Significant Enhancement of Colloidal Quantum Dots Fluorescence by 3D Disk-dot Coupled Nanoantenna Array

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Colloidal quantum dots (QDs) have many unique and attractive optical properties[1], and promise various optical applications. The QDs fluorescence can be enhanced using metallic nanostructures (plasmons). However, to date, such enhancement is only several folds. Here we experimentally demonstrate a significant QD fluorescence enhancement on a new plasmonic structure named "disk-coupled dot-on-pillar antenna array" (D2PA). The achieved enhancement factor is 255 fold (25,500 %), exceeding the previously best over 1 order of magnitude[2]. Furthermore, the enhancement is extremely uniform (~10% variation) over a large sample area, and the study of single dot at hot spot shows 10^4 enhancement indicating potential further improvement in average fluorescence.

D2PA has an Au nanodisk on top of each of periodic pillars, an Au backplane on the pillar foot, dense nanodots on each pillar sidewall, and nanogaps between the Au components[3] (Fig. 1). The Au disk and the backplane forms a vertical nanocavity which, together with the nanodots and nanogaps, can effectively collect and enhance a pumping light in its near-field zone, and efficiently couple the fluorescence emission back into the far-field, therefore enhancing the fluorescence significantly.

The fabrication process of D2PA (Fig. 2), using nanoimprint, self-alignment, and self-assembly, has: (1) SiO_2 was grown on a Si substrate; (2) a 2D Cr disk array was then patterned by nanoimprint lithography (NIL), Cr evaporation and lift-off; (3) with the Cr mask, SiO_2 nanopillars were formed by reactive ionic etching (RIE); and (4) the Au nanodisks, nanodots and backplane were formed by one step of Au evaporation (self-align and self-assebmly). The 200 nm array pitch allows a density of nanocavity as high as 2.5×10^9 cm^{-2} (Fig. 3), which enables an extraordinarily large average electric field enhancement. After fabrication, lead sulfide (PbS) QDs in PMMA (5%) was spin-coated on the D2PA.

Significant fluorescence enhancement was observed from QDs on D2PA substrate (Fig.4a). The average enhancement is 255 fold on D2PA at the peak wavelength, compared to a non-plasmonic silicon substrate covered by identical QD/PMMA layer. For a single QD at a hot spot, we observed 10^4 fluorescence enhancement on D2PA, suggesting a giant enhancement potential. The large fluorescence enhancement factor is independent with the QD concentration. Moreover, we found the D2PA enhancement factor is nearly constant over a broad range of QD concentrations, with a small variation (Fig. 4b).

In summary, we demonstrated a 255-fold fluorescence enhancement from QDs using a novel plasmonic nanocavity substrate, D2PA. Such enhancement is uniform over a large sample area and over a wide QD concentration range. This study opens the door towards a wider range of applications, such as quantum information, biosensor and quantum dot laser.

[1] J. M. Caruge, etc. *Nature Photonics* **2**, 247-250 (2008).
[2] Tanaka, K., etc., Phys. Rev. Lett. **105**, 1-4 (2010).
[3] W. Li, etc., *Optics express* **19**, 3925-3936 (2011).

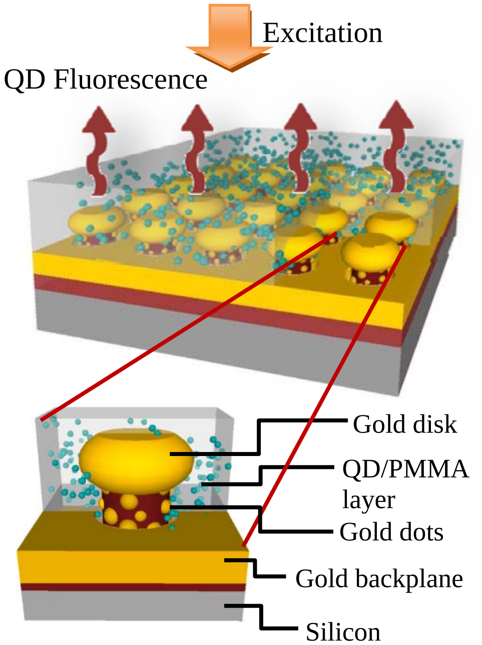

Fig. 1. Schematic of disk-coupled dots-on-pillar antenna array (D2PA). QD/PMMA layer are spinned-coated on the top to fill the small gaps.

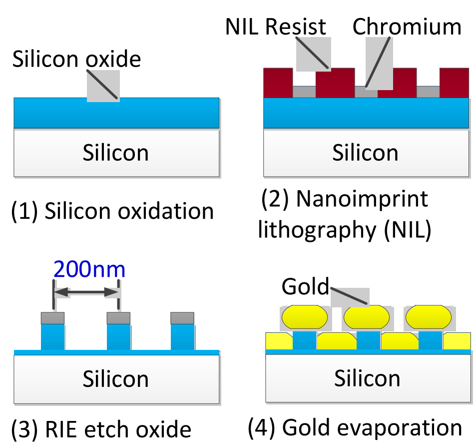

Fig. 2. D2PA fabrication steps: (1) growth of oxide layer on silicon surface; (2) nanoarray patterning with 200 nm pitch using nanoimprint lithography (NIL); (3) etching down oxide with patterned chromium as mask; (4) gold deposition.

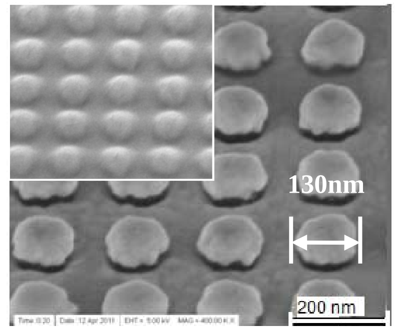

Fig. 3. Scanning electron micrographs of D2PA nanostructure. The "mushroom" shaped antenna features a 130nm disk on pillar top. Inset: D2PA covered by QD/PMMA layer.

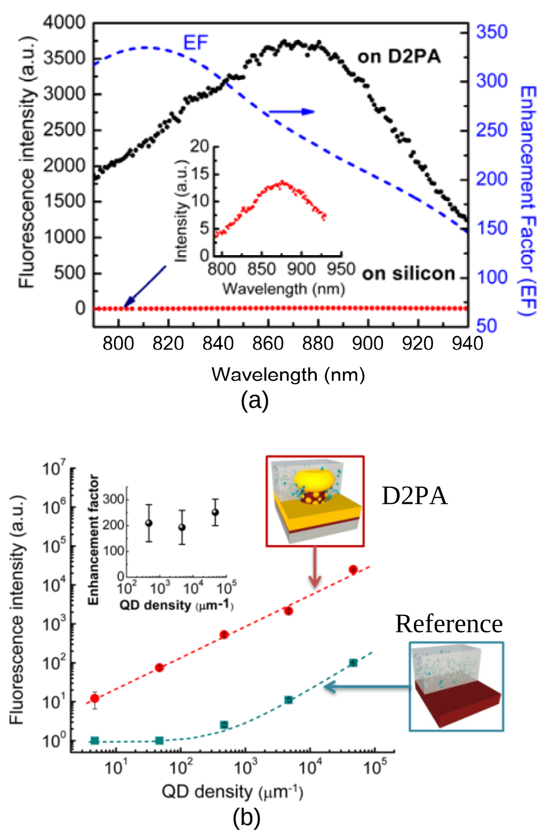

Fig. 4 (a) QD fluorescence on D2PA structure (black) and reference silicon substrate (red). Blue dashed line indicates enhancement factor spectrum. Inset: reference fluorescence in a smaller scale. (b) Fluorescence intensity as a function of QD concentration. Fluorescence signal shows near-linear relation versus QD density on D2PA (red), while for reference sample the threshold density appears (cyan). Inset: Fluorescence enhancement factors under different QD concentrations.