

# Spontaneous Emission Enhancement of CdSe Quantum Dots Embedded in a Two-dimensional Photonic Crystal L3 Nanocavity

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**Abstract**—Two-dimensional photonic crystal nanocavities were designed to tailor cavity quantum electrodynamics. Enhancing the spontaneous emission of low-quality factor nanocavity with embedded CdSe quantum dots (QDs) emitters is the aim of this study. Low concentration layer of CdSe QDs was sandwiched between two layers of  $\text{Si}_2\text{N}_3$  membrane using plasma-enhanced chemical vapor deposition. The modification rate in spontaneous emission of L3 nanocavity up to 2.3-fold has been observed at 629.5 nm in compare to bare cavities. High field confinement in the sub-wavelength regime became an interest field for quantum electrodynamics applications and good platform to study light matter interactions.

**Index Terms**—Quantum dots; L3 cavity; Photonic crystal; Purcell factor.

## 1. INTRODUCTION

The ability to confine light into small volumes is important for enhancing the light and matter interaction in the emerging field of nanophotonics. Optical confinement in the sub-wavelength regime is of high interest for quantum electrodynamics applications (Chang, et al., 2006; Gong and Vučković, 2007). They are bounding the electromagnetic excitations between semiconductors and a dielectric, leading to a strong interaction between quantum emitters and photon fields (Chang, et al., 2006). This interaction can further be enhanced through an appropriate feedback mechanism that provided by nanocavity structures. The enhancement of cavity spontaneous emission (Purcell effect) is essential for a variety of applications, such as, single-photon sources (Wie, 2019), integrated quantum optics (Mahmoodian, et al., 2016; Daveau, et al., 2017), nanoscale lasers (Oulton, et al., 2009), active meta-materials (Schulz, et al., 2016), biotechnological devices for enhanced fluorescence intensity (Lakowicz, et

al., 2003), and ultra-fast modulated LEDs (Shambat, et al., 2011). Nanolaser performances are also highly affected by light emitters that introduced within the resonator. The gain properties of semiconductor quantum dots (QDs) have a lot of advantageous due to their temperature stability, small line width enhancement factor, and the ability to suppress the non-radiative recombination (Arakawa, et al., 1982). In addition, QDs are suitable for studying cavity QED, because of their atom like quantum emission (Yoshie, et al., 2004; Reithmaier, et al., 2004; Arakawa, et al., 2012). Recently, a study has been carried on field improvement at communication wavelength using InGaAs QDs embedded in a GaAs nanowire having groove like cavity, which manifested high enhancement by 617-fold (Wie, 2019). Recently, some studies were carried out on field improvement at communication wavelength, for example, InGaAs QDs was embedded in a GaAs nanowire having groove like cavity, which manifested high enhancement by 617-fold (Wie, 2019); while using Ge/Si QDs within Si two-dimensional photonic crystal (2D PC) field enhanced by about 25 times at  $\lambda = 1.2 \mu\text{m}$  and by 34 times  $\lambda = 1.6 \mu\text{m}$  (Yakimov, 2021).

In this study, field enhancement toward visible wavelength using CdSe QDs embedded in  $\text{Si}_2\text{N}_3$  2D PC was proposed. A remarkable field enhancement was observed using even low-quality factor L3 nanocavity. Such result can be promising to study light matter interaction at visible wavelength.

## II. MATERIALS AND METHODS

In this work, the realization and characterization of a hybrid photonic crystal (PC) cavity that combines the benefits of both QDs and photonic elements has been presented. Here, an 8 nm of CdSe QDs (provided by Sigma-Aldrich) is used as an active emitter layer.

### A. 2D PC Fabrication

First, a suspended layer of  $\text{Si}_2\text{N}_3$  with a thickness of 100 nm was coated with a thin layer of 8 nm of CdSe QDs which diluted to a concentration of 0.01 mg/ml in toluene solution. Second, another layer with 100 nm of  $\text{Si}_2\text{N}_3$  is deposited by plasma-enhanced chemical vapor deposition. The whole



structure thickness becomes 200 nm with a sandwiched layer of CdSe QDs. A 200 nm polymethyl-methacrylate (PMMA) electron beam resist layer was spin coated on the  $\text{Si}_2\text{N}_3$  surface then backed on a hot plate for 1 min at  $100^\circ\text{C}$  to harden it and remove any residual solution. Subsequently, a 30 kV electron beam lithography was used to define the required PC pattern in PMMA. Then, exposed PC pattern in resist by electron beam is removed by methyl isobutyl ketone: isopropyl alcohol 1:3 developer. Finally, the PC pattern is transferred from PMMA into the  $\text{Si}_2\text{N}_3$  layer using reactive ion etching to obtain air holes through the  $\text{Si}_2\text{N}_3$  layer. The used recipe of the RIE dry etching device was based on a mixture gas of  $\text{CHF}_3$  and  $\text{O}_2$ , with flow rates 30 and 5 sccm, respectively.

### B. Photolumens (PL) Measurements

The fabricated PC is optically pumped using a CW diode pumped solid state laser at 532 nm. The beam focused through a  $100\times$  long working distance objective lens (6 mm) having a 0.77 numerical aperture (NA) to a  $2.5\ \mu\text{m}$  diameter spot on the cavity surface. The laser spot was controlling using a three-dimensional piezo motor stage (Piezosystem Jena Tritor 100, Micro XYZ Positioner stage NV 40/3s controller). The emission spectra were collected using the same objective lens and analyzed by nitrogen cooled CCD detector. Long-pass filter with a cutoff wavelength at 532 nm was used to block the excitation light from reaching the detector.

## III. RESULTS AND DISCUSSION

The QDs solution diluted to a concentration of 0.01 mg/ml in toluene and spun into the structures at 1.5 krpm (to reduce unwanted aggregation) on 100 nm  $\text{Si}_2\text{N}_3$  layer, resulting up to  $\sim 10^2$  emitters dots per  $2\ \mu\text{m}^2$ . The coating was not uniform due to the small chip size of the membrane ( $0.25\ \text{mm}^2$ ) as it is shown in Fig. 1.

PL has been taken for embedded layer of QDs as shown in Fig. 2. The emission spectrum shows a peak position at 640 nm with full width at half maximum (FWHM) 50 nm. The wide width of emission spectrum line is due to the effect of the dots aggregation which leads to red shift, besides the influence  $\text{Si}_2\text{N}_3$  layer PL emission.

The fabricated PC dimension is  $10\ \mu\text{m} \times 10\ \mu\text{m}$  with three missed holes in the center act as nanocavity. Such structure is known as PC with L3 cavity. The produced PC L3 cavity has a lattice constant  $a = 260\ \text{nm}$  (distance between two adjacent holes), hole radius  $r = 0.288a$ , and total cavity length  $0.78\ \mu\text{m}$  approximately, as shown in Fig. 3.

In general, PC as an optical system can enhance or suppress the rates of spontaneous emission for any quantum emitters. The coupling between cavity modes and the QDs as two-level system is a type of energy exchanging between them. In such system (QDs – nanocavity), photons are supplied with a certain probability that depends on their environment and density of states. The coupling between quantum emitters and cavity mode leads to a remarkable enhancement of the spontaneous emission of the output modes which is known as Purcell effect (Purcell, 1946) and the Purcell factor is totally

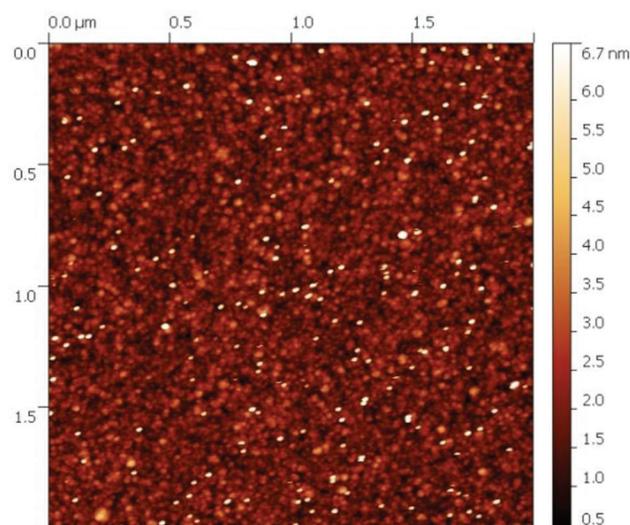


Fig. 1. Atomic force microscopy image of thin layer of 0.01 mg/mL of CdSe quantum dots coated on a thin layer of  $\text{Si}_2\text{N}_3$  membrane. Acceptable distribution and low aggregation are observed.

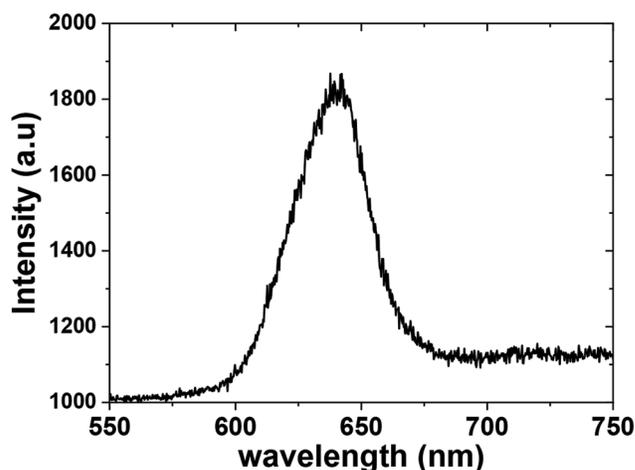


Fig. 2. Photo lumens emission of 0.01 mg/mL of CdSe quantum dots sandwiched within 200 nm layer of  $\text{Si}_2\text{N}_3$  layer.

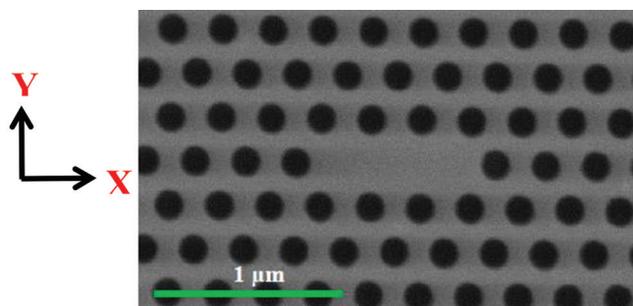


Fig. 3. Scanning electron microscope image of two-dimensional photonic crystal L3 cavity printed on  $\text{Si}_2\text{N}_3$  membrane.

depend on Q/V ratio (Ryu and Notomi, 2003):

$$F = \frac{3}{4\pi^2} \frac{Q}{V_{mode}} \left( \frac{\lambda_0}{n} \right)^3 \quad (1)$$

Where:  $Q$  quality factor,  $V_{mode}$  mode volume, and  $n$  is medium refractive index.

The enhancement of spontaneous emission rate  $\gamma$  is strongly ruled by the emitter alignment with the dipole moment  $\vec{\mu}_i$  at a point on the cavity, which can be derived from Fermi's Golden rule (Boroditsky, et al., 1999; Englund, et al., 2005).

$$\gamma = \frac{\gamma_{cav}}{\gamma_{free}} = F \times \left( \frac{\overline{E \cdot \vec{\mu}}}{|\vec{E}|_{max} |\vec{\mu}|} \right)^2 \times \frac{\Delta\lambda_{cav}^2}{4(\lambda - \lambda_{cav})^2 + \Delta\lambda_{cav}^2} \quad (2)$$

To measure the enhancement of the confined field through Purcell factor inside nanocavity before and after introducing QDs emitters, number of parameters should be estimated, such as quality factor  $Q$  and mode volume  $V_{mode}$ .

Physically, the effective cavity mode volume is known as a spatial distribution of electromagnetic energy (Balanis, 2013).

$$V_{mode} = \frac{\int \epsilon E^2 dV}{\max(\epsilon E^2)} \quad (3)$$

In purely dielectric cavities, the ratio of the volume of the active region ( $V_a$ ) to mode volume ( $V_{mode}$ ) is known as confinement factor (Chang and Chuang, 2009):

$$\tilde{A} = \frac{V_a}{V_{mode}} \quad (4)$$

Therefore, to obtain high confined cavities, the mode volume should be smaller than the active region volume, that is,  $\Gamma > 1$ . In general, the 3D nanocavity effective mode volume calculation needs to use numerical methods due to the geometric and material complexity of the cavities. Experimentally, the modes are considered completely confined to the active region volume;  $V_{mode}$  approaches the geometrical active medium volume  $V_a$  and Equation 4 becomes  $\Gamma \approx 1$  (Li, 2018).

In this experiment, cavities without QDs show no output mode enhancement (Fig. 4). However, PC L3 designed cavity supports one mode on X-axis direction (long cavity axis),  $\lambda = 625$  nm FWHM  $\Delta\lambda = 2.08$  nm (Fig. 5 red curve). Mode quality factor  $Q$  inside nanocavity which known as the ratio of stored energy to dissipated one can be calculated by  $Q = \lambda/\Delta\lambda$ . Hence, mode cavity shows low-quality factor  $Q=303$  (Fig. 5 red curve) whereas the field enhancement calculated from Purcell factor from Eq. 1 is  $F_{bare}=30.29$  using the mode volume normalized to the mode wavelength

$$V_{mode} = 0.76 \left( \frac{\lambda}{n} \right)^3$$

Introducing a thin layer of CdSe QDs which sandwiched in the PC membrane, two modes were observed  $\lambda_1 = 624.5$  nm and  $\lambda_2 = 629.5$  nm with both  $\Delta\lambda \approx 0.9$  nm as shown in Fig. 5 (black curve). Consequently, both modes' Q-factor increased significantly to  $Q_1 = 690$  and  $Q_2 = 699$ , respectively. Using the same mode volume normalized to the mode wavelength in the Purcell factor formula for the highest  $Q_2$ , factor mode

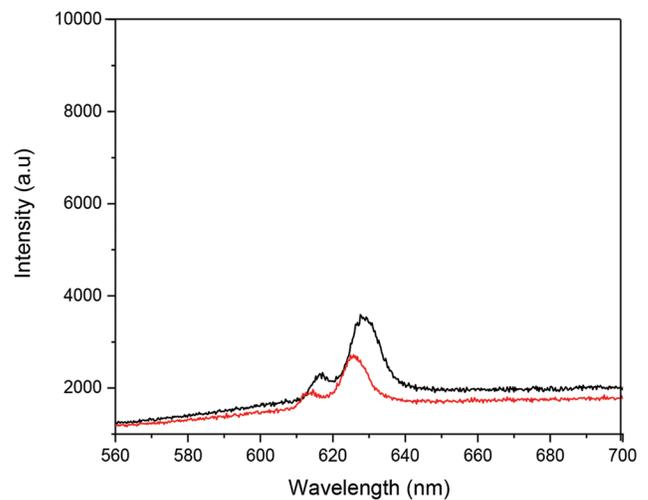


Fig. 4. Photo lumens spectrum for L3 cavity modes before introducing CdSe quantum dots (QDs) (red curve) and after introducing a thin layer of CdSe QDs (black curve).

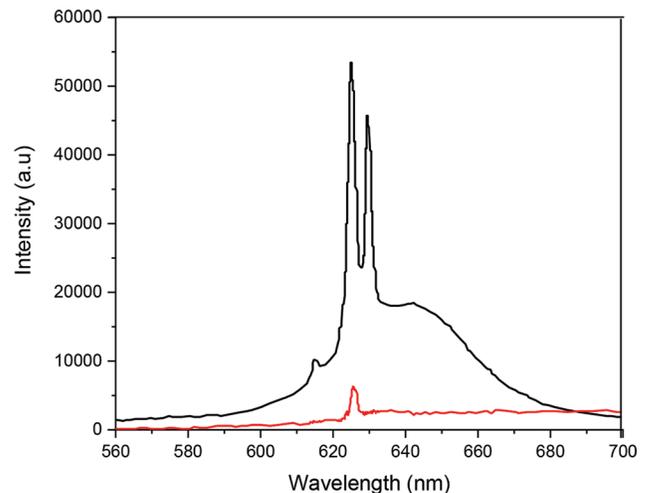


Fig. 5. Photo lumens (PL) spectrum for L3 cavity modes before introducing CdSe quantum dots (QDs) (red curve) and after introducing a thin layer of CdSe QDs (black curve). Here, QDs emission modes are highly aligned with cavity modes shows remarkable enhancement of PL.

is  $F_{QD} = 71.29$  so that the cavity field shows enhancement by 2.36-fold ( $F = F_{QD}/F_{bare}$ ).

In addition, Eq. 2 indicates that the location of quantum emitters inside nanocavities plays an essential role to either enhance or suppress the output emission. Thus, the PC cavity modes-QD exciton system on resonance should be located at the maxima electric cavity field (Vuckovic and Yamamoto, 2003). Enhancements in both  $\lambda_1$  and  $\lambda_2$  modes (Fig. 5) can be referred to the increase of coupling parameter to the decay rates ratio through strong coupling system (Vuckovic and Yamamoto, 2003; Yoshie, et al., 2001).

On the other hand, cavities that contain slightly misaligned QDs position show PL enhancement but low quality factor mode  $Q=233$  as illustrated in Fig. 6 (red curve before and black curve after introducing QDs layer).

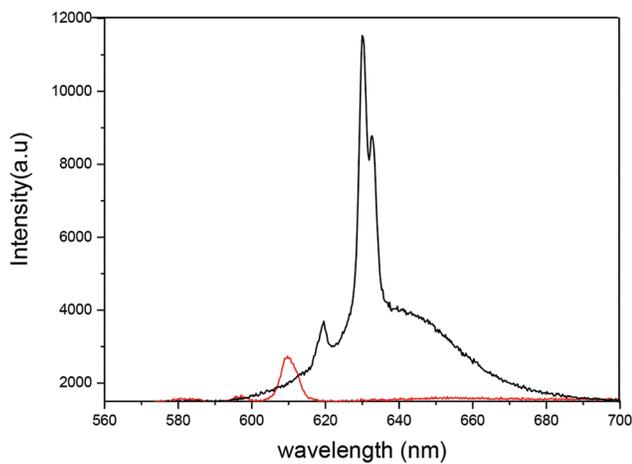


Fig. 6. Photo lumens (PL) spectrum for L3 cavity modes before introducing CdSe quantum dots (QDs) (red curve) and after introducing a thin layer of CdSe QDs (black curve). Here, QDs emission modes are slightly misaligned with cavity modes. PL emission shows enhancement but low Q factor 233.

## VI. SUMMARY

PC cavity-QD emitters system is a good platform to study light matter interaction. Mode field enhancement was observed in the order of 2.3-fold at 629.5 nm which occurs when the QD and the cavity are in resonant and the QD is spatially aligned to the cavity mode. The advantages of the used technique are the low cost and limitation of using chemicals, however, it is difficult to locate QDs at a desirable position due to random distribution of the QDs during the coating process. Using semiconductor, QDs as a gain material can improve the effective light matter interactions through nanolasers performance.

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