AN EFFICIENT SOURCE OF SINGLE PHOTONS:
A SINGLE QUANTUM DOT IN A MICROPOST MICROCAVITY

A DISSERTATION
SUBMITTED TO THE DEPARTMENT OF APPLIED PHYSICS
AND THE COMMITTEE ON GRADUATE STUDIES
OF STANFORD UNIVERSITY
IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY

Matthew Pelton
January 2002
I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Prof. Yoshihisa Yamamoto  
(Principal Adviser)

I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Prof. David A. B. Miller

I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Prof. James Harris

I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

Dr. Glenn S. Solomon

Approved for the University Committee on Graduate Studies:
Abstract

A quantum dot (QD) is a nanometer-scale inclusion of one semiconductor inside a second semiconductor with a larger bandgap. Electrons and holes in the QD can occupy only a given set of states with discrete energies, as in an atom. QD’s can thus be used to do “atomic physics” experiments in the solid state.

For example, the spontaneous emission rate from a QD can be enhanced by placing the dot inside a resonant optical cavity. We grew InAs / GaAs QD’s by strain-driven self assembly in molecular beam epitaxy. Above and below the dots, we deposited reflectors consisting of alternating quarter-wavelength thick layers of AlAs and GaAs. The sample was then plasma etched into microscopic posts. Light in the posts is trapped laterally by total internal reflection and longitudinally by the reflectors. We have seen significant emission-rate enhancement for a single QD inside such a micropost microcavity.

A single QD can also serve as a source of single photons “on demand.” Electrons and holes are introduced into the dot by pumping with a pulsed laser. For each pump pulse, the last photon emitted from the dot has a frequency different from that of all the other photons. This photon can thus be isolated by spectral filtering. When the QD is coupled to a micropost microcavity, the majority of these photons are captured by the cavity mode. We have used this technique to demonstrate an efficient source of single photons. This device has potential applications to quantum cryptography, quantum computation, and other areas.
This thesis is dedicated to my parents.

“A wise son maketh a glad father.” Proverbs x. 1.
I have been fortunate to spend my time at Stanford in the research group of Prof. Yoshihisa Yamamoto. Prof. Yamamoto has taught me not only a lot of physics, but also a way of doing research and a way of thinking, which are the most important things I have learned at Stanford. He has also gathered together an exceptionally bright and talented group of students and researchers who have helped me in countless ways and who have made the last five years lots of fun. I would like to thank all the current and former Yamamoto Group members that I have interacted with, but I mention by name only those who contributed most directly to the work reported in this thesis: Charles Santori, Bingyang Zhang, Jelena Vučković, Jocelyn Plant, Oliver Benson, Hui Deng, and Edo Waks. I would like to single out Glenn Solomon, who worked with me probably more than anybody else, and who has put up with a lot from me.

Without the technical and administrative help of several staff members, I would never have been able to do anything. Especially crucial were Tom Carver, Len Booth, Yurika Peterman, and Paula Perron. Many people outside the University also provided critical support, including David Dechaume and Bart Stevens of Riber and Bill Neill of LEO. Several members of the Harris Group were very helpful in the MBE lab, as well.

I would like to acknowledge financial support from the Stanford Graduate Fellowships program.

I would also like to acknowledge valuable discussions and advice from Prof. James Harris, Prof. David Miller, Prof. Stephen Harris, and Prof. Axel Scherer.

Finally, I would like to thank Éva Nagypál, who makes it all worthwhile.
# Contents

Abstract iv

Acknowledgements vi

1 Introduction 1

1.1 Why Single Photons? 1

1.1.1 Basics of Quantum Cryptography 1

1.1.2 The Second-Order Correlation Function 5

1.2 Sources of Single Photons 8

1.3 Outline 12

2 Self-Assembled Quantum Dots 13

2.1 Quantum Dots 13

2.2 Self-Assembly by Molecular-Beam Epitaxy 16

2.2.1 Basics of Molecular-Beam Epitaxy 16

2.2.2 Stranski-Krastanov Growth 18

2.2.3 Indium Arsenide / Gallium Arsenide Quantum Dots 20

2.3 Quantum-Dot Spectroscopy 24

2.3.1 Optical Transitions in Self-Assembled Quantum Dots 24

2.3.2 Light from Single Quantum Dots 28

3 Micropost Microcavities 35

3.1 Basics of Cavity Quantum Electrodynamics 35

3.1.1 Theory 35
3.1.2 Atomic Cavity QED ........................................... 39
3.1.3 Semiconductor Cavity QED ............................... 39
3.2 Single-Mode Microcavities ................................. 41
  3.2.1 Three-Dimensional Semiconductor Microcavities .... 41
  3.2.2 Fabrication of Micropost Microcavities .............. 43
3.3 Modes in Micropost Microcavities ......................... 53
  3.3.1 Finite-Difference Time-Domain Model .................. 55
  3.3.2 Approximate Model ....................................... 60

4 Single-Dot Lifetime Modification 65
  4.1 Quantum-Dot Cavity QED ................................... 65
  4.2 Coupling Single Dots to Single Cavity Modes .......... 66
    4.2.1 First-Generation Posts ............................... 66
    4.2.2 Emission-Rate Enhancement .......................... 68
  4.3 Second-Generation Posts ................................ 74

5 Triggered Generation of Single Photons 79
  5.1 Experimental Method ...................................... 79
  5.2 Efficient Generation of Single Photons .................. 82

6 Future Directions 92
  6.1 Device Development ........................................ 92
  6.2 Micropost Microcavity Optimization .................... 96
    6.2.1 Maximum Quality Factor .............................. 96
    6.2.2 Strong Coupling ...................................... 96
    6.2.3 Lasing ................................................ 97
  6.3 Microsphere Single-Dot Laser ............................ 99
    6.3.1 Proposed Experiment .................................. 99
    6.3.2 Laser Threshold ...................................... 102
  6.4 Entangled Photon-Pair Generation ....................... 105

7 Conclusions 110
List of Tables

3.1 Quality factor $Q$ and resonance wavelength $\lambda$ for the fundamental modes of micropost microcavities, as calculated by the finite-difference time-domain method. .................................................. 59

A.1 Stages in molecular-beam epitaxy of a microcavity containing quantum dots ................................................................. 112

A.2 Stages in the process used to make micropost microcavities ....... 113
List of Figures

1.1 Optimized secure communication rate for a realistic quantum-key-distribution system employing the BB84 protocol .......................... 9

2.1 Particle in a box ................................................................. 14
2.2 Semiconductor heterostructures and corresponding densities of states 15
2.3 Schematic of a molecular-beam epitaxy system (from [1]) .......... 17
2.4 Modes of epitaxial crystal growth: (a) Frank-van der Merve, (b) Volmer-Weber, (c) Stranski-Krastanov ................................. 19
2.5 Atomic-force-microscope images of quantum dots grown at different substrate temperatures: (a) 480 °C, (b) 487 °C, (c) 498 °C, (d) 520 °C 21
2.6 Atomic-force-microscope image of quantum dots grown under conditions optimized for low density ........................................ 23
2.7 Schematic of transitions in an InAs / GaAs quantum dot .......... 25
2.8 Schematic of the apparatus used for photoluminescence measurement 26
2.9 Photoluminescence spectrum from an ensemble of InAs / GaAs self-assembled quantum dots ................................................. 27
2.10 Photoluminescence spectrum from a sparse ensemble of InGaAs / GaAs self-assembled quantum dots ................................. 28
2.11 Photoluminescence spectra from a single InAs / GaAs self-assembled quantum dot, under above-band excitation (a) and resonant excitation (b) ................................................................. 30
2.12 Integrated luminescence intensity of the single-exciton line (circles) and biexciton line (diamonds) of a single quantum dot, as a function of above-band pump power .................................................. 31
4.3 Quality factors of the fundamental modes of micropost microcavities, as measured experimentally (points) and as calculated by the approximate method (line), including additional reflection loss in the distributed Bragg mirrors .............................................................. 69

4.4 Time-dependent luminescence intensity from quantum dots in a micropost microcavity, as measured experimentally (points) and as fitted using a rate-equation model (line) ............................................ 69

4.5 Photoluminescence lifetime for quantum dots in a micropost microcavity with a top diameter of 2 µm, as measured experimentally (points) and as predicted using the approximate model (dashed line); also shown is the photoluminescence intensity (solid line) ..................... 70

4.6 Photoluminescence lifetime for quantum dots in a micropost microcavity with a top diameter of 0.5 µm, as measured experimentally (points) and predicted using the approximate model (dashed line); also shown is the photoluminescence intensity (solid line) ..................... 72

4.7 Coupling coefficient of spontaneous emission from single quantum dots into the fundamental mode of micropost microcavities, as measured experimentally (points), and as predicted by the approximate model for dots in the center of the post (dashed line) or averaged radially over the post (solid line); the inset shows the corresponding spontaneous-emission lifetimes ............................................................. 74

4.8 Quality factors measured for first-generation and second-generation micropost microcavities ................................................................. 75

4.9 Photoluminescence from quantum dots in a second-generation micropost microcavity ............................................................. 76

4.10 Measured emission intensity from two quantum dots in a micropost microcavity as a function of pump power (points), and linear fits (lines) 77

4.11 Measured lifetimes of two quantum dots as a function of temperature-dependent detuning from a micropost microcavity mode (squares for one dot, circles for the other), and Lorentzian fits (lines) .............. 78
5.1 Schematic of the apparatus used for generation and detection of triggered single photons ........................................ 80

5.2 Photon autocorrelation for emission from a single quantum dot under pulsed, resonant excitation, with normalized peak areas indicated ........................................ 83

5.3 Photoluminescence from a micropost microcavity containing a single quantum dot ........................................ 84

5.4 Schematic of the apparatus used to measure single photons from a quantum dot in a micropost microcavity ........................................ 84

5.5 Measurement of the linear polarization of a single quantum dot in a micropost microcavity ........................................ 85

5.6 Measured autocorrelation for photons from a single quantum dot in a micropost microcavity with an incident pump power of 10.9 \( \mu \text{W} \) (points), and corresponding fit (line) ........................................ 86

5.7 Normalized area of the central autocorrelation peak for photons from a quantum dot in a micropost microcavity as a function of pump power (points), as well as a linear fit (line) ........................................ 87

5.8 Measured efficiency of the single-photon source as a function of pump power (points), together with a saturation fit (line) ........................................ 90

6.1 Schematic of the proposed structure to position a microsphere above a single quantum dot ........................................ 100

6.2 Schematic of the proposed apparatus to measure lasing with a single quantum dot coupled to the whispering-gallery mode of a microsphere cavity ........................................ 101

6.3 Calculated number of photons spontaneously emitted from a single InAs quantum dot and stored in the whispering-gallery mode of a microsphere, as a function of the microsphere-sample distance, for different sphere radii \( R \) ........................................ 105

6.4 Selection rules for conduction-band to valence-band transitions in a III/V semiconductor quantum well ........................................ 106
6.5 Measured two-photon polarization density matrix for single-exciton
and biexciton photons ........................................... 108

A.1 Optical-microscope image of a portion of the etched microcavity sample114
Chapter 1

Introduction

1.1 Why Single Photons?

1.1.1 Basics of Quantum Cryptography

Since the time of the ancient Romans, people have communicated secretly by means of ciphers. Letters in their messages were substituted with different letters, with the intention of making the message intelligible only to the intended recipient, who knew the substitution scheme. Attempts to develop increasingly sophisticated ciphers and the competing attempts to break the ciphers and read secret messages have led to advances in technology, computer science, mathematics, and physics [3]. Such advances have often resulted in the cracking of cipher systems previously thought unbreakable. “Unbreakable,” in these cases, meant that the computational resources needed to decipher a message were considered to be out of the reach of any human agency. The famous and widely-used RSA system, for example, is based on the computational difficulty of factoring the product of two large prime numbers [4]. The computation time for all known factorization algorithms increases exponentially with the size of the number of be factored. However, there is always the possibility, however remote, that an efficient factoring algorithm that works on conventional computers will be discovered. If a quantum computer is ever developed, it will be able to factor in polynomial time [5]. More prosaically, available computational power continues
to increase exponentially, meaning that previously intractable problems eventually become manageable.

The Holy Grail of cryptography is a cipher which is not just extremely difficult, but impossible, to break. However, it was shown by Shannon, the father of information theory, that the only cipher with absolute security is the one-time pad, or Vernam cipher \[6\]. The one-time pad can be implemented, for example, on the digital representation of a message by the Boolean addition of each bit to be sent with a randomly-chosen key bit. The difficulty with this system is that the key is as long as the message itself. The technical challenge becomes one of key distribution: the intended recipient (Bob) must share with the sender (Alice) the string of key bits in order to decipher the message, and the key must be kept out of the hands of any potential eavesdropper (Eve). This has prevented the widespread use of one-time pads.

However, quantum mechanics offers a way around the key-distribution problem. One of the best-known principles of quantum mechanics states that it is impossible to measure the unknown state of a single quantum system without running a risk of changing the state of the system. If the key can be encoded on the quantum states of single particles, then Bob can, in principle, detect Eve’s presence through this measurement-imposed disturbance. This idea is the foundation of the growing field generally known as “quantum cryptography,” but perhaps better called quantum key distribution \[7\].

There are several schemes for real-world quantum key distribution, but the best studied from both a theoretical and experimental point of view is known as BB84, because it was invented by Bennett and Brassard in 1984 \[8\]. It involves encoding on four non-orthogonal states of a quantum system. For example, Alice and Bob can use four linear polarization states of single photons: horizontal polarization \(|H\rangle\), vertical polarization \(|V\rangle\), polarization 45° counterclockwise from the horizontal \(|+45\rangle\), and polarization 45° clockwise from the horizontal \(|-45\rangle\). The polarizations can be grouped into two bases: \(\{ |H\rangle, |V\rangle \} \) and \(\{ |+45\rangle, |-45\rangle \}\). Each of these bases can be used to encode a bit; for example, \(|H\rangle\) could correspond to the value 0, while \(|V\rangle\) would correspond to the value 1. For each photon that she sends, Alice randomly
chooses a basis and a bit value, and sends the resulting linearly-polarized photon to
Bob. When Bob receives the photon, he randomly chooses one of the two bases in
which to measure the photon polarization. Once the entire stream of photons has
been detected by Bob, he and Alice each have an \textit{unsifted key} of sent or received bits.
However, their bits are only guaranteed to be the same when Bob used the same basis
for detection as Alice used for transmission, and when there are no errors.

The next step, then, is for Alice and Bob to communicate over a classical channel
and tell each other which bases were used in the transmission and detection. This
channel can be insecure (\textit{i.e.}, the information about the bases can be broadcast pub-
ically), but it is assumed to be \textit{unjammable}; that is, Eve is not capable of mounting
a \textit{man-in-the-middle} attack, where she breaks the classical transmission and imper-
sonates one of the communicating parties to the other. (The problem of unjammable
communication can be dealt with by classical authentication protocols [9].) Alice and
Bob retain only the bits for which they used the same basis, ending up with a \textit{sifted
key} which is, on average, half the length of the unsifted key.

Alice and Bob can then check for the presence of eavesdropping by checking for
errors in the sifted key. This is done by comparing a randomly selected subset of
the sifted key over a public channel, and subsequently discarding this subset. Any
measurement that Eve makes on the quantum transmission will impose an inevitable
\textit{back action} on the photons, which will show up as errors in the sifted key. However,
in any actual implementation of quantum key distribution, errors will also result from
the imperfect apparatus used. For example, single-photon detectors always produce
\textit{dark counts}, or false measurements of photons. Quantum key-distribution protocols
deal with these and other errors through \textit{error correction} and \textit{privacy amplification}.

Error correction allows Alice and Bob to share an identical string. This is done
by communicating over the public channel, while disclosing as little information as
possible about the corrected key. One practical scheme involves dividing the sifted
key into blocks, and comparing the \textit{parity} of those blocks (\textit{i.e.}, the sum, modulo two,
of the bits in the blocks) [10]. If the parities disagree, there must be an odd number
of errors in the block; this block is searched recursively by dividing into two sub-
blocks and comparing their parities. After the bits in error have been isolated and
corrected, Alice and Bob randomly permute the key, and repeat the procedure with larger blocks. After a sufficient number of repetitions, Alice and Bob share, with high probability, an identical key, about which Eve has learned relatively little.

At this point, Alice and Bob know the error rate, which they can assume was entirely due to Eve. Based on this and on the information leaked during the error correction step, they can determine the maximum amount of information Eve can have about the reconciled key. They can then shorten the key by the corresponding amount, so that the information Eve has is reduced to an arbitrarily small value; this is the process of privacy amplification. Provided that the initial error rate is sufficiently small, Alice and Bob can always distill a secret reconciled key, even if Eve is allowed to perform any physical operation on the transmitted photon stream [11, 12].

The BB84 protocol was first demonstrated experimentally in 1992 by its inventors [13]. Since then, it has been implemented in optical-fiber communication networks [14], with transmission distances up to 48 km [15]. Secret keys have also been transmitted through free space in daylight up to 1.6 km [16], suggesting the feasibility of earth-to-satellite quantum key distribution. However, all these demonstrations have a shortcoming: they do not use true single-photon sources.

Key bits are encoded on the polarization states of single photons; the security of the transmission arises from the fact that a single photon is indivisible and its unknown quantum state cannot be copied [17]. However, single photon sources have not been generally available. Researchers have had to resort to approximations of single photon streams, generally pulses from lasers or light-emitting diodes, attenuated to the point where the average photon number per pulse is significantly less than one. The number of photons in these pulses is described by Poissonian statistics, so that there is always a possibility of more than one photon being sent in a given pulse. Such pulses are vulnerable to a photon-number splitting attack [18, 19]. It is possible for Eve to measure the photon number in each pulse without absorbing any photons. She can then split off one photon from the pulses that originally contain more than one photon, and retain this photon until Alice and Bob discuss their bases. At this
1.1. WHY SINGLE PHOTONS?

point, she can correctly measure the photon polarization and obtain the correspond-

ing bit value without creating any error on Bob’s side. Alice and Bob can compensate

for this additional information leakage through increased privacy amplification, given

the a priori probability of emitting multiple-photon pulses. However, this reduces the

error that they can tolerate from other sources, and also reduces the length of the

final distilled key. Implementation of quantum key distribution would be aided by a

source that emitted only one photon at a time.

1.1.2 The Second-Order Correlation Function

The ideal light source for a quantum key-distribution system with a raw transmission

rate $R$ would emit one and only one photon in each time interval $T = 1/R$. Any

real light source, though, will have a non-zero probability of emitting more than one

photon in a given time interval. A smaller probability of multi-photon pulses means

less privacy amplification, and thus a higher rate of secure communication. Our goal

is to produce a device that has a reduced probability of emitting multiple-photon

pulses as compared to a Poissonian source, which we will call a single-photon source.

In order to quantify how good our single-photon source is, we use the normalized

second-order correlation function $g^{(2)}(\tau)$. Classically, this is defined as the autocorre-

lation of the optical intensity:

$$g^{(2)}(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2} = \frac{\langle E^*(t)E^*(t+\tau)E(t+\tau)E(t) \rangle}{\langle E^*(t)E(t) \rangle^2}, \quad (1.1)$$

where $I(t)$ is the intensity at time $t$, $E(t)$ and $E^*(t)$ are the corresponding electric

field and its complex conjugate, and $\langle \ldots \rangle$ indicates averaging over $t$ (equivalent to

a statistical average for ergodic systems). Simple algebraic inequalities impose two

general conditions on the classical correlation function [20]:

$$g^{(2)}(0) \geq 1, \quad (1.2)$$

$$g^{(2)}(\tau) \leq g^{(2)}(0). \quad (1.3)$$

A classical beam of constant intensity, for example, will have a constant correlation
CHAPTER 1. INTRODUCTION

function $g^{(2)}(\tau) = 1$. A pulsed source of coherent light will have a correlation function consisting of a series of identical peaks separated by the repetition period $T$; when normalized by $T$, the area of each peak is equal to unity.

Quantum mechanically, the electromagnetic field is described using operators rather than algebraic quantities. For a single-mode light field, the correlation function can then be written as

$$g^{(2)}(\tau) = \frac{\langle \hat{a}^\dagger(t)\hat{a}^\dagger(t + \tau)\hat{a}(t + \tau)\hat{a}(t) \rangle}{\langle \hat{a}^\dagger(t)\hat{a}(t) \rangle^2}, \quad (1.4)$$

where $\hat{a}^\dagger$ and $\hat{a}$ are the photon creation and annihilation operators, respectively. $g^{(2)}(\tau)$ thus represents the probability of detecting a second photon a given time $\tau$ after a first one is observed. The correlation function of a coherent source reflects its Poissonian photon statistics: the probability of detecting a photon is independent of whether one has already been detected.

In the quantum-mechanical formalism, the inequalities of Eqns. (1.2) and (1.3) no longer need be obeyed; the only general condition on $g^{(2)}(\tau)$ is that it cannot be negative. In fact, it is possible to generate light where both inequalities are violated; i.e., where the probability of detecting a second photon is lowest immediately after detecting a first photon. Such light is said to be antibunched, because of the more regular spacing of the photons. Observation of antibunching, in fact, provided the first unarguable evidence of the quantum nature of light [21].

An ideal source of single photons would have the same correlation function as that of pulsed coherent light, except that the peak centered at $\tau = 0$ would be absent. This would mean that, once a photon has been detected in a given pulse, another one will never be detected in the same pulse. A non-ideal single-photon source has a zero-delay peak with an area between zero and one. This area, which we label $g^{(2)}_o$, gives an upper bound on the probability $P(n \geq 2)$ that two or more photons are present in the same pulse:

$$P(n \geq 2) \leq \frac{1}{2} \langle \hat{n} \rangle^2 g^{(2)}_o, \quad (1.5)$$
1.1. WHY SINGLE PHOTONS?

where $\langle \hat{n} \rangle$ is the mean photon number per pulse.

This area is sufficient to describe how a single-photon source will improve the performance of a quantum key-distribution system. For the BB84 protocol, it is possible to calculate the amount of secure information shared by Alice and Bob if we limit the capabilities of Eve to individual or incoherent attacks [19]. This means that Eve can intercept any photon, store it, and make a general quantum measurement on it after Alice and Bob have publicly declared their bases. The measurement involves entangling a quantum probe with each photon to be measured, and then making a destructive measurement of the state of the probe [22]. Eve can optimize the entanglement and the probe measurement, but she must measure the probes one at a time. This limitation is imposed because it makes the calculations for realistic systems tractable, but also because it corresponds to the limit of what is possible with current technology.

Fig. 1.1 shows results calculated for realistic system parameters. The vertical axis is the number of secure bits shared by Alice and Bob, normalized by the number of bits initially sent by Alice; the horizontal axis is the total optical loss, including inefficiencies of the source and detectors and absorption loss in the transmission channel. The single-photon detectors are assumed to have a dark-count probability of $5 \cdot 10^{-8}$ per pulse. On each graph, antibunched sources are compared to two limits: a source with Poissonian statistics, and an ideal source of single photons. The mean photon number per pulse for the Poissonian source is allowed to take on any value, and is optimized for each value of channel loss. Between the two limits are the realistic single-photon sources. These sources are assumed to emit a fixed mean photon number $\langle \hat{n} \rangle$ per pulse; different graphs correspond to different values of $\langle \hat{n} \rangle$ for the realistic sources. It is also assumed that a variable attenuator is placed after the source; the attenuation is optimized for each value of channel loss. Different lines in each graph correspond to sources with different degrees of antibunching (i.e., different values of $g_o^{(2)}$).

For low channel loss, the optimized secure transmission rate for a Poissonian source is approximately 0.2 bits per pulse. In this regime, then, the Poissonian source will outperform antibunched sources with $\langle \hat{n} \rangle < 0.2$. As the channel loss increases, though,
the secure transmission rate for the Poissonian source decreases rapidly, as more and more privacy amplification is required, and the antibunched sources eventually surpass the Poissonian source. The transmission rate for all sources then continues to drop as loss increases and the number of photons reaching Bob decreases. Eventually, the errors due to the multi-photon pulses and the dark counts of Bob’s detectors overwhelm any photons that are successfully transmitted from Alice to Bob, and the transmission rate falls off abruptly. At this point, the amount of information that Bob has after the transmission about Alice’s string is less than the amount of information available to Eve. The number of bits required for privacy amplification, in other words, is less than the number of bits available to Bob. The detector dark counts and multi-photon probability thus define a maximum tolerable loss for secure communication. This cutoff loss increases with the degree of antibunching, because of the reduced ability of Eve to perform photon-number splitting attacks. In other words, if a source can be made that has a reduced probability of emitting more than one photon per pulse, quantum key distribution can be performed over longer distances.

1.2 Sources of Single Photons

The first experimental step towards a single-photon source was the production of a continuous stream of antibunched photons. This was done in 1977 by Kimble et al. by measuring resonance fluorescence from sodium atoms [21]. In these experiments, a thermal beam of atoms was excited by a dye laser tuned to an effective two-level transition. Fluorescence was collected from the atoms at a right angle to both the laser and atomic beams. The atomic flux was reduced to the point where there was a small probability for more than one atom at a time to intercept the laser beam. After a single atom emitted a photon, some time had to pass while it was re-excited before it could emit again. The photon correlation function thus vanished for zero time delay. An improvement on this experiment involved holding a single ion inside a Paul trap and using laser cooling to reduce its motion before measuring the resonance fluorescence [23].
1.2. SOURCES OF SINGLE PHOTONS

Figure 1.1: Optimized secure communication rate for a realistic quantum-key-distribution system employing the BB84 protocol
These experiments, however, required complex setups, which are not practical for integration into larger systems. Single organic molecules can be handled more easily than single atoms, by incorporating the molecules as dilute impurities in a crystalline host [24]. Luminescence from single molecules can be isolated spatially, by collecting light only from a small volume of the sample, and spectrally, by filtering out the narrow zero-phonon line of one molecule out of the inhomogeneously broadened ensemble. Antibunching from the fluorescence of single organic molecules has been seen at low temperature [25] and at room temperature [26]. However, in these experiments, the molecules generally suffer from photobleaching; i.e., the molecule stops emitting light after a certain amount of active time.

One solution to this problem has been to use nitrogen-vacancy (NV) centers in synthetic diamond crystals as the optical emitters. These defects consist of a substitutional nitrogen atom with a vacancy at an adjacent lattice position, and can be created with a controlled density by electron irradiation followed by annealing in a nitrogen environment. Light is emitted from the NV centers with high quantum efficiency and without photobleaching [27]. Antibunching in the radiation from single NV centers has been seen at room temperature [28, 29]. However, these defects intermittently stop emitting light due to shelving in metastable levels. As well, the emitted light is broadband. Finally, although diamond is well studied, it is not a mainstream semiconductor material, so that integration of the emitters into devices is impractical.

All of the sources described above emit continuous streams of antibunched photons. At least conceptually, it is straightforward to change the experimental conditions in order to produce single-photon pulses. In all cases, antibunching is due to the time required for re-excitation after a photon has been emitted. If the probability of emitting a photon and being re-excited within the excitation period can be made vanishingly small, then only one photon will be emitted for each excitation pulse. This was first demonstrated with single molecules that were pumped into their excited states using the method of adiabatic following [30]. This was later simplified by pumping with short pulses from a mode-locked laser [31, 32].

A different approach to a single-photon source involved the controlled injection
of carriers into a mesoscopic quantum well [33]. The well was alternately populated with electrons and holes by resonant tunnelling. The well area was reduced by etching small posts into the device, and the sample was held at low temperature, so that the charging energy of the wells was larger than the thermal energy of the carriers in the tunnel reservoirs. The Coulomb blockade effect then prevented more than one carrier at a time from entering the well [34]. The current regulation translated into regulated photon emission. However, the required operation temperature was less than 1 K, making the device impractical for larger systems. As well, leakage current produced a significant background of unregulated photons, limiting the degree of antibunching attainable.

An alternative semiconductor structure for single-photon generation is the quantum dot. Quantum dots can be thought of as “artificial atoms,” and photon antibunching should be possible by using quantum dots in the equivalent of atomic resonance-fluorescence experiments. This phenomenon has in fact been observed using different types of quantum dots [35, 36]. Pulsed excitation of the dots can be used to produce a triggered stream of single photons, as for single molecules [37, 38, 39].

Quantum dots offer several advantages as a source for single photons. The quantum efficiency and oscillator strength of the dots are very high. The dots do not suffer from photobleaching or shelving, and are stable indefinitely. Unlike mesoscopic quantum wells, quantum dots allow the number of confined carriers to be controlled without resorting to the Coulomb blockade effect, allowing for higher-temperature operation. The materials used to make the dots are compatible with mature semiconductor technologies, allowing for the sources to be further developed and to be incorporated into larger structures.

This last advantage is significant, because it provides a route to solving the problem of low collection efficiency. Single atoms, for example, radiate isotropically, so that only a small fraction of the emitted photons will be captured by a lens placed nearby. For quantum dots, the situation is even worse, because the majority of the photons do not leave the high-refractive-index semiconductor sample. Improvement of the collection efficiency is necessary for practical application of the source.

The collection efficiency can be improved using the techniques of cavity quantum
electrodynamics (QED). The emission characteristics of a quantum dot can be modified by placing it inside a microscopic optical cavity, or microcavity. If the dot is resonant with a cavity mode, the spontaneous emission rate into that mode will be enhanced. This means that light from the dot will be emitted mostly into a single spatial mode, which can be efficiently coupled into downstream optical elements [40]. Combining this with pulsed excitation of the dot results in an efficient source of single photons. The coupling of a single quantum dot to a single mode of a microcavity, and the production of an efficient source of single photons, are the main topics of this thesis.

1.3 Outline

This thesis will detail what is necessary to fabricate an efficient source of single photons using a single quantum dot in a microcavity. Chapter 2 presents the quantum dots that we use in our experiments. Details are given on the formation of these dots through a self-assembly process, and the optical properties of the quantum dots are presented. Chapter 3 explains how the quantum dots are coupled to microcavities. The specific single-mode optical cavities that we use are described. Details are given on how the microcavities are fabricated, and a theoretical treatment of modes in the cavities is presented. In Chapter 4, coupling between single quantum dots and single modes in the microcavities is described. Experimental results are presented for the modification of the spontaneous emission rate of single quantum dots. Chapter 5 explains how single dots in micropost microcavities are used to generate single photons. Experimental demonstration of an efficient single-photon source is presented. Finally, Chapter 6 discusses future directions for this project. Potential device improvements are mentioned, as well as the possibility of optimizing microcavity design and observing new physical phenomena. Other potential devices are discussed, including a single-dot laser and a triggered source of entangled photon pairs.
Chapter 2

Self-Assembled Quantum Dots

2.1 Quantum Dots

One of the first problems that confronts students of elementary quantum mechanics is the “particle in a box”: a massive particle such as an electron in a potential that is zero within some region and finite outside of the region. As every student who manages to pass the course knows, the wavefunction of the electron can take on only certain standing-wave patterns, with a corresponding discrete set of energy levels (see Fig. 2.1). In other words, the energy of the electron is quantized by the potential well.

Remarkably, it is possible to fabricate semiconductor structures that are very well approximated by this potential-well model. These are semiconductor heterostructures, or composite crystals of more than one semiconductor material. The states of electrons and holes in these structures can generally be described using a modified version of Bloch’s theorem, where the particle wavefunction \( \psi(\vec{r}) \) is written as a product of a periodic part \( u(\vec{r}) \) and an envelope wavefunction \( \chi(\vec{r}) \) [41]:

\[
\psi(\vec{r}) = u(\vec{r})\chi(\vec{r}).
\]  

(2.1)

The Bloch function \( u(\vec{r}) \) repeats with the periodicity of the lattice. In GaAs and InAs, \( u(\vec{r}) \) is a \( s \)-like wavefunction for electrons in the conduction band, and is a
$n = 1$

$V = V_0$

$V = 0$

$n = 2$

$n = 1$

\[ \psi_n \]

\[ \psi_n \]

$V_0$

\[ (2.2) \]

\[ \frac{\hbar^2}{2m^*} \nabla^2 + V(\vec{r}) \psi(\vec{r}) = E \psi(\vec{r}), \]

where $E$ is the carrier energy. This equation of motion is equivalent to the ordinary Schrödinger equation, except that the particle mass is replaced by an effective mass $m^*$, and the potential $V(\vec{r})$ corresponds to the energy of the appropriate band edge. That is, $V$ is the bottom of the conduction band for electrons, and it is the top of the valence band for holes.

The potential profile of the “particle in a box” can thus be simulated by a variation in bandgap. A quantum well consists of a thin sheet of a semiconductor material with a lower bandgap sandwiched between layers of a semiconductor material with a wider bandgap. Carriers are trapped in the low-bandgap material by the effective potential well. If the thickness of the layer is small enough, carriers form standing waves, and can only occupy discrete energy levels. The “quasielectric fields” created by bandgap variations allow both electrons and holes to have potential minima in the same location, something that is not possible with real electric fields.
At low enough temperatures, virtually all the carriers in a quantum well (electrons and holes) will be in the lowest quantum-confined state. The carriers are free to move, however, in the plane of the well. The well is referred to as a quasi-two-dimensional system, since the carrier motion has effectively been restricted to a plane. The corresponding density of states shows a staircase pattern. The density of states can be made more singular if the effective number of dimensions is reduced to one, in a quantum wire, or zero, in a quantum dot (see Fig. 2.2). In a quantum dot, the energy levels are fully quantized, as in an atom. Optical transitions between the valence band and the conduction band can thus occur only at discrete energies.

Fabrication of high-quality quantum dots, however, has lagged far behind fabrication of quantum wells. Two well-developed epitaxy techniques allow for the deposition of semiconductor materials with atomic-layer precision and very high purity: metal-organic chemical-vapor deposition (MOCVD) and molecular-beam epitaxy (MBE). With these methods, it has been possible to grow quantum wells since the 1970’s. However, deposition techniques that go beyond planar layer-by-layer growth were not
developed until recently. Early attempts at making semiconductor quantum dots fo-
cussed on patterning quantum wells using nanoscale lithographic techniques, such as
etching, gate deposition, stressor deposition, and ion implantation [42]. A related
technique involved patterning the semiconductor substrate before epitaxial growth.
However, damage introduced during the fabrication process generally reduced the
optical efficiency of these dots, and limitations on the tolerances of microelectronic
fabrication led to large variations in dot size.

On the other hand, uniform semiconductor nanocrystals have long been precip-
itated by chemical means [43]. However, since these dots are formed in solution or
in glasses, they are difficult to incorporate into larger semiconductor structures. As
well, they have generally been plagued by carrier traps on the nanocrystal surfaces,
which lead to photobleaching and shelving. All of these problems were solved when
it was found that quantum dots could be grown directly in MBE or MOCVD by a
strain-induced process. This has led to a recent boom in the study of semiconductor
quantum dots, fuelled by the sense that the structures will have many important
applications, particularly in optoelectronics.

2.2 Self-Assembly by Molecular-Beam Epitaxy

2.2.1 Basics of Molecular-Beam Epitaxy

Fabrication of high-quality quantum dots requires a method for high-precision epi-
taxial growth of high-purity semiconductor crystals, such as molecular-beam epitaxy.
Since its invention in the early 1970’s, MBE has grown from a specialized research
tool to a major industrial technique [1, 44]. Fig. 2.3 shows a schematic of a typical
MBE system. Elemental sources are heated in ultra-high vacuum (UHV) to produce
molecular beams, which impinge on a heated substrate. Atomic layer-by-atomic layer
deposition is achieved by using low beam fluxes, which are controlled by varying the
temperature of the source cells. The atomic mean-free path in the beams is generally
larger than the distance between the source and the substrate. Typical growth rates
of 0.1 – 1μm/hr are obtained. Shutters in front of the sources are used to control
growth time. Uniform growth is obtained by rotating the substrate during deposition. In order to obtain low impurity levels, a background pressure of less than \(5 \cdot 10^{-11}\) Torr is maintained. The UHV environment allows for \textit{in-situ} monitoring of growth conditions and growth rate by reflection high-energy electron diffraction (RHEED), laser reflectrometry, or other methods.

Layer-by-layer epitaxy of lattice-matched materials is possible within a range of growth rates and substrate temperatures. The mechanics of this growth mode are described by the Burton-Cabrera-Frank theory. According to this model, growth proceeds according to the following steps: (1) Atoms impinge on the sample surface, where they are adsorbed; (2) The atoms migrate along the surface towards atomic steps, where they are stabilized by the increased number of atomic bonds; (3) The atoms migrate along the step edges to a kink site, where they are incorporated into the lattice. Deposition involves the lateral motion of step edges or the growth of two-dimensional islands until an atomic layer is completed.

The materials most commonly grown by MBE are known as III-V compounds,
because they are composed of one element from Group III of the periodic table, and one element from Group V. In particular, we grow GaAs, AlAs, InAs, and alloys of these materials. Deposition is typically done on the (100) surface of a GaAs substrate. Nearly all the incident Ga, Al, or In is adsorbed onto the surface, while just enough As is adsorbed to maintain stoichiometric growth. The Group III flux thus determines the growth rate. The ratio of As flux to the Group III flux, known as the \( V/III \) ratio, is maintained in the range 15 – 30 for the growth of GaAs and AlAs, while the substrate temperature is maintained in the range 550 – 700°C.

GaAs and AlAs have nearly identical lattice constants, so an AlGaAs alloy can be deposited smoothly on the substrate. InAs, on the other hand, has a lattice constant about 7% larger than that of GaAs, so that thick layers cannot be deposited without forming dislocations. However, if thin layers of InAs are deposited, nanometer-scale islands may be formed.

### 2.2.2 Stranski-Krastanov Growth

Epitaxial growth of \textit{lattice-matched} material (\textit{i.e.}, epitaxial material with essentially the same lattice constant as the substrate) can proceed according to two different growth modes, as illustrated in Fig. 2.4. Layer-by-layer growth, also known as \textit{Frank-Ver der Merwe} growth, has been described above. It occurs when the sum of the epitaxial surface energy and the interface energy is less than the substrate surface energy. If the energy balance is reversed, it is favorable for the epitaxial material to form small \textit{islands} on the surface. This is known as the \textit{Volmer-Weber} growth mode.

For epitaxial growth of \textit{lattice-mismatched} material, it is necessary to consider strain energy as well as surface and interface energies. Growth can initially proceed in a layer-by-layer mode. However, the unit cells of the epitaxial material will be distorted in order to conform to the lattice constant of the substrate, leading to strain in the epitaxial layer. The energy associated with this strain increases as the layer thickness increases. It is possible to relieve the strain by forming small islands on the surface, allowing the lattice constant in the islands to relax towards its natural value. Island formation will be favorable if the energy lost by strain reduction is
greater than the energy associated with the additional surfaces [45]. Deposition of a planar wetting layer followed by three-dimensional islands is known as the Stranski-Krastanov growth mode.

Islanding competes with dislocation as a mechanism for relieving strain in epitaxial layers. If the energy lost by forming islands is greater than that lost by formation dislocations, it is possible to grow coherent islands. Under certain conditions, deposition of InAs on GaAs leads to just such dislocation-free islands. The islands have dimensions on the order of several nanometers in all three directions, making them suitable for use as quantum dots [46, 47, 48]. GaAs deposited on top of the dots forms a planar layer, resulting in a small inclusion of InAs inside a matrix of GaAs. Such self-assembled dots have been extensively studied recently, both for their physical interest and for potential applications [49, 50].
2.2.3 Indium Arsenide / Gallium Arsenide Quantum Dots

The equilibrium picture of quantum-dot formation presented above is not a complete account of how dots form in MBE. It is necessary to consider, as well, the atomic kinetics involved. The density and size of quantum dots is partially determined by the balance between the rates of adsorption and diffusion of In atoms on the sample surface. If adsorption occurs slowly compared to surface diffusion, the surface approaches thermal equilibrium at each moment during growth. This leads to a layer of dots with relatively low density and large diameters. On the other hand, if adsorption is fast compared to diffusion, the kinetics are frozen out. In atoms will tend to aggregate into dots with additional atoms are they are adsorbed nearby rather than with more distant atoms. The result is a higher density of smaller dots for the same amount of adsorbed material. The balance between adsorption and diffusion can be changed by increasing the InAs growth rate (i.e., increasing the adsorption rate), or by increasing the V/III ratio, which leads to a lower diffusion rate.

However, more dramatic differences occur when the substrate temperature is changed. A higher substrate temperature leads to a higher diffusion rate. At the same time, it increases the interdiffusion of adsorbed In atoms and underlying Ga atoms. The epitaxial quantum-dot layer is thus an InGaAs alloy, with a Ga fraction that increases with temperature. A higher Ga content means that the epitaxial layer has an effective lattice constant closer to that of the GaAs substrate. This, in turn, means that the strain energy that drives the islanding process is reduced. This combines with the decreased surface diffusion to produce larger, sparser islands.

This effect is evident in the samples shown in Fig. 2.5. For each of the samples, the equivalent of 1.9 monolayers (ML) of InAs was deposited on a GaAs substrate at a rate of 0.077 μm/hr. All MBE growth was done on a modified Varian model GenII system. A cracker was used to produce a beam of As$_2$ dimers. During InAs deposition, the V/III ratio was approximately 50. The growth was halted after the InAs deposition, so that the dots could be imaged. The exact structure of the dots is therefore different from those used in optical experiments, which are covered with a capping layer of GaAs [51]; the areal density, though, is unchanged. The images in the figure were taken using a Digital Instruments model Nanoscope atomic-force
Figure 2.5: Atomic-force-microscope images of quantum dots grown at different substrate temperatures: (a) 480 °C, (b) 487 °C, (c) 498 °C, (d) 520 °C
microscope in contact mode. The shape of the dots is somewhat blurred by the tip convolution effect. Nonetheless, a clear decrease of dot density and increase of dot size is seen with increasing substrate temperature.

Determination of the substrate temperature during growth is not entirely straightforward. There is a thermocouple on the substrate heater, but it is not in direct contact with the substrate. This leads to an offset between the measured temperature and the actual substrate temperature. The temperature was therefore determined by observing the desorption of the native oxide from the epitaxy-ready GaAs substrates before growth. As the sample is heated in the growth chamber under an As flux, the RHEED signal is monitored. At a substrate temperature of 575 °C, the oxide desorbs from the surface, and the RHEED signal changes abruptly from a diffuse pattern typical of an amorphous surface to a sharp, streaky pattern typical of a crystalline surface.

Although we can grow sparser dots by increasing the substrate temperature, this cannot be continued indefinitely. If the temperature is too high, the In atoms will not adsorb on the surface. As well, a higher substrate temperature means more alloying of the dots. If the Ga content in the InGaAs dots is too high, the bandgap difference between the dots and the surrounding GaAs matrix is low, and the confinement of carriers in the dots is weak. Carriers in the dot will then easily escape to quantum-well states in the wetting layer.

It is possible to further reduce the dot density, though, by reducing the amount of deposited InAs. Although it is difficult to generalize about dot formation, we have seen the following trend. At first, InAs forms a wetting layer with large, monolayer-high islands. After a certain thickness is deposited, a relatively small number of small dots form. As more material is deposited, the size of these dots increases, while the dot density remains nearly constant. Eventually, the size of the dots stops increasing. Further InAs deposition leads to an increase in the dot density, with little change in the average dot diameter. We attempted to stop InAs deposition at the point where the dots reached their maximum size, before the density started to increase.

Figure 2.6 shows an atomic-force microscope image of quantum dots grown under optimized conditions. These dots were grown at a substrate temperature of 530 °C.
2.2. SELF-ASSEMBLY BY MOLECULAR-BEAM EPITAXY

Figure 2.6: Atomic-force-microscope image of quantum dots grown under conditions optimized for low density
The InAs growth rate is 0.054 $\mu$m/hr, and the V/III ratio is 51. 1.75 ML of InAs was deposited. After the InAs deposition, growth is interrupted for 3 sec. in order to allow the dots to stabilize. These growth conditions were used for the efficient source of single photons described in Section 5.2. Dots described in other sections were grown under different, but similar, conditions.

2.3 Quantum-Dot Spectroscopy

2.3.1 Optical Transitions in Self-Assembled Quantum Dots

Self-assembled InAs / GaAs quantum dots are appealing as optical emitters for several reasons. The bandgaps of the two materials line up such that a large potential well is formed for both electrons and holes. InAs has a direct bandgap, which allows for efficient optical transitions. The dipole moment of the atoms in the dot effectively add up to create a single dipole with a large oscillator strength. Finally, the energy of transitions between confined states in the dots lies in the near infrared, allowing for efficient photon detection.

Our principle tool for investigating optical emission from quantum dots is photoluminescence (PL). This process is illustrated schematically in Fig. 2.7. Laser light is directed towards the sample, leading to the production of electron-hole pairs. The energy of the incident light may be larger than the GaAs bandgap; this is what we call above-band excitation. In this case, many electron-hole pairs are created in the GaAs matrix surrounding the dot. The carriers diffuse towards the dots. At low temperature, they are rapidly trapped by the dots, and quickly relax to the lowest-energy confined states [52]. They then recombine to emit a photon with a characteristic energy. Alternatively, the incident light can be tuned such that the photon energy is equal to the energy difference between higher-lying states in the dot; this is what we call resonant excitation. In this case, the diffusing and trapping steps from the GaAs are avoided, and electrons and holes are always created in equal numbers directly in the dot.
2.3. QUANTUM-DOT SPECTROSCOPY

Fig. 2.7: Schematic of transitions in an InAs / GaAs quantum dot

Fig. 2.8 shows a schematic of the experimental apparatus used for PL measurements. The sample is held at a temperature of $4 - 5 \text{ K}$ in a cryostat (Janis model ST-100 modified for close optical access, or Oxford Instruments model Microstat He). In this device, a continuous flow of liquid helium cools a copper mass. Attached to the mass is a cold finger, also made of copper, which terminates in a sample holder. The sample is soldered to the holder using high-purity indium. The cryostat is evacuated before cooling to a pressure of about $5 \cdot 10^{-3} \text{ Torr}$, in order to minimize condensation on the sample. The sample holder also has a resistive heater, which allows its temperature to be regulated. Within 3 mm of the sample surface is a window, which allows optical access from the outside of the cryostat.

A lens next to this window is used to focus pump light onto the sample. The pump source is a mode-locked Ti:sapphire laser (Coherent model Mira), pumped by a diode-pumped, frequency-doubled Nd:vanadate laser (Coherent model Verdi), which emits 3 ps-long pulses of light every 13 ns. The laser wavelength is tunable from 700 nm to 1000 nm. The laser can also be run without mode locking, for continuous-wave
CHAPTER 2. SELF-ASSEMBLED QUANTUM DOTS

Figure 2.8: Schematic of the apparatus used for photoluminescence measurement

(cw) pumping. The laser pulses are focussed onto a spot size of about 20 μm at a steep angle, while the emitted light is collected in the normal direction, so that only a relatively small amount of scattered laser light passes through the collection lens. This lens is an aspheric singlet with a numerical aperture of 0.5. A spectral filter after the lens provides additional rejection of scattered laser light. Collected light is focussed onto a removable pinhole for spatial filtering, so that a region of the sample 5 μm in diameter is effectively isolated. The cryostat is mounted on a translation stage, so that different parts of the sample can be observed.

Following the lens, the light is directed in one of two directions. A charge-coupled device (CCD) camera is used during alignment to observe the sample through the collection lens. For measurements, the emitted light is sent through a multimode-fiber bundle to a spectrometer (Acton Research model Spectra-Pro 750). In the spectrometer, the light is dispersed by a grating before being imaged onto a CCD detector array. The array consists of several columns of pixels, so that a range of wavelengths
2.3. QUANTUM-DOT SPECTROSCOPY

Figure 2.9: Photoluminescence spectrum from an ensemble of InAs / GaAs self-assembled quantum dots

can be detected simultaneously. The detected wavelength range is changed by rotating the grating using a stepper motor. The maximum attainable resolution is about 0.05 nm. The CCD array is cooled with liquid nitrogen to eliminate noise.

Fig. 2.9 shows luminescence from dots grown at a relatively low substrate temperature. The intense peak at about 845 nm, of which only part is visible in the figure, is emission from one-dimensionally confined states in the wetting layer. The weaker, inhomogeneously broadened emission in the range 900 – 1000 nm comes from the quantum dots. Since the dots are formed through a self-assembly process, they have a distribution of sizes and shapes. This leads to a variation in the confined energy levels, reflected in the large inhomogeneous linewidth. Two broad peaks can be seen. The longer-wavelength peak corresponds to excitons in the ground state of the quantum dot (referring to Fig. 2.7, electrons and holes both in the $n = 1$ state). The shorter-wavelength peak corresponds to excitons in a higher-lying confined state.

Fig. 2.10 shows luminescence from dots grown under the optimized conditions described in Section 2.2. The substrate temperature during this growth is relatively
high, so that a substantial amount of interdiffusion takes place. The wetting-layer peak is now located at about 855 nm, while the quantum-dot emission is centered around 865 nm. Since the density of dots in this sample is low, the total quantum-dot emission intensity is also low. As well, the potential well of these InGaAs dots is shallow, due to the high GaAs fraction. This means that there is no excited-state emission between the ground-state and wetting-layer emission. The quantum-dot peak is again broad, due to the size and shape variation among the dots. In order to recover the narrow homogeneous linewidth, we must isolate a single quantum dot from the ensemble.

2.3.2 Light from Single Quantum Dots

Several techniques have been explored to isolate the luminescence of a single quantum dot. A small volume of the sample can be selectively excited by cathodoluminescence [53], current injection by a scanning tunnelling microscope [54], or near-field scanning optical microscopy (NSOM) [55]. Alternatively, microscopic holes can be patterned.
into a metal layer deposited on the sample. When the sample is illuminated from above, only dots below the holes are excited, and only the light that escapes through the holes is collected [56, 57]. Perhaps the simplest way to isolate the luminescence from a single quantum dot, though, is to remove all the other dots. This can be done by etching mesas into the sample [48]. If the mesas are made small enough, and the dot density is low enough, then the average number of dots per mesa can be less than one. Mesas can then be found that contain only a single quantum dot.

We isolated single quantum dots by etching mesas with diameters down to 200 nm. (For details on the processing steps involved, see Section 3.2.) We then illuminated single mesas, and collected the resulting luminescence. Fig. 2.11 shows the PL spectra of a particular quantum dot under above-band excitation and under resonant excitation. Several narrow emission lines are seen, corresponding to different charge configurations of the dots [58, 59, 60]. The total energy of the quantum-dot system is modified by Coulomb interaction among the carriers. The quantum-confined wavefunctions of a single electron and a single hole are correlated by the electrostatic potential, forming a quasi-particle known as an exciton. Likewise, the wavefunctions of two excitons are correlated to form a biexciton. Similarly, higher-number multiexciton states can be formed, as well as charged excitons, consisting of two holes and an electron, one hole and two electrons, etc. [61]. We can identify the peaks labelled “3” and “4” in above-band excitation as charged excitons, because they disappear for resonant excitation, where only neutral exciton complexes can be formed. Peaks “1” and “2,” on the other hand, can be identified as excitonic and biexcitonic emission, respectively.

There are several types of experimental support for this identification. Fig 2.12 shows the above-band cw pump-power dependence of the intensities of lines “1” (single-exciton) and “2” (biexciton). We can see linear growth of the single-exciton peak and quadratic growth of the biexciton peak in the weak-pump limit. This is what is expected if carrier capture into the dot is a stochastic process, independent of the occupation of the dot.

Further evidence for the attribution of single-exciton and biexciton peaks comes from time-dependent luminescence measurements [62]. These were done by replacing
Figure 2.11: Photoluminescence spectra from a single InAs / GaAs self-assembled quantum dot, under above-band excitation (a) and resonant excitation (b)
the spectrometer illustrated in Fig. 2.8 with a streak camera (Hamamatsu model C5680). Light that enters the streak camera encounters a photocathode, leading to the generation of an electron pulse, which is accelerated by an electrical field through a vacuum tube. As the electrons move through the tube, they pass between a pair of electrodes. A voltage ramp is applied across the electrodes, so that the electrons are deflected laterally during their travel, with the amount of deflection being proportional to their time of generation. The voltage ramp is triggered by pulses from the pump laser. The electrons then encounter a multichannel plate, where they are amplified before striking a phosphor screen. The end result is a light pulse whose vertical position on the screen depends on the time that the incident light reached the streak camera. The image on the phosphor screen is collected and integrated by a CCD camera. At high gain, a threshold detection level can be set, and single-photon events can be observed. The images collected are corrected for noise from the electron multiplication, non-uniform sensitivity of the CCD detector, tilt and curvature of the sweep, and a small number of cosmic-ray events.
The streak camera is preceded by another spectrometer (Hamamatsu model 5095). The spectrometer disperses the light in the horizontal direction, so that streak-camera images are two-dimensional maps of intensity as a function of time and wavelength. The spectrometer has a wavelength resolution of 0.13 nm. It also determines a temporal resolution, because of the Fourier-transform relations between pulse duration and spectral width. At high spectral resolution, the time resolution of the system is about 25 ps.

Using this system, we have measured emission intensity from the single quantum dot as a function of time after excitation and of frequency. The time-dependent intensity was integrated over frequency windows that correspond to the peaks in Fig. 2.11. Resulting curves for various pump powers are shown in Fig. 2.13. The hollow lines show the results of a model fit. The results exhibit a cascaded emission process in the dot [63]. Under weak excitation, the single-exciton line appears quickly after the excitation pulse and then decays exponentially. Under higher excitation power, however, the one-exciton line reaches its maximum only after a significant delay. Most of the emission immediately after the excitation pulse now comes from the biexciton line. The biexciton must recombine before the population of the dot is reduced to one electron-hole pair, and only then can the one-exciton emission occur.

Additional support for the attribution of single-exciton and biexciton lines comes from measurements of photon cross-correlation [64]. Using the experimental configuration described in Section 5.1, we measured the correlation between photons emitted at the single-exciton frequency and photons emitted at the biexciton frequency. For example, we could determine the conditional probability of detecting a single-exciton photon at a time \( \tau + t \) after the detection of a biexciton photon at time \( t \). As expected, this detection probability is nearly zero for small negative \( \tau \), while it reaches a maximum for small positive \( \tau \). In other words, if photons are detected at both the single-exciton and biexciton frequencies, the biexciton photon will be detected first, and the single-exciton photon will be detected shortly afterwards. This indicates a deterministic two-stage decay process from the biexciton state through the single-exciton state.

We can thus isolate the luminescence of a single quantum dot, and identify the
Figure 2.13: Time-dependent luminescence intensities of the single-exciton line (black) and the biexciton line (grey) after pulsed excitation above the GaAs bandgap, for average incident pump power of 27 $\mu$W (a), 54 $\mu$W (b), 108 $\mu$W (c), and 432 $\mu$W (d)
different spectral lines in the emission. However, we can collect only a small fraction of the emission from this quantum dot. We would like to couple more of the spontaneous emission into useful directions. This can be done using the techniques of cavity quantum electrodynamics.
Chapter 3

Micropost Microcavities

3.1 Basics of Cavity Quantum Electrodynamics

3.1.1 Theory

If we want to collect more of the spontaneous emission from a quantum dot, we can redirect the light using mirrors, lenses, and other linear optical elements. In general, such an interaction will change the photon statistics of the light. However, this can be avoided by modifying the nonlinear interaction of the quantum-dot dipole with the electromagnetic vacuum, thereby modifying the spontaneous-emission characteristics of the dot. This is done by placing the dot in a microscopic optical cavity. Spontaneous-emission modification was first proposed by Purcell in 1946, and is therefore sometimes known as the Purcell effect [65]. It is also referred to by the name of cavity quantum electrodynamics (cavity QED) in the weak-coupling regime, where spontaneous emission is an irreversible process, but is modified by the presence of an optical cavity.

Spontaneous emission is a QED effect because it cannot be described without field quantization. The electron orbitals of an atom, for example, are stable solutions of Schrödinger’s equation. In a semiclassical theory, transitions from an excited state to a ground state must be driven by an incident field. The spontaneous downwards
transitions required for thermal equilibrium can be recovered by quantizing the elec-
tromagnetic field (or some other field that interacts with the atom, such as the phonon
field in the solid state).

In a second-quantized theory of the electromagnetic field, there is a non-zero
energy even when no photons are present. This vacuum state (or dark state) has a
zero-point energy equivalent to the energy of a single photon in each spatial mode.
The energy can be thought of as being carried by virtual photons, or zero-point
fluctuations. These fluctuations provoke spontaneous emission in much the same way
that real photons provoke stimulated emission. If a cavity is used to change the density
of optical modes surrounding the atom, then, the spontaneous emission properties of
the atom will be modified. This can be done by using an optical cavity. If the atomic
transition is resonant with a cavity mode, the vacuum field is enhanced, and the
spontaneous emission rate into the mode is increased. Similarly, if the transition is
non-resonant with the cavity modes, the vacuum field is reduced, and the spontaneous
emission rate is decreased.

Alternatively, the modification of spontaneous emission can be thought of as an in-
terference phenomenon involving virtual photons. An off-resonant cavity prevents an
atom from emitting a photon, because the photon would have interfered destructively
with itself had it ever existed. Similarly, for an on-resonant mode, virtual emission
events interfere constructively, and the emission rate is enhanced.

Although these heuristic models give some insight, we would like to have a more
rigorous basis for treating the modification of spontaneous emission. We start by
considering the simple Jaynes-Cummings model for a two-level atom on resonance
with a single cavity mode at frequency $\omega$. The atom is assumed to be located at
the antinode of the standing wave in the cavity, with its dipole moment oriented
parallel to the electric field. The cavity lifetime is assumed to be infinite, and the dot
linewidth is assumed to be zero. The system Hamiltonian can then be written as

$$\hat{H} = \hbar \omega (\hat{a}^\dagger \hat{a} + \hat{\pi}^\dagger \hat{\pi}) + \hbar g (\hat{\pi}^\dagger \hat{a} + \hat{a}^\dagger \hat{\pi}),$$  \hspace{1cm} (3.1)

where $\hat{a}$ is the annihilation operator for photons in the cavity mode, and $\hat{\pi} = |g\rangle \langle e|$ is
the atomic lowering operator (i.e., the operator that takes the atom from the excited state $|e\rangle$ to the ground state $|g\rangle$). The vacuum Rabi frequency $g$ describes the coupling between the atomic transition and the cavity photons, and is given by

$$g = \frac{eD}{\hbar} \sqrt{\frac{2\hbar\omega}{V_o}},$$

(3.2)

where $D$ is the atomic dipole moment, and $V_o$ is the volume of the cavity mode.

The Jaynes-Cummings Hamiltonian results in the following Heisenberg equations of motion:

$$\frac{d}{dt} \hat{\sigma}_e = ig(\hat{a}^\dagger \hat{\pi} - \hat{\pi}^\dagger \hat{a}),$$

(3.3)

$$\frac{d}{dt} \hat{\sigma}_g = -ig(\hat{a}^\dagger \hat{\pi} - \hat{\pi}^\dagger \hat{a}),$$

(3.4)

$$\frac{d}{dt} \hat{\pi} = ig(\hat{\sigma}_e - \hat{\sigma}_g)\hat{a},$$

(3.5)

$$\frac{d}{dt} \hat{a} = ig\hat{\pi},$$

(3.6)

where $\hat{\sigma}_e = |e\rangle\langle e|$ and $\hat{\sigma}_g = |g\rangle\langle g|$ are the population operators for the atomic excited state and ground state, respectively. Solutions of the equations of motion describe vacuum Rabi oscillation. Consider a system that starts with the atom in its excited state and with no photons in the cavity. Free evolution of the system will involve coherent oscillation back and forth, at frequency $g$, between this initial state and the state where the cavity contains one photon and the atom is in its ground state.

The effect of photon loss from the cavity can be accounted for by introducing damping of the cavity photons. That is, Eqn. 3.6 is replaced by the following quantum Langevin equation:

$$\frac{d}{dt} \hat{a} = -\frac{\omega}{2Q}\hat{a} - ig\hat{\pi} + \hat{F}(t),$$

(3.7)

where $Q$ is the quality factor of the cavity (that is, the ring-down time, in units of the optical period), and the fluctuation operator $\hat{F}(t)$ describes noise due to the field decay. We neglect dephasing of the atomic dipole, assuming it is slow compared to the cavity damping. We consider the weak-coupling regime, where $g \ll \omega/Q$. In this
case, solving the equations of motion for the excited-state population gives

$$\langle \sigma_e \rangle \approx \exp \left[ -(2g^2 Q/\omega)t \right], \quad (3.8)$$

where we have taken $\langle \sigma_e(t = 0) \rangle = 1$.

To determine the spontaneous decay rate for the atom without a cavity, we generalize the Jaynes-Cummings Hamiltonian to allow for a continuum of modes. The Hamiltonian becomes

$$\hat{H} = \hbar \sum_\lambda \omega_\lambda (\hat{a}_\lambda^\dagger \hat{a}_\lambda + \hat{\pi}_\lambda \hat{\pi}_\lambda) + \hbar \sum_\lambda g_\lambda (\hat{\pi}_\lambda^\dagger \hat{a}_\lambda + \hat{a}_\lambda^\dagger \hat{\pi}_\lambda), \quad (3.9)$$

where $\lambda$ indexes the different modes. The equation of motion for the atomic excited state, using the initial state $\langle \sigma_e(t = 0) \rangle = 1$, is

$$\frac{d}{dt} \langle \sigma_e(t) \rangle = - \sum_\lambda g_\lambda^2 \int_0^t dt' \exp \left[ -i(\omega_\lambda - \omega_a)(t - t') \right] \langle \sigma_e(t') \rangle, \quad (3.10)$$

where $\omega_a$ is the frequency of the atomic transition. We replace the sum over modes by an integral over the density of modes $\rho(\omega)$, giving the differential equation

$$\frac{d}{dt} \langle \sigma_e(t) \rangle \approx \pi g^2(\omega_a)\rho(\omega_a)\langle \sigma_e(t) \rangle. \quad (3.11)$$

This is identical to Fermi’s golden rule. Substituting in the free-space density of modes $\rho(\omega) = (4/3\pi)(V/(2\pi)^3)(\omega^2/c^3)$ gives the free-space decay rate:

$$\gamma_o = \frac{2e^2 D^2 \omega^3}{3\pi \hbar c^3}. \quad (3.12)$$

This is equivalent to Einstein’s $A$ coefficient.

Using Eqn. 3.2, we can thus write the ratio of the spontaneous emission rate for an atom in a cavity to the emission rate for an atom in free space:

$$\frac{\gamma}{\gamma_o} = \frac{3Q \lambda^3}{4\pi^2 V_o}, \quad (3.13)$$
where $\lambda$ is the wavelength of light in the cavity medium. This ratio is sometimes referred to as the Purcell factor.

### 3.1.2 Atomic Cavity QED

The first measurements demonstrating lifetime modification were performed by Drexhage in the early 1970’s [66]. In these experiments, single layers of molecules were deposited on a reflecting surface; the interface between the molecular layers and air acted as the upper layer of the cavity. Despite the fact that the optical transitions were not resonant with the cavity, a significant variation in emission rate was seen as the distance between the molecular layer and the mirror was changed.

Greater lifetime modification requires a cavity that has a mode volume on the order of the optical wavelength and that has a high quality factor. The first such cavities were superconducting microwave resonators, tuned to the frequency of transitions between Rydberg levels of sodium atoms in an atomic beam [67]. The spontaneous decay rate was increased by a factor of more than fifty in this configuration. Similar experiments using a parallel-plate cavity demonstrated a twenty-fold decrease in the spontaneous emission rate in the off-resonance condition [68]. Inhibited radiative decay was also seen for the cyclotron motion of a single electron in a microwave cavity formed by the electrodes of a Penning trap [69].

Modification of atomic transitions at optical frequencies is more difficult, because the required mode volume is much smaller. The first solution to this problem was to use a confocal resonator, for which all the transverse modes are degenerate. In this way, the dipole of a single atom interacted simultaneously with a multiplicity of cavity modes, and lifetime modification of up to 1.6% was observed [70]. Later, optical transitions of dye molecules were coupled to single optical modes of a microscopic Fabry-Perot cavity [71].

### 3.1.3 Semiconductor Cavity QED

If the emission rate of an atom is strongly enhanced by its interaction with a particular cavity mode, the majority of the light emitted by the atom will end up in that mode.
We can determine the coupling coefficient $\beta$ into a single cavity mode in terms of the enhanced emission rate $\gamma$ and the emission rate $\gamma_o$ in the absence of a cavity:

$$\beta = \frac{\gamma - (\gamma_o - \gamma_c)}{\gamma}$$

(3.14)

where $\gamma_c/\gamma_o$ is the fraction of radiation that would be coupled into the cavity mode in the limit of zero photon storage time (i.e., for $Q \to 0$).

High $\beta$ can mean low threshold for lasers. The lasing threshold is usually defined as the point where the gain due to an inverted medium overcomes losses [72]. Fundamental losses in a laser are due to the finite $Q$ of the laser cavity and spontaneous emission into non-lasing modes. By increasing $\beta$, then, the threshold pump power can be reduced. This was first observed for dye molecules in a microscopic Fabry-Perot cavity [73].

This observation led to an interest in using cavity-QED effects to reduce the threshold current of semiconductor lasers. Epitaxial deposition techniques such as MBE allow for the fabrication of monolithic Fabry-Perot cavities with microscopic mirror spacing, known as microcavities. The mirrors, known as distributed Bragg reflectors (DBR’s), consist of alternating layers of GaAs and AlGaAs, with the thickness of each layer equal to one quarter of the optical wavelength in the material. For a range of wavelengths and incidence angles known as the stopband, the partial reflections from all of the interfaces in the DBR’s add up in phase, making the structure a high reflector. Between the DBR’s, a spacer layer of GaAs (or AlGaAs) is grown with an optical thickness of one wavelength (or one half-wavelength). This spacer serves as the optical cavity: the reflections from the two DBR’s form a standing wave, whose antinode is located in the center of the spacer layer.

The active region is located at the antinode position. Originally, the active region consisted of one or several quantum wells (QW’s). Excitons in the quantum wells replaced the atoms in the earlier lifetime-modification experiments. The excitons are distributed throughout the well, and couple to optical modes distributed throughout the planar cavity, leading to a relatively large collective coupling. Enhancement of spontaneous emission was first observed indirectly in such a system through the
modified far-field emission pattern [74], and was later observed directly through time-resolved photoluminescence [75]. Subsequent experiments demonstrated inhibition of spontaneous emission [76], and up to 30% enhancement of the spontaneous emission rate [77]. The emission-rate enhancement was used to reduce the threshold of quantum-well lasers [78] and to increase the efficiency of light-emitting diodes [79].

The development of strain-induced self-assembly led to attempts to observe similar effects with quantum dots. The narrow homogeneous linewidth of single quantum dots allows for efficient coupling to microcavity modes, while the inhomogeneous distribution of quantum-dot energies allows for the simultaneous observation of on-resonant and off-resonant conditions. A radiative lifetime enhancement of about 30% was observed for InGaAlAs quantum dots coupled to a planar microcavity [80]. Although this is comparable to the lifetime modification obtained for quantum wells in microcavities, it is significantly lower than the modification achieved in atomic experiments. This is primarily due to the presence of leaky modes in the planar DBR microcavities. In such cavities, light is confined only in the normal direction, and can propagate in near-transverse directions through a continuum of guided modes. Although these transverse modes are not resonantly enhanced, there are many of them, so that lateral emission dominates both for the on-resonant and the off-resonant cases. In order to get larger modification of quantum-dot radiative lifetime, it is necessary to confine light in all three dimensions.

3.2 Single-Mode Microcavities

3.2.1 Three-Dimensional Semiconductor Microcavities

Various semiconductor structures have been investigated for the three-dimensional confinement of light. In order to provide significant enhancement of spontaneous emission, these cavities must have a small mode volume and a relatively high quality factor.

Microdisks have been fabricated with diameters of several microns and thicknesses of less than one micron, supported by a thin pedestal less than one micron in diameter
and several microns tall. These disks support whispering-gallery modes, where light circulates around the edge of the disk, constantly directed inwards by total internal reflection [81]. Quality factors of up to 12 000 have been obtained for disks with diameters of 2 \( \mu \text{m} \) containing InAs quantum dots [82]. However, the light in these structures leaks out in all directions due to scattering off surface imperfections. It is thus difficult to couple the light into useful directions.

An alternative structure that has attracted considerable attention is the photonic bandgap material, or photonic crystal [83]. A photonic crystal consists of a dielectric medium that is patterned into a regular three-dimensional structure with a length scale on the order of the optical wavelength. Light is Bragg-scattered by the periodic structure of the crystal. For a proper crystal design, light cannot propagate in any direction within the crystal. The range of optical frequencies that are excluded from the material is called the photonic bandgap, in analogy to the electronic bandgap in semiconductor materials [84, 85]. As in a semiconductor, a defect in the crystal structure will lead to a localized state within the bandgap. For example, if one of the voids in the photonic crystal is filled in with dielectric material, light will be trapped at that point. Such a defect mode could thus serve as a three-dimensional optical cavity with a small mode volume.

In practice, however, it has proven very difficult to fabricate high-quality three-dimensional photonic crystals. Two-dimensional photonic crystals have been investigated instead, with confinement in the third dimension provided by a planar waveguide [86, 87]. Emission from quantum dots has been coupled to single defect modes in these structures [88, 89]. The drawback of these structures is increased coupling to radiation modes, leading to lower ultimate \( Q \) values. So far, it has been difficult to simultaneously obtain the high quality factors and small mode volumes promised for such structures. As well, light extraction into useful directions is not highly efficient.

We therefore focus on an even simpler structure, where a photonic crystal provides confinement in only one direction, while a single-mode waveguide provides confinement in the other two dimensions. Such a structure can be made, for example, by etching a row of holes through a bridge waveguide [90]. However, a simpler method involves etching microscopic posts out of the planar DBR microcavities described
above. The posts act as short cylindrical waveguides, confining light by total internal reflection at the semiconductor-air interface. The DBR’s act as a one-dimensional photonic crystal, with the spacer layer acting as a crystal defect.

Such structures, which we call micropost microcavities, have been studied since the early 1990’s, generally in the context of reducing the threshold current of semiconductor lasers [91]. However, experiments were limited by the quantum-well active regions employed. Considerable non-radiative recombination occurred at the etched surfaces of the posts, drawing carriers out of the quantum wells before they could recombine radiatively. It was not until the development of self-assembled quantum dots that the carriers could be well isolated from the surfaces, and the mode structure of these cavities could be studied in detail [92, 93]. Three-dimensional modes were identified in posts with different diameters and cross-sections [94] and in coupled posts [95]. Laser emission was seen from single three-dimensionally confined modes [96]. The principle advantage of micropost microcavities as compared to other three-dimensional optical cavities is their radiation pattern [97, 98]. Light escaping from the fundamental, or lowest-energy, mode in a cylindrical micropost microcavity is well approximated by a Gaussian beam. It can thus be efficiently coupled into optical fibers, detectors, or other downstream optical components.

3.2.2 Fabrication of Micropost Microcavities

Molecular Beam Epitaxy

Planar DBR microcavities containing self-assembled InAs quantum dots can be grown in a single MBE deposition process. Accurate DBR layer thicknesses are required for high-quality cavities. We thus calibrate growth rates of AlAs and GaAs immediately before growing the microcavity. Light from a semiconductor diode laser is sent through an optical viewport onto the sample at a near-normal angle. We use a laser wavelength of about 980 nm, at which the sample is nearly transparent. The intensity of the reflected light is measured using a silicon photodetector. Sample rotation during growth causes the reflected spot to move over the detector, so we sample the maximum detected signal for each rotation period. The time trace of these samples
CHAPTER 3. MICROPOT MICROCAVITIES

shows a sinusoidal modulation due to interference between light reflected off the sample surface and the first buried interface, with an exponential decay envelope due to optical absorption in the growing layer. The period of the sinusoid is given by \( \lambda_l/(2n) \), where \( \lambda_l \) is the laser wavelength and \( n \) is the refractive index of the growing material. Using this method, growth-rate accuracy of about 0.25\% can be achieved for AlAs and GaAs, provided the substrate temperature is known accurately [99].

First-Generation Posts

Fig. 3.1 illustrates the steps we first used to fabricate micropost microcavities. A layer of poly-methyl methacrylate (PMMA) is spun onto the MBE-grown sample. The PMMA is exposed in an electron-beam lithography system, and is then developed in a solvent mixture. The result is a pattern of small holes in the resist. A thin layer of gold is then evaporated onto the sample. The remaining PMMA is dissolved in a solvent, so that the metal above it is lifted off. The resulting pattern of small metal pads is used as a mask for an anisotropic dry etch. In this way, posts with diameters from 0.5 to 6 \( \mu \)m were etched into the sample.

Fig. 3.2 shows a scanning-electron microscope image of a micropost fabricated using this process. The diameter at the top of the post is 0.5 \( \mu \)m. The etch extends through the top DBR stack, the spacer layer containing the quantum dots, and a small portion of the lower DBR. As will be explained in Section 3.3, the limited etch depth leads to a degradation of quality factor for modes in such posts. We would thus like to extend the etch through the lower DBR.

In the first-generation process, the maximum attainable etch depth was limited by two factors: erosion of the gold mask during the etch, and undercut, or reduction of post diameter with etch depth, visible in Fig. 3.2. A deeper etch requires a mask material more resistant to the etch, a thicker mask, and an etch that results in less undercut. Improving the fabrication procedure requires an understanding of the process steps and how they relate.
3.2. SINGLE-MODE MICROCAVITIES

Figure 3.1: Process steps used to make micropost microcavities
Figure 3.2: Scanning-electron microscope image of a first-generation micropost microp cavity
Electron-Cyclotron-Resonance Plasma Etching

In microelectronic fabrication, etching is the process whereby a pattern from a surface mask is transferred to a sample by removing material not protected by the mask [2]. There are two main methods of etching: wet etching, where the material is dissolved by reagents in an aqueous solution, and dry etching, where the material is eroded by low-pressure gases. Although dry etching is more complex than wet etching, it offers several advantages, including reproducibility, precise control over etch depth, and reduced contamination of etched surfaces. For etching microposts, the most important advantage of dry etching is that it allows for an anisotropic etch. That is, wet etches generally proceed at equal rates in all directions, while dry etches can be made to proceed primarily normal to the sample surface.

One of the more common dry etch techniques is plasma etching. A plasma of the reagent gases is formed in a vacuum chamber above the sample, and ions are accelerated from the plasma towards the sample by an applied bias. We performed our etches using a commercial plasma etcher (PlasmaQuest, currently sold, with some modifications, as Nexx Systems model Cirrus 150). In this system, the sample is introduced through a load lock into a vacuum chamber with a background pressure of about $10^{-7}$ Torr. Process gases enter the chamber, with individual flow rates controlled by mass flow controllers. The chamber pressure is regulated by throttling a valve between the process chamber and a turbomolecular pump. The backside of the sample is cooled by flowing helium gas, and the chuck is also cooled by a temperature-regulated recirculating fluid.

Permanent magnets around the process chamber set up a magnetic field. High-power microwaves are directed through a waveguide into the chamber. The frequency of the microwaves is set to the cyclotron frequency of electrons in the applied magnetic field. Atomic electrons are thus accelerated in circular orbits by the microwave electric field. This method can efficiently ionize and dissociate a wide variety of process gases at low pressures and temperatures. As well, it produces a large number of collisions between electrons and gas atoms, leading to a high density of ions and free radicals. Finally, the microwaves do not directly heat ions, so that the ion energy can be independently controlled.
Control over ion energy is achieved by applying an rf bias between the sample chuck and the process chamber wall. Electrons in the plasma have higher mobility than ions, and are thus better able to respond to the time-dependent electric field. The result is a time-averaged negative bias on the electrodes. The effective bias is generally less than 10 V, and can be controlled by changing the applied RF power. The bias drops across a *boundary layer* above the sample, whose thickness depends primarily on the process pressure. Ions are accelerated across the boundary layer to the sample, where they etch the semiconductor material. The relatively low ion energy minimizes etch-related sample damage.

The plasma used to etch GaAs and AlAs is formed from a dilute mixture of Cl$_2$ and BCl$_3$ in a background of Ar. The Cl$_2$ and BCl$_3$ dissociate in the plasma to produce Cl free radicals, which react with GaAs and AlAs. The Ar is inert, but supplies Ar$^+$ ions, which participate in the etch. There are four principle mechanisms involved in etching, illustrated in Fig. 3.3. In *ion milling*, or sputtering, ions bombard the surface and knock off semiconductor atoms. The process is highly anisotropic, but it is not *selective*; that is, it etches the mask material at nearly the same rate as it etches the semiconductor. *Chemical etching*, on the other hand, is highly selective, but is essentially isotropic. It involves the reaction of neutral chemical species, such as Cl, with the semiconductor material, resulting in volatile reaction products, which escape into the chamber. The other two processes involve a combination of the chemical and the physical, and are intermediate in terms of anisotropy and selectivity. In *ion-assisted chemical etching*, the etch reactions are promoted by the energy of incoming ions. *Ion-assisted protective processes* involve non-volatile reaction products that remain on the sample surface. The layer of products serves to inhibit further chemical etching unless it is first removed by ion bombardment. The main reason to use BCl$_3$ as well as Cl$_2$ gas is to promote the formation of these *passivating* layers.

In the end, there is a tradeoff between the *directionality* of the etch and its *selectivity*. The balance depends on which reaction mechanisms dominate, which in turn is determined by the energy of the ions striking the surface and by the concentration of reagent gases. There are three process parameters which can be adjusted: the relative flow rates of the gases, the RF power applied to the chuck, and the plasma
3.2. SINGLE-MODE MICROCAVITIES

Figure 3.3: Mechanisms involved in plasma etching of semiconductors: (a) ion milling, (b) chemical etching, (c) ion-assisted chemical etching, and (d) ion-assisted protective etching (from [2])

pressure. Lowering the concentrations of Cl₂ and BCl₃ reduces the chemical component of the etch. Increasing the applied RF power increases the effective DC bias, thereby increasing the ion energy and making the etch more physical. The ion energy is also increased by decreasing the process pressure and increasing the thickness of the boundary layer. Lower pressure also leads to a larger fraction of ions in the plasma, which likewise makes the etch more physical.

We have worked on optimizing the process parameters to minimize the degree of undercutting while maintaining sufficient selectivity. The undercut rate increases with etch depth, as can be seen in Fig. 3.2. We therefore divide the etch into three stages; in each subsequent stage, we decrease the partial pressure of Cl₂ and decrease the process pressure. The applied RF power is increased to maintain a roughly constant DC bias.

We also reduced the undercut by reducing sample heating. During the etch, the sample is heated by ion bombardment, which promotes isotropic chemical reactions. A major initial problem with our process was poor heat transfer between the sample
and the chuck. The difficulty was that the chuck holds only wafers that are 4” in diameter, much larger than our samples. Originally, the samples rested on a Si wafer that was held by the chuck, and thermal contact was poor. We solved this problem by mounting the sample onto the Si wafer using small pieces of double-sided adhesive copper tape (3M type 1182), which has a conductive acrylic adhesive on both sides. We then cooled the chuck to about 3 °C. The reduction of sample heating, as well as reducing undercut, decreased etch-related damage, as can be seen in Fig. 3.4.

Through this optimization, we were able to increase the aspect ratio of etched microposts. However, the tradeoff with selectivity means that the mask was completely eroded long before the etch was complete. We therefore replaced gold as a mask material with nickel, which is significantly more resistant to plasma etching [100], and used thicker layers of metal. This required a re-examination of the lithography and liftoff process.

**Electron-Beam Lithography**

Photolithography is the primary procedure used to define patterns in microelectronic fabrication. However, it is difficult to use optical lithography to expose features smaller than the wavelength of the light used. Energetic electrons, on the other hand, have de Broglie wavelengths much smaller than optical wavelengths, and can be used
to define smaller structures. *Electron-beam lithography* (EBL) is therefore a popular research tool, although its low throughput has limited its industrial application [101].

For lithography, we use a Leica model Stereoscan 440 *scanning electron microscope* (SEM). In this machine, a beam of electrons is generated by thermionic emission from a tungsten filament. The beam is sent through magnetic lenses, which collimate and focus the beam and correct for astigmatism. Apertures are used to limit the beam size, allowing a focussed spot size on the sample as small as 2 – 3 nm. Electrostatic fields deflect the beam, scanning it across the sample. A deflector higher up in the beam path allows for high-speed beam blanking. The beam column and sample chamber are evacuated to a base pressure of about $10^{-6}$ Torr. A motorized stage allows sample positioning along three axes, while manual controls adjust sample rotation and tilt.

For writing lithographic patterns, a secondary control computer is used, which runs the Nanometer Pattern Generation System produced by J. C. Nabity. Exposure involves blanking the beam, deflecting it to the point to be exposed, unblanking the beam for a fixed amount of time, and moving to the next point. The pattern is thereby filled in, point by point, in a serpentine pattern. The degree of exposure is determined by the product of the electron beam current and the dwell time per point, which is quantified as an *area dose*. The other key parameters are the *magnification* and the *beam current*. A high magnification allows small features to be exposed, but restricts the total field of view. Backlash in the motorized stage limits the accuracy of mechanical motion, meaning that patterns line up precisely only within one field of view. A high beam current, on the other hand, allows for quick exposure of the pattern, but reduces the resolution that can be obtained.

The reason for the loss of resolution is increased electron scattering. As the electrons penetrate the resist, they experience many forward scattering events, which broaden the beam. When the electrons penetrate through the resist into the substrate, they are occasionally backscattered, which can further increase the exposed area. During these processes, the electrons are continually slowing down, producing a *cascade* of lower-energy *secondary electrons*. The total area exposed to electrons thus depends on incoming electron energy and current, as well as the resist thickness and density.
This means that exposure of small patterns requires the proper choice of electron-beam resist. The most commonly used resist for EBL is poly(methyl methacrylate) (PMMA) dissolved in a solvent such as chlorobenzene. The sample is coated with the solution by placing a drop on the surface, and then rotating the sample at high speed using a spinner (made by Headway Research Co.). The thickness of the resulting film is determined by the spinning speed and by the concentration of the polymer in solution. After spinning, the sample is baked at a high temperature to remove the solvent. During exposure, high-energy electrons break long PMMA chains into short fragments which can be subsequently removed by an appropriate solvent, or developer. The sensitivity of the resist depends on the molecular weight used, with longer polymer chains requiring higher doses in order to be exposed.

PMMA gives good results in terms of exposure and development, and is easy to use. It is not sensitive to light, and once spun and baked, it can be stored almost indefinitely. It is difficult, however, to use a single layer of PMMA for liftoff of a thick Ni mask. The problem is that holes developed in the resist have nearly vertical profiles. Metal evaporated onto the sample will often be connected with metal deposited on the resist. When the resist is dissolved, some of the metal on the semiconductor will be lifted off along with the metal on the PMMA.

To get around this problem, we use a bilayer resist: a top layer of high-molecular weight PMMA, and a lower layer of a co-polymer of MMA and methacrylic acid (P(MMA-MAA)) [102]. The copolymer is dissolved at high concentration in ethyl lactate, and thus spins into a thick layer. The resist is baked after spinning, and the low-concentration PMMA is spun into a thin layer on top. The co-polymer is not soluble in non-polar solvents such as chlorobenzene, so there is no intermixing of the layers. The co-polymer has a high sensitivity, so a relatively large area is exposed by the electron beam. The PMMA, on the other hand, has a low sensitivity. As well, it is thin, which reduces the effect of forward scattering in the layer, while the thick copolymer layer largely isolates it from backscattered electrons. This means that high resolution is attainable in the PMMA layer. A mixture of a polar and a non-polar solvent develops both layers simultaneously, resulting in an undercut profile, as illustrated in Fig. 3.5.
3.3. Modes in Micropost Microcavities

The Ni layer is subsequently deposited using electron-beam evaporation in vacuum. The undercut prevents bridging between the metal on the resist and the metal on the semiconductor. Following evaporation, the resist is dissolved in acetone. The feature size after liftoff is determined by the size of the small hole in the PMMA layer. We have used this process to lift off Ni layers up to 1000 Å thick, with feature sizes down to 150 nm. The Ni can be used as a mask for deep etching of posts.

Second-Generation Microposts

The final design and process used to make second-generation micropost microcavities is summarized in Appendix A. Fig. 3.6 shows a scanning-electron microscope image of an etched micropost. This post has a top diameter of 0.6 μm and a height of 4.2 μm. The etch proceeds far into the lower DBR stack. Undercutting is still present, but is greatly reduced compared to the first-generation posts (c.f. Fig. 3.2), and occurs mostly below the quantum-dot active layer.

3.3 Modes in Micropost Microcavities

Our goals in designing and fabricating micropost microcavities are to maximize the fraction of light emitted by the quantum dot into a single microcavity mode, and to maximize the fraction of light in the cavity mode that escapes upwards in a single
Figure 3.6: Scanning-electron microscope image of a second-generation micropost microcavity
travelling-wave mode. These both require that we eliminate three primary sources of unwanted loss. The first is diffraction loss in the unetched portion of the lower DBR, as explained below. The second is loss due to the tapered shape of the micropost. These first two losses are related to the geometry of etched microposts, and can always in principle be reduced by improving the fabrication process. A more fundamental source of loss is due to the coupling between three-dimensionally confined cavity modes and radiation modes. As the mode volume is decreased, the overlap between the confined mode and travelling plane-wave modes increases, and leakage of light out of the cavity becomes more significant. Maximizing the Purcell factor thus involves a tradeoff between mode volume and radiation loss.

If the degree of unwanted loss can be estimated, the cavity can be designed such that the reflection loss of the upper DBR dominates over all the other losses. Almost all the light in the cavity mode will then escape through the upper DBR into a single-mode travelling wave. Estimating the unwanted loss requires a detailed understanding of the modes in micropost microcavities. We have therefore developed detailed models of resonant cavity modes in these structures.

3.3.1 Finite-Difference Time-Domain Model

Modes in micropost microcavities have been investigated for several years [103, 104]. However, distributed emitters were usually considered, and idealized post geometries were treated. We would like to model modes in micropost microcavities with realistic shapes and determine the degree of coupling of light from single quantum dots into these modes.

The only way to accurately model resonances of such complex structures is to solve Maxwell’s equations numerically. One of the most popular ways of doing so is the finite-difference time-domain (FDTD) method, proposed by Yee in 1966 [105]. This method is based on a discretization of the differential form of Maxwell’s equations. Specifically, the following two Maxwell’s equations are solved numerically:

\[
\mu_0 \frac{\partial \mathbf{H}}{\partial t} = -\nabla \times \mathbf{E}
\]  

(3.15)
\[ \frac{\partial \mathbf{E}}{\partial t} = \nabla \times \mathbf{H}, \]

where \( \mathbf{E} \) and \( \mathbf{H} \) are the electric and magnetic fields, respectively, \( \mu_0 \) is the susceptibility of free space, and \( \epsilon \) is the dielectric constant of the medium. A Cartesian spatial grid is defined with increments \( \Delta x, \Delta y, \) and \( \Delta z, \) and a time increment \( \Delta t \) is also defined. Any field \( F(x, y, z, t) \) can then be written in discretized form as \( F^n(i, j, k) = F(i\Delta x, j\Delta y, k\Delta z, n\Delta t). \) The electric and magnetic fields are defined on two different grids that are offset by half a step in time and space. The fields can then be alternately advanced in time, using a leapfrog method. Six coupled finite-difference equations are solved, one for each of the components of the electromagnetic fields. The computational mesh is truncated by placing a non-reflecting absorber at all boundaries [106].

This method can model the resonances of arbitrary three-dimensional cavity structures [107]. We use it to determine the modes of a micropost microcavity [108]. An initial field distribution is applied to the analyzed structure, and the fields are subsequently evolved in time. We record the time evolution of the field at a point of low symmetry, and take the Fourier transform of the resulting time series to get the cavity mode spectrum. We then identify a mode of interest with frequency \( \omega_0. \) This mode is isolated by convolving the field in time with an oscillating function of frequency \( \omega_0. \) In the frequency domain, this convolution corresponds to the application of a band-pass filter with central frequency \( \omega_0 \) and with a linewidth determined by the boundaries of the convolution integral. We ignore mode polarization in this analysis, so that any pair of degenerate modes with different polarizations is treated as a single mode.

We can take advantage of the rotational symmetry of the cavities to make the calculations more efficient, reducing the order of the computer memory requirements from \( N^3 \) to \( N^2, \) where \( N \) represents a linear dimension of the computational domain [109]. In this case, each simulation performed by the cylindrical FDTD algorithm is for a particular value of the azimuthal mode number \( m. \)

The flexibility of the FDTD method allows simulation of both ideal structures with straight walls and realistic structures with posts that are undercut as a result
of etching. We take the refractive indices of GaAs and AlAs to be 3.57 and 2.94, respectively, and the thicknesses of GaAs and AlAs mirror layers to be 70 nm and 85 nm, respectively. The central GaAs spacer layer is 280 nm thick and is sandwiched between 15 mirror pairs on top and 30 mirror pairs on the bottom. The entire structure rests on a GaAs substrate. Absorption losses are neglected. The spatial discretization is performed with a 5 nm mesh. For ideal structures, we assume straight walls and etching through the entire bottom mirror. For realistic structures, we consider the first-generation posts, where the etch extends through only the top two pairs of the bottom mirror. In these posts, the cavity diameter is taken to be constant for only 550 nm on top, after which it changes linearly with a slope of approximately 4° with respect to the micropost axis.

Fig. 3.7 shows the calculated electric field intensity of the lowest-order mode in ideal and realistic micropost microcavities with top diameters of 0.5 \( \mu m \). The intensity is represented by a grey scale. Half a longitudinal cross-section of each post is shown. The three-dimensional intensity distribution can be obtained by rotating around an axis running down the center of the post (left-hand side of the figures). Lines represent interfaces between different materials. There is a significant difference in the field distribution between the ideal and realistic posts.

The quality factor of a mode is determined by two independent methods: (1) the decay time constant for energy stored in the mode, and (2) the ratio of the energy stored in the cavity, multiplied by the mode frequency, to the power lost by radiation outside the cavity. Table 3.1 lists the values of \( Q \) calculated for the fundamental modes of ideal posts with three different diameters. The quality factor decreases as the posts get smaller, due to the blue shift of the fundamental mode (also indicated in Table 3.1). As the post diameter decreases, the mode becomes more tightly confined, and its wavelength decreases. The resonance wavelength is thus no longer the wavelength for which the DBR’s were designed, and the DBR reflectivities are reduced. At the same time, the effective incidence angle of light on the DBR’s increases. Since the mirrors are optimized for normal incidence, this also causes the mirror reflectivity to decrease slightly. We note that it would be possible, for a particular post diameter, to compensate for these effects by changing the layer thicknesses in the DBR’s.
Figure 3.7: Electric field intensity for the fundamental mode of an ideal (a) and a realistic (b) micropost microcavity, each with a top diameter of 0.5 \( \mu \text{m} \), calculated by the finite-difference time-domain method.
3.3. MODES IN MICROPOST MICROCAVITIES

Table 3.1: Quality factor $Q$ and resonance wavelength $\lambda$ for the fundamental modes of micropost microcavities, as calculated by the finite-difference time-domain method.

<table>
<thead>
<tr>
<th>Diameter ($\mu$m)</th>
<th>Ideal $Q$</th>
<th>Ideal $\lambda$ (nm)</th>
<th>Realistic $Q$</th>
<th>Realistic $\lambda$ (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>5000</td>
<td>920</td>
<td>166</td>
<td>915</td>
</tr>
<tr>
<td>1.0</td>
<td>7000</td>
<td>976</td>
<td>440</td>
<td>962</td>
</tr>
<tr>
<td>2.0</td>
<td>11 500</td>
<td>993</td>
<td>2500</td>
<td>992</td>
</tr>
<tr>
<td>$\infty$</td>
<td>12 000</td>
<td>1000</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

Figure 3.8: Quality factor and resonance wavelength for the fundamental modes of realistic micropost microcavities, calculated by the finite-difference time-domain method (points) and by the approximate method (line)
However, a much more dramatic decrease in quality factor is seen when we consider posts of realistic shape, indicating that a different loss process dominates over the effects of blue shift. Calculation results are summarized in Table 3.1 and in Fig. 3.8. The additional loss in these posts is due to diffraction in the lower DBR. Light making a longitudinal round-trip in the cavity penetrates a certain distance into the unetched portion of the lower DBR before being reflected back. As it does so, it diffracts outwards, so that only a certain fraction of the light is recaptured in the post. This is also manifest in the significant field intensity outside of the lower portion of the realistic post in Fig. 3.7 (b). This diffraction loss is our motivation to improve the etch process, as described in Section 3.2.

As well as quality factors, we calculate Purcell factors using a method presented in Ref. [110]. The total radiated power is calculated for a classical dipole in the cavity and for a dipole in bulk GaAs. The ratio of these powers gives the ratio of the spontaneous emission rate in the cavity to the emission rate in GaAs. This semiclassical analysis gives the same result as a fully quantum-mechanical analysis [111]. Fig. 3.9 shows the Purcell factors calculated for single quantum dots on resonance with the fundamental mode of cavities with realistic shapes. The quantum dots are assumed to be located near the center of the posts. The degradation in $Q$ overtakes the reduction in mode volume, reducing the Purcell factor as the post diameter decreases. Nonetheless, significant enhancement of spontaneous emission rate is seen for posts of all sizes.

### 3.3.2 Approximate Model

The FDTD calculations are demanding in terms of computer resources and processing time. We therefore developed an approximate, heuristic model, in an attempt to reproduce the major results in a simpler fashion. We followed Panzarini and Andreani [112], and assumed that the field distribution can be separated into two parts: one that depends only on the transverse coordinates $r$ and $\theta$, and one that depends only on the longitudinal coordinate $z$; i.e., $E(r, \theta, z) = E_r(r, \theta)E_z(z)$.

The longitudinal field dependence is calculated using a transfer-matrix method
3.3. MODES IN MICROPOLAR MICROCAVITIES

Figure 3.9: Purcell factors for resonant quantum dots located at the center of realistic micropost microcavities, calculated by the finite-difference time-domain method (points) and by the approximate method (line) [113]. We consider an plane wave incident on a planar microcavity in the normal direction. The forward- and backward-propagating amplitudes at any point constitute a two-component vector. The field vector $\mathbf{v}'$ after the boundary can be determined from the field vector $\mathbf{v}$ before the boundary by multiplying by the appropriate transfer matrix: $\mathbf{v}' = \mathbf{Mv}$, where

$$
\mathbf{M} = \frac{1}{2} \begin{bmatrix}
1 + \frac{k_2}{k_1} & 1 - \frac{k_2}{k_1} \\
1 - \frac{k_2}{k_1} & 1 + \frac{k_2}{k_1}
\end{bmatrix}
$$

(3.16)

In this matrix, $k_1$ and $k_2$ are the wavenumbers before and after the boundary, respectively. Propagation through a uniform layer of thickness $l$ is likewise described by a transfer matrix:

$$
\mathbf{M} = \begin{bmatrix}
e^{ikl} & 0 \\
0 & e^{-ikl}
\end{bmatrix}
$$

(3.17)

The product of successive matrices $\mathbf{M}_n\mathbf{M}_{n-1} \cdots \mathbf{M}_2\mathbf{M}_1$ gives the relation between
the fields at any two points in the structure. The field distribution in the longitudinal direction can thus be calculated. It has a maximum in the center of the spacer layer, and decays over a penetration depth $l_{\text{eff}}$ in the DBR’s. An effective refractive index $n_{\text{eff}}$ can be calculated by taking an average of the material indices across the planar structure, weighted by the electric field intensity.

The field in the transverse direction is then determined by considering an infinitely long, cylindrical, dielectric waveguide with refractive index $n_{\text{eff}}$. Modes of this waveguide can be determined by application of boundary conditions [114]. This results in a characteristic equation with, in general, multiple solutions, each one corresponding to a different waveguide mode. The boundary conditions also give the field distribution in the plane. For example, the fundamental (HE$_{11}$) mode has the following transverse electric field profile:

$$E_t(r, \theta) \propto \left\{ \frac{J_0(\beta r)}{K_0(\beta r)} \right\} (\hat{x} - i\hat{y}), \quad (3.18)$$

where $J_0$ and $K_0$ are zeroth-order Bessel functions of the first and second kind, respectively, and the transverse wavenumber $\beta$ is obtained from the characteristic equation. In this equation, the top line refers to fields inside the waveguide, and the bottom line to fields outside the waveguide. The overall wavenumber $k$ can be obtained by assuming that the longitudinal wavenumber is unchanged from the unguided value $k_0$: $k \approx \sqrt{k_0^2 + \beta^2}$. This allows, for example, the resonance wavelength of the HE$_{11}$ mode to be determined, as shown in Fig. 3.8. The blueshift with decreasing post diameter is similar to that calculated by the FDTD method.

Once the longitudinal and transverse fields have been calculated, they are multiplied together, point by point, to give the three-dimensional field distribution. Fig. 3.10 shows the field intensity calculated for the fundamental mode in the spacer layer of a post with a diameter of 0.5 $\mu$m. As in Fig. 3.7, the intensity is represented by a grey scale, and half a longitudinal cross-section of the post is shown. The distribution is similar to the results of the FDTD calculations for the ideal post.

The approximate model, though, does not account for diffraction loss in the lower DBR. The loss must added heuristically, as follows. A wave with an initial transverse profile given by the waveguide mode is allowed to propagate freely for twice the DBR
3.3. MODES IN MICROPPOST MICROCAVITIES

Figure 3.10: Electric field intensity for the fundamental mode in the spacer layer of an ideal micropost microcavity with a diameter of 0.5 μm, calculated by the approximate method

penetration depth $l_{\text{eff}}$. This represents the spreading of the field as it penetrates into the unetched lower mirror and makes a round trip back to the post. The overlap integral between the propagated field and the waveguide mode is then calculated, giving the fraction of the wave that is recaptured by the post. The remainder is an additional reflection loss in the lower DBR. Although this theory is rather approximate, it reproduces well the modal quality factors determined by the FDTD method, as seen in Fig. 3.8. This lends support to our interpretation of the mechanism for degradation of $Q$ in realistic posts.

Once the field distribution and quality factor have been calculated, the Purcell factor can be determined, according to the following formula:

$$\frac{\gamma}{\gamma_0} = \frac{Q\lambda^3}{2\pi^2 n_{\text{eff}}^3 V_0} + f, \quad (3.19)$$

where $\lambda$ is the cavity resonance wavelength, and we have assumed zero detuning between the quantum dot emission and the cavity resonance. The mode volume $V_0$ is calculated by integrating the field intensity over space and normalizing by the peak intensity. This formula differs from Eqn. (3.13) in that the decay into leaky modes at a rate $f\gamma_0$ is taken into account. Loss into unguided modes is taken to consist of two parts: light incident on the DBR’s at an angle greater than the limit of the angular stopband, and light incident on the post edges at an angle less than the critical angle for total internal reflection. For posts with diameters of 0.5 μm or less, only the
second factor contributes; in this case, \( f \) is constant at about 0.3.

Purcell factors calculated by this method, shown in Fig. 3.9, agree reasonably well with the FDTD results. This means that the heuristic model allows for a relatively simple estimation of the coupling between quantum dots and modes of micropost microcavities.
Chapter 4

Single-Dot Lifetime Modification

4.1 Quantum-Dot Cavity QED

The rate of spontaneous emission from quantum dots can be enhanced by placing the dots in micropost microcavities. In the first experiments of this kind, several layers of quantum dots were grown within a DBR microcavity, which was then etched into posts with diameters down to 0.7 μm [115]. Emission-rate enhancement by a factor of up to five was observed for the quantum dots that were on resonance with the fundamental cavity mode. Inhibited spontaneous emission was later seen for dots in micropost microcavities whose sidewalls were coated with thick gold layers [116].

Similar effects have been seen in microcavities where the lateral confinement is provided by a dielectric aperture rather than semiconductor-air interfaces. In these structures, a layer of AlGaAs with a low GaAs fraction is grown within the spacer layer of a planar microcavity with GaAs / AlGaAs DBR’s. The high-aluminum-content AlGaAs layer can be selectively oxidized at high temperature in a humid environment, producing a dielectric with low refractive index and high optical quality [117]. The oxidation proceeds laterally, and can be stopped before it is completed, leaving a micron-scale aperture. The low index of the oxide layer blue-shifts the cutoff frequency for guided modes in the planar microcavity, leading to confinement of low-energy modes in the aperture [118]. Coupling to confined modes leads to changes in emission rates by a factor of up to 2.3 [119]. This spontaneous-emission
enhancement has been used to increase the efficiency of quantum-dot light-emitting diodes [120]. Confinement in these apertured microcavities is not as strong as in microposts, though, so the maximum Purcell factor is limited.

Microdisks have also been investigated for the enhancement of the rate of spontaneous emission from quantum dots. Coupling to high-\(Q\) whispering-gallery modes has produced lifetime changes by a factor of up to 12 [121].

All of these experiments, though, involve an ensemble of quantum dots coupled to a cavity mode. Since the ensemble is inhomogeneously broadened, some of the dots are on resonance with the cavity, while others are off resonance. The large emission-rate enhancements are only for the sub-ensemble of dots that are resonant with the cavity. The emission rates for off-resonant dots are either unchanged or inhibited. The ensemble-averaged coupling into a single cavity mode is therefore poor; \textit{i.e.}, the ensemble-averaged coupling coefficient \(\beta\) is low. In order to get high \(\beta\), we require a single quantum dot coupled to a single cavity mode. This is also necessary for single-photon emission. We thus investigate micropost microcavities containing single quantum dots.

\section{Coupling Single Dots to Single Cavity Modes}

\subsection{First-Generation Posts}

Our first observation of modified spontaneous emission from single quantum dots involved our first-generation microposts, where the etch extended through only the top DBR stack, the spacer layer, and a small part of the lower DBR stack (see Section 3.2). The spacer contained a relatively dense layer of InAs quantum dots. Posts with larger diameters, then, contained many quantum dots, each with different emission energy. Photoluminescence from these posts consists of broadband emission, filtered by the cavity resonances. A sample spectrum from a post with a diameter of 6 \(\mu\)m is shown in Fig. 4.1. The post was selectively excited above-band by light from a mode-locked Ti:sapphire laser, as described in Section 2.3. The arrows are located at the resonance wavelengths calculated by the approximate method described in Section
3.3. The labels indicate the corresponding transverse waveguide modes.

Fig. 4.2 shows the filtered PL spectrum for a post with a top diameter of 1 µm. Some discrete single-dot lines are visible, but the emission is still generally broadband. Comparing to Fig. 4.1, we can see a blueshift and an increased mode spacing, due to the increased transverse confinement. Also apparent by comparing to Fig. 4.1, however, is an increase in the modal linewidth, corresponding to a decrease in the cavity quality factor $Q$. Measured quality factors for posts of different diameters are shown in Fig. 4.3. Comparing Fig. 4.3 to Fig. 3.8, we can see that the measured quality factors for posts with large diameters are lower than predicted theoretically. This is partially due to imperfections in the MBE-grown DBR mirrors, including roughness at the AlAs/GaAs interfaces and deviations between designed and actual layer thicknesses. It is also due to lateral variation in the thickness of the cavity spacer layer. These effects are accounted for phenomenologically in the approximate theory by introducing additional reflection losses in the top and bottom DBR’s. The magnitude of these losses is set to match the independently measured $Q$ of 2300 for the planar cavity. The results of the approximate theory with this additional loss
4.2.2 Emission-Rate Enhancement

For measurement of recombination rate, the emitted light is sent through a spectrometer attachment to a streak camera, giving time- and frequency-resolved luminescence intensities. The time traces are integrated over narrow frequency windows. A sample curve of emission intensity vs. time is shown in Fig. 4.4. Immediately after excitation by the laser pulse, the intensity rises, as carriers are trapped by the dots and relax to the ground state. Following this is a slower decay, as the carriers in the dot recombine.

We have been able to reproduce this behavior quantitatively using a simple rate-equation model. We consider only the lowest-energy confined states for electrons and holes in the quantum dot. Each of these states can be occupied by at most two carriers. The states are filled by electrons and holes from the surrounding GaAs matrix at rates $\Gamma_e$ and $\Gamma_h$, respectively. These rates incorporate diffusion, trapping, and
4.2. COUPLING SINGLE DOTS TO SINGLE CAVITY MODES

![Graph of quality factors vs. diameter](image1.png)

Figure 4.3: Quality factors of the fundamental modes of micropost microcavities, as measured experimentally (points) and as calculated by the approximate method (line), including additional reflection loss in the distributed Bragg mirrors.

![Graph of time-dependent luminescence intensity](image2.png)

Figure 4.4: Time-dependent luminescence intensity from quantum dots in a micropost microcavity, as measured experimentally (points) and as fitted using a rate-equation model (line).
thermalization, and are assumed to be independent of dot occupation. Recombination of excitons, biexcitons, and charged excitons is described by the rates $\gamma$, $\gamma_2$, and $\gamma_*$, respectively. The rate equations describe the time-dependent probabilities of the quantum dot being in particular states. For example, $P_0(t)$ is the probability of no carriers being in the dot, $P_{1e}(t)$ is the probability that there is only a single electron in the dot, $P_{1e1h}(t)$ is the probability that there is one electron and one hole in the dot, etc.

The temperature is assumed to be low enough that thermal excitation of carriers can be ignored. Photon decay out of the cavity is assumed to be fast compared to other rates, so that re-excitation and stimulated emission due to cavity photons can

Figure 4.5: Photoluminescence lifetime for quantum dots in a micropost microcavity with a top diameter of 2 μm, as measured experimentally (points) and as predicted using the approximate model (dashed line); also shown is the photoluminescence intensity (solid line)
be neglected. The rate equations can then be written as follows:

\[
\frac{d}{dt} \begin{bmatrix}
P_{2e2h}(t) \\
P_{2e1h}(t) \\
P_{1e2h}(t) \\
P_2(t) \\
P_{1e1h}(t) \\
P_{2h}(t) \\
P_{1e}(t) \\
P_{1h}(t) \\
P_0(t)
\end{bmatrix} = \mathbf{M} \cdot \begin{bmatrix}
P_{2e2h}(t) \\
P_{2e1h}(t) \\
P_{1e2h}(t) \\
P_2(t) \\
P_{1e1h}(t) \\
P_{2h}(t) \\
P_{1e}(t) \\
P_{1h}(t) \\
P_0(t)
\end{bmatrix},
\]

(4.1)

where

\[
\mathbf{M} = \begin{bmatrix}
-\gamma_2 & \Gamma_h & \Gamma_e & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -(\gamma_e + \Gamma_h) & \Gamma_e & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -(\gamma_e + \Gamma_h) & \Gamma_e & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -\Gamma_e & \Gamma_e & 0 & 0 & 0 & 0 \\
\gamma_2 & 0 & 0 & 0 & -\Gamma_e & \Gamma_e & 0 & 0 & 0 \\
0 & \gamma_e & 0 & 0 & 0 & -\Gamma_e & \Gamma_e & 0 & 0 \\
0 & 0 & \gamma_e & 0 & 0 & 0 & -\Gamma_e & \Gamma_e & 0 \\
0 & 0 & 0 & \gamma_e & 0 & 0 & 0 & -\Gamma_e & \Gamma_e \\
0 & 0 & 0 & 0 & \gamma_e & 0 & 0 & 0 & -\Gamma_e & \Gamma_e
\end{bmatrix}.
\]

For simplicity, we assume that the trapping rates \( \Gamma_e \) and \( \Gamma_h \) are equal, and are proportional to the number of electrons and holes in the surrounding GaAs matrix. In each period, this carrier number is set to a large initial value by the incoming laser pulse, and then rapidly decays at a rate \( \Gamma_{GaAs} \) due to radiative and non-radiative recombination. This decay rate is taken to be large compared to the rate at which carriers are trapped in the dot, so that the trapping rates are given by \( \Gamma \approx \Gamma_{e} e^{-\Gamma_{GaAs} t} \).

The rate equations are solved numerically, starting with no carriers in the dot. A pump repetition period of 13 ns is used, and the integration is continued until the solution does not change from period to period. The emitted light intensity at the one-exciton frequency is proportional to the probability \( P_{1e1h} \). Good fits to experimental data can be obtained using reasonable rates, as shown in Fig. 4.4.

Although several parameters are used in this fit, the behavior at longer times is described solely by exponential decay at the one-exciton recombination rate \( \gamma \).
regardless of the details of the model. This occurs when all of the free carriers in the GaAs matrix have recombined and there is no more than one exciton left in the quantum dot. An exponential fit to the later part of each time-dependent emission curve is therefore used to determine spontaneous emission rates.

The results for a post with a top diameter of 2 μm are shown in Fig. 4.5. There are several dots in this post, so the PL is inhomogeneously broadened. The measured lifetime reaches a minimum on resonance with the cavity mode, a clear sign that the mode is discrete and three-dimensional. As well, the lifetimes off-resonant with the cavity mode are nearly the same as the lifetime independently measured for quantum dots in the absence of a microcavity. This indicates that the lifetime modification is due to interaction with the cavity mode, and not due to some other effect, such as non-radiative recombination.
4.2. COUPLING SINGLE DOTS TO SINGLE CAVITY MODES

Fig. 4.5 also shows the emission rates predicted by the approximate model. In order to include the effect of detuning between the cavity resonance wavelength $\lambda_c$ and the quantum-dot emission wavelength $\lambda$, Eqn. (3.19) must be modified as follows:

$$\frac{\gamma}{\gamma_0} = \frac{Q\lambda^3}{2\pi^2 n^3 V_0} \frac{\Delta\lambda^2}{\Delta\lambda_c^2 + 4(\lambda - \lambda_c)^2} + f,$$

where $\Delta\lambda = \lambda/Q$ is the cavity linewidth. As well, several dots are resonant with the cavity mode, and their radial location in the dot is unknown. We must therefore average the Purcell factor over different radial positions across the post. When these factors are taken into account, good agreement with the experimental results is obtained.

Smaller posts contain only a few dots. Under high pump power, however, broadband luminescence is still seen. The origin of this luminescence is unclear; one possibility is that it results from partially localized states in the wetting layer [122, 123]. It is filtered by the cavity mode, allowing for the measurement of resonance wavelength and quality factor for small posts. For low pump power, this background signal is absent, and the luminescence from the quantum dots occurs only in discrete, homogeneously broadened lines. The discrete lines can be seen in Fig. 4.6. Also shown in this figure are the measured lifetimes for single dots on resonance and off resonance with the cavity. A significant reduction in exciton lifetime, by a factor of 4.6, is seen for the resonant dot.

The minimum lifetimes for quantum dots of different sizes are reported in the inset of Fig. 4.7. The main part of the figure shows the corresponding coupling coefficients $\beta$, calculated according to Eqn. (3.14). Two theoretical lines are shown, one averaged radially across the post, and one for dots in the center of the post. The results for larger posts agree reasonably well with the average values, while the single-dot result for the 0.5 $\mu$m-diameter post lies between the average and the maximum values. 78% of the light from this dot couples into a single three-dimensional cavity mode.
CHAPTER 4. SINGLE-DOT LIFETIME MODIFICATION

Figure 4.7: Coupling coefficient of spontaneous emission from single quantum dots into the fundamental mode of micropost microcavities, as measured experimentally (points), and as predicted by the approximate model for dots in the center of the post (dashed line) or averaged radially over the post (solid line); the inset shows the corresponding spontaneous-emission lifetimes.

Since this measurement was performed, enhanced spontaneous emission was observed for a single quantum dot coupled to a whispering-gallery mode in a microdisk [124]. However, it is difficult to couple light out of this mode into useful directions. Micropost microcavities are more practical for light-emitting devices, including single-photon sources.

4.3 Second-Generation Posts

Following the initial observation of single-dot lifetime modification, we improved our process to fabricate micropost microcavities, as described in Section 3.2. Light in the second-generation posts does not experience significant diffraction loss in the lower DBR, because the etch now extends deep into that lower mirror. Measured quality factors are thus higher, as shown in Fig. 4.8. The data shown represent the best posts observed. The lower $Q$ for large post diameters as compared to the first-generation posts is by design: fewer upper-DBR mirror pairs were deposited on this sample.
4.3. SECOND-GENERATION POSTS

Despite the improvement, the quality factor still degrades as the post diameter decreases. This has been attributed to scattering by roughness at the post sidewalls [125]. However, FDTD calculations suggest that loss is actually dominated by the tapered shape of the posts. The taper leads to coupling between the fundamental confined mode and other modes, including higher-order waveguide modes and radiation modes. The quality factors of the micropost microcavity modes could therefore still be increased by further improving the etch process.

Nonetheless, $Q$ is significantly improved as compared to first-generation posts. We thus repeated our measurement of spontaneous-emission enhancement using these second-generation posts. We modified our experiment, though, in order to address an objection that had been made to our original measurement [126]. Since the electric field in the microcavity mode varies across the post, dots located at different positions in the post will have different lifetimes, even if they have the same emission energy. In order to attribute the observed lifetime differences among the dots to detuning differences, as we have done in Fig. 4.6, we assume that we have picked a post

Figure 4.8: Quality factors measured for first-generation and second-generation micropost microcavities
that happens to have three dots located at the same radial position. This perhaps makes our demonstration of lifetime modification less convincing. We have also not completely ruled out the possibility that the on-resonant quantum dots in the small micropost microcavity have their lifetime reduced by some mechanism not related to cavity coupling, such as increased non-radiative recombination.

To address this problem, we have tuned the emission of single dots through the cavity resonance. This was done by changing the temperature of the sample. As the temperature increases, the InAs bandgap decreases, leading to a redshift of the quantum-dot emission. The GaAs and AlAs refractive indices also change with temperature, but the resulting shift in the resonance energy of the cavity is smaller than the shift of quantum-dot emission energy. It is thus possible to bring a particular quantum dot into and out of resonance with the cavity mode.

We selected a post with a top diameter of 0.5 µm that showed bright luminescence from two quantum dots, indicated by the arrows in Fig. 4.9. Fig. 4.10 shows
4.3. SECOND-GENERATION POSTS

Figure 4.10: Measured emission intensity from two quantum dots in a micropost microcavity as a function of pump power (points), and linear fits (lines)

the emission intensity from these dots as a function of pump power. The linear relationship indicates that the emission comes from single excitations, removing any ambiguity associated with the different lifetimes of biexcitons. We measured recombination rates from the two quantum dots at different temperatures using a streak camera, as described in Section 4.2. For each temperature, we also measured a spectrum under high excitation, allowing us to determine the cavity resonance frequency. In this way, we obtained a detuning between the quantum-dot emission and the cavity mode to correspond to each measured quantum-dot lifetime.

Results are shown in Fig. 4.11. The emission rate of one dot changes non-monotonically with temperature, first increasing and then decreasing. The other dot, which is initially detuned to the opposite side of the cavity resonance, has a decay rate that decreases with temperature, ruling out the possibility that a thermally-activated process is responsible for the observed lifetime changes. The lifetime for each of the dots has a Lorentzian dependence on detuning, as expected (see Eqn. 4.2). Fitted Lorentzians give Purcell factors of $3.2 \pm 0.4$ and $2.9 \pm 0.2$ for the two dots in the microcavity. The difference between the two Purcell factors is attributable to
Figure 4.11: Measured lifetimes of two quantum dots as a function of temperature-dependent detuning from a micropost microcavity mode (squares for one dot, circles for the other), and Lorentzian fits (lines)

different radial positions of the two dots in the post. The fitted linewidths of the two Lorentzians are the same within the errors, since they couple to the same cavity. The corresponding quality factors are $650 \pm 160$ and $900 \pm 160$, in agreement with the results shown in Fig. 4.8. We have thus clearly demonstrated coupling of a single quantum dot to a single mode of a micropost microcavity. This measurement removes the ambiguity concerning quantum-dot location, and also rules out an alternative mechanism for lifetime modification. Finally, it demonstrates the ability to change the coupling between the dot and the cavity by changing the sample temperature.
Chapter 5

Triggered Generation of Single Photons

5.1 Experimental Method

Excitons, biexcitons, charged excitons, and other complexes in a quantum dot all emit photons with different energies, as shown in Section 2.3. If we excite a dot with a laser pulse, creating a certain number of carriers, the carriers will recombine one pair at a time, and only the last pair will emit a photon at the single-exciton wavelength. The other wavelengths can be filtered out, leaving only one photon for each laser pulse [127]. In other words, the single quantum dot can act as a converter of laser pulses with a macroscopic number of photons into single-photon pulses, synchronized with the excitation pulses – that is, triggered single photons.

The experimental setup used to generate and detect single photons is similar to that used for photoluminescence, with the addition of a Hanbury Brown and Twiss-type apparatus for measuring the photon correlation function. A schematic is shown in Fig. 5.1. A beamsplitter sends photons from the quantum dot towards one of two single-photon detectors. The detectors are EG&G “SPCM” avalanche photodiodes, which have efficiencies of about 40% at the quantum-dot emission wavelength, and 0.2 mm-wide active areas. The small detector areas together with gratings in front of the detectors serve as small monochrometers, with the center wavelengths determined by
the detector positions. The dots are resonantly excited by pulses from a mode-locked Ti:sapphire laser. Additional rejection of scattered laser light and stray room light is provided by a 10 nm bandpass filter in front of each detector.

The electronic pulses generated by the photon detectors are used as start \( (t_1) \) and stop \( (t_2) \) signals for a time-interval counter. Histograms of delays \( \tau = t_2 - t_1 \) are collected and normalized by dividing the peak areas by the photon count rate at each detector, the laser repetition period, and the measurement time. In the limit of low overall efficiency (combined collection and detection efficiency), these normalized histograms approximate the photon correlation function [128].

This last statement can be proven as follows. \( g^{(2)}(\tau) \) is proportional to the probability \( J(\tau) \) of measuring any photon at time \( \tau \) after an initial photon is detected. The measured signal, on the other hand, is proportional to the probability \( K(\tau) \) of

Figure 5.1: Schematic of the apparatus used for generation and detection of triggered single photons
measuring the next photon a time $\tau$ after an initial photon is detected. Let $P_m(\tau)$ be the probability of detecting the $m^{th}$ photon a time $\tau$ after the initial photon is detected. Then,

$$K(\tau) = P_1(\tau), \quad (5.1)$$

$$J(\tau) = \sum_{m=1}^{\infty} P_m(\tau). \quad (5.2)$$

If the $m^{th}$ photon has been detected at time $(\tau - \tau')$, the probability of detecting the next photon at time $\tau$ is simply $K(\tau')$. We can thus write

$$\int_0^{\tau} d\tau' K(\tau') P_m(\tau - \tau') = P_{m+1}(\tau). \quad (5.3)$$

Summing over $m$, we obtain

$$\int_0^{\tau} d\tau' K(\tau') J(\tau - \tau') = \sum_{m=2}^{\infty} P_m(\tau) = J(\tau) - K(\tau). \quad (5.4)$$

Taking the Laplace transform,

$$\tilde{K}(p)\tilde{J}(p) = \tilde{J}(p) - \tilde{K}(p) \quad (5.5)$$

$$\Rightarrow \tilde{K}(p) = \frac{\tilde{J}(p)}{1 + \tilde{J}(p)}, \quad (5.6)$$

where $\tilde{K}(p)$ and $\tilde{J}(p)$ are the Laplace transforms of $K(\tau)$ and $J(\tau)$, respectively. However, $\tilde{J}(p) \propto \eta_{\text{tot}} \tilde{J}_o(p)$, where $\eta_{\text{tot}}$ is the overall efficiency and $\tilde{J}_o(p)$ is the Laplace transform of the photon detection probability in the limit of perfect efficiency. In the limit $\eta_{\text{tot}} \ll \tilde{J}_o(p)$, the detected signal approaches $g^{(2)}(\tau)$.

Our first measurement involved a single quantum dot without a microcavity, described in Section 2.3 [38]. In this case, $\eta_{\text{tot}} \approx 3 \cdot 10^{-4}$. The measured time-interval histograms are shown in Fig. 5.2. The different figures correspond to different incident laser powers, and the numbers above the peaks indicate the normalized peak areas. The central peaks around $\tau = 0$ have areas significantly less than one, indicating strong anti-bunching. For 0.88 mW average pump power, the single-exciton line
is fully saturated, and the probability of multi-photon pulses is over eight times lower than for Poissonian light.

We have thus demonstrated that a single quantum dot can serve as a single-photon source. However, the efficiency of this source is less than 0.1%, making it impractical for applications. In order to increase the collection efficiency, we couple the quantum dot to a single mode in a micropost microcavity.

5.2 Efficient Generation of Single Photons

We generated single photons efficiently using a single quantum dot in a micropost microcavity. The sample was grown and etched into posts according to the process detailed in Appendix A. We selected a particular post with a top diameter of 0.6 \( \mu m \). The low-temperature photoluminescence from the micropost was measured, as described in Section 2.3, for above-band excitation with a mode-locked Ti:sapphire laser. Results are shown in Fig. 5.3. Emission from the wetting layer is seen in the wavelength range 835 – 845 nm. We believe that the multiple peaks arise from localized states in the disordered quantum well. Spectrally isolated from the wetting-layer emission is emission from a single quantum dot at a wavelength of 855 nm, indicated by the arrow. It is this dot that will serve as a single-photon source.

Single-photon generation was confirmed through a photon correlation measurement, as described in Section 5.1. Fig. 5.4 shows a schematic of the apparatus used. As before, the emission from the dot is spatially and spectrally filtered before being sent to a Hanbury Brown and Twiss-type apparatus. A non-polarizing beamsplitter (NPBS) is followed by retarders to correct for the polarization-dependent phase shifts introduced by the beamsplitter. Each arm after the beamsplitter includes a rotatable half-wave plate to select the measurement polarization, a polarizing beamsplitter, a small monochrometer system to spectrally select the quantum dot emission line, and a single-photon detector. The electrical pulses from one photon detector served as “start” triggers for an Ortec model 566 time-to-amplitude converter (TAC), while the pulses from the other detector served as “stop” triggers for the TAC. The output of the TAC was collected and converted into a histogram by an Ortec model TRUMP-PCI
5.2. EFFICIENT GENERATION OF SINGLE PHOTONS

Figure 5.2: Photon autocorrelation for emission from a single quantum dot under pulsed, resonant excitation, with normalized peak areas indicated
CHAPTER 5. TRIGGERED GENERATION OF SINGLE PHOTONS

Figure 5.3: Photoluminescence from a micropost microcavity containing a single quantum dot

![Photoluminescence graph](image)

Figure 5.4: Schematic of the apparatus used to measure single photons from a quantum dot in a micropost microcavity

![Apparatus schematic](image)
Figure 5.5: Measurement of the linear polarization of a single quantum dot in a micropost microcavity

multi-channel analyzer (MCA); this histogram approximates the photon correlation function.

By rotating one of the half-wave plates and monitoring the change in the photon count rate at the following detector, the degree of linear polarization was determined. The measurement results are shown in Fig. 5.5. The points are experimental results, while the solid line is a sinusoidal fit. The polarization visibility is $33.1 \pm 1.8 \%$. Inserting a quarter-wave plate before the NPBS allows for a similar measurement in a circular polarization basis; this measurement showed very low visibility. We therefore model light from the quantum dot as consisting of a linearly polarized part together with an unpolarized part. This is consistent with scattering of carriers between different spins states in the dot at a rate faster than the radiative recombination rate.

The measured photon-photon correlation function for an average pump power of 10.9 $\mu$W and an integration time of 600 sec. is shown in Fig. 5.6. The points are the raw data collected by the MCA. The side peaks are separated by the laser repetition rate and all have equal areas, while the central peak is nearly absent, reflecting strong
suppression of the multi-photon probability. Due to a low emission rate from quantum dots in this sample, adjacent peaks overlap. It is therefore necessary to fit the data in order to determine the area of the central peak.

In order to reduce the number of free parameters in the fit, we measured the recombination rate of the quantum dot using a streak camera, as described in Section 4.2. The measured lifetime was $\tau = 4.4 \pm 1.2$ ns. We also measured the response time of the autocorrelation setup, using pump-laser light that is scattered off of the micropost. Since the pump pulses are only 3 ps long, the width of the peaks in the measured autocorrelation function corresponds to the timing resolution of the apparatus. The best fit gave a response time $\tau_{\text{resp}} = 473 \pm 29$ ps. This is mostly limited by electronic jitter in the SPCM’s. Each peak in the photon correlation data from the quantum dot is thus described by a two-sided exponential with a decay constant $\tau_{\text{total}} = \frac{\tau}{2} + \tau_{\text{resp}} = 2.7 \pm 0.7$ ns. A fit using this time constant is shown in
5.2. EFFICIENT GENERATION OF SINGLE PHOTONS

Fig. 5.6. There are only two adjustable parameters in the fit: the area of the central peak and the area of all the other peaks. The ratio of these two areas is $g_o^{(2)}$ (see Section 1.1).

The measurement and fit were repeated for various pump powers. The results are summarized in Fig. 5.7. For each point, the measurement polarization was set in order to maximize the photon count rate. Normalized central peak areas as low as 0.10 were obtained, reflecting a tenfold reduction in the multi-photon probability as compared to a source with Poissonian statistics. $g_o^{(2)}$ increases with pump power, suggesting that other states, apart from the desired QD emission, are being excited by the pump laser and are contributing a background of unregulated photons. These states could be in the band tail of the wetting layer, which has been observed to extend into the spectral region where sharp quantum-dot lines lie [123].

In order to measure the efficiency of our single-photon source, we must first calibrate the detection efficiency of our measurement apparatus. This was done by again scattering pump-laser light off the micropost. The scattered power was measured
immediately after the collection lens using a Coherent model LM-2 / Fieldmaster power meter. The measured power was compared to the total photon count rate at the detectors. Including light lost at the polarizers, the overall efficiency of the detection apparatus after the collection lens was determined to be $3.02 \pm 0.16\%$. The collection lens, in turn, captures only part of the light that escapes from the post. The capture fraction can be estimated by calculating the effective divergence angle of light emerging from the post. Since the electric field profile in the microcavity mode is well approximated by a Gaussian, propagation of the mode outside the cavity can be described by Gaussian beam optics [72]. The beam waist in the post is calculated by the approximate method outlined in Section 3.3. The calculations indicate that the lens with numerical aperture of 0.5 collects 22% of the light in the emitted beam.

For each pump power, the total photon count rate is normalized by the laser repetition rate, measured using a high-speed oscilloscope to be 76.3 MHz. The result is divided by the combined efficiencies of the collection lens and the measurement apparatus, giving the mean photon number per pulse $\langle \hat{n} \rangle$. The efficiency $\eta$ is then determined by assuming that the light emitted from the source consists of a statistical mixture of perfectly antibunched single photons and a small background of photons with Poissonian statistics. This mixed state can be described by the following density matrix:

$$\hat{\rho} = \sum_{n=1}^{\infty} \frac{e^{-\mu} \mu^{n-1}}{(n-1)!} |n\rangle \langle n|,$$

where $|n\rangle$ is the $n$-photon Fock state and $\mu$ describes the amplitude of the unregulated background. The mean photon number for this state is

$$\langle \hat{n} \rangle = \sum_{n=1}^{\infty} \frac{ne^{-\mu} \mu^{n-1}}{(n-1)!} = \mu + 1.$$  (5.8)

Likewise, the area of the central autocorrelation peak is given by

$$g_o^{(2)} \langle \hat{n} \rangle^2 = \langle \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a} \rangle = \sum_{n=1}^{\infty} \frac{(n+1)ne^{-\mu} \mu^{n-1}}{(n-1)!}$$  (5.10)
5.2. EFFICIENT GENERATION OF SINGLE PHOTONS

\[ = \mu(\mu + 1) + \mu. \quad (5.12) \]

Combining Eqn. 5.9 and Eqn. 5.12,

\[ g_o^{(2)} = \frac{\langle \hat{n} \rangle^2 - 1}{\langle \hat{n} \rangle^2}. \quad (5.13) \]

The coupling of this state into the travelling-wave mode leaving the top of the micropost microcavity is modelled as an attenuation by a factor equal to the efficiency \( \eta \). In other words, we let \( \langle \hat{n} \rangle^2 \rightarrow \langle \hat{n} \rangle^2/\eta \). Solving for \( \eta \) then gives

\[ \eta = \langle \hat{n} \rangle \left( \sqrt{1 - g_o^{(2)}} \right). \quad (5.14) \]

Eqn. 5.14 is applied to the measured values of \( \langle \hat{n} \rangle \) to determine the efficiency for each pump power. The results are shown in Fig. 5.8. The efficiency increases with pump power \( P \), saturating at higher powers, when more than one electron-hole pair is captured by the dot for each pump pulse. The solid line is a fit according to the saturation equation

\[ \eta = \eta_{\text{max}} (1 - e^{-P/P_{\text{sat}}}), \quad (5.15) \]

where \( P_{\text{sat}} \) is the saturation pump power and \( \eta_{\text{max}} \) is the saturated efficiency. The maximum efficiency according to this fit is 37.6 ± 1.1%. This number represents the external quantum efficiency of the device, and is the figure of merit that allows us to claim that we have produced an efficient source of single photons. It is approximately two orders of magnitude higher than the external efficiency for a quantum dot in bulk GaAs.

The device efficiency is determined by two factors: the coupling coefficient \( \beta \) and the light extraction efficiency \( \eta_{\text{extract}} \). The first factor is the fraction of emitted photons that are captured by a single cavity mode. It can be calculated from the Purcell factor according to Eqn. 3.14. In order to determine the Purcell factor, we measured the recombination lifetimes for excitons in other quantum dots on the same sample that are out of resonance with the cavity mode. We note that these dots may experience some residual emission-rate enhancement, which will mean that we
slightly underestimate the Purcell factor. The lifetimes of the off-resonant dots are too long to measure using our streak camera, so we determined recombination rates by measuring the autocorrelation of photons from these dots. We varied the peak widths when fitting the autocorrelation peaks. The fitted widths give the recombination time, once the instrument response time has been subtracted. The measured off-resonant lifetime is $25.4 \pm 1.4 \text{ ns}$. This represents a Purcell factor of $5.8 \pm 1.6$, corresponding to a coupling coefficient $\beta = 83 \pm 23\%$. To our knowledge, this is the largest emission-rate enhancement yet reported for a single quantum dot in a micropost microcavity.

The second factor in determining device efficiency is the light extraction efficiency, or the fraction of light in the cavity that escapes into a single travelling-wave mode. This is limited by light that is lost from the confined mode due to taper of the sidewalls. The fraction of light lost can be determined by comparing the quality factor $Q$ for the micropost microcavity to the quality factor $Q_o$ for the planar microcavity. $Q_o$ is almost entirely determined by the leakage rate through the top DBR. Assuming that this rate is unchanged when the cavity is etched into posts, any difference between
5.2. EFFICIENT GENERATION OF SINGLE PHOTONS

$Q$ and $Q_o$ will be due to etch-induced loss. The extraction efficiency is then simply given by $\eta_{\text{extract}} = Q/Q_o$.

In order to measure $Q$, we again pumped the micropost with high power, and observed broadband luminescence filtered by the cavity resonance. A Lorentzian fit to the filtered luminescence gives $Q = 628 \pm 69$. A similar measurement on an unetched portion of the planar microcavity gives $Q_o = 1718 \pm 13$, resulting in $\eta_{\text{extract}} = 36.6 \pm 4.0\%$. We note that the measured value of $Q$ can also be used to give a theoretical prediction for the Purcell factor, when combined with the mode volume calculated according to the approximate method. The maximum Purcell factor is predicted to be 5.7, which is consistent with our measurement, assuming that the quantum dot is located near the center of the micropost.

Combining the estimated $\beta$ and $\eta_{\text{extract}}$ gives an expected external quantum efficiency of $30 \pm 9\%$. This agrees, within the error, with the measured efficiency at saturation.

Another group has recently reported single-photon generation using a single quantum dot in a micropost microcavity [129]. Although they note that the device has improved efficiency compared to a reference quantum dot in a mesa structure, they do not provide any explicit treatment of device efficiency. We thus claim to have made the first measurement of an efficient source of single photons using a quantum dot.
Chapter 6

Future Directions

6.1 Device Development

We have demonstrated that a single quantum dot in a micropost microcavity can serve as an efficient source of single photons. However, the performance of our device is still somewhat limited. In particular, although the external quantum efficiency is about 38%, only about 8% of the emitted photons are captured by the collection lens in front of the cryostat. In principle, this can be improved simply by using a lens with a higher numerical aperture. In practice, though, it is difficult to do much better, because of the thick cryostat window between the sample and the lens. More elaborate methods, such as low-temperature confocal microscopy or direct fiber coupling, may allow for higher numerical apertures. However, it may be more straightforward to reduce the divergence of light coming out of the post by increasing the post diameter. Our present lens would collect all of the light escaping in a single mode from a post with a diameter of 2 \( \mu \text{m} \). The larger post diameter would also mean less loss from the microcavity mode due to the taper. As reported in Section 4.3, the quality factor of 2 \( \mu \text{m} \)-diameter posts is practically identical to that of the planar microcavity, indicating that unwanted losses are negligible.

The larger post diameter, though, means a larger mode volume. The resulting reduction in Purcell factor is only partially compensated by the increased cavity quality factor. According to the approximate theory, the maximum Purcell factor in
a post with a diameter of 2 μm is 3.2, corresponding to $\beta = 69\%$. The coupling in a real micropost microcavity is likely to be significantly poorer, since we are less likely to find a single dot close to the center of the larger post.

Another limitation of the current device is the residual probability of multi-photon pulses. Although our results in this respect are comparable to any that have been reported in the literature, there is still room for improvement. Since the spurious photons likely come from states in the extended wetting-layer tail, the most obvious solution is to increase the energy difference between the dot and wetting-layer states. This will require MBE growth conditions to be found that provide relatively long-wavelength dot emission while still giving low dot density.

Even if the efficiency of the device is improved and the multi-photon probability is reduced, there are several practical disadvantages that stand in the way of its widespread use. For example, it is inconvenient to use a mode-locked Ti:sapphire laser in order to operate the device. On the other hand, more compact pulsed light sources could possibly be used as the pump. For example, tunable, mode-locked semiconductor diode lasers can produce pulses about 1 to 5 ps long with repetition rates in the range of 1 to 100 GHz [130]. These compact sources could conceivably be packaged together with the microcavity sample and the necessary optical components into a small “black box.”

Compact packaging is not practical, though, if the single-photon source needs to be cooled to liquid-helium temperatures. We would like our source to operate at room temperature, or at least at temperatures that can be readily achieved using thermoelectric coolers. Unfortunately, InAs quantum dots are not efficient light sources at temperatures above about 70 K, because carriers are thermally excited out of the confined states [131]. In order to increase the maximum operating temperature, we must increase the bandgap difference between the quantum dot material and the surrounding matrix. Dots of InGaN in a matrix of GaN or AlGaN show promise in this regard, exhibiting intense photoluminescence at room temperature [132, 133]. However, they are not as well studied as InAs quantum dots, and growth of nitrides is not as well developed as growth of arsenides. High-Q DBR microcavities are also difficult to grow in this material system because of the lack of a pair of lattice-matched
CHAPTER 6. FUTURE DIRECTIONS

materials with a large refractive-index difference.

Using materials with larger bandgaps also increases the energy difference between the confined electron and hole states, meaning that the emitted photons have shorter wavelengths. InGaN quantum dots, for example, emit light around 450 nm. Optical fibers used for telecommunication, on the other hand, have maximum transmission around 1550 nm [134]. Even the InAs quantum dots we have used emit at wavelengths that are too short for long-distance fiber transmission, and are practical only for free-space communication. On the other hand, quantum dots have been developed that emit at communication wavelengths. One approach has been to reduce the quantum-dot bandgap by growing GaInNAs dots on GaAs [135]. An alternative approach is to grow InAs quantum dots within thin layers of InGaAs [136]. This reduces the confinement energy in the dots by decreasing the bandgap difference between the dots and the surrounding material. Neither of these types of dots are well studied, and it will likely be some time before it is practical to use them as single-photon emitters. Room-temperature operation and emission at communication wavelengths would also appear to be incompatible.

Another goal could be to pump the quantum dot electrically, eliminating altogether the need for the pulsed laser. An electrically driven quantum-dot single-photon source has very recently been demonstrated [137]. In this device, quantum dots are grown inside the intrinsic layer of a vertical $p$-$i$-$n$ diode. Short voltage pulses across the diode populate the dots with carriers, in much the same way as above-band pumping with a pulsed laser. A small aperture in an opaque metal layer on the device surface allows a single dot to be isolated.

The efficiency of this device could be improved by removing the surface metal and incorporating the diode into a micropost microcavity. The structure would be similar to that of vertical-cavity surface-emitting lasers (VCSEL’s). A principle difficulty with fabricating such a device would lie in contacting the doped $p$- and $n$-type reservoirs with metallic leads. Modern VCSEL’s often have contacts directly above and below the active region [138]. However, this requires an etch that extends only through the top DBR, and is thus not compatible with high-$Q$ micropost microcavities. It may be possible to contact microposts from the side, but this will be difficult for posts
with sub-micron diameters. The most practical solution may be to make contacts to the top and bottom of the micropost structure, and use the upper and lower DBR as electrical leads. The problem with this approach is that DBR’s have low electrical and thermal conductivity [139, 140]. In order to send current through the device without excessive heating, it is necessary to dope the mirrors heavily. However, this leads to significant free-carrier absorption in the mirrors, degrading the cavity quality factor. A possible solution is to use \textit{graded} interfaces in the DBR’s. With a graded transition region of 10 nm at each AlAs / GaAs interface, the resistivity can be decreased by a factor of over 300, while the reflectivity is nearly unchanged. We have calculated that graded-interface DBR’s with doping levels of $10^{17}$ cm$^{-3}$ will lead to a potential drop of 35 V across the device at a temperature of 100 K. The corresponding Ohmic heating is 40 nW, corresponding to a temperature drop across the device of 0.05 K. The free-carrier absorption loss in the DBR’s is estimated to be 1.7%, allowing for $Q$ values in excess of 3000.

Since the primary difficulty in making an electrically-pumped device lies with the DBR’s, it would be useful to be able to do away with them. This could also simplify the fabrication of optically-pumped devices. As noted in Section 3.3, spontaneous emission into leaky modes is suppressed for micropost microcavities with small diameters. This is due to total internal reflection at the sides of the posts, and does not depend on the presence of the DBR’s. It may be possible to achieve relatively high $\beta$ values by suppressing emission into radiation modes rather than enhancing emission into guided modes. This would require post diameters near the cutoff for waveguiding (190 nm for a wavelength of 855 nm) [141].

Although microposts without DBR’s would be easier to fabricate, there would still be a basic problem with yield. Since the location of the quantum dots is random, the number and position of dots in a particular etched micropost is unknown. We make hundreds of posts and look for one that has the emission characteristics we want. It would be better to have a way of growing a single quantum dot at a known location on the sample surface. Many techniques for \textit{quantum-dot placement} have recently been investigated, with varying degrees of success. The most promising methods combine self-assembly with \textit{in-situ} microfabrication. For example, one technique involves using
a scanning-tunnelling probe in ultra-high vacuum to leave nanometer-scale deposits on a GaAs surface [142]. GaAs subsequently grown by MBE avoids the deposit, leaving a nanometer-scale hole. If InAs is then deposited under the correct conditions, one dot nucleates in each hole. A similar technique involves nanodeposition using an electron beam rather than a scanning probe [143]. Alternatively, an electron beam can be used to pattern holes in an oxide mask [144]. The mask pattern can be transferred to the GaAs by etching with Cl₂, and the oxide mask subsequently evaporated. The result is again a series of holes that act as nucleation sites for individual quantum dots.

6.2 Micropost Microcavity Optimization

6.2.1 Maximum Quality Factor

Although the quality factors we have achieved for modes in micropost microcavities have allowed us to observe significant spontaneous-emission modification, higher $Q$’s are necessary to see other interesting effects. It should be possible to reduce losses by improving the etch process and reducing the taper in the post diameter. Provided that sidewall roughness can be kept low, so that scattering loss is small, next-generation posts should have significantly higher quality factors.

Eventually, as the etch process is improved, the dominant loss will be due to coupling between the confined cavity modes and radiation modes. We have used the FDTD simulation method to investigate the theoretical upper bounds for quality factors of micropost microcavities. Preliminary results indicate that modes in cavities with optimized designs may have $Q$ values as large as $10^4$ and mode volumes as small as $1.6(\lambda/n)^3$. Such high-quality cavities would allow for the observation of new phenomena, including single-dot strong coupling and single-dot lasing.

6.2.2 Strong Coupling

The emission rate from a quantum dot will not increase indefinitely as the cavity quality factor increases. Eventually, coupling between the dot and the cavity mode
becomes larger than the decay rate of either the exciton or the cavity photon. This is the regime of strong-coupling cavity QED, where the dot-cavity system exhibits coherent oscillations between an exciton in the dot and a photon in the cavity. The rate of oscillation is given by the vacuum Rabi frequency $g$ (see Section 3.1). The crossover from weak coupling to strong coupling is given by the condition

$$g > \frac{\omega}{2} \left( \frac{1}{Q} + \frac{2\pi \gamma_h}{\omega} \right),$$

(6.1)

where $\gamma_h$ is the homogeneous linewidth of the exciton.

Strong coupling was originally observed in atoms using configurations similar to those first used to observe lifetime modification [145]. It was later seen for quantum-well excitons coupled to planar microcavities [146]. In this system, the coupling is enhanced by a factor of $\sqrt{N} \approx \sqrt{A/(\pi a_o^2)}$, where $N$ is the effective number of excitons that couple identically to the cavity mode, $A$ is the coherence area, and $a_o$ is the exciton Bohr radius. More recently, strong coupling has been observed for quantum-well excitons coupled to the discrete modes of micropost microcavities [147, 148]. However, vacuum Rabi oscillation has not yet been observed for quantum dots, because only a small fraction of the quantum-dot ensemble is resonantly coupled to the cavity mode [149]. We are most interested in achieving strong coupling for a single quantum dot. In this case, the Rabi splitting is smaller than in the quantum-well case by a factor of $\sqrt{N}$, which means that we need a microcavity with a correspondingly higher quality factor and smaller mode volume. Optimized micropost microcavities may make this possible.

### 6.2.3 Lasing

Another interesting regime is that of single-dot lasing. Quantum-well lasers have long proven themselves superior to bulk-semiconductor lasers, and it was expected that the advantages would be even greater for quantum-dot lasers. Specifically, the discrete quantum-dot density of states was predicted to result in reduced temperature sensitivity and lower threshold current density [150, 151]. Not taken into account,
however, was the inhomogeneous broadening of the quantum-dot ensemble. Thus, although early devices demonstrated low threshold, the improvement was not dramatic [152]. Further reduction of the threshold current was achieved by reducing the volume of the active region and using high-$Q$ cavities, such as planar DBR microcavities [138] or whispering-gallery modes in microdisks [153, 154]. Quantum-dot lasers have in fact provided record-low threshold current densities of $5 \text{ A/cm}^2$ [155]. However, to take full advantage of the unique properties of the quantum dots, it is necessary to eliminate the inhomogeneous broadening, by isolating a single quantum dot from the ensemble.

A laser with a single quantum dot as an active medium represents an ultimate microscopic limit for semiconductor lasers. The realization of such a device should allow physical investigations similar to those afforded by the single-atom maser [156] and the single-atom laser [157]. A key difference is that the quantum dot stays fixed in place, in permanent interaction with the laser field. In this sense, the device is more like the proposed ion-trap laser [158].

The difficulty with making a single-dot laser is the low gain provided by a single quantum dot, which makes it difficult to reach laser threshold. Definition of a laser threshold in such a microscopic system is controversial [159]. If one uses the conventional, macroscopic definition of threshold, that the gain of the optical mode equals the losses, then one obtains “thresholdless” behavior in the limit $\beta \to 1$. On the other hand, it is possible to define a threshold for all values of $\beta$, at the point where stimulated emission overtakes spontaneous emission, and linear amplification is replaced by nonlinear laser oscillation. This quantum laser threshold occurs when the mean spontaneously-emitted photon number $n_{sp}$ in the laser mode is unity [160].

We use a simplified photon-flow model to estimate the occupation of the laser mode. First, an electron and hole are pumped into the QD. They spontaneously recombine and emit a photon after an average lifetime $\tau_{sp}$. Afterwards, another electron-hole pair is pumped into the QD, and the sequence repeats. The average spontaneous emission rate will thus be $N_A/\tau_{sp}$, where the inversion parameter $N_A$ is the average probability over time that the QD contains an exciton. Of all the spontaneously-emitted photons, a fraction $\beta$ will be captured by the optical cavity
mode. The captured photons will remain in the cavity for an average storage time 
\( \tau_{ph} = Q / \omega \) before leaking out. Combining all the above factors, we obtain a simple 
expression for the threshold condition:

\[
n_{sp} \approx \frac{3 \tau_{ph} N_A}{\tau_{sp}} \geq 1 .
\] (6.2)

This condition means that the cavity \( Q \) must be high in order to reach threshold. 
Optimized micropost microcavities, though, may make single-dot lasing possible.

### 6.3 Microsphere Single-Dot Laser

#### 6.3.1 Proposed Experiment

Fabrication of micropost microcavities with very high quality factors will likely prove 
difficult. We have therefore investigated other three-dimensional microcavities for 
coupling to single quantum dots. Whispering-gallery modes in glass microspheres, 
for example, can have very high quality factors [161].

Fused-silica spheres with diameters from 25 to 100 \( \mu \)m can be made by selectively 
melting the tip of an optical fiber. Surface tension shapes the glass into quasi-spherical 
shapes; a fiber stem, useful for positioning, remains connected to the sphere. Among 
the resonances of these structures are the whispering-gallery modes (WGM’s). In 
a ray-optics picture, these modes correspond to light travelling around the equator 
of the sphere, constantly deflected inwards by total internal reflection. \( Q \) values as 
high as \( 3 \cdot 10^9 \) have been observed for these modes [162]. This represents the highest 
quality factor that has been obtained in any optical microcavity.

Coupling quantum dots to microsphere modes is less straightforward than coupling 
to modes of micropost microcavities. Some progress has been made in terms of doping 
microspheres with quantum dots, but the quality factors of WGM’s in these spheres 
have been limited [163, 164]. High quality factors have been maintained by chemically 
bonding free-standing nanocrystals to the outside surface of glass microspheres [165]. 
However, the positions of the nanocrystals on the sphere are random, so it is difficult
Figure 6.1: Schematic of the proposed structure to position a microsphere above a single quantum dot to get coupling between a single quantum dot and a WGM. As well, these nanocrystals suffer the usual problem of photobleaching.

We propose to avoid these problems by coupling the microsphere to a single self-assembled InAs quantum dot, isolated in a mesa. The microsphere is positioned relative to the mesa in order to optimize the coupling between the quantum dot and a single WGM. Fig. 6.1 shows a schematic of a possible arrangement. Standard microfabrication techniques can be used to define raised ledges around the mesa containing the quantum dot. The sphere would rest in a circular cutout centered around the quantum dot. The sphere-sample distance would be determined by the radius of the sphere and the diameter of the cutout. The fiber stem that holds the sphere could be permanently fixed onto the sample.

The quantum dot could be pumped by shining laser light downwards onto the microsphere. The sphere would focus pump light onto the mesa. Light emitted by the quantum dot in appropriate directions would couple into a circulating WGM. Light could be efficiently coupled out of the sphere using an angle-polished optical fiber.
6.3. MICROSPHERE SINGLE-DOT LASER

Figure 6.2: Schematic of the proposed apparatus to measure lasing with a single quantum dot coupled to the whispering-gallery mode of a microsphere cavity
Light could then be guided with fibers out of the cryostat and into measurement equipment, such as a spectrometer or a single-photon detector. A schematic of the proposed experimental arrangement is shown in Fig. 6.2.

In order to get reasonable coupling between the quantum dot and the WGM, the sphere must be brought close to the semiconductor surface. This means that light will couple out of the WGM by frustrated total internal reflection, and the quality factor will be degraded. Nonetheless, relatively high $Q$’s of over $10^5$ are possible even for spheres in contact with a GaAs surface [167]. Another limitation of this setup is that the quantum dot is located outside of the microcavity, and thus couples only to the evanescent field of the mode. This limits the interaction between the dot dipole and the mode, making significant cavity-QED effects unlikely. On the other hand, it may still be possible to achieve single-dot lasing with this system [168]. Light travelling in a circle around the sphere would be amplified once per pass due to the interaction of the evanescent field with the excited quantum dot. The laser would thus be the microscopic analogue of evanescently coupled waveguide lasers [169, 170].

### 6.3.2 Laser Threshold

In order to determine whether it is possible to reach the laser threshold using a single quantum dot and a microsphere, we must determine the fraction $\beta$ of photons spontaneously emitted from the quantum dot that are captured by the microsphere WGM. Resonances in the microsphere can be described by three mode numbers: a resonance order $n$, an angular mode number $l$, and an azimuthal mode number $m$ [171]. WGM’s have $n = 0$ and $l \approx l_{max}$, where the maximum angular number $l_{max}$ is equal to the number of integral wavelengths that can fit around the circumference of the sphere. For each set of mode numbers, there is a transverse electric (TE) and transverse magnetic (TM) polarization mode. The field in one of these modes can be described by a scalar Hertz potential [172]. For example, in a TM mode, the radial electric field is given in terms of an electric potential $\Pi^{(e)}$:

$$E_r = \frac{\partial^2 (r\Pi^{(e)})}{\partial r^2} + k_{sph} r \Pi^{(e)}.$$  \hspace{1cm} (6.3)
In this equation, \( \mathbf{r} \) is the radial vector, and \( k_{\text{sph}} \) is the wavevector in the sphere (the free-space wavevector \( k_o \) multiplied by the refractive index \( n_{\text{sph}} \approx 1.45 \)). The WGM’s are products of spherical Bessel functions and spherical harmonics:

\[
\Pi_{lm}^{(c)} \propto j_l(k_{\text{sph}}^2 r)Y_{lm}(\theta, \phi) .
\] (6.4)

The degeneracy between modes with the same \( l \) and different \( m \) is lifted in real microspheres by a small ellipticity. We will thus consider coupling into a single TM mode with \( m = l = l_{\text{max}} \).

To determine the number of spontaneous photons in the WGM, we first calculate the capture fraction \( \beta \) using the generalization of Lorentz-Mie scattering theory due to Barton, Alexander, and Schaub [173]. The field inside the sphere is written as a sum of resonant modes. The electromagnetic radiation emitted from the dot and incident on the sphere is likewise written as a sum of orthogonal spherical modes, as is the field scattered off of the sphere. The coefficients of the three sums are related by application of boundary conditions. The result is that the amplitude \( c_o \) of the WGM in question is related only to the amplitude \( c_1 \) of the incident mode with the same mode numbers, as follows:

\[
c_o = \frac{\zeta'_l(k_o R)\psi_l(k_o R) - \zeta_l(k_o R)\psi'_l(k_o R)}{n_{\text{sph}}^2\psi'_l(k_{\text{sph}} R)\zeta'_l(k_{\text{sph}} R) - n_{\text{sph}}\zeta'_l(k_{\text{sph}} R)\zeta_l(k_o R)} c_1 .
\] (6.5)

In this equation, \( \psi_l \) and \( \zeta_l \) are the Ricatti-Bessel functions, a prime denotes the derivative, and \( R \) is the radius of the sphere. The coefficient \( c_1 \) is given by the overlap of the incident electric field \( \mathbf{E}^{\text{inc}} \) with the corresponding spherical harmonic over the surface of the sphere:

\[
c_1 = \frac{R^2}{l(l+1)j_l(k_o R)} \int_0^{2\pi} \int_0^\pi \sin \theta \, d\theta \, d\phi \, E_r^{\text{inc}}(R, \theta, \phi) \, Y_{l}^*(\theta, \phi) .
\] (6.6)

To determine the incident field, we follow the method of Lukosz and Kunz [174]. We assume that the electric dipole in the QD is randomly oriented, so that the emitted field is a spherical wave. The scalar potential inside the semiconductor sample is given...
by the following Fourier integral:

$$\Pi_o^{\text{inc}}(r) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dk_x dk_y \psi_o(k_x, k_y) \exp(i[k_x x + k_y y + k_{z,\text{sem}}(z - D)]) .$$

(6.7)

In this equation, the semiconductor-air interface is taken to be at $z = 0$, $D$ is the distance from the interface to the QD, $\psi_o$ represents a scalar spherical wave, and $k_{z,\text{sem}} = \sqrt{k_{\text{sem}}^2 - k_x^2 - k_y^2}$, where $k_{\text{sem}}$ is the wavenumber in the semiconductor. In other words, the emitted field is represented as a superposition of plane and evanescent waves, corresponding to real and imaginary values of $k_{z,\text{sem}}$. The transmission of each of these waves through the interface is given by the corresponding Fresnel coefficient.

To determine the extent of $Q$ degradation as the sphere approaches the semiconductor surface, we consider the reverse, equivalent problem of coupling a uniform, homogeneous background of plane waves from the semiconductor sample into the sphere. The capture probability for each of these waves is calculated by the modified Lorentz-Mie theory discussed above. By integrating over all plane waves, we get the total out-coupling probability $\delta_c$ per unit pass of the electromagnetic field in the WGM. The quality factor simply follows from this number and the round-trip time $T_{rt}$ in the cavity:

$$Q \approx \frac{T_{rt}}{|\delta_c|^2} = \frac{2\pi R \omega}{c |\delta_c|^2} .$$

(6.8)

The calculated quality factor and coupling coefficient can be combined to give the average number of spontaneously-emitted photons $n_{sp}$ stored in the WGM of the microsphere. We assume that the quantum dot emits one photon every 650 ns. We also assume the quantum dot is located 0.1 $\mu$m below the semiconductor surface, and that the WGM has $Q = 5 \cdot 10^8$ when the microsphere is far from the sample. Calculated results for different sphere-sample distances $d$ are shown in Fig. 6.3. At large distances, $n_{sp}$ drops off as the distance from the sample increases and the coupling of light from the dot into the WGM decreases. At short distances, on the other hand, $n_{sp}$ drops exponentially as the sphere approaches the surface because of $Q$ degradation. The rate of $Q$ degradation is nearly independent of sphere radius, but the distance at which the degradation begins is smaller for larger $R$, due to the longer round-trip time in the larger spheres. The quantum laser threshold, given by
the condition that $n_{sp} \geq 1$, is surpassed over a large range of sphere-sample distances for larger spheres. Laser oscillation should thus be possible in this microsphere laser with a single quantum dot [175].

Detailed calculations of laser characteristics for this system were performed using a master-equation model [176]. For a quantum dot pumped electrically with unity quantum efficiency, a laser threshold current of about 22 pA was calculated. This is several orders of magnitude lower than any reported threshold current for a semiconductor laser. Novel effects, such as self-quenching [177], were predicted.

6.4 Entangled Photon-Pair Generation

Finally, we suggest that it may be possible to use single quantum dots to generate polarization-entangled photon pairs [178]. The technique would be nearly identical to
that used to generate single photons. However, rather than spectrally isolating only the single-exciton peak, both the exciton and biexciton lines would be collected. The two photons collected for each pump pulse should have anticorrelated polarizations, if radiative recombination in the quantum dot follows the same selection rules as for quantum wells in III/V semiconductors (see Fig. 6.4) [41]. The selection rules permit a spin-up electron \((J_z = +1/2)\) to recombine only with a spin-up heavy hole \((J_z = +3/2)\), emitting a left-circularly polarized photon. Likewise, a spin-down electron \((J_z = -1/2)\) can recombine only with a spin-down heavy hole \((J_z = +3/2)\), emitting a right-circularly polarized photon. The selection rules should therefore translate the anticorrelation of electron and hole spins in the biexcitonic state into a photon polarization anticorrelation.

The generation of entangled photons, starting from the biexcitonic state, is similar to two-photon cascade decay in an atom [179, 180]. One of the two excitons recombines first and emits a left- or right-circularly polarized photon. The remaining exciton, with opposite carrier spins, then recombines and emits a photon of opposite polarization. If the two decay paths are indistinguishable, the two photons are in a maximally entangled (Bell) state:

\[
|\psi\rangle = \frac{1}{\sqrt{2}}(|\sigma^+\rangle_1|\sigma^-\rangle_2 + |\sigma^-\rangle_1|\sigma^+\rangle_2).
\] (6.9)
This simple picture must be modified to include band mixing due to carrier confinement in the quantum dot. In particular, the heavy-hole ground state is likely to have contributions from the $J_z = \pm 1/2$ light-hole states. Possible transitions to these states are indicated by dashed arrows in Fig. 6.4. The two photons that arise from the decay of the biexcitonic state would then no longer be perfectly anticorrelated with respect to left- and right-circular polarization. Asymmetry in the shape of the dot, strain, and piezoelectric fields would further reduce the anticorrelation [181], as would spin interactions in the dot [182].

Also excluded from the simple proposal are dephasing processes that can degrade the entanglement. For example, spin dephasing may occur between the photon emission events, while the quantum dot is in the single-exciton state. If the dephasing rate is large compared to the radiative recombination rate, the final photon state is a statistical mixture of anti-correlated photons, rather than an entangled state.

In fact, our measurements have shown strong classical polarization correlation between single-exciton and biexciton photons, but no quantum-mechanical entanglement [183]. That is, the photons are strongly correlated in a particular linear basis, but not in other, non-orthogonal bases. The apparatus used to measure two-photon correlation is the same as that described in Section 5.2 and shown in Fig. 5.4. Rotation of the half-wave plates in each arm of the Hanbury Brown and Twiss-type apparatus allows different linear polarizations to be measured, while a single quarter-wave plate can be inserted after the collection lens in order to measure circular polarizations. The monochrometers in front of the photon counters are set such that one detector measures photons from the biexciton line, while the other detector measures photons from the single-exciton line. By comparing the area of the central peak in the measured histogram of delay times to the areas of distant side peaks, the exciton-biexciton coincidence rate can be determined for a chosen pair of measurement polarizations.

The quantum dot investigated has a strong polarization anisotropy. For a particular linear polarization (designated horizontal, or “H”), the photon count rate is nearly double that for the orthogonal polarization (designated vertical, or “V”). A strong polarization correlation was measured in the H / V basis: the coincidence rate
was high when the measurement polarizations were both H or both V, and was low when one was H and the other was V. Repeating the measurement using a polarization basis rotated by 45°, however, resulted in virtually no correlation, reflecting the absence of polarization entanglement.

This can be made more explicit by measuring coincidence rates for a complete set of polarization combinations [184]. From the results of all these measurements, the complete two-photon density matrix can be deduced, using the methods of quantum state tomography. The measured matrix is represented in Fig. 6.5. The on-diagonal components display the classical polarization correlation, while the small off-diagonal components display the lack of entanglement.

In order to explain this result, we allow the biexciton to decay along two different paths: one where both photons are emitted with $\pi_x$ polarization (detected as H), and the other where both photons are emitted with $\pi_y$ polarization (detected as V) [185]. This would be consistent with energy splitting between one-exciton states with different spins, due to the electron-hole exchange interaction in an asymmetric
quantum dot. We assume that the two decay paths occur with equal probability, and we include a probability $\epsilon$ that the second photon has a polarization opposite to that of the first photon. This factor can account for spin flipping in the one-exciton state, as well as the possibility of non-ideal selection rules. Our model is capable of quantitatively reproducing the observed results. We note that, in this model, the large polarization anisotropy of the quantum dot is due to the unequal collection efficiencies of $\pi_x$ and $\pi_y$ photons. A model based on preferential decay through the $\pi_x$ path cannot simultaneously predict the observed density matrix and net polarization visibilities. We also note that, using this model, we can place a lower limit of at least 2.2 ns for the mean spin-flip time in the one-exciton state, consistent with the observations of other groups [186].

To get polarization-entangled electrons from a quantum dot, we would need to remove the spin splitting in the single-exciton state. This could possibly be done by optimizing the growth conditions so that the dots are more symmetric, or so that the dots are larger and the effect of asymmetry is minimized.
Chapter 7

Conclusions

The work reported in this thesis brings together semiconductor optoelectronics and quantum optics by focussing on single things. We have isolated single InAs / GaAs quantum dots from a self-assembled ensemble, and have observed luminescence from single excitons in the dots. We have coupled the dots to single three-dimensionally confined modes in micropost microcavities. Theoretical modelling of the microcavity modes has allowed us to improve our fabrication process, to the point where 83% of the light emitted by the single excitons is coupled to the single cavity mode. We have used pulsed excitation and spectral filtering to turn the quantum dot into a source of regulated single photons, and have seen 38% of these photons emitted into a single travelling-wave mode.

This work has been motivated by the conviction that there is a sudden demand for single photons. A practical source of single photons would be useful to experimentalists in the rapidly maturing field of quantum key distribution. As well, new applications for single photons continue to be invented. For example, a much-discussed recent proposal for experimental quantum computation involves single-photon sources, photon-number detectors, and linear optical elements [187]. Single photons may also be useful in the generation of truly random numbers [188, 189]. We are thus hopeful that our device will see continued development and significant application in the future.
Appendix A

Microcavity Design and Process in Detail

Planar microcavities containing quantum dots were grown by MBE. The dots were deposited under conditions optimized for low density, as described in Section 2.2. The deposition was done on the (001) surface of a semi-insulating GaAs substrate, which was rotated at 25 rpm during growth. The growth rate of InAs was 0.053 $\mu$m/hr, of GaAs was 0.432 $\mu$m/hr, and of AlAs was 0.425 $\mu$m/hr. The V/III ratio was 50 for InAs, 12 for GaAs, and 19 for AlAs. Table A.1 summarizes the stages in the growth process.

Following the MBE growth, microposts are etched into the sample, using electron-beam lithography and ECR dry etching, as outlined above. Table A.2 details the process steps involved. Fig. A.1 shows what part of the etched sample looks like under an optical microscope. 10 $\times$ 10 arrays of posts are etched, with the post diameter varying from array to array. Around each array is a large box, which allows for alignment during optical measurements. Arrows on the inside of the boxes point along the rows and columns of the arrays. The center-to-center spacing of posts in all arrays is 50 $\mu$m.
Table A.1: Stages in molecular-beam epitaxy of a microcavity containing quantum dots

<table>
<thead>
<tr>
<th>Step</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Desorb oxide from substrate at 625 °C</td>
</tr>
<tr>
<td>2</td>
<td>Set substrate temperature to 575 °C</td>
</tr>
<tr>
<td>3</td>
<td>Deposit 3000Å GaAs buffer layer</td>
</tr>
<tr>
<td>4</td>
<td>Deposit quarter-wavelength AlAs layer</td>
</tr>
<tr>
<td>5</td>
<td>5 sec. interrupt</td>
</tr>
<tr>
<td>6</td>
<td>Deposit quarter-wavelength GaAs layer</td>
</tr>
<tr>
<td>7</td>
<td>5 sec. interrupt</td>
</tr>
<tr>
<td>8</td>
<td>Repeat steps 3 - 6 for total of 30 mirror pairs</td>
</tr>
<tr>
<td>9</td>
<td>Deposit quarter-wavelength AlAs layer</td>
</tr>
<tr>
<td>10</td>
<td>5 sec. interrupt</td>
</tr>
<tr>
<td>11</td>
<td>Deposit GaAs layer equal to half wavelength minus 100Å</td>
</tr>
<tr>
<td>12</td>
<td>Lower substrate temperature to 530 °C and wait 10 min.</td>
</tr>
<tr>
<td>13</td>
<td>Deposit 100Å GaAs</td>
</tr>
<tr>
<td>14</td>
<td>5 sec. interrupt</td>
</tr>
<tr>
<td>15</td>
<td>Deposit 1.75 ML InAs</td>
</tr>
<tr>
<td>16</td>
<td>3 sec. interrupt</td>
</tr>
<tr>
<td>17</td>
<td>Deposit 400Å GaAs</td>
</tr>
<tr>
<td>18</td>
<td>Raise substrate temperature to 575 °C and wait 10 min.</td>
</tr>
<tr>
<td>19</td>
<td>Deposit GaAs layer equal to half wavelength minus 400 Å</td>
</tr>
<tr>
<td>20</td>
<td>Deposit quarter-wavelength AlAs layer</td>
</tr>
<tr>
<td>21</td>
<td>5 sec. interrupt</td>
</tr>
<tr>
<td>22</td>
<td>Deposit quarter-wavelength GaAs layer</td>
</tr>
<tr>
<td>23</td>
<td>5 sec. interrupt</td>
</tr>
<tr>
<td>24</td>
<td>Repeat steps 19 - 22 for total of 12 mirror pairs</td>
</tr>
</tbody>
</table>
Table A.2: Stages in the process used to make micropost microcavities

<table>
<thead>
<tr>
<th>Process Step</th>
<th>Details</th>
</tr>
</thead>
</table>
| Three-Solvent Clean | Soak 10 min. in heated degreaser (leksol)  
Three-Solvent Clean | Soak 10 min. in unheated acetone  
Soak 10 min. in heated isopropanol  
Rinse in deionized water and blow dry with clean nitrogen  
Bake 30 min. in oven at 120 °C |
| Resist Coating    | Spin MMA(8.5)MAA 13% in ethyl lactate at 3000 rpm for 40 sec.  
Bake 20 min. on hot plate at 180 °C  
Spin PMMA 950k 2% in chlorobenzene at 3000 rpm for 40 sec.  
Bake 20 min. on hot plate at 180 °C |
| E-Beam Lithography| Exposure on Leica Stereoscan 440  
Area dose = 500 μC/cm²  
Accelerating voltage = 25 kV  
Beam current = 200 pA (50 nA for large features) |
| Develop           | Develop 75 sec. in Isopropanol:MIBK 1:1  
Rinse in deionized water and blow dry with clean nitrogen  
Inspect under optical microscope and develop more if necessary |
| Liftoff           | E-beam evaporate 800 Å Ni  
Soak in acetone at least 1 hr.  
Spray with acetone to lift off metal  
Rinse in isopropanol and blow dry with clean nitrogen  
Inspect under optical microscope |
| ECR Plasma Etch   | Etch in PlasmaQuest  
Etch gases: Ar:BCl₃:Cl₂ 15:10:1  
Microwave power: 400 W  
Cool chuck as low as possible, allowing 1 hr. for temperature to stabilize  
Run dummy wafer through first  
Mount sample on doped Si wafer using copper tape  
Recipe steps:  
(1) Flow backside He at 10 Torr for 150 sec. to cool down  
(2) Start process gases flowing, with pressure = 2 mTorr  
Allow 45 sec. to stabilize  
(3) Turn on ECR power and turn on RF power to 47 W  
Etch for 600 sec.  
(4) Reduce pressure to 1 mTorr and reduce flow rates by 1/2  
Increase RF power to 50 W  
Retune microwave stubs  
Etch for 600 sec.  
(5) Reduce pressure to 0.5 mTorr and reduce flow rates by 1/2  
Stop Cl₂ flow  
Increase RF power to 52 W  
Retune microwave stubs  
Etch for 600 sec. |
Figure A.1: Optical-microscope image of a portion of the etched microcavity sample
Bibliography


[185] V. D. Kulakovskii, G. Bacher, R. Weigand, T. Kümmell, A. Forchel, E. Borovitskaya, K. Leonardi, and D. Hommel. Fine structure of biexciton emission in


