CAVITY QUANTUM ELECTRODYNAMICS WITH QUANTUM DOT - PHOTONIC CRYSTAL NANOCAVITIES

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SIGNED: Joshua R. Hendrickson
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DEDICATION

In loving memory of my grandfather, Raymond Pennington
# TABLE OF CONTENTS

LIST OF FIGURES ........................................................................................................... 8

ABSTRACT ...................................................................................................................... 10

CHAPTER 1: INTRODUCTION ................................................................................... 12

1.1 Explanation of the problems and a review of the literature ........................................ 12

1.2 Explanation of the dissertation format ..................................................................... 26

CHAPTER 2: PRESENT STUDY .................................................................................. 31

2.1 Quantum dot photonic crystal slab nanocavities ..................................................... 31

2.1.1 Strong coupling ............................................................................................... 31

2.1.2 Spectral cavity shifting through inert gas condensation ................................... 35

2.1.3 Gain - absorption effects and lasing .................................................................. 38

2.2 Improvements in the growth, fabrication, and modelling of quantum dot photonic crystal slab nanocavities ................................................................. 41

2.2.1 GaAs photonic crystal slab nanocavities: Growth, fabrication, and photon storage time .................................................................................................. 41

2.2.2 Modelling and fabrication of GaAs/InAs photonic-crystal cavities for cavity Electrodynamics ........................................................................................................ 43

2.3 Deterministic growth of quantum dots ................................................................. 46

2.4 Silicon photonic crystal nanobeam cavities ............................................................ 48

2.5 Summary of accomplishments ............................................................................... 52

2.6 Future directions ................................................................................................... 54

REFERENCES ................................................................................................................. 56
TABLE OF CONTENTS – continued

APPENDIX A: Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity ..................................................................................................................61

APPENDIX B: Scanning a photonic crystal slab nanocavity by condensation of xenon ..................................................................................................................................................66

APPENDIX C: Quantum dot photonic-crystal-slab nanocavities: Quality factors and lasing ..........................................................................................................................70

APPENDIX D: GaAs photonic crystal slab nanocavities: Growth, fabrication, and photon storage time .................................................................................................................75

APPENDIX E: Modelling and fabrication of GaAs/InAs photonic-crystal cavities for cavity electrodynamics .........................................................................................................82

APPENDIX F: In-situ annealing of InAs quantum dots on pre-structured GaAs substrates .................................................................................................................................90

APPENDIX G: Characterization of 1D photonic crystal nanobeam cavities using curved microfiber .................................................................................................................101
LIST OF FIGURES

1. Figure 1: Energy diagram for a strongly coupled system (solid lines) and for an uncoupled system (dotted line). As the detuning between the atomic and cavity frequencies approaches zero the strongly coupled system energies repel one another as opposed to the uncoupled system energies which become degenerate ..............................................................................................................14

2. Figure 2: The Jaynes Cummings ladder of energy states. The solid lines are eigenenergies of a strongly coupled system while the dashed lines indicate energy levels for an uncoupled system. A factor of $1/\sqrt{2}$ has been omitted from the eigenstates for clarity. ............................................................................................16

3. Figure 3: AFM image of quantum dots.........................................................................................................................19

4. Figure 4: Quantum dot – photonic crystal slab nanocavity .................................................................20

5. Figure 5: Computed electric field profile....................................................................................................................22

6. Figure 6: SEM image of a 1D silicon photonic crystal nanobeam cavity .................25

7. Figure 7: Schematic of experimental setup..............................................................................................................32

8. Figure 8: The two coupled-system peaks are plotted as a function of temperature and compared with the scan rates of an uncoupled QD and an empty cavity peak. ...........................................................................................................34
LIST OF FIGURES - continued

9. Figure 9: Strong coupling anti-crossing curves using xenon condensation to scan the cavity mode. The three peak positions are plotted versus cycle number, and anti-crossings are observed between the cavity mode and two distinct quantum dots. The dashed line shows the cavity mode spectra at high excitation power where coupling effects are not evident .................................................................37

10. Figure 10: Crossed polarized resonant scattering setup ........................................52
ABSTRACT

High quality factor, small mode volume photonic crystal cavities and single emitter quantum dots are the topic of this dissertation. They are studied as both a combined system with InAs quantum dots grown in the center of a 2D GaAs photonic crystal slab nanocavity as well as individually. The individual studies are concerned with passive 1D silicon photonic crystal nanobeam cavities and deterministic, site-selectively grown arrays of InAs quantum dots.

For the combined system, strong light matter coupling in a quantum dot photonic crystal slab nanocavity is discussed. Vacuum Rabi splitting is seen when the interaction strength exceeds the dissipative processes of the coupled system. In order to increase the probability of a spectral matching between cavity modes and quantum dot transitions, a technique for condensing an inert gas onto a sample is used. This can lead to a spectral tuning of up to 4 nm of the cavity mode with minimal change in the cavity quality factor while maintaining cryogenic temperatures down to 4 K. The effect of a large density of quantum dots within a quantum dot photonic crystal slab nanocavity is also addressed. Gain and absorption effects are found to occur, changing the cavity emission linewidth from that of its intrinsic value, as well as lasing with a low number of quantum dots and with high spontaneous emission coupling factors. Additionally, methods for improving the quality factor of GaAs photonic crystal cavities and better understanding different loss mechanisms are discussed.
In the individual studies, the site-selective growth of InAs quantum dots on pre-structured GaAs wafers is shown as a promising method for the eventual deterministic fabrication of photonic crystal cavities to single quantum dots. An in-situ annealing step is used to reduce quantum dot density, helping ensure that dots are not grown in unwanted locations.

Given silicon’s potential for achieving higher quality factors than its GaAs counterpart, a study of 1D passive silicon photonic crystal nanobeam cavities is carried out. Transmission through a coupled microfiber is used to measure quality factors of the cavities and compared with that of a crossed polarized resonant scattering measurement.
CHAPTER 1: INTRODUCTION

1. Explanation of the problems and a review of the literature

It is well known in the field of cavity quantum electrodynamics (CQED) that the properties of an emitter can be altered by tailoring the environment in which it is located [1]. As early as 1946 E. M. Purcell had published his article [2] describing the change in the spontaneous emission rate of a dipole when resonantly coupled to a cavity mode. This work was later extended by Jaynes and Cummings, who built up a model to describe the interaction of a two level atom with a single mode of the quantized electromagnetic field [3, 4]. In order to experimentally create such single modes of the field special cavities must be used, which led to the birth of experimental CQED.

Using a semiclassical theory, the interaction between an atom and a classical field is described in the dipole approximation by the Hamiltonian

\[ V = \mu \cdot E \]

where \( \mu \) is the dipole moment and \( E \) is the electric field. Incorporating a fully quantum picture the electric field then becomes an operator and the new interaction Hamiltonian is

\[ V = \hbar (a + a^\dagger)(g \sigma_+ + g^* \sigma_-) \]

In this equation \( a^\dagger \) and \( a \) are the field creation and annihilation operators, respectively, and create or remove an excitation in the electromagnetic field. The Pauli spin matrices \( \sigma_+ \) and \( \sigma_- \) correspond to transitions in a two level atom, \( \sigma_+ \) changing the excitation
from the ground state \( g_{nd} \) to the excited state \( g_{exc} \), and vice versa for \( \sigma_- \). The strength of the interaction is given by \( g \) which is directly proportional to the dipole matrix element and the amplitude of the electromagnetic field \( g = \frac{\mu \cdot E}{\hbar} \). Within the rotating wave approximation the full Hamiltonian of a two level atom interacting with a single mode of the quantized electromagnetic field can be expressed as

\[
H = H_0 + V = \hbar \omega \sigma_+ \sigma_- + (a^\dagger a + \frac{1}{2})h\Omega + hg(a\sigma_+ + a^\dagger \sigma_-)
\]

with \( \omega \) the atomic transition frequency and \( \Omega \) the field frequency. This equation is known as the Jaynes-Cummings Hamiltonian. Upon inspection of the interaction part of the Jaynes-Cummings Hamiltonian, one sees that coupling is only possible between states such as \( |exc, n\rangle \) and \( |gnd, n+1\rangle \). However, these states, called the bare states, are not the eigenstates of this Hamiltonian. Known as the dressed states, the eigenstates of this system are entanglements of the bare states and, at zero detuning, are

\[
|2, n\rangle = \left(|exc, n\rangle - |gnd, n+1\rangle\right)/\sqrt{2}
\]

\[
|1, n\rangle = \left(|exc, n\rangle + |gnd, n+1\rangle\right)/\sqrt{2}
\]

with eigenvalues of

\[
E_{2,n} = \hbar(n + \frac{3}{2})\Omega - hg\sqrt{n+1}
\]

\[
E_{1,n} = \hbar(n + \frac{3}{2})\Omega + hg\sqrt{n+1}
\]
The difference between these eigenvalues, $E_{1,n} - E_{2,n}$, is $2\hbar g \sqrt{n + 1}$ which can be rewritten as $\hbar R_0$, or Planck’s constant times the quantized Rabi frequency $R_0 = 2g \sqrt{n + 1}$. Figure 1 shows a plot of the energies of the coupled system as a function of the atomic transition frequency. A signature of a strongly coupled system is evident in this diagram by the characteristic anti-crossing behavior; as the detuning approaches zero the energies of the eigenstates repel one another and the energy spectrum splits, unlike the uncoupled atom and field states which become degenerate.

Figure 1: Energy diagram for a strongly coupled system (solid lines) and for an uncoupled system (dotted line). As the detuning between the atomic and cavity frequencies approaches zero the strongly coupled system energies repel one another as opposed to the uncoupled system energies which become degenerate.
It is interesting to investigate what happens with this system in the absence of any applied electromagnetic field. In the semiclassical approach the interaction energy would go to zero, however, in a fully quantum picture there is always some electromagnetic field intensity, even in the vacuum state. Theoretically creating a cavity of volume $V$, integrating the energy density within that cavity, and equating that energy to what we would find by modelling the field as a quantized harmonic oscillator we can determine the strength of the vacuum electric field

$$E_{\text{vac}} = \sqrt{\frac{\hbar \omega}{2 \epsilon_0 V}}$$

In free space where $V = \infty$ the vacuum electric field is negligibly small but as the volume decreases the field increases. This implies that one could create strong light matter interactions between a two level emitter and the vacuum field provided the volume of the cavity is sufficiently small. Since the splitting between the eigenenergies is directly proportional to the electric field, smaller volume cavities will lead to larger splittings. This energy splitting when the photon number is zero is called vacuum Rabi splitting. Figure 2 shows the first few rungs of the Jaynes Cummings ladder of energy states beginning with the unexcited atom, zero photon state.

In any real system there are always dissipative processes that one must take into account. For example, in the case of an emitter coupled to a cavity, there is the photon loss rate out of the cavity $\kappa$ and the rate of emission of the emitter into modes other than the cavity mode $\gamma$. The photon loss rate is related to the quality factor ($Q$) of the cavity which is a
measure of the amount of time energy remains inside the cavity. The atomic transition will now have a linewidth associated with it as well as the cavity mode frequency. If these linewidths are large compared to the energy splitting then the two energy peaks at zero detuning would not be resolvable. Due to the strong interactions, the linewidths of each of the two peaks are equal to the mean of the atomic and cavity linewidths. Therefore, to resolve the splitting one needs the interaction strength to be larger than the mean of the dissipative processes, or $2g \gtrsim \frac{\kappa + \gamma}{2}$. For the results provided in this dissertation the cavity loss rate well exceeds the emitter dephasing rate so the strong coupling requirement now depends on a maximization of the ratio of $\frac{g}{\kappa}$. Since

$$g \propto E_{\text{vac}} \propto \frac{1}{\sqrt{V}} \quad \text{and} \quad \kappa = \frac{\omega}{O}$$

the requirement can also be seen as maximizing $\frac{O}{\sqrt{V}}$; i.e. high quality factor, small mode volume cavities are necessary for achieving vacuum Rabi splitting.
Much of the original work in CQED in general and vacuum Rabi splitting in particular was accomplished by the atomic physics community [5]. The experimental results began in the microwave regime with Rydberg atoms passing through a Fabry-Perot cavity composed of superconducting mirrors [6]. The optical region was then accessed with the use of cesium atoms, also passing through a Fabry-Perot type cavity. Early work involved many atoms within the cavity [7], followed by experiments involving one intracavity atom at a time [8], and then progressing to the point of trapping individual atoms inside a cavity [9, 10]. At first statistics were built up by averaging over a number of different experimental runs until trapping techniques evolved to the point that holding times could be in the millisecond range, allowing for a full spectrum of data to be collected for one, single atom [11].

While the trapping time of atoms may be long compared to the length of time an experiment is run and superconducting mirrors can form cavities with ultrahigh $Q$’s, there was still a strong desire to move this field over into the realm of solid state physics. Entire optical tables are necessary to create atomic CQED systems whereas their solid state analogs can be made on semiconductor substrates with footprints on the order to microns. Plus, atoms move about while solid state emitters can remain in place indefinitely. The only question that one must ask is what do we choose for the emitter and what do we choose for the cavity [12]?
The obvious choice of emitter when solid state CQED began was the semiconductor quantum dot [13, 14, 15, 16]. While initially interface fluctuation quantum dots were thought to have superior properties eventually it was the self organized quantum dot which emerged as the top candidate. A quantum dot is composed of many atoms but is able to effectively act as a single two level emitter. In fact, single quantum dots have been shown to exhibit a number of atomic like properties such as photon antibunching [17, 18, 19], resonance fluorescence [20], Autler-Townes splitting and Mollow absorption spectra [21]. Quantum dots are often grown using molecular beam epitaxy (MBE) and utilizing the Stranski-Krastanov (SK) growth mode. Essentially a thin layer of material such as indium arsenide (InAs) is grown on a substrate such as gallium arsenide (GaAs). Under the right growth conditions, the lattice mismatch between the two materials will cause the quantum dot material to clump together into small pyramidal or spherical like structures, usually of 30-70 nm in diameter and 5-10 nm in height. An atomic force microscope (AFM) image of a single layer of quantum dots is shown in Figure 3. In the simplest approximation one can think of this as a 3D spherical potential well which, upon solving the Schrödinger equation, will lead to discrete energy levels. Of course, there are other choices of emitter such as colloidal quantum dots and nitrogen vacancy centers in diamond, however, semiconductor quantum dots have remained the most promising “artificial atom”.

Next, one must decide what type of solid state cavity to use. The choices include such systems as micropillars, microtoriods, microspheres, and microdisks but the one that
seems most promising, and that was used in the experiments described later in this dissertation, is the photonic crystal cavity [22, 23, 24, 25, 26, 27]. Starting with the notion of a photonic crystal [28], which is basically an extension of the well know one dimensional Bragg mirror into higher dimensions, a photonic crystal cavity can be made through the introduction of intentional defects. For the 2D photonic crystal slab nanocavity light confinement is achieved within the plane of the slab by Bragg reflection. This is achieved by etching a lattice of air holes into the slab to act as a periodic change in the index of refraction, thereby forming a photonic bandgap. A defect is then introduced into the lattice to allow for light localization within the bandgap; i.e. a cavity. Cavities can be created by leaving out one or more air holes or changing the size of particular air holes. By making alterations such as shifting the nearest neighbor air holes
around the defect region, the structures can be optimized [29, 30]. Changes in this shift along with hole radius, lattice constant, and slab thickness allow for high quality factor, small mode volume solid state cavities to be created at given wavelengths. Currently, fabrication tolerances have limited most photonic crystal cavities to the near IR region. The in-plane confinement occurs from total internal reflection which can be relatively good as the index of refraction of GaAs is in the range of 3.4.

Incorporating quantum dots into photonic crystal cavities is relatively straightforward. A substrate such as GaAs is placed into the MBE chamber where one first grows a buffer layer, followed by an aluminum gallium arsenide (AlGaAs) sacrificial layer, the bottom half of the photonic crystal slab, InAs quantum dots, and finally the top half of the slab. A mask of the photonic crystal pattern is then created using electron beam lithography, dry etching with sources such as chemically assisted ion beam etching for creating the air holes, and wet etching with hydrogen fluoride for sacrificial layer removal.; see Figure 4.

Figure 4: Quantum dot – photonic crystal slab nanocavity.
Now that we have our solid state CQED system, how do we reach the strong coupling regime? A properly designed and fabricated GaAs photonic crystal cavity can have mode volumes in the $\left(\frac{\lambda^3}{n}\right)$ range and quality factors exceeding 20,000 which are sufficient to achieve strong light matter coupling. There are, however, some additional requirements that can be harder to control such as spectral and spatial matching of the quantum dot and the cavity mode.

In order to meet the spectral requirement the quantum dot transition frequency must be close to that of the cavity mode. An ensemble of quantum dots typically has a ground state inhomogeneous linewidth of 50 – 100 nm, therefore, there is a certain amount of luck involved in minimizing the detuning. If one is fortunate enough to find a quantum dot spectral line within a few angstroms of the cavity mode and blue detuned then temperature tuning can be used to scan the quantum dot through the cavity. Quantum dots can suffer from large phonon broadening which is why samples are held at cryogenic temperatures. This limits the amount of tuning of the quantum dot to roughly 0.4 nm. The larger temperatures necessary for greater shifts increase $\gamma$ so much that the strong coupling regime is no longer obtainable. Alternatives to temperature tuning include techniques such as digital etching of the photonic crystal [31] or, as discussed in more detail in the following chapter, condensation of an inert gas onto the sample [32]. Gas condensation is a reversible process where a controlled amount of xenon or nitrogen gas is introduced to the cryogenically cooled sample, adhering to its surface and thus
modifying the photonic crystal parameters through increased slab thickness and decreased hole radii. This process can tune the cavity mode upwards of 5 nm with negligible changes in quality factor.

Another requirement is that the quantum dot should be located in the antinode of the electric field within the cavity; see Figure 5. Recalling that the interaction strength is equal to the dipole moment times the electric field, spatial positioning can make a big difference.

![Computed electric field profile](Image courtesy of T. Yoshie).

The SK growth mode for quantum dots results in a random spatial distribution which can make meeting the spatial requirement difficult. Several deterministic techniques have been implemented over the years and include locating individual dots with an AFM through perturbations they create on the sample surface [33], in-situ lithography [34], and
photoluminescence measurements of quantum dots as measured from highly reflective structures fabricated on the sample [35]. Each of these methods has produced positive results, however, they are not very functional if one wishes to create a network of coupled cavities. One method to address this is to create a deterministic array of site-selectively grown quantum dots which could then have photonic crystal cavities fabricated around them. The literature contains a number of techniques [36] but here I will only discuss the one relevant to this dissertation – lithographic prestructuring of substrates [37]. Beginning with a GaAs substrate, electron beam lithography and wet chemical etching are used to create an array of nanoholes roughly 50 nm in diameter and 30 nm in depth. During the MBE quantum dot growth these nanoholes act as nucleation sites where the dots preferentially form. Using in-situ annealing one can increase the probability of dot nucleation at each site and minimize nucleation in between the sites. While the optical quality is still less than that of randomly grown quantum dots, there is still much optimism that site-selective techniques will be the enabler of future networked solid state CQED systems.

Assuming one doesn’t utilize the above techniques for deterministic placement of quantum dots into cavities, it might be reasonable to suggest one use a high density of quantum dots, thereby increasing the probability of spectral and spatial matching. Unfortunately this can be detrimental to the search for strong coupling [38]. With a high density there is a large number of surrounding uncoupled quantum dots that can give rise to absorption and lower the quality factor of the cavity. On the other hand, at higher
powers one can get gain and even lasing. Lasing within these systems is quite interesting in that the number of contributing quantum dots can be very low and the spontaneous emission coupling factors can approach unity [39, 40].

Even though GaAs photonic crystals have been the workhorse of solid state CQED up until now their quality factors are more or less just above the necessary values. Theoretical simulations give results with often more then two times the quality factor as compared to some of the highest experimentally measured values. Additionally, silicon has shown to give much higher quality factors then GaAs. This has led to a desire to better understand why GaAs cavities appear to be inferior and what can be done to improve upon them [41, 42]. Sample cleanliness can be a problem that degrades the structures as well as non-uniformity due to poor lithography and non-verticality of air holes from the dry etching process. Also, if the MBE growth conditions are not properly optimized surface roughness can occur on the 2D slab. It is because of these strict demands on the growth and fabrication that many research groups in this field have not been able to reach the strong coupling regime of solid state CQED.

New designs, such as 1D photonic crystal nanobeam cavities [43, 44, 45], are actively being pursued with even higher quality factors then the 2D slab designs. These nanobeams confine the light by a photonic crystal hole array in one dimension (Bragg reflection) and total internal reflection in the other two dimensions; see Figure 6. Making such nanobeams out of silicon has resulted in quality factors upwards of 750,000 [46].
These silicon structures are mostly passive as currently there are no good quasi two level emitters that can be implemented into them. Also, being passive means that new methods besides photoluminescence must be used to measure the cavity quality factor. Such methods include fiber taper coupling [47, 48] and crossed polarized resonant scattering [49, 50].

Figure 6: SEM image of a 1D silicon photonic crystal nanobeam cavity (Image courtesy of A. Homyk).

The field of solid state CQED has seen tremendous progress in the past decade. It took over twenty five years for the atomic community to go from the many atom perturbative regime to single (one and the same) atom vacuum Rabi splitting while the same feat was accomplished in the solid state community in half that time. With improvements in cavity designs, quantum dot growth, choice of materials, fabrication techniques, and
experimental understanding of these complex systems the future of solid state CQED seems bright.

2. Explanation of the dissertation format

This dissertation is mainly composed of the seven peer reviewed and published journal articles which have been appended to this body of work. As a selection of the articles that I have co-authored over my tenure as a graduate student, these specific ones were chosen for their relevance in building a coherent story of the study of quantum dots, photonic crystals, and their interactions. I had a primary role, either as overall lead or as lead of the Tucson group, in many of the publications and made important contributions to the rest.

The first article “Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity” (Appendix A) discusses strong light matter coupling between a single InAs quantum dot and a high $Q$ mode of a GaAs photonic crystal slab nanocavity. My contribution to this work was in the taking of all of the optical spectroscopic measurements. The experimental apparatus was initially constructed by G. Rupper and the theoretical framework was provided by G. Khitrova, H. M. Gibbs, and C. Ell. The quantum dots were grown by the group of D. G. Deppe with the help of O. B. Shchekin and the photonic crystal fabrication was performed by T. Yoshie in the group of
A. Scherer. Feedback and experimental guidance for improving the fabrication was provided by G. Khitrova and H. M. Gibbs.

In the second manuscript (Appendix B) a technique for spectrally scanning a photonic crystal cavity mode by the condensation of an inert gas such as xenon or nitrogen is described. S. Mosor took the lead in the construction of the experimental apparatus along with the help of B. C. Richards and me. Improvements in the vacuum system for lower base pressure, better reproducibility, and faster pump down times were accomplished by me, B. C. Richards, and S. Mosor. The same three researchers participated in the spectroscopic analysis showing the cavity mode shift as a function of the discrete introduction of an inert gas cycled into a cryostat containing a photonic crystal cavity. I was personally responsible for the optical measurements showing strong coupling of a photonic crystal cavity mode with two different single quantum dots. Quantum dot growth and photonic crystal fabrication were again performed by those from the above publication.

In order to better understand the effect of an ensemble of quantum dots on the optical properties of a quantum dot – photonic crystal slab nanocavity, an extensive study was performed and published in the article “Quantum dot photonic-crystal-slab nanocavities: Quality factors and lasing” (Appendix C). In this article it was shown that a large density of quantum dots can give rise to both gain and absorption. This is manifest in the change of the linewidth a cavity mode, it becoming either larger or smaller depending upon the
pump power of the sample. A tradeoff is found in that while higher quantum dot densities can make the search for strong coupling easier it also increases absorption which leads to a reduced $Q$. Lasing is seen with a small number of ($<15$) of quantum dots contributing to the lasing mode and exhibiting a high spontaneous emission coupling factor. I took the lead in the optical experimentation with help from fellow group members. Once again the quantum dot growth and photonic crystal fabrication was performed by those as in the previous publications.

The fourth (Appendix D) and fifth (Appendix E) manuscripts are both related to gaining a better understanding of why GaAs photonic crystal cavities exhibit much lower $Q$’s then their silicon counterparts or as predicted in finite difference time domain (FDTD) simulations. The first of these two, “GaAs photonic crystal slab nanocavities: Growth, fabrication, and quality factor”, discusses methods for reducing sacrificial slab roughness, a characteristic that can be observed even when the top layer of the sample is smooth. While the reduction of sacrificial slab roughness has not led to an increase in $Q$, another technique is shown which does; sample cleaning with potassium hydroxide. This cleaning method removes surface debris which arises from wet etching of the sacrificial layer with hydrogen fluoride. My role in this experiment was in taking photoluminescence measurements along with B. C. Richards and helping with KOH etching with B. C. Richards and U. K. Khankhoje. B. C. Richards performed the AFM measurements and D. Litvinov performed the TEM measurements. MBE growth for reducing sacrificial layer roughness was led by G. Khitrova and H. M. Gibbs. The
second manuscript discusses two additional mechanisms leading to Q degradation. One of these mechanisms is due to crystal axis dependant roughness resulting from the epitaxial growth process and the other is due to the presence of a bottom substrate underneath the free standing photonic crystal slab. Again, I played a substantial part in the photoluminescence measurements along with B. C. Richards. The modelling of the structures was done by U. K. Khankhoje and S-H. Kim. For both of these manuscripts the quantum dot growth was done by G. Khitrova and H. M. Gibbs and the photonic crystal fabrication was done by U. K. Khankhoje.

“In-situ annealing of InAs quantum dots on pre-structured GaAs substrates” (Appendix F), the sixth publication, is concerned with using annealing techniques in order to control the density and distribution of quantum dots on a substrate that has been fabricated with nanoholes in order to induce site-selective nucleation. Electron beam lithography, AFM measurements, and photoluminescence measurements were mainly performed by me with some help in the AFM and PL measurements by M. Gehl. Wet etching and quantum dot growth were performed by M. Helfrich.

The seventh and final manuscript, “Characterization of 1D photonic crystal nanobeam cavities using curved microfiber” (Appendix G) investigates high Q, small mode volume passive silicon nanobeam cavities by monitoring the transmission of a coupled fiber. These values are compared to those obtained with a crossed polarized resonant scattering technique. The fiber taper apparatus was set up by J-Y. Kim while the fabrication of the
nanobeams was carried out by A. Homyk with modelling by U. K. Khankhoje. B. C. Richards and I took the fiber coupling and crossed polarized resonant scattering measurements. I was responsible for setting up the crossed polarized resonant scattering apparatus.
CHAPTER 2: PRESENT STUDY

The methods, results, and conclusions of this study are presented in the papers appended to this dissertation. The following is a summary of the most important findings in these publications.

2.1 Quantum dot photonic crystal slab nanocavities

The first three sections of this chapter all deal with optical experimentation of GaAs photonic crystal slab nanocavities containing an embedded, single layer of InAs quantum dots.

2.1.1 Strong coupling

The paper “Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity” shows one of the first solid state realizations of strong light-matter coupling between a quasi two level emitter and high quality factor, small mode volume cavity. The emitter that is used is an InAs semiconductor quantum dot grown using MBE and utilizing the SK growth mode, while the cavity is formed by fabricating a freestanding GaAs photonic crystal slab with three missing air holes. A quantum dot density of 400/μm² was grown in the center of the photonic crystal slab with a random spatial distribution. Photoluminescence measurements were performed in reflection geometry
with the aid of a liquid helium cryostat containing internal x-y nanopositioners. The samples were optically pumped with above band excitation using a continuous wave titanium sapphire (Ti:Sa) ring laser and the resulting emission was dispersed with a spectrometer and detected on a liquid helium cooled indium gallium arsenide (InGaAs) charged coupled device (CCD) array; see Figure 7.

Figure 7: Schematic of experimental setup.
In order to reach the strong coupling regime the cavity - quantum dot system must meet two criteria; the first is a spectral matching of the quantum dot transition with the cavity mode frequency, and the second is a spatial requirement where the quantum dot must be physically located in an anti-node of the cavity. The ensemble quantum dot emission has an inhomogenous linewidth of 50 nm, and the individual quantum dots are spatially distributed in a random fashion. These features, as well as the need for a high quality factor cavity mode, necessitated the fabrication of a large number of photonic crystal cavities which were analyzed one by one until a structure was found with the desired features. With the single quantum dot emission blue detuned from the cavity mode, the quantum dot spectrum can be swept through the cavity mode by increasing the temperature. The range of the spectral shift is around 0.5 nm. Larger shifts require higher temperatures which lead to an increasingly broadened quantum dot transition linewidth due to phonon broadening. This in turn reduces the chances of observing strong coupling, as the requirement for this regime is that the interaction strength must be greater than the mean of the cavity decay rate and the quantum dot dephasing rate.

Once a structure in the strong coupling regime was found, it was analyzed at several different pump powers and as a function of the detuning between the quantum dot transition frequency and the cavity mode frequency. At high pump powers of 690 μW the emission is dominated by the cavity peak owing to the saturation of uncoupled quantum dots and to Purcell enhancement of the spontaneous emission of quantum dots weakly coupled to the cavity mode. The quality factor measured at high pump powers
was 13,300. As the power is lowered to the intermediate value of 25 µW, the surrounding uncoupled quantum dots are no longer saturated. Their absorption results in the decrease of the quality factor down to 8,000. As the power is lowered even further to 0.78 µW one can begin to see photoluminescence of individual quantum dots. By changing the temperature one of the quantum dot transitions is scanned across the cavity mode. Due to the strong light matter coupling the two peaks repel one another, exhibiting a characteristic anti-crossing. This can be seen in more detail in Figure 8 which shows the two coupled system peaks plotted as a function of temperature. Overlaid on this plot are the uncoupled quantum dot and nanocavity resonances. At zero detuning one can see that the uncoupled resonances are degenerate while the coupled system emission is double peaked.

![Figure 8](image)

Figure 8: The two coupled-system peaks are plotted as a function of temperature and compared with the scan rates of an uncoupled QD and an empty cavity peak.
2.1.2 Spectral cavity shifting through inert gas condensation

One way in which one could increase the probability of detecting strong coupling in a quantum dot photonic crystal slab nanocavity would be to increase the likelihood of spectral matching between the quantum dot transition and the cavity mode. The original method for meeting the spectral requirement was to use temperature tuning of the quantum dot. However, the scan range of this technique is limited by phonon broadening at higher temperatures, restricting the scan range to 0.5 nm. In order to increase the scan range while maintaining low temperatures and, hence, minimizing phonon broadening, a new technique was found. This technique involves the condensation of an inert gas onto a cryogenically cooled quantum dot photonic crystal nanocavity sample. As the gas condenses onto the sample, the slab thickness is effectively increased and the photonic crystal hole diameters are effectively reduced, resulting in a redshift of the cavity mode.

This experiment was carried out by measuring the wavelength of the cavity mode emission using above band pumping with a cw Ti:Sa ring laser in reflection geometry. Essentially this is the same setup as in the previous section. The only alteration to the setup was a small port introduced into the high vacuum system attached to the cryostat and allowing for the introduction of the inert gas. The chamber is initially evacuated to a pressure of $2 \times 10^{-6}$ Torr in order to reduce any impurities. A valve to the cryostat and a valve to the vacuum pump are closed while either xenon or nitrogen gas is introduced into the chamber. Once a desired pressure of gas is obtained the valve to the cryostat is
opened and the gas condenses onto the sample. The pump valve is then opened in order to bring the pressure back down to its initial level and the process is then repeated. At each cycle the cavity mode wavelength and quality factor are measured. The xenon gas showed a maximum cavity shift of up to 5 nm with minimal degradation of quality factor while larger shifts rapidly decrease the $Q$. The amount of the shift, however, tends to be cavity dependent as another structure showed only 3 nm shift without reduction in quality factor before a slow degradation set in. For nitrogen, the maximum scan range was limited to 4 nm at a temperature of 20 K but with much less consistency in the results. Using a temperature of 10 K the scans become much more consistent and smooth but are limited to 3.4 nm.

After a condensation scan is completed one must remove the condensed gas in order to perform a new scan. This is achieved by warming up the sample which melts off the condensed gas, allowing it to be pumped back out of the system. To remove xenon gas the sample is heated up above the xenon melting temperature of 161.4 K while for nitrogen one can either heat the sample up above the nitrogen melting temperature of 63.6 K or one can use local heating by a high power laser beam. Powers above 0.5 mW were found to be sufficient to locally remove the condensed nitrogen.

Using our new condensed gas scanning method we were able to observe strong coupling again, this time with two different quantum dots both strongly coupled to the same photonic crystal cavity. As cycles of xenon gas are condensed onto the sample the cavity
mode is scanned through both quantum dot transitions with each displaying a characteristic double peak and anti-crossing. The three peak positions are plotted as a function of xenon cycle number in Figure 9. In this figure one can see that the quantum dot transition wavelengths stay constant and only the cavity mode shifts. The uncoupled cavity mode resonance is overlaid on the graph and the double peak features can be observed around cycle number 11 and cycle number 22.

Figure 9: Strong coupling anti-crossing curves using xenon condensation to scan the cavity mode. The three peak positions are plotted versus cycle number, and anti-crossings are observed between the cavity mode and two distinct quantum dots. The dashed line shows the cavity mode spectra at high excitation power where coupling effects are not evident.
2.1.3 Gain - absorption effects and lasing

As earlier work had shown, surrounding uncoupled quantum dots in a photonic crystal cavity can lead to absorption effects and lower the quality factor of a cavity, hence decreasing the likelihood of observing strong coupling. In order to better understand this process a detailed study was carried out on gain, absorption, and lasing effects in the published paper “Quantum dot photonic-crystal-slab nanocavities: Quality factors and lasing” (Appendix C). Specifically, the emission linewidths of cavity modes were measured as a function of temperature and fabrication parameters for both high and low pump powers.

The sample was mounted in a continuous flow helium cryostat with internal x-y nanopositioners and optically excited with above band pumping using either a cw Ti:Sa laser at 780 nm or by a 20 ns pulsed diode laser operating at a 2.5% duty cycle and emitting at 784 nm. The pulsed laser was used for the lasing experiments in order to decrease any possible thermal heating effects. A 0.5 numerical aperture microscope objective is used in reflection geometry, and the collected emission is dispersed in a spectrometer and detected on a liquid nitrogen cooled InGaAs CCD array.

Five specific photonic crystal nanocavities were chosen for this study. Each nanocavity had the same values of r/a = 0.27 and s/a = 0.20 (r = radius, s = shift, a = lattice constant), however, different fabrication parameters such as electron beam dosing, hole radius,
lattice constant, and end spacer shift were used. The reason for choosing various fabrication parameters was to ensure that the cavity mode emission of the five nanocavities extended across the whole ground and first excited state spectral region of the quantum dot ensemble as the temperature is tuned from 20 K up to 200 K.

Through the judicious choice of the five nanocavities and with the aid of temperature tuning, the cavity emission linewidth was measured as a function of detuning from the quantum dot ensemble ground state peak. For both high (720 μW) and low (2-10 μW) cw pump powers, far red detuned cavity emission showed a constant linewidth of roughly 0.05 meV, equating to a quality factor of 20,000. This was assumed to be the empty cavity linewidth (linewidth in the absence of any gain or absorption effects) as the quantum dots are not expected to contribute any effects when sufficiently detuned from the cavity mode. Therefore, any linewidths which were measured with values greater then 0.05 meV were assumed to be affected by quantum dot ensemble absorption effects and, likewise, any linewidths with values under 0.05 meV are assumed to be affected by gain.

What one learns from this analysis is that for low pump powers, which are required for strong coupling type experiments, the cavity linewidth is dominated by the quantum dot absorption. For energies coinciding with the quantum dot ensemble ground state peak or beyond, the cavity quality factors are reduced due to absorption of surrounding quantum dots. The highest $Q'$s are found on the low energy side of the ensemble distribution,
suggesting that this energy range would be optimal. However, the lower absorption in 
this range also corresponds to a low density of quantum dots and, therefore, a decreased 
probability of meeting the spectral requirements for strong coupling. Alternatively, 
increasing the cw pump power results in a saturation of surrounding quantum dots and 
now as one proceeds from energy values in the far red detuned range in towards zero 
detuning with the ensemble peak a narrowing of the linewidth is observed. This gain is 
observed with detunings in the range of -30 to +10 meV. For larger positive detunings 
absorption again plays an effect, and the linewidths once again exceed the empty cavity 
value; however, the broadening is not nearly as severe as when one uses low pump 
powers.

With gain effects being observed it was then decided to inspect these samples further to 
see if the single layer of quantum dots can produce lasing when excited with sufficient 
power. In these investigations the pulsed laser was used in order to minimize any thermal 
heating effects. Two nanocavities were chosen for this experiment; one lying in the 
spectral range where no gain was observed and one in a range with maximum gain. As 
the pump power is increased the quality factor of the cavity in the no-gain region 
saturated at the empty cavity $Q$ value and its intensity increased in a sublinear fashion. In 
contrast, the nanocavity in the gain region had a $Q$ which continued to increase with 
increasing pump power, eventually reaching the instrument limited value of 40,000. 
Also, its intensity increased in a superlinear fashion. Like that of other photonic crystal 
structures with multiple embedded quantum dot layers [51, 52], lasing is evident in the
gain region nanocavity through the exhibition of linewidth narrowing and a threshold like behavior in the output versus input curve. However, estimates would have us believe that the number of quantum dots contributing to the lasing here is less than 16, at least an order of magnitude lower than any previous claims at that point in time. Lasing with such a low number of emitters implies a large spontaneous emission coupling factor as was also evident by the soft threshold that was witnessed.

2.2 Improvements in the growth, fabrication, and modelling of quantum dot photonic crystal slab nanocavities

The following two sections discuss different methods used in order to understand why quality factors of GaAs photonic crystal cavities are not higher than one would expect given results from simulations and other material systems.

2.2.1 GaAs photonic crystal slab nanocavities: Growth, fabrication, and photon storage time

Short wavelength (<1000 nm) GaAs photonic crystal cavities have long suffered from low quality factors as compared to the theoretically computed values using FDTD codes and as compared to similar devices made of silicon. A study was thus undertaken in under to try to determine what the deleterious effects could be through the use of AFM,
transmission electron microscopy (TEM), and scanning electron microscopy (SEM) as well as optical spectroscopy.

In order to create a photonic crystal slab a sacrificial layer is grown under the GaAs slab which is later removed through wet etching, post fabrication. Using TEM it was discovered that the top of the AlGaAs sacrificial layer can be rough even when the top of the slab is smooth, thereby also making the bottom of the GaAs slab rough. Since surface roughness can lead to increased scattering, measures were taken in order to reduce the roughness. In order to quickly analyze samples, the sample growth process was terminated immediately after sacrificial layer growth and it was scanned with an AFM. Different growth parameters were then investigated, such as introducing a growth interruption to give time for smoothing under Arsenic pressure, growing thin layers of GaAs within the sacrificial layer, and using a misoriented substrate. AFM measurements confirmed that utilizing these different types of growth parameters can reduce the surface roughness by an order of magnitude. Unfortunately, photoluminescence measurements did not show any increase in quality factor for samples with smooth bottom slabs as compared to those with rough bottom slabs. A possible reason for consistent quality factors is that fabrication imperfections could be the dominating force. Once the fabrication process is increased to a sufficient level, surface roughness effects may well prove to have a significant effect.
Using a SEM with the sample tilted with respect to the electron beam column, a large amount of debris was found on top of the samples. This debris was not immediately evident when the sample surface was positioned perpendicular to the beam column. It was speculated that this debris originated during wet etching of the sacrificial layer with a hydrofluoric acid solution. Assuming the debris was most likely a hydroxide of aluminum the samples were dipped into a potassium hydroxide solution. AFM and SEM measurements confirmed the removal of the debris. Photoluminescence measurements were taken on ten different nanocavities both before and after the potassium hydroxide dip and showed an average increase in the quality factor of 50% and a maximum increase of 73% for one cavity in particular. In addition, the photoluminescence intensity substantially increased after the potassium hydroxide dip and the cavity modes shifted to higher energy by average of 11 meV.

2.2.2 Modelling and fabrication of GaAs/InAs photonic-crystal cavities for cavity electrodynamics

In the paper “Modelling and fabrication of GaAs photonic-crystal cavities for cavity quantum electrodynamics” (Appendix E) a study was performed for a better understanding of the relatively low quality factors found for GaAs photonic crystal slab cavities. A detailed analysis of the modelling and fabrication of these devices was undertaken as well as an investigation of several important optical loss channels.
A number of fairly obvious factors that can lead to the degradation of the cavity quality factor were initially discussed. One of these factors is the uniformity of the photonic crystal holes. Any irregularities in the lithography or etching will obviously decrease the radiative quality factor. These irregularities can be attributed to a poorly focused electron beam and, hence, a careful control of the lithographic process is paramount. A second factor is problems with sample cleanliness such as leftover remnant resist and debris. Remnant resist can be removed by using an oxygen plasma treatment as more traditional wet chemical cleaning methods were found to be inefficient, leaving behind a thin layer of the resist. As discussed in the previous section, debris on the sample surface can be removed with a potassium hydroxide dip. A third factor that can reduce the quality factor is non-verticality of the photonic crystal hole side walls. There are two major contributors to non-verticality; inadequate or excessive resist development and non-vertical flow of chlorine gas onto the substrate during the dry etching process. Side wall slopes of only 2 degrees have been shown to lower the quality factor by an order of magnitude, making hole verticality an important issue.

Most modelling software assumes perfect hole uniformity, however, fabricated samples are not perfect. To better model the actual fabricated samples a 2D contour image was extracted from SEM images and imported into the FDTD software. For comparison, average photonic crystal parameters were measured from the SEM images and also used in the FDTD simulations. In the analysis of one specific nanocavity photoluminescence measurements showed a quality factor of 10,050. Using the simulation software with
average parameter values led to a quality factor of 31,418 while using the contour image method led to a quality factor of 21,283. While the contour method is obviously much closer to the measured value than the average parameter method, it is still significantly larger. The cause of this discrepancy could easily be accounted for by things such as side wall roughness and non-vertical air holes which cannot be accounted for in the FDTD simulations.

Upon removal of the sacrificial layer an air gap is created and below this air gap is a bottom substrate composed of GaAs and having a reflectivity of ~30%. This air gap must be greater than half the vacuum wavelength in order to reduce optical loss into the bottom substrate by evanescent coupling. However, an additional source of loss can then occur from multiple non-negligible reflections between the photonic crystal slab and the bottom substrate. This can lead to interference effects in the far field emission pattern and a reduction in the quality factor. One can view this reduction in quality factor and change in emission pattern as analogous to that which occurs when a point dipole source is placed in front of a mirror, a well-known problem in CQED. Simulations show that the quality factor can be changed by as much as 10%, making the choice of sacrificial layer thickness an important concern.

The last effect that was investigated in this study was the effect of crystal axis dependent surface roughness. As described in the previous section, the underside of the GaAs slab can show RMS roughness on the order of up to 25 nm. Using TEM and AFM it was
observed that this roughness is more pronounced along the [110] direction than in the [1-10] direction. Two sets of photonic crystal samples were then fabricated with one set having their cavity axes along the rougher direction and the other along the smoother direction. In order to make a meaningful comparison the exact same parameters were used for both sets of cavities. Photoluminescence measurements revealed a difference of 20-30% in the quality factor values between the two sets of cavities.

2.3 Deterministic growth of quantum dots

Epitaxial growth using the SK growth mode leads to a random spatial distribution of quantum dots which makes it difficult to locate an emitter in the antinode of a photonic crystal cavity. This would obviously make it very difficult for one to build up a network of strongly coupled quantum dot photonic crystal cavities where photonic crystal waveguides could connect one strongly coupled system to another; a potentially important step in quantum information and computation protocols. In order to address this issue we embarked on a project to deterministically grow quantum dots in site-selective locations on a substrate with an eventual long term goal of fabricating individual photonic crystals to individual quantum dots.

This process begins with the spin coating of PMMA/MA resist onto a GaAs substrate containing a 90 nm epitaxial GaAs buffer layer. Using electron beam lithography, several square arrays of nanoholes are defined in the resist with varying lattice constants.
The nanohole arrays are transferred into the substrate by wet chemical etching with H$_2$SO$_4$:H$_2$O$_2$:H$_2$O (1:8:800) producing holes of 30 nm in depth and 50-70 nm in diameter. After resist stripping and cleaning the sample is placed into the chamber of an MBE machine where it undergoes thermal deoxidation and Ga-assisted deoxidation to remove any remaining surface contamination and surface oxides. The next step is to grow a thin Ga buffer layer of 16 nm to help compensate for any surface roughness followed by growth of InAs quantum dots.

The nanoholes etched into the substrate act as preferential nucleation sites and quantum dots tend to grow in these locations first. Depending upon the amount of material grown, nucleation can also occur in the unstructured areas as well. In order to address issues such as quantum dot density, which can be directly related to the probability of nucleation in unstructured areas, uniformity of quantum dot size, and increased probability of single as opposed to double dot nucleation, an in-situ annealing process was used. To test this process samples were grown with and without a 2:30 minute annealing step. Additionally, some samples had a GaAs capping layer grown over the InAs quantum dot layer for photoluminescence experiments while other samples were halted after the quantum dot growth so that they could be characterized by AFM.

AFM measurements of the unannealed and uncapped sample showed that there was a predominance of two quantum dots forming at each nucleation site with some amount of growth occurring between the nucleation sites. The average quantum dot diameter was
measured to be 49.1 nm with a density of $4.8 \times 10^9/cm^2$. Annealing, which causes the material to redistribute through migration of adatoms, increased the number of single quantum dots located at each nucleation site. The sizes of the quantum dots, however, was much larger with an average diameter of 82.6 nm, a consequence of dominant ripening of the dots. Longer annealing times should help reduce the dot diameter. The quantum dot density fell by roughly 2 to a new value of $2.6 \times 10^9/cm^2$. Furthermore, both samples showed a 3x larger density in the structured areas as opposed to regions of the sample far removed from the patterned nanoholes, evidence of predominance of site-selective growth.

Photoluminescence measurements of the capped samples was undertaken in order to ascertain the optical quality of the deterministically grown quantum dots. Both annealed and unannealed samples show multiple individual quantum dot peaks with linewidths on the order of 100 $\mu$eV. Although luminescence was confirmed, the intensity of the spectra was reduced and the linewidths increased as compared to samples that were grown on unpatterned substrates. Still, this is an important step in the goal of site-selective growth of single quantum dots.

2.4 Silicon photonic crystal nanobeam cavities

Silicon photonic crystal cavities have shown remarkably higher quality factors than that of their GaAs counterparts. A detailed investigation was undertaken in the
characterization of passive 1-D silicon photonic crystal nanobeam samples with the hope of one day being able to couple quasi two level emitters to these structures.

InAs quantum dots can act as an internal emission source and make the characterization of GaAs photonic crystal samples relatively easy; a photoluminescence experiment is all that is needed. The silicon cavities that are studied here are passive, therefore, new techniques must be incorporated in order to determine the quality factors of these nanobeam samples. One method that was utilized was the coupling of a curved microfiber to the photonic crystal sample. This coupling of the fiber to the sample can create an additional loss mechanism and, hence, lower the observed quality factor. In order to better appreciate the extent of this degradation a comparison was made with a crossed polarized resonant scattering measurement. This second type of measurement does not load the cavity so intrinsic quality factor measurements are possible.

When coupling a curved microfiber to a nanobeam cavity there are several parameters that should be analyzed, such as the contact position of the fiber, the angle between the fiber axis and the nanobeam axis, and the polarization of the light in the fiber. By adjusting these parameters the loading of the cavity should be able to be minimized. To experimentally measure the quality factor while addressing each of these three issues, a tunable laser is input into the fiber and passes through an in-line polarization compensator before interacting with the nanobeam cavity. The light which is transmitted is then detected by an InGaAs photodiode. As the laser is tuned through a cavity mode a
change can be observed in the transmitted laser intensity and the FWHM of this transmitted lineshape can be used to extract the quality factor.

The contact position of the fiber on the nanobeam can drastically alter the transmitted signal. When contact was made near the edge of the nanobeam the coupling was weakest and the transmission dip at the cavity resonance was small. This led to much higher quality factor measurements as opposed to positioning the fiber on the center of the nanobeam where the coupling was the strongest and the transmission dip the deepest. In addition, placing the fiber directly onto the cavity region at the center of the nanobeam modifies the local index of refraction and shifts the cavity resonance.

Another parameter that can have an influence on the transmitted signal is the angle between the fiber and the nanobeam. Since the cavity mode is polarized orthogonal to the nanobeam axis and the light with the fiber is polarized perpendicular to the fiber axis, changes in the angle between the fiber and nanobeam can affect the coupling strength. With the fiber aligned parallel to the nanobeam the polarization matching should be the best, however, the coupling in this situation, as well as the modification to the index of refraction, is very large. Regardless of where contact on the nanobeams is made, the length of the contact region extends across the nanobeam so optimization can not be made in this configuration. Therefore a parallel configuration will produce a large dip but will not yield the highest measured quality factor. Perhaps a perpendicular configuration would then be optimal as the contact between the fiber and nanobeam
would be minimized? In fact, the optical coupling is so low in this arrangement due to
the orthogonal polarizations in the fiber and in the cavity that we were not able to
measure any transmission dip. The largest quality factor measurements came when the
angle was between 20 to 60 degrees with contact made at the edge of the nanobeam. This
configuration reduces the contact region while still allowing for some overlap between
the polarization components.

To better understand how lossy the curved microfiber technique can be a comparison was
made with spectra taken using a crossed polarized resonant scattering apparatus. In this
type of experiment the input light passes through a polarizer of a given polarization, such
as vertical, and is reflected off of the nanobeam whose axis is at 45 degrees to the
incoming light. An analyzer is then placed in front of a detector and aligned
perpendicular to the original input light polarization which in this case would be
horizontal. Any light that does not interact with the cavity mode will be attenuated
because of the crossed polarizers, however, light that has interacted with the cavity mode
will be resonantly scattered at 45 degrees, allowing some portion of that light to be
transmitted through the analyzer and reach the detector; see Figure 10. The positive
aspect of this technique is that one can measure the intrinsic, unloaded quality factor, but
it does suffer from one drawback in that the lineshapes of the signal can be highly
asymmetric. This asymmetry arises from a Fano like interference between the light
resonantly scattered from the cavity and the coherent reflected background [53]. The
quality factor must then be extracted by fitting the spectra with a Fano lineshape function
using an appropriately chosen asymmetry parameter value. Doing such allowed us to measure an intrinsic quality factor of 44,100 as compared to 29,000 when measured using the curved microfiber in the optimal configuration of 45 degree angle and edge contact. This shows that the curved microfiber method only degrades the quality factor by 30%.

![Crossed polarized resonant scattering setup](image)

**Figure 10: Crossed polarized resonant scattering setup.**

### 2.5 Summary of accomplishments

The contents of this dissertation have included a variety of investigations involving photonic crystal cavities, quantum dots, and their interactions. This work has already had a large impact on the general field of solid state cavity quantum electrodynamics as evidenced by the more than 575 citations that the article “Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity” (Appendix A) has received to date. This seminal result showing strong coupling in the solid state helped make tangible the
possibility of future integrated quantum devices for quantum information and computation, quantum cryptography, nanolasers, single photon sources, and so forth. In order to practically produce such devices one would need to easily meet the spectral and spatial requirements for photonic crystal cavity – quantum dot coupling. The condensation of inert gas experiment provided a method to essentially help relax the spectral coincidence requirement. Furthermore, studies on absorption and gain have showed that large ensembles of quantum dots will not be practical for realistic devices other than lasers. Optimally, one would like a single emitter coupled to a cavity. This led to the work carried out on the deterministic, site-selective growth of quantum dots. By growing a quantum dot at a known location, a cavity could later be fabricated at the same location and, hence, maximize the probability of meeting the spatial coincidence requirement. Additionally, by increasing the spacing between the site-selectively grown quantum dots one could avoid any deleterious ensemble absorption effects.

While GaAs photonic crystal slab nanocavities have exhibited just high enough quality factors to realize solid state cavity quantum electrodynamics experiments and prove their future applicability, for any hope of real, usable devices these quality factors must be increased. The studies that were carried out on growth, fabrication, and modeling were performed with exactly this in mind. Removal of debris and remnant resist has helped to edge the $Q$ values up slightly higher as has fabricating cavities along the smoother of the two crystallographic axes. Optimizing the MBE growth led to a smoother sacrificial layer and while this has not produced higher $Q$'s as of yet it could very well lead to
improvements once other larger contributing loss factors have been overcome, such as hole verticality and uniformity.

The experiments performed with silicon nanobeam cavities were also motivated by the desire for higher quality factors. When making identical type structures in silicon and gallium arsenide, silicon always exhibits a larger quality factor. The nanobeam design itself is also quite promising and seems to be an improvement upon the more traditional 2D slab design. Having a smaller footprint, nanobeams could also allow for tinier, more compact devices. The only current drawback with silicon is the difficulty of coupling emitters as presently one can not simply grow a layer of high optical quality quantum dots within a silicon structure like one can do for gallium arsenide.

In conclusion, the work of this dissertation has helped advance the entire field of solid state cavity quantum electrodynamics. The initial strong coupling data helped open the door and the subsequent experiments have all been pushing forward the promising photonic crystal cavity – quantum dot device.

2.6 Future directions

What directions will the work of this dissertation lead to in the future? One of the beautiful features of the photonic crystal is the relative ease with which one cavity could be coupled to another by simply creating waveguides through the removal of entire rows
of air holes nearby the cavities. With increases in deterministic quantum dot growth it should only be a matter of time before one could begin creating networks of strongly coupled cavities. However, for all their promise there is still one big underlying problem that could be the Achilles heel of photonic crystal – quantum dot devices for solid state implementations of quantum information and computation and that is the fast dephasing times of these systems. If dephasing times are faster then qubit operation times then information will be lost and computation will not be able to occur.

Thinking more generally about cavities and emitters, there are other interesting directions one could take using the research explained in this dissertation as a background. Metamaterials have garnered much interested in the past decade and the split ring resonator has properties which could be very interesting from a CQED perspective. While the current quality factors of a split ring resonator is only on the order of 10, its plasmonic mode volume can be two orders of magnitude smaller then a photonic crystal cavities mode volume. Even with such low quality factors, the extremely small mode volumes can give Purcell enhancement factors in the $10^5$ range. In fact, for certain applications such as high modulation rate light emitting diodes and single photon sources the low $Q$ values could be beneficial; a low $Q$ means that light will quickly escape from the cavity. Deterministically coupling quantum dots to split ring resonators could indeed lead to very interesting future quantum metaphotonic devices.
REFERENCES


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APPENDIX A

Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity

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coupling. The excitation power used in the strong coupling experiments of 2×10^6 corresponds to 1.5×10^{13} photons s^{-1} in the cavity averaged over time; that is, interactions of the single QD with multiple photons can be neglected.

Our experiments demonstrate that long-sought solid state implementations of the strongly coupled cavity-mode-two-level-entangled systems are feasible by using single QDs in high-Q cavities with small mode volumes. With further improvements, for example using higher-Q cavities or QDs placed at the in-plane mode centre, these systems have the potential for wide application ranging from nonlinear optics to quantum information processing[^12].

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[^12]: Nature Publishing Group

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**Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity**

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Quantum systems exhibit a variety of fundamental quantum-optics phenomena, such as an enhancement of quantum decoherence and the quantum-classical boundary. Such systems also provide test beds for quantum information science. Nearly all strongly coupled cavity QED experiments have used a single atom in a high-quality-factor (high-Q) cavity. Here we report the experimental realization of a strongly coupled system in the solid state: a single quantum dot embedded in the spacer of a nanocavity, showing vacuum Rabi splitting expanding the decoherence linewidths of both the nanocavity and the quantum dot. This requires a small-volume cavity and an atomic-like two-level system. The photonic crystal slab nanocavity—which traps photons when a defect is introduced inside the two-dimensional photonic bandgap by moving out one or more holes—has both high Q and small mode volume V, as required for strong light-matter interactions. The quantum dot has two discrete energy levels with a transition dipole moment much larger than that of an atom, and is fixed in the nanocavity. The study of vacuum Rabi splitting has been an exciting subject of atomic physics since its first observation with many atoms in the early 1980s; see ref. 1 for a history of the field. After a decade of gradually improving the Q of the cavity and decreasing its volume, vacuum Rabi splitting was observed with a single atom. This opened exciting opportunities for the field of atomic cavity QED, and many experiments followed[1]. For such a truly quantum system, the optical properties are changed by the addition of a single photon or single atom, and the quantum-classical boundary can be studied[4]. But because atoms can move and even escape, their coupling is time-dependent; clearly, the next goal was to localize a cold atom inside the cavity using atomic traps[5]. In the field of semiconductor, 12 years elapsed between seeing non-perturbative normal mode coupling[6], analogous to many-atom vacuum Rabi splitting[7], and the observation of strong coupling between a single quantum dot (QD) and a small-volume crystal nanocavity. This advance, which produced opportunities for truly quantum-optics cavity QED experiments in semiconductors, owes much to the extensive studies of and improvements in SQDs and monolithic cavities. The semiconductor approximation to a two-level system in a SQD is a small semiconductor crystal confined in three dimensions by a higher-bandgap material[10]. The sharp emission lines observed from submicrometer collection spots were shown to arise from transitions between discrete energy levels of the quantum dot (QD) depending upon size and shape[11]. Coherent transient experiments were performed on these atom-like transitions[12,13], and their spontaneous emission was enhanced[14] and inhibited[15] by the Purcell effect within materials of higher than Q. The transitions of a SQD can be separated enough for the longest transition to exhibit anti-bunching, and cavity enhanced spontaneous emission can lead to one photon on demand into a desired mode[16,17].

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**Cavity quantum electrodynamics (QED) systems allow the study of a variety of fundamental quantum-optics phenomena, such as an enhancement of quantum decoherence and the quantum-classical boundary. Such systems also provide test beds for quantum information science. Nearly all strongly coupled cavity QED experiments have used a single atom in a high-quality-factor (high-Q) cavity. Here we report the experimental realization of a strongly coupled system in the solid state: a single quantum dot embedded in the spacer of a nanocavity, showing vacuum Rabi splitting expanding the decoherence linewidths of both the nanocavity and the quantum dot. This requires a small-volume cavity and an atomic-like two-level system. The photonic crystal slab nanocavity—which traps photons when a defect is introduced inside the two-dimensional photonic bandgap by moving out one or more holes—has both high Q and small mode volume V, as required for strong light-matter interactions. The quantum dot has two discrete energy levels with a transition dipole moment much larger than that of an atom, and is fixed in the nanocavity.**

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**Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity**

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Quantum systems exhibit a variety of fundamental quantum-optics phenomena, such as an enhancement of quantum decoherence and the quantum-classical boundary. Such systems also provide test beds for quantum information science. Nearly all strongly coupled cavity QED experiments have used a single atom in a high-quality-factor (high-Q) cavity. Here we report the experimental realization of a strongly coupled system in the solid state: a single quantum dot embedded in the spacer of a nanocavity, showing vacuum Rabi splitting expanding the decoherence linewidths of both the nanocavity and the quantum dot. This requires a small-volume cavity and an atomic-like two-level system. The photonic crystal slab nanocavity—which traps photons when a defect is introduced inside the two-dimensional photonic bandgap by moving out one or more holes—has both high Q and small mode volume V, as required for strong light-matter interactions. The quantum dot has two discrete energy levels with a transition dipole moment much larger than that of an atom, and is fixed in the nanocavity. The study of vacuum Rabi splitting has been an exciting subject of atomic physics since its first observation with many atoms in the early 1980s; see ref. 1 for a history of the field. After a decade of gradually improving the Q of the cavity and decreasing its volume, vacuum Rabi splitting was observed with a single atom. This opened exciting opportunities for the field of atomic cavity QED, and many experiments followed[1]. For such a truly quantum system, the optical properties are changed by the addition of a single photon or single atom, and the quantum-classical boundary can be studied[4]. But because atoms can move and even escape, their coupling is time-dependent; clearly, the next goal was to localize a cold atom inside the cavity using atomic traps[5]. In the field of semiconductor, 12 years elapsed between seeing non-perturbative normal mode coupling[6], analogous to many-atom vacuum Rabi splitting[7], and the observation of strong coupling between a single quantum dot (QD) and a small-volume crystal nanocavity. This advance, which produced opportunities for truly quantum-optics cavity QED experiments in semiconductors, owes much to the extensive studies of and improvements in SQDs and monolithic cavities. The semiconductor approximation to a two-level system in a SQD is a small semiconductor crystal confined in three dimensions by a higher-bandgap material[10]. The sharp emission lines observed from submicrometer collection spots were shown to arise from transitions between discrete energy levels of the quantum dot (QD) depending upon size and shape[11]. Coherent transient experiments were performed on these atom-like transitions[12,13], and their spontaneous emission was enhanced[14] and inhibited[15] by the Purcell effect within materials of higher than Q. The transitions of a SQD can be separated enough for the longest transition to exhibit anti-bunching, and cavity enhanced spontaneous emission can lead to one photon on demand into a desired mode[16,17].
The condition for strong coupling is more demanding on Q: the vacuum Rabi splitting, \( 2\gamma \), due to a SQD must exceed the mean of the decay rates of the cavity, \( \kappa \), and the dot, \( \gamma \). The coupling strength, \( g^2 \), is given by \( \mu \varepsilon_{\text{vac}}^2 V = \hbar \nu / 2 \). Here \( n = 3.4 \) is the semiconductor refractive index, \( \varepsilon_{\text{vac}} \) is the permittivity of vacuum, \( V \) is the mode volume, and \( \nu \) is the frequency of the transition of the quantum dot with dipole moment \( \mu \). For small-length cavities, \( \kappa = \nu g^2 / \hbar \) usually exceeds \( \gamma \); then, since \( \kappa \gg \gamma \), the challenge has been to fabricate a high-Q cavity while maintaining a very small \( V \). Recently, a breakthrough in design by Noda's group resulted in silicon photonic crystal nanocavities with \( Q = 45,000 \) and \( V = 0.07 \) \( \mu \text{m}^3 \). Clearly, the cavity with the smallest \( V \) (while maintaining high \( Q \)) yields strong coupling with a smaller dipole, that is, the dot is more quantum.

Our photonic crystal nanocavity follows the design of Noda's group, but with a new twist: a triangular lattice photonic crystal sphere with three holes missing to form a spacer (Fig. 1a, b). The vertical confinement, achieved by total internal reflection at the slab semiconductor-air interfaces, is imperfect, in that light with small in-plane wavevectors can leak out of the top and bottom. Noda's one-dimensional model showed that the key to reducing this loss is to shift out slightly the holes at the ends of the spacer: "the light has to be confined gently in order to confine it strongly." In other words, when the field envelope function is stopped abruptly, its Fourier transform has a larger overlap with small in-plane wavevectors that leak out; terminating it gently reduces that loss.

The sample, grown on a (001) GaAs substrate by molecular beam epitaxy, has a single layer of InAs quantum dots in the centre of the slab (Fig. 2a). A large array of nanocavities (~30,000 in clusters of 30 with a density of 5569 cavities \( \text{mm}^{-2} \)) is fabricated with crystal parameters changed systematically. The missing-holes spacer is surrounded by 14 periods of air holes for good in-plane optical confinement. The parameters of the photonic crystal are controlled lithographically: \( a = 300 \) \( \text{nm} \), \( r = 0.27a \), and slab thickness \( d = 0.09a \) (see Fig. 1b) for definitions of \( a \), \( r \), and \( d \).

Computation of the field strength as a function of position (Fig. 1c) shows that most of the field energy is confined to the defect region with a mode volume of \( V = 3.7 \) \( \mu \text{m}^3 \), whereas \( V \) is the resonance wavelength of light in vacuum. This \( V \) is a typical value for most of the parameter ranges. Since the intracavity field is a standing wave that oscillates from zero to a maximum every quarter wavelength (see top of Fig. 1c), there is a very limited volume of high field strength in which a SQD must be located if it is to couple strongly.

Photoluminescence (PL) measurements were performed in a temperature-controlled liquid-helium cryostat with internal \( x-y \) manipulators, essential for stability and the ability to re-find a
given nanocavity. The samples were optically pumped by the 770 nm output of a Ti:sapphire continuous wave (cw) laser. The pump beam was focused by a reflecting microscope objective (0.5 numerical aperture) to a spot size of 1 μm on the sample. The sample emission was collected by the same microscope objective, analyzed with a spectrometer, and detected by an InGaAs array integrating over 0.025 mm per pixel. We estimate that a sample area of ~10 μm² is imaged into the spectrometer, giving rise to the broad ensemble PL underlying the cavity-related emission in Figs 3 and 4.

In this geometry, we are using the leakage of the cavity mode out of the top to observe PL from a QD coupled to it. Figure 2b shows the ensemble PL spectrum with the lowest transition line at ~1,200 nm, and the first excited transition line at 1,125 nm. Figure 2c shows high-power spectra of the three highest-Q nanocavities. There has been a steady improvement in the values of Q obtained for two-dimensional photonic crystal nanocavities, fabricated for lasers, with a quantum well (Q = 250) or ~60 quantum dots (Q = 2,000) as the active medium.

We do not find a QD coupled strongly to one of the highest-Q modes displayed in Fig. 2c. But a slightly lower-Q (~13,000) mode does couple to a QD located specifically on the short wavelength side of the lowest-energy transition of the ensemble shown in Fig. 2b. At high power Fig. 3a as in Fig. 2c, the emission is dominated by the cavity peak, because QDs not coupled to the cavity are saturated: that is, a QD's emission rate is determined by its radiative decay rate, not by its excitation rate. Therefore, coupled dots emit more photons per unit time than uncoupled dots, owing to Purcell enhancement of spontaneous emission. A time-resolved experiment would be needed to see the faster decay of a coupled dot.

At intermediate power (25 μW), the increased QD absorption reduces the Q to 8,000. At low power (Fig. 3b), one can begin to see PL peaks from uncoupled QDs; we note that they all move together in the same way with temperature as does the empty cavity mode, but at a rate much faster than that mode. Therefore, a QD transition can be temperature-scanned through the cavity resonance. Figure 2b shows an anti-crossing of one QD transition with the cavity mode at 1,187.6 nm. The two normal modes repel each other in the vicinity of the crossing of the red and blue lines, showing the temperature dependence of the uncoupled QD and cavity mode resonance, respectively.

In Fig. 4a, the two coupled-system peaks are plotted as a function of temperature on an expanded wavelength scale; zero detuning where the uncoupled dot and nanocavity resonances are degenerate, the coupled system emission is clearly double-peaked. This anti-crossing behavior is characteristic of strong coupling, the regime of reversible exchange of energy back and forth between the SOD and the nanocavity—i.e., vacuum Rabi oscillations.

Figure 4a shows an independent scan over a narrower temperature range close to zero detuning. The measured zero detuning vacuum Rabi splitting is 2ω = 41 GHz = 170 μeV = 0.192 nm.

Figure 4b displays the zero-detuning emission predicted by an analytic expression. There is some uncertainty in the values of ω and γ, and even more in the location of the QD relative to the field maximum. For the plot, ω = 20.6 GHz; assuming that the dot is in the field maximum, this corresponds to μ = 29 D and a radiative lifetime of 1.82 ns (ensemble measurements gave 1-2 ns).
Even though ~10^10 photons per second are emitted by the coupled system at low power, the signal is weak because most of them stay in the 3dB-detuning through a waveguide coupled to the microcavity would be far better. If we indeed had only one SQO, there would be little emission from the cavity peak for dot-cavity detuning larger than g; but it occurs because of the high density of weakly coupled QDs. Note that the emission spectrum of a strongly coupled system is double-peak in all directions, unlike that of a quantum-well planar microcavity, which is double-peak in the nonperturbative regime, perpendicular direction while single-peak in the weak-coupling-regime, in-plane direction. This means that the energetic position of the emission peaks should be independent of detection direction.

The anti-crossing of Fig. 3b was observed many times by cycling the temperature. In a new sample, we have seen another clear anti-crossing in the 3dB-detuning vacuum Rabi splitting at 22 GHz. We have also seen the weak coupling regime of Panellenhancement of spontaneous emission: temperature scanning causes QD resonance to cross straight through the cavity resonance, but coupling increases the radiative linewidth.

We expect that our dot/microcavity system will exhibit truly quantum effects, although the linear spectroscopy that we report here has not proved this. In contrast, even though the normal-node coupling seen between a single quantum well and a microcavity also results in a two-peaked anti-crossing and is often called strong coupling by the semiconductor community, it is actually semimicrocavities—much like many atom vacuum Rabi splitting. Even a quantum-well three-dimensional microcavity with a mode diameter of only ~2 μm still requires ~300 photons to saturate its vacuum Rabi splitting; it is semi-classic, and far from the quantum regime.

There are at least two advantages of semiconductor QD cavity QED over atomic cavity QED. First, the dot position is fixed, the ability to do experiments with one and the same quantum emitter is essential for both interesting physics and applications in quantum information science. Second, the ultra-small size of the strongly coupled dot/cavity device, with ~10^9 cavities nm^2, allows us to speculate about a quantum network that would be able to store, process and distribute quantum information. The interconnections would be made by photonic crystal waveguides (using lines of holes). The essential element of a quantum network is a deterministic strong coupling of a single dot to a high-finesse optical photonic crystal cavity—shown as demonstrated here.

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Current-induced resonance and mass determination of a single magnetic domain wall

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A magnetic domain wall (DW) is a spatially localized change in magnetization configuration in a magnet. This topological object has been predicted to behave as a low-energy particle with finite mass\(^1\). This particle will couple directly with electric currents as well as magnetic fields, and its manipulation using electric currents\(^2\) is of particular interest with regard to the development of high-density magnetic memories. The DW mass sets the ultimate operation speed of these devices, but has yet to be determined experimentally. Here we report the direct observation of the dynamics of a single DW in a ferromagnetic nanowire, which demonstrates that such a topological particle has a very small but finite mass of 6.8 × 10^-14 kg. This measurement was realized by preparing a tunable DW potential in the...
APPENDIX B

Scanning a photonic crystal slab nanocavity by condensation of xenon

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Scanning a photonic crystal slab nanocavity by condensation of xenon

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Allowing xenon or nitrogen gas to condense onto a photonic crystal slab nanocavity maintained at 10–20 K results in shifts of the nanocavity mode wavelength by as much as 5 nm (~4 meV). This occurs in spite of the fact that the mode defect is achieved by emitting three holes to form the spacer.

This technique should be useful in changing the detuning between a single quantum dot transition and the nanocavity mode for cavity quantum electro-dynamics experiments, such as mapping out a strong coupling antitwisting curve. Compared with temperature scanning, it has a much larger scan range and avoids phonon broadening. © 2005 American Institute of Physics.

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Radiative coupling between a single quantum dot (SQD) and a small volume cavity alters the emission properties of the coupled system. In the weak coupling regime, the spontaneous emission has a single emission frequency, and it irreversibly escapes from the cavity. The SQD radiative emission rate is described by the Purcell factor $F_p = 3Q/4\pi V$, compared with the cavityless radiative emission rate $\gamma_e$ and $V$ are the quality factor and volume of the cavity, respectively, and $Q = \omega_0 / \kappa$ is the linewidth of the light in the material with refractive index $n$. The several single-photon-on-demand sources that have been reported operate in the weak coupling regime. In the strong coupling regime, the Rabi frequency $\Omega$ of exchange of the excitation between the SQD and the cavity, $g=\mu E_{\omega_0}/\hbar$, exceeds both the photon escape rate $\kappa$ and SQD depleting rate $\gamma$. In the formula for $g$, it is assumed that the SQD has dipole moment $\mu$ and is in the peak of the one-phonon-in-squaring intracavity field $E_{\omega_0}$ satisfying $e\Omega^2 [\mathcal{E}_{\omega_0}]^2 = \hbar \nu = 2\nu$, $e_g$ is the permittivity of vacuum, and $\nu$ is the frequency of the cavity mode. The strong coupling makes the spontaneous emission reverable, i.e., a photon emitted by the excited SQD has a higher probability of being reabsorbed than escaping the cavity. For zero detuning between the SQD and the cavity modes, the coupled-system spontaneous emission can occur at either of two frequencies separated by $2g$. If the coupled transition of the SQD is between excited state $|e\rangle$ and ground state $|g\rangle$ and the quantized field state with $n$ photons in the cavity mode is denoted by $|n\rangle$, then the two coupled-system eigenstates can be written as $|n\rangle |g\rangle$ and $|n\rangle |e\rangle$. Since neither eigenstate can be written as a product of a quantum dot (QD) state and a cavity state, each has entanglement—a basic ingredient of quantum information science. Recently, there have been three experimental claims of observing this vacuum Rabi splitting using for the nanocavity a micropillar, a photonic crystal slab, and a microdisk. The photonic crystal slab nanocavity has the smallest mode volume, $(\sqrt{3}/4)\lambda_0^2$, and it will have the largest vacuum Rabi splitting $2g$ for a given SQD dipole moment, since $E_{\omega_0} \approx 1/\sqrt{V}$.

For many years, $\kappa$ of a photonic crystal nanocavity has been larger than $\gamma$, therefore, since $\gamma < \kappa < Q$, it has been $g > \sqrt{Q}\nu / \sqrt{V}$ that needed to be maximized for strong coupling. Even when $g > 1$ has been achieved in principle for a particular nanocavity and a given SQD dipole moment, one has the problem of finding a SQD that is accidentally situated spatially close to the intracavity field maximum within the tiny mode volume and accidentally positioned spectrally close to the cavity mode frequency. To increase the statistical likelihood of this occurring, it is tempting to make the QD density very high. But if it is too high, there are two undesirable consequences. The emission lines of so many QDs start to overlap, complicating the study of cavity coupling to one particular SQD. And the ensemble absorption of the layer of QDs situated throughout the semiconductor slab reduces the $Q$. When the QD density is low enough to avoid these problems, the likelihood of finding vacuum Rabi splitting in any given nanocavity is very low. One must look for it by searching through many nanocavities, looking for the required accidental coincidences in space and frequency. In all three of the reported strong coupling experiments, the temperature was used to change the SQD nanocavity detuning, taking advantage of the fact that the SQD transition shifts to a longer wavelength with increased temperature much faster than the cavity mode. Temperature scanning, however, is limited to $\approx 0.7$ km because of the increase in QD linewidth $\nu$ due to phonon broadening at higher temperatures. A technique for changing the detuning while keeping the system at $\approx 4$ K would minimize the phonon broadening. And if the technique could scan further than 1 nm, it would make it easier to find the required coincidences at a given dot density, or it would make it possible to find them at a lower QD density. Even if one finds a way to grow a SQD in the
center of a photonic crystal slab nanocavity or to fabricate a photonic crystal nanocavity around a spatially isolated SQD, for the foreseeable future it will still be a challenge to make the SQD transition frequency coincide with the cavity frequency, i.e., tuning will still be essential. Here, we report on a 4 nm scan range achieved by condensing Xe gas onto a photonic crystal slab nanocavity at 20 K.

Figure 1 is a schematic of our photonic crystal slab nanocavity. Vertical optical confinement is by total internal reflection at both the top and bottom semiconductor/silica interfaces. The stopband of the two-dimensional photonic crystal provides in-plane "mirrors." The cavity mode is created by omitting three holes to open up a defect state within the stopband. The shifting out of the "spazer" end holes greatly increases the Q. The distribution of the intracavity field shows that the field is still appreciable at the edges of the holes around the spacer and at the semiconductor/silica interfaces where the Xe can condense. This explains why a large shift is observed even though there are no holes in the center of the spacer in which Xe can condense. Much larger shifts would be obtained using a nanocavity with the defect introduced by reducing the diameter of one hole. But for strong coupling applications, the larger spacer and larger Q of the present design are essential, and the 5 nm scan range is still large enough to be very useful.

The shift experiments are performed as follows. The wavelength of a cavity mode is observed by detecting photoluminescence (PL) from a single layer of QDs with a density of 300–400 QDs/μm², grown by molecular beam epitaxy in the center of the ~270 nm GaAs slab. A nanocavity was selected that has a mode around 1237 nm, on the long-wavelength edge of the QD ensemble PL; this avoids the reduction of Q by the ensemble QD absorption. The QDs are optically pumped nonresonantly by focusing the output of a continuous-wave titanium:sapphire laser onto the nanocavity with a spot diameter of about 2 μm, using a reflective microscope objective with numerical aperture 0.5. When the pump power exceeds roughly 100 μW, the uncoupled and very weakly coupled QDs with radiative lifetimes of 1–2 ns are almost all saturated. More strongly coupled QDs within the cavity peak are then able to emit many more photons per unit time, making the cavity peak stand out clearly. The PL is collected by the same objective in reflection geometry, dispersed in wavelength by a spectrometer, and detected by an InGaAs linear array. The sample is mounted in a CrysTec (Germany) Konti-Mikro cryostat with temperature control from 325 to 3.5 K, by adjusting the He flow and the internal heater. The sample can be moved horizontally and vertically by a computer-controlled internal nanopositioner.

![Graph](image-url)

**Fig. 2.** The shift of cavity peak (a) and the value of the cavity quality factor Q (b) as a function of the Xe condensation cycle. 0.5 Torr of Xe was introduced each cycle in a 0.75 liter volume; the temperature was 20 K. (c) Same as (a) for Xe(60 Torr per cycle; 10 K). The condensation time was approximately 3 min, and the continuous-wave excitation was at 740 nm.

This is critical for the stability of our measurements and for the ability to re-find a specific nanocavity. The Xe gas (99.999% pure) is admitted from a high-pressure (~5000 kPa) tank, through a regulator that reduces the pressure to ~50 kPa and an ultrahigh-vacuum valve, into a vacuum chamber with a volume of ~0.75 liter. Both the admission line (including the regulator) and the vacuum chamber are initially evacuated to a pressure of ~2 × 10⁻⁵ Torr, to reduce the presence of impurities (air and water vapor). Another valve isolates the vacuum chamber from the cryostat where the sample is kept at low temperature. Once the desired pressure (measured with a convection gauge) of pure Xe gas is achieved in the vacuum chamber, the Xe flow is stopped. The Xe gas accumulated in the vacuum chamber is then allowed into the cryostat. The sample and all other sufficiently cold surfaces act as a very efficient adsorption pump, condensing the Xe gas. The shift of the cavity peak is essentially simultaneous with the Xe gas condensation, and it can be monitored live. After the initial pressure is recovered, the cycle is repeated.

For our particular system, there is a very narrow Xe pressure range that can be used. For a pressure below about 0.45 Torr, there is no shift even if the cycle is repeated many times. If the pressure is much above 0.5 Torr, the ~4 nm scan limit is reached in one or a few cycles. However, in the last case, the quality factor Q of the cavity is degraded, so very fast scanning has no practical value. Some results from scanning experiments are shown in Fig. 2. The laser pump power was adjusted to 1.5 μW, to minimize laser-induced heating, and the temperature was held at 20 K. For Xe, initially the cavity peak shifts quasi-linearly with the amount of gas introduced, then the shift starts to saturate; see Fig. 2(a). Q, the quality factor of the cavity, starts with a value of ~12,000 and does not drop below 11,000, as shown in Fig. 2(b).

The behavior of Q depends upon the particular nanocavity: Another one was scanned almost 5 nm, with the Q remaining constant at 13,000 for the first 3 nm before dropping slowly to 6000.
If $N_2$ is used instead of Xe, holding the sample at a temperature of 20 K, it is also possible to scan up to 4 nm, but the results are much less reproducible. Lowering the temperature to 10 K, the scanning becomes smooth and reproducible, but the scan limit is reduced to about 3.4 nm (2.7 nm without much degradation of $Q$), as shown in Fig. 2(c). Also, the amount of $N_2$ gas used for condensation is more than 100 times that of Xe.

Of course, after scanning, one must remove the condensed gas before making a new scan. This is done by warming up the sample above the melting temperatures (161.4 K for Xe and 63.5 K for $N_2$), while continuously pumping on the system. Or, the cavity peak shifts back to its initial value due to local heating by a high-power (~0.5 mW) laser beam, for sample temperatures above 20 K. This effect occurs for $N_2$, but not for Xe. It is, as well as the behavior of the scanning above 20 K, can be attributed to the lower melting point of nitrogen. Therefore, $N_2$ is easier for multiple coarse scans, whereas Xe scans more precisely.

In fact, the idea of scanning by Xe condensation arose from lack of reproducibility of the cavity wavelength in earlier runs; we concluded this irreproducibility came from the condensation of air or water vapor due to an inadequate vacuum. Often, one does not take great care with the vacuum in a crystal when the sample is a microcavity or multiple quantum wells whose optical properties are mostly determined by layers well below the surface (which can be cleaned periodically to remove any condensation). But the photonic crystal is very sensitive to condensation, so the mode wavelength depends on both the slab thickness and the hole diameter. When we first cool down the sample, the wavelength of the cavity mode is not predictable within several mm, presumably due to the condensation of background gas. In order to recover a reproducible (shorter) wavelength, the sample is heated back up to 310 K and then immediately cooled back down. We guess this drives off the offending condensate that then recondenses somewhere else. This procedure is performed before starting the condensation.

Figure 3 illustrates that the condensation technique can indeed be used to scan the SQD (nanocavity) detuning through a strong coupling anticrossing. This technique should enable 4 K operation, thus stabilizing quantum dot dephasing that degrades strong coupling. In conclusion, condensation of xenon or nitrogen gas onto a photonic crystal slab nanocavity has been shown to be a useful way to scan the cavity mode by as much as 5 nm while maintaining the nanocavity at ~25 K.

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APPENDIX C

Quantum dot photonic-crystal-slab nanocavities: Quality factors and lasing

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Quantum dot photonic-crystal-slab nanocavities: Quality factors and lasing

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Emission linewidths of quantum dot photonic-crystal-slab nanocavities are measured as a function of temperature and fabrication parameters with low-power and high-power, cw and pulsed, nonresonant excitation. The cavity linewidth is dominated by the absorption of the ensemble of quantum dots having a density of \( n = 4 \times 10^{10} \) cm\(^{-2} \) above the absorption edge, the cavity linewidth broadens considerably compared with the empty cavity linewidth. Gain and lasing are seen for high-power pumping; it is estimated that only a small number of quantum dots contributes to the lasing.

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Recently the quality factor \( Q \) (mode energy divided by full width at half maximum [FWHM] mode energy linewidth) of photonic-crystal-slab cavities has been steadily increased by improved fabrication techniques and designs, while the wavelength was kept close to a cubic wavelength in the material. This has made possible not only quantum well and quantum dot lasers but also the observation of strong coupling\(^4\) of vacuum Rabi splitting with a single quantum dot (SQD). The role of the quantum dots (QDs) in the lasers is to provide gain, so several layers of high density QDs are often used. In contrast, strong coupling, can best be observed with an isolated SQD, suggesting the use of a single layer of low density QDs. However, to see strong coupling one must search to find two accidental coincidences. The QD must be situated close to an in cavity field maximum. This means it must be within the mode area of 0.15 \( \mu \)m\(^2\), where the in cavity field is strong. It must also have a transition frequency close to a cavity mode; our ensemble QD lowest energy transition has a FWHM of 42.5 meV at 20 K, compared with a maximum dot-nanocavity coupling strength of 0.2 meV. For a reasonable probability for both coincidences, high dot densities (900–9000 \( \mu \)m\(^2\)) have been used so far. This paper addresses two questions: Is the ensemble QD absorption detrimental to the search for strong coupling? And, if the gain is sufficient for lasing, roughly how many QDs contribute?

To fabricate a photonic-crystal-slab nanocavity, a sample is grown by molecular beam epitaxy on a (001) GaAs substrate starting with a GaAs buffer layer, 800 nm \( Al_{0.3}Ga_{0.7}As \) sacrificial layer, 40 nm GaAs, 20 nm \( Al_{0.3}Ga_{0.7}As \), single layer of self-assembled InAs QDs (density of 300–400 \( \mu \)m\(^2\)), and on top of the dots 20 nm \( Al_{0.3}Ga_{0.7}As \) and 40 nm GaAs.\(^4\) Then a two-dimensional triangular photonic-crystal-lattice with three holes missing to form a cavity spacer is fabricated to provide in-plane light confinement. The GaAs-air interfaces on the top and bottom of the 270-nm-thick slab provide vertical confinement by means of total internal reflection, but light with small in-plane wave vectors still leaks out of the cavity. As shown by Noda’s group using Si, vertical confinement is further enhanced by slightly shifting outward the holes at the ends of the spacer; the \( Q \) is increased by confining gently.\(^5\)

The quantum dots are excited by a continuous-wave Ti:Sapphire laser (wavelength 780 nm) or alternatively by a 20 ns pulsed diode laser (wavelength 784 nm). Sharp Microelectronics, 2.5% (neararity factor). A 0.5 numerical aperture (NA) microscope objective is used to focus the beam to a spot size of 1 to 2 \( \mu \)m diameter. The sample is mounted on nanopositioners inside a continuous-flow liquid-helium cryostat. The photoluminescence (PL) is collected in reflection geometry back through the same microscope objective, imaged with a f-number matching into a spectrometer, and detected by an InGaAs photodiode linear array.

Figure 1(a) shows that the ensemble QD PL spectra at 20 and 200 K are almost the same, except band gap shrinkage shifts the PL peak from 1.020 to 0.982 eV. Because of the very large inhomogeneous linewidth due to the one distribution, the absorption and PL spectra are not expected to change much with temperature.

The idea behind Figs. 1(b) and 1(c) is to determine the effect of absorption and gain on the cavity \( Q \) by measuring the nanocavity mode linewidth as a function of temperature [Fig. 1(b)] or of fabrication parameters [Figs. 1(b) and 1(c)]. As the temperature is increased from 20 to 200 K, the cavity peak \( E_{cav} \) shifts to lower energy by 8.2 meV compared with the much larger 36 meV shift of the QD ground state peak \( E_{gs} \). Therefore, temperature scans the detuning, \( E_{cav} - E_{gs} \), permitting a portion of the linewidth versus detuning curve to be determined. By using five nanocavities, labeled 1 to 5 in Fig.
FIG. 1. Quantum dot emission spectra and cavity linewidths. (a) Ensemble QD emission spectra at 20 and 200 K versus energy relative to the ensemble QD ground-state transition peak $E_0$. (b) Nanocavity FWHM linewidth for several temperatures between 20 and 200 K versus the detuning of the nanocavity mode peak from the ensemble QD ground-state transition peak. The solid squares are for low-power ($2-10 \mu W$) cw excitation, and the open circles are for high-power ($720 \mu W$) cw. Five nanocavities were selected that cover the energies of the QD ground-state and first-excited-state transitions, with the detuning increasing with temperature. All five have $r/a=0.27$ and $s/a=0.20$ and were from the same fabrication run. The data points and guide-to-the-eye solid curves show that the low-power linewidths follow the expected ensemble QD absorption spectrum. But the high-power linewidth drops below the empty-cavity straight line, indicating the presence of gain. (c) Nanocavity FWHM linewidths for low temperatures around 20 K versus detuning. The nanocavities were selected from two different fabrication runs and have a wider range of lattice constants, radius, shifts, and doping values than those in (b). Solid squares: $2-20 \mu W$ cw; open circles: $1 \text{mW}$ diode-laser peak power. The solid curves are the same ones in (b) with a slight shift upward to take account of the higher average empty-cavity linewidth. The behavior of the linewidth is clearly similar to that in (b).

1(b), and choosing different fabrication parameters such as $r$-beam doping, hole radius $r$, lattice constant $a$, and shift $s$ of “spacer” end holes (but always keeping $r/a=0.27$ and $s/a=0.20$), the ground-state and first-excited-state spectral region was covered. The top set of data and guide-to-the-eye curve in Fig. 1(b) show that the low-power linewidth spectrum has the shape expected for the QD ensemble absorption with peaks at both the ground-state and first-excited-state absorption transitions. This is essentially an intracavity measurement of the absorption's spectral profile. Both ensemble and single quantum dot absorptions have been measured previously. The absorption spectrum extracted from our linewidth data is much like that obtained from the photorefection in a waveguide containing InAs QDs (see Fig. 1 in Ref. 8). They do not find a shift between PI and absorption peaks, and we doubt that the apparent shift in our Fig. 1 is statistically significant.

One can also keep the temperature fixed at 20 K and use the fact that the nanocavity wavelength depends upon fabrication parameters; see the upper data points in Fig. 1(c). It is not surprising that this more random selection of nanocavities has greater variations in $\Delta E_{\text{absorption}}$, but the behavior is the same as Fig. 1(b). Clearly the QD ensemble absorption controls the linewidth $\Delta E$ (and therefore $Q=1/\Delta E$ of our nanocavities, except at the low-energy tail of the distribution where the absorption is small. For these data, the input power was kept low enough that saturation of the QD absorption was negligible. Uncoupled or poorly coupled QDs have longer radiative lifetimes than QDs with Purcell enhancement or strong coupling, therefore, they can be saturated by pumping hard enough— their emission rate becomes limited by their longer lifetimes. Note that the linewidths and $Q$s were the same for QD excited-state pumping as for pumping into the GaAs.

The empty cavity linewidth, determined from the large negative detuning points in Fig. 1(b) for which absorption is expected to be negligible, is about $0.05 \text{meV}$ (corresponding to $Q_{\text{empty}}=20,000$). One expects $Q_{\text{empty}}=1/Q_{\text{design}} + 1/Q_{\text{groundstate}}$, $Q_{\text{design}}$ (computed to be $40,000-50,000$ here) accounts for design losses that are present even if the fabrication is perfect, whereas $Q_{\text{groundstate}}$ comes from additional scattering losses due to fluctuations introduced by growth and etching. Since neither $Q_{\text{design}}$ nor $Q_{\text{groundstate}}$ is expected to vary much over this wavelength range, a straight line is drawn all the way across Fig. 1(b) at $0.05 \text{meV}$ to indicate $\Delta E_{\text{empty}}$.

How do these findings affect the search for QD nanocavities that exhibit strong coupling? Since the dot-nanocavity coupling $g$ must exceed the cavity loss rate $\kappa=2\pi \times 2 \text{m} / Q$, it is $g/(2\kappa)=Q/3V$ that needs to be maximized for strong coupling. We find that at the low powers required for strong coupling, the QD ensemble absorption is large enough to reduce the $Q$ by at least a factor of 2 for QD transitions near the top of the ensemble distribution or above it. From this perspective, cavities near the low-energy tail of the QD spectrum should present a better opportunity for the observation of strong coupling due to much lower QD absorption and higher $Q$s. However, the low absorption corresponds to a lower density of QDs, meaning that for a particular cavity, there is a lower likelihood of finding a QD which has the spatial and spectral coincidences necessary for strong coupling. Temperature scanning can change the dot-nanocavity detuning by about 0.7 nm before photon broadening prevents strong coupling. Recently, we have shown that condensation of nitrogen or...
xenon can give much larger shifts (~4 nm), making it easier to find the required coincidences using lower QD densities.\textsuperscript{11} In another approach (that did not achieve strong coupling), a photonic crystal nanocavity was fabricated around a particular QD, and excitation was used to scan the nanocavity peak digitally in 3 nm steps.\textsuperscript{11}

Above, we have studied the cavity linewidth and $Q$ for low-power excitation appropriate for strong coupling and showed that indeed the ensemble absorption of a single layer of high-density QDs is detrimental. A related investigation can be performed to determine if the single-layer of QDs can produce gain and lasing when excited sufficiently strongly. The lower set of data in Fig. 1(b) is taken with high-power cw excitation for which the electron and hole ground states of most of the QDs in the ensemble are occupied; gain is apparent over the range ~30 to +10 meV detuning where the linewidth drops well below $\Delta\nu_{\text{cav}}$. Even the linewidths for QDs in the lower-energy part of the first-excited-state transition drop down almost to $\Delta\nu_{\text{cav}}$. Moreover, in Fig. 1(c), the lower data points for high-power diode-laser pumping show pronounced gain, similar to Fig. 1(b).

The QD nanocavity $Q$ and emission intensity as a function of the value of the peak input diode-laser power are shown in Fig. 2; a low duty factor is used to avoid heating that prevents lasing. Generally, cavities with modes lying in the upper half of the ensemble QD lowest-energy transition, where there is no gain in Figs. 1(b) and 1(c), exhibit output versus input curves like the "nearly-lasing" curves in Fig. 2. In this case, the output increases sublinearly and $Q$ saturates at $Q_{\text{sat}}$. This implies that the QD absorption is saturated at high powers, but there is no lasing. For cavities with modes in the lower half, where there is gain in Figs. 1(b) and 1(c), the output versus input resembles the "lasing" curves in Fig. 2, where the output increases superlinearly, and $Q$ reaches $\approx 40,000$ (determined by the spectrometer array resolution). Clearly $Q_{\text{sat}}$ is exceeded because the gain from the pumped QD ensemble overcomes some or all of the cavity losses. The case that lasing is occurring is especially strong in Fig. 3, where the $Q$ saturates at 10% of the maximum pump power. As for the first photonic-crystal-nanocavity lasing using quantum dots,\textsuperscript{11} the lasing nanocavities here exhibit line-width narrowing and a threshold behavior for output versus input. The approximate power here is 100–200 $\mu$W, about the same as previously.\textsuperscript{11} However, the absorbed power in our case is five times less, due to only one layer of dots compared with their five layers of dots.

The threshold curves are "softer" as expected for a higher $\beta$ (where $\beta$ is the rate of enhanced spontaneous emission into the lasing mode divided by the rate of total spontaneous emission $\lambda_{\mu}$), leaving the threshold power ill-defined. For a SQD in the field maximum, the Purcell factor in $F_p = 3\lambda_{\mu}(2\pi\Delta\nu_{\text{cav}})^{-0.9}$ here, provided the SQD linewidth is less than the cavity linewidth (a certainty here for low temperature and low-excitation power). Since $\beta = (F_p - 1)/F_p$, then $\beta = 1$. Of course, most QDs are not in the field anti-node and the QD linewidth may exceed the cavity line-width at high-excitation levels, reducing the average $F_p$. But since the maximum $F_p$ is so large, $\beta$ must be very large, i.e., between 0.1 and 1.

In Ref. 4, it was concluded that about 80 QDs contribute to the lasing. The SQDs here are identical to those and have the same density in each layer, and both nanocavities have $\lambda_{\mu} = \lambda$. However, there is a single layer of dots compared with five layers in Ref. 4, so the number of contributing SQDs is no more than 16 here. In addition, the lattice temperature here is 10–20 K, compared with room temperature; therefore, the QD linewidth should be narrower than their 7 meV. By choosing SQDs in the 1080–1092 nm spectral region, we can see individual QD transitions and measure their linewidths as a function of diode-laser peak power. For 100–200 $\mu$W peak powers (corresponding to threshold), the QD linewidth is about 0.09 meV. From Fig. 9 of Ref. 4, one then concludes that a SQD with such a linewidth should be able to lase provided the $Q$ exceeds 1000, which is clearly the case here. Studies of lasing with an isolated SQD will be highly interesting, and the conditions here are close to those needed.\textsuperscript{4} However, SQD lasing is not claimed here, because the threshold behavior is relatively insensitive to detuning and temperature and does not change when a nanocavity is used that exhibits a strong coupling anti-crossing at low power. Most likely the gain, not from a SQD, but from several QDs in the ensemble mostly situated in the outer reaches of the cavity mode field is responsible for the lasing seen.

FIG. 2. Quality factor $Q$ and nanocavity output emission intensities versus diode-laser peak pump power. (a) The $Q$ of nonlasing nanocavities saturates at the empty-cavity $Q$, but the $Q$ of lasing cavities reaches the instrument-limited $Q$ of about 40,000. Correspondingly, both the time-averaged total (b) and peak (c) output emission energy exhibit a threshold-like behavior for the lasing cavities and a saturation behavior for the nonlasing cavities.
FIG. 3. Nanocavity $Q$ versus diode-laser peak pump power. The $Q$ of this nanocavity reaches the instrument-limited value of 40 000 at roughly 10% of the maximum input power.

here. Additionally, phonon-assisted scattering from the dot ensemble into the cavity mode could increase the number of contributing dots. Clearly, a lower density of QDs in the present nanocavity would increase the chance of observing SQD lasing.

Strong coupling was observed at power levels that we refer to as low power. The strong coupling anti-crossing, a SQD effect, could be seen even though the $Q$ was determined by an ensemble QD effect, namely the absorption of many partially saturated QDs. Both effects can occur simultaneously. For strong coupling, the QD must be situated near an anti-node, i.e., within an area of the slab of only $0.15 \mu m^2$. The product of that area and the dot density of $400 \mu m^2$ gives 60 QDs spread out over the 42.5 meV linewidth. Therefore, there is a 0.7 meV separation on average between QDs that could potentially couple strongly. However, if one computes the field distribution for our photonic-crystal-slab nanocavity, the field is nonzero over several $\mu m^2$. This implies that there are many more QDs that can influence the linewidth, $Q$, and emission properties of the nanocavity, and they are spaced much closer together spectrally. The absorption of background ensemble of QDs reduces the $Q$ at low power, and the gain of this ensemble leads to lasing. Emission from this ensemble enables the nanocavity peak to be seen even when the SQD is greatly detuned. It is intriguing that in principle an isolated SQD in our present nanocavity could exhibit lasing, but effects of the ensemble will have to be reduced considerably to see SQD lasing.

In summary, we have shown that the $Q$ of a GaAs photonic-crystal-slab nanocavity is now high enough that a single layer of high-density quantum dots introduces enough absorption to appreciably lower the $Q$, impeding the search for strong coupling, and enough gain for lasing.

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APPENDIX D

GaAs photonic crystal slab nanocavities: Growth, fabrication, and photon storage
time

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GaAs photonic crystal slab nanocavities: Growth, fabrication, and quality factor

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Abstract

In an effort to understand why short wavelength (~1000 nm) GaAs-based photonic crystal slab nanocavities have much lower quality factors (Q) than predicted (and observed in Si), many samples were grown, fabricated into nanocavities, and studied by atomic force, transmission electron, and scanning electron microscopy as well as optical spectroscopy. The top surface of the AlGaAs sacrificial layer can be rough even when the top of the slab is smooth; growth conditions are reported that reduce the AlGaAs roughness by an order of magnitude, but this had little effect on Q. The removal of the sacrificial layer by hydrogen fluoride can leave behind a residue; potassium hydroxide completely removes the residue, resulting in higher Qs.

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The small volume of a photonic crystal slab nanocavity makes it attractive for low-threshold lasing [1] and, with high-quality-factor designs [2], for semiconductor quantum optics, especially strong coupling between a single quantum dot and a single cavity mode [3–9]. The quality factor Q (cavity frequency ω divided by FWHM cavity linewidth) in a few cases has exceeded 15,000 [3,8,10,11] for fabrications on samples intended for strong coupling. More typically it is less than 10,000, particularly for the wavelength λ in the range 500–1000 nm where Si detectors greatly improve measurements. Very high-Q values have been computed for various cavity designs, but fabricated Qs are always much higher in silicon than in the GaAs system. In fact, a Q of 45,000 was already reported [2] in silicon for the L3 design used in [3] and throughout this article; optimization of that design has yielded 150,000 computationally [12] and 110,000 experimentally in Si (1.5 µm) [13]. This paper reports that whereas reducing the roughness of the top of the AlGaAs sacrificial layer did not improve Q noticeably,
using potassium hydroxide (KOH) to remove debris left behind in etching away the AlGaAs sacrificial layer did increase $Q$ appreciably.

1. Reducing roughness at the top of the AlGaAs sacrificial layer

One of the diagnostics that can be used to evaluate a sample before fabrication is to scan the surface by atomic force microscopy (AFM). Scans (40 $\mu$m x 40 $\mu$m) of our samples typically gave root mean square (RMS) values around 1 nm, indicating smooth top surfaces consistent with scanning electron microscopy (SEM) images of the fabricated nanocavities. Cross-section transmission electron microscopy (TEM) images of the sample used for our initial observation of vacuum Rabi splitting [3] revealed that the AlGaAs/GaAs interface is rough; see Fig. 1. TEM images of three of our samples grown for fabrication in the 900–1000 nm wavelength range looked almost as rough. The smoothing accomplished by growth of the first few monolayers of GaAs helped to conceal this problem, as shown in Fig. 1 – which also shows that the roughness is much larger along [1 1 0] than along [1 1 0].

What characteristics must the sacrificial layer have? The Al content $x$ of the Al$_x$Ga$_{1-x}$As needs to be in the range between 0.55 and 0.90 for etching with hydrofluoric (HF) acid. For $x > 0.90$ the etch rate is so fast that it is impractical to use an HF etch, steam oxidation of the AlGaAs in a furnace permits the use of a KOH wet etch, but it also introduces another step in fabrication which can be avoided by choosing a lower Al content. For $x < 0.55$ the etch rate is impractically slow. The AlGaAs needs to be thick enough to prevent light leakage to the substrate once the sacrificial layer has been removed; a thickness of 800–1000 nm is often used for 900 < $\lambda$ < 1200 nm of interest here. The growth of smooth AlAs layers in Bragg mirror structures in vertical cavity surface emitting lasers (VCSELs) and planar microcavities is easier, the AlAs layer is typically only 70 nm thick and alternates with a similar thickness GaAs layer which smoothens any roughness at the top of the AlAs layer. The growth of smooth Al$_x$Ga$_{1-x}$As in heterojunction lasers is also easier because $x$ rarely exceeds 0.4.

Once the AlGaAs roughness was discovered, a quick and effective approach was taken to optimize the molecular beam epitaxy (MBE) growth: stop after growing Al$_x$Ga$_{1-x}$As, immediately remove the sample, and scan it by AFM. Growth parameters investigated included introducing a growth interruption to give time for smoothing under As, growing a thin layer of GaAs, and using a misoriented substrate. Some structures can be grown as well on a flat substrate as on one polished with the normal to the surface tilted a few degrees toward a particular crystal axis. However, it is known that the growth of AlGaAs is preferentially along steps edges lying along [1 1 0]. If the surface is perfectly flat, then the surface diffusion may be inadequate to reach such an edge and island formation and 3D growth can result. This is consistent with Fig. 1 where it was found that the AlGaAs surface is rougher along [1 1 0] than along [1 1 0]. Therefore, if one has not yet identified the ideal conditions for growth on a flat substrate, growth on a tilted substrate may be better. Consequently, growth on (0 0 1) GaAs substrates misoriented by 2$^\circ$ toward [1 1 0] was tried. As summarized in Table 1, several samples grown under approximately the same conditions showed that using a tilted substrate instead of a flat substrate and increasing the number of monolayers of GaAs always
Table 1

<table>
<thead>
<tr>
<th>Sample</th>
<th>x</th>
<th>Periods</th>
<th>AlGaN (nm)</th>
<th>GaAs (MLs)</th>
<th>GI (s)</th>
<th>Cu (%)</th>
<th>r.m.s. (nms)</th>
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<td>0</td>
<td>0</td>
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<td>10</td>
<td>100</td>
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<td>3.5</td>
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<td>120</td>
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</table>

Notes: “x” is Al concentration in AlGaN, “Periods” is the number of times the sequence AlGaN, GaAs, growth interruption was repeated; “AlGaN” is the thickness of AlGaN grown before the growth of GaAs of thickness “GaAs”; “ML” is monolayer = 0.205 nm followed by a growth interruption of duration “GI”; “Cu” is the substrate misorientation angle; the root mean square (RMS) surface roughness is “r.m.s.”, and values marked with an asterisk (*) indicate the top of the GaAs slab, not grown on the other samples. QD31, grown like the AlGaN in QD24, is representative of our AlGaNs before the present study.

...decreased the r.m.s. value. However, the GaAs thickness needs to be less than one monolayer (ML) for good etching of the sacrificial layer. It was also found that decreasing the ratio of the As flux to the sum of the Al and Ga fluxes from 26 to 13 decreased the roughness. The results discussed so far were for samples grown in Tucson using a Riber 32 MBE machine. The substrate temperature was 570-580 °C. The Al concentration x was changed by varying the Ga flux, holding the Al flux constant; the AlGaN growth rate was about 0.63 (0.80) ML/s for x = 0.75 (0.55). Even flatter AlGaNs were grown on a Riber Compact 21 MBE machine at the University of Karlsruhe (samples beginning with “A”) in Table 1. There the growth rate was about 0.858 ML/s calibrated by reflection high energy electron diffraction (RHEED) oscillations, and the As pressure was typically 8 x 10^{-4} Pa, roughly half that used in Tucson (1.5-2.1) x 10^{-4} Pa. RHEED patterns were observed several times during each growth to monitor flatness and to keep the V/III flux ratio close to but above the transition to Ga-stabilized surface structure, leading to flatter growth.

2. Q values of nanocavities with smoother AlGaNs

...having determined growth conditions for AlGaNs flatter by an order of magnitude, we began growing complete structures, i.e., slabs on top of the AlGaAs. For examples, see QD41 and A0961 in Table 1, where the r.m.s. values are now for the top of the GaAs slab. The TEM images in Fig. 2 verify that the AlGaN is much flatter than in Fig. 1. The nanocavity Q

![Fig. 2](image-url)
values of these samples were measured via their photoluminescence spectra with cw nonresonant excitation at 780 nm. The fabrication run A0961-3 yielded the $Q$ values plotted in Fig. 3; a $Q$ of 16,250, one of the highest $Q$ found, is shown in the inset. For comparison, the highest $Q$ value obtained on rough-AlGaAs samples for $\lambda < 1000$ nm was 9000, whereas it reached 20,000 at $\lambda \approx 1200$ nm [110]. Therefore the data in Fig. 3 spanning 200 nm are consistent with the disjoint rough-AlGaAs data. This agreement means that taking the trouble to grow smoother AlGaAs did not improve the $Q$ with our present fabrication quality. Fig. 3 shows little change in $Q$ for wavelengths longer than 1020 nm, suggesting that $Q$ may be limited by non-vertical holes or fluctuations of hole shapes rather than Rayleigh scattering from imperfections which should increase as $\lambda^2$. QD ensemble absorption and surface state absorption may cause the reduction in $Q$ for short wavelengths.

3. KOH dip

The results just described for A0961-3 were obtained after the fabricated sample was dipped in a KOH solution (25 g/100 ml of deionized water) for 140 s just before it was placed in the cryostat and evacuated. This last step was added after an inspection of the SEM micrographs (Fig. 4a) revealed the presence of a semi-transparent object partially covering two holes in the upper left of the micrograph. AFM scans (Fig. 5) revealed the density and height variation of the debris and confirmed the hypothesis that this debris had floated and settled on the top of the slab after a successful dry etch. Speculating that this debris originated during the wet etch in the HF acid solution (1:10 = HF:H₂O by volume) and was probably a hydroxide of aluminum, we dipped the sample in a KOH solution. As is evident from the AFM scans, this process removed the debris completely. The effect of the KOH cleaning of surface debris on 10 different cavities was pronounced, showing an average improvement of 50%. One particular nanocavity from fabrication run A0961-3 showed a 79% improvement in $Q$ from 4500 to 7800. Photoluminescence intensity also increased substantially, and the cavity modes shifted to higher energy on average 11 meV for all KOH treated cavities; see Fig. 4b for typical data.
Fig. 5. AFM images of a few nanocavities of the fabrication run A0961-2 after O₂ plasma cleaning and (a) before switching in a KOH solution (1.25 g KOH in 10 ml H₂O), (b) after 60 s, and (c) after 140 s. The bright yellow spots in (a) and (b) are debris that exceed 50 nm in height; the one in the bottom left corner of (a) exceeds 600 nm.

4. Summary

It was discovered by TEM that the top of the high-Al AlGaNAs sacrificial layer in many of our MBE samples grown for fabrication of photonic crystal slab nanocavities is rough — even though the top surface of the sample (the top of the GaN slab) is relatively flat, as had already been determined by AFM. MBE growth conditions were found that decrease the AlGaNAs roughness by more than an order of magnitude. The improved smoothness will eventually be important for fabrication of high-Q nanocavities although its contribution at present is small. In addition, the efficacy of KOH in removing residual left behind in etching away the sacrificial layer is demonstrated.

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References


APPENDIX E

Modelling and fabrication of GaAs/InAs photonic-crystal cavities for cavity electrodynamics


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Modelling and fabrication of GaAs photonic-crystal cavities for cavity quantum electrodynamics

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Abstract

In this paper, we present recent progress in the growth, modelling, fabrication and characterization of gallium arsenide (GaAs) two-dimensional (2D) photonic-crystal slab cavities with embedded indium arsenide (InAs) quantum dots (QDs) that are designed for cavity quantum electrodynamics (cQED) experiments. Photonic-crystal modelling and device fabrication are discussed, followed by a detailed discussion of different failure modes that lead to photon loss. It is found that, along with errors introduced during fabrication, other significant factors such as the presence of a bottom substrate and cavity axis orientation with respect to the crystal axis, can influence the cavity quality factor (Q). A useful diagnostic tool in the form of contour finite-difference time domain (FDTD) is employed to analyse device performance.

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1. Introduction

Photonic-crystal cavities have come a long way from the initial conceptualization of photonic crystals [1, 2]. Sustained efforts in semiconductor growth, lithography and etching techniques have paved the way for the creation of high-Q, low mode volume (V) optical resonators that are ideally suited for exploring effects in quantum optics such as Purcell enhancement [3], vacuum Rabi splitting or strong coupling [4], and photon anti-bunching [5].

In our experiments, optical resonators are constructed by perturbing a 2D photonic crystal comprised of a triangular lattice of air holes in a thin slab of GaAs (see figure 1(a)), and embedded InAs QDs serve as internal light sources. Typical resonator designs involve the creation of a defect by omission of one or more holes and/or the modification of the immediate neighbourhood of the missing air hole(s). Figure 1(b) demonstrates one such tuning process to get high Q/V resonant modes, in which two air holes are shifted by x [6]. In such a geometry, horizontal in-plane optical confinement is provided by the photonic bandgap [7] which can be nearly perfect as long as the cavity is surrounded by sufficiently many layers of photonic crystals. Incomplete vertical confinement [7] happens on account of index contrast between the slab and the surrounding vacuum and is the primary cause of photon loss. The slab thickness is chosen such that only the lowest-order optical mode in the vertical direction is supported, thereby keeping the mode volume at a minimum and suppressing the coupling to the higher-order slab modes. Therefore, the slab thickness tends to be approximately close to half the wavelength of light in the material for optimal confinement.

This paper is dedicated to the modelling and fabrication of these devices, followed by a detailed investigation of the important channels for optical loss in these devices. Using three-dimensional (3D) FDTD methods and contour information extracted from the scanning electron micrograph (SEM) of a fabricated device, it is shown how various optical properties can be accurately analysed. Then, two important distinctive mechanisms of Q degradation will be emphasized: the presence of a bottom substrate and crystal-axis-dependent surface roughness resulting from the epitaxial growth process. As shown in the schematic in figure 1(a), a typical epistucture for GaAs photonic-crystal slabs includes an optically flat bottom substrate. The sacrificial layer thickness is of the order
Figure 1. (a) Convergent of a photonic-crystal slab cavity. (b) Top view of the L3 cavity design. a, r, s are the lattice constant, radius and hole shifts, respectively. (c) In-plane (centre of slab) electric-field energy densities for three typical cavity designs: L3 (3 holes missing, shifted end holes), L1 (one missing hole, shifted and shrunken nearest-neighbour holes) and L0 (no missing holes, only two shifted holes).

of ~1 μm, which has been chosen to be sufficiently larger than the vanishing tail of photonic-crystal cavity modes. However, it is still comparable to the free space emission wavelengths of InAs QDs used in this work. As a result, interesting interference effects originate from air-gap size-dependent resonances. The fabrication of these devices is fairly involved and challenging, and in our experiments we have come across several failure modes. Here, an attempt is made to catalogue these, and to suggest solutions where possible.

2. Photonic-crystal cavity modelling

For all the elegance of photonic crystals, a completely analytical description is illusory and hence these devices must be analysed numerically. Frequency-domain simulations [8] of the defect-free photonic-crystal slab reveal the extent of the photonic bandgap as a function of the in-plane Bloch wavevectors, as seen in figure 2. A subtlety must be mentioned at this point: since the structure only has 2D periodicity, light is (incompletely) confined in the vertical direction by total internal reflection. Therefore, any mode designated by (ω, k) lying above the light line, v₀ = c (ω, k) (where c is the speed of light and in-plane wavevector, respectively) will always couple to the continuum of vacuum electromagnetic (radiation) modes. Thus, the photonic bandgap in this case is defined as the frequency range that is devoid of any guided modes [9] below the light line (i.e. for ω < c k) (radiation modes will exist at all frequencies). It should be noted that all guided bands can be divided into two groups depending on the mirror symmetry with respect to the plane in the middle of the slab; even and odd (i.e. equivalently, transverse electric (TE)-like or transverse magnetic (TM)-like, to invoke the similarities to their 2D counterparts). In the case of the triangular lattice of air holes it is well known that a bandgap exists only for TE-like modes, and this can be seen in figure 2. In section 4.1.3 this characteristic of the photonic-crystal band structure will be revisited to explain an in-plane loss channel.

While frequency-domain simulations reveal highly accurate eigenfrequency and modes, they cannot be used for time-dependent phenomena, such as Q calculations, and it is here that 3D FDTD simulations [10, 11] must be employed. Using these tools, we have modelled, fabricated and characterized
Table 1. Parameter summary of typical cavity designs obtained by 3D FDTD code, $r$ refers to the radius of the perturbed holes, $\lambda$, $V$, $n$ refer to the mode wavelength, mode volume and slab refractive index, respectively.

<table>
<thead>
<tr>
<th>$r$ (nm)</th>
<th>$\lambda$ (nm)</th>
<th>$V$ ($\mu$m)</th>
<th>$n$</th>
</tr>
</thead>
<tbody>
<tr>
<td>LO</td>
<td>0.75, 0.90, 0.14, 0.30</td>
<td>0.287, 0.7109, 0.122</td>
<td></td>
</tr>
<tr>
<td>LI</td>
<td>0.55, 0.35, 0.10, 0.25</td>
<td>0.300, 0.32740, 0.422</td>
<td></td>
</tr>
<tr>
<td>LJ</td>
<td>0.75, 0.90, 0.17, 0.30</td>
<td>0.247, 0.67315, 0.392</td>
<td></td>
</tr>
</tbody>
</table>

several cavity designs that feature high $Q$s and low $V$s, 1.3 [6], 1.1 [12] and 1.0 [13] cavity geometries (see figure 1(c)). Table 1 summarizes the various salient features of these cavities.

The regime of strong coupling [14] is entered when the coupling strength, $g$, between the QD and the cavity mode exceeds the average of the individual decay rates of the QD, $\gamma_q$, and the cavity mode, $\kappa$, i.e., $2g > (\gamma_q + \kappa)/2$. The motivation to choose a high $Q$ comes from the fact that $\kappa = 1/Q$, while the need for a low $V$ comes from the fact that $g \propto 1/\sqrt{V}$. To apply these cavity designs for cQED, special care must be taken to optimize both spatial and spectral overlap between a single QD and a cavity mode. Specifically, the placement of a QD at positions very close to the GaAs surfaces must be avoided, since coupling to surface states [15] will lead to strong non-radiative losses. In light of these considerations, the L3 model which has the highest electric-field intensity at the cavity centre (see figure 1(c)) seems to be the most promising candidate for cQED experiments.

3. Fabrication and optical characterization

The GaAs material in which these resonators are fabricated is grown by molecular beam epitaxy (MBE). Details of this growth are provided elsewhere [16]. We typically start with a blank (001)-oriented GaAs wafer, onto which a 300 nm GaAs buffer is grown, followed by a 1 µm thick sacrificial layer of the type Al$_x$Ga$_{1-x}$As (with $0.7 < x < 0.94$), a 90 nm GaAs bottom slab, an atomic monolayer thick wetting layer on which form InAs QDs, and finally a 90 nm GaAs top slab. It must be noted that, in the above-described scheme, dot growth is strain-induced and hence has a spatially probabilistic distribution. Further, minute variations in QD dimensions across the wafer give rise to a spectral distribution of emission wavelengths, typically a 5% spread about a 1 µm central wavelength. It is worth mentioning here that the QD has an electric-field polarization that is primarily in-plane and thus couples to the TE-like (even) modes of the photonic crystal.

Fabrication of photonic-crystal cavities involves the following steps: cleaning of the top of the grown GaAs wafer surface, spin-coating and baking of an electron-beam resist, electron-beam lithography and subsequent resist development, pattern transfer into the substrate using a chemically assisted ion beam etch (CAIBE), removal of the underlying sacrificial layer by a wet etch and a final strip of the remaining resist.

The first step in device fabrication is the removal of any organic material from the top GaAs surface using acetone and isopropyl-alcohol (IPA). Any GaAs surface that has been exposed to atmospheric oxygen forms a thin oxide layer and this is removed by immersing the wafer in a dilute solution of hydrochloric acid (HCl) (1:1 = HCl:deionized water (DI) by volume) for 10 min. After blow drying with N$_2$ gas and heating on a hot plate at 170°C for at least 10 min, the wafer is ready for the spin-coating of a thin layer of resist. Targeting a thickness of 150 nm, a high molecular-weight positive electron beam resist (950 K poly-methyl-methacrylate (PMMA) in isopropyl alcohol) is spun on to the wafer and baked on a hot plate kept at 170°C for 30 min. Electron-beam lithography is then performed, using a Vistech EBPG-5000 | 100 kV machine that is able to expose the device masks at a resolution of 2.5 nm. Employing a low beam current in the region of 800 pA accomplishes the proper exposure of the device masks. The exposed wafer is developed for 1 min in a solution of 1:3 - methyl-isobutyl-ketone (MIBK):DI, followed by a 30 s rinse in IPA and a gentle N$_2$ blow dry. The developed devices are inspected in an SEM to confirm proper exposure and adequate development.

The developed devices are then dried etched in a CAIBE for 3 min. The etcher uses a Kaufman ion source that ionizes a 4 standard-cubic-centimetres-per-minute (sccm) flow of argon gas at a beam voltage and current of 600 V and 25 mA, respectively. Cl$_2$ gas is injected into the chamber just above the sample at a flow rate of 7 sccm. At the completion of the etch, the sample is immediately wet etched in a dilute solution of hydrochloric acid (HF) (1:1 = HF:DI by volume) for 60 s, rinsed in DI and IPA for 60 s each and allowed to dry. The sample is then agitated in a solution of IPA, chloroform and acetone (1:1:1 by volume) for 10 min to strip the resist. This removes all of the resist, except for the regions around the devices. The sample is then exposed to a gentle oxygen plasma treatment for complete resist removal. An Oxford Instruments ICP-RIE 180 is used to perform this treatment for 90 s, employing an O$_2$ flow rate of 90 sccm in a chamber at 10 mTorr, a low RF field (1 W) and a very high ICP power (600 W) that results in a gentle O$_2$ plasma that keeps the DC bias below 20 V. This is important in minimizing any surface damage that can be caused by the procedure.

The fabricated devices are placed in an evacuated liquid helium cryostat, cooled down to approximately 10 K and excited by a non-resonant Ti:sapphire laser operating at 780 nm with an approximate power of 500 µW. Photoluminescence from excited cavities is collected by a (36X) microscope objective, passed through a spectrometer and recorded on either an s detector (for wavelengths > 1 µm) or an n incouet detector (for wavelengths < 1 µm). Figure 3(c) shows the photoluminescence observed from a high $Q$ cavity. A Lorentzian fit is used to calculate the $Q$.

Through careful $Q$ measurements on a large set of fabricated devices, it is found that the measured $Q$ values are not as high as computed, even though fabricated devices look nearly perfect in all aspects: side-wall roughness, circularity of holes, etc. (see figure 3(b)). The following sections will be devoted to discussions of possible factors limiting $Q$ based on practical considerations.
4. Quality factor considerations

As has been discussed elsewhere [17], the \( Q \) of a device can be decomposed into the following factors:

\[
Q^{-1} = Q_{\text{rad}}^{-1} + Q_{\text{ext}}^{-1},
\]

where \( Q_{\text{rad}}^{-1} \) indicates the optical loss from the cavity by coupling to radiation modes in vacuum and \( Q_{\text{ext}}^{-1} \) represents a combination of the intrinsic loss in the material, the loss due to the formation of surface states that inevitably result from the oxygen termination of broken dangling GaAs bonds at the etched interfaces and any gain or loss [18] that might occur due to QD absorption in the cavity's spectral and spatial neighborhood. The loss represented by \( Q_{\text{ext}}^{-1} \) is strongly wavelength-dependent. On the one hand, GaAs shows increased loss [17] due to (sub-bandgap) surface states as one approaches the GaAs band edge at \( \approx 814 \) nm (at 10 K). Simultaneously, at wavelengths close to and lower than the QD ensemble peak (typically between 950 and 1100 nm), absorption by the QDs and the wetting layer further degrades \( Q \) from the 'empty' (no QD) cavity \( Q [18] \). \( Q_{\text{ext}}^{-1} \) can be further divided into two terms, \( Q_0^{-1} \) and \( Q_{\text{rad}}^{-1} \), where \( Q_0^{-1} \) indicates the intrinsic radiation loss in the absence of any fabrication-related error while \( Q_{\text{rad}}^{-1} \) is the loss due to (Rayleigh) scattering from surface imperfections introduced during MBE growth and device fabrication. To start with, fabrication-related failure modes that are fairly evident are presented, before moving on to \( Q \)-degrading factors that might not be obvious.

4.1. Factors related to the fabrication failure modes

4.1.1. Irregular shapes and positions of the air holes: \( Q_0 \) is optimized by a careful control of the cavity geometry and, in the case of L3 design, can be dimensionally as high as 70,000 in GaAs. (Higher \( Q_0 \) have been reported with photonic-crystal waveguide-type designs, but they are not considered here because of their higher mode volumes (calculated [19] as \( V = \frac{\int_P \sigma(E) E^2 d\varepsilon}{\int\sigma(E) E^2 d\varepsilon} \)) max \(\sigma(E) E^2 \)). Any irregularities in the lithography or etching can severely degrade \( Q_0 \). A poorly focused laser beam can be a major cause of lithographic irregularities, with the photonic-crystal holes deviating from circular shapes, as well as exposing areas larger than intended. Unless carefully controlled, the lithography step can also lead to considerable lack of reproducibility. Figure 4(b) illustrates the above-mentioned effects featuring non-circular and larger-than-intended holes which lead to the merging of holes that are closer than the lattice constant.

4.1.2. Remnant PMMA and debris. A thin layer of resist, as seen in figure 4(c) (dark, wrinkled features), is often left behind even after chemical treatment for removal. In our experience, this last layer of resist can only be removed by an oxygen plasma treatment. Moreover [16], a fine layer of sub-micron-sized microcrystallites is sometimes left behind after the completion of resist removal. These particles were speculated to be a hydride of aluminium that formed during the HF wet etch and floated from the sacrificial layer and ultimately were deposited on the top of the remnant resist. A 150 s dip in a solution of potassium hydroxide (KOH) (25 g/100 ml DI) was effective in completely removing this debris to restore the top surface to near atomic smoothness. In terms of device performance, \( Q_0 \) were found to increase [16] on average by 50% after this KOH treatment, indicating that the debris was a very significant source of scattering loss. In subsequent fabrication runs, this KOH step was incorporated in the fabrication sequence right after the HF wet etch. Figure 4(a) shows a tilted view of a surface with debris prior to the KOH treatment and figure 4(b) shows a fully fabricated device. Finally, roughness and ion-induced side-wall damage are introduced to the surfaces during the CAIBE etch which can further contribute to scattering. It is not possible to quantify the contribution of the latter source of scattering at this point, as the surfaces seem fairly smooth even in high-resolution SEMs.

4.1.3. Non-vertical side-walls. Maintaining a vertical etch profile in the dry etch is also important [20]. As explained in section 2, the presence of mirror symmetry in the ease of perfectly vertical side-walls allows one to classify slab modes into even and odd symmetry modes. However, non-verticality of etched air holes breaks this symmetry, which results in new forms of hybridized modes that can no longer be classified as...
either even or odd. A deviation from a vertical etch by even \(2^\circ\) can cause \(Q_{\text{rad}}\) to drop by an order of magnitude and this is due to the well-known TE-TM coupling loss [21]. There are at least two factors that can cause non-vertical side-walls. Firstly, inadequate or excessive resist development can give rise to non-vertical resist side-walls, which in turn are transferred to non-vertical side-walls in the GaAs substrate. Secondly, a non-vertical flow of Cl\(_2\) gas onto the substrate during the dry etch in the CAIBE can lead to asymmetric and non-vertical side-walls. Small-angle deviations \(\theta (\leq 2^\circ)\) from the vertical are hard to measure with the available resolution in an SEM, making it difficult to precisely quantify this loss channel. In general, it is easier to control etch symmetry and verticality using plasma-etch, such as in an inductively coupled plasma-reactive ion etcher (ICP RIE). Additionally, the use of photonic crystals with a complete photonic bandgap for both TH-like and TM-like modes can reduce the severity of a non-vertical etch. Significant results have been achieved using reduced symmetry photonic crystals [22–24], although in general, the spectral extent of the TM-like bandgap is much less than that of the TE-like counterpart.

4.2. Effect of a bottom substrate

Removal of the AlGaAs sacrificial layer below the photonic-crystal slab creates an air gap, exposing an optically flat surface. It is important for this air gap to be at least greater than half the vacuum wavelength to reduce optical loss into the substrate. It should be noted that reflectivity at the GaAs substrate is \(\sim 30\%\). Therefore, the effect of a bottom substrate is essentially that of a reflector below the photonic-crystal mode. For an air-gap size larger than half the vacuum wavelength, there can be multiple non-negligible reflections between the photonic-crystal slab and substrate. Thus, a fraction of the originally downward-emitted photons from the cavity are redirected upward by the bottom reflector to interfere with the originally upward-emitted photons. As a result, the far-field radiation pattern of the cavity mode is modified, changing the total emitted power [25, 26]. This is analogous to the well-known QED example of a point dipole in front of a mirror [27]. By changing the distance between the dipole source and the mirror, the original decay rate and radiation pattern are affected. Analogously, the \(Q\) of the cavity mode changes as a function of the air-gap size.

In figure 5 the \(Q\) of the L3 cavity mode is calculated by varying the air-gap size, \(t\). Even when \(t \geq 800\,\text{nm}\), \(Q\) varies by about \(\pm 5\%\) around \(Q_{0}\) (where \(Q_{0} = 67,315\) obtained in the absence of a bottom substrate). A larger variation in \(Q\) can be obtained by starting with a cavity mode that has a smaller \(Q_{0}\), since more radiative power will contribute to far-field interference. Through additional FDTD simulations, it is found that a \(Q\) variation larger than \(\pm 10\%\) is expected when \(Q_{0} \sim 50,000\). Therefore, the AlGaAs sacrificial layer thickness should be chosen carefully if \(Q\) is of primary concern in the design of photonic-crystal cavities.

4.3. Contour FDTD simulation

Although the theoretical cavity \(Qs\) can be as high as 67,000, less than half this number has been experimentally observed (see figure 3(b)). To investigate this discrepancy, the use of a 2D contour image extracted from the SEM image of a fabricated device (or the purposes of \(Q\) estimation is proposed.

Let us consider the particular case of a fabricated L3 cavity whose resonant wavelength and \(Q\) are measured to be 1141.1 nm and 10050, respectively. First, all the structural parameters \((r, r', s\) and \(a)\) characterizing the cavity are extracted from the corresponding SEM image (it should be noted that there can be \(\pm 5\%\) error in the SEM scale). 3D FDTD simulations based on these parameters reveal a resonant wavelength and \(Q\) of 1129.7 nm and 31418, respectively. Clearly, \(Q\) values estimated in this way differ significantly from the measured \(Q\).

Instead, contour data that faithfully captures all the fabrication-related imperfections (instead of using averaged cavity parameters from the SEM image) can be employed in
Figure 5. Variation of $Q_{0,4}$ with slab-to-substrate air gap for a slab of thickness 190 nm and a cavity mode at 1026 nm. (The other device parameters are the same as the L3 cavity shown in Table 1.) The dashed line corresponds to $Q_{0,4} = 67315$ when there is no substrate in the vicinity of the slab. All data is obtained by 2D-FDTD tools.

Figure 6. Electric-field intensity profiles from a contour FDTD simulation (contour data superimposed) for the three symmetry planes of the photonic-crystal cavity.

3D FDTD simulations [28]. As is evident from figure 6(c), the electric-field intensity distribution $|E|/|E_0|^2$ of the L3 mode reveals strongly localized patterns around the four air holes nearest to the cavity centre. Recalling the fact that the $Q$ of the L3 mode is highly sensitive to the fine tuning of the nearest air holes [6], even minute deviations from the ideal geometry can severely degrade $Q_{0,4}$ and this can be quantified using contour FDTD.

The extracted contour plot and simulated $|E(r)|^2$ for the structure mentioned above are shown in figure 6, in which a grid resolution of $\Delta x = \Delta y = \Delta z = 10$ nm is employed [10]. The $Q$ obtained in this way drops to 21283, which is still large but becomes closer to the measured value of 10056. The remaining discrepancy can be attributed to non-vertical air holes, side-wall roughness and material losses that cannot be incorporated in a contour FDTD simulation. Finally, for a more realistic $Q$ value, an air gap and GaAs substrate are included. The $Q$ value in the presence of a 800 nm air gap is 23576, showing about 10% improvement in comparison with the structure without the air gap.

4.4. Crystal-axis-dependent surface roughness

As we have recently reported [16], the underside of the GaAs slab can have wavy undulations that show an RMS roughness of the order of 25 nm, while maintaining an extremely smooth top surface. First by transmission electron microscopy, and then by atomic force microscopy (AFM), it was observed that the magnitude of the roughness was greater along the [110] direction than the [110] direction. To test whether this crystal-axis-dependent roughness had any bearing on cavity $Q$s, several pairs of identical cavities were fabricated with the cavity long axis (the line joining the x-slit holes in the case of the L3 designs) aligned along either of the directions mentioned above. It was found that, on average, cavities whose axis was aligned along the lower roughness direction had $Q$s that were 20–32% less (see figure 7(b)) than those aligned along the higher roughness direction. It must be noted that the overall $Q$s recorded in this experiment were low, on account of angled (non-vertical) walls of the photonic-crystal holes. Figure 7(a) shows the AFM scan of one such low $Q$ cavity. Growth improvements [16] have been detailed to ensure top and bottom slab surfaces with atomic smoothness and minimal contribution to scattering loss.

5. Conclusion

Several factors contributing to photon loss from photonic-crystal cavities designed for QED experiments are identified and addressed. Among them are irregularities with fabrication, crystal-axis-dependent losses and the presence of a bottom substrate. Lithography issues can be corrected by ensuring a properly focused electron beam, etched side-walls can be made vertical by careful control of the Cl$_2$ flow direction during dry etch; crystal-axis-dependent losses can be eliminated by optimized growth that leads to smooth slab interfaces; remnant
resist can be removed by an oxygen plasma; debris left behind by the HF undercut can be removed by treating with KOH.

Careful choice of sacrificial (AlGaAs) layer thickness can enhance cavity Q. The use of contour FDTD as a diagnostic tool for estimating cavity Qs is highlighted.

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References

APPENDIX F

In-situ annealing of InAs quantum dots on pre-structured GaAs substrates

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In-situ annealing of InAs quantum dots on pre-structured GaAs substrates

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(Dated: 29 June 2010)

We report on the effect of in-situ annealing on InAs quantum dots (QDs) grown site-selectively by molecular beam epitaxy on pre-structured GaAs substrates. We make use of surface sensitive Ga-assisted deoxidation to remove the native oxide prior to QD growth. We demonstrate that annealing combined with substrate patterning provides an additional tool to control the QD distribution and density. A morphological transition is observed where originally two QDs merge into one larger dot.

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Semiconductor quantum dots (QDs) have attracted a lot of attention due to their unique properties in the past two decades. They can act as single photon sources and generate entangled photon pairs.\textsuperscript{1,2} Combining single QDs and micro cavities such as photonic crystal slabs or micropillars offers the possibility to access and manipulate information at the quantum level. These systems provide a test bed for quantum information science\textsuperscript{3} enabling the study and exploitation of effects like quantum entanglement.\textsuperscript{4} Self-assembled QDs inherently preclude control of their location since the nucleation process is random. To precisely position QDs, different methods have been elaborated. Techniques such as electron beam lithography (EBL) have proven to define QD nucleation sites by pre-structuring the substrate.\textsuperscript{5} This process, however, commonly introduces defects at the regrowth interface which degrades the QDs’ optical properties.\textsuperscript{6} To compensate for that drawback a buffer layer (BL) is introduced and QDs are grown in stacks. Nevertheless, control of dot density and occupation number, i.e. QDs per site, was not considerably improved that way.

A different approach to access these quantities makes use of the fact that QDs undergo morphological changes during annealing. At lower temperatures they tend to ripen whereas they dissolve at higher temperatures.\textsuperscript{7,8} By choosing the right annealing conditions it should therefore be possible to control, to some extent, the final QD size as well as the QD distribution and the occupation number per site. Annealing studies on unstructured substrates have already confirmed an increase in QD size uniformity.\textsuperscript{9,10}

In our study we investigate the effect of annealing on site-selective InAs QDs grown on pre-structured GaAs substrates.

All samples were grown by molecular beam epitaxy (MBE) in a Riber Compact 21T with the sample being continuously rotated. Epi-ready (100) GaAs wafers are used as substrates and surface-patternning is performed on top of a 90 nm GaAs epitaxial layer. Conventional electron-beam lithography was used to define 50-70 nm wide holes in a PMMA/MA (polymethyl methacrylate/methacrylate) co-polymer resist on the surface. The holes were arranged on a square lattice on several arrays with varying lattice constants. The holes were etched 30 nm deep into the substrate by wet chemical etching (WCE) using H\textsubscript{2}SO\textsubscript{4}-H\textsubscript{2}O\textsubscript{2}-H\textsubscript{2}O (1:8:800 yielding an etch rate of ~1 nm/s). The resist was removed and the samples were cleaned in a series of solvent baths before being heated up to 130°C for 1 hour in the load lock chamber of the MBE system in order to get rid of volatile surface contamination.

The surface oxide was removed \textit{in-situ} by Ga-assisted deoxidation.\textsuperscript{11} Therefore, the sam-
amples were heated up to 480°C and exposed to a low Ga-flux of \( \sim 1 \text{ ML/s} \). Every 30 s of Ga-exposure was followed by a pause of 30 s such that the converted Ga\(_2\)O more easily desorbs from the surface. A total amount of 8 ML of Ga was provided before the substrate temperature was increased to 550°C and the samples were annealed for 2 min under As\(_2\)-atmosphere in order to thermally desorb remaining oxide compounds. A 16 nm GaAs buffer was then grown at 500°C followed by 1.7 ML of InAs. The growth rates for GaAs and InAs were determined as 0.3 ML/s and 0.07 ML/s with III/V beam equivalent pressure ratios of 1:10 and 1:100, respectively. In-situ annealing was performed right after QD growth. The samples were kept at growth temperature for 2:30 min and either rapidly cooled down to room temperature or capped with 80 nm GaAs. The uncapped samples were characterized by atomic force microscopy (AFM) whereas the capped samples allowed for micro-photoluminescence (\( \mu\)-PL) measurements, which were performed at 20 K with a He:Ne laser using a Si charge coupled device detector.

Holes with diameters ranging between 50-70 nm can be reproducibly defined with the existing electron beam setup. Although being round after exposure to the electron beam, the holes become rectangular after WCE. This change in morphology is due to selective etching of \{111\} GaAs sidewalls with the particular etch system in use. The hole shape ranges from square to rectangular with a prevalent elongation in the [011] direction.

Fig. 1 shows an AFM image of holes fabricated by WCE. The holes are arranged on a square lattice with a pitch of 500 nm. The average hole size is 58.4±6.1 nm. The linescan over one representative hole does not reveal its full depth since the AFM tip is probably too large. From previous calibrations of the etch rate a hole depth of about 30 nm is assumed.

After pre-structuring the sample is reintroduced into the MBE system. The surface oxide is removed with the Ga-assisted technique as described above. Deoxidation is performed at low temperature (480°C), inhibiting additional surface pitting which is observed for thermal deoxidation. The required amount of Ga to remove the surface oxide was calibrated by observing the surface configuration with a reflection high energy electron diffraction (RHEED) setup. The \( 2 \times 4 \) surface reconstruction appearing after 16 cycles of Ga-exposure is considered as an indicator for effective deoxidation. After short annealing under As\(_2\)-atmosphere, this particular reconstruction pattern is clearly visible.

In the next step a thin GaAs buffer layer was grown to compensate for surface roughness caused by WCE, particularly inside the holes. The BL growth is subject to limitations.
In general, the negative effect of surface roughness on the subsequent QD growth reduces with increasing BL thickness. However, the hole shape is influenced by the increasing buffer layer thickness with the hole becoming shallower and wider. Eventually, a single hole might develop into two separate holes if the thickness becomes too large. Since single QDs are desired this change in morphology should be avoided, which hence poses an upper limit on the BL thickness. An additional factor is the BL growth temperature, which controls the migration length of Ga adatoms. Infilling of holes is promoted at higher temperature. As a consequence, a trade off between these effects is required. We grew a thin BL of about 16 nm at moderate temperature on top of a pre-structured sample to retain the original hole shape as much as possible. This was followed by growth of InAs QDs. The amount of InAs being above the critical thickness, we expect QDs to nucleate on unstructured areas as well.

Fig. 2 shows AFM images of a set of samples with as grown (left) and annealed QDs (right). We observe in both cases QDs nucleating at locations pre-defined by the holes. The distance between two holes is 250 nm. The as grown sample shows predominant nucleation of double dots on each site. The linescan over one such pre-defined site clearly reveals the double-dot feature with the dots nucleating at the side of the hole. The center-to-center distance between these two particular dots is about 50 nm. The average diameter of the QDs is 49.1 ± 4.4 nm. As described before, we relate the occurrence of double dots to a possible change in hole shape during BL growth.

The annealed sample was fabricated the same way except additional in-situ annealing of 2.5 min after QD formation. We observe a morphological change of QDs with a tendency of original double dots to merge into single dots. The annealing step causes the material to redistribute by facilitating the migration of adatoms. The representative linescan shows that the size of the annealed QDs is increased (by a factor of 2 for the diameter) compared to as grown QDs, implying that ripening is dominant. The average QD diameter is 82.6 ± 9.5 nm. The volume of the single dots is larger than the combined volume of the double dots. According to the work of Hu et al., the observed transition of double dots to single dots is best described by a kinetic model with the ripening process being limited by attachment and detachment of atoms on the dot surface. In that context, our results suggest that the site-selective QDs are more stable than interstitial ones, i.e. the site-selective dots ripen and the interstitial ones disappear. The increased volume of the single dots is then related to the reduction of interstitial dots. This observation implies that the surface diffusion of atoms is
quite effective, at least on length scales of 100-200 nm. The application of the kinetic model is therefore justified.

To further ensure the above observations it is worth having a closer look at the material distribution. For the as grown sample, the QD densities are roughly $n_{\text{as grown}}^{\text{on-site}} \approx 4.8 \times 10^9 / \text{cm}^2$ and $n_{\text{as grown}}^{\text{off-site}} \approx 1.6 \times 10^9 / \text{cm}^2$ in the pre-structured (on-site) and unstructured (off-site, not shown) areas, respectively. For the annealed sample the densities are about $n_{\text{annealed}}^{\text{on-site}} \approx 2.6 \times 10^9 / \text{cm}^2$ and $n_{\text{annealed}}^{\text{off-site}} \approx 0.9 \times 10^9 / \text{cm}^2$ (not shown). For both samples the QD density is about three times higher in the pre-structured area compared to the unstructured one which relates to the site-selective nucleation of QDs. The difference in on-site QD density between the as grown and the annealed sample supports the above observation of the morphological transition. The merging of double dots into single QDs, as a consequence, reduces the density of QDs. The annealing step as well reduces the off-site density of randomly grown QDs by a factor of about 2. In correspondence, the on-site QD densities differ by a factor of about 2 because double dots merge into single dots and the number of random dots is halved.

Additional holes appear between the defined ones. These randomly distributed "defect" holes interfere with the attempt to precisely control QD locations since they act as nucleation sites as well. Their origin is currently under investigation. Interstitial QD nucleation is also observed which we attribute to the high amount of InAs.

The optical quality of the as grown QDs was analyzed by $\mu$-PL measurements. A set of spectra is depicted in Fig. 3. The large energy range spectra were obtained by combining several high resolution scans of small energy range. The spectra were recorded on the pattern with 250 nm spacing. Individual QD peaks are visible with linewidths above 100 $\mu$eV. We observe that the intensity of the emission is reduced and the linewidth is increased compared to QDs that were randomly grown on unstructured substrates. Due to the QD density and the limited laser spot size (1 $\mu$m) we were not able to analyze single dots. However, the QD luminescence within the pre-structured area is confirmed. Furthermore, first measurements show a comparable optical quality for annealed QDs (not shown).

In conclusion we have investigated the effect of in-situ annealing on InAs QDs grown on pre-structured substrates. We have found that site-selective QDs undergo a morphological transition due to annealing with double dots merging into single dots. As a consequence, the QD density is reduced. First $\mu$-PL measurements show luminescence of QDs in the
pre-structured area with excitonic linewidths down to 180 μeV.

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FIG. 1. (Color online) AFM image (1.75 μm × 1.75 μm) of holes fabricated by EBL and WCE. The spacing is 500 nm and the average hole size is 58.4±6.1 nm. The linescan shows the profile of a representative hole.

FIG. 2. (Color online) AFM images (2 μm × 2 μm) of as grown (left) and annealed (right) QDs (1.7 ML InAs) grown on pre-structured samples. The spacing between holes is 250 nm. A morphological transition from double dots to single dots is observed. This is confirmed by representative linescans.

FIG. 3. (Color online) Large energy range μ-PL measurements of as grown site-selective InAs QDs. The μ-PL was recorded at four different positions within the pre-structured area. Individual QD peaks are visible with minimum excitonic linewidths of 180 μeV.
Figure 1
Figure 2
Figure 3
APPENDIX G

Characterization of 1D photonic crystal nanobeam cavities using curved microfiber

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Characterization of 1D Photonic Crystal Nanobeam Cavities Using Curved Microfiber

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Abstract: We investigate high-Q small mode volume photonic crystal nanobeam cavities using a curved, tapered optical microfiber loop. The strength of the coupling between the cavity and the microfiber loop is shown to depend on the contact position on the nanobeam, angle between the nanobeam and the microfiber, and polarization of the light in the fiber. The results are compared to a resonant scattering measurement.

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References and links

1. Introduction

The use of photonic crystal nanocavities as a means of confining light has led to an active field of cavity quantum electrodynamics research in the solid state. Specifically, the interaction between such confined fields and matter has led to the observation of a number of fundamental quantum optics results in semiconductors [1-3]. The primary way of enhancing such interactions between light and matter is to increase the ratio of quality factor Q to effective mode volume V. Increasing Q provides longer photon storage times, which leads to a greater chance of interaction between the light and the matter. Decreasing V leads to higher field intensities in the cavity, and hence stronger interactions between the light and the matter. High Q and small V are pursued by the semiconductor cavity QED community because they are essential for large Purcell enhancement ($\Omega_o = Q/V$) of spontaneous emission and for a large vacuum Rabi splitting ($\Omega_{VS} = Q/V\gamma$) [1, 2, 4-6].

Cavity QED experiments with quantum dots (QDs) as the active emitters are usually performed at cryogenic temperatures. In this temperature regime, radiative recombination of excited carriers is the dominant decay mechanism, and hence the dots are easily studied by optical spectroscopic techniques. As a result, the standard technique of measuring cavity $Q$s using QD photoluminescence usually requires expensive helium cryostats. Since characterizing cavity $Q$s is a time consuming task for researchers in this field, techniques have been developed to enable measuring $Q$s independently of the active emitters, and hence at room temperatures. Measuring a probe signal in a cross-polarized resonant scattering configuration [7-8] and using a tapered microfiber probe [9-10] are two such techniques that have been developed and employed specifically for semiconductor cavity QED.

We report the results of our investigations of 1D photonic crystal nanobeam cavities by means of a microfiber tapered loop. Using this method to investigate silicon nanobeam on a silica substrate, we have measured the highest $Q/V$ ratio reported for such devices. We present the results of these experiments, as well as a comparison between the two methods of cross-polarized resonant scattering and tapered fiber transmission. We observe an asymmetric lineshape of the cavity modes using both approaches, and show that the asymmetry can be varied in the case of the tapered fiber by varying the input polarization of the probe field.

2. Photonic Crystal Design and Fabrication

The cavity considered here is a nanobeam cavity, which is essentially a wavelength-scale Fabry-Perot etalon formed by sandwiching a 1D photonic crystal waveguide between 1D photonic crystal Bloch mirrors, as shown in Figure 1. In the transverse directions, the light is confined in the nanobeam by total internal reflection. By smoothly tapering the air hole radius and the corresponding lattice constants in the mirror sections, the scattering loss is minimized and a high $Q$ is achieved [10-11]. 3D finite-difference time-domain (FDTD) simulations [13] reveal that the cavity exhibits a reasonably high $Q$ in excess of 500,000 with very low mode volumes, even though it is placed on a low index substrate. The region of tapered holes in the center of the nanobeam effectively confines the light, analogous to a Fabry-Perot spacer.
The nanobeams are fabricated using electronics grade silicon-on-insulator with a 220 nm silicon device layer and 2 μm buried oxide. To prepare the samples for electron-beam lithography, the wafers are manually cleaved, cleaned with acetone and isopropyl alcohol, and the native oxide is removed by a short dip in 10:1 buffered hydrofluoric acid. The samples are then baked at 180 °C, spin-coated with 2 percent PMMA 950K in chloroform, and baked again at 180 °C for 5 minutes. Electron-beam lithography is performed in a Leica EBPG 5000+ at 100 kV. Following electron-beam exposure, the samples are developed in 1:3 MIBK:IPA for 60 seconds, rinsed in IPA, and dried with nitrogen. After development, the wafers are etched using an Oxford Instruments Plasmalab System100 ICP380 with a mixed-mole gas chemistry consisting of SiF₆ and Cl₂F₂. Figure 2 shows an SEM of one of our nanobeam cavities.

3. Fiber Transmission Measurement

We investigate the spectral response of our nanobeam cavities by means of a curved, tapered optical fiber. The taper is fabricated in two stages. In the first stage, a standard, single-mode optical fiber is heated and pulled to a diameter of 1–1.5 μm. In the second stage, the taper is curved into a loop with 200 μm radius of curvature. The fiber taper loop is mounted onto a motorized XYZ stage and brought into contact with the nanobeam cavity. An Agilent 8164A mainframe with an Agilent 81682A tunable laser with 0.2 nm wavelength resolution is input into the fiber equipped with an inline polarization controller before propagating through the tapered region. The transmitted light is detected at the output end of the fiber by an InGaAs photodiode. The laser is tuned across the cavity resonance, and the interaction of the light with the cavity mode can be observed as a change in the transmitted intensity of the propagating field. The laser needs to have resolution better than the FWHM of the cavity mode and amplitude fluctuations must be slower than the time to scan across a mode. The Q is the transition energy of the mode divided by the FWHM energy width of the cavity mode.
nanobeam sample is mounted on stage rotating about an axis normal to the sample, so that the angle between the tapered fiber and the axis of the nanobeam cavity can be varied. Figure 3 (a) shows a typical fiber loop transmission spectrum of a nanobeam cavity, with two cavity modes visible.

![Graph showing fiber loop transmission spectrum](image)

Figure 3. (a) Fiber loop transmission spectrum of a typical nanobeam cavity at 45°, center contact (black) and spectrum directly from laser (red). (b) Fiber loop transmission spectrum of a high Q nanobeam cavity mode, at 45°, edge contact, with $Q = 75,000$.

4. Fiber Coupling to a Nanobeam

By nature of the physical interaction between the field in the fiber taper and the nanobeam, a degradation of the cavity $Q$ is expected. This is due to an additional source of losses contributed by the fiber taper, as the measurement is now of the coupled fiber-cavity system. However, the losses introduced by the presence of the fiber taper can be mitigated by careful selection of the contact parameters. The first parameter we adjust is the pressure of the fiber loop onto the nanobeam. The fiber taper loop is brought in toward the nanobeam using small steps on a motorized actuator. At some critical distance from the sample surface, Van der Waals and electrostatic forces pull the fiber taper in, causing it to stick to the surface. Once the loop is in contact with the nanobeam, the actuator can still be advanced, increasing pressure and the contact length between the fiber taper and the nanobeam. The actuator can also be pulled away slowly while the loop is still stuck to the surface, decreasing the contact length. After sufficient force is applied to pull the loop away by overcoming the sticking force, it pops off the surface of the sample.

The second parameter we adjust is the contact position along the length of the nanobeam. We have confirmed that the weakest coupling, and therefore the highest $Q$, is observed when contact is made as close as possible to the edge of the nanobeam, whereas contact in the center of the nanobeam produces the strongest coupling and hence the lowest $Q$. Contact in the center of the nanobeam also modifies the index of refraction in the vicinity of the cavity, which leads to a shift in the resonance frequency. A further degradation of the $Q$ is observed if the fiber loop pressure is too hard on the nanobeam as described above.

The third parameter we adjust is the angle between the fiber taper and the axis of the nanobeam cavity. Because the polarization of the input field is always perpendicular to the axis of the fiber, the angle between the fiber and the nanobeam will affect the coupling to the mode of the nanobeam, which is linearly polarized perpendicular to the axis of the nanobeam. One would expect that the best polarization matching would occur when the nanobeam is aligned parallel to fiber. However, this configuration also produces the strongest coupling between the field in the fiber and the nanocavity, as well as the largest index modification in the vicinity of the cavity. Even when the contact is made at the edge of the nanobeam, the length of the contact region between the fiber and the nanobeam extends over a large fraction of the nanobeam. Hence, this configuration produces a deep dip in the transmitted signal, but does not yield the highest measured $Q$s. One might think that the perpendicular configuration
between the fiber and the nanobeam would yield the highest $Q$ because of minimized contact between them as well as minimal coupling into the nanobeam. However, because of the drastically reduced coupling in the perpendicular configuration due to orthogonal polarizations, we were not able to observe any cavity modes in that configuration at the edge of the nanobeam. The highest $Q$s result with an angle ranging from 20° to 60° between the fiber taper and the nanobeam, and contact made at the edge of the nanobeam. This configuration reduces the physical contact between the fiber and the nanobeam compared to the parallel configuration, but still supports a polarization component that is matched to the nanobeam mode. The highest $Q$ we measured was 75,000 with a computed mode volume of 0.257(6)$\mu$m$^3$ in the 45° edge configuration, yielding $Q^V = 278,000$. As far as we know, this yields the highest $Q/N$ ratio that has been achieved on nanobeam cavities on substrate. The group of De La Rue [14] reported a $Q$ of 147,000 with a computed mode volume of 0.85(3)$\mu$m$^3$, yielding $Q^V = 175,000$. Figure 3 (b) shows the spectrum of the highest $Q$ nanobeam cavity mode.

5. Comparison to Resonant Scattering

As mentioned earlier, the presence of a fiber taper in contact with a nanobeam cavity provides an additional loss mechanism for light in the cavity. While this allows us to probe the $Q$ of the system by measuring the transmission through the fiber, it also reduces the $Q$ compared to the inherent $Q$ that the cavity would have by itself. In order to investigate this loss mechanism, we have compared the results of measurements with the fiber taper loop to measurements performed by cross-polarized resonant scattering [7-8, 11].

Cavity modes measured using resonant scattering are known to exhibit asymmetric lineshapes [15]. These lineshapes are attributed to a Fano interference between the resonantly scattered light and the coherent background. In order to extract a linewidth from such an asymmetric profile, we fit the signal to a Fano linehape

$$F(\omega) = A_0 + A_1 \frac{\left[ 2(\omega - \omega_0) / \Gamma \right]}{1 + \left[ 2(\omega - \omega_0) / \Gamma \right]^2}$$  \hspace{1cm} (1)

where $\omega_0$ is the frequency of the cavity mode transition, $\Gamma$ is the resonance linewidth, $A_0$ and $A_1$ are offset and scaling factors, respectively, and $g$ is the Fano parameter that quantifies the asymmetry of the linehape. Adjusting these parameters to fit a curve to our resonant scattering data, we are able to extract the underlying linewidth, and hence $Q$ of the cavity mode. Such a fit is shown in figure 4 (b). Galli, et al have reported [15] that the asymmetry of the linehape from resonant scattering data can be varied by changing the spot size of the laser beam on the sample.

![Figure 4](image)

Figure 4. (a) Fiber loop transmission spectra of a typical nanobeam cavity; $Q = 25,000$ (black) and spectrum directly from laser (red). (b) Cross-polarized resonant scattering signal of the same nanobeam cavity as (a), $Q = 44,100$ (black); fitted Fano linehape (blue); spectrum directly from laser (red).
Cavity modes measured using the fiber taper also displayed asymmetric lineshapes, which we attribute to interference between two pathways: light interacting with the cavity mode and emitted back into the fiber and light coupled into the 2D slab. However, unlike the case with the resonant scattering technique, the asymmetry of the lineshape with the fiber taper technique is strongly dependent on polarization. Hence, it was always possible to tune the polarization such that the resulting lineshape was symmetric, and this allowed us to directly fit a Lorentzian lineshape in order to extract the $Q$, without resorting to the more complicated fitting associated with the Fano Equation (1). The $Q$s measured using the resonant scattering technique were higher than those measured for the same cavity using the 45° fiber configuration at the edge of the nanobeam, consistent with the idea that the presence of the fiber loop lowers the $Q$ by introducing an additional loss mechanism.

How do the two systems compare in other ways? The fiber taper loop measurement is more difficult to set up than the resonant scattering measurement, primarily because of the equipment and experience needed to successfully taper and curve the microfibers. Once the measurement is set up, the fiber taper loop measurement is quite robust and does not suffer from the extreme sensitivity to variations in alignment of the resonant scattering technique. Both measurements can be performed at room temperature or at cryogenic temperatures; however, the fiber taper measurement would require extensive modifications to most cryostats in order to insert the fiber taper and control its motion. In contrast, the resonant scattering technique can be performed with all of the optics outside of the cryostat. The fiber taper measurement is performed in an all-fiber configuration, whereas the resonant scattering method is performed in free space. We found that absorption lines due to atmospheric nitrogen in the spectral region of our nanobeam cavity modes also appear as dips in the measured resonant scattering spectrum. In order to accurately measure the $Q$ of a cavity mode coincident with an absorption dip, as was unfortunately the case for the $Q = 75,000$ cavity in Fig. 3 (b), the mode would need to be spectrally shifted away from the absorption dip by a technique such as heating or condensation of xenon or nitrogen gas.

6. Conclusion

In conclusion, we have measured the $Q$s of 1D photonic crystal nanobeam cavities on a substrate using the transmitted signal through a fiber taper loop. Using this technique, we have measured a $Q$ as high as 75,000 with a computed mode volume of $0.27(\lambda/n)^3$, representing the highest $Q/V$ ratio reported in this system. We have observed dependence of the measured $Q$ on contact position and force between the nanobeam and the fiber taper, angle between the nanobeam and the fiber taper, and polarization of the light in the fiber. We have observed that higher $Q$s are measured when the fiber loop is contacted at the edge of the nanobeam and with a 20° to 60° angle between the fiber loop and the nanobeam. We have shown that the fiber taper loop technique is capable of measuring high $Q$s, and that by tuning the polarization in the fiber it is possible to eliminate the asymmetric lineshapes.

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