Second quantum state transition in GaAs/AlGaAs resonant Bragg structure probed by modulation reflectance spectroscopy

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SECOND QUANTUM STATE TRANSITION IN GaAs/AlGaAs RESONANT BRAGG STRUCTURE PROBED BY MODULATION REFLECTANCE SPECTROSCOPY

by

YUECHAO CHEN

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

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ABSTRACT

SECOND QUANTUM STATE TRANSITION IN GaAs/AlGaAs RESONANT BRAGG STRUCTURE PROBED BY MODULATION REFLECTANCE SPECTROSCOPY

by

Yuechao Chen

Advisors: Prof. Mim L. Nakarmi and Prof. Vladimir V. Chaldyshev

Modulation spectroscopy, ever since its introduction by B.O. Seraphin in 1964, has been considered and widely used as a sensitive experiment technique for studying and characterizing the properties of varieties of semiconductor materials. Compared to general optical reflectance spectrum which measures the absolute reflection, the modulation spectroscopy evaluates the interpretation of the changes in the optical response from the sample caused by a periodic physical perturbation applied to the sample, such as temperature, electric fields, hydrostatic pressure, uniaxial stress, etc. Those modulation spectroscopies with an external electric field perturbation are known as electroreflectance spectroscopy, which provides sharp and derivative-like spectral features in the energy region of excitonic transitions in the semiconductors while suppressing uninteresting background effects that are not affected by the electro-modulation. One interesting phenomenon is that when the excitonic transition energy in a periodic dielectric structure, for example multiple quantum well (MQW) structure, meets the Bragg resonance condition, the reflectance spectrum shows an enlarged responding effect with enhanced reflectivity and broadened transition features. This kind of a structure is known as a resonant Bragg structure (RBS) and the coincidence of the
exciton and Bragg resonances is called the double resonance condition.

In this thesis, we employed both electroreflectance and optical reflectance spectroscopies to probe excitonic transitions in a GaAs/AlGaAs RBS MQW structure. The sample structure was specially designed and fabricated to tune the double resonance condition around the second state of the heavy-hole exciton $x(e_2-hh_2)$ transitions by variation of the incident angle and temperature. The sample used in this experiment consists of 60 periods of quantum well structures with GaAs well layer (13 nm) and AlGaAs barrier layer (94 nm), grown by solid source molecular beam expitaxy on a semi-insulating GaAs substrate. We observed a significant enhancement of excitonic features at the $x(e_2-hh_2)$ exciton transitions around incident angle of $23^\circ$ in both techniques, revealing the double resonance condition at low temperature. Additionally, heavy-hole and light-hole ground state exciton transitions $x(e_1-hh_1)$ and $x(e_1-lh_1)$ were also evaluated. In the temperature dependence of optical reflectance and electroreflectance from the double resonance condition, we observed red-shift of the excitonic features. The electric field dependence measurement of electroreflectance exhibited a broadening effect for the $x(e_2-hh_2)$ exciton transition.
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Chapter 1

Introduction

One of the most important topics in photonics is understanding light-matter interaction which integrates semiconductor and optics. Various types of spectroscopies such as reflectance, transmission and absorption spectroscopies are used as tools to study them. These spectroscopies provide insightful information of the matter through its interaction with light (electromagnetic radiation), thus gives a valuable reference to study the matter and its structure. In multiple quantum wells (MQWs) structure, strong coupling of photon with excitons inside MQWs forming exciton-polaritons results in rich transition features in the spectroscopies at various exciton transitions. Although two typical kinds of excitons, light-hole and heavy-hole related excitons in a MQW structure of different materials have been well studied at their first excitation energy level, namely \( x(e_1-lh_1) \) and \( x(e_1-hh_1) \) \([1, 2, 3, 4]\), but little work has been done related to exciton transitions at higher energy levels such as heavy-hole exciton at the second state \( x(e_2-hh_2) \). The goal of this research is to observe this higher exciton transition in a GaAs/AlGaAs MQW structure using reflection.
spectroscopies. In order to achieve this, resonant Bragg structure (RBS) sample is introduced and fabricated for this study. An RBS structure is a special photonic crystal (PhC) that was proposed by Ivchenko et al. [5], that not only allows Bragg diffraction of light providing reflective and transmissive energy bands for the propagation of external light, but also exhibits resonant optical properties when the photon energy equals to the excitonic transition energy in the structure. This resonance greatly enhances the interaction of photon with excitons inside the RBS structure, resulting in magnificent transition features in the spectroscopies.

Reflectance techniques such as optical reflectance (OR), photoreflectance (PR) and electroreflectance (ER) spectroscopies are useful methods to probe the excitonic transitions in multiple quantum wells structures. OR is the most common spectroscopy method which measures the absolute optical response from the sample. PR and ER are both modulation spectroscopy techniques that evaluate the interpretation of the changes in the optical response from sample caused by a periodically applied perturbation to some measurement conditions, such as temperature, electric fields, hydrostatic pressure, uniaxial stress, etc. Both PR and ER generate differential-like spectrum in the region of exciton transitions, with enhanced and sharpened transition spectrum features, and in the meantime suppresses uninterested background noises. This dissertation work has employed all these three spectroscopy techniques on a GaAs/AlGaAs multiple quantum well Bragg structure.
1.1 Photonic Crystals

When dielectric permittivity of a medium spatially modulated periodically, it gives rise to a resonant optical reflection and transmission features due to the Bragg diffraction of the electromagnetic waves. Such media can be fabricated using a periodic system of the matter with different indices of refraction. The famous example is the dielectric photonic crystals (PhC) [6, 7, 8] such as natural and synthetic opals [9]. One-dimensional structures of this type are often referred to as distributed Bragg reflectors or dielectric mirrors with important applications in optical and optoelectronic devices, for instance, vertical cavity surface emitting lasers (VCSELs) [10].

Due to the periodically structured electromagnetic media, essentially containing regularly repeating internal regions of high and low dielectric constant media, photonic crystals affect the propagation of electromagnetic (EM) waves, typically light [11, 6]. This behavior is very similar to the periodic potential in semiconductor crystals which affects the electron motion by defining allowed and forbidden electronic energy bands. Generally, photonic crystals possess photonic band gaps: ranges of certain frequency in which light cannot propagate through the structure for which photonic crystals are also called photonic band-gap materials. Frequencies allowed to propagate through the PhCs are known as modes and groups of allowed modes form bands. Bands of disallowed frequencies (or wavelengths) are called gaps, or specifically, photonic band gaps. Varieties of optical phenomena of photonic crystals are due to this light gap property, such as high-reflecting omni-directional mirrors [12, 13, 14], low-loss wave-guiding [15, 16, 17], and spontaneous emission [18, 19, 20]. The periodicity of the photonic crystals has a length scale proportional to the
wavelength of light in the band gap.

In this thesis work, GaAs/AlGaAs multiple quantum well structure was used. It has a periods of quantum well exhibiting 1-D photonic crystal structure.

Depending on the directions of the periodicity in the material (typically dielectric), the photonic crystal structure may be periodic in one, two or three dimensions.

Properties of electromagnetic wave propagation in one-dimensional photonic crystals was first studied by Lord Rayleigh in 1887 [21] and the research began to grow extensively ever since, especially after 1987 [22, 23, 24]. Vladimir Bykov performed a detailed theoretical study of one-dimensional optical structures in 1972, and firstly investigated the effect of a photonic band gap on the spontaneous emission from atoms and molecules embedded within the photonic structure [25]. Two-dimensional photonic crystals study began in 1996, when Thomas Krauss made the first demonstration of two-dimensional photonic crystals in optical wavelengths [26], while the study on three-dimensional photonic crystal has proceeded much more slowly due to the difficulties in fabrication [27].

Study of photonic crystals led to many subsequent development in the exploration of the material and structure including their fabrication, theory, and application. The existence of photonic band gap that has certain range of forbidden of light propagation through the material, makes photonic crystals useful materials that offer us control of light propagation. One-dimensional photonic crystals are widely used in thin-film optics with coatings for lenses and mirrors (e.g. quarter-wave stack), color changing paints, and potentially optical switch [28]. Applications of two-dimensional photonic crystals in low-threshold lasers [29, 30] and high efficient LEDs [31] have been widely ex-
explored recently. Additionally, two-dimensional photonic crystal fibers are used in nonlinear devices and for guiding exotic wavelengths [27]. Three-dimensional photonic crystals also have potential applications in optical pulse control for ultra-short laser pulses [32]. Furthermore, a prospect of using three-dimensional photonic crystals for optical computing has also been proposed. The sample used in this project has periodic multiple quantum wells and can be considered as a one-dimensional photonic crystal structure.

1.1.1 Formation of band-gap in one-dimensional photonic crystals

The mechanic of formation of band gaps of light in one dimensional photonic crystals is quite similar as periodic potential in a semiconductor crystal that affects electron motion by defining allowed and forbidden electronic energy bands in solid-state physics [33].

The wave propagation equation in three-dimensional periodic media can be derived from Maxwell’s equations as an eigen problem in analogue with Schrodinger’s equation. By combining the source-free Faraday’s Law and Ampere’s Law at a fixed frequency \( \omega \), i.e. time dependence \( e^{-i\omega t} \), one can obtain an equation in only the magnetic field \( \vec{H} \):

\[
\nabla \times \frac{1}{\varepsilon} \nabla \times \vec{H} = \left( \frac{\omega}{c} \right)^2 \vec{H}
\]

(1.1)

where \( \varepsilon \) is the dielectric function \( \varepsilon (x, y, z) \) and \( c \) is the speed of light. This is an eigen value equation, with eigen value \( (\omega/c)^2 \) and an eigen operator \( \nabla \times \frac{1}{\varepsilon} \nabla \times \) that is Hermitian. Furthermore, the operator is positive-definite (for real \( \varepsilon > 0 \)), which implies that the eigen frequencies \( \omega \) are real [34].
In one-dimensional photonic crystals, a 1-D periodic function \( \varepsilon(x) = \varepsilon(x + a) \) applies for the dielectric structure. In this case, the Bloch-Floquet theorem for periodic eigen problem states that the waves propagating behavior is governed by a periodic envelope function multiplied by a plane wave:

\[
H(x) = e^{ikx} H_{n,k}(x)
\]  

(1.2)

with eigen values \( \omega_n(k) \) that are continuous and periodic functions of wave number \( k \), where \( H_{n,k} \) is a periodic envelope function satisfying:

\[
(\nabla + ik) \times \frac{1}{\varepsilon} (\nabla + ik) \times H_{n,k} = \left(\frac{\omega_n(k)}{c}\right)^2 H_{n,k}
\]

(1.3)

A complete photonic band-gap is a range of \( \omega \) in which there is no propagating (real \( k \)) solution of Maxwell’s equation for any \( k \) surrounded by propagating states above and below the gap. In other words, one can say that the wave frequency \( \omega \) selects the propagating light species, while the wave number \( k \) defines a propagating method. In a uniform media, the Maxwell’s equation has plane wave eigen solutions with frequency \( \omega \) that is linearly dependent on wave number \( k \). Especially, \( \omega = ck \) when \( \varepsilon = 1 \). In an artificial period \( a \), the \( k = -\pi/a \) mode lies at an equivalent wave vector to the \( k = \pi/a \) mode, making an artificial degeneracy at the edges of the first Brillouin zone. This degeneracy breaks when a periodic dielectric structure is presented, forming a gap in the \( \omega - k \) function diagram, i.e., a band-gap for light propagation, as shown in Fig. 1.1. More generally, it states that any periodic dielectric variation in one dimension will lead to a band-gap.
Figure 1.1: (a): Band diagram of frequency $\omega$ versus wavenumber $k$ in a uniform one-dimensional medium. Consider an artificial periodicity $a$, the dashed lines show the “folding” effect into the first Brillouin zone. (b): In a physical periodic dielectric media, the degeneracy at the edges of the first Brillouin zone splits, making the $\omega - k$ function non-continuous anymore, thus forming a band-gap.
1.2 Multiple Quantum Well Structures

Quantum wells are thin layered semiconductor structures which are so-called sandwich structures. The well consists of charge carriers (electrons and holes) in thin layers of one semiconductor “well” material sandwiched by other semiconductor “barrier” layers which have higher energy band gap than well material. The layout of the structure is shown in Fig. 1.2. It is a particular kind of hetero structure. Both electrons and holes have lower energy in the well layer compared to those in the barrier layer; hence both electrons and holes are easily confined.

Figure 1.2: The layout of a quantum well structure. The well layer is typically much thinner than the barrier.

For fabrication of multiple quantum well structures, two common techniques can be used, molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD).
1.2.1 Basic quantum well physics ("particle in a box" model)

To understand the basic properties of a quantum well structure, it’s better to start with the simple "particle-in-a-box" model. The one dimensional Schrödinger’s equation for particle states:

\[
-\frac{\hbar^2}{2m} \frac{d^2 \varphi_n}{dx^2} + V(x) \varphi_n = E_n \varphi_n
\]  

(1.4)

where \( V(x) \) is the structural potential (i.e., the quantum well potential) of the particle along the direction perpendicular to the material layers, \( m \) is the effective mass, and \( E_n \) and \( \varphi_n \) are the eigen energy and eigen function associated with the \( n^{th} \) solution to the equation [35].

In an infinite well case, the barriers on either side of the quantum well are assumed to be infinite high, which makes the wave function zero at the walls of the quantum well.

The solutions are:

\[
E_n = -\frac{\hbar^2}{2m} \left[ \frac{n\pi}{L_x} \right]^2 \quad n = 1, 2, \ldots
\]

\[
\varphi_n = \sqrt{\frac{2}{L_x}} \sin \left( \frac{n\pi x}{L_x} \right)
\]

(1.5)

where \( L_x \) is the width of quantum well.

The allowed states correspond to standing waves in the direction perpendicular to the layers. Due to the standing wave confinement, the energy levels are quantized and the wave functions are sine waves. In the formula, the energy is measured from the bottom of the well and the first allowed energy with \( n = 1 \) is above the bottom of the well. A graph of the energy levels is shown in Fig. 1.3.
The spacing between energy levels is inverse proportional to effective mass $m$ and the well width $L_x$.

![Diagram of energy levels in a quantum well](#)

**Figure 1.3: “Infinite” quantum well and associated wave functions.**

Considering the real material fabrication, the infinite well condition is ideal, while instead, a bound state is met. In this case, the wave solutions are exponentially decaying into the barrier manifesting a tunneling penetration effect. A comparison graph between an infinite well and bound well is shown in Fig. 1.4.
Figure 1.4: Comparison of wave functions in an infinite QW and a finite QW. The tunneling penetration effect can be seen in the latter case.

Figure 1.5 shows a typical band diagram for a quantum well structure. The energy levels available for both electrons and holes in a quantum well are quantized. Additionally, in quantum wells, for the direction perpendicular to the layers, we have the selection rule instead of momentum conservation, which states that only transitions between states of the same quantum number in the valence band and conduction band are allowed. Spacing between the energy levels for electrons and holes are different due to different effective mass of electron and hole.
1.2.2 Light and heavy holes

In general, there are two types of holes with different effective masses in semiconductors. They are referred to as light holes and heavy holes. Since these two kinds of holes have different masses, there are two sets of holes subbands with different energy spacing. The light holes have a considerably smaller effective mass usually comparable to that of the electron and have its sub-band spaced further away, compared to the heavy holes. The heavy-hole-to-conduction state has a slightly lower energy and is more closely spaced than the light-hole-to-conduction state, as shown in Fig. 1.6. The heavy hole state is usually dominant in optical absorption for light propagation perpendicular to the quantum well layers.

Figure 1.5: Energy band profile of a quantum well structure.
Figure 1.6: Typical band structure for light and heavy holes and split off band in quantum wells.

1.2.3 Excitonic effect

Figure 1.7 shows a typical GaAs/AlGaAs quantum well structure absorption spectrum at room temperature, along with a bulk semiconductor absorption curve and a non-excitonic quantum well absorption curve. The absorption curve data is from the reference [36]. From the absorption curves, we can also image the reflection spectra, which will exhibit similar behaviors as of absorption curves. In bulk GaAs semiconductor, the absorption behavior appears as a continuous curve as indicated in Fig. 1.7 by blue dash line. This absorption effect generally increases with the incident photon energy. While a GaAs/AlGaAs quantum well is presented in the sample, according to the “particle-in-a-box” model, the wavelength allowed for propagation in the quantum well is quantized and only a few special photon waves can be absorbed. This appears in the absorption
spectrum with some special steps as indicated by yellow dash line in Fig. 1.7, with vertical steps while assuming that the boundary of quantum well is infinite. In a real GaAs/AlGaAs quantum well structure, the actual absorption spectrum is somewhere between these two situations, which is indicated as black solid line the Fig. 1.7. From the actual absorption spectrum for a quantum well structure, we can see that the quantum well absorption is indeed quantized, with a series of steps and the results from “particle-in-a-box” model predicts the positions of the steps very well. However, a sets of peaks near the band gap energy shows such a strong effect, which cannot be obtained from the simple “particle-in-a-box” model, or in another word, the “non-excitonic” model, in which, particles are considered free. To understand this problem, the concept of excitons must be introduced [37, 38, 39, 40].
Figure 1.7: A typical GaAs/AlGaAs quantum well structure absorption spectrum at room temperature [36]. Strong peaks effect can be seen due to the excitons presents particularly near the band gap energy. A brown dashed line shows the theoretical quantum well absorption spectrum when excitons effect is neglected, calculating from the “particle-in-a-box” model. The blue dashed line is the optical absorption in bulk (i.e. 3D) semiconductors.

Electrons and holes are charged particles with negative and positive charges respectively. Due to the existence of Coulomb interaction, the electrons and holes attract each other making a bound electron-hole pair, called exciton. Hence, when we have an electron and hole pair in a semiconductor, their wave functions are not plane waves anymore. Plane waves correspond to the case of uniform independent motion of the electrons or holes.
When an exciton is created, it has less energy than a “free” electron-hole pair. The binding energy of lowest, 1S exciton is:

\[ E_B = \frac{\mu e^4}{2h^2\varepsilon^2 R\varepsilon_0} \]  \hspace{1cm} (1.6)

where \( h \) is the reduced Planck’s constant, \( \varepsilon_R \) is the relative permittivity, \( \varepsilon_0 \) is the permittivity of free space, and \( \mu \) is the reduced mass,

\[ \mu = \frac{m_e m_h}{m_e + m_h} \]  \hspace{1cm} (1.7)

This makes the possible transition at an energy

\[ E_{\text{exciton}} = E_{\text{band gap}} - E_B \]  \hspace{1cm} (1.8)

For bulk GaAs, where excitons can be clearly seen at low temperature, the binding energy \( E_B \) is approximately 4 meV, and the transition is illustrated in Fig. 1.8.

![Figure 1.8: Exciton binding energy in bulk GaAs semiconductor.](image-url)
1.3 Bragg Resonance in Multiple Quantum Wells

One dimensional propagation of light waves in a layered structure is a classical problem in solid state optics [41, 42, 43]. Theory of light reflection and transmission near the exciton resonance frequency in a multiple quantum well structure, known as resonant Bragg structure (RBS), was first introduced by E.L. Ivchenko [5]. An enhanced resonant optical reflection was predicted in such resonant Bragg structures (RBS), when the periodicity of the system meets the Bragg resonance condition at the photon energy equal to the energy of the QW excitons. In this case the Bragg scattering of light by the QW excitons leads to formation of a superradiant optical mode at the resonant frequency. Formation of the superradiant optical mode was studied theoretically [5, 39, 44, 45, 46, 47, 48, 49, 50, 51, 52] and documented experimentally for the systems of CdMnTe QWs with CdZnMgTe barriers [53, 54], InGaN QWs with GaN barriers [55], InGaAs QWs with GaAs barriers [40, 56, 57, 58] and GaAs QWs with AlGaAs barriers [1, 2, 59, 60, 61, 62, 63].

An important feature of the RBS is the ability to tune its optical properties by applying external fields that affect the excitonic states of the QWs [2, 56, 59, 60, 61, 62, 64]. For instance, the external electric field changes both the quantum confinement energies of electrons and holes and the exciton binding energy. As a result, it changes the resonant optical respond of the RBS, which can be utilized in a variety of optoelectronic and photonic devices.

The coupling between the light and the excitonic states in the RBS does not require a steady population of excitons or electrons at the desired quantum states. Thus, in contrast to the light emitting devices that usually utilize the lowest quantum states, the RBS reflectors can be build on
the base of any excitonic quantum state available in the QWs. Until recently [62], however, all the
research of RBS has been focused on the ground quantum state of the heave-hole and light-hole
excitons in periodic systems of QWs, where the formation of superradiant exciton-polariton mode
has been well documented. In References [62], and [1] V. Chaldyshev et al. and we reported the
experimental feasibility of an RBS based on the heavy-hole excitons associated with the second
quantum state of electrons and holes, x(e2-hh2), in the GaAs QWs. The ability of the x(e2-hh2)
excitons to couple with light was found to be approximately equal to that of the common excitons
at the ground quantum state x(e1-hh1).

In this thesis we report the results of an extended experimental work of the optical and
electro-optical properties of the RBS based on the x(e2-hh2) excitons in the GaAs QWs separated by
AlGaAs barriers. Although the exciton transitions at their ground states have been very well studied
both theoretically and experimentally, we didn’t find any report on RBS with exciton transitions at
higher energy levels, such as heavy-hole exciton at the second state x(e2-hh2), which is the focus of
this thesis. We already published part of our results in APL 2011 and this thesis will be a complete
and detailed explanation of our work.

1.3.1 Bragg reflection of light from MQWs structures

The multiple quantum well structure can be designed and fabricated with different parame-
ters to modify the light-matter interaction. For example, one can design and fabricate GaAs/AlGaAs
MQW samples with different well and barrier width, and number of quantum wells. As a result the
excitonic energies can vary corresponding to certain parameters.
For a normal periodic structure crystal with lattice planes separated by the interplanar distance of $d$, the normal Bragg diffraction occurs when the scattered electromagnetic waves from different planes in the system undergoes constructive interference. An enhanced reflection can be observed when the Bragg condition is satisfied. This can be described as,

$$2d \cos \theta = m\lambda$$  \hspace{1cm} (1.9)

where $d$ is the spacing between the planes in the periodic structure, $\theta$ is the incident angle of probing light, $m$ is integer, and $\lambda$ is the wavelength of the incident light.

A special structure can be designed in which the exciton energy meets the Bragg resonance wavelength. Under this circumstance, the absorption and reflectance spectrum of the MQW structure shows an enlarged responding effect, with enhanced reflectivity and broadening of the transition features. This coincidence of the Bragg resonance with the MQW excitonic energy level forms a double resonance.

Considering the Bragg resonance and exciton energy of the Bragg MQW structure, the double resonance appears when the following condition is satisfied,

$$2d\sqrt{n^2 - \sin^2 \theta} = m\lambda$$  \hspace{1cm} (1.10)

where $d$ is period of the Bragg MQW structure, $n$ is the refraction index, $\theta$ is the angle of incidence of light from external medium (air), $m$ is integer, and $\lambda$ is the wavelength of the incident light.

The optical reflectance spectrum from a resonant Bragg structure shows a Lorentzian line
shape with a half-width of $N\Gamma_0 + \Gamma$, where $\Gamma_0$ and $\Gamma$ are, respectively, the radiative and non-radiative exciton damping constants in a single quantum well, when the difference between the permittivity of the barrier layer and the background permittivity in the quantum well layer can be ignored [5]. The calculation of the reflectance spectra for an arbitrary numbers of quantum wells $N$ including an intermediate region with $N\Gamma_0$ and $\Delta = \sqrt{2\omega_0\Gamma_0/\pi}$ are comparable in magnitude, is introduced by Ikawa and Cho [48] and Pilozzi et al. [49]. Their calculations show the existence of two special frequencies at which the reflectance from the resonant Bragg structure with no dielectric constant contrast exhibits itself as of the reflectance of a homogeneous barrier material, virtually independent of the number of quantum wells, $N$.

The heterostructure containing $N$ quantum wells in this study is shown in Fig. 1.9. The sample is mounted in a vacuum environment. The sample consists a cap layer of the barrier material B with thickness of $b'$ followed by $N$ equidistant quantum wells of material A as the well layer with thickness of $a$ and material B as the barrier layer with thickness of $b$ and a semi-infinite medium of material B.
Figure 1.9: Geometry of reflection of normal incident light with amplitude $E_0$ from quantum wells consisting of $N$ QWs.

The reflectance under normal incident light at frequency $\omega$ for this structure can be approximately written as [5]:

$$r(N) = \frac{r_{01} + \tilde{r}_N e^{2i\varphi'}}{1 + r_{01} \tilde{r}_N e^{2i\varphi'}}$$

(1.11)

where $r_{01} = (1 - n_b) / (1 + n_b)$ is the amplitude reflection coefficient at the interface between vacuum and the outermost cap layer and $n_b$ ($\varepsilon_b \equiv n_b^2$) is the refraction index of the barrier material B; $\tilde{r}_N$ is the coefficient of reflection inside the structure from the system of $N$ quantum wells, i.e. the amplitude ratio of the reflected and transmitted light at the plane shifted by half width of the barrier layer $b/2$ from the first quantum well into the cap layer, as indicated in Fig. 1.9 by a dashed line; $\varphi' = k_b (b' - b/2)$, as $k_b = n_b (\omega/c)$, is the phase change a wave undergoes in traveling a
distant of \( b' - b/2 \) in the barrier, i.e. from the outmost surface of the cap layer to the dashed plane indicated in Fig. 1.9, and \( c \) is the speed of light in vacuum.

A convenient way to calculate the reflection coefficient \( \tilde{r}_N \) from \( N \) quantum wells is to use the method of transfer matrices relating the forward and backward light waves at the left and right hand boundaries of a layer [65]. Considering distance of \( b/2 \) to the left from the left hand boundary of a well layer and \( b/2 \) to the right from the right hand boundary of a well layer as the left and right hand boundaries for a quantum well, the transfer matrix \( \hat{T}^{(N)} \) involving \( N \) quantum wells is equal to that for just one quantum well raised to the \( N \)th power, as follows:

\[
\hat{T}^{(N)} = \hat{T}_{1}^{N}
\]

\[
\hat{T}_{1} = \frac{1}{\tilde{t}_{1}} \begin{bmatrix}
\tilde{r}_{1}^2 - \tilde{r}_{1} & \tilde{r}_{1} \\
-\tilde{r}_{1} & 1
\end{bmatrix}
\]

(1.12)

where the complex coefficients \( \tilde{r}_1 \) and \( \tilde{t}_1 \) are the amplitude reflection of light from and transmission through a single layer with a quantum well located at the center of the layer, respectively.

The reflection and transmission coefficients for a single quantum well at normal incidence in the vicinity of the exciton resonance can be written as [45, 66, 67, 68],

\[
\tilde{t}_1 = e^{ik_0d_{t1}}, \quad \tilde{r}_1 = e^{ik_0d_{r1}}
\]

\[
t_1 = 1 + r_1, \quad r_1 = \frac{i\Gamma_0}{\omega_0 - \omega - i(\Gamma_0 + \Gamma)}
\]

(1.13)

where \( \omega_0 \) is the exciton resonance frequency, \( d = a + b \) is the period of the quantum well structure, and \( \Gamma_0 \) and \( \Gamma \) are, respectively, the radiative and non-radiative exciton damping constants in a single
quantum well. Equation 1.13 neglects the difference between barrier layer permittivity and well layer permittivity in the quantum wells.

From that, the coefficient of reflection inside the structure from the system of \( N \) quantum wells, \( \tilde{r}_N \) can be written as [3, 68],

\[
\tilde{r}_N = \frac{\tilde{r}_1}{1 - \tilde{r}_1 \frac{\sin(N-1)Kd}{\sin N K d}}
\]  (1.14)

where \( K \) is the wave vector of the exciton polariton at a frequency \( \omega \) in an infinite periodic structure, which can be calculated as [3],

\[
\frac{1}{2t_1} \left( \tilde{t}_1^2 - \tilde{r}_1^2 - 1 \right) + \frac{1}{t_1} = \cos Kd
\]  (1.15)

The final reflection coefficient \( R_N \) is then,

\[
R_N = |r (N)|^2 = \left| \frac{r_{01} + \tilde{r}_N e^{2i\phi'}}{1 + r_{01} \tilde{r}_N e^{2i\phi'}} \right|^2
\]  (1.16)

where one crucial relationship for satisfying the Bragg resonance at the exciton transition energy is,

\[
\frac{\omega_0}{c} n_g d = \pi
\]  (1.17)

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1.4 Modulation Spectroscopy

Concept of modulation spectroscopy was first introduced by B. O. Seraphin in 1964 [69]. It is a sensitive experimental technique for the study and characterization of large number of semiconductor configurations including semiconductor bulk/thin films [69, 70, 71, 72, 73, 74, 75]; semiconductor structures such as superlattices [76, 77, 78, 79, 80], quantum wells [81, 82, 83, 84, 85, 86], quantum dots and heterojunctions [72, 82]; semiconductor interfaces/interfaces such as Schottky barriers [72], metal-insulator-semiconductor [72, 87]; actual device structures, semiconductor growth, and processing.

Instead of measuring actual optical response of a sample using other spectroscopy methods, such as photoluminescence (PL), photoluminescence excitation spectroscopy (PLE), absorption spectroscopy, photocurrent spectroscopy, spectral ellipsometry, resonant Raman scattering etc., modulation spectroscopy deals with the interpretation of the changes in the optical response of a sample caused by a periodic physical perturbation applied to the sample. Those perturbations which are modulating some parameters applied to the sample are called external modulation, while those modulating the measurement conditions themselves are called internal modulation. These observed changes with respect to modifying parameter exhibit a derivative nature of the absolute spectrum of the sample. Derivative nature of modulation spectroscopy emphasizes spectrum features of interband or intersubband transitions in semiconductor structure, and suppresses uninteresting background noises. Furthermore, due to the sensitive property of the derivative like spectrum some weak features that may not be detected in other absolute optical spectra, can be detected and
studied in modulation spectroscopy.

1.4.1 Electromodulation (EM)

Electromodulation (EM) is one of the most useful methods in measuring both reflectance and transmittance spectra on semiconductors and semiconductor microstructures. This is the technique extensionally used in this dissertation work. The method was first introduced by Seraphin [88, 89] and has been widely used ever since [69, 71, 72, 90, 91, 92, 93, 94, 95]. In electroreflectance measurements, periodic modulation of an applied electric field on a dielectric substrate produces sharp features in the reflectivity spectrum of the material when the photon energies correspond to interband or intersubband transitions. For those bulk/thin film materials with band-to-band transitions, EM under certain conditions (low field limit) produces a third-derivative spectrum, while for bound states (excitons, SQW and MQWs), the spectrum exhibits a first-derivative profile. At sufficiently high fields (either modulated or built-in), a Franz-Keldysh oscillation spectrum will be obtained [69, 71, 93, 94].

EM can be accomplished in several ways, including contact and contactless modes. The former requires the sample to be specially fabricated and has its limit in extreme conditions, such as low temperature and high modulation field. The later mode called contactless electroreflectance (CER) first introduced by Yin and Pollak [95] requires no special mounting of the sample and can be used under a variety of conditions.

A viable alternative to CER is the differential reflectance (DR) spectroscopy technique, introduced by Gal et al. in 1990 [96]. Instead of measuring one sample under a periodic electromodu-
lation, a reference sample is measured at the same time and the difference between the reflectivities of the two samples are evaluated. To realize such samples, Gal et al. introduced additional surface states in one-half of a sample while leave the another half intact. By changing the number of surface states, the surface electric field of the sample can be modified, which achieves different electric fields in the two “identical” samples, leading to differential reflectance spectroscopy which is analogous to electroreflectance.

Figure 1.10: Comparison of room temperature reflectivity and electric field-modulated reflectivity (electroreflectance) of GaAs [97].

Figure 1.10 is the work reported by Fred Pollak et al. [97] and provides a good example of the strength of modulation spectroscopy compared to regular optical reflectance spectroscopy. The
top curve in Fig. 1.10 is the reflectivity (R) spectra, while the bottom one shows the electric-field modulated spectra, also known as electroreflectance (ER) spectra. Both spectrum are taken from a bulk GaAs sample at room temperature (300 K). While the reflectivity spectra is characterized by broad features, the ER spectra has a zero base and is dominated by a series of very sharp, derivative-like features. These features are corresponding to specific transitions in the Brillouin zone (BZ) in bulk GaAs at room temperature. Compared to the reflectivity curve, the transition energies can be much easily identified in the electroreflectance spectra. Since the changes due to modulated electric-field is evaluated, thus, they cancel these uninteresting signals which are not affected by electric-field, including background light and noise.

One of the great advantages of modulation spectroscopy is the ability to perform a lineshape fitting. Since the transition features in a modulation spectroscopy are localized in photon energy, lineshape fitting provides a good method to obtain accurate values of transition energy and also broadening parameters of interband transitions. Taking the electroreflectance spectra in Fig. 1.10 for example, it is possible to determine the energies of the $E_0$, $E_0 + \Delta_0$, $E_1$ and $E_1 + \Delta_1$ peaks within a few meV even at room temperature. This provides a convenient way to study the effects of static external perturbations such as electric and magnetic fields, temperature, hydrostatic pressure, uniaxial stress, composition etc.

Besides the sharp, derivative-like transition features, electroreflectance spectra, which is driven by modulation perturbation, also contains important information in other modulation parameters. For example, modulation phase, frequency, amplitude will all have their particular effect on a modulation spectroscopy. Certain types of modulation spectroscopy can be spatially (depth) selective and thus act as probes of specific regions of the sample being studied. Considering these
effects, it is possible to investigate the effects of a certain perturbation on the sample by changing the perturbation in a specific manner.

1.4.2 Optical Modulation

Optical modulation, also known as photoreflectance spectroscopy (PR), is considered as a special electro modulation (EM) technique which is different from electroreflectance spectroscopy (ER) [97, 98]. Although both PR and ER are modulation spectroscopies which probe and evaluate the changes in the optical response from a sample under a periodically modulated electric field inside the sample, the methods to drive the modulating electric field in the sample are different. In photoreflectance (PR) measurements, modulation of the built-in electric field in the semiconductor sample is caused by photo-excited electron-hole pairs created by a laser beam pump source. This laser pump beam is modulated by a chopper at a certain frequency, serving as the modulation reference.

The photon energy of the laser pump source should be enough to create the electron-hole pair in the semiconductor material. That means, for bulk material investigation, photon energy of the laser beam should be greater than the bandgap of the semiconductor material; while for exciton transition investigation as presented in this thesis, the beam energy should be above the exciton transition energy in a semiconductor sample. A typical pump is a 5 mW He-Ne laser for room or low temperature measurements. At high temperatures, a more powerful laser beam should be used.

In a PR measurement, the laser pump beam is absorbed by the semiconductor material and create electron-hole pairs that alters the dielectric function and built-in electric field in the material,
Table 1.1: Illumination source, filter and detector options for PR measurements

<table>
<thead>
<tr>
<th>Range (nm)</th>
<th>Source</th>
<th>Filter</th>
<th>Detector</th>
</tr>
</thead>
<tbody>
<tr>
<td>220-400</td>
<td>Xenon</td>
<td>None</td>
<td>Silicon</td>
</tr>
<tr>
<td>375-730</td>
<td>Xenon</td>
<td>WG370</td>
<td>Silicon</td>
</tr>
<tr>
<td>505-990</td>
<td>Tungsten</td>
<td>OG500</td>
<td>Silicon</td>
</tr>
<tr>
<td>1000-1690</td>
<td>Tungsten</td>
<td>RG850</td>
<td>InGaAs</td>
</tr>
</tbody>
</table>

hence affects the reflectance coefficient $R$ of the material for a given illuminating beam of light. The difference of the light reflectance with or without a built-in electric filed caused by the laser pump beam is collected and evaluated as a modulation spectroscopy. This alternation is controlled by using a chopper on the laser pump beam at a certain frequency.

Due to the modulation spectroscopy, sharp and derivative-like features corresponding to the interband or intersubband transitions of the material can be obtained in PR spectra. By fitting line shape of the PR spectra, more accurate information about bandgaps, interband or intersubband transitions and broadening parameters can be extracted. And also Franz-Keldysh oscillations can be used to determine the internal fields and doping levels of the semiconductor sample.

Photoreflectance technique allows very sensitive and non-destructive characterization at room or low temperature measurements. A wide range of semiconductor materials, including silicon, III-V or II-IV compounds can be investigated with optional detectors, filters and sources, as listed in Table 1.1.

An example is presented in Fig. 1.11 which shows the photoreflectance (PR) spectra at room
temperature (300 K) from a thick GaAlAs epitaxial layer on GaAs and a series of GaAs/GaAlAs multiple quantum wells with different well widths \((L_x)\) in the vicinity of the \(E_0\) transition [82]. The transition features in the spectra are extremely rich even at room temperature. The notation \(h_n\) and \(l_n\) represent “symmetry-allowed” transitions of index \(n\) between the quantized conduction and valence subbands of heavy \((h)\) or light \((l)\) hole character. In fact, all the “symmetry-allowed” transitions were observed. The solid arrows are the energies of the various transitions calculated from theory. These rich modulation spectra from microstructures may be used to probe the “band structure” of reduced dimensional systems.

Compared to the electroreflectance technique, the weakness of a photoreflectance measurement is that it is very hard to eliminate laser scattering and reflecting signals coming into the detector, which causes magnificent noise and sometimes buries the useful transition features in the final spectra.
Figure 1.11: Room temperature photoreflectance spectra for an undoped GaAs/Ga$_{1-x}$Al$_x$As heterojunction (top trace) and three multiple quantum well samples with $x \approx 0.2$. The arrows labeled $h_1, h_2, ..., h_n, (l_1, l_2, ..., l_n)$ correspond to the calculated values of “symmetry-allowed” interband transitions between heavy (light) hole valence and conduction subbands [82].
Chapter 2

Experiment Setup

In this dissertation work, a multiple quantum well (MQW) structure is studied by three spectroscopy techniques, optical reflectance (OR), photoreflectance (PR) and electroreflectance (ER). A cryostat system was used in all these setups to hold the MQW sample, accompanied with a cooling and vacuum system. All the measurements are taken under a vacuum environment below 0.1 mTorr by the vacuum pumping system. While the cooling system can control the temperature of the sample in a range of 20 – 300 K. A tungsten light was used together with a monochromator to output a narrow band of light as the probing light source. On the computer end, we wrote a Labview program to control the stepping motor on the monochromator and collect the reflectance data. Detailed experimental setups is described in this chapter.
2.1 Optical Reflectance (OR) Setup

Figure 2.1: Optical reflectance setup.

Figure 2.1 is a schematic drawing of the experimental arrangement for optical reflectance measurement. Light from a tungsten halogen lamp source passes through a monochromator, with a bandpass value of 0.83 nm at the exit slit. The exit light then is chopped by a constant frequency chopper of 100 Hz which provides reference for the Lock-in amplifier to collect data. The incident light then is focused on the sample by means of two lens and tuned at certain incident angle. The
sample is mounted on a stage in a cryostat. The reflectance signal from the sample through another two focus lenses is collected by a silicon detector. Then a Lock-in amplifier is used to collect the silicon detector response at the same frequency as the reference frequency from the chopper. A Labview program has been written for automatic collection of final optical reflectance spectrum.
2.2 Photoreflectance (PR) Setup

The experiment setup for photoreflectance measurement is almost similar to the setup for regular optical reflectance measurement, except a laser pump beam is used to excite exciton transitions inside the sample, thus create a built-in electric field as modulation parameter. Schematic experiment setup for photoreflectance setup is shown in Fig. 2.2. A power of 5 mW and emitting wavelength of 670 nm Neon-Helium modulating laser diode is used in this measurement. The
laser beam and the illumination light beam from monochromator are shed onto the same spot on the sample. The laser beam is chopped by a chopper at frequency of 100 Hz as collecting reference for the Lock-in amplifier. To adjust the laser power, a density filter may be used in front of the laser source to block certain amount of laser shed on the sample. The monochromator selects a particular light wavelength with bandpass value of 0.83 nm from the tungsten halogen light source. This probing light is then focused onto the sample through two lens and tuned at certain incident angle. For the reflectance, a lens is used to concentrate the reflected light from the sample to the silicon detector. Then a Lock-in amplifier is used to collect the silicon detector response at the same frequency as the reference frequency from the chopper on the laser pumping beam. The final outcome is collected with a Labview program and saved for further analysis.
2.3 Electroreflectance (ER) Setup

Figure 2.3 is a schematic drawing of the experimental arrangement for electroreflectance measurement. In this case, the modulating signal is the applied voltage across the sample. Light from a tungsten halogen lamp source passes through a monochromator, with a bandpass value of 0.83 nm at the exit slit. The incident light is then focused on the sample by means of two lenses at a certain incident angle. The modulation signal is a square wave voltage generated by a Hewlett Packard 3310B function generator with a peak-to-peak amplitude of 0.5 V and frequency 100 Hz. The signal is amplified 1000 times to a peak-to-peak value of 500 V by a Trek Model 609C-6 high voltage amplifier.
amplifier and applied to the sample. The reflectance signal from the sample through another two focus lenses is detected by the silicon detector. Then a Lock-in amplifier is used to collect the silicon detector response at the same frequency as the reference frequency modulation signal. At the end, a Labview program is written and used to collect final electroreflectance spectrum.
2.4 Experiment Instruments

An optical reflectance system consist of a tungsten halogen light source, monochromator driven by step motor with Labview program, silicon detector and a temperature-controlled closed-cycle cryostat, was designed and built to measure the optical and electro reflectance from the MQW sample placed in the cryostat as shown in Fig. 2.4.
Figure 2.4: Temperature-controlled reflectance system.

The emission spectrum of the tungsten halogen light source measured directly by a silicon detector is shown in Fig. 2.5. The wavelength range for reflectance measurement locates mainly between 700 and 860 nm.
Figure 2.5: Spectrum of the tungsten halogen light source measured using the Si detector.

The PTI Model 102 monochromator from Photon Technology International Inc. is a dual-grating (1200 line/mm and 600 line/mm) quarter-meter Czerny-Turner configuration with an f-number of 4. The standard 1200 line/mm ruled grating disperses the white light spectrum across the exit slit with a mechanical scanning range from 0 to 1000 nanometers. The entrance (back) and exit (front) slits are adjustable continuously from 0 to 6 mm. For most applications when the variation with wavelength can be ignored, the bandpass ($BP$) which is the range of wavelengths with the monochromator emits about a central wavelength setting, is simply the product of the slit width ($W$) times the reciprocal linear dispersion ($Rld$):

$$Rld = 10^6 / (n \times L \times F)$$

$$BP = Rld \times W$$  \hspace{4cm} (2.1)
where \( n \) is the order integer (\( n = 1 \) for highest throughput), \( F \) is the focal length of monochromator (\( F = 200 \) mm), \( L \) is the grating (\( L = 1200 \) line/mm). In this experiment setup, the reciprocal linear dispersion (\( R_{ld} \)) can be calculated as 4.17 nm/mm. The exit slit is fixed at 0.2 mm, which gives a bandpass (\( BP \)) value of 0.83 nm.

For the silicon detector, a typical response curve is shown in Fig. 2.6. The measurement range of this experiment (700 to 860 nm) locates in the best response area for the silicon detector.

![Figure 2.6: A typical response curve of silicon detector for various illumination wavelength, ranging from 300 to 1100 nm. Graph is from https://upload.wikimedia.org/wikipedia/commons/4/41/Response_silicon_photodiode.svg.](https://upload.wikimedia.org/wikipedia/commons/4/41/Response_silicon_photodiode.svg)

The closed cycle cryogenic workstation from Cryo Industries Inc., equipped with a Model M-22 CTI Cryocooler cooling refrigerator and Model 8200 single phase water-cooled compressor
was used for temperature measurements. It can hold the sample in a vacuum environment at less than 0.1 mTorr through a Veeco Type RG-31X vacuum gauge. Temperature can be controlled ranging from 19 to 300 K by a Lakeshore 330 auto tuning temperature controller. The design of the cryostat system is shown in Fig. 2.7.

![Figure 2.7: Closed cycle cryogenic workstation, featured with CTI M-22 cooling refrigerator.](image)
2.5 Computer End Configuration

On the computer end, a Labview program called “Wavelength Scanning Program” has been developed to collect the data of the reflectance intensity on the silicon detector from the sample. The interface of the program is shown in Fig. 2.8.

![Wavelength Scanning Program interface written in Labview.](image)

Figure 2.8: Wavelength Scanning Program interface written in Labview.
The collected data can be shown in two identical data graphs in the program whose data types are controlled by two measurement selectors. A switch connected to the first set of data in Graph 1 is created to toggle the data in raw mode or normalized mode. In the normalized mode, a typical source spectrum for the exact output from monochromator is used to normalized the data points during data collection.

A brief introduction of the control parameters for each measurement is shown in Table 2.1. The “Starting WL (nm)” and “Ending WL (nm)” define the measurement range in wavelength. The behavior of the motor which controls the monochromator is controlled by the parameters “Step size (nm)” and “Step Frequency (Hz)”. The “Step size (nm)” decides the motor stepping size in nanometer for adjacent data points and “Step Frequency (Hz)” is the frequency of the trigger signal for the stepping motor. “Wait time (ms)” controls how long the Lock-in should wait for next collection, which should be smaller than the Lock-in integration time for each collection. The “No. of Points” controls how many collections Lock-in should take to evaluate each data point. The “Measurement selector” for both data graphs in the program controls the data type to collect from the Lock-in system. The value of the selector can be X (in-phase data), Y (out-phase data), R (absolute value data), phase (phase value), and noise (noise value in Lock-in). The X mode is used during an ER collection and the R mode is used in an OR collection.
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Starting WL (nm)</td>
<td>Measurement starting wavelength</td>
</tr>
<tr>
<td>Ending WL (nm)</td>
<td>Measurement ending wavelength</td>
</tr>
<tr>
<td>Step size (nm)</td>
<td>Motor stepping wavelength change in nm for each data point</td>
</tr>
<tr>
<td>Step Frequency (Hz)</td>
<td>Motor signal frequency</td>
</tr>
<tr>
<td>Wait time (ms)</td>
<td>Lock-in wait time for each collection</td>
</tr>
<tr>
<td>No. of Points</td>
<td>Number of collections for the Lock-in system for each data point</td>
</tr>
<tr>
<td>Measurement selector</td>
<td>Data collection selector</td>
</tr>
</tbody>
</table>


2.6 Typical Experiment Procedures

A typical experiment procedure is shown as follows:

1. Install the sample onto the platform of the cryostat system. After installing the sample, a metal mesh is put on top of sample to provide modulation voltage during a electroreflectance measurement.

2. Adjust the incident light to the certain angle and tune the position of the silicon detector on the path of the reflected light.

3. Start the vacuum pump for the cryostat system to keep the vacuum as low as $10^{-5}$ Torr.

4. Cool down the cryostat system to the desire measurement temperature whose value can vary from room temperature to 20 K.

5. Adjust the Lock-in collection parameters for measurement. For example, the Lock-in integration time for each collection and Lock-in sensitivity.

6. Turn on the signal chopper for an OR measurement, or turn on the square wave function generator and voltage amplifier in an ER measurement.

7. Calibrate the motor with the wavelength indicator on the monochromator and turn the motor to the starting wavelength.

8. Set the control parameters in the scanning program to proper values.

9. Turn room illumination light off and start measurement.

10. Save data after finishing measurement.
Chapter 3

Experimental Results and Discussions

In this chapter, major results of this dissertation have been presented and discussed. Both single quantum well (SQW) and multiple quantum well (MQW) samples with identical supercell structure are used for experimental measurements. In the beginning of this chapter, a simulation of reflection dependent on the number of quantum wells \( N \) had been presented to show the broadening effect of the reflection feature when the number of quantum wells changes. We also calculated the theoretical transition energies for different excitons, namely \( x(e_1-hh_1) \), \( x(e_1-lh_1) \), \( x(e_2-hh_2) \) etc., based on the structure of the MQW sample. After that, optical reflectance (OR), photoreflectance (PR) and electroreflectance (ER) measurements are performed both on the SQW and MQW samples for evaluating the existing exciton transitions under various conditions such as incident angle, temperature and electric field. The double resonance condition had been achieved and found to be around an incident angle of about 23° for the MQW sample. The exciton transition feature related to heavy hole exciton at the second state \( x(e_2-hh_2) \) was observed. Detailed experimental results
measured under various conditions will be presented here.

### 3.1 AlGaAs/GaAs MQW Bragg Sample Structure

The multiple quantum well structure sample used in this experiment consists of 60 GaAs quantum wells separated by AlGaAs barriers. The structure supercell consists of a GaAs QW sandwiched by relatively thin Al$_x$Ga$_{1-x}$As barriers with a high Al content $x = 74\%$. The rest major part of the barriers is AlGaAs with the Al content $x = 25\%$. The high and thin portion of the AlGaAs barriers improves localization of the excitons in the GaAs QWs. The schematic layer structure of the MQW sample and a band-gap diagram of the RBS structure is shown in Fig. 3.1. The RBS with 60 periods and the reference sample with the single supercell were grown by molecular beam epitaxy in a Riber 32 system on a semi-insulating 2-inch GaAs substrate with (001) orientation. The growth temperature and rate were 600 °C and ~1 μm/hr, respectively. The top AlGaAs barrier was covered by a thin GaAs cap in order to prevent an intensive surface oxidation when exposed to air. The samples were not intentionally doped. Detailed parameters values in Fig. 3.1 are shown in Table 3.1.
Table 3.1: Parameters of the resonant Bragg structure. (Designations of $a$, $b$, $c$, and $d$ are shown in Fig. 3.1.)

<table>
<thead>
<tr>
<th>Layer</th>
<th>Al content $x$</th>
<th>Thickness (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs cap</td>
<td>0</td>
<td>$g = 3.4$</td>
</tr>
<tr>
<td>Major AlGaAs barrier</td>
<td>0.25</td>
<td>$b = 88.9$</td>
</tr>
<tr>
<td>Thin AlGaAs barrier</td>
<td>0.74</td>
<td>$c = 5.4$</td>
</tr>
<tr>
<td>GaAs QW</td>
<td>0</td>
<td>$a = 12.8$</td>
</tr>
</tbody>
</table>

A well recognized problem of the RBS based on the AlGaAs/GaAs system is the interference and interplay of the Bragg reflections by the QW excitons and by the AlGaAs/GaAs interfaces [1, 2, 50, 59, 60, 61, 62, 63]. The former contribution is of interest for the concept of the RBS, whereas the latter one corresponds to conventional Bragg reflectors and should be suppressed by design as much as possible. For this we optimized the thickness of the AlGaAs barrier with high
aluminum content, so that the average index of refraction in the AlGaAs/GaAs/AlGaAs area is equal to the value in the major part of the AlGaAs barriers.
3.2 Simulation of Bragg Reflection Of MQW With Matched Dielectric Constants

Based on the theoretical analysis in Eq. 1.16, calculation of reflection spectra of a resonant Bragg structure of $N$ quantum wells with matched dielectric constant contrast (simple case) has been carried out in Matlab program and the results are shown in Fig. 3.2. The calculation was made for the case of normal incidence of light on the MQW structure from vacuum. In Fig. 3.2 (a), the calculations are done with no radiative damping, $\Gamma = 0$; in Fig. 3.2 (b), radiative damping constant $\hbar \Gamma = 100 \mu\text{eV}$; and in Fig. 3.2 (c), radiative damping constant $\hbar \Gamma = 1000 \mu\text{eV}$. Other parameters used for the calculation are shown in Table 3.2.

Table 3.2: Parameters for calculation of reflectance spectra $R_N$ of a heterostructure with $N$ quantum wells consisting of material A as the well layer and B as barrier. (Designation of $a$, $b$ and $b'$ are shown in Fig. 1.9)

<table>
<thead>
<tr>
<th>Calculation parameters</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>QW well width $a$</td>
<td>13.60 nm</td>
</tr>
<tr>
<td>QW barrier width $b$</td>
<td>97 nm</td>
</tr>
<tr>
<td>Cap layer width $b'$</td>
<td>103.64 nm</td>
</tr>
<tr>
<td>Exciton resonant frequency $\omega_0$</td>
<td>$\hbar \omega_0 = 1.629 \text{ eV}$</td>
</tr>
<tr>
<td>Barrier refraction index $n_b$</td>
<td>3.45</td>
</tr>
<tr>
<td>Radiative damping $\Gamma_0$</td>
<td>$\hbar \Gamma_0 = 50 \mu\text{eV}$</td>
</tr>
<tr>
<td>Non-radiative damping $\Gamma$</td>
<td>(a) $\hbar \Gamma = 0$, (b) $\hbar \Gamma = 100 \mu\text{eV}$, (c) $\hbar \Gamma = 1000 \mu\text{eV}$</td>
</tr>
</tbody>
</table>
(a) $\hbar \Gamma = 0$

(b) $\hbar \Gamma = 100 \mu eV$
Figure 3.2: Calculation of reflectance spectra $R_N$ of a heterostructure with $N$ quantum wells consisting of material A as the well layer and B as barrier. The difference of dielectric constants for two materials are ignored. The parameters used for calculations are: barrier material refraction index $n_b = 3.45$, radiative exciton damping constant $\hbar \Gamma_0 = 50 \, \mu eV$, and non-radiative exciton damping constant (a) $\hbar \Gamma = 0$, (b) $\hbar \Gamma = 100 \, \mu eV$ and (c) $\hbar \Gamma = 1000 \, \mu eV$. The numbers of quantum wells $N$ for calculation are ranging from 1 to 500.

Two special frequencies can be characterized from the simulation from all calculated spectra going through them, and have a reflectance coefficient close to that for a homogeneous bulk barrier material as indicated in these references [48, 49]. These two special frequencies are noted as $\omega_-$ and $\omega_+$ in Fig. 3.2. At these two frequencies, the reflectance $R_N = |r(N)|^2$ is equal to $r_{01}^2$ and nearly
independent of quantum well number $N$. $r_{01}^2$ is exactly the reflectance at the interface between vacuum and a semi-infinite bulk barrier material. When $n_b = 3.45$ is used, this reflectance is around $r_{01}^2 = |(1 - n_b) / (1 + n_b)|^2 = 0.303$. The special frequencies are referenced to the edges of the band gap of exciton polaritons.

The non-radiative damping $\hbar \Gamma = 0$ is the ideal case for theoretical illustration, where exciton radiative efficiency is 100% corresponding to a reflection of 1 at the exciton energy regardless of the quantum well number $N$, as shown in Fig. 3.2 (a). For ground state excitons, heavy-hole exciton $x(e1-hh1)$ and light-hole exciton $x(e1-lh1)$, a non-radiative damping $\hbar \Gamma = 100 \, \mu$eV which is twice as the radiative damping was used for calculation, as shown in Fig. 3.2 (b). The reflection at the exciton energy is still significant and increase with the quantum number $N$. For the second state excitons, such as heavy-hole exciton $x(e2-hh2)$, we expected a large non-radiative damping and $\hbar \Gamma = 1000 \, \mu$eV was used for calculation, as shown in Fig. 3.2 (c). The reflection at the second state exciton energy is much weaker compared to the ground state exciton case and increases with the quantum number $N$.

One interesting feature in Fig. 3.2 (c) for high non-radiative broadening and large quantum well number $N$ is a narrow peak in the reflection spectrum at the exciton frequency $\omega_0$. This is related to the Borrmann effect, which is the origin of extraordinary transmission of radiation through absorptive media [99]. For a resonant Bragg structure, this narrow peak at the exciton energy $\omega_0$ is due to the suppression of radiation absorption. As for $N \rightarrow \infty$, the steady-state distribution of radiation electric field at $\omega_0$ is a standing wave with its nodes located at the centers of QWs. Such a wave does not interact with excitons, leading to a narrow dip in the absorption spectrum and a narrow peak in the reflection spectrum at the resonant frequency $\omega_0$ [100].
The calculation also showed that the broadening of the reflectivity increases sharply with quantum number \( N \) of the structure for small \( N \) \((N < 100)\). When \( N \) goes to large numbers \((N > 400)\), the reflectance spectra inside the edges of the band gap are almost identical. The oscillation period of the spectra outside the band gap always decrease with the quantum well number \( N \). To see the broadening effects on the Bragg reflector with quantum well number \( N \) more clearly, the full width at half maximum (FWHM) is calculated for each reflectivity spectrum of \( N \), which is showed in Fig. 3.3. According to the explanation given by L. Pilozzi et al. [49], the behavior of the FWHM curve is usually divided into three different zones, indicated as I, II and III in Fig. 3.3. Namely, (i) in zone I the linear behavior is connected with the super-radiant regime; (ii) zone II is the transition zone, where the super-radiant mode redistributes; and (iii) zone III is the saturation zone, where the reflectivity shows the Bragg reflector behavior.

![Figure 3.3: Full width at half maximum of the reflectivity as a function of quantum well number N.](image)

Figure 3.3: Full width at half maximum of the reflectivity as a function of quantum well number \( N \).
The MQW sample used in our experiment consists with 60 periods of quantum wells. From the FWHM versus quantum well number analysis shown in Fig. 3.3, the FWHM of our sample will locate in zone II, thus we expect that there is decent strength of exciton transition features during OR and ER measurements.
3.3 X-Ray Diffractometry of MQW Sample

In order to evaluate the actual geometrical parameters including the thicknesses of all the grown layers and the Al content in the barriers, the samples were examined by the high-resolution x-ray diffractometry (XRD). The XRD plot for the RBS with 60 periods is shown in Fig. 3.4. The two major peaks originated from the reflection by (004) planes of the GaAs substrate ($\theta_{Br} = 66.055^\circ$) and of the RBS ($\theta_{Br} = 66.00^\circ$). The latter corresponds to the average lattice period over the whole structure. The periodicity of the structure gives rise to a periodic set of the diffraction fringes with additional modulation of their intensities due to the complex structure of the periodic supercells. A comprehensive analysis and fit of the experimental XRD plot was performed with a Bruker D8 instrument and LEPTOS application suit. The corresponding simulated curve is plotted in Fig. 3.4 with a vertical shift by two orders to distinguish the calculated curve from the experimental one since the simulation appears to be very close to the experiment. The best fit parameters of the structure are listed in Table 3.1. The thicknesses of all the layers were obtained from the XRD analysis with the precision of 0.1 nm. It should be noted that the XRD revealed no noticeable gradients of the layer thicknesses and Al concentrations across the structure.

The XRD measurements and analysis were done by our colleagues in Prof. Chaldyshev’s group at the Ioffe Physicotechnical Institute of Russian Academy of Sciences, St. Petersburg, Russia.
Figure 3.4: Experimental and simulated X-ray diffraction curves of the RBS with 60 periods. For clarity the simulated curve is shifted up by two orders.
3.4 Calculation of Exciton Energy of Quantum Well

Based on the sample structure shown in Table 3.1, energy levels of the electron and holes were estimated in the QW. The stationary one-dimensional Schrödinger equation was solved numerically with the Bastard’s boundary conditions [101]. The composition dependencies of the band gap and effective masses in the GaAs-AlGaAs system were taken from Adachi [102].

The 1-D time-independent Schrödinger equation can be written as

\[ -\frac{\hbar^2}{2m} \frac{d^2 \varphi}{dx^2} + V(x)\varphi = E\varphi \]  

(3.1)

where \( \hbar \) is the reduced Planck’s constant; \( m \) is the effective mass of particle; \( \varphi \) is the wave function; \( V(x) \) is the potential energy and \( E \) is the eigen energy.

Considering the potential energy \( V(x) = 0 \) inside the quantum well and \( V(x) = V_0 \) outside the quantum well. The solutions to the Schrödinger equation can be written as,

\[ \varphi = \begin{cases} 
\varphi_1 = F e^{-\alpha x} + G e^{\alpha x}, & x < -L/2 \\
\varphi_2 = A \sin(kx) + B \cos(kx), & -L/2 < x < L/2 \\
\varphi_3 = H e^{-\alpha x} + I e^{\alpha x}, & x > L/2 
\end{cases} \]  

(3.2)

where \( L \) is the quantum well width and,

\[ \alpha = \frac{\sqrt{2m(V_0 - E)}}{\hbar}, \quad k = \frac{\sqrt{2mE}}{\hbar} \]  

(3.3)
The boundary conditions applies that the Schrödinger equation and its first derivative must be continuous at the boundaries. The wave function also must goes to 0 when \( x \to \pm \infty \). The solutions have two cases, symmetric and antisymmetric. For symmetric, we have \( A = 0 \) and \( G = H \), and for antisymmetric, we have \( B = 0 \) and \( G = -H \). For symmetric case, we have

\[
He^{-\alpha L/2} = B \cos(kL/2)
\]

\[-\alpha He^{-\alpha L/2} = -kB \sin(kL/2)\]

which gives

\[
\alpha = k \tan(kL/2)
\]  

And for antisymmetric case, similarly, we have

\[
\alpha = -k \cot(kL/2)
\]  

Both \( \alpha \) and \( k \) are related to the energy and can be solved numerically. By introducing the dimensionless variables \( u = \alpha L/2 \) and \( v = kL/2 \), the final solution is to solve following equation,

\[
\sqrt{u_0^2 - v^2} = \begin{cases} 
  v \tan v, & (\text{symmetric}) \\
  -v \cot v, & (\text{antisymmetric}) 
\end{cases}
\]  

noting that

\[
u_0^2 = u^2 + v^2 \\
u_0^2 = mL^2V_0/2h^2
\]
and the eigen energy and be written as

\[ E_n = \frac{2\hbar^2 v_n^2}{mL^2} \]  

(3.9)

To solve Eq. 3.7 numerically is actually to find the intersection of the two curve on both sides of Eq. 3.7, respectively, \( \sqrt{u_0^2 - v^2} \) and \( v \tan v \) or \( -v \cot v \). A typical solution is shown in Fig. 3.5. The black curves correspond to \( v \tan v \) or \( -v \cot v \), and the red curve is \( \sqrt{u_0^2 - v^2} \). The numerical solution are the intersections of the curves in the Fig. 3.5.

![Figure 3.5](image)

Figure 3.5: Numerical solutions for 1-D quantum well structure with finite barrier, (a) electron, (b) light hole and (c) heavy hole. The black curves correspond for calculation of \( \sqrt{u_0^2 - v^2} \); the blue curves correspond for calculation of \( v \tan v \) and the red curves correspond for calculation of \( -v \cot v \). The numerical solutions are the intersections of the curves.

The calculation shows that there are 4 quantum states for electrons, 5 quantum states for light holes and 12 quantum states for heavy holes in the QW. With the exciton binding energies taken from Reference [103], we calculated the energies of all the allowed excitonic transitions. The
detailed parameters used for calculation are listed in Table 3.3. The calculation results and schematic drawing of the QW exciton transition energies allowed by symmetry are listed in Table 3.4 and plotted in Fig. 3.6.

Table 3.3: Parameters used for calculation of QW exciton transition energies.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value used</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs band gap (20 K)</td>
<td>$E_g(GaAs) = 1.518$ eV</td>
</tr>
<tr>
<td>AlGaAs aluminum content</td>
<td>$x = 0.74$</td>
</tr>
<tr>
<td>AlGaAs direct band gap (20 K)</td>
<td>$E_g(AlGaAs) = 2.5753$ eV</td>
</tr>
<tr>
<td>Quantum well width</td>
<td>$L = 12.5$ nm</td>
</tr>
<tr>
<td>Conduct band discontinuity</td>
<td>$\Delta E_c = 0.5$ eV</td>
</tr>
<tr>
<td>Valence band discontinuity</td>
<td>$\Delta E_v = 0.5573$ eV</td>
</tr>
<tr>
<td>Exciton binding energy</td>
<td>$E_B = 8.5$ meV</td>
</tr>
<tr>
<td>Effective mass of electron</td>
<td>$m_e = 0.067$</td>
</tr>
<tr>
<td>Effective mass of light hole</td>
<td>$m_{th} = 0.087$</td>
</tr>
<tr>
<td>Effective mass of heavy hole</td>
<td>$m_{hh} = 0.62$</td>
</tr>
</tbody>
</table>
Table 3.4: Calculated values of QW exciton transition energies.

<table>
<thead>
<tr>
<th>Exciton states</th>
<th>Calculated values (eV)</th>
<th>Exciton states</th>
<th>Calculated values (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>x(e1-hh1)</td>
<td>1.539</td>
<td>x(e1-hh3)</td>
<td>1.567</td>
</tr>
<tr>
<td>x(e1-lh1)</td>
<td>1.557</td>
<td>x(e1-lh3)</td>
<td>1.724</td>
</tr>
<tr>
<td>x(e2-hh2)</td>
<td>1.627</td>
<td>x(e3-hh1)</td>
<td>1.742</td>
</tr>
<tr>
<td>x(e2-lh2)</td>
<td>1.697</td>
<td>x(e3-lh1)</td>
<td>1.760</td>
</tr>
</tbody>
</table>

\[\Delta E_c = 0.5 \text{ eV}\]

\[E_{g \text{ (GaAs)}} = 1.518 \text{ eV}\]

\[E_{g \text{ (AlGaAs)}} = 2.575 \text{ eV}\]

\[\Delta E_v = 0.557 \text{ eV}\]

Figure 3.6: Schematic drawing of QW exciton transition energies according to the theoretical calculation. The purple lines are the electron energy levels; the blue lines are the heavy hole energy levels and the red lines are the light hole energy levels. Some allowed exciton transitions are marked by arrows and numbers.
3.5 Optical Reflectance (OR) Spectroscopy

In this section, the optical reflectance (OR) measurements and results are discussed for both SQW and MQW samples. The SQW sample (named as 7787) was grown under identical conditions as MQW sample and was prepared as a reference sample. The comparison between a SQW and a MQW sample gives a better understanding how excitonic transitions behaves with or without a Bragg enhancement which only occurs in the MQW sample with a periodic structure. The OR measurements were taken by varying conditions such as tuning the incident angle, sample temperature.

The schematic drawing of experimental setup for optical reflectance measurements is shown in Fig. 2.1. In this experiment, 0.4 to 0.8 nm bandpass (monochromator front slit width 0.1 to 0.2 mm) was used for probing light, which has enough accuracy for measurement. In the meantime it provides decent amount of light for significant transition features in the OR spectroscopy.

Optical reflectance is not a modulation spectroscopy technique as long as no sample parameter is modulated during the measurement. A chopper is used at certain frequency on the incident side to give the Lock-in amplifier the reference to pick up the useful reflected signal. The chopper passes/blocks the incident light periodically at set frequency and Lock-in amplifier will lock in the signal received on the silicon detector exactly at the same frequency as chopper. Lock-in amplifier then evaluates the difference of the incident light passed through and blocked by the chopper. This simple technique will significantly eliminate environment constant noise in the final outcome since it will be canceled in the difference evaluation by the Lock-in amplifier. In this experiment, the
chopper was set at a frequency of 100 Hz.

### 3.5.1 Optical reflectance of SQW sample

An optical reflectance (OR) spectrum of a single quantum well (SQW) sample (Sample number 7787) measured at temperature 4.2 K with normal incident light is shown in Fig. 3.7. The scan range is of photon energy from 1.4 to 1.9 eV. The measurement was done at the Ioffe Institute, St. Petersburg, Russia.

![Optical reflectance spectrum](image)

**Figure 3.7:** The optical reflectance of the SQW sample measured at temperature 4.2 K with normal incident light. Transition energies are marked with arrows.

From Fig. 3.7, we can see the whole OR spectrum has a base reflection around 0.3. This
average flat reflectivity can be accounted for by the Fresnel reflection,

\[
R = \frac{R_S + R_P}{2} = \frac{1}{2} \left\{ \frac{\cos \theta - n \sqrt{1 - \left(\frac{\sin \theta}{n}\right)^2}}{\cos \theta + n \sqrt{1 - \left(\frac{\sin \theta}{n}\right)^2}} \right\}^2 + \left\{ \frac{\sqrt{1 - \left(\frac{\sin \theta}{n}\right)^2} - n \cos \theta}{\sqrt{1 - \left(\frac{\sin \theta}{n}\right)^2} + n \cos \theta} \right\}^2
\]  

(3.10)

where \( \theta \) is the incident angle. Applying an average reflection index of the MQW dielectric material \( n = 3.45 \), the Fresnel reflection equation gives the base reflection from the sample \( R = 0.303 \), when the incident angle \( \theta = 0^\circ \). This agrees with our experimental data very well.

Transition features due to GaAs substrate, ground state excitons \( x(e_1-hh_1) \) and \( x(e_1-lh_1) \), and AlGaAs barrier are marked with arrows in Fig. 3.7. The GaAs substrate transition feature is around 1.518 eV. This agrees very well with the temperature dependence calculation of the GaAs band gap, which is given by the following equation by J. S. Blakemore [104],

\[
E_g = 1.519 - 5.408 \times 10^{-4} \times T^2 / (T + 204)
\]  

(3.11)

Above the GaAs substrate band gap, two weak transition features due to ground state exciton transitions can be identified, respectively \( x(e_1-hh_1) \) and \( x(e_1-lh_1) \), which are located around 1.544 eV and 1.554 eV. The experimental result is very close to the theoretical prediction as shown in Table 3.4. Around photon energy of 1.843 eV, the transition feature due to AlGaAs barrier can be clearly seen.

The energy of heavy-hole exciton transition at its second state \( x(e_2-hh_2) \) predicted by theory is around 1.627 eV. We did not see any transition features around this area in the SQW OR spectrum. This can be accounted for that the transition features in the SQW OR spectrum are so
weak especially the x(e2-hh2), which is totally buried by the noise in the final spectrum. This is the reason a resonant Bragg structure MQW sample is introduced with its resonant frequency near the x(e2-hh2) transition energy for specific incident angle. And in the following experiments, we will present the OR measurement on the resonant Bragg structure sample.

### 3.5.2 Optical reflectance of MQW sample with different incident angles

In order to find the exact incident angle for the double resonance condition that Bragg reflection frequency meets the 2\textsuperscript{nd} heavy exciton transition x(e2-hh2), an angle dependent OR measurement had been carried out. In this measurement, all other parameters such as temperature, incident light intensity, chopper frequency \textit{etc}. were kept the same except the probing light incident angle. By doing this, we were able to tune the Bragg reflection frequency while the 2\textsuperscript{nd} heavy hole transition x(e2-hh2) energy is not changed. At certain incident angle, the double resonance condition was satisfied and we would be able to see an overlap of Bragg reflection peak with the 2\textsuperscript{nd} heavy hole exciton transition x(e2-hh2), exhibiting an enhanced and broadened spectrum feature in OR. Our experiment showed that the double resonance condition occurred around an incident angle of about 23°.

Figure 3.8 shows the OR spectra from the RBS with 60 QWs recorded at 4.2 K at different angles of the light incidence with s-polarization. The incident angles are 0°, 23°, 45° and 68°. The experiment data are plotted as solid lines in all panels. From all the spectra of different incident angles, we can see that the background reflection increase in s-polarization probing light when the incident angle approaches the Brewster’s angle $\theta_B = 74^\circ$. This is calculated by the Brewster’s law,
giving by equation,

\[ \theta_B = \arctan(n) \]  \hspace{1cm} (3.12)

taking the refraction index of our MQW sample \( n = 3.45 \).

![Optical reflection spectra](image)

Figure 3.8: Optical reflection spectra from the RBS with 60 QWs recorded at 4.2 K for \( s \)-polarized light incident at (a) 0°, (b) 23°, (c) 45°, and (d) 68°—black solid lines. The dash line represents the calculated reflection spectra. The dot line shows the calculated reflection spectra without the exciton contribution. The vertical dot line marks the energy of the \( x(e2-hh2) \) excitons.
The major reflection peak in Fig. 3.8 is due to the Bragg reflection in accordance with the Bragg reflection law,

\[ 2d\sqrt{n^2 - \sin^2\theta} = m\lambda \]  

(3.13)

and is shifting to higher frequency with increasing incident angle. The OR feature due to the QW heavy hole excitons in the second quantum state \( x(e2-hh2) \) is clearly seen under the normal light incidence. Its energy is marked by a vertical dot line passing all panels in Fig. 3.8. At the incident angle of around 23°, the energy of the \( x(e2-hh2) \) exciton coincides with the photon energy found from the Bragg resonance condition for this RBS. This gives rise to a single strong peak of resonant reflection. At larger incident angles, the \( x(e2-hh2) \) exciton energy no longer satisfies the Bragg resonance condition and the peak in the OR spectra diminishes.

The observed behaviors in the OR spectra are supported by the calculations made in the transfer matrix approximation (see reference [51] for details). The calculated spectra (the dash curves) are also plotted in Fig. 3.8 for each examined angle of the light incidence. With the exciton energy set to 1.629 eV, radiative broadening \( \Gamma_0 = 40 \mu eV \), and non-radiative broadening \( \Gamma = 3 \text{ meV} \), the calculated curve fits the experimental data very well.

In order to reveal the exciton contribution to the aggregate resonant optical reflection under the double resonance conditions realized at 4.2 K and incident angle of 23°, we also made model calculations for a virtual system of 60 QWs without excitons. In this case, the Bragg reflectivity is still possible due to the difference between the background dielectric permittivity of the barriers and quantum wells. The calculated curve neglecting the \( x(e2-hh2) \) resonance (the dot curve in panel b
of Fig. 3.8) significantly deviates from the experimental data around the peak of \( x(e_2-hh_2) \). These comparisons of the experimental and calculated spectra show that the double resonance condition strongly enhances the exciton-mediated optical reflection.

Fig. 3.9 shows a complete scan of OR spectra on the RBS MQW sample versus varies incident angles, ranging from 11° to 59° with a 2° step. The exciton transition energies are marked as vertical dash lines in the Fig. 3.9. Around the second state heavy hole exciton transition \( x(e_2-hh_2) \), the major reflection peak is due to the Bragg reflection. As the incident angle increases, we can clearly see that the Bragg reflection peak shifts to higher frequency. When the Bragg reflection meets the \( x(e_2-hh_2) \) transition energy, we see a strong enhancement and the OR spectrum gets its maximum value at this point. This coincidence, or double resonance condition appeared at the incident angle of about 23°, as indicated by the purple line spectrum in Fig. 3.9.
Figure 3.9: Optical reflection spectra from the RBS MQW sample with 60 QWs measured at 20 K at various incident angles, ranging from 11° to 59° with a 2° step for each record. The exciton transitions, namely x(e1-hh1), x(e1-lh1) and x(e2-hh2) are marked with dash lines. (The Y axis label is account for the top most spectrum and other spectra are stacked in order.)
### 3.5.3 Optical reflectance of MQW sample at double resonance condition

Compared to the OR spectrum on the SQW sample, the OR reflectance on the MQW sample with same dielectric structure exhibits much more rich and broadened features related to both the bulk semiconductor materials and exciton transitions inside the structure.

The MQW sample used in our experiment was intentionally designed and fabricated so that makes it a special RBS sample with its Bragg resonance frequency coinciding with the second state heavy hole exciton transition \( \chi(e2-hh2) \) inside the structure at certain incident angle. This coincidence of Bragg resonance and exciton transition could enlarge the effect both in OR and ER spectroscopies, exhibiting as enhanced reflectivity and broadened transition features. Such a coincidence is always called a double resonance.

Figure 3.10 shows a typical optical reflectance spectrum on the RBS MQW sample with 60 QWs periods. The measurement was taken at temperature of 20 K and probing light incident angle of 23°. The baseline reflection is around 0.3 and well agrees with the theoretical prediction as of the SQW sample, since the materials for a single super cell for MQW and SQM samples are identical. At incident angle of 23°, the double resonance condition occurred that the Bragg resonance frequency was tuned exactly at the same frequency of the 2\(^{nd} \) heavy hole exciton transition \( \chi(e2-hh2) \) in the RBS MQW sample. Under this circumstances, we had an overlap of the Bragg reflection peak with the \( \chi(e2-hh2) \) exciton transition feature, thus exhibiting a much broadened and strong reflectance feature in the OR spectrum. This peak was fitted by third derivative of Gaussian line shape, indicated as the green line in Fig. 3.10. The energy position from the fitting is around
1.625 eV which agrees quite well with our theoretical calculation of the x(e2-hh2) transition in the RBS MQW sample at temperature 20 K.

![Optical Reflectance Spectrum](image)

Figure 3.10: A typical optical reflectance spectrum from the RBS MQW sample with 60 QWs periods, at temperature 20 K and incident angle 23°. Double resonance condition occurred under incident angle of 23° exhibiting an overlap of Bragg reflection peak with the 2nd heavy exciton transition x(e2-hh2) around 1.625 eV. Features due to the bulk GaAs band gap, 1st light and heavy hole exciton transitions are also can be seen in the OR spectrum, curve fitted and noted by arrows in the figure.

Besides the double resonance reflection band, other bulk material and QW exciton features are also identified in the MQW OR spectrum. The feature fitted with a red line in Fig. 3.10 is related to the bulk GaAs band gap transition, which is located around 1.520 eV. The theoretical
calculation showed that bulk GaAs has a band gap of 1.518 eV at temperature 20 K which is close to the results of the fitting. Above the GaAs band gap, we also identified two features, related to heavy hole x(e1-hh1) and light hole x(e1-lh1) excitons in QWs, respectively. The fitting showed the corresponding transition energies to be 1.544 eV and 1.555 eV, respectively. Our quantum mechanics calculation gave 1.539 eV for the x(e1-hh1) and 1.557 eV for the x(e1-lh1) exciton transition energies, as indicated in Table 3.4. The OR transition feature around photon energy of 1.587 eV was also fitted as indicated by a green fitting curve in Fig. 3.10. We believe this transition could be due to the ground state electron and 3rd state heavy hole exciton x(e1-hh3), whose theoretically calculated value was 1.567 eV. The detailed fitting results and corresponding theoretical calculations are shown in Table 3.5. A 2nd derivative or 3rd derivative Gaussian line shape was used for fitting depending on the symmetry of the transition feature in the OR spectrum.

Table 3.5: Exciton transition energies from OR spectrum on a 60 QWs RBS MQW sample and the corresponding theoretically calculated values.

<table>
<thead>
<tr>
<th>Exciton state</th>
<th>OR spectrum data (eV)</th>
<th>Calculated value (eV)</th>
<th>Fitting method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk GaAs</td>
<td>1.520</td>
<td>1.518</td>
<td>3rd derivative Gaussian</td>
</tr>
<tr>
<td>x(e1-hh1)</td>
<td>1.544</td>
<td>1.539</td>
<td>3rd derivative Gaussian</td>
</tr>
<tr>
<td>x(e1-lh1)</td>
<td>1.555</td>
<td>1.557</td>
<td>2nd derivative Gaussian</td>
</tr>
<tr>
<td>x(e1-hh3)</td>
<td>1.587</td>
<td>1.567</td>
<td>2nd derivative Gaussian</td>
</tr>
<tr>
<td>x(e2-hh2)</td>
<td>1.625</td>
<td>1.627</td>
<td>3rd derivative Gaussian</td>
</tr>
</tbody>
</table>
Figure 3.11: Optical reflection (OR) (a, b) and photoluminescence (PL) (c) spectra at normal light incidence from the samples with a single QW and 60 QWs recorded at 4.2 K of light reflection and emission. The PL for 1 QW is multiplied by 10 and shifted up by 0.2 for better visibility. Arrows mark excitonic transitions in GaAs QWs, GaAs substrate, and AlGaAs barriers.
Figure 3.11 shows the PL and OR spectra measured at 4.2 K from the RBS with 60 QWs and the reference sample with a single quantum well (SQW). The PL spectra are very similar showing two major peaks. One of them at 1.82 eV corresponds to the emission from the major AlGaAs barrier. The other peak at 1.543 eV originates from the excitonic states in the QWs. Due to a low excitation power, we observed only the ground state of the heavy-hole excitons associated with the first quantum levels of heavy holes and electrons x(e1-hh1).

The OR signal at an average level of about 0.3 was observed for both RBS and SQW samples. Since no antireflection coating was utilized, this OR is originated from the air-sample interface and it is well consistent with the results of calculations by the Fresnel formulas. The variations from this smooth reflectivity indicate structural features and exciton resonances in the samples. The most important excitonic lines are marked by arrows in Fig. 3.11. Two strongest features in the OR spectrum from the reference SQW sample in Fig. 3.11 correspond to the excitons in the GaAs substrate and AlGaAs barrier. Our quantum mechanical calculations for the studied QWs predicted three quantum levels for electrons and light holes and six quantum levels for heavy holes. However, only three exciton transitions related to the QWs can be seen in the spectra, namely, the ground-state excitons formed by electrons and heavy holes in their first quantum state x(e1-hh1), by electrons and light holes in their first quantum state x(e1-lh1), and by electrons and heavy holes both in their second quantum state x(e2-hh2). The single QW provides two relatively weak but well detectable peaks at 1.542 eV and 1.550 eV due to x(e1-hh1) and x(e1-lh1) quasi-2D excitons respectively. The experimentally determined energies are in a very good agreement with the calculated ones (Table 3.5). Other allowed excitonic transitions were not detected in the SQW sample.

The OR spectrum from the RBS structure is more complicated due to the Bragg reflection
band with multiple satellites. The energy of the major Bragg peak under the normal incidence is 1.610 eV. The \( x(e_1-hh_1) \) and \( x(e_1-lh_1) \) excitonic states are revealed as two sharp dips at the energies being equal to those for the SQW sample. Additionally, the OR spectrum from the RBS shows quite a strong peak at 1.628 eV, which is due to the \( x(e_2-hh_2) \) excitonic transition. The experimentally determined energy appeared close to the calculated one. Other possible allowed excitonic transitions, namely \( x(e_2-lh_2), x(e_1-hh_3), \) and \( x(e_2-hh_4) \), are not detectable in the experimental OR spectrum. Noticing that since the incident angle is 0° in this measurement, the double resonance condition is not satisfied anymore thus makes the Bragg reflection peak away from the \( x(e_2-hh_2) \) transition feature.

From Fig. 3.11, the estimated FWHM of exciton transition features in the OR spectra for both SQW and MQW samples are listed in Table 3.6. By comparing the FWHM for SQW and MQW at different exciton transitions, we found that the exciton transitions in MQW sample have only slightly bigger line widths. The small difference of line width indicates that our RBS MQW sample has very small inhomogeneity. This also agrees with our XRD measurement result of the sample.

Table 3.6: Estimated FWHM for exciton transition features of SQW and MQW samples in OR spectra.

<table>
<thead>
<tr>
<th></th>
<th>( x(e_1-hh_1) )</th>
<th>( x(e_1-lh_1) )</th>
<th>( x(e_2-hh_2) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>SQW (meV)</td>
<td>2.4</td>
<td>2.9</td>
<td>Not observed</td>
</tr>
<tr>
<td>MQW (meV)</td>
<td>2.9</td>
<td>3.4</td>
<td>4.9</td>
</tr>
</tbody>
</table>
3.5.4 Optical reflectance of MQW sample with different temperature

The temperature dependencies of the normal OR from the RBS are plotted in Fig. 3.12. With increasing temperature, the excitonic transition energies are decreasing in accord to the Varshni’s relation [105],

\[ E_g(T) = E_g(0) - \frac{\alpha T^2}{T + \beta} \]  

(3.14)

where the \( \alpha \) and \( \beta \) coefficients for GaAs and AlGaAs band gaps were determined in Ref. [31]. The red shift of the excitonic features with temperature is accompanied by enhanced broadening. The \( x(e1-hh1) \) and \( x(e1-lh1) \) excitonic features gradually disappear and become hardly detectable when the temperature is higher than 200 K.
Figure 3.12: Optical reflection spectra from the RBS MQW sample with 60 QWs measured at normal light incidence and varying temperature.

The temperature dependence of the $x(e_2-hh_2)$ excitonic line is more complicated. With the temperature elevating from 10 to 150 K, the energy of this transition becomes closer to the energy of the Bragg resonance. At 150 K, the $x(e_2-hh_2)$ exciton energy and the energy of the Bragg resonance coincide, so the double resonance condition is met. At this point Fig. 3.12 shows an excitonic enhancement of the Bragg reflection (from 48.6% at 10 K to 57.3% at 150 K) or, in other words, a Bragg enhancement of the exciton interaction with light. Further increase in the temperature detunes the two resonances, and the $x(e_2-hh_2)$ excitonic feature disappears from the OR spectra. At room temperature the excitonic states are not detectable in OR spectra due to a strong non-
radiative broadening, whereas the Bragg resonance is much less affected by temperature.

From the OR spectra of temperature dependence, we were able to identify the \(x(e_2-hh_2)\) transition energy location clearly when the temperature below 150 K. This transition energy with the relationship to temperature was then analyzed using Varshni’s equation and the result is shown in Fig. 3.13. From Fig. 3.13, we can see that the experiment data is well fitted by the Varshni’s relation, and the parameters used in the fitting are: \(E_g(0) = 1.631\), \(\alpha = 4.966E - 4\) and \(\beta = 242\). These values are very close to those in Blakemore’s equation for GaAs band gap with temperature dependence, as shown in Eq. 3.11.

![Figure 3.13](image)

Figure 3.13: 2\textsuperscript{nd} state heavy hole exciton \(x(e_2-hh_2)\) transition energy with dependency on temperature. The red curve is the fitting according to Varshni’s equation.
3.5.5 Discussion

We applied OR spectroscopy technique for both SQW and MQW samples. In the SQW OR spectrum, the exciton transition features due to the ground state heavy-hole exciton \( x(e_1-hh_1) \) and light-hole exciton \( x(e_1-lh_1) \) exhibit weak spectrum features and can just be recognized. However, the heavy-hole exciton at the second state \( x(e_2-hh_2) \) can not be observed in the SQW OR spectrum. While in the RBS MQW OR measurement, the exciton transitions are significantly strengthened. This can be well explained by our simulation of Bragg reflection of MQW. Compared to SQW, when MQW structure with N QWs is presented, the exciton radiative broadening parameters becomes N times that of the exciton in a single quantum well, thus the reflection coefficient in Eq. 1.13 for N QWs can be re-written as,

\[
r_N(\omega) = \frac{iN\Gamma_0}{\omega_0 - \omega - i (N\Gamma_0 + \Gamma)}
\]

(3.15)

and excitons become more photon active in MQW structure.

The oscillator strength and binding energy of the \( x(e_2-hh_2) \) excitons should be almost the same as that of the \( x(e_1-hh_1) \) excitons. Then we can expect almost equally effective electromagnetic coupling in the periodic systems of both excitons. This prediction is supported by the results of fitting the experimental OR spectra, which gives the parameter of radiative broadening of the \( x(e_2-hh_2) \) excitons \( \Gamma_0 \) equal to 40 ± 5 \( \mu eV \), close to the value of 37.5 \( \mu eV \) as previously determined for the \( x(e_1-hh_1) \) excitons in an AlGaAs/GaAs/AlGaAs system [2].

While the electromagnetic coupling is almost equally effective for the excitons at different
quantum states, the non-radiative broadening processes are substantially different because of additional relaxation mechanisms for upper levels. In our samples, the parameter of non-radiative broadening (one half of FWHM) appeared to be 1 meV for the x(e1-hh1) state and 3 meV for the x(e2-hh2) state.

Considering in the SQW OR spectrum, this radiative broadening parameter \( \Gamma_0 = 40 \mu \text{eV} \) is extremely small compared to the non-radiative broadening parameter \( \Gamma = 3 \text{ meV} \), leading to a low optical process efficiency. This explains why we did not observe the second state exciton transition in a SQW OR measurement. In the MQW OR measurement under double resonance condition, taking that the QW number \( N = 60 \) for our sample, this radiative broadening parameter is enlarged by 60 times \( N \Gamma_0 = 2.4 \text{ meV} \) and becomes comparable to the non-radiative broadening parameter \( \Gamma = 3 \text{ meV} \). In this circumstances, the transition due to the heavy-hole exciton at the second state x(e2-hh2) is significantly enhanced and can be clearly identified in the OR spectrum, as shown in Fig. 3.8. Our calculation showed approximately a 0.25 Bragg reflection peak with no exciton effect and this reflection becomes around 0.36 with the effect of the heavy-hole exciton at the second state x(e2-hh2) under the double resonance condition, as shown in Fig. 3.8, panel (b). This gives an overall enhancement of about 44% to the OR transition feature. This enhancement under the double resonance condition is significant and makes it possible to identify the transition of the heavy-hole exciton at the second state x(e2-hh2).

To achieve the double resonance condition, we applied two ways to manage it. The first method is to tune the incident angle. This way we change Bragg reflection frequency while maintain the exciton energy. At certain incident angle, the Bragg reflection resonance coincides with the exciton energy, typically the heavy-hole exciton at the second state x(e2-hh2) in our experiment,
and the double resonance condition is achieved. In our experiment, this incident angle is proved to be around 23° for temperature of 20 K. Another way to achieve double resonance condition is to tune the temperature. In this way, we change the exciton energy while maintain the Bragg reflection frequency, as shown in the temperature dependent OR measurement in Fig. 3.12. For a normal incidence measurement, the double resonance condition occurs around temperature of 150 K.
3.6 Photoreflectance (PR) Spectroscopy

Photoreflectance spectroscopy also known as optical modulation spectroscopy is considered to be a modulation technique which evaluate the changes in the optical response from the sample under a periodic modulated field affected by a chopped pumping laser. In this section, the photoreflectance measurement and result is discussed for the MQW sample. The schematic drawing of our setup for photoreflectance measurements is shown in Fig. 2.2.

3.6.1 PR measurement from the MQW sample

A typical photoreflectance spectrum from the RBS MQW sample with 60 QWs is shown in Fig. 3.14. The measurement was taken at temperature 20 K and an incident angle of about 23°. Compared to the optical reflectance (OR) spectroscopy, the \(x(e_1-hh_1)\), \(x(e_1-lh_1)\) and \(x(e_2-hh_2)\) exciton transitions exhibit themselves as much sharper and derivative-like spectrum features, which is due to the technique of modulation spectroscopy. The baseline of the PR spectrum locates around 0 value because that modulation spectroscopy eliminates those constant background noises during evaluation. The most significant PR feature (the biggest peak) is due to the \(x(e_1-hh_1)\) exciton transition, which is located around 1.54 eV. Above that, we also can clearly identify the \(x(e_1-lh_1)\) exciton transition. The \(x(e_2-hh_2)\) exciton transition is located around energy 1.628 eV from the PR spectrum.
3.6.2 Discussion

Photoreflectance spectrum exhibits sharp and strong features for exciton transitions. The ground state exciton transitions $x(e1-hh1)$ and $x(e1-lh1)$ are dominant in the PR spectrum, especially for the heavy-hole ground state exciton $x(e1-hh1)$. Along with that, transition feature related to the heavy-hole exciton at the second state $x(e2-hh2)$ can be clearly identified. Comparing to the OR spectrum, no transition feature related to the GaAs substrate was observed in the PR measurement. This is because the pumping laser in the PR measurement could not go deep into the GaAs substrate, resulting no modulation in the GaAs substrate of the RBS sample.
One weakness about the photoreflectance technique is that it is very hard to control the noise level since it is difficult to prevent pumping laser scattering and reflection on device surface coming into the detector. This can also be noticed in Fig. 3.14 that decent amount of noise are still present in the PR spectrum. We used different pumping lasers with varying power and found that a 5 mW power laser produces decent amount of transition features during the PR measurement and at the meantime keeps the noises at a low level.
3.7 Electroreflectance (ER) Spectroscopy

The key point in PR measurement is always how to decrease the unwanted laser noises meanwhile maintain enough transition signals. Lowering the pumping laser power will decrease the unwanted scattering and reflection laser noise, but on the other hand this also lowers the amount of exciton excitement thus produces less transition features.

Compared to photoreflectance (PR) and its background noise presence, electroreflectance (ER) spectroscopy technique has a great advantage in producing modulation spectroscopy at a much lower noise level. This is due to an external electric field is used for modulation in ER measurement instead of a pumping laser, which won’t produce any noise signals. ER measurement can be achieved in several way, including contact and contactless modes. In this experiment, the later (also known as CER) is used by mounting an metal mesh on the surface of the MQW sample to produce the required modulation voltage. In this way, we don’t need to change the sample structure and there would be no damage to the sample either.

In this section, the electroreflectance (ER) measurements and results are discussed for both SQW and MQW sample, with varies dependency conditions, such as incident angle, temperature, electric field and polarization. The schematic drawing of setup for electroreflectance measurements is shown in Fig. 2.3. In this experiment, 0.4 to 0.8 nm bandpass (monochromator front slit width 0.1 to 0.2 mm) was used for probing light, which has enough accuracy for measurement. In the meantime it provides decent amount of light for significant transition features in the final spectroscopy.
3.7.1 Electroreflectance from single quantum well sample

Figure 3.15 shows a typical ER spectrum from the SQW structure as a reference sample. The measurement was taken at the incident angle of about 23° and temperature of 20 K. The transition features are marked with arrows in the figure.

![Graph showing ER spectrum](image)

Figure 3.15: The electroreflectance (ER) spectrum measured on a single quantum well (SQW) sample (sample number 7787), at incident angle of 23°. The measurement is taken at temperature of 20 K, and a front slit width of 0.2 mm from the monochromator.

From the SQW ER spectrum, one can see a strong feature due to the bulk excitons in the
GaAs substrate, which is located around transition energy of 1.518 eV and agrees with our previous calculation for GaAs band gap at 20 K. Above the GaAs band gap, Franz-Keldysh oscillation also can be seen. A weak feature related to the x(e1-hh1) exciton state in the QW structure can be just recognized which is located around transition energy 1.544 eV and well agrees with our theoretical prediction. Higher possible exciton transitions such as x(e1-lh1) and x(e2-hh2) are below noise level and cannot be recognized. It should be noted that the first-order Stark effect is zero in symmetrical QWs and the second-order Stark effect manifests itself in ER spectra due to the existence of a bias in the system [106].

3.7.2 Electroreflectance of multiple quantum well sample

Compared to the ER spectrum of the SQW sample, the ER reflectance on the RBS MQW sample exhibits tremendously rich and strong features related to exciton transitions inside the structure. In the SQW ER spectrum, we can just recognize a small exciton transition feature related to the x(e1-hh1) exciton. For those of x(e1-lh1) and x(e2-hh2) excitons with higher transition energies, it was mostly below the noise level and can hardly be identified. On the other hand, the ER spectrum of the RBS MQW sample showed a number of strong features related to the x(e1-hh1), x(e1-lh1) and x(e2-hh2) exciton states in the QWs.
Figure 3.16: The electroreflectance (ER) spectrum measured on the RBS multiple quantum well (SQW) sample (sample number 7789) with 60 QWs, at incident angle of 23°. The measurement is taken at temperature of 20 K, and a front slit width of 0.2 mm from the monochromator. Red line is the transition features fitting using Aspnes formula [107] and fitting energies corresponding to exciton transitions are marked with arrows. The peak number notations above the GaAs substrate band gap refers to the Franz-Keldysh oscillation (FKO).

Figure 3.16 is a typical contactless electroreflectance spectrum recorded at 20 K, at the probing light incident angle of about 23° from the RBS MQW sample with 60 QWs. From the spectrum, strong transition features related to the x(e1-hh1), x(e1-lh1) and x(e2-hh2) exciton transitions can
be clearly recognized. The left most spectrum feature is due to GaAs substrate band gap, which is located around 1.517 eV and very close to the theoretical calculation value of 1.518 eV. Above the GaAs substrate band gap, a series of Franz-Keldysh oscillations coming from the substrate can be observed with its extremes marked by indexes in Fig. 3.16. After that, we see a strong feature related to the x(e1-hh1) exciton transition exhibiting as a symmetric Lorentz line shape. The x(e1-lh1) exciton transition feature can also be seen above the x(e1-hh1) exciton state. On the right part of the ER spectrum, the most significant and broadened feature is related to the x(e2-hh2) exciton transition. Due to the special structure design of the RBS MQW sample, we have the double resonance condition around the second state of electron heavy hole transition. This double resonance broadened and strengthened the exciton transition feature tremendously, as can be seen from Fig. 3.16.

In order to extract the exciton transition energies from the ER spectrum, the experiment data has been fitted with a series of Aspnes equation [107] as follows,

$$\frac{\Delta R}{R}(E) = \text{Re} \left[ Ce^{i\theta} (E - E_g + i\Gamma)^{-n} \right]$$  \hspace{1cm} (3.16)

where $C$ is the amplitude and $\theta$ is the phase factor different between the modulation signal and probing light. $E_g$ is the transition energy and $\Gamma$ is the broadening parameter of the transition. The number $n$ is also known as the critical point type and usually has a value greater than 2. Usually, $n = 3$ corresponds to two-dimensional excitons and $n = 2.5$ corresponds to three-dimensional exciton transition line shapes. In our experiment, $n = 2$ and first derivative of Lorentzian line shape fitting was used. The fitting result is plotted as red line in Fig. 3.16 and typical fitting energies are marked with arrows in the figure.
By fitting with first derivative Lorentzian line shape, we were able to estimated the exciton transition energies from the ER spectrum. The fitting values of exciton transition energies for x(e1-hh1), x(e1-lh1) and x(e2-hh2) exciton are 1.546, 1.557 and 1.628 eV, respectively. These values agree with our theoretical calculations very well, which can be seen from Table 3.7.

Table 3.7: Exciton transition energies from ER spectrum fitting and the corresponding theoretically calculated values.

<table>
<thead>
<tr>
<th>Exciton state</th>
<th>ER spectrum fitting (eV)</th>
<th>Calculated value (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>x(e1-hh1)</td>
<td>1.546</td>
<td>1.539</td>
</tr>
<tr>
<td>x(e1-lh1)</td>
<td>1.557</td>
<td>1.557</td>
</tr>
<tr>
<td>x(e2-hh2)</td>
<td>1.628</td>
<td>1.627</td>
</tr>
</tbody>
</table>

The presence of the Franz-Keldysh oscillations (FKO) in the ER spectrum is due to the intermediate electric field inside the dielectric semiconductor MQW sample, and its oscillation period is related to the strength of the internal electric field [71, 92, 108]. The relationship between the FKO period and electric field can be written as,

\[
m\pi = \phi + \frac{4}{3} \left( \frac{E_m - E_g}{\hbar \theta} \right)^{3/2}
\]

(3.17)

where \( m \) is the index of the \( m^{th} \) extreme of FKO and \( E_m \) is the photo energy corresponding to the \( m^{th} \) extreme. \( \phi = (\pi/4)(d - 1) \) with \( d \) the critical point of dimensionality. \( E_g \) is the band gap
energy and $h\theta$ is the electro-optic energy give by,

$$(h\theta)^3 = \frac{e^2 \hbar^2 F^2}{2\mu}$$  \hspace{1cm} (3.18)

where $\mu$ is the reduced mass and $F$ is the electric field.

Based on Eq. 3.17, a plot of value $(4/3\pi)(E_m - E_g)^{3/2}$ versus $m$ should exhibit as a straight line with a slope of $(h\theta)^{3/2}$. By finding this slope, we can calculate the inside electric field base on Eq. 3.18. Based on the FKO analysis, it showed that the electric field inside the RBS MQW sample is around 5.28 kV/cm. The plotting of $(4/3\pi)(E_m - E_g)^{3/2}$ versus $m$ and linear fitting can be seen in Fig. 3.17.

![Graph](image)

**Figure 3.17:** Analysis of the Franz-Keldysh oscillation (FKO) above the GaAs substrate in ER spectrum from the RBS MQW sample.
3.7.3 Angle dependent ER measurement of MQW Bragg structure

Resonance condition in the MQWs structure is given by the diffraction law, Eq. 1.10. By tuning the light incident angle $\theta$, one can modify the energy of the major Bragg reflection. Fig. 3.18 shows a series of ER spectra from the RBS MQWs sample with 60 QWs at different incident angles, ranging from 10.5° to 46°. Spectra are stacked by a distance from each other and the Y axis label is for the top most spectrum. All measurements were performed at 20 K. The features related to exciton transitions are marked by arrows, noted $x(e_1-hh_1)$ as the electron and heavy hole ground state, $x(e_1-lh_1)$ as the electron and light hole ground state, and $x(e_2-hh_2)$ as the electron and heavy hole second state. For the $x(e_2-hh_2)$ exciton transition, we can clearly see that the feature gets its maximum around an incident angle of 23°, when the double resonance condition is met, and decreases while the incident angle is detuned from the double resonance condition. Under the double resonance condition, the photon energy corresponds to both the $x(e_2-hh_2)$ exciton transition and the Bragg reflection condition, resulting a significant enhanced ER spectrum.
Figure 3.18: ER spectra from the RBS MQW sample with 60 QWs at temperature 20 K and varies incident angles. The exciton transition features are marked by arrows. (The Y axis label is account for the top most spectrum and other spectra are stacked in order.)
3.7.4 Temperature dependent ER measurement of MQW Bragg structure

According to Eq. 3.14, exciton transition energy is affected by the sample temperature. By varying the measurement temperature, it is possible to tune the double resonance condition. Figure 3.19 shows a series of ER spectra from the RBS MQW sample, with respect to different temperature ranging from 20 to 250 K. All measurements were taken with an incident angle of $23^\circ$ and plotted with constant stack in the same Y axis in Fig. 3.19. From Fig. 3.19, one can see that exciton transition features, including $x(e_1-hh_1)$, $x(e_1-lh_1)$, and $x(e_2-hh_2)$ states, all have a redshift and decrease in the magnitude with increasing temperature. At high temperatures, (e.x. above 200 K), exciton transitions are below the noise level and cannot be recognized in the spectrum anymore. Comparing the behavior of $x(e_1-hh_1)$, $x(e_1-lh_1)$ to the $x(e_2-hh_2)$ transition, one can see the feature of $x(e_2-hh_2)$ excitonic transition decrease must fast with temperature increase than the $x(e_1-hh_1)$ and $x(e_1-lh_1)$ transitions. A series of FKOs can be recognized at the left part of each spectrum above the GaAs band gap. These FKOs also exhibits a redshift with temperature increase, but the magnitude of the oscillations remain the same with temperature.
Figure 3.19: ER spectra from the RBS MQW sample with 60 QWs at incident angle of 23° and varies temperatures, ranging from 20 to 250 K. (The Y axis label is account for the bottom most spectrum and other spectra are stacked in order.)

3.7.5 Field Dependent ER measurement of MQW Bragg structure

One interesting feature of the resonant Bragg structure (RBS) is that it is possible to manipulate the optical properties by tuning the excitonic transition energy in the RBS. One of the ways is by applying an external electric field that affects the excitonic state of the QWs. The external
electric field changes both the quantum confinement energies of electrons and holes and the exciton binding energy. As a result, it affects the resonant optical respond of the RBS, which can be utilized in a variety of optoelectronic and photonic devices.

An ER measurement of the electric field dependence had been done by applying a constant DC voltage on the sample which has a thickness of 0.5 mm. The DC voltage was generated by the function generator along with the modulation square wave signal, and then was amplified by the amplifier by 1000 times. We applied the DC shift ranging from -750 to 1750 V. Beyond this range, a short-circuit occurred due to our setup. The result ER spectra with different DC shift are shown in Fig. 3.20. The ER spectra exhibits the same main transition features. The left most feature is due to bulk GaAs transition which appears around 1.518 eV. Above the GaAs band gap, a series of Frantz-Keldysh oscillation can be identified. Around 1.54 eV the major transition peak is due to the ground state heavy hole exciton \( x(e_1-hh_1) \) followed by the ground state light hole exciton \( x(e_1-lh_1) \) around 1.55 eV. The major transition feature on the right part of the spectra is the transition feature due to the heavy hole exciton at the second state \( x(e_2-hh_2) \) which occurs around 1.628 eV. A broadening effect of the \( x(e_2-hh_2) \) transition feature can be clear seen when the voltage increases from 0 to 1750 V, or from 0 to -750 V.

To further analyze the excitonic transition energy shifts due to external electric field, we used the first derivative of Gaussian lineshape (FDGL) to fit the ER spectra. The fitting equation is Eq. 3.16 and the critical point we used for a FDGL fitting is \( n = 5 \). The fitting result is shown in Fig. 3.21 and the fitting parameters are listed in Table 3.8.
Figure 3.20: ER spectra from the RBS MQW sample with 60 QWs at varies DC voltage, ranging from -750 V to 1750 V. The measurement was done at an incident angle of 23° and temperature 20 K. (The Y axis label is account for the top most spectrum and other spectra are stacked in order.)
Figure 3.21: First derivative Gaussian lineshape fitting (FDGL) of ER spectra from the RBS MQW sample with 60 QWs at varies DC voltage, ranging from -750 V to 1750 V. The experimental data is plotted as black solid lines and the fitting result is plotted as red dashed lines.
Table 3.8: Fitting parameters of first derivative Gaussian lineshape fitting (FDGL) for ER spectra from the RBS MQW sample with 60 QWs at varies DC voltage. The critical point \( n = 5 \).

<table>
<thead>
<tr>
<th>DC Voltage (V)</th>
<th>Transition feature</th>
<th>Fitting values</th>
</tr>
</thead>
<tbody>
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Figure 3.22: First derivative Gaussian lineshape fitting (FDGL) energies for the transition features from the ER spectra with electric field dependence.
Figure 3.23: First derivative Gaussian lineshape fitting (FDGL) linewidth for the transition features from the ER spectra with electric field dependence. The navy color triangle is the energy difference between the two fitting energies for the x(e2-hh2) transition feature.
The fitting energies by using first derivative of Gaussian lineshape (FDGL) for the ER spectra are plotted in Fig. 3.22. The black square points are the fitting energies for bulk GaAs transition feature. The red circle and blue triangle points are the fitting energies for the exciton transition at the ground state, corresponding to the heavy hole exciton $x(e_1-hh1)$ and the light hole exciton $x(e_1-lh1)$. The pink triangle and green square points are the two fitting energies for the heavy hole exciton transition at the second state $x(e_2-hh2)$.

According to quantum mechanics, assuming there is no build in electric field, the exciton transition energy for the ground state heavy hole and light hole excitons will shift to lower energy or long wavelength when a electric field is applied. This can be seen from Fig. 1.6. And the exciton transition energy for the second state heavy hole exciton $x(e_2-hh2)$ will shift to higher energy with increasing external electric field. From our experiment, as plotted in Fig. 3.22, the exciton transition energy for ground state heavy hole and light hole excitons almost remain at the same level and it’s hard to identify any shift. The first fitting energy of the heavy hole exciton at the second state is slightly shifted to lower energy and the second fitting energy of the heavy hole exciton at the second state almost remain constant.

This inconsistency of our experimental result to the theoretical expectation could be due to the following factors: (1) We used a mesh on top of the MQW sample to apply DC voltage and it is very hard to eliminate any gap between the mesh and the surface of the sample. Due to this vacuum gap, a large portion of the DC voltage won’t be applied on the sample thus create less electric field in the sample. (2) The mesh only contacts the surface of sample exactly under the metal lines of the mesh with a limited area. This may results in a nonuniform electric field in the sample and the surface area we were probing for reflection mainly locates between mesh grids and may have less
electric field than we expected. (3) The exciton transition energy shift due to external electric field is very small. On the other hand, the ER spectrum lineshape is dominated by the phase parameter which cannot be controlled during ER measurement. Our fitting procedure is based on the ER spectrum lineshape and the inconsistency of the ER lineshape makes it very hard to identify the small transition energy shift in the fitting procedure.

The fitting linewidth by using first derivative of Gaussian lineshape (FDGL) for the ER spectra are plotted in Fig. 3.23 along with a plot of the energy difference of the two fitting energies for the x(e2-hh2) transition feature. The broadening effect of the heavy hole exciton in the second state with increasing external electric field can be clearly seen in the graph.

### 3.7.6 Discussion

We applied ER spectroscopy both on SQW and MQW samples. For the SQW, a strong feature due to the bulk excitons in GaAs substrate and Franz-Keldysh oscillations at higher energy was observed. A weak feature related to the x(e1-hh1) exciton can be just recognized, while the features related to the x(e1-lh1) and x(e2-hh2) excitons are below the noise level. In the ER spectra of the RBS MQW sample, strong features related to the x(e1-hh1), x(e1-lh1) and x(e2-hh2) excitons were observed. The transition features are obviously different with respect to their width and shape, and overlap with the Franz–Keldysh oscillation coming from the GaAs substrate. For instance, the ER feature related to the x(e1-lh1) exciton state has a Fano line shape whereas the ER feature related to the x(e1-hh1) and x(e2-hh2) exciton states are close to a Lorentz profile. In the OR spectra, all peaks show similar bell-like shapes. The qualitative difference in the exciton-related ER lineshapes
seems to be a result of the opposite sign and different magnitude of the Stark effect for the x(e1-lh1) and x(e2-hh2) excitons in the QWs.

The full widths at half maximum (FWHM) of the excitonic features is about 4 meV for the x(e1-hh1), 3 meV for the x(e1-lh1), and 9 meV for the x(e2-hh2) excitons. Stronger broadening results in lower OR and ER intensities for the x(e2-hh2) line compared to the x(e1-hh1) line in the reference sample with the single QW. The OR and ER spectra of the RBS with 60 QWs do not follow this rule. Instead, a strong enhancement in intensity is observed for the x(e2-hh2) feature when in resonance. A significant broadening of the linewidth of the x(e2-hh2) feature is also observed when the double resonance conditions are met. This signifies the formation of a superradiant optical mode. The origin for this enhancement of the x(e2-hh2) line in the RBS with 60 QWs is the electromagnetic coupling between the x(e2-hh2) excitons in the QWs placed at the period satisfying the Bragg resonance. For the temperature of 4 – 20 K, this double resonance condition corresponds to the incident angle around $\theta = 23^\circ$. Detuning from the double resonance condition leads to a noticeable decrease in the OR and ER signals.

In the ER measurement of electric field dependence, no significant shift of the exciton transitions was observed. This could be due to the external DC electric field we applied was too small compared to the build in electric field. However, the broadening effect for the heavy-hole exciton at the second state x(e2-hh2) under various external DC electric field can be clearly seen. This proves that the transition feature of the heavy-hole exciton at the second state x(e2-hh2) is very sensitive to the external electric field.
Chapter 4

Conclusion

In this thesis, the resonant optical properties of a periodic system of the quantum well excitons were studied. In order to evaluate the heavy-hole exciton at the second state, the GaAs/AlGaAs resonant Bragg structure with multiple quantum wells was designed and fabricated to obtain the optical Bragg resonance near the exciton-polariton resonance for the heavy-hole exciton at the second state $x(e2\text{-}hh2)$ in the quantum wells. A single quantum well sample with identical supercell structure as the MQW sample was used for reference purpose.

We applied optical reflectance (OR), photoreflectance (PR) and electroreflectance (ER) spectroscopy techniques on both the SQW and MQW samples for evaluating the exciton transitions under various conditions, such as incident angle, temperature and electric field. The double resonance condition was achieved by tuning the incident angle of the light and by variation of the temperature. Under the double resonance condition, a significant enhancement of the light-matter interaction was observed, which manifests itself by strong resonant optical reflection, photoreflec-
tion and electroreflection at the exciton energies.

A simulation of Bragg reflection of MQWs with matched dielectric constants has been done to illustrate the strengthening and broadening effect of the exciton transition with the number of quantum wells. This agrees with our experiment result very well. In the OR and ER measurement of the SQW sample, only small features related to the ground state excitons were observed, while the heavy-hole exciton at the second state x(e2-hh2) was below noise level and not detected. When the RBS MQW sample was measured by OR, PR and ER spectroscopies, significant enhancement of the exciton transition features were observed, especially for the exciton x(e2-hh2) under the double resonance condition.

The double resonance condition was achieved around incident angle of 23° at temperature 20 K and normal incidence around temperature 150 K. In the OR measurement of the RBS MQW sample, the double resonance condition gave an overall enhancement of about 44% to the Bragg reflection and made it possible to identity the transition of the heavy-hole exciton at the second state x(e2-hh2). The exciton energies we got from the experiment agree with our calculation result very well. By fitting the OR spectra, we obtained parameter of radiative broadening of the x(e2-hh2) excitons $\Gamma_0$ equal to $40 \pm 5$ μeV and non-radiative broadening $\Gamma$ to be 3 meV. In the ER measurement, rich and strong features related to exciton transitions x(e1-hh1), x(e1-lh1) and x(e2-hh2) were observed. The fitting energies for excitons agree with our calculated value very well. In the field dependent ER measurement, although no significant exciton energy shift was observed, the broadening effect for the heavy-hole exciton at the second state x(e2-hh2) under various DC electric field was clearly seen, demonstrating the sensitivity of x(e2-hh2) transition with respect to an external field.
In conclusion, we demonstrated the RBS composed of a periodic system of the QW exciton-polaritons at the second quantum state. The optical lattice of the x(e2-hh2) excitons shows a strong amplification of the resonant OR and ER when the exciton-polariton resonance coincides with the Bragg resonance. The broadening of the x(e2-hh2) line when in resonance indicates the formation of the super-radiant optical mode. We experimentally prove that the second quantum state of the QW excitons is effective for the electromagnetic coupling as much as the ground state.

4.1 Future work

To further investigate the exciton transition energy shift with external electric field, we are planning to create a transparent conductive layer on top of the sample surface by spin coating. This will eliminate the vacuum gap between the electrode and the sample and generate a uniform electric field in the sample. Under this circumstances, we believe we will be able to observe the exciton transition shift due to external electric field. The result will be reported in our recent paper.
Appendix A

A.1 Matlab code for simulation of Bragg reflection

```matlab
A = 13.5973256465689504916944143041E-9;
b = 97E-9;
d = a+b;
bp = 103.644E-9;
w0 = 1.629; %eV
T0 = 50E-6; %eV
c = 3E8;
nb = 3.45;
hbar = 6.63E-34/(2*pi);
e = 1.6E-19;
r01 = (1-nb)/(1+nb);
NList = [1, 2, 10, 20, 50, 100, 200, 300, 400, 500];
for n = 1:length(NList)
    legend_string(n) = {'N=',num2str(NList(n))};
end

%% Figure 1, T = 0
T = 0;
figure(1), clf, hold all;
for n = 1:length(NList)
    N = NList(n);
    X = -0.015:0.00001:0.015;
    RNarr = [];
    for x = -0.015:0.00001:0.015
```

112
\[ w = xw0+w0; \]
\[ r1 = iT0/(w0-w-i(T0+T)); \]
\[ kb = nb*(w*e/hbar)/c; \]
\[ r1t = exp(i*kb*d)*r1; \]
\[ t1t = exp(i*kb*d)*(1+r1); \]
\[ K_d = \frac{\text{acos}((t1t^2-r1t^2+1)/2/t1t)}{\sin((N-1)*K_d)/\sin(N*K_d)}; \]
\[ rNt = \frac{r1t}{1-t1t*\sin((N-1)*K_d)/\sin(N*K_d)}; \]
\[ \phi = kb*(bp-b/2); \]
\[ rN = \frac{(r01+rNt*exp(2i*phi))/(1+r01*rNt*exp(2i*phi))}{(abs(rN))^2}; \]
\[ if \text{isnan}(rN) \]  % exclude singularity
\[ RN = RNarr(end); \]
\[ RNarr(end+1) = RN; \]
\[ end \]
\[ plot(X,RNarr); \]
\[ \text{Cal FWHM} \]
\[ \text{FWHM1}(n) = \text{fwhm}(X,RNarr); \]
\[ end \]
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\[ set(gca,'FontSize',12); \]
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\[ ylabel('$R_N$','Interpreter','LaTeX','FontSize',15); \]
\[ box on; \]
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\[ Figure 2, T = 100E-6 \]
\[ T = 100E-6; \]
\[ figure(2), clf, hold all; \]
\[ for n = 1:length(NList) \]
\[ N = NList(n); \]
\[ X = -0.015:0.00001:0.015; \]
\[ RNarr = []; \]
\[ for x=-0.015:0.00001:0.015 \]
\[ w = x*w0+w0; \]
\[ r1 = iT0/(w0-w-i(T0+T)); \]
\[ kb = nb*(w*e/hbar)/c; \]
\[ r1t = exp(i*kb*d)*r1; \]
\[ t1t = exp(i*kb*d)*(1+r1); \]
\[ 113 \]
\[ K_d = \cos((t_1t^2-r_1t^2+1)/2/t_1t); \]
\[ r_{Nt} = r_1t/(1-t_1t\sin((N-1)*K_d)/\sin(N*K_d)); \]
\[ \phi = kb*(bp-b/2); \]
\[ r_N = (r_{01}+r_{Nt}\exp(2i*\phi))/(1+r_{01}r_{Nt}\exp(2i*\phi)); \]
\[ RN = (\text{abs}(r_N))^{-2}; \]
\[ \text{if isnan}(RN) \ % \text{exclude singularity} \]
\[ RN = R_Narr(\text{end}); \]
\[ \text{end} \]
\[ R_Narr(\text{end+1}) = RN; \]
\[ \text{end} \]
\[ \text{plot}(X, R_Narr); \]

**% Format**

`set(gca,'FontSize',12);`

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`ylabel('$R_N$','Interpreter','LaTeX','FontSize',15);`

`legend(legend_string); box on;`

`text(-0.013,0.8,'(b)');`

`text(-0.0115,0.794,'\hbar\Gamma=100,\mu eV$','FontSize', 12, ...

'Interpreter','LaTeX');`

`text(-0.007,0.35,'\omega_-$','FontSize', 12, 'Interpreter','LaTeX');`

`text(0.006,0.35,'\omega_+$','FontSize', 12, 'Interpreter','LaTeX');`

`printFig(gcf,'NQWRefSim2.pdf');`

\% Figure 3, \[ T = 1000E-6 \]

\[ T = 1000E-6; \]

`figure(3), clf, hold all;`

`for n = 1:length(NList)`

\[ N = NList(n); \]

\[ X = -0.015:0.00001:0.015; \]

\[ R_Narr = []; \]

`for x=-0.015:0.00001:0.015`

\[ w = x*wo+wo; \]

\[ r_1 = i*T0/(wo-w-i*(T0+T)); \]

\[ kb = nb*(w*e/hbar)/c; \]

\[ r_{1t} = \exp(i*kb*d)*r_1; \]

\[ t_{1t} = \exp(i*kb*d)*(1+r_1); \]

\[ K_d = \cos((t_{1t}t^2-r_{1t}t^2+1)/2/t_{1t}); \]

\[ r_{Nt} = r_1t/(1-t_{1t}t\sin((N-1)*K_d)/\sin(N*K_d)); \]

\[ \phi = kb*(bp-b/2); \]

\[ r_N = (r_{01}+r_{Nt}\exp(2i*\phi))/(1+r_{01}r_{Nt}\exp(2i*\phi)); \]

\[ RN = (\text{abs}(r_N))^{-2}; \]

`\text{if isnan}(RN) \ % \text{exclude singularity} \]

\[ RN = R_Narr(\text{end}); \]

\[ \text{end} \]

\[ R_Narr(\text{end+1}) = RN; \]

`\text{end} \]

`\text{plot}(X, R_Narr); \]
A.2  Matlab code for exciton energy calculation in quantum wells

clear, clc;

%%% Constants
m0 = 9.10938291E-31;  % Free electron rest mass
h = 6.62606957E-34;   % Plank constant
hbar = h/(2*pi);      % Reduced Plank constant
ee = 1.60217657E-19;  % Electron charge

%%% Parameters GaAs/AlGaAs
L = 12.5E-9;          % GaAs QW width
Eg_well = 1.518;      % Bandgap in eV
Eg_barrier = 2.5753;  % Electron
dEc = 0.4729;         % Conduction band discontinuity
exciton_binding = 8.5E-3;  % Exciton binding energy (eV)
V0_ee = (Eg_barrier-Eg_well)*dEc*ee;  % Barrier height
me_ee = 0.067;        % Effective electron mass
V0_lh = (Eg_barrier-Eg_well)*(1-dEc)*ee;  % Barrier height
me_lh = 0.087;        % Effective hole mass
% Heavy hole
V0_hh = (Eg_barrier-Eg_well)*(1-dEc)*ee; % Barrier height
me_hh = 0.62; % Effective hole mass

u0_2_ee = me_ee*m0*L^2*V0_ee/(2*hbar^2); % u0 square
u0_2_lh = me_lh*m0*L^2*V0_lh/(2*hbar^2);
u0_2_hh = me_hh*m0*L^2*V0_hh/(2*hbar^2);

figure(1), clf, hold all; box on;
v = 0:0.0001:8;
plot(v, sqrt(u0_2_ee-v.^2));
plot(v,v.*tan(v));
plot(v,-v.*cot(v));
ylim([0 8]); title('Electron')
figure(2), clf, hold all; box on;
v = 0:0.0001:8;
plot(v, sqrt(u0_2_lh-v.^2));
plot(v,v.*tan(v));
plot(v,-v.*cot(v));
ylim([0 8]); title('Light hole')
figure(3), clf, hold all; box on;
v = 0:0.0001:20;
plot(v, sqrt(u0_2_hh-v.^2));
plot(v,v.*tan(v));
plot(v,-v.*cot(v));
ylim([0 20]); title('Heavy hole');

%% Parameters
pars = {
'Binding energy = ',num2str(exciton_binding),' eV';
'Well width L = ',num2str(L*1E9),' nm';
'Eg well = ',num2str(Eg_well), ' eV';
'Eg barrier = ',num2str(Eg_barrier), ' eV';
'Conduction discontinuity % = ',num2str(dEc);''
'Effective mass me = ',num2str(me_ee), ' m0';
'Effective mass mlh = ',num2str(me_lh), ' m0';
'Effective mass mhh = ',num2str(me_hh), ' m0';
};
A.3 Part of Labview design for scanning program

Figure A.1: Labview diagram for the main scanning program.
Figure A.2: Monochromator motor driver Labview program interface.

Figure A.3: Labview diagram for monochromator motor driver program.
Figure A.4: Lock amplifier Labview control program interface.

Figure A.5: Data saving Labview program interface.
Figure A.6: Labview diagram for data saving program.
Bibliography


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