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Influence of localized surface plasmon in a lens-shaped metal cluster on the decay dynamics of a point-dipole emitter

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ABSTRACT

Influence of localized surface plasmon in a lens-shaped metal nanoparticle on the decay dynamics of a point-dipole emitter was studied by means of numerical simulations. Dependencies of radiative and nonradiative decay rates on the distance between the particle and the dipole were calculated for dipole and quadrupole plasmon modes. The presence of the etching cone changes the nonradiative dynamics of a dipole located under a metal cluster and can be a convenient tool to reduce the surface influence on emitters. The cone also selectively enhances the nonradiative decay rate for the emitter located under the vertex enabling preferential plasmon coupling in ensembles of emitters.

1. Introduction

Interaction between plasmonic modes in metal nanoparticles and quantum emitters like color centers in crystals or semiconductor quantum dots (QDs) is of great interest for nanophotonics and quantum optics. Local enhancement of electric field caused by excitation of localized surface plasmons can result in a decrease of the life-time and significantly improve the yield of the emitter [1–3]. For that, thorough tuning of size, compound and geometry of a plasmonic particle that influence the resonance position is required. Also, mutual positioning of the particle and the emitter should be properly arranged taking into account the distribution of near fields [4–7].

One of the easy ways to produce plasmonic particles is the formation of metal droplets on the structure surface. The nucleation of droplets is a self-assembly process that can replace complicated lithography procedure and thus is attractive for plasmonic applications. The fabrication of such metal droplets over a structure containing solid-state semiconductor QDs is possible directly inside the chamber of molecular beam epitaxy (MBE) setup as the initial stage of droplet epitaxy [8]. Therefore, this method allows for fabrication of quantum emitters and metal clusters in a single growth cycle suggesting metal droplets to be a perspective type of plasmonic particles. For this method the material choice is limited with MBE compatible materials namely metals of group III (In, Ga, Al) or semimetallic antimony. In our work [9] it was demonstrated that the plasmon resonance in such particles can be tuned to the emission band of InAs/AlGaAs QDs. Recently, we experimentally demonstrated exciton–plasmon interaction between subensembles of InAs/AlGaAs QDs and indium clusters [10]. As the next step, the interaction between plasmon modes and an exciton in QDs should be studied taking into account the geometry of the real structure and

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mutual positioning. In this work we focus on the influence of plasmonic modes in a lens-shaped metal cluster on a point dipole emitter by numerical simulations.

2. Simulation method and model

ADDA package that is based on the discrete dipole approximation was used for simulations [11]. In this approach, an object of interest is described as a complex of point dipoles with their polarizabilities being derived from dielectric constants of bulk material and electrical field according to a chosen algorithm. Here, lattice dispersion relation was used [11], after that the linear system is solved to determine dipole polarizations P_i:

$$\alpha_i^{-1} P_i - \sum_{j \neq i} H_{ij} P_i = E_i^{inc}, \quad (1)$$

where E_i^{inc} is the incident electric field, α_i is the dipole polarizability, H_{ij} is the total interaction term that was chosen as interaction of point dipoles in free space, and indices i and j enumerate the dipoles. After polarizations are known, electrical field can be calculated:

$$P_i = \alpha_i E_i^{exc}, \quad (2)$$

where E_i^{exc} is a sum of E_i^{inc} and the field due to all other dipoles, but excluding the field of the dipole i itself. Then the problem of light scattering on such an object can be solved with high precision that is mainly limited by the geometrical representation.

Starting from version 1.3b4 [12], ADDA allows one to use the electrical field of a point dipole as the incident excitation [13]. Such a dipole has a certain decay rate in the free space that is modified by a metal nanoparticle presence. Then it can be separated to radiative part that corresponds to light scattering to far field and nonradiative part that is transferred to the plasmon mode and thus absorbed by the metal particle. Both decay channels are affected by the particle presence and contribute to the modified emitter dynamics [14].

In the free space, the decay cross section for a point dipole is given as $C_0 = 8\pi k^4 |p_0^2|/3$, where k and p_0 are the light wave vector and the dipole polarization, respectively. In the presence of the nanoparticle, the total decay cross section C_{tot} can be put as $C_{tot} = C_{abs} + C_{rad}$, where C_{abs} is the absorption cross section, C_{rad} is the scattering cross section of a dipole set including both ones describing the particle and the exciting dipole. Factors $F_{NR} = C_{abs}/C_0$ and $F_{Rad} = (C_{tot} - C_{abs})/C_0$ correspond to decay rate enhancements for the nonradiative and radiative decay channels of the point dipole emitter.

The metal cluster was modeled as a part of a sphere as shown in the inset in Fig. 1. The diameter of the droplet base was taken to be 340 nm with the height being 44 nm. These parameters correspond to typical ones derived experimentally by atomic force microscopy measurements of indium clusters on GaAs substrate produced by MBE in our previous work [15]. To describe the object, around 118,000 dipoles with the interdipole distance of 2.67 nm were used. It was adopted from the work [9], where it was also shown that spectral features of plasmon resonance dependencies are alike for the group III metals and antimony. Here, the indium cluster was considered as a model.

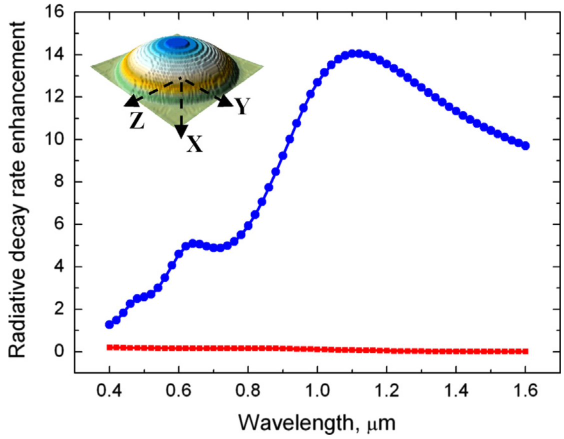

Fig. 1. Spectral dependencies of radiative decay rate enhancement. The dipole source coordinates in nanometers are (10, 170, 0) and (10, 0, 0) for blue circles and red squares, respectively. The 3D model of a lens-shaped cluster with coordinate frame is shown in the inset. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The coordinate system that was used to specify the mutual position of the dipole source and the particle is depicted in the inset in Fig. 1. The coordinate center was assigned to the droplet base center. As semiconductor QDs are buried in the matrix the positive X coordinate was attributed to the dipole source. The dipole polarization vector was put along the Y axis corresponding to the orientation of the exciton dipole moment in self-assembled QDs [16]. The coordinates of the dipole are given in nanometers. Currently, placing the dipole inside the substrate is not possible in the numerical implementation. Here, a metal particle and a dipole are considered surrounded by air that allows us to keep realistic mutual positioning. Then, in this model the presented quantitative results are likely to be upper estimates for interaction strength and lower estimates for characteristic distances in a realistic environment.

The results presented in this work have mainly qualitative character. This is determined by the limitations listed above and the essentially mesoscopic nature of QDs that is not considered in the point dipole approximation [17]. This basic approximation breaks for low distances between a cluster and a QD and the naturally limited validity of simulations for an experimental system is not affected by the demanding increase of the model precision. However, we believe that the dependencies obtained will prove to be useful and quantitative results could be used as a starting point for experimental implementations.

3. Results and discussion

The spectral dependence of radiative decay enhancement for a dipole source located 10 nm below the droplet is shown in Fig. 1. It demonstrates two resonance peaks that correspond to excitation of dipole and quadrupole