

Optical Properties of a Symmetric Coupled Quantum Dot Nanostructure

Ioannis Chatzopoulos, John Boviatsis, and Emmanuel Paspalakis

Abstract—We theoretically study the optical response of a symmetric double quantum dot nanostructure. We assume that each quantum dot contains only two energy levels and that the effect of tunneling is only included in the coupling of the upper levels. We then derive the optical susceptibility of the system under weak field excitation and under general initial conditions. The formula we obtain extends that of a previous work, as the effects of an initial superposition of the two lower states of the system are accounted for. We also present the form of the susceptibility for different initial states of the system.

Keywords—Semiconductor quantum dot, optical susceptibility, tunneling, superposition of states.

I. INTRODUCTION

Semiconductor quantum dots are nanocrystals made of semiconductor materials and exhibit three-dimensional quantum confinement [1]. They show strong quantum mechanical effects and can be described by a discrete energy spectrum. They also have novel linear and nonlinear optical properties as they have controllable size, energies and dipole matrix elements. When two quantum dots are put close together then a coupled quantum dot is formed and in this case quantum tunneling effects play an important role [2], [3]. Two examples of coupled quantum dots are the symmetric double quantum dot nanostructure [2] and the asymmetric double quantum dot molecule [3].

The interaction of the electromagnetic fields with the asymmetric quantum dot molecule has led to several interesting phenomena including tunneling induced transparency with accompanying slow light [4], [5], optical gain [6], [7], and controlled population dynamics [3], [8], [9]. The interaction of electromagnetic fields with the symmetric double quantum dot nanostructure has been widely studied with regard to controlled population dynamics and single electron transfer [2], [10], [11], [12], [13], [14]. The optical susceptibility of the symmetric double quantum dot nanostructure was studied by Ginzburg and Orenstein [15]. They showed that the system can exhibit reduced absorption and slow light [15].

In this work we revised the optical response of the symmetric double quantum dot nanostructure. We make the same

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approximations as Ginzburg and Orenstein [15] and others [2], [10], [11], [13] [(a) two energy levels per quantum dot and (b) the effect of tunneling is only included in the coupling of the upper levels] and derive again the optical susceptibility of the system under weak field excitation. The formula we obtain extends that of Ginzburg and Orenstein [15], as the effects of an initial superposition of the two lower states of the system are accounted for. We also present the form of the susceptibility for different initial states of the system. Extensions of the model are also discussed.

II. THEORETICAL MODEL AND RESULTS

The coupled quantum dot nanostructure we consider is composed of two identical quantum dots (as it is usual they are depicted with two quantum wells in Fig. 1). Each one, when isolated, possesses only two bound state energy levels. These localized states are denoted by $|L_1\rangle, |R_1\rangle$ (lower states) and by $|L_2\rangle, |R_2\rangle$ (upper states) for the left (L) and right (R) quantum dot. The lower bound state has energy ε_1 and the upper bound state has energy ε_2 . The geometrical characteristics of the dots are chosen such that the lower energy level is deep in the potential barrier and the upper energy level is near the edge of the potential barrier. These features of the energy levels are carried over to the double quantum dot nanostructure as the quantum dots are taken to be widely separated.

The Hamiltonian of the double quantum dot nanostructure, interacting with an external electromagnetic field with electric field $E(t)$ is given by

$$\begin{aligned} \hat{H} &= \varepsilon_1(|L_1\rangle\langle L_1| + |R_1\rangle\langle R_1|) \\ &+ \varepsilon_2(|L_2\rangle\langle L_2| + |R_2\rangle\langle R_2|) \\ &- \hbar U(|L_2\rangle\langle R_2| + |R_2\rangle\langle L_2|) \\ &- \mu E(t)(|L_1\rangle\langle L_2| + |R_1\rangle\langle R_2| + H.c.), \end{aligned} \quad (1)$$

where μ is the electron dipole moment for the transition $|a_1\rangle \leftrightarrow |a_2\rangle$ ($a = L, R$) (assumed real and it is taken the same for each quantum dot) and U is the electron hopping energy or the tunneling coupling coefficient between the two dots for the excited electronic states $|L_2\rangle$ and $|R_2\rangle$. The applied field is taken

$$E(t) = E_0 \cos(\omega t), \quad (2)$$

with ω being the angular frequency and E_0 being the amplitude of the electric field.

From the localized states $|L_2\rangle$ and $|R_2\rangle$ we can obtain the

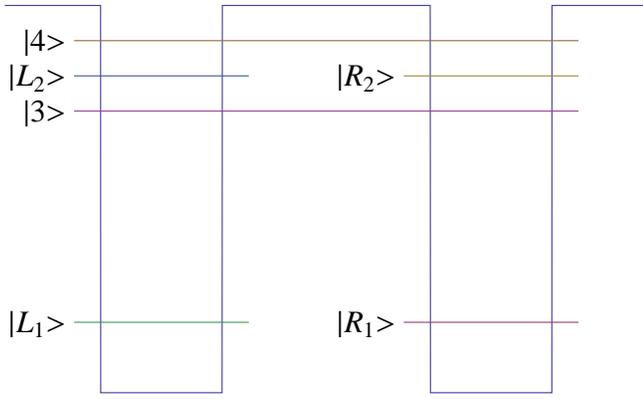


Fig. 1. Schematic diagram of the coupled quantum dot structure studied. We present the localized energy levels $|L_1\rangle, |L_2\rangle, |R_1\rangle, |R_2\rangle$ and two delocalized upper levels ($|3\rangle$ and $|4\rangle$).

delocalized (coupled) states $|3\rangle$ and $|4\rangle$, with

$$|3\rangle = \frac{1}{\sqrt{2}} (|L_2\rangle + |R_2\rangle), \quad (3)$$

$$|4\rangle = \frac{1}{\sqrt{2}} (|L_2\rangle - |R_2\rangle), \quad (4)$$

that have energies

$$\varepsilon_3 = \varepsilon_2 - \hbar U, \quad (5)$$

$$\varepsilon_4 = \varepsilon_2 + \hbar U. \quad (6)$$

For the derivation of the optical susceptibility of the system, we will use the probability amplitude approach. This approach gives proper results for weak field excitation, which is the case of interest here. The wavevector of the system at time t can be written as a superposition of two lower localized states $|L_1\rangle, |R_1\rangle$ and the two excited delocalized states $|3\rangle, |4\rangle$, such that

$$|\psi(t)\rangle = a_1(t)|L_1\rangle + a_2(t)|R_1\rangle + a_3(t)|3\rangle + a_4(t)|4\rangle. \quad (7)$$

From the time-dependent Schrödinger equation, using the Hamiltonian of equation (1) and the definitions of equations (3) - (6), we obtain the time evolution of the probability amplitudes $a_n(t)$ with $n = 1 - 4$:

$$i\hbar\dot{a}_1(t) = \varepsilon_1 a_1(t) - \frac{\mu E_0}{\sqrt{2}} \cos(\omega t) [a_3(t) + a_4(t)], \quad (8)$$

$$i\hbar\dot{a}_2(t) = \varepsilon_1 a_2(t) - \frac{\mu E_0}{\sqrt{2}} \cos(\omega t) [a_3(t) - a_4(t)], \quad (9)$$

$$i\hbar\dot{a}_3(t) = \varepsilon_3 a_3(t) - i\hbar\gamma a_3(t) - \frac{\mu E_0}{\sqrt{2}} \cos(\omega t) [a_1(t) + a_2(t)], \quad (10)$$

$$i\hbar\dot{a}_4(t) = \varepsilon_4 a_4(t) - i\hbar\gamma a_4(t) - \frac{\mu E_0}{\sqrt{2}} \cos(\omega t) [a_1(t) - a_2(t)], \quad (11)$$

where, γ denotes the decay rate of the upper states and has been included phenomenologically in the equations of the probability amplitudes.

We proceed with a change of variables

$$a_1(t) = c_1(t)e^{-i\frac{\varepsilon_1}{\hbar}t}, \quad (12)$$

$$a_2(t) = c_2(t)e^{-i\frac{\varepsilon_1}{\hbar}t}, \quad (13)$$

$$a_3(t) = c_3(t)e^{-i\frac{\varepsilon_3}{\hbar}t + i\delta t - iUt}, \quad (14)$$

$$a_4(t) = c_4(t)e^{-i\frac{\varepsilon_4}{\hbar}t + i\delta t + iUt}. \quad (15)$$

We use the new variables in equations (8) - (11), perform the rotating wave approximation and obtain

$$i\dot{c}_1(t) = -\frac{\mu E_0}{2\sqrt{2}\hbar} [c_3(t) + c_4(t)], \quad (16)$$

$$i\dot{c}_2(t) = -\frac{\mu E_0}{2\sqrt{2}\hbar} [c_3(t) - c_4(t)], \quad (17)$$

$$i\dot{c}_3(t) = (\delta - U - i\gamma)c_3(t) - \frac{\mu E_0}{2\sqrt{2}\hbar} [c_1(t) + c_2(t)], \quad (18)$$

$$i\dot{c}_4(t) = (\delta + U - i\gamma)c_4(t) - \frac{\mu E_0}{2\sqrt{2}\hbar} [c_1(t) - c_2(t)], \quad (19)$$

where, $\delta = (\varepsilon_2 - \varepsilon_1)/\hbar - \omega = \omega_{21} - \omega$.

The induced polarization of the quantum dot structure is given by

$$P = N\langle\hat{\mu}\rangle = \frac{N\mu}{\sqrt{2}} (c_3c_1^* + c_4c_1^* + c_3c_2^* - c_4c_2^*)e^{-i\omega t} + c.c. \quad (20)$$

where N is the electron density in the quantum dot structure. The induced polarization can be also written in terms of the optical susceptibility $\chi(\omega)$ as

$$P = \varepsilon_0\chi(\omega)\frac{E_0}{2}e^{-i\omega t} + \varepsilon_0\chi^*(\omega)\frac{E_0}{2}e^{i\omega t}, \quad (21)$$

where ε_0 is the vacuum permittivity. From equations (20) and (21) we obtain

$$\chi(\omega) = \frac{\sqrt{2}N\mu}{\varepsilon_0E_0} (c_3c_1^* + c_4c_1^* + c_3c_2^* - c_4c_2^*), \quad (22)$$

where the probability amplitudes c_n are calculated in steady state.

We assume that initially, at $t = 0$, the quantum dot nanostructure is at a superposition of the two lower states $|L_1\rangle$ and $|R_1\rangle$

$$|\psi(0)\rangle = \alpha|L_1\rangle + \beta|R_1\rangle, \quad (23)$$

with $|\alpha|^2 + |\beta|^2 = 1$. If the interaction of the electromagnetic field with the quantum dot nanostructure is weak, then we can apply time-dependent perturbation theory and assume that $c_1 \approx \alpha$, $c_2 \approx \beta$. We also calculate c_3 and c_4 from equations (18) and (19) in steady state. Substituting the results into equation (22), after some algebra, we obtain the optical susceptibility as

$$\chi(\omega) = \frac{N\mu^2}{\hbar\varepsilon_0} \frac{\omega_{21} - \omega - i\gamma + 2U\text{Re}(\alpha\beta^*)}{(\omega_{21} - \omega - i\gamma)^2 - U^2}. \quad (24)$$

This formula extends the result of Ginzburg and Orenstein [15] for the case of an initial superposition of the two lower states of the quantum dot nanostructure. We note that if we

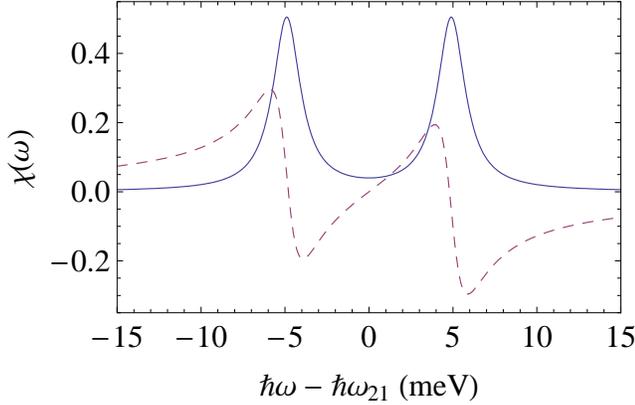


Fig. 2. The form of the real part (dashed curve) and the imaginary part (solid curve) of $\chi(\omega)$, taken from equation (24), in units $\frac{N\mu^2}{\hbar\varepsilon_0}$, for $\alpha = 1, \beta = 0$ or $\alpha = 0, \beta = 1$.

take $U = 0$ then the susceptibility gives the well-known result for a single quantum dot [16]

$$\chi(\omega) = \frac{N\mu^2}{\hbar\varepsilon_0} \frac{\omega_{21} - \omega + i\gamma}{(\omega_{21} - \omega)^2 + \gamma^2}, \quad (25)$$

which gives a Lorentzian absorption lineshape from its imaginary part and a regular dispersive lineshape from its real part.

III. FORM OF THE OPTICAL SUSCEPTIBILITY

We will now present the form of the optical susceptibility, using equation (24), for a specific quantum dot nanostructure and for different initial conditions. We take a GaAs based quantum dot, where each quantum dot has 5 nm size and are separated by a 7 nm barrier. The height of the well for each quantum dot is 450 meV. We solve the time-independent Schrödinger equation, in the effective mass approximation, for an electron in the quantum dot, using the shooting method [17], and calculate the energies, dipole matrix elements and the tunneling coupling coefficient. As we are interested in the form of the susceptibility the most important quantity in our study is the tunneling coupling coefficient that is found $\hbar U = 4.9$ meV.

Fig. 2 presents the form of the optical susceptibility in the case that $\alpha = 1, \beta = 0$ or $\alpha = 0, \beta = 1$, so the electron is initially localized in one of the quantum dots. This was the result presented by Ginzburg and Orenstein [15]. The decay rate, here and in the rest of the figures, is taken $\hbar\gamma = 1$ meV. The imaginary part of the susceptibility, which determines the absorption, shows a symmetric double-peaked structure, and the peaks are obtained at the energies $\varepsilon_3 - \varepsilon_1$ and $\varepsilon_4 - \varepsilon_1$. For $\omega = \omega_{21}$ there is a minimum in the imaginary part of $\chi(\omega)$. The real part, which can be used for the calculation of the refractive index and determines dispersion, obtains three zeroes. It shows regular dispersive behavior (negative slope) around $\varepsilon_3 - \varepsilon_1$ and $\varepsilon_4 - \varepsilon_1$. However, around ω_{21} the real part of $\chi(\omega)$ shows positive slope, leading to slow light effects [15].

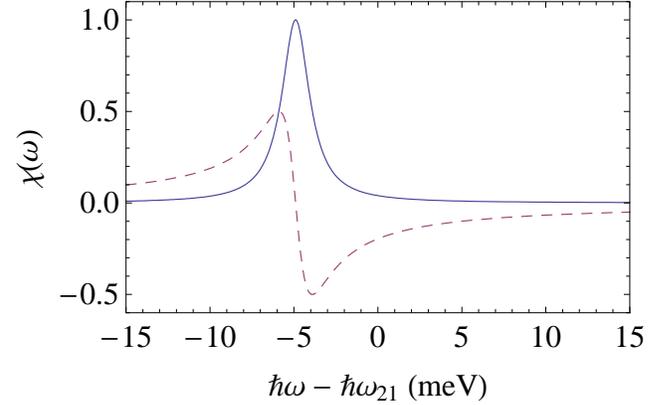


Fig. 3. The form of the real part (dashed curve) and the imaginary part (solid curve) of $\chi(\omega)$, taken from equation (24), in units $\frac{N\mu^2}{\hbar\varepsilon_0}$, for $\alpha = 1/\sqrt{2}, \beta = 1/\sqrt{2}$.

Fig. 3 presents the form of the optical susceptibility in the case that $\alpha = 1/\sqrt{2}, \beta = 1/\sqrt{2}$. This is the case of a symmetric initial superposition of states $|L_1\rangle$ and $|R_1\rangle$, which is actually the lowest energy delocalized state of the symmetric double quantum dot nanostructure. In this case we take a single Lorentzian curve for the imaginary part and a regular dispersive curve for the real part of $\chi(\omega)$. This case is similar to what happens in a single quantum dot. The maximum of the imaginary part of the susceptibility and the single zero in the real part of the susceptibility are found at energy $\varepsilon_3 - \varepsilon_1$, that shows that the lower states couple only to the delocalized state $|3\rangle$ and not to the delocalized state $|4\rangle$. This can be explained from equations (18) and (19), which show that a symmetric superposition of the lower states will only couple to state $|3\rangle$ and not to state $|4\rangle$.

Similar results are obtained in the case that $\alpha = 1/\sqrt{2}, \beta = -1/\sqrt{2}$ (see Fig. 4), which is the case of an asymmetric initial superposition of states $|L_1\rangle$ and $|R_1\rangle$. This is actually the first excited delocalized state of the symmetric double quantum dot nanostructure. The difference in comparison with Fig. 3 is that the maximum of the imaginary part of the susceptibility and the zero in the real part of the susceptibility are found at energy $\varepsilon_4 - \varepsilon_1$. This shows that the lower states couple only to the delocalized state $|4\rangle$ and not to the delocalized state $|3\rangle$, which can be also seen from equations (18) and (19).

In the next case, we consider that most of the population is initially in one of the two quantum dots, $\alpha = 1/\sqrt{5}, \beta = 2/\sqrt{5}$ [Fig. 5(a)] and $\alpha = 1/\sqrt{5}, \beta = -2/\sqrt{5}$ [Fig. 5(b)]. This means that 0.2 of the total population is initially in state $|L_1\rangle$ and 0.8 of the total population is initially in state $|R_1\rangle$. The absorption curve in this case is double-peaked but it is strongly asymmetric with one of the two peaks much higher than the other. The sign of the initial superposition determines if the maximum of the imaginary part of the susceptibility and the single zero in the real part of the susceptibility will be around $\varepsilon_3 - \varepsilon_1$ (first case) and $\varepsilon_4 - \varepsilon_1$ (second case).

In the final figure, Fig. 6, we consider a case of equal population between states $|L_1\rangle$ and $|R_1\rangle$, but with a ϕ phase

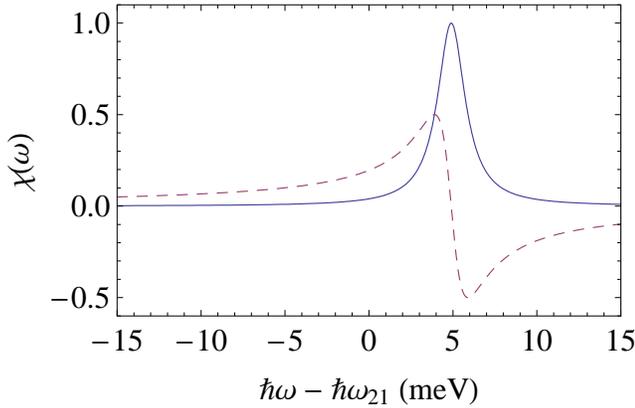


Fig. 4. The form of the real part (dashed curve) and the imaginary part (solid curve) of $\chi(\omega)$, taken from equation (24), in units $\frac{N\mu^2}{\hbar\epsilon_0}$, for $\alpha = 1/\sqrt{2}$, $\beta = -1/\sqrt{2}$.

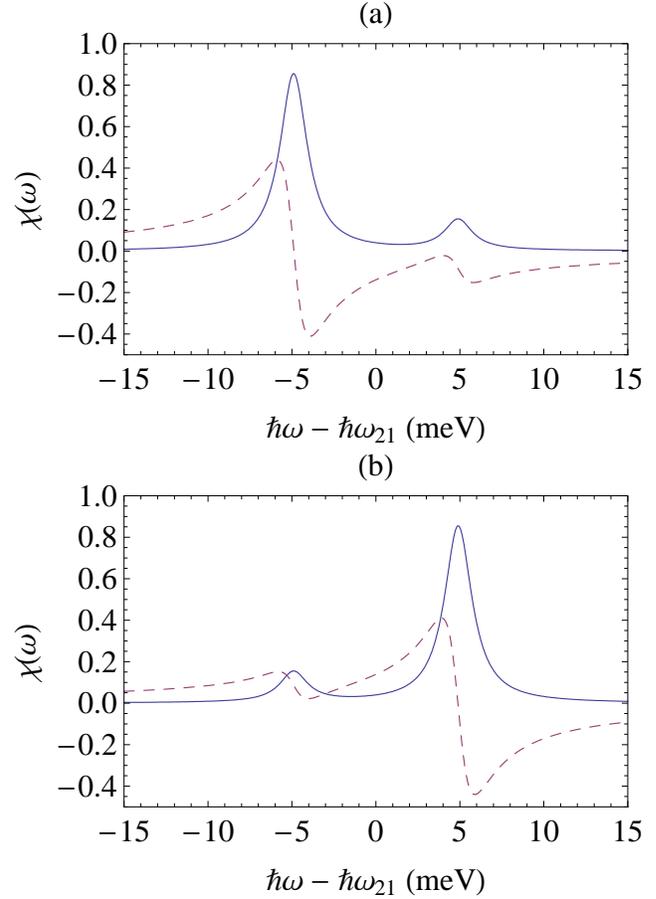


Fig. 6. The form of the real part (dashed curve) and the imaginary part (solid curve) of $\chi(\omega)$, taken from equation (24), in units $\frac{N\mu^2}{\hbar\epsilon_0}$. In (a) $\alpha = 1/\sqrt{2}$, $\beta = e^{i\pi/4}/\sqrt{2}$ and in (b) $\alpha = 1/\sqrt{2}$, $\beta = e^{i3\pi/4}/\sqrt{2}$.

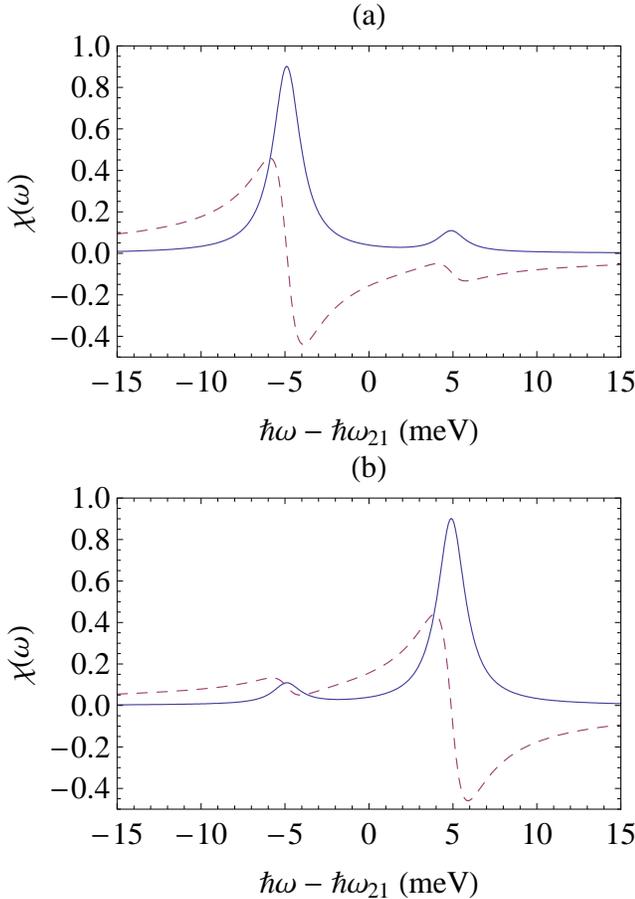


Fig. 5. The form of the real part (dashed curve) and the imaginary part (solid curve) of $\chi(\omega)$, taken from equation (24), in units $\frac{N\mu^2}{\hbar\epsilon_0}$. In (a) $\alpha = 1/\sqrt{5}$, $\beta = 2/\sqrt{5}$ and in (b) $\alpha = 1/\sqrt{5}$, $\beta = -2/\sqrt{5}$.

difference between the two states, $\alpha = 1/\sqrt{2}$, $\beta = e^{i\phi}/\sqrt{2}$. We take $\phi = \pi/4$ [Fig. 6(a)] and $\phi = 3\pi/4$ [Fig. 6(b)]. Interestingly, the behavior of the real and imaginary parts of the optical susceptibility is much closer to the strongly unequal population distribution case, shown in Fig. 5, than to the symmetric and asymmetric superposition cases, which also give equal population distributions in states $|L_1\rangle$ and $|R_1\rangle$ (Figs. 3 and 4).

IV. SUMMARY AND POSSIBLE EXTENSIONS

In summary, we theoretically analyzed the optical response of a symmetric double quantum dot nanostructure. We assumed that each quantum dot contains only two energy levels and that the effect of tunneling is only included in the coupling of the upper levels. We used a probability amplitude approach and derived the optical susceptibility of the system under weak field excitation and under a general superposition of the two lower states. The formula we obtain extends that of a previous work [15], as the effects of an initial superposition of the two lower states of the system are accounted for. We also presented the form of the susceptibility for different initial states of the system, for a specific double quantum dot nanostructure, and showed that the actual form of the real and imaginary parts of

the susceptibility depends strongly on the actual form of the initial superposition.

We intend to extend the present study in order to account for the coupling by tunneling of the lower states of the system, which is omitted here and in other studies [2], [10], [11], [13], [15]. This will allow us to study the case of two quantum dots that are close to each other and not only widely separated. In addition, with this extension we will be able to calculate the effect of the actual separation of the two quantum dots on the optical susceptibility, as well as the transition between widely separated quantum dots to closely coupled quantum dots. Finally, as slow light effects are possible in this system [15], it will be interesting to study the effects of an initial superposition state as well as the quantum dots separation on the group velocity of light.

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