Optical Dynamics of a Bistable Semiconductor Quantum Dot – Metal Nanoparticle Heterodymer

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Abstract

We perform a theoretical study of the optical dynamics of a hybrid system comprised of a closely spaced semiconductor quantum dot (SQD) and a metal nanoparticle (MNP). We show that, depending on the strength of the SQD-MNP coupling, the system can manifest bistability in the optical response. The SQD population demonstrate a hysteresis behavior upon sweeping adiabatically up and down the input intensity. We calculate the relaxation time required for the SQD population to reach its steady-state value and show that close to a critical intensity of the input, at which the SQD population switches from a lower to a higher value, the relaxation time is slowing down dramatically.

1. Introduction

Optical bistability is a fascinating nonlinear phenomenon, the essence of which is controlling the flow of light by light itself. It is of great importance for optical technologies, in particular, for optical logic and signal processing. The key ingredients for bistable response to occur are nonlinearity of the material and a positive feedback. Interplay of the two can result in a multi-valued nonlinear output within a certain range of the system parameter's space. A generic optical bistable element exhibits two stationary stable states for the same input intensity, a property which, in principle, opens the door to applications such as all-optical switches, optical transistors, and optical memories. It is of great interest to realize this phenomenon at the nanoscale. A heterodimer comprised of an SQD and an MNP provides this possibility [1, 2, 3]. From the viewpoint of the potential use of such systems in devices, the time it takes the system to be switched from one stable state to the other is of crucial importance. Studying this time is the aim of this work.

2. Model and Formalism

We consider an SQD-MNP heterodimer 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 optical transitions in the SQD occur between the valence band and discrete excitonic levels below the conduction band, requiring full quantum treatment. We will restrict ourselves to the transition between the valence band and the lowest exciton state. In the case of an MNP, the optical excitations are surface plasmons, which may be adequately described classically by its frequency dependent polarizability

$\alpha(\omega) = [\epsilon_m(\omega) - \epsilon_b]/[\epsilon_m(\omega) + 2\epsilon_b]$, where $\epsilon_m(\omega)$ is the frequency-dependent dielectric constant of the metal. All sizes of the system (SQD and MNP radii, a and r, respectively, 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.

The optical dynamics of the SQD is described using the 2×2 density matrix ρ_{mn}; $m, n = 1, 2$ denote the ground and excited SQD state, respectively. In the rotating-wave approximation, the equations of motion for the SQD read [2]

$$\dot{Z} = -\gamma(Z+1) - \frac{1}{2}\left[\left(\widetilde{\Omega}_0 - iGR\right)R^* + \left(\widetilde{\Omega}_0 - iGR\right)^* R\right] , \tag{1}$$

$$\dot{R} = -\left[(\Gamma - G_I Z) + i(\Delta + G_R Z)\right]R + \widetilde{\Omega}_0 Z \tag{2}$$

with

$$\widetilde{\Omega}_0 = \frac{1}{\epsilon'_s}\left[1 + \frac{\alpha(\omega)}{2\pi d^3}\right]\Omega_0 , \quad G = G_R + iG_I = \frac{\mu^2\alpha(\omega)}{16\pi^2\hbar\epsilon_0\epsilon_b\epsilon'_s d^6} \tag{3}$$

Here, $Z = \rho_{22} - \rho_{11}$ is the SQD population difference, R is the amplitude of the off-diagonal density matrix element ρ_{21}, γ and Γ are the relaxation constants of population and dipole moment, respectively, μ is the SQD transition dipole moment, $\Delta = \omega_0 - \omega$ is the detuning away from the SQD exciton resonance ω_0, $\Omega_0 = \mu E_0/\hbar$ is the Rabi frequency of the input field, and $\epsilon'_s = (\epsilon_s + 2\epsilon_b)/(3\epsilon_b)$ with ϵ_s being the SQD dielectric constant. The parameter $G = G_R + iG_I$ represents the SQD-MNP coupling. It is the crucial quantity in the problem, driving the optical response of the SQD-MNP heterodimer. In particular, it determines whether the system behaves in a bistable fashion or not (see below).

3. Results and discussion

The left panel in Fig. 1 shows the bistability phase diagram, the range of G_R and G_R where the bistability can occur (colored) calculated under the steady-state conditions ($\dot{Z} = \dot{R} = 0$). The boundary between white and colored regions represents the bistability threshold with respect to the parameters G_R and G_I.

The right panel in Fig. 1 demonstrates the hysteresis behavior of the SQD population difference Z obtained after solving Eqs. (1) and (2) under sweeping adiabatically up and down of the input intensity $I = |\Omega_0|^2/(\gamma\Gamma)$ (marked by the arrows). The S-shaped curve represents the steady-state solution of Eqs. (1) and (2). The calculations have been performed for a CdSe-Au heterodimer with the following set of parameters: the SQD and MNP radii $a = 3.3$ nm and $r = 10$ nm, respectively, the SQD-MNP center-to-center distance $d = 17$ nm, the SQD resonance energy $\hbar\omega_0 = 2.36$ eV, the SQD transition dipole moment $\mu = 0.65e \cdot$ nm, the SQD dielectric constant $\epsilon_s = 6.2$, the background permeability $\epsilon_b = 1$, the SQD relaxation constants $\gamma = 1.2$ ns^{-1} and $\Gamma = 3.33$ ns^{-1}, the detuning away from the SQD resonance $\Delta = 1.5\Gamma$. The polarizability of gold has been calculated using the tabulated data for ϵ_m [4]. For this set of parameters, the SQD-MNP coupling constant $G = (25.8 + 11.2i)\Gamma$ is inside the bistability region of the phase diagram (see the left panel in Fig. 1). We note here that there are two critical values of the input intensity which determine the threshold of switching of the system to the upper or lower branch of the S-shaped characteristic marked by dashed red and blues line in right panel of Fig. 1).

Figure 2 (left and middle panel) shows the evolution of the SQD population after a stepwise switching on of the input intensity around the right switching intensity $I_c = 532.47$, after which it approaches its stationary value. The calculations have been performed for a CdSe-Au heterodimer with the parameters described above. As is seen, the relaxation time of the SQD population τ depends strongly on the input intensity. The closer I is to the critical intensity I_c, the longer τ is. We defined τ as a time which the SQD population takes to reach its first peak. After that, we calculated τ as a function of the input intensity I. The results are plotted in Fig. 2, right panel. The solid line represents a fit given by $\tau = 5.48 \cdot 10^8 (I - I_c)^{-0.53}$ ns.
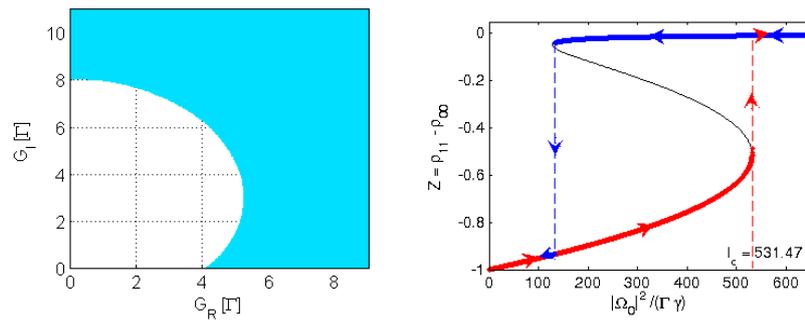
Fig. 1: Left panel – Phase diagram of the SQD bistability in the parameter space $[G_R, G_I]$. The coloured area shows the range of G_R and G_I where bistability exists. Right panel – The hysteresis loop of the SQD optical response (SQD population difference Z versus the input intensity $I = |\Omega_0|^2/(\gamma\Gamma)$) calculated for a CdSe-Au heterodimer (parameters are mentioned in the text). The S-shaped curve represents the steady-state solution of Eqs. (1). and (2)
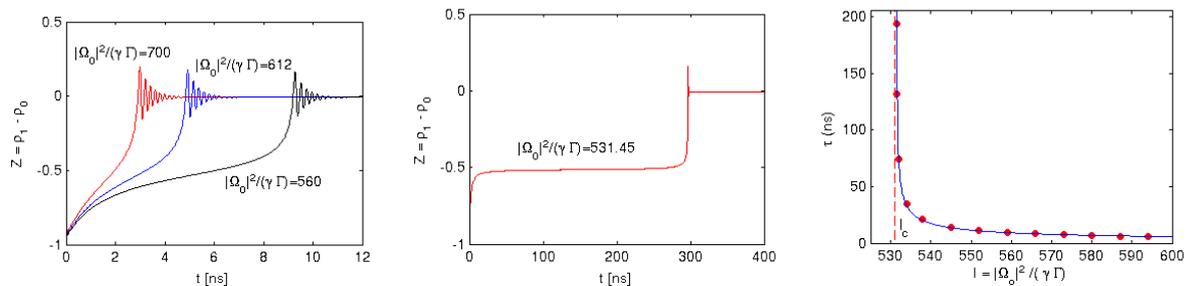
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Fig. 2: SQD population dynamics calculated after stepwise switching on of the input field (left and middle panels). Left panel – the input intensity $I = |\Omega_0|^2/(\gamma\Gamma)$ is well above the critical value I_c. Middle panel – I is close to the critical value I_c. Right panel – the relaxation time τ as a function of the input intensity I.

4. Conclusion

We studied the optical dynamics of a bistable hybrid nanodimer comprised of a closely spaced an SQD and an MNP. The system manifest bistability and hysteresis for a certain range of the SQD-MNP coupling strength. The bistability conditions can be realized for a CdSe-Au heterodimer. We found that close to a critical (switching) intensity of the input field the dynamics of the system slow down dramatically.

References

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