D3.1: Epitaxy of QDs with FSS<30μeV

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Abstract	We developed an epitaxial growth of quantum dots (QD) processing high in-plane symmetry. The QDs emit in the telecom C-band. Polarisation-dependent micro-photoluminescence measurements revealed fine structure splitting below 30μeV for neutral excitonic complexes.
Keywords	Epitaxy, quantum Dots, fine structure splitting

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EXECUTIVE SUMMARY

The QPIC1550 project aims to develop a QD-based device emitting polarization-entangled photon pairs at 1550 nm. Polarization entangled photon pairs can be generated in semiconductor quantum dots (QDs) through the radiative biexciton-exciton cascade. In this process, a biexciton (XX), consisting of two electron-hole pairs, undergoes a radiative recombination, emitting a photon and leaving behind an intermediate exciton (X). The exciton subsequently recombines, emitting a second photon. Ideally, the polarization states of the emitted photons remain indistinguishable, leading to polarization-entangled photon pairs. However, the presence of fine structure splitting (FSS), which arises due to electron-hole exchange interactions in QDs with asymmetric confinement potential of an electron-hole pair, that lifts the degeneracy of the intermediate exciton states. This splitting introduces distinguishability in the cascade's decay paths, reducing the degree of entanglement. For high-fidelity entangled photon emission, the FSS should be smaller than the emission linewidth, ensuring that the two decay paths remain spectrally indistinguishable.

Thus the aim is to develop QD epitaxy enabling a symmetric potential for exciton or biexciton states.

We developed InAs QDs on InP with tunable density and telecom C-band emission. QDs formed on pyramidal nano-posts were grown using a two-step epitaxial process, with geometry controlled via growth parameters. For optical studies, QDs were capped with a 20 nm InP layer.

Micro-photoluminescence (μPL) revealed sharp excitonic emission. Polarization-dependent μPL enabled FSS extraction, showing values below 30 μeV, though limited by the spectrometer's 70 μeV resolution. When FSS falls below this threshold, it manifests as a systematic shift in emission rather than distinct peaks. High-quality polarization data allows detecting FSS below spectrometer resolution, consistent with previous studies.

For linewidths under 200 μeV, FSS below 30 μeV is detected, though resolving below this requires advanced techniques. To achieve higher precision, we plan to implement a Fabry-Perot interferometer at DTU to resolve FSS down to a few μeV, supporting entangled photon generation.

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ABBREVIATIONS

FSS	Fine Structure Splitting
QD	Quantum Dot
μPL	Micro-photoluminescence
FWHM	Full-width half-maximum
MOVPE	Metal-organic vapor phase epitaxy
X	Neutral exciton
XX	Biexciton
CX	Charged exciton
SK	Stranski-Krastanov
DE	Droplet epitaxy

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1 SECTION: MOTIVATION

The main objective of the QPIC1550 project is to develop a universal integrated photonics platform based on silicon nitride (SiN) photonic circuitry, achieved through the integration of indium phosphide (InP), quantum dots (QDs), and indium gallium arsenide (InGaAs) detectors. This innovative platform aims to facilitate quantum photonic applications in the telecom C-band. By enabling on-demand single-photon generation, the production of entangled photon pairs, and efficient single-photon detection within a compact and scalable photonic architecture, QPIC1550 has the potential to advance the field of quantum photonics significantly.

A crucial step toward realizing a source of polarization-entangled photons at 1550 nm is the development of a QD-based emitter that exploits the biexciton (XX) – exciton (X) cascade recombination process, depicted in Figure 1.

An exciton (X) is a bound state of an electron and a hole in a semiconductor, held together by Coulomb attraction. It forms when an electron is excited from the valence band to the conduction band, leaving behind a positively charged hole. A biexciton (XX) is a bound state of two excitons, comprising two electrons and two holes. It behaves as a quasiparticle analogous to a molecular state of excitons.

The biexciton-exciton (XX-X) cascade relaxation can be a radiative decay process that involves two-photon emissions. Initially, one of the electron-hole pairs recombines to emit a photon, represented in the emission spectrum as a linearly polarized XX line. After the XX recombination, one electron-hole pair is left in the QD (X) and eventually recombines, leading to the observation of the X line with linear polarization but orthogonal to the XX. The linear polarization of X and XX spectral lines is generated by the electron-hole exchange interaction that relies on the relation between electron and hole spin states within the exciton or biexciton. Consequently, two possible XX-X recombination pathways (Fig.1, blue and red) are possible, which are defined by two possible spin state configurations within XX and X. In ideal QDs with symmetrical confinement potential, spectral identification of two orthogonally and linearly-polarized photons in the XX-X pair cannot distinguish the XX-X recombination pathway. The so-called which-path information is erased, leading to polarization-entanglement of XX and X states. However, experimentally, it is never the case, and every asymmetry in confinement potential results in a difference between the excitonic energy levels, which is called fine structure splitting (FSS). The FSS introduces spectral distinguishability between two XX-X recombination pathways and thus nullifies the entanglement.

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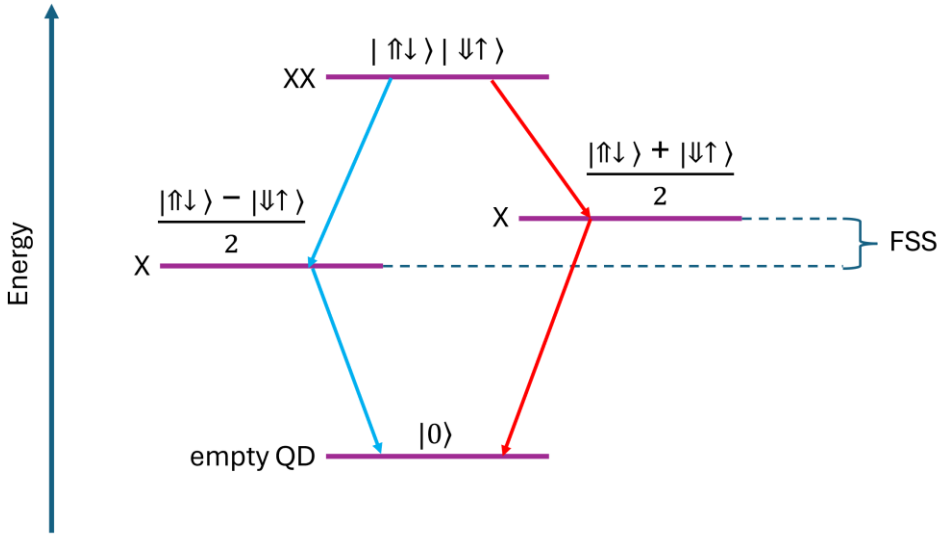

Figure 1 Scheme of the XX-X cascade. ⇑ is a spin of a hole and ↑ is a spin of an electron inside a QD. Transitions observed in the horizontal (perpendicular) linear polarization are marked as red (blue) arrows.

The value of the FSS can be experimentally obtained directly from the photoluminescence spectrum of a QD as a value of splitting between the lower- and higher-energetic lines either of X or XX, as shown in Figure 2. One can try to minimize the FSS by modifying the potential of the QD, for example, by application of the strain [1]. However, applying external strain to QD can influence the FSS only by a few μeV. In order to increase the chance of full cancellation of the FSS, the technological process has to be optimized to obtain QDs with a highly reduced initial value of FSS.

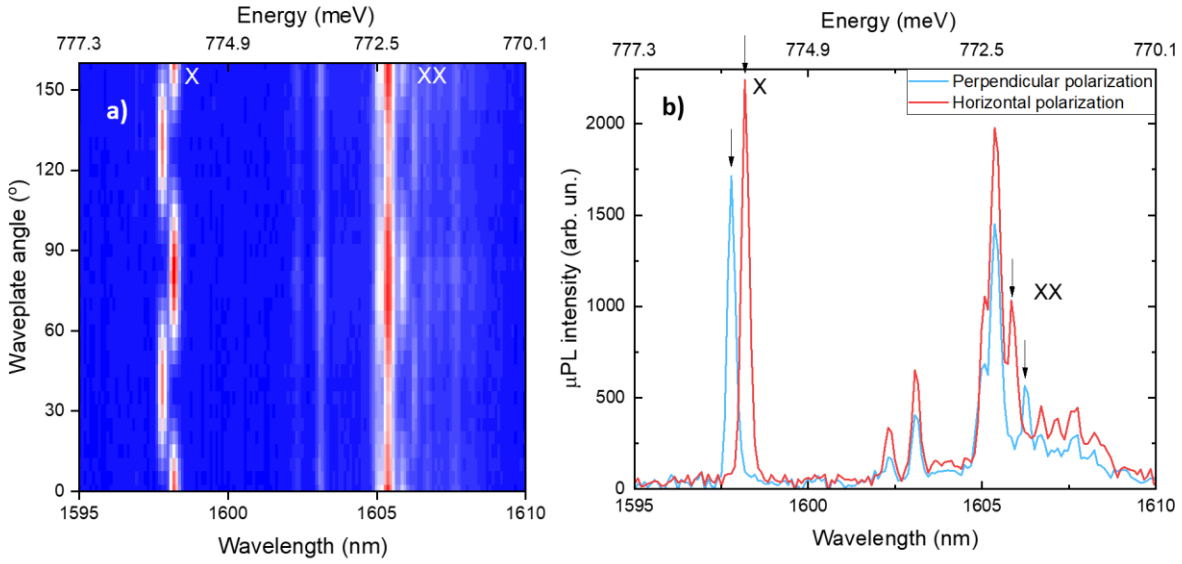

Figure 2 Experimental measurement of the FSS of an asymmetric QD. a) map of the μPL as a function of the waveplate angle. b) single spectra for orthogonal linear polarisation. The colors blue and red match the transitions marked in Figure 1. FSS of this QD is 0.1

The requirement for the FSS is, therefore, to be below the Fourier-limited emission linewidth. Consequently, the optimization of QD structural properties is a primary task and a fundamental component of Work Package 3.

QDs grown in strain-driven mechanisms, such as Stranski-Krastanov (SK) growth mode, tend to exhibit elongation due to the anisotropic nature of atoms' surface diffusion during the QD nucleation and growth process, contributing to non-zero FSS. To overcome this limitation, we investigate different epitaxial methods for QD synthesis that are less dependent on anisotropic surface diffusion and less influenced by strain during the growth process. We explore the potential of droplet epitaxy (DE), a technique that enables the fabrication of highly symmetric QDs with enhanced control over their structural and electronic properties. This approach facilitates the reduction of FSS, thereby potentially improving the fidelity of polarization entanglement.

In preliminary work, the DTU team demonstrated FSS from a droplet epitaxy InAs/InP QDs operating at telecom wavelength ~50μeV [2]. This was accomplished by fine-tuning the epitaxy process to create a symmetric confining potential for exciton and biexciton states.

Another approach we investigate is site-controlled epitaxy, in which predefined nucleation sites are created by patterning the substrate to dictate where epitaxial growth occurs. This method allows for precise spatial positioning of QDs while also enabling control over the surface diffusion of adatoms during QD nucleation. By engineering the substrate pattern, we can manipulate adatom migration and aggregation, potentially leading to more symmetric QDs with reduced FSS.

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2 SECTION: TECHNOLOGY

We developed a controlled epitaxial growth process for InAs QDs on InP, achieving tunable surface density and emission in the telecom C-band. Metal-organic vapor phase epitaxy (MOVPE) was employed as the growth technique due to its excellent scalability, high uniformity, and compatibility with industrial semiconductor fabrication, making it a promising approach for future large-scale quantum photonic applications.

The QD structures were grown using a low-pressure D125 MOVPE reactor with hydrogen (H_2) as the carrier gas. Trimethylindium (TMIn), trimethyl aluminium (TMAl) and trimethylgalium (TMGa) were used as the group III precursors, while arsine (AsH_3) and phosphine (PH_3) served as group V precursors. The growth was carried out on InP (100)-oriented substrates. The flow and, consequently, ratio of precursor gases were controlled using mass flow controllers, ensuring precise and consistent delivery of TMIn, AsH_3, and PH_3 throughout the epitaxial growth process. This level of control is critical for maintaining the desired sub-monolayer precision of the crystal growth rate. The growth temperature was controlled using an emissivity-corrected pyrometer, ensuring accurate monitoring and regulation of the substrate temperature throughout the epitaxial process. This method allows for consistent and reproducible growth conditions, which are crucial for the formation of high-quality QDs.

The QDs were selectively formed on the apex of pyramidal-shaped nano-posts (Fig. 3), ensuring precise spatial localization and uniform growth. The fabrication began with the deposition of 20 nm thick InAlAs layer on an InP substrate. The structure was then removed from the epitaxial reactor and patterned using high-resolution electron beam lithography, followed by dry etching to define the nano openings with 50-150 nm diameter. This structured template was subsequently reintroduced into the epitaxial reactor, where InP pyramids were grown, with self-assembled InAs QDs forming on their apexes.

The geometry and size of both the pyramids and QDs were finely tuned by adjusting key MOVPE growth parameters, including substrate temperature, growth rate, and the ratio between precursor fluxes. The use of MOVPE provided a high degree of reproducibility and wafer-scale integration potential, crucial for transitioning from laboratory research to industrial implementation.

For further optical characterization, the QDs were encapsulated with an additional 20 nm InP cap layer; encapsulating is required to enable carrier confinement, protect them from environmental degradation, and maintain high optical quality.

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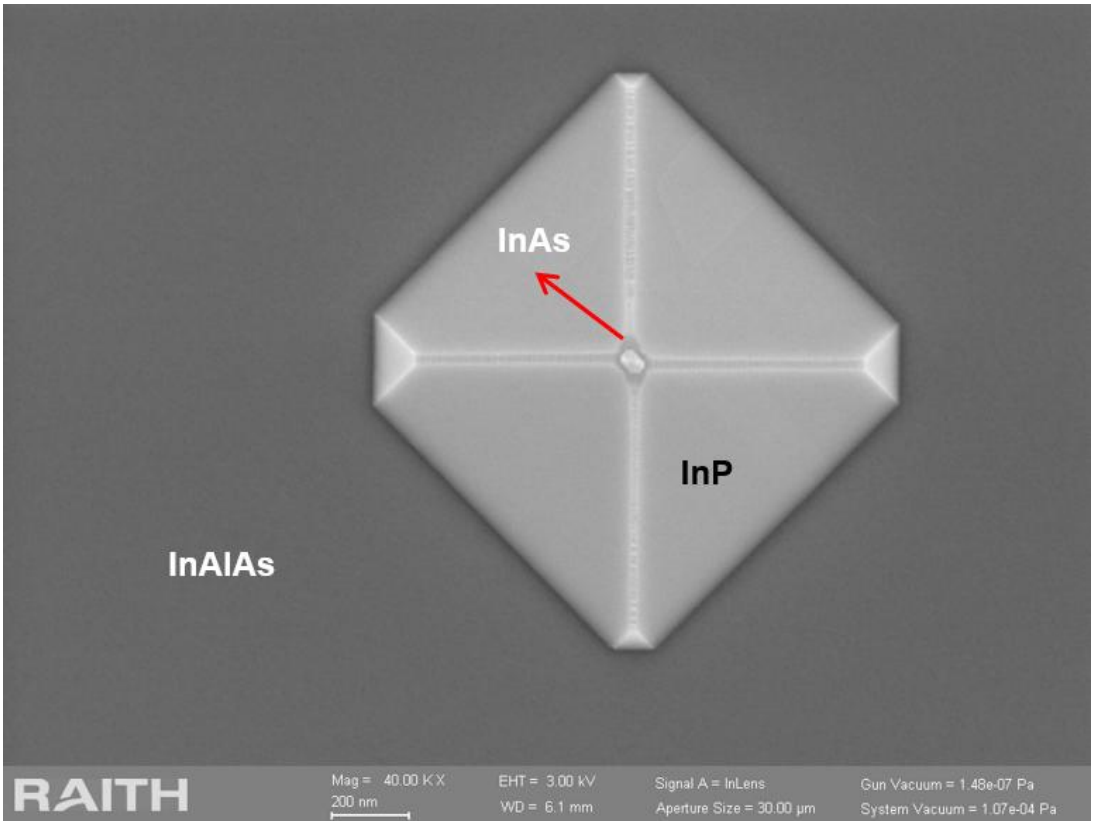

Figure 3 Plan-view scanning electron microscope image of a single uncapped InAs QD on top of InP pyramidal nanopost.

3 SECTION: EXPERIMENTAL RESULTS

Micro-photoluminescence (μPL) experiments were performed in the setup illustrated in Figure 4 The sample was cooled down in the cryostat down to 5 K and excited with a continuous-wave 640 nm laser line. Photons from the XX-X cascade were detected with linear polarization resolution using a linear polarizer of a high extinction ratio and a movable λ/2 waveplate in front of the cryostat. Changing the position of the waveplate enabled scanning of the polarization dependence of the μPL spectra, as presented in Figures 5a and 5b.

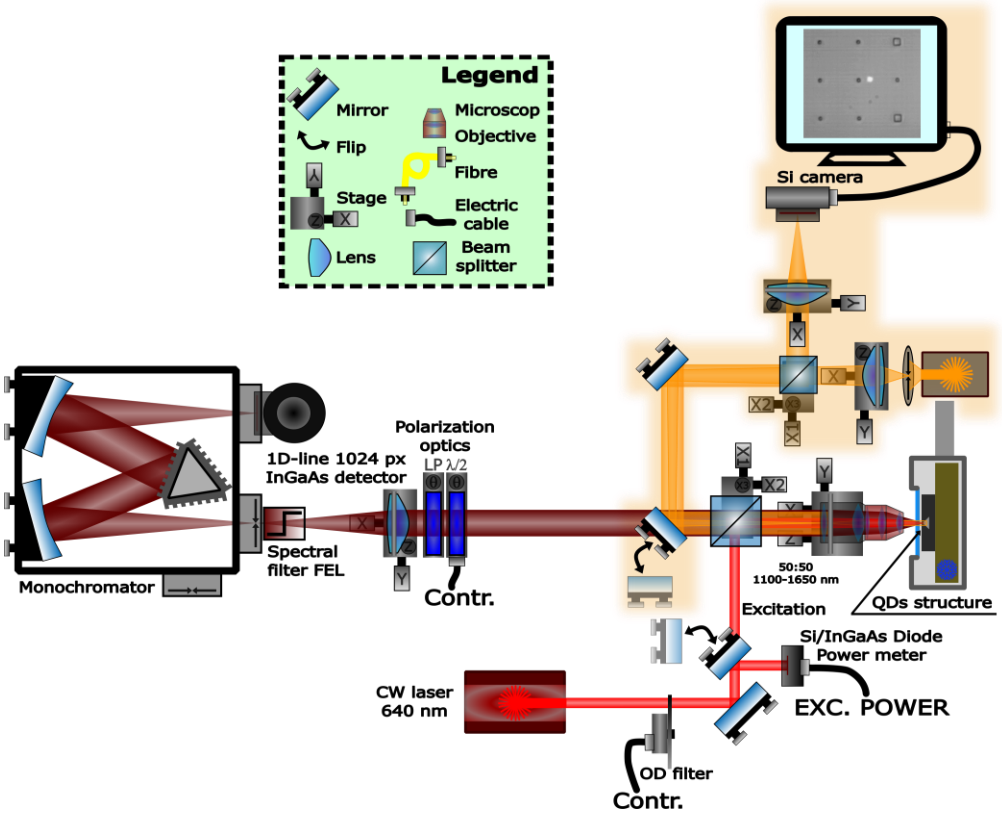

Figure 4 Scheme of the experimental setup for the polarization-dependent μPL measurement

The experiment focused on individual QDs, revealing sharp emission lines corresponding to various excitonic complexes confined within them (Fig. 5). These distinct spectral features confirm the QDs' high optical quality and suitability for quantum photonic applications.

A key parameter of interest in such systems is fine structure splitting (FSS), which arises from the anisotropic electron-hole exchange interaction that relies on particles' spin state. The FSS directly impacts the degree of polarization entanglement in photon pairs emitted via the biexciton-exciton cascade recombination. To accurately determine the FSS, we performed polarization-dependent μPL measurements, analyzing shifts in the emission peak position as a function of the polarization angle. Our results indicate that for neutral excitonic complexes, the FSS remains below 30 μeV (Fig. 5b, Table 1), which is promising for polarization-entangled photon state generation.

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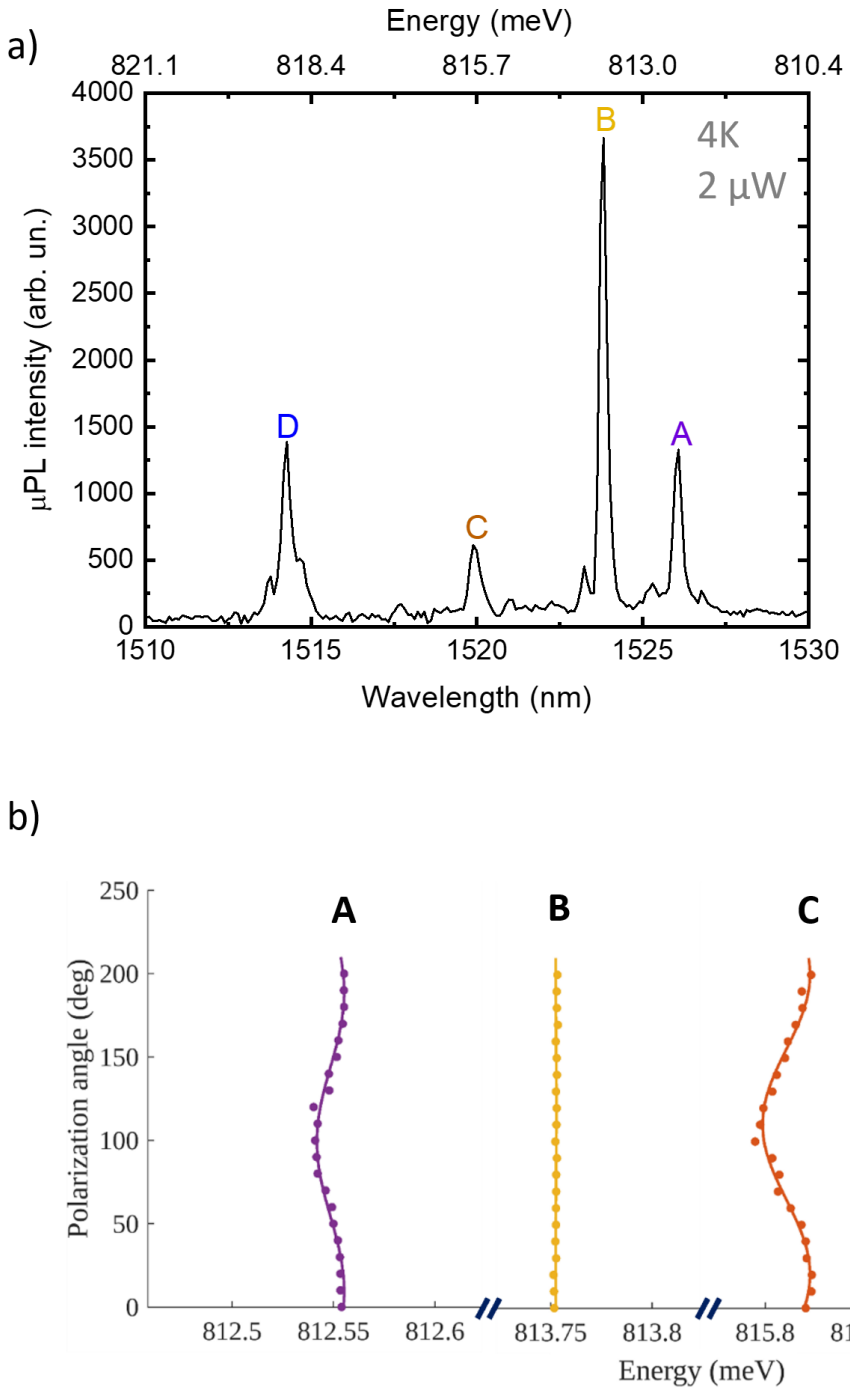

Figure 5 Representative photoluminescence measurements are taken from an individual QD: a) lines A-D, corresponding to different excitonic complexes; b) polarization-dependent measurements of corresponding excitonic complexes.

Table 1 Parameters of A-d lines from Figure 5.

	Energy of the line (meV)	Wavelength of the line (nm)	Estimated FSS (μeV)	Excitonic Complex
A	812.549	1526.061	13.42±1.8	Another QD
B	813.753	1523.804	-	CX*
C	815.810	1519.958	23.32±2.4	XX
D	818.883	1514.261	-19.85±2.9	X

**The charged exciton, CX, is a spin singlet state, so the anisotropic exchange interaction is not applicable as well as FSS. The oscillations that can be observed in the μPL spectra for the CX line are related to the heavy-hole-light-hole mixing in the CX complex*

However, a fundamental limitation in these measurements is the spectrometer's resolution, which in our case is 70 μeV. When the FSS falls below this threshold, the exciton's two bright states cannot be distinctly resolved as separate spectral lines. This limitation is further compounded by the emission linewidth (full width at half maximum, FWHM), which must remain sufficiently narrow to observe clear spectral splitting. A broader emission linewidth can obscure small FSS values, making direct measurement increasingly challenging.

Despite these constraints, polarization-dependent measurements are a viable approach for extracting FSS values, even when they fall below the spectrometer's resolution, by utilizing proper fitting tools to resolve the peaks. Instead of resolving two distinct spectral peaks, we observe a systematic shift in the central position of the Gaussian-fitted emission line as a function of polarization.

The emission spectrum was recorded as a function of the λ/2 waveplate angle. Fig. 4a illustrates the spectrum at an angle of 0°. Each spectral line is fitted using a Gaussian function, where the central value of the fit represents the emission peak position. The extracted peak positions are then plotted as a function of the waveplate angle (Fig. 4b), and a periodic function is fitted to the resulting data. In this analysis, the FSS is determined as twice the amplitude of the fitted sinusoidal curve (Table 1). This method estimates the FSS, even when it falls below the spectrometer's resolution.

The observed spectral shift exhibits a characteristic periodic dependence, where the energy difference between the extreme values corresponds to the fine structure splitting. High-quality polarization-resolved μPL data have been shown in previous studies [3] to reveal FSS values significantly smaller than the spectrometer resolution.

These results highlight the need for advanced optical characterization techniques to further improve the accuracy of FSS measurements. Future work will focus on implementing high-resolution interferometric approaches, such as Fabry-Pérot interferometry, to push the resolution to the few μeV range. Such improvements will be crucial for accurately characterizing QDs with ultra-low FSS, enabling the development of scalable quantum light sources utilizing the XX-X polarization entanglement scheme for photonic quantum technologies.

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4 CONCLUSIONS

Based on our analysis, we estimate that for emission linewidths (FWHM) below 200 μeV, fine structure splitting (FSS) values below 30 μeV can be detected. This detection is made possible by analyzing subtle changes in the emission line shape and position, even when direct spectral resolution is limited. Our findings align with previous studies [4,5], which have demonstrated that FSS values at least five times smaller than the spectrometer resolution can still be extracted through polarization-dependent measurements.

However, achieving higher precision in FSS determination requires advanced optical techniques to resolve energy splittings well below the current spectrometer resolution. Interferometric approaches, such as Fabry-Pérot interferometry, provide a promising solution by offering ultra-high spectral resolution down to a few μeV range. This level of precision is essential for accurately characterizing QDs with ultra-low FSS, which is a critical requirement for generating high-fidelity polarization-entangled photon pairs.

In the next phase of the project, we will investigate external methods for tuning QD properties, particularly FSS, through the application of electric and strain fields. This will require the engineering of photonic cavities integrated with QDs that enable effective tuning. Furthermore, to be able to investigate the new generation of devices effectively we plan to implement a Fabry-Pérot interferometer at the DTU lab to achieve this enhanced spectral resolution. This will allow us to systematically investigate the FSS of our QDs with unprecedented accuracy, enabling a deeper understanding of the fundamental mechanisms governing exciton fine structure. By optimizing QD growth, engineering the tuneable photonic devices, and further refining characterization techniques, we aim to develop highly efficient, low-FSS quantum emitters suitable for scalable entangled photon sources. These advancements will play a key role in the realization of integrated quantum photonic technologies for secure communication, quantum computing, and photonic quantum networks.

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