

Article

Plasmonic Tilted Nanocavity Modulation of Quantum Dot Luminescence

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Abstract

Quantum dots combine advantages such as strong processability via solution methods, wide color gamut coverage, and precise emission color coordinates, making them highly promising for applications in optoelectronic devices. However, they face limitations such as insufficient fluorescence intensity and low far-field extraction efficiency. Plasmonic nanocavities based on metallic nanostructures offer an efficient platform for regulating light–matter interactions. In this study, we constructed a tilted plasmonic nanocavity structure composed of a silver nanocube, CdSe/CdS nanorods, and a single-crystal silver microplate. An Al₂O₃ isolation layer prepared via atomic layer deposition was used to control the nanocavity gap, precisely matching the plasmonic resonance mode with the 620 nm fluorescence emission of the quantum dots. This coupling system significantly enhances the radiative rate in the emission band and the electric field strength in the excitation band, achieving a 187-fold luminescence enhancement of the quantum dot. Additionally, leveraging the nano-antenna effect, the fluorescence exhibits upward directional emission. Experimental and simulation results confirm the high-efficiency enhancement and directional control of quantum dot fluorescence by the tilted nanocavity, providing new insights for the integrated application of quantum dots in displays, quantum communication, and other fields.

Keywords: quantum dots; nanocavity; surface plasmons; luminescence enhancement; nano-antenna effect

1. Introduction

Plasmonics based on metal nanostructures is an emerging field with significant theoretical research value and application potential, and it has rapidly developed in recent years [1–3]. Surface plasmons are collective coherent oscillations of free charges on metal surfaces, representing a type of material element excitation coupled with a light field and electrons [4,5]. Through the localized surface plasmon resonance of metal nanostructures, dipole, quadrupole, or multipolar electromagnetic modes can be generated on the surfaces of nanostructures, confining the light field to nano- and sub-nanoscale dimensions. This enables selective enhancement of the local electromagnetic field and effective regulation and enhancement of spectral properties such as scattering, absorption, and radiation of nearby luminescent centers, as well as the interactions between light and matter [6–8]. Plasmonic nanocavities based on this principle can confine the optical field at the nanoscale, drastically reducing the mode volume, thereby significantly enhancing the local density of



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states (LDOS) and producing a strong Purcell enhancement effect [9,10]. These features render plasmonic nanocavities highly promising for applications in surface-enhanced Raman scattering, quantum optics, photocatalysis, and sensing [11–15].

Quantum dots (QDs) are zero-dimensional nanostructures, with all three spatial dimensions confined by quantum confinement effects, typically measuring between 2 and 20 nm [16,17]. Under the excitation of an external optical or electric field, QDs can emit light at characteristic wavelengths [18]. Among them, colloidal semiconductor QDs exhibit a range of exceptional optical properties, showing immense application potential in the field of display technology [19–21]. Their emission spans the entire visible spectrum, making it directly perceivable by the human eye; the emission color is precisely tunable according to their size, achieving a wide spectrum of adjustable colors; at the same time, they possess narrow emission spectra, high color purity, and strong luminescence [22]. Due to the combined effects of quantum size and quantum confinement, the emission color of a QD changes predictably with its particle size. This size-dependent precise color tunability is one of the key reasons QDs are considered core candidate materials for display devices. Additionally, QDs offer significant advantages such as high processability via solution-based methods, excellent film-forming uniformity, broad color gamut coverage, high electro-optical conversion efficiency, precise emission color coordinates, and good compatibility with flexible substrates. These characteristics have made them a research focus in the field of optoelectronic materials [23–25].

For QD-based display devices, the regulation of optical performance is a core aspect of design, primarily encompassing key parameters such as luminous intensity, emission wavelength, and color purity [26]. Currently, the approaches to achieving QD optical performance regulation are mainly divided into two categories: one involves precise control over the morphology and size of semiconductor nanocrystals, and the other involves the composite integration of QDs with optical nanocavities. Among these, composite systems of QDs and plasmonic nanocavities demonstrate excellent regulatory effects [27]. They can not only enhance the spontaneous emission rate of QDs by several orders of magnitude, resulting in a significant increase in photoluminescence, but also, through asymmetric structural design of the plasmonic nanocavities, achieve directional emission enhancement, thereby effectively improving the light emission regulation capability of QDs [28,29].

This work constructs a tilted nanocavity structure composed of a silver nanocube, CdSe/CdS nanorods, and a silver microplate. This structure combines an extremely small mode volume with ultra-low Ohmic loss characteristics. By significantly enhancing the radiation rate at the emission band and markedly increasing the electric field strength at the excitation band, it achieves a 187-fold luminescence enhancement of the CdSe/CdS nanorods. Simultaneously, leveraging the nano-antenna effect of the tilted nanocavity enables upward-directed fluorescence emission, effectively improving far-field light extraction efficiency. The efficient and directional emission control of QDs realized by this structure provides new structural design concepts for the integrated application of QDs in display devices, quantum communication, and nanolasers, and is expected to promote performance optimization and technological development of related optoelectronic devices.

2. Materials and Methods

Atomically flat single-crystal silver microplates were rapidly prepared using a wet chemical method. The required reagents for the experiment included analytically pure 4-Methylaminophenol sulfate, Metol, and silver nitrate (AgNO_3). First, 10.07 mg Metol and 10.15 mg 4-Methylaminophenol sulfate were dissolved in 10 mL of ultrapure water to form a reducing agent mixture, while 9.97 mg silver nitrate was dissolved in 10 mL of ultrapure water in the dark to prepare a silver nitrate solution. The two solutions were then mixed at

a 5:4 volume ratio by slow, dropwise addition into an amber glass bottle using syringes, and the reaction was allowed to proceed at room temperature under dark conditions to reduce silver ions to elemental silver, which gradually deposited into atomically flat single-crystal silver microplates. After the reaction was completed, the supernatant was removed, and the precipitate was washed with ethanol five times to remove unreacted impurities and solvent residues. The retained silver microplates were rinsed with ethanol, collected, and finally dispersed in ethanol. Silver nanocubes were bought from Supplier Nanoseedz. Particle size analysis of the silver nanocubes shows their average edge length is 82 ± 5 nm (Figure S1). We obtained CdSe/CdS nanorods from our collaborators. The synthetic procedure was adapted from previously reported methods [30,31]. Particle size analysis results show their average diameter is 6 ± 1 nm (Figure S1).

This paper adopts a bottom-up approach to construct plasmonic nanocavities. First, a diluted solution of single-crystal silver microplates is deposited onto glass or silicon substrates. Then, the CdSe/CdS nanorods solution is diluted in alcohol to a concentration of 3 mM, and 10 μ L of the diluted CdSe/CdS nanorods solution is spin-coated onto the single-crystal silver microplate substrate at 1000 rpm for 60 s. Afterward, the assembled samples are dried at 50 °C for 20 min. Next, a diluted solution of silver nanocubes is dropped onto the dried substrate, followed by another drying step to ensure that the silver nanocubes and CdSe/CdS nanorods adhere to the silver microplate, forming on-chip plasmonic nanocavity structures. The gap size of the nanocavities is primarily controlled by depositing Al₂O₃ of varying thicknesses. Al₂O₃ spacer layers were deposited by atomic layer deposition at 150 °C using trimethylaluminum and water as precursors, with a pulse time of 0.1 s and a purge interval of 5 s, yielding a growth rate of approximately 0.1 nm per cycle that enables precise control over the gap thickness.

For in situ single-particle dark-field scattering measurements, a halogen–tungsten lamp emitting broadband light from 400 to 1000 nm was used to illuminate the sample at an angle of 30 degrees relative to the substrate. The scattered light from the sample was then collected through an objective lens (Olympus Corporation Shinjuku, Tokyo, Japan, 50 $\times$) with a numerical aperture (NA) of 0.5. After being directed into an optical camera, multiple scattering spots with different colors emitted from various nanocavities could be observed. By moving the target nanocavity into the collection range and directing its scattered light into a spectrometer (HORIBA, Palaiseau, France, LabRAM HR Evolution), the dark-field scattering spectrum of a single nanocavity could ultimately be obtained.

Fluorescence spectral measurements were performed using a self-built confocal fluorescence microscopy system. A 532 nm continuous laser (Cobolt, Solna, Sweden, 06-MLD-532) served as the excitation source to illuminate the sample. The microscope objective (100 $\times$, NA = 0.9, Olympus) focused the laser onto the sample while collecting the emitted light, and a CCD was used to image both the sample and the emitted fluorescence. The emitted light was directed through a dichroic mirror to a spectrometer (HORIBA, Palaiseau, France, iHR550), which ultimately measured the intensity after spectrally resolving the fluorescence.

3. Results and Discussion

3.1. Design of CdSe/CdS Nanorod-Coupled Tilted Nanocavity

The construction of CdSe/CdS nanorod-coupled tilted nanocavity involves using single-crystal silver microplates with atomically smooth surfaces and low Ohmic losses as the substrate for the nanocavities. A single CdSe/CdS nanorod was placed within the nanocavity formed by the silver microplate and a silver nanocube (Figure 1a). At this stage, the silver nanocube was tilted at a small angle, thereby forming an asymmetrical plasmonic nanocavity structure. Compared to perfectly parallel nanocavities, the mode

volume is significantly compressed, offering a more localized electromagnetic field. These CdSe/CdS nanorods, with a diameter of 6 nm and a length of hundreds of nanometers, not only ensure a minimal mode volume but are also highly suitable for constructing such tilted nanocavities. The successful fabrication of the tilted nanocavities is confirmed by the characterization that one half of each CdSe/CdS nanorod is located at the bottom of the silver nanocube while the other half extends outside the cavity. In contrast, for conventional spherical QDs, additional destructive characterization techniques would be required to verify the successful fabrication of tilted nanocavities [9]. The silver nanocubes used in the experiment have an edge length of approximately 82 ± 5 nm, and their transmission electron microscopy (TEM) characterization is shown in Figure 1b. The diameter of the CdSe/CdS nanorods is about 6 ± 1 nm (Figure 1c). Full-view scanning electron microscopy (SEM) images (Figure 1d) and local magnified images of individual nanocavity on the silver microplate demonstrate that the CdSe/CdS nanorods-coupled tilted nanocavity were successfully constructed.

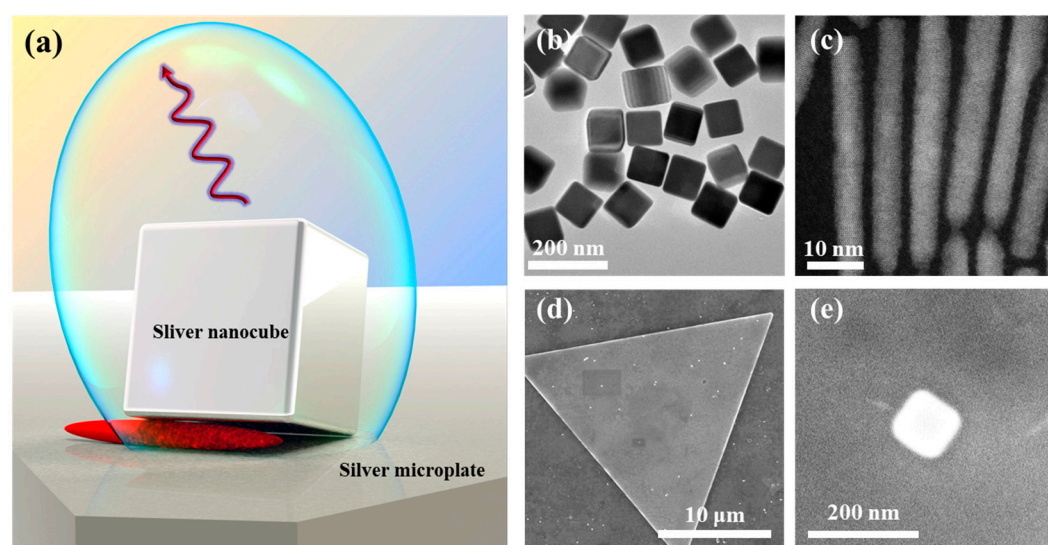


Figure 1. (a) Schematic of CdSe/CdS nanorods coupled with an inclined nanocavity; (b) TEM Characterization image of Silver Nanocubes; (c) TEM characterization image of CdSe/CdS nanorods; (d) SEM characterization image of silver nanoplate coupled nanocavities; (e) SEM characterization image of a single CdSe/CdS nanorod-coupled tilted nanocavity.

3.2. Control of Plasmonic Resonance Modes in CdSe/CdS Nanorod-Coupled Tilted Nanocavities

A schematic diagram of plasmonic tilted nanocavity enhancement QD fluorescence is shown in Figure 2a. By coordinating the plasmon mode of the nanocavity with the 620 nm fluorescence emission of CdSe QDs, the LDOS in the nanocavity can be significantly increased by the Purcell effect, thereby increasing the radiation rate of the resonance wavelength and achieving a significant enhancement of the red-light emission of QDs at 620 nm. Since the plasmonic mode of the nanocavity depends on the structure of the nanocavity and the surrounding media environment, the plasmon mode of the tilted nanocavity can be tuned to the fluorescence emission wavelength of the QD by changing the gap of the nanocavity. First, we use atomic layer deposition to plate a pure Al_2O_3 isolation layer to control the resonance wavelength of the nanocavity without introducing QDs. As shown in Figure 2b, the nanocavity plasmon formant shifts from 620 nm to 600 nm when the gap of the nanocavity changes from 10 nm to 16 nm. Therefore, in the experiment of introducing a QD, the thickness of the 10 nm isolation layer is subtracted from the 6 nm QD diameter, and we choose to plate an additional 4 nm isolation layer in the nanocavity for subsequent experiments.

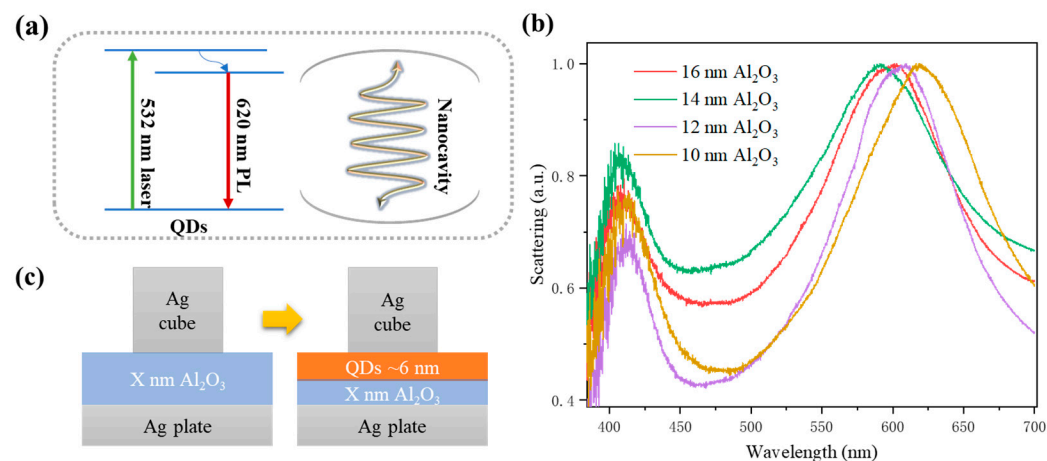


Figure 2. (a) Schematic diagram of the principle of a tilted plasmonic nanocavity enhancing QD fluorescence; (b) in situ single-particle dark-field scattering spectra of the tilted nanocavities with different gap sizes; (c) schematic diagram of the CdSe/CdS nanorod-coupled tilted nanocavity design.

3.3. Plasmonic Tilted Nanocavity Enhanced QD Emission

To quantify the strength of light–matter interaction in the constructed tilted nanocavity, the fluorescence emission spectra of single CdSe/CdS nanorod coupled to the tilted nanocavity system were measured. Under excitation with a 532 nm continuous laser, the fluorescence emission within the tilted nanocavity was significantly enhanced compared to the fluorescence of QDs deposited on a glass slide. Figure 3a shows the fluorescence spectra in three randomly selected tilted nanocavities and the QDs on a glass slide, where the red curve represents the fluorescence spectrum of QDs on the glass slide and has been amplified tenfold for easier comparison. To improve the reliability of the fluorescence enhancement factor, we have tested another 5 randomly selected nanocavities and measured their PL spectra. By comparing the average integrated intensity of the red emission of QDs at 620 nm, a direct observable fluorescence enhancement factor ranges from 134.95 to 281.35 was obtained, with an average of 187.37 and a standard deviation of 56.75. It is noteworthy that a certain splitting phenomenon was observed at the peak of the QD fluorescence spectrum in the nanocavity. We further examined the in situ single-particle dark-field scattering spectra of the nanocavity after introducing the QDs and found that the dark-field scattering spectrum also exhibited some splitting (Figure 3b). The weak dip at the emission peak (620 nm) is not a precursor to strong coupling, but a result of intermediate coupling between CdSe nanorod excitons and plasmonic modes—consistent with the Fano interference phenomenon in plasmon-exciton hybrid systems reported in two-dimensional materials [32]. Although they do not reach the stage of strong coupling, this intermediate coupling state between the nanocavity and the QD provides a fluorescence enhancement beyond the weak coupling regime for nanocavity-mediated control of QD fluorescence.

To quantitatively understand the coupling mechanism between the plasmonic nanocavity and the QDs, we moved beyond static field approximations and calculated the LDOS enhancement using finite-element simulations (COMSOL Multiphysics 6.1). The modification of the spontaneous emission rate of an emitter placed in a nanostructured environment is governed by Fermi's golden rule. In our simulations, we quantified this transition rate enhancement by calculating the Purcell factor via the integration of the energy flow density radiated by a dipole within the gap. Figure S3a displays the resulting Purcell factor spectrum, which reveals a strong resonant enhancement in the 600–700 nm range, confirming that the nanocavity acts as an efficient effector to accelerate the radiative decay of the QDs.

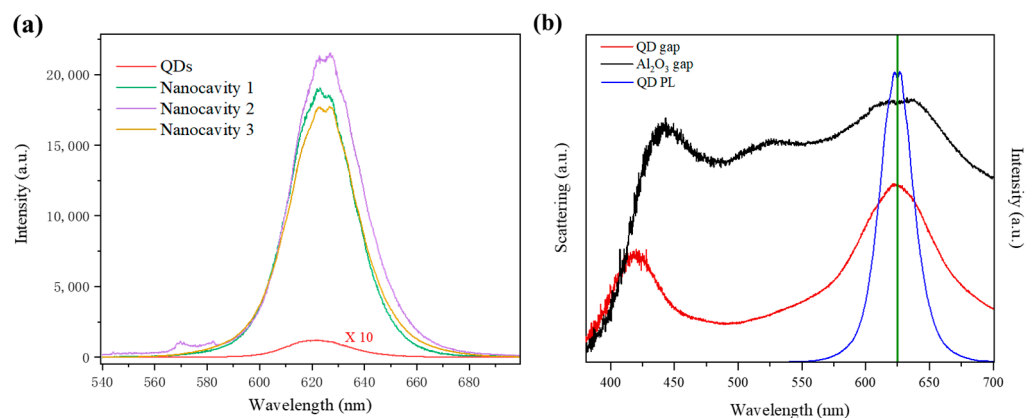


Figure 3. (a) Fluorescence spectra of the CdSe/CdS nanorods coupled with the tilted nanocavities and on a glass slide; (b) In situ single-particle dark-field scattering spectra of the nanocavities coupled with and without QDs. The green line indicates the fluorescence emission peak of the QDs.

To further validate the accuracy of our simulation model—specifically the applicability of the frequency-dependent permittivity of silver from Johnson and Christy [33]—we compared the calculated scattering cross-section (Figure S3b) with the experimental dark-field scattering spectra (Figure 3b). The simulated scattering peak appears at approximately 630 nm, showing spectral alignment with the experimental scattering resonance and the intrinsic photoluminescence of the QDs. It is worth noting that while non-local and quantum tunneling effects become significant in sub-nanometer gaps [34,35], recent studies indicate that for gap sizes exceeding 2–3 nm—such as the ~5 nm gap utilized in this work—the classical local response approximation remains valid [36,37].

3.4. Plasmonic Nanocavity-Controlled Far-Field Directional Emission of QD

In addition to enhancing the fluorescence of emitters, CdSe/CdS nanorods coupled with tilted nanocavities can also be regarded as a nanoscale patch antenna perpendicular to a silver microplate, possessing the ability to improve far-field light extraction efficiency. Using the finite element simulation method, we established a plasmonic nanocavity model with COMSOL Multiphysics and analyzed the directionality of fluorescence emission in the coupled nanocavity. For ease of analysis, we calculated the far-field radiation directions of nanocavities placed parallelly. The far-field radiation pattern of the coupled nanocavity system is shown in Figure 4. The angle θ represents the radiation direction of QD fluorescence in the y-z plane, where $\theta = 90^\circ$ denotes radiation perpendicular to the horizontal plane. Since an objective lens with a numerical aperture (NA) of 0.9 was used for excitation and light collection, the maximum measurable range of emission angles is 26° to 154° , corresponding to the yellow area in the figure. The blue and green curves represent the simulated far-field radiation intensity distribution of QD fluorescence in the nanocavity and on the glass substrate, respectively. Fluorescence on the glass substrate mainly emits to the side that cannot be collected, whereas fluorescence in the nanocavity produces upward far-field emission, with most emitted photons being collected by the objective lens. The far-field light extraction efficiency increased from 13.9% on the glass slide to 97.6% in the nano-cavity through the directional emission generated by the nano-antenna effect. Therefore, this nanocavity-coupled QD structure with highly directional far-field emission is particularly suitable for light collection.

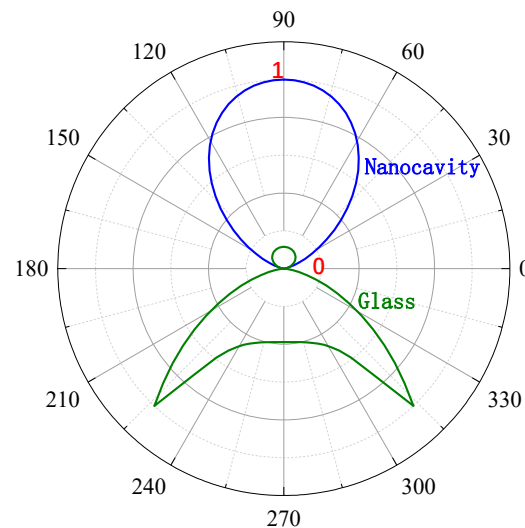


Figure 4. Far-field radiation simulation of the coupled nano-cavity system.

4. Conclusions

This study successfully constructed a silver nanocube–CdSe/CdS nanorod–single-crystal silver microplate tilted plasmonic nanocavity system and systematically investigated the regulatory effects of the coupled system on QD fluorescence. By precisely controlling the thickness of the Al_2O_3 spacer layer, the plasmonic resonance mode of the nanocavity was tuned to resonate with the 620 nm fluorescence emission of the QDs. Through the synergistic effects of local density of states enhancement induced by the Purcell effect, increased radiative rate, and strengthened excitation field, a 187-fold fluorescence enhancement of QDs was achieved. Additionally, the nanocavity's nanoantenna function significantly improved the light extraction efficiency, addressing the low fluorescence collection efficiency issue affecting conventional QD devices. This study provides a feasible approach for optimizing the performance of QD-based optoelectronic devices and holds promise for promoting their practical applications across multiple fields.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/nano16040280/s1>, Figure S1: Size distribution of silver nanocube and CdSe/CdS nanorod; Figure S2: SEM characterization image of the tilted nanocavities; Figure S3: Numerical simulations of the plasmonic nanocavity properties.

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Abbreviations

The following abbreviations are used in this manuscript:

LDOS	local density of states
QD	quantum dot
AgNO ₃	silver nitrate
AA	acrylic acid
SEM	scanning electron microscope
TEM	transmission electron microscope

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