LINEAR, NONLINEAR OPTICAL AND TRANSPORT PROPERTIES OF
QUANTUM WELLS COMPOSED OF SHORT PERIOD
STRAINED InAs/GaAs SUPERLATTICES

DISSERTATION

Presented to the Graduate Council of the
University of North Texas in Partial
Fulfillment of the Requirements

for the Degree of

DOCTOR OF PHILOSOPHY

By

Xuren Huang, B.S., M.S.
Denton, Texas
December, 1993
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In this work, ordered all-binary short-period strained InAs/GaAs superlattice quantum wells were studied as an alternative to strained ternary alloy InGaAs/GaAs quantum wells. InGaAs quantum wells QWs have been of great interest in recent years due to the great potential applications of these materials in future generations of electronic and optoelectronic devices. The all-binary structures are expected to have all the advantages of their ternary counterparts, plus several additional benefits related to growth, to the elimination of alloy disorder scattering and to the presence of a higher average indium content.

A series of ordered all-binary InAs/GaAs superlattice quantum wells was grown by either molecular beam epitaxy or migration enhanced epitaxy. The very high optical quality of these samples was demonstrated by linear optical absorption spectroscopy, photoluminescence and time-resolved photoluminescence. The nonlinear optical properties of the all-binary quantum wells were measured with picosecond differential transmission and four wave mixing techniques. Consistency of the results obtained with both techniques was demonstrated by a Kramers-Kronig analysis, and it was found that the measured nonlinearities are comparable to the best results obtained from frequently studied GaAs/AlGaAs and InGaAs/InP quantum wells. The in-well ambipolar diffusion coefficient of the all-binary quantum wells was measured at 85 K with a photo-induced transient grating technique and the results were compared with those obtained from ternary alloy InGaAs/GaAs quantum wells. The results show that in spite of the very good linear and nonlinear optical properties and the binary structure, as evidenced by transmission electron
microscopy measurement, of these InAs/GaAs superlattice quantum wells, the expected elimination of alloy disorder scattering may not have been achieved yet.

These investigations demonstrate that the unique ordered all-binary short-period strained InAs/GaAs superlattice quantum wells have technologically significant optical and electronic properties and therefore may have many applications.
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CHAPTER I

INTRODUCTION

Semiconductor quantum wells, or more generally, semiconductor nanostructures are man-made hetero- and/or modulation-doped structures of nanometer dimensions, in which the quantum size effect plays an important role in determining optical and electronic properties.

Much of the recent interest in semiconductor nanostructures has been caused by the possibility of using these structures to fabricate high performance electronic and optoelectronic devices. Semiconductor nanostructures are far superior to bulk materials for many electronic and optoelectronic device applications because of their low dimensional nature and their tailor able band structures and therefore may have tremendous applications in the future. As the technology of silicon integrated circuits approaches its fundamental limits, it is widely acknowledged that new technologies are necessary to further increase the information processing capacity of integrated circuits. Substantial improvements in performance have been achieved by using high speed electronic devices fabricated from III-V semiconductor nanostructures. Further improvements in performance may obtained by using hybrid electronic and optoelectronic systems, in which optical interconnections are used to replace conventional electrical connections, and semiconductor nanostructure may be exploited for optoelectronic devices and optical wave guides in those systems. Eventually, extremely high performance may be obtained from all-optical logic circuits in which semiconductor nanostructures might play a vital role.
Interest in semiconductor nanostructures increased sharply more than two decades ago when new types of device, such as the resonant tunneling diode and the Bloch oscillator, which could be fabricated from those structures were discussed by Esaki and Tsu (1970). Since then, many novel devices fabricated from semiconductor nanostructures have been developed. Some of the most successful examples of those devices are: low threshold, high efficiency quantum well lasers (Tsang 1987, Noblanc 1986, Burnham et al. 1984), high speed heterostructure transistors (Morkoç 1987, Linh 1987, Abe et al. 1987), and optoelectronic switches and modulators based on the quantum confined Stark effect (Miller et al. 1984).

The construction of the semiconductor nanostructure devices mentioned above has been made possible by the development of new material growth techniques, especially molecular beam epitaxy (MBE), pioneered by Cho and Arthur (1975). The MBE technique allows the control of material composition, doping and thickness on the scale of a single atomic layer, and hence enables the precise control of band gap and band structure in order to tailor the transport and optical properties of materials to suit specific applications. The precise control of the position of doping layers and the quantum confinement of excess carriers results in the spatial separation of ionized impurities and excess carriers, which is mainly responsible for the enhanced speed of carriers in transistors fabricated from nanostructures. In the case of the quantum well lasers, low threshold currents are achieved due to the two-dimensional nature of the density of states and suitable tailoring of the band structure.

Interest in semiconductor nanostructures fabricated from lattice mismatched materials did not arise until 1982, more than a decade after the first research on semiconductor nanostructures. The early research was concentrated on lattice-matched material systems, e.g. GaAs/AlGaAs, in order to avoid any built-in strain in the structure,
semiconductor nanostructures. The early research was concentrated on lattice-matched material systems, e.g. GaAs/AlGaAs, in order to avoid any built-in strain in the structure, and hence the misfit dislocations which appear if the thickness of the epitaxial layer exceeds the critical thickness determined by the limit of elastic strain containment of the layer. While this approach, which avoided the extra complications associated with strain, was reasonable in the relatively early stages of semiconductor nanostructure research, it resulted in limited spectral coverage of optoelectronic devices fabricated from these structures. In 1982, Osbourn proposed that the strain associated with the heteroepitaxy of dissimilar materials may itself offer a new functionality, namely lifting of the valence band degeneracy and alteration of the valence band structures and band-gaps, thereby allowing the attainment of wavelengths of operation inaccessible to the lattice-matched material systems. It has subsequently been seen that the advantages associated with the modified band structure far outweigh the disadvantages of the presence of strain. Many devices have exploited the possibilities created by this breakthrough in thinking. These include InGaAs/GaAs high hole mobility transistors (HHMT) and low threshold strained layer InGaAs/AlGaAs quantum well lasers (Adams 1986, Yablonovitch and Kane 1986, 1988, Choi 1990), both of which take advantage of the strain-induced splitting of valence bands and the in-plane light-hole-like nature of the uppermost valence band; and a silicon-based photodiode with an absorption edge strain-shifted to the 1.3~1.5 μm window for optical fiber communications (Temkin et al. 1986). Furthermore, the development of devices based on strained heterostructures is a big step towards the goal of integrating optoelectronic and silicon-based electronic devices (Pearsall 1990).

The most technologically important strained nanostructures which have been studied so far are fabricated from the InGaAs/GaAs. This material system is attractive because the operating wavelength of devices fabricated from it can be adjusted in the
range 0.9 to 1.1 μm, which includes the 0.98 μm required for the pump source of the Er-doped optical fiber amplifiers used in optical communication networks. Also, the effective mass of the conduction band electrons is small due to the In content; and the in-plane effective mass of uppermost valence band holes is small (m* = 0.14 m₀ for In₀.₂Ga₀.₈As on GaAs substrate) because of the biaxial compression of the InGaAs layers. Finally, the GaAs substrate is transparent at the wavelength of operation. These InGaAs/GaAs structures have been used in the construction of many high mobility p-channel transistors and low threshold lasers.

This thesis describes the study of highly strained, ordered, all binary InAs/GaAs strained layer superlattices as an alternative to the ternary InGaAs alloy. These ordered all-binary structures have all the advantages of their disordered ternary counterparts, plus several additional advantages related to growth, to the elimination of alloy scattering and to the accommodation of a higher average In content.

Chapter II is a brief review of theoretical descriptions of the electronic and optical properties of quantum wells, the ultrafast dynamics of photo-generated carriers and optical nonlinearities, and the effects of strain on quantum well band structures. Fundamental differences between two-dimensional and three-dimensional structures are described, and strained and unstrained material systems are compared. The device application of quantum wells and strained systems are considered.

Chapter III includes details of the motivation for using InAs/GaAs strained layer superlattices to replace InGaAs alloys. The procedures used to grow the structures are discussed, and the methods used to characterize the structures, including transmission electron microscopy, optical absorption spectroscopy, time-integrated photoluminescence, and picosecond time resolved photoluminescence, are described. The results show that
the InAs/GaAs strained layer superlattice structures described in this thesis are of very good quality.

The measurement of the optical nonlinearities of InAs/GaAs superlattice quantum wells is described in chapter IV. The physical mechanisms underlying the nonlinearities and the theories of pump-probe and four wave mixing techniques are discussed in §4.1. The picosecond pump-probe and four-wave techniques used to measure the optical nonlinearities are described in §4.2. In §4.3 the measured results are presented and discussed, the consistency of the results is checked via Kramers-Kronig transformation, and the measured results are compared with those of other material systems.

The measurement of the ambipolar diffusion coefficients of carriers in all-binary InAs/GaAs strained layer superlattice quantum wells and in equivalent ternary alloy quantum wells, and the comparison of the results of these measurements are described in chapter V. The measurement techniques and the theory of diffraction from transient gratings are discussed in §5.1. The samples studied and experimental setup used are described in §5.2. The nonlinear optical absorption spectra of samples at different temperatures are presented and analyzed in §5.3, as are the results of a time-resolved differential transmission measurement, and a discussion of carrier recombination behavior. The results of the measurement of ambipolar diffusion coefficient are presented in §5.4 and are discussed in §5.5.

The entire work is summarized in chapter VI.
CHAPTER II

REVIEW OF ELECTRONIC, OPTICAL AND TRANSPORT PROPERTIES OF SEMICONDUCTOR QUANTUM WELLS

A large body of work on semiconductor quantum wells (QWs) has been produced since the pioneering work of Esaki and Tsu (1970). The development of sophisticated growth techniques for layered semiconductor nanostructures has allowed QWs to be fabricated. The possibility of novel physics and the potential applications of QWs in devices exploiting fast transport, optical nonlinearities and stimulated emission have inspired much theoretical and experimental work. In this chapter, several key properties of quasi two-dimensional (2D) semiconductor nanostructures will be reviewed. These include quantum confinement and the energy levels of carriers in QWs, 2D density of states and enhanced excitonic effect in QWs, optical nonlinearities induced by photon-generated free carriers, and the effects of strain on energy levels and the conduction and valence band structures.

§ 2.1 Energy bands and the effective masses of particles in bulk semiconductors

We start by reviewing some basic concepts in semiconductor physics, specifically, energy bands and effective masses of particles in bulk semiconductors. The energy band structure of a crystal, which describes the relationship between the energy and momentum of a carrier in the crystal, originates from the periodicity of the material. It can be shown
clearly by using the somewhat over-simplified Kronig-Penney model of the onedimensional crystal (Kronig and Penney 1930, Seeger 1989), that carriers in a potential composed of a series of square wells can have an energy only in one of several energy bands, which are separated by forbidden gaps (see Fig. 2.1). While many other more complicated models (see e.g. Seeger 1989, Wolfe et al. 1989) have been used to calculate the band structures of real crystals, the results are qualitatively similar: a series of energy bands separated by forbidden gaps.

![Dispersion relation E(k) for an electron in a one-dimensional Kronig-Penney crystal which is composed of a series of square potential wells. (After Seeger 1989).](image-url)
The electrical conductivity of a crystal is largely determined by the distribution of carrier among the energy bands (see e.g. Kittel). In an insulator, electrons exactly fill one or more bands and leave the others empty. Provided that a filled band is separated by an energy gap from the next higher band, there is no continuous way to change the total momentum of the electrons if every accessible state is filled. An external electric field cannot therefore cause significant current flow in an insulator. In contrast, in metals, one of the energy bands is partially filled, hence the momentum of the electrons in this band can be changed by an external electrical field and the metal is therefore a conductor. In a semiconductor with no impurities, which is known as an intrinsic semiconductor, all the energy bands are either exactly filled or completely empty at absolute zero, and the material is therefore an insulator at this temperature. However, as the temperature increases, some electrons are thermally excited into the empty band just above the filled band, and both bands consequently become somewhat conductive. The intrinsic conductivity and intrinsic carrier concentrations are largely controlled by $E_g/k_B T$, the ratio of the forbidden band gap to temperature. Materials with very large $E_g/k_B T$ are considered to be insulators, and most intrinsic semiconductors are indeed insulators at very low temperature. Actually, there is no clear boundary to separate insulators and semiconductors: as the name suggests, semiconductors are an intermediate case between insulators and conductors.

The highest band which is filled at 0 K in a semiconductor is defined as the valence band. The energy band just above the valence band, which is empty at 0 K, is defined as the conduction band. The lowest point in the conduction band is called the conduction band edge; the highest point in the valence band is called the valence band edge. The energy difference between the conduction and valence band edges is referred to as the band gap.
An electron can be excited, thermally, optically or electrically, into a state in the conduction band from a bound state in the valence band. The movement of an electron in the conduction band can be treated approximately as a particle in a free space with a mass equal to the effective mass, which is given by

\[ m^* = \frac{\hbar^2}{d^2E/dk^2}. \] (2.1)

All the effects of the forces exerted on the electron by the crystal are included in the effective mass. For an electron in free space, \( E = \frac{\hbar^2 k^2}{2m} \), and Eq. (2.1) gives \( m^* = m \), where \( m \) is the mass of a free electron. Examining Fig. 2.1, one might notice that the deviation of the band structure away from the free particle dispersion curve increases the curvature of the band structure and therefore the quantity \( d^2E/dk^2 \) near the band minima and maxima. Therefore, in most cases, the effective mass of an electron in a semiconductor, as defined in Eq. (2.1), is smaller than that of a free electron.

Once an electron is excited into the conduction band, a vacancy is left in the valence band. This vacancy, known as a \textit{hole}, has effective positive charge. The dispersion relation for holes is much more complicated than that for electrons. The bulk hole bands are described in the Kane Model (Kane 1957) by basis functions with angular momentum \( J=3/2 \) symmetry, and therefore have 4-fold degeneracy at \( k=0 \). A typical hole band is shown in Fig. 2.2, with the heavy hole (hh) subband corresponding to \( J = 3/2, J_z = \pm 3/2 \), the light hole (lh) subband corresponding to \( J = 3/2, J_z = \pm 1/2 \), and the split off band corresponding to \( J = 1/2 \). The movement of holes can also be treated with the effective mass approximation, and the effective mass can be obtained from calculated valence band structures via Eq. 2.1.
§ 2.2 Electronic and linear optical properties of quantum wells

A semiconductor quantum well is a double heterostructure consisting of a thin layer of well material $W$ sandwiched between two thick layers of barrier material $B$. Material $B$ has a larger bandgap than that of material $W$, and the conduction and valence band offset are such that $W$ is a potential minimum for both electrons and holes, as shown in Fig. 2.2. A schematic diagram of conduction and valence band structures for a bulk direct gap semiconductor.
Fig. 2.3. Schematic diagrams of quantum wells. (a) A single quantum well. The upper part of the figure shows the material structure of a quantum well sample: a layer of well material embedded between two layers of barrier material. The lower part shows the band structure along the material growth direction, i.e., the z-direction. The conduction and valence band offsets at B-W interfaces are such that W is a potential minimum for both electrons and holes. (b) Shows a structure that is either a multiple quantum well or a superlattice, depending on the widths of the barriers and wells.

In Fig. 2.3a. For the double heterostructure to qualify as a quantum well, it is necessary that the W layer should be thin enough that the quantum size effect is significant, which...
separated values, each corresponding to a eigenenergy level. It is also necessary that the B layer should be thicker than the penetration depth of the confined particles so that the wavefunctions of particles in neighboring wells do not interfere with each other. In a structure which has many wells and with barriers thin enough that there is strong interference between particles in neighboring wells (Fig. 2.3b), there are several mini-energy-bands instead of several eigenenergies. These kind of structures are a practical realization of Kronig-Penney model (Kronig and Penney 1930), and are called superlattices (SLs).

a) Confinement of electrons in quantum wells

The energy level of electrons confined in a QW can be easily calculated in the envelope function approximation (Bastard 1981, 1982, Bastard et al. 1986). The envelope function scheme asserts that the wave function of electrons confined in a layered hetero structure takes approximately the form

$$\psi = \sum_{W,B} e^{i k_\perp z} u_{ck}^W (r) \varphi_n (z),$$

(2.3)

where $z$ is the growth direction, $k_\perp$ is the transverse electron wave vector, $u_{ck}(r)$ is the Bloch wave function in the W or B material, which has the periodicity of the host crystal, and $\varphi_n(z)$ is the envelope wavefunction. The advantages of using the envelope function formalism are that a quantum well potential, which is considered to be slowly varying compared to the scale of the lattice constant, affects only the envelope function, $\varphi_n(z)$, and that the effects of the periodic potential appear only through the effective mass. The envelope function can be determined to a good approximation by the Schrödinger-like equation.
\[
\left[ -\frac{\hbar^2}{2m^*(z)} \frac{d^2}{dz^2} + V_c(z) \right] \varphi_n(z) = \epsilon_n \varphi_n(z), \quad (2.4)
\]

where \( m^*(z) \) is the effective mass of the electron in the W or B material, \( V_c(z) \) is the energy level of the bottom of the conduction bands, and \( \epsilon_n \) is the eigenenergy of the confined electrons. Assuming that the band edge shifts abruptly at the interfaces and that the shapes of the band edges of both materials are not modified by the hetero junction, Eq. (2.4) is basically the particle in a finite square potential well problem. One of the boundary conditions for this classic example in quantum mechanics text books, which requires that the first derivative of the wavefunction be continuous between the well and the barrier, is inappropriate here because the effective mass of the electron is not necessarily the same in W and B. In order to conserve particle current, it is necessary to require continuity of \( [1/m^*(z)][\partial \varphi_n(z)/\partial z] \) at the interfaces.

Noting that the problem is symmetrical about the center of the well, which is now taken as the origin of the coordinate system, as shown in Fig. 2.4a, the solution wave functions of Eq. (2.4) can only be even or odd. Therefore, they can be written as

\[
\varphi_n(z) = A \cos k_s z, \quad \text{for } -d/2 < z < d/2
\]
\[
= B \exp[-\kappa(z-d/2)], \quad \text{for } z > d/2
\]
\[
= B \exp[\kappa(z+d/2)], \quad \text{for } z < -d/2 \quad (2.5)
\]

or

\[
\varphi_n(z) = A \sin k_s z, \quad \text{for } -d/2 < z < d/2
\]
\[
= B \exp[-\kappa(z-d/2)], \quad \text{for } z > d/2
\]
\[
= B \exp[\kappa(z+d/2)], \quad \text{for } z < -d/2 \quad (2.6)
\]

where

\[
k_s = \sqrt{\frac{2m^*_n(V_0 + \epsilon_n)}{\hbar^2}}, \quad \kappa = \sqrt{\frac{2m^*_n \epsilon_n}{\hbar^2}}, \quad -V_0 < \epsilon_n < 0 \quad , \quad (2.7)
\]
and \(A\) and \(B\) are constants. Note that the subscript in \(k_z\) was used to emphasize that \(k_z\) is only the \(z\)-component of the wavevector \(k\) for the electron confined in the QW. For the solution in Eq. (2.5), the continuity conditions at \(z = \pm d/2\) yield

\[A \cos(k_z d/2) = B,\]

and

\[(k_z / m_A^*) \sin(k_z d / 2) = \kappa / m_B^*.\]

Therefore

\[(k_z / m_A^*) \tan(k_z d / 2) = \kappa / m_B^*.\]  \hspace{1cm} (2.8)

Similarly, Eq. (2.4) yields

\[(k_z / m_A^*) \cot(k_z d / 2) = -\kappa / m_B^*.\]  \hspace{1cm} (2.9)

Eqs. (2.8) and (2.9) can be solved numerically to obtain the eigenenergies. A typical result is shown in Fig. 2.4a: a series of separated eigenenergy levels. The wavefunction corresponding to each eigenenergy is also shown in Fig 2.4a: the wavefunctions are generally sinusoidal inside the well and decay exponentially into the barriers. The band structure in the plane of the QW is a set of discrete parabolic curves as shown in Fig. 2.4b, each of which corresponds to a particular eigenenergy.

Confinement of holes in QWs can be treated in the same manner. In this case however, the heavy hole and the light hole have different sets of eigenenergies, because of the difference in their effective masses. Also, the band structure in the plane of the QW is nonparabolic, especially for large momentum, due to the interaction between different valence bands. A more detailed description of the valence band structure can be found in §2.4.
Fig. 2.4. (a) Conduction band of a semiconductor quantum well along the growth direction. The thickness of the well material is $d$. The eigenenergies of the electrons confined in the potential well are indicated by dashed lines. The corresponding wave functions are drawn near each eigenenergy level. (b) The dispersion relation of the electrons in the plane of the quantum well, i.e. perpendicular to the growth direction. Each parabolic curve corresponds to an eigenenergy. (c) The step-function like conduction band density of state of the quantum well. The onset of each step corresponds to an eigenenergy. The dashed line gives the 3D density of states of a bulk material of thickness $d$. 
b) Density of states for electrons and holes confined in QWs

Semiconductor QWs are known as quasi-2D systems because of their finite well widths and barrier heights. In a real 2D system, the wave vector of a particle has only two components, e.g., \( \mathbf{k} = k_x \hat{x} + k_y \hat{y} \). However, for an electron confined in a QW, the \( z \)-component of the wave vector can have one of the several discrete values, corresponding to the eigenenergies determined by Eq. (2.4). The wave vector corresponding to an eigenenergy \( E_n \) is therefore given by \( \mathbf{k}_n = k_x \hat{x} + k_y \hat{y} + k_z \hat{z} \). In both real and quasi-2D systems, electrons can move "freely" in two directions under the effective mass approximation (EMA), therefore, the wave vector component in these two directions can have all the allowed values determined by the size of the system.

To calculate density of states (DOS), which is the number density of allowed states per unit energy, of electrons in 2D systems, we start by considering the area of the \( k \)-surface occupied by any allowed state of a Bloch electron* (Bloch, 1928) in a 2D crystal of area \( A \) with rectangular unit cells defined by lattice vectors \( \mathbf{a}_x \) and \( \mathbf{a}_y \). There are a total of \( N = N_x N_y \) unit cells in the crystal. The boundaries in direction \( \mathbf{a}_i \) are at 0 and \( N_i \mathbf{a}_i \), where \( i = x, y \). A cyclic boundary condition \( \psi(\mathbf{r}) = \psi(\mathbf{r} + N_i \mathbf{a}_i) \) should be imposed to avoid standing electron waves (Born and Von Kárman 1912). According to Bloch's theorem, we have

\[
\psi_k(\mathbf{r} + N_i \mathbf{a}_i) = \exp(iN_i \mathbf{k} \cdot \mathbf{a}_i) \psi_k(\mathbf{r})
\]

or

\[
\psi_k(\mathbf{r} + \mathbf{R}) = \psi_k(\mathbf{r})
\]

* The term Bloch electron is used to refer to an electron that obeys the one-electron Schrödinger equation in a periodic potential. Bloch found that such electrons have wavefunctions in the form of a plane wave multiplied by a function that has the periodicity of the direct lattice. That is \( \psi_k(\mathbf{r}) = \exp(i\mathbf{k} \cdot \mathbf{r}) \psi_k(\mathbf{r}) \), where \( \mathbf{k} \) is a wavevector and \( \psi_k(\mathbf{r}) = \psi_k(\mathbf{r} + \mathbf{R}) \) for all direct lattice vectors \( \mathbf{R} \). This result is known as Bloch's theorem (Wolfe et al. 1989).
\[ \exp(iN_i k \cdot a_i) = 1. \]  \hspace{1cm} (2.10b)

The solutions of Eq. (2.10) are

\[ k_i = \frac{2\pi m_i}{N_i a_i}, \quad m_i = 0,1,2,3... \]  \hspace{1cm} (2.11)

which are the allowed \( k \) values in the crystal. The area of \( k \)-space occupied by any allowed state is

\[ \Omega_k = [k_x(m_x) - k_x(m_x - 1)][k_y(m_y) - k_y(m_y - 1)] = \frac{(2\pi)^2}{A}, \]  \hspace{1cm} (2.12)

note that \( A = N_x N_y a_x a_y \). Thus, since each allowed state can be occupied by two electrons with different spins, the total number of states per unit area between energies \( E \) and \( E + dE \) is

\[ N_{2D}(E)dE = \frac{1}{A} \frac{2}{(2\pi)^2} \frac{2\pi k(E)dk}{A} \]  \hspace{1cm} (2.13)

where \( N_{2D}(E) \) is defined as the 2D density of states. Assuming a parabolic energy band, given by

\[ E(k) = \frac{\hbar^2 k^2}{2m^*}, \]  \hspace{1cm} (2.14)

one has

\[ dk = \left( \frac{m^*}{\hbar^2} \right) dE. \]  \hspace{1cm} (2.15)

Substituting Eq. (2.15) into Eq. (2.13) gives

\[ N_{2D}(E) = \frac{m^*}{\pi \hbar^2}. \]  \hspace{1cm} (2.16)

Eq. (2.16) indicates that the 2D DOS is not a function of energy, and its value is only determined by the effective mass.
The density of states of an electron confined in a QW can be calculated in a very similar manner to the DOS in a real 2D system. Instead of \( k_z = 0 \) as in 2D systems, the \( z \)-component of the wave vector in a QW can have one of several discrete values. For each confined state, \( k_{zn} \), the DOS can be calculated with the 2D crystal model. The total DOS for an electron in a QW is therefore given by

\[
N_{QW}(E) = \sum_n N_{2D}(E) = \sum_n \frac{m^*}{\pi \hbar^2},
\]

where the summation is over all confined states with eigenenergy \( E_n < E \), for which \( k_{n\perp} \) are defined at energy \( E \) (see Fig. 2.4b). The total DOS at a given energy is then equal to \( m^*/\pi \hbar^2 \) times the number \( n \) of different confined energy levels below that energy. It can be seen from Fig. 2.4c that the QW DOS shows discontinuities at each \( E_n \). The quasi-2D DOS of QW is often referred to as the 2D DOS for simplicity, since QWs are the closest practical realization of 2D systems. Comparing with the 3D DOS given by (see e.g. Wolfe et al. 1989)

\[
N_{3D}(E) = \frac{4\pi}{\hbar^3} (2m^*)^{\nu_2} (E - E_c)^{\nu_2},
\]

the fundamental difference between the 2D and the 3D DOS is that the DOS is \textit{finite} even at the bottom of the 2D level, whereas it tends towards zero in the 3D system. This means that in a 2D system all dynamic phenomena, such as optical absorption and gain, remain finite even at low kinetic energies and temperatures.

The 2D DOS for holes can be treated in same manner. In this case the DOS is not exactly a step function because of the nonparabolic nature of the confined valence band structure. Nevertheless, a step function is still a good approximation for the hole DOS, especially in the vicinity of \( k=0 \).
The optical absorption coefficient of a semiconductor is proportional to the product of the conduction and valence band DOS (see e.g. Casey et al. 1978), therefore, for a 2D system, one should expect a step-like spectrum for optical absorption associated with creation of free electrons and holes.

c) Excitons and optical absorption in quantum wells

In a 3D semiconductor system it is well known that in addition to the free electron and hole states, there are some bound electron and hole states, which are due to the Coulomb attraction between an electron and a hole. These bound states are called exciton states (see e.g. Knox, R. S. 1963). The bound electron-hole pairs are qualitatively similar to the Hydrogen atom. The lowest bound state is one effective Rydberg (Ry*) below the continuum level and in this state, the electron and hole are separated by the effective Bohr radius \(a_B^*\). The lowest bound state is the 1s state, and its wavefunction is described by

\[
\phi_{1s}^{\text{v}}(r) = \frac{1}{\sqrt{\pi a_B^*}} \exp\left(-\frac{r}{a_B^*}\right)
\]

in \(r\) space, and

\[
\phi_{1s}^{\text{v}}(k) = \frac{8\sqrt{\pi a_B^*}}{[1+(a_B^*k)^2]^{3/2}}
\]

in \(k\) space. In the above, \(r\) is the distance between the electron and hole, and \(a_B^*\), the effective Bohr radius, is given by

\[
a_B^* = \frac{4\pi\varepsilon_0\hbar^2}{\mu e^2} = \varepsilon_r m_e a_B^*,
\]

where \(\varepsilon_r\) is the relative permittivity of the semiconductor, \(m_0\) is the free electron mass, \(\mu\)
is the reduced effective mass of electron and hole \((1/\mu = 1/m_e^* + 1/m_h^*)\), and \(a_B (=0.529\text{Å})\) is the Bohr radius. The bound state energies are

\[
E^{3D}_n = E_g - \frac{Ry^*}{n^2},
\]

(2.22)

with

\[
Ry^* = \frac{2\mu e^4}{\hbar^2 (8\pi\varepsilon)^2} = \frac{\mu}{m_e \varepsilon_r^2} Ry,
\]

(2.23)

where \(Ry (=13.6\text{eV})\) is the Rydberg constant.

Using \(\mu = 0.059\ m_o\ \varepsilon_r = 12.91\) for GaAs, one obtains \(a_B^* = 115\text{Å}\) and \(Ry^* = 4.8\) meV. For InAs, \(\mu = 0.027m_o\ \varepsilon_r = 15.15\), one obtains \(a_B^* = 297\text{Å}\) and \(Ry^* = 1.5\) meV. Therefore, in 3D systems, excitons are very weakly bound and consequently their effects cannot usually be observed at room temperature. However, the Bohr diameter \(2a_B^*\) is at least twice as large as the typical quantum well thickness of \(-100\text{Å}\), and therefore one should expect that the wavefunction and energy level of excitons confined in a QW will be substantially modified.

For a pure 2D situation, the electron-hole interaction can also be solved exactly (Shinada et. al., 1966). The wavefunction of the 1s state is

\[
\phi^{2D}_{1s}(r) = \left(\frac{2}{\pi}\right)^{1/2} \frac{2}{a_B^*} \exp\left(-\frac{2r}{a_B^*}\right)
\]

(2.24)
in \(r\)-space, and

\[
\phi^{2D}_{1s}(k) = \frac{(2\pi)^{1/2}(a_B^*/2)}{\left[1+(a_B^*k/2)^2\right]^{3/2}}
\]

(2.25)
in \(k\)-space. The bound state energies are

\[
E^{2D}_n = E_g - \frac{Ry^*}{(n-1/2)^2},
\]

(2.26)
Note that in the 2D system, the first (1s) exciton is more tightly bound \((E_{1s}^{2D} = E_g - 4Ry^*)\) than in 3D \((E_{1s}^{3D} = E_g - Ry^*)\), and also that in the 2D system the charge density maximum occurs at \(a_{2D} = a_B^* / 4\) whereas in 3D it occurs at \(a_{3D} = a_B^*\). Real QWs have finite thickness and a finite band offset, so the exciton binding energies fall between the 2D and 3D limits. The QW is therefore known as a quasi 2D system.

Theoretical determinations of the exciton binding energy in QWs have been performed by many people using variational calculations (Schmitt-Rink et al. 1989 and references therein), and will not be discussed in detail here. Nevertheless a significant enhancement of the QW exciton binding energy is predicted, with a maximum in the range \(3Ry^* > E_{\text{binding}} > 2Ry^*\) for \(1/4 < d_w/a_B^* < 1\). A typical result of the calculation of the exciton binding energy of an exciton in a QW is shown in Fig. 2.5. The increased exciton binding energy has a profound influence on QW properties: it results in the fact that the optical properties of GaAs-based (Chemla and Miller 1985) and InGaAs-based (Weiner et al. 1985) QWs are dominated by exciton effects even at room temperature.

The linear optical susceptibility, \(\chi\), and hence the absorption coefficient associated with the excitonic resonance, which is the transition between the bound and the exciton states, is much larger than that associated with the transition between bound and free electron-hole states, because of the increased electron and hole wavefunction overlap and the consequently increased oscillator strength for the exciton states. This is particularly true for QWs, because of the reduced effective Bohr radius in these quasi 2D systems. The linear optical susceptibility associated with the excitonic resonance can be obtained from the well known Elliott formula (Elliott 1957, Shinada and Sugano 1966, Haug and Schmitt-Rink 1984)

\[
\chi(\omega) = 2e^2|e_o|^2 \sum_n \frac{\left| \phi_n(r = 0) \right|^2}{E_n - \hbar \omega - i\gamma_n}, \tag{2.27}
\]
Fig. 2.5. Variation of the exciton binding energy in a GaAs/Al$_x$Ga$_{1-x}$As quantum well, as a function of quantum well thickness. The solid lines correspond to the heavy-hole exciton and the dashed lines to the light-hole exciton. Curves for the three well depths are shown: an infinite well; $\Delta E_g \approx 190$ meV and $\Delta E_g \approx 380$ meV corresponding to $x \approx 0.15$ and 0.3 respectively. (After Greene et al. 1984).

where the index $n$ runs through all exciton states (discrete and continuous), $E_n$ and $\gamma$ are the energy and width of each state, and $r_{cv}$ is the dipole matrix element, and $|r_{cv}|^2|\phi_n(r=0)|^2$ is proportional to the oscillator strength. The absorption coefficient can
be obtained from the imaginary part of $\chi$ through $\alpha(\omega) = \frac{4\pi\omega}{|cn(\omega)|} \text{Im} \chi$, where $n(\omega)$ is the refractive index. Eq. (2.27) implies that the electron-hole correlation produces strong peaks in the absorption spectrum at the energy of the $s$ exciton states with zero angular momentum, for which $|\phi_n(r = 0)|^2 \neq 0$. The usual interpretation of this result is that the absorption is enhanced by the electron-hole correlation in proportion to the increased probability of finding the electron and hole at the same site.

Combining the discussions which have been made so far, one can infer that the absorption spectrum of a QW structure consists of a series of steps corresponding to transitions from the energy levels in the valence band to the energy levels in the conduction band, with an excitonic absorption peak at the onset of each step. Such an absorption spectrum was first observed by Dingle et al. (1974).

§ 2.3 Optical nonlinearities induced by photo-generated free-carriers

Nonlinear optical phenomena can be grouped into two broad categories (see e.g. Butcher and Cotter 1990). In the first category are those phenomena in which the optical frequencies are far from the resonances of the medium (known as 'nonresonant' or 'passive' nonlinearities), and the exchange of energy between the field and the medium occurs via virtual excitations. In this case, the coherent laser field, $E$, induces a coherent polarization, $P$, in the medium, and the nonlinear polarization can couple various optical fields, which exchange photons via the material, but no net power is deposited in the medium. Examples of processes in this category are second harmonic generation, optical parametric oscillation and the optical Stark effect. In the second category are those phenomena which are dissipative (also known as 'active' or 'resonant' nonlinearities): in this case the flow of energy from and to the optical field is taken up in real energy
transitions of the medium by the processes of absorption and emission. There are two
types of real energy transition. The first type is transitions between bound electronic or
vibrational states of the medium; one example of nonlinear phenomena associated with this
type of transition is stimulated Raman scattering. The second type involves free-carrier
states, such as the creation and recombination of free-electron-hole pairs in
semiconductors. One example of a phenomena associated with either the first type or the
second type of real energy transition is the nonlinear refractive index which arises from
saturated absorption. Other examples related to free carriers are 'charge-transport-
assisted' optical nonlinearities, such as the photorefractive effect (Smirl et al. 1988) and
the 'self-electro-optic' effect (Miller et al. 1988); these occur when free carriers which are
generated by the optical field subsequently migrate and separate, resulting in the setting up
of an internal space-charge field in the material.

An important difference between these various nonlinear mechanisms is the wildly
differing response speeds that are obtained. Here the term response time represents the
longer of the 'turn on' time or the recovery time for the nonlinear optical effect. It is the
response time that limits the operational speed of devices based on the corresponding
nonlinear optical effect. In the case of nonresonant nonlinearities which involve virtual
electronic transitions, the speed of response of the medium is virtually instantaneous.
When resonant excitation is used to increase the magnitude of the nonlinearity, real
transitions occur and the important parameter is then the excited state lifetime, which
typically falls in the picosecond to nanosecond range for solid state materials. If the
displacement of free-carriers is involved, then the response time is tailorable to some
degree. Nevertheless it is usually much longer than the response time for nonresonant
nonlinearity.
In the remainder of this section, the discussion is concentrated on the nonlinear refractive index that arises from absorption saturation due to the photo-generated electron-hole plasma in semiconductor QWs, since this is the kind of optical nonlinearity to be studied in this work.

In terms of optical nonlinearities, the advantage of QWs over bulk materials arises from the fact that in both materials the modulation of the optical coefficients is dominated by phase-space filling (see discussion later on in this section) due to the photo-generated e-h pairs. Although the relative change of the oscillator strength per photo-generated electron-hole pair hardly changes as one goes from bulk GaAs to GaAs/AlGaAs QWs (Schmitt-Rink et al. 1987, Park et al. 1988), the absolute change of the oscillator strength is proportional to the linear oscillator strength, which is significantly larger in QWs because of the excitonic effects discussed in last section. The response time of the absorption saturation and nonlinear refractive index change induced by the photo-generated free electron-hole plasma is determined by the carrier lifetime, which is typically ~1 ns for III-V semiconductor QWs. This moderate lifetime allows accumulation of photo-excited carriers, and therefore optoelectronic devices fabricated with these materials can have a relatively low operational light intensity, as well as the reasonably high operational frequency of up to ~1 GHz. The impressive magnitude of the nonlinear optical effects in high quality QWs, due to the enhanced oscillator strength and long carrier lifetime, was demonstrated by the observation of degenerate four-wave mixing using a pump intensity of ~17 Wcm⁻² from a commercial CW laser diode (Miller et al. 1983b).

The creation of a photo-generated electron-hole plasma in QWs leads to the nonlinear modification of the optical properties of the QW through phase-space filling (PSF), band-gap renormalization (BGR) (Schmitt-Rink and Ell 1985, Schmitt-Rink et al.
1985), and collision-induced absorption broadening (Feng and Spector 1987). The effect of photo-generated carriers on the exciton resonance was estimated by Schmitt-Rink et al. (1985). Starting from the Elliott formula given in Eq. (2.27), the linear susceptibility, $\chi(\omega)$, is given by

$$\chi(\omega) = 2e^2 |r_{\alpha\sigma}|^2 \sum_n \frac{\left| \phi_n(r=0) \right|^2}{E_n - \hbar\omega - i\gamma_n} = \sum_n \frac{f_n}{E_n - \hbar\omega - i\gamma_n},$$

where $f_n \propto |r_{\alpha\sigma}|^2 |\phi_n(r=0)|^2$ is the oscillator strength of the transition. For frequencies in the vicinity of the 1s-exciton it is legitimate to retain only the resonant term, which yields

$$\chi \approx \frac{f_{1s}}{E_{1s} - \hbar\omega - i\gamma_{1s}}.$$

The nonlinearity arises when one or more of the three quantities $f_{1s}$, $E_{1s}$, or $\gamma_{1s}$ is changed by the optical excitation. A change of the oscillator strength $f_{1s}$ may be caused by phase-space filling and bandgap renormalization. As the photo-generated free-carrier density increases, the band gap for free electron-hole pairs becomes renormalized and moves to lower energy, although almost no change in the exciton energy is observed (Chemla et al. 1984, Knox et al. 1985). The exciton binding energy, as measured from the exciton state to the renormalized free electron state, therefore decreases and the real space volume of the exciton increases. The resonance loses oscillator strength, both because of the occupation of states from which the exciton is constructed (Pauli exclusion principle) and because of the loss of electron-hole correlation. The screening of the Coulomb interaction due to the presence of an e-h plasma also causes exciton dissociation, and hence a decrease of the exciton oscillator strength. However, the long range electrostatic screening is much weaker in 2D than in 3D (Ando et al. 1982), such that in 2D the effects of screening are weak compared with the effects of the exclusion principle, as demonstrated by Knox et al. (1986). The exciton resonance linewidth $\gamma_{1s}$ can increase as a
result of collisions between the photo-generated carriers, and the amount of broadening is
determined by the carrier density. The excitonic resonance energy, $E_{1s}$, is barely affected
by the presence of free electron-hole pairs, as mentioned earlier. The constancy of exciton
energy simply reflects the charge neutrality of the exciton, i.e., the fact that the effects of
the other free electron-hole pairs on the electron of a bound pair are strongly compensated
for by their effects on the companion hole (Schmitt-Rink et al. 1985). However, at low
temperature equilibrium, when a larger proportion of photo-generated electron-hole pairs
are in exciton states, a blue shift of the exciton resonance peak does occur due to the
repulsion between excitons (Schmitt-Rink et al. 1985). A blue shift of excitons can also
happen in structures in which an electric field is applied across the QWs, such as SEEDs
(Miller et al. 1988) and hetero nipis (Kost et al. 1988, McCallum et al. 1992), due to the
screening of the applied field by spatially separated free electrons and holes. This effect is
beyond the scope of this thesis and will not be discussed in detail here.

The electron-hole plasma can be optically generated either directly, by exciting
above the gap (Shank et al. 1983, Chemla et al. 1984, Park et al. 1988), or indirectly
following ionization of excitons created by resonant absorption (Miller et al. 1982, Weiner
et al. 1986, Tai et al. 1987). At room temperature, excitons in III-V semiconductor QWs
created via resonant excitation are unstable and will be ionized in a fraction of a
picosecond (Chemla et al. 1984). The free electrons and holes which result from the
exciton dissociation will be further thermalized to reach the lattice temperature in a few
picoseconds (Knox et al. 1985). The relative change of the exciton oscillator strength due
to the existence of one photo-generated e-h pair is

$$\left( \frac{\delta f_{1s}}{f_{1s}} \right)_{PSE} = -\sum_k \left[ f_{t_e}(k) + f_{t_h}(k) \right] \frac{\phi^{2P}(k)}{\phi^{1P}_{1s}(r = 0)}, \quad (2.30)$$
where \( f_e \) and \( f_h \) are the distribution functions of the photo-generated electrons and holes. Note that \( \phi_{1s}^{2D}(r = 0) \propto \sum_k \phi_{1s}^{2D}(k) e^{i\mathbf{k} \cdot \mathbf{r}_{\text{eq}}} \). According to the exclusion principle, transitions are not allowed if either the corresponding electron state or hole state is occupied. The summation \( \sum_k [f_e(k) + f_h(k)] \phi_{1s}^{2D}(k) \) gives the change of \( \phi_{1s}^{2D}(r = 0) \) due to the phase-space filling by one electron-hole pair.

The resonant generation of excitons yields a distribution of electrons and holes given by
\[
f_e(k) = f_h(k) = \frac{1}{2} |\phi_{1s}^{2D}(k)|^2.
\]

The physical meaning of this expression is that, since an exciton is built up from a linear combination of single-particle states distributed according to \( \phi_{1s}^{2D}(k) \), the creation of one exciton corresponds to an occupation probability in the phase space, \( |\phi_{1s}^{2D}(k)|^2 \), which is equally shared between spin-up and spin-down states.

For the free electron-hole plasma in equilibrium, and in the dilute limit, the carrier distributions are given by the Boltzmann distributions
\[
f_{e,h} = \frac{\hbar^2 \pi}{8 m_e^* k_B T} e^{-\frac{\hbar^2 k^2}{2 m_e^* k_B T}}, \tag{2.32}
\]
where \( m_e^* \) is the electron or hole effective mass, and \( T \) is the temperature.

Substituting Eqs. (2.25) and (2.31) into Eq. (2.30), one can obtain
\[
\left( \frac{\delta f_{1s}}{f_{1s}} \right)_{\text{PSF,ex}} = \frac{32}{7} \pi a_{2D}^2,
\]
which is the relative change of oscillator strength due to the phase-space filling by excitons, where \( a_{2D} = a_e^* / 4 \) is the 2D effective Bohr radius. Substituting Eqs. (2.25) and (2.32) into Eq. (2.30) one can get, in the extreme case where \( k_b T / E_{1s} >> 1 \)
which is the relative change of oscillator strength due to phase-space filling by a hot electron-hole plasma, where $E_{\text{ls}}$ is the exciton binding energy. Therefore, by comparing Eqs. (2.33) and (2.34) it can be seen that at room temperature, assuming a binding energy of 9 meV, the relative change of oscillator strength due to the phase-space filling by excitons is larger than that due to the hot electron-hole plasma. This point was demonstrated experimentally by Knox et al. (1985).

In summary, large optical nonlinearities are expected to arise from phase-space filling by room-temperature photo-excited free electron-hole plasmas in III-V semiconductor QWs, mainly due to the excitonic nature of the optical properties in these quasi 2D systems. These resonant nonlinearities persist as long as the lifetime of the photo-excited carriers. The nonlinear optical effects of phase-space filling by excitons are even larger than those associated with room-temperature electron-hole plasmas. However, at room-temperature, excitons in QWs, as well as the large nonlinearities associated with the occupancy of exciton states, will not last very long (typically a fraction of a picosecond).

§ 2.4 Valence band structures of quantum wells and the effect of strain on valence band structures

a) Valence band structures in quantum wells

For quantum wells grown on [001] oriented III-V substrates, the transition from a 3D to a 2D system lowers the symmetry of the material from $T_d$ to $D_{2d}$, which has little
effect on the in-plane band structure of electrons because of their plane wave nature, but alters the band structure of holes drastically.

The bulk hole bands are described in the Kane model by basis functions with angular momentum $J=3/2$ symmetry. The dispersion relation for holes near $k=0$ can be described by the Luttinger Hamiltonian (Luttinger 1956):

$$H = \frac{\hbar^2}{2m_0} \left( (\gamma_1 + \frac{5}{2}\gamma_2)k^2 - 2\gamma_2 (k_x^2 J_x^2 + k_y^2 J_y^2 + k_z^2 J_z^2) \right)$$

$$-4\gamma_3 [(k_x k_y + k_y k_z)(J_x J_y + J_y J_z)] + \ldots \right), \quad (2.35)$$

where $\gamma_{1,2,3}$ are the Luttinger parameters and $m_0$ is the free electron mass. For bulk material, assuming the holes propagate in the z direction, it is easy to get from Eq. (2.35) the effective masses $m_{sh}^* = m_0 (\gamma_1 - 2\gamma_2)^{-1}$ for $J_z = \pm 3/2$, and $m_{lh}^* = m_0 (\gamma_1 + 2\gamma_2)^{-1}$ for $J_z = \pm 1/2$. In the QW, the confinement lifts the degeneracy of $hh$ and $lh$ bands, because of the difference in effective mass. Furthermore, it can be found by substituting $k_y = k_z = 0$ and $k_x \neq 0$ into Eq. (2.35) that the $hh$ and $lh$ have a parabolic dispersion with a reversal of curvature in the plane of the QW, if one can neglect the effect of confinement on the dispersion and assume that the $hh$ and $lh$ do not interact. The corresponding effective masses are $m_{sx,sh}^* = m_0 / (\gamma_1 + \gamma_2)$ for $J_z = \pm 3/2$, and $m_{xy,lh}^* = m_0 / (\gamma_1 - \gamma_2)$ for $J_z = \pm 1/2$; that is, in the plane of the QW, the effective mass of the $(J_z = \pm 3/2)$ band is lighter than that of the $(J_z = \pm 1/2)$ band. This is a somewhat over simplified approach to a very complicated problem, but it gives a clear qualitative picture of band structures in a 2D system. More accurate results have been obtained by treating the confinement and the interaction between valence bands on an equal footing. This calculation is complicated and can only be carried out numerically (Ekenberg and Altarelli 1984, Broido and Sham 1985, 1986, Bastard and Brum 1986). A typical result is shown in Fig. 2.6; the solid lines represent the band structures and are clearly nonparabolic. The dashed lines represent the
dispersions when band mixing is neglected. Notice that both sets of curves are very similar near $k = 0$, which means that an estimate of effective mass made using Eq. (2.35) is valid at least at zone center.

**Fig 2.6.** In-plane valence band dispersion against wave-vector for two GaAs/Al$_{0.3}$Ga$_{0.7}$As quantum wells ((a) $d = 100$ Å, (b) $d = 150$ Å). The dashed lines show the dispersion when band mixing is neglected. (After Bastard and Brum 1986).

b) Characteristics of strained QWs fabricated from dissimilar materials

It has been known for many years that lattice-mismatched heterostructures can be grown without misfit dislocation if the layers are sufficiently thin (Frank and Van de
Merwe 1949), so that the mismatch is accommodated entirely by a uniform lattice strain. However, the common wisdom was that all strain in semiconductor devices was bad until 1982, when Osbourn proposed that the strain associated with the heteroepitaxy of dissimilar materials may itself offer a new functionality, the advantages of which might far outweigh the disadvantages of the presence of strain. The possibility of using lattice-mismatched materials to form heterostructures allows a new degree of freedom for the band gap which is useful in the design of semiconductor nanostructures. The strain in lattice mismatched heterostructures also modifies the valence band, which results in the lifting of the degeneracy at valence band center, and hence the further separation of heavy hole and light hole bands in quantum wells under biaxial compression (see e.g. O'Reilly 1988, Marzin et al. 1989, People and Jackson 1990 and Mailhiot and Smith 1990). A material system in which this is observed is InGaAs quantum wells grown on a GaAs substrate. Strained InGaAs QWs, as well as many other structures, hold promise for high mobility p-channel transistors (Drummond et al. 1986, Lee et al. 1987, Zipperian et al. 1988 and Daniels et al. 1988) and low threshold lasers (Adams 1986 and Yablonovitch and Kane 1986, 1988).

A schematic cross section of In$_x$Ga$_{1-x}$As grown on a [001] GaAs substrate is shown in Fig. 2.7. The lattice constant of free-standing In$_x$Ga$_{1-x}$As is 5.6533±0.405Å. For a sufficiently thin layer of InGaAs, all the strain is incorporated in the layer. The layer is under biaxial compression such that its in-plane lattice constant is equal to the substrate lattice constant. In response to the biaxial compression, the layer relaxes along the growth direction. The stability of strained layers was first studied by Frank and Van de Morwe (1949). They calculated the critical layer thickness below which the mismatched layer is expected to be uniformly strained instead of having misfit-generated defects. Since then, there have been a number of further calculations of the critical layer thickness (Matthews...
and Blakeslee 1974, 1975, 1976, People and Bean 1985 and People 1986). Most of these calculations differ from each other in the energy assumed to be stored in a strained layer, but the basic idea is the same: to find a point at which the energy stored in the strained layer exceeds the energy required to introduce a dislocation. The most systematic experimental study of the critical thickness of a In\(_x\)Ga\(_{1-x}\)As layer grown on a GaAs substrate was performed by Weng (1989), and the result is shown in Fig. 2.8.

![mismatched layer](image)

![strained layer](image)

Fig. 2.7. Schematic cross-section InGaAs and GaAs. (a) Free standing InGaAs and GaAs crystal. The lattice constant of InGaAs is larger than that of GaAs. (b) A thin InGaAs layer is grown on a GaAs substrate. The lattice mismatch is elastically accommodated in the InGaAs layer by a uniform biaxial compression.
Fig. 2.8. Surface morphology observed under a phase-contrast optical microscope and the corresponding PL peak strength for the InGaAs strained single QW samples plotted as a function of the strained layer thickness and the In content. Open squares indicate samples with smooth surfaces and strong PL features. Open circles indicate samples with cross-hatched surfaces and reasonably good PL peaks; triangular symbols represent samples with cross-hatched or rough surfaces and insignificant PL features; the inverted triangular symbol represents the sample with a rough surface and a noticeable PL peak. (After Weng 1989).
Fig. 2.9. (a) A schematic representation of the band structure of an unstrained direct-gap semiconductor. The light hole and heavy hole bands are degenerate at $k = 0$. (b) Under biaxial compression the mean band gap increases and the valence band degeneracy is lifted at $k = 0$. The highest band is heavy along the material growth direction, $z$, and comparatively light in directions perpendicular to the $z$-direction. (c) Under biaxial tension the mean band gap decreases, the valence band splitting is reversed so the highest valence band is now light along the $z$-direction and comparatively heavy in directions perpendicular to the $z$-direction. (After O'Reilly 1989)
The biaxial compression in, e.g., an InGaAs layer grown on a GaAs substrate, or biaxial tension in other material systems, alters the relative position of atoms in the crystal. That obviously causes the band gap of the material to change. Furthermore, the strain lowers the symmetry of the semiconductor, in case of the growth on [001] substrate, from $T_d$ to $D_{2d}$ for III-V compounds (Marzin and Gérard 1990). This symmetry change is the same as that experienced when going from a bulk material to a quantum well, and has the same effect: lifting the valence band degeneracy at $k=0$, and producing anisotropic band structures. The strain effects on band structures are shown in Fig. 2.9 (O'Reilly 1989).

Fig. 2.9a is a schematic representation of the band structure of an unstrained direct-gap semiconductor. The heavy-hole (hh, corresponding to $J=3/2$, $J_z=\pm 3/2$) and light-hole (lh, corresponding to $J=3/2$, $J_z=\pm 1/2$) bands are degenerate at the $k=0$ and the spin-split-off (so, corresponding to $J=1/2$, $J_z=\pm 1/2$) band lies $\Delta$ lower in energy. The bottom of the conduction band (cb) is separated by the band-gap energy $E_g$ from the top of the valence bands. Fig. 2.9b shows the band structures under biaxial compression. The hydrostatic component of the compression increases the mean band gap, while the axial component splits the degeneracy of the valence band maximum and introduces an anisotropic valence band structure, with the highest band corresponding to $J=3/2$, $J_z=\pm 3/2$, which, as in the unstrained 2D system, is heavy-hole like along the $k_z$ direction and is light-hole like in the plane of biaxial compression. Fig. 2.9c shows schematically the band structure of a direct-gap semiconductor under biaxial tension. In this case, the mean band-gap reduces and the valence splitting is reversed so the highest band is now light-hole-like along $k_z$ and heavy-hole-like in the plane of biaxial tension. Quantum wells corresponding to the band structures in Fig. 2.9 are shown in Fig. 2.10. The ($J=3/2$, $J_z=\pm 3/2$) "heavy"-hole and the ($J=3/2$, $J_z=\pm 1/2$) "light"-hole effectively see different quantum wells in heterostructures under biaxial compression. Here we use the terms *heavy* and *light* in
Fig. 2.10. (a) An unstrained quantum well structure with confinement energies indicated for electrons (——), heavy holes (---------), and light holes (------------). (b) When the quantum well layers is under biaxial compression, the heavy-hole well gets deeper by comparison with the light-hole well, increasing the hh1-lh1 energy splitting. (c) For a quantum well under biaxial tension, the light-hole well deepens and the highest confined valence state can be lh1. (After O'Reilly 1989).
quotation mark to emphasize that they designate spin states rather than effective masses. In case of biaxial compression, the "heavy"-hole band is above the "light"-hole band, and the highest confined "heavy"-hole level and "light"-hole level in QWs are further separated due to the difference in effective mass. In case of biaxial tension, the "light"-hole confined level could be above, equal to or below the "heavy"-hole confined level, depending only on the amount of tension and the effective masses.

As stated at the beginning of the last paragraph, the anisotropy of the valence band structures is a result of the reduced symmetry in a biaxially strained semiconductor, similar to the case of an unstrained 2D system. Although the presence of strain, e.g., the biaxial compression in InGaAs/GaAs QWs, further reduces the in-plane effective mass of the J=3/2, J_z=±3/2 valence band from the unstrained 2D confinement case, the in-plane "heavy" hole effective mass is not closely correlated to the amount of strain applied to the material (Schirber et al. 1985; People et al. 1984). Nevertheless, the amount of valence band splitting is related to the strain, and the advantages associated with the strain induced splitting of "heavy"-hole and "light"-hole bands are closely correlated to the amount of strain in the material.

The separation of "heavy"-hole and "light"-hole bands, in a biaxially compressed system, e.g., InGaAs grown on GaAs, makes it possible to preferentially populate the "heavy"-hole band, which has a lighter in-plane effective mass, resulting in a high-mobility p-type material. An in-plane hole effective mass of m* = 0.14m_0 has been observed by Schirber et al. (1985) in In_{0.2}Ga_{0.8}As/GaAs strained quantum wells.

Strain can also be employed to reduce the threshold current in semiconductor laser diodes. The inversion condition in a semiconductor laser was derived by Bernard and Durrafourg (1961) to be E_f_c - E_f_v > E_g where E_f_c and E_f_v are the quasi Fermi-levels for
Fig. 2.11. Schematic of band-filling for conduction and valence band states for equal numbers of injected electrons and holes in (a) unstrained and (b) strained QWs. The biaxial compression in the strained QW, which reduces the DOS of the uppermost valence band and further separates hh and lh confinement energy, results in a reduced threshold carrier density.

electrons and holes, respectively, and $E_g$ is the band-gap. The positions of quasi Fermi-levels are determined by the number density of injected or photo-generated electrons and holes, and the density of states in the conduction and valence band. In unstrained QW structures (see Fig. 2.11a), $E_{fv}$ is usually quite far above the confined valence levels because the valence band consists of several closely spaced levels with a high total density of states. The occupancy factor $f_v(E)$ is therefore small, in order to maintain the charge
neutralit. Hence, in order to reach the inversion condition, the unstrained diode laser has to be pumped hard to push the electron quasi Fermi level well into the conduction band. In strained QWs, the increased splitting of the various hole states as well as the reduction of the in-plane hole effective mass significantly lowers the hole quasi Fermi level at a given hole density, as shown in Fig. 2.11b, which in turn lowers the threshold injection current.
CHAPTER III

GROWTH AND CHARACTERIZATION OF InAs/GaAs STRAINED-LAYER SUPERLATTICE QUANTUM WELLS

In this chapter, the motivation behind our investigation of all-binary InAs/GaAs short-period strained-layer superlattices (SPSLS) as a possible alternative to InGaAs alloy quantum wells for device applications will be discussed. The growth methods and the structure of these samples will also be discussed. Transmission electron microscopy (TEM), linear absorption spectroscopy and photoluminescence (PL) have been used to characterize the all-binary structures and the results indicate that these structures are of very high quality.

§ 3.1 Motivation

In spite of all of the advantages of strained ternary alloy InGaAs/GaAs QWs, which were discussed in §1 and §2.3, some limitations to the performance of these structures have been encountered. Firstly, because of the ternary nature of InGaAs alloy QWs, alloy disorder scattering is unavoidable, therefore, at low temperatures, carrier mobilities in InGaAs/GaAs QWs are not higher than those in GaAs/AlGaAs QWs, in spite of the lower effective masses of carriers in the first structure (Kastalsky et al. 1982). Secondly, the alloy disorder introduces an additional contribution to the excitonic line width (Basu 1990). Thirdly, the alloy concentration of the ternary well, and hence the
wavelength of the $n=1$ excitonic absorption peak, is difficult to reproduce, because it is
difficult to reliably measure and control the group III flux ratio during the InGaAs growth
(Hasenberg et al. 1990). We should emphasize here that wavelength reproducibility is
essential to practical photoelectron device applications. Finally, the critical thickness
parameter (Weng 1989) places an upper limit on the indium mole-fraction of
InGaAs/GaAs QWs with a given well width, restricting the flexibility in varying the indium
mole-fraction and well width to obtain increased spectral coverage.

An attempt has been made to use ordered all-binary (InAs)(GaAs) SPSLS as an
alternative to the ternary InGaAs alloy, not only to overcome the previously stated
difficulties, but also to gain extra advantages. Firstly, because of the binary nature of
(InAs)(GaAs) SPSLS/GaAs QWs, we might expect to eliminate, or at least reduce, the
problems associated with the alloy disorder in InGaAs/GaAs QWs. This has led to
predictions of a substantial reduction or even elimination of alloy disorder scattering and
to the proposed use of (InAs) (GaAs) SPSLS as the channel material in modulation-doped
field effect transistors to improve the speed of these devices (Singh 1986). Secondly,
during the growth of (InAs)(GaAs) SPSLS, accurate control of the InAs and GaAs layer
thicknesses is possible by monitoring the layer thickness with in-situ reflection high-energy
electron diffraction (RHEED) oscillations (Ploog 1989, Chen et al. 1990). This diagnostic
allows the desired effective composition and band gap to be more readily obtained.
Thirdly, we have found experimentally that the SPSLS QWs can accommodate a high
average indium mole fraction (30%-40%) in wide wells (10-20 nm), compared to the
indium mole-fraction of less than 20% for 10 nm wide InGaAs QWs (Weng, 1989),
without evidence of strain relaxation due to misfit dislocation formation. This too has led
to the expectation of higher electron mobilities because of the lower electron effective
mass of InAs.
Differences between the ternary alloy and binary superlattice QWs are shown schematically in Fig. 3.1. The upper part of Fig. 3.1a shows the three pieces of material of which an InGaAs/GaAs QW is composed. The difference in the heights of the InGaAs and GaAs blocks reflects the difference in lattice constants of free standing InGaAs and GaAs crystals. The middle part of Fig. 3.1a shows an InGaAs/GaAs QW, the difference in lattice constants of InGaAs and GaAs is entirely accommodated as elastic strain in the InGaAs layer, assuming the structure is grown on a GaAs substrate. The lower part of Fig. 3.1a shows the conduction band structure of the InGaAs/GaAs QW and the n=1 wave function of electrons confined in the QW. For comparison, a superlattice QW is shown in Fig. 3.1b. The upper part of Fig. 3.1b shows the free standing InAs and GaAs layers used to compose the superlattice QW. Note the large difference in lattice constants of InAs and GaAs (~7.1 %). The middle part of Fig. 3.1b shows the superlattice QW, again, the difference in lattice constants of InAs and GaAs is entirely accommodated as elastic strain in the InAs layers, assuming the structure is grown on a GaAs substrate. The lower part of Fig. 3.1b shows the conduction band structure of the InAs/GaAs superlattice QW. There are many thin potential wells in the superlattice, each of which corresponds to an InAs layer. The GaAs barriers are too thin (only a few atomic layers thick) to effectively confine carriers, and the wave functions of carriers in adjacent thin wells strongly interfere with each other. As a result, the entire superlattice functions as a single QW. Although the wave functions of carriers confined in the superlattice QW have small modulations corresponding to the periodicity of the superlattice, they are nearly identical to the wave functions of carriers confined in a single QW, as shown in the lower part of the Fig. 3.1b for n=1 electron wave function.
Fig. 3.1. Schematic comparison of strained ternary alloy InGaAs QW and strained all-binary InAs/GaAs superlattice QW. The upper part of the figure shows the free standing material pieces used to compose (a) the ternary alloy and (b) the binary superlattice QWs. The middle part of the figure shows the strained (a) InGaAs alloy and (b) InAs/GaAs superlattice QWs. The lower part of the figure shows conduction band structures and $n=1$ electron wave functions of (a) the ternary alloy and (b) the binary superlattice QWs, and shows that the superlattice functions as a single QW.
§ 3.2 Sample structures, notations and growth conditions

We have grown various \((\text{InAs})_i(\text{GaAs})_j\) SPSLS and used them as the quantum well in multiple quantum well (MQW) structures. The notation \(i \times j \times k\) is employed to specify the SPSLS. Specifically, \(i\) and \(j\) give the thickness, in monolayers (ML), of each layer of InAs and GaAs respectively, and \(k\) designates the number of layers of InAs that alternate with the \((k-l)\) layers of GaAs to form each SPSLS. All the SPSLS QW samples we have studied are listed in Table 3.1.

<table>
<thead>
<tr>
<th>Sample Notation</th>
<th>Well Composition</th>
<th>Number of InAs Layers</th>
<th>Well Width</th>
<th>Equivalent In Mole-fraction of SPSLS</th>
<th>GaAs Barrier Width</th>
</tr>
</thead>
<tbody>
<tr>
<td>(2 \times 5 \times 6)</td>
<td>((\text{InAs})_2(\text{GaAs})_5)</td>
<td>6</td>
<td>11.0 nm</td>
<td>32 %</td>
<td>20 nm</td>
</tr>
<tr>
<td>(2 \times 5 \times 8)</td>
<td>&quot;</td>
<td>8</td>
<td>15.2 nm</td>
<td>31 %</td>
<td>&quot;</td>
</tr>
<tr>
<td>(2 \times 5 \times 10)</td>
<td>&quot;</td>
<td>10</td>
<td>19.3 nm</td>
<td>31 %</td>
<td>&quot;</td>
</tr>
<tr>
<td>(2 \times 4 \times 4)</td>
<td>((\text{InAs})_2(\text{GaAs})_4)</td>
<td>4</td>
<td>6.0 nm</td>
<td>40 %</td>
<td>&quot;</td>
</tr>
<tr>
<td>(2 \times 4 \times 6)</td>
<td>&quot;</td>
<td>6</td>
<td>9.6 nm</td>
<td>38 %</td>
<td>&quot;</td>
</tr>
<tr>
<td>(2 \times 4 \times 8)</td>
<td>&quot;</td>
<td>8</td>
<td>13.2 nm</td>
<td>36 %</td>
<td>&quot;</td>
</tr>
<tr>
<td>(2 \times 4 \times 10)</td>
<td>&quot;</td>
<td>10</td>
<td>16.8 nm</td>
<td>36 %</td>
<td>&quot;</td>
</tr>
</tbody>
</table>

Table 3.1 Notations and structures of SPSLS QWs.

The SPSLS QWs have been grown on (100) GaAs substrates by molecular beam epitaxy (MBE) as well as by migration enhanced epitaxy (MEE). Excellent results have been obtained with both growth techniques. In all of the samples, a 400 - 500 nm GaAs buffer was grown with a substrate temperature of 600°C after successfully desorbing the native oxide layer. Then, the substrate temperature was ramped down to 530°C in 5 min. as GaAs growth was continued. Next, the SPSLS QWs were grown on the buffer layer by either MBE or by MEE. During the growth, the in-situ RHEED oscillation method was used to monitor the layer thicknesses. Fifty identical QWs were grown on each sample.
Finally, a 100 nm thick GaAs cap layer was grown on top of the MQW. A schematic diagram of sample 2x5x6 is shown in Fig. 3.2.

Transmission electron microscopy (TEM) measurements have been performed on sample 2x5x8, and a typical result is shown in Fig. 3.3. The TEM picture shows clear evidence of layered structure of the QWs.

Fig. 3.2. Schematic diagram of a 2x5x6 InAs/GaAs strained layer superlattice quantum well grown on a GaAs substrate.
Fig. 3.3. Transmission electron microscopy (TEM) image for the 2x5x8 sample. Two sets of eight thin strips across the picture are the clear evidence of the layered structure of the sample. The striped areas represent the (InAs)$_2$(GaAs)$_5$ superlattice well, and the rest of the picture represents the GaAs barrier.
§ 3.3 Linear optical absorption spectra of SPSLS MQWs

It is necessary to verify that these rather complicated SPSLS structures do function as QWs, and are of good quality. In order to do that, the most straightforward technique to use is linear absorption spectroscopy, not only because the measurement is simple, but also because it provides much useful information. The observation of step-like features in the optical absorption spectra is the most important indication of the onset of quantum size effects and of the quasi 2D nature of the QWs. The line width of the $n = 1$ excitonic absorption peak and the Stokes shift of the photoluminescence peak with respect to the absorption peak can also be used as indicators of sample quality. Furthermore, precise data concerning details of the SPSLS electronic band structure and the band offset of the QWs can be extracted from the energies of the optical transitions associated with the MQWs.

The linear optical absorption spectra of the samples were measured with the apparatus shown in Fig. 3.4. The light source was a Xenon arc lamp, a portion of the output spectrum of which was selected by a computer-controlled 1/4 meter monochromator. The input and output slits of the monochromator were adjusted so that the resolution of the system was about 1 nm. The output from the monochromator was chopped with a mechanical chopper, then collimated and focused on to the sample. The samples were mounted in a cryostat which allowed the temperature of the samples to be varied between 15 K and room temperature. The temperature of the samples was measured with a Au-Chromel thermocouple mounted on the sample frame, close to the samples. The transmitted light was focused onto silicon photodiode D1. Because the output power of the Xenon lamp was strongly dependent on wavelength, it was necessary to monitor this power in order to obtain an accurate transmission spectrum of the sample.
This was done with a second photodiode D2, identical to D1, which monitored light reflected from the front window of the cryostat. The output from each diode was passed through a lock-in amplifier which was tuned to the chopper frequency. The transmission spectrum of the sample was obtained by scanning the monochromator and dividing the signal from D1 by that from D2. A correction was made for reflection from the sample surface by assuming unity transmission at a wavelength much longer than that of the n=1 excitonic absorption peak and scaling the measured transmission spectrum to 1 at this point. The quantity used on the ordinate of most of our absorption spectra is absorbance which is defined as $-\log(1/T)$, and it should be noted that log represents the decadic logarithm.

![Diagram](image)

**Fig. 3.4.** Experiment apparatus for low temperature linear absorption coefficient measurement. The following abbreviations are used: B.S. = beamsplitter, D = detector, L = lens.

The room temperature absorption spectra of all the samples were also measured with Perkin-Elmer Lambda 9 spectrophotometer, and the results are consistent with those obtained with the method described in last paragraph.
Fig. 3.5 shows the room temperature absorbance spectra of five of the samples. The primary features of note are the strong, clearly-resolved n=1 excitonic absorption peaks between 960 and 1030 nm. Note that the spectral position of the peak for a given superlattice composition can be adjusted by varying the well width, i.e. the overall length of the superlattice. This is consistent with each of these superlattices functioning as a single quantum well. Also note that, by fine-tuning the composition, it should be possible to use this material system as the gain medium for 980 nm room-temperature laser diodes. As we discussed in §2.3, this wavelength is appropriate for efficient photo-pumping of erbium-based lasers for the important 1.55 μm and 2.8 μm regions.

Fig. 3.5. Room temperature absorbance of the InAs/GaAs strained layer superlattice QW samples.
By comparing the n=1 excitonic absorption peak linewidth and the oscillator strength of 2x5xk and 2x4xk structures, it is clear that the quality of the 2x4xk structures is not as high as that of the 2x5xk's. The reason for that might be that the growth of (InAs)$_2$(GaAs)$_4$ has not yet been optimized, since only one attempt has been made to grow these 2x4xk structures. Therefore, the analysis will be mostly focused on (InAs)$_2$(GaAs)$_5$ structures.

Low-temperature (15 K) absorption spectra of the 2x5xk samples are shown in Fig. 3.6. All of these spectra display step-like features, which correspond to the step-like 2D density of states discussed in §2.1, plus excitonic absorption peaks at the onset of each step. Note that the magnitudes of the first step of all three spectra are virtually the same, in spite of the fact that the 2x5x10 wells are almost twice as thick as the 2x5x6 wells. This is more evidence of the 2D-like nature of these structures. Note that as the width of the QW is increased, the n=1 heavy hole exciton peak, which is located between 900 and 920 nm, shows an increase in width. This tendency can be attributed to the decrease of exciton binding energy with increasing well width (Greene et al. 1984). The linewidths of the n=1 hh excitonic peaks (full width at half maximum) are estimated to be 4.8, 5.4 and 6.1 meV for the 2x5x6, 2x5x8 and 2x5x10 wells, respectively. These linewidths are comparable to the very best InGaAs alloy quantum wells (Dahl et al. 1987). The next-strongest feature in each spectrum is located at ~ 850, 868 and 885 nm for k = 6, 8 and 10, respectively. Because of the strength of these peaks and because they dramatically shift to longer wavelength with increasing well width, we tentatively identify them with the n=2 hh exciton. In each spectrum, there is also a weaker peak located at a roughly constant energy above the n=1 hh peak at 870, 877, and 880 nm for k = 6, 8, and 10, respectively. The relatively weak dependence of this separation on well width and the
lower strength of the peak suggest that it is associated with the $n = 1$ light hole (lh) excitonic transition.

Fig 3.6 Low temperature absorbance of the $2\times5\times k$ InAs/GaAs strained layer superlattice QW samples.

The linear absorption spectra obtained from our high quality SPSLS QWs show clear evidence of the 2D nature of these samples, and are indicative of the elimination of material imperfections, e.g. well width fluctuations and interface roughness, which would cause the broadening of excitonic absorption peaks. Compared to the excitonic features reported in earlier published works (Marzin et al. 1990) on (InAs)(GaAs) SPSLS QWs (see insert in Fig 3.6), our results show a great improvement in sample quality.
Photoluminescence (PL) is another very important method which can be used to characterize QW structures. In our PL experiment, carriers were photo-excited ~90 meV above the bandedge of the material. These carriers lose excess energy by transferring to the lattice via phonon scattering and reach lattice temperature in ~100 ps. At low lattice temperatures, many of these photo-excited carriers form excitons and undergo radiative recombination on a time scale of ~1 ns and therefore the PL signal of the QW is usually dominated by excitonic recombination on the n=1 hh line. As a result, the linewidth of the PL spectrum is directly related to the exciton linewidth. Furthermore, comparison between PL and absorption data yields the Stokes-shift due to the localization of exciton states, which is believed to be the result of material imperfections such as interface roughness, defect and alloy disorder. By time-resolving the PL signal, we can obtain information about carrier cooling processes and carrier lifetimes.

Fig. 3.7 shows the apparatus used for the PL measurement. The excitation source was a synchronously mode-locked Styryl 9 dye laser, pumped by the frequency-doubled output of a mode-locked Nd:YAG laser. This source produced pulses of ~5 ps duration, with a bandwidth of ~2 meV, at a 76 MHz repetition rate. The wavelength of the dye laser was tunable so that the excess energy of the photo excited carriers could be controlled. These pump pulses were then focused by a f=15cm best form lens onto the sample. The photoluminescence was collected and focused with f/4 optics onto the entrance slit of a 0.25 m spectrometer. The spectrally dispersed output from the spectrometer was then arrayed spatially along a slit, which was in turn imaged onto the photocathode of the streak camera. Electrons were promptly emitted from the cathode by means of the photoelectric effect, and were rapidly accelerated through a mesh towards
the anode. The resulting electron beam current, as a function of time, closely resembled the envelope function of the photoluminescence $I(t)$, and, as a function of position, closely resembled the spectrum on the input slit, $I(x)$. The focus cone provided an electrostatic lensing field which sharply imaged the slit onto the back phosphor screen. A measurement of the temporal profile of electron beam current was obtained by sweeping the electron beam across the phosphor screen by applying a linear voltage ramp to a set of deflection plates. This occurred because those electrons leaving the photocathode at earlier times arrived at the phosphor at one position, while those that leaving at later times arrived at a different position, and the function of time $I(t)$ was thereby effectively transformed into a function of position $I(y)$. The spectrally and temporally dispersed signal $I(x,y)$ at the
phosphor was intensified and monitored with a CCD camera. The streak camera was operated in synchroscan mode, i.e. the ramp field was synchronized with the excitation laser pulses, which had a 76 MHz repetition rate. The system has an overall spectral resolution of ~0.6 meV and a temporal resolution of ~20 ps.

Fig. 3.8. 15 K temporally integrated photoluminescence spectra for 2x5xk samples.

Single, intense, narrow emission lines are observed in the temporally integrated PL spectra shown in Fig. 3.8. The temporally dispersed signal is numerically integrated over the range -20 ps to 1800 ps. These measurements were taken at 15K with pump laser tuned to 840 nm, which is below the band gap of the GaAs substrate at this temperature,
ensure that carriers are generated in the wells. At an initial average in-well carrier density estimated to be \( \sim 4 \times 10^{10} \text{cm}^{-2} \) (differing by, at most, a factor of 1.25 for the different samples), this PL is considered to be predominantly excitonic. The linewidths vary from \( \sim 7.5 \text{ meV} \) for the 2x5x6 structure to \( \sim 9 \text{ meV} \) for the 2x5x10. These linewidth are comparable to those of high quality InGaAs alloy material and are an improvement over those previously reported (Marzin et al. 1989) for InAs/GaAs SPSLSs, further attesting to the quality of the samples.

Yet another indication of the high material quality of the SPSLS samples is the remarkably close correspondence between the absorption and PL peaks. An example of the 15 K absorption and PL results, taken for the 2x5x6 sample, is shown in Fig. 3.8(a). In this example, the red shift of the PL peak with respect to the \( n=1 \) excitonic absorption peak is merely \( \sim -0.5 \text{ meV} \). A similar correspondence was observed for other 2x5xk samples. Red-shifts of several meV of the PL emission relative to the \( n=1 \) hh excitonic absorption have been commonly observed at low temperature for all but the very highest quality MQWs and have been attributed to excitonic localization due to material imperfections within the wells. This shift has been particularly difficult to reduce in the case of ternary alloy wells because of the additional localization due to alloy disorder. The absence of a significant red-shift in the data is consistent with the elimination of the alloy disorder contribution in all-binary samples and is indicative of high interface quality. For comparison, an example of the absorption and PL results obtained for the 2x4x6 sample is shown in Fig. 3.9b. The \( \sim 4 \text{ meV} \) red shift of the PL peak relative to the absorption shown in Fig. 3.9b is typical for 2x4xk samples, which, as mentioned previously, have not yet been optimized.

In Fig. 3.10, the spectrally integrated PL signal (integrated for about 10 meV across the emission peak of each individual sample) of 2x5xk samples were plotted as
Fig. 3.9. Excitonic absorbance and photoluminescence at 15 K for (a) 2x5x6 sample and (b) 2x4x6 sample.
functions of time, with the display limited to ~2 ns by the streak camera temporal window. The PL decays of Fig. 3.10 are nearly exponential, consistent with the exponential decay characteristics of monomolecular exciton-dominated recombination. The time constants are approximately 1800 ps, 1650 ps and 1400 ps for 2x5x6, 2x5x8 and 2x5x10 respectively. These long life-times provide additional evidence of the high material quality. The PL rise time of ~200 ps shown in Fig. 3.10 is fully resolved. Since the carriers were clearly photo-generated within the wells, rather than the barrier material, this rise cannot be attributed to carrier transport to, and/or trapping by, the wells, but rather to carrier cooling within the wells. Indeed, remarkably similar time-resolved PL traces have been obtained for these samples for excitation at 800 nm (above the GaAs band edge).
This suggests that the carrier trapping by the wells is both efficient and very rapid ( < 10 ps), in agreement with earlier measurements in AlGaAs QWs (Göbel et al. 1988).

§ 3.5 Summary

The ordered, all-binary InAs/GaAs SPSLS QW samples were grown in order to study the possibility of using them as an alternative to InGaAs alloy QWs in device applications. These samples were characterized with TEM, linear absorption and photoluminescence techniques, and the results indicate that the material properties of these all-binary structures are at least as good as those of the QWs fabricated from other well characterized alloy material systems, and are a great improvement over previously studied (InAs)(GaAs) SPSLS QWs.
CHAPTER IV

OPTICAL NONLINEARITIES OF InAs/GaAs STRAINED-LAYER SUPERLATTICE QUANTUM WELLS

In this chapter, a description is made of the measurement of the nonlinear absorption cross section, $\sigma_{eh}$, and the nonlinear refraction coefficient, $n_{eh}$, associated with bleaching of the excitonic absorption of InAs/GaAs SPSLS QWs at room temperature. The nonlinear absorption cross section was obtained directly from the results of a differential transmission measurement employing the pump-probe technique. The nonlinear refraction coefficient was extracted from the efficiency of diffraction from the photo-generated transient grating and the known $\sigma_{eh}$. The consistency of the differential transmission and the transient grating measurements is demonstrated via a Kramers-Kronig analysis.

§ 4.1 Mechanism, theory and techniques for measurement of optical nonlinearities

The optical nonlinearities discussed in this section are induced by a photo-generated and thermalized electron-hole plasma. These should be clearly distinguished from the nonlinearities due to excitation below the absorption edge, in which case the laser field induces a coherent polarization of the valence electrons that persists essentially only as long as the laser is applied. The nonlinear polarization can couple various optical fields, which exchange photons via the material, but no net power is deposited in the
material. On the other hand, photo-excitation above the absorption edge generates real electron-hole pairs, and hence induces changes in the optical properties of the material mainly via phase-space filling by the photo-generated carriers (Schmitt-Rink et al. 1985, Knox et al. 1986). The optical nonlinearity due to photo-generated electron and hole populations persist as long as the electrons and holes themselves do not recombine.

The electron-hole plasma can be optically generated either directly above the gap (Shank et al. 1983, Chemla et al. 1984, Park et al. 1988) or indirectly following exciton creation by resonant absorption (Miller et al. 1982, Weiner et al. 1986, Tai et al. 1987). Excitons that are generated by resonant excitation within the absorption peak are only stable at low temperature where they can last as long as a few ns. In the case of III-V QWs at room temperature, the excitons are thermally unstable and very quickly transform into free electron-hole pairs, since the LO-phonon energy is much larger than the exciton binding energy and excitons will be ionized via a single collision with a phonon. At room temperature, the exciton lifetime is only 400 fs in GaAs QWs (Chemla et al. 1984), 250 fs in InGaAs QWs (Schmitt-Rink et al. 1989) and 150 fs in the InAs/GaAs SPSLS QWs studied in this work (measured with the method used by Chemla et al. 1984)). Furthermore, ultrafast photo-generation can produce non-equilibrium distributions of electron and hole states, and it will take a few ps for them to reach thermal equilibrium with the lattice (Schmitt-Rink et al. 1989). Once photo-generated carriers are thermalized, it has been found experimentally that the changes in the absorption spectrum do not depend on the wavelength of the excitation pulse or on its duration, they depend only on the density of excited electron-hole pairs (Chemla et al. 1988).

The change of the absorption coefficient, $\Delta \alpha$, induced by photo-generated carriers in InAs/GaAs SPSLS QW samples was measured with the degenerate pump-probe technique. As shown in Fig. 4.1, a strong pump pulse is used to optically excite the
sample and therefore to change its absorption coefficient. The value of $\Delta \alpha$ is obtained by measuring the transmission of a weak probe pulse. The probe pulse is 10's of ps behind the pump pulse to allow the complete thermalization of the photo-generated carriers, and is much weaker than the pump so that the probe itself will not cause any significant changes in the sample. Since the directly measured value is transmittance, this technique is also known as the degenerate differential transmission measurement. The differential transmission, $\Delta T/T$, is defined as

$$\frac{\Delta T}{T} = \frac{T_{on} - T_{off}}{T_{off}},$$

where $T_{on}$ is the probe transmission after photo-excitation by the pump pulse, $T_{off}$ is the probe transmission without the pump pulse, i.e. the linear transmission. The change of absorption coefficient, $\Delta \alpha$, can be simply calculated by

$$\Delta \alpha = \frac{-1}{d} \ln \left( 1 + \frac{\Delta T}{T} \right),$$

where $d$ is the length of the sample. The term "degenerate" is used to indicate that both pump and probe pulses emanate from a single tunable laser and have the same frequency. The spectrum of $\Delta \alpha$ can be measured by tuning the laser across a certain wavelength range. As stated earlier, for the type of nonlinearity studied here, the excitation wavelength makes no difference. The only thing that is important is the photo-generated carrier density. Due to the shape of the absorption spectrum and the variation of the laser output power with wavelength, the photo-generated carrier densities for degenerate pump-probe measurements are wavelength dependent. However, if the slight broadening and red shifting effects are ignored, within the unsaturated region the change of absorption is linearly proportion to the carrier density (Schmitt-Rink et al. 1985),

$$\Delta \alpha = \sigma_{\text{ef}} N,$$
where $\sigma_{eh}$ is defined as the absorption cross section, which is the change of absorption coefficient per photo-generated electron-hole pair, and $N$ is the number density of photo-generated electron-hole pairs. Using Eqs. (4.2) and (4.3) and the linear absorption spectrum of the sample, $\sigma_{eh}$ can be easily extracted from the degenerate differential transmission measurement. The introduction of $\sigma_{eh}$ makes it easier to interpret the degenerate differential transmission results, and more importantly, makes it easier to compare nonlinearities of different samples measured under different conditions.

Fig. 4.1. Schematic diagram of pump-probe measurement. A strong pump pulse is used to optically excite the sample and therefore to change its absorption coefficient, $\alpha$. The value of $\Delta \alpha$ is obtained by measuring the change in transmission of a weak probe pulse. A time delay between pump and probe pulses is necessary to allow the photo-excited carriers to be thermalized.
In the unsaturated region, the spatial distribution and temporal evolution of the nonlinearities, e.g. \( \Delta \alpha \), are simply determined by the corresponding distribution and evolution of the photo-generated carrier densities. Assuming that the pump pulse has a Gaussian spatial intensity distribution, and that the duration of the pump pulse is much shorter than the lifetime of the photo-generated carriers, the carrier density in quasi 2D systems, such as the QW samples studied here, after the ultrafast photo-excitation can be described by

\[
\frac{\partial N(x, y, t)}{\partial t} + \frac{N(x, y, t)}{\tau_R} - D_a \nabla^2 N(x, y, t) = 0, \tag{4.4a}
\]

with

\[
N(x, y, t = 0) = \frac{\alpha F(x, y)}{h\nu} = N_0 \exp \left( -\frac{x^2 + y^2}{\omega_o^2} \right), \tag{4.4b}
\]

where \( N \) is number density of photo-generated electron-hole pairs, \( N_0 \) is the number density at the center of the laser spot, \( \tau_R \) is the recombination lifetime of the photo-generated electron-hole pairs, \( D_a \) is the ambipolar diffusion coefficient of the host material, \( F \) is the fluence of the pump pulse, \( \omega_o \) is HW1/eM of the intensity of the pump pulse, \( \alpha \) is the linear absorption coefficient of the material, and \( h\nu \) is the photon energy of the excitation laser. The solution of Eq. (4.4a) with initial conditions given by Eq. (4.4b) is (Appendix I)

\[
N(x, y, t) = \frac{N_0}{1 + 4D_a t / \omega_o^2} \exp \left( -\frac{x^2 + y^2}{\omega_o^2 + 4D_a t} \right) e^{-t/\tau_R}. \tag{4.5}
\]

Eq. (4.5) indicates that the number density \( N \), and therefore \( \Delta \alpha \) as defined in Eq. (4.3), decays as time progresses, and that the decay rate depends on the carrier lifetime, the laser spot size and the diffusion coefficient of the material. When \( t = \omega_o^2 / 2D_a \),

\[
N(\rho = 0) = \frac{N_0}{3} \approx \frac{N_0}{e}.
\]

For the typical pump spot size of about 60 \( \mu \text{m} \) (FW1/eM)
used in our experiment, and using \( D_a = 75 \text{ cm}^2/\text{s} \), the value of \( \omega_0^2/2D_a \) is 60 ns. Considering the \( \sim 1 \text{ ns} \) recombination life-time in all the samples that have been studied here, it is reasonable to neglect the effect of carriers diffusing away from the photo-excited area. Also, if only the center part (\( \sim 1/3 \) of the FW1/eM of intensity) of the photo-excited area is studied with the probe pulse, it is a good approximation to assume that the probed area of the pump spot has a uniform intensity.

The optically induced changes in the absorption are accompanied by refractive index changes. As for \( \Delta \alpha \), in the unsaturated regime the change of refractive index, \( \Delta n \), is linearly proportional to the number density of photo-generated electron-hole pairs, i.e.

\[
\Delta n = n_{eh}N. \tag{4.6}
\]

with \( n_{eh} \) defined as the nonlinear refraction coefficient, which is the change of refractive index per photo-generated electron-hole pair. For the very thin samples (\( d \sim 1\mu \text{m} \)) studied here, and in the unsaturated regime of the nonlinearity, the change in the optical path length, \( \Delta nd \), due to photon excitation is very difficult to measure directly by, e.g., interferometry. Nevertheless, \( n_{eh}(\omega) \) may be obtained from \( \sigma_{eh}(\omega) \) via the Kramers-Kronig relation.

The refractive index and the absorption coefficient of materials containing a fixed number of carriers are related by the Kramers-Kronig relations (Stern 1963). The absorption coefficient and the refractive index of a photo-excited semiconductor are therefore also related by the Kramers-Kronig relations (Miller et al. 1983a, Chemla et al. 1984). The change of refractive index associated with the change of absorption coefficient is given by

\[
\Delta n(\omega) = \frac{c}{\pi} \int_0^\infty \frac{\Delta \alpha(\omega')d\omega'}{\omega^2 + \omega'^2}, \tag{4.7}
\]

where \( P \) indicates that the principal value of the integral has to be evaluated.
Fig. 4.2. Schematic diagram of transient grating measurement. Two identical pump pulses separated by angle $\theta$ are spatially and temporally coincident at the sample. The interference between two pump pulses modulates the intensity and therefore creates a photo-excited carrier grating, which decays via diffusion and recombination. The decay of the grating is monitored by measuring the efficiency with which a weak probe pulse is diffracted from the grating.

For the degenerate differential transmission experiments, the $\Delta \alpha$ spectra are obtained by tuning the laser wavelength across the $n = 1$ hh excitonic absorption peak. The measurements are therefore not made at fixed carrier density, since both linear absorption coefficient and laser output power are functions of wavelength. In order to use the Kramers-Kronig relations, it is necessary to convert the $\Delta \alpha(\omega)$ to $\alpha_{hh}(\omega)$ by using the
definition given in Eq. (4.3). Both $\alpha_{eh}(\omega)$ and $n_{eh}(\omega)$ are independent of carrier density over the range of interest and are therefore related by Kramers-Kronig relations

$$n_{eh}(\omega) = \frac{c}{\pi} \int_0^\infty \frac{\sigma_{eh}(\omega') d\omega'}{\omega'^2 - \omega^2}. \quad (4.8)$$

Information on $\sigma_{eh}$ and $n_{eh}$ can also be obtained from the transient grating diffraction efficiency spectrum of the sample. The geometry of the transient grating measurement is shown in Fig. 4.2. Two identical pump pulses separated by an angle $\theta$ are spatially and temporally coincident at the sample. The interference between the two pump pulses modulates their intensity, and hence the fluence experienced by the sample along the $x$-direction. Under the plane wave approximation, assuming plane wavefronts and a uniform spatial intensity distribution of the incident pump pulses, the modulated fluence is given by

$$F(x) = 2F_1 \left[ 1 + \sin \left( \frac{2\pi x}{\Lambda} \right) \right]. \quad (4.9)$$

where $F_1$ is the fluence of each pump pulse, $\Lambda$ is the grating spacing which is given by

$$\Lambda = \frac{\lambda}{2 \sin (\theta/2)}, \quad (4.10)$$

where $\lambda$ is the laser wavelength. A sinusoidally modulated distribution of carriers, i.e. a carrier grating, will be created via direct absorption of two interfering pump pulses. This grating will decay through recombination and in-well diffusion as described by

$$\frac{\partial N(x,t)}{\partial t} + \frac{N(x,t)}{\tau_R} - D_a \nabla^2 N(x,t) = 0, \quad (4.11a)$$

with initial condition

$$N(x,t=0) = \frac{\alpha F(x)}{hv} = N_0 \left[ 1 + \cos \left( \frac{2\pi x}{\Lambda} \right) \right]. \quad (4.11b)$$
which assumes instantaneous excitation. The solution of Eq. (4.11) is

\[ N(x,t) = N_0 \left[ 1 + e^{-t/\tau_D} \cos \left( \frac{2\pi x}{\Lambda} \right) \right] e^{-t/\tau_R}, \]  

(4.12)

where \( \tau_D \) is the diffusive decay time. Eq. (4.12) indicates that the magnitude of the number density modulation will decay with a time constant \( \tau_D \) due to the in-well diffusion, and the entire carrier population will decay with time constant \( \tau_R \) due to the carrier recombination. Substituting Eq. (4.12) into Eq. (4.11a) gives

\[ \frac{1}{\tau_D} = \frac{4\pi^2 D_a}{\Lambda^2}. \]  

(4.13)

The grating decay rate, \( \tau_G \), is defined as

\[ \frac{1}{\tau_G} = \frac{1}{\tau_D} + \frac{1}{\tau_R} = \frac{4\pi^2 D_a}{\Lambda^2} + \frac{1}{\tau_R}. \]  

(4.14)

The carrier density distribution given by Eq. (4.12) will modulate the absorption and refractive index, according to Eqs. (4.3) and (4.6), forming a diffraction grating. The efficiency, \( \eta \), with which a weak probe pulse is scattered from a thin sinusoidal grating is given by (Appendix II):

\[ \eta \approx \frac{1}{4} \exp \left( -\alpha d - \sigma_{eh} e^{-t/\tau_R} N_0 d - 2t / \tau_G \right) \left[ \left( \frac{2\pi n_{eh}}{\lambda} \right)^2 + \left( \frac{\sigma_{eh}}{2} \right)^2 \right] N_0^2 d^2, \]  

(4.15)

where \( d \) is the sample thickness, \( N_0 \) is the average number density of photo-generated electron-hole pairs, and \( \alpha \) is the linear absorption coefficient. Eq. (4.15) indicates that the diffraction efficiency decays with time constant \( \tau_G/2 \), due to both in-well diffusion and carrier recombination. Using \( \tau_R = 1 \) ns, which is typical among all the samples studied in this work, \( D_a = 18 \) cm²/s as measured at room temperature in the best 2x5xk (InAs)(GaAs) SPSLS samples, and \( \Lambda = 8.5 \) µm, which was the grating spacing used in the
transient grating experiment, Eq. (4.14) yields $\tau_G = 500$ ps. When $t \ll \tau_G$ and the carrier density is low enough so that $\Delta \alpha = \sigma_{eh} N_0 \ll \alpha$, Eq. (4.15) becomes

$$\eta = \frac{1}{4} e^{-\sigma_{eh} \left( \frac{2 \pi n_{eh}}{\lambda} \right)^2 + \left( \frac{\sigma_{eh}}{2} \right)^2} N_0^2 d^2.$$

(4.16)

The diffraction efficiency, measured before significant decay by in-well diffusion has happened, can be used as a check on the consistency of the measured $\sigma_{eh}$ and $n_{eh}$ calculated from the $\Delta T/T$ measurement and the Kramers-Kronig transformation.

It should be emphasized here that Eq. (4.15) was derived, and will only be valid, under the following approximations and conditions:

(1) Plane wave approximation, which assumes that the pump pulses have plane wavefronts, uniform spatial intensity distributions and infinite cross sections. These assumptions are appropriate when the sample is set at the beam waist, only the central 1/3 part of the pump laser spot is probed and the laser spot is sufficiently large that the effects of carrier diffusion out of the photo-excited area are negligible.

(2) The fluence of the pump pulses are sufficiently low, i.e. within the unsaturated region of the nonlinearity, so that sinusoidal modulations of $\alpha$ and $n$ will be produced.

(3) The active sample is thin enough that the thin grating condition is satisfied (see discussion in Appendix III). All the samples studied here are thin enough that the use of Eq. (4.15) is appropriate.

(4) The duration of the pump pulses should be much shorter than the carrier recombination time and the diffusion decay time.

(5) Measurements should be made a few ps after the optical excitation, when photo-generated carriers have been thermalized.
Care was taken to ensure that all these conditions were fulfilled in the experiments.

§ 4.2 Experimental apparatus for nonlinear optical measurements

The nonlinear optical properties of the samples in the wavelength region of the $n = 1$ hh exciton were measured with the apparatus shown in Fig. 4.3. The excitation source was a synchronously mode-locked Styril 13 dye laser, which was pumped by the frequency-doubled, fiber-and-grating compressed output of a mode-locked Nd:YAG laser. The dye laser, which was tunable from 950 nm to 990 nm, produced pulses of duration $\sim 500$ fs at a repetition frequency of 76 MHz. Tuning of the laser output wavelength was provided by a two-plate birefringent filter, which was operated by a computer-controlled stepper motor. At 970 nm, the average output power of the dye laser was $\sim 40$ mW.

The output of the dye laser was vertically polarized when it reached the beam-splitter, BS1, and was split by BS1 into a strong beam and a weak beam. The strong beam was further split by a 50/50 beam splitter, BS2, to form pump beams of equal intensity. The weak beam from BS1 went through a periscope which rotated the polarization by 90 degrees, and was attenuated by neutral density filters to form a probe beam. All three beams were aligned so that they were in the same plane and propagating in the same direction, and a $f = 15$ cm focal length best form lens was used to focus all three beams to the same spot on the sample. The probe beam was expanded by a 1:3 telescope, before it reached the final focusing lens, to allow the probe to be focused to a smaller spot than the pump. Therefore, we are able to use the plane wave approximation in the extraction of $\sigma_{eh}$ and $n_{eh}$, as discussed in preceding sections. The focused pump and probe beam were measured, with a 5 $\mu$m diameter pin-hole, to be 57 and 18 $\mu$m (full
width at 1/e maximum) respectively. The relative arrival time of all three pulses at the sample site could be adjusted individually by means of delay stages.

![Experimental apparatus diagram](image)

Fig. 4.3. Experimental apparatus used to perform the nonlinear optical measurements. The following abbreviations are used: BS = beamsplitter, C = chopper, ND = neutral density filter, and P = periscope. BS2 is removed during the differential transmission measurements.

Differential transmission, ΔT/T, was measured by replacing BS2 with a mirror to provide a single, strong pump beam. The probe pulse was retarded by 30 ps with respect to the pump pulses, so that photo-generated carriers had thermalized by the time that the probe pulse arrived at the sample. The typical time scale for the thermalization is only a few picoseconds (Knox et al. 1985, Knox et al. 1986). The pump beam and the probe beam were chopped by choppers C1 and C2 at 27 Hz and 2.1 kHz respectively. A silicon
Fig. 4.4. Schematic sketch of the photodiode output signal for nonlinear optical measurement. (a) Signal for differential transmission measurement. The detector was set to collect the transmitted probe light. The larger signal at "pump on" region reflects the reduction of absorption due to the photo-excitation. (b) Signal for grating diffraction efficiency measurement. The detector was set to collect the probe light diffracted from the photo-generated grating.
photodiode was set behind the sample to detect the transmitted probe beam. A polarizer was set in front of the photodiode to block the scattered pump light, which was cross polarized with respect to the probe light. An idealized output waveform of the photodiode is shown in Fig. 4.4a. Note that the waveform is the product of two square waves with frequencies corresponding to the chopping frequency of the pump and the probe beam respectively. The difference between the amplitude of the 2.1 kHz square wave in the "pump off" and the "pump on" regions corresponds to the change of transmission of the sample due to the phase-space filling caused by the carriers generated by the pump beam. One might expect the minimum value of the signal to be zero, but from inspection of Fig. 4.4a it can be seen that in the "pump on" region the signal has a DC background which is due to the incomplete blockage of the scattered pump light. Cascaded lock-in amplifiers were used to detect the signal modulated at both chopping frequencies, and the output from the second lock-in amplifier gave the change of transmission, $\Delta T$, on an arbitrary scale. In order to measure the linear transmission, $T$, on the same scale, and correct for instrument response, the pump beam was blocked, the probe beam was chopped at both 27 Hz and 2.1 kHz, and the same photodiode and lock-in amplifiers were used to detect transmitted probe light in the absence of any pump light. The signal from the second lock-in amplifier was digitized and recorded by a computer-controlled data acquisition system and these data were used to calculate $\Delta T / T$.

Note that in Fig. 4.4 individual laser pulses are not resolved, because the response time of the photodiode is much longer than the $\sim 13$ ns time interval between consecutive laser pulses and the output from the photodiode is therefore an accumulation over many pulses. However, obtaining the temporal behavior of the nonlinearities did not rely on resolving individual laser pulses, instead it was obtained by varying the relative delay between pulses in the pump and probe beams. The probe pulses yielded information on
the material in a time window, equal in length to the probe pulse duration, at some delay after the sample was excited by the pump pulses. The ~1 ns recombination lifetime of the samples, measured with time resolved PL, indicates complete recovery of the samples in the ~13 ns time interval between consecutive laser pulses.

The wave forms in Fig. 4.4 were idealized by neglecting the power fluctuation of the dye laser output. In the experiments, a second photodiode was used to detect the signal reflected from a glass slide set in the probe beam path. Data were taken only when the power fluctuation monitored by the second photodiode was within ~±5%. The second photodiode was also used to measure the laser output power as a function of wavelength.

Diffraction efficiencies in the forward traveling four-wave mixing geometry were measured with the full apparatus depicted in Fig. 4.3. Transient grating measurements were performed with full temporal overlap of the pump beams at the sample site. As was the case in the differential transmission measurements, the probe beam was retarded by 30 ps with respect to the pumps. For the grating period of 8.5 μm used for these measurements, the diffusion decay time constant, $\tau_D$, was ~200 ps, therefore, diffusion resulted in no significant decay of the grating during the first 30 ps after excitation. An idealized output wave form of the photodiode used to detect the diffracted probe beam is shown in Fig. 4.4b. In the "pump off" region, where one would expect no diffraction, the signal came from the scattered probe light. In the "pump on" region, the wave form contains a 2.1 kHz square wave, which was a sum of the diffracted and the scattered probe light, plus a DC signal from the scattered pump light. Again, it is necessary to use cascaded lock-in amplifiers to separate the diffracted probe from the rest of the signal. The diffraction efficiency is defined in Eq. (4.24) as the ratio of diffracted probe power to the incident probe power. To obtain the diffraction efficiency, the same photodiode and
lock-in amplifiers were used to measure the probe beam chopped at both 27 Hz and 2.1 kHz, in the absence of any pump light.

§ 4.3 Results and discussion

Differential transmission spectra for the 2x5x6 sample, obtained with pump fluence ranging from $5.0 \times 10^{-7}$ Jcm$^{-2}$ to $7.5 \times 10^{-6}$ Jcm$^{-2}$, are plotted in Fig. 4.5. These spectra are consistent with a pump-induced bleaching and broadening of the $n = 1$ hh excitonic absorption line. The spectra are similar to those measured in other quantum-confined systems (Miller, A. et al. 1989, Chemla et al. 1984, Tai et al. 1987), where the bleaching has been mainly attributed to phase-space filling. One notable difference between the present spectra and those previously described is the isolation of the heavy-hole resonance from the light-hole resonance, a result of the large strain-induced splitting of the heavy- and light-hole states. As fluence increases, the differential transmission peak increases and broadens to the high energy side. At the highest fluence, the spectrum develops a tail on the high energy side, which is consistent with absorption bleaching due to band filling. Also, the peak of the negative lobe increases and moves to the low energy side with increasing fluence, which is consistent with collisional broadening of the excitonic absorption line (Schmitt-Rink et al. 1989). The peak measured differential transmission was 50% at a wavelength of 967 nm and at a pump fluence of $7.5 \times 10^{-6}$ Jcm$^{-2}$.

From close examination of the data in Fig. 4.5, it can be seen that differential transmission near the exciton peak saturates with increasing fluence. This can be seen more clearly by plotting $\Delta T/T$ at the exciton peak against fluence, as has been done in Fig. 4.6. From this figure, it is evident that $\Delta T/T$ initially increases linearly with fluence, but
begins to saturate at a fluence of approximately $1.8 \times 10^{-6}$ J cm$^{-2}$. To extract meaningful values of $\sigma_{eh}$ and $n_{eh}$, it is necessary to use $\Delta T/T$ and diffraction efficiency data obtained at a fluence considerably less than this saturation value, i.e., in the fluence regime where each incident photon causes the same change in carrier number density. This condition is also necessary in order to obtain the sinusoidal grating described in §4.1. Similar results to those displayed in Fig. 4.5 and Fig. 4.6 were obtained for the other 2x5x$k$ samples.

The nonlinear absorption cross section, $\sigma_{eh}$, may be simply calculated from Eqs. (4.2) and (4.3), in the unsaturated regime of the nonlinearity, in which the change of
Fig. 4.6. Fluence dependence of the measured differential transmission for the 2x5x6 sample near the peak at 967 nm.

absorption, $\Delta \alpha$, and the change of refractive index, $\Delta n$, are linearly proportional to the intensity of the pump beam. The $\sigma_{eh}(\omega)$ spectra of all three 2x5xk samples were obtained from the measured $\Delta T / T$, using Eqs. (4.2) and (4.3), and are plotted in Fig. 4.7a. In this figure, the abscissa represents the energy offset from the low fluence excitonic absorption line center. This allows a direct comparison between the profiles and magnitudes of the $\sigma_{eh}$ spectra for the different samples to be made.

The $n_{eh}(\omega)$ for each 2x5xk sample corresponding to the data in Fig. 4.7a was calculated using the Kramers-Kronig transformation shown in Eq. (4.8), and was plotted
in Fig. 4.7b. Again, the abscissa represents the energy offset from the low fluence excitonic line center.

<table>
<thead>
<tr>
<th>Sample Label</th>
<th>Well Width (nm)</th>
<th>Exciton Linewidth @15K (meV)</th>
<th>$\tau_R$ (ns)</th>
<th>$\sigma_{eh}$ $(10^{-14}\text{cm}^2)$</th>
<th>$n_{eh}$ $(10^{-19}\text{cm}^3)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>2x5x6</td>
<td>11</td>
<td>4.8</td>
<td>2.2 ± 0.1</td>
<td>4.2</td>
<td>2.4</td>
</tr>
<tr>
<td>2x5x8</td>
<td>15</td>
<td>5.4</td>
<td>1.7 ± 0.1</td>
<td>4.3</td>
<td>2.5</td>
</tr>
<tr>
<td>2x5x10</td>
<td>19</td>
<td>6.2</td>
<td>0.65 ± 0.05</td>
<td>2.7</td>
<td>1.7</td>
</tr>
</tbody>
</table>

Table 4.1. Peak nonlinear absorption cross section, $\sigma_{eh}$, and nonlinear refraction coefficient, $n_{eh}$, for the three 2x5xk samples.

The peak extracted values of $\sigma_{eh}$ and $n_{eh}$ for each sample are shown in Table 4.1. From this table and Fig. 4.7, it can be seen that the peak values of $\sigma_{eh}$ and $n_{eh}$ for the 2x5x6 and 2x5x8 samples are very similar, but that the corresponding values for the 2x5x10 sample are considerably smaller. This is thought to be a consequence of the broader and weaker excitonic absorption profile and shorter carrier life-time in the latter sample, which we believe results from the reduced quantum confinement in the broad wells and the poorer sample quality.

Information on $\sigma_{eh}$ and $n_{eh}$ can also be obtained from the transient grating measurement, as indicated in Eq. (4.16). The consistency of the differential transmission and the transient grating measurement can be used as a check on the measured values of $\sigma_{eh}$ and $n_{eh}$. Fig. 4.8 shows the relationship among all the relevant quantities. There are two sets of values which could be compared in order to check the consistency. Firstly, the $n_{eh}$ calculated from the measured $\sigma_{eh}$ via the Kramers-Kronig transformation and the $n_{eh}$
Fig. 4.7. (a) Wavelength dependence of the nonlinear absorption cross-section, $\sigma_{eh}$. (b) Wavelength dependence of the nonlinear refraction coefficient, $n_{eh}$, for the 2x5xk samples.
Fig. 4.8. Relationships among the measured and the calculated nonlinear absorption cross-sections, nonlinear refraction coefficients, and diffraction efficiencies.

extracted from both differential transmission and transient grating measurements via Eq. (4.16) should be consistent. Secondly, the measured diffraction efficiency and the \( \eta \) calculated from the measured \( \sigma_{eh} \) and the calculated \( n_{eh} \) should be consistent. It is clear from Fig. 4.8, these two conditions of consistency are the same condition expressed in different ways. Therefore, only one of them needs to be checked.

Using the data shown in Fig 4.7, a diffraction efficiency profile was calculated by substituting the \( \sigma_{eh} \) and \( n_{eh} \) obtained from Eq. 4.3 and Eq. 4.8, respectively, into Eq. 4.16, for each of the 2x5xk samples. The measured and calculated data for the 2x5x6 sample
are plotted in Fig. 4.9. For this sample the measured and calculated diffraction efficiency spectra agree very well, both in magnitude and in shape, indicating that the differential transmission and transient grating measurements are consistent. For the other two samples, similarly good agreement between the shapes of the calculated and measured efficiency profiles was obtained, although the magnitudes differed by 25% for the 2x5x8 and 50% for the 2x5x10 sample. The agreement between the shapes of the profiles still allows us to conclude that the measured $\sigma_{eh}$ and $n_{eh}$ are mutually consistent. The discrepancy between the magnitudes of the predicted and calculated efficiencies could be caused by making differential transmission and corresponding transient grating measurements on different areas of the sample, together with the slight spot-to-spot variations in surface quality.

<table>
<thead>
<tr>
<th>Well/Barrier Composition</th>
<th>Well Width (nm)</th>
<th>$\sigma_{eh}$ (10^{-14}\text{cm}^2)</th>
<th>$n_{eh}$ (10^{-19}\text{cm}^3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>This Work</td>
<td>(InAs)$_2$(GaAs)$_5$/GaAs</td>
<td>11</td>
<td>4.2</td>
</tr>
<tr>
<td>Chemla et al. 1984</td>
<td>GaAs/AlGaAs</td>
<td>9.6</td>
<td>7</td>
</tr>
<tr>
<td>Tai et al. 1987</td>
<td>InGaAs/InP</td>
<td>10</td>
<td>2.8</td>
</tr>
</tbody>
</table>

Table 4.2. Comparison of nonlinear optical coefficients for QWs fabricated from different material systems.

To allow easy comparison of the nonlinear optical properties of the InAs/GaAs SPSLS samples with those of other QW materials, the measured nonlinear optical coefficients of 2x5x6 are listed in Table 4.2, along with the measured values for QWs of similar well width fabricated from other frequently studied material systems, GaAs/AlGaAs and InGaAs/InP. It is clear that the results for the (InAs)$_2$(GaAs)$_5$/GaAs
QWs are comparable to what are believed to be the best results obtained from the other material systems, which leads us to conclude that the nonlinear optical properties of QWs containing short-period strained layer superlattice are at least as good as those of QWs made from other conventional material systems.

Fig. 4.9. Measured and calculated diffraction efficiencies for the 2x5x6 sample.
CHAPTER V

CARRIER TRANSPORT IN QUANTUM WELLS COMPOSED OF ALL BINARY InAs/GaAs SHORT-PERIOD SUPERLATTICES OR THE EQUIVALENT TERNARY InGaAs ALLOY

Quantum wells containing SPSLS's may be expected to exhibit higher in-plane carrier mobilities than equivalent InGaAs ternary alloy quantum wells because of their large average indium content and the elimination of alloy disorder scattering in the SPSLS structures (see discussion in §3.1). In this chapter we describe the measurement of the ambipolar diffusion coefficient of all-binary \((\text{InAs})_2(\text{GaAs})_5\) SPSLS quantum wells by time-resolving the differential transmission signal and the efficiency of diffraction from a photo-generated transient grating. The results are compared to those obtained from InGaAs ternary alloy QW's of similar well width and confinement energies.

§ 5.1 Methods, conditions and considerations for measuring and comparing carrier transport properties of all-binary \((\text{InAs})_2(\text{GaAs})_5\) SPSLS and equivalent InGaAs alloy quantum wells

In order to measure the ambipolar diffusion coefficient of the quantum wells, a transient grating was created in the same way as described in chapter IV. However, in this case, consideration was given to the temporal decay of the diffracted signal due to in-plane diffusion and the recombination of photo-generated carriers. The laser was tuned to the
n=1 heavy hole exciton resonance of the sample to be studied, and the two pump pulses were temporally and spatially overlapped in the sample. The interference between the two pulses modulated the intensity distribution observed by the sample and an exciton grating was generated via direct absorption of the pump light. Due to the interaction with LO-phonons in the sample, the exciton grating became a thermalized electron-hole plasma grating on a time scale of a few ps, as discussed in Chapter IV. Once the grating was formed, it could decay via in-well diffusion and carrier recombination. The decay of the grating was monitored by measuring the efficiency with which a weak probe pulse was diffracted from the grating, as a function of time delay of the probe pulse with respect to the pump pulses. The decay rate of the diffraction efficiency is determined mainly by the ambipolar diffusion coefficient of the sample, the grating spacing, and the recombination lifetime of photo-generated carriers. Recall Eqs. (4.15) and (4.14)

\[
\eta = \frac{1}{4} \exp\left(-\alpha d - \sigma_{eh} e^{-t/\tau_G} N_0 d - 2t / \tau_G \right) \left[ \left( \frac{2 \pi \eta_{\text{eh}}}{\lambda} \right)^2 + \left( \frac{\sigma_{\text{eh}}}{2} \right)^2 \right] N_0^2 d^2,
\]

\[
\frac{1}{\tau_G} = \frac{1}{\tau_D} + \frac{1}{\tau_R} = \frac{4 \pi^2 D_a}{\Lambda^2} + \frac{1}{\tau_R},
\]

which give the efficiency, \(\eta\), with which the probe pulse is diffracted from the photo-generated grating as a function of material nonlinearities, grating parameters and delay time of the probe pulse with respect to pump pulses. A few picoseconds (which is the time needed for carriers generated by ultrafast laser pulses to become thermalized) after a grating is written by pump pulses, the efficiency of light diffracted from the photo-generated grating can be expressed as

\[
\eta = \eta_0 \exp\left(-\frac{2t}{\tau_G}\right) = \eta_0 \exp\left[-\left(\frac{8 \pi^2 D_a}{\Lambda^2} - \frac{2}{\tau_R}\right)t\right],
\]

(5.1)
since only the temporal decay of the diffraction efficiency is of interest for measuring the ambipolar diffusion coefficient. In Eq. (5.1), \( \eta_0 \) is a constant determined by the depth of modulation of the photo-generated carrier grating and the material nonlinearities, \( \tau_G \) is the grating decay time, \( D_a \) is the ambipolar diffusion coefficient, \( \Lambda \) is the grating spacing, \( \tau_R \) is the carrier recombination time and \( t \) is the delay time of the probe pulse with respect to the pump pulses.

There are two different ways to extract the ambipolar diffusion coefficient from grating decay measurements. Firstly, one can measure the decay rate of the diffraction efficiency, which is given by

\[
\Gamma = \frac{8\pi D_a}{\Lambda^2} \frac{2}{\tau_R},
\]

for a certain grating spacing. The recombination time, \( \tau_R \), can be measured independently by time resolving the differential transmission signal, since the photo-induced change of optical absorption in the samples studied in this work is mainly due to the bleaching, as evidenced in Fig. 4.5, not the shifting of excitonic absorption peak. As discussed in Chapter IV,

\[
\frac{\Delta T}{T} = e^{-\Delta \alpha d} - 1
\]

\[
\equiv -\Delta \alpha d = -\sigma_{eh}dN_0e^{-\tau_Rs}, \quad \text{for } \Delta \alpha d \ll 1
\]

where \( d \) is the sample thickness, \( N_0 \) is the initial carrier density, and \( \sigma_{eh} \) is the nonlinear absorption coefficient. With knowledge of \( \Gamma \), \( \Lambda \) and \( \tau_R \), the ambipolar diffusion coefficient can be easily extracted using Eq. 5.2. Secondly, one can measure the diffraction efficiency decay rate, \( \Gamma \), at many different grating spacings. If the measured values of \( \Gamma \) are plotted as function of \( 8\pi/\Lambda^2 \), one would expect to obtain a straight line, with slope equal to \( D_a \) and y intercept equal to \( 2/\tau_R \).
The latter method can also give a sense of the quality of the measurements, i.e. if all the data points lie close to a straight line, theory and experiment are in good accord and data taken at different grating spacings yield consistent results. Also, if many data points are taken, the accuracy of the final result can be improved. However, the large amount of time required to perform the alignment for each grating spacing makes it impractical to perform the measurement with a large number of different grating spacings. Finally, if the carrier recombination is not simply exponential, as was found at low carrier density for most of the samples studied in this work, care must be exercised in using the second method. Most of the results reported here in were measured with the first method, and a few were measured with the second method in order to check consistency.

The optically-induced transient grating and the pump-probe techniques employed here have several advantages over the Hall measurement technique which is commonly used to determine the mobility of carriers in semiconductor materials. Firstly, it is not necessary to grow an intentionally doped sample for the mobility measurement. Secondly, the photo-generated carrier density can be easily controlled so that comparison of mobilities in different samples at the same carrier density is possible. However, the optical method also has some drawbacks. Firstly, because photo-excitation creates carriers in pairs, the electron and hole mobilities cannot be measured separately. The ambipolar diffusion coefficients measured with the optical method are dominated by the heavier species and therefore usually provide information on the hole mobility only. Secondly, the grating theory discussed in § 4.1 is valid only at relatively low carrier density.

The temperature at which the ambipolar diffusion coefficients of 2x5xk InAs/GaAs SPSLS QWs and equivalent InGaAs alloy QWs were measured was chosen to be 85 K, since the main purpose of this measurement was to explore the elimination of alloy disorder scattering in ordered all binary SPSLS QWs. At 85 K, the influence of optical
phonon scattering on the carrier mobility is greatly reduced, and one might expect alloy disorder scattering to play an important role in the transport properties of the alloys, as shown in Fig 5.1 (Hayes et al. 1982). In addition, alloy disorder scattering would be completely eliminated in a perfect binary structure, leading to the expectation of faster charge transport in such structures.

Fig. 5.1. Hole mobility as a function of temperature for a GaAsInP alloy. The experimental points are matched to a curve representing a mobility derived by combining the mobility limits imposed by (a) polar optical phonon scattering, (b) non-polar optical and acoustic scattering, (c) alloy scattering, and (d) ionized-impurity scattering. (After Hayes et al., 1982)
§ 5.2 Experimental Configurations and Sample Structures

Three SPSLS samples, identified as 2x5x6, 2x5x8, and 2x5x10, were studied here. The description and characterization of these samples can be found in chapter 3, and will not be repeated here. For comparison with the SPSLS results, transient grating and pump-probe measurements were also conducted on two nearly-equivalent InGaAs alloy MQW structures, one grown in the same MBE machine as the SPSLS structures, and one grown in a Varian Gen II MBE machine at Harry Diamond Laboratories (HDL). By "equivalent" we mean that the width of the well and the absorption energy of the excitonic ground state in the alloy MQW are approximately the same as for the corresponding SPSLS MQW, ensuring comparable confinement energies. Specifically, each ternary MQW sample was grown on a [001]-oriented GaAs substrate and each contains 50 wells composed of In$_{0.14}$Ga$_{0.86}$As and separated by GaAs barriers. The ternary sample grown at HDL has 11-nm-wide wells and is the alloy equivalent of the binary sample 2x5x6, and is designated T11. The schematic diagrams of sample T11 and corresponding 2x5x6 are shown in Fig. 5.2. The ternary sample grown at HRL has 19-nm-wide wells, is designated T19, and is the ternary equivalent of binary 2x5x10. It is interesting to note that, for equal n = 1 heavy hole excitonic absorption energies, the mole fraction of indium in the SPSLS wells was approximately twice that in the corresponding ternary alloy wells.

All optical measurements were performed at temperatures between 85 K and 300K with a synchronously mode-locked Styryl 13 dye laser (Dawson et al 1988) pumped by the stabilized and compressed, frequency-doubled output of a CW mode-locked Nd:YAG laser. The laser system is shown schematically in Fig 5.3. The mode-locked Nd:YAG laser produces pulses of 1.06 µm, 100 ps at repetition rate of 76 MHz. The output from the YAG laser is then compressed, with a fiber-grating compressor, to about 5 ps and
Fig. 5.2. Schematic diagram for the all binary 2x5x6 superlattice and the "equivalent" ternary alloy QWs. Both samples have GaAs barriers. The term "equivalent" is defined to mean that both the binary and ternary samples have the same well and barrier width, and that the In concentration of the ternary well has been adjusted so the n= 1 hh excitonic peaks are almost identical.
frequency doubled to 532 nm. These green pulses are used to pump the dye laser. The dye laser produces pulses of approximately 1 ps duration which are tunable from 910 to 990 nm, allowing optical interrogation of the ground state excitonic transitions in each sample. The dye laser incorporated a cavity dumper that allowed the repetition rate of the laser to be varied between 1 MHz and 76 MHz, although most of the results discussed herein were taken at a repetition rate of 1 MHz. As shown in Fig. 5.4, the beam from the dye laser was split into three parts to form two pump beams and a weak probe beam. Two identical lenses were used to focus the two s-polarized pump pulses onto the same spot on the sample. In order to change the period of the grating, the angle between the beams at the sample site could be adjusted in the range 5 to 25 degrees. The fluence ratio of the two pump pulses was 20:1. The decay of the grating was monitored by measuring the diffraction efficiency of the time delayed, p-polarized, probe pulse, which was focused onto the optically excited region of the sample using a third lens. This resulted in a probe diameter at the sample that was roughly three times smaller than that of the pumps, ensuring that the probe was diffracted by an approximately uniform grating. The probe fluence was set to be a factor of 35 lower than the weaker pump. Comparing the experimental setup shown in Fig 4.3 and Fig 5.4, one might notice that there are two main differences. Firstly, in the latter case, two pump beams and the probe beam were focused onto the sample by three different lenses instead of a single lens. This made it easier to adjust the grating spacing and laser spot size independently. Secondly, in the latter case, the ratio between intensities of the two pump pulses was 20:1 instead of 1:1, so that the modulation of carrier density was somewhat smaller than the average carrier density, and the effect of the carrier density dependence of carrier recombination on the transient grating decay is reduced.
Fig. 5.3. Schematic diagram for the laser and control system. The output from the Nd:YAG laser is stabilized, compressed and frequency doubled, and then used as the pump source of the synchronously mode-locked Styryl 13 dye laser, which produces tunable (910-990 nm) sub-picosecond pulses. The repetition rate of these pulses either equal to the 76 MHz repetition rate of the YAG laser or able to be adjusted between 148 kHz to 9.5 MHz via a cavity dumper. The wavelength of the dye laser is tuned via a birefringent filter operated by a computer controlled stepper motor.
Fig. 5.4. Schematic diagram of the experiment apparatus for the photoexcited transient grating decay measurements. The following abbreviations are used: BS = beamsplitter, C = chopper, D = detector, and P = periscope. This apparatus can also be used for differential transmission measurements by removing the BS2.

In addition to the grating decay measurements, the recombination time for each sample was independently determined by blocking one pump beam and measuring the time dependence of the pump-induced change in the absorption coefficient. Independent knowledge of the recombination time allows us to extract the ambipolar diffusion coefficient from the grating decay measurements, as discussed in §5.1.
§ 5.3 Carrier Lifetime Measurements

We start by looking at the \( \Delta T \) spectra of the sample 2x5x6 taken at 85K at different carrier densities. The spectra shown in Fig 5.5 are evidently quite different when compared to those obtained from the same sample at room temperature, which are shown in Fig. 4.5. At low carrier densities, the spectra of Fig. 5.5 have large negative peaks on high energy side, an indication of exciton blue shift, which is attributed to the exciton-exciton interaction (Schmitt-Rink et al. 1985). This blue shift occurs only at low temperature, because the percentage of carriers in the exciton states at low temperature equilibrium is larger then that in the room temperature situation. This blue shift of the exciton peak made the interpretation of transient grating decay results somewhat more difficult. As the carrier density decreases via recombination, the positive peak of the \( \Delta T \) signal remains at the same energy and decreases in amplitude, whereas the negative peak of the \( \Delta T \) signal moves to a lower energy as well as decreasing in amplitude. If a significant amount of photo-generated carriers recombine on the same time scale in which the grating is washed out via in-well diffusion, Eqs. (4.15) and (5.1) are no longer valid, since these equations are obtained under the assumption that the differential transmission spectrum is linearly proportional to the carrier density. More seriously, the definitions of \( \sigma_{eh} \) and \( n_{eh} \) given in chapter IV are not valid in the situation involving shifting of exciton peak. Fortunately, if the carrier lifetime is much longer than the grating decay time, as the case in this investigation, the effects of exciton peak shifting on the diffraction efficiency can be neglected, because the average carrier density does not undergo a significant change before the grating has been washed out, mainly via in-well diffusion. In this limit, Eq. 4.24 and hence Eq. (5.1) are still valid, although one cannot extract \( \sigma_{eh} \) and \( n_{eh} \) from the result of degenerate differential transmission measurement. In fact, \( \sigma_{eh} \) and \( n_{eh} \) are not
defined for optical nonlinearities involving the shifting of excitonic absorption peaks (McCallum et al. 1992).

![Diagram showing differential transmission spectra for different values of N.](image)

Fig. 5.5. 85 K differential transmission spectra of the 2x5x6 sample over a range of fluence.

The results of measuring the temporal dependence of the pump-induced change in probe transmission, $\Delta T$, for all five samples are shown in Fig. 5.6. These data were obtained using a repetition rate of 1 MHz and a pump fluence that produced a sheet charge density of $10^{11}$ cm$^{-2}$ in the quantum wells at 85 K. The central wavelength of the laser was tuned to the n=1 heavy-hole exciton peak of each sample. For delays beyond approximately 30 ps, a straight line is a good fit to the decay curve for each sample on these semilogarithmic plots, indicating an exponential decay. The carrier lifetimes
extracted from these fits ranged from 450 ps in the ternary T19 to 2.2 ns in the binary 2x5x6. The results are summarized in Table 5.1. We note that binaries 2x5x6 and 2x5x8 have substantially longer lifetimes than binary 2x5x10 and the two ternaries. Furthermore, there is a one to one correspondence between the measured lifetimes and the sharpness of the excitonic absorption resonances, i.e. narrow excitonic absorption resonances are associated with long lifetimes.

<table>
<thead>
<tr>
<th>Sample Label</th>
<th>Well Composition</th>
<th>Well Width (nm)</th>
<th>Exciton Linewidth @15K (meV)</th>
<th>(\tau_R) (ns)</th>
<th>(D_a) (cm²/s)</th>
<th>(\mu_h) (cm²/Vs)</th>
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</thead>
<tbody>
<tr>
<td>2x5x6</td>
<td>(InAs)₂(GaAs)₅</td>
<td>11</td>
<td>4.8</td>
<td>2.2 ± 0.1</td>
<td>73 ± 5</td>
<td>5300</td>
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<tr>
<td>2x5x8</td>
<td></td>
<td>15</td>
<td>5.4</td>
<td>1.7 ± 0.1</td>
<td>73 ± 5</td>
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<tr>
<td>2x5x10</td>
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<td>15.0</td>
<td>0.45 ± 0.05</td>
<td>44 ± 3</td>
<td>3200</td>
</tr>
</tbody>
</table>

Table 5.1 Growth parameters and measured properties for samples 2x5x6, 2x5x8, 2x5x10, T11 and T19: the composition and widths of the quantum wells, the measured excitonic absorption line width at 15 K, the lifetime for carrier recombination at 85 K, the measured ambipolar diffusion coefficient, and the estimated lower limit for the hole mobility.
Fig. 5.6. Pump-induced change in transmission as a function of probe delay for each of the 2x5xk and the "equivalent" ternary alloy samples discussed in the text. Each set of data has been arbitrarily scaled as indicated for clarity.

The initial rapid decay shown for some of these samples appears in all samples at sheet charge densities below \( \sim 10^{11} \text{ cm}^{-2} \) and becomes more prominent as the carrier density is decreased, as shown in Fig 5.7 for sample 2x5x6. There is also some indication that the decay time associated with the transient increases with decreasing carrier densities. The transient feature is insignificant in 2x5x6 as well as in all other samples at densities above a few times \( 10^{11} \text{ cm}^{-2} \). These characteristics may indicate the presence of a saturatable trapping level in these structures. The recombination dynamics of the
ternary Tl1 are even more complicated: this sample not only shows the fast transient response, but the time constant associated with the slower decay feature is dependent upon laser repetition rate, varying from approximately 1 ns at 10 MHz to 1.5 ns at 76 MHz. We also observe an anomalous behavior in the transport properties of this sample, as discussed below.

Fig. 5.7. Pump-induced change in transmission as a function of probe delay for the 2x5xk sample over a range of pump fluences.

Although these structures clearly display interesting and complicated recombination dynamics, we emphasize that the transport properties of these materials were measured in a regime that minimized the effects of the fast trapping process. All the
data were obtained at a sheet charge density of $\sim 10^{11} \text{ cm}^{-2}$, a density at which the slower recombination process dominates. Although the faster decay process is even less pronounced at higher sheet charge densities, the nonlinear response of the material begins to saturate (see §4.3) at densities above $10^{11} \text{ cm}^{-2}$. Hence, densities above this value were avoided. The carrier dynamics in the three SPSLS's and the ternary T19 were found to be independent of the laser repetition rate, while the dynamics in ternary T11 were found to be independent of repetition rate only for rates below $\sim 10 \text{ MHz}$. Hence, by operating the laser at a repetition rate of 1 MHz, we ensured that the results for all samples, including T11, were not influenced by this parameter.

§ 5.4 Diffusion Measurements

The transient grating measurements were used to determine the ambipolar diffusion coefficients. The decay of the grating diffraction efficiency for each sample is shown in Fig. 5.8 for a grating period of 6 μm and an average photo-generated sheet-charge density of $10^{11} \text{ cm}^{-2}$. After the initial transient feature, which again can be attributed to the fast trap-related recombination, the decay in each case follows a straight line on this semilogarithmic plot, indicating that the decays are exponential as predicted by Eq. (5.1). The 6-μm grating spacing provided an overall decay time for the diffraction efficiency for each sample that was much longer than the decay time associated with the fast trapping process, thus ensuring that the latter did not influence the extraction of the ambipolar diffusion coefficients. The ambipolar diffusion coefficient for each sample was extracted, using the measured decays of the diffraction efficiencies, the carrier lifetimes, and Eq. (5.2). The coefficients measured at 85 K are summarized in Table 5.1.
Fig. 5.8. Diffraction efficiency as a function of probe delay for each of the 2x5xk and the "equivalent" ternary alloy samples discussed in the text. Each set of data has been arbitrarily scaled as indicated for clarity.

Additional measurements at other grating periods were used to verify that the grating decay rates determined using the above procedure depended linearly on $1/\Lambda^2$, as predicted by Eq. (5.2). A representative result for sample 2x5x6 is shown in Fig. 5.9. These data were taken at a repetition rate of 76 MHz, and the diffusion coefficient of 68 cm$^2$/s obtained from a linear fit to the data is in good agreement with the value obtained at 1 MHz by simply using the measured carrier lifetime, the known grating period, and Eq. (5.2). Similar agreement was found for samples 2x5x6, 2x5x8, and T19. Measurements at 76 MHz on the ternary T11, however, resulted in a diffusion coefficient
of only 50 cm$^2$/s as opposed to the value of 68 cm$^2$/s obtained at 1 MHz. This anomalous behavior, like the repetition-rate dependence of the carrier lifetime in this sample, is not presently understood.

![Graph showing grating decay rate versus $4\pi^2/\Lambda^2$ for the 2x5x6 sample.](image)

Fig. 5.9. Grating decay rate versus $4\pi^2/\Lambda^2$ for the 2x5x6 sample. The diffusion coefficient can be obtained from the slope of the linear fit through the data and the intercept of this fit with the vertical axis gives the carrier recombination time.

Because of the exciton blue shift at 85K, one might believe that the diffusion at low temperature is exciton diffusion instead of ambipolar diffusion. In order to clarify this, the diffusion coefficients were measured as function of temperature. Fig. 5.10 shows
the result for sample 2x5x6, which is typical for all the samples. The data points in Fig. 5.10 can be fitted by $T^{-1.1}$, a typical result for diffusion restricted by phonon scattering (Seeger 1985). Clearly, there is no evidence of a transition from ambipolar diffusion at room temperature to exciton diffusion at low temperature, since exciton diffusion is an order of magnitude slower than ambipolar diffusion (Hillmer et al. 1989). We argue that although the exciton density is high enough to cause a blue shift, the majority of photo-generated carriers are still in unbound states at as low as 85K.

![Graph showing ambipolar diffusion coefficient plotted as a function of temperature.](image)

**Fig. 5.10.** Measured ambipolar diffusion coefficient plotted as a function of temperature.
§ 5.5 Discussion

Examination of Table 1 reveals an obvious correspondence between the carrier lifetimes and the measured diffusion coefficients, i.e., the shorter the lifetime the smaller the diffusion coefficient. The shorter lifetimes and smaller diffusion coefficients measured in the binary 2x5x10 and the ternary T19 may indicate that these samples have a higher defect density than the other three samples. This conclusion is also consistent with the poorer linear optical properties of these two samples.

From the data contained in Table 1, we can compare the transport properties of the SPSLS's and the equivalent ternary alloy MQWs. Comparing the 10-period SPSLS (sample 2x5x10) with the ternary MQW containing 19-nm wells (sample T19), which was grown in the same MBE machine, we see that the SPSLS has an ambipolar diffusion coefficient that is approximately a 20% larger. However, the carrier lifetime for the SPSLS is also longer than that of the ternary. Given the general trend of increased diffusion coefficient with increased carrier lifetime observed in all the samples, this improved diffusion in the SPSLS sample relative to the ternary cannot be unambiguously attributed to the binary nature of the SPSLS. The second ternary sample (T11), which has 11-nm wells and was grown in the Varian MBE machine at HDL, is to be compared with the 6-period SPSLS (2x5x6) grown in the Perkin Elmer MBE machine at HDL. We find that the diffusion coefficient for the ternary is almost as large as that of the binary, in spite of the considerably longer lifetime in the binary. Although further optimization of growth conditions may lead to realization of the predicted improvement in transport properties in the binary SPSLS MQWs, the present results imply that such an improvement has not yet been achieved. This may indicate that indium-rich island formation and/or the introduction of large numbers of interfaces tend to offset any enhancement in mobilities that may be
associated with the reduction in alloy scattering, larger average indium content, and larger local strain in the SPSLS. It should be emphasized, however, that the carrier lifetimes, diffusion coefficients, and optical properties of the best SPSLSs are at least as good as the better of the two ternaries.

We now relate the measured ambipolar diffusion coefficients to the hole mobilities in these structures. For equal electron and hole densities, the ambipolar diffusion coefficient is given in terms of the mobilities by (see e.g. Seeger 1989):

$$D_a = \frac{2k_B T \mu_n \mu_p}{e(\mu_n + \mu_p)}.$$  

(5.3)

Here, $\mu_n$ and $\mu_p$ are the electron and hole mobilities, respectively, $k_B$ is Boltzmann's constant, $e$ is the magnitude of the fundamental electronic charge, and $T$ is the carrier temperature. Note that the measured diffusion coefficients are significantly smaller than would be expected for purely electronic diffusion in these materials at this temperature, indicating that the ambipolar diffusion is likely hole-dominated. Hence, in spite of the expected light-hole nature of the in-plane mass of the valence band in these strained structures, we assume that the electron mobility is still large compared to the hole mobility. In this case, Eq. (5.3) reduces to $D_a \approx 2k_B T \mu_p/e$, which allows us to estimate a lower limit for the hole mobility in each sample. These values are also shown in Table 1 and are all comparable to previously reported (Fritz et al. 1986) hole mobilities for strained InGaAs alloy wells of similar geometry, well width, and sheet-charge density at ~80K.

In summary, ambipolar diffusion coefficient in $(\text{InAs})_2(\text{GaAs})_5$ SPSLS MQWs grown on GaAs substrates have measured for the first time, and the results have been compared to those obtained in equivalent InGaAs alloy MQWs. We find that the
ambipolar transport properties of the best all-binary structures are comparable to those of the better alloy MQW, while the carrier lifetimes in these binaries are approximately twice as long as those in the alloy. The absence of a clear enhancement in the transport properties of the SPSLS's relative to the ternary alloy materials may indicate that the potential improvements in transport in the SPSLS's that one might expect as a consequence of reduced alloy scattering, large local strain, and higher average indium content may be offset by perturbations in the crystalline potential associated with interface roughness and island formation. It should be emphasized, however, that both TEM and X-ray diffraction measurements demonstrate the existence of the binary structure in the SPSLS MQWs, and continued optimization of the growth parameters may allow the realization of improved ambipolar transport in the SPSLS's.

It should be noted that, since the measured ambipolar diffusion coefficients are likely to be dominated by holes, the electron transport properties of the reported samples have yet to be determined. Improved electron mobilities have been demonstrated by incorporating SPSLS channels in high electron mobility transistors (HEMT's) (Hasenberg 1990, Onda 1990). Again, the SPSLS samples can accommodate, without the formation of misfit dislocations, unusually high indium concentrations (>30% for each structure) with respect to the high quality InGaAs alloy quantum wells of the same well width (11-19 nm) (Fritz et al. 1985, Anderson et al. 1989, Weng 1989, Elman et al. 1989). The improved electron transport observed in the SPSLS's may therefore be a consequence of a lighter electron mass associated with the more InAs-like nature of the conduction band. Furthermore, the measurements reported here were conducted at sheet charge densities considerably below those appropriate to practical high-speed electronic devices. Again the limitation of the present measurements is the saturation of the nonlinearity that produces the grating. Although we have seen no clear evidence of any density
dependence of the diffusion coefficient in any of these samples for densities up to $10^{11}$ cm$^{-2}$, we note that alloy disorder scattering may play a more important role in the ternaries at still higher densities (Ogale and Madhukar 1984).
CHAPTER VI

SUMMARY

The ternary alloy InGaAs has been the subject of intense investigation recently because its high electron mobility, large Γ-L valley separation, and small band gap make it attractive for high-speed electronic device and optoelectronic device applications, such as the high electron mobility transistor and the diode laser. The properties of InGaAs grown on GaAs substrates are of particular interest for device applications. The difference between the lattice constants of InGaAs and GaAs place an upper limit on the thickness of InGaAs which can be grown lattice-matched to a GaAs substrate. However, the lattice mismatch between InGaAs and a GaAs substrate can be entirely accommodated as elastic strain, if for a given indium mole fraction, x, the InGaAs layer is thinner than the critical thickness for dislocation formation. In addition to the attractive intrinsic properties of InGaAs, the introduction of strain results in new features that are important for device design. For example, the strain lifts the degeneracy in the valence band providing a large splitting between light- and heavy-hole bands and producing an uppermost band with a light-hole-like in-plane effective mass. Strained InGaAs holds promise for high mobility p-channel transistors and low threshold lasers.

In this work, all-binary (InAs)$_m$(GaAs)$_n$ short-period strained-layer superlattices (SPSLS's) have been investigated as a potential alternative to the random ternary InGaAs strained alloys. The all-binary materials offer the prospect of improved electron and hole transport as the result of reduced alloy scattering and larger average indium content. For
example, the ordered nature of superlattices has led to predictions of a substantial reduction or elimination of alloy scattering and, thus, to their proposed use as the channel material in modulation-doped field effect transistors in order to improve the speed of these devices. In addition, a considerably larger average indium content can be accommodated in quantum wells composed of SPSLS's than in wells of the same width composed of InGaAs alloy material. This too has led to the expectation of higher electron mobilities, because of the observed increase in electron mobility with increasing indium content in the ternary alloys. In addition, the all-binary materials offer potential advantages in material growth and wavelength reproducibility.

A series of ordered all-binary InAs/GaAs short-period strained layer superlattice (SPSLS) quantum well (QW) samples were grown on GaAs substrate by either conventional molecular beam epitaxy (MBE) or migration enhanced epitaxy (MEE). Optical characterization shows that these are of very good quality structures. Clearly-resolved excitonic absorption peaks have observed at room temperature in all the structures. In optimized samples at 15 K, photoluminescence (PL) and excitonic linewidths are below 10 meV, with the PL Stokes-shifted by less than 1 meV. Another evidence of high sample quality is the long PL lifetime (~1.5 ns) for optimized samples at 15 K.

Nonlinear optical properties of the (InAs)$_2$(GaAs)$_5$ SPSLS QWs were measured. Specifically, picosecond differential transmission and transient grating techniques were used to measure the nonlinear absorption cross-section, $\sigma_{eh}$, and the refractive index change per photo-generated electron-hole pair, $n_{eh}$, near the $n = 1$ heavy-hole excitonic resonance. By applying a Kramers-Kronig analysis, consistency between the results of the differential transmission and transient grating measurements was demonstrated. The
measured $\sigma_{eh}$ and $n_{eh}$ are comparable to those of high quality GaAs/AlGaAs and InGaAs/InP QWs.

The in-well ambipolar diffusion coefficients and carrier lifetimes in $(\text{InAs})_2(\text{GaAs})_5$ SPSLS QWs were measured using picosecond optically-induced transient grating and pump-probe techniques. The results obtained in the SPSLS's are compared to those obtained in InGaAs alloy quantum wells of comparable well width and confinement energies. It was found that the ambipolar diffusion coefficients and carrier lifetimes in the SPSLS's are at least as large as those measured in the equivalent alloys. The absence of a dramatic improvement in these properties, however, may indicate that, e.g., interface roughness and island formation in the SPSLS's may be offsetting their possible advantages. This may occur in spite of the fact that optical measurements of the SPSLS's indicate high material quality and X-ray and transmission electron microscopic measurements demonstrate the binary nature of these structures. The results, therefore, indicate that further optimization of the growth conditions for the SPSLS's may be required to realize the full potential of these novel structures.

In conclusion, growth of high quality quantum wells composed of ordered all-binary InAs/GaAs short-period strained layer superlattices has been demonstrated. These structures might be used to replace the ternary alloy InGaAs/GaAs structures in device applications resulting in some advantages related to growth, to the elimination of alloy scattering and to the accommodation of a higher average In content. The linear and nonlinear optical properties of these all-binary structures are comparable to those of high quality ternary alloy quantum well samples. The results of the in-well transport measurements do not show clear evidence of the elimination of alloy disorder scattering at relatively low carrier densities, and further investigations of this effect are necessary.
APPENDIX I

SOLUTION OF THE 2D DIFFUSION EQUATION WITH GAUSSIAN INITIAL CONDITIONS
In a pump-probe experiment in which it is assumed that the pump pulse has a Gaussian spatial intensity distribution, and that the duration of the pump pulse is much shorter than the lifetime of photo-generated carriers, the carrier density in quasi 2D systems, such as the QW samples studied here, after the ultrafast photo-excitation can be described by

\[
\frac{\partial N(x,y,t)}{\partial t} + \frac{N(x,y,t)}{\tau_R} - D_a \nabla^2 N(x,y,t) = 0,
\]
(A1.1a)

with

\[
N(x,y,t = 0) = \frac{\alpha F(x,y)}{h\nu} = N_0 \exp \left( -\frac{x^2 + y^2}{\omega_0^2} \right)
\]
(A1.1b)

where \(N\) is number density of photo-generated electron-hole pairs, \(N_0\) is the number density at the center of the laser spot, \(\tau_R\) is the recombination lifetime of the photo-generated electron-hole pairs, \(D_a\) is the ambipolar diffusion coefficient of the host material, \(F\) is the fluence of the pump pulse, \(\omega_0\) is HW1/eM intensity of the pump pulse, \(\alpha\) is the linear absorption coefficient of the material, and \(h\nu\) is the photon energy of the excitation laser.

Assuming that the recombination lifetime is not a function of carrier density, we have

\[
N(x,y,t) = U(x,y,t) e^{-t/\tau_R},
\]
(A1.2)

and Eq. (A1.1) becomes

\[
\frac{\partial U(x,y,t)}{\partial t} - D_a \nabla^2 U(x,y,t) = 0.
\]
(A1.3)

Assume all variables in \(U(x,y,t)\) are separable, so that

\[
U(x,y,t) = X(x)Y(y)T(t),
\]
(A1.4)
$U$ can be represented by a product of three single variable functions. Substituting Eq. (A1.4) into Eq. (A1.3) and dividing it by $D a U$, we have

$$\frac{T'}{D a T} = \frac{X''}{X} + \frac{Y''}{Y},$$  \hspace{1cm} (A1.5)

since both sides of the equation have different variable(s), they can be equal only if they are both equal to a constant, e.g.

$$\frac{T'}{D a T} = \frac{X''}{X} + \frac{Y''}{Y} = 2\omega^2.$$   \hspace{1cm} (A1.6)

Considering the symmetry of the problem, we also have

$$\frac{X''}{X} = \frac{Y''}{Y} = \omega^2.$$   \hspace{1cm} (A1.7)

The solutions for Eq. (A1.6) and (A1.7) are

$$T(t) \propto e^{-2\omega^2 D a t},$$

$$X(x) \propto e^{i\omega x},$$

$$Y(y) \propto e^{i\omega y}.$$   \hspace{1cm} (A1.8)

Therefore the solution corresponding to constant $\omega$ is

$$U(x, y, t, \omega) = A(\omega) e^{-2\omega^2 D a t} e^{i\omega (x+y)},$$   \hspace{1cm} (A1.9)

Considering the independence of $X(x)$ and $Y(y)$, and again the symmetry of the problem, it is safe to write

$$U(x, y, t, \omega) = X(x, t, \omega) Y(y, t, \omega),$$   \hspace{1cm} (A1.10)

and

$$X(x, t, \omega) = A_x(\omega) e^{-\omega^2 D a t} e^{i\omega x},$$

$$Y(y, t, \omega) = A_y(\omega) e^{-\omega^2 D a t} e^{i\omega y}.$$/

(A1.11)

The final solution is the superposition of all the possible solutions.
\[ X(x,t) = \int_{-\infty}^{\infty} A_x(\omega) e^{-\omega^2 D_a t} e^{i\omega x} d\omega \]  

(A1.12)

where \( A_x(\omega) \) is the Fourier transform of \( X(x,t = 0) = \sqrt{N_0} e^{-x^2/2\sigma_0^2} \)

\[ A_x(\omega) = \frac{\sqrt{N_0}}{2\pi} \int_{-\infty}^{\infty} e^{-\xi^2/2\omega_0^2} e^{-i\omega \xi} d\xi. \]  

(A1.13)

Substituting Eq. (A1.13) into Eq. (A1.12) gives

\[ X(x,t) = \int_{-\infty}^{\infty} \sqrt{N_0} e^{-\xi^2/2\omega_0^2} \left[ \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-\omega^2 D_a t} e^{i\omega(x-t)} d\omega \right] e^{i\omega t} d\ξ. \]  

(A1.14)

Using the integration formula \( \int_{-\infty}^{\infty} e^{-\xi^2} e^{\rho i\xi} d\xi = \frac{\sqrt{\pi}}{\rho} e^{\rho^2/4} \), Eq. (A1.14) becomes

\[ X(x,t) = \int_{-\infty}^{\infty} \sqrt{N_0} e^{-\xi^2/2\omega_0^2} \left[ \frac{1}{2\sqrt{D_a \pi t}} e^{(x-\xi)^2/(4D_a t)} \right] e^{i\omega t} d\xi \]

\[ = \frac{\sqrt{N_0}}{\sqrt{1 + 4D_a t / \omega_0^2}} e^{-x^2/(2\sigma_0^2 + 4D_a t)} \]  

(A1.15)

In the same way we can get

\[ Y(y,t) = \frac{\sqrt{N_0}}{\sqrt{1 + 4D_a t / \omega_0^2}} e^{-y^2/(2\sigma_0^2 + 4D_a t)} \]  

(A1.16)

Combining Eq. (A1.15), Eq. (A1.16), Eq. (A1.10) and Eq. (A.1.2) gives

\[ N(x,y,t) = \frac{N_0}{1 + 4D_a t / \omega_0^2} \exp \left( \frac{x^2 + y^2}{\omega_0^2 + 4D_a t} \right) e^{-t/\tau_R}. \]  

(A1.17)
APPENDIX II

EFFICIENCY OF LIGHT DIFFRACTED FROM A PHOTO-GENERATED TRANSIENT THIN GRATING
In this appendix, a theory of dynamic free-carrier gratings induced by ultra-short laser pulses in semiconductor QW structures will be laid out along the lines of H. J. Eichler's work (Eichler et al. 1981, Eichler et al. 1986).

For simplicity, the efficiency with which light is diffracted from a photo-generated transient thin grating will be calculated under the plane wave approximation, that is the pump beams will be assumed to be plane waves of uniform intensity. In addition, diffusion due to the finite laser spot size will be neglected. In our transient grating measurement, two pump pulses separated by an angle $\theta$ were temporally and spatially coincident on the sample. The interference between the two pulses modulated the intensity across the sample. The direct absorption of the two interfering pump pulses produced a transient free-carrier grating, as described by Eq. 4.12.

$$N(x,t) = N_0 \left[ 1 + e^{-t/\tau_D} \cos \left( \frac{2\pi x}{\Lambda} \right) \right] e^{-t/\tau_R} , \quad (4.12)$$

where $N$ is number density of photo-generated electron-hole pairs, $N_0$ is the number density at the center of the laser spot, $\tau_R$ is the recombination lifetime of the photo-generated electron-hole pairs, and $\tau_D$ is the diffusive decay time. As we have discussed in §4.1, the change of excitonic absorption and the accompanying change of refractive index are the result of phase-space filling of the photo-generated carriers. When the carrier density is sufficiently low, the change of absorption coefficient, $\Delta \alpha$, and the change of refractive index, $\Delta n$, are linearly proportional to the carrier density,

$$\Delta \alpha = \sigma_{eh} N, \quad (4.3)$$

$$\Delta n = n_{eh} N, \quad (4.6)$$

where, $\sigma_{eh}$ is defined as the nonlinear absorption cross section, and $n_{eh}$ is defined as the nonlinear refraction coefficient. Therefore, the free-carrier grating can be described in
terms of a transmission grating and a phase grating. Using Eqs. 4.3, 4.6 and 4.12, the photo-generated grating can be described by a spatially periodic amplitude transmittance, $t_a$, where

$$t_a = \exp\left[-(\alpha + \Delta \alpha)d\right] \exp\left[\frac{i2\pi(n + \Delta n)d}{\lambda}\right]$$

$$= \exp\left[-\alpha d + \frac{i2\pi nd}{\lambda} - \left(\sigma_{eh}N_0d - \frac{i2\pi n_{eh}N_0d}{\lambda}\right)\left(1 + e^{-i\pi R_x} \cos\frac{2\pi x}{\Lambda} e^{-i\pi R_x}\right)\right],$$  \hspace{1cm} (A2.1)

and $\alpha$ is the linear absorption coefficient of the sample, $n$ is the linear refractive index of the sample, $\lambda$ is the laser wavelength, and $d$ is the sample thickness.

For simplicity, we will treat the grating as being thin by neglecting the effects of finite grating thickness, and will discuss in appendix III the conditions under which this approximation is valid. A thin photo-generated carrier grating with fringe spacing $\Lambda$ is shown schematically in Fig A2.1. The grating spacing, $\Lambda$, is determined by the angle between two the pump beams and is given by

$$\Lambda = \frac{\lambda}{2\sin(\theta / 2)}. \hspace{1cm} (A2.2)$$

An incident probe light wave, propagating in a direction perpendicular to the grating surface, is described by

$$E_i = \frac{A_i}{2} e^{i(\omega t - kz)} + c.c. \hspace{1cm} (A2.3)$$

The field strength just behind the grating at $z = 0$ is

$$E = \frac{A_i}{2} t(x) e^{i\omega t} + c.c. \hspace{1cm} (A2.4)$$

The amplitude of the light wave just behind the grating, $A_i t(x)$, can also be described by a superposition of plane waves (each of which corresponds to a diffracted partial wave) with amplitudes $A_m$. 
Because $t(x)$ is periodic with $\Lambda$, Eq. A2.5 corresponds to a Fourier series development of $t(x)$ if $k_{mx}$ is chosen as

$$k_{mx} = \frac{m2\pi}{\Lambda}, \quad m = 0, \pm 1, \pm 2, \ldots$$

The amplitude $A_m$ of a diffracted partial wave is given by the $m$th Fourier coefficient of the transmittance $t(x)$:

$$A_m = \frac{A_0}{\Lambda} \int_0^\Lambda t(x)e^{im2\pi x/\Lambda} dx$$

Fig. A2.1. Grating geometry: a thin grating induced by two pump beams separated by an angle $\theta$. 

$$A_1 t(x) = \sum_m A_m e^{-ik_{mx}x} \quad \text{(A2.5)}$$
The direction of the \( m \)th order diffraction is determined by

\[
\sin \phi_m = \frac{k m}{k} = \frac{m \lambda}{2 \pi} = \frac{m \lambda}{\lambda} = 2m \sin \left( \frac{\theta}{2} \right)
\]  
(A2.8)

Substituting Eq. A2.1 into Eq. A2.7 gives

\[
A_m = \frac{A_1^A}{\Lambda} \exp \left[ -\alpha d + i \left( \frac{i \sigma_{eh}}{2} + \frac{2 \pi n_{eh}}{\lambda} \right) \right] N_0 d \left( 1 + e^{-i2\pi x} \cos \frac{2\pi x}{\Lambda} \right) e^{-im2\pi x/\Lambda} dx,
\]  
(A2.9)

where we neglected the term \( e^{i2\pi n/\lambda} \) in Eq. A2.1 since it only induces a constant phase shift. Making

\[
\phi = \left( \frac{2\pi n_{eh}}{\lambda} + \frac{i \sigma_{eh}}{2} \right) N_0 d
\]  
(A2.10)

Eq. A2.9 becomes

\[
A_m = \frac{A e^{-ad/2}}{\Lambda} \exp \left( i \phi e^{-i2\pi \xi} \right) \left[ \exp \left( i \phi e^{-i2\pi \xi} \cos \frac{2\pi x}{\Lambda} \right) + \frac{im2\pi x}{\Lambda} \right] dx.
\]  
(A2.11)

Using the dimensionless variable

\[
\xi = 2\pi x/\Lambda
\]  
(A2.12)

we have

\[
A_m = A e^{-ad/2} \exp \left( i \phi e^{-i2\pi \xi} \right) \left[ \frac{1}{2\pi} \int_{0}^{2\pi} \exp \left( i \phi e^{-i2\pi \xi} \cos \xi \right) \cos (m\xi) d\xi \right]
\]

\[
= A e^{-ad/2} \exp \left( i \phi e^{-i2\pi \xi} \right) \left[ \int_{m\xi}^{m\xi} \phi e^{-i2\pi \xi} \right].
\]  
(A2.13)
where $J_m$ is the $m$th order Bessel function defined in the square bracket in Eq. A2.13. For $|\phi e^{-t/\tau_\phi}| \ll 1$ the following approximations are valid

$$J_0(\phi e^{-t/\tau_\phi}) = 1,$$

$$J_1(\phi e^{-t/\tau_\phi}) = J_{-1}(\phi e^{-t/\tau_\phi}) \equiv \frac{1}{2} \phi e^{-t/\tau_\phi}. \quad (A2.14)$$

The efficiency for the first diffracted order is

$$\eta = \left| \frac{A_{m=1}}{A_i} \right|^2$$

$$= \exp\left(-\alpha d - \sigma_{eh} e^{-t/\tau_e} N_0 d\right) \left| J_1(\phi e^{-t/\tau_\phi}) \right|^2$$

$$\approx \frac{1}{4} \exp\left(-\alpha d - \sigma_{eh} e^{-t/\tau_e} N_0 d\right) \left| \phi e^{-t/\tau_\phi} \right|^2 \quad (|\phi e^{-t/\tau_\phi}| \ll 1)$$

$$= \frac{1}{4} \exp\left(-\alpha d - \sigma_{eh} e^{-t/\tau_e} N_0 d - 2t/\tau_\phi\right) \left[ \left(\frac{2\pi n_{eh}}{\lambda}\right)^2 + \left(\frac{\sigma_{eh}}{2}\right)^2 \right] N_0^2 d^2. \quad (A2.15)$$

This equation describes the first order diffraction efficiency of a transient photo-generated thin grating as a function of material nonlinearity, photo-generated carrier density, and time. Care should be taken not to use this equation at $t = 0$. As we have discussed in §2.3, it takes a few picoseconds for the photo-generated carriers to be thermalized, and $\sigma_{eh}$ and $n_{eh}$ have different values before the thermalization of the carriers.

When $t \ll \tau_\phi$, Eq. A2.15 can be simplified as

$$\eta \approx \frac{1}{4} \exp\left(-\alpha d - \sigma_{eh} N_0 d\right) \left[ \left(\frac{2\pi n_{eh}}{\lambda}\right)^2 + \left(\frac{\sigma_{eh}}{2}\right)^2 \right] N_0^2 d^2. \quad (A2.16)$$

If the carrier density is low enough so that $\Delta \alpha = \sigma_{eh} N_0 \ll \alpha$, we have

$$\eta \approx \frac{1}{4} e^{-\alpha d} \left[ \left(\frac{2\pi n_{eh}}{\lambda}\right)^2 + \left(\frac{\sigma_{eh}}{2}\right)^2 \right] N_0^2 d^2. \quad (A2.17)$$
Note again that all the results in this section were obtained under the plane wave approximation, which assumes plane wave fronts and uniform intensity across the beam. This approximation is appropriate as long as the sample is set at the beam waist, the spot size of the pump beam is large enough so that the effect of carriers diffusing away from the excited area can be neglected, and the size of the probe beam is much smaller (<1/3) than that of the pump beam.
APPENDIX III

THE THIN GRATING CONDITION
In general, a grating of finite thickness can be divided into a number of thin grating elements as shown in Fig. A3.1. The phase difference between the beams diffracted by the first and the last grating elements, respectively, is

\[
Q = \frac{2\pi n}{\lambda} (d - dc \cos \phi_m),
\]  

(A3.1)

where \( \phi_m \) is the diffraction angle inside the sample. Using Eq. (A2.8) and \( \sin \phi_1 = \phi_1, \cos \phi_1 \approx 1 - \frac{1}{2} \phi_1^2 \) (\( \phi_1 << 1 \)), for the first diffraction, one obtains

\[
Q = \pi d \lambda / \Lambda^2 n.
\]  

(A3.2)

If the phase difference is sufficiently small, so that \( Q << 1 \), the beams from all the grating elements interfere constructively. This is known as the thin grating condition. In this case, the whole grating can be treated as a single thin grating. For our experiments, the samples have active thicknesses of \( \sim 1.5 \) \( \mu \)m, and refractive index of \( \sim 3.5 \), the laser wavelength, \( \lambda \), is \( \sim 0.97 \)\( \mu \)m, and the grating spacing, \( \Lambda \), is \( \sim 8 \)\( \mu \)m. Substituting these values
into Eq.(A3.2) yields $Q = 0.02 << 1$. Therefore, it is appropriate to use the thin grating theory to interpret our experimental results.
APPENDIX IV

KRAMERS-KRONIG RELATION
The refractive index and the absorption coefficient of a medium are related by the Kramers-Kronig relations (Stern 1963), which remain valid for a material containing a fixed number of carriers. The absorption coefficient and the refractive index of a semiconductor containing a fixed number of photo-generated carriers are therefore also related by the Kramers-Kronig relations (Miller *et al.* 1983a, Chemla *et al.* 1984). The change of refractive index, \( \Delta n(\omega) \), associated with the change of absorption coefficient, \( \Delta \alpha(\omega) \), is

\[
\Delta n(\omega) = \frac{c}{\pi} \mathcal{P} \int_0^\infty \frac{\Delta \alpha(\omega') d\omega'}{\omega'^2 - \omega^2} \tag{A4.1}
\]

where \( \mathcal{P} \) indicates that the principal value of the integral has to be evaluated.

In our experiments, the \( \Delta \alpha \) spectra were obtained by tuning the laser wavelength across the \( n = 1 \) hh excitonic absorption peak. The measurements therefore, were not made at fixed carrier density, since both linear absorption coefficient and laser output power were functions of wavelength. In order to use the Kramers-Kronig relations, we need to convert the \( \Delta \alpha(\omega) \) and \( \Delta n(\omega) \) to \( \alpha_{eh}(\omega) \) and \( n_{eh}(\omega) \) by using the definition given in Eq. (4.3) and (4.6) respectively. Both \( \alpha_{eh}(\omega) \) and \( n_{eh}(\omega) \) are for fixed carrier density, and are therefore related by Kramers-Kronig relations, i.e.

\[
n_{eh}(\omega) = \frac{c}{\pi} \mathcal{P} \int_0^\infty \alpha_{eh}(\omega') d\omega' \tag{A4.2}
\]

\( \alpha_{eh}(\omega) \) is not zero only in a limited range around the \( n = 1 \) hh excitonic absorption peak, especially for the low carrier densities at which the \( \alpha_{eh} \) measurements were performed, therefore, the limits of the integral in Eq. A4.2 can be set to some finite value without introducing error. Numerical calculation of Eq. A4.2 is difficult because of the resonant denominator. However, the convergence problems can be avoid by converting a discrete set of measured \( \alpha_{eh}(\omega) \) into a continuous set by linear interpolation as shown in Fig. A4.1.
Fig A4.1. A discrete set of measured $\sigma_{eh}$ converted into a continuous set by linear interpolation. (After Kost et al. 1988).

(A. Kost et al. 1988), and carrying out the integration analytically. In Fig. A4.1, $\omega_i$ is the frequency of the $i$th points of the absorption spectrum. The slope, $m_i$, and the intercept, $b_i$, of the straight line connecting two measured points $\sigma_{eh}(\omega_i)$ and $\sigma_{eh}(\omega_{i+1})$ are given by

$$m_i = \frac{\sigma_{eh}(\omega_{i+1}) - \sigma_{eh}(\omega_i)}{\omega_{i+1} - \omega_i} \quad (A4.3)$$

and

$$b_i = \sigma_{eh}(\omega_i) - m_i \omega_i. \quad (A4.4)$$

The Kramers-Kronig relations Eq. (A4.2) becomes
\[ n_{eh}(\omega) = \frac{c}{\pi} \sum_{i=0}^{n} \frac{m_i \omega_{i+1} + b_i}{\omega_{i+1}^2 - \omega_i^2} \, d\omega_i \]

\[ = \frac{c}{\pi} \sum_{i=0}^{n} \left( \frac{m_i}{2} \ln \left| \frac{\omega_{i+1}^2 - \omega_i^2}{\omega_{i+1}^2 - \omega_i^2} \right| + \frac{b_i}{2\omega_i} \ln \left| \frac{\omega_{i+1} - \omega_i + \omega_i}{\omega_{i+1} + \omega_i - \omega} \right| \right). \tag{A4.5} \]
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