Lifetimes and Quantum Efficiencies of Quantum Dots Deterministically Positioned in Photonic-Crystal Waveguides

Chu, Xiao-Liu; Pregnolato, Tommaso; Schott, Ruediger; Wieck, Andreas D.; Ludwig, Arne; Rotenberg, Nir; Lodahl, Peter

Published in:
Advanced Quantum Technologies

DOI:
10.1002/qute.202000026

Publication date:
2020

Document version
Peer reviewed version

Citation for published version (APA):
Lifetimes and quantum efficiencies of quantum dots deterministically positioned in photonic-crystal waveguides

Xiao-Liu Chu*, Tommaso Pregnolato, Rüdiger Schott, Andreas D. Wieck, Arne Ludwig, Nir Rotenberg and Peter Lodahl

Dr. Xiao-Liu Chu, Dr. Tommaso Pregnolato, Prof. Nir Rotenberg and Prof. Peter Lodahl
Center for Hybrid Quantum Networks (Hy-Q), Niels Bohr Institute, University of Copenhagen, Blegdamsvej 17, DK-2100, Copenhagen, Denmark
E-mail: xiao-liu.chu@nbi.ku.dk

Prof. Rüdiger Schott
ETH Zürich, Otto-Stern-Weg 1, 8093 Zürich, Switzerland

Prof. Andreas D. Wieck and Dr. Arne Ludwig
Lehrstuhl für Angewandte Festkörperphysik, Ruhr-Universität Bochum, Universitätsstraße 150, 44801 Bochum, Germany

Keywords: quantum nanophotonics, single-photon sources, quantum dots, nanofabrication

Interfacing single emitters and photonic nanostructures enables modifying their emission properties, such as enhancing individual decay rates or controlling the emission direction. To achieve full control, the single emitter must be positioned in the nanostructures deterministically. Here, spectroscopy is used to gain spectral and spatial information about individual quantum dots (QDs) in order to position each emitter in a pre-determined location in a unit cell of a photonic-crystal waveguide (PhCW). Depending on the spatial and spectral positioning within the structured nanophotonic mode, the quantum dot emission is observed to either be suppressed or enhanced. These results represent an important step towards unlocking the full potential of nanophotonic systems and will be crucial to the creation of complex multi-emitter quantum photonic circuits.

1. Introduction

Photonic-crystal waveguides (PhCWs) control the flow of light and reshape the electromagnetic landscape at the nanoscale, a powerful combination in the context of quantum optics. By confining light to a small region, and slowing down its velocity, PhCWs enhance light-matter interactions enabling, for example, efficient photon-pair generation by spontaneous parametric down conversion. These same effects allow PhCWs to act as an
efficient interface to solid-state quantum emitters such as quantum dots (QDs)\textsuperscript{[3]} or defect centers in diamond.\textsuperscript{[4]} In fact, PhCWs also strongly suppress emission into free-space,\textsuperscript{[3, 5]} meaning that they can almost perfectly couple the emitters to guided photon modes.\textsuperscript{[6]}

The physics of emitter-photon coupling is captured by the Purcell factor, which quantifies the enhancement (or suppression) of emission into the PhCW, $\Gamma_{wg}$, relative to the emission rate in bulk $\Gamma_B$.\textsuperscript{[7]} For PhCWs, the frequency $\omega$ and position $r$ dependent Purcell factor can be written as\textsuperscript{[8, 9]}

$$F_P(\omega, r) = \frac{3\pi a c^2}{\omega^2 \sqrt{\varepsilon(\omega, r)}} n_g(\omega) |d^* \cdot e(\omega, r)|^2,$$

where $a$ is the lattice constant of the PhCW, $c$ is the speed of light in vacuum, and $\varepsilon$ is the dielectric constant of the material in which the emitter is embedded. The group index $n_g$ sets the velocity of the photonic mode according to $v_g = c/n_g$ and linearly enhances the emission rate, as shown in Equation 1. Ideally, $n_g$ diverges at the photonic bandedge, as shown in the calculation presented in Figure 1. In practice, unavoidable fabrication imperfections break the discrete symmetry of the photonic crystal, leading to unwanted scattering losses and capping the value of $n_g$ at $\approx 500$.\textsuperscript{[10]}

![Figure 1.](image-url) (Position and wavelength dependent optical properties of a PhCW. The calculated group index $n_g$ diverges at the bandedge (detuning $\Delta \lambda = 0$), greatly enhancing light-matter interactions in the region. Insets: spatial map of the normalized amplitude of the in-plane electric field distributions for fast ($n_g = 5$) and slow ($n_g = 50$) light modes in a unit cell. The)
nanoscopic nature of the features in these light fields is evident as they are the same size as the holes of the PhCW (white circles, radius \( r = 77 \text{ nm} \)).

Finally, the Purcell factor depends on the overlap between the transition dipole moment \( \mathbf{d} \) (located at \( r \) and oscillating with angular frequency \( \omega \)) and the electric field of the photonic mode \( \mathbf{e} (\omega, \mathbf{r}) \), which is normalized such that the integrated electrical energy over the unit cell is unity, at that position. The nanoscale geometry of PhCWs results in a complex and finely structured \( \mathbf{e} (\omega, \mathbf{r}) \) that is known to contain regions of various possible field polarizations. \cite{11} Examples of the in-plane field distributions \( e_x (\omega, \mathbf{r}) \) and \( e_y (\omega, \mathbf{r}) \), calculated for different \( n_g \) modes, are shown in Figure 1. Here, the structure of the light fields, with nanoscopic features, is readily visible.

In total, the ability to engineer \( n_g \) and \( e (\omega, \mathbf{r}) \) means it is possible to design PhCWs that enhance, \cite{12} suppress \cite{13} or even directionally couple emission, \cite{14,15} but only if the emitter can be precisely positioned. \cite{16} High quality indium arsenide (InAs) QDs with excellent coherence properties \cite{17} that produce indistinguishable photons with high count rates \cite{18,19} demanded by emerging quantum technologies are normally grown via molecular-beam epitaxy using the Stranski-Krastanov method. \cite{3} The growth process results in self-assembled and randomly distributed QDs, both spatially throughout each wafer, and in their emission wavelength. Unlocking the full power of the PhCW platform, as represented by Equation 1, has therefore not been possible, and typical experiments involved the fabrication of many samples followed by a search for those that contain an emitter in a favorable position.

Although early pioneering theoretical work on photonic crystals showed that the emission dynamics of a single emitter will have very strong position dependence as expressed by the local density of optical states, \cite{20,21} it has been a challenge to observe experimentally. Here, we take the important step by mapping the dynamics of single-photon emission of QDs deterministically placed in PhCWs with sub-wavelength precision. Furthermore, we show that the nanostructure is capable of improving the radiative properties of the emitter by controlling the radiative part of the decay rate. Spectroscopic and lifetime measurements of the QDs, both before and after nanofabrication, allow us to quantify the effect of the PhCW on the emission. We observe a spatial dependence of the emission properties and a significant improvement of the quantum efficiency of the QDs. Moreover, we demonstrate that this can be done on relatively large scales, here integrating 35 QDs into waveguides, each within an area of only \( 0.254 \times 0.33 \mu \text{m}^2 \) of a photonic chip.

2. Results and Discussion

2.1. Deterministic Integration of Quantum Dots in Photonic-Crystal Waveguides

The first step of precise placement of QDs within PhCWs is to locate them and characterize their behavior in bulk samples. The sample is a doped GaAs membrane that forms a p-i-n
diode, which stabilizes and tunes the emission wavelength of the QDs. 

Whilst scanning electron microscope (SEM)- or atomic force microscope (AFM)-based methods have been utilized to find the QD position with great precision, they do not provide the spectral knowledge of the emitters. Alternative approaches such as cathodoluminescence (CL) or micro-photoluminesence (mPL) bridge this gap, with CL-based approaches, characterization and nanofabrication is performed in the same machine, meaning that alignment markers are not needed and the chip need never be exposed to the environment. In contrast, mPL separates these two steps, allowing for parallel detection and nanofabrication as would be required in large-scale production. To locate the QDs, we therefore choose to use the mPL protocol outlined in ref. [30] and correlate images of QD emission to those of a global reference frame (here a grid of gold crosses, see Figure 2a, to determine the absolute position of each QD. All measurements unless noted otherwise, are taken at a bias voltage of 0.3 V.

Using this procedure, we locate each QD with an accuracy of 9.2 nm relative to global alignment markers (see Supplementary materials for additional information), similar to that of...
other mPL protocols\textsuperscript{[28, 31–34]}. The resulting QD-PhCW alignment accuracy consists of a randomly distributed standard deviation of 33 nm and a small systematic shift of only 1 nm\textsuperscript{[30]}, which we attribute to our mPL imaging method. We then measure the emission spectra of the brightest QDs and use this information to precisely design a PhCW around the QD, as shown in Figure 2b. Here, the PhCW hole radii range from 71 to 76 nm and the lattice constant from 233 to 247 nm, nominally placing the bandedge within 10 nm of the emission wavelength (near 915 nm).

Before nanofabrication, we carry out time-resolved measurements on the neutral excitons, $X^0$, of 13 QDs in bulk, which we determine from voltage spectra maps. The measured decay curves are well-modelled as single-exponential decays despite the fact that InAs QDs have two optically bright states, $|X\rangle_b$ and $|Y\rangle_b$, corresponding to orthogonally oriented, in-plane dipoles.\textsuperscript{[13]} Knowledge of the dipole orientation relative to the crystal planes of the GaAs membrane allows us to design our PhCWs such that they lie longitudinally and transversely with respect to $|X\rangle_b$ and $|Y\rangle_b$, as shown in Figure 2b.

In bulk, a single-exponential decay is not surprising, as the radiative recombination rates of the two states are nearly-identical. In fact, the QDs still behave as effective two-level systems, even in the presence of significant non-radiative recombination. In the experiment, single-exponential decay curves are observed consistently throughout the experiment both for QDs in bulk samples and in the PhCWs. This stems from the fact that significant non-radiative recombination was found in the QDs, implying that only the well-coupled dipole was observed while recombination from the weaker coupled dipole and dark excitons was suppressed\textsuperscript{[35]} (See Supplementary Information for additional details). Consequently, the QDs effectively behave as 2-level emitters with a total decay rate:

$$\Gamma_i = \gamma_{i,r} + \gamma_{i,nr},$$  \hspace{1cm} (2)
where \( i = B \) or \( wg \), depending on whether the QD is embedded in bulk or in a waveguide, respectively, and all the non-radiative processes are included in \( \gamma_{i,nr} \). Alternatively, each QD can be characterized by its quantum efficiency,

\[
QE_i = \frac{\gamma_{i,r}}{\gamma_{i,r} + \gamma_{i,nr}}
\]

which quantifies the probability that the QD will emit a photon upon excitation.

We summarize our bulk lifetime measurements as a histogram in Figure 2c. A normal distribution fit to this data (red curve) reveals a mean decay rate (\( \Gamma_B \)) of 3.9 ns\(^{-1}\) with a standard deviation of \( \pm 0.7 \) ns\(^{-1}\). This decay rate is significantly faster than the estimated radiative decay rate \( \gamma_{B,r} = 1 \) ns\(^{-1}\), which is based on both calculations and measurements on different QD samples in bulk semiconductor.\(^{[36, 37]}\) Indeed, this confirms the presence of significant non-radiative recombination, consistent with previous measurements of QDs that emit near 910 nm (rather than the more standard 945 nm). Such QDs tend to be smaller and suffer from a poor overlap between the electron and hole wavefunctions.\(^{[38]}\) Consequently, as shown in Figure 2c, we used Equation 3 to obtain \( QE_B \) that range between 0.2 and 0.4, well below the near-unity values routinely measured with the larger QDs.\(^{[13]}\)

Having located and characterized the QDs in bulk, we now fabricate 35 PhCWs with QDs equally distributed between 7 positions, as shown in Figure 3. At these positions, \( F_P \) for a transversely oriented dipole \( d_y \) varies by at maximum a factor of 4 (Figure 3a). We show an example of the final, suspended PhCW with integrated grating couplers in Figure 3b. For each waveguide, micro-photoluminescence spectroscopy measurements (see inset to Figure 4, for an example) reveal whether or not a QD was successfully interfaced with the structure. From these, we find a yield of 94% for structures with a nominal QD-surface separation > 100 nm,
and 44% for smaller separations. The latter positions are at (0, 0.5a), (0, 0.8a) and (0, 1.3a), as shown in Figure 3a, corresponding to nominal distances of 77, 49, and 31 nm from the nearest surface for \( a = 247 \text{ nm} \) and \( r = 76 \text{ nm} \). Moreover, it is likely that the effective separation is smaller due to a depletion region around the holes, where the QDs cannot be efficiently biased, \(^{30, 39}\) meaning that the observed yield is consistent with our protocol accuracy.

Figure 3. ((Deterministic integration of QDs into PhCWs (a) Designated QD positions (green crosses) within a unit cell of a PhCW, overlapped with the calculated \( F_P \) for a \( \hat{y} \)-oriented transition dipole. Also shown are the holes of the PhCW (black circles) and the 33 nm alignment uncertainty of our protocol (blue curves). (b) SEM of a PhCW with integrated grating couplers. (c) A zoomed-out SEM shows many PhCWs, each of which contains a single, positioned QD. The randomness of the PhCW positions reflect that of the QDs.))

We measure the transmission spectrum of each waveguide that contains a QD using a tunable laser, presenting an example in the inset to Figure 4 (green curve). This allows us to extract the detuning \( \Delta \lambda \) between each QD and the band edge, where the transmission drops precipitously. A histogram of \( \Delta \lambda \) for all QDs is shown in the main part of Figure 4, demonstrating that with few exceptions we are able to design and fabricate each individual PhCW such that the QDs lie within 10 nm of the band edge, where the group index \( n_g \) is changing more rapidly. We obtain a spectral accuracy of around \( \pm 1 \text{ nm} \), which we attribute to changes to the stress and strain within the GaAs membrane due to the removal of the sacrificial layer, as detailed in ref. [30]. As seen in Figure 1, each \( \Delta \lambda \) corresponds to a different \( n_g \), and hence emission enhancement factor. Although the exact curvature of this
function depends on the exact geometry of the PhCW, we give a rough indication of typical $n_g$’s for each separation in the top axis of Figure 4.

In total, these results demonstrate that we are able to precisely position QDs, both spatially and spectrally, with respect to individual PhCWs.

**Figure 4.** ((Spectral response of the QDs and the PhCWs. This is summarized in a histogram of the detuning $\Delta \lambda$ between the QD emission and the photonic band edge. Typical $n_g$ values for these detunings are given on the top axis. Inset: Example of a transmission curve and a QD emission spectrum, from which we calculate $\Delta \lambda$)

2.1. Position Dependent Decay Rate and Quantum Efficiency

We can reformulate the radiative decay rate of QDs inside PhCW (Equation 2) using the Purcell factor (Equation 1) as

$$\gamma_{wg,r}(\omega, r) = F_p(\omega, r) \gamma_{B,r} \approx A(r)n_g(\omega)\gamma_{B,r},$$

(4)

where in the last step we assume that $A(r)$, which is determined by the prefactor and the mode-dipole overlap, has only a weak wavelength dependence. This assumption is reasonable in view of Figure 1, where only minor changes to the electric field distributions were seen as
$\Delta \lambda$ changed. We are therefore able to first study the effect of $\Delta \lambda$ on the emission, before proceeding to the more complex spatial dependence.

We begin by considering two emitters situated close to the photonic band edge, where the largest decay rate enhancement is expected. We consider two QDs, QD1 and QD2, located at $(0.2a, 0)$ and $(0.4a, 0)$ in the unit cell and within 2 nm of the photonic band edge. Since QD2 lies closer to the mode maximum it should experience a slightly larger $F_P$ (c.f. Figure 3a). At 0.3 V, the QDs emit at 915.1 nm and 904.7 nm respectively, which we change by scanning the external bias voltage between 0-0.6 V (the detuning dependence can be seen in the voltage-wavelength PL spectra (Figure S2) in the supplementary material). At each detuning, we measure and fit the decay rate, again observing single-exponential dynamics as discussed above and as can be seen by the exemplary measurements presented in the inset to Figure 5. The main body of Figure 5 summarizes the dependence of the lifetime (symbols) of both QDs on their respective detuning from the bandgap. As expected, a significantly increased $\Gamma_{wg}$ is observed as the QD emission wavelength approaches the band edge, due to the increase in $n_g$.

**Figure 5.** (Photonic band-edge enhancement of $\Gamma_{wg}$). Lifetime measurements (symbols) reveal a sharp increase in the decay rate of two QDs as they are electrically tuned towards the bandedge (dashed line), well above the bulk values (solid region). The data are well-fitted using Equations 2 and 4 (solid curves). Note that relatively large uncertainty in detuning arises due to difficulty in precisely placing the band-edge of the PhCW, while the uncertainty in the decay rate is very small (within the data marker) attesting to the quality of our fits.
Inset: examples of two lifetime measurements, corresponding to the two solid symbols in the main figure.)

Although both QDs in Figure 5 display the same wavelength dependent trend, they differ in their details. Decay rates of QD1, for example, can be tuned between 2.4 ns$^{-1}$ and 6.1 ns$^{-1}$, meaning that it begins slower and ends up faster than the average $\Gamma_B$ (shaded region). This can be understood by considering that QD1 is located at position $(0.2a, 0)$, where we do not expect an enhancement of emission due to the Purcell factor $F_P$ (Figure 3a). This suppression is then overcome through electrical tuning towards the band edge, and a corresponding increase of $n_g$ and $F_P$. In contrast, QD2 is positioned close to the field maximum, where its radiative decay rate is enhanced. We also calculate the $QE_{\text{wg}}$ of these two QDs, using the radiative and non-radiative decay rates extracted from the fits of Equations 2 and 4 to the data of Figure 5 (solid curves, see Supplementary Material for more information on the fitting process). From these, we calculate that when QD1 is 2 nm away from the band edge $QE_{\text{wg}} = 0.54 \pm 0.03$, which can be increased to $0.76 \pm 0.05$ as $\Delta \lambda \rightarrow 0$. Similarly, for QD2 the $QE_{\text{wg}}$ increases from $0.78 \pm 0.05$ to $0.86 \pm 0.06$ as its emission wavelength approaches the bandedge. These results, and the agreement between our measurement and model, confirm that we observe strong wavelength-dependent slow-light emission enhancement. [12, 40, 41]

We proceed to measure the decay rate $\Gamma_{\text{wg}}$ for the rest of the QDs in the PhCWs, and group them by their position in the unit cell. We normalize each data point to the $n_g$ experienced by that QD, and present the position dependent decay rates in Figure 6. Here, the mean and standard deviations are given by the black circles and error bars. For comparison, we also show the calculated normalized decay rate (at $n_g = 20$) for both transverse ($\hat{y}$-oriented) and longitudinal ($\hat{x}$-oriented) dipoles, in the absence of non-radiative recombination (i.e. unity QE), using red and blue shaded regions that reflect the positional uncertainty of the QDs. In this figure we also show the branching ratio $\gamma_{\text{wg},r} (\hat{y}\text{-dipole}) / \gamma_{\text{wg},r} (\hat{x}\text{-dipole})$ for each nominal
emitter position (black curve, right axis), demonstrating that the relative emission rates of the two bright excitons can be controlled through a careful positioning of the QD. For most of the chosen positions the branching ratio is typically larger than five, while the remaining positions \((y/a \geq 0.5)\) exhibited very small count rates and therefore a small signal to noise ratio.

**Figure 6.** ((Normalized decay rate \(\Gamma_{\text{wg}} / (n_g \Gamma_B)\) of QDs located at different positions within a PhCW unit cell. The QDs are located at (a) \(y = 0\) and shifted along \(x\) or (b) \(x = 0.5a\) and shifted along \(y\), as shown in Figure 3a. The mean values for the QD decay rates can be seen as red circles and the black squares are the mean values. The shaded regions correspond to the calculated normalized decay rates of the \(\hat{x}\)-oriented (blue) and \(\hat{y}\)-oriented (red) dipoles, and reflect the 1σ (dark colors) and 2σ (light colors) positional uncertainty of our protocol. The calculated branching ratio between the two dipoles, at each nominal position for \(n_g = 20\), is also given (black curve, right axis).))

Qualitatively, we observe that the average normalized decay rate of the QDs placed along the center of the waveguides (Figure 6a) increase as they are positioned closer to the mode maximum (i.e. \(x\) increases to 0.5\(a\)). In contrast, the average normalized decay rate decreases as we position the QD away from the center, increasing \(y\) (Figure 6b). Excitingly, we are still able to observe emission from QDs positioned only 60 nm, and even 30 nm, away from the hole edge (at \(y = 0.8a\) and 1.3\(a\), respectively), even though a local depletion of carriers near surfaces effectively increases the region within which we cannot activate these emitters. \(^{30, 39}\)
Additional effects that have been observed within 15 nm of the surface, such as trapped surface states that increase the non-radiative decay rate and tunneling out of the QD, could further alter its optical properties \cite{29,42}. It is also clear from Figure 6 that the measured normalized decay rates are lower than what is predicted by the calculations, shown as the shaded region. This is because in the theory we only consider the ideal case with unity QE. The addition of non-radiative recombination, which is unaffected by the Purcell factor $F_p$, effectively reduces the influence of the waveguide on the emitter. This results in a flattening and lowering of the calculated position-dependent decay rate curves of Figure 6b, as can be seen in the Supplementary materials, in agreement with the measurements.

In fact, by considering the group of QDs in each position separately, we can extract information about their radiative and non-radiative decay rates and thus their $QE_{wg}$. Two examples of this procedure are presented in Figure 7, in (a) for QDs located at $(0.5a, 0.5a)$ where we expect a strong suppression of emission, and in (b) at $(0.5a, 0)$ at the mode maximum (c.f. Figure 3a).
Figure 7. ((The decay rate $\Gamma_{wg}$ of QDs located at (a) $(0.5a, 0.5a)$ and (b) $(0.5a, 0)$ as a function of their respective detuning $\Delta \lambda$ (bottom axis) or respective $n_g$ (top axis). For each group, Equations 2 and 4 are used to fit the data, holding $\Gamma_{wg}/n_g$ constant for all QDs and allowing $\gamma_{wg,nr}$ to vary (values given in plot). From the fit parameters we calculate $\text{QE}_{wg}$ finding QDs whose emission is suppressed (red text), normal values for QDs in bulk (yellow text), or enhanced (green text), due to slow-light interactions.))

We assume that all QDs at the same position have an identical $A(r)$, and all the data for QDs at the same position can therefore be simultaneously modelled using Equations 2 and 4, where only $n_g$ and the non-radiative rate $\Gamma_{wg,nr}$ are allowed to vary. In Figure 7 we show the results of this fit, where the value of $\Gamma_{wg,nr}$ that defines each curve is given. We extract both the radiative
and non-radiative decay rates from the fits, which allow us to calculate $Q E_{w g}$ of each QD using Equation 3. As expected, the radiative emission of the QD located at $(0.5a, 0.5a)$ (Figure 7a) and well away from the band edge is suppressed, resulting in a low $Q E_{w g} = 0.11 \pm 0.01$. Decreasing the detuning, and operating in the region where $n_g = 25 - 35$, compensates for this suppression. For these QDs, we measure a $Q E_{w g}$ that ranges between 0.21 and 0.23, commensurate with bulk $Q E$ values (c.f. Figure 2c), even if these QDs have larger $\Gamma_{w g, n r}$. Here, we expect a low branching ratio and hence a biexponential decay, but do not observe it due to the large non-radiative rate $\Gamma_{w g, n r}$. We observe a similar trend for QDs located at the mode maximum at $(0.5a, 0)$ (Figure 7b). Here, for a QD located well away from the band edge where $n_g = 8$, and which has a large $\Gamma_{w g, n r}$, we find a low $Q E_{w g} = 0.13$. Increasing the $n_g \approx 15 - 20$ recovers bulk $Q E_B$, with values of 0.19-0.31 measured. Further increase of $n_g$ to beyond 25 results in a large enhancement of $\Gamma_{w g, r}$, and measured $Q E_{w g}$ values of 0.45 and 0.72, well beyond what we saw for bulk QDs.

2. Conclusions

In this article, we use a photoluminescence alignment procedure to experimentally study both the temporal response and quantum efficiency of QDs deterministically distributed throughout a unit cell of a PhCW. We encounter a low $Q E$ due to the use of non-ideal QDs as already seen in bulk and show that emission can be further suppressed due to interaction with the highly structured photonic mode. This suppression can be overcome by exploiting the slow-light effect of the PhCW, resulting in enhanced $Q E$. We use the position control to extract the $Q E$, which we have previously done on QD ensembles, $^{[38]}$ and now extend to single QDs. In total, these results demonstrate the power of PhCWs to control the emission of single photons, and show that we can systematically and precisely unlock this ability by pre-determining the spectral and spatial position of emitters. Using this procedure in conjunction with emitters with $Q E = 1$, we now have the toolbox to map out the spatial dependence of the local density
of states of PhCWs and other complex nanophotonic structures. By positioning the QDs at the mode maximum in the center of the unit cell, we are also able to deterministically fabricate photon-emitter interfaces with near-unity coupling efficiencies. These capabilities are critical to the large-scale fabrication of optimal single- and entangled-photon sources, respectively, and the creation of complex quantum photonic circuits, as deterministic positioning is a key method for controllably scaling up nanophotonic systems and hence enable the coupling of multiple QDs.

**Funding Information**

We gratefully acknowledge financial support from the Danish National Research Foundation (Center of Excellence “Hy-Q”), the Europe Research Council (ERC Advanced Grant “SCALE”), the European Union’s Horizon 2020 research and innovation program under the Marie Skłodowska Curie grant agreement no. 753067 (OPHOCS), Innovation Fund Denmark (Quantum Innovation Center “Qubiz”), and the Danish Research Infrastructure Grant (QUANTECH).

**Supporting Information**

See Supplement 1 for supporting content.

**Conflict of Interest**

The authors declare the following competing financial interest(s): Peter Lodahl is the founder of Sparrow Quantum. All other authors declare no conflicts of interest.

**Acknowledgements**

The authors gratefully acknowledge Sandra Ø. Madsen and Tim Schröder for her help in the construction of the optical setup and Leonardo Midolo and Zhe Liu for assistance in fabrication.

Received: ((will be filled in by the editorial staff))
Revised: ((will be filled in by the editorial staff))
Published online: ((will be filled in by the editorial staff))
References


Keywords
Quantum nanophotonics, single-photon sources, quantum dots, nanofabrication

X.-L. Chu*, T. Pregolato, R. Schott, A. D. Wieck, A. Ludwig, N. Rotenberg and P. Lodahl

The emission dynamics of quantum dots deterministically placed throughout photonic crystal waveguide unit cells are mapped. This precise spatial and spectral control unlocks the full power of nanophotonics and is an important step towards large-scale fabrication of optimal single- and entangled-photon sources, and hence the creation of complex quantum photonic circuits.


Supporting Information

Supplement 1

Xiao-Liu Chu*, Tommaso Pregnolato, Rüdiger Schott, Andreas D. Wieck, Arne Ludwig, Nir Rotenberg and Peter Lodahl

1. Distribution of QD alignment uncertainty

Using an improved mPL approach we were able to locate each QD relative to global alignment markers optically before nanofabrication and found an average positional uncertainty of 9.2 nm (see Figure S1). More details about the methodology can be found in ref. [29].

![Figure S1. The absolute position of each QD relative to alignment markers using the mPL approach to image the emitters and markers. The final uncertainty varies, but is on average 9.2 nm. Taken with permission from ref. [4].](image-url)
2. Voltage-wavelength photoluminescence spectrum

Our InAs QDs are self-assembled on a GaAs surface. A p-i-n diode is formed by doping the surrounding GaAs membrane and thus allowing us to control the charge environment and tune the QDs with an external voltage source. A standard voltage-wavelength photoluminescence spectrum is shown in Figure S2, where we use a laser at 795 nm to optically excite the QD.

![Voltage-wavelength photoluminescence spectrum](image)

Figure S2. Voltage-wavelength photoluminescence spectrum of a single QD optically excited with a laser (Tsunami) at 795 nm.

3. Calculating the rate equations

\[
\begin{align*}
|X\rangle_b & \quad \gamma_{bd} \quad |X\rangle_d \\
\gamma_{br} & \quad \gamma_{db} \quad \gamma_{b,nr} \quad \gamma_{d,nr} \\
|g\rangle &
\end{align*}
\]
Figure S3. A diagram of the energy levels for a neutral exciton. Both bright |$X_b$⟩ and dark |$X_d$⟩ states have radiative and non-radiative transitions.

The rate equations for a three level system:

$$
\dot{\rho}_1 = \gamma_{b,r} \rho_2 + \gamma_{b,nr} \rho_2 + \gamma_{d,nr} \rho_3
$$

$$
\dot{\rho}_2 = -\gamma_{b,r} \rho_2 - \gamma_{b,nr} \rho_2 - \gamma_{bd} \rho_2 + \gamma_{db} \rho_3
$$

$$
\dot{\rho}_3 = \gamma_{bd} \rho_2 - \gamma_{db} \rho_3 - \gamma_{d,nr} \rho_3.
$$

(S1)

We use the following boundary conditions: $\rho_2(0) = 0.5$; $\rho_1(0) = 0$; $\rho_3(0) = 0.5$.

We also make the following assumptions:

1. At the temperatures of this experiment ($T = 10$ K), and using the formula $\gamma_{bd} = e^{\delta_{bd}/k_B T} \gamma_{db}$, where $\delta_{bd}$ is the energy splitting between the bright and dark state (hundreds $\mu$eV ) [1], we find that the spin-flip rates are the same, i.e. $\gamma_{bd} = \gamma_{db}$.

2. Experiments have shown that the non-radiative decay rates from the dark and bright states are the same [2, 3]: $\gamma_{b,nr} = \gamma_{d,nr}$

Solutions to the rate equations are therefore:

$$
\rho_2 = A_f e^{-\gamma_f t} + A_s e^{-\gamma_s t},
$$

(S2)

where

$$
\gamma_f = \frac{\gamma_{b,r}}{2} + \gamma_{b,nr} + \gamma_{db} + \sqrt{\gamma_{db}^2 + \gamma_{b,r}^2 / 4}
$$

$$
\gamma_s = \frac{\gamma_{b,r}}{2} + \gamma_{b,nr} + \gamma_{db} - \sqrt{\gamma_{db}^2 + \gamma_{b,r}^2 / 4}
$$

$$
A_f = \frac{1}{4} \left( 1 + \frac{\gamma_{b,r} - 2 \gamma_{db}}{\sqrt{\gamma_{b,r}^2 + 4 \gamma_{db}^2}} \right)
$$
\[ A_2 = \frac{1}{4} \left(1 - \frac{\gamma_{b,r} - 2\gamma_{db}}{\sqrt{\gamma_{b,r}^2 + 4\gamma_{db}^2}}\right). \]  

(S3)

For small spin-flip processes \( \gamma_{b,r} \gg \gamma_{db} \), as has been reported in previous work [2], the above expression simplifies to the following:

\[ \gamma_f \rightarrow \gamma_{b,r} + \gamma_{b,nr} \]

\[ \gamma_s \rightarrow \gamma_{b,nr} \]

\[ A_f \rightarrow \frac{1}{2} \]

\[ A_z \rightarrow 0 \]  

(S4)

In bulk, the two dipole contributions cannot easily be separated and hence a single exponential is observed. In the waveguide, we expect the lifetime measurements to show a bi-exponential decay. Instead a significant non-radiative contribution to the decay that also dominates the weakly coupled dipole result in well-modelled single-exponential decays.

We used Equation S2 and measured the instrument response function (IRF) in order to fit our lifetime measurements. An example of a lifetime measurement and fit can be seen in Figure S4.
2. Calculating the normalized decay rate

We measured the decay rate $\Gamma_{wg}$ for all of the QDs in the PhCWs, and group them by their position in the unit cell. The details of the lifetime fitting procedure can be found the section above. Each data point is normalised to the $n_g$ experienced by that QD, and the position dependent decay rates is presented in Figure 6 in the paper and Figure S3. Each individual measurement is represented by a red circle, while the mean and standard deviations are given by the black circles and error bars. The calculated normalized decay rate (at $n_g = 20$) is also shown for comparison, for both transverse ($y$-oriented) and longitudinal ($x$-oriented) dipoles, using red and blue shaded regions that reflect both the $2\sigma$ and $1\sigma$ positional uncertainty of the QDs. By comparing our measurements to the calculations at positions containing many QDs, e.g. at $x = 0.5a$; $y = 0a$ near the mode maximum, we conclude that the large observed spread...
in decay rates arises due to the presence of non-radiative recombination. This is because in the ideal case with unity QE, shown as the shaded region in Figure 6 in the paper, our calculated normalized decay rate spread of X and Y for QDs within 1σ and 2σ of this position is smaller than the measurement. If we instead proceed to increase the non-radiative decay rate, and hence lower the QE of the emitter, we find that the calculated normalized decay rates decrease, as is presented in Figure S3. This can be understood as the Purcell factor acts on the radiative rate γr only. A larger non-radiative contribution to the bulk decay rate therefore results in less Purcell enhancement of the decay rate and in doing so one can recover the large spread as observed experimentally.

Figure S5. The measured normalized decay rate Γwg/(n_gΓ_B) of QDs located at different positions within a PhCW unit cell. The QDs are located at (a) y = 0 and shifted along x or (b) x = 0.5a and shifted along y. Individual decay rates for each QD can be seen as red circles and the black squares are the mean values. The calculated branching ratio between the two dipoles, at each nominal position for n_g = 20, is also given (black curve, right axis).
shaded regions correspond to the calculated normalized decay rates of the x-oriented and y-oriented dipoles, and reflect the 2σ positional uncertainty of our protocol for different QE (=0.1, 0.2, 0.3, ..., 1).

References


