Shaping spontaneous emission from a single quantum dot into a narrow beam pattern

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Abstract: We experimentally observe directional spontaneous emission with angular half-width of ~10 degrees from a single CdSe/ZnS quantum dot positioned in a slit nanoaperture surrounded by periodic corrugations. ©2009 Optical Society of America
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Unlike dielectric or semiconductor microcavities, their metal counterparts can be reduced to volumes significantly smaller than λ^3, enabling very tight light confinement. Thus, metal nanocavities are promising potential candidates for obtaining enhanced nonlinear-optical effects, as well as for building single-photon sources and low-threshold lasers. The critical issue for such sub-wavelength structures, however, is the efficiency of coupling the light between them and a desired free-space mode without much loss due to scattering into other modes. One example of a sub-wavelength metal cavity is a narrow slit made in a metal film, which is equivalent to a rectangular box with metal walls. One dimension of such box can be made arbitrarily small [1]. It was demonstrated experimentally [2] that, by surrounding the slit with periodic corrugations, the emission pattern from it could be concentrated in a narrow range of angles despite the slit's sub-wavelength width. We have recently extended the first-principles approach [3,4] to show that more than 40% coupling to a macroscopic Gaussian mode can be achieved with losses into other modes less than 30% (and with the remaining 40% reflected back into the nanocavity) [5,6]. Another method of forcing the spontaneous emission into a particular mode is using the Purcell effect in a high-Q microcavity [7]; however, coupling from that mode to the desired free-space mode may still be rather inefficient, and only a fraction of the total number of emitted photons is captured. In this paper, we show the modification of the emission pattern from a single quantum dot by the corrugated metal nanostructure, which makes it easier to couple it to the desired external mode. For that purpose, we have not tried to maximize Q of the metal nanocavity formed by the slit, which could further lead to increase in the spontaneous emission rate.

Our beam shaping nanostructure is similar to that in [1], consisting of a 65-nm-wide slit surrounded by N=7 40-nm-wide grooves (depth h=110 nm) on each side, with period d=500 nm. The 20-μm-long pattern is made by focused ion beam milling a 300-nm-thick thermally evaporated silver film (Fig. 1a). We analyze the structure by a confocal fluorescence microscope in diascopic (transmission) illumination mode with collecting dry objective with NA=0.85. The collected light is processed by a modified version of spatially resolving spectrometer [8] placed in the Fourier plane of the objective. This allows us to directly measure angularly resolved emission spectra from the corrugated pattern, shown in Fig. 1c (corresponding theoretical predictions are shown in Fig. 1b).

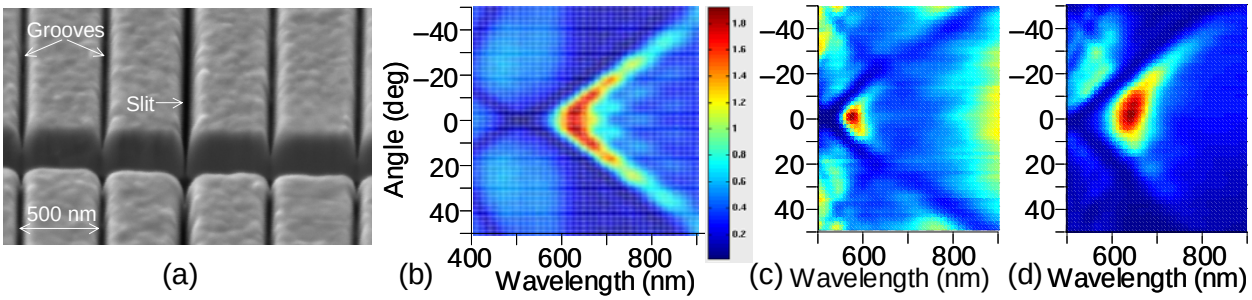

Fig. 1. (a) Cross-section of one of the samples showing slit nanoaperture surrounded by periodic corrugations. (b) Modeling predictions for angular emission pattern from the nanostructure with N=7 grooves on each side of the slit, h=110 nm depth of the grooves, and d=500 nm groove period. (c) Experimentally measured emission pattern (without quantum dots). (d) Experimentally measured emission pattern with several quantum dots positioned inside the slit.

Next, we position several colloidal CdSe/ZnS core-shell quantum dots with average emission peak near 630 nm into the central slit using a selective chemical attachment technique. The resulting pattern shown in Fig. 1d demonstrates single emission peak with full width of ~20° (quantum dots are excited by blue portion of illuminating white light).

In another sample, we place a single quantum dot into the slit. The corresponding emission pattern under 532-nm excitation is shown in the left insert of Fig. 2a. The emission spectrum and angular pattern are shown in Figs. 2a and 2b, respectively. We also compare these data with the spectra and angular patterns obtained from single

quantum dots on non-corrugated silver film and on a glass slide, also shown in Fig. 2. We can easily see a drastic reduction in the angular spread of the quantum dot's spontaneous emission in the presence of the corrugated nanostructure. The structure concentrates most of the emission into a single peak compatible with numerical apertures of common optical fibers, thereby promising a great improvement in the fluorescence collection efficiency for future quantum-optics applications.

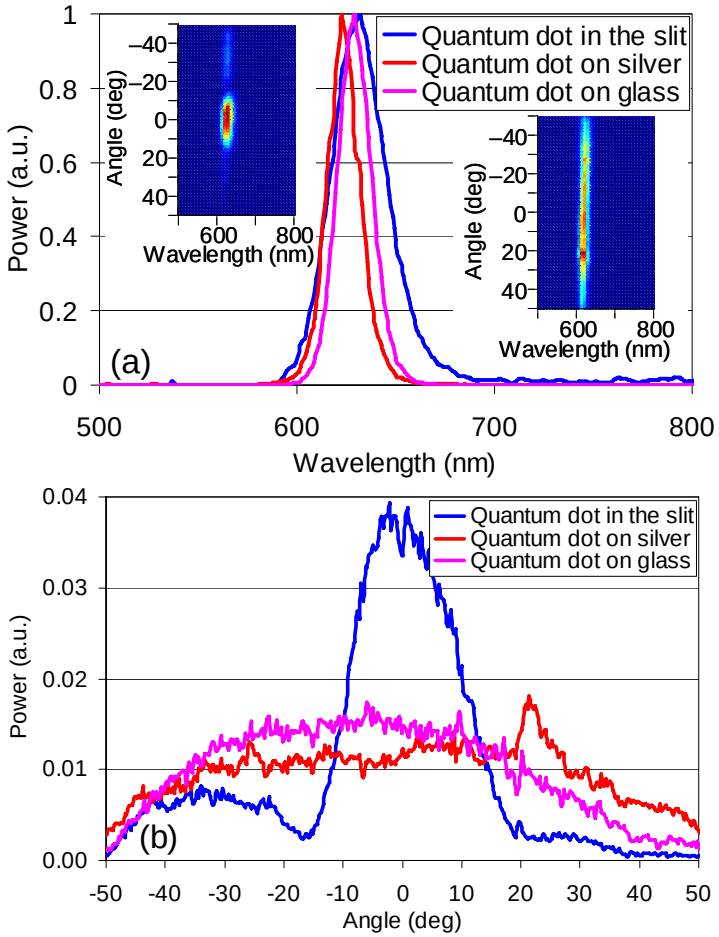

Fig. 2. (a) Angularly-integrated spectra of fluorescence from the CdSe/ZnS core-shell quantum dot positioned either inside the slit nanoaperture, or on non-corrugated silver film, or on glass. (b) Corresponding spectrally-integrated angular emission patterns, showing dramatic narrowing of the fluorescence pattern by the corrugated nanostructure. The left and right inserts on (a) show spectrally resolved emission patterns observed from the quantum dot positioned inside the slit and on non-corrugated silver film, respectively.

To summarize, we have demonstrated dramatic narrowing of the far-field fluorescence pattern from a quantum dot positioned inside a rectangular (slit) nanoaperture surrounded by periodic corrugations. Most (80%) of the emerging spontaneous emission is concentrated in a beam with angular half-width at half-maximum of ~10°, which indicates the potential for using this corrugated nanostructure for efficient coupling into and out of the slit nanocavity for enhancement of nonlinear- and quantum-optics effects.

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