



Transient properties of transparency of a quantum system near a plasmonic nanostructure

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ABSTRACT

We study theoretically the time evolution of the susceptibility of a four-level double-V-type quantum system near a plasmonic nanostructure. In the quantum system under study one V-type transition is influenced by the interaction with surface plasmons while the other V-type transition interacts with free-space vacuum. As a plasmonic nanostructure we consider a two-dimensional array of metal-coated dielectric nanospheres. We analyze the case of a linearly polarized weak laser field that couples the lowest state with the upper states in the free-space transitions. We show that the time evolution of the susceptibility of the system is strongly dependent on the distance between the quantum system and the plasmonic nanostructure. For example, while for some distances only transient absorption appears, for others transient gain without inversion is possible.

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1. Introduction

A relatively new area of research in photonics that combines nanophotonics and quantum optics is the study of the interaction of light with quantum emitters, such as atoms, molecules and quantum dots, in the presence of plasmonic nanostructures. Examples of the phenomena studied in this area include the modification of spontaneous emission [1–5] and of resonance fluorescence [6,7], the creation of quantum interference effects in spontaneous emission [8–12], the creation and enhancement of entanglement [13–16] between distant quantum systems, and the creation of quantum coherence and interference effects such as optical transparency [17,18], slow light [18,19], and gain without inversion [20,21].

In this work we study the time evolution of the susceptibility of a four-level double-V-type quantum system near a plasmonic nanostructure. In the quantum system under study one V-type transition is influenced by the interaction with surface plasmons while the other V-type transition interacts with free-space vacuum. As a plasmonic nanostructure we consider a two-dimensional array of metal-coated dielectric nanospheres wherein we calculate the relevant decay rates by a rigorous electromagnetic Green's tensor technique [8–10,18]. The structure also interacts with a linearly polarized weak laser field, which couples the lowest state with the upper states in the

free-space transitions. The optical response of the system is studied by a density matrix methodology. Using numerical solutions of the relevant density matrix equations we show that the time evolution of the susceptibility of the system for a specific frequency is strongly influenced by the presence of the plasmonic nanostructure and is strongly dependent on the distance of the quantum system from the plasmonic nanostructure as well as on the free-space spontaneous decay.

Recently, Hatf and Singh studied the effects of spontaneous emission interference in absorption and dispersion properties of metallic photonic crystals doped with quantum dots in the V-type configuration interacting with either a single probe laser field or a probe and a coupling laser field [17]. In the same work they studied the time evolution of the susceptibility in the case of a single laser field. Very recently, we have analyzed the steady state optical response of the same coupled quantum-plasmonic system that is studied here and found, in particular, that optical transparency occurs at a specific laser frequency, which is also accompanied with very steep positive dispersion leading to slow light effect [18]. We note that the transient optical properties of other types of optical transparency, such as electromagnetically induced transparency [22–25], transparency near a photonic band edge [26], transparency via vacuum induced coherence [27,28], tunneling-induced transparency in semiconductor quantum wells [29,30], and voltage-controlled transparency in semiconductor quantum dots [31], have been studied in detail in the literature. Besides the creation of transient gain without inversion, a topic of

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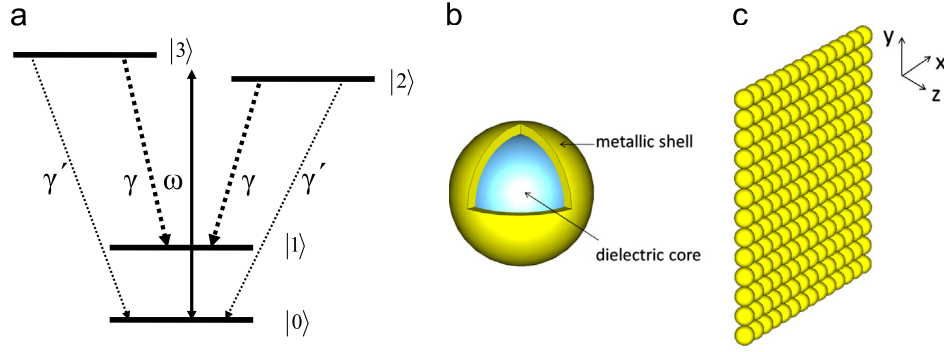


Fig. 1. (a) The quantum system under study is a double-V-type system where two upper states $|2\rangle$ and $|3\rangle$ decay with spontaneous emission to the two lower states $|0\rangle$ and $|1\rangle$. The system interacts with a probe laser field that couples state $|0\rangle$ with states $|2\rangle$ and $|3\rangle$. (b) A metal-coated dielectric nanosphere. (c) The two-dimensional array of metal-coated dielectric nanospheres used in this work.

fundamental scientific interest, the results of our study can also be used in enhancing the power generated by a photovoltaic device [32,33] and the efficiency of a photodetector [34].

The article is organized as follows. In the next section we present the relevant density matrix equations. Then, in Section 3 we present results for the time evolution of the susceptibility of the system from the numerical solution of the density matrix equations, for different distances of the quantum system from the plasmonic nanostructure, with or without free-space spontaneous decay rate. Finally, in Section 4 we summarize our findings.

2. Theoretical model

The quantum system of interest is shown in Fig. 1(a). We consider a four-level system with two closely lying upper states $|2\rangle$ and $|3\rangle$, and two lower states $|0\rangle$ and $|1\rangle$. We will call this system a double-V-type system, in order to identify easily two different three-level V-type transitions in the structure. We note that the system is also topologically equivalent to the so-called double Λ -type system [11,12]. The quantum system is located in vacuum at distance d from the surface of the plasmonic nanostructure. We take states $|2\rangle$ and $|3\rangle$ to characterize two Zeeman sublevels. Then, the dipole moment operator is taken as $\vec{\mu} = \mu'(|2\rangle\langle 0|\hat{e}_- + |3\rangle\langle 0|\hat{e}_+) + \mu(|2\rangle\langle 1|\hat{e}_- + |3\rangle\langle 1|\hat{e}_+) + \text{H.c.}$, where $\hat{e}_\pm = (\mathbf{e}_z \pm i\mathbf{e}_x)/\sqrt{2}$ describe the right- (\hat{e}_+) and left-rotating (\hat{e}_-) unit vectors; μ, μ' are taken to be real.

The quantum system interacts with a linearly polarized continuous wave laser field with electric field $\vec{E}(t) = \hat{z}E_0 \cos(\omega t)$, where E_0 is the electric field amplitude and ω is the angular frequency of the electric field. The laser field couples state $|0\rangle$ with states $|2\rangle$ and $|3\rangle$. The Hamiltonian that describes the interaction of the electromagnetic field with the quantum system, in the dipole and rotating wave approximations, is given by

$$H = \hbar \left(-\delta - \frac{\omega_{32}}{2} \right) |2\rangle\langle 2| + \hbar \left(-\delta + \frac{\omega_{32}}{2} \right) |3\rangle\langle 3| - \frac{\hbar\Omega}{2} (|0\rangle\langle 2| + |0\rangle\langle 3| + \text{H.c.}). \quad (1)$$

Here, $\delta = \omega - \bar{\omega}$ is the detuning from resonance with the average transition energies of states $|2\rangle$ and $|3\rangle$ from state $|0\rangle$, with $\bar{\omega} = (\omega_3 + \omega_2)/2 - \omega_0$, $\omega_{32} = (\omega_3 - \omega_2)/2$, and Ω is the Rabi frequency defined as $\Omega = \mu'E_0/(\sqrt{2}\hbar)$. Also, $\hbar\omega_n$ with $n=0-3$ is the energy of state $|n\rangle$.

Both excited states $|2\rangle$ and $|3\rangle$ decay spontaneously to state $|0\rangle$ with decay rates $2\gamma'_2, 2\gamma'_3$, respectively, and to state $|1\rangle$ with decay rates $2\gamma_2, 2\gamma_3$, respectively. We assume that the transitions $|2\rangle, |3\rangle$ to $|1\rangle$ lie within the surface-plasmon bands of the plasmonic nanostructure, whereas the transitions $|2\rangle, |3\rangle$ to $|0\rangle$ are spectrally far from the surface-plasmon bands and are not influenced by the plasmonic nanostructure. Therefore, as in the transitions $|2\rangle, |3\rangle$ to

$|0\rangle$ the spontaneous decay occurs due to the interaction of the quantum system with free-space vacuum modes, we refer to this decay as free-space spontaneous decay.

We choose the energy difference of states $|2\rangle$ and $|3\rangle$ to be rather small, i.e. ω_{32} to be just a few Γ_0 , where Γ_0 is the decay rate of states $|2\rangle$ and $|3\rangle$ to state $|1\rangle$ the vacuum. The latter is taken to be the same for both states. We can therefore assume that $\gamma_2 = \gamma_3 = \gamma$ and $\gamma'_2 = \gamma'_3 = \gamma'$ [9].

Using the Hamiltonian of Eq. (1) we obtain the following equations for the density matrix elements of the system, assuming a Markovian response:

$$\begin{aligned} \dot{\rho}_{00}(t) &= 2\gamma'[\rho_{22}(t) + \rho_{33}(t)] - i\frac{\Omega}{2}[\rho_{02}(t) - \rho_{20}(t)] \\ &\quad - i\frac{\Omega}{2}[\rho_{03}(t) - \rho_{30}(t)], \end{aligned} \quad (2)$$

$$\begin{aligned} \dot{\rho}_{22}(t) &= -2(\gamma + \gamma')\rho_{22}(t) + i\frac{\Omega}{2}[\rho_{02}(t) - \rho_{20}(t)] \\ &\quad - \kappa[\rho_{23}(t) + \rho_{32}(t)], \end{aligned} \quad (3)$$

$$\begin{aligned} \dot{\rho}_{33}(t) &= -2(\gamma + \gamma')\rho_{33}(t) + i\frac{\Omega}{2}[\rho_{03}(t) - \rho_{30}(t)] \\ &\quad - \kappa[\rho_{23}(t) + \rho_{32}(t)], \end{aligned} \quad (4)$$

$$\begin{aligned} \dot{\rho}_{20}(t) &= \left(i\delta + i\frac{\omega_{32}}{2} - \gamma - \gamma' \right) \rho_{20}(t) - i\frac{\Omega}{2}\rho_{22}(t) \\ &\quad - i\frac{\Omega}{2}\rho_{23}(t) + i\frac{\Omega}{2}\rho_{00}(t) - \kappa\rho_{30}(t), \end{aligned} \quad (5)$$

$$\begin{aligned} \dot{\rho}_{30}(t) &= \left(i\delta - i\frac{\omega_{32}}{2} - \gamma - \gamma' \right) \rho_{30}(t) - i\frac{\Omega}{2}\rho_{33}(t) \\ &\quad - i\frac{\Omega}{2}\rho_{32}(t) + i\frac{\Omega}{2}\rho_{00}(t) - \kappa\rho_{20}(t), \end{aligned} \quad (6)$$

$$\begin{aligned} \dot{\rho}_{23}(t) &= (i\omega_{32} - 2\gamma - 2\gamma')\rho_{23}(t) + i\frac{\Omega}{2}\rho_{03}(t) - i\frac{\Omega}{2}\rho_{20}(t) \\ &\quad - \kappa[\rho_{22}(t) + \rho_{33}(t)], \end{aligned} \quad (7)$$

with $\rho_{00}(t) + \rho_{11}(t) + \rho_{22}(t) + \rho_{33}(t) = 1$ and $\rho_{nm}(t) = \rho_{mn}^*(t)$. Here, κ is the coupling coefficient between states $|2\rangle$ and $|3\rangle$ due to spontaneous emission in a modified anisotropic vacuum [35] and is responsible for the appearance of quantum interference [36].

The values of γ and κ are obtained by [37–40]

$$\gamma = \frac{\mu_0\mu^2\bar{\omega}^2}{\hbar} \hat{e}_- \cdot \text{Im} \mathbf{G}(\mathbf{r}, \mathbf{r}; \bar{\omega}) \cdot \hat{e}_+ \quad (8)$$

$$\kappa = \frac{\mu_0\mu^2\bar{\omega}^2}{\hbar} \hat{e}_+ \cdot \text{Im} \mathbf{G}(\mathbf{r}, \mathbf{r}; \bar{\omega}) \cdot \hat{e}_+. \quad (9)$$

Here, $\mathbf{G}(\mathbf{r}, \mathbf{r}; \omega)$ is the dyadic electromagnetic Green's tensor, where \mathbf{r} refers to the position of the quantum emitter, and μ_0 is the permeability of vacuum. Also, $\bar{\omega} = (\omega_3 + \omega_2)/2 - \omega_1$. From Eqs.

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