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**OPTICAL TWO-WAVE MIXING
IN
IRON-DOPED INDIUM PHOSPHIDE**

by

Nick C. Peereboom, B.A.Sc.

**A thesis submitted to the
Faculty of Graduate Studies and Research
in partial fulfillment
of the requirements for the degree of**

Master of Engineering

**Ottawa-Carleton Institute for Electrical Engineering
Department of Electronics
Faculty of Engineering
Carleton University**

March 1992

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"OPTICAL TWO-WAVE MIXING IN IRON-DOPED INDIUM PHOSPHIDE"

**submitted by Nick C. Peereboom, B.A.Sc.
in partial fulfillment of the requirements for
the degree of Master of Engineering**

Thesis Supervisor

Chair, Department of Electronics

**Carleton University
March 6, 1992**

Abstract

The thesis reviews the theory of photorefractive two-wave mixing in InP:Fe, including derivations of response time and optical gain. Iron concentration [Fe] in InP:Fe was measured using absorption spectra. The photorefractive response time and optical gain in InP:Fe and GaAs crystals were measured experimentally as functions of polarization, grating spacing, optical intensity and external bias. Characterizing the InP:Fe crystal led to experimental demonstration of a photorefractive temperature-intensity resonance which peaked at $T=16^{\circ}\text{C}$ and $I_0=3.7 \text{ mW/cm}^2$ for $\lambda=1.064 \mu\text{m}$. The ratio of the iron species concentrations $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ was measured using the photorefractive effect while the crystal was under illumination. The ratio changes as a function of optical intensity. A maximum photorefractive gain of 7 cm^{-1} , or 6 cm^{-1} net gain, was demonstrated in bulk InP:Fe. The result was a five millimeter long 15 dB optical amplifier.

There are four contributions to theory. First, room temperature optical cross-section spectra of $[\text{Fe}^{+2}]$ and $[\text{Fe}^{+3}]$ were predicted using published data for 77°K and theoretical multi-band cross-section models. Second, quantitative investigation of the source of deep level optical absorption in InP:Fe was performed. Third, the concentration of iron in InP:Fe was measured using optical absorption. Iron was resolved as two charge species Fe^{+2} and Fe^{+3} using room temperature sub-band gap absorption spectra for $[\text{Fe}^{+2}]$ and $[\text{Fe}^{+3}]$ cross-sections. Fourth, PISCES finite element modelling was performed to simulate the photorefractive effect. The dynamic formation of photorefractive space charge electric field was observed with steady-state amplitudes greater than 20 kV/cm (gain = 6 cm^{-1}).

The two-wave mixing apparatus consisted of single-mode fiber-optic arms which formed two coherent laser beams. The beams passed directly through the crystal without the aid of folding mirrors. The technique is a practical contribution to the experimental methods because it simplifies set-up of photorefractive experiments. In addition, the fiber technique makes two-wave mixing compatible with integrated optics applications.

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CHAPTER 1

INTRODUCTION

The thesis examines optical two-wave mixing (2WM) in semi-insulating InP. 2WM is a branch of nonlinear optics with application to optical signal processing, real time image processing, and amplification of wide band telecommunications signals. In addition to many useful applications, 2WM is one of the more fundamental processes for the study of photorefractive materials. 2WM is worthy of investigation in its own right because it is a fast nondestructive technique for measuring III-V material parameters.

Study of photorefractive 2WM in InP began in 1984. The motivation for looking at III-V semiconductors such as InP for photorefractive media is a combination of two capabilities: relative speed and telecommunication compatibility. III-V semiconductors have faster response times compared with other photorefractors such as BaTiO₃, and they operate in the infrared making them compatible with direct parallel processing of telecommunications optical fiber signals.

The object is to report a single channel optical amplifier using 2WM in InP:Fe, and to model the effect using the semiconductor's material parameters.

1.1 Photorefractive Effect (PRE)

1.1.1 Description

The photorefractive effect is the light induced change of the refractive index in a crystal based on spatial modulation of photocurrents through deep level absorption of nonuniform illumination [1]. Optical absorption, carrier transport, and the electro-optic effect combined together produce an energy exchange between two beams. The energy redistribution in 2WM arises from two-beam coupling between the incident beams in the dynamic phase volume grating created by the electro-optic effect. The phase grating wave is shifted by some angle from the intensity wave. The beam coupling equations show that the $\pi/2$ shifted component of the phase grating causes one optical beam to be parametrically amplified, and the other optical beam depleted.

The photorefractive effect results from the sequence of events depicted in plan view in Figure 1.1 [2], where one type of charge carrier is assumed for the sake of

Figure 1.1 - The photorefractive effect in two-wave mixing [2].

simplicity. **Figure 1.1** shows the sequence in six panels (A) through (F), each panel illustrating a different parameter of the same mixing region.

In panel (A), the optical waves travel in the $+z$ direction, with some $\pm x$ component, entering the bottom of the depicted medium and leaving at the top. There are approximately 20 optical wavelengths between the bottom and the top of the medium. The peak of each wave period is represented by an almost horizontal slanted line. The lines indicate the phase front of the wave. For waves heading in the $-x$ direction, the phase front lines slope left. For waves heading in the $+x$ direction, the phase front lines slope right. The Huygens' construction shows how the optical waves interfere, forming an optical standing wave with optical maxima in the middle and on the far left and right sides of the panel.

Panel (B) shows the steady state result of the standing optical wave. The dots represent charge carriers. More photons are absorbed by deep level traps at the optical peaks than in the valleys. The absorption of photons frees trapped charge carriers, and space periodic charge clouds form based on the point-to-point differences in photon absorption rate. The carriers that make up the clouds drift and diffuse away from the optical peaks and tend to collect in the optical valleys in the pure diffusion case.

Panel (C) shows the result of the charge transport in panel (B): regions of fixed ionized traps (labeled "+") are separated by $\Lambda/2$ from regions of mobile free charge (labeled "-").

Panel (D) shows the space charge electric field E_{sc} generated by the charge separation in panel (C).

Panel (E) shows the result of E_{sc} in panel (D): the field distorts the crystal lattice by stretching and compressing interatomic bonds. The small total distortion is exaggerated to emphasize the points of maximum change.

Panel (F) shows the result of the optically distorted crystal lattice: a modulated refractive index in the x axis. Panels (E) and (F) show the electro-optic

Figure 1.2 - Vector diagram demonstrates momentum conservation in exact Bragg scattering of the pump into the signal.

interaction called the Pockels effect. The result is the volume phase grating $n(x)$. Due to net diffusion of free carriers, part of $n(x)$ is shifted by $\pi/2$ from the optical standing wave.

The $\pi/2$ quadrature phase condition results in one way energy transfer between the two interfering waves. The one way energy transfer is a result of the coupling constant introduced by $n(x)$. The coupling is described by the coupled wave equations. Without a signal beam, there is no coupling even though the phase grating remains for many milliseconds. In other words, the pump is coupled into the signal instead of classically diffracted into the signal. The coupling speed depends on the transverse optical lattice vibration frequency, on the order of ten terahertz, or 1000 gigahertz.

Note that the phase grating $n(x)$ at any phase angle relative to the optical standing wave automatically satisfies the Bragg condition to scatter one beam into the other, as can be seen in a momentum matching diagram. For example, see **Figure 1.2**. However, the photorefractive effect is not just a fancy name for a diffraction grating. The grating would be just a first order Bragg scatterer if the grating were an amplitude grating. Two interfering waves would scatter into each other with some small diffraction efficiency. Instead, the photorefractive crystal is more than a Bragg scatterer. The phase shifted component of $n(x)$ modulates the propagation constant in the medium, and the propagation

constant modulates the two wave coupling constant. The coupling constant describes the coupling between the pump wave and the signal wave. Either beam by itself is not noticeably affected by the grating. However, when the two beams are together, a one way energy transfer occurs, proportional to the sine of the phase shift between $n(x)$ and the intensity grating (see Section 4.2). Energy goes from one wave to the other wave but not the opposite way. The direction of the transfer depends on the majority photocarrier, the sign of the electro-optic coefficient, and the sign of the phase angle of $n(x)$. The coupling comes from the linear optical susceptibility and the optical polarization density waves being proportional to the instantaneous signal beam electric field and interaction with the out of phase refractive index grating (see equations (4.52) and ff for details).

1.1.2 Pockels Effect

The Pockels effect, also known as the linear electro-optic effect, is the name given to the electro-optic phenomenon where the refractive index change is proportional to the signed magnitude of the electric field. The Pockels effect is only found in those relatively few materials which have acentric unit cells. The most common are ferroelectric crystals such as LiNbO_3 and BaTiO_3 , photoconductors such as BSO, and III-V semiconductors such as GaAs and InP. Acentric unit cells lack inversion symmetry. The lack of symmetry leads to nonreciprocal energy transfer through the electro-optic coefficient.

In photorefractive experiments, a DC electric field applied in one direction may cause a small positive change in refractive index due to small changes in the electric field distributions in the interatomic covalent bonds. Reversing the field causes a negative change in the index as a result of the asymmetrical electric bonding. This is the origin of the Pockels effect [3].

GaAs and InP have cubic $[43m]$ symmetry. Therefore GaAs and InP lack inversion symmetry and they exhibit photorefractive. On the other hand, silicon and germanium do not exhibit handedness because of their cubic $[m\bar{3}m]$ inversion symmetry [4]. Therefore silicon and germanium do not exhibit photorefractive. Note that with high optical intensities and internal electric fields, one could induce photorefractive second order index changes in silicon through the Kerr effect, or else energy level shifts through the Stark effect. However, the Pockels effect requires much less optical power and is

therefore preferred for most applications.

The Pockels effect causes photorefractive media to be relatively efficient nonlinear media [5]. The optical gain and coupling coefficients do not depend on the optical power density. For power densities as low as 1 mW/cm² the photorefractive effect can occur, in contrast to the relatively power hungry Kerr effect which operates through the third-order optical susceptibility χ_3 .

The Pockels effect is described by this relationship between refractive index and electric field:

$$\Delta\left(\frac{1}{n^2}\right) = r_{ijk} E_x \hat{k} \quad \text{and} \quad \Delta n = -\frac{1}{2} r_{ijk} n^3 E_x \hat{k} \quad (1.1)$$

where in a Taylor series expansion it has been assumed that $|\Delta n/n| \ll 1$. In the case of InP:Fe:

n	=	material refractive index	= 3.29
r_{ijk}	=	electro-optic coefficient	= 1.34 pm/V
$E_{sc} \hat{k}$	=	space charge field in direction $\hat{k}$	= 10 kV/cm (typical)

For example, InP:Fe exhibits a peak to peak refractive index change of 4.8×10^{-7} under a modulated 10 kV/cm space charge field. From the coupled wave theory for 2WM, the Pockels effect with an optical standing wave leads to photorefractive optical gain Γ [6]:

$$\Gamma = \frac{2\pi n^3 r_{41} \xi E_x}{\lambda} \quad (1.2)$$

where

ξ = competition factor between electrons and holes, $-1 \leq \xi \leq 1$.

In other words, a periodic space charge field of amplitude 1 kV/cm can introduce a maximum photorefractive gain $\Gamma = 0.282 \text{ cm}^{-1}$. For $\Gamma = 7 \text{ cm}^{-1}$, the amplitude of the periodic photoinduced space charge field must be at least 25 kV/cm. Such substantial periodic steady state fields are required for a practical photorefractive device.

The Pockels effect - and the photorefractive phenomena - are based on the electro-optic effect, which in turn exists if and only if there is the previously discussed asymmetry of the crystal's unit cell. There are practical applications to this behaviour.

Because 2WM is not symmetrical, the optical two-beam coupling experiment is like an optical transistor. For example, if energy from the left hand going beam transfers to the right hand going beam, no energy transfers the other way. The isolation allows one to build photorefractive optical transistors and amplifiers. For example, Kawata et. al. analyze and demonstrate the photorefractive optical transistor's two dimensional action in a BSO crystal[7]. They control cells of alphanumeric letters in an optical beam array, turning individual letters on and off by modulating a reference beam to the BSO crystal. Applications are reviewed in more detail in Chapter 7.

1.2 History

Nonlinear photoinduced refractive index change was first observed as "optical damage" at Bell Labs in 1966 by Ashkin et. al. in LiNbO_3 during experiments in second-harmonic generation [8]. The index change was a nuisance because a few minutes after the laser was turned on, the crystal began to distort the high intensity beam. The laser light had altered the optical properties of the material.

In 1969 Chen reported nonlinear photoinduced refractive index change in LiNbO_3 [9]. Chen correctly identified the problem of "optical damage" as coming from photoexcited electrons drifting out of the illuminated region to deep level traps near the edge of the illuminated region. This is the basic model of the photorefractive effect. Chen observed a refractive index diffraction grating formed by intersecting plane waves in LiNbO_3 , but he saw no optical amplification.

Before 1976, the modeling efforts in photorefraction ignored the effect of the index grating on the intensity distribution of the beams in optical mixing. Then in 1976, Vinetskii et. al. considered the index grating, correctly analyzed the steady state photorefractive diffraction efficiency, and described the process of holographic light amplification[10]. Kukhtarev generalized this work to the transient case[11], deriving the fundamental spatial Fourier harmonic of the steady state space charge field and the complex response time, including build up, decay, and oscillatory response. Between 1976 and 1979, several papers came out from the Kukhtarev group. Then in 1979 Kukhtarev et. al. produced the current coupled wave theory of the photorefractive effect [12] and photorefractive amplification[13]. This theory with some modifications

explains all of the phenomena observed due to the photorefractive effect.

Most of the theoretical work since 1979 has been to extend charge carrier transport models from the original one hop photoexcitation solutions of the Kukhtarev - Vinetskii equations to new formulations where the charge carriers move in multiple photoexcitations and retrappings[14].

1.2.1 Observed Gain

The first observation of optically induced changes in the index of semiconductors was in 1967 when Ashkin et. al. observed CdS[15]. However, this result in CdS was not attributed to the electro-optic effect, but to the Franz-Keldysh effect. In 1984, Glass et. al. [16] and Klein[17] published the first PR experiments in III-V semiconductors. Research has concentrated on GaAs and InP due to their relative availability. In 1990 work was extended to study photorefractive beam coupling in other III-V semiconductors such as CdTe:V[18], and undoped GaP[19].

1.2.1.1 Observed Gain in GaAs

The first observed 2WM differential gain in GaAs was $\Gamma=0.4 \text{ cm}^{-1}$ at $\lambda=1.06 \mu\text{m}$ for zero bias by Klein in 1984 [20]. In 1987 Walsh et. al. reported 2WM gain $\Gamma=0.5 \text{ cm}^{-1}$ in GaAs:Cr with absorption $\alpha=2.2 \text{ cm}^{-1}$ at $\lambda=1.09 \mu\text{m}$ [21]. In 1987 Kumar et. al. reported 2WM gain $\Gamma=0.8 \text{ cm}^{-1}$ in GaAs:Cr using 4 and 7 MHz AC external bias fields to stabilize the gain. Their experimental parameters were $\Lambda=18 \mu\text{m}$, $\lambda=1.064 \mu\text{m}$, $E_0=3.5 \text{ kV/cm}$ [22]. In 1988 Imbert et. al. reported 2WM gain $\Gamma=7 \text{ cm}^{-1}$ (30 dB/cm) in GaAs:Cr with fringes moving at 1 mm/sec, $\Lambda=18 \mu\text{m}$, $\lambda=1.064 \mu\text{m}$, $E_0=16 \text{ kV/cm}$, and beam ratio $\beta>1000$ [23]. In 1989 Walsh et. al. reported 2WM gain $\Gamma=2.3 \text{ cm}^{-1}$ exceeding the absorption loss of 2.2 cm^{-1} using a 780 kHz, 6 kV/cm amplitude AC field for $\lambda=1.09 \mu\text{m}$, $\Lambda=45 \mu\text{m}$, $\beta=30$, and $I_0=10 \text{ mW/cm}^2$ [24].

1.2.1.2 Observed Gain in InP

2WM coupling studies in InP began in 1984 [25], when Glass et. al. observed a diffraction efficiency of $\eta=0.005$. Picoli et. al. predicted 2WM gains of up to 20 cm^{-1} (80 dB/cm) in InP:Fe at room temperature using the photorefractive resonance model [26]. In 1990, Ozkul et. al. reported gains of 11.4 cm^{-1} (50 dB/cm) and 5 cm^{-1} (20

dB/cm) at $\lambda=1.06 \mu\text{m}$ and $1.32 \mu\text{m}$, respectively, using 10 kV/cm dc bias [27]. This exceeds the absorption of $\approx 1 \text{ cm}^{-1}$. In the same year, Miller et. al. reported net gain of 14 cm^{-1} (61 dB/cm) at $\lambda=0.970 \mu\text{m}$ using 10 kV/cm dc bias [28].

In the zero bias case, Strait et. al. observed net gain over absorption in InP:Fe at $\lambda=1.32 \mu\text{m}$ for no bias in 1990 [29], where $\Gamma=0.194 \text{ cm}^{-1} > \alpha=0.145 \text{ cm}^{-1}$.

1.3 Band Gap Engineering

The photorefractive sensitivity of a III-V semiconductor to light is critically dependent on the position of the trapping centers in the band gap. The position influences the optical excitation cross section σ^p of the center and the free carrier trapping cross section σ_n . Figures 1.3 and 1.4 show the positions of most deep levels in InP [30] [31] and GaAs [32], as well as many shallow donors and acceptors. The levels are given in milli electron volts, relative to conduction band and valence band for donors and acceptors, respectively. Band gap engineering allows one to tailor material for specific applications in specific bands by doping with various elements. Transition metals such as Ti, Fe, and Cr are particularly useful as deep levels because they act to make the semiconductors semi-insulating. These deep levels are noteworthy since they make GaAs and InP into low loss substrates suitable for microwave and optical transmission.

In addition to using the deep levels, alloy selection such as in InGaAsP enables one to build semiconductors with exactly the right band gap and deep levels. Multiple quantum well (MQW) structures potentially are useful for band gap engineering the photorefractive effect, but papers on the subject are scant at the time of writing. Pressure [33] can be used to raise the edge of the valence band independent of the conduction band and the deep levels to exactly control the valence band's energy relative to the deep levels and to the conduction band. Finally, material selection gives one great control over the wavelength of photorefractive operation. GaP is useful between 550 and 900 nm; GaAs is useful between 900 and 1300 nm; and InP is useful between 1000 and 1600 nm.

Figure 1.3 Deep level dopants in InP [30] [31].

1.4 Optical Signal Processing

The logic behind doing research into 2WM in InP:Fe and other III-V semiconductors is the promise of coherent optical signal processing functions using power densities as low as 1 mW/cm^2 . Bulk wide band multichannel optical amplifiers are capable of performing well in telecommunications networks. Longer term promise is in optical computing. 12 dB optical gain blocks covering only 3 mm^2 of an opto-electronic hybrid circuit are feasible in opto-electronic integrated circuits. 2WM offers the potential of processing data and images at high speed by using the parallelism inherent to optical components. As in electrical computing, one implements an optical computer with a few basic components for data processing such as holographic interconnects, spatial light modulators, optical memories, mass storage, and optical transistors. The optical control components require optically nonlinear materials, just as electrical control components require electrically nonlinear materials. 2WM in photorefractive material such as InP and GaAs can provide these components. 2WM has been observed in GaAs and InP since 1984 [34]. These III-V semiconductors and their alloys are already the industrial basis of high speed electrical integrated circuits. One could build on established industry production line

techniques for GaAs and InP to make all of the optical structures required for an optical computer.

1.5 Thesis Organization

Chapter 1 is the introduction. It reviews the logic behind research into photorefractives. Photorefractive research is based on the promise of cost effective, high speed, low power optical signal processing. The photorefractive effect that leads to 2WM is described. The introduction concludes with a short history of 2WM and the photorefractive effect.

Chapter 2 reviews the general Kukhtarev model. It is basis of the current understanding of the photorefractive effect.

Chapter 3 describes deep level absorption in InP:Fe, and explains how it drives 2WM. A technique is developed to estimate deep level concentrations from InP absorption spectra. The demonstration is unique in the available literature in that it is the

Figure 1.4 Deep level dopants in GaAs [32].

first time that absorption spectra have been used in this way to evaluate deep level concentrations and charge species ratios.

Chapter 4 moves on from the theory of optical absorption to review solutions to the Kukhtarev model for response time τ and gain Γ in optical mixing amplifiers. The significance of the phase shift between the intensity grating and the phase grating is explained. Since not all of the material parameters were measured, some assumptions are made about charge carrier mobility, impurity concentrations, and the uniformity of the InP:Fe sample under study.

Chapter 5 uses numerical finite element modeling to observe the dynamic formation of the photorefractive grating, and predicts photorefractive gain for series of external biases, grating spacings, and modulation depths. The simulation is performed with the commercially available program called **PISCES**.

Chapter 6 reports experiments performed during the thesis. The two primary experimental set-ups are discussed. The experiments measure 2WM gain Γ and response time τ . Thirty decibels per centimeter optical gain is observed. Observations and models for the results are discussed, and compared with others' work.

Chapter 7 presents applications for 2WM. The applications are divided into two main groups: telecommunications applications and optical computing applications. For telecommunications, the optical signal amplification capability of 2WM implemented in different venues dominates the various potential devices. For optical computing, 2WM finds application to devices for optical memory and image processing.

Chapter 8 concludes the thesis with overall results and some statements about future directions for research. The work is not unique because similar results have been found before by others in InP:Fe and other III-V semiconductors.

CHAPTER 2

KUKHTAREV MODEL

This chapter considers the basic model of the photorefractive effect. As described in the Introduction, Kukhtarev's model assumes a single charge carrier in a photorefractive medium such as LiNbO_3 . In general for semiconductors, one must consider two carriers: electrons and holes. Strohkendl et. al. extend the equations of Kukhtarev to include both electrons and holes. He obtains a set of differential equations for the periodic space charge field E_{sc} formed by the optical interference pattern $I(x)$ inside the crystal [35]. This chapter follows Strohkendl's extension, discusses Picoli's thermal model, and discusses the effective electro-optic coefficient in III-V semiconductors as a function of signal polarization.

2.1 Kukhtarev Model including Electrons and Holes

In any sample of III-V semiconductor such as InP:Fe , there is a background of shallow impurities concentrated near the edges of the conduction and valence bands. Let N_A be the density of shallow acceptors, and N_D be the density of shallow donors. Typically, nominally pure InP may have concentrations of $N_A \approx 10^{14} \text{ cm}^{-3}$ and $N_D \approx 10^{15} \text{ cm}^{-3}$ - an n-type material. In the absence of optical excitation and deep level traps, the steady state electron density is:

$$n_e = N_D - N_A = 10^{15} - 10^{14} = 9 \times 10^{14} \text{ cm}^{-3} \quad (2.1)$$

and since the intrinsic concentration $n_i = 1.2 \times 10^8 \text{ cm}^{-3}$ at 293°K , the hole concentration is about 16 cm^{-3} - negligibly small.

$$n_h = \frac{n_i^2}{n_e} = 16 \text{ cm}^{-3} \quad (2.2)$$

At room temperature the shallow levels are completely ionized and do not contribute trap levels to the photorefractive model [36].

The model for charge concentrations is more complicated in general because iron sites present in all InP boules form deep level traps. The iron compensates for the electron donors by absorbing free electrons and liberating holes. A large enough iron concentration compensates for all donors and renders InP:Fe semi-insulating. Let $[\text{Fe}]$ be the total

concentration of the deep level iron dopant. The iron sites are divided into two distinct mid-gap species with distinct optical and electrical characteristics: electrically neutral donor sites, $[Fe^{+2}]$, and electrically positive acceptor sites $[Fe^{+3}]$. $[Fe] \equiv [Fe^{+2}] + [Fe^{+3}]$. In the dark, the two species continuously change charge state. The populations are in dynamic steady state with each other in two balanced reactions:

The $[Fe^{+2}]/[Fe^{+3}]$ ratio is determined by the shallow donor concentrations, and temperature, and optical intensity. It will be analytically expressed and measured later.

When light is introduced in the crystal, the rate equation for ionized iron dopant Fe^{+3} is:

$$\frac{\partial [Fe^{+3}]}{\partial t} = \sigma_n^o I [Fe^{+2}] - \gamma_e n_e [Fe^{+3}] - \sigma_p^o I [Fe^{+3}] + \gamma_h n_h [Fe^{+2}] \quad (2.4)$$

where

- I = spatially periodic optical intensity
- σ_p^o, σ_n^o = optical cross-section for electrons, holes
- γ_e, γ_h = recombination coefficients for electrons, holes
- n_e, n_h = densities of free electrons, holes

The rate equation for $[Fe^{+3}]$ says that the rate of creation of Fe^{+3} is dependent on four competing processes, represented by the four terms in the equation.

Term 1: $\sigma_n^o I [Fe^{+2}]$ is the rate that Fe^{+2} is converted into Fe^{+3} via photogeneration of an electron:

Term 2: $-\gamma_e n_e [Fe^{+3}]$ is the rate that Fe^{+3} is converted into Fe^{+2} via electron recombination:

Term 3: $-\sigma_p^o I [Fe^{+3}]$ is the rate that Fe^{+3} is converted into Fe^{+2} via photogeneration of a hole:

Term 4: $\gamma_h n_h [Fe^{+2}]$ is the rate that Fe^{+2} is converted into Fe^{+3} via hole recombination:

These reactions will be discussed more completely in Chapter 3 (absorption) and Chapter 4 (recombination time).

The current densities for electrons j_e and holes j_h are:

$$j_e = en_e\mu_e E + \mu_e k_B T \nabla n_e \quad (2.9)$$

$$j_h = en_h\mu_h E - \mu_h k_B T \nabla n_h \quad (2.10)$$

where the drift and diffusion terms are the first and second terms. The variables are:

μ_e, μ_h = mobilities for electrons, holes
 E = vector electric field
 k_B = Boltzmann's constant
 T = temperature

The continuity equations for electrons and holes considering current, optical absorption, and recombination are:

$$\frac{\partial n_e}{\partial t} = \frac{\nabla j_e}{e} + \sigma_a I [Fe^{+2}] - \gamma_e n_e [Fe^{+3}] \quad (2.11)$$

$$\frac{\partial n_h}{\partial t} = -\frac{\nabla j_h}{e} + \sigma_p I [Fe^{+3}] - \gamma_h n_h [Fe^{+2}] \quad (2.12)$$

The Poisson equation considering shallow donors and deep level traps is:

$$\nabla E(x,t) = \nabla^2 V(x,t) = -\frac{\rho(x,t)}{e} = \frac{e}{e} (N_D - N_A + n_h(x,t) - n_e(x,t) + [Fe^{+3}](x,t)) \quad (2.13)$$

where time varying parameters are indicated and where $[Fe^{+2}](x,t)$ is not included in the expression because Fe^{+2} is a neutral site and so does not contribute to the charge density. Imposing a fixed external voltage V_o across a sample of length L introduces a boundary condition because the electric field in the crystal must contribute to a fixed applied bias $V_o/L = E_o$:

$$E_o = \frac{V_o}{L} = \int_0^L E(x,t) dx \quad (2.14)$$

Assuming a one dimensional variation in optical intensity $I(x)$ and a constant external electric field in the x direction E_o , the above equations reduce to a set of seven equations in seven unknowns: $n_e(x,t)$, $n_h(x,t)$, $[Fe^{+3}](x,t)$, $[Fe^{+2}](x,t)$, $j_e(x,t)$, $j_h(x,t)$, and $E(x,t)$. In the steady state regime $d/dt=0$, reducing the equations even further. The equations can either be solved

analytically with many limiting approximations as outlined here in Chapter 2 and later on in Chapter 4, or else numerically as in Chapter 5.

2.2 Standard Model for E_{sc} and τ

Most analytical models of the photorefractive effect start with a sinusoidal optical interference pattern $I(x)$ and an optical modulation index m :

$$I(x) = I_0(1 + me^{ikx}) \text{ where } m = \frac{2(I_s I_p)^{1/2}}{I_0} = \frac{2}{\beta^{1/2}} \text{ and } k = \frac{2\pi}{\Lambda} \quad (2.15)$$

where I_s and I_p are the intensities of the copolarized signal and pump, β is the ratio of the pump to the signal intensity, I_0 is the average optical intensity, and Λ is the photorefractive grating spacing set by the angle of beam intersection 2θ and the free space wavelength λ :

$$\Lambda = \frac{\lambda}{2\sin\theta} \quad (2.16)$$

For example, where the pump beam is 10^2 times more intense than the signal beam, the modulation index $m=0.2$.

Strohkendl et. al. [37] make two assumptions. First, they assume through the quasi-CW illumination approximation that one may neglect the time derivatives of the two continuity equations. Quasi-CW illumination means that the time to establish or erase a grating is much longer than the free carrier lifetimes. Second, they assume that optical intensity is small enough so that free carrier densities are not large enough to significantly effect $E(x,t)$. For this to be true, optical intensity must be less than two limits. The limits are set by the lifetime and the absorption coefficient due to each carrier. After Strohkendl:

$$I_0 < \frac{4\pi^2 h \nu e k_B T}{\Lambda^2 e^2 \tau_e \alpha_e} \text{ for electrons, and } I_0 < \frac{4\pi^2 h \nu e k_B T}{\Lambda^2 e^2 \tau_h \alpha_h} \text{ for holes} \quad (2.17)$$

In Chapter 3 this thesis investigates the role of optical absorption in InP:Fe. A total absorption of 0.93 cm^{-1} is measured at $\lambda=1.064 \text{ }\mu\text{m}$. Based on fitting the absorption spectrum, $[\text{Fe}^{+2}]/[\text{Fe}]=0.29$ and $[\text{Fe}^{+3}]/[\text{Fe}]=0.71$. Therefore $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]=0.41$. Since $\sigma_p^0 \approx 10.7\sigma_n^0$, the optical absorption coefficients in the InP:Fe sample break down as $\alpha_e \approx 0.034 \text{ cm}^{-1}$ and $\alpha_h \approx 0.896 \text{ cm}^{-1}$. The carrier lifetimes are found to be $\tau_e \approx 500 \text{ psec}$ and $\tau_h \approx 2 \text{ }\mu\text{sec}$. Therefore I_0 at $\lambda=1.064 \text{ }\mu\text{m}$ is limited in this model by hole photoexcitation to $70,000 \text{ mW/cm}^2$. The limit is not approached in any known experimental paper on

photorefractive InP:Fe. This is because absorptive heating causes large temperature fluctuations in InP:Fe for CW experiments whenever $I_0 \geq 1000 \text{ mW/cm}^2$.

Since the driving optical forcing function is sinusoidal with a small modulation index, one assumes that the spatial variation of all other quantities is also sinusoidal, with a DC term and a time varying phase and magnitude. The DC terms are labelled sub 0 (zero) for the photogenerated free carrier densities:

$$n_{e0} = \frac{\sigma_n \sigma I_0 [Fe^{+2}]_0}{\gamma_e [Fe^{+3}]_0} \quad \text{and} \quad n_{h0} = \frac{\sigma_p \sigma I_0 [Fe^{+3}]_0}{\gamma_h [Fe^{+2}]_0} \quad (2.18)$$

As a result, where $k=2\pi/\Lambda$, the magnitude of the fundamental component of E_{sc} , $E_1(t)$, varies with time according to:

$$\begin{aligned} \frac{dE_1(t)}{dt} = E_1(t) & \left(\frac{n_{e0} e \mu_e}{e} \frac{1 + \frac{k(k-iV)}{k_0^2}}{1 + \frac{k(k-iV)}{K_e^2}} + \frac{n_{h0} e \mu_h}{e} \frac{1 + \frac{k(k+iV)}{k_0^2}}{1 + \frac{k(k+iV)}{K_h^2}} \right) \\ & + i m \frac{k_B T}{e} \left(\frac{n_{e0} e \mu_e}{e} \frac{k-iV}{1 + \frac{k(k-iV)}{K_e^2}} - \frac{n_{h0} e \mu_h}{e} \frac{k+iV}{1 + \frac{k(k-iV)}{K_h^2}} \right) \end{aligned} \quad (2.19)$$

where

$$K_e^{-2} = \frac{k_B T \mu_e}{e \gamma_e [Fe^{+3}]_0} \quad \text{and} \quad K_h^{-2} = \frac{k_B T \mu_h}{e \gamma_h [Fe^{+2}]_0} \quad (2.20)$$

$$k_0 = \frac{e^2 [Fe^{+2}]_0 [Fe^{+3}]_0}{e k_B T [Fe]} \quad \text{and} \quad V = \frac{e E_0}{k_B T} \quad (2.21)$$

$1/K_e$ and $1/K_h$ are the average distances traveled by the electrons and the holes between excitation and recombination. Here, the recombination coefficients and the mobilities are from [Chapter 4](#):

$$\begin{aligned}
\gamma_e &= 2.5 \times 10^{-9} \text{ cm}^3/\text{s} \\
\gamma_h &= 2.0 \times 10^{-10} \text{ cm}^3/\text{s} \\
\mu_e &= 1470 \text{ cm}^2/(\text{V}\cdot\text{s}) \\
\mu_h &= 140 \text{ cm}^2/(\text{V}\cdot\text{s}) \\
[\text{Fe}^{+2}]_0 &= [\text{Fe}^{+3}]_0 = 0.5 \times 10^{16} \text{ cm}^{-3}
\end{aligned}$$

and so the average charge carrier travel ranges are 17.5 μm and 19.1 μm at room temperature for electrons and holes, respectively. k_0^{-1} is the Debye screening length, 0.083 μm . V is a measure of the carrier drift, equal to 38.5 μm^{-1} for $E_0 = 10 \text{ kV/cm}$.

The method of undetermined coefficients yields the particular solution to the differential equation. The steady state magnitude and phase of E_{sc} is results. The homogeneous solution to the differential equation yields the time constant and the period of any oscillations. In general, the fundamental component of E_{sc} is E_1 :

$$E_1 = -i m k_0^2 \frac{k_B T}{e} \frac{1 - \frac{\sigma_p^0 [\text{Fe}^{+3}]_0}{\sigma_n^0 [\text{Fe}^{+2}]_0} \frac{k+iV}{k-iV} \frac{K_i^2 + k(k-iV)}{K_h^2 + k(k-iV)}}{\frac{k_0^2 + k(k-iV)}{k-iV} + \frac{\sigma_p^0 [\text{Fe}^{+3}]_0}{\sigma_n^0 [\text{Fe}^{+2}]_0} \frac{k_0^2 + k(k+iV)}{K_h^2 + k(k+iV)} \frac{K_i^2 + k(k-iV)}{k-iV}} \quad (2.22)$$

with time constant τ :

$$\tau = \frac{\frac{h\nu}{I_0 [\text{Fe}]}}{\left[\frac{\sigma_n^0}{[\text{Fe}^{+3}]_0} \frac{k_0^2 K_i^2 + k_0^2 k^2 + k^2 K_i^2 + k^4 + k^2 V^2}{K_i^4 + 2k^2 K_i^2 + k^4 + k^2 V^2} + \frac{\sigma_p^0}{[\text{Fe}^{+2}]_0} \frac{k_0^2 K_h^2 + k_0^2 k^2 + k^2 K_h^2 + k^4 + k^2 V^2}{K_h^4 + 2k^2 K_h^2 + k^4 + k^2 V^2} \right]} \quad (2.23)$$

and a damped oscillatory period T :

$$T = \frac{\frac{2\pi h\nu}{I_0 [\text{Fe}]}}{\left[\frac{\sigma_n^0}{[\text{Fe}^{+3}]_0} \frac{k_0^2 kV - kK_i^2 V}{K_i^2 + 2k^2 K_i^2 + k^4 + k^2 V^2} + \frac{\sigma_p^0}{[\text{Fe}^{+2}]_0} \frac{k_0^2 kV - kK_h^2 V}{K_h^2 + 2k^2 K_h^2 + k^4 + k^2 V^2} \right]} \quad (2.24)$$

Through the Pockels effect, this solution for a periodic electric field results in a periodic modulation in the refractive index of the medium. The ensuing two-beam optical coupling

leads to one beam being depleted, and the other beam being increased in intensity. In **Chapter 4** it is shown that in the steady state, for a small enough optical signal relative to the pump, the signal's intensity inside the crystal changes according to:

$$I_s(z) = I_s(0)e^{\Gamma - \alpha z} \text{ where } \Gamma = \frac{2\pi n^3 r_{eff}}{\lambda} \xi E_{sc} \quad (2.25)$$

$\xi(I_0)$ is the reduction factor $-1 \leq \xi \leq 1$ due to two-band coupling, where electrons and holes interfere with each other with the net result of screening and reducing the electric field. At high temperatures, $|\xi| < 1$, and thermal carriers wash out 2WM:

$$\xi(I_0) = \frac{I_0 (\sigma_p^o [Fe^{+3}] - \sigma_n^o [Fe^{+2}])}{h\nu (e_{po} [Fe^{+3}] + e_{no} [Fe^{+2}])} \quad (2.26)$$

where e_{no} and e_{po} are the electron and hole thermal emission rates plus the optical emission rates per donor site:

$$e_{no} = e_n^{th} + \sigma_n^o I_0 \quad (2.27)$$

$$e_{po} = e_p^{th} + \sigma_p^o I_0 \quad (2.28)$$

The gain Γ may be positive, zero, or negative, depending on the crystal orientation as discussed below in the index ellipsoid section. Only when Γ exceeds absorptive loss α can there be net gain in the crystal. Under zero bias conditions, E_{sc} is limited by the diffusion field E_D and the trap density space charge field E_q :

$$\frac{1}{E_{sc}} = \frac{1}{E_D} + \frac{1}{E_q} \quad (2.29)$$

E_D is the steady state value of the space charge field formed just by diffusion:

$$E_D = \frac{2\pi k_B T}{e\Lambda} \quad (2.30)$$

E_q is the maximum field that the crystal can support when all of the available deep level charges are separated from N_E fixed deep levels by $\Lambda/2$:

$$E_q = \frac{eN_E\Lambda}{2\pi e} \text{ and } N_E = \frac{[Fe^{+2}][Fe^{+3}]}{[Fe]} \quad (2.31)$$

In general E_{sc} is small when there is no applied bias, resulting in low gain ($\approx 0.2 \text{ cm}^{-1}$). Applying bias transports more free carriers away from the deep levels and much larger

values of E_{sc} and gain ($\approx 7 \text{ cm}^{-1}$) are found.

2.3 Thermal Model

The model in the last section does not take thermal generation of carriers into account. It turns out that at room temperature, the thermal generation rate of electrons in InP:Fe is close to the optical generation rate of holes when $I_0 \approx 10 \text{ mW/cm}^2$. Picoli et. al. [38] consider this thermal effect for the case of negligible injection and weak illumination. Negligible injection is the condition where the applied external bias voltage produces a constant and uniform current in the crystal, resulting in a uniform analytical electric field across the sample. This is true for InP:Fe for $E_0 \leq 10 \text{ kV/cm}$, but not true for all semiconductors, such as GaAs:Cr.

Under weak enough illumination and negligible injection, the Kukhtarev equations are linearized by assuming that the deviations from DC values of terms are small and the first Fourier components are linear in I_0 . Picoli found that the component of space charge field E_{sc} at phase angle $\psi = 90^\circ$ to the optical intensity is:

$$E_1 = \text{Im}(E_{sc}) = i m I_0 \frac{\sigma_p^0 [Fe^{+3}] - \sigma_n^0 [Fe^{+2}]}{e_{no} [Fe^{+2}] \left(\frac{1}{E_q} + \frac{1}{E_D - iE_0} \right) + e_{po} [Fe^{+3}] \left(\frac{1}{E_q} + \frac{1}{E_D - iE_0} \right)} \quad (2.32)$$

where the imaginary factor "i" is used to designate the 90° phase shift. This form allows a simple discussion of the mechanism behind the temperature-intensity resonance of the 90° spatially shifted component of E_{sc} . However, the equation is valid only for $\Lambda < 5 \mu\text{m}$ due to simplifying approximations. The equation is a generalized form for E_{sc} originally given by Kukhtarev, since setting $\sigma_n^0 = 0$, $e_{no} = 0$, and in the limit as I_0 goes to infinity, E_{sc} above reduces to:

$$E_{sc} = i m \frac{1}{\frac{1}{E_q} + \frac{1}{E_D + iE_0}} \quad (2.33)$$

Picoli found that E_{sc} consists of real and imaginary parts. The real part is in phase with the optical standing wave, and the imaginary part is 90° out of phase. When the rates of electron and hole creation are equal, the real part disappears. In general, the imaginary

component of E_x is:

$$E_x = i m \xi(I_o) \frac{A(E_o, k)}{A^2(E_o, k) + \left(\frac{e_{no}[Fe^{+2}] - e_{po}[Fe^{+3}]}{e_{no}[Fe^{+2}] + e_{po}[Fe^{+3}]} \right) B^2(E_o, k)} \quad (2.34)$$

where:

$$A(E_o, k) = \frac{1}{E_o} + \frac{E_D}{E_o^2 + E_D^2} \quad (2.35)$$

$$B(E_o, k) = \frac{E_o}{E_o^2 + E_D^2} \quad (2.36)$$

2.4 Electro-Optics

The electro-optic tensor is a 6x3 matrix which relates the vector electric field to a change in refractive index: $(\Delta(1/n^2))_i = R_{ij} E_j$. The electro-optic tensor "R" for III-V semiconductors has three equal non-zero elements r_{41} :

$$R = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{41} \end{pmatrix} \quad (2.37)$$

where the axes are the principal 43m crystal axes, $[001]$, $[110]$, and $[1\bar{1}0]$.

2.4.1 Effective Electro-Optic Coefficient

The effective electro-optic coefficient r_{eff} determines the beam coupling coefficient Γ , as shown later on in [Chapter 4](#). It is different from the III-V semiconductor electro-optic coefficient r_{41} . The effective electro-optic coefficient r_{eff} is a tensor function of the crystal orientation, the direction of the space charge field $\mathbf{R}$, and the polarization

vectors of the signal and pump beams $\hat{e}_1$ and $\hat{e}_2$. r_{eff} changes the refractive index in the crystal according to:

$$n(x,y,z) = n_o + \Delta n = n_o + \frac{1}{2} r_{\text{eff}} n_o^3 E(x,y,z) \quad (2.38)$$

where equation (1.1) was used. Two principal geometries were studied in the thesis: $\hat{k}$ in [001] and $\hat{k}$ in [110]. Both geometries are depicted in Figure 2.1. The shaded box represents the photorefractive crystal. ϕ is the angle of polarization from s-polarization. θ is the half angle of intersection of the beams inside the crystal. r_{eff} is given by [39]:

$$r_{\text{eff}} = \hat{e}_2^* \cdot \mathbf{e}, [R \cdot \hat{k}] \cdot \mathbf{e}, \hat{e}_1 \quad (2.39)$$

which is commonly used but less commonly explained. It is derived later on in Section 4.2 of this thesis. From the same reference, R can be expressed as a sum of direct tensor, or Kronecker products using the crystal's unit vectors $\hat{x}$, $\hat{y}$, and $\hat{z}$:

$$R = r_{41}(\hat{x} \otimes \hat{y} \otimes \hat{z} + \hat{x} \otimes \hat{z} \otimes \hat{y} + \hat{y} \otimes \hat{x} \otimes \hat{z} + \hat{y} \otimes \hat{z} \otimes \hat{x} + \hat{z} \otimes \hat{x} \otimes \hat{y} + \hat{z} \otimes \hat{y} \otimes \hat{x}) \quad (2.40)$$

Since electric field vector $\mathbf{E}$ is parallel or anti-parallel to $\hat{z}$ in both [001] and [110] geometries, the expression can be simplified:

$$\begin{aligned} (\hat{x} \otimes \hat{y} \otimes \hat{z}) \cdot \hat{k} &= (\hat{x} \otimes \hat{y})(\hat{z} \cdot \hat{k}) \\ &= \pm \hat{x} \otimes \hat{y} \end{aligned} \quad (2.41)$$

where the sign is positive for [001] and negative for [110]. As a result, $R \cdot \hat{k}$ in the equation for r_{eff} is:

$$\begin{aligned} R \cdot \hat{k} &= r_{41}(\hat{x} \otimes \hat{y} + \hat{y} \otimes \hat{x}) \\ &= r_{41} \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix}_{\hat{k} = [001]} = \frac{r_{41}}{\sqrt{2}} \begin{pmatrix} 0 & 0 & -1 \\ 0 & 0 & 1 \\ -1 & 1 & 0 \end{pmatrix}_{\hat{k} = [110]} \end{aligned} \quad (2.42)$$

Referring to Figure 2.1, the polarization unit vectors $\hat{e}_1$ and $\hat{e}_2$ for $\hat{k}$ in [001] are:

$$\hat{e}_1 = \begin{pmatrix} -\sin(\phi)\sin(\theta) \\ +\cos(\phi) \\ +\sin(\phi)\cos(\theta) \end{pmatrix} \quad \hat{e}_2 = \begin{pmatrix} +\sin(\phi)\sin(\theta) \\ +\cos(\phi) \\ +\sin(\phi)\cos(\theta) \end{pmatrix} \quad (2.43)$$

Figure 2.1 - Two-wave mixing geometries for $\hat{k}$ in $[001]$ and $[110]$ directions.

and the polarization unit vectors $\hat{e}_1$ and $\hat{e}_2$ for $\hat{k}$ in $[110]$ are:

$$\hat{e}_1 = \begin{pmatrix} -\sin(\phi)\sin(\theta) \\ -\cos(\phi) \\ -\sin(\phi)\cos(\theta) \end{pmatrix} \quad \hat{e}_2 = \begin{pmatrix} +\sin(\phi)\sin(\theta) \\ -\cos(\phi) \\ -\sin(\phi)\cos(\theta) \end{pmatrix} \quad (2.44)$$

As a result of the explicit expressions for $\hat{k}$, $\hat{e}_1$ and $\hat{e}_2$, and since ϵ_r terms are diagonal with three equal nonzero terms each equal to the refractive index n_o^2 , the equations for $r_{\text{eff}}(\phi, \theta)$ reduce to:

$$\begin{aligned} r_{\text{eff}} &= -r_{41}n_o^4 (\sin^2(\phi)\sin^2(\theta) + \cos^2(\phi)) \quad \text{for } \hat{k} = [001] \\ &= \frac{r_{41}n_o^4}{\sqrt{2}} \sin(2\phi)\cos(\theta) \quad \text{for } \hat{k} = [110] \end{aligned} \quad (2.45)$$

The equations for $r_{\text{eff}}(\phi, \theta)$ are useful for predicting angles at which r_{eff} and consequently two wave coupling is maximized. In the case $\hat{k} = [001]$, r_{eff} is maximized when $\phi = 0^\circ$, that is, both pump and signal beams are s-polarized. In the case $\hat{k} = [110]$, r_{eff} is maximized when $\phi = 45^\circ$, that is, both pump and signal beams are slanted at 45° relative to s-polarized light. The normalized values from the theoretical functions, r_{eff}/r_{41} , are depicted as a function of ϕ in Figure 2.2, with θ as a parameter. θ is 0° (co-propagating beams), 45° (orthogonal beams), and 90° (counter-propagating beams).

2.4.2 Index Ellipsoid

The crystal orientation and the imposed "low-frequency" electric field combine to form a three dimensional dielectric surface known as an index ellipsoid. The index ellipsoid determines the refractive index change seen by a light beam, depending on the polarization angle ϕ and the angle of incidence θ .

InP is an optically isotropic crystal with a spherical constant energy surface. D_k is the electric displacement vector in direction k . E_l is the electric field in direction l . D_k and E_l are related by the tensor:

$$D_k = \epsilon_{kl} E_l \quad (2.46)$$

where ϵ_{kl} is the kl^{th} element of the dielectric tensor $\epsilon_o \epsilon_r$. Considering the stored energy

density w_e in D space, the constant energy surface is [40]:

$$\frac{D_x^2}{\epsilon_x} + \frac{D_y^2}{\epsilon_y} + \frac{D_z^2}{\epsilon_z} = 2w_e \epsilon_0 \quad (2.47)$$

where ϵ_x , ϵ_y , and ϵ_z are the relative dielectric constants on the principal axes of the crystal. The index ellipsoid formula results by replacing $D/(2w_e \epsilon_0)^{1/2}$ with r and setting $\epsilon_x = n_x^2$:

$$\frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1 \quad (2.48)$$

In InP:Fe and other III-V semiconductors, the photorefractive effect changes the index ellipsoid through the space charge field from the degenerate $n_x = n_y = n_z$ spherical condition to a nondegenerate ellipsoid with unequal axes. The principal axes change both in length and in direction, depending on the direction of the applied electric field in the crystal's coordinate system. The equation of the index ellipsoid in an electric field is:

$$\left(\frac{1}{n^2}\right)_1 x^2 + \left(\frac{1}{n^2}\right)_2 y^2 + \left(\frac{1}{n^2}\right)_3 z^2 + \left(\frac{1}{n^2}\right)_4 yz + \left(\frac{1}{n^2}\right)_5 xz + \left(\frac{1}{n^2}\right)_6 xy = 1 \quad (2.49)$$

Figure 2.2 - Effective normalized electro-optic coefficient r_{eff}/r_{41} as a function of polarization angle ϕ .

where changes in the constants $(1/n^2)_{1,2,3...6}$ are defined by the product of the electro-optic tensor R and the vector electric field:

$$\begin{pmatrix} \Delta\left(\frac{1}{n^2}\right)_1 \\ \Delta\left(\frac{1}{n^2}\right)_2 \\ \Delta\left(\frac{1}{n^2}\right)_3 \\ \Delta\left(\frac{1}{n^2}\right)_4 \\ \Delta\left(\frac{1}{n^2}\right)_5 \\ \Delta\left(\frac{1}{n^2}\right)_6 \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{41} \end{pmatrix} \times \begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ r_{41}E_x \\ r_{41}E_y \\ r_{41}E_z \end{pmatrix} \quad (2.50)$$

For $\mathbf{R}=[001]$, $E_x=E_y=0$ and $E_z=E$. The resulting index ellipsoid is:

$$\frac{x^2 + y^2 + z^2}{n_o^2} + 2r_{41}E xy = 1 \quad (2.51)$$

which is not in canonical ellipsoid form. To get into the standard form, the variables are changed from $\{x\}$ to $\{x'\}$ by rotating $+45^\circ$ around the z axis using the transformation:

$$\begin{pmatrix} x' \\ y' \\ z' \end{pmatrix} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 & 0 \\ -1 & 1 & 0 \\ 0 & 0 & \sqrt{2} \end{pmatrix} \times \begin{pmatrix} x \\ y \\ z \end{pmatrix} \quad (2.52)$$

The transform converts the ellipsoid into canonical form:

$$\left(\frac{1}{n_o^2} + r_{41}E\right)x'^2 + \left(\frac{1}{n_o^2} - r_{41}E\right)y'^2 + \frac{1}{n_o^2}z'^2 = 1 \quad (2.53)$$

Comparing this expression to the standard form and using a Taylor series approximation gives the refractive index accessed by light polarized in the direction of the new set of principal axes $\{x'\}$ for $\mathbf{R}=[001]$:

$$\begin{aligned} n_{x'} &= n_o - \frac{1}{2}r_{41}n_o^3E \\ n_{y'} &= n_o + \frac{1}{2}r_{41}n_o^3E \\ n_{z'} &= n_o \end{aligned} \quad (2.54)$$

The same sort of derivation applies for $\mathbf{R}=[110]$, $E_x=E/\sqrt{2}$, $E_y=E/\sqrt{2}$, and $E_z=0$. The resulting index ellipsoid is:

$$\frac{x^2 + y^2 + z^2}{n_o^2} + \sqrt{2}r_{41}E(yz + xz) = 1 \quad (2.55)$$

which is not in canonical ellipsoid form. To get into the standard form, two rotations are required. First, the variables are changed from $\{x\}$ to $\{x'\}$ by rotating $+45^\circ$ around the z axis. Next, $\{x'\}$ is transformed to $\{x''\}$ by rotating $+45^\circ$ around y' . The transformation operation is:

$$\begin{pmatrix} x'' \\ y'' \\ z'' \end{pmatrix} = \frac{1}{2} \begin{pmatrix} 1 & 1 & \sqrt{2} \\ -\sqrt{2} & \sqrt{2} & 0 \\ -1 & -1 & \sqrt{2} \end{pmatrix} \times \begin{pmatrix} x \\ y \\ z \end{pmatrix} \quad (2.56)$$

The transform converts the ellipsoid into canonical form:

$$\left(\frac{1}{n_o^2} + r_{41}E\right)x''^2 + \frac{1}{n_o^2}y''^2 + \left(\frac{1}{n_o^2} - r_{41}E\right)z''^2 = 1 \quad (2.57)$$

Comparing this expression to the standard form and using a Taylor series approximation gives the refractive index accessed by light polarized in the direction of the new set of principal axes $\{x''\}$ for $\mathbf{R}=[110]$:

$$\begin{aligned}
 n_x'' &= n_o - \frac{1}{2}r_{41}n_o^3E \\
 n_y'' &= n_o \\
 n_z'' &= n_o + \frac{1}{2}r_{41}n_o^3E
 \end{aligned}
 \tag{2.58}$$

2.4.3 Ellipsoid Intersection with Polarization Vector

The index ellipsoid by itself is not enough to indicate the effective amplitude of the refractive index grating presented to light with arbitrary polarization and direction. The polarization unit vector $\hat{e}_1$ multiplied by some constant A will intersect the index ellipsoid, where A is equal to the refractive index presented to the light with polarization ϕ and direction θ . The intersection of the polarization vector with the ellipsoid defines $n(\phi, \theta)$ for $\mathbf{R}=[001]$ as:

$$n(\phi, \theta) = n_o + \frac{r_{41}n_o^3E}{2} \sin(2\phi)\sin(\theta) \tag{2.59}$$

where the index variation is a maximum for $\phi=45^\circ$. The variation of the normalized accessed refractive index $\Delta n/(n_o r_{41})$ is depicted in Figure 2.3 for 1 kV/cm electric field. The result disagrees with the effective electro-optic coefficient r_{eff} above, which indicates maximum beam coupling for $\phi=0^\circ$. The disagreement appears to be caused by $n(\phi, \theta)$ coming from just a consideration of $\hat{e}_1$, instead of both $\hat{e}_1$ and $\hat{e}_2$ as is the case with r_{eff} . The beam coupling involves more than just one beam diffracting off the phase grating. The second beam contributes to the value of r_{eff} and does not show up in $n(\phi, \theta)$.

For $\mathbf{R}=[110]$, $n(\phi, \theta)$ is:

$$n(\phi, \theta) = n_o - \frac{r_{41}n_o^3E}{2\sqrt{2}} (\sin(2\phi)\cos(\theta) + \sin^2(\phi)\sin(2\theta)) \tag{2.60}$$

where the index variation is a maximum for $\phi=45^\circ$ for $\theta=0^\circ$. The value of ϕ corresponding to the maximum value of $n(\phi, \theta)$ increases to $\phi=62.6^\circ$ for $\theta=45^\circ$ and goes to $\phi=67^\circ$ for $\theta=90^\circ$. The result agrees with r_{eff} above, in that maximum beam coupling exists nominally for $\phi=45^\circ$, since nominally $\theta=0$. The form of $n(\phi, \theta)$ differs from r_{eff} in that $n(\phi, \theta)$ has two trigonometric terms and r_{eff} has only one term. The difference comes from the difference

between the meanings of $n(\phi, \theta)$ and r_{eff} as described above for $\mathbf{R}=[001]$.

$\Delta n(\phi, \theta)$ and $r_{\text{eff}}(\phi, \theta)$ may be anywhere between in phase and 90° out-of-phase with each other. The out-of-phase condition is clearly seen comparing Figure 2.2 and Figure 2.3. In other words, $\Delta n(\phi, \theta)$ and $r_{\text{eff}}(\phi, \theta)$ are independent of each other. The independence of $\Delta n(\phi, \theta)$ and $r_{\text{eff}}(\phi, \theta)$ indicates that $\Delta n(\phi, \theta)$ is not important to the photorefractive effect in III-V semiconductors, although both are proportional to r_{41} and the electric field. The relative importance of $r_{\text{eff}}(\phi, \theta)$ can be seen from Figure 2.2. The effective electro-optic coefficient is multiplied by a factor of typically 100. $\Delta n(\phi, \theta)/n_o$, on the other hand, is maximized near 5×10^{-13} .

2.5 Summary

This chapter presented the Kukhtarev theory for photorefraction. Using Strohkendl et. al.'s techniques, solutions for the time constant and the fundamental component of the space periodic electric field are given. There was some discussion of

Figure 2.3 - Accessed refractive index for arbitrary polarization angle ϕ for $\mathbf{R}=[001]$ and $\mathbf{R}=[110]$.

thermal effects, and the effective electro-optic coefficient was calculated for the grating vector $\mathbf{K}$ in the major III-V semiconductor crystallographic directions [001] and [110].

Other directions of the grating vector $\mathbf{K}$ can provide improvements to the photorefractive gain. For example, by rotating InP:Fe about its [110] axis, Strait, Reed and Kukhtarev demonstrated a 15.5 percent gain enhancement (0.63 dB/cm) when $\mathbf{K}$ is along the $\langle 111 \rangle$ direction [41]. The improvement arises because the orientation takes advantage of all three electro-optic axes. Crystal orientation for maximizing r_{eff} is not considered further because the potential for improved gain is relatively small, and because experimentally the thesis is limited to grating vector $\mathbf{K}$ in [110] and [001].

The next two chapters will build on the original Kukhtarev theory. Specific theory for photorefractive InP:Fe is developed. Chapter 3 considers the origin of optical absorption in InP:Fe for wavelengths longer than the band gap. Chapter 4 considers the time constant and steady state photorefractive gain.

CHAPTER 3

OPTICAL ABSORPTION IN InP:Fe

3.1 Introduction

3.1.1 Purpose

This chapter discusses the process of optical absorption in InP:Fe and how it applies to photorefractive two-wave mixing. Deep level absorption is the driving process behind electro-optic based photorefractive, and so it is considered here in some detail. Absorption is explored starting from the atomic structure of indium, phosphorous, and the deep level iron trap. Electron and hole photoionization processes are explained for sub-band gap photons. The origin of the semiconductor's sub band gap absorption spectrum is explained. The optical absorption spectrum between $\lambda=950$ nm and $\lambda=1600$ nm is predicted, and compared with the measured optical absorption spectrum for InP:Fe. This chapter leads directly into **Chapter 4**, where absorption limits the time required to set up photorefractive electric fields.

The InP:Fe configuration coordinate diagram is used to model the electronic transitions active in photorefractive. A modified Lucovsky model for photoionization cross-section σ^0 , and Takanohashi and Nakajima's measured cross-section data at 77°K are used to predict σ_p^0 and σ_n^0 spectra at room temperature. With the thesis's measurement of InP:Fe optical absorption at room temperature and the predicted σ^0 spectra, the iron concentration in InP:Fe is estimated. Given the estimate for [Fe] and the two predicted σ^0 spectra, the measured absorption spectrum for InP:Fe is compared with the model for a variable charge species ratio $0 \leq [\text{Fe}^{+2}]/[\text{Fe}] \leq 1$. A best fit RMS error technique compares measured and modeled absorption spectra and leads to estimates of $[\text{Fe}^{+2}]$ and $[\text{Fe}^{+3}]$. The cross-sections and the iron species concentrations will be used later on in modeling the 2WM time constant and the 2WM gain in InP:Fe.

The technique for estimating charge species has not been performed before in InP:Fe according to available literature. The same sequence may be applied to other III-V semiconductors, such as GaAs:EL2 and GaAs:Cr. Results were obtained but are not reported in the thesis.

3.1.2 General Considerations

Optical absorption drives the photorefractive effect by freeing charge carriers from deep level traps¹ to diffuse and drift into clouds of space charge. Deep level acceptors and donors interact with photons to form either valence band holes or conduction band electrons or both, depending on the exact position of the deep level in the band gap and the fine structure (if any) associated with it.

Optical absorption arises from the deep levels. Deep levels are physical impurities and defects in the otherwise perfectly ordered tetrahedral cubic crystals. For example, one part in 10^6 of Fe in InP - approximately 10^{16} cm^{-3} - replaces indium in the lattice and creates a $\text{Fe}^{+2}/\text{Fe}^{+3}$ level 0.63 eV below CB Γ_1 at 300°K. The deep level traps electrons and holes from unavoidable low concentrations of shallow impurities. Thus, the deep level compensates for shallow impurities and tends to make the material semi-insulating. This action is fortuitous because both InP and GaAs have unavoidable deep level traps. In InP, Fe is present and cannot be cost effectively processed out of the raw indium material. In GaAs, the mid gap As antisite defect cannot be processed out of the material because it occurs naturally. The traps at large enough concentrations convert GaAs, GaAs:Cr, and InP:Fe into low conductivity semi-insulating materials that are useful for electronic, opto-electronic, and photonic applications.

Linear optical absorption " α " is described empirically as the sum of the effects of all of the absorbing centers (i.e. deep levels and other sites) in the material. Assume that N_{Ai} is the concentration of acceptor absorbing center "i", and N_{Dj} is the concentration of donor absorbing center "j". Center "i" has optical cross-section σ_{pi}^0 (producing holes) and center "j" has σ_{nj}^0 (producing free electrons). Together the ensemble of centers will result in total absorption coefficient α :

¹ Deep levels, deep level traps, and deep centers are synonyms for severe local perturbations in a semiconductor crystal lying at or near the center of the fundamental band gap. They introduce bound states that are much more localized than those of shallow impurities and act primarily as traps and recombination centers. They control the lifetime of the charge carriers, and play a major role in drift and diffusion in the photorefractive effect.

$$\alpha = \alpha_{int} + \sum_{i=1}^m N_{Ai} \sigma_{pi}^o + \sum_{j=1}^n N_{Dj} \sigma_{nj}^o \quad (3.1)$$

where

- α_{int} = absorption intrinsic to material not related to the deep levels.
- m = number of deep level acceptor species
- n = number of deep level donor species

When light travels through the medium, it loses power at rate α "nepers²" per unit distance. The optical intensity varies in such a medium according to the empirical Beer-Lambert rule for linear optical absorption [42]:

$$I(z) = I_o e^{-\alpha z} \quad (3.2)$$

where

- I_o = intensity of light just inside the crystal
- $I(z)$ = intensity of light at distance z into the medium
- z = distance of light travel in the medium

The macroscopic absorption α is based on the deep levels $\{N_i\}$. The deep levels are described quantum mechanically by localized wave functions. The III-V unit cell's crystal field acts on the substitutional impurity to introduce crystal field splitting of the orbitals [43]. The orbitals introduce levels that are split by between 0.3 eV to 1.0 eV. The orbitals are usually nondegenerate. They split into finer lines by first and second order spin-orbit and spin-spin interactions. The separation between these levels is between 0.001 eV and 0.1 eV. These levels are discussed in Sections 3.4 and 3.5.

3.1.3 InP:Fe Absorption Characteristics

The sub-band gap absorption model for InP:Fe is one of the better developed absorption models among the III-V semiconductors. For example, the absorption model for InP is more satisfactory in predicting deep level absorption than in GaAs. This is unexpected since the first semi-insulating InP became available in 1975, ten years after SI GaAs became available. The reason is the simpler InP physics. Absorption in InP:Fe has

² Nepers are the natural logarithmic measure of loss, similar to decibels. To convert to decibels, multiply nepers by 4.34 decibels per neper. When discussing gain and loss, the standard optical engineering practice is to use "<number> per centimeter"; "nepers" per centimeter is implicit and never stated.

a relatively simpler analytical explanation and is consequently better understood than in the more common III-V semiconductors GaAs:EL2 and GaAs:Cr. Unlike GaAs, no antisite defect in InP's band gap plays a role in optical absorption. The defect exists but since its electronic rest states are far from the band gap, it is decoupled from infrared interactions. This is different from GaAs where the As antisite defect EL2 complicates things in the fundamental band gap, no matter what other deep levels are present.

3.2 Chemistry

The electronic structure of neutral Fe atoms is:

$$Fe : 1s^2 2s^1 2p^6 3s^2 3p^6 3d^6 4s^2 \quad (3.3)$$

where the $3d^6 4s^2$ components contribute to the chemistry of lattice bonding. The Fe replaces the indium cation "In" in InP:Fe. The neutral In electronic structure is:

$$In : 1s^2 2s^1 2p^6 3s^2 3p^6 3d^{10} 4s^2 4p^6 4d^{10} 5s^2 5p^1 \quad (3.4)$$

where in an InP lattice, In shares three electrons in the $5s^2 5p^1$ outer shells to covalently bond with four phosphide anion "P" nearest neighbours. The phosphide has neutral structure:

$$P : 1s^2 2s^1 2p^6 3s^2 3p^3 \quad (3.5)$$

and its $3p^3$ orbitals are filled to $3p^6$ by sharing three electrons with In. The orbitals are called sp^3 hybrid bond orbitals because they are a linear combination of s and p orbitals, where it is three times as likely to find the electron in the p state than in the s state [44]. The lattice spacing for InP is 5.8687 angstroms at 291°K [45]. The spacing is comparable to that for GaAs, 5.6533 angstroms at 300°K. The approximate atomic radii of In, P, and Fe are 1.4, 1.0, and 1.3 angstroms[46].

When Fe joins the lattice, Fe's $3d^6 4s^2$ orbitals replace the In's $5s^2 5p^1$ orbitals. The deep iron level in the band gap results from the obvious mismatch: differences in shape and size between the Fe orbitals and the In orbitals distort the InP lattice. Fe donates three electrons to the phosphide atoms. Although it occupies a neutral unit cell, the Fe "donates" three electrons and is denoted "Fe⁺³". Its structure changes since Fe loses one d and two 4s orbitals to P, and the Fe outer shell becomes $3d^5$. The five orbital states available to d electrons split into a triplet and a doublet in the tetrahedrally bonded InP matrix [47]. The crystal field splitting and the net electron loss occurs because the semiconductor

covalent bond is only covalent for the In and the P lattice. Iron's 3d electrons are relatively more loosely bound than In's 5s and the 5p electrons, and the iron becomes ionized. Instead of sharing three 3p³ electron wave functions from four phosphide nearest neighbours, iron gives up three electrons and receives only two in return. Without covalent bonding to the lattice, iron is relatively decoupled from the lattice compared with In. The Fe⁺² and Fe⁺³ outer orbitals are:

$$Fe^{+3} : 3d^5 \quad Fe^{+2} : 3d^6 \quad (3.6)$$

The ionic character of the iron site renders the Fe⁺³ slightly electro-negative, although the crystal unit cell remains neutral. Fe⁺³ will trap any free electrons in the immediate atomic vicinity in the luminescent (0.63 eV at room temperature) reaction:

and this results in semi-insulating InP:Fe when the iron concentration exceeds the shallow donor concentration: $[Fe] \gg \Sigma[N_D]_i \rightarrow SI$. In SI InP:Fe therefore one expects to find the Fe population divided between a population of Fe⁺² and a population of Fe⁺³. Much of the latter portion of this chapter is spent evaluating these two populations through absorption spectra.

3.3 Band Structure

The band structure at 300°K is depicted in Figure 3.1. It is based on the model for Fe⁺² based photoconductivity advanced by Look [48] in 1979, and supported by Tapster [49] et. al. in 1981. It consists of the 1.34 eV fundamental band gap separating the valence band "VB" Γ_{15} and the conduction band "CB" Γ_1 . Two higher energy satellite conduction bands, L₁ and X, lie above Γ_1 :

- L₁ is the CB minimum in the <111> crystal direction, 0.50 eV above Γ_1 ,
- X is the CB minimum in the <001> crystal direction, 0.75 eV above Γ_1 , and
- Γ_1 lies in the center of the Brillouin zone.

Inside the band gap there is a structure consisting of two main energy levels 5E and 5T₂ separated by 0.35 eV. The levels are due to Fe⁺² and Fe⁺³. 5E and 5T₂ are the crystal field split levels of the 3d ground state of Fe⁺² (3d⁶) in tetrahedral symmetry. 5E and 5T₂ are not simple levels, but are nondegenerate due to spin-orbit splitting, generating more

Figure 3.1 InP:Fe Band Structure at 300°K.

levels in the band gap. The 5E level is within plus or minus a few millivolts of the Fermi level in SI InP:Fe. The energy levels are discussed further in **Section 3.4: Fe 3d Levels in InP:Fe**.

The band structure and the levels are determined by a variety of techniques, such as Arrhenius plots, deep level optical spectroscopy (DLOS), deep level transient spectroscopy (DLTS), photoluminescence (PL), magneto-circular dichroism (MCD), electronic paramagnetic resonance (EPR), and electron spin resonance (ESR). Discussion of these basic techniques is out of the scope of the thesis, and the results from the techniques are presented without the background details. More can be found in Pantelides's review of deep centers in semiconductors [50].

Fe fixes the Fermi level in InP:Fe near 5E at approximately 0.71 eV above the valence band Γ_{15} . This is about 0.63 eV below the conduction band Γ_1 . The Fermi level therefore is locked almost exactly between the valence and the conduction band, making InP:Fe semi-insulating near room temperature. The semi-insulating property is not quenched at lower temperatures because the Fe site is partially decoupled from the lattice through its ionic bonds. In GaAs:EL2 it is quenched below 140°K because the defect is part of the covalent lattice. The quenching behaviour is discussed in Pantelides.

The InP band gap varies as a function of temperature. Since InP is similar to GaAs, it is assumed that the general form of the GaAs gap dependence used by Sze applies to InP. Modifying coefficients to match the known InP E_g at 4°K, 77°K, and 300°K, equation (3.8) is obtained:

$$E_g = 1.42 - \frac{3.92 \times 10^{-4} T^2}{T + 204} \text{ eV} \quad (3.8)$$

This equation, together with Takanohashi and Nakajima's results, show that as temperature increases, CB, 5E, and 5T2 remain relatively unaffected, while at room temperature, VB Γ_{15} rises [51] at $\approx 0.33 \text{ meV/K}$. In addition, the equation is used to predict photoionization cross-sections σ_p° later on in the thesis when temperature scaling factors are employed.

3.4 Fe 3d Levels in InP:Fe

Fe has two crystal split levels in the InP fundamental band gap. The level corresponding to the $\text{Fe}^{+2}\text{-Fe}^{+3}$ transition lies near the center of the energy gap in InP:Fe. Electron spin resonance (ESR) techniques are used to probe the Zeeman splitting of the Fe^{+3} level ground state in an applied magnetic field. ESR provides information about the immediate environment of transition metals in III-V semiconductors with non-zero orbital angular momentum. The Fe^{+2} levels are explored using other techniques.

The $3d^6 \text{Fe}^{+2}$ levels are shown in Figure 3.2. They can be observed through optical scanning exciting d-d interactions. The two main levels, 5E and 5T2, are important to the photorefractive effect. They originate from the 3d orbital fields interacting with the cubic fields of the InP unit cell. Absorption studies near $\lambda = 3 \mu\text{m}$ indicate 5E-5T2 level interactions. Photoluminescence and acoustic paramagnetic resonance experiments show that the Fe^{+2} 5E level is nondegenerate and splits into an orbital doublet, ending up as a ground state pentet of levels known as $5\Gamma_3$ [52]. These levels are almost evenly split by $\approx 0.01 \text{ eV}$ and are due to third-order spin-orbit interaction. The levels are labeled 1,2,3,4, and 5 in Figure 3.2. 0.35 eV above the 5E level there is the excited Fe^{+2} state 5T2 which is also nondegenerate. Spin-spin and spin-orbit interactions convert the basic 5T2 level into an orbital triplet, a doublet, and a singlet. 5T2 becomes an excited state sextet known as $5\Gamma_5$. These levels are separated by $\approx 0.01 \text{ eV}$. The levels are labeled 6,7,8,9,10, and 11 in

Figure 3.2 - Fe^{+2} 3d levels in InP:Fe.

the Figure 3.2.

The $3d^5 \text{Fe}^{+3}$ levels coincide closely to the $5E 3d^6 \text{Fe}^{+2}$ level. They can be observed through ESR techniques. The $3d^5 \text{Fe}^{+3}$ levels are different from the Fe^{+2} levels because Fe^{+3} has one less 3d electron than Fe^{+2} . One electron significantly changes the physics of the site. The most important change is that there is no large optical 3d structure, but instead a fine optical structure based on spin. $\text{Fe}^{+3} 3d^5$ has a non-zero orbital angular momentum, compared with $\text{Fe}^{+2} 3d^6$ having zero orbital angular momentum. Therefore Fe^{+3} produces a clear five line ESR spectrum out of six allowed levels separated by ≈ 0.01 eV, while Fe^{+2} is more or less invisible to ESR with only ≈ 3 micro-eV spacing between levels. The anomalous Fe^{+2} levels are caused by crystal field splitting introducing a small non-zero component to its angular momentum. Such small separations between Fe^{+2} ESR levels are invisible to optical scanning.

The differences between Fe^{+3} and Fe^{+2} can be summarized as follows. Fe^{+3} in InP:Fe shows a clear four line ESR spectrum at energies of ≈ 0.01 eV. However, there is no d-d structure for Fe^{+3} in the optical range. Fe^{+2} shows a much weaker five line ESR spectrum ($0.000,003$ eV) and sharp d-d transitions in the ≈ 0.3 eV optical range. These differences result from the different spin (ESR) and the different allowed levels (d-d) for

the two wave functions. Even though there are differences, both centers are significant in absorbing light in the range $950 \text{ nm} \leq \lambda \leq 1600 \text{ nm}$. Fe^{+3} absorbs light near $\lambda = 1.064 \mu\text{m}$ by accepting a VB electron into a 3d orbital and producing a hole. Fe^{+2} absorbs light by losing a 3d electron and producing a free electron. These transitions drive 2WM and they are explored in Section 3.5.

3.4.1 5T2 Population

The 0.35 eV separation of the crystal split 5E and 5T2 Fe^{+2} levels invites speculation about the population of the 5T2 excited state of Fe^{+2} , known as Fe^{+2*} , at room temperature. Considering Fe^{+2} as a two level system of Fe^{+2} (5E level) and Fe^{+2*} (5T2 level), one may model it with a Maxwell-Boltzmann distribution function. Fe^{+2} and Fe^{+2*} have degeneracies g_i , 5 and 6, and energies ϵ_i , 0 and 0.35 eV, respectively, as described in the last few paragraphs. The population of the various levels is [53]:

$$\frac{N_j}{N} = \frac{g_j \exp\left(-\frac{\epsilon_j}{k_B T}\right)}{Z} \quad (3.9)$$

where:

N_j = population of species "j"
 N = total population of Fe^{+2}
 Z = partition function, where

$$Z = \sum_j g_j \exp\left(-\frac{\epsilon_j}{k_B T}\right) = 5 \exp(0) + 6 \exp(-13.5) = 5.000008 \quad (3.10)$$

Therefore one finds that $[\text{Fe}^{+2}_{\text{rest}}]/[\text{Fe}^{+2}] \approx 1.0$, and $[\text{Fe}^{+2*}]/[\text{Fe}^{+2}] \approx 1.4 \times 10^{-6}$. The excited state Fe^{+2*} forms a small fraction of the Fe^{+2} . Later on this chapter shows that $[\text{Fe}^{+2}] \approx 3.4 \times 10^{15} \text{ cm}^{-3}$. $[\text{Fe}^{+2*}]$ is small: only $\approx 5 \times 10^{10} \text{ cm}^{-3}$. 99.99986 percent of Fe^{+2} is in the rest state. Therefore while the excited state is partially populated, it is assumed to be negligible in this thesis.

3.4.2 Chemical Potential

The chemical potential μ of Fe^{+2} may be determined from the partition function Z . μ is important in various thermal considerations and is found using:

$$\frac{1}{Z} = \exp\left(\frac{\mu}{k_B T}\right) \therefore \mu = -0.042 \text{ eV} \quad (3.11)$$

μ is found to be -0.042 eV, which is close to the thermal energy at room temperature $k_B T = 0.026 \text{ eV}$.

3.5 Active Transitions

Deep level optical absorption is the basic transition that begins the photorefractive effect. Generally, a transition is the event when an electron's wave function transfers from one lattice energy level to another. In InP:Fe there are five transitions important to the photorefractive effect, and other simultaneous transitions that are not so important. There are thermal transitions, optical transitions, and trap transitions both inside the deep levels, and between the deep levels, VB, and CB. In InP:Fe at room temperature and for $1000 \leq \lambda \leq 1600 \text{ nm}$, the three dominant transitions are thermal ionization $5E \rightarrow CB$, and photoionization $VB \rightarrow 5E$ and $VB \rightarrow 5T_2$. The photoionization rate of holes is greater than the photoionization rate for electrons because $\sigma_p^0[Fe^{+3}] = 1.00 \text{ cm}^2$ $\gg$ $\sigma_n^0[Fe^{+2}] = 0.06 \text{ cm}^2$ at $\lambda = 1.064 \text{ }\mu\text{m}$. The result is that holes are the dominant photocarrier in InP:Fe.

The two other important transitions involve charge carriers losing energy. These transitions occur when free electrons and holes are trapped back to the $5E$ mid band gap level. The trap transitions influence the photorefractive effect by replenishing electron populations in the band structure diagram. While other transitions operate at the same time, only these five major transitions are considered in the model for 2WM.

For one reason or another, not all level transitions are allowed due to restrictions in spin, dipole moments, and optical energy thresholds. In particular, the direct transition $VB \rightarrow CB$ is not seen in the usual wavelength range of electro-optic photorefractive work because usually the photons have less energy than the band gap. For this thesis, $\lambda = 1.064 \text{ }\mu\text{m}$ and $1.152 \text{ }\mu\text{m}$, and $VB \rightarrow CB$ is not significant. However, in general the transition is important because it allows photorefraction in crystals of any symmetry such as Si and Ge, and to polycrystals through Franz-Keldysh electrorefractive photorefraction.

The Franz-Keldysh effect is "an electric field-induced [linear] change of the complex dielectric constant of a direct band-gap semiconductor, occurring at optical energies close to the band-gap energy." [54] It is not the Kerr effect since the Kerr effect is quadratic in electric field. Electrorefractive photorefractive is one part of the Franz-Keldysh effect, based on changes to the refractive index component of the dielectric constant. Koehler, Garmire, Partovi, Glass, and Klein found that space charge clouds alter the absorption and thence the refractive index through mixed conduction and the Franz-Keldysh effect³. In InP the Franz-Keldysh effect may be an order of magnitude larger than the Pockels effect when operating within several tens of nanometers of the band-gap, and therefore it is a possible narrow band substitute for photorefractive beam coupling.

Electrorefractive photorefractive in InP is based on wavelengths near the gap wavelength, which is $\lambda_g = 926$ nm at 300°K. For example, Partovi et. al. observed net gain of 13.3 cm^{-1} in GaAs:EL2 for $\lambda = 939$ nm, a moving grating with $\Lambda = 7 \text{ }\mu\text{m}$, $E_0 = 10 \text{ kV/cm}$, $I_0 = 140 \text{ mW/cm}^2$, and beam ratio = 5000. The large gain was obtained by combining electro-optic photorefractive with electrorefractive photorefractive [55]. Millerd et. al. report two-wave mixing gain as large as 14 cm^{-1} without the Pockels effect when operating at 970 nm in InP:Fe [56]. However, since this thesis explores the electro-optic photorefractive effect and not the electrorefractive photorefractive effect, the high absorption band edge effects are not considered further.

Other transitions become important in InP:Fe for longer wavelengths. Past $\lambda = 1.6 \text{ }\mu\text{m}$, electrons become the dominant photocarrier in InP:Fe. Photoexcitation in the range $0.59 \leq h\nu \leq 0.70 \text{ eV}$ or $1.8 \leq \lambda \leq 2.1 \text{ }\mu\text{m}$ is caused by the $5E \rightarrow \Gamma_1$ transition. Photoexcitation in the range $0.27 \leq h\nu \leq 0.59 \text{ eV}$ or $2.1 \leq \lambda \leq 4.6 \text{ }\mu\text{m}$ is caused by the $5T2 \rightarrow \Gamma_1$ transition [57].

Deep level to deep level transitions also play a role in InP:Fe. Fe^{+2} electrons are photoexcited from 5E to 5T2 using $0.27 \leq h\nu \leq 0.59 \text{ eV}$ or $2.1 \leq \lambda \leq 4.6 \text{ }\mu\text{m}$ light. Once in 5T2, electrons thermalize down through the 5T2 sextet until they reach level six (see Figure

³ The magnitude of Franz-Keldysh electrorefraction can be determined by applying the Kramers-Kronig relationship to measurements of electroabsorption.

3.2). From level 6, electrons fall to 5E states 1,2,3, or 4 in the indicated transitions. The fall results in photoluminescence around $\lambda=3 \mu\text{m}$. Note that the (6-1) transition $5T_2 \rightarrow 5A_2$ is not observed since it is forbidden by electric dipole mismatch. These long wavelength transitions are allowed for $\lambda=1.064 \mu\text{m}$ during 2WM work, but they have not been observed to alter the results of the experiments. Therefore, while they are mentioned for completeness, they are not considered further.

The next three subsections consider the four transitions that are important to photorefractive two-wave mixing.

3.5.1 Thermal Excitation Transitions

Thermal excitation of electrons at room temperature happens due to phonon absorption. The loosest bound electron in the Fe^{+2} deep level is excited to the conduction band in the transition $5F \rightarrow \text{CB}$. There are three such possible transitions corresponding to the three conduction bands: Γ_1 , L_1 , and X_1 . Each transition requires a different phonon energy and has a different thermal cross-section. Since at room temperature $k_B T \approx 0.026 \text{ eV}$, only the lowest energy transition is probable: 0.63 eV to Γ_1 , or $24k_B T$. The transitions to L_1 and X require 1.13 ($43k_B T$) or 1.33 eV ($51k_B T$) respectively, and are insignificant compared to Γ_1 . The transition to Γ_1 is:

The thermal cross-section of Fe^{+2} is $\sigma_n^\infty = 3.5 \times 10^{-14} \text{ cm}^2$ [58]. The rate of thermal electron excitation to Γ_1 per $[\text{Fe}^{+2}]$ site is [59]:

$$\frac{1}{\tau} = v_{\text{rms}} n_c \sigma_n^\infty \exp\left(-\frac{e_i}{k_B T}\right) \quad (3.13)$$

where

- v_{rms} = rms thermal velocity
- n_c = effective conduction band density of states
- σ_n^∞ = electron capture cross-section at infinite temperature in Fe^{+2}
- T = temperature
- e_i = transition energy $5E \rightarrow \Gamma_1$

The product of v_{rms} and n_c is:

$$v_{\text{rms}} n_c = \left(\frac{m_e k_B^2}{h^3} \right) \times (96 \pi^3)^{\frac{1}{2}} \times T^2 \times \left(\frac{m^*}{m_e} \right) = 3.25 \times 10^{25} [m^{-2} K^{-2} s^{-1}] T^2 \left(\frac{m^*}{m_e} \right) \quad (3.14)$$

where

m_e = electron rest mass
 m = electron effective mass in Γ_1
 h = Planck's constant

The total rate of thermal excitation of electrons from 5E for all Fe^{+2} sites is:

$$\text{Thermal electron rate} = 3.25 \times 10^{25} [m^{-2} K^{-2} s^{-1}] T^2 \frac{m_e^*}{m} \sigma_n \exp\left(-\frac{e_i}{k_B T}\right) [Fe^{+2}] \quad (3.15)$$

The thermal rate of electron excitation may equal the optical rate of hole excitation at room temperature for relatively low power densities. When the rates are equal a resonance is produced that is important for photorefractive gain. The resonance is discussed in **Chapter 6**.

Thermal excitation of holes happens less frequently. A thermal hole is formed when a valence band electron is sent to 5E or 5T2. The required energy is 0.71 eV ($27k_B T$) and 1.06 eV ($41k_B T$) respectively. The hole thermal cross-section $\sigma_p^o = 3 \times 10^{-16} \text{ cm}^2$ is one hundred times smaller than the electron thermal cross-section. The transition is ignored for the purposes of photorefractive in all available references.

3.5.2 Optical Excitation Transitions

Photoionization forms holes by driving electrons from the valence band Γ_{15} to the Fe^{+3} level, yielding Fe^{+2} in either 5E or the 5T2 form. This requires 0.71 or 1.06 eV respectively. The transition is:

$Fe^{+2}(5T2)$ quickly relaxes from its excited state by luminescing at ≈ 0.35 eV in a transition to $Fe^{+2}(5E)$. The total optical cross-section for Fe^{+3} σ_p^o is $330 \times 10^{-18} \text{ cm}^2$ for light at $\lambda = 1.064 \mu\text{m}$ and 300°K. The rate of optical hole excitation is:

$$\text{Hole rate} = \sigma_p^o \frac{I_o}{h\nu} [Fe^{+3}] \frac{\text{holes}}{\text{cm}^3 \text{-second}} \quad (3.17)$$

Photoionization forms electrons by driving electrons from the Fe^{+2} 5E deep level to CB, either Γ_1 , L_1 , or X_1 . This requires 0.63, 1.13, or 1.33 eV respectively. The transition is:

The total optical cross-section for Fe^{+2} σ_n^o is $1.7 \times 10^{-18} \text{ cm}^2$ for light at $\lambda = 1.064 \text{ }\mu\text{m}$ and 300°K. The rate of optical electron excitation is:

$$\text{Electron rate} = \sigma_n^o \frac{I_o}{h\nu} [Fe^{+2}] \frac{\text{electrons}}{\text{cm}^3\text{-second}} \quad (3.19)$$

At $\lambda = 1.064 \text{ }\mu\text{m}$ the optical cross-section for holes in Fe^{+3} is 194 times larger than that for electrons in Fe^{+2} . Therefore when $194[Fe^{+3}] > [Fe^{+2}]$, holes dominate optical absorption in InP:Fe. This is true for 2WM at $\lambda = 1.064 \text{ }\mu\text{m}$.

3.5.3 Carrier Trap Transitions

Thermally and optically excited carriers diffuse and drift around the InP crystal until trapped by an acceptor. Electrons are captured by Fe^{+3} in $CB \rightarrow 5E$:

There is no agreement on the recombination cross-section of electrons σ_n^r , but there is consensus on the order of magnitude. Valley finds σ_n^r is 10^{-14} cm^2 at 300°K [60]. Look finds σ_n^r is 10^{-15} cm^2 [61]. The thesis uses $\sigma_n^r = 3.5 \times 10^{-14} \text{ cm}^2$.

Holes are captured by Fe^{+2} in $5E \rightarrow \Gamma_{15}$:

There is no agreement on the recombination cross-section of holes σ_p^r either. Valley finds the recombination cross-section for holes σ_p^r is $3 \times 10^{-16} \text{ cm}^2$ at 300°K [62]. Picoli et. al. use $\sigma_p^r = 10^{-15} \text{ cm}^2$ [63]. The thesis uses $\sigma_p^r = 3 \times 10^{-16} \text{ cm}^2$.

Electron and hole capture rates are discussed with respect to the photorefractive time constant in Section 4.1 of this thesis.

3.6 Configuration Coordinate Diagram

The configuration coordinate diagram is a state diagram depicting the energy relationships between allowed energy states in the first reduced Brillouin zone, as described by Pantelides. The diagram is a graph of wave function total energy (potential plus kinetic) versus normalized linear momentum "q" in the crystal's principle axes: L_1 $\langle 111 \rangle$ and X_1 $\langle 001 \rangle$. Before discussing the configuration coordinate diagram, one needs to become familiar with the Franck-Condon parameter.

3.6.1 Franck-Condon Parameter d_{FC}

The Franck-Condon parameter d_{FC} is the difference between thermal activation energy and the optical activation energy for the same transition. In general, the thermal energy to drive a transition is greater than or equal to the optical energy. They are equal only for direct transitions. For indirect transitions, the two energies are different because the potential minima of the levels do not have the same momentum. For example, consider two hypothetical parabolic states A and B that are slightly offset in momentum from Γ_1 . After optical excitation $A \rightarrow B$ from A's minimum to B's curve, the electron must lose energy to assume steady state at B's lowest potential energy. It does this by moving to B's potential minimum. From the relatively stable B minimum, $B \rightarrow A$ relaxation may occur, but the energy separation is different from $A \rightarrow B$.

d_{FC} partly explains the temperature dependence of the optical cross-sections of electrons and holes [64], and the difference between two states potential energy separation and the actual energy involved in a transition. For example, d_{FC} plus the thermal ionization energy is approximately the optical ionization energy. This helps understanding the variety of transitions on the configuration coordinate diagram. The configuration coordinate diagram for InP:Fe is detailed in the next section.

3.6.2 InP:Fe Details

The configuration coordinate diagram for InP:Fe at 300°K is shown in Figure 3.3 [65], with the iron level information coming from Bremond et. al. [66]. The figure shows the levels that are important for the photorefractive effect in InP:Fe. The levels for InP:Fe are to scale on both axes. The levels for Fe are to scale on the vertical axis but the exact curvature and the location of the potential energy minima were not found in the

the available literature. Higher and lower energy levels are not shown because they do not contribute to the physics of the photorefractive effect.

As expected from the band structure diagram, the Fe^{+2} levels are positioned between CB Γ_1 and VB Γ_{15} . 5E and 5T2 are slightly offset from the zero wave vector point

Figure 3.3 InP:Fe configuration coordinate diagram with normalized Brillouin zone at 300°K.

in the X <001> direction. Clearly there is an energy difference between optical excitations (vertical transitions) and thermal excitation (horizontal and vertical transitions) in, for example, transitions involving the 5E $\text{Fe}^{+2}/\text{Fe}^{+3}$ level like $\Gamma_{15} \leftrightarrow 5\text{E}$, and $5\text{E} \leftrightarrow \Gamma_1$. The difference is the Franck-Condon parameter d_{FC} .

The three depicted band transitions shown are optical and thermal transitions. The 0.85 eV and the 1.2 eV transitions are optically excited hole creation events. The 0.7 eV transition is either a thermally or an optically excited free electron creation event. A number of other levels surround the 1.34 eV material band gap at 300°K. The heavy hole level Γ_{15} defines the lower edge of the valence band. Light holes approach the heavy hole energy at the zero wave vector point. The conduction band is defined by the Γ_1 minimum, and the two higher energy satellite bands, L_1 <111> and X <001>. X has not been modeled in the photorefractive effect by other researchers. It is used in this chapter to model absorption spectrum of InP:Fe and to predict the iron concentration.

Defect deep levels like iron differ from the usual configuration coordinate diagrams because the defects are by definition not periodic. Therefore the deep levels in the configuration coordinate diagram do not repeat every Brillouin zone, while CB and VB levels repeat.

3.7 Photoionization

Photoionization cross-sections for electrons and holes σ_n^0 and σ_p^0 determine the bulk absorption coefficient, the competition between electrons and holes, and the suitability of a material for use as a photorefractive two-beam coupling medium. In this section, the model for σ_n^0 and σ_p^0 is considered and compared with measured values. The model is used to scale the measured data from 77°K to 300°K. The scaled room temperature data is used in Section 3.8 to evaluate $[\text{Fe}]$ and $[\text{Fe}^{+2}]/[\text{Fe}]$ from room temperature optical absorption spectra of InP:Fe.

3.7.1 Lucovsky Model

The Lucovsky model [67] expresses the photoionization cross-section σ^0 of deep impurity centers such as Fe in InP:Fe for forbidden direct transitions such as $5E \rightarrow \Gamma_1$. It differs qualitatively from σ^0 for shallow impurities because low energy ionization (≈ 0.05 eV) is based on the relatively unperturbed and uniform Coulomb covalent crystal field ($V \propto 1/r$). In deep levels, though, larger energy for ionization (0.70 eV) is needed due to the perturbed and locally ionic crystal field. The long range Coulomb effect is neglected and the ion core is assumed to be a delta function. σ^0 is based on the model in which the ion core is responsible for the ground state binding energy. The normalized ground state wave function for deep levels used by Bethe and Morrison is [68]:

$$\psi(r) = \sqrt{\frac{\alpha}{2\pi}} \frac{e^{-\alpha r}}{r} \text{ where } \alpha = \sqrt{\frac{m^* E_i}{2\pi^2 \hbar^2}} \quad (3.22)$$

The deep level wave function is clearly different from the 1s hydrogenic wave function for shallow levels [69]:

$$\psi_{1s}(r) = \frac{1}{\sqrt{\pi}} \alpha^{\frac{3}{2}} e^{-\alpha r} \text{ where } \alpha = \sqrt{\frac{m^* e^2}{4\pi^2 \hbar^2}} \quad (3.23)$$

The deep level has a singularity at its center while the shallow level is well behaved. In addition, the deep level is much more localized. In considering the optical cross-section, Bethe and Morrison treated the interaction between photons and deuterons. Dexter extended it to interactions in a crystal lattice between photons and deep levels. The result for σ^0 is given by Lucovsky:

$$\sigma^o(h\nu) = \frac{1}{n} R^2 \frac{16\pi e^2 h}{3m^* m_e c} \frac{E_i^{1/2} (h\nu - E_i)^{3/2}}{(h\nu)^3} \quad (3.24)$$

where:

σ^o	=	photoionization cross-section
n	=	index of refraction
R	=	effective field ratio for the radiation inducing the transition
e	=	electric charge
h	=	Planck's constant
ν	=	optical frequency (radians per second)
c	=	speed of light
m_e	=	electron mass
m^*	=	effective mass
E_i	=	measured transition ionization energy
c	=	speed of light

R , the effective field ratio for the radiation inducing the transition, is the ratio of the electric field needed to induce the deep level transition to the electric field in the unperturbed lattice.

Takanohashi and Nakajima measured the wavelength dependence of σ_p^o for holes and σ_n^o for electrons in n-InP:Fe using photocapacitance spectroscopy (PHCAP) [70] at 77°K in 1989. Their results are depicted as the symbols at 77°K in Figure 3.4. The PHCAP experimental technique is described below because it is important in checking trends in the Lucovsky model predictions of σ^o vs λ .

3.7.2 PHCAP

PHCAP is transient photocapacitance spectroscopy of electron and hole photoionization cross-sections of semiconductor deep levels. The experiment requires a junction diode where at low temperatures, under reverse bias, the transient and steady state capacitance is measured after application of monochromatic light.

To perform the measurement, Takanohashi and Nakajima grew 5 μm of SI InP:Fe onto a (100) oriented n^+ -InP:Sn substrate. They prepared an abrupt p^+ -n junction on this by diffusing Cd into the InP:Fe to a depth of 1.2 μm . AuZn formed the p^+ contact. AuSn formed the n^+ contact. 1/2 mm diameter mesa-type p^+ -n junction diodes were

Figure 3.4 InP:Fe photoionization cross sections for electrons and holes at 77°K with experimental data points from Takanohashi [70]. Hole data points for 300°K are scaled.

prepared by wet-chemical etching the wafer.

The PHCAP measurements are performed by cooling to 77°K in the dark at zero bias. Then a five volt reverse bias is applied. The depletion region fills the diode and a steady state condition is achieved. Then the diode is illuminated with monochromatic light. The transient capacitance is recorded as a function of time. The total change in the depletion region capacitance between steady state in darkness and steady state illumination is $\Delta C(\infty)$. $\Delta C(\infty)$ is proportional to the difference between the total trap concentration and the steady state number of excited centers, since as centers become excited, the depletion region empties, the equivalent capacitor plates get further apart, and the capacitance falls.

The photoionization cross-sections of electrons and holes is determined by appropriately analyzing PHCAP data. Two equations can be produced relating σ_p° and σ_n° to the data. One equation relates the steady state total capacitance change to the cross-sections:

$$n_i(\infty) = f(\Delta C(\infty)) = [Fe] \left[\frac{\sigma_p^\circ}{\sigma_p^\circ + \sigma_n^\circ} \right] \quad (3.25)$$

where

$n_i(\infty)$ = steady state number of excited centers
 $[Fe]$ = total trap concentration

$n_i(\infty)$ is calculated based on $\Delta C(\infty)$.

The second equation relates the capacitive transient to the cross-sections:

$$e^\circ = (\sigma_p^\circ + \sigma_n^\circ)N \quad (3.26)$$

where

e° = charge carrier emission rate
 N = optical flux density photons/cm²/sec

These simultaneous equations yield σ_n° and σ_p° for one λ . By performing the measurement at many different λ s one obtains the σ_n° and σ_p° spectra.

3.7.3 σ_n° and σ_p° Measurements

Takanohashi and Nakajima modeled only two of the four photorefractive cross-sections using the Lucovsky model and a two band model accounting for dominant transitions. They modeled the direct transitions $5E \rightarrow \Gamma_1$ and $\Gamma_{15} \rightarrow 5E$. Indirect transitions $5E \rightarrow L_1$ and $\Gamma_{15} \rightarrow 5T_2$ were not modeled. While they did refer to the optical absorption coefficient of the deep level impurity, they did not extend it to estimate total iron concentration or the charge species ratio $[Fe^{+2}]/[Fe^{+3}]$. In 1990 Strait, Reed, Valley and Klein extended Takanohashi and Nakajima's data to estimate $[Fe^{+2}]$ and $[Fe^{+3}]$ [71]. Strait et. al. did not, however, use σ° to estimate total iron concentration. No one has used the technique in other III-V semiconductors, probably because σ_n° and σ_p° are not as well known for GaAs:EL2 and GaAs:Cr. It is due to GaAs sub-band gap processes being physically more complicated than such processes in InP.

The Lucovsky model is fitted to five transitions to produce a σ° for each transition. These σ° s are summed to obtain two composite cross-sections. Then this model's composite σ° parameters are fitted to the observed σ° data. The model is compared to Takanohashi and Nakajima's data. Then Takanohashi and Nakajima's σ° data is scaled from 77°K to 300°K in order to estimate the total iron concentration and the charge species ratio $[Fe^{+2}]/[Fe^{+3}]$.

According to Takanohashi and Nakajima, the Lucovsky model gives the photoionization cross-section spectrum for the forbidden⁴ direct transition $5E \rightarrow \Gamma_1$ (producing free electrons) at all temperatures as:

$$\sigma_n^\circ(5E-\Gamma_1) = 1.0 \times 10^{-17} \left[\frac{(h\nu - 0.63)^{\frac{3}{2}}}{(h\nu)^3} \right] \text{ cm}^2 \quad (3.27)$$

The σ_n° equation for allowed indirect transitions $5E \rightarrow L_1$ and $5E \rightarrow X_1$ is similar to the σ_n° equation for forbidden direct transitions. There are two differences: first, the activation energy is $E_i = 1.13$ eV and 1.33 eV respectively at all temperatures, and second,

⁴ "Forbidden" means a transition from one wave function at a stable potential energy minimum to an unstable point on another wave function that is somewhat displaced from the local potential energy minimum by some value of kinetic energy.

the $h\nu - E_i$ term is taken to the power 2 instead of to the power $3/2$ [72] [73]. The powers are assigned by transition type as seen on the configuration coordinate diagram. The formalism is given in Table 3.1:

Table 3.1 - Optical Cross-section Power Dependence

TRANSITION TYPE	SYNONYM	POWER	EXAMPLES
ALLOWED DIRECT	DIRECT OPTICAL	1/2	$e-h^0 + h\nu \rightarrow e'_{T1} + h^+$
FORBIDDEN DIRECT	DIRECT OPTICAL	3/2	$Fe^{+3} + e-h^+ + h\nu \rightarrow Fe^{+2}(5E)$ $Fe^{+2}(5E) + h\nu \rightarrow Fe^{+3} + e'_{T1}$
ALLOWED INDIRECT	THERMAL + OPTICAL	2	$Fe^{+2}(5E) + h\nu \rightarrow Fe^{+3} + e'_{X1}$
FORBIDDEN INDIRECT	THERMAL + OPTICAL	3	$e-h^0 + h\nu \rightarrow e'_{X1} + h^+$

In general, the form of the equations used for these transitions are as follows:

Allowed direct transitions:

$$\sigma^0 = k_{\text{allowed direct}} \times (h\nu - E_i)^{1/2} \quad (3.28)$$

Forbidden direct transitions:

$$\sigma^0 = k_{\text{forbidden direct}} \times \frac{(h\nu - E_i)^{3/2}}{(h\nu)^2} \quad (3.29)$$

Allowed indirect transitions:

$$\sigma^0 = k_{\text{allowed indirect}} \times \frac{(h\nu - E_i + h\omega_q)^2}{\exp\left(\frac{h\omega_q}{k_B T}\right) - 1} \quad (3.30)$$

Forbidden indirect transitions:

$$\sigma^0 = k_{\text{forbidden indirect}} \times \frac{(h\nu - E_i + h\omega_q)^3}{\exp\left(\frac{h\omega_q}{k_B T}\right) - 1} \quad (3.31)$$

where ω_q is the transverse phonon frequency, around 10 THz. While the indirect transitions

have an explicit temperature dependence, temperature dependence is implicit in all terms through the transition energy E_t .

Three of the four types of electronic transitions are depicted in Figure 3.5, where the configuration coordinate diagram has been redrawn with transitions indicated by zig-zag lines. Allowed direct transition from valence to conduction band is not shown because it would clutter the figure too much.

Figure 3.5 InP:Fe configuration coordinate diagram showing different types of electronic transitions.

Takanohashi and Nakajima's paper supplied the data to fit the power model, performed in the course of the thesis. One assumes that σ_n^o behaves additively with both σ_n'' components adding to the best fit curves. The combined curve for the three transition σ_n^o is:

$$\sigma_n^o(\lambda) = \sigma_n^o(\lambda)_{F_0 \rightarrow 5E - \Gamma_1} + \sigma_n^o(\lambda)_{F_0 \rightarrow 5E - L} + \sigma_n^o(\lambda)_{F_0 \rightarrow 5E - X} \quad (3.32)$$

The best fit of the total $\Sigma\sigma_n^o$ at 77°K is:

$$\sigma_n^o(\lambda) = 1 \times 10^{-17} \frac{(h\nu - 0.63)^{\frac{3}{2}}}{(h\nu)^3} + 2 \times 10^{-15} (h\nu - 1.095)^2 + 5 \times 10^{-14} (h\nu - 1.18)^2 \text{ cm}^2 \quad (3.33)$$

where the transitions to L_1 and X bands are modeled as allowed indirect transitions, as can be seen from the configuration coordinate diagram. To obtain the best fit, the two indirect transition components were modified by adjusting the multiplicative parameters on the 1.11 eV and the 1.18 eV terms, which correspond to the L_1 1.13 eV and the X_1 band 1.33 eV transitions respectively. The fit is depicted in **Figure 3.4** as the continuous lines. The diamond symbols are Takanohashi and Nakajima's data. The continuous curve "ELECTRONS" is the model. The model agrees with the data for $1.0 \mu\text{m} < \lambda < 1.8 \mu\text{m}$. For $\lambda < 0.95 \mu\text{m}$, the data diverges from the model due to more complicated transitions with the higher energy photons. Direct VB→CB absorption accounts for the disagreement in the $0.9 < \lambda < 0.95 \mu\text{m}$ range. Photoionization of electrons in InP is adequately modeled in the region of experimental interest for this thesis, $0.95 < \lambda < 1.6 \mu\text{m}$.

Takanohashi and Nakajima model σ_p^o for $\Gamma_{15} \rightarrow 5E$ as a forbidden direct transition (producing holes) at room temperature:

$$\sigma_p^o(\Gamma_{15} \rightarrow 5E) = 4.8 \times 10^{-16} \left(\frac{(h\nu - 0.71)^{\frac{3}{2}}}{(h\nu)^3} \right) \text{ cm}^2 \quad (3.34)$$

This work models transition $\Gamma_{15} \rightarrow 5T_2$ as an allowed indirect transition with activation energy $E_a = 1.13 \text{ eV}$ at 77°K. Takanohashi and Nakajima did not model it. In addition, when the temperature changes to 300°K, the transition energy changes to 1.06 eV. They did not model this either. The combined curve for the two transition σ_p^o is:

$$\sigma_p^o(\lambda) = \sigma_p^o(\lambda)_{\Gamma_{15} \rightarrow F_0 \rightarrow 5E} + \sigma_p^o(\lambda)_{\Gamma_{15} \rightarrow F_0 \rightarrow 5T_2} \quad (3.35)$$

The best fit of the total $\Sigma\sigma_p^\circ$ at 77°K is:

$$\sigma_p^\circ(\lambda) = +4.8 \times 10^{-16} \frac{(h\nu - 0.78)^{\frac{3}{2}}}{(h\nu)^3} + 1.5 \times 10^{-14} (h\nu - 1.11)^2 \text{ cm}^2 \quad (3.36)$$

Again a best fit was performed on the indirect transition component by adjusting the multiplicative parameter in the 1.11 eV term, which corresponds to the 5T2 1.13 eV transition. The fit is depicted in Figure 3.4. The triangles are Takanohashi and Nakajima's data. The continuous curve "HOLES" is the model. The model agrees with the data for $0.95 < \lambda < 1.6 \mu\text{m}$. However, for $\lambda < 0.95 \mu\text{m}$, the measured value of σ_p° falls off. This is not accounted for by Lucovsky or by Takanohashi and Nakajima. No one has advanced a quantitative explanation for the moderate disagreement in the $0.90 < \lambda < 0.95 \mu\text{m}$ range. Clearly, though, the fall is a band edge effect due to direct valence band to conduction band transitions. The direct transition produces electron-hole pairs and overpowers the effect of the iron transitions. Iron is only one part in a million or so in InP:Fe, and so one expects iron absorption effects to be swamped near the intrinsic band gap.

In spite of the band edge effect, photoionization of holes in InP is adequately modeled in the region of experimental interest for 2WM in this thesis, $1.0 \mu\text{m} < \lambda < 1.2 \mu\text{m}$.

The σ_p° model given above is valid only for 77°K, while a model for room temperature operation is desired. No published photoionization spectra exist for individual Fe^{+2} and Fe^{+3} species' σ_p° at 300°K. Scaling the data using the composite model provides σ_p° information at other temperatures, given knowledge of the changes in the transition level energies. At 300°K the band gap E_g is 1.350 eV ($k_B T = 0.026$ eV) instead of 1.412 eV ($k_B T = 0.007$ eV) from equation (3.11). The difference of 0.062 eV is the change in the valence band level. From Takanohashi and Nakajima, to first order, the Fe levels and the conduction band levels are constant. Therefore the energy terms in the σ_p° expression fall ≈ 0.07 eV from 0.78 eV and 1.11 eV to 0.705 eV and 1.04 eV respectively. It seems that the change is not exactly the 0.062 eV band gap change because of thermal effects and possible small errors in the used transition levels for 300°K as shown in Figure 3.1 for the InP:Fe band structure. The multiplicative terms remain the same at 300°K. Therefore σ_p° at 300°K is:

$$\sigma_p^o(\lambda) = +4.8 \times 10^{-16} \frac{(h\nu - 0.705)^{\frac{3}{2}}}{(h\nu)^3} + 2.5 \times 10^{-14} (h\nu - 1.04)^2 \text{ cm}^2 \quad (3.37)$$

The temperature shifted curve for σ_p^o at 300°K is shown in Figure 3.4 above the curve for 77°K. At 1.06 μm at 300°K, experimentally Valley has found $\sigma_p^o = 3.1 \times 10^{-16} \text{ cm}^2$ and $\sigma_n^o = 4 \times 10^{-18} \text{ cm}^2$. The model here gives $\sigma_p^o = 3.29 \times 10^{-16} \text{ cm}^2$ and $\sigma_n^o = 3.9 \times 10^{-18} \text{ cm}^2$. The measured and theoretical hole cross-sections are within six percent of each other. The electron cross-sections differ by three percent in the region of interest. The general agreement is important to modeling in the next section. The agreement facilitates measurement of iron concentration and discrimination of Fe^{+2} and Fe^{+3} breakdown from an InP:Fe absorption spectrum.

Measured σ^o data is considered to be more reliable than the model, even when the measured data is scaled to a new temperature. Takanohashi and Nakajima's data can be scaled from 77°K to 300°K assuming that trends in σ_p^o can be modeled with the composite Lucovsky model:

$$\begin{aligned} \sigma_p^o(\lambda, 300\text{K}) = & +4.8 \times 10^{-16} \frac{(h\nu - 0.705)^{\frac{3}{2}}}{(h\nu)^3} \\ & + 1.5 \times 10^{-14} (h\nu - 1.04)^2 \times \frac{\exp\left(\frac{h\omega_g}{k_B T (77^\circ\text{K})}\right) - 1}{\exp\left(\frac{h\omega_g}{k_B T (300^\circ\text{K})}\right) - 1} \end{aligned} \quad (3.38)$$

The first term (VB→5E) changes because the transition energy changes. Nothing else is needed when considering such an allowed direct transition. The second term changes because both the transition energy changes and because allowed indirect transitions (ie VB→5T2) have the indicated exponential temperature dependence.

The trends in σ_n^o can be modeled in a similar form:

$$\sigma_n^o(\lambda, 300K) = +1.1 \times 10^{-17} \frac{(h\nu - 0.63)^{\frac{3}{2}}}{(h\nu)^3} + (2 \times 10^{-15}(h\nu - 1.095)^2 + 5 \times 10^{-14}(h\nu - 1.18)^2) \times \frac{\exp\left(\frac{h\omega_g}{k_B T (77^\circ K)}\right) - 1}{\exp\left(\frac{h\omega_g}{k_B T (300^\circ K)}\right) - 1} \quad (3.39)$$

The photoionization cross-section due to allowed direct transition $5E \rightarrow \Gamma_1$ is unaltered with the temperature change. Indirect transitions $5E \rightarrow L_1$ and $5E \rightarrow X$ are altered using the same scaling factor as in the hole photoionization cross-sections in the last equation. Strait et. al. did not take all this into account. Instead they were interested in σ^o for $\lambda > 1.3 \mu m$, and they claimed that there is not much difference between σ_p^o at $77^\circ K$ and $300^\circ K$.

The cross-section models are used to produce theoretically calculated values for σ_p^o as shown in Table 3.2 and for σ_n^o as shown in Table 3.3 for various wavelengths. Other workers' experimentally measured values are shown in Table 3.4. The trends are depicted in Figure 3.4 for $77^\circ K$ and $300^\circ K$ [74]. The data points are scaled from Takanohashi's paper. For $300^\circ K$ σ^o is larger for holes but smaller for electrons. The difference for the holes is that the relevant transition energies are 0.07 eV smaller for holes at $300^\circ K$ than they are at $77^\circ K$ due to the falling valence band energy level. The difference for electrons is that even though the transition energies stay approximately the same, the absorption coefficient corresponding to electron transitions as allowed indirect processes have the following general temperature dependence on their multiplicative coefficients:

$$\alpha = constant \times \frac{(h\nu - E_i + h\omega_g)^2}{\exp\left(\frac{h\omega_g}{k_B T}\right)} \quad (3.40)$$

The temperature change from $77^\circ K$ to $300^\circ K$ results in the absorption coefficients due to Fe^{+2} for some transitions falling to twelve percent of the $77^\circ K$ values.

Table 3.2 Theoretical Calculated Hole Photoionization Cross-section in InP:Fe.

λ [μm]	$h\nu$ [eV]	σ_p° [10^{-18}cm^2]					
		$\Gamma_{15} \rightarrow SE$		$\Gamma_{15} \rightarrow 5T_2$		TOTAL XS	
		300K $\rightarrow 0.71\text{eV}$	77K $\rightarrow 0.78\text{eV}$	300K $\rightarrow 1.06\text{eV}$	77K $\rightarrow 1.13\text{eV}$	300°K	77°K
1.064	1.17	94	74	235	46	329	120
1.152	1.08	88	64	24	0	112	64
1.320	0.94	66	39	0	0	66	39
1.550	0.80	27	3.7	0	0	27	3.7

Table 3.3 Theoretical Calculated Electron Photoionization Cross-Section in InP:Fe.

λ [μm]	$h\nu$ [eV]	σ_n° [10^{-18}cm^2]							
		$5E \rightarrow \Gamma_1$		$5E \rightarrow L_1$		$5E \rightarrow X$		TOTAL XS	
		300K $\rightarrow$ 0.63eV	77K $\rightarrow$ 0.63eV	300K $\rightarrow$ 1.13eV	77K $\rightarrow$ 1.13eV	300K $\rightarrow$ 1.25eV	77K $\rightarrow$ 1.25eV	300°K	77°K
1.064	1.17	2.7	2.7	1.2	10	0	0	3.9	12.7
1.152	1.08	2.6	2.6	0	0	0	0	2.6	2.6
1.320	0.94	2.3	2.3	0	0	0	0	2.3	2.3
1.550	0.80	1.5	1.5	0	0	0	0	1.5	1.5

Table 3.4 Experimental Measured Photoionization Cross-section in InP:Fe.

Valley:[73], Takanohashi:[69].

λ [μm]	$h\nu$ [eV]	σ_p° [10^{-18}cm^2]		σ_n° [10^{-18}cm^2]	
		77°K	300°K	77°K	300°K
1.064	1.17		310 Valley		4.0 Valley
		120 Takanohashi	320 scaled	15. Takanohashi	
1.152	1.08	70 Takanohashi	130 scaled	2.8 Takanohashi	
1.320	0.94	38 Takanohashi	63 scaled	2.2 Takanohashi	
1.550	0.80	4 Takanohashi	16 scaled	1.5 Takanohashi	

3.8 Absorption

Optical spectral absorption in semiconductors is a technique for measuring deep level concentrations. Densities such as [Fe] in InP:Fe, [EL2] in GaAs:EL2, and [Cr] in GaAs:Cr are all available through the spectra, assuming that the intrinsic material

absorption is negligible, which is generally true in these media in the $1.0\ \mu\text{m} \leq \lambda \leq 1.6\ \mu\text{m}$ band. Intrinsic free carrier absorption may be neglected when the free electron concentration is less than $10^{18}\ \text{cm}^{-3}$ [75]. That is the case for all SI materials.

This section presents the end results of the absorption chapter. After considering all of the atomic transitions and lattice details, absorption spectra are interpreted. A Perkin Elmer spectrophotometer result for InP:Fe absorption spectra is presented. The photoionization cross-sections of InP:Fe are related to the absorption spectra. Two techniques for extracting [Fe] are demonstrated, and a problem is outlined.

3.8.1 InP:Fe Absorption Spectrum

The measured optical absorption coefficient of InP:Fe falls monotonically as λ increases in the 0.9 to 1.6 μm band, as shown by the data points in Figure 3.6. A thin 0.080 cm thick sample of Crystacomm InP:Fe was used. The relatively thin sample was used in order to observe the band edge effects near 950 nm. A thicker sample would obscure the large optical absorption coefficients in the area due to the limited ≈ 40 dB dynamic range of the Perkin Elmer spectrophotometer. A thicker sample could be used to find more accurate spectra in the $\lambda > 1.4\ \mu\text{m}$ range.

The sample in the absorption measurement came from the bottom of the LEC boule from which the thicker sample used later on in the photorefractive measurements was cut. Both samples came from within one centimeter of the edge in the same quadrant of the five centimeter diameter boule. The physical proximity of the two samples gives one some confidence in the similarity of electrical and optical characteristics between the thin absorption sample and the thicker sample in which photorefractive experiments were made.

Figure 3.6 shows that the band gap wavelength at $\lambda = 0.929\ \mu\text{m}$ is close to the last data point where $\lambda = 0.948\ \mu\text{m}$ and $\alpha = 137.4\ \text{cm}^{-1}$. At $\lambda = 1.064\ \mu\text{m}$, $\alpha = 0.93\ \text{cm}^{-1}$, and by $\lambda = 1.600\ \mu\text{m}$, $\alpha = 0.09\ \text{cm}^{-1}$. The absorption was measured in a Perkin Elmer Lambda 9 spectrometer using 1 nm steps, 2 second settling time, and the data is unsmoothed. To the first order, surface reflections were eliminated by tilting the crystal by $\approx 15^\circ$ from normal incidence. Reflection and wavelength dependent refractive index variation were taken into

Figure 3.6 - Experimental data points "▽" and best RMS fit line to the scaled photoionization model for absorption in InP:Fe at 300°K. $[\text{Fe}^{+2}]/\{[\text{Fe}]\} = 0.92 \pm 0.02$.

account by updating the index in post-measurement spreadsheet manipulations for every 10 nm, using published and interpolated refractive index data.

Consider the InP:Fe crystal in the spectrophotometer. The crystal has refractive index n , and absorption coefficient α at wavelength λ . The spectrophotometer measures absorbance A , the total loss from the crystal due to absorption, reflection, and scattered light. The 9x9 mm cross-section crystal was polished, so scattered light was not expected to be important. The maximum cross-section of the crystal was presented to the beam to allow the maximum amount of light to pass through the sample. By using the largest practical optical aperture in the Perkin Elmer spectrophotometer given the limited sample size, the useful dynamic range of the experimental results was maximized.

A portion T of the light intensity incident from the air on the crystal is transmitted into the crystal medium:

$$T = \frac{4n}{(n+1)^2} \quad (3.41)$$

When the light reaches the other side of the crystal of thickness L , the fraction of light remaining T_1 is:

$$T_1 = T e^{-\alpha L} \quad (3.42)$$

The fraction of light emerging from the crystal T_2 is:

$$T_2 = T^2 e^{-\alpha L} \quad (3.43)$$

A certain amount of T_1 is reflected back into the crystal. This component reflects back and forth inside the crystal and produces a significant reading on the spectrophotometer when the refractive index is large. The multiple reflections result in a net value of transmission through the crystal T_∞ , a Taylor series that reduces to:

$$T_\infty = \frac{T^2 e^{-\alpha L}}{1 - T^2 e^{-2\alpha L}} = 10^{-A} \quad (3.44)$$

where A is in units of decibels/10. By manipulating the equation for T_∞ into a quadratic form, one solves for the absorption coefficient α :

$$\alpha = -\frac{1}{L} \ln \left(\frac{-10^4 + \sqrt{10^8 + \frac{4}{T^2}}}{2} \right) \quad (3.45)$$

This equation produced values of α that were smaller than observed α at $\lambda = 1.064 \mu\text{m}$, $1.152 \mu\text{m}$, and $1.32 \mu\text{m}$. In addition, absorption spectra from the equation had large mid-band deviations from the predicted photoionization based absorption spectra. The form has problems because of imperfections in the crystal and the crystal's tilt, which removed most of the reflections from the spectrophotometer's entrance pupil. Considering only the first reflection produced a much closer fit to the measured laser absorptions and to the modeled absorptions:

$$\alpha = \frac{1}{L} \ln(10^4 T) \quad (3.46)$$

The simplified equation for α assumes that the incident beam from the spectrophotometer makes only one transit through the crystal - in other words, multi-reflections are negligible. The initial reflection at the start of the InP:Fe crystal is considered in the "T" term, but other reflections are neglected.

The raw absorbance data showed some noise near $\lambda = 1400 \text{ nm}$, presumably due to a small reflection resonance in the sample. The noise apparently comes from multipass reflections that start to become significant to the optical transmission in the low absorption wavelength region for certain bands, such as observed at $\lambda = 1400 \pm 50 \text{ nm}$ and $\lambda = 2800 \pm 100 \text{ nm}$. The noise is not evident in the data used in Figure 3.5 due to undersampling the hundreds of data points in the Perkin Elmer spectrophotometer's data output for easier manipulation of the data.

The falling α as a function of λ is the result of the afore-mentioned decreasing photoionization cross-sections of both electrons and holes in this band. To first order, the optical absorption coefficient α is equal to:

$$\alpha = [\text{Fe}^{+3}] \sigma_p^0 + [\text{Fe}^{+2}] \sigma_n^0 \quad (3.47)$$

where it is assumed that only an iron impurity exists in the band gap to contribute to

absorption, and there is no intrinsic absorption in InP. The value of α in InP:Fe at several common wavelengths is given in Table 3.5.

Table 3.5 Measured Absorption Coefficient for InP:Fe at 300°K.

λ (μm)	ABSORPTION COEFFICIENT (cm^{-1})
1.064	0.93
1.152	0.41
1.320	0.21
1.550	0.10

An equivalent expression for $\alpha(\lambda)$ is obtained by introducing the Fe^{+2} fraction

f:

$$\alpha(\lambda) = \{\sigma_n^o(\lambda)f + \sigma_p^o(\lambda)(1-f)\}[Fe] \quad (3.48)$$

where:

- σ_n^o = photoionization cross-section of Fe^{+2} donor
- σ_p^o = photoionization cross-section of Fe^{+3} acceptor
- f = fraction of Fe in Fe^{+2} donor form = $[\text{Fe}^{+2}]/[\text{Fe}]$
- 1-f = fraction of Fe in Fe^{+3} acceptor form = $[\text{Fe}^{+3}]/[\text{Fe}]$
- [Fe] = total concentration of Fe

To obtain an estimate of Fe^{+2} fraction f, one uses the model for α and Takanohashi and Nakajima's measured cross-sections (scaled to 300°K) of the $\text{Fe}^{+2}/\text{Fe}^{+3}$ deep level. A best RMS fit to the α versus λ curve is obtained by varying f. First, before finding f, one uses a special wavelength to find bounds for the total iron concentration [Fe]. The special wavelength is explained in the next section.

3.8.2 Magic Wavelength

There is a "magic" wavelength which varies as a function of temperature where the optical absorption cross-sections of the deep level traps Fe^{+2} and Fe^{+3} are equal. For example, at 77°K, from Takanohashi and Nakajima's data, $\sigma_n^o = \sigma_p^o = 4.5 \times 10^{-16} \text{ cm}^2$ for $\lambda = 0.976 \mu\text{m}/1.27 \text{ eV}$. For a given temperature, at the magic wavelength, the optical absorption coefficient leads directly to the total Fe concentration [Fe], assuming that only

Figure 3.7 Estimated iron concentration spectrum in InP:Fe versus wavelength.

iron transitions are responsible for optical absorption at the wavelength of interest. One solves for [Fe] using this magic wavelength calculation:

$$[Fe] = [Fe^{+2}] + [Fe^{-3}] = \frac{\alpha}{\sigma^0} \quad (3.49)$$

Figure 3.7 depicts the spectrum of possible solutions to this equation at 300°K. The curve labeled "HOLES" is the curve using σ_p^0 , and the curve labeled "ELECTRONS" uses σ_n^0 . The curves will cross where $\sigma_p^0 = \sigma_n^0$. At this point the ordinate indicates the iron concentration. Note that the curves do not cross at 300°K for InP:Fe due to the temperature variation of the photoionization cross-sections. The crystal would have to be cooled to 77°K measure [Fe] exactly using this technique. However, it can be observed that the curves approach each other. They approach because the cross-sections are within a factor of ten of each other for $\lambda < 0.960 \mu\text{m}$. At 77°K there is no problem because for $\lambda = 0.976 \mu\text{m}$ σ^0 is $4.5 \times 10^{-16} \text{ cm}^2$ for both electrons and holes. For the purposes of this thesis, the spectral measurement at 77°K was not possible given the available experimental equipment.

Although the curves do not cross at 300°K, it is possible to obtain a ball park estimate of [Fe] near the magic wavelength. Extrapolating the ELECTRONS curve between $\lambda = 950 \text{ nm}$ and $\lambda = 1000 \text{ nm}$ to the holes curve indicates a value of [Fe] between $2 \times 10^{16} \text{ cm}^{-3}$ and $10 \times 10^{16} \text{ cm}^{-3}$. The estimate is close to Crystacomm's SIMS estimate that nominal doping is $6 \times 10^{16} \text{ cm}^{-3}$ for the bottom of one of their LEC boules. Crystacomm claims that the nominal [Fe] at the seed of the LEC boule is $0.9 \times 10^{16} \text{ cm}^{-3}$. [Fe] increases towards the bottom of the boule (the tang) and it is not expected to exceed the 10^{17} cm^{-3} precipitation level.

3.8.3 $[Fe^{+2}]/[Fe]$

Given the measured absorption spectrum and the photoionization model for the absorption spectrum, varying [Fe] and $f = [Fe^{+2}]/[Fe]$ as parameters yields fits of varying goodness to the measured spectrum. The goodness is measured using a normalized RMS fit equation:

$$\epsilon_{RMS} = \sqrt{\frac{\sum_{i=1}^N \left(\frac{\alpha_{experimental}(\lambda_i) - \alpha_{model}(\lambda_i)}{\alpha_{experimental}(\lambda_i)} \right)^2}{N}} \quad (3.50)$$

Figure 3.6 shows measured absorption spectrum data points against the best fit absorption model line, where the fit considered all data points between $\lambda = 1000$ and $\lambda = 1400$ nm. The fit was evaluated by varying $[Fe]$ from 10^{15} to $10 \times 10^{16} \text{ cm}^{-3}$. At each value of $[Fe]$, f is varied between 0 and 1. The scaled RMS error varied from 0.14 to 0.28. For example, if $[Fe]$ were taken to be $10 \times 10^{16} \text{ cm}^{-3}$, then $f = 0.97$ minimizes the RMS error at 0.28.

There was a broad error floor for $3 \times 10^{15} \leq [Fe] \leq 4 \times 10^{16} \text{ cm}^{-3}$ where the error was ≤ 0.14 . Since the wafer comes from the tang, one takes the largest value of $[Fe]$ that minimizes the RMS error. The calculated value of $[Fe]$ is $3.7 \pm 0.3 \times 10^{16} \text{ cm}^{-3}$. At this point, $f = [Fe^{+2}]/[Fe] = 0.92 \pm 0.01$.

Most of the iron - 92 percent - is reduced to Fe^{+2} . The result is not expected. One expects that larger values of $[Fe]$ in the tang should suck up all the stray electrons and result in mostly Fe^{+3} and a very small fraction of Fe^{+2} . One would have thought that the Fe^{+2} fraction would be less, for example, near ten percent instead of 92 percent. The data indicates that only eight percent of the iron is free to trap CB electrons. The relatively large Fe^{+2} concentration may be because of other effects at the bottom of the boule. At the bottom of the boule one finds the largest concentration of defects and impurities from the melt, resulting from drawing the boule. The extra defects and impurities generate more free electrons and fill the Fe^{+3} acceptors. Fe^{+3} is converted into more Fe^{+2} than is the case throughout most of the boule. One could test this hypothesis by doing absorption spectra for wafers from the top and the bottom of an InP:Fe boule. A qualitative shift in the iron tail of the spectrum would indicate a change from Fe^{+2} to Fe^{+3} domination.

Figure 3.6 shows reasonable agreement for this fit, with some larger errors for the low energy tail $1500 \leq \lambda \leq 1600$ nm. The problem lies in lower energy unmodeled processes becoming dominant in that range.

The modeled iron tail spectrum alone rises too slowly as one approaches the band gap. The error is due to intrinsic bandgap absorption mediated by optical phonons ($hw_q = 0.041$ eV for 10 THz TO phonons). To model the measured data, two VB→CB transitions were introduced: one allowed indirect transition and one forbidden indirect transition. The energies of the transitions were calculated by:

$$E_{vb \rightarrow cb} = E_g - k_B T - hw_q = 1.34 - 0.026 - 0.041 = 1.27 \text{ eV} \quad (3.51)$$

The resulting absorption model is:

$$\begin{aligned} \alpha(\lambda) = & \sigma_n^o(\lambda)[Fe^{+2}] + \sigma_p^o(\lambda)[Fe^{+3}] \\ & + 1.5 \times 10^5 \left(\frac{hc}{\lambda} - 1.28 \text{ eV} \right)^2 + 2 \times 10^7 \left(\frac{hc}{\lambda} - 1.29 \text{ eV} \right)^3 \text{ cm}^{-1} \end{aligned} \quad (3.52)$$

Even though the sample used for absorption measurement was only 0.080 cm thick, no exciton edge was seen in the InP:Fe near the band gap energy because the loss through the sample near the band gap exceeded the Perkin Elmer spectrometer's dynamic range. The exciton edge is supposed to occur around 1000 cm^{-1} while the best one could measure even with the thin sample was $\approx 100 \text{ cm}^{-1}$. The exciton edge would become visible given the existing equipment if one used a piece of polished InP:Fe that was less than 0.008 cm (80 μm) thick.

3.8.4 Comparison with Other Work

It is worthwhile to observe that the InP:Fe samples used by Ozkul et. al. [27] had measured α between 1.5 and 2.6 cm^{-1} , while the NRC sample is at 0.93 cm^{-1} at $\lambda = 1.064 \mu\text{m}$. The NRC sample was cut from the bottom of an LEC grown boule, where the iron solution is near a maximum concentration before precipitating. Originally it was expected that the bottom would yield the largest values of absorption for the boule. The thesis, however, has shown that it may be expected to instead reduce the absorption due to Fe^{+2} dominating at the bottom.

Discussions with the crystal grower⁵ revealed that many researchers in

⁵ George Antypas of Crystacomm, Mountain View, California.

photorefractive InP:Fe use the top of the boule instead of the bottom of the boule. The reason for this is that although optical absorption may be less at the top since defects, unwanted impurities, and thence sites for [Fe] increase from seed to tail in a boule, Fe^{+3} concentration is increased at the seed. The improvement is marked for increasing the Fe^{+3} concentration relative to the Fe^{+2} concentration. Therefore on the basis of $[\text{Fe}^{+3}]$, the seed seems to be the source of choice for photorefractive crystals. However, there may be other factors making a better quality crystal nearer the seed than towards the tang.

Later on in this thesis $[\text{Fe}^{+2}]/[\text{Fe}]$ is measured using a different technique. It is observed that under laser illumination at $\lambda = 1.06 \mu\text{m}$, for $I_0 = 4 \text{ mW/cm}^2$, $[\text{Fe}^{+2}]$ is 0.57 of the total iron concentration. The fall in $[\text{Fe}^{+2}]$ from $f=0.92$ can be explained by optical excitation driving electrons out of the deep level and into the conduction band, changing Fe^{+2} into Fe^{+3} .

3.9 Summary

In this chapter optical absorption in InP:Fe was reviewed. Chemistry, band structure, thermal, and optical transitions were considered. A sample of Crystacomm InP:Fe was compared with published photoionization cross-section and material parameters were extracted. The sample is found to exhibit the following absorption-related parameters at 300°K in the dark:

α	=	0.93	cm^{-1} at $\lambda = 1.064 \mu\text{m}$.
$[\text{Fe}]$	=	3.7×10^{16}	cm^{-3}
$[\text{Fe}^{+2}]/[\text{Fe}]$	=	0.92	[unitless]
$[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$	=	11.5	[unitless]
$[\text{Fe}^{+2}]$	=	3.4×10^{16}	cm^{-3}
$[\text{Fe}^{+3}]$	=	0.3×10^{16}	cm^{-3}
σ_p^∞	=	3×10^{-16}	cm^2
σ_n^0	=	1.7×10^{-18}	cm^2
σ_n^∞	=	3.5×10^{-14}	cm^2
σ_p^0	=	3.3×10^{-16}	cm^2

In general, one can find bounds to [Fe] in InP:Fe by determining the absorption coefficient at or near the "magic" wavelength $\lambda = 0.976 \mu\text{m}$ at 77°K. There the photoionization cross-sections of iron Fe^{+2} and Fe^{+3} are equal: $\sigma^0 = \sigma_n^0 = \sigma_p^0 = 4.5 \times 10^{-16} \text{ cm}^2$

at 77°K. [Fe] is calculated by dividing α by σ^0 , assuming only deep level components to optical absorption are present.

Without the "magic" wavelength, iron concentration in InP:Fe cannot easily be measured with a single optical absorption measurement, unless all of the iron is in a known charge state (Fe^{+2} or Fe^{+3}). Optical measurement of [Fe] requires a least squares fit of known $[\text{Fe}^{+2}]$ and $[\text{Fe}^{+3}]$ absorption spectra to the measured crystal spectra.

CHAPTER 4

PHOTOREFRACTIVE PARAMETERS

This chapter considers photorefractive 2WM in InP:Fe. The two most important experimental and theoretical parameters, time constant τ and gain Γ , are considered based on the Kukhtarev equation (**Chapter 2**) and the deep level optical absorption model (**Chapter 3**). Time constant is considered first because of the logical chain of events from absorption in the previous chapter into other processes that lead to photorefractive gain.

4.1 Time Constant in InP:Fe

4.1.1 Introduction

The $1/e$ time to set up a photorefractive grating is the photorefractive time constant τ . As experimentally measured, it depends not only on the presence of the photorefractive grating but also on the overlapping photochromic grating. τ depends on the physics of both these gratings, involving material parameters and external variables such as I_0 and E_0 . The presence of electrons and holes with different photoionization cross-sections, different mobilities, and different recombination probabilities adds another dimension to τ .

A photorefractive material may exhibit three kinds of volume holograms: amplitude holograms due to photochromic gratings, phase holograms due to photorefractive gratings, and amplitude/phase holograms due to combined photochromic and photorefractive gratings [76]. Both types have been observed in InP:Fe and in GaAs:Cr. The two gratings have different time constants because the photochromic grating is formed as the first step in the photoexcitation, charge transport, trap, and Pockels effect sequence from which a photorefractive grating is created. The photochromic grating always has a shorter time constant than the photorefractive grating since the absorption process leads any photorefractive result. The steady state contribution of the photochromic grating may surpass the contribution of the photorefractive index grating even when conditions favour the photorefractive effect.

Each step in the photorefractive effect has its own individual additive effect on the overall time constant of a photorefractive index grating. In practice, most researchers assume that the transient processes obey an exponential law with one time constant [77] [78]. Recently Osipov and Sturman suggested that one time constant is only partially applicable to grating recording [79]. Observations and theory show that the transient effects in 2WM energy transfer may be oscillatory overdamped or underdamped solutions to the Kukhtarev coupled wave equations, which were discussed in Chapter 2. The transient effects are particularly important when the interbeam angles are small or when the input beam intensities are appreciably different, for example in the case of a photorefractive amplifier.

4.1.2 Fundamental Speed Limits

There are four fundamental lower limits involved in the response time of the PRE. They are based on the sequence of four fundamental processes involved in the photorefractive effect: photoexcitation, transport, trap, and the Pockels effect [80] [81]. If there is more than one charge carrier involved in the process, the number of time constants doubles to eight time constants. Some observations in the course of the thesis work indicated at least two of these time constants involved in PR grating formation. Electron and hole effects apparently "competed" for photorefractive gain as the direction of optical energy transfer first went one way, and then settled on the other. The independent processes are considered one-by-one below. Later the combined results of the fundamental limits are considered.

4.1.2.1 #1: Photoexcitation

The time required after the pump beam is turned on to have the number of absorption events equal the number of free carriers in a photorefractive grating with optical gain Γ is given by Yeh [82] [83]. The analysis is based on uniform light incident on an InP crystal with absorption coefficient α_p . The light causes N_p photoexcitation events per unit volume per second. At some time t after turning on the 2WM beams of combined mean intensity I , neglecting recombination, N_p is:

$$N_p = \frac{I\alpha_p t}{h\nu} \quad (4.1)$$

A certain number of these charges do not recombine and contribute to useful space charge separation for 2WM. The number of available charges N depends on quantum efficiency η , where $0 \leq \eta \leq 1$. Usually $\eta \approx 0.5$. N is:

$$N = \frac{I \alpha_p t}{h\nu} \eta \quad (4.2)$$

The 2WM interference pattern forms a simple periodic charge density distribution of amplitude ρ_0 (forgetting about phase angle with respect to the optical excitation for this subsection):

$$\rho(x) = \rho_0 \cos\left(\frac{2\pi x}{\Lambda}\right) \text{ where } \rho_0 = \pi e N = \pi I \alpha_p \frac{e}{h\nu} \eta t \quad (4.3)$$

Using the Poisson equation $\partial^2 V / \partial x^2 = -\rho(x)/\epsilon$, $\rho(x)$ is integrated to solve for the space charge electric field $E_{sc}(x)$:

$$E_{sc}(x) = \int \rho(x) dx = -\frac{I \alpha_p}{2} \frac{e}{h\nu} \frac{\Lambda \eta t}{\epsilon} \sin\left(\frac{2\pi x}{\Lambda}\right) \quad (4.4)$$

The Pockels effect relates E_{sc} to a change in refractive index Δn . Using equation (1.5) the depth of the phase hologram Δn is:

$$\Delta n = -\frac{1}{2} n^3 r_{41} E_{sc} = \frac{I \alpha_p}{4} \frac{e}{h\nu} \Lambda \eta Q t \quad (4.5)$$

The "Q" variable is the usual sensitivity figure of merit for photorefractive media. Q is valid for media performance comparisons for $\Lambda \gg$ Debye length, and is given in equation (4.6):

$$Q = \frac{n^3 r}{e} \quad (4.6)$$

where the variables are:

n	=	refractive index
	=	3.29 in InP:Fe at $\lambda = 1.064 \mu\text{m}$
	=	3.48 in GaAs at $\lambda = 1.064 \mu\text{m}$
r_{41}	=	appropriate electro-optic coefficient for given geometry
	=	1.34×10^{-12} meter/volt in InP:Fe at $\lambda = 1.064 \mu\text{m}$
	=	1.43×10^{-12} meter/volt in GaAs at $\lambda = 1.064 \mu\text{m}$
ϵ	=	medium permittivity
	=	$\epsilon_0 \times 8.854 \times 10^{-12}$ Farad/meter
ϵ_0	=	medium dielectric constant

- = 12.4 in InP:Fe
- = 12.9 in GaAs

In InP:Fe, $Q=0.43 \text{ m}^2/\text{Coulomb}$, and for GaAs, $Q=0.53 \text{ m}^2/\text{Coulomb}$ at $\lambda=1.064 \text{ }\mu\text{m}$.

Γ is the photorefractive coupling constant - the gain. It ranges between 0 to 20 cm^{-1} . The maximum diffusion limited gain Γ at any time t is a function of Δn and λ :

$$\Gamma(t) = \frac{2\pi\Delta n}{\lambda} = \left(\frac{e}{h\nu}\right)\left(\frac{\Lambda}{\lambda}\right)\alpha_p \frac{\pi\eta I_0 Q}{2} t \quad (4.7)$$

As a result, the time required after the pump beam is turned on to have the number of absorption events equal the number of free carriers in a photorefractive grating is:

$$\tau(\Gamma) = \left(\frac{h\nu}{e}\right)\left(\frac{\lambda}{\Lambda}\right)\left(\frac{\Gamma}{\alpha_p}\right)\left(\frac{2}{\pi\eta I_0 Q}\right) \quad (4.8)$$

where the variables are:

- $h\nu$ = photon energy = $1.17 \text{ eV} = 1.87 \times 10^{-19} \text{ Joules}$ typically
- λ = wavelength = $1.064 \text{ }\mu\text{m}$ typically
- Λ = grating spacing = 1 to $20 \text{ }\mu\text{m}$ typically
- α_p = photorefractive absorption coefficient
- = 0.93 cm^{-1} in InP:Fe at $\lambda=1.064 \text{ }\mu\text{m}$
- = 0.41 cm^{-1} in InP:Fe at $\lambda=1.152 \text{ }\mu\text{m}$
- I_0 = optical intensity = 5 mW/cm^2 typically

Experimentally, 2WM measurements involve finding the ratio between the steady state signal beam intensity with the pump and the signal beam intensity without the pump. The ratio is γ . It is related to the photorefractive gain coefficient Γ by [84]:

$$\gamma = \frac{I_s(\text{with pump})}{I_s(\text{without pump})} = \exp(\Gamma L) \therefore \Gamma = \frac{1}{L} \ln(\gamma) \quad (4.9)$$

where:

- L = interaction length of the crystal

and from this expression one also has the beam coupling gain coefficient Γ .

Equation (4.8) says that the photon limited response time is equal to the energy required to write a grating with a specified coupling constant Γ divided by the intensity I_0 . Equation (4.8) says, inter alia, that the photorefractive response time is inversely

PHOTOREFRACTIVE RESPONSE TIME

Figure 4.1 Photon-limited response time of the photorefractive effect in InP:Fe for various values of gain Γ , for $\Lambda=10\text{ }\mu\text{m}$ and $\lambda=1.064\text{ }\mu\text{m}$.

proportional to the optical intensity. At low pump intensities, PRE will take a long time to respond. For example, see in Figure 4.1 where the theoretical photon limited response time in InP:Fe is plotted versus intensity for five values of Γ over the experimental range of intensity 0.01 to 100 mW/cm². The parameters are $\lambda = 1.064 \mu\text{m}$, $\Lambda = 10 \mu\text{m}$, $0.1 \leq \Gamma \leq 10.0 \text{ cm}^{-1}$, and quantum efficiency $\eta = 0.5$. Assume $\Gamma = 10 \text{ cm}^{-1}$. At $I_0 = 1 \text{ mW/cm}^2$, InP:Fe takes $\approx 0.5 \text{ msec}$ to produce the required number of carriers to populate the required charge clouds. At $I_0 = 10 \text{ mW/cm}^2$, InP:Fe takes $\approx 0.05 \text{ msec}$ to respond. At $I_0 = 100 \text{ mW/cm}^2$, InP:Fe takes $\approx 0.005 \text{ msec}$ to respond. No matter how fast the other three processes leading to photorefractive are, the response time for the four combined processes cannot be less than the absorption-limited response time.

4.1.2.2 #2: Transport

The second important limit of photorefractive response time is the time required for carriers to displace from the standing wave optical intensity peaks far enough to form a periodic space charge electric field, E_{sc} [85]. The distance is about half a grating period, where $1 \leq \Lambda \leq 10 \mu\text{m}$. The main movement is from the intensity peaks into the intensity valleys, a distance of $\Lambda/2$. Assume room temperature mobility of 1470 cm²/V-s (electrons) and 140 cm²/V-s (holes) in InP:Fe, and 5000 cm²/V-s (electrons) and 400 cm²/V-s (holes) in GaAs [86]⁶. There are two independent transport times: one due to drift (movement in an electric field) called τ_{drift} , and another due to diffusion (random movement) called τ_D . They are discussed below.

The drift transport time τ_{drift} depends on the electric field, mobility, and the grating spacing. Given $\Lambda = 1 \mu\text{m}$ and an electric field $E_{sc} = 5 \text{ kV/cm}$ field, the drift time is on the order of 34 psec (electrons)/ 360 psec (holes) in InP:Fe, and 10 psec (electrons)/ 125 psec (holes) in GaAs, where equation (4.10) is used:

$$\tau_{\text{drift}} = \frac{\Lambda}{2\mu E_{sc}} \quad (4.10)$$

⁶ The thesis attempted mobility measurements on SI InP:Fe and GaAs samples from the same wafers that the 2WM samples were cut. Diffused ohmic contacts were grown and tested on van der Pauw geometry samples. The resistivity was so large that the Hall voltage was lost in Johnson noise. The problem and a solution are described in the Conclusion.

The equation is a simple time equals distance ($\Lambda/2$) divided by speed (μE_{sc}) formula.

The diffusion transport time τ_D depends on mobility and grating spacing. Valley calculated the electron diffusion time in GaAs:EL2 at $\lambda = 1.06 \mu\text{m}$ and predicts $\tau_D \approx 5$ psec [87] (electrons)/ 77 psec (holes) and in InP:Fe, 21 psec (electrons)/ 222 ps (holes):

$$\tau_D = \frac{\Lambda^2}{4\pi D_n} \quad (4.11)$$

where D_n is the diffusion coefficient for electrons as determined by the Einstein relation between mobility for a classical gas and diffusivity [88]:

$$\mu = \frac{e}{k_B T} D_n \quad D_n = \frac{k_B T}{e} \mu \quad (4.12)$$

If a faster response is required, the Einstein relation suggests and experimental observations indicate lower temperatures cause the mobility μ to increase, allowing even faster response. The response may be faster, but photorefraction may be lost. The physics of PRE can change completely with some phenomena starting up and others being curtailed. While results in InP:Fe at low temperatures are rare in the available literature, a number of low temperature observations have been performed in GaAs:EL2. For example, at $T = 11^\circ\text{K}$, Tajima et. al. [89] observed photoquenching of absorption in GaAs:EL2 at $\lambda = 1.064 \mu\text{m}$ due to the reaction:

Time constant ≈ 1.5 seconds @ $I_0 = 100 \text{ mW/cm}^2$

As a result of photoquenching, the material fails to absorb any more light and so the photorefractive phase grating is not sustained. The absorption recovered under illumination at $\lambda = 1.32 \mu\text{m}$ in the reaction:

Time constant ≈ 22 minutes @ $I_0 = 100 \text{ mW/cm}^2$

Low temperatures do provide some improved photorefractive results. For example, in GaAs:EL2 at low temperatures, photorefractive 4WM efficiency is enhanced by a factor of eight for a short period through a direct optical modulation of the Fermi level

for $60 < T < 140^\circ\text{K}$. The quenched grating is equivalent to a periodic "p-i-p" structure, since the illuminated (quenched) regions of the grating have their Fermi levels driven towards the valence band. The gradient in the Fermi level defines an electric field that works with diffusion to transport charge into the darker regions. Therefore in this temperature regime as EL2 is optically quenched, the Fermi level modulation leads to much larger values of E_{sc} [90]. Once EL2 is completely quenched, though, the diffraction efficiency drops off and the crystal is no longer semi-insulating.

Another cause of PRE quenching is a reservoir of charge in partially occupied defect/impurity levels which screens the electric field [91]. The reservoir of minority charges tends to redistribute through thermal and photoionization and recapture, in response to the majority carriers' local electric potential. Eventually, nonequilibrium occupation of such minority carrier traps may lead to complete screening of PRE. This has been observed in semi-insulating InP:Fe between 100 and 300°K [92].

Because PRE depends on either holes, electrons, or both holes and electrons simultaneously, the real transport time is more involved than the numbers given by equations (4.10) and (4.11). Mixed conduction becomes more complex, especially in InP:Fe where free electrons are produced both thermally and by optical absorption and while holes are produced by optical absorption but not thermally to any great extent. The two main modes of charge carrier transport - diffusion and drift - are discussed below for both electrons and holes.

4.1.2.2.1 Diffusion Only

Without an applied electric field, Brownian-motion type diffusion transports the charge carriers away from the optical intensity peaks. The diffusion length L_D of carriers is the FWHM distance that a charge carrier will travel before being trapped. The trap time is τ_R , and it represents the time constant for diffusion to move the charges by one Debye length away from the peak optical intensity point. It is discussed in a section below by itself. L_D depends on the temperature and on the mobility-lifetime product:

$$L_D = \left(\frac{k_B T \mu \tau_R}{e} \right)^{1/2} \quad (4.13)$$

4.1.2.2.2 Drift and Diffusion

With an applied electric field, longer average transport lengths are achieved by adding drift to diffusion. It takes the charge carriers longer to reach steady state, and larger space charge fields can be supported. As a result, larger 2WM gains can be supported with an applied electric field. The drift length L_E is equal to:

$$L_E = \mu \tau E_x \quad (4.14)$$

4.1.2.2.3 Photovoltaic Effect

In some photorefractive materials such as the ferroelectrics, the photovoltaic effect dominates PRE. However, in III-V semiconductors, the photovoltaic effect is not as important because they have higher dark conductivities σ_d , and no polar axis. In semi-insulating InP and GaAs, for example, $10^{-6} \leq \sigma_d \leq 10^{-8} (\Omega\text{-cm})^{-1}$. For LiNbO₃, $10^{-8} \leq \sigma_d \leq 10^{-19}$, and in BaTiO₃ $\sigma_d \approx 10^{-12} (\Omega\text{-cm})^{-1}$. Since the maximum value of E_{sc} is proportional to $1/\sigma_d$, E_{sc} is orders of magnitude smaller in InP than in the ferroelectrics, for the same L_{ph} and photovoltaic constant κ .

The polar axis plays an important role in photovoltaic phenomena. Ferroelectrics have a polar axis with dielectric constants on the order of 1000 compared with ≈ 10 on other axes, while III-V semiconductors are isotropic and to third order, have no polar axis. The largest observed photovoltaic effects rely on current on the polar axis.

Bulk photovoltaic effects are not important in semiconductors. It is negligible because the maximum photovoltaic carrier transport distance is just the mean carrier scattering length, and this distance is short compared to drift and diffusion lengths in semiconductors. In higher resistivity substances, especially ferroelectrics such as LiNbO₃ and BaTiO₃, photovoltaic transport is important, but not in semi-insulating semiconductors. For this reason, photovoltaic transport is ignored for the rest of the thesis.

Therefore, due to higher conductivity and no polar axis, photovoltaic effects are much less important to photorefractive in III-V semiconductors than in ferroelectrics. A search of INSPEC and available reference literature failed to turn up photovoltaic constants for InP or for GaAs. Therefore the photovoltaic effect in InP is assumed to be

negligible.

4.1.2.3 #3: Trap

The characteristic trap time τ_R is the time constant for the life of a free carrier in a semiconductor. It depends on the trap densities $[Fe^{+2}]$ & $[Fe^{+3}]$ and the recombination coefficients γ_R for electrons and holes. τ_R has a leading roll in the overall time constant of the photorefractive effect because it controls the diffusion lengths and the drift times as observed above.

Various authors state that the trap time for electrons τ_{Re} is related to the electron recombination coefficient γ_{Re} and the number of acceptors $[Fe^{+3}]$ by [93] [94] [95]:

$$\tau_{Re} = \frac{1}{\gamma_{Re}[Fe^{+3}]} \quad (4.15)$$

and similarly for holes: the trap time for holes τ_{Rh} is related to the hole recombination coefficient γ_{Rh} and the number of donors $[Fe^{+2}]$ by:

$$\tau_{Rh} = \frac{1}{\gamma_{Rh}[Fe^{+2}]} \quad (4.16)$$

These equations do not physically explain τ_R . The $[Fe^{+3}]$ term is understandable since one expects the time for a free electron to recombine to be inversely proportional to the number of acceptors. However, the γ_R number with its units of cm^3/sec is not explained. From physical considerations and balancing of units, γ_R is the product of the acceptor cross-section σ_A^e and the carrier thermal velocity v_{th} :

$$\gamma_R = \sigma_A^e v_{th} \quad (4.17)$$

and it leads to a physical basis for τ_R [96]:

$$\tau_R = \frac{1}{\sigma_A^e v_{th} [Fe^{+3}]} \quad (4.18)$$

For example in another III-V semiconductor material, consider GaAs:EL2 where traps $[EL2]$ and $[EL2^-]$ take the place of $[Fe^{+3}]$ and $[Fe^{+2}]$. Valley and Smirl estimated $\tau_{Re} \approx 38$ nsec and $\tau_{Rh} \approx 3.2$ μ sec in GaAs:EL2, where $[EL2] = 1.4 \times 10^{15} cm^{-3}$ and

$[EL2] = 1.3 \times 10^{15} \text{ cm}^{-3}$. Calculated τ_R for electrons is 38 nsec, and τ_R for holes is 2.8 μsec . The result is close to the published estimates. Valley and Smiri attributed various shorter recombination times in the literature to larger impurity concentrations in other samples [97].

Table 4.1 Characteristic Trap Time τ_R Model in InP:Fe

		ELECTRON TRAP: $[Fe^{+3}]$	HOLE TRAP: $[Fe^{+2}]$	
σ_A^0 recombination XS at infinite temperature		3.5×10^{-14}	3×10^{-16}	$[\text{cm}^2]$
v_{th} thermal velocity		4.1×10^7	1.6×10^7	$[\text{cm/sec}]$
N_A # acceptors		$[Fe^{+3}] = 5 \times 10^{16}$	$[Fe^{+2}] = 5 \times 10^{15}$	$[\text{cm}^{-3}]$
τ_R	CALC:	1.1 nsec	6.2 μnsec	
	EXP:	500 psec	2 μnsec	

Calculated values of γ_R are: $\gamma_R = 2.5 \times 10^{-9} \text{ cm}^3/\text{sec}$ for electrons and $2 \times 10^{-10} \text{ cm}^3/\text{sec}$ for holes. These numbers are close to the direct electron-hole recombination coefficient of $\gamma_R = 2 \times 10^{-10} \text{ cm}^3/\text{sec}$ as stated by Von Lehman and Ballantyne [98].

From nanosecond absorption measurements in InP:Fe at $\lambda = 1.064 \mu\text{m}$, Halter et. al. observe $\tau_{Re} \approx 500 \text{ psec}$ and $\tau_{Rh} \approx 2 \text{ usec}$ [99], for $[Fe^{+2}] \approx 5 \times 10^{15} \text{ cm}^{-3}$, and $[Fe^{+3}] \approx 5 \times 10^{16} \text{ cm}^{-3}$. Table 4.1 shows the results of the equation on Halter's parameter and a comparison is made between the experimental results and the equation for τ_R . The thermal velocities come from Picoli et. al. [100]. The calculated numbers are within a factor of three from the experimental values. Halter et. al. also observed linear absorption coefficients of $\alpha = 1.95 \text{ cm}^{-1}$ at $\lambda = 1.064 \mu\text{m}$, and $\alpha = 0.67 \text{ cm}^{-1}$ at $\lambda = 1.3 \mu\text{m}$ in their sample⁷. Because these results are based on pulsed measurements, the iron species concentrations

⁷ Some reported values of absorption are inconsistent with the photoionization cross section spectra detailed in Chapter 3. At $\lambda = 1.064 \mu\text{m}$, the InP:Fe absorption coefficient should be greater than 15 cm^{-1} , ten times the measurement of Halter et. al. The problem seems to lie in the Halter group using $\sigma_p^0 = 5 \times 10^{-17} \text{ cm}^2$ instead of $3.3 \times 10^{-16} \text{ cm}^2$. The issue will surface again in Chapter 6.

are changed by the light. The recombination times are expected to change in low intensity CW mode. In particular, τ_{Re} will become longer as $[Fe^{+3}]$ falls, and τ_{Rh} will become longer as $[Fe^{+2}]$ falls.

Recombination becomes important in GaAs:Cr on time scales longer than 100 psec [101]. Weiner and Yu measured recombination times in GaAs:Cr of 90 psec ($[Cr] = 1.6 \times 10^{17} \text{ cm}^{-3}$) $\leq \tau_R \leq 170$ psec ($[Cr] = 10^{15} \text{ cm}^{-3}$). Note that the larger the compensating level's concentration, the shorter the recombination time. This is because given increased $[Cr]$ gives electrons a larger density of free sites to recombine. Recombination time was 250 μ sec for GaAs:O ($[O] = 6 \times 10^{17} \text{ cm}^{-3}$), and 1200 psec for undoped GaAs ($N_A, N_D < 10^{14} \text{ cm}^{-3}$). Scaling similar results in InP:Fe leads to concluding that recombination is important for time scales longer than one nsec. Therefore, for the CW measurements in this thesis, recombination is always important.

4.1.2.4 #4: Pockels Effect

The response time of the linear EO Pockels effect, T_{EO} , is the last delay before the photorefractive index grating is formed and two beam coupling can then occur. T_{EO} is about 0.1 psec, short enough to be negligible in all reported CW measurements.

T_{EO} is approximately equal to the reciprocal of the fundamental transverse optical lattice vibration frequency f_{TO} . Physically interpreted, it says that the time for the unit cell's interatomic bonds in the crystal to stretch or contract in response to the space charge field is on the order of one lattice vibration period. This stretching/contraction of the atomic bonds is what causes the reduced/increased electron density, respectively. The changed electron density causes the refractive index to change proportional to the signed value of the space charge field. The refractive index depends on the crystal orientation, the light polarization, and the direction of the grating vector $\mathbf{K}$ through the index ellipsoid that results from the space charge field.

Typical ferroelectric oxides such as BaTiO₃ have measured TO lattice vibration frequencies in the range $1 \leq f_{TO} \leq 10$ THz, resulting in $0.1 \leq T_{EO} \leq 1$ psec [102]. Semiconductor materials such as InP:Fe and GaAs:Cr also fall in this range. Measured

PM-1 3½"x4" PHOTOGRAPHIC MICROCOPY TARGET
NBS 1010a ANSI/ISO #2 EQUIVALENT

PRECISIONSM RESOLUTION TARGETS

PKXNELN3 IN METHYLENE BLUE TESTING SINCE 1974

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phonon spectra for InP show the TO frequency varies $7.4 \leq f_{\text{TO}} \leq 9.5$ THz (0.030 to 0.038 eV), increasing from L to X to Γ oriented phonons [103]. The TO frequency results in delay $0.10 \leq T_{\text{EO}} \leq 0.14$ psec. Measured phonon spectra for GaAs show the TO frequency varies $7.7 \leq f_{\text{TO}} \leq 8.0$ THz (0.031 to 0.032 eV), for reduced wave number $0 \leq q/q_{\text{MAX}} \leq 1$ [104]. The result is a delay of $0.12 \leq T_{\text{EO}} \leq 0.13$ psec between a signal beam changing in amplitude and the pump amplifying the signal at its new amplitude.

The subpicosecond Pockels delay is negligible in all of the experiments with a low power CW signal and a pump chopped at a rate between 1 Hz and 100 Hz. However, it may be observed in other photorefractive experiments using powerful picosecond laser pulses and materials with very short transport times. Yeh claims that there are materials with transport times as short as 1 psec [105]. Valley et al have observed PRE in GaAs using 43 psec pulses [106], and in GaAs:EL2 and in InP:Fe using 29 psec pulses [107].

4.1.3 Drift-Diffusion Approximations

4.1.3.1 Photorefractive Response Time: τ Based Combined Expression

A more satisfactory estimate of the photorefractive response time τ_c combines the effects of the photoexcitation time, the transport time, and the trap time. To start with it is based on the one band transport model with a single charge carrier. The model considers the effects of the dielectric relaxation time, diffusion and drift as the sum of different additive periods.

This sort of combined expression for the photorefractive response time τ_c is presented by Kukhtarev [108], Valley, Klein [109], Gunter, and Huignard [110]. Derived from equation (2.22), the expression features a discrete term for each event in the photorefractive process, with the exception of the Pockels effect. The exemption of the Pockels effect is not important unless time scales of one picosecond are found in the experimental regime. These authors found that the transient response has an overdamped oscillatory behaviour. The analysis is valid for CW illumination, small index modulation, quasi-steady state in the conduction electron number density, and charge transport lengths that are equal to or longer than the grating period Λ . The $1/e$ response

time τ_e is:

$$\tau_e = \tau_{di} \frac{\left(1 + \frac{\tau_R}{\tau_D}\right)^2 + \left(\frac{\tau_R}{\tau_E}\right)^2}{\left(1 + \frac{\tau_R \tau_{di}}{\tau_D \tau_I}\right) \left(1 + \frac{\tau_R}{\tau_D}\right) + \left(\frac{\tau_R}{\tau_E}\right)^2 \left(\frac{\tau_{di}}{\tau_I}\right)} \quad (4.19)$$

where τ_R the recombination time is defined in equation (4.21) and Table 4.1. Note that there are two time constants in the one equation: one for the hole component of photorefraction, and the other for the electron component of photorefraction.

The oscillatory frequency of the space charge electric field is ω_e :

$$\omega_e = \frac{1}{\tau_{di}} \frac{\frac{\tau_R}{\tau_E} \left(\frac{\tau_{di}}{\tau_I} - 1\right)}{\left(1 + \frac{\tau_R}{\tau_D}\right)^2 + \left(\frac{\tau_R}{\tau_E}\right)^2} \quad (4.20)$$

The dielectric relaxation time τ_{di} is:

$$\tau_{di} = \frac{e_o \epsilon}{4\pi\sigma} = \frac{e_o \epsilon}{4\pi \left(\sigma_d + \frac{\partial \sigma}{\partial I_o} \times I_o \right)} \quad (4.21)$$

where σ_d represent the dark photoconductivity. 99 percent of the dark conductivity is attributed to electrons since they are the primary dark carrier in InP:Fe.

The drift time τ_E is:

$$\tau_E = \frac{\Lambda}{2\pi\mu_e E_o} \quad (4.22)$$

The diffusion time τ_D is:

$$\tau_D = \frac{e\Lambda^2}{4\pi^2\mu_e k_B T} \quad (4.23)$$

The inverse of the sum of photoproduction and ion recombination rates, τ_I is:

Figure 4.2 - Photorefractive response time versus optical intensity for InP:Fe for $0.01 \leq I_0 \leq 1000 \text{ mW/cm}^2$, $\lambda = 1.064 \text{ } \mu\text{m}$, $\Lambda = 10 \text{ } \mu\text{m}$, $E_0 = 1.0 \text{ kV/cm}$, and $T = 289^\circ\text{K}$.

$$\tau_I = \frac{1}{\sigma^0 I_0 + \gamma_R n_0} \quad (4.24)$$

where σ^0 is the photoionization cross-section of the charge carrier under consideration. The zeroth order charge number densities n_{oe} and n_{oh} are the mean value of the periodic space charge carrier distributions for electrons and holes, respectively:

$$n_{oe} = \frac{\sigma_e^0 I_0 [Fe^{+2}]}{\gamma_{Re} [Fe^{+3}]} \quad n_{oh} = \frac{\sigma_h^0 I_0 [Fe^{+3}]}{\gamma_{Rh} [Fe^{+2}]} \quad (4.25)$$

Values calculated from the $\{\tau_i\}$ based expression for photorefractive response time τ_e , equation (4.22), are shown in **Figure 4.2** as a function of I_0 at $\lambda=1.064 \mu m$ in InP:Fe. The crystal is biased at 1 kV/cm, and the material parameters are $[Fe]=3.7 \times 10^{16} \text{ cm}^{-3}$, $[Fe^{+2}]/[Fe]=0.92$. The response time is broken down into two parts: one part that is due to electrons, and another part that is due to holes. Since holes are the majority photocarrier in InP:Fe, the hole response time (labeled "COMBINED - HOLES") is the important temporal parameter. It can be seen that as optical intensity increases, the response time generally decreases - the same general tendency seen in **Section 4.1.2.1** for strictly absorptive reasons. Here, the shape of the curve is different. The curve starts off flat for low intensities where $\tau_{eh} \approx 5$ seconds. The curve has a knee near $I_0 = 1 \text{ mW/cm}^2$, after which τ_{eh} falls by one decade for every factor of ten increase in intensity. In the model, the flat low intensity section comes from the conductivity. Using an experimentally measured function for the intensity dependent conductivity in the expression for the dielectric relaxation time (first section of **Chapter 6**), the conductivity begins to change near 0.1 mW/cm^2 from being dominated by the constant dark conductivity term to being dominated by the linearly increasing photoconduction term. The increasing conductivity decreases the dielectric relaxation time, as can be seen in **Figure 4.2** in the curve labeled "Dielectric relaxation - h". The dielectric relaxation time has the same knee as the photorefractive response time. The only difference between the two curves is that the dielectric relaxation time is almost 10^4 times less than the photorefractive response curve. The curve labeled "Photo & ionize time - h" is the τ_I term in equation (4.27). It falls from 70 seconds to 3×10^{-4} seconds as the intensity increases.

The model for photorefractive time constant is adequate for optical intensities

greater than 1 mW/cm^2 . However, the low optical intensities where the knee occurs is valid only for higher resistivity materials like BaTiO_3 . The photorefractive effect at room temperature in InP:Fe is washed out by thermal electrons for pump power less than 1 mW/cm^2 . Therefore the model depicted in **Figure 4.2** is useful, with the caution that low power work tests the limits of the approximations used in the equation.

Values calculated from the $\{\tau_i\}$ based expression for the photorefractive response, (equation (4.22)), are shown in **Figure 4.3** as a function of grating spacing at $\lambda = 1.064 \text{ } \mu\text{m}$, $I_0 = 5 \text{ mW/cm}^2$ in InP:Fe . The grating spacing is varied from zero to $15 \text{ } \mu\text{m}$. The crystal bias and material parameters are the same as in **Figure 4.2**. Since holes are the majority photocarrier in InP:Fe , the hole response time (labeled "COMBINED - HOLES") is the important temporal parameter. As grating spacing increases, the response time is quasi-constant at 50 msec. The exception is an isolated transition from 100 msec for $0 \text{ } \mu\text{m} < \Lambda < 2 \text{ } \mu\text{m}$. Expanding equation (4.22) shows that in the limit as grating spacing increases to infinity, the photorefractive response time converges to the dielectric relaxation time, which is the observed 50 msec quasi-constant. As the grating spacing goes to zero, the response time converges to τ_1 , which is a function of optical intensity as shown in equation (4.27).

The changes in the diffusion time constant (labeled "DIFFUSION - HOLES") from 10^{-12} sec to 10^{-8} sec and the drift time constant (labeled "DRIFT - HOLES") from 10^{-11} sec to 10^{-7} sec have no effect on the photorefractive time constant under this set of device conditions. The situation changes when the electric field is reduced to $\approx 0.1 \text{ kV/cm}$. In that regime, the photorefractive time constant is no longer a quasi-constant. It falls towards the rising diffusion curve and drift curve as grating spacing increase. The rising diffusion curve and drift curve cause the photorefractive time constant to rise again and converge for large enough value of grating spacing.

The model shown in **Figure 4.3** for photorefractive time constant is adequate for grating spacing less than or about equal to the charge transport length. The charge transport length is calculated as the effective hole range [111]:

Figure 4.3 - Photorefractive response time versus grating spacing for InP:Fe for $0 < \Lambda \leq 15$ μm , $I_0 = 5$ mW/cm^2 , $\lambda = 1.064$ μm , $E_0 = 1.0$ kV/cm , and $T = 289^\circ\text{K}$.

$$r_h = \left(\frac{\mu_h k_B T}{e \gamma_{RH} [F e^{-2}]} \right)^{\frac{1}{2}} \quad (4.26)$$

which is 18 μm for holes and 8 μm for electrons. Therefore the model is useful over the depicted range 0 μm to 15 μm of grating spacing. The only drawback to this model in comparison to the absorption based model is that there is no reference to photorefractive gain. The model just states a time to settle, and that is all. Therefore while the current model offers insight into how the charge transport times and the dielectric response times effect the photorefractive response time, there is no connection to overall gain, the most important photorefractive parameter. The problem will be settled in Section 4.1.4.

4.1.3.2 Photorefractive Response Time: Λ , E_0 Sum

The expression in the previous section for photorefractive response time $\tau_e = \tau_e(\tau_E, \tau_D, \tau_R, \tau_I, \dots)$ can be converted into a more manageable three part series with a diffusion function $f_D(\Lambda)$ and a drift function $f_E(\Lambda)$ multiplied by E_0^2 . For $\Lambda \gg$ Debye screening length, the response time is given by Strait and Glass [112] and Klein [113]:

$$\tau_e = \tau_d \left[1 + f_D(\Lambda) + f_E(\Lambda) E_0^2 \right] \quad (4.27)$$

Separation of Λ and E_0 variables in the equation demonstrates that response time increases with applied bias. This corrects the wrong idea that bias acts to speed up the photorefractive effect. Even though bias does speed up individual charge transport, the total time to stabilize the photorefractive grating increases.

The diffusion term $f_D(\Lambda)$ is:

$$f_D(\Lambda) = \frac{4\pi^2 \mu \tau_R k_B T}{e \Lambda^2} \quad (4.28)$$

The drift term $f_E(\Lambda)$ is:

$$f_E(\Lambda) = \frac{4\pi^2 e (\mu \tau_R)^2}{e \Lambda^2 + 4\pi^2 \mu \tau_R k_B T} \quad (4.29)$$

The Debye screening length l_d for both electrons and holes is:

$$l_{se} = \left[4\pi^2 \epsilon \epsilon_o \frac{k_B T}{e^2 [Fe^{-3}]} \right]^{1/2} \quad l_{sh} = \left[4\pi^2 \epsilon \epsilon_o \frac{k_B T}{e^2 [Fe^{-2}]} \right]^{1/2} \quad (4.30)$$

and using $[Fe]=3.7 \times 10^{16} \text{ cm}^{-3}$ and $[Fe^{+2}]/[Fe]=0.92$, $l_{se}=0.48 \text{ } \mu\text{m}$ and $l_{sh}=0.14 \text{ } \mu\text{m}$. Using either charge carrier, the Debye length is too short to measure directly using infrared light with wavelength greater than one micron.

The variables in the Λ , E_o sum expression for photorefractive time response are:

- τ_e = the transport relaxation time of a photorefractive grating
- $f_D(\Lambda)$ = diffusion component of transport relaxation
- $f_E(\Lambda)$ = drift component of transport relaxation
- E_o = externally applied electric field
- l_s = Debye screening length

The curve for photorefractive response time τ_e as a function of grating spacing for $0 \text{ } \mu\text{m} < \Lambda \leq 15 \text{ } \mu\text{m}$ is shown in **Figure 4.4** at $\lambda=1.064 \text{ } \mu\text{m}$ for InP:Fe. External bias is 1 kV/cm, and the optical pump power is 5 mW/cm^2 . The curves labeled "SUM" are from the drift/diffusion term sum given in equation (4.30). For comparison, the figure also shows data points for the tau-based approximation of **Section 4.1.3.1**. The tau-based curves are identified the "COMBINED" label and with symbols: " Δ " for holes and "o" for electrons. For convenience, the time constant derived from the tau-based expression is referred to as "COMBINED_e" or "COMBINED_h", and the time constant derived from the sum expression is referred to as "SUM_e" or "SUM_h".

All time constants fall as grating spacing increases. The SUM and the COMBINED curves for electrons are very close to each other. Only for $\Lambda < 0.5 \text{ } \mu\text{m}$ is there a significant separation. The worst case is at $\lambda=0.1 \text{ } \mu\text{m}$, where $\text{SUM}_e=3.8 \text{ sec}$ and $\text{COMBINED}_e=1.2 \text{ sec}$. For larger values of Λ , SUM_e and COMBINED_e equal each other. At $\Lambda=15 \text{ } \mu\text{m}$, they equal $1.45 \times 10^{-2} \text{ sec}$. The SUM hole time constants are separated by a factor of two for $2 \text{ } \mu\text{m} < \lambda < 15 \text{ } \mu\text{m}$. At $\Lambda=15 \text{ } \mu\text{m}$, $\text{SUM}_h=9.36 \times 10^{-2} \text{ sec}$ and $\text{COMBINED}_h=4.92 \times 10^{-2} \text{ sec}$. They diverge for smaller values of Λ for a worst case of $\text{SUM}_h=23 \text{ sec}$ and $\text{COMBINED}_h=0.1 \text{ sec}$. Therefore while the SUM and the COMBINED models give similar trends, there is no exact equality between the two models.

Figure 4.4 PR response time versus grating spacing, based on the sum of diffusion and drift terms in InP:Fe for $1 \leq \Lambda \leq 15$ μm , $E_0 = 1$ kV/cm, $I_0 = 5$ mW/cm², and $T = 289^\circ\text{K}$.

The dielectric response times for electrons and holes was found to be 1.35×10^{-5} sec and 1.83×10^{-5} sec, respectively. These times are about a factor of 1000 smaller than the smallest indicated photorefractive response time. Drift and diffusion add a lot to the medium's response time.

The curve for photorefractive response time τ_c as a function of optical illumination for $0.01 \text{ mW/cm}^2 \leq I_0 \leq 1000 \text{ mW/cm}^2$ is shown in Figure 4.5 at $\lambda = 1.064 \text{ } \mu\text{m}$ for InP:Fe. External bias is 1 kV/cm, and the grating spacing is 10 μm . Close agreement is evident between the SUM and the COMBINED curves for both electrons and holes. As intensity increases, the response time falls. For large enough intensity, the time falls proportional to $1/I_0$. Considering holes, at $I_0 = 0.01 \text{ mW/cm}^2$, $\text{SUM}_h = 6.39 \text{ sec}$ and $\text{COMBINED}_h = 5.74 \text{ sec}$. At $I_0 = 1000 \text{ mW/cm}^2$, $\text{SUM}_h = 2.32 \times 10^{-4} \text{ sec}$ and $\text{COMBINED}_h = 1.25 \times 10^{-4} \text{ sec}$. Considering electrons, at $I_0 = 0.01 \text{ mW/cm}^2$, $\text{SUM}_e = 1.62 \times 10^{-2} \text{ sec}$ and $\text{COMBINED}_e = \text{SUM}_e$. At $I_0 = 1000 \text{ mW/cm}^2$, $\text{SUM}_e = 8.1 \times 10^{-4} \text{ sec}$ and $\text{COMBINED}_e = 6.8 \times 10^{-4} \text{ sec}$.

Under different conditions, the curves in Figure 4.5 change. For example, in the limit as E_0 approaches zero bias, both the COMBINED_e and SUM_e curve set and the COMBINED_h and SUM_h curve set converge. As the grating spacing changes, the curves tend to diverge. If $\Lambda = 0.1 \text{ } \mu\text{m}$ instead of 10 μm , $\text{SUM}_{e,h}$ curves are greater than their $\text{COMBINED}_{e,h}$ counterparts by a factor of 100. If $\Lambda = 100 \text{ } \mu\text{m}$, $\text{SUM}_{e,h}$ curves are less than their $\text{COMBINED}_{e,h}$ counterparts by a factor varying between 1.1 and 3.

Figure 4.5 PR response time versus intensity, based on the sum of diffusion and drift terms in InP:Fe for $0.01 \text{ mW/cm}^2 \leq I_0 \leq 1000 \text{ mW/cm}^2$, $E_0 = 1 \text{ kV/cm}$, $\Lambda = 10 \text{ } \mu\text{m}$, and $T = 289^\circ\text{K}$.

The curve for photorefractive response time τ_e as a function of external bias $0.001 \text{ kV/cm} \leq E_0 \leq 100 \text{ kV/cm}$ is shown in **Figure 4.6** at $\lambda = 1.064 \text{ } \mu\text{m}$ for InP:Fe. The optical pump power is 5 mW/cm^2 and the grating spacing is $10 \text{ } \mu\text{m}$. Both the SUM_e curve and the SUM_h curve are flat for small values of external bias, equal to $4.0 \times 10^{-4} \text{ sec}$ and $2.3 \times 10^{-3} \text{ sec}$, respectively. The knee at $E_0 = 0.1 \text{ kV/cm}$ corresponds to the drift process of the photorefractive effect beginning to dominate the diffusion process. After the knee, the response time varies with the square of the external bias, as indicated in equation (4.30). For $E_0 = 1 \text{ kV/cm}$, $\text{SUM}_h = 9.55 \times 10^{-2} \text{ sec}$ and $\text{SUM}_e = 1.55 \times 10^{-2} \text{ sec}$. For $E_0 = 10 \text{ kV/cm}$, $\text{SUM}_h = 9.32 \text{ sec}$ and $\text{SUM}_e = 1.51 \text{ sec}$. The electron saturation velocity in InP:Fe near 15 kV/cm is not modeled in the SUM equation. Therefore SUM_e is not accurate for $E_0 > 15 \text{ kV/cm}$. The SUM model was compared with the COMBINED model, as shown in **Table 4.2**. The models are close for small bias, but diverge for $E_0 > 1 \text{ kV/cm}$.

Table 4.2 - Photorefractive Response Time as a Function of External Bias for $\Lambda = 10 \text{ } \mu\text{m}$

BIAS FIELD	MODEL: SUM		MODEL: COMBINED	
	τ_e	τ_h	τ_e	τ_h
[kV/cm]	[sec]	[sec]	[sec]	[sec]
0.001	4.0×10^{-4}	2.3×10^{-3}	4.24×10^{-4}	2.42×10^{-3}
1	1.55×10^{-2}	9.55×10^{-2}	1.54×10^{-2}	4.99×10^{-2}
10	1.51	9.32	8.14×10^{-1}	1.03×10^{-1}
100	151	931	1.74	0.104

The curves change as the parameters are altered. For example, decreasing Λ from $10 \text{ } \mu\text{m}$ to $1 \text{ } \mu\text{m}$ has the result of causing both SUM_e and SUM_h to rise by 100 while maintaining the same separation. Increasing Λ to $100 \text{ } \mu\text{m}$ causes SUM_e to fall by ten, and SUM_h falls by 30. For another example, increasing optical intensity from 5 mW/cm^2 to 50 mW/cm^2 causes the SUM_e and the SUM_h curves to approximately coincide.

Figure 4.6 PR response time versus external bias, based on the sum of diffusion and drift terms in InP:Fe for $0.001 \text{ kV/cm} \leq E_0 \leq 100 \text{ kV/cm}$, $\Lambda = 10 \text{ } \mu\text{m}$, $I_0 = 5 \text{ mW/cm}^2$, and $T = 289^\circ\text{K}$.

4.1.4 Exact Numerical Solution: ΓL Series

The $SUM_{e,h}$ and $COMBINED_{e,h}$ expressions that are explained in the last few sections are complicated and not always accurate expressions for the photorefractive response time. They do not satisfy the experimentalist's desire for a straightforward relationship between the two major observable photorefractive parameters: the gain and the time constant.

Vachss has proposed a simple piecewise analytic solution for two-wave mixing coupling response time τ_e [114] as a function of the total photorefractive gain. His solution is based on the Solymar/Heaton solution to photorefractive two-wave coupling in the undepleted pump limit [115], which in turn comes from the original basic equations by Kukhtarev [116]. τ_e is the time required for the signal beam intensity to reach $1/e$ of its steady state value from its zero pump beam amplitude. The solution is valid for arbitrary coupling strength, so long as the pump beam intensity is at least twenty times as large as the signal beam intensity throughout the crystal. The photorefractive response time τ_e is expressed as the $\tau_e(\Gamma L, \tau_{PRE})$ where:

$$\begin{aligned}\Gamma &= 2WM \text{ gain} \\ L &= 2WM \text{ interaction length} \approx \text{crystal length} \\ \tau_{PRE} &= \text{photorefractive response time without beam coupling}\end{aligned}$$

τ_{PRE} is the photorefractive response time without beam coupling and is used by Cronin-Golomb [117]. τ_{PRE} is the period evaluated in the drift-diffusion SUM and COMBINED expressions in the previous sections. The expression for $\tau_e = \tau(\Gamma L, \tau_{PRE})$ is in two parts. The first part is for $\Gamma L \leq 2.2$ for photorefractive gain less than 10 dB:

$$\tau_e = \tau_e(\Gamma L, \tau_{PRE}) = \tau_{PRE}(1 + 0.41\Gamma L + 0.074(\Gamma L)^2 + O(\Gamma L)^3) \quad (4.31)$$

The second part is for $\Gamma L > 2.2$ for photorefractive gain greater than 10 dB:

$$\tau_e = \tau_e(\Gamma L, \tau_{PRE}) = \tau_{PRE} \frac{\Gamma L}{2} \left(1 + 1.479 \left(\frac{1}{\Gamma L} \right)^{1/4} + O\left(\frac{1}{\Gamma L} \right) \right) \quad (4.32)$$

τ_e increases almost linearly with ΓL for $\Gamma L > 2.2$, and becomes less and less linear as the ΓL coupling decreases.

The two and three term analytic formulas approximate the Bessel function solution for τ_e

and are accurate to ± 2.5 percent. They are satisfying because of their simple form and because they corroborate the correlation between experimentally observed large 2WM gains and slow response times.

The solution to the Vachss expression is depicted in **Figure 4.7**. The two curves in the Vachss expression nearly meet where $\Gamma L \approx 2.2$. For comparison, the photon limited response time from equation (4.8) is also shown. The photon-limited response time is for a one centimeter long crystal of InP:Fe with pump power 5 mW/cm^2 , absorption coefficient 0.93 cm^{-1} , and grating spacing $10 \text{ }\mu\text{m}$. The photon limited time is normalized by dividing through by an typical photorefractive response time of 12 msec in order to conform to the normalized response time used by Vachss. The photon-limited response time is consistently lower than the Vachss expression. The difference is expected since Vachss considers absorption, drift, diffusion, and the settling of the charges into the photorefractive grating, while the photon limited time considers only the time required to photogenerate enough charge carriers to eventually form the photorefractive grating.

4.1.5 Summary

The time required to set up PRE in InP and other III-V semiconductors is a function of many variables. It depends on the optical intensity, external electric field, temperature, grating spacing, and several material parameters. It is observed that the time varies between $100 \text{ }\mu\text{sec}$ and 1000 msec in InP:Fe.

There are four fundamental speed limits in the process: photoexcitation, transport, trap, and Pockels effect. Photoexcitation time is inversely proportional to optical intensity. Transport time is inversely proportional to mobility and the grating spacing. Trap time is inversely proportional to the number of acceptors. The Pockels effect time is inversely proportional to the fundamental transverse optical lattice vibration frequency. The photoexcitation time and the transport time are by far the largest contributors to the time to set up PRE.

For any given material the larger gains are always accompanied by longer time constants to set up the grating. This was experimentally observed in InP:Fe during high gain

Figure 4.7 Vachss's expression for photorefractive two beam coupling response time τ_c .

measurements. The increase in time constant with gain has been simplified by Vachss into two terms. One term considers material and experimental components in a fundamental zero gain term. The second multiplicative term depends only on the ΓL product.

4.2 Optical Gain in InP:Fe

This section is the end of the theoretical part of the thesis. It ties together the areas that have already been considered into the most important photorefractive parameter, the optical two-beam coupling gain Γ . The section considers photorefractive two beam coupling gain in III-V semiconductors in general, with particular application to InP:Fe. The definition for gain Γ is based on the space periodic space charge field E_{sc} already being present as described by equation (2.22). The field has been accounted for by the action of simultaneous (1) photon absorption, (2) drift, (3) diffusion, and (4) recombination processes described in **Chapter 2** (Kukhtarev equation leading to E_{sc}), **Chapter 3** (optical absorption), and **Section 4.1** (time constant). The $\pi/2$ shifted component of E_{sc} produces the phase hologram that couples the two optical beams together. The quantitative amount of coupling is predicted by the coupled wave equation. Many versions of the coupled wave equation exist. The different versions all predict the same general one way energy transfer result seen in the photorefractive effect. The first version was derived by Kogelnick in 1969. The coupled wave equation is presented below, with some necessary modifications for the case of InP:Fe. It is followed by a section deriving the photorefractive gain in two standard InP:Fe geometries.

4.2.1 Coupled Wave Equations

Optical waves propagating in a medium obey the scalar wave equation:

$$\nabla^2 E(x,z) + k(x,z)^2 E(x,z) = 0 \quad (4.33)$$

where $E(x,z)$ is the complex amplitude of the optical wave with temporal frequency ω . The propagation constant $k(x,z)^2$ is spatially modulated by the photorefractive space charge electric field E_{sc} :

$$k(x,z)^2 = \frac{\omega^2}{c^2} \epsilon(x,z) - iv\mu\sigma(I_{sc}x,z) \quad (4.34)$$

where

$\epsilon(x,z)$ = permittivity of the medium

μ = permeability of the medium
 σ = conductivity of the medium

In the previous chapters it has been shown how two coherent interfering waves in a semi-insulating medium can generate a steady state space periodic electric field in the crystal with amplitude E_{sc} :

$$E_{medium} = E_o + E_{sc} \exp(i(k \cdot x + \psi)) \quad (4.35)$$

where

E_o = a DC externally applied field
 ψ = phase shift between E_{sc} and the optical standing wave.

E_{medium} varies slowly in time compared with the optical fields, although the spatial periods of E_{medium} and the optical fields are comparable at order of magnitude one micron in dimension. The space periodic component of E_{medium} modulates the optical propagation constant $k(x,z)$ in the scalar wave equation. The real, lossless part of $k(x,z)^2$ is proportional to permittivity $\epsilon(x,z)$. $\epsilon(x,z)$ is modulated by E_{sc} through the Pockel's effect in equation (1.1):

$$\begin{aligned} \epsilon(x,z) &= (\epsilon_r + \Delta \epsilon_r(x,z)) \epsilon_o \\ &= (\epsilon_r + r_{eff} E_{sc}(x,z)) \epsilon_o \\ &= (\epsilon_r + r_{eff} E_{sc} \exp(i(k \cdot x + \psi))) \epsilon_o \end{aligned} \quad (4.36)$$

where

ϵ_o = permittivity of free space
 ϵ_r = relative permittivity of the medium
 $\hat{k}$ = photorefractive grating vector

The imaginary term in the expression for $k(x,z)^2$ is proportional to conductivity $\sigma(I_o, x, z)$. $\sigma(I_o, x, z)$ contributes an optical absorption grating in the crystal which diffracts either beam into each other, but it does not contribute significantly to two-beam coupling.

Starting from the scalar wave equation, Kogelnik considered two interfering optical frequency electric fields:

$$E = E_s + E_p = E_s \hat{e}_s \exp(i(\nu t + k_s \cdot x)) + E_p \hat{e}_p \exp(i(\nu t + k_p \cdot x)) \quad (4.37)$$

and expressed the two-beam coupling as [118]:

$$\begin{aligned}\cos(\theta)\frac{\partial E_1}{\partial z} + \alpha^* E_1 &= -i \left(\frac{\pi \Delta n}{\lambda} \right) E_2 \\ \cos(\theta)\frac{\partial E_2}{\partial z} + \alpha^* E_2 &= -i \left(\frac{\pi \Delta n}{\lambda} \right) E_1\end{aligned}\quad (4.38)$$

where

- E_1 = signal beam electric field amplitude
- E_2 = pump beam electric field amplitude
- Δn = amplitude of refractive index variation
- α^* = absorption coefficient for E-fields
- θ = half angle of intersection of the mixing optical beams.

Picoli et. al. expressed the coupled wave equations in terms of optical intensity, which is easier to work with [119]:

$$\begin{aligned}\frac{dI_1(z)}{dz} &= \Gamma \frac{I_1(z)I_2(z)}{I_o(z)} - \alpha I_1(z) \\ \frac{dI_2(z)}{dz} &= -\Gamma \frac{I_1(z)I_2(z)}{I_o(z)} - \alpha I_2(z)\end{aligned}\quad (4.39)$$

where

- $I_1(z)$ = $E_1 \cdot E_1^* / Z$
- = intensity of signal beam
- $I_2(z)$ = $E_2 \cdot E_2^* / Z$
- = intensity of pump beam
- $I_o(z)$ = $(I_1^2 + I_2^2)^{1/2}$
- = total optical intensity
- α = absorption coefficient
- Γ = gain coefficient

The gain coefficient Γ is derived in the next section. In general, the signal intensity I_1 is much smaller than I_o . Therefore the solution of the coupled wave equation for $I_1(z)$ is:

$$I_1(z) = I_1(0)\exp((\Gamma - \alpha)z) \quad (4.40)$$

For the unsaturated case the pump intensity $I_2(z)$ is undepleted throughout the interaction region, and so $I_2(z)$ and $I_o(z)$ are relatively constant at least over short lengths. $I_1(z)$ exponentially increases or decreases depending on the sign of Γ . The photorefractive gain manifests itself in the ratio of the signal intensity propagating through the medium alone to

the signal intensity with the pump beam on. The ratio is $\exp(\Gamma L)$, where L is the length of the two-beam interaction region. Experimentally, L is usually made to equal the length of the crystal. Real gain depends upon whether the average gain Γ through the photorefractive volume is greater than the combined effects of the average absorption loss α and the reflection losses at the input and output facets of the crystal medium.

4.2.2 Photorefractive Gain Derivation

To find a general expression for the photorefractive gain coefficient Γ , consider an extension to Feinberg et. al.'s derivation [120]. The reference is basic because it is the source of Γ used by Fabre et. al. [121] and by Boothroyd et. al. [122]. They begin by considering the photorefraction as a statistical process where W_n is the probability that a charge occupies site n under optical intensity I_n . The rate of change of occupancy probability W_n is proportional to the sum over the ensemble of "m" sites of the product of the site-to-site hopping rate constant D_{mn} with the difference between the value of the diffusion factor $W_n I_n$ multiplied by the drift factor $\exp(qV_{nm}/(2k_B T))$ at sites m and n :

$$\frac{\partial W_n}{\partial t} = - \sum_m D_{mn} \left(W_n I_n \exp\left(\frac{qV_{nm}}{2k_B T}\right) - W_m I_m \exp\left(\frac{qV_{mn}}{2k_B T}\right) \right) \quad (4.41)$$

where V_{nm} is the voltage between sites n and m . The qualitative goodness of this model can be judged by considering two limiting cases under quasi-uniform illumination. First, if $W_n = 1$ and all other $W_m = 0$, then $dW_n/dt < 0$: one charge tends to diffuse or drift away from site n to take up another site m if all m sites are empty. The probability that the charge will occupy original site n falls. Second, if $W_n = 0$ and all other $W_m = 1$, then $dW_n/dt > 0$: charges from the other sites, under illumination, tend to occupy site n . The probability that a charge will move to occupy site n increases. These probability tendencies are exaggerated in the presence of a DC electric field, as expected: electric field accelerates charge motion.

Into the statistical equation two coherent beams of light are introduced, described by optical electric fields E_1 and E_2 from before. They interfere and produce a standing intensity wave $I(x)$, with spatial wave vector $k = k_1 - k_2$:

$$\begin{aligned}
 E_1(x) &= \text{Re } E_1 \hat{e}_1 \exp(ik_1 x) \\
 E_2(x) &= \text{Re } E_2 \hat{e}_2 \exp(ik_2 x) \\
 I_{1,2}(x) &= I_0 + \text{Re } I_{01} \exp(ik \cdot x) = I_0(1 + \text{Re } m \exp(ik \cdot x))
 \end{aligned}
 \tag{4.42}$$

where

$$\begin{aligned}
 I_0 &= \text{average total optical intensity} \\
 &= |E_1|^2 + |E_2|^2 / [n_o(\mu/\epsilon)^{1/2}] \\
 I_{01} &= \text{periodic modulation on total optical intensity} \\
 &= 2E_1 \cdot E_2^* \hat{e}_1 \cdot \hat{e}_2^* / [n_o(\mu/\epsilon)^{1/2}] \\
 m &= \text{optical depth of modulation} \\
 &= 2E_1 \hat{e}_1 \cdot \hat{e}_2^* / E_2
 \end{aligned}$$

and it has been assumed that the magnitude of electric field E_2 is much greater than the magnitude of electric field E_1 . The periodic spatial variations in the optical intensity drive the statistical equation so that in steady state the probability $W(x)$ that a charge is at a site at position x is of the same form as the equation for $I(x)$:

$$W(x) = W_0 + \text{Re } W_{01} \exp(i(k \cdot x + \psi + \pi)) \tag{4.43}$$

where ψ is the phase angle between the optical peaks and the probability minima, and the $e^{i\pi}$ factor is introduced because the probability minima tend to lie at the intensity maxima in the purely diffusive case. Poisson's equation relates the probability of occupancy at any point x , $W(x)$, to the static potential distribution in the crystal $V(x)$:

$$\begin{aligned}
 \frac{\partial^2 V(x)}{\partial x^2} &= -\frac{\text{net charge density}}{e_r e_o} = -\frac{W(x) \rho q}{e_r e_o} \\
 \therefore V(x) &= -\frac{\rho q}{e_r e_o} \left(V_1 + V_2 x + \frac{W_0 x^2}{2} - \frac{W_{01}}{k^2} \exp(i(k \cdot x + \psi + \pi)) \right)
 \end{aligned}
 \tag{4.44}$$

where ρ is the total density of empty plus occupied sites, and the V_1 and V_2 constants of integration are ignored because they do not contribute significantly to the periodic grating.

The periodic static potential $V_{01}(x)$ modifies the linear optical susceptibility $X(x)$ of the medium through the electro-optic tensor R and the grating vector k :

$$X(x) = e_o e_r (R \cdot \hat{k}) e_r, \quad V_{01}(x) = e_r (R \cdot \hat{k}) \frac{W_{01} \rho q}{k} \exp(i(kx + \psi + \pi)) \tag{4.45}$$

In the steady state, the product of susceptibility $X(x)$ and the dominant optical field E_2 modifies the wave vector of the signal E_1 in the scalar wave equation, resulting in photorefractive gain to E_1 . To show this, consider the statistical equation again. In steady state, the time derivative goes to zero. The statistical equation can be normalized and rewritten:

$$\frac{\partial w}{\partial t} = -DI_0 k_0^2 l^2 ((w + m)\alpha(\alpha + i\alpha f) + w) = 0 \quad (4.46)$$

$$\therefore w = -\frac{m\alpha(1 + if)}{1 + \alpha^2 + i\alpha f}$$

where

$$\begin{aligned} w &= \text{normalized charge wave amplitude} \\ &= W_{01}/W_0 \\ \alpha &= \text{normalized grating wave vector} \\ &= k/k_0 \\ f &= \text{normalized electric field amplitude} \\ &= E_{sc}/f_0 \end{aligned}$$

and the normalizing factors W_0 , k_0 , and f_0 are defined as:

$$k_0 = \text{characteristic wave vector} = \sqrt{\frac{\rho W_0 q^2}{e_o e_s k_B T}} \quad (4.47)$$

$$f_0 = \text{characteristic field} = \frac{k_0 k_B T}{q}$$

Substituting $W_{01} = wW_0$ into the expression for linear optical susceptibility $X(x)$ yields:

$$X(x) = -2e_o e_s (R \cdot \hat{k}) e_s \frac{(\alpha + if)}{1 + \alpha^2 + i\alpha f} \frac{E_1}{E_2} f_0 (\hat{e}_1 \cdot \hat{e}_2^*) \exp(i(kx + \psi + \pi)) \quad (4.48)$$

$X(x)$ is directly proportional to the smaller optical field E_1 . The relation can be simplified if $|\alpha| \ll f$, that is, if the amplitude of the space charge field E_{sc} is greater than 0.16 kV/cm for $\Lambda = 10 \mu\text{m}$. Usually E_{sc} is greater than 1 kV/cm and so susceptibility $X(x)$ simplifies to:

$$X(x) = -2ie_o e_s (R \cdot \hat{k}) e_s E_{sc} \frac{E_1}{E_2} f_0 (\hat{e}_1 \cdot \hat{e}_2^*) \exp(i(kx + \psi + \pi)) \quad (4.49)$$

The standing wave of optical polarization density $P(x)$ induced in the crystal

is:

$$\begin{aligned} P(x) &= \text{instantaneous polarization} \\ &= \frac{1}{2} \epsilon_0 X(x) \hat{e}_2 E_2 + \text{higher order nonlinear terms} \end{aligned} \quad (4.50)$$

where the dominant E_2 term multiplies $X(x)$ and is cancelled by the E_2 term in the denominator of $X(x)$. The polarization density $P(x)$ is proportional to smaller optical field amplitude E_1 . $P(x)$ alters the propagation k_1 of optical wave E_1 by altering the effective dielectric constant in the scalar wave equation. Since the value of $\Delta\epsilon$ as seen by E_1 is the projection of its electric field polarization $\hat{e}_1$ on the optical polarization density $P(x)$, the vector propagation constant k_1 and the projection of the altered propagation constant $k_1 \delta k_1$ can be written:

$$\begin{aligned} k_1^2 &= \frac{\omega^2}{c^2} \epsilon - i\omega\mu\sigma \dots \text{but in SI media, } \sigma \sim 0 \\ \dots k_1^2 &= \frac{\omega^2}{c^2} \left(\epsilon_0 \epsilon_r + \frac{1}{2} \hat{e}_1 \cdot X \hat{e}_2 E_2 \right) \\ \text{and } k_1 \cdot \delta k_1 &= \frac{\omega^2}{4c^2 \epsilon_0} \hat{e}_1 \cdot X \hat{e}_2 \frac{E_2}{E_1} \end{aligned} \quad (4.51)$$

by taking the vector derivative, and using the identity $dX/dE_1 \equiv X/E_1$.

The photorefractive gain Γ comes from substituting the expression for the optical susceptibility X into the modified propagation constant $k_1 \delta k_1$:

$$\hat{k}_1 \cdot \delta k_1 = - \frac{i\pi}{\lambda} \frac{r_{eff}}{n_0} E_x (\hat{e}_1 \cdot \hat{e}_2^*) \exp(i(\psi + \pi)) \quad (4.52)$$

where k_1 is normalized by dividing through by $|k_1| = \sqrt{\epsilon_r} \omega/c$, and the expression is simplified by setting the directional components surrounding the electro-optic tensor to r_{eff} :

$$r_{eff}(\theta, \phi) = \hat{e}_1 \cdot \mathbf{e}_r (R \cdot \hat{x}) \mathbf{e}_r \hat{e}_2 \quad (4.53)$$

The resulting electric field $E_1(x)$ and the optical intensity $I_1(x)$ is:

$$\begin{aligned}
 E_1(x) &= \hat{e}_1 E_1(0) \exp((k_1 + \hat{k}_1 \cdot \delta k_1)x) \\
 I_1(x) &= \frac{E_1(0)^2}{n_o \sqrt{\frac{\mu}{\epsilon}}} \exp((2k_1 + 2\hat{k}_1 \cdot \delta k_1)x)
 \end{aligned}
 \tag{4.54}$$

where the optical electric field expression was converted into an optical intensity expression by squaring the electric field and dividing by the impedance of the medium $n_o(\mu/\epsilon)^{1/2}$. The combined presence of the space charge field and the optical pump field E_2 change the optical intensity $I_1(x)$ by a factor $\exp(2\hat{k}_1 \cdot \delta k_1 L)$, where L is the length of the interaction region. The gain Γ is identified as $\Gamma \equiv \text{Re}[2\hat{k}_1 \cdot \delta k_1]$. Therefore the general expression for photorefractive gain $\Gamma(\Theta, \phi, \psi)$ is:

$$\Gamma = \mp \frac{2\pi}{\lambda} \frac{r_{eff}(\Theta, \phi)}{n_o} E_x \sin(\psi) \frac{\hat{e}_1 \cdot \hat{e}_2^*}{\cos(\Theta_{in})}
 \tag{4.55}$$

where gain is negative for holes, and positive for electrons. Gain Γ is proportional to r_{eff} , space charge field amplitude E_x , space charge field phase shift $\sin(\psi)$, and the projection of the signal beam polarization on the pump beam polarization $\hat{e}_1 \cdot \hat{e}_2^*$. The importance of the space charge field phase shift of 90° is clearly demonstrated. With zero phase shift, there is zero gain. The imaginary term $\exp(2\hat{k}_1 \cdot \delta k_1 L)$ imposes a constant phase shift in $E_1(L)$ and no gain, and so it is neglected.

The $1/\cos(\Theta_{in})$ term is optional geometry factor, introduced at the end of the derivation to correct for the slanting of the beam through the crystal. Another way of expressing it is to say that in a crystal of length L , the path travelled by light angled at Θ to the normal is $L/\cos(\Theta_{in})$. Fabre and Picoli have a modulation depth factor m in the denominator of the equation for Γ . This thesis does not find such a term and it does not seem to effect the results. The $1/m$ term can be found in some papers concerning high gain ferroelectrics such as BaTiO_3 , but it is not often associated with III-V semiconductor photorefractive gain.

In InP:Fe , the effective electro-optic coefficient r_{eff} is different for the two

principal crystal orientations: $\mathbf{R}=[001]$ and $\mathbf{R}=[110]$. Using the expressions for r_{eff} in equation (2.44) which were derived in Chapter 2, the photorefractive gain terms become:

$$\begin{aligned}\Gamma_{[001]} &= -\frac{2\pi}{\lambda} r_{41} n_o^3 E_x (\sin^2(\phi) \sin^2(\theta) + \cos^2(\phi)) \sin(\psi) \\ &\quad \times (\sin^2(\phi) \cos(2\theta) + \cos^2(\phi)) \quad \text{for } \hat{k} = [001] \\ \Gamma_{[110]} &= -\frac{\sqrt{2}\pi}{\lambda} r_{41} n_o^3 E_x \sin(2\phi) \cos(\theta) \sin(\psi) \\ &\quad \times (\sin^2(\phi) \cos(2\theta) + \cos^2(\phi)) \quad \text{for } \hat{k} = [110]\end{aligned}\tag{4.56}$$

where using equations (2.42) and (2.43), $\hat{e}_1 \cdot \hat{e}_2 = \sin^2(\phi) \cos(2\theta) + \cos^2(\phi)$ was substituted.

4.2.3 Summary

The two-wave mixing coupled wave equations were presented and a solution was obtained for one optical wave which has much less intensity than the other. As the smaller intensity signal beam travels through the crystal, its intensity increases or decreases depending on the difference between the exponential gain Γ and the characteristic material loss coefficient α . The larger intensity pump beam decreases in intensity as it travels through the same crystal, primarily depending on the exponential material loss coefficient α and depending to second order on the rate of energy transfer to the weaker signal beam.

A general expression for the exponential gain coefficient Γ was derived using Feinberg's statistical method of 1980 [121]. The derivation is based on steady state statistical view of the migrating charges. The dependence of the gain on the nominally 90° phase shift between the standing wave of optical intensity and the space charge electric field was demonstrated. The dependence on dominant charge carrier, beam polarization, and the effective electro-optic coefficient was shown for two principal crystal orientations: $\mathbf{R}=[001]$ and $\mathbf{R}=[110]$.

CHAPTER 5

NUMERICAL MODELING WITH PISCES

This chapter considers numerical modeling of the photorefractive effect. The modeling was performed by applying the finite element method to solve the Poisson differential equation and continuity equation in two spatial dimensions. A narrow bar of InP:Fe was considered as indicated in the next section. The reason for employing the finite element method to study 2WM is that classical techniques involving closed form solutions for photorefractive parameters (gain and response time) are valid only under specific conditions. The classical techniques were already discussed in **Chapter 2** on the Kukhtarev model, **Chapter 3** on optical absorption, and **Chapter 4** on photorefractive parameters. Numerical modeling overcomes simplifying but restricting approximations on device conditions because it is a general technique and allows one to predict gain and response time under arbitrary situations. The optical intensity, modulation depth, and grating spacing are all programmable, giving the user some control in observing the simulated photorefractive effect.

5.1 Description

PISCES is the acronym for **Poisson and Continuity Equation Solver**. It is a commercial program⁸ that simultaneously solves Poisson's equation and the continuity equation for electrons and holes under conditions of photogenerated charge carriers. **PISCES** determines electrostatic potential, and electron and hole concentrations in a user-defined two dimensional structure. To set-up the device, the user defines the device extents, and the finite element mesh as depicted in **Figure 5.1**. The default device size is 20 optical standing waves in the direction of the external bias, and one standing wave in the plane of incidence.

Given the dimensioned device, the next step is to define the physical locations of electrodes, doping levels, dopant position in the band gap, band gap, mobility, recombination time, Fermi-Dirac statistics, external bias voltage, the temperature,

⁸ **PISCES** Version S, Revision 9033 is run on SUN SparcStations. It is supplied by Technology Modeling Associates, Inc., of Palo Alto, CA.

material type, and permittivity. Solutions are obtained for the Poisson equation and the continuity equation at each node of each triangular finite element in **Figure 5.1** using simple boundary conditions. For an adequately fine mesh, the photorefractive effect can be simulated. Exactly how fine the simulation mesh must be is explored later on in this **Chapter**.

Once the device is described, **PISCES** solves for the steady state of the InP:Fe without the presence of light. The dark steady state condition is the starting point for the photorefractive effect. Due to potential instabilities in the numerical method, care must be taken when introducing electric field in the crystal. Therefore before light is applied, bias electric field in the horizontal direction in **Figure 5.1** is ramped up in several solution iterations. This eliminates convergence problems. Newton's method is used to solve the coupled Poisson and continuity equations subject to the default convergence tolerances applied at each node:

►Potential tolerance	10^{-5}	[kT/q]
►Carrier concentration tolerance	10^{-5}	[unitless]
►Poisson equation tolerance	10^{-26}	[Coulomb/micron]
►Continuity equation tolerance	5×10^{-18}	[Ampere/micron]

Figure 5.1 - Finite element mesh for $\Lambda=10 \mu\text{m}$: 19 grid points per Λ in the direction of the electric field, 1 grid point per Λ in the plane of incidence.

The solution is based on a first-order backward difference time discretization. It is used instead of the default second-order time discretization in order to speed convergence in some simulation conditions.

Into the solution for a dark, steady state, biased, bulk crystal, an s-polarized optical standing wave is introduced using the PISCES **PHOTOGEN** feature. The geometry is same as in **Figure 1.1**, where the light beams enter the crystal from the bottom, intersect, interfere, photons are absorbed, and eventually the two beams optically couple before leaving at the top of the crystal. The **PHOTOGEN** feature models the spatial and temporal generation of charge carriers in a device. The photogeneration model is discussed in more detail in the next section. The optical intensity waveform is sinusoidal and is defined at the center of each finite element in the mesh. In general 19 points per standing wave are used to define the sinusoidal exciting waveform. The number of points per standing wave was varied between 4 and 38 points. 19 points was found to be adequate for convergence. See **Section 5.4.10** for more details.

Once the device is set-up, the light is introduced at time $t=0$. PISCES evaluates an initial solution at $t=0.1$ psec. The initial solution is used as the basis for another solution, stepped in time by a factor of 1.414. Stepping by $t_2/t_1=1.414$ continues until $t=1.00$ psec is reached. After evaluating intermediate solutions at 0.1, 0.141, 0.200, 0.282, 0.400, 0.566, 0.800, and 0.900 psec, PISCES solves for the device state at $t=1.00$ psec. The device state consists of local potential, ionization states of acceptors and donors, and hole and electron concentrations for each node of each finite element. The 1.00 psec solution is saved. Next, the solution is evaluated for $t=10$ psec using the previous intermediate times scaled by a factor of ten. The 10 psec solution is saved. The sequence continues in decades of time until $t=1.0$ second. At one second the device is observed to be in a quasi-steady state for most simulation situations applicable to III-V semiconductors.

Using the thirteen decades of time solution files ranging from one picosecond to one second, PISCES plots the space charge electric field E_{sc} , electron concentration, hole concentration, net charge concentration, and net carrier concentration across the optical intensity standing wave grating. The resulting curves show the growth dynamics of a photorefractive grating.

5.2 Modeling Photorefraction

The PISCES program for modeling the photorefractive effect is structured into two main parts. The first part develops the solutions and displays intermediate results every decade in time. The intermediate results are at times 1 psec, 10 psec, 100 psec, ... one second. The device is laid out, the sinusoidal optical intensity pattern is defined by sampling a sinusoid at every $1/19^{\text{th}}$ of the sinusoidal grating spacing, and eventually solutions are saved. The second part of the program uses the thirteen solutions to present all thirteen decades of data versus position at once for electric field, charge carrier concentration, net carrier concentration, net charge concentration, and mid gap potential. Representative results of the program are given in Section 5.3 for the zero bias and the 30 kV/cm cases.

5.2.1 PHOTOGEN Model

The PHOTOGEN model is used to simulate the generation of free electrons and holes resulting from optical absorption at deep level iron sites. The process is called "photogeneration". Photogeneration works by specifying the coordinates of the starting and ending points of the optical path in the device. The photocarrier generation rate $G(z, r, t)$ is defined in three dimensions: (1) along the optical axis (z), (2) radially from the optical path (r), and (3) in time (t):

$$G(z, r, t) = Z(z)R(r)T(t) \quad (5.1)$$

Each z , r , and t variable is independent and the terms due to $Z(z)$, $R(r)$, and $T(t)$ are separable from the others. The optical beam has a Gaussian intensity cross-section in beam profile and in time. The time dependence $T(t)$ is:

$$T(t) = \frac{2\exp\left(-\left(\frac{t-T_o}{TC}\right)^2\right)}{TC\sqrt{\pi}\operatorname{erfc}\left(-\frac{T_o}{TC}\right)} \quad (5.2)$$

where

- T_o = time offset for the generation of the pulse, located at the peak of the pulse.
[seconds]
 TC = characteristic time of the generation pulse

[seconds]

T_0 is set to zero because charge generation starts at $t=0$. TC is set to 10,000 seconds in order to simulate a constant illumination for time between zero and one seconds. Therefore $T(t)$ is approximately constant and equal to 1.1284×10^{-4} [unitless] during this time.

The z dependence $Z(z)$ is:

$$Z(z) = A_1 + A_2 z + A_3 \exp(A_4 z) + k \left(C_1 (C_2 + C_3 z)^{C_4} + L(z) \right) \quad (5.3)$$

where the "A" terms describe the length dependence of photogeneration, and the "k", the "C" and the " $L(z)$ " terms describe the linear energy transfer in single-event upset simulations, useful in X-ray or ion bombardment simulation. The linear energy transfer terms are all set to zero for sub-bandgap photon absorption. The other terms are:

- A_1 = the constant depth dependence of charge generation
[carriers/cm³]
- A_2 = the linear dependence of charge generation
[carriers/cm³/μm]
- A_3 = the pre-exponential depth dependence of charge generation
[carriers/cm³]
- A_4 = the exponential absorption coefficient
[μm⁻¹]

Terms A_1 and A_2 are set to zero. Only terms A_3 and A_4 pertain to sub-band gap absorption. Term A_3 is dependent on optical intensity, absorption cross-section of the iron deep level, and the iron concentration. A_3 is the most important term for the photogeneration model, and is calculated using:

$$A_3 = k_1 \frac{I_0}{h\nu} \left(1 + m \cos\left(\frac{2\pi x}{\Lambda}\right) \right) \sigma_h [Fe^{-3}] \quad (5.4)$$

where

k_1 = a special term

and all of the other terms were defined before. The constant k_1 is set equal to 8862, the reciprocal of $T(t)$ between zero and one seconds, in order to account for an undocumented feature in the PISCES program that was uncovered in the course of this thesis. Without the k_1 term, the photogeneration rate reported by PISCES is less by a factor of about 10^4 than

the rate calculated by hand.

Term A_4 is the absorption term, and it is less significant than A_3 because the sub band gap optical absorption is negligible over the optical axis device length of a few microns. A_4 is dependent on the bulk absorption coefficient. Only hole generation is considered here because, as discussed in Chapter 3 (Optical Absorption), α_h is about ten times larger than α_e at $\lambda=1.064 \mu\text{m}$. Because holes are known to be the dominant photocarrier in InP:Fe, the hole generating optical absorption coefficient is arbitrarily set at 90 percent of the measured sample absorption $\alpha=0.93 \text{ cm}^{-1}$. This gives $A_4=8.4 \times 10^{-5} \mu\text{m}^{-1}$. The optical absorption is very small over the space of $10 \mu\text{m}$; only 0.09% of the intensity is lost over $10 \mu\text{m}$.

The radial dependence $R(r)$ is a Gaussian profile:

$$R(r) = \exp\left(-\left(\frac{r}{DCHR}\right)^2\right) \quad (5.5)$$

where

DCHR = the characteristic radial distance for charge generation
[microns]

The user program written to implement the photorefraction model defines the optical intensity every grating spacing. The DCHR variable is set at 100 grating spacings to eliminate the variation of the optical intensity across each individual finite element. As a result, $R(r)$ is always unity.

5.2.2 Approximations

The PISCES PHOTOGEN model considers only electron-hole pair production as the result of optical absorption events. This is due to the PISCES program development history. The PHOTOGEN feature was originally designed as a single event upset subroutine. As a result, PISCES does not model the absorption of a sub-band gap photon by a deep level iron site, resulting in one charge carrier and one fixed empty trap. Neither does PISCES model the inter-conversion between Fe^{+2} and Fe^{+3} due to thermal ionization and optical absorption. Deep levels and their ionization states are not modeled by PISCES. The straight PISCES model is unsuited to modeling photorefraction in InP:Fe at $\lambda=1.064$

μm , and so modifications are made.

Altering PISCES by adding appropriate software modules was considered and rejected because the length and complexity of the code indicated a "conceptually simple" task that would last many months. Testing the code would also be a major project with little pay off. Instead, it was decided to use a few approximations.

Although PISCES does not simulate the deep iron level, it allows the user to modify all of the material parameters. By setting the electron mobility to zero, the creation of an electron hole pair creates a mobile hole and a fixed electron. The fixed electron acts just like the Fe^{+2} trap that results in deep level absorption: it is immobile, and traps holes. In addition, experience with the program indicates that any nonzero value of electron mobility causes the photorefractive grating to completely disappear. Setting μ_e to zero is the major approximation in using PISCES to model the photorefractive effect.

Modeling mobile photoelectrons is not possible in the $\mu_e=0$ approximation. However, thermal electrons can be modeled to some extent by invoking the partial ionization of shallow donors feature and carefully choosing the Fe^{+2} band gap position and concentration. However, their mobility is still locked at zero. Zero mobility is expected to increase the response time of the photorefractive effect modeled in the PISCES package.

The absence of deep iron trap levels changes the meaning of the material parameters. Assigning electrons to account for Fe^{+2} traps causes the Shockley-Read-Hall (SRH) recombination time of electrons "TAUN0" to mean a combined recombination time for electrons and Fe^{+2} . For example, values of TAUN0 near 10^{-3} second caused electric fields to be maximized in the crystal, while electron recombination times are typically 0.5 nsec. 10^{-3} sec is a compromise between the real value of τ_e and the mean time to ionize any given Fe^{+3} iron trap site given $[\text{Fe}]=10^{17} \text{ cm}^{-3}$ with $I_0=10 \text{ mW/cm}^2$, about two seconds.

The hole SRH recombination time is fixed at 2×10^{-6} sec for most of the simulation work.

5.3 Results of Single PISCES Simulations

This section presents results from three representative PISCES simulations. The simulations were conducted based on a default set of canonical parameters given in Table 5.1 below. The parameters were selected by taking published numbers, and then perturbing the numbers in many series of PISCES simulations until large periodic space charge fields were produced, with amplitudes on the order of many kilovolts per centimeter. The parameters were further refined until the field amplitudes were large enough to generate experimentally observed photorefractive gain of 7 cm^{-1} .

TABLE 5.1 - PISCES Simulation Canonical Parameters

PARAMETER	VALUE	PARAMETER	VALUE
Λ grating spacing	$10 \mu\text{m}$		
E_o external bias	30 kV/cm	Wavelength λ	$1.064 \mu\text{m}$
I_o optical intensity	10 mW/cm^2	σ_e	$4.7 \times 10^{-18} \text{ cm}^2$
m modulation index	0.1	σ_h	$4.2 \times 10^{-16} \text{ cm}^2$
EDB donor level	0.25 eV	μ_e	$0 \text{ V/cm}^2/\text{s}$
EAB acceptor level	0.29 eV	μ_h	$140 \text{ V/cm}^2/\text{s}$
$[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$	0.99	τ_e	1 msec
[Fe]	10^{17} cm^{-3}	τ_h	2 μsec
Temperature	289 °K	Band gap E_g	1.34 eV
Simulation mesh spacing	$\Lambda/19$	Crystal dimensions	$\Lambda(\text{opt}) \times 20\Lambda(E_o)$
# of grid points	762	# of triangles	760

All of the canonical parameters are within an order of magnitude of observed photorefractive InP:Fe device properties, except for electron mobility which is a special case and has already been discussed. These parameters are individually varied in the thesis and their effects on photorefractive gain Γ are discussed in Section 5.4.

The first simulation uses a special value of SRH hole recombination time $\tau_h = 0.01$ sec in order to clearly demonstrate the net motion of the peaks of the net charge concentration in the crystal from the peaks of the optical standing wave to a steady state value some 90° away.

The second simulation uses modulation index $m=1$ and external bias $E_o = 30$ kV/cm. A modulation index of 1.0 was selected to demonstrate the maximum values of E_{sc} and the driving charge distributions inside the InP:Fe crystal.

The third simulation uses $m=1$ and $E_o = 0$ kV/cm to demonstrate the photorefractive effect under zero external bias conditions. Sinusoidal optical standing wave excitation with a modulation index of unity results in a nonsinusoidal asymmetrical periodic electric field.

5.3.1 First Canonical Simulation: Charge Transport

Figure 5.2 depicts the evolution of the net charge concentration between one picosecond and one second after light is introduced in the crystal. The region depicted is 22 microns in the middle of the photorefractive crystal. The x-axis covers about two grating spacings. The y axis represents the signed logarithm of the net charge concentration. The crystal is biased at 30 kV/cm according to the canonical parameter in **Table 5.1**. The time corresponding to any particular curve in the figure may be determined using the legend in the middle right of the box. "+" represents a data point at $t=1$ msec, " Δ " represents $t=10$ msec, "o" represents $t=100$ msec, and so on. One may observe the dynamics of the photorefractive phase shift being set up between the optical standing wave and the net charge concentration, mainly around $t=1$ msec simulation time. The steady state net charge phase shift is 117° instead of 180° because of the drift component of the spatial phase shift induced by the external bias. The time corresponds roughly to the programmed value of τ_e , which the simulation uses as the mean lifetime for Fe^{+2} charge sites instead of SRH electron recombination time. The simulation used a much larger value for τ_h than the canonical $\tau_h = 2 \times 10^{-6}$ sec. The longer time has the effect of delaying the phase shift from the microsecond regime into the millisecond regime. The delay causes the phase shift/charge transport event to happen in the millisecond time frame when the amplitude of the net charge concentration is visible, instead of in the microsecond regime when the concentration

amplitude is still small.

Figure 5.2 - Growth of phase shift due to net charge transport through drift and diffusion effects. Standing wave optical maxima are at 1 μm , 11 μm , and 21 μm . Optical minima are at 6 μm and 16 μm .

5.3.2 Second Canonical Simulation: External Bias

This section presents the graphic output of a single canonical PISCES simulation with the usual external electric field $E_0=30$ kV/cm, and the modulation index increased to 1. The modulation index was increased to demonstrate a maximum in photorefractive gain. Electric field, current versus time, charge carrier distribution, net charge concentration, and net carrier concentration are depicted in the next pages over 13 decades of time from one picosecond to one second after light is turned on.

5.3.2.1 Electric Field

The increase in the periodic space charge field for a distance of $\pm 1.1\lambda$ around the center of the crystal is depicted in Figure 5.3. E_{sc} grows from zero to 13,250 V/cm, while the dc value of the field falls from 30 to 25.25 kV/cm between 1 psec and 1 second. The corresponding steady state photorefractive gain is 3.7 cm^{-1} . Notice that the steady state peak occurs at position $12.25 \mu\text{m}$, not at position $11 \mu\text{m}$, where the maximum optical excitation occurs. The phase shift is 45° instead of 90° . The carrier concentration peak has moved under the electric field. As a result, the periodic electric field is offset a few degrees from the ideal 90° phase shift by the drift transport mechanism. Later on, the observed phase shift is seen to be 90° for the zero bias case because the drift mechanism is no longer operating (see Section 5.3.2.2).

Figure 5.4 depicts the dynamic electric field over the full 20A of simulated crystal length. In steady state there is a uniform sinusoidal pattern superimposed on a DC level extending most of the way across the crystal. The dc value of the field falls after light is turned on in most of the bulk crystal. The reason is clear from the figure. As the light frees mobile holes, the holes are repelled from the positive electrode on the left hand side of the crystal. The affected region is 10 μm wide with relatively few mobile charge carriers, and relatively high resistance compared with the rest of the crystal. The electric field is large at the left hand end of the crystal by to the relatively high resistance there. There is no similar depletion region at the negative electrode on the right hand side of the crystal. The reason is that electrons are not mobile in this simulation: they are repulsed but not moved, so no depletion region forms.

Figure 5.4 - E_{sc} for $E_0=30$ kV/cm, $m=1$: extents of the crystal.

5.3.2.2 Current

Figure 5.5 depicts the current drawn at the electrodes by the simulated InP:Fe crystal after the light is turned on. The damped exponential growth in current indicates a realistic convergence to a steady state condition. Current increases from 31×10^{-7} A/ μm to 180×10^{-7} A/ μm when 10 mW/cm^2 light is applied. In a one millimeter wide electrode the current would increase from 0.3 mA to 1.8 mA. Assuming that the experimentally observed linear relationship between conductivity and intensity holds in PISCES, the corresponding

Figure 5.5 - Bulk current versus time after light has been turned on for $E_0 = 30$ kV/cm, $m = 1$.

intensity dependent conductivity σ from Figure 5.5 is:

$$\sigma(I) = (10 + 6I)10^{-4}(\Omega\text{-cm})^{-1} \quad (5.6)$$

Illumination increases the conductivity as expected, but the dark and the illuminated conductivity are each 10^4 times larger than experimental measurements in SI InP:Fe. The dark conductivity should be between $\sigma_d = 10^{-7}$ and $10^{-8} (\Omega\text{-cm})^{-1}$. The disparity that is observed here is isolated and has not affected the other results. The disparity may be related to the previously described undocumented feature in the PHOTOGEN model for time dependence. That feature obliged the modeling to use a multiplicative factor of 10^4 in the optical intensity. It may explain the factor of 10^4 in the photoconductivity, but it does not explain the factor of 10^4 in the dark conductivity. The extra intensity did not cause any other problems. In particular, when the response time is observed with the factor of 10^4 included in the optical intensity, the response time is reasonably close to values that are observed experimentally, between 10 msec and 1 second, depending on material and experimental parameters.

5.3.2.3 Hole Distribution

Figure 5.6 shows the hole distribution for $\pm 1.1\text{A}$ about the center of the crystal. It shows the sinusoidal modulation of hole distribution in steady state varying between 5×10^{14} and $2 \times 10^{15} \text{ cm}^{-3}$. The hole distribution changes by a factor of just 20

Figure 5.6 - Hole distribution for $E_0=30$ kV/cm, $m=1$: middle $\pm 1.1\text{\AA}$ of the crystal.

between one picosecond and one second. The $t=0$ value concentration was 10^{14} cm^{-3} . The ordinate scale is kept small to allow easy comparison with the large changes in the electron distribution which are discussed in the next sections.

Figure 5.7 shows the hole distribution over the full 20 $\text{\AA}$ of the simulated crystal. There is a clear repulsion of holes from the left hand positive electrode, showing in the depleted hole concentration in that area. A higher electric field results near the left hand

Figure 5.7 - Hole distribution for $E_0=30$ kV/cm, $m=1$: extents of the crystal.

electrode.

5.3.2.4 Electron Distribution

Figure 5.8 depicts the electron distribution for $\pm 1.1\lambda$ about the center of the crystal. It shows a periodic cycloid-shaped modulation of electron density in steady state varying by a factor of 250 between 8×10^{12} and $2 \times 10^{15} \text{ cm}^{-3}$. The periodic electron density increases by a factor of 10^9 between one picosecond and one second. Up to the one millisecond point, the electron density increases by a factor of ten for every decade of time.

Figure 5.8 - Electron distribution for $E_0 = 30 \text{ kV/cm}$, $m=1$: middle $\pm 1.1\lambda$ of the crystal.

The large increase is because the dark concentration of electrons is negligible in the simulated InP:Fe. Up to the SRH recombination time $\tau_e = 10^{-3} \text{ sec}$, the electron concentration rises. The dark initial electron concentration is around 10^4 cm^{-3} . Combined with the dark hole concentration around 10^{14} cm^{-3} , the intrinsic concentration in the simulated device is 10^9 cm^{-3} . The simulated number is close to the published value of $n_i = 1.2 \times 10^8 \text{ cm}^{-3}$ at 293°K [123]. The rise in electron density stops abruptly when the peak electron density equals the peak hole density at $2 \times 10^{15} \text{ cm}^{-3}$.

Because electrons are constrained from motion in their $\mu_e = 0$ role as the Fe^{+2} trap, the valleys are very deep when the optical modulation index $m=1$. The depth of the valleys is depicted in Figure 5.9 for the extents of the crystal. There is no depletion region

Figure 5.9 - Electron distribution for $E_0=30$ kV/cm, $m=1$: extents of the crystal.

or accentuation in electron concentration near either electrode since the electrons are immobile.

5.3.2.5 Net Charge and Net Carrier Distribution

Figure 5.10 depicts the net charge concentration for $\pm 1.1\lambda$ about the center of the crystal. It shows that between one picosecond and 100 microseconds, the net charge

Figure 5.10 - Net charge distribution for $E_0=30$ kV/cm, $m=1$: middle $\pm 1.1\lambda$ of the crystal.

density including contributions of donors, acceptors, electrons, and holes is a constant at 10^{15} cm^{-3} . At one millisecond, the magnitude of the density begins to decrease near the minima in optical excitation. By ten milliseconds, the curve has completely transformed from a flat line into a periodic boxcar function alternating between positive 10^{14} cm^{-3} and negative 10^{15} cm^{-3} . The alternating regions of net positive and net negative charge generate the photorefractive electric field. The positive regions are due to hole transport from the optical maxima. The hole dominated regions are 115° wide and centered 33° away from the optical minima towards the positive electrode. The pure diffusion case is for 0° away from the optical minima. The 30 kV/cm external bias has shifted the net charge pattern by $\approx 30^\circ$.

Figure 5.11 depicts the net carrier concentration for $\pm 1.1\lambda$ about the center of the crystal. Similar to the net charge concentration, the net carrier density plot shows that between one picosecond and ten microseconds, the net carrier density (electrons plus holes) is a constant at $+10^{14} \text{ cm}^{-3}$. At 100 microseconds, the sign of the density changes

Figure 5.11 - Net carrier distribution for $E_0=30 \text{ kV/cm}$, $m=1$: middle $\pm 1.1\lambda$ of the crystal.

at the optical maxima, and the magnitude increases slightly near the optical minima to form a box car function. By one millisecond, the curve has completely transformed from a flat line into a box car function alternating between positive 10^{14} cm^{-3} and negative 10^{14} cm^{-3} . The alternating regions of net holes and net electrons (as Fe^{+2} hole traps) is the well known result of diffusion and drift transport mechanisms. The steady state hole dominated regions

are 180° wide and centered 33° away from the optical minima towards the positive electrode. The net carrier distribution is shifted by 33° from the optical minima by the external bias. The electron dominated regions grow from zero width to 180° between ten microseconds and ten milliseconds.

5.3.3 Third Canonical Simulation: Zero Bias

The third canonical simulation presents the graphic output of a single canonical PISCES simulation with zero external electric field, and the modulation index increased from the canonical parameter $m=0.1$ to $m=1$ in order to demonstrate the maximum electric field. Electric field, charge carrier distribution, net charge concentration, net carrier concentration, and mid-band potential are depicted in the next sections over thirteen decades of time from one picosecond to one second after light is turned on.

5.3.3.1 Electric Field

Figure 5.12 shows the increase in the periodic space charge field for a distance of $\pm 1.1\lambda$ around the center of the crystal. The steady state waveform is an asymmetrical function, not a sinusoid, even though the driving function is a sinusoid. The asymmetry is most pronounced when the modulation index is near unity. The electric charge tends to

Figure 5.12 - E_{sc} for $E_0=0$ kV/cm, $m=1$: middle $\pm 1.1\lambda$ of the crystal.

leave the bright regions and bunch around regions of darkness. The bunching of charges causes the asymmetrical electric field. This effect can be seen at $x=6 \mu\text{m}$ and $x=16 \mu\text{m}$ in

the figure, where the light intensity falls to zero. The electric field intensity is skewed towards the dark regions. Such a result was theoretically predicted by Gunter in $\text{KNbO}_3:\text{Fe}$ [124].

The asymmetrical electric field arises because the symmetrical optical intensity standing wave has deep minima. Charge tends to collect in the optical minima and stay there. The net charge concentration at the optical minima is more concentrated than at the optical maxima because it is not being excited. The concentration at the optical maxima is always being excited and so excited carriers (holes) diffuse further than at the optical minima. Another way of looking at it is to observe that the dielectric relaxation time varies on a micron to micron basis as a function of local optical intensity (see equation (4.24)). The relaxation time increases in the absence of light. Therefore while carriers diffuse quickly away from the optical maxima, carriers stay and are trapped at the optical minima through the low electrical conductivity at the dark regions. The result is a narrow large concentration of net holes at the optical minima. The tightly confined charge causes the electric field to skew and concentrate around the narrow optical minima regions.

Figure 5.12 depicts E_{sc} growing from zero to 260 V/cm between one picosecond and one second. The corresponding steady state photorefractive gain is 0.07 cm^{-1} . Notice that the steady state peak occurs at position $7.5 \mu\text{m}$, not at position $11 \mu\text{m}$, where the maximum optical excitation occurs. The 126° phase shift is shifted from the ideal 90° by a saturation mechanism. The mechanism results in an asymmetrical nonsinusoidal field, and is seen only for large values of modulation index. For smaller modulation indices, the phase shift is exactly 90° : the electric field's peak is at $8.5 \mu\text{m}$, it is sinusoidal. The ideal 90° photorefractive phase shift and the sinusoidal field can be observed between one and 100 microseconds, before the crystal enters steady state.

Figure 5.13 depicts the dynamic electric field over the full 20λ of simulated crystal length. In steady state there is a uniform periodic pattern extending most of the way across the crystal. On a linear scale the asymmetrical steady state solution assumes a sharply peaked curve. It is that way only for zero bias cases with large modulation depth. When $m=0.1$, the curves are nearly sinusoidal. E_{sc} exceeds 1000 V/cm at the edges of the crystal.

5.3.3.2 Hole Distribution

HOLE DENSITY IN PHOTOREFRACTIVE INP:Fe

log[Concentration (cm⁻¹)]

Distance (microns)

0 MV/cm	EXTERNAL BIAS
10 nm	GRATING PERIOD
10 mW/cm²	OPTICAL INTENSITY
1.77	MODULATION INDEX
1.0E-17 cm⁻³	[Fe]
0.9 ...	[Fe(-Δ)]/[Fe]
9.34E09-16 photons/cm²/sec	PHOTON FLUX
1.7E-17 cm²	Fe(-Δ) optical NS
1.2E-16 cm²	Fe(Δ) optical NS
0.037 cm⁻¹	INP:Fe absorption at 1.0E-16 W

■	1 psec
□	10 psec
●	100 psec
○	1 msec
×	10 msec
+	100 msec
*	1 sec
◇	10 usec
◊	100 usec
△	1 msec
▽	10 msec
◓	100 msec
◔	1 sec

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Figure 5.14 - Hole distribution for $E_0=0$ kV/cm, $m=1$: middle $\pm 1.1\Lambda$ of the crystal.

the hole density does not change between one picosecond and one second. The vertical scale is kept small to allow visual comparison with the large changes in the electron distribution which are depicted in the next section.

Figure 5.15 shows the hole distribution over the full 20 μ m of the simulated crystal. The hole density has a cycloid pattern, even though it is excited with a sinusoidal optical distribution. Unlike the high voltage bias case, there is no 10 μ m wide depletion

Figure 5.15 - Hole distribution for $E_0=0$ kV/cm, $m=1$: extents of the crystal.

region at the left hand positive electrode. Instead, there is a one micron wide region at both electrodes. The small regions are so close to the electrodes that a significant number of mobile holes diffuse into contact with the electrodes and are lost to electron-hole recombination.

5.3.3.3 Electron Distribution

Figures 5.16 and 5.17 depict the electron distribution for ± 1.1 A and ± 10 A about the center of the crystal respectively. They show a periodic cycloid-shaped modulation of electron density in steady state varying by a factor of 250 between 8×10^{12} and 2×10^{15} cm $^{-3}$. The electron distribution is exactly the same as in the external bias case because the electrons are constrained with zero mobility. As a result, bias does not move electrons around. The comments of the HV bias Section 5.3.2.3 apply and are not repeated here.

Figure 5.16 - Electron distribution for $E_0=0$ kV/cm, $m=1$: middle $\pm 1.1\lambda$ of the crystal.

5.3.3.4 Net Charge and Net Carrier Distribution

Figure 5.18 depicts the net charge concentration for $\pm 1.1\lambda$ about the center

Figure 5.17 - Electron distribution for $E_c=0$ kV/cm, $m=1$: extents of the crystal.

of the crystal. It shows that between one picosecond and one microsecond, the net charge density including contributions of donors, acceptors, electrons, and holes is a constant at $-3.9 \times 10^{14} \text{ cm}^{-3}$. At ten microseconds, the magnitude of the density begins to decrease near

the minima in optical excitation, and increase at the maxima. By ten milliseconds, the curve has changed from a flat line into a periodic function with wide valleys at $-5 \times 10^{14} \text{ cm}^{-3}$ and sharp 72° wide peaks at $-3.8 \times 10^{14} \text{ cm}^{-3}$ centered on the optical minima. The alternating regions of charge generate the photorefractive electric field. The more positive darker regions are due to accumulation of holes coming from the optical maxima by diffusion. As observed in the electric field in Section 5.3.3.1, the charge concentrations center on the dark optical minimas at $x=6 \text{ }\mu\text{m}$ and $x=16 \text{ }\mu\text{m}$.

The zero bias charge density is different from the HV bias density in two ways. First, the zero bias peaks coincide with the optical minima (diffusion transport), while the HV peaks are 33° shifted towards the positive electrode. Second, the zero bias peaks are much smaller than the HV bias peaks, where the sign of the net charge alternates. Both differences are due to the drift transport mechanism not being present in this simulation.

Figure 5.19 depicts the net carrier concentration for $\pm 1.1\lambda$ about the center of the crystal. Similar to the net charge concentration, the net carrier density plot shows that between one picosecond and one microsecond, the net carrier density (electrons plus holes) is a constant at $+10^{14} \text{ cm}^{-3}$. At ten microseconds, the magnitude increases slightly at the optical minima to form a smooth periodic function. By ten milliseconds, the curve has changed from a flat line into a function alternating between $+4.5 \times 10^{12} \text{ cm}^{-3}$ and $+1.2 \times 10^{14}$

Figure 5.19 - Net carrier distribution for $E_0=0$ kV/cm, $m=1$: middle $\pm 1.1\text{\AA}$ of the crystal. cm^{-3} . The regions are all dominated by holes.

The zero bias net carrier density is different from the HV bias density in two ways. First, the zero bias holes dominate, while with bias, hole regions alternate with equal width electron regions. Second, the zero bias carrier density is a smooth continuous curve with fluctuations less than a factor of 100, while with bias, the curve is a flat-top boxcar function with sharp periodic transitions that go ten orders of magnitude. As with charge density, these differences are due to drift transport.

5.3.3.5 Mid Band Potential

Figure 5.20 depicts the mid band potential for $\pm 1.1\text{\AA}$ about the center of the modeled crystal. The mid band potential is modulated with the net charge distribution. It has little or no effect on the photorefractive effect, and is included here for completeness. Trapping and excitation involved in the formation of electric field and photorefractive effect are all local phenomena, so the overall potential is not important. Further PISCES output shows that the relative positions of the deep levels and the valence and conduction bands are quantitatively unchanged.

Figure 5.20 - Mid band potential for $E_0=0$ kV/cm, $m=1$: middle $\pm 1.1\text{\AA}$ of the crystal.

5.4 Results of Series of PISCES Simulations

Series of PISCES simulations were conducted based on a default set of canonical parameters given in **Table 5.1** in **Section 5.3**. Space charge electric field amplitude E_{sc} , photorefractive gain Γ , and response time τ were extracted, and their variations were considered as a function of the following variables:

▶ optical intensity I_0	▶ external bias E_0
▶ grating spacing Λ	▶ modulation index m
▶ temperature T	▶ $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ ratio
▶ donor level EDB	▶ $[\text{Fe}]$
▶ acceptor level EAB	▶ mobility μ_e, μ_h
▶ recombination times τ_e, τ_h	▶ simulation mesh spacing

These sets of simulations demonstrate the generation of a space charge field in quadrature with the optical standing wave, the dark carrier equivalent irradiance, Debye length, sufficient E_{sc} to produce 6 cm^{-1} of photorefractive gain Γ , and a limited T-I resonance. To convert the periodic $\pi/2$ shifted space charge electric field amplitude to gain, equation (1.2) is used, assuming $\xi=1$. The equation indicates $E_{sc}=1$ kV/cm produces a photorefractive gain $\Gamma=0.282 \text{ cm}^{-1}$. It is assumed that the gain is positive: PISCES does not account for crystal orientation. The observed gain could be positive or negative, depending

on the orientation of the crystal relative to the beam polarizations. Other values of Γ are linearly scaled from the correspondence.

An in-depth investigation of any one of the simulation sets could be performed by detailed comparisons with analytic photorefractive models. It was decided instead that it would be better to perform a survey of the simulation model's behavior under all possible device conditions. The survey is presented in the next pages.

Figure 5.21 Photorefractive gain vs optical pump intensity.

5.4.1 Gain Γ vs Optical Intensity

Figure 5.21 depicts photorefractive gain Γ vs intensity I_0 . The curve consists of a number of connected data points. Each data point is the result of a twenty minute PISCES simulation run. The curve shows more or less what is experimentally observed: gain Γ increases from zero until I_0 grows comparable to the equivalent dark irradiance in InP:Fe. The equivalent dark irradiance is the intensity of light that it would take to excite the same rate of electrons into the conduction band as does the background thermal fluctuations in the crystal. Typically, the equivalent dark irradiance in InP:Fe is between 10 and 100 mW/cm². This is borne out in the figure since an intensity of ≈ 30 mW/cm² saturates the gain. However, the model does not show the temperature-intensity resonance, which in InP:Fe is a sharp peak at the dark irradiance intensity. Instead, Γ drops only a little from the peak, and then flattens out. This simulated behaviour is close to what is seen experimentally in GaAs (no T-I resonance) and InP:Fe when the InP:Fe is not temperature stabilized. However, when InP:Fe is temperature stabilized, the T-I resonance comes out and the PISCES model fails to show the experimentally observed resonance behaviour.

What is not considered?

- ▶ Thermal run-away due to optical absorption and conductive heating is not included in the model. For example, 1000 mW/cm² at $\lambda = 1.064 \mu\text{m}$ induces a temperature rise of 0.5 °C/sec. 10 mW/cm² and 10 kV/cm bias induces a temperature rise of 0.9 °C/sec.
- ▶ I_0 changes the $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ ratio through absorption, which changes the dynamics of the absorption. It is not considered.

Gain Γ/E_{dc} vs Optical Intensity

Figure 5.22 shows a plot of Γ/E_{dc} vs I_0 shows Γ/E_{dc} increasing from zero until I_0 grows comparable to the equivalent dark irradiance in InP:Fe. However, the model does not show the temperature-intensity resonance, with a sharp peak at the dark irradiance

Figure 5.22 Scaled photorefractive gain Γ/E_{dc} vs pump intensity.

point. Instead, Γ/E_{dc} flattens out, displaying no peak. This is exactly what is seen experimentally in GaAs (no T-I resonance) and InP:Fe when the InP:Fe is not temperature stabilized. When InP:Fe is stabilized, the T-I resonance comes out and the PISCES model fails to accurately represent the experimentally observed photorefractive gain. This is due to PISCES not simulating the competing deep level traps Fe^{+2} and Fe^{+3} , and their different photoionization cross-sections.

5.4.2 Gain Γ vs Temperature

Figure 5.23 is a plot of PISCES data for gain vs temperature. It shows more or less what is experimentally observed: gain increases from zero to a peak. However, the peak is much wider than that observed in practice, and gain does not fall to zero for temperatures greater than the observed optimum temperature. The PISCES model gets some but not all of the features of the T-I resonance.

Figure 5.23 Photorefractive gain vs temperature.

5.4.3 Gain Γ vs External Bias

Figure 5.24 is a plot of PISCES data for gain Γ vs external bias E_0 . It shows more or less what is experimentally observed: gain increases with E_0 squared. What is not seen in the simulation is the fall off in gain near 12 kV/cm due to the fall off in electron mobility. This is a limitation of the PISCES model: electron mobility is artificially fixed at zero to simulate fixed Fe^{+2} sites.

For E_0 around 30 kV/cm, the depletion zone near the positive electrode is about 30 μm thick.

5.4.4 Gain Γ vs Grating Spacing Λ

Photorefractive gain Γ vs grating spacing Λ is modeled for zero bias and for HV DC bias under nonsaturation and saturation conditions.

Figure 5.24 Photorefractive gain vs external bias E_0 .

5.4.4.1 Gain vs Grating Spacing

Figure 5.25 is a plot of PISCES data for gain vs grating spacing for $E_0=0$. The plot shows two curves: one curve for $I_0=10 \text{ mW/cm}^2$ (unsaturated case) and the other curve for $I_0=1000 \text{ mW/cm}^2$ (saturated case). Both cases show a peak in gain, and then a fall off to zero for $\Lambda > 3 \mu\text{m}$. The peak for the saturated case ($0.15 \mu\text{m}$, 9.14 cm^{-1}) is higher and at a shorter grating spacing than for the unsaturated case ($1 \mu\text{m}$, 0.025 cm^{-1}). The difference in peaks is due to the greater rate of photocarrier production in the saturated case driving the charge carriers to partially overcome the negative effects of short range diffusion and charge repulsion. The fall off for larger values of Λ is due to space charge shielding effects and diffusion transport not being able to take the mobile charge carriers the $\Lambda/4$ distance before the carriers are trapped or lost due to recombination.

The plot is similar to published results, where the Debye length peak in the saturated curve is around $0.9 \mu\text{m}$, and in the unsaturated case, the peak (not the Debye length) is around $3 \mu\text{m}$. The difference with the simulation is that here the Debye length is about six times smaller than the one observed experimentally. The difference is due to

Figure 5.25 Photorefractive gain vs grating spacing for zero bias: Debye length measurement shows the difference between saturated and unsaturated conditions.

less effective charge transport in the simulation since there are no mobile electrons, and the mobile holes are much slower (mobility is eleven times smaller). Using equation 4.33 for Debye screening length, the canonical parameters, and $\text{Fe}^{+2} = 5 \times 10^{16} \text{ cm}^{-3}$, the hole Debye length $l_{\text{sh}} = 0.12 \mu\text{m}$. PISCES numerically predicted a hole Debye length $l_{\text{sh}} = 0.15 \mu\text{m}$, within twenty percent of the value from the equation. The unlooked for closeness of the PISCES approximation to analytical theory for Debye length with holes invites a different simulation, where hole mobility is locked to zero and electron mobility is $1500 \text{ cm}^2/\text{V}\cdot\text{s}$.

For larger values of grating spacing, even though there was no applied voltage on the crystal, the simulation indicates $10^{-6} \text{ A}/\mu\text{m}$ current⁹ coming out of the ends of the crystal starting at 10^{-11} seconds and ending at 10^{-4} seconds after light is turned on. This short-term transient current is due to hole-hole charge repulsion near the electrode. The larger grating spacings have larger contiguous ionized regions, in which hole-hole repulsion occurs. Large enough regions produce the small unexpected current that shows up in the

⁹ $\text{A}/\mu\text{m}$ current is converted into absolute amperes by multiplying by the depth of the photorefractive crystal.

simulation.

Figure 5.26 Photorefractive gain vs grating spacing Λ for external bias.

5.4.4.2 External Bias

Figure 5.26 depicts gain Γ vs grating spacing Λ under external bias. It shows gain increasing from zero as Λ increases, leveling off at $\approx 0.8 \text{ cm}^{-1}$ for $\Lambda \geq 100 \mu\text{m}$. The increase conforms to theory. Experimentally, this thesis has observed the increase in gain with grating spacing for $1.3 \leq \Lambda \leq 15 \mu\text{m}$. Therefore experiment and the PISCES model agree over the experimental domain.

5.4.5 Gain Γ vs Time

Figure 5.27 depicts five curves for gain vs time. The curves show gain increasing from zero in the same way for several values of electron recombination time. The curves diverge after $100 \mu\text{sec}$, to either level off at some gain, or else peak near 1 msec and fall off. In all the data, hole recombination time was fixed at $2 \mu\text{sec}$. The coarse data points are due to the design of the program used to generate the data. Data is saved every factor of 10 in time: at $1 \mu\text{s}$, $10 \mu\text{s}$, $100 \mu\text{s}$,... etc.

The growth of gain with time is approximately a smooth exponential only for

Figure 5.27 Photorefractive gain vs time for various electron recombination times.

some conditions: i.e., for electron recombination time $TAUN0=0.001$ and $TAUN0=0.0001$. This indicates that the common assumption of the exponential model for gain is not generally valid, as pointed out earlier in the thesis. For $TAUN0 > 0.001$, gain overshoots its steady state value and comes to rest after a damped oscillation.

Both the simulated smooth settling and the underdamped oscillation are observed in experiments conducted for this thesis and more completely in published results by other workers. Judging from available published work, it is the first time this behaviour has been observed with a finite element type analysis. Therefore experiment and the basic PISCES model without deep level traps agree at least qualitatively over the experimental domain.

5.4.6 Gain Γ vs Modulation Index

This section explores the effect of the modulation index of the optical standing wave on the photorefractive gain in the PISCES model. Zero bias and HV bias cases are considered.

Figure 5.28 Photorefractive gain vs intensity modulation index: $E_o=0$.

5.4.6.1 Gain Γ vs Modulation Index: $E_o=0$

Figure 5.28 depicts gain vs modulation index m for zero bias. It shows gain increasing 1:1 with m for $0.001 \leq m \leq 1$. It is intuitive to think that the photorefractive field increases proportionally with the depth of modulation, since the m drives the amplitude of the photogenerated charge clouds and the resulting diffusion gradient. As a result, charge separation, E_{sc} , and 2WM gain is proportional to the pump-to-signal ratio β .

The graph shows five curves, investigating response between $\Lambda=0.15 \mu\text{m}$ and $10 \mu\text{m}$. For $\Lambda=0.15 \mu\text{m}$, two variations from the canonical parameters are employed: I_o is increased to 1000 mW/cm^2 to saturate the crystal, and $TAUP0$ is increased to 10^{-3} seconds. The changes were suggested by other series of PISCES runs. They indicated that changing these variables would increase gain. The result is maximum photorefractive gain $\Gamma=3.4 \text{ cm}^{-1}$, greater than the intrinsic absorption $\alpha=0.9 \text{ cm}^{-1}$.

The positive sloped curves do not explain experimentally observed high gain with undepleted pumps and small values of signal. The disparity is explained by the PISCES

model not simulating the deep level iron trap. The iron trap increases the field by trapping both mobile holes and electrons. A small signal can set up a large enough field for photorefractive gain with the iron trap. Therefore, accurate photorefractive device simulations with PISCES will be delayed until deep level traps are adequately modeled.

Figure 5.29 Photorefractive gain vs intensity modulation index: $E_0=30$ kV/cm applied bias.

5.4.6.2 External Bias

Figure 5.29 depicts gain vs modulation index m for 30 kV/cm bias. It shows the same trend as in the zero bias case: gain increases 1:1 with m for $0.001 \leq m \leq 1$. The difference is that the maximum fields are about three times larger in the HV case. The reason for this is the more efficient drift plus diffusion charge transport with the external bias.

The graph shows two curves. The lower curve is for the canonical case, with $I_0=10$ mW/cm² and the hole recombination time $TAUP_0=2 \times 10^{-6}$ seconds, resulting in a maximum photorefractive gain $\Gamma=3.7$ cm⁻¹. The upper curve changes I_0 to 30 mW/cm² and $TAUP_0$ to 10^{-3} seconds, since work with PISCES suggested that these changes would increase gain by factors of 1.18 and 1.9 respectively, or by a factor of 2.27 in total. The resulting maximum photorefractive gain is 8.7 cm⁻¹, an increase of 2.3. This is close to what the independent changes suggest. This exceeds the absorption loss $\alpha=0.9$ cm⁻¹ and the observed maximum gain in the experimental part of this thesis, $\Gamma=7$ cm⁻¹.

Even larger values of gain are observed by taking larger values of grating spacing. For example, with $\Lambda=100\text{ }\mu\text{m}$ instead of $10\text{ }\mu\text{m}$ as above, $\Gamma=15.2\text{ cm}^{-1}$. This exceeds known published experimentally observed gains in InP:Fe at $\lambda=1.06\text{ }\mu\text{m}$.

5.4.7 Gain Γ vs Mobility

This Section investigates how charge carrier mobility affects gain in PISCES. Both electron and hole mobility are considered, although in general electron mobility has to be kept at zero in order to model fixed deep level Fe^{+2} for the holes. Otherwise, the photorefractive gain goes to almost zero.

5.4.7.1 Gain Γ vs Hole Mobility

A plot of gain vs hole mobility μ_h shows a flat line. μ_h was varied between 0.00001 and 10000 V/cm²/sec and Γ was constant. Hole mobility plays a small role in the steady state value of gain.

The PISCES field dependent mobility model was employed in an attempt to observe the effect of the high field electrode regions and the periodic field on the photorefractive gain Γ . However, the model caused gain to go to zero while the DC field in the crystal remained constant. This is a PISCES model problem, and has to do with the unmodified commercial software package. Since hole mobility is constant for more than ten times the electric field that electron mobility is, the μ_h is set as a constant at 140 cm²/V-sec. The constant μ_h is reasonable because most published work use it for InP:Fe.

5.4.7.2 Gain vs Electron Mobility

Figure 5.30 depicts gain vs electron mobility μ_e . It shows that gain falls to zero as μ_e increases from 10^{-7} to $10^{-3}\text{ cm}^2/\text{V-s}$. For all other PISCES simulations μ_e is set to zero. Mobile electrons are sacrificed in order to simulate fixed Fe^{+2} hole traps. This detracts from the simulation in that the electron saturation velocity at $\approx 10\text{ kV/cm}$ is not evident in PISCES plots of gain vs external bias.

5.4.8 Gain Γ vs Deep Level Parameters

In the PISCES model, the deep level of iron affects photorefractive gain Γ in three main ways: iron concentration $[\text{Fe}]$, the donor to acceptor ratio $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$, and

Figure 5.30 Photorefractive gain versus the constant electron mobility, "MUN0".

the position of the deep levels in the band.

5.4.8.1 Gain Γ vs Iron Concentration

Figure 5.31 depicts gain vs [Fe]. It shows gain increasing from zero for $[\text{Fe}] > 10^{15} \text{ cm}^{-3}$. At the lower values of iron concentration ($[\text{Fe}] < 2 \times 10^{16}$), there is no DC field in the crystal. Instead, the field is a ramp plus a small amplitude sinusoid going across the crystal. There are no relative peaks and valleys in the electric field in this regime. For larger values of [Fe], photorefractive space charge field amplitude exceeds the DC field. Eventually, for the largest values of [Fe], gain saturates because the iron produces proportionally more free thermal electrons and no more optical holes. The electrons wash out the grating.

The plot shows the difference between gain for external bias of 5 kV/cm and 30 kV/cm. The ratio between gains is not constant, but varies by a factor of five above and below each other. For $[\text{Fe}] > 2 \times 10^{16}$ the ratio is more or less constant.

Figure 5.31 Photorefractive gain vs iron concentration [Fe].

5.4.8.2 Gain Γ vs $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$

Figure 5.32 depicts the zero bias plot of gain vs $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$. The curve shows a resonance behaviour near $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 1$. There are two sharp peaks in gain: 0.045 cm^{-1} at $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 0.99$ and 0.038 cm^{-1} at $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 1.18$, both corresponding to the equality of the thermal electron creation rate and the optical hole creation rate. For $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] \leq 1.0$, $E_{dc} > 0$. However, when $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] > 1$, $E_{dc} \equiv 0$. For most PISCES simulations, $[\text{Fe}^{+2}/\text{Fe}^{+3}]$ is set to 0.99 in order to get the maximum value of gain and the drift transport mechanism resulting from non-zero E_{dc} . $[\text{Fe}^{+2}/\text{Fe}^{+3}] = 0.99$ is the canonical value used in all other simulations.

5.4.8.3 Gain Γ vs Deep Level Position

PISCES models partially ionized shallow acceptor and donor levels, but PISCES does not model deep levels. To model the deep iron level, the iron is split into independent acceptor (Fe^{+3}) and donor (Fe^{+2}) species. The acceptor is defined by its population and by its level EAB relative to the valence band. The donor is defined by its population and its level EDB relative to the conduction band. The model does not account

Figure 5.32 Photorefractive gain vs $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$.

for acceptor/donor dependence, resulting from inter-conversion between Fe^{+2} and Fe^{+3} , and their differing photoionization cross-sections. Therefore the model is limited in its application.

5.4.8.3.1 Gain Γ vs EAB ACCEPTOR LEVEL

Figure 5.33 depicts gain Γ vs the Fe^{+3} acceptor level "EAB" for a simulation without donors. There is a peak in gain at $\text{EAB}=0.290$ eV. The peak corresponds to the point where the optimum number of deep level partially ionized acceptors are available to trap the steady state holes, since EAB impacts on the level's recombination time by controlling the number of thermally ionized acceptors. Varying EAB changes the hole thermal excitation rate.

The peak seen in **Figure 5.33** is not due to the T-I resonance point where the excitation rate of free electrons equals the optical excitation rate of holes, because optical excitation rate is independent of acceptor level in the PISCES model, and because thermal excitation of free electrons is independent of Fe^{+3} acceptor level.

Figure 5.33 Photorefractive gain vs acceptor level "EAB" relative to the valence band.

5.4.8.3.2 Gain Γ vs EDB DONOR LEVEL

Figure 5.34 depicts gain Γ vs the Fe^{+2} donor level "EDB" for $E_0 = 5 \text{ kV/cm}$ and $N_0/N_s = [\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 1.18$. It is different from the other canonical simulations since usually $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 0.99$. This is usually the case because it gives larger values of Γ and nonzero E_{ac} . However, there is only a flat Γ characteristic for the usual case. So the more interesting case was investigated, where there is a peak. The peak in gain is at EDB = 0.25 eV, presumably corresponding to the point where the excitation rate of free electrons equals the optical excitation rate of holes. The drawback to the $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 1.18$ condition is that E_{ac} is zero. This is undesirable because it says that there is no bias assisted carrier transport.

5.4.9 Gain Γ vs Recombination Time

PISCES allows the user to define the SRH lifetime of electrons and holes. This Section investigates the effect that recombination time has on photorefractive gain. The rudimentary model used in the simulation has a few problems. In particular, electron recombination time is really the time for Fe^{+2} to convert into Fe^{+3} , not τ_e . Hole

Figure 5.34 Photorefractive gain vs donor level "EDB" relative to the conduction band.

recombination time is a combination of τ_h and the time for Fe^{+3} to convert into Fe^{+2} .

PISCES defines τ_h as a function of TAUP0, NSRHP, and $N_{\text{total}}(x,y)$, and defines τ_e as a function of TAUN0, NSRHN, and $N_{\text{total}}(x,y)$:

$$\tau_e(x,y) = \frac{TAUN0}{1 + \frac{N_{\text{total}}(x,y)}{NSRHN}} \quad (5.7)$$

$$\tau_h(x,y) = \frac{TAUP0}{1 + \frac{N_{\text{total}}(x,y)}{NSRHP}}$$

where:

- TAUP0, TAUN0 = SRH carrier lifetimes
- NSRHP, NSRHN = SRH concentration parameters
- $N_{\text{total}}(x,y)$ = total local impurity concentration

Figure 5.35 Photorefractive gain versus the SRH hole recombination time for holes, TAUP0.

5.4.9.1 Gain Γ vs Hole Recombination Time TAUP0

Figure 5.35 depicts gain Γ vs TAUP0 the hole recombination time. It shows gain increasing from a plateau at 0.37 cm^{-1} for $\text{TAUP0} < 10^{-4} \text{ sec}$ to a peak of 0.75 cm^{-1} at $\text{TAUP0} = 10^{-3} \text{ sec}$. Gain falls off after this. In all this data, electron recombination time was fixed with $\text{TAUP0} = 8 \times 10^{-4} \text{ sec}$. The peak is due to the hole mobility. For larger TAUP0, holes go further due to drift and diffusion. After the optimum point, the holes drift too far and go past the optical intensity valleys to detract from the overall grating field.

5.4.9.2 Gain Γ vs Electron Recombination Time TAUN0

Figure 5.36 depicts gain Γ vs TAUN0 the electron recombination time. It shows gain increasing from zero for $\text{TAUN0} < 10^{-6} \text{ sec}$ to a peak of 0.45 cm^{-1} at $\text{TAUN0} = 1.3 \times 10^{-3} \text{ sec}$. Gain falls off after this. Hole recombination time was fixed at $2 \mu\text{sec}$. The electron/ Fe^{+2} peak is attributed to a balance between the hole traps not lasting long enough to do their job and trap holes, and lasting too long and not trapping holes, resulting in either case in a washed out grating.

Figure 5.36 Photorefractive gain versus the SRH recombination time for electrons, TAUN0.

5.4.10 Gain Γ vs Number of Finite Elements per Δ

Figure 5.37 depicts gain Γ versus the number of finite elements per grating period. Clearly, for less than 15 elements per grating, gain falls off due to insufficient samples. For around 20 elements per grating, the steady state periodic electric field stabilizes and become less dependent on grating spacing. For this reason, 19 elements per grating spacing was used as a canonical parameter. The relatively slow fall in gain for more than 25 elements per grating is due to the crystal being shortened in length as the number of finite elements increases to keep the total number of triangle elements roughly constant. The shortening is needed due to a software limitation on the number of nodes. As the grid gets denser and the crystal shortens, the depletion region at the positive electrode accounts for larger fraction of the total crystal, and a greater fraction of the applied bias is lost in it. In an ideal world - ideal in the sense that there is enough memory to accommodate all of the required nodes - the length of the crystal could be held constant for all finite element spacings, and the plot would be flat for greater than 20 elements per grating.

Figure 5.37 Photorefractive gain vs the number of finite elements per grating spacing Λ .

5.5 Summary

PISCES models the photorefractive effect in InP:Fe adequately, although the photogeneration model does not account for the deep iron level. The solutions are well behaved and demonstrate experimentally observed and theoretically indicated trends. It is possible to control material parameters and tailor device conditions to achieve desired response times and photorefractive gain. Values of gain as high as 15 cm^{-1} were observed. PISCES generates plots of electric field and charge carrier concentration which assist in understanding the dynamics of creating the photorefractive effect for both zero bias and high field situations.

A small temperature-intensity resonance was observed near 30 mW/cm^2 and 300°K . The photorefractive gain peak is similar to but markedly smaller than published results and experimental results given later in this thesis.

PISCES indicates that gain increases with the square of the external bias. This

is what is generally observed experimentally. PISCES fails to show the peak in gain vs E_{on} , which indicates the saturation velocity of electrons at approximately 12 kV/cm, because the mobility of the electrons is set to zero.

The Debye length is observed in the PISCES model at $\Lambda=0.15 \mu\text{m}$. The peak Λ is seven times shorter than the usual peak Λ at approximately $1 \mu\text{m}$ which is usually experimentally observed. The difference is because the only mobile charge carriers are holes, which have about eleven times smaller mobility than electrons. Implementing the deep iron level would restore mobility to free electrons and clear up this simulation problem. In addition, a peak in gain vs Λ was observed for the unsaturated case at $\Lambda=1 \mu\text{m}$. The increase in Λ_{peak} is experimentally observed and theoretically explained later on in this thesis.

When a 30 kV/cm external bias is applied to the crystal, the Debye length peak in gain vs Λ disappears and is replaced by a damped exponential rise in between $\Lambda=1 \mu\text{m}$ and $\Lambda=500 \mu\text{m}$. Gain increases monotonically by a factor of ten over that interval.

PISCES models of photorefractive gain as a function of time indicates the experimentally observed regimes: damped oscillation and damped exponential increase in gain. The nature of the time response depends on material parameters. Measurement of current as a function of time after light is turned on shows the expected damped exponential rise in current to a steady state value.

The PISCES model showed gain increasing linearly with the index of optical modulation under all device conditions for three orders of magnitude. This aspect of the PISCES model is intuitively sound, but it fails to give a satisfactory explanation of high gain with small signal intensities and large pumps.

Investigation of what happens when the electron mobility $\mu_e=0$ restriction is relaxed shows that the photorefractive gain falls to zero for $\mu_e \geq 10^{-3} \text{V/cm}^2/\text{s}$. This is fallout from the PISCES model failing to simulate the deep iron level.

PISCES investigation of gain vs iron concentration shows that providing $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ is 0.99, increasing the iron concentration also increases gain. A note of

caution is warranted at this point. The improvement in gain depends on the device bias conditions. One should not take the simplistic view from the trend that more iron in InP:Fe necessarily makes a better photorefractor. The $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ ratio is intuitively dependent on $[\text{Fe}]$, and is critical to obtaining gain. Small $[\text{Fe}]$ dependent changes in the ratio invariably result in greatly reduced photorefractive performance.

PISCES investigation of SRH hole recombination time showed that gain could be increased by a factor of two by changing τ_h from 2×10^{-6} μsec (a canonical parameter) to 10^{-3} μsec . The peak is due to optimizing the hole lifetime so that the average photogenerated hole travels $\Lambda/2$ into the optical intensity minima and the drift/diffusion transport is most efficient. A similar peak is observed in the SRH electron recombination time, and the peak is due to a different aspect of the same transport mechanism. The electron/ Fe^{+2} peak is attributed to a balance between the hole traps not lasting long enough to trap holes, and lasting too long and not trapping holes, resulting in either case in a washed out grating.

PISCES investigation of gain as a function of acceptor and donor levels indicate that there are optimum dopant levels which maximize gain.

PISCES allowed a search for the optimum value of the $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ ratio. A resonance is observed near $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]=1$, corresponding to the equality of thermal electron creation rate to the optical hole creation rate. $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]=0.99$ results in a maximum in photorefractive gain. This number maximizes the photorefractive electron-hole competition coefficient ξ at 1.

The effect of finite element grating spacing on the steady state value of the photorefractive gain was modeled. Nineteen elements per grating was found to be adequate to model the effect.

PISCES did not indicate an optimum material for photorefractive gain. An adequate deep level trap model must be implemented before PISCES can suggest appropriate materials.

CHAPTER 6

EXPERIMENTAL TWO-WAVE MIXING

This chapter considers experimental results from laboratory work in photorefractive 2WM in InP:Fe. It builds on the theoretical models in Chapter 2, Chapter 3, and Chapter 4, and the numerical modeling in Chapter 5.

6.1 Description

The experiments and models used to observe and explain the results of two-wave mixing in InP:Fe will be described. Two experiments were performed: one in which the grating spacing Λ was held constant at $\Lambda = 1.3 \mu\text{m}$, and the crystal orientation was changed, and a second in which the grating spacing was varied and the orientation was held constant. The experiments measured gain Γ and characteristic response time τ as a function of optical intensity I_o , grating spacing Λ , externally applied electric field E_o , and pump and signal optical polarization angles ϕ_p and ϕ_s .

The same crystal was used for both experiments. It was among three crystals from the same wafer in the same boule that were characterized for photoconductivity. The semi-insulating InP:Fe wafer was a one mm thick $\langle 001 \rangle$, cut from the bottom of an LEC grown boule. It was cut from the bottom (tang) in an attempt to maximize [Fe] and so maximize the photorefractive properties of the crystal.

Dark and illuminated conductivities were measured. The nominal manufacturer specified dark conductivity is $\sigma_d \approx 10^{-8} (\Omega\text{-cm})^{-1}$. Using silver painted electrodes on three samples from the edge of the wafer, the measured dark conductivity was $\sigma_d = 0.62 \times 10^{-8} (\Omega\text{-cm})^{-1}$ at room temperature. The sample had to be in the dark because even dim incandescent room light caused the conductivity to rise. Under illumination at $\lambda = 1.064 \mu\text{m}$, the measured conductivity σ was found to vary as a linear function of optical intensity I_o :

$$\sigma(I_o) = (0.62 + 0.10I_o)10^{-8} (\Omega\text{-cm})^{-1} \quad (6.1)$$

where I_o is in units of mW/cm^2 and varied between 0 and $17 \text{ mW}/\text{cm}^2$, and measurements were made under a field of $5 \text{ kV}/\text{cm}$ DC. Similarly, under illumination at $\lambda = 1.152 \mu\text{m}$, the measured conductivity σ varies as:

$$\sigma(I_o) = (0.62+0.0089I_o)10^{-8} (\Omega\text{-cm})^{-1} \quad (6.2)$$

where I_o was varied between 0 and 120 mW/cm². I_o at $\lambda=1.064 \mu\text{m}$ has ten times the effect on photoconductivity than I_o at $\lambda=1.152 \mu\text{m}$ due to the difference in absorption coefficient. The difference in absorption coefficient was considered in Chapter 3.

The σ results reported here at $\lambda=1.064 \mu\text{m}$ are comparable to other workers' data for $\sigma(I_o)$ at $\lambda=1.064 \mu\text{m}$. Results at $\lambda=1.152 \mu\text{m}$ have not been reported in the available literature. Ozkul reports a variety of results for various samples at $\lambda=1.064 \mu\text{m}$ (#101A and #154Q3) [125]:

$$\sigma(I_o) = (1.03+0.14I_o)10^{-8} (\Omega\text{-cm})^{-1} \quad (6.3)$$

$$\sigma(I_o) = (0.67+0.3I_o)10^{-8} (\Omega\text{-cm})^{-1} \quad (6.4)$$

Gravey reported [126]:

$$\sigma(I_o) = (1.1+0.1I_o)10^{-8} (\Omega\text{-cm})^{-1} \quad (6.5)$$

and Mainguet reported [127]:

$$\sigma(I_o) = (0.83+0.031I_o)10^{-8} (\Omega\text{-cm})^{-1} \quad (6.6)$$

These conductivities are all close to the measurement made in the sample used in photorefractive experiments in the thesis. The comparison gives one confidence that others' 2WM experimental results should be repeatable using the experimental sample.

The dark conductivity quoted for SI InP:Fe by the factory (Crystacomm) was $\sigma_d \approx 10^{-8} (\Omega\text{-cm})^{-1}$. The number is close to the measured value of $\sigma_d = 0.62 \times 10^{-8} (\Omega\text{-cm})^{-1}$.

6.2 Experiment 1: Γ and τ vs I_0

This section discusses the constant grating spacing Λ experiment used to measure gain Γ and τ as a function of I_0 , at two wavelengths: $\lambda = 1.064 \mu\text{m}$ and $\lambda = 1.152 \mu\text{m}$.

6.2.1 Experimental Set-up

The experimental set-up is depicted in **Figure 6.1**. The source consists of two lasers, at $1.064 \mu\text{m}$ or $1.152 \mu\text{m}$. The $1.064 \mu\text{m}$ laser is a Spectra-Physics diode pumped Nd:YAG laser. It is randomly polarized and puts out 100 mW in TEM_{00} . The $1.152 \mu\text{m}$ laser is an old Spectra-Physics SP-124B HeNe laser with three or four lively longitudinal modes each separated by 200 MHz in the 1 GHz gain profile of the medium. It is vertically polarized and puts out 2 mW in TEM_{00} .

One laser is selected using mirror M1. M1 reflects the beam into M2. After M2, the beam passes through polarizing beam splitter PBS, which makes the beam have vertical orientation with 30 dB extinction ratio. PBS is followed by a neutral density wedge, and focusing lens L1. After L1 the beam is split by dielectric beam splitting mirror BS into pump and signal components. The pump is reflected by mirrors M3 and M4 into the chopper. The chopper cuts the pump beam on and off, providing a modulation to the signal beam. Half-wave plate " $\lambda/2$ " rotates the pump polarization to the desired angle ϕ_p . Then the pump enters the InP:Fe crystal. The crystal is AR coated on both sides for $1.152 \mu\text{m}$.

The signal beam is reflected from BS towards mirror M5. Beam stop BSTOP is used to prevent internal reflections from BS from reaching the crystal. M5 reflects the beam into half-wave plate $\lambda/2$ and thence into the crystal with the pump beam. The crystal is oriented with Λ in $\langle 110 \rangle$. The beam intensity ratio varies between 12.5 and 44.7. The linearly polarized beams have small unwanted orthogonally polarized components due to the dielectric beam splitting mirror BS and due to wavelength dependent trade-offs in the $\lambda/2$ plates. The pump extinction ratio is 23/23 dB, and the signal extinction ratio is 35/27 dB at $\lambda = 1.064 \mu\text{m}$ and $1.152 \mu\text{m}$ respectively. The maximum gain was found for signal and pump beams polarized at 45° rotated from s-polarization, i.e., 45° from vertical.

Figure 6.1 First experimental set-up for 2WM in InP:Fe.

After mixing with the pump in the crystal, the signal beam enters the detector assembly. A neutral density wedge adjusts the power so that the detector does not saturate or have too little light to detect. Microscope objective lens L2 at f/20 collects the Gaussian beam and focuses it through long pass filter LPF onto the 0.3 mm diameter surface of the biased photodiode detector. The uncooled detector consists of a reverse biased InGaAs pin diode (Fujitsu FID13532WS) and a high gain op amp with $R_f = 3.3 \text{ M}\Omega$ feedback resistance. The signal is measured on a voltmeter and a digital storage oscilloscope. The oscilloscope is connected to a computer through GPIB. The computer saves digital pictures from the oscilloscope for later processing.

Many of the components are mounted on positioners. All of the mirrors are on $\theta_x \theta_y$ mounts. Mirror M3 is mounted on a y-z positioner to allow for the equalization of the pump and the signal path lengths. The $\lambda/2$ wave plates are mounted on rotary stages to allow precise polarization positioning. The InP:Fe crystal is mounted on a xyz stage with $\theta_x \theta_y$ positioning. This allows the crystal to be positioned precisely in the beams, and tilted so that the small reflected pump from the crystal's front face reflects back up the signal beam's path. This ensures a clean passage of both pump and signal beams through the crystal.

Objective lens L2 and the photodiode PD are mounted on an xy stage to allow the signal beam to be acquired in a minimum of time.

6.2.1.1 Laser Heating

The InP crystal continuously absorbs laser energy during 2WM. Assuming that all of the absorbed energy eventually becomes thermal energy in the crystal, the rate of temperature increase is related to the flux I_o by the absorption constant α , the bulk specific heat capacity C , and the material density ρ :

$$\frac{\partial T}{\partial t} = \frac{\alpha I_o}{C\rho} = \frac{0.93 \text{ cm}^{-1}}{0.35 \frac{\text{J}}{\text{gram} \cdot ^\circ\text{C}} \times 5.32 \frac{\text{gram}}{\text{cm}^3}} I_o = 0.00050 I_o \quad (6.7)$$

where

$$\begin{aligned} C &= 0.35 \text{ J(gram} \cdot ^\circ\text{C)}^{-1} \\ \rho &= 5.32 \text{ gm cm}^{-3} \end{aligned}$$

I_0 = intensity in mW/cm^2

Therefore one predicts the following rates of temperature rise in the experimental crystal of InP:Fe as given in Table 6.1.

Table 6.1 - Laser Induced Temperature Rise

I_0 [mW/cm^2]	LASER HEATING $\partial T/\partial t$ [$^{\circ}\text{C/sec}$]
1	0.0005
3	0.0015
10	0.005
30	0.015
100	0.05
300	0.15
1000	0.5

For $I_0 \approx 1 \text{ mW/cm}^2$ the rate of temperature rise is $0.0005^{\circ}\text{C/sec}$. It takes 30 minutes to change the crystal temperature by 1°C . Such heating is negligible, because the heat is constantly being exchanged with the environment through conductive, convective, and radiative processes. When the laser is turned on, the crystal is in quasi-steady state with its thermal surroundings. It does not affect the thermal rate or the band gap energy.

The situation is different when the optical intensity is increased. When I_0 is 1000 mW/cm^2 , temperature changes 1°C in 2 seconds. This is too fast for the crystal to be in quasi-steady state with its environment under CW illumination unless the crystal is cooled. Even with cooling, local laser heating introduces thermal spots in the pump beam's path in the crystal. It increases the thermal electron excitation rate and narrows the band gap. Therefore for $I_0 > 30 \text{ mW/cm}^2$, a cooling apparatus is needed. In the absence of a cooler, one must take data rather quickly after turning on the laser pump to obtain uniform cool down for a few minutes. The heat is dissipated through thermal diffusion and by thermal conductivity ($K=0.46 \text{ W (cm}^2\text{-sec)}$) according to the heat conduction equation:

$$\frac{\partial T}{\partial x} = \frac{K}{C} \nabla^2 T + 0.0005 I_0 = D \nabla^2 T + 0.0005 I_0 \quad (6.8)$$

where D is the thermal diffusivity, $0.247 \text{ cm}^2\text{C/sec}^2$. The exact temperature distribution depends on the boundary conditions of the crystal. On the bottom of the crystal there is either no heat exchange ($\partial T/\partial x=0$ in Section 6.1), or else a heat sink ($T=\text{constant}$ in Section 6.2). On the top of the crystal, there is an air-semiconductor interface. The top is not well characterized because the day-by-day flow of air was variable. Blowing air over the crystal was found to speed the process of cooling the laser-warmed crystal after several minutes of illumination. On the sides of the crystal there are touch-on stainless steel electrodes. It is assumed that the electrodes do not significantly contribute to the temperature distribution.

Without illumination, solving the heat conduction equation by separation of variables under steady state yields two results. First, with the crystal mounted on an mylar insulator, $\partial T/\partial x=0$, and the solution is $T=T_{\text{air}}$. Second, with the heat sink, assuming that the top of the crystal is at air temperature and the bottom of the crystal is at heat sink temperature, the equation yields a straight line, where the temperature at the center of the crystal is the average of the air and the heat sink temperatures.

With illumination, the laser heating source term upsets the thermal equilibrium. However, under low illumination conditions this term was considered unimportant. Experimental measurements were taken immediately after turning on the pump, and so heating was not a concern.

6.2.1.2 Power Measurement

Absolute measurement of power was performed using a Photodyne 22XL power meter (serial #10303) and sensor head 550 (serial #12539). The five millimeter diameter active area on the sensor head allowed accurate measurement of distributed beam power. To ensure that the power did not saturate the detector, a calibrated set of precision metallic ND filters was employed to attenuate the beam into the detector's linear region.

Figure 6.2 Fringe visibility of the HeNe laser at $\lambda = 1.152 \mu\text{m}$.

6.2.1.3 Coherence Length

The coherence length of the lasers was the critical parameter to performing 2WM. The coherence requirement was that the optical standing wave interference pattern between the pump and the signal must be stationary for enough time for carrier excitation and drift to set up the periodic electric field. The time stability requirement is equivalent to a bandwidth constraint on the laser, and bandwidth translates into a certain length of the laser beam in which the beam is coherent with itself. The length is called the "coherence length", and it was measured as part of characterizing the 2WM experiments.

The coherence lengths of the two lasers were determined using a different apparatus than shown in **Figure 6.1**. The coherence length at $\lambda = 1.152 \mu\text{m}$ was measured using two arms with a piezoelectrically driven $\pm 14\pi$ single mode fiber optic phase shifter in one arm providing fringes at 420 Hz, and a corner reflector in the other arm providing a variable difference in path length. Fringe visibility $V(z)$ is a function of corner reflector position z and is equal to:

$$V(z) = \frac{I_{\max} - I_{\min}}{I_{\max} + I_{\min}} \quad (6.9)$$

where $I_{\max}$ and $I_{\min}$ are the detected intensities of the peaks and the valleys in the fringes. The result is shown in **Figure 6.2**. The squares are measured points, and the curve

Figure 6.3 Fringe visibility of the Nd:YAG laser at $\lambda=1.064 \mu\text{m}$.

represents the expected fringe visibility given by equation (6.10). There are peaks every 27.5 inches, corresponding to the 27.6 inch separation of the HeNe mirrors. Every peak is about three inches wide at 90% of V_{max} . This corresponds to a coherence length of six inches/fifteen centimeters since the measurement was performed with folded optics. The width and shape of the peak corresponds to the spectral bandwidth and the spectral shape of the laser. However, there is so much 60 Hz AC power supply switching noise in the laser that the spectral shape information is lost. One may still estimate the spectral bandwidth by assuming that the shape of the measured visibility curve is close to a $\text{sinc}(z/L)$ function, where $L \approx 15 \text{ cm}$:

$$V(z) = \left| 0.8 \times \text{sinc} \left(\frac{0.133z}{\text{coherence length}} \right) \right| \quad (6.10)$$

Then the Fourier transform of the visibility curve is the spectrum of the laser, in which the $\text{sinc}(z/L)$ approximation is a box car function of width $c/L \approx 2.0 \text{ GHz}$. The width is near to the known $\approx 1.5 \text{ GHz}$ gain profile of the HeNe medium.

Figure 6.3 shows the same sort of curve as shown in **Figure 6.2** except the laser has been changed. The Nd:YAG laser at $\lambda=1.064 \mu\text{m}$ is being employed. The data points were obtained using a different apparatus, and the lines represent the fringe visibility calculation for the Nd:YAG. The apparatus used a nonpolarizing beam splitter dividing the

Figure 6.4 2WM gain using the Nd:YAG laser at $\lambda = 1.064 \mu\text{m}$.

vertically polarized beam into two orthogonal arms of approximately equal length. One arm was $\pm 3\pi$ phase modulated with a piezo driven retroreflecting mirror, and the other arm was a plain retroreflecting mirror mounted on a positioner. The simple interferometer geometry combined the collimated reflected beams in the nonpolarizing beam splitter and detected them with the detector apparatus of **Figure 6.1**. In order to observe maximum fringe visibility, ND wedges were placed in each arm to equalize the optical intensities.

Figure 6.3 shows that the 90% visibility coherence length is ≈ 2 mm. The line represents the $V(z) = \text{sinc}(z/L)$ model:

$$V(z) = \left| \text{sinc}\left(\frac{0.465 z}{\text{coherence length}}\right) \right| \quad (6.11)$$

where coherence length $L \approx 4.3$ mm. This corresponds to $c/L \approx 70$ GHz. As expected there is a relationship between the coherence length L and the ≈ 60 GHz gain bandwidth of the Nd:YAG medium.

Figure 6.4 shows the effect of the coherence length on the 2WM gain Γ at $\lambda = 1.064 \mu\text{m}$. The graph comes from the set-up procedure in experiment #2 in **Section 6.2** using fiber optics and no folded optics to get the path lengths equal. The peak of the 2WM gain Γ is found only for a path length difference of ± 1 mm. The result corroborates the

finding of the coherence length being ≈ 4.3 mm from Figure 6.3. It says that the coherence length corresponding to 90% of the maximum Γ is more significant to 2WM than a conventional $V(z)$ FWHM coherence length, although the two are related. Figure 6.4 also shows small peaks in Γ spaced 4.5 mm apart. The peak separation corresponds to the cavity mirror spacing of the Nd:YAG rod that is the source for the laser.

6.2.1.4 Beam Diameter

Figure 6.5 Knife edge profile of the YAG beam at $\lambda = 1.064$ μm .

Figure 6.5 shows the profiles of the laser beams at $\lambda = 1.064$ μm . The profiles were obtained using a vertically oriented razor edge mounted on a digital micrometer. The micrometer moved the blade through the beams and the power passing the blade was measured. Although the beam is not exactly Gaussian in profile, the beams are modeled as Gaussian since the approximation covers most of the beam power. Under the Gaussian model, one finds that the FWHM is twice the 3 $\rightarrow$ 6.1 dB loss distance, and the $1/e^2$ diameter is twice the 3 $\rightarrow$ 16.4 dB loss distance. The ratio of $1/e^2$ and FWHM in a true Gaussian beam is a constant. The calculated ratio for the Nd:YAG laser from knife edge measurements is within ± 15 percent of the ideal Gaussian ratio.

Figure 6.6 shows the profiles of the laser beams at $\lambda = 1.152$ μm . The profiles were obtained using a vertically oriented razor edge. The calculated ratio for the HeNe

Figure 6.6 Knife edge profile of the HeNe beam at $\lambda = 1.152 \mu\text{m}$.

laser from knife edge measurements is within ± 15 percent of the ideal Gaussian ratio.

6.2.2 Observations

Figure 6.7 shows the experimental results for photorefractive gain Γ vs optical intensity I_0 at $\lambda = 1.064 \mu\text{m}$. Each point represents a 1000 times averaged measurement. The pump was chopped at 15 Hz. The gain rises from zero with I_0 . The intensity varied

Figure 6.7 Photorefractive gain in InP:Fe: gain Γ versus I_0 at $\lambda = 1.064 \mu\text{m}$, $\Lambda = 1.18 \mu\text{m}$

from 1 to 600 mW/cm². Grating spacing was equal to 1.18 μm. The gain rises proportional to the log of the intensity until it appears to level off, or saturate, at $\Gamma \approx 0.45 \text{ cm}^{-1}$ for $I_0 > 400 \text{ mW/cm}^2$. In saturation the optical intensity dominates over the thermal carriers. For optical intensity less than 400 mW/cm², the gain varies proportionally to the log of the optical intensity. The relation is shown as the straight line in the graph:

$$\Gamma = c_1 \ln \left(\frac{I_0}{I_{\text{dark}}} \right) \quad (6.12)$$

where

$$\begin{aligned} c_1 &= 0.085 \text{ cm}^{-1} \\ I_{\text{dark}} &= 1.3 \text{ mW/cm}^2 \end{aligned}$$

The log form of the data and the straight line fit are similar to the PISCES result depicted in Figure 5.21 and Figure 5.22. The beam FWHM diameter was 0.618 mm. The pump to signal intensity ratio was 30, corresponding to a modulation index of 0.35. The extinction ratio of the pump/signal was 21 dB/26 dB respectively. The interaction length was limited by the beam diameter to 2.5 mm.

Figure 6.8 shows the experimental results for photorefractive gain Γ vs optical intensity at $\lambda = 1.152 \text{ μm}$. The curve depicts the gain rising to 0.22 cm⁻¹ as the intensity varies from 1 to 125 mW/cm². Grating spacing was equal to 1.29 μm. The gain does not

Figure 6.8 Photorefractive gain in InP:Fe: Γ versus I_0 at $\lambda = 1.152 \text{ μm}$, $\Lambda = 1.29 \text{ μm}$

Figure 6.9 Response time in InP:Fe: τ versus I_0 at $\lambda=1.064 \mu\text{m}$, $\Lambda=1.18 \mu\text{m}$

level off the way it did for $\lambda=1.064 \mu\text{m}$. The gain does not level off because there was insufficient power at $\lambda=1.152 \mu\text{m}$ to saturate the crystal. The gain follows approximately an exponential s-curve:

$$\Gamma = c_2 \left[1 - \exp\left(-\frac{I_0}{I_{\text{dark}}}\right) \right] \quad (6.13)$$

where

$$\begin{aligned} c_2 &= 0.25 \text{ cm}^{-1} \\ I_{\text{dark}} &= 65 \text{ mW/cm}^2 \end{aligned}$$

The form of the equation is different from that form for $\lambda=1.064 \mu\text{m}$. It is not known whether the difference is attributed to the difference between processes in the crystal at the two wavelengths, or something else. The beam FWHM diameter was 0.272 mm. The pump to signal intensity ratio was 27, corresponding to a modulation index of 0.37. The extinction ratio of the pump/signal was 21 dB/13 dB respectively. The interaction length was limited by the beam diameter to 1.1 mm.

Figure 6.9 shows the experimental results for response time τ vs optical intensity I_0 at $\lambda=1.064 \mu\text{m}$. The chop rate was 100 Hz, instead of 15 Hz used for the gain measurement, because of timing jitter introduced by the chopper's internal timing electronics at the lower rate. Each point represents between 100 and 1000 averaged chops

of the pump. The variability in the averaging number is due to manually monitoring the averaged signal over time and terminating the averaging when the shape of the signal changed. The observed shape of the detected signal changed after turning on the pump due apparently to laser induced heating of the InP:Fe medium. The data points exhibit some scatter. The scatter seems to be due to material parameters changing and causing the photorefractive beam coupling process to deliver differing gains. The coupling did not always turn on in a well behaved damped exponential fashion. There were oscillations in the 2WM signal, leading to some guessing about the level to measure the time constant. Vignetting caused by the finite mechanical chopping time is not believed to be the source of error for two reasons. First, the mechanical chop time was approximately 0.5 milliseconds at the 100 Hertz chopping rate with a 1.5 mm beam diameter. This is less than all of the measurements. Second, the $1/e$ timing measurements were made after the 0.5 millisecond pump beam turn-on time.

For these low power levels, the response time is roughly proportional to the inverse of the optical intensity, as expected from Chapter 4. The best fit for the response time, represented by the curve labeled $1/(I_0 + \text{offset})$ in the figure, is given by:

$$\tau = \frac{1}{c_3 I_0 + \frac{1}{\tau_{\text{offset}}}} \quad (6.14)$$

Figure 6.10 Response time in InP:Fe; τ versus I_0 at $\lambda = 1.152 \mu\text{m}$, $\Lambda = 1.29 \mu\text{m}$.

where

$$c_3 = 0.0040 \text{ cm}^2/\text{mW}/\text{msec}$$

$$\tau_{\text{offset}} = 2.32 \text{ msec}$$

This curve fits the measurements well except for the lowest optical pump intensity. A second curve in the figure, labeled " $1/I_o$ ", depicts the inverse optical intensity fit to the response time measurements:

$$\tau = \frac{100}{I_o} \quad (6.15)$$

This is the expected best fit. However, it does not fit the measurements well.

Figure 6.10 shows the experimental results for response time τ vs optical intensity I_o at $\lambda = 1.152 \mu\text{m}$. The response times for equal intensity are longer than with $\lambda = 1.064 \mu\text{m}$. For example, the response time for $I_o = 100 \text{ mW}/\text{cm}^2$ is 1.2 msec and 4.0 msec at $\lambda = 1.064 \mu\text{m}$ and $1.152 \mu\text{m}$, respectively. The increase is expected because the InP:Fe absorption coefficient is 2.3 times smaller at the longer wavelength. Extra scatter in these points comes from a large 60 cycle power supply rectification hum on the $\lambda = 1.152 \mu\text{m}$ signal that had some 1 KHz frequency components. Two theoretical best fit curves are depicted in **Figure 6.10**. They show usual the reciprocal intensity dependence (labeled $1/I_o$) where

$$\tau = \frac{70}{I_o} \quad (6.16)$$

and the better approximation

$$\tau = 11.9 \times \left(\frac{1}{I_o} \right)^{\frac{1}{4}} \quad (6.17)$$

where the response time depends on the -0.25^{th} power of the optical intensity. The result is not predicted from the classic photorefractive theory outlined in **Chapter 4**. Without a physical mechanism or other's findings to explain the result, the result would appear to be a systematic measurement error coming from the AC spikes on the laser.

Figure 6.11 shows a normalized typical oscilloscope trace of the 2WM signal at $\lambda = 1.152 \mu\text{m}$. The signal rises more or less exponentially from the time that the pump turns on. When the pump turns off, the signal quickly falls back to its original level.

Figure 6.11 Normalized 2WM oscilloscope data showing the increase in the signal attributed to optical photorefractive gain.

Figures 6.12 and 6.13 plot $1/\tau_{\text{measured}}$ against I_0 at $\lambda = 1.064 \mu\text{m}$ and $\lambda = 1.152 \mu\text{m}$. The graphs show there is no reciprocal relationship between τ and I_0 .

Figure 6.12 Reciprocal response time in InP:Fe: $1/\tau$ versus I_0 at $\lambda = 1.064 \mu\text{m}$, $\Lambda = 1.18 \mu\text{m}$

6.2.3 Summary

This section described the constant $\Lambda=1.18 \mu\text{m}$ and $\Lambda=1.29 \mu\text{m}$ photorefraction experiment in InP:Fe at $\lambda=1.064 \mu\text{m}$ and $1.152 \mu\text{m}$ respectively. The effects of laser heating and coherence length are discussed. The beam diameter and beam power measurement techniques are detailed. Experimental measurements for gain and response time were presented and compared to theory.

It was found that the InP:Fe behaved consistently with the model based on the Kukhtarev equations. Maximum gain Γ was found to be 0.47 cm^{-1} and 0.22 cm^{-1} for $\lambda=1.064 \mu\text{m}$ and $1.152 \mu\text{m}$ and $I_0=550 \text{ mW/cm}^2$ and 120 mW/cm^2 respectively. Gain did not exceed absorption in this experiment, where $\alpha=0.93 \text{ cm}^{-1}$ and 0.41 cm^{-1} respectively. The response times were proportional to I_0^{-1} . Minimum $1/e$ response times were 0.9 msec and 2.2 msec respectively. As I_0 decreased, the required time to build up a steady state photorefractive effect would increase to 6.5 msec and $10. \text{ msec}$ for $I_0=5 \text{ mW/cm}^2$ and 1 mW/cm^2 ,

Figure 6.13 Reciprocal response time in InP:Fe: $1/\tau$ versus I_0 at $\lambda=1.152 \mu\text{m}$, $\Lambda=1.29 \mu\text{m}$. respectively.

6.3 Experiment 2: Γ vs Polarization, E, and Λ

This Section discusses the variable grating spacing Λ experiment: gain versus polarization angle ϕ , gain versus externally applied electric field E_0 , and gain versus Λ measurements are reported.

6.3.1 Experimental Set-up

6.3.1.1 Single Mode Fiber-Based Beam Formation

The experimental set-up is depicted in **Figure 6.14**. The randomly polarized diode pumped Nd:YAG laser at $\lambda = 1.064 \mu\text{m}$ is coupled into an FC connectorized single mode fiber using an Optics for Research HCC thread mount. The light is routed through a fusion splice into a Gould 200748 1x2 directional coupler. The coupler is used as a power splitter. Out of the coupler comes two fibers, a signal fiber and a pump fiber. The signal and pump fibers were cut to one meter lengths and match each other in length to within five millimeters. Due to the relatively short length of fiber before the launch into free space, steps had to be taken to reduce the magnitude of higher order core modes causing multimoding and interfering with the desired single mode output.

Both core and cladding modes were considered. First, extraneous core modes were eliminated using experimentally selected tightly wound mandrels. Several 24 millimeter diameter mandrels were wound immediately after the directional coupler to eliminate multiple core modes generated in the coupler junction and to obtain single mode TEM_{00} output. Second, cladding modes were eliminated using mode strippers at both the input and output of the directional coupler. Each mode stripper was implemented by stripping the plastic jacket from the glass fiber cladding over a two centimeter length. The stripped fiber was then placed in index matching gel surrounded by a protective four centimeter length of 3/16 inch diameter glass tubing. As a result of these precautions, only the desired single mode light was observed at the end of the optical fibers.

The fibers end in two fiber launching stages, each consisting of an xyz chuck for positioning and an f/20 objective lens. The objective lenses collimate the light coming from the fiber ends, and focus it to pass through the one mm thick, one cm long InP:Fe crystal aperture positioned approximately 75 centimeters away. The beams were observed to be TEM_{00} as expected coming from single mode fiber incorporating both cladding mode protection and multimode protection.

6.3.1.2 Beam Characteristics

Polarizing beam splitters depicted as squares just in front of the objective lenses pass only the vertically polarized component. The signal polarization is then modified

Figure 6.14 Second experimental set-up for 2WM in InP:Fe.

as desired with the $\lambda/2$ waveplates. To calculate the incident power intensity, the total power and the Gaussian beam diameters are measured. The signal and the pump beam diameters are determined at the crystal by reflecting the light with a flat test mirror positioned a few centimeters before the crystal. The folding mirror is not shown in **Figure 6.14**. The micrometer controlled knife edge is placed the same number of few centimeters after the added mirror, and the knife cuts through the beam. FWHM and $1/e^2$ diameters are measured at 3→6.1 dB and 3→16.4 dB points respectively. The diameter ratios were found to be ± 15 percent of the ideal Gaussian ratio of 2.885. Because the knife edge cuts the beam at the same z-axis point in the laser beams as the crystal normally is without the folding mirror, one may be confident that the measured profile is the actual profile at the crystal.

6.3.1.3 Chopper

In the first experimental set-up, the 10 Hz chopper blade cut the four millimeter diameter beam in about three milliseconds. The time is comparable to the PR

Figure 6.15 Single beam adjustment of chopper assembly for minimum chopper time, with no 2WM.

response time. Therefore vignetting interfered with the measurement of response times less than ten milliseconds long.

To eliminate vignetting, in the second experiment the chopper was modified from the original set up. Two lenses were placed in the beam around the chopper as shown in **Figure 6.14** to focus the beam to a small spot of diameter $\approx 20 \mu\text{m}$. The chopper blade cuts the narrower pump beam faster than before. The increase in speed results in more accurate measurements of the time response of 2WM. The lenses were ten millimeter diameter diode laser glass achromatic doublets with 25 mm focal length. They were mounted in 25 millimeter diameter lens holders. The holders slid up and down a brass barrel, and were separated by the chopper blade. The lenses were positioned for maximum performance as follows. The entrance pupil lens was held in approximately the correct position 25 millimeters from the chopper by tightening a thumbscrew. The position of the chopper blade and the exit pupil lens was modified until the minimum chopping time was observed. Then the chopper mount and the exit lens were screwed down. The detected pump beam without 2WM is shown in **Figure 6.15**. The 10 Hz chopper cuts the beam and the signal rises to its 90 percent point in about $35 \mu\text{sec}$. Without the lens one expects approximately three millisecond chop time if the beam is 1.5 mm in diameter. A factor of 85 improvement was obtained after deburring the chopper blade with a light application of steel wool and increasing the beam diameter into the entrance pupil using the pump beam's objective lens at the end of the pump fiber. One expects the minimum chop time to be the point where the blur circle of diameter D due to diffraction effects equals blur circle diameter due to the spherical aberration glass effects [128]:

$$D = \frac{0.067 \, d^3}{f^2} = \frac{1.27 \lambda f}{d} = 8 \, \mu\text{m} \quad (6.18)$$

glass
spherical
abberation

diffraction

where d is the laser beam diameter at the entrance pupil, and f is the lens focal length. Here the optimum laser beam diameter is four millimeters and the ideal estimated chop time is calculated to be $17 \mu\text{sec}$. This ideal chop time is not attained. Instead, the fastest observed chop time was $34 \mu\text{sec}$. The observed chop time is two times longer than the

predicted chop time. The difference is due to hand adjustments of the focusing lenses, the non-ideal entrance beam diameter, and the finite thickness of the chopper blade.

For situations where the photorefractive time constant approached one second, the chopper was not able to function at the required 0.1 Hz. The chopper was disabled, and the beam was modulated by manually moving a one half inch thick block of rubber into the beam.

6.3.1.4 Crystal Environment

The crystal is cooled with a thermoelectric (TE) cooler. The crystal is thermally connected and electrically insulated from the TE element by 22 mm of MACOR¹⁰ and a thin film of DOW CORNING 340 silicone heat sink compound. The TE cooler is mounted on a copper heat sink and the whole apparatus is on a xyz0,0,0, positioner. Extra cooling comes from a nearby miniature box fan. The Cambion temperature controller drives the TE cooler using the resistance of the YSI precision thermistor YSI 44011. The thermistor is embedded in silicone in the MACOR half way between the InP:Fe crystal and the TE cooler. The temperature at the top of the MACOR was characterized as shown in Figure 6.16. It shows temperature versus Cambion controller setting, where the temperature is measured using a Newport chrome-alumel thermocouple held solidly against the MACOR and surrounded by silicone thermal compound, one millimeter away from the thermistor. The error bars in this graph represent three standard deviations from the mean temperature over ten separate readings. The system was left to settle for five minutes for each new temperature.

For the experiments, the crystal is mechanically held in place using a nylon screw forced down with a weight on a vertical slider. This ensures adequate thermal contact for the crystal through a minimum of silicone to the MACOR. Electrodes are applied to the crystal and touch it from above, not from the side. The electrodes are soft spring loaded to allow some degrees of freedom to the crystal as it is being positioned. The reason behind applying voltage from the top is that the crystal is a large 1x2 cm piece of InP:Fe wafer, and only the light passing through the middle of the piece is used for 2WM. The

¹⁰ MACOR is a registered trademark of CORNING Corporation.

electrodes must be close to the one millimeter wide optically active region because of the relatively high conductivity of the illuminated volume. Most of the voltage is lost to the dark low conductivity regions when the electrodes are placed on the sides of the large piece of InP:Fe. The dark part of the crystal acts like a gigaohm voltage dropping resistor. It reduces or eliminates the 2WM gain Γ enhancement due to externally applied field because the low conductivity regions shield the optically active region from the external field.

The $\langle 001 \rangle$ cut crystal in **Figure 6.15** is oriented in the same manner as in **Figure 6.1** in the first experiment (log book 6 page 40/60) with the grating vector Λ and the applied electric field E_0 in $[110]$. The pump is again moving in the $[110]$ direction. The z-axis is again $[1-10]$.

6.3.1.5 Detection

After passing through the crystal, the signal beam is detected in the same fashion as in **Figure 6.1**, with a couple of differences. The similarity in detection is that the

Figure 6.16 Thermo-electric (TE) cooler performance.

beam passes through a ND wedge, an objective lens, a band pass filter, and into a photodiode. The signal is observed in a digital storage oscilloscope and data is transferred to a computer. The first difference in detection is that the signal beam is deflected by two mirrors before detection. The folded geometry isolates the signal beam from scattered pump light from the crystal. The isolation arises from having the signal and the pump travelling in opposite directions at the detector. The second difference in detection used in the second experiment is that the pump beam is also detected and is used to trigger the oscilloscope. Since introducing the faster chopper in the pump path, the pump is a much better trigger than the signal. The trigger is useful because although the signal under ideal conditions has a large 2WM gain and can trigger itself, under other conditions there is less gain and the signal is not a reliable trigger.

6.3.2 2WM vs Polarization

Figure 6.17 shows photorefractive gain as a function of beam polarization angle ϕ from the vertical. The maximum 2WM gain Γ was found when the polarization of both pump and signal was $\approx 45^\circ$ tilted from vertical, approximately in the [111] direction. The gain vs polarization data points fit the curve:

$$\Gamma(\phi) = 0.45 \sin\left(\frac{\pi}{180}(2\phi + 5.0^\circ)\right) + 0.15 \text{ cm}^{-1} \quad (6.19)$$

When the signal and the pump are at $\phi = 45^\circ$ wrt vertical, 2WM accesses the maximum index variation available on the index ellipsoid as discussed in Chapter 4, Section 4.2.2 for the experimental configuration with Λ in [110]. The observed equation corresponds to equation (4.59) assuming that the 5° term in equation (6.19) is an experimental error. The 0.45 cm^{-1} amplitude sinusoidal part to the gain curve is due to the photorefractive grating. The 0.15 cm^{-1} DC part of the curve is due to the photochromic grating. From equation (4.59), the maximum value of E_{xc} for zero bias in this sample of InP:Fe is equal to 2.3 kV/cm , based on the 0.45 cm^{-1} amplitude coefficient in equation (6.19).

3

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3

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Figure 6.17 Photorefractive gain versus polarization angle for Λ in $[110]$ and the optic axis in $[1\bar{1}0]$.

6.3.3 Zero Bias

This section relates the zero bias gain versus grating spacing measurements to the Debye length l_d in semiconductors. A technique is demonstrated to extract a value for iron concentration [Fe]. Experimental plots of gain versus grating spacing under zero bias have a relative maximum where the grating spacing Λ is equal to the Debye space charge screening length l_d for the dominant photocarrier.

l_d is the maximum distance over which a large difference in the number of positive and negative charges can occur. It increases with the square root of temperature and decreases with the square root of the number of acceptors. Qualitatively the Debye length is range dividing two photorefractive space charge domains. For ranges shorter than l_d , an acceptor exerts a significant force on a carrier. This is space charge screening. For ranges longer than l_d , an acceptor exerts little force on a carrier and quasi-neutrality must exist [129]. Positive and negative clouds emerge and interact with each other through Poisson's law. l_d is the upper limit to short range individual particle interactions, before charge clouds form.

The Debye length is the space charge screening length of electric tightening by E_{sc} . It turns out to be the distance that allows for the maximum space charge field in unbiased crystals. The reason for this is that it optimizes longer range space charge diffusion and shorter range space charge screening. The Debye length l_d is equal to:

$$l_d = \left(\frac{\pi e_o k_B T}{e N_A} \right)^{1/2} \quad (6.20)$$

where N_A is $[Fe^{+2}]$ or $[Fe^{+3}]$, depending on the photocarrier, holes or electrons respectively.

For $\Lambda \gg l_d$, the space charge field is the diffusion field. The charge separation is limited by the diffusion length l_d [130]:

$$l_d = 2\pi \left(\frac{\mu k_B T}{e N_A \tau_R} \right)^{1/2} \quad (6.21)$$

l_d is the $1/e$ hop length of the average excited carrier. l_d is different from l_s because while l_s considers temperature and acceptor concentration, l_d also depends on mobility μ and

recombination time τ_R .

For $\Lambda < l_s$, the space charge clouds diffuse and interfere with each other, partially screening and reducing the space charge field. The maximum in gain for zero bias happens when the diffusion field E_D equals the theoretical limiting space charge field E_q for N_T photorefractively mobile charges [131]:

$$E_D = \frac{2\pi k_B T}{\Lambda e} = \frac{N_T e \Lambda}{2\pi \epsilon \epsilon_0} = E_q \quad (6.22)$$

where N_T must be less than or equal to the deep level trap density in SI InP:Fe.

A larger gain is not physically realizable under zero external bias because the crystal does not support any larger values of space charge field with the optimum value of Λ . The reason for this is charge screening: the photocarriers are so close to the traps that they are quickly trapped, and cannot pool into clouds. As a result, N_T may be determined by knowing the value of grating spacing that gives the largest gain [132] [133] [134]:

$$N_T = \frac{4\pi^2 k_B T \epsilon \epsilon_0}{e^2 \Lambda^2} \quad (6.23)$$

The formula applies when there is only one photocarrier, and when the optical intensity I_0 saturates the gain. If there are two photocarriers, N_T is the effective number of photorefractively mobile charges. This requires $I_0 \geq 100 \text{ mW/cm}^2$ in InP:Fe, 100 mW/cm^2 , and 400 mW/cm^2 in GaAs:EL2. For example, Klein used $I_0 = 4000 \text{ mW/cm}^2$ in his determination of $[\text{EL2}^+] = 1.3 \times 10^{15} \text{ cm}^{-3}$ in GaAs:EL2 [135]. Because in this thesis the optical fiber based gain vs grating spacing set up allowed maximum power densities of 5 mW/cm^2 , it is impossible to use the observed peaks for InP:Fe and GaAs:Cr at about $\Lambda = 4 \mu\text{m}$ to determine $[N_A]$. For $\Lambda \leq 4 \mu\text{m}$, and $I_0 < I_{\text{sat}}$, experimental space charge field is both screened and washed out by thermal effects. As a result, it is not possible to use the formula for a straight forward solution to find the iron concentration in InP:Fe, $[\text{Fe}] = [N_A]$ in the experiment.

Figure 6.18 shows gain versus grating spacing for the sample of Crystacomm

Figure 6.18 Photorefractive gain versus grating spacing for zero bias: InP:Fe, crystal orientation 1-C, $\phi = +45^\circ$ with respect to vertical, Λ in [110].

LEC grown InP:Fe wafer at $\lambda = 1.064 \mu\text{m}$. The crystal dimensions are 1.0 cm $\langle 110 \rangle$ x 2.0 cm $\langle 110 \rangle$ x 0.11 cm $\langle 001 \rangle$, on the optical axis, the bias axis, and the thermal axes, respectively. The grating vector is [110]. The copolarized pump and probe beams are polarized at $\phi = 45^\circ$ with respect to vertical. Temperature was constant at 16°C . The error bars represent plus or minus one standard deviation on the data. The clear peak at around $\Lambda = 4 \mu\text{m}$ indicates where unsaturated shorter range effects begin to dominate long range diffusion, but the formula for $[N_A] = [\text{Fe}^{+3}]$ above does not apply because the gain Γ is not

saturated with optical intensity I_0 .

Figure 6.19 shows gain versus grating spacing for the sample of Cominco LEC grown GaAs:Cr wafer at $\lambda = 1.064 \mu\text{m}$. Again the crystal is unsaturated and $N_A = [\text{Cr}^{+3}]$ does not apply. The crystal dimensions are $0.50 \text{ cm } \langle 110 \rangle \times 0.35 \text{ cm } \langle 001 \rangle \times 0.50 \text{ cm } \langle 1\bar{1}0 \rangle$, on the optical, bias, and thermal axes respectively. The grating vector Λ is $[001]$. The copolarized pump and probe beams are linearly polarized at $\phi = 0^\circ$ with respect to vertical

Figure 6.19 Photorefractive gain versus grating spacing for zero bias: GaAs:Cr, small sample, crystal orientation 1-D, $\phi = 0^\circ$ (vertical polarization), Λ in $[001]$.

along the $\langle 1\bar{1}0 \rangle$ crystal axis. Temperature was constant at 16°C.

It may be possible to use these experimental curves to determine an approximate value for $[N_A]$ even though the crystal's deep level traps were not optically saturated. Such an analysis has not been published in the available literature.

6.3.4 External Bias

The next two sections detail the experimentally measured maximum photorefractive gain characteristics of the InP:Fe crystal under conditions of DC high voltage bias. The available bias varied from 0 to 15 kV/cm. Electron saturation velocity effects are observed when bias exceeded 10 kV/cm. **Section 6.3.4.1** investigates the temperature-intensity resonance and measures a modest gain peak in an InP:Fe crystal. **Section 6.3.4.2** uses the resonance information to obtain gains of up to 7 cm^{-1} by varying the grating spacing.

6.3.4.1 2WM Optimum Temperature

A temperature intensity resonance was observed in InP:Fe. The resonance allows one to calculate the material parameter $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$. The most recent model of 2WM in InP:Fe was presented by Picoli et. al. at Centre National d'Etudes des Télécommunications (CNET) in 1989 [136]. It predicts a resonance in an arbitrary crystal of semi-insulating InP:Fe. For any given optical pump intensity, 2WM gain is a maximum for one specific crystal temperature. The specific temperature depends on wavelength and crystal parameters. For example, at the optimum temperature for a certain intensity, Partovi et. al. predict 20 cm^{-1} gain Γ at wavelength $\lambda = 1.06 \mu\text{m}$ for a fringe spacing $\Lambda = 15 \mu\text{m}$ and an applied DC electric field $E_0 = 10 \text{ kV/cm}$. At any other intensity, the gain is less. The thesis observed an intensity tuned peak in gain. The gain peak had a width FWHM of about 30 percent of I_0 . The CNET theoretical FWHM increases approximately linearly with temperature from 20 percent to 30 percent between 17°C and 32°C. **Figure 6.20** depicts the intensity giving the maximum gain increasing with temperature. It shows the theoretical and experimental results reported by Picoli et. al. along with the thesis's experimental result labeled "NRC EXP". The results show agreement with each other and the theoretical curve. The theoretical curve "NRC THEORY" is developed in the next few

OPTIMUM INTENSITY vs InP:Fe TEMPERATURE

Figure 6.20 Optimum intensity versus crystal temperature for InP:Fe. The line represents the theoretical curve. The data points represent experimental results and CNET theory.

pages.

6.3.4.1.1 Observation of T-I Gain Resonance

The observed result in the thesis is a maximum gain of 0.87 cm^{-1} for 16°C , $I_0 = 3.7 \text{ mW/cm}^2$, and $\Lambda = 2.5 \text{ }\mu\text{m}$. It was found by varying the pump intensity while keeping the pump to signal intensity ratio constant at approximately 700 to 1 (modulation index ≈ 0.076). The results showing 2WM gain versus pump intensity are plotted in **Figure 6.21**. The error bars represent plus and minus one standard deviation of each collection of data points. The probability that the true value of gain passes through these bars is 67 percent. The middle of the crystal's temperature is estimated as the average of the TE cooler stabilized MACOR surface of the crystal stage (12°C) and the room ambient air temperature (20°C).

I_0 is the intensity at the center of the pump beam inside the AR coated InP:Fe crystal. I_0 is calculated using knife edge versus power measurements on the pump beam. I_0 decreases through the crystal through optical absorption. In the one centimeter length the InP:Fe absorption coefficient is 0.93 cm^{-1} , and I_0 falls to 39 percent of its original value. Therefore the resonance condition can not be met throughout all of the crystal volume with a simple temperature control scheme. Two such schemes are discussed in the summary at the end of this section.

Figure 6.21 Gain in InP:Fe versus optical intensity for $\Lambda = 2.5 \text{ }\mu\text{m}$ in [110], 16°C crystal temperature, and 10 kV/cm DC bias. Optical polarization $\phi = \pm 45^\circ$ from vertical.

The intensity-temperature resonance results from two independent processes exciting electrons and holes. They balance the mixed conduction in semi-insulating InP:Fe, causing the rate of hole creation to equal the rate of free electron generation.

6.3.4.1.2 Optical Hole Contribution

When the rate of hole creation (optical intensity driven) equals the rate of free electron creation (temperature driven), the gain is a maximum because the equality produces a $\pi/2$ phase shift between the internal electric field and the optical standing wave given a DC field (4-10 kV/cm typically). Under exposure to light at $\lambda = 1.064$ microns, holes are created by single photon absorption:

$$Fe^{+3} + h\nu \rightarrow Fe^{+2}(5E) + h^+(\Gamma_{15}) \quad (6.24)$$

in which a valence band electron's energy increases from the valence band Γ_{15} to the 5E deep iron center level in the middle of the InP:Fe band gap. These transitions are depicted in Figure 3.1 for $300 \pm 20^\circ K$. As temperature rises the valence band energy level rises (i.e., transition energies fall) at around 0.27 meV/ $^\circ K$, while the energy relations between 5E, 5T₂, and the conduction band remain relatively constant. The temperature dependent transition energies can be calculated as discussed in Chapter 3.

The rate of hole creation is e_p^o , and was already considered in Chapter 3. e_p^o has units of holes/available Fe^{+3} site/sec. Recalling equation (3.57),

$$e_p^o = \sigma_p^o \frac{I_o}{h\nu} \quad (6.25)$$

where

$$\begin{aligned} \sigma_p^o &= \text{photoionization cross-section for holes at } \lambda = 1.064 \text{ } \mu\text{m and } 300^\circ K^{11} \\ &= 3.3 \times 10^{-16} \text{ cm}^2 \\ \sigma_n^o &= \text{photoionization cross-section for electrons at } \lambda = 1.064 \text{ } \mu\text{m and } 300^\circ K \\ &= 1.7 \times 10^{-18} \text{ cm}^2 \end{aligned}$$

Holes dominate optical absorption due to the large value of σ_p^o relative to σ_n^o . At $\lambda = 1.064 \text{ } \mu\text{m}$ the photoionization cross-section is 194 times larger for holes (Fe^{+3}) than

¹¹ Photoionization cross section σ_p^o varies with temperature, but the variation is not taken into account here because all of the work is around room temperature $\pm 20^\circ C$. Most of the photorefractive work is conducted at $16^\circ C$. For example, at $77^\circ K$ the cross section for holes is $1.2 \times 10^{-16} \text{ cm}^2$. The optical cross-section has risen about 1.2 percent per degree Celcius in a temperature increase of 222° .

it is for electrons (Fe^{+2}) at room temperature. In semi-insulating InP:Fe, the ratio of absorption due to holes to absorption due to electrons α_h/α_e is always greater than unity. For example, in the dark, even when $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 11.5$, $\alpha_h/\alpha_e = \sigma_p^0[\text{Fe}^{+3}]/\sigma_n^0[\text{Fe}^{+2}] = 17$. In the light the Fe^{+2} iron is converted into Fe^{+3} and so the ratio becomes even larger. Under all illumination conditions, sources for optical holes are a bigger target than sources for optical electrons.

Picoli et. al. estimated the $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ population ratio to be ≈ 0.30 under 2WM optimum T-I resonance conditions. The thesis found that in the dark, $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 11.5$ in the absorption crystal sample. Under 2WM illumination the thesis found $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] \approx 0.84$ in the 2WM crystal sample. Both the absorption and the 2WM crystal samples were cut from the edge of the same wafer. Assuming that the crystal samples are almost identical, one may conclude that the change in $[\text{Fe}^{+2}]/[\text{Fe}^{+3}]$ comes from optical excitation. Laser light changes the steady state of the crystal, causing Fe^{+3} to be converted into Fe^{+2} .

The optical pump creates holes at a rate linearly proportional to the CW optical intensity. However, there is an intensity limit where so many holes and electrons have been created that the InP:Fe becomes conductive. Current increases suddenly over a period of a few seconds from 100 μamp to supply limited current greater than 1000 μamp . The phenomenon is observed for $I_0 > 200 \text{ mW/cm}^2$ and $E_0 > 5 \text{ kV/cm}$. When the event occurs, 2WM gain falls to $\approx 0.005 \text{ cm}^{-1}$ from typically 5 cm^{-1} . It takes the crystal several minutes of darkness and zero field before large photorefractive gains can be produced again. Presumably the time is required for the crystal's carriers to be trapped and become semi-insulating again.

6.3.4.1.3 Thermal Electron Contribution

Second, consider the thermal creation of free electrons. The main thermal creation process is the direct transition $5E \rightarrow \Gamma_1$, with 0.63 eV activation energy. $5E \rightarrow \Gamma_1$ promotes electrons from the negatively charged lattice trap Fe^{+2} site to the conduction band, thermally mediated by an acoustic phonon. The transition is:

The rate of generation of thermal electrons under steady state conditions is [137] e_n^{th}

electrons per available Fe^{+2} site per second, as was already considered in Chapter 3. From equation (3.18),

$$e_n^{th} = 3.25 \times 10^{25} T^2 \frac{m_e^*}{m} \sigma_n^- \exp\left(-\frac{e_i}{k_B T}\right) m^{-2} K^{-2} sec^{-1} \quad (6.27)$$

where

T	=	temperature in °K
m_e^*/m	=	0.078
σ_n^-	=	electron capture cross-section at infinite temperature in Fe^{+2}
	=	$3.5 \times 10^{-14} cm^2$
e_i	=	$5E \rightarrow \Gamma_1$ thermal activation energy
	=	$0.63 \pm 0.01 eV$
k_B	=	Boltzmann's constant
	=	$1.38 \times 10^{-23} Joules/K$
	=	$8.6 \times 10^{-5} eV/K$

6.3.4.1.4 Solve for $[Fe^{+2}]/[Fe^{+3}]$

The temperature-intensity resonance in Γ happens when the optical creation rate of holes equals the thermal creation rate of free electrons:

$$e_p^o [Fe^{+3}] = e_n^{th} [Fe^{+2}] \frac{\text{charge carriers}}{cm^3 - second} \quad (6.28)$$

and using the expressions for e_p^o and e_n^{th} from above,

$$\sigma_p^o \frac{I_o}{h\nu} [Fe^{+3}] = 3.25 \times 10^{25} T^2 \frac{m_e^*}{m} \sigma_n^- \exp\left(-\frac{e_i}{k_B T}\right) [Fe^{+2}] \quad (6.29)$$

and the value of optical pump intensity I_o for the maximum 2WM gain can then be expressed as a function of temperature:

$$I_o = 3.25 \times 10^{25} \frac{m_e^*}{m} \frac{\sigma_n^-}{\sigma_p^o} h\nu T^2 \frac{[Fe^{+2}]}{[Fe^{+3}]} \exp\left(-\frac{e_i}{k_B T}\right) \quad (6.30)$$

which reduces from the constants given above to:

$$I_o = 5.03 \times 10^7 \frac{[Fe^{+2}]}{[Fe^{+3}]} T^2 \exp\left(-\frac{7304}{T}\right) \quad (6.31)$$

where I_o is in units of watts/meter². Using the expression for I_o , it is possible to evaluate the gain optimizing intensity I_o at any temperature if one knows the InP:Fe material parameter $[Fe^{+2}]/[Fe^{+3}]$. The $[Fe^{+2}]/[Fe^{+3}]$ parameter is not supplied by the manufacturer. Therefore it is necessary to measure $[Fe^{+2}]/[Fe^{+3}]$ by sweeping either I_o or T and looking

for a maximum in photorefractive gain. The operation was performed as shown in **Figure 6.21** by sweeping intensity I_0 . For the 2WM sample of InP:Fe, gain is maximized for $I_0 = 3.7$ mW/cm² = 0.037 W/cm², and $T \approx 16^\circ\text{C}$. For this data the T - I_0 relationship yields $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 0.84 \pm 0.07$ assuming $\pm 1^\circ$ uncertainty in the temperature. The number is comparable to $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] \approx 0.43$ obtained by Picoli et. al. In the experimental sample, the expression for the value of I_0 in units of mW/cm² for maximum photorefractive gain at a given temperature T reduces to:

$$I_0 = 4.23 \times 10^7 T^2 \exp\left(-\frac{7304}{T}\right) \frac{\text{W}}{\text{m}^2} = 4.23 \times 10^6 T^2 \exp\left(-\frac{7304}{T}\right) \frac{\text{mW}}{\text{cm}^2} \quad (6.32)$$

The equation was used to generate the theoretical curve shown in **Figure 6.20**.

For $I_0 = 3.7$ mW/cm², photoconductivity $\sigma_p = 3.7 \times 10^{-9} (\Omega\text{-cm})^{-1}$ as per the measured data represented by equation (6.1). The photoconductivity is comparable to the dark conductivity $\sigma_d = 6.2 \times 10^{-9} (\Omega\text{-cm})^{-1}$. The proximity of the photoconductivity and the dark conductivity may be expected under T - I_0 resonance conditions. At resonance the dark conductivity is almost exclusively due to thermal electrons from the $\text{Fe}^{+2} \rightarrow \Gamma_1$ electron transition, and photoconductivity is almost exclusively due to optically excited holes from the $\Gamma_{15} \rightarrow \text{Fe}^{+3}$ electron transition. Intuitively, the resonance situation where thermal electrons and optical holes contribute equally to the bulk properties of the medium points towards equal contributions to the medium conductivity from photoconductivity and dark conductivity.

The value of $[\text{Fe}^{+2}]/[\text{Fe}^{+3}] = 0.84$ indicates that in the InP:Fe under optimal 2WM illumination conditions there are comparable populations of Fe^{+2} and Fe^{+3} , although $[\text{Fe}^{+3}]$ is slightly greater. Since Fe^{+2} and Fe^{+3} are the only states of Fe in InP:Fe, one has expressions for $[\text{Fe}^{+2}]/[\text{Fe}]$ and $[\text{Fe}^{+3}]/[\text{Fe}]$:

$$[\text{Fe}^{+3}] = \frac{1}{\left(\frac{[\text{Fe}^{+2}]}{[\text{Fe}^{+3}]} + 1\right)} [\text{Fe}] \quad [\text{Fe}^{+2}] = \frac{\frac{[\text{Fe}^{+2}]}{[\text{Fe}^{+3}]}}{\left(\frac{[\text{Fe}^{+2}]}{[\text{Fe}^{+3}]} + 1\right)} [\text{Fe}] \quad (6.33)$$

From **Chapter 3**, $[\text{Fe}] = 3.7 \times 10^{16} \text{ cm}^{-3}$. Therefore $[\text{Fe}^{+3}]/[\text{Fe}] = 0.54 \pm 0.02$, giving

$[Fe^{+3}] = 2.0 \pm 0.1 \times 10^{16} \text{ cm}^{-3}$. $[Fe^{+2}]/[Fe] = 0.46 \pm 0.02$, giving $[Fe^{+2}] = 1.7 \pm 0.1 \times 10^{16} \text{ cm}^{-3}$.

6.3.4.1.5 GaAs:Cr Resonance?

GaAs:Cr was measured for a similar temperature-intensity resonance. The result is shown in Figure 6.22 for 11.4 kV/cm, in peak measurements, 100X averages, and for zero bias. The peak measurements are much larger than the 100X measurements because the 2WM signal is very unstable in GaAs:Cr when external DC field exceeds 5 kV/cm due apparently to periodic charge injection into the GaAs:Cr volume. The curves show that unlike InP:Fe, GaAs:Cr has no low optical power 2WM gain resonance in this I_0 regime. Under HV DC, GaAs:Cr appears to launch electrical domains into the bulk volume. The domains travel to the other electrode and they form a travelling grating in the medium. The grating spacing varies between 0.9 and 3.4 mm and the rate of propagation varies between 15 and 120 mm/sec, depending on the electric field and the optical intensity [138]. This injection phenomenon has not been observed in InP:Fe. Picoli et. al. suggest that while their theory fits InP:Fe well and should fit other III-V semiconductor compounds, one needs a theory that includes injection current [139]. The present theory does not account for an injection region.

While there is no known paper looking at temperature and intensity for high bias in GaAs:Cr, GaAs:Cr was investigated by Cheng and Partovi for 2WM gain depending on temperature and intensity for zero bias electric field [140]. Cheng found that gain fell in GaAs:Cr between 290 and 390°K for intensities between 1.4 and 98 mW/cm². Cheng attributed this to the dark conductivity σ_d increasing above the photoconductivity σ_p and thermally washing out the photorefractive grating. Cheng used the one-carrier and one-level model to adequately describe his experimental results for zero electric field bias.

6.3.4.1.6 Grating Spacing Effect

Grating spacing Λ does not affect the thermal ionization rate or the optical ionization rate for large pump to signal ratios (i.e., in 2WM gain applications). Therefore one neither expects nor observes a variation in the optimum optical intensity for a fixed temperature for Λ between 2 μm and 14 μm [141].

Figure 6.22 Photorefractive gain in GaAs:Cr versus optical intensity for $\lambda=2.5 \mu\text{m}$ in [001], $T=16^\circ\text{C}$, $E_0=11.4 \text{ kV/cm}$ DC bias, $\phi=0^\circ$ in [110].

6.3.4.1.7 Cross-section Effect

Photoionization Cross-section of Holes and Electrons

The intensity-temperature resonance in 2WM gain is the result of holes dominating the optical absorption process and electrons dominating the thermal excitation process. The reason is that the photoionization cross-section of holes σ_p^0 is greater than the photoionization cross-section of electrons σ_n^0 at $\lambda=1.064 \mu\text{m}$. At different wavelengths this does not always hold true. As discussed in Section 3.7, the wavelength dependence of σ_p^0 and σ_n^0 in InP:Fe is significant.

The photoionization cross-section for $5E \rightarrow \Gamma_1$ (producing free electrons) and the photoionization cross-section for $\Gamma_{15} \rightarrow 5E$ (producing holes) at room temperature is a power law depending on λ and the band gap. σ_n° and σ_p° values were tabulated in Tables 3.2, 3.3, 3.4, and 3.5 for various λ . The trends are clearly depicted in Figure 3.4 for 77°K. At 300°K the photoionization cross-sections are larger for holes but the same for electrons. This is because the relevant transition energies are 0.07 eV smaller for holes at 300°K than they are at 77°K due to the rising valence band energy level. The higher the temperature, the smaller the transition energy, and the easier it is for one photon to create a hole.

Optical Absorption Coefficient α of the Crystal

The optical absorption coefficient of InP:Fe falls monotonically as λ increases in the 0.9 μm to 2.0 μm band. This is a direct result of the afore-mentioned decreasing photoionization cross-sections of both electrons and holes in this band. To first order, the optical absorption coefficient α is represented by equation (3.50). It is assumed that only iron in the band gap contributes to absorption, and that there is no intrinsic absorption in InP. The measured values of α for InP, and other samples of III-V compound semiconductors are given in Table 6.2, from results of experimental work in the course of this thesis.

TABLE 6.2 Measured Absorption Coefficients for III-V Compound Semiconductors

λ [μm]	ABSORPTION COEFFICIENT α [cm^{-1}]		
	InP:Fe	GaAs:Cr	GaAs:EL2
1.064	0.93	1.45	2.02
1.152	0.41	1.11	1.44
1.320	0.21	0.54	0.47
1.550	0.10	0.17	0.14

At longer wavelengths, the falling value of the optical absorption coefficient means that more optical pump power is needed to create enough holes to equal the rate of

thermal excitation of free electrons. This condition is essential to drive photorefractive gain into the intensity-temperature resonance. Quanta of longer λ are not absorbed at the same rate per cubic centimeter in the volume of the crystal, resulting in fewer holes being created for the same value of I_0 . This was predicted and observed by Ozkul, Picoli, and Wolffer in moving from $\lambda=1.064 \mu\text{m}$ to $\lambda=1.320 \mu\text{m}$ [142]. Their 0.72 cm long crystal's (Sample # 101A) α decreased from 2.6 cm^{-1} to $\approx 0.5 \text{ cm}^{-1}$ (factor of 5.2)¹², and the required power increased from 15 mW/cm^2 to 100 mW/cm^2 (factor of 6.7). The temperature was 17°C , $E_0=10 \text{ kV/cm}$, beam intensity ratio $\beta \approx 1000$, and $\Lambda=5 \mu\text{m}$. The average crystal gain Γ increased at the longer wavelength to 5.3 cm^{-1} from 4.4 cm^{-1} . This can be attributed to reducing α and the resulting extended region of the crystal's volume that works under near optimum intensity conditions at $\lambda=1.064 \mu\text{m}$.

Decreasing α has two effects of 2WM, one positive and one negative. On the plus side, decreasing α increases the volume of the crystal that can work under optimum intensity conditions. On the minus side, decreasing α demands that one deliver proportionally higher optical pump power to the crystal to achieve the optimum intensity conditions. For a given length of crystal, one may engineer the maximum gain from an amplifier by trading off α against I_0 .

It is worthwhile to observe that the InP:Fe samples used by Ozkul et. al. had measured α between 1.5 cm^{-1} and 2.6 cm^{-1} , while for this work the sample exhibited 0.93 cm^{-1} at $\lambda=1.064 \mu\text{m}$. This work's sample was cut from the tang of an LEC grown boule, meaning that for that boule the concentration of iron was a maximum. Obtaining a wafer from the bottom was originally planned to yield the maximum possible absorption. However, for whatever reason, Ozkul's samples had more than twice the absorption than in this thesis. It implies that purchasing samples distributed from the seed to the tail and running absorption spectra would give an instructive variety of results. For example, discussions with the manufacturer (George Antypas at Crystacomm) indicated that researchers in the photorefractive InP:Fe area use the seed of the boule instead of the tang

¹² The InP:Fe used in this thesis α fell from 0.93 cm^{-1} to 0.21 cm^{-1} (factor of 4.4) over the same band. By scaling against Ozkul's experimental result (factor of 1.29), one predicts that the required value of I_0 in the thesis sample at $1.320 \mu\text{m}$ is $\approx 4.4 \times 1.29 \times 3.7 \text{ mW/cm}^2 = 21 \text{ mW/cm}^2$.

of the boule. Although [Fe] is less at the seed since defects, and unwanted impurities are less, α may be larger at the seed due to larger $[\text{Fe}^{+3}]$.

6.3.4.2 Maximum Gain

The last Section 6.3.4.1 investigated the temperature-intensity resonance at fixed grating spacing and measured a modest photorefractive gain peak as a function of optical intensity in an InP:Fe crystal. This section locks the temperature of the crystal and the optical intensity to the measured T- I_0 resonance maximum (16°C - 3.7 mW/cm^2). Then the grating spacing Λ is varied in a search for maximum photorefractive gain $\Gamma(\Lambda)$. Gains of up to $\Gamma=7 \text{ cm}^{-1}$ or 30 dB/cm are found at $\lambda=1.064 \mu\text{m}$, less absorptive loss α of 0.93 cm^{-1} or 4 dB/cm. Over a 0.5 cm interaction length, the result is an optical amplifier with 13 dB net gain.

6.3.4.2.1 Grating Spacing Effect

The standing wave grating spacing Λ was varied from $1.35 \mu\text{m}$ to $12.3 \mu\text{m}$ in

Figure 6.23 Photorefractive gain in InP:Fe as a function of grating spacing for $\lambda=1.064 \mu\text{m}$, $\beta=500$, $T=16^\circ\text{C}$, $I_0=3.7 \text{ mW/cm}^2$, $E_0=12.5 \text{ kV/cm}$, polarization $\phi=45^\circ$, and Λ in $\langle 110 \rangle$.

the <110> crystal direction with the polarization angle ϕ at 45° . The pump to signal intensity ratio β was fixed at $\beta=500$ so that the pump intensity would always dominate over the signal intensity. The best results were obtained for $E_0=12.5$ kV/cm, and they are depicted in the **Figure 6.23** for various values of external bias. The figure shows data points labelled " $\square$ " for $E=7.5$ kV/cm, " $\diamond$ " for $E=10$ kV/cm, and " x " for $E=12.5$ kV/cm. Points for smaller values of external bias are not depicted in the figure because the gain was close to zero. The data points for $E=12.5$ kV/cm show that gain increases from ≈ 1 cm $^{-1}$ to ≈ 7 cm $^{-1}$ as the grating spacing increases. Physical limitations on experimental geometry such as the length of available optical bench space and the finite width of the fiber optic to free space launchers prevented larger values of grating spacing from being used.

The data points can be described by functions as shown in the curves in the figure. The three curves represent the same empirically derived exponential function, except that the multiplicative constant $\Gamma_{\max}$ varies from curve to curve:

$$\Gamma(\Lambda) = \Gamma_{\max} \times \left(1 - \exp \left(- \left(\frac{\Lambda}{4.5 \mu m} \right)^2 \right) \right) \quad (6.34)$$

The oscilloscope response of the best gain result is depicted in **Figure 6.24** for a period of about 40 seconds. For $0 \text{ sec} \leq t \leq 7 \text{ sec}$ the signal beam alone is present in the bulk crystal. The pump beam turns on at $t \approx 7$ seconds and is removed at $t \approx 47$ seconds. The figure shows measured data (noisy line) and an empirical model (bold line) for the signal beam rising from 130 millivolts detected output to 4.775 volts detected output. There is a peak in the gain near ten seconds, and the gain falls to an intermediate value after that. The empirical model for the time variation of the amplified signal during the time that the pump beam is present is:

$$\begin{aligned} \text{Detected Signal Voltage}(t) &= 2.4 \times \left(\exp \left(- \left(\frac{t-12}{20 \text{ sec}} \right)^2 \right) + 1 \right) \text{ volts} \\ &\text{or alternatively} \\ \text{Gain } \Gamma(t) &= 7.2 \times \left(\exp \left(- \left(\frac{t-12}{20 \text{ sec}} \right)^2 \right) + 1 \right) \text{ cm}^{-1} \end{aligned} \quad (6.35)$$

where photorefractive gain has the same form as the detected signal.

In the equation the time constant of the photorefractive effect in the experimental set-up is approximately 20 seconds. In this case the time constant is different from previously considered time constants. Instead of being related to the time to set up the space charge electric field, the time constant in this case is a thermal parameter. The peak of the photorefractive gain in the figure slowly fell over time after turn-on of the pump beam because of laser-induced crystal heating. As presented in equation (6.7) for the zero bias case, the absorption based laser heating over 40 seconds is just $\Delta T_1 = 0.07^\circ\text{C}$. 0.07°C is not enough to explain the eventual fall of photorefractive gain over time. An adequate increase in temperature can be attributed to the external bias. Although the crystal is temperature stabilized using TE cooling, ohmic heating was observed to upset the crystal's temperature distribution. Heating caused by the dark electrons and photocarriers moving in the electric field increases the crystal temperature according to the bulk thermal temperature rise formula:

$$\left(\frac{\partial T}{\partial t}\right)_{\text{electrical}} = \frac{\text{resistive dissipation per unit volume}}{C \rho} = \frac{\sigma E^2}{C \rho} \quad (6.36)$$

Using the measured conductivity relation for InP:Fe given in equation (6.1) and the

Figure 6.24 Time response of the photorefractive effect in InP:Fe for maximum gain showing maximum gain $\Lambda = 7.2 \text{ cm}^{-1}$.

measured $T-I_0$ optical resonance intensity $I_0 = 3.7 \text{ mW/cm}^2$, the InP:Fe conductivity is $\sigma = 2.3 \times 10^{-9} (\Omega\text{-cm})^{-1}$. For $E = 12.5 \text{ kV/cm}$, the rate of temperature rise is $0.193 \text{ }^\circ\text{C/sec}$, or $\Delta T = 7^\circ\text{C}$ in 40 seconds. Typical values of temperature rise due to the ohmic heating are given in Table 6.3 for $I_0 = 10 \text{ mW/cm}^2$. Any 7°C rise in temperature is enough to explain the observed detuning after 40 seconds in Figure 6.24.

Table 6.3 - Ohmic Heating Induced Temperature Rise

$$I_0 = 10 \text{ mW/cm}^2, \sigma = 1.6 \times 10^{-8} (\Omega\text{-cm})^{-1}$$

EXTERNAL BIAS [kV/cm]	$\delta T/\delta t$ [$^\circ\text{C/sec}$]
0	0
1	0.0087
3	0.078
10	0.87
30	7.8
100	87.

The increase in crystal temperature over time detuned the crystal from the $T-I_0$ resonance, resulting in a non-optimal value of pump intensity. Resistive heating dominates photorefractive $T-I_0$ detuning under the application of external bias because resistive heating effects is one hundred times greater than laser absorption heating. Resistive heating dominates over laser heating at least in CW applications from low optical intensities up to at least $I_0 = 1000 \text{ mW/cm}^2$.

The amplified signal envelope increases for about five seconds before falling off. The increase indicates that the crystal temperature was about one degree Celcius below the exact $T-I_0$ resonance temperature at turn-on time.

The measured noise in the figure is not part of the photorefractive effect, but is a result of air movement in the experimental lay out. The time constant of the photorefractive effect was measured around 100 milliseconds in this set-up. Such a long time constant and random

air movements in the paths of the interfering beams caused the photorefractive gain magnitude to be unstable. Fluctuations in the phase of the intensity standing wave relative to the space charge electric field in the crystal caused the gain to go through a series of maxima and minima. Techniques to improve the stability due to air motion were tried and they are presented in the Conclusion.

The peak gain $\Gamma = 7 \text{ cm}^{-1}$ corresponds to a minimum periodic space charge electric field amplitude E_{sc} of 25 kV/cm. Here equation (1.2) has been used assuming that the electron-hole competition factor $|\xi| = 1$. If $|\xi| < 1$, then E_{sc} is proportionally larger. Comparable and larger periodic fields in the crystal were predicted in Chapter 5 by numerical finite element modelling using the commercial PISCES software package.

6.3.4.1.2 External DC Bias Effect

The applied external electric field E_0 was varied in the $\langle 110 \rangle$ crystal direction from 0 kV/cm to 12.5 kV/cm. The results for $\Lambda = 1.86 \mu\text{m}$ and $\Lambda = 12.3 \mu\text{m}$ are representative and are depicted in Figure 6.25. The figure shows the gain increasing monotonically from

Figure 6.25 Photorefractive gain in InP:Fe as a function of electric field for $\lambda = 1.064 \mu\text{m}$, $B = 500$, $T = 16^\circ\text{C}$, $I_0 = 3.7 \text{ mW/cm}^2$, $\Lambda = 1.86 \mu\text{m}$ and $12.3 \mu\text{m}$, polarization $\phi = 45^\circ$, and Λ in $\langle 110 \rangle$.

0.01 cm⁻¹ to 7 cm⁻¹ as applied electric field increases from zero to 12.5 kV/cm. The gain increases exponentially at first, but by 12.5 kV/cm the gain increases more slowly due to saturation of the charge carrier velocity. For E₀ greater than 15 kV/cm one expects that the gain will fall due to the electron velocity saturation effect. Such hard saturation was not observed because it was not possible to reach 15 kV/cm with the given equipment.

The data points can be described by functions as shown in the curves in the figure. The two curves are observed to follow the same exponential power function, except that the multiplicative constant $\alpha(\Lambda)$ varies according to the empirical function:

$$\Gamma(E_0) = \alpha(\Lambda) \times \left(\frac{E_0}{8 \frac{kV}{cm}} \right)^6 \times \exp \left(- \left(\frac{E_0}{8 \frac{kV}{cm}} \right)^2 \right) \quad (6.37)$$

where $\alpha(\Lambda) = 1.8 \text{ cm}^{-1}$ and 5.5 cm^{-1} at $\Lambda = 1.86 \text{ } \mu\text{m}$ and $\Lambda = 12.3 \text{ } \mu\text{m}$ respectively. In general, the gain increased for longer grating spacing.

6.4 Summary

This section experimentally showed how under external DC bias, balancing the rates of free electron formation and hole formation can be used to optimize 2WM gain Γ in InP:Fe. For $\Lambda = 12 \text{ } \mu\text{m}$ and $E_0 = 12.5 \text{ kV/cm}$, a maximum gain of $\Gamma = 7 \text{ cm}^{-1}$ was observed as optical intensity was varied.

The gain is due to the decoupling of the free electron formation process from the free hole formation process. Electrons depend mostly on thermal excitation at room temperature, and to first order thermal holes can be ignored due to $e_n^{\text{th}} \ll e_p^{\text{th}}$. Holes depend mostly on optical excitation at room temperature, and to first order optical electrons can be ignored due to $\sigma_n^0 \ll \sigma_p^0$. The concentrations of Fe⁺² and Fe⁺³ are about equal under $I_0 = 3.5 \text{ mW/cm}^2$ laser illumination. The laser light has caused [Fe⁺²] to fall from the dark value of [Fe⁺²]/[Fe] = 0.92 (Chapter 3) to 0.46 (T-I resonance). The laser induced concentration change is due to a transient initial conversion of Fe⁺² into Fe⁺³ by optical excitation of conduction band electrons.

The published T-I resonance model of Picoli et. al. has two problems with

erroneous physical constants. The first problem is that the Picoli model employs $[\text{Fe}^{+3}]$ photoionization cross-section $\sigma_p^o = 5 \times 10^{-17} \text{ cm}^2$. It is 6.6 times smaller than the value measured by Valley and modeled in Chapter 3, $\sigma_p^o = 3.3 \times 10^{-16} \text{ cm}^2$. It impacts the resonance model by increasing the estimated resonance intensity by a factor 6.6. Picoli et. al. did not recognize this as a problem. Second, the Picoli model employs a nonphysical value for the thermal ionization of electrons $5E \rightarrow \Gamma_1$ transition energy, using 0.72 eV instead of 0.63 eV. It impacts the model by reducing the resonance intensity by a factor of 32.4. Picoli et. al. recognized the problem, but fail to offer an explanation except that it is convenient to fit the data. Together, both problems act to reduce the predicted optimum intensity by a factor of 4.9. The problems were not imported into the model used in the thesis. Neither problem was found to be necessary to fit the experimental data to the resonance model used in the thesis. The two apparently bogus numbers used by Picoli et. al. as constants appear to be a way to fit their data to theory.

The model predicts an illumination induced rise in the relative ionic concentration $[\text{Fe}^{+3}]/[\text{Fe}]$. The rise is a cause of concern. The absorption constant increases with $[\text{Fe}^{+3}]$ because σ_p^o (due to Fe^{+3}) $= 194\sigma_a^o$ (due to Fe^{+2}). When the relative $[\text{Fe}^{+3}]/[\text{Fe}]$ concentration rises from 0.08 to 0.54, the optical absorption should increase about six times, from $\approx 1 \text{ cm}^{-1}$ to $\approx 6 \text{ cm}^{-1}$. Such a loss should reduce or eliminate any photorefractive gain. However, laboratory observation indicates that for whatever reason, such is not the case. Under the conditions of temperature-intensity resonance predicted by the Picoli model, when the pump is added to the signal beam, the signal power increases dramatically as the two beams are coupled. The absorptive loss does not appear to increase, unless the photorefractive gain is on the order of 12 cm^{-1} . That would result in the observed maximum gain of $\approx 7 \text{ cm}^{-1}$.

The temperature-intensity resonance cannot be satisfied throughout the volume of the crystal because the pump intensity falls due to optical absorption. One way to satisfy the resonance condition throughout the volume is to decrease the crystal temperature along the path of the pump. In this manner, as the pump intensity falls, the required pump intensity for maximizing Γ also falls. The entire path can be made to deliver the optimum 2WM gain for the available pump power. The entire crystal would produce maximum 2WM gain, and it has not been implemented to the best available knowledge.

Another way to satisfy the resonance condition is to tailor the absorption in the crystal by increasing α along the path of the pump. It can be done by illuminating the crystal with incoherent light propagating backwards along the path of the pump and the signal. The light causes Fe^{+2} to be converted into Fe^{+3} . The phenomenon was observed in the thesis in the difference between $[\text{Fe}^{+2}]/[\text{Fe}]$ when the crystal is in the dark to when it is illuminated under 2WM conditions. The backwards illumination causes more $[\text{Fe}^{+3}]$ at the end of the pump than at the beginning of the pump's path in the crystal. The pump would experience larger absorption constant the deeper it travels in the crystal, since σ_p^o (due to Fe^{+3}) $\approx 194\sigma_n^o$ (due to Fe^{+2}). The volume rate of pump optical energy absorption through the crystal becomes more equal because α is least when the pump is at maximum intensity. α increases as the pump is absorbed through the crystal. Equalizing the optical energy absorption rate per unit volume causes the rate of hole photoexcitation to become equal along the length of the crystal. Assuming uniform temperature, the new steady state satisfies the T-I₀ resonance throughout all of the length of the crystal and produces an optimized amplifier.

Satisfying the resonance condition gives a potential reward of 80 dB/cm ($\approx 20 \text{ cm}^{-1}$) gain, potentially making a half centimeter long crystal of InP:Fe into a 40 dB amplifier block. A millimeter of crystal would be a 10 dB gain block. Many such small devices could be integrated into hybrid photonic integrated circuits, for example, to boost the output of many lossy devices.

An effort was made to observe a similar intensity-temperature resonance in GaAs:Cr under identical conditions. No gain enhancement was observed in the nominally similar III-V semiconductor. The distinct performance is due to different physics in GaAs:Cr - i.e., the band structure is different. Therefore σ_n^o , σ_p^o , e_n^{th} , and are all different. The balancing of the thermal electron rate against the optical hole rate in GaAs:Cr is washed out by too many optical electrons.

CHAPTER 7

APPLICATIONS

This chapter considers applications of the photorefractive effect. The photorefractive effect in semiconductors is a relatively new and unexploited phenomenon. Photorefractive two-wave mixing has been explained theoretically in Chapters 2, 3, and 4, studied with a finite element analysis in Chapter 5, and observed experimentally in Chapter 6. Chapter 7 now endeavors to list some practical applications of photorefractive devices in telecommunications and in optical computing. Some examples are fast nondestructive III-V semiconductor wafer characterization, low level optical detection, image processing, optical novelty filters, optical computing, and beam cleanup using phase conjugation. Gunter and Huignard surveyed many of these applications in 1989 [143]. The most immediate uses are as low cost optical amplifier devices for telco services in long distance, CATV, and LANs. For example, the HDTV distribution system needs amplifiers for secondary and tertiary neighbourhood services distribution hubs. This market may become significant as CATV converts to optical fiber and when or if HDTV becomes the standard CATV medium. All these signal distribution networks based on optical fiber need the optical in-line amplifier to boost the received signal.

7.1 Telecommunications

7.1.1 Signal Amplification

Photorefractive crystals may be used as signal amplifiers in fiber optics communications systems. The possibility of amplifying signals with data rates significantly faster than implied by the typically one millisecond photorefractive response time was first recognized by Hamel de Montchenault et. al. in 1987 [144]. The first observation of AM signal amplification was by Bylsma et. al. in 1988 in a 0.9 cm long InP:Fe sample [145]. 13 dB gain and 11 dB gain were observed on a 4 kHz square wave signal at $\lambda = 1.06 \mu\text{m}$ and $1.3 \mu\text{m}$ respectively. The crystal response frequency was 0.3 kHz for 10 kV/cm bias. In 1990 Dolfi et. al. reported a two channel photorefractive amplifier in BaTiO_3 amplifying signals at 1.6 and 2.2 GHz [146]. To date, the ultimate bandwidth of the photorefractive amplifier has not been reached and the state of the art is four orders of magnitude away from approaching the high frequency limit.

7.1.1.1 Bandwidth

The ultimate optical bandwidth of photorefractive 2WM amplifiers has not been experimentally measured. For mutually coherent lasers, once the photorefractive two-beam coupling has been set up, the signal bandwidth only depends on the speed of the Pockels effect. As detailed in Section 4.1.2.4, Pockels effect takes about ≈ 0.12 psec to set up, limited by fundamental transverse optical lattice vibration frequency. This corresponds to about 8 THz bandwidth, or three percent of the 300 THz optical carrier frequency. This is more than adequate for most fiber amplifier applications.

Although mutual coherence is taken for granted in 2WM, the coherence of the mixing waves is an important limit when considering the maximum pulse throughput. This is because modulation on the optical carrier causes the optical spectrum to spread out. For ASK signals, the modulation consideration results in a quasi-coherence constraint. The pulse frequency must be considerably less than the optical frequency, which is ≈ 300 THz.

Mutual coherence due to spectral spreading is less significant than the Pockels response limit. Therefore one may assume that 2WM gain will fall off at around 8 THz band to Pockels effect alone. This appears to be the fundamental limit of photorefractive amplifiers. It has not been discussed in the available literature.

7.1.1.2 Noise Figure

A 2WM amplifier is less noisy than any semi-lasing element such as a double-heterostructure injection gain medium, an ionized gas, or erbium-doped fibers. The amplifiers exhibit an intrinsically low noise figure due to operation as a parametric amplifier [147]. The noise figure is theoretically 0 dB, but practically it must be somewhat increased due to insertion loss and internal optical scattering. The photorefractive gain is the result of the action of a degenerate parametric amplifier [148]. This happens since the grating with spacing Λ acts as an idler wave inside the crystal to transfer the energy from the pump to the signal. The energy exchange is governed by the Manley-Rowe energy relations, where the original derivation [149] is changed to account for the optical wave vectors.

Equivalently, one may explain that gain with low noise is the result of

stimulating bulk PRE instead of quantum mechanical spontaneous emission. 2WM is a stimulated bulk material process as opposed to the noisy spontaneous quantum mechanical transitions that characterize other laser-based amplifiers.

7.1.1.3 Comparison with Photochromic Devices

Photochromic gratings may exist in the same material as photorefractive gratings. However, they only diffract pump energy into the signal. There is no net addition to the signal power. Therefore photochromic effects should be avoided when building a gain device.

7.1.2 Repeater Gain Block

Optical repeaters are becoming more important as high speed fiber optics transmission spreads from just long haul applications into LANs. To compensate for intrinsic fiber losses and coupling losses to customers, the signal needs to be boosted whenever it falls below a reliable detection level. The function is performed with fiber optic repeaters.

Telecommunication fiber optic repeaters may be implemented in bulk semiconductors using PR amplifiers as shown in Figure 7.1. 20 dB is readily achievable in one centimeter of photorefractive crystal. This implementation is better than conventional

Figure 7.1 - All optical photorefractive in-line fiber optic repeater.

electric repeaters because it is an all optical implementation: it is simpler and it has a wider bandwidth. The photorefractive repeater is better than erbium-doped fiber optical repeaters because it is smaller, it may be integrated with optoelectronic ICs, and it introduces no spontaneous noise. The disadvantages are the high voltage dc bias, polarization sensitivity and the requirement for a frequency tracking LO pump laser. The frequency tracking is needed to provide a quasi-coherent pump to the bulk crystal.

7.1.3 Multichannel Optical Amplifier

As demonstrated in this thesis, PR amplifiers allow one to amplify one signal with a coherent pump beam. Gilbreath showed three beam mixing in bulk BaTiO₃ where the pump beam amplifies two signal beams simultaneously [150]. By extension, it is possible to amplify many AM or FM signals with a quasi-coherent pump beam in a single bulk crystal. This cost effective application is depicted in Figure 7.2. The device may be integrated into a two dimensional waveguide on some suitable substrate for a photonic integrated circuit.

7.1.4 Low Level Optical Detection

The 2WM process may be configured so that as well as amplifying the signal,

Figure 7.2 - Multichannel photorefractive in-line optical fiber amplifier using a quasi-coherent pump.

part of the pump is diffracted into the signal. The operation integrates an amplification function with a beam combination function in a heterodyne receiver. The result is a more sensitive, simpler, cheaper optical heterodyne receiver [151]. Boothroyd et. al. discuss coherent detection with photorefractive GaAs:Cr, and the need for matching the polarization angles of pump and signal beams to index ellipsoid [152].

7.1.5 NxN Multichannel Coupler

In a typical NxN multichannel coupler, signal losses are at least $20\log_{10}N$ decibels. That is, for a 16x16 coupler, the insertion loss is greater than 14 dB. This large loss can be eliminated in radiation type couplers by filling the propagation volume with a photorefractive medium such as InP and pumping the crystal. If the pump is quasi-coherent with the signals, then the signal power in each of the N channels is increased. The quasi-coherency condition is satisfied at the output of a central switching station and in a bundle derived from the same optical reference frequency. Over a one centimeter crystal interaction length, the power of each channel signal may be increased 20 dB or more. One foresees the possibility of almost lossless NxN couplers.

7.2 Optical Computing

7.2.1 Optical Memory

7.2.1.1 Optical EPROM

An all-optical erasable EPROM (Erasable Programmable Read Only Memory) may be fabricated based on a directional coupler scheme originally designed in LiNbO₃ with a control dielectric waveguide between two signal guides as suggested by Zang and Ristic at the University of Toronto [153]. Aronson and Hesselink used a different geometry in LiNbO₃ to fabricate a 50x50 array of photorefractive integrated-optical switches in an active area of 5x5 mm [154]. If one switched from LiNbO₃ to semi-insulating InP, one could construct similar photorefractive structures. This could act as a useful optical PROM device for optical time division switching on a single InP substrate. Resulting devices would have switching speeds on the order of one microsecond. They would also be optically controlled, and optically read out.

Optical computing using photorefractive GaAs and InP promise direct optical digital processing of telecom λ s 1.3 and 1.5 μ m. Since the photorefractive effect can operate

in the preferred telecom λ s, i/o is simpler and faster because no optical to electrical conversion is required on received signals, and no electrical to optical conversion is required on transmission.

7.2.1.2 Optical Mass Storage

In addition to optical RAM, an optical computer requires optically accessible mass storage. One possibility is the prototype "holostore" device manufactured by Microelectronics and Computer Technology Corp. (MCC) of Austin, Texas. This hard disk replacement is the first commercial application of the photorefractive process [155], and is designed for current electrical computers. Holostore uses packed arrays of hundreds of tiny crystal columns of cerium-doped strontium barium niobate ($\text{SrBaNbO}_3\text{:Ce}$) to photorefractively record data in eight Kbyte pages. MCC uses a patented nondestructive read technique to perform <10 microseconds read and <100 microseconds write times per page. This is about one thousand times faster than hard disk performance, and the prototype 1 gigabyte storage capacity is about ten times larger than the usual hard disks that fit into the same $5\frac{1}{4}$ inch form factor in one's PC. The potential for 100 gigabyte removable modules with even better access time points to the photorefractive effect meeting demands for more high bandwidth, nonvolatile storage.

7.2.2 Image Processing

There are a number of demonstrated image processing applications for 2WM. In 1988, Feinberg and MacDonald report using photorefractive phase conjugation for $1\ \mu\text{m}$ photolithography, image correlation, convolution, and subtraction, edge and defect enhancement, and associative memory [156]. For example, Figure 7.3 depicts a photorefractive image adder and subtractor. The phase conjugate mirror generates two retrodirected wavefronts, one for image A, and one for image B. The amplitude subtraction operation $A-B$ is performed by arranging for the retrodirected coherent beams to be out of phase across the wavefront. $A+B$ happens when the coherent beams are geometrically manipulated to be in phase.

In 1988, Huignard and Gunter describe photorefractive applications in photolithography, image storage, beam deflection, image convolution and correlation, edge enhancement, image subtraction, and parallel optical logic [157]. For example,

Figure 7.3 Image adder and subtractor. After Feinberg [157].

Figure 7.4 Image correlation using 2WM. After Huignard [134].

Figure 7.4 depicts 2WM performing image correlation of image B against image A. Images A and B are focused into the photorefractive crystal in 2WM geometry. The interference hologram recorded in the crystal is the convolution product of the optical fields when viewed in the Fourier plane of the exit lens. A reference beam at a longer wavelength λ_R is used to read the phase grating, and the result $A \times B$ is shown. The filter is used to pass λ_R and to eliminate the image wavelength from the output.

P. Yeh et. al. published a review paper on photorefractive optical computing in 1989 describing image amplification, image subtraction, optical interconnects, matrix-vector, and matrix-matrix multiplication [158]. These applications involve both 2WM and the closely related four-wave mixing.

Cheng reports 2WM based devices in semi-insulating GaAs performing fast image convolution, and autocorrelation [159]. Undemonstrated but possible developments of the technology are fast programmable lenses and programmable beamsplitters [160].

The following sections briefly discuss image amplification, novelty filters, and wafer characterization.

7.2.2.1 Image Amplification

A pump beam will amplify an image beam inside a photorefractive medium the same way that the thesis demonstrates amplification of a single beam. In 1989, Yeh reports spatially uniform amplification in BaTiO_3 of 40 dB gray scale in a 40 dB image amplifier [161]. In 1990, Rajbenbach reports noise-free amplification with 34 dB gain in BaTiO_3 , where he achieved the noise-free image by rotating the crystal at 5 millidegrees/sec [162]. Because the noise sources are defects - bonded to the lattice - rotating the crystal washes out the photorefractive effect associated with these sources.

7.2.2.2 Novelty and Monotony Filters

A novelty filter filters out parts of an image that are unchanging and only transmits new items in the image [163]. It works by temporal high-pass filtering using the time constant of the photorefractive effect in the crystal. A novelty filter using 2WM is depicted in Figure 7.5. The optical novelty filter uses a real-time holographic storage medium. The medium records the time exponential average of the scene. The scene is impressed on the pump as a two dimensional amplitude, phase or polarization image. A change in the pump beam results in energy transfer to the second beam to be momentarily defeated, and the new feature in the pump is transmitted. Soon, however, photorefractive energy transfer begins again and the new feature is no longer transmitted since it is coupled into the signal. In this way, interference simultaneously subtracts the stored scene from the

live scene, giving peak contrasts of up to thirty to one.

The optical novelty filter is simple to build and is more cost effective than conventional implementations of novelty filters which employ modified liquid crystal television displays. It has several uses. For example, in biomedical research, a novelty filter can be placed at an image plane in a microscope to filter out a nonbirefringent stationary background and show only moving cells. The military can use novelty filters as motion indicators in battlefield imaging systems. A potent electronics warfare use is integration of a novelty filter as a second channel in a Bragg cell spectrum analyzer. The filtered channel would eliminate noisy CW and chopped CW signals to only amplify and measure radar pulse widths between fifty nanoseconds and one microsecond.

The optical monotony filter is the complement to the novelty filter. The 2WM based novelty filters described above all have both a novelty and a monotony port, corresponding to the two wave fronts coming out of the crystal. The monotony port uses the photorefractive effect to filter time varying parts of an image and the port displays only the constant components of a scene. The port is a temporal low-pass filter based on the time it takes for the photorefractive effect to set up. This response time can be arbitrarily set by adjusting the optical pump intensity, the bias voltage or the crystal temperature. For lower pump intensity, higher bias voltage, or closer to optimum gain Γ crystal temperature, the crystal has a longer time constant τ . The transmitted image changes more slowly new

Figure 7.5 Optical novelty filter using 2WM. After Anderson and Feinberg [164].

features in the input image. Therefore, only things that have been still for at least time τ appear in the output image on the monotony channel.

7.2.2.3 Nondestructive III-V Wafer Characterization

This work demonstrates how to use absorption spectra between $\lambda=900$ and 1600 nm to solve for deep level trap concentrations in small samples of III-V semiconductors. Others have used single λ absorption at room temperature to image the concentration of deep level traps such as Fe in InP:Fe, EL2 in GaAs, and EL2 and Cr in GaAs:Cr. This nondestructive test tells one the uniformity of the wafer, distribution of traps, and the resistivity of the material. If required, more information is available by taking absorption spectra at 77°K. This is because the spectral lines of weaker transitions are invisible at room temperature, while at lower temperatures, they become clear.

While absorption imaging yields trap density information, it does not yield the majority photocarrier. The photocarrier may be quickly determined using the photorefractive 2WM [164]. The direction of energy transfer is controlled by the majority photocarrier for a particular set of crystal axes. If the carrier is electrons, then energy goes one way and if it is holes, then energy goes the other way. This gives one a room temperature characterization of p or n type at one λ and Λ . In addition, 2WM delivers a second estimate of deep level trap concentration, and a sensitive indication of the transport properties of the crystal in the Debye length.

The absorption and 2WM tests are nondestructive and only require wafers that are polished on both sides. The tests determine material parameters and give fast go - no go data on wafer suitability for processing into digital and microwave chips. This increases the process yield since one may guarantee particular substrate parameters by imaging the parameters of each wafer. One is not limited by destructive and expensive batch sampling techniques such as scanning imaging mass spectroscopy ("SIMS"). Using these tests would save money for the semiconductor industry by reducing fabrication waste. It would increase chip reliability by verifying critical parameters of each wafer before fabrication. The main operational cost in the test is in polishing both sides of each wafer.

7.3 Summary

Two-wave mixing in InP:Fe have demonstrated direct optical signal processing applications wavelengths $1.3\ \mu\text{m}$ and $1.5\ \mu\text{m}$. Since the photorefractive effect can operate in these preferred wavelengths for telecommunications, operation is simpler and faster because no optical to electrical translations are required. The most promising near term applications are to amplifying wide band multichannel optical telecommunications signals, and in analog image processing. Longer term application is in devices for all optical digital computers. Computers based completely on photorefractive devices described here are not likely due to the relatively slow one channel switching speed.

CHAPTER 8

SUMMARY AND CONCLUSIONS

Chapter 1 introduced the reader to the photorefractive effect for III-V semiconductors in general, and for InP:Fe, in particular. The concept of optical gain in a photorefractive crystal was presented followed by a brief discussion of the history of the research. **Chapter 2** described the Kukhtarev formulation and the electro-optic effect as it applies to the photorefractive effect. **Chapter 3** explained the sub-band gap optical absorption mechanism in InP:Fe. **Chapter 4** presented theoretical derivations of the response time and the ultimate optical gain of the photorefractive effect. **Chapter 5** numerically modeled the photorefractive effect in InP:Fe using the finite element method. **Chapter 6** presented experimental laboratory work and demonstrated photorefractive gain up to 7 cm^{-1} , corresponding to a 15 dB net gain parametric optical amplifier, theoretically with a scattering limited zero dB plus insertion loss noise figure. **Chapter 7** explained some of the possible unexploited commercial applications of photorefractive in semiconductors.

8.1 Summary

SI InP:Fe was characterized by evaluating $[\text{Fe}]$, $[\text{Fe}^{+2}]$, and $[\text{Fe}^{+3}]$ using absorption spectra and a composite Lucovsky model for photoionization cross-section of holes and electrons. $[\text{Fe}]$ was $3.7 \times 10^{16} \text{ cm}^{-3}$, and $[\text{Fe}^{+2}]/[\text{Fe}]$ was 0.92 in the dark. A temperature intensity resonance was observed at 289°K and $I_0 = 4 \text{ mW/cm}^2$. The resonance indicated that $[\text{Fe}^{+2}]$ and $[\text{Fe}^{+3}]$ changed under 2WM at $\lambda = 1.064 \text{ }\mu\text{m}$ so that $[\text{Fe}^{+2}]/[\text{Fe}] = 0.46$. The change shows that the laser beam changes the charge species breakdown. Fe^{+2} was reduced to Fe^{+3} . At the resonance, $[\text{Fe}^{+2}] = [\text{Fe}^{+3}]$. The rough equality is regarded as the condition for optimum 2WM because the laser beam continuously replenishes the population of Fe^{+2} as thermal excitation replenishes the population of Fe^{+3} . The balance between thermal electrons and photo-excited holes sets up a steady state for maximum 2WM gain.

The maximum small signal optical gain for $\lambda = 1.064 \text{ }\mu\text{m}$ was $\Gamma = 7 \text{ cm}^{-1}$ at $\alpha = 0.93 \text{ cm}^{-1}$, $\Lambda = 12.3 \text{ }\mu\text{m}$, and $E_0 = 12.5 \text{ kV/cm}$. It corresponds to periodic grating fields of at least 21.6 kV/cm . The real net gain of the AR coated InP:Fe crystal was 15 dB. The result is representative of others' work in InP:Fe. The observed gain was modeled

analytically using the estimate for [Fe].

Numerical modeling using the program PISCES was used to simulate the formation of a photorefractive space charge field. The model produced a maximum $\pi/2$ shifted electric field amplitude of 54 kV/cm, corresponding to gain $\Gamma = 15.2 \text{ cm}^{-1}$. Fields on the order of 20 kV/cm were simulated, and the model's behaviour was studied as a function of [Fe], external bias, depth of modulation, temperature, optical intensity, and grating spacing. Both the simulated electric field amplitudes and response times resembled measured laboratory data, even though PISCES does not implement optical absorption at deep level sites. Approximations were made to make PISCES approximate the observed data.

The time constant required to set up large gains was modeled and measured. An increase in the time constant was observed as the gain increases. Increasing gain from 0.1 to 7 cm^{-1} was accompanied by the lengthening of the time constant from 1 to 100 msec. The longer time constant is due to the extra time required to generate enough carriers to form the necessary larger charge clouds. The numerical modeling with PISCES indicated the same order of magnitude of time constants.

8.2 Conclusions

There were four contributions to theory. First, room temperature optical cross-section spectra of $[\text{Fe}^{+2}]$ and $[\text{Fe}^{+3}]$ were predicted using published data for 77°K and theoretical multi-band cross-section models. Second, quantitative investigation of the source of deep level optical absorption in InP:Fe was performed. Third, the concentration of iron in InP:Fe was measured using optical absorption. Iron was resolved as two charge species Fe^{+2} and Fe^{+3} using room temperature sub-band gap absorption spectra and $[\text{Fe}^{+2}]$ and $[\text{Fe}^{+3}]$ cross-section spectra. Fourth, PISCES finite element modelling was performed to simulate the photorefractive effect. The dynamic formation of photorefractive space charge electric field was observed with steady-state amplitudes greater than 20 kV/cm (gain = 6 cm^{-1}).

Building single-mode fiber-optic arms into the two-wave mixing apparatus was a practical contribution to experimental technique because it simplifies set-up. The fiber

technique makes two-wave mixing compatible with integrated optics applications.

The field of photorefractive 2WM in III-V semiconductors is important to telecommunications and optical computing since these devices may someday find practical roles outside the laboratory. A very low noise photorefractive amplifier for telecommunications is one useful building block from a long list of new applications.

The amplifier can be built in bulk InP:Fe or else in an integrated epitaxial InP waveguide. Light can be coupled in and out on optical fibers. The bandwidth of these amplifiers is estimated to be ≈ 8 THz, and gains of 40 dB per centimeter of crystal have been achieved by other workers.

Although the study of two-wave mixing amplifiers in III-V semiconductors only began in 1984, many papers have been published on the subject. Interest originally focused on GaAs:EL2 and GaAs:Cr, but recently InP:Fe has become the medium of choice due to higher experimental and predicted gains. In the future, other media such as CdTe:V and undoped GaP may become available, offering new wavelengths of operation. GaP is especially interesting because it is photorefractive in the visible range. GaP has advantages in that it exhibits less intrinsic absorption, in that it is mechanically more rugged than InP:Fe and GaAs, and in that it exhibits photorefraction between wavelengths of $\lambda = 600$ nm and $\lambda = 900$ nm. This provides a medium for work with 800 nm wavelength laser diodes.

The 1979 Kukhtarev model still describes most of the observed photorefractive gain and the dynamic time variations observed in the experiments.

Characterizing InP:Fe with room temperature optical absorption spectra between wavelengths 900 nm and 1600 nm provides immediately useful qualitative information, such as approximate location of the band gap. However, the spectra are difficult to interpret in terms of device parameters. In particular, the derivation of iron doping level in InP from absorption spectra is not a well known technique. Most workers use a mass spectroscopy technique to find the iron concentration even though that technique is destructive and it offers no insight into the electronic state of the iron sites. The absorption spectrum method is preferred, but the process is complicated by the two iron

charge species in the band gap, each species with an independent photoionization cross-section that varies as a function of temperature.

8.3 Directions for Future Research

8.3.1 General Ideas

In a 1990 study, Zielinger and Tapiero stated [165] that any major progress towards industrial application of photorefractive InP:Fe, GaAs, and other III-V semiconductors is dependent on fulfilling three interdependent research thrusts:

- (1) Crystal growth and processing.
- (2) Systematic physical and chemical characterization.
- (3) Investigation of photorefractive properties and nonlinear experiments.

Thrust (2) - the systematic characterization of semi-insulating InP and GaAs is the most difficult research thrust due to the high resistivity of the semi-insulating media. The problem is that classical semiconductor techniques are based on either strong n type doping or else strong p type doping, with the resulting abundant mobile charge carriers. This de facto assumption severely limits one's ability to characterize materials when there are almost no carriers, as in the case of SI InP:Fe studied in this thesis. For example, classical Hall mobility measurements¹³ are impractical and almost impossible to perform due to the high voltage required to produce any transverse voltage at all greater than Johnson noise through the semiconductor.

Although classical methods may not work well in characterizing SI III-V material parameters, new techniques are available. Absorption spectra, photoconductivity spectra, and emission spectra are the most common tools. Magnetic circular dichroism is not used much but it is well suited to the study of high resistivity media.

8.3.2 Specific Experiments

This section discusses specific follow-on experiments that would extend the body of knowledge on photorefractive effects.

¹³ van der Pauw geometry was used in Hall measurements at 77°K and 300°K.

8.3.2.1 Absorption Spectra under Illumination

It was observed that under dark conditions, Fe^{+2} formed 29% of the total available Fe, while under optimum 2WM conditions, it formed 57%. To characterize the shift in ionic concentration, one could perform absorption spectra measurements while the InP:Fe sample is under uniform illumination with $1.064\ \mu\text{m}$ light. By varying the intensity between 0.5 and $1000\ \text{mW/cm}^2$, one expects to observe change in the spectral shape corresponding to changes in the relative concentrations of Fe^{+2} and Fe^{+3} . Two critical factors must be considered when doing the experiment. First, scattered light from the laser will saturate the detector if it is not eliminated at the detector entrance pupil. This requires two 60 dB band stop notch filters (one for the reference and one for the sample channel in the spectrophotometer) centered at $\lambda=1.064$ and less than 20 nm wide. Second, the temperature of the sample must be closely controlled because it influences the significant rate of thermal electron generation. The laser illumination is the source of most thermal problems. $1000\ \text{mW/cm}^2$ causes temperature rise of 2.0°C per second. Temperature stabilized absorption spectra results would be useful in modeling 2WM performance.

8.3.2.2 Mobility in SI InP:Fe

Carrier mobility is important in modeling 2WM in InP:Fe. Measuring it in SI materials poses a problem. The resistivity of the samples is high enough that even with 50 volts and one microamp current sources, the Hall voltage at the sample points fluctuates with what appears to be Johnson noise even when using ohmic contacts diffused into the InP:Fe sample¹⁴. The solution is to uniformly illuminate the sample with intensities between 10 and $100\ \text{mW/cm}^2$ at $\lambda=1.064\ \mu\text{m}$. This would reduce the resistance by at least an order of magnitude, and would provide noise-free Hall mobility measurements. Again, absorptive heating is a problem because it causes the crystal to heat up and changes the mobility results. To eliminate the problem, use a TE cooler bonded to all four sides of the crystal sample or else a liquid nitrogen bath and deliver the laser to the sample using a fiber bundle.

¹⁴ Using van der Pauw geometry and three $5\times 5\times 1\ \text{mm}$ samples from the edge of the same wafer as was used to cut the 2WM sample.

8.3.2.3 Debye Length Measurement

Debye length measurements reveal the mean free diffusion distance in the crystal. To measure it using 2WM, one must provide at least $\approx 1000 \text{ mW/cm}^2$ in order to saturate the crystal by ionizing all of the available iron centers. Temperature control must be used in this experiment because of previously described absorptive heating effects.

8.3.2.4 Other Directions

Based on Zielinger and Tapiero's statement, there are many interesting directions in which to proceed, and some which may not be worth while. In practical terms, one can envision the local research institute carrying out physical characterization and photorefractive experiments, but not special dedicated crystal growth experiments. Construction of an automated photorefractive telecommunications test bed at $\lambda = 1.3 \text{ }\mu\text{m}$ with a one meter coherence length, single mode fiber optics, a 20 GHz AM modulator, motor-driven stages, a dedicated digital oscilloscope, and a modern spectrum analyzer with a YIG preselector is a good place to start characterization and experimental work. Such a test bed permits repeatable physical characterizations and the ability to quickly investigate various photorefractive properties. Fast consistent results from the outlined set up are particularly important for today's results-driven laboratory programs. There is no leisure to pursue experimental details and related but unfunded side aspects of material behaviour, even though they may have some long term academic merit. In particular, serious photorefractive work requires computer automation of photorefractive gain versus grating spacing data acquisition. The test bed should standardize on one particular crystal size such as $5 \times 10 \times 10 \text{ mm}$ to facilitate the automation. When not working with integrated optics, it is important to route the photorefractive arms through closed tubes to stop random air fluctuations from disrupting the wavefronts and the interference pattern in the medium. This is especially important for experiments with high optical gain and the attendant long set-up time constants.

Many useful tests have not been performed on photorefractive amplifiers. For example, noise figure, bandwidth, and intermodulation products have not been experimentally addressed. A test should be set-up to measure the bandwidth of the photorefractive amplifier, looking at optical fiber signals AM modulated out to at least to

20 GHz. As explained in **Section 7.1.1.1**, one expects the 3 dB bandwidth of a photorefractive gain element to be about 8 THz.

From the test bed, the most important parameters about what is needed in a photorefractive integrated optic device could be tested. This could lead to production and testing of an integrated photorefractive amplifier.

A follow-on integrated optics application is building an optical EPROM as described in **Section 7.2.1.1**. Even though a EPROM is a more complicated fabrication, any demonstration of an EPROM would be a significant accomplishment.

8.3.2.5 Iron Concentration Verification

The absorption spectrum based iron concentration measurement performed in **Chapter 3** did not have any independent measurement to confirm that the iron parameters extracted from the absorption spectrum were true. A series of three extra experiments need to be performed to verify the measurement. The first experiment is an electronic spin resonance (ESR) test to measure the Fe^{+3} ionic content. The second experiment is a photoluminescence test near $\lambda=3 \mu\text{m}$ or else an acoustic paramagnetic resonance (APR) test to measure the Fe^{+2} ionic content. The third experiment is mass spectroscopy to determine the total iron concentration. The third experiment must be performed last of all because it destroys the InP:Fe crystal sample.

8.3.2.6 Telecommunications Amplifier

The photorefractive amplifier has been tested by others up to two gigahertz. It is theoretically capable of amplifying amplitude modulated signals up to eight terahertz. The gain and the noise figure response of the amplifier should be measured up to twenty gigahertz, using standard single mode telecommunications equipment at $\lambda=1.32 \mu\text{m}$ up to ten gigabits per second. The response to 20 GHz can be tested using fast single mode fiber modulators and wide band low power microwave equipment. The response at millimeter wave frequencies i.e., 30 to 40 GHz, could be tested using experimental single mode fiber modulators. Testing at 60 GHz, 100 GHz, and beyond may have to wait for the development of the next generation(s) of single mode fiber modulators.

8.3.3 Numerical Modeling

The thesis demonstrates application of the numerical modeling program PISCES to observe the growth and the steady state form of the optical absorption induced space charge electric field in InP:Fe. Future work could begin by examining the phase angle of the space charge electric field as a function of the applied external bias.

Further work should modify the modeling of deep levels with PISCES. A problem exists in the program in that PISCES does not implement optical absorption at deep level sites. Instead, PISCES simulates absorption by creating both a free electron and a hole. For the photorefractive effect, only one or the other is created. PISCES source code should be modified to allow for partial optical ionization of deep levels. The modification would divide the deep iron level into two deep level charge species Fe^{+3} and Fe^{+2} which transform into each other as a result of optical absorption events. The model would have different programmable optical cross-sections for hole ionization from Fe^{+3} and electron ionization from Fe^{+2} . The model would also have to allow for partial thermal ionization of trapped electrons in Fe^{+2} to the conduction band, for laser induced absorptive heating, and for ohmic heating caused by photoconductivity in the presence of external bias.

A second modification to PISCES would be to solve for the 90° shifted component of the space charge field. It would assist in the model of the coupling between signal and pump in the photorefractive effect. The modification would divide the crystal into one hundred slices, each slice $100\ \mu\text{m}$ thick. Together they would comprise a one centimeter thick crystal. Each slice would consist of a mesh of finite elements as described in Chapter 5. The modified PISCES would, at each simulation time, calculate the energy transfer due to the coupled wave equations in one slice. The information would be passed on to the next slice. The energy transfer would be calculated again in each subsequent slice, based on the constant transferred energy (gain Γ) and the constant absorptive loss (a function of the deep levels) over the $100\ \mu\text{m}$ slice thickness. The slice calculations would go on until all 100 slices were considered for time t . Then the model would go on to time $t+1$. And so on, until steady state is reached (nominally at one second).

The first modification should allow simulation to observe the temperature-intensity resonance in InP:Fe. The resonance was observed and modeled analytically in the

thesis but the effect was very weak in the numerical PISCES modeling. The weakness of the effect in PISCES is clearly attributable to the lack of modeling photoionization processes in deep levels and deep level trap occupancy. Generally, accurate numerical modeling would allow planning of new experiments and explanation of results. The PISCES model would be useful in designing integrated optics waveguide devices such as MOSFETs with optical waveguides because of the ability in the program to build arbitrary structures with varying dopant levels and refractive indices. For example, a monolithic FET and a microwave transmission line could be added to a photorefracting device, and the interaction between the fields could be examined.

8.3.4 Coherence Requirement

The photorefractive effect takes some time to set up in a crystal. Even though the operational bandwidth of a photorefractive amplifier is ≈ 8 THz, the physical setting up of the photorefractive effect can take a few milliseconds to set up. Therefore, the intensity interference pattern must be reasonably stable for at least a few milliseconds. This is only achieved if the interfering beams are mutually coherent. If the two beams are incoherent with respect to each other, there is no PRE. In this work, the coherency requirement demands that the path lengths of the signal and the pump beams, derived from the same laser, must be equal within a fraction of the coherence length.

The allowable error in path length is proportional to the coherence length of the laser. Coherence lengths vary between microns and kilometers, depending on the laser. In this work, coherence length at $1.064 \mu\text{m}$ was two millimeters. It was a day long tedious job to mechanically align the 2WM path lengths when coherence length was so short. Other workers published similar observations even with much longer coherence lengths. Picoli et. al. found it "somewhat time-consuming" to set up photorefractive experiments when the coherence length was less than ten millimeters [166]. Gilbreath commented on the difficulty when the coherence length was fifty millimeters [167]. Scaling from Gilbreath's work in photorefractive BaTiO_3 , using the YAG in this thesis, the experiments in this thesis using the Nd:YAG at $\lambda = 1.064 \mu\text{m}$ can tolerate path length differences of three percent of the coherence length, or sixty microns. The idea of alignment with such small tolerances is impractical when dealing with hand positioned folded optical path lengths. Typically the experimental part of the thesis used path lengths of seventy five centimeters,

which according to the sixty micron maximum error, requires a positioning tolerance of 0.008 percent.

Apart from buying a better laser with a longer coherence length, there are three ways to get around the short coherence problem. The first way is to use an etalon to filter out longitudinal modes and to extend the coherence length of the YAG and the HeNe lasers. For example, Gilbreath used a temperature-stabilized etalon to extend coherence length from fifty millimeters to approximately one meter. As a result, she tolerated path length differences of a few centimeters, and beam coupling was unaffected by loss in coherence due to errors of a few centimeters.

The second way to obtain coherence from short coherence sources is to use a set of first order holographic gratings to diffract the beam into two beams and then use two lenses - one for collimating, and the other for focusing onto the crystal. One grating is needed for each value of Λ . This was used successfully by Picoli [168]. He found that the mechanical set-up time was greatly reduced. In addition, the photorefractive results were repeatable. Picoli's method is superior to Gibreath's method in that the holographic grating does not throw away the beam's modal power, while the etalon does.

The third way to obtain coherence was employed in the second experiment in the thesis. A single mode fiber optic directional coupler and fiber beam launchers on precision positioners were used to obtain path length equality and optical coherence in the crystals. It works reliably, the results are repeatable, but it is more tedious than Gilbreath's and Picoli's methods. It requires hours of fine adjustments in search of the coherence peak for every value of Λ .

8.3.5 Optimum [Fe] in InP:Fe

The thesis studies one InP:Fe crystal and as a result, there is no experimental data on the effect of [Fe] on optical gain. Other workers have considered the relationship between [Fe] and optical gain. In general, if you can live with the extra optical absorption loss, more $[\text{Fe}^{+3}]$ is better, limited only by the precipitation of iron in InP:Fe at $\approx 10^{17} \text{ cm}^{-3}$. The potential trouble with more iron is that the Fe^{+2} concentration may become much larger than the Fe^{+3} concentration. This increases the equivalent dark irradiance. The

increased number of thermal electrons means that more optical intensity is needed to reach the T-I₀ resonance condition. Picoli et. al. modeled the optimum concentration of iron for maximum gain and they found that the more iron given a fixed Fe⁺²/Fe⁺³ ratio, the more exponential gain. For 10¹⁶cm⁻³ ≤ [Fe] ≤ 10¹⁷cm⁻³, the optimum gain increases from 7 to 20 cm⁻¹ as

$$\Gamma \sim 7 + 14 \times \left(1 - e^{-\frac{[\text{Fe}] - 10^{16}}{3 \times 10^{16}}} \right) \quad (8.1)$$

The gain levels off around 20 cm⁻¹. The large gain is only obtained if [Fe⁺²]/[Fe⁺³] is correct: Picoli gives ≈0.43 [169]. The number is comparable to 0.84 found in the T-I₀ resonance **Section 6.3.4.1.4** for the InP:Fe crystal used in the thesis.

Godefray et. al. reported that holographic efficiency, optical sensitivity, and electro-optic coefficients are maximized for 0.075 percent iron in the melt [170]. The influence of iron is larger on the electro-optic coefficient than on the space charge electric field. No optimum iron concentration has been observed in a systematic study of photorefractive InP or in any other III-V semiconductors. Strait suggests that for maximum photorefractive gain at λ=1.3 μm and zero external bias, one should minimize iron concentration, while retaining semi-insulating crystal characteristics.

Chapter 9
REFERENCES

1. P. Gunter, and J.P. Huignard, "Photorefractive Effects and Materials". in Photorefractive Materials and their Applications I: Fundamental Phenomena, edited by P. Gunter and J.P. Huignard, Springer Verlag, page 7, 1988.
2. D.M. Pepper, J. Feinberg, and N.V. Kukhtarev, "The Photorefractive Effect", Scientific American, page 64, 1990.
3. J. Feinberg, "Phase conjugation with nonlinear crystals", Lasers and Optronics, page 57, 1990.
4. A. Yariv, P. Yeh, "Optical Waves in Crystals", J. Wiley, page 224, 1984.
5. P. Yeh, A.E. Chiou, J. Hong, P. Beckwith, T. Chang, M. Khoshnevisan, "Photorefractive nonlinear optics and optical computing", Optical Engineering, 28, page 328, 1989.
6. J. Strait, J.D. Reed, G.C. Valley, M.B. Klein, "Net Gain in Photorefractive InP:Fe at $\lambda = 1.32 \mu\text{m}$ without an Applied Field", Applied Physics Letters, 57, page 951, 1990.
7. Y. Kawata, S. Kawata, and S. Minami, "Locally controllable image amplification by two-wave coupling with a BSO crystal", Japanese Journal of Applied Physics, Part 2: Letters, 29, page 1547, 1990.
8. A. Ashkin, G.D. Boyd, J.M. Dziedzic, R.G. Smith, A.A. Ballman, J.J. Levinstein, and K. Nassau, "Optically induced refractive index inhomogeneities in LiNbO_3 and LiTaO_3 ", Applied Physics Letters, 9, page 72, 1966.
9. F.S. Chen, "Optically induced change of refractive indices in LiNbO_3 and LiTaO_3 ", Journal of Applied Physics, 40, page 3389, 1969.
10. V.L. Vinetskii, N.V. Kukhtarev, V.B. Markov, S.F. Odulov, and M.S. Soskin, Abstracts of Reports, First Soviet-Japanese Symposium on Ferro-electrics, Novosibirsk, 1976.
11. N.V. Kukhtarev, "Kinetics of hologram recording and erasure in electro-optic crystals", Soviet Technical Physics Letters, 2, page 438, 1976.
12. N.V. Kukhtarev, V.B. Markov, S.G. Odulov, M.S. Soskin, and V.L. Vinetskii, "Holographic storage in electrooptic crystals. I. Steady state", Ferroelectrics, 22, page 949, 1979.
13. N.V. Kukhtarev, V.B. Markov, S.G. Odulov, M.S. Soskin, and V.L. Vinetskii, "Holographic storage in electrooptic crystals. II. Beam coupling - light amplification", Ferroelectrics, 22, page 961, 1979.

14. G.C. Valley, and J.F. Lam, "Theory of Photorefractive Effects in Electro-optic Crystals", in Photorefractive Materials and their Applications I: Fundamental Phenomena, edited by P. Gunter and J.P. Huignard, Springer Verlag, page 77, 1988.
15. A. Ashkin, B. Tell, J.M. Dziedzic, "Laser induced refractive index inhomogeneities and absorption saturation effects in CdS", IEEE Journal of Quantum Electronics, 3, page 400, 1967.
16. A.M. Glass, A.M. Johnson, D.H. Olson, W. Simpson, A.A. Ballman, "Four-wave mixing in semi-insulating InP and GaAs", Appl. Phys. Lett., 44, page 948, 1984.
17. M.B. Klein, "Beam coupling in undoped GaAs at 1.06 μm using the photorefractive effect", Optics Letters, 9, page 350, 1984.
18. A. Partovi, J. Millerd, E.M. Garmire, M. Ziari, W.H. Steier, S.B. Trivedi, and M.B. Klein, "Photorefractivity at 1.5 μm in CdTe:V", Appl. Phys. Lett., 57, page 846, 1990
19. K. Kuroda, Y. Okazaki, T. Shimura, H. Okamura, M. Chihara, M. Itoh, and I. Ogura, "Photorefractive effect in GaP", Optics Letters, 15, page 1197, 1990.
20. M.B. Klein, "Beam coupling in undoped GaAs at 1.06 μm using the photorefractive effect", Optics Letters, 9, page 350, 1984.
21. K. Walsh, T.J. Hall, "Influence of polarization state and absorption gratings on photorefractive two-wave mixing in GaAs", Optics Letters, 12, page 1026, 1987.
22. J. Kumar, G. Albanese, W.H. Steier, and M. Ziari, "Enhanced two-beam mixing gain in photorefractive GaAs using alternating electric fields", Optics Letters, 12, page 120, 1987.
23. B. Imbert, H. Rajbenbach, S. Mallick, J.P. Herriau, and J.P. Huignard, "High photorefractive gain in two-beam coupling with moving fringes in GaAs:Cr crystals", Optics Letters, 13, page 327, 1988.
24. K. Walsh, and T.J. Hall, "Gain exceeding absorptive losses in photorefractive GaAs", Applied Optics, 28, page 16, 1989.
25. A.M. Glass, A.M. Johnson, D.H. Olson, W. Simpson, A.A. Ballman, "Four-wave mixing in semi-insulating InP and GaAs", Appl. Phys. Lett., 44, page 948, 1984.
26. G. Picoli, P. Gravey, C. Ozkul, and V. Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: A new mechanism of resonance", J. Appl. Phys, 66, page 3798, 1989.
27. C. Ozkul, G. Picoli, and N. Wolffer, "High gain coherent amplification in thermally stabilized InP:Fe crystals under dc fields", Applied Optics, 29 page 2711, 1990.

28. J.E. Millerd, S.D. Koehler, E.M. Garmire, A. Partovi, A.M. Glass, and M.B. Klein, "Photorefractive gain enhancement in InP:Fe using band-edge resonance and temperature stabilization", *Appl. Phys. Lett.* 57, page 2776-2778, 1990.
29. J. Strait, J.D. Reed, G.C. Valley, and M.B. Klein, "Net gain in photorefractive InP:Fe at $\lambda = 1.32$ μm without an applied field", *Applied Physics Letters*, 57, page 951, 1990.
30. A.M. Glass, J. Strait, "The Photorefractive Effect in Semiconductors", in Photorefractive Materials and their Applications I: Fundamental Phenomena, edited by P.Gunter and J.P. Huignard, Springer-Verlag, page 240, 1988.
31. C.D. Brandt, A.M. Hennel, T. Bryskiewicz, K.Y. Ko, L.M. Pawlosicz, and H.C. Gatos, "Electronic and optical properties of Ti-doped GaAs and InP; semi-insulating InP", *J. Appl. Phys.* 65, page 3459, 1989.
32. S.M. Sze, Physics of Semiconductor Devices, Second Edition, John Wiley, page 21, 1981.
33. H. Muller, R. Trommer, and M. Cardona, "Pressure dependence of the direct absorption edge of InP", *Physical Review B*, 21, page 4879, 1980.
34. M.B. Klein, "Beam coupling in undoped GaAs at 1.06 μm using the photorefractive effect", *Optics Letters*, 9, page 350, 1984.
35. F.P. Strohkendl, J.M.C. Jonathan, and R.W. Hellwarth, "Hole-electron competition in photorefractive gratings", *Optics Letters*, 11, 1986, page 312.
36. G. Picoli, P. Gravey, C. Ozkul, and V. Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: a new mechanism of resonance", *J. Appl. Phys.*, 66, 1989, page 3798.
37. F.P. Strohkendl, J.M.C. Jonathan, and R.W. Hellwarth, "Hole-electron competition in photorefractive gratings", *Optics Letters*, 11, 1986, page 312.
38. G. Picoli, P. Gravey, C. Ozkul, and V. Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: a new mechanism of resonance", *J. Appl. Phys.*, 66, 1989, page 3798.
39. JC Fabre, JMC Jonathan, G Roosen, "43m Photorefractive Materials in Energy Transfer Experiments", *Optics Communications*, 65, page 257, 1988.
40. A. Yariv, Quantum Electronics, Third Edition, John Wiley, 1989, page 90.
41. J Strait, JD Reed, NV Kukhtarev, "Orientational dependence of photorefractive two-beam coupling in InP:Fe", *Optics Letters*, 15, 1990, page 209.
42. M.J. Weber, Handbook of Laser Science and Technology, Volume IV Optical Materials: Part 2, CRC Press, page 287, 1986.

43. T.L. Brown, H. E. LeMay, Jr., Chemistry: the Central Science, Prentice-Hall, page 708, 1977.
44. R.K. Willardson, A.C. Beer, Indium Phosphide: Crystal Growth and Characterization, Academic Press, 1990, page 251.
45. O Madeling, M Schultz, H Weiss, "Numerical Data and Functional Relationships in Science and Technology", Group III: Crystal and Solid State Physics, Volume 17: Semiconductors, Subvolume a, Landolt-Bornstein, Springer-Verlag, 1982, page 287.
46. TL Brown, HE LeMay, Chemistry: the Central Science, Prentice-Hall, 1977, page 198.
47. S.T. Pantelides, Deep Centers in Semiconductors, Gordon and Breach, 1986, page 1 ff.
48. D.C. Look, "Model for Fe^{2+} intracenter-induced photoconductivity in InP:Fe ", Physical Review B, 20, page 4160, 1979.
49. P.R. Tapster, M.S. Skolnick, R.G. Humphreys, P.J. Dean, B. Cockayne, and W.R. MacEwan, "Optical and capacitance spectroscopy of InP:Fe ", J. Phys. C: Solid State Phys., 14, page 5069, 1981.
50. S.T. Pantelides, Deep Centers in Semiconductors, Gordon & Breach, 1986.
51. T. Takanohashi, and K. Nakajima, "Photoionization cross section of InP:Fe ", J. Appl. Phys, 65, page 3933, 1989.
52. S. Bishop, "Iron Impurity Centers", in "Deep Centers in Semiconductors", edited by S.T. Pantelides, page 547, 1986.
53. F.W. Sears, G.L. Salinger, Thermodynamics, Kinetic Theory, and Statistical Thermodynamics, Third Edition, Addison-Wesley, 1975, page 336.
54. TE Van Eck, LM Walpita, WSC Chang, HH Wieder, "Franz-Keldysh electrorefraction and electroabsorption in bulk InP and GaAs ", Appl. Phys. Lett. 48, 1986, page 451.
55. A. Partovi, A. Kost, E.M. Garmire, G.C. Valley, M.B. Klein, "Band-edge photorefractive effect in semiconductors", Appl. Phys. Lett. 56, 1990, page 1089.
56. J.E. Millerd, S.D. Koehler, E.M. Garmire, A. Partovi, A.M. Glass, and M.B. Klein, "Photorefractive gain enhancement in InP:Fe using band-edge resonance and temperature stabilization", Appl. Phys. Lett. 57, page 2779, 1990.
57. Pantelides, Deep Centers in Semiconductors, Gordon and Breach, 1986, page 541 ff.
58. G. Bremond, A. Nouailhat, G. Guillot, "Deep Level Spectroscopy in InP:Fe ", Electronics Letters, 17, 1981, page 55.

59. PR Tapster, MS Skolnick, RG Humphreys, PJ Dean, B Cockayne, and WR MacEwan, "Optical and capacitance spectroscopy of InP:Fe", J. Phys. C: Solid State, 14, 1981, page 5069.
60. G.C. Valley, S.W. McCahon, "Photorefractive measurement of photoionization and recombination cross section in InP:Fe", J. Appl. Phys, 64, page 6684, 1988.
61. DC Look, "Model of Fe+2 intracenter-induced photoconductivity in InP:Fe", Physical Review B, 20, page 4160, 1979.
62. G.C. Valley, S.W. McCahon, "Photorefractive measurement of photoionization and recombination cross section in InP:Fe", J. Appl. Phys, 64, page 6684, 1988.
63. G Picoli, P Gravey, C Ozkul, V Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: A new mechanism of resonance", J. Appl. Phys. 66, page 3798, 1989.
64. A. Chantre, G. Vincent, D. Bois, "Deep level optical spectroscopy in GaAs", Physical Review B, 23, page 5335, 1981.
65. R.K. Willardson, A.C. Beer, Indium Phosphide: Crystal Growth and Characterization, Academic Press, 1990, page 250.
66. G Bremond, A Nouailhat, and G Guillot, "Fe Deep Level Optical Spectroscopy in InP", Solid State Communications, 41, 1982, page 477.
67. G. Lucovsky, "On the photoionization of deep impurity centers in semiconductors", Solid State Communications, 3, page 299, 1965.
68. H.A. Bethe, P. Morrison, Elementary Nuclear Theory, Second Edition, John Wiley, 1956, pages 44-47, 75-84.
69. E.E. Anderson, Modern Physics and Quantum Mechanics, Saunders Company, 1971, page 279.
70. T. Takanohashi and K. Nakajima, "Photoionization cross section of InP:Fe", J. Appl. Phys., 65, page 3933, 1989.
71. J. Strait, J.D. Reed, G.C. Valley, and M.B. Klein, "Net gain in photorefractive InP:Fe at $\lambda = 1.32$ μ m without an applied field", Applied Physics Letters, 57, page 951, 1990.
72. S.M. Sze, Physics of Semiconductor Devices, Second Edition, John Wiley, 1981, page 41.
73. O Madelung, M Schultz, H. Weiss, Numerical Data and Functional Relationships in Science and Technology. Volume 17, Subvolume a, Semiconductors, Springer-Verlag, 1982, page 11.

74. G.C. Valley, S.W. McCahon, "Photorefractive measurement of photoionization and recombination cross section in InP:Fe", J. Appl. Phys, 64, page 6684, 1988.
75. G. Martin and S. Makram-Ebeid, "Mid-gap Donor Level EL2", edited by S.T. Pantelides, Gordon and Breach, page 432, 1986.
76. Chong Hoon Kwak, Seo Yeon Park, Hak Kyu Lee, and El-Hang Lee, "Exact solution of two-wave coupling for photorefractive and photochromic gratings in photorefractive materials", Optics Communications, 79, page 349, 1990.
77. N.V. Kukhtarev, V.B. Markov, S.G. Odulov, M.S. Soskin, and V.L. Vinetskii, "Holographic storage in electro-optic crystals I. Steady State", Ferroelectrics, 22, page 949, 1979
78. D.R. Erbschloe, and T. Wilson, "A simple analytic solution for transient two-wave mixing in photorefractive materials", Optics Communications, 72, page 135, 1989
79. Yu. M. Osipov, and B.I. Sturman, "Transient processes for two-beam interaction in photorefractive crystals", Optics Communications, 79, page 345, 1990
80. A.M. Glass, M.B. Klein, and George C. Valley, "Fundamental limit of the speed of photorefractive effect and its impact on device applications and material research: comment", Applied Optics, 26, page 3189, 1987.
81. P. Yeh, "Fundamental limit of the speed of photorefractive effect and its impact on device applications and material research: author's reply to comment", Applied Optics, 26, page 3190, 1987.
82. P. Yeh, "Fundamental limit of the speed of photorefractive effect and its impact on device applications and material research", Applied Optics, 26, page 602, 1987.
83. P. Yeh, "Two-wave mixing in nonlinear media", IEEE Journal of Quantum Electronics, 25, page 484, 1989.
84. A. Partovi, A. Kost, E.M. Garmire, G.C. Valley, M.B. Klein, "Band-edge photorefractive effect in semiconductors", Appl. Phys. Lett., 56, 1990, page 1089.
85. A.M. Glass, M.B. Klein, and G.C. Valley, "Fundamental limit of the speed of photorefractive effect and its impact on device applications and material research: comment", Applied Optics, 26, page 3189, 1987.
86. G.C. Valley, J. Dubard, A.L. Smirl, and A.M. Glass, "Picosecond photorefractive response of GaAs:EL2, InP:Fe, and CdTe:V", Optics Letters, 14, page 961, 1989.
87. A.L. Smirl, G.C. Valley, K.M. Bohnert, and T.F. Boggess, "Picosecond photorefractive and free-carrier transient energy transfer in GaAs at 1 μ m", IEEE Journal of Quantum Electronics, 24, page 289, 1988.

88. C. Kittel, H. Kroemer, Thermal Physics, Second Edition, Freeman, page 406, 1980
89. M. Tajima, H. Saito, T. Iino, and K. Ishida, "Photoquenching and Recovery Effects of EL2 Absorption in GaAs", Japanese Journal of Applied Physics, 27, page L 101, 1988
90. D.D. Nolte, D.H. Olsen, and A.M. Glass, "Spatial modulation of the Fermi level by coherent illumination of undoped GaAs", Physical Review B, 40, page 10650, 1989-II
91. D.D. Nolte, A.M. Glass, "Nonequilibrium charge transfer and other low-temperature photorefractive effects", Photorefractive Materials, Effects, and Devices, Aussois, France, page 168, 1990.
92. D.D. Nolte, D.H. Olsen, and A.M. Glass, "Nonequilibrium Screening of the Photorefractive Effect", Physical Review Letters, 63, page 891, 1989.
93. R.A. Mullen, " Photorefractive measurements of Physical Parameters", in Topics in Applied Physics Volume 61. Photorefractive Materials and their Applications I: Fundamental Phenomena, edited by P. Gunter and J.P. Huignard, Springer-Verlag, page 109, 1988
94. G.C. Valley, "Short-pulse grating formation in photorefractive materials", IEEE Journal of Quantum Electronics, 19, page 1637, 1983.
95. G.C. Valley, M.B. Klein, R.A. Mullen, D. Rytz, B. Wechsler, "Photorefractive Materials", Ann. Rev. Mater. Sci., page 168, 1988.
96. J.S. Weiner, and P.Y. Yu, "Free carrier lifetime in semi-insulating GaAs from time-resolved band-to-band photoluminescence", J. Appl. Phys., 55, page 3889, 1984.
97. G.C. Valley, A.L. Smirl, "Theory of Transient Energy Transfer in Gallium Arsenide", IEEE Journal of Quantum Electronics, 24, page 304, 1988.
98. A. Von Lehman and J.M. Ballantyne, "Investigation of the nonlinearity in the luminescence of GaAs under high-density picosecond photoexcitation", J. Appl. Phys, 58, page 958, 1985.
99. P.U. Halter, J.C. Fabre, and G. Roosen, "Kinetics of induced nanosecond absorption in photorefractive InP:Fe", Photorefractive Materials, Effects, and Devices, Aussois, page 33, 1990.
100. G Picoli, P Gravey, C Ozkul, V Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: a new mechanism of resonance", J. Appl. Phys., 66, page 3798, 1989.
101. J.S. Weiner, and P.Y. Yu, "Free carrier lifetime in semi-insulating GaAs from time-resolved band-to-band photoluminescence", J. Appl. Phys, 55, page 3889, 1984.

102. A.M. Glass, M.B. Klein, and G.C. Valley, "Fundamental limit of the speed of photorefractive effect and its impact on device applications and material research: comment", *Applied Optics*, **26**, page 3189, 1987.
103. G.F. Alfrey, P.H. Borchers, "Phonon frequencies from the Raman spectrum of indium phosphide", *J. Phys. C: Solid State Phys.*, **5**, page L275, 1972.
104. S.M. Sze, Physics of Semiconductor Devices, Second Edition, page 40, 1981.
105. P. Yeh, "Fundamental limit of the speed of photorefractive effect and its impact on device applications and material research: author's reply to comment", *Applied Optics*, **26**, page 3190, 1987.
106. G.C. Valley, A.L. Smirl, and M.B. Klein, "Picosecond photorefractive beam coupling in GaAs", *Optics Letters*, **11**, page 647, 1986.
107. G.C. Valley, J. Dubard, A.L. Smirl, and A.M. Glass, "Picosecond photorefractive response of GaAs:EL2, InP:Fe, and CdTe:V", *Optics Letters*, **14**, page 961, 1989.
108. N.V. Kukhtarev, "Kinetics of hologram recording and erasure in electrooptic crystals", *Soviet Technical Physics Letters*, **2**, page 438, 1976.
109. G.C. Valley, M.B. Klein, "Optimal properties of photorefractive materials for optical data processing", *Optical Engineering*, **22**, page 704, 1983.
110. P. Gunter, J.P. Huignard, "Photorefractive Effects and Materials", in Topics in Applied Physics Volume 61: Photorefractive Materials and their Applications I: Fundamental Phenomena, edited by P. Gunter and J.P. Huignard, page 56, 1988.
111. P. Gunter, J.P. Huignard, Photorefractive Materials and their Applications I, Springer-Verlag, 1988, page 175.
112. J. Strait and A.M. Glass, "Time-resolved photorefractive four-wave mixing in semiconductor materials", *J. Opt. Soc. Am. B*, **3**, page 342, 1986.
113. M.B. Klein, "Beam coupling in undoped GaAs at 1.06 μm using the photorefractive effect", *Optics Letters*, **9**, page 350, 1984.
114. F. Vachss, "An analytic expression for the photorefractive two beam coupling response time", *Photorefractive Materials, Effects, and Devices*, Aussois, France, page 140, 1990.
115. L. Solymar, J. Heaton, "Transient energy transfer in photorefractive materials: an analytic solution", *Opt. Comm.*, **51**, page 76, 1984.
116. N.V. Kukhtarev, V.B. Markov, S.G. Odulov, M.S. Soskin, V.L. Vinetskii, "Holographic storage in electro-optic crystals, II: Beam coupling", *Ferroelectrics*, **22**, page 961, 1979.

117. M. Cronin-Golomb, Photorefractive Materials, Effects, and Devices, edited by G. Valley and P. Yeh, page 142, 1987.
118. H Kogelnik, "Coupled wave theory for thick hologram gratings", Bell System Technical Journal, 48, 1969, page 2909.
119. G Picoli, P Gravey, C Ozkul, V Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: A new mechanism of resonance", J. Appl. Phys., 66, page 3798, 1989.
120. J Feinberg, D Heiman, AR Tanguay, RW Hellwarth, "Photorefractive effects and light-induced charge migration in barium titanate", J. Appl. Phys., 51, 1980, page 1297.
121. JC Fabre, JMC Jonathan, G Roosen, "43m Photorefractive Materials in Energy Transfer Experiments", Optics Communications, 65, page 257, 1988
122. SA Boothroyd, J Chrostowski, MS O'Sullivan, "Photorefractive GaAs for Coherent Detection", Photonic Networks, Components, and Applications, editors J. Chrostowski, J. Terry, World Scientific, 1991, page 228.
123. Numerical Data and Functional Relationships in Science and Technology, Volume 17 Semiconductors, Subvolume a, Landolt-Bornstein, Springer-Verlag, 1982, page 288.
124. P. Gunter, CRC Handbook of Laser Science and Technology, Volume IV, Optical Materials: Part 2, Marvin J. Weber, 1986, page 348.
125. C. Ozkul, G. Picoli, P. Gravey, and N. Wolffer, "High gain coherent amplification in thermally stabilized InP:Fe crystals under dc fields", Applied Optics, 18, 1990, page 2711.
126. P. Gravey, G. Picoli, and J.Y. Labandibar, "Stabilization of photorefractive two-beam coupling in InP:Fe under high DC fields by temperature control", Optics Communications, 70, 1989, page 190.
127. B. Mainguet, "Characterization of the photorefractive effect in InP:Fe by using two-wave mixing under electric fields", Optics Letters, 13, 1988, page 657.
128. Optics for Research Design Memo # 1, "Focusing a Laser Beam to a Minimum Spot", 1989.
129. Brittanica, 23, page 703.
130. N.V. Kukhtarev, "Kinetics of hologram recording and erasure in electro-optic crystals", Soviet Technical Physics Letters, 2/12, December 1976, page 438.
131. R. Mullen, "Photorefractive measurements of physical parameters", in Topics in Applied Physics Volume 61, Photorefractive Materials and their Applications and their Applications I: Fundamental Phenomena, Editors: P. Gunter, J.-P. Huignard, 1988, page 167.

132. M.B. Klein, "Photorefractive Properties of BaTiO₃", in Topics in Applied Physics Volume 61, Photorefractive Materials and their Applications and their Applications I: Fundamental Phenomena, Editors: P. Gunter, J.-P. Huignard, 1988, page 193.
133. NV Kukhtarev, VB Markov, SG Obulov, MS Soskin, and VL Vinetskii, "Holographic Storage in Electro-optic crystals. I. Steady State", *Ferroelectrics*, 22, 1979, page 949.
134. A.M. Glass, and J. Strait, "The Photorefractive Effect in Semiconductors", in Topics in Applied Physics Volume 61, Photorefractive Materials and their Applications and their Applications I: Fundamental Phenomena, Editors: P. Gunter, J.-P. Huignard, 1988, page 237.
135. M.B. Klein, "Beam coupling in undoped GaAs at 1.06 um using the photorefractive effect", *Optics Letters*, 9, 1984, page 350.
136. G. Picoli, P. Gravey, V. Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: A new mechanism of resonance", *Journal of Applied Physics*, 66, page 3798, 1989.
137. *ibid*, G. Picoli et al.
138. H. Rajbenbach, J.M. Verdiell, and J.P. Huignard, "Visualization of electrical domains in semi-insulating GaAs:Cr and potential use for variable grating mode operation", *Applied Physics Letters*, 53, page 541, 1988.
139. G. Picoli, P. Gravey, V. Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: A new mechanism of resonance", *J. Appl. Phys.*, 66, page 3798, 1989
140. L.J. Cheng and A. Partovi, "Temperature and intensity dependence of photorefractive effect in GaAs", *Applied Physics Letters*, 49, page 1456, 1986.
141. C. Ozkul, G. Picoli, and N. Wolffer, "High gain coherent amplification in thermally stabilized InP:Fe crystals under dc fields", *Applied Optics*, 29, page 2711, 1990.
142. C. Ozkul, G. Picoli, and N. Wolffer, "High gain coherent amplification in thermally stabilized InP:Fe crystals under dc fields", *Applied Optics*, 29, page 2711, 1990.
143. P. Gunter and J.P. Huignard, Photorefractive Materials and their Applications II, Survey of Applications, Springer Verlag, 1989.
144. G. Hamel de Montchenault, B. Loiseaux, and J.P. Huignard, "Amplification of high bandwidth signals through two-wave mixing in photorefractive BSO crystals", *Topical Meeting on Photorefractive Materials, Effects, and Devices*, Paper WD5, Los Angeles, CA, 1987.
145. R.B. Bylisma, A.M. Glass, D.H. Olsen, "Optical signal amplification at 1.3 um by two-wave mixing in InP:Fe", *Electronics Letters*, 24, page 360, 1988.

146. D. Dolfi, B. Forestier, B. Loiseaux, H. Rajbenbach, J.P. Huignard, "Two-wave mixing using dual frequency signals in BaTiO₃ - application to the amplification of microwave signals", Photorefractive Materials, Effects, and Devices, Aussois, France, page 390, 1990.
147. Y. Yamamoto, T. Mukai, "Fundamentals of optical amplifiers", Optical and Quantum Electronics, 21, page S1, 1989.
148. O.P. Gandhi, Microwave Engineering and Applications, Pergamon, page 426, 1985.
149. H.E. Rowe, "Some general properties of nonlinear elements. II. Small signal theory", Proceedings of the IRE, page 48, 1958.
150. G.C. Gilbreath, "Forward multibeam mixing in diffusion-driven photorefractive media", Journal of Lightwave Technology, 9, page 105, 1991.
151. F. Davidson, L. Boutsikaris, M. Krainak, "Coherent optical detection through two-wave mixing in photorefractive materials", Optics Letters, 13, page 506, 1988
152. S.A. Boothroyd, J. Chrostowski, M.S. O'Sullivan, "Photorefractive GaAs for Coherent Detection", Photonic Networks, Components, and Applications, editors J. Chrostowski, J. Terry, World Scientific Publishing Co., 1991, page 228.
153. D.Y. Zang, V.M. Ristic, "All-Optical Erasable Programmable Read-Only Memory (EPROM) Based on LiNbO₃ Channel Waveguides", IEEE Photonics Technology Letters, 1, page 323, 1989
154. L.B. Aronson, L. Hesselink, "Photorefractive integrated-optical switch arrays in LiNbO₃", Optics Letters, 15, page 30, 1990
155. T. Parish, "Crystal Clear Storage", BYTE, page 283, November 1990.
156. J. Feinberg and K.R. MacDonald, "Phase-conjugate mirrors and resonators with photorefractive materials", Photorefractive Materials and their Applications II: Survey of Applications, edited by Gunter and Huignard, page 151, 1988
157. J.P. Huignard, and P. Gunter, "Optical processing using wave mixing in photorefractive crystals", Photorefractive Materials and their Applications II: Survey of Applications, edited by Gunter and Huignard, page 205, 1988
158. P. Yeh, A.E. Chiou, J. Hong, P. Beckwith, T. Chang, M. Khoshnevisan, "Photorefractive nonlinear optics and optical computing", Optical Engineering, 28, page 328, 1989
159. G. Gheen, L.J. Cheng, "Image processing by four-wave mixing in photorefractive GaAs", Applied Physics Letters, 51, page 1481, 1987
160. Tabiani, NRC Lecture. 22 November 1989, MIT Sabbatical & Guest Worker at Ottawa University.

161. P. Yeh, A.E. Chiou, J. Hong, P. Beckwith, T. Chang, M. Khoshnevisan, "Photorefractive nonlinear optics and optical computing", *Optical Engineering*, 28, page 328, 1989.
162. H. Rajbenbach, A. Delboulbe, and J.P. Huignard, "Noise-free photorefractive image amplifiers", *Photorefractive Materials, Effects, and Devices*, Aussois, France, page 329, 1990
163. D.Z. Anderson, J. Feinberg, "Optical Novelty Filters", *IEEE Journal of Quantum Electronics*, 25, page 635, 1989
164. A.M. Glass, M.B. Klein, and G.C. Valley, "Photorefractive determination of the sign of photocarriers in InP and GaAs", *Electronics Letters*, 21, page 220, 1985
165. J.P. Zielinger and M. Tapiero, "Considerations on the optimization and relevant characterization of photorefractive materials", Photorefractive Materials, Effects, and Devices, Aussois, France, page 58, 1990.
166. C. Ozkul, G. Picoli, P. Gravey, and N. Wolffer, "High gain coherent amplification in thermally stabilized InP:Fe crystals under dc fields", *Applied Optics*, 29, page 2711, 1990.
167. G.C. Gilbreath, "Forward Multibeam Mixing in Diffusion-Driven Photorefractive Media", *Journal of Lightwave Technology*, 9, page 105, 1991.
168. C. Ozkul, G. Picoli, P. Gravey, and N. Wolffer, "High gain coherent amplification in thermally stabilized InP:Fe crystals under dc fields", *Applied Optics*, 29, page 2711, 1990.
169. G. Picoli, P. Gravey, C. Ozkul, V. Vieux, "Theory of two-wave mixing gain enhancement in photorefractive InP:Fe: a new mechanism of resonance", *J. Appl. Phys.* 66, 1989, 3798.
170. G Godefray, G Ormoncey, P Jullien, P Lompre, W Ousi, Y Semonov, "Crystal Growth, Electro-optic and Photorefractive Properties of Iron-doped BaTiO₃", *Topical Meeting on Photorefractive Materials, Effects, and Devices*, August 12-14, 1987, page 159.

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