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Altering Degenerate Four-Wave Mixing and Third-Harmonic Generation in a Coupled Quantum Dot-Metallic Nanoparticle Structure with the Use of the Purcell Effect †

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Abstract: We study the modification of the phenomena of degenerate four-wave mixing and third-harmonic generation in a quantum dot that is coupled to a spherical metallic nanoparticle. We find that the strong alteration of the quantum dot's spontaneous decay rate near the metallic nanoparticle gives strong variation, either enhancement or suppression, to the phenomena of degenerate four-wave mixing and third-harmonic generation for different distances of the quantum dot from the surface of the metallic nanoparticle, depending on the electric dipole direction of the quantum dot. We also show that the degree of enhancement or suppression of the nonlinear optical susceptibilities differs for the studied phenomena and it is stronger for degenerate four-wave mixing than for third-harmonic generation. This work may have important potential applications in the creation of nanoscale photonic devices for various technological applications.

Keywords: third-order nonlinear optical susceptibility; quantum dot; metallic nanoparticle; degenerate four-wave mixing; third-harmonic generation

1. Introduction

The modification of the optical properties of semiconductor quantum dots near plasmonic nanostructures have attracted significant attention in recent years due to the several potential applications of the coupled nanostructures in optoelectronics, biophotonics and quantum technologies, including sensors, light harvesting, quantum information processing and quantum communication, imaging, photocatalysis, solar cells, and others [1]. One of the methods for modifying the nonlinear optical susceptibilities in quantum dots near plasmonic nanostructures uses the change of the spontaneous decay rates of quantum emitters due to the Purcell effect in a tailored nanophotonic environment [2–6]. This method was initially proposed by Bermel et al. [7] for quantum dots in photonic crystals and was later explored for the modification of linear and nonlinear absorption and dispersion [2,4], second- [5] and third-harmonic generation [6], and nonlinear optical rectification [3,5] of quantum dots and molecules near simple and complex plasmonic nanostructures.

In this paper, following the approach of Refs. [2–7], we study the modification of the phenomena of degenerate four-wave mixing and third-harmonic generation in a quantum dot close to a relatively large gold nanosphere. In our study we use a metallic nanosphere with radius 60 nm, which behaves as a nanophotonic environment and alters (enhances or suppresses) the spontaneous decay rate of the nearby quantum dot. The variation of the spontaneous decay rate of the quantum dot leads to strong modification to the degenerate four-wave mixing and the third-harmonic generation. Specifically, both

enhancement and suppression of the degenerate four-wave mixing and the third-harmonic generation effect can occur depending on the distance between the quantum dot and the metallic nanosphere. The direction of the electric dipole of the quantum dot strongly influences degenerate four-wave mixing and third-harmonic generation. We find that the degree of enhancement or suppression of the nonlinear optical susceptibilities differs for the studied phenomena, and it is stronger for degenerate four-wave mixing than for third-harmonic generation.

2. Methods and Materials

We consider a quantum dot placed near a spherical metallic nanoparticle and the whole system is embedded in a dielectric background with relative dielectric constant ϵ. The metallic nanoparticle is treated as a classical particle of radius R described by a relative dielectric function $\epsilon_m(\omega)$. The system interacts with two electromagnetic fields with angular frequencies ω_1 and ω_2. We study a spherical metallic nanoparticle made of gold with radius 60 nm. The electromagnetic response of the metallic nanosphere is described by the dielectric function $\epsilon_m(\omega)$ of Ref. [8]. For the background we consider an isotropic dielectric with relative dielectric constant $\epsilon = 2.25$.

For the semiconductor quantum dot, which is placed at distance d from the surface of the spherical nanoparticle along the x-axis, we consider the excitonic transition of a GaAs/AlGaAs quantum dot with transition energy $\hbar\omega_{10} = 1.517$ eV and electric dipole matrix element $\mu_{01} = 1.2\, e$ nm. For the spontaneous decay rate of the exciton in the absence of the metallic nanoparticle we take $\gamma_0 = 10^{10}$ s^{-1}. The quantum dot's pure dephasing rate is zero in this study. For the calculation of the quantum dot's spontaneous decay rate we use the MNPBEM toolbox [9].

3. Results and Discussion

We place the quantum dot at various distances d from the surface of the spherical metallic nanoparticle, along the x-axis. In Figure 1 we present the modification in the quantum dot's spontaneous decay rate for two different quantum dot's electric dipole directions, along the x-axis (γ_x) and along the z-axis (γ_z), as a function of distance d. The obtained behavior is typical, see, for example, refs. [4–6,9,10]. Both decay rates are enhanced for small distances between the quantum dot and the metal nanoparticle, but as this distance increases the decay rate corresponding to the electric dipole direction along the x-axis decreases monotonically and the value of the decay rate remains larger than the spontaneous decay rate in the absence of the metallic nanoparticle γ_0. For electric dipole direction along the z-axis, with the increase of the distance between the quantum dot and the metallic nanoparticle, the value of the decay rate shows a rapid decrease and can become quite smaller than the spontaneous decay rate in the absence of the metallic nanoparticle γ_0. After a specific distance, about $d \approx 23$ nm, the quantum dot's spontaneous decay rate increases again, but its value remains lower than γ_0 in the regime we present here.

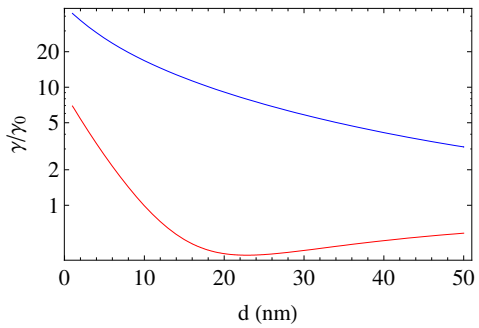

Figure 1. The quantum dot's spontaneous decay rate as a function of the distance d from the surface of a spherical gold nanoparticle with radius 60 nm. We consider two different directions of the electric dipole moment of the quantum dot. The dipole moment is either along the z-axis (red curve) or along the x-axis (blue curve).

Then, in Figures 2 and 3 we present results for the third-order nonlinear optical susceptibility [11] and specifically for the phenomena of degenerate four-wave mixing and third-harmonic generation for a quantum dot close to the metallic nanosphere. For the modification of the decay rates by the metallic nanosphere we use the results of Figure 1. In Figure 2a we present the results for the degenerate four-wave mixing coefficient $|\chi^{(3)}_{DFWM}|$ as function of the photon energy $\hbar\omega$ for quantum dot's electric dipole direction along the z-axis. We note that we obtain either suppression or enhancement of the degenerate four-wave mixing coefficient, in comparison to the value in the absence of the metallic nanoparticle (red curve), depending on the value of the distance d. The suppression of the degenerate four-wave mixing coefficient occurs for $d = 5$ nm (green curve). For $d = 10$ nm (blue curve) the value of the of the degenerate four-wave mixing coefficient is almost the same as the value in the absence of the metallic nanoparticle. The enhancement of the degenerate four-wave mixing coefficient occurs for all other distances, with the largest enhancement, about eighteen times in comparison to the value in the absence of the metallic nanoparticle, found for $d = 25$ nm (turquoise curve). In addition, the widths of the various degenerate four-wave mixing coefficients vary when altering the distance d, and we obtain the greatest width for $d = 5$ nm, which coincides with the strongest suppression, while the narrowest width is found for $d = 25$ nm, which coincides with the largest enhancement.

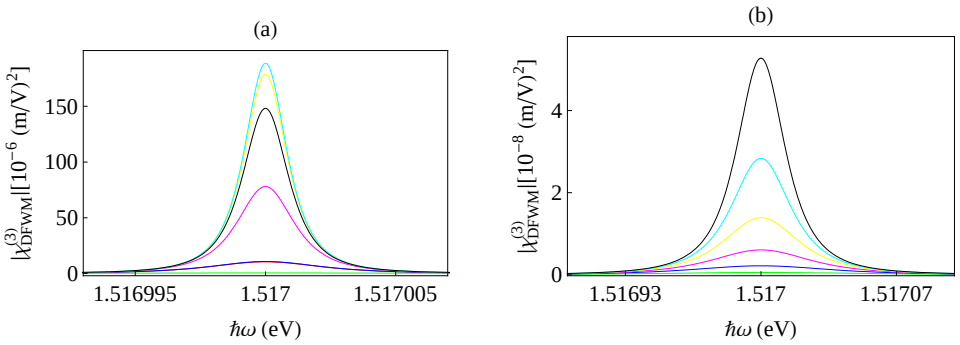

Figure 2. The degenerate four-wave mixing coefficient $|\chi^{(3)}_{DFWM}|$ as a function of the photon energy $\hbar\omega$ for a quantum dot with electric dipole orientation along the z-axis (**a**) and along the x-axis (**b**) and for different distances from the surface of the metallic nanosphere: $d = 5$ nm (green curve), $d = 10$ nm (blue curve), $d = 15$ nm (magenta curve), $d = 20$ nm (yellow curve), $d = 25$ nm (turquoise curve), and $d = 30$ nm (black curve). The red curve in (**a**) describes the case in the absence of the metallic nanosphere.

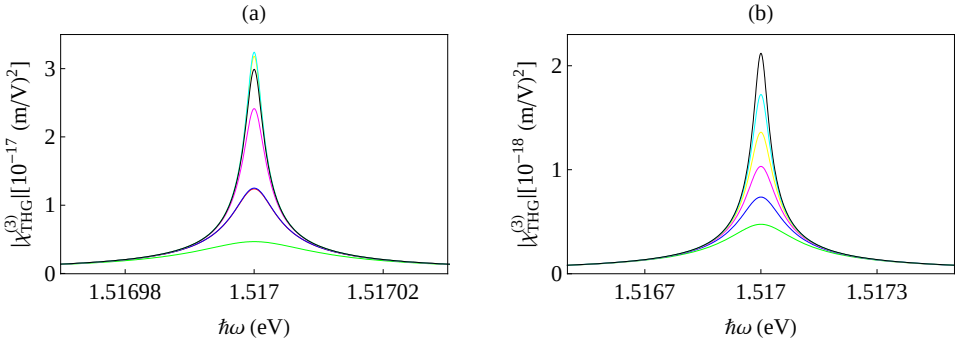

Figure 3. The third-harmonic generation coefficient $|\chi^{(3)}_{THG}|$ as a function of the photon energy $\hbar\omega$ for a quantum dot with electric dipole orientation along the z-axis (**a**) and along the x-axis (**b**) and for different distances from the surface of the metallic nanosphere: $d = 5$ nm (green curve), $d = 10$ nm (blue curve), $d = 15$ nm (magenta curve), $d = 20$ nm (yellow curve), $d = 25$ nm (turquoise curve), and $d = 30$ nm (black curve). The red curve in (**a**) describes the case in the absence of the metallic nanosphere.

Next, in Figure 2b we study the dependence of the degenerate four-wave mixing coefficient on the quantum dot's electric dipole orientation and consider a x-oriented electric dipole. We find that all the results in the case that the metallic nanosphere is present lead to suppression of the degenerate four-wave mixing coefficient in comparison to the case in the absence of the metallic nanoparticle [red curve of Figure 2a]. The suppression weakens with the increase of the distance between the quantum dot and the metallic nanoparticle. The widths of the different curves also change with the change of the distance, and we find greater widths for shorter distances, i.e., coinciding with larger suppression of the degenerate four-wave mixing coefficient.

The third-harmonic generation is shown in Figure 3. The behaviour of the third-harmonic generation coefficient is quantitative similar to the degenerate four-wave mixing coefficient. However, there are some qualitative differences. The maximum enhancement of the third-harmonic generation coefficient, shown in Figure 3a, is found also for $d = 25$ nm, and it is about 2.6 times in comparison to the case without the plasmonic nanoparticle for z-oriented electric dipoles. Also, the suppression of the third-harmonic generation coefficient in comparison to results in the absence of the plasmonic nanoparticle for x-oriented electric dipoles is significantly weaker than that found for the degenerate four-wave mixing coefficient.

4. Conclusions

In this work we showed that the phenomena of degenerate four-wave mixing and third-harmonic generation of a quantum dot can be tailored by coupling the quantum dot with a spherical metallic nanoparticle. We showed that the strong alteration, enhancement or suppression, of the quantum dot's spontaneous decay rate near the metallic nanoparticle can lead to strong change of the degenerate four-wave mixing and third-harmonic generation coefficients, which can be either enhanced or suppressed depending on the distance of the quantum dot from the surface of the nanoparticle and on the quantum dot's electric dipole direction. We also found that the degree of enhancement or suppression of the degenerate four-wave mixing is stronger than that of third-harmonic generation.

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