



Polarization-dependent fluorescence of CdSe/ZnS quantum dots coupling to a single gold-silver alloy nanotube



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ABSTRACT

The fluorescence intensities of CdSe/ZnS quantum dots on a silver nanowire and a gold-silver alloy nanotube with different molar ratios of gold and silver are investigated under excitation with different polarization angles. The ratio of the fluorescence intensity of perpendicular and parallel polarization excitations is measured to be 4.05 for a silver nanowire and decreases to 1.30, 1.10 and 1.07 for nanotube with Au:Ag molar ratios of 1:4.0, 1:2.5 and 1:1.5, respectively. The numerical simulation results show that the aforementioned experimental phenomena can be attributed to the variation in the distribution of the surface plasmon electromagnetic field under perpendicular polarized light excitation. The electromagnetic field is primarily distributed on the outer surface of the silver nanowire, while the field partially penetrates the interior of the silver nanotube and becomes completely concentrated at the interior of the gold nanotube. This research also found that the productivity and robustness of the chemical synthesis of the alloy nanotubes can be greatly improved by using a microwave reactor.

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1. Introduction

Control over surface plasmon is essential in a variety of plasmonic applications, such as ultrasensitive biosensors [1], energy harvesting and conversion techniques [2,3], high-resolution imaging [4], and plasmonic waveguides [5]. In addition to the size [6], morphology [7], composition and coupling of the metal nanostructures themselves [8–10], there are some other factors that can influence the surface plasmon electromagnetic field. Firstly, by altering the dielectric environment of the metallic nanostructure through hybridization [11,12], the use of an electrical field or different dielectric materials as a coating or covering on the metal [13–15], the frequency and intensity of the local surface plasmon can be altered [11–15], and the propagation of the surface plasmon can travel in a described manner [5]. Secondly, an increase in temperature can also result in both a redshift and a stronger field intensity of the surface plasmon resonance at hot spot in the dimer [6]. In addition, by applying an external dc bias across a narrow gap between the coupled metal nanostructures, the plasmonic resonances of the system can be modified in a reversible manner [16].

The surface plasmon inherits one important property from photons: polarization. Therefore, controlling the polarization of the excitation light is another effective and simple pathway towards controlling the electromagnetic field of the surface plasmon. The polarization of the excitation light not only affects the excitation efficiency of the surface plasmon of some metal nanostructures [17,18], but also affects the coupling efficiency between two adjacent metal nanostructures [9,10]. Furthermore, by precisely controlling the polarization of the incident light, specific surface plasmon modes can be obtained from self-assembled gold nanoparticle clusters [19], the coupling surface plasmon mode can be selected from integrated nanoantennas [20], and the directionality of launched surface plasmon polaritons can be continuously tuned [21]. In contrast, surface plasmon polaritons have also been reported to yield polarization conversions due to the phenomenon of the polarization dependence of the surface plasmon [22].

Metallic nanotubes represent a class of hollow nanostructures with a unique set of features that are different from their solid counterparts, including a high surface-to-volume ratio, a low density, a superior load-carrying ability, and tunable optical properties [23]. In addition, metal nanotubes possess two interfaces: a metal interface and a dielectric interface. Consequently, the surface plasmon for nanotubes are thought to be different from those of

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nanowires. Certain unique properties of the surface plasmon of metallic nanotubes have been discovered through numerical simulations [24–27], i.e., hybridization of the plasmon modes for concentric nanoshells [24], non-local dielectric effects in Au nanoshells [25], optical forces on silver nanotubes [26], and a set of morphology-dependent resonances due to the total internal reflection of light at the surface [27].

In this paper, we comparatively investigate the dependence of fluorescence intensities from CdSe/ZnS core/shell quantum dots (QDs) on a single silver nanowire and on gold-silver alloy nanotubes with different gold-to-silver molar ratios under the excitation of polarized light. To confirm the experimental results, the electromagnetic field simulation of a silver nanowire, a silver nanotube and a gold nanotube are also presented. Additionally, a modified chemical method for preparing gold-silver alloy nanotubes without aggregation is presented.

2. Experimental section

2.1. Preparation of silver nanowires and CdSe/ZnS QDs

Ag nanowires were prepared using a solvothermal method. Briefly, 10 μL of an ethylene glycol (EG) solution containing 0.1 M FeCl_3 was added to 10 mL of an EG solution of polyvinylpyrrolidone (0.166 g) under vigorous stirring. Then, with a syringe, the prepared

mixed solution was injected drop by drop into 10 mL of an EG solution of AgNO_3 (0.17 g). The final mixed solution was placed in a 25-mL Teflon-lined autoclave tube. The tube was sealed, heated at 160 $^\circ\text{C}$ for 2.5 h and then cooled naturally to room temperature. For the measurements, Ag nanowires were washed and collected by adding a large amount of ethanol, followed by centrifugation. Spherical CdSe/ZnS QDs with a diameter of 5–6 nm in chloroform were prepared by an organometallic synthetic approach [28].

2.2. Preparation of silver-gold alloy nanotubes

Silver-gold alloy nanotubes were prepared by using silver nanowires as sacrificial templates in the replacement reaction with a slight modification of the process with the introduction of a microwave reactor for heating the solution [29]. Typically, 2 mL of as-prepared silver nanowires in an EG solution was mixed with 50 mL of deionized water under stirring and then heated to boil in the microwave reactor. Next, 1 mL of a deionized water solution of 1 mM HAuCl_4 was injected into the reaction solution. The mixed solution was taken into the microwave reactor again for continuous boiling for 3 min. Finally, gold-silver alloy nanotubes were collected by centrifugation for further experiments and characterization. The molar ratios of Au:Ag in the alloy nanotubes were adjusted by altering the amount of HAuCl_4 in the replacement reaction.

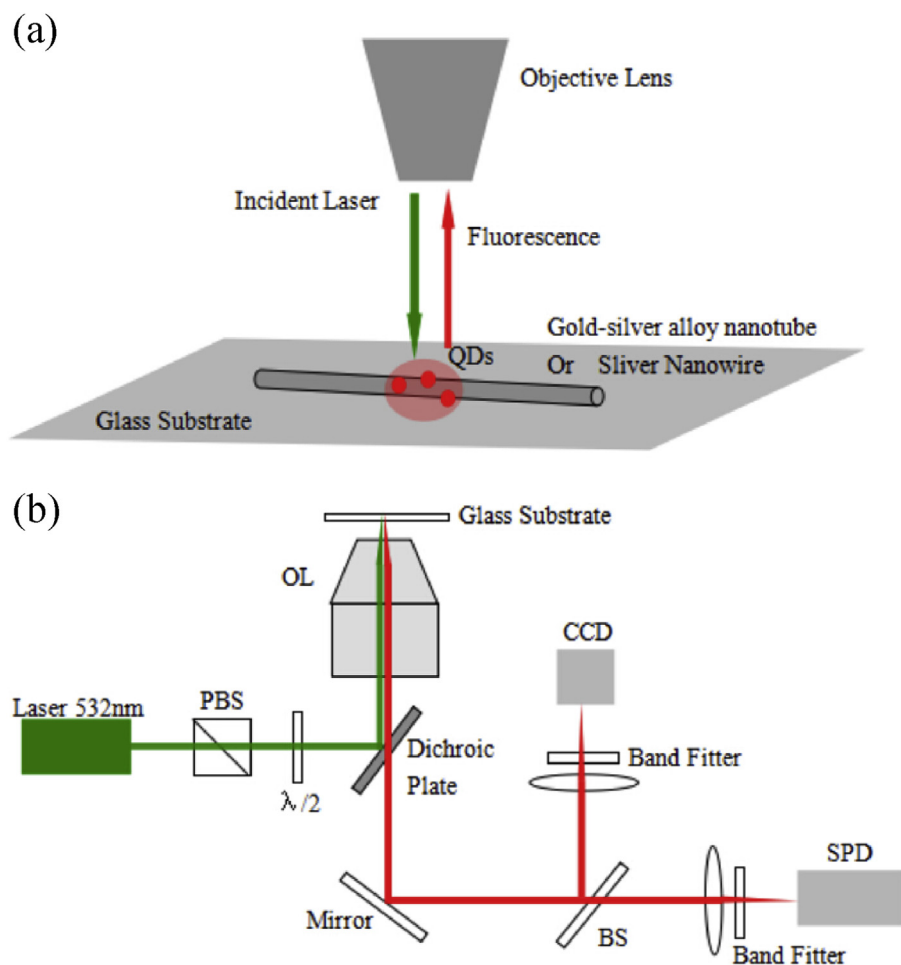


Fig. 1. Schematics of (a) the sample and (b) the experimental setup. The pump laser illuminates the sample from the top, and the fluorescence signals of the QDs are collected using the same objective lens. CdSe/ZnS QDs and the nanowire or nanotubes are deposited onto a glass substrate.

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