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Growth and characterisation of InAsP single quantum dots in InP nano/micro- cavities for single photon source applications

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Declaration

I certify that this dissertation does not incorporate, without acknowledgement, any material previously submitted for a degree or diploma in any university and that to the best of my knowledge, it does not contain any material previously published or written by another person except where due reference is made in the text. The work in this dissertation is my own, except for the contributions made by others as described in the Acknowledgements.

Xiaoying Huang

August 2026

Acknowledgements

Starting a PhD journey is my own decision, but obtaining a PhD degree needs a lot of people's decision: whether the boss will give me the position, whether the tool manager will give me access, whether the MOCVD will be working, whether the editor will accept my paper, etc. All of these are important elements for completing my PhD, and also what we call in Chinese the combination of favourable timing (天时), geographical advantage (地利), and human harmony (人和). Next, I would like to divide my acknowledgements into these three main parts.

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Abstract

Single-photon sources or emitters are at the core of quantum information technologies, ranging from quantum communication to quantum cryptography and quantum computing. Among various types of single-photon emitters, epitaxially grown semiconductor quantum dots have been recognised as highly promising candidates due to their superior optical performance, including high brightness, purity, and indistinguishability. The integration of single quantum dots into optical cavities to enhance the spontaneous emission rate is crucial for achieving high-quality single-photon emitters. While top-down fabrication approaches have been widely explored, deterministic positioning of single quantum dots remains a significant challenge. In this thesis, we develop a hybrid approach that combines droplet epitaxy with selective area epitaxy to deterministically position single quantum dots in various optical cavities, enabling both optically pumped and electrically injected single-photon source applications.

We first demonstrate the combination of droplet epitaxy with high-quality InP nanowire arrays to deterministically position a single quantum dot in each nanowire, enabling scalable single-photon emitters. By systematically investigating the parameter influence of droplet density on individual nanowires (diameter, pitch size, height, and precursor deposition time) — as well as the structural properties of the quantum dots and the shell capping layer conditions (group V-to-group III ratio, pitch size, and temperature), we achieve uniform dot-in-nanowire arrays. The arrays exhibit high brightness, reaching up to one million counts per second under 1 nW laser excitation. This high brightness originates from the single quantum dot emission being coupled to the dominant optical mode of the nanowire, resulting in a Purcell enhancement and a short radiative lifetime of 350 ± 5 ps. FDTD simulation indicates a Purcell factor of 3.5, with the coupled mode identified as the TM_{01} mode. Measurement of the second-order correlation function yields a $g^{(2)}(0)$ value of 0.05, confirming its operation as a high-purity single-photon source.

After achieving optically pumped and deterministically grown single-photon sources in nanowires, the second part of this thesis focuses on the realisation of electrically driven devices to eliminate the need for expensive and bulky laser systems. We demonstrate the design of a core-shell quantum dot-in-nanowire structure for electrically pumped, vertically emitting

single-photon sources. The nanowire core-shell structure consists of a p-doped core, an intrinsic shell layer where the single quantum dot is grown, followed by an n-doped shell. I–V characteristics of the fabricated devices exhibit typical p–n junction rectifying behaviour, confirming successful junction formation. Quantum dots that are coupled to the mode supported by the nanowire cavity exhibit excellent optical performance, with a brightness of up to one million counts per second under a 1.5 μA injection current. Furthermore, highly stable emission wavelength and photon count rate are obtained under continuous current injection for over one hour.

Finally, in the third part of this thesis, we extend our hybrid droplet epitaxy with selective area epitaxy technique to incorporate quantum dots in the recently developed bottom-up ring cavities, which support multiple whispering gallery modes with high-quality factors compared to nanowire cavities, thereby offering a higher probability of achieving coupling. After optimising the growth conditions, we are able to incorporate low-density quantum dots in the good morphology rings with high quality factors (3000-4000). Single-photon emitter with a measured $g^{(2)}(0)$ value of 0.24 and an ultra-short radiative lifetime of 70 ± 1.8 ps are obtained. Through simulation, we show this is a result of the quantum emission being coupled to one of the TE modes of the whispering gallery mode cavity, with a corresponding Purcell factor of 12. By increasing the ring diameter from 4 to 10 μm , more and closely spaced high-quality factor whispering gallery modes can be supported, thereby not only enhancing the probability of coupling with the quantum dots, but also the possibility of multi-wavelength single photon emitters from the same device.

The study presented in this thesis demonstrates the feasibility of deterministically incorporating single quantum dots into various bottom-up cavities using droplet epitaxy and selective area epitaxy, offering a promising pathway toward on-chip integrated quantum photonic devices for miniaturised quantum information processing platforms.

Publications

Journal Articles

1. **Xiaoying Huang**, Jake Horder, Wei Wen Wong, Naiyin Wang, Yue Bian, Karin Yamamura, Igor Aharonovich, Chennupati Jagadish, and Hark Hoe Tan, “Scalable bright and pure single photon sources by droplet epitaxy on InP nanowire arrays”, *ACS Nano*, 2024, 18(7), 5581-5589.
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Acronyms

APD	Avalanche photodiode
BS	Beam Splitter
CL	Cathodoluminescence
CPS	Counts Per Second
DE	Droplet epitaxy
DWL	Direct Write Lithography
EBL	Electron Beam Lithography
EDX	Energy Dispersive X-ray Spectroscopy
FDTD	Finite-Difference Time-Domain
FFT	Fast Fourier Transform
FM	Frank-Van der Merwe
HBT	Hanbury Brown-Twiss
HOM	Hong–Ou–Mandel
HRTEM	High Resolution Transmission Electron Microscopy
IPA	Isopropyl Alcohol
I-V	Current-Voltage
NW	Nanowire
MOCVD	Metal-Organic Chemical Vapour Deposition
PECVD	Plasma-Enhanced Chemical Vapour Deposition
PL	Photoluminescence
QD	Quantum Dot
Q-factor	Quality factor
RIE	Reactive Ion Etching
SAE	Selective Area Epitaxy
SEM	Scanning Electron Microscopy
S-K	Stranski-Krastanov
SPD	Single Photon Detector
SPS	Single Photon Source
STEM	Scanning Transmission Electron Microscopy
TEM	Transmission Electron Microscopy

TMIn	Trimethylindium
TRPL	Time-Resolved Photoluminescence Spectroscopy
VLS	Vapour-Liquid-Solid
VW	Volmer-Weber
WGM	Whispering Gallery Mode
WZ	Wurtzite
ZB	Zinc-Blende

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CHAPTER 1

1.Introduction

1.1. Research Background

The last few decades have witnessed unprecedented growth in information technologies, largely driven by the steady miniaturisation of electronic components and the performance gains guided by Moore's law. However, as traditional semiconductor technologies approach their physical and practical limits, the need for fundamentally new paradigms has become urgent ^[1]. As a result, the emerging field of quantum technologies—leveraging the principles of quantum mechanics—offers a path forward. Central to this field is the ability to generate, manipulate, and detect individual quanta of light, namely single photons. The development of reliable single-photon sources is not only scientifically intriguing but also critical for the implementation of scalable quantum communication, quantum computing, and quantum sensing platforms.

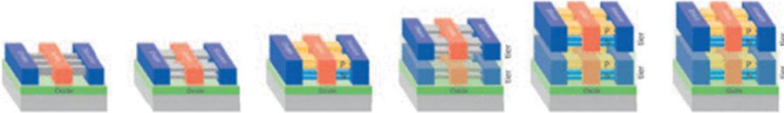
1.1.1 End of Moore's law

Moore's law, proposed by Gordon Moore in 1965, gives an insight in the development of semiconductor industry—the number of transistors on an integrated circuit (unit per chip) doubles about every two years. In principle, increasing number of transistors per unit area can be achieved by either making smaller devices or stacking them vertically or combining them laterally in sophisticated packages. Indeed, in the past few decades, the transistor size has decreased from micrometer to nanometer scale and the number of transistors also increases exponentially ^[2]. The size of the transistor that can be fabricated by the patterning and etching, however, is limited by the wavelength of light for lithography. Currently, A sub-2 nm logic chip can be potentially achieved by using a high-NA extreme ultraviolet lithography light ^[3]. To further continue Moore's law, researchers address the state-of-the art challenges from the complex system of devices processing, design, and manufactures. For example, a monolithic 3D integration of transistor technique has been proposed by stacking the transistor vertically

while the length barely changed as seen in Table 1.1. In this road map, the transistor density may keep increasing up to 0.5 nm node in 2037 by combining several stacks.

Table 1.1 CMOS transistor scaling strategy: limited gate length reduction and increasing use of monolithic 3D layer stacking (2022–2037) ^[4]

Year of production	2022	2025	2028	2031	2034	2037
Node label	"3nm"	"2nm"	"1.5nm"	"1,0nm"	"0.7nm"	"0.5nm"



Vdd(V)	0.7	0.65	0.65	0.6	0.6	0.6
Gate length (nm)	16	14	12	12	12	12
Nr of stacked tiers	1	1	1	2	4	6
Digital block area scaling	1	0.74	0.55	0.26	0.13	0.08
Digital block energy scaling	1	0.81	0.72	0.56	0.5	0.49

As further scale down the transistor to sub-nm level, the Moore's law has encountered significant physical and practical limitations. These include quantum tunnelling effects, which compromise transistor switching reliability, extreme lithographic precision requirements, which approach atomic dimensions, the escalating cost and complexity of advanced semiconductor fabrication. To overcome these limitations and address the increasing demands of next-generation computational problems, there is an urgent need for novel technologies and alternative computing paradigms beyond conventional metal-oxide-semiconductor (CMOS) devices ^[5].

1.1.2 The Second Quantum Revolution

The first quantum revolution of the early 20th century, which introduced foundational principles such as wave-particle duality of light, nature at the atom and energy quantisation, contributed to technologies like lasers, transistors and atomic clocks. The second quantum revolution happening today, as outlined by Dowling and Milburn ^[6], is defined by applying the established quantum principles to solve problems through the technical ability to engineer, control and utilise individual quantum objects (e.g., single photons, atoms and particles) ^[7]. This is currently

driving the developments of key quantum technologies such as quantum computing, quantum communication and quantum sensing ^[6, 8].

At the heart of this revolution is the transition from merely observing quantum effects to actively harnessing quantum coherence and entanglement as operational resources ^[9]. In classical physics, information is encoded in binary states (0 or 1), but in quantum systems, quantum bits (qubits) can exist in a superposition of both states simultaneously. Moreover, entangled states allow distant particles to share nonlocal correlations with high security that cannot be described by classical statistics ^[10]. These unique properties enable quantum-enhanced capabilities in cryptography, communication and sensing.

One of the most prominent outcomes of the second quantum revolution is quantum computing ^[11], which offers exponential speedups for specific tasks such as prime factorisation, unstructured database search and quantum system simulation. Quantum algorithms such as Shor's and Grover's highlight the power of quantum processing ^[12], particularly for problems that are computationally intractable for even the most advanced classical supercomputers. In parallel, quantum communication has emerged as a secure method for transmitting information using quantum key distribution ^[13, 14], which guarantees security based on the fundamental laws of quantum mechanics rather than computational assumptions. Technologies such as quantum repeaters and quantum networks are now being developed to extend quantum communication over long distances ^[15, 16].

Another rapidly advancing field is quantum sensing and metrology ^[17, 18], which leverages quantum states to achieve unprecedented precision in measurements of time, magnetic fields, gravitational acceleration and other physical quantities. For instance, atomic clocks based on optical transitions ^[19] and nitrogen-vacancy centres in diamond for magnetic field detection ^[20, 21] represent breakthroughs in timekeeping and imaging resolution. These sensors have immediate applications in navigation, biomedical diagnostics, geology and even in testing the boundaries of fundamental physics.

The realisation of these technologies depends heavily on progress in quantum materials, quantum control techniques and quantum device engineering ^[22]. Photons, atoms, ions, superconducting circuits and semiconductor quantum dots are all being explored as platforms for qubit implementation, each offering unique trade-offs between coherence time, scalability and ease of integration. In particular, solid-state quantum emitters, such as III-V semiconductor

quantum dots, play a crucial role in the development of on-demand single-photon sources, which are essential components for photonic quantum technologies.

1.1.3 Solid-state quantum light source

Quantum light sources that can emit non-classical light, such as single photons or entangled photon pairs, are fundamental components in emerging quantum technologies. Among the various platforms, solid-state quantum light sources^[23] have garnered significant interest due to their scalability, compatibility with existing semiconductor processing techniques and potential for integration into on-chip photonic circuits.

A solid-state quantum light source usually includes a quantum emitter placed inside a crystalline host material. When excited optically or electrically, the emitter relaxes by releasing a single photon, which can be defined as a single excitation of a mode of quantised electromagnetic field. The field mode is an orthonormal solution to Maxwell's equations described by a set of parameters, including the centre angular frequency ω , spectral and temporal shape, transverse mode profile, and propagation constant^[24]. Unlike traditional light sources, like lasers or LEDs, these quantum emitters can prevent multi-photon emission events and generate exactly one such excitation on demand, allowing for true single-photon generation.

The most popular material system is quantum dots (QDs)—typically within 10 nm in at least one dimension—in which the motion of charge carriers is confined leading to localisation of electrons and holes in one or more spatial directions^[25]. This results in the formation of discrete energy levels in the conduction and valence bands, and the transitions between these levels become similar to those in single atoms. Thus quantum dots are sometimes called “artificial atoms”. Based on the material system and fabrication approach quantum dot-based solid-state quantum light sources can be broadly classified into two categories: epitaxial semiconductor quantum dots and colloidal quantum dots. Epitaxial semiconductor quantum dots which are directly grown on a substrate such as CdSe and CdTe (II-VI materials), and InAs, InGaAs and InGaN (III-V materials). III-V QDs are the most promising candidate for applications in quantum information technologies^[26]. II-VI QDs are also appealing material candidates at room temperature single emitters^[27]; However, their practical applications are limited by fluorescence intermittency (blinking) from charge trapping processes^[28] and low efficiency due to Auger recombination^[29]. Colloidal QDs like CsPbX₃ (X = Cl, Br, I) also as emerging candidate for room-temperature single emission with high purity and narrow linewidth^[30, 31],

but the instability of the material system ^[32, 33] limits its application for on-chip integration for quantum network.

Another widely studied platform includes colour centres in wide-bandgap materials, such as nitrogen-vacancy ^[34] and silicon-vacancy centres in diamond ^[35, 36], and SiC defects, ^[37] respectively. These materials are embedded with impurities of dopant in the crystal lattice of another material or an isolated crystallographic point defect (a vacancy or an atom at an irregular point in the lattice), resulting in atom-like properties. These emitters offer room-temperature stability, long coherence times and compatibility with nanofabrication, making them attractive for practical quantum applications ^[38].

In addition, silicon defect-based emitters are optically active point defects formed by intrinsic lattice defects or impurity–defect complexes in crystalline silicon. The W centre, associated with a silicon tri-interstitial complex, exhibits single-photon emission with a zero-phonon line at ~ 1218 nm ^[39]. The carbon-related G centre emits at ~ 1278 nm and can be fabricated in silicon-on-insulator platforms, offering compatibility with established silicon photonics ^[40]. The T centre is particularly attractive for quantum networking, as it combines telecom O-band emission at ~ 1326 nm with an optically addressable, long-lived electron spin ^[41, 42]. These silicon defects therefore provide a promising route towards scalable single-photon sources and spin–photon interfaces directly integrated with mature silicon photonic technologies.

Furthermore, 2D materials, namely atomically thin two-dimensional materials formed by van der Waals force such as hexagonal boron nitride and transition metal dichalcogenides ^[43, 44], have recently demonstrated single-photon emission from localised defects, offering new avenues for ultra-compact, atomically thin quantum photonic devices ^[45, 46]. However, the layer nature from mechanically transferring or aligning individual flakes or emitters onto photonic circuits make it difficult in scalable and repeatable integration with photonic circuits ^[47].

Despite their promise, solid-state sources face challenges including spectral inhomogeneity, photon indistinguishability, degradation due to phonon interactions and fabrication reproducibility. Addressing these issues through improved material growth, strain engineering, and photonic integration is crucial for the deployment of high-performance, on-chip quantum photonic networks.

1.2. Significance of III-V semiconductor single quantum dots in quantum technologies

III–V semiconductor single QDs have emerged as promising building blocks in the rapidly advancing field of quantum technologies by allowing on-demand generation of single photons and entangled photon pairs. These photons, also called flying qubits, are capable of carrying quantum information over long distance. Figure 1.2 illustrates an envisioned quantum internet with high-performance QDs embedded in microcavities connecting different applications of quantum teleportation, quantum memory, quantum sensing and quantum computing. The detailed advantages of III-V QDs will be discussed in this section.

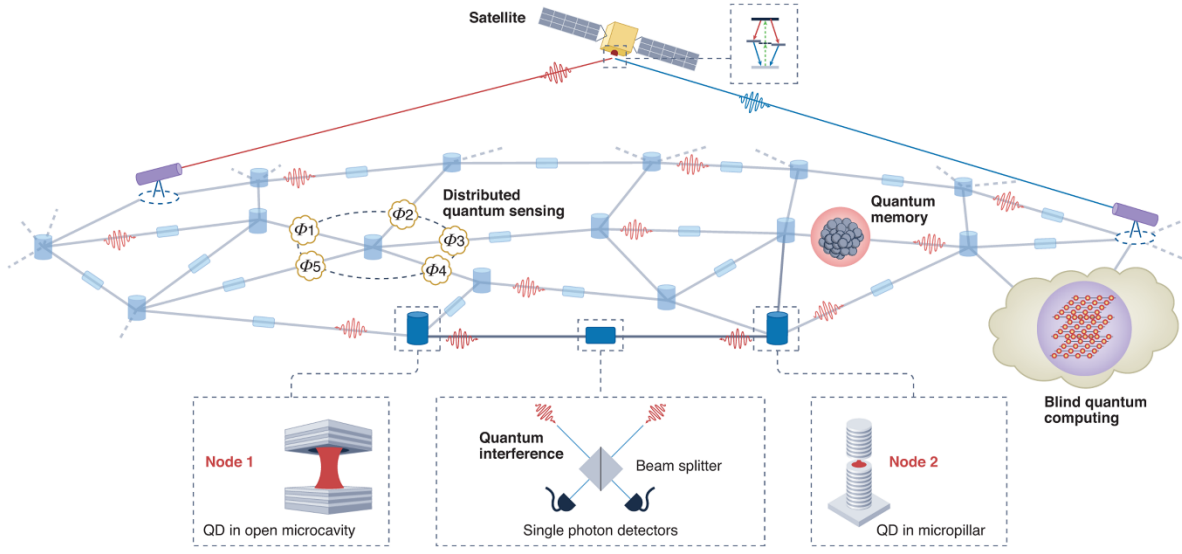


Figure 1.1. An envisioned quantum internet with III-V semiconductor quantum dots ^[48].

1.2.1 Superior optical performance

A key component to realise quantum internet is an ideal quantum light source with high brightness, purity and indistinguishability. By incorporating the QDs to optical cavities and realising coupling between the QD emission and the mode supported by the optical cavity, many desirable properties can be achieved such as enhanced self-emission rate, above-threshold purity and indistinguishability, narrow optical transition linewidth ^[49, 50], short radiative lifetime ^[51] and near-unity quantum efficiency ^[52]. Specifically, InAs/GaAs QDs are the most extensively studied ^[53, 54], offering ultra-low single-photon emission probability with measured second-order correlation values $g^{(2)}(0) < 0.005$ under pulsed non-resonant or resonant excitation ^[55]. The first report of single photon emission using III-V QDs was achieved

by coupling InAs quantum emission (~ 940 nm) to high quality factor GaAs microdisk [56]. Since then, significant improvements have been made by carefully engineering photonic nanostructures to enhance collection efficiency such as micropillars [57, 58], nanobeam waveguides [59], bullseye structure [60, 61] or photonic crystal cavities [62, 63]. These QDs can achieve collection efficiencies exceeding 70%, making them some of the brightest single-photon sources benefit from Purcell-enhanced emission, and support GHz repetition rates. Under resonant π -pulse laser excitation, InAs/GaAs QDs routinely emit photons with indistinguishability exceeding 90–95%, approaching the Fourier-transform limit [64], with coherence times close to twice the radiative lifetime, indicating transform-limited emission.

Similarly, GaAs/AlGaAs QDs offer equally impressive optical performance with additional advantages. Due to their strain-free growth and high symmetry by local droplet etching [65], these QDs exhibit minimal spectral diffusion [66], negligible fine-structure splitting [67], and naturally emit polarisation-entangled photon pairs with fidelities exceeding 90% [68], without requiring post-growth tuning. Their emission wavelength lies in the visible to near-infrared range (700–800 nm), and under resonant excitation, they can emit transform-limited single photons with indistinguishability above 95% [69], setting a benchmark for high-coherence quantum emitters.

Additionally, GaN and InGaN QDs, which emit photons in the visible-light range (400–600 nm), offer the unique advantage of room-temperature operation due to their large band gap [70–72]. These systems have demonstrated clear antibunching behaviour ($g^{(2)}(0) < 0.2$), although challenges remain in improving photon indistinguishability and reducing spectral diffusion due to stronger electron–phonon interactions and wider inhomogeneous broadening [73, 74].

Telecom-wavelength quantum light sources play a pivotal role in the development of scalable and long-distance quantum communication systems [75]. In addition, telecom-wavelength photons are inherently compatible with the existing global fibre-optic infrastructure, facilitating seamless integration of quantum networks with current classical communication systems [76, 77]. InAs/InP QDs have shown strong potential, with emission centred around 1.3–1.55 μm , which aligns well with low-loss transmission windows of standard silica optical fibres, with attenuation as low as 0.2 dB/km at 1550 nm [78]. Although slightly lower in photon indistinguishability compared to InAs/GaAs systems (typically 70–85%), recent improvements in resonant excitation and phonon suppression techniques have led to significant advances. The single-photon purity of InAs/InP QDs can reach as high as $g^{(2)}(0)$ value of $(4.2 \pm 0.2) \cdot 10^{-4}$

[79] and continued development of photonic integration strategies (e.g., with dielectric or semiconductor cavities) is helping to boost brightness and coherence. Table 1.2 illustrates a survey of the state-of-the art achievement with quantum dot-based telecom photon emitters and their optical performance.

Table 1.2. A survey of the state-of-the art achievement with quantum dot-based telecom photon emitters and their optical performance. This table summarises the optical performance metrics experimentally, including photon efficiency (η), Purcell enhancement (F_p), life-time ($T_{1,opt}$), single photon purity ($g^{(2)}(0)$), two photon interference (TPI) visibility for telecom-band-emitting III-V QDs, under different excitation schematics when integrated them into different optical structures. Reproduced from P. Holewa et.al [24].

Matrix	λ [μm]	Structure	η [%]	F_p	$T_{1,opt}$	$g^{(2)}(0)$	TPI visibility V	Excitation	Ref.
Quantum dots									
InP	1.56	Ph. C.		5	0.2 ns	(0.10 ± 0.02)		Nonres.	[240]
InP	1.58	SPH			1.21 ns	$(4.4 \pm 0.2) \times 10^{-4}$		p -shell	[63]
GaAs, MB	1.55	Planar			1.7 ns	(0.072 ± 0.104)	PS: $(89 \pm 11) \%$	RF & TPE	[241]
GaAs	1.30	CBG	23	4	0.3 ns	0.01		p -shell	[242]
InP	1.56	DBR	13.3		1.3 ns	0 ± 0.083		Nonres.	[217]
InP	1.54	Mesa	10		1.7 ns	0 ± 0.038		Nonres.	[218]
InP	1.53	Mesa			1.21 ns	$(5 \pm 4) \times 10^{-3}$	$(35 \pm 3) \%$	TPE	[103]
GaAs	1.32	CBG	11.2	4.2	0.3 ns	(0.087 ± 0.003)		Nonres.	[225]
GaAs	1.33	CBG		5.5	0.29 ns	(0.193 ± 0.022)		Nonres.	[226]
GaAs, MB	1.56	CBG	17	4	0.52 ns	$(5.2 \pm 0.1) \times 10^{-3}$	$(8.1 \pm 3.4) \%$	p -shell	[220]
InAsP/InP	1.31	NW	27.6		2.1 ns	0.021		Nonres.	[91]
GaSb/AlGaSb*	1.47	SIL			0.43 ns	(0.16 ± 0.02)		Nonres.	[62]
InP	1.55	CBG	16.6	5	0.4 ns	$(3.2 \pm 0.6) \times 10^{-3}$	$(19.3 \pm 2.6) \%$	LO-phonon	[102]
GaAs	1.30	CBG		3.9	0.4 ns	(0.055 ± 0.034)	PS: $(61.1 \pm 3.2) \%$	Nonres.	[243]
InP	1.53	CBG	24	5.3	0.78 ns	(0.078 ± 0.016)	PS: $(33.4 \pm 0.1) \%$	Nonres.	[244]
GaAs, MB	1.55	CBG		4.3	0.46 ns	(0.076 ± 0.001)	$(50.4 \pm 1.8) \%$	SUPER	[245]
GaAs, MB	1.55	CBG		4.3	0.46 ns	(0.069 ± 0.001)	$(66.4 \pm 0.4) \%$	LA-phonon	[245]
InP	1.55	NB	12.4		1.87 ns	(0.015 ± 0.003)	PS: $(84 \pm 6) \%$	Nonres.	[52]
DE, InP	1.52	Ph. C.		5	0.34 ns	(0.164 ± 0.009)		LA-phonon	[246]
InAlGaAs/InP	1.55	CBG		6.7	0.18 ns	(0.057 ± 0.004)		p -shell	[68]
InAlGaAs/InP	1.55	CBG		4.7	0.26 ns	(0.0307 ± 0.0004)	$(71.9 \pm 0.2) \%$	p -shell	[56]
InAsP/InP	1.27	NW	28		2.35 ns	(0.006 ± 0.003)	5.6 %	Nonres.	[144]

1.2.2 On-chip integration with quantum photonic platforms

As nanoscale light sources embedded in solid-state matrices, QDs can be deterministically positioned and integrated with on-chip photonic structures using mature semiconductor fabrication techniques to realise complex quantum functionalities, offering a scalable and flexible route toward quantum photonic integrated circuits. Many high-performance components in classical photonics can be directly leveraged and interfaced with QDs, however, this requires efficiently mediated optical interactions enabled by low propagation losses as well as scalable control of individual QDs. In general, material platforms with high refractive index contrast are advantageous for realising compact photonic devices due to their strong mode confinement capabilities. However, they are more susceptible to scattering losses caused by surface and sidewall roughness introduced during non-ideal nanofabrication processes. The overall insertion loss of an optical switch is determined by the footprint and transmission loss of the integrated waveguides, as well as the excess loss introduced by active tuning mechanisms. In contrast, low-index-contrast materials such as SiO_2 and silicon oxynitride (SiON) inherently exhibit lower propagation loss. In particular, the refractive index contrast of silica waveguides closely matches that of standard single-mode optical fibres ^[80], enabling efficient coupling. These waveguides are often fabricated using direct laser writing, which avoids the roughness typically introduced by semiconductor etching, though this comes at the cost of a relatively larger device footprint. High-index-contrast platforms such as GaAs ^[81], Si ^[82], LiNbO_3 ^[83], and Si_3N_4 ^[84] offer index contrasts ranging from 20% to 50%, depending on the cladding material. These platforms support tight optical confinement and significantly reduced device dimensions, making them suitable for dense photonic integration despite their increased sensitivity to fabrication imperfections. The development of integrated quantum technologies either monolithically or heterogeneous and their full potential critically hinges on the compatibility of QD structures with a given photonic integrated circuits platforms ^[85].

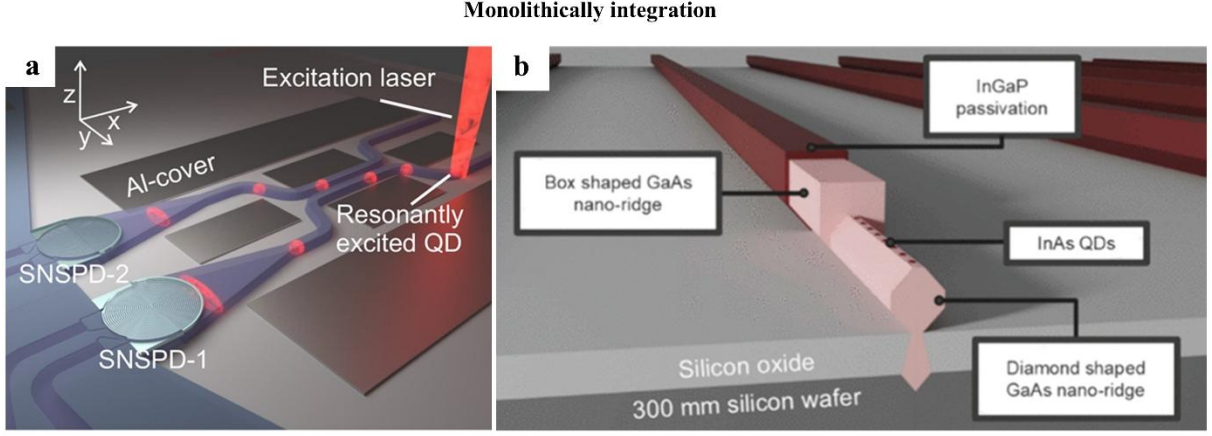


Figure 1.2. Examples of monolithic integration. (a) A fully-GaAs-integrated device. Individual QDs embedded in a single-mode GaAs ridge waveguide are optically excited via a resonant pump laser from top. The resulting emission is guided along the waveguide to an evanescent field coupler, where it is split into two separate waveguide branches. Each branch is tapered at the output and integrated with superconducting nanowire single-photon detectors ^[86]. (b) Realisation of deterministically single photon source by selective growth of InAs QDs within box-shaped GaAs nano-ridges with InGaP shell as passivation layer ^[87].

Monolithic integration which refers to the fabrication of multiple functional components—such as light sources, waveguides, modulators, and detectors—on a single substrate through a unified deposition and lithographic processes. This approach enables the realisation of compact, robust, and highly scalable photonic and optoelectronic systems, as it eliminates the need for post-fabrication alignment and interconnection between discrete elements. Furthermore, monolithic integration also minimises coupling loss between different components on the chip, which is the dominant loss of a quantum photonic system on a hybrid platform. Among different material platforms, (Al)GaAs system is undoubtedly an attractive platform for quantum applications, as single quantum dot devices can be epitaxially grown and are compatible with on-chip detectors as Figure 1.2 (a). Selective growth of GaAs waveguide on large 300 mm Si wafers by using nano-ridge engineering, as illustrated in Figure 1.2 (b), provides a significant advantage for large-scale fabrication as it is compatible with CMOS foundries. Additionally, (Al)GaAs has tight mode confinement offered by its large refractive index contrast of 40% ^[81]. However, the waveguide transmission loss is still large, ranging from tens to even hundreds dB/cm, thus limiting the scalability on the (Al)GaAs platform. Besides, monolithic integration poses

significant challenges in material compatibility and fabrication complexity, especially when combining active and passive elements or integrating dissimilar material systems.

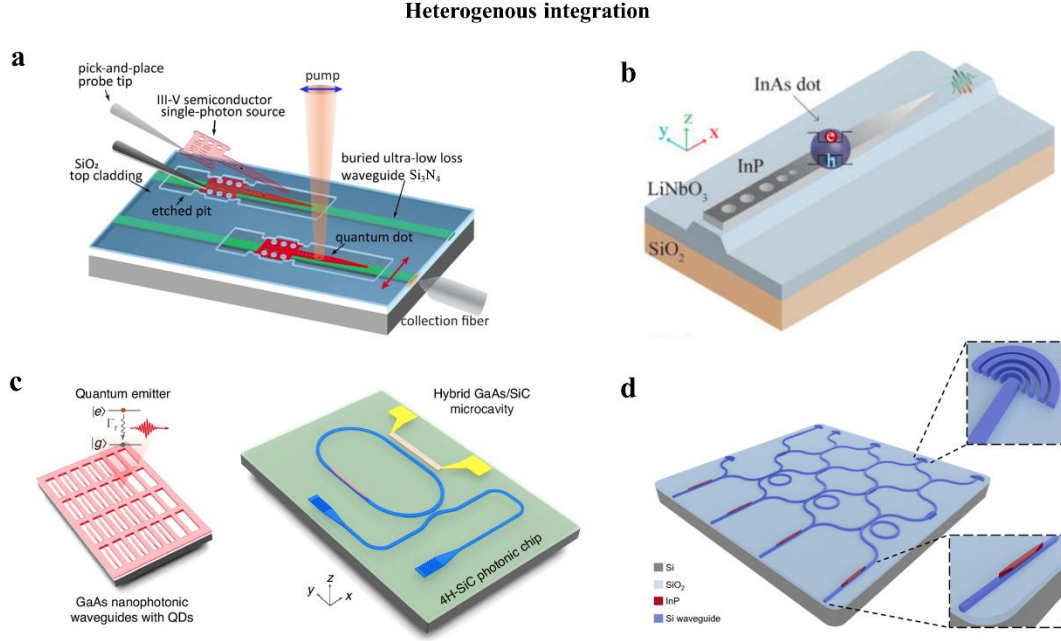


Figure 1.3. Examples of heterogenous integration using different method: (a-b) pick-and-place method, (a) GaAs nanophotonic devices containing InAs QDs were picked up onto an ultra-low loss Si_3N_4 waveguide^[88]. (b) InAs QDs embedded in an InP nanobeam with Bragg reflector on one side and the adiabatic taper on the other side, this structure was integrated with LiNbO_3 waveguide for high-efficient single photons^[89]. (c) Transfer-printing method: tapered GaAs nanophotonic waveguides with QDs transferred to 4H-SiC waveguides, which were prepared by electron beam lithography and plasma dry etching techniques, separately. The GaAs waveguides was transferred and placed onto the 4H-SiC resonator to form a hybrid GaAs/4H-SiC cavity^[90]. (d) Wafer-bonding method: InAs QDs grown epitaxially are integrated into a tapered InP waveguide placed on top of an SOI silicon waveguide, enabling efficient single-photon coupling into the chip and offering a scalable, cost-effective solution for multi-source quantum photonic systems^[91].

Heterogeneous integration involves the combination of multiple functional components fabricated from different material systems onto a single chip or substrate, typically through

wafer bonding, transfer printing or flip-chip assembly techniques. Heterogeneous integration enables the incorporation of diverse materials—each optimised for specific functionalities—within a unified photonic or electronic platform in providing a viable solution to enhance the circuit complexity, functionality and compatibility. Furthermore, it offers a flexible modular approach to build the reconfigurable quantum photonic integrated circuits and exploit the advantages of different material platforms. The integrations of QD single emitters into photonic nanostructures of different materials like Si, Si_3N_4 , LiNbO_3 , and SiC have been demonstrated. Figure 1.3 (a-d) present some examples of heterogeneous integration by three commonly used integration techniques: 1. Pick-and-place: III-V QDs embedded nano-waveguides are integrated onto (a) low-loss Si_3N_4 waveguide and (b) LiNbO_3 waveguide. 2. Transfer-printing: (c) GaAs waveguides with single QDs are transferred and placed onto the 4H-SiC resonator to form a hybrid GaAs/4H-SiC cavity. 3. Wafer-bonding: (d) Self-assembled InAs QDs embedded in tapered InP waveguide positioned over the SOI-defined Si waveguide. The first two methods allow for separate preparation of single-photon emitters and photonic circuits prior to assembling them together, thus affording high flexibility in fabrication and the possibility to preselect high performance quantum light source before integration. The pick-and-place approach uses robotic or mechanical actuators to individually position components, offering high placement precision but at the cost of lower throughput. In contrast, transfer printing typically employs an elastomeric stamp to retrieve and deposit multiple micro- or nano-devices from a donor to a target substrate, enabling high-throughput and scalable integration. Similarly, wafer-bonding allows scalable technique in which entire wafers or large-area dies from dissimilar materials are physically bonded through direct, adhesive or metal-assisted bonding methods.

1.2.3 Tunable emission wavelength

A key advantage of epitaxial semiconductor QDs for scalable quantum photonic technology is their emission wavelength tunability, enabling spectral matching between multiple emitters or alignment with the resonance mode of the optical cavities. The following briefly outlines several approaches to control the QD emission wavelength.

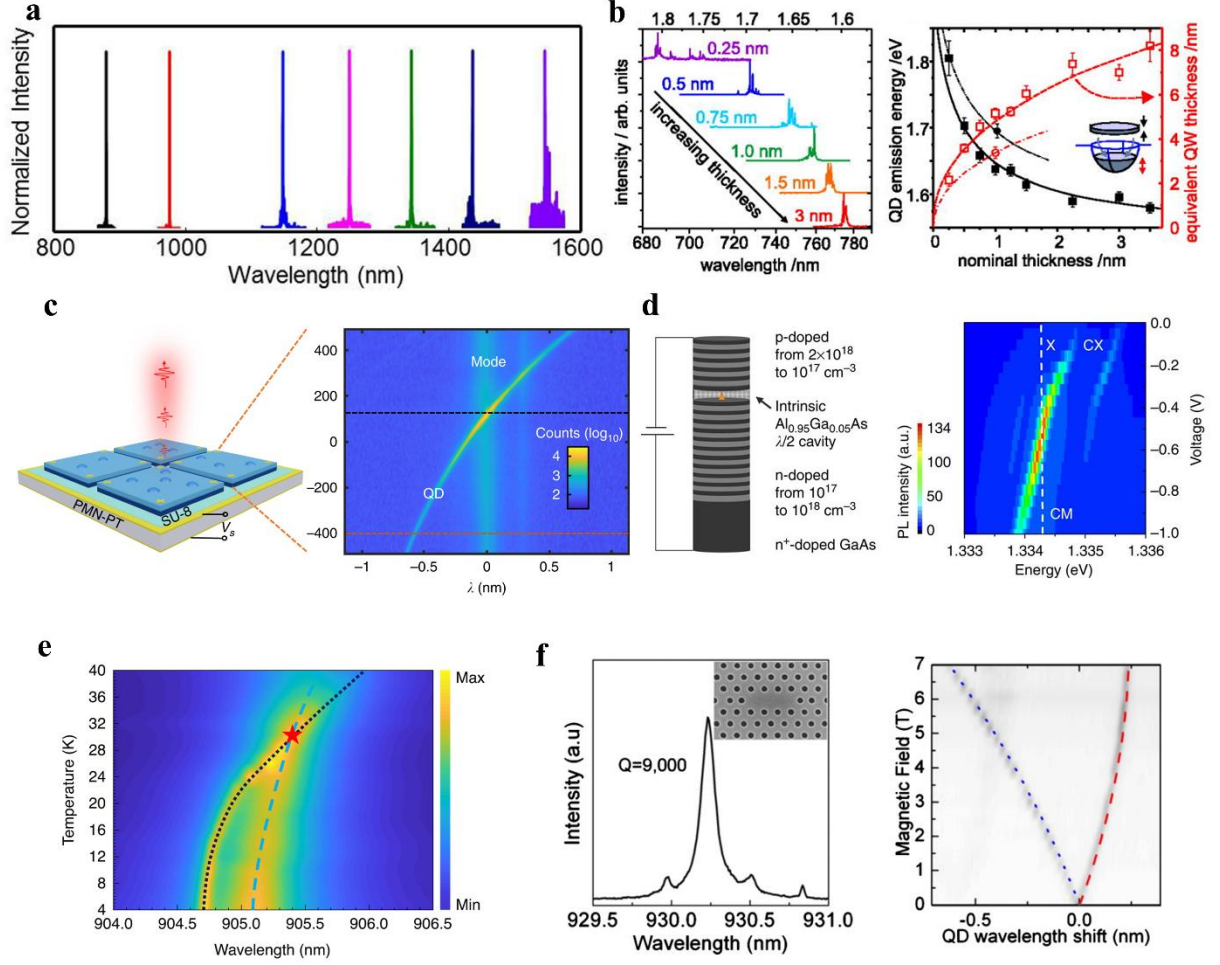


Figure 1.4. QD wavelength adjusting via different approaches. (a) InAsP QD in InP nanowire, wavelength shifting from 880 nm to 1550 nm by adjusting the arsenic composition and growth time (size) [92]. (b) GaAs QD wavelength shifts from ~690 nm to 780 nm with different GaAs thickness filling the holes formed by Ga droplet local etching, and corresponding theoretical calculation of QD emission energy [93]. (c) Strain running of QD wavelength to couple to cavity mode by sweeping the applied voltage from -500 V to 500 V [94]. (d) Electrically tuning the exciton state of QD to couple to cavity mode by applying electrical field to a p-i-n diode pillar [95]. (e) Temperature dependent to red shift the QD wavelength to couple to cavity mode of the ring [96]. (f) Magnetic field tuning of a QD strongly coupled to photonic crystal cavity [97].

(1) Engineering QD size and composition

The emission wavelength of QDs can be primarily defined during the epitaxial growth process by controlling parameters such as dot size and composition to engineer their band gaps^[98]. For instance, Figure 1.4 (a), by increasing the QD growth time (size) and the arsenic composition, the InAsP QD wavelength can be shifted from 880 to 1550 nm. Similarly, in Figure 1.4 (b) the increasing thickness of GaAs filling thickness in the hole created by Ga droplet local etching, resulting a red shift of the QD emission wavelength. This approach enables emission tuning across a wide spectral range, including the telecom bands. Though limited to coarse and stochastic tuning across ensembles, growth engineering remains critical for fabricating QDs targeted at specific wavelength regimes.

(2) Strain tuning

The lack of symmetric distribution between $[110]$ and $[1-10]$ direction for QDs grown along the $[001]$ direction results in energy level splitting - the so-called fine structure splitting, thereby limiting the fidelity of entangled photon pairs^[99]. Direction strain tuning modifies the QD band structure based on multi-axis strain^[100] control by applying controlled mechanical deformation using piezoelectric substrates or microelectromechanical systems^[101]. In Figure 1.4 (c), by integrating QDs onto a piezo-actuator and applying a strain field, the spectral mismatch between QD emission and the cavity mode can be compensated. This technique allows for broadband and highly stable spectral tuning over tens of μeV without introducing significant decoherence or altering the internal structure of the QD^[102].

(3) Electric field tuning

Electric field tuning exploits the quantum-confined Stark effect^[103, 104], wherein an external bias (electric field) applied across a QD embedded in a diode structure shifts its emission energy levels. This allows for continuous, reversible tuning of the QD emission typically in the range of a few to tens of meV. As seen in Figure 1.4(d) the QD emission lines shift towards lower energy and the excitonic emission is tuned across the cavity mode by applying a negative voltage. Beyond tuning, electric fields can also control charge occupancy and suppress fine-structure splitting, enabling generation of entangled photon pairs^[105-107].

(4) Temperature tuning

Temperature tuning relies on the temperature dependence of the QD bandgap energy. As the temperature increases, the bandgap narrows, resulting in a redshift of the emission wavelength

^[108, 109]. As seen in Figure 1.4 (e), by increasing the temperature to redshift the QD wavelength which will also redshift the cavity mode (albeit at a much slower rate), they couple together at one point. This approach is straightforward and reversible, and can offer a tuning range of several meV. However, it often comes at the expense of increased phonon interactions, which degrade coherence and photon indistinguishability ^[110]. Therefore, it is mainly used for coarse tuning or for aligning QDs with cavities at cryogenic conditions.

(5) Magnetic field tuning

Magnetic field tuning modifies the QD emission spectrum by introducing Zeeman splitting and diamagnetic shifts in the excitonic energy levels ^[111]. When a magnetic field is applied, the degeneracy of spin states is lifted, leading to the splitting of exciton and biexciton lines into spin-resolved components ^[112, 113]. This allows for precise control over spin-selective optical transitions, which is essential for realising spin-photon interfaces—a key building block for quantum memories, spin-based qubits and entanglement protocols ^[114]. Figure 1.4 (f) shows the Zeeman splitting exciton lines of an InAs QD strongly coupled to a photonic crystal cavity by magnetic field tuning.

1.2.4 Site-controllable fabrication

The conventional epitaxial growth of single QDs on planar substrates typically results in random spatial and spectral distribution ^[115], which poses a significant challenge for their use in scalable quantum photonic networks. Without precise site control, it is difficult to deterministically position QDs within the photonic structures or to spectrally match multiple emitters—a requirement for quantum interference and multi-photon entanglement ^[116]. Furthermore, planar QD structures generally suffer from poor light extraction efficiency ^[117], due to total internal reflection and isotropic emission, which seriously limits their brightness and usefulness in quantum applications that require high-purity, bright and indistinguishable single or entangled photons. To overcome these limitations, site-controlled QD growth and QD–cavity integration techniques have been developed. Single QDs can be fabricated either by bottom-up or top-down approaches, allowing deterministically position QD at predefined locations with high spatial precision. When coupled to optical microcavities such as micro-ring resonators, photonic crystal cavities, or micropillars, the emission properties of QDs can be significantly enhanced through the Purcell effect, leading to increase spontaneous emission rates and improved photon extraction efficiency ^[118, 119]. This cavity- quantum electrodynamics regime enables the realisation of bright, fast, and high-coherence single-photon sources, suitable for integration in on-chip quantum photonic circuits. As such, the combination of deterministic QD positioning with cavity-enhanced emission represents a critical advancement toward the development of practical and scalable solid-state quantum light sources.

a. Bottom-up epitaxial growth

Growth of single QDs on a pre-patterned substrate represents one of the most direct and effective strategies to achieve site-controlled quantum dot arrays, enabling deterministic integration with photonic and electronic devices. This approach can be realised either by directly patterning the substrate surface (e.g., through lithographically defined nanoholes or nanoislands) or by employing patterned dielectric masks such as SiO_x or SiN_x . The former approach patterns the substrate (such as InP, GaAs, GaN) directly with nanoholes arrays, using high-resolution lithography techniques such as nanoimprint lithography ^[120] and electron beam lithography ^[121] followed by etching, that serve as preferential nucleation sites for QD deposition. For example, single InAs QDs only nucleate inside the hole as shown in Figure 1.5 (a-b). However, the defects created by the etching process result in low QD quality and nonradiative recombination, thus lower the quantum efficiency ^[122]. In contrast, the latter

approach by growing on the patterned dielectric mask, which provides spatial confinement for adatom diffusion and defines nucleation sites, is superior and can achieve QDs with high crystal quality and low defect density. This method has been successfully applied to grow a wide range of III–V semiconductor nanostructures such as those shown in Figure 1.5 (c–d), including InAs QDs on GaAs^[123, 124], InGaN QDs on GaN^[125, 126], and InAsP QDs in InP nanowires (NWs)^[127, 128] □ each tailored for specific optoelectronic or quantum applications. Moreover, dielectric-mask-based selective area epitaxy (SAE) is inherently scalable and compatible with CMOS processing, making it a promising route toward the mass production of deterministic quantum emitters integrated within photonic circuits or microcavity systems.

In addition to conventional lithography-based techniques, several in situ substrate patterning approaches have been developed to enable site-controlled growth of single QDs without the need for complex lithographic alignment or masking steps. These techniques directly engineer the substrate morphology or surface energy during growth preparation, offering an elegant and potentially more scalable solution for deterministic QD placement. One notable example is the substrate-encoded size-reducing epitaxy technique^[129], as illustrated in Figure 1.5(e). In this method, a pre-patterned substrate undergoes a controlled overgrowth process that gradually reduces the lateral size of the pattern features. This engineered evolution of surface morphology results in sharply localised growth fronts, enabling the deterministic formation of single QDs at precise spatial locations. This approach provides tight control over both QD positioning and vertical alignment, which is critical for enhancing light extraction efficiency and Purcell enhancement when integrating QDs into resonant photonic structures such as distributed Bragg reflectors or microcavities^[130–132]. Another in situ strategy utilises laser interference to create periodic nanoisland arrays on the substrate surface, as shown in Figure 1.5(f). By interfering coherent laser beams at specific angles, a large-area periodic pattern of intensity maxima is generated, which is then transferred to the substrate through material modification or etching^[133]. These nanoislands act as preferential nucleation sites for QD growth due to local variations in surface energy or strain, effectively guiding InAs QD nucleation with high regularity and periodicity. This technique is highly attractive for large-scale fabrication, as it can rapidly pattern centimeter-scale areas with high spatial uniformity without the need for serial writing.

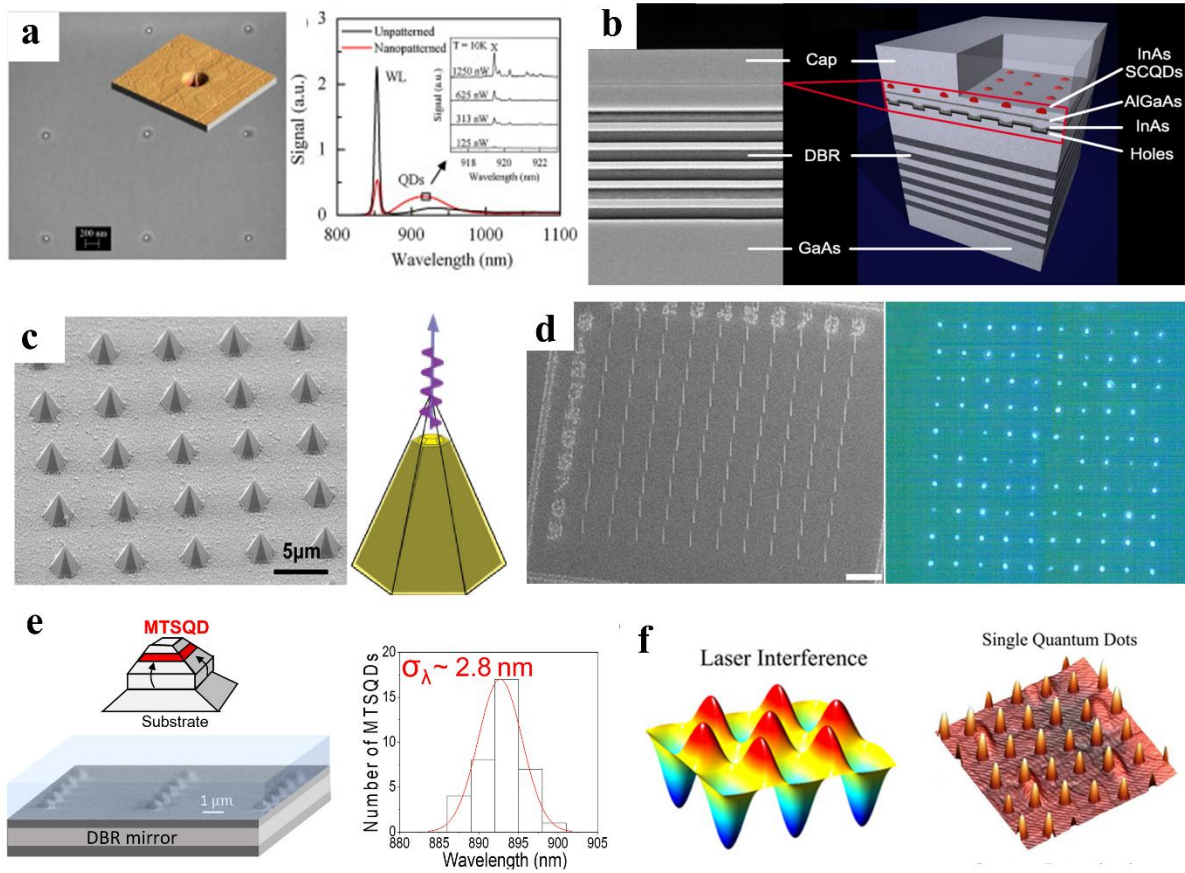


Figure 1.5. Bottom-up site-controllable growth of single quantum on pre-patterned substrate. Patterns on substrate directly: (a) Soft UV-nanoimprint lithography creates hole arrays on GaAs substrate for InAs QD deposition ^[122]. (b) Etching of nanohole arrays as nucleation centre for QD growth ^[134]. Selective epitaxy on patterned dielectric substrate: (c) InGaN single QDs on GaN pyramid array ^[72], (d) InAsP single QDs in InP NW array ^[135]. In situ approaches for patterning: (e) Substrate encoded size reducing epitaxy for single QD array on DBR mirror ^[136]. (f) Laser interference for nanoisland array creation, which can as preferential nucleation sites for InAs QDs ^[137].

b. Top-down nanoscale positioning

Many deterministic fabrication methods have been developed to preselect a single QD with the desired optical properties, such as suitable wavelength, narrow linewidth and high brightness, by a combination of nanoscale positioning techniques. Some early works used microscopy imaging, like scanning electron microscopy and atomic force microscopy in figure 1.6 (a-b), to directly position a single QD on a photonic crystal cavity and achieve strong coupling effect.

However, the epitaxial QDs used in quantum device applications are usually buried beneath a capping layer (larger bandgap than QD materials for quantum confinement) ^[138, 139], preventing adverse influence from the environment and thus enabling stable optical properties. For positioning these invisible QDs, a combination of luminescence measurement before fabrication is needed. A cathodoluminescence (CL) -assisted in-situ electron beam lithography (EBL) has been developed in Figure 1.6 (c) to identify and check the optical properties which is patterning the area around the QD for subsequent fabrication. This method allows direct fabrication of QD in cavity with a spatial resolution ~ 40 nm. Other studies also demonstrated the integration of QDs with different cavities to achieve high quality quantum light sources, indicating a promising path toward significant scaling of chip-based quantum photonics ^[140, 141].

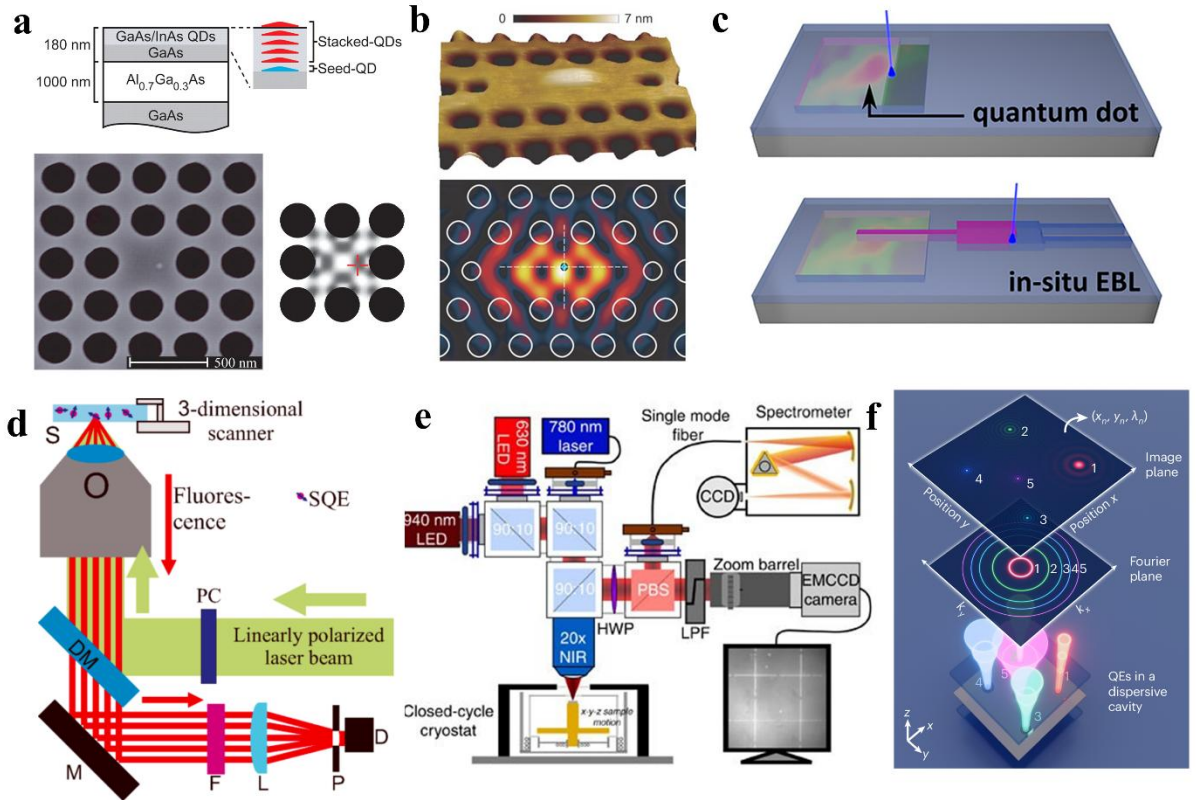


Figure 1.6 Nanoscale positioning skills based on top-down fabrication. (a) Scanning electron microscope is used to image the InAs QD (white dot), which is grown on the GaAs photonic crystal cavity with the layer structure shown in the top panel ^[142]. (b) Atomic force microscopy is used to identify the QD position on a photonic crystal nanocavity, enabling its alignment with the cavity electric field maximum, as shown in the bottom panel ^[143]. (c) In-situ electron beam

lithography of deterministic integration of an InAs QD into a 50/50 multimode interference beam splitter using low-temperature cathodoluminescence spectroscopy ^[144]. (d) Laser-scanning confocal microscopy system for visualisation of arbitrarily oriented single emitters ^[145]. (e) Wide-field PL imaging system to identify the emitting QDs by exciting a large area of sample via a LED then detected by a camera ^[146]. (f) Super-resolved nanophotonic snapshot hyperspectral imaging technique for high efficiency extract both the spatial position and emission wavelength of a large number of single quantum emitters ^[147].

The limitation of CL-assisted EBL in-situ fabrication method is inability to determine single photon emission of the QD before fabrication. To overcome this issue, a few ex-situ optical methods have been developed. Laser-scanning confocal microscopy has been broadly used to position the single emitters, the system setup as seen in Figure 1.6 (d). When the excitation laser scans across the sample, a point-by-point collection will show a photoluminescence (PL) map of the sample. The position of the isolated fluorescing objects can be determined at a level well below the diffraction limit, and the position accuracy can be as high as 10 nm ^[148, 149]. However, the largest scanning area is limited by the size of confocal plane (usually within 100 μm) and the time duration for scanning is long, limiting the overall throughput of this approach. In contrast, wide-field PL imaging-based positioning technique enable higher throughput and spatially multiplexed of the emitters by exciting a large scale of sample with normally a LED and collecting the luminescence information of the whole area by a camera as seen in Figure 1.6 (e). Thus, wide-field PL imaging has the advantage of extracting in a parallel fashion the spatial positions of a large number of quantum emitters with an accuracy of deep subwavelength using the super localisation technique ^[150]. However, the ultimate goal for these positioning skills is to select a promising candidate with highly spectral matching between the single emitters and the nanostructure cavity modes. Therefore, one-by-one individually spectrum measurement is unavoidable, which makes it as the most time-consuming process with low efficiency. To overcome these bottlenecks, Liu et.al developed a super-resolved nanophotonic snapshot hyperspectral imaging technique as seen in Figure 1.6 (f), by taking advantage of the wide-field imaging technique, enabling the simultaneous extraction of both spatial position and emission wavelength of a large number of single emitters, with a spatial and spectral accuracy down to 15 nm and 0.4 nm. This technique shows the ability for future large-scale integration of compact quantum photonic devices with various functionalities.

1.3. Challenges

Despite QD-based solid-state emitters having shown their capability to generate single photons on-demand, highly pure and indistinguishable single photons for quantum information technology application, several key challenges remain. Firstly, spectral inhomogeneity across self-assembled QDs poses a major obstacle to scalable systems, as slight differences in size, shape, and composition lead to non-uniform emission wavelengths, limiting photon indistinguishability between sources. Secondly, random spatial positioning in conventional growth methods makes deterministic coupling of QDs to photonic structures (e.g., cavities, waveguides) difficult, reducing coupling efficiency and scalability. Thirdly, while high optical quality can be achieved at cryogenic temperatures, room-temperature operation remains challenging due to phonon interactions that degrade coherence and photon indistinguishability, especially for low band gap material of In(Ga)As/GaAs, In(As)P/InP, GaAs/AlGaAs. Additionally, charge noise, spectral diffusion, and blinking can impact photon purity and stability. Integration with photonic integrated circuits also requires high-precision fabrication, materials compatibility (especially for hybrid systems like III-V QDs on silicon), and reproducibility across large wafers.

Despite these challenges, QD-based single-photon emitters is highly promising. Advances in site-controllable epitaxial growth and post fabrication, strain, magnetic field, electric field tuning, monolithic and heterogeneous integration techniques have enabled precise control over QD placement and emission properties. Progress in cavity quantum electrodynamics has enhanced brightness, purity, and speed via Purcell enhancement. Moreover, the development of telecom-wavelength QDs and frequency conversion techniques is opening the door to long-distance quantum networks compatible with existing fibre infrastructure. With continued improvement in fabrication precision, material platforms, and noise suppression, QD-based single-photon sources are poised to become a cornerstone of future on-chip quantum networks, quantum communication, and quantum computing architectures.

1.4. Thesis outline

In this thesis, we present a solution for the deterministic growth of InAsP single quantum dots within InP nano/micro-cavities using a metalorganic chemical vapour deposition (MOCVD) system, covering the development from optically pumped to electrically driven single-photon source applications. This thesis is divided into seven chapters.

In Chapter 2, we introduce the basic concepts and theoretical background relevant to this work, starting from the epitaxial growth in the MOCVD system, including its working mechanism, selective area epitaxy, and single QD growth. We then discuss cavity effects by positioning a single QD inside optical cavities to enhance single-photon optical performance. Finally, we introduce the basic concepts of single-photon sources and their excitation schemes, including both optical and electrical excitation.

In Chapter 3, we briefly introduce a series of experimental techniques involved in this work. These include techniques used for substrate preparation for selective area growth in the MOCVD system, device fabrication for electrically driven devices, characterisation methods for both material and optical properties, and theoretical simulations.

Starting from Chapter 4, we present the main research results of this PhD project. Chapter 4 introduces the combination of In droplet epitaxy with selectively grown InP NW arrays to deterministically position a single QD within an InP NW cavity. We systematically investigate the In droplet density, structural properties of both NWs and QDs, optical performance of the QDs, as well as theoretical simulations. Finally, we achieve scalable, bright, and pure single-photon emission from QDs embedded in nanowire arrays.

In Chapter 5, we further integrate single QDs into InP NW p–i–n diodes to fabricate electrically driven single-photon devices. The chapter begins with the pattern design used to reproduce the experimental results from Chapter 4, enabling the growth of a single QD in a single NW diode. With the aid of numerical simulations, we demonstrate electrically driven single quantum dot emission successfully coupled to the Fabry–Pérot mode of the NW. The device exhibits excellent performance in terms of single-photon purity, brightness, and long-term stability.

In Chapter 6, droplet epitaxy is further applied to bottom-up grown high-quality-factor InP micro-ring resonators. We systematically optimise the growth conditions to achieve high-Q InP rings with low-density In droplets on individual rings. The coupling of quantum dot emission into the whispering gallery modes of the rings results in Purcell-enhanced single-photon emission. Furthermore, we engineer the emitter–cavity coupling efficiency by optimising the

whispering gallery mode spacing, demonstrating high coupling probability and multi-wavelength single-photon emission with larger rings.

Finally, Chapter 7 summarises the main contributions of this thesis and provides three future research directions based on this thesis.

Chapter 2

2. Theoretical Background

This chapter introduces some of the theoretical background relevant to this thesis to provide a better understanding of the work. In section 2.1, some theoretical background knowledge related to epitaxial growth are discussed, including the mechanisms underlying MOCVD, selective area epitaxy, and single QD growth. Cavity quantum electrodynamic effect of single quantum dots in optical cavities is presented in Section 2.2 with photon antibunching in Section 2.3 and quantum dot excitation schematic in Section 2.4. These contents are helpful to better understand the epitaxial growth and characterisation of InAsP single QDs in InP optical cavities for single photon source.

2.1 Epitaxial Growth

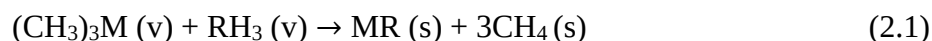
This section introduces the fundamental principles and processes involved in epitaxial growth using MOCVD, as well as the growth strategies of the fabrication of single QDs.

2.1.1 Basic mechanism of MOCVD

MOCVD is one of the most widely employed epitaxial growth techniques for fabricating high-quality III–V compound semiconductors such as GaAs, InP, InAs, and their alloys. Compared to molecular beam epitaxy, which operates under ultra-high vacuum and offers exceptional control over atomic-layer deposition, MOCVD is more scalable and industry-compatible, making it the preferred method for large-scale device manufacturing. Moreover, MOCVD enables higher growth rates, better material uniformity over large wafer areas, and flexibility in chemical composition tuning through the use of metal-organic precursors. Its ability to precisely control layer thickness, composition, and doping at the monolayer scale makes MOCVD particularly attractive for applications requiring both high crystal quality and integration scalability, such as quantum photonic devices, light-emitting diodes, and high-speed transistors.

The growth of III–V materials such as InP, GaAs, InAsP, and related ternary/quaternary alloys is achieved through the chemical reaction of group III metal-organic precursors (e.g.,

trimethylindium (TMIn), trimethylgallium (TMGa) with group V hydride gases (e.g., AsH₃, PH₃) in the vapour phase. The process occurs in either a horizontal or vertical flow reactor, where the precursors are introduced via carrier gases (typically H₂ or N₂) and react thermally on a heated crystalline substrate. The processes involved in the crystal growth are extremely complex, but can be simplified as:



where M represents metal in the metal-organic compounds (Ga, In, etc), R represents group V materials like P and As.

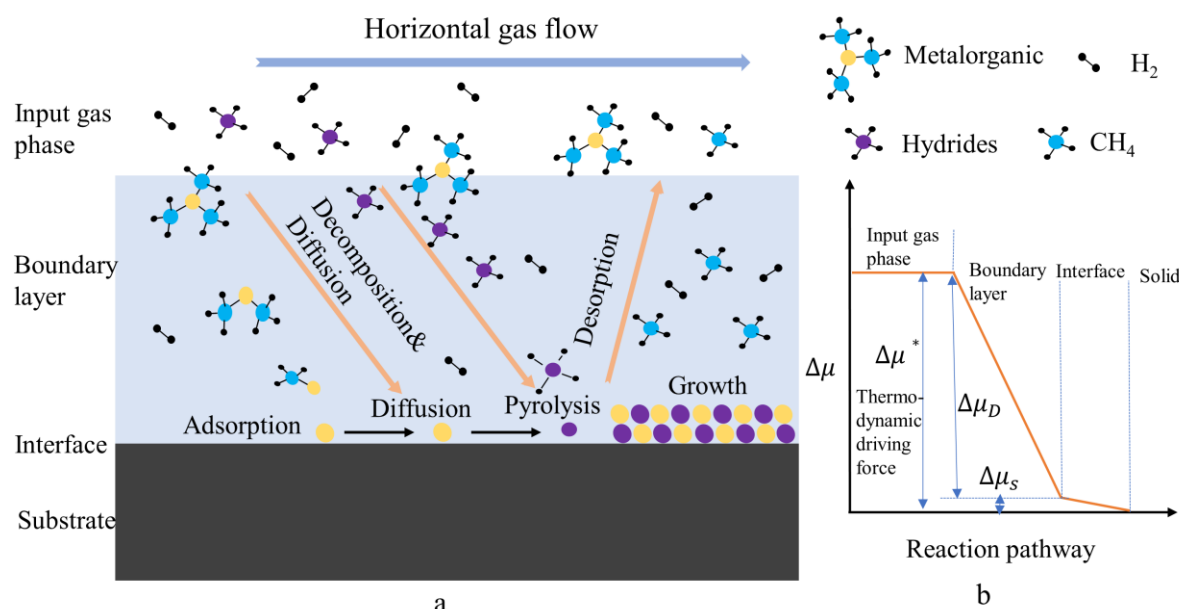


Figure 2.1. MOCVD growth mechanism. (a) Schematic diagram illustrates the fundamental growth process in MOCVD for III-V crystal growth. The material growth typically involves gas phase transport and adsorption, surface diffusion, Pyrolysis and decomposition, incorporation and layer growth, desorption of by-products. (b) Chemical potential versus reaction coordinate, showing the drop in chemical potential required for each step as a thermodynamic driving force in the growth sequence.

As illustrated in Figure 2.1(a), the MOCVD growth process involves a series of surface and gas phase interactions:

(a) Gas Phase Transport and Adsorption

The precursors are carried to the substrate by laminar horizontal gas flow. Upon reaching the substrate, the metalorganic and hydride molecules adsorb onto the surface. This adsorption step is essential for initiating surface reactions and is influenced by substrate temperature, surface energy, and precursor flux.

(b) Surface Diffusion

After adsorption, the precursor molecules or their decomposition fragments undergo lateral surface diffusion. This mobility allows atoms to locate energetically favourable lattice sites, contributing to crystal quality and uniform coverage.

(c) Pyrolysis and Decomposition

The adsorbed molecules thermally decompose (pyrolyse) near the substrate surface. For instance, TMIn breaks down into indium adatoms and methyl by-products (e.g., CH_3), while hydrides such as PH_3 release group V elements (P, As). These atomic species are then available for incorporation into the growing lattice.

(d) Incorporation and Layer Growth

The group III and V adatoms react and incorporate into the crystalline substrate following the lattice orientation. This epitaxial process results in atomically smooth, compositionally uniform films. The growth rate is determined by the decomposition kinetics and surface incorporation efficiency.

(e) Desorption of By-products

Volatile reaction by-products (e.g., CH_4 , H_2) desorb from the substrate and are removed via the carrier gas flow. Efficient desorption is critical to avoid surface contamination and ensure a clean growth environment.

The overall crystal quality and composition of the grown layers depend heavily on key process parameters, including substrate temperature, V/III ratio, chamber pressure, and precursor flow rates.

A nonequilibrium situation is intentionally created to form the driving force for solid state product growth in the MOCVD system. Figure 2.1 (b) illustrates chemical potential drops in each step as a thermodynamic driving force in the growth. The overall thermodynamic driving force is the chemical potential difference between precursors in gas phase and atoms in solid phase, representing by $\Delta\mu^*$, * represents the chemical potential of the input gas phase. The growth rate is proportional to the atomic flux transported—typically via diffusion—through the gas phase to the interface, which equals the flux of atoms incorporated into the solid across the interface. This schematic illustrates the driving force necessary to sustain this flux ($\Delta\mu_D$) for the diffusion process and the surface reaction ($\Delta\mu_S$). Thus, for the growth of III-V solid compounds, the driving force can be described as:

$$\Delta\mu = \mu_{III}^v + \mu_V^v - \mu_{III-V}^S = RT \times \ln \frac{P_{III}P_V}{P_{III}^e P_V^e} \quad (2.2)$$

where the middle part represents the chemical potential of the group III and group V gas reactants and grown solid state III-V compound; R, T represent the gas constant and temperature, respectively. P_V (P_V^e) and P_{III} (P_{III}^e) are the partial pressures of group V and group III, respectively, with the superscript e representing the partial pressure at equilibrium. Even the whole process is nonequilibrium, near-equilibrium condition maybe exist at the growing solid state surface with a large supersaturation in the input vapor phase ($\Delta\mu^* \gg 0$), which requires the interface kinetics to be much faster than the diffusion kinetic ($\Delta\mu_D \gg \Delta\mu_S$).

2.1.2 Selective Area Epitaxy

SAE is a powerful epitaxial growth technique that enables the site-controlled deposition of III–V nanostructures by locally modulating growth using a patterned dielectric mask—typically SiO_2 or Si_3N_4 —on the substrate surface. Unlike conventional blanket growth, SAE confines crystal growth to exposed openings (windows) defined by lithography, while growth is suppressed in the masked regions. The detail sample preparation processes for SAE will be discussed in Chapter 3.2. This technique is highly suitable for the integration of low-dimensional quantum structures (e.g., quantum dots, quantum wells, nanowires) and heterogeneous photonic architectures on-chip.

Figure 2.2 schematically illustrates the mass transport mechanisms during SAE of InP nanowires on a patterned InP substrate. In the initial stage of growth (Figure 2.2a), group III precursors (TmIn) and group V hydrides (PH_3) are introduced into the reactor. Growth occurs

selectively within the openings on the patterned substrate, while no deposition occurs in the surrounding mask area. Adatoms are supplied through two main pathways: surface diffusion from the surrounding mask region (process 1) and direct impingement into the window openings (process 2). These adatoms accumulate within the exposed substrate area, promoting vertical nucleation and the onset of nanowire formation.

Once the hole is filled and axial nanowire growth initiates (Figure 2.2b), material continues to be incorporated both axially, at the nanowire tip, and laterally, along the sidewall facets (process 3). The competition between axial and lateral growth can be controlled by the growth parameters. Furthermore, the pitch between the openings—defined as the centre-to-centre distance between adjacent openings—can also play a crucial role in regulating local adatom flux and nanowire morphology. At smaller pitch size, the competition for adatoms between neighbouring openings increases, leading to reduced effective material supply per site and lower radial growth rates. Moreover, closer spacing enhances the likelihood of nanowire coalescence due to lateral overgrowth, which can degrade verticality and uniformity. In contrast, larger pitch size allows more efficient adatom collection per site, enhancing radial growth and enabling better separation between nanowires, albeit at the cost of lower integration density.

Therefore, precise optimisation of pitch size, in combination with growth parameters such as temperature and V/III ratio, is essential to achieve high aspect ratio, uniform, and vertically aligned InP nanowire arrays. Understanding and controlling these mechanisms is critical for the deterministic fabrication of site-controlled quantum emitters and photonic devices based on nanowire platforms.

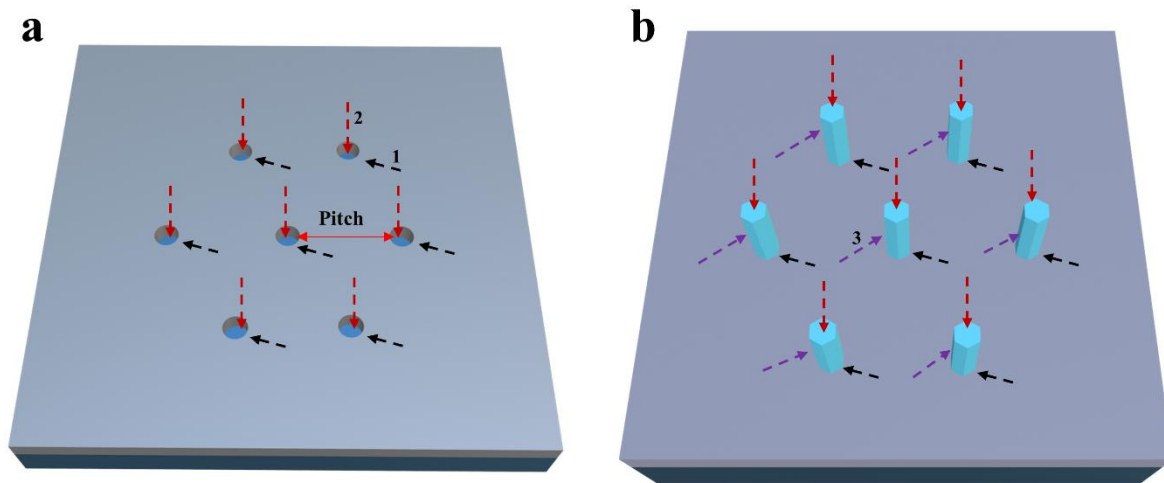


Figure 2.2. Schematic illustration of mass transport processes for growth of SAE nanowire with a fixed pitch. (a) The initial stage of adatoms filling the holes, from (1) mask diffusion (black arrow) and (2) direct growth (red arrow) in the hole opening. (b) After filling the hole, nanowire growth occurs both axially and laterally. (3) Indicates lateral growth at the sidewall facets of the nanowire (purple arrow).

In addition to morphology, the crystal phase of InP nanowires grown via SAE is strongly influenced by the local growth conditions and kinetic environment. III–V semiconductors like InP typically crystallise in the zinc blende structure in bulk form; However, when grown in nanoscale geometries under nonequilibrium conditions—especially at reduced dimensions and higher growth temperatures—wurtzite phase segments can emerge. The competition between wurtzite and zinc blende phases during nanowire growth is governed by several factors, including growth direction, V/III ratio, temperature, and growth rate.

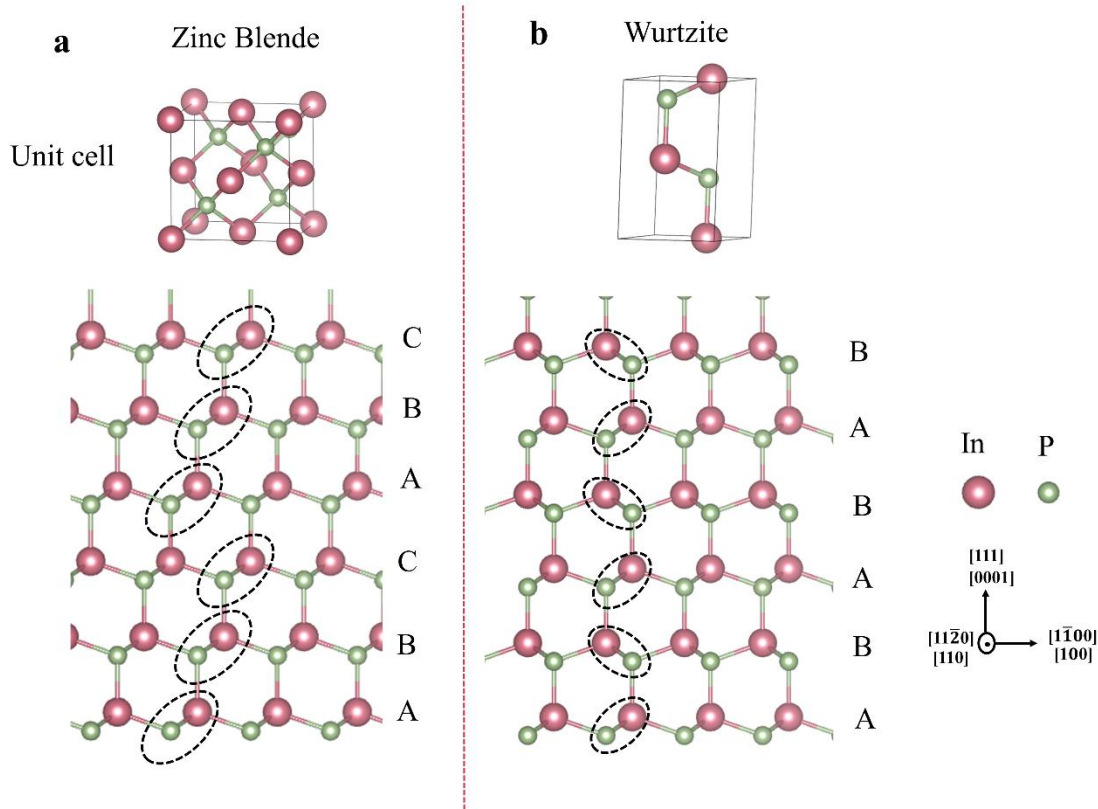


Figure 2.3. Ball-and-stick models of the two crystal structures. (a) cubic zinc blende and (b) hexagonal wurtzite unit cell, and their corresponding atomic plane stacking sequence, viewed from $[110]$ or $[11\bar{2}0]$ direction, respectively. The dashed line circles represent the repeating sequence with ABCABC... in zinc blende and ABAB... in wurtzite, respectively ^[151].

Once the III-V nanostructure is grown, their crystal structure needs to be investigated. Figure 2.3 presents the ball-and-stick models of the two fundamental crystal structures: (a) zinc blende and (b) wurtzite. From the unit cell of each structure, both structures are composed of tetrahedrally coordinated group III and group V atoms; however, they differ in their atomic stacking sequence and crystal symmetry. The zinc blende structure, crystallises in a cubic lattice with an ABCABC... stacking sequence along the $[111]$ direction. In contrast, the wurtzite structure adopts a hexagonal lattice and follows an ABAB... stacking sequence along the $[0001]$ (c-axis) direction. The dashed circles highlight the repeating bilayer sequences: in zinc blende, every third layer repeats, while in wurtzite, the sequence alternates every two layers. This variation in stacking results in differences in symmetry and physical properties, such as band structure, spontaneous polarisation, and strain relaxation behaviour. The easily distinguishable atomic stacking sequence along the $[110]$ or $[11\bar{2}0]$ directions helps the investigation of growth materials using atomically resolved transmission electron microscopy.

2.1.3 Self-assemble single quantum dot

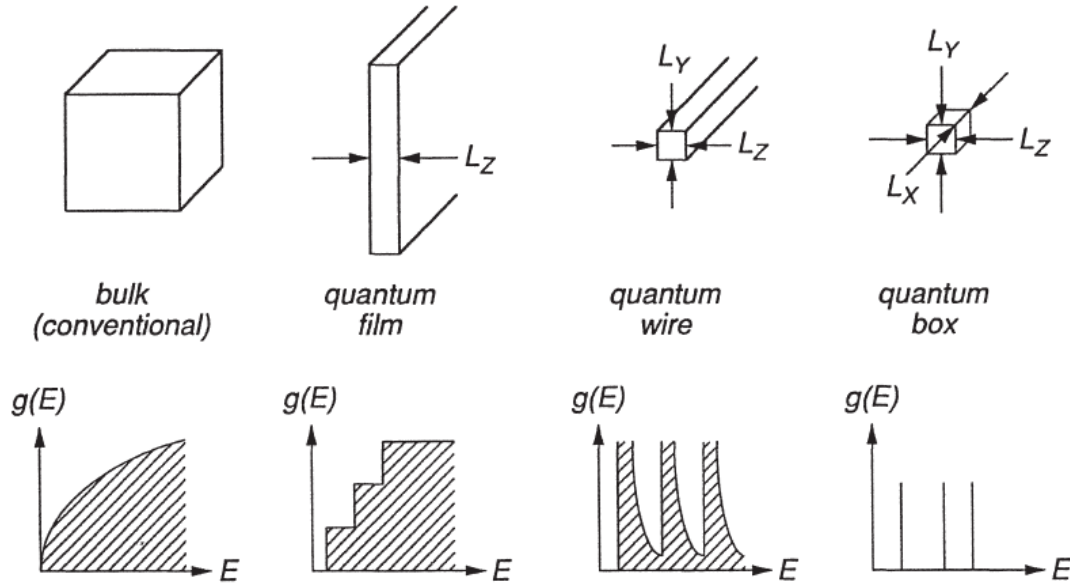


Figure 2.4. Quantum confinements in nanostructures and their density-of-states of electrons as a function of energy ^[152].

Self-assembled III–V QDs are semiconductor nanostructures that confine charge carriers—electrons and holes—in all three spatial dimensions. As depicted in Figure 2.4, quantum confinement becomes increasingly significant as the dimensionality of the structure is reduced. In a bulk (3D) semiconductor, carriers are free to move in all directions, and the density of states, $g(E)$, increases as a square root of energy, resulting in a continuous energy spectrum. When carriers are confined in one dimension (quantum well or film), the density of states becomes step-like. Further confinement in two dimensions (quantum wire) causes the density of states to exhibit sharp peaks (Van Hove singularities). Finally, in quantum boxes or QDs, carriers are confined in all three spatial directions, leading to discrete atomic-like energy levels and a delta-function-like density of states. This transition from continuous to discrete states profoundly impacts the optical and electronic properties of the material.

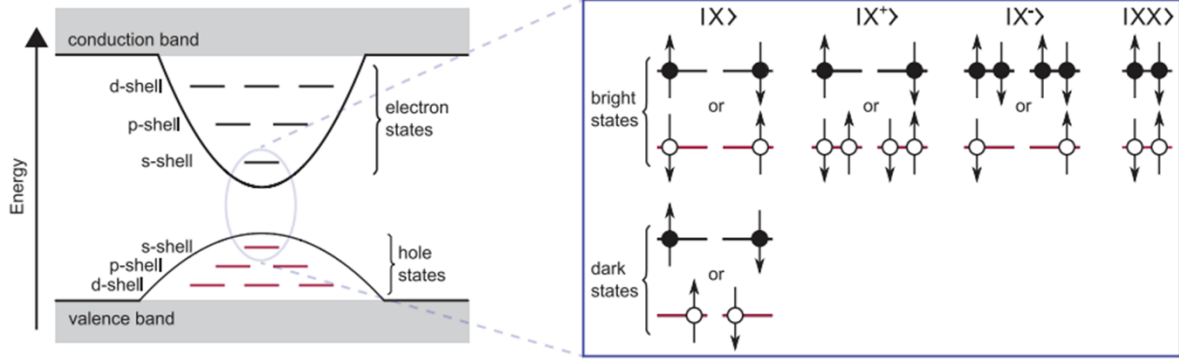


Figure 2.5 Schematic energy potential of a QD ^[153]. The electron states (black lines) and hole states (red lines) can be named as s-shell, p-shell, d-shell, et.al, depending on effective masses. The right panel shows a zoomed-in view of the nine s-shell carrier configurations. The resulting quantum states are defined by the number of electrons (filled circles) and holes (open circles), as well as their respective spin orientations (indicated by arrows). The dark exciton state corresponds to a parallel spin configuration, rendering it optically inactive and leading to a longer lifetime.

QDs confine electrons and holes in all three spatial dimensions, leading to a discrete energy level structure that resembles the orbital structure of atoms. These discrete energy levels, shown schematically in Figure 2.5, are typically labelled as s-, p-, and d-shells, reflecting the angular momentum characteristics of the wavefunctions. The shell structure arises from the harmonic or parabolic potential that approximates the confinement landscape of the QD, with level spacing and degeneracy governed by the lateral size of the dot and the effective masses of the carriers. Electrons, which typically have smaller effective mass than holes, occupy higher energy levels under the same confinement potential. Within the lowest-energy s-shell, combinations of single and multiple carrier occupancies result in a range of few-particle quantum states, defined not only by the number of confined carriers but also by their spin configurations.

As illustrated in the right panel of Figure 2.5, different excitonic configurations can be formed depending on the number and spin states of electrons and holes occupying the s-shell. These include the neutral exciton (X), positively and negatively charged excitons (X^+ and X^-), and the biexciton (XX). For the neutral exciton, an electron-hole pair can form either bright or dark exciton states depending on their spin configuration. The bright exciton states ($J_z = \pm 1$) are

optically allowed and are initially associated with opposite circular polarisations, whereas the dark exciton states ($J_z = \pm 2$) are optically forbidden. The charged excitons contain an additional hole (X^+) or electron (X^-), while the biexciton consists of two electron–hole pairs occupying the same QD. The biexciton is particularly important for quantum light generation, as its radiative decay through the $XX-X$ cascade can generate pairs of polarisation-entangled photons when fine-structure splitting (FSS) is sufficiently small.

In an ideal quantum dot with high in-plane symmetry, the two bright exciton states are degenerate. However, in real InAs quantum dots, shape anisotropy, strain, and anisotropic electron–hole exchange interactions lift this degeneracy, giving rise to FSS. The resulting eigenstates are two orthogonally and linearly polarised states, conventionally denoted H (horizontally polarised) and V (vertically polarised). The magnitude of the FSS is particularly important for polarisation-entangled photon generation through the biexciton–exciton ($XX-X$) cascade, as a finite FSS can provide the path information and reduce the degree of photon entanglement.

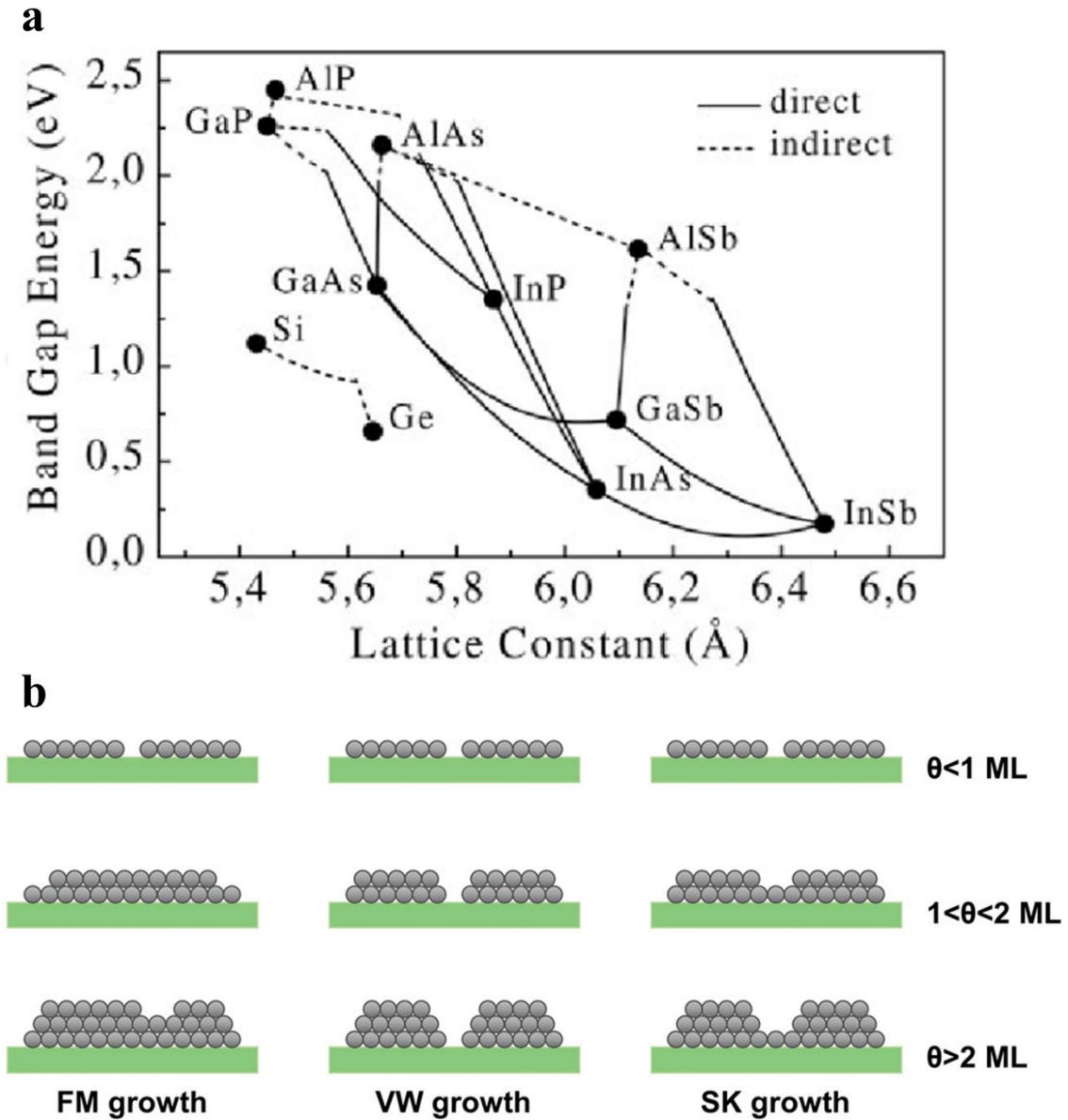


Figure 2.6. The relationship between epitaxial growth and lattice constant. (a) Lattice constant versus band gap energy of some common IV, III-V, and related semiconductor compounds ^[154]. (b) three primary epitaxial growth modes: Frank–van der Merwe, Volmer–Weber, and Stranski–Krastanov ^[155].

The formation of self-assembled III–V semiconductor QDs via epitaxial techniques is fundamentally governed by the lattice mismatch between the epitaxial layer and the underlying substrate. As illustrated in Figure 2.6 (a), a plot of band gap energy versus lattice constant for common group IV, III–V, and related compound semiconductors with solid line representing direct band gap and dashed line representing indirect band gap semiconductors. With similar

lattice constants (e.g., GaAs and AlAs), smooth planar layer can be grown. However, when a material with a significantly different lattice constant is deposited onto a substrate (e.g., InAs on GaAs or InP), strain builds up in the film, which profoundly alters the epitaxial growth mode. There are three primary epitaxial growth modes as seen in Figure 2.6 (b): Frank–van der Merwe (FM), Volmer–Weber (VW), and Stranski–Krastanov (SK). In the FM or layer-by-layer growth mode, the adatoms have stronger interactions with the substrate than with each other, leading to the formation of successive atomic monolayers. This occurs under low lattice mismatch conditions and is typically used to grow smooth quantum wells, superlattices, or bulk materials. In contrast, VW or island growth occurs when adatom–adatom interactions dominate, and the deposited material poorly wets the substrate. This leads to immediate nucleation of 3D islands without an initial wetting layer, often observed in highly mismatched or chemically incompatible systems like GaN grown on sapphire. The SK growth mode, which is intermediate between FM and VW modes, is of particular interest for self-assembled QD formation. This mode usually happens between two materials which have lattice mismatch between $\sim 2\%$ to 8% .

Figure 2.7 (a) schematically illustrates the formation of QD by SK mode (such as InAs QDs grown on GaAs), the InAs initially form a few monolayers (wetting layer) on the GaAs substrate, but once the critical thickness is exceeded, accumulated elastic strain energy relaxes due to lattice mismatch, driving the transition from 2D layer to 3D island/dot formation. This strain-induced islanding allows coherent, dislocation-free QDs to form spontaneously if they are small enough. InAs on GaAs ($\sim 7\%$)^[156] and InAs on InP ($\sim 3.2\%$)^[157] are commonly used to realise high-quality, coherently strained self-assembled QDs. The growth of single QDs for single photon emission with density as low as 1 dot per μm^2 in SK mode needs the growth temperature, flux rate, as well as the deposition time to carefully adjusted, as the strain relaxation is hard to control. In contrast, droplet epitaxy (Fig. 2.7b) provides an alternative strain-independent approach to grow QDs on lattice-matched or mismatched substrates. In the conventional droplet epitaxy process, group-III metal droplets (Ga, In, etc) are first deposited under group-V-deficient conditions on the surface^[158-160]. These droplets are then crystallised by exposing them to group-V flux (As or P), leading to the formation of QDs. Since this process does not rely on lattice mismatch or strain, droplet epitaxy allows for more flexibility in substrate choice and typically enables symmetrical, dome-shaped dots. However, one of its drawbacks is the rather low temperature required during the crystallisation stage (to avoid bimodal growth), leading to low quality material and optical properties^[161, 162].

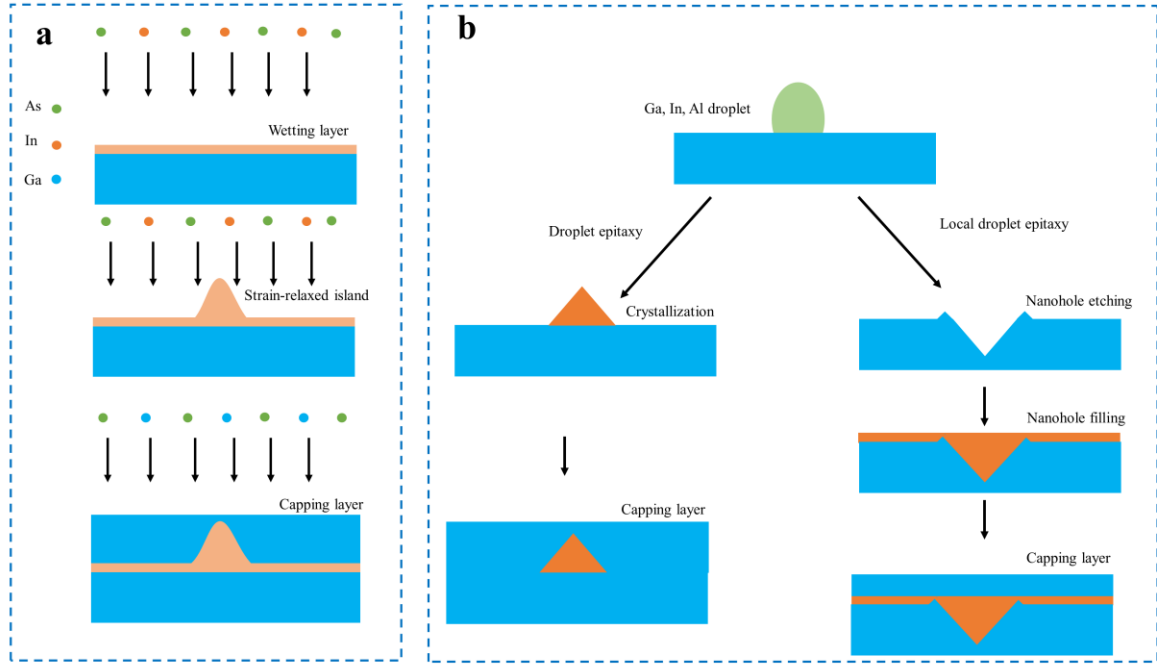


Figure 2.7. Self-assembled single QD growth approaches. (a) Stranski–Krastanov mode (b) droplet epitaxy mode, which can be divided into droplet epitaxy and local droplet epitaxy.

A different version of this technique, local droplet epitaxy, uses the metal droplets to drill the nanoholes for further QD deposition ^[68]. Local droplet epitaxy basically has four steps: (1) Metal droplet formation, as in droplet epitaxy; (2) Local etching to form nanoholes; (3) QD formation by hole filling; (4) QD capping to form confinement. The fine control of all parameters (e.g. temperature, droplet size, substrate facets) determines the nanohole depth, shape, and lateral extension. Local droplet epitaxy mode usually happens at high temperature, resulting in high quality, defect free, as well as strain free QDs with excellent optical properties ^[99, 163].

QDs grown on conventional (100) substrates often suffer from asymmetrical lateral strain and shape anisotropy, leading to anisotropic electron–hole exchange interactions and consequently to exciton fine structure splitting of the exciton levels, which degrades the polarisation entanglement of emitted photon pairs. However, both droplet epitaxy and local droplet epitaxy can be used on (111)A substrate with a C_{3v} crystal symmetry ^[164, 165], which is more isotropic in-plane, reducing the electron–hole exchange interaction. Combined with symmetric droplet nucleation, droplet epitaxy can lead to strong suppression of fine structure splitting, a critical requirement for entangled photon pair emission via the biexciton–exciton cascade ^[69, 166].

2.2 Quantum dot – cavity effects

In this section, we explore the effects of optical cavities on the emission properties of semiconductor QDs. Coupling QDs to resonant cavities enables control over spontaneous emission through cavity quantum electrodynamic effects. We first discuss the weak coupling regime, where the Purcell effect enhances the emission rate, and the strong coupling regime, marked by Rabi splitting and coherent energy exchange between the QD and the cavity mode. Additionally, we address how cavity structures can significantly improve light extraction efficiency, a key factor in realising high-performance quantum light sources for quantum information applications.

2.2.1 Cavity quantum electrodynamic effect

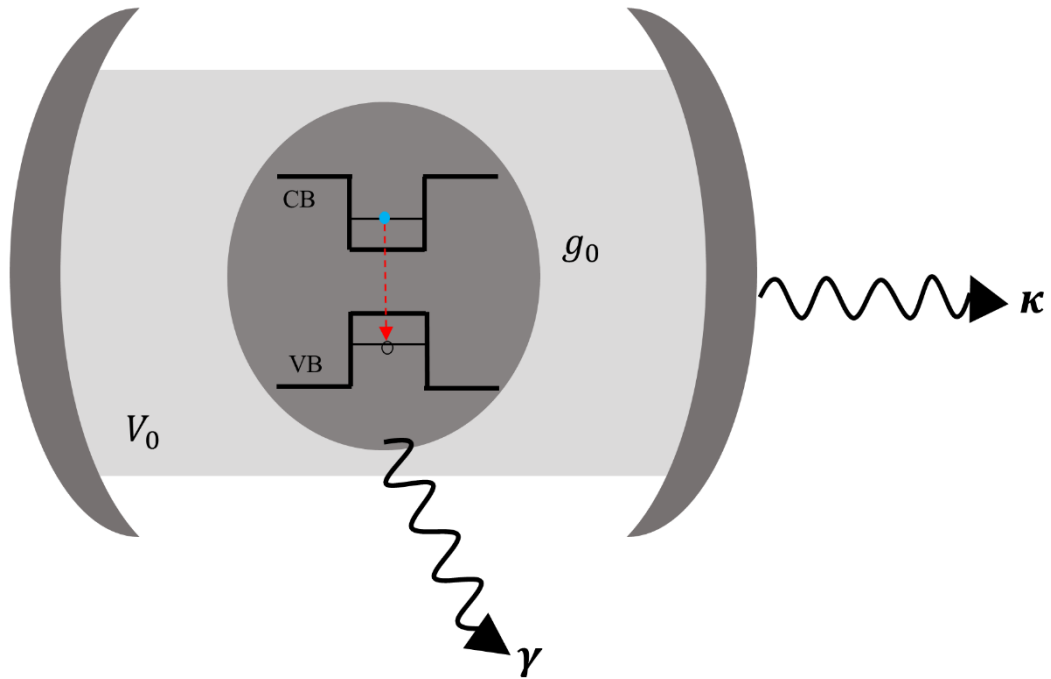


Figure 2.8. A two-energy level system in a resonance cavity with modal volume of V_0 . The cavity is described by three parameters: g_0 , κ , and γ , which represent the atom-cavity coupling, the photon decay rate, and the non-resonant decay rate, respectively.

Cavity quantum electrodynamics explores the interaction between a quantum emitter and the electromagnetic field confined within an optical cavity. In this thesis, particular attention is given to the case where the transition frequency of a QD aligns with one of the cavity's resonant modes. We treat the QD as a two-energy level system for fixed transition frequency analysis which is determined by the bandgap. As shown in Figure 2.8, the relative strength of the QD-cavity interaction is determined by three parameters: the atom-photon coupling parameter g_0 , the photon decay rate of the cavity κ , and the non-resonant decay rate γ . These three parameters each define a characteristic time-scale for the dynamic of the atom-photon system. Basically, the coupling strength can be classified as either weak or strong.

(1) Weak coupling

Weak coupling happens when the QD-cavity coupling constant g_0 is smaller than the loss rate due to either leakage of photons from the cavity κ or decay to non-resonant modes γ , which can be described as,

$$g_0 \leq (\kappa, \gamma) \quad (2.3)$$

where (κ, γ) represents the smaller of κ or γ . Thus, the loss from the system is faster than the characteristic interaction time between QD and cavity. In this case, QD emission from the cavity is irreversible, to free space. Then the cavity mode is resonant with the QD emission, the photon spontaneous emission rate is enhanced in a weak coupling regime, which is called the Purcell effect. The Purcell factor F_P is used to defined how much enhancement or suppression the photon emission rate of the QD in free space ^[167],

$$F_P = \frac{\tau_R^{free}}{\tau_R^{cavity}} \quad (2.4)$$

where τ_R^{free} and τ_R^{cavity} represent the photon decay rate is free space and cavity, respectively. F_P can also be defined as,

$$F_P = \frac{3Q(\lambda/n)^3}{4\pi^2 V_e} \quad (2.5)$$

here Q represents the quality factor of the cavity, λ represents the emission wavelength of the QD, n represents the refractive index of the medium inside the cavity, V_0 represents the effective mode volume of the resonant cavity mode. Purcell factor is a convenient parameter to characterise the effects of QD in an optical cavity, $F_P > 1$ imply the QD spontaneous emission rate is enhanced by the cavity. Conversely, $F_P < 1$ imply the cavity suppresses the QD emission.

Equation 2.5 shows that large Purcell factor requires the cavity to have a high Q factor and small effective modal volume.

Spontaneous emission coupling factor (β) is another useful parameter to describe the cavity effect, which is the fraction of the number of photons emitted into the cavity mode to the total number photons emitted:

$$\beta = \frac{F_p}{1+F_p} \quad (2.6)$$

The higher the β value, the more photons are coupled to the cavity mode. If the β value is near unity, the Purcell factor is large.

(2) Strong coupling

Strong coupling occurs when the QD-cavity coupling constant g_0 is much larger than the cavity decay rate set by the cavity lifetime and the non-resonant atomic decay rate, which is described as,

$$g_0 \gg (\kappa, \gamma) \quad (2.7)$$

Here, the interaction between the photons in the cavity mode and QD is reversible. When the QD emits a photon into the resonant cavity mode, it is reflected between the mirrors and reabsorbed by the QD before being lost from the system. In this regime, the emitter no longer undergoes irreversible spontaneous emission; instead, it engages in a coherent and periodic exchange of energy with the cavity photon, manifested as Rabi oscillations occurring on timescales shorter than the inverse cavity field decay rate. This coherent and reversible light–matter interaction defines the cavity quantum electrodynamics regime.

The coupling strength g_0 is related to the optical transition dipole moment μ , local electric field E , oscillator strength f , and cavity mode volume V can be expressed as ^[168]:

$$g_0 = \mu \cdot E = \hbar \sqrt{\frac{\pi e^2 f}{4\epsilon_r \epsilon_0 m_0 V}} \quad (2.8)$$

where ϵ_r ϵ_0 is the dielectric constant of the cavity material and m_0 is the free electron mass. Equation (2.8) shows that the coupling strength g_0 increases with the oscillator strength f , and decreases with the cavity mode volume V . Therefore, achieving the strong-coupling regime requires a small cavity mode volume V , a large oscillator strength f , a narrow line emitter (small γ) and a high-quality-factor (Q-factor) cavities with low decay rate κ .

In the strong coupling regime, the excited states are doubly degenerate as a result of the electric-dipole interaction between the two-energy level QD and the resonant cavity mode. Each level consists of a doublet with splitting:

$$\Delta E_n = 2\sqrt{n}\hbar g_0 \quad (2.9)$$

This mixed atom-photon states are called dressed states and the ladder of doublets is called the Jaynes-Cumming ladder. An important point for this system is the splitting even for the first rung of ladder, which is called the vacuum Rabi splitting, and its magnitude is equal to:

$$\Delta E^{vac}(N) = \sqrt{N}\Delta E^{vac} = \sqrt{N}\left(\frac{2\mu_{12}^2\hbar\omega}{\epsilon_0 V_0}\right)^{1/2} \quad (2.10)$$

where N is the number of atoms interacting with the single-mode field of a resonant cavity in the Jaynes-Cummings model.

2.2.2 Light extraction efficiency

Light extraction efficiency is a critical parameter in the realisation of high-performance single-photon sources based on QDs. It directly determines the brightness of the emitter—that is, the photon counts that can be delivered to a single photon detector. Even if a QD exhibits excellent intrinsic properties such as high quantum efficiency, spectral purity, and photon indistinguishability, these advantages are significantly undermined if only a small fraction of the emitted photons escape the host material and are collected. A QD embedded in a bulk semiconductor structure typically suffers from poor extraction efficiency—often below 1–5%—due to the high refractive index contrast between the semiconductor host (e.g., $n_{\text{GaAs}} \sim 3.5$) and the surrounding medium (e.g., $n_{\text{air}} \sim 1.0$). This mismatch results in strong total internal reflection at the semiconductor-air interface and severe photon loss into non-collectible modes.

To overcome these challenges, QDs are routinely integrated into optical microcavities, which serve dual roles: they enhance the radiative emission rate through cavity quantum electrodynamic effects (as discussed in Section 2.2.1), and reshape the angular emission profile, enabling a more directional and collectable output. This section focuses on the mechanisms and strategies for improving light extraction efficiency η from QD-cavity systems.

Light extraction efficiency ϵ can be defined as ^[169]:

$$\epsilon = \beta \cdot \eta \quad (2.11)$$

where β is the spontaneous emission coupling factor, η is the collection efficiency of the photons that have been emitted into cavity mode.

$$\eta = \frac{\frac{1}{Q_C}}{\frac{1}{Q_C} + \frac{1}{Q_0}} = 1 - \frac{Q_T}{Q_0} \quad (2.12)$$

where Q_T is the total Q factor of the optical cavity that can be divided into two terms, corresponding to the optical losses either collected (Q_C) or not collected (Q_0). High- β factors (>90%) are achievable in high-Q/low-V mode cavities, when the emitter is spectrally and spatially aligned with the resonant mode. The collection efficiency η , in contrast, depends on the geometry and photonic design of the structure and typically ranges from 20–80%, depending on fabrication precision. Table 2.1 gives an indication of light extraction efficiency from different cavity geometry designs.

Table 2.1. An overview of the light extraction efficiency from different cavity geometry designs.

Cavity Design	Extraction efficiency η	β	Single photon purity $g^{(2)}(0)$
Micropillar ^[170]	85%	0.8	0.016
Bullseye ^[171]	16.6%	0.83	0.0032
Tapered NW ^[172]	72%	0.95	0.008
Optical antenna ^[173]	65%		0.002
Topological bulk cavity ^[174]	92%	0.62	–

2.3 Photon antibunching

One of the defining characteristics of a true single-photon emitter is its ability to emit photons one at a time, a phenomenon known as photon antibunching. This quantum optical behaviour, fundamentally distinct from classical or coherent light sources, serves as a critical signature for verifying the quantum nature of light emitted from quantum dots and other single-photon sources. In this section, we discuss the concept of photon antibunching by starting with the definition of light sources, and how it is experimentally measured, particularly through second-order correlation function $g^{(2)}(0)$ measurements using a Hanbury-Brown-Twiss interferometer.

2.3.1 Definition and types of light source

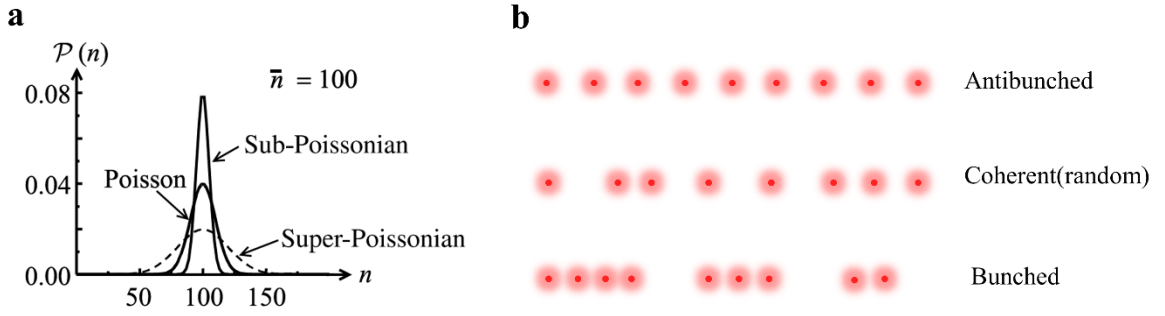


Figure 2.9. Photon statistics comparison. (a) Photon statistics with the same mean photon number $\bar{n}$ for light with Poissonian distribution, sub-Poissonian distribution, and super-Poissonian distribution. (b) Comparison of the photon streams for antibunched, coherent and bunched light.

Photons are with discrete nature, and the photon number fluctuates on short time-scales. These fluctuations can be described by photon statistics as shown in Figure 2.9(a). Three types of light sources are defined by comparing probability distribution $p_{(n)}$ of detecting n photons with same mean photon number $\bar{n} = 100$ of light and the photon distribution properties in Figure 2.9 (b).

Poissonian Distribution (Coherent Light):

$$\Delta n = \sqrt{\bar{n}} \quad (2.13)$$

The solid black curve represents a Poissonian distribution, characteristic of coherent light sources such as lasers. For these sources, the photon number fluctuations follow the relationship above is the average photon number. This distribution serves as a reference for "random" photon emission without correlations.

Sub-Poissonian Distribution (Antibunched Light):

$$\Delta n < \sqrt{\bar{n}} \quad (2.14)$$

The narrow peak labelled as "Sub-Poissonian" corresponds to light sources exhibiting reduced photon number fluctuations compared to the Poissonian case. This behaviour is typical of single-photon emitters, such as isolated quantum dots or trapped atoms, where photon antibunching leads to suppressed simultaneous multi-photon emission. Sub-Poissonian light is a signature of non-classical photon statistics.

Super-Poissonian Distribution (Bunched Light):

$$\Delta n > \sqrt{\bar{n}} \quad (2.15)$$

The broader distribution labelled "Super-Poissonian" reflects enhanced photon number fluctuations, characteristic of thermal or bunched light sources (e.g., blackbody radiation, spontaneous emission). In such cases, photons tend to arrive in groups, resulting in larger-than-Poissonian variances.

2.3.2 Single photon sources

Single-photon sources (SPSs) are non-classical light source with anti-bunching photon distribution as discussed in section 2.3.1 and capable of producing one and only one photon at a time in response to a trigger pulse, either optically or electrically.

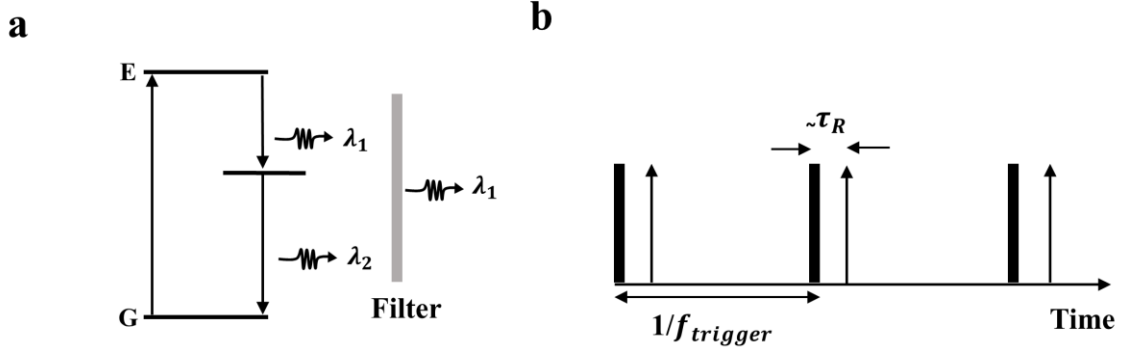


Figure 2.10. Schematic illustration of single atom in response to a trigger pulse. (a) Under optical or electrical excitation, a quantum emitter is promoted from the ground state to an excited state. The subsequent cascade results in emission of photons with different wavelength. After emission, the system returns to the ground state, ensuring that only one photon is emitted per excitation cycle. (b) Photon emission sequence under trigger repetition rate of $1/f_{trigger}$. The thick lines and arrows represent the trigger pulses and photon emission events, respectively. The time different between them is roughly the radiative lifetime (τ_R) of the single photon.

$$y = y_0 + A_1 \cdot \exp(-x/\tau_R) \quad (2.16)$$

In general, a single-photon emitter operates based on a quantum two-level or three-level system, in which a single excitation leads to a single radiative recombination event. Figure 2.10(a) illustrates the basic excitation and emission process of a single atom with three-level system. Upon absorption of energy, the emitter is excited from its ground state to an excited state. After cascading the ground state, two types of different wavelength photons are emitted. By using the optical filter, it is possible to select photons with single wavelength for single photon source applications. The quantum nature of this transition ensures that only one photon is emitted per excitation cycle, provided the emitter is not re-excited during the emission event. Under periodically pulse excitation with trigger pulse frequency of $f_{trigger}$, the atom will emit exactly one photon for each pulse as illustrated in Figure 2.10 (b). The time between pulse excitation

and photon emission is roughly the radiative lifetime of the single photons. If the time separation which designed by $f_{trigger}$ is significantly longer than τ_R , the trigger pulses control the separation of subsequent photons.

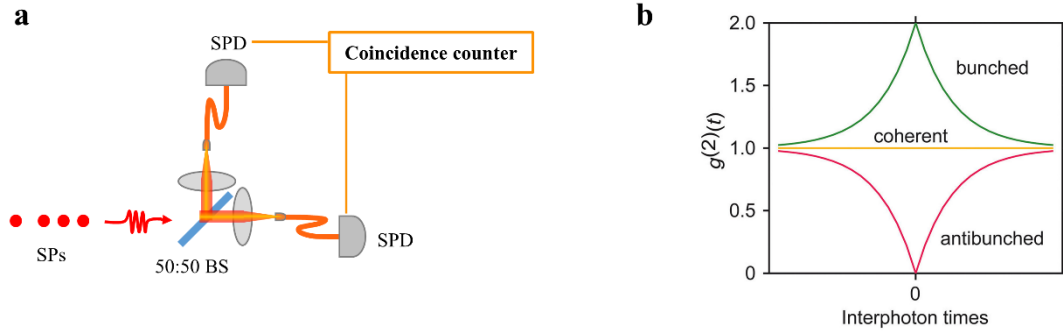


Figure 2.11. Photon antibunching measurement. (a) Schematics of Hanbury-Brown-Twiss experiment set up for single photon purity measurement. (b) Theoretical calculation of $g^{(2)}(0)$ values of different types of light sources.

Figure 2.11(a) shows a schematic of the Hanbury–Brown and Twiss (HBT) experimental setup used to characterize single-photon purity. In this setup, the emitted photons are split by a 50:50 beam splitter and directed to two single-photon detectors. By recording the coincidence counts as a function of the time delay between detection events, the second-order correlation function is obtained as:

$$g^{(2)}(\tau) = \frac{\langle I(t) + I(t+\tau) \rangle}{\langle I(t) \rangle \langle I(t+\tau) \rangle} \quad (2.17)$$

where $I(t)$ is the intensity of light beam at time t , $I(t) = \langle I(t + \tau) \rangle$ if a source with constant average intensity, when $\tau \gg \tau_c$. The intensity fluctuation at time t and $t + \tau$ will be completely uncorrelated with each other:

$$I(t) = \langle I \rangle + \Delta I(t) \quad (2.18)$$

With $\Delta I(t) = 0$,

$$\langle I(t) + I(t + \tau) \rangle_{\tau \gg \tau_c} = \langle I \rangle^2 \quad (2.19)$$

Therefore,

$$g^{(2)}(\tau \gg \tau_c) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle \langle I(t+\tau) \rangle} = \frac{I_0^2}{I_0^2} = 1 \quad (2.20)$$

In contrast, when $\tau \ll \tau_c$, the fluctuation between the two times will have correlation. In particular when $\tau = 0$,

$$g^{(2)}(0) = \frac{\langle I(t)^2 \rangle}{\langle I(t) \rangle^2} \quad (2.21)$$

Thus, in any conceivable time dependence of $I(t)$, it will be always the case that,

$$g^{(2)}(0) \geq 1 \quad (2.22)$$

The above discussion is based on the intensity correlation. In real measurement, number of photons will be measured by single photon detectors, since it is proportional to the light intensity, equation 2.17 can also be as:

$$g^{(2)}(\tau) = \frac{\langle n_1(t)n_2(t+\tau) \rangle}{\langle n_1(t) \rangle \langle n_2(t+\tau) \rangle} \quad (2.23)$$

where $n_i(t)$ is the number of photon counts registered on detector i at time t . In anti-bunching light, there is a regular gap between the photons. Thus, the probability of detecting a photon at detector 2 after detecting one photon on detector 1 is small for small values of τ and then increases with τ . Therefore, as shown in the Figure 2.11(b):

$$g^{(2)}(0) < g^{(2)}(\tau), \quad (2.24)$$

$$g^{(2)}(0) < 1 \quad (2.25)$$

Second-order correlation function can also be explained via various form of light that we consider in classical optics. For instance, the thermal or chaotic light emitted from an atomic discharge lamp can be described using simplified source models. If the spectral line is Doppler broadened, it can be approximated by a Gaussian line shape.

$$g^{(2)}(\tau) = 1 + \exp\left[-\pi\left(\frac{\tau}{\tau_c}\right)^2\right] \quad (2.26)$$

Similarly, if the spectra line is a lifetime-broaden source with a Lorentzian line shape,

$$g^{(2)}(\tau) = 1 + \exp(-2|\tau|/\tau_0) \quad (2.27)$$

Ideal single photon source

There are mainly three key parameters that describe an ideal single photon source for quantum information technology applications: brightness, purity and indistinguishability.

Brightness

Brightness refers to the probability of generating and collecting a single photon per excitation cycle. It quantifies how efficiently a single-photon source emits photons into the desired optical mode. High brightness is essential for practical quantum information technologies, as it directly determines the photon generation rate and the overall system performance. The brightness depends on several factors, including the internal quantum efficiency of the emitter, the coupling efficiency between the emitter and the optical mode, and the collection efficiency of the detection system. Achieving high brightness typically requires optimising the emitter–cavity coupling, minimising optical losses, and improving photon extraction through nanophotonic engineering.

Purity

Purity describes the probability that a single-photon source emits exactly one photon per excitation event, without any multi-photon contribution. It is commonly quantified by the second-order correlation function at zero-time delay $g^{(2)}(0)$ as presented above, where an ideal single-photon source exhibits $g^{(2)}(0) = 0$. High purity is crucial for secure quantum communication and reliable quantum computation, as multi-photon emission events can lead to information leakage or logical errors. Factors affecting purity include background emission, carrier re-excitation, and timing jitter. Careful control of the excitation conditions, material quality, and device design is essential to suppress unwanted multi-photon generation and achieve near-ideal single-photon statistics. In practical applications, a value of $g^{(2)}(0) < 0.5$ confirms single-photon emission, while high-performance quantum technologies typically require $g^{(2)}(0) < 0.1$ or even lower.

Indistinguishability

Indistinguishability characterises how identical consecutive photons are in all degrees of freedom, including frequency, polarisation, temporal profile, and spatial mode. For photons to interfere quantum mechanically — a key requirement for quantum logic operations and

entanglement generation — they must be perfectly indistinguishable. Indistinguishability is often evaluated through two-photon interference experiments, such as the Hong–Ou–Mandel test. In practice, photon indistinguishability can be degraded by dephasing, spectral diffusion, and timing jitter arising from interactions with the solid-state environment. Achieving high indistinguishability therefore requires emitters with narrow linewidths, stable emission energy, and fast radiative recombination, often realised through coupling to optical cavities or operating at cryogenic temperatures. Indistinguishability of the emitted photons is not discussed in detail in this thesis, as it has not been experimentally measured in our work.

2.4 Excitation schematics

The excitation of single QDs is a fundamental step in generating quantum light, and the method of excitation significantly influences the emission properties, including brightness, coherence, and timing precision. QDs can be excited either optically or electrically, each offering distinct advantages and practical considerations depending on the target application. In this section, we discuss the schematics and basic mechanism of these two excitation approaches.

2.4.1 Optical excitation

Optical excitation refers to the process of generating electron–hole pairs in a QD through the absorption of photons from an external light source, typically a laser. When the incident photon energy exceeds the bandgap of the surrounding material or the QD itself, carriers are excited into higher energy states and subsequently recombine radiatively, leading to the emission of single photons. Optical excitation is widely used in laboratory settings due to its experimental flexibility and precise control over excitation parameters such as wavelength, power, and polarisation. The emission is then characterised in their spectrum, lifetime, and photon statistics. Figure 2.12 illustrates three commonly employed optical excitation schematics^[175]: above-band, quasi-resonant, and resonant excitation.

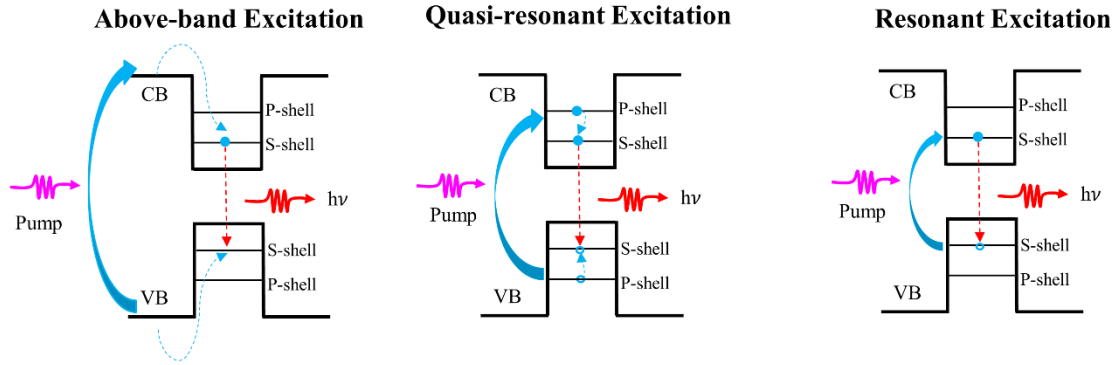


Figure 2.12. Three different optical excitation schemes: Above-band excitation (left), quasi-resonant excitation (middle), and resonant excitation (right).

In above-band excitation ^[176], a pump laser with photon energy greater than the bandgap of the surrounding matrix material creates electron–hole pairs in the barrier. These carriers subsequently relax via phonon interactions and are captured into the quantum dot, typically occupying higher excited states (p-shell), followed by rapid non-radiative relaxation to the lowest energy level (s-shell), before recombining radiatively. Although this method is straightforward and widely used, it introduces significant background emission and phonon scattering, which can degrade the indistinguishability and temporal precision of the emitted photons.

In quasi-resonant excitation, the pump laser is tuned to an excited state within the QD, such as the p-shell ^[177], allowing direct excitation into the quantum dot without generating free carriers in the surrounding material. The excited carriers then relax quickly and non-radiatively to the s-shell, and finally recombine radiatively to emit a photon. This approach offers a compromise between simplicity and coherence, improving emission purity while reducing background noise and temporal jitter compared to above-band excitation.

Similarly, resonant excitation involves tuning the pump laser precisely to the s-shell transition energy of the QD ^[178, 179], enabling coherent excitation and de-excitation within the same energy level. This scheme allows for deterministic single-photon emission with high purity and indistinguishability, as it minimises phonon interactions and avoids carrier relaxation pathways. However, due to the spectral overlap between the excitation laser and emitted photon,

sophisticated cross-polarisation or orthogonal excitation-detection schemes are often required to isolate the emission signal.

2.4.2 Electrical injection

Electrical excitation represents a critical step toward scalable, chip-integrated quantum light sources. In this scheme, an InAs QD is embedded in an InP p-i-n diode structure as illustrated in the band energy diagram in Figure 2.13, and an external voltage bias is applied to drive electrons from conduction band of the n-type material and holes from valence band of p-type material into the QD. Recombination of these carriers within the confined energy states of the QD results in the emission of a single photon, making this structure suitable for electrically driven single-photon sources.

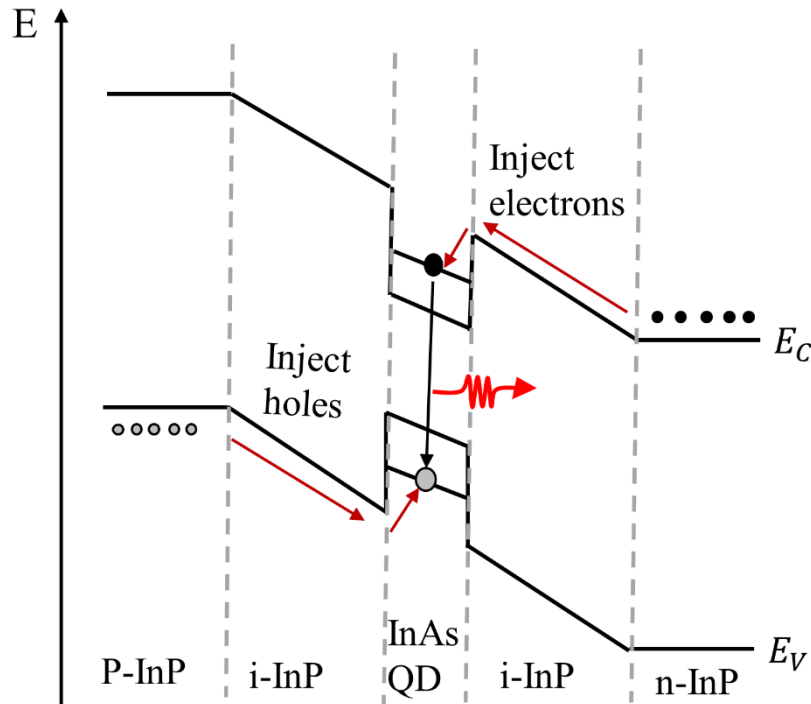


Figure 2.13. Band structure of InAs/InP single quantum dot with p-i-n diode. Electrons are injected from conduction of the n-InP side and holes from valence band of the p-InP side into the intrinsic region, where the InAs quantum dot is located. Carrier recombination within the single quantum dot leads to single-photon emission.

Compared to optical methods, electrical injection offers two major advantages for real-world devices:

1. Compactness and integrability, as it eliminates the need for external lasers.
2. Scalability, through wafer-level fabrication and integration with silicon photonics or CMOS-compatible circuits.

Nevertheless, electrically driven sources face additional challenges, such as background noise from nearby charge traps, current leakage, and thermal broadening, all of which can degrade photon coherence. To address these issues, advanced heterostructures with tunnel barriers, charge stabilisation layers, and site-controlled QDs are being developed.

2.5 Summary

In this chapter, we systematically discuss the theoretical background of the bottom-up growth of QDs embedded in optical cavities for single-photon source applications, covering both material growth and optical aspects. The chapter begins with the fundamental mechanisms of epitaxial growth, including selective area epitaxy in MOCVD systems and self-assembled QD formation. We then examine light–matter interactions by placing a QD within an optical cavity, introducing the concepts of cavity quantum electrodynamics and light extraction efficiency. To elucidate the operation principles of a single-photon source, we explain photon antibunching behaviour by comparing photon bunching statistics and the second-order correlation function. Finally, we present the excitation schemes for both optical pumping and electrical injection to provide a comprehensive understanding of optical data collection.

CHAPTER 3

3. Experimental Techniques

3.1. Introduction

This chapter discusses procedures of sample preparation for selective area epitaxy (SAE) and NW device fabrication for electrically driven single photon source, as well as their corresponding basic working principles of the major experimental techniques used in this thesis.

3.2. Substrate preparation for selective area epitaxy

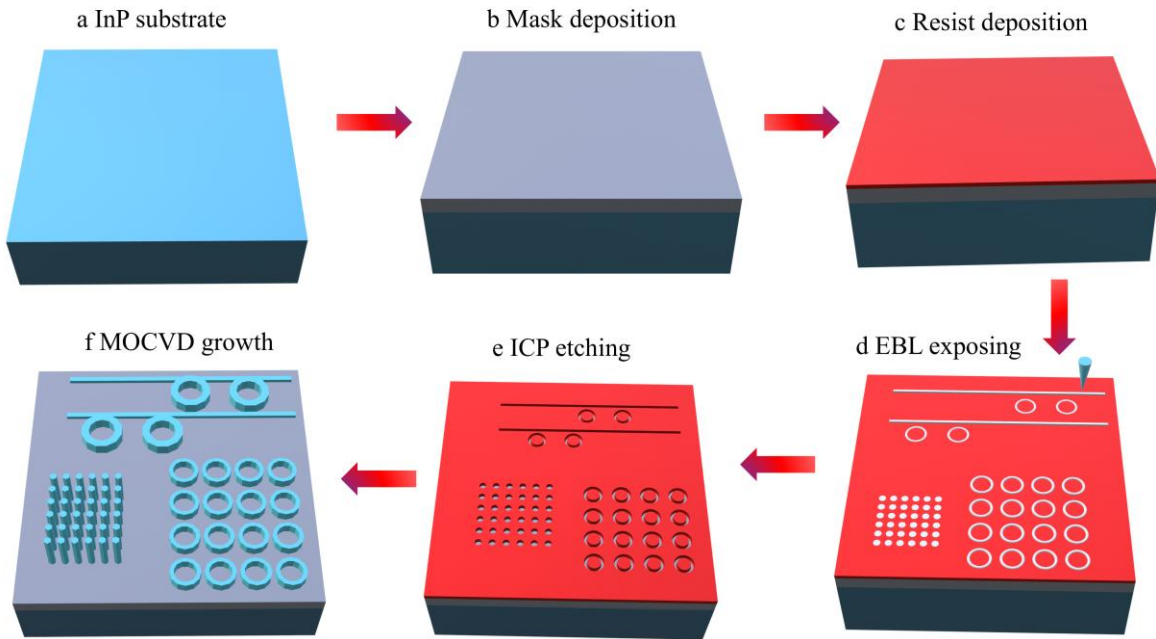


Figure 3.1 Schematic of patterning nanostructures on InP substrate for selective area epitaxy. (a) InP substrate. (b) SiO_x deposition on InP substrate. (c) Spin coating of electron resist. (d) Electron beam lithography for different geometries: rings, holes, and waveguides. (e) inductively coupled plasma reactive ion etching of the exposure area to get the pattern for growth. (f) InP nanostructures grown from the corresponding pattern openings on the patterned InP substrates.

Figure 3.1 schematically illustrates the key processing steps for selective area epitaxy of nanostructures. To achieve selective-area growth of nanostructures with desired geometries, a substrate pre-patterned with a dielectric mask such as silicon dioxide or silicon nitride is required^[180]. First is to select the right orientation substrate. For III-V ($A^{III}B^V$) semiconductors, $\{111\}$ planes are polar and can be divided into (111)A and (111)B^[181, 182]. Selective area epitaxy is achieved by the nucleation and growth under standard vapour-solid growth mode, and previous study shows that hexagonal nucleus along the $\langle 111 \rangle$ direction has the smallest Gibbs free energy^[183], however, the selection between the polar $\langle 111 \rangle$ A and $\langle 111 \rangle$ B directions is primarily governed by surface termination and growth conditions, such as the V/III ratio and surface reconstruction, rather than thermodynamics alone. In this thesis, (111)A InP substrates are used for samples in Chapters 4-6. As seen in Figure 3.1(b), a layer of SiO₂ mask is deposited on the substrate by plasma-enhanced chemical vapour deposition. In Figure 3.1(c), a layer of electron-beam resist (ARP-6200, ZEP 520 A, mr-EBR-0.3) is spin-coated on the substrate and baked by a hotplate. (Baking time depends on the resist, ARP-6200 and mr-EBR-0.3 are at 150 °C for 1 min, ZEP 520 A is at 170 °C for 2 minutes). The resist is then patterned by an electron beam lithography system as illustrated in Figure 3.1(d), after which it is developed by the corresponding developer for one minute and rinsed by isopropyl alcohol for one minute. Subsequently, the patterned substrate is transferred to the inductively-coupled plasma reactive ion etching chamber for dry etching to remove the exposed SiO₂ regions (Figure 3.1(e)) followed by soaking the sample in a resist remover overnight to remove the resist. The pattern is then striped by O₂ plasma in barrel etcher for 2 min to ensure complete removal of the resist. Finally, the prepared patterned substrate is trim-etched by in H₃PO₄, H₂O₂, H₂O with a ratio of 1:1:3 for 5 minutes before loading into the MOCVD reactor for growth.

3.2.1. Plasma-enhanced chemical vapour deposition

Plasma-enhanced chemical vapour deposition is one of the most matured chemical vapour deposition techniques for various thin film (SiO_x, SiN_x, etc) with high uniformity and deposition rate at relatively low temperature^[184]. For all the pattern fabrication in this thesis, SiO₂ was used as the mask and deposited by an Oxford Plasmalab 100 Dual Frequency system, the schematic of which is shown in Figure 3.2. The reactant gases are introduced between two electrodes in a parallel plate configuration - a grounded electrode on bottom and an RF biased electrode at the top. Its dual frequency 13.56 MHz and 100 kHz power applied to upper electrode enables stress control and film densification. The substrate is arranged on a tray and the heating system under the tray can be used to heat the substrate.

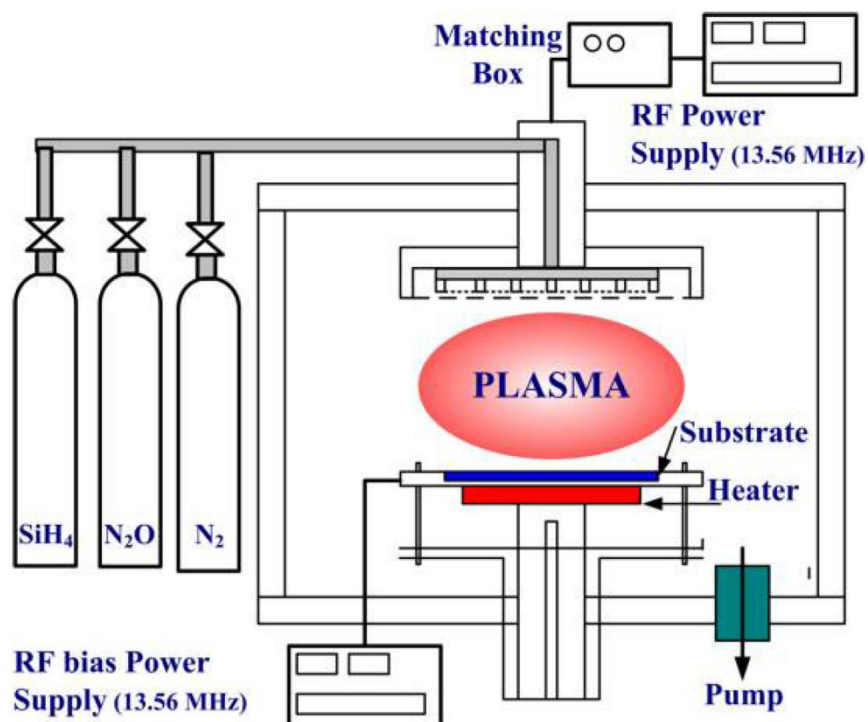
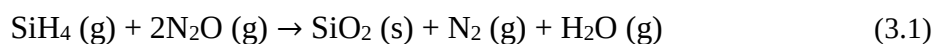


Figure 3.2: Schematic of the plasma-enhanced chemical vapour deposition system ^[185].

SiH₄ and N₂O were used as precursor gases, and N₂ was as carrier gas with the flow rate of 4.5, 355, 81 sccm, respectively, at a pressure of 650 m Torr and temperature of 300 °C . The SiO₂ thickness in Chapter 4, 5, 6 are 30, 100, 440 nm, respectively. The SiO₂ layer is deposited by a series chemical reactions of the reactant gases, created by the plasma generating from capacitive coupling discharge between two electrodes under the radio frequency power of 20 watts. The deposition rate of SiO₂ film under this condition is 0.49 nm/s.

The basic reaction is as indicated below:



3.2.2. Electron beam lithography

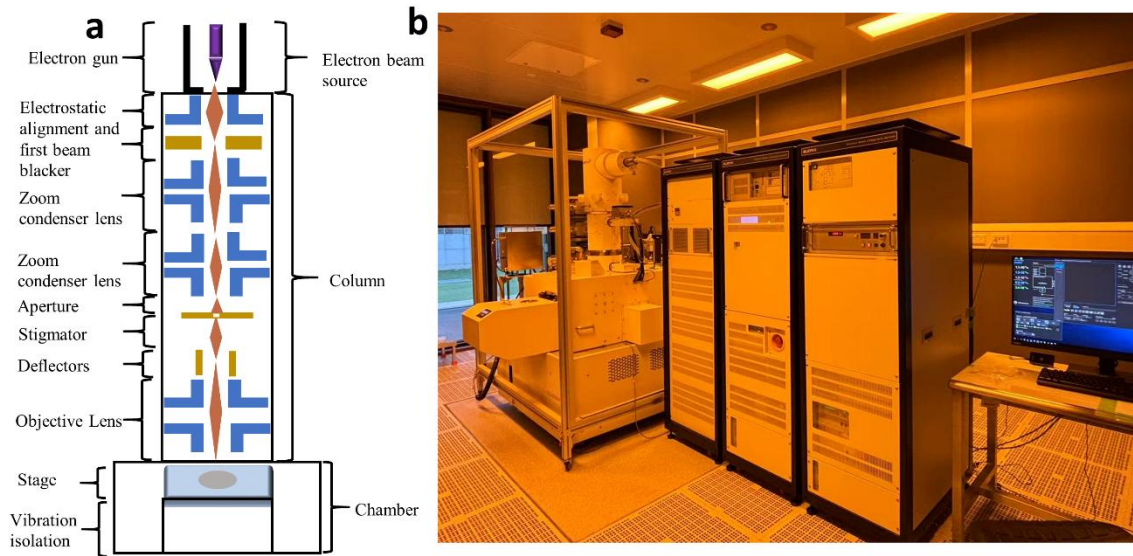


Figure 3.3. (a) Schematic diagram of the electron beam lithography system. (b) A photograph of the Elionix FS-125 system used in the thesis.

Electron beam lithography is a high-resolution patterning technique widely used in nanofabrication for creating nanostructures that are too small to fabricate with conventional photolithography^[186-188]. The schematic of EBL is shown in Figure 3.3 (a). Basically, in a high-vacuum column, electrons are initially generated by an electron gun—either thermionic or field emission—and accelerated by an electrostatic field to 125 keV. These electrons are then shaped and directed using electron-optical components, such as electromagnetic lenses and deflection coils, before being focused onto a substrate coated with electron beam resist. The deflection coils allow the electron beam to be steered to write the desired pattern without physically moving the substrate. The current of the system can be selected from 50 pA to 100 nA, depending on the feature size of the pattern. In general, the system features a field emission electron gun that provides a highly coherent and focused beam, allowing for feature sizes below 10 nm depending on beam current, step size, and exposure time.

During exposure, the incident electrons induce chain scission in resists altering their solubility. Following exposure, the resist is developed in a selective solvent leaving behind the unexposed (if positive resist like PMMA, ARP, ZEP, mr-EBR is used)^[189] or the exposed (for negative resist like HSQ, SU-8)^[190] areas. In this thesis, Raith 150 was used in Chapter 4 and 6, Elionix FS-125 (Figure 3.3 (b)) was used in Chapter 5 in fabricating patterns for selective area epitaxy.

Three types of positive resists - ZEP, mr-pos EBR, and ARP- were used depending of the thickness of the SiO₂ and the pattern size. ZEP was used for the NW patterns while mr-pos EBR was used for the ring patterns as they required different thickness of SiO₂. A field size of 500 μm , current of 2 nA, dose values between 400 to 700 uc/cm^2 were used in this thesis.

3.2.3. Inductively-coupled plasma reactive ion etching

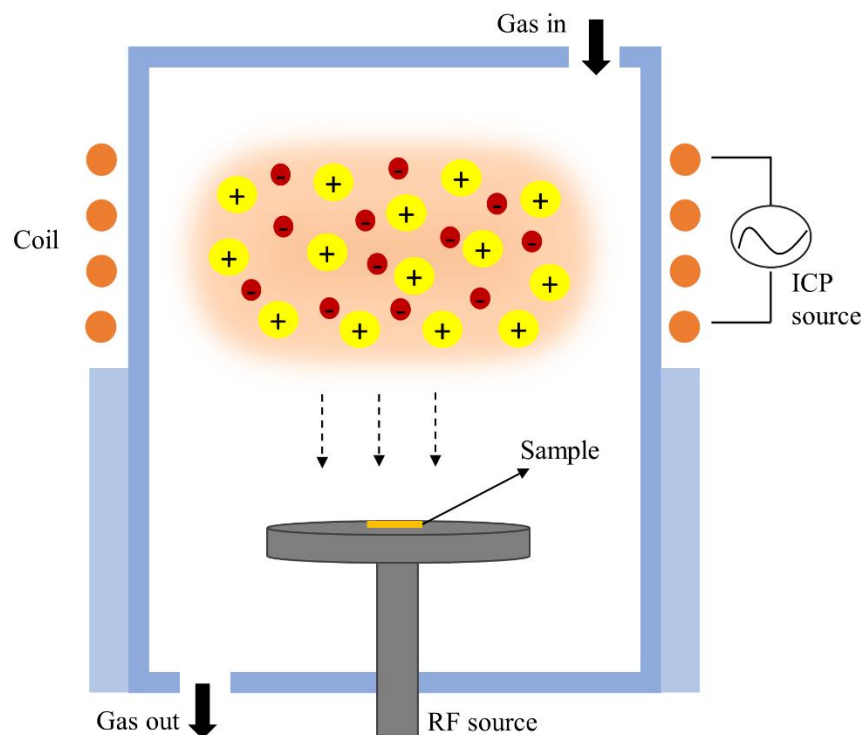


Figure 3.4. Schematics of an inductively-coupled plasma reactive ion etching system.

Inductively-coupled plasma reactive ion etching is a widely used dry etching technique in the fabrication of micro- and nanoscale devices in oxides^[191, 192], nitrides^[193], metals and semiconductors^[194-196] based on silicon, III-V and III-N materials. Different gas chemistries are employed depending on the target material. For instance, fluorine-based gases such as SF₆ and CHF₃ are commonly used for etching silicon^[197] and silicon dioxide^[192], respectively, while chlorine-based chemistries (e.g., Cl₂, BCl₃) are suitable for III-V semiconductors such as GaAs and InP^[196]. Additive gases like O₂ or Ar are often used to modify the etch characteristics or improve sidewall passivation^[198]. In general, it combines the benefits of high-density plasma

generation with precise control over ion bombardment energy, offering superior anisotropy, high etch rates, and excellent process flexibility. The system typically consists of two independently controlled radio frequency power sources. The first source, applied to a planar coil located above the plasma chamber, inductively couples energy into the gas to generate a high-density plasma. This component primarily governs the concentration of reactive species, such as radicals and ions. The second power source is capacitively coupled to the substrate stage, creating a bias voltage that accelerates ions toward the substrate surface. This configuration allows independent tuning of plasma density and ion energy, enabling precise control over the etching profile.

In this thesis, an Oxford PlasmaLab 80 Plus RIE system was employed to etch the patterned SiO₂ mask layer. CHF₃ was used as reactive gas with a flow rate of 25 sccm under 30 mTorr pressure, with ICP and RF power of 100 W and 70 W, respectively - resulting in an etch rate of ~ 0.17 nm/s.

3.2.4. Barrel etching

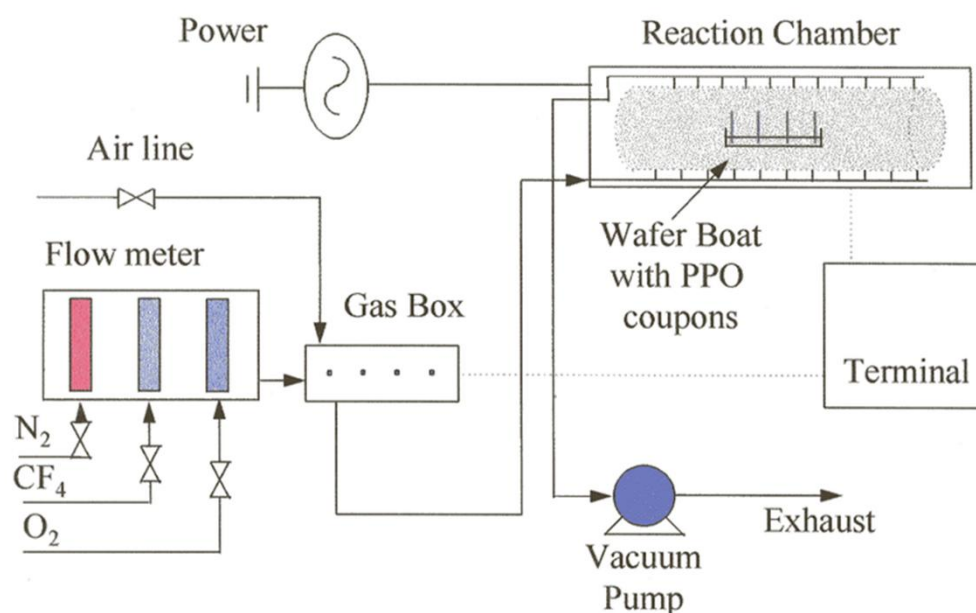


Figure 3.5. Schematics of the barrel etching system ^[199].

The barrel etcher, also known as a plasma asher, is a type of low-pressure plasma processing system primarily used for isotropic dry etching and photoresist removal (ashing) in semiconductor ^[200] and microfabrication processes ^[201]. It operates on the principle of capacitively coupled plasma generation in a cylindrical chamber, where samples are exposed to chemically reactive species in a largely non-directional manner.

In this thesis, a barrel etcher is used for cleaning the pattern opening under O₂ plasma to avoid any resist residual accumulating within the opening after development. This step ensures a clean opening for subsequent inductively-coupled plasma reactive ion etching to remove the exposed SiO₂ regions. An O₂ flow rate of 300 sccm and an RF power of 500 W were applied for 2 minutes.

3.2.5. Metalorganic chemical vapour deposition

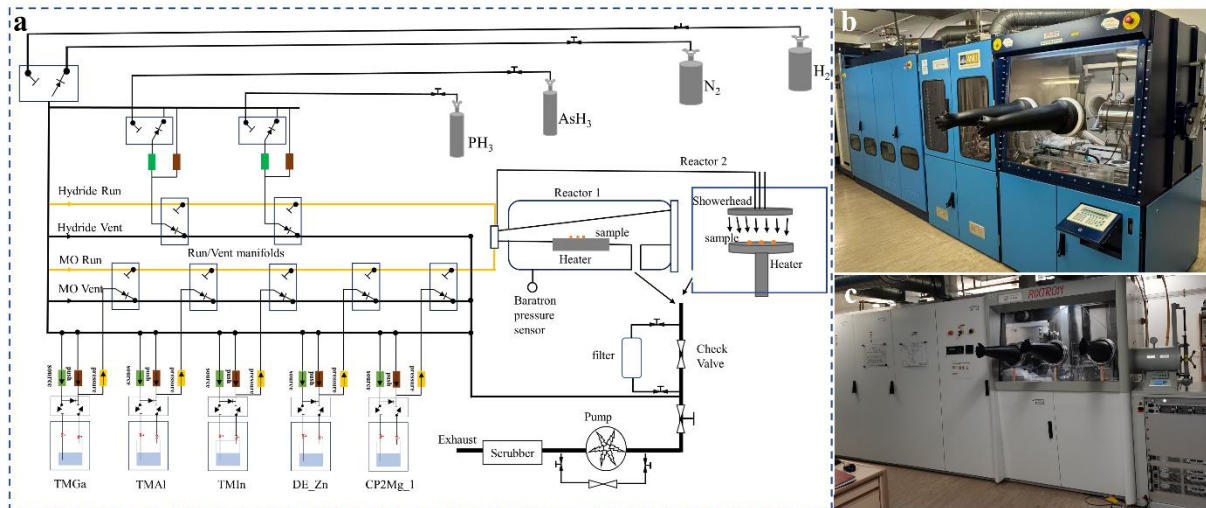


Figure 3.6. Schematic of the two types of MOCVD in a, b is the photo of Aixtron 200/4 horizontal MOCVD, c is the photo of the CCS MOCVD.

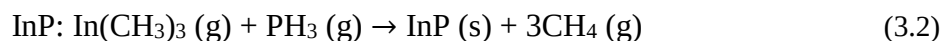
MOCVD, also name as metalorganic vapour phase epitaxy or MOVPE, is a widely used epitaxial growth technique for fabricating high-quality compound semiconductor thin films or nanostructures, particularly those based on III-V such as GaAs, InP, GaN and II-VI materials such as ZnS, ZnO, ZnSe and their ternary or quaternary alloys ^[202]. It is one of the most competitive techniques for the commercial mass production of optoelectronic components due to its high growth rate and its precise control over composition, doping, and layer thickness without the requirement of high vacuum such as in molecular beam epitaxy ^[203, 204]. As a result,

MOCVD has become the dominant method for the production of optoelectronic devices such as LEDs ^[205], lasers ^[202, 206], photodetectors ^[207], and solar cells ^[208].

MOCVD is an extremely complicated system as seen in the schematic in Figure 3.5a. Despite the difference in reactor configurations, the horizontal and closed-coupled shower head systems, they are similar to each other in many aspects. The fundamental principle of MOCVD involves delivering volatile chemical precursors into a heated reactor chamber where they undergo surface-mediated chemical reactions to deposit solid material on the substrate. For growth of III-V compound semiconductors, the typical precursors are metal-organic compounds (e.g. trimethylgallium, trimethylindium, trimethylaluminum that are stored in sealed stainless steel cylinders and maintained at a fixed temperature and pressure as the group III source (e.g. Ga, In, Al). Hydrides (e.g. AsH₃, PH₃, NH₃) are typically used as the group V source (e.g. As, P, N). They are typically in gaseous form and stored in pressurised cylinders. In addition, diethylzinc (Zn(CH₃)₂) and SiH₄ are used as the doping source for p- and n-type doping, respectively. These precursors are transported into the reactor by an inert carrier gas, H₂ or N₂. Figure 3.5a illustrates the gas lines for delivering metalorganics and hydrides consist of run and vent lines, the run lines are connected to the reactor while the vent lines are connected to the exhaust. This design helps to stabilise gas flow when they are switched between in and out of the reactor to minimise fluctuation and turbulence.

Inside the reactor, the substrate is heated to temperatures typically between 500 °C and 1000 °C, which causes the precursors to decompose at or near the substrate surface. The group III atoms from the metal-organics and the group V atoms from the hydride gases react to form a solid crystalline compound, while the organic ligands and by-products are evacuated through the exhaust system. The growth rate is kinetically controlled by adjusting temperature, pressure, gas flow rates, and III/V ratio, allowing precise tuning of film properties and layer structures. We note that safe MOCVD operation is ensured through dedicated ventilated gas cabinets, continuous toxic-gas monitoring, automated gas shut-off and safety interlocks, inert-gas purging, and an exhaust scrubber system for hazardous precursors.

In this thesis, the studies in chapters 4-6 used an Aixtron 200/4 horizontal system. The reactor is operated at a low pressure (100 mbar), which reduces parasitic nucleation and improve the atomic interface sharpness. As this thesis mainly focus on the InP growth, and QD growth by droplet epitaxy, the chemical reactions can be expressed as below,





These represent the simplified overall reactions, as the actual decomposition involves multiple intermediate steps.

3.3. Device fabrication processes

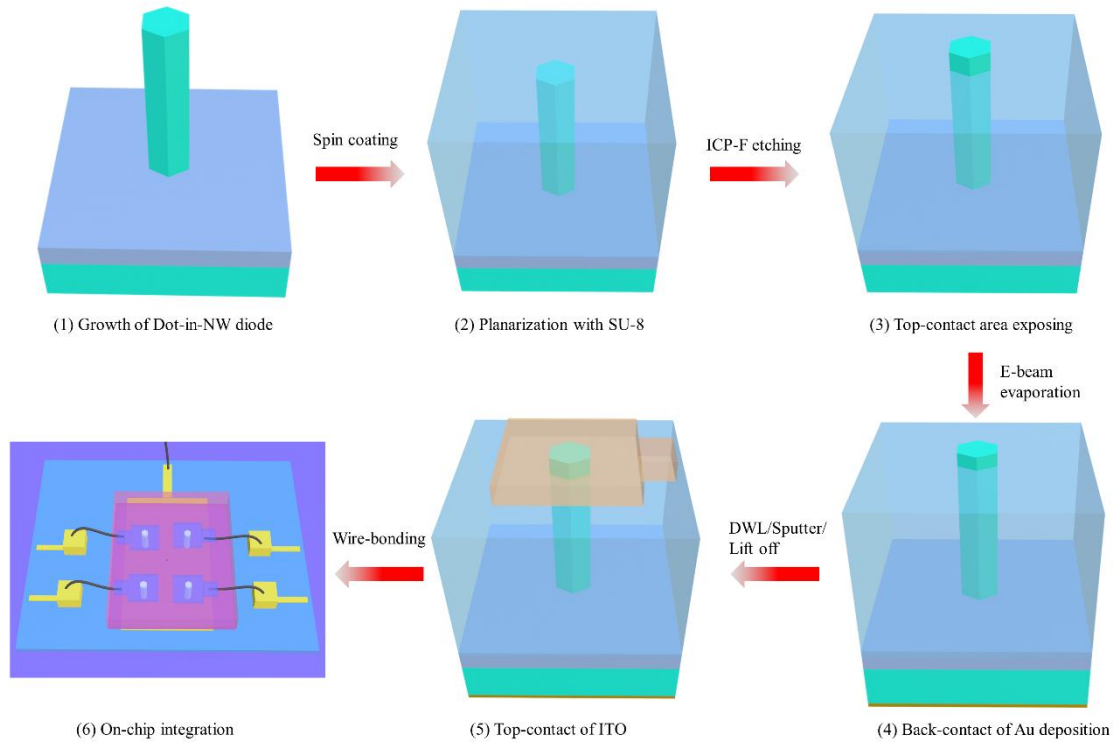


Figure 3.7. Device fabrication process flow: (1) Growth of a single QD within an InP p-i-n NW. (2) Spin-coating of SU-8 for planarisation of the NW. (3) ICP-RIE etching to expose the NW tip for contact formation. (4) Electron beam evaporation of Au on the backside of the substrate. (5) Lithography to define the top-contact area, followed by ITO deposition via sputtering and lift-off. (6) Mounting the device on a PCB package, including wire bonding.

The device fabrication process begins with the epitaxial growth of the QDs in p-i-n InP NWs by MOCVD. The NWs are then planarised by spin-coating a layer of SU-8 photoresist. Subsequently, the substrate is transferred into an inductively coupled plasma reactive ion etching system, where part of the SU8 is etched using 10 sccm flow of CHF_3 to expose the tip of the NWs. To remove the native oxide on the backside of the substrate, the sample is dipped in 1% HF solution prior to depositing a 50 nm Au back contact by electron beam evaporation. For the top contact, direct laser writing lithography is employed to define the contact area. A

layer of AZ-5214 photoresist is spin-coated on top of the sample and exposed with a 405 nm laser. After pattern definition, a 100 nm-thick indium tin oxide layer is deposited by sputtering, followed by soaking in acetone overnight for the lift-off process. Finally, the device is mounted onto a PCB pad using silver paste, and wire-bonded for electrical connection.

3.3.1. Electron beam evaporation

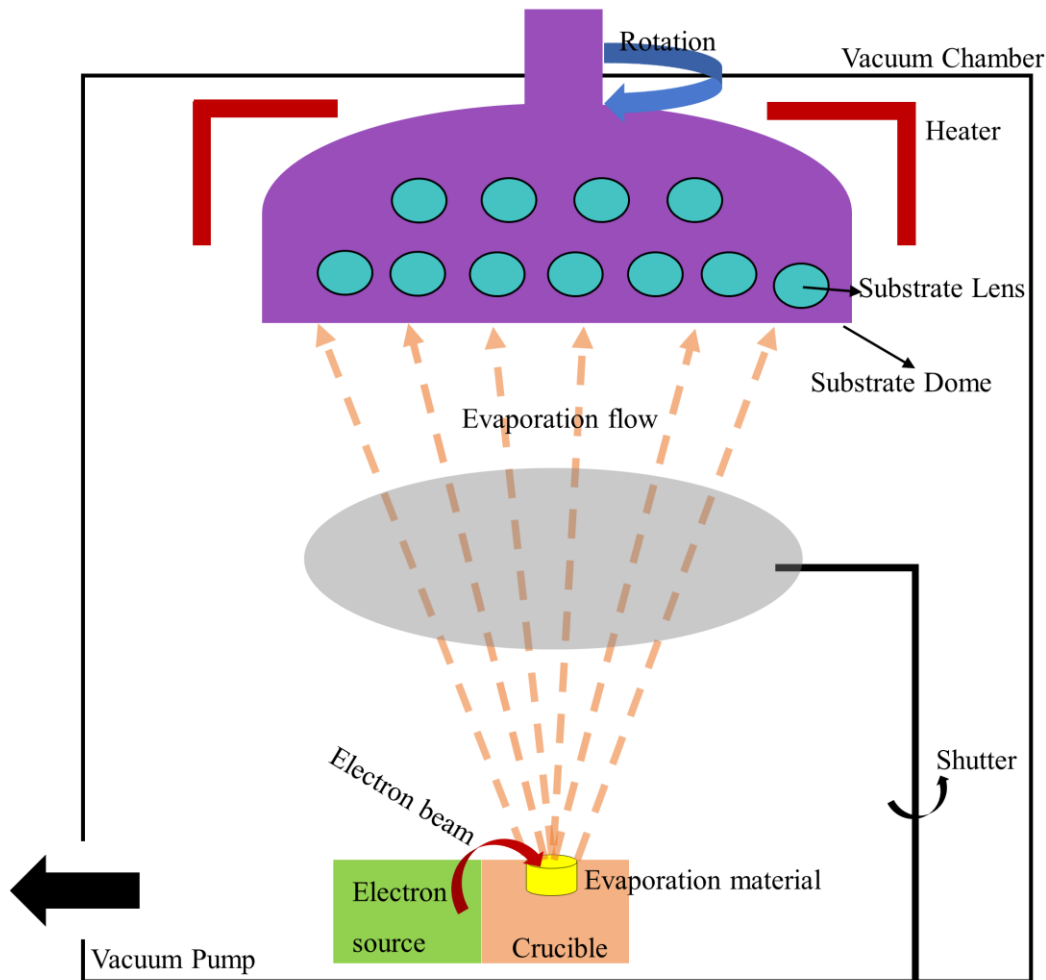


Figure 3.8. Schematic diagram of electron beam evaporation process ^[209].

Electron-beam evaporation is a physical vapour deposition technique in which a high-energy electron beam is used to locally heat and evaporate a source material under high vacuum, allowing the evaporated atoms to condense on a substrate and form a thin film. The deposition is performed in a high-vacuum chamber (typically $<10^{-6}$ Torr) to minimise contamination and ensure good adhesion and film quality. The target material is placed in a crucible, where it is

bombarded by a focused electron beam, resulting in highly localised heating and evaporation. The evaporated vapour then travels towards the substrate, where the samples are mounted. A shutter is used to control the start and stop of deposition, and a rotating sample holder ensures uniform thickness across the wafer.

A Temescal BJD 2000 e-beam evaporator system was used to deposit Ti and Au as the contacts for all the devices in this thesis.

3.3.2. Direct write lithography

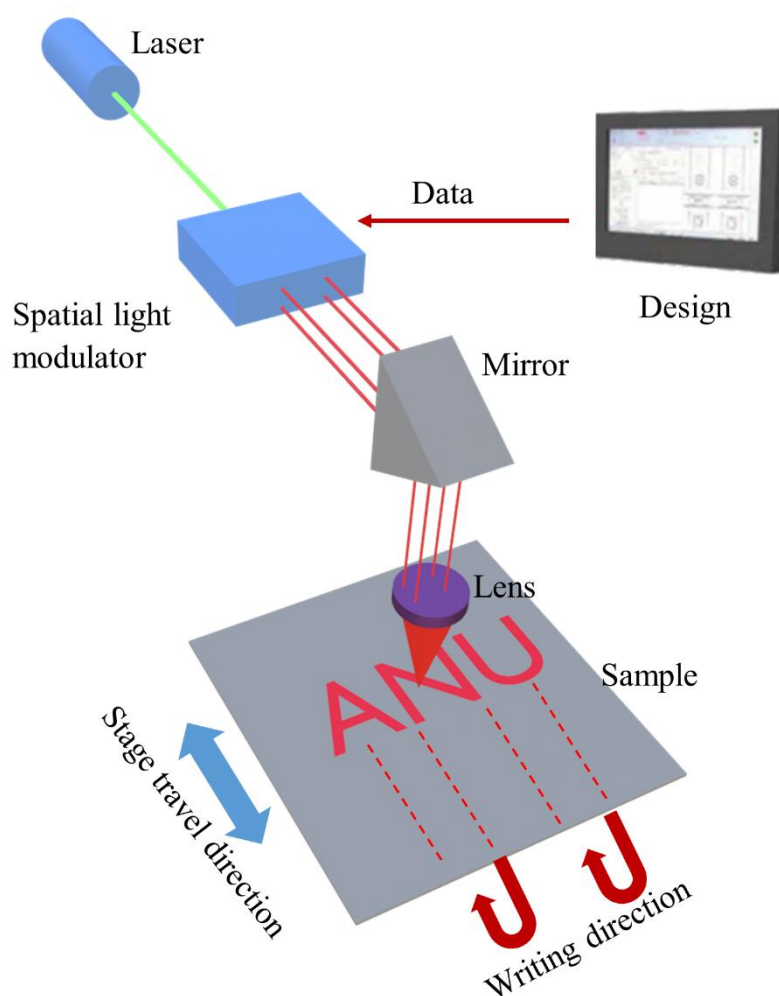


Figure 3.9. Schematic of direct write lithography

Figure 3.9 illustrates the working principle of the Heidelberg Instruments MLA150 maskless lithography system, which enables high-resolution, flexible micro-patterning without the use of a physical photomask. The process begins with a coherent 405 nm UV laser beam, which is

directed into a spatial light modulator, typically a digital micromirror device, to dynamically modulate the beam according to the re-designed patterns. The modulated light is then reflected by a mirror and focused by a lens onto the photoresist-coated sample surface. A precision stage with computer controlled movement to enable stitch-free, large-area exposures. The system also includes automatic focusing, levelling, and alignment functions, which enhance pattern fidelity and reproducibility. In this thesis, we used AZ 5214 photoresist with a dose of 250 mJ/cm^2 for the pattern exposure.

3.3.3. Sputter deposition

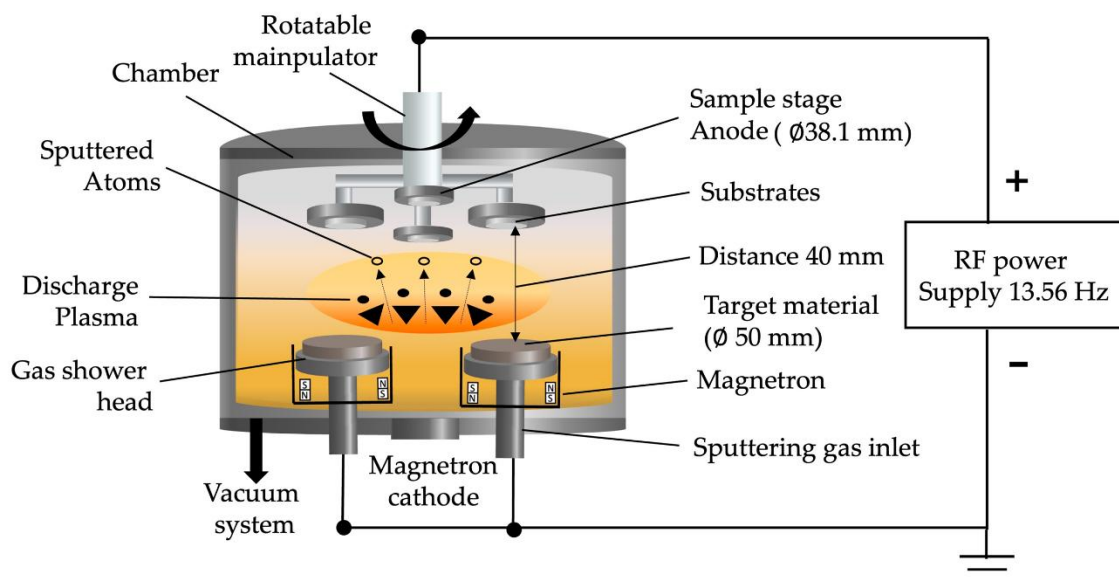


Figure 3.10. Schematic diagram of a direct current magnetron sputter deposition system ^[210].

Sputtering is a widely used physical vapour deposition technique for depositing thin films of metals, semiconductors, and insulators with excellent adhesion and step coverage. In this work, a radio frequency and direct current magnetron sputtering system was utilised for the deposition of contact layers. The sputter deposition process is carried out in a high-vacuum chamber typically below 5×10^{-6} Torr. After reaching vacuum, a sputtering gas—commonly Ar—is

introduced to a controlled working pressure (usually ~ 10 mTorr). A high voltage is then applied between the target (cathode) and the substrate (anode), generating a plasma. For insulating materials such as oxides or nitrides, radio frequency power is used to prevent charge accumulation on the target, while direct current power is used for conductive targets like metals. An AJA sputter system was used for ITO deposition in this thesis.

3.3.4. Wire bonding

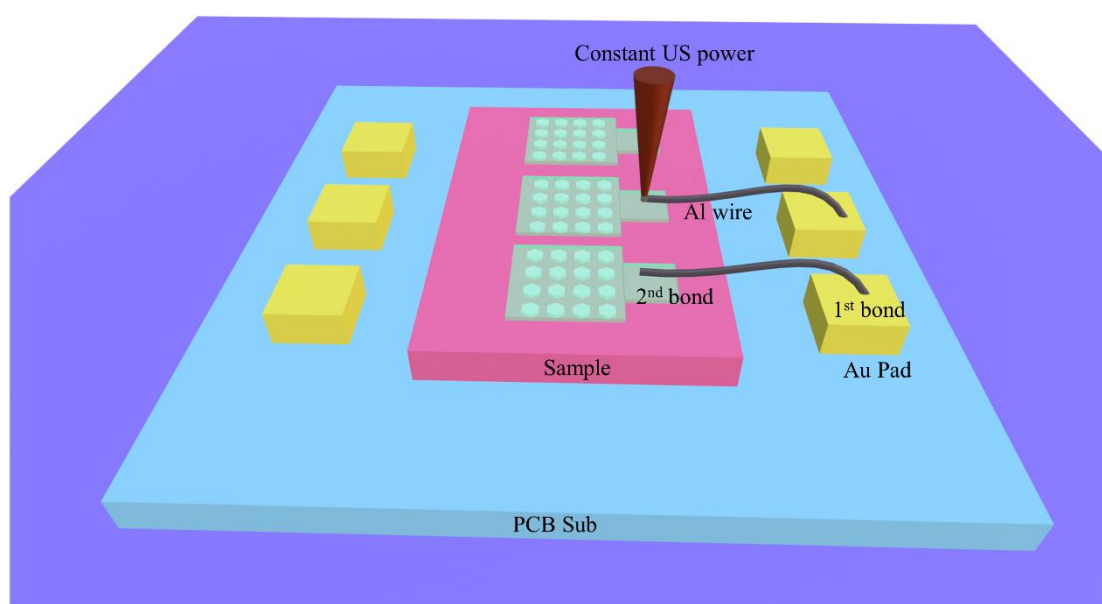


Figure 3.11. Schematic of wire-bonding of the NW devices onto a printed circuit board.

Figure 3.11 illustrates the wire bonding process used to make electrical connections between microfabricated device pads and external circuitry on a printed circuit board. Briefly, a needle with Al wire applying constant ultrasonic power to form the first bond between the Al wire and the Au pad on the printed circuit board. Ultrasonic vibrations, combined with moderate pressure, clean the interface and ensure strong metallurgical adhesion. The needle then rises to loop height position, moves to the top of NW contact region and creates the second bond, completing the electrical path.

3.4. Material property characterisation techniques

3.4.1. Scanning electron microscopy

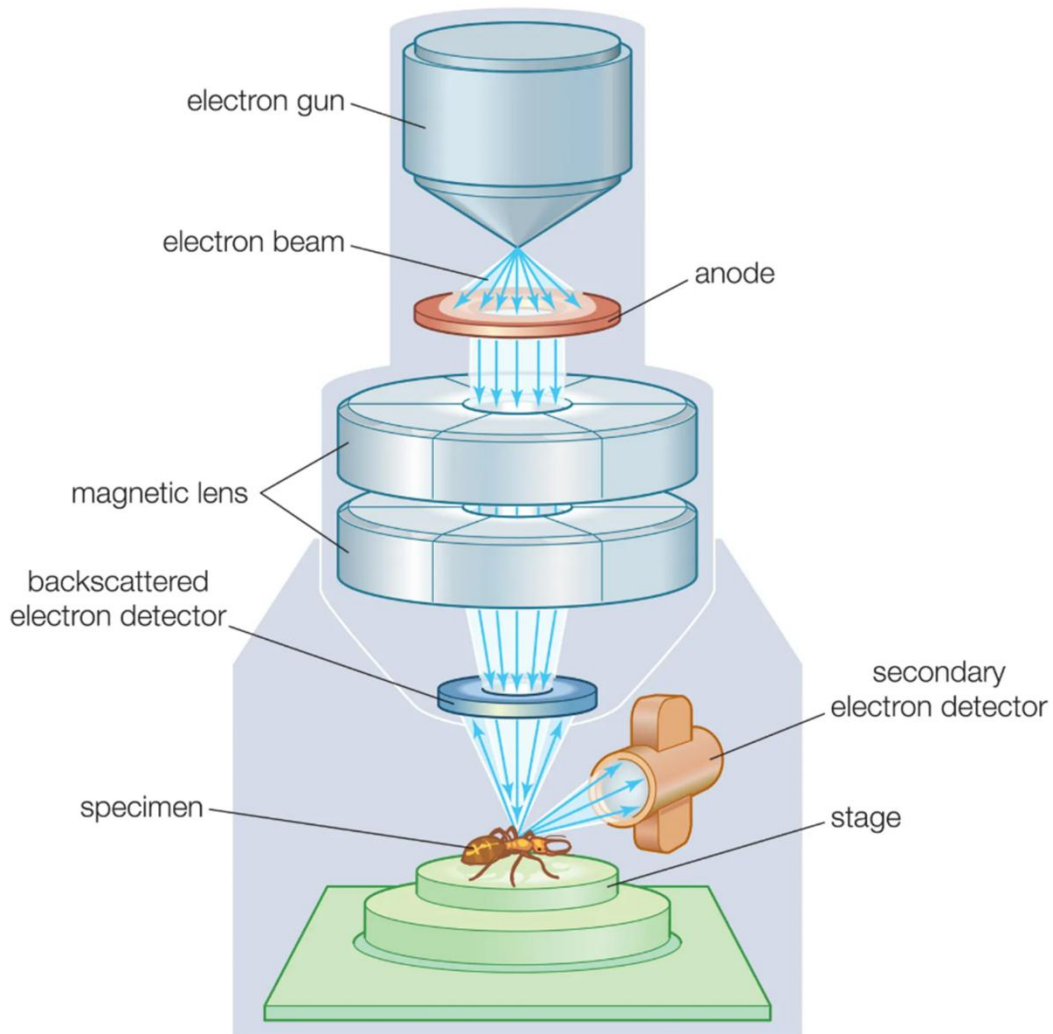


Figure 3.12. Schematic of a scanning electron microscope system.

Scanning electron microscopy is a powerful imaging tool widely used in nanoscience and materials research to obtain high-resolution surface morphology and topographical information^[211]. Figure 3.12 illustrates the basic working schematic of the system - a high-energy focus beam generated by the electron gun scanning across the surface of the sample. When the electron beam interacts with the sample, various signals are generated, including secondary

electrons, backscattered electrons, and characteristic X-rays. The secondary electrons are collected by the detector to provide detailed information about surface features with a resolution down to a few nanometers. Besides imaging, various detectors integrated into the microscope can also deliver crucial insights into the specimen's surface topography and composition. For example, an energy-dispersive X-ray detector enables elemental analysis by detecting characteristic X-rays emitted from the sample, allowing for qualitative and semi-quantitative compositional mapping. In this thesis, a FEI Verios 460 system was used to characterise the nanostructure morphology.

3.4.2. Transmission electron microscopy

Transmission electron microscopy is a powerful imaging and analytical tool that can be used to direct observation of the internal structure of materials at nanometer and even atomic scales. Unlike scanning electron microscopy, which detects surface-emitted electrons, TEM operates by transmitting a highly focused, high-energy electron beam (typically 100–300 keV) through an ultra-thin specimen, usually less than 100 nm thick. As the electrons pass through the sample, they are scattered by the atomic potentials within the material. The transmitted electrons are then collected and magnified by a series of electromagnetic lenses to form high-resolution images on a fluorescent screen or camera.

TEM enables various imaging modes, including bright-field, dark-field, and high-resolution, which provide detailed structural information. Additionally, diffraction patterns obtained from selected area electron diffraction allow for phase identification and analysis of crystal orientation. Moreover, transmission electron microscopes are often equipped with energy dispersive X-ray spectrometer and electron energy loss spectrometer systems for local compositional analysis.

A JEOL 2100F FEGTEM equipped with an energy dispersive X-ray spectroscopy system at the Centre for Advanced Microscopy of Australian National University is used for the structural characterisation of NWs and QDs at a voltage of 200 kV, and a current of 131 μA in this thesis.

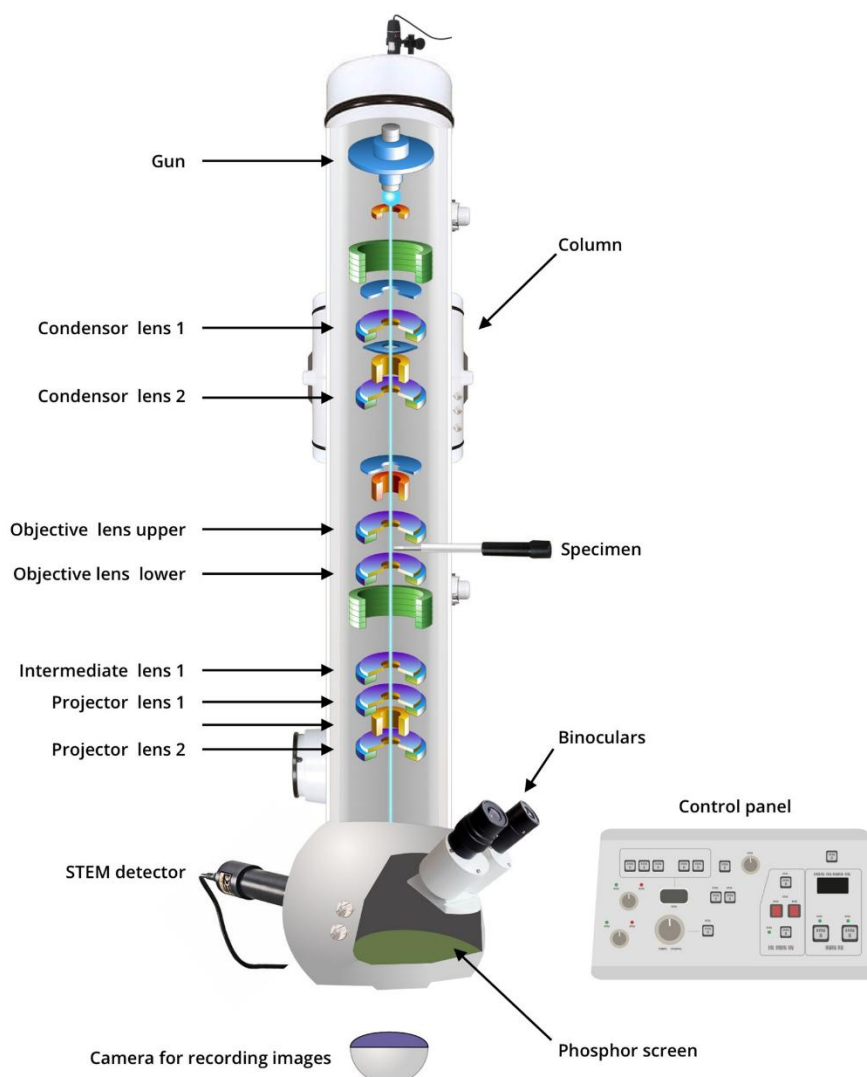


Figure 3.13. Schematic of a transmission electron microscope.

3.4.3. Atomic force microscopy

Atomic force microscopy is a popular technique to analyse the surface morphology, roughness, and nanoscale features. It operates based on the principle of scanning a sharp tip attached to a flexible cantilever across the sample surface. As the tip interacts with the surface—via van der Waals forces, electrostatic forces, or repulsive contact forces—the cantilever deflects in response to local topography. A laser beam reflected from the back of the cantilever is detected by a photodiode, converting mechanical deflection into high-resolution surface height data. A Bruker Dimension Icon system was used for nanostructure roughness, QD size measurement under peak force tapping mode, and conductive atomic force microscopy was used to measure

the current map of the NW tip in this thesis. The system offers excellent mechanical and thermal stability, along with a closed-loop scanner for high-precision measurements over large scan areas.

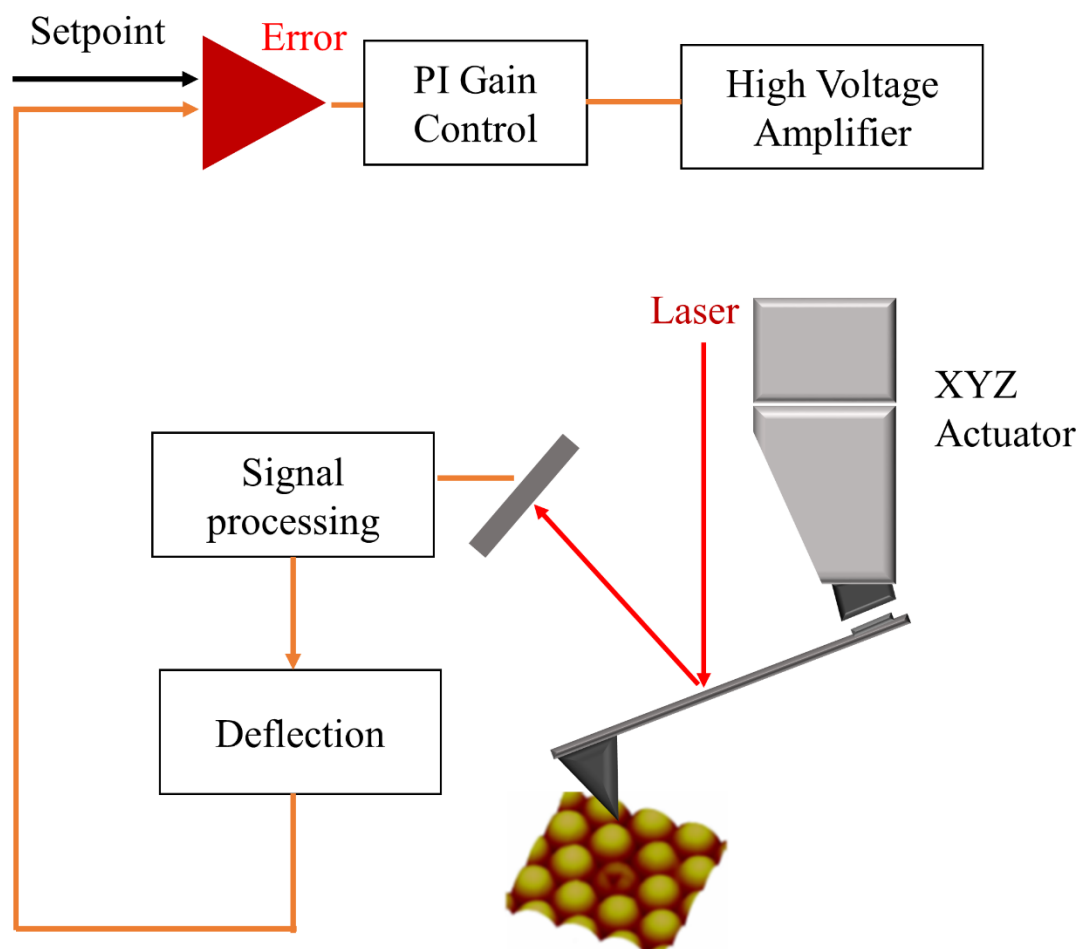


Figure 3.14. Schematic of the atomic force microscopy system.

3.4.4. Ellipsometry

An ellipsometer is a powerful, non-destructive optical instrument used primarily to measure the thickness and complex dielectric properties (such as refractive index, extinction coefficient, surface roughness, bandgap, mobility) of thin films. Ellipsometry is based on the measurement of changes in polarisation of light upon reflection (or transmission) from a sample surface. The schematic of the technique in Figure 3.15 shows when linearly polarised light hits the surface of a sample at an oblique angle, it becomes elliptically polarised due to differences in reflection between the p- and s-polarised components. The instrument detects this change, which is sensitive to the sample's optical characteristics and layer thickness. A J.A. Wollam M2000 ellipsometer was used to measure the thickness of SiO₂ and resists in this thesis.

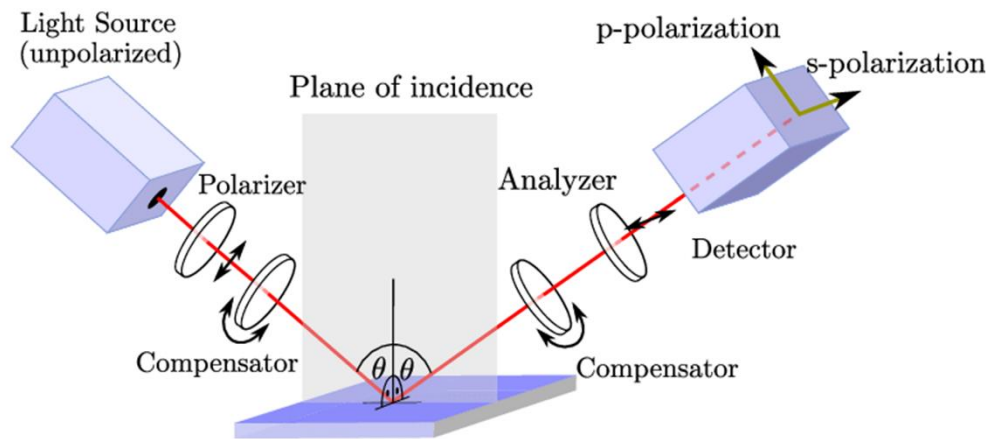


Figure 3.15. Schematic of the ellipsometry technique ^[212].

3.5. Optical and electrical characterisation techniques

In this thesis, the optical properties of the QDs in NWs and rings were measured by photoluminescence, based on which we can further determine other properties such as lifetime, single photon purity, and linewidth. Cathodoluminescence was used to measure the position of the QD along the NW. Electroluminescence and current-voltage were used for QD-in-NW device characterisation in chapter 6.

3.5.1. Cathodoluminescence

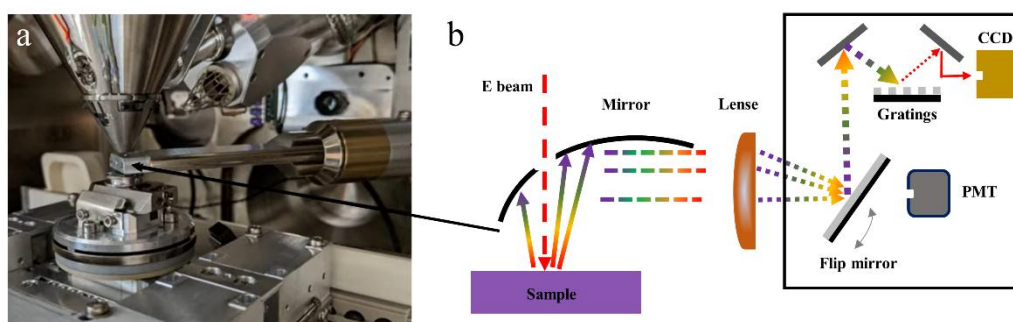


Figure 3.16. Working mechanism of a cathodoluminescence system. (a) An image of a Gatan MonoCL4 Elite cathodoluminescence spectroscopic system equipped in the FEI Verios 460 scanning electron microscope. (b) Schematic of the cathodoluminescence system.

Cathodoluminescence (CL) is a photon emission phenomenon that occurs when high-energy electrons interact with a material, inducing radiative electronic transitions ^[213]. It is usually integrated with a scanning electron microscope to use its high energy electron beam as the excitation source. When an electron beam strikes the sample surface, it transfers energy to the electrons in the material, promoting them from the valence band to the conduction band or exciting defect or impurity states. These excited charge carriers (electrons and holes) then relax back to lower energy states. If this relaxation occurs via radiative recombination, photons are emitted. The energy (and therefore wavelength) of the emitted photons corresponds to the energy gap between the involved states, such as the bandgap or defect levels.

Figure 3.16(a-b) shows the schematic of a Gatan MonoCL4 Elite cathodoluminescence spectroscopic system equipped on the FEI Verios 460 scanning electron microscope. The

electron beam goes through the hole on the parabolic mirror to the sample surface, whilst the emitted light can be collimated by the mirror onto the detector. There are two different analysis modes in CL monochromatic mode and panchromatic mode, which map the imaging area at a selected narrow wavelength range of the entire spectral information, respectively. For panchromatic mapping, the collected light is directed to a high sensitive UV optimised photomultiplier tube detector (200 – 900 nm), while for monochromatic mode, the collected light is first passed through a grating before being detected by a Princeton instrument PIXIS PX100BX CCD (Si based, 200 – 1000 nm). Measurements can be taken from 80 – 300 K.

In this thesis, all the measurements were taken at 80 K (under liquid nitrogen cooling) with a voltage of 15 kV and current of 100 pA. In chapter 4 for QD-in-NW study, CL was performed to determine the position of QD along the NW as well its emission wavelength.

3.5.2. Confocal microscopy

In conventional wide-field microscopy, the entire sample is illuminated and detected simultaneously. In contrast, confocal microscopy is an advanced optical imaging technique that enhances the resolution and contrast of fluorescence or reflected light microscopy by using point illumination and spatial filtering to eliminate out-of-focus light ^[214]. Specifically, in the confocal setup illustrated in Figure 3.17, a laser beam is directed through a pinhole and is tightly focused onto a single point on the sample through an objective lens. The light emitted or reflected from this point is collected by the same lens and passes through another confocal pinhole placed in front of the detector. This pinhole allows only light from the focal plane to reach the detector while blocking out-of-focus light from above or below the plane. The result is a sharp optical section of the sample at a specific depth, which helps to excite and collect only one isolated quantum emitter within the excitation area.

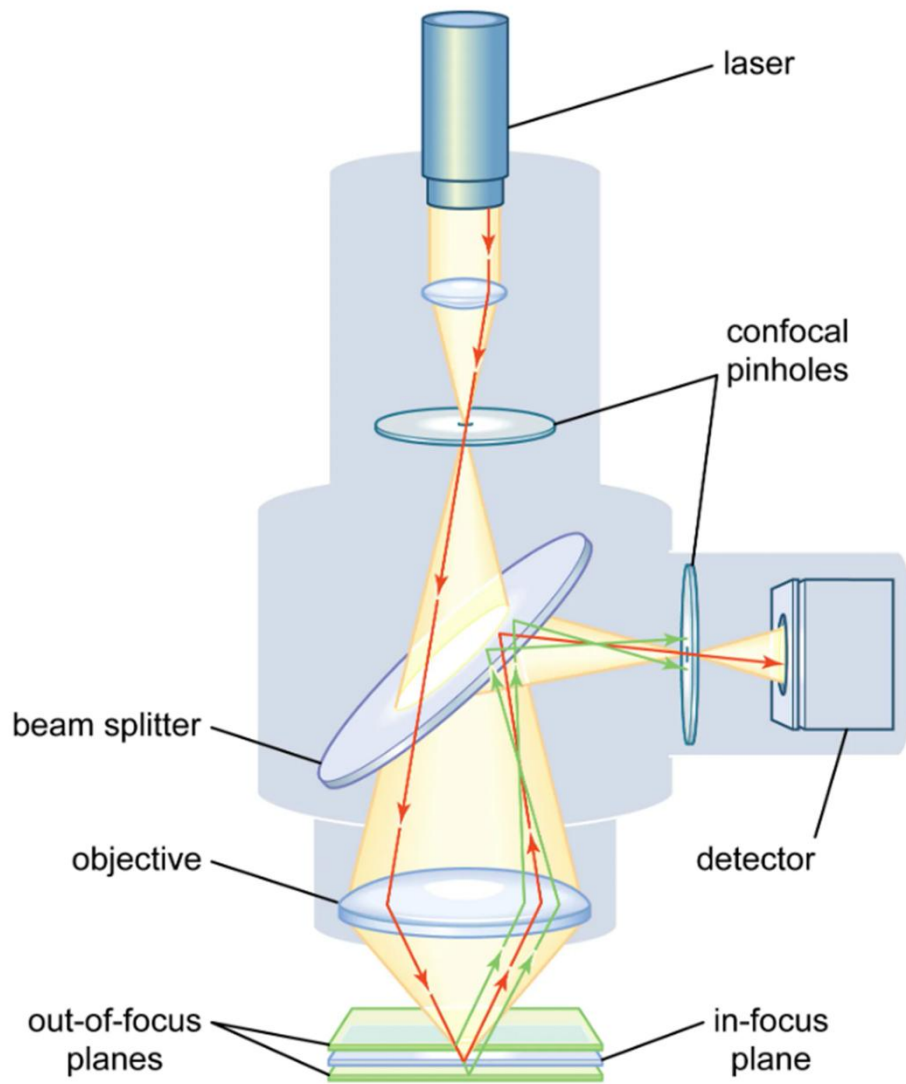


Figure 3.17. Schematic of the working principle in confocal microscopy.

The resolution of the confocal microscopy is dependent on the diffraction limit of the laser, where lateral resolution of $0.37\lambda/\text{NA}$ and axial resolution of $1.5n\lambda/\text{NA}$ for the ideal confocal microscope.

A variation of the confocal microscopy technique is scanning confocal microscopy^[215], where an additional scan mirror is put in front of the first confocal pinhole. A luminescence map can then be obtained by collecting data point-by-point from the in-focus plane when scan the mirror to change the excitation position. The scanning confocal microscopy was extensively used throughout this thesis for mapping QDs in the NW arrays or micro-rings.

3.5.3. Photoluminescence and electroluminescence

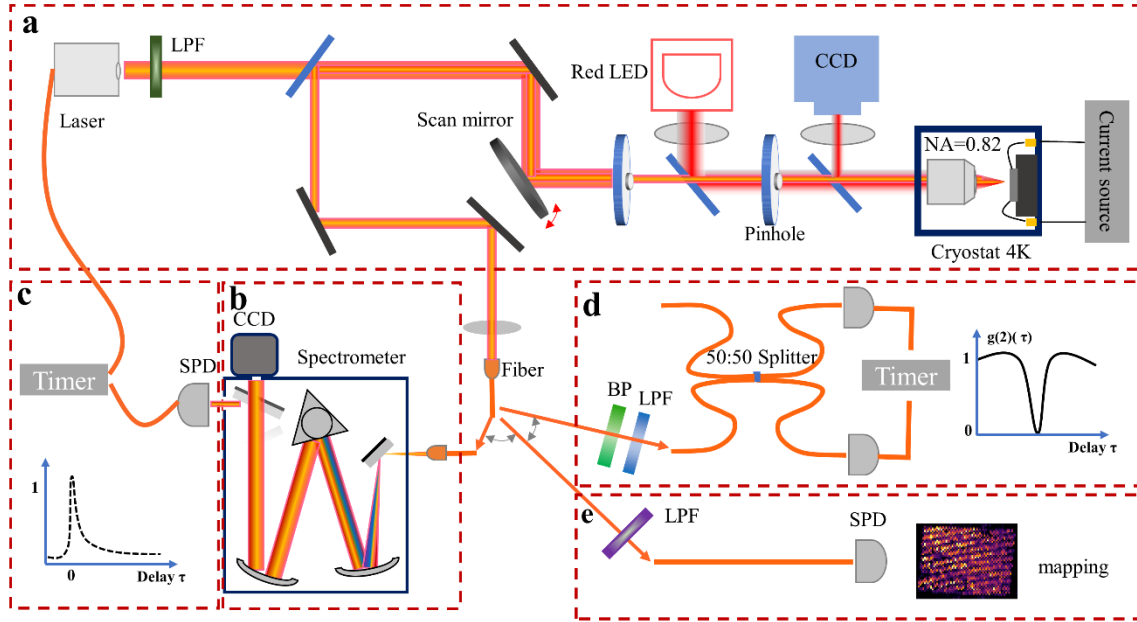


Figure 3.18. Schematic of the confocal micro-luminescence system setup for photoluminescence and electroluminescence measurements of the samples. (a) Excitation path (laser for optically pump, current source for electrically pump). (b) Light collection to spectrometer for measuring the luminescence signal. (c) Lifetime measurements set up. (d) Hanbury-Brown-Twiss setup for second-order correlation $g^{(2)}(0)$ measurements. (e) Signal mapping setup.

Figure 3.18 illustrates the schematic of the confocal micro-luminescence system for photoluminescence and electroluminescence measurements of the samples at cryogenic temperatures. The samples were maintained at a temperature of 4 K in a closed-loop cryostat (AttoDRY800), Figure 3.18(a) shows the excitation path which focuses the pump laser (for optical excitation of the QDs) on the sample through two pinholes and a 65X objective lens (NIR, NA = 0.82). The combination of an LED and a CCD camera is used for positioning the sample with respect to the imaging system. A Keithley source meter (2612B) was used to apply current for the electrical injection for the QD-in-NW devices. The luminescence signal was measured using a spectrometer (Andor shamrock) with 300, 600, and 1800 lines/mm grating and a silicon CCD camera (Newton), as shown in Figure 3.18(b). An extra collection path through the grating inside the spectrometer to a single photon detector (SPD, PDM) together

with a time-triggering device (HydraHarp 400) were used to measure the time delay between the luminescence signal and laser as shown in Figure 3.18(c). Time-resolved photoluminescence measurements were carried out using a 532 nm femtosecond pulsed laser with a repetition rate of 10.2 MHz and pulse duration of 200 ps. The instrument response function for the lifetime measurement is 50 ps. Figure 3.18(d) shows the second-order correlation measurement using a Hanbury-Brown-Twiss setup by filtering the emissions from the QDs before directing the signal to two single-photon detectors. Figure 3.18(e) depicts the optical path for the QD emission signal for mapping measurements, where the signal is filtered with a long-pass filter to remove signals with wavelengths shorter than 870 nm, before being collected by a single-photon detector (Excelitas SPCM-AQRH-14-FC). The integration time for the confocal mapping was 25 ms per pixel.

In this thesis, all the photoluminescence and electroluminescence measurements, including single photon purity and mapping were performed at University of Technology Sydney. Lifetime and temperature dependent photoluminescence were performed at the Australia National University.

3.5.4. Current–voltage measurements

To evaluate the electrical performance of the fabricated p–i–n QD-in-NW devices, current–voltage (I–V) measurements were carried out using a Keithley Tektronix Source Meter (2612B), which provides integrated voltage sourcing and current measurement capabilities with high resolution and stability. This characterisation is essential for verifying the diode functionality of the NW junctions and assessing their suitability for electrically driven single-photon emission. I–V curve provides key insights into the junction quality, contact quality, and carrier transport mechanisms within the device. All I–V measurements in this thesis were performed at both room and cryogenic temperatures.

A typical I–V curve of the semiconductor p–n junction diode is shown in Figure 3.19, with current increasing exponentially after a turn-on voltage and being blocked in the reverse bias direction, which allows only a small leakage current until breakdown. Specifically, in an ideal p–n junction, the I–V relationship is given by,

$$I = I_0(e^{\frac{qV}{nkT}} - 1) \quad (3.4)$$

where I is the measure current, I_0 is the reverse saturation current, n is the ideality factor, V is the applied voltage. k , T , and q are Boltzmann constant, temperature, and electron charge, respectively. In the forward region, when the bias exceeds the internal barrier, A positive current rises sharply, showing a non-linear curve; the corresponding voltage is referred to as the threshold (turn-on) voltage. In contrast, under reverse bias, only a very small negative current flows, mainly due to thermally excited electrons diffusing across the p–n junction. However, when the reverse bias becomes sufficiently large, the negative current increases rapidly as the diode undergoes breakdown.

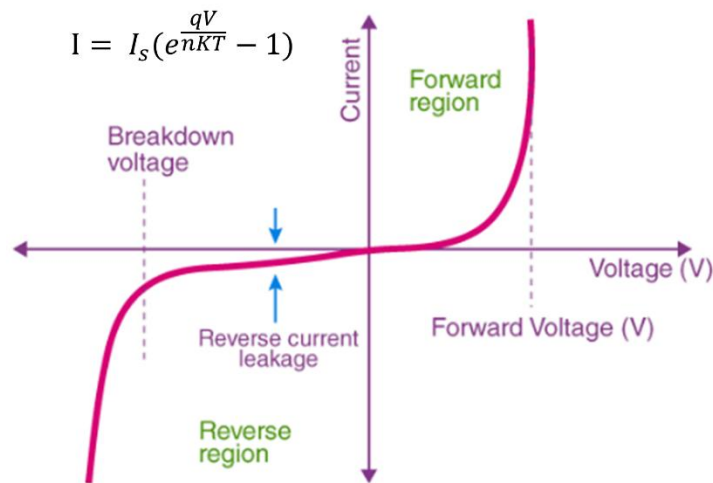


Figure 3.19. Typical current–voltage curve of semiconductor p–n junction diode.

3.6. Theoretical simulation

3.6.1. Finite difference time domain

Finite difference time domain (FDTD) is a numerical method used to solve Maxwell's equations in the time domain by discretising both space and time. It enables the simulation of electromagnetic wave propagation in complex nanophotonic structures and is widely used to analyse optical field distributions and device performance ^[216]. It's a powerful tool for simulating how light interacts with materials and structures. In this thesis, we used a commercially available software package Ansys Lumerical FDTD to design single QD emission in InP NW and micro-ring cavities and calculate their Purcell factor, Q-factor, as well as the resonant mode profiles.

In the simulations, a dipole source, representing emission from the QD, was placed at the field antinode of the fundamental cavity mode to maximise coupling. The dipole was polarised along the axis of the NW or ring symmetry, and simulations were typically carried out across a wavelength range of 900–1000 nm for InAs(P) QD emission. Perfectly matched layer boundary conditions with a distance of at least half a wavelength distance were applied in all directions to eliminate reflections and emulate an open-space environment.

Purcell factors were calculated from the ratio of the power emitted by the dipole source in a NW/ring structure to the power emitted by the same dipole in a homogeneous (bulk) medium. Electric and magnetic field monitors were placed strategically within the cavity to record the resonant field distributions, decay dynamics, and spatial mode profiles. Time monitors were placed within the cavities, which can be used to calculate the Q-factor by finding the resonance frequencies of the signal and measuring the full width half maximum of the resonant peaks.

3.7. Summary

In this chapter, the basic operations and principles of experimental techniques related to this thesis are described. In Section 3.2, The details of fabricating patterns for selective area epitaxy and equipment used such as minutes before loading into the MOCVD reactor for growth. Plasma-enhanced chemical vapour deposition, electron beam lithography, Inductive-coupled plasma reactive ion, barrel etcher, MOCVD are discussed. Section 3.3 discusses the NW device fabrication processes and related fabrication techniques of electron beam evaporation, direct write lithography, sputter deposition, and wire bonding. Section 3.4 discusses the material characterisation techniques for morphological, structural, compositional and optical analyses of the nanostructures, including scanning electron microscopy, transmission electron microscopy, atomic force microscopy, ellipsometry. Section 3.5 discusses the techniques that used for optical characterisation – cathodoluminescence equipped with scanning electron microscopy system, photoluminescence, electroluminescence, single photon purity, temperature dependent measurements based on confocal luminescence spectroscopy, and current-voltage measurement to investigate the quality of the p-n junction. Section 3.6 discusses the numerical technique using Lumerical finite difference time domain for designing QD emission within the NW/micro-ring cavity.

CHAPTER 4

4. Scalable Bright and Pure Single Photon Sources by Droplet Epitaxy on InP Nanowire Arrays

4.1. Introduction

Quantum technologies such as quantum simulation ^[217], and quantum computing ^[218], have witnessed substantial advances in recent years. A high-performance quantum light source is a key component of these systems, as they provide single or entangled photon pairs ^[219, 220]. As discussed in Chapter 1, III-V semiconductor single QDs is one of the most promising candidates with their superior advantages. However, the realisation of scalable and deterministic single QDs in optical cavities still challenging ^[136]. While top-down approach is time-consuming in identifying suitable sparse single QD regions ^[221] and unavoidable sidewall roughness leads to poor optical output efficiencies ^[222], bottom-up methods provide a more efficient route for fabricating single QDs in high-quality optical cavities. Among these cavities, the quantum dot-in-nanowire (QD-in-NW) configuration is the most favoured bottom-up device architectures ^[223], mainly grown by the vapour-liquid-solid (VLS) and selective area epitaxy (SAE) techniques, as discussed in Chapter 2. The VLS method often employs foreign catalysts such as gold and operates at relatively low temperatures, which can degrade crystal quality ^[224, 225]. In contrast, QDs grown via SAE are generally constrained by the size of the mask openings, making it difficult to achieve diameters of 20 nm or smaller ^[226, 227]. Moreover, forming uniform QD-in-NW heterostructures (e.g. InAs QDs in InP or GaAs NWs) remains challenging in SAE. This largely due to the memory effect of the system with short growth time of QDs (normally a few seconds) ^[228-230], resulting in fluctuations of exciton emission lines ^[231]. Consequently, producing small QDs (≤ 20 nm) with strong carrier confinement, which is essential for reliable single-photon emission, continues to be a significant challenge ^[223, 232].

In this chapter, we demonstrate the growth of single InAsP QDs in SAE-grown InP NW arrays by using the droplet epitaxy (DE) technique. We systematically investigate droplet density on one single NW by changing different growth parameters (diameter of NW, pitch size, In precursor deposition time and height of NW). One droplet in a NW is achieved in an array scale

with high uniformity. The structural properties of the droplets and the QDs after crystallisation are then systematically investigated. InP shell layer to surround the QDs is also optimised by adjusting different parameters like V/III ratio, array pitch size, and growth temperature. Optical properties, including emitter brightness and QD wavelength distribution, were investigated using low-temperature confocal microscopy. Based on these measurements, further characterisation of the emission lifetime and single-photon purity were performed. FDTD simulation was used to investigate the simulated Purcell factor and coupling mode when the QD is coupled to the NW dominant mode. This Chapter shows great promise for bottom-up, scalable fabrication of SQDs in various optical cavities in the development of quantum optical circuits.

4.2. Experimental methods and design

Material growth: NWs were grown on the SiO₂ patterned semi-insulating InP (111) A substrates using a horizontal flow metalorganic vapour phase epitaxy system operated at a pressure of 100 mbar. Prior to growth, the substrates were thermally deoxidised at the temperature of 750 °C for 10 mins. InP NWs were initially grown at 730 °C with PH₃ and TMIIn flow rates at 4.9×10^{-4} mol/min and 6.07×10^{-6} mol/min, respectively, corresponding to a V/III ratio of 81. The NW height (~1, ~2, ~3, and ~4 µm) was controlled by adjusting the growth time (2, 2.5, 3, and 3.5 mins). After growth, the substrate temperature was reduced to 400 °C under continuous PH₃ flow and stabilise for 3 minutes. Subsequently, the PH₃ supply was stopped and the chamber was purged for 30 seconds to remove residual group V species. TMIIn was then introduced at a flow rate of 2×10^{-6} mol/min for a fixed amount of time, followed by immediate cooling to study droplet formation. For TEM analysis, ~3 µm NWs were exposed to TMIIn for 0.25 s and then crystallised at 520 °C using AsH₃ and PH₃ flow rates of 1.2×10^{-4} mol/min and 1.78×10^{-4} mol/min, respectively, for 5 mins. The ratio of AsH₃ / (AsH₃ + PH₃) was maintained to 0.4, followed by substrate cooling down process. For optical characterisation, samples were prepared with a bilayer InP shell grown after droplet crystallisation. The first shell layer was grown at a temperature of 520 °C for 3 mins, using TMIIn and PH₃ flow rates of 2.99×10^{-6} mol/min and 1.2×10^{-2} mol/min, respectively. Then the substrate temperature was increased to 600 °C and stabilised for 3 mins under PH₃ flow and the second InP shell layer was grown for 10 mins with a TMI and PH₃ flow rate of

6.07×10^{-6} mol/min and 4.91×10^{-3} mol/min, respectively. The high temperature layer shell was to anneal the QDs and the shell layer to further improved their crystal quality.

Material properties characterisation: the density and size of the droplets on InP NWs were characterised by a FEI Verios 460 SEM system operated at an accelerating voltage of 10 kV and a beam current of 50 pA. CL measurements were conducted in the same system at 15 kV and 100 pA, equipped with a Gatan MonoCL4 Elite Module and a circulating liquid nitrogen cooling system to maintain a temperature of 77 K. Structural and compositional characterisation were carried out using an aberration-corrected scanning transmission electron microscope (JEOL 2100F FEGTEM) fitted with an energy dispersive X-ray spectroscopy system, operated at an accelerating voltage of 200 kV, a beam current of 131 μ A.

Experimental design:

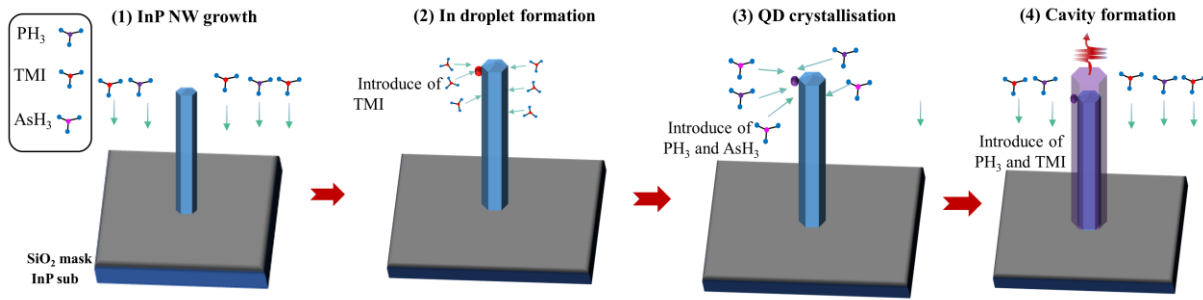


Figure 4.1. Combination of droplet epitaxy with selective area epitaxy to grow single QDs in InP NWs. (1) Selective area growth of InP NW, (2) Introduce of TMI gas to deposit In droplet on the NW, (3) Introduce of PH₃ and AsH₃ to crystallise droplet into QD, (4) Growth the shell layer of InP to form the cavity.

As discussed in Chapter 2.1.3, the Stranski-Krastanov (SK) and DE methods are the two most widely used bottom-up approaches for forming QDs in III-V materials. The SK mechanism depends on strain arising from lattice mismatch between the QD material and the substrate material, whereas DE forms QDs through the crystallisation of group III metal droplets ^[233]. For growth of Single QDs in SAE-grown NW cavities, DE offers several advantages over the SK growth mode, including: (i) a greater ease of formation on high-Miller-index surfaces ^[234], (ii) less temperature dependent for sparse QDs ^[235], (iii) fewer limitations on material

combinations (composition of QDs) for tuning the emission wavelength ^[236], and (iv) the absence of wetting layer avoids hybridised quantum states ^[237]. In this thesis, we take the advantages of DE in combination with SAE to deterministically grow single quantum dots in optical cavities. Figure 4.2 illustrates the experimental procedure for growing single QDs in NWs by combining In droplet epitaxy with selective-area epitaxial growth of InP NWs. The InP NWs are first grown on an InP substrate using PH₃ and TMI as precursor gases, with a SiO₂ layer serving as a mask containing small openings, as shown in (1). In the next step (2), TMIn is introduced in the absence of group-V precursors, decomposing to form small In droplets at high-energy sites near the NW tip. Once the droplets are established, group-V sources such as PH₃ and AsH₃ are supplied to crystallise the droplets into QDs (3). Finally, a shell layer of InP is grown by reintroducing PH₃ and TMI, encapsulating the QD and simultaneously forming an optical cavity.

4.3. Engineering the droplet density on single NWs

In this study, indium droplets are utilised as nucleation seeds for the formation of QDs such that the droplet density directly reflects the QD density. For single-photon source applications, the optimal configuration corresponds to one droplet (i.e. QD) per NW. In the following section, we systematically examine the influence of key growth parameters, including NW diameter, pitch size, height, and the TMI flow rate, on the resulting droplet density.

4.3.1. Diameter of NW

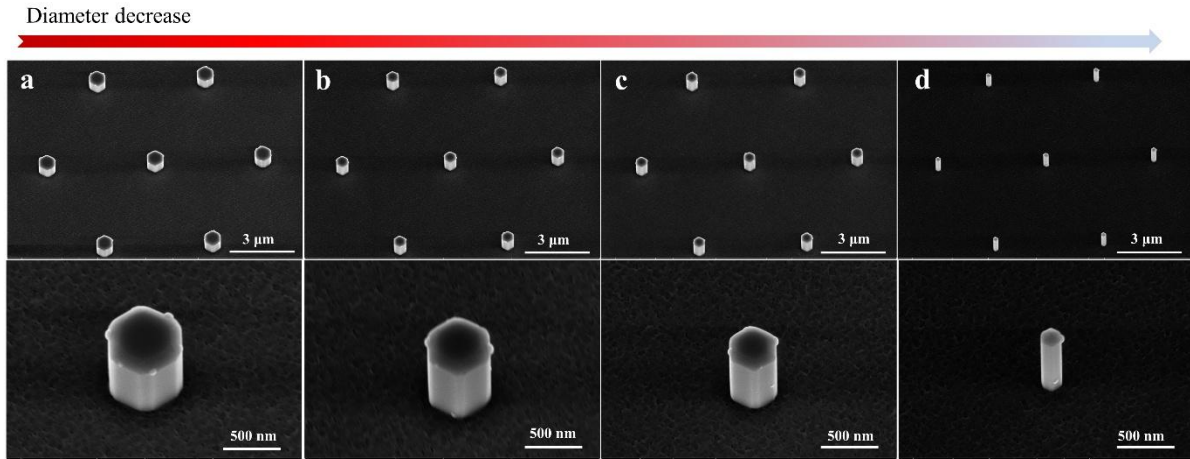


Figure 4.2 SEM images showing droplet density as a function of NW diameter, at a fixed pitch of 3 μm , a TMI flow rate of 4.0×10^{-6} mol/min, and a height of ~ 0.5 μm . The results are shown for NW diameters of (a) ~ 700 nm, (b) ~ 500 nm, (c) ~ 300 nm, and (d) ~ 200 nm.

To elucidate the formation of QDs in NWs, we first examined the influence of NW diameter. InP NW arrays were grown to the target diameters and heights with a pitch of 3 μm ; the growth temperature was then reduced to 400 $^{\circ}\text{C}$ in a PH_3 ambient. PH_3 was subsequently purged, and TMI was introduced into the growth chamber at a flow rate of 4.0×10^{-6} mol/min for 0.5 s to form In droplets. Figure 4.3 shows the droplet density as a function of NW diameter: (a) ~ 700 nm, (b) ~ 500 nm, (c) ~ 300 nm, and (d) ~ 200 nm. The droplets are In droplets formed as a result of TMI decomposition in the absence of group V precursors (AsH_3 or PH_3), and predominantly nucleate at high-index facets located at the vertices of the hexagonal NWs — formed by the intersection of the (0001) top facet and the $(1\bar{1}20)$ sidewalls — and therefore are more likely to form at the NW tips than on the sidewalls. Larger-diameter NWs present greater contact area and more available nucleation sites for In adatoms, resulting in a higher droplet density on ~ 700

nm NWs. Conversely, decreasing the NW diameter reduces the probability of nucleation and leads to fewer droplets, as shown in (d).

4.3.2. Pitch size of NW

For single-photon applications, the QD density must be sufficiently low to allow spatial identification during optical measurements, ideally below 1 dot/ μm^2 . In SAE growth, the pitch size of the NWs provides a direct means to control the spatial location of QD formation. Figure 4.3 illustrates the effect of array pitch size on droplet density under a fixed TMIn flow rate of 2.0×10^{-6} mol/min and a NW height of 3 μm : (a) 1 μm , (b) 2 μm , and (c) 3 μm . The indium droplets form through adatom diffusion from the substrate surface as well as direct impingement onto the NWs. At smaller pitch sizes, each NW has a reduced adatom capture area, so droplet formation is mainly governed by direct impingement and limited diffusion to specific nucleation sites. In the 1 μm pitch array, each NW tip hosts a single droplet, while almost no droplets appear along the NW sidewalls. As the pitch size increases, the capture area per NW becomes larger, enabling some droplets to nucleate along the NW length, as observed for the 2 μm array. When the pitch size increases to 3 μm , most NWs exhibit two distinct droplets.

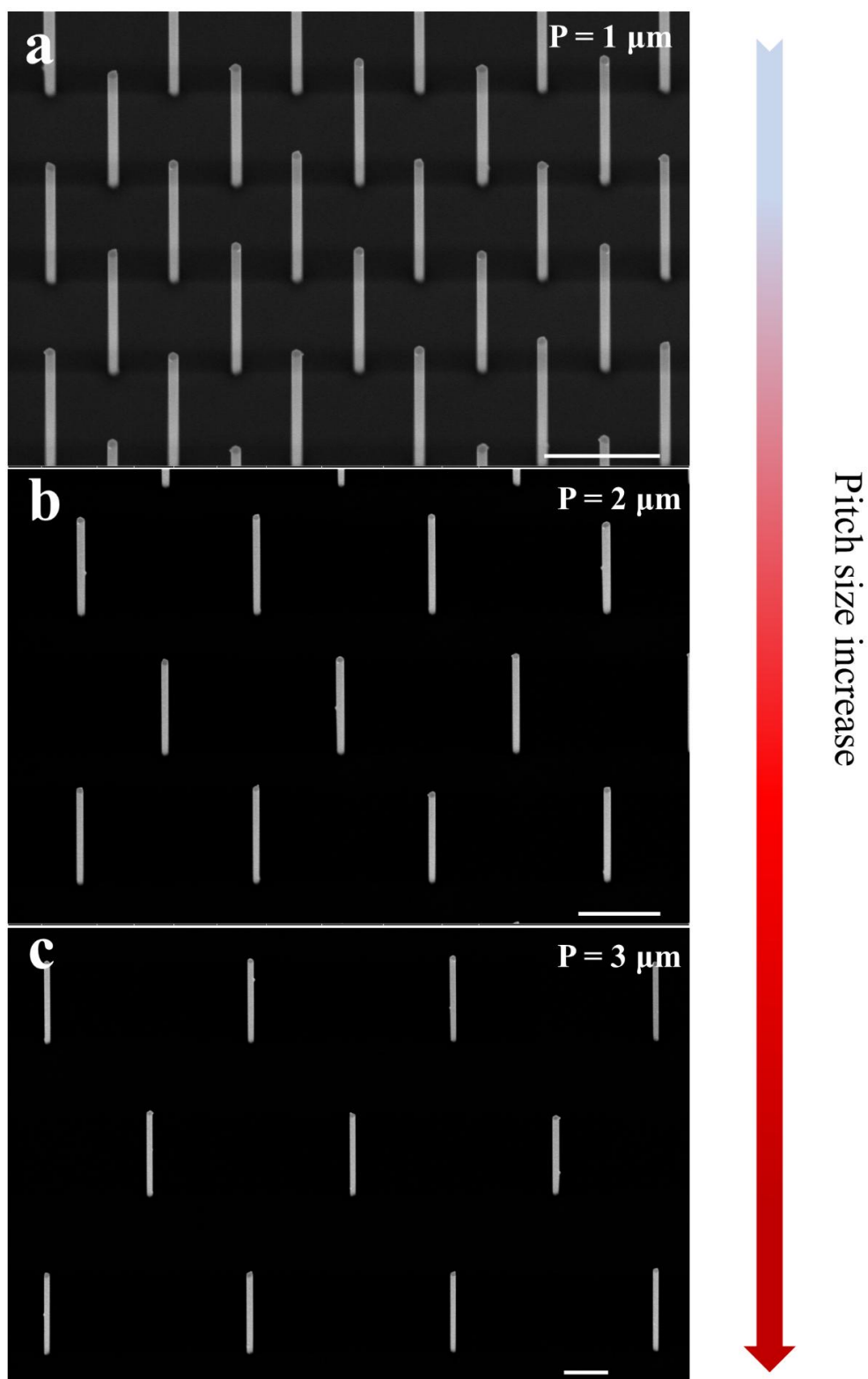


Figure 4.3. SEM images illustrating the effect of array pitch size on droplet density under a fixed TMI flow rate of 2.0×10^{-6} mol/min and a NW height of 3 μm . The investigated pitch sizes were (a) 1 μm , (b) 2 μm , and (c) 3 μm . All scale bars are 1 μm .

4.3.3. Deposition time of TMI and height of NW

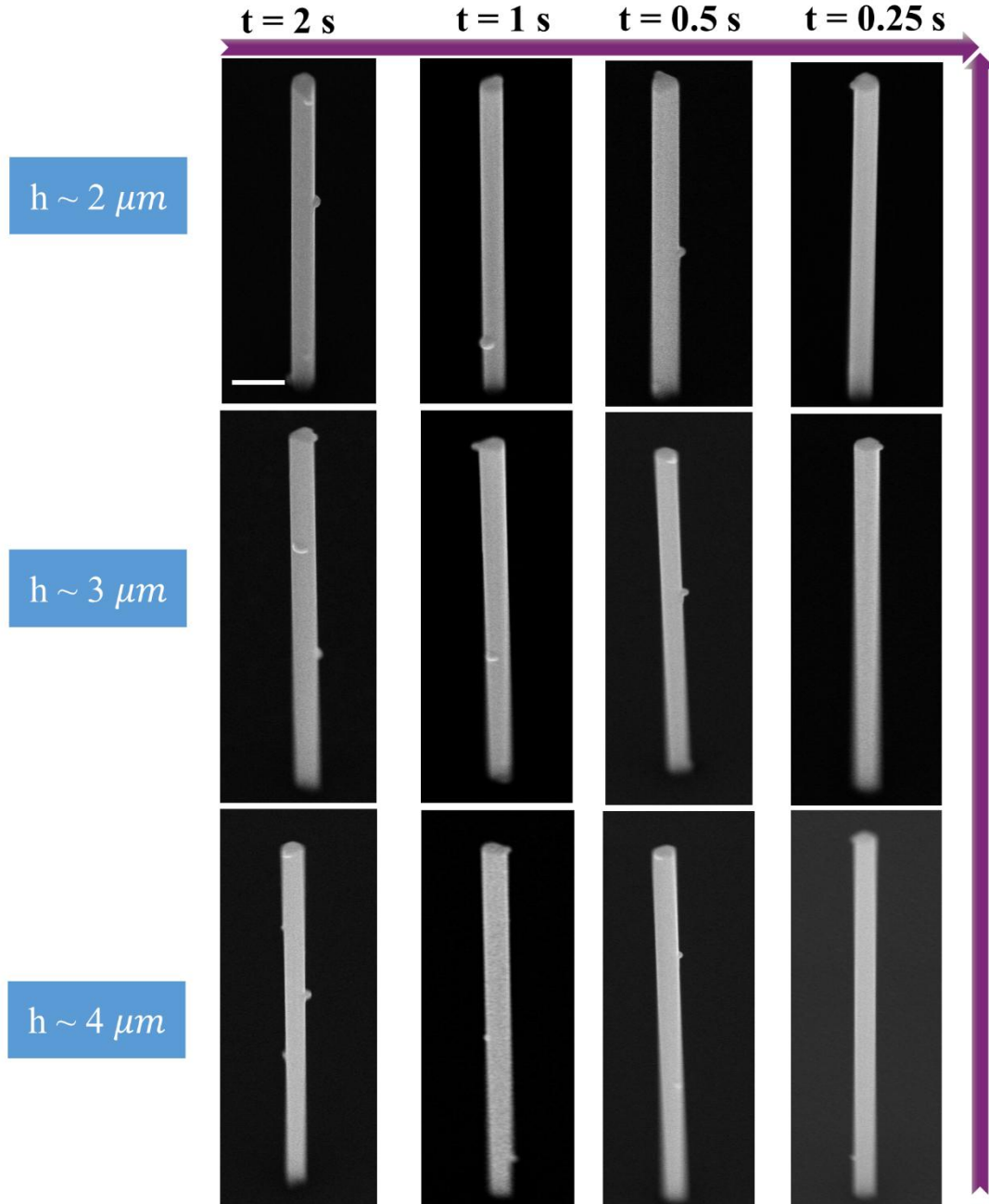


Figure 4.4. SEM images showing droplet density on a representative single NW from 3 μm pitch arrays as a function of TMI deposition time and NW height, under a fixed deposition temperature of 400 °C and a TMI flow rate of 2.0×10^{-6} mol/min. The scale bar is 100 nm.

Next, we study the droplet density on individual NW with a pitch size of 3 μm . Figure 4.4 illustrates droplet formation on the NW as a function of both NW height and TMIn deposition time. In all samples, a droplet is consistently observed at the tip of the NW, while additional droplets form along the sidewalls. By optimising both the NW length and TMIn deposition time, it is possible to achieve the formation of a single droplet exclusively at the NW tip. For example, reducing the NW length as shown in the columns decreases the density of droplets on sidewall. Similarly, shortening the TMIn deposition time as shown in the rows also reduces the droplet density. We finally achieve only one droplet is formed on the tip of the NW with TMIn deposition time of 0.25 s and NW length 3 μm . Considering that the diffusion length of In adatoms on InP at this temperature is approximately 1 μm ^[238], droplet formation is governed by both direct impingement and surface diffusion toward favourable nucleation sites. These sites are typically located at high-index facets, such as those formed at the vertices of the hexagonal NW due to the intersection of the $(11\bar{2}0)$ sidewalls. At the tip of the NW, nucleation is even more favourable because of the presence of high-energy top facets ^[239], making droplet formation more likely there than along the sidewalls. By reducing the length of the NW there is lower density of nucleation sites along the length of the NW and hence we can deterministically position the droplet at the tip of the NW. Therefore, shortening the NW reduces the density of available nucleation sites along its length, enabling deterministic positioning of the droplet at the tip. In addition, controlling the supply of In adatoms—by adjusting the TMIn deposition time—ensures that only a single droplet forms at the NW tip.

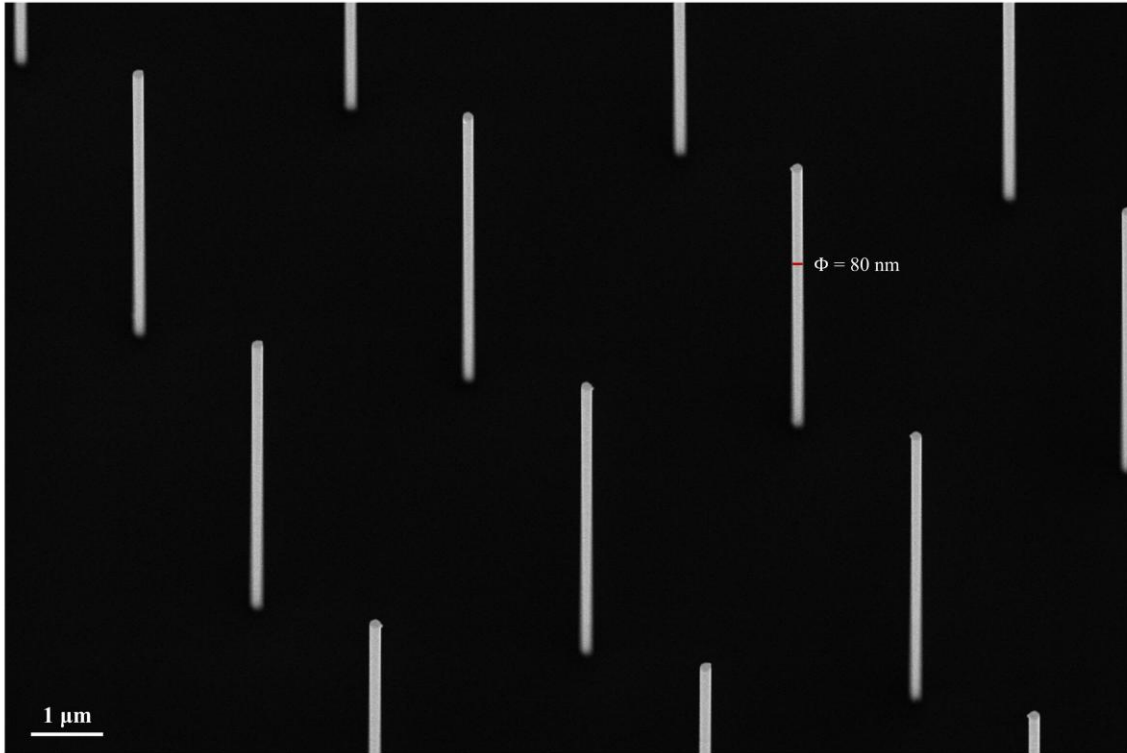


Figure 4.5. Optimised droplet density on $\sim 3 \mu\text{m}$ long NWs with a diameter of 80 nm in a $3 \mu\text{m}$ pitch array, with only one droplet formed on each NW.

After a series of optimisations, we achieved deterministic growth of a single indium droplet on each NW, as shown in Figure 4.5. The droplets were formed on $\sim 3 \mu\text{m}$ long NWs with a diameter of 80 nm arranged in a $3 \mu\text{m}$ pitch array, with only one droplet observed per NW.

4.4. Droplet (QD) size and structure properties

In this section, we investigate the size of droplets and also the structure properties of the QDs after crystallisation. The QD size provides direct information about its emission wavelength, while the structural quality is crucial for achieving good optical performance.

4.4.1. Droplet Size

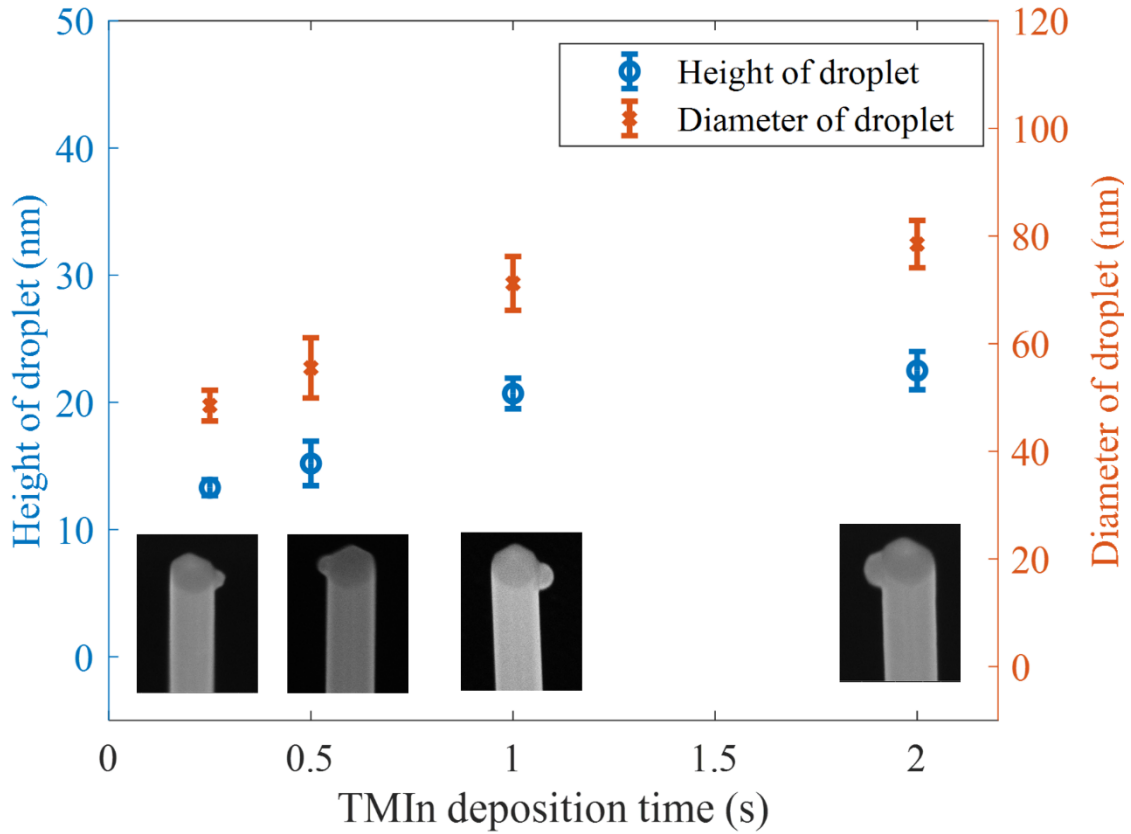


Figure 4.6. Size of the droplets at the nanowire tip as a function of TMIn deposition time, with representative SEM images for each deposition duration shown in the insets.

Figure 4.6 shows that the diameter and the height of the droplet on the tip of the NW varies as a function of TMIn deposition time. The size of the droplet decreases with shorter TMIn deposition time. A small droplet size with a diameter of 48.5 ± 2.85 nm and a height of 13.3 ± 0.63 nm can be achieved at 0.25 s, which is later used for optical performance measurements. The size of the droplet provides a direct reference for the size of QD, offering one freedom to

engineer the emission wavelength. In general, the larger the droplet (QD) the longer the emission wavelength. Another freedom is the QD composition, by changing the $\text{AsH}_3/\text{PH}_3+\text{AsH}_3$ ratio during the droplet crystallisation process, we can potential shift the QD composition from InAsP to InAs, leading to the redshift of the emission wavelength. This will be illustrated in Section 4.4.2.

4.4.2. Structural properties of the quantum dots

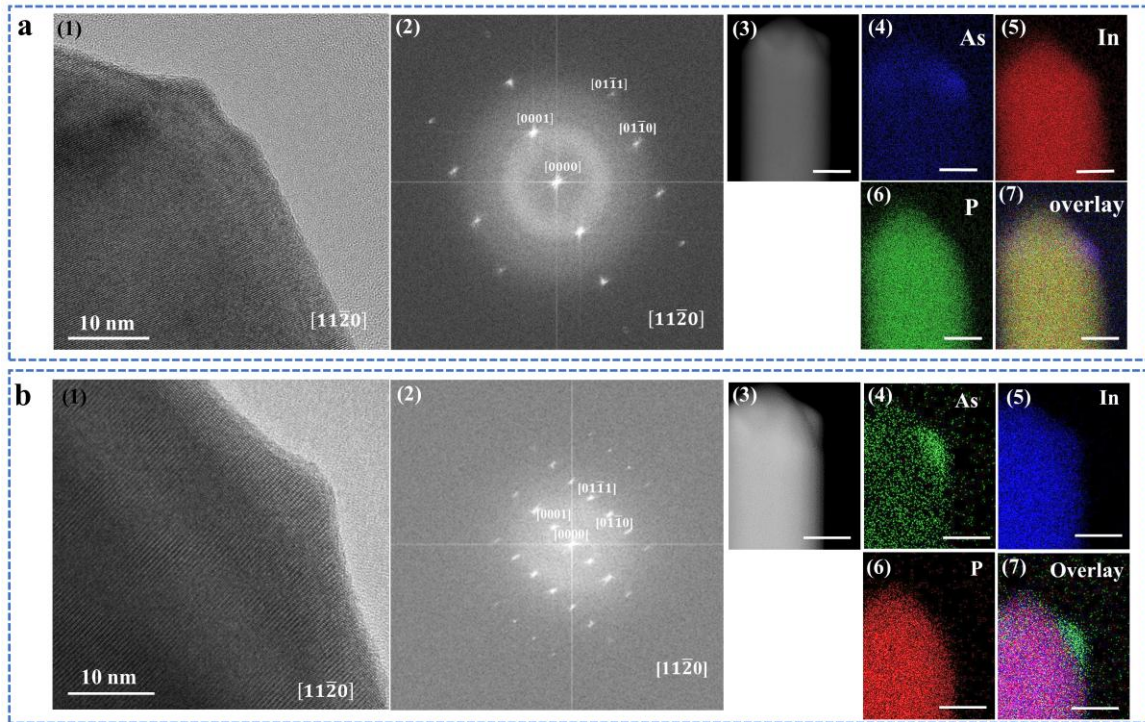


Figure 4.7. Structural properties of the QD on NW after crystallisation of the In droplets with different TMIn deposition times: (a) 0.25 s and (b) 0.5 s. (a-1) and (b-1) are the HRTEM images taken along the $[11\bar{2}0]$ crystallographic direction of the NW tip region containing QDs, (a-2) and (b-2) are the corresponding diffraction patterns, respectively. (a-3) and (b-3) are the STEM image taken at the QD regions of the NW tips, their corresponding energy dispersive X-ray spectroscopy compositional maps: (a-4) and (b-4) As, (a-5) and (b-5) In, (a-6) and (b-6) P, (a-7) and (b-7) over laid elemental maps combining the As, In, and P maps. The unlabelled scale bars are 20 nm.

The QD was formed by introducing group V precursor gases (AsH_3 and PH_3) after droplet formation. The size and composition of the QDs were analysed using (S)TEM, as shown in

Figure 4.7, under different TMIn deposition times and crystallisation conditions: (a) 0.25 s with an $\text{AsH}_3 / (\text{AsH}_3 + \text{PH}_3)$ ratio of 0.4, and (b) 0.5 s with AsH_3 only. The excellent crystalline quality of the NW is confirmed by high-resolution transmission electron microscopy (HRTEM) images taken along the $[11\bar{2}0]$ crystallographic direction, as presented in Figure 4.7 (a-1) and (b-1). The corresponding QD diameters and heights are 13 / 3.5 nm and 22.5 / 6.1 nm, respectively. The apparent size reduction is mainly attributed to the transformation of the liquid droplet into a more compact crystalline quantum dot, accompanied by atomic redistribution during recrystallisation. Both the NW and QD exhibit a WZ crystal phase with high bandgap compared to ZB, as shown in Figure 4.7 (a-2) and (b-2). This is essential for maintaining high crystal quality, particularly at the QD–NW interface within the shell layer, thereby minimising carrier scattering and trapping. Energy-dispersive X-ray spectroscopy was performed to investigate the QD composition. Figures 4.7 (a-4) and (b-4) correspond to As, (a-5) and (b-5) to In, (a-6) and (b-6) to P, and (a-7) and (b-7) to the overlaid elemental maps combining As, In, and P. These results confirm that the QDs consist of InAsP and InAs, respectively. The observed differences in size and composition provide a means to engineer and tune the emission wavelength of the QD.

4.5. Effect of growth parameter on InP shell quality

A uniform morphology of the NW is required to form a high-quality cavity for single photon emission enhancement. To study the optical properties of the QDs, samples with a double-layer InP shell were grown after droplet crystallisation. The first shell layer in all samples was grown at a crystallisation temperature of 520 °C to prevent QD evaporation after formation. The second layer was grown at a temperature of 600 °C to improve crystal quality and enable thermal annealing of the QDs for enhanced optical performance. In this section, the influence of different experimental parameters on the quality of the second InP shell layer is discussed.

4.5.1. V/III ratio

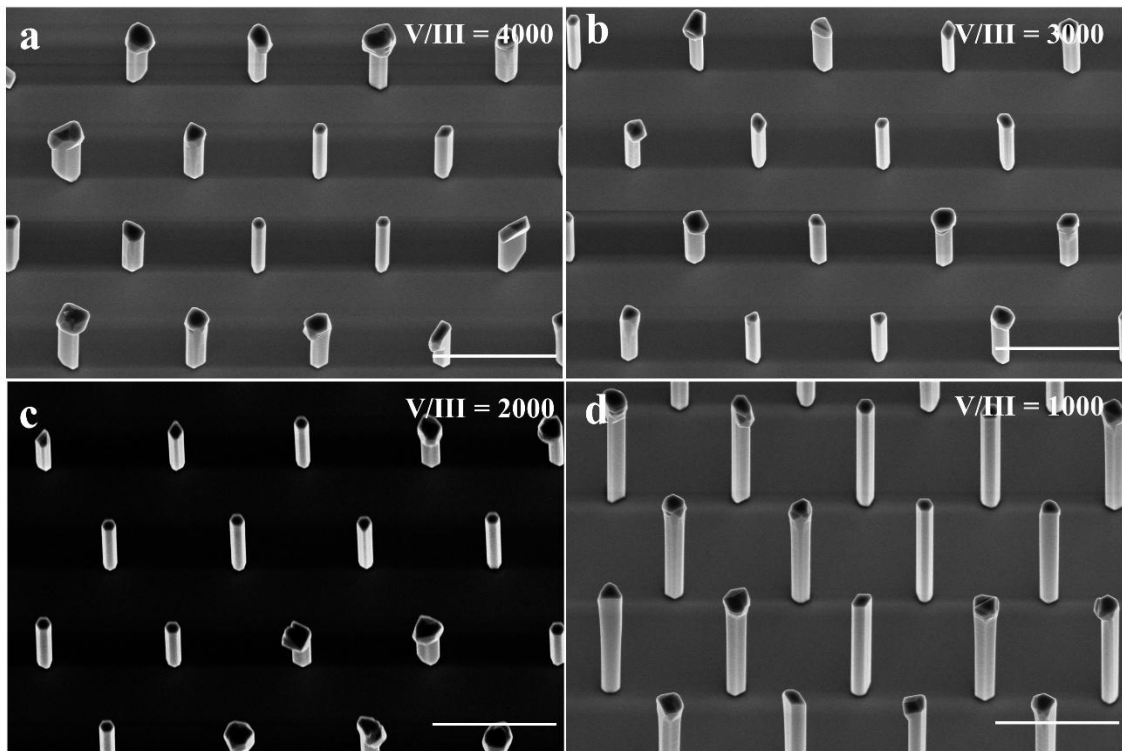


Figure 4.8. SEM images demonstrating the optimisation of the InP shell layer as a function of the V/III ratio: (a) 4000, (b) 3000, (c) 2000, and (d) 1000. All scale bars are 3 μm .

Figure 4.8 shows SEM images of NWs grown at the temperature of 650 °C under different V/III ratios: (a) 4000, (b) 3000, (c) 2000, and (d) 1000. The uniformity of the shell layer strongly depends on the V/III ratio during growth. At a high V/III ratio like 4000, the NW sidewalls

exhibit nonuniform shell coverage and irregular top facets. This is mainly due to the excessive supply of group-V species, which suppresses the In adatom diffusion across the NW, leading to nonuniform lateral growth and shell thickness variations along the NW. As the V/III ratio decreases to 1000, the sidewall coverage becomes smoother and more uniform. A lower V/III ratio enhances the diffusion of In adatoms over the NW surface, allowing them to redistribute more evenly before incorporation. Consequently, the shell layer forms with improved uniformity and smoother morphology. However, when the V/III ratio is further reduced at such a high growth temperature, the diffusion rate of In adatoms increase significantly, which may explain the slight nonuniform crystal accumulation observed at the top regions of some NWs.

4.5.2. Pitch size

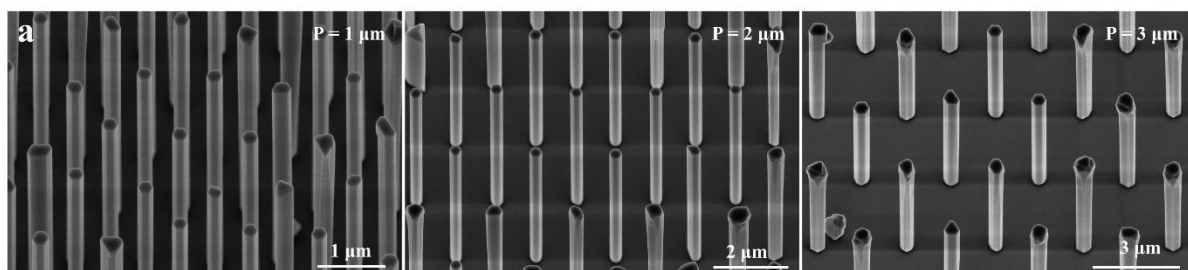


Figure 4.9. SEM images showing the optimisation of InP shell layer as a function of pitch size of the NW arrays: (a) 1 μm , (2) 2 μm , and (3) 3 μm .

The influence of pitch size is investigated at a fixed InP shell growth V/III ratio of 1000, and temperature of 650 $^{\circ}\text{C}$. From Figure 4.9, it is observed that NWs grown with smaller pitch sizes exhibit more uniform lengths across the array. This behaviour can be attributed to the diffusion and consumption dynamics of precursor species during growth. When the pitch size is large, each NW acts as an independent growth site, and the local precursor concentration above each opening may vary significantly due to nonuniform depletion of precursors. In contrast, when the pitch size is reduced, the diffusion fields of adjacent openings overlap, resulting in a smoother precursor concentration profile on the substrate surface. Such diffusion coupling minimises fluctuations in the local growth rate caused by variations in precursor supply or surface kinetics, thereby leading to more uniform axial growth and improved NW length uniformity. However, to avoid excitation of neighbouring NWs during single-NW optical

characterisation, the shell growth conditions were further optimised for a large pitch size of 3 μm .

4.5.3. Temperature

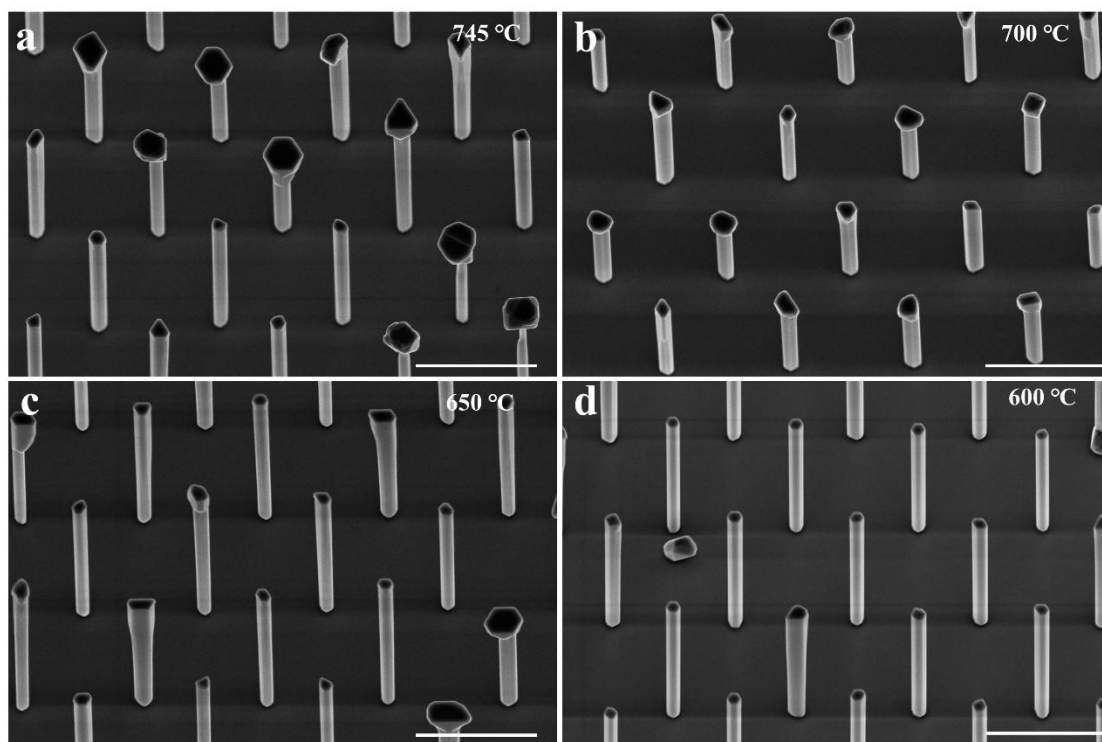


Figure 4.10. SEM images showing the optimisation of InP shell layer as a function of growth temperature: (a) 745 °C, (2) 700 °C, (3) 650 °C, and (4) 600 °C. All scale bars are 3 μm .

Figure 4.10 is the SEM images showing the evolution of the InP shell layer morphology as a function of growth temperature. As the growth temperature decreases from 745 °C to 600 °C, a clear improvement in the shell uniformity can be observed. At the highest temperature (745 °C), the tip of the NWs appears rough and some of them have a clot of InP crystal, which is likely due to higher In adatom diffusion leading to accumulation at the tip of the NW. At intermediate temperatures (700–650 °C), the shell becomes more conformal along the NW sidewalls, however some of the NWs sidewalls are still rough. By reducing the temperature further to 600 °C, the shell growth along the whole array become more uniform and smoother with good hexagonal structure due to a balanced In adatom diffusion and incorporation rate.

4.6. Optical properties characterisation

In this section, we examine the optical properties of the QDs embedded in the NWs after shell capping. Cathodoluminescence measurements are conducted to determine the position of the quantum dot along the NW, while photoluminescence (PL) is performed to evaluate the emission uniformity of the QD-in-NW array and its single-photon emission properties.

4.6.1. SEM-cathodoluminescence

So far, we have demonstrated good quality of QD-in-NW structures in Section 4.5. To investigate QD position along the NW, we transfer two QD-in-NW structures with different length and TMIn deposition time: (a): $\sim 4.5 \mu\text{m}$ and 0.25 s, (b): $\sim 6 \mu\text{m}$ and 0.5 s. The different in height is achieved by different NW core growth time of 3 mins and 4 mins, respectively.

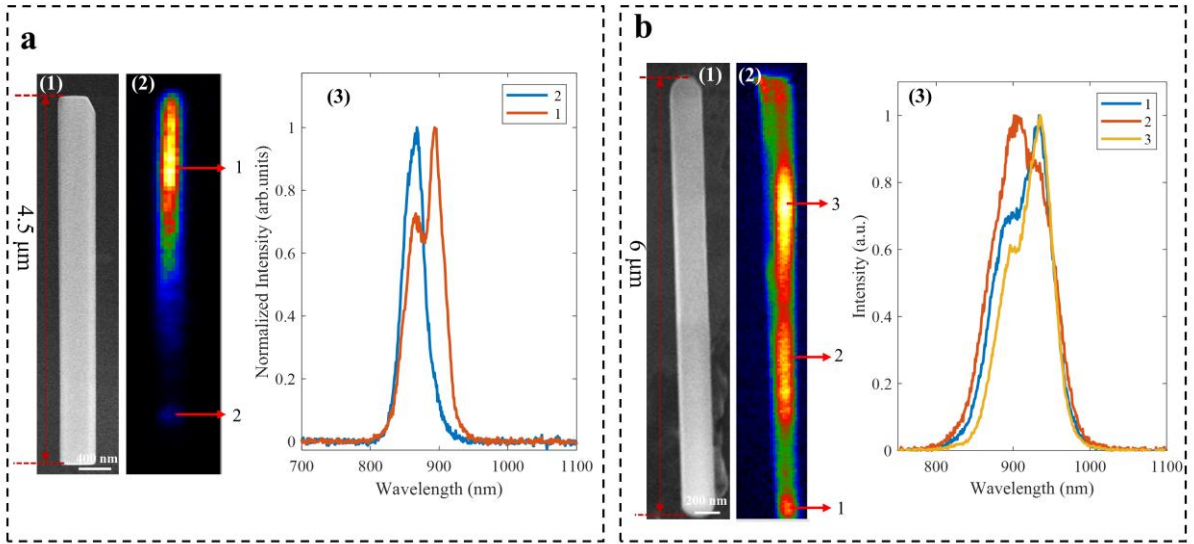


Figure 4.11. SEM-CL measurements of Dot-in-NW structures with different NW length after transfer onto Si substrates: (a): $\sim 4.5 \mu\text{m}$, (b): $\sim 6 \mu\text{m}$. (a-1) and (b-1) are the SEM images of the measured QD-in-NW, with the corresponding panchromatic CL maps at 77 K displayed in (a-2) and (b-2), respectively. (a-3) and (b-3) are the CL spectra obtained from the emission spots.

The QD-in-NW structures were transferred onto Si substrates for CL measurements. Figure 4.11(a-2) presents the panchromatic CL mapping corresponding to the NW shown in Figure 4.11(a-1), where a bright emission spot is clearly observed approximately $3 \mu\text{m}$ from the base of the NW. The CL spectra collected at points 1 and 2 in Figure 4.11(a-2) reveal emission peaks

at 865 nm and 900 nm, originating from the InP NW and the InAsP QD, respectively. This confirms the deterministic positioning of a single QD within the NWs, achieved through optimisation of the In droplet density as described in Figure 4.5. In contrast, increasing the NW length enhances the possibility of more droplet formation along the NW, as evidenced in Figure 4.11(b-2), where multiple bright spots corresponding to QD emissions around ~ 920 nm as observed in the corresponding CL spectra.

4.6.2. PL and single photon characterisation

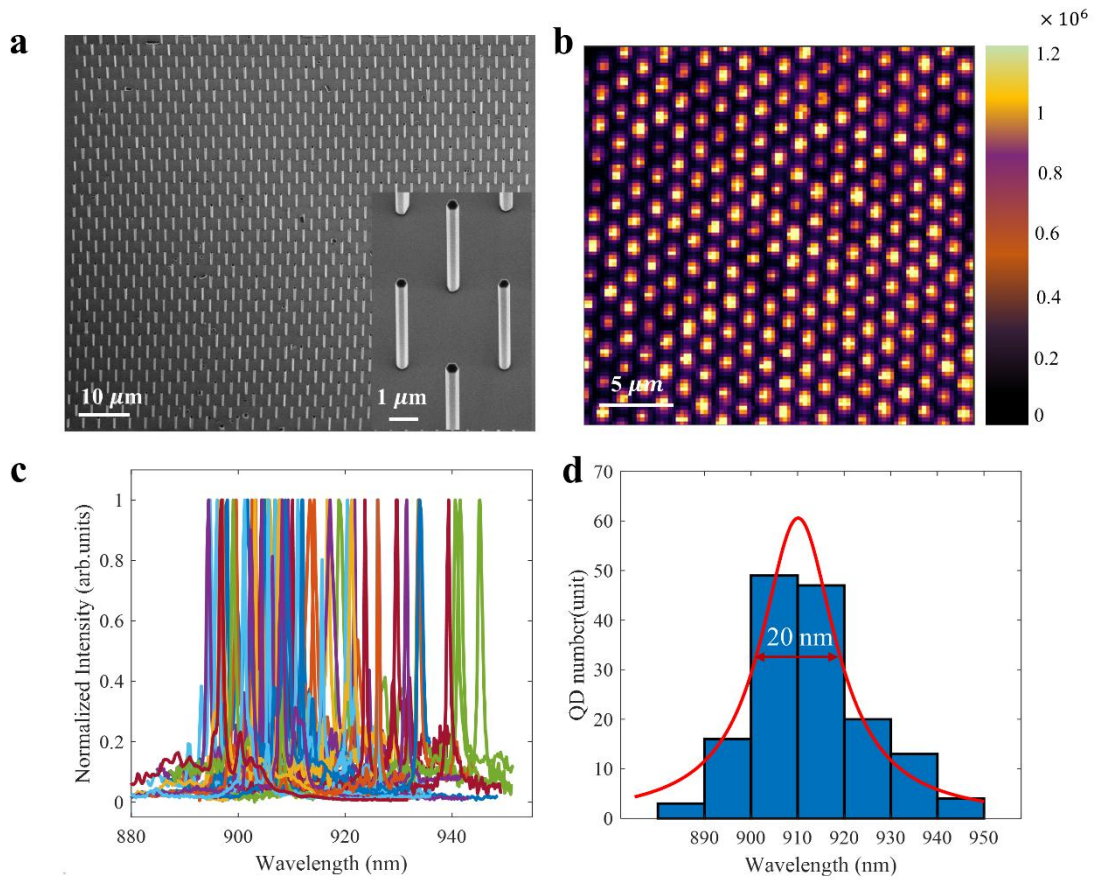


Figure 4.12. Statistical measurements of the QD-in-NW array. (a) SEM image of a $90\ \mu\text{m} \times 90\ \mu\text{m}$ QD-in-NW array after capping the QDs with InP shell layer, and the inset is a high-magnification SEM image showing the QD-in-NW array having a uniform morphology with hexagonal sidewall facets. (b) Photon intensity map of the NW array measured under 1 nW excitation from a 675 nm CW laser at 4 K. (c) Photoluminescence spectra of 70 QD-in-NWs. (d) Distribution of the emission wavelengths from 150 QD-in-NWs.

Figure 4.12(a) presents an SEM image of the NW array with a pitch size of 3 μm , demonstrating a highly uniform morphology across the entire array following the growth optimisation described in Section 4.5. After capping the QDs, individual NW cavities exhibit consistent hexagonal cross-sections (as shown in the inset), reflecting the high structural quality of the NWs. Such uniformity is essential for forming high-quality optical cavities that enhance QD emission. Micro-photoluminescence ($\mu\text{-PL}$) spectroscopy measurements were carried out in a confocal setup at the temperature of 4 K under above-band excitation using a 675 nm continuous-wave laser. Figure 4.12(b) displays the intensity map of the NW array for emission wavelengths above 870 nm. The strong uniformity observed in the mapping is consistent with the excellent morphology of the NW array. In addition, each NW exhibits high brightness, with photon count rates reaching the order of millions of counts per second using a single-photon detector under only 1 nW excitation power. Figure 4.12(c) shows the PL spectra collected from 70 QD-in-NW structures, with emission wavelengths spanning 890–945 nm. The statistical distribution of emission wavelengths from 150 samples, shown in Figure 4.12(d), follows a normal distribution centred at 910 nm, with a relatively narrow full width at half maximum (FWHM) of 20 nm. This spectral variation is attributed to slight differences in QD size and composition, and could be further reduced through improved control over droplet formation and crystallisation.

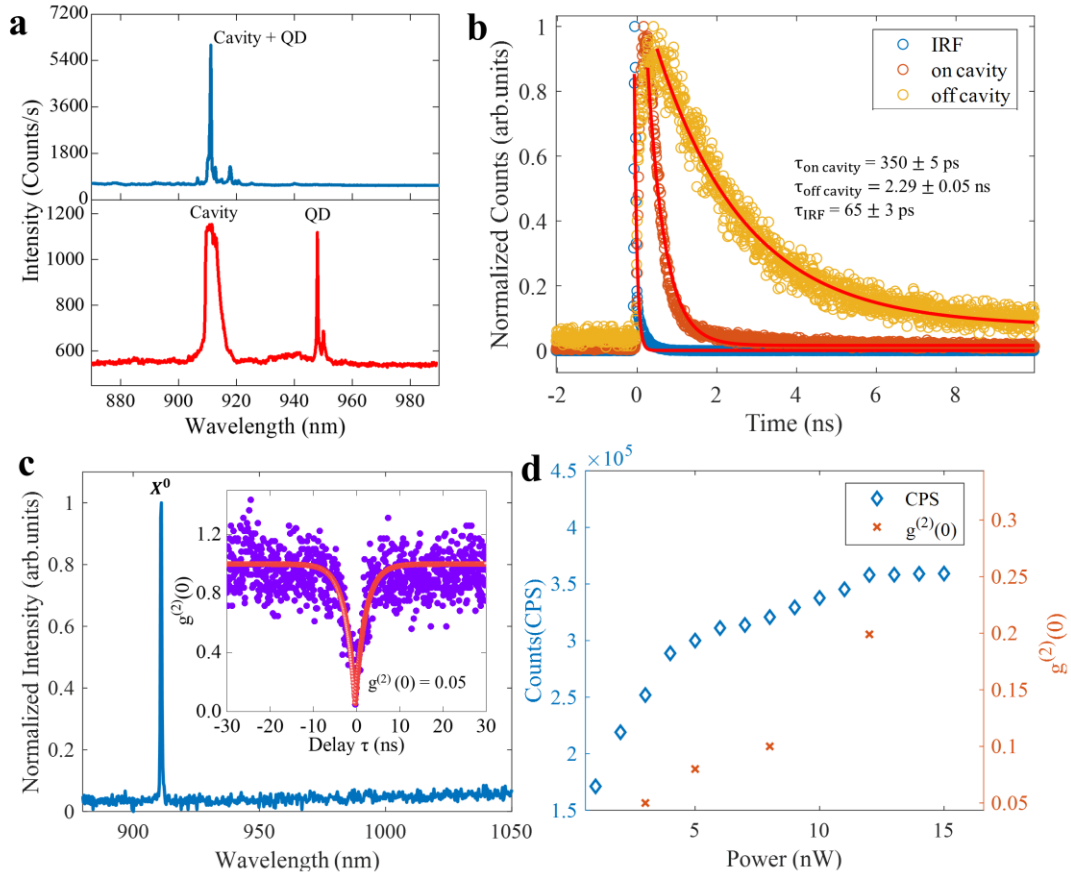


Figure 4.13. Single photon performance measurement. (a) PL spectra measured from two QD-in-NW samples, where blue line corresponds to QD spectrally coupled to the NW cavity, while the red line represents QD not coupled to the cavity. (b) Time-resolved PL decay dynamics for the coupled and uncoupled QD emission are presented, along with the instrument response function (IRF). The decay curves are fitted using single-exponential functions, shown as red solid lines. (c) PL spectrum of the excitonic state of the coupled QD (blue spectrum in (a)) after suppressing background signals using a set of optical filters. The inset displays the second-order autocorrelation measurement, yielding a fitted value $g^{(2)}(0) = 0.05$, confirming single emission. (d) Power dependent measurement of photon count rates and $g^{(2)}(0)$.

Previous study has reported that QDs incorporated in NWs have pronounced Purcell enhancement with NW radius above 180 nm [240]. Two different types of QD-in-NWs PL spectra were observed as shown in Figure 4.13(a). The blue (upper) spectrum shows a strong and narrow peak at ~912 nm which is superimposed on the lower intensity emission from the InP NW, whilst the red (lower) spectrum clearly shows a QD emission peak (at ~948 nm) quite separated from the InP NW emission (at ~912 nm). Furthermore, the intensity of the blue spectrum is substantially higher than that of the red spectrum. The differences in the spectra

can be understood in terms of the coupling between the QD emission and the dominant guided mode of the NW. To further confirm this interaction between QD emission and the NW cavity mode, time-resolved PL measurements were carried out to compare the carrier dynamics in the two samples. The photon decay data in Figure 4.13(b) reveals a significantly shorter carrier lifetime in the sample where the QD emission is coupled to the NW cavity (350 ± 5 ps) compared to the sample where the QD and NW cavity emissions are not coupled (2.29 ± 0.05 ns). This pronounced difference in lifetime originates from the enhanced radiative recombination rate enabled by the resonant coupling between the QD and the cavity mode ^[96]. Consequently, the coupled system exhibits both enhanced emission intensity and accelerated recombination dynamics. The single-photon emission characteristics of the coupled QD-in-NW system were investigated using a Hanbury–Brown and Twiss set-up under continuous-wave excitation at 675 nm. The QD emission was spectrally isolated using a combination of bandpass and long-pass optical filters as shown in Figure 4.13(c). Even without the use of a high-resolution grating for narrow spectral selection, a low $g^{(2)}(0)$ value of 0.05 was obtained at an excitation power, confirming the high single photon purity of our QD-in-NW structure. With increasing excitation power, the photon count rate increases, while the single-photon purity gradually degrades, as shown in Figure 4.13(d). Nevertheless, the emission begins to saturate beyond approximately 10 nW excitation power. Even at saturation, a relatively low $g^{(2)}(0)$ value of 0.19 is still maintained. This performance can be further improved by employing alternative excitation schemes, such as above-band pulsed excitation ^[241], p-shell resonance excitation ^[125], and s-shell resonance excitation ^[118], in combination with higher-resolution spectral filtering techniques.

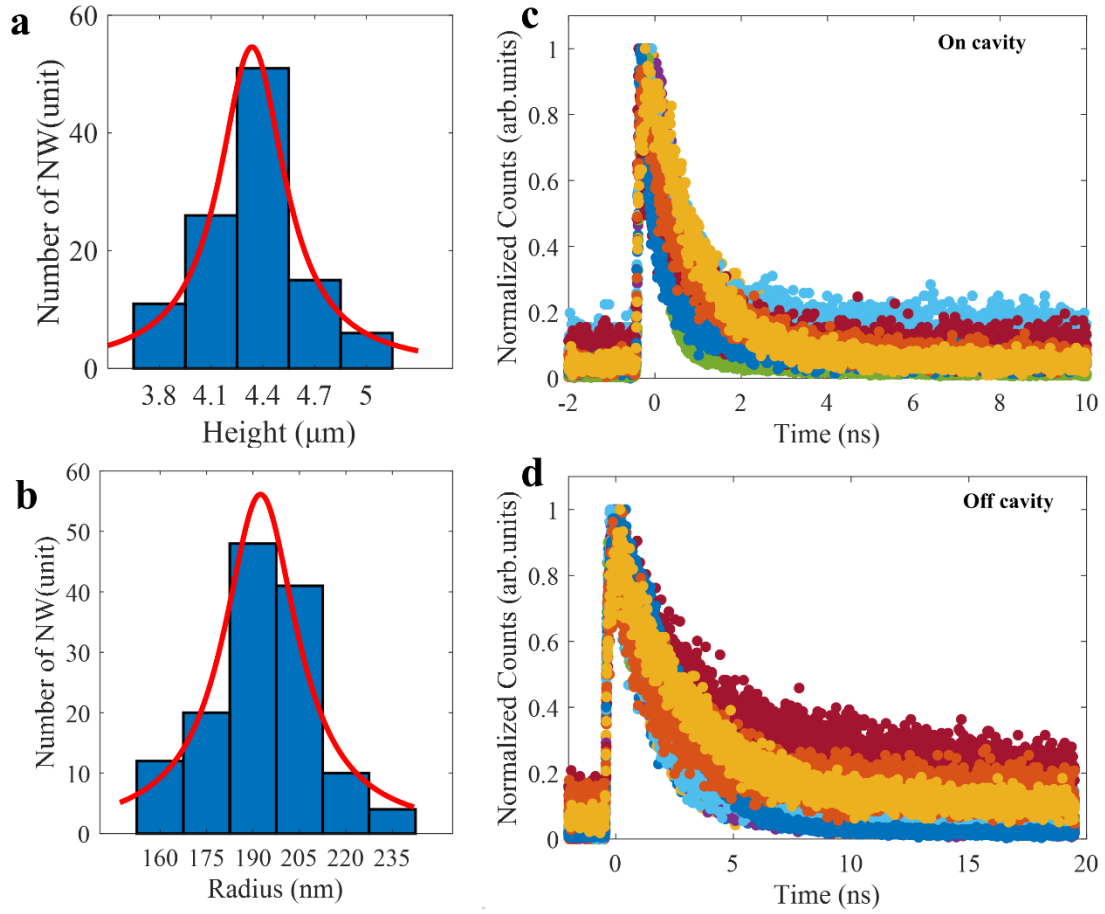


Figure 4.14. Statistical distributions of the (a) NW radius and (b) NW height. Time-resolved data of 10 on-cavity QDs (c) and 10 off-cavity QD-in-NWs (d).

To evaluate the uniformity of the NWs, we analysed the size distribution of 150 NWs by measuring their height and radius. Statistical measurements of our NW arrays indicate the average height Figure 4.14 (a) and radius Figure 4.14 (b) of the NWs are 4.4 μm (FWHM of 0.47 μm) and 192 nm (FWHM of 14.4 nm), respectively. The fluctuations in NW dimensions and QD emission wavelengths result in some QDs being off-resonant and thus decoupled from the NW cavity modes. We study the lifetime difference by measuring the lifetimes from 10 QDs with their emissions coupled and uncoupled into the NW cavities which give average lifetimes of 790 ps and 2.33 ns, respectively.

4.7. FDTD simulation

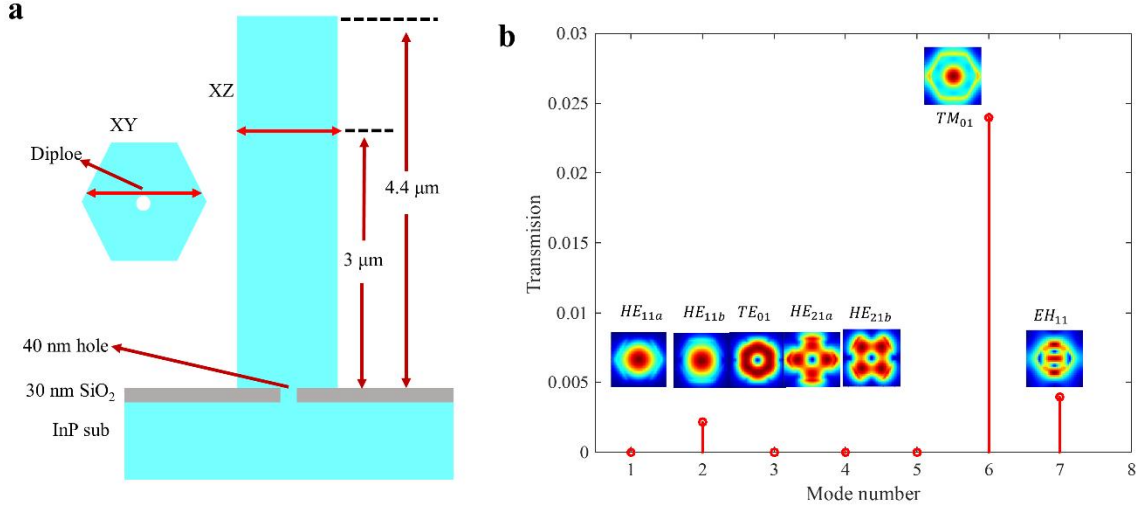


Figure 4.15. FDTD simulation. (a) Schematics of the simulated QD-in-NW structure showing the XY and XZ planes. (b) Simulated transmission power of the first 7 modes of a 380 nm diameter InP NW at the wavelength of 870 nm and corresponding electric field intensity profiles.

FDTD simulation is used to investigate which NW mode is coupled to the QD emission and to study the Purcell enhancement effect of NWs with different sizes. FDTD simulations were performed using the Lumerical 3D Electromagnetic Simulator commercial solver package. A perfectly matched layer was placed around the structure with a distance of at least half a wavelength distance of the NW boundary. Figure 4.15(a) shows the design of the simulation structure based on our experimental results, with a 30 nm SiO₂ growth mask layer placed on the InP substrate, a mask opening size of 40 nm, NW with a height of 4.4 μm and a diameter of 380 nm. A broadband dipole source with a bandwidth spanning from 800 to 1250 nm was placed at a distance of 3 μm from the bottom and the edge of the mask opening in the XY plane.

The Purcell factor is calculated by equation 4.1,

$$F_P = \frac{3Q(\lambda/n)^3}{4\pi^2 V_e} \quad (4.1)$$

Here, the V_e is the coupling mode volume calculated using,

$$V = \frac{\int_V \varepsilon(r) |E(r)|^2 d^3r}{\max(\varepsilon(r) |E(r)|^2)} \quad (4.2)$$

where $\varepsilon(r)$ is the material permittivity at position of r . A time monitor is set up in the middle of the NW to start recording the field after 50 fs then the transient fields have decayed so that only resonant field of the structure remain. By plotting the simulated spectrum result from the time monitor, we can find the resonant frequency over the broadband range. NW is a relatively low Q cavity as the field decay completely from the simulation in a time that can be simulated reasonably by FDTD. Thus, the quality factor can be determined from the Fourier transform of the field by finding the resonance frequencies of the signal and measuring the full width half maximum of the resonant peaks,

$$Q = \frac{f_R}{FWHM} \quad (4.3)$$

where, the f_R is the resonant frequency.

To confirm the coupling mode, we first investigate the dominant optical mode of the NW by placing 100 dipole sources at random positions along the NW, each emitting at a wavelength of 870 nm (corresponding to InP emission). A mode expansion monitor is positioned at the centre of the NW, and seven major modes are selected to calculate the power contribution of each mode. Figure 4.15(b) shows the optical power distribution in the NW cavity among seven major waveguide modes, illustrating the dominant mode of NW is TM_{01} with the highest transmission power. In the simulation of a single QD coupled to the NW cavity, a frequency-domain field profile monitor is positioned at the same location as the mode expansion monitor. The simulation frequency is set to a single point corresponding to the QD emission wavelength of 912 nm. We compare the simulated electrical field profile at 912 nm with the modes in figure 4.15 (b) to check the final result.

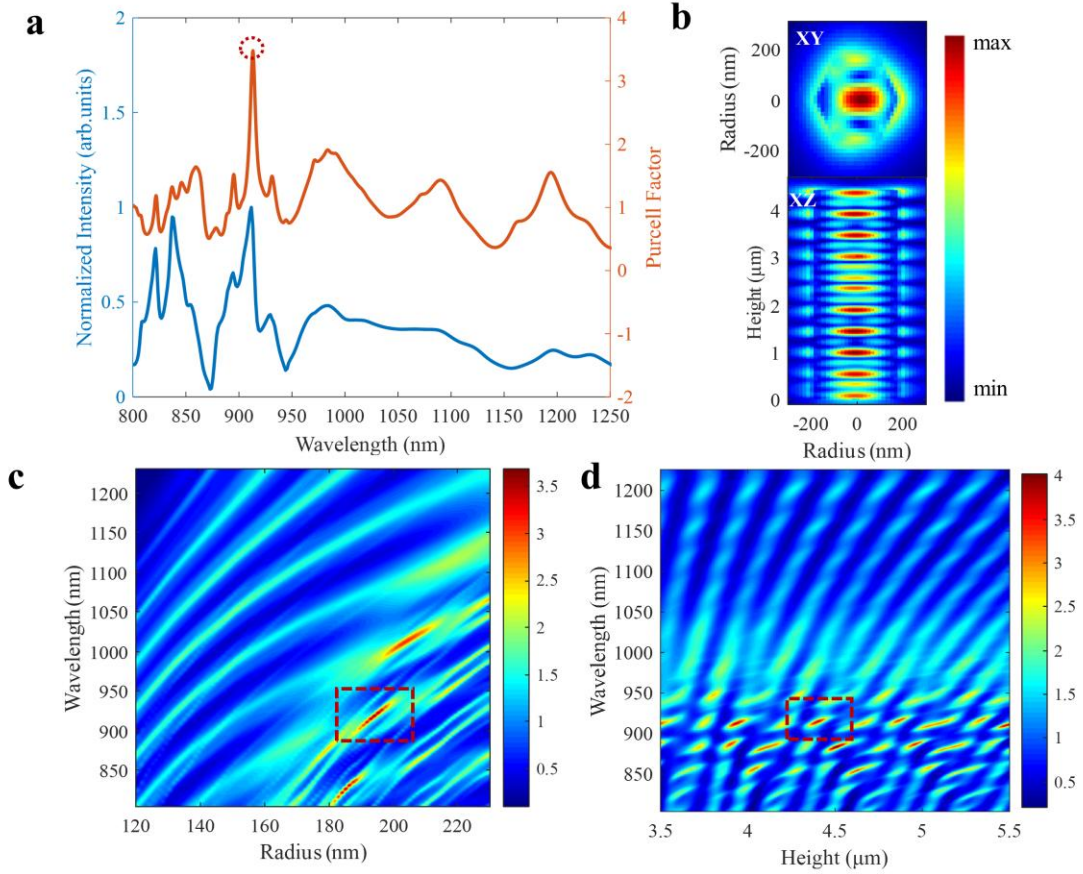


Figure 4.16. FDTD simulation of Purcell enhancement of NW. (a) Simulated spectrum (blue) and Purcell factor (orange) of a QD-in-NW structure. (b) Electrical field profile on the XY and XZ planes at the wavelength with maximum Purcell factor, indicated by the red circle in (a). (c) Simulated Purcell factor as a function of NW radius and QD wavelength. (d) Simulated Purcell factor as a function of NW height and QD wavelength.

Simulation results of the spectra and Purcell factor of the NW coupled single QD in Figure 4.13 (a) with a height of 4.4 μm and a radius of 192 nm, using a broadband dipole source ranging from 800 to 1250 nm are shown in Figure 4.16(a). Both the spectra and Purcell factor demonstrate a strong peak at around 912 nm. The Purcell factor at this wavelength is 3.5, which is in good agreement with our experimental results. We note that the discrete peaks beside 912 nm are generated by longitudinal Fabry-Perot mode with a spacing of $\Delta\lambda = 17.74 \text{ nm}$ [242]. Figure 4.16(b) has shown the dominant mode of the InP NW is TM_{01} mode. The XY and XZ electrical field profiles at the wavelength of 912 nm in Figure 4.17 (b) confirm the QD emission is coupled into the TM_{01} mode of the NW. Figure 4.16 (c) shows the Purcell factor as a function of QD wavelength and NW radius by sweeping across NW radii and heights ranging from 120

to 230 nm and 3.5 to 5.5 μm , respectively. A distinct feature of this plot is that the Purcell factor is maximum for a combination of wavelengths and radius. In general, larger QDs or higher As composition in the QDs (longer emission wavelength) requires a NW with a larger diameter to ensure coupling between QD emission and NW mode. Figure 4.16(d) shows the map of the Purcell factor as a function of QD wavelength and NW height. This provides design flexibility, allowing NWs with different lengths to exhibit similar Purcell factors. The regions highlighted by the two red rectangles in Figures 4.16(c) and (d) correspond to the range of NW radius and length used in this work, where the Purcell factor varies from 2 to 3.9. Statistical measurements of the NW height and radius in Figure 4.14 (a-b) indicate that most QDs are coupled to the dominant optical mode of the NW.

4.8. Summary

In this chapter, we demonstrate a technique that can potentially realise scalable bottom-up single photon sources by employing droplet epitaxy of InAsP QDs on InP NW arrays. We systematically investigate droplet density on the NWs by changing NW diameter, height, pitch size and TMIn deposition time. The size of the droplet (QD) can be tuned by changing the diameter and pitch size of NW, TMIn deposition time, and NW height. For better cavity formation, we also optimise the InP shell growth condition by systematically investigating the effect of V/III ratio, pitch size, and growth temperature. Our technique allows us to deterministically grow one QD at the tip of each NW with a uniform morphology across the entire array. The QDs exhibit superior optical performance with high brightness of millions counts per second by 1 nW excitation power, and a narrow wavelength distribution with a FWHM of 20 nm. By coupling the QD emissions into the mode of the NWs, we achieve a short radiative lifetime of 350 ± 5 ps, a single-photon purity of $g^{(2)}(0) = 0.05$. FDTD is performed to simulate Purcell factor and the dominant mode of the NW for the QD to couple with, showing a simulated Purcell factor of 3.5 and the QD is coupled to the TM_{01} mode of the NW. Compared with conventional planar grown self-assembled QDs, where the emitter position is intrinsically random, this approach enables deterministic positioning of individual QDs through selective-area nanowire growth. It also directly integrates the QD with an optical cavity. The demonstrated brightness and Purcell enhancement are promising for bottom-up single-photon sources. However, further improvements in photon extraction efficiency and indistinguishability are required to compete with state-of-the-art III–V devices. Importantly, this site-controlled growth approach provides a potential route towards large arrays of QD–cavity devices for integrated quantum photonic circuits.

CHAPTER 5

5. InAs Single Quantum Dots in p-i-n InP Nanowires for Electrically Driven Single-Photon Sources

5.1. Introduction

Various single photon emitter systems such as defects in 2D materials ^[45, 243], single molecules ^[244], single atoms ^[245], and colloidal quantum dots ^[246] have been investigated. However, most of them require optical pumping, which limits their applications in practice. In contrast, the electrically driven approach offers a compact, portable and low energy consumption path ^[247]. In this respect, III–V semiconductor QD based single photon sources are attractive not only in their superior optical performance ^[26, 236] as discussed in Chapter 1, but also in their capability to be monolithically integrated into p–i–n diode structures through advanced epitaxial growth techniques such as molecular beam epitaxy and metal–organic chemical vapour deposition ^[248, 249]. Moreover, the compatibility of these materials enables heterogeneous integration with diverse platforms, enabling scalable and compact on-chip quantum photonic circuits ^[77, 89].

However, there are still challenges in using epitaxially-grown QDs as electrically driven single photon sources. Firstly, carrier injection should ensure that electron-hole pair recombination occurs only in the QD and form well-defined exciton states ^[95]. However, electrical injection often suffers leakage currents, background emission, and charge noise ^[250], resulting in the degradation of the purity and stability of the single photon emission. This demands careful doping design, optimisation of the layers, and suppression of charge fluctuation ^[251]. Secondly, bottom-up growth of single QDs in optical cavities offers a highly efficient approach to deterministically position the single emitters in the cavities ^[225], with QD-in-NW ^[92, 252] as an famous example as shown in Chapter 4. Saniya *et al.* have demonstrated the first electrically driven QD-in-NW single-photon source device, by collecting photons from a horizontally lying GaN nanowire diode embedded with an InGaN QD ^[253]. Despite this important milestone, electrically driven single photon emission has not yet been reported in InP NW material system. This gap highlights the challenges in identifying and fabricating contact areas in horizontal nanowire after they are transferred from their growth substrates ^[254], owing to their small

dimensions with lateral size of a few hundred nanometers and axial length of a few microns [255]. In addition, light traveling along the nanowire waveguide limits the collection efficiency from the top. In comparison, fabricating vertically light emitting NW devices can avoid the transferring process and vertical NWs have been presented as high-efficiency single photon sources cavities [256, 257]. Furthermore, InAs QDs embedded in InP material systems can operate at telecom-band wavelengths, ensuring direct compatibility with the existing low-loss, cheap, and reliable telecom optical fibre network for quantum networks [48, 258].

In this chapter, we overcome these challenges by designing a core-shell p-i-n structure with a deterministically grow an InAs single QD in an InP NW diode. The QDs are grown by droplet epitaxy while the NWs are fabricated by SAE as presented in Chapter 4. For the optimisation of the p-doped InP core, a high growth temperature was employed to enable the growth of thin nanowires, while the Zn precursor flow rate was varied to maximise the Zn doping concentration. For the n-doped InP shell optimisation, we investigate the growth temperature to obtain uniform hexagonal NW structure as good optical cavity for light propagation. We confirm this core-shell doping structure by conducting conductive atomic force microscope, where the current map of the NW tip region indicates that electrons and holes are injected laterally. Two devices are fabricated with same NW dimension but different QD wavelength. Current-voltage measurements of these two devices show good rectifying behaviour, characteristic of a p-n junction. One device shows strong QD emission coupled to the NW FP mode, while the off-coupled device exhibits much a weaker QD emission. The coupled QD acts as single photon emission with a measured $g^{(2)}(0)$ value of 0.21 and high brightness as high as a million counts per second under a current injection of 1.5 μA . Moreover, the device shows long-term stability - maintaining the same emission wavelength and photon count rate over an hour. FDTD simulation proves the QD has been coupled to the TM_{01} mode of the NW with a simulated Purcell factor of 4.3. Our bottom-up approach shows great promise for scalable fabrication of electrically driven single photon sources in III-V semiconductor platforms for future development of quantum networks.

5.2. Experimental methods and design

Substrate preparation for selective area epitaxy: 150 nm SiO₂ layer was deposited on p-doped InP (111) A substrates by plasma-enhanced chemical vapour deposition, following by spin coating of 300 nm electron beam resist (ARP 6200.09). The pattern for NW growth was designed with circles of 40 nm diameter, which was then exposed by electron beam lithography (Elionix F125) with a voltage of 125 kV and a current of 2 nA. The pattern was transferred onto the SiO₂ layer after inductively coupled plasma reactive ion etching (ICP-RIE) with CHF₃. The patterned substrates were trim-etched in a mixed solution of 20% H₂O₂, 20% H₃PO₄ and 60% DI water for 2 mins.

MOCVD growth: Epitaxial growth was conducted on the InP substrates with SiO₂ masks using a horizontal flow metalorganic vapour phase epitaxy system (Aixtron 200/4) at a pressure of 100 mbar. Prior to growth, the substrates were thermally deoxidised for 10 mins at the temperature of 750 °C under PH₃ flow. TMIn, PH₃, and AsH₃, were used as precursors for In, P, As, whilst diethylzinc (DEZn) and SiH₄ were used for Zn, and Si doping, respectively. P-doped InP core was first grown at the temperature of 735 °C for 3 mins with PH₃, TMIn and DEZn flow rate of 4.9×10^{-4} , 6.07×10^{-6} , and 9.6×10^{-6} mol/min, respectively, corresponding to a V/III ratio of 81. The substrate was then cooled to 400 °C under PH₃ protection with the same flow rate and stabilised for 3 minutes. PH₃ was then removed and the chamber purged for 30 s to remove any residual group V source, after which TMIn at a flow rate of 2×10^{-6} mol/min was introduced for a fixed amount of time (0.1 and 0.2 s). The substrate was cooled down immediately to investigate the droplet formation. For TEM measurement, the droplets were then crystallised into QDs at 520 °C with an AsH₃ flow rate of 5×10^{-4} mol/min, and then cooled down under the protection of AsH₃ until the temperature was below 350 °C. For the complete device, the QDs were capped by a layer of undoped InP grown at the temperature of 520 °C for 1.5 mins, with a V/III ratio of 2000 using TMIn and PH₃ flow rates of 2.99×10^{-6} and 5.98×10^{-3} mol/min, respectively. Then the substrate temperature was increased to 620 °C and stabilised for 3 mins under PH₃ flow. Finally, an n-doped InP shell layer was grown for 2 mins with a V/III ratio of ~800, corresponding to TMIn, PH₃, and SiH₄ flow rates of 6.07×10^{-6} , 4.91×10^{-3} , and 1.7×10^{-6} mol/min, respectively.

Material characterisation: The NW morphology and In droplet size were characterised by an FEI Verios 460 SEM system operated at a voltage of 5 kV and a current of 100 pA. The NW conductivity was characterised by conductive AFM (Bruker, Dimension Icon) under contact

mode. Structural and compositional analyses were performed by an aberration-corrected scanning TEM system (JEOL 2100F FEGTEM) equipped with an energy dispersive X-ray spectroscopy system, operating at a voltage of 200 kV, and a current of 131 μA .

Device characterisation: The current-voltage (I - V) characteristics were measured with a Keithley 2612 B source unit. Experimental set-up of luminescence measurement is schematically illustrated in Figure Chapter 3.5.3.

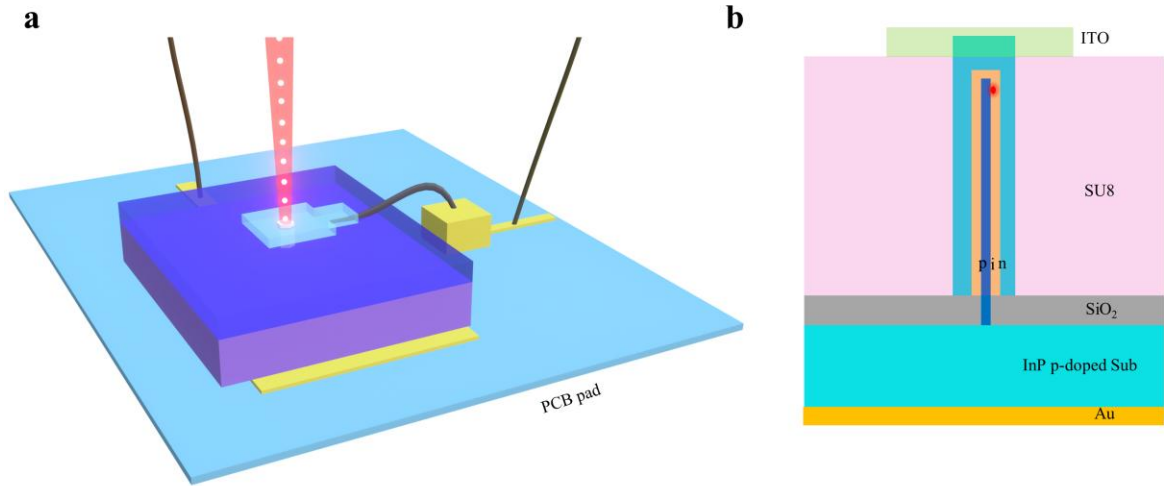


Figure 5.1. Device packaging and design. (a) Device packaging by bonding the NW diode with PCB pad for electrically driven characterisation. (b) Design of single NW device: a cross-sectional view of a core-shell p-i-n InP NW diode with a single InAs QD embedded in the intrinsic region. SiO_2 is used as the mask layer, SU-8 as the planarisation layer, Au and ITO serve as the back and top contacts, respectively.

Electrically driven QD-in-NW single photon devices in the literature are all based on horizontal lying NWs^[253, 259], which have several challenges. First, individual NWs need to be transferred on another substrate and painstakingly identified to make the electrical contacts. This is time consuming and requires electron beam lithography^[260, 261] to fabricate contacts to both ends of the NW (for an axial p-i-n structure). Furthermore, light emitted from the NW is scattered by the underlying substrate resulting in poor collection efficiency. Therefore, we design vertically emitting devices with the NWs standing on their growth substrate. The design also allows us to use simple lithography to make the contacts on both bottom and top region of the NW as shown in Figure 5.1 (a). Detail structure of the device as illustrated in Figure 5.1 (b), a core-shell p-i-n nanowire with the QD embedded in the i-region of the NW. A p-doped InP NW core was first

grown by selective area epitaxy on a (111)A p⁺ InP substrate, followed by the growth of a single InAs QD (red circle). The QD was grown by droplet epitaxy as reported previously [262]. An intrinsic (i-layer) InP shell was then deposited, and finally an n-doped InP shell layer was grown. Au was deposited as the back contact, while indium tin oxide (ITO) was used as the electron-selective top contact after a planarisation and etch-back step. Details of the device fabrication processes have been discussed in Chapter 3.3.

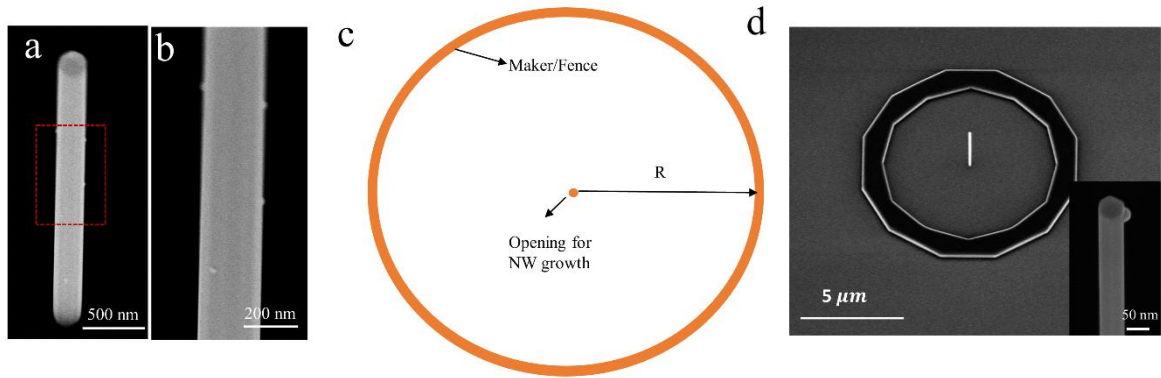


Figure 5.2. (a) SAE growth of single undoped InP NW with 0.1 s TMI deposition time at a flow rate of $2 \times 10^{-6} \text{ mol/min}$ and at a temperature of 400 °C. NW with much larger lateral growth and multiple droplets on the sidewall of the NW as seen in the zoom in SEM image of the red rectangular region from (a) in (b). (c) Pattern design to grow single QD-in-NW diodes: the orange ring with a radius of R as the maker for the single NW (orange circle in the centre), and also acts as a fence to control the diffusion rate of the adatoms. (d) Growth results of the In droplet on undoped single NW with the pattern in (b).

In chapter 4.3.3, we have demonstrated that on $\sim 3 \mu\text{m}$ high NWs in a $3 \mu\text{m}$ - pitch array, only one droplet is formed on each NW. The core NW growth condition was with a V/III ratio of 81 at a temperature of 740 °C for 3 mins, and the single droplet formation was achieved with 0.25 s TMI deposition time at a flow rate of $2 \times 10^{-6} \text{ mol/min}$ and at a temperature of 400 °C. However, the fabrication of electrically driven QD-in-NW arrays remains challenging, as it is difficult to ensure uniformity among all NWs due to small fluctuations in precursor flow rates, leading to variations in resistance between NWs. This, in turn, hinders stable current injection. Therefore, optimisation of single QD-in-NW diodes is necessary. We first applied the same growth parameters to deposit droplets on single NWs, pronounced lateral growth and multiple

droplet formations on the sidewalls were observed, as shown in the SEM images in Figure 5.2(a-b). This effect is mainly attributed to the higher diffusion of adatoms toward the single opening on the bare substrate. To address this issue, we introduced a circular ring fence with a radius R surrounding the small central opening for NW growth, as illustrated in Figure 5.2(c). This ring opening not only limits the adatom diffusion rate and simulates the environment of array growth, but also serves as a marker for locating the single NW during the device fabrication process. We repeated the growth using the new pattern design. The SEM image in Figure 5.2(d) shows that both the InP micro-ring and the NW exhibit highly uniform structures, with minimal lateral growth of the NW, as shown in the inset. A single In droplet is also observed on the tip of the NW. With the aid of the ring marker, we successfully reproduced the growth parameters for depositing single droplets on individual NWs.

5.3. InP NW core-shell structure quality engineering

The parameters for droplet formation and the i-layer shell growth are the almost the same as optically pump samples in Chapter 4. Thus, from the growth part in this section, we mainly discuss the growth parameter for the p-doped InP core as well as the n-doped InP shell, aiming to obtain a good optical cavity for light propagation.

5.3.1 p-doped NW core

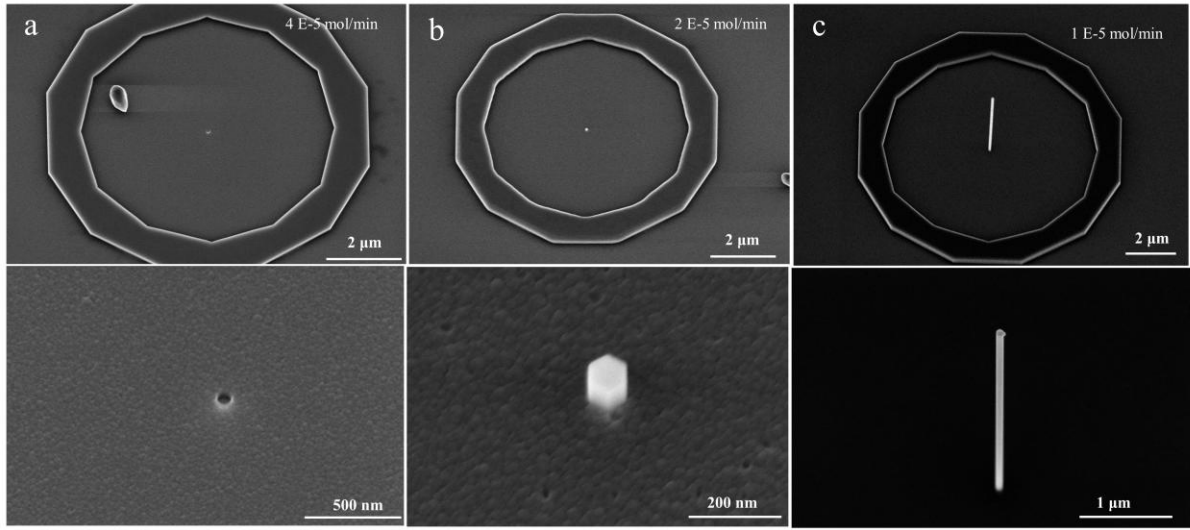


Figure 5.3. SEM images of the p-doped NW core growth with a V/III ratio of 81 and a temperature of 730 °C in different DEZn doping flow rate and growth time: (a) $4 \times 10^{-5} \text{ mol/min}$ and 20 mins, (b) $2 \times 10^{-5} \text{ mol/min}$ and 10 mins, (c) $1 \times 10^{-5} \text{ mol/min}$ and 4 mins. The below SEM images are the zoom-in image of the above ones.

DEZn (Diethylzinc, $\text{Zn}(\text{C}_2\text{H}_5)_2$) is a common p-type dopant precursor in III–V semiconductor growth by MOCVD. It supplies Zn adatoms which substitute of In, introducing acceptor levels and thereby providing holes in the valence band ^[263, 264]. However, Zn adatoms exhibits a tendency toward doping saturation in InP and possesses a high diffusion coefficient ^[265], especially at high temperature. But the core NW is based on selective area epitaxy which usually requires high temperature growth. To achieve higher Zn doping at the growth temperature of 730 °C, we used high DEZn flow rates to obtain p-doped core NWs. The SEM images in Figure

5.3 show the results of p-doped NW growth under a V/III ratio of 81 and a temperature of 730 °C with different DEZn flow rates and growth times. At a DEZn flow rate of 4×10^{-5} mol/min (Figure 5.3a), no NWs emerged from the openings even after a 20 min growth. This high DEZn flow rate suppresses the diffusion of In and P adatoms, thus preventing NW growth. When the flow rate was reduced to 2×10^{-5} mol/min (Figure 5.3b), short NWs (~ 100 nm) were obtained after 10 min of growth. However, the growth rate was still too slow to reach a few microns in height. Further decreasing the flow rate to 1×10^{-5} mol/min (Figure 5.3c) resulted in the NWs with a height of ~ 3 μm and a diameter of ~ 60 nm. A small droplet on the tip region was formed by introducing TMIn at a flow rate of 2×10^{-6} mol/min for 0.1 s at a deposition temperature of 400 °C. With the aid of the ring marker, we were able to deterministically position a single droplet on a p-doped NW. It is worth noting that the growth temperature used here (730 °C) is higher than typical planar growth conditions (~ 650 °C), under which similar DEZn flow rates can result in Zn doping concentrations on the order of 10^{18} cm^{-3} [266]. However, Zn incorporation in InP nanowires is known to decrease significantly at growth temperatures above ~ 700 °C [267]. Therefore, the actual Zn concentration in the nanowires is expected to be lower than that estimated from planar growth calibrations.

5.3.2 n-doped NW shell

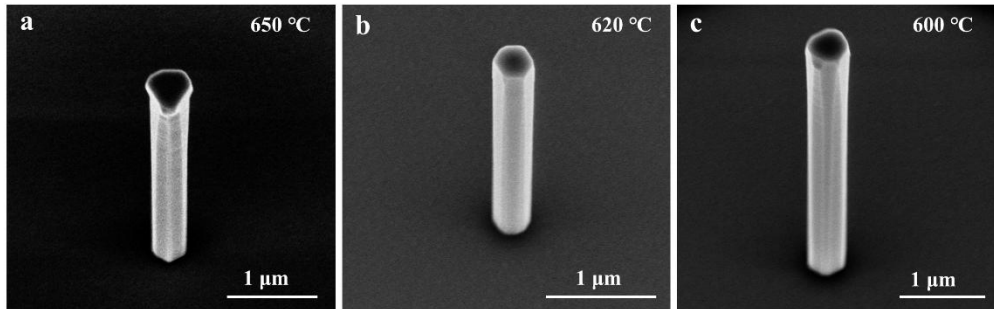


Figure 5.4 SEM images of the InP NW diode after the final capping of the QD with an n-doped shell layer for 2 mins with a V/III ratio of ~ 800 , and a SiH_4 flow rates of 1.7×10^{-6} mol/min at different growth temperatures: (a) 650 °C, (b) 620 °C, and (c) 600 °C.

We optimised the n-doped InP shell growth by varying the growth temperature, as shown in Figure 5.4. The SEM images illustrate the evolution of the InP shell morphology as a function of growth temperature. Silicon is widely used as an n-type dopant in InP due to its relatively high incorporation efficiency and its ability to provide shallow donor levels ^[268, 269], enabling effective electron activation at room temperature. The n-doped InP NWs were grown with a V/III ratio of ~ 800 and a SiH_4 flow rate of 1.7×10^{-6} mol/min for 2 min. Based on calibration from planar InP growth under similar conditions, this SiH_4 flow rate corresponds to an electron concentration on the order of $\sim 10^{18} \text{ cm}^{-3}$. It should be noted that, in nanowire structures, the effective doping concentration may deviate from planar calibration due to enhanced surface effects and differences in incorporation kinetics. Nevertheless, the chosen SiH_4 flow provides a reliable reference point for achieving moderate n-type doping in InP nanowires. From Figure 5.4, it can be observed that the growth temperature of 620°C results in the best NW morphology, which is consistent with that of the undoped NWs discussed in Section 4.5.3. This indicates that the introduction of Si does not significantly affect the growth of the InP shell.

5.4. Device characterisation

In this section, we first investigate the performance of optically pumped quantum dots to determine the optimal NW diameter for achieving Purcell enhancement. Next, conductive atomic force microscopy is employed to examine the electron–hole recombination region. Finally, electrically driven devices are fabricated, and the optical performance of single QDs is systematically studied.

5.4.1. PL of single NW

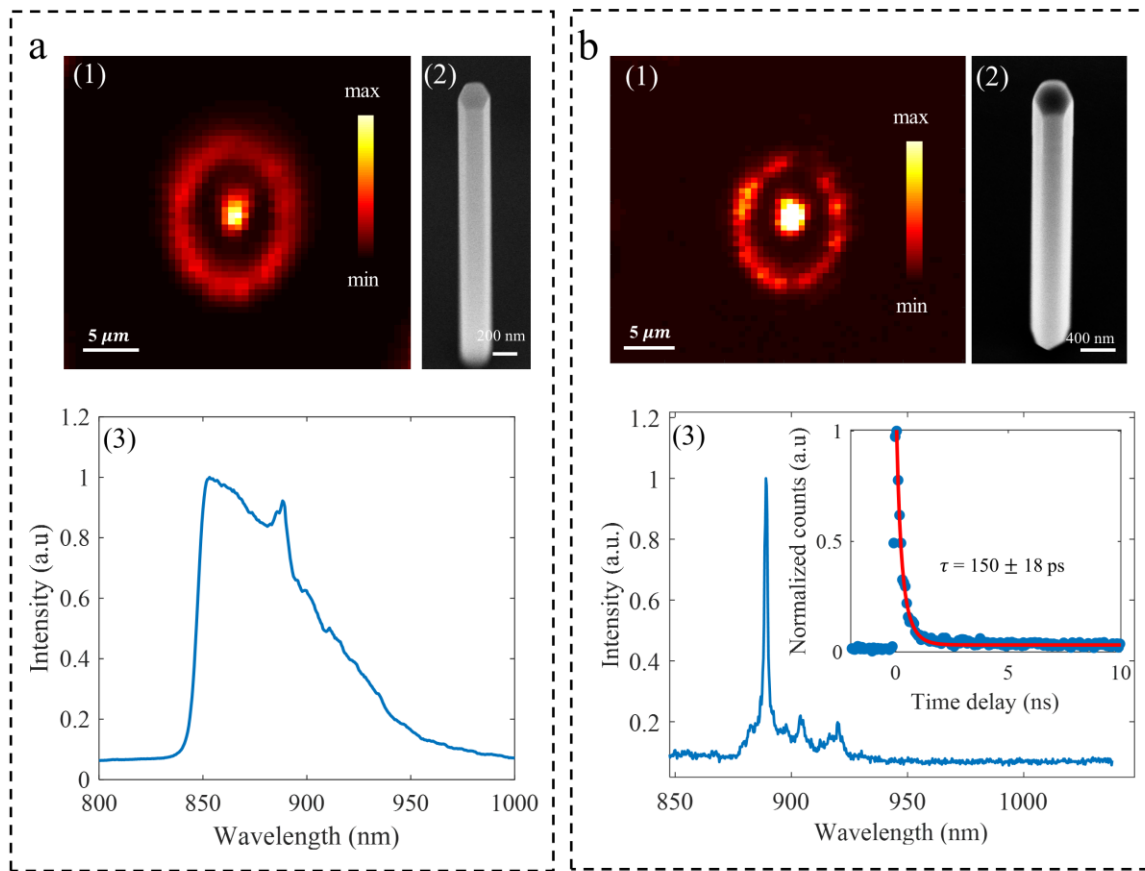


Figure 5.5. Optical excitation of the QD-in-NW diodes with different NW diameters. (a) 200 nm, (b) 410 nm. (a-1) and (b-1) are the PL intensity maps under 10 nW of 780 nm CW laser excitation at 5 K. (a-2) and (b-2) are the corresponding SEM images. (a-3) and (b-3) are the corresponding PL spectra excited under 20 nW excitation, the inset is the time-resolved trace of the 887 nm peak, showing a lifetime of 150 ± 18 ps.

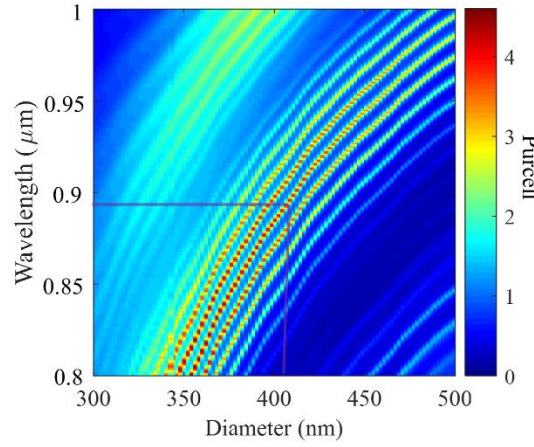


Figure 5.6. Simulated map of Purcell factor as a function of NW radius and QD emission wavelength.

Prior to device fabrication, micro-photoluminescence spectroscopy was carried out at 5 K using a 780 nm CW laser to identify the QD emission wavelength and to investigate the coupling effect. Figure 5.5(a-1) is the PL map under 10 nW excitation from the NW shown in the SEM image in Figure 5.5(a-2). Figure 5.5(a-3) presents the corresponding spectrum of the device, in which a sharp emission peak at 887 nm, which can be attributed to the QD, appears over a broad background emission originating from the doped InP NW. The latter is due to doping-induced bandgap modification, which leads to broader emission compared to an undoped InP NW. In addition, the small diameter of the NW could not support the mode at ~ 887 nm for coupling the QD emission, resulting in a low signal to noise ratio of the QD emission. When we increase the diameter of the NW to 410 nm in (b), brighter emission from the QD is observed. From the PL map in Figure 5.5(b-1), under the same excitation power, there is much brighter emission. Moreover, Figure 5.5 (b-3) shows a sharp and strong peak at ~ 887 nm representing the QD emission, while the much smaller regular peaks indicate the modes of the NW being excited. The inset shows a fast decay rate of the main peak at 150 ± 18 ps, which confirms the coupling of the QD emission with the NW cavity mode.

To understand to optical behaviour difference between the 200 nm and 410 nm diameter NWs, FDTD simulation is performed to simulate the Purcell factor as a function of nanowire diameter and emission wavelength as shown in Figure 5.6. The simulation results in Figure 5.6 show

that the highest Purcell factor at 887 nm can be achieved from a NW with a diameter of 410 nm, which explains the stronger QD emission from Figure 5.5 (b). Note that the regular peaks correspond to the Fabry–Pérot modes of the NW, which can be calculated by ^[270]:

$$\Delta\lambda = \left(\frac{\lambda^2}{2L}\right) \left(n - \lambda\left(\frac{dn}{d\lambda}\right)\right)^{-1} \quad (5.1)$$

where L is the NW height, n is the refractive index at wavelength of λ , and $n - \lambda\left(\frac{dn}{d\lambda}\right)$ is group refractive index.

5.4.2. Conductive atomic force microscopy

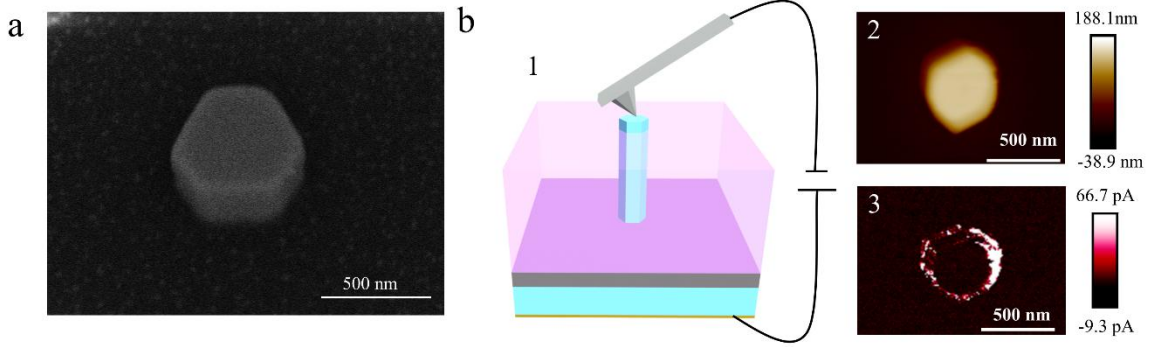


Figure 5.7. (a) SEM Image showing the sample after planarisation and etch-back for conductive AFM measurement. (b) Conductive-AFM measurement of the device: (1) Schematic diagram illustrating the measurement setup; (2) AFM image showing the height profile and (3) the corresponding current map.

To further confirm our core-shell p-i-n structure and lateral carrier injection, we carried out conductive atomic force microscopy (C-AFM) on the single NW diode after the planarisation and etch-back step. The SEM image in Figure 5.7 (a) shows the NW for C-AFM measurement after etch-back. The measurement schematic is shown in Figure 5.7(b-1), where a conductive AFM tip scans across the top facet of the NW while serving as a local top electrode, and the Au

back contact on the substrate is connected to the bottom electrode, forming a complete electrical circuit. This configuration enables simultaneous acquisition of topography and local electrical transport characteristics. Figure 5.7(b-2) shows the AFM image of the nanowire tip with a height of 227 nm after planarisation and etch-back. The corresponding current map shown in Figure 5.7(b-3) indicates a bright region along the periphery of the nanowire, attributed to a lower resistance path with higher conductivity, indicating preferential lateral carrier injection and resulting in higher current levels in this region.

5.4.3. Current-voltage measurement

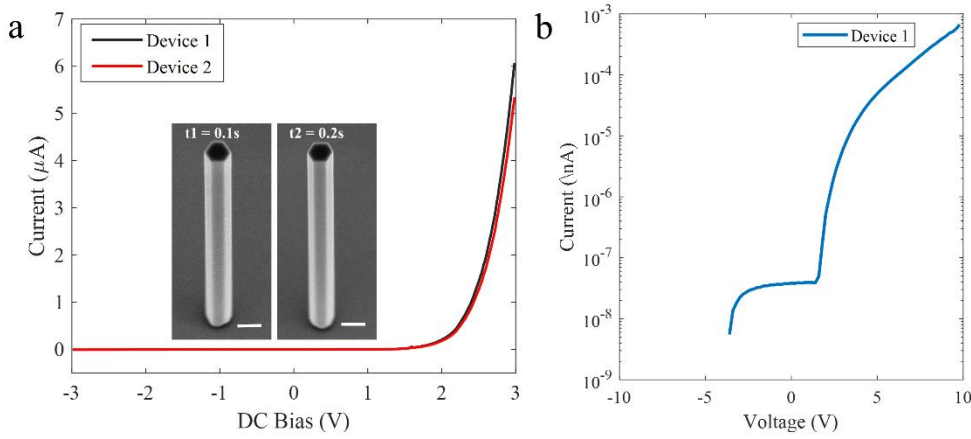


Figure 5.8. (a) I-V characteristics of device 1 and device 2. Insets show the corresponding SEM images of the two devices. (b) I-V plot of device 1 from -10 to +10 V, showing a rectification ratio of $\sim 1 \times 10^5$.

To investigate the coupling of the QD emission to the NW optical cavity modes, we fabricated two devices with different QD emission wavelengths by tuning the droplet deposition time (device 1: 0.1 s; device 2: 0.2 s), while keeping the NW dimensions identical. The low-temperature (5 K) I-V characteristics of both devices, measured from -3 to $+3$ V, are shown in Figure 5.8(a). During the measurement, the source meter was configured to operate in voltage-source mode, where a sweep of bias voltage was applied across the device while the corresponding current was recorded. The bias was typically swept from -3 V to $+3$ V with 301

data points, and the resulting current was measured with picoampere-level sensitivity. A compliance current limit of 1 mA was set to prevent device damage due to overcurrent, particularly important for fragile single NW-based structures. Measurement timing and integration settings were optimised to reduce noise while maintaining measurement speed. Both devices exhibit typical diode-like rectifying behaviour, with negligible current under reverse bias and a turn-on voltage of ~ 2 V. The low leakage current in the reverse-bias regime indicates that the p-i-n junction in the nanowires is well-formed. The near overlap of the two I–V curves further confirm the reproducibility of the epitaxial growth and device fabrication processes. The insets in Figure 5.8(a) show the SEM images of the two devices, revealing uniform nanowire morphology with comparable dimensions of ~ 6 μm in height and ~ 410 nm in diameter. A rectification ratio of $\sim 1 \times 10^5$ at ± 10 V of device 1 can be seen in Figure 5.8(b).

5.4.4. Electroluminescence

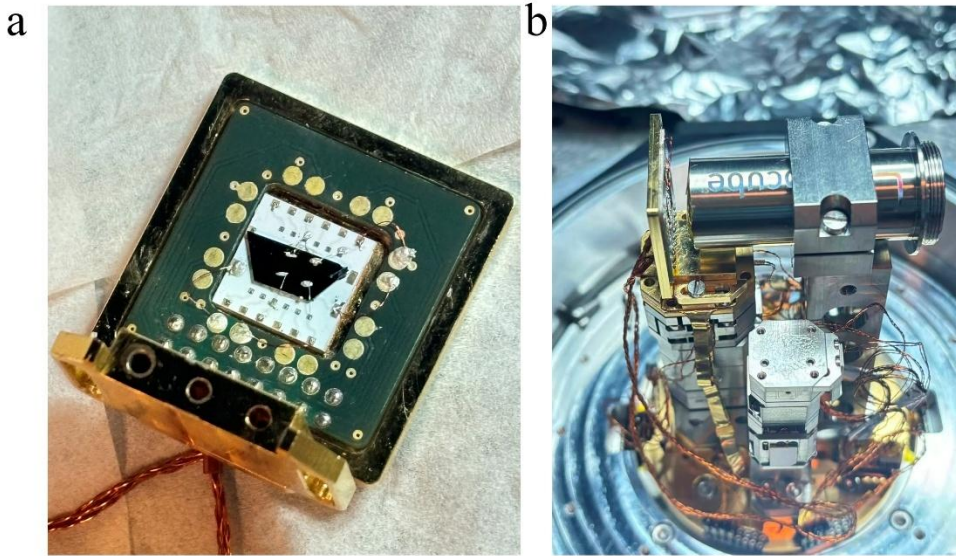


Figure 5.9. Integration of the device with the cryostat for low-temperature, electrical characterisation. (a) The fabricated devices on the white PCB pad, wire-bonded with conductive wires, are mounted onto the L-shaped copper holder (with electrodes indicated by yellow circles) using silver paste. (b) The assembled PCB pad is subsequently connected to the cryostat, where the electrodes are connected to the internal pins that feed through to external terminals for BNC connections to the current source.

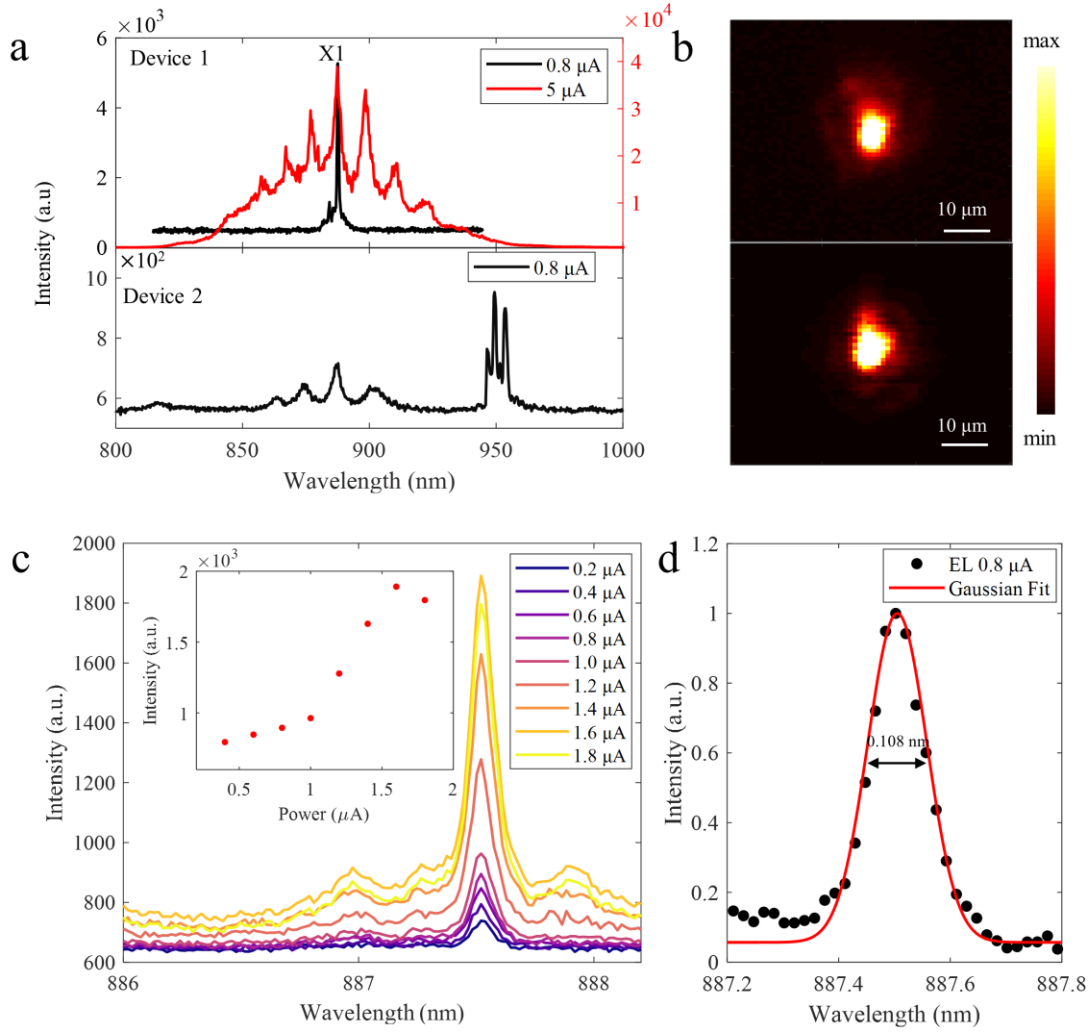


Figure 5.10. (a) EL spectra of the two devices: black lines correspond to measurements under 0.8 μA , and the red line of Device 1 corresponds to 5 μA . X1 indicates the emission peak from the QD. (b) EL intensity maps at 0.8 μA for both devices. (c) Injection current dependent measurement of X1 at 887.5 nm in Device 1 with 1800 g/mm grating. Insert shows EL intensity plot as a function of injection current. (d) EL spectrum of the X1 emission under 0.8 μA injection current. The Gaussian fit gives a full width at half maximum of 0.108 nm.

EL measurements were performed in a confocal microscopy system at 5 K under continuous current injection (The experimental set-up is schematically shown in Chapter 3.5.3). The

mounting of devices onto a PCB in the cryostat is shown in Figure 5.9. After device fabrication, we first integrate the device to the self-design white printed circuit board (PCB, copper based) pad, and after wire-bonding, we solder the conductive wires for back and top contact of each device. Then the white PCB pad is mounted onto the L-shaped copper holder (with electrodes indicated by yellow circles) using silver paste to fix the wires as seen in Figure 5.9 (a). Finally, the assembled PCB pad is connected to the cryostat (Figure 5.9 (b)), where the electrodes are connected to the internal pins that feed through to external terminals for BNC connections to the current source for electrically driven measurements.

Figure 5.10(a) presents the EL spectra of the two devices. At an injection current of $0.8 \mu\text{A}$, device 1 (black line) exhibits a much stronger QD emission peak at $\sim 887 \text{ nm}$, compared with device 2 where the QD emission is weaker and occurs at $\sim 950 \text{ nm}$ (The peaks in the $860\text{--}900 \text{ nm}$ range are due to emission from the InP NW). The difference in the QD emission peaks can be attributed to the difference in size between the two QDs, whilst the intensity difference is due to the coupling between the QD emission and the nanowire cavity modes in device 1. To further verify this effect, the injection current in device 1 is increased to $5 \mu\text{A}$. Under this condition, the energy levels of the QD become saturated, and additional electron–hole recombination occurs in the InP region, producing a broad emission spanning $\sim 830\text{--}930 \text{ nm}$ (red line). Furthermore, this broadband emission excites the Fabry–Pérot modes of the NW cavity, as evidenced by the distinct periodic peaks ($\Delta\lambda = 11.1 \text{ nm}$) in the spectrum. The overlap between the QD emission and the cavity modes confirms their optical coupling. A similar set of mode peaks is observed in device 2 at low injection current with comparable mode spacing, which further confirms the nearly identical NW dimensions in both devices. The corresponding EL intensity maps of the two devices, obtained under $0.8 \mu\text{A}$ current injection, are shown in the right panel of Figure 5.10(b). The bright emission spot from the top of each nanowire indicates that emission is in the vertical direction. The measurement results of the QD emission (labelled X1) in device 1 under different current injection levels with an 1800 g/mm high resolution grating are shown in Figure 5.10(c). The X1 peak increases in intensity as a function of injection current, with several additional smaller side peaks being excited at higher currents. The inset in Figure 5.10(c) shows that the intensity of X1 increases linearly and saturates at a current of $1.6 \mu\text{A}$. The linewidth of the emission peak at $0.8 \mu\text{A}$, determined from a Gaussian fit of the EL spectrum in the Figure 5.10 (d) is 0.108 nm .

5.4.5 Single photon properties measurement

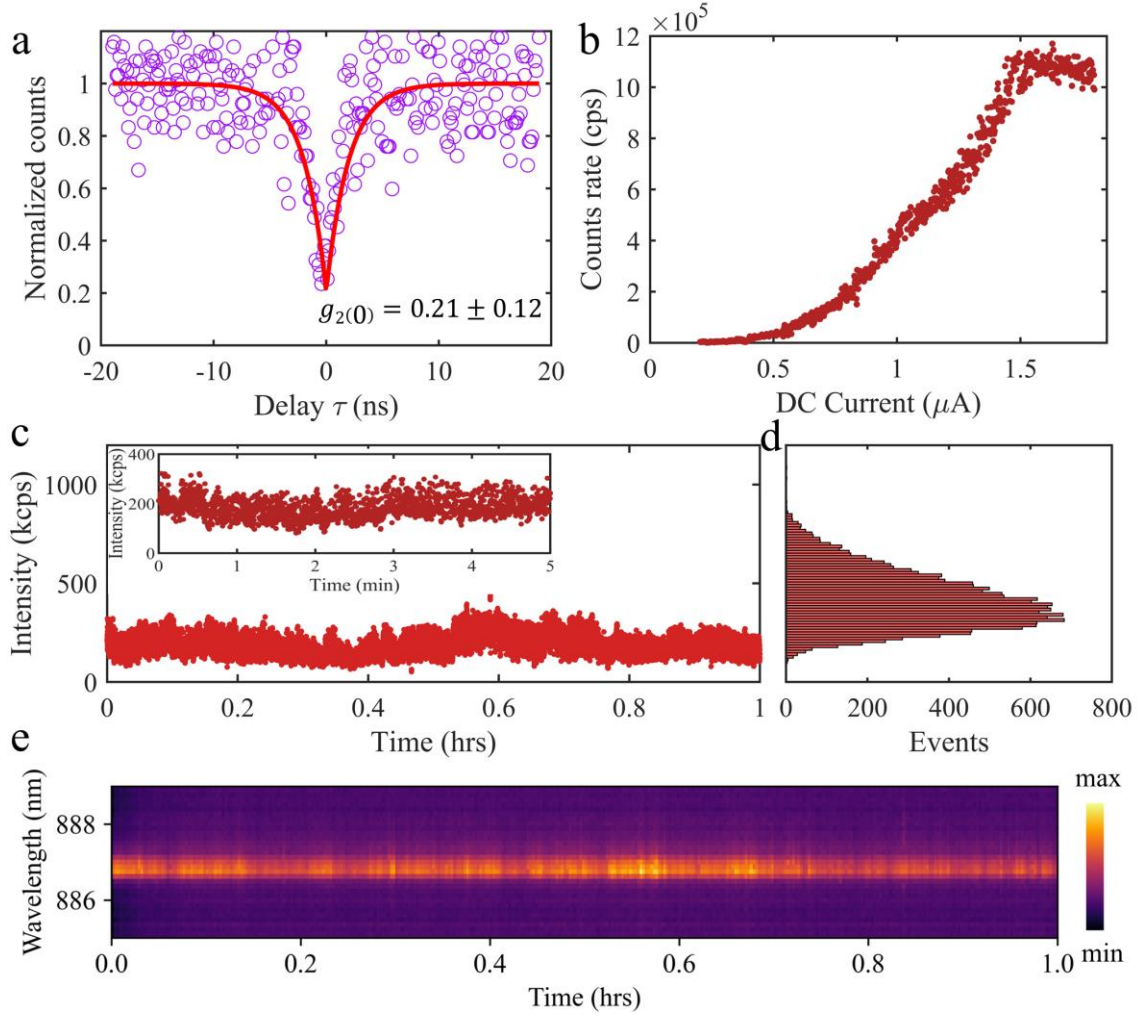


Figure 5.11. Optical characteristics of the QD emission in device 1. (a) Second-order correlation measurement results under an injection current of 0.8 μA , where $g^{(2)}(0) = 0.21$ is obtained from the fitting data. (b) Photon count rate as a function of injection current. (c) Temporal response of the photon count rate over one hour, with the inset showing the intensity in the first 5 minutes. (d) The corresponding histogram of the distribution in (c). (e) Spectral map at 0.8 μA continuous current injection over one-hour duration.

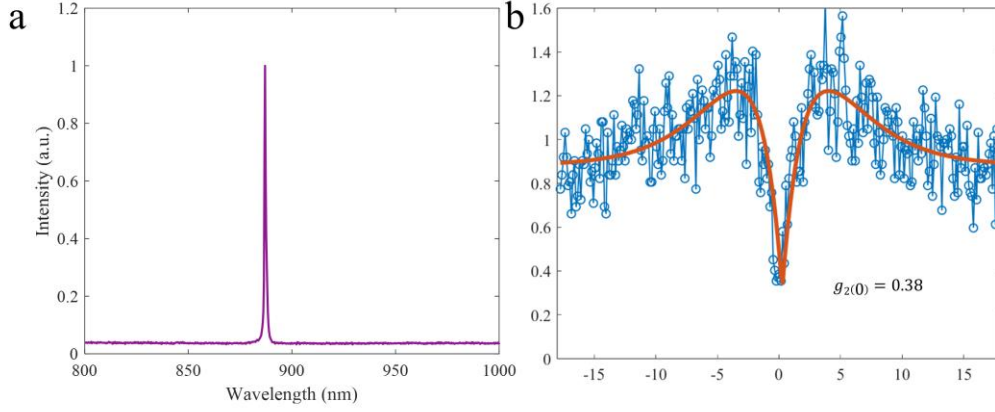


Figure 5.12. (a) Filtered spectrum of the X1 emission used for the $g^{(2)}(0)$ measurements. (b) Saturation $g^{(2)}(0)$ under an injection current of 1.6 μA , showing a value of 0.38, proves the single emission at high injection current.

Single photon purity measurements of the X1 state were performed using the Hanbury–Brown and Twiss interferometer under an injection current of 0.8 μA . The measured second-order correlation function is shown in Figure 5.11(a). After spectral filtering (the filtered QD spectrum is shown in Figure 5.12(a)), a zero-delay value of 0.21 in the inset was obtained, confirming our QD-nanowire device is indeed a single photon emitter. The measured single photon purity is likely underestimated, as the experiment was limited by the time jitter of the avalanche photodiodes ^[271] and the use of continuous direct current injection, both of which are known to degrade the measured $g^{(2)}(0)$ value. Therefore, the intrinsic $g^{(2)}(0)$ value is expected to be lower. The brightness of the single emitter was evaluated through power-dependent photon count measurements, as shown in Figure 5.11(b). The photon count rate increases with injection current and reaches approximately one million counts per second at $\sim 1.5 \mu\text{A}$. This high brightness is attributed to efficient coupling between the QD emission and the nanowire cavity modes. Saturation $g^{(2)}(0)$ under an injection current of 1.6 μA , showing a value of 0.38, proves the single emission at high injection current in Figure 5.12(b). The long-term operation stability of the device was also investigated under an injection current of 0.8 μA , as shown in Figure 5.11(c-e). Figure 5.11(c) presents the temporal evolution of the photon count rate monitored continuously over 1 hour, with the inset highlighting the stability during the first 5 minutes. The histogram in Figure 5.11(d) further quantifies the statistical distribution of the count rate. Although there are some fluctuations in the emission count rate over this time span,

the dominant peak centered at ~ 200 kcps in the histogram indicates the long-term stability of the photon emission can be maintained, which is comparable to optically pump device ^[272]. These fluctuations are attributed to blinking effects triggered by enhanced non-radiative Auger recombination in the presence of excess charges under a high electric field (~ 50 nm intrinsic region, corresponding to $\sim 10^6$ V/cm) ^[273, 274]. This issue can likely be mitigated by increasing the thickness of the i-layer above 100 nm, thereby reducing the electric field by approximately one order of magnitude. The sudden fluctuation around ~ 0.55 h is likely attributed to instrumental fluctuation. Figure 5.11(e) depicts the EL spectral map recorded over one hour. The persistent and stable emission line at 887.5 nm indicates that electrons and holes recombine deterministically within the single quantum dot, with no observable heating effect during prolonged current injection that could otherwise redshift the emission wavelength, thus detune it from the cavity mode ^[96]. $g^{(2)}(0)$ measurement on another device with the same growth parameter of Device 1 in Figure S8 proves the reproducibility. An electrically injected QD-in-nanowire array device performance in Figure S9 shows the yield, and scalability.

5.5. FDTD simulation

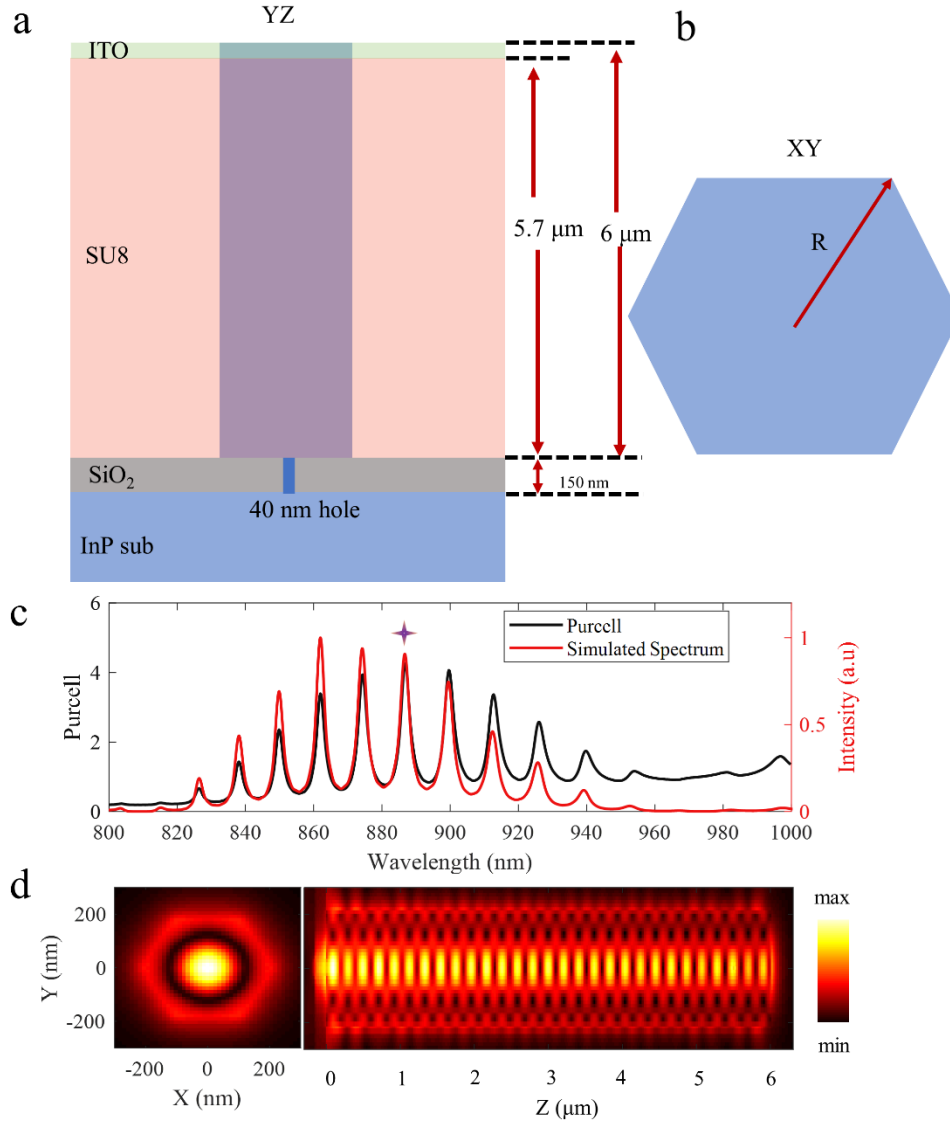


Figure 5.13. Schematic of the structure used for FDTD simulation viewed from the (a) YZ plane and (b) XY plane. (c) Simulated spectrum (red) and Purcell factor (black) of device 1. (d) Simulated electrical field profile on the XY plane and XZ plane of X1 emission.

FDTD simulations are performed using the Lumerical 3D Electromagnetic Simulator commercial solver package. A boundary condition of perfectly matched layer is used to place around the structure with a distance of at least half a wavelength distance of the NW boundary. Figure 5.12 shows schematic of the structure for simulation viewed from YZ plane (Figure 5.13 (a)) and XY plane (Figure 5.13 (b)). The InP NW with a height of 6 μm and a diameter of R and is sitting on 150 nm SiO₂ mask with an opening width of 40 nm. The NW is embedded

with a 5.7 μm -thick layer of SU-8, except the top 300 nm, which is covered with ITO. The refractive indices of the different materials are set to: $n_{\text{SiO}_2} = 1.5$, $n_{\text{InP}} = 3.45$, $n_{\text{SU-8}} = 1.5$, $n_{\text{ITO}} = 1.48$. A broadband TM mode dipole source with a bandwidth from 800 to 1000 nm is placed at the centre of the NW at a height of 5.5 μm from the bottom of SiO_2 to excite the structure. FDTD simulation of the NW with the same experimental dimension confirms the coupling as shown in Figure 5.13(c), with the results of simulated Purcell factor (black line) and spectrum (red line) of device 1. The purple star indicates the position of the X1 peak of device 1 in Figure 5.10(a), where a Purcell factor of 4.3 is obtained. This value can be further increased by increasing the thickness of the SiO_2 layer to reduce light leakage into the substrate. The QD emission peak of Device 2 at ~ 950 nm exhibits a calculated Purcell factor of ~ 0.9 , indicating no Purcell enhancement. Simulated electrical field profiles on the XY and XZ planes at the wavelength of 887.5 nm in Figure 5.13 (d) show the QD emission is coupled to the TM_{01} mode of the NW.

5.6. Study of n-i-p QD in NW

In the previous section, we focused on the p-i-n structure with a p-doped NW core, and an n-doped NW shell. One of the main reasons for this design is the need to choose a transparent electron-selective contact. ITO is selected because it combines high optical transparency with high electrical conductivity and a work function that favours electron transport when interfaced with n-type InP. In contrast, its work function is less favourable for hole injection into p-type InP, resulting in a larger contact barrier and higher contact resistance. Therefore, ITO is more suitable for the n-type shell than for direct contact with the p-type InP shell. However, the InP core requires high temperature growth to ensure its thin diameter making it challenging for the p-doping. Thus, we also explore the n-i-p structure with an n-doped core and a p-doped shell. The structure of the device as illustrated in Figure 5.14 (a), a core-shell n-i-p nanowire with the QD embedded in the i-region of the nanowire. The n-doped InP nanowire core was first grown by on a (111)A n+ InP substrate for 4 mins at 740 $^{\circ}\text{C}$ with a V/III ratio of 81, and a SiH_4 flow rate of 1.7×10^{-6} mol/min. The quantum dot and i-layer were grown under the same conditions as the p-i-n structure. The red circle in between the layers represents the single InAs QD. The p-doped InP shell layer was grown at 620 $^{\circ}\text{C}$ for 2 mins with a V/III ratio of 800 and the DEZn flow rate of 4×10^{-6} mol/min. 50 nm Au was deposited as the back contact. Here

we used 7 nm Au and 100 nm ITO as the hole-selective top contact after a planarisation and etch-back step. The thin Au layer was introduced to improve band alignment.

Figure 5.14 (b) shows the SEM image of the In droplet on a n-doped InP NW grown at 400 °C with a TMIn deposition time of 0.1 s and a TMIn flow rate of 2×10^{-6} mol/min. Figure 5.14 (c) shows the SEM image InP NW diode after capping with a layer of p-doped shell. The NW has a diameter of ~ 80 nm in (b) and uniform hexagonal structure in (c) showing unaffected and repeatable process even swapping the doping order.

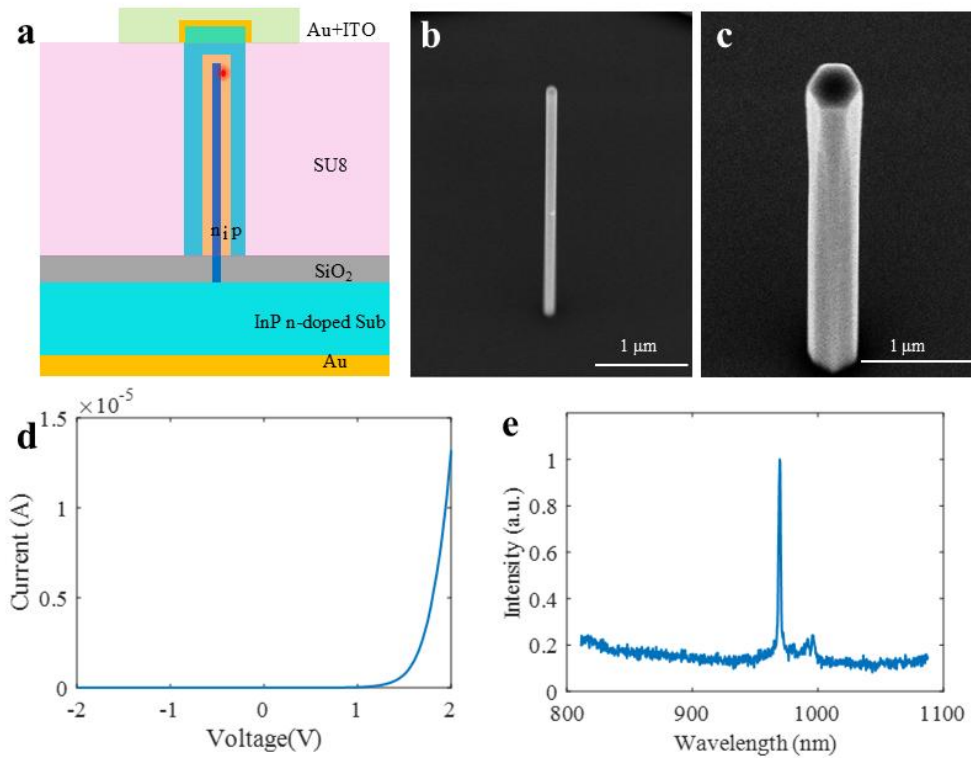


Figure 5.14. n-i-p QD-in-NW diode. (a) Schematic cross-sectional view of a core-shell n-i-p InP NW diode with a single InAs QD (red dot) embedded in the intrinsic region. SiO₂ is used as the mask layer, SU-8 as the planarisation layer, Au as the back contact and Au/ITO as the top contact. (b) SEM image of In droplet on n-doped InP NW core. (c) SEM image of the NW after capping with a layer of p-doped shell. (d) Current-voltage (I-V) characteristics of the device. (e) The corresponding EL spectrum of the device under an injection current of 1 μA.

Device fabrication processes were the same as the p-i-n structure. We first measured the I-V curve to prove there is p-n junction. Figure 5.14(d) is the I-V curve of the n-i-p device measured

from -2 V to +2 V, the devices also exhibit typical diode-like rectifying behaviour, with negligible current under reverse bias and a turn-on voltage of ~ 1.4 V. The low leakage current in the reverse-bias regime showing a good p-n junction. Figure 5.14 (e) is the EL spectrum measured at 5 K under an injection current of 1 μ A. The sharp peak at ~ 980 nm is from the QD emission. Our study shows another potential approach to achieve electrically injected single photon emitter.

5.7. Summary

In this section, we successfully grow single InAs QDs within a core-shell InP NW diode structure to realise electrically driven single-photon emitters. A hole-in-ring pattern is designed to reproduce the growth conditions as those of the NW array. The p-doped core NW exhibits good morphology and crystalline quality. We also investigate the influence of growth temperature on the n-doped InP shell layer and achieve uniform hexagonal NWs after capping. The PL of single QDs is studied, and the maximum Purcell enhancement is simulated prior to device fabrication. Furthermore, C-AFM measurements confirm the core-shell structure. The current-voltage characteristics of the fabricated devices exhibit typical p-n junction rectifying behaviour. We further demonstrate that the QD emission couples to the Fabry-Perot mode of the NW under high current injection, as supported by FDTD simulations showing coupling to the TM_{01} mode of the NW with a simulated Purcell factor of 4.3. The QD is confirmed to operate as a single-photon emitter, with a measured $g^{(2)}(0)$ value of 0.21 and a brightness exceeding one million counts per second under a 1.5 μ A current injection. Moreover, the device exhibits long-term stability, maintaining the same emission wavelength and photon count rate over an hour.

Electrical injection provides a compact route to operate the single-photon source without external optical excitation. The bottom-up architecture further enables direct integration of a single QD within the nanowire cavity. However, deterministic spectral matching between the QD emission and the FP mode remains challenging. The photon extraction efficiency and indistinguishability also require further investigation. Future work could optimise the QD position and nanowire geometry to improve QD-cavity coupling and photon extraction. These advances could enable scalable electrically driven single-photon sources for integrated quantum photonics.

CHAPTER 6

6. Bottom-up single quantum dots in micro-ring resonators for on-chip integrated single photon emitters

6.1 Introduction

On-chip integrated quantum photonics is a compelling platform in providing low-noise, scalable and compact single photon sources^[275-277] for quantum communication^[278], quantum teleportation^[279], and quantum memories^[280]. For on-chip integration, two major components, namely optical waveguides and ring resonators, are required in generating stable state photons^[116]. While the former is used to guide and route photons^[281], the later plays a key role in generating functional single photons such as pure orbital angular momentum state^[96], chiral or spin single photon emission^[282, 283], and interference entangled photons^[284, 285]. However, achieving such performance demands the integration of single emitters with ring cavities. We have already demonstrated that by combining droplet epitaxy with SAE, a single QD can be deterministically grown in a single nanowire and coupled with cavity mode of the nanowire in Chapter 4. However, due to the size and composition distribution of the QDs^[115, 136], coupling the QD emission into the nanowire cavity mode is a ‘hit-or-miss’ exercise. In comparison, ring resonators have superior advantages as they can support multiple high-Q factor whispering-gallery modes (WGMs) over a broad spectral range^[286, 287]. By taking advantage of this property, there is a higher possibility of coupling the QD emissions to one of these modes.

In this chapter, we further apply the droplet epitaxy technique with bottom-up InP micro-rings to incorporate InAsP single QDs for single photon sources. The growth parameters (i.e. temperature, TMIn flow rate and deposition time) for micro-ring, droplet density, ring capping layer are optimised to achieve low density QDs in a high Q-factor ring resonator. Our micro-rings have high Q-factors (~ 3000 for the $4\ \mu\text{m}$ diameter rings and ~ 4000 for the $10\ \mu\text{m}$ diameter rings) with a low surface roughness of $0.17\ \text{nm}$. We also show that the QD coupled into the cavity mode of a $4\ \mu\text{m}$ diameter ring results in an ultra-short lifetime of $70 \pm 1.8\ \text{ps}$ with a single photon purity $g^{(2)}(0)$ of 0.241 . FDTD simulations show the single emitter has

been coupled with the TE (82,1,2) WGM of the ring with a Purcell enhancement factor of 12. By increasing the diameter of the rings from 4 μm to 10 μm , more and spaced closer high-Q WGMs can be supported, contributing to higher probability for the single QDs to be coupled with. Our results contribute to the development of bottom-up quantum light sources in on-chip photonic integrated circuits for quantum technologies.

6.2. Methods and theory

6.2.1 Experimental methods

Pattern preparation: 440 nm SiO_2 layer was deposited on semi-insulating InP (111)A substrates by plasma-enhanced chemical vapour deposition, followed by spin coating of 400 nm electron beam resist (mr-ebr-0.3). Patterns were designed with rings, with a width of 40 nm for different diameters, which were then exposed by electron beam lithography (Raith 150) at a voltage of 20 kV and a current of 0.023 nA. The patterns were transferred onto the SiO_2 layer using inductively coupled plasma reactive ion etching (ICP-RIE) with CHF_3 and Ar. The patterned substrates were trim etched in a mixed solution of 20% H_2O_2 , 20% H_3PO_4 and 60% DI water for 10 mins.

Material growth: Epitaxial growth was conducted on the SiO_2 patterned substrates using a horizontal flow metalorganic vapour phase epitaxy system with at a pressure of 100 mbar. The substrates were thermally deoxidised for 10 mins at the temperature of 750 $^\circ\text{C}$ under PH_3 flow prior to growth. InP core rings were first grown for 4 mins at different temperatures of 750, 725, 700, 680 $^\circ\text{C}$ with a PH_3 and TMIn flow rate of 2.1×10^{-3} and 6.091×10^{-6} mol/min, respectively, corresponding to a V/III ratio of 350. For droplet formation, the reactor was then cooled to the desired temperatures under PH_3 protection with the same flow rate and stabilised for 3 minutes. PH_3 was then removed, and the chamber purged for 30 s to remove any residual group V source, after which TMIn was introduced into the reactor. Various TMIn flow rates and deposition times were investigated. The substrates were cooled down immediately to room temperature to investigate the droplet density. For optical properties measurement, we crystallised the droplets at 520 $^\circ\text{C}$ with an AsH_3 flow rate of 1.2×10^{-4} mol/min and a PH_3 flow rate of 1.78×10^{-4} mol/min for 4 mins, where the ratio of $\text{AsH}_3 / (\text{AsH}_3 + \text{PH}_3)$ was fixed to 0.4. The substrate temperature was then increased to 625 $^\circ\text{C}$ under the protection of PH_3 protection with a flow rate of 2.1×10^{-3} mol/min and stabilised for 2 mins. InP shell layer was subsequently grown with the same V/III ratio as the initial ring for 1 min after which

TMIn flow was removed and the substrate temperature increased to 700 °C under PH₃ flow protection and stabilised for 3 mins. The purpose of this additional step is to anneal the QDs and the shell layer to further improved their crystal quality.

Material characterisation: Density and size of the droplets on InP rings were characterised by a FEI Verios 460 scanning electron microscope operated at a voltage of 5 kV and a current of 100 pA. Surface roughness of the rings and QD size measurements were carried out by Atomic Force Microscopy (Bruker, Dimension Icon) under peak force tapping mode.

6.2.2 Experimental design

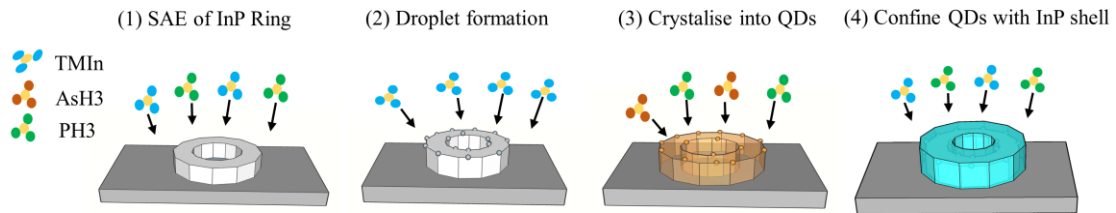


Figure 6.1. In droplet epitaxy on bottom-up InP micro-ring resonators to grow QDs in the rings. (1) Growth of InP micro-ring by introducing TMIn and PH₃ gases, (2) Introduction of TMIn precursor to form In droplets on the micro-ring, (3) Crystallisation of In droplets into InAsP QDs by introducing PH₃ and AsH₃. (4) Growth of InP shell layer to confine the QDs.

As discussed in Chapter 4, the use of droplet epitaxy to form low density QDs allows a larger growth window, in particular the droplet formation can be carried out at a wide range of growth conditions like high Miller index surfaces, temperatures, etc.^[236]. Figure 6.1a schematically illustrates the droplet epitaxy of InAsP QDs on an InP micro-ring grown by SAE. The process consists of four steps, first is SAE growth of InP micro-ring patterned with a ring opening on the SiO₂ mask by introducing TMIn and PH₃ precursors. After growth of the rings, the temperature was reduced to the desired value under PH₃ ambient. PH₃ was then removed before introducing TMIn into the chamber for a fixed amount of time to form In droplets. QDs are then formed by introducing the group V gases of AsH₃ and PH₃ to crystallise the droplets. Finally, an InP capping layer is grown by increasing the temperature to 625 °C and stabilising for 3 mins for high quality cavity formation.

6.2.3 Theoretical background of whispering gallery modes

The concept of WGMs originates from acoustics, first observed by Lord Rayleigh in 1878 in St. Paul's Cathedral ^[288], where whispered sounds could travel along the circular wall of the dome and be heard clearly on the opposite side. The optical analogue of this phenomenon occurs when light waves are confined by continuous total internal reflection along the curved boundary of a dielectric microstructure, such as a microsphere, micro-disk, or micro-ring. These circulating optical modes are referred to as WGMs.

In a WGM resonator, light propagates azimuthally around the periphery like Figure 6.2, trapped by total internal reflection at the interface between the high refractive index cavity and the surrounding lower-index medium (typically air, $n=1$). WGMs are excited when the optical path length for one complete round trip is an integer multiple of the wavelength ^[289]. The stable resonance condition can be expressed as:

$$m\lambda_m = 2\pi n_{eff} R \quad (6.1)$$

where m is the azimuthal mode number, λ_m is the resonance wavelength, n_{eff} is the effective refractive index of the mode, and R is the radius of the micro-resonator. By rearranging this equation, the resonant wavelength can be expressed as,

$$\lambda_m = \frac{2\pi n_{eff}^m R}{m} \quad (6.2)$$

The free spectral range (FSR) is wavelength difference between resonant peaks,

$$FSR = \lambda_m - \lambda_{m+1} = \frac{2\pi n_{eff}^m R}{m} - \frac{2\pi n_{eff}^{m+1} R}{m+1} = \frac{2\pi n_{eff}^m R}{m(m+1)} \quad (6.3)$$

If $m \gg 1$,

$$FSR = \frac{2\pi n_{eff}^m R}{m(m+1)} \approx \frac{\lambda^2}{2\pi n_{eff}^m R} \quad (6.4)$$

Here, it can be observed that FSR is inversely proportional to the size and refractive index of the microcavity. Therefore, a smaller ring size results in a larger FSR, and vice versa.

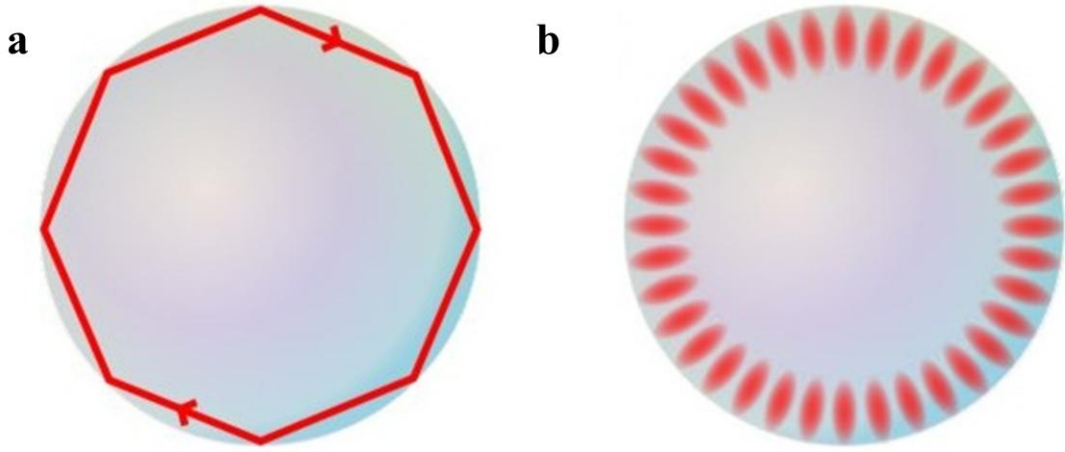


Figure 6.2. Light propagates azimuthally around the periphery in a whispering gallery mode resonator. (a) Ray-optics representation of a whispering gallery mode, where light propagates along the cavity boundary via repeated total internal reflection. (b) Corresponding the electric field intensity distribution ($|E|^2$), with discrete field maxima (antinodes) along the periphery of the cavity.

The Q-factor represents the ratio between the total optical energy stored in the resonant cavity and the energy lost per optical cycle during light propagation within the microcavity ^[290]. It reflects the confinement capability of the cavity for the electromagnetic field. A higher Q-factor indicates stronger confinement and lower energy loss, while a lower Q-factor corresponds to weaker confinement and higher loss.

$$Q_{\text{Factor}} = \frac{\nu}{\Delta\nu} \quad (6.5)$$

Where, ν represents stored energy, $\Delta\nu$ represents the loss energy per cycle, respectively.

Another important parameter is the finesse of the cavity, which is defined as the ratio of FSR and resonance width,

$$\text{Finesse} = \frac{FSR}{FWHM} \quad (6.6)$$

It measures the sharpness of the resonances relative to their spacing.

6.3. Epitaxial growth of single InAsP quantum dots in InP micro-ring resonators

In this section, we systematically investigate the influence of growth parameter on both ring and droplet density for the growth of QDs in InP micro-rings for single photon sources.

6.3.1. Effect of growth temperature on InP ring

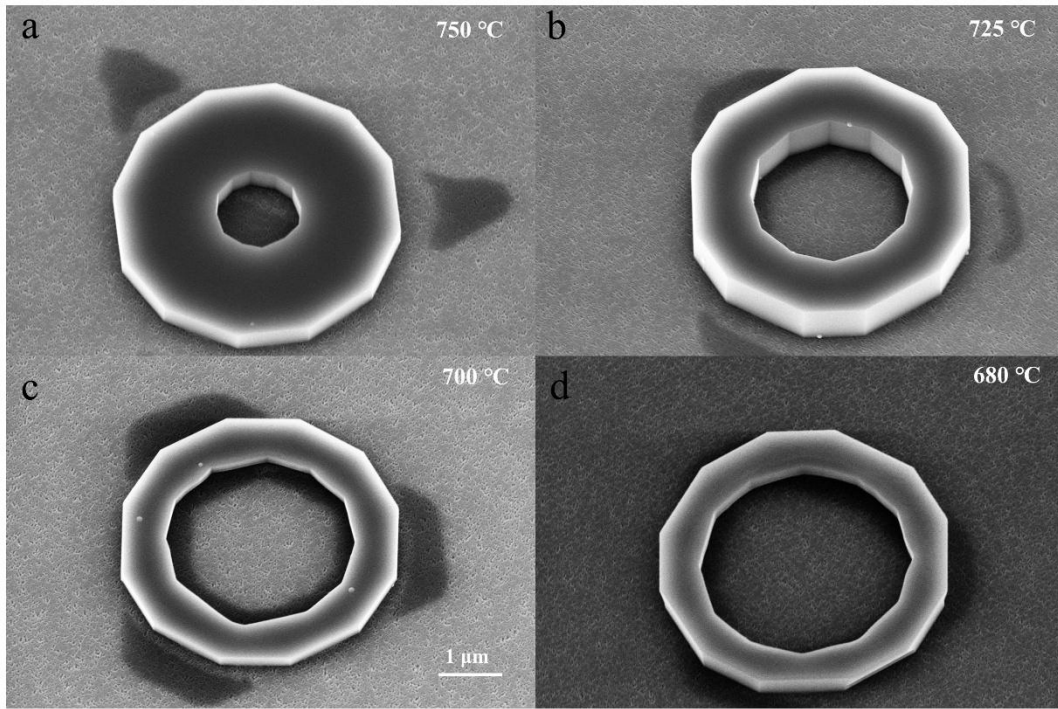


Figure 6.3. InP ring growth under different growth temperature: (a) 750 °C, (2) 725 °C, (3) 700 °C, (4) 680 °C.

Figure 6.3 illustrates the effect of growth temperature on the lateral growth behaviour of InP rings, with a fixed V/III ratio of 350 and a growth time of 4 minutes. As the growth temperature decreases from 750 °C to 680 °C, the extent of lateral overgrowth is progressively suppressed. At higher temperatures, enhanced adatom diffusion leads to significant sidewall deposition, resulting in thicker structures that tend towards disk-like morphology. In contrast, lower growth temperatures reduce the adatom diffusion length, thereby limiting lateral migration and preserving well-defined ring structures with minimal overgrowth.

These results demonstrate that growth temperature is a key parameter for controlling the lateral growth of InP, enabling the tuning of the morphology from disk-like structures to well-defined rings. It should be noted that other growth parameters, such as the V/III ratio and growth time, also play important roles in determining the final morphology. However, in this work, the growth temperature is isolated as the primary variable to elucidate its influence on lateral growth behaviour.

6.3.2. Investigation of droplet (quantum dot) density

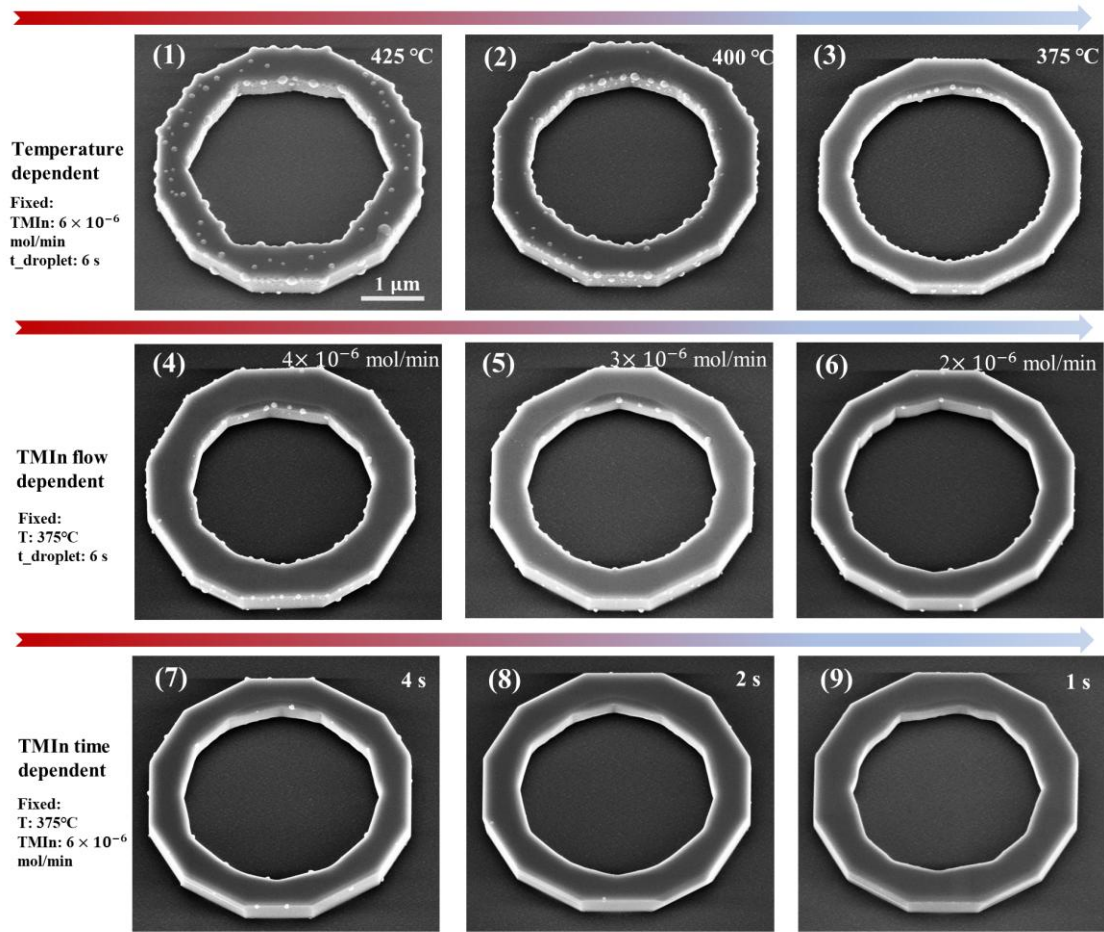


Figure 6.4. SEM images of In droplet density on 4 μ m diameter InP micro-rings for different deposition conditions: (1-3) deposition temperature: 425 °C, 400 °C, 375 °C, respective; (4-6) TMIn flow rate: 4×10^{-6} mol/min, 3×10^{-6} mol/min, 2×10^{-6} mol/min, respectively. (7-9) TMIn deposition time: 4s, 2s, 1s, respectively. All images have the same magnification.

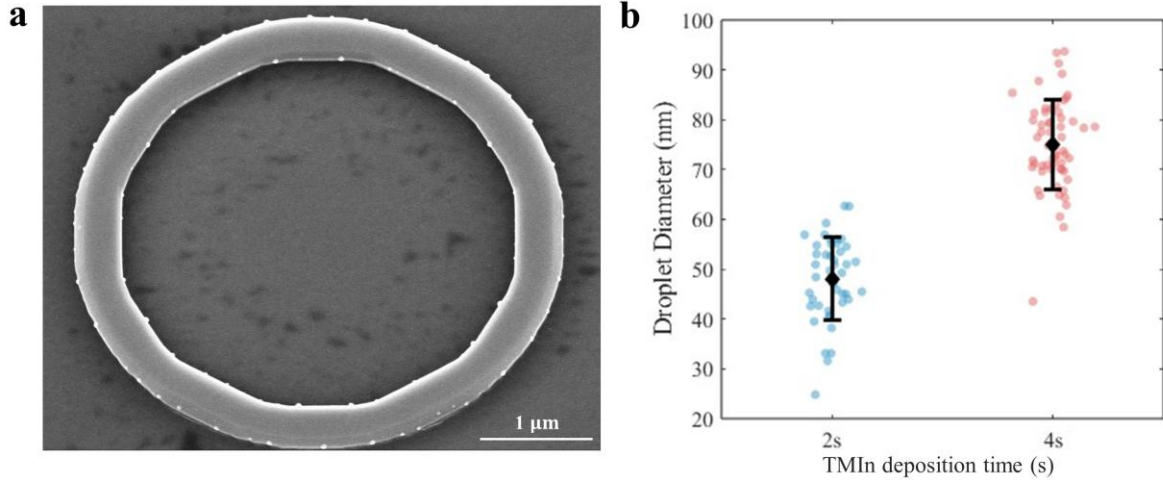


Figure 6.5. (a) SEM image showing In droplets forming on a 4 μm InP ring with a TMIn flow rate of 6×10^{-6} mol/min and deposition time of 4 s at a temperature of 350 $^{\circ}\text{C}$. The droplets only form on edge region (the intersection of sidewalls facets ($\{11\bar{2}0\}$ and $\{1\bar{1}00\}$) and top facet $\{0001\}$), where the surface energy is high. Statistical measurements of droplet diameters for ~ 50 droplets from TMIn deposition time of 4 s and 2 s show an average diameter of 75.0 and 48.1 nm, respectively.

After growth of the rings, the temperature was reduced to the desired value under PH_3 ambient. PH_3 was then removed before introducing TMIn into the chamber for a fixed amount of time to form In droplets. For single photon emitter applications, ideally only a single QD is required inside the micro-ring. However, the area of the InP micro-ring is too large and more than one droplet can simultaneously form on the ring. Hence, we investigate the formation of In droplets on the micro-rings under different growth conditions (TMIn deposition temperature, flow rate, deposition time in Figure 6.4) to achieve as low density as possible. The effect of growth temperature on the droplet density on the 4 μm ring at a fixed TMIn flow rate of 6×10^{-6} mol/min and deposition time of 6 s is shown in Figure 6.4(1-3). In adatoms prefer to nucleate at regions where the surface energy is high especially at the edges of the ring, i.e. the intersection of sidewalls facets ($\{11\bar{2}0\}$ and $\{1\bar{1}00\}$) and top facet $\{0001\}$. Droplets start to form at the top facets at higher temperature (400 and 425 $^{\circ}\text{C}$) as a result of surface decomposition thereby forming nucleation sites. (Figure 6.5(a) shows that the droplets only form at the edges of the ring even with a high TMIn flow rate at 350 $^{\circ}\text{C}$ as the top facets are not decomposed lower temperatures) However, as the growth temperature is lowered, the density

of In droplets on the rings is also substantially reduced due to less decomposition of TMIn. Figure 6.4(4-6) shows the effect of TMIn flow rate on the density of In droplets at a fixed temperature of 375 °C and deposition time of 6 s. As expected, the density of droplets decreases with decreasing TMIn flow rate and indeed at $2 \times 10^{-6} \text{ mol/min}$ only a few droplets are formed on the sidewalls. However, this may still be undesirable as single photon emitters and we can further reduce the density of the droplets by reducing the deposition time of TMIn as can be clearly seen in Figure 6.4(7-9). Only several droplets are formed at 2 s deposition time while no droplet is formed at 1 s. Figure 6.5 (b) shows statistical measurements of droplet diameters for ~50 droplets from TMIn deposition time of 4 s and 2 s show an average diameter of 75.0 ± 8.89 and 48.1 ± 8.32 nm, respectively.

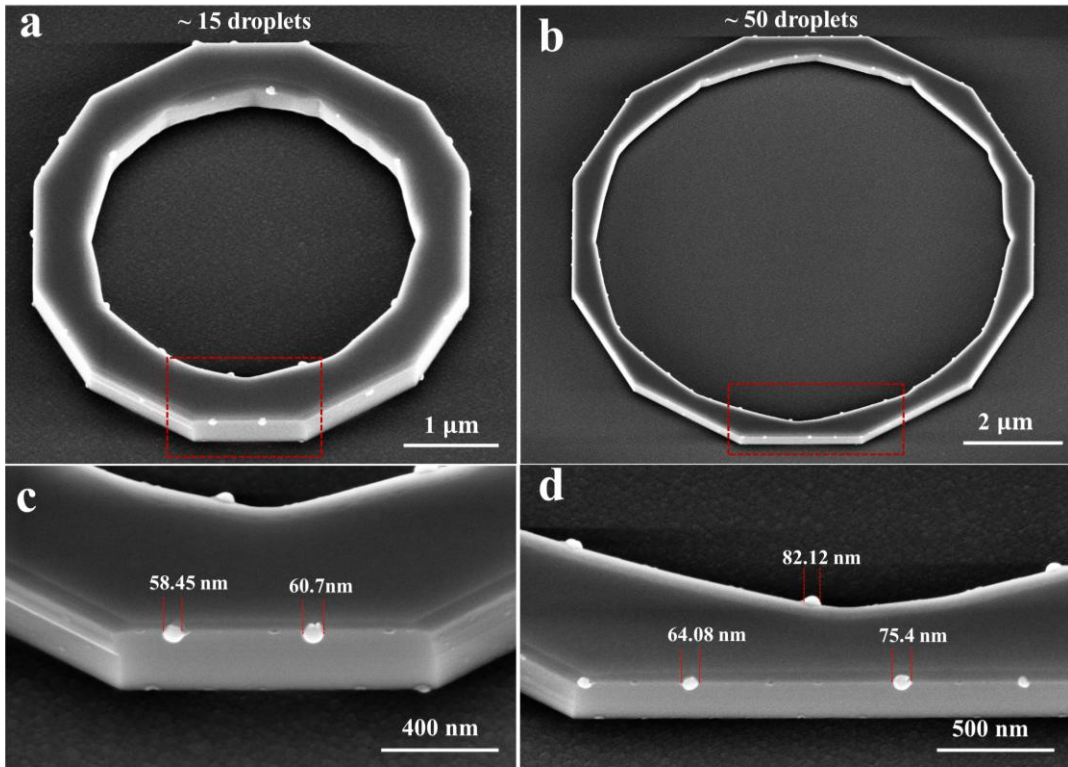


Figure 6.6. SEM images of In droplets with a TMI flow rate of $2 \times 10^{-6} \text{ mol/min}$ and a deposition time of 4 s at the temperature of 400 °C on InP micro-rings with different sizes for single photon source measurements: (a) 4 μm ring (~ 15 droplets) and (c) the corresponding higher magnification image of the region highlighted by the red rectangle in (a), (b) 10 μm ring (~ 50 droplets) and (d) the corresponding higher magnification image of the region highlighted by the red rectangle in (b).

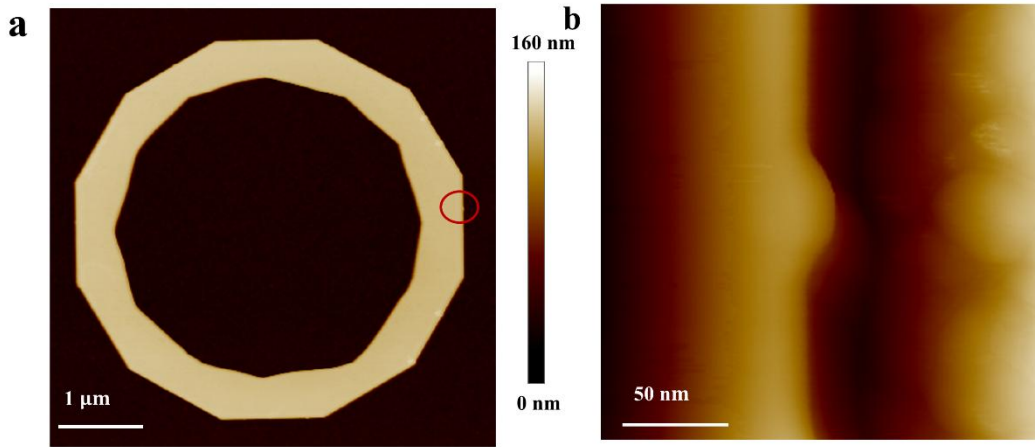


Figure 6.7. (a) AFM image of a 4 μm ring after crystallising the In droplets into InAsP QDs. (b) High magnification AFM image of the region highlighted by the red circle in (a), showing the QD has a diameter of 50 nm and a height of 16 nm.

For optical measurements, we directly increase the temperature to 625 $^{\circ}\text{C}$ for InP capping layer growth after crystallisation of the droplets into QDs at 520 $^{\circ}\text{C}$. We have investigated the droplet density on one 4 μm ring and obtained the lowest density of 5 droplets (QDs) in Figure 6.4 (8). However, PL measurements on the sample did not show any emissions from the QDs, most likely due to the decomposition of the QDs or intermixing of the QDs on the surrounding InP layer as a result of the high growth temperature of the capping layer. Therefore, for subsequent measurements, we used samples which were grown with a higher density of droplets (and hence QDs) where the TMIn deposition time was increased from 2 s to 4 s as shown in the SEM images in Figure 6.6. Figure 6.6 (a) is the SEM image of 4 μm ring, about 15 droplets can be seen. In the magnified SEM image in (c), the representative droplet size is around 60 nm. The size of the QDs was measured by AFM after crystallising the droplets (but before capping) as shown in Figure 6.7. The representative QD shows a diameter of 50 nm and a height of 16 nm, however, it is expected the final size of the QDs would be reduced during the temperature ramp up for growth of the capping layer. For the larger 10 μm diameter ring in Figure 6.6 (b), about 50 In droplets could be seen. In the high magnification SEM image in Figure 6.6(d), the size of the droplets ranges from 60 to 80 nm in diameter. This fluctuation is due to the In adatom diffusion rate difference between the inner and out facets of the ring.

6.3.3. InP capping layer quality

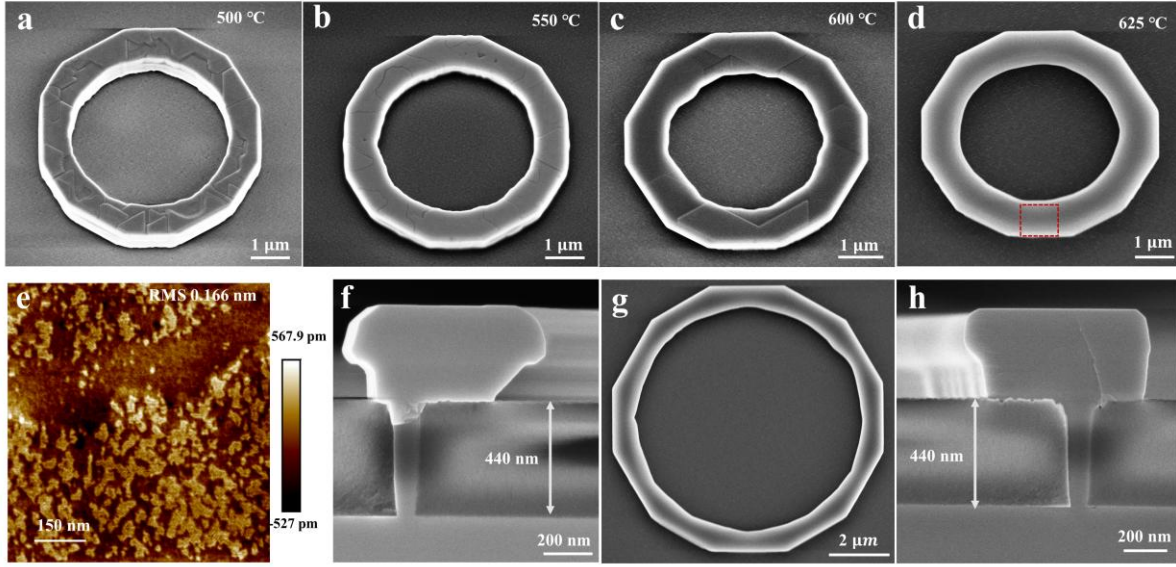


Figure 6.8. SEM images of 4 μm InP ring grown with a high density of QDs and after capping the QDs with different shell growth temperature: (a) 500 $^{\circ}\text{C}$, (b) 550 $^{\circ}\text{C}$, (c) 600 $^{\circ}\text{C}$, (d) 625 $^{\circ}\text{C}$. (e) AFM image of the red rectangular region shown in (d). (f) SEM image of the cross-section view of InP ring in (d). (g) SEM image of 10 μm InP ring after capping the QDs at the temperature of 625 $^{\circ}\text{C}$ and corresponding cross-section in (h).

As mentioned in the introduction, one of the main advantages of a ring resonator/cavity is its high Q-factor, which allows strong coupling between the QD emissions and the cavity modes of the ring^[281]. To evaluate the Q-factor of the rings, we grew a relatively high-density of QDs as a broadband light source to illuminate the cavity modes in the rings. After depositing the In droplets using the same conditions as Figure 6.4(a-4), AsH_3 and PH_3 were introduced at 520 $^{\circ}\text{C}$ for 3 minutes in order to crystallise them into InAsP QDs. A layer of InP was then grown over the QDs to provide confinement. It is important for this layer to have good crystal quality and with excellent morphology that conforms to the underlying ring so as not to compromise the Q-factor of the ring^[291]. To minimise the decomposition of the QDs whilst ramping up the temperature after QD formation, group V flow was maintained. Figure 6.8(a-d) shows the SEM images of the InP micro-rings after shell capping for 1 minute at various temperatures with a V/III ratio of 350, with a subsequent annealing at 700 $^{\circ}\text{C}$ for 3 minutes. At a growth temperature of 500 $^{\circ}\text{C}$ for cap layer deposition, the ring surface is rough due to the low diffusion rate of the

adatoms. However, increasing the growth temperature contributes to higher and more uniform diffusion rate of the adatoms, thus the ring morphology improves, especially at 625 °C , where a smooth and excellent dodecagon ring is seen. Figure 6.8(e) is the AFM image of the rectangular region indicated in Figure 6.8(d), showing the root mean square (RMS) surface roughness is 0.17 nm, which clearly illustrates the facets are smooth. Figure 6.8(f) is the SEM image of the cross-section view of InP ring in (d), showing the InP grew laterally over the 440 nm-thick SiO₂. Figure 6.8(g) is the SEM image of the 10 μm InP ring after capping the QDs with an InP layer at 625 °C and the corresponding cross-section in (h). Both images show excellent ring morphology, smooth facets, indicating the reproducibility of our technique to obtain high quality micro-rings even for larger ones.

6.4. Optical performance

This section mainly investigates the optical performance of the QDs in InP micro-ring cavities, including quality factor, emission spectra, lifetime as well as the single photon purity in difference size of the rings.

6.4.1. Quality factor measurement

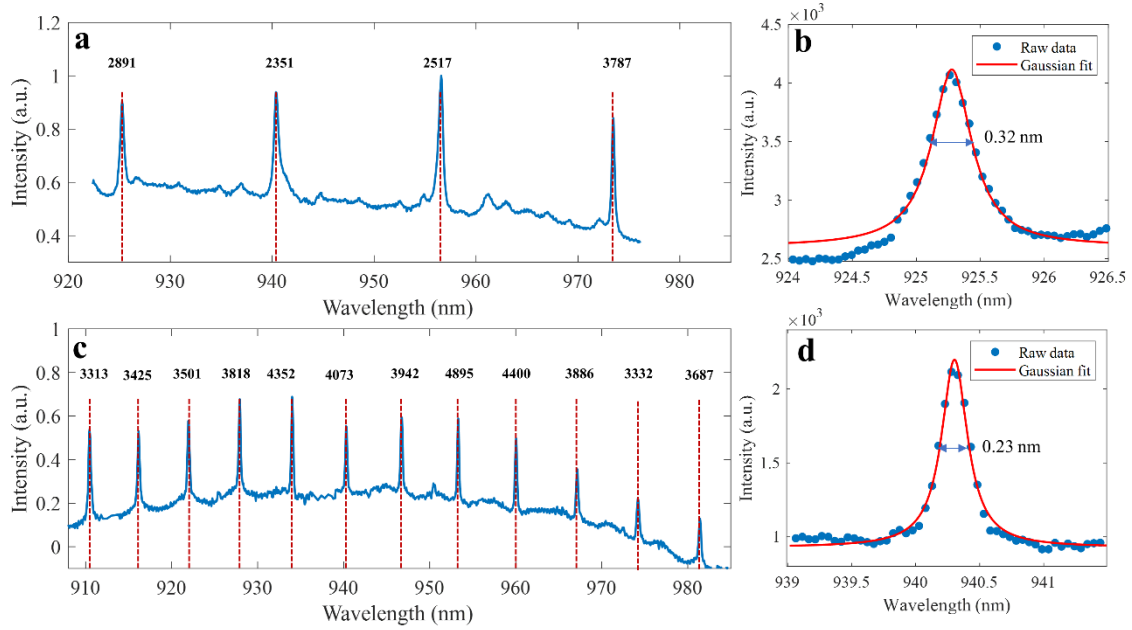


Figure 6.9. InP micro-rings with a high density of InAsP QDs as a broad-band source for cavity modes and quality factor measurements. (a) PL spectra of 4 μm InP ring showing the mode spacing and (b) the corresponding Gaussian fit of the 925.3 nm peak. (c) PL spectra of 10 μm InP ring showing the mode spacing and (d) the corresponding Gaussian fit of the 940.4 nm peak.

InP micro-rings with a high density of InAsP QDs as a broad-band source were used for cavity modes and quality factor measurements. PL measurements were taken under 100 μW excitation from a 520 nm femtosecond pulsed laser at 5 K. Figure 6.9 (a) shows a PL spectrum of the InAsP QDs imbedded in the 4 μm InP micro-ring, where four dominating cavity modes can be clearly observed with Q-factors ranging from 2000 to 3000. The experimental Q-factor is calculated by $Q = \lambda/\text{FWHM}$, and the representative Gaussian fit of the 925.3 nm peak in (b) shows a full wide at half maximum is 0.32 nm. Figure 6.9 (c) is the PL spectrum of the 10 μm InP ring, showing that more cavity modes are supported, as expected for a ring with larger

diameter. Moreover, the Q-factors of these cavity modes are substantially higher than those in the smaller ring due to less bending loss ^[292]. The representative Gaussian fit of the 940.3 nm peak in (d) shows a full width at half maximum of 0.23 nm. Note that the spectral resolution in our system is limited by the 1200 lines/mm grating, and the measured Q-factors would be even higher using higher-resolution gratings ^[281].

6.4.2. Single quantum dot in ring measurement

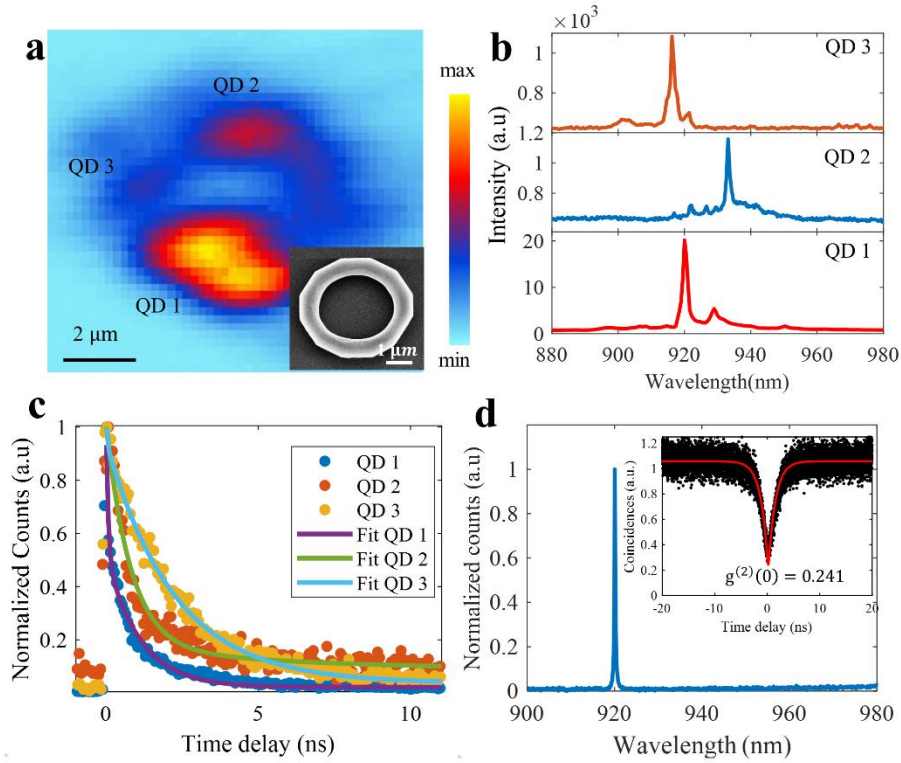


Figure 6.10. Optical characterisation of the QDs in 4 μm ring. (a) PL map of a 4 μm ring with low density QDs measured under 1 μW excitation from a 750 nm continuous wave laser at 5 K. (b) PL spectra of the three QDs indicated in (a). (c) Time-resolved PL measurements taken at the peak intensity of each of the three QDs and corresponding single exponential fitting. (d) Second-order correlation measurement of the peak emission of QD 1.

Figure 6.10(a) shows the PL map of a 4 μm ring with low density QDs sample measured under 1 μW laser power excitation. The SEM image of the ring is shown in the inset. Only three QD emissions could be observed from this sample with the emission from one QD dominating over the other two. Although this sample has about 15 droplets (Figure 6.6(a)), subsequent growth

of the InP capping layer resulted in the dissociation of most of the QDs. The PL spectra of the three QDs under 2 μW excitation power are shown in Figure 6.10(b). The intensity originating from QD 1 is about a factor of 20 times higher than those from the other two QDs, suggesting that the emission is coupled to one of the cavity modes of the ring. Temporal photon decay measurement of the peak intensity of each QD in Figure 6.10(c) reveals that QD 1 has a significantly shorter carrier lifetime of 70 ± 1.8 ps compared with QD 2 of 810 ± 9.2 ps and QD 3 of 1.68 ± 0.13 ns. The dramatic decrease in carrier lifetime can be explained by the significantly enhanced radiative rates when the QD emission is resonantly coupled to a cavity mode of the ring^[143, 293]. Single photon purity measurement of QD 1 using the HBT set up after filtering with a 920 nm bandpass filter is shown in Figure 6.10(d) while the inset shows a $g^{(2)}(0)$ value of 0.241, conclusively proving it is indeed a single photon emitter. Since our single photon purity measurements were limited to time-jitter of the avalanche photodiodes^[271] and non-resonant cw excitation^[118, 125], which are known to degrade the purity results of the emitted photons, it is most likely the $g^{(2)}(0)$ value could be even lower.

We now investigate the optical properties of the 10 μm ring grown using the same growth conditions for the QDs and the capping layer as those used for the 4 μm ring. We have shown the SEM images in Figure 6.6(b) with a droplet density of ~ 50 droplets for this larger ring before capping. Figure 6.11(a) shows an excellent ring morphology after capping the QDs and Figure 6.11(b) is the corresponding PL map where emission from the QDs could be seen. There are seven QD emissions from this sample with much brighter emission from QD 1 and QD 3 compared to the other five QDs. Figure 6.11(c) shows the PL spectra of all the seven QDs under 2 μW laser excitation. The peaks at ~ 900 nm are from the underlying InP substrates. The variation of the QD wavelength is presumably due to the size/composition difference which will affect their bandgap. QD 1 has a much higher intensity compared with other QDs, implying the coupling to the cavity modes. In addition, some QDs like QD 2, 6, and 7 have broader background, presumably as a result of changing QD morphology during the heating up stage and/or growth of the capping layer at higher temperature^[294]. The relatively broad linewidths of QD 6 and QD 7 may not arise solely from cavity coupling, but could also result from emission from multiple closely spaced QDs or defect-related spectral broadening. Figure 6.11(d) shows the corresponding PL decay plots for the seven QD peaks. QD 1, 3, 4, 5, and 6 all have a short lifetime (up to ~ 300 ps) compared with QD 2 and QD 7 (1-7 ns), indicating they are coupled to cavity modes of the ring. Figure 6.12 shows the second-order correlation measurement of QD 1, a $g^{(2)}(0)$ value of 0.3179 proves the coupled QD is single emission. The coupling

efficiency of the emitters to the optical cavities depends not only on the matching of the emission wavelength to the optical modes supported by the cavities, but also by their spatial overlap ^[94, 295].

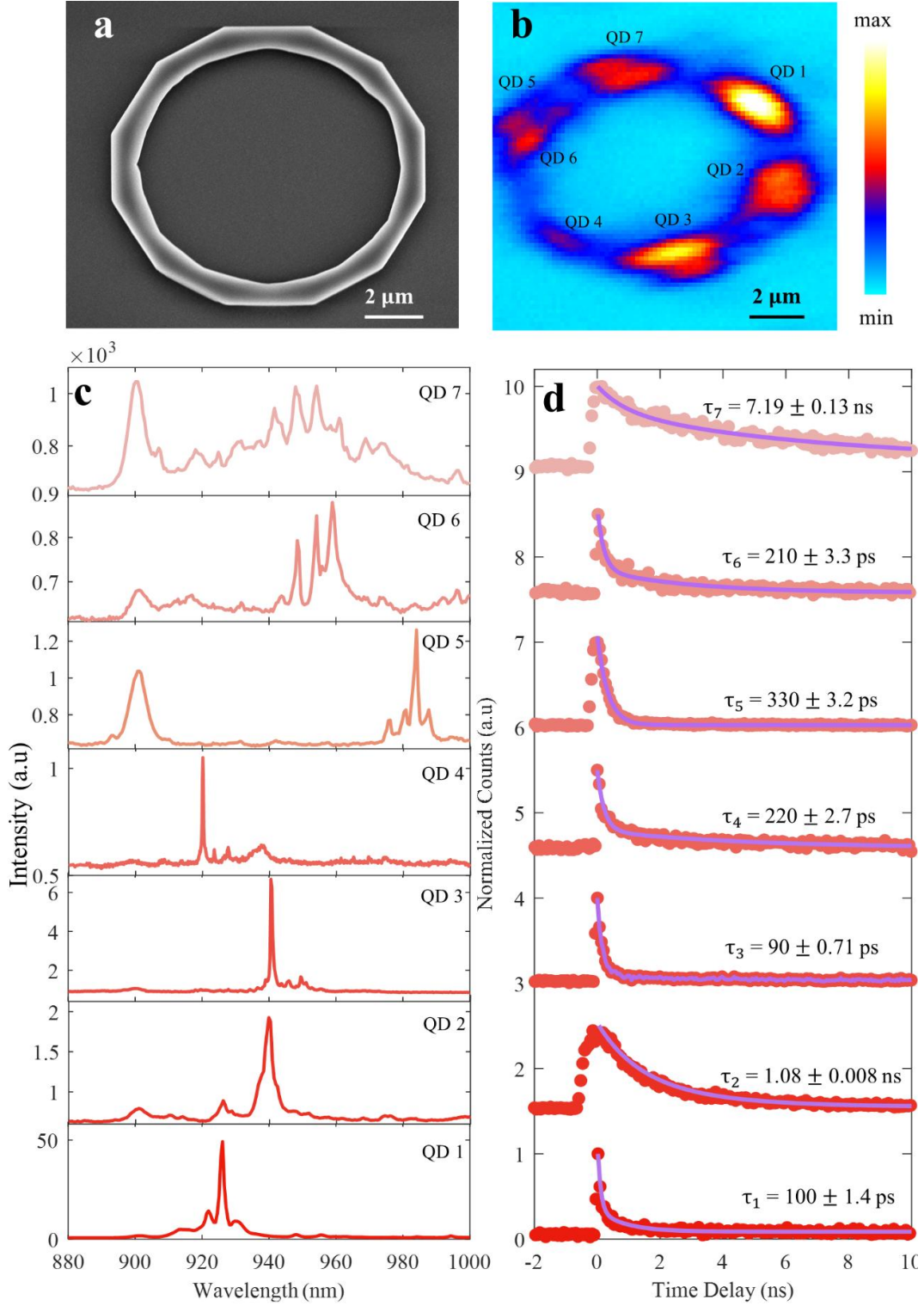


Figure 6.11. Optical characterisation of QDs in the 10 μm ring. (a) SEM image of the 10 μm ring and (b) the corresponding PL map measured under 1 μW excitation from a 750 nm cw laser at 5 K. The colour bar indicates emission intensity. (c) PL spectra of the seven QDs shown in (b). (d) Time-resolved measurements taken at the peak intensity of each QD. The plot is fitted with a single exponential decay.

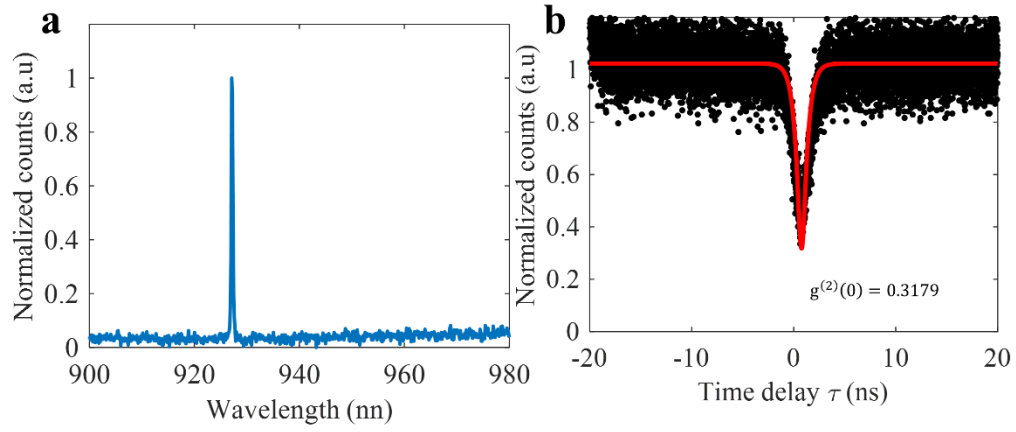


Figure 6.12. Single photon purity of the coupled QD in the 10 μm ring. (a) Spectrum of QD 1 in filtered with a 930 nm bandpass filter and (b) second-order correlation measurement of single photon purity giving a $g^{(2)}(0)$ value of 0.3179, proving its single emission.

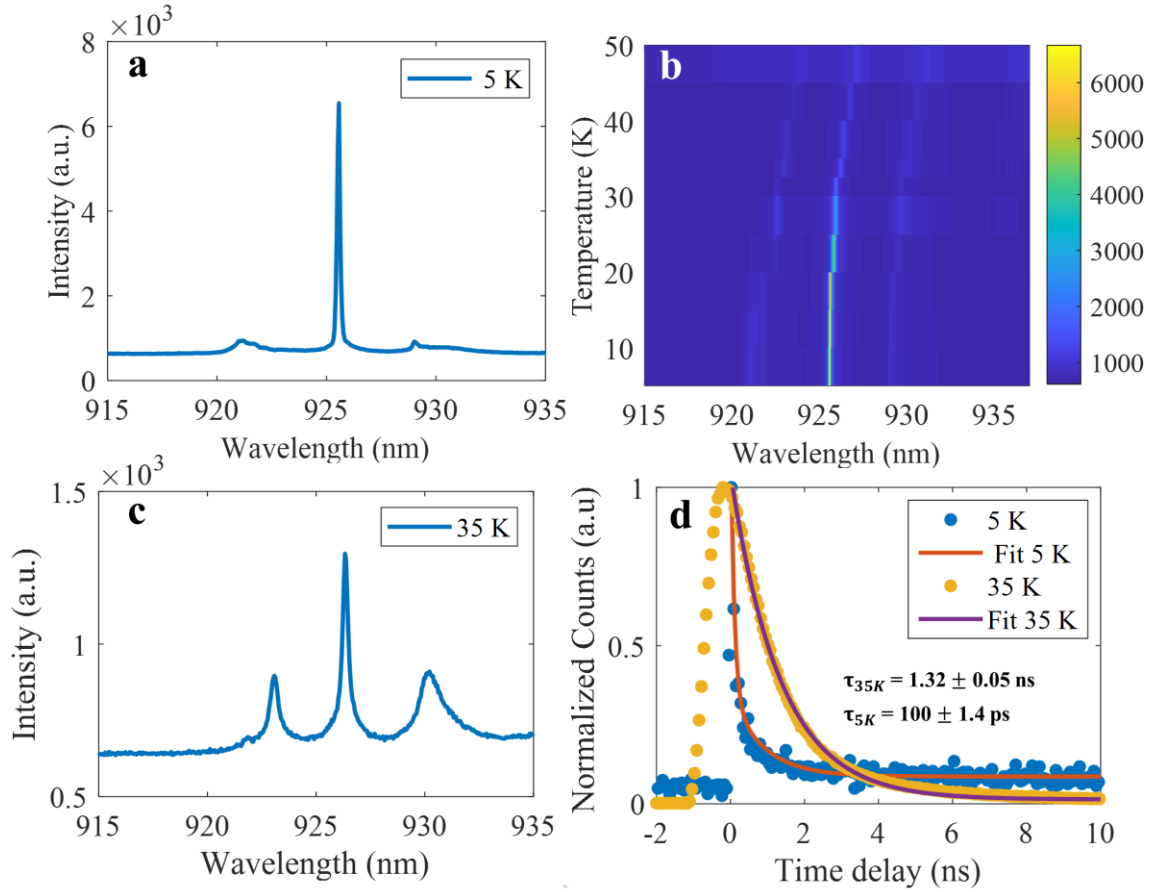


Figure 6.13. (a) PL spectrum of QD1 in the 10 μm ring at 5 K. (b) Temperature dependent measurement from 5 to 50 K showing the detuning of the QD wavelength at 925.56 nm from the ring cavity mode. The intensity drops significantly when the emission peak is off-resonance with the cavity mode with increasing temperature. (c) PL spectrum of QD1 in the 10 μm ring at 35 K. (d) Time-resolved PL measurements taken at 5 and 35 K from QD1 and corresponding single exponential fitting showing the lifetime increases from 100 ± 1.4 ps to 1.32 ± 0.05 ns, further proving the detuning of QD-cavity coupling with increasing temperature.

To prove QD-cavity coupling, temperature dependent measurement is performed on QD1 of the 10 μm ring. Figure 6.13(a) is the PL spectrum of QD1 measured under 1200 g/mm grating, showing a dominant peak at 925.6 nm and other two peaks from different states of the QD at 921.1 and 929.0 nm, respectively. Figure 6.13(b) is PL map from 5 to 50 K showing the detuning of the QD wavelength at 925.56 nm from the ring cavity mode. The intensity drops significantly when the emission peak is off-resonance with the cavity mode with increasing temperature as shown in the 35 K PL spectrum in (c). Further time-resolved PL measurements taken at 5 and 35 K from QD1 and corresponding single exponential fitting showing the lifetime

increases from 100 ± 1.4 ps to 1.32 ± 0.05 ns, further proving the detuning of QD-cavity coupling with increasing temperature.

6.5. FDTD simulation

FDTD simulations were performed using the Lumerical 3D Electromagnetic Simulator commercial solver package to calculate the Purcell factor and Q-factor of the ring. The Purcell factor is obtained from the ratio of the dipole power emitted in the structure to that emitted in the vacuum, representing the spontaneous emission enhancement. However, ring is a high Q factor cavity thus the electromagnetic field cannot completely decay from the simulation in a reasonable time. Therefore, the Q-factor of the cavity mode is determined from the slope of the envelope of the time-domain field decay signal using the formula,

$$Q = \frac{-2\pi f_R \log_{10}(e)}{2m} \quad (6.7)$$

where f_R is the resonant frequency of the mode, and m is the slope of the decay in electromagnetic field.

The section below discusses the derivation of the Q factor in equation (6.7). From Chapter 6.2.3, we know that the Q factor is defined as,

$$Q = \frac{\omega_r}{FWHM} \quad (6.8)$$

where ω_r is the resonance frequency,

$$\omega_r = 2\pi f_R \quad (6.9)$$

and FWHM is the full width half maximum of the resonance peak. The time domain signal of the resonance is described by

$$E_{(t)} = e^{-r(\alpha - i\omega_r)} v_t \quad (6.10)$$

where α is the decay constant. Using Fourier transform,

$$|E_{(\omega)}|^2 = \frac{1}{\alpha^2 + (\omega - \omega_r)^2} \quad (6.11)$$

The max value of $|E_{(\omega)}|^2$ occurs when $\omega = \omega_r$. Also, when $\omega = \omega_r + \alpha$ or $\omega = \omega_r - \alpha$, the half max frequency $|E_{(\omega)}|^2 = \frac{1}{\alpha^2} \times \frac{1}{2}$. Therefore, $FWHM = 2\alpha$. Substituting this value into the original Q formula and solving α gives,

$$\alpha = \frac{\omega_r}{2Q} \quad (6.12)$$

Now we have connected the decay constant α to Q . Next is to determine the slope of the time signal decay and connect to Q . Taking the log of the time signal to make the envelope a linear function, the following equation is obtained.

$$\log_{10}(|E(t)|) = \frac{-\omega_r t}{2Q} \log_{10}(e) = mt \quad (6.13)$$

where m is the slope of the log of the time signal envelope. Thus, solving Q , we get

$$Q = \frac{-\omega_r \log_{10}(e)}{2m} \quad (6.14)$$

as in equation (6.7).

Whispering-gallery mode identification

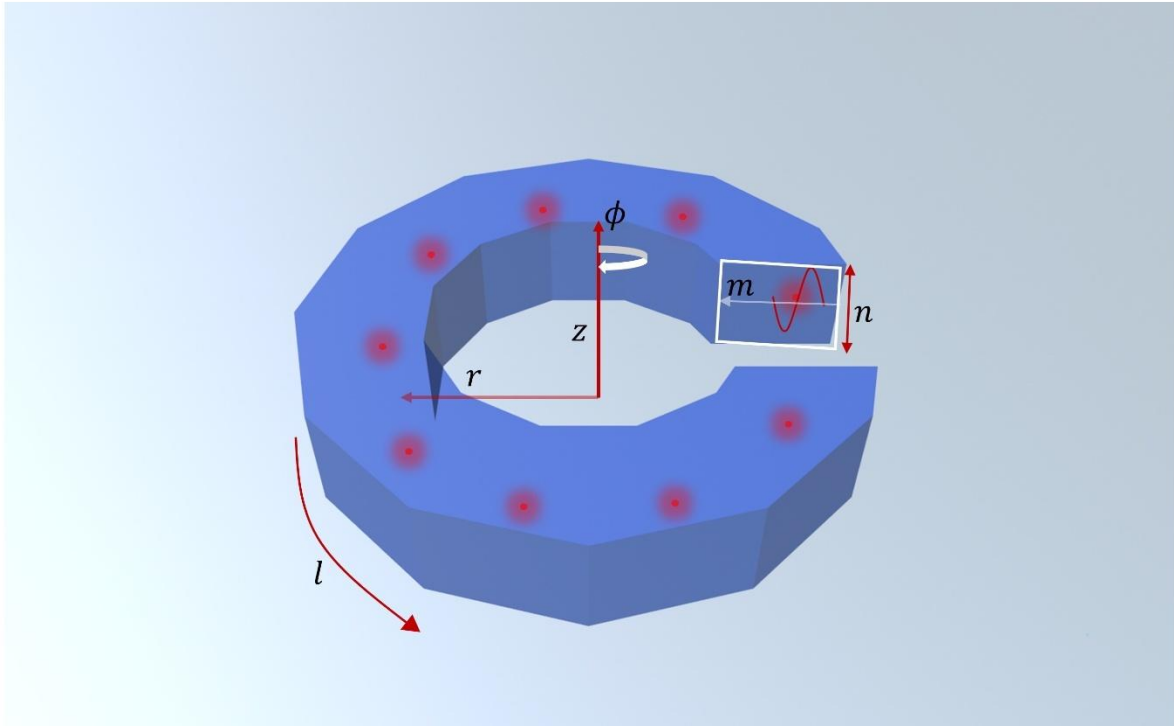


Figure 6.14. WGM identification in polar coordinates. The mode numbers l , m , and n are used to represent the number of electric field maxima along the azimuthal direction $\hat{\phi}$, radial direction $\hat{r}$, and vertical direction $\hat{z}$, respectively. WGMs are named on the basis of their field polarisation followed by the mode numbers l, m, n .

6.5.1. Optical simulation of the 4 μm ring

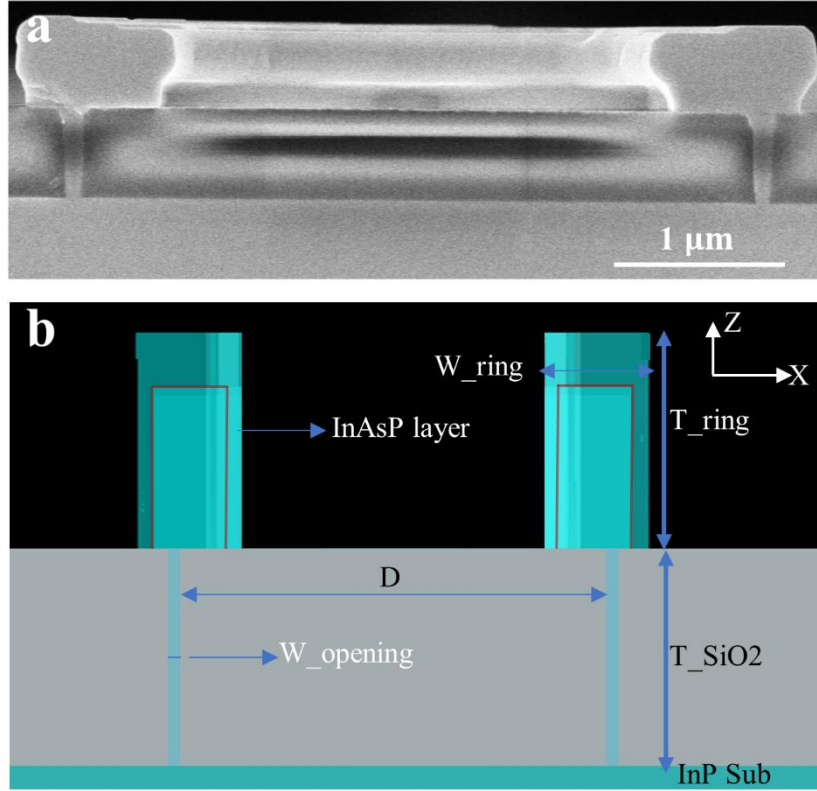


Figure 6.15 (a) Cross-section SEM image of the 4 μm ring after capping of the QDs. (b) Schematic showing the cross-section and various dimensions of the micro-ring used in the simulation.

In this simulation, a mesh size of $10\text{ nm} \times 10\text{ nm} \times 10\text{ nm}$ is used with a perfectly matched layer boundary. The QD emitter is simulated by putting a dipole source with transverse-electric (TE) mode and a wavelength from 900 to 1000 nm. Figure 6.15 (a) shows the cross-section SEM image of the 4 μm ring after capping of the QDs. Figure 6.15(b) is the corresponding simulation design of XZ plane view of the structure. The parameters are based on our experimental results: $T_{\text{SiO}_2} = 440\text{ nm}$, $T_{\text{ring}} = 400\text{ nm}$, $W_{\text{opening}} = 90\text{ nm}$, $W_{\text{ring}} = 800\text{ nm}$, $D = 4\text{ }\mu\text{m}$. The refractive index of InP and SiO₂ are 3.456 and 1.45, respectively. We incorporate a thin InAsP layer as indicated by the red lines with refractive index of 3.6 to better simulate the real structure, which represent As-P adatoms exchange during the introduction of AsH₃ and PH₃ at the crystallisation stage of the QDs. The InAsP layer thickness is set to 10 nm as it is the lowest mesh cell we can simulate.

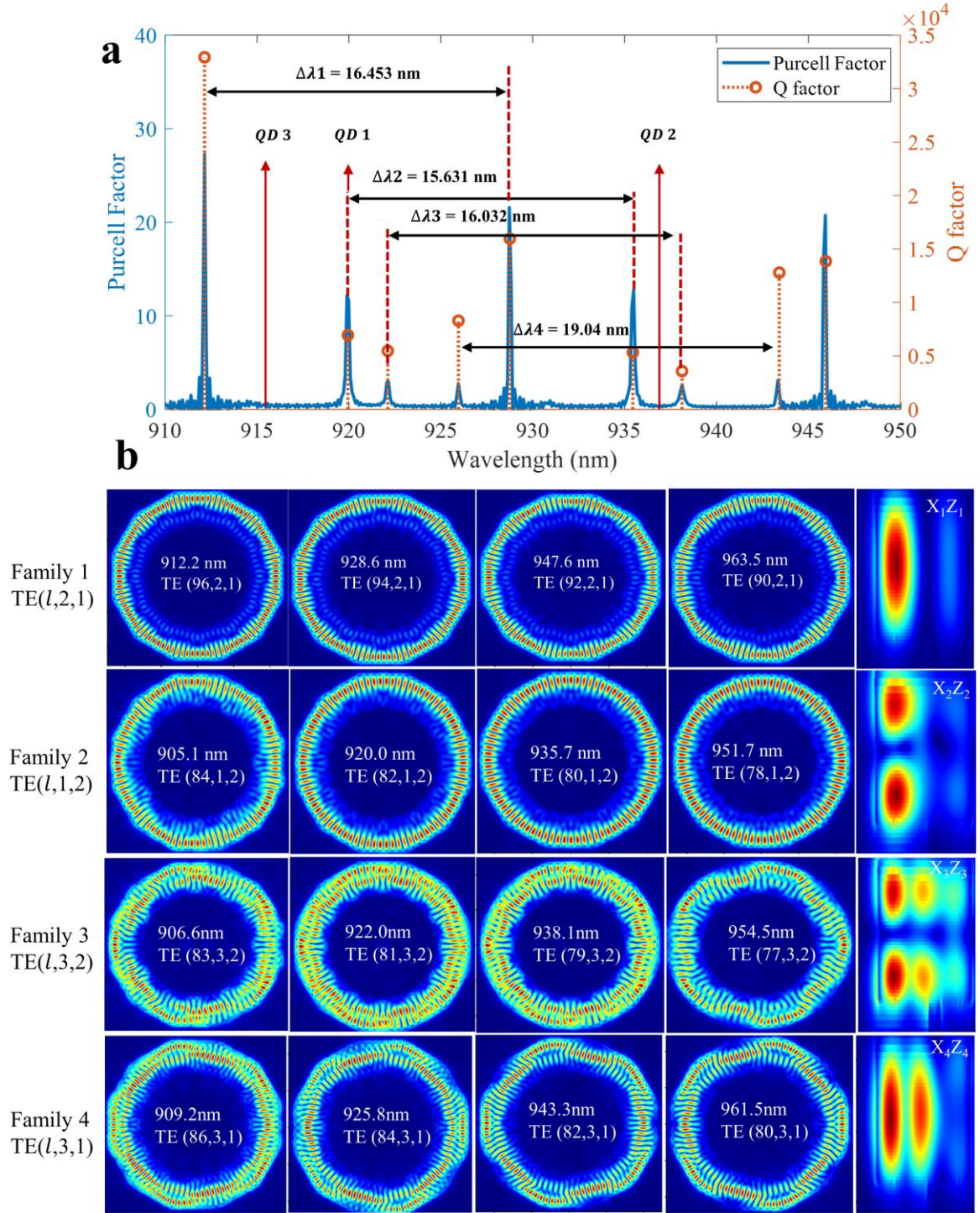


Figure 6.16. FDTD simulation results of the 4 μm ring. (a) Simulated Purcell and Q-factors of the ring using dimensions obtained from SEM results, showing four WGM family with different free spectrum range, the three red arrows represent the three QD emission peak positions of the 4 μm ring. (b) Simulated XY and XZ plane electrical field intensity profiles for four representative WGMs of each family at four adjacent wavelengths.

Figure 6.16(a) shows the simulated Purcell enhancement and Q-factors of the ring, where the three arrows indicate the major peak positions of the three QDs (QD1-3) from Figure 6.10(b). The simulation results show that QD 1 is indeed coupled to a cavity mode with a Q-factor of 6940, resulting in a Purcell factor of 12. In addition, the ring supports four types of WGMs with free spectrum range of $\Delta\lambda_1 = 16.453 \text{ nm}$, $\Delta\lambda_2 = 15.63 \text{ nm}$, $\Delta\lambda_3 = 16.032 \text{ nm}$, $\Delta\lambda_4 = 19.04 \text{ nm}$, respectively. Figure 6.16(b) presents the simulated XY and XZ electric field intensity profiles of the four WGM families (TE ($l,2,1$), TE($l,1,2$), TE($l,3,2$), TE($l,3,1$), respectively, where l represents the electric field maxima at azimuthal direction.) at four adjacent wavelengths. A clear trend is observed in which the number of field maxima decreases by two between successive modes, corresponding to a reduction in mode order. Despite these differences in the XY plane, all modes share a similar electric field distribution in the XZ plane, as illustrated in the last column. The simulation proves the 920 nm peak at QD 1 coupled to TE (82, 1, 2) mode in WGM family 2 of the ring. WGM identification in polar coordinates are shown in Figure 6.14. The mode numbers l , m , and n are used to represent the number of electric field maxima along the azimuthal direction $\hat{\phi}$, radial direction $\hat{r}$, and vertical direction $\hat{z}$, respectively. WGMs are named on the basis of their field polarization followed by the mode numbers l , m , n , respectively ^[296].

6.5.2. Optical simulation of the 10 μm ring

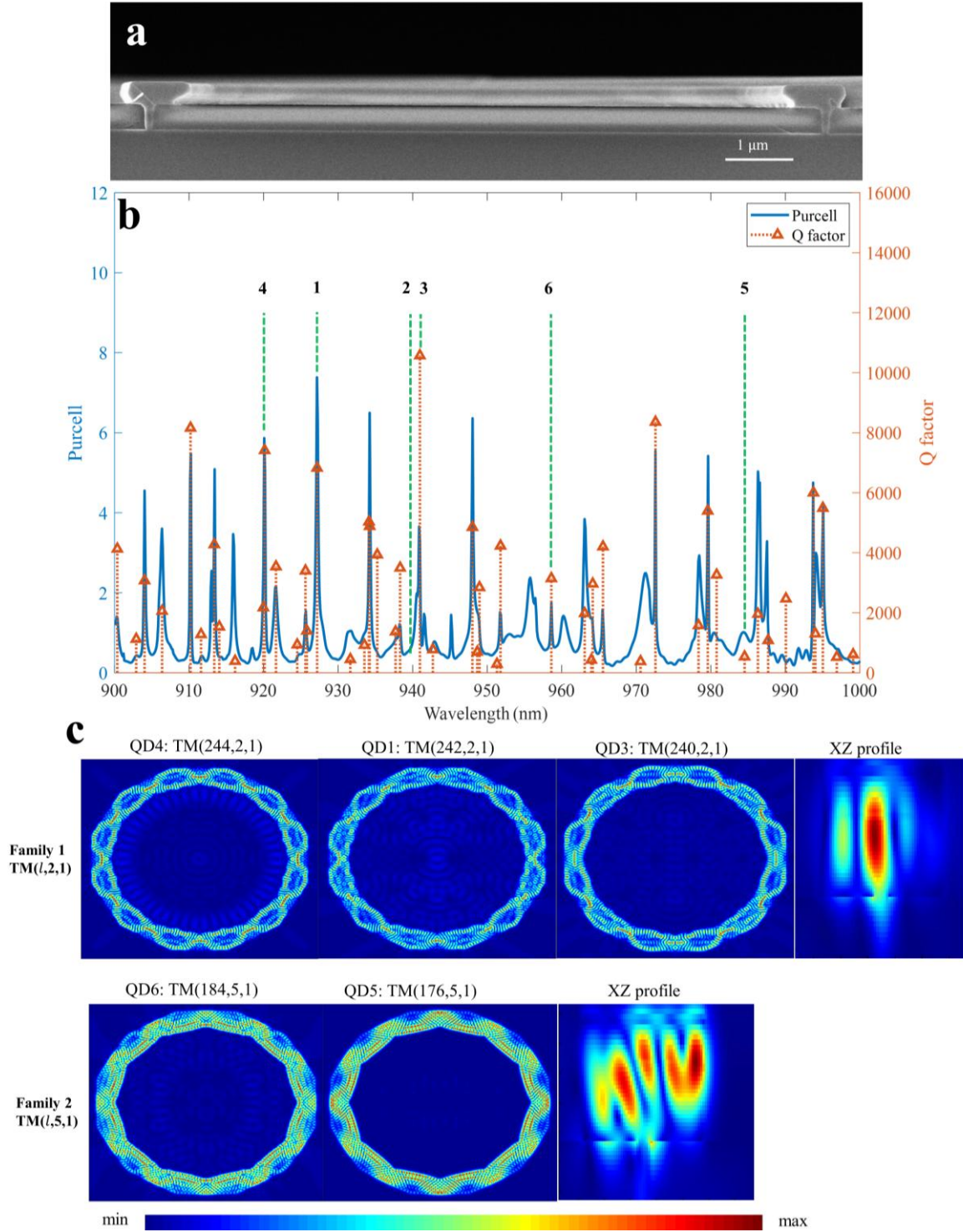


Figure 6.17. FDTD simulation results of the 10 μm ring (a) Cross-section SEM image of the 10 μm ring after capping of the QDs. (b) Simulated Purcell factor and Q-factor of the 10 μm ring. The green dash lines are the peak positions of QD 1-6. (c) Electric field intensity profiles in the XY and XZ plane of the two WGM families.

The simulation conditions of the 10 μm ring are the same as those of the 4 μm ring. The ring parameters are set based on experimental measurements: $T_{\text{SiO}_2} = 440 \text{ nm}$, $T_{\text{ring}} = 400 \text{ nm}$, $W_{\text{opening}} = 90 \text{ nm}$, $W_{\text{ring}} = 900 \text{ nm}$, $D = 10 \mu\text{m}$ from the cross-section SEM image in Figure 6.17 (a). The simulation time for this larger ring is demanding with fine mesh setting of $10 \text{ nm} \times 10 \text{ nm} \times 10 \text{ nm}$. Therefore, we only simulated a short period of (10000 fs) to check corresponding wavelength of the modes by the combination of Purcell and Q-factors, as the mode wavelength would not change with longer simulation times. Figure 6.17(b) shows the simulated Purcell factor and Q-factor, the green dash lines are the peak positions of the QDs from Figure 6.11 (c), with the QD 1, 3, 4, 5, and 6 being coupled to the modes of the rings. Figure 6.17(c) shows the XY plane and XZ plane electrical field intensity profiles of the coupled QDs, where QD 1,3 and 4 are coupled to the same WGM families of TM ($l,2,1$) while QD 5 and 6 are coupled to another WGM family of TM($l,5,1$).

6.6. Summary

In this chapter, we further applied droplet epitaxy to incorporate InAsP QDs in bottom-up InP micro-rings to couple their emissions with the whispering gallery modes of the ring resonators for single photon source applications. We optimised the width of InP rings prior to In droplet deposition to achieve better spatial coupling. By systematically optimising the growth parameters (deposition temperature, TMIn flow rate and deposition time) of In droplet deposition, we achieved only a few droplets in the 4 μm ring with a TMIn flow rate of $2 \times 10^{-6} \text{ mol/min}$ and a deposition time of 4 s at the temperature of 400 $^{\circ}\text{C}$. A smooth ring surface with a roughness of 0.17 nm was achieved by increasing the InP capping shell growth temperature to 625 $^{\circ}\text{C}$ with a V/III ratio of 350, resulting in high Q-factor of the rings (~ 3000 of 4 μm ring, ~ 4000 of 10 μm ring). By coupling the QDs into the 4 μm ring, we showed single photon emission with a $g^{(2)}(0)$ value of 0.241 and a short radiation lifetime of 70 ± 1.8 ps. FDTD simulations further confirmed the coupling of single photon emission into the TE (82,1,2) mode with a simulated Purcell factor of 12 and a Q-factor of 6940. Moreover, we achieved different densities of QDs in the two types of rings. The larger ring of 10 μm supports more high Q-factor WGMs giving higher chances for more QDs to be coupled, resulting in multiple Purcell-enhanced single photon emissions from the ring at different wavelengths.

The bottom-up growth approach provides good reproducibility in micro-ring geometry and surface morphology, enabling consistent optical properties across different devices. The main limitation instead arises from the QDs incorporated within the rings. Although the droplet density can be controlled through the growth conditions, deterministic formation of a single QD at the optimal position within the cavity mode remains challenging. Variations in QD size and composition also result in variations in emission wavelength, limiting deterministic spectral matching between individual QDs and specific cavity modes. Further improvements in selective-area growth, droplet nucleation control and ring geometry are therefore required to achieve reproducible and identical single-QD–cavity coupling across large device arrays. Addressing these limitations will be important for extending this bottom-up approach towards scalable on-chip quantum photonic circuits.

CHAPTER 7

7. Conclusion and Outlook

7.1 Conclusions

This thesis investigates a technique for the deterministic positioning of single QDs within two types of bottom-up grown NW and micro-ring optical cavities. The QD density and cavity quality are optimised under different growth parameters for both electrically and optically driven single-photon emission.

Chapters 1 to 3 present the research background, motivation, and current challenges associated with III–V semiconductor QD-based single-photon sources. The theoretical background, including material growth mechanisms, light–matter interactions in optical cavities, and the definition and characterisation of single photons, are discussed to provide a working understanding of this work. In addition, the experimental techniques employed throughout the thesis—such as selective area epitaxy, device fabrication, and material and device characterisation—are introduced.

Chapter 4 introduces In droplet epitaxy combined with selective area epitaxy of InP NW arrays to deterministically position single QDs within the NWs. A single droplet at the tip of each NW is achieved by reducing the NW diameter, height, and TMIn deposition time. A uniform QD-in-NW array is obtained by optimising the V/III ratio, pitch size, and shell-layer growth temperature. Optical characterisation of the array demonstrates scalable and bright single-photon emission, with most QDs coupled to the dominant optical mode of the NW.

Chapter 5 demonstrates electrically driven single-photon emission by deterministically positioning an InAs QD within a p–i–n InP NW diode. The core–shell diode structure is designed to facilitate efficient electrical contacts and strong vertical emission. Guided by photoluminescence measurements and numerical simulations, the QD emission is shown to couple to the Fabry–Pérot mode of the NW, yielding a high brightness of up to one million counts per second and a single-photon purity with $g^{(2)}(0)=0.21$. Finite difference time domain simulations confirm that the coupled mode corresponds to the TM_{01} mode, with a simulated

Purcell factor of 4.3. Furthermore, the device exhibits long-term stability in both emission wavelength and photon count rate under continuous current injection for over one hour.

Chapter 6 extends the droplet epitaxy approach to high Q-factor, bottom-up grown InP ring resonators to achieve coupling between QD emission and whispering gallery modes. A low QD density within 4- μm rings is realised by reducing the growth temperature, TMIn flow rate, and deposition time. The InP capping layer is optimised to achieve a low surface roughness of 0.17 nm, enabling high Q-factor rings (~ 3000 for a 4- μm ring and ~ 4000 for a 10- μm ring). Emission from a single QD is coupled to the whispering gallery mode of the ring, resulting in an ultrafast radiative lifetime of 70 ± 1.8 ps and a single-photon purity of $g^{(2)}(0)=0.24$. Finite difference time domain simulation proved the coupling to the TE (82,1,2) mode with a simulated Purcell factor of 12. Increasing the ring diameter to 10 μm supports more closely spaced high-Q WGMs, increasing the probability of QD–optical mode coupling.

7.2 Outlook

In this thesis, we have demonstrated the success of combining droplet epitaxy with selective area epitaxy for site-control of QD incorporation within an optical cavity for single photon emitters. Based on the insights gained from this study, three potential research directions can be pursued in future work to further broaden the capability and functionality QD-in-cavity system: 1). Electrically driven single photon emitter in telecommunication wavelengths; 2). On-chip integration with a waveguide; 3). Coupling of QD with circular Bragg grating to enhance vertical collection efficiency.

7.2.1 Electrically driven single photon emitter in the telecommunication wavelengths

Realisation of electrically driven single emitters in the telecommunication wavelength range is of great importance for long-distance quantum communication ^[297], as it coincides with the low-loss transmission window of optical fiber ^[298]. Building on the successful realisation of electrically driven single-photon emission from InAs QDs in InP NW diodes (~ 880 nm) presented in Chapter 5, achieving electrically driven single emitters in the telecommunication wavelength range is highly feasible. This can be accomplished by optimising the QDs to obtain an appropriate size and composition. Figure 7.1 shows the 5 K PL spectra of samples using above-band excitation with a 522 nm pulsed laser. The QDs were grown under different In

droplet deposition times of 0.3, 0.4, 0.5, and 0.6 s. By increasing deposition time, the droplet size is increased, with the diameter/height evolving from 43.2/11.6 nm, to 52.2/15.5 nm, to 65.7/21.6 nm, and finally to 76.6/22.2 nm, as shown in the inset scanning electron microscopy (SEM) images. This increase in droplet size leads to larger QD sizes upon crystallisation with the group V precursor (AsH_3) and consequently induces a red-shift of the emission wavelength toward the telecommunication band. We note here that it is also possible to introduce PH_3 during crystallisation of the In droplets to form InAsP QDs, which gives us an additional degree of control to the emission wavelength by changing their composition. The NW growth and device fabrication processes would be very similar to what have been investigated and developed in this dissertation.

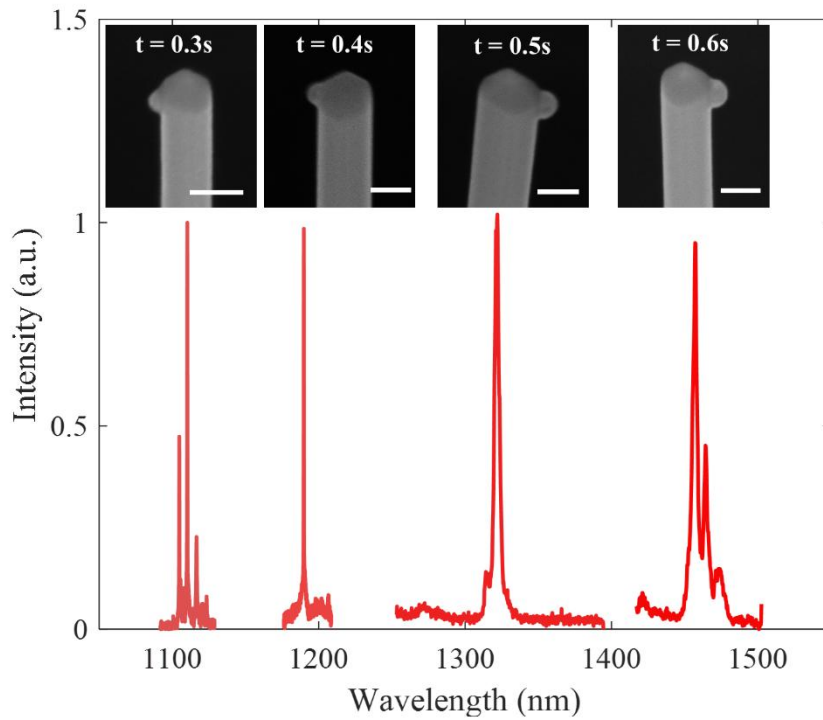


Figure 7.1 Engineering of the QD wavelength toward the telecom-band. PL spectra of four different QDs measured at 5 K with QD grown under different In droplet deposition time of 0.3, 0.4, 0.5, and 0.6 s. Insets show the corresponding droplet size. The increase of droplet deposition time leads to a larger QD during the crystallisation process and hence a red-shift of the emission wavelength. All scale bars are 50 nm.

7.2.2 On-chip integration with a waveguide

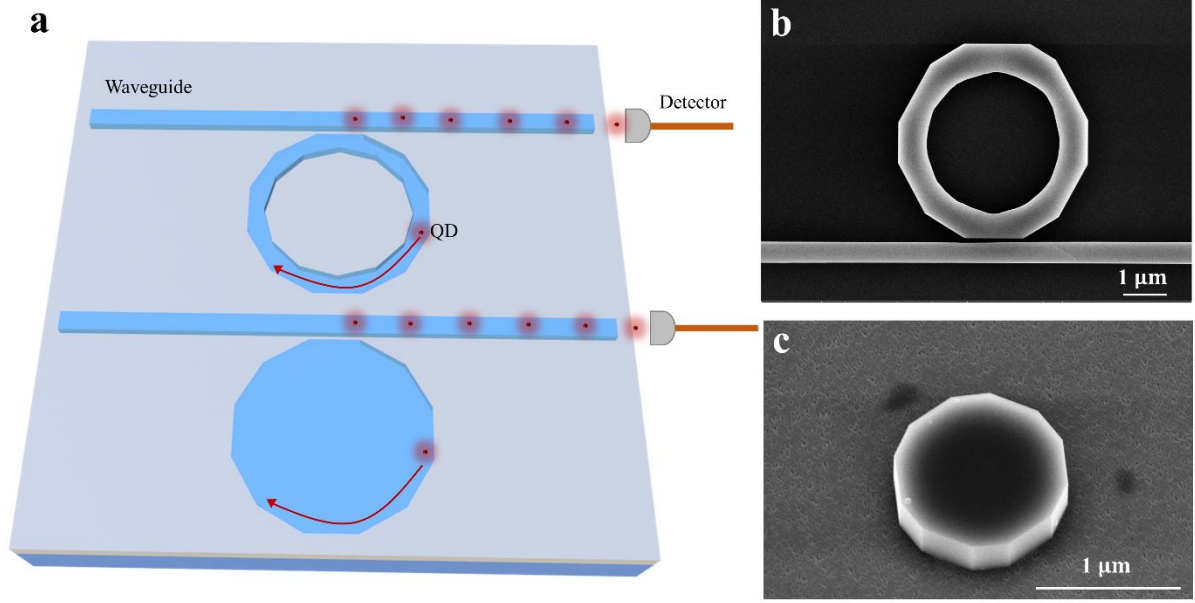


Figure 7.2. Bottom-up micro-ring/disk on-chip integration with waveguide. (a) Illustration of on-chip integration of an optical waveguide with QD in a micro-ring/disk structure. (b) SEM image of the bottom-up InP micro-ring integrated with a waveguide. (c) SEM image of bottom-up InP micro-disk. Note that a few droplets can be seen in the image.

In Chapter 6, we demonstrated the coupling of QD emission to the whispering gallery modes (WGMs) of bottom-up InP micro-rings. However, the light collection efficiency from the top is very low, as the optical field is primarily confined within the in-plane direction. Therefore, future research could focus on the on-chip integration of QDs in ring or disk structures with waveguides to achieve efficient single-photon routing within photonic circuits, as illustrated in Figure 7.2(a). The coupling between the WGMs of the ring and the guided modes of the bus waveguide can enable high coupling efficiency and controlled photon transfer on-chip ^[299]. Such integration represents an important step toward scalable and fully integrated quantum photonic platforms ^[300]. Figures 7.2(b–c) present SEM images of the preliminary work carried out within this thesis on bottom-up micro-rings with waveguides and micro-disks, respectively, demonstrating the feasibility of directly growing both structures simultaneously. Nevertheless, achieving deterministic single-photon emission within a photonic network requires careful design and engineering of the coupling gap, ring diameter, and QD position to ensure efficient coupling and stable performance for chip-based quantum communication and computation architectures.

7.2.3 Coupling of QD in bottom-up with circular Bragg grating to enhance vertical collection efficiency

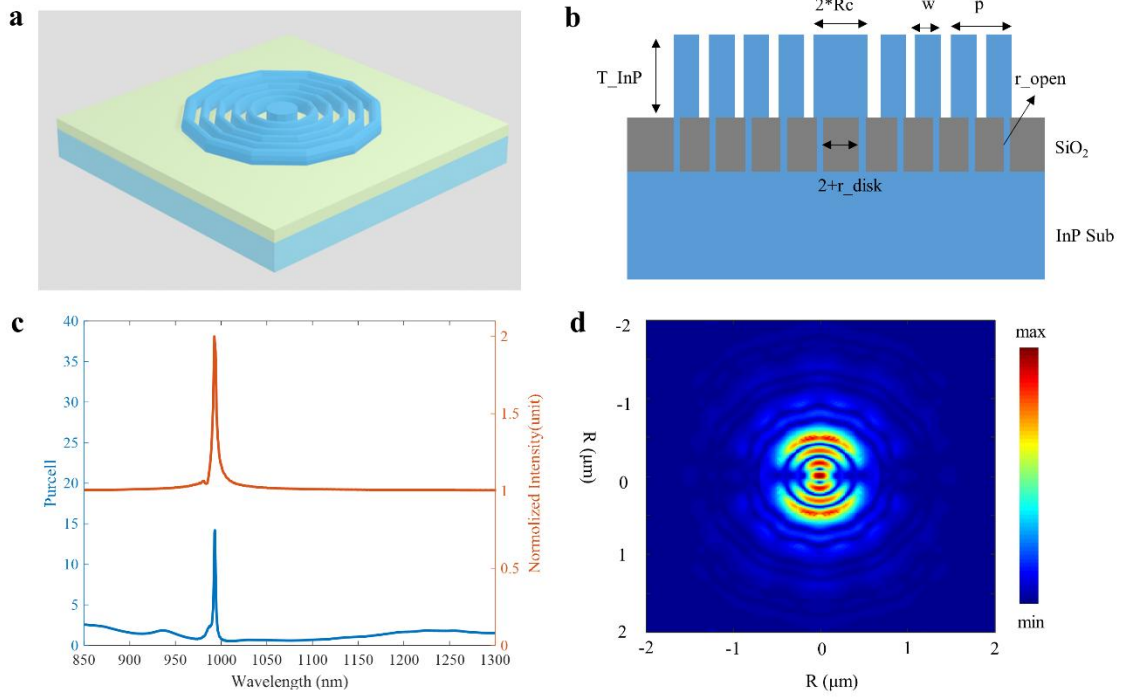


Figure 7.3. Design bottom-up circular Bragg grating to enhance vertical extraction efficiency. (a) Schematic of the structure: a central disk surrounding by multiply concentric decagonal InP rings. (b) Cross-sectional view of the structure with different geometrical parameters. (c) Simulation results of Purcell factor and spectrum. (d) Simulated electrical field profile at plane view of the disk at the resonance peak position.

Another promising research direction is the integration of single QDs with bottom-up grown circular Bragg grating cavities to achieve enhanced vertical light extraction ^[301]. Unlike whispering gallery modes that mainly confine light in the in-plane direction, circular Bragg grating structures provide a photonic environment that redirects the emitted photons vertically ^[241, 302], significantly improving collection efficiency. This structure typically consists of a central disk with QDs and multiple concentric rings served as gratings as illustrated in Figure 7.3 (a). All previously reported structures of this type are based on top-down etching, which involves multiple independent processes, including QD growth and positioning, cavity design and fabrication, and substrate transfer ^[303-305].

In contrast, in the bottom-up fabrication approach, the QD and the circular Bragg grating structure can be grown within the same run. Figure 7.3(b) shows a cross-sectional view of the design, where multiple concentric ring patterns are defined through an SiO₂ mask, and the central disk has a radius of r_{disk} ; Other parameters such as height of the InP layer (T_{InP}), disk radius (R_c), grating width (w), grating period (p), gap between gratings ($p-w$), and total number of grating rings. By designing these parameters properly, both the far-field emission pattern and the Purcell factor can be enhanced substantially ^[306]. Figure 7.3(c) presents the preliminary simulation results, showing a resonance peak at approximately 990 nm with a simulated Purcell factor of 15, while the in-plane electric field profile at the peak wavelength, shown in Figure 7.3(d), indicates strong confinement within the central disk region. Yet, further simulation studies of light extraction efficiency need to be done.

From a growth perspective, the key challenge lies in achieving deterministic droplet positioning at the centre of the disk, since droplets preferentially nucleate at high-energy sites along the ring edges. To overcome this, a central ring structure was designed to promote droplet formation at the centre, with the aim of forming a disk via increased lateral growth during the QD capping process. Figure 7.4 presents our preliminary growth results: (a) shows an SEM image of the concentric rings after etching, which exhibit uniform widths; (b) shows the In droplet formed on the InP Bragg grating structure after growth, with the magnified image of the central ring in (c). The droplet deposition conditions are the same as those used in Chapter 6 for low-density QDs, and the droplets tend to form at the edges of the rings. Figure 7.4(d) shows the final SEM image after capping the QDs. Nevertheless, the lateral growth rate of the central ring is not high enough to form a disk, further studies such as simulations, and pattern design need to be conducted. However, the smooth and uniform morphology after capping demonstrates the feasibility of this approach. The successful realisation of bottom-up circular Bragg grating structures would open new approach for high light extraction efficiency single emitters.

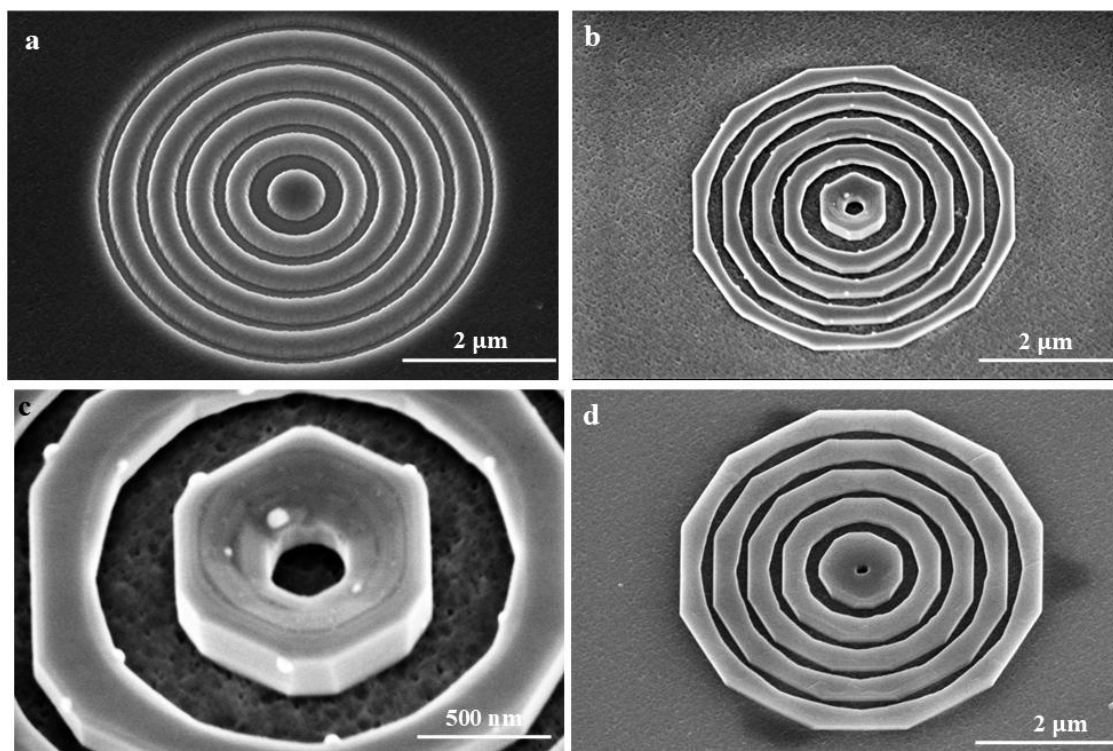


Figure 7.4. Preliminary results of bottom-up circular Bragg grating structure. SEM of (a) the pattern with concentric rings after etching, (b) In droplets formed on the concentric rings, (c) the magnified central ring region in (b), (d) the concentric rings after capping the QDs.

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