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Efficient Biexciton State Preparation in a Semiconductor Quantum Dot Coupled to a Metal Nanoparticle with Linearly Chirped Gaussian Pulses †

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Abstract: We consider a hybrid nanostructure composed of a semiconductor quantum dot placed near a spherical metallic nanoparticle, and study the effect of the nanoparticle on the population transfer from the ground state to the biexciton state of the quantum dot, when using linearly chirped Gaussian pulses. For various values of the system parameters (biexciton energy shift, pulse area and chirp, interparticle distance), we calculate the final population of the biexciton state by performing numerical simulations of the nonlinear density matrix equations describing the coupled system as well as its interaction with the applied electromagnetic field. We find that for relatively large values of the biexciton energy shift and not very small interparticle distances, the presence of the nanoparticle improves the biexciton state preparation, since it effectively increases the area of the applied pulse. We expect that our results may be exploited for the development of novel nanoscale photonic devices or future quantum technologies.

Keywords: semiconductor quantum dots; biexciton; coherent control; adiabatic rapid passage

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1. Introduction

Manipulating the exciton and biexciton states in semiconductor quantum dots using laser fields is an intense research area, because such systems provide a promising solid state platform for modern quantum technologies [1]. Within this framework, several works study the optical properties of hybrid systems composed of semiconductor quantum dots (SQDs) coupled to plasmonic nanostructures [2]. By coherently controlling the quantum part of these composite nanosystems, they behave as active nanophotonic structures which are expected to find major applications in fields like nanotechnology and quantum technology. As an example we mention that a composite structure consisting of a semiconductor quantum dot (SQD) and a metal nanoparticle (MNP), is more efficient than a quantum dot alone for optical phenomena like the creation of single photons on demand [3,4] and polarization-entangled photons [5]. In order to take advantage of the superior properties provided by the coupled SQD-MNP system regarding these quantum technology applications, an important problem is to efficiently prepare the biexciton state starting initially from the grounds state of the quantum dot, when the nanoparticle is present.

In our recent works we have tackled this problem using resonant hyperbolic secant [6] and on-off pulses [7], as well as pulses designed using the methodology of shortcuts to adiabaticity [8]. Although these methods appear to be successful in theory, they may present some problems in the experimental implementation. Specifically, the resonant pulses might not have the necessary robustness against unexpected frequency detunings, while the shortcut pulse profiles might be difficult to implement experimentally. For these reasons, here we investigate the problem of biexciton state preparation in the SQD-MNP

hybrid structure using easily implementable linearly chirped Gaussian pulses. Note that such pulses have been successfully used for population transfer to the biexciton state in a single quantum dot [9,10], without the metal nanoparticle, while here we study the problem in the presence of the MNP. We find with numerical simulations that the desired population transfer can be implemented with the used pulses. We also find that for large biexciton energies the nanoparticle improves the robustness of the population transfer.

2. System

The coupled SQD-MNP system is displayed in Figure 1. The Hamiltonian of this structure, in the dipole approximation, is written as [6–8]

$$H = E|1\rangle\langle 1| + (2E + E_b)|2\rangle\langle 2| - \mu\mathcal{E}_{\mathcal{SQD}}(t)\left(|0\rangle\langle 1| + |1\rangle\langle 2| + H.c.\right). \quad (1)$$

Here, E is the energy of the single-exciton state $|1\rangle$ and E_b the energy shift of the biexciton state $|2\rangle$. For simplicity, we have taken the energy of the ground state $|0\rangle$ as the zero of the energy. Also, μ denotes the dipole moment of the semiconductor quantum dot corresponding to the ground-exciton transition and the exciton-biexciton transition (for simplicity this is taken the same for both transitions), and $\mathcal{E}_{\mathcal{SQD}}$ represents the electric field inside the semiconductor quantum dot. We note that we have considered a symmetric quantum dot and due to selection rules there is no direct ground to biexciton transition with a single photon. We describe the dynamics of the quantum dot - metal nanoparticle hybrid system using the density matrix equations, see the detailed equations for this specific structure in Refs. [6–8].

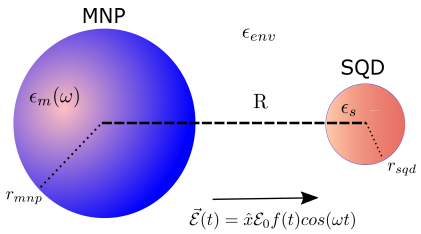

Figure 1. Semincoductor quantum dot-metal nanoparticle coupled systems.

3. Results and Discussion

We numerically simulate the density matrix equations with the parameter values that have been used in many studies of the systems at hand, see for example Refs. [6–8], and typically correspond to CdSe-based quantum dots. For the metallic nanosphere we use a gold nanosphere of radius 7.5 nm. We take the SQD initially in the ground state, and study the efficiency of population transfer in the biexciton state in the presence of the MNP, when using chirped Gaussian pulses with initial duration $\tau_0 = 0.75$ ps, for various values of pulse area and chirp parameter.

In Figure 2 we display contour plots of the final biexciton population as function of the pulse area and the chirp parameter, for biexciton energy shift $E_b = 7$ meV and four values of the interparticle distance. When $R = 100$ nm, Figure 2a, a distance for which MNP has practically no effect on the population transfer, we observe that the biexciton state can in general be robustly prepared for larger values of the chirp parameter a, as long as the pulse area exceeds some threshold. For smaller distances, like $R = 15$ nm and $R = 12$ nm, we observe from Figure 2b,c that the pulse area threshold is lowered and thus the robustness of the transfer is increased, due to the presence of the MNP. For even shorter distances, as in Figure 2d where $R = 11$ nm, we observe that the performance is degraded compared to the previous two cases, although still can be found large parameter areas for which the population transfer is robust.

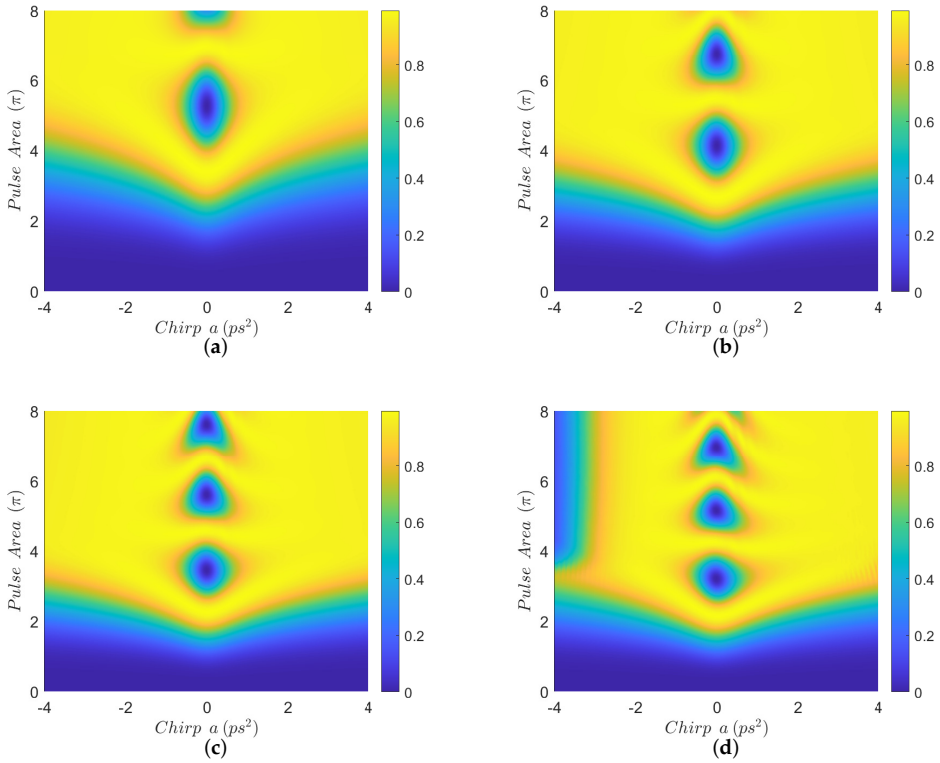

Figure 2. Contour plots of the final biexciton population as function of the pulse area and the chirp parameter a of the applied Gaussian pulse, for biexciton energy shift $E_b = 7$ meV and four different interparticle distances: (**a**) $R = 100$ nm, (**b**) $R = 15$ nm, (**c**) $R = 12$ nm, and (**d**) $R = 11$ nm.

In order to understand the behavior observed in Figure 2, where a nonzero E_b is used, we need to adapt the point of view of Ref. [9] to the case where a MNP is placed next to the SQD. In that work the authors study the population transfer to the biexciton state in a SQD withouth MNP, when using linearly chirped Gaussian pulses. They explain their results by considering the effect of E_b on the eigenenergies of the three-level biexciton system. Here we will adopt the same point of view and additionally consider the influence of the MNP. Specifically, the effect of the MNP on our system is two-fold. First, it effectively increases the pulse area of the effective Rabi frequency [6–8]. Second, the terms in the density matrix equations involving the self-interaction term, see Refs. [6–8], act as a perturbation, inducing transitions between the energy levels. As explained in Ref. [9], for large values of E_b, like in Figure 2, and for pulse areas above threshold, the spacing between the energy eigenvalues is large enough to allow the adiabatic population transfer from the ground to the biexciton state, as shown in Figure 2a. The large value of the biexciton energy shift E_b, which determines the detuning between the central pulse frequency ω and the energy of the exciton level, guarantees that the perturbation terms involving G do not induce transitions between exciton and biexciton states. Thus, the only effect of the nanoparticle is to increase the pulse area and thus robustness, as is demonstrated in Figure 2b,c. We emphasize that this phenomenon has not been observed in Ref. [9], since no MNP is considered there. Only for shorter distances, where parameter G increases, the robustness is undermined by the presence of the MNP, as in Figure 2d.

4. Conclusions

We showed with numerical simulations that the biexciton state can be efficiently prepared in a coupled semiconductor quantum dot-metal nanoparticle system, using easily implemented linearly chirped Gaussian pulses. This population transfer problem in this hybrid system is quite important, since such systems present enhanced properties for

quantum technology applications, like single-photon generation. We also found that for large values of the biexciton energy shift the presence of the nanoparticle enhances the robustness of the population transfer.

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References

1. Michler, P. (Ed.) *Quantum Dots for Quantum Information Technologies*; Springer: Berlin/Heidelberg, Germany, 2017; ISBN 978-331-956-377-0.
2. Szychowski, B.; Pelton, M.; Daniel, M.C. Preparation and properties of plasmonic-excitonic nanoparticle assemblies. *Nanophotonics* **2019**, *8*, 517.
3. Ridolfo, A.; Di Stefano, O.; Fina, N.; Saija, R.; Savasta, S. Quantum plasmonics with quantum dot-metal nanoparticle molecules: Influence of the Fano effect on photon statistics. *Phys. Rev. Lett.* **2010**, *105*, 263601.
4. Straubel, J.; Filter, R.; Rockstuhl, C.; Slowik, K. Plasmonic nanoantenna based triggered single-photon source. *Phys. Rev. B* **2016**, *93*, 195412.
5. Maksymov, S.I.; Miroshnichenko, A.E.; Kivshar, Y.S. Plasmonic nanoantennas for efficient control of polarization-entangled photon pairs. *Phys. Rev. A* **2012**, *86*, 011801(R).
6. Paspalakis, E.; Smponias, A.; Stefanatos, D. Coherent preparation of the biexciton state in a semiconductor quantum dot coupled to a metallic nanoparticle. *J. Appl. Phys.* **2021**, *129*, 223104.
7. Smponias, A.; Stefanatos, D.; Paspalakis, E. Efficient biexciton preparation in a quantum dot—Metal nanoparticle system using on-off pulses. *Nanomaterials* **2021**, *11*, 1859.
8. Smponias, A.; Stefanatos, D.; Paspalakis, E. Efficient biexciton state preparation in a semiconductor quantum dot-metallic nanoparticle hybrid structure using transitionless quantum driving. *Ann. Phys.* **2022**, *534*, 2100316.
9. Glässl, M.; Barth, A.; Gawarecki, K.; Machnikowski, P.; Croitoru, M.D.; Lüker, S.; Reiter, D.E.; Kuhn, T.; Axt, V.M.; Biexciton state preparation in a quantum dot via adiabatic rapid passage: Comparison between two control protocols and impact of phonon-induced dephasing. *Phys. Rev. B* **2013**, *87*, 085303.
10. Kaldewey, T.; Lüker, S.; Kuhlmann, A. .; Valentin, S.R.; Ludwig, A.; Wieck, A D.; Reiter, D.E.; Kuhn, T.; Warburton, R.J. Coherent and robust high-fidelity generation of a biexciton in a quantum dot by rapid adiabatic passage. *Phys. Rev. B* **2017**, *95*, 161302(R).