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Dimensionality of InGaAs Nonlinear Optical Response

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(Ph.D. Thesis)

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DIMENSIONALITY OF InGaAs NONLINEAR OPTICAL RESPONSE

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Ph.D. Thesis

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Abstract

Dimensionality Dependence of InGaAs Nonlinear Optical Response

by

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Doctor of Philosophy in Physics

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In this thesis the ultrafast optical properties of a series of InGaAs samples ranging from the two to the three dimensional limit are discussed. An optical system producing 150 fs continuum centered at 1.5 microns was built. Using this system, ultrafast pump-probe and four wave mixing experiments were performed. Carrier thermalization measurements reveal that screening of the Coulomb interaction is relatively unaffected by confinement, while Pauli blocking nonlinearities at the band edge are approximately twice as strong in two dimensions as in three. Carrier cooling via phonon emission is influenced by confinement due both to the change in electron distribution function and the reduction in electron phonon coupling. Purely coherent band edge effects, as measured by the AC Stark effect and four wave mixing, are found to be dominated by the changes in excitonic structure which take place with confinement.

Dimensionality of InGaAs Nonlinear Optical Response

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Chapter 1

Introduction

1.1 Motivation

Ultrafast dynamics in semiconductors have been studied with great vigor ever since the development of ultrafast laser sources in the early 1980's. These dynamics are of fundamental interest for understanding fundamental principles of condensed matter physics. For example, previous work has examined the physics of scattering at the Fermi surface [1], the electron phonon interaction [2,3], electron-electron scattering and electron hole scattering [4,5,6], as well as the physics of non-thermal distributions [7].

Simultaneously with the advent of ultrafast sources, sample growth techniques were rapidly improving, allowing for the production of complex structures made of layers of different compounds with monolayer accuracy. The ability to grow such multilayers led very rapidly to the development of quantum confined structures, in which two materials with different band gaps are layered, producing spatial discontinuities in the band gap along the growth direction. [8] When several layers of these materials are placed in sequence, the resulting structures are known as semiconductor quantum wells, because electrons and holes are confined to the low band gap layers, which, if they are made sufficiently thin, cause the carriers to live in a quasi two dimensional world.

Studies of quantum well structures were quickly realized to be of both fundamental and technical importance. Clearly, semiconductor physics in two dimensions would not be the same as in three, as was observed early on in studies of the effect of quantum confinement on linear absorption and transport. Early studies (both theoretical and experimental) of quantum wells revealed that they have a very different density of states than do bulk samples, and that their excitonic absorption is enhanced by confinement. [10]

The enhanced density of states at the band edge and increased excitonic singularity has led many workers to develop electro-optical devices based on quantum confined structures. These devices include semiconductor lasers and amplifiers, as well as modulators and transistors. Operation of these devices is enhanced over their bulk counterparts in the sense that they require reduced total carrier density to achieve the same amplification, due to the increase in the band edge density of states. This results, for example, in lower threshold currents for lasing in quantum confined than in bulk structures. Devices have also been developed that were based on effects that only exist in the case of quantum confinement, such as the quantum confined stark effect. [9].

For all of these devices it is the confinement induced change in the steady state, linear optical properties which initially motivated use of quantum confined structures. However, the point of using electro-optic rather than all electrical devices in communications is the increased speed which is available. Thus, an important question for device development is the effect of quantum confinement on the ultrafast optical properties of semiconductors. The physics which underlies the answer to this question is not very well understood. It is known that a number of many body interactions which affect ultrafast properties, such as screening, Pauli-blocking, and the electron phonon interaction are different in two dimensions than in three. However real structures exist in a regime intermediate between the absolute two and three dimensional limits, and in this regime very little theoretical or experimental work has been done. In fact, virtually all of the ultrafast dynamics of real semiconductors result from a complex interplay among many

different physical interactions. Because of this complexity, it is very difficult to model exactly how confinement will affect the ultrafast properties of a particular structure.

In order to answer these questions we undertook a study of the ultrafast optical properties of a series of samples with varying quantum confinement. We selected InGaAs/InAlAs for our heterostructures. This material system is excellent for studies of confinement because InGaAs has a relatively small band gap (800 meV), which leads to large, weakly bound excitons. The exciton Bohr radius in this system is $290\text{\AA}$, more than twice that in the more commonly studied material GaAs. The large Bohr radius allows us to reach the strongly confined limit, in which the Bohr radius is much smaller than the well width, without having to go to wells so narrow that inhomogeneous broadening due to well width fluctuations becomes important. We studied wells ranging in width from $100\text{\AA}$ (one third the Bohr radius) to $6000\text{\AA}$ (twenty times the Bohr radius). InGaAs is also technologically important because its band gap is near the zero dispersion point for silicon based optical fibers, and thus it has been widely exploited for electro-optical devices.

As mentioned above, there has been substantial work on the ultrafast optical properties of semiconductors, [11] however the vast majority of this work has been on GaAs and larger band gap materials. Although studies of both narrow quantum wells [5,6,7] and bulk materials [12] have been performed, no experiments in which the transition from two to three dimensions is examined have been carried out. One study of the effect of confinement on spin relaxation dynamics was performed recently, [13] however in this case the variation in confinement was achieved by varying the composition of the barriers, which varies the depth of the wells, rather than their width. Changing the

depth of the wells varies the penetration of the wavefunction into the barrier layers very strongly, which is not the case for work in which well width is varied. Some work on ultrafast properties of InGaAs based materials has been done [14,15], however all of this work has been limited to single wavelength studies, in which the dynamics of only one energy level of the system can be probed.

Our experiments are performed with a broad-band, ultrafast source at 1.5 microns, which allows the use of pump-broadband probe techniques in which the dynamics of a wide spectral range of the system can be explored. We studied a variety of ultrafast optical properties, including transient band edge nonlinearities, carrier thermalization and cooling dynamics, as well as coherence dynamics at the band edge. This allows us to understand the effects of confinement on a number of the physical interactions governing the system, including screening, Pauli-blocking, the electron phonon interaction, and carrier carrier scattering. These are the first ultrafast, multiwavelength studies performed in the InGaAs system, and in addition they are the only systematic exploration of the evolution of ultrafast dynamics from two to three dimensions.

1.2 Overview of Thesis Content

In order to perform the broad-band, ultrafast measurements of our samples, it was necessary to develop a unique source centered at 1.5 microns. The development and characterization of our NaCl based laser, amplifier and continuum generation system is covered in Chapter two. This chapter also discusses the basics of our low temperature,

spectrally resolved pump broad band probe and four wave mixing experiments. In the process of developing our laser system we encountered some unexpected nonlinear dynamical effects. These are discussed in Appendix 2.

In Chapter three the effects of confinement on the linear optical properties of semiconductors are presented. The modifications in the densities of states of the carriers are discussed, and excitonic linear absorption is reviewed. In this chapter I present the material details and linear optical spectra of our samples, and draw conclusions about homogeneous and inhomogeneous broadening of the excitons.

Chapter four gives an overview of the theory of semiconductor nonlinear dynamics based on the semiconductor Bloch equations. Solutions to these equations are obtained in the case of no Coulomb interaction among the dipoles for both pump probe and four wave mixing experiments. The importance of the Coulomb interaction for semiconductor systems is discussed, and some cases in which the semiconductor Bloch equations are not applicable are presented.

Chapter five begins the presentation of our ultrafast pump probe measurements. This chapter focuses on measurements of carrier thermalization and cooling after excitation high above the band edge. Comparisons between bulk, quantum well, and intermediate dimensionality samples are made for screening, Pauli blocking, band edge transients, and the electron phonon interaction.

In Chapter six our measurements of the AC Stark effect for all samples are presented. A review of the principles of the Stark effect is given, followed by an analysis

of the data in terms of recently developed theories of the ultrafast ACSE, which differs markedly from the steady state results.

Chapter seven explores the coherent dynamics at the band edge in four of our samples. Spectrally resolved four wave mixing measurements at low and high carrier densities are presented. The temporal and spectral behaviors of the four wave mixing signals are analyzed in terms of both dynamic and steady state properties of the exciton.

Chapter eight presents an overview of the conclusions of our work, and presents some open questions and possibilities for future research.

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Chapter 2

Experimental System

2.0 Introduction

In this chapter I will discuss the experimental system used for our measurements. Section 2.1 details the requirements of the experiment and outlines the techniques used to make the measurements. Section 2.2 outlines the optical system we developed. This system is covered in more detail in sections 2.3, 2.4, 2.5 and 2.6, which explain the physics of the gain medium (NaCl), laser, amplifier, and continuum generation respectively. In section 2.7 I present a summary of our results on the system.

2.1 Experimental Requirements

The experiment we undertook involved the measurement of transient, nonlinear optical effects in a series of InGaAs heterostructures of various well widths. The well widths we used ranged from 100Å to 6000Å, and thus the band edge at low temperature varied from 1.425 to 1.550 μm . In addition to the behavior of carriers excited at the band edge, we also were interested in exciting carriers well above the band, to study carrier relaxation, and well below the band, to study the AC Stark effect. For all of these measurements we needed to be able to measure the effects of the excitation over a broad range of energy in the sample, ranging from below the band edge to approximately one LO phonon (50 nm) above the band.

We used two techniques to make our measurements. The first is the spectrally resolved pump probe technique, which has been used by many other workers in the visible and near infrared (800nm) to study carrier dynamics in GaAs. [1] In this technique, the sample is excited with an intense, relatively narrow band (transform limited) pump pulse. A broadband probe pulse is then used to measure the absorption spectrum of the sample as a function of time delay between the pump and probe pulses. In order to measure small signals and account for any spectral structure on probe, it is helpful to use a differential

technique. In this technique the absorption spectrum of the sample in the *presence* of the pump (T) is normalized to the absorption in the *absence* of the pump (T_o), giving a differential transmission spectrum (DTS) which is defined as follows:

$$DTS \equiv \frac{T_o - T}{T_o} \equiv \Delta T / T$$

The detailed theory of the DTS will be treated in the section 4.4.

An overview of the physical layout of the experiment is shown in figure 2.1. The light from the laser/amplifier/continuum generation system is split into pump and probe beams by a polarizing beam splitter cube. This is a convenient way to split the two beams as the ratio of the two can be easily adjusted with a wave plate before the cube. The two beams are then spectrally filtered appropriately using interference filters from Spectrogon. The variable delay between pump and probe is controlled using a Klinger stepper motor in the pump line. The pump and probe beams are focussed onto the sample in a small liquid Helium finger cryostat. Generally, the pump beam is focussed to a spot approximately three times the size of the probe spot so that the probe will see a relatively uniform pump intensity. The probe beam is then sent through a 1/2 meter single grating spectrometer to an InGaAs optical multichannel analyzer (Princeton Applied Research) for spectral measurements. Alternatively, the output of the spectrometer can be sent to a single element Germanium detector for detailed time measurements at a fixed wavelength. In this case the pump beam is chopped at 10 to 25 Hz and the output from the detector is sent to a lock-in amplifier. In both cases, the data acquisition is controlled through a GPIB board run with ASYST. The acquisition software is titled ACQUIRE, and was written by Jason Stark at AT&T Bell laboratories.

The second experimental technique we used is spectrally resolved four wave mixing. In this technique two pulses incident on the sample in directions k_1 and k_2

Figure 2.1 Schematic of infrared laser/amplifier/continuum generation system.

interfere to produce a third beam which propagates in the direction $2k_1 - k_2$ or $2k_2 - k_1$. One diffracted beam is sent to the spectrometer and OMA, where its spectrum is resolved as a function of time delay between the two incident beams. We used this technique, which is discussed in more detail in section 4.3, to study coherent dynamics at the band edge of our samples.

In order to perform these measurements, we thus required a broadband coherent source producing wavelengths from at least $1.30\mu\text{m}$ to $1.65\mu\text{m}$. Many of the processes we chose to study, such as thermalization of a non thermal carrier distribution, take place on the sub- 100fs time scale. Thus short pulses are a necessity for this experiment. The intensities required for the experiments varied somewhat. The order of magnitude necessary for the pump energy is given by the saturation fluence of the excitons in the sample, on the order of 1 nJ. Thus, if we excite the sample with 1 nJ of energy anywhere above the band edge we can expect to create very strong perturbations of the absorption spectrum of the pump, giving an easily measureable signal. For weaker nonlinearities, such as the AC Stark effect, a 1nJ pump gives a much weaker signal, on the order of a few percent. Our detection system can pick up differential signals of about .5%. Thus, approximately 1nJ of energy in a 20nm (transform limited) bandwidth is required for the pump beam in all of our experiments, meaning that we need at least 1 nJ of energy in 20 nm over the spectral range from 1.3 to $1.65\mu\text{m}$. For the probe pulse, the requirements are somewhat different. The role of the probe pulse is to measure the absorption spectrum of the sample over the energy range of interest. For some experiments, such as the AC Stark effect, the nonlinearity in the system is strongly dominated by the exciton. In this case the probe can be quite spectrally narrow, with a full width half maximum on the order of 30 nm. For the carrier relaxation experiments, however, it is necessary for the probe's spectral window to cover the region from well above the pump excitation to well below the band edge. This is generally approximately 150 nm. Our maximum spectral range was limited by the size of the OMA and the groove spacing of the spectrometer grating.

For 150 lines per inch the spectral range of the OMA is 250 nm, with approximately 1 nm resolution. In principle, since we use a differential measurement, it would be acceptable for the probe spectrum to be quite non-uniform in intensity. In reality however, the range over which the probe intensity can vary is limited by the dynamic range of the OMA, which for InGaAs OMAs which operate at 1.5 microns, is only about twenty. Thus, in order to measure our signals over the relevant spectral range it is necessary that the probe spectrum be flat to within about a factor of five, over a 150nm bandwidth. Of course, in order to obtain meaningful data about the temporal evolution of the semiconductor nonlinearities, it is critical that the pump and probe beams are temporally flat, that is, that the whole spectral width of the probe beam arrives at once, without chirp. The remainder of this chapter will address the optical system that we built in order to fulfill these requirements.

2.2 Optical system overview

While some time-resolved measurements can be done with a single-wavelength laser source, the investigation of many physical processes in semiconductors requires nondegenerate (multiple-wavelength) pump-probe measurements in order to "see" what is happening at different energy levels. This requires either a synchronized, femtosecond, dual-wavelength laser source, or a femtosecond continuum system. Continuum systems, in particular, have gained widespread acceptance due to their versatility and broad spectral coverage. Such systems have proven to be very valuable in studies of GaAs/AlGaAs heterostructures at wavelengths near 800 nm [1]. For our work, a similar system operating near 1.5 μm is necessary. In fact, interest in such a system is not limited to the study of semiconductors but includes chemical and biological processes as well.

The last few years have seen tremendous advances in short pulse technology in the infrared. These advances have been based primarily on color-center laser technology.

Perhaps the single most important development was the soliton laser [2]. This was followed soon afterward by more general coupled cavity techniques (such as Additive Pulse Modelocking [3,4] and/or Coupled Cavity Modelocking [5]) which do not rely on soliton effects. Both the soliton laser and the APM/CCM lasers have produced pulses as short as 60 fsec. Coupled-cavity techniques have been successfully applied to the modelocking of Erbium-doped fiber lasers as well [6]. Additionally, workers have succeeded in passively modelocking color-center lasers using either InGaAs/InP multiple quantum wells or bulk InGaAsP as a saturable absorber [7]. Such passively modelocked F-center lasers have produced pulses as short as 200 fsec. A significant development toward dual-wavelength sources has been the demonstration of a femtosecond optical parametric oscillator (OPO) pumped by a colliding pulse modelocked (CPM) laser [8]. This source can produce two synchronized, tunable infrared pulses at different wavelengths ($\lambda_{\text{sig}} = 1.0 \mu\text{m}$, $\lambda_{\text{idler}} = 1.6 \mu\text{m}$) with pulsewidths as short as 100 fsec. This source has been used in measurements of relaxation times in InGaAs. Even more promising are the femtosecond OPO's which are pumped by modelocked Ti:sapphire lasers [9,10]. Finally, amplified, femtosecond Ti:sapphire lasers can generate continuum with average powers approaching 1-Watt [11]. Preliminary measurements indicate a substantial amount of continuum light in the $1.5 \mu\text{m}$ range. This review is by no means comprehensive; several other techniques (such as difference frequency generation and fiber Raman-soliton generation) are available as well.

Existing femtosecond laser sources at $1.5 \mu\text{m}$ (such as the APM, or the passively modelocked color-center laser) can produce pulses with approximately 1 nanojoule of energy, with peak powers of approximately 10 kilowatts. This is ample power for many types of nonlinear experiments. However, continuum generation with 100 fsec pulses requires pulse energies on the order of 1 microjoule. Thus, it is necessary to amplify the pulses from an APM laser (for example) by roughly a factor of 1000 to obtain continuum generation.

In this section, I describe an amplified, infrared laser system which generates femtosecond, optical pulses near $1.5\ \mu\text{m}$ to energies exceeding 50 microjoules. Short pulses (140 fsec) from an additive-pulse modelocked NaCl laser are amplified in a multipass amplifier in which NaCl F-centers serve as the gain medium. This system can generate peak powers approaching 1 GW, with wavelengths tunable from $1.53\ \mu\text{m}$ to $1.60\ \mu\text{m}$. The high-power pulses are capable of generating an extremely broadband femtosecond continuum which is usable not only in the $1.5\ \mu\text{m}$ wavelength region, but extends from the UV (400 nm) to the mid-infrared ($3.5\ \mu\text{m}$).

2.3 NaCl F-centers as a gain medium

There are few choices for gain media near 1.5 microns. The need for broad gain bandwidth, which is necessary to produce short pulses, restricts the field even further. A number of gain media were considered as candidates for a femtosecond oscillator/amplifier system. The final choice of NaCl F_2O^+ centers was due largely to the requirements of the amplifier, as a number of different oscillators can operate in this regime. Once NaCl was selected for the amplifier, the oscillator was chosen to be NaCl as well, in order to optimize the overlap of the oscillator and amplifier gain spectra. Some alternatives are presented here.

Erbium-doped fibers would provide the convenience and simplicity of coupling the pump and the femtosecond pulses into an optical fiber. However, the gain bandwidth is too narrow for 100 fsec pulses, which would result in considerable spectral narrowing (and thus temporal broadening) of a broadband femtosecond pulse. The narrow gain bandwidth would also afford little or no tunability. In addition, we could expect a host of deleterious nonlinear effects (such as stimulated Raman scattering and/or damage) which would occur at such high pulse powers in a long fiber, although this might be alleviated somewhat by the use of chirped pulse amplification. New solid-state laser media doped

with Cr^{4+} such as Cr:YAG provide broad bandwidth in this wavelength range and appear to be very promising for femtosecond applications [Cr:YAG]. However, they are not yet widely available.

The color-centers in KCl:Tl and NaCl seem to be the best choices. However, both systems suffer from the same drawback: the need for cryogenics. KCl:Tl would be a suitable gain medium [12]. It combines the advantages of a moderately long lifetime ($3\mu\text{s}$), no need for auxiliary radiation (unlike NaCl), and a sufficiently broad bandwidth. Its gain spectrum is centered around $1.5\mu\text{m}$. To date, KCl:Tl lasers have generated the shortest pulses (60 fsec) of any of the color-center lasers. However, the crystal processing is considerably more difficult (involving e-beam irradiation to create damage centers in the crystal) and the production of large crystals suitable for an amplifier has not been attempted by anyone to our knowledge. Also, KCl:Tl has little tolerance for handling at room temperature, in normal room light, which is required for polishing the large areas necessary for an amplifier crystal. NaCl, on the other hand, can tolerate a reasonable degree of handling at room temperature in normal room light. We chose NaCl on the basis of its availability, relative ease of handling, and its superior tunability.

Figure 2.2 Gain spectrum of the NaCl F_2 center.

In NaCl the the laser active center (denoted by F_2O^+) consists of an F_2^+ center bound to an O^{2-} impurity. The F_2^+ center consists of two chlorine vacancies bound together in the NaCl lattice, with one missing electron. This gives a center with physics

similar to that of a helium atom. It provides a four level system for lasing, in which excitation is followed by two fast configurational relaxations to a metastable state approximately 800meV above the ground state. The binding of the F_2^+ to the O^{2-} impurity prevents the F_2^+ center from migrating in the crystal, which causes irreversible fading during laser operation. For convenience, some of the relevant radiative properties of F_2O^+ centers in NaCl have been summarized in Table I. A detailed description of the properties of the F_2O center in NaCl can be found in ref. [13]. The gain spectrum of F_2O centers in NaCl is shown in Figure 2.2, where we have also shown the theoretical spectrum of a transform-limited 70 fsec pulse for comparison. Due to their large gain bandwidth ($\Delta\lambda \sim 200\text{nm}$), NaCl F-centers provide a nearly ideal medium for amplification of short pulses at 1.5 μm . The gain cross-section, σ , and thus the saturation fluence (E_{sat}) is very similar to that of organic laser dyes. The upper state lifetime is 150 nsec, and is almost purely radiative. The absorption band for the desired transition peaks near 1.06 μm , making Nd:YAG or Nd:YLF lasers ideal as pump sources. The energy storage density is roughly a few millijoules per cm^3 .

Gain bandwidth*	$\Delta\lambda = 200 \text{ nm}$
Gain cross section*	$\sigma = 7 \times 10^{-17} \text{ cm}^2$
Gain lifetime	$\tau_R = 150 \text{ ns}$
Saturation energy*	$E_{\text{sat}} = 1.8 \text{ mJ/cm}^2$
Saturation intensity*	$I_{\text{sat}} = 20 \text{ kW/cm}^2$
F-center density	$N_o \sim 4 \times 10^{16} \text{ cm}^{-3}$
Energy storage density	$\rho_e = 3 \text{ mJ/cm}^3$

Table I. Properties of $F_2^+ : O^{2-}$ Centers in NaCl:OH⁻

Like many other F-center systems, the F_2O centers in NaCl exhibit orientational bleaching, which degrades the gain and can seriously affect laser performance. For NaCl, this effect is counteracted by the use of auxiliary radiation in addition to the normal pump light. This auxiliary radiation is most commonly implemented using blue light from a filtered Mercury arc-lamp, or green light derived from either an argon laser (514 nm) or a frequency-doubled YAG laser (532 nm).

Here, with aid of figure 2.3, I will briefly describe the process of orientational bleaching and the methods used to counteract it. For a more complete description of this process see Ref. [15]. Figure 2.3 schematically shows the F-center dipoles (A , $\bar{A}$, B , $\bar{B}$, C , $\bar{C}$) which are oriented along the $[110]$ axes of the crystal. The polarization vectors of the YAG pulse and the $1.5 \mu\text{m}$ pulses are denoted by $PYAG$ and PIR respectively. Horizontally polarized YAG light excites centers B , $\bar{B}$, C , $\bar{C}$ but not A or $\bar{A}$ because $PYAG$ is orthogonal to A and $\bar{A}$. This has two effects, gain dichroism and orientational bleaching. Since centers B , $\bar{B}$, C , $\bar{C}$ are excited by the pump, they all contribute to the gain if PIR is horizontal as well. However, if PIR is vertical, then it can

derive gain only from centers along B , $\bar{B}$. Thus, vertically polarized IR pulses experience less gain than do horizontally polarized pulses for the case of horizontally polarized pump light. Therefore we expect gain dichroism when pumping with linearly polarized light.

Orientational bleaching is due to the spontaneous reorientation of the *excited* centers under intense optical pumping; i.e., the excited centers can sometimes fall into any of the other possible orientations. *Unexcited* centers do not undergo this reorientation. In the case of horizontal PYAG, centers oriented along B , $\bar{B}$, C , or $\bar{C}$ are excited and undergo random reorientations. The result is an accumulation of centers along A , $\bar{A}$ and a depletion of centers along B , $\bar{B}$, C , $\bar{C}$ which results in strongly reduced gain. This reorientational effect is commonly counteracted by exposing the crystal to some form of auxiliary radiation, such as blue light from a mercury arc-lamp or blue/green light from an argon laser. This short wavelength excitation raises the centers in the undesired orientations (in this case, A , $\bar{A}$) to higher excited states and makes it possible for them to flip back into one of the desired orientations.

It is worth mentioning that other F-centers have been used in optical amplifiers and in high-power systems as well. The first application of color-centers to optical pulse amplification, to our knowledge, used KCl:Li F-centers to amplify nanosecond pulses at a wavelength of $\lambda \sim 3 \mu\text{m}$ from another F-center laser [14]. This was a single-pass amplifier with a small-signal gain of about $G=10$ per pass. More recently, nanosecond pulse amplification was demonstrated at wavelengths in the 2.0-2.4 μm range using lithium $\text{F}_2^+ \text{A}$ centers in KCl [15]. The single-pass amplifier had a small-signal gain of 15, and an output pulse energy of over 2 millijoules with a pump pulse energy of 30 millijoules. Color-centers have even been used in high-energy laser systems. A laser with a large LiF: F_2^+ crystal (640 cm^3 volume) was able to generate 100 joule pulses near 1.14 μm when pumped with a 700 Joule YAG laser [16].

NaCl itself has been used in moderately high-power laser systems. A room-temperature NaCl oscillator has produced up to 8 mJ when pumped by 160 mJ from a Q-

switched YAG laser [17]. The combination of large energy storage capabilities with the broad gain bandwidth suggests that NaCl is well suited to moderately high energy femtosecond pulse amplification.

2.4 Additive Pulse Modelocked Laser

A schematic of the entire oscillator/amplifier system is shown in Fig. 2.4. The starting point is the femtosecond laser oscillator. This oscillator is an Additive Pulse Modelocked (APM) NaCl laser which is pumped by a modelocked Nd:YLF laser [18]. The APM laser consists of a synchronously pumped NaCl laser coupled to an external cavity which contains a length of single-mode optical fiber. It produces tunable, femtosecond pulses near $1.5 \mu\text{m}$ with pulse energies of $E_p \sim 1\text{nJ}$, at a repetition rate near 76 Mhz.

The cavity design of our laser is shown in figure 2.5. The gain medium is a NaCl crystal with dimensions $7 \times 5 \times 3\text{mm}$, which is colinearly pumped along the 7mm axis with a modelocked Nd:YLF laser at 76 Mhz. It is also possible to use considerably thinner crystals for lasers of this type, in fact this crystal was originally designed to be pumped along the 3mm dimension. We have found, however, that the additional gain allowed by the longer crystal greatly enhances the operation of the laser, and the crystal is sufficiently uniform that the increased length of the gain medium is not detrimental. The crystal is maintained at 77K in a cryogenic dewar, which has 50mm focal length lenses for its windows. These are mounted on gimbal-like mounts and translation stages. This design allows for tight focussing in the crystal, as well as independent adjustment of focal plane and astigmatism. We have found that the addition of these mounts greatly enhances our ability to control the quality of the laser mode. Depending on the crystal quality, the lasers in our laboratory typically give 10% to 20% efficiencies. Pumping with 5 Watts at $1.06\mu\text{m}$, the laser typically gives 300 to 400mW at line center. Because of the high gain

Figure 2.4 Schematic of the optical system. Elements are beamsplitter (B/S), Faraday isolator (ISO), pockels cell (PC) and photodiode (PD).

of NaCl, the laser is most efficiently operated with output couplers of about 30%. The laser is tuned using the diving angle birefringent tuning plate of Nagano , Lenz and Ippen [19]. This plate has its optical axis at 66 degrees with respect to the surface normal, which allows one to tune the plate over several (approximately seven) orders. Since the different orders have different band pass widths, the plate can be tuned to optimize the bandpass for the combination of stability (from bandwidth limiting) and short pulses (from broad bandwidth.) We have found that this flexibility increases the stability of the laser in the shortest pulse regimes over ordinary tuning plates. Operating in this configuration the synch-pumped laser produces pulses of 5-10 psec when pumped with 80 ps pulses.

Figure 2.5 Schematic of the additive pulse modelocked laser cavity.

In an APM laser, pulses are shortened from 10 ps to 150fs through interferometric recombination of two pulses in a manner which enhances the pulse peak and decreases the pulse wings. This pulse shaping takes place through the addition of a Kerr nonlinearity (in our case 10 cm of dispersion shifted, single mode, optical fiber) in an external cavity coupled to the main cavity. In addition to the fiber, the external cavity contains a 55%/45% beamsplitter and an end mirror. As a pulse leaves the main cavity, it is split by the beamsplitter and 45% of it is sent to the external cavity, the remainder acting as the laser output. The pulse in the external cavity passes through the fiber, is retroreflected

and then sent back through the fiber to the main cavity. The main and external cavities are maintained at the same length. As the pulse passes through the fiber, it undergoes an intensity dependent phase shift due to the Kerr nonlinearity ($n_2(I)$). The pulse is then returned to the main cavity, where it combines interferometrically at the beamsplitter with a pulse coming from the main cavity. The shape of the pulse which is reinjected into the laser depends in detail on the relative phase shifts of the peak and wings of the pulse. Since this phase shift is intensity dependent, the entire external cavity can be treated exactly as an intensity dependent mirror. The details of the pulse shaping dynamics in an APM are explored elsewhere [20], [21].

Given the mechanism of pulse shaping in the APM, it is clear that two parameters are critical. The first is the total phase shift undergone by the pulse in the external cavity, which is given by the combination of the fiber length and the intensity of the pulses in the fiber. The second is the precise matching of the two cavity lengths. In our laser this is maintained through an active feedback system which monitors the laser intracavity power with a photodiode, compares it to a reference level, and gives a feedback voltage to a piezo mounted on the back of the external cavity end mirror. When the cavity is synchronously pumped, the intracavity power oscillates due to the fact that the APM will "choose" its own frequency of operation, which is generally a little different from the repetition rate of the modelocked YLF. The power will then oscillate at the difference frequency between these two repetition rates (generally a few kHz.) It is also possible [22] to operate the APM in a self starting mode, where the YLF modelocker is turned off and the YLF is allowed to run CW. In this case, the pulsing in the APM builds up from modebeating in the Nd:YLF laser. Although the self-starting APM generally gives a quieter output (since it has no oscillations due to cavity walk-off) the parameter range for its operation is very narrow, and so it often drops out and will not restart without intervention. Thus, for our experiments we have used the laser in the non self-starting configuration.

The output of our laser is tunable from about 1.55 to 1.62 μm , and is generally about 150 mW after the beamsplitter. The pulse widths vary widely depending on the details of power, wavelength, and position of the birefringent tuner. For experiments we use the shortest stable pulses, which are sech^2 with about 140 fs duration, and operate near the maximum of the gain at 1.58 μm .

The output of the APM laser is sent to the amplifier through an adjustable telescope (for mode-matching), a Faraday isolator, and a pockels cell switchout. When trying to generate continuum, it became necessary to insert the Faraday isolator between the oscillator and the amplifier because back-reflections from the continuum generator would get re-amplified and shot back to the APM. This not only drove the APM laser unstable, it also damaged the control fiber and the gain crystal of the APM.

All timing and triggering is done by the pockels cell driver (MEDOX E-O) which counts pulses from the APM laser. When a set number of pulses has been counted, the driver sends out trigger pulses to the pockels cell and the Q-switched YAG through channels which have adjustable time delays. By this method, we can obtain less than 10 nsec jitter between the Q-S YAG pulse and the modelocked pulse train. Since the Q-S YAG pulses are 80 nsec long and the gain lifetime is 150 nsec, the timing can be adjusted so that the 10 nsec jitter is inconsequential. It is important to note that even though the APM laser is synchronously pumped, the system timing must be derived from the APM pulses themselves as opposed to the RF of the YLF modelocker. This is because APM lasers (as well as the soliton laser) are passively modelocked and thus run at their own repetition rate, which may differ from that of the pump laser by several kilohertz (see for example ref. [21]).

2.5 Amplifier Construction

The starting point of the amplifier is the gain crystal. NaCl crystals were obtained from the Crystal Growth Facility at Cornell University and from Werner Gellerman at the University of Utah. Crystals from these sources were approximately equivalent in terms of gain and other parameters. The amplifier crystals were approximately 6 x 6 x 20 mm. Such large crystal sizes require considerably longer coloration times (up to six hours) than the one-hour coloration normally used, although the coloration pressure (20 torr) was the same as that used for small crystals.

A crude polishing procedure was used in the first amplifier versions, which produced curved crystal surfaces, resulting in beam distortion. A more refined procedure produces relatively flat surfaces (approximately one wave over the central 70% of the crystal face). This procedure begins with mounting the crystal in a polishing jig (3 inches diameter) which has three NaCl feet. The crystal is then polished using a three-step process:

- 1) shaping with 100 μ m grit emery paper,
- 2) smoothing with 8 μ m polishing paper, and
- 3) polishing with Linde A abrasive and isopropyl alcohol on a polishing pad.

To insure flatness, it is important to minimize the amount of polishing in the final step. Using this method two crystal faces can be shaped and polished in about two hours..

The crystals were held at 77° K in a liquid-nitrogen cryostat available from Fall Creek Instruments, Ithaca, NY. which has a hold time of about 36 hours. Optical access to the crystal chamber is provided through two anti-reflection coated windows. Each crystal was mounted to a copper cold-finger which extends into the crystal chamber. A piece of indium foil was squeezed between the crystal and the cold finger for good thermal contact. Before cooling, the crystals were aggregated for about 40 minutes at room temperature using filtered blue-violet light from a Mercury arc-lamp. After cooling to 77K, the crystals were exposed again to the blue light for a few minutes (this step was accidentally skipped in one of our amplifiers with no apparent ill effects). The side and

top windows provided easy optical access for the auxiliary radiation. For auxiliary radiation, we have tried both a filtered Hg arc-lamp like that used for photo-aggregation, and an air-cooled Argon ion laser (514 nm). We have found that the Argon laser is much more reliable than the Hg-arc lamp.

Several amplifier layouts were considered, most of them based on the designs of existing femtosecond dye amplifiers. However, there are some important differences between dyes and F-centers which make simple duplication difficult, and even undesirable. First, and most obvious, is the necessity of putting the gain medium in a liquid nitrogen cryostat. Second is the moderately long gain lifetime of the NaCl F-centers ($\tau \sim 150 \text{ nsec}$). This has advantages and disadvantages. The main disadvantage is that the gain persists for a time which is much longer than the 13 nsec pulse separation coming from the APM oscillator, resulting in a train of several amplified pulses rather than a single amplified pulse. As will be seen later, a pulse switchout must be used if a single amplified pulse is desired. On the positive side, the long gain lifetime relaxes requirements on the pump-pulse width; i.e., efficient pumping can be obtained with YAG pulses as long as 100 nsec. It also relaxes restrictions on the total trip time through the amplifier. Third, the density of centers is relatively low (approx $4 \times 10^{16} / \text{cm}^3$), which results in lower single-pass gains than are obtained in dye amplifiers of the same length. A 7-mm thick NaCl crystal can produce a small-signal gain of $G=7$ when totally inverted. Contrast this with "bow-tie" dye amplifiers where a single-pass gain of 8 is obtained in a jet which is only 1 mm thick. The lower gain/unit-length means that we must either use thicker crystals or take more passes through the crystal. In practice, it turns out that using thicker crystals is the most advantageous. We should also note that while one can easily manipulate the concentration of dye solutions, this is not easily done with F-centers. So far, attempts to increase the F_2O^+ center densities to greater than $\rho = 1 \times 10^{17} \text{ cm}^{-3}$ have resulted in the formation of colloidal aggregates which act as scattering and absorption centers for the laser radiation.

A noncollinear design, fashioned after the widely used "bow-tie" dye amplifier design [23], was chosen (Fig. 2.6). Several variations of this design were built and tested. The different variations of the amplifier used crystal thicknesses ranging from 6 mm to 20 mm, which is much thicker than the 1 mm jets which are used in bow-tie dye amplifiers. This requires that the beams, while not collinear, must be as close to collinear as possible to avoid clipping the edges of the crystal, and to maximize overlap with the narrow cylindrical pumping volume. The actual amplifier is, therefore, in the shape of a very narrow bow-tie. In Figure 2.6, the angles have been exaggerated for clarity. One guiding principle which we followed was to use relatively tight focusing ($w \sim 300 \mu\text{m}$) for the first two passes through the crystal in order to minimize the beam distortion.

Figure 2.6 Schematic of amplifier system.

During operation the crystal is end pumped by a Quantronix 117 Q-switched YAG laser which produces 80 nsec pulses at $1.064\text{-}\mu\text{m}$ with a pulse energy of 2 mJ, at a repetition rate of 1 kilohertz. (The YAG laser can be operated at repetition rates of up to 10 kHz, however at high repetition rates the amplifier crystal appears to undergo some irreversible damage, perhaps due to heating.) The YAG beam was weakly focussed to a

spot-size of approximately 2 mm in the crystal. Since the gain lifetime is over 150 nsec, it is not necessary to have a very short pump pulse for efficient operation of the amplifier. The 80 nsec pump pulse is sufficiently short to take advantage of the gain storage time. This allows the use of CW-pumped Q-switched YAG lasers which can operate at repetition rates of well over 10 kilohertz. This is an important advantage over flash-pumped Q-S YAG systems which commonly have much lower repetition rates. The higher repetition rates greatly reduce data acquisition time and also enable the use of lock-in techniques for signal averaging. This advantage has been clearly demonstrated with the Copper-vapor pumped dye amplifiers which have repetition rates up to 8 kilohertz.

A noncollinear 3-pass amplifier was built using a 6 x 6 x 20 mm crystal. The use of a longer crystal (20 mm) greatly increased the single-pass gain, and enabled the production of microjoule pulses in as few as two passes. The crystal was end-pumped along the 20 mm dimension (through the 6 x 6 face) by the YAG laser. When both the pump and APM beams were tightly focussed in the crystal, (spot size <1 mm) the single-pass gain could be as high as $G_{sp} = 80$. However, we normally used a larger YAG spot size (~2 mm) which gave a single-pass gain of 30-40. With such high single-pass gains, it is especially important to wedge the crystal faces by a few degrees to prevent parasitic lasing.

Without the pockels cell switchout, the 150 nsec gain lifetime results in an amplified pulse train containing several pulses as shown in figure 2.7(a), while figure 2.7(b) shows a single amplified pulse with the switchout activated (i.e., single pulse injection). Note that the single pulse energy in 2.7(b) is greater than the peak pulse energy in 2.7(a), which indicates some degree of gain saturation. Also visible in 2.7(b) are the neighboring pulses which are suppressed by the switchout, but not entirely. The switchout contrast ratio is limited to 30:1 by the stray birefringence in the LiNbO₃ pockels cell. In the photo, the actual ratio is about 20:1.

Figure 2.7 Amplified pulse train with switchout disabled (a) and with switchout activated (b). Pulse energy in (b) is over $20\mu\text{J}$.

The optical losses of the NaCl crystal were measured in situ in the amplifier, using 1.55 μm light from the NaCl laser. The crystal had an IR transmittance of $T_{\text{IR}} \sim 80\%$. Accounting for the 4% Fresnel losses at the crystal surfaces, this gives an internal transmittance of $T_{\text{bulk}} \sim 87\%$. One might expect that this 13% absorption would be due to the colloidal aggregates which often form during photoaggregation or repeated room temperature handling and thermal cycling. However, we have found that this bulk absorption persists even after the number of aggregates has been reduced by annealing. The origin of this loss is not well understood, but it might be attributable to OH^- ions or other absorbing or scattering defects.

The pump absorption is a useful quantity to measure in order to obtain an upper limit on the possible energy efficiency. It can also give some idea of the maximum attainable single-pass gain in the case where the absorption cross section, σ_a , and emission cross section, σ_g , are roughly equal. This is approximately true for F_2O^+ centers. Then the maximum possible gain coefficient, $\gamma_0 = N_0 \sigma_g$ is almost the same as the small-signal absorption coefficient, $\alpha = N_0 \sigma_a$, for the pump laser, where N_0 is the density of laser-active centers. For this crystal, the small-signal pump transmittance was $T_{\text{YAG}} = 2.4\%$ as measured with a low power CW YAG laser. Assuming that the absorption and emission cross-sections are approximately equal, we would expect a maximum single-pass gain of $G_{\text{max}} \sim 40$. Comparing this with the measured single-pass gain of $G_{\text{sp}} \sim 80$, we obtain absorption and gain coefficients of $\alpha = 1.8 \text{ cm}^{-1}$ and $\gamma = 2.2 \text{ cm}^{-1}$, respectively. Under intense pumping by the Q-S YAG laser, the pump transmittance increases to $T_{\text{YAG}} = 20\%$, indicating a significant degree of pump saturation and inversion.

There are two possible approaches to increasing the total gain experienced by the laser as it passes through the amplifier. The first is to increase the number of passes through the crystal. Aside from the difficulty of aligning many noncollinear passes, this approach proved to be troublesome because inhomogeneities in the amplifier crystal and

slight curvature of the crystal surfaces cause distortion of the beam to increase with each pass. In our experience, it is not practical to make more than four passes through even the most carefully polished crystal, as the beam distortion becomes too great. The best approach to increasing the total gain this problem was to increase the single-pass gain, by lengthening the amplifier crystal, so that fewer passes were needed.

As with NaCl laser oscillators, orientational bleaching can degrade the performance of this amplifier significantly. In NaCl oscillators (such as the APM laser described earlier), orientational bleaching can completely extinguish the laser within a few seconds if some form of auxiliary radiation is not provided. This effect also occurs in the amplifier, but is much less severe due to the much looser focusing. Pump spot sizes in the amplifier are approximately $w_0 = 1\text{mm}$ compared with the oscillator where the spot size is roughly $w_0 = 30\mu\text{m}$. Thus, in the amplifier, fading takes minutes instead of seconds. The repetition rate of the amplifier also influences the fading rate. At 5kHz, fading takes place on the order of thirty seconds, compared to 10 minutes at 1 kHz. This indicates that the severity of fading in the amplifier with respect to the laser may also be reduced by the fact that the amplifier operates at only a few kHz, rather than 76Mhz. As is expected for these color centers, the choice of pump polarization has a strong effect on the gain and the fading. Another technique which minimizes orientational bleaching is to pump the crystal with circularly polarized light. In the present amplifier geometry, this should excite all centers roughly equally, and therefore should not cause orientational bleaching or gain dichroism (at least in theory). What we find experimentally agrees well with the expected behavior for linearly polarized pump light; i.e. there is severe fading in the absence of auxiliary radiation, and strong gain dichroism, even with auxiliary radiation. Pumping with circularly polarized light produces results which partially agree with expectations: both the gain dichroism and the fading are reduced somewhat, but are not completely eliminated. Even under circularly polarized (CP) pumping, the crystal exhibits some gain dichroism, favoring vertically polarized light. Of course, auxiliary radiation greatly reduces the fading

effect in both cases. Electron-trapping might also partially explain the gain fading under CP-pumping with no auxiliary radiation. In one version of the amplifier, the degree of fading decreased after a few days of nearly constant use. Apparently, the crystal underwent some sort of "burning-in" process which is not clearly understood.

We tried several combinations of pump polarization, IR polarization, and auxiliary radiation, each time measuring the transient gain (before fading) and the steady-state gain (after fading). These results are summarized in Table II. The main observations are:

1. Fading is faster and more complete for linear pumping than for circular pumping.
2. The presence of auxiliary radiation is beneficial in all cases.
3. Sustainable gains of $G \geq 30$ can occur under the conditions:
[YAG--Horiz.Pol., IR--Horiz.Pol., Aux--on].
4. A gain of $G = 60$ can be sustained indefinitely for the conditions:
[YAG--Circ.Pol., IR--Vert.Pol., Aux--on].
5. The gain dichroism is strong for linearly polarized pumping (up to a 7:1 ratio), but it can also be strong for CP-pumping (almost 3:1).
6. High gains can be produced transiently (for ~ 3 min.) by suddenly rotating both the pump and IR polarizations by 90° .

YAG polarization	IR polarization	Auxilliary Radiation	Gain per pass (transient)	Gain per pass (sustained)
Horizontal	Horizontal	On	25	25
$\sigma+$	Horizontal	On	20	20
$\sigma+$	Vertical	On	60	60
$\sigma+$	Vertical	Off	60	35
$\sigma-$	Vertical	Off	50	<50
Vertical	Vertical	On	80	40
Vertical	Vertical	Off	80	<20

Table II. Polarization effects on gain.

The auxiliary radiation appears to work by a different mechanism in the amplifier than in the oscillator. This is supported by two observations. First, varying the polarization of the green light does not seem to affect the amplifier gain. This is in stark contrast to the behavior of the NaCl oscillator, where a misalignment of the polarization by only a few degrees can completely extinguish the laser. Second, after fading, the gain can be fully restored by the green light even if the YAG laser is blocked. These two observations suggest that the green light is working via a ground-state absorption (GSA) which is not as polarization sensitive as the excited-state absorption (ESA) to which the reorientation effect is normally attributed. This notion is also supported by the fact that the centers are only in the excited state for 150 nsec out of every 1 msec (for a 1 kHz repetition rate). Thus, the ESA mode of reorientation operates with a duty-factor of 10^{-4} while the GSA mode works constantly. This suggests that much greater efficiency (of

reorientation) could be obtained by frequency-doubling the QS-YAG laser to provide maximum green power during the excitation period. More efficient reorientation would allow us to exploit the higher gain available with linearly polarized pumping.

Microjoule energies were easily obtained with these NaCl amplifiers. Amplified pulse energies, as measured by pulse contrast ratio, were always cross-checked against average power measurements. The pulse energies quoted in this section refer to the peak pulse in the pulse train unless otherwise stated.

Without the switch-out, the amplifier normally produced pulses of 10 μJ energy under the conditions of circularly polarized pumping along with auxiliary radiation. These pulse energies could be sustained indefinitely under these conditions. These energy flux levels ($20 \mu\text{J} / \text{mm}^2$) are near the saturation fluence ($18 \mu\text{J} / \text{mm}^2$) of the F-centers. With single-pulse injection, the amplifier produced up to 25 μJ in a single pulse. Again, the energy flux levels were near near the saturation fluence.

In order to get an estimate of the total extractable energy in the crystal, an oscillator was constructed by placing end mirrors on each side of the crystal. One end mirror was a flat high-reflector (for $1.55\text{-}\mu\text{m}$) and the output-coupler was a flat 55% reflector. The cavity length was 20 cm and the lensing of the crystal surfaces created a stable cavity. Collinear pumping was achieved through the HR end mirror, which transmitted 90% of the pump light. When pumped with 2 mJ, this laser oscillator produced up to 270 μJ , achieving up to 20% quantum efficiency. Considering that the output coupling loss (45%) is only slightly higher than the round-trip parasitic losses (36%), quantum efficiencies of over 30% should be attainable if it is possible to reduce the optical losses in the crystal.

The output energy of this oscillator can be considered to be an upper limit to the extractable pulse energy, and it appears reasonable to expect over 100 μJ output with improvements in the crystals. Such improvements could be achieved either by reducing the losses, or by increasing the F-center density somewhat. Even more importantly, an

improvement in the optical quality (i.e., reducing the beam distortion) would allow more passes through the amplifier. This would both allow for increased efficiency and better optical quality of the output beam.

There are two types of unwanted background radiation which need to be suppressed in high-gain optical amplifiers such as this one: amplified spontaneous emission (ASE), and unamplified background. ASE is especially severe in amplifiers which contain a very high-gain stage (some dye amplifiers have a gain of 1000 in the first stage). ASE is less of a problem in the bow-tie dye amplifiers where each stage has a relatively modest gain in the range of 5 to 8. We would expect this amplifier to fall somewhere between the two.

We attempted to measure ASE by simply blocking the injected pulses to the amplifier and detecting the ASE with the same photodiode used to measure the amplified pulse train. In the early, relatively low gain amplifiers, the measured ASE was so small that it was difficult to detect. The ASE energy (in the direction of the laser beam) was easily less than 1% of the amplified pulse energy. Note that this is a directional measurement (i.e., in the direction of the injected and amplified beam). The total ASE integrated over all directions is not known. The low level of ASE is probably due to the noncollinear geometry and the moderate single-pass gain ($G_{sp} \sim 40$) used in this amplifier. More recent amplifier designs, using newer crystals, have given single pass gains of up to 500. In these amplifiers, ASE and lasing are significant problems. To minimize lasing in the amplifier, we have inserted an optical isolator between the amplifier and continuum generation. We chose this location because the continuum generation cell itself, where the amplified beam is tightly focussed, seems to act as an end mirror in the ASE and lasing process. Although this did somewhat reduce the ASE, we still have as much as half of the energy in this high gain system going into ASE. The ASE itself is not a problem for our experiments as we always operate far from the narrow spectral band where the ASE

occurs. We would still like to reduce the ASE, however, as it uses up useful energy which could otherwise be converted into amplification.

The unamplified pulses comprise a second portion of the background. The amplified pulses are superimposed on the unamplified pulse train from the oscillator. For the amplifier without the switch-out, the unamplified background constitutes 30 to 40% of the average power. The pockels cell switchout significantly reduces this background. The amplified pulse makes up over 95% of the total average power while the unamplified background represents less than 5% of the total average power. Acoustic ringing in the pockels cell (which is particularly severe in LiNbO_3) is responsible for most of this power leakage. If the acoustic ringing were to be suppressed [24], then this leakage could be reduced by roughly a factor of 3 so that it would represent less than 2% of the total average power.

Further suppression of the background requires a saturable absorber. For 620 nm systems, Malachite-green is the absorber dye commonly used. For 800 nm systems, GaAs/AlGaAs Quantum-wells can be used [23] as well as bulk GaAs. For wavelengths near $1.5\mu\text{m}$, bulk InGaAs should be suitable as a saturable absorber. The proper thickness should be between 10 and $14\mu\text{m}$ which should have an optical density of 3 or 4. We had $12\mu\text{m}$ of bulk InGaAs grown (lattice matched) on an InP substrate. Inserting this saturable absorber after the second pass in the amplifier increased the contrast ratio by roughly 2 orders of magnitude, but two-photon absorption in the substrate ($300\mu\text{m}$ thick), created unacceptable losses. It is therefore necessary to thin the substrate as much as possible when using this type of saturable absorber, as is done with GaAs at 800 nm.

High repetition rates are desirable for good signal averaging. The results reported so far were obtained at 1 kilohertz repetition rates. In fact, Q-switched YAG lasers are capable of repetition rates well above 10 kilohertz. However, as the repetition rate is increased beyond 1 kHz, four deleterious effects occur:

1. the pulse energy from the YAG laser begins to roll off,

2. the pulses become longer,
3. the peak-to-peak power fluctuations increase,
4. the pulse timing jitter increases.

This obviously degrades the amplifier performance. For example, at 5 kilohertz, the pulsewidth from the YAG laser has increased from 80 nsec to about 150 nsec. This hurts the pumping efficiency for a gain medium whose lifetime is 150 nsec. The effects are seen in figure 7 which shows both the pump pulses (upper traces in each frame) and the amplified pulse trains (lower traces) at repetition rates of 1 kHz, 2.5 kHz, and 5 kHz. At a repetition rate of 5 kHz, the amplified pulse energies are 5 times smaller than those at 1 kHz. This is due to the combined effects of the YAG laser's decreasing pulse energy and increasing pulse width.

These results do not represent a limit on the amplifier itself, but on the YAG laser used to pump it. Q-switched YAG lasers are commercially available which can generate, at 5 kHz, the same pulse energy (2 mJ) and duration (80 nsec) that our present YAG laser can only achieve at 1 kHz. While the present system can generate continuum at repetition rates up to 3 kilohertz, a simple substitution of YAG lasers would immediately enable continuum generation at repetition rates exceeding 7 kHz. Improvements in the amplifier itself should additionally boost the repetition rate or the pulse energies. Also, the pockels cell switchout limits the repetition rate to a few kilohertz due to the acoustic ringing which lasts about 100 μ sec. Thus, the acoustic ringing must be reduced not only to eliminate the unamplified background, but to increase the repetition rate as well.

The exceptionally broad gain bandwidth of NaCl not only enables femtosecond pulse amplification, it also provides tunability of the entire system. The amplifier has tunability over several linewidths for 100 fsec pulses. As expected, no spectral narrowing occurs. The system can generate continuum from starting wavelengths anywhere from 1.52 to 1.64 μ m. The amplifier tunability can be considered to be greater than this range if submicrojoule energies are desired. Currently, the tuning range is limited not by the

amplifier, but by the APM laser (1.55-1.62 μm) which, in turn, is limited by the birefringent tuning element. With improvements in the APM (and in the amplifier), the limits of the tunability should extend from below 1.50 μm on the short side, to almost 1.7 μm on the long side.

Little or no pulse broadening has been observed in this amplifier. Pulses as short as 100 fsec have been amplified without being measurably broadened. Material dispersion can be a severe problem for femtosecond pulses at visible wavelengths, but at 1.5 μm it is of little consequence for the optical elements typically used. In the amplifier, the beam passes through about 2 cm of glass, and 6 cm of NaCl. The group velocity dispersions (GVD) of glass and NaCl at 1.55 μm are:

$$D_{\text{NaCl}} = .03 \text{ fsec}/(\text{nm})(\text{mm})$$

$$D_{\text{Glass}} = -.015 \text{ fsec}/(\text{nm})(\text{mm})$$

Note that the GVD of NaCl and glass have opposite signs, resulting in a partial cancellation of the total GVD in the amplifier. A simple calculation predicts only about a 15% broadening for 100 fsec pulses due to material dispersion. With the addition of the pockels cell, the pulses travel through an additional 5 cm of lithium niobate, and still no broadening is observed when 140 fsec pulses are amplified.

2.6 Continuum Generation

For the purpose of multiple wavelength spectroscopy, we have generated continuum using the amplified pulses. This was accomplished by focusing the amplified pulses into various thicknesses of glass and other materials. The first and most striking observation was the clear generation of strong visible continuum even though the starting wavelength was 1.55 μm . The emission patterns of the continuum vary depending on the input pulse widths. The continuum has a better beam profile than the pulse used to generate it. Even though the beam from the amplifier is somewhat distorted and is not

diffraction limited, the visible portion of the continuum is emitted in a uniform, circular, diffraction-limited lobe, which indicates that self-focusing is very important under these conditions. The central lobe contains the entire emission spectrum, and is surrounded by a diffuse, multi-colored ring pattern. This ring pattern progresses from red (in the innermost rings) to blue and violet (in the outer rings). All of the above observations obtain when pulsewidths of 120 to 150 fsec are used. However, when pulses as short as 100 fsec are used, the ring pattern no longer appears and the continuum is emitted in a single circular lobe, although the focusing properties of this emission have not been determined. This single-lobed pattern is reminiscent of the continuum generated in ethylene glycol jets by visible or 800 nm pulses.

Continuum could be generated in a variety of media including glass, BaF_2 , sapphire, calcite, and a jet of ethylene-glycol. The continuum threshold energy was about $2 \mu\text{J}$ in the solids, but was almost $5 \mu\text{J}$ in the ethylene-glycol jet. This rather high value for the threshold in ethylene-glycol is probably due to a smaller nonlinearity at this long wavelength, and may also be due to the beam quality. In all of these materials, it was possible to generate continuum for several hours without inducing optical damage. Attempts to generate continuum in silicon, NaCl , CaF_2 , ZnSe , and ZnS resulted in optical damage.

Figure 2.8(a) shows the amplified pulse spectrum and the continuum spectrum from a 3 mm thick piece of BaF_2 . These spectra were obtained by simply moving the BaF_2 in and out of the focus of the lens. Figure 2.8(b) shows the continuum on a logarithmic scale. Approximately 10% of the pulse energy goes into the continuum. Attempts to improve the pump depletion by changing the focus result in optical damage to the BaF_2 .

Figure 2.8 Continuum spectra from BaF₂ (3mm thick). (a) Spectrum of fundamental (dotted) and continuum (solid). (b) Continuum spectrum on a log scale, taken with four separate detectors.

Of all the materials tried thus far, the continuum generated from BaF₂ is the broadest in spectral extent, on both the short and long wavelength sides. The continuum spectrum from BaF₂, shown in figure 2.8, extends from below 400 nm to beyond 3.5 microns. This spectrum is a composite of four separate spectra which were obtained using a variety of detectors (Si, Ge, PbS, InSb) and spectrally dispersing media (glass prism, grating, and silicon prism).

The surprising breadth of the continuum makes this a very versatile spectroscopic tool. The far-reaching infrared continuum should be useful for femtosecond studies of vibronic states in molecules and solids. Nearer the fundamental, the continuum signal at 1.2 μm contains about 2nJ of energy in an 80 nm bandwidth. After dispersing this through a .25 meter spectrometer, it is over 50 times stronger than the intensity needed to saturate an infrared (InGaAs) optical multichannel analyzer (OMA). Of course, the intensity increases as one approaches the fundamental wavelength. The wavelength region of 1.2- to 1.6 μm is of particular interest for the study of InGaAs heterostructures. In the visible

and UV, there is easily enough intensity to use a visible OMA. The flatness of the visible continuum should make it particularly easy to use.

The continuum generated around 1.5 microns by the source discussed in section 2.5 was used to perform several types of experiments on InGaAs samples. For all of these experiments the continuum generated in 1mm of sapphire was split into two paths, forming the pump and probe pulses for the system. The pump and probe were spectrally filtered appropriately, using interference filters from Spectragon Inc. By performing cross correlations with narrow band filters across various regions of the spectrum, we found that the light coming from these filters was not significantly chirped within 200nm, the useful bandwidth of the system. Cross correlations in this wavelength regime have some inherent difficulties, however, due to the nature of the upconversion crystals that must be used.

In our auto and cross correlation set up, we used a 3mm thick POM crystal for upconversion. Although this crystal has very high conversion efficiency, the available crystals are both thick and optically quite inhomogeneous. Thus when performing cross correlations with this set up, we were concerned about the effects of the crystal thickness, group velocity mismatch, and crystal inhomogeneities on the results. We thus performed a second, more spectrally uniform check of the temporal characteristics of the continuum, using two photon absorption in InP.

We initially obtained two photon absorption around 1.5 microns in InP by accident, while performing experiments on InGaAs samples grown on thick (600 micron) InP substrates. We noticed in our first pump probe measurements a broad, spectrally flat increase in probe absorption right around zero time delay. After some puzzlement, we realized that this must be due to a two photon process, in which one photon from the pump beam and one from the probe coincide to produce real absorption at 1.5 microns. The band gap of InP is 1.35eV, so it is normally transparent to 1.5 μ m (800 meV) light. So long as one is well above the band edge, the two photon absorption coefficient of InP

should reflect simply the three dimensional continuum density of states, that is, it should be quite spectrally featureless. Thus, a measurement of the spectrally and temporally

resolved two photon absorption pump probe signal, using a spectrally narrow pump and spectrally broad probe, should give an accurate view of the chirp in the pulses, with sub 100fs resolution. If zero time delay occurs for the whole bandwidth of the probe pulse at one time, (no chirp) the pump probe signal will rise and fall simultaneously across the entire probe spectrum, following the temporal profile of the pump pulse. If however, the probe pulse is substantially chirped, the pump probe signal will peak at different time delays for different wavelengths, and will be seen to sweep across the spectrum in time. Figure 2.9 shows two photon pump probe signals taken for 80nm around 1350 nm. Note the spectral flatness of this signal, which has a temporal width of 150 fs. The chirp on this signal is quite small, about 50 fs over 100nm. This type of low chirp signal was found for all of our operative wavelengths, with one exception.

Figure 2.10 Autocorrelations of 30nm bandwidths selected from continuum. (a) 1470 nm (b) 1535 nm (continuum generation at cell focus) (c) 1535 nm (continuum generation 1mm away from cell focus.)

If wavelengths very close to the fundamental were examined, they were found to be substantially chirped (on the order of 2 fs/nm). For this reason, as well as others noted below, we avoided taking data near the laser fundamental, and tuned the laser appropriately so that our measurements were always made at least 40nm away. Although we did not quantify the accuracy of this technique using known chirps, judging from the temporal and spectral resolution of the system we should be able to resolve a chirp on the order of a 30 fs/200 nm. A final rough check that the continuum was relatively chirp free across 200nm was that a 200nm slice would autocorrelate to about 150fs, showing that it was not significantly temporally broadened from the laser.

In order to establish the duration of the pulses at various wavelengths, we also performed autocorrelations of various spectral slices of the continuum. Autocorrelations at a few representative wavelengths are shown in figure 2.10. Again, we find that the temporal profile of the continuum is fairly well behaved for most wavelengths, reproducing the laser pulse width of 150 to 200fs, with the exception of light very near the fundamental. Very near the fundamental, the duration of the pulses was extremely

sensitive to the amount of self-focussing in the continuum generation cell, varying with intensity and focus from about 200fs to more than 500fs. Once again, this encouraged us to avoid the spectral regime near the fundamental for our measurements.

It should be noted that although the temporal behavior we found for our continuum is quite similar to that generated in ethylene glycol at shorter wavelengths by other workers [25], the physics of the generation is probably somewhat different. This is evident, for example, in the spatial properties of the emission from our continuum cell, which is not evident in the emission from the much thinner ethylene glycol jets used by other workers. It has been recently noted [26] that the mechanism for supercontinuum generation in the case of extended media, such as our 1 mm thick sapphire substrate, must be considered as a combination of the effects of self phase modulation and self focussing. Even in the absence "catastrophic" self focussing, in which there is clear beam collapse, the presence of spatial and spectral as well as phase structure on the pulse must be taken into account. In particular, the presence of the self focussing nonlinearity has been shown to result in a particular conical emission pattern of continuum, in which different wavelengths are emitted at different angular positions. This can be shown to arise from the fact that the full continuum spectrum is generated at all points in the pulse, in contrast to the model of self phase modulation alone, where (for positive n_2) the longer wavelengths are generated at the leading edge of the pulse while the shorter wavelengths are generated in the trailing edge. Another result of the self-focussing is that much less self phase modulation is necessary in order to generate a very broad band continuum. This means that the temporal shift (chirp) between the high and low energy wings of the continuum can be expected to be reduced from that predicted by self phase modulation alone.

2.7 Conclusions

To summarize, we have built and operated a tunable, femtosecond, NaCl amplifier which can produce multi-microjoule pulse energies at repetition rates of over 1 kHz. This amplifier is pumped by a simple, inexpensive Q-switched YAG laser. The pulse broadening in this amplifier is negligible, and the broad gain bandwidth of NaCl provides at least 120 nm of tunability. An extremely broad continuum can be generated in BaF₂ using this system, which is usable possibly beyond 2.5 μm . Additionally the center wavelength of the continuum is tunable, owing to the tunability of the NaCl laser system. Continuum has been generated at repetition rates up to 3 kHz using the present system, and should be easily scalable to beyond 7 kHz by improving the crystals somewhat, or by using a more powerful pump source.

This system is used in a pump broad-band probe experiment on InGaAs samples, in which the probe transmission through the sample is measured as a function of time delay between pump and probe.

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Chapter 3

Linear Absorption

3.1 Introduction

Since the development of techniques for producing atomically precise growth of semiconductor crystals in the late 1970s, it has been known that very thin layers have optical properties different from those of bulk material. One of the first discoveries about so called "quantum well" systems was that their absorption spectra are strongly modified from bulk spectra. Many of these modifications can be predicted using very simple quantum mechanical models. This chapter has two major aims. The first is to introduce the changes in electronic structure which result from confinement, and to discuss the effects of these changes on absorption spectra. This background, which will appear in sections 3.2 and 3.3, will be important later in understanding the modifications in absorption which are caused by the presence of strong, ultrafast, excitation. The second part of this chapter, sections 3.4 through 3.6, will discuss specifically the physics influencing linear absorption in InGaAs/InAlAs heterostructures, and will present a simple analysis of our absorption data in light of the theory. Section 3.5 gives the structural details of our samples.

3.2 Effects of Confinement on Electronic Structure

Strong confinement affects the electronic behavior in a system when the confinement width becomes comparable to the important natural length scales of the system which characterize its quantum mechanical behavior. These include the coherence length for the electrons in the system, as well as the excitonic Bohr radius, a_0 . In this case,

the electronic wave function “feels” the boundary conditions imposed by the barriers, and must adjust accordingly. The treatment of confined carriers can be considered in two parts. First, I will treat the problem of a non-interacting gas of electrons (or holes) in a well of finite width. Although this problem is quite tractable, it only gives a very approximate picture of the effects of confinement in a semiconductor, because the carriers in fact interact strongly via the Coulomb interaction. In the second part of this section I will discuss the effects of confinement on the excitonic bound states of the system, as well as on the unbound, or scattering states in the continuum.

The wave function for an electron in a one dimensional well of depth V_B is a well known problem in introductory quantum mechanics. [1] In the case where the well is infinitely deep, the allowed states of the system are quantized into a series of levels with

energy $E = \frac{\hbar^2 n^2 \pi^2}{2L_z^2 m}$. Here n are non-zero integers, and L_z is the width of the well. The

wavefunctions corresponding to these energy levels are given by $\Psi = A \sin \frac{n\pi}{L_z} z$ for n

odd, and $\Psi = A \cos \frac{n\pi}{L_z} z$ for n even. This is in contrast with the allowed states for an

unconfined system, which are given by $E = \frac{\hbar^2 n^2 \pi^2}{2mL^2}$, where L is now the size of the

crystal. For the case of a macroscopic crystal, L is generally so large that E is taken to be a continuous variable for most purposes. In the case of a thin layered semiconductor, the particles are quantum confined in the z direction and free to move in the x - y plane. Thus we treat the system with the combination of a two dimensional continuum of states and a one dimensional set of quantized states. There are two major effects of this quantization

on the density of allowed states of the system. In three dimensions the density of states

per unit volume, $\rho(E)$, is given by $\rho(\varepsilon) = \frac{m}{\hbar^2 \pi^2} \left(\frac{2m\varepsilon}{\hbar^2} \right)^{1/2}$. In the quantum confined case,

however, we obtain $\sum_n \rho_n(\varepsilon) = \sum_n \frac{m}{\pi \hbar^2} \Theta(\varepsilon - E_n)$, where n are the allowed levels.

These two densities of states are plotted in figure 3.1. For wells of finite depth, of course, the wavefunctions are altered in that they do not go to zero immediately at the edges of the well, but rather leak into the barriers, decaying exponentially inside them. Despite this modification, however, the density of states for a well of finite depth still maintains the steplike structure shown in figure 3.1, although the exact positions of the steps (given by the energies of the bound states) are shifted.

Figure 3.1: Densities of states for two and three dimensions.

Of course, for a finite well there are only a limited number of bound states, given by the number of infinite well states which have energy less than the well depth, V . For any non-zero well depth, however, there is at least one bound state (as is always the case for one and two dimensional systems.)

As mentioned before, the Coulomb interaction plays a critical role in semiconductors, as can be seen immediately from the fact that real absorption spectra never look like the noninteracting densities of states in figure 3.1. The mutual repulsion of

electrons (which can also be thought of as the attraction of electrons and holes) yields a new set of states in the system. Because the Coulomb interaction is always three dimensional (even in the barriers, which for our case have a dielectric constant very similar to that in the wells), the presence of the Coulomb interaction makes it much less simple to decouple the motion of the electrons in the x-y plane and the confined z direction. Let us first consider the influence of the Coulomb interaction in the absolute two and three dimensional cases. [2] In the effective mass approximation, the Hamiltonian for a single interacting electron and hole is exactly that of a hydrogen atom, with the appropriate dielectric constant and effective masses. Thus we obtain for the radial part of the relative motion of the electron and hole a series of Laguerre polynomials. The angular portion of the wavefunction is expressed in terms of the usual spherical harmonics. The center of mass motion is unaffected by the Coulomb interaction and can still be described using plane waves. Using these approximations, several differences between the two and three dimensional solutions emerge. [3] First, the allowed energy states in the two dimensional case are given by $E_n = \frac{-Ry}{(n+1/2)^2}$, where $n=0,1,2,\dots$. This is in contrast to the three dimensional expression, $E_n = \frac{-Ry}{n^2}$, where $n=1,2,3,\dots$ (In both cases Ry is the three dimensional binding energy of the lowest exciton state—approximately 2.5 meV in the case of InGaAs/InAlAs.) Note that this implies that the lowest exciton state is *four times more strongly bound in two dimensions* than in three. In addition, the radial wave functions, $\phi(r)$, for the relative motion of electrons and holes in two and three dimensions differ in such a way that the excitonic Bohr radius is twice as large in three dimensions as in two,

that is $\phi_{3D}(r) = \frac{1}{\sqrt{\pi}} \frac{e^{-r/a_0}}{a_0}$, while $\phi_{2D}(r) = \frac{2\sqrt{2}}{\sqrt{\pi}} \frac{e^{-2r/a_0}}{a_0}$. For physical properties it is often

more important to consider the charge distribution, $\rho(r) \equiv |\phi(r)|^2$, than the wave function, $\phi(r)$. In this case, then, the two dimensional excitonic radius is actually $a_0/4$. This shrinking of the exciton, accompanied by increased binding energy has been measured in a number of systems including GaAs, InGaAs, and CdS, and will prove to be very important in both the steady state and ultrafast behavior of confined systems. As well as affecting the bound states of excitons, confinement also affects the scattering or continuum states. The expressions for the wavefunction in these states are quite complicated.[3] I will discuss the most important results for optical properties in section 3.3.

3.3 Effects of Confinement on Optical Absorption

The absorption coefficient in a material for which the ground state is entirely occupied and all excited states are unoccupied is simply proportional to the product of the probability for a transition between the initial and final states multiplied by the appropriate density of states and summed over all states. The relevant transition probabilities can be found from Fermi's Golden rule, yielding an expression for absorption which is

$$\alpha = \frac{4\pi^2 e^2}{m^2 c \omega \eta} \sum_{i,f} \left| \langle f | \hat{V} | i \rangle \right|^2 \delta(\epsilon_f - \epsilon_i - \hbar\omega). \text{ Here } V \text{ is the perturbation at frequency } \omega$$

which is causing the transition, and η is the real part of the dielectric constant. In the presence of the Coulomb interaction, and assuming that the interband transition matrix

elements, P_{cv} , are relatively k independent, we then obtain for the excitonic absorption

$$\alpha = \frac{4\pi^2 e^2}{m^2 c \omega \eta} |P_{cv}|^2 \sum_n |\phi_n(r=0)|^2 \delta(\varepsilon_{n0} - \hbar\omega) \delta_{kk'} \delta_{K,0}. \text{ Note that it is now clear that the}$$

absorption coefficient is proportional to the overlap of the electron and hole wavefunctions. This implies that the confinement induced shrinking of the exciton will result in an increase in excitonic absorption in two dimensions, and that the Coulomb attraction of the electron and hole increases absorption for both the bound and scattering states of the exciton. In fact, it can be shown that the excitonic oscillator strength is inversely proportional to exciton volume.[4] A summary of some relevant parameters for absorption in two and three dimensions is shown in Table 3.1. Clearly, we have only taken into account the absolute two and three dimensional limits. Complete calculations including both the Coulomb coupling and finite well widths are quite complicated, however some numerical solutions can be obtained. [5,6]. Here $\alpha \equiv \sqrt{\hbar\omega - E_g + Ry}$.

	2D	3D
Binding energy of lowest exciton	4Ry	Ry
$ \phi_n(r=0) ^2$ bound states	$[\pi\alpha_0^2(n-1/2)^3]^{-1}$	$[\pi\alpha_0^3 n^3]^{-1}$
$ \phi_n(r=0) ^2$ unbound states	$\frac{e^{\pi\alpha}}{\cosh \pi\alpha}$	$\frac{\pi\alpha e^{\pi\alpha}}{\sinh \pi\alpha}$
ratio of 1s to 2s absorption	$f_{1s}=27f_{2s}$	$f_{1s}=8f_{2s}$
density of states	$\frac{\mu}{\pi\hbar^2}$	$\frac{\mu}{\pi\hbar^2} (Ry + \hbar\omega - E_g)^{1/2}$

Table 3.1 Parameters for excitonic absorption in two and three dimensions.

3.4 Excitons in InGaAs/InAlAs Heterostructures

In real heterostructures things are not always as simple as discussed in the previous section. Some of the reasons for this are fundamental, and lie in the approximations made in the calculations. Others are more practical, having to do with material quality or details of band structure. In this section I aim to discuss some of these important points for the case of our material system, in order to better understand what sorts of effects we should see in our data.

Among the fundamental shortcomings of the calculations in sections 3.2 and 3.3 was the absence of any account of Fermi statistics for the electrons and holes. In reality we need to use a many particle wave function for the electrons and holes that has been properly antisymmetrized. For the very low densities usually produced in linear absorption measurements this is generally not an important effect, however for high densities such as those encountered in our femtosecond experiments it can not be ignored. Indeed, even though excitons have a total spin of zero, they are composite particles and can only be treated as bosons in the low density limit where the electrons and holes of different excitons are “far” from one another. One can show that the commutator for excitons (similar to that for other composite particles, such as Cooper pairs) consists of a boson like part and an additional density dependent term. That is,

$$[\hat{X}, \hat{X}^\dagger] = \delta_{k,k'} + O\left[(Na_o^3)^2\right].$$

In addition we have assumed that the masses of electrons and holes are identical in the barriers and the wells. As will be discussed further in Chapter 5, this is not, in general, the case. For low lying energy levels in very deep wells, which do

not have much wavefunction leakage into the barriers, this is not a critical consideration. However, for understanding superlattice systems and high lying energy states it is quite important.

There are several more practical issues involved with the study of our material system, InGaAs/InAlAs. First, this III-V zinc blende semiconductor, like many others, has a complicated valence band structure which strongly affects the absorption spectrum. The top of the valence band consists of a $J=3/2$ upper valence band multiplet and a $J=1/2$ lower valence band which is depressed by the spin-orbit splitting. [7,8]. The $J=3/2$ bands are degenerate at zone center, but are separated for k not equal to zero because of the smaller curvature of the $J_z = \pm 3/2$ multiplet compared to the $J_z = \pm 1/2$ one. The $3/2$ band is thus known as the heavy hole band, while the $1/2$ is known as the light hole. In the presence of confinement, the degeneracy of these two bands is lifted by the difference in effective masses, leading to two series of excitons which are energetically split from one another. Away from $k=0$, mixing of these two bands leads to very strong band non-parabolicity, which increases with increasing confinement. In fact, for an infinite well the heavy and light hole bands will cross at $k = \frac{\pi\sqrt{2}}{L_z}$. Of course, band mixing will lift this degeneracy and give an anticrossing, however the very strong non-parabolicity of the bands remains. In particular, this can affect the excitonic absorption. Band non-parabolicity results in an excitonic effective mass which increases with decreasing well width, resulting in an additional shrinkage of the exciton wave function and thus an increase in the excitonic oscillator strength. This causes an excitonic oscillator strength which increases faster with confinement than would be predicted by simple models. [9].

In most of sections 3.2 and 3.3 we treated the case of infinitely deep wells. Clearly this is unphysical, although it is more true for the case of $\text{In}_{0.47}\text{Ga}_{0.53}\text{As}/\text{In}_{0.47}\text{Al}_{0.53}\text{As}$, where the valence and conduction band offsets are 293meV and 440meV respectively, than in GaAs/AlGaAs, where they are almost fifty percent smaller. The finite depth of the well has the greatest effect in the narrowest wells, where leakage of the wavefunctions into the barrier layers is most important.

Very important effects in the InGaAs/InAlAs system originate from the fact that this is an all ternary system. Since pure InAs and pure GaAs have very different band gaps, alloy disorder in this system can have profound effects. In particular, it has been found in spectral hole burning studies [10] that at low temperatures inhomogeneous broadening due to alloy fluctuations is very strong, and results in localization of most excitons. Although multiple quantum well structures also show inhomogeneous broadening due to well width fluctuations, it appears that for our samples alloy broadening is a much stronger effect, as will be discussed in detail in section 3.6.

3.5 Sample structures

Our samples are made of $\text{In}_{0.47}\text{Ga}_{0.53}\text{As}$ wells and $\text{In}_{0.47}\text{Al}_{0.53}\text{As}$ barriers, which I will refer to throughout this thesis as InGaAs and InAlAs. Some important material properties of this system at low temperature are detailed in table 2. Our samples consist of wells ranging in width from 75 to 6000Å, however the total amount of InGaAs is maintained constant at 6000Å by varying the number of wells in the structure, so that

optical measurements on the samples can be directly compared. Barriers between wells are 70Å wide, which for our large band offset (733meV) results in no coupling among wells. The structures are grown lattice matched on semiinsulating InP substrates, and are capped with an InAlAs layer for protection. Before measurements are made the InP is thinned to approximately 50 microns to reduce two photon absorption contributions to the nonlinear signal.

	InGaAs	InAlAs
m_e	.041 m_0	.071 m_0
m_{lh}	.377 m_0	.580 m_0
m_{hh}	.052 m_0	.140 m_0
E_g	.810eV	1.543eV

Table 3.2 Material properties of InGaAs and InAlAs

3.6 Linear Absorption

Data and Models

The linear absorption spectra of all of our samples taken at approximately 10K are shown in figures 3.2(a) and (b). From these data a number of effects are immediately apparent. First, we see the anticipated blue shift of the absorption onset with increasing confinement. This shift should be proportional to $\frac{1}{L_z^2}$ if penetration

Figure 3.2 (a) Linear absorption spectra of four samples, ranging from two to three dimensions.

of the wavefunction into the barrier is minimal. In figure 3.3 the onset of absorption is plotted vs. $\frac{1}{L_z^2}$. Note that although the rise is nearly linear for large well widths, it starts to roll over for the narrowest wells, indicating that for the 100 and 75Å samples, the wavefunctions are starting to penetrate into the barriers substantially. This is also substantiated in figure 3.4, which shows the excitonic oscillator strength (which is proportional to the integrated area under the excitonic peak) versus $1/L_z$. It is known [4] that the excitonic oscillator strength should be inversely proportional to exciton volume. For relatively narrow wells, where the exciton's extension along the z direction is

Figure 3.2 (b): Band edge absorption spectra for (a) 100Å (b) 200Å (c) 500Å and (d) 6000 Å samples.

approximately given by L_z , we thus expect that the excitonic oscillator strength will be proportional to $1/L_z$. Looking at figure 3.4, we find that the oscillator strength follows this linear behavior for well widths between 500 and 100Å. It is expected that the bulk sample will not follow this trend, as in this case the exciton volume is independent of well width. Indeed, the fact that the excitonic oscillator strength is approximately the same for the 500 and 6000 Å wells indicates that from 500 Å up the exciton volume is relatively unaffected by confinement. This makes sense given that the Bohr diameter of bulk excitons is approximately 580 Å, thus wells on this order and larger should not confine the exciton significantly. There is also a deviation from linearity for the narrowest quantum well. In this case, as seen also in the blue shift data, the wavefunctions of the electron and hole are beginning to penetrate significantly into the barrier, so that increasing confinement does not necessarily result in decreasing exciton volume.

The intermediate dimensionality case is particularly interesting since as well width increases the splitting of the heavy hole and light hole bands, as well as the subband splitting, decrease. This can be seen quite clearly in the 500Å sample, in which the subband spacing is reduced to only a few meV, while the heavy-hole light-hole splitting is no longer visible. Thus this sample lies in the intermediate dimensionality regime both in the sense of the continuum structure and in the excitonic confinement.

In binary quantum well systems, such as GaAs/AlGaAs, the major source of inhomogeneous broadening is well width fluctuations. This trend is revealed in increasing excitonic linewidth with decreasing well width. Even at low temperatures, bulk samples are virtually only homogeneously broadened. In ternary systems such as our InGaAs/InAlAs samples, this is not the case. The measured linewidths of the 1s excitons in all samples are approximately 3 meV, virtually independent of well width. This implies that alloy broadening, which is well width independent, is the primary source of inhomogeneous broadening in these samples. In all samples the exciton lines are strongly

inhomogeneously broadened at low temperatures, as will be seen in more detail in the

Four wave mixing experiments discussed in chapter 7.

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Chapter 4

Theory of Pump-Probe and Four Wave Mixing

4.1 Introduction

In order to completely model the behavior of a semiconductor excited by a strong, ultrafast, optical field, it is necessary to have exact knowledge of all of the possible excited states (including free electron hole pairs, excitons, biexcitons, triexcitons, etc.) that can be produced by the excitation. In particular, it is necessary to know all of the many-particle states possible in the system. Since the possibilities for many particle states are essentially endless, some approximations must be made. The approach that is most frequently used is to take a generalized Hartree Fock approximation, in which all of the many particle states are approximated by products of single e-h pair states. [1] In this chapter I will give a general overview of the theory that has been developed based on this approach. The general approach to the problem will be treated in section 4.2. Sections 4.3 and 4.4 will give analytic solutions for four wave mixing and pump probe experiments for the simplest approximation, where Coulomb coupling is neglected. Section 4.5 will discuss some experiments which demonstrate the need to go beyond the Hartree Fock approximation.

4.2 The Semiconductor Bloch Equations

In order to obtain expressions for the time evolution of the population and interband polarization, the single pair states discussed in section 4.1 are treated as an ensemble of non-degenerate two level systems in the density matrix formalism. In order to do this we must take the effective mass approximation, and also assume that the

semiconductor is spatially homogeneous and isotropic. The density matrix for the e-h pair state with momentum k is written

$$4.1 \quad \hat{n}_k(t) = \begin{pmatrix} n_{ck}(t) & \psi_k(t) \\ \psi_k^*(t) & n_{vk}(t) \end{pmatrix}.$$

We take the electron and hole to have the same momentum as they are optically excited and we approximate the photon momentum by zero. The Hamiltonian acting on this ensemble includes both the coupling of the system to the light field in the dipole approximation, as well as the Coulomb coupling among the various k states. Thus the Hamiltonian is written [2]

$$4.2 \quad \hat{H}_k(t) = \begin{bmatrix} \varepsilon_{ck}^0 & -\mu_k E(t) \\ -\mu_k^* E^*(t) & \varepsilon_{vk}^0 \end{bmatrix} - \sum_{k'} V_{k,k'} \hat{n}_{k'}.$$

The Coulomb coupling can thus be thought of as renormalizing the energies ε_{ck} , ε_{vk} of the system, as well as the coupling of the field to the e-h pair. The last equation can be interpreted as if each two level system sees an effective field which includes both the externally applied field and the field produced by the other dipoles in the system which interact via the Coulomb interaction. Thus we can write the renormalized coupling as

$$\Delta_k = \mu_k E_p + \sum_{k'} V_{k,k'} \psi_{k'}$$

and the renormalized energies as $\varepsilon_{jk} = \varepsilon_{jk}^0 - \sum_{k'} V_{k,k'} n_{jk}$. In the

Hartree-Fock approximation the anharmonicity in the pair states which is induced by coupling to phonons and other e-h pairs is treated by including a phenomenological relaxation term in the Liouville equation. In practice, the relaxation will be different for the diagonal (population) and off diagonal (polarization) terms. The Liouville equation

the uncoupled problem for semiconductors, but must always be included directly in calculations. In general, the Coulomb interaction should not be taken as its bare value, but rather should be screened in appropriately. This screening is of course dependent on the population n_k , which increases the nonlinearity inherent in equations 4.4 and 4.5. The scattering rates, γ , contain carrier carrier scattering, as well as carrier-phonon interactions and scattering with impurities. They too are inherently dependent on the density and k space distribution of the population which has been excited. Frequently, as will be the case for the simple calculations presented in this chapter, the γ are taken to be k independent. This is only appropriate when the physical mechanisms of scattering, as discussed above, are relatively k independent, and is questionable as a general assumption. As a result of these complexities, the semiconductor Bloch equations (SCBE) are not in general soluble exactly, but must be solved by numerical techniques. If one goes to the electron hole representation, in which $n_{ck} \rightarrow n_k$ and $n_{vk} \rightarrow (1 - n_k)$, then equation 4.4 can be written (4.6)

$$i\frac{\partial}{\partial t}\psi_k + [i\gamma_{i,k} - \varepsilon_k]\psi_k + \sum_k V_{k,k'}\psi_{k'} = -(1 - 2n_k)\mu_k E_p + 2\left[\sum_{k'} \psi_{k'} V_{k,k'} n_{k'} - \sum_{k'} n_k V_{k,k'} \psi_{k'}\right].$$

The SCBE contain implicitly the nonlinearities which lead to signal in pump probe and four wave mixing experiments. The Pauli-blocking or phase space filling term appears in the first term on the right hand side of equation 4.6. The second term on the right hand side contains nonlinearities induced by carrier-carrier interactions, including screening, and is most important for dense media.

work in the rotating wave approximation, assuming that we are near a resonance, and ignore components originating from the $e^{i\omega t}$ term. In order to obtain general equations for the n th order polarization, we use the perturbation approximation, in which both the density matrix and the macroscopic polarization can be expressed as a sum over the various orders, $\hat{n} = \hat{n}^{(0)} + \hat{n}^{(1)} + \hat{n}^{(2)} + \dots$ and similarly $\langle \bar{P} \rangle = \langle \bar{P}^{(0)} \rangle + \langle \bar{P}^{(1)} \rangle + \langle \bar{P}^{(2)} \rangle + \dots$.

The macroscopic polarization is obtained in terms of the density matrix as the sum over the interband polarizations. We thus obtain expressions for the n th order population and polarization. These are then solved by integrating, one order at a time, starting with the assumption that at thermal equilibrium everything is in the ground state, so that we know the zero order population. From the zero order population we can integrate to find the first order polarization, and thus the second order population, and finally the third order polarization which leads to the four wave mixing signal. The result has four components, which propagate along k_1 , k_2 , $2k_2 - k_1$ and $2k_1 - k_2$. Selecting the only those components of the result which propagate in the direction $k_3 = 2k_2 - k_1$, we arrive at the result for the third order interband polarization

$$\begin{aligned} \psi^{(3)} = & -2in^{(0)} \left(\frac{\mu}{\hbar} \right)^3 e^{[-(\gamma + i\Delta\omega) + ik_3 r]} \times \\ & \left\{ \int_{-\infty}^t \int_{-\infty}^{t'} \int_{-\infty}^{t''} \left\{ E_2(r, t') E_2(r, t'') E_1(r, t''') \exp[(\gamma - \gamma_t)(t' - t'') + \gamma_t t''' + i\Delta\omega(t' + t'' - t''')] \right\} dt''' dt'' dt' \right. \\ & \left. + \int_{-\infty}^t \int_{-\infty}^{t'} \int_{-\infty}^{t''} \left\{ E_2(r, t') E_1(r, t'') E_2(r, t''') \exp[(\gamma - \gamma_t)(t' - t'') + \gamma_t t''' + i\Delta\omega(t' - t'' + t''')] \right\} dt''' dt'' dt' \right\} \end{aligned}$$

In order to obtain an analytic result for this case, we can consider the case of electric fields which are much shorter in time than either of the relaxation times, and can be treated as delta functions in time. This yields, for the case of homogeneous broadening,

$$P^{(3)}(r, t) = -in^{(0)} N \mu \theta_1 \theta_2^2 \exp[ik_3 r - \gamma_i(t - t_1)]$$

for $t > t_2$, and zero for $t < t_2$. Here θ_1 and θ_2 are the pulse areas of the first and second pulses, and t_1 and t_2 are their arrival times. N gives the total number of dipoles in the ensemble. The case of inhomogeneous broadening can be obtained by summing over a gaussian distribution of oscillators with width $\delta\omega$. In this case the result for the third order polarization in the k_3 direction is

$$P^{(3)}(r, t) = -in^{(0)} N \mu \theta_1 \theta_2^2 \exp\left[ik_3 r - \gamma_i(t - t_1) - \frac{1}{4}(t - 2t_2 + t_1)^2 (\delta\omega)^2\right]$$

for $t > t_2$, and zero for $t < t_2$. Note that in the homogeneously broadened case, the polarization is emitted immediately after the arrival of the second pulse, whereas in the inhomogeneously broadened case, the polarization appears as a "photon echo", peaking at time $t = t_2 + (t_2 - t_1)$, assuming $t_2 > t_1$. In ordinary FWM measurements, a slow detector is used to integrate the total intensity emitted along k_3 , as a function of time delay between t_1 and t_2 . Integrating the previous results, we obtain for the signal on the slow detector

$$J = A \exp[-4\gamma_i(t_2 - t_1)] \left\{ 1 + \Phi\left[\frac{\delta\omega}{\sqrt{\pi}}(t_2 - t_1)\right] \right\} \text{ for inhomogeneous broadening, where}$$

$$\Phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-t^2} dt. \text{ For homogeneous broadening integrated intensity is}$$

$$J = B \exp[-2\gamma_i(t_2 - t_1)]. \text{ In both cases the signal is non-zero only for } t_2 > t_1.$$

Thus we obtain the result that the decay of the FWM signal depends essentially only on the transverse relaxation time for the approximations discussed above. Although this is a very useful result, more recent work on semiconductors has revealed that the situation is considerably more complicated. When the Coulomb coupling term, V , is not taken to be zero, as is indeed necessary for any real semiconductor, a number of effects are observed which deviate from the simple relaxation behavior. For excitation resonant with the exciton, exciton-exciton interactions cause a component at negative time delays, as well as a polarization wave scattering term which produces a peak in $P^{(3)}$ away from zero time delay even in the case of homogeneous broadening.[4,5,6] Although some modeling has been done for this case [7], it is numerical in nature and is thus too complex to reproduce here. It should be noted, however, that with increased spectral and temporal resolution more and more deviations from the simple uncoupled model are being found. Several of these are discussed at greater length in the context of our results of Chapter 7.

4.4 Pump-Probe Measurements

With the advent of broadband, ultrafast sources (generally from continuum generation) the technique of spectrally resolved pump-probe measurements to examine energy relaxation in semiconductors has become quite popular. In this technique a strong, narrow band pump excites the system, which is then probed by a weak, broadband probe beam. The probe beam thus interrogates the changes in absorption induced by the strong pump. The spectrum of the probe beam transmitted through the sample is then measured

as a function of time delay between the pump and the probe. In order to describe this type of measurement with the SCBE, one writes the electric field in two parts, the strong pump field E_p propagating along k_1 , and the weak probe field E_t propagating along k_2 . The strong pump field gives rise to an excited state whose dynamics are described by the SCBE, while the probe field yields the linear excitation spectrum of the excited state. For the case of uncoupled two level systems we start with the identical dynamical equations as was the case for the FWM experiment, however we excite the system with only one beam and then solve for $P^{(3)}$ which propagates along the probe direction, k_2 . In this case when we solve the SCBE iteratively we are interested in obtaining terms out to second order in the pump and first order in probe. Using this approach, the polarization $P^{(3)}$ which propagates in the probe direction is found to be [8]

$$4.7 \quad P^{(3)} = N_0 F_2(t) \otimes \left\{ \begin{aligned} &E_2(t) [F_1(t) \otimes (E_p(t) \cdot P_p^*(t) - E_p^*(t) P_p(t))] \\ &+ E_p(t) [F_1(t) \otimes (E_t(t) P_p^*(t))] \\ &- E_p(t) [F_1(t) \otimes (E_p^*(t) P(t))] + E_t(t) \end{aligned} \right\}$$

$$\text{here we have defined } F_1(t) = \begin{cases} 0(t < 0) \\ \frac{2i\mu}{\hbar} \exp(-\gamma t)(t > 0) \end{cases} \text{ and}$$

$$F_2(t) = \begin{cases} 0(t < 0) \\ \frac{i\mu}{\hbar} \exp(-\gamma t) \exp(-i\Omega t)(t > 0) \end{cases}. \quad \Omega \text{ is the detuning from the pump frequency,}$$

and N_0 is the initial population difference between the ground and excited states. P_p and P_t are the first order polarizations induced by the pump and probe fields respectively, and

are calculated via $P_p(t) = F_2(t) \otimes E_p(t)$ and $P_t(t) = F_2(t) \otimes E_t(t)$. The symbol $\otimes$ denotes convolution and is defined by $F(t) \otimes E(t) \equiv \int dt' F(t-t')E(t')$.

The measurement which is usually made in spectrally resolved pump probe experiments is the difference in the absorption spectra of the sample in the absence and presence of the pump beam. The absorptive response of the system in the frequency domain is proportional to the imaginary part of the susceptibility, $\chi(\omega)'' \equiv \text{Im} \frac{P(\omega)}{E(\omega)}$. This is obtained by Fourier transforming equation 4.7, and dividing by the spectrum of the probe signal, $E_t(\omega)$. This results in a differential transmittance signal which is given by

$$4.8 \quad \frac{\Delta T}{T} = -\text{Im} \left\{ \frac{f_2(\omega)}{E_t(\omega)} \Im \left[\begin{array}{l} E_t(t) N_p^{(2)}(t) \\ + E_p(t) [F_1(t) \otimes (E_t(t) P_p^*(t))] \\ - E_p(t) [F_1(t) \otimes (E_p^*(t) P_t(t))] \end{array} \right] \right\}.$$

Here $N_p^{(2)}(t)$ is the second order population induced by the pump field, given by $N_p^{(2)} = F_1(t) \otimes [E_p(t) P_p^*(t) - c.c.]$. $\Im[A(t)]$ is the Fourier transform of $A(t)$, while $f_2(\omega)$ is the Fourier transform of $F_2(t)$.

The interpretation of equation 4.8 is, in fact, quite straightforward. The first term is the level population term. It is proportional to the real population changes induced by the pump which are still present when the probe arrives. It decays with the energy relaxation rate, γ , so that only the population distribution which exists at the instant of the probe's arrival contributes. It is this term which allows us to follow the evolution of the energetic distribution of carriers as they thermalize among themselves and cool to the lattice temperature. This term is non-zero only when the pump precedes the probe.

The second term reflects the coupling between the pump-induced polarization and the probe field. It is non-zero only when there is temporal overlap of the pump and probe, right around zero time delay. It reflects the changes in population created by the interaction of the pump induced polarization with the probe field. The pump field then interacts with this population modulation to create a third order polarization which emits along the probe direction.

The third term is the perturbed free induction decay term. It occurs because the broadband probe field induces a coherent polarization in the material which decays at the transverse relaxation rate, γ_t . This free induction decay, which is the same as the one probed in four wave mixing experiments, is perturbed by the arrival of the pump pulse, which shifts the resonances of the system. This term exists only when the probe pulse precedes or overlaps the pump pulse, that is, for negative time delays, and it rises as the time delay goes to zero with a time constant $1/\gamma_t$ for a homogeneously broadened system. For a narrow resonance this term causes spectral oscillations with a period given by $1/\Delta t$.

We see, then, that for different time delays different terms contribute to the pump-probe spectra. In general, pump probe experiments are used to measure population dynamics, which are reflected in the first term of equation 4.8. This is a valid interpretation of the pump probe data for time delays in which the pump precedes the probe by more than one pulse-width. For these positive time delays, only the population term contributes to the signal. For negative time delays and delays very near zero, however, we see that the two other contributions to the signal are important, and dynamics observed in the spectra can no longer be directly interpreted in terms of

population evolution. As seen in chapters 5 and 6, all of three contributions to the pump probe signal are visible in our data.

As in the case of four wave mixing, the addition of Coulomb coupling among the two level systems in the model is clearly necessary to obtain quantitative agreement between theory and experiment. This can be done, and it results in additional contributions to all three of the signals discussed above. Because there are such a large number of dynamic processes contributing to pump-probe data, however, it is not in general possible to use fitting of pump probe signals to obtain precise measures of the coupling among two level systems, as was done in FWM by Bigot et.al.

4.5 Beyond the SCBE

In recent years more detailed measurements using both pump probe and four wave mixing techniques have revealed a number of results which can not be explained by the semiconductor Bloch equations. These include measurements of pump probe and FWM signals derived from two photon absorption and emission measurements [9]. Two photon and higher order processes are explicitly excluded in the approximations of the SCBE, however these two photon signals can be quite strong. Measurements of semiconductors under strong magnetic field have also revealed dynamics which can not be predicted under the SCBE. These are found for the case of strongly non-Lorentzian resonances, in which the Coulomb coupling between a bound state and a set of continuum states strongly modifies both linear absorption and dynamical properties. [10] It is clear that models

which go beyond the semiconductor Bloch equations will be increasingly important in the future, when the availability of increasingly short (sub 10fs) pulses will allow for further examination of processes which are non-Markovian in nature. In this case, the assumption of a single scattering parameter, γ , is not appropriate, as "memory" effects must be included.

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Chapter 5

Carrier Relaxation Dynamics

5.1 Introduction

In this chapter I will address pump-probe experiments we performed in which the InGaAs heterostructures were excited well above the band edge, but with insufficient excess energy to emit LO phonons. This sort of excitation allowed us to study the effects of confinement on a number of ultrafast processes. The evolution of the population is

sketched out in figure 5.1. First, the sharply peaked, non-thermal population (a) thermalizes through Coulomb mediated carrier carrier scattering to a hot Boltzman distribution (b). This process takes about 250fs and is discussed in section 5.2. Next, the hot carriers begin to cool to the lattice temperature by the emission of phonons, eventually resulting in a Fermi distribution in equilibrium with the lattice. The majority of carriers have insufficient excess energy to emit LO phonons, and so must cool by the much slower acoustic phonon emission process. Thus, cooling takes several hundred ps. It is discussed in section 5.6. Our measurements also allow us to observe the influence of confinement on a number of transient nonlinearities. Section 5.3 addresses the influence of confinement on the phase space filling nonlinearity. Section 5.4 addresses the transient fermi edge singularity, which produces transient nonlinearities near the excitation energy at very early

times before the carriers have thermalized, while section 5.5 treats transient nonlinearities at the band edge.

Understanding of carrier dynamics is particularly important in InGaAs because of its role as a material system for optoelectronic devices. Optimization of these devices depend on a thorough knowledge of these dynamics, as they provide the fundamental speed limits for communication systems. Measurements of carrier dynamics in semiconductors have thus far focused on GaAs, for which systems generating broadband ultrashort optical pulses are readily available. In contrast, InGaAs has not been carefully studied due to the lack of such systems in the region of 1.55 μm . There have been several single-wavelength, $\sim 100\text{fs}$ time-resolved measurements of the optical properties of InGaAs (bulk) [1], and quantum wells (QW) [2,3], and of InGaAs-based active devices such as QW traveling wave amplifiers [4, 5]. However, because in these experiments only a single energy is probed, the motion of populations in phase space cannot be followed and the effects they induce at other energies cannot be observed.

For our measurements, the central energy of the pump photons was chosen to be approximately 30 meV above the lowest bound exciton state. Accounting for the electron and hole effective masses, ($m_{hh} = 0.377 m_0$, $m_{lh} = 0.052 m_0$, $m_e = 0.041 m_0$) this gives an initial electron distribution with an excess energy of approximately 26 meV, and a hole distribution with excess energy approximately 3 meV. InGaAs has two LO phonon branches, the GaAs-like, with energy of 35 meV, and the InAs-like, with energy 28 meV. We thus expect all of the holes and the majority of the electrons to have insufficient energy to emit LO phonons. Therefore they will thermalize among themselves and cool to the lattice temperature largely *without* LO phonon emission. Early thermalization should be dominated by carrier-carrier scattering, while at late times the majority of electrons will cool via acoustic phonon emission. The absorption spectra for our samples show that at our excitation energy, the density of states for the electrons is very similar, $\sim 1 \times 10^{16} \text{ meV}^{-1} \text{ cm}^{-3}$ for the bulk, and $\sim 2 \times 10^{16} \text{ meV}^{-1} \text{ cm}^{-3}$ for the QWs, accounting for the well

widths. From the absorption spectra and the depth of the spectral hole we estimate the excitation density for all samples to be roughly $N=5 \times 10^{16} \text{cm}^{-3}$ distributed over approximately 10 meV.

5.2 Coulomb interactions and carrier thermalization

Population thermalization by carrier-carrier scattering has been examined both theoretically and experimentally in a number of studies. As it is a scattering process, its efficiency is governed by two major factors: the strength of the scattering interaction, and the phase space available for scattering.[6] The strength of the Coulomb interaction which governs this scattering is of course critically dependent on screening. Accurate models of screening for non thermal populations, however, are very difficult to develop for a number of reasons. First, screening models are strongly dependent on accurate modeling of the k space distribution of the carriers involved. Simple models generally take either a Fermi or a Boltzmann distribution, leading to the Thomas Fermi or Debye Huckel approximations for static screening. For the case of thermalization, however, the population distribution is evolving on a very short time scale, and can not be accurately modelled by any simple functional form. Furthermore, the tractable models for screening generally take a static model, in which the rearrangement of carriers to most effectively screen the potential has no internal dynamics. On the time scales of our experiment, this is clearly not a good approximation. Despite these inherent difficulties, a number of approaches have been used to model carrier-carrier scattering. Perhaps the most successful of these have been the Monte Carlo simulations [7]. Although these still maintain a static, random phase approximation type screening model, they do include appropriate carrier distributions which change in time in a self-consistent way. That is, they model the two particle scattering rate based on the instantaneous one particle distribution function. In general it is assumed that the most important scatterings for carrier thermalization are electron-electron collisions, as particles with identical masses scatter most inelastically, and the

holes are excited with very little excess energy. Experimental work in n- and p- doped GaAs samples have tended to lend support to this view. [8] Monte Carlo simulations based on these assumptions have given qualitative agreement with many aspects of experiments in GaAs quantum wells. In particular, the thermalization times are approximately correct. Such models are limited in their application to our problem, however, as they are only tractable in the absolute two and three dimensional regimes, and cannot account for wells of finite thickness. In addition, these studies do not include the effects of valence band complexity, or of many body effects such as correlations and excitonic effects.

Despite the difficulty in making predictions about the details of carrier thermalization, it has generally been assumed that carrier thermalization should occur faster in quantum wells than in bulk samples. This is due to the expectation that the efficiency of screening is reduced in two dimensions. A simple argument for this reduction can be seen in figure 5.2. Although the densities of carriers are equivalent in the

bulk and quantum well samples, the carriers in the bulk are free to move to screen a test charge, while those in the quantum well are limited in their motion. This argument depends critically on the fact that for samples such as ours the barriers and the quantum wells have very similar dielectric constants, so that while the carriers are confined to the wells, the electric field lines are essentially three dimensional. An example of this effect can be seen for the case of Debye-Huckel screening in two and three dimensions. In three

dimensions the screening wavevector κ has the form: $\kappa = \left(\frac{6\pi e^2}{\epsilon_0} \frac{N}{\epsilon_f} \right)^{1/2}$, where the screened Coulomb potential is given by the Yukawa form, $V_s(r) = \frac{e^2}{\epsilon_0 r} e^{-\kappa r}$. In two dimensions, however, above a critical density the screening wavevector saturates at a finite value, $q_0 = \frac{2me^2}{\epsilon_0 \hbar}$, where the dielectric constant is given by $\epsilon(q, 0) = 1 + \frac{q_0}{q}$. It should be noted that q_0 is inversely proportional to the excitonic Bohr radius, which leads to the interpretation that in two dimensions, once excitons are closely packed screening is maximized and can not increase any further. For the two dimensional case, q gives the component of momentum in the quantum well plane. Thus, at high densities screening is seen to be more efficient in three dimensions than in two.

The other major influence on the carrier carrier scattering rate is the density of final states available for scattering. For our experiments, at least for the early, high energy scatterings the density of states is very similar among all our samples. Thus, we do not expect a very strong influence due to this effect.

Studies in GaAs have shown that thermalization is very dependent on the density of carriers, as would be expected for a Coulomb mediated process in which screening is important. In GaAs quantum wells, pump/broadband probe studies have shown that this Coulomb mediated thermalization occurs very fast, within 100fs or less for carrier densities on the order of 10^{10} cm^{-2} . [9] Studies have also been performed on bulk GaAs [10], which found time constants for thermalization approximately twice those found in quantum wells at similar densities, in qualitative agreement with theoretical predictions. Since these experiments were performed with somewhat different techniques and on samples of different quality, however, direct comparison of their results is questionable.

In our measurements we examined both detailed dynamics at a single wavelength as well as full spectral information to try to extract the effects of confinement on carrier thermalization. In order to better understand the data, it is necessary here to briefly

review the signatures of various physical effects. As presented in Chapter 4, when a population exists at a particular energy in the conduction and valence band of the semiconductor, it results in a decrease in absorption at that energy due to phase space filling. In addition to this phase space filling nonlinearity, the absorption spectrum is also affected by changes in oscillator strength due to screening. At very early times coherent effects also influence the signal, as discussed in Chapter 4, however I reserve discussion of these until section 5.5. Thus, in our experiment, the signal can be expected to evolve as follows (see figure 5.1) When the carriers are first injected high in the band, there will be a phase space filling signal at the excitation energy, reflecting the nonthermal electron-hole plasma (hereafter known as the spectral hole). In addition, there will be a signal at the exciton reflecting screening of the exciton by the nonthermal population, which broadens the exciton and reduces its oscillator strength. Once the carriers have thermalized to a hot, Boltzmann distribution, the spectral hole will have dissipated, and the signal at the band edge will be increased, as it now reflects phase space filling of the states that make up the exciton as well as screening of the exciton by the thermalized population. The hot distribution will have a temperature given by the excess energy with which the distribution was initially excited, well in excess of the lattice temperature. (It should be noted that the time for formation of excitons from band edge free carriers is very long, on the order of hundreds of picoseconds, because it requires acoustic phonon emission.) Finally, as the carriers cool to the lattice temperature the signal at the band edge will again increase, as the population which overlaps the excitonic states increases.

We thus expect the evolution of the signals at the band edge and spectral hole at early times to reflect the carrier thermalization dynamics. Figure 5.3 shows the arrival of carriers at the band edge for the bulk and 100Å quantum well samples, over 1.4ps around $\Delta t=0$. Contrary to predictions that the thermalization should be substantially faster in two than in three dimensions, the two signals are essentially identical, giving a thermalization time of approximately 250fs. The time

resolution of the system is about 100fs, so differences between the two thermalization times should be resolvable. (Indeed, we are able to resolve the effects of changing carrier density on the carrier thermalization time with this method.) Similar measurements taken on samples with 150, 250, and 500Å well widths give the same results. This indicates, then, that for densities around 10^{16}cm^{-3} the rates of carrier-carrier scattering by the nonthermal population are not significantly different in two and three dimensions. Recalling that the density of states at the excitation energy is similar among all the samples, the equivalence of carrier-carrier scattering rates implies that the Coulomb interaction among the non-thermal carriers is equally effective in two and three dimensions. This contradicts the prediction that the Coulomb interaction should be weaker in three dimensions than in two, due to the increased effectiveness of screening. This surprising result can be confirmed by considering the two other forms of data. First, we can look at the full spectral profiles taken for 250fs around $\Delta t=0$. (Figure 5.4). Looking at the time for the narrow spectral holes to dissipate, we again see similar times for all samples of about 250 fs. As the carriers thermalize among themselves they assume a Boltzmann distribution. Thus, in addition to the carriers which

Figure 5.4 Time evolution of the differential absorption spectrum. (a) 100 Å quantum well (b) bulk.

scatter towards the band edge, some of the carriers scatter up into the band, forming the high energy tail of the distribution. A final approach to measuring the carrier-carrier scattering time is thus to examine the time necessary for carriers scattered to the high energy side of the spectral hole to arrive in the tail of the distribution. This turns out to be an extremely sensitive measure of the carrier thermalization time, which can detect small differences in rate caused by a factor of three change in carrier density. Using this measure, we again find that the carrier thermalization, and thus efficiency of screening among nonthermal carriers, is not influenced by well width. Since we do not expect the dimensionality dependence of screening to depend strongly on which carriers are being screened, we believe that for our conditions the efficiency of screening of the exciton by the optically excited carrier distribution is also relatively independent of well width.

5.3 Phase Space Filling

Having concluded that screening of the Coulomb interaction does not seem to be strongly affected by well width, we can move on to study the other major influence on the band edge signal, the phase space filling nonlinearity. Phase space filling originates in the Fermionic nature of the carriers, which requires that no two carriers can occupy the same state at the same time. Thus, if a state is already occupied, optical absorption to that state is forbidden, or more mathematically, the absorption in the presence of carriers is given by $\alpha = \alpha_0(1 - f_e - f_h)$, where f_e and f_h are the electron and hole occupancies, and α_0 is the absorption strength in the absence of carriers. The phase space filling nonlinearity, Δ_{psf} , is obtained from the definition of the respective oscillator strengths of the exciton with and without the presence of carriers. In the approximation where the interband matrix elements are taken to be independent of k , the oscillator strength is given by [11]

$$5.1 \quad f_n = 4\pi e^2 |r_{cv}|^2 |\phi_n(r=0)|^2 \left\{ 1 - \frac{1}{\phi_n(r=0)} \sum_k (f_e + f_h) \phi_n(k) \right\}$$

Here ϕ_n are the excitonic wave functions and r_{CV} are the interband dipole matrix elements. This leads immediately to the result for the change in oscillator strength due to phase space filling

$$5.2 \quad \frac{\Delta f}{f} = \left\{ \frac{1}{\phi_n(r=0)} \sum_k (f_e + f_h) \phi_n(k) \right\}.$$

We are interested in particular in the effects of phase space filling by a hot carrier distribution. At time delays longer than 300 fs, we know (section 5.2) that the carriers have already thermalized to a Boltzmann distribution. Carrier cooling through acoustic phonon emission does not begin to become significant until at after at least 1ps of time delay. Thus, the phase space filling contribution to the band edge signal for times between approximately 300fs and 1ps should be due to a thermalized distribution which has not yet started to loose energy to the lattice. To calculate this contribution, we need to introduce into equation 5.2 the k space expressions for the excitonic wave functions and the hot carrier distributions in two and three dimensions. For the two dimensional case,

$$\phi_{1s}(k) = \frac{\sqrt{8\pi\Lambda^2}}{(1 + \Lambda^2 k^2)^{3/2}}. \text{ In three dimensions } \phi_{1s}(k) = \frac{\sqrt{64\pi\alpha_0^{3/2}}}{(1 + \alpha_0^2 k^2)^2}. \text{ Here } \Lambda = a_0/2. \text{ Since}$$

the electrons we excite have 30 meV of excess energy, it is legitimate to approximate their distributions with Boltzman distributions. The holes have very little excess energy, so their distributions hardly change with time, and can be approximated as Fermi-like. After substituting and integrating, we obtain for the electron contributions to phase space filling, $S_e = (\pi\alpha_0^2 N_e) 2\alpha_{e2D} I_{2D}$ in two dimensions, and $S_e = \alpha_0^3 N_e 8\pi^{1/2} \alpha_{e3D}^{3/2} I_{3D}$ in three dimensions. Here $\alpha_{e2D} = \frac{m}{m_e} \frac{4E_0}{kT}$ and $\alpha_{e3D} = \frac{m}{m_e} \frac{E_0}{kT}$. Here the I are integrals over error

functions (see appendix 1) which can be evaluated explicitly for any given carrier density and excess energy. For 10^{17}cm^{-3} carriers with an excess energy of 30 meV, we find that phase space filling is predicted to be approximately *twice* as effective in two as in three dimensions.

In order to compare this result with our experiment, we recall that at very early time delays, when the carriers are in a nonthermal distribution, the signal at the band edge is due entirely to screening. Once the carriers have thermalized, phase space filling by the hot population contributes to the signal as well as screening. Thus, by comparing differential absorption spectra around zero time delay with those at 500fs time delay, we can get an estimate of the relative importance of screening and phase space filling in two and three dimensions. In section 5.2, we showed that screening does not appear to be very strongly affected by confinement for our carrier densities. Thus, a comparison of early to late band edge signals for different well widths gives a measure of the effect of confinement on the strength of phase space filling. Figure 5.5 shows DAS of the 100Å quantum well and bulk samples taken at zero time delay and at 500fs. In comparing the band edge signals, we find that as predicted by our model, PSF appears to be approximately twice as effective in the quantum well as in the bulk. This is in direct agreement with our calculation. We have measured this ratio in intermediate dimensionality samples, and find a smooth extrapolation from the two dimensional to the three dimensional limit. This is one of the first measurements of the evolution of many body effects from two to three dimensions.

5.4 Transient Fermi edge singularity

Figure 5.6 shows a close-up of DAS at time delays of zero and +33fs. The dotted line shows the pump spectrum used for each experiment. For all the samples, it can be seen that the spectral hole is red shifted from the pump spectrum by approximately 5meV. This shift is accompanied by a broad induced absorption on the high energy side of the pump. This effect is known as the transient Fermi edge singularity, and has been observed before in both pump-probe and four wave mixing experiments in GaAs. [12,13,14]. The origin of this effect is most easily understood by comparison with the Fermi edge

Figure 5.5 Differential absorption spectra for zero time delay (dotted) and +500fs (solid). (a) 6000Å sample (b) 100Å sample.

singularity [FES] and Mahan exciton which are well known from linear spectroscopy of doped samples [15] and from x-ray spectroscopy of atoms [16]. When n-doped samples are weakly excited, the electron Fermi sea is attracted to the photogenerated holes, resulting in a very weakly bound state in which many electrons orbit one hole. This is known as the Fermi edge singularity. It is very strongly dependent on the sharpness of the Fermi sea, and thus is only measurable below 10K and in very pure samples. (The singularity is rarely seen in p-doped samples, as it depends on the minority carriers being very heavy. Indeed, many models of the FES use holes of infinite mass.)

Figure 5.6 Differential transmission spectra at zero time delay (dashed) and +33fs (solid) with pump spectra (dotted) for (a) 100Å well (b) 200Å well (c) 500Å well (d) bulk.

When a sharp population distribution is excited high in the band with a relatively narrow pump pulse, such as ours, a similar effect occurs. Figure 5.7 shows a schematic of the process. Again, we are concerned primarily with the distribution of electrons which is created high in the band, as the distribution of holes is virtually at the band edge. As the

electron distribution is created high in the band, it can be thought of as the sum of electron and hole Fermi seas. (see figure 5.7) These distributions then perturb the probe absorption just as would carriers due to doping. Photogenerated holes are attracted to the electron Fermi sea, resulting in an increased absorption just above the non thermal distribution. The hole Fermi sea, on the other hand, repels photogenerated holes, causing a decrease in absorption below the excitation energy. The net result is a red shift of the spectral hole.

This shift is thus due to a coulomb mediated many body process, and might be expected to be affected by confinement due to the changes in screening. Consistent with our thermalization measurements, however, at these excitation densities very similar shifts are seen for all of our samples, implying again that screening is not strongly affected by confinement.

5.5 Transient band edge nonlinearities

Several other nonlinearities are seen near the band edge for times very close to zero time delay. In all samples, the band edge signal builds up from a small signal which is present well before $\Delta t=0$. The majority of this signal is due to the coherent contribution to the pump probe response (see Chapter 4). For above resonance excitation, this coherent contribution produces a signal which is negative below the resonance and positive at and above the resonance. As discussed in section 4.4, this signal should rise with a time constant T_2 coming in from negative time, and then fall away with the overlap of the pulses. This is clearly the case for the bulk sample, as shown in figure 5.8. In the 100Å and 150Å quantum well samples, however, a negative signal below the exciton persists out for about 400fs, well beyond the overlap of the pump and probe pulses. In these samples, as discussed in Chapter 3, the excitons are very strongly bound, yielding large oscillator strengths. These clearly defined excitons undergo collisional broadening due to the electron hole plasma excited by the pump. We believe it is this incoherent collisional broadening which causes the longer lived negative signal. Although the weaker excitons in the wider samples are almost certainly broadened as well, due to the fact that the excitonic resonances are not as clearly defined collisional broadening is harder to see in these samples.

Band gap renormalization (BGR), the reduction in band gap due to the screening of the e-h correlation at the band edge states by the free carriers high up in the band, should also be visible in the bulk sample. BGR in this sample would appear as an induced absorption below the original band gap at early times. At later times, redistribution of the carriers in renormalized bands would partially fill the states near the band edge, reducing the induced absorption. The time scale for this redistribution, on the order of 200fs, is very similar to the time scale of the pulses which governs the persistence of the coherence effects. Thus, with our time resolution it is difficult to differentiate between the effects of

Figure 5.8 Differential absorption spectra for (a) 100A sample and (b) 6000A sample.

BGR and coherent signal at the bulk band edge. This effect will be further masked in the narrower quantum wells, where the band edge is dominated by bound excitonic states, as these shift very little due to screening. However, a band gap shift in these samples might still be observed because as the band edge moves closer to the excitonic bound states oscillator strength shifts away from the bound exciton. Thus, BGR in the narrow quantum well samples would appear as a loss of excitonic oscillator strength. Since screening and coherent signals also give a loss of oscillator strength on the same time scale, however, BGR in narrow wells can not be isolated from other effects.

5.6 Electron phonon interactions

Interactions between carriers and lattice vibrations allow hot carriers to cool to the lattice temperature, which is, on a long time scale, in equilibrium with the surroundings.

(In our case, the sample is tied to a copper cold finger which is at approximately 7K.)

Studies of these interactions have become quite active in the last decade, as carrier cooling dynamics are critical to the recovery times of many devices. These studies have in large part been restricted to the electron phonon interaction, because the hole phonon interaction is greatly complicated by the complexity of the valence band structure. In our experiments, it is legitimate to concentrate on cooling of the electrons, as they have the large majority of excess energy at excitation. This continues to be true once the carriers have thermalized, as collisions between electrons and holes are likely to be nearly elastic due to the large ratio of hole to electron mass.

In general, when considering the electron-phonon interaction, [17] one must consider four types of processes.

1. Deformation potential interactions between electrons and acoustic phonons.
2. Deformation potential interactions between electrons and optical phonons.
3. Piezoelectric interactions between electrons and acoustic phonons.
4. Polar interactions between electrons and longitudinally polarized optical phonons.

Deformation potential interactions are those in which the interaction potential is due to mechanical strain, which alters the band structure by shifting energies. These energy shifts can be directly proportional to displacement (in the case of optical phonons)

or to differential displacement (in the case of acoustic phonons). Deformation potentials occur for all types of materials. The polar and piezoelectric interactions, on the other hand, only occur in polar materials, and rely on polarizations set up in the material by the motions of different types of atoms within a unit cell. When they are allowed, these types of interactions generally are far stronger than those based on the deformation potential. In particular, for III-V materials such as InGaAs and InAlAs, the polar interaction is generally by far the strongest for intravalley scatterings. The selection rules for each type of process depend on the symmetry of the band in which scattering takes place. We will be concerned only with Γ valleys, in which LA, TA, LO, and TO nonpolar processes are allowed, as well as LA, TA, and LO polar processes. The strongest polar interaction is that between electrons and LO phonons. This is due to the geometry of the atomic displacements. For LO phonons, the opposite motions of the different types of atoms within the unit cell set up a long range polarization which is directly proportional to displacement, that is, there is a nonzero gradient of the scalar potential set up by the motion. In the case of TO phonons the gradient is zero. In general, only small wavevector phonons can participate in this type of scattering, due to the requirements for conservation of energy and momentum. The piezoelectric scattering of electrons with acoustic phonons, though also a polar process, is much weaker than the LO phonon scattering because acoustic phonons can only produce a second order polarization, that is, the polarization depends on the differential displacements of the atoms.

In the general procedure for obtaining energy and momentum relaxation rates from the electron phonon interaction one treats the interaction as a small, time dependent

perturbation to the electronic wave functions and then uses Fermi's Golden rule to calculate appropriate transition rates. This is fairly straight forward for bulk materials provided that one treats cases with relatively simple conduction band structures. The main difficulty is quantitative calculation of the strengths of the relative interactions, however these can be obtained empirically via measurements of bulk properties, such as compressibility. One complication occurs in the case of high carrier density, however, as in this case the polar interactions are screened, and the screening must be treated appropriately. A second effect of high carrier densities arises because the rate for hot electrons to emit an LO phonon may be much higher than the rate for LO phonons to decay into acoustic phonons. Since only acoustic phonons can actually drain energy from the sample and equilibrate it with its surroundings, in this case a large population of nonequilibrium LO phonons may develop. These are known as hot phonons, and they decrease the rate of carrier cooling by increasing the likelihood that a carrier will reabsorb a phonon. The effects of these hot phonons have been measured in a number of studies in GaAs. [18,19]

The addition of confinement to this picture introduces a number of new issues. Perhaps most obviously, the electronic wave functions must be modified to take account of the confinement. Boundary conditions for doing this are well understood. [20]. A more complicated problem is posed by the effect of confinement on the phonon modes. For long wavelength acoustic modes, the frequencies in the well and barrier generally overlap quite well. In this case, the acoustic modes see the multiple quantum well structure as a superlattice, which leads to zone folding. In many cases, however, this does

not introduce a strong perturbation to the strength of the interactions and the acoustic phonons can be treated as bulk modes. For optical phonons, however, the situation is very different. In this case, the frequencies of long wavelength modes in the wells and barriers may be very different, making it impossible for phonons to propagate. In this case we must treat confined phonon modes. The development of appropriate boundary conditions for these modes remains controversial. In addition to confined versions of ordinary optical modes, there exist also new interface modes whose importance in electron phonon scattering is also the subject of intensive study. The presence of a transition with well width from propagating to confined phonon modes in GaAs/GaAlAs has been shown in Raman scattering experiments.[21]

In the simplest model of confined phonon modes, phonons are taken to have zero amplitude at the barriers, leading to a quantization scheme very similar to that used for electrons in infinite wells. We obtain for the allowed wavevectors of the phonons,

$$q_z \frac{L}{2} = (n+1)\pi \text{ for } n \text{ odd, and } q_z \frac{L}{2} = (2n+1)\frac{\pi}{2} \text{ for } n \text{ even.}$$

Confined modes will have a

weaker polar optical interaction with electrons than do bulk modes, as this interaction goes as $1/q^2$, and for confined modes there exists a minimum q .

In addition to these effects, it is clear that other, more subtle effects are produced by confinement. These include a change in the strength of screening of the polar interaction as well as alterations of the overlap of the electron and phonon wavefunctions. It should be noted that the majority of both experimental and theoretical work on the effects of confinement on the electron phonon interaction have treated electron populations that are very high above the band edge, on the order of 200 to 400 meV, in

lattices that are at room temperature. This focus has occurred because for devices in which carriers are electrically (rather than optically) injected, the carriers are likely to start out very high in the bands. In this case, the LO phonons are by far the most important as the electrons have so much excess energy, and there is already an equilibrium phonon population which can result in absorption as well as emission of phonons. Furthermore, intersubband processes as well as intervalley scatterings are important in this case.

In the case of our experiment, the electrons are excited with about 26 meV of excess energy. The lowest LO phonon mode in InGaAs is at about 28 meV, however this "InAs like" mode interacts far more weakly with electrons than the "GaAs-like" mode at 35 meV. [22]. Thus we expect that when first excited, less than 5% of the electrons, (and none of the holes) have sufficient energy to emit LO phonons. The LO phonon modes in InAlAs are about 10 meV higher at zone center than those in InGaAs, and thus we do expect some confinement of these modes in the narrowest wells.[23]

In order to look for effects of confinement on electron phonon interactions in our samples, we measured the rate of arrival of carriers at the band edge over long times, up to 50 ps. Since carrier thermalization is complete within a few hundred fs, we expect the gradually increasing signal at the band edge to reflect the cooling of the electron distribution, which results in a slowly increasing band edge population. Data for the 100 Å quantum well and the 6000 Å bulk sample are shown in figure 5.9. Note that the band edge signal in the bulk saturates considerably faster than does that in the quantum well, indicating that in the bulk sample, the carriers cool to a Fermi-Dirac distribution more quickly than in the quantum well. Fitting these signals with a simple exponential model,

which assumes single phonon scatterings, we find a 7ps time constant for the bulk, compared to a 21ps time constant for the quantum well.

One might interpret this difference as a reduction in electron-acoustic phonon scattering due to confinement. However this interpretation is problematic because the time constants we obtained are far too short. Typical electron-acoustic phonon scattering times for III-V materials are on the order of 100-300 ps. [24] Our scattering times are far more consistent with those obtained both theoretically and experimentally for GaAs with high enough carrier densities to cause hot phonon effects. (~7ps) [24]. The densities we excite, on the order of 10^{11} - 10^{12} cm⁻² are definitely in this regime.

This puzzle is resolved by considering more carefully the Boltzman distributions the electrons take up once they have thermalized. The electronic distributions in two and three dimensions are given by the product of the appropriate Boltzmann distribution and the appropriate density of states: $\rho(\epsilon) = f(\epsilon)D(\epsilon)$. In this case the Boltzmann distributions are the same, however the densities of states near the band edge are very different (rising as $\epsilon^{1/2}$ in three dimensions, independent of ϵ in two.) Figure 5.10 (a)

shows the energy distributions of our hot electron populations as calculated from their excess energy. In Figure 5.10 (b), the populations have been integrated to show the total percentage of carriers below a given energy. In both figures the strongest LO phonon, at 35meV, is marked. From this figure we see that in the

thermalized distributions, in contrast to the initial non thermal distribution, a large fraction of the carriers are able to cool by emission of LO phonons. This fraction, however, is confinement dependent, being considerably larger (30%) in the bulk case than in that of the quantum well (15%).

This difference of carrier distribution implies that the bulk sample will have a time constant for carrier cooling that is approximately half that of the quantum well. Our data show a somewhat larger difference. We attribute this additional slowing of electron cooling in the quantum well sample to a reduction in the electron-LO phonon coupling constant due to phonon confinement. It should be noted that once the electrons have cooled to a temperature of about 20meV, virtually no carriers remain high enough in energy to emit LO phonons. At this point, acoustic phonon emission will dominate, and the carrier cooling rate will be vastly reduced. The remaining cooling is expected to take place on the time scale of several hundred picoseconds, which is unfortunately not experimentally accessible for us.

5.7 Conclusions

Our studies of the behavior of carriers excited well above the band edge in InGaAs heterostructures have revealed a number of important results. We find that carrier thermalization to a hot, Boltzman distribution occurs within 250fs of excitation, and is relatively independent of well width. This indicates that the Coulomb interaction among carriers is not strongly modified by confinement at our densities. Pauli blocking of the

exciton by this hot distribution, however, is influenced by confinement, being approximately twice as effective in two dimensions than in three, and extrapolating smoothly between the two and three dimensional limits. The relative strength this non-linearity in two and three dimensions agrees well with our calculations.

While transient nonlinearities high in the band, such as the transient Fermi edge singularity, are relatively unaffected by confinement, transient nonlinearities at the band edge are quite sensitive to it. This is due in large part to the very different density of states at the band edge in two and three dimensions, including the enhancement of exciton binding energy and oscillator strength with confinement.

Dynamics on a long (hundreds of picoseconds) time scale are also affected by confinement. This is due to a modification of the efficiency of electronic cooling by phonon emission. Confinement appears to effect this process in two ways. First, the dimensionality dependent change in carrier distribution which originates in the density of states results in an increased proportion of the thermalized electron distribution which can emit LO phonons in three dimensions versus two. Second, confinement of the phonons by the ionic mass difference between barriers and wells results in a modified phonon density of states, reducing the electron-LO phonon coupling.

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Chapter 6

The AC Stark Effect

6.1 Introduction

In the previous chapter we have discussed dynamic nonlinearities which occur with resonant excitation of the semiconductor, due to the presence of a real carrier population in the conduction and valence bands. The time scales of these nonlinearities are in general determined by carrier dynamics, such as carrier carrier and carrier phonon scattering times. In this chapter we treat a different type of nonlinearity, in which the carriers are excited non-resonantly (below the band edge). In this case no real carrier population is produced, however the ground state of the semiconductor is modified by the presence of strong ac electromagnetic fields. There are several ways to view this modification. Perhaps the simplest is to realize that when fields which are comparable in strength to atomic fields are imposed on the semiconductor, the eigenstates of the system are dressed, resulting in a new ground state which now contains some superposition of the unperturbed excited states. In this sense, the name AC Stark Effect for these shifts due to nonresonant excitation makes sense in analogy to the well known DC Stark Effect, in which atomic levels are shifted by the presence of a DC Electric field.

An alternative way to view the change in the system is in terms of virtual excitations. Although classically no population may be excited by the nonresonant pump beam, the Heisenberg uncertainty principle states that there is some flexibility here, given by $\Delta E \Delta t \geq \frac{\hbar}{2}$. Thus, for a detuning given by ΔE , a carrier population may be produced which exists for a time of order $\Delta t = \hbar/2\Delta E$. These short lived populations are known as virtual carriers, although they exhibit all of the same types of nonlinearities as real carriers.

They do not participate in relaxation processes, however. In particular, a virtual population can exhibit phase space filling effects, resulting in shifts in the absorption of a test beam. A third way to understand these nonresonant effects is in terms of anharmonic charge density fluctuations which are induced by the optical field and can in turn interact with other (probe) optical fields. All of these approaches to nonresonant excitations are important to understanding different aspects the AC Stark Effect. We will appeal to them as they become relevant.

The first two sections of this chapter cover the AC Stark Effect (ACSE) in the steady state limit. In section 6.2 we will examine the effects of nonresonant excitation on two level systems, a picture which is appropriate both for isolated atoms and for semiconductors in the case where Coulomb coupling among different k states is neglected. Section 6.3 will cover the alterations to the two level picture which arise in the presence of Coulomb correlation. The effects of short pulse excitation, which is in fact necessary to observe the ACSE in semiconductors, are explained in section 6.4. In section 6.5 I present our data on the ACSE as a function of well width, and section 6.6 contains our conclusions.

6.2 The Two Level System Picture

The AC Stark effect was first understood by Rabi [1] in 1937 in relation to the interaction between atoms and radiation. Consider a two level system which is excited by a nearly resonant pump beam at frequency ω_0 (see figure 6.1). In the dressed state picture,

the presence of the pump beam results in two nearly degenerate levels, $|g,1\rangle$ and $|e,0\rangle$.

The interaction between the system and the field, however, splits these levels. In the dipole approximation and the rotating wave approximation, this splitting (known as the generalized Rabi splitting) can be found from perturbation theory and is given by

$$6.1 \quad \Delta E = \left[(E_{eg} - \hbar\omega)^2 + 4|\mu_{eg}E_p|^2 \right]^{1/2},$$

where μ_{eg} is the dipole moment of eg , E_p is the pump field, and E_{eg} is the energy splitting between g and e . In the weak excitation regime, the new energy splitting between the dressed ground and excited states is then given by

$$6.2 \quad \Delta E_{eg} = \frac{2|\mu_{eg}E_p|^2}{\hbar^2 \Delta\Omega}.$$

In the absence of damping, the atoms will oscillate between the ground and excited states with a frequency given by $\omega = \Delta E/\hbar$, which is known as Rabi flopping.

These two level system results can be applied to a semiconductor system if Coulomb coupling between different k states e.g. excitonic effects, are ignored. In this case the energy splittings given in equations 6.1 and 6.2 apply individually to each pair of conduction and valence band states which have the same k (assuming that

we can ignore the photon momentum, which is always a good approximation for our

experiments.) Note that this implies that the different k states will have different energy shifts in the presence of the pump beam, depending on their detuning from resonance. For our case, in which the semiconductor is excited near the band edge, the states at the band edge will shift the most, as they are least detuned, and states farther away will shift less. The end result, then, is a decrease in dispersion of both bands, which can also be viewed as an increase of the effective mass of both electrons and holes. The situations in which this two level picture is appropriate for semiconductors will be discussed in the next section.

6.3 Steady State Picture with Coulomb interaction

In order to properly take into account the effects of Coulomb correlation on the ACSE we need to solve the semiconductor Bloch equations (see Chapter 4) in the appropriate limits. Since we excite no real population, scattering processes can be neglected and the pair amplitude follows the applied field instantaneously. This is known as adiabatic following, and has the result that relaxation term in the SCBE can be neglected. Again going into the rotating wave approximation, the SCBE reduce to

$$6.3 \quad i \frac{\partial}{\partial t} \psi_k = \{E_c(k) - E_v(k) - \omega\} \psi_k - \{n_c(k) - n_v(k)\} \Delta_k$$

$$6.4 \quad \frac{\partial}{\partial t} n_c(k) = -\frac{\partial}{\partial t} n_v(k) = -i(\psi_k^* \Delta_k - \psi_k \Delta_k^*)$$

where $\Delta_k = \mu_k E + \sum_{k'} V_{kk'} \psi_{k'}$. In general these equations can only be solved numerically,

as each function depends on an integral over the others. However, in the case of weak

excitation, where $\Delta_k \ll E_g$, the energies ε_k can be solved for analytically, leading to an absorption shift ΔE_k for each k given by

$$6.5 \quad \Delta E_k = \left(E_g - \omega + \frac{\hbar^2 k^2}{2m} \right) + \frac{2|\Delta_k|^2}{\left(E_g - \omega + \frac{\hbar^2 k^2}{2m} \right)}.$$

Note that as in the two level system case, the single particle states all blue shift, resulting in non parabolic bands. However, the shift is modified in that each state shifts due not only to the applied electric field (μE term) but also due to the *local field* caused by Coulomb interactions among k states.

This result gives the shifts for unbound states, however we are also interested in the results for bound excitonic states. Again, analytical results are only obtainable in the weak excitation regime. The calculation in this case is quite lengthy, requiring the solution of a Wannier equation in the presence of the pump beam which renormalizes the single particle states. The result, however, is quite intuitively satisfying. The blue shift of the exciton is given by [2,3]

$$6.6 \quad \Delta \omega_x = \frac{2|\mu_{cv} E_p|^2}{\Delta \Omega} \frac{|\phi_x(r=0)|^2}{N_{psf}}$$

where N_{psf} is the exciton saturation density (see section 6.4) and $\phi_x(r=0)$ is the unperturbed excitonic wavefunction for $r=0$. Thus, as for the single particle states, the exciton blue shift is just the same as for the two level system, with the exception that the shift is enhanced by the Coulomb correlation, and limited by phase space filling.

The major difference between the two level system case and the excitonic case, however, comes not in the blue shift, but in the the oscillator strength of the system. In

the two level system case, the resonance is shifted, however its oscillator strength remains the same. This is not the case for excitons. Two opposing contributions to the excitonic oscillator strength exist in the case of the ACSE. First, as mentioned in section 6.2, the effective masses of the electrons and holes near the band edges are increased by the presence of the pump. The binding energy of excitons is proportional to reduced mass, and thus the increase in effective mass results in an increased binding energy, reduced Bohr radius, and hence increased oscillator strength. On the other hand, as seen in equation 6.6, phase space filling plays a role even for virtual carriers. The presence of virtual excitons occupying phase space reduces the opportunity for absorption into these states, thus reducing the excitonic oscillator strength. The combination of these two effects gives the following result for the excitonic oscillator strength, f_x , in the presence of the non resonant pump, [8]

$$6.7 \quad f_x = f_x^0 \left(1 - \frac{N}{N_{psf}} \right) \left(\frac{m^*}{m} \right)^d$$

Here m^* is the renormalized reduced mass, and d is the dimensionality of the sample. N is the density of virtual carriers excited, which is given by

$$6.8 \quad N = \frac{|\mu_{cv} E|^2}{\Delta \Omega} \frac{|\phi_x(r=0)|^2}{N_{psf}}$$

The dimensionality dependence arises due to differences in relative effectiveness of phase space filling in two and three dimensions, as well as to differences in the excitonic wavefunctions. It should be noted that these expressions apply for the case in which all of the virtual population is excited into one excitonic state, and there are no coupled

continuum states near by. They predict almost no change in oscillator strength for 2D, and a slight increase in oscillator strength for 3D.

Although these predictions have led to a number of studies, they are not in fact verifiable experimentally, because they account only for the steady state regime. As discussed in the next section, experiments on the semiconductor ACSE can only be performed with short pulses, and these have important effects on the results.

6.4 Transient AC Stark Effect: Theory and Experiment

As mentioned in the previous sections, until recently all theoretical work on the ACSE was restricted to the steady state domain. These studies are not directly applicable to current experimental work because the high field intensities necessary to produce a measurable Stark shift are thus far only available with short pulse (ps or fs) lasers. It has been found experimentally [4,5] that the results of such short pulse experiments are in fact quite dependent on the spectral and temporal properties of the pulses used. Recently, several works have been published [6,7,8] which treat the ACSE problem in the short pulse regime. In this section I will discuss some of the results of these studies, and use them to explore our data on the ACSE in InGaAs heterostructures.

In order to obtain a quantitative theory for the transient ACSE, it is necessary to obtain full numerical solutions of the semiconductor Bloch equations. As in the model discussed in section 6.2, screening can be neglected due to the very low density of carriers produced. Essentially then, it is necessary to solve equations 6.2 and 6.3 self consistently

for the case of time dependent variables. This results in a number of modifications to the steady state results.

Our experimental technique for the ACSE is similar to that described in Chapter 5, with the exception that the 30nm FWHM pump beam is tuned approximately 30meV *below* the lowest excitonic resonance for each sample. The excitation energy is still approximately 1nJ per pulse, leading to a peak power density in the sample of about 100 MW/cm². In this case we are in the relatively low excitation regime, where $\mu E \ll \Delta\Omega$. Once again, we measure the change in transmission of the pump beam as a function of time delay between pump and probe. Differential transmission spectra for 100, 200, 500 and 6000 Å samples are shown in figure 6.2.

Perhaps the most noticeable feature of our ACSE data is that, contrary to what one might expect for a perturbation that lasts only as long as the pump pulse, the signal persists for five or six hundred femtoseconds in all the samples. This is due not to the production of real carriers, but rather to a strong negative time delay signal which originates in the coherence effects discussed in Chapter 4. This is precisely the same effect which gives a negative time signal in the experiments of Chapter 5. In this case, however, since we are pumping below resonance, the perturbation of the free induction decay by the pump results in a spectral shift of the excitonic resonance. This leads to a bipolar oscillation, with its zero crossing near the peak of the exciton, rather than a bleaching signal which peaks at the exciton as is the case for real excitation. The persistence of this signal out to negative times gives, in the case of homogeneously broadened lines, gives a measure of the coherence decay time, T_2 . A plot of these decay times for our samples is

Figure 6.2 ACSE spectra for (a) 100A sample (b) 200A sample (c) 500A sample (d) 6000A sample. 1nJ pump energy, 30meV detuned.

shown in figure 6.3. The somewhat unexpected well width dependence of these times can not be immediately attributed to differences in decay times however, as our samples may be considerable inhomogeneously broadened. This will be discussed further in Chapter 7, where four wave mixing results are presented.

A second dynamic effect which is observable for all the samples is the evolution of the signal from bipolar to unipolar with increasing time delay. On close observation, it is found that for all samples the unipolar, late time signal is centered at the exciton, whereas the bipolar signal has its zero crossing, rather than its peak, centered at the exciton. This indicates that the negative time signal is due predominantly to a spectral shift, while at later times the signal is dominated by a reduction of oscillator strength. This evolution is in fact predicted by the numerical results of Binder and Koch, which indicate that as the virtual population increases during the pulse overlap, phase space filling causes a decrease in oscillator strength which follows the pulse profile adiabatically. For negative time

delays, in contrast, no population is present and the signal is due to a spectral shift of the exciton which takes place during free induction decay.

The maximum bleaching of the band edge signal can be seen in figure 6.4 to differ between the bulk and quantum well samples. There are several possible explanations for this difference. The change in oscillator strength of an exciton due to phase space filling by the virtual population can be easily calculated in the manner outlined in section 5.5. In order to obtain an analytical result, one assumes that the virtual population is excited only into the lowest excitonic state. In this case, the change in oscillator strength is given by

$$\Delta f = \frac{N}{N_{psf}}, \text{ where } N_{psf} \text{ is the saturation density. This saturation density varies with}$$

dimensionality, primarily due to the decrease in Bohr radius of the exciton with increasing confinement. The values for the two and three dimensional limits are $N_{psf}^{2D} = \frac{7}{32} [\pi a_{2d}^2]^{-1}$

$$\text{and } N_{psf}^{3d} = \frac{8}{21} \left[\frac{4\pi}{3} a_{3d}^3 \right]^{-1}. \text{ If the densities of virtual carriers excited in two and three}$$

dimensions were equal, this would lead to an approximately five fold larger loss of oscillator strength in three dimensions than in two. However, in the static approximation, the density of virtual carriers is proportional to the expectation value of the excitonic wave function at $r=0$. (See equation 6.8). If we take the absolute two and three dimensional values for $\phi(r=0)$, we find that in fact the loss of oscillator strength due to phase space filling should be approximately two times greater in two dimensions than in three.

Recall from equation 6.7 that there is an additional contribution to the change in excitonic oscillator strength from the (time dependent) renormalization of electron and

Figure 6.4 Absorption spectra with (dotted) and without (solid) pump excitation, and differential transmission spectra. Samples were excited 30meV below the band edge. (a) 6000Å sample (b) 100Å sample.

hole masses. The size of this contribution depends on the relative time scales of the renormalization and the internal time scales of the system. In particular, the time scale of this renormalization should be compared to the Bohr orbital time for the exciton. In the case where the pulses are of approximately the same duration as the Bohr orbital time, the excitonic wave function is significantly perturbed by the mass renormalization, and the binding energy and oscillator strength increase. Pulses much shorter than the Bohr orbital time, however, do not strongly perturb the exciton, and thus in this case the mass renormalization does not compensate as completely for the loss of oscillator strength due to phase space filling. Thus the time scales in the problem have a strong effect on the ACSE response.

Object	Energy(meV)		Timescale (fs)	
	2D	3D	2D	3D
Bandgap	880	800	2	2
Pulse Duration	15	15	120	120
Binding Energy	8	2.5	250	1000
Detuning	35	35	50	50
X linewidth	5	?	400	?

Table 6.1 Time and energy scales for two and three dimensional samples.

Table 6.1 shows the relevant time and energy scales for our narrowest and widest samples. Note that the Bohr orbital time in the bulk is about four times longer than that in

the quantum well, and thus ratio of pulse width to Bohr orbital time is *considerably smaller* for the bulk than the quantum well. As discussed above, we expect these differences in time scale to influence the ACSE response. We thus have two counteracting forces on the change of oscillator strength for the two samples. The loss of oscillator strength due to phase space filling should be greater in the quantum well, however the compensation for this loss due to mass renormalization should also be greater. Without numerical studies it is not possible to estimate the relative sizes of these two effects. Looking at figure 6.4, we see that the bulk bleaches considerably more than does the quantum well. This may reflect the increased effectiveness of mass renormalization due to the shorter time scale in the quasi two dimensional excitons.

In the preceding discussion we have assumed that only a single excitonic resonance participates in the shift and bleaching. There are two parts to this assumption. First, we have assumed that there is a single excitonic resonance which is much closer in energy to the pump than is any other level, and thus that the whole spectral change may be taken to come from this resonance. Although this approximation may be good for the narrowest wells, where the excitonic binding energy is approximately 10meV, it is certainly not the case for the bulk or 500Å samples, in which the existence of bound excitonic states is not clear, and there are certainly many very closely spaced levels at the band edge. Even for the narrowest wells, the higher lying excitonic states which are only a few meV away from the 1s resonance call this approximation into question. Second, we have assumed that the virtual population we excite occupies only the 1s k states. In the numerical studies of Binder and Koch it has been shown that this is not the case. In fact, the virtual population

distribution extends over at least twice as much k space as does the $1s$ state. This means that our approximations about phase space filling factors may not be appropriate.

Qualitatively, however, these approximations are useful in gaining an intuitive feel for the physics behind the numerical calculations of the full time dependent problem, allowing us to understand which influences on the ACSE may be most important.

In addition to the effects of short pulses on the change in oscillator strength seen in the ACSE, time dependent effects also play a role in the blue shift. In particular, the use of pulses much shorter than the coherence time of the system, T_2 , results in a considerably reduced blue shift in comparison with the cw estimates. This is inherent in the way the measurement is performed. We measure the spectrum of the probe pulse, which is the Fourier transform of the polarization emitted in the probe direction, that is,

$$P(\omega) \propto \int_{-\infty}^{\infty} P(t)e^{-i\omega t} dt. \text{ Polarization is emitted for a time dictated by } T_2, \text{ however the}$$

perturbation to the system exists only during the pump pulse. Since the integral over time must be performed out to times long enough that the polarization has decayed completely, the measured blue shift will be reduced from the cw value by the ratio of τ_p/T_2 . In our samples T_2 , as measured by FWM in Chapter 7, ranges between three and five times the pulse width, resulting in a substantial decrease in the measured blue shift.

Finally, in figure 6.4 we note a negative absorption below the exciton in the presence of the pump pulse for the $100\text{\AA}$ sample. This effect has been predicted by several workers [7, 8] and is again unique to the short pulse regime. It originates in the combination of a temporally short pump pulse, and a spectrally wide probe pulse. The short duration of the pump pulse results in a sudden cutoff of the perturbation to the

system, with a characteristic time τ_p . Transforming to the spectral domain, this implies a change in the system with characteristic frequency $\omega = \tau_p / \hbar$. Detailed numerical calculations indeed predict this effect, which manifests itself as oscillations with this characteristic frequency centered around the exciton. In particular, the first dip in the oscillations appears as gain just below the excitonic resonance. (Oscillations further below resonance are not visible as there is no absorption there.) We have made the first observation of this gain, to our knowledge. Other workers, using GaAs samples [5], have not observed this effect. It is likely that this is due to higher intensities used in their experiments, which are known to produce real carriers through two photon absorption. The presence of this carrier distribution would alter many of the band edge effects considered here.

Finally, we note that the total differential absorption signal we observe decreases with increasing well width. This is consistent with the dependence of the ACSE on the oscillator strength at the band edge, which decreases as confinement is relaxed. Furthermore, as the samples become wider, the excitons are more weakly bound and thus the time scales for excitonic behavior become longer. For the same length of pulse, then, we expect reduced perturbation of the exciton. In particular, gain below the exciton is noted only in the narrowest sample, in which both time and oscillator strength serve to enhance the ACSE.

6.5 Conclusions

We have performed the first measurements of ACSE in InGaAs on a femtosecond time scale. These reveal several previously unmeasured effects, including gain below the excitonic line and the evolution from shifting to bleaching, that have been predicted theoretically. In addition, we find a considerable qualitative dependence on well width, which originates both from changes in excitonic oscillator strength and from the related changes in Bohr orbital time with confinement.

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Chapter 7

Four Wave Mixing

7.1 Introduction

In this chapter I will discuss experiments we have performed to understand the coherent dynamics of our system. In the semiconductor Bloch equations there are two phenomenological decay times introduced to account for the true dynamics of the system. The first of these, known as " T_1 ", gives the energetic relaxation time of the population excited from its ground state. For carriers excited near the band edge, this can be very long, on the order of nanoseconds in the case of InGaAs. The second time, T_2 , is known as the coherence or phase relaxation time. This time measures how long it takes for the coherent polarization initially set up in the system by the optical field to decay away. This loss of coherence takes place via the same types of collisions which are important in carrier thermalization, such as electron-electron scattering, electron phonon scattering, electron impurity scattering, however in the case of polarization relaxation we are interested in *elastic* collisions, which cause a change of phase, as well as the inelastic ones which result in a redistribution of energy. An examination of the dimensionality dependence of T_2 thus gives additional information about how confinement affects various scattering mechanisms in our heterostructures.

In the past ten years, the technique of time resolved four wave mixing (FWM) has been developed and exploited to gain access to information about coherent dynamics in a number of contexts. In section 7.2 I will present a basic physical picture of FWM. The mathematical details of FWM measurements are discussed in Chapter 4. In section 7.3 I discuss the influence of confinement on the various scattering mechanisms that can cause

dephasing. Section 7.4 covers the results of spectrally resolved FWM measurements made on four of our samples, including some unexpected results obtained in the bulk sample. Section 7.5 addresses the conclusions we can draw from our measurements.

7.2 Principles of Four Wave Mixing

Figure 7.1 Schematic of FWM geometry. Four wave mixing directions shown in bold.

Time resolved four wave mixing measurements are performed using two non-collinear pulses which are time delayed from one another. The two pulses interfere on the sample, forming a grating (of intensity, in the case of parallel polarized beams, or polarization, in the case of perpendicularly polarized beams.) A third photon, from one of the beams, is diffracted from the grating into the momentum conserving direction $2k_1 - k_2$ or $2k_2 - k_1$. (Figure 7.1) The presence of the signal in this direction requires a non-zero third order polarization, $P^{(3)}$. This third order polarization is generated in the following

manner: One photon from the first pulse creates a first order polarization. When the second pulse arrives, a second order population is created by the combination of the field of the second pulse and the polarization remaining from the first pulse. Finally, a second photon from the second pulse is scattered off the grating which results from the interference of the first two photons, giving a third order polarization emitted in the direction $2k_2 - k_1$. In order for the total polarization emitted in the FWM direction to be non-zero, the first order polarization produced by the first pulse must be non-zero when the second pulse arrives. This requires that the dipoles initially excited by this pulse are still oscillating in phase. Initially, when the dipoles have just been created by a coherent laser pulse, they all oscillate in phase. As time passes, however, the carriers undergo scattering which randomizes their phases. In addition, if the dipoles have resonant frequencies which differ slightly from one another (inhomogeneous broadening), they will drift out of phase with each other as time passes. If relative phase coherence among the dipoles still exists when the second pulse arrives, then a non-zero $P^{(3)}$ will be produced in the FWM direction. On the other hand, if the dipoles have lost their coherence before the arrival of the second pulse, no signal will be produced. The time for the dipoles to dephase, known as the coherent or transverse relaxation time, can thus be measured by mapping out the intensity of the FWM signal as a function of the time delay between the first and second pulses. It should be noted that $P^{(3)}$ contributes to the signal along k_1 and k_2 , as well as in the FWM direction. The signals along k_1 and k_2 , however, also contain the first order contributions from the two beams being transmitted through the sample,

whereas the signal in the FWM direction contains third order coherent contributions only.

The directions $2k_1 - k_2$ and $2k_2 - k_1$ are thus referred to as being "background free."

In the case where the laser excites a system of uncoupled two-level dipoles, it can be shown (see section 4.3) that the FWM intensity decays exponentially as a function of time delay, with a characteristic time $T_2/2$ for an homogeneously broadened system, or $T_2/4$ for an inhomogeneously broadened one. A number of studies, both experimental and theoretical, however, have shown that a simple exponential decay is the exception rather than the rule in semiconductors. This is due primarily to the fact that a semiconductor can not be described as an ensemble of independent two level systems. In many cases, several different resonances are excited by the pump beam (for example the light and heavy holes) leading to an interference in the FWM signal known as quantum beats.[1] Even in the case where a single excitonic level is excited, Coulomb correlations substantially modify the FWM mixing signal, leading to non-exponential decay. [2,3]. Finally, in the case of optically thick samples it has been shown that even for a simple two level system (e.g. a strong resonance in a noble gas), the model of exponential decay fails due to propagation effects. [4].

These various effects substantially complicate the interpretation of FWM signals in terms of dephasing dynamics. In order to extract more accurate information than is available in a simple FWM experiment, workers have developed techniques to better characterize the FWM signal. The simplest FWM experiment measures the total intensity of the emitted signal as a function of time delay between the two pulses. This is known as time integrated four wave mixing. One alternative is to time resolve the emitted signal at

fixed time delay, by upconverting it in a nonlinear crystal with a second, well characterized, pulse, (usually coming directly from the laser). This technique, known as time resolved four wave mixing, has allowed, for example, an analysis of the effects of the density dependence of exciton-exciton interaction, which causes a delayed response in the emission. Another possibility is to spectrally resolve the FWM emission. In the case where several resonances are emitting simultaneously, spectral resolution may allow a deeper understanding of the origin of non-exponential decays. In addition, the detailed line shapes observed can be used to understand interactions in the system, such as Coulomb coupling among states [5], or coupling among Landau levels. [7] In principle, the time resolved and spectrally resolved signals are related by Fourier transforms, and contain the same dynamical information. In practice, however, since the two measurements are always made over finite intervals, they provide somewhat different and complementary information. In particular, measurements of very early time dynamics are better resolved with $S(t)$, while measurements of peak positions or long temporal delays are better resolved in $S(\omega)$. Finally, one can make interferometric measurements of the phase of the FWM emission. Although this gives a great deal of insight into the dynamics of dephasing, it requires an extremely stable optical system in order to give reproducible results. In our case neither time resolved nor phase measurements are possible due to the low repetition rate of the amplifier/continuum generation system. We have, however, made spectrally resolved measurements of the FWM signal as a function of time delay, which give considerable information on the dynamics of our samples. Our measurements were made with excitation just below the lowest exciton level of each sample, in order to

avoid the creation of a large free electron-hole population. Although the physics of dephasing in the presence of such a population is certainly of great interest, the high scattering rate which results generally gives a dephasing rate much faster (~ 10 fs) than can be measured with our laser system.

7.3 Physical Processes Affecting Excitonic Dephasing

As mentioned in section 7.2, dephasing times can be influenced by any number of scattering processes. Early work [8] demonstrated a strong temperature dependence to excitonic FWM decay times, indicating the importance of exciton phonon scattering. In our case, however, all measurements were performed at low (10K) temperatures, and thus the phonon population is quite small. A second scattering mechanism which strongly affects dephasing is exciton electron or exciton hole scattering. As mentioned above, we attempted to excite our samples so that very little energy was available to excite the free carrier states above the exciton. Thus we expect that for the narrow samples, where the excitons are more strongly bound, few free carriers are produced, at least in the case of low density excitation. For high density excitation, where the excitonic resonances are nearly saturated, production of unbound electrons and holes again becomes important. In the case of the 500Å and bulk samples, however, which do not show excitonic resonances which are clearly separated from the continuum, exciton-electron and exciton-hole scattering may be important at all densities.

The interactions among excitons have been shown to strongly affect [3,4,5] FWM emission. In addition, it has been shown that in cases where the excitons are reduced in spatial extent (and other mechanisms for dephasing, such as free carriers and phonons, have been suppressed) dephasing times tend to lengthen. In one measurement of dephasing times in 100Å InGaAs quantum wells and InGaAs bulk, [6] it was found that the dephasing time was six times longer in the quantum wells. When excitons are reduced in spatial extent by the application of a magnetic field, their dephasing times also tend to increase. This increase in dephasing time with decreasing spatial extent of the exciton has two possible contributions. The first is exciton-impurity scattering. As excitons are reduced in size, they are less likely to encounter and thus scatter from impurities. How the increase in dephasing time should scale with exciton diameter, however, is not clear due to the extended nature of the ionized impurity Coulomb potential. Another possibility for the increase in dephasing time would be a decrease in exciton-exciton scattering. Schultheis et. al. have shown that for low temperatures and low excitation densities, exciton exciton scattering can dominate the dephasing rates. Free carriers, however, scatter much more strongly and thus dominate excitonic dephasing whenever they are present in non-negligible densities.

For the low temperature, low density, case, then, we would expect the dimensionality dependence of the dephasing time to be dominated by changes in the exciton interaction with material defects. However, there are two opposing contributions to this effect. As discussed above, the decrease in exciton cross section with confinement would be expected to reduce the excitonic dephasing time for the case of a fixed impurity

concentration. However, as one goes to very narrow quantum wells, where the well width is substantially smaller than the exciton diameter, the importance of scattering from defects in the interfaces will increase. (Although for the infinite barrier case, the excitonic wave function has a node at the interfaces, and thus excitons should not directly "contact" the interfaces, charged impurities in the interface will affect the excitons over long distances. Furthermore, for finite barriers, the electron and hole wavefunctions do leak somewhat into the barrier layers, and this leakage is known to increase with decreasing well width.) Depending on details of sample quality, then, confinement could either increase or reduce excitonic dephasing times.

7.4 Results of Four Wave Mixing Experiments

Spectrally resolved FWM experiments were performed on samples of width 100, 200, 500, and 6000 Å. The measurements were performed over densities ranging from approximately 10^{15} - 10^{17} cm⁻³ (10^9 - 10^{11} cm⁻²). For each sample, measurements were performed both by sending the signal to a spectrally integrating detector and by spectrally resolving the signal with a spectrometer and an InGaAs optical multichannel analyzer. It turned out that for the signal intensities we obtained the OMA gave considerably better dynamic range than did the integrating detector, allowing for measurements at considerably lower densities. Thus data shown here is all obtained from the spectrally resolved measurements, which can of course be integrated to obtain total emitted intensity

as a function of time delay. I will discuss data from each of the samples in turn, starting with the narrowest quantum well, which has in many respects the simplest response.

The 100Å quantum well has its 1s heavy hole exciton peaked at 865 meV. This sample was excited with 15meV (FWHM) pulses centered around 850meV. The results of spectrally resolved FWM are shown in figure 7.2 (a) and (b) for both high (10^{11}cm^{-2}) and low (10^9cm^{-2}) density excitation. Figures 7.2 (c) and (d) show the spectrally integrated signal, which have exponential decays with a time constant of 325fs for low density excitation, and 190fs for the high density case. If we assume that the excitonic line in this case is inhomogeneously broadened, the two level system result for this sample then gives dephasing times of 750fs and 1.3ps for the high and low densities, respectively. The assumption of inhomogeneous broadening in this case is quite reasonable, as we have a ternary alloy sample at low temperature, which is likely to produce excitons which are somewhat localized at the various band-gap minima (see Chapter 3). If we take 1.3 ps for the limiting dephasing time, we obtain an upper limit for the homogeneous linewidth for the exciton of approximately 2meV. Absorption measurements of the exciton give linewidths at of least 3meV, thus indicating that the sample is, in fact, quite substantially inhomogeneously broadened. It is likely, in fact, that if we were able to make FWM measurements at even lower densities we would obtain even longer dephasing times, and thus narrower homogeneous linewidths.

Closer examination of the FWM spectrum for the high and low density cases reveals further dynamical information. Figure 7.3 shows the FWM spectra at high and low densities for $\Delta t=0$, overlaid with the pump spectrum. Note that with increasing density,

Figure 7.2 FWM signal from 100Å sample. (a) $N=10^{11}/\text{cm}^2$ (b) $N=10^9/\text{cm}^2$

Figure 7.2 Spectrally integrated FWM signal from 100A sample. (c) $N=10^{11}/\text{cm}^2$ (d) $N=10^9/\text{cm}^2$

Figure 7.3 FWM spectra of 100A sample taken at $N=10^9/\text{cm}^2$ (dotted) and $N=10^{11}/\text{cm}^2$ (dashed) overlaid with pump spectrum (solid).

the emission both broadens considerably and shifts towards the pump spectrum. The increase in breadth of the emission at high densities has been seen before in GaAs [5]. It is attributed to the fact that at 10^{11}cm^{-2} , the exciton is nearly saturated and thus the exciting pulses generate a substantial free carrier population. This causes both a sharp decrease in the dephasing time and a shift from excitonic emission only to exciton plus free carrier emission. In experiments on GaAs at room temperature, Bigot et. al. find a strongly assymetrical line shape of the exciton at low densities, indicative of a non-exponential temporal decay due to exciton-exciton interaction. We do not observe this deviation from a Lorentzian line shape, even at the lowest densities, which are comparable to those of Bigot et.al.. There are several possible explanations for this difference. The most likely stems from the fact that we make our measurements at liquid He temperatures, where the exciton lineshape is strongly inhomogeneously broadened. In this case, the broad distribution of resonant frequencies under the excitonic line masks any change in the lineshape of individual resonances due to interactions. Similarly, we see a simple exponential decay in the time domain measurements. It is also possible that exciton-exciton interactions are weaker in InGaAs than in GaAs, for example due to exciton localization in the ternary material.

The intermediate dimensionality samples (200 and 500Å) show behavior similar to that in the narrowest well, in that they have dephasing times which increase strongly with density. In these samples background scattering was a problem, and it was not possible to go to densities below 10^{10}cm^{-2} . Figures 7.4 and 7.5 show the spectrally integrated and spectrally resolved signals for each sample. We obtain a dephasing time for the 200Å wells

Figure 7.4 FWM signal from 200A sample. (a) Spectrally resolved (b) Spectrally integrated

of 530 fs at 10^{10}cm^{-2} , considerably shorter than is measured in the 100 Å sample at the same density. This is interesting in comparison with the perturbed free induction decay (PFID) data shown in Chapter 6, in which the 200 Å sample showed a considerably longer decay time than did the 100 Å sample. This difference is probably due to the fact that for inhomogeneously broadened lines, the PFID signal decay is affected both by the dephasing time and by the inhomogeneity of the line. The contribution of inhomogeneous broadening may be increased in the 100 Å sample due to increased localization from interface discontinuities, while its homogeneous linewidth is somewhat narrower than that of the 200 Å sample, due to the reduction in exciton Bohr radius and the corresponding reduction in exciton scattering from ionized impurities. This accounts for the fact that the linear absorption spectra show similar linewidths in the 100 Å and 200 Å samples, while the 100 Å sample has a longer dephasing time.

Interpretation of the dephasing times in the 500 Å sample is complicated by the fact that it shows a strongly non-exponential decay. This was also observed in the PFID data on this sample. We attribute this behavior to the presence of several very closely spaced resonances near the band edge of this sample. The heavy hole and light hole 1s excitons are just barely split from one another in the linear absorption. Although we do not resolve separate peaks from these two resonances in the FWM spectrum, we do observe a wide emission spectrum which does not coincide with the excitation spectrum. This wide, non-gaussian emission is likely to be due to the presence of (at least) two bound states which emit simultaneously and interfere to give a strongly non-exponential decay in time.

(a)

(b)

Figure 7.5 FWM signal from 500A sample. $N=10^{10}/\text{cm}^2$

(a) Spectrally resolved (b) Spectrally integrated

The FWM signals from the bulk sample taken at high and low density are shown in figure 7.6. In this sample we see a qualitative change in the FWM spectrum as we decrease the carrier density. At high densities (above 10^{17} cm^{-3}) the emission is very broad and decays quickly, within 100fs. As the density is lowered, however, a narrow peak emerges which has a much longer decay time. The broad emission is centered near $\Delta t=0$, while the narrow emission appears to be somewhat time delayed, peaking at approximately $\Delta t=150\text{fs}$. Figure 7.7 shows the narrow (late time) and broad (early time) signals superimposed on the linear transmission spectrum. The narrow line does not correspond to any feature observed in the linear absorption spectrum. If the sample is excited somewhat above the band edge only the broad response is seen. We interpret this in terms of two types of emission. The broad, instantaneous emission derives from the continuum of unbound states near the band edge. The longlived, narrow line is due to the 1s excitonic bound state, which is not resolved from the continuum in linear absorption. The decay time of the narrow line is similar to that found for the quantum well. Again, in this case we do not see a strong asymmetry of the FWM spectrum even at low densities, indicating that this narrow line is still strongly inhomogeneously broadened. The resolution of a narrow excitonic feature from a broadband continuum by the difference in dephasing time is the first such measurement to our knowledge. It explains in a very satisfying way the presence of resonant PFID signals in the ACSE and carrier thermalization data, despite the absence of any clear exciton in the absorption spectra.

Figure 7.6 · FWM spectra for 6000Å sample. (a) $N = 10^{15}/\text{cm}^3$ (b) $N = 10^{17}/\text{cm}^3$.

Figure 7.6 Spectrally integrated FWM intensity, 6000A sample.
 (c) $N=10^{17}/\text{cm}^3$ (d) $N=10^{15}/\text{cm}^3$

Figure 7.7 FWM spectra of 6000Å sample for $N=10^{15}/\text{cm}^2$. pump spectrum (solid), absorption spectrum (dotted), FWM at $\Delta t=0$ (dashed) FWM at $\Delta t=+300\text{fs}$ (dash-dot).

7.5 Conclusions

Our spectrally resolved FWM measurements reveal an interesting array of behaviors as one goes from the three dimensional to the two dimensional limit. Rather than obtaining phase relaxation times which evolve smoothly with confinement, we find that for each confinement regime (quasi 2D, intermediate, bulk) unique features in the FWM signal emerge, resulting from different aspects of the band edge. In the narrowest samples we find spectra which reflect emission from a single line, and which give an exponential decay in time. The decay times obtained from these samples indicate a phase relaxation time which increases with confinement. In the 500Å sample, the FWM spectra are dominated by the fact that in this intermediate dimensionality regime many resonances lie very close together. This results in emission from several resonances at once and a strongly non-exponential decay, indicating strong coupling among the resonances. In the bulk sample the presence of many resonances is again evident, this time in the simultaneous emission from both a bound excitonic state and a group of continuum states. The phase relaxation time for the bulk excitonic state is similar to that for the narrowest quantum well, indicating that there is not a strong change in phase relaxation time due to confinement alone. Our measurements thus show that the evolution of the band edge states as one goes from two to three dimensions is very complex, and that FWM as well as linear measurements are required in order to fully reveal the underlying processes.

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Chapter 8

Conclusions

8.1 Summary of results

We have designed and built a laser, amplifier and continuum generation system to produce 150fs continuum centered around 1.5 microns. The continuum extends from at least 300nm to 3 microns, and is relatively unchirped away from the fundamental. This system produces several nJ in a 30nm bandwidth for wavelengths ranging from 1.3 to 1.7 microns.

We have used this system to perform a series of studies of InGaAs heterostructures, ranging from 100Å quantum wells to bulk. Our studies of these samples have revealed a number of aspects of the evolution of ultrafast dynamics from two to three dimensions. We find that for densities on the order of $10^{10}/\text{cm}^2$, screening of the Coulomb interaction is relatively unaffected by confinement, contrary to predictions. As a result, thermalization times for electron distributions are approximately the same, 250fs, independent of confinement. In contrast, we find that the Pauli blocking nonlinearity at the band edge is approximately twice as effective in two as in three dimensions.

Purely coherent band edge effects, as measured by the AC Stark effect and four wave mixing, are found to be dominated by the changes in excitonic structure that take place with confinement. In particular, the AC Stark response is strongly affected by the change in Bohr orbital time scale that takes place as the exciton shrinks with confinement. Spectrally resolved four wave mixing results, which reflect polarization relaxation dynamics, show a wide variety of behaviors with changing well width. The signals clearly indicate that alloy induced inhomogeneous broadening is the dominant effect in the

excitonic line shape in these samples. Furthermore, coherent techniques reveal excitonic features in the bulk sample which are invisible in linear measurements. It is clearly necessary to go beyond linear measurements to fully understand the evolution of excitonic structure with confinement. Ours are the first studies of the evolution of ultrafast dynamics with dimensionality.

8.2 Future Prospects

The studies discussed in this thesis represent the first systematic exploration of the changes in ultrafast dynamics that take place with confinement. As such, they have revealed a number of interesting questions which we are unable to answer with our current system. Clearly, it would be advantageous to be able to explore a much wider range of carrier densities in our studies of carrier thermalization and cooling. This would allow us to explore the changes in carrier scattering which result from confinement over a range of regimes, from the strongly screened, metallic limit, to the weakly screened limit. We are prevented from doing this by signal to noise limitations, many of which originate with our additive pulse modelocked laser. Recently, lasers based on Cr^{4+} doped YAG have been developed which operate around 1.48 microns.[1] These lasers utilize Kerr lens modelocking, which is inherently far more stable and has less pulse to pulse jitter than does APM. The wavelength range of these systems is such that it would be possible to combine a Cr:Yag laser with our current amplifier system. This would result in a far quieter

continuum which could thus be used over a wider range of both intensities and wavelengths.

Recent work on high repetition rate Titanium sapphire laser/amplifier systems has resulted in a vast increase in understanding of solid state amplifier systems.[2] It might be possible to develop a similar system based on the NaCl F center. The increase in amplifier repetition rate which would result would allow a whole new range of experiments to be performed in the InGaAs system. In addition to the carrier thermalization and cooling experiments discussed above, these would include the high accuracy upconverted four wave mixing experiments which have recently been used in GaAs to reveal new aspects of the exciton exciton interaction and even the dynamics of phase.[3] It would be very interesting to perform experiments such as these on a series of samples ranging from two to three dimensions, as the evolution of the exciton-exciton interaction (which is governed by screening) would provide an excellent test of changes in the Coulomb interactions among carriers with dimensionality.

Our studies have revealed that for many types of interactions, the change in excitonic structure with confinement is very important to dynamics. InGaAs has particularly large excitons, allowing access to the strongly confined regime. It would be interesting to compare our experiments with a series of measurements of materials in which excitons are more strongly bound, such as GaAs and CuCl. Although it would be harder to reach the strongly confined limit for excitons in these systems, the continuum states would still become quasi-two dimensional. By comparing results in several

materials it would be possible to more clearly separate the effects of confinement of carriers from those due to changes in excitonic structure.

Finally, it would be nice to be able to answer the question I am most frequently asked at conferences. "Do you know anything about the evolution from two to one dimension?". One approach to addressing this would be to make a series of samples ranging from the two to the one dimensional limit. Unfortunately, the difficulties with production of reliable, narrow quantum wires still preclude doing a study of this type. Some understanding of the transition from two to zero dimensions has been obtained from studies of two dimensional systems placed under strong magnetic fields.[4] Similarly, three dimensional systems placed under high magnetic field become quasi one dimensional. However, with further studies of ultrafast dynamics under magnetic confinement it has become increasingly clear that magnetic fields change semiconductor states in very complex ways, and a simple analogy with "shrinking" the system by two dimensions is not sufficient.[5]

Developments in laser technology that have taken place over the years I have been in graduate school have already opened up many new possibilities for understanding the influence of confinement on ultrafast dynamics in the evolution from three to two dimensions. In a few years developments in growth techniques are certain to allow new understanding of the 2D to 1D and 1D to 0D transitions.

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Appendix 1

Details of PSF Calculation

Appendix 1: Mathematical Details of Phase Space Filling calculation.

The integral I_{3D} which appears in section 5.3 is given by $I_{3D} = \int_0^{\infty} \frac{\alpha^{3/2} e^{-\alpha} \sqrt{x}}{(1+x)^2} dx$.

The function is evaluated in figure 1, below.

Figure 1.1 Three dimensional integral, I_{3D} .

The integral I_{2D} is given by $I_{2D} = \int_0^{\infty} \frac{\alpha e^{-\alpha}}{(1+x)^{3/2}} dx$. The function is evaluated in figure 2,

below.

Figure 1.2 Two dimensional integral, I_{2D} .

Appendix 2

Nonlinear Dynamics in the APM Laser

Appendix 2

The principles of nonlinear dynamics and chaos provide useful tools for understanding the complex behaviors of real systems. This is especially true for optical systems where nonlinear dynamics have proven most useful in understanding laser instabilities. Although instabilities in femtosecond lasers have been frequently studied, only a few such studies [1,2] relate these instabilities to contemporary principles of nonlinear dynamics. Establishing such relationships is vital to obtaining a better understanding of the stability (and instability) regions of modelocked lasers.

It is known, though rarely mentioned in the literature, that excessive fiber nonlinearity causes severe instabilities in additive-pulse modelocked (APM) lasers. However, the nature of the path to instability has not been studied. Given the highly nonlinear nature of APM lasers, it should not be surprising that they can exhibit chaos and other complex dynamics. In this appendix, I will discuss our observation of period-doubling and quasi-periodicity in an additive-pulse modelocked NaCl F-center laser. Using numerical modeling, we have examined these paths to instability in APM lasers. While our work concentrates on APM lasers, the principles discussed here are relevant to other types of passively modelocked lasers which use some form of fast saturable absorber modelocking. That this is so, is borne out by our observation of period-doubling in a self-modelocked Ti:Sapphire laser as well.

Experiments were performed with an NaCl F-center APM laser [3,4] shown in figure 1. This is the Fabry-Perot APM configuration, as opposed to the Michelson configuration which is also widely used. The laser has an output coupler with $T=45\%$,

Figure A2.1 Schematic of NaCl APM laser cavity. Main cavity contains the gain (NaCl), birefringent tuner plate (BTP), and output coupler (OC). The control cavity contains 10cm of single mode fiber (SMF), and a 50% beamsplitter (BS). The cavity length is adjusted via the end mirror, M_2 , which is mounted on a piezoelectric transducer (PZT).

(sometimes $T=30\%$ is used) and the control cavity contains a beamsplitter which sends 55% of the laser power to a 10 cm length of dispersion-shifted fiber ($\lambda_0=1.55$ microns). Both cavities are 2 meters in length, corresponding to a repetition rate of 76 MHz (i.e., a pulse repetition period of 13 nsec), and "synchronous pumping" is provided by a modelocked Nd:YLF laser which has a repetition rate of 76 MHz. Without the external cavity, the synchronously pumped NaCl laser produces pulses of approximately 10 psec duration. With the external cavity coupled to the main cavity, the APM laser becomes passively modelocked and produces pulses of 120 fsec duration, as measured by noncollinear autocorrelation. The pulses are tunable from 1550-1620 nm.

Normal modelocked behavior is shown in figure 2(a), while figure 2(b) shows the period-doubling behavior. This is a stable mode of operation, and it can be produced most readily by increasing the laser power and adjusting the cavity-length mismatch to lock on to any one of several fringes which support this behavior. The period-doubled mode is most easily observed by fully resolving the pulse train on a fast (<1 nsec) photo diode; however, one must take care to properly trigger the scope to discriminate between the large and small pulses, or else the pulse train will appear "normal" as in figure 2(a). One cannot rely on the measured pulse autocorrelations or optical spectra, as they *give no evidence of this behavior*. The period-doubling sets in when the output power from the main cavity exceeds $P_{\text{main}} = 240$ mW. At this power level, we couple about 80 mW of power into the fiber, although the actual power during APM operation is slightly less. Note that this period-doubled operation is not to be confused with multiple pulsing--there is only one pulse in each cavity at any given time.

Figure 2

Figure A2.2 Pulse train under (a) normal modelocking conditions, (b) period-doubled modelocking, and (c) quasiperiodic modelocking.

As with femtosecond modelocking, period-doubling is a direct consequence of coupling to the control cavity. If the control cavity is blocked, the laser reverts back to active modelocking (10 psec pulses) and period-doubling does not occur. In addition, period-doubling only occurs when the laser is passively modelocked in the femtosecond regime.

At higher laser power, ($P_{\text{main}} > 300$ mW) the APM laser produces a quasiperiodic pulse train, shown in figure 2(c). This form of quasiperiodicity couples period-doubling with a 5 Mhz modulation. In fact, the large pulses evolve into small pulses and back again with a periodicity of ~ 5 Mhz. This modulation does not necessarily correspond to a subharmonic of the 76 Mhz fundamental. The observation of such a pulse train requires very delicate triggering of the scope.

In order to understand the nature of these instabilities, we have performed numerical simulations of the APM laser using a laser model described by the recursion relations[6]

$$\begin{aligned} a_1^N(t) &= \hat{G}\hat{B}b_1^N(t) \\ a_2^N(t) &= \gamma^2 b_2^N(t) \exp \left[i \left(\Phi_0 + \gamma^2 n_2 |b_2^N(t)|^2 \right) \right] \\ b_1^{N+1}(t) &= r a_1^N(t) + \sqrt{1-r^2} a_2^N(t) \\ b_2^{N+1}(t) &= \sqrt{1-r^2} a_1^N(t) - r a_2^N(t) \end{aligned}$$

where $a_1(t)$, $a_2(t)$, are the pulse amplitudes incident on the output coupler, and $b_1(t)$, $b_2(t)$ are the pulse amplitudes reflected from the output coupler. The subscripts 1

and 2 correspond to the amplitudes in the main and control cavities, respectively. The superscript, N , is the round trip index. Each of these pulse amplitudes is a 1024-point complex vector. The operator $\hat{G}$ represents the effects of saturable gain ($\hat{G} = \exp[g_0/(1+E/E_{\text{sat}})]$) where g_0 is the unsaturated gain, E_{sat} is the saturation energy, and $E = \int |b_1(t)|^2 dt$ is the pulse energy in the main cavity. The operator $\hat{B}$ represents bandwidth limiting by a Lorentzian filter function. Equation 1(b) represents the self-phase modulation which occurs in the fiber. The output coupler reflectivity is r^2 , and γ^2 is the reflectivity of the beamsplitter. We used values of $\gamma^2 = .55$, $g_0 = 0.5$, $E_{\text{sat}} = 30$, and $r^2 = .55$ (for 45% OC) or $r^2 = .70$ (for 30% OC). The normalized Kerr nonlinearity in the fiber, n_2 , is varied from 0 to 10. These equations contain two simplifying assumptions with respect to those of ref.6: (i) dispersionless propagation in the fiber and main cavities and, (ii) near perfect temporal overlap of the main cavity pulse, $a_1(t)$, and the pulse returning from the control cavity, $a_2(t)$, when they recombine interferometrically at the output coupler. This corresponds to the case of zero cavity-length mismatch, except for a static phase bias ($\Phi_0 = -2.5$ radians). The zero-dispersion approximation is justified by the fact that we are operating the laser very near the zero-dispersion point of the fiber in the external cavity, and by the fact that the main cavity has very little group velocity dispersion (GVD). [7] However, unlike the analytic theory developed in ref. 6 we do not make approximations to the operators, nor is any self-consistent solution assumed.

These simulations reveal several interesting behaviors. Figure 3 shows a bifurcation diagram in which we plot the control cavity pulse energy, $E_{\text{fib}} = \int |b_2(t)|^2$, versus the kerr nonlinearity, n_2 , using a 45% output coupler. Note that the output

pulse energy (as seen experimentally) is related to the control cavity by $E_{\text{out}} = \gamma^2 E_{\text{fib}}$. As the fiber nonlinearity is increased from zero, the (simulated) laser proceeds from CW lasing, to stable modelocking (pulsewidth decreasing as n_2 increases) to period-doubled modelocking, and then a sudden transition to apparent chaotic behavior. As the nonlinearity is then decreased, this transition exhibits a hysteresis. If we use a 30% output coupler in the simulation, we see bifurcations up to period-8 (limited by sampling interval of n_2) before chaos sets in. Figure 3 also shows that period-doubling occurs over a range of nonlinearity values which is broader than the range for normal modelocking. In the actual laser, period doubling sets in when the output power of the main cavity is about 240 mW, and continues on up at least through 350 mW (the upper limit of our measurements), although more complex behavior (quasiperiodicity) can also set in at about $P_{\text{main}}=300$ mW depending upon which fringe the APM is locked onto. While the analogy between increasing power and increasing nonlinearity is not exact, it does give some indication of agreement between theory and experiment.

When we examine the pulse solutions in the region of period-doubling, we find that the large pulses (corresponding to the upper branch of fig. 3) have a different intensity profile and spectrum than the small pulses, which correspond to the lower branch. This pulse reshaping is illustrated in figure 4(a) which shows the simulated pulse intensity profiles of the "upper-branch" and "lower-branch" pulses in both the main and control

Figure A2.3 Bifurcation diagram generated by simulations of laser pulse energy (in the control cavity) versus fiber non-linearity, n_2 , for an APM laser with a 45% output coupler ($r=.8367$) and 55% beamsplitter ($\gamma=.7416$) and phase bias $\Phi_0=-2.5$

cavities. Figure 4(a) also indicates that the control cavity pulse energy is a maximum when the main cavity pulse energy is a minimum, and *vis versa*, suggesting that the coupled cavities exchange a parcel of excess energy back and forth on alternating round trips. Figure 4(a) shows the intensity profile of this "exchange pulse" (*i.e.*, the large pulse intensity minus the small pulse intensity) in the main and control cavities. We find that this difference is very nearly the same for the two cavities, even though the main cavity pulse energy is substantially larger than the control cavity pulse. The energy exchange between the cavities has been verified experimentally by placing photodiodes symmetrically about the output coupler and monitoring the pulse trains of the main and control cavities simultaneously. The pulse trains, shown in figure 4(b), are in perfect anti-phase, clearly demonstrating the exchange of energy between the two cavities. Note also that the pulse contrast ratio is larger for the control cavity than for the main cavity. This is also predicted by the simulations.

Quasiperiodicity is not seen in the simulations if perfect pulse overlap is assumed, but it is observed when a cavity length mismatch corresponding to about 10% of the final pulsewidth is assumed. This produces a temporal mismatch of the pulse envelopes in the two cavities. The pulse reshaping which occurs under quasiperiodic operation is considerably more complex than that associated with period-doubling, and will not be discussed further in this letter.

A few additional points are worth emphasizing. First, the calculated pulse intensity autocorrelations and optical spectra for the simulated laser output show no evidence of the period-doubled behavior and deviate only slightly from transform limited behavior. Thus,

Figure A2.4 (a) Calculated pulse intensity profiles $|b_1(t)|^2$ and $|b_2(t)|^2$ for both the “upper branch” pulses (heavy solid) and the “lower branch” pulses (thin solid) in both the main and control cavities. The “exchange pulses” are shown to the left of these (thin solid). (b) The measured pulse trains in the main and control cavities (lower and upper traces, respectively) confirm that energy is exchanged between the cavities.

the simulations predict that these commonly used diagnostics fail to inform the user that this behavior is occurring, which is in total agreement with experimental observations.

Second, because there is no dispersion in the model (and very little in the laser), soliton effects are not present, so the period-doubling instability is *not* due to high-order soliton effects which have been reported in self-modelocked Ti:Sapphire and CPM lasers. [8,9]

Third, these instabilities are produced by the same fast saturable absorber mechanism which provides the pulse shortening, and are not related to rate equation or laser-Bloch equation dynamics which are predominant in most other lasers which have been studied for their complex dynamics.

While it has been demonstrated that instabilities are caused by excessive nonlinearity, the question remains as to *how much* nonlinear phase shift causes them. Intuitively, we expect that peak nonlinear phase shifts of $\phi_{nl} \cong 2\pi$ would cause severe pulse distortion upon recombination with the main cavity pulse at the output coupler, and might therefore lead to instability. The simulations show that the first bifurcation occurs at a peak nonlinear phase shift of only $\phi_{nl} \cong 1$ radian. This is true for both the 45% and 30% output couplers. This is surprising in light of the fact that various groups [8,9] have reported stable operation of APM lasers even with peak nonlinear phase shifts well in excess of $\phi_{nl} \cong \pi$. In recent experiments, Steinmeyer *et al* [10] directly measured the dynamical behavior of this type of interferometric pulse recombination in a nonlinear ring cavity injected with picosecond pulses. They observed the first bifurcation when the peak nonlinear phase shift reached $\phi_{nl} \cong \pi$.

Although there have been very few systematic studies of instabilities in APM lasers, there have evolved some "common-sense" guidelines for obtaining stable modelocking, such as using sufficient bandwidth limiting and avoiding excessive amounts of nonlinearity in the fiber. We have shown that these guidelines can be linked to familiar principles of nonlinear dynamics. This should enable us to better understand instabilities in passively modelocked lasers which use coupled-cavity fast saturable absorbers.

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