A Scalable Quantum Computation Platform: Solid State Quantum Memories Coupled to Photonic Integrated Circuits

by

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Submitted to the Department of Electrical Engineering and Computer Science
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Abstract

Quantum computation and communication systems exploit quantum mechanical effects to
surpass their classical counterparts in certain applications. However, while proof-of-principle
experimental demonstrations have been performed, these are limited to a handful of nodes
with limited - and often immutable - connectivity. Here we demonstrate an integrated plat-
form for solid state quantum information processing.

Pre-characterized solid state quantum nodes (nitrogen vacancy centers in diamond nanopho-
tonic structures) are placed into a photonic integrated circuit which allows for low-loss and
phase-stable collection, routing, and detection of photons as well as on-chip state manipu-
lation and classical control. Moreover, the fabrication of high-quality photonic resonators
in diamond allows for the increased emission and collection rates of photons coherent with
the spin state. These two advances promise an on-chip entanglement rate much larger than
the decoherence rate, allowing the creation and maintenance of cluster states for quantum
computation.

Thesis Supervisor: Dirk R. Englund
Title: Assistant Professor of Electrical Engineering and Computer Science
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Chapter 1

Introduction

Quantum sensing, communication, and computation systems leverage unique properties of quantum mechanics to surpass their classical counterparts in certain applications. Theoretical work promises enhanced sensitivity \[2\], secure communication networks \[3\], and computation speed-ups for problems that are classically intractable \[4, 5, 6\]. However, while proof-of-principle experimental demonstrations have been performed, these are limited in scale due to high decoherence, low entanglement rates, and large experimental overhead. This thesis focuses on architectures for quantum information processing at the scale necessary for quantum advantage - around 100 qubits for the first demonstrations of quantum supremacy over classical hardware, or even millions for the demonstration of fault-tolerant universal computation. There are two main thrusts towards the goal of scalability:

- Increased entanglement rate between spatially separated quantum nodes, and
- Scalable architectures for quantum information processing.

1.1 Increased Entanglement Rate

Entanglement between remote quantum nodes is a key resource for quantum sensing, communication, and computation. This entanglement can be mediated via the coherent coupling between matter-based quantum memories and well defined optical modes \[7\]. However, the maximum size of a quantum network is limited by the ratio between the entanglement rate and the coherence time of the matter qubits. Currently, the maximum demonstrated size of a distributed quantum network is limited to two optically connected quantum memories,
mainly due to low collection efficiency into spatially and spectrally indistinguishable optical modes.

This thesis focuses on the negatively charged nitrogen vacancy (NV) center in diamond, which is a promising solid state quantum memory. The electron spin state of the NV center has millisecond spin coherence times \[8\] and can be mapped onto the occupation of a single optical mode \[9, 10\] for mediating entanglement between spatially separated qubits. Moreover, the electron spin state can be mapped onto nearby nuclear spins, increasing the coherence time to seconds. Chapter 2 gives an introduction to the spin and optical properties discussed in this thesis.

Recent work has demonstrated high fidelity entanglement \[10\] between two spatially separated NV centers but even these impressive results demonstrate entanglement rates more than twenty times slower than the nuclear spin decoherence rate, limiting the maximum network size to two. The maximum network size will increase with increased collection efficiency into a single useable optical mode while maintaining a long spin coherence time. To increase emission into the desired frequency mode, and collection into the desired spatial mode, it is necessary to modify the electromagnetic environment of the defect center. Chapter 3 describes the diamond fabrication techniques developed to pattern the diamond on the nanoscale. Chapter 4 describes the simulation and measurement of optimized resonant diamond devices for increased collection of photons coherent with the spin state.

### 1.2 Scalable Architectures

As quantum systems grow, so do the required control and measurement systems. Thus, it is necessary to develop experimental architectures in which the control structure does not limit the performance of the quantum computer. Solid state quantum nodes naturally remove the need for complex trapping architectures as the atomic-like defects are naturally trapped in a potential created by the host crystal. However, it is still necessary to route the optical and microwave control fields to each node with low temporal and spatial cross talk and to route and detect the emission in a reconfigurable manner. Chapter 5 discusses the demonstration of monolithic and hybrid integrated quantum systems. The thesis ends with an outlook on the future of large-scale engineered quantum systems.
Chapter 2

The Negatively Charged Nitrogen Vacancy Center in Diamond

There is a myriad of defects in diamond, known as color centers, with emission across the visible and near infrared spectrum [11]. The optical and spin properties of many of these color centers have been studied extensively. This thesis will focus on the negatively charged nitrogen vacancy (NV) center. The NV center comprises a nitrogen in a carbon lattice site and a vacancy at an adjacent site, as shown in Fig. 2-1(a). In the negatively charged state, there are six electrons trapped at the defect - three electrons from the three adjacent carbons, two electrons from the nitrogen, and a final electron from the conduction band of the diamond lattice. The $sp^3$ orbital is thus missing 2 electrons, and we consider the two-hole wave function when describing the physics of the NV center, which is equivalent to a two electron system. This spin-1 system gives rise to the spin triplet ground and excited state levels that are used for storing quantum information [12].

2.1 NV Creation

Nitrogen can be found in natural diamond or incorporated during the growth process of diamond grown via chemical vapor deposition, and some of these nitrogens will naturally form NV centers. However, large scale quantum information processing systems based on NV centers cannot rely on purely random NV creation. It is possible to implant nitrogen ions into the diamond crystal [13]. The energy of the ion beam gives control over the depth of the N atoms. The implantation of N ions also creates vacancies in the diamond lattice by
knocking carbon atoms out of the lattice [13]. NV centers are formed after implantation by annealing above 600°C which promotes the diffusion of the vacancies which are then trapped in the low-energy NV configuration. It has been found that annealing up to 1200°C improves the spin and optical coherence properties as multi-vacancy defects are broken up [14]. The conversion probability from an N atom in the diamond lattice to a NV center is generally only 1-5% [13].

![Diagram of NV center in diamond](image)

Figure 2-1: (a) Atomic structure of the NV center in diamond. (b) Level structure of the NV in zero magnetic field. (c) Optically detected magnetic resonance. (d) Spectrum of the NV center at 10K with above band (532 nm) pumping.

2.2 Spin Properties

Even at room temperature, the electron spin of the NV center has a long coherence time. Fig. 2-1(b) shows the level diagram of the NV center at 0 magnetic field, including the ground and excited state spin triplets and a long-lived singlet state. Even in the absence of an external magnetic field, the $m_s = 0$ and $m_s = \pm 1$ states are split due to a spin-spin interaction. The degeneracy between the $m_s = 1$ and $m_s = -1$ sub levels can be lifted by
the application of a magnetic field. These spin sub levels have long coherence times with longitudinal electron-spin relaxation times measured up to $T_1 = 200\,\text{s}$ [15], and typical spin phase coherence time of $T_2^* \sim 1 - 10\,\mu\text{s}$ which can be extended by decoupling sequences, typically to a few $100\,\mu\text{s}$ [16] and even up to $T_2 = 1\,\text{s}$ [8]. The state of the electron spin can also be coherently mapped to the state of nearby nuclear spins which can extend the coherence time [17] and could provide encoding of error-corrected logical qubits [18].

The $m_s = 0$ excited states decay directly to the $m_s = 0$ ground state, while the $m_s = \pm 1$ excited state levels couple to a long-lived singlet state, labeled as $^1A$ in Fig. 2-1(b), which decays non-radiatively to the $m_s = 0$ electronic ground state. This leads to a spin dependent fluorescence, and allows for optically detected magnetic resonance (ODMR) measurements, as seen in Fig. 2-1(c), allowing for measurement of the spin population and simple and high-fidelity initialization into the $m_s = 0$ ground state.

2.3 Optical Properties

Unlike the exceptional spin coherence of the NV center, the coherence of the optical transitions is poor. The shading in Fig. 2-1(b) denotes the quasi-continuum of phonon modes in the diamond lattice, and is expanded in the inset of Fig. 2-1(d). These phonon modes allow for the incoherent excitation of the NV, as above-band excitation excites both the NV and phonon modes in the diamond lattice. These phonons decay within picoseconds. Unfortunately, the complementary process also occurs — when the NV excited state decays some energy is lost to the diamond lattice due to the excitation of vibrational modes [19]. This gives rise to a broad phonon side band (PSB) in the emission spectrum as seen in Fig. 2-1(d). Purely radiative decay produces photons at the transition frequency of the NV, known as the zero phonon line (ZPL). The ratio between these decay pathways is described by the Debye-Waller factor, and is only $\sim 3\%$ in bulk diamond. Only the ZPL photons can be used for photon-mediated entanglement between NV centers because the other transitions lose information to the diamond lattice that cannot be retrieved, and thus are not fully entangled with the spin state.

The collection of the ZPL photons is further hindered by the high refractive index of diamond ($n = 2.4$) which causes high total internal reflection and the dipole emission pattern of the NV which is not easily collected into a single guided mode such as a fiber or a
waveguide. A major part of this thesis describes the design, fabrication, and measurement of nanostructures to optimize the photonic density of states at the ZPL of the NV center to increase the emission of photons at the ZPL, suppress the photonic density of states in the phonon side band, and to increase collection of those photons into guided spatial modes.

2.3.1 Low Temperature Properties

For the creation of entanglement between spatially separated NV centers via photonic channels it is necessary to have spin-conserving optical transitions with good coherence properties. The excited state properties of the NV center are determined by spin-spin and spin-orbital interactions, as well as any external strain [20], electric [21], or magnetic fields [22]. The excited state structure consists of a spin triplet and an orbital doublet. At high temperatures, electron-phonon interactions average the orbital populations. This mixing rate follows a $T^5$ dependence, and is negligible below $\sim 10$ K [23, 20]. The degeneracy between the orbital doublets is split with any non-axial strain field [23], and the degeneracy between the $m_s = +1$ and $m_s = -1$ spin states is lifted with an external magnetic field. Fig. 2-2(a) shows the six allowed optical transitions.

All of the optical transitions are spin conserving, but the orbital angular momentum can change depending on the polarization of the photon emitted. The two $m_s = 0$ transitions are spin conserving and provide good cycling transitions. The other four excited states are entangled spin and orbital states. Moreover, the $m_s = \pm 1$ excited states are not cycling due to coupling to the metastable singlet state which decays to the $m_s = 0$ ground state. This thesis considers only the two cycling $m_s = 0$ transitions, which we call $E_x$ and $E_y$.

All of the six optical transitions can be used to transfer the electronic spin state of an NV center to the state of a photon in various ways. For instance, Togan et al. showed high fidelity entanglement between the electronic spin state and the polarization of the emitted photon by using the degeneracy between the $m_s = \pm 1$ states at $B = 0$ to create entanglement between the electronic state of the NV and the polarization of the emitted photon [9]. Bernien et al. demonstrated entanglement generation between spatially separated NVs by first entangling the spin state with the population of a photonic mode via selective excitation of the $m_s = 0$ transition [10].

Any implementation of entanglement generation between spatially separated NV centers relies on the indistinguishability and temporal coherence of the photons. For a system well
isolated from the environment, the spectral width of a transition will be inversely proportional to the transition’s lifetime: \( \Delta \nu \Delta \tau = 1/2\pi \). The lifetime of the \( m_s = 0 \) transition is typically 13 ns in a bulk diamond, and thus we expect a lifetime-limited linewidth of about 12 MHz for the \( E_x \) and \( E_y \) transitions. Initial studies of the temporal coherence properties of NV centers were promising. Although dephasing was seen at high pump powers [24], single-scan linewidths down to the lifetime limit were measured in natural NVs in bulk diamond [21, 25]. Unfortunately, repeated scans across the transitions showed an inhomogeneous distribution of center frequencies. This temporal inhomogeneous broadening of spectroscopic lines is seen in all solid state emitters and is referred to as spectral diffusion.

The spectral diffusion in NV centers is attributed to changes in the local charge environment by the excitation laser which leads to changes in the transition frequencies due to the Stark effect.

The spectral diffusion of NV centers is exacerbated by the ionization dynamics. Resonant excitation causes photo-ionization out of \( \text{NV}^- \), into \( \text{NV}^0 \) charge state. This can be reversed via high energy (e.g. 532 nm) excitation that excites charge impurities in the environment and re-initializes the NV center to the \( \text{NV}^- \) state. However, this necessarily causes large changes in the charge environment of the NV center, which increases the inhomogeneous distribution of transition frequencies. For instance, even NV centers with nearly lifetime-limited single line scans are broadened by an order of magnitude with the introduction of a
532 nm repump beam [25, 26, 27, 28]. It has been shown that resonant control of the charge state of the NV by pumping at the NV\(^0\) zero phonon line (575 nm) can reduce spectral diffusion [29, 14]. Presumably spectral diffusion is reduced because the power needed to switch from the NV\(^0\) to NV\(^-\) charge state is reduced as 575 nm light can ionize electrons from the conduction band of diamond and resonantly excite the NV\(^0\) where it then can trap the extra electron to switch to NV\(^-\) charge state.

Even with a 532 repump beam and the subsequent spectral diffusion, two photon quantum interference of photons emitted by the same NV has been shown [26, 27], and the interference of photons emitted by two NV centers can be achieved using the Stark effect to overlap the transition frequencies of interest with external DC electric fields [26, 28]. To achieve high fidelity quantum entanglement between NV centers, researchers used a combination of the yellow repump and periodic checks to ensure that the NV is on resonance before attempting entanglement [10, 30].

Most of the initial excited-state spectroscopy measurements and all of the interference and entanglement generation demonstrations have focused on single "hero" NV centers with superb optical and spin coherence properties. Unfortunately, every NV experiences a different electronic environment due to the distribution of lattice defects, and this problem is exacerbated in near-surface NV centers. Fig. 2-2(c) shows the distribution in linewidths of implanted NV centers for two different EG samples to illustrate the spread in spectral diffusion over one sample, and between samples.

As we start to scale up NV-based systems to the size needed for quantum information processing, it will be necessary not only to pick the best NV centers but to fully understand the origins of this inhomogeneous distribution of linewidths to systematically prepare diamonds for consistent creation of NV centers in nanostructures with lifetime-limited linewidths.

### 2.4 Surface Termination

All previous demonstrations of interference or entanglement between separated NV centers have been performed on NV centers deep within the diamond, using SILs to increase photon collection. Thus, the surface properties of the diamond had no effect on the coherence of the optical transitions. However, the collection rate is fundamentally limited by the Debye-Waller factor. Nanophotonic devices such as waveguides and photonic crystal cavities can
improve the emission and collection efficiency into a desired spectral and spatial mode. However, these structures all must be patterned on the order of the wavelength of interest in diamond ($\lambda_{\text{ZPL}}/n$). This necessarily brings the diamond surfaces close to the NV center and thus it is necessary to understand and manipulate the properties of the diamond surface. This section provides an overview of the literature on diamond surface chemistry.

The charge state of the NV center can be affected by large changes in the electric field surrounding it. In standard operating regimes, the charge state is stable, though the ratio of NV$^-$ to NV$^0$ is affected by surface termination. It has been found that oxygen termination (via a 475°C Oxygen anneal) results in a higher NV$^-$ concentration [31, 32]. Fluorinated surfaces also show increased NV$^-$ concentration [33, 34, 35, 36]. In contrast, a hydrogenated surface increases the NV$^0$ concentration [37]. A wetting layer is formed due to the H-C bonds at the surface, pulling electrons from the valence band and causing a depletion band where the NV$^0$ charge state is more stable. This has also been exploited for the active control of single NV centers via Schottky junctions made from a hydrogenated diamond surface and Al contacts [38, 39], electrolyte gates [40], diamond PIN diodes [41], and in-plane gate structures [42]. Finally, phosphorous and boron doping of diamond has also been shown to affect the relative concentration of NV$^-$ and NV$^0$, although surface termination effects are still dominant to 60 nm [43].

The above literature uses band bending induced by the surface termination to explain the charge switching effects. However, there has also been simulation work suggesting that the effects arise from surface image states and NV-related gap states [44].

The techniques used to stabilize the charge state of the NV center discussed above give only coarse tuning of the electronic environment that the NV center feels, and as of now there's no literature to suggest any effect on spectral diffusion of these techniques. Unlike charge state stabilization, spectral stability is affected by local changes in the electronic environment. These changes are caused by the high power 532 nm laser used to initialize the charge and spin state, though the dynamics of this process are not fully understood, especially for near surface NV centers. Of the studies mentioned above, only one looks into the spectral stability of NV centers, and finds that localized charge traps still exist [31].

There has been one paper showing stable NV centers 100 nm deep in diamond [14] after high temperature annealing and a tri-acid clean. However, this work was not done on nanostructured diamond, the number of measured NVs is small, so it's unclear how general
the results are, and they do not discuss the surface termination of the diamond, although we can presume it to be mainly oxygen terminated as the tri-acid removes any graphite layer.

For stable NV centers in nanostructures, we would like a stable reconstruction of the bonds at the surface of the diamond. A lot of theoretical and experimental work has been done on the surface chemistry of diamond. Early studies showed that the \{100\} and \{111\} facets of diamond can reconstruct cleanly and reproducibly, but the \{110\} facet does not [45]. This is especially important because the quasi-isotropic etch that we use to create nanostructures in diamond (Sec. 3.4) leaves only \{100\} faces exposed.

Beyond carbon terminations, theoretical work has studied the coverage of H and O terminations of \{100\} diamond [46], and termination has been studied experimentally, although without considering the effect on NV centers [47, 48].

Simulations suggest that nitrogen-terminated \{111\} diamond can provide a stable electrostatic environment [49], though it has not been shown experimentally. Nitrogen termination of \{100\} diamond has been achieved, though the NV properties were not studied [50, 51]. Simulations also suggest that fluorination and chlorination can provide stable terminations of the diamond facets [52]. Many experimental studies of fluorination have been done, but the effects on the spectral diffusion of NV centers have not been studied [53, 54, 55].

In conclusion, while there is a wealth of information on diamond surface chemistry, it still remains to be seen how - and if - the surfaces of nanostructures can be passivated to stabilize the charge state and transition frequencies of NV centers in nanostructures.

### 2.5 Entanglement Protocols

Optically-mediated entanglement between spatially separated NV centers hinges on the distinguishability of the photons they emit. Spectral indistinguishability can be achieved by an external DC electric field via the Stark effect. Spatial indistinguishability can be achieved by coupling into single modes and erasing the which-path information through a 50:50 beam splitter. The basic steps of an optically-mediated entanglement protocol are outlined in Fig. 2-3.

There are many proposed techniques for optically-mediated entanglement of spatially separated atomic qubits. The rate and fidelity of entanglement for each case is affected by residual distinguishability, collection efficiency, and unwanted counts (due to electrical
Figure 2-3: A simplified version of photon-mediated entanglement between 2 spatially separated NV centers. (a) The spin each of the NV centers is prepared in a superposition \(|0\rangle + |1\rangle\). (b) The spin of each NV center is entangled with the state of a photonic channel. (c) The path information of the photons is erased with a 50:50 beam splitter, and a detection event heralds entanglement.

noise, leaking laser light, or undesired radiative atomic decay pathways. The choice of a specific protocol should be driven by the requirements of a particular application, and the noise sources and losses of a particular implementation. Previous implementations of entanglement between the spin states of spatially separated solid state qubits have followed a protocol first outlined by Barret and Kok [56]. While we do not perform entanglement in this thesis, the steps of the protocol motivate the need for high collection efficiency of indistinguishable photons from the NV center:

- We first prepare both spins a superposition: 
  \[
  |\Psi\rangle_{AB} = (|0\rangle_A + |1\rangle_A) \otimes (|0\rangle_B + |1\rangle_B)
  \]
- We then excite only the \(|1\rangle\) to \(|e\rangle\) transition: 
  \[
  |\Psi\rangle_{AB} = (|0\rangle_A + |e\rangle_A) \otimes (|0\rangle_B + |e\rangle_B)
  \]
- Each NV center then decays and (with probability \(\eta\)) emits a photon into the collection mode, or the photon is lost to the environment with a probability \((1-\eta)\). The collection modes are impinged on a beam splitter, and the outputs are sent to single photon detectors. A detection event collapses the atomic state, but not to a fully entangled state. 
  \[
  |\Psi\rangle_{AB} = |11\rangle_{AB} + |01\rangle_{AB} + |10\rangle_{AB}
  \]
- To get rid of the unwanted mode, we flip both spins, and the \(|1\rangle\) to \(|e\rangle\) transition is excited again. 
  \[
  |\Psi\rangle_{AB} = |00\rangle_{AB} + |01\rangle_{AB} + |10\rangle_{AB} + |11\rangle_{AB}
  \]
- Again, these modes are impinged on a beam splitter, and the outputs are sent to detectors where a click on the same (different) detector creates a fully entangled state 
  \[
  |\Psi\rangle_{AB} = |01\rangle_{AB} + |10\rangle_{AB} \quad |\Psi\rangle_{AB} = |01\rangle_{AB} - |10\rangle_{AB}
  \]
The fidelity of this protocol is reduced both by dark counts and indistinguishability between the photon modes. This fidelity is not limited by a low collection rate due to the double heralding, although the rate of entanglement is proportional to $\eta^2$. This two-round scheme also reduces the requirements on phase stability, requiring interferometric stability only during the time of the two rounds, as the phase difference accumulated due to a difference in path lengths between the atoms and the detectors is canceled out [10]. The analysis of the achievable entanglement rate has also been extended to include frequency mismatch between the transition frequencies of the two NV centers. Interestingly, it is possible to obtain unity efficiency with different (but known) frequencies with sufficiently fast detectors [27].

The rate of this protocol is limited by the collection efficiency of photons entangled with the spin state. At the time of writing, the fastest reported entanglement rate is 1 every 250 s [57], or 0.004 Hz, much slower than the electronic or nuclear spin decoherence rate. In order to create cluster states with 10s or 100s of qubits within the spin coherence time, the entanglement rate must be improved by at least five orders of magnitude so that hundreds of successful entanglement operations can be performed within the spin coherence time.

### 2.6 Experimental Techniques

All experiments in this thesis make use of confocal scanning laser microscopes with slight modifications. We pump the sample with 532 nm, 575 nm, or 637 nm light, and collect the reflection and fluorescence through a high NA objective. The fluorescence image is separated from the reflected pump beam with various long pass optical filters depending on the application. A pinhole is used to spatially filter the fluorescence. We use room temperature setups for easy characterization of photonic structures, and cryogenic setups for excited state spectroscopy. We use closed-cycle cryostats and generally work around 4K.

There are two main spectroscopy techniques that are used in this thesis. The first is photoluminescence (PL) spectroscopy where we excite at 532 nm and collect the broadband NV fluorescence. Fig. 2-1 shows a standard room temperature spectrum collected under 532 nm excitation. PL is used to identify NV centers, measure spin populations, and quantify the effect of engineered nanostructures on the emission properties of the NV center. We also use photoluminescence excitation (PLE) spectroscopy to explore the structure of the excited
state levels at cold temperatures. In PLE experiments, we tune a narrow linewidth laser
stabilized to an external wavemeter across the transition frequency of the ZPL, around
637 nm. We then filter out the excitation laser and collect photons from the phonon side
band with a 650 nm long pass filter. In PLE measurements either a 532 nm or 575 nm laser is
used to stabilize the charge state of the NV center. PLE measurements allow us to measure
the frequency-dependent excitation cross section of different NV centers to understand the
spectral diffusion of NV centers in different samples.
Chapter 3

Diamond Fabrication

3.1 Introduction

Color centers in diamond - including the NV center - have emerged as important tools for quantum information processing. Almost all applications depend directly on the collection efficiency of photons emitted by these color centers. For example, the rate of photonic-mediated entanglement is proportional to the square of the collection efficiency as outlined in Sec. 2.5. Unfortunately, diamond has a high refractive index ($n = 2.4$), and due to total internal reflection, only up to 8% of the light can be collected from an NV center beneath an unpatterned \{100\} diamond surface [58]. Thus, it is necessary to pattern the diamond to enhance the emission and collection of the frequency range of interest.

While there are micro-devices that can increase the collection efficiency of light into a single mode [59, 60, 30], it is desirable to pattern diamond at length scales comparable to the wavelength of relevant transition frequency to achieve the greatest control over the local photonic density of states. Many photonic systems that confine light on the order of the wavelength are based on thin film substrates which are either suspended or supported by a lower index of refraction material to achieve total internal reflection. For single mode devices operating resonantly with color centers in the visible range, such a thin film has to be on the order of 200 nm in thickness. However, in contrast to many other semiconductor materials, single crystal diamond can generally only be grown on other single crystal diamond, although single-crystal growth has been demonstrated on iridium substrates [61]. The lack of single-crystal diamond grown on a sacrificial substrate precludes the use of an underlying sacrificial layer or lower index material for optical confinement. The first two sections of this chapter
review previous techniques used to make suspended nanophotonic structures in diamond. These techniques were used by collaborators to fabricated some of the structures measured during the course of the thesis. The final chapter discusses in detail a novel fabrication technique developed in the course of this thesis.

3.2 Thin Film Fabrication

*Portions of this section have appeared in Reference [62].*

3.2.1 Thin Film Creation

There are many methods for the fabrication of diamond thin films from bulk diamond substrates. Focused ion beam (FIB) milling can be used to separate small diamond slabs from the bulk [63]. However, the highly physical nature of the ion bombardment causes crystal damage as evidenced by Raman spectroscopy, photoluminescence [63], and transmission electron microscopy [64]. Reactive Ion Etching (RIE) of slabs causes less crystal damage [64] in the final slabs, allowing for spin coherence times approaching 100 μs [65]. However, this RIE method only allows for the production of small (≈ 10 × 10 μm²) membranes, which limits the ability to post-process and fabricate more complex photonic structures.

There has also been work towards the separation of a diamond film from the bulk via the controlled creation of a damage layer. MeV ions accelerated at the diamond crystal will stop at an energy-dependent depth. The damage caused by collisions with the lattice will create a well-localized graphite layer that can be removed via a wet-etch step [66, 67]. Crystal damage is inevitably induced in the removed membrane. However, this can be mitigated with an etch of the damaged side [68] and subsequent diamond overgrowth [69, 70, 71], allowing for defect centers with high quality spin properties [70].

Finally, one can start from diamond membranes of a few 10s of μms and further thin the membranes via RIE [72]. While this can create high quality diamond membranes with precisely tuned average thickness, it is difficult to get a consistent thickness across a full substrate due to initial variations in thickness.
3.2.2 Patterning

Even after a diamond membrane has been fabricated and transferred to a sacrificial substrate it is still difficult to pattern at the nanoscale. Hydrogen silsesquioxane (HSQ) resist is a high resolution electron beam resist that can be used to pattern diamond [73]. Its modest intrinsic selectivity to standard diamond RIE etch recipes can be enhanced by post-development electron curing [74]. The etch selectivity can be further enhanced with other mask layers patterned via lift-off, or an initial short dry or wet etch step. Such recipes have been used to demonstrate diamond nanowires [75], suspended waveguide and nanobeam cavities [76, 77, 78], diamond plasmonic apertures [73], and gratings [79].

FIB milling of diamond is a maskless process which can be used for fabrication of diamond photonic devices [80, 81, 82]. The spatial resolution is limited by the ion beam width. This gives several advantages for diamond patterning: a mask is not required, eliminating the need for special handling or resist spinning, and optical isolation from the bulk is achieved simply by tilting the stage to etch at an angle relative to the beam and to undercut the structure [80]. FIB milling has been used to demonstrate nanobeam cavities [80] and free-standing, undercut bridge structures [83] in bulk diamond, and two-dimensional (2D) photonic cavities [81] in a single crystal diamond layer on a buffered Si substrate. However, this technique is limited by the long milling time. Moreover, inclined side walls [81] lead to limited cavity quality-factors, and the residual damage to the diamond material results in reduced color center properties as well as additional optical and spin background. The material damage from ion milling can be partially removed either by acid treatment and oxidation [81] or using electron beam induced local etching [83]. This minimizes the optical losses and fluorescence background from the ion contamination, though has not been shown to remove it entirely.

To avoid spin-coating small diamond samples and exposing them to electron, ion, or UV radiation, a novel nanofabrication technique [84] relying on pre-patterned mask transfer was developed. Instead of defining a mask directly on the diamond substrate, a frameless single-crystal silicon membrane mask is pre-patterned from (SOI) samples and placed onto the diamond substrate silicon using membrane-transfer techniques [84]. This enables pattern transfer with feature sizes down to 10 nm, etch selectivity of over 38 for the subsequent oxygen RIE, and automatic positioning of ion implantation apertures with respect to the photonic structure [85, 86] with alignment accuracy guaranteed by the EBL writing. This
method leverages all the high quality Si fabrication techniques that have been developed.

By applying one of two complimentary transfer techniques, both small and large masks can be placed on diamond substrates with sizes from $200 \times 200 \mu m$ to $1 \times 1 mm$, as required by the sample size to be patterned. For small masks, a pick-and-place method is applied based on nanomanipulation of a polydimethylsiloxane (PDMS) adhesive attached to a tungsten probe tip. For large masks, a stamping approach is used with a transparent polytetrafluoroethylene (PTFE) sheet [84]. After the pattern is transferred to diamond by oxygen etching, the Si membrane masks are mechanically removed, avoiding solvent-based mask removal procedures on diamond substrates.

The Si mask patterning was applied for various photonic nanostructures in both bulk samples [87] and diamond thin films with sizes down to hundreds of square micrometers [72]. To pattern diamond membranes with $200 - 300 \text{ nm}$ thickness, the diamond membranes are adhered to a Si substrate, the patterned Si mask is put on the diamond membrane using the pick-and-place technique, photonic patterns are transferred into the diamond membrane by oxygen RIE, the Si mask is mechanically removed, and finally isotropic $SF_6$ plasma is applied to undercut the etched diamond photonic structure for optical measurements. While mask transfer enables patterning of small substrates, the membrane technique still suffers from low yield as it is near-impossible to achieve nm-scale thickness uniformity across a sample when thinning from tens of micrometers to 200 nm.

### 3.3 Triangular Etching of Diamond

As discussed in Sec. 3.2, the fabrication of large uniform thin film diamond samples has not yet been developed. This limits the fabrication of diamond devices that guide and capture light when 2D confinement is required. To achieve optical isolation for photonic structures in bulk diamond and to circumvent the need for large thin film diamond samples for patterning, alternative fabrication approaches have been demonstrated that enable the monolithic patterning of waveguide and cavity structures into bulk diamond.

One design concept is based on suspended devices with triangular cross-section. Such designs can be realized by angled etching, either by rotating the sample and milling by FIB [80], or by guiding the trajectory of ions in an RIE process. To guide the ions in an RIE chamber, a triangular Faraday cage is aligned to the sample to produce an angular dry
etch at 45° with respect to the diamond surface [76, 87, 77] as shown in Fig. 3-1. For the triangular structures discussed in Chapter 5.2, the pattern is defined with a high-quality silicon hard mask that is patterned on a silicon-on-oxide (SOI) wafer and released from the oxide substrate and transferred onto the diamond substrate with a hydrofluoric acid wet etch and float down technique [84]. Masks as large as 1.75 mm × 1.75 mm were transferred onto bulk diamond with this method.

![Figure 3-1: (a) Faraday cage etching.](image)

Angular etching was used to demonstrate race-track patterns with ultra high quality factor (Q factor) [76] and one-dimensional (1D) nanobeam cavities [77, 87]. More recently, a modified version of this etch was demonstrated using reactive ion etching with a collimated beam of oxygen ions which enabled higher Q factors in racetrack resonators and uniformity across a 25 mm sample [88].

Unfortunately, replicating this etch in the tools available at MIT proved difficult. While we were able to make a small Faraday cage to fit in the load lock of the SAMCO 200iP, adequate thermal and electrical contact was impossible without adding contamination.

### 3.4 Quasi-Isotropic Diamond Etching

*Portions of this section have appeared in Reference [89]*

Another technique to produce free-standing structures is a quasi-isotropic oxygen undercut. This technique is based on a combination of standard vertical RIE and zero forward bias oxygen plasma etching at an elevated sample temperature. The zero bias etching takes advantage of the low directionality of the oxygen ions and the thermally activated diamond surface leading to a quasi-isotropic chemical etch. Initial demonstrations of this technique
enabled high $Q$ racetrack resonators [90], and high mechanical quality factor waveguides [91]. However, the wavelength-scaled features needed for photonic crystal cavities were not possible within the limitations of the initial demonstration. This section gives a quick overview of the steps, and then describes the process of optimizing the method on the tools available at MIT to fulfill the strict fabrication constraints for making sensitive resonant structures.

3.4.1 Fabrication Steps

Figure 3-2: (a-k) Fabrication steps for rectangular nanobeam photonic crystal cavities, where gray, green, red, and black indicate bulk diamond, SiN, ZEP, and Al$_2$O$_3$, respectively. Steps are labeled in the figure, and details can be seen in Table 3.1.

Fig. 3-2(a-k) outline the steps of the fabrication process. A 180 nm-thick low-stress SiN layer, deposited with plasma-enhanced chemical vapor deposition (PECVD), functions as a hard mask to pattern the diamond. Electron-beam lithography defines the nanobeam
cavities (ZEP 520A exposed at 500\,\mu C/cm^2 and developed at 2°C in ortho-xylene for 90 s). Following resist development, a tetrafluoromethane (CF\textsubscript{4}) RIE step transfers the pattern into the SiN hard mask. Any remaining resist is removed with N-methyl-2-pyrrolidone (NMP) to limit contamination of the etch chamber. This pattern is transferred into the diamond (Figure 3-2(a)) using an ICP-RIE etch.

A conformal layer of 20 nm of aluminum oxide (Al\textsubscript{2}O\textsubscript{3}) produced by atomic layer deposition (ALD), protects all sides of the nanobeam cavity for the subsequent etch steps. A CF\textsubscript{4} reactive ion etch removes the top Al\textsubscript{2}O\textsubscript{3} layer, leaving only the sides covered. A second anisotropic oxygen etch removes an additional 1 \mu m of diamond. A quasi-isotropic etch then undercuts the nanobeam structure. Once the desired nanobeam height is achieved, the residual SiN and Al\textsubscript{2}O\textsubscript{3} are removed using 49% hydrofluoric acid leaving a suspended diamond structure. The sample is dried out of isopropyl alcohol (IPA) on a 180°C hotplate to reduce the surface tension during drying and achieve near unity yield of suspended devices.

<table>
<thead>
<tr>
<th>Number</th>
<th>Step</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Clean</td>
<td>5 min sonication Acetone, IPA, Methanol, DI water</td>
</tr>
<tr>
<td>2</td>
<td>Pirahna</td>
<td>3:1 Sulfuric Acid:Hydrogen Peroxide 15 min, RT</td>
</tr>
<tr>
<td>3</td>
<td>SiN deposition</td>
<td>Mixed Frequency 150-180 nm</td>
</tr>
<tr>
<td>4</td>
<td>Ash</td>
<td>100 W 5 min</td>
</tr>
<tr>
<td>5</td>
<td>ZEP</td>
<td>6 krpmin 1 min</td>
</tr>
<tr>
<td>6</td>
<td>Bake</td>
<td>180°C 1.5 min</td>
</tr>
<tr>
<td>7</td>
<td>E-spacer</td>
<td>2×4 krpmin 1 min</td>
</tr>
<tr>
<td>8</td>
<td>E-beam</td>
<td>125 keV, 500pA, 500 \mu C/cm\textsuperscript{2}</td>
</tr>
<tr>
<td>9</td>
<td>E-spacer removal</td>
<td>10 s dip DI water</td>
</tr>
<tr>
<td>10</td>
<td>Development</td>
<td>90 s 2°C</td>
</tr>
<tr>
<td>11</td>
<td>SiN Patterning</td>
<td>CF\textsubscript{4}, 10 mT, 200 W 70 nm/min</td>
</tr>
<tr>
<td>12</td>
<td>ZEP removal</td>
<td>NMP overnight</td>
</tr>
<tr>
<td>13</td>
<td>O\textsubscript{2} Ash</td>
<td>100 W 3 min</td>
</tr>
<tr>
<td>14</td>
<td>Anisotropic Etch</td>
<td>O\textsubscript{2}, 1 mT, ICP 500 W, RF 240 W, 3 min, 10 s</td>
</tr>
<tr>
<td>15</td>
<td>ALD Al\textsubscript{2}O\textsubscript{3}</td>
<td>220 cycles, 250°C</td>
</tr>
<tr>
<td>16</td>
<td>Top ALD removal</td>
<td>CF\textsubscript{4}, 10 mT, 200 W 70 nm/min 5-10 nm/min</td>
</tr>
<tr>
<td>17</td>
<td>Anisotropic Etch</td>
<td>O\textsubscript{2}, 1 mT, ICP 500 W, RF 240 W, 5 min</td>
</tr>
<tr>
<td>18</td>
<td>Isotropic Etch</td>
<td>O\textsubscript{2}, 200°C, 20 mT, ICP 900 W, RF 0 W, ~ 85 min</td>
</tr>
<tr>
<td>19</td>
<td>Mask removal</td>
<td>HF 10 min</td>
</tr>
<tr>
<td>20</td>
<td>Dry</td>
<td>180°C out of IPA</td>
</tr>
</tbody>
</table>

Table 3.1: Full process for making suspended beams, with a cross section of ~ 250×250 \textwidth.

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3.4.2 Process Optimization

Mask Choice and Patterning

A mask should be both easy to pattern and provide a good etch selectivity. We began with Hydrogen silsesquioxane (HSQ) as an etch mask, as it has been shown to provide up to 13:1 etch selectivity for an $O_2$ diamond etch [92, 75, 76, 77, 78, 73, 79]. We used a 5 nm Cr layer between the diamond and the HSQ as a conduction layer for EBL, a reflective layer for the height sensor, and an adhesion layer. Unfortunately, this led to further issues as it was necessary to remove the Cr layer before the initial anisotropic oxygen etch through a Cl plasma etch which could ruin the optical and spin properties of NVs. Moreover, the selectivity of HSQ is modest.

As HSQ is similar in composition to SiO\(_2\), we tried evaporated SiO\(_2\). Unfortunately, it also did not adhere well to diamond and demonstrated stress-induced cracking. SiO\(_2\) produced with PECVD also cracked due to stress. To combat this, we deposited low-stress PECVD SiN. The low stress deposition was achieved with a mixed frequency deposition (alternating between 13.56 MHz for 7.5 s and 380 KHz for 2.5 s, at 30 W at 900 mtorr, 300°C with 1960 sccm N\(_2\), 40 sccm SiH\(_4\) and 40 sccm NH\(_3\)). These deposition conditions give a low-stress SiN layer with a deposition rate that varies depending on the tool conditioning but is approximately 18 nm/min. The etch selectivity for the anisotropic diamond etch described below is 30:1, and there was no obvious degradation of the SiN mask during the quasi-isotropic diamond etch. For the fabrication of waveguides and 1D PhC nanobeams with cross sections of $\sim 250 \times 250$ nm, we use a mask thickness of 150-180 nm. For wider or thicker structures a thicker mask is necessary [93].

ZEP 520a, a high resolution positive-tone electron-beam resist, is used to define the structures. We spin the resist at 6,000 rpm for 1 minute, and then bake at 180°C for 1.5 minutes. We use E-spacer, a conductive polymer suspended in water, to prevent charging during the e-beam write. We spin two layers, each at 4,000 rpm for 1 minute. The patterns are written with an Elionix F125 which has a 125 kV electron beam. We use 500 pA beam current and a dwell time of 0.01 µs with 500,000 points in a $500 \times 500$ µm write field for a dose of 500 µC/cm\(^2\). The E-spacer is removed directly after writing with a 10 s dip in DI water. We develop the written structures in ortho-Xylene at 2°C for 90 s. This cold temperature allows for slow, controlled development for increased repeatability and higher

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The SiN is patterned with a tetrafluoromethane (CF$_4$) RIE at 10 mTorr and 200 W. This etches the SiN at ~60-70 nm/min, depending on the chamber condition. The etch rate is calibrated on a test SiN on Si sample before each etch. Due to RIE-lag we etch for 1.5 times the breakthrough time to ensure that the holes of the photonic crystal cavities are fully etched into the mask. The ZEP is removed completely after 6 minutes of the etch. Thus, we etch for approximately 4 minutes to ensure the holes are fully etched and the SiN is left intact. The remaining ZEP is removed with an overnight soak in N-methyl-2-pyrrolidinone (NMP) at room temperature. Finally, the sample is cleaned with a 100 W ash for 3 minutes to ensure all the resist is removed and does not contaminate the ICP-RIE chamber.

Anisotropic Oxygen Etch

It is necessary to optimize the diamond anisotropic etch to be as smooth and straight as possible. Sidewall roughness causes scattering, reducing the experimental $Q$ of nanophotonic cavities, and sidewall slant will ruin the anisotropic etch of the Al$_2$O$_3$ (Fig. 3-2(c)), as the Al$_2$O$_3$ will also be removed from the sidewalls if they are slanted.

Various mixtures of Argon and Chlorine plasmas can be used to etch diamond, but there is evidence that Cl atoms remain on the diamond surface and can lodge themselves deep into the diamond lattice during the etch process, ruining the spin coherence properties of NV centers. Thus we use a purely O$_2$ etch.

All diamond etches are done in a SAMCO 200iP, ICP-RIE tool, which allows the user to control the plasma density and bias voltage independently. A load lock system ensures that the chamber is always kept under vacuum, improving etch repeatability. We also only etch diamond in the chamber which eliminates micromasking due to deposition of contaminants from the chamber's sidewalls. There are five parameters that can be tuned to achieve the desired etch properties: bias, ICP power, pressure, flow rate, and temperature.

As we are optimizing for a vertical smooth etch, we would like a long mean free path for the oxygen ions to minimize collisions between ions, such that all ions are perpendicularly incident at the surface. Thus, we etch at low pressure (0.4 Pa). Tests at higher pressures showed a qualitative increase in surface roughness and slanted sidewalls under SEM evaluation. We also work at room temperature (30°C) and find that this is adequate. The etch begins with a higher pressure step to ensure that the plasma sparks consistently. We
use a flow rate of 30 sccm as the plasma consistently sparks and is maintained at that flow rate (this is not the case at 15 sccm). To first order, increasing the plasma power increases the etch rate, and increasing the bias power increases etch rate and decreases the sidewall slant. Our final recipe uses 240 W RF power and 500 W bias. In this regime, the etch rate is linearly proportional to the bias power. We pick a low enough etch rate such that the initial fluctuations in plasma density do not effect the final etch depth within our measurement accuracy.

The initial anisotropic oxygen etch step is ~2.5 times as deep as the final desired cavity height. This ensures that the beam is thicker than the desired final height when it achieves a rectangular cross section, allowing for precise tuning of the final beam thickness. An SEM of a photonic crystal nanobeam cavity after the initial anisotropic etch is shown in Fig. 3-3(a) with an intact SiN hard mask layer. The second anisotropic etch is optional, but reduces the amount of quasi-isotropic etch time necessary to undercut the structure.

![SEM images](image)

Figure 3-3: (a) An SEM of a diamond photonic crystal nanobeam cavity after the initial anisotropic etch. (b) An SEM of the cavity during the isotropic etch, before mask removal. (c) An SEM of the suspended cavity after mask removal.
Conformal Coating

The initial demonstration of quasi-isotropic etching for the formation of rectangular suspended structures in diamond used a conformal coating of LPCVD SiN to protect the sidewalls during the isotropic etch [90, 91]. However, when we used PECVD SiN to coat the sidewalls, it did not conformally coat the holes of the photonic crystal cavity (~200 nm in radius), and thus did not protect the sidewalls of the holes in the subsequent steps. Thus, we moved to atomic layer deposition (ALD) which can conformally coat all surfaces. We tried both HfO and Al₂O₃, and used Al₂O₃ in the end as it could be etched with the same CF₄ etch as the SiN and is removed in the HF acid etch that also removes the SiN mask. We use 220 cycles of ALD, which gives a ~22 nm layer. We see no measurable degradation of this layer during the isotropic etch. It is also thin enough to allow periodic SEM measurements of the nanobeam height during the isotropic etch, as seen in Fig. 3-3(b), which allows precise tuning of the beam height during the isotropic etch.

Quasi-Isotropic Oxygen Etch

The quasi-isotropic etch is a nearly entirely chemical etch with facet-dependent etch rates. While we did not do a quantitative test of the effect of temperature on facet etch rates, increasing the temperature from 30°C to 200°C results in a significantly increased etch rate. We also create a strong plasma with high pressure and high power to increase the density of O₂ ions and to remove any directionality from the etch. We found that lower pressures resulted in a bumpy surface. Recently, the bias power has been varied to control the directionality of the etch to fabricate pedestals with a range of shapes [98].

The optimized quasi-isotropic etch parameters are 900 W RF, no forward bias, 3 Pa, and at 200°C. The etch rate is facet dependent with the {110} facet etching fast and the {100} facet etching slowest [91]. We use {100} diamond and align the beams to the {110} facet so that the beam underetches completely before slowly etching up the {100} facet, allowing precise tailoring of the final beam thickness. To reach a final thickness of 230 nm of a 250 nm wide beam with an initial anisotropic etch of 610 nm and a second anisotropic etch of 1 μm, the total etch time is approximately 85 minutes, though we do one etch of 60 minutes and then iteratively etch in steps of 5 minutes and monitor the beam thickness with the SEM. Critically, this is possible because there is contrast between the electrons scattering by 20 nm
of Al₂O₃, and 20 nm Al₂O₃ with diamond. This can be seen clearly in Fig. 3-3(b).

**Mask Removal**

When the beam is the desired thickness, we remove both the SiN and Al₂O₃ with an aqueous 49% hydrofluoric acid (HF) etch for 10 minutes. This does not etch diamond at all. While it is not ideal to use such high concentration HF for safety reasons it allows us to cleanly and completely remove both masks in one step while the diamond is unaffected. The HF is rinsed for 10 minutes in water and then transferred to IPA, taking care to keep the diamond submerged at all times. We dry the diamond by placing it on a Si wafer at 180°C out of IPA. This reduces the surface tension during drying, increasing the yield of structures. We initially used a critical point dryer, but found that it was not necessary due to the strength of diamond.

### 3.5 Results

Faithful fabrication of simulated cavities relies on a rectangular cross section and a smooth bottom facet. To evaluate this, we break beams and place them on their side as seen in Fig. 3-4 so we can evaluate the cross-section and compare the top and bottom facets. The bottom facet (Fig. 3-4(b)) does have a different profile than the top, and the holes are smaller due to an increase in sidewall slant of the high aspect ratio holes. A more quantitative understanding of the etch could be achieved via FIB milling and direct measurements of the cross section.

![Figure 3-4: An SEM of a similar cavity as shown in Fig. 3-3 broken and flipped to compare the top (a) and bottom (b) facets. Scale bars show 1 μm.](image)

Obtaining high yield of suspended structures can be difficult due to the stress exerted during drying and the fragility of high aspect ratio structures. Luckily, diamond is the
hardest naturally occurring material in nature which helps us get near unity yield of sus-
pended nanobeam structures. We tested the structural stability of structures with up to
\( \sim 250 \text{ nm} \times 250 \text{ nm} \times 20 \text{ \mu m} \) with tethers on one or two sides. Near-unity yield is maintained
for all the structures tested, so it is possible we could fabricate longer structures. A variety
of such 1D suspended structures is seen in Fig. 3-5(a-c).

![Figure 3-5](image)

Figure 3-5: (a) An SEM of a one-side photonic crystal nanobeam cavities for coupling to
a PIC (see Ch. 4-5). (b) An SEM of an alligator cavities (see Ch. 4). (c) An SEM of a
double-sided photonic crystal for coupling to a PIC (see Ch. 5).

All the results and SEMs discussed so far have been fabricated on single crystal diamond
with a \{100\} top facet as that is most commonly available commercially. However, it will
ultimately be necessary to fabricate cavities in \{111\} diamond for optical alignment between
the NV emission dipoles and the cavity mode polarization [12]. Unfortunately, it is not
possible to take advantage of the large difference between etch rates of the \{110\} and \{100\}
facets. Moreover, the orientation of the crystal planes within the diamond was not known
a priori in the samples used. Fig. 3-6(a,b) show the uneven etching that results from the
quasi-isotropic etch acting along a random etch plane. The nanobeam suffers from sidewall
roughness and uneven thickness, although it is more uniform than the bottom surface would

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suggest. It is possible that with more process optimization similar results could be obtained in \{111\} diamond as in \{100\} diamond.

For applications such as monolithic photonic integrated circuits in diamond for quantum [87, 99] or nonlinear [100] photonics it would be helpful to have wafer-scale substrates. While the size of high quality single crystal diamond pieces is limited to only a few millimeters due to growth constraints, high purity polycrystalline diamond can be grown on the wafer scale, and NV centers in polycrystalline diamond have been shown to have similar spin and optical coherence properties to bulk single crystal diamond [101]. Initial tests of applying the quasi-isotropic etch technique to polycrystalline diamond proved successful as long as the structures stayed within the grain boundaries as seen in Fig. 3-6(c,d). Unfortunately, the diamond at the grain boundaries is lower quality than the diamond within the grains due to a higher dislocation density [101], and thus etches significantly faster, precluding the fabrication of devices across grain boundaries in this first iteration. However, this could be overcome if the grain boundaries were pre-mapped and guiding structures were enlarged over the grain boundaries.

![Figure 3-6](image-url)

Figure 3-6: (a) An SEM of a nanobeam etched into \{111\} diamond. (b) An SEM of a wheel etched into \{111\} diamond to test angle dependence of the quasi-isotropic undercut. (c,d) SEMs of ring resonators coupled to single mode waveguides etched into polycrystalline diamond.
3.5.1 Failure Modes

The fabrication procedure presented in this chapter has enabled the high-yield fabrication of suspended nanophotonic structures from bulk diamond. However, the process was developed over months and more than one hundred trials. Two of the most common failure mechanisms are shown in Fig. 3-7. Fig. 3-7(a,b) show the result of a combination of over-development, over-etching of the SiN mask, and over-etching of the ALD layer. This, along with degradation due to the anisotropic O$_2$ etch steps results in near-complete removal of the SiN, leaving only a 20 nm thick wall of Al$_2$O$_3$.

![Figure 3-7: (a,b) SEM of structures with over-etched SiN, leaving walls of Al$_2$O$_3$ but no protection on the top of the diamond. (c) SEM of a nanobeam PhC cavity which should have holes with decreasing radius at the center of the cavity though the smallest holes did not come out. The inset shows the SiN mask of the same structure.

Fig. 3-7(c) shows a nanobeam photonic crystal cavity which should have holes of decreasing radius towards the center of the cavity. While the large holes are etched completely, the second smallest holes are not etched fully, and the smallest holes are not seen at all. The inset of Fig. 3-7(c) shows the SiN mask. Although the mask seems to have holes, we could not determine the depth. This is a result of a combination of under-development and under-etching. This can be remedied with careful stabilization of the development temperature to achieve consistent development times. It can also be remedied by over-etching the SiN.
mask to ensure that the holes are completely etched, even with RIE-lag, although as seen in Fig 3-7(a,b) this can lead to further troubles.

To increase the yield of our fabrication, we carefully calibrate the etch steps. Furthermore, when we are working with high purity (>1 ppb N) diamonds which are more expensive and can be difficult to obtain, we fabricate a low purity diamond patterned with the same parameters before the high purity diamond. Finally, we also design structures that are robust to these fabrication imperfections by always using holes with the same radius (Sec. 4.3.1) or even by designing cavities without holes (Sec. 4.3.3).

3.6 Outlook

The quasi-isotropic fabrication technique developed and demonstrated here allows for the creation of high quality suspended nanophotonic structures from bulk diamond. A detailed analysis of the 1D photonic crystal cavities created with this method is given in Ch. 4, showing record high quality factors and high uniformity across samples. 2D photonic crystal cavities have also been demonstrated [93].

However, there are still many improvements that can be made. We are currently limited to etching one set of dimensions on one chip (e.g. we can only successfully fabricate only \( \sim 250 \text{ nm} \times 250 \text{ nm} \) nanobeams or \( \sim 1 \mu\text{m} \times 250 \text{ nm} \) 2D structures on one chip, but not both) because to achieve a rectangular cross section for the wide structures, we will have completely etched away the narrow structures. However, the etch rate is strongly dependent on the trench size surrounding the beam, with smaller trenches giving slower etch rates. Thus it could be possible to tailor the trench size and fabricate structures of varying sizes on the same chip. However, the exact relation between trench size and etch rate has not been studied. If this relationship can be quantified — for instance with FIB milled cross sections — the utility of this etch would increase and it would be possible to successfully fabricate more complicated structures. Finally, a more quantitative understanding of the etch rates of different diamond planes would help in transferring this etch to \{111\} and polycrystalline diamond.

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Chapter 4

Cavity Enhancement of NV Emission

4.1 Introduction

Many impressive experiments have been performed with NV centers found deep in the diamond lattice. However, even with micro-patterned structures for collection enhancement [60, 59, 30] and engineered collection optics, the rate of collection of ZPL photons is fundamentally limited by the radiative lifetime (13ns), the Debye-Waller factor (3%), and the finite collection efficiency of these structures (75% in simulation [59]). Because of these limitations, the highest demonstrated collection rate of ZPL photons is only 35kHz [30] at saturation.

Direct engineering of the local electro-magnetic environment of the NV center can improve the spin-photon interface through spontaneous emission rate modification of cavity-resonant transitions. In particular, the Purcell Effect describes the change of a quantum emitter’s spontaneous emission rate due to an engineered density of states. The maximum enhancement of emission at the resonant frequency ($\lambda_c$) is described by the Purcell Factor [102]:

$$F_p^{\text{max}} = \frac{3}{4\pi^2} \left( \frac{\lambda_c}{n} \right)^3 \left( \frac{Q}{V} \right)$$  \hspace{1cm} (4.1)

where $n$ is the refractive index of diamond, $Q$ is the quality factor of the cavity mode, and $V$ is the mode volume of the cavity mode. In experiments, the measured Purcell factor is reduced by non-radiative transitions and spatial and polarization misalignment between the NV dipole transition and the cavity field. To maximize the enhancement, it is necessary to use cavities with high $Q$ and low $V$. Photonic crystal cavities are a natural choice as
they have can small mode volumes \( V \sim (\lambda/n)^3 \) and high quality factors \( Q \sim 1 \times 10^6 \) in simulation.

In analogy to the electronic bandgap in materials that arises from the periodic potential created by the crystal structure, the patterning of a photonic crystal (PhC) can lead to a photonic bandgap, prohibiting the propagation of certain frequencies of light. In this thesis, we discuss only 1D photonic crystals in which periodic patterning in 1D prohibits propagation along that direction due to deconstructive interference. The light is confined in the other two directions by total internal reflection. After the successful formation of a 1D bandgap, a defect in the PhC creates a localized spatial mode supporting the existence of a narrow band of frequencies. It is also possible to engineer the loss channels for high efficiency collection into a single well-described spatial mode (for instance a single fiber or waveguide mode).

This chapter discusses the design, simulation, and measurement of one-dimensional photonic crystal cavities resonant with the NV center’s ZPL. The fabrication procedures used to create these cavities are described in detail in Ch. 3. Various designs are explored in order to achieve cavities with high \( Q \) factors and low mode volumes while ensuring that the NV center remains as far as possible from all surfaces to limit spectral diffusion.

4.2 Triangular Cavities

*Portions of this section have appeared in Reference [87], and Igal Bayn fabricated the measured cavities.*

Initial demonstrations of suspended diamond structures used a triangular etching technique (details in Ch. 3). Although the triangular cross section is unconventional, periodic structuring can create both TE and TM bandgaps, allowing for the creation of high \( Q \), low \( V \) modes in simulation [87, 80, 77].

A bandgap is created by a periodic series of rectangular holes in a triangular nanobeam. The rectangular holes (shorter in the propagation direction) allow for a stronger index contrast which leads to a larger bandgap than achievable with square holes. A defect region is created by parabolically increasing the lattice constant from \( a_7 \) at the center of the cavity to \( a_0 \) in the waveguide regions over \( N = 7 \) periods according to \( a_N = a_0 + (N/N_0)^2 \) \( a \). 3D Finite-Difference-Time-Domain (FDTD) simulations in conjunction with a gradient descent
optimization algorithm are used to optimize cavity parameters for a high quality factor ($Q$) and low mode volume ($V$). As illustrated in Fig. 4-1, we found an optimal $Q/V$ ratio for parameters $a_0 = 0.9a$, $k = 0.1$, $H = 1.35a$, $W = 2a$, $W_x = 0.525W$, $W_y = 0.5a$ with a mode at frequency $a/\lambda = 0.339$ with $Q = 2.51 \times 10^6$, $V_m = 1.062 \times (\lambda/n)^3$ with 22 periods of air holes on either side of the defect. The largest loss in these unloaded cavities is in the $+z$ direction, easing characterization under confocal excitation and collection. The cavities can be loaded into one waveguide direction by removing some holes on one side of the cavity.

We fabricated the cavities in single-crystal diamond grown by chemical vapor deposition (CVD, Element6) with a nitrogen defect density of 1ppm, and thus a native NV center density of approximately 1 ppb. These NV centers serve as internal light sources that allow us to easily characterize the cavity fabrication.

In order to ensure the fabrication of cavities near the desired wavelength (637 nm) in the presence of fabrication imperfections, we produced cavities with a range of parameters: $a = 220 - 240$ nm, $W = 2a$, $W_x = 0.29W - 0.5W$, and $W_y = 0.15W - 0.25W$. We
characterized these cavities by confocal spectroscopy, using an air objective with NA=0.9 and 532 nm laser excitation. Fluorescence was detected by a silicon avalanche photodiode (APD, Perkin Elmer). Cavity Q factors as high as 3000 were measured (Fig. 4-1). The discrepancies between simulated and measured Q factor and resonant wavelength are due to many factors. First, the diamond had an initial surface roughness of \( Ra = 30 \) nm, which could be reduced in future runs through mechanical or chemical polishing. Second, it is difficult to fabricate rectangular holes with sharp corners, and the SEMs show ellipsoidal holes (Fig. 4-2(a)). This discrepancy could be solved by optimizing a structure with circular or ellipsoidal holes. Third, the fabricated cross-section of the beam does not have the intended shape as seen in the SEM of a FIB milled cross section seen in Fig. 4-2(c). Again, this could be corrected in subsequent rounds of optimization and fabrication. Finally, the SEMs show significant surface roughness (Fig. 4-2(a,b)) on the cavity sides which will cause scattering and increased losses. This could be improved with fine tuning of the fabrication recipe.

4.3 Rectangular Cavities

While the triangular PhC cavities showed promise, fabrication imperfections limited the \( Q \), and improvements proved difficult with the tools available at MIT. Moreover, the triangular waveguide modes give lossy coupling to standard rectangular waveguides used in Ch. 5. The development of the quasi-isotropic diamond etch for the fabrication of suspended beams from bulk diamond allowed for the fabrication of conventional 1D PhC cavities with rectangular a crosssection.

4.3.1 L0 Nanobeam cavities

*Portions of this section have appeared in Reference [89]*

Cavity Optimization

We design PhC nanobeam cavities to support a low-mode-volume (\( V \)) mode with a high quality factor \( Q \) at the NV ZPL (637 nm), and with the electric-field maximum concentrated in the diamond. A schematic of the nanobeam cavities is shown in Fig. 4-3(a). The design process begins with approximate cavity parameters derived from band structure simulations
and optimizes the cavity $Q$ of the lowest order TE mode (intensity profile shown in Figure 4-3(b)) at $\lambda_{NV} = 637$ nm by FDTD simulations. The final design consists of a diamond waveguide ($W = 250$ nm and $H = 230$ nm) periodically patterned with holes with radius $r = 58$ nm and spacing $a = 192$ nm. The defect supporting the cavity mode is introduced by linearly decreasing the hole spacing to $a = 171$ nm over 5 periods. With 25 holes on either side of the cavity region, this cavity has a radiation-limited $Q$ factor of greater than 1 million in simulation. The cavity also supports a TM mode (Figure 4-3(c) shows the intensity profile) at 615 nm with a $Q$ of 13,000 in simulation, as well as other TE modes with mode maxima in the outer hole regions with lower $Q$ values – 5,500 and 2,500 in simulation at 649 nm and 653 nm respectively shown in Fig. 4-3(d,e). These higher order modes are denoted as M3 and M4 in the measured spectra in Figures 4-5 and 4-6.

![Figure 4-3: (a) Artistic rendering of an array of rectangular nanobeam cavities fabricated from bulk diamond. (b) Re($E$) for the first order TE mode. (c) Re($E$) for the first order TM mode. (d) Re($E$) for a lower frequency TE mode localized in the mirror holes (Mode 3 in text). (e) Re($E$) for a lower frequency TM mode localized in the mirror holes (Mode 4 in text). Scale bar for (b-e): 1 μm](image)

**Collection Enhancement**

To increase the overall entanglement rate through increasing the collection rate of ZPL photons, it is not enough to increase the emission rate into the desired frequency mode. It is also important to engineer the cavity’s loss channels for increased collection into the desired single spatial mode. We will collect light into a single mode waveguide for routing through a photonic integrated circuit, which provides a scalable architecture for entanglement generation and quantum information processing as described in Ch. 5. Thus, we would like to
engineer photonic structures that are loaded into the diamond waveguide mode.

Unfortunately, optimized 1D PhC cavities generally minimize loss into the waveguide as that is the only propagation direction with a bandgap. For instance, the optimized 1D PhC cavity has only $\sim 1\%$ loss into the waveguide, and thus does gives little enhancement in the number of ZPL photons collected into a single waveguide mode. Work has been done in silicon to engineer high $Q$ cavities directly loaded into the feeding waveguide [103]. However, the results could not be replicated in diamond, perhaps due to the lower refractive index. Instead, we simply remove holes from one side of the cavity which naturally loads the mode into the waveguide, although the $Q$ factor is lowered. We pick the number of holes that maximizes the collection enhancement of the frequency of interest. Fig. 4-4(a) shows the decrease in $Q$ and change in the percentage of the loss collected into the waveguide mode as the number of holes on one side of the cavity is reduced. Fig. 4-4(b) shows the overall enhancement to the rate of photons collected into a single waveguide mode. At the optimum ($N = 13$), 60% of the light is collected into the waveguide mode.

![Figure 4-4: (a) Effect of the number of mirror holes on the $Q$ and the percentage of loss collected into the diamond waveguide mode. (b) An approximation of the collection enhancement for increasing number of holes.](image)

Measurements

We fabricated these nanobeam cavity designs in a $3\text{mm} \times 3\text{mm} \times 0.3\text{mm}$ single-crystal diamond with a $\{100\}$ top face grown by chemical vapor deposition (CVD, Element6) with a
nitrogen defect density of less than 1 ppm, and thus a native NV density of approximately 1 ppb. We measured the fabricated cavities at 4 K by photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy. The native population of NV centers, excited using a 532 nm laser, provides an internal light source for the cavity. The cavity PL is collected with a 0.9 NA objective and resolved on a spectrometer with a resolution of 0.06 nm which corresponds to \( Q \sim 14,000 \). To ensure that we fabricate cavities with resonances near the ZPL frequency even with systematic errors (e.g. in beam thickness due to the resolution limit of the SEM thickness measurement, or in hole size due to exposure or development inaccuracy), we sweep the width of the beam in steps of 5 nm from \( W = 240 \) nm to \( W = 260 \) nm and the radius of the holes in 3 nm steps from \( r = 52 \) nm to \( r = 64 \) nm. Increased width should increase the resonant wavelength, while increased radius should decrease the resonant wavelength.

Fig. 4-5 shows two cavity spectra: Cavity A (\( W = 260 \) nm, \( r = 55 \) nm) and Cavity B (\( W = 245 \) nm, \( r = 58 \) nm). Fig. 4-5(a) shows the cavity-modified PL spectrum of the NV ensemble at Cavity A under 532 nm excitation. Four modes appear in the spectrum, corresponding to the first-order TE and TM modes (profiles shown in Figure 4-3(b,c)), as well as the higher-order modes, M3 and M4. Fig. 4-5(b) shows the high-Q first-order TE mode at 636.1 nm, as well as the inhomogeneously-broadened ZPL of the excited ensemble of NV centers at 637 nm. The linewidth of the cavity mode is limited by the spectrometer’s resolution, as confirmed by comparing to the spectrum of a sub-0.5 MHz laser at the same wavelength. Fitting the spectrum with a pseudo-Voigt function \([19]\) takes into account the effect of the Gaussian spectrometer filter on the Lorentzian cavity spectrum. This fit indicates a lower bound of \( Q \geq 16700 \), above the resolution of the spectrometer (\( Q \geq 14,000 \)).

The spectrum of Cavity B (Fig. 4-5(c)) reveals the first-order TE mode overlapping with the inhomogenous distribution of ZPLs in the NV ensemble. We performed PLE spectroscopy to better measure the \( Q \) of this cavity and study the cavity-enhanced excitation of the NV ensemble. The PLE consisted of NV phonon side band (PSB) detection (filtered > 650 nm) while scanning a \(< 0.5 \) MHz-linewidth laser over the NV ZPL and cavity frequencies. The inset of Fig. 4-5(c) shows the PLE spectrum of the unpatterned nanobeam 7 \( \mu \)m from the cavity center. This reveals an inhomogeneously broadened absorption spectrum of the excited population of NV centers centered at 470.48 THZ (637.2 nm) with a full width half
Figure 4-5: (a) PL spectrum of Cavity A with lowest order TE and TM cavity peaks fit with Lorentzian functions. (b) Zoom in of the fundamental TE mode of Cavity A. The red line shows a fit to the spectrometer-limited cavity peak, fit with a pseudo-Voigt function. (c) PL spectrum of Cavity B showing the inhomogeneously broadened ZPL emission from the ensemble of excited NVs at 637 nm and a cavity peak at 637.1 nm. (d) Photoluminescence excitation (PLE) spectrum of Cavity B showing a PLE peak at NV ZPL position and a cavity-enhanced PLE peak 100 GHz detuned center from the inhomogeneous ZPL distribution. The inset shows a PLE measurement of the same nanobeam 7 µm from the cavity center showing only the inhomogeneous distribution of NV centers in the sample.
maximum of 42.7 GHz. A PLE scan at the center of the cavity shows a second peak that
is absent on the rest of the sample; we attribute this to the cavity-enhanced absorption of
NV centers coupled to the cavity mode. A Lorentzian fit of the peak reveals a measured
cavity $Q$ of 14,700. This provides a lower bound on the $Q$ of the bare cavity mode, as the
PLE spectrum may be broadened by the interaction with the inhomogeneous distribution
of emitters coupled to this mode of the cavity [104].

![Graph showing frequency distribution of resonant frequencies](image)

Figure 4-6: (a) Frequency distribution of resonant frequencies of Mode 4 for all 25 sets of
parameters. The error bars show the standard deviation $\pm \sigma$ of the resonant frequencies
for the 5 cavities fabricated for each set of parameters. (b) The spectra of the 5 cavities
fabricated with $W = 260\,\text{nm}$ and $r = 58\,\text{nm}$ (circled in (a)) showing the distribution of Mode
3 and Mode 4.

A survey of all of the fabricated devices demonstrates the consistency and high yield
of our fabrication technique. Fig. 4-6(a) shows the measured cavity resonances of mode
M4 across the full range of parameters. The error bars show the standard deviation of the
wavelength position for the 5 cavities fabricated with each parameter set. We used mode M4
as it has the highest vertical loss of the four modes, and thus the highest SNR in spectrometer
measurements. This survey reveals the expected trends: the resonance wavelengths increase
with larger beam widths and decrease with larger holes radii. The standard deviation within
each parameter is low with an average of $\pm 2.2\,\text{nm}$ deviation from the mean in each parameter
set, showing the consistency of the fabrication process. The spectra of M3 and M4 of the 5
cavities with $W = 260\,\text{nm}$ and $r = 58\,\text{nm}$ (circled in Fig. 4-6(a) are shown in Fig. 4-6(b)).
4.3.2 L3 cavities

The L0 cavities described and measured in Sec. 4.3.1 provide high $Q/V$ even experimentally. However, a NV center at the mode maximum will only be 32 nm from the 2 holes at either side of the cavity center. This motivated an attempt to simulate a high $Q$ nanobeam cavity with 3 holes missing at the center in analogy to the L3 cavity in 2D photonic crystals [105]. The cavity $Q$ was optimized, starting with the parameters found during the optimization of the L0 cavity. Unfortunately, the largest cavity $Q$ achieved was only 20,000 and the mode volume was $>10(\lambda/n)^3$. For a design with a mode volume $V \sim 3(\lambda/n)^3$ the maximum $Q$ achieved in simulation was 6,000. The measured cavity $Q$ values of these cavities are limited to 1000. Results with single NV centers are outlined in Sec. 4.4.

4.3.3 Alligator Cavities

PhC cavities are usually formed by etching air holes into a material. For 1D PhC cavities, this ensures the largest possible modulation in effective refractive index. However, it means that an NV center optimally coupled to the cavity mode will be less than 100 nm from surfaces (Sec. 4.3.1), or the $Q$ will be low (Sec. 4.3.2). Thus, we took inspiration from "Fishbone" PhC cavities in which semicircles are etched into the side of a nanobeam [106]. Unfortunately, when we fabricated these structures mask degradation rounded the corners, and we were not able to faithfully replicate the geometry.

To overcome the fabrication constraints, we instead turned to an "Alligator" cavity design with a periodic effective refractive index created with a sinusoidal modulation of the nanobeam width as seen in Fig. 4-7(a,b). These cavities were first explored by the Kimble group at Caltech to couple neutral trapped atoms to PhC cavities [107]. There are many parameters that could be modified to create a localized mode. We chose to reduce the depth of the modulation linearly over five periods. This increases the minimum distance between any surface and an optimally coupled NV center at the center of the beam to 92 nm in the optimized design. The mode profile ($|E|^2$) of the optimized cavity with $Q = 1.4 \times 10^5$ with is shown in Fig. 4-7(a).

We fabricated an array of Alligator cavities with a parameter sweep around the set of optimized parameters. An SEM of one of the fabricated cavities is shown in Fig. 4-7(b). We then measured a set of the cavities. The fabricated cavities were thicker than the design
Figure 4-7: (a) Mode profile ($|E|^2$) of the optimized fundamental TE mode. (b) An SEM of an Alligator cavity. (c) Spectrum of a cavity mode ($Q = 10,802$, at the spectrometer limit). (d) Distribution of $Q$ values for 15 measured cavities showing consistently high $Q$.

(−300 nm) causing the resonances to be at larger wavelengths than expected. Nevertheless, the cavities showed high $Q$ values as shown in the distribution in Fig. 4-7(d). The resolution limit of the spectrometer used for these measurements is ~11,000. The spectrum of a cavity with $Q \geq 10,802$ is shown in Fig. 4-7(c). This is near the spectrometer limit and the actual $Q$ may be higher.

4.4 Spectral Properties of Cavity-Coupled NV Centers

Coupling single NV centers to nanophotonic devices such as waveguides and cavities will boost the NV-NV entanglement rate by increasing the emission and collection rate of photons entangled with the spin. However, this is only possible if it is possible to fabricate nanostructures that do not diminish the coherence of the optical transitions of the NV center. Unfortunately, while NV centers with lifetime-limited linewidths have been demonstrated in bulk diamond [14], they suffer from spectral diffusion in nanostructured diamond due to an increase in surface charge traps.
4.4.1 Linewidths in Nanostructures Fabricated from Diamond Membranes

We first measured the linewidths of NV centers in single-mode diamond waveguides fabricated from diamond membranes [108] used in Sec. 5.3. We performed photoluminescence excitation (PLE) measurements at cryogenic temperatures (18K). Fluorescence above 650 nm was collected under repeated resonant laser line scans of several GHz around the ZPL of a single NV inside a μWG. We use a resonant laser with 200 nW of power. The laser frequency is scanned using a piezo-controlled grating in a New Focus Velocity laser. Each step in the sweep was integrated for 5 ms making each sweep take 2-3 seconds. The step speed was limited by our hardware and was more than 1000 times longer than necessary [109, 14]. A 600 μs green laser pulse of ~100 μW is delivered prior to each scan to re-initialize the NV− center into the $m_s = 0$ ground state and NV− charge state.

Multiple PLE peaks are contained in one scan due to spectral diffusion caused by the resonant laser during the long integration time of 5 ms per frequency step. To determine the fundamental linewidth, the number of PLE peaks, $n$, was determined using a threshold, and the sum of $n$ Gaussian functions were fit to the data. Finally, the full-width at half-maximum
data was binned to analyze the spectral linewidth as indicated in Figure 4-8(c).

Figure 4-8(a,b) shows a multiscan measurement of a representative NV, and an example single-line scan respectively. Figure 4-8(c) shows the distribution of the measured linewidths of signal peaks in the PLE of consecutive single-line scans of NV C, with a maximally occurring linewidth of 393 MHz. This linewidth is about 30 times broader than the expected lifetime limited linewidth of 13 MHz for the $m_s = 0$ transitions in bulk diamond [110, 14]. We partially attribute this broadening to in-situ spectral diffusion occurring during a single line scan. This broadening could be reduced by more suitable power and timing parameters.

Figure 4-8(d) shows the mean and standard deviation of the measured linewidths of three NV centers in individual waveguides. Although these measured linewidths are larger than the expected lifetime limited linewidth, they are up to 20 times lower than the previously reported linewidth of an NV in nanostructured diamond [111] and the dephasing times are within the time resolution of detection equipment, allowing for measurements of two-photon interference towards distributed entanglement [10]. However, these measurements do not characterize the full inhomogeneous linewidth due to spectral diffusion, which is the limiting factor for efficient, high-fidelity quantum network protocols.

### 4.4.2 Linewidths in Nanostructures Fabricated from Bulk Diamond

The quasi-isotropic etch introduced in Sec. 3.4 provides consistent fabrication of high quality nanostuctures from bulk diamond, but it is still not known if it supports NV centers with lifetime-limited linewidths in nanostructures. In order to characterize the effect of fabrication on the optical properties of NV centers we begin with diamond samples with only 1 ppb N, implant $N^{15}$ at 85 keV and anneal at 1200°C to promote the creation of NV centers. This creates a layer of NV centers ~100 nm deep with a random distribution in the $x, y$ plane at a density of ~1 per 5 $\mu$m$^2$. It is possible to align structures to pre-localized defects [112, 113], and work is on-going in our lab to do this on the chip-scale for > 10,000 NV centers simultaneously. However, this capability was not ready, so we simply fabricated arrays of cavities and characterized them post fabrication to locate ones with single NV centers.
Broadband Devices

We first characterize NV centers in single mode waveguides (250×250 nm). We take spectra of 19 NV centers at cryogenic temperatures (4 K) with 532 nm excitation directly after fabrication. The spread in ZPL transition frequencies seen in the top panel of Fig. 4.4.2(a) indicates strain in the diamond induced by the fabrication of these free-standing structures. To relieve the strain, we anneal the sample at high temperatures and high vacuum (1200°C, 10⁻⁸ mbar). The reduction of strain is demonstrated by the narrowing of the wavelength distribution of cryogenic spectra taken after the anneal as seen in the bottom panel of Fig. 4.4.2.

![Histogram of ZPL wavelengths before and after anneal](image)

**Figure 4-9:** (a) Top: Histogram of the ZPL wavelengths of 19 NV centers in single mode waveguides directly after fabrication. Bottom: Histogram of the ZPL wavelengths of 28 NV centers in single mode waveguides after a high temperature, high vacuum anneal. (c) Histogram of inhomogeneously broadened linewidths of 28 NV centers in single mode waveguides (after the anneal).

In order to measure the effect of spectral diffusion, we perform resonant excitation scans on 28 NV centers found in single mode waveguides. To probe an NV center's full inhomogeneous linewidth caused by spectral diffusion, we perform a slow scan, where 1 × 10⁴ data points are taken at each frequency, with a 532 nm repump before each point. The distribution of linewidths (Fig. 4.4.2(c)) indicates that all measured NV centers experience levels of spectral diffusion lower than have been previously reported for NV centers in nanostructures, with a full width at half maximum of 3.53±1.36 GHz. Unfortunately, these NV centers
still have linewidths that are almost 300 times the expected lifetime limited linewidth.

**Resonant Devices**

While these waveguides can be used to increase the collection efficiency of ZPL photons into a single spatial mode [108], it is necessary to also enhance the emission probability into the ZPL of the NV by coupling to a cavity mode with a high $Q$ factor and low mode volume. To this end, we measured NV centers coupled to L0 photonic crystal nanobeam cavities (Sec. 4.3.1). These cavities have a $Q$ of $10^6$ in simulation, and up to the resolution of the spectrometer (14,000) in experiment [89]. Unfortunately, resonant scans of NV centers coupled L0 photonic crystal nanobeam cavities show spectral diffusion of tens of GHz. A PLE scan of a representative NV with a FWHM of 30.6 GHz is shown in Fig. 4.4.2(a). This large spectral diffusion is presumably due to the proximity of etched surfaces to the NV center.

![Figure 4-10: (a) PLE scan of an NV in a L0 cavity (FWHM 30.6 GHz). (b) Spectrum of an L3 cavity with an NV at the center. (c) Averaged PLE scan of the NV seen in (c) showing the $E_x$ (5.56 GHz FWHM) and $E_y$ (3.84 GHz FWHM) transitions.](image)

We also measure L3 (Sec. 4.3.2) cavities on the same chip. These cavities have a $Q$ factor limited to 6,000 in simulation and the largest measured experimental $Q$ factor is 1,000. However, an optimally coupled NV center will be 125 nm from the top and bottom surfaces, 140 nm from the sides, and 350 nm from the holes. The spectrum of one such cavity (Fig. 4.4.2(b)) shows the modes of the cavity ($Q = 600$) as well as the ZPL of an NV. A PLE scan on a NV spatially coupled to this cavity shows the $E_x$ and $E_y$ transitions fitted with two Gaussian functions with FWHM of 5.56 GHz and 3.84 GHz, similar to the linewidths measured in the single mode waveguides (Fig. 4.4.2(c)).
4.5 Outlook

The quasi-isotropic etch technique enables the consistent fabrication of high-quality nanostructures with the desired functionalities. Considering only the properties of the cavities, it should be possible to significantly increase the entanglement rate between spatially separated NV centers. Unfortunately, the optical coherence of NV centers in nanostructures is degraded. Nevertheless, the ~GHz linewidths measured in L3 cavities are promising. Moreover, the Alligator cavities described in Sec. 4.3.3 support a high-$Q$ fundamental TE mode while ensuring that the NV center remains 115 nm from the top and bottom, 133 nm from each side, and tens of microns from any surface in the X direction. Table 4.1 summarizes these results.

<table>
<thead>
<tr>
<th>Cavity Type</th>
<th>Simulated $Q$ factor</th>
<th>Experimental $Q$ factor</th>
<th>Linewidth</th>
</tr>
</thead>
<tbody>
<tr>
<td>L0</td>
<td>$1 \times 10^6$</td>
<td>$1.4 \times 10^4$</td>
<td>30 GHz</td>
</tr>
<tr>
<td>L3</td>
<td>$6 \times 10^3$</td>
<td>$1 \times 10^3$</td>
<td>3.8 GHz</td>
</tr>
<tr>
<td>Alligator</td>
<td>$1.4 \times 10^5$</td>
<td>$1.1 \times 10^4$</td>
<td>?? \textsuperscript{1}</td>
</tr>
</tbody>
</table>

Table 4.1: Summary of cavity results. \textsuperscript{1} The distance between the NV center and sidewalls suggest that a cavity-coupled NV center could have a linewidth of $\sim 1$ GHz.

Novel designs such as the Alligator cavity, along with further improvements in surface quality, should allow for the coupling between high-$Q$ cavities and single NV centers with stable linewidths of $\leq 1$ GHz. This would bring the field to a useful regime, as a high-$Q$ cavity will decrease the radiative lifetime via the Purcell effect, which will in turn give rise to a larger lifetime-limited linewidth. Going forward, it will be important to design high-$Q$ cavities in which the surfaces are as far from the NV center as possible to limit the effects of spectral diffusion, explore surface terminations for passivation of surface charge traps, and better understand the dynamics of spectral diffusion.
Chapter 5

Photonic Integrated Circuits for Quantum Information Processing

5.1 Introduction

Free-space optical setups have enabled some of the first verifications of quantum mechanics [114, 115]. Initial implementations of photon-mediated entanglement between spatially separated quantum memories have relied on bulk free-space optics or fiber-based systems [10, 116]. However, free-space approaches are bulky and suffer from phase instability. While fiber systems can be more compact, they suffer from loss, phase instability, and polarization instability. As we strive to engineer quantum systems with tens, hundreds, and ultimately millions of qubits, the space and stability constraints prohibit the use of bulk optics or fiber-based systems. Instead, it is necessary to move to an integrated approach.

In direct analogy with the electrical integrated circuits that drove the growth of classical computing, it will be necessary to build up the architecture for a photonic integrated circuit (PIC) to enable the scaling of quantum computing. Translating distributed quantum networks of spatially separated qubits connected via photonic channels to a PIC architecture can provide scalability through low-loss, phase-stable, polarization-maintaining, and small-footprint optical routing. Further implementation of on-chip microwave control, optical detection, and classical logic will enable high rate and high fidelity entanglement generation between spatially separated quantum bits and finally quantum computation.

Over the past decade, nanofabrication techniques have been developed and perfected...
and it is now possible to fabricate single-mode waveguides with low-loss optical propagation with a variety of material platforms. This has led to the demonstration of on-chip quantum optics experiments, and the development of a platform for on-chip linear optical quantum computation (LOQC) [117] with both passive and active photonic elements [118, 119, 120]. The work in this thesis extends these quantum information processing (QIP) platforms to include the integration of quantum memories into a scalable integrated photonic platform.

This chapter discusses the feasibility of many platforms for integrated QIP with NV center quantum memories. When comparing different implementations the important figures of merit are (1) quantum node yield, (2) passive and active component fabrication quality, and (3) background fluorescence level. The chapter concludes with an outlook on the promise of fully chip-integrated quantum processors going forward.

5.2 An All-Diamond Architecture

Portions of this section have appeared in Reference [87] and [85]. Igal Bayn fabricated the diamond waveguides discussed in this section.

It is natural to first consider an all-diamond PIC. In this work, we aimed to build a chip with NV centers coupled to cavities directly loaded to diamond waveguide modes. An all-diamond network would enable efficient collection of NV fluorescence into a single spatial mode, and limit the necessary fabrication steps. In this work we began with a CVD-grown bulk diamond and created isolated triangular photonic structures as discussed in Sec. 3-1.

5.2.1 Demonstration of PIC Components

The triangular etch creates air-cladded waveguides with a triangular cross-section. The height of the beam is fixed at $H = 0.5W$ due to the 45° angle etch. A waveguide with $W = 500$ nm is single mode for 637 nm light. The TE-like and TM-like modes are shown in Fig. 5-1(a,b) respectively.

As discussed in more detail in Chapter 4, it will be necessary to couple individual NV centers to cavities to increase emission and collection into a single spectral and spatial mode for high rate and fidelity entanglement. Thus, we also demonstrated 1D PhC cavities. The design and measurements of these cavities are discussed in Sec. 4.2. These cavities can be loaded directly into the waveguide mode. Because these waveguides are suspended
and not built on a low-index substrate as is often done (e.g. in standard silicon-on-oxide platforms) it is necessary to include supports for structural integrity. We used a tapered one-sided support structure where the waveguide width is increased at the site of the bridge to minimize scattering [121]. We optimized the design of the taper using 3D FDTD simulations to minimize transmission loss. Simulations of the optimized design predict a transmission loss of 0.05dB at 637 nm.

The fabricated suspended cavity-waveguide network was mechanically stable due to the introduction of support structures every 20 μm. However, the fabricated supports did not perform as well as in simulation and caused 40-50% scattering at each support. This made it impossible to reliably measure the intrinsic diamond waveguide loss and prohibited the use of these networks for a quantum information application.

5.2.2 Targeted Implantation

In the initial demonstration of the components needed for a simple all-diamond PIC, we used diamond with a native concentration of nitrogen with 100s of NV centers per diffraction limited spot to ease characterization measurements. However, in a final implementation it will be necessary to isolate single NV centers coupled to PhC cavities. For experiments with single NV centers, we begin with high-purity diamond with a defect concentration of less than 1 ppb, implant nitrogen ions at the desired depth, and anneal the diamond to promote the creation of NV centers as described in Ch. 2. However, this leads to a random distribution of NV centers which then limits the yield of nodes in which a single NV center with sufficient optical and spin coherence is well-coupled to a PhC cavity that has a sufficient Q.

To increase the yield of nodes with a well-coupled NV-cavity system, we implemented a self-aligned lithographic technique for the targeted implantation of nitrogen ions in photonic
nanostructures [85, 86]. A Si hard mask was used to define waveguides and 1d PhC cavities as described in Sec. 5.2.1. In addition to the photonic structures, implantation apertures were defined in the same e-beam exposure with widths less than 50 nm, much smaller than the photonic features which have a minimum feature size of ~100 nm. The small width (and thus large aspect ratio) prevents oxygen ion etching due to RIE lag [96]. Thus these holes do not affect the final diamond structure. Instead, they serve only to mask the diamond during nitrogen implantation. Because the photonic structures and implantation apertures are written in the same e-beam step, these rectangular implantation apertures are lithographically aligned with the waveguide and cavity patterns and the implantation volume is restricted to the mode maxima. After implantation, the mask is removed and the sample is annealed. The NV yield per aperture follows a Poisson distribution, with a 26% single-NV yield per implantation aperture [85]. This will give an increased yield of NV centers well coupled to the cavity mode.

5.2.3 Conclusion

An all-diamond architecture is a simple path towards an integrated architecture for entangling NV-based nodes as emission can be directly collected into the waveguide mode, and high-purity diamond with no background fluorescence is available commercially. Moreover, large-scale lithography via mask transfer or direct writing can produce chip-scale devices directly in diamond, and improvements in diamond fabrication and device design could enable low-loss propagation in single mode diamond waveguides and other passive diamond components [76, 77, 99], and initial demonstrations of nonlinear photonics such as four wave mixing have been demonstrated in diamond waveguides [100].

However, any monolithic approach will suffer from scalability if the yield of the components is low. Without targeted implantation, the probability of fabricating a cavity on resonance with a well-coupled NV center is well below 1%. Even with targeted implantation, the probability of fabricating a high-\(Q\) cavity on resonance with the coupled NV center is low and requiring good optical and spin properties further restricts the node yield. This makes the fabrication of a fully connected monolithic quantum network intractable, as the number of attempts needed to create a network with \(N\) fully connected nodes with a per-node-yield of \(Y\) would take on average \(N^{1/Y}\) attempts until success. For instance, a successful monolithic PIC with 10 quantum nodes with a yield of 26% (e.g. the implantation yield shown
in Ref. [85] disregarding the need for suitable NV properties or cavity yield) would take on average more than 7,000 fabrication attempts until all N nodes are successful at once.

5.3 A Hybrid Architecture

*Portions of this section have appeared in Reference [108].*

As discussed in the previous section, building a monolithic diamond PIC for QIP with NV centers is prohibitively difficult due to challenging diamond fabrication and the low yield of quantum nodes (e.g. single NV centers with adequate optical and spin properties well coupled to a high-Q PhC cavity). Thus, we instead address the challenge of fabricating a scalable quantum network architecture with a hybrid approach in which quantum nodes based on NV centers are pre-characterized and integrated into a PIC that can be fabricated with near-unity yield. The pre-selection characterization step guarantees that every node in the PIC contains a single NV center with adequate optical and spin characteristics. With this bottom-up approach, the number of fabrication attempts necessary to create a quantum network scales linearly with the number of desired nodes. For example, a 10 node network with a 27% node yield will now take on average only 170 node fabrication attempts in contrast to the 7,000 PIC fabrication attempts necessary for a monolithic network.

5.3.1 Silicon Nitride PICs

*Carl Poitras (then in Michal Lipson’s group) fabricated the SiN waveguides used in this section*

As an initial demonstration of a hybrid architecture for QIP, we use a Silicon Nitride (SiN) photonic backbone. SiN-based photonics relies on well-developed fabrication processes [122], is CMOS-compatible [123], and has a large band gap (\(\sim 5\,\text{eV}\)) and high index of refraction \((n = 2.1)\) which makes it ideal for the low-loss routing of the visible emission of NV centers.

In a hybrid photonic network, it is necessary to engineer low-loss coupling between the diamond photonic structure into the SiN waveguide. In this proof-of-principle demonstration, each quantum node consists of an NV in a simple diamond single-mode micro-waveguide (200 x 200 nm) (\(\mu\)WG). The SiN waveguide also supports single mode propagation over the NV emission spectrum with a cross section of 400 x 400 nm.
We fabricated the diamond \( \mu \)WGs from a 200 nm-thick single crystal diamond membrane which was thinned from a 5 \( \mu \)m diamond slab produced by chemical vapor deposition. The diamond slab was thinned with anisotropic oxygen plasma etching. The diamond \( \mu \)WGs were patterned from the membrane with oxygen plasma [124] in a reactive ion etching chamber using a transferrable patterned silicon membrane as a hard etch mask [72, 84].

To create the SiN PIC, 400 nm SiN was deposited on a thermally oxidized (~4 \( \mu \)m) Si wafer in a furnace at 800°C. After a piranha clean, MA-N negative resist and electron-beam lithography were used to define the waveguides. After resist development the SiN was dry etched in a \( \text{CHF}_3/\text{O}_2 \) chemistry. The resist is stripped in an oxygen plasma and the wafer was annealed at 1200°C for 180 minutes in an ambient nitrogen environment. ~3 \( \mu \)m of SiO\(_2\) was then deposited as a cladding material with plasma-enhanced chemical vapor deposition (PECVD) [122]. To access the coupling regions, SPR positive tone resist and contact lithography were used to define 50 \( \times \) 50 \( \mu \)m windows in the SiO\(_2\) over the coupling region for the integration of the diamond. The development of the exposed resist exposes the windows, and they were etched in a buffered oxide etch until the SiN waveguides were fully uncovered. The thermally oxidized SiO\(_2\) etches slower than the PECVD SiO\(_2\) layer and acts as an etch-stop layer. Finally, the chip was spun with photo-resist to protect the devices, the chip was cleaved near the waveguide ends, and the edges were polished back to provide a clean facet for fiber coupling.

**Diamond - PIC coupling**

We investigated four structures for efficient coupling between a diamond waveguide mode and a SiN waveguide mode. We are interested in a structure which has high coupling efficiency from the diamond waveguide mode to the SiN waveguide mode and which allows for easy alignment.

To predict and optimize the coupling efficiency from an NV center in a 200\( \times \)200 nm single mode diamond \( \mu \)WG to a 400\( \times \)400 nm single mode SiN WG, we simulated our system using FDTD simulations. The NV center is represented as an electric dipole placed in the center of the diamond \( \mu \)WG, oriented perpendicular to the propagation axis of the \( \mu \)WG and 55° off-horizontal. This is consistent with a diamond slab oriented in the \{100\} direction, as we use in our experiments. For the four investigated structures, we placed Poynting flux monitors (i) to either side of the NV (ii) at each end of the SiN WG, and (iii) surrounding
the entire structure to monitor where electromagnetic power is lost. The ratio of (i) to (iii) gives the NV coupling efficiency to the initial waveguide mode, and the ratio of (ii) to (iii) yields the total coupling efficiency of the device. We simulated symmetric devices, but the percentage of light coupled into a single waveguide mode could be doubled by including a distributed bragg reflector (DBR). For each structure we swept the diamond width and height, though kept the SiN dimensions fixed as these were decided by our collaborator. We also swept the taper lengths, always finding a saturating coupling efficiency with increased length.

![Diagram](image)

**Figure 5-2:** (a) Schematic of a mode coupling device with a tapered diamond waveguide placed over a SiN waveguide. (b) Schematic of a mode coupling device with a tapered diamond waveguide placed over a gap in a SiN waveguide (also with tapered ends. (c) Simulation of the coupling efficiency and loss channels for the device shown in (a). (d) Simulation of the coupling efficiency and loss channels for the device shown in (b).

In the first device, a diamond $\mu$WG is placed on top of the SiN waveguide, and the diamond is tapered, as shown in Fig. 5-2(a). The tapering allows for adiabatic mode coupling from the initial SiN-diamond mode to the all-SiN mode. This eases the fabrication resolution needed for the underlying PIC, and allows for node placement at any point along the waveguide. To optimize the coupling, the diamond taper length ($t$), width ($W$), and height ($H$) of the diamond waveguide were swept. The optimal coupling is found at $W = 280$ nm, $H = 240$ nm, and $t = 8 \mu$m, although the coupling is insensitive to the taper length. This design yields an overall dipole-to-WG coupling efficiency of 50% (25% coupled to each side)
as seen in Fig. 5-2(c).

The second device is similar to the first, though the diamond waveguide is placed over a gap in the SiN waveguide, and the ends of both the SiN and diamond waveguides are tapered as seen in Fig.5-2(b). Again, this promotes adiabatic mode coupling from the single-mode diamond waveguide to the single-mode SiN waveguide. This allows for better coupling of the dipole to the waveguide mode, so more of the light can be captured into the diamond waveguide mode initially. It also reduces the coupling from the laser to the waveguide in our top-down excitation scheme used in the experimental implementation below. To optimize the coupling, we sweep the diamond and SiN taper lengths, as well as the width and height of the diamond waveguide while keeping the gap length at 2 μm. The tapers are aligned such that the taper regions in SiN and diamond do not overlap, minimizing the photon loss due to scattering. \( W = H = 200 \text{ nm} \) is the optimal diamond waveguide dimension for coupling into the \( 400 \times 400 \text{ nm} \) SiN waveguide. Based on the sweep results, we chose a diamond μWG taper length of 6 μm and a SiN WG taper length of 5 μm. This gives a diamond μWG which is 24 μm long, and yields an overall dipole-to-WG coupling efficiency of 80% (40% coupled to each side) as seen in Fig. 5-2(d).

![Figure 5-3](image)

Figure 5-3: (a) Schematic of a mode coupling device with a tapered diamond waveguide placed next to a SiN waveguide. (b) Schematic of a mode coupling device with a tapered diamond waveguide placed next to a gap in a SiN waveguide (also with tapered ends. (c) Simulation of the coupling efficiency and loss channels for the device shown in (a). (d) Simulation of the coupling efficiency and loss channels for the device shown in (b).
The third and fourth devices are based on a geometry where the diamond waveguide is placed on the SiO₂, directly abutting the SiN waveguide. In analogy to the first two devices, we simulate structures with and without a gap as seen in Fig. 5-3(a,b). Again, we optimize the width and height of the diamond waveguide, as well as the length of all tapers. Without a gap (Fig. 5-3(c)), the maximum dipole-to-WG coupling efficiency is 58% (29% to each side). With a gap (Fig. 5-3(d)) this increases to 68% (34% to each side). Similar to the first two designs, this is due to less coupling between the NV and initial waveguide mode without a gap, as the NV is not at the mode maximum of the hybrid SiN-diamond mode.

For the initial demonstration of coupling, we use the second design which provides the highest collection efficiency, although placing the diamond μWG next to the SiN waveguide is considerably easier and should be considered in the future.

Diamond Integration

_Luozhou Li fabricated the diamond waveguides discussed in this section._

The diamond μWGs are detached and picked up from the initial array with a tungsten microprobe (Ted Pella) mounted to a 3-axis piezo micro manipulator. The sample is placed on a 2-axis X-Y stage and rotation stage to align the tip perpendicular to the diamond μWGs. Fig. 5-4 shows a 12 μm long diamond μWG held by a tungsten probe with a 500 nm radius tip. Adhesion between the diamond and the tip is promoted by a solvent clean of the tungsten tip immediately before picking up the μWG. Each μWG is transferred to the SiN chip, which sits on a 2-axis X-Y stage and a rotation stage. The diamond μWG is aligned to the coupling region of a SiN waveguide and placed in its center, as shown in Fig. 5-4(b). Adhesion between the SiN and diamond waveguides is promoted via O₂ plasma cleaning of the SiN prior to placement (100 W, 5 minutes). We found that this O₂ plasma clean step was integral to the successful placement of a diamond node. It is also time sensitive and the placement must be done within a few hours of the plasma treatment. Fig. 5-4(c) shows the complete placement of the diamond μWG over a gap in the SiN waveguide.

Experimental Results

The scanning electron microgram (SEM) in Fig. 5-4(d) shows a typical array of diamond μWGs. In this experiment, we used 12 μm-long, 200 nm wide μWGs with 4 μm-long tapers down to 100 nm on either side fabricated from a diamond membrane. This minimal taper
Figure 5-4: (a) A diamond μWG held by a tungsten tip. (b) Aligning the diamond μWG to the SiN waveguide. (c) The final integrated node. (d) An SEM of the initial array of diamond μWGs. (e) A scanning confocal image of the array of diamond μWGs in (d).

<table>
<thead>
<tr>
<th>WG number</th>
<th>CPS in the WG</th>
<th>CPS in the objective</th>
</tr>
</thead>
<tbody>
<tr>
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<td>$3.8 \times 10^5$</td>
<td>$1.8 \times 10^5$</td>
</tr>
<tr>
<td>2</td>
<td>$2.5 \times 10^5$</td>
<td>$1.7 \times 10^5$</td>
</tr>
<tr>
<td>3</td>
<td>$2.4 \times 10^5$</td>
<td>$4.4 \times 10^5$</td>
</tr>
<tr>
<td>4</td>
<td>$1 \times 10^5$</td>
<td>$2.6 \times 10^5$</td>
</tr>
</tbody>
</table>

Table 5.1: Overview of coupling into the waveguide and into the objective from four diamond nodes placed on the SiN PIC at 100μW excitation.

size is larger than optimal, but the fabrication yield was significantly increased compared to thinner ends. This is no longer a constraint with the quasi-isotropic etch (Sec. 3.4). FDTD simulations indicate that this non-ideal geometry should yield a 52.5% coupling efficiency from the NV ZPL to the SiN waveguide. We used photoluminescence (PL) confocal raster scans (NA = 0.9, 532 nm excitation) to identify and characterize four μWGs with single NV centers near the center of the μWG. As seen in the confocal scan in Fig. 5-4(e), this is a rare occurrence in this randomly implanted sample. The pre-selected diamond μWGs are picked and transferred onto four consecutive coupling regions of WGs in the SiN PIC. Fig. 5-4(c) shows an optical image of a representative placement.

We evaluated the coupling efficiency into the SiN PIC from each μWG by confocally exciting the single NV through the objective and collecting the fluorescence both through the objective and through the SiN waveguide into a single mode fiber. We used a lensed single mode fiber with a focal spot of 1 μm (and working distance of 4 μm), which approximately matches the expected spot size of the tapered SiN waveguide at the edge of the chip. The fiber is held on a 3-axis piezo stage for fine positioning. We optimize coupling from the waveguide to the fiber by back-coupling a 632 nm HeNe laser through the fiber into the
waveguide and maximizing transmission. The pump is filtered from the waveguide collection port by coupling into free space, filtering with a notch filter, and coupling back into a fiber for routing to the APD.

Table 5.1 summarizes the photon count rates obtained into the objective and into the SiN waveguide for a 532 nm pump power of 100 μW. The total count rate is separated into NV and background due to PL from the SiN waveguide by performing autocorrelation measurements. The emission rate into the waveguide is consistently high with variation due to differences in the μWG placement, fabrication imperfections, and the position and dipole orientation of the NV centers.

![Images of PL raster scans and auto-correlation measurements](image)

Figure 5-5: (a) High resolution confocal PL raster scan of the NV with confocal collection and (b) waveguide collection. (c) normalized autocorrelation measurement $g^{(2)}(\tau)$ via confocal collection confirming single photon emission. (d) cross-correlation measurement between photons collected via the confocal setup and through the waveguide confirming that the majority of photons collected through the waveguide originate from a single NV. Solid curves in (c,d) are fits to the data. (e) Spectrum of the emitter collected via the waveguide. The solid curve is a model with parameter values taken from measured data as detailed in the Appendix. (f) Saturation measurements acquired on the same emitter with confocal (dashed) and waveguide (solid) collection. (g) fits from (f) without background and corrected for measured collection losses (see text).

In the following, we focus on integrated system 1 in Table 5.1. Fig. 5-5(a,b) show PL raster scans of the NV under confocal excitation with confocal and waveguide collection, respectively. For Fig. 5-5(b), the background was subtracted using spin-selective fluorescence [125, 126]. Fig. 5-5(c) shows the normalized auto-correlation measurement of the NV
in Fig. 5-5(a,b) with $g^{(2)}(0) = 0.17$ at 100 µW of 532 nm excitation. We also performed cross-correlation measurements between photons collected via the confocal and waveguide setups. The normalized cross-correlation measurement in Fig. 5-5(d) indicates clear anti-bunching at 60 µW of excitation. The increase in $g^{(2)}(0)$ relative to the confocal autocorrelation measurement is attributed to the generation of background fluorescence due to coupling of the excitation laser into the SiN WG.

Fig. 5-5(e) plots the spectrum of the fluorescence collected through the waveguide including both NV fluorescence and the unwanted PL caused by laser propagation through the SiN waveguide. The PL originating from the diamond µWG experiences an etalon effect due to the diamond µWG facets. Over the wavelength region of interest (600-780 nm), the dispersion of the waveguide mode is linear. The group velocity dispersion in the 12 µm diamond waveguide is negligible as seen in Fig. 5-5(f) and does not affect the elatoning effect. Thus, the detected spectrum is modeled simply as

$$WG(\lambda_0) = a \left[ \frac{(1 - r^2)}{1 - r^2 e^{-4i\pi nL/\lambda_0}} \right]^2 NV(\lambda_0) + bBG(\lambda_0)$$

where $NV(\lambda_0)$ is the spectrum collected via the confocal setup and $BG(\lambda_0)$ is the spectrum of the SiN fluorescence collected through the waveguide, both of which are plotted in Fig. 5-5. $n = 1.65$ is the expected effective refractive index of the diamond waveguide mode determined via eigenmode analysis of the waveguide, $L = 12$ µm is the total length of the diamond waveguide, and $r^2 = 0.17$ due to the waveguide-air interface.

To evaluate the enhancement in collection efficiency through the waveguide, we performed emitter saturation measurements with both confocal and waveguide collection as seen in Fig. 5-5(f). In each case, the excitation polarization was tuned to maximize the signal-to-noise ratio. For confocal collection, this entailed maximizing the NV excitation rate, while under waveguide collection this entailed minimizing the coupling of the 532 nm excitation into the waveguide to minimize background fluorescence. The optimized polarization for waveguide collection reduces the NV excitation rate, and as such increases the saturation intensity from 135 µW via confocal collection to 350 µW via waveguide collection. For confocal (waveguide) collection, 16 kcps (55 kcps) were detected at 60 µW of excitation (with polarization optimized for waveguide signal to noise ratio), as used to measure the cross-correlation function seen in Fig. 5-5(d).
Figure 5-5(g) shows the fits in Figure 5-5(f) without the linear background terms, and corrected for the measured collection efficiencies of each collection pathway. The efficiency of the waveguide collection pathway was estimated to be \( \sim 25\% \) with a transmission measurement using a 635 nm HeNe laser. The confocal setup was measured to have an upper-bound efficiency of 17% from the sample to the fiber-coupled detector, again from transmission of the 635 nm laser. Both signals are measured with a Si avalanche photodiode (APD) with quantum efficiency \( \eta = 0.65 \). Without these system inefficiencies, we estimate that \( 0.38 \times 10^6 \) NV photons/second are collected into the objective at saturation, while \( 1.45 \times 10^6 \) NV photons/second are collected into one direction of the single mode SiN waveguide. This is more than 3.5 times that collected into the objective, demonstrating the efficiency of this hybrid approach even with a simple waveguide and imperfect fabrication, NV placement, and \( \mu \)WG placement. This coupling efficiency could surely be improved in future iterations.

The creation of an efficient network of entangled quantum nodes depends not only on the efficient optical coupling demonstrated above, but also on the spectral and spin properties of the quantum nodes. The pre-characterization demonstrated here is easily scaled to include these, and other relevant properties.

![Figure 5-6](image.png)

Figure 5-6: (a) ODMR of an NV under no magnetic field with waveguide collection. (b) The \( m_s = 0 \) to 1 transition is driven off resonance and 3 Ramsey frequencies are observed due to coupling between the NV electronic spin and the host N\(^{14}\) nuclear spin, with a decay due to the surrounding spin bath (\( T_2^* = 2 \mu s \)). (c) The \( \pi \) and \( \pi/2 \) times of an on-resonance driving field are used to construct a Hahn-Echo sequence to decouple the NV from the surrounding spin bath and measure \( T_2 > 120 \mu s \) from the exponential decay of the coherent revivals.

In Figure 5-6 we present the electron spin properties of the NV center in a second integrated system (WG number 2 in Table 5.1(f)) in which \( 0.8 \times 10^6 \) photons/second were collected into one direction of the waveguide at saturation. Refer to Ch. 2 for details about the NV center’s magnetic sublevels. Figure 5-6(a) plots the ODMR fluorescence signal.
collected through the waveguide under continuous-wave laser excitation with no external magnetic field.

For state manipulation, the degeneracy of the $m_s = \pm 1$ levels is lifted by the application of an magnetic field of $\sim 56$ Gauss projected onto the NV axis. A Ramsey sequence, consisting of two $\pi/2$ pulses with a MW frequency, slightly detuned from the $m_s = 0$ to $m_s = 1$ transition frequency, separated by a free evolution time $\tau$, was used to probe the spin environment experienced by the NV. From this, we deduce an ensemble phase coherence time $T_2^* \approx 2 \mu s$ as seen in Figure 5-6(b). We also performed Hahn-Echo decoupling [127] to filter low frequency noise components arising from the spin bath. These measurements indicate a spin coherence time of $T_2 \approx 120 \mu s$, see Figure 5-6(c). This long spin coherence time is similar to values observed in the parent diamond crystal. We anticipate that using isotropically purified $^{12}$C diamond together with dynamical decoupling should enable spin coherence times in excess of tens of milliseconds [128].

![Figure 5-7: (a) Normalized spectra of the NV via confocal collection (black) and the SiN waveguide fluorescence (gray) under 532-nm excitation. (b) Effective index of the diamond waveguide mode. (c) Laser attenuation from DBR filters integrated into similar single-mode waveguides measured in transmission.](image)

The initial demonstration of a hybrid quantum PIC illustrates the efficacy of this approach. The node yield was 100%, even with an initial low yield of suitable diamond nodes. Also, the simple taper geometry was shown to give consistently high collection efficiency into the PIC mode. However, the saturation and cross-correlation measurements make it clear that the background PL caused by the SiN would limit the entanglement fidelity in this architecture. Fig. 5-7(a) shows a comparison of the background PL measured through SiN waveguide and a typical NV room temperature spectrum. To reduce SiN fluorescence in future iterations, we designed, fabricated, and tested DBRs integrated into single-mode
waveguides to stop propagation of the 532nm excitation laser through the waveguide. The DBRs were designed with periodic etched and full regions to give coherent reflection. The full regions are terminated with a semi-circle in simulation to take into account e-beam resolution limits. The widths and lengths of the two regions were optimized to maximize the ratio of reflection of excitation light and transmission of 637nm ZPL signal. FDTD simulations of 60 periods of the optimized structure gives greater than 40 dB attenuation for elliptically polarized light at the excitation wavelength with a FWHM of ~30 nm, and over 95% ZPL transmission. The first fabricated iteration of these structures results provided greater than 14dB of attenuation at 515 nm as seen in Fig. 5-7(b) excitation, a common direct-bandgap laser wavelength, which can be used to efficiently excite the NV [129, 130].

5.3.2 Alternative Platforms

As demonstrated in Sec. 5.3.1, SiN provides a low-loss platform for QIP with NV centers. However, while mature fabrication techniques enable low-loss passive components, the background fluorescence due to the amorphous nature of SiN and the lack of active capabilities limit its use as a scalable platform for QIP. Therefore, we also investigate other material platforms.

A suitable material for large scale on-chip QIP will have:

- (i) a large band gap (≥ 2eV) to ensure low absorption of propagation ZPL photons;
- (ii) low autofluorescence to maximize signal-to-noise ratio;
- (iii) active capabilities for tunable phase shifters and filters;
- (iv) mature fabrication techniques for low-loss passive and active components.

A list of the suitable materials that we investigated is found in Table 5.2. There are of course other materials that allow propagation of visible light that may arise as contenders for QIP in the visible spectral range in the future.

**Su8**

*Igal Bayn fabricated the Su8 waveguides measured in this section.*

SU-8 is an epoxy-based polymer that is commonly used as a negative photoresist and in the production of bio-MEMS structures. It has an index of refraction of \( n = 1.59 \) and is
Table 5.2: Overview of materials suitable for a visible PIC for QIP. ¹The exact bandgap of SiN can vary due to deposition parameters and Si content [1]. ²The Kerr nonlinearity can be increased in Si-rich SiN. ³Broadband PL seen in low-quality films.

<table>
<thead>
<tr>
<th>Material</th>
<th>Band Gap</th>
<th>PL</th>
<th>Active</th>
<th>Fabrication</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiN</td>
<td>4.5 eV</td>
<td>Broadband</td>
<td>None ²</td>
<td>Yes</td>
</tr>
<tr>
<td>Su8</td>
<td>3.4 eV</td>
<td>Low</td>
<td>None ²</td>
<td>Yes</td>
</tr>
<tr>
<td>GaP</td>
<td>2.25 eV</td>
<td>Low³</td>
<td>r₃₃ = 1 pm/V</td>
<td>No</td>
</tr>
<tr>
<td>LiNb</td>
<td>4.9 eV</td>
<td>Low</td>
<td>r₃₃ = 32.2 pm/V</td>
<td>No</td>
</tr>
<tr>
<td>AlN</td>
<td>6 eV</td>
<td>Low</td>
<td>r₃₃ = 0.6 pm/V</td>
<td>Yes</td>
</tr>
<tr>
<td>GaN</td>
<td>3.4 eV</td>
<td>Low</td>
<td>r₃₃ = 1.9 pm/V</td>
<td>Yes</td>
</tr>
</tbody>
</table>

often used to efficiency couple from a waveguide mode with higher effective refractive index to a fiber mode with comparable effective refractive index [141]. As it is a negative resist, high-aspect ratio structures are easily fabricated, and low loss propagation in the visible spectrum has been demonstrated [142]. Su8-on-SOI samples fabricated in-house showed low autofluorescence, and waveguides were demonstrated to have a loss of 3 dB/mm, but bends showed high loss. The loss could be reduced by baking the sample after fabrication to smooth the sidewalls.

Unfortunately, the difference in mode size between the Su8 and diamond waveguide modes made it difficult to efficiently couple between modes. Fig. 5-8(a,b) show a simulation of the fundamental TE modes of the Su8 and diamond single mode waveguides used in experiment. As in the SiN case, we model the coupling efficiency from an NV in a diamond waveguide with tapered ends to a final Su8 waveguide mode, both with and without a gap in the Su8 waveguide (see Sec. 5.3.1 for details about the structures and simulations). Fig. 5-8(c,d) show the coupling efficiency from the NV to the Su8 waveguide with and without a gap respectively. Unfortunately, the maximum coupling efficiency is only 48% (24% in each direction) and most of the light is lost to the substrate (+Z direction).

Gallium Phosphide

*Igal Bayn fabricated the GaP waveguides measured in this section*
Gallium Phosphide (GaP) has a bandgap of 2.25 eV allowing for low-loss propagation of visible light, and has been used in hybrid GaP-diamond devices for Purcell enhancement [143] and on-chip routing of NV fluorescence [144, 145]. Unfortunately, high quality GaP can only be grown on AlGaP sacrificial layers which does not allow for the fabrication of single-mode low-loss propagation of visible light. Thus, it must be undercut and used suspended or transferred onto a different substrate, and it is difficult to create robust, stable PICs. We were unable to measure the transmission of suspended, air-clad GaP waveguides fabricated in-house due to low yield of long waveguides. Moreover, background fluorescence due to bad film quality and excessive processing made it unusable for single photon measurements. Recently, optical structures have been demonstrated in a GaP-on-SiO$_2$ platform [146], perhaps improving the prospects for the fabrication of large-scale photonic integrated circuits in GaP.
Aluminum Nitride

Aluminum Nitride (AlN) is the leading choice for large scale active photonic integrated circuits for operation in the visible spectrum due to the large bandgap. Low loss propagation, nonlinear optical devices, and MEMs components have all been demonstrated in an AlN platform [137, 147]. Recent work in our lab has shown the fabrication of record low loss waveguides in visible and even the UV range, making an AlN platform applicable to QIP with trapped ions as well [148].

Lithium Niobate

Lithium Niobite (LiNb) is an attractive choice for quantum information in a PIC architecture. As seen in Table 5.2, LiNb has electro-optic and piezo-electric coefficients more than an order of magnitude larger than any of the other materials considered, and has a long history of use for active optical components in bulk optics. Moreover, wafers of LiNb on SiO₂ are available commercially for single-mode waveguide fabrication. Unfortunately, LiNb is difficult to etch, and all demonstrated etches so far are purely physical causing large surface roughness, mask degradation, and redeposition. Moreover, the thermoelectric effect causes high $E$ fields when heating the substrate which complicates many processing steps, for example with depositing SiO₂ for cladding. Until recently, this has prohibited the fabrication of low-loss waveguides in LiNb. Recently, however this has been overcome and extremely low-loss waveguide propagation has been demonstrated [149, 150]. As it becomes possible to fabricate high-quality integrated components, LiNb is emerging as the most promising candidate for a full PIC architecture for QIP in the visible spectrum.

5.3.3 Hybrid Devices

Portions of this section have appeared in Reference [151]

The results reported thus far have focused on passive devices, with no added functionality coming from the hybrid nature of the PIC. However, we can also exploit the hybrid nature of the PIC to make fully hybrid nanophotonic structures which add further functionality.

Photonic nanocavities in diamond have emerged as useful structures for interfacing photons and embedded atomic color centers, such as the NV center as described in Chap. 4. However, there are currently two main challenges limiting progress in scaling to multiple
coupled emitter-cavity systems for distributed quantum networks. To improve the entanglement rate, the cavity design must not only enhance the emission of the desired transition via a high $Q$ mode at the transition frequency, but more importantly increase the overall collection rate of that transition. Thus the loss pathways of the cavity must be engineered to funnel the enhanced light into a single useable mode, such as a waveguide mode for further routing and manipulation. Secondly, fabrication inconsistencies across a single chip cause a spread in the resonant frequencies of the final devices that can severely decrease the enhancement of the desired transition. Thus, post-fabrication tuning at each individual node is necessary.

Figure 5-9: (a) Structure of the hybrid cavity. Two suspended AlN beams (gray) periodically patterned with holes sit on either side of a width-modulated diamond (blue). (b) Cross section through $x = 0$ plane of the cavity. (c) Unloaded cavity mode profile ($|E|^2$).

Here, we present a hybrid cavity design that addresses both of these challenges: 75% of the mode at each cavity is coupled into a single waveguide mode of the underlying photonic integrated circuit (PIC) that connects all nodes, and the frequency is tunable over 10 linewidths while maintaining a $Q$ factor within 50% of the maximum value. AlN is chosen to create the optical band-gap at the cavity region – and as the backbone PIC material – as it has a wide direct band-gap ($\sim 6.1$ eV at 4 K) that allows low-loss single-mode operation at the NV ZPL wavelength (637 nm). Moreover, its favorable piezoelectric properties have made it a widely used material for free-standing nanoelectromechanical (NEMS) devices [152, 153], and electro-optic and frequency conversion devices [154, 147] have been demonstrated, making it a promising platform for reconfigurable quantum circuits that can

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interface with fibers for long-distance quantum communication. This active ability also enables the NEMS needed to tune the cavity frequency, as discussed below.

The AlN backbone consists of single mode waveguides with all beam splitters and active components needed for routing the optical signal between cavity nodes. As indicated in the schematic of Figure 5-9(a) and the cross section in Figure 5-9(b), the substrate at the cavity region is etched away to leave the AlN suspended in air. A diamond waveguide containing a single, well-characterized NV center is placed into the cavity region. The diamond waveguide is suspended at the cavity region, with both ends resting on the substrate and locked into AlN alignment features.

As shown in Figure 5-9a, the cavity region consists of two suspended parallel AlN waveguides with a height \( H/\lambda_{NV} = 0.345 \) and width \( W/\lambda_{NV} = 0.383 \) where \( \lambda_{NV} = 637 \text{ nm} \) is the free-space ZPL wavelength. These are patterned with periodic holes with constant spacing \( (a/\lambda_{NV} = 0.356) \) and constant radius \( (r/a = 0.295) \). A diamond slab with the same height as the AlN layer containing a single NV center at the center of the beam is centered between the two AlN beams \( (D_1 = D_2 = 0.235\lambda_{NV}) \). The width of the suspended diamond membrane increases parabolically from \( W_1/\lambda_{NV} = 0.157 \) to \( W_2/\lambda_{NV} = 0.314 \) over 7 periods. The periodic patterning of the two AlN waveguides creates a bandgap in the \( x \) direction. The increased width of the diamond membrane at the center of the structure increases the
effective refractive index in a localized spot in the otherwise periodic structure. Moreover, the diamond refractive index \(n_{\text{dmd}} = 2.42\) is higher than that of AlN \(n_{\text{AIN}} = 2.10\), and so the mode resides in the diamond as seen in the energy distribution \(|E|^2\) shown in Figure 5-9c. The gentle parabolic width increase reduces scattering \([105]\), and finite difference time domain (FDTD) simulations show a radiation-limited \(Q\) factor of \(> 1\) million with 50 holes on either side of the center of the cavity.

FDTD simulations reveal a mode volume of \(V \approx 10(\lambda_{\text{NV}}/n_{\text{dmd}})^3\). Due to the large transverse extent of the hybrid structure, this mode volume is larger than other nanobeam cavity designs, which provide mode volumes on the order of \((\lambda/n)^3\). However, the high \(Q\) ensures a large Purcell enhancement for an optimally placed and oriented NV center. The mode maximum is at the center of the structure, or 110 nm in depth and 100 nm from each side of the diamond. The mode is within 90\% of the maximum within ±20 nm, ±30 nm and ±60 nm for the \(x\), \(y\), and \(z\) directions respectively.

The central positioning of the NV center (≥ 100 nm from each surface) is also beneficial for the optical properties of the NV. Although the NV exhibits long-lived electronic spin states, they are especially sensitive to stray electric fields, and the nanofabrication of PhC cavities can introduce defects in the crystal and on the surface due to the ion bombardment that is necessary to etch the host material. These defects trap charges in unstable configurations that lead to pure dephasing and spectral diffusion that in turn leads to a decrease in entanglement rate due to reduced indistinguishability between photons emitted by distinct emitters. The spectral diffusion of solid state emitters increases with proximity to fabricated surfaces \([111, 108]\), and previous work in bulk diamond has demonstrated lifetime limited linewidths of NV centers 100 nm from the surface \([14]\), suggesting that this design could provide the same with adequate surface treatment. In this design, the diamond is only minimally patterned, and instead the periodic change in the dielectric environment needed to produce a band gap is provided by the patterning in the AlN.

To efficiently collect the NV emission into the backbone PIC, the cavity must be coupled to the waveguide. The unloaded design described above has a high \(Q\), but the emitted light cannot be efficiently collected into the waveguide as it radiates mainly into the \(z\)-plane. Thus, despite the large Purcell enhancement due to the high \(Q/V\), there is only a marginal increase of light collected into a single AlN waveguide mode over a simpler, broadband architecture of an NV coupled to a single diamond waveguide mode that is adiabatically tapered to transfer
the optical mode to the underlying PIC mode [108]. To increase the collection enhancement, the cavity is loaded into a single AlN waveguide mode by reducing the number of holes on one of the four sides of the cavity, as seen in Fig. 5-10(a). In general, the fraction of emission into the waveguide can be estimated from the ratio $F = Q_{i}/(Q_{i} + Q_{l})$ where $Q_{i}$ is the intrinsic $Q$ factor, and $Q_{l}$ is the loaded $Q$ factor. However, modifying the cavity geometry can also increase scattering into other loss pathways. Moreover, the figure of merit of interest is the overall collection enhancement, which is a function of $Q$ and $F$. Therefore, we performed detailed FDTD simulations to measure the collection enhancement into the waveguide port. As seen in Fig. 5-10(a), the collection enhancement is maximized when the number of holes leading to the output waveguide is $N_{out} = 16$. At this configuration, the loaded $Q$ is 55,000, with 75% of the light coupled into the waveguide as calculated with FDTD simulations. The collection spectra into the waveguide, as well as into the $y$ and $z$ planes is shown in Fig. 5-10(b). Fig. 5-10(c) shows the mode profile ($\log(|E|^2)$) of the loaded cavity, with clear coupling into the bottom right waveguide.

For high fidelity entanglement between two NV centers, they both must emit into the same lifetime-limited frequency mode. The ZPL transition frequency of 2 or more NV centers can be tuned to overlap via the Stark effect [21]. However, the cavities’ resonances must also be at the same frequencies. Unfortunately, fabrication imperfections across a single chip can cause inhomogeneities in the resonant frequencies. Therefore, it is necessary to tune the frequency of the cavity post-fabrication without a significant drop in the quality factor of the cavity. Previous works have demonstrated cavity tuning in other material systems. For instance, the fast and reversible tuning of PhC cavities in semiconductors such as Si and
GaAs has been demonstrated via injection of free carriers, either electrically \cite{155} or via two photon absorption \cite{156}. However, PN junctions are notoriously difficult to make in diamond and doping the diamond may have deleterious effects on the NV center. Previous cavity results in diamond \cite{72, 157} have used gas adsorption and sublimation to tune a cavity’s resonant frequency, but this approach affects all cavities on the chip simultaneously and cannot be easily used to stabilize the resonant frequencies of many cavities on the same chip over a long period of time.

Works in other material systems employed nano-electrical-mechanical systems (NEMS) to couple two cavity modes \cite{158, 159} or modify the evanescent field of the cavity mode to induce wavelength shifts \cite{160, 161, 162}. This is not currently feasible in an all-diamond system as large-scale free-standing membranes are not widely available, and moving the diamond would cause strain which would shift the NV center’s optical transitions \cite{23}. In the hybrid design presented in this paper, the AlN layer can provide the needed NEMS functionality using electrostatic or piezoelectric \cite{152, 153} actuation. FDTD simulations show that displacing the two patterned AlN beams changes the cavity’s effective refractive index, and thus the resonant frequency. The tuning simulations were performed on the loaded cavity. Moving the waveguide-coupled AlN beam a certain distance \((D_1)\) provides different tuning than moving the non-coupled beam the same distance \((D_2 = D_1)\). Figure 5-11(a) summarizes the results showing the effect of beam displacements on resonant frequency and \(Q\), plotted against \((D_2 - D_1)\). Figure 5-11(b) demonstrates that the cavity can be tuned over 10 linewidths while maintaining \(Q \geq 0.5Q_{max}\). While this device’s tuning range is small due to the high \(Q\), it allows the individual tuning of multiple cavities on the same chip with no crosstalk. If necessary, coarse tuning of cavities can be accomplished by a variety of other techniques such as photochromic \cite{163} or chalcogenide index-changing \cite{164} materials. The cavity fine-tuning, as proposed here using NEMS actuation is required to tune and maintain many high-\(Q\) on-chip cavities on resonance for the realization of on-chip quantum networks and entanglement distribution in envisioned quantum repeater architectures \cite{165}.

Finally, this hybrid cavity design is compatible with the scalable hybrid assembly of a photonic backbone populated with pre-selected quantum nodes \cite{166, 108, 167, 168}. The proposed approach requires careful alignment between the diamond and AlN components. Vertical alignment \((z)\) is assured by the diamond membrane’s physical contact with the plane of the substrate. \(x\) alignment must be within \(\pm 30\ nm\) to maintain an enhancement \(\geq 90\%\)
of the maximum. Various pick-and-placement techniques have already been demonstrated with excellent in-plane alignment, including optical microscopy with better than 200 nm in-plane tolerance [108, 169], and scanning-electron microscope based alignment with 25 nm tolerance [170, 171, 172, 173], aided by lithographically defined alignment structures as indicated in Figure 5-9(a). Finally, any remaining error is correctable in-situ using AlN NEMS actuators in \( x \) and \( y \) directions.

In conclusion, we introduced a novel hybrid cavity design that enhances the collection of photons from a transition of an embedded quantum emitter that is at the resonant frequency of the cavity. This is achieved by loading a high-\( Q \) cavity directly into a single mode PIC network. Moreover, the emitter remains \( \geq 100 \) nm away from every surface, reducing spectral diffusion due to trapped charges on the surface. Finally, NEMS actuation permits the reversible and stable tuning of the resonant frequency. The implementation of this design across a multi-node PIC will enable efficient multi-qubit entanglement across the PIC.

5.4 Outlook

The work presented in this chapter focused on the initial demonstration of quantum memories integrated with on-chip photonic routing, showing it will be possible to build up a hybrid architecture where the benefits of each material can be fully utilized. However, to gain the full advantages of processing quantum information on-chip it will be necessary to integrate more functionality on-chip, and to successful integrate many more quantum nodes on one chip.

Moreover, the entanglement protocol used to mediate entanglement between spatially separated NV centers maintains a high fidelity even in the presence of phase instability and loss, though at the expense of entanglement rate. However, PIC architectures provide a low-loss platform with high phase and polarization stability, and nanophotonic structures will increase the coupling between single NV centers and the desired optical mode. Thus, it is necessary to reconsider the choice of entanglement protocol, perhaps valuing high rate entanglement over unity fidelity to create the large cluster states needed for computation.
Chapter 6

Conclusion and Outlook

Quantum information science is at a turning point. Theoretical results and initial experimental demonstrations have spurred governmental and private funding, with the understanding that the successful development of quantum computation could change the world as classical computation did in the last half century. However, we are far away from the scale needed to demonstrate quantum supremacy in communication or computation. Quantum computers and quantum repeaters both require multiple entangled quantum memories that can be individually controlled and measured. However, building and maintaining these states is challenging due to the many decoherence pathways any experimental system encompasses. While there has been rapid experimental progress in developing entangled networks using stationary qubits connected via photons, the network size is limited to two due to low entanglement rates and bulk infrastructure.

The research presented in this thesis aims to increase the maximum quantum network size by increasing the entanglement rates and introducing a scalable architecture for the on-chip creation, control, and measurement of entangled states. While, it was not possible in this thesis, the work presented here should lead to a demonstration of high-rate, high-fidelity entanglement on-chip.

6.1 Increased Entanglement Rates

This thesis presents the design, fabrication, and measurement of high quality nanophotonic devices [87, 151, 89, 108, 174, 175, 85, 93] to increase the optical density of states at the ZPL frequency of the NV center and to tailor the loss channels for collection into the
desired optical mode. Cavities fabricated directly from bulk diamond show record high quality factors [89], and a hybrid design allows for the wavelength tuning over ten times the linewidth for matching frequencies for all nodes [151].

Unfortunately, the optical coherence of NV centers in these structures were degraded due to proximity to the diamond surface. To increase the entanglement rate without sacrificing fidelity, it will be necessary to improve the optical coherence of NV centers in nanostructures. The first step in reducing spectral diffusion will necessarily be large-scale characterization of the optical properties of NV centers – both in bulk diamond and in nanostructures – to understand the dynamics of spectral diffusion and charge stability. Once the dynamics are well understood, it should be possible to control and stabilize the charge state and optical coherence of NV centers near surfaces via improved diamond preparation, surface chemistry, or electrical control.

The spin properties of NV centers make them an obvious choice for a quantum memory. However, the optical properties must be improved. To this end, there has also been a lot of research into other defects in diamond (and other solid state hosts). For instance the Silicon and Germanium di-vacancy centers have no permanent dipole and thus do not suffer from spectral diffusion to the first order [86, 176], and their Debye-Waller factors are much higher. Unfortunately, their ground state consists of spin-orbital states that allow phonon-mediated spin mixing and require dilution fridge temperatures to achieve reasonable spin coherence times [177, 178]. Going forward it will be important to continue investigating these and other defects in the hopes of engineering a quantum system that provides both good optical and spin coherence.

6.2 Scalable Architectures

This thesis develops and demonstrates a hybrid architecture [108] in which pre-characterized diamond nodes are placed into a photonic integrated circuit. This decouples the low-yield process of fabricating high quality quantum memories from the final result, leading to a high yield final system where every node is pre-selected. However, for a demonstration of on-chip entanglement, it will be necessary to design, fabricate, and test cryo-compatible passive and active electrical and optical components for control and signal routing, filtering, and detection.

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Fig. 6-1 shows a schematic of an on-chip QIP platform. While this thesis focuses on NV centers, most of the requirements for an on-chip QIP architecture are agnostic with respect to the actual quantum memory, and scaling up other quantum systems such as solid state defects, trapped ions, and trapped neutral atoms will also require optical and MW control, optical detection, and on-chip classical logic. The need for microwave systems and on-chip logic also apply to non-optical qubits such as superconducting qubits and charged quantum dots in semiconductors.

Laser excitation is necessary for optically active qubits. When comparing sources, wavelength, linewidth, modulation depth and speed, power, and wavelength tunability should all be considered. On-chip lasers cannot currently provide the linewidth, power, and tunability necessary, although the field is constantly improving [179]. Vertical-cavity surface-emitting lasers coupled to waveguides with gratings [180] could also be used. In the near-term, it will be easiest to use conventional laser systems coupled on-chip to take advantage of the power, linewidth, and wavelength tunability available in commercial lasers. As all operations will necessarily be performed on a clock we can also use commercial bulk acoustic-optical modulators for fast and high-isolation modulation to create pulse streams that are sent to all
nodes, and use MEMS-based switches at each node which are slow but provide high optical isolation [181].

Microwave control of spin states is necessary for solid-state and ion systems, and DC electric fields are necessary for trapping ions and for Stark tuning of the optical transitions of solid-state defects. In the near term, these AC and DC electric fields can be generated off-chip and delivered to nodes with on-chip striplines. However, in the long-term it will be necessary to leverage the multi-billion dollar industry of complementary metal-oxide-semiconductor (CMOS) systems which are routinely used to create high bandwidth microwave fields.

To build the large entangled cluster states necessary for quantum computation, or even to mediate entanglement between pairs of NV centers in a quantum repeater node, it will be necessary to route the optical channels to interfere the necessary modes. This has been demonstrated for LOQC in Si and SiO$_2$ waveguide platforms with a large tuneable Mach-Zender Interferometer (MZI) array which allows for coupling between nodes. The development of these systems for visible light will have to take into account loss and device footprint. This is a mature field in Si waveguides for C and L band propagation, but has only recently been considered in platforms compatible with low-loss visible propagation.

To create entanglement between nodes it is also necessary to have high efficiency photon detection for each mode. Si APDs have been demonstrated in a CMOS process [182], with specifications rivaling commercial systems. On the other hand, superconducting nanowire single photon detectors (SNSPDs) can have significantly higher quantum efficiency and lower jitter [183]. These can be directly integrated with waveguides [169], or in the near-term the light can be coupled off-chip with fiber and then routed to detectors. Finally, any quantum system will need a classical logic layer for maintaining a clock shared across all nodes, and for fast feedback from measurement results to PIC reconfiguration. Again, logical layers are routinely implemented in CMOS chips.

6.3 Outlook

In conclusion, the research presented in this thesis represent a step towards the implementation of large-scale quantum systems. With improved optical coherence of NV centers in nanophotonic devices, it could be possible to demonstrate entanglement rates order of mag-
nitude faster than the decoherence rate, and the integration of low-cross talk optical and microwave control at each node, active photonic routing, and on-chip classical logic could enable on-chip entanglement. Together, these advances could enable the on-chip generation of cluster states for quantum computing [184], and on-chip quantum repeater nodes for long distance quantum communication [185].
Bibliography


