Integrated Optical Components for Quantum Key Distribution

Yasir Jamal Noori



Department of Physics Lancaster University

February 2017

This thesis is submitted in partial fulfilment of the requirements for the degree of Doctor of Philosophy

Declaration

I declare that this thesis, unless stated otherwise, is the result of my own work which I carried out in Lancaster University between September 2013 and August 2016. To the best of my knowledge and belief, it contains no material previously published or produced by another party in fulfilment, partial or otherwise, of any other degree or diploma at another university or institute of higher learning, except where due acknowledgement and reference is made in the text. This thesis is not the same as any others I have already submitted, or in the process of submitting, for any degree at any university or institution. The word count on this thesis does not exceed the maximum limit of 80,000 words.

mode

Y. J. Noori, Lancaster, 28 Feb 2017

Acknowledgements

I would first and foremost like to direct my sincere appreciation and gratitude to my supervisor, Robert J Young, for his excellent supervision and invaluable support and guidance throughout the period of my PhD. His great passion and motives for research has and continues to fuel that of mine. I would like to thank him for giving me the opportunity to be part of his research project, many thanks for all the fascinating projects he offered and for the freedom I was given during my work. Rob has helped defining the researcher I am today and I do not doubt that without him, this project would not have been possible.

I would like to thank my second supervisor Utz Roedig for supervising my work from the computer science side and for being always ready to give me the support and guidance when I needed him. A lot of gratitude to Phillip Speed for being first a great friend and secondly a very wise business partner, who can always boost my energy to let me provide more for the sake of my research and future career.

I've been very lucky to have had the chance to work with a number of amazing people during my studies in Lancaster University. The friendship I developed with Yameng, Jonny, Chris, Ramon, Ethem and Matt have been very effective in reducing the pressure that I could have otherwise faced without them from working hard during my PhD. I feel very happy to have been easily able to blend in well with them despite the cultural and language barriers.

A lot of thanking goes to Andrew R J Marshall and Aiyeshah Alhodaib for growing the InAs nanowires, Peter Hodgson for growing the GaSb quantum dots, Yuri Pashkin, Atif Aziz and Richard George for giving me the trainings I needed in the Quantum Technology Centre and the invaluable experimental support and discussions that have helped me with my work.

My parents Nada and Jamal, my siblings Dhafer and Rania and my love Dania, I could have never have reached anywhere close to where I am now without your love, support and constant encouragements. Thank you for teaching me to believe in myself and in my dreams. I have and will always try to make you proud. I love you all.

To my current and previous house mates, Maysara, Kabeer, Mohammad, Sharih, Ahmed and Iznan, the joyful time I spend with you every night after a long day of work has been very effective at clearing my head from work matters and keeping my stress levels down.

Finally, I extend my gratitude to the faculty of Science and Technology for funding me throughout my fascinating doctorate journey.

List of publications

- J. R. Orchard, C. Woodhead, J. Wu, M. Tang, R. Beanland, Y. J. Noori, H. Liu, R. J. Young, D. J. Mowbray, "Silicon-based single quantum dot emission in the telecoms C-band" ACS Photonics, 4, 1740-1746 (2017)
- C. S. Woodhead, J. Roberts, Y. J. Noori, Y. Cao, R. Bernardo-Gavito, P. Tovee, A. Kozikov, K. Novoselov and R. J. Young, "Light extraction from 2D materials using liquid formed micro-lenses" IOP 2D Materials, 4, 015032 (2016)
- Y. J. Noori, Y. Cao, J. Roberts, C. S. Woodhead, R. Bernardo-Gavito, P. Tovee and R. J. Young, "Photonic crystals for enhanced light extraction from 2D materials" ACS Photonics, 3, 2515-2520 (2016)
- J. Roberts, I. E. Bagci, M. A. M. Zawawi, J. Sexton, N. Hulbert, Y. J. Noori, C. S. Woodhead, M. Missous, M. A. Migliorato, U. Roedig and R. J. Young, "Atomic-scale authentication with resonant tunnelling diodes" MRS Advances 1, 1625 (2016)
- J. Roberts, I. E. Bagci, M. A. Zawawi, J.Sexton, N.Hulbert, Y. J. Noori, M. P. Young, C. S. Woodhead, M. Missous, M. A. Migilorato, U. Roedig and R. J. Young, "Using quantum confinement to uniquely identify devices" Nature Sci. Rep. 5, 16456 (2015)
- M. P. Young, C. S. Woodhead, J. Roberts, Y. J. Noori, M. T. Noble, A. Krier, E. P. Smakman, P. M. Koenraad, M. Hayne and R. J. Young, "Photoluminescence studies of individual and few GaSb/GaAs quantum rings" AIP Advances 4, 117128 (2014)
- R. Bernardo-Gavito, J. Roberts, J. Sexton, B. Astbury, H. Shokeir, T. McGrath, Y. J. Noori, C. S. Woodhead, M. Missous, U. Roedig, R. J. Young, "Extracting random numbers from quantum tunnelling through a single diode" <u>arXiv:1706.07348</u> (Manuscript submitted to Nature Electronics, 2017)
- Y. Cao, A. J. Robson, A. Alharbi, J. Roberts, C. S. Woodhead, Y. J. Noori, R. Bernardo-Gavito, D. Shahrjerdi, U. Roedig, V. I. Fal'ko, R. J. Young, "Optical identification using imperfections in 2D materials" <u>arXiv:1706.07949</u> (Accepted for publishing in IOP 2D Materials, 2017)
- 9. Y. J. Noori, R. J. Young. "A photonic device" GB1706216.7 (18 July 2017)
- A. Alhodaib, P. J. Carrington, Y. J. Noori, Ana M. Sanchez, M. D. Thompson, M. C. Rosamond, Edmund H. Linfield, A. Krier, A. R. J. Marshall, "Room temperature mid-infrared emission from faceted InAsSb multi quantum wells embedded in InAs nanowires" (Submitted to Nature Communications, 2017)

Abstract

The security of current public key cryptosystems, such as RSA, depends on the difficulty of computing certain functions known as trapdoor functions. However, as computational resources become more abundant with the fast development of super- and quantum computers, relying on such methods for communication security becomes risky. Quantum key distribution (QKD), is a potential solution that can allow theoretically secure key exchange for future communications. Chip-scale integration of this solution for securing communication of embedded systems and hand held devices demands miniaturizing the optical components that are used in typical QKD boxes, hence reducing its size and cost. The aim of the work in this thesis is firstly investigating novel approaches to realising integrable single photon sources and detectors for applications such as QKD, and secondly proposing a chip-scale integrated QKD system with efficient and optimised optical components.

In the first part of the thesis, a model for coupling 2D material emitters to rod-type photonic cavities is studied for room temperature single photon sources. Our investigated approach allows better coupling between the emitter and the cavity modes than conventional methods, while increasing light collection ratio. In the second part, site-controlled growth of semiconductor III-V nanowires on Si for photodetection applications is achieved by fabricating the sites using electron-beam lithography and wet etching. Studies were also carried out to investigate the effect of the wafer's growth temperature on the nanowire formation. Finally, a model was proposed for realising a chip-scale QKD system using photonic crystals as a photonic circuit platform. The work involves increasing the Q-factor of the cavity single photon source, increasing cavity waveguide coupling, reducing losses in beam splitters and outcouplers. A final model of a chip-scale QKD system which involves the optimised components is proposed at the end of the thesis.

Table of Contents

Chapter	1	1-1
1.1.	Classical Key Sharing Schemes	1-1
1.2.	Quantum Cryptography for Secure Key Sharing	1-3
1.3.	Practical Challenges in QKD	1-6
1.4.	Thesis Outline	1-8
Biblio	graphy	1-11
Chapter	2	2-1
2.1.	Semiconductors and Quantum Dots	2-1
2.1.	2. Self-Assembled Quantum Dots	2-3
2.1.	3. Defects in 2D Materials	2-3
2.2.	Photonic Crystals	2-4
2.2.	1. Photonic Band Structure	2-4
2.2.	2. Photonic Crystal Resonators and Waveguides	2-6
2.2.	3. Purcell Enhancement	2-10
2.3.	Single Photon Detectors	2-12
2.3.	1. P-I-N Junctions	2-13
2.3.	2. Avalanche Multiplication	2-14
2.4.	Differential Phase QKD Protocols	2-15
Biblio	graphy	2-19
Chapter	3	
3.1.	Introduction	3-1
3.2.	Fabrication Techniques	
3.2.	1. Electron-Beam Lithography	
3.2.	2. Thermal (Resistive) Evaporation	
3.2.	3. Lift-Off	
3.2.	4. Molecular Beam Epitaxy	3-9
3.2.	5. Etching	3-9
3.2.	6. RIE and RIE-ICP	3-11
3.3.	Scanning Probe Microscopy	3-14
3.4.	Fabrication of Hole-Type Photonic Crystals	3-16
3.5.	Numerical Modelling With Finite Difference Time Domain	
3.6.	Micro-Photoluminescence Spectroscopy	3-25
Biblio	graphy	3-27

Chapter -	4	4-1
4.1.	Introduction	4-1
4.2.	TMDC Monolayers as Quantum Light Emitters	4-2
4.3.	Cavity Fabrication and Emitter Transfer	4-11
4.4.	Conclusions	4-17
Biblio	graphy	4-19
Chapter	5	5-1
5.1.	Introduction	5-1
5.2.	Site-Controlled Nanowire Fabrication	5-6
5.3.	InAs and GaAs Nanowire Growth	5-9
5.4.	Conclusion	
Biblio	graphy	
Chapter	б	6-1
6.1.	Introduction	6-1
6.1.	1. Motivation	6-2
6.1.	2. GaAs as an Optoelectronic Material for QKD	6-3
6.2.	Components for QKD	6-5
6.2.	1. Single Photon Cavity for QKD	6-5
6.2.	2. Integrated Waveguides for QKD	6-7
6.2.	3. Photon Out-Coupling for a QKD Chip	6-10
6.3.	Numerical Optimisation of QKD Components	6-11
6.3.	1. Optimising Cavity Confinement for Single Photon Emission	6-11
6.3.	2. Optimising Waveguide-Cavity Coupling for Efficient Beam splitting	6-16
6.3.	3. Y junction Photonic Crystal Beam splitters for Integrated QKD Systems	6-19
6.3.	4. Optimised Light Out-Coupling Efficiency from a Photonic Crystal Waveg	uide6-20
6.4.	Assembling a Chip-Scale QKD System	6-23
6.5.	Preliminary Experimental Results for Photonic Crystals Using GaSb QDs	6-23
6.6.	Conclusions	6-26
Biblio	graphy	
Chapter	7	7-1
7.1.	Conclusions	7-1
7.2.	Future Work	7-6
Appendi	х	A-1

Chapter 1

Introduction

1.1. Classical Key Sharing Schemes

Since the advent of the Internet, continuous development in its security has been a major challenge for researchers and industrial organisations that specialise in this field. Today, the importance of the Internet's security is not solely based on its usage for social media or online shopping, but if it were to be breached by attackers, human lives could be put in danger and many modern countries' national securities could become vulnerable. This has always been a driving motivation to further research and develop the security of communication systems. An example of the serious importance of network and Internet security is its involvement in online voting and crisis management. In the present day, the internet is used extensively for national economy control and simultaneously for the resolution of military tasks. These sensitive uses of the Internet make its security an essential goal that needs to be achieved at all times.

Today, when valuable data such as credit card numbers are sent to an online store for a payment, public key cryptography protocols such as Transport Layer Security (TLS) are used to authenticate the store's server¹. After the authentication, TLS is used to negotiate a symmetric session key, which can later be used for encrypting data, such as the credit card's number, transferred between the two parties. This process is also known as key exchange/distribution/negotiation/establishment. The exchanged key can later be used by both parties for data encryption and decryption. The security of encryption depends

entirely on the key and the encryption scheme. If a secure encryption scheme was used, in combination with a secret key, the only way for an eavesdropper to have knowledge about the transmitted information is through knowing the key used for encryption. Hence, firstly, the key exchange mechanism used for sensitive applications should be ultimately secure to prevent eavesdroppers from obtaining any information about the key. Secondly, the key should be used with a provably uncrackable encryption technique such as the one-time pad. This will prevent an eavesdropper from obtaining any information about the key exchange and encryption are executed correctly.

Common existent techniques of key exchange between multiple parties depend on what's known by mathematicians and cryptographers as trapdoor functions. RSA is a popular protocol that exploits trapdoor functions to allow key exchange. The security of RSA for key exchange relies on the assumption that it is computationally infeasible to factorise a large number into its prime factors. If an eavesdropper, Eve, can listen to what is sent across a communication channel during an RSA protocol between a sender, Alice, and a receiver, Bob, then she should be able to decrypt the cipher using just Bob's publicly revealed key, but only if she has enough computational power. If that was the case, Eve would be able to decrypt the cipher and recover the symmetric key. In other words, current cryptography systems that use RSA and complexity related schemes for key exchange are vulnerable if an attacker were to have access to high computational resources, making one way functions reversible.

This security scheme, to some extent is a temporary solution. The constant development of super and quantum computers can lead to faster and more efficient breaking of RSA schemes by eavesdroppers who have access to such technology. Although the latter is not yet a reality, the basic principles have been proven in laboratories and theoretically have been shown to be especially capable for computing complex mathematical equations such as trapdoor functions^{2, 3}. If such computation capabilities are achieved, a future adversary could break the encryption of many communication channels that depend on RSA for their security, introducing a retroactive vulnerability overnight. One of the theories that was published and led to an increase in security development efforts was Shor's algorithm for quantum computing. The algorithm was developed for factorising large prime numbers in an exponentially faster

time by using a quantum computer. This led to a great increase in research efforts in this field after 1994. Since then, researchers have been pushing the limits of data transfer rates and the maximum achievable communication distances using quantum phenomena-based provably secure systems.

1.2. Quantum Cryptography for Secure Key Sharing

Provably secure systems use quantum mechanical principles for securing communication, this is commonly known as quantum key distribution (QKD). QKD prevents an eavesdropper from listening to the QKD channel without being discovered using the unbreakable fundamental laws of physics. This sets upper bounds on the amount of knowledge that Eve can obtain about the transferred data between Alice and Bob.

Like classical key distribution methods, QKD is also not immune against active man in the middle attacks. Thus, prior to Alice and Bob using QKD as a key distribution protocol, they need to have an authenticated quantum and classical channel. This ensures they do not succumb to an active man in the middle attack. This means a seed key is needed prior to starting the QKD scheme, to be used by Alice and Bob to authenticate each other. For this reason, QKD is often referred to as a key growing protocol.

The first QKD protocol, BB84, was proposed by Bennett and Brassard in 1984, and since then several recognised security proofs have been reported to prove its security^{4, 5}. The protocol begins with Alice preparing her random digital key that she would like to share with Bob. Next, Alice and Bob agree on different polarisation bases for bit encoding. For example, in the rectilinear basis, Alice and Bob can use vertically and horizontally polarised photons to represent 0 and 1, respectively. On the other hand, they represent 0 and 1 using photons polarised at 45 and 135 degrees in the diagonal basis. In sending these bits, Alice uses a trusted random number generator (RNG) to make her choice for the basis she uses for encoding each qubit on each photon pulse, before sending the pulses to Bob for measurements. On receiving the qubits, Bob also performs his measurements using a random number generator to make his basis choices.

Following the measurements stage, Alice and Bob publicly broadcast their chosen encoding basis and measurement basis for every single qubit. A comparison session between the encoding and measurement bases is then undertaken to filter the bits that will later form the key. Any bit where the corresponding qubits had been encoded and measured with non-matching bases is dropped from the bit sequence by Alice and Bob. This is because there is a probability of 50% that a qubit measured by Bob with a non-matching basis will be opposite to Alice's intended bit making the key bit Bob ends up with differ from Alice's. Therefore, Alice and Bob only keep the bits that were measured by matching bases. Table 1 shows an example of a typical BB84 scenario carried out between Alice and Bob. Revealing of information related to the basis choices through a public channel can reveal information about a percentage of the key bit string but not the total bit string. Knowledge about the bit information leaked to the eavesdropper in this process can be minimised in the privacy amplification stage.

Alice's random bit		0	0	1	0	1	1	0	+ Rectilinear Basis
Alice's random basis choice	+	X	+	X	X	+	X	+	× Diagonal Basis
Photon polarization	♦	1	1	\mathbf{N}	1	1	\mathbf{Z}	1	\rightarrow 0 degree polarisation
Bob's random measuring basis		X	X	X	+	X	+	+	45 degree polarisation
Bob measurement	↔	7	$\mathbf{\mathbf{N}}$	\mathbf{N}	↔	/	†	1	90 degree polarisation
Shared secret key	1	0		1				0	🔪 135 degree polarisation

Table 1: A typical BB84 scenario showing the random basis choices by Alice and Bob for bit encoding and measurement, and the key both parties agreed on after they compare their basis choices. Black cells in the table correspond to ignored photons where Alice and Bob used different basis.

Now assuming an eavesdropper, Eve, was trying to gain information by listening to the channel in which the photons have been transmitted through. She will do this using what is known as the intercept-resend attack⁴. In this attack, Eve intercepts the incoming photons and attempts to measure them with two different polarizing splitters for the two bases Alice and Bob previously agreed on. When Eve intercepts the stream she will have no knowledge about which of the two bases Alice has used for encoding every bit, and will have to measure and send the photons using a random basis of her choice. On average, only half of the measurement bases she chooses will match that of Alice's choices. If Eve selects a different basis from Alice to measure a qubit, she will force the photon polarization to her own basis' 0 or 1 polarisation. For example, if a photon is prepared in the 45 degrees state (i.e in a

superposition of horizontal and vertical polarisations), and Eve uses the rectilinear basis, her measurement act will destroy the superposition and render the photon in either the horizontal or vertical polarisation with equal probabilities. When Eve sends the photon she measured to Bob, he will also randomly select a basis for his splitter to measure the bit value. If he selects a similar basis to Alice that is different from Eve, there is a 50:50 chance that he will measure a different polarization which could give the opposite bit from Alice. When Alice and Bob compare their results they will find that this particular bit is flipped, which dictates the presence of an eavesdropper who attempted to measure the transmitted photon and was unlucky enough to have selected the wrong measuring basis for that particular bit. However, if Eve performs her measurement with the wrong basis but the result of Alice and Bob's bits agree, Eve will be lucky enough to get away without being detected.

Alice's random bit	1	0	0	1	0	1	1	0
Alice's random basis choice	+	X	+	X	X	+	X	+
Photon polarization	↔	/	1	\mathbf{N}	/	+	\mathbf{N}	1
Eve random measuring basis	+	+	X	‡	X	X	X	‡
Eve measurement	1	1	$\mathbf{\mathbf{N}}$	1	$\mathbf{\mathbf{N}}$	1	>	1
Bob's random measuring basis	+	X	X	X	‡	X	\$	†
Bob measurement	÷	\mathbf{N}	\mathbf{N}	\mathbf{N}	ŧ	7	ŧ	1
Shared secret key	1	1		1				0
Errors (C orrect or Incorrect) bit	C	I		С				С

Table 2: A typical BB84 scenario showing how the presence of an eavesdropper will result in bit flips when the basis used by Eve for measurement don't match that for Alice and Bob. This is exactly the case with the second bit in the bit sequence from the table. This bit flips can be later be spotted by Alice and Bob when a portion of the final key is compared, indicating the presence of an eavesdropper.

In order to be able to detect an eavesdropper, Alice and Bob will need to compare their bit results for the photons that they both prepared and measured with matching bases. If the set of bits they compare is large enough, then Eve's probability in matching Alice and Bob's bases for the whole set is very low. If the bit sequence Alice and Bob compare match on both sides, then they can be certain that there is no eavesdropper listening to the channel. After transferring the key bit sequence and completing the basis comparison step, Alice and Bob randomly select a subset of bits to check for the presence of Eve. If no error is found between Alice's and Bob's bits strings then they can use the rest of the key (sifted key) for subsequent symmetric data encryption or signing.

Bob has a 50% chance of measuring each received photon with the correct basis. The same probability applies to Eve's chance of choosing the correct basis. Detecting Eve takes place only when she chooses the wrong basis while Bob choses the right basis, hence there is a 25% chance of this happening. If the explained situation takes place, there is a 50% probability in Bob receiving a bit flip at his end. Therefore, in the presence of Eve constantly intercepting and measuring photons without any knowledge of the basis Alice or Bob use for sending and measuring, there is a theoretical probability of 12.5% of every bit being flipped due to her act and arrive inverted at Bob's end. The presence of Eve listening to the quantum channel will increase the Quantum Bit Error Rate (QBER) in the distributed key, where QBER is the ratio of an error rate to the key rate. Practically, the presence of Eve listening to the quantum channel is not the only factor that increases the QBER. The different protocols available in the literature can cope with different QBERs, however, for common BB84 coherent one way systems this is in the order of 11%.

1.3. Practical Challenges in QKD

QKD was first demonstrated experimentally in 1989 where the results were published by Bennett, Bessette et al. in 1992⁶. Since then, QKD systems have routinely been realised in labs and commercially sold by several companies⁷⁻¹⁰. However, there are many practical challenges that prevent widespread adoption such as:

• *Distance* – Since QKD operates at the single photon level, light absorption and scattering in fibre-optics limit the achievable communication distance to a few hundred kilometres before the use of repeaters is necessary. However, due to the no-cloning theorem in quantum physics, repeaters used in classical communications cannot be used in a QKD channel. Quantum repeaters are still far from being practical, mainly due to decoherence. An important missing component is a robust quantum memory, which is an essential part of a quantum repeater¹¹⁻¹³.

Hence, the current method to share a key between two distant parties is to use one or multiple trusted nodes^{14, 15}, Charlie, who operates as a receiver with Alice and as a sender with Bob. The drawbacks of light absorption and scattering does not reduce the achievable distance only, but it also has a direct effect on the achievable key bit rate¹⁶.

- Single photon sources current commercial QKD products use attenuated coherent lasers light as their single photon sources. However, the photon number characteristics for an attenuated laser pulse exhibit a Poisson distribution, making the probability of emitting more than a single photon per pulse finite; this gives Eve the chance to perform photon number splitting attacks¹⁷. To limit the probability of emitting more than a single photon, the mean number of photons per pulse is usually around 0.2 photons per pulse. This means, on average, less than one fifth of the pulses are used to carry information, this limits the achievable key bit rate. Realising an on demand single photon source is essential for increasing the bit rate of QKD without introducing retroactive security concerns.
- Single photon detectors state of the art QKD boxes that operate at telecom wavelengths use InGaAs infrared single photon detectors¹⁸. However, the limited quantum efficiency of these detectors and their high dark count rate makes them a limiting factor to the practical achievable key bit rate. Most of these issues are solved by cooling the systems to low temperatures, however, for practicality purposes, approaches to realising efficient room temperature single photon detectors, such as using nanowires are being investigated.
- *Bit rate* As discussed before, this obstacle is directly related to the achievable distance, nonideal single photon sources and single photon detectors. In addition, other reasons that might affect the bit rate could be the use of inefficient error correction and privacy amplification algorithms that might be lossy and slow to run with current available processing hardware.
- *Cost and size* the size and cost requirements of current commercial QKD boxes prevents the technology from inhabiting any consumer grade electronics such as PCs, phones, embedded systems and makes it monopolised by bodies with fund abundance such as banks and

government organisations. This is due to the challenges faced in integrating the necessary optical components of the system, including the single photon source, the waveguides, the single photon detector and its cooling hardware, the processing electronics and the power supplies.

The extensive work in the development of integrated optical devices has shown strong potential for advancing the integration of quantum technologies such as quantum computing and QKD. The aim of this thesis is to use currently available integrated photonics technologies to make advancements toward tackling the single photon source and the single photon detector issues. The last part of the thesis, will be aimed at proposing a model to directly solve the size and the cost issue for the whole QKD system by designing an integrated photonic chip that involves all the optical components needed to realise a QKD chip including the single photon source.

1.4. Thesis Outline

This thesis work concentrates on developing integrated optical components for quantum cryptography applications and addresses many of the integration challenges from theoretical and experimental perspectives. The long-term goal is to achieve practical and efficient fully integrated QKD systems that includes integrated single photon sources and detectors operating at room temperature. Although the aspect of this work is focused on quantum cryptography applications, the concepts and technologies introduced can be used to enable many other application in quantum information processing that are being developed in parallel, such as quantum computing

Chapter 2 of the thesis will start by providing a brief summary of quantum dots as single photon sources. The necessary background for the main theory and principles of photonic crystals as the suggested platform for integrated optical cavities and waveguides will follow. Next, a summary about Purcell enhancement using optical cavities for increasing the spontaneous emission rate of single photons will be given. The chapter will also involve the key physical concepts needed to understand the basics of semiconductor photon detectors, which will be needed when discussing the work of chapter 5

concerning nanowire single photon detectors. Finally, Differential Phase Shift (DPS) QKD will be introduced as the protocol used in the suggested QKD chip that was discussed in chapter 6.

Chapter 3 will start with a summary of the experimental techniques and working principles of the tools used in fabricating the optical components discussed in this thesis, such as electron-beam (e-beam) lithography, dry and wet etching techniques such as inductively coupled plasma and reactive ion etching and micro-photoluminescence. In the following section the fabrication methods for hole-type photonic crystals which involves a new e-beam resist with high temperature dry etching will be explained.

Chapter 4 will propose a new model for enhancing light emission from 2D materials and increasing light-matter interaction through the use of rod-type photonic crystal cavities. Finite Difference Time Domain simulation results of the cavity mode together with the enhancement obtained as the cavity rods radius is changed is demonstrated. In addition, enhancements obtained as the 2D material is allowed to dip inside the cavity is shown. Finally, a summary of the approach taken toward realising the model experimentally with preliminary results will be discussed.

Chapter 5 will introduce a new scheme for realising photodetectors with site controlled III-V semiconducting nanowires by epitaxial growth on Si. The introduction will briefly list the advantages of nanowire based photodetectors over conventional planar photodiodes. The chapter will later present results from fabricating the nanowire sites using state of the art e-beam lithography and CSAR as a positive e-beam resist. Electron micrographs of the epitaxial growth results of InAs and GaAs nanowires will be shown. The chapter will include investigation of the effect of growth temperature on the successful formation of nanowires and their achieved lengths. Image analysis of electron micrographs will be carried out and a discussion will follow to analyse the achieved nanowires lengths for photodetection applications.

Chapter 6 will introduce current state of the art integrated QKD systems. An alternative approach that harnesses quantum dot technology for single photon emission using GaAs as a host optoelectronic material will be suggested. Comparisons will be made between the different approaches to realising DPS-QKD integrated components. Numerical modelling for optimising each component and

preliminary experimental results will be presented before putting the proposal for the fully integrated QKD chip.

Chapter 7 will summarise the results achieved in this thesis including the challenges and the limitations faced throughout the research period. The chapter will discuss future plans that are currently being (or will be) investigated to take the research of 2D material based rod-type photonic crystals single photon sources, nanowire based photodetectors and chip based QKD an additional step further.

Bibliography

¹ T. Polk, K. McKay, S. Chokhani. Guidelines for the selection, the configuration, and use of transport layer security (TLS) implementations. NIST special publication 800-52 (2014)

² P. W. Shor. Algorithms for quantum computations: discrete logarithms and factoring. Proc. 35nd Annual Symposium on Foundations of Computer Science, IEEE Computer Society Press, 124-134 (1994)

³ D. Goodin. How the NSA can break trillions of encrypted web and VPN connections. ARS Technical UK. <u>https://arstechnica.co.uk/security/2015/10/how-the-nsa-can-break-trillions-of-encrypted-web-and-vpn-connections/</u> (accessed 10/10/2016)

⁴ P. W. Shor, J. Preskill. Simple proof of security of the BB84 quantum key distribution protocol. Phys. Rev. Lett., 85, 441-444 (2000)

⁵ R. Renner, N. Gisin, B. Kraus. Information-theoretic security proof for quantum-key-distribution protocols. Phys. Rev. A., 72, 012332 (2005)

⁶ C. H. Bennett, F. Bessette, G. Brassard, L. Salvail, J. Smolin. Experimental quantum cryptography. J. Cryptology, 5, 3-28 (1992)

⁷ B. Korzh, C. C. W. Lim, R. Houlmann, N. Gisin, M. J. Li, D. Nolan, B. Sanguinetti, R. Thew, H. Zbinden. Provable secure and practical quantum key distribution over 307 km of optical fibre. Nature Photon., 9, 163-168 (2015)

⁸ P. Jouguet, S. Kunz-Jacques, A. Leverrier, P. Grangier, E. Diamanti, Experimental demonstration of long-distance continuous-variable quantum key distribution. Nature Photon., 7, 378-381 (2013)

⁹ Toshiba Cambridge Research laboratory. Toshiba QKD system. <u>http://www.toshiba.eu/eu/Cambridge-Research-Laboratory/Quantum-Information-Group/Quantum-Key-Distribution/Toshiba-QKD-system/</u> (last accessed 12/12/2016)

¹⁰ ID Quantique limited. An overview of QKD. <u>http://www.idquantique.com/quantum-safe-crypto/qkd-overview/</u> (last accessed 12/12/2016)

¹¹ P. Kok, C. P. Williams, J. P. Dowling. Construction of a quantum repeater with linear optics. Phys. Rev. A, 68, 022301 (2003)

¹² K. Azuma, K. Tamaki, H. Lo. All-photonic quantum repeaters. Nature Commun. 6, 6787 (2015)

¹³ W. J. Munro, J. Azuma, K. Tamaki, K. Nemoto. Inside quantum repeaters. IEEE Journal of Selected Topics in Quantum Electronics, 21, 6400813 (2015)

¹⁴ M. Sasaki, M. Fujiwara, H. Ishizuka, et al.. Field test of quantum key distribution in the Tokyo QKD network. Optics Express, 19, 10387 (2011)

¹⁵ C. Elliott. Building the quantum network. New Journal of Physics, 4, 46.1-12 (2002)

¹⁶ E. Diamanti, H. Lo, B. Qi, Z. Yuan. Practical challenges in quantum key distribution. NPJ Quantum Information, 2, 16025 (2016)

¹⁷ G. Brassard, N. Lutkenhaus, T. Mor, B. C. Sanders. Limitations on practical quantum cryptography. Phys. Rev. Lett., 85, 1330-1333 (2000)

¹⁸ J. Zhang, M. A. Itzler, H. Zbinden, J. Pan. Advances in InGaAs/InP single-photon detector systems for quantum communication. Light: Science & Applications, 4, e286 (2015)

Chapter 2

Background and Theory

In this thesis, optoelectronic components for integrated quantum information devices are studied, specifically for uses in quantum communications. The operation principles of these components utilise the fundamental physical properties of semiconductors, quantum confinement and photonic crystals (PhCs). This chapter will provide the necessary background materials and theory needed for understanding these properties and how they are utilised to realise useful optoelectronic devices.

2.1. Semiconductors and Quantum Dots

2.1.1. Basics

Semiconductor materials have their conductivity lie between that of insulators and conductors at room temperatures. An attractive aspect of semiconductors is that the conductivity in these materials can be engineered to suit different purposes¹. Due to the periodicity of the crystal lattice structure of semiconductor materials, an electron wave-function travelling through the lattice interacts with a periodic electric potential. This periodicity results in discrete energies where the electron wave-function is allowed to exist whilst providing other energies that the electrons are forbidden to have. The forbidden and allowed energy ranges can be calculated by solving the Schrödinger equation for an electron wave-function in the periodic lattice potential, where the solutions are known as Bloch waves². These solutions can be described visually as the electronic band structure for the lattice³. At both ends of the forbidden energy range, otherwise known as the bandgap, exist the valence band and the conduction

bands. For intrinsic semiconductors, the valence band is the highest range of electron energies that the electrons can have at absolute zero temperature. On the other hand, the conduction band is the lowest range of vacant electronic states. It will be shown later in this chapter that photonic band structures can also be found for a photon wave-function travelling through a periodic structure of materials of different dielectric constants.

Quantum dots (QDs) are formed in semiconductors by altering the system's band structure such that exciton confining potential wells are made⁴. This can be achieved by three dimensionally modifying the physical structure of the system on a length scale smaller than the electron's de Broglie wavelength. If the confinement is large enough, an electrons' and/or holes' wave-function can be confined in all dimensions. An electron in the QD material can be excited from the valence band to the conduction band by absorbing a photon, leaving behind a hole, where the electron and the hole can bind together due to attractive columbic forces forming an exciton. When the electron relaxes back to the valence band, by combining with the hole, a single photon is emitted. This makes QD systems ideal for single photon source applications such as quantum key distribution (QKD)⁵.

The energy of a photon emitted by a quantum dot is the sum of the bandgap energy of the quantum dot material, the confinement energy and the exciton Coulomb energy, as shown in equation 2.1.

$$E = E_{bandgap} + E_{confinement} + E_{Coulomb}$$
(2.1)

The confinement energy of an electron wave-function is directly dependent on the bandgap difference between the host material and the QD material, and on the QD size. This can be calculated using the particle in a box approximation. Coulombic attraction between the negatively charged electron and the positively charged hole can influence the total photonic energy of the quantum dot. This attraction is directly proportional to Rydberg's energy and inversely proportional to the square of the dielectric constant of the QD material.



Figure 2.1: Illustration of the self-organised growth of semiconductor QDs. A QD material is deposited on the substrate using epitaxial methods. The lattice constant difference between the QD material and the substrate causes the QD layer to cluster into QDs as its thickness is increased.

Conventionally, there are different forms of QD systems, for example, colloidal QDs⁶, Josephson junction QDs⁷, however, the two forms that are of interest in this work are self-assembled QDs⁸ and zero-dimensional defects in 2D materials⁹.

2.1.2. Self-Assembled Quantum Dots

Self-assembled QDs are made by growing a thin semiconductor material very slowly, using Molecular Beam Epitaxy (MBE) or Chemical Vapour Deposition (CVD), on top of another semiconductor material of differing lattice constant^{10,11}. After growing a few monolayers, the strain caused by the lattice mismatch between the two semiconducting materials causes the uniform layer structure to collapse and form three-dimensional islands as shown in Figure 2.1. This is known as the Stranski-Krastanov growth technique¹². The QDs are usually capped with a thick layer of the original material to enhance their formation due to strain and protect them.

Different materials have been used to create self-assembled QDs such as GaSb/GaAs¹³, InAs/GaAs¹⁴, InN/GaN QDs¹⁵.

2.1.3. Defects in 2D Materials

2D materials are crystalline materials that are comprised of a single or a few layer atoms. Since the discovery of graphene, 2D monolayer materials with unique optical properties, such as transition metal dichalcogenides (TMDCs), have been investigated. Examples of these include molybdenum disulphide

(MoS₂) and tungsten diselenide (WSe₂). These were found to exhibit similar optical properties to semiconductor materials such as GaAs. Many TMDC monolayers, such as the ones previously mentioned, have a direct bandgap and their optical emission wavelength lies in the visible range making them good light absorbers/emitters. In addition, TMDC monolayers benefit from the unique optical properties of quantum wells due to their two-dimensional nature giving excitons discrete energy states.

The simplest and most abundant type of QDs in semiconductor TMDCs are vacancies, while the less commonly found type are dopants in the lattice¹⁶. The introduction of defects such as atomic vacancies can be done by fluctuating the growth conditions of CVD grown TMDC monolayers. Dopants in the lattice can be introduced through ion/electron irradiation, high temperature annealing and plasma processing. Similar to QDs in bulk semiconductor materials such as GaAs, QDs in 2D materials have been reported to be capable of emitting single photons⁹, making them a potential source of single photons for quantum information applications.



Figure 2.2: (a) A schematic illustration of the atomic structure of a typical, (a) pure, (b) defective and (c) impure MoS2 monolayer flakes.

2.2. Photonic Crystals

2.2.1. Photonic Band Structure

In the first part of this chapter, the basics of the electronic band structure in semiconductor crystal materials was summarised. In this section, an overview of the photonic band structure in PhCs will be introduced. Interestingly, the theory of the electronic wave-functions in a semiconductor crystal and photonic wave-functions in PhC are very similar¹⁷. A PhC consist of a periodic optical structure of different materials with different dielectric constants. PhCs can be found in nature in different places, an example of this is the colourful pattern on an insect's eye as shown in Figure 2.3.



Figure 2.3: *An image of a female Tabanus horse fly showing the colourful pattern on the fly's eye which results from the diffraction of white light into different colours (a). An electron micrograph image of a fly's eye showing the periodic pattern that causes the colourful appearance of the eye (b). *Royalty free from: <u>http://www.earthporm.com/11-incredible-eye-macros/</u>



Figure 2.4: Illustrations of a 1D (a) and a 3D (b) PhC. The blocks with different colours, yellow, grey and cyan represent materials with supposedly different dielectric constants.



Figure 2.5: Examples of 2D PhCs showing (a) a hexagonal array hole-type and (b) a square array rod-type lattice. The grey materials is usually made of different dielectric constants in comparison to the surrounding material. Both of those types will be explored and used for making photonic devices in this thesis, see chapters 4 and 6.

The periodicity in the photonic structure can be in one, two or three spatial dimensions, Figure 2.4 shows examples of a one and three-dimensional PhC, whilst Figure 2.5 shows examples of two-dimensional PhCs, which will be the focus of this thesis.

The different properties of PhCs including the periodic lattice constant of the lattice, the shape and size of the periodic materials and the different dielectric constants of the materials allows the propagation of light with certain wavelengths and prohibits the propagation of other wavelengths. The range of wavelengths that are prohibited from propagating through the lattice is known as the photonic bandgap and is caused by the destructive interference of the light's wave-function due to reflection at the alternating layers. This is analogous to the concept of an electronic bandgap that prohibits the propagation of electronic wave-functions with certain energies. To illustrate this idea, Figure 2.6 shows the transmission spectrum obtained as a light wave travels through a square PhC structure made of high dielectric rods in air, and is collected by a flux region at the opposite side of the source¹⁸.



Figure 2.6: An example of a simulation setup of a wave emitted from a plane wave source and propagating through a PhC towards a detector on the other side. (b) The transmission spectrum obtained from the detector showing a wide bandgap range from 0.250 to 0.425 with photonic edge states on both sides of the band gap.

The Poynting vector is the rate of energy transfer per unit area of an electromagnetic field¹⁹. In the Finite Difference Time Domain (FDTD) simulation in Figure 2.6, the power spectrum transferred through a flux region for a field frequency ω is calculated via the integral of the Poynting vector over the flux region, and subsequently accumulated over *n*-time steps. The process is repeated for every frequency to construct the flux spectrum.

2.2.2. Photonic Crystal Resonators and Waveguides

If a defect in the PhC lattice is introduced, a photonic cavity can be created in the structure. Normally, a defect in the lattice can be made by removing one or more of the lattice cells. Other methods of

creating defects is by changing the size and/or the dielectric constant of one or more of the lattice cells. In the example shown in Figure 2.7 (a), removing one of the rods creates a monopole defect cavity. Introducing a defect can cause a localised state to form within the bandgap of the dispersion spectrum of the crystal. This means when a light wave travels through the lattice, somewhere within the bandgap transmission curve, a state can be found corresponding to the cavity mode, as illustrated in Figure 2.7 (b).

In the one-dimensional Bragg mirror approximation, a photonic cavity in a material with refractive index, n (n = 1 in the illustration of Figure 2.7), has a mode when the cavity length L_{cav} is equal to an integer multiple, m, of the intra-cavity half wavelength, λ , and is given by:

$$L_{cav} = \frac{m\lambda}{2n} \tag{2.2}$$

The resonance occurs when there is a constructive interference caused by reflections of the wave due to the photonic bandgap. This constructive interference produces a sharp cavity mode within the bandgap region.



Figure 2.7: An example of a simulation setup of a wave emitted from a plane wave source and propagating through a PhC structure towards a detector with a removed hole forming a cavity. (b) The transmission spectrum obtained from the detector showing an opened cavity mode state within the band gap. The inset shows the E_y field profile of such a cavity.

The defect within a 2D PhC can be designed to confine a desired cavity mode wavelength within the plane of the crystal. To confine the cavity mode in the third dimension, previous work has used total

internal reflection rather than Bragg reflection. Krauss et al. were the first people to show experimentally a 2D PhC cavity mode that harnesses total internal reflection for light confinement²⁰.

The confinement strength of any cavity is determined primarily by the Q-factor of the cavity. The Q-factor of a cavity is defined mainly by two parameters related to the cavity mode, these are the mode's frequency (or wavelength), ω , and the lifetime of a photon confined inside the cavity, τ . The lifetime of a photon inside the cavity can be rationally thought of as the duration a photon can stay within the cavity before it is scattered or absorbed. Ideal cavities have an infinite lifetime, i.e. the photons can be trapped inside them without escaping. In practice however, the lifetime of a cavity is finite. The inverse of the lifetime is the cavity decay rate, κ :

$$\kappa = \frac{1}{\tau_{cav}} \tag{2.3}$$

The Q-factor can then be defined as:

$$Q = \frac{\omega}{\kappa} \tag{2.4}$$

The most common PhC cavity design comprises one or more air-holes removed from a PhC hole lattice fabricated on a high dielectric constant material such as GaAs or Si. The most common of these are the H1 and the L3 cavities that are made of one and three removed holes, respectively. Scanning Electron Microscopy (SEM) images of the H1 and L3 PhCs are shown in Figure 2.8. Another common cavity design is based on the removal of one rod of a high dielectric constant periodic rod structure surrounded by air. SEM images of such a structure is shown in Figure 2.9. The latter cavity design will be the main type used in chapter 4, whilst the former will be mainly used in chapter 6.

The PhC cavities discussed before are defects within the photonic lattice and can be considered as a localised disruption to the periodicity that act as optical resonators capable of confining electromagnetic modes of discrete wavelengths located within the photonic bandgap wavelength range. A PhC waveguide can be considered as a series of specially coupled point defects. Coupling between these defects leads to the formation of a transmission band or guided mode in which the electromagnetic wave



Figure 2.8: (a) An example of an H1 hole-type PhC cavity. (b) An example of an L3 hole-type PhC cavity. The two figures show one and three removed holes of the lattice forming H1 and L3 cavities, respectively.



Figure 2.9: An example of a hexagonal lattice H1 rod-type PhC cavity, showing one missing rod from the hexagonal rods lattice to form a rod-type photonic cavity.

is confined in all directions but is allowed to travel along the series of defects. In a similar fashion to PhC cavities, PhC waveguides can utilise total internal reflection to confine light in the third dimension whilst being confined due to the photonic bandgap in the other two dimensions.

A single mode waveguide can be created by removing a row of holes in a hole-type PhC, such that there exists only one guided mode wavelength within the bandgap of the lattice. This makes PhC waveguides very useful in light transfer on a photonic chip, beside its potential use in integrated wavelength filters



due to its selectivity to couple to a discrete set of wavelengths²¹. A row of missing holes is called a W1 type waveguide and will be the main type considered in this thesis, see Figure 2.10.

Figure 2.10: SEM image of a typical W1 PhC waveguide designed for coupling to GaSb QDs with emission wavelengths of \sim 1250 nm. The waveguide is formed by creating a line defect in the lattice by simply omitting a line of holes to create a propagating mode in the photonic bandgap system.

W1 waveguides are important since they can cause slow light effects²². Slow light is a term used to describe light travelling through a medium with a group velocity that is much lower than the speed of light in that medium. Slow light has many important applications in quantum information. This is because by slowing light as it travels through a medium, stronger light-matter interactions can take place, providing many opportunities for applications in optical buffering and quantum memories^{23,24}. In chapter 6, slow light is suggested to be used in phase modulators, hence shortening the needed waveguide length for creating π -phase modulation compared to conventional phase modulators.

2.2.3. Purcell Enhancement

The advantage in using a photonic cavity system to couple to a single photon emitter such as a QD is to increase the spontaneous emission rate of single photons from the QD^{25} . This rate of single photon emission is directly related to the bit rate achievable when the system is used in a QKD chip. By placing a QD inside a photonic cavity, the electric field of the cavity mode can interact with the QD's ground and excited state. Chapter 3 will show how a cavity is fabricated around a QD for this type of experiment to be carried out.

The enhancement in the spontaneous emission rate of an emitter such as a QD coupled to a cavity resonator is known as Purcell enhancement²⁶. When an emitter decays in a cavity, the cavity confines the photon spatially in a small region and enhances the electric field locally. The Purcell factor is maximised when the QD that is in resonance with the cavity mode and its dipole is aligned with the maximum of the cavity mode's electric field. When this is the case, the Purcell factor can be written as follows:

$$F = \frac{3}{4\pi^2} \frac{Q}{V} \left(\frac{\lambda}{n}\right)^3 \tag{2.4}$$

where V is the mode volume. This means that the Purcell effect is directly related to the Q-factor of the cavity. Increasing the Q-factor whilst keeping other parameters constant transfers the cavity-QD coupling from the weak coupling regime to the strong coupling regime where the cavity and the QD have similar life times²⁷. However, strong coupling is not required for the case of a QD coupled to a micro-cavity for single photon source applications. The mode volume, on the other hand, is related to the cavity mode's wavelength divided by the refractive index of the cavity. Small mode volumes enhance the interaction strength between the cavity mode's electromagnetic field and the quantum emitter. For example, H1 and L3 cavities have been shown to have mode volumes in the order of $\left(\frac{\lambda}{n}\right)^3$, where *n* is the refractive index of the semiconductor material in which the mode is confined in, making them useful for single photon emission applications¹⁷. There are a number of tools that can be used to find these parameters. An example of one of these tools that was mainly used in this thesis is discussed in section 3.5.

2.3. Single Photon Detectors

Single photon detectors (SPDs) are one of the main components that determine the speed and efficiency of a QKD system²⁸. An ideal SPD is expected to click every time it receives a photon, at an arbitrary speed^{29,30}. Practical SPDs that are incorporated in QKD systems are far from ideal. For example, the quantum efficiency, which is the percentage of photons incident on the device's photoreactive surface and produce charge carriers, may be less than 100%. The dark count, which is the rate the detector outputs pulses in the absence of any incident photon, and the dead time, which is the time in which a detector is not capable of detecting a photon after a photon detection event, are both usually non-zero in practice. The dead time also reduces the "photon number resolution", which is the ability to distinguish the number of photons in an incident pulse. The time jitter is also an important factor, it can be defined as the time delay variations between the input of the optical signal and the output of the electrical signal. This in practice may also be non-zero.

There are different physical methods that allow the realisation of SPD devices. The first uses superconducting nanowires that are typically 5 nm thin, 100 nm in width and a few hundreds of microns in length patterned in a grid like structure³¹⁻³³. During operation, the wire is cooled below its superconducting critical temperature and a DC bias current that is close to but less than its superconducting critical current is applied. When a photon is absorbed by the nanowire grid, it breaks electron Cooper pairs and reduces the local critical current below that of the bias current, causing an increase in resistance at the region of the incident photon. This perturbation in resistance causes a change in the bias current, typically measurable by sensitive electronic instruments. Superconducting nanowire SPDs are known to have high quantum efficiency and relatively small dark counts, in comparison to avalanche photodetectors (APDs) that will be described later. However, the limiting factor that prevents this technology from being incorporated in commercial products in the future, is their operation temperature which is typically required to be only a few degrees above absolute zero.

A more interesting technique in realising SPDs is by exploiting the avalanche effect when a P-I-N junction is reverse biased. Such detector types have been implemented on Silicon and III-V materials.

In the next section, a brief overview of P-I-N junctions together with the working principles of APDs will be given as background material for chapter 5 which involves using nanowire P-I-N junctions for photodetection.

2.3.1. P-I-N Junctions

In section 2.1.1, the basics of semiconductor materials were introduced. It was introduced that semiconductors have been widely used in electronic devices due to the ability to engineer their conductivity for different applications. Changing the conductivity can be done by doping the semiconductors with atoms to introduce negative (n-doping) and positive (p-doping) charge carriers, electrons and holes. In the n- and p-doped regions, the carrier concentrations are a few orders of magnitude higher than the concentration of carriers in the intrinsic region. Sandwiching an intrinsic semiconductor region between an n-doped and a p-doped region makes a p-i-n junction. This structure has many applications in high power electronics and is suitable for fast switches, attenuators and photodetectors. The latter is the main use for p-i-n junctions in this thesis. Between the p-i-n junction, there exists a layer even larger than the intrinsic region, known as the depletion region. This is formed by charge carriers combining and annihilating one another through diffusion from the p- and the n-doped regions. This diffusion becomes limited by the electric field created by positive and negative ions left behind in the n and p regions, respectively.

Under forward bias, the p-type region is connected to the positive terminal of a power source and the n-type is connected to the negative terminal. Electrons and holes from the n-type and the p-type regions are forced towards the intrinsic region due to the potential difference of the source. Some charge carriers recombine at the intrinsic region and others continue to flow through the opposite side of the junction creating an electric current.

When a p-i-n junction is reverse biased, the depletion region widens and ordinarily the junction does not conduct, except for a small leakage current. If a photon of sufficient energy is absorbed by the depletion region of the diode, the absorption process creates an electron-hole pair within the depletion region, as shown in Figure 2.11. The applied reverse bias field pulls the electrons and holes, out of the region, toward the source's terminal, causing a current flow³⁴.



Figure 2.11: (Left) p-i-n junction under reverse bias, showing the regions of majority electron and hole carriers, an incident photon on the intrinsic region and the created electron-hole pair. (Right) Band diagram for the corresponding p-i-n junction with the created electron-hole pair due to an incident photon on the intrinsic region. The excited electron and hole carriers are attracted toward the n and p-regions respectively due to reverse bias, causing a current spike which can be amplified and measured by sensitive electronics.

2.3.2. Avalanche Multiplication

Avalanche multiplication is a phenomenon that can occur in semiconductor and insulating materials where an electric current multiplication allows high currents to be generated in an otherwise insulating materials²⁷. In p-i-n junctions, the phenomena occurs when, following the absorption of a photon, carriers are accelerated from the depletion region by the electric field of an applied reverse bias. If the charge carriers have enough kinetic energy, collisions with other bound electrons can excite them across the bandgap and create other free electron-hole pairs (secondary carriers). These secondary carriers accelerate due to the applied bias and gain enough energy to create even more carriers in a cascaded process. Hence a single photon can create a sudden increase in current at the junction. The current created by the multiplicative process of carriers due to the absorption of a single photon makes avalanche multiplication an ideal process for realising semiconductor SPDs.

2.4. Differential Phase QKD Protocols

The QKD scheme described in chapter 1, encodes quantum information on photon polarisations using two non-orthogonal bases. An alternative encoding approach uses the phase difference between successive photon pulses interfering at a beam splitter at the end of a Mach-Zehnder interferometer. This method of encoding information is known as Differential Phase Shift (DPS) QKD³⁵.

One of the key components in a DPS-QKD systems is a Mach-Zehnder interferometer. To illustrate the operation principles of DPS-QKD, it is important to explain how a Mach-Zehnder interferometer work for large photon number pulses. In this type of interferometers, the input laser beam is split in a 50:50 beam splitter, resulting in two beams, one can be called the modulated beam (marked yellow) and the other can be called the reference beam (marked red), as shown in Figure 2.12.



Figure 2.12: A block diagram of a typical DPS-QKD system with two pulses, coloured red and yellow, taking the different path in the Mach-Zehnder interferometers. The laser is input from the bottom left as shown by the blue arrow and split by a beam splitter. One arm contains a phase modulator which causes phase shifting one arm relative to the other. Constructive and destructive phase shifting can be recorded by detectors 0 and 1 respectively.

Let's define l_1 and l_2 as the total path length for the modulated and reference beams respectively. When the light passes through a glass beam splitter it exhibits a phase shift equivalent to $2\pi t/\lambda$, where t is the thickness of the beam splitter. For detector 0, the modulated beam takes a phase shift of π from the first beam splitter, a π phase shift from the top mirror, a $2\pi t/\lambda$ from the last beam splitter, and the path length of $2\pi l_1/\lambda$, before reaching detector 0. This gives a total of

$$2\pi + 2\pi \left(\frac{l_1 + t}{\lambda}\right)$$

The reference beam takes a phase shift of $2\pi t/\lambda$ as it goes through the first beam splitter, a π phase shift as it gets reflected by the bottom mirror and another π phase shift as it gets reflected by the second beam splitter toward detector 0. Hence the phase difference between the two beams ϕ , which is caused by the phase modulator is:

$$2\pi + 2\pi \left(\frac{l_1 + t}{\lambda}\right) - 2\pi - 2\pi \left(\frac{l_2 + t}{\lambda}\right) = \phi$$
$$2\pi \left(\frac{l_1 - l_2}{\lambda}\right) = \phi$$

In a similar fashion, the phase difference between the two arms for detector 1 is:

$$2\pi + 2\pi \left(\frac{l_1 + 2t}{\lambda}\right) - \pi - 2\pi \left(\frac{l_2 + 2t}{\lambda}\right) = \phi$$
$$\pi + 2\pi \left(\frac{l_1 - l_2}{\lambda}\right) = \pi + \phi$$

Hence, it is clear to note that when the phase modulator causes a zero phase shift, there is a constructive interference on the path to detector 0, and a destructive interference to detector 1. By varying ϕ , this condition can be changed, hence changing the probability of arrival at either detector.

A simplified example of a DPS-QKD system is shown in Figure 2., where Alice generates a train of single photon pulses and passes it through a Mach-Zehnder interferometer. In the interferometer, Alice splits the signal through a beam splitter, and applies a random phase modulation of 0, $\frac{\pi}{2}$, $\frac{3\pi}{2}$, or π on one arm and recombines the two arms at the junction of a second beam splitter. She later sends the signal to Bob through a quantum channel such as a fibre optic or free space. At Bob's side, he performs a similar Mach-Zehnder interferometry process, where he splits the signal into two arms, performs a random 0

or $\frac{\pi}{2}$ phase-shift through one arm and recombines the two arms in a beam splitter. There are three detection time slots at which Bob can measure. The first is when two consecutive pulses take the long paths on Alice's and Bob's interferometers. The second is when the two pulses both take the short paths of the interferometers. Both of these events aren't of use in this protocol. The only used events are when a pulse takes the long and short arm of Alice and Bob sides while the other takes the short and long arms of Alice and Bob respectively, as shown in Figure 2.. If the interferometers on both sides are matched, as when Alice and Bob use a similar basis, a 0 or π phase shift takes place between two successive photon pulses, causing trigger events in either detector 0 or 1, as shown in Figure 2.. In a real implementation, each single photon pulse is accompanied by one proceeding bright pulse for timing purposes. The bright photon pulse will appear as two pulses on Bob's detectors as it goes through the two long and two short paths in Alice and Bob sides.



Alice

Bob

Figure 2.13: A block diagram of a typical DPS-QKD system with two pulses, coloured red and yellow, taking the different path in the Mach-Zehnder interferometers.

After receiving the qubits, Bob announces publicly the time slots at which a photon was detected by one of his detectors, but does not reveal which detector detected it. From Alice's modulation data, she can know which detector in Bob's end recorded the event. By designating detection events recorded by detector 0 and 1 as key bit values 0 and 1, respectively, they can share an identical bit string.

In an ideal system where there are no errors in the detection events, the sifted key created is unconditionally secure. However, in practice, as discussed in chapter 1, all QKD systems have a baseline error rate, hence error correction and privacy amplification procedures are essential in a practical DPS-QKD system.


Figure 2.14: An example of a click event observed on detector 0 due to a 0 degrees phase shift representing a 0 bit. This occurs when two consecutive pulses take opposite and alternate arms on Alice and Bob's ends. The figure also shows timing pulses measured by the detector.

After error correction, when the error threshold is low enough to allow carrying on with the protocol, to nullify any information that may have been obtained by an eavesdropper, Eve, privacy amplification is performed³⁶. This allows making Eve's information of the final shared key extremely small. In privacy amplification, Alice and Bob apply hash algorithms that uses the error free shared key as an input and outputs a new shorter key.

Bibliography

¹ D. A. Neamen. Semiconductor physics and devices, basic principles. Third edition. McGraw Hill, ISBN 0-07-232107-5 (2003)

² F. Bloch, Z. Physik. Uber die Quantenmechanik der Elektronen in Kristallgittern 52, 555-600 (1928)

³ R. Kronig, W. G. Penney. Quantum Mechanics of Electronics in crystal lattices. Proc. Roy. Soc., A130, 499-513 (1931)

⁴ P. Michler. Single quantum dots, fundamentals, applications and new concepts. Springer, ISBN 3-540-140220-0 (2003)

⁵ N. Gisin, G. Ribordy, W. Tittel, H. Zbinden. Quantum Cryptography. Rev. Mod. Phys., 74, 145-195 (2002)

⁶ C. B. Murray, C. R. Kagan, M. G. Bawendi. Synthesis and characterization of monodisperse nanocrystals and close-packed nanocrystal assemblies. Annual Review of Materials Research, 30 (1), 545-610 (2000)

⁷ Y. Makhlin, G. Schön, A. Shnirman. Quantum-state engineering with Josephson-junction devices. Rev. Mod. Phys., 73, 357 (2001)

⁸ Z. M. Wang. Self-assembled quantum dots. Springer, ISBN 978-0-387-74190-1 (2008)

⁹ C. Chakraborty, L. Kinnischtzke, K. M. Goodfellow, R. Beams, A. N. Vamivakas. Voltage-controlled quantum light from an atomically thin semiconductor. Nature Nanotechnol. 10, 507-512 (2015)

¹⁰ P. Frigeri, L. Seravalli, G. Trevisi, S. Franchi. Molecular beam epitaxy: an overview. Comprehensive Semiconductor Science and Technology, 3, 480-522 (2011)

¹¹ R. C. Jaeger. Introduction to microelectronic fabrication. Upper Saddle River: Prentice Hall, volume 5, ISBN 0-201-44494-1

¹² I. N. Stranski, L. Krastanow. Zur Theorie der orientierten Ausscheidung von Ionenkristallen aufeinander. Abhandlungen der Mathematisch-Naturwissenschaftlichen Klasse IIb. Akademie der Wissenschaften Wien, 146, 797-810 (1938)

¹³ F. Hatami, N. N. Ledentsov, M. Grundmann, J. Böhrer, F. Heinrichsdorff, M. Beer, D. Bimberg, S. S. Ruvimov, P. Werner, U. Gösele, J. Heydenreich, U. Richter, S. V. Ivanov, B. Y. Meltser, P. S. Kop'ev, Zh. I. Alferov. Radiative recombination in type-II GaSb/GaAs quantum dots. Appl. Phys. Lett., 67, 656-658 (1995)

¹⁴ J. Y. Marzin, J. M. GéRard, A. Izraél, D. Barrier, G. Bastard. Photoluminescence of single InAs quantum dots obtained by self-organised growth on GaAs. Phys. Rev. Lett., 73, 716-719 (1994)

¹⁵ O. Briot, B. Maleyre, S. Ruffenach. Indium nitride quantum dots grown by metalorganic vapour phase epitaxy. Appl. Phys. Lett., 83, 2919-2921 (2003)

¹⁶ Z. Lin, B. R. Carvalho, E. Kahn, R. Lv, R. Rao, H. Terrones, M. A. Pimenta, M. Terrones. Defect engineering of two-dimensional transition metal dichalcogenides. IOP 2D Matter, 3, 022002 (2016)

¹⁷ J. D. Joannopoulos, S. G. Johnson, J. N. Winn, R. D. Meade. Photonic crystals: modelling the flow of light, Second edition. Princeton University Press, ISBN 978-0-691-12456-8 (2011)

¹⁸ S. G. Johnson, S. Fan, P. R. Villeneuve, J. D. Joannopoulos. Guided modes in photonic crystal slabs. Phys. Rev. B: Condens. Matter. Phys., 60, 5751-5758 (1999)

¹⁹ J. A. Stratton. Electromagnetic theory (1st ed.). New York: McGraw-Hill. ISBN 978-0-470-13153-4 (1941)

²⁰ T. F. Krauss, R. M. De La Rue, S. Brand. Two-dimensional photonic-bandgap structures operating at near-infrared wavelengths. Nature, 383, 699-702 (1996)

²¹ Y. A. Vlasov, M. O'Boyle, H. F. Hamann, S. J. McNab. Active control of slow light on a chip with photonic crystal waveguide. Nature, 438, 65-69 (2005)

²² T. Baba. Slow light in photonic crystals. Nature, 2, 465-473 (2008)

²³ M. Notomi, K. Yamada, A. Shinya, J. Takahashi, C. Takahashi, I. Yokohama. Extremely large group-velocity dispersion of line-defect waveguides in photonic crystal slabs. Phys. Rev. Lett., 87, 253902 (2001)

²⁴ A. Javadi, I. Sollner, M. Arcari, S. Lindskov Hansen, L. Midolo, S. Mahmoodian, G. Kirsanske, T. Pregnolato, E. H. Lee, J. D. Song, S. Stobbe, P. Lodahl. Single-photon non-linear optics with a quantum dot in a waveguide. Nature Commun., 6, 8655 (2015)

²⁵ P. Lodahl, S. Mahmoodian, S. Stobbe. Interfacing single photons and single quantum dots with photonic nanostructures. Rev. Mod. Phys., 87, 347-400 (2015)

²⁶ E. M. Purcell. Spontaneous emission probabilities at radio frequencies. Phys. Rev., 69, 674 (1946)

²⁷ J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, A. Forchel. Strong coupling in a single quantum dot semiconductor microcavity system. Nature, 432, 197-200 (2004)

²⁸ V. Scarani, H. Bechmann-Pasquinucci, N. J. Cerf, M. Dusek, N. Lutkenhaus, M. Peev. The security of practical key distribution. Rev. Mod. Phys., 81, 1301-1350 (2009)

²⁹ M. D. Eisaman, J. Fan, A. Migdall, S. V. Polyakov. Invited review article: Single-photon sources and detectors. Rev. Sci. Instrum., 82, 071101 (2011)

³⁰ L. C. Comandar, B. Frohlich, M. Lucamarini, K. A. Patel, A. W. Sharpe, J. F. Dynes, Z. L. Yuan, R. V. Penty, A. J. Shields. Room temperature single-photon detectors for high bit rate quantum key distribution. Appl. Phys. Lett., 104, 021101 (2014)

³¹ Robert H Hadfield. Single-photon detectors for optical quantum information applications. Nature Photon., 3, 696-705 (2009)

³² F. Marsili, V. B. Verma, J. A. Stern, S. Harrington, A. E. Lita, T. Gerrits, I. Vayshenker, B. Baek, M. D. Shaw, R. P. Mirin, S. W. Nam. Detecting single infrared photons with 93% system efficiency. Nature Photon., 7, 210-216 (2013)

³³ C. M. Natarajan, M. G. Tanner, R. H. Hadfield. Superconducting nanowire single-photon detectors: physics and applications. Supercond. Sci. Technol., 25, 063001 (2012)

³⁴ W. T. Tsang. Semiconductors and semimetals. Part D photodetectors. Academic Press, ISBN 978-0-12-752153-4 (1985)

³⁵ K. Inoue, E. Waks, Y. Yamamoto. Differential Phase shift quantum key distribution. Phys. Rev. Lett., 89, 037902 (2002)

³⁶ C. H. Bennett, G. Brassard, C. Crepeau, U. Maurer. Generalized privacy amplification. IEEE Trans. Inf. Theory, 41, 1915 (1995).

Chapter 3

Device Fabrication and Experimental Techniques

3.1. Introduction

Many tools and experimental techniques were used for designing, simulating, growing, fabricating and testing the devices discussed in this thesis. Designing and simulating the photonic components for quantum key distribution (QKD) were done using MIT's Electromagnetic Equation Propagation (MEEP), which is a Finite Difference Time Domain (FDTD) tool written by Oskooi et al¹. Growth of the quantum dots (QDs) and nanowires was done using molecular beam epitaxy (MBE). Fabrication of the devices was achieved using electron-beam (e-beam) lithography, thermal evaporation and inductively-coupled plasma reactive ion etching (ICP-RIE). Fabrication inspections of the devices were done using atomic force microscopy (AFM) and scanning electron microscopy (SEM). Micro-photoluminescence spectroscopy was carried out using an in-house built setup to perform optical characterisation of the devices. In this chapter of the thesis, a summary of these tools and techniques will be given. The following sections will summarise and discuss the fabrication recipes and the challenges faced to realise the hole-type photonic crystals (PhCs).

Some of the novel aspects in this chapter involve the use of CSAR62, a resist used as an alternative to PMMA for e-beam lithography. A comparison will be made between using CSAR62 and PMMA as etching masks in ICP-RIE dry etching. Selectivity results with etching GaAs using chlorine based gases

will be given. Other novelties in this chapter include performing high temperature dry etching to achieve high aspect ratios in etching hole-type PhCs. A comparison will be made between the achievable etching profiles as the table temperature of the ICP-RIE is increased.

3.2. Fabrication Techniques

3.2.1. Electron-Beam Lithography

Semiconductor device fabrication involves many processes and techniques used to create integrated electronic and photonic circuits for use in a wide range of applications. Device fabrication usually involves a series of steps such as lithography, chemical processing and physical etching etc. before achieving the final desired structure on a semiconductor material.

In nanofabrication, lithography is a technique used in patterning an integrated circuit structure on a thin film, such as a resist, or the bulk substrate. The most common type of lithography techniques are photolithography² and e-beam $(e-beam)^3$ lithography. There are other methods for performing lithography such as nanoimprint lithography⁴, interference lithography⁵, X-ray lithography⁶, magneto lithography⁷ and scanning probe lithography⁸. The photonic structures investigated in this thesis requires nanometre resolution lithography techniques. This high-resolution requirement is particularly important in fabricating PhCs operating in the visible/near-infrared wavelength range. The challenge in fabricating PhCs lies in their requirement to be precisely fabricated as needed in the design and simulation process to ensure they keep their optical properties and prevent photonic losses. This makes e-beam lithography that typically has sub-nanometre resolution, the ideal tool for fabricating PhCs. The disadvantage of using e-beam lithography is its slow throughput speed, where multiple devices can only be written in a serial manner. For this reason, it is not generally suitable for the mass manufacturing of devices as the processing time scales linearly with the number of devices being produced, whilst other parallel techniques can scale sub-linearly. A potential technique that can be utilised in the future for PhC fabrication is nanoimprint lithography, which can achieve a resolution lower than 10 nm, thus suitable enough for visible wavelengths PhCs. Previous reports have shown PhC structures fabricated with scanning probe lithography. Since most of the devices presented in this thesis are fabricated with e-beam lithography, the focus of this section will be on introducing e-beam lithography.

E-beam lithography is the process of using a beam of electrons to write a pattern on an e-beam sensitive resist. The resist layer is normally span coated on a substrate, and by exposing and scanning the resist with an e-beam, using an e-beam writer, the desired pattern can be achieved. Long molecular chains that the resist consists of break after the application of the e-beam making the exposed areas on the resist dissolvable in a suitable chemical solution. The high achievable resolution with e-beam lithography is due to the sub-nanometre size de Broglie wavelength of electrons, which is much shorter than UV light wavelengths, that is typically used in photolithography. The e-beam writer used in fabricating the devices presented in this thesis is a JEOL-JBX-5500ZD. A schematic illustration of a typical e-beam writer is shown in Figure 3.1.



Figure 3.1: A simplified schematic illustration of a typical e-beam writer, similar to that of a scanning electron microscope. The figure shows the e-beam path as it travels through the anode, magnetic lens, scanning coils before exposing the sample. Secondary electrons emitted by the sample and backscattered electron are detected by different detectors for imaging and calibration.

In the JEOL system, an e-beam is generated from a cold field emission electron gun, and the e-beam is attracted by an anode that is normally at a potential of 50 kV. The gun is made of a crystalline tungsten tip that is very sharp, typically less than 0.1 μ m at its end, hence creating a strong electric field at the tip. The beam is then passed through a series of electromagnetic condensing lenses that reduce the diameter of the beam. The beam is then passed through a beam limiting aperture, where the aperture size used in the JEOL system is normally 40 μ m. Finally, the beam diameter is focused on the sample by an objective lens where the diameter size of the beam typically reaches < 1 *nm*. During the writing and alignment process, the scanning coils are used to scan the beam on the sample and pattern the desired structure. The backscattered and secondary electrons detectors are used to image the surface by detecting electrons scattered and those emitted by the sample, respectively.

The system is typically operated under high vacuum to prevent electron scattering and losses as they travel through the column. Unless stated otherwise, most devices are patterned using a 50 pA beam current, however the base dosage together with the modulation were altered. The high acceleration voltage of the system provides the necessary resolution for fabricating nanostructures. This is due to the reduced angle of backscattering, as shown in Figure 3.2. For PhC devices that have dense structures and require precise writing patterns, a dosage modulation tool, known as Beamer, is utilised to reduce shape changes in the written pattern due to forward and backscattered electrons. This is also known as Proximity Effect Correction (PEC).

In forward scattering, an accelerated electron from the beam collides with another electron from one of the atoms within the substrate/resist. The incident electron will change its direction of travel while transferring parts of its energy to the atom which becomes ionised, creating a secondary electron. In backscattering, an electron collides with a much heavier nucleus from a resist/substrate, causing it to elastically scatter. The scattering angle due to backscattering is usually much larger than that for forward scattering. Some electrons return back through the resist at a significant distance from the incident beam point. This causes enlargement in the exposed regions of the resist as shown in Figure 3.3 (a and b). Hence, PECs are utilised to reduce the enlargement caused by backscattered electrons.



Figure 3.2: A schematic illustration showing a comparison between the spread of a beam accelerated by a 20kV and a 50kV acceleration voltage as it penetrates through a substrate. Electrons with higher accelerating voltages can penetrate to greater depths due to their high velocity, reducing backscattering angles.

In brief, there are mainly two methods to perform PEC, the first is shape modification and the second is dosage modification. Performing the PEC correction often needs the accelerating voltage of the used e-beam writer and the materials of the substrate, resist and developer as inputs. In shape modification, a single dosage is used for the entire structure. The software uses an algorithm to modify the patterned structures whilst taking into account the base dosage applied and other variables to achieve a closer approximation to the desired structure, Figure 3.3 (c and d). In dose modification, the pattern is divided into segments or pixels and the software uses an algorithm to calculate the dosage needed for every pixel in the mask, while taking into account the dosage applied to nearby pixels. This can be done by calculating the convolution of multiple Gaussian functions from exposing every pixel. The disadvantage of the dosage in the EBL system to account for the multi-dosage layers that the mask typically composes of. However, compared to the shape modification method, closer matching between the desired and the fabricated structures can be made using the dosage modification technique. On the other hand, the disadvantage of the shape modification method is its incompetence in handling masks that

involve large and small structures close to each other, since modification of the shape may lead to an overlap between the different structures. A hybrid method of dosage and shape modification can also be used for PECs. For example, to fabricate the desired structure shown in Figure 3.3 (e), the exposed area is reduced and the structures is divided into regions of different dosage levels. Dividing the structures into many dosage levels (typically tens to a few hundreds) with small increments between each level allows realising the desired structure with good geometrical accuracy, as shown in Figure 3.3 (f). Considering the rectangle shown in Figure 3.3 it is important to notice that the rectangle is exposed with low dosages near the centre, the dosage is increased though toward the edges of the rectangle. This is because the average dosage per area accumulated near the centre will be higher compared to those near the edges when a single dosage was used for the whole rectangle.



Figure 3.3: An illustrative diagram of proximity effects on the achieved pattern. (a and b) An illustration of the enlargement occurs as the pattern is exposed with one constant dosage. (c and d) The enhancement obtained in the developed shape size, as the shape of the exposed region is changed. (e and f) Even better enhancement obtained as the pattern is shrunk and divided into regions of different dosages, where outer regions are exposed to higher dosage compared to inner regions, hence maintaining the same average dosage throughout the pattern.

3.2.2. Thermal (Resistive) Evaporation

Resistive evaporation⁹ is a popular technique in the semiconductor microfabrication industry used to accurately deposit nanoscale films of metals or other materials on a substrate. The metal is either evaporated by applying an electric current through a filament crucible or sublimated off a tungsten wire. A schematic of a resistive evaporator system is shown in Figure 3.4. This method should not be confused with Molecular Beam Epitaxy which can also be classified as a form of thermal evaporation, MBE systems are not included in this discussion (see section 3.2.4).

Inside the chamber, an oscillating crystal is fitted to measure the deposition rate, which can be used to calculate the achieved thickness¹⁰. The working principle behind the oscillator is that the oscillation frequency is changed as the mass of the oscillator is increased when the material is deposited on it. An electronic instrument continuously reads the resonance frequency of the oscillator and applies appropriate algorithms to convert the frequency data to deposition rates in real time.

There are many other methods that can be used for thin film metal deposition such as e-beam evaporation and plasma sputtering⁹. The advantage that resistive evaporation has over other methods is its simplicity, cheap system maintenance and compatibility with most metals. Thermal evaporation of Chromium for its use as a hard etching mask in fabricating the rod-type PhC is discussed in chapter 4.

3.2.3. Lift-Off

Lift-off is a process used widely in the microfabrication industry for transferring a pattern of a specific material, usually metals, on a substrate using a sacrificial material, such as a resist¹¹. The desired pattern is first patterned on a sacrificial layer, where areas of the sacrificial layer are removed exposing the underlying substrate to be deposited on, see Figure 3.5 (a). A metal layer is then deposited to cover the exposed regions of the substrate and cover remaining regions of the sacrificial layer, as shown in Figure 3.5 (b). The lift-off process involves inserting the whole substrate in a solution that dissolves the sacrificial layer and removes it together with the capping metal layer, as shown in Figure 3.5 (c and d). It is necessary that the lift-off solvent used is not reactive with the metal deposited but only dissolves the sacrificial layer material.



Figure 3.4: A schematic illustration of a thermal evaporator system. The chromium is coated around a tungsten filament and is sublimated when a current is applied across the tungsten filament. Heating of the tungsten filament is made by a variable current source. The deposition rate is measured by a crystal oscillator connected to an amplifier and calibration electronics.



Figure 3.5: A schematic illustration of the lift off process. (a) First the inverse pattern is transferred on the substrate using a resist (b) A metal (such as Chromium) is deposited, covering the exposed regions of the substrate and the regions capped with a resist. (b) The whole sample is placed in lift-off solvent such as acetone, which dissolves the resist and removes the metal layer on top of them, leaving only regions where the metal is stuck to the substrate unaffected.

It will be shown later in chapter 4 that a lift-off process is used in fabricating rod-type PhCs utilising hard metal mask to provides better etching selectivity.

3.2.4. Molecular Beam Epitaxy

Molecular beam epitaxy (MBE) is a common technique for thin film deposition of materials such as metals and semiconductors¹². The word epitaxy originates from ancient Greek, where epi means "above" and taxis means "ordered" or "arranged". Hence, the word epitaxy can be translated as "arranging upon" or "ordered above". Compared to Chemical Vapour Deposition (CVD)13, MBE systems do not use organic chemical precursors which normally increase background levels of deposited carbon. In addition, the ultra-high vacuum that MBE operates at allows reducing other environmental impurities within the chamber. Furthermore, MBE can provide very accurate composition of the monolayer deposition and doping control. However, the complexity, price and maintenance costs of MBEs make them less favourable for industrial processing in comparison to CVD methods. In an MBE system, elements such as Gallium and Arsenic are heated in effusion cells to evaporation temperatures and are allowed to condense on the wafer. Computerised shutters are placed between the effusion cells and the wafer holder to allow precise control of the thickness of the deposited material. The temperatures of the effusion cells are accurately controlled to regulate the deposition rate. On the other hand, controlling the wafer's temperature allows controlling the rate of hopping or desorption of the deposited material¹⁴. To monitor the growth of the crystal layers on the wafer, reflection high energy electron diffraction (RHEED) is used. Growing the GaSb QDs and the different nanowires discussed in chapter 5 was carried out using a VG V80h MBE system.

3.2.5. Etching

Plasma dry etching is an essential tool used in the semiconductor fabrication industry. Plasma dry etching together with wet etching were heavily used in fabricating most of the devices in this thesis. In this section, a brief introduction to the concept of etching will be made together with overviews of dry and wet etching¹⁵. In the section that follows, two very common techniques for dry etching will be explained, these are Reactive Ion Etching (RIE)¹⁶ and Inductively Coupled Plasma (ICP)¹⁷.

In micro and nano-fabrication, etching is the process of removing layers of a wafer to achieve a certain structure. Before carrying out the etching process, some regions of the wafer are usually covered with a mask to protect the underlying substrate against etching, whilst the uncovered areas are exposed to the etchant material and etched away. The mask is usually defined by a lithography process; where the mask may be the resist used in the lithography. There are two types of etching employed in the semiconductor industry, these are wet and dry etching.

In wet etching, liquid chemical etchants are used to remove material from a wafer. The liquid is chosen so that it reacts with the substrate and create other products that are usually dissolved and carried away by the etchant solution. The substrate is normally rinsed in a different solution to stop the chemical reaction by diluting any remaining etchant, and removing any by-products created as a result of the reaction. The advantage that wet etching has is that it is simple and the etchants needed are normally cheap. However, achieving anisotropic etching profiles with wet etching is usually to do. This makes wet etching unsuitable in fabricating structures that need high etching verticality, as is the case for PhCs. In chapter 5, hydrofluoric acid wet etching will be used to etch SiO₂ for creating the nanowire sites for their deterministic growth. Wet etching will also be used for releasing the suspended PhC bridge structures in chapter 6.

In dry etching, a plasma is utilised together with etchant gasses to remove unmasked areas of a substrate. There are two primary mechanisms that take place when dry etching a substrate, the first is chemical etching and the second is physical etching (sometimes known as mechanical etching). While this may sound simplistic, careful examination of the used recipe in dry etching to select between these two types of etching mechanisms is necessary for optimised control over the etch. This is especially important in etching fine features such as those in PhCs. In chemical etching, etchant gases react with the substrate causing other materials to form. The by-products that result from the chemical etching process are removed by the vacuum pump. In physical dry etching, high kinetic energy ions, electrons or photons are used to etch the substrate. The high-energy particles bombard the substrate causing top-down

sputtering of the atomic layers from the substrate. In chapter 4, dry etching will be used to create the rods for the rod-type PhCs and will be used later in chapter 6 to etch the holes in the hole-type PhCs.

To create and sustain the plasma required for dry etching, energy is supplied by coupling an external electric field and/or magnetic field to ionise the etchant gas and supply the ions with enough kinetic energy to perform the etching. In the following sections, it will be shown that the different methods of coupling the gases to different energy sources can provide control over the plasma. Compared to wet etching, dry etching can provide a good control over the etch profile by adjusting parameters such as the chamber pressure, the etchant gases' flow rates, the etching time and the applied energy that ionises and drives the ionised gas.

3.2.6. RIE and RIE-ICP

RIE is a technology utilised in the semiconductor micro-fabrication industry for dry etching¹⁵⁻¹⁷. RIE uses a plasma to remove materials from a wafer substrate by chemical and physical means under low pressures. Compared to RIE-ICP, RIE typically uses a parallel plate capacitor configuration (the configuration is sometimes known as Capacitively Coupled plasma) to create an external electric field for creating and sustaining the plasma inside the chamber. The wafer is normally situated in the bottom of the chamber facing the plasma as shown in

. The RIE system used in this work is an Oxford Instrument Plasma Pro NGP80 supplied with Ar, O_2 , SF_6 , CHF_4 gases.

The electric field is applied between the capacitor plates using an RF generator operating at an industrial frequency of 13.56 MHz, where the source power can range from a few watts to several hundred watts. The oscillating electric field from the capacitor plates ionises the gas molecules by knocking out their electrons. These electrons are accelerated within the oscillating plates knocking out other electrons to further ionise the gas atoms, where those oscillate hitting the chamber walls and the lower electrode. The chamber walls are grounded, hence electrons hitting the walls are removed. However, the accumulation of electron charge on the bottom electrode builds up a potential difference that can be measured by voltage measuring instruments. The potential difference between the lower electrode and



Figure 3.6: The internal configuration of an RIE system. The figure shows the top and lower parallel plates electrodes responsible for igniting and sustaining the plasma, together with the position of the wafer with respect to the arrangement. The chamber is pumped and maintained at a desired pressure by a variable pressure valve. Different gasses lines are connected to the gas inlet with mass flow controllers to control their flow.

the top is known as DC bias, and it is an important parameter that needs to be controlled for optimised control over the etching. Ions that are much heavier than electrons are attracted toward the wafer electrode due to the potential difference and the negative voltage the wafer has. The DC bias can be as little as a few volts and as high as several hundred volts. The drift of ions toward the wafer can make the ions react chemically with the wafer to etch it (discussed previously as chemical etching) or physically sputter the wafer due to the ions high kinetic energy (discussed previously as physical etching).



Figure 3.7: A schematic illustration of an ICP-RIE system, showing the addition of the ICP coils in comparison with the RIE system shown in

An ICP-RIE system is similar in structure to an RIE system except for the addition of an N-turn magnetic coil around the plasma discharge region. The inductor is connected to an RF generator and delivers the energy to the gas via a time varying magnetic field inside the chamber. A simplified ICP-RIE configuration is shown in Figure 3.7. The separate control over the inductively and capacitively supplied energies to the plasma allows one to control the plasma density and other plasma parameters without significantly changing the energy of the ions that bombard the sample. In other words, the ICP can control the number of ions reaching the substrate for chemical etching while the CCP can control the momentum of the ions that bombard the substrate. The ICP-RIE system used in this work is an Oxford Instruments Plasmalab System 100 supplied with N₂, H₂, BCl₃, Cl₂, Ar, SF₆, O₂ gases. In the following sections, discussions and analysis will be made of the effect of changing the etching parameters on the achieved etching results.

3.3. Scanning Probe Microscopy

Most of the fabricated features in this thesis required e-beam lithography due to the high resolution that it provides compared to photolithography. Normally, visual and topological investigation of the resultant developed patterns on an e-beam resist is essential to assist with identifying any source of fabrication issues. This was especially important with using PMMA in fabricating the hole-type PhCs, due to incomplete developing of the small and dense patterns that were exposed with fairly low dosages. Using SEM for this purpose will expose the e-beam sensitive resist and cause further exposure to the resist. This makes any further development to the pattern, if necessary, useless. Further exposure to electrons may also cause cross linking in the resist, making it harder to strip off in later stages of the device fabrication. For this reason, scanning probe microscopy (SPM) was frequently utilised in this work to characterise the topographic structure of the resist after exposure.

SPM is a large branch of microscopy that utilises sharp probes to investigate a sample's topological, mechanical and electrical characteristics. A simplified illustrative figure for a scanning probe microscope is shown in Figure 3.8. A sharp probe is mounted on a cantilever and allowed to contact the sample by mounting the sample on an x-y-z piezo stage and moving it upwards until contact between the tip and the sample is achieved. An applied external voltage across the piezo stage allows the sample to be scanned beneath the probe to investigate the sample's surface. A laser is calibrated to reflect from the cantilever toward the centre of a four-quadrant photodiode. When the surface of a sample is scanned, the probe is deflected based on the topography of the sample, causing the reflected laser spot to shift from the centre of the photodiode. This shift is registered with time and a correlation between the topography and probe position is recorded to output a topography map. The probe used in this work is an Al coated Si tip with a pyramid height ranging from 4-6 μ m, whilst the tip's diameter is 12 ± 4 nm. This aspect ratio is usually sufficient enough to allow the probe to reach the bottom of a fabricated PhC hole with a radii of 55 nm and a depth of 120 nm. The AFM systems used in this work was either a Veeco model NanoMan VS or a Veeco AFM CP-II.



Figure 3.8: A simplified schematic diagram of the working principle of a scanning probe microscope. When the tip is scanned laterally across the sample's surface, deflections in the cantilever caused by the sample's topology are measured by a quadrant photodetector. The sample is mounted on a piezoelectrically driven stage to allow it to move during the scan process.

There were two imaging modes used to inspect the fabrication of the devices discussed in this thesis, namely contact and tapping modes. In the contact mode, the tip approaches the sample and makes contact throughout the whole scanning process. As the sample is moved during the scanning process, the signal recorded at the photodetector is transferred to the control electronics where a feedback loop allows the height of the sample to change in order to keep the force on the cantilever constant. The change in the sample stage's height is recorded to output the sample's topography. The tapping method, on the other hand, operates in a different way. In this method, the cantilever is oscillated at the cantilever mechanical resonance frequency. Hence, the tip makes contact with the sample for very short periods of time that depend on the resonance frequency, this is in contrast to the contact mode where the tip is constantly in contact with the sample. In tapping mode, when the topography underneath the oscillating tip is changed, the tip oscillation amplitude changes, hence the sample-tip separation is varied using a feedback loop to maintain a constant oscillation amplitude of the tip. The topography map is then inferred from sample-tip separation. Tapping mode was found to produce better images when small features such as the PhC holes from Figure 3.11 are imaged. It was found that using contact mode, the

tip struggles to reach the bottom of the exposed holes due to the low aspect ratio of the pyramidal shaped tips.

3.4. Fabrication of Hole-Type Photonic Crystals

In this section, the fabrication methods and the challenges faced in fabricating the hole-type PhC cavities discussed in chapter 6 will be explained. This includes the problems faced with developing PMMA and the methods in which they were solved using CSAR62. A summary of the best dry etching recipes that give the most anisotropic sidewalls for the holes will be given together with the effect of different parameters that affect the etching profile. The steps for fabricating the structure are summarised in Figure 3.9.

Initial attempts at fabricating the PhC were done using PMMA as an e-beam resist¹⁸. The sample was spin-coated with a 200 nm PMMA layer and baked at 180 °C. Following this, the samples are patterned with a 50 pA e-beam at 240 μ C/cm², which was found to be about the average dosage needed for PMMA on a GaAs substrate using a 50kV writer. Using a different substrate such as Si, the dosage is usually increased to account for the lower backscattering of electrons from the lighter atoms of Si compared to GaAs. During the development process, two different solutions were used in removing the exposed regions of the sample, these were Isopropanol (IPA):Deionized water (DI) 7:3 and Methyl Isobutyl Ketone (MIBK):IPA 1:3. Both water and IPA are non-solvents for PMMA at room temperature. In comparison to highly polar and the heavy molecular weight of MIBK, the less polar and lighter IPA/water mixture can provide much better sensitivity because it can disintegrate and washoff the small fractions of molecules that have been exposed by the e-beam.

However, there were two main issues faced when using PMMA as an e-beam resist for fabricating the holes. In all fabrication attempts it was found that the PMMA resist was not developed deep enough to the point where the underlying substrate was exposed. This was still the issue even when either of the two developers (IPA:DI and MIBK:IPA) were used for long developing time periods. The developing depth of the PMMA layer could have only been increased when the dosage was increased. However,



Figure 3.9: A schematic illustration showing for the steps of fabricating the hole-type PhC. (a) The resist is spun on the substrate before the pattern was written with an e-beam writer and developed as shown in (b). (c) ICP-RIE dry etching is performed to pattern the hole structure on the top GaAs layer. (d) Wet etching with HF is used to suspend the PhC structure by removing the underlying AlGaAs layer.

for the purpose of fabricating the PhC this had the effect of increasing the holes' radii due to increased backscattering of electrons from the substrate, which is expected to reduce the cavities Q-factor and change their resonance wavelength.

An alternative resist that was used to replace PMMA is CSAR62¹⁹. Similar to PMMA, this is a positive e-beam resist that provides very high sensitivity and good contrast that is suitable for fabricating features that require sub 10 nm resolution. The advantage that the cheap CSAR62 provides over PMMA is its higher selectivity in dry etching, making it a strong competitor of ZEP520. The increased selectivity in etching CSAR62 with GaAs makes the needed resist thickness that is required to mask the substrate much smaller. This allows making holes with radii closer to those desired, which is aided by the thinner resist typically having less faceting after development. In addition, due to the higher sensitivity of CSAR62 compared to PMMA, a much smaller dosage is required to expose the pattern needed, this reduces the increase that is expected to happen to the rod's radii due to backscattering effects.

The incomplete developing issue that was faced with PMMA was easily solved using CSAR62. The patterns are exposed with $110 \,\mu\text{C/cm}^2$ and developed using o-xylene. AFM measurements showed that the exposed regions are removed completely after around 2 minutes of developing. It is worth mentioning that whilst developing, sonication was done at a 50 % power (of 250 W maximum power and 32-38 KHz average operating frequency) at room temperature, using a Grant ultrasonic water bath of model XUB5 to aid the developing process, but it is not expected to be vital with CSAR62.

It was always planned that a resist would be more suitable to operate as a mask for fabricating the holetype PhC. This is because the hole-type PhCs are much smaller and are more prone to fabrication imperfections in comparison to the rod-type PhCs. Using a hard mask for lift-off like that for rod-type PhCs creates a large room for error, such as a change in the holes radius and a reduction in the circularity of the achieved holes after the pattern is transferred. It is also expected to increase the hole sidewall roughness after etching, reducing the quality of the photonic structure.

After developing and inspecting the pattern, dry etching using the ICP-RIE method is performed. The gases introduced in the chamber are BCl₃ with a flow rate of 2.5 standard cubic centimetres per minute (sccm) and Ar with a flow rate of 7.0 sccm. The reason behind using Ar in the recipe was to promote physical etching rather than chemical etching which is expected to cause undercutting shapes. The RF power which represents the power introduced in the parallel plates is 70 W and the ICP power was 100 W. The chamber pressure was kept at the minimum achievable pressure by the pump which was roughly around 2.1 mTorr. The wafer temperature was approximately 13 °C. In this recipe, it was found that the etch rate was approximately 104 ± 12 nm/min. The GaAs:PMMA etching selectivity using this recipe is approximately $1.1:1 (\pm 0.12)$. This was measured by etching larger patterns exposed on the PMMA resist, as performing this measurement with hole-type PhCs patterns exposed on PMMA was impossible due to the incomplete developing issue discussed previously. This selectivity is expected to be less in etching PhC holes. The reason is because in etching small features, it is harder to extract ions used for etching that tend to have lower kinetic energy as they get trapped inside the small patterns in the resist, leading to slower etching rates, as is the case in PhC holes. On the other hand, the selectivity obtained

in etching GaAs:CSAR62 was 1.9:1 (\pm 0.12). This greatly reduces the required thickness needed for the resist to etch the holes completely through the GaAs layer until the AlGaAs layer is reached.

At this point, the current problem with fabricating the PhC was the large faceting obtained in the sidewall profiles of the etched holes as shown in Figure 3.10 (a and b). The faceting can also be observed from the top view image of the holes which show the dim surface inside the holes that are cause by reflections of the electrons from the facets. The second problem which needed to be addressed was the relatively slow etch rate obtained for etching the holes compared to etching larger features such as the out-couplers. To solve these problems, the table temperature, and hence the wafer's temperature, was increased from 13 °C to 50 °C, whilst keeping the other etching parameters unchanged. This made the etch rate of the holes comparable to that of the large features such as the out-coupler rings, while the sidewalls profile shows better verticality. Both changes are attributed to an increase in the chemical etching rate which sped up the etching process and helped undercutting the sidewalls, hence reducing the facets. It was important to continue reducing the facets further to achieve holes with vertical sidewalls. This was done by further increasing the table temperature to 100 °C while keeping other etching parameters the same; the fabricated holes are shown in Figure 3.11. Clearly, from the top view image of the holes there is a great reduction in the facets, identified by the sharply shaped circles in comparison with those from Figure 3.10. The tilted image can also show the shadow of the side wall profiles of the holes closest to the out-coupler rings. However, it is expected that this recipe cannot be used for etching much deeper holes while maintaining the sidewalls vertical profile. This is because the higher temperature provided an increased chemical etching rate in comparison with physical etching, thus increasing the etching time for deeper sidewalls will likely form bow shapes due to increased undercutting which is normally associated with chemical etching.

Beside the improvement in the etch rate of small features and the enhanced anisotropy in the etching profile, etching at higher temperatures was found to improve the etching selectivity. At 100 °C, this was measured to be $3.6:1 (\pm 0.12)$ for GaAs:CSAR62. This was firstly noticed when the colour of the

remaining resist after etching was found to be blue-green compared to violet when the temperature was 13 °C. The selectivity was then measured more accurately using a surface profiler.

To summarise, the etching of GaAs was best done using CSAR62 as an etching mask. Etching GaAs was done using the recipe: $BaCl_3$ (2.5 sccm), Ar (7.0 sccm), RF(CCP power) = 70 W, ICP power= 100 W, P = 2.1 mTorr, T = 100 °C. Etching at high temperature was vital for improved etching rate of small features, for an improved sidewall profile and for increased selectivity.



Figure 3.10: (a and b) top and tilted views of the etching profile obtained from etching the sample at 13 °C temperature showing the large resultant etching facets. (c and d) Top and tilted views of the etching profiles at 50 °C showing the improved etch rate of the small holes and the slightly reduced faceting due to increased chemical etching.



Figure 3.11: The etching results obtained from etching at 100°C showing much better sidewalls from the top (a and b) and tilted (c and d) view images showing optimised anisotropy of the etched sidewall profile in comparison to Figure 3.10.



Figure 3.12: (a) An optical microscope image showing the etched bright yellow region as an indicator of under etching compared to the darker unetched regions. The figure also shows the etched trenches fabricated to allow larger surface contact between the AlGaAs layer and the HF solution. (b) An optical microscope image of a group of PhC bridge structures, all had successful wet etching without any bridge collapse.

After dry etching the holes to the required depth of the bridges, hydrofluoric acid (HF) wet etching was needed for removing the underlying Al_{0.8}Ga_{0.2}As layer (refer to Figure 3.9). It is expected that the AlGaAs etch rate scales with the Aluminium to Ga ratio, in this case the ratio 0.8:0.2 of Al:Ga was chosen from the literature²⁰. It is important to have large etched features surrounding the PhC structure to allow a higher surface area contact between the acid and the AlGaAs layer and allow better extraction of etched AlGaAs fragments. Here, the dry etched sample is inserted into a 0.8:0.2 HF:DI solution for 10 seconds and subsequently rinsed in DI for 10 seconds. This process is repeated three times, before finally rinsing the sample in IPA for 2 minutes²⁰. Dividing the etching process into multiple etch and rinse steps was found to help in extracting any remaining AlGaAs fragments from under the created bridge. Interestingly, there were very few cases where a created bridge would collapse after etching the underneath AlGaAs layer. This means that the PhC structure production yield is high and the structure is mechanically robust. However, it is worth mentioning that when performing sonication on the etched chip, even at very low powers, most of the created bridge on the chip collapsed and washed away. The wet etched structures were inspected under the microscope and successful wet etching would typically display bright yellow regions surrounding the etched structures due to the removed AlGaAs layer, see Figure 3.12.

3.5. Numerical Modelling With Finite Difference Time Domain

FDTD was the main tool used to simulate the photonic devices discussed in this thesis. These devices were mainly the PhC components and the out-couplers. The software packages used were mainly Meep¹ and Lumerical. It is beyond the scope of this thesis to dwell into the details of the theory and working principles of FDTD. However, a brief summary will be given next to give the necessary introduction for the very basics of FDTD as an electromagnetic wave simulation algorithm. For a comprehensive presentation of the method and all of its technical aspects, the reader is referred to references 22 and 23.

The FDTD simulation method is one of the most widely used numerical techniques for solving electromagnetic boundary value problems. FDTD is based on numerically solving Maxwell's equations in space and time, specifically those including the curl of E and H:

$$\mu \frac{\partial H}{\partial t} = -\nabla \times E \tag{3.1}$$

$$\varepsilon \frac{\partial E}{\partial t} = \nabla \times \boldsymbol{H} - \sigma \boldsymbol{E} \tag{3.2}$$

Where E and H are the Electric and Magnetic field vectors respectively, ε and μ are the electric permittivity and magnetic permeability of the dielectric material respectively. To illustrate how FDTD uses the above equations, two-dimensional simulations will be considered for simplicity. In two-dimensional simulations, light only propagates in the x-y plane, it is assumed that the simulated structure's geometry and the field are invariant in the z-direction, hence any partial derivatives with respect to the z-dimension are equal to zero. In this case, for six field components, Maxwell's equations in 2D involves two sets of independently coupled differential equations, each with only three field components:

$$-\frac{\partial E_z}{\partial y} = \mu_0 \frac{\partial H_x}{\partial t} \qquad -\frac{\partial H_z}{\partial y} = \varepsilon_0 \varepsilon_r \frac{\partial E_x}{\partial t} + \sigma E_x$$

$$-\frac{\partial E_z}{\partial x} = \mu_0 \frac{\partial H_y}{\partial t} \qquad -\frac{\partial H_z}{\partial y} = \varepsilon_0 \varepsilon_r \frac{\partial E_y}{\partial t} + \sigma E_y \qquad (3.3)$$

$$\frac{\partial H_y}{\partial x} - \frac{\partial H_x}{\partial y} = \varepsilon_0 \varepsilon_r \frac{\partial E_z}{\partial t} + \sigma E_z \qquad \frac{\partial E_x}{\partial y} - \frac{\partial E_y}{\partial x} = \mu_0 \frac{\partial H_z}{\partial t}$$

The first set which involves only magnetic components H_x , H_y in the x-y plane and E_z in the z direction is designated the TM mode. Likewise, the second set includes E_x and E_y in the x-y plane and H_z in the z-direction is designated the TE mode. The coupling between the field components in each of the two sets requires three different fields to be calculated for every mode.

The computational space is sampled into computational cells, where the cells are small enough in comparison to the considered wavelengths to minimise errors. The electric and magnetic field components for the TM and TE modes are calculated for every computational cell at successive time

steps, where the time steps are set small enough in comparison to the considered inverse frequencies to minimise errors. To perform the simulation, an electromagnetic wave source is added by increasing the field values at the desired position for the electromagnetic source, and the wave propagates by computing the fields for the other cells at successive times, this process is demonstrated in Figure 3.13.

There are two advantages of FDTD over frequency domain methods. Firstly, the computational power requirement for FDTD scales linearly with the size of the computational space without introducing any change in the simulation power needed as the simulated structure is changed. This makes the simulation power requirement for irregular structures similar to regular ones such as PhCs. Secondly, the method is simple to implement, even if there exist complicated scattering structures within the computation space, because each lattice cell can be assigned arbitrary dielectric values. This also permits simulating arbitrarily shaped, lossy, dispersive or anisotropic structures in the computation space.



Figure 3.13: Two-dimensional FDTD mesh showing the TM (a) mode and the TE (b) mode with magnetic (electric) field components labelled as H (E) distributed around the mesh. Arrows indicate in-plane components, while dots indicate out-of-plane components. Electromagnetic wave propagation and interaction is mapped into the simulation mesh by assigning appropriate values of permittivity to each electric field vectors and appropriate values of permeability to each magnetic field vector in the mesh.

By using transmission flux regions as presented in section 2.2.1, it was possible to calculate power spectra obtained from cavity and waveguide modes. The flux region is set up to integrate the Poynting vector of the electromagnetic field over the flux region for a frequency ω , these are then accumulated over *n*-time steps. The output spectra can be used to manually measure confinement modes and Q-

factors. An alternative method to extract modes from the calculated spectra and measure their Q-factors was using the harmonic inversion method that is embedded in the Meep package. For more details on this method, the reader is referred to reference 21.

FDTD simulations with Meep were done using a computer cluster, known as high end computing (HEC), and is operated by Linux. The HEC provided an abundance of memory and processing power for simulations to be carried out with time efficiency. In most simulations, about 16 GB of memory was used with up to 64 Intel cores clocked at 2.2 GHz. An example of a code used to simulate an optimised L3 hole-type PhC cavity can be found in Appendix A. On the other hand, Lumerical was operated on a windows workstation operated by an Intel Octa-core i7 processor with a Haswell microarchitecture. The advantage that Lumerical provides over Meep is the visual interface of the simulation setup. This is normally coded in Meep and required to be compiled before it can be visually inspected, consuming a considerable amount of time. However, being able to run Meep in a Linux operation system make it compatible with computer clusters, hence allowing simulations to run much faster when enough processing resources are available.

3.6. Micro-Photoluminescence Spectroscopy

In this section, the methods for obtaining the μ -photoluminescence measurements are explained. The measurement system was built in-house as shown in Figure 3.14. The excitation laser used in all the measurements was a continuous wave Nd:YAG frequency doubled 532 nm laser. The beam is first passed through neutral density (ND) filter wheels to control the incident power. The beam then passes through a beam sampler where some of the light is reflected and measured by a photodiode power meter. The laser is focused on the sample using a 50x objective lens with a numerical aperture of 0.65 such that the laser spot size is reduced to approximately 1 μ m². The objective is mounted on a piezo controlled xyz stage to allow fine tuning of the laser spot position. The emitted light is collected with the same objective lens and directed to an Andor Shamrock 500 spectrometer for spectroscopic analysis. The spectrometer uses a silicon electron-multiplying charge coupled device (EMCCD) for measurements between 500-900 nm and an InGaAs array for measurements in the wavelength range 800-1600 nm.

Both CCDs are Peltier cooled to -85 °C. For optical imaging purposes, the sample is illuminated with a red LED and the image is collected from the objective lens and output to a computer screen via a USB infrared camera.



Figure 3.14: The micro-photoluminescence setup used in taking the photoluminescence measurements in this thesis. The figure shows the optical path of the laser as it goes to the sample and is reflected to the spectrometer. The laser light is attenuated with neutral density (ND) filter wheels, a beam sampler is used to direct a portion of the light to a power meter, before reaching the sample. The laser is focused with an objective lens which also collects emitted light by the sample and directs it toward the spectrometer. The optical path of the illuminating red beam from the LED source to the digital camera that was used for optical imaging can also be seen in the figure.

Bibliography

¹ A. F. Oskooi, D. Roundy, M. Ibanescu, P. Bermel, J. D. Joannopoulos, S. G. Johnson. MEEP: A flexible free-software package for electromagnetic simulations by the FDTD method. Computer Physics Communications, 181, 687-702 (2010)

² M. J. Madou. Fundamentals of microfabrication: the science of miniturization, second edition. Lithography, ch.1. CRC press (2002)

³ M. A. Mohammad, M. Muhammad, S. K. Dew, M. Stepanova. Fundamentals of electron beam exposure and development. Nanofabrication, ch.2. Springer (2012)

⁴ S. Y. Chou, P. R. Krauss, P. J. Renstrom. Nanoimprint lithography. Journal of Vacuum Science & Technology B, Nanotechnology and Microelectronics: Materials, Processing and Phenomena, 14, 4129-4133 (1996)

⁵ V. Berger, O. Gauthier-Lafaye, E. Costard. Photonic band gaps and holography. Journal of Applied Physics, 82, 60-64 (1997)

⁶ A. Heuberger. X-ray lithography. Journal of Vacuum Science & Technology B, Nanotechnology and Microelectronics: Materials, Processing, Measurement, and Phenomena, 6, 107-121 (1988)

⁷ A. Bardea, N. Burshtein, Y. Rudich, T. Salame, C. Ziv, O. Yarden, R. Naaman. DNA and RNA molecule detection and identification using a patterned capillary tube (PCT). Ann. Chem., 83, 9418-9423 (2011)

⁸ R. Garcia, A. W. Knoll, E. Riedo. Advanced scanning probe lithography. Nature Nanotechnol., 9 577-587 (2014)

⁹ R. C. Jaeger. Introduction to microelectronic fabrication, second edition. Volume 5. Film deposition, ch.6. Upper Saddle River: Prentice Hall (2002)

¹⁰ K. H. Behrndt. Long-term operation of crystal oscillators in thin-film deposition. Journal of Vacuum Science and Technology, 8, 622-626 (1971)

¹¹ S. Franssila. Introduction to micro fabrication. Structures by deposition, ch.23. Wiley (2004)

¹² W. P. McCray. MBE deserves a place in the history books. Nature Nanotechnol., 2, 259-261 (2007)

¹³ J. R. Creighton, P. Ho. Chemical vapor deposition. Introduction to chemical vapor deposition (CVD), ch. 1. (2001)

¹⁴ V. A. Shchukin, D. Bimberg. Spontaneous ordering of nanostructures on crystal surfaces. Rev. Mod. Phys., 71, 1125-1171 (1999)

¹⁵ S. Franssila. Introduction to micro fabrication. Structures by deposition, ch.11. Wiley (2004)

¹⁶ H. Jansen, H. Gardeniers, M. de Boer, M. Elwenspoek, J. Fluitman. A survey on the reactive ion etching of silicon in microtechnology. Journal of Micromechanics and Microengineering, 6, 14-28 (1996)

¹⁷ M. D. Henry. ICP etching of silicon for micro and nanoscale devices. Doctorate thesis, California Institute of Technology (2010)

¹⁸ U. K. Khankhoje, S-H Kim, B. C. Richards, J. Hendrickson, J. Sweet, J. D. Olitzky, G. Khitrova, H. M. Gibbs, A. Scherer. Modelling and fabrication of GaAs photonic-crystal cavities for cavity quantum electrodynamics. Nanotechnology, 21, 065202 (2010)

¹⁹ CSAR 62 (AR-P 6200), Allresist, <u>http://www.allresist.com/csar-62-ar-p-6200/</u> (last accessed 01/02/2017)

²⁰ A. Faron. Locally controlled photonic crystal devices with coupled quantum dots: physics and applications. Doctorate thesis, Stanford University (2009)

²¹ V. A. Mandelshtam, H. S. Taylor. Harmonic inversion of time signals and its applications. J. Chem. Phys., 107, 6756-6769 (1997)

²² A. Taflove, S. C. Hagness. Computational Electrodynamics: The finite-difference-time-domain method, third edition. Boston: Artech House (2005)

²³ D. H. Choi, W. J. R. Hoefer. The Finite-difference-time-domain method and its application to eigenvalue problems. IEEE Transactions on Microwave Theory and Techniques, 34, 1464-1470 (1986)

Chapter 4

Photonic Crystals for Coupling to 2D Materials

4.1. Introduction

Demonstrating reliable and miniaturised quantum light sources has been a major challenge in the past couple of decades¹. The realisation of such sources could resolve many challenges in creating integrated quantum technology-based applications such as quantum key distribution and quantum computing^{2,3}. A brief introduction to single photon sources was presented in chapter 2. In this chapter of the thesis, the strong potential for quantum light emission from 2D materials will be introduced. Following this, a model for coupling transition metal dichalcogenides (TMDCs), a specific type of 2D material, to resonant cavities based on photonic crystals (PhCs) is suggested. Numerical simulations using finite difference time domain (FDTD) are used to prove that the suggested cavity design provides improved light-matter interactions and enhancement in the light collection efficiency. In addition, analysis of the reliability and robustness of the suggested design against fabrication and experimental uncertainties will be carried out for claim support. Later, preliminary fabrication results of the suggested cavities and the transfer of TMDC flakes on top, using both new and pre-developed techniques, will show the maturity that this research direction has reached for realising chip-based quantum light emitters.

4.2. TMDC Monolayers as Quantum Light Emitters

A dramatic increase in scientific research interest in 2D materials has taken place in the past decade due to their extraordinary mechanical, electrical, thermal and optical properties⁴⁻⁷. For quantum technology applications, their unique optical properties, particularly those based on transition metal dichalcogenide materials, make them prospective candidates for single photon sources. Recently, multiple demonstrations of single photon emission from defects in Tungsten Diselenide (WSe₂) and MoS₂ have been reported using electrical and optical excitation^{8,9}. Lattice defects in TMDC monolayers, such as the ones shown in Figure 4.1 (b), can form localised electronic states inside the band gap of the TMDC monolayer. These localised excitons, otherwise known as quantum dots (QDs) due to their zero-dimensional confinement, appear with narrow linewidths in the optical photoluminescence signature of the TMDC monolayers. On the other hand, impurities due to doping of the lattice, can have similar optical properties and be harnessed in a similar fashion for quantum light applications.



Figure 4.1: (a) A schematic illustration of the atomic structure of a typical, (a) pure, (b) defective and (c) impure MoS_2 monolayer flakes

Currently, a bottleneck for the application of 2D materials is their low optical absorption, quantum yield and extraction efficiency of approximately a few percent. This severely reduces the luminescence efficiency of light collected from these materials, hence reducing their potential optoelectronic implementations¹⁰. Existing approaches to work around this issue are material specific, such as using organic superacid to treat MoS_2^{11} , using tunable Bragg mirrors for MoS_2 and BN heterostructures and enhancing the luminescence of MoS_2 by fabricating plasmonic structures^{12,13}. Therefore, a universal approach that offers enhancement for all TMDCs would be advantageous in solving many issues related to light output from 2D materials. Using PhC cavities is one technique to improve light-matter interactions and maximise light extraction.

Monolayer-cavity coupling has been demonstrated recently using a hole-type PhC¹⁴⁻¹⁷. Lasing and enhancement in the spontaneous emission rate for light emitted from 2D materials has also been shown. Whilst hole-type PhC cavities can have high Q-factors due to reduced mode loss in the vertical direction due to the index contrast at the bridge-air interface, this method suffers from a few disadvantages. The first is that the cavity mode is confined within the bridge structure comprising the high index region, whereas TMDC surface emitter monolayers are conventionally exfoliated or wet transferred above this structure. This causes reduced coupling between the emitter and the cavity mode's maximum, leading to lower light-matter interactions. The second disadvantage in using hole-type PhCs is that it results in most of the cavity's resonant mode being confined laterally within the high index slab, rather than vertically coupled out of the plane to interact with the TMDC. This limits the collection efficiency of light collected from these TMDC monolayers. Thirdly, the large dielectric-to-air volume ratio that exists in the structures of hole-type PhCs results in undesirable light absorption by the dielectric material, which is particularly critical when Si or GaAs is used as the dielectric material while the system operation wavelength is in the visible regime. These disadvantages were the main motivation for attempting to couple cavities and monolayer emitters using rod-type PhCs.

Using rod-type PhC cavities, the previously mentioned issues with hole-type PhCs can be solved, as will be shown later. Similar to other forms of cavities, the suggested rod-type PhCs in this thesis comprise a hexagonal array of rods with a lattice constant, *a*, and a rod radius, *r*. The cavity that will be mainly discussed in this thesis is an H1 type of cavity where only one rod is missed from the structure to create the localised state in the photonic bandgap of the lattice.

In previous work, carried out in Lancaster University, researchers have shown that 2D materials dip when transferred onto hollowed regions such as etched trenches on a substrate¹⁸. In this work, we suggest exploiting the flexible nature of TMDC monolayers to allow the dipping of a monolayer



Figure 4.2: A schematics illustration of our suggested method for TMD-cavity coupling, showing the bowing of a monolayer flake above a rod-type PhC cavity to allow optimised coupling to the centre of the cavity where the field maximum exists.

transferred onto a rod-type PhC to make it couple to the cavity's centre region where the mode's antinode exists. Figure 4.2 shows an illustrative image of the suggested cavity design.

Interestingly, the reason behind choosing a hexagonal array PhC structure over the square array one is due to the latter's small radius pillar requirement which can become a limitation in the fabrication stage. For rod-type cavities, square lattice arrays such as the one explained in reference 8, have been extensively studied and a transverse magnetic (TM) bandgap has been shown. However, for wavelengths in the visible regime such as those emitted by TMDC monolayers, the size of a square lattice PhC designed to have its bandgap around this wavelength will require small dimensions compared to a hexagonal lattice designed for the same wavelength range. For example, assume a square lattice PhC is designed for molybdenum disulphide (MoS₂), i.e. its bandgap range covers the central emission wavelength of MoS₂, which is approximately 660 nm, as shown in Figure 4.4 (a); an example of a MoS₂ flake is shown in Figure 4.4 (b). Kay et al. show that the for a PhC lattice of a = 1 and r = 0.2a, an H1 cavity would have its cavity mode at $\lambda = 2.56a$, in normalised units¹⁹. In other words, a PhC cavity with a mode at $\lambda = 660 nm$, requires its lattice constant to be, a = 258 nm and the radius to be, r = 52 nm, with rod heights of at least 660 nm. Designing rods with such dimensions is very challenging and requires an extremely accurate fabrication process. Furthermore, small fabrication
uncertainties that changes the rod's radius, the lattice constant, sidewall roughness and/or their vertical height can shift the cavity mode and change its extraction efficiency. An H1 hexagonal lattice PhC cavity of similar dimensions has its cavity mode at a much smaller wavelength. Making a cavity with a mode wavelength of 660 nm makes it less dependent on fabrication limitations. The cavity suggested in this work can achieve this goal using a = 595 nm and r = 95 nm, over twice as large as the square lattice crystal.

The PhC hexagonal lattice proposed in this work is surrounded by air for optimum index contrast. The rods have a refractive index of 3.9, corresponding to the index of refraction for silicon at the MoS_2 monolayer emission wavelength of approximately 660 nm. MoS_2 was the material used for this work because of its large neutral and charged exciton binding energies, making it a great candidate for room



Figure 4.3: (a) Our suggested cavity design, top and cross section screenshots from the modelling software showing the flux region and the cavity structure. The lattice constant *a* is measured as the separation between the centres of two adjacent holes in the lattice. B and c show the top and side views of the setup of the proposed rod type PhC cavity with the position of the flux region above it. The simulation region is surrounded by a PML layer to prevent light reflection from the boundaries of the simulation cell.



Figure 4.4: (a) Room temperature photoluminescence emission from a MoS2 monolayer showing central emission peak near 660 nm. (b) A microscope image of a typical MoS_2 mechanically exfoliated sample, showing a monolayer, a bilayer and a thick flake. The thickness of 30 layers was roughly estimated from the reflected lime flake colour.



Figure 4.5: Top view of a time slice of the E_x (a) and E_y (b) field components of the PhC cavity. (c) Cross section of the E_y field components within the microcavity. Red (blue) illustrate the positive (negative) values of the electric field. The figure shows that maximum cavity mode field occurs in the centre of the cavity.

temperature quantum light sources. Nevertheless, the PhC design is universal, as the normalised parameters may be scaled and adjusted to match any light-emitting TMDC. It consists of rods with a lattice constant, a = 1, and a radius, r = 0.165a, where the rods are made by etching the silicon substrate. The fabrication process will be discussed in section 4.3. Unsurprisingly, the PhC rod's height also influences the photonic confinement. If the rods are too short, the cavity mode's shape can extend into the air above the PhC. This results in a reduction in the spatial interface between the mode and the rods, lowering the light collection ratio. Conversely, if the rods are too tall, higher order and propagating modes can form within the cavity structure. Furthermore, etching high aspect ratio rods will always require complex dry etching techniques to achieve the desired degree of anisotropy. Previous work has revealed that the maximum gap size for a square-lattice rod-type PhC is achieved for rod heights of approximately 2.3*a*, corresponding to approximately two cavity modes wavelengths²⁰. During the period of this work, similar results were found for the suggested hexagonal lattice. Therefore, the rods' height was fixed to 2.3*a* in all simulation runs.

The Plane Wave Expansion (PWE) method was the first step in the simulation process to model the bandgap region for the designed structure. 2D PWE simulations of the PhC structure shown in Figure 4.3 (a), before creating the defect cavity, is presented in Figure 4.5a. A bandgap in the frequency range $1.05 < \lambda < 1.17$ is found. Once the photonic band diagram was simulated, an H1 cavity is created. Subsequently, FDTD simulations was used to test the cavity's performance. Initial 2D simulations was carried out to confirm the presence of the cavity mode and analyse its central wavelength. It is worth mentioning that Q-factor measurements in 2D simulations will show astronomical values since leakage in the vertical direction is virtually zero compared to the 3D case. The main reason behind performing 2D simulations in the first place is to save computational time and power.

3D FDTD simulations of the cavity are shown in Figure 4.5 (a-c). Initially an E_x polarised source was placed at the centre of the cavity, with a central wavelength λ of 1.1*a* and a Full-Width Half Maximum (FWHM) of $d\lambda = 0.1$. Using Meep, a visual image of the light propagation can be observed. A few steps after the initialisation of the dipole, where edge state and propagating ones are allowed to leak, the field components of the cavity mode can be easily observed on its own, as shown in Figure 4.5 (c). The cavity is later simulated with an E_y polarised dipole. The electric field distribution for the cavity mode is shown in Figure 4.5 (b). Using the Harmonic inversion (harminv) tool embedded in Meep, the cavity mode was found to be at a wavelength $\lambda = 1.12a$ with a Q-factor of 300. On the other hand, Figure 4.5 (c), shows the field distribution for a cross section slice along the x direction through the cavity. It is easy to observe that although the cavity's top is primarily air light can be confined in the vertical direction due to the index differential at the top of the pillars, this makes reasonable confinement of light components leaking at a wide angle vertically, parallel to the norm of the substrate. This makes this cavity structure which is based on a rod-type PhC structure appealing for enhancing the light collection efficiency toward the collecting optics. This is because only the vertical components of light

are allowed to leak upward, while components leaking at other angles are confined due to the photonic band gap laterally and due to the high index contrast at the rods' top-air interface. It is important to mention here that absorption due to the dielectric material is not considered here. Absorption due to the dielectric material is expected to reduce the cavity's Q, however this becomes less important for low Q cavities such as the ones shown here where the light escapes due to low confinement in the vertical direction. This makes this technique potentially capable of enhancing light from quantum emitters to be efficiently collected in the vertical direction by external collecting optics.

Measuring the enhancement in the light collection efficiency requires outputting the light flux spectrum due to the cavity. This is then compared to the case when the source is placed directly above a substrate without a PhC structure, representing a TMDC monolayer mechanically exfoliated directly on the substrate. The enhancement ratio, η_e , is calculated as the power flux with and without the cavity according to equation 1.

$$\eta_e = P_{cav}/P_0 \tag{1}$$

Figure 4.3 (b and c) shows screenshots taken directly from Lumerical's user interface for the simulation cells. The mesh resolution used in this simulation is 20 elements per lattice constant in every dimension. Corresponding to approximately one element per 30 nm in real units. It was found that increasing the number of elements does not add much more accuracy to the obtained results, hence the resolution was fixed for all subsequent simulations at this value to save computation time and power. To measure the enhancement in the light collection efficiency, the flux region was made to have an area of $23a^2$ and setup above the cavity, collecting light vertically radiated upward, mimicking an objective lens with numerical aperture of 0.65.



Figure 4.6: (a) FDTD simulations of the enhancement spectra for rods with a radius ranging from 0.155a and 0.170a. (b) Normalised flux spectrum for a crystal with radius r = 0.161a, in comparison with the emission from a source on a bulk substrate, showing a 400% enhancement in the obtained flux.

When the source is placed directly on the substrate, a Gaussian curve corresponding to the dipole is recorded by the flux region. Later, a series of simulations were carried out by having a source inside a PhC cavity containing rods with different radii. The aim was primarily to investigate the robustness of this design to fabrication imperfections that are likely to occur due to the relatively small structure. Causes of shifts in the radius of the fabricated rods could be due to inaccurate selection of exposure dosage in the electron-beam (e-beam) writing process. Other reasons could be due to non-anisotropic sidewall profile development of the exposed regions of the resist, due to temperature variations of the developing solution and/or the samples themselves. Hence, the radius was varied between 0.155a and 0.170a. Maximum enhancement was achieved for a PhC cavity having rods with a radius, r = 0.161a, where the cavity mode's central wavelength, $\lambda = 1.104a$, as shown in Figure 4.6 (a). The enhancement obtained here is attributed to radiation modes from the cavity leaking vertically toward the flux region. As the radius deviates from r = 0.161a, the collected power flux starts to decrease, reducing the enhancement ratio, η_e . It is clear from Figure 4.6 to note that reducing the radius of the rods tends to blue-shift the cavity mode's energy, thus reducing its wavelength. This is expected, as the relationship between the PhC's cell radius, whether it is hole-type or rod-type, and the cavity mode's energy was anticipated through previous studies²⁰. Figure 4.6 (b) plots a comparison between the obtained fluxes for two cases. The first case is when a flake is exfoliated directly onto a flat substrate. The second case is when the flake is transferred on top of the PhC structure, dipping to half the height of the rods where

the cavity's field maximum exists, as shown in in Figure 4.7. Later, the effect of having the source away from the cavity's centre will be discussed.



Figure 4.7: The cavity's emitted flux spectra as the dipole position within the cavity is varied along the z-axis, where maximum enhancement is obtained when the source is at a height, h/2. The enhancement reduces as the source height is varied along the z-direction of the cavity. The figure on the right illustrates the degrees of bowing of the 2D material from the top of the rods for each of the cases shown on the left.

Throughout the scope of this work, the simulations were ran assuming the flake is dipped to about half the height of the rods. This is expected to be the ideal case since the source will be coupled to the field's antinode, creating maximum spatial coupling between the source and cavity. Dipping of the monolayer to about half the height of the rods can promote the creation of defects due to strain, creating localised excitons within the monolayer, however this might not be the case in reality and is discussed later in the fabrication section. Performing simulations to anticipate the amount of dipping of a monolayer over a typical structure depends on many factors. These include the monolayer material, the rods' surface roughness, and even the transfer temperature. These typically require setting up finite element simulations whilst taking into account the van der Waals, Coulombic and gravitational force, which can be exhaustive. Hence, a series of simulations were run with the dipole source emitter placed at different heights along the vertical direction inside the cavity, to investigate the effect of dipping of the monolayer emitter on the collection efficiency. Even though the source position in the vertical direction was changed along the height of the cavity, it is important to mention that the flux region height above the photonic structure was not changed. The cavity spectra for the different heights are shown in Figure 4.7. It is expected that maximum Purcell enhancement for a light emitter should be achieved when the source is spatially aligned with the cavity mode's electric field maximum. This exists at the centre of the cavity, one wavelength above the substrate, as was previously shown in Figure 4.5 (c). Enhancement decreases as the source is moved toward the top of the rods due to reduced spatial coupling between the emitter and the cavity mode, whilst also coupling to radiative modes in air. If the source is placed at the bottom of the cavity, the main cause of reduction in enhancement is due to coupling to substrate-mediated leaky modes.

Compared to H1 cavities, L3 cavities are usually favoured for better coupling to PhC waveguides, and tend to have lower confinement quality factors of roughly 150. In L3 cavities, the enhancement in the extraction efficiency is reduced due to non-vertical components of the cavity mode leaking at a wide range of angles and missing the flux region.

4.3. Cavity Fabrication and Emitter Transfer

Fabricating hole-type PhCs has been achieved by many groups. This is due to its suitability in coupling self-assembled QDs grown by the Stranski-Krastanov method to the cavity mode which is normally confined within the bridge, where the dots normally exist (see chapter 3). Rod-type PhCs have been fabricated and tested by fewer groups. Hence, much less literature was available to guide in the fabrication steps of rod-type PhCs. This part of the chapter will explain the details for the approach taken to fabricating this type of PhC.

Details of the working principles of the fabrication tools used in realising the rod-type PhC structure are explained in chapter 3. While Silicon and GaAs have both similar refractive indices for light in the visible regime, silicon was the substrate material used since it has a wider industrial use. The important advantage Silicon holds over GaAs is the reduced electron and nuclear spin interaction in the optical characteristics of single QDs. In addition to improved light collection efficiency, this is one of the future research directions that this work can also lead to.

The fabrication process starts with a (100) Silicon substrate with the oxide removed via HF etching. The substrate is spin-coated with CSAR62 (AR-P 6200), which is a positive e-beam resist developed by Allresist as a cheap alternative to ZEP520A. Compared to poly(methyl-methacrylate) (PMMA), CSAR62 was found to achieve higher contrast and requires less exposure dosage, making the patterning process much faster and more cost effective. In chapter 3, the advantage of the high selectivity to dry etching that CSAR62 benefits from compared to PMMA was shown as a key factor for fabricating holetype PhCs. The resist material is usually diluted in a 1:1 ratio of CSAR62: Anisole to reduce the concentration and allow thin film spin coating. It is important to choose the film thickness wisely, since there will be a hard mask metal lift-off process that will take place after the e-beam exposure. A rule of thumb known in the industry is that there needs to be a minimum of a 3:1 thickness ratio between the resist and the lift-off metal. Spinning too thin a layer will cause the lift-off process to fail, on the other hand, spinning a thick layer will reduce the contrast of the achieved pattern. Producing high contrast patterns is especially important in this work since the cavity mode's central wavelength and extraction efficiency are directly related to the fabricated radius of the rods. The thickness used for realising the structure was thus 120 nm. The spin-coated sample is subsequently baked on a hot plate at 180 °C for 3 minutes.

The mask was designed with AutoCAD and corrected with BEAMER to reduce proximity effects. The pattern writing was done with a 50kV JEOL (JBX-5500ZD) using a beam current of 50 pA and an exposure dosage of $120 \,\mu\text{C/cm}^2$. The pattern was later developed with o-xylene for 140 s, during which gentle sonication took place for 15 s. It was found that gently sonicating the sample while it is in the developing solution helps removing the exposed resist layer completely, until the substrate is exposed to air. The issue of partial development of the resist is more prominent with PMMA for small features, such as those found in fabricating the hole-type PhC structure, as was shown in chapter 3. This was another advantage for using CSAR62 over PMMA.

During the period of this work, many tests were carried out to identify the material that acts as the best hard mask for dry etching Silicon using Reactive Ion Etching (RIE). A judgement was made based on the etching selectivity and the ease of pattern transfer through the lift-off process. Using the dry etching recipe developed during this work (which will be explained later), the selectivity achieved in etching Silicon with an Aluminium hard mask was 13:1. Nickel was found to have a selectivity of 18:1, whilst with Chromium the selectivity was 49:1. Previous work has shown high selectivity in etching Si using SiO₂, where it was calculated that the selectivity can range from 5-35:1 depending on the etching recipe²¹. Hence, efforts were primarily focused toward transferring the pattern to the substrate using Cr as a hard mask.

To measure the etching selectivity, a Veeco AFM CP-II was used. The resist thickness was measured after spinning using the developed patterns. After etching, another measurement was made to measure the sum of the etch depth and the remained resist. Finally the resist is stripped off the substrate and the actual etch depth was measured. Given the etch time period, this set of measurements can be used to calculate the etch rate of the resist and the etch rate of the semiconductor, hence the selectivity.

Cr is bought attached to a tungsten wire from which it can sublimate when a current of 60-70 A and a voltage of 2 V is applied at 10^{-6} Pa. Importantly, the desired rod height should be approximately 2.25 times the lattice constant for a maximum Q-factor and extraction efficiency²⁰. In this case, the rods should be etched by approximately 1320 nm, for efficiently coupling the cavity to a MoS₂ monolayer that has a central emission wavelength of 660 nm. With a 49:1 selectivity, the Cr thickness should be at least 27 nm. However, to ensure enough Cr is available for the etching process, the metal thickness was made approximately 40 nm. This is the reason that the resist thickness was targeted for 120 nm, to follow the 3:1 resist:metal lifeoff rule of thumb.

The lift-off process was done in PG remover 1165, which is a mixture of pure organic materials including N-methyl-2-pyrrolidine, compatible with many metals including Chromium and manufactured by Microposit. The sample is inspected under a long working distance objective to identify if the lift-off process was successful. Sonication is usually desirable if the lift-off process took more than 24 hours to remove any remained resist quickly. However, it was found that sonicating for

long times tends to strip off the Cr discs that are attached to the substrate, which is a crucial factor that will prevent formation of the vertical rods.

The sample is dry etched using Sulphur hexafluoride (SF₆) and Oxygen (O₂) based RIE etching. The flow of SF₆ and O₂ were 10 and 5.5 sccm, respectively. The etching RF power was 130 W and the chamber pressure was 5 mTorr. This recipe produced an etching rate of approximately 90 nm per minute, so typically the etching period is approximately 15 minutes. Figure 4.8 shows examples of fabricated rod-type PhC cavities with rod heights of approximately one cavity mode wavelength. The reason one cavity mode wavelength was used instead of the value referenced above, was due to the reduced verticality of the sidewalls after etching beyond 1 micron. However, although using a height of one wavelength is not optimum, it is still expected to provide good coupling between the cavity and the source.



Figure 4.8: Scanning electron micrographs showing examples of a L3 fabricated cavity with high aspect ratio rods. Realising the rods with this level of etching anisotropy typically requires careful optimisation of the etching parameters.

The MoS₂ crystal material was bought from an external supplier in centimetre size bulks and exfoliation is carried out using an exfoliation tape to de-attach atomically thin layers from of MoS₂ from the crystal. The MoS₂ were bought from a manufacturing company called Manchester Nanomaterials. The MoS₂ is synthesised using chemical vapour transport techniques which is normally used to grow atomic layer thick flakes, but can grow thick layers if it was run for durations that can be as long as multiple days²². To transfer a monolayer flake on the cavity structure, mechanical exfoliation is performed on a sacrificial substrate. This substrate is usually made of Silicon with a deposited 300 nm SiO₂ layer that

covers it. The SiO₂ layer plays a vital role in providing enhanced optical contrast for finding exfoliated monolayers under the optical microscope. Using the Si/SiO₂ substrate, Polyvinyl Alcohol (PVA) is spin-coated at 2000 rpm to create a layer approximately 100 nm in thickness. Subsequently, PMMA is spin-coated on top of the PVA layer at 4000 rpm and baked at 150 °C to create a layer with a thickness of 200 nm. Finally, the desired TMDC is mechanically exfoliated and transferred to the Si/SiO₂/PVA/PMMA stack. Monolayers are then visually identified under an optical microscope and those that look similar in colour and brightness to that shown in Figure 4.4 (b) are prepared for transfer.

The flake is transferred using the wet transfer process, which involves stamping the flake over the cavity²³. A schematic of the wet transfer process is shown in Figure 4.9. Trenches are etched around the monolayer to create an isolated island using a sharp pointer pin that scratches through the PVA and PMMA. Water droplets are allowed in the trenches to dissolve the sacrificial PVA layer and let the top PMMA layer float on the water. The sample is then immersed in a water bath such that the PMMA layer detaches from the sample and floats on the surface of the bath, isolating the PMMA layer. The floating PMMA film is scooped from the water surface using a metal washer ring and the washer is placed on a hot plate to allow adhesion between the PMMA layer and the washer. The washer is flipped and mounted on a tool made in-house to allow clipping of the washer to a slow descending arm which lowers the washer/PMMA toward the sample. A microscope is used to align the flake on top of the cavity before mounting. It is desirable to have the sample on a hotplate before the actual transfer to improve adhesion between the flake and the cavity. Figure 4.10 shows the initial results achieved in transferring monolayers of MoS₂ onto rod-type PhC cavities.

Room temperature μ -photoluminescence measurements of the sample shown in Figure 4.8 did not show any cavity enhancement to the emission of the MoS₂ monolayer, the emission obtained is shown in Figure 4.11. The figure shows a strong bi-layer emission with a peak central wavelength of 810 nm and a weaker monolayer emission around 660 nm, without any indication of cavity mode coupling¹¹. SEM imaging proves that this is caused due to miss-alignment of the flake as it was transferred above the cavity which can be observed in Figure 4.10 (b).



Figure 4.9: A schematic illustration of the steps involved in the dry transfer technique. (a) The initial structure of the wet transfer is made of Si with PVA and PMMA spin coated on the substrate and the flake exfoliated on top of the structure. (b) Trenches are made and water droplet are allowed passed through the trenches to allow dissolving the PVA layer. (c) The PMMA layer together with the flake are detached from the wafer and float on a water bath. (d) The PMMA layer is then scooped out of the bath and stamped on the rods structure.



Figure 4.10: Scanning Electron Micrographs showing initial results from transferring monolayer flakes on top of a fabricated PhC cavity made in silicon by e-beam lithography and dry RIE etching. The figures shows how the topology of a flake can be varied by the structure of its hosting substrate. This was an initial attempt though, perfect alignment of a flake above a cavity was not achieved.



Figure 4.11: Room temperature μ -photoluminescence emission from the MoS₂ flake shown in Figure 4.10 excited with a laser power of 3 mW. The sample was placed under vacuum in the order of 10⁻³ mtorr to ensure that the cavity surrounding has a refractive index of as close to 1 as possible, matching that of the simulated cavity. The figure shows emission signatures of a monolayer and a bilayer corresponding to central wavelengths of 660 and 810 nm.

4.4. Conclusions

In conclusion, a model was proposed to couple 2D material-based light emitters to a resonant cavity efficiently. The model that was proposed which is based on rod-type PhCs provides advantages over current methods for 2D material-cavity coupling. These advantages include spatial coupling between the field's maximum and the quantum emitter and improved light collection efficiency from these cavities. The photonic band diagram for the designed structure was solved and the field distribution within an H1 cavity was simulated. Simulations of the collected flux spectra was measured for a range of PhC rod radii and maximum enhancement in the collection efficiency of about 400% was shown for r = 0.161a. A series of simulations were carried out to investigate the effect of having the monolayer dipping at different heights inside the cavity. It was shown that maximum enhancement is achieved when the source spatially overlaps with the cavity mode's maximum and reduces as the source is moved up or down from the cavity due to reduced spatial coupling and coupling to leaky modes outside the cavity, respectively.

Details of the fabrication process of these PhCs were explained and preliminary results were shown. The method of transferring monolayers on the cavity and the results achieved were demonstrated.

Enhancing the collection efficiency further could be done using the author's recent work in Solid Immersion Lenses^{24,25}. Glass SILs can be reliably positioned on top of the cavities with micrometer-scale accuracy. These kind of SILs were shown to give over 300% enhancement in the light collection efficiency. While SILs made of other materials such as GaAs can achieve over 1000% enhancement as discussed in reference 24. Combining SILs with the PhC cavities can lead to a greater increase in the light collected from TMDC monolayers. Another advantage in placing a SIL on top of the cavity can be enhancing the vertical confinement due to total internal reflection at the air-glass interface. Initial results from FDTD simulations show that adding a slab with a similar refractive index to that of glass showed that the cavity's Q-factor can be increased by four folds.

Bibliography

¹ M. D. Eisaman, J. Fan, A. Migdall, S. V. Polyakov. Single-photon sources and detectors. Rev. Sci. Instrum., 82, 071101 (2011)

² N. Gisin, G. Ribordy, W. Tittel, H. Zbinden. Quantum cryptography. Rev. Mod. Phys., 74, 145-195 (2002)

³ T. D. Ladd, F. Jelezko, R. Laflamme, Y. Nakamura, C. Monroe, J. L. O'Brien. Quantum computers. Nature, 464, 45-53 (2010)

⁴ D. Akinwande, C. Brennan, J. Scott Bunch, et al.. A review on mechanics and mechanical properties of 2D materials-Graphene and beyond. Extreme Mechanics Letters, 13, 42-77 (2017)

⁵ D. Wei, Y. Liu, Y. Wang, H. Zhang, L. Huang, G. Yu. Synthesis of N-doped Graphene by chemical vapour deposition and its electrical properties. Nano Lett., 9, 1752-1758 (2009)

⁶ A. A. Balandin. Thermal properties of graphene and nanostructured carbon materials. Nature Mater., 10, 569-581 (2011).

⁷ F. Xia, H. Wang, D. Xiao, M. Dubey, A. Ramasubramaniam. Two-dimensional material nanophotonics. Nature Photon., 8, 899-907 (2014)

⁸ C. P. Berraquero, M. Barbone, D. M. Kara, G. Ilya, D. Yoon, A. K. Ott, J. Beitner, K. Watanabe, T. Waniguchi, A. C. Ferrari, M. Atature. Atomically thin quantum light emitting diodes. Nature Commun., 12978 (2016)

⁹ C. Chakraborty, L. Kinnischtzke, K. M. Goodfellow, R. Beams, A. N. Vamivakas. Voltage-controlled quantum light from an atomically thin semiconductor. Nature Nanotechnol., 10, 507-511 (2015)

¹⁰ K. R. Paton, J. N. Coleman. Relating the optical absorption coefficient of nanosheet dispersion to the intrinsic monolayer absorption. arXiv:1511.04410 (2015)

¹¹ M. Amani, D. H. Lien, D. Kiriya, et al.. Near-unity photoluminescence quantum yield in MoS₂. Science, 4, 1065-1068 (2015)

¹² S. Dufferwiel, S. Schwarz, F. Withers, et al. Exciton-polaritons in van der Waals heterostructures embedded in tunable microcavities. Nature Commun., 6, 9576 (2015)

¹³ K. C. J. Lee, Y. Chen, H. Lin, C. Cheng, P. Chen, T. Wu, M. Shih, K. Wei, L. Li, C. Chang. Plasmonic gold nanorods coverage influence on enhancement of the photoluminescence of two-dimensional MoS₂ monolayer. Sci. Rep., 5, 16374 (2015)

¹⁴ S. Wu, S. Buckley, A. M. Joes, J. S. Ross, N. J. Ghimire, J. Yan, D. G. Mandrus, W. Yao, F. Hatami, J. Vuckovic, A. Majumdar, X. Xu. Control of two-dimensional excitonic light emission via photonic crystal. IOP 2D Mater., 1, 011001 (2014)

¹⁵ X. Gan, Y. Gao, K. F. Mak, X. Yao, R. Shiue, A. Van der Zande, M. E. Trusheim, F. Hatami, T. F. Heinz, J. Hone, D. Englund. Controlling the spontaneous emission rate of monolayer MoS_2 in a photonic crystal nanocavity. Appl. Phys. Lett., 103, 181119 (2013)

¹⁶ Y. Ye, Z. J. Wong, X. Lu, H. Zhu, X. Chen, Y. Wang, X. Zhang. Monolayer exciton laser. Nature Photon., 9, 733-737 (2015)

¹⁷S. Wu, S. Buckley, J. R. Schaibley, L. Feng, J. Yan, D. G. Mandrus, F. Hatami, W. Yao, J. Vuckovic. Monolayer semiconductor nanocavity laser with ultralow thresholds. Nature, 520, 69-72 (2015)

¹⁸ N. D. Kay, B. J. Robinson, V. I. Falko, K. S. Novoselov, O. V. Kolosov. Electromechanical sending of substrate charge hidden under atomic 2D crystals. Nano Lett, 14 (6), 3400-3404 (2014)

¹⁹ P. R. Villeneuve, S. Fan, J. D. Joannopoulos. Microcavities in photonic crystals: mode symmetry, tunability, and coupling efficiency. Phys. Rev. B, 54, 7837-7842 (1996)

²⁰ C. Gui, Y. Zhang, J. Du, J. Xia, J. Wang. Experimental demonstration of analog signal transmission in a silicon photonic crystal L3 resonator. Optics Express, 23, 13916-13923 (2015)

²¹ N. R. Rueger, M. F. Doemling, M. Schaepkens, J. J. Beulens, T. E. F. M. Standaer, G. S. Oehrlein. Selective etching of SiO₂ over polycrystalline silicon using CHF₃ in an inductively coupled plasma reactor. Optics Express, 23, 13916-13923 (2015)

²² R. Roldan, A. Castellanos-Gomez, E. Cappelluti, F. Guinea. Strain engineering in semiconducting two-dimensional crystals. J. Phys.: Condens. Matter, 27, 313201 (2015)

²³ A. Castellanos-Gomez, M. Buscema, R. Molenaar, V. Singh, L. Janssen, H. S. J. Van der Zant, A. G. Steele. Deterministic transfer of two-dimensional materials by all-dry viscoelastic stamping. IOP 2D Mater, 1, 011002 (2014)

²⁴ C. S. Woodhead, J. Roberts, Y. J. Noori, Y. Cao, R. Bernardo-Gavito, P. Tovee, A. Kozikov, K. Novoselov, R. J. Young. IOP 2D Mater., 4, 015032 (2016)

²⁵ K. A. Serrels, E. Ramsay, P. A. Dalgarno, B. D. Gerardot, J. A. O'Connor, R. H. Hadfield, R. J. Warburton, D. T. Reid. Solid immersion lens applications for nanophotonic devices. J. Nano-photonics, 2, 021854 (2008)

Chapter 5

Catalytic Free Deterministic Positioning of III-V Nanowires on Silicon for Photodetector Applications

5.1. Introduction

Avalanche photodetectors (APDs) exploit the photoelectric effect to convert photons incident on a photodiode into small electrical pulses that are strengthened by avalanche multiplications to make them measurable by modern electronic counters^{1,2}. As discussed previously in chapter 2, a semiconductor avalanche photodetector is a p-i-n junction that is reverse biased under high voltages, but not higher than their breakdown voltages. When a photon is absorbed by the intrinsic region, an electron-hole pair is created. The electron and hole are subsequently separated and accelerated by the electric field to the n and p sides, respectively. This causes other electron-hole pairs to be created due to the high kinetic energy that the initial electron-hole pair exhibits due to the applied electric field. Although state-of-the-art APDs have a higher dark current and lower quantum efficiency compared to superconducting nanowire detectors, APDs do not require cryogenic temperatures and can operate reasonably well at practically achievable temperatures for use in applications such as Quantum Key Distribution (QKD). Despite the differences in the fabrication and integration complexity, both of the previously mentioned technologies for single photon detection can be integrated. Although, one is more efficient, has a higher signal to noise ratio, but requires cryogenic temperatures and is expensive to run, while the other is less

efficient but can operate at low temperatures achievable by thermoelectric coolers. The current effort is directed toward realising a room temperature SPD that has a high quantum efficiency and small dark current for faster QKD bit rates and more practical quantum technology applications.

In recent years, III-V semiconductor nanowires have attracted much attention as potential building blocks for a variety of basic optical components including solar cells, frequency electronics, single photon sources and photodetectors³⁻¹⁰. Nanowires are traditionally chemically synthesised, or grown by epitaxial means using vapour-liquid-solid methods, where the latter has the advantage of realising nanowires with much lower impurities that may otherwise cause atomic defects to appear in the crystal lattice structure. While silicon is the most universally used material in the electronics industry, integration of III-V semiconductors onto Si can result in compact integrated optoelectronic circuits. However, current III-V semiconductor devices, such as photodiodes, image sensors and lasers, are fabricated via a hybrid bonding of III-V bulk substrates onto complementary metal oxide semiconductor (CMOS) integrated circuits^{11,12}. This bonding process can drastically limit the minimum size of the device and entails a large increase in production costs. To solve this issue, epitaxial integration of III-V semiconductor by Si platform can ease large-scale integration and reduce production costs^{13,14}.

Compared to thin film photodetectors, reports have shown that ultra-high photocurrent gains are achievable using GaN p-i-n nanowire based photodetectors, which is three orders of magnitude higher than for those that use thin film structures¹⁵. Moreover, their InAs counterparts have been reported to exhibit high photoresponsivity at the C-band telecommunication wavelength^{16,17}. Also, core-shell GaAs/AlGaAs nanowire photodetectors were shown to have a response time of only a picosecond, making them suitable for high speed performance photodetectors¹⁸. Core-shell InAs/InP nanowires, on the other hand, were reported to have a very high mobility at room temperature¹⁹. In addition to the ultra-high photocurrent gain, higher photo-responsivity, shorter response time and higher mobility, the photonic absorption of nanowire arrays has been increased, by optimising the nanowire's length and diameter, to levels matching those for planar device photodetectors, despite having 10% the volume. The attractive structural aspect of nanowires that makes them advantageous over planar structures is

their wider light collection angle. This increases the probability that an impinging photon on the nanowire at a wide range of angles can be absorbed and converted to an electric current.

The operation temperature of photodetectors is crucial for integrated photonic applications, where the cooling systems can limit the size of the overall device and increase its power requirements. It has been shown recently that adding Sb as a group V material together with As during epitaxial growth of InAs nanowires allows the realisation of low leakage current p-i-n nanowires for photodetection applications at room temperature²⁰. Furthermore, incorporating Sb can improve the nanowire crystal quality and reduce the stacking fault density. Sourribes et al. observed a sharp decrease in the stacking fault density by increasing the Sb content up to 15%, while increasing the field effect mobility by more than three times²¹. These advantages can in practice make nanowire based photodetectors a promising approach that can offer better photodetection solutions in comparison to their planar thin film counterparts.

A wide range of operation wavelengths within the near-infrared spectrum can be achieved for nanowire photodetectors. By tuning the temperature and the material composition that forms the nanowires, the operation wavelength can be tuned. For example, by doping InAs with Sb, the bandgap of the material can be reduced, hence red-shifting the operation wavelength²⁰. In addition, other semiconductor materials such as GaAs and InP can be exploited to realise nanowire-based photodetectors that can operate at other wavelengths such as those in the popular C-band and O-band of the telecommunications spectrum. Developing photon detectors in these frequency ranges is essential since long distance fibre optic telecommunications are solely operated in these bands where fibre attenuation is minimum. The attractive low leakage current achieved previously with InAs nanowires photodetectors together with the high bandgap property of GaAs for room temperature telecom wavelength single photon detection, make these materials attractive for studying in this work.

There are two common methods of biasing a p-i-n nanowire before using it for photodetection applications. The first is done by fabricating metal contacts and placing a nanowire horizontally on the substrate such that each end of the nanowire is connected to a contact pad, as shown in Figure 5.1:. The second is achieved by fabricating contacts on top of a free-standing nanowire grown on a doped substrate used as the back contact, as shown in Figure 5.2. The first method suffers from many problems.

Firstly, the nanowires are usually grown free-standing on a substrate using MBE, PECVD and detached from the substrate by sonicating the substrate in a solution. The wires are then drop cast on a substrate and the solution is allowed to dry, which leaves the nanowires randomly distributed. Spatially positioning the nanowires at a desired location can be done using probe techniques, but is extremely difficult and not scalable enough to be adapted for mass manufacturing. This is especially the case when multiple nanowires are needed to be biased in a nanowire photodetector array. Secondly, there isn't an effective method to optically couple a nanowire photodetector in this form to an integrated waveguide, but can be coupled better to vertical light illumination. On the other hand, exploiting free-standing nanowires for photodetector applications, as shown in Figure 5.2:, can provide many advantages. First, this method can benefit from the advantage of deterministic positioning of nanowires on a substrate by patterning and etching a layer of SiO_2 on a Si substrate, this method will be explained in detail later. Second, a large grid of nanowires can be biased simultaneously using contacts fabricated with more scalable techniques such as photolithography. Thirdly, a nanowire array biased in this form can be fabricated and coupled efficiently to a PhC waveguide with a tapered mode out-coupler, as shown in Figure 5.3. Using a top gold contact is advantageous for reflecting light travelling away from the nanowire, therefore realising better light confinement within the SU8 layer. Alternatively, an optically transparent Ohmic contact material such as indium tin oxide can be used if light incident on the photodetector is coupled vertically.



Figure 5.1: Biasing a nanowire that lies horizontally on a substrate with each end connected to one contact pad, where the light is typically applied from above the substrate. The nanowire is broken off its growth substrate by sonication and transferred on a pre-patterned substrate using the drop casting method.



Figure 5.2: Biasing a free-standing nanowire grown with MBE by etching a hole on the SiO_2 layer and exposing the Si substrate to allow the nanowire to seed. The top gold contact is patterned on an optically transparent material such as SU8 after dry etching a portion of it to expose the top of the N-type region of the nanowire.



Figure 5.3: A suggested example for coupling a PhC waveguide to a p-i-n nanowire photodetector using a tapered out-coupler that direct the light beam from the photonic waveguide toward the intrinsic region of the nanowire.

This chapter will firstly present a simple and effective method to deterministically position III-V material nanowires on a silicon substrate, a potential technique for large scale integration of nanowirebased photodetectors in photonic chips. An effective approach toward fabricating the nanowire sites using electron-beam (e-beam) lithography and chemical wet etching is also presented. Later, results from growing nanowires and the effect of changing the MBE growth temperature on InAs nanowire formation is explored. Results will also be presented from using the same method to selectively grow GaAs nanowires together with preliminary photoluminescence measurements results.

5.2. Site-Controlled Nanowire Fabrication

Epitaxial and synthetic growth of nanowires on a substrate is normally a random process where the nanowires grow uniformly across a substrate. The growth density depends on many parameters such as the substrate's temperature and the abundance of the nanowire composition material in the chamber. Nanowire growth engineering for integrated photonic detectors requires deterministic positioning of the nanowires with a spatial accuracy that is smaller than the system's operation wavelength for an optimised signal collection efficiency. Moreover, nucleation of nanowires on a flat substrate often requires metal alloy seeding to catalyse the growth. However, intense research efforts are currently underway to develop catalyst-free growth techniques, hence avoiding metal contamination whilst favouring full compatibility with Si technology²⁰. The effect of metal incorporation into the nanowire during growth has adverse effects on the optical and electrical properties of the devices, as presented in multiple reports²²⁻²⁵. In addition, avoiding metal seeding reduces the number of technological steps needed to realise the nanowires. The results shown in this chapter include the catalyst-free growth seeding of III-V nanowires on Si substrates.

Integrating high performance III-V nanowires with mainstream Si technology encounters many longterm challenges due to the lattice mismatch, different crystal structures and thermal coefficients. While this process of spatial growing of III-V material nanowires is done on Si, the results presented here can be improved if a lattice-matched III-V material is used as the host substrate. In this work, all the Si substrates used as hosts for the nanowires are p-type (111) with a thermally grown 25 nm SiO₂ coating.

The mask for the nanowire sites constitutes primarily of circularly shaped 50 nm diameter holes etched into the SiO_2 layer until the Si substrate is exposed. Across the mask, different arrays containing different hole diameters and a varying pitch were used to investigate the effect of the resultant structure's geometry on the formed pattern and the growth ratio of nanowires in these holes. Figure 5.4 shows sections of the mask used in this process. Because the mask contains regions with arrays of varying hole density, proximity effect correction was vital to ensure the dosage distribution was uniform throughout the mask.

Chapter 5 - Deterministic Positioning of III-V Nanowires on Silicon for Photodetector Applications



Figure 5.4: A screenshot of the mask used in the fabrication of nanowire sites for photodetectors. The large dense square in the middle of the mask is used for PL measurements of the nanowires to characterise their emission in the mid infrared spectrum. The smaller squares are arrays of many sites with varying densities as shown by the red insets. Alignments marks are made around the whole pattern to mark its edges.

The Si substrate is initially cleaned and sonicated with acetone and Isopropyl alcohol (IPA) before an e-beam resist is spin-coated on the substrate to form a layer thickness of 80 nm. CSAR62 was used as the e-beam resist, where an exposure dosage of $110 \,\mu\text{C/cm}^2$ is used to write the pattern on the wafer, as shown in Figure 5.5 (a). The high dosage sensitivity of CSAR62 compared to PMMA allows the fabrication of patterns with a lower exposure dosage, hence reducing the exposure time and improving contrast of the exposed patterns due to reduced electron forward scattering. Using PMMA required much higher dosages to completely develop the resist such that the underlying substrate is exposed. This issue is especially prominent for small features such as the 50 nm diameter nanowire sites in this work. After exposing the CSAR62 resist, the pattern is developed with o-xylene for 90 s, during which gentle sonication took place for approximately 15 s. This agitates the solution to allow faster extraction of the remaining developed resist from within the holes and allows fresher developing solution to develop the exposed holes further until the SiO₂ layer is exposed. Following developing, the wafer is rinsed in IPA for approximately 2 minutes to stop the developing process, and then dried with Nitrogen gas. An electron micrograph of the developed resist is shown in Figure 5.6.

The SiO₂ layer was wet-etched using a buffered oxide etcher (BOE) (NH₄F (35%), H₂O (60%) and HF (5%)) for 40 s. The pattern is then rinsed in deionised water for 60 s and inspected under an optical microscope. This exact etching process was previously made by Thompson et al in the exact same conditions²⁰. The work has involved AFM imaging of the etched nanowire sites to ensure that the holes have etched completely through the oxide layer exposing the Si substrate. After the BOE etching, CSAR62 did not show any thickness reduction or display signs of removal, this means a high etching selectivity of over 10:1 can be achieved using CSAR62 as a resist. This is in contrast with PMMA which normally has an etching selectivity of less than 2:1 with SiO₂. The resist is then stripped off from the substrate using acetone for 5 minutes before being rinsed with IPA for 2 minutes. The pattern normally appears faint due to the thin SiO₂ layer and its transparent nature. Immediately before loading into the MBE system, the pattern is etched with BOE for 10 s to remove any oxidation due to exposure to air that might have taken place during the resist strip off or microscope inspection stages. Wet etching the pattern was found to increase the holes' diameters to about 90 nm. This is likely to be due to undercutting of the etched holes which is a normal effect that takes place with chemical wet etching as shown in Figure 5.7. To solve this issue, it is possible to utilise plasma methods to dry etch the pattern using reactive ion etching. Using CSAR62 is particularly easier than PMMA in this case due to its higher etching selectivity. In this work, wet etching was the only method used in all the etching attempts.



Figure 5.5: Illustrations of the steps to fabricate hole sites for nanowire growth on SiO₂. (a) Positive CSAR62 e-beam resist is spin-coated on the substrate. (b) Holes are created by exposing the resist with an e-beam and developing using o-xylene. (c) The SiO₂ layer is etched using buffered oxide etcher, however this causes undercutting in the oxide layer (d) the resist is stripped off with acetone.



Figure 5.6: The resultant exposed hole array on CSAR62 after developing, showing holes of 60 nm, slightly larger than the exposed 50 nm holes, which could be resulted from backscattering.



Figure 5.7: Hole arrays of different densities after wet etching the SiO_2 layer and stripping off the etching resist. The figure shows an increase in the holes diameter to approximately 80 nm and the formation of small particles near the edges of the holes. The large rim surrounding the holes is an indication of the large facets caused by wet etching.

It was noted that, the etching step usually leaves remains of SiO_2 at the edge of the etched holes, as shown by the red circles in Figure 5.7. It is anticipated that these remains affect the nanowire growth location within the site, as will be shown in the next section.

5.3. InAs and GaAs Nanowire Growth

The five InAs nanowire samples were grown using In 2D growth rate of 0.07 ML/s, As growth rate of 1.8 ML/s and a Sb growth rate of 1.15 ML/s, where the calibration was made using high energy electron diffraction. The high group V flow rate during the growth process was used to compensate for the material evaporated from the substrate due to the low boiling point of group V materials. The time of

growth was fixed to 1 hour and 20 minutes. After loading the wafer in the MBE growth chamber, it was annealed at an MBE set point temperature of 700 °C (corresponding to an actual temperature of 588 °C, approximated using the oxide desorption temperature of a n-doped GaAs substrate and the GaAs growth transition temperature) for 10 minutes before it is allowed to cool down and remain at a constant set point temperature of 560 °C (494 °C actual) for growth. The aim of this is to remove any native oxide (expected to be around 1 nm thick) that had formed on the opening areas of the masked substrate when transferring the samples into the MBE chamber. In addition, this step helps removing any other contaminants and moisture from the substrate surface. The growth is initiated by opening the respective shutters for the different materials. Once the III-V material reaches the substrate, it diffuses across the SiO₂ until it preferentially bonds to the exposed Si substrate where the nanowire sites have been fabricated. This is due to its monocrystalline structure relative to the thermally grown SiO₂.

The InAs nanowire growth was optimised by only adjusting the wafer's temperature during the growth process. It was found that the optimum growth rate for the nanowires was achieved in a small temperature window. Nanowires of only a few hundred nanometres would require thinner resists for contacts to be fabricated, which is challenging using UV lithography. On the other hand, long nanowires of a few microns that enable simpler fabrication, requiring much thicker resists, but this length reduces light confinement in the vertical direction and causes waveguide losses. The first growth attempt (sample number 601) was completed by growing the InAs nanowires with a substrate temperature of 560 °C, with the corresponding results is shown in Figure 5.8. It is easy to observe that the grown nanowires are relatively short. The lengths of 50 nanowires were measured from the micrograph and a histogram of the achieved lengths is shown in Figure 5.13. The measured data shows a mean length of 0.31 μ m, while the maximum and minimum recorded lengths are 0.43 μ m and 0.18 μ m respectively. The short mean length is expected to be attributed to a reduction in the diffusion length of the III-V atoms on the substrate caused by the low substrate temperature, which reduces the formation of long nanowires. The micrographs show nanoparticles of InAs seeding throughout the whole substrate including on the SiO₂ layer. Reducing the substrate temperature might increase the chance of seeding

of deposited atoms on the SiO_2 layer which would otherwise not bond if their kinetic energy is higher at higher wafer temperatures.

By increasing the wafer's growth temperature to 570 °C (501 °C actual), longer nanowires were grown using a similar growth procedure (sample number 576), with a large reduction in seeding of nanoparticles on non-patterned regions of SiO₂, see Figure 5.9. This is a clear indication that the nanoparticles formed on the surface, as shown in Figure 5.8, are constituted of the growth material rather than being external contaminants or deformation of the SiO₂ layer into nanoparticles shaped structures. The lengths of 50 nanowires were similarly measured using image analysis techniques. The blue histogram shown in Figure 5.13 shows the lengths distribution of the nanowires. The mean is measured to be 0.98 μ m, with the maximum and minimum length recorded as 1.65 μ m and 0.39 μ m respectively. In comparison with previous results, this length range allows easier contact fabrication for biasing nanowire based photodetectors. A second attempt was made to repeat the nanowire growth results achieved at 570 °C. It can be seen from Figure 5.10 that the nanowire growth takes the square array pattern of the etched SiO_2 hole nanowire sites. The micrograph shows that the achieved growth success of the nanowires can be over 70%. From Figure 5.13, the centre of the distribution was found to be approximately 0.87 μ m, with maximum and minimum recorded lengths of 1.22 μ m and 0.34 μ m respectively. The larger nanowire lengths achieved at higher temperatures is a result of the higher migration mobility of indium atoms over the substrate before bonding to a nanowire. Arsenic is not expected to migrate in a similar way to In, but desorb from the substrate without bonding to it. This is the reason that higher flow rates were used for arsenic in comparison to In. The same applies with growing GaAs nanowires as will be shown later. This should change when the wires are grown on a much less dense pattern. It is clear to see that the oxide disrupts the nucleation process of III-Vs at 570 °C, this is proved by the lack of any seeding on the non-patterned regions. This is in contrast to Si, which can form strong bonding, particularly with group III atoms.

Attempts were also made to grow nanowires at higher temperatures. Increasing the wafer growth temperature to 580 °C and 590 °C (501 °C and 516 °C actual temperatures respectively) was found to allow clean seeding in the hole sites, however, thermal desorption of As from the silicon substrate reduce

Chapter 5 - Deterministic Positioning of III-V Nanowires on Silicon for Photodetector Applications



Figure 5.8: Micrograph results from MBE growth of InAs nanowires with a substrate temperature of 560 °C, grown for 1 hour and 20 minutes, showing short nanowires (red circles) and large InAs particles forming throughout the substrate.



Figure 5.9: (a) A tilted micrograph image at 70° showing results from MBE growth of InAs nanowires with a substrate temperature of 570 °C grown for 1 hour and 20 minutes, showing nanowire lengths closer to the desired value of approximately 1 μ m with fewer scattered nanoparticles (b) top view showing the formation of nanowires and nanoparticles on the square array of nanowires sites. The bright spots represent nanowires while darker spots represent vacant sites.



Figure 5.10: A second of InAs nanowires sample grown using a similar substrate growth temperature of 570 °C to the sample shown in the Figure 5.9 and grown in the same system, by Thompson et al. from reference 20. (a) A tilted image at 30°, showing nanowire formations with lengths over 1 μ m. (b) Top view image of the square array of nanowires showing the high growth ratio of nanowires on the patterned sites, shown as bright dots.

Chapter 5 - Deterministic Positioning of III-V Nanowires on Silicon for Photodetector Applications



Figure 5.11: Micrograph results from an attempt to grow InAs nanowires at 580 °C for 1 hour and 20 minutes. The figure shows the failure of nanowires formation, and the presence of Indium nanoparticles formed in the nanowire sites with a very clean substrate surface in unpatterned regions. The high temperature is likely to have largely reduced random seeding on unpatterned SiO₂ regions.



Figure 5.12: Micrograph images from an attempt to grow InAs nanowires at 590 °C showing similar cluster formation resulted in from growing at 580 °C, the growth duration was remained constant for 1 hour and 20 minutes.

the abundance of the material on the surface which causes failure in the formation of the nanowires, see Figures 5.11 and 5.12. The high temperature also reduces the $In-SiO_2$ bonding strength leading to almost no formation of nanowires on the un-patterned oxide regions, in comparison to small diffusion lengths of Ga atoms above the substrate at low growth temperatures.

Interestingly, it can be observed from Figure 5.9 (b) that the bright spots of the square array represent the successful growth of vertical nanowires, while the relatively darker spots are the unsuccessful formation of nanowires and the formation of nanoparticles. The physical reason behind the brightness difference between the two can be explained by the regular arrangement of the crystal in the nanowires,

which reflects more electrons than the irregular crystal arrangement of the nanoparticles. This irregular crystal arrangement makes them exhibit random shapes. Figure 5.14 (a-c) show close up views of the seeded nanowires in the holes with diameters of 300 nm. Almost all the nanowires were found to seed and grow near the circumferences of the holes, which is likely to take place over the remains of SiO_2 that appear after wet etching as was previously shown in Figure 5.7.

Furthermore, two nanowires can occasionally grow within a single hole when the diameter is large enough, this can be seen in Figure 5.14 (c). From analysing electron micrographs, the length of the grown nanowires is found to be dependent on the spatial density of the holes, the difference can be noted by comparing Figure 5.14 (a-c) with Figure 5.14 (d) and/or Figure 5.15 (a). Longer nanowires were found to grow as the density of the array of holes was decreased. This is due to the proximity effect where the largely spaced nanowires collect a greater fraction of precursor material diffused from larger surrounding areas of the substrate^{26,27}. When the fabricated sites have a high spatial density, materials that migrate from the surrounding areas are shared with a larger number of sites, hence reducing the nanowire length per site. The hexagonal shape of the grown nanowires is evident, as shown in Figure 5.15 (b) and Figure 5.16. However, the hexagonal shape symmetry is not preserved for all the nanowires throughout the substrate, it is expected that this is due to different growth rates of the different facets, which is likely to be related to the shape of the nucleation centre.

Hexagonal GaAs nanowires, sample QA618, were grown in a similar fashion on Si with a substrate growth temperature of 750 °C (corresponding to 631 °C actual temperature, approximated using the oxide desorption temperature of a n-type GaAs substrate and the GaAs transition growth temperature), following the method proposed by Trukhin et al²⁸. Previous studies have also reported growth of GaAs nanowires using other methods such as nanoimprint lithography, e-beam lithography and lithography free techniques²⁹⁻³¹. The reported growth temperatures and rates parameters are close to our used parameters, except the longer time duration which was grown in this work, which produced longer wires. The aim for realising long nanowires is made primarily for easy fabrication of nanowire-based photodetectors in the future. The higher band gap of GaAs allows photodetection at smaller wavelengths, closer to the visible range. The same mask that was used previously in growing InAs

nanowires, was used for fabricating sites for GaAs nanowires. The lengths of the grown nanowires were found to be larger than those for the previously grown InAs nanowires, for example the red circled nanowire shown in Figure 5.17 (a) has a length of approximately 4.85 μ m compared to 1 μ m for the one in Figure 5.9. Proximity effects were also influential on the nanowires length, for example, the nanowire shown in Figure 5.18 has a length of approximately 4.85 μ m, longer than those shown in Figure 5.17 (b) that have a length of 4.22 μ m. This is due to the lower



Figure 5.13: Length distribution of approximately 50 InAs nanowires measured from the samples in Figure 5.8 (red chart), Figure 5.9 (blue chart) and Figure 5.10 (green chart) respectively. The mean length of the nanowires from each sample was found to be approximately 0.31 μ m, 0.98 μ m and 0.87 μ m. The growth of short wires is mainly due to low growth wafer temperature.



Figure 5.14: A selection of InAs micrographs grown at 570 °C, grown for 1 hour and 20 minutes, showing preferential spatial growth of InAs nanowires on a grid array of holes with varying diameters, without much seeding on the non-patterned oxide regions (a-d). When the nanowire sites are large in diameter, more than one nanowire can grow adjacent to each other as shown in (c).



Figure 5.15: (a) Deterministic growth of InAs nanowires in the patterned hole sites showing long nanowires, relative to those in Figure 5.14 (a-c) due to large spatial distribution between the sites. (b) The hexagonal shape of a typical InAs nanowire formed on Si. The growth temperature of the wires was 570 °C. The growth duration was remained constant for all the samples for 1 hour and 20 minutes.



Figure 5.16: Growth results of InAs nanowires grown at 570 °C for 1 hour and 20 minutes, showing over 60% growth ratio in the fabricated sites square array. The figures show that the grown nanowires exhibit varying hexagonal geometries.



Figure 5.17: (a) Growth results of the QA618 GaAs nanowires sample showing a 10×10 square array of hole sites separated by 5µm. (b) Growth results of GaAs nanowires grown on a 10×10 square array of holes sites separated by 1µm. The figure shows the difference in the grown nanowires lengths for the two arrays due to the different arrays densities.



Figure 5.18: A close up view of a single GaAs (sample QA618) nanowire with a length of 4.85 μ m showing cylindrical formation of nanowires for GaAs in comparison to the previously shown hexagonal nanowires for InAs.



Figure 5.19: Length distribution of 50 GaAs nanowires measured from the sample shown in Figure 5.17. The distribution shows most nanowires grew to a length of 6 μ m, with the maximum length from this batch being 6.27 μ m. The figure also shows how the nanowire's length distribution tails down for shorter lengths. The minimum length was measured to be 3.24 μ m.



Figure 5.20: µPL measurements at 20 K of an ensemble of nanowires grown on a square array of nanowires sites separated by 300 nm showing a broad emission spectrum from 825 nm to over 1000 nm with sharp features in the spectra indicating nanowire confinement emission.



Figure 5.21: μ PL measurement spectrum of an individual GaAs nanowire taken at 20 K. The spectrum shows a sharp spike from a single nanowire, possibly due to an impurity or defect in the lattice. The single nanowire shown here is grown in an array of sites fabricated with a separation of 5 μ m. The large separation is made to ensure exciting only a single nanowire with the 1 μ m laser spot size in the μ -PL setup.

spatial density of the nanowires sites, making each site share larger amount of material with nearby neighbouring sites in comparison to lower density arrays. Figure 5.19 shows a histogram for 50 GaAs nanowires measured from different micrographs. It is easy to observe that much larger lengths were achieved using GaAs at a similar growth temperature. The largest portion of the nanowires were found to have lengths between 5.60 μ m and 6.30 μ m, where 6.30 μ m was also the maximum length. From this histogram, it can be seen that shorter nanowires can be found, with the smallest recorded one being 3.24 μ m. It was also noted that the growth of GaAs nanowires was not always vertical. A large proportion of the nanowires were found to be grown slanted at a sharp angle relative to the substrate. Interestingly, by analysing the figures, the angle distribution was found to be small, which could mean that this angle is related to the crystal's facet orientation, making the nanowire's grow at an angle, in this case aligned with the (111) plane.

Preliminary micro-photoluminescence measurements were carried out for both an ensemble of GaAs nanowires and individual ones at 20 K, as a first step toward fabricating p-i-n structures for photodetection applications. The central emission peak obtained from the ensemble's μ -PL measurement show a central emission peak at approximately 868 nm as shown in Figure 5.20. The central wavelength of the main peak corresponds to the bandgap of GaAs. A long tail for the central

peak was also found to extend to 1 μ m. Power dependence measurements on a nanowire ensemble have shown red shifting in the central emission wavelength for the peak as the power is reduced. Growing nanowires in a largely spaced grid allows μ -PL of individual nanowires to study their confinement characteristics. The μ -PL spectrum of the nanowire circled in Figure 5.17 (a) is shown in Figure 5.21. The spectrum shows a sharp spike with a full width half maximum of 0.8 nm at 851 nm, which could correspond to quantum-confined states in the nanowire.

Realisation of photodetectors using InAs-based p-i-n structures has been performed recently and is shown in reference 20. Presently, work is being carried out toward growing similar p-i-n structures in GaAs for detecting lower wavelengths. Following this, fabrication of front and back gold contacts on the free-standing nanowires will be carried out.

5.4. Conclusion

In summary, the work done in this chapter involved investigating a potential technique of enhancing single photon detection using nanowire based photodetectors. Results from site-controlled catalyst-free growth of III-V (InAs and GaAs) nanowires on Si substrates is shown and discussed. To selectively grow nanowires spatially, e-beam lithography of nanowire hole sites was carried out to preferentially grow nanowire arrays on a predefined area. The growth of III-V nanowires in this chapter was done on a p-type Si substrate, solving a huge challenge in the integration of III-V optoelectronic technology on the common Si platform. InAs and GaAs nanowires for mid- and near-infrared operating wavelengths were subsequently grown on the fabricated sites. Results show that the growth temperature is vital for optimum growth of InAs nanowires. Growing at lower temperatures may lead to the formation of short nanowires due to reduced diffusion length of deposited atoms on the substrate while growing at higher temperatures may reduce the abundance of V material due to its lower evaporation temperature. Initial micro-photoluminescence measurements show sharp signatures from an ensemble of nanowires as well as individual ones.
Bibliography

¹ W. T. Tsang. Semiconductors and semimetals. Part D photodetectors. Academic Press, ISBN 978-0-12-752153-4 (1985)

² J. C. Campbell. Recent advances in telecommunications avalanche photodiodes. Journal of Lightwave Technology, 25, 109-121 (2007)

³ N. Han, F. Y. Wang, J. J. Hou, S. P. Yip, H. Lin, F. Xiu, M. Fang, Z. X. Yang, X. L. Shi, G. F. Dong, T. F. Hung, J. C. Ho. Tunable electronic transport properties of metal-cluster-decorated III-V nanowire transistors. Advanced Materials Communication, 25, 4445-4451 (2013)

⁴ T. Takahashi, K. Takei, E. Adabi, Z. Y. Fan, A. M. Niknejad, A. Javey. Parallel array InAs nanowire transistors for mechanically bendable, ultrahigh frequency electronics. ACS Nano, 4, 5855-5860 (2010)

⁵ A. C. Ford, J. C. Ho, Y. L. Chueh, Y. C. Tseng, Z. Y. Fan, J. Guo, J. Bokor, A. Javey. Diameterdependent electron mobility of InAs nanowires. Nano Lett., 9, 360-365 (2009)

⁶ M. S. Gudikssen, L. K. Lauhon, J. F. Wang, D. C. Smith, C. M. Lieber. Growth of nanowire superlattice structures for nanoscale photonics and electronics. Nature, 415, 617-620 (2002)

⁷ Y. J. Dong, B. Z. Tian, T. J. Kempa, C. M. Lieber. Coaxial group III-nitride nanowire photovoltaics. Nano Lett., 9, 2183-2186 (2009)

⁸ C. Thelander, L. E. Froberg, C. Rehnstedt, K. Samuelson, L. E. Wernersson. Vertical enhancementmode InAs nanowire field-effect transistor with 50-nm wrap gate. IEEE Electronic Device Lett., 29-206-208 (2008)

⁹ H. Xia, Z. Y. Lu, T. X. Li, P. Parkinson, Z. M. Liao, F. H. Liu, W. Lu, W. D. Hu, P. P. Chen, H. Y. Xu, J. Zou, C. Jagadish. Distinct photocurrent response of individual GaAs nanowires induced by n-type doping. ACS Nano, 6, 6005-6013 (2012)

¹⁰ R. E. Wahl, F. Y. Wang, H. E. Chung, G. R. Kunnen, S. P. Yip, E. H. Lee, E. Y. B. Pun, G. B. Raupp, D. R. Allee, J. C. Ho. Stability and low-frequency noise in InAs NW parallel array thin-film transistors. IEEE Electron Device Lett., 34, 765-767 (2013)

¹¹ L. Rogalski. Infrared detectors: status and trends. Prog. Quant. Electron., 27, 59-210 (2003)

¹² L. Becker. Current and future trends in infrared focal plane array technology. Proc. SPIE, 5881, 588105 (2005)

¹³ T. Martensson, C. P. T. Svensson, B. A. Wacaser, M. W. Larsson, W. Seifert, K. Deppert, A. Gustafsson, L. R. Wallenberg, L. Samuelson. Epitaxial III-V nanowires on silicon. Nano Lett., 4, 1987-1990 (2004)

¹⁴ M. Paladugu, C. Merckling, R. Loo, O. Richard, H. Bender, J. Dekoster, W. Vandervorst, M. Caymax, M. Heyns. Site selective integration of III-V materials on Si for nanoscale logic and photonic devices. Crys. Growth Des., 12, 4696-4702 (2012)

¹⁵ R. S. Chen, H. Y. Chen, C. Y. Lu, K. H. Chen, C. P. Chen, L. C. Chen, Y. J. Yang. Ultrahigh photocurrent gain in m-axial GaN nanowires. Appl. Phys. Lett., 91, 223106 (2007)

¹⁶ J. Miao, W. Hu, N. Guo, Z. Lu, X. Zou, L. Liao, S. Shi, P. Chen, Z. Fan, J. C. Ho, T. Li, X. S. Chen, W. Lu. Single InAs nanowire room-temperature near infrared photodetectors. ACS Nano, 8, 3628-3635 (2014)

¹⁷ H. W. Shin, S. J. Lee, D. G. Kim, M. Bae, J. Heo, K. J. Choi, W. J. Choi, J. Choe, J. C. Shin. Shortwavelength infrared photodetector on Si employing strain-induced growth of very tall InAs nanowire arrays. Sci. Rep., 5, 10764 (2015)

¹⁸ E. M. Gallo, G. N. Chen, M. Currie, T. McGuckin, P. Prete, N. Lovergine, B. Nabet, J. E. Spainer. Picosecond response times in GaAs/AlGaAs core/shell nanowire-based photodetectors. Appl. Phys. Lett., 98, 241113 (2011)

¹⁹ X. C. Jiang, Q. H. Xiong, S. Nam, F. Qian, Y. Li, C. M. Lieber. InAs/InP radial nanowire heterostructures as high electron mobility devices. Nano Lett., 7, 3214-3218 (2007)

²⁰ M. D. Thompson, A. Alhodaib, A. P. Craig, A. Robson, A. Aziz, A. Krier, J. Svensson, L. Wernersson, A. M. Sanchez, A. R. J. Marshall. Low leakage-current InAsSb nanowire photodetectors on silicon. Nano Lett., 16, 182-187 (2016)

²¹ M. J. L. Sourribes, I. Isakov, M. Panfilova, H. Liu, P. A. Warburton. Mobility enhancement by Sbmediated minimisation of stacking fault density in InAs nanowires grown on silicon. Nano Lett., 145, 1643-1650 (2014)

²² M. D. Kelzenberg, D. B. Turner-Evans, B. M. Kayes, M. A. Filler, M. C. Putnam, N. S. Lewis, H. A. Atwater. Photovoltaics measurements in single-nanowire silicon solar cells. Nano Lett., 8, 710-714 (2008)

²³ Y. Cui, Z. Zhong, D. Wang, W. U. Wang, C. M. Lieber. High performance silicon nanowire field effect transistors. Nano Lett., 3, 149-152 (2003)

²⁴ D. Wang, B. A. Sheriff, J. R. Heath. Silicon P-FETs from ultrahigh density nanowire arrays. Nano Lett., 6, 1096-1100 (2006)

²⁵ M. D. Kelzenberg, S. W. Boettcher, J. A. Petykiewicz, et al.. Enhanced absorption and carrier collection in Si wire arrays for photovoltaic applications. Nature Mater., 9, 239-244 (2010)

²⁶ H. J. Joyce, Q. Gao, H. H. Tan, C. Jagadish, Y. Kim, J. Zou, L. M. Smith, H. E. Jackson, J. M. Yarrison-Rice, P. Parkinson, M. B. Johnston. III-V semiconductor nanowires for optoelectronic device applications. Progress in Quantum Electronics, 35, 23-75 (2011)

²⁷ M. H. Madsen, M. Aagesen, P. Krogstrup, C. Sørensen, J. Nygård. Influence of the oxide layer for growth of self-assisted InAs nanowires on Si(111). Nanoscale Res. Lett., 6, 516 (2011)

²⁸ V. N. Trukhin, A. S. Buyskikh, N. A. Kaliteevskaya, A. D. Bourauleuv, L. L. Samoilov, Y. B. Samsonenko, G. E. Cirlin, M. A. Kaliteevski, A. J. Gallant. Terahertz generation by GaAs nanowires. Appl. Phys. Lett., 103, 072108 (2013)

²⁹ A. M. Munshi, D. L. Dheeraj, V. T. Fauske, D. C. Kim, J. Huh, J. F. Reinertsen, L. Ahtapodov, K. D. Lee, B. Heidari, A. T. J. Helvoort, B. O. Fimland, H. Weman. Position-controlled uniform GaAs nanowires on silicon using nanoimprint lithography. Nano Lett., 14, 960-966 (2014)

³⁰ S. Plissard, G. Larrieu, X. Wallart, P. Caroff. High yield of self-catalyzed GaAs nanowire arrays grown on silicon via gallium droplet positioning. Nanotechnology, 22, 275602 (2011)

³¹ E. Koivusalo, T. Hakkarainen, M. Guina. Structural investigation of uniform ensembles of selfcatalyzed GaAs nanowires fabricated by lithography-free technique. Nanoscale Research Letters, 12:192 (2017)

Chapter 6

A Quantum Key Distribution Chip Design

6.1. Introduction

QKD has recently attracted great research attention for the potential it offers in securing everyday digital communications¹⁻³. In the present day, the largest obstacle that prevents QKD systems from inhabiting many consumer grade electronics is their large size and cost. This makes QKD used solely by organisations, banks and governments that have large financial resources. This chapter will introduce a design that is aimed at miniaturizing QKD systems to the chip level, thereby paving the way towards integrating QKD in handheld devices and embedded systems for securing their communications.

In this chapter, current research into integrated optical components, such as cavities, waveguides and out-couplers will be introduced. A discussion will follow to compare the different types of integrated optical components, and the most practical of these will be used for proposing a full system. Numerical simulations of these components and design optimisations will be performed to improve many of their aspects, such as increasing their coupling efficiencies and reducing their losses. Finally, a full QKD system made of the optimised optical components will be presented as a strong candidate for future integrated QKD systems.

6.1.1. Motivation

Secure key sharing with QKD was explained in chapter 1 using the polarisation degree of freedom for encoding quantum information on photons⁴. However, these qubits can also be encoded with the phase difference between successive photons pulses. Realising a QKD system by encoding 1s and 0s on the phase difference between photon pulses is known as Differential Phase Shift (DPS) QKD⁵. Practically, DPS-QKD is much less susceptible to environmental fluctuations, relative to polarisation encoding. In addition, while integrated polarisation rotators have been introduced before, integrated phase modulators have been extensively studied and have shown great promise for optical digital communications⁶⁻⁸.



Figure 6.1: A block diagram of a double Mach-Zehnder interferometer DPS-QKD system. Successive pulses from the single photon source (component A) on Alice's side are phase modulated (using component C) and interfered before being out-coupled (using component D) from the chip. Bob performs similar interferometric measurements using a similar setup (Components BCB). Interfered photons are then recorded by detectors 1 or 0, depending on their phase difference.

A state-of-the-art integrated DPS-QKD system was demonstrated by Sibson et al⁹. The system benefits from the already established Si optoelectronic technology, using an attenuated laser source that operates at telecom wavelengths. However, since QKD is prone to the photon number splitting attacks when a Poisson source is used, the average number of photons per pulse is largely reduced, hence reducing the achievable bit rate¹⁰. This makes an alternative method that involves a high bit rate, integrated source of indistinguishable photons necessary for fast and perfectly secure QKD systems. In addition, the QKD system demonstrated in reference 9 requires the complicated growth of structures with multiple lithography processes to realise the full system including the laser source, the waveguides and the phase modulator. In this thesis, a much simpler system that requires less complicated growth structures that can be fabricated with a single lithography process based on PhCs will be suggested. Using a PhC platform based on GaAs, shorter phase modulators can be fabricated leading to further miniaturization of the technology.

The different components A, B, C and D shown in Figure 6.1 that are being used as parts of a typical DPS-QKD system will be introduced and discussed. PhC cavities will be suggested and optimised as a potential platform for realising single photon sources. For part B of the system, a new beam splitting scheme in which light couples to two waveguides following emission from the cavity will be simulated and the cavity-waveguide coupling efficiency will be enhanced for reducing losses. Different methods in realising part C of the system will be discussed. Finally, as part of improving part D of the system, current techniques for light out-coupling will be explained and numerical modelling will be performed for optimised coupling between a PhC waveguide and a Bragg mirror out-coupler.

6.1.2. GaAs as an Optoelectronic Material for QKD

One of the most critical components in a secure QKD system is a pure single photon source¹⁰. Many of the requirements of an ideal single photon source are discussed in chapter 4. While ideal single photon sources are still far from being commercially available, InAs quantum dots (QDs) are one of the most promising sources of single photons¹¹. The drawback that InAs QDs suffer from which makes them impractical for commercial applications is their operation temperature. However, research into single photon sources based on InAs for quantum information applications has shown that such systems have many advantages over others, including the operation wavelength, photon indistinguishability, short lifetime and compatibility with optoelectronics materials such as GaAs^{12,13}. These advantages are the main driving motivation towards achieving a secure GaAs chip-based QKD system.

There has been a great debate over the years about which is the most ideal material for optoelectronic applications. The top two that were debated for were Silicon and Gallium Arsenide. Compared to GaAs, Silicon is one of the most abundant materials on earth and compatible with current industrial device fabrication processing. In terms of its optical properties, Si's indirect bandgap means it is an inefficient light absorber and emitter and renders it of little use in most optoelectronic applications. On the other hand, the direct bandgap of GaAs and InP means they emit and absorb light efficiently at different wavelengths¹⁴. In contrast to InP, GaAs has been studied extensively and has a lattice match with alloys

such as AlGaAs and Al₂O₃ that are widely involved in many GaAs-based optoelectronic applications for creating tunnel barriers, laser diodes and photodetector heterostructures.

For integrated optical components, it is desirable to operate at a wavelength where there is minimum absorption. Due to GaAs's direct bandgap, its absorption reduces to virtually zero at wavelengths above 870 nm, while Silicon absorption is two orders of magnitudes higher at that wavelength. It is known that the wavelength emission range of InAs QDs is centred at 915 nm and spans a width of approximately 60 nm. Figure 6.2 show a typical spectrum obtained from self-assembled InAs QDs at 20 K. In addition, operating in this wavelength range makes QKD systems compatible with the commercially available single photon detectors that normally have maximum quantum efficiency in this range¹⁵. Finally, the property that both GaAs and InAs possess is that they are a III-V semiconductor, this makes GaAs an ideal environment material for the epitaxial growth of InAs QDs due to matching structural lattices. Hence, fabricating an integrated QKD chip that harnesses the single photon emission property of InAs should be done on GaAs.



Figure 6.2: A typical micro-photoluminescence spectrum recorded for an InAs QD ensemble at 20 K and 10 μ W laser power. The figure shows clear signatures of sharp emission lines due to the QD exciton confinement with a possible signature of biexciton lines at around 935 nm.

Researchers have previously demonstrated single photon emission from InAs QDs capped with GaAs¹⁶. Much of this work was motivated by proposed applications in quantum computing and quantum communications. Efforts have also been made toward engineering photonic structures such as optical cavities and waveguides around InAs QDs¹⁶, mainly to manipulate the quantum states that emitted photons carry. As a result, on chip Purcell enhancement in QDs¹⁷, single photon guiding and dipole induced transparency¹⁸ have all been reported. This was made possible by the developments of lithography techniques such as electron-beam (e-beam) lithography and dry etching techniques on GaAs. Observing the progress and extensive studies being made on InAs QDs using a GaAs substrate was one of the key motivational forces toward designing a chip-based QKD system. The current aim is epitaxial integration of all the optical components that assemble a QKD system into a single chip, preferably including integration of GaAs and other III-V unique optoelectronic materials with conventional Si technology.

6.2. Components for QKD

In this section of the chapter, an introduction to the main components needed for QKD will be given and current techniques to realise these components will be summarised. A comparison will be made between the different techniques needed to realise every component and an option will be favoured based on the goal of achieving a chip-based QKD system.

6.2.1. Single Photon Cavity for QKD

Over the years, different GaAs cavities have been proposed and constructed¹⁹. The ultimate aim is to improve light-matter coupling by confining the light's electric and magnetic field components inside a structure where a two-level light emitter exists such as a QD^{20-22} , an $atom^{23,24}$, a nitrogen vacancy centre in diamond²⁵, etc. This coupling increases the emitter's absorption and spontaneous emission rate.



Figure 6.3: Schematic illustration of different proposed cavity designs for enhancing the spontaneous emission rate from QDs. The figure shows a common (a) micro-pillar (b) micro-disc and (c) L3 PhC cavity. The blue flare shown in (a) represents the direction of light travel, while those in (b) are a visual representation of the whispering gallery mode in the micro-disc.

One design is known as a pillar micro-cavity¹³. In this cavity, the light is trapped within the vertical height of a GaAs pillar which has a diameter on the order of one cavity mode wavelength. Along the vertical height of the cavity, a distributed Bragg mirror is grown epitaxially to trap the light in the active region of the cavity which includes the quantum emitter. Previous work has shown Purcell enhancement from coupling an InAs QD situated in the centre of the cavity to the cavity mode¹³. The number of Bragg mirror layers grown above the cavity is slightly smaller than below to allow the cavity to preferentially radiate light modes vertically upward, as shown by the blue flares in Figure 6.3 (a). The distributed Bragg structure could be considered as a 1D PhC that can confine the light due to the photonic band in the x-z and y-z planes of the pillar. Light is confined laterally in the x-y plane due to total internal reflection at the GaAs-air interface of the pillar.

A second type of cavity constitutes another class of resonator where the light is trapped in the x-y plane due to a Whispering Gallery mode at the rim of a GaAs disk²⁶, as in Figure 6.3 (b). Finally, another GaAs cavity design is based on a 2D-bridge type PhC, as in Figure 6.3 (c)¹⁷. In this type of cavities, the light is confined in the x-y plane due to the photonic bandgap and confined in the z-plane due to total internal reflection at the GaAs-air interfaces at the top and bottom. More of the basics of PhCs and related references can be found in chapter 2.

Each of these cavity types have advantages over the other. In terms of the quality factor, it is known that whispering gallery modes in a micro-disc are usually higher than those achieved in micro-pillars and PhC cavities¹⁹. The highest Q-factor achieved experimentally in coupling whispering gallery modes to InAs QDs was reported by Srinivasan et al.¹⁹ to be 4×10^5 . The highest Q-factors achieved in

coupling GaAs bridge-type PhCs and micro-pillar cavities to InAs QDs were reported by Hennessy et al. and Loo et al. as 3×10^5 and 6×10^4 , respectively²⁶⁻²⁸. Due to the nature of the whispering gallery mode, the mode volume for micro-disc cavities is usually much higher than those of a bridge-type PhC and micro-pillar cavities. The latter usually has the smallest achievable mode volume of $\frac{1}{10} \left(\frac{\lambda}{n}\right)^3$, while being on the order of $\left(\frac{\lambda}{n}\right)^3$ and $10 \left(\frac{\lambda}{n}\right)^3$ for bridge-type PhCs and micro-disc cavities.

Fabrication wise, it is widely known that PhC cavities are more prone to fabrication imperfections compared to micro-disc and micro-pillar cavities. Surface roughness of the bridge and/or the etching profile of the fabricated holes can reduce its Q-factor dramatically and affect its mode volume. Nonetheless, decent results have been reported over the past few years, and the availability of many of the fabrication tools required make it a routinely achievable challenge.

For QKD applications where photon manipulation and processing is carried out on a chip, coupling light efficiently from a cavity is critical. Previously, coupling of light from a micro-disc cavity was carried out using a single mode fibre optic spatially positioned within a few hundred nanometres from the rim of the micro-disc²⁹. This coupling process requires precise positioning of the fibre optic making the setup extremely sensitive to vibrations, hence it is unsuitable for mass manufacturing. Meanwhile, due to light being emitted from a micro-pillar vertically, coupling light back to the chip for processing using mirrors or a focusing objective is also sensitive to optical alignments and could introduce losses. Coupling a PhC cavity to a PhC waveguide can be easily achieved by engineering the photonic band structure of the crystal. PhC cavity-waveguide coupling can be easily simulated and different designs can be reliably fabricated and tested using e-beam lithography and Inductively Coupled Plasma (ICP) etching on GaAs³⁰. The decent Q-factors achievable with the medium mode volume and efficient coupling that PhC cavities can offer makes it an ideal choice for integrated QKD systems.

6.2.2. Integrated Waveguides for QKD

Integrated waveguides for on chip photonic interconnects divide into two common types. The first is a ridge waveguide in which light is confined in the centre core of the guiding layer due to the index contrast that surrounds the guiding layer, as shown in Figure 6.4 (a)^{6.31}. Ridge waveguides are more

common in Silicon and Indium Phosphide (InP) photonic technologies since the surrounding material that provides the index contrast can be SiO_2 . The second kind of integrated waveguides is a bridge-type PhC waveguide, as shown in Figure 6.4 (b)¹⁸. This type constitutes a row of missing holes from the PhC lattice, opening a confined state within the photonic bandgap that can be used as a single mode waveguide. Similar to PhC cavities, light in PhC waveguides is confined in the x-y plane due to the photonic bandgap and in the z direction due to total internal reflection of the bridge structure.



Figure 6.4: Schematic illustrations of a ridge (a) and PhC (b) waveguides. The ridge waveguide shown in the figure is made of a material of different refractive index to the substrate material for achieving index contrast for light confinement.

PhC waveguides have less losses compared to most forms of ridge waveguide in principle, however, due to the high precision requirement in fabricating PhC waveguides, the reported experimental losses are usually higher³². Previous work has showed minima of 1.8 db/cm losses for PhC waveguides and 0.7db/cm for ridge waveguides^{33,34}. However, most of these measurements are made at telecom wavelengths. If measurements were made where GaAs is two orders of magnitude less absorbent than Silicon, such as that of the emission wavelength of InAs QDs, loss results from GaAs can be much smaller.

It is necessary for the chosen waveguide used in a DPS-QKD chip to have a model in which phase modulation can be carried out. Differential phase shifting in QKD can be carried out by applying an electric field across one arm of a Mach-Zehnder interferometer. The amount of phase modulation is dependent on the material the photons are guided through³⁵. For transverse modulation the change in phase can be calculated using the equation below:

$$\Delta \phi = \frac{2\pi}{\lambda} r_{41} \frac{\mathrm{V}}{\mathrm{d}} L$$

where r₄₁ is the 4-1 component of the electro-optic matrix for GaAs, E is the electric field applied across the waveguide, d and L are the width and length of the waveguide respectively. The same applies for Silicon, however, different tensor components should be taken into account for different guided modes (TE or TM) because the electro-optic modulation is different when the electric field is applied at different orientations.

The waveguide's length is important here to ensure π phase modulations are performed as the photons travel through the waveguide, without applying high voltages and facing heating issues. Applying a high voltage per pulse can also be difficult to generate electronically. Furthermore electron tunnelling and/or arcing might occur in this small length scale. These issues become even more prominent when operating at high operation frequencies for fast QKD bit rates. Compared to ridge waveguides, PhC waveguides can play a vital role in reducing the length of the waveguide by harnessing slow light effects. Reduction in the group velocity of light due to slow light effects can shorten the waveguide by an order of magnitude while achieving the same phase modulation using similar applied voltages. The symmetric strong electro-optic modulation and low absorption of GaAs at the InAs emission wavelength, combined with the slow light effect of PhCs makes the two ideal for integrated phase modulation.

Recently, researchers have proposed and tested a scheme for PhC phase modulators. In this scheme, pi-n doped regions are made across a PhC waveguide to provide a strong electric field for electro-optic modulation³⁶. The waveguide is fabricated with e-beam lithography, dry and wet etching, and doped using ion-implantation. It is important to take doping's effect on the refractive index into account when designing PhC phase modulators. Changes in the doping between the n and p regions can require modification to the symmetry of the crystal in the y-direction to compensate for this. To compensate for the change in index in regions surrounding the waveguide, the photonic crystal lattice may be altered by modifying the radii of the holes in these regions or modifying their lattice constant.



Figure 6.5: The proposed phase modulation scheme used in the QKD system. The schematic shows the different doping regions needed to allow phase modulation to guided modes in the PhC waveguide.

6.2.3. Photon Out-Coupling for a QKD Chip

Coupling light efficiently out of a chip for carrying out chip to chip communications, for example, is important for reducing photon losses and increasing key bit rates in a QKD system. There have been a few different designs suggested for light out-coupling, such as using a Bragg mirror based or tapered mode out-coupler, as shown in Figure 6.6.

The difference between the Bragg and tapered mode out-couplers is that the first directs light in the vertical direction out of the chip, while the second can only emit the light in a parallel direction to the chip. Although practically, it is much easier to collect the light from above the chip using an objective lens or a lensed fibre aligned above the out-coupler, it is still challenging to perform coupling without precise alignments, therefore addressing this challenge becomes difficult industrially for consumer grade devices.



Figure 6.6: Schematic illustrations of different proposed out-of-chip light couplers showing (a) Bragg mirror and (b) tapered mode out-couplers. The train of red flares indicate the direction of light travel out of each out-coupler. The figure shows how the Bragg out-coupler can be used to direct light away from the chip, while the tapered mode out-coupler can allow directing light to adjacent photonic systems, such as photodetectors, on the same chip.

A tapered beam out-coupler has the advantage of efficient coupling of light to other optical components situated next to the device on the same chip, such as nanowire photodetectors. Due to the symmetry of the structure, using a Bragg reflector mirror means only 50% of the light travels vertically upward from the out-coupler, while 50% travels downward. However, this issue isn't a major problem as some of the light that travels downward away from the collection optics, gets reflected at the underlying air-substrate interface. The number of lost photons that travel downward can be reduced by over 99% and be redirected in the vertical direction by epitaxially growing a distributed Bragg reflector underneath the PhC bridge. In section 6.3.4, simulations will be carried out to optimise coupling between the waveguide and the out-coupler, thereby reducing reflections and scattering at the out-coupler's end.

6.3. Numerical Optimisation of QKD Components

6.3.1. Optimising Cavity Confinement for Single Photon Emission

As discussed in chapter 2, the Purcell effect and efficient light-matter coupling all depend directly on a cavity's Q-factor and inversely on the cavity's mode volume. Coupling cavities to integrated photonic circuitry for chip-scale quantum photonic applications was previously achieved in a number of



Figure 6.7: The design of an unmodified L3 cavity, indicating the direction of displacement of holes A-E. The displacement is the distance holes A-E moved from their normal position.

reports^{16,37}. Simple cavities such as the L3, H0 and H1 cavities are widespread designs due to their simplicity, compactness, high Q-factor and the small mode-volume they have, making them practical for integrated photonic circuit applications.

In designing a PhC structure for GaAs, it is important to take into account the exact refractive index that corresponds to the wavelength of interest. For GaAs, the refractive index for the central emission wavelength of 915 nm in which InAs QDs emit is approximately 3.59.

H0 and H1 have shown large Q-factors and very small mode volumes. Their small mode volume make them inefficient in waveguide coupling, which is important for an integrated QKD system. In addition, H0 cavities mode maximum is not centred in the middle of the cavity, in contrast to L3 cavities³⁸. The larger mode volumes for L3 cavities make them more attractive in such applications. Optimising cavity Q-factors can cause large Purcell enhancements, hence allowing the realisation of high bitrate single photon sources.

Optimising the Q-factor for L3 cavities can be done by changing the radii and positions of the nearby holes surrounding the cavity. These holes will be labelled A to E based on their distance to the cavity.

Figure 6.7 shows these holes and the arrows that indicate the direction of displacement, S, for the holes. This simple method has been shown to be capable of increasing the cavities Q-factor by orders of magnitude while maintaining the same mode volume³⁸. Using this technique, a series of simulations were completed to suggest different improved designs that give high Q-factors from an L3 cavity. The mode wavelengths will be presented in normalised units of the lattice constant, a = 1. For an explanation of the methods used to perform FDTD simulations of the cavities, see chapter 2.



Figure 6.8: The relationship between changing the radius of holes, A, on the cavity resonant wavelength (a) and the cavity mode's volume (b). The figure shows increasing the holes radius causes the cavity mode to blue shift. The Q-factor however, can fluctuate, hence careful selection of the holes radius is necessary for optimised Q cavities.



Figure 6.9: An x-y cross section of the simulated PhC cavity through the bridge structure. The 3D FDTD simulations of the photonic cavity mode show time slice images of the confined (a) E_x (b) E_y and (c) H_z field components within the bridge structure of the cavity. Red (blue) indicate positive (negative) components of the electric field.

Unmodified L3 Cavity

The unmodified L3 cavity is made of three missing holes from the lattice, without any modification in the positions or radii of holes A-E. A series of simulations were carried out to investigate at which radius is the maximum Q-factor obtained, before proceeding with performing small modifications to

the nearby holes for optimisation. For unmodified cavities with a refractive index matching that of GaAs at the InAs QD emission wavelength, results from Figure 6.8 show that the cavity mode's wavelength reduces proportionally to the holes radius. This is expected to be the case since the dielectric to air ratio is reduced, causing reduction in the mode's wavelength. The Q-factor is expected to vary with many factors such as the dielectric index, holes radius and cavity modifications. The Q-factor was found to fluctuate above 5600 for radius values ranging from 0.27a to 0.33a. Increasing the radius further is not expected to increase the Q-factor much more than the range shown. The same is expected to take place when the radius is reduced.

Optimised Cavity Design 1

The first optimisation was done by shifting holes A so that $S_A = 0.15a$, by following the work of Akahane et al.³⁹. The cavity mode's field distribution is shown in Figure 6.9, where the mode has a Q-factor of 4.5×10^4 at a resonance wavelength of $\lambda = 3.707a$.



Figure 6.10: The relationship between changing r_A and its effect on the resonance wavelength (a) and Q-factor (b) for an L3 cavity with $S_A=0.15$.

The first improvement to the design is done by changing the radius of holes A and measuring the corresponding achieved Q-factor. The radius of the holes were changed from $r_A = 0.10a$ to $r_A = 0.25a$, and a maximum Q-factor was achieved for holes with a radius $r_A = 0.2a$, measured as 1.3×10^5 . The resonant mode wavelength was found to blue shift as the radius of the A holes, r_A , was increased, as shown in Figure 6.10. The intuitive reasoning behind this is basically as the A holes radius is increased,

the volume of the cavity reduces, leading to smaller wavelengths confined in the structure. The resonant mode wavelength for the design with the maximum Q-factor is $\lambda = 3.886a$.

It is interesting to mention that performing simulations where $S_A = 0.15a$ with a similar r_A sweep on an H1 cavity, does not produce maximum Q-factor with $r_A = 0.2a$, as was shown previously with L3 cavities. Figure 6.11 shows results from similar simulations to the ones done previously on L3 cavities, but now on H1 cavities.



Figure 6.11: The relationship between changing r_A and its effect on the resonance wavelength (a) and Q-factor (b) for an H1 cavity with $S_A=0.15$.

Optimised Cavity Design 2

This design was based on modifications done by Minkov et al.³⁸ where the radii of holes A-E were kept constant but the hole positions were shifted so that, $S_A = 0.3370a$, $S_B = 0.2706a$, $S_C = 0.0874a$, $S_D = 0.3223a$ and $S_E = 0.1737$. These correspond to the hole shifts suggested by Minkov et al. on a PhC structure with a radius r = 0.25a for optimising the cavity Q-factor for telecom wavelengths in Silicon. However, the refractive index for Silicon at telecom wavelengths is lower than that for GaAs at the emission wavelength of InAs QDs. Investigating for which displacements S_{A-E} the is Q-factor maximised is a large parameter space and requires extensive studies, the aim of this work is providing an alternative design with a high Q-factor, while operating with InAs QD emission wavelengths. Hence, the results are expected to be different when similar simulations were done for a different index. Results from similar studies to the ones previously completed are shown in Figure 6.12. Interestingly, when similar sweeps were carried out, the maximum Q-factors were achieved for a radius of a PhC, r =

0.26*a*, whereas Minkov et al. reported a different value. This means, performing any modifications on a certain cavity with a certain slab index to improve its Q-factor does not necessary imply that the modification can be universal and work on any cavity with any effective slab index. In addition, the normalised cavity mode's wavelength and its Q will not be the same either.



Figure 6.12: The impact of changing the PhC holes radius on the resonance wavelength (a) and Q-factor (b) for an L3 cavity with $S_A = 0.3370a$, $S_B = 0.2706a$, $S_C = 0.0874a$, $S_D = 0.3223a$ and $S_E = 0.1737$.

6.3.2. Optimising Waveguide-Cavity Coupling for Efficient Beam splitting

As aforementioned, one of the main components in a DPS-QKD setup is a Mach-Zehnder interferometer. In a Mach-Zehnder interferometer, a single light beam is split into two separate channels via a beam splitter, a phase shift is then applied to one side of the split beam before the two channels are recombined, resulting in an interference effect, at the junction of a second beam splitter.

The most common method of beam splitting using a PhC platform is with a Y-shaped waveguide. However, this type of beam splitter is known to be lossy, due to the high leakage losses at the splitting junction and the 60 degree bends. In previous work, suggestions were made to modify the holes at the regions where losses are encountered to improve the Y-shaped beam splitter design⁴⁰. In this section, a different model for beam splitting will be proposed and optimised using two waveguides coupled to an L3 resonant cavity, as shown in Figure 6.13.

It is crucial to couple the light in the cavity to the two waveguides efficiently, thereby reducing losses and improving the QKD bit rate. From looking at the field distribution in Figure 6.14 (a), it is easy to notice that the electric field components of the cavity mode tend to exponentially decay away from the



Figure 6.13: The proposed method of light beam splitting at the cavity as an alternative to Y shaped PhCs beam splitters. Having the cavity oriented in this way allows the tails of the cavity mode to align with the waveguide.

cavity through the photonic structure at angles of 45, 135, 270 and 315 degrees, relative to the positive x-axis of the slice. The radiative decay at these angles can be exploited to selectively position a waveguide that the cavity can couple to efficiently. For this reason, a tilted cavity positioned next to two L waveguides is suggested for optimised coupling efficiency. Figure 6.13 shows a rendered image of the tilted cavity design modified according to cavity design 1, with two waveguides coupled to its ends for efficient light beam splitting.

In previous work by Faron et al.⁴⁰ where they demonstrated coupling tilted cavities to waveguides, the Q-factor was shown to be directly dependent on the number of holes that separate the cavity and the waveguide. Reducing the number of holes leads to higher leakage from the cavity to the waveguide, thereby reducing the cavity's Q-factor. In that work, the coupling was investigated by measuring the Q-factor ratios between that of the cavity and the waveguide. A more rigorous method to estimate this involves directly computing the power flux coupled to the waveguide in the two separate cases.

By coupling the horizontal cavity design 1 to two waveguides, where the index of the PhC's dielectric material matches that of GaAs at 915 nm, it was found that the Q-factor reduces to 3.4×10^3 . On the other hand, the Q-factor achieved by coupling the optimised tilted design 1 to two waveguides is 3.6×10^3 .

10³. By recording the flux through either of the two waveguides that are coupled to the cavity, the ratio between power fluxes transmitted from a tilted and a horizontal cavity is calculated, hence the enhancement in coupling efficiency. It is important to mention that this method of measuring the coupling efficiency enhancement can only be done when the simulation time steps for both cases is the same and the difference in the cavity Q-factor is negligible. If this wasn't the case, confinement differences between the horizontal and the tilted cavity would lead to differences in light leakage through the waveguide and hence the collected power flux.



Figure 6.14: The electric field distribution for the optimised horizontal (a) and tilted (b) cavity coupled waveguides. The figure shows a clear increase in the field distribution along the waveguides that are coupled to a titled cavity due to an increased coupling efficiency.



Figure 6.15: The power fluxes collected from the end of one of the two waveguides that are coupled to the cavity. The figure shows clear enhancement in the collected power flux from a tilted cavity compared to a horizontal cavity, while maintaining similar Q-factors between the two cases.

Figure 6.15 shows that there is over 300% enhancement in the collected flux from the cavity; this is larger than similar work done previously⁴¹. Interestingly, the coupling of higher order modes and/or edge state modes to the waveguides diminish when a tilted cavity structure is used. This may mean that these states might not exist within a tilted cavity, and if they do they will be in other wavelength ranges.

6.3.3. Y junction Photonic Crystal Beam splitters for Integrated QKD Systems

While two waveguides optimally coupled to a tilted cavity can serve as an effective beam splitter scheme for QKD, an optimised Y shaped beam splitter based on PhCs is necessary to realise an efficient Mach-Zehnder interferometer on Bob's side. In this work, we will briefly discuss a method in which light can be efficiently split into two arms with little reflection at the Y-junction; this method was suggested initially in reference 42. In this section, the aim is to quantify the effect of modifying the Yjunction by measuring the power flux collected at the end of the beam splitter's arms using a flux region, hence providing better estimate to the enhancement obtained by modifying the beam splitter. The splitting efficiency of an unmodified beam spliter design similar to the one shown in Figure 6.16 (a) is first measured. The beam splitter is then modified as shown in Figure 6.16 (b). The modification involves adding an extra hole at the Y-junction with r = 0.23a which is shifted by S = 0.30a. After simulating the two different cases, the results were presented in Figure 6.17. From the E_v field distribution, it is easy to see even splitting of the light in the two arms in the modified beam splitter. Furthermore, the field in the two arms is much less intense when an unmodified beam splitter is used. This becomes much more obvious when the flux spectrum is output Figure 6.17 (b). It is clear to see that modifying the beam splitter has caused the power coupled to the arms to approximately double. This means that over 50% of the power is lost at the Y-junction when an unmodified beam splitter is used. In a QKD arrangement similar to the one shown in Figure 6.1, which typically uses 5 beam splitters, losses like these due to poor beam splitter design can cause a large reduction in the achievable bit rate.



Figure 6.16: Schematics images of the unmodified (a) and the proposed modified (b) beam splitter. The modification is made by adding a smaller hole, with r = 0.23a, shifted by S = 0.30a,



Figure 6.17: The field distribution inside a modified Y beam splitter structure (left) and the power flux spectrum (right) obtained from modifying the beam splitter structure as per Figure 6.16.

6.3.4. Optimised Light Out-Coupling Efficiency from a Photonic Crystal Waveguide

In this section, a study was carried out to investigate variations in the collected flux as the out-coupler is displaced along the PhC waveguide. This was performed to find the out-coupler's ideal position in which the collection efficiency is optimised. To perform this study, a simulation was setup to measure the flux spectra at the end of a PhC waveguide (before the out-coupler), and after the out-coupler, as shown by the red rectangles in Figure 6.18. The Bragg out-coupler is designed to have alternating half circular rings of width $\lambda/_{4n}$ where n is the refractive index of GaAs. These Bragg mirror based out-

couplers introduce reflections of light back to the waveguide leading to the formation of a standing mode along the waveguide, as shown in Figure 6.19. As discussed in section 6.2.3, the maximum collected power flux shouldn't be more than 50% the collected flux at the waveguide. This is because the other 50% should be directed in the opposite direction of the large flux region, due to the symmetry of the structure. However, if a distributed Bragg mirror is simulated underneath the out-coupler, the coupling ratio could reach close to unity.



Figure 6.18: Enhancing the light coupling efficiency from a Bragg out-coupler. A schematic representation of the simulation setup showing the position of the flux regions. The flux region at the waveguide should extend in the z-axis through the slab.



Figure 6.19: An x-y slice of the initial (a) and final (b) positions of the Bragg out-coupler. The large red arrow shows the direction of displacement of the out-coupler from the initial position to the final position.

In every successive simulation, the out-coupler is shifted by 0.1a out for a total displacement of 2a, and the percentage of the coupled power flux collected at the top of the out-coupler is compared to that

at the waveguide. The results of this are shown in Figure 6.21 (a). The initial position was randomly chosen to be the left end of one of the holes closest to the waveguide region, Figure 6.20 gives a visual illustration of the initial and final position of the out-coupler right edge. The results show that the coupling efficiency can change dramatically as the out-coupler is displaced along the waveguide, in fact both an increase and decrease in coupling efficiency is observed as the displacement is changed. A maximum efficiency of 94.5% (the circled point in Figure 6.21 (a)) was achieved when the position of the right edge of the out-coupler is displaced by 0.4a from the initial position as illustrated by Figure 6.20. Hence, careful design of the device is necessary to allow maximum coupling of single photons from the chip for QKD applications. The optimised position of the out-coupler, together with a cross section field profile at the out-coupler is shown in Figure 6.21.



Figure 6.20: A visual illustration of the chosen initial and final positions of the right edge of the out-coupler. The figure also shows the position that gives the highest coupling efficiency of 94.5%, as shown by the circled point in Figure 6.21 (a).



Figure 6.21: (a) A scatter graph of the coupling efficiency obtained from displacing the Bragg out-coupler along the x-direction. (b) Graphical representation of the out-coupler's position that couples maximally to the waveguide with an efficiency of 95%. The inset shows a slice along the red line of the electric field as it leaves the waveguide and travels upwards and downwards, working affectively as a dipole source.

6.4. Assembling a Chip-Scale QKD System

After discussing and optimising cavities, waveguides, phase modulators and out-couplers for out-ofchip coupling, the DPS-QKD chip can be discussed as a whole, as illustrated in Figure 6.22.

As shown previously in Figure 6.1, the DPS-QKD system starts with a single photon source. In this work, this was proposed to be an InAs QD, coupled to either of the optimised cavity designs 1 or 2. After the source, the photons are split into two arms. This was proposed to be accomplished by coupling two waveguides to a tilted cavity. Using two waveguides directly coupled to the cavity helps to reduce losses that can be encountered if a Y junction PhC beam splitter was used. The cavity is suggested to be tilted to improve light coupling to the waveguides. Photons that travel through one arm should encounter a phase modulation compared to the other. This is suggested through use of a PhC phase modulator. The light is then coupled out of the chip through both arms using a Bragg mirror out-coupler. The position of the out-coupler with respect to the PhC waveguide is necessarily tuned according to the simulations performed in section 6.3.4 for optimum light out-coupling and reduced reflections at the waveguide-out-coupler interface. Ideally, a distributed Bragg mirror tuned to the InAs QD wavelength can be used underneath the out-coupler to reflect downward travelling photons to the collecting optics above the chip. The photons from either arm are then collected and combined through a fibre beam splitter before travelling to the receiver.

6.5. Preliminary Experimental Results for Photonic Crystals Using GaSb QDs

GaSb QDs are type-two systems that have an extremely large hole confinement potential, resulting in efficient operation at room temperature. Furthermore, by manipulating their growth, quantum rings can



Figure 6.22: An illustrative image of the proposed QKD chip showing the optimised cavity design and out-coupler with the suggested phase modulator.

be formed which benefit from an increased overlap of the electron-hole wave function uncommon in type-two systems, giving them great potential as quantum emitters⁴².

Due to their operation temperature, QDs based on GaSb have promising applications in quantum technologies. Having similar optical properties to InAs QDs, the room temperature operation that GaSb benefits from has made them a target for initial attempts at PhC fabrication, mainly to master the fabrication process. The devices were made on a GaAs substrate using e-beam lithography and Chlorine based ICP etching. The fabrication process is explained in detail in chapter 3. On excitation at 170 µW laser power, a cavity mode peak was observed at around 1250 nm with a FWHM of around 0.3 nm (Lorentzian fitted), resulting in a cavity Q-factor of over 4000. Figure 6.23 (a) shows an SEM image of a fabricated device using the optimised design 2, together with the micro-photoluminescence emission from the cavity mode shown in Figure 6.23 (b). A µ-PL spectrum of an ensemble of GaSb QDs without a PhC structure is displayed in Figure 6.23 (c) for comparison. The high QD density of this sample made precise Purcell enhancement measurements difficult to perform since more than one QD can be emitting at the same cavity mode wavelength. It is important to mention that while the cavity and the QD light emitter work at room temperature, it is necessary for the photonic structure to be under vacuum for the cavity mode to be observed. This is because it is likely that atmospheric pressure and moisture in the air tend to increase the refractive index surrounding the structure, reducing the index contrast in the structure. It is clear to see from Figure 6.23 (b) that there is emission near the edge of the bandgap, these are likely to be edge state modes that arise from localised states through the photonic lattice.



Figure 6.23: (a) Scanning Electron Micrograph of a fabricated PhC cavity according to optimised design 2 from Minkov et al. (b) Micro-photoluminescence measurement of the emission spectrum obtained from the cavity showing the cavity and an edge state mode coupled to a dense GaSb spectrum, excited with a 0.5 μ W laser power (c) uPL emission from an ensemble of GaSb QDs at 5 μ W without a PhC structures. The discontinuity in the spectrum is because the plot is made of multiple spectra recorded at different central wavelengths. All the measurements were made at room temperature.



Figure 6.24: (a) Scanning electron micrograph of a fabricated out-coupler coupled to a PhC waveguide that itself is coupled to an L3 resonant cavity as a first step toward a chip scale integrated QKD experiment. (b) The spectrum obtained from the out-coupler only in comparison to that obtained from both the out-coupler and cavity/waveguide radiative leaky modes. Measurements were done using confocal μ -photoluminescnece.

To test the coupling efficiency, fabrication of out-couplers coupled to a PhC waveguide was performed and the results of the fabricated system are shown in Figure 6.24. It can be seen from the corresponding spectrum of the device in Figure 6.24 that a cavity mode was found at a wavelength of 1304 nm, which is different from the 1250 nm found in the previous device. The difference can be associated to uncertainties in the lithography and etching of the devices. The Q-factor for this cavity was measured to be approximately 1210, which is lower than the L3 cavity case shown in Figure 6.23 (b), however, this is mainly due to the adjacent waveguide near the cavity which can allow the cavity mode to leak into it, hence reducing the cavities Q-factor. Four different peaks were also found between 1210 nm and 1250 nm that are likely to be associated with photonic edge states. To measure the out-coupling efficiency, the laser peak is focused on the L3 cavity and emission was collected in two situations. First emission was collected from the entire structure which includes emission from the cavity/waveguide radiative leaking modes together with the out-coupler's emission. Measurements were then made to collect light emitted by the out-coupler only. These measurements were done using a confocal system which involves an iris that can cover the entire structure except the out-coupler, hence collecting light emitted by the out-coupler only. To calculate the out-coupling efficiency, the ratio of the out-coupler's emission to radiative emission was taken. This was found to be approximately 25% for the case shown in Figure 6.24. This efficiency can be improved by fine tuning the position of the out-coupler and the growth of a Bragg mirror below the structure that can redirect light emitted toward the substrate toward the photoluminescence objective lens.

6.6. Conclusions

After proposing a scheme in which QKD can be realised using a PhC platform, the aim of this chapter was to optimise the optical components that assemble the QKD system, thereby improving its achievable performance and efficiency. The chapter starts with an introduction to state-of-the-art integrated QKD and the benefit that this proposal can bring to the technology. Discussions were first made to compare current methods of realising photonic cavities, waveguides, phase modulators and out-couplers and comparisons were made to put forward the most suitable system. Numerical simulations were performed using finite difference time domain methods to optimise practical PhC cavities and efficiently couple them to PhC waveguides. To enhance out-of-chip coupling efficiency, numerical simulations were also performed to optimise the out-coupling efficiency of light from the chip using a Bragg mirror out-coupler. Enhancement in the coupling efficiency at Y-junction beam splitters was also performed and coupling of over 90% was achieved. After proposing and optimising all the necessary components to realise a DPS-QKD system, a potential design model for a typical 'Alice' chip was shown which involves the optimised tilted cavity coupled to the newly proposed beam splitting scheme by coupling the cavity to two waveguides. Contacts for phase modulation were fabricated around one arm of the waveguide, whilst doping the waveguide to create p-i-n regions provided a reduction of the distance between the two electrodes, thereby creating a higher electric field to minimise the waveguide's length and the voltage needed for phase modulation.

Bibliography

¹ N. Gisin, G. Ribordy, W. Tittel, H. Zbinden. Quantum cryptography. Rev. Mod. Phys., 74, 145-195 (2002)

² H. Lo, M. Curty, K. Tamaki. Secure quantum key distribution. Nature Photon., 8, 595-604 (2014)

³ V. Scarani, H. Bechmann-Pasquinucci, N. Cerf, M. Dusek, N. Lutkenhaus, M. Peev. The security of practical quantum key distribution. Rev. Mod. Phys., 81, 1301-1350 (2009)

⁴ C. Bennett, G. Brassar. Quantum cryptography: public key distribution and coin tossing. In Proceedings of IEEE International Conference on Computers, Systems and Signal Processing, 175, 8-12 (1984)

⁵ K. Inoue, E. Waks, Y. Yamamoto. Differential Phase shift quantum key distribution. Phys. Rev. Lett., 89, 037902 (2002)

⁶ J. Wang, A. Santamato, P. Jiang, D. Bonneau, E. Engin, J. W. Silverstone, M. Lermer, J. Beetz, M. Kamp, S. Hofling, M. G. Tanner, C. M. Natarajan, R. H. Hadfield, S. N. Dorenbos, V. Zwiller, J. L. O'Brien, M. G. Thompson. Gallium arsenide (GaAs) quantum photonic waveguide circuits. Optics Communications, 327, 49-55 (2014)

⁷ H. C. Nguyen, S. Hashimoto, M. Shinkawa, T. Baba. Compact and fast photonic crystal silicon optical modulators. Optics Express., 20, 22465-22474 (2012)

⁸ Q. Lu, W. Guo, D Byrene, J. F. Donegan. Design of low V_{π} high-speed GaAs travelling-wave electrooptic phase modulators using an n-i-p-n structure. IEEE Photonic Technology Letters, 20, 1805-1807 (2008)

⁹ P. Sibson, C. Erven, M. Godfrey, S. Miki, T. Yamashita, M. Fujiwara, M. Sasaki, H. Terai, M. G. Tanner, C. M. Natarajan, R. H. Hadfield, J. L. O'Brien, M. G. Thompson. Chip-based quantum key distribution. Nature Commun., 13984 (2017)

¹⁰ G. Brassard, N. Lutkenhaus, T. Mor, B. C. Sanders. Limitations on practical quantum cryptography. Phys. Rev. Lett, 85, 1330-1333 (2000)

¹¹ M. D. Eisaman, J. Fan, A. Migdall, S. V. Polyakov. Invited review article: single-photon sources and detectors. Rev. Sci. Instrum., 82, 071101 (2011)

¹² L. Landin, M. S. Miller, M. E. Pistol, C. E. Pryor, L. Samuelson. Optical studies of individual InAs quantum dots in GaAs: few-particle effects. Science, 280, 262-264 (1998)

¹³ X. Ding, Y. He, Z. C. Duan, N. Gregersen, M. C. Chen, S. Unsleber, S. Maier, C. Schneider, M. kamp, S. Hofling, C. Lu, J. Pan. On-demand single photons with high extraction efficiency and nearunity indistinguishability from a resonantly driven quantum dot in a micropillar. Phys. Rev. Lett., 116, 020401 (2016)

¹⁴ X. Ziang, L. Shifeng, Q. Laixiang, P. Shuping, W. Wei, Y. Yu, Y. Li, C. Zhijian, W. Shufeng, D. Honglin, Y. Minghui, G. G. Qin. Refractive index and extinction coefficient of CH₃NH₃PbI₃ studied by spectroscopic ellipsometry. Optical Material Express, 5, 29-43 (2015)

¹⁵ M. McClish, R. Farrell, K. Vanderpuye, K. S. Shah. A re-examination of silicon avalanche photodiode gain and quantum efficiency. IEEE Nuclear Science Symposium Conference Record, DOI: 10.1109/NSSMIC.2005.1596952 (2005)

¹⁶ A. Schwagmann, S. Kalliakos, I. Farrer, J. P. Griffiths, G. A. C. Jones, D. A. Ritchie, A. J. Shields. On-chip single photon emission from an integrated semiconductor quantum dot into a photonic crystal waveguide. Appl. Phys. Lett., 99, 261108 (2011)

¹⁷ D. G. Gevaux, A. J. Bennett, R. M. Stevenson, A. J. Shields, P. Atkinson, J. Griffiths, D. Anderson, G. A. C. Jones, D. A. Ritchie. Enhancement and suppression of spontaneous emission by temperature tuning InAs quantum dots to photonic crystal cavities. Appl. Phys. Lett., 88, 131101 (2006)

¹⁸ A. Javadi, I. Sollner, M. Arcari, S. Lindskov Hansen, L. Midolo, S. Mahmoodian, G. Kirsanske, T. Pregnolato, E. H. Lee, J. D. Song, S. Stobbe, P. Lodahl. Single-photon non-linear optics with a quantum dot in a waveguide. Nature Commun., 6, 8655 (2015)

¹⁹ P. Lodahl, S. mahmoodian, S. Stobbe. Interfacing single photons and single quantum dots with photonic nanostructures, Rev. Mod. Phys., 87, 347-400 (2015)

 20 S. Tomic, J. Pal, M. Migilorato, R. J. Young, N. Vukmirovic. Visible spectrum quantum light sources based on In_xGa_{1-x}N/GaB quantum dots. ACS Photonics, 2, 958-963 (2015)

²¹ C. Santori, M. Pelton, G. Solomon, Y. Dale, Y. Yamamoto. Triggered single photons for a quantum dot. Phys. Rev. let., 86, 1502-1505 (2001)

²² M. Holmes, K. Choi, S. Kako, M. Arita, Y. Arakawa. Room temperature triggered single photon emission from a III-Nitride site controlled nanowire quantum dot. Nano Lett., 14, 982-986 (2014).

²³ H. J. Kimble, M. Dagenais, L. Mande, Photon antibunching in resonance fluorescence. Phys. Rev. Lett., 39, 691-695 (1977)

²⁴ F. Diedrich, H. Walther. Nonclassical radiation of a single stored ion. Phys. Rev. Lett., 58, 203-206 (1987)

²⁵ C. Jurtsiefer, S. Mayer, P. Zarda, H. Weinfurter. Stable solid-state source of single photons. Phys. Rev. Lett., 85, 290-293 (2000)

²⁶ K. Srinivasan, O. Painter. Linear and nonlinear optical spectroscopy of a strongly coupled microdiskquantum dot system. Nature, 450, 862-865 (2007)

²⁷ K. Hennessy, A. Badolato, M. Winger, D. Gerace, M. Atature, S. Gulde, S. Fält, E. L. Hu, A. Imam Oğlu. Quantum nature of a strongly coupled single quantum dot-cavity system. Nature, 455, 896-899 (2007)

²⁸ V. Loo, C. Arnold, O. Gazzano, A. Lemaitre, I. Sagnes, O. Krebs, P. Voisin, P. Senellart, L. Lanco. Optical nonlinearity for few-photon pulses on a quantum dot-pillar cavity device. Phys. Rev. Lett., 109, 166806 (2012)

 29 P. E. Barclay, K. Srinivasan, O. Painter, B. Lev, H. Mabuchi. Integration of fiber-coupled high-Q SiN_x microdisk with atom chips. Appl. Phys. Lett., 89, 131108 (2006)

³⁰ U. K. Khankhoje, S-H. Kim, B. C. Richards, J. Hendrickson, J. Sweet, J. D. Olitzky, G. Khitrova, H. M. Gibbs, A. Scherer. Modelling and fabrication of GaAs photonic-crystal cavities for cavity quantum electrodynamics. Nanotechnology, 21, 065202 (2010)

³¹ M. Smit, X. Leijtens, H. Ambrosius, et al.. An introduction to InP-based generic integration technology. Semicond. Sci. Technol., 29, 083001 (2014)

³² C. J. M. Smith, H. Benisty, S. Olivier, M. Rattier, C. Weisbuch, T. F. Krauss, R. M. De La Rue, R. Houdre, U. Oesterle. Low-loss channel waveguides with two-dimensional photonic crystal boundaries. Appl. Phys. Lett., 77, 2813-2815 (2000)

³³ Y. Sugimoto, Y. Tanaka, N. Ikeda, Y. Nakamura, K. Asakawa. Low propagation loss of 0.76 db/mm in GaAs-based single-line-defect two-dimensional photonic crystal slab waveguides up to 1 cm in length. Optics Express, 12, 1090-1096 (2004)

³⁴ G. Z. Mashanovich, M. M. Miloševic, M. Nedeljkovic, N. Owens, B. Xiong, E. J. Teo, Y. Hu. Low loss silicon waveguides for the mid-infrared. Optics Express, 19, 7112-7119 (2011)

 35 Q. Lu, W. Guo, D. Byrne, J. F. Donegan. Design of low V_{π} high-speed GaAs travelling-wave electrooptic phase modulators using an n-i-p-n structure. IEEE Photonics Technology Letters, 20, 1805-1807 (2008)

³⁶ H. C. Nguyen, S. Hashimoto, M. Shinkawa, T. Baba. Compact and fast photonic crystal silicon optical modulators. Optics Express, 20, 22465-22474 (2012)

³⁷ F. S. F. Brossard, X. L. Xu, D. A. Williams, M. Hadjipanayi, M. Hugues, M. Hopkinson, X. Wang, R. A. Taylor. Strongly coupled single quantum dot in a photonic crystal waveguide cavity. Appl. Phys. Lett., 97, 111101 (2010)

³⁸ M. Minkov, V. Savona. Automated optimization of photonic crystal slab cavities. Sci. Rep., 4, 5124 (2014)

³⁹ Y. Akahane, T. Asano, B. Song, S. Noda. High-Q photonic nanocavity in a two-dimensional photonic crystal. Nature, 425, 944-947 (2003)

⁴⁰ A. Faron, E. Waks, D. Englund, I. Fushman, J. Vuckovic. Efficient photonic crystal cavity-waveguide couplers. Appl. Phys. Lett., 90, 073102 (2016)

⁴¹ FDTD simulation software with CrystalWave. Photon Design. <u>https://www.photond.com/products/crystalwave/crystalwave_features_fdtd_01.htm</u> (last accessed 10/02/2017)

⁴² M. P. Young, C. S. Woodhead, J. Roberts, Y. J. Noori, M. T. Noble, A. Krier, E. P. Smakman, P. M. Koenraad, M. Hayne, R. J. Young. Photoluminescence studied of individual and few GaSb/GaAs quantum rings. AIP Advances, 4, 117127 (2014)

Chapter 7

Conclusions and Future Work

7.1. Conclusions

In this thesis, potential approaches to realising optical components for integrated chip-scale quantum key distribution (QKD) systems are numerically and experimentally studied. While the primary application for carrying out research in this direction is QKD, these components can be utilised in many other applications such as quantum computing, optical communication, metrology, sensing etc. Firstly, numerical studies for coupling of transition metal dichalcogenides (TMDC) QDs with optical cavities based on rod-type photonic crystals (PhC) were done for the first time to improve light-matter interaction and provide enhancement in the light collection efficiency. Secondly, deterministic spatial growth of III-V nanowires on silicon was achieved for single photon detection applications. Thirdly, a proposal for integrating a QKD system using PhCs was presented for the first time. Numerical optimisation of the individual components needed for QKD was achieved and initial fabrication results were presented.

The single photon source is one of the most important components found in a typical QKD system. It directly contributes to the bit rate and security of the distributed key in QKD. Chapter 4 of this thesis aims at utilising the unique optical properties of TMDCs for realising integrable single photon sources. A rod-type PhC cavity design was proposed to efficiently couple to TMDC monolayers. Compared to using air-bridge hole-type PhC cavities, this technique, in principle provides optimised coupling

between the cavity mode's maximum and the light emitter. In addition, the reduced dielectric-to-air volume ratio of rod-type PhCs minimises light absorption by the dielectric material. Finite difference time-domain (FDTD) method was used to simulate the optical properties of the PhC cavity, output its Q-factor and measure the enhancement in the collected flux emitted by a TMDC light source coupled to the cavity. Simulations of a hexagonal lattice PhC show a bandgap forming for TE-like modes between $1.05a < \lambda < 1.17a$. By observing the field profile for E_x and E_y, a cavity confinement mode was found at $\lambda = 1.12a$ with a Q-factor simulated to be more than 300 for r = 0.165a and rods height h = 2.3a. Next a flux region was setup above the cavity to mimic a photoluminescence objective with numerical aperture of 0.65. A pulse source inside the cavity is initiated and the power flux transmitted through a flux region is accumulated as the field decays from the cavity with time. The power flux is compared to the case where the source is placed directly on the substrate without the presence of a rodtype PhC structure. After comparing the power fluxes, it was found that the photonic cavity can provide about 400 % enhancement in the light collection efficiency. To study the effect of variations in the rods' radius on the extraction efficiency due to fabrication fluctuations, simulations were carried out for rranging from 0.155a to 0.170a. Results have shown that maximum enhancement takes place when the rods' radius is r = 0.161a. The collected power flux starts to reduces, hence reducing the collection enhancement to lower percentages as the radius deviates away from r = 0.161a. The comparison between the different flux spectra as r is varied has shown that reducing the radius of the rods causes a blue-shift in the cavity mode's energy, while increasing r causes a red-shift in the cavity mode's energy. Numerical modelling was also carried out to investigate the effect of dipping the monolayer to different heights within the cavity on the achieved enhancement in the collected light. Simulations were done while the source position is moved from vertically from the bottom of the cavity to the top in steps. The comparison between the different power flux spectra collected from every step showed maximum enhancement when the source vertical height is near the central height of the rods where the cavity mode's maximum exists. The enhancement reduces as the source is moved toward the top of the rods due to reduced spatial coupling between the emitter and the cavity mode. In addition coupling to radiative modes to air causes further reduction in the enhancement ratio. The enhancement reduces as

the source is moved downward, away from the central height of the rods. This is minimised at the bottom of the cavity where coupling to substrate-mediated leaky modes becomes more dominant.

Experimental fabrication of rod-type PhCs using CSAR62 as a positive e-beam resist has been shown. Cr was transferred to the substrate as a hard mask to provide high selectivity in the dry etch of Silicon. Reactive ion etching technique was utilised to form the rods by etching silicon using SF_6 and O_2 gasses at 130W RF power, achieving an etch rate of approximately 90nm per minute. The transfer of a monolayer on the PhC was followed using the wet transfer method. Finally, results from successful PhC fabrication and monolayer transfer were presented.

In a QKD system, single photon detectors are essential components that directly influence the achieved key bit rate. Chapter 5 aims at advancing the recently emerged application of nanowire based photodetectors to grow III-V nanowires on Si material in a spatially deterministic way. The unique geometrical, electrical and optical properties of nanowires that make them superior over conventional methods of realising photodetectors were discussed together with the current state research in the field. Different methods of biasing free standing and drop-casted nanowires were introduced. Next, a proposal was shown for optically coupling epitaxially grown free standing nanowires photodetector to a photonic chip. For spatially selectively growing these nanowires, results from the fabrication of nanowire sites using e-beam lithography were given. The e-beam CAD pattern is firstly corrected for proximity effects using Beamer. The pattern is written by exposing a CSAR resist which was previously spin coated on a Si/SiO₂ substrate. The exposed regions are then developed in o-Xylene solution. The pattern is then transferred to the oxide layer by wet etching with buffered oxide HF etcher. The resist is then stripped from the substrate using acetone. Electron micrographs of the fabricated nanowire sites were shown for the post-developing and post-etching stages. Enlargement in the transferred nanowire sites was observed due to isotropic chemical etching of the oxide layer. Following the etching immediately, epitaxial growth of InAs and GaAs nanowires was achieved using molecular beam epitaxy. Electron micrographs show that the nanowire successfully seed on the etched nanowire sites where the silicon is exposed, while almost no growth was observed above the oxide covered regions. However, this varies dramatically with temperature as the longest achieved nanowires growth was found to take place within a small growth temperature window around 570 °C. Growing at low wafer temperatures was found to lead to the growth of short nanowires, this is likely to be attributed to reduced migration distance of the nanowire's composition material above the substrate. Operating at a higher substrate's growth temperatures provided enough migration of the composite material which caused growth within the nanowires sites only, while left oxide regions clean. However, this was expected to have led to reducing the amount of group V material available for the growth of nanowires due to their low evaporation temperature. The growth ratio of nanowires per the number of available sites have been measured to be over 70%. The micrographs have shown that all the nanowires exhibited hexagonal structures with different symmetries. The variation of the symmetry could have been due to nanowires seeding on the particle remains of SiO₂ that were found to appear after wet etching the oxide layer. Results from growing GaAs nanowires on nanowires grew following the 111 facet. Optimising this could be done by fine tuning the growth temperature and the material flow rate.

Prior to experimentally fabricating a photonic circuit for QKD, spending a large amount of effort for modelling the components is essential for designing an optimised system with optimum efficiency. Chapter 6 proposed a model to miniaturise conventional differential phase shift (DPS) QKD to a chip level using PhCs as a platform for integrated photonic circuitry on GaAs. Single photon sources based on InAs quantum dots (QDs) embedded in the bridge structure of a typical hole-type PhC cavity are suggested. An L3 cavity design is proposed for Purcell enhancing light emission from InAs QDs by increasing the field intensity within the vicinity of the emitter due to the cavity mode. Different variations to the standard L3 cavity design were proposed and simulated using FDTD. The design was optimised by investigating the effect of changing the holes' radius on the standard cavity. As the radius of the holes is increased, the cavity mode's wavelength was found to reduce. Maximum Q factor for the cavity was found with a radius r = 0.3a. Optimising the cavity's design was carried out by shifting the holes adjacent to the cavity, while measuring its achieved Q-factor. The first optimised design was found by setting $r_A = 0.2a$, which formed a cavity with $Q = 1.3 \times 10^5$. Simulations have also shown that the cavity mode's volume blueshifts as r_A is increased. The same relation was observed for H1 cavities, the optimised Q-factor for H1 cavities was found for $r_A = 0.15a$ though. The second optimised design was taken from literature, however, the work done in this chapter aimed at investigating the optimised design with GaAs for wavelengths emitted by InAs QDs, in contrast to using Si material for a cavity mode wavelength in the mid infrared spectrum as was presented in the original work. Simulation results have shown that Q-factors over 1.5×10^5 were achievable.

To extract single photons out from the cavity in a QKD chip, a PhC waveguide is spatially coupled to the cavity to allow the confinement mode to leak into the waveguide mode. A study was carried out to compare the enhancement in the power flux collected through a PhC waveguide when it is coupled to a tilted and horizontal cavity. FDTD have shown over three times enhancement in the flux collected from a waveguide with a tilted cavity compared to a horizontal cavity. This optimisation in principle can lead to higher bitrate when utilised in a QKD chip.

A Mach-Zehnder is an important component in DPS-QKD systems. To reduce losses in a Mach-Zehnder, optimised Y-junction beam splitters that have minimum losses is essential and was one of the main motivations for carrying out this work. The simulation was performed by recording the flux as it travels toward the Y-junction and at the arms of the beam splitter. A conventional Y-beam splitter was optimised by adding a PhC hole at the beam splitter's junction, where the holes have a smaller radius compared to others in the crystal. This modification was found to greatly reduce scattered light at the junction. This improved the light splitting and coupling to the two waveguides. Further optimisation of the beam splitter could be done by modifying the PhC near the 60° bends to reduce scattering losses. It is anticipated that increasing the diameters of the holes surrounding the bend can improve light bending and dramatically reduces losses.

To couple the light efficiently from the photonic chip, a Bragg mirror out-coupler was investigated. The effect of varying the position of a two layer Bragg out-coupler along the PhC waveguide was found to effect the percentage of light emitted out by the out-coupler. Careful positioning of the out-coupler with respect to the PhC holes that the crystal is composed of can lead to coupling efficiencies of as high as 95%. The optimised components are then collected together to make the final proposal of the chip-scale QKD system. The system is composed of the optimised tilted cavity which is coupled to two waveguides

one of which includes a PhC phase modulator. The waveguides are each coupled to the optimised outcouplers.

Finally, preliminary results from testing the optimised PhC L3 fabricated on GaAs using GaSb as light emitters were presented as a first step towards realising chip-scale QKD using PhCs.

7.2. Future Work

One of the main advantages of rod-type PhC cavity is allowing optimised coupling between the cavity mode and a 2D emitter which increases light-matter interaction and provides enhancement in the light extraction from these emitters. Performing computer simulations that can best predict experimental results can only be done by including silicon's absorption in the simulations. This could be done by assigning complex refractive indices to the high dielectric material that resemble silicon. Absorption by the dielectric rods and the substrate can lead to reduced cavity Q-factor and affect the extraction efficiency ratio. In addition, placing a 2D monolayer above the cavity can cause perturbation to the dielectric symmetry of the cavity structure, taking this into account may lead to reduction in the achieved Q-factor, despite the fact that the perturbation might be very small.

In chapter 4, simulations were carried out to investigation the effect of varying the emitter's height in the cavity on the achieved enhancement. While emission from artificially created defects is promoted when the monolayer is maximally strained, simulating changes in the extraction efficiency as the emitter's position is varied in the x-y plane is useful for a comprehensive study of the cavity. Finite element methods of the dipping of the emitter can be utilised to help predicting the dipping of a monolayer in the cavity.

Vertical light confinement is necessary to allow light guiding on-chip with minimum losses. One method of achieving vertical confinement with rod-type PhCs can be accomplished by surrounding the rods with an optically transparent material which could be realised by spin coating the chip or bottom up growth of the material using chemical vapour deposition. The refractive index contrast between the transparent material with air on top and the dielectric substrate at the bottom causes total internal reflection. This technique has never been used with rod-type PhC structures but is expected to increase

the cavity Q-factor, encouraging a wider use of rod-type PhCs for integrated photonic applications. This allows integrating QKD systems using rod-type PhCs. Future plans also involve controlling the dipping of a monolayer inside a cavity using probe techniques.

Having fabricated nanowire sites and successfully managed to isolate single and arrays of nanowires, work has recently started in growing p-i-n junction GaAs nanowires for photodetection applications. Work in this direction has already been carried out using InAs as composition material of the nanowires and results have been published showing mid-infrared photodetection. The path toward growing p-i-n junction nanowires with GaAs should be sufficiently clear, considering similar work was done with InAs nanowires. Following the growth, free standing nanowire will biased by fabricating contacts using the approach explained in chapter 5 as this approach is perfectly suitable for spatial coupling to integrated PhC waveguides. The mask for fabricating the contact pads used to bias the nanowire grid arrays is shown in Figure 7.1. The large pads at the bottom of the figure are contact pads, while the pads at the top of the figure are used to probe the nanowire that grow in the hole arrays (black squares and dots in the figure). Studies can be then follow to investigate the efficiency of photodetection using a single and an array of nanowires.



Figure 7.1: A schematic diagram of the designed mask for fabricating contact pads for biasing the nanowire grid arrays. Enclosed pink regions indicate areas that will be exposed to UV light. The top end of the pads align with the nanowire arrays while the bottom pads are used for making probe contact or wire bonds.

Integrating a QKD system in hand held devices could only be achieved by miniaturizing single photon detectors to the chip-level. For example light from Bob's photonic circuit can be coupled to an external off-the-shelf Si APD using beam out-couplers. The out-coupler directs the light from the waveguide toward the detector chip which is typically situated adjacent to the measurement chip. The layer structure of a QKD chip can also be grown so that both the Si APD detector layers and those required for fabricating photonic crystals are done by molecular beam epitaxy or chemical vapour deposition. Fabricating the detector's contacts and the PhC can then follow using conventional lithography techniques. Another approach to integrating the detectors used in QKD could emerge from successfully realising nanowire based photodetectors. To achieve this, research is needed to be carried out to tune the operation wavelength to match that of InAs QD's emission.

Further work can be carried out to optimise light guiding in hole-type PhC waveguides by making modification to its design. Increasing the confinements of these waveguides and reducing their losses is essential for many applications other than QKD. Waveguides with lower losses can increase the QKD bit rate in QKD and allows fabricating long phase modulators, reducing the voltage required to establish high electric fields for short waveguides. This reduces heat resultant from operating at high switching frequencies.

Successful growth and fabrication of the full QKD system will be followed by many measurements such as photoluminescence, coupling efficiencies between the different optical components, losses by the different components and key generation.

The QKD system discussed in this thesis uses the differential phase shift keying, however, polarisation encoding QKD can also have strong integration potential on a chip scale. Each of these schemes have advantages and disadvantages in their fabrication complexity, bit rate, achieved distance, etc, and the best scheme should be chosen based on the QKD application the system is designed for.

Appendix A

This Appendix contains an example of a MEEP code used to simulate an L3 hole-type photonic crystal cavity embedded in GaAs bridge.

; resetting the software to

(reset-meep) ; clearing memory from MEEP

(define-param res 20); defines the pixel size of the simulation cell (define-param edge-shift 0.15); shift of holes near the cavity

(define-param tdecay 500); define number of time step to run simulation

(define-param pad-holes 8); number of holes at each side

(define-param pad-air 1.5); thickness of air region above cavity

(define-param eps-gaas 12.827859); dielectric constant of GaAs

(define-param hole-radius 0.29); radius of holes

(define-param hole-radius 20.20); radius of small holes near cavity

(define-param slab-thickness 0.5); bridge/slab thickness

(define-param dpml 1); width of pml layer

(define-param fcen 0.273712876505613); central frequency of source

(define-param df 0.1); spectral width of source

(define-param nfreq 10000); number of sampled frequencies

; switching off averaging dielectric values which is a feature in meep

(set! eps-averaging? false)

; defining the dimensions of simulation cell

(define sx (+ 2 (* 2 pad-holes) (* 2 dpml)))

(define sy (+ (* (sin (/ pi 3)) 2 pad-holes) (* 2 dpml)))

(define sz (+ slab-thickness (* 2 pad-air) (* 2 dpml)))

; setting dimension of simulation cell

(set! geometry-lattice (make lattice (size sx sy sz)))

; defining default material to be air

(set! default-material (make dielectric (material air)))

(set! pml-layers (list (make pml (thickness dpml)))); defining pml layer and dimensions

; setting up geometry of the bridge

```
(set! geometry
 (list
   (make block
     (center 0 0 0)
     (size infinity infinity slab-thickness)
     (material (make dielectric (epsilon eps-gaas))))))
; defining the array of PhC lattice
(set! geometry
 (append geometry
   ; even rows above
   (geometric-objects-duplicates
    (vector3 0 (* 2 (sin (/ pi 3))) 0) 1 (/ pad-holes 2)
     (geometric-object-duplicates
       (vector3 -1 0 0) 0
       (+ 2 (* 2 pad-holes))
       (make cylinder
         (center (+ 1 pad-holes) 0)
         (radius hole-radius)
         (height infinity)
         (material air))))
   ; even rows below
   (geometric-objects-duplicates
    (vector3 0 (* 2 (sin (/ pi 3))) 0) (/ pad-holes -2) -1
     (geometric-object-duplicates
       (vector3 -1 0 0) 0
       (+ 2 (* 2 pad-holes))
       (make cylinder
         (center (+ 1 pad-holes) 0)
         (radius hole-radius)
         (height infinity)
         (material air))))
   ; odd rows
   (geometric-objects-duplicates
```

(vector3 0 (* 2 (sin (/ pi 3))) 0)

```
(/ pad-holes -2) (- (/ pad-holes 2) 1)
  (geometric-object-duplicates
    (vector3 -1 0 0) 0
    (+1 (* 2 pad-holes))
    (make cylinder
      (center (+ pad-holes 0.5) (sin (/ pi 3)))
      (radius hole-radius)
      (height infinity)
      (material air))))
; middle row
(geometric-object-duplicates
(vector3 -1 0 0) 0 (- pad-holes 2)
  (make cylinder
    (center -3 0 0)
    (radius hole-radius)
    (height infinity)
    (material air)))
(geometric-object-duplicates
(vector3 1 0 0) 0 (- pad-holes 2)
  (make cylinder
    (center 3 0 0)
    (radius hole-radius)
    (height infinity)
    (material air)))
; the offset cells
(list
  (make cylinder
    (center (+ 2 edge-shift) 0 0)
    (radius hole-radius2)
    (height infinity)
    (material air))
  (make cylinder
    (center (- -2 edge-shift) 0 0)
    (radius hole-radius2)
```

```
(height infinity)
       (material air)))))
; switching on mirror symmetry for quicker simulations
(set! symmetries (list (make mirror-sym (direction X) (phase 1))
                    (make mirror-sym (direction Z) (phase 1))
                    (make mirror-sym (direction Y) (phase -1))))
; setting up pixel size (resolution) of simulation cell
(set-param! resolution res)
; setting up source
(set! sources (list
 (make source
   (src (make gaussian-src (frequency fcen) (fwidth df)))
     (component Ey)
     (center 0 0 0))))
(define (print-mode-volume) (print; measuring mode volume
                           "mode_volume: " (meep-time)
                           ", " (electric-energy-in-box (meep-fields-total-volume fields))
                            ", " (meep-fields-electric-energy-max-in-box fields (meep-fields-total-
volume fields))
                           ", " (meep-fields-modal-volume-in-box fields (meep-fields-total-volume
fields))
                            "\n"))
; defining flux transmission region
(define trans
    (add-flux fcen df nfreq
           (make flux-region
            (center 1 0 0) (size 0 1.5 1.5) (direction X))))
; setting up simulation parameters, such as time steps of simulation, outputting epsilon function, using
harminy to find modes and calculating their Q-factors, outputting mode volume and e-field profile.
```

(run-sources+ tdecay

(at-beginning output-epsilon) (after-sources (harminv Ey (vector3 0 0 0) fcen df)) (after-sources+ (- tdecay (/ .25 fcen)) print-mode-volume) (at-end output-efield-y))

(display-fluxes trans)