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**Ultrafast nonlinear optical processes: Cross-phase-modulation
and stimulated hyper-Raman scattering**

Wang, Quan-Zhen, Ph.D.

City University of New York, 1993

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**ULTRAFAST NONLINEAR OPTICAL PROCESSES:
CROSS-PHASE-MODULATION
AND
STIMULATED HYPER-RAMAN SCATTERING**

by

QUAN-ZHEN WANG

A dissertation submitted to the Graduate Faculty in Physics in partial fulfillment of the requirement for the degree of Doctor of Philosophy, The City University of New York.

• 1993 •

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Abstracts

ULTRAFAST NONLINEAR OPTICAL PROCESSES: CROSS-PHASE-MODULATION AND STIMULATED HYPER-RAMAN SCATTERING

by

QUAN-ZHEN WANG

Advisors: Professor R. R. Alfano and Professor P. P. Ho

Nonlinear optics (NLO) is an important field of science and engineering. NLO can be used to generate, transmit and control the spectrum of laser pulses in all states of matter: solids, liquids, and gases. The objective of this thesis is to study some of the high order NLO processes. These NLO processes are cross-phase-modulation (XPM) and stimulated hyper-Raman scattering (SHRS). XPM is an intrinsic NLO process which occurs in many nonlinear optical processes. XPM is a powerful method to control the spectral, temporal, and spatial properties of ultrashort optical pulses. SHRS is a fifth order nonlinear optical process which is important for additional spectral and temporal control of ultrashort optical pulses.

This thesis is divided in eight chapters, The introduction to this thesis is presented in Chapter One. The second chapter reviews the theory of nonlinear optical susceptibilities. Some of the basic properties of the second- and third-order nonlinearities are reviewed and discussed. In the third chapter, the basic nonlinear wave equations are derived from Maxwell equations. These wave equations can be used to present a very good quantitative description of the cross-phase-modulation effects on light pulse propagation.

The research achievements of my thesis work are described in the fourth through seventh chapters.

In the fourth chapter of this thesis, the theory and experimental observation of second-harmonic cross-phase-modulation effect is described. Experimental investigation on the spectral broadening and the temporal behavior of the second-harmonic cross-phase-modulation effect in ZnSe crystal are presented.

In the fifth part of this thesis, the theoretical analysis of degenerate-cross-phase-modulation in multimode optical fibers is presented. Experimental measurements of the spectral broadening and distribution of picosecond laser pulses propagating through short multimode glass optical fibers are presented. Comparison of the measurements in multimode optical fibers with those in single mode optical fibers is also given. The study is important for the understanding the propagation of ultrashort pulses in large core multimode optical fibers.

The sixth chapter of the thesis presents our theoretical research that predicts the effects of degenerate-cross-phase-modulation on the spectral and temporal profiles of the pump and probe pulses for both single- and double pump pulses. Numerical solutions for the spectral and the temporal distributions of the pump and probe pulses are calculated. The influence of input intensity and initial time delay between the pulses are discussed. The effect of degenerate four-wave mixing on pulse amplification and compression are described. The experimental measurements of the predicted pulse compression and amplification effects in optical fibers are given in this chapter.

In the seventh chapter of the thesis, theoretical study and experimental observation of stimulated hyper-Raman scattering in an organic crystal are presented. A parametric instability theory is applied to compute the intensity dependence of SHRS signal.

Finally, in the eighth chapter of the thesis, I present insights to the future applications of cross-phase-modulation for ultrafast communication and computation systems at ultrahigh rates.

Dedicated to
my parents
and
my wife, Li-Wei Wang

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Chapter 1

Introduction

In this chapter, the basic concepts of nonlinear optics will be introduced. Self-phase-modulation, cross-phase-modulation, and stimulated hyper-Raman scattering will be briefly reviewed. The thesis statement and the organization of this thesis will be presented.

1.1 Introduction

The material response to an applied optical field at high intensity is often nonlinear.¹ Some of the nonlinear optical phenomena, such as optical pumping,² were observed before the invention of the laser. However, ordinary light can induce detectable nonlinear effects only in the cases when resonant enhancement exists.² The advent of high power laser pulses opened the door for the nonlinear optics field.³ Fascinating effects have been discovered since that time.²⁻¹⁵ Many discoveries have not only greatly enhanced our knowledge about the interaction of light with matter, but also created a revolutionary change in optics and opened the door for photonics.

The physical origin of the nonlinear optical phenomena lies in the fact that the strong field of a laser pulse is capable of affecting the atomic and molecular systems of materials. The medium then reacts back and interacts with the field of the laser pulse to produce various nonlinear effects. The nonlinear optical response can be described by expressing the polarization $P(t)$ as a power series in the field strength $E(t)$ as

$$P(t) = \chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \chi^{(4)}E^4(t) + \chi^{(5)}E^5(t) + \dots, \quad (1.1)$$

where $\chi^{(1)}$ is known as the linear susceptibility, the quantities $\chi^{(2)}$, $\chi^{(3)}$, $\chi^{(4)}$, and $\chi^{(5)}$ are known as the second-, third-, fourth-, and fifth-order nonlinear susceptibilities, respectively. For simplicity, the polarization $P(t)$ and the field $E(t)$ have taken to be scalar quantities in writing Eq.(1.1). In general, the polarization and the field are vectors, $\chi^{(1)}$ is a second rank tensor, and $\chi^{(2)}$ a third rank tensor, etc.

The lowest order nonlinearity is the second order nonlinearity and is responsible for second harmonic generation, sum- and difference-frequency generation, rectification, two photon absorption, optical parametric oscillation and amplification, and Pockels electro-optical effects. The third order harmonic generation, phase conjugation, two-photon absorption, Raman scattering, self-focusing, Kerr effect, self-phase-modulation (SPM), and cross-phase-modulation (XPM) are the typical third order nonlinear processes. Fifth order harmonic generation, six-wave mixing and stimulated hyper-Raman scattering (SHRS) are fifth order nonlinear processes.

This thesis work concentrates on the studies of nonlinear processes associated with XPM and SHRS.

1.2 Cross-Phase-Modulation

Two important nonlinear phenomena in the propagation of ultrashort optical pulses in matter are self-phase-modulation (SPM) and cross-phase-modulation (XPM).

SPM refers to the phenomenon in which a laser beam propagating in a medium interacts with the medium and imposes a phase modulation on itself. It is the main mechanism responsible for the generation of picosecond and femtosecond supercontinuum light.¹⁶⁻²⁰

XPM is similar to self-phase-modulation, but corresponds to the phase modulation caused by the nonlinear refractive index induced by another intense optical pulse.^{19, 21-}

²³ It has an important potential for applications based on picosecond and femtosecond technology. XPM is intrinsic to the generation of stimulated Raman scattering (SRS), second harmonic generation (SHG), and stimulated four photon mixing (SFPM). When the phase of the stimulated Raman scattering pulse is modulated by the index change caused by the primary pumping laser pulse, the process is called stimulated Raman scattering cross-phase-modulation (SRS-XPM). When the phase of the second harmonic generation pulse is modulated by the index change caused by the pumping laser pulse, the process is called second harmonic generation cross-phase-modulation (SHG-XPM). If the phase modulations are produced by two or more laser pulses with the same frequencies interacting in condensed matter, the process is called degenerate cross-phase-modulation (DXPM).²⁴⁻²⁶

1.3 Stimulated Hyper-Raman Scattering

Stimulated Raman scattering (SRS) is one of the few nonlinear optical effects discovered in the early 1960s. Over the years, many useful applications have been developed out of this process. SRS is a two-photon process in which one photon at ω_L is absorbed and one photon at $\omega_L - \omega_p$ is emitted, while a phonon at ω_p is created. From the wave interaction point of view, a two-photon transition is a third-order process. Therefore, SRS can be described as a third-order wave coupling process between the pump and Stokes waves. Stimulated hyper-Raman scattering (SHRS) is a three-photon analogue of SRS. In SHRS, two photons of the pumping laser beam at frequency ω_L are absorbed and a photon at $2\omega_L - \omega_p$ and a phonon at ω_p are created. SHRS is a fifth order nonlinear optical process described by the nonlinear susceptibility $\chi^{(5)}$ as compared to SRS which is described by the third-order nonlinear susceptibility $\chi^{(3)}$. SHRS has been observed in connection with two-photon excitation of alkali-metal and alkali-earth-

metal atoms²⁶⁻²⁸ and in molecules.²⁹ Recently, SHRS has been observed in a thin organic crystal.³⁰

1.4 Thesis Statement

This thesis will focus on the theoretical and experimental studies of XPM of ultrashort optical pulses propagating in condensed matter and the SHRS in organic crystals.

The spectral and temporal properties of XPM are essential for the understanding and applications of XPM process. First, the second-harmonic generation XPM (SHG-XPM) is studied.

Theoretical analysis of SHG-XPM is given. The spectral and temporal properties of SHG-XPM of picosecond pulses in ZnSe crystals are investigated by using a spectrometer combined with an optical multichannel analyzer and a picosecond streak camera. Then the degenerate-XPM (DXPM) is investigated. The spectral broadening of picosecond pulses in multimode optical fibers due to the DXPM between different modes is studied. Pulse compression using DXPM is investigated both theoretically and experimentally. The influence of the degenerate-four-wave-mixing (DFWM) on pulse compression using DXPM is studied. The SHRS in an organic crystal (-) 2-(α -methylbenzylamino)-5-nitropyridine (MBA-NP) is also measured and compared with the theoretical analysis.

1.5 Thesis Organization

This thesis is organized into eight chapters.

In Chapter 2, brief discussions on the nonlinear susceptibilities from both the classical oscillator model and the quantum mechanical model are presented.

In Chapter 3, the coupled electromagnetic waves equations which govern the nonlinear effects in media are derived.

In Chapter 4, theoretical analysis on the SHG-XPM is presented. The SHG-XPM effect is experimentally studied with ZnSe crystal which is a very suitable material to study the SHG-XPM since it possesses both large $\chi^{(2)}$ and $\chi^{(3)}$. Both the spectral profiles and temporal profiles of the SHG-XPM pulses are measured and theoretically modeled.

In Chapter 5, the XPM effects in multimode optical fibers are investigated theoretically by applying the wave equations obtained in Chapter 3. The experimental results of the spectral broadening caused by XPM in multimode optical fibers are presented. Theoretical fitting of the experimental results are provided. This investigation will allow us to study the propagation of ultrashort laser pulses in multimode optical fibers.

In Chapter 6, the theory of DXPM is given. The results of numerical calculations of DXPM are presented. The analysis indicated that DXPM can be used to compress optical pulses in condensed matter in both normal and anomalous dispersion regimes without the use of prisms and gratings. Experimental results of pulse compression are presented.

In Chapter 7, the fifth order nonlinear effect, stimulated hyper-Raman scattering (SHRS) in a thin organic crystal is reported. A parametric theory for SHRS is developed to explain the intensity dependence of the hyper-Raman signal.

In Chapter 8, insight into the future research on pulse compression and amplification using XPM are given.

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Chapter 2

Nonlinear Optical Susceptibilities

The nonlinear optical susceptibilities characterize the nonlinear optical response in a medium and depend on the detailed electronic and molecular structure of the medium. We introduce the nonlinear polarization and nonlinear optical susceptibilities in Section 2.1. In Section 2.2 simple anharmonic oscillator models are considered to illustrate the origin of the optical nonlinear susceptibilities. Section 2.3 gives a brief discussion of nonlinear susceptibilities starting from the microscopic theory.

2.1 Maxwell Equations in Nonlinear Media

An optical pulse propagating in a nonlinear medium is governed by the Maxwell's equations for the electric and magnetic fields $\mathbf{E}(\mathbf{r},t)$ and $\mathbf{B}(\mathbf{r},t)$:¹⁻³

$$\begin{aligned}\nabla \times \mathbf{E} &= -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}, \\ \nabla \times \mathbf{H} &= \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t} + \frac{4\pi}{c} \mathbf{J}, \\ \nabla \cdot \mathbf{D} &= 4\pi\rho, \\ \nabla \cdot \mathbf{B} &= 0,\end{aligned}\tag{2.1}$$

where $\mathbf{J}(\mathbf{r},t)$ and $\rho(\mathbf{r},t)$ are the free current and charge densities, respectively. In nonlinear optics, we are often interested in the solution of Maxwell's equations in regions of space that contain no free charges and currents, so that $\mathbf{J}(\mathbf{r},t)=0$ and $\rho(\mathbf{r},t)=0$. For nonmagnetic materials, we also have $\mathbf{B}=\mathbf{H}$.

The electric displacement $\mathbf{D}(\mathbf{r},t)$ is given by the constitutive equations^{1,4}

$$\mathbf{D} = \epsilon \mathbf{E} = \mathbf{E} + 4\pi \mathbf{P}, \quad (2.2)$$

where the constitutive parameter ϵ is known as permittivity tensor, $\mathbf{P}$ is the electric polarization. From the first two equations in Eqs (2.1), the wave equation that governs optical wave propagation in the medium can be obtained:

$$\left(\nabla \times (\nabla \times) + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \mathbf{E}(\mathbf{r}, t) = - \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}(\mathbf{r}, t), \quad (2.3)$$

where $\mathbf{P}$ is a function of spatial coordinates and time, and is the only time-varying source term.

When the electric and magnetic fields are not strong, the quantity ϵ can be assumed independent of the fields. That is the case of linear optics.

In the linear case, $\mathbf{P}$ takes a simple linearized form

$$\mathbf{P}(\mathbf{r}, t) = \int_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r}-\mathbf{r}', t-t') \cdot \mathbf{E}(\mathbf{r}', t') d\mathbf{r}' dt' \quad (2.4)$$

where $\chi^{(1)}$ is the linear susceptibility. If $\mathbf{E}$ is a monochromatic plane wave with

$$\mathbf{E}(\mathbf{r}, t) = \mathbf{E}(\mathbf{k}, \omega) = \mathbf{A}(\mathbf{k}, \omega) \exp(i\mathbf{k} \cdot \mathbf{r} - i\omega t), \quad (2.5)$$

then Fourier transform of (2.3) yields the relation

$$\mathbf{P}(\mathbf{k}, \omega) = \chi^{(1)}(\mathbf{k}, \omega) \cdot \mathbf{E}(\mathbf{k}, \omega)$$

with

$$\chi^{(1)}(\mathbf{k}, \omega) = \int_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r}, t) \exp(-i\mathbf{k} \cdot \mathbf{r} - i\omega t) d\mathbf{r} dt. \quad (2.6)$$

The linear dielectric constant $\epsilon(\mathbf{k}, \omega)$ is related to $\chi^{(1)}$ by

$$\epsilon(\mathbf{k}, \omega) = 1 + 4\pi \chi^{(1)}(\mathbf{k}, \omega). \quad (2.7)$$

In the electric dipole approximation, $\chi^{(1)}(\mathbf{r}, t)$ is independent of $\mathbf{r}$, and hence both $\chi^{(1)}(\mathbf{k}, \omega)$ and $\epsilon(\mathbf{k}, \omega)$ are independent of $\mathbf{k}$. Unless specified otherwise, we use the electric dipole approximation throughout this thesis.

In the nonlinear case, the electric polarization $\mathbf{P}$ can be expanded into a power series of $\mathbf{E}$:⁵

$$\begin{aligned}
\mathbf{P}(\mathbf{r}, t) = & \int_{-\infty}^{\infty} \chi^{(1)}(t-t') \cdot \mathbf{E}(\mathbf{r}, t') dt' \\
& + \int_{-\infty}^{\infty} \chi^{(2)}(t-t_1; t-t_2) \cdot \mathbf{E}(\mathbf{r}, t_1) \mathbf{E}(\mathbf{r}, t_2) dt_1 dt_2 \\
& + \int_{-\infty}^{\infty} \chi^{(3)}(t-t_1; t-t_2; t-t_3) \\
& \quad : \mathbf{E}(\mathbf{r}, t_1) \mathbf{E}(\mathbf{r}, t_2) \mathbf{E}(\mathbf{r}, t_3) dt_1 dt_2 dt_3 + \dots
\end{aligned} \tag{2.8}$$

where $\chi^{(n)}$ is the n th-order nonlinear susceptibility. If $\mathbf{E}$ can be expressed as

$$\mathbf{E}(\mathbf{r}, t) = \int_{-\infty}^{\infty} \mathbf{E}(\mathbf{r}, \omega) d\omega \quad \text{with } \mathbf{E}(\mathbf{r}, \omega) \propto \exp(-i\omega t), \tag{2.9}$$

then, the induced polarization in a medium can be expressed as

$$\mathbf{P}(\mathbf{r}, t) = \mathbf{P}^{(1)}(\mathbf{r}, t) + \mathbf{P}^{(2)}(\mathbf{r}, t) + \mathbf{P}^{(3)}(\mathbf{r}, t) \tag{2.10}$$

where

$$\begin{aligned}
\mathbf{P}^{(1)}(\mathbf{r}, t) &= \int_{-\infty}^{\infty} \chi^{(1)}(t-t') \cdot \mathbf{E}(\mathbf{r}, t') dt', \tag{2.11} \\
\mathbf{P}^{(n)}(\mathbf{r}, t) &= \int_{-\infty}^{\infty} \chi^{(n)}(t-t_1; t-t_2; \dots; t-t_n) \cdot \mathbf{E}(\mathbf{r}, t_1) \mathbf{E}(\mathbf{r}, t_2) \dots \mathbf{E}(\mathbf{r}, t_n) dt_1 dt_2 \dots dt_n \\
&= \int_{-\infty}^{\infty} \chi^{(n)}(\omega = \omega_1 + \omega_2 + \dots + \omega_n) : \mathbf{E}(\omega_1) \mathbf{E}(\omega_2) \dots \mathbf{E}(\omega_n) d\omega_1 d\omega_2 \dots d\omega_n,
\end{aligned} \tag{2.12}$$

and the n th-order susceptibility is

$$\begin{aligned}
\chi^{(n)}(t-t_1; t-t_2; \dots; t-t_n) &= \int_{-\infty}^{\infty} \chi^{(n)}(t-t_1; t-t_2; \dots; t-t_n) \exp[i\omega_1(t-t_1) + \\
&\quad + i\omega_2(t-t_2) + \dots + i\omega_n(t-t_n)] d\omega_1 d\omega_2 \dots d\omega_n.
\end{aligned} \tag{2.13}$$

We note that, strictly speaking, only for a set of applied monochromatic fields can we write

$$\mathbf{P}^{(n)}(\mathbf{r}, \omega) = \chi^{(n)}(\omega = \omega_1 + \omega_2 + \dots + \omega_n) : E(\omega_1) E(\omega_2) \dots E(\omega_n). \quad (2.14)$$

In the case of instantaneous response (corresponding to a dispersionless medium),⁶ we have

$$\mathbf{P}^{(n)}(\mathbf{r}, t) = \chi^{(n)} \cdot [\mathbf{E}(\mathbf{r}, t)]^n. \quad (2.15)$$

The linear and nonlinear susceptibilities characterize the optical properties of a medium. If $\chi^{(n)}$ is known for a given medium, the n -th order nonlinear optical effects in this medium can be predicted from the Maxwell's equations. Physically, $\chi^{(n)}$ is related to the microscopic structure of the medium and can be properly evaluated with a full quantum mechanical calculations. In the following sections, the origin of the nonlinear optical susceptibilities are discussed.

2.2 Physical Origin of Nonlinear Polarization

--- Classical Oscillator Model

The optical polarization of dielectric media is due mostly to the outer, loosely bound valence electrons that are displaced by the optical field. Denoting the electron deviation from the equilibrium position by x and the density of electrons by N , the polarization P is given by

$$P(t) = -Nex(t). \quad (2.16)$$

2.2.1 Noncentrosymmetric Media

If the deviation x is sufficiently small, the equation of motion of the electron coordinate in the presence of a driving force F for the case of noncentrosymmetric media is^{4,5,7}

$$\frac{d^2x}{dt^2} + \Gamma \frac{dx}{dt} + \omega_0^2 x + \alpha x^2 = F, \quad (2.17)$$

where αx^2 is nonlinear term and α is a parameter that characterizes the nonlinearity of the response. We consider here the response of the oscillator to an applied field with Fourier components at frequency $\pm\omega_1$ and $\pm\omega_2$. Then the driving force can be written as

$$\begin{aligned} F &= -\frac{e}{m} E(t) \\ &= -\frac{e}{m} [E_1(e^{-i\omega_1 t} + e^{i\omega_1 t}) + E_2(e^{-i\omega_2 t} + e^{i\omega_2 t})]. \end{aligned} \quad (2.18)$$

We use a procedure analogous to that of the Rayleigh-Schrodinger perturbation theory. We replace $E(t)$ in Eqs. (2.17) and (2.18) by $\lambda E(t)$, where λ is a perturbation parameter which will be set equal to one at the end of the calculation. We now seek a solution in the form of a power series expansion in the strength λ of the perturbation:

$$x = \lambda x^{(1)} + \lambda^2 x^{(2)} + \lambda^3 x^{(3)} + \dots \quad (2.19)$$

Substituting Equation (2.19) into Equation (2.17) one gets a equation contains λ^1 , λ^2 , and λ^3 . If this equation is to be valid for all times t , the coefficients of λ^1 , λ^2 , and λ^3 on both sides of the equation must be equal.

Equating the coefficients of λ^1 gives

$$x^{(1)}(\omega_j) = \frac{-(e/m)E_j}{(\omega_0^2 - \omega_j^2 - i\omega_j\Gamma)} e^{-i\omega_j t} \quad (2.20)$$

where $j=1$ and 2 , respectively.

Equating the coefficients of λ^2 and using the expression of $x^{(1)}(\omega_j)$ leads to the solutions for the lowest-order correction $x^{(2)}$:

$$\begin{aligned} x^{(2)}(\omega_1 \pm \omega_2) &= \frac{-2\alpha(e/m)^2 E_1 E_2}{(\omega_0^2 - \omega_1^2 - i\omega_1\Gamma)(\omega_0^2 - \omega_2^2 \mp i\omega_2\Gamma)} \\ &\quad \times \frac{1}{\omega_0^2 - (\omega_1 \pm \omega_2)^2 - i(\omega_1 \pm \omega_2)\Gamma} e^{-i(\omega_1 \pm \omega_2)t}, \end{aligned} \quad (2.21)$$

$$x^{(2)}(2\omega_j) = \frac{-\alpha(q/m)^2 E_j^2}{(\omega_0^2 - \omega_j^2 - i\omega_j\Gamma)^2 (\omega_0^2 - 4\omega_j^2 - i2\omega_j\Gamma)} e^{-i2\omega_j t} , \quad (2.22)$$

$$x^{(2)}(0) = -2\alpha\left(\frac{e}{m}\right)^2 \frac{1}{\omega_0^2} \left(\frac{E_1 E_1^*}{(\omega_0^2 - \omega_1^2)^2 + (\omega_1\Gamma)^2} + \frac{E_2 E_2^*}{(\omega_0^2 - \omega_2^2)^2 + (\omega_2\Gamma)^2} \right) \quad (2.23)$$

The appearance of zero optical frequency polarization component is known as optical rectification.

By successive iteration, higher order solutions can also be obtained. As seen from the second order solutions, new frequency components of the polarization at $\omega_1 \pm \omega_2$, $2\omega_1$, $2\omega_2$, and 0 have appeared through quadratic interaction of the field with the oscillator via the anharmonic term. The oscillating polarization components will radiate and generate new electromagnetic waves at $\omega_1 \pm \omega_2$, $2\omega_1$, and $2\omega_2$. Thus, lowest order nonlinear effects, sum- and difference-frequency generation and second harmonic generation are obtained.

With the solutions of $x(t)$, it can be readily shown that the linear and nonlinear susceptibilities are

$$\chi^{(1)}(\omega_j) = \frac{N(e^2/m)}{(\omega_0^2 - \omega_j^2 - i\omega_j\Gamma)}, \quad (2.24)$$

$$\begin{aligned} \chi^{(2)}(\omega_1 \pm \omega_2) &= \frac{N\alpha(e^3/m^2)}{(\omega_0^2 - \omega_1^2 - i\omega_1\Gamma)(\omega_0^2 - \omega_2^2 \mp i\omega_2\Gamma)} \\ &\quad \times \frac{1}{\omega_0^2 - (\omega_1 \pm \omega_2)^2 - i(\omega_1 \pm \omega_2)\Gamma} , \end{aligned} \quad (2.25)$$

$$\chi^{(2)}(2\omega_j) = \frac{N\alpha(e^3/m^2)}{(\omega_0^2 - \omega_j^2 - i\omega_j\Gamma)^2 (\omega_0^2 - 4\omega_j^2 - i2\omega_j\Gamma)} , \quad (2.26)$$

and for optical rectification

$$\chi^{(2)}(0=\omega_j-\omega_j) = N\alpha\left(\frac{e^3}{m^2}\right) \frac{1}{\omega_0^2[(\omega_0^2 - \omega_j^2) + (\omega_j\Gamma)^2]} \quad (2.27)$$

respectively.

As we can see from above, the second order nonlinear susceptibility is the lowest-order nonlinear susceptibility of a noncentrosymmetric material. The higher-order nonlinear susceptibilities can be obtained by extending the analysis. For example, the third-order nonlinear susceptibility $\chi^{(3)}$ can be obtained by equating the coefficient of λ^3 .

2.2.2 Centrosymmetric Media

For the case of a centrosymmetric medium, the equation of motion for the electron displacement from equilibrium can be written as⁷

$$\frac{d^2\mathbf{r}}{dt^2} + \Gamma\frac{d\mathbf{r}}{dt} + \omega_0^2\mathbf{r} - \beta\mathbf{r}(\mathbf{r}\cdot\mathbf{r}) = \mathbf{F}, \quad (2.28)$$

where $-\beta\mathbf{r}(\mathbf{r}\cdot\mathbf{r})$ is the nonlinear term. To simplify the problem, we express the applied field as

$$\mathbf{E}(t) = \sum_n \mathbf{E}(\omega_n)e^{-i\omega_n t}. \quad (2.29)$$

The method of solution is analogous to that used above for noncentrosymmetric medium.

We replace $\mathbf{E}(t)$ by $\lambda\mathbf{E}(t)$ and let the $\mathbf{r}(t)$ have a form of

$$\mathbf{r}(t) = \lambda\mathbf{r}^{(1)} + \lambda^2\mathbf{r}^{(2)} + \lambda^3\mathbf{r}^{(3)} + \dots, \quad (2.30)$$

With the same technique used above, one may find

$$\mathbf{r}^{(1)}(t) = \sum_n \mathbf{r}^{(1)}(\omega_n)e^{-i\omega_n t} \quad (2.31)$$

where

$$\mathbf{r}^{(1)}(\omega_n) = \frac{-e\mathbf{E}^{(1)}(\omega_n)/m}{(\omega_0^2 - \omega_n^2 - i\omega_n\Gamma)}. \quad (2.32)$$

Hence, the linear susceptibility is

$$\chi_{ij}^{(1)}(\omega_n) = \frac{Ne^2/m}{(\omega_0^2 - \omega_j^2 - i\omega_j\Gamma)} \delta_{ij} , \quad (2.33)$$

where δ_{ij} is the Kronecker delta, which is defined such that $\delta_{ij} = 1$ for $i = j$ and $\delta_{ij} = 0$ for $i \neq j$.

The steady state solution for the second-order response of the system is $\mathbf{r}^{(2)} = 0$. This indicates that *the second-order nonlinear susceptibility vanishes in centrosymmetric media*.

The solutions for the third-order response are

$$\mathbf{r}^{(3)}(t) = \sum_q \mathbf{r}^{(3)}(\omega_q) e^{-i\omega_q t} , \quad (2.34)$$

with

$$\begin{aligned} \mathbf{r}^{(3)}(\omega_q) = & -\beta \left(\frac{e}{m} \right)^3 \sum_{(mnp)} \frac{[\mathbf{E}(\omega_m) \cdot \mathbf{E}(\omega_n)] \mathbf{E}(\omega_p)}{(\omega_0^2 - \omega_q^2 - i\omega_q\Gamma)(\omega_0^2 - \omega_m^2 - i\omega_m\Gamma)} \\ & \times \frac{1}{(\omega_0^2 - \omega_n^2 - i\omega_n\Gamma)(\omega_0^2 - \omega_p^2 - i\omega_p\Gamma)} . \end{aligned} \quad (2.35)$$

The nonlinear susceptibility is

$$\begin{aligned} \chi_{ijkl}^{(3)}(\omega_q = \omega_m + \omega_n + \omega_p) = & N\beta \left(\frac{e^4}{m^3} \right) \sum_{(mnp)} \frac{\delta_{jk} \delta_{il}}{(\omega_0^2 - \omega_q^2 - i\omega_q\Gamma)(\omega_0^2 - \omega_m^2 - i\omega_m\Gamma)} \\ & \times \frac{1}{(\omega_0^2 - \omega_n^2 - i\omega_n\Gamma)(\omega_0^2 - \omega_p^2 - i\omega_p\Gamma)} . \end{aligned} \quad (2.36)$$

Eq.(2.36) is a perfectly adequate expression for the nonlinear susceptibility. It is conventional to define nonlinear susceptibilities in a manner that displays the symmetry, known as intrinsic permutation symmetry. Since there is six possible permutations of the orders in which the electric field components may be taken, we define the third-order susceptibility to be one-sixth of the sum of the six expressions analogous to Eq. (2.36) with the input fields taken in all possible orders. When we carry out this prescription, we

find that only three distinct contributions occur and that the resulting form for the nonlinear susceptibility is given by

$$\begin{aligned} \chi_{ijkl}^{(3)}(\omega_q=\omega_m+\omega_n+\omega_p) &= \frac{1}{3}N\beta\left(\frac{e^4}{m^3}\right) \\ &\times \sum_{(mnp)} \frac{\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}}{(\omega_0^2 - \omega_q^2 - i\omega_q\Gamma)(\omega_0^2 - \omega_m^2 - i\omega_m\Gamma)} \\ &\times \frac{1}{(\omega_0^2 - \omega_n^2 - i\omega_n\Gamma)(\omega_0^2 - \omega_p^2 - i\omega_p\Gamma)} . \end{aligned} \quad (2.37)$$

From the above solutions, we can see that the nonlinear response depends not only on the material, but also on the frequencies of applied fields. If the applied frequencies ω_j are near the resonance frequency, the resonant denominator may drastically increase the nonlinear susceptibility and the nonlinear effects can be much stronger. Consequently, resonant nonlinear effects can be detected with much weaker light intensity.

With resonant enhancement, the nonlinear effects might be so strong that the perturbation expansion is no longer valid. If this is the case, the full nonlinear expression of P as a function of E must be included in the calculation. The problem then falls into the domain of strong interaction of light with matter which is beyond our discussion here.

2.3 Physical Origin of Nonlinear Polarization

--- Quantum Mechanical Model

Nonlinear optical susceptibilities depend on the detailed electronic and molecular structure of the media. Quantum mechanical calculations are needed to find the microscopic expression for the nonlinear susceptibilities.⁸

A simple quantum mechanical perturbation theory of the atomic wave function can be used to make accurate predictions of the nonresonant response of atomic and molecular

systems.⁷ The effects of near-resonant excitation cannot be adequately described by this formalism. The more complicated density matrix formalism provides results that are more generally valid. In the following, we calculate the nonlinear optical susceptibilities through the use of the density matrix formulation of quantum mechanics.^{6,7,9}

The density matrix operator is defined as

$$\rho = \overline{|\psi\rangle\langle\psi|}, \quad (2.38)$$

where $|\psi\rangle$ represent the ket vector for some state of the system, $\langle\psi|$ represents the Hermitian adjoint of $|\psi\rangle$, and the overbar represents an ensemble average. The ensemble average of a polarization $\mathbf{P}$ is given by

$$\begin{aligned} \langle\mathbf{P}\rangle &= \overline{\langle\psi|\mathbf{P}|\psi\rangle} \\ &= \text{Tr}(\rho\mathbf{P}). \end{aligned} \quad (2.39)$$

From the definition of ρ in (2.38) and from the Schrödinger equation for $|\psi\rangle$, we can readily obtain the equation of motion for ρ

$$\frac{\partial\rho}{\partial t} = \frac{1}{i\hbar} [\hat{H}, \rho]. \quad (2.40)$$

The Hamiltonian $\hat{H}$ is

$$\hat{H} = \hat{H}_0 + \hat{V}(t), \quad (2.41)$$

with $\hat{H}_0$ is the Hamiltonian for a free atom and $\hat{V}(t)$ describes the interaction of the atom with the electromagnetic field. The interaction Hamiltonian can be usually written as

$$\hat{V}(t) = -\hat{\mu} \cdot \mathbf{E}(t), \quad (2.42)$$

where $\hat{\mu} = -e\hat{\mathbf{r}}$ is the electric dipole moment operator, $-e$ is the charge of the electron, $\mathbf{E}(t)$ is the electric field. There are still certain interactions (such as those resulting from collision between atoms) that can not conveniently be included in a Hamiltonian description. Those interactions can lead to a change in the state of the system, and hence to a nonvanishing value of $d\rho/dt$. We include such effects in the formalism by adding phenomenological damping terms to the equation of motion (2.40). Hence, the equation of motion becomes

$$\frac{\partial \rho}{\partial t} = \frac{1}{i\hbar} [\hat{H}, \rho] + \left(\frac{\partial \rho}{\partial t} \right)_{\text{relax}} \quad (2.43)$$

where

$$\left(\frac{\partial \rho_{mn}}{\partial t} \right)_{\text{relax}} = -\Gamma_{mn}(\rho_{mn} - \rho_{mn}^{(0)}) \quad (2.44)$$

The second term on the right-hand side of (2.43) is a phenomenological damping term, which indicates relaxation to an equilibrium value $\rho_{mn}^{(0)}$ at a rate Γ_{mn} . Since Γ_{nm} is a decay rate, we assume that $\Gamma_{mn} = \Gamma_{nm}$. In addition, we make the physical assumption that

$$\rho_{mn}^{(0)} = 0 \quad \text{for } n \neq m. \quad (2.45)$$

For most problems of physical interest, Eq. (2.43) cannot be solved analytically. To find the nonlinear polarization and nonlinear susceptibilities of various orders, we need to use a perturbation expansion in the calculation. In order to do so, we replace $\hat{V}$ by $\lambda \hat{V}$ in Eq. (2.42), and seek a solution in the form of a power series in λ

$$\rho = \rho^{(0)} + \lambda \rho^{(1)} + \lambda^2 \rho^{(2)} + \dots \quad (2.46)$$

By comparing the coefficients for each power of λ in Eq. (2.43), we obtained

$$\begin{aligned} \frac{\partial \rho^{(1)}}{\partial t} &= \frac{1}{i\hbar} ([\hat{H}_0, \rho^{(1)}] + [\hat{V}, \rho^{(0)}]) + \left(\frac{\partial \rho^{(1)}}{\partial t} \right)_{\text{relax}}, \\ \frac{\partial \rho^{(2)}}{\partial t} &= \frac{1}{i\hbar} ([\hat{H}_0, \rho^{(2)}] + [\hat{V}, \rho^{(1)}]) + \left(\frac{\partial \rho^{(2)}}{\partial t} \right)_{\text{relax}}, \\ \frac{\partial \rho^{(3)}}{\partial t} &= \frac{1}{i\hbar} ([\hat{H}_0, \rho^{(3)}] + [\hat{V}, \rho^{(2)}]) + \left(\frac{\partial \rho^{(3)}}{\partial t} \right)_{\text{relax}}, \end{aligned} \quad (2.47)$$

It can be shown that the solutions for Eq. (2.46) can be expressed as⁷

$$\rho_{mn}^{(q)}(t) = \frac{1}{i\hbar} \int_{-\infty}^t [\hat{V}, \rho^{(q-1)}]_{mn} e^{(i\omega_{mn} + \Gamma_{mn})(t'-t)} dt', \quad (2.48)$$

where $\omega_{mn} = (E_m - E_n)/\hbar$.

The optical polarization then can be written as

$$\langle \mathbf{P}^{(n)} \rangle = \text{Tr}(\rho^{(n)} \mathbf{P}). \quad (2.49)$$

We are interested in the response to a field that can be decomposed into Fourier components,

$$\mathbf{E}(t) = \sum_{\mathbf{p}} \mathbf{E}(\omega_{\mathbf{p}}) e^{-i\omega_{\mathbf{p}}t}. \quad (2.50)$$

Then, since

$$\hat{\mathbf{V}} = \sum_{\mathbf{p}} \hat{\mathbf{V}}(\omega_{\mathbf{p}}), \text{ , and } \hat{\mathbf{V}}(\omega_{\mathbf{p}}) \propto e^{-i\omega_{\mathbf{p}}t}, \quad (2.51)$$

the operator can also be expanded into a Fourier series

$$\rho^{(n)} = \sum_{\mathbf{p}} \rho^{(n)}(\omega_{\mathbf{p}}). \quad (2.52)$$

With

$$\frac{\partial \rho^{(n)}(\omega_{\mathbf{p}})}{\partial t} = -i\omega_{\mathbf{p}} \rho^{(n)}(\omega_{\mathbf{p}}), \quad (2.53)$$

Eq. (2.47) can be solved explicitly for $\rho^{(n)}(\omega_{\mathbf{p}})$ in successive orders. The first-, second- and third-order solutions are

$$\begin{aligned} \rho_{mn}^{(1)}(\omega_{\mathbf{p}}) &= \frac{(\hat{\mathbf{V}}(\omega_{\mathbf{p}}))_{mn}}{\hbar((\omega_{\mathbf{p}} - \omega_{mn}) + i\Gamma_{mn})} (\rho_{nn}^{(0)} - \rho_{mm}^{(0)}) \\ \rho_{mn}^{(2)}(\omega_{\mathbf{p}} + \omega_{\mathbf{q}}) &= \frac{[\hat{\mathbf{V}}(\omega_{\mathbf{p}}); \rho_{mn}^{(1)}(\omega_{\mathbf{q}})]_{mn} + [\hat{\mathbf{V}}(\omega_{\mathbf{q}}); \rho_{mn}^{(1)}(\omega_{\mathbf{p}})]_{mn}}{\hbar((\omega_{\mathbf{p}} + \omega_{\mathbf{q}} - \omega_{mn}) + i\Gamma_{mn})} \\ \rho_{mn}^{(3)}(\omega_{\mathbf{p}} + \omega_{\mathbf{q}} + \omega_{\mathbf{r}}) &= \frac{[\hat{\mathbf{V}}(\omega_{\mathbf{p}}); \rho_{mn}^{(2)}(\omega_{\mathbf{q}})]_{mn} + [\hat{\mathbf{V}}(\omega_{\mathbf{q}}); \rho_{mn}^{(2)}(\omega_{\mathbf{p}})]_{mn}}{\hbar((\omega_{\mathbf{p}} + \omega_{\mathbf{q}} + \omega_{\mathbf{r}} - \omega_{mn}) + i\Gamma_{mn})} \end{aligned} \quad (2.54)$$

Higher order solutions can be obtained readily, although the derivation is long and tedious.

With the solutions for $\rho^{(n)}$, the linear and nonlinear susceptibilities can be found to be

$$\chi_{ij}^{(1)}(\omega_{\mathbf{p}}) = \frac{e^2 N}{\hbar} \sum_{mn} \rho_{nn}^{(0)} \left(\frac{(r_i)_{nm}(r_j)_{mn}}{(\omega_{mn} - \omega_{\mathbf{p}}) - i\Gamma_{mn}} + \frac{(r_i)_{mn}(r_j)_{nm}}{(\omega_{mn} + \omega_{\mathbf{p}}) + i\Gamma_{mn}} \right) \quad (2.55)$$

where $\omega_{mn} = -\omega_{nm}$ and $\Gamma_{mn} = \Gamma_{nm}$ have been used. If we assume all the population in one level (typically the ground state) denoted as level g ($\rho_{gg}^{(0)} = 1$ and $\rho_{nn}^{(0)} = 0$ for $n \neq g$),

we have

$$\chi_{ij}^{(1)}(\omega_{\mathbf{p}}) = \frac{e^2 N}{\hbar} \sum_m \left(\frac{(r_i)_{gm}(r_j)_{mg}}{(\omega_{mg} - \omega_{\mathbf{p}}) - i\Gamma_{mg}} + \frac{(r_i)_{mg}(r_j)_{gm}}{(\omega_{mg} + \omega_{\mathbf{p}}) + i\Gamma_{mg}} \right) \quad (2.56)$$

The second order nonlinear susceptibility tensor can be obtained as

$$\begin{aligned}
\chi_{ijk}^{(2)}(\omega=\omega_p+\omega_q, \omega_p, \omega_q) = & -\frac{e^3 N}{2\hbar^2} \\
& \times \sum_{mnl} \left\{ (\rho_{mm}^{(0)} - \rho_{ll}^{(0)}) \left(\frac{(r_i)_{mn}(r_j)_{nl}(r_k)_{lm}}{[(\omega_{nm} - \omega_p - \omega_q) - i\Gamma_{nm}][(\omega_{lm} - \omega_p) - i\Gamma_{lm}]} \right) \right. \\
& + \left(\frac{(r_i)_{mn}(r_j)_{ml}(r_k)_{lm}}{[(\omega_{nm} - \omega_p - \omega_q) - i\Gamma_{nm}][(\omega_{lm} - \omega_q) - i\Gamma_{lm}]} \right) \\
& - (\rho_{ll}^{(0)} - \rho_{nn}^{(0)}) \left(\frac{(r_i)_{mn}(r_j)_{lm}(r_k)_{nl}}{[(\omega_{nm} - \omega_p - \omega_q) - i\Gamma_{nm}][(\omega_{nl} - \omega_p) - i\Gamma_{nl}]} \right) \\
& \left. + \left(\frac{(r_i)_{mn}(r_j)_{lm}(r_k)_{nl}}{[(\omega_{nm} - \omega_p - \omega_q) - i\Gamma_{nm}][(\omega_{nl} - \omega_q) - i\Gamma_{nl}]} \right) \right\},
\end{aligned} \tag{2.57}$$

The third order nonlinear susceptibility is given by

$$\begin{aligned}
\chi_{kji h}^{(3)}(\omega=\omega_p+\omega_q+\omega_r, \omega_p, \omega_q, \omega_r) = & \frac{e^4 N}{\hbar^3} P \sum_{vnml} \rho_{ll}^{(0)} \\
& \times \left\{ \frac{(r_k)_{lv}(r_j)_{vn}(r_i)_{nm}(r_h)_{ml}}{[(\omega_{vl} - \omega_p - \omega_q - \omega_r) - i\Gamma_{vl}][(\omega_{nl} - \omega_p - \omega_q) - i\Gamma_{nl}][(\omega_{ml} - \omega_p) - i\Gamma_{ml}]} \right. \\
& + \frac{(r_h)_{lv}(r_k)_{vn}(r_j)_{nm}(r_i)_{ml}}{[(\omega_{nv} - \omega_p - \omega_q - \omega_r) - i\Gamma_{nv}][(\omega_{mv} - \omega_p - \omega_q) - i\Gamma_{mv}][(\omega_{vl} + \omega_p) + i\Gamma_{vl}]} \\
& + \frac{(r_i)_{lv}(r_k)_{vn}(r_j)_{nm}(r_h)_{ml}}{[(\omega_{nv} - \omega_p - \omega_q - \omega_r) - i\Gamma_{nv}][(\omega_{vm} + \omega_p + \omega_q) + i\Gamma_{vm}][(\omega_{ml} - \omega_p) - i\Gamma_{ml}]} \\
& + \frac{(r_h)_{lv}(r_i)_{vn}(r_k)_{nm}(r_j)_{ml}}{[(\omega_{mv} - \omega_p - \omega_q - \omega_r) - i\Gamma_{mv}][(\omega_{nl} + \omega_p + \omega_q) + i\Gamma_{nl}][(\omega_{vl} + \omega_p) + i\Gamma_{vl}]} \\
& + \frac{(r_j)_{lv}(r_k)_{vn}(r_i)_{nm}(r_h)_{ml}}{[(\omega_{vn} + \omega_p + \omega_q + \omega_r) + i\Gamma_{vn}][(\omega_{nl} - \omega_p - \omega_q) - i\Gamma_{nl}][(\omega_{ml} - \omega_p) - i\Gamma_{ml}]} \\
& + \frac{(r_h)_{lv}(r_j)_{vn}(r_k)_{nm}(r_i)_{ml}}{[(\omega_{nm} + \omega_p + \omega_q + \omega_r) + i\Gamma_{nm}][(\omega_{mv} - \omega_p - \omega_q) - i\Gamma_{mv}][(\omega_{vl} + \omega_p) + i\Gamma_{vl}]} \\
& + \frac{(r_i)_{lv}(r_j)_{vn}(r_k)_{nm}(r_h)_{ml}}{[(\omega_{nm} + \omega_p + \omega_q + \omega_r) + i\Gamma_{nm}][(\omega_{vm} + \omega_p + \omega_q) + i\Gamma_{vm}][(\omega_{ml} - \omega_p) - i\Gamma_{ml}]} \\
& + \frac{(r_h)_{lv}(r_i)_{vn}(r_j)_{nm}(r_k)_{ml}}{[(\omega_{ml} + \omega_p + \omega_q + \omega_r) + i\Gamma_{ml}][(\omega_{nl} + \omega_p + \omega_q) + i\Gamma_{nl}][(\omega_{vl} + \omega_p) + i\Gamma_{vl}]} \\
& \left. \right\},
\end{aligned} \tag{2.58}$$

where P is the intrinsic permutation operator, which means that everything to the right of it is to be averaged over all possible permutations of the input frequencies ω_p , ω_q , and ω_r , with the Cartesian indices h, i, j permuted simultaneously. For the general case in which

ω_p , ω_q , and ω_r are distinct, six permutations of the field frequencies occur. The expression for $\chi^{(3)}$ consists of 48 different terms once the permutation operator is expanded.

Perturbation calculations can be facilitated with the help of diagrams. Double-sided Feynman diagram^{5,10} can be used to provide a simple picture to the corresponding physical process as well as allowing one to write down immediately the corresponding mathematical expressions for $\chi^{(n)}$.

From the above expressions, we can see that nonlinear susceptibilities arise from the interaction of the applied radiation field with electric dipole moment of the atomic and molecular systems of media. If the frequencies of the applied fields near the resonance frequencies of the atomic and molecular system of the media, the resonant denominator may drastically increases the nonlinear susceptibilities.

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Chapter 3

Nonlinear Wave Equations

In the pervious chapter, we have seen that the nonlinear response of a material system to an intense laser field can cause the polarization of the medium to develop new frequency components. Propagation of an intense laser pulse in a medium leads to various nonlinear optical phenomena. SPM and XPM are two important nonlinear phenomena. The wave equation description can give very good quantitative descriptions of SPM and XPM effects. In this chapter, we derive the nonlinear wave equations from Maxwell equations in Sections 3. and 3.2. In Section 3.3, we use the derived wave equations to describe the SPM.

These equations will be frequently used in later chapters.

3.1 Wave Equations in A Nonlinear Medium

From the Maxwell equations (2.1), we can find the wave equations that governs optical wave propagation in a medium:

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r},t) + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{D}(\mathbf{r},t) = 0 . \quad (3.1)$$

Since the electric displacement vector is

$$\mathbf{D} = \mathbf{E} + 4\pi\mathbf{P}, \quad (3.2)$$

we can obtain

$$\nabla \times \nabla \times \mathbf{E} + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E} = - \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P} . \quad (3.3)$$

This is the most general form of the wave equation in nonlinear optics. Under certain conditions it can be simplified. For example, by using an identity from vector calculus, we can write the first term on the left-hand side of Eq. (3.3) as

$$\nabla \times \nabla \times \mathbf{E} = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}. \quad (3.4)$$

In the linear optics of isotropic source-free media, the first term on the right-hand side of this identity vanishes because the Maxwell equation $\nabla \cdot \mathbf{D} = 0$ implies that $\nabla \cdot \mathbf{E} = 0$. However, in nonlinear optics this term in general is nonvanishing even for isotropic materials, since the more general relation between $\mathbf{D}$ and $\mathbf{E}$ is much more complicated than a linear proportion. Fortunately, in nonlinear optics the first term on the right-hand side of Eq. (3.2) can usually be dropped. For example, if $\mathbf{E}$ is of the form of a transverse, infinite plane wave, $\nabla \cdot \mathbf{E}$ vanishes identically. More generally, the first term can often be shown to be small, even when it does not vanish, especially when the slowly-varying amplitude approximation is valid.

It is convenient to split $\mathbf{P}$ in to its linear and nonlinear parts as

$$\mathbf{P} = \mathbf{P}_L + \mathbf{P}_{NL}, \quad (3.5)$$

where $\mathbf{P}_L = \mathbf{P}^{(1)}$ is proportional to the electric field $\mathbf{E}$. Hence Eq. (3.2) can be written as

$$\begin{aligned} \mathbf{D} &= \mathbf{E} + 4\pi\mathbf{P}_L + 4\pi\mathbf{P}_{NL} \\ &= \mathbf{D}_L + 4\pi\mathbf{P}_{NL}, \end{aligned} \quad (3.6)$$

where $\mathbf{D}_L = \epsilon \cdot \mathbf{E}$ is the linear electric displacement vector, ϵ is the linear dielectric tensor which reduces to a real scalar quantity for the case of an isotropic, lossless material. Substituting Eqs. (3.4) and (3.6) into Eq. (3.3), and using the approximation $\nabla \cdot \mathbf{E} = 0$, we get the wave equation

$$\nabla^2 \mathbf{E}(\mathbf{r}, t) - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{D}_L(\mathbf{r}, t) = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}_{NL}(\mathbf{r}, t). \quad (3.7)$$

3.2 Time-Dependent Nonlinear Wave Equations in Dispersive Media.

Since both SPM and XPM processes involve propagating waves with time varying amplitudes, we need to find the time-dependent nonlinear wave equations in dispersive media. In general, the electric field of a linearly polarized laser pulse can be approximately expressed as a quasi-monochromatic plane wave when the change along the transverse coordinates are negligible:

$$\mathbf{E}(\mathbf{r},t) = \hat{\mathbf{e}} E(z,t) = \hat{\mathbf{e}} A(z,t)\exp[i(k_0z - \omega_0t)], \quad (3.8)$$

where $\hat{\mathbf{e}}$ is the unit vector of the polarization direction of the electric field, ω_0 is the carrier frequency, k_0 is the carrier wave number, and $A(z,t)$ is the slowly varying amplitude of the electric field.

Since the fields depends only on the spatial coordinate z , we can replace ∇^2 by $\partial^2/\partial z^2$ in Eq. (3.7). We then obtain

$$\nabla^2 E(z,t) = \exp[i(k_0z - \omega_0t)] \left(\frac{\partial^2 A(z)}{\partial z^2} + 2ik_0 \frac{\partial A(z)}{\partial z} - k_0^2 A(z) \right). \quad (3.9)$$

When solving the wave equations, several simplifying approximations are often made.¹⁻³ Among them is the slowly varying amplitude approximation, which is valid for many nonlinear effects. Since the energy among waves usually travels over a distance much longer than their wavelengths, we expect

$$\left| \frac{\partial^2 A(z)}{\partial z^2} \right| \ll \left| k_0 \frac{\partial A(z)}{\partial z} \right|. \quad (3.10)$$

So we have

$$\nabla^2 E \approx \exp[i(k_0z - \omega_0t)] \left(2ik_0 \frac{\partial A(z)}{\partial z} - k_0^2 A(z) \right). \quad (3.11)$$

If $E(z,t)$ is expressed in terms of the Fourier integral

$$E(z,t) = \int_{-\infty}^{\infty} \tilde{E}(z,\omega) \exp(i\omega t) d\omega, \quad (3.12)$$

then we have

$$D(z,t) = \int_{-\infty}^{\infty} n_0^2(\omega) \tilde{E}(z,\omega) \exp(-i\omega t) d\omega, \quad (3.13)$$

where $n_0(\omega) = \sqrt{\epsilon}$ is the linear index of refraction which is ω dependent. We can write the linear polarization term on the left-hand side of Eq. (3.7) as

$$\begin{aligned} -\frac{1}{c^2} \frac{\partial^2}{\partial t^2} D(z,t) &= -\frac{1}{c^2} \int_{-\infty}^{\infty} n_0^2(\omega) \tilde{E}(z,\omega) \frac{\partial^2}{\partial t^2} \exp(-i\omega t) d\omega \\ &= \int_{-\infty}^{\infty} \frac{n_0^2(\omega) \omega^2}{c^2} \tilde{E}(z,\omega) \exp(-i\omega t) d\omega \\ &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} k^2(\omega) A(z,t') \exp[i\omega(t'-t)] \\ &\quad \times \exp[i(k_0 z - \omega_0 t)] d\omega dt'. \end{aligned} \quad (3.14)$$

The derivation of the wave equation then proceeds by expanding $k^2(\omega)$ about the carrier frequency ω_0 in the form:

$$k^2(\omega) = k_0^2 + 2k_0 k_0^{(1)} (\omega - \omega_0) + k_0 k_0^{(2)} (\omega - \omega_0)^2 + \dots, \quad (3.15)$$

where $k_0 = k(\omega_0)$ is the propagation constant, $k_0^{(1)} = \left(\frac{\partial k}{\partial \omega} \right)_{\omega=\omega_0} = v_g^{-1}$ is the inverse of the group velocity, and $k_0^{(2)} = \left(\frac{\partial^2 k}{\partial \omega^2} \right)_{\omega=\omega_0}$ is the inverse of group velocity dispersion.

It is then possible to evaluate the integral of Eq. (3.14) by using the convenient delta function identities

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \exp[i(\omega - \omega_0)(t' - t)] d\omega = \delta(t' - t), \quad (3.16)$$

as well as

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} (\omega - \omega_0) \exp[i(\omega - \omega_0)(t' - t)] d\omega = i\delta^{(1)}(t' - t), \quad (3.17)$$

and

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} (\omega - \omega_0)^2 \exp[i(\omega - \omega_0)(t' - t)] d\omega = -\delta^{(2)}(t' - t). \quad (3.18)$$

In these relations, $\delta^{(n)}(t' - t)$ is the n th-order derivative of the Dirac delta function, with the property that

$$\int_{-\infty}^{\infty} \delta^{(n)}(t - t_0) f(t) dt = \left(\frac{\partial^n f(t)}{\partial t^n} \right)_{t=t_0} \quad (3.19)$$

when applied to a function $f(t)$. Substituting Eq. (3.15) into Eq. (3.14) and using Eqs. (3.16) to (3.19), the second term on the left-hand side of Eq. (3.7) becomes

$$\begin{aligned} & -\frac{1}{c^2} \frac{\partial^2}{\partial t^2} D(z, t) \\ &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} [k_0^2 + 2k_0 k_0^{(1)} (\omega - \omega_0) + k_0 k_0^{(2)} (\omega - \omega_0)^2] A(z, t') \\ & \quad \times \exp[i\omega(t' - t)] \exp[i(k_0 z - \omega_0 t)] d\omega dt' \\ &= \int_{-\infty}^{\infty} [k_0^2 \delta(t' - t) + i2k_0 k_0^{(1)} \delta^{(1)}(t' - t) - k_0 k_0^{(2)} \delta^{(2)}(t' - t)] A(z, t') \\ & \quad \times \exp[i(k_0 z - \omega_0 t)] dt' \\ &= [k_0^2 A(z, t) + i2k_0 k_0^{(1)} \frac{\partial A(z, t)}{\partial t} - k_0 k_0^{(2)} \frac{\partial^2 A(z, t)}{\partial t^2}] \exp[i(k_0 z - \omega_0 t)]. \end{aligned} \quad (3.20)$$

Inserting Eqs. (3.11) and (3.20) into Eq. (3.7) with the approximation of

$$\frac{\partial^2 P_{NL}(z, t)}{\partial t^2} = -\omega_0^2 P_{NL}(z, t) \quad (3.21)$$

yields the time-dependent nonlinear wave equation:

$$\boxed{i \left(\frac{\partial A(z, t)}{\partial z} + \frac{1}{v_g} \frac{\partial A(z, t)}{\partial t} \right) - \frac{1}{2} k_0^{(2)} \frac{\partial^2 A(z, t)}{\partial t^2} = -\frac{2\pi\omega_0^2}{k_0 c^2} P_{NL}(z, t) \exp[-i(k_0 z - \omega_0 t)]} \quad (3.22)$$

where v_g is the group velocity. In Eq.(3.22), the first two terms on the left hand side describes the envelope propagation at the group velocity ; the third term determines the temporal pulse broadening due to the group velocity dispersion; and the term on the right-hand side characterizes the nonlinear polarization which is responsible for different kinds of nonlinear phenomena.

3.3 Application of The Time-Dependent Nonlinear Wave Equation

--- Self- and Cross-Phase-Modulation

The nonlinear wave equation (3.22) can be further reduced to

$$\frac{\partial A(z,t)}{\partial z} + \frac{1}{v_g} \frac{\partial A(z,t)}{\partial t} = - \frac{\omega_0 n_2}{2c} |A(z,t)|^2 A(z,t), \quad (3.23)$$

where $n_2 = 3\pi\chi^{(3)}/8n$ is the nonlinear refractive index coefficient. Eq. (3.23) was derived using the following approximations: (1) linearly polarized electric field, (2) homogeneous radial fields, (3) slowly varying envelope, (4) isotropic and nonmagnetic medium, (5) negligible Raman effect, (6) frequency-independent nonlinear susceptibility $\chi^{(3)}$, and (7) neglecting group velocity dispersion, absorption, self-steeping, and self-frequency shift.

The basic mechanism and properties of SPM can be explored using the simplified wave equation (3.23).⁷ Denoting a and α the amplitude and phase of the electric field envelope $A=a \exp(i\alpha)$, Eq. (3.23) reduces to

$$\frac{\partial a(z,t)}{\partial z} + \frac{1}{v_g} \frac{\partial a(z,t)}{\partial t} = 0 \quad (3.24)$$

and

$$\frac{\partial \alpha(z,t)}{\partial z} + \frac{1}{v_g} \frac{\partial \alpha(z,t)}{\partial t} = \frac{\omega_0 n_2}{2c} a^2(z,t). \quad (3.25)$$

The analytical solutions for the amplitude and phase are

$$a(\tau) = a_0 F(\tau) \quad (3.26)$$

and

$$\begin{aligned} \alpha(\tau, z) &= \frac{\omega_0 n_2}{2c} \int_0^z a^2 dz' \\ &= \frac{\omega_0 n_2}{2c} a_0^2 F^2(\tau) z, \end{aligned} \quad (3.27)$$

where a_0 is the amplitude, $F(\tau)$ the pulse envelope, and τ the local time $\tau = t - z/v_g$.

The electronic field envelope solution of Eq. (3.23) is then given by

$$A(z, \tau) = a(\tau) \exp\left(i \frac{\omega_0 n_2}{2c} a_0^2 F^2(\tau) z\right). \quad (3.28)$$

The main physics behind the supercontinuum generation by SPM is contained in Eq. (3.28) and is displayed in Fig. 3.1. As shown in Fig. 3.1(a), the index change becomes time dependent and, therefore, the phase of a pulse propagating in a distorted medium becomes time dependent, resulting in SPM. The frequency of the electric field is continuously shifted (Fig. 3.1(c)) in time. This process is most important in the generation of femtosecond pulses.

Since the pulse duration is much larger than the optical period $2\pi/\omega_0$ (slowly varying approximation), the electric field at each given time τ within the pulse has a specific *local* and *instantaneous* frequency which is given by

$$\omega(\tau) = \omega_0 + \delta\omega(\tau) \quad (3.29)$$

where

$$\begin{aligned} \delta\omega(\tau) &= -\frac{\partial\alpha}{\partial\tau} \\ &= -\frac{\omega_0 n_2}{2c} a_0^2 z \frac{\partial F^2(\tau)}{\partial\tau}. \end{aligned} \quad (3.30)$$

The $\delta\omega(\tau)$ is the frequency shift generated at a particular local time τ within the pulse shape. This frequency shift is proportional to the derivative of the pulse envelope, which corresponds to the generation of new frequencies resulting in a wider spectrum. Fig. 3.1(c) shows the frequency distribution within the pulse shape. The leading edge is red

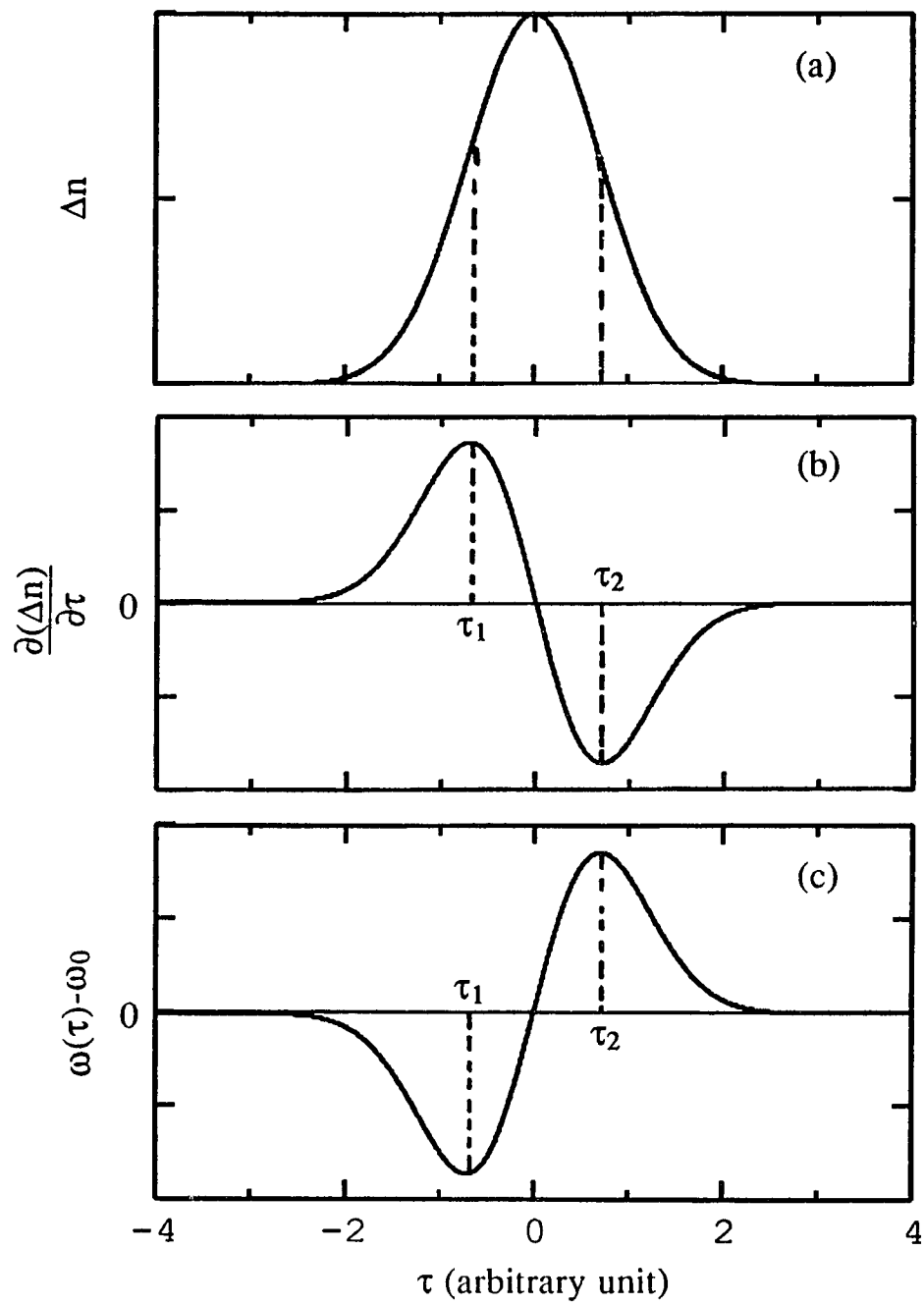


Figure 3.1. Time dependent index, phase and frequency shift generated by self-phase-modulation (SPM). (a) time-dependent nonlinear index change; (b) time rate of index change; (c) time distribution of SPM-shift frequencies $\omega(t) - \omega_0$.

shifted, the frequency of the pulse peak is nonshifted and the trailing edge is blue shifted, respectively.

The spectrum of the SPM pulse is obtained by taking the Fourier transform of the complex temporal envelope $A(z, \tau)$:

$$A(\Omega, z) = \frac{1}{2\pi} \int_{-\infty}^{\infty} A(z, \tau) \exp(i\Omega\tau) d\tau, \quad (3.31)$$

where $\Omega = \omega - \omega_0$. The intensity spectrum is given by

$$S(\Omega, z) = \frac{c}{4\pi} |A(\Omega, z)|^2. \quad (3.32)$$

In particular cases, the intensity spectrum is large compared with π , and the stationary phase method leads to

$$\Delta\omega(z)_{\max} = \frac{\omega_0 n_2}{2c} a_0^2 z \left(\left. \frac{\partial F^2(\tau)}{\partial \tau} \right|_{\tau_2} - \left. \frac{\partial F^2(\tau)}{\partial \tau} \right|_{\tau_1} \right) \quad (3.33)$$

where $\Delta\omega(z)_{\max}$ is the maximum frequency spread, τ_1 and τ_2 are the pulse envelope inflection points.

For a Gaussian laser pulse given by

$$F(\tau) = \exp\left(-\frac{\tau^2}{2\tau_0^2}\right) \quad (3.34)$$

where τ_0 is the 1/e pulse duration, the maximum frequency extent is

$$\Delta\omega(z)_{\max} = \frac{\omega_0 n_2 a_0^2 z}{c\tau_0}. \quad (3.35)$$

The maximum frequency shift (Eq. (3.35)) indicates the following salient points:

- The frequency extent is inversely proportional to the pump pulse duration. The shorter the incoming pulse, the greater the frequency extent. The first white light broadband supercontinuum pulses were generated using picosecond laser pulses.⁸
- The spectral broadening is proportional to n_2 . The supercontinuum generation can be enhanced by increasing the nonlinear refractive index.⁹
- The spectral broadening is proportional to ω_0 and z .

- The spectral broadening is linearly proportional to the amplitude a_0^2 . Therefore, multiple-excitation laser beams at different wavelengths may be used to increase the supercontinuum generation. Since the electronic field at each given time τ within the pulse has a specific *local* and *instantaneous* frequency, it is possible to induce desired frequency shift of a weak probe pulse by varying the time interval between the weak probe pulse and a strong pump who induces the time dependent index change. These features lead to the basic principle behind XPM.¹⁰⁻¹⁹

Spectral broadening due to the XPM is sketched in Fig. 3.2. An intense pulse E_1 at frequency ω_1 is used to induce the time-dependent nonlinear index change and dynamically alter the phase and frequency of a weak probe pulse E_2 at ω_2 . The spectral shift of XPM can be controlled by adjusting the time delay between the pump and probe pulses. When the pump and probe pulses overlap totally, the probe pulse has a frequency shift similar to that of the pump pulse. The leading edge of the probe pulse is red-shifted and the trailing edge is blue shifted. When the probe pulse overlaps with the leading edge of the pump pulse, the phase of the probe pulse is modulated by the index change caused by the leading edge of the pump pulse and its frequency is red-shifted. When the probe pulse overlaps with the trailing edge of the probe pulse, the phase of the probe pulse is modulated by the index change caused by the trailing edge of the pump pulse and its frequency is blue-shifted. This effect is illustrated in Fig. 3.3.

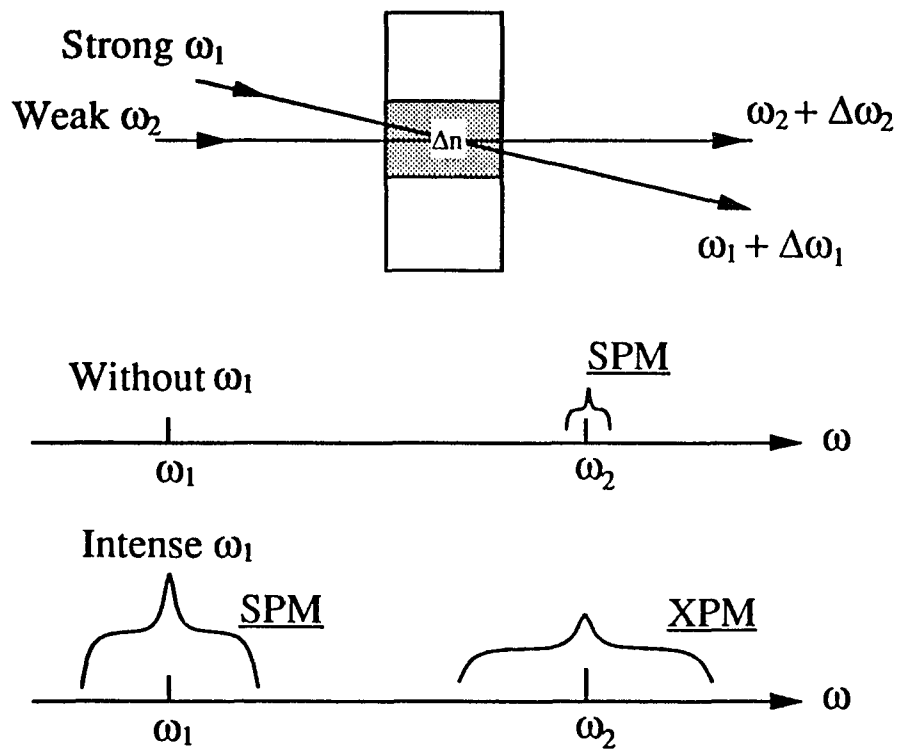


Figure 3.2. A schematic of XPM process. The induced spectral broadening of a weak probe pulse at ω_2 modulated by an intense laser pulse at ω_1 .

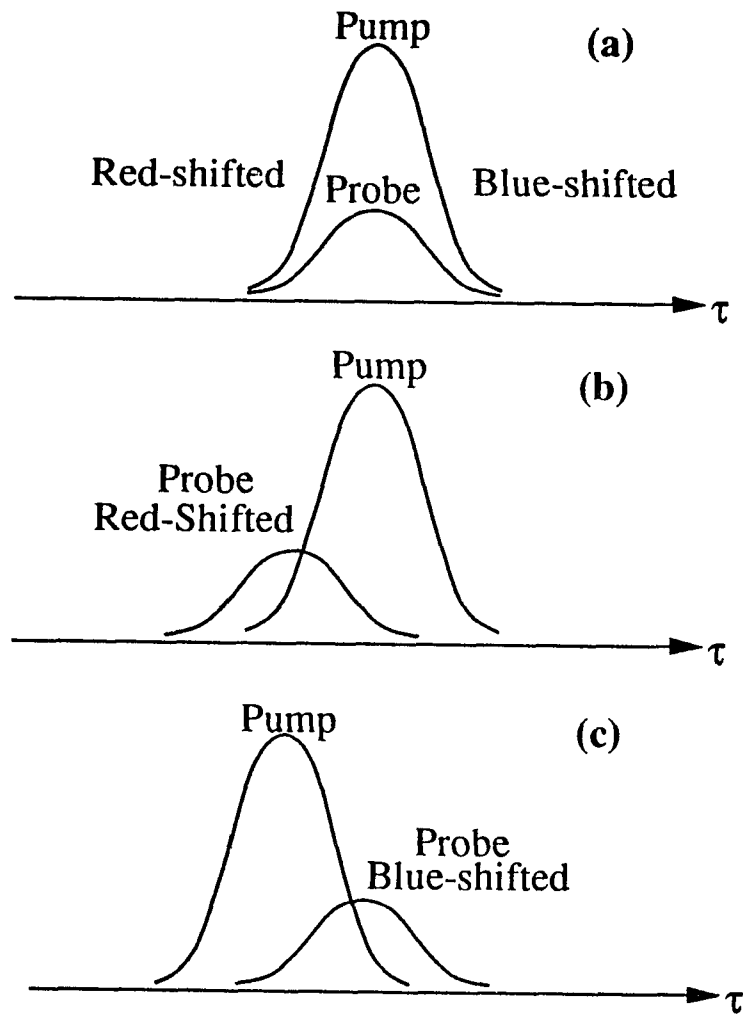


Figure 3.3 XPM with different time delays between the strong pump and weak probe pulses. (a) when the probe pulse and pump pulse are totally overlapped, the leading edge of the probe pulse is red-shifted and the trailing edge is blue-shifted; (b) when the weak probe pulse overlaps with the leading edge of the pump pulse, its frequency is red-shifted; and (c) when the weak probe pulse overlaps with the trailing edge of the pump pulse, its frequency is blue-shifted

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Chapter 4

Second Harmonic Generation Cross-Phase-Modulation

Second harmonic generation (SHG) was the first nonlinear optical process observed with lasers.¹ It is one of the most basic nonlinear effects.²⁻⁴ Over the years, SHG has been used to generate new optical frequencies. It forms the heart of the temporal auto- and cross-correlators, and is one of the elements in optical computations and logic operations.⁵ Efficient SHG is produced from a large $\chi^{(2)}$ and by phase matching the anisotropic indices of refraction. Recently, owing to the high power and short pulse duration of laser pulses, SHG has been observed in quasi-isotropic materials such as optical glass fibers.^{6,7} The third order nonlinearity $\chi^{(3)}$ is responsible for third harmonic generation and self-phase-modulation (SPM).⁸⁻¹¹ Many materials possess both large second order nonlinear susceptibility $\chi^{(2)}$ and third order nonlinear susceptibility $\chi^{(3)}$. Both SHG and SPM occur at the same time and can be coupled together. The interference between SHG and SPM alters the emission spectrum and has been called *second harmonic generation cross-phase-modulation* (SHG-XPM).¹²⁻¹⁴

SHG-XPM is an important nonlinear effect which can be used to modulate and change the spectral and the temporal distribution of the generated SHG pulse. In this chapter, a theoretical analysis of SHG-XPM using the nonlinear wave equation obtained in Chapter 3 will be given. Experimental studies on the spectral broadening and the temporal behavior of the SHG-XPM in ZnSe will be presented. Theoretical results will be compared with the experimental observations.

4.1 Second Harmonic Generation Cross-Phase-Modulation Theory

4.1.1 Wave Equations for Second Harmonic Generation Cross-Phase-Modulation

When a quasi-monochromatic laser pulses with frequencies of ω_0 and $2\omega_0$ propagates along the z axis through a noncentrosymmetric medium possessing both $\chi^{(2)}$ and $\chi^{(3)}$, the total electric field can be written as

$$E(z,t) = E_1(z,t) + E_2(z,t), \quad (4.1)$$

where

$$E_1(z,t) = \frac{1}{2} \{ A_1(z,t)\exp[-i(k_{10}z - \omega_0 t)] + \text{c.c.} \} \quad (4.2)$$

is the incident primary wave and the generated second harmonic wave is assumed to be

$$E_2(z,t) = \frac{1}{2} \{ A_2(z,t)\exp[-i(k_{20}z - 2\omega_0 t)] + \text{c.c.} \}. \quad (4.3)$$

The nonlinear polarization then can be written as

$$P_{NL}(z,t) = \chi^{(2)}(E(z,t))^2 + \chi^{(3)}(E(z,t))^3, \quad (4.4)$$

where $\chi^{(2)}$ and $\chi^{(3)}$ are the second- and third-order nonlinear susceptibilities, respectively. The third-order nonlinear susceptibility $\chi^{(3)}$ can be written as $\chi^{(3)} = \chi^{(3)'} + i\chi^{(3)''}$. The real part of $\chi^{(3)}$ contributes to the phase-modulation, while the imaginary part of $\chi^{(3)}$ arises from the induced absorption.¹⁵⁻¹⁷

If we keep only the terms synchronized with ω_0 and $2\omega_0$, the nonlinear polarization can be split into two components

$$\begin{aligned} P_{NL1}(z,t) &= \frac{1}{2} \chi^{(2)} A_1^*(z,t) A_2(z,t) \exp[-i(k_{20}z - k_{10}z - \omega_0 t)] \\ &\quad + \frac{3}{8} \chi^{(3)} \{ |A_1(z,t)|^2 A_1(z,t) + 2|A_2(z,t)|^2 A_1(z,t) \} \exp[-i(k_{10}z - \omega_0 t)] \\ &\quad + \text{c.c.} \\ P_{NL2}(z,t) &= \frac{1}{4} \chi^{(2)} A_1^2(z,t) \exp[-i(2k_{10}z + 2\omega_0 t)] \\ &\quad + \frac{3}{8} \chi^{(3)} \{ 2|A_1(z,t)|^2 A_2(z,t) + |A_2(z,t)|^2 A_2(z,t) \} \exp[-i(k_{20}z - 2\omega_0 t)] \end{aligned} \quad (4.5)$$

$$+ \text{c.c.} \quad (4.6)$$

Inserting the nonlinear polarization given by Eqs. (4.5) and (4.6) into the nonlinear wave equation (3.22) and neglecting the group velocity dispersion effect ($k_0^{(2)}=0$), we obtain the nonlinear wave equations which describe the SHG-XPM:

$$\begin{aligned} \frac{\partial A_1(z,t)}{\partial z} + \frac{1}{v_{g1}} \frac{\partial A_1(z,t)}{\partial t} = i \frac{2\pi\omega_0^2}{k_{10}c^2} \left\{ \frac{1}{2} \chi^{(2)} A_1^*(z,t) A_2(z,t) \exp[-i(k_{20}-2k_{10})z] \right. \\ \left. + \frac{3}{8} \chi^{(3)} [A_1(z,t)|^2 + 2|A_2(z,t)|^2] A_1(z,t) \right\} - \delta A_1(z,t), \quad (4.7) \end{aligned}$$

$$\begin{aligned} \frac{\partial A_2(z,t)}{\partial z} + \frac{1}{v_{g2}} \frac{\partial A_2(z,t)}{\partial t} = i \frac{8\pi\omega_0^2}{k_{20}c^2} \left\{ \frac{1}{4} \chi^{(2)} A_1^2(z,t) \exp[-i(2k_{10}-k_{20})z] \right. \\ \left. + \frac{3}{8} \chi^{(3)} [2|A_1(z,t)|^2 + |A_2(z,t)|^2] A_2(z,t) \right\} - \alpha A_2(z,t), \quad (4.8) \end{aligned}$$

where δ is linear absorption coefficient for the wave at frequency ω_0 and α is the linear absorption coefficient for the wave at frequency $2\omega_0$, v_{g1} and v_{g2} are the group velocities for the wave at frequencies ω_0 and $2\omega_0$, respectively. The first term on the right hand side of Eq. (4.7) is the loss caused by the energy transfer into the second harmonic generation at $2\omega_0$, the second term is SPM, the third term is the XPM caused by the second harmonic generation at and the fourth term is the loss caused by the material absorption at $2\omega_0$. For the equation describing the second harmonic generation wave at $2\omega_0$, the first term on the right hand side of Eq. (4.8) is the second harmonic generation, the second term is SPM, the third term is the XPM caused by the incident primary pulse at ω_0 , and the fourth term is the loss caused by the material absorption at $2\omega_0$.

For non-phase-matched SHG we can assume that $|A_1(z,t)| \gg |A_2(z,t)|$. If the absorption coefficient δ is small, Eq (4.7) can be approximated as follows:

$$\frac{\partial A_1(z,t)}{\partial z} + \frac{1}{v_{g1}} \frac{\partial A_1(z,t)}{\partial t} = i \gamma A_1(z,t)^2 A_1(z,t), \quad (4.9)$$

where the parameter $\gamma = \frac{\omega_0 n_2}{c}$, $n_2 = 3\pi \chi^{(3)}/8n$ is the nonlinear refractive index coefficient, the XPM term caused by the second harmonic pulse has been neglected. $\gamma =$

$\gamma + i\gamma'$ since $\chi^{(3)}$ is a complex quantity. Eq. (4.8), the equation for the second harmonic pulse envelop $A_2(z,t)$, reduces to

$$\frac{\partial A_2(z,t)}{\partial z} + \frac{1}{v_{g2}} \frac{\partial A_2(z,t)}{\partial t} = i\beta A_1^2(z,t) \exp[-i(2k_{10} - k_{20})z] + i4\gamma |A_1(z,t)|^2 A_2(z,t) - \alpha A_2(z,t), \quad (4.10)$$

where the parameter $\beta = \frac{2\omega_0 n_1}{c}$, $n_1 = \pi\chi^{(1)}/4n$, and the SPM of the second harmonic pulse has been neglected.

The second harmonic pulse is generated from the incident primary laser pulse, so the amplitude as well as the phase of the second harmonic pulse is coupled with the primary laser pulse. It is impossible to solve the problem analytically. However, we can simplify the problem by considering the spectral broadening and temporal distribution separately.

4.1.2 Spectral Broadening of Second Harmonic Generation

Cross-Phase-Modulation

Now we focus on the problem of spectral broadening of SHG-XPM. To simplify the problem, we exclude the amplification of the amplitude of the second harmonic pulse by the primary pump pulse. We assume that γ is a real quantity and the amplitudes of both primary and second harmonic pulses are not changing with time and propagate with the same group velocity. Eq. (4.10) can be further reduced to

$$\frac{\partial A_2(z,t)}{\partial z} + \frac{1}{v_{g1}} \frac{\partial A_2(z,t)}{\partial t} = i4\gamma |A_1(z,t)|^2 A_2(z,t). \quad (4.11)$$

Changing variables

$$\tau = t - \frac{z}{v_g}, \quad (4.12)$$

$$z = z \quad (4.13)$$

and denoting by a_j and α_j the amplitude and phase of the electric envelope, respectively,

A_j can be written as

$$A_j(z,\tau) = a_j(z,\tau) \exp[i\alpha_j(z,\tau)], \quad (4.14)$$

where τ is the local time of pump pulse and $j = 1$ and 2 , Eqs. (4.9) and (4.11) reduce to

$$\frac{\partial a_1}{\partial z} = 0, \quad (4.15)$$

$$\frac{\partial \alpha_1}{\partial z} = \gamma a_1^2, \quad (4.16)$$

$$\frac{\partial a_2}{\partial z} = 0 \quad (4.17)$$

$$\frac{\partial \alpha_2}{\partial z} = 4\gamma a_1^2. \quad (4.18)$$

We assume that the pulses at $z=0$ are Gaussian pulses and given by

$$A_1(z=0, \tau) = \sqrt{\frac{P_1}{A_{\text{eff}}}} \exp\left(-\frac{\tau^2}{2T_0^2}\right), \quad (4.19)$$

$$A_2(z=0, \tau) = \sqrt{\frac{P_2}{A_{\text{eff}}}} \exp\left(-\frac{\tau^2}{T_0^2}\right), \quad (4.20)$$

where P_1 are the peak power of the pulses, A_{eff} is the effective cross section area, T_0 is the 1/e pulse duration of the primary pump pulse, and 1/e pulse duration of the second harmonic pulse is $T_0/\sqrt{2}$. With the initial conditions defined by Eqs. (4.19) and (4.20),

we obtain the analytical solutions for Eqs. (4.15) to (4.18)

$$A_1(z, \tau) = \sqrt{\frac{P_1}{A_{\text{eff}}}} \exp\left(-\frac{\tau^2}{2T_0^2}\right) \exp[i\alpha_1(z, \tau)], \quad (4.21)$$

$$A_2(z, \tau) = \sqrt{\frac{P_2}{A_{\text{eff}}}} \exp\left(-\frac{\tau^2}{T_0^2}\right) \exp[i\alpha_2(z, \tau)], \quad (4.22)$$

$$\alpha_1(z, \tau) = \gamma \frac{P_1}{A_{\text{eff}}} \exp\left(-\frac{\tau^2}{T_0^2}\right) z, \quad (4.23)$$

$$\alpha_2(z, \tau) = 4\gamma \frac{P_1}{A_{\text{eff}}} \exp\left(-\frac{\tau^2}{T_0^2}\right) z. \quad (4.24)$$

The instantaneous SHG-XPM induced frequency chirp is obtained by differentiating the phases α_1 and α_2 with respect to time τ . They are

$$\begin{aligned} \Delta\omega_1(z, \tau) &= \frac{\partial \alpha_1(z, \tau)}{\partial \tau} \\ &= \gamma \frac{P_1}{A_{\text{eff}}} \frac{\tau}{T_0^2} \exp\left(-\frac{\tau^2}{2T_0^2}\right) z \\ &= \frac{2\omega_0 n_2}{c A_{\text{eff}}} \frac{P_1}{A_{\text{eff}}} \frac{\tau}{T_0^2} \exp\left(-\frac{\tau^2}{2T_0^2}\right) z, \\ \Delta\omega_2(z, \tau) &= \frac{\partial \alpha_2(z, \tau)}{\partial \tau} \end{aligned} \quad (4.25)$$

$$\begin{aligned}
&= 4\gamma \frac{P_1}{A_{\text{eff}}} \frac{\tau}{T_0^2} \exp\left(-\frac{\tau^2}{2T_0^2}\right) z \\
&= \frac{8\omega_0 n_2}{cA_{\text{eff}}} \frac{P_1}{A_{\text{eff}}} \frac{\tau}{T_0^2} \exp\left(-\frac{\tau^2}{2T_0^2}\right) z,
\end{aligned} \tag{4.26}$$

where $\Delta\omega_1 = \omega_1 - \omega_0$, and $\Delta\omega_2 = \omega_2 - 2\omega_0$.

The spectral broadening for the primary pump pulse arises from SPM. The spectral broadening for the second harmonic pulse arises from XPM. Eqs. (4.25) and (4.26) indicate that the frequency extents for both pulses are inversely proportional to the pulse duration; the spectral broadening is proportional to the nonlinear index coefficient n_2 ; the spectral broadening is proportional to the interaction length z ; and the broadening is proportional to the input powers of the pump pulses. It also should be noticed that the spectral broadening of the second harmonic pulse caused by SHG-XPM is 4 times that of the primary pump pulse caused by SPM.

4.1.3 Temporal Behavior of Non-Phase-matched Second Harmonic Generation Cross-Phase-Modulation

In a conventional non-phase-matched SHG the third-order nonlinearity usually can be neglected, that is $\gamma = 0$. The self-non-phase-matched term will determine the pulse shape of the SHG pulse.

To study the temporal behavior of the second harmonic pulse generated in SHG-XPM we need to solve Eqs (4.9) and (4.10) without neglecting the second harmonic generation term. When we take $F(\tau)$ to be the pulse envelope function for the primary pump pulse,^{13,14,18} that is

$$A_f(z,t) = A_{10} F(\tau/T), \tag{4.27}$$

where A_{10} is the initial amplitude of the primary pump pulse at ω_0 , $\tau = t - z/v_{g1}$ is the local time of the pump pulse and T is the pulse duration parameter, we can obtain a formal solution of $A_1(z,t)$ and $A_2(z,t)$ for the Eqs. (4.9) and (4.10)

$$A_1(z,t) = A_{10} F(\tau/T) \exp[i\gamma A_{10}^2 F^2(\tau/T)z], \quad (4.28)$$

$$\begin{aligned} A_2(z,t) = & i\beta A_{10}^2 \exp\left[-\alpha z - (2\gamma' A_{10}^2) \int_0^z F^2(U + \eta z') dz'\right] \\ & + i(2\gamma A_{10}^2) \int_0^z F^2(U + \eta z') dz' \int_0^z dz' F^2(U + \eta z') \exp(\alpha z') \\ & \times \exp\left[-2\gamma' A_{10}^2 F^2(U + \eta z')z' + 2\gamma'' A_{10}^2 \int_0^{z'} F^2(U + \eta z'') dz''\right. \\ & \left. - i\xi z' + i2\gamma' A_{10}^2 F^2(U + \eta z')z' - i2\gamma' A_{10}^2 \int_0^{z'} F^2(U + \eta z'') dz''\right], \end{aligned} \quad (4.29)$$

where $U = (t - z/v_{g2})/T$; $\eta = [n(2\omega_0) - n(\omega_0)]c\tau$ is the group velocity dispersion parameter; $\xi = [n^P(2\omega_0) - n^P(\omega_0)]2\omega_0/c$ is the phase velocity mismatching parameter; $n(\omega_0)$, $n^P(\omega_0)$ and $n(2\omega_0)$, $n^P(2\omega_0)$ are the group and phase indices of refraction at ω_0 and $2\omega_0$, respectively. The second harmonic envelope function of $A_2(z,t)$ in Eq. (4.29) depends on a complicated function of the product of β and γ . This equation is used to analyze the temporal behavior of the second harmonic pulse in SHG-XPM process.

Four calculated curves for $A_2(z,t)$ are plotted in Fig. 4.1, using Eq. (4.29) with appropriately chosen constant parameters under different conditions. The horizontal axis is a time scale. The vertical axis is the pulse intensity. Figs. 4.1(a) to 4.1(c) show the temporal profiles of SHG pulses without XPM ($\gamma = 0$). In Fig. 4.1(a) the dashed curve is the incident primary pulse, and the solid curve is the second harmonic pulse. Since the group velocity dispersion is assumed to be zero, both the primary and the second harmonic pulses travel with the same group velocity. The pulse duration of the second harmonic pulse is $1/\sqrt{2}$ of that of the primary pulse. In Fig. 4.1(b) two cases are presented in which the group velocity dispersion is nonzero. The dotted curve represents

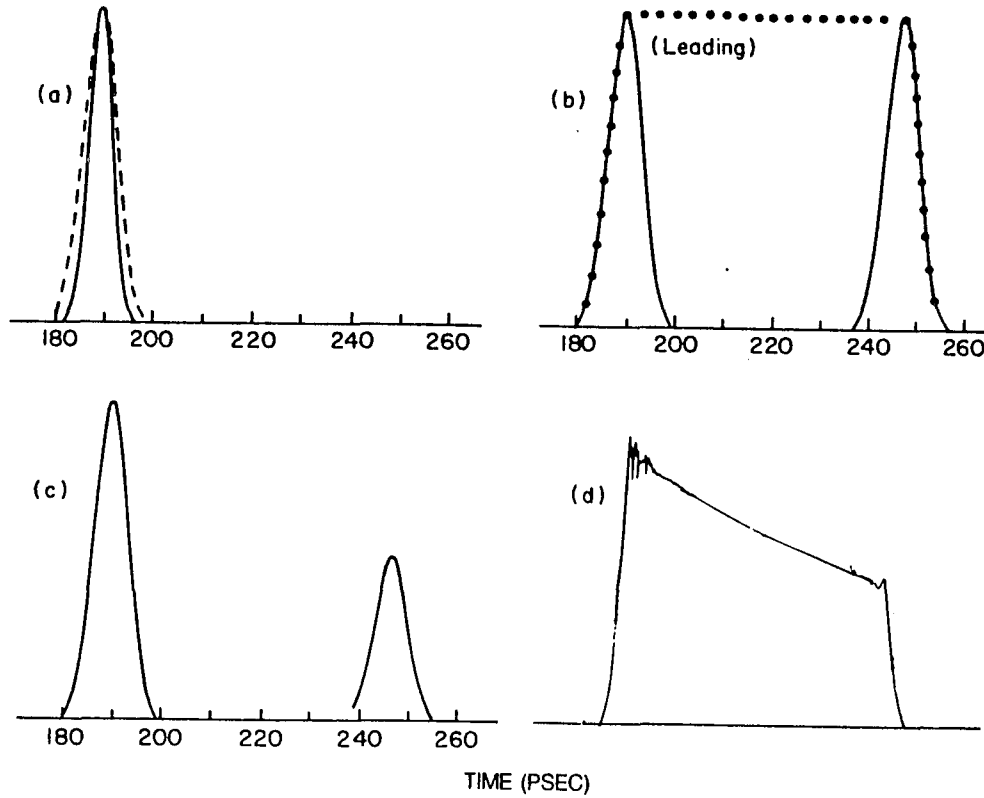


Figure 4.1. Temporal behavior of SHG-XPM. The horizontal axis is time with an arbitrary zero point and the vertical axis is the square of the electrical field amplitude. The peak heights of all curves are normalized to 1. (a) Simple phase-matched SHG: no GVD ($\eta=0$), no XPM ($\gamma=0$), and phase matching ($\xi=0$); (b) SHG with GVD ($\eta \neq 0$) and no XPM ($\gamma=0$); The dotted curve represents SHG signal in a phase-matched case ($\xi=0$). the solid curves are non-phase matched SHG. In the calculation $\xi=2.5 \times 10^4 \text{ cm}^{-1}$ was used. Two pulses are generated at the entrance and exit surface of the crystal. The separation of the two pulses depends on the length of the crystal and the pulse duration is related to the coherent length ($=1/\Delta k$). (c) Non-phase matched SHG with linear absorption. The intensity of SHG from the entrance surface is attenuated. In the calculation $\xi=2.5 \times 10^4 \text{ cm}^{-1}$ and $\alpha=0.3 \text{ cm}^{-1}$ were used. (d) SHG-XPM pulse. In the calculation, $\xi=2.5 \times 10^4 \text{ cm}^{-1}$, $\alpha=0.3 \text{ cm}^{-1}$ and induced absorption $0.23 \times 10^{-4} \gamma E_{10}^2$ were used.

the SHG pulse when the phase velocity is matched ($\xi = 0$). Efficient second harmonic signals are generated by the primary pulse through out the nonlinear medium and form a longer pulse. When the phase velocity is not matched, the SHG pulse will only be observed within the given coherent length ($1/\Delta k$) of the nonlinear crystal and the middle portion of the SHG pulse is wash out through fast oscillation of the time integration of Eq. (4.29). As long as the index difference and the crystal length are large enough, two separated peaks in time may be observed under the assumption of nondepleted input and no absorption of the generated pulse. The separation of peaks is $\Delta nL/c$, where L is the length of the crystal. Only within the coherent length of the front and exit surface of the nonlinear medium do two peaks exist. Fig. 4.1(c) shows that adding a linear absorption loss of the SHG pulse in the nonlinear material will cause the trailing edge to be attenuated. Fig. 4.1(d) displays the temporal distribution of the non-phase matched second harmonic pulse of SHG-XPM where $\gamma \neq 0$. We assume that both linear loss and induced absorption exist. Owing to the XPM induced-phase-matching, the interference of the SHG which cancels the generated SHG inside the nonlinear medium is destroyed.

The influence of the strength of third-order nonlinearity on the temporal profile of the second harmonic pulse in SHG-XPM process is displayed in Fig. 4. 2. When γE_{10}^2 is greater than a certain value ($>4 \times 10^3$), the intensity between the two peaks of the SHG pulse generated in the entrance surface and exit surface increases due to an induced phase term that washes out the destructive interference term $\exp(i\xi z')$. Further increase of γE_{10}^2 will not increase the intensity of the middle part of the SHG pulse, but will wash out the intensity oscillations of the pulse profile produced by the interaction of the XPM and the destructive interference caused by phase mismatching.

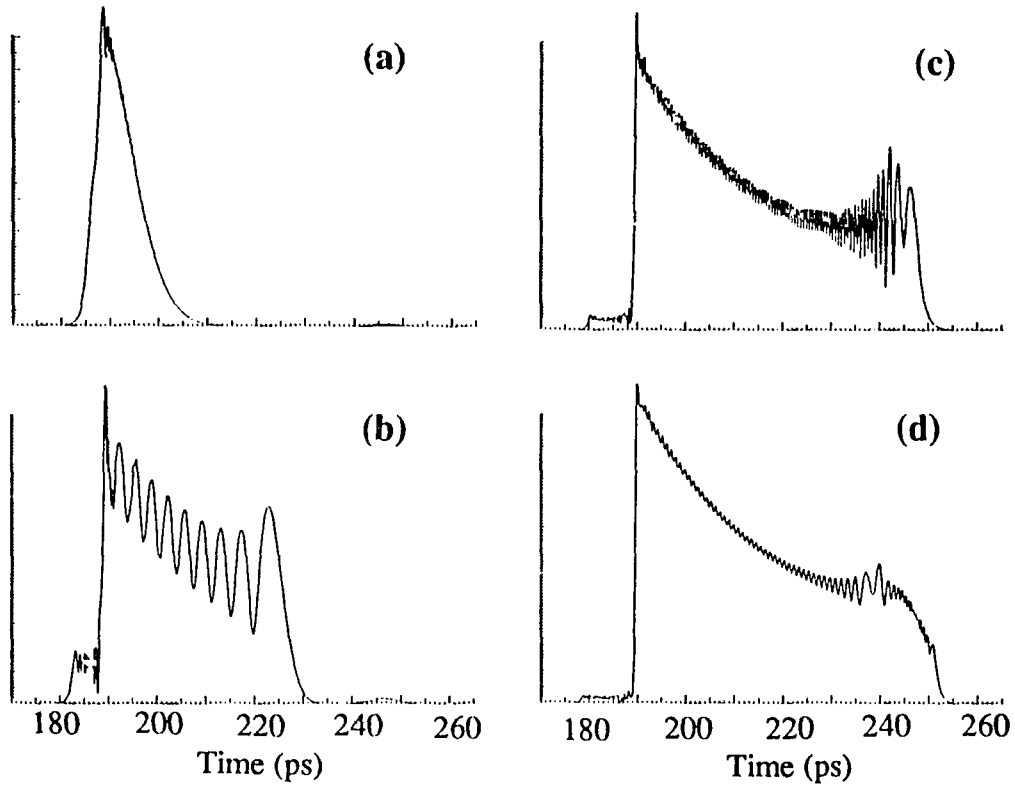


Figure 4.2. Temporal profiles of a SHG-XPM pulse as function of strength of XPM. (a) $\gamma E_{10}^2 = 5 \times 10^3$; (b) $\gamma E_{10}^2 = 10^4$; (c) $\gamma E_{10}^2 = 3 \times 10^4$; (d) $\gamma E_{10}^2 = 6 \times 10^4$. In the calculation induced absorption was used.

4.2 Experimental Studies of Second Harmonic Generation Cross-Phase-Modulation in ZnSe

4.2.1 Sample Preparations

ZnSe crystals possess both large second- and third-order nonlinearities. They are very suitable for the investigation of SHG-XPM process. The second-order nonlinearity, Pockels coefficient r_{ijk} , of ZnSe is about 2×10^{-12} m/v (ref. 19) and the third-order nonlinear index of refraction n_2 measured from the optical Kerr effect by us is about half the value of $n_2(\text{CS}_2) = 2 \times 10^{-11}$ esu.

The linear index of refraction and dispersion of ZnSe crystal at different wavelengths are listed in Table 4.1.

In the experiment, two types of ZnSe materials were used: single crystal and polycrystalline. A single crystal of ZnSe of about 1.6 cm in length was grown by Philips Laboratories.¹⁹ Polycrystalline ZnSe samples of 0.2, 0.5, 2.2, and 5.0 cm was purchased from Janos Inc.. All the samples are polished at the input and output ends.

An organic crystal (-)-2-(α -methylbenzylamino)-5-nitropyridine (MBA-NP) of 1.9 cm in length was also used in this experiment. The second-order nonlinear susceptibility of MBA-NP crystal is about 10 times of that of LiNO_3 .²⁰ Its third-order nonlinear susceptibility is about the order of 10^{-3} esu.²¹

4.2.2 Laser System

A mode-locked Nd:glass laser was the workhorse of ultrafast nonlinear optics and spectroscopy in 1970's and is still in use today. The laser system we used to investigate the SHG-XPM is shown in Fig. 4.3. The generation of a single intense picosecond laser pulse is usually accomplished by using a glass mode-locked oscillator, a single pulse selector, and an amplifier. The laser medium was phosphate glass. The phosphate glass has a large gain coefficient and lower nonlinear index n_2 than silicate glass by at least

Table 4.1. Linear index of refraction and dispersion in multicrystalline ZnSe.

λ (nm)	n	$dn/d\lambda$ (nm ⁻¹)	$n-\lambda(dn/d\lambda)$	$\Delta t/L$ (ps/mm)*
1100	2.4805	-82.6	2.571	-0.04
1060	2.4842	-93.0	2.583	0.00
560	2.6577	-99.8	3.217	2.21
550	2.6699	-1070.0	3.259	2.25
540	2.6830	-1160.0	3.309	2.42
530	2.7124	-1250.0	3.360	2.59
520	2.7124	-1350.0	3.414	2.77
510	2.7289	-1460.0	3.474	2.97
500	2.7468	-1590.0	3.542	3.20

* $\Delta t/L$ is time delay per unit length relative to 1060 nm light.

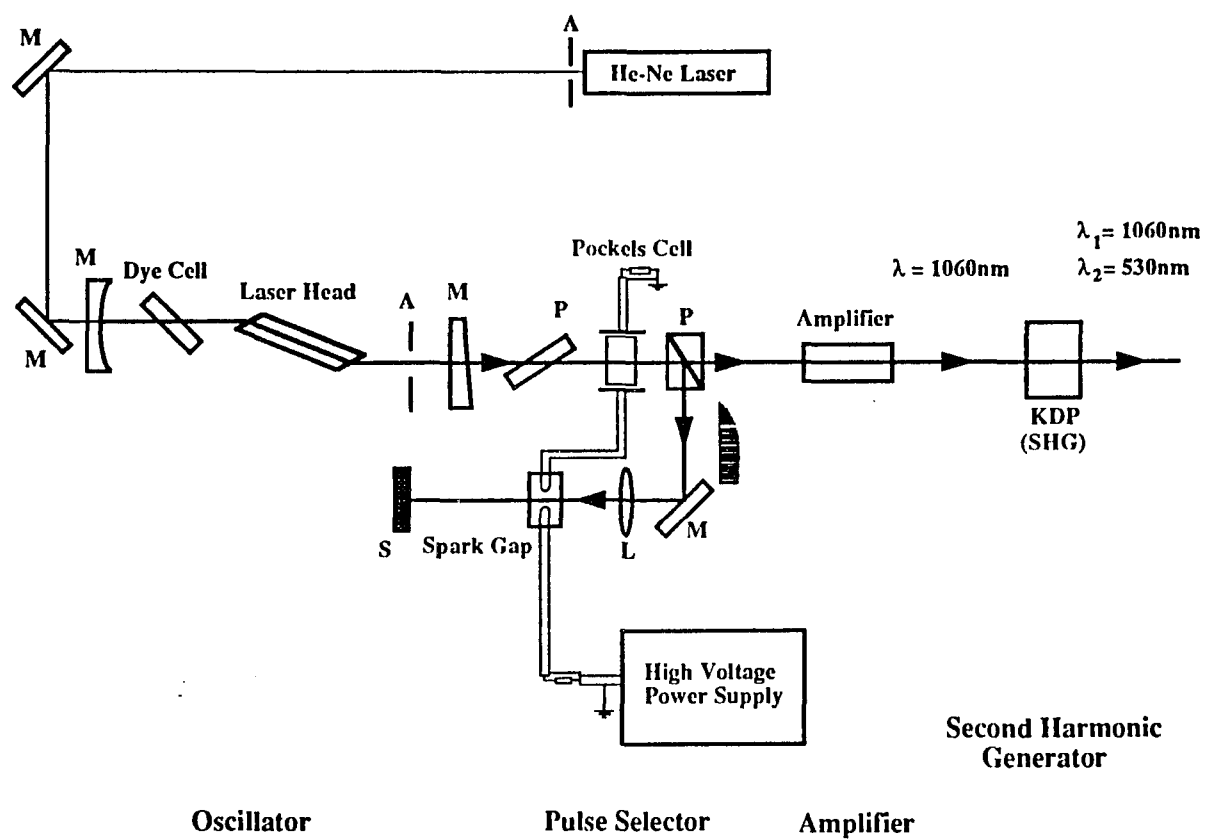


Figure 4.3. Mode-locked Nd:glass laser system. He-Ne laser is used for alignment. M: mirrors; P: polarizers.

a factor of 1.4 and 1.1, respectively.²² The characteristics of Nd-doped glass as a mode-locked oscillator-amplifier system have been reported by many investigators.²²⁻²⁴

The output pulse train from the oscillator contains about 100 pulses at the wavelength of 1054 nm separated by the round trip time of a pulse in the cavity. A single pulse is selected from the pulse train with a transverse laser-triggered spark gap connected by a double-shielded coaxial cable with a KD*P Lasermetric Pockels cell. The single pulse is then amplified by a one stage amplifier. The pulse duration measured by two-photon-fluorescence or a streak camera for a pulse selected out of the train is 8 ps. A visible pulse at 527 nm is generated by a KDP crystal using the second harmonic generation of the amplified pulse at 1054 nm.

The output laser pulse at 1054 nm and 527 nm wavelengths have about 2 mJ and 200 μ J pulse energies, respectively. The fundamental wavelength pulse and the second harmonic pulse can be separated using color filters R85 and 1-75.

4.2.3 Experimental Setup for Spectral Broadening of Second Harmonic Generation Cross-Phase-Modulation

The experimental arrangement used to generate and detect the SHG-XPM spectral broadening is shown in Fig. 4.4. The 1054 nm laser pulse generated from the laser system was weakly focused into the sample. The incident laser energy was controlled by changing the neutral-density filter F_1 . The spot size at the sample was about 1.5 mm in diameter. A polycrystalline ZnSe sample of 2.2 cm in length and a single crystal of ZnSe of about 1.6 cm in length were used in the experiment. The second harmonic produced in the polycrystalline ZnSe sample was about 10 nJ when the full power of the laser pulse was used. The output signal light was sent through a 1/2 m Jarrell-Ash spectrograph to measure the spectral distribution of the signal light. The 1054 nm incident laser was filtered out by using color filters F_2 .

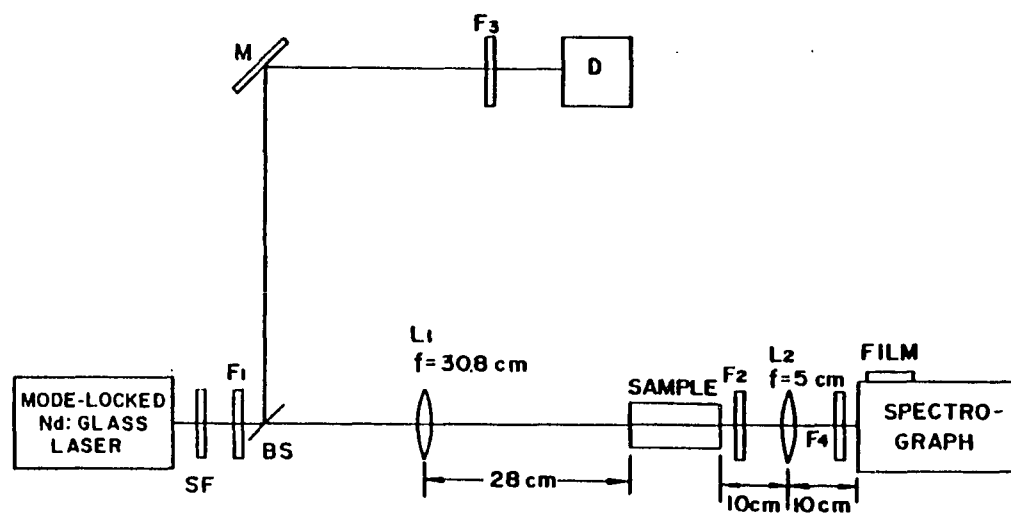


Figure 4.4. Experiment setup for SHG-XPM spectral broadening. L_1 and L_2 are lenses, SF is a spectral filter to select either 1060 nm or 530 nm or both wavelength lasers. F_1 , F_3 , and F_4 are sets of color filters and neutral density filters. F_1 consists of Hoya R720, Corning 2-3-67, and variable neutral density filters. F_2 is a set of Corning color filters.

4.2.4 Experimental Results of Spectral Broadening of Second Harmonic Generation Cross-Phase-Modulation

Typical spectra for non-phase-matching SHG-XPM generated in the 2.2 cm polycrystalline ZnSe crystal by 1054 nm laser pulse of various pulse energies are displayed in Fig. 4.5. The spectrum from a quartz sample was included in Fig. 4.5 for reference. Fig. 4.6 displays the spectral profiles of SHG-XPM generated in 2.2 cm ZnSe crystal pumped 1054 nm laser pulse with different intensities.

The salient features of the SHG-XPM spectrum indicates that the extent of the spectral broadening about the SHG line at 527 nm depends on the incident 1054 nm pump laser pulse. When the incident laser pulse energy was 2 mJ, there was significant spectral broadening of about 1100 cm^{-1} on the Stokes side and 770 cm^{-1} on the anti-Stokes side. There was no significant difference in the spectral broadening distribution measured in the single and polycrystalline materials. The intensity of induced spectral broadening was greater in polycrystalline ZnSe due to the longer length of the crystal.

The spectral width of SHG-XPM is plotted for the Stokes and anti-Stokes sides as a function of the incident laser energy in Fig. 4.7. The salient feature of Fig. 4.7 is that the Stokes side of the SHG-XPM is broader than the anti-Stokes side of the SHG-XPM. When the incident pulse energy was less than 1 mJ, the spectral broadening was found to be monotonically increasing with the pulse energy of the 1054 nm pump pulse, this is very close to the theoretical prediction for the spectral broadening in Eq. (4.26).

The spectral broadening generated by sending an intense 8 ps laser pulse at 527 nm alone through these ZnSe crystals was also measured to compare with the SHG-XPM spectral broadening. The observed spectral broadening was only about 200 cm^{-1} when the energy of the 527 nm pulse was over 0.2 mJ. This measurement suggests that the SPM process from the 10 nJ SHG pulse in ZnSe is negligible in comparison with the SHG-XPM which induce 1000 cm^{-1} spectral broadening.

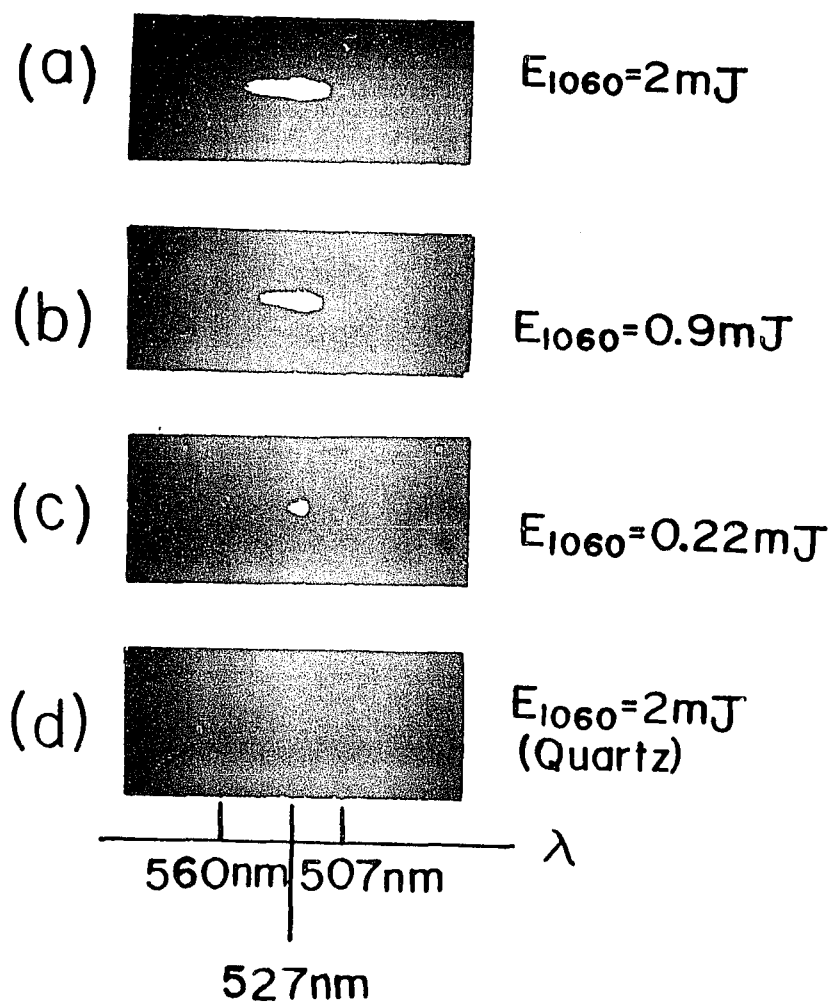


Figure 4.5 SHG-XPM spectra in ZnSe crystal excited by an intense 1060 nm laser pulse. (a) $E_{1060}=2$ mJ, (b) $E_{1060}=0.9$ mJ, (c) $E_{1060}=0.22$ mJ, and (d) ZnSe crystal was replaced by a 3.7-cm-long quartz crystal, $E_{1060}=2$ mJ.

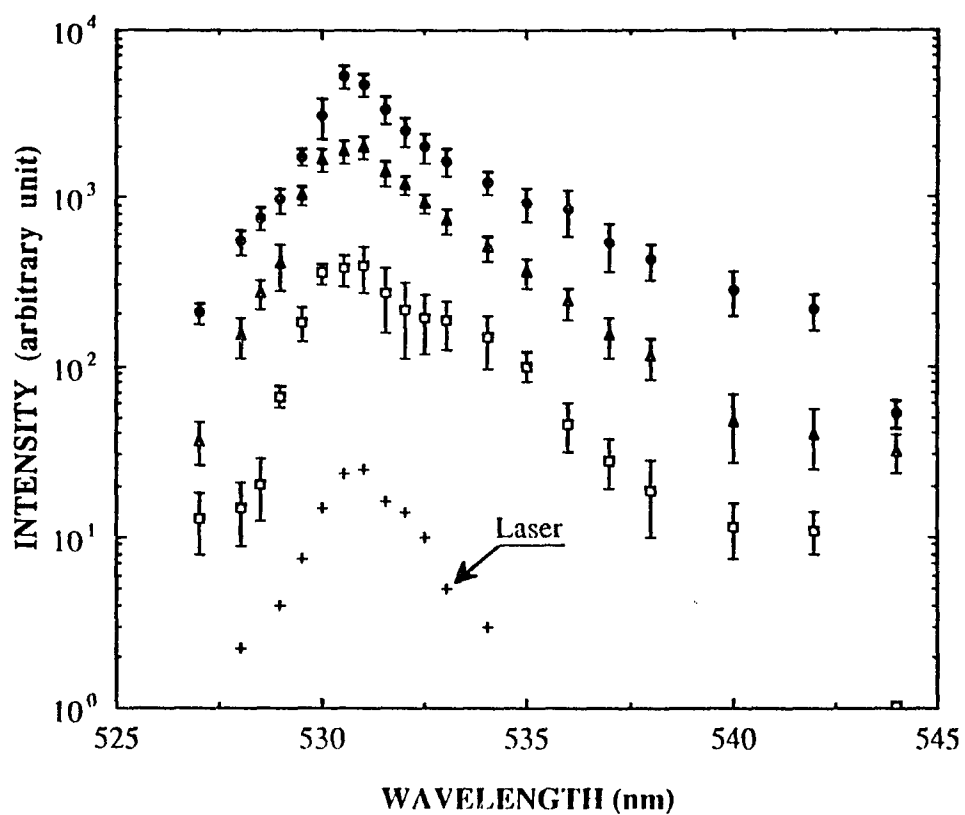


Figure 4.6. Spectral profiles of SHG-XPM generated in 2.2 cm ZnSe crystal pumped by 1060 nm laser pulse. ●: full intensity I_0 , Δ: $0.268I_0$, and □: $0.0697I_0$.

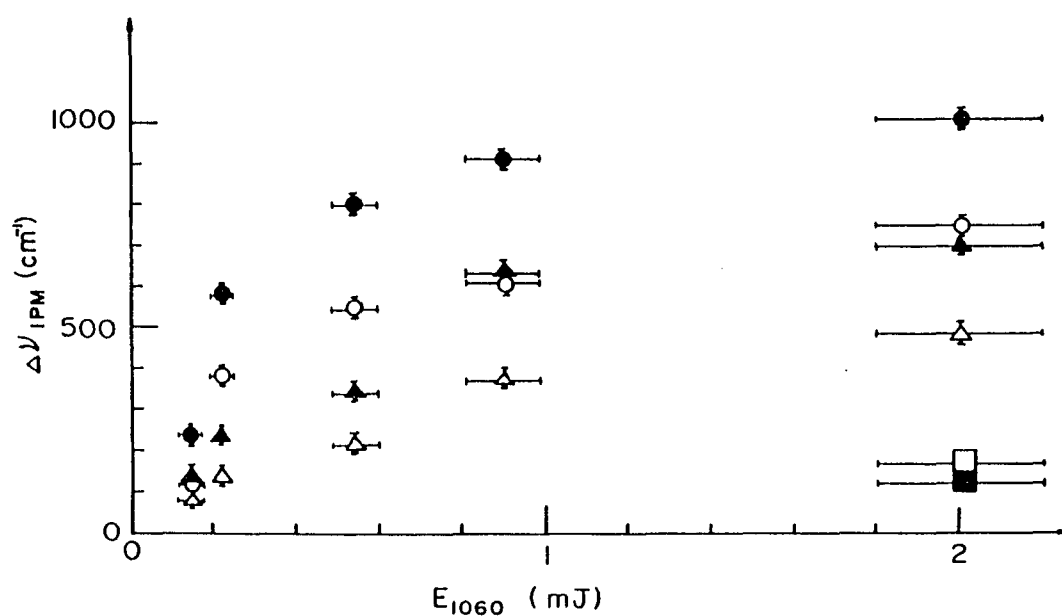


Figure 4.7. Intensity dependence of SHG-XPM in ZnSe pumped by a 1060 nm laser pulse. The horizontal line is the incident laser energy. ●: 2.2-cm-long polycrystalline ZnSe anti-Stokes broadening. ○: 2.2-cm-long polycrystalline ZnSe Stokes broadening. ▲: 1.6-cm-long single crystal ZnSe anti-Stokes broadening. △: 1.6-cm-long single crystal ZnSe Stokes broadening. □: 3.7-cm-long quartz crystal Stokes broadening. The measured $\Delta\nu$ is defined to be the frequency spread from 530 nm to the furthest detectable wavelengths either photographically or by an OMA.

The frequency shift of the primary pump pulse produced by the SPM process was also measured to compare with the spectral broadening of the SHG pulse. Fig. 4.8 shows the frequency shifts of the primary pump pulse and the SHG pulse. The dotted curve is the frequency shift of the SHG pulse and the empty-squared curve is the frequency shift of the primary laser pulse. The pump pulse energy is about 2 mJ for both curves. The salient feature of Fig. 4.8 is that the spectral broadening of the SHG pulse is larger than that of the primary laser pulse. This feature can be quantitatively explained by the theory of the spectral broadening which shows that spectral broadening of the second harmonic pulse caused by SHG-XPM is larger than that of the primary pump pulse caused by SPM [see Eqs. (4.25) and (4.26)].

4.2.5 Experimental Setup for Temporal behaviors of Second Harmonic Generation Cross-Phase-Modulation

The experimental setup for the measurement of the temporal properties of the SHG-XPM is shown in Fig. 4.9. The laser system used was the same as the system used for the spectral broadening measurement. The only difference is that a portion of the primary laser pulse at 1054 nm was converted into its second harmonic in the system by a KDP crystal. This SHG pulse at 527 nm was used as a reference pulse. The spot size of the pump laser at the sample was about 1.5 mm in diameter. The SHG produced in ZnSe sample was about 10 nJ. The incident laser energy was controlled by changing the neutral-density filter F_1 . Using a beam splitter, the primary pump pulse at 1054 nm and the SHG pulse at 527 nm were separated into two different paths. The weaker 527 nm reference pulse was used for calibration and passed through a fixed distance in air to set the zero reference time for the streak camera. Only 1054 nm pulse was used to pump the ZnSe samples. The pump pulse was almost collimated through the entire length of the sample. The temporal profile and the propagation time of the pulses from the ZnSe and the reference 527 nm pulse were measured by a Hamamatsu 2 ps streak camera

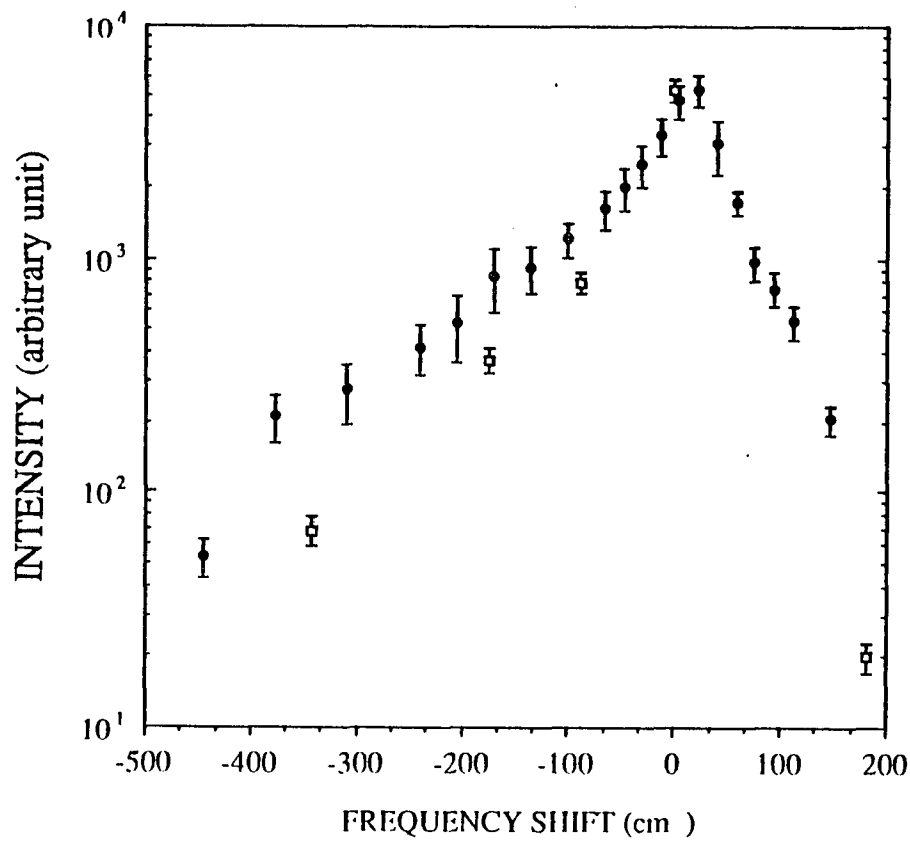


Figure 4.8. Frequency shifts of the primary pump laser and the SHG pulses of SHG-XPM in 2.2cm-long polycrystalline ZnSe. ●: frequency shift of SHG, □: frequency shift of primary pump laser.

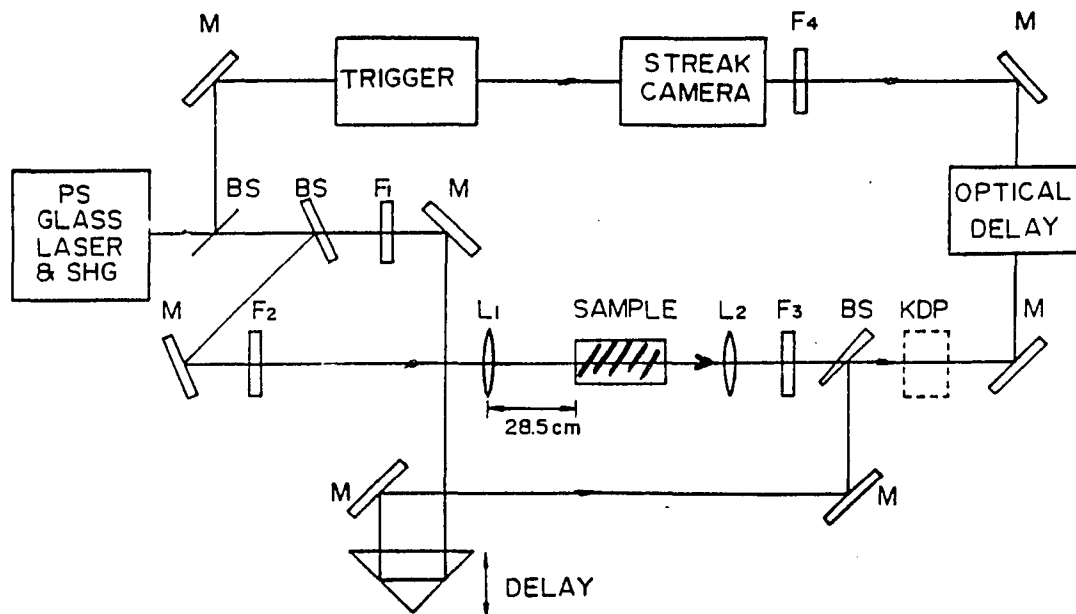


Figure 4.9. Experimental setup to measure the temporal profile and propagation time of the pulses. BS: beamsplitter; F's: filters; M: mirrors; L: lens; dashed KDP: 100-mm-thick KDP crystal which was only inserted during the measurement of the propagation delay time of 1060 nm pump laser pulse travel through air or ZnSe.

system. The zero time between reference 527 nm and primary pump pulse at 1054 nm was determined with the help of a 100 μm thick KDP crystal placed in the path of 1054 nm pulse. The absolute time delay for the spectrally broadened pulse traveling through the sample was determined by measuring the time separation between the 527 nm reference pulse and the spectrally broadened signals in the streak camera. Polycrystalline ZnSe samples of thickness 0.2, 0.5, 1.0, 2.2, 5.0 cm, and a single crystal ZnSe of 1.6 cm thickness were used in the experiment. The SHG pulse profile of 1.9 cm MBA-NP crystal is also measured.

4.2.6 Experimental Results for The Temporal Behavior of The Second Harmonic Generation Cross-Phase-Modulation

The temporal profiles and propagation time of an intense 1054 nm pump pulse and SHG-XPM pulses at different λ propagating through a 2.2 cm ZnSe polycrystalline sample are shown in Fig. 4.10. The reference pulse was used to mark an arbitrary zero time point. A pulse delay of 119 ps at 1054 nm was observed in Fig. 4.10(a) when an intense 1054 nm pulse passed through the crystal, while the total signal of SHG-XPM with a wavelength spread from 500 to 570 nm displayed in Fig. 4.10(b) indicated a sharp spike at 119 ps and a long plateau spanning from 119 to 149 ps. Using 10 nm bandwidth narrow band filters, pulses of selected wavelengths from the SHG-XPM have also been measured. Fig. 4.10(c) - (d) show the temporal distribution of two pulses with wavelengths centered at 530 nm and 550 nm, respectively. All the traces from Figs. 4.10(b) - (d) indicate that the induced spectrally broadened pulses have one major component emitted at nearly the same time as the 1054 nm incident pulse [Fig. 4.10(a)]. The selected 10 nm SHG-XPM away from the second harmonic wavelength has shown a dominant pulse distribution generated at the end of the crystal. When a weak 3 nJ calibration pulse at 527 nm propagated alone through the 2.2 cm ZnSe, an output pulse at about 149 ps was observed which is 60 ps later than the 1054 nm pulse. The

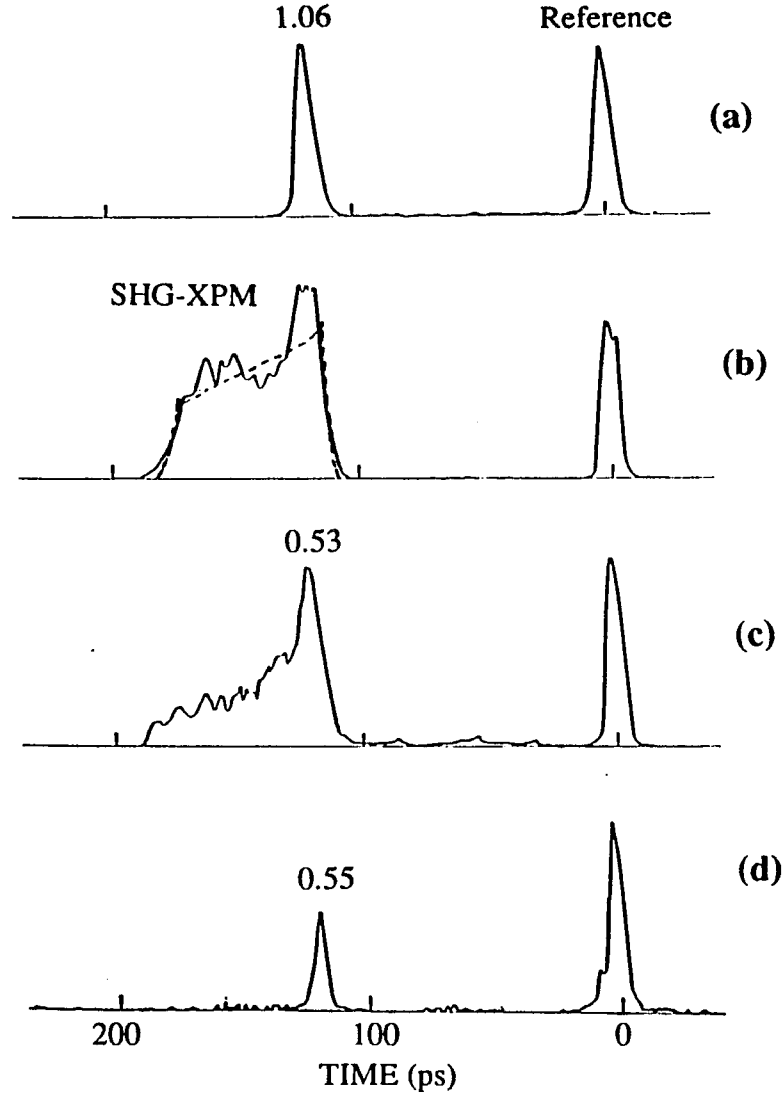


Figure 4.10. Temporal profile and propagation delay time of the SHG-XPM in 2.2 cm ZnSe crystal. (a) incident 1060 nm laser pulse, (b) SHG XPM signal of all visible spectra, (c) selected 530 nm from SHG XPM, and (d) selected 550 nm from SHG XPM. Time profiles were measured with a 2-ps resolution streak camera system. The right-handside of the time scale is the leading time. The vertical scale is an arbitrary intensity scale. The dashed curve in (b) is calculated from Eq. (4.29) with $\xi=2.5\times10^4\text{ cm}^{-1}$, $\alpha=0.3\text{ cm}^{-1}$ and induced absorption $0.23\times10^{-4}\gamma E_{10}^2$.

measurement is in very good agreement with the results calculated using the data provided in Table 1.

A dashed curve calculated for the temporal distribution of the SHG-XPM from Eq. (4.29) is plotted in Fig. 4.10(b) to fit the experimental curve. We obtain good fit to the experimental results by choosing suitable parameters.

Fig. 4.11 displays the temporal profiles of SHG-XPM pulses at different λ propagating through polycrystalline ZnSe of 0.2 cm in length. Fig. 4.12 shows the pulse shapes of the SHG-XPM pulses generated from 5 cm ZnSe crystals. These results are similar to the results obtained from the 2.2 cm ZnSe crystal. The change in the sample length results in the change of the pulse duration of the SHG-XPM pulse.

The temporal distribution of the SHG signal of the primary laser pulse passing through the 1.9 cm MBA-NP crystal is displayed in Fig. 4.13. The salient feature of the SHG pulse is that the signal has two separate peaks in time generated from the entrance and exit surfaces of the sample. This is due to the the destructive interference arising from the phase mismatching. Since MBA-NP possesses a small third-order nonlinearity, the SHG-XPM effect is not large enough to wash out the destructive interference.

4.3 Discussions

Although the simplified theory of XPM can quantitatively explain the spectral broadening features of SHG-XPM, there are some differences between the theoretical prediction of Eqs. (4.25) and (4.26) the experimental results. Eq. (4.26) shows that the spectral broadening of SHG-XPM is linear with the input intensity of the primary pulse. The experimental results in Figs. 4.4 and 4.5 show a more complicated relationship between the spectral broadening and the intensity of the pump pulse. Eqs. (4.25) and

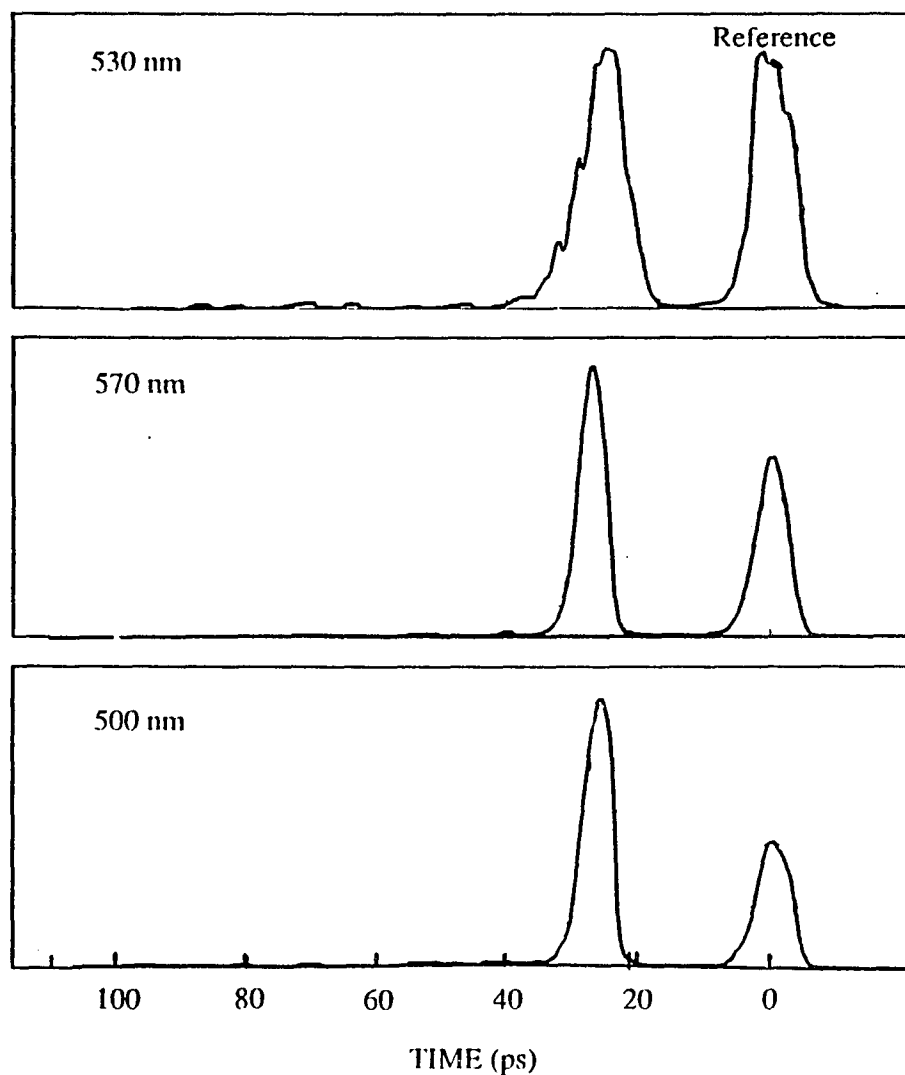


Figure 4.11. Temporal profiles of SHG XPM pulses at different wavelength generated from a 0.2-cm-long ZnSe crystal pumped by 1060 nm laser pulse. The reference pulse marks an arbitrary zero time.

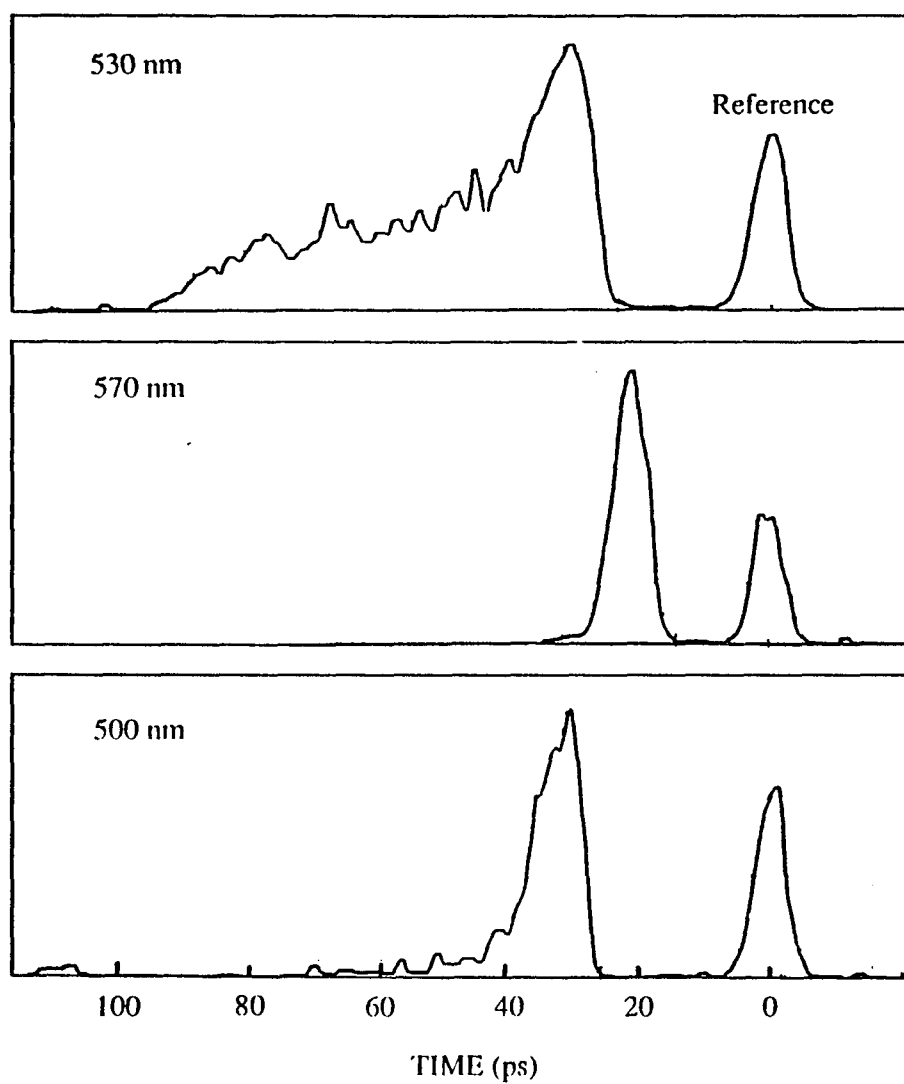


Figure 4.12. Temporal profiles of SHG XPM pulses at different wavelength generated from a 5-cm-long ZnSe crystal pumped by 1060 nm laser pulse. The reference pulse marks an arbitrary zero time.

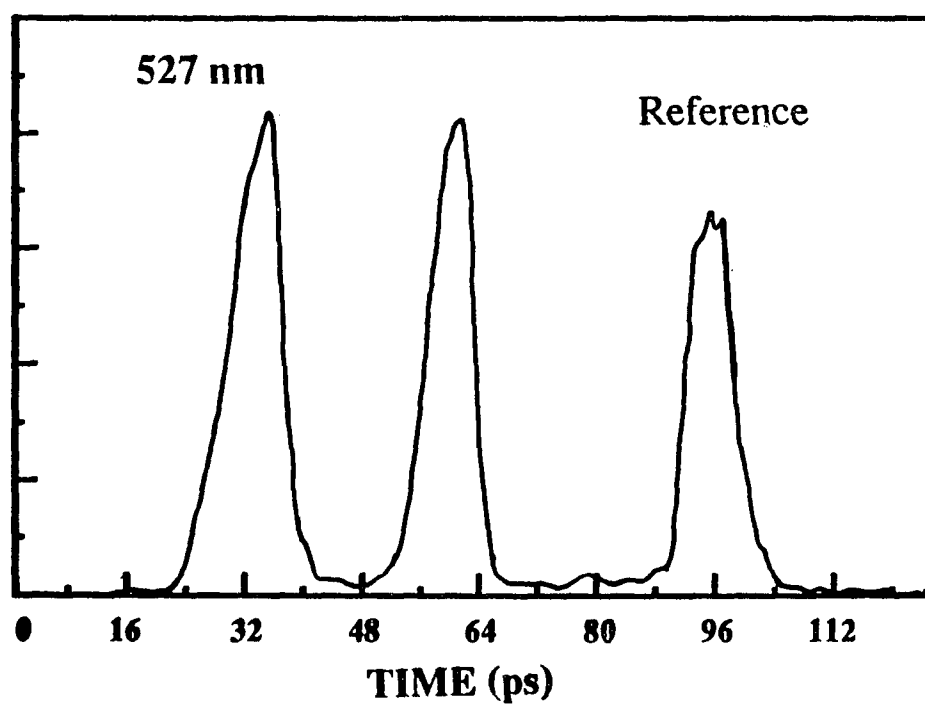


Figure 4.13. Temporal profiles of nonphase matched SHG pulse generated from a 1.9-cm-long MBA-NP crystal. The two peaks were generated from the entrance and exit surfaces of the crystal, respectively.

(4.26) show that the spectral broadening of the second harmonic pulse caused by SHG XPM is 4 times that of the primary pump pulse caused by SPM. While the experimental results in Fig. 4.7 show that the spectral broadening of the second harmonic pulse caused by SHG-XPM is about 1.5 times of that of the primary pump pulse caused by SPM. These differences may be explained by the assumption made in the theoretical evaluation. To simplify the problem, we neglected the generation of the SHG through out the sample and assumed there is no group velocity dispersion between the primary pulse and the SHG pulse. But in the experiment, the SHG pulse is generated throughout the whole sample, and there is a group velocity mismatch between the pump pulse and SHG pulse.

The sharp spike of the selected 550 nm pulse profile in Fig. 4.8(d) can be qualitatively explained via the walk-off cancellation of the phase modulation. At any given location in the crystal except the exit layer, the red-shifted front part of the phase-modulated SHG pulse can eventually be canceled by the blue-shifted trailing part of the phase modulated SHG pulse. Therefore, spectral broadening about the second harmonic frequency of the pump laser pulse will mainly occur at the end of the crystal where the walk-off between 527 and 1054 nm is insignificant. This is another evidence of XPM.

The SHG-XPM can be used to control the spectrum and temporal profiles of the SHG by changing the intensity of the input primary pulse, the nonlinearities and the length of the nonlinear material.

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Chapter 5

Degenerate Cross-Phase-Modulation in Multimode Optical Fibers

Self-phase modulation¹⁻⁵ and cross-phase modulation² processes in single mode optical fibers⁶⁻⁸ play important roles in pulse propagation. Large core multimode optical fibers are commonly used to effectively collect signal and produce uniform intensity distribution among the modes. These large core optical fibers are used in wideband optical signal transmission over short distances, especially in medical and semiconductor diagnostic systems. Recently it was shown⁹⁻¹⁰ that the energy of a pulse can be distributed among a few lower modes, using nonlinear optical effects of cross-phase-modulation and stimulated Raman scattering. This research⁹⁻¹⁰ demonstrates the possibility of using large core multimode optical fibers for long distance high speed communication links. Furthermore, future-generation medical diagnosis and ablation fiber-based systems^{11,12} will depend on the spectral as well as temporal properties of high intensity optical pulses traveling in short-multimode optical fibers. Therefore, linear and nonlinear optical pulse propagation, modal distribution,¹³ induced focusing,⁹ SFPM effect,¹⁴ and spectral broadening and distribution^{15,16} in multimode optical fibers are of great interest.

In this chapter, the spectral broadening caused by DXPM¹⁷⁻¹⁹ between different modes in short multimode optical fibers are investigated both theoretically and experimentally.

The theoretical analysis of DXPM in multimode optical fibers using the nonlinear wave equation obtained in Chapter 3 is given in Section 5.1. In Section 5.2, the experimental investigation of the DXPM spectral broadening of picosecond laser pulses

propagating through short multimode glass optical fibers are presented. Comparison of the measurements in multimode optical fibers with those in single mode optical fibers is also presented.

5.1 Simple Model of Degenerate-Cross-Phase-Modulation in Multimode Optical Fibers

When a light pulse is incident upon a multimode optical fiber along the z direction, it excites several modes, which are allowed by the characteristics of the fiber. The polarization of the light in the different modes can be assumed to be the same as the polarization of the input pulse in short fibers. For short fibers, the coupling between different modes can be neglected. So the electric field of the optical pulse in j th mode can be expressed as

$$E_j(\mathbf{r}, t) = \frac{1}{2} \{ A_j \Phi_j(x, y) \Theta_j(z, t) \exp[i(k_j z - \omega_0 t)] + \text{c. c.} \} \quad (5.1)$$

where A_j is the amplitude, $\Phi_j(x, y)$ is the transverse distribution function determined by the characteristics of the fiber, and $\Theta_j(z, t)$ is the pulse shape, and k_j is the propagation constant of the pulse in j -th mode of the optical fiber, respectively.

In long, low loss, multimode optical fibers it is known that the energy is almost uniformly distributed among the excited modes.²⁰ It has been demonstrated that in a nonlinear multimode optical fiber under certain conditions the energy can be distributed among only a few lowest modes.²¹ This effect was measured in the time and space domains experimentally.^{9,10,22} If the details of the transverse distribution of each mode are neglected, the total electric field can be expressed as²³

$$E(z, t) = \frac{1}{2} \left(\sum_j C_j \Theta_j(z, t) \exp[i(k_j z - \omega_0 t)] + \text{c. c.} \right) \quad (5.2)$$

where C_j is the effective amplitude of the j -th mode. The nonlinear polarization is

$$P_{NL}(z,t) = \chi^{(3)}(E(z,t))^3. \quad (5.3)$$

The components of the nonlinear polarization can be shown to be

$$P_{NLj}(z,t) = \frac{3}{8} \chi^{(3)} \left\{ |C_j \Theta_j(z,t)|^2 + 2 \sum_{k \neq j} |C_k \Theta_k(z,t)|^2 + 2 \sum_{k \neq j, l} C_k \Theta_k(z,t) C_l^* \Theta_l^*(z,t) \exp[-i(k_k - k_l)z] \right\} \exp[i(k_j z - \omega_0 t)], \quad (5.4)$$

where the first term on the right-hand side is the SPM term and the second and third terms are the XPM terms. Since the frequency for all the different modes are the same, it is degenerate XPM (DXPM). Within the picosecond time envelope, the third term oscillate rapidly since $k_k \neq k_l$ for $k \neq l$. Their contributions to the nonlinear polarization are washed out. Therefore, the nonlinear polarizations reduces to

$$P_{NLj}(z,t) = \frac{3}{8} \chi^{(3)} \left(|C_j \Theta_j(z,t)|^2 + 2 \sum_{k \neq j} |C_k \Theta_k(z,t)|^2 \right) \exp[i(k_j z - \omega_0 t)]. \quad (5.5)$$

By inserting the nonlinear polarization (5.5) into the nonlinear wave equation (3.22), the nonlinear wave equation for the DXPM in multimode optical fibers is obtained:

$$\frac{\partial \Theta_j(z,t)}{\partial z} + \frac{1}{v_{gj}} \frac{\partial \Theta_j(z,t)}{\partial t} = i \frac{\omega_0 n_2}{c} \left(|C_j \Theta_j(z,t)|^2 + 2 \sum_{k \neq j} |C_k \Theta_k(z,t)|^2 \right) \Theta_j(z,t) \quad (5.6)$$

where $n_2 = 3\pi\chi^{(3)}/8n$ is the nonlinear refractive index coefficient. In deriving Eq. (5.6), the group velocity dispersion effects was neglected.

Each mode is assumed to be Gaussian in shape at $z = 0$, given by

$$\Theta_j(z = 0, t) = \exp[-t^2/T^2], \quad (5.7)$$

where T is the $1/e$ pulse duration. Neglecting the mode coupling in short multimode optical fibers,^{15, 23} the solutions for Eq. (5.6) are obtained:

$$\Theta_j(z,t) = \exp[-(t - z/v_{gj})^2/T^2] \exp(i\Delta\phi_j(z,t)), \quad (5.8)$$

where the phase change for the j -th mode induced by SPM and DXPM is

$$\Delta\phi_j(z,t) = \frac{\omega_0 n_2}{c} \left(|C_j \Theta_j(z,t)|^2 + 2 \sum_{k \neq j} |C_k \Theta_k(z,t)|^2 \right) z. \quad (5.9)$$

The total electric field then can be written as

$$E(z,t) = \frac{1}{2} \left(\sum_j C_j \exp[-(t - z/v_{gj})^2/T^2] \exp[i\Delta\phi(z,t)] + \text{c.c.} \right) \quad (5.10)$$

The time derivative of the induced phase change $\Delta\phi_j(z,t)$ yields the spectral broadening for the j -th mode. The spectral distribution for the total pulse can be obtained by the Fourier transform of the electric field (5.10)

$$E(z,\omega) = \frac{1}{2} \int \sum_j C_j \exp[-(t - z/v_{gj})^2/T^2] \exp[i\Delta\phi(z,t)] \exp[i\omega t] dt + \text{c.c.} \quad (5.11)$$

5.2 Experiment on Degenerate-Cross-Phase-Modulation in Multimode Optical Fibers

5.2.1 Sample Preparation

Multimode silica-glass optical fibers with core diameters of 100 μm and numerical aperture of 0.2 were obtained from Spectran Corporation. For comparison, single mode glass fiber was also used. The single mode glass fiber operating in the visible region with a core diameter of 2.5 μm and a cladding diameter of 79 μm was manufactured by the Corning Glass Company. The cutoff wavelength of the fiber was 462 nm, and the refractive index difference between the core and the cladding was 0.24%.

The plastic protection jacket of a fiber was removed by emerge the ends of the fiber into methylene. After taking off the protection jacket, the fiber was carefully cut. The cutting quality was checked with a 300 $\times$ microscope. If the cutting was good then the fiber was used in the experiment. After performing the experiment, the length of the fiber was measured. The lengths of the fibers used in the experiment varied from 20 to 100 cm.

5.2.2 Experimental Setup

The experimental setup for the measurement of the spectral broadening of DXPM in multi-mode optical fibers is shown in Fig. 5.1. The laser system used was the same as in Chapter 4. An 8-ps laser pulse at 527 nm with pulse energy of 2 mJ was selected from the laser system and then coupled into the optical fiber by a 10× microscope objective lens. The output signal pulse was collected with a 20× microscope objective lens and passed through a 0.25-m Jarrell-Ash spectrograph. An EG&G Princeton Applied Research OMA II system was connected to the spectrograph to measure the spectral distribution.

5.2.3 Experimental Results

The output spectra of the laser pulses propagating in multimode optical fibers of different lengths are displayed in Fig. 5.2. The spectrum of the incident laser pulse is shown in Fig. 5.2(a). The bandwidth of the wavelength centered at 527 nm is about 1.5 nm. The spectral broadening for multimode optical fibers shown in Figs. 5.2(b) - (d) increases with the fiber length. When the length of the fiber was longer than 50 cm, stimulated Raman scattering with $\Delta\omega_R \sim 450 \text{ cm}^{-1}$ was observed. This is shown in Fig. 5.2(d). In all cases, the energy of the incident laser pulse was held nearly constant. The salient features of the spectrum of the output laser pulse from multimode optical fibers are displayed in Fig. 5.2. The spectral broadening of the pulse in multimode optical fibers is asymmetric about the incident laser frequency. The spectral broadening is shifted much more to the Stokes side than to the anti-Stokes side. The peak wavelength of the output pulses is shifted to the anti-Stokes side.

For comparison, similar measurements of spectral broadening in single mode optical fibers under the same experimental conditions were measured and are displayed in Fig. 5.3. In single mode optical fibers, the spectral broadening was asymmetric to the anti-Stokes side. This is a typical feature of the SPM spectral broadening. The spectrum was

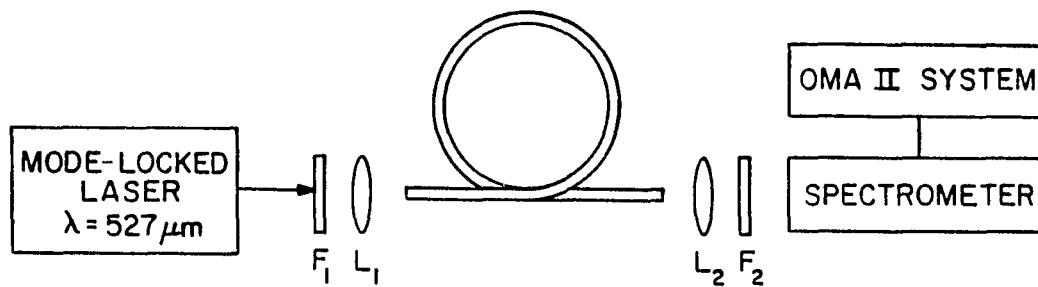


Figure 5.1. Experimental setup for spectral broadening of DXPM in multimode optical fibers. F_1 and F_2 are sets of color and neutral density filters; L_1 : 10× microscope objective lens, and L_2 : 20× microscope objective lens.

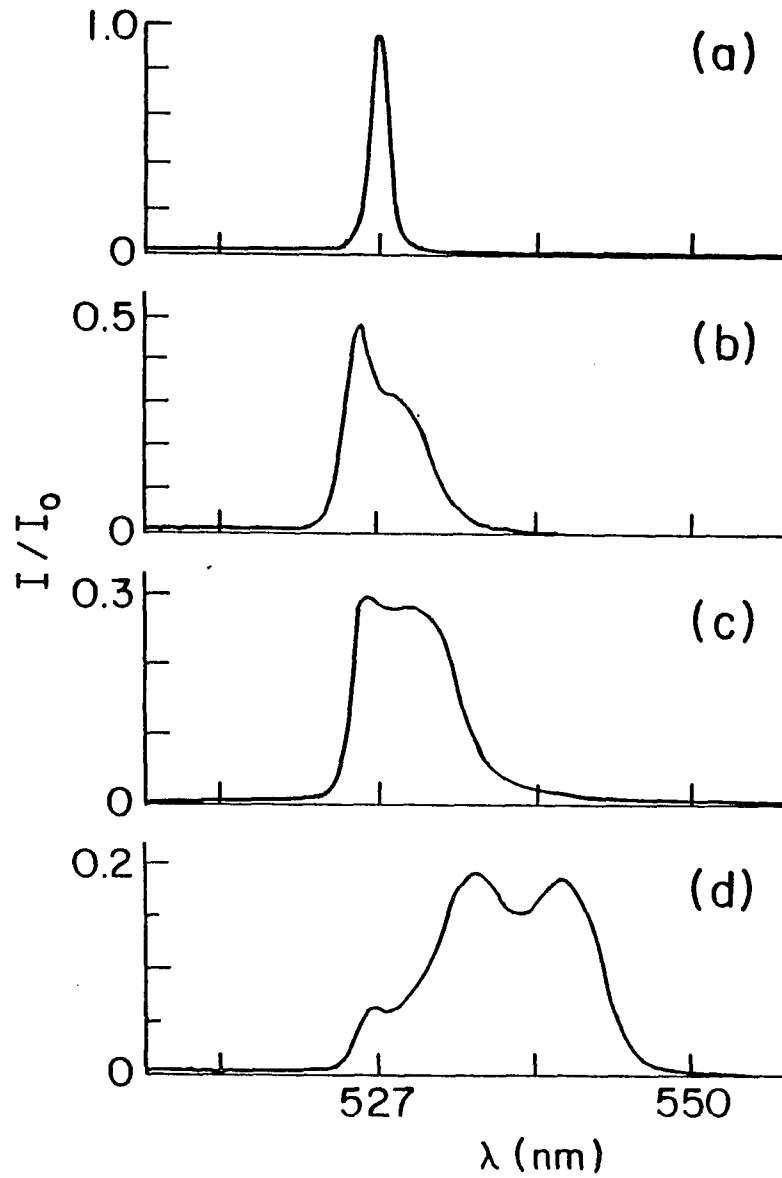


Figure 5.2. Output DXPM spectral for 8 picosecond laser pulse at 527 nm from multimode optical fibers with different lengths. The horizontal axis is wavelength and the vertical axis in normalized intensity. (a) No fiber; (b) 22cm; (c) 42 cm; (d) 84 cm.

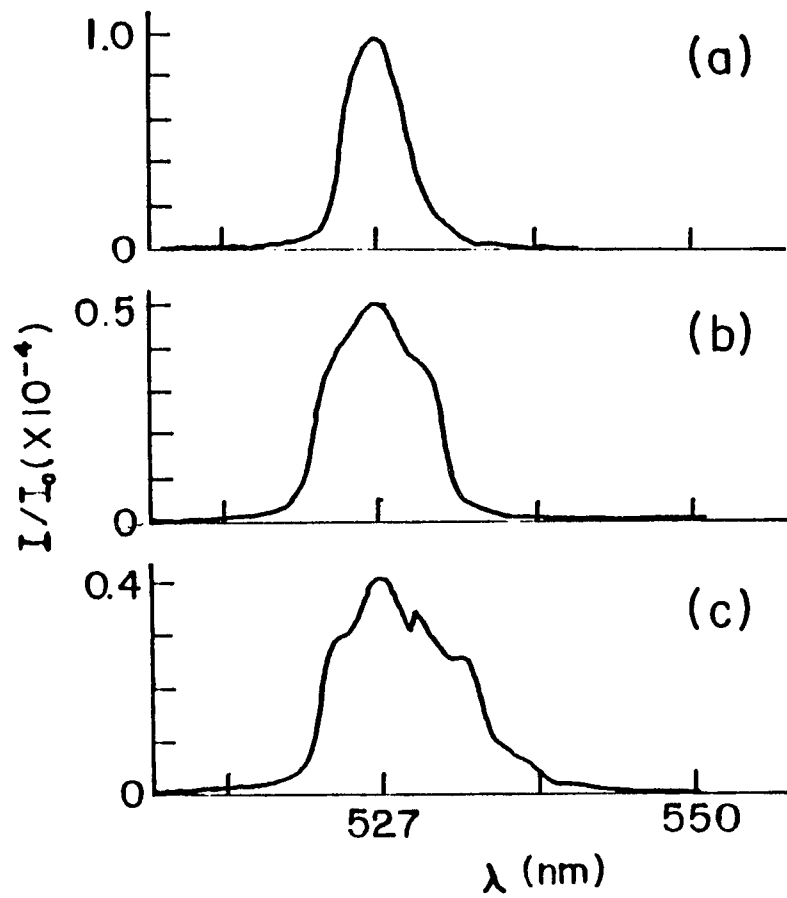


Figure 5.3. Output SPM spectral for 8 picosecond laser pulse at 527 nm from single mode optical fibers with different lengths. The horizontal axis is wavelength and the vertical axis is normalized intensity. (a) 20 cm; (b) 40 cm; (c) 50 cm.

dominated by SPM at low intensity ($< 10^7$ W/cm²) or when the fiber was short (< 20 cm). When the laser intensity was higher ($> 10^9$ W/cm²) or the length of the fiber was longer (> 50 cm), the Stokes spectra were much broader than the anti-Stokes spectra owing to the stimulated Raman scattering and XPM. In contrast, the spectral broadening for multimode optical fibers was asymmetrically shifted toward the Stokes side when the intensity was low ($< 10^7$ W/cm²) or the fiber was short (< 20 cm). In the case of high intensity ($> 10^9$ W/cm²) or a long fiber (> 60 cm), the stimulated Raman scattering builds up. At intermediate laser intensity, a spectral peak with a wavelength between the laser and the first stimulated Raman scattering often appears. In addition, the anti-Stokes spectral broadening of pulses propagating in multimode optical fibers were much wider than the Stokes broadening.

5.3 Discussion

The asymmetric spectral broadening of laser pulses propagating in multimode optical fibers was measured. To understand this behavior, the numerical simulation of the data is developed showing the asymmetrical spectral broadening.

Using Eqs. (5.10) and selected appropriate effective amplitudes of the different modes and the fiber lengths, three calculated output pulse spectra are plotted in Fig. 5.4. In the calculation, 16 modes, with effective amplitudes $C_1^2=1.00$, $C_2^2=0.80$, $C_3^2=0.65$, $C_4^2=0.50$, $C_5^2=0.40$, $C_6^2=0.30$, $C_7^2=0.25$, $C_8^2=0.20$, $C_9^2=0.15$, $C_{10}^2=0.12$, $C_{11}^2=0.10$, $C_{12}^2=0.08$, $C_{13}^2=0.06$, $C_{14}^2=0.04$, $C_{15}^2=0.02$, and $C_{16}^2=0.01$, were chosen for the best data fitting. The unit is 3.7×10^{10} (V/m)². $n_2=2.3 \times 10^{-22}$ (m/V)², $v_{g1}=3 \times 10^8/1.5$ m/sec, $v_{g16}=1.9600 \times 10^8/1.5$ m/sec, and $v_{gi}=v_{g1}-(i-1)(v_{g1}-v_{g16})/(16-1)$ or $i=2$ to 15 . The total energy is 2 μ J. The salient features of the calculated spectral distribution are that the broadening to the Stokes side is

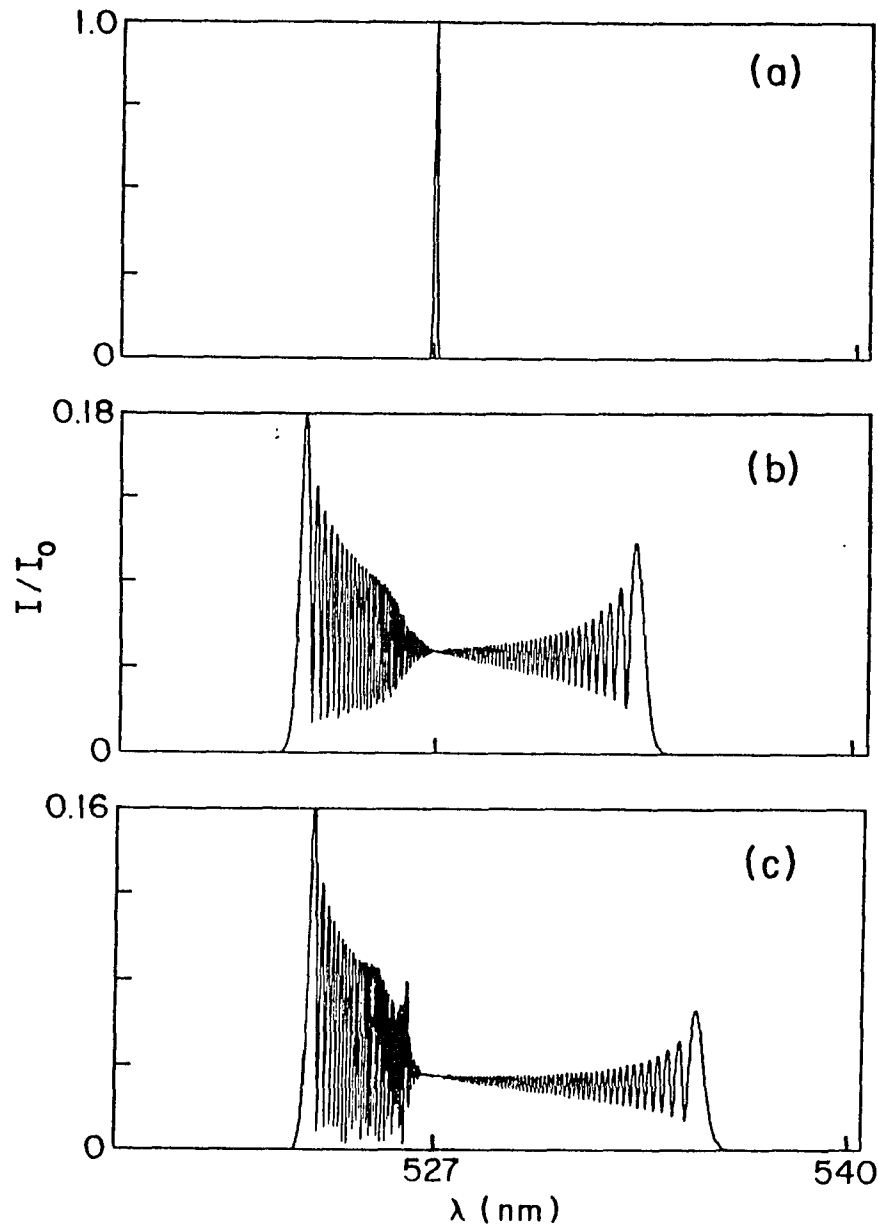


Figure 5.4. Calculated DXPM spectral broadening in multimode optical fibers as a function of fiber length for an 8-picosecond laser pulse at 527 nm. The horizontal axis is wavelength and the vertical axis in normalized intensity. (a) No fiber, (b) 20 cm, (c) 40 cm. In the calculation, 16 modes, with effective amplitudes , were chosen for the best data fitting. The unit is 3.78(a) No fiber, (b) 20 cm, (c) 40 cm.

wider than that to the anti-Stokes side and the peak frequency is shifted to the anti-Stokes side. The calculated values of the spectral broadening on both the Stokes and anti-Stokes sides as functions of the fiber length are plotted in Fig. 5.5 for comparison with the measured data. The calculated curves are in good agreement with the experimental data. Only at $L = 84$ cm is the measured spectrum much broader, this is caused by the additional Raman XPM, which is not considered in the calculation.

The observed spectra from multimode optical fibers in Fig. 5.2 did not show any of the modulation structure shown in the calculated spectra in Fig. 5.4. The lack of the modulation structures in the experimental spectra can be explained as follows. First, the calculated modulation structure has a period of 0.13 nm, which is below the spectral resolution of the measurement system of about 0.5 nm. The poor resolution of the measurement system may wash out the modulation structure. Secondly, The FWHM of the spectral the laser pulse was about 1 nm, which is much greater than the modulation period of the calculated spectra. Other difference between the calculated curves and the measured spectra need to be mentioned. The calculated spectra do not show a strong feature at the pumping laser line. The output wavelength peaks are located near the edge of the anti-Stokes side. The calculated peak shift is approximately six times greater than that of the measured spectra. These differences may be accounted for by the approximations in the calculations. First, in the calculations only sixteen modes were used to simplify the calculation. The characteristics of the fiber allow for more than 100,000 modes to be excited. Therefore, mixing in the spectra will wash out the modulation structure. Secondly, the pulse distortion of the pulse in each mode was not considered in the calculation. Thirdly, in the calculation Fourier transform limiting bandwidth was used, while the laser pulse was highly chirped.

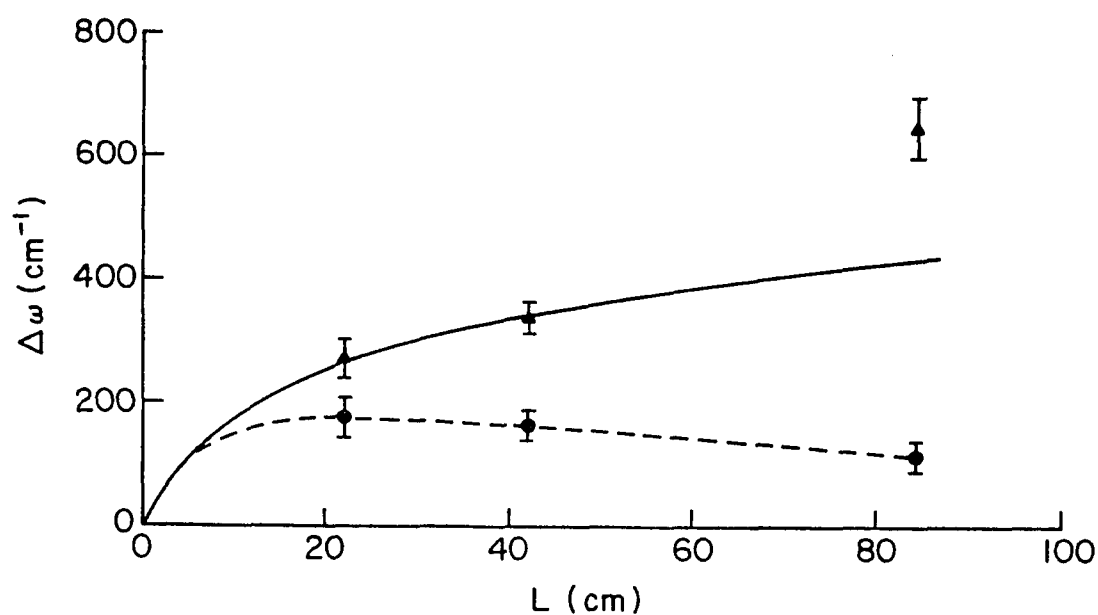


Figure 5.5. DXPM spectral broadening as a function of multimode optical fiber length (L). Δ : Stokes side; $\bullet$: anti-Stokes side. The solid and the dashed curves are the calculated spectral broadening to the Stokes and anti-Stokes sides, respectively, with respect to fiber length.

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Chapter 6

Degenerate Cross-Phase-Modulation and Pulse Compression

Degenerate cross-phase-modulation (DXPM)¹⁻⁴ is a new class of cross-phase-modulation (XPM)⁵⁻¹⁴ in which two or more laser pulses with the same frequency but different polarizations interact in condensed matter. Linearly polarized strong pump pulses induce an index change. The index change can be used to modulate the phase of a perpendicularly polarized weak probe pulse and change its spectral and temporal profile. By adjusting the intensity of the pump pulses and the delay between the pump and probe pulses, the spectral and temporal changes of the weak probe pulse can be controlled using DXPM.

This chapter presents both theoretical research that predicts the effects of DXPM on the spectral and temporal profiles of the pump and probe pulses for both single- and double pump pulses and the experimental measurements of the predicted effects. In Section 6.1, we compute the nonlinear polarizations for DXPM and apply to the nonlinear wave equation that was obtained in Chapter 3. Numerical solutions for the spectral and the temporal distributions of the pump and probe pulses are calculated. The influence of input intensity and initial time delay between the pulses are discussed. The effect of degenerate four-wave mixing (DFWM) on pulse amplification and compression are particularly discussed. In Section 6.2 we present experimental measurements of pulse compression and amplification in optical fibers. Finally, a discussion on the theory and the experiments of DXPM is given in Section 6.3.

6.1 Theory of Degenerate Cross-Phase-Modulation

6.1.1 Nonlinear Polarizations and Wave Equations of Degenerate Cross-Phase-Modulation

To simplify the problem of DXPM, the following assumptions are made: the pulse duration is much longer than the response time of the medium; and the frequency dependence of the nonlinear susceptibility is not taken into account. Considering only third-order nonlinearities, the nonlinear polarization is expressed as:

$$P_i^{NL}(\omega_4) = \sum_{jkl} \chi_{ijkl}^{(3)} (-\omega_4, \omega_1, \omega_2, \omega_3) E_j(\omega_1) E_k(\omega_2) E_l(\omega_3), \quad (6.1)$$

where $\omega_4 = \omega_1 + \omega_2 + \omega_3$ by the conservation of energy, $\chi^{(3)}$ has 81 elements in general. In isotropic media there are 21 non-zero elements, of which four can be different and only three are independent.¹⁵ In the quasi-monochromatic approximation, the total electric field of two perpendicular linearly polarized optical pulses can be written in the form of :

$$\mathbf{E} = \frac{1}{2} \left[\hat{x} A_x(z, t-t_d) \exp[-i\omega_0 t_d + i(\omega_0 t - k_x z)] + \hat{y} A_y(z, t) \exp[i(\omega_0 t - k_y z)] \right] + \text{c.c.}, \quad (6.2)$$

where t_d is the time delay between the pump pulse polarized along the x axis and the probe pulse polarized along the y axis. Substituting Eq. (6.2) into Eq. (6.1) and keeping only the terms synchronized with ω_0 , one obtains

$$P_x^{NL} = \frac{1}{8} \chi_{1111}^{(3)} \left[3|A_x|^2 A_x + 2|A_y|^2 A_x + A_y^2 A_x^* \exp(i2\omega_0 t_d - i2\Delta k z) \right] \exp[i(\omega_0 t - k_x z) - i\omega_0 t_d], \quad (6.3a)$$

$$P_y^{NL} = \frac{1}{8} \chi_{1111}^{(3)} \left[3|A_y|^2 A_y + 2|A_x|^2 A_y + A_x^2 A_y^* \exp(-i2\omega_0 t_d + i2\Delta k z) \right] \exp[i(\omega_0 t - k_y z)], \quad (6.3b)$$

where $\Delta k = k_x - k_y$ is the propagation constant mismatch between the two orthogonally polarized waves, and $\exp(\pm i\omega_0 t_d)$ is the delay phase factor. The first term on the right hand side in Eqs. (6.3a) and (6.3b) is the SPM term, the second term is the XPM term

and the last term arises from the degenerate four-wave-mixing (DFWM). Their contributions to the XPM only appears when the two orthogonally polarized waves have the same frequency because of the degenerate nature of the polarization components.

Applying Eqs. (6.3a) - (6.3b) to Eq. (3.22), we obtain the coupled nonlinear wave equations for DXPM:

$$\frac{\partial A_x}{\partial z} + \frac{1}{v_{gx}} \frac{\partial A_x}{\partial t} + i \frac{1}{2} k^{(2)} \frac{\partial^2 A_x}{\partial t^2} = i \frac{\omega_0 n_2}{c} \left[(|A_x|^2 + \frac{2}{3} |A_y|^2) A_x + \frac{1}{3} A_y^2 A_x^* \exp(i2\omega_0 t_d - i2\Delta k z) \right], \quad (6.4a)$$

$$\frac{\partial A_y}{\partial z} + \frac{1}{v_{gy}} \frac{\partial A_y}{\partial t} + i \frac{1}{2} k^{(2)} \frac{\partial^2 A_y}{\partial t^2} = i \frac{\omega_0 n_2}{c} \left[(|A_y|^2 + \frac{2}{3} |A_x|^2) A_y + \frac{1}{3} A_x^2 A_y^* \exp(-i2\omega_0 t_d + i2\Delta k z) \right], \quad (6.4b)$$

where v_{gx} and v_{gy} are the group velocities for the pulses polarized along the x axis and y-axis, respectively, $k^{(2)}$ is the group velocity dispersion, $n_2 = 3\pi\chi_{1111}^{(3)}/8n$ is the nonlinear refractive index coefficient,

The first terms in the right hand sides of Eqs. (6.4a) and (6.4b) are the self-phase-modulation (SPM) terms, the second terms are the XPM terms, and the last terms are the DFWM terms.

The influence of the DFWM to the XPM process depends on the extent of the phase matching of the two orthogonally polarized pulses characterized by the beat length $L_B = \frac{2\pi}{k_x - k_y}$. If the length of the medium $L \gg L_B$, the contributions of the last term in Eqs. (6.4a) and (6.4b) can be neglected. In this case the DXPM between the two polarization components is similar to the conventional XPM with two pulses with the same polarization but different frequencies. The coupling term for the DXPM is weaker than that of XPM since the coefficient for the cross term is 2/3 for DXPM instead of 2 for XPM.¹⁻⁶ In some cases, such as in nonbirefringent media or with the use of modulation instability¹⁰ to compensate for the birefringence in a medium, Δk can be zero. Under this

condition ($\Delta k=0$), the contributions from the DFWM term to the amplitude and phase modulations must be taken into consideration.

6.1.2 Spectral Broadening of Degenerate Cross-Phase-Modulation

Numerical methods are used to solve Eqs. (6.4a) and (6.4b) for general cases. However, analytical solutions can be obtained when the group velocity dispersion and temporal broadening are neglected.

For large birefringence, the DFWM terms have large phase mismatch and can be neglected. The last term is set to zero since its contribution washes out. Then the nonlinear coupled wave equations reduce to:

$$\frac{\partial A_x}{\partial z} + \frac{1}{v_g} \frac{\partial A_x}{\partial t} = i \frac{\omega_0 n_2}{c} (|A_x|^2 + \frac{2}{3} |A_y|^2) A_x, \quad (6.5a)$$

$$\frac{\partial A_y}{\partial z} + \frac{1}{v_g} \frac{\partial A_y}{\partial t} = i \frac{\omega_0 n_2}{c} (|A_y|^2 + \frac{2}{3} |A_x|^2) A_y, \quad (6.5b)$$

where $v_g = v_{gx} = v_{gy}$ has been assumed. Changing variables

$$\tau = t - \frac{z}{v_g}, \quad (6.6a)$$

$$z = z \quad (6.6b)$$

and denoting by a_j and α_j the amplitude and phase of the electric envelope, respectively,

A_j can be written as

$$A_j(z, \tau) = a_j \exp[i\alpha_j(z, \tau)], \quad (6.7)$$

where τ is the local time and $j = x, y$. Eqs. (6.5a) and (6.5b) reduce to

$$\frac{\partial a_x}{\partial z} = 0, \quad (6.8a)$$

$$\frac{\partial \alpha_x}{\partial z} = \frac{\omega_0 n_2}{c} \left[a_x^2 + \frac{2}{3} a_y^2 \right], \quad (6.8b)$$

$$\frac{\partial a_y}{\partial z} = 0,$$

$$(6.8c)$$

$$\frac{\partial \alpha_y}{\partial z} = \frac{\omega_0 n_2}{c} \left[a_y^2 + \frac{2}{3} a_x^2 \right]. \quad (6.8d)$$

The analytical solutions for Eqs.(6.8a) - (6.8d) can be obtained as

$$A_x(z,t) = a_x(z=0,t)\exp(i\alpha_x) , \quad (6.9a)$$

$$A_y(z,t) = a_y(z=0,t)\exp(i\alpha_y) , \quad (6.9b)$$

with

$$\alpha_x = \frac{\omega_0 n_2}{c} \left(a_x^2 + \frac{2}{3} a_y^2 \right) z , \quad (6.9c)$$

$$\alpha_y = \frac{\omega_0 n_2}{c} \left(a_y^2 + \frac{2}{3} a_x^2 \right) z . \quad (6.9d)$$

These results are similar to those obtained from conventional XPM. The difference is that the coefficient in the coupling term of DXPM is 2/3 instead of 2 for XPM. The amplitude shapes of the pump and probe pulses remain the same as their input values.

The instantaneous DXPM induced frequency chirp can be obtained by differentiating the phases α_x and α_y with respect to time τ . The envelope of the Gaussian pulses at $z=0$ are given by

$$A_x(z=0,t) = \sqrt{\frac{P_x}{A_{\text{eff}}}} \exp\left(-\frac{(\tau - \tau_d)^2}{2T_0^2}\right) , \quad (6.10a)$$

$$A_y(z=0,t) = \sqrt{\frac{P_y}{A_{\text{eff}}}} \exp\left(-\frac{\tau^2}{2T_0^2}\right) , \quad (6.10b)$$

where P_j are the peak power of the pulses, A_{eff} is the effective cross section area, τ_d is the local time delay between the pulses, and T_0 is the 1/e pulse duration.

The instantaneous DXPM induced frequency chirp are:

$$\Delta\omega_x = \frac{2\omega_0 n_2}{cA_{\text{eff}}} \left(P_x \frac{(\tau - \tau_d)}{T_0} \exp\left(-\frac{(\tau - \tau_d)^2}{2T_0^2}\right) + P_y \frac{\tau}{T_0} \exp\left(-\frac{\tau^2}{2T_0^2}\right) \right) z , \quad (6.11a)$$

$$\Delta\omega_y = \frac{2\omega_0 n_2}{cA_{\text{eff}}} \left(P_y \frac{\tau}{T_0} \exp\left(-\frac{\tau^2}{2T_0^2}\right) + P_x \frac{(\tau - \tau_d)}{T_0} \exp\left(-\frac{(\tau - \tau_d)^2}{2T_0^2}\right) \right) z , \quad (6.11b)$$

where $\Delta\omega_j = \omega_j - \omega_0$. The first and second terms on the right hand side of Eqs. (6.11a) and (6.11b) are contributions arising from SPM and XPM, respectively. Eqs. (6.11a) and (6.11b) indicate that the frequency extents for both pulses are inversely proportional to the pulse duration; the spectral broadening is proportional to the nonlinear index coefficient n_2 ; the spectral broadening is proportional to the interaction length z ; and the broadening is proportional to the input power of the pulses.

If the power of the probe pulse P_y is much smaller than that of the pump pulse P_x , the XPM term in Eq. (6.11a) and the SPM term in Eq. (6.11b) can be neglected. If we assume that there is no time delay between the pump and probe pulses, the probe pulse has the same spectral broadening as the pump pulse: the leading part is red shifted and the trailing part is blue shifted. If we assume that the pump pulse is leading the probe pulse by about the amount of the pulse duration, τ_d is a negative quantity, then the leading part of the probe pulse meets the trailing part of the pump pulse and is modulated by the index change induced by the trailing part of the pump pulse. The modulation produces a blue shift in the frequency of the leading part of the probe pulse. Since the trailing part of the probe pulse does not see much of the induced index change, the frequency of that part almost remain unchanged. If the probe pulse is leading the pump pulse, its trailing part is red shifted by the modulation of the induced index of refraction by the leading part of the pump pulse. If the two pump pulses are properly separated and the probe pulse is located between them, the spectral broadening effect is a combination of the above two cases: the leading part of the probe pulse is blue shifted by the modulation of the induced index of refraction by the trailing part of the first pump pulse, while the trailing part of the probe pulse is red shifted by the modulation of the induced index of refraction by the leading part of the second pump pulse. This effect is particularly important in DXPM pulse compression without using gratings or prisms. This effect will be discussed later.

Spectral profiles affected by DXPM can be obtained by computing the Fourier transform of the temporal pulse distribution as:

$$S_j(z, \omega - \omega_0) = \frac{1}{2\pi} \int a_j(\tau, z) \exp[i(\alpha_j(\tau, z))] \exp[(i(\omega - \omega_0)\tau)] d\tau, \quad (6.12)$$

where $j=x, y$, and $|S_j(z, \omega - \omega_0)|^2$ is the spectral intensity distribution of the pulses.

The numerical solutions for the change of spectra of DXPM is displayed in Fig. 6.1 when the group velocity is considered. The initial pulses are Gaussian. The input peak power of the pump pulse is 1000 times that of the probe pulse. $N=(L_D/L_{NL})^{1/2}$, where

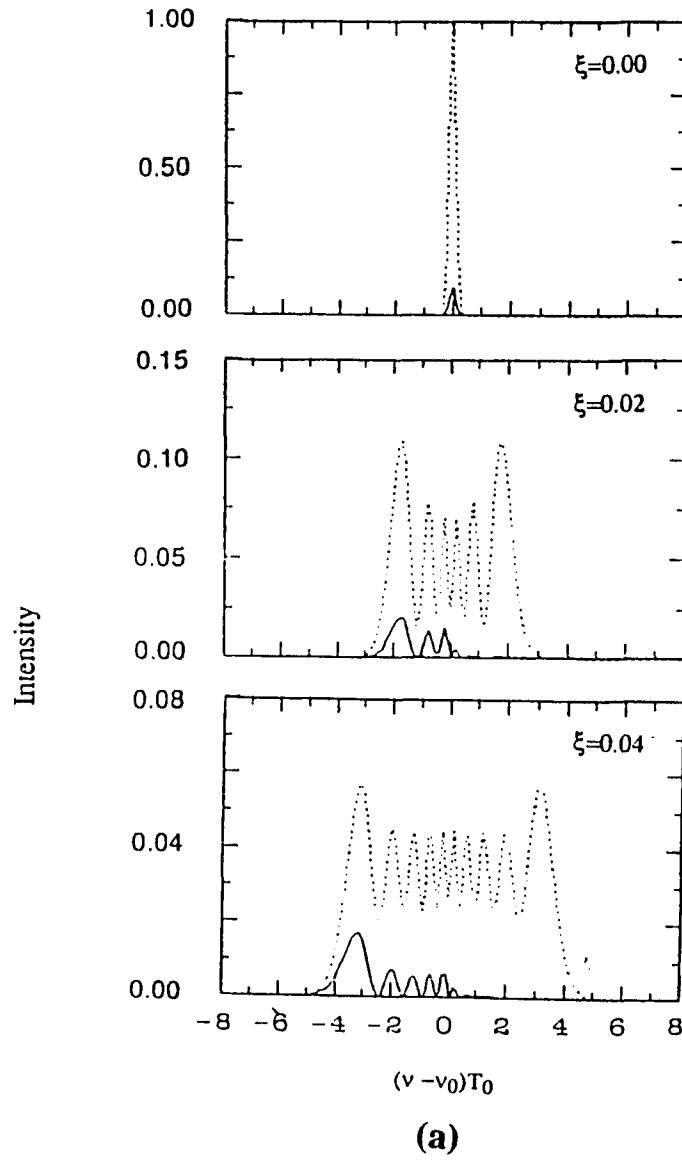


Figure 6.1. The spectral distribution of DXPM. Solid curves are the profiles of probe pulses enlarged by a factor of 100. Dotted curves are the spectral profiles of the pump pulses. $N=30$. (a) single pump pulse, $T_D=1$; (b) single pump pulse, $T_D=2$; and (c) double pump pulses, $T_D=2$. $N=30$.

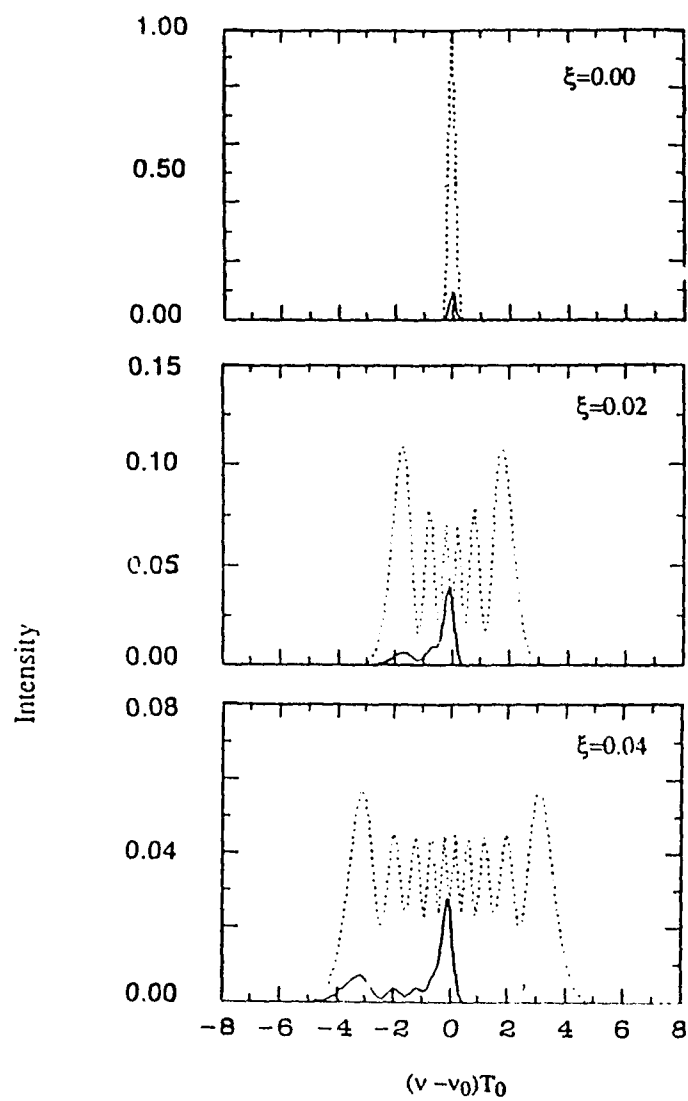
**(b)**

Figure 6.1 continued

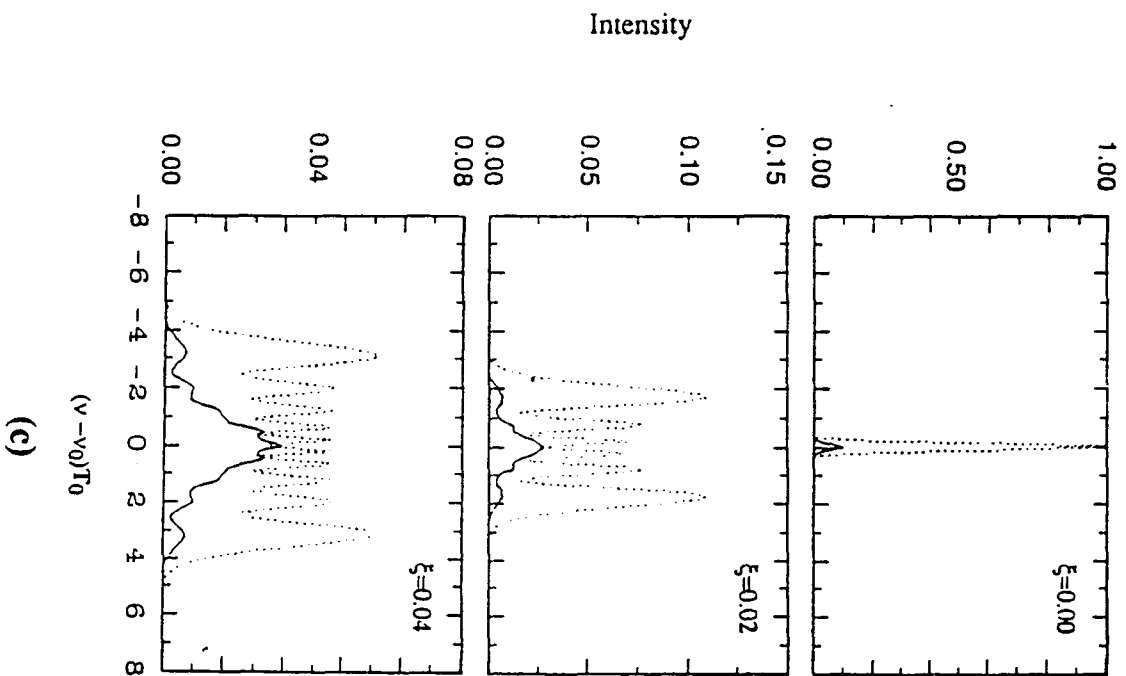


Figure 6.1 continued

$L_D = T_0^2 / |k^{(2)}|$ is the dispersion length, $L_{NL} = cA_{eff} / (\omega_0 n_2 P_0)$ is the nonlinear length, T_0 is the $1/e$ pulse duration, A_{eff} is the effective cross-section area, and P_0 is the input peak power of pump pulses. The dimensionless variables T and ξ are defined as $T = \tau / T_0$ and $\xi = z / L_D$. When the probe pulse is leading the pump pulse and the normalized delay $T_D = 1$, the induced chirp of the probe pulse is over the center part of the pulse. A large portion of the probe pulse is modulated by the leading part of the pump pulse and the frequency is chirped. There is a small peak at the input frequency ν_0 as shown in Fig. 6.1(a). When the normalized delay $T_D = 2$, the induced chirp of the probe pulse is over the trailing part and the rest part of the pulse remain unchanged. There is a large peak at the input frequency ν_0 as displayed in Fig. 6.1(b). In the case of double-pump pulse and the delay between the probe pulse and each pump pulse $T_D = 2$, the induced chirp of the probe pulse occurs at its both leading and trailing parts. The center part of the pulse remain unchanged. The spectrum distribution profile is symmetric and most energy of the pulse is near the input frequency as shown in Fig. 6.1(c). In all the cases, the modulation period of the probe pulse is the same as that of the pump pulse.

6.1.3 Pulse Compression and Amplification of Degenerate Cross-Phase-Modulation

When GVD effects are considered, $(\alpha_x - \alpha_y) \neq 0$ for general cases. The DFWM terms modulate the amplitudes as well as the phases. Since the derivatives of the amplitudes with respect to z vary sinusoidally, in certain regions one pulse is amplified while the other is attenuated. In other regions the situation is reversed. The total effect of the amplification and the attenuation is the result of the integration over the interaction length. The amplification and attenuation period of the amplitude depends on the phase difference between the pump and the probe pulses. The phase difference depends on the pulse amplitudes. Since the phases and the amplitudes of both the pump and probe pulses are

coupled together, it is extremely difficult to obtain an analytical solution for the nonlinear coupled

In a birefringent medium, the coupled nonlinear wave equations reduced to

$$\frac{\partial A_x}{\partial z} + \frac{1}{v_g} \frac{\partial A_x}{\partial t} + i \frac{1}{2} k^{(2)} \frac{\partial^2 A_x}{\partial t^2} = i \frac{\omega_0 n_2}{c} (|A_x|^2 + \frac{2}{3} |A_y|^2) A_x, \quad (6.13b)$$

$$\frac{\partial A_y}{\partial z} + \frac{1}{v_g} \frac{\partial A_y}{\partial t} + i \frac{1}{2} k^{(2)} \frac{\partial^2 A_y}{\partial t^2} = i \frac{\omega_0 n_2}{c} (|A_y|^2 + \frac{2}{3} |A_x|^2) A_y, \quad (6.13b)$$

where the two DFWM terms which, for large birefringence, have large phase mismatch and can be neglected; and $v_g = v_{gx} = v_{gy}$ has been assumed to simplify the calculations.

Now we introduce the dimensionless variables

$$T = \frac{\tau}{T_0}, \quad (6.14a)$$

$$\xi = \frac{z}{L_D}, \quad (6.14b)$$

$$U_j = \frac{A_j}{\sqrt{P_{\text{pump}}/A_{\text{eff}}}}, \quad (6.14c)$$

where

$$L_D = \frac{T_0^2}{|k^{(2)}|} \quad (6.15)$$

is the dispersion length, over which the pulse duration increases about 40% owing to the dispersion effect. The reduced coupled nonlinear wave Eqs. (6.13a) and (6.13b) reduce to

$$i \frac{\partial U_x}{\partial \xi} = \pm \frac{1}{2} \frac{\partial^2 U_x}{\partial T^2} - N^2 [(|U_x|^2 + \frac{2}{3} |U_y|^2) U_x] \quad (6.16a)$$

$$i \frac{\partial U_y}{\partial \xi} = \pm \frac{1}{2} \frac{\partial^2 U_y}{\partial T^2} - N^2 [(|U_y|^2 + \frac{2}{3} |U_x|^2) U_y] \quad (6.16b)$$

where

$$N^2 = \frac{L_D}{L_{\text{NL}}}, \quad (6.17)$$

with

$$L_{\text{NL}} = \left(\frac{\omega_0 n_2 P_{\text{pump}}}{c A_{\text{eff}}} \right)^{-1} \quad (6.18)$$

the nonlinear length. The choice of the sign in the dispersion term depends on the sign of $k^{(2)}$: a plus sign is for normal GVD ($k^{(2)} > 0$), and a minus sign is for anomalous GVD

($k^{(2)} < 0$). For example, $\lambda < 1300$ nm is the normal dispersion regime for silica fibers. At $\lambda = 530$ nm, the dispersion $k^{(2)} \approx 0.06$ ps²/m. From Eq. (6.15) $L_D \sim 100$ m for $T_0 \approx 3 - 5$ ps. Give a single mode fiber and $P_{\text{pump}} \sim 100$ W, from Eq.(6.18), $L_{\text{NL}} \sim 1$ m. Substituting these values into Eq. (6.17) one can find that parameter $N \sim 10$. $\lambda > 1300$ nm is the anomalous-dispersion regime for glass optical fibers. At $\lambda = 1550$ nm, dispersion $k^{(2)} \sim -0.02$ ps²/m. In this case, $L_D \sim 300$ m for $T_0 \approx 3 - 5$ ps. and $L_{\text{NL}} \sim 1$ m for $P_{\text{pump}} \sim 100$ W. Thus $N \sim 10$.

Fig. 6.2 shows a numerical solution of Eqs. (2.18a) and (2.18b) for an initial pulse shape $U_j(T,0) = \text{sech}(T)$ for both pump and probe pulses in the anomalous GVD regime when $N = 3$. In the calculation, the peak power of the probe pulse is chosen to be one percent of that of the pump pulse. There is no time delay between the pump and probe pulses. Both pulses are compressed. The compression effect is the same as the soliton-effect compression¹⁶ except that the frequency chirp of the weak probe pulse is induced by the strong pump pulse. It is obvious that to get the probe pulse compressed the initial delay between the two pulses must be zero.

Fig. 6.3 displays the numerical solution of Eqs. (2.18a) and (2.18b) for probe pulses (solid curves) under a single pump pulse in the normal dispersion regime with different time delays between the input pump and probe pulses. The initial pulse shapes are Gaussian for both pump and probe pulses. The physical mechanism behind the pulse compression is the pump-induced frequency chirp. For our cases, the induced chirp is nearly linear and negative over the leading part of the probe pulse. In the normal GVD regime, compression occurs in the leading part of the pulse, as can be seen in Fig. 6.3. The compression effect depends on the initial delay between the two pulses and the interaction length.

For the case when only one pump pulse is used, the compression occurs on one side of the probe pulse. The compressed pulse is asymmetric in time. To improve the compression distortion and the compression effect, one may use two pump pulses to

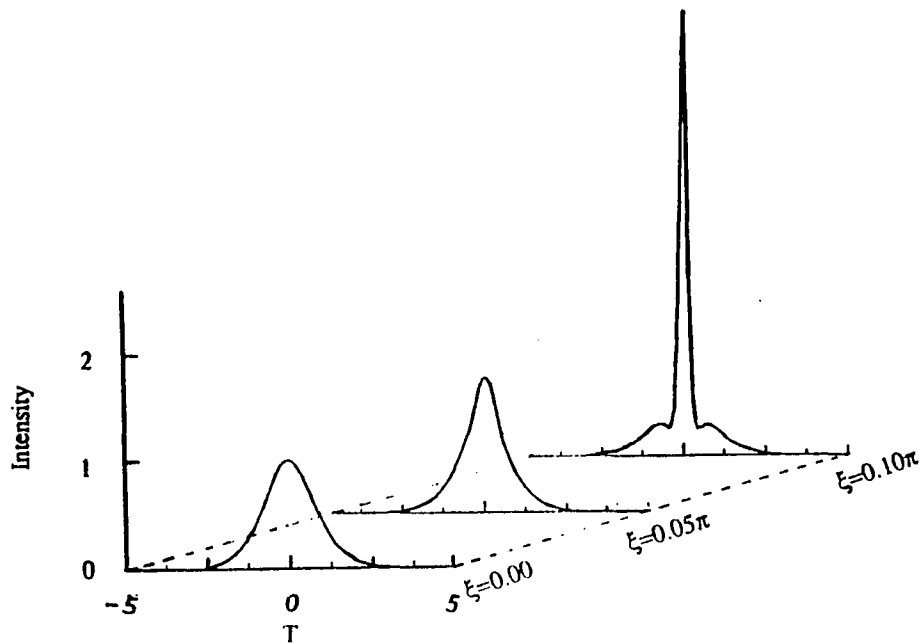


Figure 6.2. Changes in envelope shape of the pulse in time by DXPM in anomalous dispersion regime for $N=30$ and $T_D=0$ along the propagation length. The evolution of the pulse shape of the pump pulse is the same as that of the probe pulse.

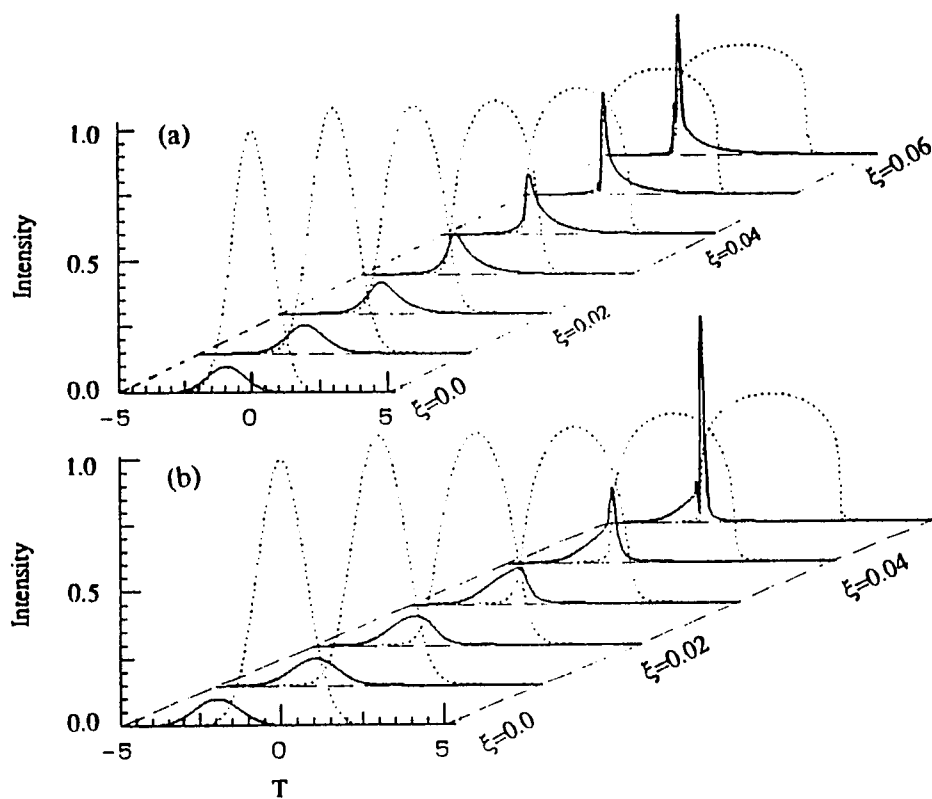


Figure 6.3. Changes in the envelope shape of the pulse in time by DXPM in the normal dispersion regime for a single pump pulse and $N=30$ along the propagation length. (a) $T_D=1$, (b) $T_D=2$. Solid curves show the evolution of the probe pulse shape enlarged by a factor of 100. Dotted curves are the changes of the pump pulse shape.

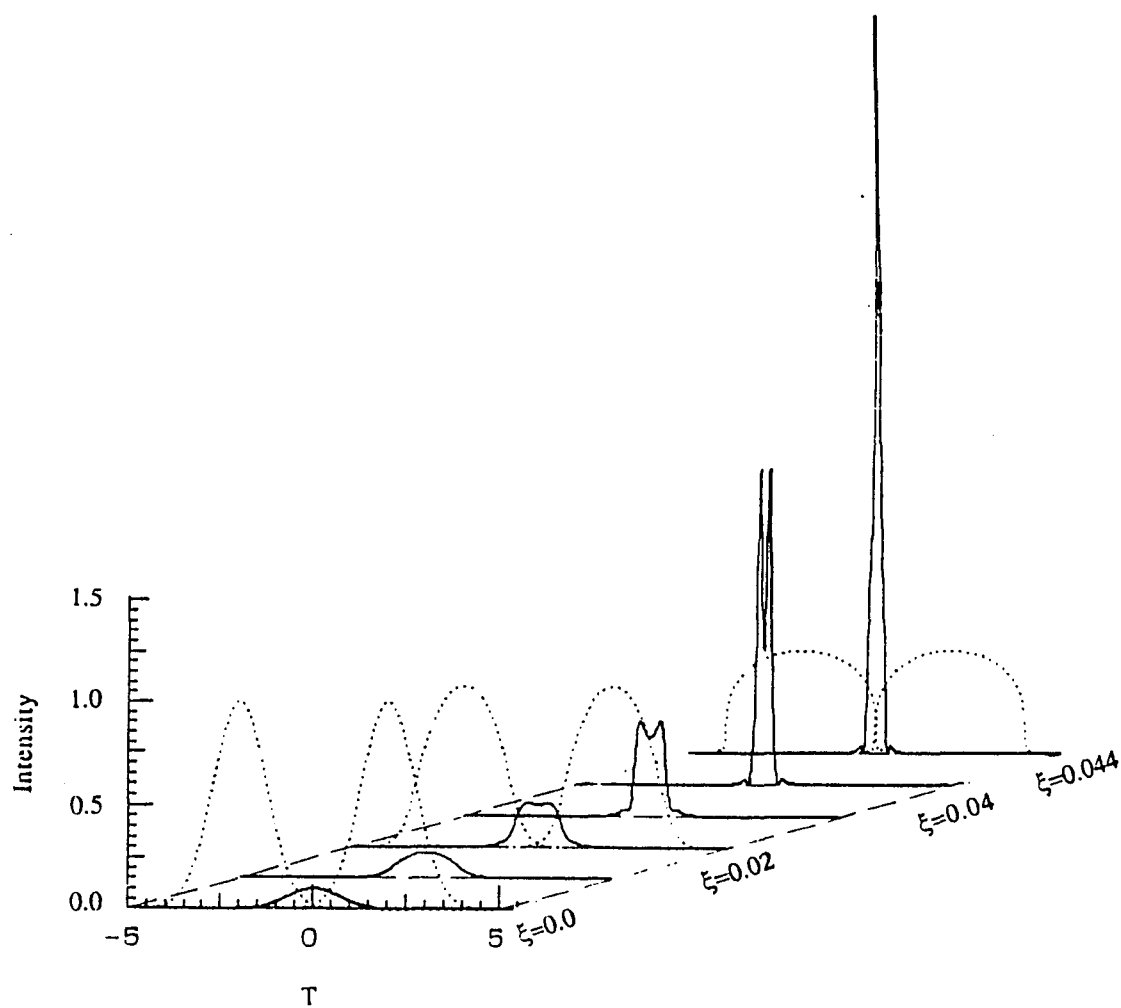


Figure 6.4. Changes in the envelope shape of the pulse in time by DXPM in the normal dispersion regime. The probe pulse shape is enlarged by a factor of 100. Dotted curves are the changes of the pump pulse shape.

compress a weak probe pulse propagating between them. The results of numerical simulation of a double-pump DXPM is displayed in Fig. 6.4. The compressed pulse is symmetric in time and the pulse duration can be reduced by a factor of 2.

6.1.4 Effect of Degenerate Four-Wave Mixing on Pulse Compression and Amplification in Degenerate Cross-Phase-Modulation Process

When phase matched degenerate four-wave mixing (DFWM) can exist ($\Delta k = 0$), the weak probe pulse can be amplified owing to DFWM. In addition, the DFWM can generate a chirp and increase XPM. The weak probe pulse can consequently be further compressed owing to the group-velocity dispersion.

To investigate the influence of DFWM on the phase modulation and amplitude modulation of the probe pulse induced by the pump pulse, one needs to solve the coupled-wave equations (6.4a) and (6.4b) with condition $\Delta k = 0$:

$$\frac{\partial A_x}{\partial z} + \frac{1}{v_{gx}} \frac{\partial A_x}{\partial t} + i \frac{1}{2} k^{(2)} \frac{\partial^2 A_x}{\partial t^2} = i \frac{\omega_0 n_2}{c} \left[(|A_x|^2 + \frac{2}{3} |A_y|^2) A_x + \frac{1}{3} A_y^2 A_x^* \exp(i2\omega_0 t_d) \right], \quad (6.19a)$$

$$\frac{\partial A_y}{\partial z} + \frac{1}{v_{gy}} \frac{\partial A_y}{\partial t} + i \frac{1}{2} k^{(2)} \frac{\partial^2 A_y}{\partial t^2} = i \frac{\omega_0 n_2}{c} \left[(|A_y|^2 + \frac{2}{3} |A_x|^2) A_y + \frac{1}{3} A_x^2 A_y^* \exp(-i2\omega_0 t_d) \right]. \quad (6.19b)$$

Using the same method that we used in the previous section, the coupled wave equations (6.19a) and (6.19b) can be reduced to

$$i \frac{\partial U_x}{\partial \xi} = \pm \frac{1}{2} \frac{\partial^2 U_x}{\partial T^2} - N^2 \left[(|U_x|^2 + \frac{2}{3} |U_y|^2) U_x + \frac{1}{3} U_y^2 U_x^* \exp(i\theta) \right], \quad (6.20a)$$

$$i \frac{\partial U_y}{\partial \xi} = \pm \frac{1}{2} \frac{\partial^2 U_y}{\partial T^2} - N^2 \left[(|U_y|^2 + \frac{2}{3} |U_x|^2) U_y + \frac{1}{3} U_x^2 U_y^* \exp(-i\theta) \right], \quad (6.20b)$$

where $\theta = \omega_0 t_d$ is the delay phase change.

Figs. 6.5 and 6.6 display the numerical solutions of Eqs. (6.20a) and (6.20b) with $\theta = n\pi$ for single and double pump pulses, respectively. The solid curves are the changes of the probe pulses. The dotted curves are the changes of the pump pulses. The initial

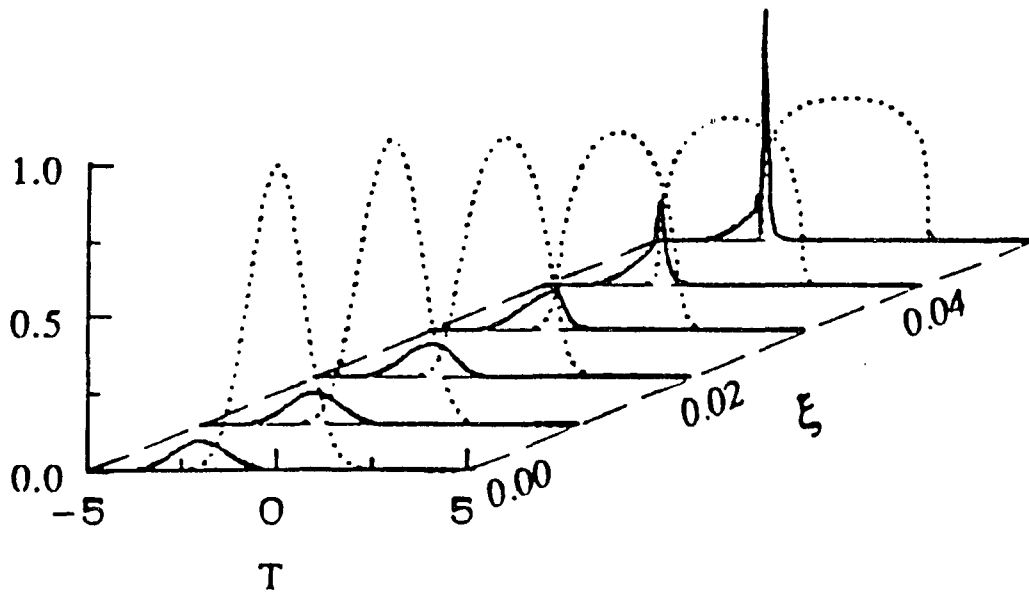


Figure 6.5. Changes in the envelope shapes of the probe and pump pulses in time in the normal dispersion regime under single pump pulse DXPM for $N=30$, $T_D=2$, $\Delta k=0$, and $\theta=n\pi$. The effect of phase matched DFWM is considered. The solid curves show the evolution of the probe pulse shape enlarged by a factor of 100. Dotted curves are the changes of pump pulse shape.

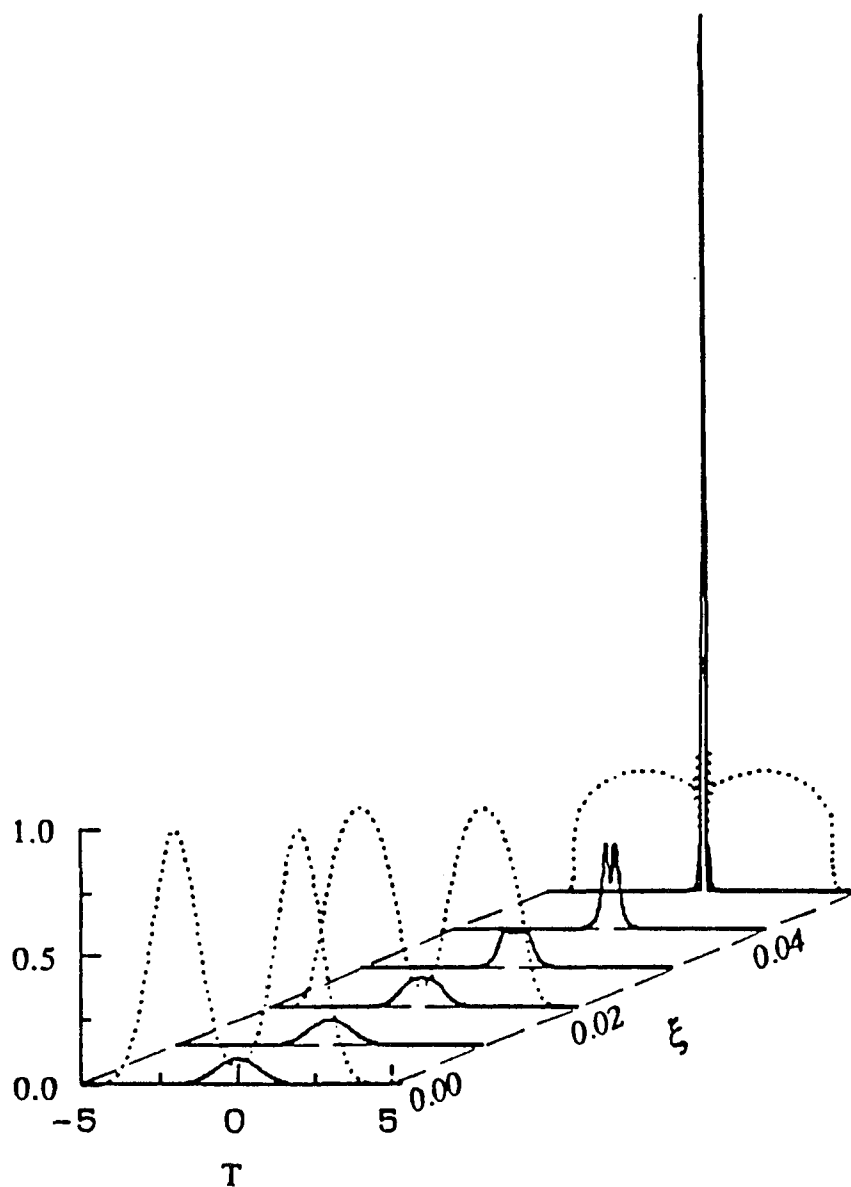


Figure 6.6. Changes in envelope shapes of the probe and pump pulses in time in the normal dispersion regime under double pump pulse DXPM for $N=30$, $T_D=2$, $\Delta k=0$, and $\theta=n\pi$. The effect of phase matched DFWM is considered. Solid curves show the evolution of the probe pulse shape enlarged by a factor of 100. Dotted curves are the changes of pump pulse shape.

pulse shapes are Gaussian for both the pump and probe pulses. The input peak power of the pump pulse is 1000 times that of the probe pulse. In the calculation $N = 30$ and $T_d = 2$ are used. The XPM of the probe pulse on the pump pulse is also taken into consideration. It can be seen that when the DFWM exists, the weak probe pulse can be amplified as well as compressed. The amplification through DFWM improves the effect of pulse compression. In the double pump-pulse compression scheme with a normalized time delay between the probe and the pump pulses of $T_d = 2$, the probe pulse is symmetric, and the pulse duration is reduced by a factor of 15 with an energy gain of 2.3 times. This result gives a power gain of 34 times.

Wave-breaking oscillations¹⁶⁻¹⁸ occur at both sides of the probe when two pump pulses are employed. Wave-breaking is manifested by the appearance of oscillations in the wings of the pulses. The oscillations are due to the interference between the shifted instantaneous frequency components induced by SPM and XPM and unshifted carrier component in the wings of the pulse. Figs. 6.7 show the expansions of the profiles of the pump and probe pulses at $\xi = 0.05$ of Figs. 6.4 and 6.6. When DFWM is not considered, the negative chirped trailing edge of the first pump pulse interferes with the positive chirped leading edge of the second pump pulse. This results in a large intensity oscillation in the region in which the two pump pulses overlap. The phase of the probe pulse is mainly modulated by the pump pulses through XPM since the intensities of the pump pulses are much larger than that of the probe pulse, and the combination of the GVD and XPM results in wave-breaking oscillations of the pump pulse with the same oscillation period as the pump pulses. Similar intensity oscillations of the pump pulses occur when the DFWM is taken into consideration. The probe pulse has a much narrower intense peak at the center and develops rapid oscillations in the wings as a result of XPM and SPM. The oscillation period of the probe pulse is the same as that of the oscillation formed in the overlap region of the pump pulses.

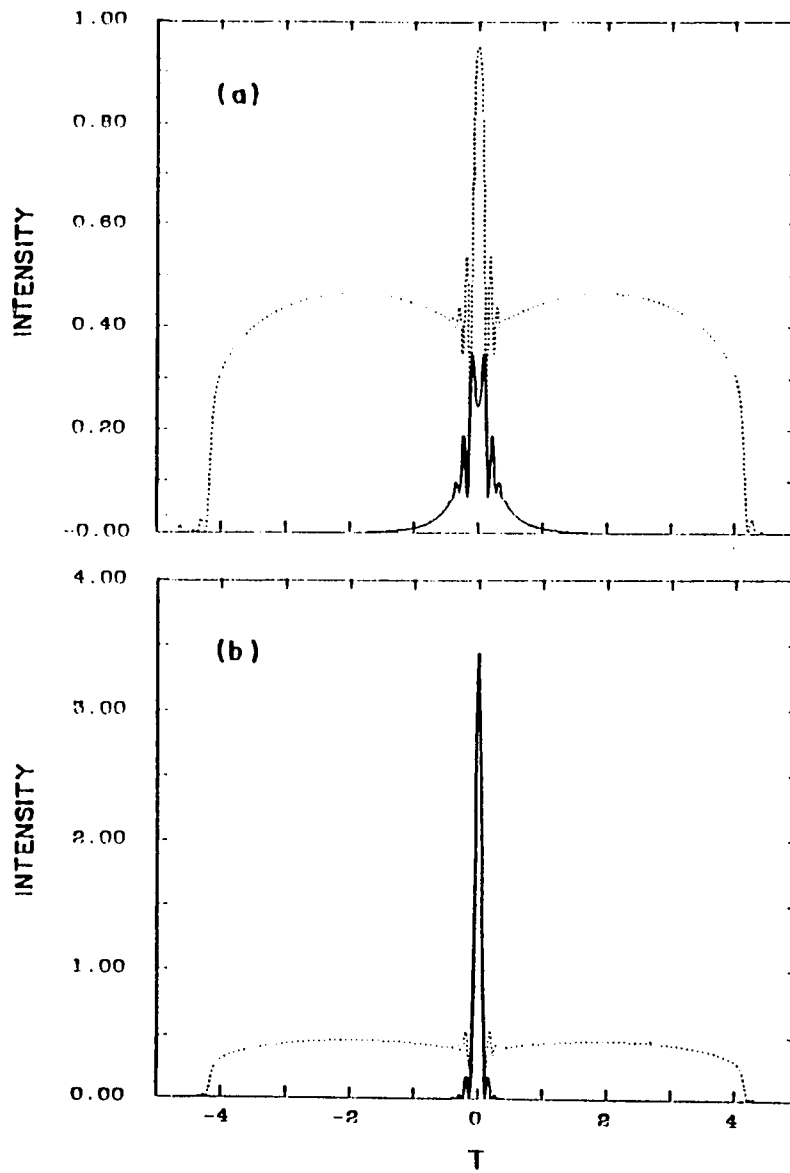


Figure 6.7. Expansion of pulse profiles at $\xi=0.05$ under double pump pulse DXPM. (a) Without DFWM, (b) with DFWM. The solid curves show the evolution of the probe pulse shape enlarged by a factor of 100. Dotted curves are the changes of pump pulse shapes.

6.1.5 The Influence of Intensity Ratio and Delay Time

The influences of the intensity ratio and the delay time between the pump and probe pulses are demonstrated in Figs. 6.8 and 6.9. Fig. 6.8 shows the changes of the envelopes of the pump and probe pulses with different intensity ratios. When the intensities of the pump and probe pulses are the same, the SPM and XPM of both pulses have the same strengths. The blue shift of the frequency caused by the trailing edge of the first pump pulse is compensated by the red shift induced by the leading edge of the probe pulse. At the center of the overlapping part of the trailing edge of the first pump pulse and the leading edge of the probe pulse, the frequencies remain unshifted. The frequency of the probe pulse earlier than the point is blue shifted induced by the first pump pulse. The frequency of the probe pulse between this point and its peak is red shifted due to SPM. The interference of the instantaneous frequency components in the overlap region resulting in large intensity oscillations. In the overlap region of the trailing edge of the probe pulse and the leading edge of the pump pulse, intensity oscillations are induced by a similar interference. This effect causes strong pulse distortion as shown in Fig. 6.8(a). When the intensity ratio of the pump and probe pulses is greater than 10, the SPM and XPM induced by the pump pulses dominate the frequency-shifting process. The leading edge of the probe pulse is blue shifted and its trailing edge is red shifted. In normal GVD region, the blue shifted leading edge slows down and the red shifted edge speeds up and resulting pulse compression of the probe pulse as shown in 6.8(c) - (f).

Fig. 6.9 displays the envelope shape change of pump and probe pulses in time for $T_d=1.7$ and $T_d=2.3$ and $\theta=n\pi$. The change of the time delay between pump and probe pulses changes the pulse compression. When $T_d=1.7$, the pulse duration of the probe pulse reduces to 1/15 of its input value at $\xi=0.05$. While pulse duration reduces by a factor of 15 at $\xi=0.06$ when $T_d=2.3$. This feature indicates that when the interaction length is given, the compression of the probe pulse can be adjusted by changing the delay between the pump and probe pulses.

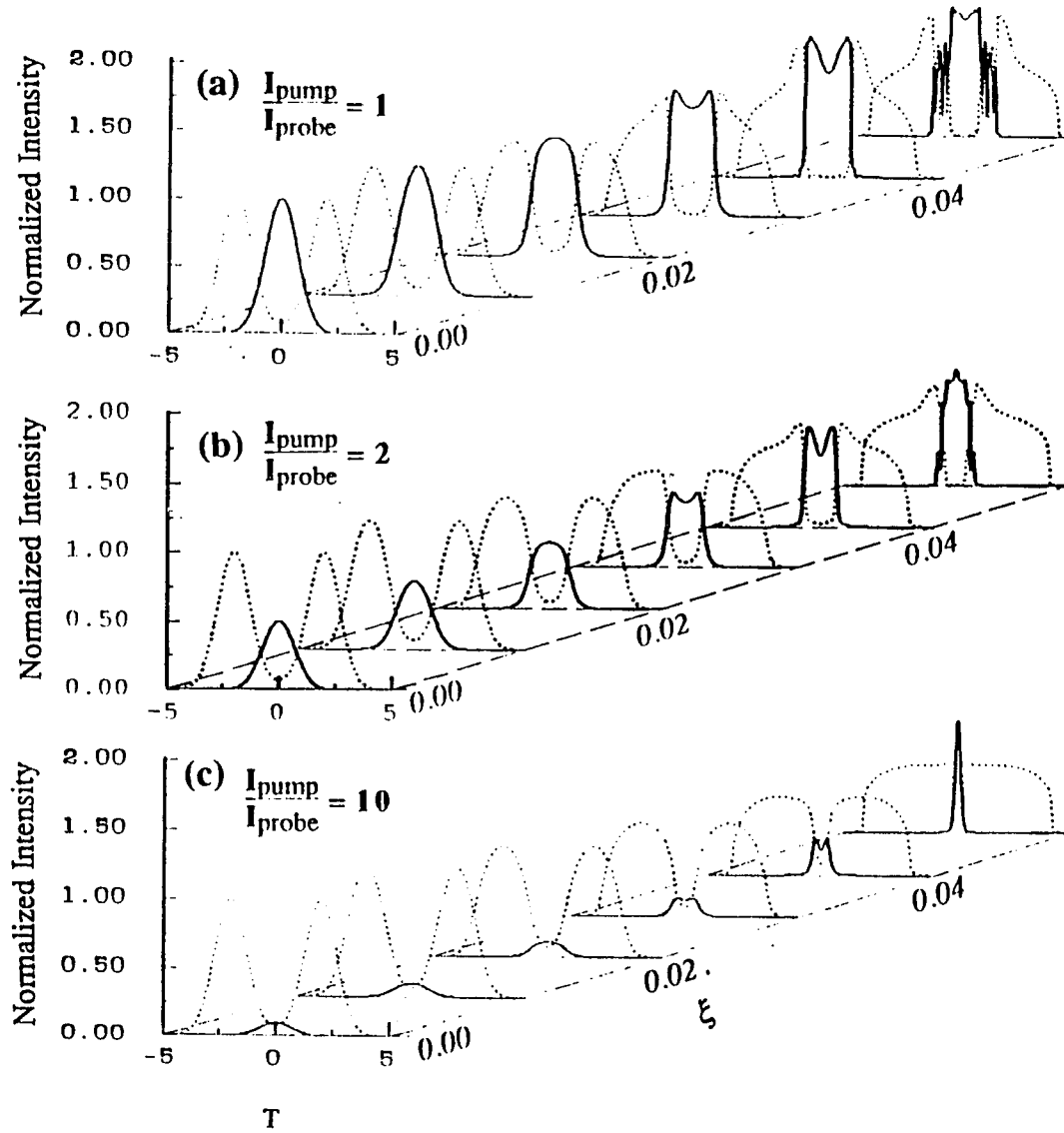


Figure 6.8. Changes in the envelope shapes of the probe and pump pulses in time in the normal dispersion regime under double pump pulse DXPM as a function of intensity ratio of the pump to probe pulses. $N=30$, $T_D=2$, $\Delta k=0$, and $\theta=n\pi$. The effect of phase matched DFWM is considered. The dotted curves show the changes of pump pulse shape. The solid curves show the evolution of the probe pulse shape enlarged by a factor of: (a) 1, (b) (1), (c) 1, (d) 3, (e) 10, and (f) 100.

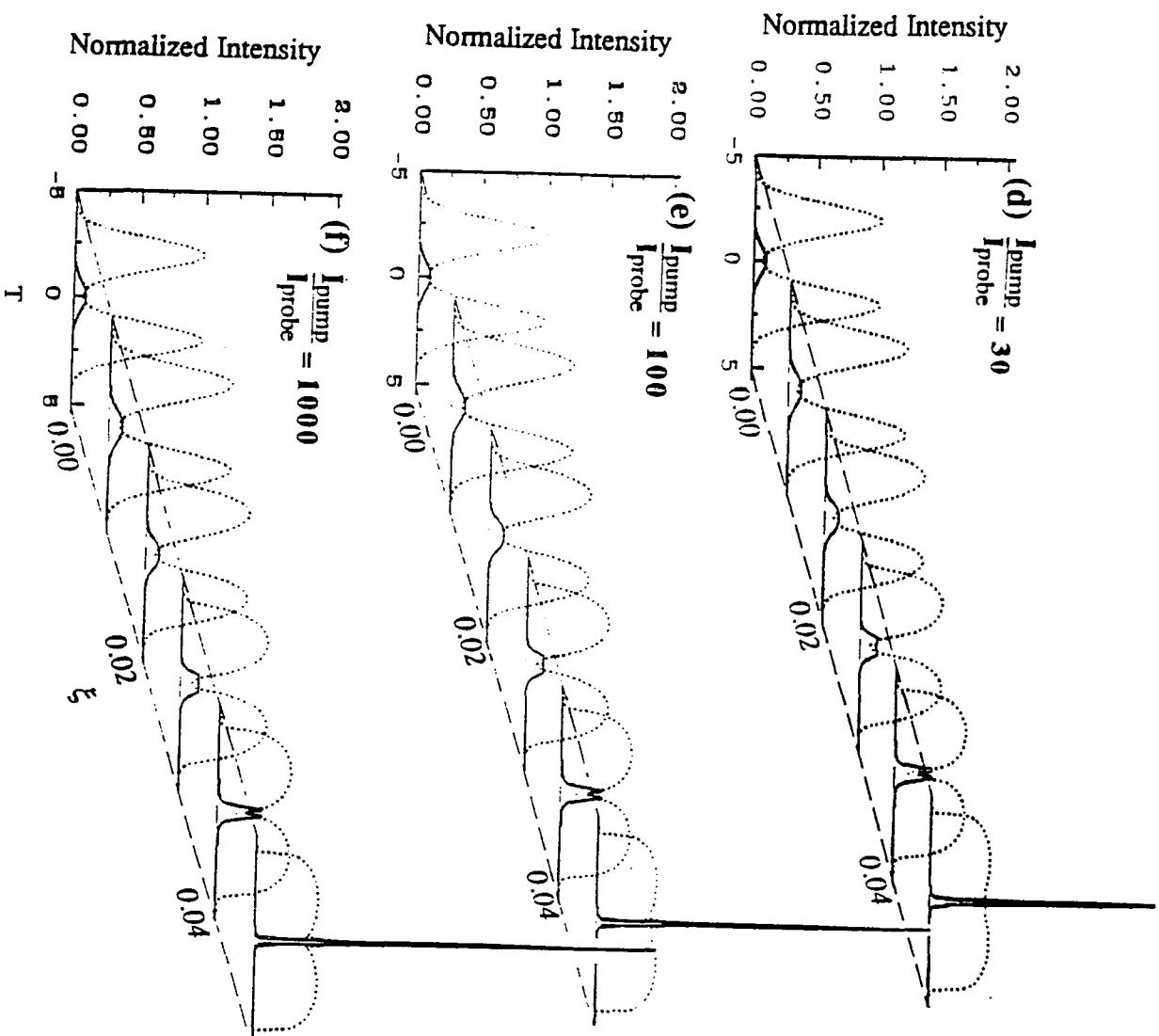


Figure 6.8 continued.

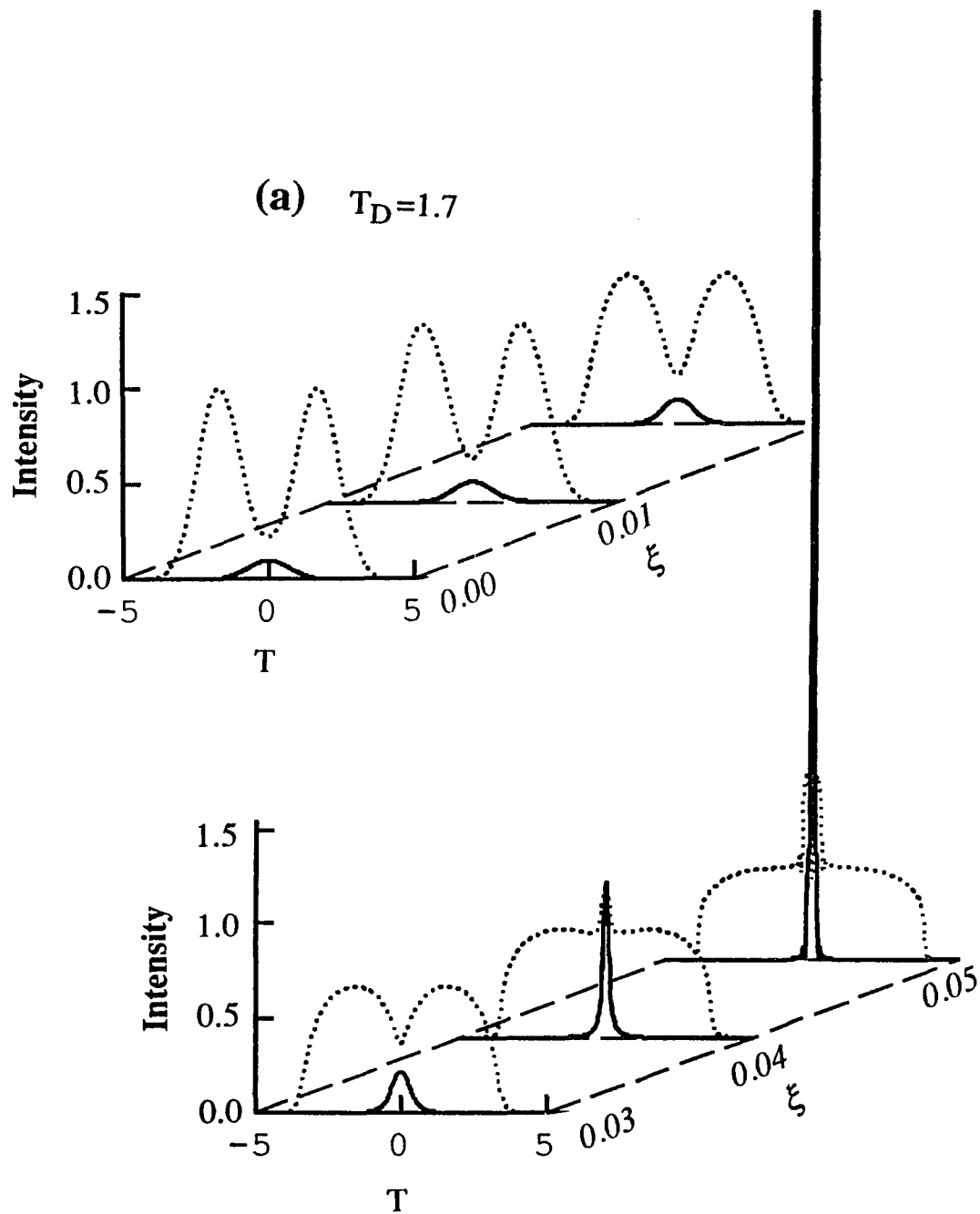


Figure 6.9. Changes in the envelope shapes of the probe and pump pulses in time in the normal dispersion regime under double pump pulse DXPM as a function of initial time delay. (a) $T_D=1.7$, (b) $T_D=2.3$. $N=30$, $T_D=2$, $\Delta k=0$, and $\theta=n\pi$. Dotted curves are the changes of pump pulse shape. The solid curves show the evolution of the probe pulse shape enlarged by a factor of 100.

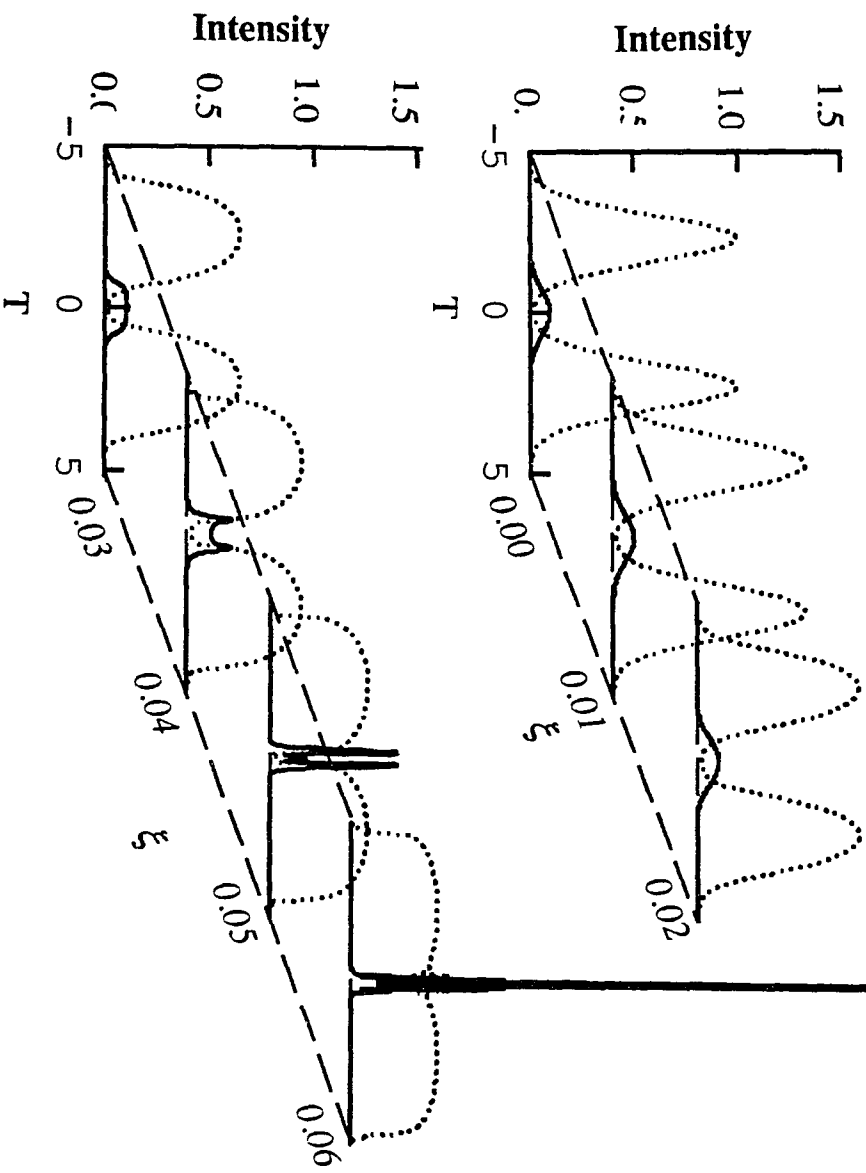
(b) $T_D = 2.3$ 

Figure 6.9 continued.

6.2 Degenerate Cross-Phase-Modulation Experiments

6.2.1 Sample Preparation

In the experiment both normal optical fibers and birefringent optical fibers are used. The single mode glass fiber (F-SA) operating in the visible region with a core diameter of $3.2\text{ }\mu\text{m}$ and cladding diameter of $125\text{ }\mu\text{m}$ was purchased from Newport Corporation. The numerical aperture of the fiber was 0.11. The group velocity dispersion at 550 nm was 425 ps/km·nm. The polarization preserving single mode optical fiber (F-SPA) was also bought from Newport Corporation. The core diameter and cladding diameters were 1.5 and $125\text{ }\mu\text{m}$, respectively. The numerical aperture of the fiber was 0.1. The beat length is less than 5 mm.

The plastic protection jacket of the fiber was removed by emerging the ends of the fiber into methylene chloride. After taking off the protection jacket, the fiber was carefully cut. The cutting quality was checked with a $300\times$ microscope.

6.2.2 Laser Systems

The laser used in this experiment was a 30 ps Quantel mode locking Nd:YAG laser. The laser rod of the oscillator is cut at Brewsters' angle and is pumped by two Xenon flash lamps. Inside the cavity, an acousto-optical crystal is used as an active mode locker and a saturable absorber dye (Kodak) is used as a passive mode locker. The acousto-optical crystal is electrically modulated by a radio frequency signal. The modulation period is equal to the round trip time of the laser pulse in the cavity. The combined action of the saturable absorber and the acousto-optical modulator allows the production of a pulse train of typically twelve to sixteen 40 ps pulses within the Gaussian envelope. The total energy in the train is about 7 mJ. The pulses are p-polarized. The saturable absorber has the greater effect on decreasing the pulse duration, while the acousto-optical modulator has the greater effect on the shot to shot stability.

The train of pulses is then directed into a Pockels cell pulse selector. This device essentially consists of an electro-optic crystal and polarizing optics. A laser pulse can be selected from the laser pulse train, by applying a well defined voltage pulse across the electro-optic crystal, which rotates the polarization of the selected pulse from the train such that it can pass through a crossed polarizer. The pulse selected from the train is then amplified by a double pass ring amplifier which contains a half wave plate, polarizer, and polarizing beam splitters. The single laser pulse can be amplified to 75 mJ and is linearly polarized. Finally, the amplified pulse passes through a type I second harmonic generator (KDP) crystal which converts up to twenty percent of the incident 1064 nm laser pulse into its second harmonic frequency pulse. The pulse duration of the second harmonic laser is approximately 30 ps. A set of color filters was used after the laser systems to select the laser pulses.

6.2.3 Experimental Results

The experimental setup is shown in Fig. 6.10. The second harmonic pulse at 532 nm from the laser system was selected. The laser light is linearly polarized. A rotator was used to change the polarization direction. The laser beam was directed into a modified polarizing Michelson interferometer. The interferometer produces a pair of orthogonally polarized pulses by the polarization beam splitter. Two quarter wave plates were placed into two beam paths. The stronger beam was further split into two by a 50:50 beam splitter to produce two pump pulses. Each of those three beams was reflected back along its incident direction by a mirrors perpendicular to the incident beam. The relative time delays can be adjusted by changing the positions of the three reflection mirrors. The combination of the 1/4 wave plates and the polarization beam splitter prevents any power from being reflected back toward the laser. By rotating the polarization of the incident pulse, the relative intensities in the two polarizations are controlled. The three out put pulses from the modified Michelson interferometer are focused into the optical fibers by a

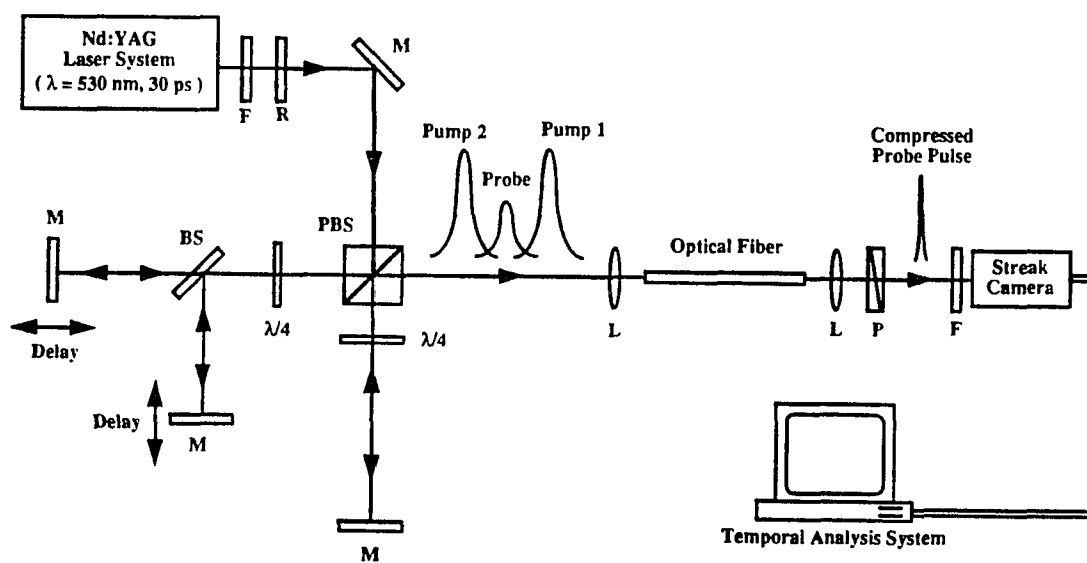


Figure 6.10. Experimental setup for pulse compression using DXPM. BS: beam splitter; PBS: polarization beam splitter; F: filters; L: 20× microscope objective lenses; M: mirrors; P: polarizer; and R: polarization rotator.

20× microscope objective lens. The probe pulse is extracted from the optical fiber output by a polarizer. The intensity was measured by a Hamamatus 2-ps streak camera.

The measured probe pulse from a 1.2 m polarization preserving single mode optical fiber is shown in Fig. 6.11. The energy of each pump pulse was about 90 nJ and the probe pulse energy was about 1/300 of that of the pump pulse. The delays between the probe pulse and the pump pulses were about 30 ps. The pulse duration of the output probe pulse was compressed to 1/3 of its original value. If the energy of the input pump pulse was increased to 120 nJ, stimulated Raman scattering occurred and the input pump and probe pulses were distorted.

The measured probe pulse from a 1.17 m single mode optical fiber is displayed in Fig. 6.12. The energy of each pump pulse was about 80 nJ and the probe pulse energy was about 1/300 of that of the pump pulse. The delays between the probe pulse and the pump pulses were about 30 ps. The pulse duration of the output probe pulse was compressed to 1/3 of its original value. No obvious amplification, as predicted by the theoretical calculations, was observed for the probe pulse. If the energy of the input pump pulse was increased to 90 nJ, the polarization of the input pump pulses changed and mixed with the probe pulse. The probe pulse could not be extracted from the output of the optical fibers.

6.3 Discussion

Both theoretical and experimental studies show that DXPM process can compress the pulse duration of a weak probe pulse by a combination of XPM and GVD without the use of gratings and prisms. The theoretical calculation also predicts the amplification of the weak probe pulse by the strong pump pulses in DXPM. However, this prediction was not seen in the experiment. Only pulse compression was observed. The reason is

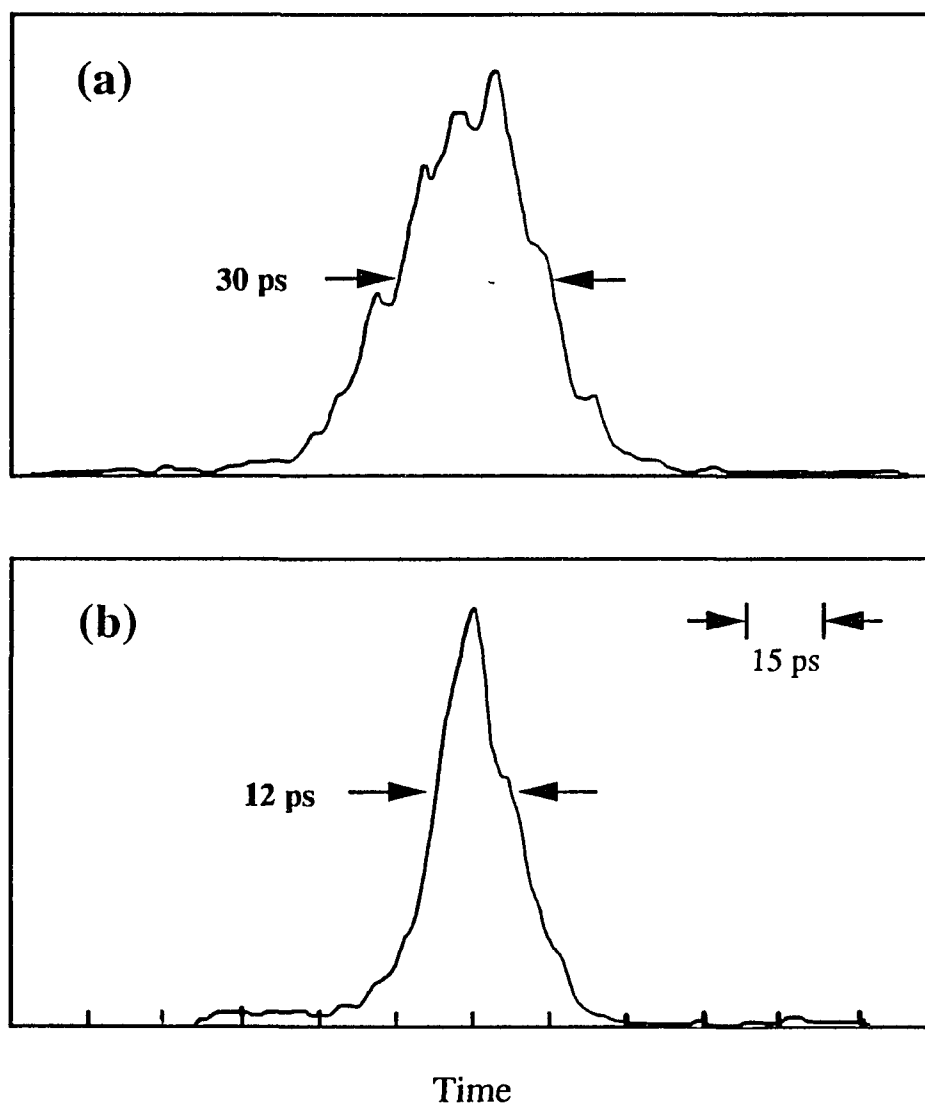


Figure 6.11. Pulse compression in polarization preserving optical fiber using DXPM. (a) input pulse; (b) compressed pulse. The length of fiber is 1.2 m. The pump pulse energy is about 90 nJ and the energy of the probe pulse is about $1/300$ of that of the pump pulse. The pulses are measured with a 2 picosecond streak camera.

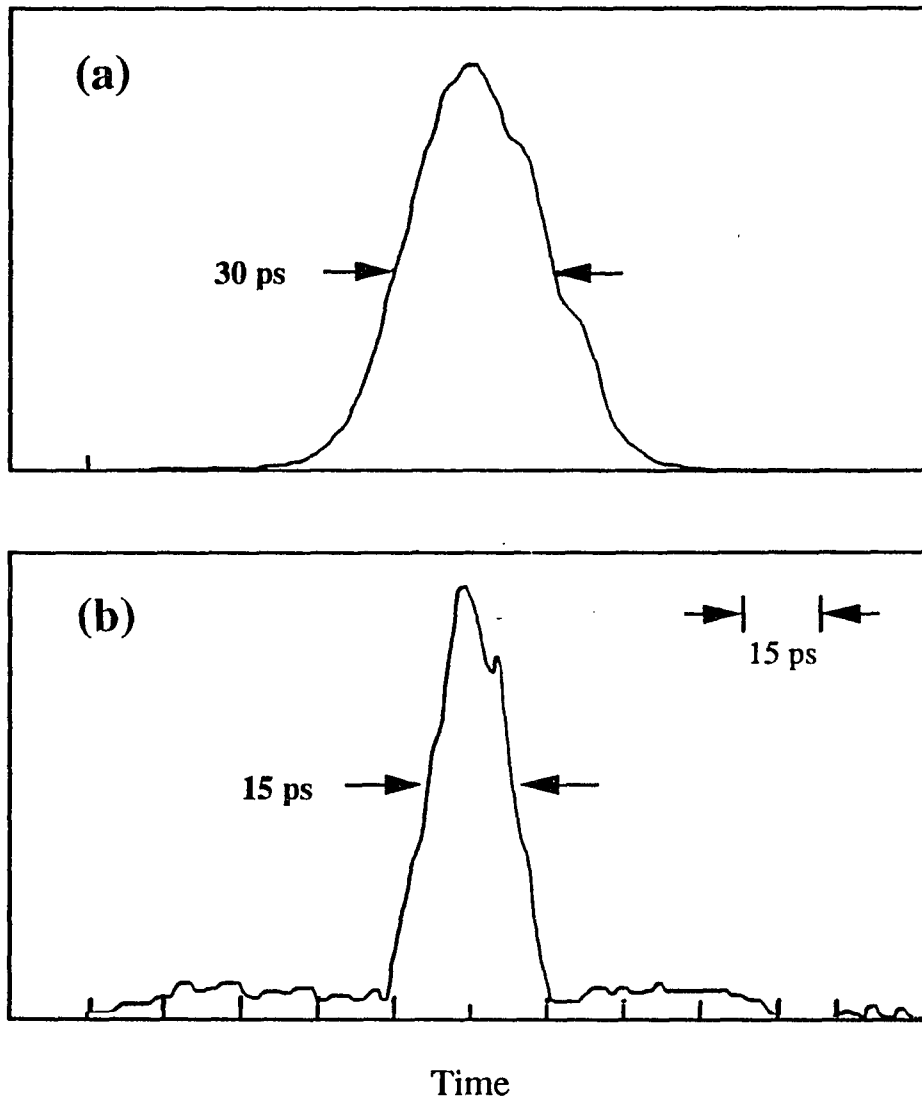


Figure 6.12. Pulse compression in single mode optical fiber using DXPM. (a) input pulse; (b) compressed pulse. The length of fiber is 1.7 m. The pump pulse energy is about 90 nJ and the energy of the probe pulse is about $1/300$ of that of the pump pulse. The pulses are measured with a 2 picosecond streak camera.

that the amplification effect becomes strong only when the weak pulse is compressed to less than one third of its original duration, as shown in Figs. 6.4. Due to the competition between SPM, XPM and the stimulated Raman scattering, the pulse duration of the probe pulse can be only compressed to about one third of its input value. Therefore, the amplification of the probe pulse was not dramatic.

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Chapter 7

Stimulated Hyper-Raman Scattering

Stimulated hyper-Raman scattering (SHRS)¹⁻⁶ is the three-photon analog of stimulated Raman scattering. In SHRS two pump photons are annihilated and a Stokes photon is created. SHRS is described by the fifth order ($\chi^{(5)}$) in the expansion of the electrical polarization.

This chapter presents our theoretical study and the experimental observation of SHRS based on $\chi^{(5)}$ in (-)-2-(*a*-methylbenzylamino)-5-nitropyridine (MBA-NP) crystal. In Section 7.1, a parametric instability theory⁷ is applied to compute the intensity dependence of SHRS signal. In Section 7.2 we present the experimental measurements of SHRS based on $\chi^{(5)}$ in an ultrathin MBA-NP crystal.

7.1 Theoretical Study of The Intensity Dependence of Stimulated Hyper-Raman Scattering

7.1.1 Equations of Motion of Occupation Numbers

The basis of the use of parametric instability theory in SHRS is the well-known result of quantum mechanics.^{8,9} The probability per unit time for a system originally in state $|i\rangle$, the initial state, to make a transition to another state $|f\rangle$, the final state, is

$$TP = \frac{2\pi}{\hbar} |\langle f|H|i\rangle|^2 \delta(\hbar\omega_f - \hbar\omega_i), \quad (7.1)$$

where H is the hamiltonian responsible for the scattering between the states.

In SHRS processes, two photon of the pump field are absorbed and one photon of the signal field and one optical phonon are created. A diagram of this process is shown in Fig. 7.1. The Hamiltonian for this four-particle process in which momentum is conserved can be written as

$$H = \sum_{klmn} V_{klmn} \Delta(K_k + K_l - K_m - K_n) b_k b_l b_m^\dagger b_n^\dagger + \text{c.c.}, \quad (7.2)$$

where b and $b^\dagger$'s are the creation and annihilation operators, K_i 's are the momenta, Δ is delta function, c.c. denotes hermitian conjugate, and V_{klmn} depends on k, l, m , and n in general. This hamiltonian has matrix elements for both the hyper-Raman process and its inverse process. For example, the matrix element of this hamiltonian for the hyper-Raman process of Fig. 7.1(a) is

$$\begin{aligned} & \langle n_1-1, n_2-1, n_3+1, n_4+1 | \sum_{klmn} \Delta(K_k + K_l - K_m - K_n) \\ & \times (V_{klmn} b_k b_l b_m^\dagger b_n^\dagger + V_{klmn}^* b_n b_m b_l^\dagger b_k^\dagger) | n_1, n_2, n_3, n_4 \rangle, \end{aligned} \quad (7.3)$$

with $l, k = L$, and $m = HR$, and $n = P$, and L denotes for laser, HR denotes for hyper-Raman, and P denotes for phonon. The only nonvanishing terms in the sum over $klmn$ are the terms with

$$k=1, \quad l=2, \quad m=3, \quad n=4;$$

$$k=1, \quad l=2, \quad m=4, \quad n=3;$$

$$k=2, \quad l=1, \quad m=3, \quad n=4;$$

$$k=2, \quad l=1, \quad m=4, \quad n=3;$$

and

$$k=3, \quad l=4, \quad m=2, \quad n=1;$$

$$k=4, \quad l=3, \quad m=2, \quad n=1;$$

$$k=3, \quad l=4, \quad m=1, \quad n=2;$$

$$k=4, \quad l=3, \quad m=1, \quad n=2;$$

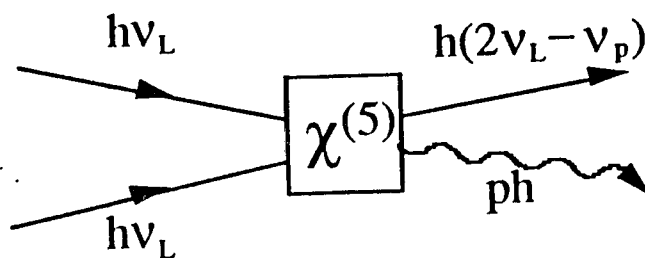


Figure 7.1. Diagram of hyper-Raman scattering process.

because the creation and annihilation operators $b_k b_l b_m^\dagger b_n^\dagger$ must operate on the initial state $|n_1, n_2, n_3, n_4\rangle$ to give the final state $|n_1-1, n_2-1, n_3+1, n_4+1\rangle$. Therefore, the matrix elements are

$$\begin{aligned}
 & \langle n_1-1, n_2-1, n_3+1, n_4+1 | \Delta(K_1 + K_2 - K_3 - K_4) (V_{1234} b_1 b_2 b_3^\dagger b_4^\dagger + \\
 & + V_{1243} b_1 b_2 b_4^\dagger b_3^\dagger + V_{2134} b_2 b_1 b_3^\dagger b_4^\dagger + V_{2143} b_1 b_2 b_4^\dagger b_3^\dagger) + \\
 & + \Delta(K_3 + K_4 - K_1 - K_2) (V_{4321}^* b_1 b_2 b_3^\dagger b_4^\dagger + V_{3421}^* b_1 b_2 b_4^\dagger b_3^\dagger + \\
 & + V_{4312}^* b_2 b_1 b_3^\dagger b_4^\dagger + V_{3412}^* b_2 b_1 b_4^\dagger b_3^\dagger) | n_1, n_2, n_3, n_4 \rangle \\
 & = [\Delta(K_1 + K_2 - K_3 - K_4) (V_{1234} + V_{1243} + V_{2134} + V_{2143}) \\
 & + \Delta(K_3 + K_4 - K_1 - K_2) (V_{4321}^* + V_{3421}^* + V_{4312}^* + V_{3412}^*)] \\
 & \times [n_1 n_2 (n_3 + 1) (n_4 + 1)]^{1/2}. \tag{7.4}
 \end{aligned}$$

Similarly, the nonvanishing matrix elements of the Hamiltonian for the inverse process are the terms with

$$k=3, \quad l=4, \quad m=1, \quad n=2;$$

$$k=4, \quad l=3, \quad m=1, \quad n=2;$$

$$k=3, \quad l=4, \quad m=2, \quad n=1;$$

$$k=4, \quad l=3, \quad m=2, \quad n=1;$$

and

$$k=1, \quad l=2, \quad m=3, \quad n=4;$$

$$k=1, \quad l=2, \quad m=4, \quad n=3;$$

$$k=2, \quad l=1, \quad m=3, \quad n=4;$$

$$k=2, \quad l=1, \quad m=4, \quad n=3.$$

The matrix elements are

$$\begin{aligned}
 & \langle n_1+1, n_2+1, n_3-1, n_4-1 | \Delta(K_4 + K_3 - K_2 - K_1) (V_{4321} b_4 b_3 b_2^\dagger b_1^\dagger + \\
 & + V_{3421} b_3 b_4 b_2^\dagger b_1^\dagger + V_{4312} b_4 b_3 b_1^\dagger b_2^\dagger + V_{3412} b_3 b_4 b_1^\dagger b_2^\dagger) + \\
 & + \Delta(K_1 + K_2 - K_3 - K_4) (V_{1234}^* b_4 b_3 b_2^\dagger b_1^\dagger + V_{1243}^* b_3 b_4 b_2^\dagger b_1^\dagger + \\
 & + V_{2134}^* b_4 b_3 b_1^\dagger b_2^\dagger + V_{2143}^* b_3 b_4 b_1^\dagger b_2^\dagger) | n_1, n_2, n_3, n_4 \rangle
 \end{aligned}$$

$$\begin{aligned}
&= [\Delta(K_4 + K_3 - K_2 - K_1)(V_{4321} + V_{3421} + V_{4312} + V_{3412}) \\
&\quad + \Delta(K_1 + K_2 - K_3 - K_4)(V_{1234}^* + V_{1243}^* + V_{2134}^* + V_{2143}^*)] \\
&\quad \times [(n_1 + 1)(n_2 + 1)n_3 n_4]^{1/2}.
\end{aligned} \tag{7.5}$$

By using the standard quantum mechanical transition-probability result (7.1) and

$$\frac{dn_{HR}}{dt} = \sum_{K_L K_L K_P} (TP_{n_{HR}} \text{ increased by 1} - TP_{n_{HR}} \text{ decreased by 1}), \tag{7.6}$$

one gets that the rate equation for the creation of hyper-Raman photon per volume V by the hyper-Raman scattering process is

$$\frac{dn_{HR}}{dt} = C [n_L^2 (n_{HR} + V^{-1})(n_P + V^{-1}) - (n_L + V^{-1})n_{HR} n_P], \tag{7.7}$$

where

$$C = \frac{2\pi}{\hbar^2} |V_{HR}|^2 V^3 \delta(\omega), \tag{7.8}$$

with

$$\begin{aligned}
|V_{HR}|^2 = & |V_{1234} + V_{1243} + V_{2134} + V_{2143} + \\
& + V_{1234}^* + V_{1243}^* + V_{2134}^* + V_{2143}^*|^2,
\end{aligned} \tag{7.9}$$

is the hyper-Raman matrix element, the argument of $d(\omega)$ is

$$\omega \equiv 2\omega_L - \omega_{HR} - \omega_P. \tag{7.10}$$

By considering the values of the occupation numbers of hyper-Raman photon and phonon in a small element dx of the laser beam at position x , we obtain the equations of motion of the occupation numbers

$$\begin{aligned}
\frac{\partial n_{HR}}{\partial t} = & C[(n_{HR} + n_P + V^{-1}) n_L^2 - 2 n_{HR} n_P n_L - n_{HR} n_P V^{-1}] \\
& - v \alpha_{HR} n_{HR} - v \frac{\partial n_{HR}}{\partial x},
\end{aligned} \tag{7.11}$$

and

$$\begin{aligned}
\frac{\partial n_P}{\partial t} = & C[(n_{HR} + n_P + V^{-1}) n_L^2 - 2 n_{HR} n_P n_L - n_{HR} n_P V^{-1}] \\
& - \Gamma_P (n_P - \bar{n}_P) - v_P \frac{\partial n_P}{\partial x},
\end{aligned} \tag{7.12}$$

where α_{HR} is the absorption coefficient, V the volume of the sample, v is the velocity of the hyper-Raman light in the medium, v_P is the phonon velocity, $\bar{n}_P$ is thermal-

equilibrium value of n_p , and Γ_p is the inverse lifetime of phonon n_p . The last term in Eq. (7.12) is negligible since the phonon velocity is very small compare the velocity of the light.

7.1.2 Intensity Dependence of SHRS Signal

The intensities of the laser and hyper-Raman scattering signal is $I_L = \hbar\omega_L c_L n_L$ and $I_{HR} = \sum \hbar\omega_{HR} v n_{HR}$, where the sum is over the hyper-Raman modes and c_L the velocity of laser light. Dispersion is neglected where $c_L = v$.

Assuming $n_p \gg V^{-1}$ and $n_p \gg n_{HR}$, Eq. (7.12) reduces to

$$\frac{\partial}{\partial t} (n_p - \bar{n}_p) = \Gamma_p \left(n_p \frac{n_L^2}{n_C} - (n_p - \bar{n}_p) \right), \quad (7.13)$$

where $n_C = [\Gamma_p / C]^{1/2}$ is the critical value of n_i . It can be seen from Eq. (7.13) that when the pumping laser density is greater than n_C , the phonon density n_p becomes unstable since $\frac{\partial}{\partial t} (n_p - \bar{n}_p) > 0$ for all n_p . From the experimental point of view it follows that at a certain critical effective laser density $I_C = \hbar\omega_L c_L n_C$ an explosive build up of the phonon occupation number, n_p , can take place. The increase in phonons cause a similar increase in the hyper-Raman Stokes radiation intensity.

Since the hyper-Raman scattering is a three photon process in which a system is excited with two photons of laser at n_L and emits one photon at the frequency $2\nu_L - \nu_p$, the photon occupation number at the laser frequency can be written as

$$n_{Leff} = n_i - 2 \sum n_{HR}, \quad (7.14)$$

where n_i is total photon occupation number of the incident laser pulse. Substituting Eq.(7.14) into Eqs.(7.11) and (7.12), the equation of motion for n_{HR} in the steady state case can be written as

$$\left(\frac{dn_{HR}}{dx} + \alpha_{HR} n_{HR} \right) \left(n_{LC}^2 - (n_i - 2 \sum_M n_{HR})^2 + 2 n_{HR} (n_i - 2 \sum_M n_{HR}) \right)$$

$$= \beta_P \left((n_{HR} + n_P) (n_i - 2 \sum_M n_{HR})^2 - 2 n_{HR} n_P (n_i - 2 \sum_M n_{HR}) \right) \quad (7.15)$$

where $\beta_P = \Gamma_P/v$. Since the phonon velocity v_P and value of one phonon per volume V are small, the last term in Eq.(7.12) and the terms with V^{-1} have been neglected in the derivation of Eq.(7.15).

For a first order approximation, $\sum_M n_{HR}$ in Eq.(7.15) can be formally replaced by Mn ,

where M is the number of effective modes and n is the average value of the hyper-Raman-photon occupation number in each mode. Eq.(5) reduces to

$$\frac{dI_{HR}}{dx} = \beta_P \frac{(I_{HR} + I_{SP})(I_i - I_{HR})^2 - I_{HR} I_{SP} (I_i - I_{HR})/M}{I_C - (I_i - I_{HR})^2 + I_{HR} (I_i - I_{HR})/M} - \alpha_{HR} I_{HR}, \quad (7.16)$$

where $I_{HR} = \hbar\omega_{HR}Mn$, $I_{SP} = \hbar\omega_{HR}M\bar{n}_P$, and $I_i = \hbar\omega_L n_i$.

When I_{HR} is much smaller than I_i , the explicit solution to Eq.(7.16) is

$$I_{HR} \propto I_{SP} (\exp(gx) - 1) + I_{HR}(0) \exp(gx). \quad (7.17)$$

where $g = \beta_P (I_i)^2 / [I_C^2 - (\gamma I_i - I_{HR})^2]$. The first term on the right-hand side of Eq.(7.17) is the amplified spontaneous emission of hyper-Raman and the second term is stimulated hyper-Raman scattering. When $gx \ll 1$ and $I_{HR}(0) = 0$, then $I_{HR} \propto I_{SP} \beta_P I_L^2 x$, which is the result of the spontaneous hyper-Raman intensity.¹⁰

A numerical method is used to solve Eq.(7.16) for the general case. Fig. 7.2 shows the numerical solution of Eq. (7.16) which describes an ideal case of SHRS process. As the laser power increases, the SHRS first increases linearly because it is generated from spontaneous scattering, and then grows quasi-exponentially when stimulated scattering sets in. At a certain input power, the output rises suddenly with a large gain. It finally levels off as a result of depletion of the laser power.

In practical cases, the gain may not be as great as the theoretical result due to the existence of other nonlinear processes, such as the enhanced phonon relaxation.¹¹ In the

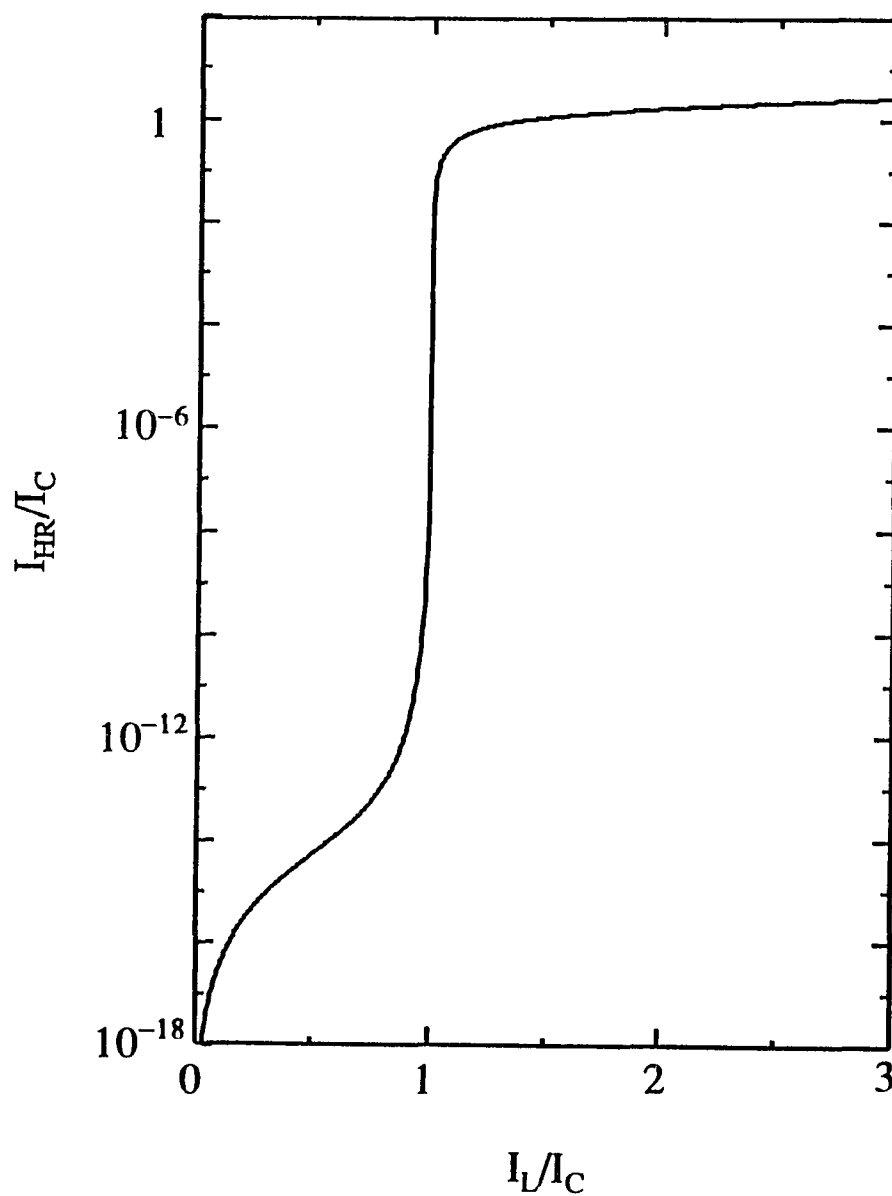


Figure 7.2. Intensity of SHRS signal as a function of laser intensity without induced phonon decay. The hyper-Raman signal increases rapidly as the input laser intensity and saturates due to the depletion of the pump laser.

following section, the intensity dependence of the SHRS is calculated when the phonon relaxation is enhanced by the phonon instability.

7.1.3 Effect of Phonon Relaxation on SHRS

An important effect which influences the intensity dependence of SHRS is the effect of a parametric instability of phonons.¹¹⁻¹⁴ In the hyper-Raman scattering process, two laser photons are annihilated and a hyper-Raman photon and fundamental phonon are created. The created phonon may split into other phonons. Here we consider the created phonon in the hyper-Raman scattering process splits into two phonons, and the splitting is subject to parametric instability.¹¹

Two central results of the phonon-parametric instability study are that the fundamental-mode occupation number is limited to values below a critical value and that there is an effective inverse life time of the fundamental phonon that is greater than the usual linear value as a result of an increase in the occupation number of the phonon modes into which the fundamental phonon decays.¹¹⁻¹⁴ Consequently, there will be no depletion of the laser power since the phonon occupation number participating in the hyper-Raman process is limited to a value below a threshold value.

With the consideration of the phonon instability, the equations of motion of the hyper-Raman phonon occupation number and hyper-Raman photon number become

$$\frac{dn_P}{dt} = \tilde{\Gamma}_P \{ [(n_{HR} + n_P + V^{-1}) n_L^2 - 2 n_{HR} n_P n_L - n_{HR} n_P V^{-1}] \tilde{n}_C^{-2} - (n_P - \bar{n}_P) \}, \quad (7.18)$$

$$\frac{\partial n_{HR}}{\partial t} = \tilde{\Gamma}_P [(n_{HR} + n_P + V^{-1}) n_L^2 - 2 n_{HR} n_P n_L - n_{HR} n_P V^{-1}] \tilde{n}_C^{-2} - v \frac{\partial n_{HR}}{\partial x}, \quad (7.19)$$

where $\tilde{\Gamma}_P$ is the effective inverse life time of the phonon enhanced by the phonon instability, and the critical value $\tilde{n}_C$ is defined as

$$\tilde{n}_C = [\tilde{\Gamma}_P / \tilde{C}]^{1/2}, \text{ and } \tilde{C} = \frac{2\pi}{\hbar^2} |V_{HR}|^2 V^3 \delta(\tilde{\omega}), \quad (7.20)$$

where V_{HR} is the hyper-Raman matrix element, and $d(\tilde{\omega})$, with $\tilde{\omega} \equiv 2\omega_L - \omega_{HR} - \omega_P$, is the line-shape factor obtained by replacing the δ function by a Lorentzian of width $\tilde{\Gamma}_P$.

On resonance, $\tilde{n}_C \propto \tilde{\Gamma}_P$, as can be seen from the Eq. (7.20). Thus, we can let

$$\tilde{n}_C = n_{C0} \gamma, \quad \gamma = \tilde{\Gamma}_P / \Gamma_P, \quad (7.21)$$

where n_{C0} is the linear value of $\tilde{n}_C$ with $\tilde{\Gamma}_P = \Gamma_P$ and γ is the enhancement factor for the relaxation of the phonon mode. The $n_{HR}n_P n_L$ and $n_{HR}n_P V^{-1}$ terms in Eq. (7.18) are important in the saturation region. This depletion of the laser beam power is neglected here. In this case, the steady state solution to Eq. (7.18) is

$$n_P = \frac{\gamma^2 \tilde{n}_P + I_{LC}^2 (n_{HR} + V^{-1})}{\gamma^2 - V^{-1} I_{LC}^2}, \quad (7.22)$$

where

$$I_{LC} = \frac{I_L}{I_C} = \frac{n_L}{n_{C0}}, \quad (7.23)$$

with $I_L = \hbar\omega_{LC} n_L$ and $I_C = \hbar\omega_{LC} n_{C0}$. Equation (7.22) reduces to the previous result in Section 7.1.2 in the limit of $\gamma=1$.

When the depletion of the laser beam power is neglected, the equation of motion for the number of hyper-Raman photons reduces to

$$\frac{\partial n_{HR}}{\partial t} = \tilde{\beta}_C I_{LC}^2 \gamma^{-1} (n_{HR} + n_P + V^{-1}) \nu - \nu \frac{\partial n_{HR}}{\partial x}, \quad (7.24)$$

where $\tilde{\beta}_C = \tilde{\Gamma}_P / \nu$.

Substituting (7.22) into (7.24) and solving the resulting equation for the case of $\gamma=1$ gives a result similar to that obtained in the previous section. Here we are interested in the effects of the enhanced phonon relaxation on the hyper-Raman intensity. The behavior of the solution can be determined by considering limiting cases. From References 11-14 we have

$$\gamma = \frac{1}{1 - n_P/n_U}, \quad (7.25)$$

where n_U is an upper bound to n_P . Substituting Eq. (7.25) into Eq. (7.22) and neglecting $\gamma^2 \tilde{n}_P + I_{LC}^2 V^{-1}$, we have

$$\gamma^2 I^2 + (\gamma^2 - 1) I_{LC}^2 - \gamma^2 (\gamma^2 - 1) = 0, \quad (7.26)$$

where $I^2 = I_{LC}^2 n_{HR}/n_U$. The solution for γ^2 is

$$\gamma^2 = \frac{1}{2} (1 + I_{LC}^2 + I^2) \left(1 + \left(1 - \frac{4 I_{LC}^2}{(1 + I_{LC}^2 + I^2)^2} \right)^{1/2} \right) \quad (7.27)$$

When I^2 is very small, the solution is $\gamma^2 = 1$. If I^2 is large enough, that is in the case of a large phonon enhanced relaxation effect, Eq. (7.27) gives

$$\gamma^2 = I^2. \quad (7.28)$$

Substituting this result into Eq. (7.24) gives

$$\begin{aligned} \frac{dI_{HR}}{dx} &= \frac{\beta}{\gamma} I_{HP} I_{LC}^2 \\ &= \beta \frac{1}{(I^2)^{1/2}} I_{HR} I_{LC}^2, \end{aligned} \quad (7.29)$$

The solution to Eq. (7.29) is

$$I_{HR} = \frac{1}{4} \beta^2 I_{LC}^2 I_{UX}^2 + \text{constant}. \quad (7.30)$$

where $I_U = \hbar \omega_{LC} n_U$.

The behavior of I_{HR} as a function of the intensity of the pump laser according to the limiting case of Eq. (7.30) and the result of last section is illustrated schematically in Fig. 7.3, where $n_{U0} = (\bar{n}_P + 1) \exp(\beta x)$. For low pump laser intensity ($I_{LR} < 1$), the phonon enhanced relaxation can be neglected and the hyper-Raman signal amplified from its spontaneous noise with a enhanced gain. When the input pump laser intensity is large, the hyper-Raman photon and the phonon numbers increase and cause the enhanced relaxation of the hyper-Raman phonons. The enhanced phonon relaxation limits the hyper-Raman phonons and cause the saturation of the hyper-Raman signal. The saturation level of the signal depends on the value of the upper bound of the phonon numbers. If the upper bound value of the phonons is large, the hyper-Raman signal saturates at a large value. If the upper bound of the phonons is small, the hyper-Raman signal saturates at a small value.

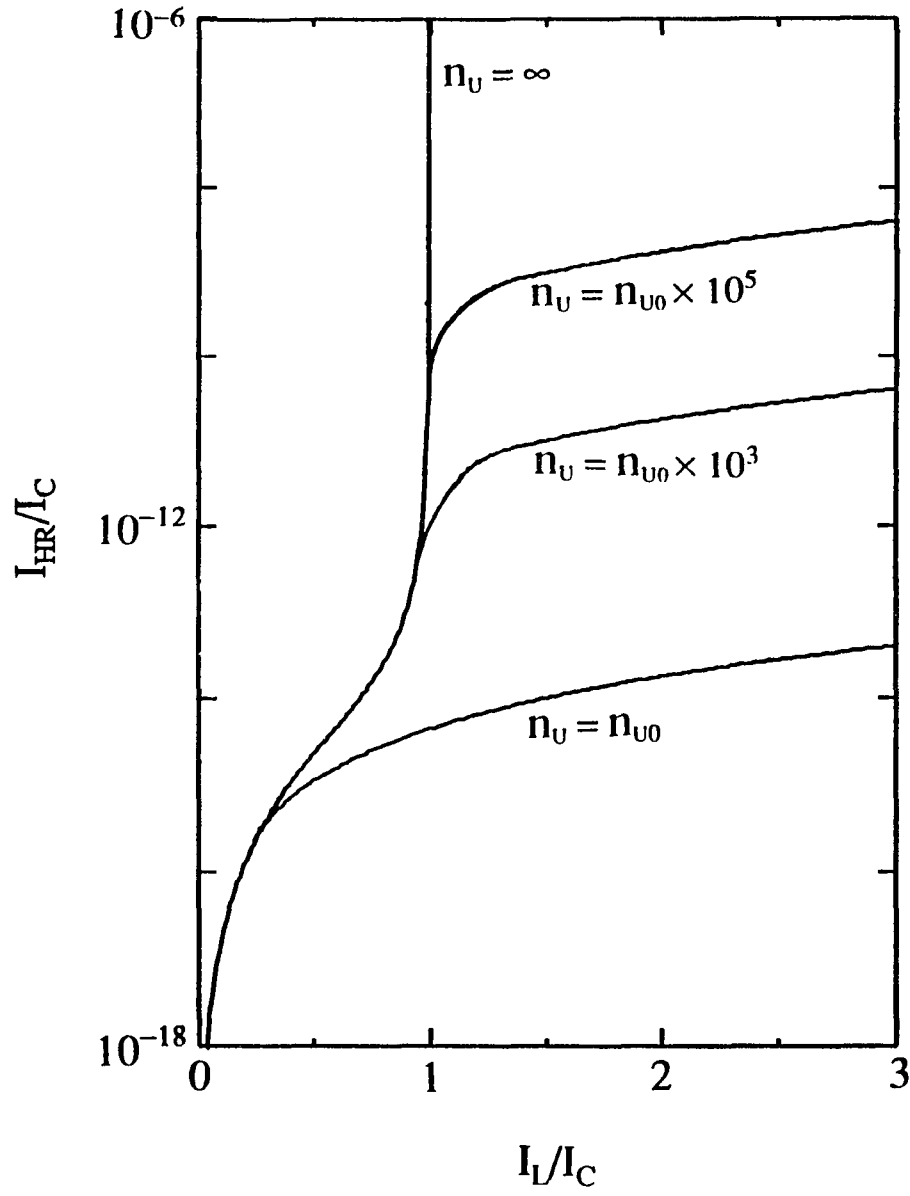


Figure 7.3. Schematic illustration of the effect of the phonon instability on hyper-Raman signal intensity. $n_{U0} = (\bar{n}_p + 1) \exp(\beta x)$.

7.2 Experiments on Stimulated Hyper-Raman Scattering

7.2.1 Sample Preparation

Future photonic devices will require optical materials which possess extremely large and fast nonlinear coefficients^{15,16}. Nonlinear optical materials based on p-conjugated organic materials have the potential to fulfill some of these basic requirements¹⁷⁻²² for nonlinear optical devices for communication and computation. The organic crystal (-)-2-(*a*-methylbenzylamino)-5-nitropyridine (MBA-NP) has attracted much interest because of its extremely large $\chi^{(2)}$ and high optical damage threshold.

The MBA-NP crystal was grown at British Telecom Research Laboratories in England.²³ Crystals of this material belong to the monoclinic system with the non-centrosymmetric space group $P2_1$, point group 2. It has unit cell parameters $a=5.392 \text{ \AA}$, $b=6.354 \text{ \AA}$, $c=17.924 \text{ \AA}$, and $\beta=94.6^\circ$. Fig. 7.4 shows the absorption curve of a 1.9-cm MBA-NP crystal. Fig. 7.5 displays IR spectrum of a 40-mm-thick MBA-NP crystal and the spontaneous Raman spectrum of MBA-NP crystal pumped by a 80-mW cw argon laser at 488 nm. The IR spectrum was measured with a FTS-40 Digilab Fast Fourier Transform IR spectrometer system. The Raman signal was detected by an optical multichannel analyzer system (EG & G) in combination with a Triplemate monochromator (Spex Industries Inc.). The IR spectrum shows that the mode at 992 cm^{-1} is IR active. No Raman signal was observed at this mode. Furthermore, the IR absorption in the possible Raman line region pumped by 1054 nm laser make this crystal be suitable for to the study of hyper-Raman scattering.

7.2.2 Experimental Setup

The experimental arrangement used to measure the hyper-Raman scattering signal is shown in Fig. 7.6. The laser system is the same as in Chapter 4. The 1054 nm laser pulse generated from the laser system with a spectral width of about 80 cm^{-1} , peak

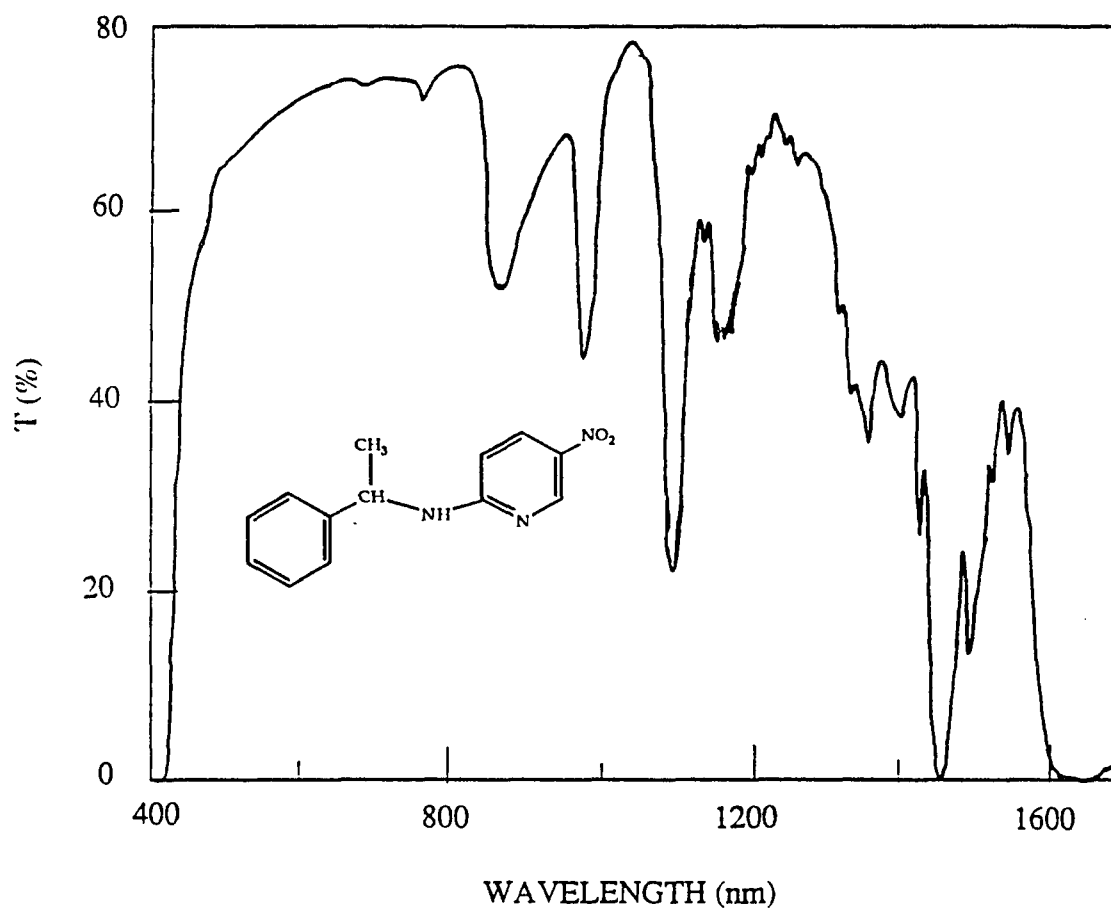


Figure 7.4. Absorption spectrum of a 1.9-cm MBA-NP crystal. The insert shows the molecular structure of MBA-NP.

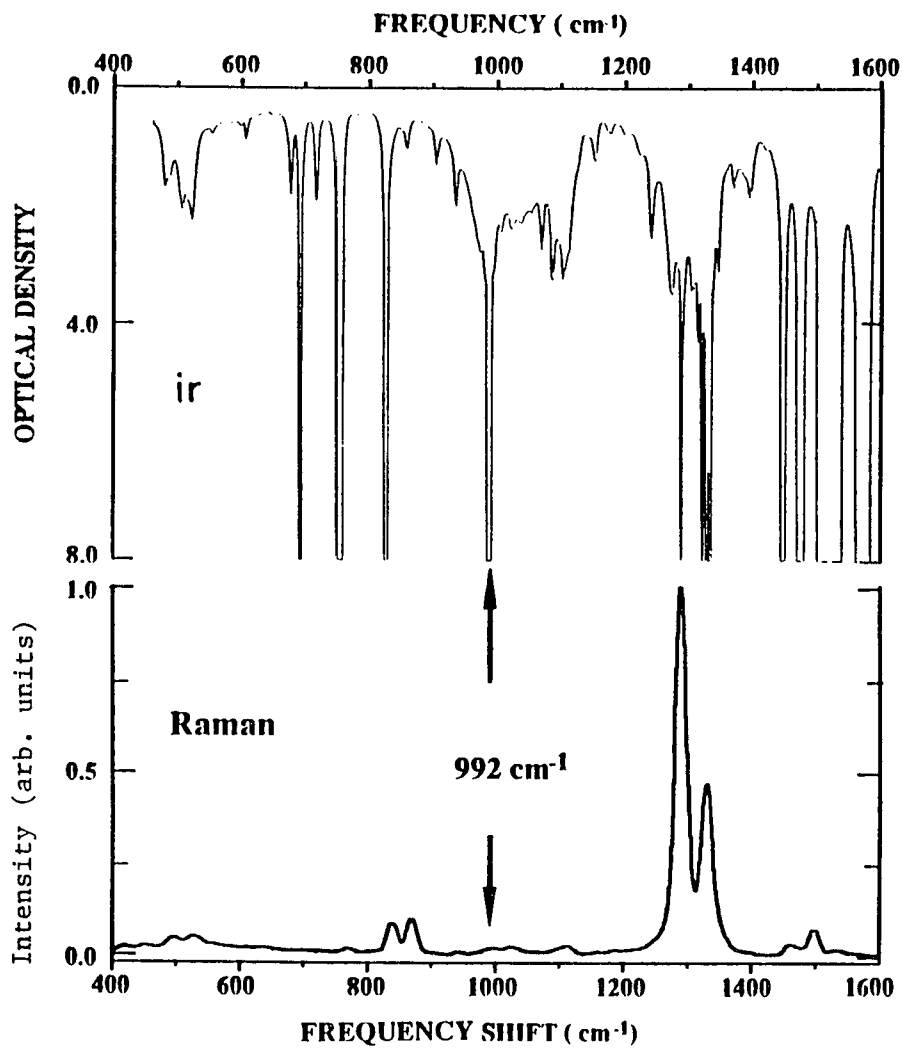


Figure 7.5. IR absorption spectrum of MBA-NP crystal with 40 μm thickness, and Raman spectrum of a MBA-NP crystal pumped by a cw laser at 488 nm.

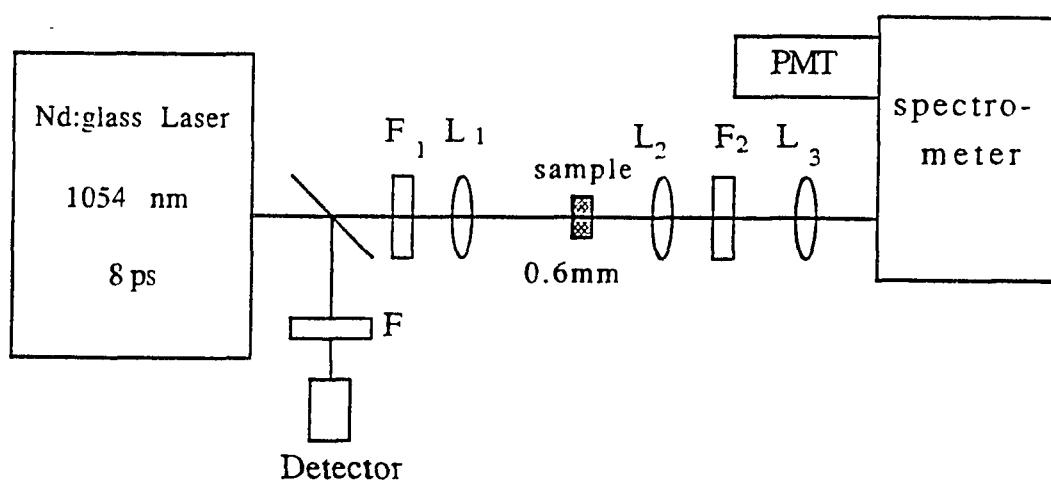


Figure 7.6. Experimental setup for SHRS. F: filters; L: lenses. The signal in IR regime is detected with S-1 photomultiplier tube or Ge-photodiode, or PdS-photodiode. The signal in visible and UV regimes are detected with MAO II system.

power about 109 W and the pulse duration of 8 ps. A KDP crystal was used to generate the second-harmonic of the 1054 nm laser. The laser beam was focused with a 28.5 cm focal length lens into a 0.6 mm thickness single crystal of MBA-NP. The beam diameter at the sample was about 1 mm. The incident laser energy was controlled by changing the neutral-density filter F_1 . The output signal in the visible and UV regions were collected by a 10 cm focal length lens and directed into a 0.25-m Jarrell-Ash spectrograph with a Par OMA III video readout system for spectral recording and analysis or a 0.5 m Jarrell-Ash spectrometer with a photomultiplier tube for intensity measurement. The output light in near-IR region was measured using either a 0.5 m Jarrell-Ash spectrometer with a Ge-photodiode or a PbS-photodiode detector, or a 0.25 m spectrometer with a CCD camera readout system.

7.2.3 Experimental Results

A typical nonlinear spectrum from ultra thin 600 μm MBA-NP crystal under 1054 nm, 10 ps pulses excitation is displayed in Fig. 7.7. The salient feature of the curve displayed indicates that there are four distinct maxima located at 527 nm (SHG), 556 nm, 351 nm (THG), and 364 nm. The two new spectral features are located at 556 nm and 364 nm, which are shifted by 992 cm^{-1} from SHG and THG signals, respectively. However, there was no observable stimulated signals in the infrared region due to absorption. Fig. 7.8 shows the intensity dependence of the hyper signal at $2\nu-\nu_p$ (556 nm) with the 1054 nm pump intensity. The signal intensity at 556 nm increases rapidly and saturates as shown in Fig. 7.8.

The temporal behavior of the SHG at 527 nm and 556 nm signals from the 1054 nm laser pulse passing through a 1.9 cm long MBA-NP crystal have been measured by a 2 ps resolution streak camera and are displayed in Fig. 7.9. The SHG signal has two separate peaks in time generated from the entrance and exit surfaces, respectively. This

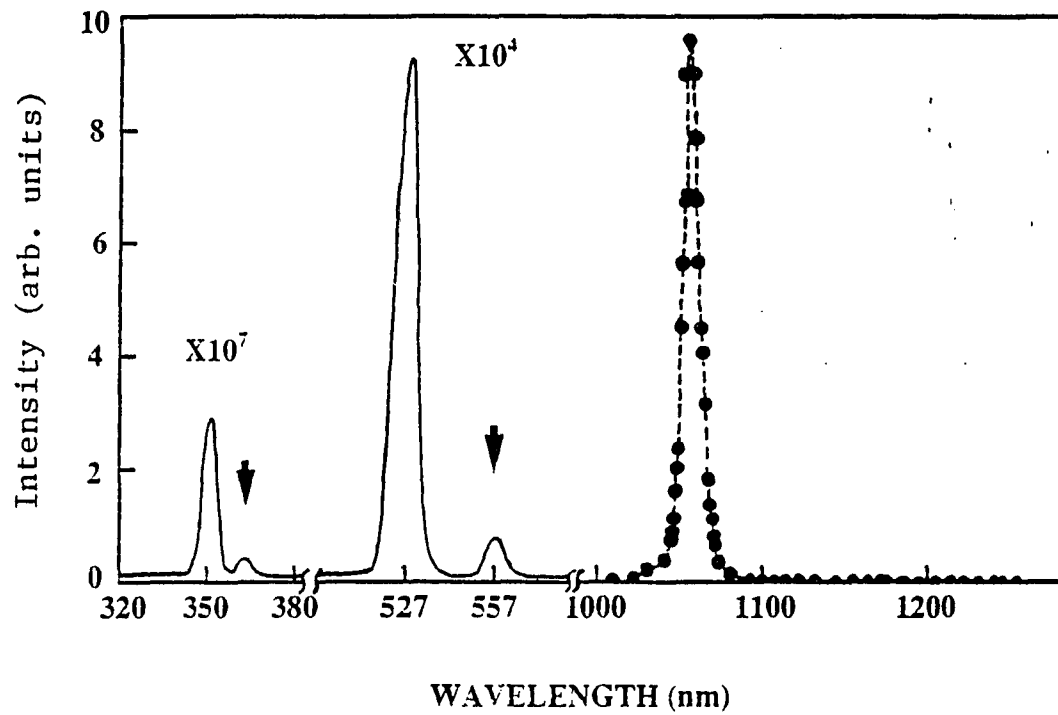


Figure 7.7. Nonlinear signal from 600-mm MBA-NP in UV and visible spectral regions generated by 1054 nm, 8-ps laser pulses. Arrows point to the observed salient features. The intensity from 330 to 370 nm and from 500 to 600 nm displayed have been multiplied by 10^7 and 10^4 , respectively.

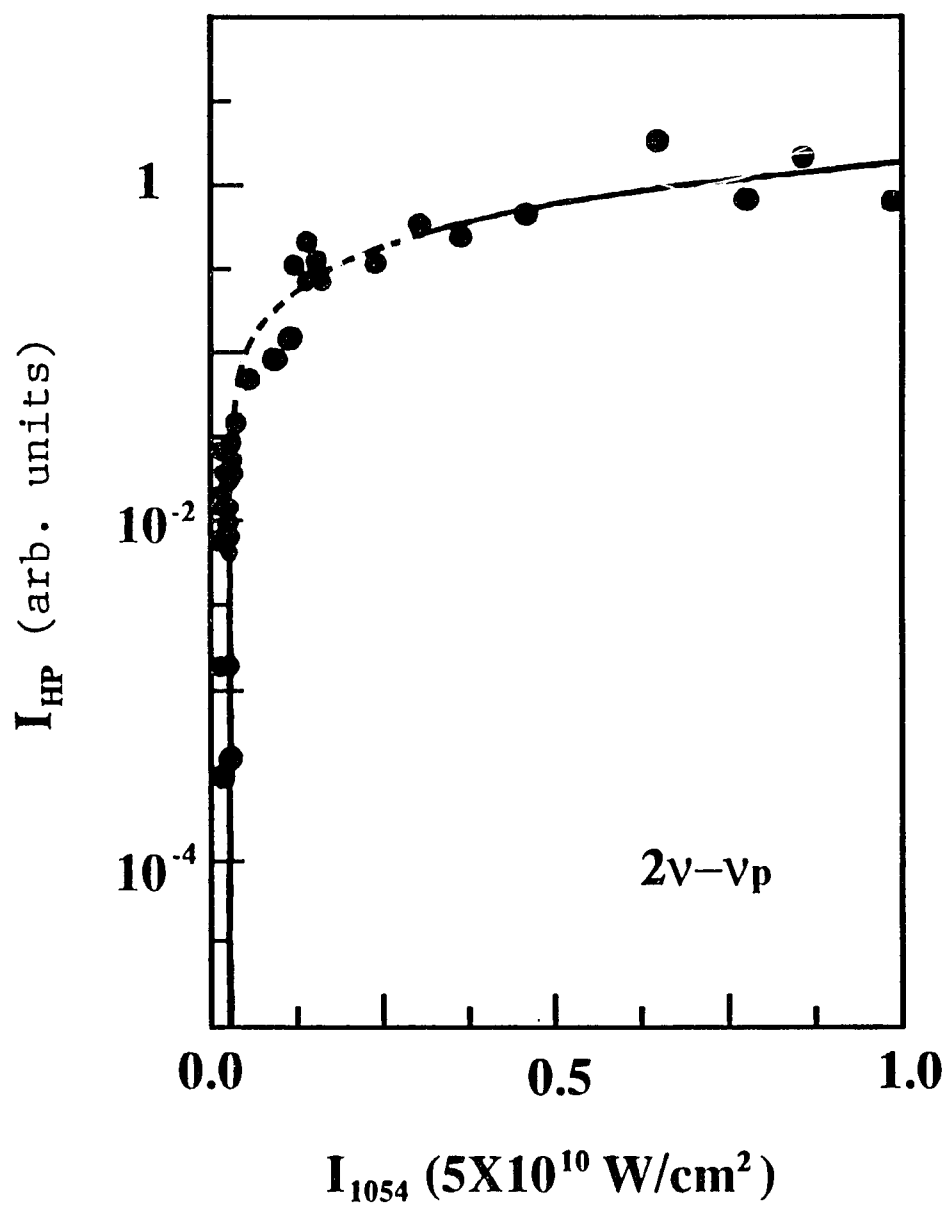


Figure 7.8. Intensity dependence of SHRS signal from ultrathin MBA-NP crystal. The solid curve is calculated from Eqs. (7.16) and (7.29).

signal is due to the phase mismatch and the interference of the signals generated in different positions. Only a sharp peak in time at 556 nm generated from the exit end of the crystal can be observed. This feature is a clear signal of stimulated emission pulse. The temporal distributions of the SHG and 556 nm signals generated from 600 μm MBA-NP are similar. Both have one clear peak with the pulse duration about the same as that of the pump pulse.

One key characteristic of the measured nonlinear spectral signal is that the intensity ratio I_{556}/I_{527} is almost equal to I_{364}/I_{351} when the input 1054 nm intensity changes from 3×10^9 to $5 \times 10^{10} \text{ W/cm}^2$. The intensities at 556 nm and 364 nm are about one tenth of the intensities at 527 nm and 351 nm, respectively. The signal intensity at the SHG frequency 527 nm varies as $I_{2\nu} \propto I_\nu^2$. The signal intensity at THG is found to be almost linear to that at SHG. Phase-match second harmonic generation was not observed because of the poor quality of the sample and orientation.

The signal at 3 ν can arise from both the third harmonic generation [$\propto \chi^{(3)}$] and the three wave mixing [$\propto \chi^{(2)}$]. To find out which signal generating process is responsible for 3 ν , both transmission and reflection methods were performed. Our experimental results indicates that the intensity at 3 ν is linearly proportional to the intensity at 2 ν for both cases when the input laser intensity at 1054 nm was high and kept constant. These observations indicate that the signal at 3 ν mainly originates from the three wave mixing via $\chi^{(2)}$ from signals at 2 ν and ν . It was estimated experimentally that about less than 10% of the intensity of the signal at 3 ν was generated from THG through $\chi^{(3)}$. The absorption depths of both MBA-NP and ZnSe are comparable to the coherence length. A standard model of THG^{16,17} was used to estimate the value of the third-order susceptibility of MBA-NP. It was found that the $\chi^{(3)}$ of MBA-NP is less than one percent of that of ZnSe, about the order of 10^{-12} esu.

From experiments, the signals at frequency 3 ν - ν_p mostly arises from three wave mixing with strong signals at ν and 2 ν - ν_p through $\chi^{(3)}$. The signal intensity at 3 ν - ν_p was

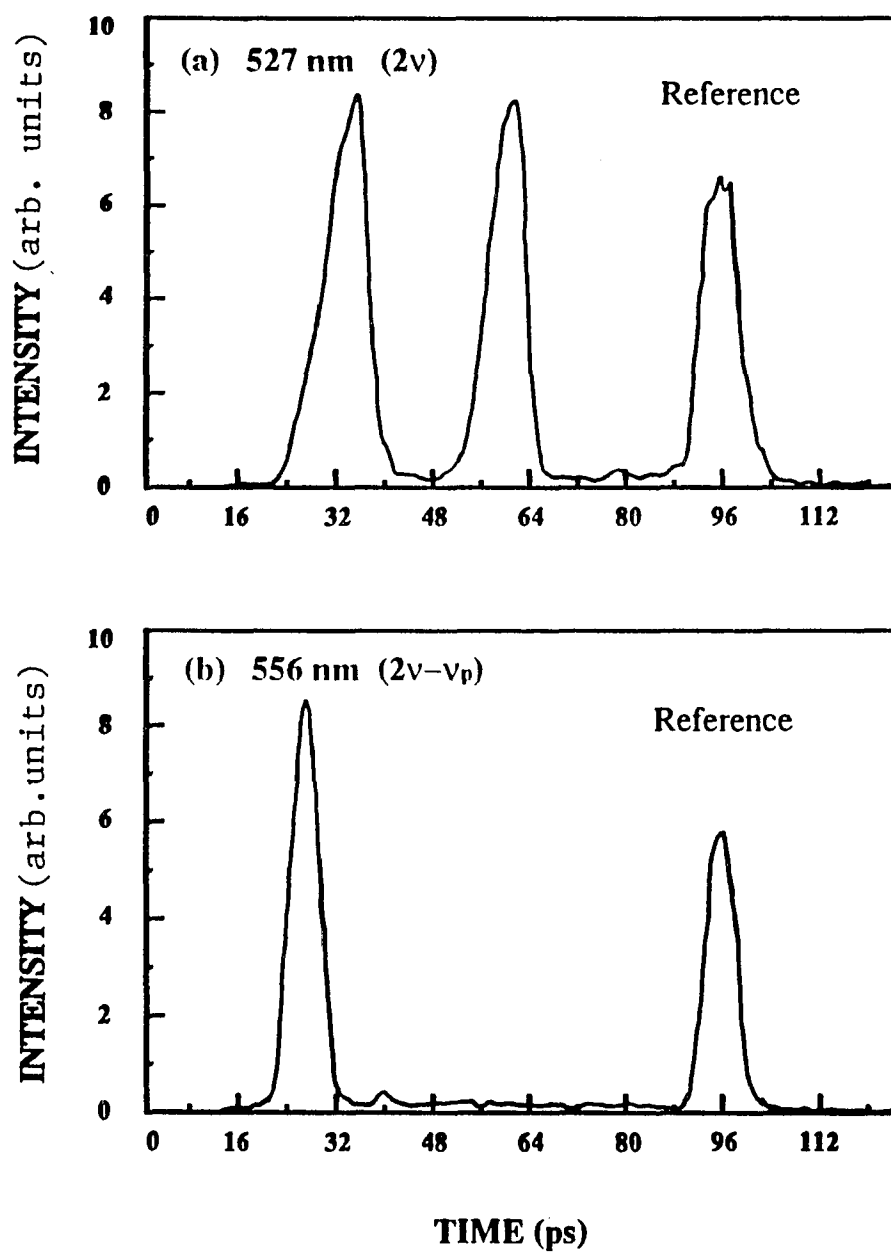


Figure 7.9. Temporal behavior of the nonlinear signals generated from a 1.9 cm long MBA-NP crystal. (a) SHG signal at 527 nm; (b) hyper-Raman signal at 556 nm.

found to vary as $I_{3\nu-\nu_p} \propto (\chi^{(2)})^2 I_{2\nu-\nu_p}$. Therefore, the ratio of the intensity at $2\nu-\nu_p$ to the intensity at 2ν is identical to the ratio of $3\nu-\nu_p$ to 3ν ,

$$\frac{I_{3\nu-\nu_p}}{I_{3\nu}} = \frac{(\chi^{(2)})^2 I_{2\nu-\nu_p} I_\nu}{(\chi^{(2)})^2 I_{2\nu} I_\nu} = \frac{I_{2\nu-\nu_p}}{I_{2\nu}}. \quad (7.30)$$

Besides the possibility of stimulated hyper-Raman scattering¹²⁻¹⁷ for our observed signal generated at $2\nu-\nu_p$, there may be three other possibilities: (1) stimulated Raman scattering via 2ν ; (2) second harmonic generation of the stimulated Raman of the pump laser; (3) parametric hyper-Raman generation of the incident laser ν and its stimulated Raman scattering ($\nu_p = 992 \text{ cm}^{-1}$) at $\frac{1}{\nu-\nu_p} = 1177 \text{ nm}$. However, the later three

possibilities can be ruled out by the following experimental results.

When an external 527 nm laser pulse with an intensity comparable to the SHG generated by 1054 nm passed through the MBA-NP crystal, no signal at $2\nu-\nu_p$ was observed. When the incident intensity of the primary pumping laser was kept constant, the output intensity at $2\nu-\nu_p$ remained almost constant when the intensity at 2ν was changed externally. In addition, no Raman signal at 992 cm^{-1} was observed in the spontaneous Raman spectrum. These observations rule out the first possibility. Due to the IR absorption in the possible Raman line region, no strong stimulated Raman of the pump laser was observed in IR region as shown in Fig. 7.7. This is the key factor supporting the stimulated hyper Raman scattering hypothesis. The lack of stimulated Raman scattering of the primary pump laser in the IR region makes the stimulated hyper Raman scattering the first nonlinear process in the MBA-NP crystal pumped by 1054 nm laser pulses. Furthermore, there was no sinusoidal oscillation, which is the typical feature of the signal from frequency up-conversion parametric process, in the temporal distribution of the signal at 556 nm. These results rule out the second and third possibilities. The IR absorption spectrum shows that there is a strong absorption band about $\nu_p = 992 \text{ cm}^{-1}$. Therefore, the signal at $2\nu-\nu_p$ appears to arise from hyper-Raman scattering of the primary pump laser.

7.3 Discussion

The solid line in Fig. 7.9 is a fit to the experimental results on the intensity dependence of hyper-Raman scattering using Eqs. (7.16) and (7.29). Eq. (7.16) is used for $I_{LC} < 1$, and Eq. (7.29) is used for $I_{LC} > 1$. In the region where $I_{LC} \approx 1$, the situation is between the two limiting cases and is more complicated. In the calculation, the value of $\beta_p = 0.7 \text{ cm}^{-1}$ was used which corresponds to $\Gamma^{-1} \approx 10^{-11} \text{ s}$. By adjusting the value of I_C , excellent agreement has been achieved over five decades between the theoretical calculation and the experimental results when input laser intensity is higher than $1 \times 10^9 \text{ W/cm}^2$. When the laser intensity was lower than $1 \times 10^9 \text{ W/cm}^2$, the hyper-Raman signal reduced to about 10^{-6} of the saturation value. That value was below our experimental sensitivity.

Although the parametric instabilities of phonons¹¹⁻¹² were used to explain the observed saturation of the SHRS signal, no direct experimental evidence has been observed for the phonon instability. The number of vibrational modes at any k value in a molecular crystal is large. In MBA-NP crystals, there are four molecules in a unit cell. Each molecule contains 31 atoms. There are 372 vibrational modes per k value. Therefore, the phonon structure and dynamics in MBA-NP crystal are very complicated. More work on the dynamics of phonons in MBA-NP is needed to explain the saturation of the SHRS signal.

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Chapter 8

Future Directions of XPM Research

This chapter describes some examples of the possible applications using XPM-based processes for signal processors to develop optical communication networks and optical computers with data stream in the terahertz regime. The research presented in this thesis can lead to several applications.

8.1 Ultrashort Optical Pulse Compression and Amplification for Long Distance Optical Communications

Figure 8.1 displays a possible method using degenerate cross-phase-modulation (DXPM)¹⁻³ for the pulse compression and amplification of long distance optical fiber communication system. After passing through a long distance fiber, the signal pulse broadens due to the group velocity dispersion and the pulse energy losses due to the attenuation. To retrieve the intensity and pulse shape, a pulse amplification and compression unit based on DXPM is used. In this amplification and compression unit, the broadened output optical signal pulse from optical fibers is split into two. One beam with a small portion of the energy is used as a trigger signal for the semiconductor pump laser system and the other, after a well defined optical delay system, is coupled with two orthogonally polarized pump laser pulses with the same intensities produced by the pump laser system into the condensed matter where the signal pulse is amplified and compressed via DXPM. The delay time between two pump pulses can be varied by adjusting the delay of the reflecting mirrors. The amplified and compressed signal pulse

is extracted from the output of the amplification and compression unit by a polarizer and re-coupled into the fiber chain for the long distance transmission. This process can be repeated to compensate the dispersion effect in fiber link communication systems.

8.2 Ultrashort DXPM A/D Converter

The schematic of an DXPM based optical A/D converter is shown in Fig. 8.2. It consists of two main components: a nonlinear optical material for DXPM processing and a frequency demultiplexer (DEMUX) to separate the digitized signal. The nonlinear optical materials can be either a glass fiber,⁴ a polymer waveguide,⁵ a semiconductor micro-structure,⁶ or bulk semiconductors⁷. The DEMUX can be a grating or a prism. In the DXPM A/D process, a sampled linear polarized ω signal will be used as the pump pulse. A pulse with the same frequency but perpendicularly polarized to the pump pulse will be used as the probe pulse. The output frequency of the probe pulse will be shifted depending on the intensity of the pump pulse. From the results of Chapter 6, the magnitude of the frequency shift is linearly proportional to the pump pulse intensity. After a DEMUX, the frequency shifted probe pulse will be dispersed into different angles as a function of the induced frequency shift. For a 4-bits operation, a photo-diode array detector with 16-channels can be used to record the modulated probe signal at different frequencies. For example, when $0 \leq I_{in} < \Delta I$, the induced frequency shift is $0 \leq \Delta\omega < \Omega$, the DEMUX output of the probe pulse will be recorded by the Channel 0, where ΔI is a designated intensity unit for the digitization and Ω is the corresponding unit value of the induced frequency shift. when $(N-1)\Delta I \leq I_{in} < N\Delta I$, the induced frequency shift is $(N-1)\Omega \leq \Delta\omega < N\Omega$, the DEMUX output of the probe pulse will be recorded by the Channel (N-1). Analog intensity of the input pump pulse can be digitized into coded numbers from 0 to 15.

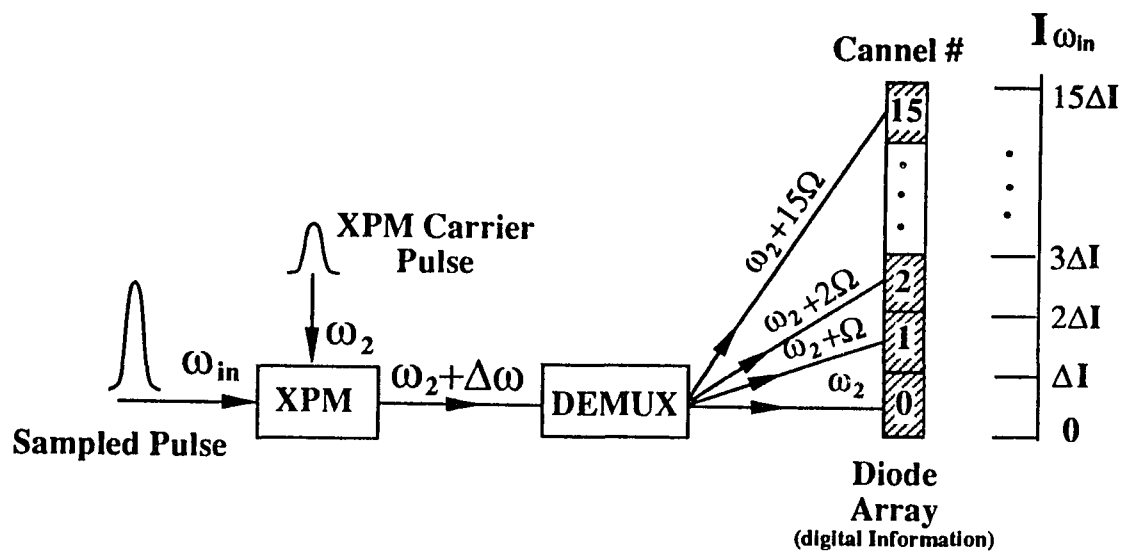


Figure 8.2. Diagram of a XPM analog/digital signal converter.

The above angular digitized information in decimal format can be converted into the binary format using an anamorphic optical system. The anamorphic (ANA) optics which uses the combination of cylindrical and spherical lenses can convert the angular dispersed optical beam after the DEMUX into a line source.⁸ A schematic of the setup is displayed in Fig. 8.3. After the lens system, the probe pulse is spread into 16-line format according to the intensity of the sampled analog pulse. A binary mask can map each individual line into binary digital forms in time. For example, in a 4-bit binary mask, 1 will be converted by the mask into (0001), 2 will be converted into (0010), and so on. After this position mask, signals will be correspondingly delayed into a set of 4 pulse train in time.

Higher A/D bit accuracy, such as 8-bit A/D converter, can be obtained by increasing the frequency shift. The induced frequency shift can be increased by increasing the interaction length and the third-order optical nonlinearity of the material. DXPM can be used to increase the interaction length due to the small walk-off effect between the pump and probe pulses.

8.3 Ultrashort Optical Pulse Attraction and Repulsion Using Cross-Phase-Modulation: Pulse Position Modulation

An extension of XPM is an induced velocity change by the induced index. Fig. 8.4 shows the schematic diagrams of ultrashort optical pulse attraction or repulsion by the induced index changes. In the absence of a pump pulse, the weak probe pulse passes through an undisturbed medium. When the signal pulse copropagates in the nonlinear medium with a pump pulse leading it, if the response time of the nonlinearity is longer than the time interval between the pump and probe pulses, the effective index of refraction for the probe pulse will be changed. Hence, the probe pulse will be sped up if

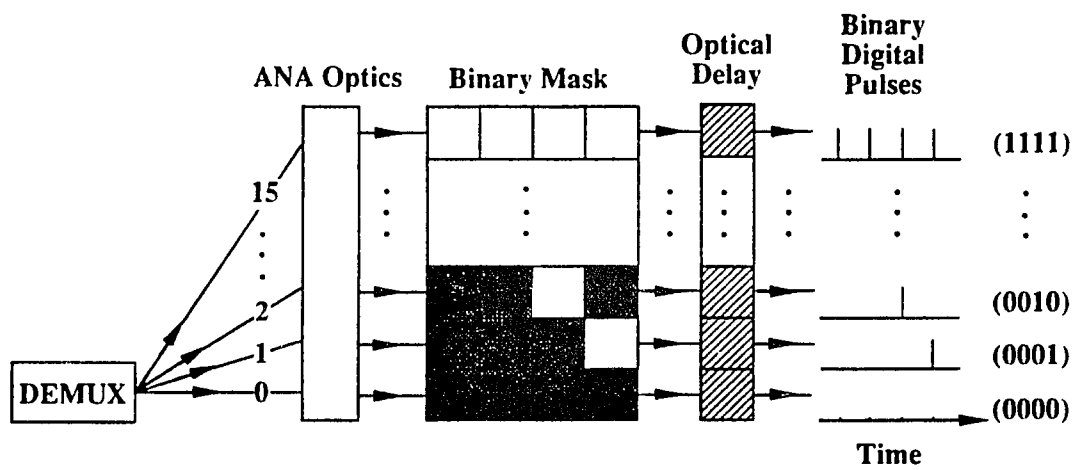


Figure 8.3. Schematic illustration of binary digital pulse generation.

the nonlinearity is negative and will be slowed down if the nonlinearity is positive. In the former case the probe pulse is "attracted" by the pump pulse and in the latter case it is "pushed" by the pump pulse. The magnitude of the attraction and repulsion depends on the intensity of the pump pulse. In Fig. 8.4, the signal pulse S1 and S2 were shifted by Δt_1 and Δt_2 by the pump pulses P1 and P2, respectively, while S3 is not affected by the stream of pump pulses.

If the pump pulses are a sampled analog signal pulse train and the probe pulses are a pulse train containing pulses with the same time interval and same amplitudes, the time interval between pulse in the output probe pulse train will be changed. The position of the pulses within the time slot allocated to each pulse conveys the amplitude information. The pulse delay relative to some reference point in the slot is proportional to the amplitude of the sample. This is called pulse-position modulation (PPM).⁹

This induced position change of pulses can be also used to scramble and unscramble the ultrashort pulse optical signals. If a positive nonlinear medium within certain magnitude of the nonlinearity is used to scramble the signal, a negative nonlinear medium with the same magnitude of the nonlinearity can be used to unscramble the signal.

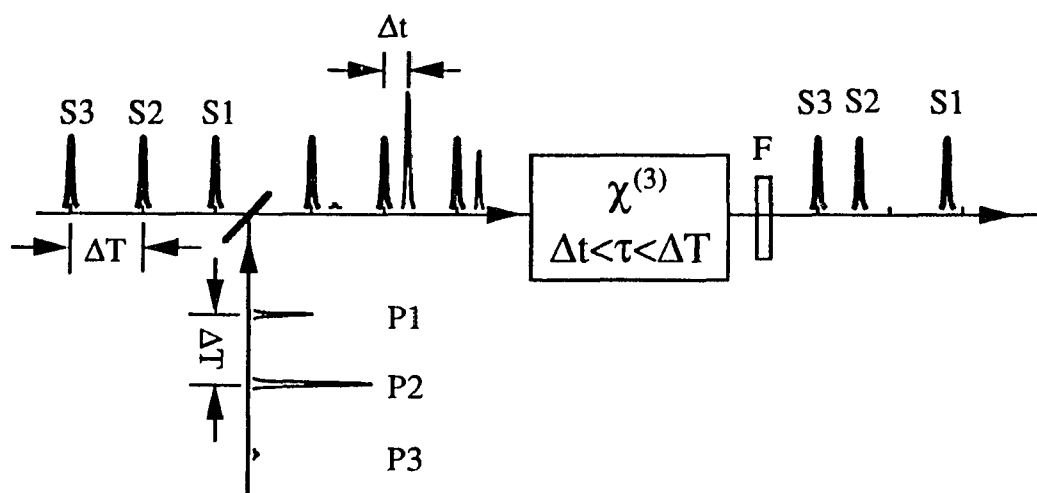


Figure 8.4. Pulse position modulation by induced nonlinear index change.

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Appendix

SHG-XPM.f

```

c   This program is used to calculate the temporal
c   profiles of SHG-XPM
double precision f1,f2,ap1,ap2,ac1,ac2,u,x,s1,s2,ps,h
open(11,file="da3.d")
n=1000
h=0.022/float(n)
m=250
d=100.0/float(m)
do 20 j=101,150
  u=170.0+j*d
  ap1=f1(u,0.0)+f1(u,0.022)
  ap2=f2(u,0.0)+f2(u,0.022)
  x=0.0
  ac1=0.0
  ac2=0.0
  do 4 i=1,n-1
    x=x+h
    ac1=f1(u,x)+ac1
4   ac2=f2(u,x)+ac2
    ap1=2.0*ac1+ap1
    ap2=2.0*ac2+ap2
    x=-0.5*h
    ac1=0.0
    ac2=0.0
    do 10 i=1,n
      x=x+h
      ac1=f1(u,x)+ac1
10   ac2=f2(u,x)+ac2
      s1=(4.0*ac1+ap1)*h/6.0
      s2=(4.0*ac2+ap2)*h/6.0
      ps=sqrt((s1)**2+(s2)**2)
20   write(11,100) u,ps
100  format(2f16.12)
close(11,status="keep")
end
double precision function f1(u,x)
double precision u,x
f1=dexp(-2.0*(u/6.79-36.29+381.4*x)**2+100.0*x)*dcos(23028.0*x
1 -(1.0d+6)*x*dexp(-2.0*(u/6.79-36.29+381.4*x)**2))
return
end
double precision function f2(u,x)
double precision u,x
f2=dexp(-2.0*(u/6.79-36.29+381.4*x)**2+100.0*x)*dsin(23028.0*x
1 -(1.0d+6)*x*dexp(-2.0*(u/6.79-36.29+381.4*x)**2))
return
end

```

multidxpm.f

```

c   This is a program used to calculate the
c   spectral distribution of DXPM in multimode
c   optical fibers
parameter(n=2048)
dimension c(16)
real f(n),g1(n),g2(n)
double precision u,f0,t
open (11,file="d1.d")
t=110.0/dfloat(n)
l=60.0
a=1.5
b=2.0*1*4.9e-3
c(1)=1.0
c(2)=0.8
c(3)=0.65
c(4)=0.5
c(5)=0.4
c(6)=0.3
c(7)=0.25
c(8)=0.2
c(9)=0.15
c(10)=0.12
c(11)=0.1
c(12)=0.08
c(13)=0.06
c(14)=0.04
c(15)=0.02
c(16)=0.01
do 10 i=1,n
u=t*float(i)
f(i)=0.0
d=0.0
do 5 k=1,16
f(i)=f(i)+c(k)*exp(-2.0*(u/6.8-a-b*float(k-1))**2)
5   d=d+c(k)
f(i)=f(i)/d
write (11,100) u,f(i)
g1(i)=f(i)*cos(500.0*float(i)-17.0*1*f(i))
10  g2(i)=f(i)*sin(500.0*float(i)-17.0*1*f(i))
call fftc(n,g1,g2)
open (12,file="d9.d")
do 20 i=1,2001
s=sqrt(g1(i)**2+g2(i)**2)
f0=3.0d+7/527.0
u=3.0d+7/(f0-dfloat(i-866)/1.1)
20  write (12,100) u,s
100 format (2f16.8)
close (12,status="keep")
end

```

singledxpm.f

```

c      This is a Program to calculate the temporal and
c      spectral distributions of the pump and probe pulses for
c      single pump pulse DXPM in a nonlinear dispersive medium
c      in normal dispersion region. The DFWM terms
c      are neglected.
      implicit double precision (a-h, o-z)
      common /char/anam, bnam
      common /per/rat, irat, nt, delt
      character * 12 fk, f
      character * 13 anam, bnam
      complex u(1001),u1(1001),w(1001),w1(1001)
      complex d,v1,v2,v3,v4
      print *, "input names of datafiles for PUMP and PROBE"
      read (5,1000) anam,bnam
1000   format (A4,3x,A4)
      print *, anam,bnam
      print*, "enter tstart, tstop, delt"
      read (5,1100) tstart,tstop,delt
1100   format (f4.1,3x,f4.1,3x,f9.6)
      print *, tstart, tstop, delt
      print *, "enter zstart, zstop, delz, zn"
      read (5,1200) zstart,zstop,delz, zn
      print *, zstart,zstop,delz,zn
1200   format (f4.1,3x,f4.1,3x,f12.9,3x,f4.1)
      print*, "enter N"
      read (5,1300) n
1300   format (I3)
      print *, n
      print *, "enter Ipump/Iprobe and deltau"
      read (5,1500) rat, deltau
1500   format (f7.1,3x,f9.6)
      print *, rat, deltau
      b=delz/delt**2/2
      nt=(tstop-tstart)/delt+1
      ntt=nt-1
      nz=(zstop-zstart)/delz
      z1=(zstop-zstart)/zn
      d=cmplx(0,1)
      pi=3.1415926540
c
      do 500 k=6,7
      k1=int(k/10.0)
      k2=int(k-k1*10.0)
      kk1=k1+48
      kk2=k2+48
      fk=char(kk1)//char(kk2)
      z=zstart
      do 10 i=1,nt
      t=tstart+(i-1)*delt
      t1=t-(5-k)
      th=thita*pi
      u(i)=exp(-t1*t1/2.0-d*th)

```

```

    u1(i)=u(i)
    w(i)=exp(-t*t/2)/sqrt(rat)
    w1(i)=w(i)
10  continue
    j=-1
20  j=j+1
    j1=int(j/10.0)
    j2=int(j-j1*10.0)
    jj1=j1+48
    jj2=j2+48
    f=char(jj1)//char(jj2)
    k1=int(k/10.0)
    anam=anam(1:4)//fk(1:2)//f(1:2)//".d"
    bnam=bnam(1:4)//fk(1:2)//f(1:2)//".d"

c    store temporal distributions of pulses
    open (1, file=anam)
    open (2, file=bnam)
    write (1,*) "z=",z
    write (2,*) "z=",z
    if (rat.lt.20) go to 140
    irat=int(rat/10.)
    do 130 i=1,nt
        t=tstart+(i-1)*delt
        uu=u1(i)*conjg(u1(i))
        write (1,*) t,uu
        ww=irat*w1(i)*conjg(w1(i))
        write (2,*) t,ww
130  continue
    go to 160
140  do 150 i=1,nt
        t=tstart+(i-1)*delt
        uu=u1(i)*conjg(u1(i))
        write (1,*) t,uu
        ww=w1(i)*conjg(w1(i))
        write (2,*) t,ww
150  continue
160  close (1,status="keep")
    close (2,status="keep")

c    Compute the spectral distributions
    call ftsp(u1,w1)

c
    if (z.ge.zstop) go to 500
    zz=zstart+(j+1)*z1
200  do 250 i=2,ntt
        v1=u(i+1)-2*u(i)+u(i-1)
        v2=u(i)*conjg(u(i))+2.0/3.0*w(i)*conjg(w(i))
        u1(i)=u(i)-d*(b*v1-deltz*n*n*v2*u(i))
        v3=w(i+1)-2*w(i)+w(i-1)
        v4=w(i)*conjg(w(i))+2.0/3.0*u(i)*conjg(u(i))
        w1(i)=w(i)-d*(b*v3-deltz*n*n*v4*w(i))
250  continue
    do 300 i=1,ntt

```

```

      u(i)=u1(i)
      w(i)=w1(i)
300  continue
      z=z+delz
      if(z-zz)200,20,20
500  continue
      stop
      end

      subroutine ftsp(u1,w1)
c      This subroutine is used to calculate the spectral
c      distributions (Fourier Transformation) of pump and
c      probe pulses for DXPM in a nonlinear dispersive medium
c      in normal dispersion region.
      implicit double precision (a-h, o-z)
      common /char/anam, bnam
      common /per/rat, irat, nt, delt
      character * 6 s
      character * 13 anam, bnam, cnam, dnam
      complex u1(1000), w1(1000)
      real ar(1000), ai(1000), br(1000), bi(1000)
      s="sp"
      cnam=s(1:2)//anam
      dnam=s(1:2)//bnam
      if (rat.lt.20) go to 140
      sirat=sqrt(float(irat))

c
      nnt=nt-1
c
      do 130 i=1,nnt
      ar(i)=u1(i)
      ai(i)=aimag(u1(i))
      br(i)=sirat*w1(i)
      bi(i)=sirat*aimag(w1(i))
130  continue
      go to 160
140  do 150 i=1,nnt
      ar(i)=u1(i)
      ai(i)=aimag(u1(i))
      br(i)=w1(i)
      bi(i)=aimag(w1(i))
150  continue
c
160  call fftc(nnt,ar,ai)
      call fftc(nnt,br,bi)
c
      ntt=nnt/2
      ftt=nt/(nnt*delt)
c
      open(1,file=cnam)
      open(2,file=dnam)
      do 200 i=1,nnt
      l=ntt+i
      if (l.gt.nnt) l=i-ntt

```

```
fre1=(l-1)/(nnt*delt)
if (fre1.ge.fitt) fre1=fre1-1./delt
aa=(ar(l)*ar(l)+ai(l)*ai(l))/nnt/25.132724761963
bb=(br(l)*br(l)+bi(l)*bi(l))/nnt/25.132724761963
write (1,*) fre1, aa
write (2,*) fre1, bb
200 continue
close (1, status="keep")
close (2, status="keep")
return
end
```

ddsxpm.f

```

c      This is a Program for calculation of
c      double pulse DXPM in a nonlinear dispersive medium
c      in normal dispersion region.
      implicit double precision (a-h, o-z)
      character * 12 fk, f
      character * 13 anam, bnam
      complex u(1001),u1(1001),w(1001),w1(1001)
      complex d,v1,v2,v3,v4
      print*, "input names of datafiles for PUMP(two pulses) and PROBE"
      read (5,1000) anam, bnam
1000   format (A4,3x,A4)
      print *, anam, bnam
      print*, "enter tstart, tstop, delt"
      read (5,1100) tstart, tstop, delt
1100   format (f4.1,3x,f4.1,3x,f9.6)
      print *, tstart, tstop, delt
      print *, "enter zstart, zstop, delz"
      read (5,1200) zstart, zstop, delz
      print *, zstart, zstop, delz
1200   format (f4.1,3x,f4.1,3x,f12.9)
      print*, "enter N"
      read (5,1300) n
1300   format (I3)
      print *, n
c      read in phase mismatching
      print *, "enter thita"
      read (5,1400) thita
1400   format (f14.10)
      print *, thita
      b=delz/delt**2/2
      nt=(tstop-tstart)/delt+1
      ntt=nt-1
      nz=(zstop-zstart)/delz
      z1=(zstop-zstart)/10.0
      d=cmplx(0,1)
      pi=3.1415926540
c
      do 500 k=7,7
      k1=int(k/10.0)
      k2=int(k-k1*10.0)
      kk1=k1+48
      kk2=k2+48
      fk=char(kk1)//char(kk2)
      z=zstart
      do 10 i=1,nt
      t=tstart+(i-1)*delt
      t1=t+(5-k)
      t2=t-(5-k)
      th=thita*pi
      u(i)=exp(-t1*t1/2.0-d*th)+exp(-t2*t2/2.0+d*th)
      u1(i)=u(i)
      w(i)=exp(-t*t/2)/sqrt(1000.0)

```

```

w1(i)=w(i)
10  continue
    j=-1
20  j=j+1
    j1=int(j/10.0)
    j2=int(j-j1*10.0)
    jj1=j1+48
    jj2=j2+48
    f=char(jj1)//char(jj2)
    k1=int(k/10.0)
    anam=anam(1:4)//fk(1:2)//f(1:2)//".d"
    bnam=bnam(1:4)//fk(1:2)//f(1:2)//".d"
    open (1, file=anam)
    open (2, file=bnam)
    write (1,*) "z=",z
    write (2,*) "z=",z
    do 150 i=1,nt
        t=tstart+(i-1)*delt
        uu=u1(i)*conjg(u1(i))
        write (1,*) t,uu
        ww=100.*w1(i)*conjg(w1(i))
        write (2,*) t,ww
150  continue
        close (1,status="keep")
        close (2,status="keep")
        if (z.ge.zstop) go to 500
        zz=zstart+(j+1)*z1
200  do 250 i=2,ntt
        v1=u(i+1)-2*u(i)+u(i-1)
        v2=u(i)*conjg(u(i))+2.0/3.0*w(i)*conjg(w(i))
        v2=v2*u(i)+1.0/3.0*w(i)*w(i)*conjg(u(i))
        u1(i)=u(i)-d*(b*v1-deltz*n*n*v2)
        v3=w(i+1)-2*w(i)+w(i-1)
        v4=w(i)*conjg(w(i))+2.0/3.0*u(i)*conjg(u(i))
        v4=v4*w(i)+1.0/3.0*u(i)*u(i)*conjg(w(i))
        w1(i)=w(i)-d*(b*v3-deltz*n*n*v4)
250  continue
        do 300 i=1,ntt
            u(i)=u1(i)
            w(i)=w1(i)
300  continue
            z=z+deltz
            if(z-zz)200,20,20
500  continue
        stop
    end

```

ihprm.r

```

#      This program calculate the intensity dependence
#      of hyper-ramman
#      find I(hyper-Raman)
#
#      External eqns, print
common /nmbr/beta,xa,xb,r1
common /data/errpar,dt,x,tstart
common E10
#
real x(2),dx,dt,tstart
real errpar(2)
character *12 fn
#
print*,"enter beta, Isp, mode# "
read*,beta, xa, xb
tstop=0.06
tstart=0.0
print*,"enter dt, errpar(1),errpar(2)"
read*, dt, errpar(1), errpar(2)
print*,"enter N, step and file name"
read*, n, st, fn
do i=1,n {
  E10=0.01+(i-1)*st
  x(2)=E10
  open (3, file=fn)
  #
  call odes(eqns,x,2,tstart,tstop,dt,errpar,print)
}
stop
end
#
subroutine eqns (t,x,n,dx)
common /nmbr/beta,xa,xb,r1
#
real t,x(1),dx(1)
#
a=(x(2)-x(1))
b=(x(1)+xa)*a**2-x(1)*xa*a/xb
c=1-a**2+x(1)*a/xb
dx(1)=beta*b/c
#
return
end
#
subroutine print (t0,x0,t1,x1,n,dt,tstop,e)
common E10
#
real t0,x0(n),t1,x1(n),dt,tstop
real e(n)
#
if(t0 == T1) return
#

```

```
jout=i1mach(2)
write(jout,9998) t1,x1(1),x1(2),dt
9998 format(2x,1P3E20.8,1PE14.2)
if (t1>=tstop) { write (3,*) E10, log10(x1(1))}
#
if (t1 <= tstop) return
return
end
```

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