Strong Purcell enhancement of emission from close-packed colloidal quantum-dots in a photonic-lattice cavity

T-S. Luk, S. Xiong, B. G. Farfan, W. W. Chow, I. El-Kady, X. Miao, P. J. Resnick, M. F. Su, G. Subramania, M. R. Taha, C. J. Brinker
Sandia National Laboratories, Albuquerque, New Mexico 87185
tsluk@sandia.gov
505-844-8931

Abstract

High Purcell spontaneous emission enhancement factor of 116 is achieved by integrating a self-assembled, close packed monolayer of colloidal PbS quantum dots with a L3-type silicon photonic crystal cavity.

High Q optical microcavities can be used to control the radiative process of dipole emitters, such as quantum dots, through photonic density of states control. By enhancing the spontaneous emission, heat dissipating non-radiative relaxations can be suppressed therefore resulting in greater light emitting efficiency and providing a possibility of extracting multi-exciton energy by the emission process. In order to study the radiative control of quantum dots, it is important to be able to incorporate these emitters without perturbing the optical properties of the cavity. Serious challenges remain in integration of the photonic-crystal cavity structure and quantum dots without significant Q factor degradation. Standard spin-coating techniques fail to retain the inherent Q factor and achieve high quantum-dot concentration, consistent film thickness and controlled uniform particle separation from the surface.

This paper reports progress in meeting some of these challenges. We achieved an enhanced emission factor of 116 from close-packed PbS quantum-dot/polymer monolayers transferred to a L3 Si photonic-crystal cavity with Q factor around 2860. This is a significant improvement over previously reported Purcell factor[1] of 10 involving colloidal quantum dots[2]. The key factor enabling this result is the ability to deposit a 20nm thick monolayer of close-packed quantum dots onto a photonic-crystal cavity to provide direct contact with the cavity. The quantum-dot density of 10^4 μm^{-2}, which is over two orders of magnitude higher than typical with Stranski-Krastanow growth, directly contributes to high emission intensity and mitigates the difficulty of placing dots selectively at the anti-nodes of the optical modes. An optimal mode confinement factor is guaranteed by the uniformity (on the scale of an optical wavelength) of dot density throughout the cavity volume.

The experiments were performed with L3 (a row of three missing air holes) photonic-crystal

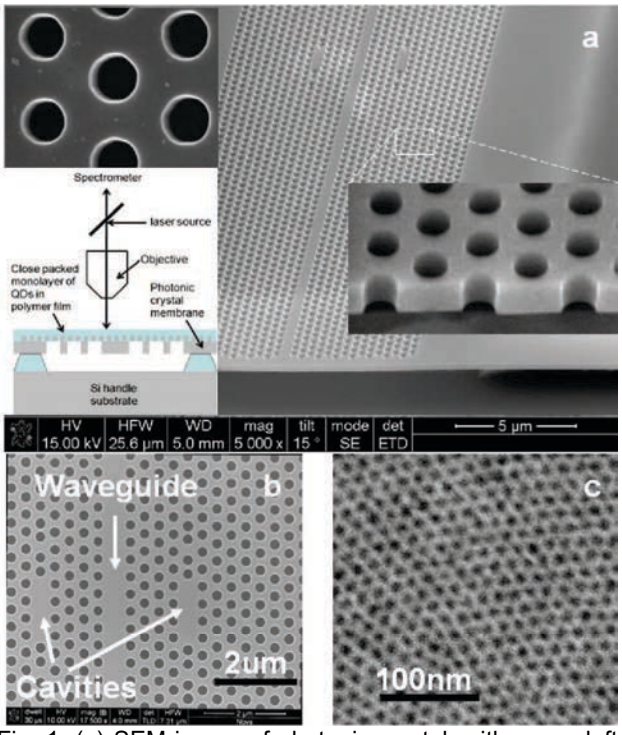

Fig. 1. (a) SEM image of photonic crystal, with upper left inset providing a detailed view of the holes (240nm diameter) and lower right inset showing the hole edges and sidewalls. The lower left inset provides a schematic diagram of the photoluminescence measurement geometry. (b) Top view of two identical L3-type photonic crystal cavities. (c) TEM image of a free-standing PbS quantum dot/P3HT polymer monolayer to be transferred to the photonic crystal surface.

cavities in a triangular lattice shown in Fig. 1, with lattice constant in the Γ-K direction of a = 415nm. In this design (referred to as the L3P cavity), the positions of the three holes at both ends of the cavity are shifted by 0.18a, 0.025a and 0.18a, as in the design by Akahane et. al.[3]. The air-hole radii are 0.29a and the silicon slab thickness is 0.63a. For convenience, the direction of the waveguide (a missing row of air holes) also serves as a reference frame for describing polarization direction. For the experiments, 2-D photonic crystals were fabricated from 150mm silicon-on-insulator (SOI) substrate (Fig 1a and b). Each device has a 250nm thick patterned layer and a 3μm thick buried-oxide layer (BOx). As recently reported[4], we create a close-packed PbS quantum-dot monolayer by evaporation-induced nanoparticles/polymer self-assembly at a fluid interface, followed by monolayer transfer.

The solid curve in Fig. 2 shows the measured PL spectrum from photo-excited quantum dots on the L3P cavity. A schematic of the micro-photoluminescence (PL) apparatus together with the excitation and detection geometries are shown in Fig. 1a inset. The excitation source is a cw 830nm diode laser delivered by a single-mode 5.6μm core fiber collimated to match the entrance pupil of a 0.65NA Mitutoyo 50X NIR HR objective.

In the experiment, the cavity resonances are determined by locating the enhanced emission frequencies using a micro-PL apparatus. This radiation is from the leakage of the trapped radiation in the cavity. An expanded view of the emission lineshape centered at 1574nm is depicted in the inset. The narrowest linewidth of 0.55nm implies a loaded-cavity Q factor of 2860, which is the highest of all our structures. The enhanced emission abve the background occurs only when the excitation region is in the cavity and the emission is polarized in accordance to the cavity mode. This background is from photo-excited quantum dots outside of the cavity and scattering from cavity imperfections.

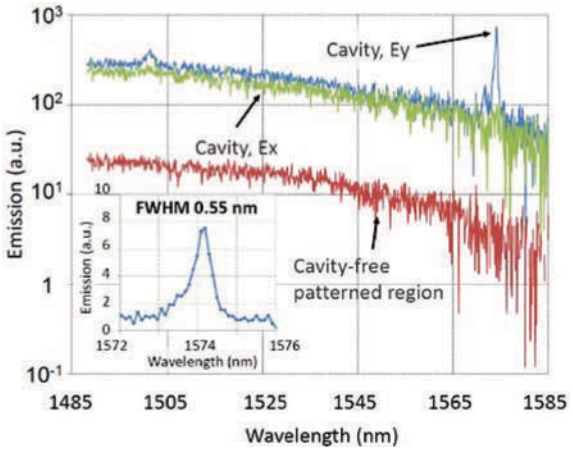

Fig.2. PL from photo-excited PbS quantum dots. The emission spectra from the L3P cavity are for polarizations perpendicular (blue trace) and parallel (green trace) to the waveguide. The spectrum for emission from the cavity-free patterned region (red trace) is also perpendicularly polarized and normalized to the cavity area. The inset shows an expanded plot of the cavity resonance.

In conclusion, we demonstrate significant spontaneous emission enhancement, achieving a maximum Purcell factor of 116, which is the highest yet reported for a colloidally-derived quantum-dot device. Our experiments are based on the integration of a self-assembled, close packed monolayer of PbS quantum dots with a L3-type silicon photonic crystal cavity. The key factor enabling this performance is our ability to transfer a macroscopically uniform, close-packed, 20-nm thick QD monolayer to the photonic crystal. This fabrication approach maintains direct QD contact with the cavity, achieves the highest possible QD density, and guarantees an optimal emitter-to-optical-mode confinement factor. Our results emphasize that combining top-down lithography and bottom-up self-assembly is a viable approach for advancing the nanophotonics field.

References

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