Strongly inhibited spontaneous emission of near-IR PbS quantum dots in 3D silicon photonic band gap crystals

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Introduction

Central in quantum optics and cavity quantum electrodynamics (cQED) ⇒ Control properties of matter via the properties of light!

- Famous: control radiative rate of elementary quantum emitters, like dye molecules, ions, or quantum dots (QDs) [1]
- Crucial for applications: miniature lasers & light-emitting diodes [2], photocatalysis & photochemistry [3,4], and sensing [5,6]

Challenge: study emitters placed at well-defined positions in 3D photonic band gap crystals

Vacuum Fluctuations & LDOS

Optical emitters typically in quantum regime ⇒ Emission induced by both radiative reaction (“antenna effect”) & by vacuum fluctuations [7]

- Vacuum fluctuations: fluctuations of quantized EM-fields (red wavelets), even in free space
- Combined effects captured by local density of radiative states (LDOS) [8]
- LDOS controlled by surrounding emitter by suitably tailored dielectric environment
- Most radical control: 3D photonic band gap: emission strongly inhibited

From Band Gap to Theory Inhibition in Experiments

(a) Reduced density of states (DOS, a lattice parameter, c = speed of light) for a 3D inverse woodpile photonic crystal from silicon, calculated for pores with radii R/a = 0.252 (red circles). Effective medium (blue); free space (green curve).
(b) Relative DOS equal to ratio of crystal and effective medium DOS (red circles). 

Main result: measured intensity of QDs in the band gap crystal (black squares) normalized to similar QDs on a Si surface ⇒ Broad & deep gap apparent!

Our Photonic Crystals

(a) Scanning electron micrograph (SEM) of a 3D inverse woodpile photonic crystal (pore radii R=160 nm) made by us at MESA+
(b) Schematic of the unit cell, zooming in one a pore to show how we position QDs by targeted polymer surface-chemistry: ATRP initiator layer (orange), polymer chains forming brushes (green), covalently attached PbS quantum dots (black) on silicon (blue). The positioning was verified by X-ray fluo tomography [9].

Strongly Inhibited Emission

Time-correlated single photon counting of PbS QDs on flat Si (black squares), in crystal S4 (blue triangles), crystal S12 (red circles), in suspension (green hexes) near the PBG crystal ⇒ Emission time inhibited ⇒ Calculated relative DOS for relevant pore radii (connected black symbols) ⇒ Excellent reproduction between different crystals with same pore radii. Systematic shift of the band gap with varying crystal dimensions

References