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Dynamics of Few Electrons in Self-Assembled Coupled Quantum Dots

A Dissertation submitted in partial satisfaction of the

requirements for the degree Doctor of Philosophy

in Physics

by

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DEDICATION

While it is debatable whether terahertz (THz) technology will be useful for any particular application, e.g., quantum information processing (QIP), it is clear from current trends that more general applications are approaching from the horizon. Conventional computer processor clock speeds continue to increase into the low GHz at the time of this writing, and no end of demand is in sight. Frequent computing industry projections regarding characteristic device sizes, electric fields, numbers of charges per bit, heat dissipation etc., and their respective impending fundamental challenges, have become cliché. Still, there is clearly a technological frontier opening up, both in terms of THz science and QIP, that will eventually be used to solve these looming crises.

More generally, though, one should step back and reflect on the needs and desires of our actual, root employers: the taxpaying public of the United States of America. In that respect, it is not at all clear that our driving goal should be the exploitation of undeveloped bandwidth, or the creation of machines that make unbreakable codes, however the proxies of the public may claim that this should be the case.

In the end, public decisions are often based as much, if not more, on æsthetics as on logic. And when we communicate to the public or its proxies, our core motivating factors, our driving visions come through. It is the author's firm belief and experience that it is of real, practical benefit to remind ourselves, our students, our colleagues and all facets of the outside world of the æsthetic value of our work. When we hone

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that vision, and broadcast it clearly, blow as the political and economic winds might, some fraction of our employers exist who will hear, who will listen, who will believe.

To wit:

We are building new worlds, bit by bit. We are manipulating artificial atoms and man-made molecules in fantastic crystal structures where the very physical constants of our world – masses, permittivities, etc. – are effectively changed. By filling this THz technology gap, we will be able to speak and hear in languages long used by the natural world, but to which we have been largely deaf and dumb, even today. In QIP is the promise of understanding the fabric of nature by thinking the way nature thinks, on a more fundamental level than ever before. We have been given the tools to make artificial atoms; it is only natural that we use these same tools to fill out the periodic table. To these noble ends is focused the author's vision of this work.

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Studies in Far-Infrared Spectroscopy of Coupled Quantum Dots

Studies in Characterization of Charge Loading and Storage in Quantum Dots

Studies in Mid-Infrared Spectroscopy of Stored Charge in Quantum Dots

ABSTRACT

Dynamics of Few Electrons in Self-Assembled, Coupled Quantum Dots

by

Kulvinder Singh Gill

An experimental approach to the study of various lifetimes of charge states in self-assembled, coupled and uncoupled quantum dots is presented. Using conventional methods of tunneling charges into ensembles of coupled quantum dots, we explore far-infrared absorption in, and saturation of, electric-field-tunable transitions between bonding and antibonding quantized states. To develop a framework for the study of such dynamics while independently controlling the quantum dot charge density, we present a method of charge storage in ensembles of uncoupled quantum dots, independent of tuning electric fields. This method includes detection of stored charge via electrical means. Finally, we use this method to explore mid-infrared dynamics of stored charge in uncoupled quantum dots.

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1 Introduction

Semiconductor technology is rapidly approaching two major physical challenges: the terahertz technology gap, and computation on the quantum scale. The terahertz range of the electromagnetic spectrum, which bridges between, on one side, the microwave frequency range used in high-speed electronic circuitry, and on the other, the near-infrared frequency range used in optoelectronics, is conspicuously poor both in terms of sources and detectors, lasers and cameras. Quantum-scale computation is not only inevitable as devices shrink in scale, but also holds the promise of solving problems, both computational and social, that are practically impossible for both the non-quantum technology of today, or any of the future. This project addresses pieces of both issues, by investigating a potential method of quantum computation using terahertz technology. Specifically, we are working on placing single electrons into orbit around artificial atoms, and manipulating those electrons with terahertz frequency radiation. Our vision is to use electrons in these orbits to perform elementary quantum computations, while working in conventional semiconductors to allow rapid scaling of this technology for real-world applications.

1.1 Summary

Here we present an experimental approach to the study of various lifetimes of charge states in ensembles of self-assembled, coupled and uncoupled indium arsenide (InAs) quantum dots. Our work had two main goals: to study and test current approximations of the complex energetic landscape that defines these quantum dots,

and to investigate them as components of a proposed quantum information processor. First, we constructed ensembles of coupled quantum dots in a conventional metal-insulator-semiconductor field-effect transistor-like growth structure allowing simultaneous control of the charge density (σ) and growthdirection electric field (E_z); this coupling of the quantum dot electronic states was evidenced by our observation of far-infrared absorption, tunable via E_z -induced Stark shift. A single parameter controlled σ and E_z . Due to the range of E_z needed to induce electronic coupling, these transitions were necessarily executed in systems with $\sigma \sim 6$ electrons per quantum dot[1]. Through a far-infrared saturation experiment, we deduced a lower bound on the dephasing time of approximately 40 ps[2]. We expect a much longer possible dephasing time limited by acoustic phonons, so an investigation of transitions between single-electron states was sought.

For such an investigation, we developed a novel scheme, using a dopedinsulating-doped structure and a combination of near-infrared laser pulses and voltage biases, to write, store, and read charges in ensembles of quantum dots. This procedure allows manipulation of E_z during the storage time. We characterized this storage procedure, and found very long storage times, 10 seconds and greater, at relatively high temperatures, 80 K and greater. Finally, we put this novel device to use by observing the mid-infrared spectroscopy of charges in the quantum dots during this cycle by using the sample itself as a mid-infrared detector.

1.2 Motivation

There were two major motivating factors behind our study of the dynamics of few charges in self-assembled InAs quantum dots. The first was to test several models of electron and hole states in quantum dots. The second was to probe the various lifetimes of such states for their applicability to quantum information processing.

1.2.1 Models of Charge States in Quantum Dots

Though the literature is replete with experimental spectroscopic characterizations of InAs quantum dots[3-10], a combination of factors makes theoretical predictions of the energetic landscape challenging at best[11-16]. First, due to the Stransky-Krastanow growth mechanism, the size and shape of InAs quantum dots grown on GaAs, though somewhat controllable, vary widely[14]. Also, the InAs/GaAs alloy composition is known to vary throughout the quantum dot region[17]. Finally, the lattice strain that initiates quantum dot growth also alters estimates of the bandgap, effective masses and other parameters[14].

Nonetheless, by estimating the size, shape, alloy, and strain, several models can be identified[11-17]. Further, recent experimental advances in mapping projections of self-consistent quantum dot potentials have given even greater insight to aid theory[18]. We sought to expand the toolbox available for quantum dot characterization. With our system, we investigated the possibility of MIR and FIR spectroscopy of ensembles of quantum dots, while varying σ and E_z independently in-situ.

1.2.2 Investigating Quantum Dots for Quantum Information Processing

Various approaches have been pursued over time for specific kind of implementation of a quantum information processor[19]. This device would consist of quantum bits (qubits) realized as either InAs quantum dots or impurity-quantum dots in GaAs, with internal energy spacings in the FIR. They would be isolated from the environment by their location within a solid-state resonator, possibly a defect cavity within a terahertz (THz) photonic bandgap crystal. Due to the long wavelengths associated with THz radiation, the cavity would be large compared to the quantum dots and would house perhaps thousands of qubits. The individual qubits would then be individually addressed via interband NIR lasers. By using the ac Stark effect of the strong NIR lasers, the energy spacings could then be tuned into and out of resonance with the cavity mode, allowing coupling of the individual qubits with the population state of the cavity mode. By simultaneously manipulating separate gubits within the same cavity via separate NIR beams, one could, in principle, perform two-qubit logic operations. This would provide the necessary components of a quantum information processor in the solid state, capable of performing arbitrary quantum computations, while remaining scalable through quasi-

conventional solid state processing techniques[20]. A sketch of such a processor is shown in Figure 1.2.2-1.



Figure 1.2.2-1 A conceptual drawing of a model THz Quantum Information Processor. Note interband lasers in red, addressing individual quantum dots in photonic bandgap crystal defect.

There are three important parameters for today's models for cavity-based quantum information processors, irrespective of physical implementation: the maximum operation rate, often related to the Rabi frequency of the system; the leakage rate, which measures how often the system interacts with the environment, allowing you to observe the state; and the dephasing rate T_2^{-1} , which measures the

rate at which the internal state of the system decays. One would prefer that the various rates be ordered such that: (Rabi frequency) > (leakage rate) > $(T_2^{-1})[21]$.

In our proposed system, the parameter of greatest concern is T_2 . A very direct way to measure T_2 in an ensemble of quantum dots would be via a photon-echo experiment[22]; unfortunately that is beyond the scope of this work.

Instead, we have focused on constructing a system which allows us to maximize T_2 in certain respects. For electronic states in solid-state systems, coupling to phonons is problematic[19]; therefore we have attempted to avoid optical phonons by seeking systems with energy spacings below the longitudinal optical (LO) phonon limit. For ease of growth and processing, we chose to work with GaAs, so $hf_{LO} \approx 36$ meV. Measurements are thus in the THz frequency region (2.5 THz \approx 10 meV), which has other advantages described above. Energy spacings in single InAs quantum dots are typically 40 meV or greater[1], so we focused more on the bonding-antibonding transitions of coupled (§2.1.2) quantum dots. These transitions are polarized in the growth direction, so for coupling to occur, the individual dot levels must be tuned into resonance by the choice of an appropriate Ez;; the same Ez can tune the resonance itself within some range. Since T_2 may be sensitive to electron-electron scattering within the coupled quantum dots, we aim to have a wellcontrollable σ , preferably ≤ 1 electron/dot-pair. Hence our search for separate, independent control of E_z and $\sigma.$

2 Previous Work

In order to appreciate this work and its place in the field, it is instructive to first review some significant accomplishments by other workers. Specifically, we will briefly discuss quantum dots in general, coupling them with one another, a few methods of loading charges into them, and typical techniques of quantum dot midinfrared spectroscopy.

2.1 Quantum Dots, A Brief Overview

For our purposes, a quantum dot can be defined as a system that confines charges – electrons, holes or both – in all three dimensions. Further, the quantization energies imposed by the confinement in each of these dimensions are greater than both the thermal energies of the system, and the width in energy of the individual states. In such a system unperturbed by external fields, the entire state of the system will eventually be determined by the number of carriers stored in the dot[23].

2.1.1 Various Quantum Dot Systems

The most well-studied quantum dot systems are atoms; indeed, quantum dots are often called "artificial atoms." With their three-dimensional Coulombic potential and extremely sharp linewidths, atoms and ions pose natural choices as candidate systems for quantum information processing (QIP)[24]. The challenge is keeping the

atoms isolated from the environment, to preserve their beautiful spectral characteristics, typically in the optical frequency regime.

Atomic impurities in crystals are similar to atoms in many respects. Specifically, our group has built on prior studies of donor impurities in GaAs. Electrons bound to these donors behave quite analogously to hydrogen atoms, with a few added complications from phonons and nuclear spin interactions. Since the energetics are rescaled by the bulk semiconductor properties, the relevant energies are once again in the 10 meV range, and much of the prior discussion (§1.2.2) holds[25].

Also heavily studied are lithographically-defined quantum dots[26]. Usually, they consist of metal gates judiciously deposited on top of a semiconductor twodimensional electron gas (2DEG). The gates, when properly biased, define depletion regions in the 2DEG which then confine electrons in the un-depleted region. In fact, the gates can also be used to carefully inject and remove electrons from the quantum dots. Because of the energetics of the 2DEG and the size limitations on lithographical confinement, the dynamics require temperatures in the range of 10^{-3} K.

A local perturbation in the crystal strain in a quantum well, usually resulting from a buried self-assembled quantum dot (§2.1.1), can cause charges to localize in the region of the strain, resulting in a strain-induced quantum dot[27].

Nanometer-scale particles of semiconductor, e.g. CdSe, result in regions where conduction band electrons can exist bounded by large three-dimensional potential – the surface of the particle itself. These particles, in order to prevent aggregation and unintentional quantum dot-quantum dot interaction, are often coated with polymer

ligands, allowing their suspension in a solution. These colloidal quantum dots are under intense study for their applications as biological markers[28], and polymeric photonics[29], not to mention QIP. A few issues with colloidal quantum dots are that their very isolation makes difficult the injection of charge and that the ligands have time-dependent perturbing effects on the confining potential.

This list of types of quantum dots is by no means exhaustive, but is quite representative.

2.1.2 Self-Assembled InGaAs Quantum Dots

A 7% mismatch exists between the lattice constants of GaAs and InAs[23]. When InAs is deposited on GaAs, a few monolayers of highly-strained InAs is initially grown – a so-called "wetting layer." If GaAs deposition is resumed at this point, the result is a strained InGaAs quantum well bounded by GaAs (InGaAs/GaAs QW). The QW is InGaAs instead of InAs due to diffusion of Ga into the well. If, instead, InAs deposition is continued, eventually, under the proper growth conditions, the InAs layer relieves the strain by de-wetting, forming threedimensional mounds of InAs on top of the wetting layer[30]. This is the so-called "Stransky-Krastanow" (SK) growth mode. GaAs growth is then resumed, resulting in an InGaAs quantum dot, bounded by a GaAs "capping layer" on top, and a the narrow InGaAs effective QW of the wetting layer. InGaAs/GaAs QWs are type I heterostructures, resulting in two-dimensional confinement of both electrons and holes. Similarly, InGaAs/GaAs quantum dots confine both carriers in threedimensions. Henceforth in §2.1 we will refer to InGaAs/GaAs quantum dots as "InAs quantum dots," and afterward as simply "quantum dots."

Inherent in the SK growth mode, the InAs quantum dots nucleate at random sites along the plane of the wetting layer, where the average density of dots can be controlled by growth conditions. In certain cases, including pre-patterning of the initial GaAs surface, the nucleation sites of quantum dots can be directed[31].

Also, the InAs quantum dots vary in size and shape[32], resulting in a spread of about 10% in the various dimensions. Again, by careful control of the growth conditions, certain variations can be minimized. For example, if the capping layer growth is interrupted, the dots may be only partially covered by GaAs. Then, the growth conditions may be altered so as to evaporate the yet-uncovered InAs. The GaAs growth is resumed, resulting in quantum dots with uniform heights. This procedure is referred to as the "cap and flush" technique[33].

If the capping layer over one layer of InAs quantum dots is sufficiently thin, a strain field will propagate through to the GaAs surface. If another layer of quantum dots is then grown on this GaAs surface as described before, the strain field perturbation from the first layer will incite the nucleation of the second layer at sites highly correlated with the first layer. In fact, this will result in a two-dimensional layer of vertically-aligned InAs quantum dot pairs. If the intervening GaAs layer is sufficiently thin, coupling of the electronic and hole states may occur. This is the coupling referred to in §1.2.2[34].

Several factors greatly complicate the prediction of the bound states of InAs quantum dots. The first is the fluctuation in both size and shape described earlier. The second is the related problem of the varying indium vs. gallium concentration in the quantum dot. The final factor is the very complex influence of strain. All of these factors combine to make the potential profile, and hence the (bare) bound states of InAs extremely difficult to predict, though many workers have had measured success using a variety of simplifying assumptions and atomistic models[11-17].

Also, many experiments include multiple charges in each quantum dot, usually a combination of electrons and holes; a self-consistent solution to both the Schrödinger and Poisson equations is required. Here, again, valiant efforts have been made, with varying degrees of success[12, 17].

2.2 Quantum Dot Coupling and Tuning

Since the goal of this work is the study of coupled quantum dots, it is instructive to review the elementary aspects of the theory of coupling, along with the deviations from such a simplified model, and various supporting observations.

2.2.1 The Ideal Double-Square-Well Model

To get a feel for the behavior of the energy states of a coupled quantum dot system, it is useful to consider a model with some simplifying assumptions. Undoubtedly, many of the following assumptions are not strictly necessary for our conclusions, but the full generalization is beyond our scope. We will assume that the full Hamiltonian and final three-dimensional wavefunctions are separable, and for the study of coupling will only consider the potential and wavefunction profiles in the growth- or *z*-direction, since that is the primary direction of confinement. We will assume *z*-confinement such that there is only one bound state. We will neglect differences in material properties (e.g., effective masses).

First, consider a square potential well with width a (nm) and finite depth V_0 (eV). The ground state of this potential is the well-known oscillatory function within the well and the exponential decay in the barriers. When the well is shallow enough, a single bound state solution can be approximated

$$\Psi(z) = \begin{cases} D \exp(\kappa z) \\ C \cos(kz) \\ D \exp(-\kappa z) \end{cases} \begin{cases} z < -\frac{a}{2} \\ |z| < \frac{a}{2} \\ z > \frac{a}{2} \end{cases}$$
$$k = \sqrt{\frac{2m\varepsilon_1}{\hbar^2}}; \kappa = \sqrt{\frac{2m(V_0 - \varepsilon)}{\hbar^2}}$$
$$V_0 < \frac{\pi^2 \hbar^2}{2ma^2} \Rightarrow \varepsilon \approx V_0 (1 - \frac{V_0 ma^2}{2\hbar^2})$$

Equation 2-1

Where m is the mass of the electron in the material and ε is the energy of the single bound state. D and C are constants that can be found by normalization and boundary-matching conditions. The last condition on V₀ determines one bound state[23]. Next, consider two identical wells as before, with z-axis separation between them b (of nm magnitude). As long as b is finite, the exponential behavior of the wavefunctions in the barrier regions ensure that the above wavefunctions will not describe energy eigenstates of the system, i.e. coupling will occur. This is to say that the ground state wavefunction of each well will be perturbed by the presence of the other well. Under our previous assumptions, in fact, the ground states of the combined system can be described as linear combinations of the above single-well ground state wavefunctions:

 $\psi'(z) = \alpha \psi(z - z_1) + \beta \psi(z - z_2)$

Equation 2-2

With associated energies:

$$\begin{split} \varepsilon_{\pm} &= \varepsilon \pm \frac{c}{1 \pm f} + \frac{d}{1 \pm f} \\ c &= \langle \psi(z - z_2) | V_0(z - z_2) | \psi(z - z_1) \rangle; \\ d &= \langle \psi(z - z_2) | V_0(z - z_1) | \psi(z - z_2) \rangle; \\ f &= \langle \psi(z - z_2) | \psi(z - z_1) \rangle \end{split}$$

Equation 2-3

Where the last, overlap term, f, is often small and therefore neglected[35].

Clearly, the energy difference between the symmetric (bonding) and antisymmetric (anti-bonding) states, which varies as c, is exponentially dependent on b. To generalize one step further, consider an electric field E_z applied in the zdirection between the wells, yielding a perturbing potential $V_1(z) = z \cdot E_z$, and a resulting potential difference between the wells $\Delta V \approx (a + b) \cdot E_z$. As long as $\Delta V \ll V_0$ and $a \cdot E_z \ll V_0$, the resulting states are still well-approximated by linear combinations of the original ground state wavefunctions. Naturally, in the limit of small $\Delta V \ll c$, we return to the prior, zero-field description. In the limit of $\Delta V \gg c$, we return to the original, single-well descriptions for each well, with small "contaminations" from the other well.

From this discussion, we see that, by manipulating the electric field within some range in this toy system, we can drastically sweep the bonding-antibonding energy splitting. This very tunable aspect of coupled quantum dots draws us, as we search for a solid-state implementation of quantum computing. The next step for such an inquiry would be to observe such a tunable transition, and measure its various lifetimes. In this discussion, though, we've glossed over some complicating factors.

2.2.2 Deviations from the Ideal Model

The first practical deviation from the preceding picture is the size of quantum dots. Since the growth dimension can be carefully controlled (§2.1.2), we refer mainly to the lateral dimension. As it happens, the strain field propagation that causes vertical alignment of quantum dots may contribute to the observed lengthening of the lateral dimensions of subsequent layers of quantum dots. Such broadening can be seen in electron microscopy. This lengthening is not well-controlled, and results

in a slight lowering of the top quantum dot's ground state energy with respect to the bottom quantum dot[36].

The shape of a quantum dot is often assumed to resemble a pyramid, a lens, or a hemisphere by various workers. The most precise measurement of quantum dot shape is performed by AFM, but that is restricted to un-capped quantum dots, and so has limited applicability. Cross-sectional microscopy such as SEM and TEM are limited not only by resolution and contrast but also by the ability to make repeated, precise slices of the same dot or similar dots[37].

A related concern is the uncertainty of the exact alloy composition of quantum dots. During the growth process, there is some intermixing of gallium and indium into the InAs and GaAs regions, respectively. Even within the quantum dots, the indium concentration is observed to vary. This process makes the approximation of well-defined, high-contrast potential walls troubling at best[38].

The problem of calculating the effects of strain on the quantum dot's properties is complicated by the uncertain shape – for the sake of boundary conditions – and composition – the source of strain in the first place – but it is also a challenging issue in its own right. Even for bulk, strained InAs grown on GaAs, there are gaps in typical cataloged parameters, especially at low temperatures[13, 39].

Finally, there is the issue of population. In the previous section we analyzed bare-potential wavefunctions. For populated quantum dots, the problem must, of course, be solved self-consistently, using the Poisson and Schrödinger equations. This is not the end of the story, though. Consider E_z , introduced in the last section.

We confined this field to exist only between the wells, because if it exists everywhere uniformly as an imposed field might, the steady-state solution would be for all charge to (eventually) tunnel out of both dots. So stored charge has a finite lifetime, for non-zero E_z .

Similarly, since the wavefunctions are altered due to extant charge, one would prefer to tune the bonding-antibonding transition under a constant charge density, to measure any dynamics as a function of field alone. This idea is a significant motivating factor for this work[2, 12].

2.3 Loading Schemes

To be sure, much interesting physics has been explored, and continues to be explored, using neutral quantum dots[35, 40]. Indeed, some quantum computing approaches have even been developed using only excitons in otherwise-bare and neutral quantum dots[41]. However, charged excitons are very interesting animals, and so are singly- and muliply-charged quantum dots[42, 43]. Since we are interested in *intra*band dynamics, the successful loading of charges into, and storage of charges within, quantum dots is very important to us. It is instructive, then, to look over others' various means of quantum dot loading.

2.3.1 MISFET Structures

Consider a metal-insulator-semiconductor field-effect transistor (MISFET) structure. Such a device could be realized in three stages: first grow a doped GaAs back-gate (BG), then an insulating GaAs layer, and then deposit an aluminum Schottky front-gate (FG) on top. This will form a diode, with a turn on voltage of approximately 0.6 V on the FG in reference to the BG (V_{FG}). If a layer of quantum dots is grown within the insulating layer, sufficiently close to the BG, V_{FG} will control the relative voltage of the quantum dot layer and the BG (V_{QD}). In fact, these two voltages are proportional; this is the so-called "lever arm" effect. Control of V_{QD} allows the tunneling of charge between the BG and the quantum dot layer. This charge tunneling back and forth only occurs when the Fermi level of the BG is resonant with an energy state in the quantum dot layer; this rate is proportional to the density of states of the quantum dot, at the resonant energy. Such charge tunneling can be measured via capacitance-voltage spectroscopy (CV)[3]. In this manner the quantum dot density of states can be mapped out, and the quantum dots can be filled with charges sequentially, allowing for various types of spectroscopy (near- and mid-infrared, magneto-optical, etc.) as a function of charge density. Some researchers have even been able, using a transverse magnetic field and Fourier analysis, to map out the self-consistent wavefunctions at various charge densities[44].

Effective as this method has been, it is not quite suited to our purposes. Namely, the charge density is completely determined by V_{FG} , which simultaneously controls E_z , so independent control is not possible. Hence, though we ourselves have done some work using such a method (§3), we must look elsewhere.

2.3.2 Resonant Interband Excitation

Several schemes for storing charges in quantum dots have been developed using near-infrared (NIR), or interband excitations. The few I will describe here are: QW X-valley transfer[27], resonant selective diffusion and neighbor ionization[45], and resonant quantum dot excitation and tunneling[46]. Many other techniques appear in the literature[47-60], but these are representative.

The structure required for QW X-valley transfer is, in essence, an AlAs barrier layer sandwiched between a layer of quantum dots on one side and a GaAs QW on the other. The strain field from the quantum dots propagates through the thin AlAs to form strain-induced quantum dots (SIQDs) in the GaAs QW. Excitons are resonantly excited in the SIQDs, while a bias is maintained to drive electrons toward the barrier. AlAs has an X-valley conduction band minimum, into which an electron excited in an SIQD may tunnel. That electron may then tunnel out of the barrier and into the InAs quantum dot on the other side. The charges may be stored in this manner for some seconds, the electron in the quantum dot, the hole in SIQD, until the bias is reversed, and the hole is coaxed to recombine with the electron in the InAs quantum dot. The emitted light from the recombination within the quantum dot can then be observed, to confirm the storage and retrieval success[27].

The X-valley technique has a few drawbacks, though. The origin of the lifetime limitation is unknown, though it may arise from the holes escaping from the SIQD into the surrounding QW. Alternatively, the hole may recombine with the electron,

since they are stored in such proximity. In any case, we search for a storage method that doesn't keep the opposite charge carrier so close by[27].

The second scheme, resonant selective diffusion and neighbor ionization, actually exploits two different mechanisms along with resonant excitation near the GaAs band-edge energy, E_g^{GaAs} . In the first case, the excitation energy is higher than E_g^{GaAs} by hv_{ex} , which is related to the kinetic energy of the resultant excitons. The electron and hole then diffuse, but since the diffusivity of the electron is much greater than the hole, electrons are more often captured by quantum dots, resulting in a net negative charge. In fact, as hv_{ex} is varied, an oscillatory behavior is observed, where the quantum dot is either doubly negatively charged or neutral, depending on whether hv_{ex} is equal to an integer number of LO phonon energies, hv_{LO} . That is, if, after a cascade of LO phonon emissions, the exciton still has some residual kinetic energy, the electrons will be efficiently captured by quantum dots. In the second case, the excitation energy is varied between the band-edge of the wetting layer, E_g^{WL} , and E_g^{GaAs} . Above a threshold energy, acceptors in the bulk are excited, which contribute electrons to the quantum dots[46].

In both of these cases, a clear advantage is that no external voltage bias is needed to charge the dots. The major disadvantage is that both of these cases require fine control of the excitation energy, while continually interrogating the photoluminescence (PL) of a single quantum dot, to monitor its charge state[46]. Since we desire to charge ensembles of quantum dots, we must look elsewhere.
The third process is resonant quantum dot excitation and tunneling. Much like the MISFET approach, the quantum dots are embedded in an insulating region of a MISFET structure. In this case, though, carriers are selectively excited within the quantum dots, the FG is biased so that one species, say the hole, tunnels out to the BG, while the other, in this case the electron, is prevented from tunneling out by a tunneling barrier placed between the quantum dots and the FG. Through such a process, charges have been stored for longer than 25 microseconds. To read out how much charge remains in the quantum dots, the sample is simply reverse-biased, so as to inject the complimentary charge from the BG into the quantum dot; the light produced from the recombination of the electron and injected hole can then be observed[46].

For our purposes, though, this approach appears to have the same drawbacks as the MISFET structure. Namely, we require the ability to tune the quantum dot region, via an external field, close to flat-band conditions. Achieving such a situation with this method would be quite challenging.

2.4 MIR Spectroscopy

We now move our attention to the last focus of our review, mid-infrared (MIR) spectroscopy. A full review of the myriad techniques used to study the MIR behavior of quantum dots is beyond our scope; a limited review is more appropriate. Typically either a wedged waveguide[61-63] or a photocurrent routine[64-68] is used to extract the absorption profile of bound-to-bound and bound-to-continuum

transitions in quantum dots. For this discussion, we shall consider quantum dots of materials other than strictly InAs (e.g., SiGe).

2.4.1 Wedge-Waveguide Spectroscopy

In this approach, many – up to hundreds! – of layers of quantum dots are grown, often in a vertically-coupled manner (§2.1.2). The charge states are not necessarily coupled, however. Since the normal-incidence cross sectional area of quantum dots is so small, on the order of 100 nm²/QD, the multiple layering is meant to increase the total absorption. Often, the absorption is further enhanced by creating a wedged waveguide. First, the top and bottom surfaces of the sample are metallized, which confines the MIR light within the sample. Then, the sides of the sample are polished at some angle, usually 45°, and the MIR beam is sent into one polished facet so as to bounce back and forth inside the waveguide before exiting the other facet. This allows the MIR many passes through the quantum dot layers, enhancing the absorption by several times. Typically, the quantum dot regions are also doped so as to provide carriers to perform the intraband absorption. The MIR source is usually a broadband Globar® source from a MIR Fourier-transform infrared (FTIR) spectrometer[8].

2.4.2 Vertical and Lateral Photocurrent

Several workers have used photocurrent (PC) schemes to probe bound-tocontinuum transitions in quantum dots[69, 70]. For vertical PC, a doped quantum dot is located in an insulating region under a mild electric field in the growth direction. Injected MIR light excites the bound charge to states in or near the continuum; the electric field then sweeps the charge away, and a PC signal is registered. This is the same principle behind many quantum dot infrared photodetectors (QDIPs) [69, 70]. For lateral PC, a high-mobility conduction path is provided *within* the plane, so that once a charge is excited out of a quantum dot, it falls into the channel and registers as PC. In these devices, two types of conduction paths are usually used: either the existence of the wetting layer (WL) in contact with all the quantum dots on each plane is exploited, or a modulation-doped two-dimensional electron gas (2DEG) is inserted to serve the same purpose. Once again, the quantum dots are usually doped during growth, and the MIR source is usually a broadband Globar®. In this case, however, since the sample itself emits a PC signal proportional to carrier escape, it can be used as a detector; the detectivity peaks at the bound-to-continuum transition energies, so these energies may be mapped out using an FTIR system[64, 65, 67].

In an initial study of quantum dot systems, bound-to-continuum transitions are interesting to study. This method has the same disadvantage of starting with doped quantum dots as in §2.4.1. Also, this structure allows some background level of current (dark current), which must be subtracted away. But, like in §2.4.1, this is also an interesting technique that should be kept in mind.

3 Simultaneous Loading and Tuning: MISFET Structure

3.1 Theoretical Background

Two main metal-insulator-semiconductor field-effect transistor (MISFET) designs were used in this work; both are described in detail elsewhere (§9.1.1.1). The first was the extensively studied two-terminal device; here the mechanism of charge loading and some discussion of the spectroscopic analysis is given. The second was a three-terminal MISFET device, using DX centers (see §4). The proposed charge loading mechanism and some issues with the design are given here, as well.

3.1.1 Loading and Tuning Quantum Dots

As discussed in the previous chapter, much work on quantum dots has been done using the standard MISFET design (Figure 3.1.1-1). In this design, there are two gates; a buried, doped GaAs layer which serves as a back gate (BG); and a metal Schottky front gate (FG), usually Al, deposited on the epilayer surface. These gates are separated by a GaAs insulating region, with an $Al_{0.3}Ga_{0.7}As$ tunneling barrier located close to the FG. Quantum dots are embedded in the GaAs insulating region, close to the BG, and charges tunnel into the quantum dot layer from the BG, when an appropriate voltage, V_{FG} , is applied to the FG relative to the BG.



Figure 3.1.1-1 MISFET Conduction Band Diagram

3.1.1.1 Loading, Tuning, and Calibration

To load quantum dots using MISFET structures, the 'lever-arm' effect is generally employed. For any V_{FG} voltage applied, the actual band alignment at the surface must include the Schottky barrier. The Schottky barrier height is determined by the alignment of the metal's Fermi level with that of the semiconductor surface on which the metal is deposited[23]. Since the surface is nominally undoped, the Fermi level should be pinned approximately mid-gap. Therefore, the Schottky barrier height from the voltage of the FG to the conduction band of GaAs is approximately one-half the GaAs band gap, or 0.7 V. This value varies from sample to sample depending on how the metal-semiconductor interface is treated, as charge traps at the

epilayer surface can have significant effects on the Fermi level alignment. Also, due to the lack of doping, the solution of the Poisson equation between the Schottky FG and the BG yields a linear voltage drop across the insulating region -a constant electric field E_z. This field creates a potential difference between the quantum dot layer and the BG, V_{QD} . The BG is heavily n-doped (10¹⁸ cm⁻³), so the Fermi level is very close to the GaAs conduction band. The quantum dot layer is close to the BG (250 Å), and so electrons may tunnel from the BG to the quantum dot layer if any states are available in the quantum dots, at or below the Fermi level. For the quantum dots used (height ~ 6 nm, diameter ~ 20 nm), the lowest bound electron energy state was expected to be \sim 330 meV below the GaAs conduction band. To barely empty the dots, one should then have $V_{OD} \sim -0.33$ V, so that the lowest quantum dot state is just barely above the Fermi level. Since the distance from the quantum dot to the BG is $\sim 1/7^{\text{th}}$ of the distance from the BG to the FG (a 'lever arm' of 6.72, to be precise), this implies that $V_{FG} + V_{Schottky} \sim 6.72 \text{ x } V_{QD}$, or $V_{FG} = -1.52$ V. Indeed, for some of the samples, we used just this value of V_{FG} to empty the quantum dots.

To tune the quantum dot layers with respect to each other, only a slight modification is required. If the quantum dots are identical, the resonant coupling of their energy levels requires flat-band conditions, or zero electric field in the insulating region. This implies that $V_{FG} = 0.7$ V, to counteract the built-in $V_{Schottky}$. This is the reason for the Al_{0.3}Ga_{0.7}As tunneling barrier near the FG; without such a barrier, electrons would readily flow across the device, from the BG to the FG, at this voltage. To tune the quantum dot layers 10 meV out of resonance with each other, with a distance between the layers of 7 nm, one needs to swing V_{FG} by 0.24 V, to 0.46 V. From these two exercises, one can easily see that this is not the ideal system to use for studying electronic coupling, as the coupling is so intertwined with the charge loading.

To calibrate the quantum dot charge loading, capacitance-voltage spectroscopy is an excellent tool(§9.4.1.1). Typically, we applied both a constant V_{dc} and a small (mV), sinusoidal (~34 Hz) 'tickle' V_{ac} to the FG ($V_{FG} = V_{dc} + V_{ac}$), while measuring the current (I_{BG}) flowing through the BG, which was pinned to a virtual ground. Using standard lock-in techniques, we detected the amplitude and phase of the current signal with respect to the V_{FG}, and from this signal calculated the capacitance, C_s , of the sample. This C_s varies as a function of V_{dc} ; this is the C(V) spectrum. Because $V_{ac} \ll V_{dc}$, V_{ac} is too small to effect the population of the quantum dots significantly. As V_{dc} increases and drives energy levels in the quantum dots into resonance with the BG Fermi level, charges tunnel back and forth from the BG to the quantum dots rapidly. As long as the 'tickle' frequency is slow enough, the BG and quantum dots are effectively shorted together; the effective distance from the BG to the FG is decreased by a factor of $\sim 6/7$, and the effective C_s is increased by a factor of \sim 7/6. As V_{dc} continues to increase, the resonant quantum dot level fills, and charge may no longer tunnel back and forth as before. Then, since the BG and quantum dot layers are no longer effectively shorted, C_s should in principle return to its initial state. This process results in resonant peaks in the CV spectra, which can be

correlated to the energy level structure of the quantum dots in the quantum dot layer. Since there is a $\sim 10\%$ variation in the size of the quantum dots, the peaks are significantly broadened, which makes the more closely-spaced higher quantum dot levels particularly difficult to discern. Further, since the quantum dots don't occupy the total area in their plane, the effective front gate-back gate distance is actually a mixture of the two distances. This has the effect of lowering the peak value of the capacitance. The peaks also appear on top of a background, as traps in the bulk are similarly filled[3].

CV spectroscopy is therefore extremely useful in terms of loading charges into quantum dots in MISFET structures, by determining the exact voltages $V_{FG} = V_{dc}$ at which charges may tunnel into the quantum dots from the BG. Indeed, the integration of $C(V_{FG})$ with respect to V_{FG} yields the actual charge density loaded. This provides a good check against the estimated quantum dot density from the growth.

3.1.1.2 Far-Infrared Spectroscopy and Analysis

A great deal of information on the quantum dots under study can be obtained through far-infrared (FIR) spectroscopy. It is useful, therefore, to review a few concepts that will come into play in this analysis.

In linear absorption, the attenuation coefficient κ on resonance for absorption in a material with index of refraction *n*, number density of absorbers *N*, line half-width $\Delta \omega$, and dipole matrix element for the transition μ is[22]:

$$\kappa = \frac{4\sqrt{\pi}N\mu^2}{\hbar cn\Delta\omega}$$

Equation 3-1

Knowledge of the material length and the change in transmission due to the absorbers (as opposed to that change due to reflections or absorption in the bulk) then allows an estimate of the attenuation coefficient. From Eq. 3-1 and knowledge of the values of N, n, and $\Delta\omega$, the dipole matrix element can be readily calculated.

Given any two-level system with a finite decay rate, there exists some intensity I_s of illumination under which the rate of excitation is equal to the rate of spontaneous emission. I_s is the saturation intensity, since once the upper and lower state populations are equal, the sample does not absorb additional intensity. One can then plot the differential absorption ∂A (1 - ∂T , actually) as a function of intensity I, and fit the plot to a function of this form, which is correct for inhomogeneously broadened transitions[71]:

$$\partial A = (1 - \partial T) = 1 - \frac{A_0}{\sqrt{1 + \frac{I}{I_s}}}$$

Equation 3-2

Where ∂T is the differential transmission, the percentage change in the transmission with and without loaded charges, and A_o is ∂T for $I \ll I_s$.

Consider a MISFET structure as described above with a Schottky FG and a doped GaAs BG. The FG-BG distance is only $\sim .2 \ \mu$ m; the wavelength of light at our

energy of interest (10 meV) is ~100 μ m in vacuum, or ~30 μ m in GaAs. This means that the entire device is small compared to the FIR wavelength in the direction of polarization. Under high FIR intensities on the order of 100 W/cm², the ∂V_{FG} induced on the device is about .3 mV. Clearly, this is small – it may be enhanced due to waveguide effects – but we nevertheless observe significant current increase when the device is exposed to intense FIR via the FEL (§3.2.4). We interpret this current as arising from a half-wave rectification of the THz frequency voltage, which gives rise to an effectively higher V_{FG}. The author is uncertain whether this incremental ∂V_{FG} has any effect on the loading of the quantum dots, or their absorption under high fields.

From the saturation intensity I_s , and the knowledge of the dipole matrix element μ , one can determine the product of the longitudinal (T₁) and the transverse (T₂) relaxation times via[22]:

$$I_s = \frac{nc\hbar^2}{4\pi T_1 T_2 \mu^2}$$

Equation 3-3

 T_1 and T_2 are related by [22]:

$$T_2^{-1} = \frac{1}{2}T_1^{-1} + \gamma_{\phi}$$

Equation 3-4

Where γ_{ϕ} is the pure dephasing rate. We can then see that $T_2 \leq 2T_1$, so that knowledge of the product T_1T_2 sets limits on T_1 and T_2 individually:

$$\frac{{T_2}^2}{2} \le T_1 T_2 \le 2{T_1}^2$$

Equation 3-5

Theoretical predictions and simulations based on observational trends of quantum dot lifetimes disagree, yielding acoustic-phonon-limited lifetimes from 10's of ns – the 'phonon bottleneck' – to 10's of ps[72, 73]. For situations where the WL may also be populated, others have predicted Auger scattering that shortens lifetimes to 10's of ps[74, 75].

3.2 Experimental Data

As described in more detail in the Appendices (§9.4.1), we used MISFET-type structures to study coupled quantum dots. First, we used CV and FTIR spectroscopy to calibrate the loading of electrons and tuning of the coupling, respectively. Then, we probed the system with strong THz fields from the free-electron laser (FEL) to determine a saturation intensity for the bonding-antibonding transition. This intensity gave us some information on the lifetimes of the bonding-antibonding antibonding superpositions.

3.2.1 Sample Growth Characteristics

We started with three samples, A, B, and C. The relevant differences between the samples ($\S9.1.1.1$) were the heights of the two quantum dot layers grown, and the distances between the layers. Sample A was intentionally asymmetric; the two quantum dot layers were grown 7 nm apart, the first with a height $\cong 1.5$ nm, the second \approx 6 nm. Samples B and C were both symmetric, with the height of both layers of quantum dots ~ 6 nm. However, the quantum dot layers were grown 7 nm and 7.5 nm apart for samples B and C, respectively.

3.2.2 Capacitance-Voltage Characterization

To map out the loading of the coupled quantum dots as a function of V_{FG} (§3.1.1.1), we performed CV spectroscopy on all three samples (Figure 3.2.2-1, Figure 3.2.2-2). The first feature we see in the data on sample C is the sequential filling of the lowest energy states of the bottom quantum dot layer, at about -0.78 V and -0.63 V. At these large negative voltages, the quantum dot layers are detuned out of the coupling regime, so the states are localized within the separate dot layers. The voltage difference between the first and second electron is due to the Coulomb blockade effect. Next we see a shoulder which represents loading of electrons into the p-states of the bottom quantum dot layer, between -0.3 and -0.2 V. Superimposed on the bottom quantum dot filling are the resonances from the filling of the top quantum dot states. Due to the lever arm effect, the expected voltages for the filling of the s- and p-states for the top quantum dots are -0.27 and 0.27 V, respectively. We can see a weak shoulder at 0.25 V, which may confirm the top quantum dot p-state loading. At 0.5 V, we see filling of the wetting layer (WL) states. The flat-band condition, at which V_{FG} offsets the Schottky barrier, should occur at 0.7-0.8 V. At these voltages, we should see coupling of s- and p-states of the bottom quantum dots with the s- and p-states of the top quantum dots,

respectively. Of course, at these voltages, the dots are also full of charges, so we begin to see the problem that arises when we load and tune coupled quantum dots by varying a single parameter (§3.1.1.1).



Figure 3.2.2-1 CV of Sample C. Note quantum dot loading resonances.

The data for samples A and B (Figure 3.2.2-2) are also given. It is unclear why the background capacitance, seen as the capacitance and large, negative voltages, is so different between A and B on the one hand, and C on the other (16 and 17 nF, respectively). This may be due to a small parasitic capacitance in parallel with C. Also, the charging of sample A definitely appears at more positive V_{FG} than C, and B appears to do so, as well: 0.15 and -0.4 V, respectively. This implies that the ground states are closer to the GaAs band edge than in sample C. The shift in A is explained by the fact that the bottom quantum dots in A are much smaller in the growth direction (1.5 nm vs. 6 nm for the others), the primary dimension of confinement. This pushes the ground state energies for the bottom quantum dots in A higher. The apparent shift in B may be explained by variations in thickness or composition of the quantum dots in B – it may be that the resonances are smeared out in this way. The s- and p-states of the top quantum dots in sample B are expected to be filled at 0.01 and 0.58 V, respectively.



Figure 3.2.2-2 CV of Samples A and B. Note quantum dot loading resonances.

3.2.3 FTIR Characterization

To spectrally locate the expected bonding-antibonding transitions, we used farinfrared (FIR) Fourier-transform infrared (FTIR) spectroscopy (§9.3), in the waveguide geometry, with the FIR polarized in the growth direction. Here, we used the results obtained from CV to take FIR spectra at V_{FG} where the coupled quantum dots were expected to be charged, and normalized them by spectra where they were expected to be empty. To compare various spectra at once, we offset the traces for clarity (Figure 3.2.3-1, Figure 3.2.3-2, Figure 3.2.3-3).

Looking first to sample B FTIR vs. V_{FG} (Figure 3.2.3-1), we see two separate absorption peaks, a weak one at 22 meV for $V_{FG} \ge 0.6$ V, and the other at 12 meV which grows to 5.5%, and is distinguishable for $V_{FG} \ge 0.65$ V. We also see some slight tuning at larger V_{FG} , but no noticeable increase in absorption, so we conclude that higher voltages don't impact the charge state of the coupled quantum dots. Looking at A vs. B FTIR vs. V_{FG} (Figure 3.2.3-2), we see that A also exhibits the absorption at 22 meV, but not the one at 12 meV, which suggests that the latter is linked to the bottom quantum dot height, i.e. that in sample A there is no coupling.



Figure 3.2.3-1 FTIR Sample B. Normalized at 1.0 V, offset for clarity.



Figure 3.2.3-2 FTIR Transmission for Samples A and B. Normalized by transmission at V_{FG} of 1.0 and 1.1 V, respectively.

At these frequencies, we have identified 4 possible mechanisms to explain absorption for these samples: 1) transitions between tunnel-split quantum dot states; 2) transitions between p-states and the WL states; 3) transitions between tunnelsplit coupled WL (CWL) states; 4) intersubband transitions in a 2DEG that may arise at a nearby GaAs/AlGaAs interface. The transition energy between s- and pstates is expected at 50 meV from PL, and the necessary polarization for such a transition is within the plane, in any case. A simple one-dimensional calculation of the expected tunnel splitting of the CWL states is given by assuming 0.5 nm-thick InAs QWs separated by a 7 nm-thick barrier, and yields an expected absorption at 24 meV. Since the WLs are positioned identically for both samples, we suspect that the 22 meV absorption is due to mechanism (3). Looking at the 12 meV absorption, since it exists for B but not for A, and since the GaAs/AlGaAs interfaces are identical for both samples, we rule out (4) as an explanation. Similarly, (2) and (3) don't explain the difference between A and B, as both mechanisms would occur for both samples in this voltage range. So we're left with (1) as an explanation for the 12 meV absorption.

The association of the 12 meV absorption with transitions between tunnel-split bonding and antibonding states also explains other features observed. This absorption isn't found in A, because the quantum dot heights are too dissimilar to allow coupling. Also, this type of absorption should only appear when the quantum dot levels are loaded with electrons and brought close to resonance. In this case, from CV, we know that B is loaded with electrons, and that it is close to flat-band conditions at $V_{FG} = 0.65$ V, when this absorption arises. If the 22 meV absorption was correctly identified, then the redshift between these two makes sense, since the tighter (looser) bound states of the quantum dot (WL) should lead to smaller (larger) tunnel-splitting. The 12 meV absorption is broader, which would be expected from the size-distribution of quantum dots. Finally, from CV, we would expect charges to exist in both the s- and p-states of both bottom and top quantum dots in sample B at $V_{FG} \ge 0.58$ V, so the observed absorption should be between the tunnel-split p-

states. State-filling should preclude observation of s-state absorption at these voltages.

We see a few confirmations of our previous assignments in the FTIR spectra of sample C (Figure 3.2.3-3). The high-energy absorption remains near 24 meV, and we again identified it as CWL tunnel-splitting. This peak grows with increasing bias. The low-energy absorption appears at 10 meV and $V_{FG} \approx 0.45$ V, grows to maximum absorption at $V_{FG} \approx 0.7$ V, and shifts slightly to higher energies before vanishing at $V_{FG} \approx 1.1$ V. The lower energy of the absorption (10 meV vs. 12 meV for B) is consistent with a smaller tunnel splitting due to the larger tunneling barrier, as the thickness of the GaAs layer between the dots is larger (7.5 nm vs. 7nm for B). So, we again assign the low-energy absorption to the tunnel-splitting of the p-states. The quenching of this absorption at 1.1 V is consistent with a complete filling of the p-states. That this occurs in our range of investigation for C but not for B is also consistent with the CV data, which suggested that the quantum dot states for B were slightly higher (closer to the GaAs band edge) than for C. Hence, for increasing V_{FG} , the 12 meV absorption didn't change in the spectra for B. Also, for V_{FG} near and above that for flat-band conditions, current flow across the device may disturb any steady-state quantum dot populations.



Figure 3.2.3-3 FTIR Sample C. Normalized at 1.5 V, offset for clarity.

3.2.4 FIR Saturation

Using a Free-Electron Laser (FEL) tuned to the observed FIR resonance (§3.2.3) at 85 cm⁻¹ (10 meV), we analyzed samples B and C, as a function of power and V_{FG} (Figure 3.2.4-1, Figure 3.2.4-2, Figure 3.2.4-3). We used transmission at V_{FG} = -1 (-1.5) V to normalize transmission data for sample B (C). Looking first to our transmission curves for B and C as a function of V_{FG} (Figure 3.2.3-1, Figure 3.2.3-3), we see an absorption onset at around V_{FG} = 0.8 V, where we expect the p-states of both samples to be partially full. At this point, the absorption from the coupled quantum dot bonding-antibonding transition is superimposed on free carrier and

CWL absorption, where the coupled quantum dots in sample B (C) contribute 4 (2.2) ± 0.3 % of the total absorption. These figures are found by subtracting the absorption at 85 cm⁻¹ for $V_{FG} = 0.85$ (0.7) V from the expected background for sample B (C). The expected background is not the normalizing background, but rather the non-coupled quantum dot background extrapolated as a smooth connection of the non-coupled quantum dot absorption at lower and higher energies in analogy with the spectrum of sample A (§3.2.3). From the integrated absorption and the number of quantum dots measured via CV, we calculate a dipole matrix element of 12 ± 0.3 (5.5 ± 0.2) nm for sample B (C). The difference between these two values may be a result of differing numbers of optically active quantum dots in the two samples. Also, from our previous discussion (§3.2.2), we expect that the variation in dot height may be greater in B, and that the dots are more loosely binding in B than in C. Both of these would support a larger vertical cross-section.



Figure 3.2.4-1 Transmission vs. V_{FG} , Samples B and C. Normalization Shown. FTIR (red line), FEL (blue points), overlaid.

Looking again at our fixed energy (85 cm⁻¹) transmission vs. V_{FG} spectra (Figure 3.2.4-1), we see good agreement between the FTIR data (solid lines) and the linear-regime FEL data (diamonds, squares for B, C).

Maintaining a constant energy of 85 cm⁻¹ and V_{FG} of 0.85 V while varying the FEL intensity on sample B (Figure 3.2.4-2), we clearly see a saturation, where the absorption is bleached at 100 W/cm². We attribute this saturation to the coupled quantum dot p-states. The absorption at this energy doesn't entirely saturate, which suggests that the background absorption doesn't saturate at these intensities. A least-squares fit to a standard, inhomogeneously broadened saturation curve (solid line) yields a saturation intensity of 0.25 ± 0.15 W/cm². This intensity is of the same

order as reports of intersubband transitions[76], but 6 orders of magnitude less than intraband quantum dot transitions at 10 μ m[77]. A combination of differences in relaxation times and dipole matrix elements may explain this stark difference.



Figure 3.2.4-2 Transmission vs. Power, Sample B. Normalization Shown. Fit yields $I_{sat} = 0.25 \pm 0.15$ W/cm².

Looking now at the saturation of sample C (Figure 3.2.4-3), again at 85 cm⁻¹, we observed the absorption at two different voltages, $V_{FG} = 0.7$ and 0.9 V to check the difference in behavior when the coupled quantum dot p-states are partially and completely full, respectively. Again, we see a saturation for $V_{FG} = 0.7$ V, but no distinct change at all for 0.9 V. This is expected, since the absorption should already be quenched at $V_{FG} = 0.9$ V due to state-filling, leaving only the observed background

absorption, which again is unsaturated at these intensities. The saturation curve for $V_{FG} = 0.7 \text{ V}$, as before, yields a saturation intensity of $4.5 \pm 3.5 \text{ W/cm}^2$. The magnitude of the saturation is less than 1%, where the total coupled quantum dot absorption was measured to be over 2%; this discrepancy has been reported to be due to the rectification effect of the high FIR intensities on the sample. At high FIR intensities, the current flow through the sample increases, suggesting an increase in the effective V_{FG} . It is argued that the falling-off of absorption of the coupled quantum dots in sample C with increasing effective V_{FG} explains the non-bleaching at high FIR intensities. This argument is somewhat lacking; assuming the states were being filled, they would indeed contribute less absorption, and so at high FIR intensities the transmission should be even closer to 1. Unfortunately, no satisfying alternative explanation has been found.



Figure 3.2.4-3 Transmission vs. Power, Sample C. Normalization Shown. Loaded (0.7 V, red) and Overloaded (0.9 V, blue). Fit (green) yields $I_{sat} = 4.5 \pm 3.5 \text{ W/cm}^2$

From the saturation intensities, we calculate a T_1T_2 product of 5500 (1700) ps² for sample B (C) (§3.1.1.2); part of this difference may result from different coupling efficiencies. From these values we can estimate a lower limit: the longest T_2 possible is $2T_1$. Assuming this is the case, and taking account of the uncertainty associated with the least-squares fit that determined the saturation intensity, we arrive at ranges for possible T_1 of 40 - 85 (20 - 65) ps for sample B (C). Another factor of 2

uncertainty is embedded in the absolute value of the coupled intensity, which applies another 30-40% variation on T_1 .

One source of broadening that may decrease T_1 is the interaction between electrons. Because of the high V_{FG} needed to achieve the flat-band conditions necessary for coupling, many carriers are injected into the coupled quantum dots as well as the coupled wetting layer states. In fact, short relaxation times have been predicted via Auger processes when a high density 2DEG is close to quantum dots[74, 75]. It is to investigate T_1 and T_2 that we proceed from this structure to one that admits independent control of the loading and tuning of coupled quantum dots.

3.3 Summary

We observed a tunable absorption peak in FIR transmission which we attribute to the bonding-antibonding transition of coupled p-states in InAs quantum dots. We see that absorption near 85 cm⁻¹, and we further observe a bleaching of the transition at high FIR intensities. From this bleaching, we deduce a lower bound on the longitudinal relaxation time T_1 of approximately 40 ps. We expect that the intertwined nature of loading charges into our dots and coupling the dots with one another leads to a shortening of the relaxation time by Auger or other processes.

4 Separate Loading and Tuning: MISFET-DX Structure

4.1 Theoretical Background

The fatal flaw in the MISFET design, for the purposes of few-electron coupled quantum dot spectroscopy, was that the tuning of the coupling and the loading of the charge were done *simultaneously*; a change in V_{FG} would change both the internal alignment of the coupled quantum dot energy levels, and also the alignment of those levels with the BG Fermi level, which would load or empty dots simultaneously. We designed a challenging new structure, completely new to our knowledge, that used thermally-activated carriers, DX centers, as a kind of disposable gate.



Figure 3.2.4-1 MISFET-DX Center Conduction Band Diagram. Sample is Warm and V_{FG} is referenced to the DX Center layer.

DX centers, in our case Si dopants in Al_{0.3}Ga_{0.7}As, are both donors and acceptors at warm temperatures. When they change charge state, there is a local lattice relaxation to a lower total energy state. This deformation requires assistance from phonons, so that at cold temperatures, below 100 K or so, they cease donating and accepting charge, and 'freeze out'. Even at cold temperatures, though, DX centers exhibit persistent photocurrent, which means they can be photoexcited into donating charge; pains must be taken to make sure that they are kept in darkness whenever possible[78]. We imagined making a 'DX gate' that would donate a controllable amount of charge to our coupled quantum dots at warm temperatures (Figure 3.2.4-1). In this way we could adjust V_{FG} in reference to the DX layer to controllably load charge into the coupled quantum dots as before. But, since the DX centers would freeze out at low temperatures, they would also allow us to adjust the field across the coupled quantum dots without worrying about charge flowing back into the DX centers. This would allow us to tune the dots at a constant charge density by now referencing V_{FG} to a deeper buried BG (Figure 3.2.4-2).



Figure 3.2.4-2 MISFET-DX Center Conduction Band Diagram. Sample is Cold and V_{FG} is referenced to the doped quantum well back gate.

Our first attempt at such a structure was to make several changes to the original MISFET device (§9.1.2.6). First, we used a modulation-doped GaAs/Al_{0.3}Ga_{0.7}As QW as our BG, rather than the n-doped GaAs of before. This was to improve the FIR throughput for FTIR, by avoiding absorption by the heavily-doped layer. The surrounding Al_{0.3}Ga_{0.7}As continued above the BG for about 0.5 μ m, at which point we used Si-doped Al_{0.3}Ga_{0.7}As for a DX center layer. The Al_{0.3}Ga_{0.7}As continued after the DX center layer for 30 nm before a 15 nm GaAs QW was deposited, embedded in which were our coupled quantum dot layers. Growth of Al_{0.3}Ga_{0.7}As resumed up to a surface capping layer of GaAs. The coupled quantum dot-surface distance was much shorter here than before (45 vs 100 nm). Further growth details are given elsewhere (§9.1.2.6).

The specifics given here outline the concerns of our first attempt. The main concern was to have the BG as far below the DX layer as reasonably possible. This is to ensure that the NiAuGe ohmic contacts that we used to electrically contact each of these layers would not spike down to short them together. The result is a BG-FG distance of ~550 nm, and a DX-FG distance of ~80 nm.

Consider, now the loading condition for the coupled quantum dots at warm temperatures, when the DX centers are still active (Figure 3.2.4-1). The voltages of the FG and (postulated at) the coupled quantum dot layers are now referenced to the DX center Fermi level, which is 0.12 V below the Al_{0.3}Ga_{0.7}As conduction band. This places the Fermi level 0.21 V above the GaAs conduction band, and 0.42 V above the lowest coupled quantum dot bound state. The lever-arm effect implies,

then, that the total voltage at V_{FG} = -0.42 V * (80 nm / 30 nm) + 1.03 V = -0.09 V. The last 1.03 V is due to the Schottky offset to the $Al_{0.3}Ga_{0.7}As$ band edge. This seems reasonable, so far. However, once cold, the DX centers freeze out, and it is then necessary to bring the coupled quantum dots back to flat-band condition for coupling (Figure 3.2.4-2). At that point, however, there is a field remaining between the DX center layer and the FG. This field is maintained by a charge density frozen into the DX layer, and compensating charge drawn to the FG. To bring the coupled quantum dots into flat-band conditions, the entire remaining field must be cancelled. Now V_{FG} must be held in reference to the BG. If, during the previous manipulations, the BG had been held shorted electrically to the DX center layer, it is still at an equipotential with the floating DX centers and the V_{FG} in reference to the BG is still -0.09 V. The voltage necessary to bring the coupled quantum dots into resonance is $V_{FG} = -0.09 + 1.12 \text{ V} * (500/80) = 6.91 \text{ V}$. Keeping in mind that the turn-on voltage for an Al Schottky diode, even on Al_{0.3}Ga_{0.7}As, is about 1.03 V, this method should result in incredible current flow.

There are several problems with this procedure. The first is that the BG is too far from the DX and FG, so that the second lever arm multiplies the tuning field into a completely unreasonable forward V_{FG} . The second is that there is no significant tunneling barrier between the BG and DX layers, so that any decent forward voltage in reference to the BG would immediately draw current. The third is that the DXcoupled quantum dot distance was too close, meaning the loading field in that region had to be quite high. The fourth is that the BG was initially shorted to the DX

centers. If, instead, the BG were held at a positive voltage in reference to the DX layer, then the total BG-FG voltage could be reduced by that much, at the last step. Of all these problems, the huge BG-DX distance was the worst.

To correct these problems, we shortened the distances quite a bit (§9.1.2.7). We put the BG-DX distance to ~100 nm, and the DX-FG distance to ~220 nm. At cold temperatures, the DX centers were to freeze out, and so electrical contact to that layer would be lost. But since using CV to measure the charge loading would be useful, contact to either the source or drain of charge was necessary. It is impossible to electrically contact all the quantum dots (they would cease to be quantum dots), so we replaced the quantum dots with InAs CQWs (9.1.2.8). The plan was to electrically contact the QWs and monitor their charge state via CV. In this case, the CQW contact would be left floating, and we would only detect the AC signal for our CV measurement. The CQWs were placed ~100 nm from the DX centers. We also inserted a tunneling barrier between the BG and DX layers.

Thus, the structure grew from a two-terminal MISFET design, to a nonfunctional three-terminal MISFET-DX design, to a critically flawed four-terminal MISFET-DX CQW design. The problem now was the distances; they were just too short. Keeping in mind that we wanted three separate (electrically independent) ohmic contacts to three charge layers spaced ~100 nm apart, the spiking distance of our NiAuGe ohmic contact recipe, at 100 - 400 nm, was non-ideal.

A two-terminal processing of the device was attempted. In this case, only the FG, DX and BG would be contacted, and the QW would be left floating. Even the

DX and BG would not be contacted separately, but would actually be shorted together via the annealed ohmic contacts. When warm, the structure would work as previously described. When cold, however, the DX centers would, in principle, freeze out and leave the joint BG-DX contact only electrically connected to the BG. To test the charge in the CQWs, one might then look at the photoluminescence, which would broaden somewhat for filled wells versus empty ones.

4.2 Experimental Data

As reviewed in §4.1, it should, in principle, be possible to construct a device that works in an analogous manner to the previous MISFET devices, but allows storage of a fixed amount of charge. This method would free the V_{FG} to vary independently of the charge density in the coupled quantum dots, thereby allowing independent loading and tuning of electrons in coupled quantum dots, as the title of this section demands.

In short, one would go from a two-terminal MISFET structure – one for the doped-GaAs BG, and one for the Al Schottky FG – to a three-terminal structure we called MISFET-DX. In the order of growth, the gates would be: 1) doped-GaAs BG, 2) Si-doped Al_{0.3}Ga_{0.7}As (DX center) gate, 3) Al Schottky FG. The region with DX centers would be a metallic gate a high temperatures, but at low temperatures (T \leq 100 K) would be an insulating region — indeed, a tunneling barrier between the BG and the coupled quantum dots.

Such a device was attempted but, despite multiple revisions, was eventually abandoned due to growth, processing, and tuning difficulties (§9.2.3, 9.6).

4.3 Summary

We attempted to construct a new device that would allow tuning of coupled quantum dots via a growth-direction electric field while maintaining a constant population of charge within the quantum dots. This structure was designed to use DX centers as a sort of disposable gate. This effort was not successful, due partly to processing difficulties.

5 Separate Loading and Tuning: NIN Structure

5.1 Theoretical Background

Our most recent device design has also been our most successful. Here, we will discuss this n+-doped–insulator–n+-doped (NIN) structure, its origins, modifications, and operational principles. We will also project our expectations of what data may be gained by interrogating this device in several different experiments.

5.1.1 Origins and Previous Work

The original idea for this device came from discussions on photoinduced charge storage, and previous work in the dissertation of W. V. Schoenfeld (WVS)[35]. WVS had designed a structure to electrically isolate coupled quantum dots so that no charge would tunnel in or out of them from any nearby gates, while at the same time allowing for an overall field to be applied in the growth direction E_z . He did so, in order to probe the coupled quantum dots using μ -PL as a function of E_z , in the hope of seeing evidence of electronic coupling in the single-quantum dot spectra. He was unsuccessful in that particular objective, but he did manage to electrically isolate his coupled quantum dots quite well, while maintaining access for the injection of interband light. Meantime, we were looking for a structure that would allow us to inject charges optically into the GaAs medium surrounding our quantum dots, while

applying a suitable E_z so as to control the loading of those charges into the quantum dots. WVS's structure was an almost perfect fit.

5.1.2 Structural Changes

The device grown by WVS differed in several respects from our NIN structure, shown in Figure 5.1.3-1 (a). The two devices were both quite symmetric, with two n^+ -doped GaAs gates surrounding an insulating region. The insulating region was also symmetric, with two AlGaAs barriers sandwiching a central GaAs layer. In the GaAs layer were embedded the quantum dot layers. The differences between the devices start at the barriers. WVS used Al_{0.5}Ga_{0.5}As, but X-valleys, traps, etc. that might arise from such a high Al concentration posed a concern. We chose, instead, to use lower Al_{0.3}Ga_{0.7}As barriers, 20 times as wide. Positioned at the very center of the central GaAs section, far from the Al_{0.5}Ga_{0.5}As barriers, WVS grew a single layer of coupled quantum dots; our device was designed to use the Al_{0.3}Ga_{0.7}As barriers as 'backboards' of a sort, to help funnel charge into the quantum dot layers. So, we put one quantum dot layer on either side of the GaAs range. We also wanted to study the storage mechanism alone, before moving into coupled quantum dots, so we used only single quantum dot layers, on both sides.

5.1.3 Loading Procedure

In this device, electrical control is maintained by separate ohmic contacts to each of the n⁺-doped layers. The bottom (top) layer is referred to as the back (front) gate or BG (FG). To write, store and read electrons using quantum dots in the NIN
structure, four steps are necessary: 1) application of an initial write/store voltage to the FG in reference to the BG, V_{FG} , equal to a DC component V_{dc} and a modulated component V_{amp} ($V_{FG} = V_{write} = V_{dc} + V_{amp}$); 2) NIR illumination normal to the surface with duration T_L and frequency f_{NIR} ; 3) application of a read voltage $V_{FG} =$ V_{dc} after a total time T_{app} ; 4) NIR illumination normal to the surface with duration T_{UL} and frequency f_{NIR} . Each of these steps will now be discussed in detail.



Figure 5.1.3-1 The Loading/Unloading Cycle, Energy Band Diagrams



Figure 5.1.3-2 Loading/Unloading timing diagram.

5.1.3.1 Write/Store Voltage

The entire operating voltage range, both with and without illumination, was experimentally determined via IV and CV measurements (§5.2.1.3). In terms of the IV, the sample exhibits a diode-like turn-on at both positive and negative voltages, which change slightly under illumination. These turn-on voltages were avoided, since current flowing across the structure was assumed to be poor for charge storage purposes. In terms of CV, under zero illumination the capacitance of the device is determined by the growth and processing geometry. We assumed a parallel-plate model, and, with the input of the various dielectric constants of GaAs and $Al_{0.3}Ga_{0.7}As$, were able to predict a capacitance well within an order of magnitude. Specifically, the formula for the capacitance is:

$$C_{T}^{-1} = C_{b}^{-1} + C_{i}^{-1} + C_{b}^{-1};$$

$$C_{T} = \frac{C_{b}C_{i}}{2C_{i} + C_{b}};$$

$$C_{b(i)} = \frac{\varepsilon_{0}\varepsilon_{b(i)}A}{d_{b(i)}};$$

$$\varepsilon_{b(i)} \approx 12.2(13.2);$$

$$d_{b(i)} \approx 500(200)nm;$$

Equation 5-1

Where *A* is the area of the device, and $d_{b(i)}$ is the thickness of the Al_{0.3}Ga_{0.7}As barrier (GaAs interior) layer. The voltage range of operation, then, must also fall within the limits of the sample behaving as a capacitor with a constant CV profile. This range was usually smaller than that determined by IV. Given these two voltage constraints, V_{dc} and V_{amp} are free to be chosen.

The purpose of V_{dc} and V_{amp} are to prepare the sample for the photoinjection of charge (§5.2.2.1). Since charge is created only in the GaAs regions, but not in the barriers, excessive current should not flow under illumination. The capacitance, though should change significantly. While illuminated, enough carriers should be injected into the interior that the GaAs behaves as a conductor rather than a dielectric. In this case, the first line of Equation 5-1 should have $C_i \rightarrow \infty$, to keep the model reasonable. Again, even this simple model is able to predict the change in

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capacitance within a factor of 2. We predicted $C_T^{lit} \sim C_T^{dark}$ *1.2. Under illumination, the suitable CV range also diminished from the dark CV.

When illuminated, charge will separate in the GaAs region according to the electric field resultant from V_{FG} (Figure 5.1.3-1 b). As long as illumination and V_{FG} remain, charge will continue to separate until the built-up charge one either side of the interior region induces a field to balance the initial field (Figure 5.1.3-1 c). At this point, charge may be injected, but, sensing no net field, does not continue to separate. Since there is one quantum dot layer on each side of the GaAs region, we assumed that the built-up charge was stored in the quantum dots. The charge density σ_0 at the quantum dots is, then:

$$\sigma_{0} = \frac{V_{FG}C_{T}}{A} + \sigma_{growth} = \frac{V_{FG}C_{b}}{2A} + \sigma_{growth} = \frac{V_{FG}\varepsilon_{0}\varepsilon_{b}}{2d_{b}} + \sigma_{growth};$$

$$\sigma_{0} \approx \sigma_{growth} + V_{FG} \times 6.8 \times 10^{10} \frac{e}{cm^{2}}$$

Equation 5-2

Where σ_{growth} is the initial charge density due to the built-in fields due to asymmetries in the growth, etc. These units were very convenient, since we expected our quantum dot densities to be ~10¹⁰ cm⁻². From Equation 5-2, we can see that, to study the storage of charge in quantum dots as a function of charge density, it would be useful to know σ_{growth} , or at least cancel it out by appropriate choice of V_{dc} . When we measure the stored charge in step 4 (Figure 5.1.3-1 d), we are actually measuring the incremental charge density σ_{amp} due to the modulation of V_{amp} between illumination steps. Suspecting that the storage lifetime of σ_{amp} would be shortened by large baseline charge, we measured the lifetime as a function of V_{dc} , and chose V_{dc} based on that data. V_{amp} was chosen to load a few (1-6) electrons in each quantum dot.

5.1.3.2 NIR Illumination 1

The first pulse of NIR illumination, NIR1, was designed to provide sufficient photogenerated carriers in the GaAs region to make maximum use of the applied field. The wavelength was chosen to be 800 nm, so as to avoid creating charges in the barrier regions. A sufficient flux of light is necessary to maintain electron and hole gasses of two-dimensional *densities* much greater than the quantum dot density. These gasses would have to be maintained for a *time* sufficient for a) the carriers to transit the GaAs region in the growth direction, and b) the device itself to reach a new electrostatic equilibrium. Brief calculations shows that (b) is the longer time scale, essentially the RC time of the system.

The number of photons that make it from the surface to be absorbed in the GaAs region is quite small. I calculate 30% loss from reflection (at normal incidence), 28% of the remainder absorbed in the doped GaAs of the FG, and only 7.5% of what's left absorbed in the GaAs interior, assuming a transparent top barrier. The net result is that only 3.8% of the incident photons are used. Assuming 1 mW of power is incident on the surface (§9.4.3.2), and a device area of 0.1 cm² we have 1.5×10^{15} photons cm⁻² s⁻¹ in the active region – plenty for our purposes.

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Since the RC time of our system depends on the sample capacitance, which depends on illumination, we chose to experimentally determine the duration of NIR1, T_L . We did that by running our experiment at varying T_L , monitoring the charge read, and found a threshold above which the stored charge did not change much for an incremental change in T_L . We then fit the data to exponential rises, but the results are clear enough to be read off the data by eye (Figure 5.2.2-4).

5.1.3.3 Storage Time

After NIR1, $V_{FG} = V_{dc} + V_{amp}$ was maintained for a time T_{wait} . The total time that V_{amp} was applied was T_{app} . $T_{app} = T_{wait} + T_L + T_{RC}$, where T_{RC} was a settling time chosen to be a few RC time constants. Because T_L , T_{RC} (100 μ s – ms) « T_{wait} (10 ms – 10 s), $T_{app} \sim T_{wait}$. The maximum storage time was assumed to occur when there was no field to drive the charges out of the quantum dots. This occurred with $V_{FG} = V_{dc} + V_{amp}$ right after NIR1, since the field had just been cancelled near the quantum dots by NIR1. So the default storage condition was with $V_{FG} = V_{dc} + V_{amp}$. After T_{wait} , V_{amp} was removed and only $V_{FG} = V_{dc}$ remained, in preparation for the unloading pulse.

5.1.3.4 NIR Illumination 2

 T_{RC} after the end of T_{wait} , the system was in a peculiar condition. The total voltage FG-BG was $V_{FG} = V_{dc}$, so one might expect the external charge density (on the gates) to depend only on V_{dc} . However, recall that, in step 2, the quantum dots on either side of the GaAs interior stored a charge σ_0 , as in Equation 5-2. Now, with

 V_{amp} turned off, that charge density remained, including the amount of charge due to V_{amp} , σ_{amp} . This incremental stored charge now draws its own compensating charge σ'_{amp} onto the gates:

$$\sigma_{amp}' = \frac{V_{amp_i}C_T}{A} = \frac{A\sigma_{amp}C_T}{C_iA} = \frac{\sigma_{amp}C_T}{C_i};$$
$$\sigma_{amp}' \approx \frac{\sigma_{amp}}{6.4}$$

Equation 5-3

Where V_{amp-i} is the voltage induced across the internal GaAs layer by the stored incremental charge σ_{amp} .

After T_{wait} , another NIR pulse is sent to the sample, NIR2, for a duration T_{UL} . NIR2 again injects charges into the GaAs interior, which now separate only because of the field induced by σ_{amp} . The charges separate in the opposite direction as for NIR1, now to the point where they cancel out σ_{amp} 's induced field. In so doing, they recombine with σ_{amp} within the quantum dots, and then stop separating and have no more net effect. The result is to release σ'_{amp} from the gates, which then flows through the circuit. We monitored this current flow; its integral, properly scaled, gave a measure of the amount of σ_{amp} had remained in the device for the time T_{wait} .

We chose the time T_{UL} in much the same way as T_L . That is to say we observed the charge storage as a function of T_{UL} , and observed an exponential rise. This showed us that increasing T_{UL} beyond a certain range would no longer increase the amount of charge extracted during the unloading stage. Similarly, we chose T_{int} , the time over which we integrated the unloading current pulse, by varying T_{int} until we saw that we were capturing the vast majority of the charge in our integration. Again, we were looking for a situation where small changes to T_{int} (~10 ms) would not effect the amount of charge measured.

5.2 Experimental Data

Charge storage was first observed in a dark (non-optical) cryostat with the LED as an excitation source. Because of the cryogenic instabilities and the broad spectral emission, the LED was abandoned in favor of a diode laser. In an optical cryostat, charge storage was not initially observed, so the laser was coupled into the dark cryostat using an optical fiber (§9.4.3). Afterward, we moved our sample back to the optical cryostat with a good deal more success (§9.5.1). We suspect that our first trial in the optical cryostat subjected the sample to too much stray room light; during our second trial, we were able to see significant diminution of the storage signal when room lights were allowed onto the sample.

5.2.1 Sample Characterization

Before we could be confident using this new structure as designed in a loading/unloading scheme, it was necessary to characterize its basic properties, optical and electrical.

5.2.1.1 Sample Growth Characteristics

We used two different n-i-n samples for these experiments, Cersei and Sansa (C and S, respectively), whose growth parameters were designed to be identical, except that C had quantum dots imbedded in the GaAs section of the insulating region (§9.1.3), whereas S, a reference sample, had no quantum dots (and no associated WLs) at all. The FG doped layer for both C and S extended to the epilayer surface, which distinguishes them from previous n-i-n growths.

5.2.1.2 Photoluminescence

As described in the appendices (§9.4), we used a fiber to couple our short-passfiltered NIR (800 nm) laser diode into the cryostat to perform the loading/unloading cycle. We also used the same fiber to collect the photoluminescence. Looking at Sample C's spectrum at 4 K, taken at 7.85 mW laser diode output, with 0.5 mm slitwidths on our long-pass-filtered spectrometer, we see a broad peak with a highenergy tail (Figure 5.2.1-1). The width is characteristic of ensembles of quantum dots, as is the tail, which is a result of higher-energy excitonic states that result from state-filling.



Figure 5.2.1-1 Photoluminescence, Sample C. Short-pass-filtered laser source.

When the short-pass filter is removed from the laser diode, the low-energy luminescence of the laser diode, reflected off the surface of the sample, also registers in the spectrum of C. When we look at the unfiltered laser diode spectrum, we can see this luminescence (Figure 5.2.1-2). Here, we are modulating the laser diode with an applied voltage, that actually modulates the current through the laser diode. Hence, the power varies as the voltage, but the relationship is not quite linear. Yet, if we look at the quantum dot peak as the excitation power decreases, we see that the peak of the PL shifts to lower energies. This also agrees with observations that, at lower powers, only lower-energy recombination is observed.



Figure 5.2.1-2 Photoluminescence vs Laser Diode Power, Sample C. Unfiltered laser source.

We also looked at the PL as a function of FG-BG bias across the structure (Figure 5.2.1-3). Here we see no significant Stark shifting, which is reasonable, considering the voltage drop across the dot is only 1% of the applied voltage. Hence, a 10% change in wavelength would require approximately 10 V to be applied.



Figure 5.2.1-3 Photoluminescence vs. V_{FG}, Sample C

Finally, we compare the previous data with the power and filter dependence of S (Figure 5.2.1-4). Immediately, we see that the quantum dot peak is gone, as we would expect. However, even with the short-pass filter on the laser diode, the low-energy luminescence of the laser diode is still apparent. This is probably due to the fact that, when taking PL, we optimize the alignment by maximizing the signal from the wavelength we are trying to detect. In the case of S, since there was no quantum dot signal to optimize, we focused on the laser diode luminescence, which is simply reflected off the sample surface. In contrast, when we were measuring C, by optimizing the quantum dot peak, we were able to effectively spatially filter out some of the laser diode luminescence.



Figure 5.2.1-4 Photoluminescence vs Laser Diode Power, Sample S. All traces with short-pass filtered Laser Diode, except for top blue curve, at max power.

5.2.1.3 Current- and Capacitance-Voltage Characterization

To make sure the samples were working as expected, we took current-voltage (IV) and CV traces. Electrically, our structure should look something like a pair of back-to-back diodes: when cold and not illuminated, in some voltage range around zero, the device should be insulating and have a constant capacitance. The expected values and experimental methods are described elsewhere (§5.1.3.1).

The electrical connections to the samples simply consisted of squashing a bit of indium between a gold wire and the gold surface of an ohmic contact pad that had

been deposited onto the device surface. But, inconsistent results were obtained. The IV plot of C shows very different IV characteristics exhibited on different dates (Figure 5.2.1-5). We do see a region of small current around zero, and label this as our operating regime. But, outside of this region, the IV behaves quite peculiarly. First, the earlier mounting M1 has a much sharper turn-on at positive voltages, and a much slower turn-on at negative voltages, at zero illumination. A partial explanation of this could be that there was some parallel, diode-like current path available during the first mounting, that allowed current to flow at positive, but not negative, voltages. This doesn't explain things fully, since there would have to be a diode-like current path for the second mounting M2, oriented in the opposite direction, for the negative-voltage behavior to make sense. As we increase the intensity of the laser incident on the sample, we might expect to see an increase in the current flow at larger voltages, but the voltage range over which the sample is insulating should remain constant. Indeed, that seems to be the case, except that at negative voltages, especially for M1, there appeared a shoulder at about -2 V. As this negative-voltage current-shoulder has been a fairly general trend with this structure, to a greater or lesser extent with various mountings, it may involve an inherent asymmetry in the growth, possibly Si dopants floating up into the AlGaAs barrier near the BG; fortunately, the voltage range over which the sample remained insulating under illumination was broad enough that we did not need to worry overmuch about currents at the higher voltages. In fact, most of our measurements in the next section were performed using the second mounting.

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Figure 5.2.1-5 I-V vs Laser Diode Power, Sample C, two different mountings of the same sample, M1 and M2, shown.

The IV curve for S matches up quite nicely, having even less leakage current than C (note the scale, Figure 5.2.1-6). S also exhibits no shoulder, at least for the mounting exhibited.



Figure 5.2.1-6 I-V vs Laser Diode Power, Sample Sansa, 0 to 15 mW

The CV data are a bit more complicated, and were used to confirm the electrical behavior of these devices, as mentioned in the first paragraph of this section. We would expect to have a relatively constant capacitance in the insulating voltage region, while under no illumination. As described above (§5.1.3.1), we would expect the capacitance to rise under intense 800 nm-wavelength illumination, eventually saturating when the GaAs section of the insulating region is effectively 'metallized' by the rapid photoinjection of carriers. For C, the capacitance did saturate, but at a significantly higher capacitance than predicted (Figure 5.2.1-7). This may mean that a larger fraction of the insulating region of the sample was 'metallized', perhaps parts

of the AlGaAs barriers. We also see some strange behavior around -0.5 V, where it seems the capacitance isn't stable; perhaps current was being drawn at those voltages. For Sample S, the capacitance fluctuated as a function of voltage, much more than for Sample C (Figure 5.2.1-8). We did not concern ourselves with the fluctuations, though, as they amounted to only a fraction of the total capacitance. The average capacitance at saturation, though, did match up better for Sample S than for Sample C. So, keeping these things in mind, we took ± 1 V as our operating voltage range.



Figure 5.2.1-7 C-V vs Laser Diode Power, Sample C. Predicted saturation based on 0 mW.



Figure 5.2.1-8 C-V vs Laser Diode Power, Sample S. Predicted saturation based on 0 mW.

5.2.2 Loading and Unloading

Now that we had some idea of the devices' behavior under DC and AC voltages, with and without illumination, we were able to test them using our loading/unloading procedure. The procedure has many free parameters, though, so we spent some time feeling our way around parameter-space, experimentally optimizing the system. Then, we confidently performed several experiments, testing the properties of the storage lifetime.

5.2.2.1 Loading/Unloading Cycle, Oscilloscope Trace

The details of the design and implementation of the loading/unloading (L/UL) cycle are explained elsewhere (§5.1.3), but a brief overview with a few cogent details is in order, here. The cycle (Figure 5.2.2-1) begins with both sets of dots nominally empty, and a constant voltage applied to the front gate (FG) in reference to the back gate (BG) to balance out any built-in fields, $V_{FG} = V_{dc}$. Then we apply an amplitude voltage V_{amp} , so $V_{FG} = V_{dc} + V_{amp}$. This causes charge to accumulate on the gates, just like a capacitor. After a relaxation time, we then shine a pulse, NIR1 (800 nm, ~15 mW) for a time, T_L, to inject charge into the GaAs region, and load the quantum dots. We leave $V_{FG} = V_{dc} + V_{amp}$ for a total time T_{app}, before resetting $V_{FG} = V_{dc}$. After another relaxation time, we shine a second pulse, NIR2, identical except for a different duration T_{UL}, to unload the quantum dots. At this point, we are back to our initial condition.



Figure 5.2.2-1 Timing diagram of Loading/Unloading Cycle

Throughout this cycle, the current flowing to the sample is monitored by a) measuring the voltage across a load resistor in series with the sample, or b) using a transimpedance amplifier. Method (a) is very simple to interpret, but the data are noisy, and the circuit RC-time is slowed. Method (b) takes a little calculation to interpret, but the data are cleaner and the RC-time is much improved.

A sample scope trace (taken with transimpedance amplifier), shows a piece of this cycle with sample C (Figure 5.2.2-2 a). Not shown is the large current pulse that results from the application of V_{amp} . As marked, we see the current signature due to NIR1, and we see its duration T_L . The cycle continues with an abbreviated time axis, and the current pulse from the removal of V_{amp} is not shown. We then see the end of

the cycle; we see the current signature of NIR2, for duration T_{UL} . This is the most important part of the cycle, since the current that flows due to NIR2 is a measure of the amount of charge stored in the device. Throughout the cycle, we see the oscillation of 60 Hz noise.



Figure 5.2.2-2 Raw scope signal. Signals from changes in V_{FG} not shown; time scale abbreviated. Shows (a) complete L/UL cycle for Sample C; same cycle but without loading pulse NIR 1 showing NIR2-associated photocurrent background (b); full cycle minus background photocurrent (c); and the same difference trace for Sample S (d).

Whenever light shines on the sample, there is some photoresponse, which results in a large photocurrent when voltage is applied. Hence the discontinuities around the NIR1 signature. There is even some photovoltage (a shifting of the IV curve) when no voltage is applied, and we see a small blip from this when NIR2 ends. To isolate the NIR2 current due to photoresponse and that due to charge storage, we repeat the same procedure without NIR1. With no NIR1, no charge should be loaded into the quantum dots; the only NIR2 signature should be the photoresponse background.

In the next plot (Figure 5.2.2-2 b), we see the same signal, this time with NIR1 removed. This is our background, which we subtract from the foreground to get the NIR2 charge-unloading signal (Figure 5.2.2-2 c). Then, because the voltage is proportional to the current, the integrated signal is proportional to the charge flowing off the sample, which is in turn proportional to the stored charge. To eliminate the 60 Hz noise, we fit the non-pulse data to 60 Hz sine-waves and subtract the residual from the raw trace.

We can also look at the identical difference-curve of S, taken under similar conditions to the previous C data (Figure 5.2.2-2 c). Here, we see that, at NIR2, there is no signature that changes depending on NIR1's presence. Hence, there is no charge-dependent signal, so no charge storage.

5.2.2.2 Optimizing Loading, Unloading Pulse Energies

The cycle described above and in other sections (§5.1.3) depends on having an excess of flux during both the NIR1 and NIR2 pulses. Because the efficiency of our

fiber-coupling system was so low (§5.1.3.1), we kept the NIR pulses at the maximum power we could, ~15 mW, and adjusted the pulse lengths, T_L and T_{UL} , to make sure we had enough flux. All of these (§5.2.2.2) measurements were performed using the load resistor setup, not the transimpedance amplifier, with V_{app} (V_{dc}) = 0.2 (0.0) V, and at 4 K. First, we picked a T_L (0.1 ms), and varied T_{UL} , while measuring the integrated charge over a 10 ms window triggered on NIR2. By varying T_{UL} with all other conditions constant, we were able to measure a saturation time T_{UL-sat} , where no more charge could be extracted by increasing the NIR2 flux (Figure 5.2.2-3). We then observed how T_{UL-sat} varied when T_{app} varied, to get an idea of any time-dependence on the extraction of charge. The roughly decreasing nature of T_{UL-sat} with increasing T_{app} lead us to choose 2.5 ms as a safe T_{UL-sat} for the rest of our experiments.



Figure 5.2.2-3 Charge Extracted vs T_{UL} at varioius T_{app}.

We then held T_{app} constant at 6 ms, and took the same Q vs T_{UL} curves while varying T_L . The data in this case was fairly noisy, and some of the data didn't fit the model at all (Figure 5.2.2-4). But, in the end, the fits to the data shown didn't give us much more information than looking at the data itself. That is, no drastic changes in the saturation behavior of Q vs T_{UL} were observed as T_L was varied over 2 orders of magnitude. We estimated that T_{UL} and T_L of 2.5 and .5 ms, respectively, should be sufficient to put the system in a stable regime, where small variations in T_{UL} and T_L should have minimal impact on the charge storage cycle.



Figure 5.2.2-4 Charge Extracted vs. T_{UL} at various T_L.

It should be kept in mind that these measurements of stable pulse lengths may be affected by the charge measurement apparatus - i.e., the times are probably longer than they would be by using the transimpedance amplifier to measure the charge, as the sample can only equilibrate as fast as the RC time constant of the overall circuit.

5.2.2.3 Storage Lifetimes

Armed with stable, though arbitrary, choices for T_{UL} and T_L , we then measured the storage lifetime of C by recording the stored charge as a function of T_{app} . As mentioned earlier, T_{app} is a good approximation for the total time between the loading and unloading of charge in the quantum dots. In this case, we took {V_{amp}, V_{dc}, temperature} to be {0.2 V, 0.3 V, 80 K}, and used the transimpedance amplifier instead of the load resistor. The effect of the V_{dc} offset is explained in (§5.1.3.1). When we plot the stored charge vs. time in a log-linear format (Figure 5.2.2-5), some interesting facts emerge. Normally, a single exponential decay curve plotted in a log-linear fashion would produce a single straight line with constant slope, the slope being the exponential coefficient. In fact, we see at least two different slopes; we see that the short-time data ($T_{app} < 1$ s) has a relatively fast decay time of ~1 s, while the long-time data ($T_{app} \ge 1$ s) has a much longer decay time of ~20 s. Though dual, shorter, decay times for charge stored in quantum dots have been reported elsewhere[27], the cause of the dual delay times is still unclear. Characteristics of these storage lifetimes are investigated below.



Figure 5.2.2-5 Stored Charge vs. T_{app} at 80 K. Fit to two separate storage times, 1.1 ± 0.2 and 18 ± 6 s, with amplitudes 1.03 ± 0.02 and 0.71 ± 0.06 e/QD, respectively.

5.2.2.4 Storage Lifetimes vs DC Offset

As described in more detail elsewhere (\$5.1.3.1) V_{dc} applied throughout the cycle changes the baseline amount of charge in the quantum dots. This is a result of the fact that what we measure during the NIR2 pulse is the *difference* in charge between the initial state, and the state right after NIR1. There may also be built-in fields resulting from asymmetries in the growth, which nevertheless can affect the storage.

So it was necessary to observe the behavior of the storage lifetimes on V_{dc} . For this data, we took $V_{app} = 0.2$ V, at 15 K, and used the transimpedance amplifier.

We took data for $-0.1 \text{ V} \le V_{dc} \le 0.7 \text{ V}$, with a total voltage swing, keeping in mind $V_{app} = 0.2 \text{ V}$, of $-0.1 \text{ V} \le V_{app} + V_{dc} \le 0.9 \text{ V}$. We see that, for the V_{dc} investigated, there is no appreciable change in either lifetime (Figure 5.2.2-6). However, the long lifetimes do seem a bit more stable, inasmuch as they fit better to exponential decay curves, at $V_{dc} \sim 0.3 \text{ V}$. So, for much of the data taken on lifetimes, we took $V_{dc} = 0.3 \text{ V}$.



Figure 5.2.2-6 Decay Lifetimes vs. V_{dc} at 15 K.

5.2.2.5 Charge Storage vs Voltage Amplitude

This device is designed so that the amount of charge stored is determined completely by the voltage applied during NIR1. We checked the amplitude of the stored charge at 5 K, for { V_{dc} , T_{app} } = {0.3 V, 20 ms}, using the transimpedance amplifier, as a function of V_{amp} . In principle, the stored charge should be linear in V_{amp} , with a coefficient of ~6 electrons/QD/Volt (§5.1.3.1). Our measured values were between 3.7 and 4.7 e/QD/V (Figure 5.2.2-7). Though we are encouraged by the linearity, the discrepancy between the estimated and measured values is yet troubling. More troubling, still is the observation that subsequent experiments in an optical cryostat have yielded results that suggest up to 12 e/QD/V. The predicted value is based only on the geometry of the device and the quantum dot density. It may be that the different illumination procedures result in different effective areas, but results are not yet conclusive.



Figure 5.2.2-7 Charge Stored vs. V_{amp} , at 5 K, V_{dc} of 0.3 V, T_{app} of 20 ms. Two effective capacitances are found, 3.75 ± 0.05 and 4.7 ± 0.1 e/QD/V.

5.2.2.6 Storage Lifetimes vs Temperature

Our final preliminary characterization of this device was to observe the behavior of the storage lifetimes as a function of sample temperature. As discussed in \$5.1.3.1, the binding energies of electrons (holes) in these quantum dots is predicted to be ~330 (230) meV. For charges bound by energies this large, one should see no temperature dependence of carrier escape, up to 100 K and higher. Indeed, this is exactly what is observed (Figure 5.2.2-8), as we set {V_{amp}, V_{dc}} = {0.2 V, 0.3 V} and measure the lifetimes at various temperatures from 5 to 80 K. The lifetimes we measure range from 1 second to a minute or more.



Figure 5.2.2-8 Lifetimes vs. Temperature. Data spans two different days.

5.3 Summary

We succeeded in developing a system that allows careful control of quantum dot population. This population is determined, not by the growth direction electric field E_z during the storage time, but rather by E_z imposed during a loading NIR pulse. Therefore, in principle, the device should also allow control of the E_z tuning of coupled quantum dots at a constant quantum dot charge population.

6 NIN Loading Experiment: FTIR/MIR

6.1 Theoretical Background

6.1.1 MIR Experiments

The above section (§5) detailed our typical loading, unloading experiment. If, on the other hand, the NIR2 pulse (§5.1.3.4) wasn't used, and instead the sample was illuminated with broadband MIR light, the quantum dots might still be unloaded, and the signature current might still be seen. The idea is that, during the time that the V_{amp} was off, the quantum dots are biased such that the stored charge is drawn toward the center of the device. If the quantum dots absorb MIR light of the appropriate energy, either carrier of charge could be promoted to the quasicontinuum of the GaAs band-edge. The extant field would then accelerate the freed charge away toward the other quantum dot layer, and the corresponding amount of compensating charge on the gates would be released. If the MIR were then chopped, lock-in techniques could be used to monitor this small current during the unloading phase.

This experiment is quite ambitious, so a first step is to leave the MIR on in a continuous-wave mode, while running the loading experiment as originally planned. Then, one might expect the lifetime of the storage to be reduced by the MIR-induced ionization. One issue is that the device is highly sensitive to interband light. So, when placing it in the beam path of an FTIR, pains must be taken to filter the MIR beam. Not only the visible HeNe alignment laser must be removed, but also the high-energy interband tail of the broadband MIR source. For these purposes, room-temperature bulk InAs is a fine filter as it absorbs all light above about 350 meV.

6.1.2 Expectations for NIN Experiments

There are several features of the data from these experiments that could be predicted. First, the binding energies of the quantum dots are expected to be quite large: \sim 330 meV for electrons and \sim 230 meV for holes. This range has been borne out by other experiments. For binding energies this large, the temperature would have to approach 2700K for k_BT to become comparable. So, purely thermal ejection is not expected for these quantum dots in reasonable temperature ranges. In fact, significant escape has not been observed for rather large temperatures, in other experiments on similar quantum dots.

The quantum dot to quantum dot dimensional fluctuations of about 10% should result in a similar spread in binding energies, and hence lifetimes, as well as other spectral features that might be observed.

The unloading current is insensitive to which carrier type, electrons or holes, is being ejected or cancelled within the quantum dots. This means that if a MIR light source were used to eject charges from the quantum dots, and the resultant current were measured, some other method of de-convolving the data would have to be used to separate out the ejection of electrons versus holes. One such method might be FTIR.

The quantum dots should be able to store only on the order of 10 electrons per dot. If the device were biased so as to store more than that, the excess shouldn't be successfully stored. Furthermore, as the filling of the quantum dots increases, the incremental charge should be bound less tightly, and so the lifetime of that incremental charge should decrease. Also, if the wavelength of MIR absorption necessary to eject charge were measured, as the filling increased, there should appear a red tail, as lower energies of light excite the least-bound charges.

If, while stored, the device is biased so that charges should tunnel toward the center, the lifetime of storage should decrease. To a limited extent, the inverse should also be true, when charges are biased toward their nearest barrier. Likewise, in the two bias cases, the MIR absorbed should develop red and blue tails, respectively. Finally, if biased sufficiently while stored, all of the charge should tunnel out of the quantum dots, and the lifetime and the absorption should be quenched.

6.2 Experimental Data

To further characterize the charge storage, we began mid-infrared spectroscopy on C. We first moved the sample C back into the optical cryostat, and simply coupled the NIR laser into the sample by shining on it through a zinc-selenide (ZnSe) window. We confirmed that the charge storage still functioned properly, and then moved the entire setup into the FTIR spectrometer, the setup of which is described elsewhere (§9.5). Most importantly, we were then able to simultaneously shine our NIR laser onto the sample, as well as the broadband MIR light from a Globar® source in the FTIR.

Our goal was to use the sample as a MIR detector. The idea is that the unloading current signal results from charge being released from the quantum dots, and that this would occur regardless of whether electrons or holes were being eliminated, and regardless of the method of elimination. So, by using a broadband MIR source that covers the frequency range that includes the expected binding energies of electrons and holes (330 and 230 meV, respectively), a current signal might be seen as a result of the MIR source, taking the place of NIR2. The loading/unloading cycle would then be completed by directly kicking the electrons and holes out of the quantum dots, rather than by creating more electron-hole pairs in the GaAs region, as with the original cycle. In fact, by using the unloading current, which should be proportional to the MIR intensity, to map out the FTIR's interferogram, we could then map out the exact MIR energies that were used in charge ejection.

6.2.1 Storage Lifetimes vs V_{dc} vs MIR

As a first step, we measured the dependence of the charge storage on the MIR. Using the fact that the steady-state population of the quantum dots, Q_{dc} , is proportional to V_{dc} , we sought whether the higher the energy levels occupied by the varying charge, Q_{amp} , would be ejected more readily by the MIR. We expected the lifetimes to shorten under constant MIR illumination, especially under high V_{dc} . In

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this case, we investigated for $T_{app} \le 1s$, and so should only capture the shorter storage times.

When we look at data on the storage lifetime behavior taken over several days, we see two main differences (Figure 6.2.1-1). First, the lifetimes, even when not illuminated by MIR, change over several orders of magnitude, in stark contrast to data taken earlier, in the non-optical cryostat. Second, though the lifetimes are, in general, suppressed by the MIR, no drastic change occurs at high V_{dc} , in the range investigated.



Figure 6.2.1-1 Short Lifetimes vs V_{dc} , with (blue) and without (red) cw MIR illumination. Data taken over several days (shapes).

It should be noted that we had to filter the MIR beam to block the alignment HeNe laser from hitting the sample. As seen in the next subsection, we had to longpass the MIR beam, as well. We used a piece of InAs wafer to block all energies higher than the InAs (300 K) bandgap. Initially, the InAs filter was a bit small, so we had to block the remaining beam completely, leading to approximately 50% lower throughput than the June data sets. Also, the 3 June data was taken under dubious alignment conditions, such that the stored charge seemed to be about 50% smaller than normal. Therefore, this data should be taken as preliminary in nature.

6.2.2 Step-Scan MIR Spectroscopy

By chopping the MIR beam at about 200 Hz and using standard lock-in techniques, we attempted to use the sample as a MIR photodetector. In this case, we used the NIR1 loading pulse, but not the NIR2 unloading pulse. Thus, any unloading should be entirely due to the MIR intensity. As mentioned in the previous section, we partially filtered the MIR beam, but allowed some unfiltered MIR to hit the sample. Under these conditions, we were able to see an interferogram (Figure 6.2.2-1). The corresponding spectrum is also given, along with the calculated 1200 K blackbody spectral curve, both normalized and scaled up by 800 times(Figure 6.2.2-2). Since the Globar® source is expected to approximate a blackbody, neglecting beamsplitter effects, the IR intensity hitting C should be about the same as that calculated, up to some overall factor. In the spectra, we clearly see a feature at about 1.5 eV. This is the energy of GaAs absorption, and we can interpret this as the small piece of Globar® emission at these NIR energies. In fact, this is the equivalent of having a weak, chopped NIR source, unloading C in just the same manner as the NIR2 pulse did in the standard cycle.



Figure 6.2.2-1 Step-Scan Interferogram from Sample C. Unfiltered MIR source.



Figure 6.2.2-2 Step-Scan spectra, Sample C (red). Unfiltered MIR source, approximately 1200 K. 1200 K Blackbody curves, normed (blue) and scaled to fit (green).

From the normalized blackbody curve, we see that a huge fraction of the intensity of the MIR beam is actually centered near our range of interest, 200-350 meV. The shape of the high-energy tail of the GaAs feature matches quite well with the scaledup version of the blackbody curve, indicating that the absorption there may be limited by the intensity of NIR light in the high-energy blackbody tail, rather than by the density of states of GaAs, near the conduction band. In fact, the temperature range suggested by the low-energy turn-on, according to the temperature dependence of the GaAs band-gap, is 240-250 K. Unfortunately, the temperature was not recorded for this measurement.

When the entire MIR beam was long-pass filtered with the InAs, so that the GaAs was not excited, no sensible interferogram was obtained. Clearly, these are only preliminary measurements on this system, and much can be done to reduce the noise and take advantage of the (relatively) enormous intensities at our energies of interest.

6.3 Summary

We attempted some elementary MIR spectroscopy of charges stored in quantum dots, using the NIN system we developed. We observed definite lifetime-shortening, indicating that MIR light of the appropriate energies can and does eject charge from our quantum dots. We tried to extract more definite spectroscopic data using our sample as a photodetector, but were only marginally successful. We note that these results are encouraging for future MIR and other experiments using this system, as it is capable of tuning quantum dots with a growth-direction electric field at a constant charge density.

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7 Conclusion

7.1 Summary and General Remarks

Initial project goals included: 1) To engineer systems capable of interrogating bound electrons in quantum dots with far-infrared (FIR) energy splittings; 2) To experimentally observe dynamics in such systems; 3) To study that system as a function of charge density; 4) To study that system as a function of the energy splitting itself; 5) To analyze the suitability of that system for the purposes of quantum information processing. As work progressed, these initial goals changed, but it is still interesting to consider the degrees of success toward each of these very ambitious goals.

We were clearly successful in our first goal. We used the well-studied technology of quantum dots embedded in metal-insulator-semiconductor field-effect transistor (MISFET) structures, which had previously been used to investigate single quantum dot layers having mid-infrared (MIR) energy splittings (§2.3.1), and incorporated coupled quantum dots to achieve FIR splittings, predicted to be in the 10 meV range (§3.2.3 and §3.2.4). Furthermore, we took another existing design of coupled quantum dots embedded in a metal-insulator-metal (NIN) structure, previously used to study microphotoluminescence (§5.1.1), and extended it for use as a charge storage device for single quantum dot layers (§5.1.3). We achieved charge storage times on the order of minutes (§5.2.2.6). In principle, this device could also be

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implemented with coupled quantum dots, which would again give us the desired FIR splittings.

In our second goal, we were also quite successful. In the MISFET system, we clearly observed electron absorption in the energy range of 7 to 15 meV (§3.2.3). A more fundamental, but no less interesting, type of dynamics was observed in the NIN system. There, we experimentally confirmed no temperature dependence for temperatures ranging from 5 K to 80 K and above (§5.2.2.6).

Our third and forth goals met with qualified successes. In the MISFET structure, we were able to modify the charge density and the energy splittings by changing the growth-direction electric field (§3.2.3). Indeed, we were able to tune the charge state through a range of 0 to 12 electrons per quantum dot (§3.2.2). However, due to issues inherent in the design, we were unable to modify those two parameters independently (§3.2.4). We attempted to modify the MISFET structures using DX centers (§4.1), but results were inconclusive (§4.2). The NIN device, however, did allow study of the storage behavior as a function of the total charge density, by varying a voltage offset (§5.1.3.1). There, again, we found very little dependence of the storage lifetime on charge density (§5.2.2.4).

Our fifth goal was also partially successful. In the MISFET structure, we were able, through observing the saturation of the FIR absorption, to deduce a lower bound on the longitudinal relaxation time T_1 for the coupled quantum dots to be in the range of 20 - 85 ps (§3.2.4). Such a low T_1 is very unexpected, and we suspected electron-electron interactions both within the quantum dots and between quantum

dots and the nearby wetting layer to be responsible. Certainly, for study of dynamics of few electrons in coupled quantum dots, the NIN design arises as a fitting successor to that of the MISFET. In that, rather limited, sense, the NIN device has also been partially analyzed for quantum information processing suitability.

7.2 Future Directions

Here, we detail a few comments on the directions this project and those like it could and should go in the future. We will present only the most general of concepts, all involving the NIN device, as the details are best worked out by the experimentalists involved. Should there be a drought of ideas from unsuccessful brainstorming, the author's clouded mind is always available.

7.2.1 Near-term

The next steps for this project are clear. MIR detection via capacitive coupling (§6) has shown signs of promise of resolving the bound-to-continuum transitions, and should be pursued vigorously. Similarly, MIR transmission through the sample also appears straightforward, as a further confirmation of the spectral characteristics of the stored charge, and the bound-to-bound absorption mechanisms. In the bound-to-bound case, though, polarization has to be maintained in the plane, and single-pass absorption may not yield much signal. A wedged-waveguide geometry (§2.4.1) may be in order, in that case. Observation of the GaAs band-edge absorption (§6.2.2) should be investigated further, especially as a means of confirming the lattice temperature optically. Finally, it should be relatively simple, using circularly

polarized light for NIR1 (§5.1.3.2), to write spin-polarized electrons into the quantum dots, and read out the spin polarization using the opposite handedness for NIR2.

In terms of system upgrades, there are only a few immediate ideas to mention. First, the windows currently used for NIR/MIR transmission are ZnSe, and should probably be switched to CaF₂. The data collection apparatus should undergo another level of automation, despite inconveniences to users. Lastly, since the storage mechanism is robust up to 80 K, using liquid nitrogen instead of helium would offer multiple benefits including savings of both time and money.

7.2.2 Medium-term

The device design may undergo a couple of improvements. The RC time constant of the device, critical as the experimental repetition rate increases, should be reduced by shrinking the device area. This will also necessitate a more sensitive measurement apparatus, as the current system measures total charge, which should also be proportional to area. Another type of bound-to-continuum detection that may be used is in-plane photocurrent (§2.4.2). This would involve contacting the wetting layers (WL), and the process engineering may be tricky.

In the growth of subsequent samples, several parameters should be studied for adjustment. If the operating voltage range can be kept more or less constant, thinner tunneling barriers would increase the stored-charge-to- V_{amp} proportionality (§5.1.3.1), thus assisting the access of the filled-quantum dot and WL states. Clearly,

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a sample with coupled quantum dots on at least one side of the device, should be grown and tested soon. For FIR transmission spectroscopy, one might use a waveguide structure as with the MISFET devices (§3.1.1.2). Care should be taken to calculate the absorption of the doped gates; modulation-doped quantum well (QW) gates may be in order. Finally, there is no reason, in principle, why this device cannot be multiplexed; that is, an N-period AlGaAs/GaAs superlattice quantum dot structure could provide N charge-separation layers and 2N sites for charge storage, with advantages over the N=1 case studied here. This would most directly aid MIR transmission experiments, but benefits to FIR experiments are clear, as well.

With coupled quantum dots, MIR measurements can also be done, especially if bound-to-continuum excitation from the coupled states can be resolved from each other. But FIR spectroscopy would be the main focus, and transmission and saturation spectroscopy should proceed as it had before (§3.2.3, 3.2.4).

7.2.3 Long-term

In the future, co-opting of μ -PL technology should proceed posthaste. Proposed experiments implementing the ac Stark effect should be thoroughly investigated, and μ -PL should help. This may require lower quantum dot density and a transparent front gate contact. The lower quantum dot density will lower photocurrent signal, however.

FIR photonic cavity integration should also be seriously considered for our coupled quantum dots. The location of gate layers may be tricky; placing

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modulation-doped single-subband QWs at cavity vertical nodes may help. Current efforts at FIR generation within the cavity via a femtosecond laser pulse may prove helpful, for characterization purposes.

Major advances in the FEL Pulse-Slicer technology should enable photon echo experiments. This would be especially interesting in combination with the previous two long-term ideas.

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9 Appendices

9.1 Sample Growths

9.1.1 MISFET Samples

9.1.1.1 Included in Experiments

9.1.1.1.1 B (90116-3)

Featured in both linear and saturation spectroscopy experiments.

Semi-insulating GaAs Substrate

GaAs 1000Å

AlAs/GaAs short-period superlattice $40x(20\text{\AA}/20\text{\AA})$

- GaAs 800Å
- n-GaAs 200Å (4e18 cm⁻³)
- GaAs 250Å

InAs QD layer, grown at $4x \rightarrow 2x$ transition temperature, sample not rotated

during QD growth

GaAs 70Å

InAs QD layer, same as before

GaAs 300Å

AlAs/GaAs short-period superlattice 27x(30Å/10Å)

GaAs 50Å

Surface

9.1.1.1.2 A (90210-1)

Featured in linear spectroscopy experiments.

Semi-insulating GaAs Substrate

GaAs 1000Å

AlAs/GaAs short-period superlattice 40x(20Å/20Å)

GaAs 800Å

n-GaAs 200Å (4e18 cm⁻³)

GaAs 250Å

InAs QD layer, grown at $4x \rightarrow 2x$ transition temperature, sample not rotated

during QD growth, 10Å recipe

GaAs70ÅInAs QD layer, same as before, except normal (not 10Å) recipeGaAs300ÅAlAs/GaAs short-period superlattice27x(30Å/10Å)GaAs50ÅSurface

9.1.1.2 Other Growths

9.1.1.2.1 Strain-induced quantum dots (990222)

Never pursued as a coupled quantum dot experiment.

GaAs not intentionally doped substrate + buffer

300Å $Al_{0.3}Ga_{0.7}As$ Si delta doping 10¹¹ cm⁻² 100Å $Al_{0.3}Ga_{0.7}As$ GaAs 85Å $Al_{0.3}Ga_{0.7}As$ 100Å Si delta doping 10¹¹ cm⁻² $Al_{0.3}Ga_{0.7}As$ 300Å 5000Å GaAs InAs QD layer, 500Å in diameter 250Å $Al_{0.3}Ga_{0.7}As$ 85Å GaAs $Al_{0.3}Ga_{0.7}As$ 25Å GaAs 75Å Al_{0.3}Ga_{0.7}As 100Å Si delta doping 10¹¹ cm⁻² Al_{0.3}Ga_{0.7}As 900Å 100Å GaAs Surface

9.1.1.2.2 Reference sample for strain-induced quantum dots (990221)Not necessary, due to Sample 990222 not being used.

GaAs not intentionally doped substrate + buffer

 $Al_{0.3}Ga_{0.7}As$ 300Å

 Si delta doping 10^{11} cm⁻²

 $Al_{0.3}Ga_{0.7}As$ 100Å

 GaAs
 85Å

 $Al_{0.3}Ga_{0.7}As$ 100Å

 Si delta doping 10^{11} cm⁻²

 $Al_{0.3}Ga_{0.7}As$ 100Å

 Si delta doping 10^{11} cm⁻²

 $Al_{0.3}Ga_{0.7}As$ 300Å

 GaAs
 5000Å

InAs QD layer, 500Å in diameter [I assume the QDs are not included in the

reference sample, but that was not explicit in my records.]

250Å Al_{0.3}Ga_{0.7}As GaAs 85Å $Al_{0.3}Ga_{0.7}As$ 25Å 75Å GaAs Al_{0.3}Ga_{0.7}As 100Å Si delta doping 10¹¹ cm⁻² 900Å $Al_{0.3}Ga_{0.7}As$ 100Å GaAs Surface

9.1.2 MISFET-DX Growths

9.1.2.1 Coupled QDs with DX 90918-2

Back Gate doping too low for ohmic contact.			
Semi-insulating GaAs substrate + buffer layer			
Al _{0.3} Ga _{0.7} As	200Å		
Si delta doping 2*10 ¹¹ cm ⁻²			
Al _{0.3} Ga _{0.7} As	100Å		
GaAs 85Å			
Al _{0.3} Ga _{0.7} As	100Å		
Si delta doping 2*10 ¹¹ cm ⁻²			
Al _{0.3} Ga _{0.7} As	4500Å		
Al _{0.3} Ga _{0.7} As	100Å n-doped 10^{18} cm ⁻³		
Al _{0.3} Ga _{0.7} As	300Å		
GaAs 15Å			
InAs QD layer, same QDs and recipe as 90116-3			
GaAs 70Å			
InAs QD layer, same QDs and recipe as 90116-3			
GaAs 70Å			
Al _{0.3} Ga _{0.7} As	450Å		
GaAs 100Å			
Surface			

9.1.2.2 Coupled QDs with DX 90918-3

Back Gate doping too low for ohmic contact.				
Semi-insulating GaAs substrate + buffer layer				
Al _{0.3} Ga _{0.7} As	200Å			
Si delta doping 2*10 ¹¹ cm ⁻²				
Al _{0.3} Ga _{0.7} As	100Å			
GaAs 85Å				
Al _{0.3} Ga _{0.7} As	100Å			
Si delta doping 2*10 ¹¹ cm ⁻²				
Al _{0.3} Ga _{0.7} As	4500Å			
Al _{0.3} Ga _{0.7} As	100Å n-doped 10^{18} cm ⁻³			
Al _{0.3} Ga _{0.7} As	300Å			
GaAs 15Å				
InAs QD layer, same QDs and recipe as 90116-3				
GaAs 60Å				
InAs QD layer, same QDs and recipe as 90116-3				
GaAs 70Å				
Al _{0.3} Ga _{0.7} As	450Å			
GaAs 100Å				

9.1.2.3 Coupled QDs with DX 91213-1

Back Gate and DX layer doping too low for ohmic contact.

Semi-insulating GaAs substrate + buffer layer

200Å $Al_{0.3}Ga_{0.7}As$ Si delta doping 5*10¹¹ cm⁻² 100Å Al_{0.3}Ga_{0.7}As GaAs 85Å Al_{0.3}Ga_{0.7}As 100Å Si delta doping 5*10¹¹ cm⁻² 4500Å $Al_{0.3}Ga_{0.7}As$ 100Å n-doped 10^{18} cm⁻³ Al_{0.3}Ga_{0.7}As 300Å $Al_{0.3}Ga_{0.7}As$ 15Å GaAs InAs QD layer, same QDs and recipe as 90116-3 70Å GaAs InAs QD layer, same QDs and recipe as 90116-3 70Å GaAs $Al_{0.3}Ga_{0.7}As$ 450Å 100Å GaAs

9.1.2.4 Coupled QDs with DX 91213-2

Back Gate and DX layer doping too low for ohmic contact.

Semi-insulating GaAs substrate + buffer layer

200Å $Al_{0.3}Ga_{0.7}As$ Si delta doping 5*10¹¹ cm⁻² 100Å Al_{0.3}Ga_{0.7}As GaAs 85Å Al_{0.3}Ga_{0.7}As 100Å Si delta doping 5*10¹¹ cm⁻² 4500Å $Al_{0.3}Ga_{0.7}As$ 100Å n-doped 10^{18} cm⁻³ Al_{0.3}Ga_{0.7}As 300Å $Al_{0.3}Ga_{0.7}As$ 15Å GaAs InAs QD layer, same QDs and recipe as 90116-3 60Å GaAs InAs QD layer, same QDs and recipe as 90116-3 70Å GaAs $Al_{0.3}Ga_{0.7}As$ 450Å 100Å GaAs

9.1.2.5 Coupled QDs with DX 00110-1

Finally realized error in growth structure. See dissertation body.

Semi-insulating GaAs substrate + buffer layer

500Å $Al_{0.3}Ga_{0.7}As$ Si delta doping 2*10¹² cm⁻² 100Å Al_{0.3}Ga_{0.7}As GaAs 85Å $Al_{0.3}Ga_{0.7}As$ 100Å Si delta doping 2*10¹² cm⁻² 4500Å $Al_{0.3}Ga_{0.7}As$ 200Å n-doped 2*10¹⁸ cm⁻³ $Al_{0.3}Ga_{0.7}As$ $Al_{0.3}Ga_{0.7}As$ 300Å 15Å GaAs InAs QD layer, same QDs and recipe as 90116-3 (not rotated, see note) 70Å GaAs InAs QD layer, same QDs and recipe as 90116-3 70Å GaAs $Al_{0.3}Ga_{0.7}As$ 450Å 100Å GaAs

NB: If looking at half-wafer epilayer surface, top-down, so that it looks like a letter "D", the low density side is toward the top of the "D", and the high density $(10^{10} \text{ cm}^{-2})$ side is toward the bottom of the "D"

9.1.2.6 Coupled Quantum Wells with DX 00110-2

Quantum Wells to compare to Sample 00110-1. Finally realized error in growth structure. See dissertation body.

Semi-insulating GaAs substrate + buffer layer

200Å $Al_{0.3}Ga_{0.7}As$ Si delta doping 2*10¹² cm⁻² 100Å Al_{0.3}Ga_{0.7}As 85Å GaAs $Al_{0.3}Ga_{0.7}As$ 100Å Si delta doping 2*10¹² cm⁻² $Al_{0.3}Ga_{0.7}As$ 4500Å 200Å n-doped 2*10¹⁸ cm⁻³ $Al_{0.3}Ga_{0.7}As$ 300Å $Al_{0.3}Ga_{0.7}As$ 75Å GaAs $Al_{0.3}Ga_{0.7}As$ 25Å 85Å GaAs $Al_{0.3}Ga_{0.7}As$ 450Å GaAs 100Å

Surface

9.1.2.7 MISFET-DX with QD redesign (00925A)

Quantum Wells instead of QDs, but QWs spaced too closely together. See note.

330Å GaAs AlAs/GaAs SPS 20x(10Å/10Å) 130Å GaAs AlAs/GaAs SPS 100x(1.5Å/3.5Å) Si delta doping 1 Å AlAs/GaAs SPS 20x(1.5Å/3.5Å) 85Å GaAs AlAs/GaAs SPS 20x(1.5Å/3.5Å) Si delta doping 1 Å AlAs/GaAs SPS $2x(2.3\text{\AA}/5.3\text{\AA})$ AlAs/GaAs SPS 2x(3Å/4.5Å) AlAs/GaAs SPS 2x(3.8Å/3.8Å) AlAs/GaAs SPS 2x(4.5Å/3.0Å) AlAs/GaAs SPS $2x(5.3\text{\AA}/2.3\text{\AA})$ AlAs/GaAs SPS 2x(6.0Å/1.5Å) AlAs/GaAs SPS 2x(4.5Å/.5Å) AlAs 650Å 50Å GaAs

AlAs 50Å

AlAs/GaAs SPS 2x(6.8Å/.8Å)

AlAs/GaAs SPS 2x(6.0Å/3.0Å)

AlAs/GaAs SPS 2x(5.3Å/2.3Å)

AlAs/GaAs SPS 2x(6.0Å/4.0Å)

AlAs/GaAs SPS 2x(5.0Å/5.0Å)

AlAs/GaAs SPS 2x(4.0Å/6.0Å)

AlAs/GaAs SPS 20x(3.0Å/7.0Å)

AlAs/GaAs SPS 200x(1.5Å/3.5Å)

GaAs 10Å

InGaAs 45Å

GaAs 10Å [but had ordered 60Å here, so next growth is correction]

InGaAS 55Å

GaAs 10Å

AlAs/GaAs SPS 490x(1.5Å/3.5Å)

GaAs 100Å

9.1.2.8 MISFET-DX with Quantum Well redesign Tam (010215A)

Larger spacing between QWs. See Sample 00925A.

GaAs 1000Å

AlAs/GaAs SPS 75x(20Å/20Å)

GaAs 1000Å

30%AlGaAs 500Å 5.7Å Si delta doping 100Å 30%AlGaAs 85Å GaAs 100Å 30%AlGaAs 5.7Å Si delta doping 25Å 40%AlGaAs 50%AlGaAs 25Å 60%AlGaAs 25Å 25Å 70%AlGaAs 25Å 80%AlGaAs 90%AlGaAs 25Å 650Å AlAs GaAs 50Å 50Å AlAs 90%AlGaAs 25Å 25Å 80%AlGaAs 25Å 70%AlGaAs 50Å GaAs 60%AlGaAs 25Å 25Å 50%AlGaAs 50Å GaAs

40%AlGaAs		25Å	
Al _{0.3} Ga _{0.7} As-DX		200Å	
30%AlGaAs		1000Å	
GaAs	10Å		
InGaAs	45Å		
GaAs	60Å		
InGaAs	55Å		
GaAs	10Å		
30%AlGaAs		2450Å	
GaAs 100Å			

9.1.3 NIN Samples

9.1.3.1 Sample C, Cersei (9030130A)

Semi-insulating GaAs substrate				
Smoothing superlattice				
30%AlGaA	As	200Å		
n-GaAs	1000Å	$10^{18} \mathrm{cm}^{-3}$		
GaAs	100Å			
30%AlGa	As	5000Å		
C •	200 %			

GaAs 300Å

InAs QDs 1.85 monolayers

GaAs 2000Å

InAs QDs 1.85 monolayers

GaAs 300Å

30%AlGaAs 5000Å

GaAs 100Å

n-GaAs 1000Å 10¹⁸ cm⁻³

9.1.3.2 Sample S, Sansa NIN without QDs (031204B)

Should be exactly like Sample C, Cersei, but without InAs at all.

9.2 Processing

9.2.1 General Procedure

9.2.1.1 Cleaning

9.2.1.1.1 General Cleaning

- Use dedicated glassware, tweezers for every step.
- 5 min. in cold Acetone
- 5 min. in warm* Methanol (*if dedicated, limited, no-flashfire hotplate exists, otherwise use cold Meth)
- 5 min warm* Isopropanol
- 5 min cold DI water
- After every touch to sample, rinse and blow-dry with DI, and same for tweezers

- * After hot steps, blow sample cool before rinsing
- Dishes generally only need Ace step before rinsing and going into oven to dry
- Use 120 degree oven for glassware, 90 degree oven for photoresist (later)

9.2.1.1.2 Pre-E-beam Cleaning

Before e-beam, do etch #1 for 20s, blowdry, then immediately start e-beam procedure

9.2.1.1.3 Post-Liftoff Cleaning

- Metal should come off w/ACE like photoresist
- If not, try rubbing gently w/big Q-tips
- use squeezie bulbs to repeatedly squirt air bubbles and ACE at the sample while sample is immersed in ACE to remove stubborn metal
- DO NOT allow sample to dry with any metal still on it that you are uncomfortable with
- then rinse ACE 5 min
- ISO 5 min
- DI 3 min
- Bake out moisture 10 min

9.2.1.2 Photolithography

9.2.1.2.1 Standard Photolithography

- Fill 5 mL syringe with photoresist (PR), attach 40 µm filter
- Syringe will be hard to refill, use new each time
- Squrit evenly onto sample, make sure no air bubbles are left on sample
- Drag bead of PR around sample to cover edges
- Throw out PR after 6 months
- Clean vacuum chuck off w/big swabs and ACE
- Check vacuum
 - o if clogged, remove chuck, try to clear underside
- Use 4110 PR
- Spin on PR for 30s at 5500 RPM
- Soft bake 96 degrees for 1 min
- Aligning
 - Adjust fine vertical control such that mask makes contact just

before the contact switch maxes out

- Make sure deep UV filter is *not* in (used only for small features)
- Expose for 25s
- Post-exposure bake for 1 min, 95 degrees
- If liftoff is imminent, soak in toluene for 10 min before developing
- Develop in diluted (1:4) AZ 400K developer for 45s
- 9.2.1.2.2 Double-Layer Photolith
 - Spin OGC825 at 4000 RPM for 1 min
- Bake 95 degrees for 1min
- Flood expose 4-10 s (longer, more undercut, but less resolution/contrast)
- Continue w/ standard Photolith as usual

9.2.1.2.3 Image reversal

- Mark Su
 - o 5214 PR, spin 3000 RPM, 30s
 - Softbake 1 min
 - o Expose w/mask 15s
 - to reverse image,
 - hardbake 1 min
 - flood expose 30 s
- Maura Raburn (small features)
 - o expose mask 20 s
 - o hardbake 105 degrees, 1 min
 - o flood w/blue (deep UV) filter, 1 min
- Develop 1:5.5 AZ400K

9.2.1.3 Etching

- 9.2.1.3.1 Etchants
 - Rate estimates for GaAs, faster for AlGaAs
 - NH₄OH : H₂O::1:10, removes native oxides in 15s

- H₂SO₄:H₂O₂:H₂O::1:8:55, 15 Å/s
- H₃PO₄: H₂O₂:H₂O::3:3:100, 30 Å/s
- NH₄OH: H₂O₂:H₂O::10.5:1.8:150, 100 Å/s
- HCL:H₂O::1:1, removes Indium from substrate, 3hours to overnight etch.
 Remember to mask entire epilayer before attempting!
- NH₄OH: H₂O₂:H₂O::5.8:1:83, 75 Å/s

9.2.1.3.2 Etching procedure

- Use dedicated glassware.
- Mix up main etch (see above) in etch beaker
- Place on stirplate at 80 RPM for 30 min
- Mix up oxide etchant (#1 above)
- Dip in oxide etchant 20 s
- Rinse in DI tap 15 s
- Dunk in stirring main etchant for desired etch time
- Remove, rinse in DI for 15 s
- Dunk in ACE to strip away PR
- Dek-Tak to find actual etch depth, adjust main etch time appropriately
- Always strip off PR and check the etch depth for each sample. This may require quite a few etch steps, especially if the etch rate is off. This is a price well-worth paying for etch accuracy.

9.2.1.3.3 Etches in action

The actual etches used for the MISFET structures were taken from Jon Williams of the Sherwin Group, and so are detailed in his thesis. The ones used for NIN structures were similarly taken from Winston Schoenfeld of the Petroff Group, and so his thesis should be similarly consulted.

9.2.1.4 Metal depositions

- For use after 20s oxide dip
- For Ohmic contacts
 - o Jon Williams' recipe
 - 100 Å Ge
 - 100 Å Au
 - X Å Ge
 - Y Å Au
 - 100 Å Ni
 - 3000 Å Au
 - Choose X, Y, such that (100 + Y)/(100 + X) = 1.977
 - Typically used X, Y = 77, 250, respectively
 - Can use 100 Å Ni precursor to JW recipe to prevent spiking
 - Yuvaraj Dora's recipe
 - 70 Å Ni
 - 400 Å Ge
 - 600 Å Au

- 100 Å Ni
- 2000 Å Au
- For Schottky contacts
 - o Just dump Al on epilayer surface
 - Make *sure* surface is clean, as contaminants can ruin barrier quality
 - Used either e-beam 3 or thermal evaporator

9.2.1.5 Annealing

- Used only for Ohmic contacts
- Using Rapid Thermal Annealer
- 60s at 0 degrees C
- 15s ramp to 430
- 45s at 430, varied time and plateau temperature in various runs
- 1s ramp down to 0
- 120s at 0
- N₂ gas flow at 3000 sccm

9.2.2 MISFET trials

In the processing of the MISFET structures, we followed closely the procedures developed by Jon Williams, so his thesis should be consulted for details.

Specifically, the Ohmic contacts and waveguide structures were all the same as his.

9.2.3 MISFET-DX trials

Several techniques were attempted to separately contact the various, closelyspaced layers in this structure. First, simply calibrating the annealing temperature, time was tried. Second, we tried various metal recipes for shallow Ohmic contacts, as well as annealing times and temperatures. Neither case provided repeatable results, possibly due to surface states that get involved in shallow Ohmic contacts. Passivation of the surface was not attempted. Third, we tried isolating etches to judiciously cut off electrical paths of the various layers, one by one. The problem here was that, as an etched surface approaches a conduction path, surface states start to perturb the conduction long before a sharp cut-off can be observed. Ohmic contacts via shallow anneal into a side was not attempted.

9.2.4 NIN trials

Generally, the Ohmic contacts to the back gate always worked as described. The front gate doping layer needed to extend right to the sample surface for our Ohmic contacts to work there, though.

9.3 FTIR-FIR Measurements

For the MISFET and MISFET-DX structures, we performed FIR transmission measurements using the Bruker IFS 66V FTIR. In general, we used the 23 μ m beamsplitter and the Hg arc-lamp source. The sample and the 1.6K Si bolometer were located in the same cryostat, in the same geometry as that used in Jon Williams'

experiments on transmission of quantum wells, so reference should be made to his thesis for details.

9.4 Loading and Tuning

9.4.1 MISFET L&T

9.4.1.1 CV

Capacitance-Voltage measurements on the MISFET structures were performed exactly as similar measurements done by Jon Williams. In short, a DC voltage and an AC tickle voltage were both supplied by an SR830 lock-in amplifier. The voltages were scaled, added and applied to the sample front gate, in reference to the back gate, which was kept at virtual ground. Then, the current driven through the circuit was measured via a transimpedance amplifier and sent back into the SR830. The SR830 was accessed via GPIB for all measurements. All of the electronics between the SR830 and the sample (voltage adder, transimpedance amplifier, etc.) were contained in a 'CV box', which also allowed comparison to some standard capacitances for reference (~1 nF, .1 nF, etc.).

9.4.2 MISFET-DX L&T

CV was not attempted as a tool to measure charge loading in the MISFET-DX samples. This was because, at cold temperatures where CV gives a reasonable measurement due to high shunt resistances, no charge should move within the sample as the DX centers would be frozen out.

Instead, we tried to measure charge loading via FTIR absorption. Without the ability to change the charge density while cold, we could not use the empty dot state as a reference spectra. So, we cooled the sample down under bias conditions that we were fairly confident would load the dots. Then we took spectra at various bias voltages between the front-gate and back-gate, and divided these spectra by each other, expecting to see some signature of a shift of resonance frequency. We were able to see an FIR absorption, but it did not tune with bias. Instead, it disappeared and reappeared as a function of voltage, which we did not expect. In fact, the disappearance of the absorption was rather a signal that charge was moving in and out of the active region as a function of bias which our sample was designed explicitly *not* to do.

9.4.3 NIN L&T

9.4.3.1 LED

Our first successful loading measurement used a purple scrap IR LED. Strangely enough, these same LEDs were used in prior experiments with DX centers in the Sherwin Group. We mounted the LED pointing perpindicular to the sample surface, and applied voltages to the LED to turn it on. However, the turn-on voltage of the LED was very large when the LED was cold, so this required using a high-voltage (50V) power supply. But then, when the LED was turned on, the effective temperature must have increased, because the current through the LED would spike up and max out (~1A), limited by the power supply. This meant that each LED

would burn out relatively quickly, because no square-pulse voltage could be picked that would both turn the LED on quickly and not blow it up at cold temperatures.

9.4.3.2 Laser Diode

Our first successful attempt at L&T with the laser diode involved simply coupling the LD into a fiber coupler, and having the light spill out of the stripped fiber inside the cryostat, with the fiber tip laying on the epilayer, pointing tangentially toward it. Later, we wanted to use the same setup to do a crude PL measurement, but the collection efficiency of the fiber oriented parallel to the surface wasn't sufficient.

Our second successful attempt involved two more optics: a right-angle glass prism and a tuned 1064nm mirror. As the stripped fiber approached the sample, we bonded a 2mmx2mm glass prism on the end of the fiber. This allowed the light to reflect internally in the prism, and be redirected at 90 degrees from parallel to the epilayer to a perpendicular orientation. This also allowed light to be collected back through the fiber and out the fiber coupler, to measure PL. To separate the incoming LD optical path from the outgoing PL path, we inserted a dielectric mirror. The incoming laser then passed through the rough, uncoated back surface of the mirror and out the dielectric surface into the fiber coupler. The returning PL then hit the dielectric surface, and was then reflected into our spectrometer/PMT setup.

We soon found that there was significant low-energy luminescence coming from the LD, so we short-pass filtered it before it entered the fiber coupler. To eliminate

the LD light from coming into the spectrometer, we long-pass filtered the luminescence.

9.4.3.3 Electronics

9.4.3.3.1 Indium contacts

Many different types of contacts were tried to make electrical connections between gold wires and the gold surface of the Ohmic contacts. Silver paint works on occasion, but usually doesn't create a stable bond to the sample surface. We used a blob of Indium, and smushed our gold wire into this blob, which often stayed connected to the sample surface. The gold wires could then be threaded into pin sockets, and then the sockets could be filled with silver paint, to ensure socket-wire contact.

9.4.3.3.2 Current measurements, load resistor

For our initial measurements, we applied a square pulse from a pulse generator to the front gate in reference to the back gate. To measure the current flowing through this circuit (especially during the light pulses), we inserted a load resistor (100 kOhms) and measured the voltage across the resistor. This method was trivial to treat, as we could simply divide the voltage by the resistance to get the current. However, the load resistor did slow down the circuit, and because the resistor was at room temperature, it also added considerable noise.

9.4.3.3.3 Current measurements, transimpedance amplifier

For our subsequent experiments, we replaced the resistor with a transimpedance amplifier. In that way, we set our back gate to virtual ground, and measured the voltage on the output side of a feedback resistor. Had the feedback resistor been the only feedback element, the calculation would have gone as before. However, we also put a feedback capacitor in parallel with the feedback resistor, to prevent oscillations of the amplifier op amp. This complicates the voltage to current conversion a bit. Now, the voltage must be divided by the impedance of the feedback element, rather than the resistance alone. Since the impedance is a function of frequency, and complex, different frequency components translate into different scales of current for a given voltage. So, a fourier transform is necessary, but the result is trivial, and will be left as an exercise to the reader. Suffice it to say that the integrated-voltage-tocharge conversion must then include a term that is the multiplication of the feedback capacitor and the total change in voltage. That is, if you start integrating the voltage signal and stop integrating the voltage signal with identical endpoint voltages, the feedback capacitor contributes nothing to the total charge measurement.

9.4.3.3.4 Summary

To be clear, first one measures the current flowing through the circuit during the unloading pulse (NIR2). Then one integrates that current to find a charge that has flowed off of the sample. Then one subtracts that integration from an identical measurement with no loading pulse (NIR1). Then one converts that external charge to the equivalent internal charge to estimate the charge per quantum dot.

9.5 FTIR-MIR and Loading and Tuning

9.5.1 Optics

To perform MIR experiments, we first mounted the sample in an optical cryostat with a ZnSe window. We then eliminated the 1064 nm mirror and the fiber setup, and so had a much higher throughput of light onto the sample (~10 mW compared to ~1 mW). When the stray lights from the room were blocked, the loading/unloading procedure worked as before. To couple the MIR onto the sample, the cryostat was mounted into the Bruker, and the MIR spot was aligned onto the sample using the IR viewer. In this way, both the NIR and the MIR could be aligned onto the same spot. The HeNe alignment laser had to be blocked, since its wavelength would have effectively shorted the sample, so we used a room-temperature InAs wafer to absorb the HeNe, and all optical energies of the MIR source above ~350 meV. For the MIR lifetime measurements shown, the optics were arranged in just this way.

For the step-scan MIR measurements, we allowed some high-energy MIR light to hit the sample, to allow us to confirm some signal at all.

9.5.2 Electronics

For the MIR lifetime measurements, no change in the electronics was necessary.

For the step-scan measurements, things were a bit more complicated. We eliminated the NIR2 unloading pulse, in favor of looking at the MIR unloading our sample. We also chopped the MIR using a mechanical chopper located right between the source and the aperture in the FTIR. We then locked-in to the component of the loading/unloading voltage signal that oscillated at just the frequency of the chopper (~200Hz). When observing the in-phase component of the signal through the external input, the Bruker could then take step-scan spectra.

Care should be taken to ensure that all appropriate connections are made among the logic boards of the Bruker, and that all preparatory steps have been completed according to the Bruker step-scan manual.

It should be noted that, in order to do step-scan, one needs to extract the signal from a detector and send it back into the Bruker external input port. If one wishes to do step-scan with one of the internal Bruker detectors, it is a bit challenging. This is because, when the Bruker is directed to analyze the signal from the external detector port, the mirrors are also moved to pass the beam out to the external beam port side. This has the effect of blocking the beam from any internal detector, much to the user's frustration. No solution to this problem has been found to date, though Bruker, Inc. has not been queried.

9.6 DX centers

Well, we know of no reason, in principle, why such a structure could not work. However, our experience is that it simply did not. Our first challenge was one of growth as our first attempt was designed incorrectly. Our second challenge was one of processing. We wanted to separately contact two buried layers (BG and DX) with ohmic contacts, while the electrostatics of the structure demanded that they be grown relatively closely together. After approximately a year (!) of processing, we

reverted to a simpler biasing scheme that allowed joint contact of the BG and DX layers. Finally, we were able to see an absorption signal, but it did not tune with gate voltage, and so could have been CWL absorption. Further, we grew an identical sample with coupled quantum wells (CQWs) instead of coupled quantum dots, to observe the analogous filling of the wells via PL broadening, but found no such broadening; the well-width we chose was too narrow, and the PL therefore too broad, to see any noticeable broadening.

By this time, several years (!) into it, we were quite tired and frustrated with this approach.