Abstract

Novel Fabrication Techniques and Transport Characterization of
Semiconductor Nanostructures

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With the advent of epitaxial growth techniques such as molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD), and the development of nanoscale lithography techniques (i.e., electron-beam lithography), the design and implementation of electronic systems that exhibit a range of both classical charging effects and/or quantum mechanical effects is now possible. The focus of my dissertation concerns several studies which examine both the physics at these size scales in different material systems and structures, as well as novel fabrication techniques for creating these structures. After discussing the background theory and the major experimental methods that I have developed, transport results from two studies are presented. The first is a study of the density of states in superlattice structures in the GaAs/AlGaAs material system. The second is a study of transport in vertical quantum dot structures, also using GaAs and AlGaAs to form the wires and barriers. The ‘dot’ in this case is an InGaAs well, which allows for the study of electron transport in a more linear regime. The challenges involved in the fabrication of vertical quantum dots are the motivation for the study which follows; a technique for obtaining self-assembled quantum dot structures. Following this, the results of a study that explores fabrication attempts to realize two kinds of novel charge quantized device structures in metals will be shown. Lastly, the results of an attempt to realize a heterostructure system in Si/CaF$_2$ (similar to the GaAs/AlGaAs material system) will be presented.
Novel Fabrication Techniques and Transport Characterization of Semiconductor Nanostructures

A Dissertation
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by
Jeffrey W. Sleight
Advisor: Prof. Mark A. Reed
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Acknowledgments

Dedicated to my parents, who first interested me in science, my freshman physics teacher, who taught me the beauty of physics, and my future wife, who inspired me to keep at it.

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<tr>
<td>A</td>
<td>Area</td>
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<tr>
<td>Å</td>
<td>Ångström</td>
</tr>
<tr>
<td>$a_B$</td>
<td>Bohr radius</td>
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<tr>
<td>AFM</td>
<td>Atomic force microscopy</td>
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<tr>
<td>AlGaAs</td>
<td>Aluminum Gallium Arsenide</td>
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<tr>
<td>Au</td>
<td>Gold</td>
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<tr>
<td>B</td>
<td>Magnetic field</td>
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<tr>
<td>C</td>
<td>Capacitance</td>
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<tr>
<td>CaF$_2$</td>
<td>Calcium fluoride</td>
</tr>
<tr>
<td>$C_G$</td>
<td>Gate capacitance</td>
</tr>
<tr>
<td>DBRTS</td>
<td>Double-barrier resonant-tunneling structure</td>
</tr>
<tr>
<td>$\Delta E$</td>
<td>Change in energy</td>
</tr>
<tr>
<td>$\delta E$</td>
<td>Energy spacing between single-particle states</td>
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<tr>
<td>$\epsilon$</td>
<td>Dielectric constant</td>
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<tr>
<td>$e$</td>
<td>Electron charge</td>
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<tr>
<td>$E_C$</td>
<td>Charging energy</td>
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<tr>
<td>$E_F$</td>
<td>Fermi energy</td>
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<tr>
<td>$f(\Delta E)$</td>
<td>Fermi function</td>
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<tr>
<td>$\Gamma$</td>
<td>Tunneling rate</td>
</tr>
<tr>
<td>Symbol</td>
<td>Description</td>
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<tr>
<td>--------</td>
<td>-------------</td>
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<tr>
<td>$G$</td>
<td>Conductance</td>
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<tr>
<td>GaAs</td>
<td>Gallium Arsenide</td>
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<tr>
<td>Ge</td>
<td>Germanium</td>
</tr>
<tr>
<td>$h$</td>
<td>Planck's constant</td>
</tr>
<tr>
<td>$\hbar$</td>
<td>Planck's constant divided by $2\pi$</td>
</tr>
<tr>
<td>I</td>
<td>Current</td>
</tr>
<tr>
<td>InAs</td>
<td>Indium Arsenide</td>
</tr>
<tr>
<td>InGaAs</td>
<td>Indium Gallium Arsenide</td>
</tr>
<tr>
<td>$J$</td>
<td>Current density</td>
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<tr>
<td>$k$</td>
<td>Wavevector</td>
</tr>
<tr>
<td>$k_B$</td>
<td>Boltzmann's constant</td>
</tr>
<tr>
<td>LO</td>
<td>longitudinal optical</td>
</tr>
<tr>
<td>$m^*$</td>
<td>Effective mass</td>
</tr>
<tr>
<td>MBE</td>
<td>Molecular Beam Epitaxy</td>
</tr>
<tr>
<td>MOCVD</td>
<td>Metalorganic Chemical Vapor Deposition</td>
</tr>
<tr>
<td>MQT</td>
<td>Macroscopic quantum tunneling</td>
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<tr>
<td>nm</td>
<td>nanometer</td>
</tr>
<tr>
<td>NDR</td>
<td>Negative differential resistance</td>
</tr>
<tr>
<td>$\Phi$</td>
<td>Potential energy</td>
</tr>
<tr>
<td>PMMA</td>
<td>Polymethylmethacrylate</td>
</tr>
<tr>
<td>P/V</td>
<td>Peak-to-valley</td>
</tr>
<tr>
<td>Q</td>
<td>Charge on junction</td>
</tr>
<tr>
<td>QPC</td>
<td>Quantum point contact</td>
</tr>
<tr>
<td>$R_T$</td>
<td>Barrier resistance</td>
</tr>
<tr>
<td>$R_K$</td>
<td>Quantum unit of resistance</td>
</tr>
<tr>
<td>RIE</td>
<td>Reactive ion etching</td>
</tr>
<tr>
<td>Abbreviation</td>
<td>Full Form</td>
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<td>--------------</td>
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<tr>
<td>RTD</td>
<td>Resonant tunneling diode</td>
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<tr>
<td>SEM</td>
<td>Scanning electron microscopy</td>
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<tr>
<td>SET</td>
<td>Single electron transistor</td>
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<tr>
<td>Si</td>
<td>Silicon</td>
</tr>
<tr>
<td>SiO₂</td>
<td>Silicon dioxide</td>
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<tr>
<td>T(E)</td>
<td>Transmission probability</td>
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<tr>
<td>TE</td>
<td>Thermoelectric</td>
</tr>
<tr>
<td>TEM</td>
<td>Transmission electron microscopy</td>
</tr>
<tr>
<td>Ti</td>
<td>Titanium</td>
</tr>
<tr>
<td>τₑ</td>
<td>Lifetime of quasi-bound quantum state</td>
</tr>
<tr>
<td>Uₖ</td>
<td>Single electron charging energy</td>
</tr>
<tr>
<td>V</td>
<td>Voltage</td>
</tr>
<tr>
<td>W</td>
<td>Depletion depth</td>
</tr>
<tr>
<td>WD</td>
<td>Working distance</td>
</tr>
<tr>
<td>Ω</td>
<td>Ohms</td>
</tr>
<tr>
<td>1D</td>
<td>One dimensional</td>
</tr>
<tr>
<td>2D</td>
<td>Two dimensional</td>
</tr>
<tr>
<td>2DEG</td>
<td>Two dimensional electron gas</td>
</tr>
<tr>
<td>3D</td>
<td>Three dimensional</td>
</tr>
<tr>
<td>(111)</td>
<td>A specific crystal plane designated by its Miller indices</td>
</tr>
<tr>
<td>&lt;111&gt;</td>
<td>A family of directions</td>
</tr>
</tbody>
</table>
Chapter 1

Introduction

With the advent of epitaxial growth techniques such as molecular beam epitaxy (MBE) and metal-organic chemical vapor deposition (MOCVD), and the development of nanoscale lithography techniques (i.e., electron beam lithography), the design and implementation of nanometer electron devices that exhibit a range of both classical charging effects and/or quantum mechanical effects is now possible. The motivation for creating these structures has historically had two major thrusts: 1) The creation of nano-scale laboratories to examine physics, 2) The development of an electronics technology that will both be far smaller and faster than conventional silicon technology. The first goal has been and continues to be an incredibly active field. Discoveries such as the Integer [59] and Fractional Quantum Hall Effects [105] were cases where new, unexpected, physics was explored. The recent observation of Bloch Oscillations in superlattice structures [108] is an example of where expected physics has finally been seen in experiment. While much of the work involving resonant tunneling structures, quantum dots, and coulomb blockade structures is not new physics, it does provide a new testing ground for verification of basic concepts concerning how electron transport is affected at scales where spatial and charge quantization dominate.

The second motivation (next generation electronics), remains far more elusive. Indeed, it currently seems that beyond certain niche applications, electronic device technologies based
upon either spatial or charge quantization are not likely to become the next generation electronics, at least in the materials which they are currently fabricated. For devices that rely on charge quantization, the prospects are daunting. Early predictions by Likharev[63] indicating that 10 nm junction diameters are sufficient for room temperature operation turned out to be far too optimistic. More recent estimates for these structures show that for device structures fabricated at the current lithographic limits ($\sim 10$ nm), operation at less than 100K is required. For room temperature operation of such devices, they would have to be scaled to areas encompassing only a few atoms. To date, there is no high throughput technology capable of achieving this. The only hope for room temperature operation in the field of single electronics comes from self-assembled structures. Instead of building a structure in a very fabrication intensive manner, one can make the structure in a parallel manner a million times over (literally by mixing the right things together in the beaker). While there have been many demonstrations of processes involving self-assembly [50], very little work has examined the electron transport in these systems. There most likely will be a large amount of basic materials science that will have to occur before the prospects of it as a candidate for the next generation of electronic devices can be ascertained.

In the general field of quantum effect devices, the situation is less clear. There are certainly applications which currently use such devices at a discrete level. The resonant tunneling diode (RTD) has been employed in high frequency oscilloscopes, where the need for a very fast trigger makes it a natural choice. There are also major efforts underway to exploit the advantages that could arise from multi-valued logic (i.e., a transistor that operates in the quantum regime can go from off to on an arbitrary number of times). By using multi-valued logic, fewer active components are required. For this to be economically worthwhile, it will have to be less expensive than what is done in silicon. Although room temperature operation has been obtained for quantum effect devices, the major obstacle in utilizing them in a large scale integrated design is related to fluctuations of the well and barrier dimensions when they are made. In order to achieve quantum spatial confinement,
structures with small wells and barriers must be created (typically on the order of 10Å to 50Å). As one can easily imagine, monolayer variations in the wells and barriers often occur. These variations have a large impact on both the resultant quantum energy levels and on the current density through the structure, thus the integration of these devices is a formidable task.

The operational voltage and temperature range of any new electron device technology is an very important factor in determining its feasibility. Figure 1.1 summarizes the temperature and voltage operation ranges for various nanostructures. The horizontal dashed white line marks 300K (room temperature operation), and the vertical dashed white line ($\Delta E_Q/e$) shows the voltage operation limits imposed by the separation of quantized states (either through confinement or charge). The majority of these structures (waveguides, quantum interference, quantum point contacts (QPC), metal SET's, and depletion-defined lateral structures) are restricted to operation at cryogenic temperatures (less than 100K; less than 1K in some cases). Only epitaxial heterojunction tunneling devices have successfully achieved room temperature operation.

The focus of my dissertation concerns examining the physics at these size scales, rather than on building the next generation of electronics from these materials, although some of what will be presented definitely examines this problem as well. Whatever electronic technology prevails, it will undoubtedly have to deal with issues that become important as size scales shrink (quantization, single electron charging, tunneling). Before launching into the experimental work, in Chapter 2 I will outline the basics of the relevant theory and background needed to appreciate the experimental work. Chapter 3 will show the more important experimental techniques and procedures. The results consist of studies on six different types of materials and structures, where the emphasis has always been on either electron transport measurements, or on the fabrication of a novel system to examine electron transport. Chapter 4 describes work involving superlattice structures in the GaAs/AlGaAs material system. By repeating a barrier/well structure over many periods, one can create
Figure 1.1: Comparison of quantum effects in nanostructures. Adapted from reference [82].
a lattice (superlattice), and as in a crystalline solid, a band structure will be formed. The 
sensitivity of the electron transport properties to fluctuations in the epitaxial structure is 
*much less* than that present in a simple double barrier/single well system (i.e., the RTD).

Chapter 5 is concerned with vertical quantum dot structures. Previous work examining 
these structures was largely in the GaAs/AlGaAs system, where due to depletion effects, 
when electron spectroscopy reveals the presence of quantization, the injected electrons are 
quite hot (i.e., a bias as large as 100-200 mV must be applied). By utilizing an InGaAs well, 
injection can occur at far lower bias levels, placing the transport much closer to the linear 
regime, which is accessible to the theorists more readily. Chapter 6 describes a technique 
for obtaining self-assembled quantum dot structures, rather than fabricating them. Chapter 
7 explores fabrication attempts at realizing two kinds of novel charge quantized device 
structures. In Chapter 8, the electronic system is simple (a single, thin barrier), but the 
material system used, Si/CaF$_2$/Si, is new. Chapter 9 will include concluding remarks.
Chapter 2

Theoretical and Experimental Background for Charging and Energy Quantization

2.1 Introduction

As dimensions of an electronic structure become small (<100Å), there are two major effects that result. The first, Coulomb blockade, has classical origins, whereas the second, energy quantization due to spatial confinement, is strictly quantum mechanical in nature. In metal systems, the short Fermi wavelength (~0.5 nm) has prevented the observation of effects due to quantum confinement. Thus all of the work in this area has focused almost exclusively on Coulomb blockade effects. In semiconductors, the Fermi wavelength is typically on the order of 5 nm, making both quantization and charging effects experimentally accessible. In this chapter, the basic theory behind both Coulomb blockade and energy quantization in nanostructures will be given. Also, some results from the landmark experimental work will be discussed.
2.2 Coulomb Blockade

Observation of Coulomb charging effects due to single electrons is not a new field. It was first seen several decades ago[40, 75, 61, 62, 44]. However, at that time, it was only seen in granular metallic films. With the advent of nanofabrication techniques, metallic junctions with capacitances in the femto or even atto farad regime are possible.[38] Although much of the literature in this field has been concerned with metal/insulator/metal junctions, the same arguments also apply for heterostructure semiconductor systems.[94] The only complication arising in the semiconductor system is that energy quantization may occur as well.

The motivation for achieving systems with such small capacitances is to increase the charging energy, $E_C$. For a simple parallel plate capacitor, the capacitance is given by

$$C = \frac{\epsilon A}{d}, \quad (2.1)$$

where, $\epsilon$ is the dielectric constant of the insulator, $A$ is the area of the junction, and $d$ is the thickness of the insulator. The charging energy of such a junction is

$$E_C = \frac{Q^2}{2C}, \quad (2.2)$$

where $Q$ is the charge on the junction. It is clear that in order to obtain a large charging energy, we need to make $C$ as small as possible, by making $A$ as small as possible. As with any process that is observed at a finite temperature, it is required that

$$E_C \gg k_B T. \quad (2.3)$$

An additional requirement is that the barrier used must have a resistance ($R_T$) that exceeds the quantum unit of resistance ($R_K$),

$$R_T \gg R_K = \frac{h}{e^2} \approx 25.8k\Omega. \quad (2.4)$$

This requirement is due to the need to localize the electron long enough to charge the junction. Equation 2.4 follows from 2.2 and the Heisenberg uncertainty principle, which
gives the lifetime energy broadening by $\hbar/\tau$, where $\tau$ is the time the electron spends in the junction.

### 2.2.1 Single Barrier Junction

The simplest case to consider is that of a current biased single barrier junction, shown schematically in figure 2.1. In this case, the change in energy, $\Delta E$, for one electron either entering or leaving the junction is

$$\Delta E = \left(\frac{1}{2C}\right) (Q^2 - (Q \pm e)^2) = \left(\frac{1}{C}\right) (q \pm \frac{e}{2}).$$

(2.5)

The tunneling rate, $\Gamma$, is proportional to $\Delta E^2$, and for a finite temperature can be written as

$$\Gamma = \left(\frac{1}{e}\right) I(V) f(\Delta E) = \left(\frac{1}{e^2}\right) R_T f(\Delta E),$$

(2.6)
where \( f(\Delta E) \) is the Fermi function, and \( I(V) \) is the current-voltage characteristic of the junction. At zero temperature this becomes

\[
\Gamma = \left( \frac{1}{e^2} \right) R_T \Delta E. \tag{2.7}
\]

Thus, at low temperatures (where the effect of thermally assisted processes can be neglected), tunneling is only possible for positive \( \Delta E \), requiring a charge of \( \pm e/2 \) to be supplied to the junction. This results in a theoretical current-voltage \( I(V) \), as shown in figure 2.2. In the voltage range \(-e/2C < V < (e/2C)\), no tunneling current is possible. This is known as Coulomb blockade, as a Coulomb interaction leads to a suppression of the current flow.
Another important theoretical aspect of a current-biased single junction are the single electron tunneling oscillations. As is shown in figure 2.3, the current source will charge the junction until the critical charge $e/2$ is reached. Then, the electron tunnels and the current source will recharge the junction. This process will cause voltage oscillations with a frequency of $I/e$ to appear across the junction. For single electron tunneling (SET) oscillations, a time correlation of tunneling leads to voltage oscillations.

In practice, neither of these effects has ever been observed[45]. There are several reasons for this. First, it is almost impossible to experimentally achieve an ideal current source. The leads to the junction are resistive and cause current biasing to become voltage biasing. With voltage biasing, the bias forces the junction to maintain a fixed voltage, quenching the
SET oscillations. Also, the environmental modes of the leads[54], dampen SET oscillations and obscure the Coulomb blockade signature in the I(V) with their own structure.

2.2.2 Double Barrier Junction

Double junctions have met with considerably more experimental success [38]. Part of this success is that they are voltage biased, so the lead resistance is no longer a problem. Shown in figure 2.4 is the schematic of a voltage biased and gated double junction. The two junction capacitances and resistances are \((C_1,R_1)\) and \((C_2,R_2)\) respectively. The structure is assumed to be symmetrically biased by two sources \((-V/2\) and \(+V/2\)), and the gate is assumed to have a capacitance of \(C_G\) and a potential of \(U\). Considering the system shown in figure 2.4, the capacitance of the system is \(C_1\) added in parallel to \(C_2\), and therefore the total charge \(Q\) in this system is given by

\[
Q = \frac{C_1Q_1 + C_2Q_2}{C_1 + C_2}.
\] (2.8)

While the total charge, \(Q\), is coupled to the outside via the leads, the difference \(Q_1 - Q_2\), which gives the charge, \(-ne\) (\(n\) electrons on the metal island formed between the two junctions) is isolated from the environment[45]. The deciding factor on whether electrons will tunnel depends on the change in the free energy,

\[
E = \sum_{i=1}^{N} \frac{q_i^2}{2C_i} - \sum_{j=1}^{M} q_j V_j.
\] (2.9)

The sum over \(i\) accounts for the energy stored in the \(N\) capacitors of the system. The sum over \(j\) accounts for the work done by the \(M\) voltage leads[6]. For the case of a double junction with a gate (as shown in figure 2.4), this again leads to a situation where a bias voltage will cause tunneling when

\[
\Delta E = \frac{e}{2C} \left[ \left( \frac{C_2 + \frac{1}{2}C_G}{C_1 + C_2 + C_G} \right) V + C_G U + q - e/2 \right] = 0.
\] (2.10)

In the case without a gate, and where \(C = C_1 = C_2\), this reduces to a voltage,

\[
V = \frac{e}{2C}.
\] (2.11)
Figure 2.4: Schematic of a double barrier gated junction, symmetrically biased.
again resulting in a Coulomb blockade of current inside a voltage range. This has been experimentally observed in both metals [38] and semiconductors [60]. By tuning the gate potential, \( U \), it is possible to tune where the blockade voltage will occur, and also to select the number of electrons that reside on the middle island.

There is a second-order process, known as charge macroscopic quantum tunneling (MQT), where charge tunnels simultaneously through both junctions. The rate for this process is

\[
\Gamma \propto \left( \frac{R_K}{R_T} \right)^N,
\]

(2.12)

where \( N \) is the number of junctions in series. For reasonable tunneling currents, there is not much design latitude in changing the tunnel junction resistance \( (R_T) \). The only way to reduce the amount of MQT leakage current is to increase the number of junctions.

### 2.2.3 One-D Array of Junctions

For a one-dimensional array of junctions, a Coulomb blockade of current occurs for biases less than

\[
V = \frac{e}{2C}(N - 1),
\]

(2.13)

where \( N \) is the number of junctions in the array [39]. Arrays of junctions are usually treated with a capacitance matrix approach [7].

\[
C_{ij} = C_0 \text{ for } i = j
\]

\[
C_{ij} = C \text{ for } i = j \pm 1
\]

\[
C_{ij} = 0 \text{ for } i \neq j, j \pm 1
\]

(2.14)

This model assumes a self-capacitance, \( C_0 \), for every island, and a coupling capacitance, \( C \), between adjacent islands. All other interactions are neglected. This generates a set of linear equations which describe the potentials,

\[
-C\phi_{i-1} + (2C + C_0)\phi_i - C\phi_{i+1} = q_i \quad i = 1, ..., N - 1.
\]

(2.15)
The solution of this model leads to charge solitons, whose motion is correlated both in time and space, due to the soliton-soliton repulsion. This is speculated to be a better way to search for SET oscillations than using single junctions. There has been limited experimental success in observing these oscillations [26].

2.3 Energy Quantization

In semiconductor heterostructures that utilize layers for wells and barriers, it is possible to examine quantum confinement effects in the electron transport characteristics of the structure. There are a few important cases to consider. In this section, a simple quantum well will be discussed first. Next, the basic theory of superlattices will be shown. Lastly, quantum dot structures will be discussed. For these systems, lateral confinement becomes important, and as well as the charging effects discussed in the previous sections.

2.3.1 Quantum Wells

In a quantum well with infinitely high barriers, the electron energy of the \( n \)’th state is given by

\[
E_n = \left( \frac{\hbar^2}{2m^*} \frac{n\pi}{d} \right)^2.
\]  

In a real structure with a low lying state (i.e., GaAs wells, AlGaAs barriers), the finite barrier height usually has a large effect only for higher lying states.

Many material systems have been used for artificially creating quantum well structures for electronic purposes [109]. The most common is the GaAs/AlGaAs system. The quantum well structure forms the basis for a resonant tunneling diode (RTD) shown in figure 2.5. The resonant tunneling diode (RTD) consists of two electrodes (an emitter and a collector) that contact a quantum well. Figure 2.5a shows a conduction energy band diagram for a RTD under zero bias. The shaded regions in the emitter (to the left of the well) and in the collector (to the right of the well) correspond to occupied states in the GaAs conduction
Figure 2.5: Conduction band diagram of a resonant tunneling diode (RTD) structure under 0 mV (a), 100 mV (b), and 800 mV (c) bias.
band lying below the Fermi level. Under bias, little current flows through the structure until the quantum well state (black line in well) comes into resonance with the Fermi level in the emitter, as shown in figure 2.5b. At that point, a large amount of current flows through the device until the bias pulls the state below the emitter conduction band edge, at which point the device turns off (figure 2.5c). Figure 2.6 schematically shows the resultant I(V).

Thus, by using the quantum well, we can construct a unipolar diode that has four states (off/on/off/on). The additional state allows for logic compression over schemes that only employ conventional two state devices. However, a major problem with this approach is growth uniformity. Monolayer variations in the GaAs well and barrier width have large effects on the resonant bias of the device.

### 2.3.2 Superlattices

A superlattice is an artificial structure that creates a periodic potential. It is very similar in nature to an actual crystal lattice in that a summation of discrete states combines to form continuous energy bands. In the case of most semiconductor superlattices, the potential
modulation is at most only a few hundred meV. Therefore, the resultant energy band width
is usually on the order of tens of meV. Consequently, the convention is to distinguish the
artificially created band structure from the crystal band structure, by referring to the bands
and bandgaps as minibands and minibandgaps (or simply minigaps).

A simple way to solve for the minibands and minigaps in a superlattice structure is to
use the Kronig-Penny form

\[ \cos qd = \cos k A L A \cos k B L B - \frac{1}{2}(1/\xi - \xi) \sin k A L A \sin k B L B \]  

(2.17)

for bound states (below the barrier height) and

\[ \cos qd = \cos k A L A \cos k B L B - \frac{1}{2}(1/\xi + \xi) \sin k A L A \sin k B L B \]  

(2.18)

for unbound states, where \( \xi = m^*_A k A / m^*_B k_B \). \( L_A \), \( k_A \), and \( m^*_A \) are the well dimension,
wavevector, and effective mass, and \( L_B \), \( k_B \), and \( m^*_B \) are the corresponding values for
the barrier. \( q \) is the Bloch wave vector. Note that this solution takes into account the
effective mass discontinuity that occurs at a heterostructure interface. These equations
have solutions for \(-1 \leq \cos qd \leq 1\). The bands in figure 2.7 were calculated by applying
this model to a GaAs/AlGaAs superlattice with a conduction band offset (barrier height)
of 200 meV. The plotted curve is either equation 2.17 or 2.18 (depending upon the energy
being evaluated is a bound or unbound state). The shaded regions shown are where 2.17
and 2.18 have solutions, and thus depict the minibands. The spaces between them are the
forbidden energy regions (i.e., the minigaps).

More exact calculations to determine the band structure of superlattice can be done in
two ways. For finite period superlattices, a numerical heterostructure modeling program,
BandProf [35], may be employed (examples of BandProf solutions are shown in Chapter 5).
BandProf is a heterostructure modeling tool that calculates both band-bending and quantum
effects in a variety of heterostructure systems. From a description of the heterostructure
parameters (thickness, composition, and doping of layers), a self-consistent Poisson solution
Figure 2.7: Kronig-Penny model solution for a GaAs/AlGaAs superlattice with $\Delta E_C = 200$ meV, a 21Å well, and a 125Å barrier. The curve results from equations 2.17 and 2.18, and has solutions between -1 and 1. The shaded regions represent minibands, the white areas represent the minigaps, where there are no solutions, and thus no allowed energies.
is found. Once the potential is known, the quantum states in that potential can be solved for. This approach has the benefit that it can be used for a device under a bias, as this simply modifies the potential used for solving the quantum states.

An alternative approach is to solve both Poisson’s and Schrödinger’s equations self-consistently. This approach is computationally far more difficult and time consuming, and in practice does not predict superlattice bandstructure that is significantly different from the approach using BandProf alone.

2.3.3 Vertical Quantum Dot Structures

To date, almost all of the experimental work concerning quantum dot structures can be divided into two categories: lateral and vertical with regard to the direction of electron transport. Lateral quantum dot structures are usually formed by electrostatically defining a confining potential in a 2D electron gas formed in a modulation doped inversion layer. This approach has the advantage that it is relatively easy to place a gate near the dot. This locally affects the potential. Since the fabrication techniques that create the structure never actually expose the layers that form the conducting channels, it is usually possible to maintain high quality, high mobility conducting layers throughout the processing. The main disadvantage to such structures is that the potential walls are relatively soft (i.e., the barriers are not high or steeply sloped). The “softness” of the potential is due to the distance the gates are from the 2D electron gas. The gates that define the potential are usually a few hundred Angströms away from the conducting channel.

In the vertical case, which is studied in this dissertation, the confinement is through epitaxial layering in one dimension, and through fabrication created physical surfaces in the other dimensions. Much “harder” walls and higher barriers are obtained. The two main disadvantages to this approach are that these devices can not easily be gated (although this has been accomplished with 1 μm diameter devices [11, 24]) and that the fabrication required to make such a structure depends upon a critical highly anisotropic reactive ion
etching step. The underlying physics is similar for both cases. A vertical structure is presented in the sections that follow. Figure 2.8 illustrates the differences between (a) the starting epitaxial material, and (b) the method used for quantization.

Quantization in Zero Magnetic Field

Consider a quantum well which has been quantized in the vertical dimension through epitaxial growth. In the lateral dimension through fabrication, we create a system that is a quasi-0-D dot connected to the outside world by two 1-D leads. The vertical confinement in such a structure is modeled by a rectangular potential. The horizontal confinement, which occurs due to Fermi level pinning on the side walls, is modeled as a parabolic potential,

$$\Phi(r) = \Phi_T [1 - (R - r)/W]^2,$$

(2.19)

where $\Phi_T$ is the Fermi-level pinning energy at the sidewalls, $R$ is the lateral physical dimension, $r$ is the radial coordinate, and $W$ is the depletion depth.

Schrödinger’s equation for such a structure is best expresses in cylindrical coordinates. There is no $\theta$ dependence, and since $\Phi(r,z)$ can be written as $\Phi(r) + \Phi(z)$, separation of variables is possible. The single electron eigenstates are

$$E_N = E_z + E_{n,l}.$$  

(2.20)

$E_z$ comes from the vertical quantization, and this will almost always be $E_0$, the quantum well ground state in the vertical direction. $E_{n,l}$ are the eigenstates that result from the parabolic potential.

$$E_{n,l} = (M + 1)\hbar \omega_0$$

(2.21)

where $M = (2n + |l|)$, the radial quantum numbers $n = 0, 1, 2, ...$, and the azimuthal quantum numbers $l = 0, \pm 1, \pm 2, ...$. If we assume that we are in the first vertical subband, $E_0$, and that the lateral dimension is reduced to $2W$ or less, then the energy spacing between
Figure 2.8: (a) A comparison of the starting epitaxially grown material for a lateral quantum dot structure (right) to a vertical structure (left). (b) Same structures after the fabrication that forms the quantum dot.
states is
\[ \Delta E = \hbar \omega = \left( \frac{2\Phi_T}{m^*} \right)^{1/2} \frac{\hbar^2}{R}. \]
(2.22)

For a GaAs well, with AlGaAs barriers, typical values for energy state separation are on order tens of meV.

**Current versus voltage characteristics**

The current versus applied bias for such a structure is described by
\[ I(V) = e \sum_n \int_{E_F - eV}^{E_F} N_n(E) v_n(E) T_n(E, V) dE, \]
(2.23)
where the sum is over all 1D subbands, \( n \), below the Fermi energy \( E_F \) in the emitter. \( N_n(E) \) (\( \propto E^{-1/2} \)) is the 1D density of states of the \( n \)th subband in the emitter. \( v_n(E) \) (\( \propto E^{1/2} \)) is the velocity of electrons at energy \( E \) in subband \( n \), and \( T_n(E, V) \) is the transmission probability through the double-barrier structure. For the special case of 1D subbands, the product \( N_n(E) v_n(E) = 1/\hbar \) is constant. Assuming that \( T_n(E, V) \) near resonance is approximated by a Lorentzian line shape, with linewidth \( \Gamma_{e} \) and peak transmission of \( T_{0,n}(V) \), a current contribution of
\[ \Delta I_n = e \pi T_{0,n}(V) \Gamma_{e} / \hbar \]
(2.24)
is obtained for each spin state in every occupied emitter subband [102].

**Finite Temperature and Scattering**

Under finite temperature and in the presence of inelastic scattering processes, the line shape due to tunneling into a 0-D state is broadened. If we write the transmission \( T_n(E) \) as
\[ T_n(E) = \frac{T_{0,n}}{1 + \{2(E - E_n)/\Gamma\}^2}, \]
(2.25)
where \( E_n \) is the energy of a 0-D dot state, and \( \Gamma \) is the total line width which is composed of three parts, as shown in equation 2.26.
\[ \Gamma = \Gamma_{e} + \Gamma_{s} + \Gamma_{T} \]
(2.26)
$\Gamma_e = \hbar/\tau_e$ is related to the intrinsic lifetime, $\tau_e$, of the quasi-bound quantum state. $\Gamma_s = \hbar/\tau_s$ is related to the inelastic scattering time, $\tau_s$. $\Gamma_T \sim k_B T$ arises from finite temperature effects. For vertical quantum dot samples, $\Gamma_T$ is usually neglected below 1 K. $\Gamma_s$ is the dominant broadening effect. Scattering is due either to electron relaxation in the dot (by scattering into other subbands) or to phonon emission[101].

**Quantization in Finite Magnetic Field**

In the presence of a magnetic field, $E_{n,l}$ is now given as

$$E_{n,l} = (n + \frac{1}{2} + |l|/2)\hbar (\omega_c^2 + 4\omega_0^2)^{1/2} + (l/2)\hbar \omega_c,$$  \hspace{1cm} (2.27)

where $\omega_c = eB/m^*$ is the cyclotron frequency and $\hbar \omega_0$ is the energy spacing at $B = 0$.

As expected, for $B = 0$ this reduces to equation 2.21, having a $(M + 1)$-fold degeneracy for each spin. Under magnetic field, this degeneracy is lifted. For $\hbar \omega_0 = 0$ (no confining potential), equation 2.27 becomes

$$E_{n,l} = (N + \frac{1}{2})\hbar \omega_c.$$  \hspace{1cm} (2.28)

$N = n + l/2 + |l|/2$ where $N$ is the Landau-level.

**Coulomb Charging Effects on a Quantum Dot**

Until now, electron-electron interactions have been neglected. As discussed in section 2.2.2, charging effects are important as the lateral device area is scaled down. For a cylindrical device, charging by a single electron leads to a charging energy,

$$E_C = \frac{e^2}{2C},$$  \hspace{1cm} (2.29)

where

$$C \approx \frac{e_0 \pi a^2}{4d}, \hspace{0.5cm} a/d \gg 1.$$  \hspace{1cm} (2.30)

In the expression for the capacitance, $a$ is defined as the device's electrical width ($a \approx 2(R - W)$), and $d$ is the barrier thickness. If we assume a quantized energy spacing given by
a 0-D box, $\Delta E \approx \hbar^2 / m^* a^2$, and the ratio of the charging energy, $E_C$, to the quantization energy, $\Delta E$, is given by

$$\frac{E_C}{\Delta E} \approx \frac{d e^2 m^*}{4\pi \epsilon \epsilon_0 \hbar^2} = \frac{d}{a_B},$$

where $a_B \simeq 10$ nm is the effective Bohr radius in GaAs. This means that for $d = 10$ nm, the ratio is unity and single electron charging should be apparent in vertical quantum dot structures when $d/a_B \leq 1$.

How the combination of quantization and charging effects impacts the electron spectroscopy depends on many factors. Consider the case of an asymmetric structure[94], where $T_E$, $T_C$ are the transmission coefficients through the emitter and collector respectively. Let $T_E \ll T_C$. Under positive bias (injection through emitter), electrons that tunnel through the emitter at a rate $\Gamma_E$ leave faster than they are replaced (as $\Gamma_E \ll \Gamma_C$). In this case, a Coulomb blockade effect occurs for the first electron filling the quantum dot. Below the voltage threshold, $V_{th}$, no current flows through the structure. After $V_{th}$ is reached, a step in the current appears. The current is then modulated by the quantum dot density of states (DOS), as described in equations 2.20, 2.21, and 2.22. This is shown schematically in the forward bias region of figure 2.9. In this bias configuration, occupation of the dot by a second electron is not probable. For negative bias (injection from the collector), since $\Gamma_C \gg \Gamma_E$, charging has a much more important role. It is now probable to charge the quantum dot with more than one electron. In this bias direction, one sees a series of current steps with increasing bias, which correspond to incremental charging of the quantum dot by an additional electron. On top of this structure, one is also able to distinguish the quantum dot DOS (shown in reverse bias of figure 2.9). It should be noted that the charging energy for having one electron in the well, $U_{1e}$, is usually less than that of $U_{2e}$ (two electrons in the well). For two electrons in the well, electron-electron repulsion energy contributes. In general, the energy required for the transition from an $N$ to an $N + 1$ state involves the complex interactions of the many-electron system.
Figure 2.9: Theoretical current versus voltage for an asymmetric vertical quantum dot structure.
Chapter 3

Experimental Methods

3.1 Introduction

Three major experimental systems that consist of an electron beam lithography system, a long working distance thermal evaporator, and a cryogenic electrical characterization system were developed during the course of this dissertation. As these systems were crucial for performing much of the work described in this dissertation, they will be documented in some detail. The electron beam lithography system is crucial to all of the work where sub-micron lithography is required. It is also used in a novel crystallite growth inhibition study discussed in chapter 7. In addition to having sub-micron lithography capabilities, it is important to have a long working distance metal evaporation system to deposit metal structures. A high vacuum deposition system was designed and built to obtain very small metal lift-off features. Also, in order to electrically characterize low dimensional structures, cryogenic and specialized low noise electronics systems were developed.
3.2 Yale $\mu$ELM Electron Beam Lithography System

3.2.1 Hardware

The Yale $\mu$ELM Electron Beam Lithography System was constructed at Yale University from components received from JEOL (Japanese Electron Optics Ltd.) and from custom components and software created at Yale. Figure 3.1, shows a block diagram of the electron beam lithography system. The first major component is a JEOL 6400 Scanning Electron Microscope (SEM). This SEM has an ultimate spot size of 35Å, using a LaB$_6$ source in the electron gun. The LaB$_6$ source has advantages for use over a tungsten source as it is about ten times brighter (in C/cm$^2$). Since electron beam lithography is a serial process, this translates into accomplishing the lithography in one tenth the time. This point cannot be overemphasized. A project requiring a ten hour write time might not be attempted, whereas the one hour write would be. Beam current typically varies from the pA range for very fine scale (<100nm) lithography, to levels approaching 100 nA for large area (micron size) pixels. Fine scale lithography is performed at an electron beam working distance (WD) of 8 mm. Coarse lithography (>100nm) may be accomplished at 39 mm, even though the beam dispersion is greater at higher WD (having a negative effect upon the resolution).

This SEM has a few additions to it, supplied by JEOL, which aid in its conversion to a lithography tool.

I. External Scan Control: This unit allows an externally provided analog signal to control the electron beam. Normally, an SEM raster scans the sample. With this option, it is possible to move the beam to any position inside the given scan field (determined by the magnification used).

II. Beam Blanking Device: This allows an external signal to blank the beam. It operates by electromagnetically deflecting the beam, and can be driven at speeds of up to 700 kHz.
Figure 3.1: Block diagram of the Yale μELM electron beam lithography system.
III. Probe Current Detector: This unit measures the current the electron beam provides, and is essential for obtaining reproducible dosages.

IV. Optical Camera: At a working distance of 39mm, a removable optical camera can be used. This is extremely valuable for aligning samples (in x, y, and θ) without exposing the materials that are routinely used for electron beam resists (i.e., polymethylmethacrylate (PMMA)). It also aids in work at 8mm. Alignment is achieved at 39mm, and then the camera is retracted. The sample is then brought to a WD of 8mm.

V. External Computer Control: This option allows for external control of most of the SEM functions via a serial line.

VI. Stage Automation: A dedicated 486 PC controls the stage automation system, which controls the position of the stage along three axes (x, y, and z). This system allows for the repositioning of the stage to preset positions with accuracy of ±2μm. It also has external control, via a serial line, as the SEM does. A problem with this system is that, while the motors that move the stage can step with an accuracy of 100Å, and built-in corrections take care of backlash, the actual stage is an SEM stage (not designed with lithography intended). This has two major drawbacks. First, the X and Y axes are not orthogonal. It is typical to line up the Y axis, travel a few mm along the X axis, and see the system systematically drift by tens to hundreds of microns. The further you travel, the worse it gets. What is even more problematic is that the axes are also non-linear in regions. This issue is discussed further in the software section, 3.2.2.

In addition to what is supplied by JEOL, another 486 PC was purchased to control the lithography system. The Control PC communicates serially to both the SEM and the
stage computer. It also has a custom board, developed at Yale, that controls both the beam blanking device and two 16 bit digital to analog converters (DACS). This allows the Control PC to completely control electron beam dosage. It positions the beam, opens the beam up for a specified time, and then shuts the beam. By repeating this process, point by point, any shape can be written. The NorthGate also has a National Instruments, general purpose, analog to digital (A/D) and digital to analog (D/A) board, used in the A/D mode to convert signals from the secondary electron detector into images for the automatic alignment routines, which are described further in the next section.

3.2.2 Software: Align 6.0

The software that controls the system has gone through many revisions. The current code was written by myself and Mr. James McCormack, and is detailed in a separate manual.[70] A brief summary of its capabilities follows.

One of the main goals in the design of the program was to avoid using proprietary computer aided design (CAD) programs that are costly, and are only available on workstation platforms. Therefore, the CAD input file that Align 6.0 uses is the DXF format file, which can be created with several commercially available CAD packages for Macintoshes and PC’s. This allows the user to easily create the lithographic pattern without having to use the computer that runs the electron beam system.

Once the program loads in the DXF file, many options are possible. The simplest option is just to write the current pattern. The user has to select a magnification (for field size), a beam current (which relates to beam size), and a writing pixel size. The beam current divided by the area of the writing pixel size is used to calculate the deposited dose. Since many of these parameters are inter-related, updating one of them may affect others. In general, you select the magnification size first. This determines the field size (100x = 1000\(\mu\)m x 1000 \(\mu\)m, 1000x = 100 \(\mu\)m by 100 \(\mu\)m). Once the field size is set, the number of DACS units per micron can be determined. Then, the pixel size (typically 20\(\AA\), 50\(\AA\), 100\(\AA\),
200Å, 500Å, or 0.3 μm) is set. From an empirically determined look-up table, a suggested beam current is calculated. Once the SEM is properly focused and aligned to the sample, writing may be started.

There are a number of options beyond simply writing one isolated pattern.

I. Step and Repeat: A pattern is stepped and repeated an arbitrary number of times in X and Y. While the step and repeat is being done, a program parameter allows for increasing the exposure dosage at each step. This is very useful in determining the correct exposure parameters for a given resist at a set beam condition.

II. Stage Correction: As mentioned in the section 3.2.1, the stage design leaves much to be desired. Fortunately, many of the problems can either be corrected or greatly alleviated via software. A routine was written that allows user to specify the correct stage locations at every write position. This requires a pre-existing layer to be fabricated on the substrate, used for these alignment purposes. This usually succeeds in achieving alignment to better than 2 μm. Alignment to better than 1 μm may be obtained in two ways. First, one may use a central alignment mark on the sample, expose that region to view, and center it. The other possibility is using automatic alignment (see following item).

III. Automatic Alignment: A recent addition to the program allows for a layer in the drawing to specify an expected alignment layer pattern. This pattern is not written. Instead, the program uses the electronics that it normally writes with to scan the sample and form a two bit image. Currently, the signal from the secondary detector is amplified and filtered by a PAR 113 preamplifier. It is then brought into the NorthGate using the National Instruments A/D card. The scanned pattern is compared to what is expected, and the stage computer is then instructed to move accordingly. A variety of approaches, as well as
alignment patterns may be used. For a given setup, alignment better than 1 
\( \mu m \) may be achieved after optimizing the parameters. It is also possible to have 
the program electronically shift the pattern to achieve final alignment within 
\( \pm 0.25 \mu m \).

IV. Native File Type: In addition to reading DXF files, the program can output 
and input files in its own format, designed to be easy to read and edit with 
any generic text editor. This is useful in situations where you want to alter the 
order in which entities are written (especially in difficult cases involving resist 
charging).

V. Variable Exposure Control: Objects on the same write layer, in CAD, may 
be given different colors. These are interpreted as corresponding to different 
dosages. This gives a primitive, but very useful, first order proximity effect 
correction mechanism. This is utilized in situations where the optimum exposure 
level for the small area structures is too high for the large area structures.

3.2.3 System Results

The system has been used in many collaborations. This section shows a sampling of these 
results. More results from this system are shown in later chapters. The resist that is used 
for all of the metal lift-off results is a bilayer. The primary layer is a 5% solution of 150k 
m.wt. PMMA spun on at 3000 RPM (which typically results in a layer approximately that 
is 2000Å thick). The top layer is a 2% solution of 950k m.wt. PMMA spun on at 8000 
RPM to achieve a layer that is only 500Å thick. In all cases, the solvent is 100% MIBK. 
The development is always a 3:1 mixture of isopropanol alcohol to MIBK, for 30 seconds. 
Picking the correct electron beam dosage to achieve proper development at 30 seconds is 
observed to give the best resist profiles (in terms of aspect ratio and broadening).
Figure 3.2: 20 - 30 nm gold dots formed using electron beam lithography on a Si substrate.

Small Dots

One of the first tests of any lithographic system is to determine what is the smallest feature that can be produced. Figure 3.2 shows a closeup of some gold dots that were formed via lift-off on a Si substrate. This is a step which is important in the fabrication of vertical quantum dot structures. As can be seen, 20 to 30 nm resolution is obtained. What is even more impressive is the creation of a large array of these dots. Figure 3.3 shows a 25 by 25 array of these dots, and figure 3.4 shows this pattern stepped and repeated with the stage control.

Fields of dots are also used to estimate proximity effect. Shown, in figure 3.5, are a series of dots, that starting from the upper left hand corner, are spaced further and further apart. The large cluster in the upper left are five dots in the CAD that resulted in one large dot in the metal lift-off, due to the proximity effect which exposed the resist in the regions
Figure 3.3: A 25 by 25 array of gold dots on a Si substrate, showing the written field (A lower magnification view of figure 3.2).
Figure 3.4: A 5 by 5 array of gold dots on a Si substrate, showing the field in figure 3.3 stepped and repeated.
Figure 3.5: A series of dots that starting from the upper left hand corner, are spaced at increasing distance from each other. The cluster in the upper left hand corner occurs due to the proximity effect.

between the closely spaced dots.

**Lines**

Lines are the next obvious entity to investigate. Lines as thin as 50-100 nm have been fabricated using this system. Figure 3.6 shows a 250 nm metal line that was fabricated for creating a modulated magnetic field in a two-dimensional electron gas (2DEG).

**Rings and Stadiums**

The fabrication of more complex structures has also been achieved. Figure 3.7 shows a metal ring created on top of a GaAs 2DEG. The kink at the bottom of the ring illustrates a resist charging problem. Due to the order in which objects were written, the charging
Figure 3.6: A 250 nm wide meandering line, Au/Ti on Si substrate.

deflected the beam causing the kink.

Another project that required extensive use of the electron beam lithography system involved the creation of sub-micron cavities in a high mobility GaAs 2DEG. These were created to study the differences between chaotic and non-chaotic transport systems. For example, more chaotic-like signatures in transport are expected to be seen from an electron undergoing collisions with a stadium shaped potential, than from a circular shaped potential. The transport and analysis of these structures is given in great detail in Dr. Mark Keller’s dissertation[58]. In order to create these potentials, electron beam lithography, with a 1000Å monolayer of PMMA, is used to create an etch mask for the GaAs 2DEG. Etching away the layer in the selected regions defines the potential. The figures that follow (3.8, 3.9, 3.10, and 3.11 ) show a series of atomic force microscopy (AFM) images of the PMMA after lithography and development.
Figure 3.7: A metal ring fabricated on top of a GaAs 2DEG.
Figure 3.8: AFM image showing four different types of small structures used in the quantum chaos project. The dark background is where the PMMA has been removed in the developer. Clockwise, from the upper left corner, are a ‘stomach’, a stadium, a 1-D wire, and a polygon.
Figure 3.9: AFM image showing a closeup of the polygon structure, created in PMMA on GaAs.
Figure 3.10: AFM image showing a closeup of the ‘stomach’, created in PMMA on GaAs.
Figure 3.11: AFM image showing a closeup of the stadium, created in PMMA on GaAs.
3.3 Very Long Working Distance Thermal Evaporator

A problem that arises in defining very small metallic features using lift-off is shown in figure 3.12. For a finite thermal metal source size, $d_s$, the lithographic feature in the top layer of the resist, $d_L$, will become broadened due to shadowing ($d_f > d_L$). The improvement that is expected for typical resist and source geometries, is roughly a factor of three for this system over the Varian thermal evaporator in the Yale cleanroom. The broadening of features in the Varian is on order 10 nm, versus 3 nm for the new system.

The system is cryopumped, and has a Fomblin based mechanical pump to minimize hydrocarbon contamination. The minimum base pressure is approximately $5 \times 10^{-8}$ Torr. This can be reached in under 24 hours from venting to atmosphere. The thermal evaporation power source is a Sorenson DC supply, capable of providing up to 7 Volts at up to 300 Amps. A high current switch allows for selecting between one of three boats. Pressure is monitored using thermocouple gauges from atmosphere to 50 mTorr, a Pirani gauge from 2000 mTorr to 1 mTorr, a Baratron gauge from 100 mTorr to 0.01 mTorr, and an ion gauge from $1 \times 10^{-3}$ Torr to $1 \times 10^{-8}$ Torr. A thickness monitor, capable of storing parameters for eight different films is also present. The sample is clamped to a block which is coupled via a thermoelectric (TE) cooler to a water cooled block. Without using the TE cooler, the sample maintains the temperature of the cooling water during the deposition (a thermoelectric temperature sensor is situated near the sample). Using the TE cooler, the sample temperature is lowered to about 70°C below the water temperature. This TE cooler system is used to regulate the sample temperature during a deposition, in order to provide more consistent results. The system also has a controlled leak valve gas inlet which is currently attached to oxygen. This is used to oxidize a metal (i.e., aluminum) just after a deposition.
Figure 3.12: Schematic of finite source size effects on thermal evaporation of metal through a PMMA bilayer resist.
3.4 Electrical Measurements at Low Temperatures

The electrical characterization of quantum-size-effect structures typically necessitates measurements at low temperatures. It is relatively easy to obtain temperatures from 300K to 1.2K in a commercially available cryostat using $^4$He as a cryogen. Temperatures below 1.2K are generally not possible without the use of more elaborate refrigeration techniques. A simple way to get to 300 mK is use $^3$He instead of $^4$He, however this adds greatly to the complexity of the cryogenic system. Due to the high cost of $^3$He, it must be recovered. Therefore a closed-cycle pumping system is required. Using a mixture of $^3$He and $^4$He, a dilution refrigerator, which will be discussed later, typically obtains temperatures in the 10 mK range.

As $kT$ decreases, experimental voltage resolution becomes increasingly important. Many transport effects that show characteristics in the I(V) are dependent upon how the Fermi level in the sample broadens. At room temperature, $kT \approx 26$ meV, so mV resolution is adequate. At 1.2K, $kT = 0.1$ meV, and at 30mK, $kT = 2.5 \mu$eV. This means that unless we can measure the sample with $\mu$V resolution, we are not be able to resolve the sharp features that result from the increasingly sharp edge of the Fermi function.

3.4.1 Cryostat Systems

Janis Supervaritemp

For the measurements taken between room temperature and 1.2K, a Janis Research Supervaritemp cryostat is used. This system consists of an outer liquid nitrogen dewer, separated from an inner liquid helium dewer by a vacuum wall. The system can be operated in a variety of modes. The sample can be immersed in He, and by varying the pressure over the sample with a Walker regulator, temperatures between 1.2K and 4.2K are maintained. Temperatures higher than 4.2K are generally reached by heating the He as it passes through the capillary into the sample reservoir. Although this may be used to regulate temperatures
from 4.2K to 300K, in practice, it is usually used up to 120K. At this point the He flow is shut off. The system temperature rises slowly enough to allow for acquisition of data every ten degrees, with no significant thermal averaging.

Several components were added to this system to aid in the ease of use. The pre-cool of the outer liquid nitrogen dewer typically takes 12-24 hours, and several re-fills of liquid nitrogen. In order that this be automatically accomplished overnight, an Oxford Instrument liquid nitrogen level sensor was acquired. A valve controlled by this sensor fills the system when the level drops below 30%. A LakeShore 330 Temperature Controller is used to monitor the temperature of the silicon diode sensors placed at the bottom of the liquid He dewer. This aids transferring liquid He, as you can tell by the temperature when liquid He is accumulating on the bottom of the dewer. Additional sensors are placed at the He vaporizer coil, and at the sample. This controller is used to regulate the temperature in the system, or simply to read out the temperature. It has a computer interface, and together with a Macintosh IIci and a HP4145 Semiconductor Parameter Analyzer (SPA), form the basis for an automated current versus voltage and temperature system. Automating this system was crucial, in that the warm up time from 1.2K to room temperature is on the order of days.

The software written to control the systems allows the user to take I(V)'s with the SPA at a pre-set temperature list (either a start value, a stop value, and an increment, or a file of desired temperatures). In order to make transferring He more efficient, and to monitor \(^4\text{He}\) consumption during experiments, an Oxford Helium Level sensor was added.

The sample is mounted on a removable stick, which is wired for both a standard 16 pin DIP and a Yale 8 pin non-magnetic header. The system, as delivered, contained four quartz optical windows. However, for the work in this dissertation, these windows were removed and replaced with metal blanks to ensure that electrical characterization occurring in the dark.
Oxford Kelvinox Dilution Refrigerator

For measurements under 1.2K, an Oxford Kelvinox 25 dilution refrigerator is used. The measured cooling power of the Yale dilution refrigerator is 100µW at 100 mK, and a base temperature of 27 mK has been obtained. The sample is mounted on either the Yale 8 pin header, a 16 pin commercial DIP, or in a custom 24 pin socket. A 9 Tesla superconducting magnet is available for magnetotransport studies. 11 Tesla is obtained by pumping on the lambda plate attached to the magnet, thus reducing the magnet temperature.

Again for this system, computer control is important, as low noise I(V)’s take 5-10 minutes to acquire. Stepping from 0 to 9 Tesla at fine increments (i.e., 2000 Gauss) takes 40 hours, or more. For the magnetotransport experiments, the control program that ran the Janis was modified to also control the magnet supply in the dilution refrigerator. In this manner, I(V,B) traces are acquired with almost no user intervention required (except to monitor the dilution refrigerator).

The dilution refrigerator is also under computer control, which allows monitoring of temperature sensors at the various important points in the dilution refrigerator (the still, the mixing chamber and sample, and the 1K pot). Details of this and other aspects concerning the operation of the refrigerator, and the specifics concerning the Yale refrigerator, are covered in the Oxford manual[8] and in Dr. Mark Keller’s dissertation[58].

3.4.2 Low Noise DC Electronics

As indicated in the earlier section, a great deal of data collecting automation occurs in many of the experiments, utilizing a computer to control the various experimental parameters and data acquisition. In most cases, the heart of the data acquisition system is an HP4145 Semiconductor Parameter Analyzer (SPA). Unfortunately, while the SPA has excellent current resolution (around 50 fA), its minimum voltage resolution is only 1 mV. As was previously discussed, this is not adequate for measurements at 1K and below.

In order to make measurements at a finer voltage scale, a special ultra low-noise cur-
Figure 3.13: Schematic for the low-noise voltage/current preamplifier.

The basis of operation is as follows. A symmetric bipolar voltage signal, typically on the order of tens of volts, is supplied by the SPA as the input to the preamp. This signal is immediately attenuated in the preamp by a selectable factor of two, twenty, or two hundred. This results in a corresponding reduction in the signal to noise ratio of the input signal. The preamp is battery powered to reduce 60Hz noise, the ground of the box is isolated from the ground of the SPA to kill grounding loop pickup, and capacitors to the box ground serve to shunt high frequency noise components.

The attenuated signal is then routed across two bias resistors, in series with the sample,
as shown in figure 3.14. The voltage across the sample is determined from the instrumentation amplifier, IA1. The current is determined by measuring the total bias voltage with IA3, and subtracting from it the voltage across the sample, given by IA2, by using the output of IA2 for the reference signal of IA3. For a given bias resistor $R_b$, the current for the sample is related to the output of IA3 by

$$V_{out3} = G_I V_0 - G_I V = G_I 2R_b I$$

(3.1)

The instrumentation amplifiers (IA1, IA2, and IA3) consist of three discrete OP-amps, using two Burr-Brown OPA111BM and one Burr-Brown INA105BM (shown in figure 3.15). These instrumentation amplifiers have $\mu$V level voltage noise and approximately 10 fA level current noise in the frequency range where the measurements are made.

Extreme care was taken in the construction of this unit, as very high resistance bias resistors are used for $R_b$. These resistors are mounted on a machined Teflon board, and much of the internal wiring is teflon coated (teflon resistance is greater than $10^{15}$ Ohms). All inputs to the instrumentation amplifiers are brought in on flying teflon coated leads (not attached to the printed circuit board, which can have resistances of only $10^{12}$ Ohms). Also, high resistance ceramic selector switches are used.
Figure 3.15: Schematic of instrumentation amplifiers used in figures 3.13 and 3.14.

Figure 3.16 shows the improvement in I(V) characteristics that are obtained using this box. The sample used is a quantum dot structure, cooled to 50 mK. The step-like features in the I(V) are due to tunneling through the discrete dot states, and the width of the leading edge is very sensitive to temperature. This work will be discussed in detail in chapter 6. It is apparent that without the preamp setup, much of the important structure in the data is lost. It is also critical that the correct value of $R_b$ be used, as regions that exhibit negative differential resistance will not be measured if the load line determined by $R_b$ is not steep enough. This is seen by the blank regions in the $R_b = 10$M curve in figure 3.16.
Figure 3.16: IV data from a vertical quantum dot structure acquired using the low-noise preamplifier (two different values of \(R_b\) shown), and also with the HP4145 SPA alone.
Chapter 4

Spectroscopic Study of Intra-miniband and Inter-miniband Tunneling in Finite Superlattices

4.1 Introduction

The artificial creation of periodic structures, superlattices, and the characterization of them using electron transport and optical techniques has been a very active research area. In the GaAs/AlGaAs material system, varying the barrier and well thicknesses as well as the barrier heights gives great control over the created lattice, and therefore the type of band structure one can study. In weakly coupled superlattices it has been shown [21] that the perpendicular transport proceeds via sequential tunneling, whereas under the proper conditions a miniband forms[30,27,89] possessing a constant 2D density of states inside the band.
In the limit where there are few superlattice periods (N) and the miniband width (ΔW) is relatively wide, it is possible to experimentally probe the individual eigenstates which compose the miniband when \( kT/(ΔW/N) \ll 1 \). Relatively little work has focused on this aspect of miniband formation, where the transport in the structure shifts from sequential tunneling through coupled quantum wells to coherent transport through minibands. Davies and co-workers [23] were the first to investigate a superlattice / barrier / superlattice structure. Under external bias, electrons are injected through a barrier from the Fermi level of one superlattice (hereafter defined as the emitter) into the downstream (collector) superlattice allowing a study of the density of states of the collector superlattice. More recently, [1] work involving superlattice / barrier / superlattice structures (superlattice tunnel diode) has shown the existence of the individual finite superlattice eigenstates. Presented here is a tunneling density of states study of the transition from a superlattice miniband to a coupled well structure for both inter-miniband and intra-miniband cases, and evidence for intra-miniband tunneling processes involving longitudinal optical (LO) phonon emission.

The aim of this study is to investigate both the bandstructure of superlattices and the electron transport through superlattice minibands. The superlattices are characterized by photoluminescence and transmission electron microscopy, as well as electrical spectroscopy.

### 4.2 Experiment

Figure 4.2 is a transmission electron micrograph (TEM) of the type of superlattice tunnel diode structure investigated in this study. The three superlattice emitter, barrier, and superlattice collector are clearly visible. TEM measurements were used throughout this study to verify epitaxial dimensions. Figure 4.1 is a self-consistent conduction band diagram of the structure shown in figure 4.2 under resonant bias. This specific example is identical to the initial work of Davies, et al.[23] The band diagram is determined from a self-consistent finite temperature Thomas-Fermi zero-current calculation, and the quantum states are found
by direct evaluation of the resonances of the effective-mass Hamiltonian.[35] Under voltage bias, current will flow through the superlattice tunnel diode until the top of the first collector miniband crosses the bottom of the available emitter electron supply. Then a decrease in current occurs due to the requirement to conserve both energy and momentum.

This superlattice tunnel diode structure consisting of a superlattice emitter, a relatively thick barrier, and a superlattice collector is utilized to study the density of states of a series of superlattice structures. The barrier is used primarily to narrow the energy distribution of the electrons that tunnel into the collector. Table 4.2 illustrates the series of superlattice structures investigated. Superlattice dimensions are determined from TEM’s and the Fermi level and miniband positions are calculated using a fully self-consistent Poisson-Schrödinger calculation.[67] All structures were grown in a Riber 2300 MBE on In-free mounted GaAs wafers. Sample 1 was grown nominally identical to that of Davies, et al. [23] in order to
Figure 4.2: A TEM (transmission electron micrograph) of sample 1. The central line is 55Å for scale.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$d_{\text{GaAs}}/d_{\text{AlGaAs}}$ (Å)</th>
<th>$E_{\text{SL,n}} - E_{\text{C,GaAs}}$ (meV)</th>
<th>Width ($\Delta W$) (meV)</th>
<th>$E_{\text{F,SL}} - E_{\text{C,GaAs}}$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>52/21</td>
<td>45,204</td>
<td>44,199</td>
<td>47</td>
</tr>
<tr>
<td>2</td>
<td>34/42</td>
<td>87</td>
<td>33</td>
<td>87</td>
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<tr>
<td>3</td>
<td>41/10</td>
<td>35</td>
<td>165</td>
<td>46</td>
</tr>
<tr>
<td>4,5,6</td>
<td>36/9</td>
<td>37</td>
<td>214</td>
<td>47</td>
</tr>
<tr>
<td>7</td>
<td>117/25</td>
<td>19,77,176</td>
<td>9,44,74</td>
<td>27</td>
</tr>
</tbody>
</table>

Table 4.1: A summary of parameters for the seven superlattice samples. The first column lists the sample number. The second column lists the thickness of the well and barrier ($d_{\text{GaAs}}/d_{\text{AlGaAs}}$), the third column lists the bottom of the superlattice minibands relative to the GaAs conduction band edge ($E_{\text{n,SL}} - E_{\text{C,GaAs}}$), the fourth column lists the miniband widths ($\Delta W$), and the last column lists the Fermi level in the superlattice minibands relative to the GaAs conduction band edge ($E_{\text{F,SL}} - E_{\text{C,GaAs}}$).
compare to previous work; however TEM reveals some differences in our structure (i.e., somewhat smaller barrier and wells widths than in Davies work, although their TEM's revealed ±5Å variances across the structure, as well as an asymmetry between the collector and emitter superlattices). The remaining samples consist of a Cr-doped semi-insulating GaAs substrate, a 0.5 mm undoped GaAs buffer, a 1 mm $1 \times 10^{18}$ cm$^{-3}$ Si-doped n$^+$ GaAs bottom contact, a 420Å $1 \times 10^{17}$ cm$^{-3}$ (last 20Å undoped) GaAs contact to superlattice transition region, a superlattice / tunnel barrier / superlattice region symmetric about the tunnel barrier, a 420Å $1 \times 10^{17}$ cm$^{-3}$ (first 20Å undoped) GaAs contact to superlattice transition region, a 400Å $2 \times 10^{18}$ cm$^{-3}$ GaAs top contact, and an InGaAs top non-alloyed ohmic contact. Samples 1, 2, 3, 4, 5, and 7 contain 10 period superlattices. The GaAs wells of these superlattices are doped at $1 \times 10^{17}$ cm$^{-3}$, the barriers are nominally undoped, and the tunneling barrier is kept fixed at 88Å of AlGaAs (except for 7 which has a 167Å barrier). To insure accurate superlattice spectroscopy, the tunneling barrier must be the dominant impedance in the structure and be sufficiently thick to decouple the ten period superlattices on either side of the barrier. Sample 2 has the same approximate miniband width (33 meV) as Sample 1, except the position of the miniband is higher (approximately by a factor of two) in energy with respect to the conduction band of GaAs, making the second miniband completely "virtual" (i.e., the second mini-band is entirely above the conduction band edge of the superlattice barriers). Sample 3 is designed to have the same miniband centroid as Sample 2, except with a miniband width five times larger (165 meV). In addition, it has symmetric 400Å $1 \times 10^{17}$ cm$^{-3}$ contact regions adjacent to the superlattices to study the effect of contacts. Sample 4 is designed to have a very large miniband width (214 meV, with the top 51 meV of the first miniband "virtual", with respect to the AlGaAs tunnel barrier) having easily experimentally resolvable eigenstates spaced by approximately 21 meV. Sample 5 is identical to Sample 4, except that the bottom superlattice is replaced with bulk GaAs (though the doping modulation is identical), to investigate injection into a superlattice from a three dimensional system and vice versa. Sample 6 is identical to
Sample 5, with a change in the number of superlattice periods from 10 to 20. Sample 7 explores the intra-miniband tunneling regime by having three minibands: a very narrow (9 meV) n=1 miniband, and much wider n=2 (44 meV) and n=3 (74 meV) minibands.

For each superlattice structure designed for transport measurements, a photoluminescence control wafer and an electrical device wafer were grown. The photoluminescence structures consist of undoped superlattices clad in AlGaAs. Photoluminescence of the nominally identical superlattices are used to verify the superlattice bandgap. All photoluminescence measurements hereafter quoted are for the undoped superlattice control samples, not the transport samples. Devices are fabricated using contact lithography, lift-off metallization processes, a conventional alloyed Ni/Ge/Au composite bottom contact, and wet chemical etching to form various size mesa devices, as small as $4 \mu m^2$. A SiN/polyimide passivation layer is deposited over the entire structure prior to fabrication of vias and contacts to the individual devices. For all of the results presented, positive bias will correspond to injection from the emitter (top contact) and negative bias will correspond to injection from the collector (substrate side).

4.3 Results

4.3.1 Narrow Miniband Samples

Sample 1 is a repeat of the structure of Davies, et al. [23]. The superlattice wells are 52Å in width, with the middle 17Å doped n-type to $4 \times 10^{17} \text{cm}^{-3}$. The superlattice barriers are 21Å wide, and the tunneling barrier separating the two superlattices is 55Å thick. We calculate that the first miniband lies 45 meV above the GaAs conduction band edge and extends for an additional 44 meV. The second miniband ranges from 204 meV to 403 meV above the GaAs conduction band edge, creating a minigap of 105 meV. The calculated superlattice Fermi level lies 10 meV above the first miniband edge. Low temperature photoluminescence data shows a superlattice bandgap of 1.580 eV, compared to a calculated bandgap of 1.578 eV.
Figure 4.3: Sample 1 current-voltage and conductance-voltage characteristics of a $4\mu$m$^2$ superlattice tunnel diode at $T=4K$ and 300K. Negative (positive) voltages correspond to injection from the surface (substrate) superlattice.

Devices $4\mu$m$^2$ in size are used for characterization of Sample 1. Our experimental zero bias device resistance and contact resistance at liquid helium temperatures are 5 k$\Omega$ and 31 $\Omega$, respectively. Figure 4.3 shows the current-voltage (I-V) and conductance-voltage (G-V) characteristics of this structure at 4K and 300K. We see a major low voltage resonance at $\pm 50$ mV, with less evident resonances observable in the derivative at about $\pm 400$ mV and $>500$mV. There is little change in the characteristics from 10K to 100K. From 100K to 300K the valley current increases monotonically. Our calculations show that the emitter and collector minibands no longer overlap at $\pm 50$ mV. For biases between the current peak
and valley (50mV < V < 150 mV), the emitter Fermi level is aligned with the first collector minigap. This indicates that the 50 mV resonance is the n=1 emitter to n=1 collector inter-miniband transition. Notice the finite zero bias conductance due to the equilibrium line-up of these minibands. The higher bias structure at about ±400 mV corresponds to the n=1 emitter to n=2 collector transition. This is in good agreement with the calculated value of 420 mV. The higher (>500 mV) structure is unidentified, being either injection into the indirect band minima or Gunn effect. There is little change in the low voltage peak position with increasing temperature. However the higher resonance shifts to lower bias, reflecting the increase of inelastic thermally generated background current. The low bias (n=1 emitter miniband to n=1 collector miniband tunneling transition) conductance peak only shifts up by a constant conductance with increasing temperature, indicating that thermally generated inelastic tunneling is the dominant leakage current. Compared to Davies, et. al.[23], the stronger 300K resonant structure here implies that material parameters are important in determining the magnitude of thermally generated inelastic tunneling, in this case about 500 nS/K above 100K.

We now investigate how these characteristics change as a function of the miniband position using Sample 2, which has a similar miniband width as Sample 1, and also has a “virtual” second miniband. Sample 2 is a ten period, 34Å well / 42Å barrier superlattice on either side of a 88Å tunnel barrier. The superlattice has a thin bound miniband, 33 meV wide, located 87 meV above the GaAs conduction band edge (versus 45 meV for Sample 1). The second miniband, a virtual band, lies from 263 meV to 454 meV, which is above the GaAs conduction band edge. The calculated Fermi energy of the superlattice at 4.2 K is 2 meV above the lower edge of the first miniband. Low temperature photoluminescence shows an experimental bandgap of the superlattice of 1.622 eV, in excellent agreement with the calculated bandgap of 1.622 eV. Figure 4.4 shows the I-V and G-V data of the Sample 2 superlattice tunnel diode structure at 173 K and 300 K. At low temperature (T < 150 K), the I-V is asymmetric in current, and the position of the n=1 emitter to n=1
Figure 4.4: Sample 2 current-voltage and conductance-voltage characteristics of a 9μm² superlattice tunnel diode at T=173K and 300K. Negative (positive) voltages correspond to injection from the surface (substrate) superlattice.
collector resonance for both the positive and negative voltages shifts to lower biases with an increase in temperature. The asymmetry in I-V characteristics at low temperature can be explained by carrier freeze out in the high resistance contacts (note the asymmetric epitaxial contact structure). Thus, here I concentrate on higher temperature data where this is not a contributing factor. Even at 173 K a slight asymmetry in the G-V around V=0 is observable. Variable temperature measurements show a forward peak current maximum at 187 K, a reverse at 110 K, and maximum peak to valley current ratios of 1.62 for forward bias and 2.34 for reverse bias occurring at the same temperature that the current peak occurs. This implies that the peak to valley ratio is dominated, even at relatively high temperature, by the elastic tunneling current. The valley current dependence does not reveal an activated process. The major resonances of the device occur at +196 mV and at -188 mV at T=173K. Higher bias data (> 1V) shows little structure at any temperature. Compared to Sample 1, there is a dramatic increase in the strength of the n=1 to n=1 tunneling resonance due to changing the position and width of the miniband. The 300K I-V of Sample 2 (with peak-to-valley (P/V) current ratio of 1.3 at 300K (2.4 at 4.2K)) is quite similar to the 4.2K I-V of Sample 1 (P/V of 2). The peak conductance is 100mS higher in Sample 1, consistent with a higher energy miniband. Aside from the major resonances (with the exception of an LO phonon emission shoulder present in Sample 1, to be discussed later), there is no apparent additional structure in the conductance greater than the 1mV experimental resolution for either Sample 1 or Sample 2, at a temperature of 4.2K.

4.3.2 Medium / Wide Miniband Sample

Now a single miniband sample with a medium and wide range miniband width superlattice will be examined. In addition, the contacts to the superlattices are altered to avoid the freeze out effects seen in Sample 2. Sample 3 is a structure with 41Å superlattice wells and 10Å superlattice barriers. The wells are doped n-type to 1x10¹⁷ cm⁻³. The structure has an 88Å tunneling barrier. The first miniband of Sample 3 is calculated to lie 35 meV above the
Figure 4.5: Sample 3 current-voltage and conductance-voltage characteristics of a 9µm² superlattice sample at T=4.2K. Negative (positive) voltages correspond to injection from the surface (substrate) superlattice. Note the fine structure in the conductance trace, due to the collector superlattice eigenstates.

Conduction band edge of GaAs and extends for 165 meV. The second miniband ranges from 294 to 810 meV above the GaAs conduction band edge. The calculated superlattice Fermi level lies 5 meV above the lower edge of the first superlattice miniband. Low temperature photoluminescence shows an experimental superlattice bandgap of 1.581 eV, compared to a calculated bandgap of 1.575 eV.

Figure 4.5 shows the low voltage I-V and G-V characteristics of Sample 3 at 4.2K. The ~ ±120mV major peak corresponds to the line-up of the first collector minigap with
the emitter Fermi level. A series of peaks on the low bias side of the major peak are apparent. Note that these biases correspond to electric fields well below that expected for Stark localization.[57,17] The condition for Stark localization of a superlattice is

\[ eEd > \Delta W, \]  

where \( E \) is the applied electric field, \( d \) is the superlattice period, and \( \Delta W \) the width of the miniband under consideration. At the biases considered here, the Stark splitting is less than 10meV, compared to a miniband width of 105 meV. The “sub-resonant series” starts to degrade above 20K, and is unobservable (except for the highest sub-resonance peak) above 50K.

The widest single miniband structure investigated (Sample 4) has a 36Å well and a 9Å barrier. The tunnel barrier of the structure is again 88Å and the superlattice wells were doped n-type to 1x10^{17} \text{ cm}^{-3}. The first miniband in this structure is calculated to extend for 214 meV, from 37 to 251 meV above the GaAs conduction band edge. The calculated superlattice Fermi energy lies in the first miniband, 55 meV above the GaAs conduction band edge. Low temperature photoluminescence gives an experimental bandgap of 1.575 eV, compared to a calculated bandgap of 1.578 eV.

Figure 4.6 shows the low temperature I-V and G-V characteristics of a 9\mu m^2 superlattice device. The major resonances of this device occur at +432 mV and -484 mV. In comparison to Sample 3, this sample exhibits a more pronounced sub-resonance series, as can be viewed better in the conductance derivative (Figure 4.7). The higher bias peaks of this series are evident even at room temperature.

There are two possible explanations for the structure: Stark localization of the miniband, as previously discussed, or the existence of individual quantum states within the miniband. In the present case, the maximum Stark splitting would be 30 meV, much lower than the miniband width of 214 meV. Thus the structure appears at too low a voltage to attribute the features to Stark localization of the miniband.
Figure 4.6: Sample 4 current-voltage and conductance-voltage characteristics at T=4.2K. Negative (positive) voltages correspond to injection from the surface (substrate) superlattice. Note the fine structure in the conductance trace, due to the collector superlattice eigenstates.
Figure 4.7: Sample 4 conductance-voltage and derivative conductance-voltage characteristics for the same sample and conditions as in 4.6.
Figure 4.8: Experimental (circle) and theoretical (cross) resonant crossings of the collector finite superlattice transmission peaks with the emitter superlattice Fermi level in Sample 4. The calculated resonant crossings were determined from mapping the finite superlattice transmission peaks onto the self-consistent band structure and determining the bias at which they cross the emitter Fermi level.

To determine if the structure is due to the finite extent of the superlattice, the single electron transmission coefficient of the 10-period superlattice/coupled quantum well system is calculated and then these 10 resonant peaks are mapped onto the self-consistent band structure. Figure 4.8 shows the calculated resonant crossings of the collector finite superlattice transmission peaks with the emitter Fermi level, compared with the experimental resonant peaks. The calibration of the top resonance is determined by the number of periods in the finite superlattice, and the low peak cut-off is determined from the superlattice Fermi level. The agreement between calculated and experimental peak position is qualitatively (a $V^{1/2}$ behavior) and quantitatively good. High voltage deviation may indicate that
Figure 4.9: Conductance-voltage and derivative conductance-voltage characteristics of Sample 5 (same as Sample 4 except one of the superlattices replaced with bulk GaAs) at T=10K. Positive bias corresponds to electron injection from the bulk GaAs into the finite superlattice. Note the absence of structure in the reverse bias.

A zero-current model is no longer valid. Sample 4 provides an example of a superlattice whose number of periods is sufficiently small that the effects of finite periodicity on the transmission coefficient can be seen in electron transport. To experimentally check that the resonances are indeed arising from the collector density of states, Sample 5 is examined, which is identical to Sample 4 but with bulk GaAs on one side of the superlattice. Figure 4.9 shows the G-V characteristics of this structure at 10K. Positive bias corresponds to electron injection from the bulk GaAs into the finite superlattice. Under this condition,
the position and number of the sub-resonance peaks compares well with that of the finite superlattice injector sample. As has been pointed out earlier, there is no structure in the reverse bias direction since the collector is bulk. It should be noted that the lower Fermi level in the bulk GaAs (versus the replaced superlattice) accounts for the voltage shift of the sub-resonant peaks. Sample 6 is used as another check to assure that the collector density of states is the source of the resonances seen in both Samples 4 and 5. As stated previously, this sample is identical to Sample 4, except that the number of superlattice periods on either side of the tunnel barrier is now 20 instead of 10. To first order, this should double the number of resonances seen in the electron spectra, as now the miniband is composed of 20 eigenstates, compared to 10 in Sample 4. Seen in figure 4.10 is evidence of approximately 18 resonances from 0 to 400 mV, in reasonable agreement with the expected observable 18-19 states (since 1 or 2 are hidden below the collector Fermi level).

The absence of structure in Samples 1 and 2, and the observation of structure in Samples 3 to 6, implies that the transition from an indistinguishable miniband to a coupled well structure has been experimentally observed. In energy, this implies the transition occurs between state splittings of 3.7 meV (the maximum in Sample 1) and 13.6 meV (the minimum observable in Sample 3), when kT < the state splitting E(i+1) - E(i). Note that this is a function of the position of eigenstate i within the miniband. The origin of the eigenstate inhomogeneous broadening mechanism (such as epitaxial or alloy fluctuations) is not known.

4.3.3 Multiple Miniband Sample

In previous samples, transport in only one miniband is investigated. The last superlattice structure investigated is a multi-miniband structure designed to show tunneling through a spectrum of higher order minibands. In order to increase the number of minibands under the AlGaAs conduction band edge, the superlattice well is widened. This structure has a 117Å well and a 27Å barrier. In order to compensate for the larger well, the resonant tunneling barrier is increased to 167Å. The calculated superlattice Fermi level lies 8 meV
Figure 4.10: Conductance-voltage and derivative conductance-voltage characteristics of Sample 6 (Sample 4 with 20 superlattice periods, instead of 10) at T=4.2K.
above the lower edge of the first miniband. Low temperature photoluminescence of the control superlattice shows a superlattice bandgap of 1.540 eV, compared to a calculated bandgap of 1.533 eV. Figure 4.11 shows a zero-bias Thomas-Fermi calculation[35] of the conduction band edge and minibands at low temperature for Sample 7. Three minibands are apparent. Using a more accurate fully self-consistent Poisson-Schrödinger calculation,[67] we calculate the positions of the minibands (referenced to the GaAs conduction band edge) to be: the first from 19 to 28 meV, the second from 77 to 111 meV, and the third from 176 to 250 meV. Figure 4.16 shows the device under a bias of 120 mV. In this sample, the Fermi level is calculated to lie at 27 meV above the GaAs conduction band edge, placing it near the top of the first, narrow (9 meV wide) miniband; thus, injection is occurring from near the top of the emitter miniband (e.g., from the Fermi level). The narrowness of the first emitter miniband also aids in the collimation of the electron energy distribution.

Figure 4.13 shows the experimental current and conductance versus voltage for Sample
Figure 4.12: Same as in figure 4.11 , with an applied bias of 120 mV. Note injection is now into the mini-gap between the collector n=2 and n=3 minibands.

7. The conductance peak near zero volts is due to transport from the n=1 emitter miniband into the n=1 collector miniband. The conductance peak near 100meV is due to transport into the n=2 collector miniband, and the peak near 200meV is due to transport into the n=3 collector miniband. The position of these current peaks, and the valleys between them, are verified using a self-consistent Thomas-Fermi model, evaluated at various device bias levels. Unlike Samples 3 through 6, fine structure is not as clearly visible in the conductance traces for 7, due to the smaller width of the minibands in Sample 7 which makes resolution of the eigenstates more difficult. Figure 4.14 shows two conductance derivative curves for Sample 7, taken at two different temperatures, 1.4K (solid) and 16.5K (dashed). Fine structure is apparent in both of these traces. Nominally identical traces are obtained after thermally cycling the device to room temperature. Many traces taken at temperatures between 16.5K and 1.2K show a gradual sharpening as temperature decreases, consistent with Fermi-level broadening in the emitter.
Figure 4.13: Sample 7 current-voltage and conductance-voltage characteristics of a 4μm² superlattice sample at T=4.2K. Positive voltages corresponds to injection from the surface superlattice. Note the small fine structure in the conductance trace due to the collector superlattice eigenstates.
Figure 4.14: Derivative conductance-voltage characteristics of Sample 7 at $T=1.4\,\text{K}$ (solid) and $T=16.5\,\text{K}$ (dashed).
In order to better understand the fine structure apparent in the conductance derivative data for Sample 7, a self-consistent Thomas-Fermi model is evaluated at several bias points to determine the conduction band profile. Knowing the conduction band profile, the eigenstate energies of the superlattice can be calculated, as in Figure 4.11. Figure 4.15 shows how the eigenstate energies of the collector superlattice move with bias, with respect to the Fermi level in the emitter. When these states pass through the emitter Fermi level, an increase in conduction is expected due to the enhanced probability of tunneling into one of the collector superlattice eigenstates. Note how the top-most states in the n=1 and n=2 minibands ‘peel off’ due to Stark localization of the state near the depletion layer next to the barrier. As is shown in figure 4.16, the Fermi level crossings with the superlattice states occur in regimes where there are no ‘peeled-off’ states, and these are thus not observable in the experiment. For the n=1 to n=1 transitions, where ΔW is 9 meV, it should be possible to observe individual states for T ≲ 10.4 K if the inhomogeneously broadened linewidth is less than 0.9 meV. For the n=1 to n=2 and n=1 to n=3 transitions, we expect that we should be able to clearly resolve effects due to the individual eigenstates, provided T ≲ 51 K (n=2) and 86 K (n=3) and the inhomogeneously broadened linewidth is less than 4.4 meV (n=2) or 7.4 meV (n=3).

Figure 4.17 shows the comparison of this modeling to five experimental conductance derivative curves (arbitrarily vertically offset) at temperatures from 1.2 K to 16.5 K. The two sets of dotted lines labeled n=2 and n=3 indicate where the peaks are expected from the modeling, and this qualitatively agrees where some structure occurs. The experimentally observed peak separation for the n=2 transitions are in excellent agreement with theory. For the n=2 labeled region, ΔV ≈ 4 mV (giving ΔE = 3.4 meV using a energy/voltage conversion ratio of 0.85, determined from the bandstructure modeling), as compared to a theoretical state separation of 3.4 meV. Note that the observed inhomogeneous broadened eigenstate linewidths are ~1.7 meV, probably due to epitaxial structural imperfections and/or impurities, implying that the n=1 states overlap and are thus not observable.
Figure 4.15: Calculated collector superlattice eigenstate energies as a function of applied voltage. Note how the top eigenstate in the $n=1$ and $n=2$ miniband peel off and enter the $n=2$ and $n=3$ minibands respectively.
Figure 4.16: Same as in figure 4.15, with an applied bias of 120 mV. Note the injection into the mini-gap between the collector \(n=2\) and \(n=3\) minibands.
Figure 4.17: Derivative conductance-voltage characteristics of Sample 7 at 1.4K, 3.0K, 6.0K, 10.0K, and 16.5K. The shaded region indicates where the n=1 emitter states intersect the collector superlattice n=1 states. The two series of dashed lines (n=2 and n=3) indicate where the theory predicts the occurrence of conductance resonances, due to the eigenstates in the collector superlattice. The two brackets labeled “n = 2 + 1 LO” show where features due to GaAs and AlAs LO phonon assisted tunneling events would occur.
Additional peaks are observed between the expected \( n=2 \) and \( n=3 \) resonances. The similar temperature dependence of these additional peaks with those of the \( n=2 \) and \( n=3 \) peaks imply that these are also individual state peaks. Up until now, transitions involving an LO phonon emission have not been considered. Using phonon energies of 36 meV for GaAs and 48 meV for AlAs, we expect additional transitions to lie at biases that are an LO phonon energy higher, after taking into account the energy/voltage conversion ratio. Since we have both GaAs and AlAs modes, we generate two additional bands of transitions which are calculated to lie between the \( n=2 \) and \( n=3 \) main peaks. The brackets labeled \( n=2 + 1 \) LO and \( 2 + 1 \) LO indicate where features would arise from these processes. The bands span the entire \( n=2 \) to \( n=3 \) gap (no phonons), accounting for the additional observed structure. The \( n=1 \) phonon replicas lie close to the \( n=2 \) no phonon resonances, and thus may account for them not being observable. The strength of these phonon replicas is not well understood. The only other sample in this study that shows an LO phonon replica is Sample 1 (only visible when magnifying the region near 100 mV in Figure 4.3, see figure 4.18), however in this case, as expected, no discrete structure is seen. For the other samples studied, the bias voltage to enter the LO phonon region is either too high (onset of noisy conduction at high bias) or the LO phonon region significantly overlapped the miniband region, which presumably has a stronger effect on the tunneling spectroscopy.

4.4 Conclusions

The transition in a GaAs/AlGaAs superlattice from a continuous narrow miniband to a larger miniband has been observed, where the finite period nature of the superlattice becomes important. Structure in tunneling spectroscopy for finite period single and multiple mini-band superlattice / barrier / superlattice samples that is attributed to the discrete eigenstates which form the miniband was also evident. Variable temperature data shows thermal broadening of the conductance structure due to changing the energy distribution.
Figure 4.18: Conductance voltage and derivative conductance-voltage characteristics of Sample 1 at T = 4.2K, showing the LO phonon assisted tunneling, near V = 100 mV.
of the electrons near the Fermi level in the emitter, as expected. The features in the conductance spectra were determined to arise from the collector density of states by both examining an asymmetric structure with no superlattice on one side of the barrier, and by varying the superlattice periodicity. The experimental electron spectroscopy results agree well with self-consistent Thomas-Fermi calculations. Evidence for GaAs and AlAs LO intra-miniband phonon-assisted (emission) transitions is present in the spectroscopy.
Chapter 5

Transport in Vertical InGaAs Quantum Dots

5.1 Introduction

Tunneling in low dimensional semiconductor structures has been a very active research field, both experimentally [102, 94, 81, 83, 24, 98, 97, 73, 71, 60, 80] and theoretically,[66, 64, 14, 106, 5, 72, 46, 18, 56] Electron confinement is usually achieved either through fabrication imposed confinement of a double-barrier resonant-tunneling structure (DBRTS) or through electrostatic confinement of a 2D electron layer. Both techniques create a potential which spatially localizes an electron in a region, a dot, and quantize the allowed energy levels in this region. These two techniques also allow for the coupling of electrodes to the sample. One electrode, the emitter, probes the density of quasi-OD states by injecting electrons into the dot. The other electrode, the collector, allows injected electrons to leave the dot.

In the case of 2D electron layers that are electrostatically confined, the single-electron charging energy, $U_C$, is usually much greater than $\delta E$, the spacing between single-particle states in the dot. In the case of a double barrier semiconductor heterostructure, $\delta E$ and $U_C$ can be of the same order,[94] or $U_C$ can be less than $\delta E$. In the regime where they are
on the same order, it is difficult to distinguish between the transport phenomena caused by each of these effects[94, 46] and proper modeling[5] is required to appropriately assign the observed structure to either spatial quantization or single electron charging.

Much of the prior work in double barrier structures utilizes a GaAs well, between two AlGaAs barriers to form the dot.[102, 94, 98, 97, 80] Typically, the well and barriers are undoped, and have an undoped spacer layer (~100 Å), to prevent the diffusion of dopants into the AlGaAs barriers during growth, situated immediately prior to the barriers. Dopants in AlGaAs are known to form deep donor levels (DX centers), and it is preferable to avoid complicating the electron spectroscopy with events due to these levels.[74] These spacer layers are usually contacted by highly doped GaAs. This results in a conduction band profile for the bulk epitaxial material as shown in figure 5.1a. The first quantized state is relatively far away from the Fermi level in the emitter, due to band-bending in the spacer layer region (lateral quantization pulls the dot states up even further). In this example, biases on the order of hundreds of millivolts are needed to pull these states down and to inject electrons into the dot. By using an In$_x$Ga$_{1-x}$As well, the quantum states in the bulk may be brought down to, or below, the Fermi level (figure 5.1b). There are a few advantages to this structure. First, by bringing the quantum levels further down in the well, their lifetime is increased, and thus their intrinsic broadness in energy is reduced. Also, there is far less distortion of the emitter/dot/collector potential needed to examine the density of states of the dot, as the dot states may be brought into resonance with less applied bias, compared to systems using GaAs wells.[102,94,98,97,80] Additionally, due to the lower applied bias required, there is far less power dissipation and local electron heating in the dot region, allowing for measurements of the system closer to equilibrium.
Figure 5.1: Conduction band diagram in tunneling direction for (a) a GaAs quantum well (unconfined laterally) and the corresponding diagram for (b) an In$_{0.1}$GaAs$_{0.9}$ quantum well. The shaded regions correspond to occupied electron states below the Fermi level.
5.2 Sample Growth and Fabrication

Samples are grown using molecular beam epitaxy on a Si-doped (100) GaAs substrate. The dot region consists of a 50Å In_{0.1}Ga_{0.9}As quantum well, enclosed by a pair of 40Å thick Al_{0.25}Ga_{0.75}As barriers. This region, along with 100Å spacer layers of GaAs that contact the barriers, is undoped. The spacer layers (which serve to prevent the diffusion of dopants into the Al_{0.25}Ga_{0.75}As barriers) are contacted by GaAs doped with Si at a density of $3 \times 10^{18}$ cm$^{-3}$.

Small (~ 100nm) AuGe/Ni/Au ohmic top-contact dots are defined by electron-beam lithography on the surface of the grown resonant tunneling structure. A bilayer polymethylmethacrylate (PMMA) resist and lift-off method is used. The metal dot ohmic contact serves as a self-aligned etch mask for highly anisotropic reactive ion etching (RIE) using BCl$_3$ as an etch gas. The resonant tunneling structure is etched through to the bottom n$^+$ GaAs contact. The resulting pillars, which contain the quantum dots, are shown in figure 5.2. The smaller pillars are approximately 1000Å wide and are standing adjacent to 1 μm wide pillars. Contact to the top of the structures is achieved through a planarizing/etch-back process employing polyimide and an O$_2$ RIE.[81] A gold contact pad is then evaporated over the columns. Bottom contact is achieved through the conductive substrate. Sample growth and fabrication was performed at the Central Research Laboratory of Texas Instruments.
Figure 5.2: An SEM showing the free standing pillars containing the quantum dots, prior to polyimide planarization and top contact. The thinner pillars are approximately 1000Å in diameter. SEM courtesy of J.N. Randall, Central Research Laboratories, Texas Instruments.
5.3 Theory I: Analytic Results

Consider a quantum well which has been quantized in the vertical dimension through epitaxial growth. In the lateral dimension through fabrication, we can create a system that is a quasi 0D dot connected to the outside world by two 1D leads. The vertical confinement in such a structure is modeled by a rectangular potential. The fabrication imposed lateral confinement, which is enhanced due to Fermi level pinning on the side walls,[94,81] may be modeled in a few different ways.

5.3.1 Lateral Quantization

Typically the lateral potential is modeled by a parabolic potential, as the incorporation of magnetic field into the Hamiltonian of this system can be accomplished analytically[81,94, 102]. For the more general case, we can write the potential as a parabolic potential at the sidewalls with a flat bottom in the center,

\[ \Phi(r) = \Phi_T [1 - (R - r)/W]^2 \text{ for } r > R - W; \quad \Phi_T(r) = 0 \text{ for } r < R - W, \]  \hspace{1cm} (5.1)

where \( \Phi_T \) is the Fermi-level pinning energy at the sidewalls, \( R \) is the lateral physical dimension, \( r \) is the radial coordinate, and \( W \) is the depletion depth. Figure 5.3 shows a schematic of the vertical potential (left) (similar to figure 5.1b) formed by the epitaxial structure and the lateral potential (bottom) formed by Fermi level mid-gap pinning at the surface. \( E_{C,\Gamma} \) denotes the \( \Gamma\)-point conduction band energy.

Schrödinger’s equation for such a structure is best expressed in cylindrical coordinates. Due to rotational symmetry, there is no \( \theta \) dependence, and since \( \Psi(r, z) \) can be written as \( \psi(r) \psi(z) \), separation of variables is possible. The single electron eigenstates, \( E_N \), are composed of two parts,

\[ E_N = E_z + E_{n,l}. \]  \hspace{1cm} (5.2)

The values of \( E_z \) (\( z = 0, 1, 2, 3... \)) result from the vertical quantization, and for the experimental electron spectroscopy of a DBRTS, this will almost always be \( E_0 \), the quantum
Figure 5.3: Schematic illustration of the vertical (a-b) and lateral (c-d) potentials of a vertical column containing a quantum dot. (adapted from reference [81])
well ground state in the vertical direction. The $E_{n,l}$ are the eigenstates that result from the lateral potential. For a parabolic model,

$$E_{n,l} = (2n + |l| + 1)\hbar \omega_0$$

(5.3)

where the radial quantum numbers $n = 0, 1, 2, \ldots$, the azimuthal quantum numbers $l = 0, \pm 1, \pm 2, \ldots$, and $\hbar \omega_0$ is the energy state spacing in the parabolic well. If we assume that we are in the first vertical subband, $E_0$, and that the lateral dimension is reduced to $2W$ or less (i.e., the potential is purely parabolic, no flat bottom), then the energy spacing between states is

$$\Delta E = \hbar \omega = \left(\frac{2\Phi_T}{m^*}\right)^{1/2} \frac{\hbar}{R}.$$  (5.4)

For a GaAs well, with AlGaAs barriers, typical values for energy state separation are on the order of tens of meV, for $\Phi_T = 0.8$ eV and $R \approx 500 \text{Å}$.

### 5.3.2 Magnetic Field

The next obvious question is that of how these localized states depend upon magnetic field. An approach that can be employed when the observed magnetic shifts are small compared to the lateral state energy spacings, is to use first order perturbation theory. There are two important field directions to consider: field parallel to the current direction and field perpendicular to the current direction.

**Field Parallel to Current**

For this orientation, the Hamiltonian can be written as

$$H = (\mathbf{p} + e\mathbf{A})^2/2m^* + V_{lateral}(r, z, \theta).$$

(5.5)

Picking a vector potential $\mathbf{A} = (1/2)B_0r\hat{\theta}$, the shift of the quantum dot states in field, $\delta E_{B||}$, is given by

$$\delta E_{B||} = \left(\frac{e\hbar}{2m^*}\right)\ell B + \frac{e^2B^2}{8m^*} <r_0^2>.$$  (5.6)
Note that spin is neglected in equation 5.6, as the spin splitting energy is $g\hbar B/2m$ (note $m$, not $m^*$), which is only 0.25 meV at 10 Tesla. The shift for a localized level with $\ell = 0$ is *entirely* due to the diamagnetic term,

$$\delta E_{\text{diamagnetic}} = \frac{e^2 B^2 \langle r_0^2 \rangle}{8m^*}$$

where $\langle r_0^2 \rangle$ is the spatial extent of the localized wavefunction. Note that if a diamagnetic shift is observed in the experimental data, the only unknown in equation 5.7 is $\langle r_0^2 \rangle$. Therefore, by the experimental diamagnetic shift is a *direct measure* of $\langle r_0^2 \rangle$.

$\langle r_0^2 \rangle$ can also be calculated theoretically, by assuming a form for the lateral potential and resultant wavefunctions. For a given choice of wavefunctions, $\psi(r)$,

$$\langle r_0^2 \rangle = \frac{\int \psi(r) r^2 \psi^*(r) r \, dr}{\int \psi(r) \psi^*(r) r \, dr}. \quad (5.8)$$

This result only assumes that the solutions in $r$, $z$, and $\theta$ are separable. Therefore, the choice of potential is only limited to those which are separable. There are two extremes for the potential used in analytically determining the wavefunctions. The first is the hardwall form (i.e., $V = \infty$ at the edge of the dot, zero inside the dot), which results in Bessel wave functions. The other choice is a radially symmetric parabolic well, which results in hypergeometric wave functions. In the hardwall case, the size of the potential is the free parameter that may be adjusted to match the experimentally observed diamagnetic shift. In this case, the size of the potential will also correspond to the physical dot size, minus depletion effects. As the diamagnetic shift measures the spatial extent of the state wave function, it can also describe the shape of the lateral potential by examining the shifts of a series of higher order states. In principal, this problem must be solved self-consistently in order for the potential shape and resultant wavefunctions to match the observed diamagnetic shifts.
Field Perpendicular to Current

For a field perpendicular to the current \((B = B_0 \hat{x})\), it is easier to express the Hamiltonian in rectangular coordinates,

\[
H = (p + eA)^2 / 2m^* + V_{\text{lat}}(x, y, z),
\]

with the vector potential \(A = (0, -zB_0, 0)\) and the three-dimensional confining potential of the quantum dot \(V_{\text{con}}(x, y, z)\). The first order perturbation term in this case is more difficult to evaluate, due to the \(y\) and \(z\) coupling involved. Numerical results indicate an increase in dot energy states by a diamagnetic shift term, which is predicted to be less than 0.5 meV at 10 Tesla for a GaAs/AlGaAs quantum dot system with the same well width (50 Å) as in this study. The small value for the shift in this field orientation is due to the interaction of a relatively weak magnetic potential in comparison to the strong confinement in the \(z\) direction.

5.3.3 Current versus Voltage Behavior

The vertical quantum dot structures are typically electrically probed using a two terminal current versus voltage measurement. Theoretically, the current versus applied bias for such a structure is described by

\[
I(V) = e \sum_n \int_{E_{F} - eV}^{E_{F}} N_n(E) v_n(E) T_n(E, V) dE,
\]

where the sum is taken over all 1D subbands, \(n\), below the Fermi energy \(E_F\) in the emitter. \(N_n(E) \propto E^{-1/2}\) is the 1D density of states of the \(n\)th subband in the emitter. \(v_n(E) \propto E^{1/2}\) is the velocity of electrons at energy \(E\) in subband \(n\), and \(T_n(E, V)\) is the transmission probability through the double-barrier structure. For the special case of 1D subbands, the product \(N_n(E)v_n(E) = 1/h\) is constant. Assuming that \(T_n(E, V)\) near resonance is approximated by a Lorentzian line shape, with linewidth \(\Gamma_e\) and peak transmission of \(T_{0,n}(V)\), a current contribution of

\[
\Delta I_n = e \pi T_{0,n}(V) \Gamma_e / h
\]
is obtained for each spin state in every occupied 1D emitter subband.[102] Thus, for low bias where \( T_{0,n} \) is not a strong function of bias, we expect to see a sharp staircase structure in the \( I(V) \), with a step for every \( \Delta I_n \).

In the presence of inelastic scattering processes, the line shape due to tunneling into a 0D state is broadened. The transmission coefficient then may be written as

\[
T_n(E) = \frac{T_{0,n}}{1 + \{2(E - E_n)/\Gamma\}^2},
\]

where \( E_n \) is the energy of a 0D dot state, and \( \Gamma \) is the total line width which is composed of two parts, \( \Gamma = \Gamma_e + \Gamma_s \).[93] The intrinsic state broadening is given by \( \Gamma_e = \hbar/\tau_e \), where \( \tau_e \) is the lifetime of the quasi-bound quantum state. \( \Gamma_s = \hbar/\tau_s \) is related to the inelastic scattering time, \( \tau_s \). \( \Gamma_s \) is the dominant broadening effect. Scattering is usually attributed to either electron relaxation in the dot (by scattering into other subbands) or to phonon emission.[101]

### 5.3.4 Charging Effects

Until now, electron-electron interactions have been neglected. Charging effects are important as the lateral device area is scaled down. Charging can be introduced into the usual single electron model, by modeling the Coulomb charging energy of quantum dot as arising from a single effective capacitance \( C \).[102] This approach uses the semi-classical geometric capacitance (the sum of an emitter capacitance \( C_e \) and a collector capacitance \( C_c \) in series),

\[
C = C_e + C_c \approx \frac{\epsilon_0 \pi a^2}{4} \left( d_e^{-1} + d_c^{-1} \right),
\]

where \( d_e \) and \( d_c \) are the thickness of the emitter and collector barriers, respectively. For the structures in this paper, \( C \approx 2.6 \times 10^{-16} \text{F} \) using \( \epsilon_{AlGaAs} \approx 11.6 \), and \( a \approx 800 \text{Å} \). The value \( a \) is the electrical diameter which is the extrapolated value determined from current density measurements on large area samples. In this case, the estimated charging energy, \( E_C \), where

\[
E_C = \frac{e^2}{2C},
\]
is approximately 0.31 meV. The simplest inclusion of charging effects is to add in a charging energy term (as in equation 5.14) to equation 5.2.

Figure 5.4 illustrates the qualitative low bias I(V) curve expected from theory. For the pure quantization ($E_c = 0$), a series of spin degenerate plateaus is expected from equation 5.11. If the quantization and charging energies are made comparable, the spin degeneracy is lifted, and more steps will be observed. Also, for this case the spacing between steps becomes more complicated, as both charging and quantization energies contribute to the resonance bias for a dot conduction channel.

We now combine all of the effects discussed (i.e., quantization, magnetic field, and charging) to determine the overall dependence of the resonance bias. In order to convert between device bias and energy, $\alpha$ is defined to be the energy to bias (meV/mV) conversion constant. The resonant voltage at which the steps in current occur is given as a function of the total number of electrons residing on the dot, $N$, and the magnetic field, $B$.

$$V_{res}(n,I,B,N) \approx \left( \frac{1}{\alpha e} \right) \{E_z + E(B) + \left[ \frac{e^2}{C} \left( N - \frac{1}{2} \right) \right] - E_F \} \quad (5.15)$$

The factor $E(B)$ is the dependence of the quantized states on magnetic field. $E_{C,N} = e^2/C(N - \frac{1}{2})$ gives the Coulomb charging energy for $N$ electrons on the dot.[102,43]

5.4 Theory II: Numerical Results

Section 5.3 gave analytic expressions for the behavior of the electron eigenstates in the 0D dot. While many of these are very useful to first order in determining expected diamagnetic shifts and state spacings, the treatment of the lateral potential as either a parabolic or hardwall potential is an oversimplification of the problem. In order to treat the real lateral potential, two methods (1D and fully 3D) have been pursued to more accurately determine

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Note that in Tewordt’s expression for equation 5.13 [102], a factor of $4\pi$ is incorrectly placed in the denominator. The $4\pi$ only belongs in the CGS unit expression for capacitance between two parallel plates. Tewordt also uses a value for $E_c$ as $e^2/C$, whereas it should be $e^2/2C$. This reduces Tewordt’s predicted charging energy for his structures substantially from 6.25 meV to 0.25 meV.
Figure 5.4: Qualitative I(V) expected for a vertical quantum dot at low applied bias. The left staircase is for the case where charging energy is negligible, the right staircase represents the case where the charging and quantization energies are comparable. The numbers near the steps indicate the dot channel through which conduction is proceeding.
the expected level spacings and diamagnetic shifts.

5.4.1 1D Results

The 1D results for the lateral potential were obtained using a modified version of BandProf, a heterostructure modeling program that is capable of self-consistently solving the Poisson equation.\(^2\) Figure 5.5 shows, using a 1D calculation, the states in a 800Å wide well with sidewalls that are parabolically pinned to 800 meV. Quantum states are indicated by dashed lines in the figure. The state spacing in energy is not constant, as is especially evident in the lower lying states in figure 5.5b. Figure 5.6 shows the lateral state energy versus state index. For a purely parabolic potential, this would be a straight line. However, this line clearly has curvature. Taking the derivative (as shown in the dashed curve for figure 5.6) gives the state separation in energy as a function of state index. Note that each of these states is spin degenerate, and also degenerate by an additional factor of two if a 2D potential is assumed.

5.4.2 3D Modeling

The complete 3D numerical modeling of the dot structure is achieved in a manner similar to that of the 1D calculation. Starting from a physical description of the structure, the Poisson equation is self-consistently solved, determining the potential diagram for the dot.\(^1\) Figure 5.7 shows a 3D plot of \(V(r, z)\) from a calculation for the InGaAs dot structure, using a physical radius of 60 nm, which is close to the physical radius of a typical dot structure. In this figure, the axis labeled ‘\(\rho\)’ corresponds to \(r\). Note the Fermi level pinning at \(\rho = 60\) nm. Wavefunctions, energy eigenstates, and their dependence upon magnetic field can also be calculated.

\(^2\)The lateral potential was modeled in BandProf by using an AlAs/InAs conduction band edge. In order for the state calculation to be correct, the electron effective masses for AlAs and InAs were set to be that of In\(_{0.1}\)Ga\(_{0.9}\)As.
Figure 5.5: (a) 1D calculation of the lateral potential and quantum levels of an In$_{0.10}$Ga$_{0.90}$As quantum dot. (b) Vertical expansion near the bottom of the potential. The number of dashes in the states corresponds to the number of nodes in the wavefunction.
Figure 5.6: Lateral state energy versus state index, and the lateral state energy separation as a function of state index from the potential and states shown in figure 5.5.
Figure 5.7: A plot of the potential in a InGaAs quantum dot for $R = 60$ nm.
5.5 Experimental Results

Several different types of transport measurements were performed to investigate the electronic properties of the vertical InGaAs quantum dot structures. First, large area samples (i.e., no lateral confinement) were examined to confirm the epitaxial (z) potential and to determine current densities in the bulk material. Following this, small area dot structures were measured at low temperatures. Variable temperature data, as well as a variety of magnetotunneling measurements will be presented.

5.5.1 Large Area Samples

Large area resonant tunneling diodes (RTDs) are fabricated to examine the peak positions and current densities for the epitaxial material. Figure 5.8 shows a current versus voltage trace for a 1024 $\mu$m$^2$ sample measured at 4K. The very symmetric response is consistent with the symmetric epitaxial structure. The negative differential resistance (NDR) occurs at $\pm 23$ mV with a peak current density of 56 A/cm$^2$. The conductance is 25 mS for the 1024 $\mu$m$^2$ device, and this value is observed to scale linearly with device area, as expected, providing a means of extrapolating the upper limits for the electrical sizes of the smaller devices.

5.5.2 Zero Magnetic Field Measurements: Small Area Samples

The small area quantum dot samples are measured at milliKelvin temperatures in a dilution refrigerator. Figure 5.9 shows a current versus voltage I(V) curve for a sample ($R \approx 100$ nm) under zero magnetic field, at a mixing chamber temperature of 50 mK. Figure 5.10 shows a close-up near the zero bias region. The conductance for this sample ($\sim 0.4 \mu$S) yields a value of approximately 800Å for the electrical diameter, $a (= 2r$, see figure 5.3).

Step-like staircase structure in current is observed in figures 5.9 and 5.10 in both bias directions, especially at low biases. Note the qualitative agreement between this and what
Figure 5.8: Current versus voltage trace for a 32\( \mu m^2 \) sample fabricated from the same epitaxial material used for the quantum dots. T = 4.2K.
Figure 5.9: Current versus voltage characteristic for an In$_{0.1}$Ga$_{0.9}$As quantum dot. Note the step-like structure in current near zero bias. $T_{\text{mix}} = 50\text{mK}$. 
Figure 5.10: A closeup of the low bias regime of figure 5.9.
is theoretically expected (figure 5.4). Similar structure in the low bias regime for DBRTS has been previously reported by other groups. [94, 101] The major difference in our work is the use of an InGaAs dot, as well as the use of a symmetric epitaxial structure. As expected from the theory (section 5.3.3), the steps are relatively flat at low bias, when the transmission coefficient, $T_{0,n}$, (equation 5.11) is not a strong function of applied bias.

In order to analyze the spectroscopy in greater detail, we need to determine the bias to energy conversion factor, $\alpha$. The parameter $\alpha$ may be determined from the temperature dependence of the plateau edges, which is the focus of the next section.

### 5.5.3 Variable Temperature Measurements

The sharpness of the plateau edges are expected to depend upon temperature, as for higher temperature, the Fermi distribution in the emitter broadens. The current versus voltage characteristics as a function of temperature, are shown in figures 5.11 and 5.13. Figures 5.12 and 5.14 show corresponding conductance (dI/dV) versus voltage data. As expected, the plateau edges (the sharp peaks in conductance at approximately 7.5 mV and 17.5 mV in the reverse bias and 18.5 mV and 30.0 mV in the forward bias) are very sensitive to changes in temperature.

As previously mentioned, the voltage to energy conversion factor, $\alpha$, is calculated from fitting the Fermi function $[I(V, T) = I_0 f(\alpha V/kT)]$ to the first current plateau ($I_0$ is the value of the current on the plateau) in the variable temperature data.

$$I(V, T) = \frac{I_0}{\{1 + \exp[-\alpha e(V - V_{th})/(kT)]\}} \quad (5.16)$$

$V_{th}$ is threshold voltage for the current plateau and can be accurately determined from the intersection point of the $I(V, T)$ curves in figures 5.11 and 5.13 as the current given by equation 5.16 does not depend upon temperature for $V = V_{th}$. This value for $\alpha$ is determined to be 0.37 meV/mV in the forward bias direction and 0.50 meV/mV in the reverse bias direction. This implies an asymmetry in the emitter and collector (either in
Figure 5.11: Reverse bias current versus voltage for a quantum dot at 35mK, 2.0K, 4.0K, 5.0K, 7.5K, 10.0K, and 14.0K.
Figure 5.12: Reverse bias conductance versus voltage for a quantum dot at 35mK, 2.0K, 4.0K, 5.0K, 7.5K, 10.0K, and 14.0K. The inset shows an expansion of the sub-threshold peaks that arise as temperature is increased.
Figure 5.13: Forward bias current versus voltage for a quantum dot at 35mK, 2.0K, 4.0K, 5.0K, 7.5K, 10.0K, and 14.0K.
Figure 5.14: Forward bias conductance versus voltage for a quantum dot at 35 mK, 1.2K, 2.0K, 4.0K, 5.0K, 10.0K, and 14.0K. The inset shows an expansion of the sub-threshold conductance plateau that arises as temperature is increased.
doping, or in the barrier or spacer layer thickness) in contrast to the symmetric response seen in the large area device (figure 5.8). This is not surprising as we are now probing a very localized region instead of averaging over non-uniformities in barriers or series doping, as in the large area device.

It should also be noted that only temperatures greater than 1 K were used for this fit, since below 1 K, the line shape can reach either the intrinsic linewidth that is limited by inelastic scattering[101] or the temperature limit for cooling the electrons in the dilution refrigerator. The electron temperature in the sample is often not the same as that of the mixing chamber, or of the sample thermometer, at milliKelvin temperatures. In both the reverse and forward bias directions, the sample’s electrons appear to stop cooling at 100-200 mK. This is consistent with other samples measured in the same experimental system. This places an upper limit on the intrinsic state linewidth ($\Gamma$ at $T = 0$) of approximately $1.9 \times 10^{-11}$ seconds.

As previously mentioned, the current value at the intersection point ($V = V_{th}$) for the different temperature $I(V)$ curves in figures 5.11 and 5.13 should be $I_0/2$ (see equation 5.16). However, in both figures 5.11 and 5.13, the average plateau current is only about 75% of the expected value, $I_0$. The assumption that the plateau current, $I_0$, will be twice the current at $V = V_{th}$ assumes a constant density of states in the 1D emitter as the bias is increased. If we are near a 1D subband edge at $V = V_{th}$, as the bias is increased the density of states, and current will decrease. It is also difficult to determine what the plateau current is in the presence of the fine structure which modulates the current.

**Fine Structure on Plateaus: Using the 0D Dot States to Probe the Emitter**

Another effect which is observed in current and conductance in figures 5.11, 5.12, 5.13, and 5.14 is the presence of fine structure peaks on top of the current plateaus. These fine structure peaks exhibit less of a dependence upon temperature, especially those located far away from the plateau edges (note the conductance peak at 23 mV in the reverse bias and
at 12 mV in the forward bias). This relative temperature insensitivity indicates tunneling from the electrons below the emitter Fermi level, where the emitter state occupation is not a strong function of temperature. Therefore, this weaker structure is attributed to emitter states below the Fermi-level, which pass into and out of resonance with the narrow 0D dot levels, as the applied bias is varied. If this model is correct, we expect that as the sample temperature is increased, states above the Fermi level could be populated.

**Thermally Activated Emitter State Tunneling**

Figure 5.15 schematically shows how the occupation of discrete emitter levels (depicted as black lines in (a) and (b)) is affected by increasing temperature. At very low temperatures, only states in the emitter at or below the Fermi level, $E_F$, are occupied. These are the only emitter states available for tunneling. Electrons in the states nearest to the Fermi level will tunnel into the dot state first. States below the Fermi level then contribute to the tunneling as the bias is increased.

At finite temperature, occupation of the states above the Fermi level becomes more probable. Contributions of emitter states above the Fermi level become important, since the product of level occupation and tunneling probability for these states will become comparable to that of the states near the Fermi level. Due to the occupation of states above the Fermi level as $kT$ is increased, resonances below (in voltage) the first plateau may be observed. The insets of figures 5.12 and 5.14 show that this is observed. As expected, the conductance peak in the inset of figure 5.12 rises with temperature as the state occupation increases.

The pre-resonant structure shown in the inset of figure 5.12 may be fitted to the derivative of equation 5.16 (since $I(V) \propto f(E)$, the Fermi function, then the conductance, $G(V) \propto df(E)/dE$). Figure 5.16 shows the experimental and theoretical conductance for the pre-resonant conductance peak as a function of temperature. The theoretical conductance curve was determined assuming that an emitter state 1.75 meV above the Fermi level
Figure 5.15: (a) At T=0, the Fermi function, f(E), is a step function (black line) which results in a very sharp transition from occupied to unoccupied discrete emitter states. (b) At finite temperature, f(E) becomes rounded near the Fermi energy, $E_F$, causing a gradual transition from states that are mostly occupied to states that are mostly unoccupied.
becomes thermally activated at higher temperatures. It should be noted that the only fitting parameter other than the preresonant peak location is $I_0$, which was within 5% of the value determined from the $I_0/2$ intersection at $V = V_{th}$ for the first current plateau in the reverse bias in figure 5.11. Since $I_0$ depends upon $T(E)$, this indicates that the transmission probability for a transition from the first emitter state lying above the Fermi level to the ground state in the dot is approximately the same as the transmission probability for the emitter states near the Fermi level, as we expect.

**Variable Temperature Results Summary**

From the variable temperature results, the broadening of the plateau edges is observed and found to be consistent with the expected Fermi level broadening in the emitter. From this dependence, the bias to energy conversion factor, $\alpha$, is determined, allowing for spectroscopic interpretation of the plateau widths and positions in terms of energy. Additionally, fine structure on top of the plateaus was observed. This fine structure depends more weakly upon temperature than the plateau edges and is strong evidence for this structure originating from discrete states in the emitter below the Fermi level. This interpretation also implies that thermally activated fine structure below the plateau threshold bias should also be apparent. This is observed in the data and in excellent agreement with the theory.
Figure 5.16: Experimental conductance versus temperature (for the peak in the inset of figure 5.12) and the predicted theoretical conductance.
5.5.4 Magnetotunneling Measurements

A variety of magnetotunneling measurements were performed. They can be divided into two categories. The first, which is discussed in this section, are measurements which include a large bias range (similar to that in figure 5.9), over relatively large field steps (approximately 0.2 Tesla) from 0 to 9 Tesla. Both bias directions were covered, in magnetic field both parallel and perpendicular to the current (i.e., perpendicular and parallel to the quantum well respectively). In order to better characterize the magnetoconductance of the fine structure on the plateaus, a reduced bias region (~ ±45mV) that contains the first few steps (similar to that shown in figure 5.10) was examined with higher current sensitivity and smaller field steps (approximately 0.02 Tesla) for both bias directions and field orientations. This data is discussed in section 5.5.6.

Wide Bias Magnetoconductance in Parallel Field

In magnetic field parallel to current, diamagnetic shifts of the dot ground state levels are expected (see section 5.3.2). Figures 5.17 and 5.18 show the reverse and forward current-voltage characteristics versus magnetic field parallel to current, ranging from 0 to 9 Tesla. The mixing chamber temperature ($T_{mix}$) is 30 mK and the step in magnetic field between traces is 1875 Gauss. The traces are offset by a constant current value for clarity.

In magnetic field parallel to the current, there is a diamagnetic movement of all steps to higher bias with field (this can be better observed in the fan diagrams in figures 5.30, 5.31, 5.32, and 5.33). At 9 Tesla, the shift is approximately $+1.4-2.6$ meV ($+2-4$ mV), depending on the step. Table 5.1 lists the values for the diamagnetic shifts and current plateau widths for the first six steps. Figure 5.19 shows the threshold voltage for the first plateau in reverse bias versus magnetic field squared. Note the excellent agreement to the theoretically expected diamagnetic $B^2$ behavior (see equation 5.7). Accounting for $\alpha$, this slope yields a wavefunction of radial extent of approximately 100Å for the ground state in both bias directions. The implications of this value for the radial wavefunction extent upon
Figure 5.17: Current-voltage characteristics versus magnetic field parallel (0-9 Tesla in 0.1825 Tesla steps) to the current for the reverse bias direction.
Figure 5.18: Current-voltage characteristics versus magnetic field parallel (0-9 Tesla in 0.1825 Tesla steps) to the current for the forward bias direction.
Table 5.1: Experimentally observed diamagnetic shifts and current plateau widths.

<table>
<thead>
<tr>
<th>Plateau Index (N)</th>
<th>$\Delta E_{\text{diamagnetic}}$ (meV, $B=9$ Tesla) (reverse, forward bias)</th>
<th>Plateau Width (meV) (reverse, forward bias)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.4,1.6</td>
<td>5.0,3.3</td>
</tr>
<tr>
<td>2</td>
<td>1.5,1.6</td>
<td>6.5,3.3</td>
</tr>
<tr>
<td>3</td>
<td>1.9,2.6</td>
<td>6.5,2.6</td>
</tr>
<tr>
<td>4</td>
<td>2.0,1.6</td>
<td>9.0,4.4</td>
</tr>
<tr>
<td>5</td>
<td>2.1,1.6</td>
<td>1.1,3.0</td>
</tr>
<tr>
<td>6</td>
<td>2.1,1.0</td>
<td>3.5,2.2</td>
</tr>
</tbody>
</table>

the dot spectroscopy will be elaborated upon after discussing the results for the magnetic field perpendicular to current orientation.

A complication that should be noted here is that all resonances shift to higher bias with only a diamagnetic trend. This is in contrast to the single-electron theories for two-dimensionally and three-dimensionally confined, nearly cylindrical quantum dots in a magnetic field.[34,90,48] These theories show that some states increase in energy with magnetic field, while other states decrease in energy. Starting from zero field, the ground state always shifts upward in energy, while the second state always shifts downward (ignoring spin) for low field (typically below 3 Tesla). This is due to the Zeeman term (the term linear in $B$) in equation 5.6. Figure 5.20 shows a numerical calculation of the dependence on the energy eigenstates upon parallel magnetic field for the 3D potential shown in figure 5.7. The solid lines show $\ell = 0$ states, which exhibit only diamagnetic shifts, the dotted lines show the $\ell > 0$ states. The absence of a linear behavior in magnetic field in the experimental data implies that either we can not probe $\ell > 0$ states, possibly due to orthogonality between the emitter wave functions and $\ell > 0$ dot levels, or this simply tells us that $\ell$ is not a good quantum number for this system. This implies the assumed cylindrical symmetry has been distorted significantly. It should also be noted that other groups[102,94,86] investigating similar systems always observe a diamagnetic dependence only.
Figure 5.19: Threshold voltage for the first current plateau in reverse bias versus magnetic field squared. Note the excellent agreement with the theoretical fit.
Figure 5.20: Calculation of the energy eigenstate dependence upon magnetic field (field parallel to current) for the potential shown in figure 5.7. The solid lines denote $\ell = 0$ states, the dotted lines denote $\ell > 0$ states.
Wide Bias Magnetoconductance in Perpendicular Field

As discussed earlier, for field directions in the plane of the lateral quantum dot potential, the quantum dot states should not be greatly perturbed in energy by the magnetic field (<0.5 meV at 10 Tesla), due to the strong z confinement. Figures 5.21 and 5.22 show the reverse and forward current-voltage characteristics versus perpendicular magnetic field, for the same temperature and bias conditions as in figures 5.17 and 5.18. As expected, all of the current steps attributed to 0D dot states are not affected greatly by perpendicular field, in contrast to the diamagnetic movement observed in parallel field. This is confirmation that the electron spectroscopy is the result of tunneling through the laterally localized dot states that reside in the strong confinement of the quantum well.

Current Suppression in Perpendicular Field

An additional observation is that the plateau current is suppressed for the perpendicular magnetic field orientation in both bias directions. Especially note the lower plateaus in figure 5.21, which appear to disappear due to the current scale. Figures 5.23 and 5.24 show how dramatic these effects are. Current versus magnetic field squared is shown for the maximum and minimum current observed on the first plateau in the forward bias (figure 5.24) and reverse bias (figure 5.23) directions, in both parallel and perpendicular field orientations. In parallel field, the difference between the current minimum and maximum remains constant on a logarithmic scale over the field range in both bias directions, and only slight suppression is observed at 9 Tesla. The current suppression for perpendicular field at 9 Tesla is very large for the reverse bias (~100 for the plateau current minimum and ~30 for the current maximum). In forward bias, a factor of approximately 10 is observed for both the reverse and forward bias directions. The noise limit for current is approximately 0.1 pA, so we are still at least a factor of 10 above the noise level in these measurements.

This current suppression has been observed previously in a coupled-dot system,[99] and was attributed to a decreasing 0D-0D transition probability between the dots. In
Figure 5.21: Current-voltage characteristics versus magnetic field perpendicular (0-9 Tesla in 0.1825 Tesla steps) to the current for the reverse bias direction.
Figure 5.22: Current-voltage characteristics versus magnetic field perpendicular (0-9 Tesla in 0.1825 Tesla steps) to the current for the forward bias direction.
Figure 5.23: Maximum and minimum current values for the first plateau in reverse bias as a function of perpendicular and parallel magnetic field.
Figure 5.24: Maximum and minimum current values for the first plateau in forward bias as a function of perpendicular and parallel magnetic field.
our case, since there is one quantum well, the perpendicular field can \textit{only} be responsible for decreasing the 1D emitter to 0D dot transition probability. At higher biases in the forward direction, the current at high magnetic fields is actually larger than at zero field (examine the difference in high bias behavior between 0 T and 9 T in figure 5.22). This was also been observed in a coupled-dot system.\cite{99} A possible mechanism for this increased current is inelastic contributions to the tunneling current, which increase with increasing perpendicular field.

Note the extremely linear behavior of the current suppression for the perpendicular field in figures 5.23 and 5.24. If the emitter and dot lateral wavefunctions are modeled as Gaussians, the Log($I$) versus $B^2$ trend can be accounted for. The current is proportional to the overlap between the emitter and dot wavefunctions,

$$I \propto \int \psi_{\text{Emitter}} \psi_{\text{Dot}} = \lambda \sqrt{\frac{\pi}{2}}, \quad (5.17)$$

and where $\psi \propto \exp(-y^2/\lambda^2)$. Under the application of magnetic field that shifts the emitter wavefunctions with respect to the dot wavefunction by a displacement $y_0$,

$$\psi_{\text{Emitter}} \propto \exp\left[-(y - y_0)^2/\lambda^2\right]. \quad (5.18)$$

In this case,

$$\int \psi_{\text{Emitter}} \psi_{\text{Dot}} = \left(\lambda \sqrt{\frac{\pi}{2}}\right) \exp\left(\frac{-y_0^2}{2\lambda^2}\right). \quad (5.19)$$

As current is proportional to this overlap, Log($I$) $\propto y_0^2$. If $y_0 \propto B$, we have the Log($I$) $\propto B^2$ behavior observed experimentally.

To consider whether this trend is reasonable, note coupling to the strong potential in $z$ for the dot prevents a substantial shift of the wavefunction center.\cite{3} The shift of the emitter wavefunction, $y_0$, may be estimated using a parabolic approximation to the lateral potential in magnetic field and assuming that in field the potential is offset by $y_0$.

$$V = \frac{1}{2} \omega_c(y^2 - y_0^2) \quad (5.20)$$

\footnote{If the wavefunction in the dot shifted significantly, the confinement energy would also change, which is not seen experimentally (i.e., no plateau shift for magnetic field perpendicular to current).}
From equation 5.9 the displacement $y_0$ is given by

$$y_0 = \frac{p z \omega_c}{m^*(\omega_c^2 + \omega_0^2)}.$$  \hspace{1cm} (5.21)

For $\omega_0 > \omega_c$, this term varies as $B$, as required. If this model is correct, and if the collector and emitter contacts are similar, then upon reversing bias and field direction, similar suppression effects should be observed. However, in practice, the emitter and collector contacts may be dissimilar enough to prevent observation of this.

Sakai and coworkers[85] have proposed a similar model to account for the current suppression observed through impurity tunneling in a quantum well. In their model, current as a function of voltage and magnetic field perpendicular to current is given by

$$J(V, B) = -\frac{2e\pi n^*}{\hbar} \int_{k_0 - k_c}^{k_0 + k_c} \frac{|M(k_y, k_v)|^2}{k_v} dk_y.$$ \hspace{1cm} (5.22)

The value $M(k_y, k_v)$ is determined primarily by the overlap integrals between the emitter and dot wave functions, and therefore this model is qualitatively similar to ours. The value $k_c$ depends upon applied bias, and can range from $k_F$, the Fermi wavevector, to $0$. $k_v$ is the value of $k_x$ determined from energy conservation for a given value of $k_y$, $k_v^2 = k_c^2 - (k_y - k_0)^2$. In this model, $k_0$ is the separation of the dispersion relationships between the emitter and the dot, given by $eB\Delta S/\hbar$, where $\Delta S$ is the separation between the emitter and dot states. In reference [85], the emitter is 3D, so it is assumed that it can be written as a plane wave in the $x$ and $y$ directions. In our case, this is not the case. Only certain values for $k_x$ and $k_y$ will be allowed, due to the 1D nature of the lead.

**Wide Bias Range Magnetotunneling Summary**

The results from the wide bias range magnetotunneling experiments can be divided into two categories, magnetic field perpendicular to current and magnetic field parallel to current. For the field parallel to the current, the expected diamagnetic shifts of the current plateau edges were observed and the magnitude of these shifts corresponds to a dot wavefunction
extent of approximately 100 Å. Only diamagnetic shifts were observed in the data, which implies that only \( \ell = 0 \) states are present. For field perpendicular to the current, little shift of the plateau edges is observed and the plateau current is observed to decrease linearly as \( \log(I) \) versus \( B^2 \). This trend is understood in terms of the decreasing overlap between the radial emitter and dot wavefunctions, assuming that the dot wavefunction remains fixed, while the emitter wavefunction moves in magnetic field.

5.5.5 Spectroscopy Interpretation: The N-Dot Model

As discussed in section 5.5.4, the wide bias magnetotransport studies point out some clear inconsistencies with the expected quantum dot system. The magnitude of the diamagnetic shift implies a spatial extent for the wavefunction of approximately 200 Å in diameter. For this dimension, a charging energy of 5 meV is expected,\(^1\) and a quantization energy, \( E_Q \), of 35-45 meV between the ground state and first excited state (the lower limit is the parabolic result, the upper limit is the hardwall result). While the charging seems reasonable to account for the observed state to state separations (see table 5.1), the quantization energy is far too large to account for the spacing from the second to third plateau. As the ground state is only doubly degenerate due to spin, we would expect to observe only two plateaus, separated by 5 meV/\( \alpha \), and then nothing should occur in the I(V) until at least 70 mV higher. Clearly a model beyond simple charging and quantization is required to explain the experimental data.

To explain similar diamagnetic shifts in previous work, a model of conducting filaments through the dot has been proposed by Tewordt and coworkers.\(^{102}\) The filaments are assumed to be caused by the potential created by dopant atoms near or in the dot which enhance the lateral quantization already present in the dot. Since on average all filament

\(^1\)In estimating the charging energy, \( E_C \), the crucial parameter is the junction area. The geometrical capacitance argument assumes a metallic density of states, which is a questionable assumption in the emitter and collector electrodes, and certainly suspect in the dot. To provide a better estimate, a charging radius \( R_C \) is assumed. \( R_C \) is estimated from the wavefunction extent determined from the diamagnetic shift (100 Å), which results in \( E_C \approx 5.0 \) meV.
Figure 5.25: Energy ladder diagram for the two dot model. For no electrons in the dot (N=0), there is a ground state spin degenerate level for the left and right dot (E_{0L} and E_{0R}), separated by a small offset, $\delta E_Q$. When the dot is occupied by one electron (N=1e), it will occupy the lowest state available, $E_{0L}$, and the spin degeneracy of that state will be split by the Coulomb energy, $e^2/2C$, into $E_{00L}$ and $E_{01L}$. The right state is also assumed to be lifted by the charging energy. The second electron (N=2) will enter the next lowest energy state ($E_{01L}$). The third electron (N=3) will then enter the ground state of the right dot.
states are laterally localized by about the same amount, all of the peaks should exhibit similar diamagnetic shifts, which is observed. This also yields a more complicated density of states for the dot. However, transport through the system is still proceeding via tunneling through a highly localized dot state. Note that this model also provides for the distortion of the cylindrical symmetry that is needed to prevent observation of $\ell > 0$ levels. Therefore, this model is consistent with the experimental data. Applying this to our case, we expect to have a series of Coulomb-split plateau pairs with similar current step heights ($\Delta I_n$), current suppression (for $B \perp I$), and diamagnetic shifts. Restated, the conduction model is that we are probing $N$ dots in parallel with the emitter. For this specific sample, $N \approx 2 - 3$. In examining table 5.1, note that for reverse bias, the diamagnetic shift is the same for $N = (1,2)$ and for $N = (3,4)$, and that the energy spacing between all of these is close to what is expected for the charging energy. In the forward bias, a similar trend is observed for the $N = (1,2)$ and $N = (4,5)$ pairs. Figure 5.25 shows the conceptual energy ladder diagram for this transport process.

The current suppression and current step height ($\Delta I_n$) for these pairs should also track together. Table 5.2 lists the approximate $\Delta I_n$ at 0 and 9 Tesla ($B \perp I$) and suppression values for reverse and forward bias for the first five plateaus. There is some degree of uncertainty in estimating $\Delta I_n$, as the assumption is that each current plateau is independent of the others, and that the background line shape the plateau is riding on top of (caused by the preceding plateaus) is flat. This approximation begins to break down around the fourth plateau in both bias directions. For the $N = (1,2)$ pairs in both bias directions, there is good agreement between both the $\Delta I_n$ values and the suppression. For the $N = (3,4)$ pair in reverse bias and the $N = (4,5)$ pair in forward bias, the agreement is not as good, although as previously mentioned there is great uncertainty in $\Delta I_n$ in this regime. However, it is interesting to note that for the $N = 3$ plateau in forward bias (which exhibited a large diamagnetic shift compared to the other steps) exhibits a suppression value different from the other pairs. This could indicate it is the result of a third dot; however its absence in
<table>
<thead>
<tr>
<th>Plateau Index</th>
<th>$\Delta I_n$ (pA, B=0 Tesla)</th>
<th>$\Delta I_n$ (pA, B=9 Tesla)</th>
<th>Suppression</th>
</tr>
</thead>
<tbody>
<tr>
<td>N</td>
<td>(reverse,forward bias)</td>
<td>(reverse,forward bias)</td>
<td>(reverse,forward bias)</td>
</tr>
<tr>
<td>1</td>
<td>200,600</td>
<td>8,70</td>
<td>25,8,6</td>
</tr>
<tr>
<td>2</td>
<td>200,500</td>
<td>11,60</td>
<td>18,8,3</td>
</tr>
<tr>
<td>3</td>
<td>800,1100</td>
<td>80,240</td>
<td>10,4,6</td>
</tr>
<tr>
<td>4</td>
<td>2300,1600</td>
<td>500,570</td>
<td>4,6,2,8</td>
</tr>
<tr>
<td>5</td>
<td>2500,2500</td>
<td>1400,580</td>
<td>1,8,4,3</td>
</tr>
</tbody>
</table>

Table 5.2: Experimentally observed current step values in zero field and also for B=9 Tesla (B \(\perp\) I). Suppression values are the ratio of $\Delta I_n(B = 0)/\Delta I_n(B = 9T)$.

Forward bias can not be explained. While the N-dot model can not be conclusively proven with this data, it is consistent with many of the features observed (i.e., state separation, magnetic field dependance, current suppression, and the lack of Zeeman shifts).

**Spectroscopy Summary**

From the variable temperature data, an energy scale is determined. The magnetotunneling data both confirms the localization of the dot states in the epitaxial quantum well and also determines the lateral localization length. From this, a model of conduction through multiple dots in parallel is proposed and shown to be consistent with the observed energy spacings, the lack of Zeeman shifts, and the grouping of plateaus into pairs with similar diamagnetic shifts and values of current suppression. Now that we have accounted for the major spectroscopy, the focus will turn to the fine structure superimposed on the current plateaus.
5.5.6 Magnetoconductance of the Fine Structure

The thermal dependence of the conductance has already been presented as evidence that the plateau edges result from localized 0D dot states crossing the Fermi level, and that the fine structure results from states below the Fermi level in the emitter becoming resonant under bias with the sharp 0D dot states. Although the magnetotunneling data presented in section 5.5.4 allowed for tracking the motion of the plateau edges in field, the field steps were too large to observe the magnetic field dependence of this fine structure. Figure 5.26 shows conductance versus voltage traces for the forward bias direction at low applied bias. The major peaks at 15.5 mV, 24.5 mV, and 30.0 mV (shown with arrows) correspond to the plateau edges in current. Note how the fine structure between these plateau edges changes dramatically as the field is stepped from 0 to 1200 Gauss, in 100 Gauss increments (seen especially in the region between the second and third major peaks in the conductance).

The fine structure characteristics are reproducible, even after thermally cycling the sample to 300 K. The field steps are more than an order of magnitude smaller than those shown in figures 5.17 and 5.18. It is clear that the evolution of this fine structure in field would be difficult to determine without stepping field on the 100 Gauss level (compare the top and bottom trace on figure 5.26). It is also apparent that this field dependence is different from that of the dot states. Other groups [102,94,42,100] have seen similar fine structure, however their field steps were on the order of Tesla, rather than 100 Gauss, so the dependence of this fine structure upon field appeared random.

In order to better characterize this fine structure, data was acquired in the low bias regime (approximately ± 45 mV), every 187.5 Gauss from 0 to 9 Tesla. Over one thousand I(V) curves were acquired over a time scale of hundreds of hours in the dilution refrigerator, at a mixing chamber base temperature of 30 mK. Data was acquired in both device bias directions and in both parallel and perpendicular magnetic field. Figures 5.27, 5.28, and 5.29 show selected portions of this data for the forward bias direction. In figure 5.27, the magnetic field is parallel to the current and ranges from from 0 Tesla (bottom) to 2.25 Tesla.
Figure 5.26: Conductance versus voltage, arbitrarily offset for clarity, showing the evolution of the fine structure with magnetic field. Field is varied from 0 Gauss (bottom trace) to 1200 Gauss (top trace), in 100 Gauss increments.
(top). Figure 5.28 shows the same magnetic field orientation, now at higher field values (6.75–9.0 Tesla). Note that the fine structure is still clearly present at high field. Figure 5.29 shows the same magnetic field range as in figure 5.27, however now with the magnetic field perpendicular to the current orientation. The I(V) traces in these figures have been arbitrarily offset, so the movement of the fine structure with field is apparent. Note that unlike the case of localized dot states, the fine structure exhibits motion in both parallel and perpendicular field orientations.
Figure 5.27: I-V data in parallel field, ranging from 0 Tesla (bottom) to 2.25 Tesla (top). Curves are arbitrarily vertically offset.
Forward Bias
$B=6.75-9$ Tesla ($B \parallel I$)

Figure 5.28: I-V data in parallel field, ranging from 6.75 Tesla (bottom) to 9.0 Tesla (top). Curves are arbitrarily vertically offset.
Figure 5.29: I-V data in perpendicular field, ranging from 0 Tesla (bottom) to 2.25 Tesla (top). Curves are arbitrarily vertically offset.
Fan Diagrams

To generate fan diagrams that show how the fine structure peaks depend upon field, the data was transformed numerically into conductance versus voltage curves. A peak finding algorithm searched the conductance traces for peaks. The conductance peak positions in voltage were then plotted against magnetic field, generating fan diagrams.

Figure 5.30 shows the results of this procedure for the reverse bias and figure 5.31 shows this for the forward bias in parallel field ($B \parallel I$). In the reverse bias, three lines due to the plateau edges at 10 mV, 20 mV, and 32 mV ($B=0$ Tesla) are clearly visible with a diamagnetic shift (previously discussed, see section 5.5.4) apparent in the curvature of this lines. Between plateau edges, the fine structure peaks form a complex lattice of crossing lines. In the forward bias direction, there are lines at 15 mV, 24 mV, 33 mV, and 40 mV at $B=0$ Tesla, which again correspond to plateau edges that exhibit a diamagnetic shift. Note the weaker lines seen at 35 mV and 42 mV at $B=0$ Tesla, move toward lower voltage with increasing field. The strength of these peaks is much less than that of the plateaus (they are barely visible in figure 5.18). There is also weak evidence of this occurring in the reverse bias (starting at 43 mV, $B=0$ Tesla, extending to 31 mV at 9 Tesla, there is a weak line).

Figures 5.32 and 5.33 show fan diagrams for the magnetic field perpendicular to current case, where there is little motion of the stronger lines associated with quantum dot states. For the reverse bias case (figure 5.32), these are the lines at 18 mV, 29 mV, and 41 mV. For the forward bias case, (figure 5.33) these occur at 7 mV, 17 mV, and 31 mV. Note that these are shifted in voltage from the values seen in figures 5.30 and 5.31. While the fine structure, number of plateaus observed, and plateau width are consistent through device thermal cycling, the spectra is sometimes offset in voltage. We believe this is due to the

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Although these weaker lines are not well understood a possible explanation for these features is the observation of the diamagnetic shift of 1D subbands in the emitter, which would shift to lower voltage in the spectroscopy as they rose to higher energy in the emitter. However, these lines do not move in perpendicular field, which strongly implies that they are associated with the dot.
Figure 5.30: Fan diagram showing peak location versus voltage and magnetic field parallel to the current for the reverse bias direction.
Figure 5.31: Fan diagram showing peak location versus voltage and magnetic field parallel to the current for the forward bias direction.
Figure 5.32: Fan diagram showing peak location versus voltage and magnetic field perpendicular to the current for the reverse bias direction.
Figure 5.33: Fan diagram showing peak location versus voltage and magnetic field perpendicular to the current for the forward bias direction.
Figure 5.34: Histogram showing numerical Fourier transform of the first plateau in the fan diagram shown in figure 5.30. X denotes the mean value for the fine structure peak separation, σ the standard deviation.
Figure 5.35: Histogram showing numerical Fourier transform of the first plateau in the fan diagram shown in figure 5.31. X denotes the mean value for the fine structure peak separation, $\sigma$ the standard deviation.
Figure 5.36: Histogram showing numerical Fourier transform of the first plateau in the fan diagram shown in figure 5.32. $X$ denotes the mean value for the fine structure peak separation, $\sigma$ the standard deviation.
Figure 5.37: Histogram showing numerical Fourier transform of the first plateau in the fan diagram shown in figure 5.33. X denotes the mean value for the fine structure peak separation, σ the standard deviation.
charging configuration that is in place in the emitter and collector upon cool down, which may change from run to run. Note that the weaker lines at 27 mV and 33 mV in the forward bias (figure 5.33) correspond to the weaker lines seen at 35 mV and 42 mV in parallel field (figure 5.31).

Note how the fine structure motion in figures 5.30 and 5.31 clearly correlates with varying magnetic field. From measuring the slope of many of these lines, in reverse bias the fine structure typically moves at a rate of 2.5 mV/T (1.25 meV/T) versus 5.0 mV/T (1.85 meV/T) in the forward direction. Both of these values are on order of the value for $eB/m^*$ (1.8 meV/T in In$_{0.1}$Ga$_{0.9}$As and 1.7 meV/T in GaAs). Similar field dependence of the fine structure is seen in the perpendicular orientations (figures 5.32 and 5.33), which confirms that source of the fine structure does not reside in the dot. Instead, the lack of dependence upon field orientation strongly implies the presence of emitter states. For emitter states, the magnetic field dependence on conductance should not be dependent upon the field orientation, as there is no strong confining potential in the transport direction, as exists for the dot.

The average energy separation between the fine structure peaks on the first plateau in both bias and field directions has been calculated by performing a numerical Fourier transform upon the fan diagrams (figures 5.30, 5.31, 5.32, and 5.33). The transforms are represented using histograms and are shown in figures 5.34, 5.35, 5.36, and 5.37. Table 5.3 shows the values for the mean fine structure separation, $\bar{x}$, and the standard deviation, $\sigma$. As expected, no dramatic change in the peak separation is observed when the field orientation is varied.

In previous work by Su and Goldman,[94,42] the fine structure was observed to quench in magnetic field. This was viewed as evidence against the fine structure being attributed to a discrete density of states in the emitter. This is not the case observed for the samples studied in our work. Even at fields of 11 Tesla (see figure 5.38), the fine structure is still strong and clearly present. In strong perpendicular field, the fine structure is observed to


<table>
<thead>
<tr>
<th>Plateau</th>
<th>Fine Structure Separation (Bias, Field)</th>
<th>$\bar{x} \pm \sigma$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reverse, $B \perp I$</td>
<td>0.91±0.4</td>
<td></td>
</tr>
<tr>
<td>Reverse, $B \parallel I$</td>
<td>0.83±0.3</td>
<td></td>
</tr>
<tr>
<td>Forward, $B \perp I$</td>
<td>0.58±0.2</td>
<td></td>
</tr>
<tr>
<td>Forward, $B \parallel I$</td>
<td>0.81±0.3</td>
<td></td>
</tr>
</tbody>
</table>

Table 5.3: Average fine structure separation in meV, determined from the histograms shown in figures 5.34, 5.36, 5.35, and 5.37.

quench at the same rate as the current steps. Both are suppressed due to a decrease in the emitter-dot transition probability in perpendicular field, and this is the expected result if the fine structure is due to the density of states in the emitter.

5.5.7 Origin of the Fine Structure

From the temperature dependence, we know that fine structure is due to tunneling processes from states below the Fermi level in the emitter. The thermal activation of a pre-resonant fine structure peak is also in agreement with this model. Other groups have recently shown that the fine structure is a result of states in the emitter by examining transport in an asymmetric structure.[86] In these experiments, transport employing a thin emitter and a thick collector shows no fine structure, as $T_C$ (the collector transmission coefficient) controls the tunneling current. In the other bias direction, when injecting from the collector, the fine structure is observed. This clearly indicates that the fine structure is dependent upon the emitter. In our symmetric structure, we observe the fine structure in both bias directions, as expected. The observation that the fine structure is generally more pronounced in the reverse bias (compare figures 5.11 and 5.13) is also expected. The voltage to energy conversion factor, $\alpha$, for the reverse bias is slightly larger than $\alpha$ for forward bias. This implies that the emitter is thicker for the reverse bias and thinner for the forward bias, which is consistent with the extreme case presented in reference [86]. Another piece of evidence that
Figure 5.38: Current and conductance versus voltage characteristics for the quantum dot at 11 Tesla.
links the fine structure to the emitter in this work is the equal suppression of both the fine structure and overall plateau current in parallel field. The motion of the fine structure in magnetic field, which is on the order of $\frac{\hbar \omega_c}{c}$, is consistent with what one expects for states in GaAs when the confinement energy is weak. The lack of dependence upon magnetic field orientation excludes states localized in the dot, as states in the dot exhibit very different response in parallel ($B || I$) versus perpendicular ($B \perp I$) magnetic field orientations due to the strong $z$ confinement.

1D Subbands?

The observed energy separations of 0.6-0.9 meV are too small to arise from 1D subbands in the leads. For a 1400Å wide lead, the 1D subband leads are calculated to be separated by 15 meV near the Fermi level. Even if the lead diameter is increased to 3400Å (much larger than the upper possible fabrication size of approximately 1500Å,) the calculated energy spacings are still 5 meV.

Other possible models to account for this close energy separation, including interference effects[15,95], universal conductance fluctuations (UCF)[92], or effects due to chaotic transport[55, 36, 37] can be individually excluded. Interference effects require sharp corners in the lateral potential, which we do not expect, and predict far larger values for the energy separation of the fine structure than is observed (on order 50 meV). UCF effects are expected to show a random motion in magnetic field, not the monotonic dependance upon field that is exhibited by the fine structure. Additionally, the observation of a thermally activated peak, the lack of a strong temperature dependance of the fine structure, and the observation of the fine structure at temperatures greater than 10K also make UCF effects unlikely as an explanation for the fine structure. The localization extent of the dot states makes any chaotic trajectories inside the dot unlikely as well.
Discrete Dopants and Localization in \( z \)

This small energy separation between the fine structure peaks in conductance indicates that they may be due to the effect that individual donors have upon the 1D density of states in the emitter. For the dopant levels used in this structure, there are approximately two electrons in a 100 Å cube in the emitter (the value of 100 Å is used as it is the mean free path in the doped lead). This places about one hundred dopant atoms with electrons contributing to the degenerate doping of the conduction band in this volume, and for a calculated Fermi level of 80 meV, this results in level spacings that are approximately 0.8 meV in energy (2.2 mV in forward bias, 1.6 mV in reverse bias). This in good agreement with the observed 0.6-0.9 meV spacing. Another possibility is to assume that due to fluctuations in the 1D emitter lead cause localization in the \( z \)-direction (current direction). While this could be occurring, it is still difficult to obtain energy spacings of 0.6-0.9 meV from this effect without increasing the localization length to very large values (i.e., >3400 Å). To investigate the responsible process further, samples with different dopant levels and spacer layer thicknesses could be fabricated to see if the fine structure energy spacing is related to the discrete dopants.
5.6 Conclusions

The electron spectroscopy of the quasi-0D vertical InGaAs quantum dot resonant tunneling structure is investigated as a function of magnetic field and temperature. Steps in current due to the discrete nature of the dot density of states are observed. Under magnetic field parallel to the current, the step edges exhibit a diamagnetic shift as expected. The magnitude of this diamagnetic shift is used as a direct measure of the radial extent of the wavefunction in the dot. In the perpendicular (in-plane) field orientation, no shift is observed, which conclusively shows that the current steps are due to states localized in the epitaxial quantum well. Inconsistencies in the step spacings, the diamagnetic shifts, and lack of Zeeman splittings indicate that transport is proceeding through more than one channel in parallel (i.e., two dots, or more). A model incorporating this type of transport is proposed and found to be consistent with the first few plateaus.

The highly localized dot states are then utilized as ‘probes’ to examine the electronic structure of the emitter electrode. Discrete states in the emitter are observed through the presence of fine structure on top of the normal current plateaus that modulates over a period of approximately 0.6-0.9 meV. The fine structure is observed to be reproducible, both upon repeated traces and upon thermal cycling. Variable temperature results indicate that this fine structure originates from below the Fermi level in the emitter. With increasing temperature, states above the Fermi level are populated. The observation of preresonant thermally activated peaks as temperature increases is in excellent agreement with the emitter discrete state model. In both parallel and perpendicular magnetic field orientations, the fine structure peaks shift at approximately $\hbar \omega_c = 1.7 \text{ meV/T}$, which is again consistent with emitter states. Figure 5.39 shows a summary of the spectroscopic features, and their assignments in this model.
Figure 5.39: Absolute value of current versus voltage for the lower two plateaus in reverse and forward bias. The emitter states on top of the plateaus have been highlighted and the index of the plateaus in the $N$-dot model is given.
Chapter 6

Controlled III-V Semiconductor Cluster Nucleation and Epitaxial Growth via Electron Beam Lithography

6.1 Introduction

The fabrication of low dimensional semiconductor structures usually involves some type of post-growth lithographic pattern transfer strategy, such as dry-etching of topographic features, which tends to create electrically and optically active defects[19,79]. An alternative approach would be to attempt selective-area deposition by altering the substrate surface chemistry or orientation (i.e., (100), (111), ...) prior to growth. However, this methodology (i.e., growth through windows in dielectric masks or on patterned substrates) typically requires a complex processing scheme and is very sensitive to growth and surface conditions [10,68]. For example, electron beam irradiation has been employed to modify the naturally
occuring oxide on a GaAs substrate, thereby providing a patterned mask for Cl₂ based dry etching and subsequent low-temperature growth [52].

A combined electron beam lithography / epitaxial growth procedure for controlling the nucleation and growth of two-dimensional (2-D) InAs and GaAs thin films and three-dimensional (3-D) InAs clusters on GaAs substrates has been developed. This technique provides sub-micron lateral resolution and does not require pattern transfer or post-growth fabrication. While others have shown that a statistical distribution of InAs clusters may exhibit quantum-size effects[13,32], the technique described here can be used to isolate and register such structures to a pre-defined lithographic pattern. This potentially can create isolated quantum dot structures, that could be electrically probed in a method similar to that used in chapter 6.

6.2 Procedure

The starting material consists of semi-insulating (100) GaAs that has been pre-coated with 100 nm of Al₀.₆Ga₀.₄As via metalorganic chemical vapor deposition (MOCVD). The resulting Al₀.₆Ga₀.₄As/GaAs wafer has been cleaved into several 10 x 10 mm² samples and stored under ordinary laboratory conditions. Consequently, a thin layer of native oxide forms on the surface (Figure 6.1a). Electron-beam (e-beam) exposure has been performed at an accelerating potential of 40 keV, a current of 100 nA, a beam diameter of 0.25 μm, and a base pressure of 2x10⁻⁷ torr. A variety of lithographic patterns have been written on each sample, and the integrated charge density, \( \sigma_e \) has been varied between 10 and 100 C/cm². This procedure yields a thin, chemically resistant native oxide layer and an e-beam irradiated semiconductor volume beneath the surface which is roughly 5 μm in diameter (Figure 6.1b). The sample is then etched in a hot HCl:H₂O solution (\( \sim 75^\circ C \)) to remove the Al₀.₆Ga₀.₄As masking layer and thereby eliminate surface contamination. Finally, the sample is transferred into the MOCVD reactor and annealed at 700°C for 360 seconds.
This combined wet-etching / thermal annealing process is identical to our standard GaAs substrate preparation procedure. This is known to yield an “epi-ready” GaAs surface (figure 6.1c).

The growth of InAs is initiated by lowering the susceptor temperature, and introducing a pre-stabilized TMIn / H₂ gas phase mixture. Scanning electron and atomic force microscopies were used to study the topology of the InAs clusters. Figure 6.2 is a typical SEM image, and figure 6.3 shows an AFM of similar clusters. Note the alignment of the clusters to the GaAs substrate orientation. Figure 6.1d is a schematic cross-section of a generic InAs-on-GaAs sample after e-beam lithography and epitaxial growth. The scanning electron microscope (SEM) image in figure 6.4 shows an e-beam written array consisting of 0.5 x 0.5 μm² pixels on 2 μm centers with σₑ = 50 C/cm². The center-to-center pixel spacing is small enough, and the e-beam dose large enough, such that cluster nucleation is completely eliminated over a 50 x 50 μm² area (region 1), even though the nominal InAs film thickness is 10 nm. The equilibrium cluster density has been pre-determined by selecting the appropriate growth conditions.[111] Note that in this particular case, the Al₀.₆Ga₀.₄As masking layer has not been completely removed in the e-beam irradiated region as evidenced by the small anomaly in the upper left hand corner (region 3).

6.3 Auger Analysis

Auger electron spectroscopy (AES) has been performed in order to determine the chemical composition in these three distinct regions. In all cases (figure 6.4b, 6.4c, and 6.4d), the carbon and oxygen contamination on the sample surface, caused by long-term exposure to ordinary laboratory conditions, is removed by surface sputtering. In figure 6.4b, the peak attributed to indium is eliminated after only 2 nm of sputtering indicating the presence of only a thin InAs wetting layer (region 1). The lack of a measurable Al signal verifies that the Al₀.₆Ga₀.₄As has been completely removed via the wet-etching procedure. A separate
Native Oxide ($\leq 10$ nm) $\downarrow$

Al$_{0.6}$Ga$_{0.4}$As (6-100 nm)

InAs (10-200 nm)

Figure 6.1: Schematic diagram of the combined electron beam lithography / epitaxial growth process.
Figure 6.2: Typical SEM image of the InAs clusters on (100) GaAs substrate.
Figure 6.3: AFM image of the InAs clusters on (100) GaAs substrate.
Figure 6.4: Scanning electron microscope image in (a) of selective-area InAs cluster nucleation ($T_d = 650$ C$^\circ$, $R_g = 0.15$ nm/s). The Auger electron spectra taken from region 1, 2, and 3, are plotted in (b), (c), and (d), respectively. In each case, the upper (lower) spectrum was measured before (after) argon ion sputtering removed $\sim$20Å of material.
optical microscope study shows that the etch resistant (e-beam exposed) oxide is undercut by first etching through the native (un-exposed) oxide and then attacking the remaining Al$_{0.6}$Ga$_{0.4}$As from all sides (i.e., lift off occurs). In Figure 6.4c, outside the e-beam irradiated area but between InAs clusters (region 2), the Auger data shows evidence of a slightly thicker InAs wetting layer (i.e., a small In signal remains after sputtering). The large Al signal in figure 6.4d is attributed to remnants of the Al$_{0.6}$Ga$_{0.4}$As masking layer (region 3).

### 6.4 Atomic Force Microscopy Studies of Surface

Atomic Force Microscopy (AFM) studies were performed to see how the e-beam exposure modifies the sample surface morphology, prior to growth. Figure 6.5 is a study of how the electron beam sweep rate (the frequency the beam sweeps back and forth across the sample to deposit a given dose), affects the surface of samples with a 1000Å Al$_{0.6}$Ga$_{0.4}$As cap layer. AFM images are shown, with corresponding cross sections. In all figures, there are three important regions. First, in region 1 there is the background level, which is farthest from the e-beam exposure. This surface is not changed during the process as expected, and high resolution AFM shows atomic terracing. The next region, region 2, is the 15 - 20 Å high, 10µm wide feature. The third region occurs in the center of region 2. In the case of a) (1 Hz Sweep Rate) there is a depression in the center of this region 2, in b) (500mHz) there is a depression with a peak in the center, and in c) (50mHz) there is no depression and a peak. The location of the peak corresponds to where the e-beam dosage was actually swept, and the width ( 1µm) corresponds to the lithographic width of the feature. At a higher sweep rate of 1 Hz, the depression which forms centered around where the e-beam hit the surface, and the width of the depression varies according to the sweep rate. The width of region 2 is approximately 10µm, consistent with the length scale for energy loss of 40 keV electrons. The magnitude of deformation in both regions 2 and 3 is e-beam dosage dependent. Contamination of the sample from the e-beam polymerization of hydrocarbons
Figure 6.5: Atomic Force Microscopy (AFM) images (left) and corresponding crossections (right) of three lines written at different e-beam frequencies. (a) 1 Hz, (b) 500 mHz, and (c) 50 mHz.

(either already present on the sample from storage in ambient conditions or from the e-beam lithography system diffusion pumps) is the most likely explanation for this surface topology. What is unique in this case from other contamination studies[53] is that at more rapid sweep rates, the contamination is observed to be reduced in the center. Additionally, high resolution AFM reveals atomic terracing is still present in all written regions, as shown in figure 6.6.

A similar study examined the dosage effects at a fixed e-beam frequency. Figure 6.7 shows a series of lines, all written with a 50 mHz e-beam sweep rate, increasing from five
Figure 6.6: High resolution AFM image showing atomic terracing. The total height variation is 11Å.
Figure 6.7: AFM images of a series of lines written at 50 mHz, varying the number of beam passes.
beam passes (bottom), where the broad background becomes apparent, to ten, twenty, and finally forty passes (top). The surface topology becomes more enhanced with the increasing number of beam passes. As in the frequency dependance study, this was also written on a 1000Å Al$_{0.6}$Ga$_{0.4}$As cap layer. This indicates that even for single passes, that deposition of a contamination layer occurs. Grids of written dots were also examined, and these yielded a contamination array on the surface, as seen in figure 6.8.

An indepth study of this contamination was performed which will be published elsewhere[2]. The major results of this study was that for a fixed e-beam dosage, the contamination build-up is observed to be a function of the e-beam frequency, and approaches a saturation value for slow frequencies. Figure 6.9 shows AFM crossections for a intermediate range of e-beam sweep frequency rates, prior to the range where the onset of saturation is observed.

In addition to the separate optical microscope study that shows that e-beam exposed area is undercut, a separate study tested whether the sacrificial AlGaAs layer really lifts off the contamination on top of it. Titanium / Gold features (100Å Ti, 1000Å Au) were photolithographically defined on top of the AlGaAs layer. The standard etch was then used, and undercut and lift off of the Ti/Au features was observed. This confirms that even though contamination may be present on top of the AlGaAs layer, it should be removed prior to growth.

### 6.5 Growth Inhibition Results

The scanning electron microscope (SEM) images in figures 6.10 and 6.11 demonstrate that this method of selective-area deposition is effective even for a 20-fold increase in InAs film thickness. In both figure 6.10 and 6.11, the width of the denuded trenches is considerably larger than the lithographic feature size ($\sim$ 1 μm). Nevertheless, the length scale over which growth is inhibited is directly proportional to the integrated charge density. For example, a
Figure 6.8: AFM of an e-beam written AlGaAs surface. The dark regions (depressions) in the four by four array are where the e-beam dose was deposited.
Figure 6.9: AFM cross-sections across an e-beam exposed dot region. The contamination peak height increases for slow frequencies.
50% increase in $\sigma_e$ yields almost a 2-fold improvement in the aspect ratio (height-to-width ratio of the as-grown feature).

Given that growth inhibition does not result simply from surface contamination or an etch resistant native oxide, we believe that e-beam irradiation alters one of the characteristic energies of the epitaxial growth process, i.e., adsorption, surface diffusion, or cluster formation[107]. If surface diffusion were the limiting step, then we should observe a large build-up of islands at the edges of the e-beam written area accompanied by a monotonic decrease to the equilibrium density further away from the lithographic pattern (which is generally not the case). Moreover, the cluster formation energy is primarily a function of deposit-atom to deposit-atom bond strength, and should not be greatly affected by an e-beam modified substrate-deposit interaction. Thus, by a process of elimination, we conclude
Figure 6.11: Same as in figure 6.10, except now the integrated charge density is increased to 30 C/cm$^2$. 
that a reduction in the energy of adsorption must be responsible for our observations. This could possibly be a consequence of an e-beam induced surface reconstruction owing to the large charge density deposited in the substrate or of surface relaxation caused by structural damage.

A similar investigation has been performed on samples with only a 6 nm Al$_{0.6}$Ga$_{0.4}$As masking layer. In this particular case, the Auger spectra from the e-beam irradiated area exhibits strong Al and O signals between the InAs wetting layer and the GaAs surface even after 4 nm of sputtering. In contrast, no Al or O is detected in the un-exposed region. These observations are consistent with earlier reports involving GaAs native oxide masking,[52] except that in the present case the e-beam exposed oxide is stable even after high temperature annealing (700 C°). From a limited set of experiments, we have determined the following: (1) heteroepitaxy of InAs on GaAs is inhibited over a wide range of growth conditions ($475 \leq T_g \leq 650$C° and $0.15 \leq R_g \leq 1$ nm/s), (2) growth inhibition is also exhibited on n$^+$ (100) and (111)B GaAs substrates, and (3) this process can be employed for selective homoepitaxy (i.e., GaAs-on-GaAs). We mention that growth inhibition has also been achieved without the use of a sacrificial Al$_{0.6}$Ga$_{0.4}$As layer. However, for this special case, we have not yet confirmed that the GaAs substrate is “epi-ready” prior to growth.

The SEM images of figures 6.12 and 6.13 demonstrate that this phenomenon is useful for sub-micron lithography. Figure 6.12 shows a 50 x 50 $\mu$m$^2$ array of InAs clusters for which the equilibrium density has been matched to the e-beam pixel density. Consequently, the InAs clusters have nucleated at the cross-hairs of the undamaged channels. Figure 6.12 shows that the region of growth inhibition can be restricted to sub-micron lines simply by optimizing the e-beam exposure conditions. Figure 6.14 illustrates how the distance between the 0.5 x 0.5 $\mu$m$^2$ centers influences the growth. The completely bare upper right corner has the same center to center distance as in figure 6.12; however, the exposure dosage was higher. As the center to center distance is increased, the points where the e-beam exposed the sample become clearly visible.
Figure 6.12: Scanning electron microscope image of a 2-D array of InAs clusters. The absolute position of the individual clusters has been manipulated via e-beam lithography, etching (6 nm Al$_{0.6}$Ga$_{0.4}$As mask), and epitaxial growth. The computer generated e-beam patterns consist of 0.5 x 0.5 $\mu$m$^2$ on 2 $\mu$m centers. The growth rate is $R_g = 0.15$ nm/s, and the growth temperature is 650 °C.
Figure 6.13: Similar to figure 6.12, except with 0.5 μm wide lines with variable center-to-center spacing. Growth temperature was 550°C.
Figure 6.14: An SEM image showing that the InAs clusters do not nucleate in the e-beam exposed regions. Pixel spacings are 2 μm (upper right), 4 μm (lower right), 6 μm (upper left), and 8 μm (lower left); a second 2 μm spaced lattice is partially seen in the far lower left, overlapping the 8 μm lattice.
Figure 6.15: Outline of a Hall Bar in an InAs film. The bright line is where growth was inhibited.

It is possible to grow 2D layers of relatively high quality InAs on (111) GaAs.[110] Work is currently underway to create a Hall Bar structure in the as-grown film by using the e-beam inhibited growth to form an isolation region. This is the first application of this technique to create a useful, though relatively large, electronic structure. Figure 6.15 shows an initial attempt at achieving such a structure. These Hall Bars have recently been created, and await mobility measurements.

6.6 Conclusion

We have described a combined e-beam lithography / epitaxial growth process for achieving controlled III-V semiconductor cluster nucleation and epitaxial growth. Selective-area deposition has been observed for InAs-on-GaAs heteroepitaxy, over a wide range of conditions, as well as for GaAs-on-GaAs homoepitaxy. By removing a sacrificial Al$_{0.6}$Ga$_{0.4}$As
layer prior to growth, we have demonstrated that growth inhibition is not caused by surface contamination or a residual native oxide, but results from an e-beam induced change in the GaAs surface energetics. Finally, we have shown that this technique can be employed to achieve ex-situ registration of quantum-size structures.
Chapter 7

Fabrication of Novel Metallic Structures

7.1 Introduction

The lowest value of capacitance for metallic Coulomb blockade structures assembled with conventional nanofabrication techniques is typically limited to approximately $5 \times 10^{-17}\text{F}$ [6]. The constraining factor is the inability to define a junction area smaller than approximately $30 \times 30 \text{nm}^2$, which is currently the smallest area definable using state of the art electron-beam lithography. The reduction of the capacitance in these structures is crucial to higher temperature operation, as the charging energy varies inversely with the capacitance.

When considering the useful operating temperature of any large scale circuit employing Coulomb charging effect devices, such as the single electron transistor (SET), the probability for thermally activated tunneling events, $\Gamma_T$, must be small. $\Gamma_T < 10^{-50}\text{s}^{-1}$ is a reasonable requirement for running a VLSI circuit for a long time [6]. In the limit of many junctions and for $k_B T \ll e^2/C_0$, $\Gamma_T$ can be approximated as

$$\Gamma_T \sim \frac{G}{C_0} \exp \left[ -\frac{e^2}{16C_0k_BT} \right], \quad (7.1)$$
where $G$ is the junction conductance, and $C_0$ is the junction self-capacitance. For the above parameters, this yields an operating temperature of 300 mK.

For reliable operation at a more reasonable temperature (i.e., liquid Helium temperatures), the junction area must be reduced to below 100 nm$^2$, or the capacitance below $5 \times 10^{-18}$F. While some experiments utilizing scanning tunneling microscopy (STM) have studied systems with capacitances as low as $1 \times 10^{-18}$F[87], these systems are not very robust. They are difficult to measure at low temperature and they are certainly not useful in making more than one device at any give time. The subject of this chapter concerns two alternative fabrication attempts to create more robust structures with very low capacitances, using novel nanofabrication techniques.

7.2 Aluminum Towers

7.2.1 Introduction

The fabrication of small area aluminum / aluminum oxide junctions usually employs a resist-bridge, shadow evaporation technique[38]. This results in a lateral layer of junctions on top of a substrate (typically silicon). The lateral junctions have some disadvantages.

I. Offset Charge: One of the most serious problems with utilizing SETs is the non-uniform offset charge that must be compensated for with a tuned voltage offset on the gate electrode. While this is possible to do for a few devices manually (electrometer applications), it becomes very tedious, or practically impossible for applications requiring hundreds to thousands of SET devices (current standards).

II. Awkward Mask Design: The resist-bridge technique for creating junctions is not space efficient. Large empty spaces and areas filled with non-device metal are created. This again is not a problem for only a few devices; however it is a
Figure 7.1: Schematic of a vertical Al/Al₂O₃ double junction structure.

serious issue for possible high density applications.

An alternative to this type of structure would be to create a structure depicted in figure 7.1. The fabrication of such a structure will be discussed in detail later. The two main features are an electron-beam lithography step, which defines the radius of the tower, and a thermal evaporation deposition step, which defines the Al and Al₂O₃ layers in the transport direction. There are a number of potential advantages to a vertical structure.

I. Very Small Metal Islands: Using this technique, the metal island may be made extremely small. In a typical SET the island is usually on the order of a micron. Using this structure, the vertical limit is controllable by evaporation, so it may
be made arbitrarily thin. In the lateral direction, the size is limited by the
lithographic technique employed, which can be as small as 20 to 30 nm using
electron-beam lithography.

II. Metal Superlattices: It is possible with this technique to create more uniform
one-dimensional arrays of junctions vertically, by a repeated sequence of depo-
sitions and oxidations.

III. Offset Charge: Since these junctions are not residing directly on top of an SiO$_2$
layer, as the lateral junctions are, there is likely to be less of a problem with offset
charge. Another attractive feature is the side-wall oxidation that passivates the
sidewalls of the tower.

7.2.2 Tower Fabrication

The starting material is a pre-oxidized n-type Si (100) wafer. The oxide thickness is ap-
proximately 200Å. The wafer conductivity is 1-10 Ω-cm. This ensures that the substrate
is a relatively good conductor at room temperature, when the electron-beam lithography
is done, and an insulator at cryogenic temperatures, when the structure will be electrically
measured. Photolithography and lift-off is then used to define the bottom gold contact layer.
Then the PMMA bilayer (as described in chapter 3) is used to lift-off aluminum/aluminum
oxide towers. The aluminum / aluminum oxide tower is deposited in the evaporation system
described in chapter 3. After this step a planarizing polyimide coating is etched to expose
the tops of the towers which are then contacted with gold metallization using photolithog-
raphy. This fabrication sequence is very similar to the process steps used for the creation
of vertical quantum dot structures.
7.2.3 Tower Fabrication Results

A number of fabrication related problems currently prevent a vertical junction from being realized. The first major problem encountered was the lack of adhesion of the submicron Al towers to the gold contact lines. It is observed that any process involving the use of photo-resist which covers the contact gold at any time creates adhesion problems. As seen with AFM, even after an extensive clean-up, approximately 30 Å of residue is left behind. This layer causes severe adhesion problems for submicron structures. The only workable solution is to arrange the processing so that the e-beam step is immediately after the gold contact layer (i.e., the gold contact layer is never covered with photoresist).

In the course of resolving the adhesion problem, it became apparent that using AFM as a fabrication diagnostic is crucial for this type of system. It was observed that scanning electron microscopy was very ineffective in resolving the towers, as the contrast between a sub-100nm metal tower and the underlying gold wire is quite poor. Figures 7.2 and 7.3 show AFM images of the highest aspect ratio towers fabricated. In figure 7.2, an array of aluminum towers is shown, with some of the array overlapping the underlying gold wire. The ridges seen at the edges of the 2 µm gold contact wire are related to the photolithography lift-off process that defines the gold wire. Figure 7.3 shows a three dimensional rendering of the towers. Note in this AFM, four towers are observable (two on the gold wire, two on the substrate). The towers are not uniform in diameter, and have a pointed top.

Most importantly, it is observed that towers reach a height limit which is related to their lithographic size, rather than the evaporated thickness. Figure 7.4a is a 1 × 1µm image of 80 nm (size in lithographic design) Al towers and 7.4b shows a 2 × 2µm image of 30 nm Al towers. Corresponding crosssections for the Al towers in these images are shown in figures 7.5a and 7.5b respectively. Two effects are obvious. First, note the more pronounced peak on the top of the smaller tower in figure 7.5b. The peak for this tower is both sharper and smaller than in figure 7.5a. This illustrates a fundamental problem in attempting to create high aspect ratio structures using thermal evaporation / resist bilayer techniques. The resist
Figure 7.2: An AFM showing an array of aluminum towers. The 2 \( \mu \text{m} \) wide line is the gold contact wire.
Figure 7.3: A 3D rendering of the bottom central region of figure 7.2.
Figure 7.4: (a) $1 \times 1 \mu$m AFM image of 80 nm Al towers, (b) $2 \times 2 \mu$m AFM image of 30 nm Al towers.
Figure 7.5: (a) AFM cross sections of a typical Al tower in 7.4a and (b) in 7.4b.
holes for the smaller structures close off due to the finite source size of the Al source and the working distance of the system. In the system used, both the working distance and the Al source size were adjusted to minimize this effect. However, the desired aspect ratio towers at sub-50nm requires a different fabrication approach.

An alternative method for fabricating high aspect ratio metallic structures has been demonstrated by Chou and coworkers [22]. In their work, electron beam lithography is used in conjunction with a high aspect ratio developer to create narrow, deep holes in a PMMA mask. Then, nickel is electroplated through the holes. The PMMA is removed, leaving behind the nickel towers. Towers with an aspect ration of up to 10 have been achieved, with minimum tower diameters of approximately 350 Å. However, it may be quite difficult to adapt an electroplating technique for use in a charging structure. Establishing good junction interfaces is critical to device performance, and there is uncertainty regarding how smooth an electroplated surface is upon termination, oxidation, and then continued electroplating. Also, electroplating does not typically yield very pure metals, which is important to minimize offset charges.

7.3 Small Gaps

7.3.1 Introduction

Another method for studying both Coulomb charging effects at small size scales, and also possibly conduction through polymers, is the fabrication of very small gaps (<50 nm) between two electrodes. Since the gaps may not be quite that small, a gold island deposition may be used to make even smaller gaps in between a fabricated gap. This is illustrated in figure 7.6a. It is expected that the system shown in figure 7.6b will exhibit extremely strong Coulomb blockade effects. The capacitance of an isolated metal island may approach $1 \times 10^{-18}$F, where the single-electron is observable at room temperature.
Figure 7.6: Schematics showing how small gaps between two electrodes, in conjunction with a sub-continuous deposited gold film can be used to examine transport in (a) conducting polymers and (b) tunneling through discrete metal islands. (c) shows a side view of (a).
7.3.2 Gap Fabrication

The substrate chosen is again Si n-type, having a conductivity of 1-10Ω-cm and a thermally oxidized 1000Å SiO2 layer on top. This structure is fabricated using a resist-bridge created with e-beam lithography and a PMMA bilayer. Figures 7.7 and 7.8 show a 2D and 3D rendering respectively of an AFM image of the resist-bridge. Even over a relatively long distance (1 μm), the bridge appears continuous. The fabrication process is shown in figure 7.9. After e-beam exposure, two angled gold evaporation are used to create a gap that is smaller than the resist bridge width (a very useful method of going beyond the minimum feature width obtainable with e-beam lithography)[38]. The bottom of figure 7.9 shows an enlargement of an SEM image showing a fabricated gap. Note the thinner metal near the gap, due to the angled deposition.

The gap electrodes are connected to the photolithographically defined gold lines and contact pads as shown in figure 7.10. Great care must be taken in handling these structures, as static electric discharge can destroy them. Also, care must be taken at the junction of the e-beam and photolithographic metal. Ridges at the edges of the photolithographically defined gold lines can prevent the two wires from contacting. The resistance of a continuous line in the e-beam layer is used as a control to establish that the layers successfully contact each other. This line is not present in figure 7.10, and was added to the e-beam later, in place of the lower-most central gap.

7.3.3 Gap Fabrication Results

This technique can be used to reproducibly create gaps with a variety of sizes, as shown in the SEM images of figure 7.11 (note for this figure and figure 7.12 only a single perpendicular evaporation was performed). Narrow gaps between electrodes as wide as 1 μm have also been achieved (see figure 7.12). Close-up SEM images of the gaps show the metal shadowing from the resist bridge (see figures 7.13 and 7.14). Gaps as small as 10 nm have been fabricated (see figure 7.13).
Figure 7.7: An AFM image of a resist bridge form in a PMMA bilayer.
Figure 7.8: A 3D rendering of the resist-bridge shown in figure 7.8
Figure 7.9: Fabrication diagram for the angle deposition resist bridge process. The lower part of the figure is an SEM of a fabricated gap.
Figure 7.10: An optical micrograph and a diagram showing the metallization in the e-beam field and the contact to the photolithographically defined metal.
Figure 7.11: A series of SEM images showing a variety of gap sizes. The actual gap size is smaller than that in the design (number over the SEM images), due to the proximity effect exposing resist in the gap region.
Figure 7.12: An SEM of a narrow gap between 1 μm thick electrodes, formed using a PMMA resist bridge similar to figure 7.8.
Figure 7.13: A high magnification SEM image of the gap region, showing the metal thickness variations that are caused by the two angled deposition. The gap size is approximately 10 nm.
7.3.4 Gold Island Deposition

To reduce the fabricated gap size, a discontinuous gold deposition is used. Figure 7.15 shows the typical stages of gold film deposition in a thermal evaporator. First, nucleation of small gold islands occurs. Then, the islands grow, until they coalesce. As the thickness increases, the holes between the islands fill in until a completely continuous film is achieved. In order to exploit this process, we require that the deposition be stopped during the time when the islands are growing, before they coalesce. To achieve this, junction resistances are monitored in situ, and the deposition rate used is very slow ($<1\text{Å/s}$).

Figure 7.16a shows an AFM image of an island film in which only 10Å of gold has been deposited. (This is an average thickness determined from a crystal thickness monitor.) Clearly distinct gold islands are apparent. In figure 7.16b, 40Å of gold has been deposited. At this thickness, the beginning of gold island coalescence is observed. It should be noted
1. Initial nucleation

2. Island growth

3. Island coalescence

4. Hole fill in

5. Complete film

Figure 7.15: Steps of metal film deposition.
Figure 7.16: An AFM image of (a) gold island nucleation, and (b) coalescence for gold thermally evaporated onto a silicon substrate.
that the AFM images do not convey the actual detail of the island edges correctly. In practice, the islands will appear larger in AFM images than they are physically as the AFM cantilever tip has a finite radius (on order 100Å). The curvature of the tip broadens features in the line sweep, and in the AFM image.

7.3.5 Preliminary Results

Fabricated gap chips are bonded to 16 bin DIP headers prior to the metal island deposition, to allow for monitoring the junction resistance during the deposition. The deposition is ended when the conduction through the islands approaches a resistance of a few hundred kΩ. Besides gold, platinum and palladium were also used. Figure 7.17 shows a record of the junction conductance versus time for these three metals for large area test junctions. Starting from an initial value after the deposition, the conductance in all three cases decreases while the film is still in vacuum. When the chamber is vented with dry nitrogen gas, the conductance abruptly changes, and then abruptly decreases when the exposed to air. Over time, the resistance of all of the junctions tends to steadily increase. This phenomenon has been observed previously in granular metal films [75]. While the mechanism responsible for these conductance changes in the junctions is not well understood, a combination of both the cooling of the deposited islands and substrate, as well as a metal migration effect, is most likely responsible for the long term resistance increase. This effect makes the realization of a charging structure difficult, because the intra-island distance may keep increasing, and the tunneling current drops exponentially with this distance.

Results from a variable temperature experiment are shown in figure 7.18. The zero bias conductivity versus $1/T$ (figure 7.18b) is similar to results obtained by others in large area granular film experiments [75]. In these systems, transport is governed by thermally activated tunneling, as the charge carrier density is given by

$$n = N \exp\left(-E_a/kT\right),$$

(7.2)
Figure 7.17: Conductance versus time for gap junctions with platinum, palladium, and gold islands.
where $N$ is the total number of charges in the film, and $E_a$ is the activation energy. The activation energy can be determined by fitting the linear regime of figure 7.18b ($1/T < 0.020$) to equation 7.2 which results in an activation energy of 30 meV. As this activation energy is the result of Coulomb charging in the film, it should be on order $e^2/r$, where $r$ is the average particle size. Using this assumption, the particle size in this case is approximately 13 nm, which is close to expected. Neugebauer and Webb[75] obtain very similar results for granular gold films.

Since no single electron charging effects are apparent, and the results are almost identical to that observed in large area granular film experiments, the conclusion is that although we have successfully deposited the thin film, the conduction is not occurring predominantly through the localized region in the gap. This may indicate that the metal in the gap region migrates away from the gap in room temperature. Future work will concentrate on finding a metal or a metal/insulator system that is more stable over time.
Figure 7.18: (a) A series of I(V) curves for a gap structure with gold islands. The lowest resistance trace is taken at 300K. Subsequent traces are shown every 20K until 20K. (b) A plot of the log of the conductance (for the traces in (a)) versus inverse temperature.
Chapter 8

Vertical Conduction in Thin Si/CaF$_2$/Si Structures

8.1 Introduction

There exists a very real economic incentive for transferring newly discovered quantum electronic effects seen in III-V heterostructure systems into a silicon-based material system. The work that is described in this chapter is the beginning of an attempt to achieve resonant tunneling structures in a heterostructure system based in silicon/CaF$_2$.

The silicon - calcium fluoride system has long been considered as a candidate for electrical isolation of silicon, especially in the area of 3D device integration.[77] The relatively small 0.6% lattice mismatch at 300°C makes CaF$_2$ very promising. Additionally, CaF$_2$ has the attractive property that it may be patterned directly with electron-beam and ion-beam lithography.[69,103] However, little attention has been given to the transport properties of the Si-CaF$_2$ heterojunction.

To date, much of the work in this system has either focused on very thick layers (i.e., gate dielectrics, >1000Å)[91] or very thin layers (i.e., a few monolayers of CaF$_2$). Thin layers of CaF$_2$ on Si have been studied using a variety of techniques, including Auger spectroscopy
and core level photoemission[84, 51], medium-energy ion scattering[104], resonant three-wave-mixing spectroscopy[49], and transmission electron microscopy.[28,9] The emphasis of this work has been on understanding the electronic and structural properties of the Si/CaF$_2$ interface. Almost all of this work is concerned with exposed CaF$_2$ layers on top of Si, rather than on a CaF$_2$ layer buried in Si. Very little attention has been focused on the intermediate regime of a 100-1000Å layer of CaF$_2$ on Si.

It has been observed that CaF$_2$ grows most readily on Si(111) oriented substrates.[88] On Si(111), A and B phase CaF$_2$ growth can occur. Most groups have only observed the growth of B phase.[3,78,65] Only limited evidence for thin layers of A phase CaF$_2$ on silicon exists.[9] We have found that we can grow exclusively A phase CaF$_2$ material by ramping the substrate temperature during growth.[20] The ability to grow A phase CaF$_2$ may be crucial to achieving good quality films. Figure 8.1 schematically shows A phase CaF$_2$ and B phase CaF$_2$ grown on a stepped Si(111) surface. A Si substrate step in A phase growth can be accommodated, and only creates a few dangling bonds. However in B phase growth, the 180° rotation of the CaF$_2$ lattice in the $<111>$ direction creates line defects at every step site in the silicon substrate.

The growth of thin CaF$_2$ layers on silicon, and subsequent epitaxial regrowth of silicon, allows the possibility of a heteroepitaxial system in silicon, making heteroepitaxial devices realizable. A few groups have investigated Si/CaF$_2$/Si structures[4, 33], however, the thickness of the CaF$_2$ layer that was used was rather large (3000-5000Å), which precludes making electron tunneling measurements through these CaF$_2$ barriers. Given the large conduction band offset of around 2.5eV[51], the CaF$_2$ layers will have to be either thin enough to permit substantial penetration of the electron wave function, and/or possess a light electron effective mass in order to observe a reasonable tunneling current.

In this work, I have examined the simplest possible structure in the Si/CaF$_2$/Si heteroepitaxial system, a single thin barrier of CaF$_2$ in n-type Si. Ideally, by examining electron transport through CaF$_2$ barriers of different thicknesses, the conduction band offset and the
Figure 2: a) Schematic of the A-phase CaF$_2$ on a stepped Si(111) surface and b) B-phase CaF$_2$ on a stepped Si(111) surface. Note the accommodation of the step in the Si(111) surface in the A-phase CaF$_2$ versus the gap that forms in the B-phase CaF$_2$. 
CaF₂ electron effective mass may be determined. The values obtained in a Si/CaF₂/Si heterostructure for the conduction band offset should be more reliable than those obtained from a measurement on a few monolayers of CaF₂ on top of Si, as the properties of the interface may change substantially when additional monolayers of CaF₂ are present.

8.2 Growth and Fabrication

The single barrier samples studied were all grown in a VG molecular beam epitaxy (MBE) system. As illustrated in figure 8.2, starting with an N-type Si(111) substrate, a passivating epitaxial layer of n⁺Si, 2000Å to 1 μm thick, was grown via chemical vapor deposition. The sample temperature was then either ramped from 100°C to 600°C for MBE growth of A phase CaF₂, or held at 700°C for growth of B phase CaF₂. The phase of the CaF₂ was confirmed through x-ray diffraction. A 2000Å-5000Å top layer of n⁺ silicon was then deposited either via CVD. Depending on the growth temperature, the top silicon layer was either single crystalline or polycrystalline.

Devices were fabricated using conventional photolithography and lift-off to create aluminum mesas which were used as self-aligned etch masks. Mesa size ranged from 8x8μm²
to 256x256μm². Reactive ion etching (RIE) was used to remove the top level of silicon. Both SF₆ and CF₄ type reactive ion etching were used, although SF₆ gave better results in terms of creating devices with lower leakage. CaF₂ was observed to serve as an excellent etch stop for SF₆ reactive ion etching. Electrical contact was made from the mesa top to the backside.

8.3 Experimental Results

A series of devices were studied to determine differences in the electrical quality of A versus B phase CaF₂. Nominal identically structures were grown with CaF₂ barrier layers 100Å, 200Å, and 500Å thick. In these devices, the top silicon layer was polycrystalline. As anticipated, the B phase devices exhibited far worse characteristics than the A phase devices, with currents several orders of magnitude higher for identical size devices. This is most likely due to the inability of B phase CaF₂ to compensate for steps in the Si surface, resulting in line defects and pinholes occurring everywhere a step in the Si substrate occurs. Additionally, at the growth temperature for B phase CaF₂ the lattice mismatch is 2.1%, which is much higher than the value of 0.7% where A phase growth is started.

Figures 8.3 and 8.4 show experimental current versus voltage characteristics for 100Å and 200Å layers of A phase CaF₂ respectively for four different size devices (256x256μm, 128x128μm, 64x64μm, and 32x32μm). Several features are apparent. First, an irrecoverable breakdown was suffered at a field of about 3x10⁶ V/cm (3 volts) in the 100Å sample, and at around 1x10⁶ V/cm (2 volts) in the 200Å sample. We believe that this trend of lower breakdown fields in thicker layers is consistent with increasing defect formation due to the increased strain in the thicker layer. The smallest device measured in the 100Å CaF₂ layer exhibited no breakdown until a field of about 7x10⁶ V/cm, which is consistent with the dielectric breakdown of CaF₂[16], rather than a breakdown process via defects. This implies a defect density of one per 1000μm² in the 100Å A phase CaF₂.
Figure 8.3: Current versus voltage characteristics for 100Å A-phase CaF$_2$ barrier device for different area measurements. $T = 300$K.
Figure 8.4: Current versus voltage characteristics for 200Å A-phase CaF$_2$ barrier device for different area measurements. T = 300K.
Although the results in the A phase devices in the low bias regime are more promising than in the B phase devices, they are still not consistent with tunneling through an epitaxial layer. This is seen by considering the 100Å and 200Å devices. The currents scale with area in the lower bias regime, as expected. However, as shown in figure 8.5, theoretical calculations of the tunneling currents in Si/CaF$_2$/Si structures with thicknesses of 100Å and 200Å show currents that are nine orders of magnitude different in the lower bias regime, while the experimental curves for both the 100Å and 200Å device have currents that are on the same order of magnitude. Assumptions made in the theoretical tunneling current model are a conduction band offset of 2.5eV between CaF$_2$ and Si, and an effective mass of 0.02 for electrons in CaF$_2$. This choice is somewhat arbitrary. However, the situation does not improve by picking a larger effective mass value. One reference [29] places an upper limit of 0.2 on m$^*$ in CaF$_2$. The calculations involve the use of a three band model (one band for each layer), thus taking into account the effective mass discontinuity at the Si/CaF$_2$ interface. The current density equation[47] that is numerically integrated is

$$J_x = \frac{-4\pi e kT m}{\hbar^3} \int_0^\infty \ln \left( \frac{1 + \exp\left(\frac{E_F - E_x}{kT}\right)}{1 + \exp\left(\frac{E_F - V(x) - E_x}{kT}\right)} \right) \exp \left( \frac{-4\pi}{\hbar} \int_{S_1}^{S_2} \left(2m(V(x) - E_x)\right)^{1/2} dx \right) dE_x,$$

where $J_x$ is the current density in the vertical direction, $E_F$ is the Fermi level, $E_X$ is the energy of the electron in the direction of tunneling, $V(x)$ is the potential barrier (CaF$_2$), and $S_1$ and $S_2$ are the spatial locations of the beginning and end of the barrier. This model ignores the depletion/accumulation effects on the conduction band, as it was used only to illustrate how different the current through these two layers should be.

Devices were also fabricated in a 100Å A phase CaF$_2$ barrier structure that was grown with a single crystal silicon top layer. The current versus voltage and temperature characteristics for a 100Å A phase Si/CaF$_2$/Si structure is shown in figure 8.6. Figure 8.7 shows the same data in a three dimensional view. Ideally, transport through the barrier should be governed by direct tunneling at low applied voltages, and Fowler-Nordheim tunneling [96] at higher voltages, where the discontinuity in the current versus voltage curve marks
Figure 8.5: Theoretical current versus voltage curves for 100Å and 200Å CaF$_2$ barrier devices. $T = 300$K. Experimental data shown (solid line) for 100Å A-phase CaF$_2$. 
the crossover between these two regimes. However, variable temperature measurements indicate that this kind of transport does not occur in our devices.

As can be seen in figures 8.6 and 8.7, the higher bias regime has a much different temperature dependence than that of the lower bias regime, which should not be the case if transport is occurring via tunneling through an epitaxial layer of CaF$_2$. The only temperature dependence in the tunneling current comes from the number of electrons at a given temperature available to tunnel, given by:

$$N(T) = 2 \left( \frac{2\pi m_0^* kT}{\hbar^2} \right) M_C \exp \left( \frac{-(E_C - E_F)}{kT} \right)$$  \hspace{1cm} (8.2)

Since at a given temperature the number of electrons, $N(T)$ is a constant, changes of $N(T)$ that are temperature induced will affect both direct tunneling and Fowler-Nordheim tunneling in the same way. Therefore, they should both have approximately the same temperature dependence.

A subsequent fabrication of these devices was done, identical to before, except now a 1000Å indium layer was deposited on top of the aluminum contact. During biasing of the device, it is observed that in the higher bias regime, voids in the indium layer formed on the order of one per every 900μm$^2$. This value is consistent with the value seen in the previous 100Å A phase CaF$_2$ devices. These appear to form as a result of the electromigration of indium, most likely caused by locally high current densities through pin-hole defects in the CaF$_2$ layer. Figure 8.8 shows a contact before and after biasing. A control experiment was also performed, where the probe tip that contacts the indium/aluminum pad directly contacts the aluminum layer on bottom. Similar results were obtained, indicating that the effect was independent of whether or not the current was actually flowing through the indium layer.
Figure 8.6: Current versus voltage characteristics of a 100Å A-phase CaF$_2$ barrier, with a single crystal top Si layer at various temperatures.
Figure 8.7: Current versus voltage and temperature characteristics of a 100Å A-phase CaF₂ barrier, with a single crystal top Si layer.

Figure 8.8: Indium/Aluminum metalization before (left) and after (right) bias.
8.4 Conclusions

We have fabricated thin Si/CaF$_2$/Si junctions in both A and B phase CaF$_2$. It has been shown that the electrical properties of A phase CaF$_2$ barriers are superior to those of B phase CaF$_2$. However, even in the A phase material studied, conduction through the CaF$_2$ layer is dominated by conduction through defects. Before this system can be useful for creating heterostructures, more work will have to be done investigating the improvement of the CaF$_2$ material quality, especially in regards to reducing pin-hole defects. The use of CaF$_2$ as a gate dielectric may have more promise as recent results indicate a radiation hardness that is a factor of two better than SiO$_2$.[41]
Chapter 9

Summary and Conclusions

In this section, I will summarize the major results of this dissertation. In Chapter 3, three major experimental systems that I developed are discussed: the Yale electron-beam lithography system, the long working distance evaporator, and the ultra-low noise electronics. In Chapter 4, the electron spectroscopy of finite period minibands in GaAs/AlGaAs superlattices is observed to reveal the presence of the individual eigenstates that form the miniband. This is observed for both inter-miniband tunneling and for the first time in intra-miniband tunneling. In Chapter 5, extensive electron spectroscopy studies on InGaAs quantum dots are discussed. The magnetic field dependence of the current plateau edges conclusively shows that these plateaus are due to laterally localized states in the quantum well. The diamagnetic shift of these states is used to determine the lateral extent of the dot states. From this, the model of conduction through \( N \) dots in parallel is developed. Additionally, the current suppression in perpendicular field is measured in detail for the first time in this system, and found to be consistent with a decreasing overlap between the emitter and dot wavefunction. For Chapters 5 and 6, samples were grown at the Central Research Laboratory of Texas Instruments. Theoretical support for both of these projects was provided by Prof. William Frensley’s group at the University of Texas at Dallas.

The work in Chapter 6 is largely motivated by the challenges of fabricating the quantum
dot structures discussed in Chapter 5. This work is the result of very close collaboration with Roger Welser (see his dissertation for more details concerning InAs nucleation\cite{110}).

This is the first result of its type, a method that uses high-dosage electron-beam lithography to inhibit the nucleation of submicron crystallites. Chapter 7 discusses attempts to create two types of novel charge quantized structures. While some of the fabrication techniques developed are impressive (especially the reproducible 100Å gaps), there is still further work required to realize charge quantization. Finally, Chapter 8 shows the results of a study that attempts to realize a heterojunction system in Si. Using both A and B phase CaF$_2$ as the barrier, it is observed that the A phase material has better electrical properties. These results were the first published on transport through A phase films of CaF$_2$. However, conduction through pinholes still dominates, and further work on improving the material is required before devices with reasonable electrical properties may be fabricated.
Bibliography


