Optical Apsorption and Bistability of Semiconductor Quantum Dot-Metal Nanoparticle Dimers

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Abstract

We perform a theoretical study of the optical response of a hybrid system comprised of a closely spaced semiconductor quantum dot (SQD) and a noble metal nanoparticle (MNP). We show that this system exhibits fascinating properties originating from the strong dipole-dipole SQD exciton - MNP plasmon interaction. In the linear regime, this interaction leads to the creation of a modified exciton-plasmon excitation with a red-shifted resonance energy and a broadened energy absorption spectrum. In the nonlinear regime, the coupling results in the formation of narrow exciton-plasmon Fano resonances with tunable properties. Additionally, the presence of the MNP leads to a self-action (feedback) of the SQD that can result in bistability of the hybrid dimer optical response. We provide a detailed analysis of the bistability phase diagram.

1. Introduction

Nowadays, modern nanotechnology allows us to assemble a variety of hybrid nanocomposites. These systems have been studied for a range of applications such as light nanosources for nanophotonic devices [1] and resonance energy transfer [2]. Among ubiquitous combinations, the mostly studied hybrids are noble metal-semiconductor nanocomposites [3, 4]).

The SQD-MNP dipole-dipole interaction for closely spaced nanoparticles can be relatively strong. This coupling dramatically modifies the optical response of such hybrid systems and gives rise to a number of interesting phenomena, among them nonlinear Fano resonances and bistability [4]. The novel properties exhibited by the hybrid systems held the potential to revolutionize optoelectronics and nanophotonics.

We perform a theoretical analysis of the optical response of a simple hybrid system comprising a spherical MNP and SQD, both in the weak-field (linear) and the strong-field (nonlinear) regime. The system is assumed to be embedded in a dielectric host with permeability ε_b and driven by a linearly polarized (along the system axis) external field with amplitude E_0 and frequency ω. The SQD is modeled as a two-level system with transition frequency ω_0 and optical transition dipole moment μ. It is treated quantum mechanically within the framework of the 2x2 density matrix (DM) formalism with relaxation constants γ (population) and Γ (polarization). The MNP is considered classically via its frequency dependent

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scalar polarizability $\alpha(\omega) = a^3[\varepsilon_M(\omega) - \varepsilon_b]/[\varepsilon_M(\omega) + 2\varepsilon_b]$; a is the MNP radius and $\varepsilon_{\rm M}(\omega)$ is the dielectric function of the metal. All sizes of the system (SQD and MNP radii and center-to-center distance d) are assumed to be small enough to neglect the retardation effects and to consider both nanoparticles as point dipoles.

2. Bistability phase diagram

Within the rotating wave approximation for the DM and under steady-state conditions, it is straightforward to derive an equation for the SQD population difference $Z = \rho_{11} - \rho_{00}$. It reads

$$\frac{|\widetilde{\Omega}_0|^2}{\gamma\,\Gamma} = -\frac{Z+1}{Z}\,\frac{|(\Gamma - G_{\rm I}Z) + i\,(\Delta + G_{\rm R}Z)|^2}{\Gamma^2}\,, \tag{1}$$

$$\widetilde{\Omega}_0 = \frac{1}{\varepsilon_s'}\left[1 + \frac{2\alpha(\omega)}{d^3}\right]\Omega_0\,, \qquad G = G_r + iG_i = \frac{4\mu^2\alpha(\omega)}{\varepsilon_b\,\varepsilon_s'\,\hbar\,d^6}\,, \tag{2}$$

where $\Omega_0 = \mu E_0/\hbar$ and $\varepsilon_s' = (\varepsilon_s + 2\varepsilon_b)/(3\varepsilon_b)$. Equation (1) is of third order in Z and therefore may have three real solutions, depending on the values of the off-resonance detuning $\Delta = \omega_0 - \omega$, the polarization relaxation constant Γ, and the SQD-MNP coupling constant $G = G_r + iG_i$.

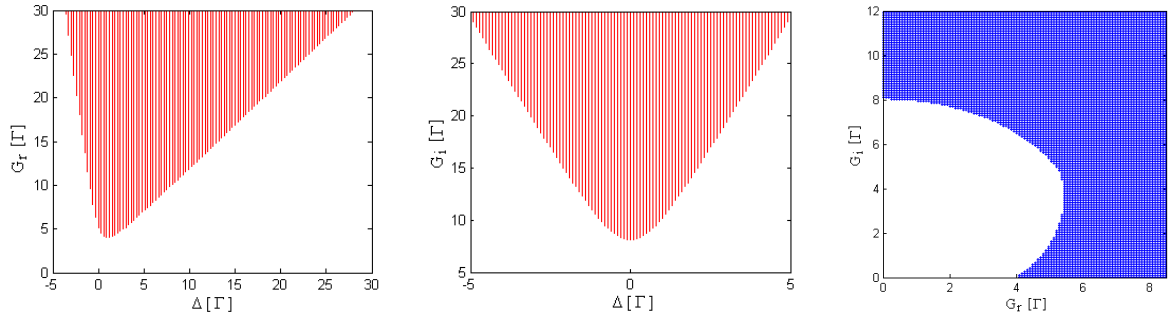

Fig. 1: Bistability phase diagrams of the SQD optical response. Shaded regions show the parameter space where bistability exists. From left to right: phase diagram in the parameter space $[G_r, \Delta]$ at $G_i = 0$, $[G_i, \Delta]$ at $G_r = 0$, and $[G_r, G_i]$ at arbitrary Δ.

Bistability phase diagrams for different set of parameters Δ, G_i, and G_r are presented in Fig. 1. From these data, we derive that both coupling constants, G_r and G_i, gives rise to bistable optical response. At $G_r = 0$, the critical value for bistability to occur is $G_i = 8\Gamma$, while at $G_i = 0$, the critical value of $G_r = 4\Gamma$. For a nonzero detuning Δ, higher values of G_r and G_i are required.

3. Spectra

The total energy Q absorbed by the hybrid system is due to the absorption by both the SQD and the MNP: $Q = Q_{\rm SQD} + Q_{\rm MNP}$. The $Q_{\rm SQD(MNP)}$ is calculated as

$$Q_{\rm SQD(MNP)} = \frac{1}{2}\omega{\rm Im}(\chi_{\rm SQD(MNP)})|E_{\rm SQD(MNP)}|^2\,, \tag{3}$$

where $\chi_{\rm SQD(MNP)}$ and $E_{\rm SQD(MNP)}$ are, respectively, the susceptibility of SQD(MNP) and the field amplitude acting on SQD (MNP). We consider a SQD in the vicinity of an Au MNP and use the following set of SQD parameters: the transition energy $\hbar\omega_0 = 2.5$ eV, the SQD transition dipole moment $\mu = 0.7\,e\cdot{\rm nm}$, the SQD relaxation constants $\gamma = 1.25$ ns^{-1} and $\Gamma = 3.33$ ns^{-1} [4], the SQD dielectric

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constant $\varepsilon_s = 6.2$, the MNP radius $a = 10$ nm, $d = 16$ nm, the background dielectric constant $\varepsilon_b = 1$. The metal dielectric constant $\varepsilon_{\rm M}$ was taken from the tabulated data for the permittivity of gold [5].

Figure 2 shows the dependence of the spectrum on the center-to-center distance d (smaller d increases the SQD-MNP dipole-dipole coupling). The left (middle) plot corresponds to the weak (strong) field limit, specified in the figure caption. In the weak limit, the absorption peak is red-shifted and broadened when reducing d, while in the strong field limit it demonstrates an asymmetric Fano shape. Moreover, the hybrid absorption reveals a discontinuity, dependent on which branch of Z it is measured (see the right panel and the insert).

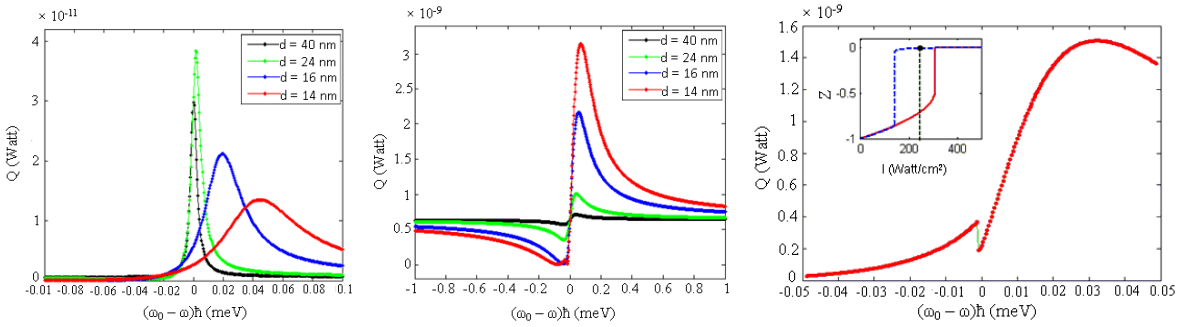

Fig. 2: Energy Q absorbed by a hybrid system comprised of a SQD and a Au MNP. Left panel: spectra calculated for different center-to-center SQD-MNP distances in the limit of low external field intensity, $I = \Omega_0^2/\sqrt{\gamma\Gamma} < 1$. Middle plot: the same, but now in the limit of high external field intensity, $I = \Omega_0^2/\sqrt{\gamma\Gamma} > 1$. Right plot: discontinuity in the absorbed energy Q when exciting the system at the upper branch of the bistability characteristic Z vs I (see the insert).

4. Conclusion

We studied the optical responce of a hybrid nanodimer comprised of a closely spaced semiconductor quantum dot and a metal nanoparticle. In the linear regime, the strong dipole-dipole SQD exciton-MNP plasmon interaction results in the formation of hybrid excitation with a shifted and broadened energy absorption peak. In the nonlinear regime, the SQD-MNP coupling results in the formation of a narrow hybrid exciton-plasmon Fano resonance, which can be controlled by changing the SQD-MNP center-to-center distance. Furthermore, the self-action of the SQD via the MNP can result in bistability of the optical response, giving rise to a discontinuity in the absorption spectra. We conducted a detailed study of the bistability phase diagram of the composite and calculated critical parameters for bistability to occur. The considered hybrid system may serve as an element of all-optical logic.

References

[1] P. Viste et al., Enhancement and quenching regimes in metal-semiconductor hybrid optical nanosources, *ACS Nano,* vol. 4, no. 2, pp. 759-764, 2010.

[2] K. K. Haldar, T. Sen, and A. Patra, Metal conjugated seniconductor hybrid nanoparticle-based fluorescence resonance energy transfer, *J. Phys. Chem. C,* vol. 114, no. 11, pp. 4869-4874, 2010.

[3] A. O. Govorov et al., Exciton-plasmon interaction and hybrid excitons in semiconductor-metal nanoparticles assemblies, *Nano Lett.*, vol. 6, no. 5, pp. 984-994, 2006.

[4] R. D. Artuso and G. V. Bryant, Strongly coupled quantum dot-metal nanoparticle systems: Exciton induced transparency, discontinuous response, and suppression as driven quantumoscillator effects, *Phys. Rev. B,* vol. 82, p. 195419-10, 2010.

[5] P. B. Johnson and R. W. Christy, Optical constants of the noble metals, *Phys. Rev. B,* vol. 6, no. 12, pp. 4370-4379, 1972.

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