THE PHYSICS AND FEMTOSECOND APPLICATIONS
OF PHOTOREFRACTIVE MULTIPLE QUANTUM WELLS

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Robert Mark Brubaker

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To all those who make the struggle worthwhile.
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Although there is only one name on this thesis, it represents countless contributions from many people besides the author. Of course, the support and inspiration of Prof. David D. Nolte was critical for success. This work follows the footsteps of Dr. Qingnan Wang, who discovered nonreciprocal energy transfer and developed the hot-electron model, in addition to his other work on the photorefractive effect. Prof. M. Melloch generously provided all the AlGaAs growths. Prof. A. Weiner provided guidance for the femtosecond pulse diffraction in the last chapter as part of our focused research initiative to shape pulses with photorefractive quantum wells. Prof. D. Elliott taught the author basic laser physics and nonlinear optics. Eastman Kodak Corp. provided support for a Kodak Fellowship, which included funds for equipment and travel. María Aguilar measured the two-wave mixing spectra, and Indrajit Lahiri performed the moving grating experiments, both in collaboration with the author. Thanks also goes to all the students who have been part of the lab: Ranbir Rana, Nien-Po Chen, Karrin Kwolek, Mihaela Dinu, Indrajit Lahiri, John McKenna, María Aguilar, John Coy, Dave Crouse, Peter Rakich and others. Good luck to Yi Ding, the post-doc who is building on the work with shaping pulses. Thank to the committee (Prof. Nolte, Prof. Melloch, Prof. Grabowski, Prof. Muzikar) for their patience and insightful questions, the staff of the physics department (Linda Gretter, Virginia Messick, Sandy Formica, Judy McKinley and others) for keeping the author on track and out of trouble, and other students (Dennis Krause, Dave Schaefer, Dan Lovall and others) for their fellowship and good humor.
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ABSTRACT


Photorefractive quantum wells are dynamic holographic devices that operate at low optical intensities. A spatial variation in the incident light creates a corresponding refractive index and absorption pattern. Photocarriers are generated, then transport by diffusion and drift, and trap at deep level defects. The resulting space charge field is converted into a diffraction grating by an electro-optic effect. The thin-film grating is probed in the Raman-Nath regime by degenerate four-wave mixing, two-wave mixing, and nondegenerate four-wave mixing. The material system is Aluminum Gallium Arsenide epilayers and AlGaAs/GaAs multiple quantum wells grown by molecular beam epitaxy. The devices are operated in the transverse or Franz-Keldysh geometry, for which an electric field is applied in the plane of the quantum wells (perpendicular to the growth direction). The transport is parallel to the grating vector, and the electro-optic effect is the field ionization of excitons. The purpose of the first part of this work was to complete the study of transverse geometry photorefractive structures. The main result is conclusive evidence proving that hot-electrons cause the photorefractive phase shift which produces nonreciprocal energy transfer. The electron velocity-field nonlinearity is shown to correspond to the onset of the photorefractive phase shift. The second part demonstrates that these devices are useful for characterizing and controlling ultrashort pulses with a duration of 100 fs using nondegenerate four-wave mixing. Pulses were characterized using photorefractive quantum wells to perform electric-field correlation where the pulses write the grating in the photorefractive quantum wells, and a continuous wave diode laser probes the grating. The shape of a femtosecond pulse diffracted from photorefractive quantum wells is measured when a continuous wave diode laser writes the grating, and the femtosecond pulse probes the grating. The shape of the diffracted pulse is measured with electric-field cross correlation and spectral interferometry. The diffracted pulse is broadened by the reduction of its spectrum from the bandwidth of the
diffraction spectrum, but remains nearly transform limited. Therefore, photorefractive quantum wells are suitable for use in a spectral holography pulse shaping apparatus.
1. INTRODUCTION

For many optical processing applications, an ideal optical nonlinearity operates at low incident intensities with a fast response time. In addition, the nonlinearity should occur in a convenient material, such as semiconductors which can be grown with precise control, and integrated into devices with electronics or other optical processing elements. The work presented in this thesis is an investigation of a particular optical nonlinearity, the photorefractive effect, in the Aluminum Gallium Arsenide system, which possesses many desirable properties for nonlinear optical processing. The investigation is motivated by the application of the photorefractive effect to characterize and control femtosecond laser pulses.

To obtain an optical nonlinearity of a desired magnitude, there are trade-offs between interaction length, speed, incident intensity and the strength of the nonlinearity. Fiber optic wave guides posses small nonlinearities in the glass core, but offer extremely long interaction lengths in a strongly confined path. Shorter wave guides are also used in materials with larger nonlinearities. Second harmonic generation is accomplished on the scale of millimeters or less by focusing a beam into the crystal. Bulk photorefractive materials are several millimeters thick, while photorefractive quantum wells are only a few micrometers thick.

The photorefractive effect is a change in the refractive index driven by the spatial variation of an illumination pattern. The illumination pattern is created by the interference of two beams, and therefore the process is holographic. The simplest possible hologram is the interference of two lowest mode gaussian beams, which are essentially plane waves. More complex patterns are generated by interfering beams that have some transverse spatial pattern, such as an image, on one or both beams. Operations such as joint Fourier transform image correlation have been demonstrated in photorefractive quantum wells.

The photorefractive effect operates through the generation, transport and trapping of charge at deep level defects, which creates a space-charge distribution, and through an electro-optic effect, which converts the space-charge electric-field into a change in refractive
index. Since charges are promoted across the bandgap (not a virtual transition), this effect is resonant, and not instantaneous. The response time depends on the incident intensity, so one can integrate a small intensity for a long time, or use high intensities to operate at short response times.

The dynamic holographic processing of images is an important and inherent property of the photorefractive effect. The processing and switching of two dimensional images is potentially useful in communications, as well as other applications. The parallelism of image processing makes the notion of an 'optical computer' distinct from the digital electronic computer. If one attempts to make an optical computer that mimics a digital electronic computer, there are several fundamental roadblocks. Photons are difficult to store, since there is no compact optical 'capacitor'. Photons are difficult to make to interact. A nonlinear medium is necessary to control one photon with another photon. Therefore, it is difficult to make an 'optical transistor'. To process an image, the parallelism of an image allows the processor to have a much lower 'clock speed' than the serial digital computer that operates on each pixel separately. Serial processing would not be the best use of an all optical device.

The photorefractive effect was found in LiNbO3, and was initially considered to be optical damage because it was an undesirable effect at the time. Other ferroelectric crystals have been investigated extensively, as well as many semiconductor systems. The ferroelectric crystals generally have large electro-optic effects, but suffer from long time constants (from seconds to hours or years!) due to the small carrier mobility. Semiconductors, on the other hand, have much higher mobilities, and therefore much smaller response times, but have weaker electro-optic effects. Semiconductors also have the advantage of highly refined growth techniques, both for bulk crystals as well as atomic layer control of multilayer structures. Semiconductor systems can be integrated with existing electronic systems, such as sources and detectors, for packaging in a device using the current technologies of photolithography and monolithic integration.

The photorefractive response of a material can also be used to study its solid state properties, that is, the transport and trapping of carriers, which include carrier dynamics and deep level defect physics. The photorefractive response in GaAs with EL2 has been used to study Fermi-level modulation, and the photorefractive effect in diluted magnetic semiconductors can be controlled with an applied magnetic field. Phase conjugation has also been demonstrated, where the wave front of a beam can be reversed. The system is fundamentally nonlinear from nonlinearities in the transport equations, and the electro-optic effect.
The material system used in this thesis is the AlGaAs system grown by molecular beam epitaxy (MBE). The interaction length is on the order of a micron, so a very large nonlinearity is used over this small interaction length. The wavelengths used have a photon energy near the bandgap of the semiconductor so that the effect is resonant, and depends on strong absorption. The structure of the crystal is either epilayers or a series of quantum wells grown into the crystal by varying the Al fraction of the material. This system is generally well understood and is grown with precise control and high quality (few defects and few impurities). The electro-optic effect used in these thin film quantum well structures depends on the manipulation of the oscillator strength of the excitons. The effect considered in this work is exciton field ionization, but there is also the quantum confined Stark effect, and other effects in bulk materials.

This work on transverse-field photorefractive quantum wells is only one aspect of photorefractive quantum wells. The alternate perpendicular geometry uses the quantum confined Stark effect, with the transport perpendicular to the grating vector. Intentional design of the structure to be a Fabry-Perot cavity can increase the diffraction efficiency, and lead to other micro cavity effects. For communications technology, a structure has been designed in another material system for 1.55 μm wavelengths. For design of diffraction with wide bandwidths, Fibonacci sequences of quantum wells have been explored, raising challenging questions about the engineering of the spectrum for oscillator strength. Overall, the research spans from the basic physics to proof-of-principle for applications.

This thesis is structured as follows: §2 contains an explanation of the quantum well structure and its basic optical properties, including the electro-optic effect. The photorefractive effect and the wave mixing used to probe the grating are presented in §3, with the most important result being the confirmation of a theory for the unexpected two-wave mixing in these structures. §4 makes a transition from the physics of these structures to applications, as well as a change from CW lasers to pulsed laser use. The use of photorefractive quantum wells as an femtosecond pulse electric-field correlator is shown. Finally, §5 describes the diffraction of femtosecond pulses from quantum wells, with the objective of controlling the shape of a femtosecond pulse by manipulating its Fourier components, that is, the amplitude and phase of its wavelength spectrum.
2. TRANSVERSE-FIELD ELECTRO-OPTIC SPECTROSCOPY OF ALUMINUM GALLIUM ARSENIDE QUANTUM WELLS AND THIN FILMS

2.1. Introduction

This chapter contains a comprehensive study of transverse-field electro-optic properties of GaAs and AlGaAs quantum wells and epilayers, for five representative structures spanning a range of Al fractions. Quantum well structures are known to show large electroabsorption in the quantum-confined Stark geometry [1-5], but epilayers and quantum wells also have appreciable electroabsorption in the transverse-field geometry [3, 6-8]. The photorefractive effect in thin semiconductor films was first explored in quantum wells [9] in a transverse-field geometry. However, homogeneous epilayers also have strong photorefractive responses, including two-wave mixing [10]. The properties of excitons are discussed in §2, the description of sample growth and transmission spectra are given in §3, and the electro-optic coefficients are reported in §4.

2.2. Excitons in Epilayers and Quantum Wells

2.2.1. Exciton Energy Levels

The exciton is a bound state of an electron and a hole [11]. It is the lowest energy excitation of a perfect semiconductor crystal. In the AlGaAs system, excitons are analogous to hydrogen atoms, with the hole playing the role of the proton. When ionized, the electron and hole are free to travel infinitely far from each other. The wavefunction is spread out over many lattice sites, so the Coulomb binding is modified by the relative dielectric constant $\varepsilon$ of the material. The energy for the $n^{th}$ level is

$$E_n = -\frac{\mu e^4}{2\hbar^2 \varepsilon^2 n^2} = -\frac{\mu}{m_e \varepsilon^2} \frac{1 \ R}{n^2} \quad (2.1)$$
in CGS units, where \( m_e \) is the free electron mass, \( R = 13.61 \text{ eV} \) is the Rydberg and \( \mu \) is the reduced mass of the exciton

\[
\frac{1}{\mu} = \frac{1}{m_e^*} + \frac{1}{m_h^*}
\]

(2.2)

where \( m_e^* \) and \( m_h^* \) are the electron and hole effective masses, respectively. The separation of the electron and hole is

\[
a_n = \frac{n^2 \hbar^2}{\mu(e^2/\varepsilon)} = n^2 e^2 m_e \frac{\mu}{\varepsilon} a_0
\]

(2.3)

where \( a_0 = 0.529 \text{ Å} \) is the Bohr radius. For GaAs, \( m_e^* = 0.067 m_e \), \( m_h^* = 0.62 m_e \) for the heavy hole, and \( \varepsilon = 13.18 \) [12], so \( E_1 = -4.7 \text{ meV} \) and \( a_1 = 115 \text{ Å} \). At room temperature, the phonon energy of 25 meV easily ionizes the exciton, making it difficult to see an exciton absorption feature in the transmission spectrum. At low temperatures, the exciton is clearly resolved [13].

The binding energy of an exciton is greatly increased by spatial constraint in a quantum well structure, as shown in Fig. 2.1 for Al\(_{0.3}\)Ga\(_{0.7}\)As barriers and GaAs wells. The band offset between the barrier and well confines both types of carriers to the same layer, and the structure is called a Type I quantum well. If the band offset traps the carriers in different layers, the structure is a Type II quantum well.

If the barrier were impenetrable, narrowing of the well width could confine the exciton to be two-dimensional, with a ground state Coulomb binding energy of \( 4E_1 \) [14]. However, the penetration of the exciton wavefunction into the barriers only allows an increase of the Coulomb binding energy to approximately \( 2.5E_1 \), or 10 meV for AlGaAs quantum wells. A simple calculation shows the basic physics of excitons in a well (following the Ben Daniel-Duke model [15]). Each type of carrier can be considered independently in its respective well to find their confinement energies without Coulomb binding. The only complication from the usual textbook calculation is the different effective masses in the well and the barrier. For a well with depth \( V > 0 \) and width \( L \) the bound state wavefunction is
Fig. 2.1. Energy-band diagram for quantum confined excitons. The energy levels shown do not include Coulomb binding. The transition shown is between the electron and heavy hole ground state (n=1) levels. The ground state wave functions are schematically depicted, with the exponential tails penetrating into the barriers.

\[
\Psi(z) = \begin{cases} 
  A \exp(\beta z), & z < -L/2 \\
  B \cos(\alpha z) + C \sin(\alpha z), & |z| < L/2 \\
  D \exp(-\beta z), & z > L/2 
\end{cases}
\] (2.4)

where

\[
\alpha = \sqrt{\frac{2m_w}{\hbar^2} (E + V)} \quad \text{and} \quad \beta = \sqrt{\frac{2m_b}{\hbar^2} (-E)}
\] (2.5)

and \( m_w \) and \( m_b \) are the effective masses of the carrier in the well and the barrier, respectively, and the energy level \( E < 0 \) is measured from the top of the barrier. The solution of the time-independent Schrödinger equation with boundary conditions
\[ \Psi(z) \quad \text{continuous} \]
\[ \frac{1}{m(z)} \frac{\partial \Psi(z)}{\partial z} \quad \text{continuous} \]  

yields equations for the energy levels

\[ \frac{\beta}{m_B} = -\frac{\alpha}{m_W} \cot \left( \alpha L / 2 \right), \quad \text{odd parity} \]
\[ \frac{\beta}{m_B} = \frac{\alpha}{m_W} \tan \left( \alpha L / 2 \right), \quad \text{even parity} \]  

which are similar to those for spatially invariant masses. For a Al\textsubscript{0.3}Ga\textsubscript{0.7}As barrier and GaAs well, the difference in the band gap energy between the barrier and the well is 374 meV, which is distributed 65% to the conduction band offset and 35% to the valence band offset, so that the well depth is \( V_e = 243 \) meV for the electron, and \( V_h = 131 \) meV for the hole. For a 75 Å well width, the solution to Eq. 2.7 is \( E_e = -197 \) meV for the electron. In a quantum well, the degeneracy of the valence band is lifted. The different effective masses for the light and heavy holes result in different energy levels. The energy for the heavy hole is \( E_{hh} = -123 \) meV and \( E_{lh} = -99 \) meV for the light hole. The energy of the exciton transition, not including Coulomb binding, is just the bandgap of the barrier plus the electron and hole energies. For the heavy hole exciton, the transition energy is 1.477 eV, and for the light hole exciton it is 1.501 eV. Excitonic transition energies for other barrier heights are shown in Fig. 2.2, using the selection rule \( \Delta n = 0 \). The exciton transition energy for various well widths is shown in Fig. 2.3. These values are only estimates to gain insight into the system.

2.2.2. Excitons in an Electric Field

The application of an electric field to a semiconductor changes the absorption of the material for photon energies near the bandgap. This effect is the Franz-Keldysh effect [16, 17]. The original calculations did not include the contribution of the exciton to the Franz-Keldysh effect, which greatly increases the magnitude of the absorption change [18]. The Franz-Keldysh effect induces a large change in the absorption near the band gap, and an oscillating change for energies higher than the gap.
Fig. 2.2. Quantum well exciton transition energy vs. barrier Al fraction for 7.5 nm wells.

Fig. 2.3. Quantum well exciton transition energy vs. well width for 30% Al barriers.
Without Applied Field  

\[ \text{In-plane e-h Pair Potential} \]

With Applied Field  

\[ \text{Exciton Field Ionization} \]

Field Geometry  

\[ \text{Electric-field} \]

\[ \text{Quantum Confined Stark Effect} \]

CB  

Quantum Confined Potentials

VB

Fig. 2.4. Electro-optic effect geometries. Exciton field ionization occurs when the field is applied parallel to the quantum well planes (perpendicular to the growth direction). Field ionization is similar in homogeneous and quantum well materials. The quantum confined Stark effect occurs when an electric field is applied perpendicular to the quantum well planes (parallel to the growth direction).

The anisotropy of quantum wells defines two directions, one parallel to the wells, and one perpendicular to them, as shown in Fig. 2.4. Application of a field perpendicular to the wells gives rise to the quantum confined Stark effect (QCSE) [2, 3]. The energy of the exciton is shifted to lower energy, and the shift is a quadratic function of field, to lowest order. Eventually, as the magnitude of the field is increased, the carriers can tunnel out of the wells. The decrease of the lifetime increases the linewidth of the transition. This lifetime broadening is only of secondary importance compared with the QCSE.

With the field applied parallel to the wells, the effect is similar to the bulk Franz-Keldysh effect, where the field acts to reduce the lifetime of the exciton by lowering the relative Coulomb potential [19]. This can be described as exciton field ionization. Because the direction of the applied field should produce no difference in the effect, the change in
absorption is quadratic in field to lowest order. At high fields, the energy of the exciton is shifted, but this is a much weaker effect than the lifetime broadening.

The absorption spectrum $\alpha(\lambda)$ and the refractive index spectrum $n(\lambda)$ of the material are related by the Kramers-Kronig transformations [20]

$$n(\lambda) = 1 - \frac{\lambda^2}{2\pi^2} P \int_0^\infty \frac{\alpha(s)}{s^2 - \lambda^2} ds$$

$$\alpha(\lambda) = 8P \int_0^\infty \frac{n(s)}{s^2 - \lambda^2} ds$$

(2.8)

where P is principal value, and the functions are given in terms of wavelength for convenience in analyzing the data. When an electric field is applied to the sample, both the materials absorption and refractive index change. With the notations

$$\Delta n(\lambda, F) = n(\lambda, F) - n(\lambda, 0)$$

$$\Delta \alpha(\lambda, F) = \alpha(\lambda, F) - \alpha(\lambda, 0)$$

(2.9)

the changes in absorption and refractive index can also be related by

$$\Delta n(\lambda) = -\frac{\lambda^2}{2\pi^2} P \int_0^\infty \frac{\Delta \alpha(s)}{s^2 - \lambda^2} ds$$

$$\Delta \alpha(\lambda) = 8P \int_0^\infty \frac{\Delta n(s)}{s^2 - \lambda^2} ds$$

(2.10)

which are useful because the changes are only non zero over a narrow range near the band gap. In contrast, for calculating refractive index from the absorption, it is necessary to know or estimate absorption at all wavelengths. (Actually, for calculating changes, other transitions should ideally be taken into account, but they are either far from the gap or weak, like the $n = 2$ transition.)

2.3. Structure Growth and Transmission Spectra

The electro-optic and photorefractive properties of five different multiple quantum well (MQW) and epilayer structures are presented here. Two of the structures are
Proton Implanted  
$10^{12}$ cm$^{-2}$ at 160 keV  

<table>
<thead>
<tr>
<th>5 nm</th>
<th>GaAs</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 nm</td>
<td>Al$<em>{0.2}$Ga$</em>{0.8}$As</td>
</tr>
<tr>
<td>60</td>
<td>10 nm</td>
</tr>
<tr>
<td>7.5 nm</td>
<td>GaAs</td>
</tr>
<tr>
<td>50 nm</td>
<td>GaAs</td>
</tr>
<tr>
<td>100 nm</td>
<td>Al$<em>{0.2}$Ga$</em>{0.8}$As</td>
</tr>
<tr>
<td>50 nm</td>
<td>AlAs</td>
</tr>
<tr>
<td>1 μm</td>
<td>GaAs</td>
</tr>
<tr>
<td>n$^+$ GaAs Substrate 052992C</td>
<td></td>
</tr>
</tbody>
</table>

(a)  

Multiple Quantum Wells (1.05 μm)  
Incident Light  

(b)  

Fig. 2.5. Sample structure and preparation: (a) sample structure diagram, with growth proceeding upward from the substrate; (b) sample mounted on glass slide.

homogeneous epilayers of GaAs and of Al$_{0.2}$Ga$_{0.8}$As, and three structures are Al$_x$Ga$_{1-x}$As multiple quantum well structures, all with 60 periods of 7.5 nm GaAs wells and 10.0 nm Al$_x$Ga$_{1-x}$As barriers, where x is 0.05, 0.10 and 0.30. The sample structure diagram is shown in Fig. 2.5(a). All five structures were grown by molecular beam epitaxy in a Varian Gen II chamber on 0.5 or 1 μm of GaAs on a GaAs substrate, with stop-etch and selective etch layers. The MQW and epilayers were 1 μm thick and sandwiched between 100.0 nm layers of either Al$_{0.2}$Ga$_{0.8}$As (for the $x = 0.05$ and $x = 0.10$ multiple quantum wells) or Al$_{0.3}$Ga$_{0.7}$As. A 5 nm GaAs cap was deposited on top of the growth to protect the device from oxidation.

After growth, each wafer was proton implanted to make the material semi-insulating. The protons themselves are not important, rather it is the interstitial and vacancy defects that they create. A photorefractive semiconductor must be semi-insulating in order to slow dielectric relaxation, and to provide deep traps to store the photo-induced space-charge [21]. The proton implant was made on the episurface with 160 keV protons at a dose of $10^{12}$ cm$^{-2}$. Three deep level defects are created on average by each proton [22] resulting in a defect density of ~3x10$^{16}$ cm$^{-3}$, which is sufficient to compensate residual~10$^{15}$ cm$^{-3}$ shallow defects. The defects are not dense enough to broaden the room temperature spectra [23]. The samples were cleaved to approximately 3 mm x 3 mm, the substrates were removed using a lift-off technique [24], and the free-standing films were
Fig. 2.6. Experimental configuration for transmission and differential transmission measurements. The long pass filter blocks harmonics of the fundamental frequency. The lockin amplifier is referenced to the chopper for transmission measurements, and to the applied field for differential transmission measurements.

"Van der Waals" bonded [25] to glass microscope slides, as shown in Fig. 2.5(b). The samples were not anti-reflection coated. Gold contacts were evaporated on the samples with a 1 mm spacing, across which the electric field was applied in the plane of the wells. The device resistivities were \( \sim 10^7 \ \Omega \) cm for total sample resistances of several gigaohms.

The absorbance spectrum for each sample was obtained from the measured transmission using the equation

\[
\alpha(E) = -\frac{1}{L} \ln \left( T(E) \right)
\]

where \( L \) is the thickness of the sample and \( E \) is the photon energy. The transmission was measured using a 600 W Tungsten source, a SPEX 1403 Czerny-Turner 0.85 m spectrometer and a biased silicon diode photodetector with lock-in detection, as shown in Fig. 2.6. The room-temperature absorption spectra are shown in Fig. 2.7 for each sample. As expected for room temperature, no exciton absorption feature is visible for the epilayer structures. The exciton features for the quantum wells become stronger as the barrier energy become larger, and therefore as the quantum confinement increases. The quantum confinement lifts the degeneracy of the valence band, resulting in a heavy-hole exciton feature at lower energy and a light-hole exciton feature at higher energy. Fabry-Perot
Fig. 2.7. Absorption vs. photon energy for each of the five structures. The exciton features of the quantum wells become more pronounced as the binding energy increases, while there is no obvious exciton absorption feature for the epilayers.

Fringes are visible at photon energies below the band gap in each sample. These fringes are ignored here, but may be tuned to optimize various diffraction properties of the structures when desired [26].

2.4. Transverse-Field Electro-Optic Spectra

The transverse-field electro-optic effect in the quantum well structures with the applied electric field in the plane of the quantum wells consists of a change in the absorption and refractive index, due to the broadening of the excitonic transition, as shown in Fig. 2.8 for a large applied field. To lowest order in field, the change in absorption varies as the square of the field, because the broadening does not depend on the direction of the applied field. Therefore, for small electric fields the electroabsorption has a quadratic dependence on field. We define the quadratic coefficients $s_1$ and $s_2$ that characterize the low-field electro-optic effect as
Fig. 2.8. Absorption vs. wavelength for the 10\% Al barrier MQW without and with an applied field of 10 kV/cm. With an applied field, the excitons are ionized, so the absorption feature is broadened while its area remains constant.

\[
\Delta \tilde{n}(F) = -\frac{1}{2}n^3 \tilde{s} F^2
\]

where

\[
\tilde{s} = s_1 + is_2
\]

and

\[
\Delta \tilde{n} = \Delta n + i \frac{\hbar c}{4\pi E} \Delta \alpha
\]

(in SI units) where \(\tilde{n}\) is the complex refractive index. The changes in the refractive index and absorption are therefore

\[
\Delta n = -\frac{1}{2}n^3 s_1 F^2
\]

\[
\Delta \alpha = -\frac{2\pi E}{\hbar c} n^3 s_2 F^2
\]

where \(F\) is the applied field, \(E\) is the photon energy, and \(n\) is the refractive index, taken to be \(n = 3.5\) for all structures.
Fig. 2.9. Electro-optic properties of the 10% Al MQW: (a) electroabsorption magnitude vs. applied field for a photon energy near an electroabsorption peak, and (b) electroabsorption and electrorefraction vs. photon energy for an applied field of 2 kV/cm.

The electroabsorption was measured with the same apparatus as for transmission, with the lock-in amplifier referenced to the applied field. The electroabsorption spectrum was calculated from the differential transmission spectrum using the equation
\[ \Delta \alpha(F) = - \frac{1}{L} \ln \left( 1 + \frac{\Delta T(F)}{T(0)} \right) \] (2.14)

where the differential transmission was measured with a sinusoidal field varying between zero and a maximum field with a frequency of 277 Hz, and the transmission at zero-field was measured by chopping the source, also at 277 Hz. The electroabsorption is insensitive to frequency from DC to many kHz. The electrorefraction was calculated from the measured electroabsorption using the Kramers-Kronig transformation in Eq. 2.10. The field across the aperture is not uniform, but when averaged across the entire window, the change in transmission is acceptably accurate [27].

The dependence of electroabsorption on field is shown in Fig. 2.9(a) for the 10% Al quantum well sample. For fields below 3 kV/cm, the electroabsorption is nearly quadratic. The quadratic coefficient was determined by measuring the electroabsorption at a field of 2 kV/cm, where the quadratic dependence holds for every structure. Representative electroabsorption and electrorefraction spectra are shown in Fig. 2.9(b) for the 10% Al quantum well structure. The quadratic coefficients for the five structures studied are shown in Figs. 2.10-14. For every sample, the largest change in index occurs near the lowest photon energy for which the electroabsorption changes sign. The three samples, GaAs, 5% Al quantum wells and 10% Al quantum wells all have coefficients of comparable magnitudes. In the progression from GaAs to 5% Al quantum wells to 10% Al quantum wells to 30% Al quantum wells, both the excitonic binding energy and the excitonic oscillator strength increase, which should result in larger saturated electroabsorption. However, the sensitivity to field decreases for this progression. The competition between increasing oscillator strength and decreasing field sensitivity apparently results in the three lower oscillator strength structures having comparable electroabsorption at small field. It is interesting to note that there are oscillations in the electroabsorption for higher photon energy for the 5% Al and 10% Al quantum well structures. The oscillations might be due to weak coupling between the wells that occurs as the barrier is lowered.

Despite the lack of a clear absorption line for the epilayers, the electroabsorption in these structures is also strong. As with the quantum wells, the saturated electroabsorption for the Al0.2Ga0.8As epilayer should be larger than for the GaAs epilayer, but its sensitivity to field should be smaller. Apparently, the decreased sensitivity to field results in the low-field electroabsorption for the Al0.2Ga0.8As epilayer being much smaller than for the GaAs epilayer.
Fig. 2.10. Quadratic electro-optic coefficients vs. photon energy for the GaAs epilayer.

Fig. 2.11. Quadratic electro-optic coefficients vs. photon energy for the 5% Al MQW.
Fig. 2.12. Quadratic electro-optic coefficients vs. photon energy for the 10% Al MQW.

Fig. 2.13. Quadratic electro-optic coefficients vs. photon energy for the 30% Al MQW.
Fig. 2.14. Quadratic electro-optic coefficients vs. photon energy for the 20% Al epilayer.

The energy for the electroabsorption peak is the same as the exciton transition energy for exciton field ionization. Measured transition energies are compared with calculated values in Table 2.1. The data are taken from the low field electro-absorption spectra, and the calculated values are from Eq. 2.7. The calculated values should be 5 to 10 meV larger since Coulomb binding is neglected.

2.5. Conclusion

This chapter has presented the electro-optic effect of semi-insulating quantum wells and epilayers in the transverse geometry. In particular, the three lower Al fraction structures have quadratic electro-optic coefficients of approximately 2x10^{-11} cm^2/V^2 for both refractive index and absorption in the small field limit.

Table 2.1. Heavy hole exciton transition energies. The data is from the peaks in the low-field electroabsorption spectra, and the calculation neglects Coulomb binding.

<table>
<thead>
<tr>
<th>Structure</th>
<th>Experiment (eV)</th>
<th>Calculation (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>1.421</td>
<td>1.424</td>
</tr>
<tr>
<td>5% Al barrier</td>
<td>1.452</td>
<td>1.451</td>
</tr>
<tr>
<td>10% Al barrier</td>
<td>1.466</td>
<td>1.461</td>
</tr>
<tr>
<td>30% Al barrier</td>
<td>1.476</td>
<td>1.477</td>
</tr>
</tbody>
</table>
2.6. References


3. THE PHOTOREFRACTIVE EFFECT IN TRANSVERSE GEOMETRY ALUMINUM GALLIUM ARSENIDE QUANTUM WELLS AND THIN FILMS

3.1. Introduction

The photorefractive effect is a low-intensity optical nonlinearity. A spatial pattern in incident intensity produces a corresponding refractive index pattern. The photogeneration, transport and trapping of charge creates a space-charge electric field that modifies the refractive index (and absorption) through an electro-optic effect. The previous chapter presented the specific electro-optic properties for photorefractive quantum wells in the transverse geometry. This chapter continues with the photorefractive properties of GaAs and AlGaAs quantum wells and epilayers. The process of probing the refractive index grating is useful both for studying the solid state physics of these structures, and for optical processing, as shown in the following two chapters. The climax of this chapter is the confirmation of a model for two-wave mixing that is based on the hot-electron effect [1]. Nonreciprocal energy transfer in two-wave mixing occurs in the transverse-field geometry with the electric-field applied parallel to the grating vector, using a CW laser and an applied DC electric field. Nonreciprocal energy transfer has not been seen in the perpendicular geometry where the electric field is applied perpendicular to the grating vector [2-4], without moving gratings [5]. The organization of this chapter is as follows: a general discussion of the photorefractive effect is given in §2; four-wave mixing and two-wave mixing measurements are described in §3; and the analysis of the photorefractive phase shift is presented in §4, and a moving grating experiment is presented in §5.

3.2. Photorefractive Transport and Thin-Film Diffraction

The simplest geometry for the photorefractive effect is shown in Fig. 3.1, where two coherent laser beams interfere in the thin-film sample. Although real laser beams have a gaussian profile, plane waves are a useful approximation. The interference creates a sinusoidal intensity pattern \( I(x) = I_0(1 + m \cos Kx) \), where \( I_0 \) is the average intensity (the sum of the individual intensities), \( m \) is the modulation index (the depth of the interference fringes), and \( K = 2\pi / \Lambda \) is the grating wavevector, \( \Lambda = \lambda / 2\sin \theta \) is the fringe spacing, \( \lambda \)
Fig. 3.1. Geometry for interference of two laser beams in a photorefractive quantum well device.

(a)

I(x) \Rightarrow \rho(x) \Rightarrow E_{sc}(x) \Rightarrow \Delta n(x)

GENERATION TRANSPORT TRAPPING

ELECTRO-OPTIC EFFECT

(b)

Dark Light Dark

CB Deep Level Defects

Energy x

(c)

Fig. 3.2. Charge transport in the photorefractive effect: (a) geometry for applied field and grating writing beams; (b) steps in the formation of a grating; (c) energy band diagram of the semiconductor, showing the transport of photocarriers.
is the wavelength and $\theta$ is the half-angle between the beams. A schematic of the steps in photocarrier transport is shown in Fig. 3.2(c). Photorefractive quantum wells are illuminated with a photon energy near the bandgap of the semiconductor structure, so that the electrons and holes are generated at the same rate by cross-band absorption. (This is in contrast to the photorefractive effect in bulk materials where the photon energy is selected to excite carriers from the deep level defects [6].) After generation, the carriers diffuse and drift under the influence of an applied field. Finally, the carriers trap at deep level defects. The transport equations are [7, 8]

\[
\begin{align*}
\frac{\partial n}{\partial t} - \frac{1}{e} \mathbf{V} \cdot \mathbf{j}_e &= I\alpha + (\beta_e + s_e I)N_D^0 - \sigma_e n \nu e N_D^+ - \gamma_e n p \\
\frac{\partial p}{\partial t} + \frac{1}{e} \mathbf{V} \cdot \mathbf{j}_h &= I\alpha + (\beta_h + s_h I)N_D^+ - \sigma_h p \nu h N_D^0 - \gamma_e p n \\
\mathbf{j}_e &= e\mu_e n \mathbf{E} + k_B T \mu_e \nabla n \\
\mathbf{j}_h &= e\mu_h p \mathbf{E} - k_B T \mu_h \nabla p \\
\frac{1}{e} \mathbf{V} \cdot \left( \mathbf{j}_e + \mathbf{j}_h \right) &= \frac{\partial}{\partial t} \left( n + N_A^- - n^+ D \right) \\
\mathbf{V} \cdot \mathbf{E} &= -\left( \frac{1}{\varepsilon \varepsilon_0} \right) \left( n + N_A^- - n^+ D \right)
\end{align*}
\]

(3.1)

where the first two equations are the generation-recombination equations for electrons and holes, the next two equations define the current densities, the fifth equation describes charge conservation, and the last equation is Gauss’ Law. The subscript $e$ (h) denotes a quantity for electrons (holes), and the free carrier concentrations for electrons (holes) is $n$ ($p$). The current density is $\mathbf{J}$, the incident laser intensity is $I$, the absorption is $\alpha$, the carrier thermal emission rate is $\beta$, the defect optical cross section is $s$, the defect capture cross section is $\sigma$, the carrier thermal velocity is $\nu$, the carrier mobility is $\mu$, the lattice temperature is $T$, and the electron-hole direct recombination cross section is $\gamma_{eh}$. It is also assumed that out of the total number density of deep level defects $N_D$, some compensate for shallow level density $N_A$, so that

\[
N_D = N_D^0 + N_D^+
\]

(3.2)

where $N_D^0$ is the density of neutral deep level defects, and $N_D^+$ is the number of ionized deep level defects.
Several assumptions can be made to solve these equations. One dimensional equations are adequate, with the axis along the direction of the applied field. To fully describe transport properties into and out of quantum well barriers, a second dimension should be included, but this is not necessary to explain most of the phenomena. After reduction to one dimension, the next challenge is that the set of equations is inherently nonlinear because of the products of free carrier densities with defect densities in the first two equations and with the electric field in the third and fourth equations. This obviously makes the equations more difficult to solve, but also more interesting. The seven equations with seven unknowns can be reduced to three, for \( n, p \) and \( E \). In the limit where the modulation index \( m \) is small (that is, a large intensity ratio between the writing beams), the equations can be linearized. In the zero order approximation, the uniform field is just the applied field, and the equations for \( n \) and \( p \) can be solved. The first order equations lead to a \( 3 \times 3 \) matrix equation, which can be solved with several assumptions. For instance, several small terms can be removed, such as thermal generation of carriers and direct recombination of electrons and holes. Although the equations can be solved for small \( m \), the experiments are usually carried out with \( m \) near unity to maximum the detected signal.

The space charge grating generated by the intensity pattern creates both a refractive index and an absorption grating through the electro-optic effect. The electro-optic effect is nonlinear; in addition there is a nonlinearity from the transport, which can be especially strong when \( m \) is near unity. Therefore the refractive index and absorption gratings are possibly far from sinusoidal. In general, the space-charge field can be expressed as a Fourier series. The total field is the sum of the applied field and the space charge field

\[
E_{TOTAL}(x) = E_0 + E_1 \cos \left( Kx - \phi_1 \right) + E_2 \cos \left( 2Kx - \phi_2 \right) + \ldots
\]  

(3.3)

where \( E_0 \) is the applied field. For low field where the electro-optic effect is quadratic, the complex refractive index grating is proportional to square of the total electric field
\[ \Delta \tilde{n}(x) \propto E_{TOTAL}^2(x) = \left\{ E_0^2 + \frac{1}{2} E_1^2 + \frac{1}{2} E_2^2 \right\} \\
+ \left\{ -2E_0E_1\cos \left( Kx - \phi_1 \right) + E_1E_2\cos \left( Kx + \phi_1 - \phi_2 \right) \right\} \\
+ \left\{ -2E_0E_2\cos \left( 2Kx - \phi_2 \right) + \frac{1}{2} E_1^2\cos \left( 2Kx - 2\phi_1 \right) \right\} \tag{3.4} \\
+ \ldots \]

where only terms up to the second harmonic are included. Therefore, there can be higher harmonic gratings from which diffraction can occur [9]. In addition, there is an offset in the uniform refractive index and absorption due to the presence of the grating.

The diffraction grating created by the photorefractive effect in the transverse geometry can be probed in three ways, as shown in Fig. 3.3. For thin-film structures, the diffraction is in the Raman-Nath regime, and therefore does not need to satisfy the Bragg condition. The first two phenomena depicted in Fig. 3.3, degenerate four-wave mixing (D4WM) and two-wave mixing (2WM), happen simultaneously when the writing beams have a photon energy in the spectrum of the electro-optic effect. The third geometry, nondegenerate four-wave mixing (ND4WM) is accomplished by writing the grating with a photon energy above the spectrum of the electro-optic effect, and probing the grating with a photon energy in the spectrum of the electro-optic effect.

3.2.1. Raman-Nath Diffraction

Raman-Nath diffraction can be derived simply by considering the wave front modulation of a transmitted beam, as shown in Fig. 3.4(a) and following Ref. [10]. The refractive index of the thin film layer is

\[ \tilde{n}(x) = \tilde{n} + \Delta \tilde{n}\cos \left( Kx - \phi \right) \tag{3.5} \]

where \( \tilde{n} = n + i\alpha / 2k \) is the uniform complex index that depends on the average field in the material, and \( \Delta \tilde{n} = \Delta n + i\Delta \alpha / 2k \) is the modulation of the complex refractive index due to just the fundamental grating (higher harmonics can also be taken into account). The incident electric field is expressed as
Fig. 3.3. Three geometries for probing the refractive index grating.

Fig. 3.4. Diffraction of Raman-Nath orders: (a) diffraction of one beam alone; (b) diffraction of both beams, showing both the four-wave mixing and the interference that causes two-wave mixing.
\[ E^{IN}(\tilde{r},t) = \delta E^{IN} \exp \left( i(\tilde{k} \cdot \tilde{r} - \omega t) \right) \]  

(3.6)

where \( \delta \) is the unit polarization vector. The polarization of the two writing beams are taken to be parallel, so the polarization vector is dropped from the equation, and the time dependence can also be factored out. The transmitted pulse undergoes a spatially varying phase change

\[ \delta(x) = \bar{n}(x) k L / \cos(\theta') \]  

(3.7)

where \( L \) is the thickness of the sample, and \( \theta' \) is the angle inside the material. Therefore, the field exiting the material is

\[ E^{OUT}(\tilde{r}) = E^{IN}(\tilde{r}) \exp \left( i\delta(x) \right) \]  

\[ = E^{IN} \exp \left( i\tilde{k} \cdot \tilde{r} \right) \exp \left( i\delta \right) \exp \left( i\Delta \delta \cos (Kx - \phi) \right) \]  

(3.8)

and using the relation

\[ \exp \left( iz \cos (\delta) \right) = \sum_{M = -\infty}^{\infty} i^{M} J_{M}(z) \exp \left( iM\delta \right) \]  

(3.9)

the output field can be separated into the diffracted orders

\[ E^{OUT}(\tilde{r}) = E^{IN} \exp \left( i\tilde{k} \cdot \tilde{r} \right) \exp \left( i\delta \right) \sum_{M = -\infty}^{\infty} i^{M} J_{M}(\Delta \delta) \exp \left( iM(Kx - \phi) \right) \]  

\[ = E^{IN} \exp \left( i\delta \right) \sum_{M = -\infty}^{\infty} J_{M}(\Delta \delta) i^{M} \exp \left( -iM\phi \right) \exp \left( i(\tilde{k} \cdot \tilde{r} + MKx) \right) \]  

(3.10)

\[ = \sum_{M = -\infty}^{\infty} E_{M} \exp \left( i(\tilde{k} + MK) \cdot \tilde{r} \right) \]

where the direction of the diffracted order is determined by the last exponential. The Bessel function can be approximated for small arguments

\[ J_{M}(z) = \frac{z^{M}}{2^{M} M!} \text{ for } M > 0 \]

\[ J_{-M}(z) = (-1)^{M} J_{M}(z) \]  

(3.11)
because the refractive index modulation and absorption modulation are small in this geometry. Therefore, the diffracted field $E_M$ is

$$E_M = E_I^N \exp \left( i \delta \right) \exp \left( -iM \phi \right) \Delta \delta / 2^M M!$$ (3.12)

in the direction $\hat{k} + MK\hat{z}$. The direction can be rewritten as

$$\sin \theta_M = \sin \theta + MK / k$$ (3.13)

where $\theta_M$ is the angle of the diffracted beam from the normal. Usually, only the $M = \pm 1$ and 0 order beams are useful, because diffraction efficiency drops as a power of $M$.

Eq. 3.12 and Eq. 3.13 are used below to calculate the diffracted field for each of the three diffraction geometries.

3.2.2. Degenerate Four-Wave Mixing

In the degenerate four-wave mixing geometry shown in Fig. 3.4(b), the two beams that write the grating also diffract from the grating. For symmetric incident beams the two incident wave vectors are

$$\hat{k}_1 = k \cos(\theta) \hat{x} + k \sin(\theta) \hat{z}$$
$$\hat{k}_2 = -k \cos(\theta) \hat{x} + k \sin(\theta) \hat{z}$$ (3.14)

with the diffracted field from $E_I$ being

$$E_M = E_I^N \exp \left( i \delta \right) \exp \left( -i\phi \right) \Delta \delta / 2$$ (3.15)

which has the intensity

$$I_M = I_I^N \exp \left( \text{Re} \left( i \delta \right) \right) \Delta \delta^2 / 2$$

$$= I_I^N \exp \left( -\alpha L \right) \frac{1}{4} \left[ \left( \frac{\Delta n k L}{\cos \theta'} \right)^2 + \left( \frac{\Delta \alpha L}{2 \cos \theta'} \right)^2 \right].$$ (3.16)
The efficiency of the grating in diffracting the beams is quantified by either the input
diffraction efficiency

$$
\eta_{IN}^{M=1} = \frac{I_{M=1}}{I_{IN}} = \exp \left(-\alpha L\right) \frac{1}{4} \left[ \left( \frac{\Delta n}{\cos \theta'} \frac{k L}{\cos \theta} \right)^2 + \left( \frac{\Delta L}{\cos \theta'} \right)^2 \right]^{1/2}
$$

which includes the effect of transmission, or the output diffraction efficiency

$$
\eta_{OUT}^{M=1} = \frac{I_{M=1}}{I_{IN}} \exp \left(-\alpha L\right) = \frac{1}{4} \left[ \left( \frac{\Delta n}{\cos \theta'} \frac{k L}{\cos \theta} \right)^2 + \left( \frac{\Delta L}{\cos \theta'} \right)^2 \right]^{1/2}
$$

which does not include transmission, but is more convenient to calculate from experimental
measurements.

The direction of the diffracted beam can be simplified starting from Eq. 3.13, since
the probe beams are also the writing beams. The grating vector magnitude is $K = 2k\sin \theta$, so

$$
\sin \theta_M = (1 + 2M)\sin \theta
$$

in the case of degenerate four-wave mixing.

3.2.3. Two-Wave Mixing

Two-wave mixing is also shown schematically in Fig. 3.4(b), where each of the
zero order transmitted beams interferes with one of the first diffracted orders from the other
beam. This interference can cause net energy transfer between the two beams. Again, the
field in the direction of beam one is

$$
E_{1}^{OUT} = E_{M_1=0} + E_{M_2=1}
= E_{1}^{IN} \exp \left(i\delta\right) + E_{2}^{IN} \exp \left(i\delta\right) i \exp \left(-i\phi\right) \Delta \delta/2
$$

$$
= \exp \left(i\delta\right) \left(E_{1}^{IN} + E_{2}^{IN} i \exp \left(-i\phi\right) \Delta \delta/2\right)
$$
and the intensity is

\[ I_{1\text{OUT}}^{\text{OUT}} = \exp(-\alpha L) \left( I_{1\text{IN}}^{\text{IN}} + 2\sqrt{I_{1\text{IN}}^{\text{IN}} I_{2\text{IN}}^{\text{IN}}} \Re \left( i \exp(-i\phi) \Delta \delta/2 \right) + I_{2\text{IN}}^{\text{IN}} \Delta \delta/2 \right)^2 \]  

(3.21)

\[ = \exp(-\alpha L) \left( I_{1\text{IN}}^{\text{IN}} + \sqrt{I_{1\text{IN}}^{\text{IN}} I_{2\text{IN}}^{\text{IN}}} \left( \frac{\Delta n k L}{\cos \theta'} \sin \phi - \frac{\Delta \alpha L}{2 \cos \theta'} \cos \phi \right) \right) \]

where the second order term is negligibly small and neglected. Similarly, for the second beam,

\[ I_{2\text{OUT}}^{\text{OUT}} = \exp(-\alpha L) \left( I_{2\text{IN}}^{\text{IN}} + \sqrt{I_{1\text{IN}}^{\text{IN}} I_{2\text{IN}}^{\text{IN}}} \left( -\frac{\Delta n k L}{\cos \theta'} \sin \phi + \frac{\Delta \alpha L}{2 \cos \theta'} \cos \phi \right) \right) \]  

(3.22)

where the sign of the refractive index term has changed. The absorption \( \alpha \) depends on both the applied field and the presence of the grating, as discussed with Eq. 3.4. The absorption can be rewritten as \( \alpha = \alpha(E) + \alpha \), where \( \alpha(E) \) is the absorption for the uniform applied field and \( \alpha \) is the change in the average electroabsorption due to the space-charge field and the nonlinear electro-optic effect. The two-wave mixing ratio is the ratio of the mixed intensity to the unmixed intensity:

\[ \gamma = \frac{I_{2\text{OUT}}^{\text{OUT}}}{I_{1\text{IN}}^{\text{IN}} \exp(-\alpha(E)L)} = \exp(-\alpha L) \left( 1 + \sqrt{\beta} \left( \frac{\Delta n k L}{\cos \theta'} \sin \phi + \frac{\Delta \alpha L}{2 \cos \theta'} \cos \phi \right) \right) \]  

(3.23)

where the beam ratio is \( \beta = I_{2\text{IN}}^{\text{IN}} / I_{1\text{IN}}^{\text{IN}} \). A simple physical picture for nonreciprocal energy transfer is shown in Fig. 3.5. The photorefractive effect generally displaces the refractive index grating by a non-zero spatial shift described in terms of the photorefractive phase shift \( \phi \) from the incident intensity pattern. If the change of refractive index pattern is viewed to occur as step functions, then the step from the index \( n \) to the index \( \Delta n \) can be viewed as an interface. Following the propagation of beam one, the beam impinges upon the interface from high to low index (because \( \Delta n < 0 \)), so the reflected beam does not undergo any phase change, and interferes constructively with the transmitted part of beam
two. Following beam two, the beam impinges upon the interface from low to high index, and therefore the reflected beam undergoes a $\pi$ phase change, and interferes destructively with the transmitted part of beam one. Therefore, there is a net transfer of energy from beam one to beam two. This energy transfer depends on the sine of the phase shift, and is maximum for a shift of $\pm \pi/2$. The energy transfer is a relative transfer between the two transmitted beams, and not necessarily net gain in either of the beams. The absorption of the photorefractive quantum wells is generally larger than the energy transfer, so that neither beam can be amplified.

3.2.4. Nondegenerate Four-Wave Mixing

This is conceptually the most straightforward geometry, because the beams that write the grating are different from the beam that probes the grating. In this case, $\vec{k} = k\hat{z}$
for a normally incident probe. The diffraction efficiency is defined the same as for degenerate four-wave mixing, except that the direction can be written as

\[ \sin \theta_M = \sin \theta_{PR} + \frac{\lambda_{PR}}{\lambda_{WR}} \sin \theta_{WR} \]  

(3.24)

where the subscript \( PR \) indicates the probe beam, and the subscript \( WR \) indicates the writing beam. For nondegenerate four-wave mixing, the photon energy of the writing beams will always be larger than for the probe beam, so \( \lambda_{WR} < \lambda_{PR} \) always. Also, two-wave mixing does not occur. There are issues with respect to the relative intensity of the writing beams and probe beam, because the probe beam acts to erase the gratings [11].

3.3. Photorefractive Experiments

The samples used in photorefractive experiments are prepared as discussed in Chapter 2. Proton implantation provides the deep level defects to trap the space charge. The experiments are performed using a CW Ti-Sapphire laser as the source, which is pumped by an Argon Ion laser. Ti-Sapphire lasers have an amazing tuning range of over 300 nm, approximately from 700 nm to 1000 nm, which straddles the range of photon energies useful for studying AlGaAs materials. The experimental geometry for degenerate four-wave mixing and two-wave mixing is shown in Fig. 3.6, and for nondegenerate four-wave mixing in Fig. 3.7. The grating writing laser beam is split into two beams (by a cube beamsplitter) which interfere in the multiple quantum well sample without focusing. The diffracted beams are detected using biased silicon p-i-n photodiodes. The output of each photodetector is read with a lockin amplifier, which is either referenced to the chopper or to a sinusoidal field applied to the sample. Experiments are typically performed with a 10 \( \mu \)m fringe spacing. The diffraction efficiency drops as the fringe spacing is decreased, due to carrier diffusion [8]. The critical fringe spacing is approximately 3 \( \mu \)m. A fringe spacing longer than 25 \( \mu \)m is inconvenient, due to the close spacing between the transmitted and diffracted beams. The applied field is typically 4 to 10 kV/cm, and total incident intensities are 1 to 100 W/cm\(^2\) (on the order of 1 mW total optical power). There is a trade-off between incident intensity and the response time of the sample. Photorefractive quantum wells have one of the worlds lowest saturation intensities. The saturation intensity is defined as the intensity at which the photoconductivity equals the dark conductivity, and is
Fig. 3.6. Experimental geometry for degenerate four-wave and two-wave mixing.

Fig. 3.7. Experimental geometry for nondegenerate four-wave mixing. The diode laser 
writes the grating with a photon energy above the bandgap of the multiple quantum well 
structure. The diode laser beam is shown with a dashed line.
typically 10 μW/cm² [8]. The saturation intensity is low due to the high resistivity of the material. The response time will be discussed more in §5.

The upper limit on optical and electrical power applied to the sample is Joule heating of the sample by the motion of photocarriers in the applied field. Since the samples are bonded to a glass slide, the heat is not conducted efficiently away from the sample. To monitor the photocurrent through the sample, a load resistor \( R_L \approx 100 \, \text{kΩ} \) is in series with the sample. The voltage \( V_L \) over the resistor is used to calculate the current. A rule of thumb is that more than 10 mW of Joule heating is detrimental due to spectral shift of the band gap with temperature. Since the resistance of the sample is at least an order of magnitude larger than \( R_L \), the Joule heating is \( P = V V_L / R_L \), that is, just the power from the voltage supply.

Experimental results for the three diffraction geometries follow.

3.3.1. Degenerate Four-Wave Mixing

The output diffraction efficiency in Eq. 3.17 can be simplified to

\[
\eta_1^{\text{OUT}} = \left( \frac{\pi L \Delta n}{\lambda} \right)^2 + \left( \frac{L \Delta \alpha}{4} \right)^2
\]

(3.25)

for small incident angles. The diffraction efficiency vs. photon energy for the 10% Al barrier MQW sample is shown in Fig. 3.9 for a fringe spacing of 10 μm with a sinusoidal time varying applied field of 4 kV/cm. The fit is calculated from the electroabsorption and electrorefraction according to Eq. 3.25, with the magnitude being the only free parameter. Although the fit is good, there is some discrepancy. One source of error is due to Fabry-Perot effects, since the samples are not anti-reflection coated. Another more difficult error is the actual modulation of the space charge field in the sample. Electro-optic measurements are made with a 100% modulation depth, that is, the field is varied from zero to some maximum value. However, the modulation depth of the space charge in the sample is not known. As discussed above, the calculations for large intensity modulation have not been performed for photorefractive quantum wells. Also, the modulation depends on the precise value of the material parameters through the carrier transport.

The diffraction efficiency vs. applied field is shown in Fig. 3.10 for three structures at the lowest-energy zero-crossing of the electroabsorption, using a DC applied field, a fringe spacing of 11 μm, a beam ratio near unity, and a total incident intensity of
Fig. 3.8. Electro-optic effect for 10% Al barrier MQW for 4 kV/cm applied field.

Fig. 3.9. Diffraction efficiency vs. photon energy for the 10% Al MQW. The fit is calculated from the electroabsorption and electrorefraction with only the magnitude as a free parameter.
Fig. 3.10. Diffraction efficiency vs. applied field for the GaAs epilayer, the 10% Al MQW and the 20% Al epilayer, measured at the lowest energy zero-crossing in electroabsorption for each structure.

20 mW/cm². The strong saturation of the diffraction efficiency in the GaAs epilayer and the quantum well is not a result of electroabsorption saturation. As can be seen in Fig. 2.8(a), the electroabsorption is increasing linearly in this field range, and the four-wave mixing depends quadratically on the electro-optic effect from Eq. 3.25. Therefore, the saturation of the four-wave mixing diffraction efficiency must be due to a transport effect. The nonlinear velocity-field property of hot electrons is shown to be the cause of the saturation of the diffraction efficiency in §4.

3.3.2. Two-Wave Mixing

Photorefractive two-wave mixing in semiconductor quantum wells and thin films provides a versatile spectroscopy not only of the electro-optic properties of the materials, but also of electronic transport and space-charge accumulation phenomena. The photorefractive effect is sensitive to transport and space-charge effects through the so-called photorefractive phase shift. The photorefractive phase shift is defined as the ratio of the spatial shift of the optical gratings relative to the intensity interference pattern caused by the interference between two coherent laser beams. Photorefractive phase shifts are common in many photorefractive materials [6], but were not anticipated to occur in photorefractive
quantum wells. Photorefractive phase shifts were discovered in transverse-field photorefractive quantum wells [12], and have been speculated to arise from hot-electron effects and transport nonlinearity [13]. Therefore, two-wave mixing spectra provide both optical and transport information.

In two-wave mixing between a pump beam (I₁) and a probe beam (I₂), the first diffraction order of each beam propagates in the direction of the other beam [6]. In the case of photorefractive semiconductor quantum wells and thin films the combined absorption and refractive index gratings participate in the diffraction in the Raman-Nath regime. The intensities of the two emerging beams can be expressed as given in Eq. 3.21 and Eq. 3.22. The contribution to the transmitted intensity from the absorption grating is symmetric with respect to the photorefractive phase-shift φ, while the contribution from the index grating is asymmetric. By reversing the direction of the applied electric field it is possible to determine both contributions.

In the case of small α, the exponential in the two-wave mixing ratio may be expanded, and the two-wave mixing ratio becomes

\[
γ = 1 - αL + \sqrt{β} \left( \frac{Δn k L}{\cos θ} \sin φ + \frac{Δα L}{2 \cos θ} \cos φ \right)
\]

(3.26)

The nonreciprocal energy transfer arises from the refractive index contribution to the two-wave mixing ratio, which is obtained by taking the difference in the two-wave mixing between the positive and negative field directions (the antisymmetric part of the two-wave mixing ratio):

\[
γ_n = \frac{1}{2} \left( γ(E) - γ(−E) \right) = \sqrt{β} \frac{Δn k L}{\cos θ} \sin φ
\]

(3.27)

The antisymmetric two-wave mixing ratio γₙ is related to the photorefractive gain

\[
Γ = \frac{2 Δn k}{m \cos θ} \sin φ \quad \text{(where} \ m = \frac{2\sqrt{β}}{1 + β}\text{)}
\]

by the equation

\[
γ_n = \frac{β}{1 + β} ΓL
\]

(3.28)

The symmetric modulation arises from absorption and is given by the symmetric part of the two-wave mixing ratio:
\[ \gamma_\alpha = \frac{1}{2} (\gamma(E) + \gamma(-E)) = 1 - \alpha L + \sqrt{\beta} \frac{\Delta \alpha L}{2 \cos \theta} \cos \phi \] (3.29)

The two-wave mixing experiments were performed as discussed above. For two-wave mixing, the structures are biased by a DC voltage whose polarity can be reversed, and the measurements are performed by monitoring the intensity modulation of the transmitted probe beam I₁ as the pump beam I₂ is chopped at a frequency of 277 Hz. Only the diffracted terms of the above equations are therefore detected and the experimentally measured parameter \( \gamma_\alpha^{\text{exp}} \) is related to \( \gamma_\alpha \) through the relation \( \gamma_\alpha^{\text{exp}} = \gamma_\alpha - 1 \). Chopping the pump beam may introduce a systematic error due to the redistribution of the electric field within the sample. To eliminate this contribution we also measured the modulation of I₁ when the writing beams were cross-polarized and subtracted the cross-polarized signal from the co-polarized signal.

The symmetric and antisymmetric contributions to Eq. 3.26 are shown in Figs. 3.11-15 for the four quantum well structures and epilayers, with a fringe spacing of 12 \( \mu \text{m} \), an applied field of 7.5 kV/cm, an incident intensity of 10 mW/cm\(^2\) and a beam ratio near unity. The experimentally measured electroabsorption spectra were scaled to fit the symmetric data, and the electrorefraction spectra were scaled to fit the antisymmetric data. The experimental electroabsorption spectra were measured for an applied field of 7.5 kV/cm, and the electrorefraction was calculated using the Kramers-Kronig relation. The scaling parameter could not be predicted, because the effective modulation index is unknown and the phase shift was not measured here. The agreement between the data and the electro-optic spectra is good for all the samples except the GaAs. For all samples, there is also some discrepancy for higher photon energies. The parameters used to scale the electro-optic data to fit the two-wave mixing ratios are given in Table 3.1, with estimated values of the photorefractive phase shift calculated using the parameters and Eq. 3.27.

The two-wave mixing vs. applied field is shown in Fig. 3.16 for three samples at the lowest-energy zero-crossing in electroabsorption, for a fringe spacing of 11 \( \mu \text{m} \), a beam ratio near unity, and a total incident intensity of 20 mW/cm\(^2\). As in the case of four-wave mixing vs. field, the saturation of the GaAs and 10% Al quantum-well two-wave mixing (which depends linearly on the electro-optic effect from Eq. 3.26) at a field where the electro-optic effect is increasing linearly with field indicates that there is a transport effect causing the saturation.
Fig. 3.11. Symmetric and antisymmetric two-wave mixing ratios for the GaAs epilayer.

Fig. 3.12. Symmetric and antisymmetric two-wave mixing ratios for the 5% Al MQW.
Fig. 3.13. Symmetric and antisymmetric two-wave mixing ratios for the 10% Al MQW.

Fig. 3.14. Symmetric and antisymmetric two-wave mixing ratios for the 30% Al MQW.
Fig. 3.15. Symmetric and antisymmetric two-wave mixing ratios for the 20% epilayer.

Fig. 3.16. Antisymmetric two-wave mixing ratio vs. applied field for the GaAs epilayer, the 10% Al MQW and the 20% Al epilayer, measured at the lowest energy zero-crossing in electroabsorption for each structure.
Table 3.1. Parameters for fitting electro-optic data to two-wave mixing data and the estimated phase shift.

<table>
<thead>
<tr>
<th>Structure</th>
<th>Anti-symmetric fitting parameter</th>
<th>Symmetric fitting parameter</th>
<th>Estimated phase shift</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>1.75</td>
<td>$7.5 \times 10^{-6}$</td>
<td>$58^\circ$</td>
</tr>
<tr>
<td>5% Al MQW</td>
<td>11</td>
<td>$2.75 \times 10^{-5}$</td>
<td>$70^\circ$</td>
</tr>
<tr>
<td>10% Al MQW</td>
<td>8</td>
<td>$1.5 \times 10^{-5}$</td>
<td>$75^\circ$</td>
</tr>
<tr>
<td>30% Al MQW</td>
<td>3.5</td>
<td>$4.0 \times 10^{-5}$</td>
<td>$31^\circ$</td>
</tr>
</tbody>
</table>

Fig. 3.17. Nondegenerate four-wave mixing diffraction efficiency vs. probe wavelength compared with degenerate four-wave mixing for the double multiple quantum well structure. The diffraction peak for the lower energy set of quantum wells is suppressed in nondegenerate four-wave mixing compared to the same peak in the degenerate case.

3.3.3. Nondegenerate Four-Wave Mixing

In principle, the spectra for nondegenerate four-wave mixing diffraction efficiency should be the same as for degenerate four-wave mixing. The nondegenerate and degenerate output diffraction efficiency for a "double" quantum well structure is shown in Fig. 3.17, where there is a marked difference in the two spectra. This structure consists of 75 periods of quantum wells with 15% Al barriers, and another 75 periods of quantum wells with 30% Al barriers, which have an exciton energy level at the higher energy (lower
wavelength). The higher energy diffraction peak is smaller for nondegenerate four-wave mixing, relative to the lower energy peak. The photon energy of the writing beam for nondegenerate mixing is 1.810 eV, while the band gap of the 15% Al is 1.611 eV, and the gap of the 30% Al barrier is 1.798 eV. The writing beam photon energy is near the gap of the higher barrier, and far above the gap of the lower barrier. Therefore, it appears that the higher the writing beam photon energy above the exciton resonance, the higher the diffraction efficiency. Although there may be some influence from Fabry-Perot effects, the large magnitude of the effect indicates that it is more likely due to some aspect of the carrier transport. In particular, the transport perpendicular to the wells should be considered.

3.4. Photorefractive Phase Shift

Nonreciprocal energy transfer was observed in transverse-field photorefractive quantum wells [12]. The presence of nonreciprocal energy transfer in the two-wave mixing of all five structures implies there is a spatial shift of the index grating relative to the light intensity pattern. The characteristic field for onset of the phase shift is near 3 kV/cm, which is also where the electron velocity becomes nonlinear for GaAs [14, 15]. This field is substantially larger than the diffusion field [16-18] (~0.2 kV/cm) and substantially smaller than the field for space-charge limitation [6, 19] (~200 kV/cm). To account for the possibility that the electron velocity-field nonlinearity could affect transport and cause nonreciprocal energy transfer in two-wave mixing, the usual linear transport equations were modified to include the nonlinear electron velocity [20]. For an applied field \( E_0 \) and a grating vector \( K \), the space-charge electric field may be written as

\[
E_{sc}(K, x) = \frac{-m E_0 \exp(iKx)}{1 - \left(1 + i KL_{Eh}^{-1}\right) \frac{L_{nonlinear}}{L_{bipolar}}} \tag{3.30}
\]

when diffusion fields are neglected. This assumption is valid in the high-field limit because diffusion fields of ~0.2 kV/cm are much smaller than applied fields over 1 kV/cm. We define a nonlinear transport length by

\[
L_{nonlinear} = \left(\frac{v(E_0)}{E_0} - \frac{d v(E_0)}{d E_0}\right) E_0 \tau_e \tag{3.31}
\]
where the linear bipolar drift length is \( L_{E_{bipolar}} = L_{E_h} + L_{E_e} \). The quantities \( L_{E_h} \) and \( L_{E_e} \) are the linear transport lengths for holes and electrons, and are equal to \( \mu_h E \tau_h \) and \( v(E) \tau_e \), respectively. The carrier recombination lifetimes are \( \tau_h \) and \( \tau_e \), the hole mobility is \( \mu_h \), and the field-dependent electron velocity is \( v(E) \).

Eq. 3.30 describes a screened static sinusoidal field with a nonlocal (complex) screening dielectric function. When the nonlinear transport is strong, the imaginary term in the denominator is large, and the space-charge field is spatially shifted from the intensity pattern, representing a nonlocal response of the material to the external static perturbation. Physically, velocity saturation in trap-dominated semiconductors disables dielectric relaxation of space-charge, depleting charge from the bright fringes and preventing the space-charge trapped in the dark fringe from relaxing. In extreme cases the internal field amplitude can be nearly entirely imaginary, predicting a maximum spatial shift of the field by a quarter fringe spacing relative to the intensity pattern.

The electron velocity-field nonlinearity is caused by intervalley scattering in AlGaAs: as an electric field is applied to the material, carrier heating promotes electrons from the \( \Gamma \) valley to the \( L \) valleys [15]. The smaller mobility in the \( L \) valley results in a saturation of the electron velocity with field. The larger the energy difference \( \Delta E_{\Gamma L} \) between the \( \Gamma \) valley and the \( L \) valley, the stronger the nonlinearity, and the larger the maximum of the nonlinear transport length defined in Eq. 3.31.

To test the nonlinear transport hypothesis as the source of nonlocal response, we measured two-wave and four-wave mixing in three of the five structures. These structures each have different transport nonlinearities originating from different \( \Gamma - L \) valley energy separations, which are expected to produce three distinct signatures for the phase shift. One of the structures chosen was the 10\% Al quantum well sample, similar to structures that had exhibited the original nonlocal response [12]. In addition to the quantum well sample, the GaAs and Al\(_{0.2}\)Ga\(_{0.8}\)As epilayers were studied. The GaAs epilayer has a \( \Gamma - L \) valley energy separation of 284 meV, the 10\% Al quantum-well structure has a \( \Gamma - L \) valley energy separation of 224 meV in the barrier, and the Al\(_{0.2}\)Ga\(_{0.8}\)As epilayer has the smallest \( \Gamma - L \) valley energy separation (of our three structures) of 163 meV [21]. These quantum well and epilayer structures were chosen to provide a trend of decreasing \( \Gamma - L \) valley separation and decreasing transport nonlinearity from GaAs through the multiple quantum well to Al\(_{0.2}\)Ga\(_{0.8}\)As. The multiple quantum well sample exhibits an additional nonlinearity determined by quantum-well transport (alloy scattering at the well-barrier interface and quantum tunneling into the barrier) and real-space transfer effects [22]. The
quantum-well properties were candidates for providing a mechanism for the phase shift seen originally in the quantum well sample, but these mechanisms could be tested and discounted by comparing the quantum-well sample against the epilayers.

The carrier lifetimes in the three structures were engineered to eliminate a possible competing mechanism that could affect the nonlocal response. For instance, if the Al$_{0.2}$Ga$_{0.8}$As epilayer had significantly smaller carrier lifetimes than the GaAs epilayer, then the difference in photorefractive phase shift between these samples could be entirely due to trap limitation [6], independent of transport nonlinearity. Therefore, we engineered the carrier lifetimes in all three samples by proton implantation, and verified the lifetimes using femtosecond saturated absorption pump-probe measurements [23]. The Al$_{0.2}$Ga$_{0.8}$As epilayer had a 160 ps electron lifetime, the 10% Al multiple quantum well structure had a 143 ps electron lifetime, and the GaAs epilayer had a 48 ps electron lifetime. This monotonic trend represents increasing defect concentrations, which would produce decreasing photorefractive phase shifts under the trap limitation mechanism, opposite to the trend expected for the nonlinear transport mechanism. Therefore the sample lifetimes were specifically controlled to give a clear and unambiguous experimental differentiation between the trap-limitation mechanism and the nonlinear-transport mechanism.

The phase shift was determined by measuring two-wave mixing and four-wave mixing performed simultaneously at a wavelength where the electroabsorption is zero. The phase shift was then calculated from

$$
\sin (\phi) = \frac{\Delta n}{|\Delta n|} \frac{\gamma}{2\sqrt{\beta} \eta}
$$

(3.32)

From the four-wave mixing data and two-wave mixing data shown in Figs. 3.10 and 3.16, the sine of the photorefractive phase shift vs. applied field was calculated, with the results shown in Fig. 3.18. The GaAs epilayer has a significant photorefractive phase shift that increases for fields up to 4 kV/cm, and saturates at a value of $\phi = 50^\circ$. The Al$_{0.2}$Ga$_{0.8}$As epilayer, which is expected to have the smallest nonlinear transport based on intervalley scattering, has a phase shift of only $\phi = 21^\circ$ at 7.5 kV/cm, and is not yet saturated for this field. The multiple quantum well structure saturates at an intermediate phase shift of $\phi = 30^\circ$ for fields above 5 kV/cm. The phase shifts and the saturation fields in each of the structures follow a monotonic trend from the GaAs epilayer, through the multiple quantum
Fig. 3.18. Sine of the photorefractive phase shift vs. applied field for the GaAs epilayer, the 10% Al MQW and the 20% Al epilayer.

Fig. 3.19. Ratio of nonlinear to linear transport lengths as a function of Gamma-L energy separation. The curve is an exponential fit to the data indicating the exponential increase of the nonlinear transport as a function of intervalley separation. The inset shows the conduction band energy vs. wavevector in the L direction.
well to the \( \text{Al}_{0.2}\text{Ga}_{0.8}\text{As} \) epilayer, reflecting the monotonic trend in the \( \Gamma-L \) energy separation. The saturation field increases and the photorefractive phase shift decreases as the role of intervalley scattering is diminished in each of these structures. GaAs has the largest nonlinear transport and produces the largest phase shifts at the lowest saturation fields, while \( \text{Al}_{0.2}\text{Ga}_{0.8}\text{As} \) has smaller transport nonlinearities because of the reduced \( \Gamma-L \) valley energy difference.

The dependence of the nonlocal response is shown explicitly in Fig. 3.19 as a function of the \( \Gamma-L \) valley energy difference for a field \( E_0 = 7.5 \text{ kV/cm} \). The ratio of the nonlinear transport length to the linear transport length \( \frac{L_{\text{nonlinear}}}{L_{\text{bipolar}}} \) is calculated from the data in Fig. 3.18 using

\[
\frac{L_{\text{nonlinear}}}{L_{\text{bipolar}}} = \frac{\tan \phi}{K \mu_h E_0 \tau_e + \tan \phi}
\]  

(3.33)

where \( K = 2\pi/\Lambda, \Lambda = 11 \text{ \( \mu \)m, and } \mu_h = 600 \text{ cm}^2/\text{Vs} \). Eq. 3.33 is Eq. 3.31 solved for \( \frac{L_{\text{nonlinear}}}{L_{\text{bipolar}}} \), except that the measured \( \tau_e \) was used in place of \( \tau_h \), because the hole lifetime is not known. However, the hole lifetime is assumed to be proportional to the electron lifetime. The calculated values are shown in Table 3.2. The curve in the figure is an exponential fit to the data, indicating the Boltzmann factor for the ratio of electron populations in the indirect and direct valleys. The trend of the measured nonlinearity increasing with intervalley separation firmly establishes the direct role of transport nonlinearity in generating the nonlocal photorefractive phase shift in thin trap-dominated semiconductor films.

The photocurrent as a function of applied field was measured for the GaAs epilayer, as shown in Fig. 3.20. The bend in the curve occurs at the same field where the phase shift rises.

<table>
<thead>
<tr>
<th>Structure</th>
<th>( \sin \phi )</th>
<th>( \tau_e ) (ps)</th>
<th>( \frac{L_{\text{nonlinear}}}{L_{\text{bipolar}}} )</th>
<th>( \Delta E_{\Gamma L} ) (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>0.76</td>
<td>47.6</td>
<td>0.49</td>
<td>284</td>
</tr>
<tr>
<td>10% Al MQW</td>
<td>0.49</td>
<td>143.7</td>
<td>0.13</td>
<td>224</td>
</tr>
<tr>
<td>( \text{Al}<em>{0.2}\text{Ga}</em>{0.8}\text{As} )</td>
<td>0.36</td>
<td>159.6</td>
<td>0.085</td>
<td>163</td>
</tr>
</tbody>
</table>

Table 3.2. Photorefractive phase shifts, carrier lifetimes, calculated transport length ratios and intervalley energy separations for the three structures in the phase shift experiment.
Fig. 3.20. Sine of the photorefractive phase shift compared with the current-field curve as a function of applied field. The strong onset of the photorefractive phase shift coincides with the onset of intervalley scattering, seen as a marked deviation from ohmic behavior for fields above 3 kV/cm.
3.5. Moving Grating Experiments

One method to determine the response time of the photorefractive effect is to introduce moving the gratings in the material and measure the diffraction efficiency as a function of the fringe velocity. For gratings moving with a constant velocity, the incident intensity pattern is

\[ I(x,t) = I_0 \left( 1 + m \cos \left( k x - \Omega t \right) \right) \tag{3.34} \]

The photorefractive material must now screen a moving grating, which reveals the rate at which the screening occurs. This time is approximately the dielectric relaxation time, which depends on the incident intensity. Therefore, the trade-off between intensity and response time can be quantified. This experiment was performed using acousto-optic modulators. A modulator was placed in each of the writing beams in the (now near) degenerate geometry. The first diffracted order from each modulator was interfered in the sample. The frequency for one modulator was fixed at 80 MHz, while the other could be varied to 80 MHz ± 30 MHz. The first diffracted order from an acousto-optic modulator is shifted in frequency by the frequency of the modulator. Therefore, the frequency in Eq. 3.33 is the frequency difference between the two modulators.

Two-wave mixing and degenerate four-wave mixing were performed as a function of the moving grating frequency difference, as shown in Fig. 3.21. The sample used is the double quantum well from §3.3.3, and the wavelength was tuned to the lowest energy zero in the electro-absorption, so that the gratings are entirely refractive index gratings. The intensity incident on the sample is approximately 10 mW/cm². The output diffraction efficiency drops with increasing frequency difference as expected. The critical frequency is approximately 10 kHz for this incident intensity. The critical frequency should depend on the applied field partly because of the nonlinear electron velocity.

The two-wave mixing as a function of both positive and negative frequency difference is also shown in Fig. 3.21, along with the phase shift calculated from Eq. 3.32. The positive direction is chosen in the direction of the applied field, which is also the direction of positive phase shift. The graphs show that phase shift can be "pushed" and "pulled" with the moving gratings, as generally expected for the phase of a driven system.
Fig. 3.21. Two-wave mixing and degenerate four-wave mixing with moving gratings.
Similarly, this technique has been used to induce a phase shift in a perpendicular geometry structure, where two-wave mixing does not occur with static gratings.

3.6. Conclusion

Degenerate four-wave mixing spectra can be understood in a straightforward manner, but nonreciprocal energy transfer in two-wave mixing is not predicted by the usual transport theory. The phase shift associated with the nonlocal optical response observed in two-wave mixing was measured in three structures. In conjunction with measurements of the photocurrent vs. applied voltage, the phase shift measurements confirm that the hot electron velocity-field nonlinearity is the transport effect responsible for nonreciprocal energy transfer in two-wave mixing. Nondegenerate four-wave mixing spectra indicate that vertical transport may play a role in the transverse geometry. Finally, moving gratings reveal the response time of grating formation.
3.6. References


4. STEADY-STATE FOUR-WAVE MIXING IN PHOTOREFRACTIVE QUANTUM WELLS USING FEMTOSECOND PULSES

4.1. Introduction

Calibrated ultrafast optical spectroscopy relies on autocorrelation techniques for the characterization of ultrafast pulses. Subtle changes in the intensity profile or in the phase of a pulse after transit through a dispersive medium can yield important information concerning the physics of the medium. Autocorrelation techniques used to observe these effects fall broadly into two classes: intensity and field autocorrelation [1-5]. Intensity autocorrelation provides information on the instantaneous intensity of the pulse. Nonlinear optical effects, such as second harmonic generation, are typically used for this class of techniques [1, 6]. Field autocorrelation provides information on the electric field, including the phase, of the pulse. For many applications, field autocorrelation provides more useful information. Field autocorrelation can be achieved through interferometric second harmonic generation [4, 7], or through induced-grating techniques using multiwave mixing in intensity-integrating materials. Such induced-grating techniques have provided many varieties of autocorrelators [8-10].

Multi-wave mixing of ultrafast pulses is attractive as a field-autocorrelation technique because of its relative ease of implementation. This technique was demonstrated early in thin films [5, 11], and has continued in broader applications, for instance measuring coherence times of broadband CW lasers [12]. Johnson et al. [13] first demonstrated the use of photorefractive materials to measure the coherence length [14] of picosecond pulses. Photorefractive materials have the advantage of high sensitivity, requiring low light intensities to generate large induced refractive index gratings. The low light intensities are a result of the relatively long response time of the materials, allowing low photon fluxes to be integrated over time. The photorefractive technique is part of a much broader group of techniques that includes interferometric second harmonic generation [15]. One of the benefits of photorefractive ultrafast mixing is the possibility for temporal encoding, i.e., encoding temporal information by spatially distributing the information in
the photorefractive crystals. The temporal encoding allows real time autocorrelation [16], or femtosecond reconstruction [17].

In this paper we describe femtosecond mixing in photorefractive AlGaAs/GaAs multiple quantum wells. The quantum well samples are thin films with quantum-confined excitons that are sensitive to applied electric fields. The importance of thin films stems from the thin interference gratings that operate in the Raman-Nath diffraction regime and do not require phase matching to observe a diffracted beam. Our samples operate in the transverse field geometry, also known as the Franz-Keldysh geometry. The operation of these photorefractive quantum wells has been described in detail elsewhere [18]. The important feature of the transverse field geometry is the presence of an aperture defined by gold electrodes that are used for applying the electric field. The aperture of the device and the diffraction properties of thin gratings define several possible field-autocorrelation modes. These include two-wave mixing, degenerate four-wave mixing and nondegenerate four-wave mixing.

The theory of ultrashort pulse interference is presented in section 4.2, and the theory of four-wave mixing diffraction from photorefractive quantum wells is described in section 4.3. Experimental results are presented in section 4.4 and discussed in section 4.5.

4.2. Interaction Area and Photorefractive Gratings

In the geometry of induced-grating autocorrelation, two laser beams intersect inside a sensitive nonlinear material. The intersecting optical pulses generate interference fringes if the pulses are coherent. The intersection volume depends on the crossing angle and on the pulse duration \( t_p \) (full width at half maximum). A representative intersection volume is shown in Fig. 4.1 for symmetric beams incident on a thin film each by an angle \( \theta \). The beams require no focusing. The delay of beam 2 relative to beam 1 is expressed as \( \tau \). The intersection volume intersects the plane of the photorefractive quantum well in a well-defined area within the aperture defined by the gold electrodes. The position of the interaction area is fixed for a constant delay \( \tau \). The displacement of the center of the interaction area relative to the center of the aperture is given by

\[
x_0(\tau) = \frac{c \tau}{2 \sin \theta}
\]

(4.1)

Within the coherence volumes of the pulses, the average light intensity is modulated by the interference between the two beams. The average light intensity is given by
Fig. 4.1 Intersection volume and interaction area for two colliding pulses intersected by a thin film. The center position of the interaction area within the window defined by the electrodes depends on the relative time delay and on the crossing angle. Degenerate four-wave mixing is shown in (a) and nondegenerate four-wave mixing in (b). Only the intersection volume of the two pulsed beams is shown in (b).
\[ I(x) = I \left[ 1 + m(x, \tau) \cos(Kx + \phi) \right] \]  \hfill (4.2)

where \( x \) is the distance across the aperture, \( \phi \) is an arbitrary phase, \( \bar{I} \) is the average laser intensity and \( K = 2\pi/\Lambda \) is the grating wavevector of the fringe pattern with a fringe spacing \( \Lambda \). All quantities are time averages. The most important component of the equation is the modulation index function \( m(x, \tau) \). The modulation function provides all the information concerning the coherence properties. As shown in Fig. 4.1, the modulation index is maximum at \( x_0(\tau) \), with width \( \Delta x \) (full width at half maximum).

In photorefractive materials, only time-average light intensities are relevant for the formation of induced gratings. The photon flux is integrated and stored as trapped space charge in defects in the material. If the relaxation time of the trapped charge is much longer than the repetition time of the laser pulses, then the material only responds to the total flux integrated over the relaxation time. The slow response and long time integration of the induced gratings makes it possible to operate with low average light intensities. In photorefractive semiconductors, the response time is typically microseconds, compared with typical repetition times of nanoseconds. The induced gratings in these pulsed experiments are therefore under steady-state conditions.

The modulated light intensity generates both refractive index gratings and absorption gratings in the photorefractive quantum well samples. In the transverse electric field geometry that we use [18], an electric field is applied parallel to the plane of the quantum wells and parallel to the grating vector \( K \). The modulated light intensity generates screening carriers that are trapped as space-charge at deep defects. The space charge modulates the applied electric field. Because the photorefractive quantum wells rely on the quadratic electroabsorption of the excitons, the refractive index and absorption grating amplitudes depend on the square of the local electric field through the equations

\[ \Delta n = -\frac{1}{2} n^3 s_1 E^2 \quad \Delta \alpha = -\frac{2\pi}{\lambda} n^3 s_2 E^2 \]  \hfill (4.3)

where \( s_1 \) and \( s_2 \) are quadratic electro-optic coefficients. The local electric field is given by \( E = E_0 + E_{sc} \), where \( E_0 \) is the externally applied electric field and \( E_{sc} \) is the space charge field. The grating modulation is therefore given by [18, 19]
\[ \Delta n = -n^3 s_1 E_0^2 m(x, \tau) \quad \Delta \alpha = -\frac{4\pi}{\lambda} n^3 s_2 E_0^2 m(x, \tau) \]  

(4.4)

where we have assumed full screening of the applied field.

The laser electric fields of the intersecting beams are

\[ \vec{E}_{1,2}(x, t) = \hat{e}_{1,2} E_{1,2}(t - \tau_{1,2} - \vec{k}_{1,2} \cdot \vec{x}/c) \exp \left[ i \left( \omega(t - \tau_{1,2}) - k_{1,2} \vec{k}_{1,2} \cdot \vec{x} \right) \right] \]  

(4.5)

where \( \hat{e}_{1,2} \) is the polarization vector for each beam, \( E_{1,2}(t) \) is the envelope function of each pulse, \( \tau_{1,2} \) is the pulse delay for each beam, and \( \vec{k}_{1,2} \) is the unit propagation vector for each beam, with \( k_1 = k_2 = k = \omega/c \) for the center wavelength of the pulse. The transverse beam intensity profile is neglected, which is valid for unfocused beams. The \( x \) direction is across the aperture and the \( z \) direction is normal to the sample, giving \( \vec{k}_{1,2} = \pm \sin(\theta) \hat{x} + \cos(\theta) \hat{z} \), where \( \theta \) is the angle from normal. The beams are both polarized perpendicular to the plane formed by the beams, with \( \hat{e}_{1,2} = \hat{y} \).

For the experimental intensities and electric fields, the response time of the system is 10 \( \mu \)s, much longer than the 13 ns between pulses. Therefore, the grating builds up over many pulses and is affected negligibly by one pulse. The intensity in the sample is averaged over the pulse repetition time \( T \) to find the modulation index. Taking \( \tau_1 = 0 \) and \( \tau_2 = \tau \), with \( z = 0 \) at the plane of the sample,

\[
I(x, \tau) = \frac{1}{T^2} \int_{-T/2}^{T/2} \left| \vec{E}_1(x, t) + \vec{E}_2(x, t) \right|^2 dt 
= \bar{I} \left[ 1 + m(x, \tau) \cos \left( Kx - \omega \tau + \phi(x, \tau) \right) \right]
\]  

(4.6)

where \( Z \) is the wave impedance, \( \bar{I} \) is the average intensity at the sample, and \( \phi \) is a phase that depends only weakly on \( x \) and \( \tau \). For \( E(t) = E_1(t) = E_2(t) \), the expression for the modulation index is
\[ m(x, \tau) \propto \left| \int_{-\pi/2}^{\pi/2} dt \, E(t - x \sin(\theta)/c) \, E^*(t - \tau + x \sin(\theta)/c) \right| \]

\[ \propto \left| \int_{-\infty}^{\infty} dt \, E(t + \tau - 2x \sin(\theta)/c) \, E^*(t) \right| \]

(4.7)

\[ \propto \Gamma(\tau - 2x \sin(\theta)/c) \]

where \( \Gamma(\tau) \) is the autocorrelation of the electric field envelope function, also called the coherence function. This can be calculated directly using the Wiener-Khinchin theorem by taking the Fourier transform of the power spectrum \( P(\omega) \).

For the case of a Gaussian pulse, the modulation index can easily be calculated. The intensity of the pulse is taken to be

\[ I_{1,2}(\bar{x}, t) = \bar{I}_{1,2} \frac{T}{t_p} \sqrt{\frac{4\ln 2}{\pi}} \exp \left[ -4\ln 2 \left( \frac{t - \tau_{1,2} - k_{1,2} \bar{x}/c}{t_p} \right)^2 \right] \]

(4.8)

where \( \bar{I}_{1,2} \) are the time-average intensities of each beam, \( T \) is the time between pulses, and \( t_p \) is the pulse duration. Assuming a transform-limited pulse, the electric field for each beam is

\[ E_1(\bar{x}, t) = \sqrt{2ZI_1(\bar{x}, t)} \exp \left[ i(\omega t - k_1 \bar{x}) \right] \]

\[ E_2(\bar{x}, t) = \sqrt{2ZI_2(\bar{x}, t)} \exp \left[ i(\omega(t - \tau) - k_2 \bar{x}) \right] \]

(4.9)

where \( Z \) is the wave impedance. For equal beam intensities, the modulation index is therefore

\[ m(x, \tau) = \exp \left[ -\frac{8 \ln(2)}{c^2 t_p^2} \sin^2 \theta \left( x - x_0(\tau) \right)^2 \right] \]

(4.10)

where the delay time \( \tau \) is implicitly included in the position \( x_0 \) defined in Eq. (4.1). The modulation function is a Gaussian with a full width at half maximum of
\[ \Delta x = \frac{c t_p}{\sqrt{2} \sin \theta} \]  

(4.11)

Small interaction area widths result from large crossing angles. The angular control of the interaction area width provides control of the autocorrelator time resolution.

4.3. Autocorrelator Geometries: Diffraction Efficiency

We define two autocorrelator geometries based on the photorefractive quantum wells. The first geometry operates from self diffraction in degenerate four-wave mixing. The time resolution in this geometry is determined by the crossing angle \( \theta \) and the electrode spacing \( W \). The second geometry operates with an independent probe laser in a nondegenerate four-wave mixing geometry. The time resolution in the nondegenerate geometry can be made independent of both the electrode spacing as well as the crossing angle by focusing the probe beam. The nondegenerate geometry has the added advantage that the autocorrelator can operate for any pump laser wavelengths shorter than the wavelength corresponding to the bandgap of the photorefractive quantum well sample, providing broadband operation in the near infrared and visible spectrum.

4.3.1. Degenerate Four-Wave Mixing

Degenerate four-wave mixing relies on self-diffraction of the laser beams from the photorefractive gratings, as shown in Fig. 4.1(a). The diffracted signal is detected in the first diffraction order by a large-aperture photodetector. The diffraction efficiency from an absorption and phase grating is proportional to

\[ \eta \propto \left( \frac{\pi \Delta n L}{\lambda} \right)^2 + \left( \frac{\Delta \alpha L}{4} \right)^2 \]  

(4.12)

for a thin grating of thickness \( L \). The modulated amplitudes \( \Delta n \) and \( \Delta \alpha \) from Eq.(4.3) depend linearly on \( m(x, \tau) \), leading to a dependence of the diffraction efficiency on the square of the modulation. The total diffracted intensity is proportional to

\[ \eta(\tau) \propto \frac{1}{W} \int_{w/2}^{w/2} m^2(x, \tau) dx \]  

(4.13)
where the integral is taken over the aperture of width $W$ defined by the electrodes, and the transverse beam intensity profile is neglected. Eq. (4.13) implicitly assumes that the photodetector detects all of the diffracted intensity. For a transform-limited Gaussian pulse, the integral in Eq. (4.13) yields

$$\eta(\tau) \propto \text{erf}\left(\frac{W/2 + x_0(\tau)}{\Delta \nu \sqrt{8 \ln 2}}\right) + \text{erf}\left(\frac{W/2 - x_0(\tau)}{\Delta \nu \sqrt{8 \ln 2}}\right)$$  (4.14)

which can be evaluated numerically. The width $\Delta \tau$ (full width at half maximum) of $\eta(\tau)$ cannot be given in closed form, but $\Delta \tau = \sqrt{2} \; t_p$ in the limit when $W$ approaches zero.

From Eq. (4.13) and Fig. 4.1, it is clear that the sensitivity of four-wave mixing for measuring the field autocorrelation depends on the ratio of the aperture width $W$ to the interaction width $\Delta x$ given by Eq. (4.11) for Gaussian pulses. When $W$ is large compared to $\Delta x$, the diffraction does not change appreciably as $x_0$ moves over the interaction region, so no information is gained as a function of pulse separation $\tau$. This is shown in Fig. 4.1 for clarity. When $W$ is small compared to $\Delta x$, then changes in $x_0$ cause changes in diffraction, resulting in excellent time resolution as $\tau$ is varied. To illustrate variations in resolution for two different electrode spacings, the calculated pulse delay width $\Delta \tau$ of the diffraction is shown in Fig. 4.2 as a function of pulse duration $t_p$ for transform-limited Gaussian pulses. The narrower electrode spacing allows sensitive measurements down to much smaller pulse widths than for the wider electrode spacing. For $\theta = 2.44^\circ$, the minimum resolvable pulse width $t_p$ is 155 fs for $W = 1$ mm, and 35 fs for $W = 0.23$ mm for a transform-limited Gaussian pulse.

4.3.2. Nondegenerate Four-Wave Mixing

A drawback of the degenerate four-wave mixing geometry for autocorrelation is that it is only useful for a narrow band of laser wavelengths close to the exciton transition wavelength. We can exploit the thin film character of the photorefractive quantum wells to perform above-bandgap holography [20]. Because of the short 1 micron interaction length of the photorefractive quantum well sample, any laser with a photon energy larger than the bandgap can be used to write the holograms. A second CW laser selected close to the exciton transition energy is then used to probe the photorefractive gratings. The probe laser can be an inexpensive low-power laser diode.
Fig. 4.2 Calculation of diffraction width as a function of pulse duration for transform limited Gaussian pulses. The narrower aperture sample is sensitive to much shorter pulses than the wider aperture sample, down to less than 50 fs pulse durations.

The nondegenerate four-wave mixing autocorrelation geometry is nearly identical to the degenerate case, except for the inclusion of the CW probe laser, as shown in Fig. 4.1(b). The detection scheme can again rely on a broad-area detector that detects the diffracted beam when the interaction area is within the aperture defined by the electrodes. The thin grating character of the photorefractive quantum wells insures that no Bragg condition is necessary to observe the diffracted beam. The conclusions concerning the time resolution are the same as for the degenerate case. An additional possibility, in the case of nondegenerate mixing, is to focus the probe beam to width d within a large aperture, such that d < W. The time resolution in this case is given by comparing Δx to d rather than W. Therefore, nondegenerate mixing provides a way of improving the time resolution of the autocorrelation.

4.4. Experimental Operation

We used two different photorefractive GaAs/AlGaAs multiple quantum well samples [19]. Sample QW-2 and RB-10 are equivalent for this experiment, except that QW-2 has an aperture width of W = 1 mm and RB-10 has an aperture width of W = 0.23 mm. Both samples were grown by molecular beam epitaxy. The optically active region of
QW-2 consists of 60 periods of 75 Å GaAs wells and 100 Å Al$_{0.3}$Ga$_{0.7}$As barriers. The structure for RB-10 is the same as QW-2 except the barriers are 150 Å of Al$_{0.4}$Ga$_{0.6}$As. The optical properties of RB-10 are similar to those of QW-2. Both samples were proton implanted with a dose of $10^{12}$ protons/cm$^2$ at an energy of 160 keV to make the samples semi-insulating. The substrates were removed for transmission experiments. To remove the substrate of QW-2, the sample was epoxied to a glass slide, then etched with a selective etch of citric acid and H$_2$O$_2$ [21]. A lift-off technique was used to remove the substrate of RB-10 [22], and the resulting thin film was van der Waals bonded to a glass slide [23]. Two gold electrodes were evaporated onto each sample to apply an electric field parallel to the quantum wells. The aperture between the gold electrodes of QW-2 had a width $W = 1$ mm, and the aperture of RB-10 had a width $W = 0.23$ mm.

The femtosecond source for the experiment was a Coherent Mira Model 900-F laser. It is a modelocked Ti:Sapphire laser with a 76 MHz repetition rate, tunability from 800 to 900 nm, and pulse durations adjustable around 100 fs. For part of the experiment, the laser was operated in CW mode, but only at discrete wavelengths spaced approximately 2 nm apart. Part of the laser output was diverted into a 0.25 m spectrometer to measure the power spectrum, and part of the output was diverted into an autocorrelator to measure the pulse width.

As shown in Fig. 4.3, the main beam was split into two beams that intersected without focusing in the sample. Before entering the sample, one of the beams passed through a variable delay line. The ratio of the beam intensities was near unity, with a total intensity of approximately 10 mW/cm$^2$. The half angle between the beams was $\theta$, corresponding to an interference fringe spacing of $\Lambda = \lambda / 2 \sin(\theta)$. The sample was oriented with the electric field parallel to the grating vector. Two silicon detectors were placed to detect light transmitted through the sample, one to measure the direct beam, and one to measure first-order diffraction from four wave mixing. The output of each detector was measured with a lock-in amplifier. The lock-in was referenced to the voltage applied to the sample when AC voltage was used, and to a chopper when the voltage was constant. The signals with an AC applied voltage were detected at twice the applied frequency because of the quadratic electro-optic effect.

The results of four-wave mixing for sample QW-2 are shown in Fig. 4.4 for both CW and modelocked operation, with $\theta = 2.6^\circ$, and $\Lambda = 9.6 \mu$m at $\lambda = 840$ nm. An AC field with an amplitude of 5.30 kV/cm was applied to the sample at a frequency of 350 Hz. The CW diffraction efficiency was consistent with previous data [18], while the modelocked diffraction efficiency was reduced in magnitude from the CW measurement.
Fig. 4.3 Experimental geometry. Both degenerate and nondegenerate mixing are shown together, but performed separately. The monochromator and autocorrelator used to characterize the pulses are not shown.

Fig. 4.4 Degenerate 4WM for CW and modelocked operation. The prediction for CW mixing is calculated from the electroabsorption, with the magnitude adjusted to fit the data. The modelocked prediction is calculated from the CW data with no adjustable parameters.
The reduction is caused by the spectral broadening of the laser line during modelocked operation. In the mixing experiment, the broadband pulse may lead to an angular spread of the diffracted beam. However, in our experiments the large area detector is positioned close enough to the sample to capture all the diffracted power. For a Gaussian pulse, the line shape function is given by

\[
g(\lambda, \lambda') = \frac{1}{\Delta \lambda} \sqrt{\frac{4 \ln 2}{\pi}} \exp \left[ -4 \ln 2 \left( \frac{\lambda - \lambda'}{\Delta \lambda} \right)^2 \right]
\]  

(4.15)

where \(\Delta \lambda\) is the full-width at half-maximum. The measured modelocked diffraction efficiency is therefore the average of the CW diffraction efficiency over the line-shape function, given by

\[
\eta_{ML}(\lambda) = \int_0^\infty \eta_{CW}(\lambda') g(\lambda, \lambda') d\lambda'
\]  

(4.16)

In this expression, the only cause of the reduced diffraction efficiency is the broadband detector. If a monochromator were placed after the sample and before the detector, the CW and modelocked diffraction efficiency would be identical.

The predicted four-wave mixing diffraction efficiencies are also shown in Fig. 4.4. The CW prediction is calculated from Eq.(4.12), where the electroabsorption was determined from a transmission experiment and the change in refractive index was calculated from the electroabsorption using the Kramers-Kronig transformation. The peak height was adjusted to fit the data. For the modelocked prediction, Eq.(4.16) was used, assuming the lineshape function was Gaussian. There were no adjustable parameters.

The dependence of degenerate four-wave mixing on relative pulse delay for modelocked laser operation is shown in Fig. 4.5 for sample RB-10 with an electrode spacing of \(W = 0.23\) mm and an applied field of 4.3 kV/cm AC. The average intensity incident on the sample is 6 mW/cm², which is far above the saturation intensity [18], but low enough to prevent Joule heating. The half-angle between the beams is \(\theta = 2.44^\circ\), and the laser wavelength is \(\lambda = 837\) nm, resulting in a fringe spacing of \(\Lambda = 9.8\) \(\mu\)m. By comparing autocorrelator data with the power spectrum, the pulses were determined not to be transform limited. The theoretical curves were calculated from the power spectrum data.

For nondegenerate four-wave mixing, the dependence on pulse delay is shown in Fig. 4.6 for sample QW-2. The pulsed pump beams have a photon energy above the band
Fig. 4.5 Degenerate 4WM for modelocked operation vs. time delay for an aperture width $W = 0.23$ mm. The data is the average of three runs and the predictions are calculated from the Fourier transform of the power spectrum for two pulse bandwidths.

Fig. 4.6 Nondegenerate 4WM for modelocked operation vs. time delay for $W = 1$ mm. The CW probe beam is from a diode laser with a wavelength near the exciton absorption features, while the pulsed pump beams have a photon energy above the band gap of the material. The data is the average of three runs and the predictions are calculated from the Fourier transform of the power spectrum for two pulse bandwidths.
gap, with a central wavelength of 798 nm. The half-angle between the pump beams is $\theta = 2.44^{\circ}$, resulting in an 9.4 $\mu$m fringe spacing $\Lambda$. The probe beam was from a Micralase CW diode laser beam incident normally on the sample, with a wavelength of 836 nm tuned close to the exciton transition. The 1.5 mW/cm$^2$ probe beam intensity was chosen smaller than the 6 mW/cm$^2$ pump beam intensity to prevent erasure of the grating. Again, the pulses were determined not to be transform limited. As before, the theoretical curves are calculated from the power spectrum.

4.5. Discussion

The difference between the data and the calculations in Fig. 4.5 and Fig. 4.6 may be due to field inhomogeneity in the sample. The voltage applied to the sample through the gold electrodes was assumed to result in a uniform field across the aperture. A simple electrostatic calculation shows that the field near the electrodes should be larger than in the middle of the aperture. Also, the gold electrodes may make non-ohmic contact with the semi-insulating sample. Non-ohmic contact could lead to a depletion region near each contact, which would again lead to a stronger field near the electrodes.

In addition to the present experiment, the diffraction could be imaged directly with a CCD camera or linear array for a wide aperture sample. Instead of measuring the diffraction efficiency as a function of the pulse delay, the diffraction efficiency across the array would be detected. The width $W$ of the aperture for this experiment should be at least twice $\Delta x$, so that the entire diffraction region could be imaged at once. This could be achieved by increasing $W$ and increasing $\theta$, which is exactly the opposite of what was required for good resolution in the present experiment. To prevent the interpretation from being unnecessarily complicated, the beams would need to be expanded to approximately plane waves over the width of the aperture. This technique is very similar to the one used in Ref. 16, except that here the different time delays are encoded across a thin hologram, not in the bulk of a volume hologram.

A significant advantage of the photorefractive quantum wells that should be mentioned is the sensitivity of the autocorrelation schemes. The photorefractive quantum well structures represent one of the highest-sensitivity nonlinear optical materials [18]. Under CW operation, the saturation intensity is only 10 $\mu$W/cm$^2$. With a 76 MHz repetition rate and 100 fs pulse durations, pulses with peak intensities of only 1 W/cm$^2$ are sufficient to produce fully-developed photorefractive gratings. Autocorrelation measurements on such weak pulses using conventional nonlinear crystals is extremely
difficult without focusing. In practice, such low powers may not be practical because the autocorrelation relies on diffraction. Diffraction efficiencies in our structures are typically $5 \times 10^{-4}$. Therefore, our practical lower limit is estimated to be 200 $\mu W/cm^2$ average power, and peak powers of 20 W/cm$^2$. The experiments described above were performed with 10 mW/cm$^2$ average power. The low diffraction efficiencies in our samples are not a fundamental property of the photorefractive quantum wells. Diffraction efficiencies for optimized photorefractive quantum well structures are anticipated to approach several percent.

In conclusion, we have demonstrated the use of a photorefractive quantum well structure as an electric-field autocorrelator for 100 fs pulses. Intensity gratings induced by the writing beams have a modulation index that is proportional to the field-autocorrelation or coherence function. Through the photorefractive effect, the changes in the modulation index as a function of pulse delay can be detected by degenerate and nondegenerate four-wave mixing. Degenerate mixing was performed at wavelengths near the excitonic absorption features. Nondegenerate mixing was performed with pulsed writing beams of 800 nm wavelength and a CW diode laser with a wavelength of 840 nm. In principle, the pump could have any photon energy above the band gap of the material. This technique could easily be extended to sensitive cross-correlation of two different ultrafast pulses.
4.6. References


5. DIFFRACTION OF FEMTOSECOND PULSES FROM PHOTOREFRACTIVE QUANTUM WELLS

5.1. Introduction

Ultrafast pulses contain a wide bandwidth, with 4 THz for a typical 100 fs pulse duration, or 10 nm at a center wavelength of 850 nm. The successful use of this bandwidth is desirable for high throughput of coded data in fiber optic communications channels. One method to achieve this is to combine N parallel channels each with bandwidth B into one channel with bandwidth NB, such as through wavelength division multiplexing, with the total bandwidth divided into N frequency intervals that are each modulated independently over the bandwidth B. Another method is time division multiplexing, with the modulation over the entire bandwidth using N time intervals, one for each input channel. Unfortunately, no modulator is available that can operate at 4 THz. One technique to shape ultrafast pulses over their full bandwidth is to use a Fourier-domain pulse shaper to manipulate the pulse in the frequency domain to obtain the desired pulse characteristics [1]. This technique has been extensively demonstrated for fixed Fourier masks [2] and programmable liquid crystal modulator Fourier masks with phase control [3] and amplitude and phase control [4]. Placement of a holographic material in the Fourier plane enables more complex and nonlinear operations, such as reversal of a pulse in time [5]. For dynamic pulse shaping, an ideal diffraction material has flat amplitude and phase response over the bandwidth of the pulses, and a fast response time for high repetition rates. Photorefractive quantum wells are a candidate for the dynamic holographic medium in a pulse shaper [6, 7]. Diffraction from quantum wells relies on absorption and index gratings with strong wavelength dependence. The chief question investigated in this section is whether these strong dispersion effects significantly distort the ultrafast pulse. Photorefractive quantum wells are evaluated for use in a pulse shaper by measuring the shape of a diffracted femtosecond pulse.

Previous work in bulk photorefractive crystals with two-wave mixing of femtosecond pulses includes pulse characterization [8, 9] and pulse shaping [10, 11]. Bulk ferroelectric materials have the advantage of large photorefractive response, but the
disadvantage of long grating formation times. Also, two-wave mixing is trivially phase-matched, but four-wave mixing requires tedious Bragg matching of a third beam in bulk materials. Short pulses can also suffer from broadening by dispersion when traversing a long interaction length in a bulk crystal. In contrast, photorefractive multiple quantum wells have a short grating formation time, and thin film diffraction allows four-wave mixing without a Bragg condition. Quantum wells can also be engineered in several material systems.

In the nondegenerate four-wave mixing geometry described here, femtosecond pulses diffract from a grating written by an above-gap CW diode laser. This is in contrast to previous work with photorefractive quantum wells where a near gap CW diode laser beam diffracts from a grating written by above-gap femtosecond pulses to find the electric-field correlation function. This was accomplished both with direct interference [12], and interference in a Fourier-transform joint correlator geometry (time-to-space mapping) [13]. In both cases, photorefractive quantum wells are used to characterize the shape of femtosecond pulses, but not change their shape, by obtaining the square of the electric-field correlation envelope. The conceptual power of time-to-space mapping is the correspondence to spatial image processing techniques. These techniques are exploited in spectral holography.

The first section presents the theory of diffraction of femtosecond pulses from photorefractive quantum wells and the second section describes measurement of pulse shapes by two different interferometric techniques, electric-field cross correlation and spectral interferometry. The third section describes the photorefractive quantum well optical properties and the experimental procedures, and the fourth part, data and analysis.

5.2. Femtosecond Pulses and Photorefractive Multiple Quantum Wells

In this section, the temporal shape of a femtosecond pulse will be theoretically calculated for pulses transmitted through and diffracted from a photorefractive quantum well. The most convenient method is to Fourier transform the input pulse into the frequency domain. The transmission or diffraction of a pulse is represented by a filter function, which is multiplied by the input electric field and then inverse Fourier transformed to obtain the temporal shape of the output pulse.

Femtosecond pulses are emitted from a modelocked laser in a train with repetition time $T_{REP}$, and a pulse duration is given as the full-width half-maximum (FWHM) $t_p$. The
center frequency is \( \omega_c \), the center wavelength is \( \lambda_c = 2\pi c/\omega_c \), the propagation vector is \( \hat{k} \) and the unit propagation vector is \( \hat{k} \). The electric field for one pulse is

\[
\mathbf{E}(\mathbf{x},t) = \hat{\mathbf{e}} E_0 f(t - \hat{k} \cdot \mathbf{x}/c) \exp \left( i \left( \hat{k} \cdot \mathbf{x} - \omega_c t \right) \right)
\]  

(5.1)

where \( \hat{\mathbf{e}} \) is the polarization vector, \( E_0 \) is the electric field amplitude, and \( f(t) \) is the pulse shape function. Note the sign convention in the exponential. In what follows, the polarization and position are known and fixed, so the polarization vector is dropped, and \( \mathbf{x} \) is set to 0. The electric field simplifies to

\[
e(t) = E_0 f(t) \exp \left( -i\omega_c t \right)
\]  

(5.2)

and the intensity is

\[
I(t) = \frac{1}{2Z} |e(t)|^2 = I_{AVG}T_{REP} |f(t)|^2
\]  

(5.3)

where \( Z \) is the wave impedance, and the normalization of the pulse shape function is chosen to be

\[
\int_{-\infty}^{\infty} |f(t)|^2 dt = 1
\]  

(5.4)

so that

\[
\frac{1}{T_{REP}} \int_{-\infty}^{\infty} I(t) dt = I_{AVG}
\]  

(5.5)

The function \( f(t) \) is typically a hyperbolic secant or a gaussian function for modelocked pulses. For a gaussian,

\[
f(t) \propto \exp \left( -2\ln2 \left( \frac{t}{t_p} \right)^2 \right)
\]  

(5.6)

where \( t_p \) is the FWHM duration.
Transformations between the time and frequency domains are accomplished with the Fourier transforms

\[ E(\omega) = F\{e(t)\} = \int_{-\infty}^{\infty} e(t) \exp \left( i\omega t \right) dt \]

\[ e(t) = F^{-1}\{E(\omega)\} = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(\omega) \exp \left( -i\omega t \right) d\omega \]  \hspace{1cm} (5.7)

which, for gaussian pulses, yields the relationships

\[ I(t) \propto \exp \left( -4\ln 2 \left( \frac{t}{t_p} \right)^2 \right) \quad \quad \quad |E(\omega)|^2 \propto \exp \left( -\frac{1}{4\ln 2} \left( t_p (\omega - \omega_c) \right)^2 \right) \]

\[ \downarrow \quad \uparrow \quad \quad \quad \quad \downarrow \quad \uparrow \]

\[ e(t) \propto \exp \left( -2\ln 2 \left( \frac{t}{t_p} \right)^2 - i\omega_c t \right) \quad \quad \quad E(\omega) \propto \exp \left( -\frac{1}{8\ln 2} \left( t_p (\omega - \omega_c) \right)^2 \right) \] \hspace{1cm} (5.8)

The time bandwidth product for a gaussian is \( t_p \Delta \nu = t_p 4\ln 2 / 2\pi t_p = 2\ln 2 / \pi \approx 0.441 \). If a varying phase is added to the pulse, the pulse may become distorted. Generally, the phase can be expanded in a power series of the frequency. The constant term is trivial. The linear term is also trivial, since it only causes a shift in the conjugate variable. The lowest nontrivial term is the quadratic term, which causes chirp. There may also be higher order terms. Chirp may be caused by dispersion of a pulse transmitted through a material. In this case, the relationships are
\[ I(t) \sim \exp \left( -4 \ln 2 \left( \frac{t}{t_p} \right)^2 \right) \]

\[ E(\omega) \sim \exp \left( -\frac{1}{4 \ln 2} \left( \frac{t_p(\omega - \omega_c)}{\sqrt{1 + \beta^2}} \right)^2 \right) \]

\[ e(t) \sim \exp \left( -2 \ln 2 (1 + i\beta) \left( \frac{t}{t_p} \right)^2 - i\omega_c t \right) \]

\[ E(\omega) \sim \exp \left( -\frac{1}{8 \ln 2} (1 - i\beta) \left( \frac{t_p(\omega - \omega_c)}{\sqrt{1 + \beta^2}} \right)^2 \right) \]

(5.9)

where \( \beta \) is a parameter characterizing the amount of chirp. The sign of \( \beta \) is chosen so that \( \beta > 0 \) corresponds to positive chirp (low frequencies ahead of high frequencies), and normal dispersion \( (dn/d\omega > 0) \). If the chirp is caused by dispersion, then the bandwidth remains constant, that is \( t_p / \sqrt{1 + \beta^2} \) is constant, so larger dispersion results in a longer pulse duration. The time bandwidth product for a chirped gaussian pulse is \( t_p \Delta \nu = 2 \ln 2 \sqrt{1 + \beta^2} / \pi \). Transform limited pulses are the shortest possible pulse for a given power spectrum. Therefore, increasing dispersion causes longer pulses.

The transmission or diffraction of a pulse can be represented by a complex filter function \( H(\omega) \) in the frequency domain. For transmission,

\[ E_T(\omega) = E_{IN}(\omega) H_T(\omega) \]  

(5.10)

Neglecting Fresnel reflection at the surfaces of the thin film and the associated Fabry-Perot effects, the filter function for transmission is

\[ H_T(\omega) = \exp \left( i\tilde{n}(\omega) kL \right) \]  

(5.11)

where \( \tilde{n}(\omega) = n(\omega) + i\alpha(\omega) / 2k \) is the complex refractive index, \( k \) is the propagation constant in free space, and \( L \) is the sample thickness. The transmission spectrum is
\[ T(\omega) = \left| H_T(\omega) \right|^2 = \exp \left(-\alpha(\omega) L\right), \quad (5.12) \]

which can be measured.

In nondegenerate four-wave mixing, femtosecond pulses diffract from a grating written in the semiconductor thin-film by a CW laser with a photon energy above the bandgap of the semiconductor, as shown in Fig. 5.1. Photorefractive quantum wells operate by the photogeneration, transport and trapping of charge at deep level defects, and an electro-optic effect. The interference of the two writing beams creates a periodic intensity pattern \( I(x) \) which induces a refractive index pattern

\[ \tilde{n}(x,\omega) = \tilde{n}(\omega) + \Delta \tilde{n}(\omega) f(Kx + \phi) \quad (5.13) \]

through the photorefractive effect, where \( \Delta \tilde{n}(\omega) = \Delta n(\omega) + i\Delta \alpha(\omega) / 2k \) is the complex change in refractive index caused by the electro-optic effect, which implicitly depends on the applied electric field as well as beam ratio, fringe spacing, and material parameters involving transport. The grating vector is \( K \), and \( \phi \) is the phase offset between the intensity pattern and the index pattern. The function \( f(Kx + \phi) \) is a periodic function with period \( 2\pi \), and is generally not a sinusoid due to the nonlinearities in the transport and the electro-optic effect. The writing beams completely determine the refractive index in Eq. 5.13, where the frequency \( \omega \) is for the probe beam. The spectral dependence in Eq. 5.13 is determined by the transmission and electro-optic spectra of the thin-film structure. The photon energy of the writing beams is implicitly included in \( K \).

In the thin grating limit, the spatially varying phase shift diffracts the incident pulse into many orders without requiring phase-matching. The strongest diffraction is from the lowest harmonic (fundamental) grating, so \( f(Kx + \phi) \) is taken to be \( \cos(Kx + \phi) \) [14]. The total electric-field at the exit face of the material is

\[ E_{ALL}(\omega) = E_{IN}(\omega) \exp \left(i\tilde{n}(\omega) kL\right) \exp \left(\Delta \tilde{n}(\omega) kL\cos (Kx + \phi)\right). \quad (5.14) \]

Using the relation

\[ \exp \left(iz\cos (\delta)\right) = \sum_{m=-\infty}^{\infty} i^m J_m(z) \exp (im\delta) \quad (5.15) \]
and the previous definition for $H_T(\omega)$,

$$E_{ALL}(\omega) = E_{IN}(\omega)H_T(\omega) \sum_{m=-\infty}^{\infty} i^m J_m(\Delta n(\omega) kL) \exp\left(iKx + \phi\right)$$

(5.16)

which contains all diffracted orders. The first-order diffracted electric-field is for $m = 1$,

$$E_D(\omega) = E_{IN}(\omega)H_T(\omega) iJ_1(\Delta n(\omega) kL) \exp\left(iKx + \phi\right).$$

(5.17)

Using the approximation $J_m(z) \approx z^m / 2^m m!$ for small $z$, and neglecting the phases, which are unimportant, the electric field is written in terms of a filter function $E_D(\omega) = E_{IN}(\omega)H_D(\omega)$, where

$$H_D(\omega) = H_T(\omega) \left[\frac{1}{2} \left( \Delta n(\omega) kL + i\Delta \alpha(\omega) \frac{L}{2} \right) \right]$$

(5.18)

and the power spectrum for the diffracted pulse is

$$|E_D(\omega)|^2 = |E_{IN}(\omega)|^2 |H_D(\omega)|^2$$

$$= |E_{IN}(\omega)|^2 |H_T(\omega)|^2 \frac{1}{4} \left( \Delta n(\omega) kL + i\Delta \alpha(\omega) \frac{L}{2} \right)^2$$

(5.19)

and the input diffraction efficiency is $\eta_{IN}(\omega) = |E_D(\omega)|^2 / |E_{IN}(\omega)|^2 = |H_D(\omega)|^2$ and the output diffraction efficiency is $\eta_{OUT}(\omega) = |E_D(\omega)|^2 / |E_T(\omega)|^2 = |H_D(\omega)|^2 / |H_T(\omega)|^2$.

In the time domain, the electric field is $e_D(t) = F^{-1}\{E_D(\omega)\}$. The shape of the pulse is $I_D(t) = |E_D(\omega)|^2 / 2Z$. Unfortunately, it is impossible to directly measure the temporal profile of an ultrashort pulse. Therefore, several indirect techniques must be used, including linear electric-field interference of the signal pulse with a reference pulse, and nonlinear correlation of the intensity profile with a reference pulse. With a known reference pulse, the signal pulse can be fully characterized.
Fig. 5.1. Experimental geometry for diffracting pulses from the photorefractive quantum wells. The shape of the diffracted pulses is inferred from electric-field cross-correlation as well as spectral and spectral interferometry methods. An above-gap CW diode laser writes the grating for nondegenerate four-wave mixing. The CW beam is shown as the dashed line.

5.3. Pulse Shape Measurements

Three linear techniques can be used to determine the shape of the diffracted pulse, as shown in Fig. 5.1. These techniques include finding the electric-field cross-correlation of the diffracted pulses with the reference pulse, finding the spectra of the diffracted and reference pulses, and finding the spectral interference of the diffracted pulse and the reference pulse. The interference of the signal pulse (the transmitted pulse or the diffracted pulse) and the reference pulse (the input pulse) result in a total electric field

\[ E_{TOTAL}(t, \tau) = E_S(t) + E_R(t - \tau) = E_S f_S(t) \exp(-i \omega_S t) + E_R f_R(t - \tau) \exp(-i \omega_R(t - \tau)) \]  \hspace{1cm} (5.20)

where \( \tau \) is the delay of the reference pulse relative to the signal pulse. The reference pulse is the input pulse, and the intensity of the interference is
$$I_{\text{TOTAL}}(t, \tau) = \frac{1}{2Z} \left| E_{\text{TOTAL}}(t, \tau) \right|^2$$

$$= \frac{1}{2Z} \left| E_S(t) \right|^2 + \frac{1}{2Z} \left| E_R(t - \tau) \right|^2 + \frac{1}{Z} \text{Re} \left( E_S(t) E^*_R(t - \tau) \right)$$

$$= I_S(t) + I_R(t - \tau) + 2 \sqrt{I_S I_R} \text{Re} \left( f_S(t) f^*_R(t - \tau) \exp \left(-i\Delta \omega \tau \right) \exp \left(-i\omega_c \tau \right) \right)$$

(5.21)

where $\Delta \omega = \omega_S - \omega_R$. In the following, the frequency difference is incorporated into $f_S$, and $\Delta \omega = 0$. A standard silicon photodetector cannot respond faster than 10 ns, which is much longer than 100 fs. Therefore, the detected intensity is averaged

$$I_{\text{TOTAL}}(\tau) = \int_{-\infty}^{\infty} I_{\text{TOTAL}}(t, \tau) dt$$

$$= I_S + I_R + 2 \sqrt{I_S I_R} \text{Re} \left( \exp \left(-i\omega_c \tau \right) \int_{-\infty}^{\infty} f_S(t) f^*_R(t - \tau) dt \right)$$

$$= I_{\text{TOTAL}} \left( 1 + m \text{Re} \left( \exp \left(-i\omega_c \tau \right) \gamma_{S,R}(\tau) \right) \right)$$

(5.22)

$$= I_{\text{TOTAL}} \left( 1 + m \left| \gamma_{S,R}(\tau) \right| \cos \left( \arg(\gamma_{S,R}(\tau)) - \omega_c \tau \right) \right)$$

where $m = 2 \sqrt{I_S I_R} / (I_S + I_R)$ is the modulation depth of the interference, and

$$\gamma_{S,R}(\tau) = \int_{-\infty}^{\infty} f_S(t) f^*_R(t - \tau) dt$$

(5.23)

is the correlation function, which contains all the useful information about the interference. For a gaussian signal and a gaussian reference pulse, when both are transform limited,

$$\left| \gamma_{S,R}(\tau) \right| \propto \exp \left( -4 \ln 2 \frac{\tau^2}{2(t_S^2 + t_R^2)} \right)$$

(5.24)

which has a width $\tau_{S,R} = \sqrt{2(t_S^2 + t_R^2)}$, and reduces to $\tau_{R,R} = 2t_R$ for an autocorrelation. The correlation function can also be found for the case where the signal pulse is a chirped gaussian, but the analytic expression is complicated, and therefore not directly useful.
Generally, the width of the correlation will be smaller than expected for a transform limited pulse of the same duration, due to the reduced coherence between the signal and reference pulses. In the special case where signal is a chirped gaussian with the same power spectrum as the reference, the duration of the signal pulse is $t_S = t_R \sqrt{1 + \beta^2}$ and the correlation width is $\tau_{S,R} = 2 t_R \sqrt{1 + (\beta/2)^2}$. This is the case where the pulse is transmitted through a transparent material with dispersion.

A connection between the time domain and the frequency domain can be made through the Fourier transform of the correlation function

$$\gamma_{S,R}(\tau) \quad \text{Fourier} \quad F_S(\omega)F_R^*(\omega) \tag{5.25}$$

and in the case of autocorrelation reduces to

$$\gamma_{R,R}(\tau) \quad \text{Fourier} \quad \left| F_R(\omega) \right|^2 \sim \left| E_R(\omega) \right|^2 \tag{5.26}$$

which is just the power spectrum. This is useful for checking consistency between time domain electric-field autocorrelation and the power spectrum.

In the frequency domain, there is interference of the spectra [15, 16]. Following Ref. [16], the electric-field in the frequency domain is

$$E_{TOTAL}(\omega, \tau) = E_S(\omega) + E_R(\omega)\exp\{i\omega\tau\}$$
$$= E_SF_S(\omega - \omega_R) + E_RF_R(\omega - \omega_R)\exp\{i\omega\tau\} \tag{5.27}$$

and the detected intensity from spectral interference is

$$\left| E_{TOTAL}(\omega, \tau) \right|^2 = \left| E_S(\omega) \right|^2 + \left| E_R(\omega)\exp\{i\omega\tau\} \right|^2$$
$$+ 2\text{Re} \left( E_S(\omega)E_R^*(\omega)\exp\{-i\omega\tau\} \right)$$
$$= \left| E_S(\omega) \right|^2 + \left| E_R(\omega) \right|^2 + 2 \left| E_S(\omega) \right| \left| E_R(\omega) \right| \cos \left( \Delta\phi_{S,R}(\omega) - \omega\tau \right) \tag{5.28}$$

where $\Delta\phi_{S,R}(\omega) = \arg \left( E_S(\omega) \right) - \arg \left( E_R(\omega) \right) = \arg \left( F_S(\omega - \omega_R) \right) - \arg \left( F_R(\omega - \omega_R) \right)$ is the phase of the signal spectrum relative to the phase of the reference spectrum. The
interference may seem counterintuitive, because there is interference even when the pulses are separated by a delay much greater than the pulse length. However, since the maximum delay is inversely proportional to the resolution of the spectrometer, a particular wavelength interval is effectively spread in time to a shape that is the inverse Fourier transform of the spectrometer bandpass function. Therefore, a resolution that is ten times smaller than the pulse bandwidth will allow spectral interference when the pulses are separated by as much as 10t_p. An equivalent viewpoint is that the delay must be short enough for the spectrometer to resolve the interference fringes. If the maximum delay is exceeded, the fringes are washed out so that the measured quantity is the sum of the individual power spectra.

The spectral interference contains both individual spectra as well as the interference. The individual spectra are each measured separately and subtracted from the interference to obtain the interference spectrum

\[
S(\omega, \tau) = \left| E_{TOTAL}(\omega, \tau) \right|^2 - \left| E_S(\omega) \right|^2 - \left| E_R(\omega) \right|^2
= 2 \text{Re} \left( E_S(\omega) E_R^*(\omega) \exp(-i\omega \tau) \right)
= 2 E_S E_R \text{Re} \left( F_S(\omega - \omega_R) F_R^*(\omega - \omega_R) \exp(-i\omega \tau) \right)
\]

which has a functional form similar to the correlation function, i.e., a pulse function on a carrier or an envelope with oscillations. Also, both the correlation and the spectral interferometry data are real parts of a complex function. To calculate the envelope magnitude and the phase of the oscillations, one can recover the complex function from the real function. For a function \( x(t) \) (such as the electric field correlation),

\[
x(t) = 2 \text{Re} \left( \tilde{x}(\tau) \exp(-i\omega_c \tau) \right)
= \tilde{x}(\tau) \exp(-i\omega_c \tau) + \tilde{x}^*(\tau) \exp(i\omega_c \tau)
\]

\[
F\{ x(t) \} = \tilde{X}(\omega - \omega_c) + \tilde{X}^*(-\omega - \omega_c)
\]

\[
\theta(\omega) F\{ x(t) \} = \tilde{X}(\omega - \omega_c)
\]

\[
F^{-1}\{ \theta(\omega) F\{ x(t) \} \} = \tilde{x}(\tau) \exp(-i\omega_c \tau)
\]
where \( \theta(\omega) \) is the step function that selects the positive frequency values. For a function \( X(\omega) \) (such as the spectral interference),

\[
X(\omega) = 2\text{Re}\left( \tilde{X}(\omega) \exp\left( i\omega \tau \right) \right) \\
= \tilde{X}(\omega) \exp\left( i\omega \tau \right) + \tilde{X}^*(\omega) \exp\left( -i\omega \tau \right)
\]

\[
F^{-1}\{X(\omega)\} = \tilde{x}(t - \tau) + \tilde{x}^*(t - \tau) \tag{5.31}
\]

\[
\theta(t)F^{-1}\{X(\omega)\} = \tilde{x}(t - \tau)
\]

\[
F\left\{ \theta(t)F^{-1}\{X(\omega)\} \right\} = \tilde{X}(\omega) \exp\left( i\omega \tau \right)
\]

In both cases, the transform of the real function is the sum of the desired function shifted to the right and its complex conjugate shifted to the left. The overall operation is valid when the function and its conjugate do not overlap at the origin, so that the step function selects all of the desired function and none of its conjugate. For the electric-field cross-correlation using Eq. 5.30, the condition is that the bandwidth of the pulse is much smaller than the center frequency. This is equivalent to a slowly varying envelope approximation where there are many oscillations under the envelope of the function. This is true for 100 fs pulses in the infrared, but breaks down for 10 fs pulses. Similarly, for the spectral interference using Eq. 5.31, the condition is that the delay must be much larger than the pulse width, again so that there are many oscillations under the envelope. In this case, the delay can be chosen to satisfy this condition.

Both interferometric techniques depend on the characteristics of the reference pulse. Spectral interference of two pulses provides the relative phase of the pulses. If the reference pulse is close to transform limited, the phase difference is the phase of the signal pulse. The combination of second-harmonic autocorrelation and measurement of the spectrum of the reference pulse can be used to estimate how close the pulse is to transform limited. When the reference pulse is not transform limited, a technique such as frequency resolved optical gating (FROG) \(^{17}\) must be used to fully characterize the reference pulse. Standard techniques are sufficient for this work because the reference pulses are assumed to be well behaved and the frequency difference between the signal pulse and the reference pulse is the quantity of interest.

Second harmonic generation correlation depends on the intensity of the pulse. For background free second harmonic correlation, the second harmonic output intensity is
\[ I_{\text{2\omega}}^{BF}(\tau) = \langle I_{\text{2\omega}}(t, \tau) \rangle = \langle I_S(t) I_R(t - \tau) \rangle \]  

(5.32)

where \( \langle \rangle \) denotes time average over a repetition period. The interferometric second harmonic generation correlation depends on the relative phase of the two pulses. If the amplitudes are taken to be equal, the second harmonic intensity (using the total electric field from Eq. 5.20) is

\[
I_{\text{2\omega}}^{\text{INT}}(\tau) = \langle I_{\text{2\omega}}(t, \tau) \rangle \\
\propto \langle |E_{\text{2\omega}}(t, \tau)|^2 \rangle \\
\propto \langle |E_{\text{TOTAL}}^2(t, \tau)|^2 \rangle \\
\propto \langle |f_S(t)|^4 + |f_R(t - \tau)|^4 + 4|f_S(t)|^2|f_R(t - \tau)|^2 \\
+ f_S^2(t)f_R^2(t - \tau)\exp(-2i\omega_c\tau) \\
+ 2f_S^2(t)f_S^*(t)f_R^*(t - \tau)\exp(-i\omega_c\tau) \\
+ 2f_S(t)f_R(t - \tau)f_R^*(t - \tau)\exp(-i\omega_c\tau) \rangle .
\]

(5.33)

The measured intensity at the peak is eight times the value for large pulse separations. When the interferometric intensity is averaged over one interference fringe,

\[
\omega_c \int_0^{\omega_c^{-1}} I_{\text{2\omega}}^{\text{INT}}(\tau - \tau')d\tau' = \langle |f_S(t)|^4 + |f_R(t - \tau)|^4 + 4|f_S(t)|^2|f_R(t - \tau)|^2 \rangle \\
= I_S^2 + I_R^2 + 4I_{\text{2\omega}}^{BF}(\tau)
\]

(5.34)

which is the background free result with a background due to the second harmonic of each pulse. For gaussian pulses,

\[ I_{\text{2\omega}}^{BF}(\tau) \propto \exp\left(-4\ln2 - \frac{\tau^2}{t_S^2 + t_R^2}\right) \]

(5.35)
which has a width of \( \tau_{S,R}^{BF} = \sqrt{\tau_S^2 + \tau_R^2} \), and reduces to \( \tau_{R,R}^{BF} = \sqrt{2} \tau_R \) for autocorrelation.

5.3. Experiment

The multiple quantum well structure used in our experiment was grown by molecular beam epitaxy in a Varian Gen II chamber. The growth began with 0.5 \( \mu \)m GaAs on a semi-insulating GaAs substrate, followed by 500 nm of Al\(_{0.5}\)Ga\(_{0.5}\)As, 20 nm of AlAs, and 10 nm of GaAs was grown for substrate removal etches. After 250 nm of Al\(_{0.1}\)Ga\(_{0.9}\)As, 85 periods of 7.5 nm GaAs wells and 10 nm of Al\(_{0.1}\)Ga\(_{0.9}\)As barriers were deposited, followed by 150 nm of Al\(_{0.1}\)Ga\(_{0.9}\)As. After growth, the samples were proton implanted with a dose of 10\(^{12}\) cm\(^{-2}\) at 160 keV and 5 x 10\(^{11}\) cm\(^{-2}\) at 80 keV to create deep level defects and render the material semi-insulating [18, 19] and photorefractive [6, 20]. The implanted wafer was cleaved into 3 mm x 3 mm pieces. To remove the substrate for transmission experiments, the substrate was lapped to 100 \( \mu \)m, and then etched with a 1:19 ammonium hydroxide/hydrogen peroxide etch to the 50\% Al stop etch layer [21]. Then an HF acid etch removed the 50\% Al layer and the AlAs layer [22]. Gold contacts are evaporated directly onto the low bandgap GaAs layer with a 1 mm aperture across which a field is applied in the plane of the quantum wells, perpendicular to the growth direction.

The transmission and electro-optic effect of the photorefractive quantum wells are shown in Fig. 5.2. The data are measured using a 1000 W halogen source filtered by a 0.85 m double spectrometer. There is a peak absorption due to the quantum-confined heavy-hole exciton of approximately 10\(^4\) cm\(^{-1}\) for the 1.9 \( \mu \)m thickness of the quantum well region. The change in absorption and the change in the refractive index spectra due to the electro-optic effect are shown in Fig. 5.2(b). The electro-optic effect with a field applied in the plane of the quantum wells is due to the field ionization of the excitons, which broadens both the light-hole exciton absorption feature and the heavy-hole absorption feature, resulting in a characteristic double dip. The differential transmission is measured by applying a sinusoid that varies between 0 V and 400 V with a frequency of 277 Hz. The change in absorption is calculated using

\[
\Delta \alpha(F,\lambda) = -\frac{1}{L} \ln \left( \frac{\Delta T(F,\lambda)}{T(\lambda)} + 1 \right)
\]  

(5.36)
Fig. 5.2. Optical properties of the photorefractive quantum wells: (a) transmission spectrum; (b) spectra for change in absorption and change in refractive index due to electro-optic effect, with 4 kV/cm applied field. The refractive index change is calculated from the change in absorption using the Kramers-Kronig transform.
Fig. 5.3. Calculated diffraction spectra of the photorefractive quantum wells: (a) input diffraction efficiency; (b) diffraction phase. The diffraction efficiency is calculated using both the transmission spectrum and electro-optic effect. The phase is only calculated from the electro-optic effect. Note that the phase is flat near 850 nm.
where $L$ is the thickness and $F$ is the applied field. The change in refractive index was calculated using the Kramers-Kronig transformation

$$
\Delta n(\omega) = \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\Delta \alpha(s)}{s^2 - \omega^2} ds
$$

(5.37)

with a maximum absorption change of 1000 cm$^{-1}$ and a maximum refractive index change of 0.005. The CW input diffraction efficiency spectrum shown in Fig. 5.3(a) is calculated directly from the transmission and electro-optic spectra using Eq. 5.19. Since the heavy-hole exciton oscillator strength is much larger than light-hole exciton oscillator strength, the peak is almost entirely due to the heavy hole exciton, with a maximum input diffraction efficiency of $10^{-4}$. The spectrum of the diffracted phase shown in Fig. 5.3(b) is calculated only from the changes in absorption and refractive index, and not from the background index spectrum, which is not known in detail. Despite the oscillation in the electro-optic spectrum, the phase is relatively flat. That is, the change in refractive index spectrum is approximately the same as for the change in absorption, but shifted by a constant phase, because of the relationship through the Kramers-Kronig transform.

The femtosecond pulse is diffracted from the photorefractive quantum well using non-degenerate four-wave mixing, as shown in Fig. 5.1. The femtosecond pulse source is a Clark NJA-4 modelocked Ti-Sapphire laser (with 100 MHz repetition rate), pumped by a Coherent Innova-310 Argon Ion laser. A 685 nm diode laser is the source for the writing beams, with a photon energy above the bandgap of the quantum well barriers. The fringe spacing of the interference pattern is 10 $\mu$m, which is far above the resolution limit of 3 $\mu$m and below the limit where scattered light from the transmitted beam overlaps with the diffraction. The incident intensity from the writing beams is 2 mW total with a beam ratio of unity, while there is 2 mW in the probe pulsed beam, which results in a total intensity of approximately 40 mW/cm$^2$. To obtain the largest diffraction efficiency, the writing beams should be much stronger than the probe, but diffracted power is maximized when the ratio of the writing beam power to the probe power is near unity. An advantage of photorefractive quantum wells is their ultra-low sensitivity, where the gratings are fully developed for a saturation intensity of 10 $\mu$W/cm$^2$. There is a trade-off between response time and intensity, with shorter response times for higher intensities. At an intensity of 40 mW/cm$^2$, the response time is about 10 $\mu$s. Over the duration of one 100 fs pulse, the photorefractive grating remains essentially static. To obtain diffraction, a field is applied by
applying 400V DC over a 1 mm gap for an average field of 4 kV/cm, and the writing beams are chopped at 1.0 kHz.

The shape of the pulse is measured using the linear techniques described above, which use an undistorted reference pulse. The delay line for the reference pulse uses a hollow retroreflector mounted on a stepper-motor driven translation stage with a 0.1 \( \mu \text{m} \) step size. A biased p-i-n silicon photodetector is used for electric-field correlation. Spectra are measured with a 0.275 m spectrometer with a CCD for capture. Nonlinear correlation is accomplished with 0.5 mm thick BBO with a photomultiplier tube to detect the second harmonic. Both field and second-harmonic correlation scans are performed by chopping the writing beams and measuring the signal with a lockin amplifier. For correlation scans, the envelope function can be used to estimate the signal pulse shape, given the properties of the reference pulse. However, the combination of the uncertainty of the translation stage and drift of the relative path lengths as the translation stage is scanned makes phase information unreliable for correlation scans. There is no drift between adjacent points in the spectra because the CCD captures all points simultaneously.

5.4. Data and Analysis

The characteristics of the reference pulse are important for interferometric and correlation techniques. Pulses from the laser used in this experiment are typically 100 fs in duration and are nearly transform limited gaussian pulses. The reference pulse electric field autocorrelation is shown in Fig. 5.4(a), and the interferometric second harmonic autocorrelation is shown in Fig. 5.4(b). The sign convention is that positive delays correspond to the reference pulse arriving after the signal pulse at the detector. Therefore, the leading edge of the pulse is on the left, so the pulse appears to be moving from right to left. With a 100 nm step size for the translation stage and approximately 850 nm center frequency, there are over four points per interference fringe. The heavy line in Fig. 5.4(a) is the envelope of the correlation function \( \gamma_{R,R}(\tau) \) calculated using Eq. 5.30, which has a width of 213 fs. The power spectrum is shown in Fig. 5.5(a) with a width of 8.8 nm, and the average of the second harmonic autocorrelation is shown in Fig. 5.5(b) with a width 158 fs. Both curves in Fig. 5.5 are fitted with gaussian functions to show that the pulse is approximately a gaussian shape. Electric field autocorrelation of the reference and the Fourier transform of the spectrum match well, as shown in Fig. 5.6. These curves should match according to Eq. 5.26, which verifies that the spectral data is consistent with the correlation data. The time-bandwidth product is calculated directly from the spectrum.
Fig. 5.4. Reference pulse auto-correlation vs. delay: (a) electric field auto-correlation of reference pulse; (b) interferometric second harmonic autocorrelation of reference pulse. In (a), the heavy line is the calculated envelope.
Fig. 5.5. Gaussian fits to reference pulse: (a) gaussian fit to reference pulse spectrum; (b) gaussian fit to reference pulse second harmonic autocorrelation, which is the average of the data in Fig. 5.4(b). Both curves are well fit by a gaussian. The reference pulse is nearly transform limited.
using $t_p \nu = t_p c \Delta \lambda / \lambda^2$, where $c = 300$ nm/fs, and $t_p$ is calculated from the second harmonic autocorrelation width using Eq. 5.30. The time-bandwidth product for the reference pulse is 0.41, which is consistent with a transform limited gaussian pulse.

The absorption and refractive index spectra distort the transmitted pulse. The electric field cross-correlation of the transmitted and reference pulses is shown in Fig. 5.7(a), and the interferometric cross-correlation is shown in Fig. 5.7(b). The transmitted pulse has a tail on the following edge of the pulse. The power spectrum is shown in Fig. 5.8(a) and the phase spectrum is shown in Fig. 5.8(b), calculated from the spectral interference of the transmitted pulse with the reference pulse. The tail is due to either the amplitude or phase of the transmission function $H_T(\omega)$. In Fig. 5.9, the power spectrum data is compared with the power spectrum calculated from the product of the transmission in Fig. 5.2(a) and from the power spectrum of the reference pulse. In Fig. 5.10, the electric-field cross-correlation data is compared with calculations from spectral measurements. The calculation using both the amplitude and the phase nearly matches the data, while the calculation without the phase fails to produce a tail. Therefore, photorefractive quantum wells distort transmitted pulses due to both absorption and refractive index variation over the bandwidth of the pulse.
Fig. 5.7. Cross-correlation of transmitted pulse with reference pulse vs. reference pulse delay: (a) electric field cross-correlation; (b) interferometric second harmonic cross-correlation. The transmitted pulse shape has gained a tail on the following edge of the pulse.
Fig. 5.8. Spectrum of transmitted pulse: (a) transmitted and reference pulse spectra; (b) transmitted pulse phase measured by spectral interference. There is a notch in the transmitted pulse spectrum due to the heavy hole exciton absorption feature.
Fig. 5.9. Spectrum of transmitted pulse and calculated spectrum.

Fig. 5.10. Electric field cross-correlation of transmitted pulse. The first calculation uses the amplitude and phase from the spectral measurements, while the second calculation uses only the amplitude, with the phase set to zero. The phase is necessary to describe the tail on the correlation.
The peak of the diffraction efficiency spectrum in Fig. 5.3(a) broadens the diffracted pulse by reducing its bandwidth. The electric-field cross-correlation of the diffracted and reference pulses is shown in Fig. 5.11. The calculated envelope has a width of 433 fs, more than twice as long as the reference field autocorrelation width of 213 fs. Assuming the diffracted pulse is a transform limited gaussian, Eq. 5.24 can be used to estimate that the diffracted pulse duration is

\[ t_s = \sqrt{\frac{t_{SR}^2}{2} - t_R^2} \]  \hspace{1cm} (5.38)

which is 285 fs, where the reference pulse duration is the same as used above. The increased duration of the diffracted pulse compared to the reference pulse is in general due to a combination of the decreased bandwidth of the pulse, and the variation of the refractive index over the bandwidth, that is, chirping (or higher order) changes in the phase. In Fig. 5.12(a), the spectra of the reference and diffracted pulse are shown. The diffraction spectrum appears to also be nearly gaussian, and the spectrum and the estimate of the pulse length are used to calculate a time-bandwidth product of 0.43, also very close the that for a gaussian. The phase in Fig. 5.12(b) is nearly flat, confirming that the pulse is nearly transform limited.

The spectrum of the diffracted pulse is calculated from the electro-optic data and compared with the data in Fig. 5.13. The difference between the data and the calculation is likely due to the difference between the modulation depth of the space-charge field during photorefractive mixing and the modulation of the field used for differential transmission. Even with a beam ratio of unity in the writing beams, the effective modulation of the space-charge field will be limited by the transport. If the effective space-charge modulation were known, then a precise comparison could be made. Some error may also be due to nonuniform distribution of the field between the contacts [23], but this effect should be small since the incident intensity is high.

The electric-field correlation of the diffracted and reference pulses is calculated from the spectral data using Eq. 5.18 and compared with the data in Fig. 5.14. The calculation with and without the phase are nearly identical, indicating that the phase variation of the diffracted pulse is small, so the pulse is nearly transform limited.

In conclusion, pulses diffracted from photorefractive quantum wells are broadened, but remain nearly transform limited. The broadening is due to the bandwidth of the diffraction efficiency spectrum, which is determined by the electro-optic properties of the
quantum wells. This indicates that photorefractive devices will be useful for pulse shaping. Future work includes the design of structures with wider bandwidth through bandgap engineering of the quantum well structure [24]. To obtain more diffracted power, asymmetric Fabry-Perot structures [25], and perpendicular geometry p-i-n structures [26] are being explored. Finally, photorefractive materials that operate at a wavelength of 1.5 μm [27] are required for pulse shaping compatible with fiber-optic communications technology.

Fig. 5.11. Electric field cross-correlation of the diffracted pulse and the reference pulse. The diffracted pulse is estimated to be 2.5 times longer than the reference pulse.
Fig. 5.12. Spectra of the diffracted pulse: (a) power spectrum of the diffracted pulse compared to reference pulse; (b) phase of the diffracted pulse. The diffracted pulse is also nearly a gaussian shape.
Fig. 5.13. Spectrum of the diffracted compared with calculation from transmission and electro-optic data: (a) linear scale; (b) log scale.
Fig. 5.14. Electric-field cross correlation of diffracted pulse with reference pulse. The calculation is from spectral data. The small difference between the calculation with and without phase indicates the pulse is nearly transform limited.
5.5. References


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Robert M. Brubaker was born in San Diego, California on December 27, 1965, the only child of Elfrieda and Robert H. Brubaker. He spent most of his rambunctious years in Maryland, enjoying both the Appalachian Mountains to the west, and the Chesapeake Bay and Atlantic Ocean to the east. While attending Seneca Valley High School in Germantown, Maryland, he developed a strong interest in math and science, and decided to pursue a degree in electrical engineering. After attending the University of Maryland College Park, he discovered that engineering is fun, but physics is the bedrock of understanding. He entered Purdue University in 1989 to study experimental condensed matter physics, and was fortunate to be the third student of Prof. David D. Nolte. He has accepted a position with the Eastman Kodak Corp. in the Microelectronics Technology Division, which sponsored him as a Kodak Fellow. He looks forward to the bagels and snow in Rochester, New York.