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Efficient Excitonic Population Transfer in a Coupled Quantum Dot–Metal Nanoparticle Structure Interacting with a Chirped Laser Pulse †

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Abstract: We apply the method of rapid adiabatic passage to a semiconductor quantum dot coupled to a plasmonic nanostructure, specifically a metal nanoparticle, and examine the excitonic state preparation efficiency for different distances between the quantum dot and the metal nanoparticle. In particular, results for the interaction of the coupled quantum dot–metal nanoparticle structure with linearly chirped Gaussian laser pulses are presented. We find that efficient population transfer occurs for a wide range of system parameters, like pulse areas and chirp rates, for different distances between the quantum dot and the metal nanoparticle. The presence of the metal nanoparticle influences significantly the population transfer to the exciton state, when the distance between the quantum dot and the metal nanoparticle becomes small.

Keywords: semiconductor quantum dot; metal nanoparticle; plasmonics; rapid adiabatic passage; chirped laser pulse

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1. Introduction

A relatively new area of active research combining nanophotonics, quantum optics and quantum technology studies the optical properties of complex structures containing plasmonic nanostructures and quantum systems, such as molecules and semiconductor quantum dots [1]. Coherently controlled quantum systems coupled to plasmonic nanostructures are considered active nanophotonic structures and are expected to have important applications in many fields, such as nanotechnology and quantum computing.

An important problem studied in these systems is the effect of the plasmonic nanostructure on controlled population transfer to the exciton state of the semiconductor quantum dot, starting from the ground state of the quantum dot [2–8]. The studies to date on the controlled population dynamics of quantum dots coupled to plasmonic nanostructures have dealt mainly with the preparation of the exciton state by resonant methods [2–7], while more recently there has been work on optimal pulses as well [8]. The resonant methods give excitonic population with very high efficiency, but only for a specific combination of pulse width and electric field value. Also, the performance of the resonance methods is quite sensitive to variations in the parameters of the laser fields used. Alternatively, there are very important methods of population transfer and quantum control that are adiabatic and are not sensitive to moderate changes in the parameters of the laser fields. One of these methods is rapid adiabatic passage [9]. This method has been used extensively in isolated quantum dots, both theoretically [10] and experimentally [11,12].

In the present work, we apply the method of rapid adiabatic passage to a semiconductor quantum dot coupled to a plasmonic nanostructure, specifically a spherical metal nanoparticle, and examine the excitonic state preparation efficiency for different distances

between the quantum dot and the metal nanoparticle. In particular, results for the interaction of the coupled quantum dot–metal nanoparticle structure with linearly chirped Gaussian laser pulses are presented. We find that efficient population transfer occurs for a wide range of system parameters, like pulse areas and chirp rates, for different distances between the quantum dot and the metal nanoparticle. We also show that the presence of the metal nanoparticle influences significantly the population transfer to the exciton state, when the distance between the quantum dot and the metal nanoparticle becomes small.

2. Methods and Materials

The coupled nanostructure that we consider contains a spherical metal nanoparticle of radius a and a small semiconductor quantum dot placed along the z-axis. The structure is embedded in an environment with real dielectric constant ε_{env}. The distance between the two particles is denoted by R (center-to-center distance). The quantum dot is described as a two-level quantum system, with $|1\rangle$ being the ground state and $|2\rangle$ being the single exciton state. The coupled system interacts with a linearly polarized electromagnetic field with $\vec{E}(t) = \hat{z}E_0 f(t)\cos[\omega t + \phi(t)]$. Here, $\hat{z}$ is the polarization unit vector, which is taken along the z direction, E_0 is the electric field amplitude, $f(t)$ is the dimensionless pulse envelope, ω is the angular frequency, and $\phi(t)$ is the time-dependent phase of the applied field, which leads to the creation of a chirped pulse. Also, ε_S is the dielectric constant of the semiconductor quantum dot and we treat the metal nanoparticle as a classical dielectric particle with dielectric function $\varepsilon_m(\omega)$. The dynamics of the population of the quantum dot is described by the density matrix equations, see, e.g., Ref. [5]. In the equations, we include both the modification of the electric field seen by the quantum dot, as well as, the exciton-plasmon coupling between the quantum dot and the metal nanoparticle.

We consider a Gaussian shape, linearly chirped laser pulse, that interacts with the coupled nanostructure, similar to that described in Refs. [9,10], meaning that the initial pulse has the Gaussian form $f(t) = e^{-(t-t_0)^2/2\tau_0^2}$, which is chirped and obtains a time-dependent frequency with linear dependence in $\dot{\phi}(t)$. We solve the density matrix equations for specific parameters of the quantum dot and different pulse areas, chirp rates and distances between the quantum dot and the metal nanoparticle and obtain the population of the exciton state. The parameters used in the calculations are $\varepsilon_{env} = \varepsilon_0$, , with ε_0 being the dielectric constant of the vacuum, $a = 7.5$ nm, the electric dipole moment of the quantum dot is $\mu = 0.65\, e$ nm, the energy of the exciton state is $\hbar\omega_0 = 2.5$ eV while the ground state energy is zero, and the dielectric constant of the quantum dot is $\varepsilon_S = 6\varepsilon_0$. For the metal nanoparticle, we use the dielectric function from experimental data. Also, the decay and dephasing times of the quantum dot are taken $T_1 = 0.8$ ns and $T_2 = 0.3$ ns, respectively.

3. Results and Discussion

The basic results of this work are shown in Figure 1. We present the population of the exciton state at the end of the pulse for the case that $\tau_0 = 1$ ps for different values of the pulse area and the spectral chirp. In Figure 1a, where $R = 30$ nm, the influence of the metal nanoparticle to the population transfer is essentially insignificant and the results obtained are similar for a two-level quantum system without the metal nanoparticle [9,10]. This means that we obtain very high population in the exciton state in the absence of the chirp for pulse areas that are odd multiples of π, while for pulse areas that are even multiples of π there is no population transfer. However, when the pulse is chirped, we obtain population transfer to the exciton state with high efficiency for a wide range of parameters. Also, the efficiency of the transfer is symmetric for positive and negative values of the chirp rate.

When the quantum dot comes closer to the metal nanoparticle, efficient population transfer to the exciton state still occurs for a wide range of pulse areas and chirp rates. However, in this case the influence of the metal nanoparticle to the efficiency of population transfer to the exciton state is significant, as it is shown in Figure 1b–d. We first note that the regions of parameters where the efficiency of population transfer is very high become more narrow as the quantum dot and the metal nanoparticle approach each other; see,

for example, the results shown in Figure 1c,d, where the regions of efficient population transfer is significantly narrower than in the results presented in Figure 1a,b. Also, in the absence of the chirp, we do not obtain the maximum of population of the exciton state for pulse areas that are exactly odd multiples of π. The latter phenomenon has been discussed earlier for resonant excitation of the system with hyperbolic secant pulses [5]. In addition, the efficiency of transfer is no longer symmetric for positive and negative values of the chirp rate. Specifically, it may happen for short interparticle distances, see Figure 1d, that the same value of the pulse area leads to high population transfer efficiency for a specific negative chirp rate, while for the same positive chirp rate it gives very low population transfer efficiency, and vice versa.

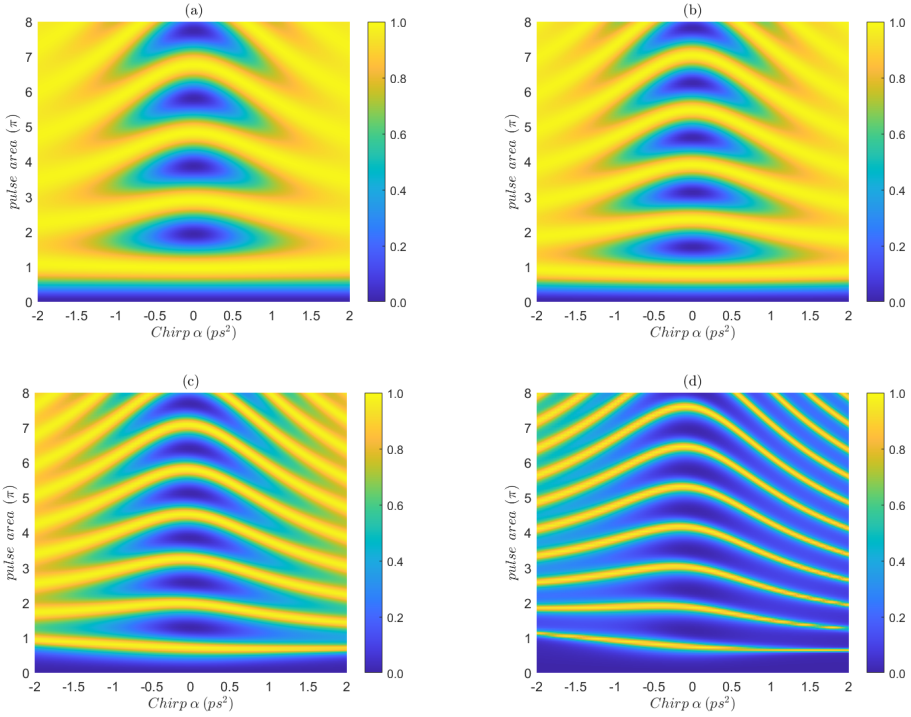

Figure 1. The population of the exciton state at the end of the pulse for different values of the pulse area and the spectral chirp rate α. The results are shown for different distances between the quantum dot and the metal nanoparticle. In (**a**) $R = 30$ nm, in (**b**) $R = 15$ nm, in (**c**) $R = 12$ nm, and in (**d**) $R = 11$ nm.

4. Conclusions

In this work, we presented a study on the application of the method of rapid adiabatic passage for preparing the excitonic state of a quantum dot coupled to a plasmonic nanostructure, specifically a metal nanoparticle. We applied the widely used methodology of the nonlinear density matrix equations, which contain the exciton-plasmon coupling [2–8], and examined the excitonic state preparation efficiency for different distances between the quantum dot and the metal nanoparticle. In particular, initial results for the interaction of the coupled quantum dot–metal nanoparticle structure with linearly chirped Gaussian laser pulses were presented. We found that, although efficient population transfer occurs for a wide range of system parameters, like pulse areas and chirp rates, the presence of the metal nanoparticle may influence significantly the population transfer to the exciton state, when the distance between the quantum dot and the metal nanoparticle is small.

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Conflicts of Interest: The authors declare no conflict of interest.

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