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Optical properties of new hybrid nanoantenna in submicron cavity

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Abstract. An essential area of nanophotonics is the creation of efficient quantum emitters operating at high frequencies. In this regard, plasmon nanoantennas based on nanoparticles on metal (nanopatch antennas) are incredibly relevant. We have created and investigated a new hybrid nanoantenna with a cube on metal and quantum emitters. We demonstrate an increase up to 60 times for the rate of spontaneous emission and the gap-plasmon mode changing for nanopatch antenna in the metallic well. The results show the possibility of creating plasmon antennas in a controlled way by creating an array of regularly arranged nanoscale cavities-resonators.

1. Introduction

Nanoparticles on metal (nanopatch antennas, NPA) have been actively studied for more than 5 years and have shown their effectiveness for nanophotonics tasks [1]. Thus, high spontaneous emission rates (up to 10-100 ps) were obtained for various emitters, such as organic molecules [2,3], quantum dots [4–6], and nanodiamonds with NV centres [7], based on nanopatch antennas. Moreover, NPAs can provide a single-photon generation with single quantum dots and NV centres [7,8]. However, there are still several problems with this type of antenna.

The first problem is related to the uncontrolled placement of nanoparticles on the substrate since it is somewhat challenging to create nanoantennas in a given location (for colloidal nanoparticles). To solve this problem, the authors of the paper [9] propose to use the method of two-photon polymerization, fixing gold nanoparticles with emitters in a given place. The second problem is significant ohmic losses in the metal that makes up many plasmon nanoantennas [10]. Losses in metal nanoantennas can be reduced by changing the system geometry [11] or adding dielectric components [12,13]. A different approach is proposed in the research [14]. The authors show the opportunity of reducing non-radiation losses by hybridizing the nanoparticle's plasmon and the external resonator modes.

We decided to develop the idea of adding an external resonator to the nanopatch antenna. Thus, we have developed a new hybrid nanoantenna consisting of a nanocube and a round metal cavity. Previously, we showed that the nanocube absorption could decrease due to its placement



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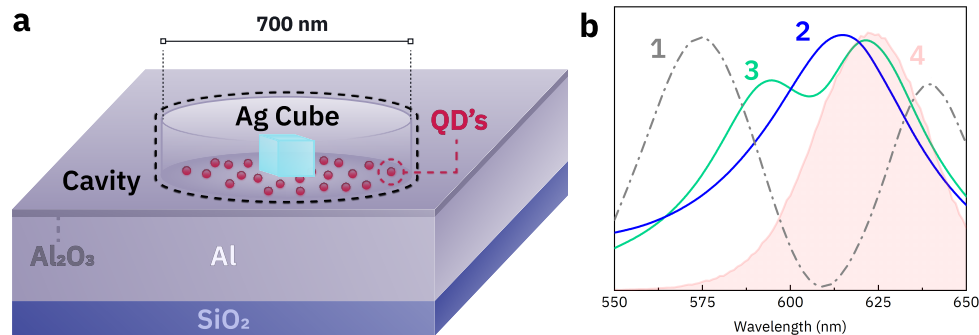


Figure 1. (a) - Diagram of a hybrid nanoantenna based on a nanocube and a circular cavity. (b) - Calculated extinction cross-section spectra of a nanocube placed in a circular cavity: the extinction cross-section spectrum of a cube located in the cavity centre (1); near the cavity wall at a distance of 55 nm (2, 3). The incident radiation is polarized in the cube-displacement direction (2) and perpendicular to the displacement relative to the cavity centre (3). Measured luminescence spectrum of CdSe/CdS quantum dots in hexane (4).

in a submicron cavity [15]. Simultaneously, the antenna's plasmon resonance could split into two resonances, independent of the exciting radiation's polarisation (Fig. 1b, curve 1) at specific parameters.

This paper experimentally shows a significant change in the QDs spontaneous emission rate and intensity placed in the hybrid nanoantenna (HNA) gap at room temperature (Fig. 1a). In this case, the studied antennas are placed in a particular structured array of a given geometry (Fig. 2).

2. Materials and Methods

Figure 1a shows a hybrid nanoantenna scheme. The nanoantenna consists of a circular aluminium cavity with a diameter of 700 nm and a depth of 300 nm. A silver cube is placed on a layer of quantum dots inside the cavity. The cube-size is about 80 nm covered by a PVP (polyvinylpyrrolidone) shell, and the QDs average diameter is about 8 nm.

Samples with an ordered array of manufactured nanoscale wells were obtained by deposition of an Al film to a Si/SiO₂ substrate by magnetron sputtering. Using the ion lithography system (FIB), the wells were etched sequentially. The Al film quality and the wells etching result were analyzed using a scanning electron microscope (SEM).

In the next step, we deposit quantum dots and silver nanocubes. CdSe/CdS quantum dots were synthesized according to the method [16]. The maximum luminescence wavelength of the synthesized quantum dots is 625 nm (Fig. 1b, curve 4).

The obtained samples were examined on a scanning fluorescent confocal microscope using an 100×/1.40 oil immersion objective with a numerical aperture (NA) of 1.4. The quantum dots in hybrid nanoantennas were excited by a diode laser at a wavelength of 375 nm (pulse mode). The radiation was collected on a single photon avalanche diode with pre-filtering of the reflected laser radiation.

3. Results and discussions

The study of the array of holes showed both bright and dim centres (Fig. 2d). This phenomenon must be due to the presence or absence of nanocubes in the cavities. To accurately determine cubes presence in the cavities, the sample was examined on a scanning electron microscope (Fig. 2a-c).

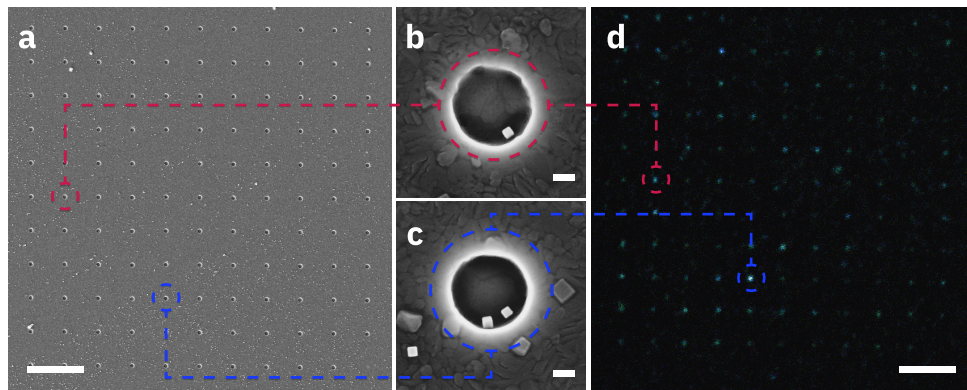


Figure 2. (a) - Scanning electron microscope image of an array of cavities and individual cavities (b, c) with applied quantum dots and cubes. The scale bars are $10\ \mu\text{m}$ and $200\ \text{nm}$, respectively. (d) - Fluorescence-lifetime image (FLIM) of an array of cavities. The scale bar is $10\ \mu\text{m}$.

Based on the measurement of the bright points fluorescence, we obtained kinetic curves. Figure 3a shows examples of the kinetic emission spectra of quantum dots in different environments. For QDs placed on glass, the main luminescence decay time is $\tau_{\text{glass}} = 6,2 \pm 0,3\ \text{ns}$. When a layer of quantum dots is placed on a metal, the radiation change properties dramatically. The spontaneous emission time decreases to $\tau_{\text{metal}} = 0,76 \pm 0,02\ \text{ns}$. Simultaneously, the radiation intensity decreases that indicates the luminescence quenching effect by metal.

The emission of quantum dots in hybrid nanoantennas shows a time reduction of 1-2 orders of magnitude ($\tau_{\text{HNA}} = 0,13 \pm 0,01\ \text{ns}$). In this case, the intensity in cavities with cubes is up to 5 times higher than the radiation of quantum dots in wells without nanoparticles. In Figure 3b, we show the intensity distribution of the measured cavities as a function of the luminescence decay time. The figure shows that some selected cavities luminescence time without cubes (red dots) is close to the radiation time of the layer of quantum dots on the metal.

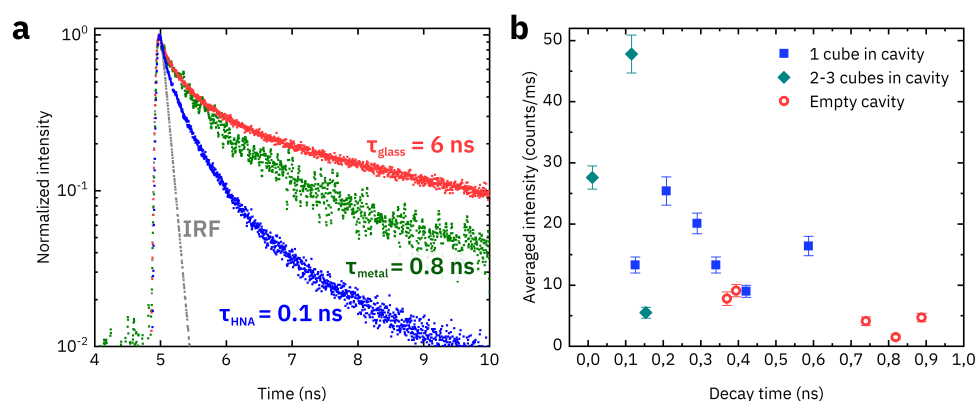


Figure 3. (a) - Comparison of the measured time-resolved fluorescence decay for quantum dots on glass (red curve), on metal (green curve), and in the HNA gap (blue curve). The grey curve is marked by the IRF detector. (b) - Comparison of the luminescence intensity of quantum dots from the characteristic time of spontaneous emission in different environments: blue squares – a cavity with one cube, green diamonds - cavities with several cubes, and hollow circles - cavities without cubes.

We also obtained brighter cavities with a short spontaneous relaxation time (about 0.4 ns) without cubes. This luminescence enhancement can be caused by the cavity optical resonance, which lies in the region of 600-610 nm. This condition can change the rate of luminescence decay, but the effect of this gain is lower compared to the gain from the plasmon resonance of the cube on the metal.

Note that the cubes were located not in the cavity centre in the resulting sample but closer to the wall. If the cube is in the centre, then two-hybrid resonances occur in this system. The resonance splitting effect weakens if the cube is located near the cavity wall (Fig. 1b, curves 2-3). We show that in this case, splitting is observed only in the case when the polarization of the incident radiation is perpendicular to the cube-displacement relative to the centre of the cavity (Fig. 1b, curves 3). Thus, the antenna resonance overlap with the QD-layer radiation in the antenna gap may change. This effect explains the difference in the intensity of individual cavities antenna radiation (Fig. 3b).

4. Conclusion

This work demonstrated a new hybrid nanoantenna with quantum emitters and showed a spontaneous emission rate increase up to 60 times. The results show the possibility of creating plasmon antennas in a controlled way by creating a cavities array. Moreover, this system can be combined with other structures, such as the bull's-eye system [17]. The proposed mechanisms open up new ways to solve the main problems of using plasmon nanostructures in photonics to create fast, ultra-bright photon sources with controlled resonance.

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