Optical Properties of a Symmetric Coupled Quantum Dot Nanostructure

Ioannis Chatzopoulos[1,2], John Boviatsis[3] and Emmanuel Paspalakis[2]

[1]Department of Physics and Astronomy, University of Southampton, Southampton, UK
[2]Materials Science Department, University of Patras, Patras, Greece
[3]Technological and Educational Institute of Western Greece, Patras, Greece

European Union
European Social Fund
OPERATIONAL PROGRAMME
EDUCATION AND LIFELONG LEARNING
investing in knowledge society
MINISTRY OF EDUCATION & RELIGIOUS AFFAIRS
MANAGING AUTHORITY
NSRF 2007-2013
programme for development
EUROPEAN SOCIAL FUND
Co-financed by Greece and the European Union

Semiconductor Quantum Dots

- Semiconductor quantum dots [1] are nanocrystals made of semiconductor materials and exhibit three-dimensional quantum confinement.
- They show strong quantum mechanical effects and can 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 important role.
- An example of a coupled quantum dot is the symmetric double quantum dot nanostructure [2].

1. A. Tartakovskii, *Quantum Dots: Optics, Electron Transport and Future Applications*, Cambridge: Cambridge University Press, 2012.
2. L. A. Openov, Phys. Rev. B, vol. 60, pp. 8798-8803, 1999.

Scope of the Talk

•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 two quantum dots are coupled by tunneling.

• We derive the optical susceptibilities of the system under weak field excitation and under general initial conditions.

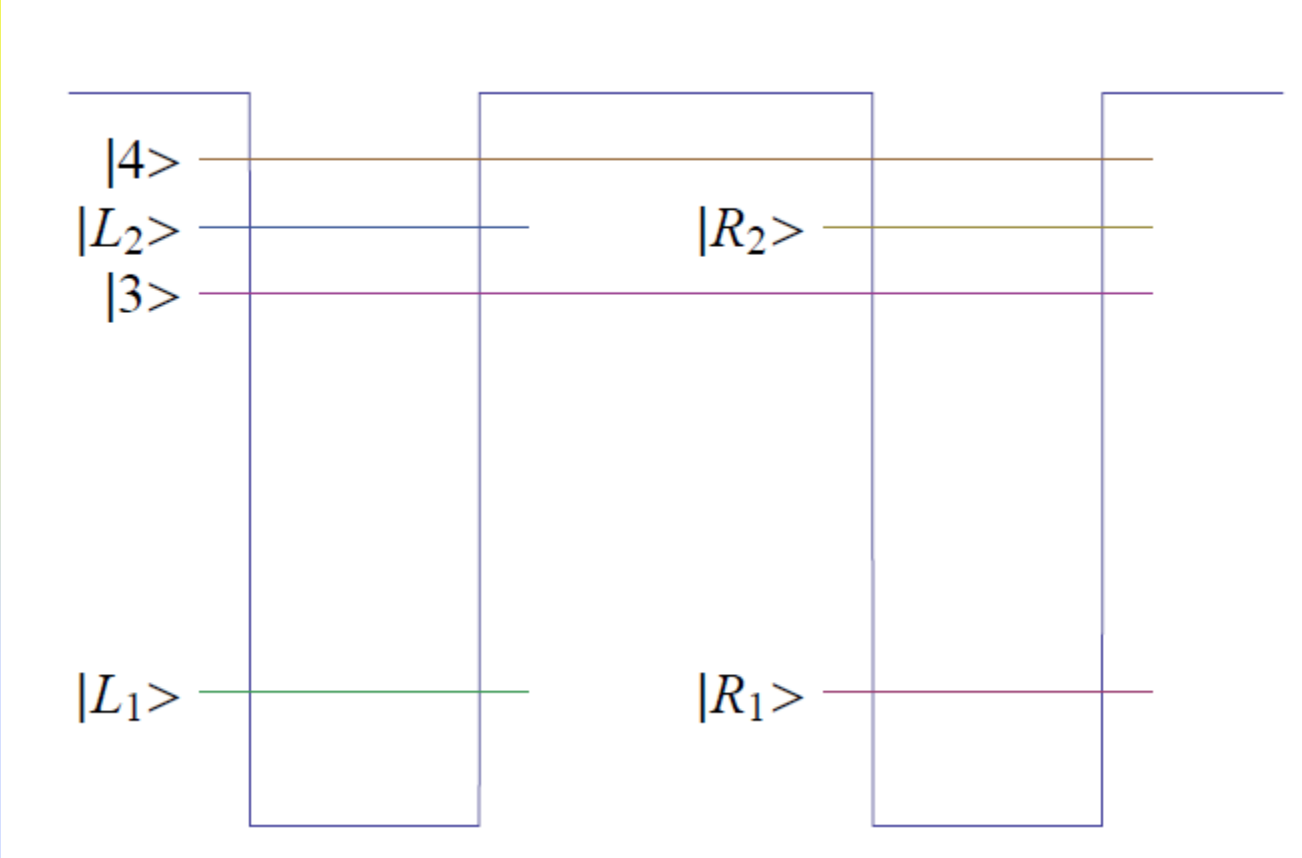
• The formulae we obtain extends that of a previous work, as

(a) the effects of an initial superposition of the two lower states of the system are accounted for

(b) the tunneling of two lower states is included in the model.

• We also present specific results for the form of the susceptibility for different initial states of the system.

Symmetric Double Quantum Dot Nanostructure
First Model



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

First Model – Historic Account

- It was initially proposed by Openov [1] for fully controllable single electron transfer between two quantum dots.
- Since then, different solutions of this problem in the system have been proposed by several authors [2-4].
- The optical response of the system under a weak probe field was studied by Ginzburg and Orenstein [5], giving emphasis to the effects of absorption reduction and creation of low group velocities (slow light).
- In their work the case when the system is initially in a superposition of the two lower states (general initial condition) was not considered.

1. L. A. Openov, Phys. Rev. B, vol. 60, pp. 8798-8803, 1999.
2. E. Paspalakis, Z. Kis, E. Voutsinas, and A. F. Terzis, Phys. Rev. B, vol. 69, art. no. 155316, 2004.
3. A. V. Tsukanov, Phys. Rev. B, vol. 73, art. no. 085308, 2006.
4. S. G. Kosionis, A. F. Terzis, and E. Paspalakis, Phys. Rev. B, vol. 75, art. no. 193305, 2007.
5. P. Ginzburg and M. Orenstein, Opt. Express, vol. 14, pp. 12467-12472, 2006.

Hamiltonian of the System

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

- ε_n is the energy of states $|L_n\rangle$ and $|R_n\rangle$, n = 1,2.
- U is the coupling tunneling coefficient of the two upper levels.
- $E(t)$ is the applied electric field.
- μ is the electric dipole matrix element for the individual quantum dot.

$$E(t) = E_0 \cos(\omega t)$$

Delocalized (Coupled) States

$$
\begin{aligned}
|3\rangle &= \frac{1}{\sqrt{2}}\left(|L_2\rangle + |R_2\rangle\right) , \\
|4\rangle &= \frac{1}{\sqrt{2}}\left(|L_2\rangle - |R_2\rangle\right) ,
\end{aligned}
$$

$$
\begin{aligned}
\varepsilon_3 &= \varepsilon_2 - \hbar U , \\
\varepsilon_4 &= \varepsilon_2 + \hbar U .
\end{aligned}
$$

Probability Amplitudes

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

$$\begin{aligned}
i\hbar\dot{a}_1(t) &= \varepsilon_1 a_1(t) - \frac{\mu E_0}{\sqrt{2}}\cos(\omega t)\left[a_3(t) + a_4(t)\right] , \\
i\hbar\dot{a}_2(t) &= \varepsilon_1 a_2(t) - \frac{\mu E_0}{\sqrt{2}}\cos(\omega t)\left[a_3(t) - a_4(t)\right] , \\
i\hbar\dot{a}_3(t) &= \varepsilon_3 a_3(t) - i\hbar\gamma a_3(t) \\
&- \frac{\mu E_0}{\sqrt{2}}\cos(\omega t)\left[a_1(t) + a_2(t)\right] , \\
i\hbar\dot{a}_4(t) &= \varepsilon_4 a_4(t) - i\hbar\gamma a_4(t) \\
&- \frac{\mu E_0}{\sqrt{2}}\cos(\omega t)\left[a_1(t) - a_2(t)\right] ,
\end{aligned}$$

Change of Variables – Rotating Wave Approximation

$$
\begin{aligned}
a_1(t) &= c_1(t)e^{-i\frac{\varepsilon_1}{\hbar}t}, \\
a_2(t) &= c_2(t)e^{-i\frac{\varepsilon_1}{\hbar}t}, \\
a_3(t) &= c_3(t)e^{-i\frac{\varepsilon_3}{\hbar}t+i\delta t-iUt}, \\
a_4(t) &= c_4(t)e^{-i\frac{\varepsilon_4}{\hbar}t+i\delta t+iUt}.
\end{aligned}
$$

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

$$
\begin{aligned}
i\dot{c}_1(t) &= -\frac{\mu E_0}{2\sqrt{2}\hbar}\left[c_3(t) + c_4(t)\right], \\
i\dot{c}_2(t) &= -\frac{\mu E_0}{2\sqrt{2}\hbar}\left[c_3(t) - c_4(t)\right], \\
i\dot{c}_3(t) &= (\delta - U - i\gamma)c_3(t) \\
&\quad - \frac{\mu E_0}{2\sqrt{2}\hbar}\left[c_1(t) + c_2(t)\right], \\
i\dot{c}_4(t) &= (\delta + U - i\gamma)c_4(t) \\
&\quad - \frac{\mu E_0}{2\sqrt{2}\hbar}\left[c_1(t) - c_2(t)\right],
\end{aligned}
$$

Polarization

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

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

$$\chi(\omega) = \frac{\sqrt{2}N\mu}{\varepsilon_0 E_0}(c_3c_1^* + c_4c_1^* + c_3c_2^* - c_4c_2^*)$$

N is the electron density in the quantum dot structure.

Linear Susceptibility

Initial Condition

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

$$|\alpha|^2 + |\beta|^2 = 1.$$

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

- This formula extends the work of Ginzburg and Orenstein [1] for the case of an initial superposition of the two lower states.

1. P. Ginzburg and M. Orenstein, Opt. Express, vol. 14, pp. 12467-12472, 2006.

Linear Susceptibility – Single Quantum Dot

$$U = 0$$

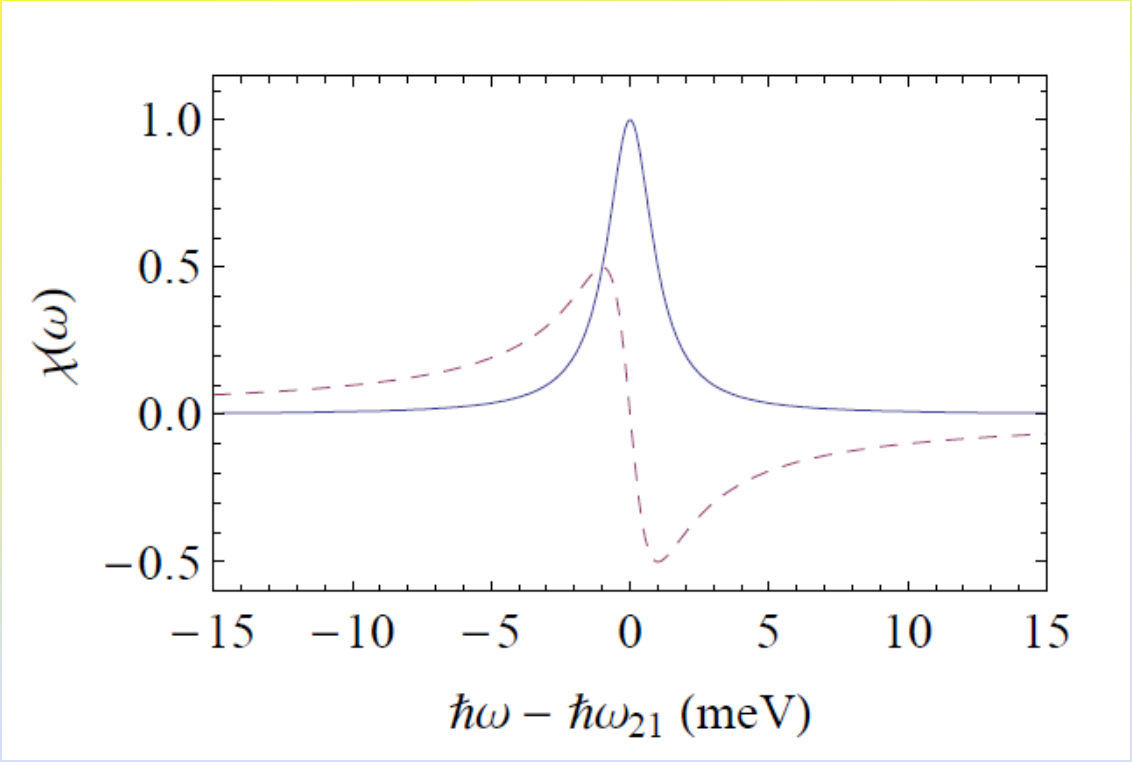
$$\chi(\omega) = \frac{N\mu^2}{\hbar\varepsilon_0} \frac{\omega_{21} - \omega + i\gamma}{(\omega_{21} - \omega)^2 + \gamma^2}$$

Parameters

- We consider GaAs quantum dots.
- The size of each individual quantum dot is 5 nm .
- The height of the well in each quantum dot is 450 meV.
- The barrier length between the two quantum dots is 7 nm.
- We use the shooting method [1] for the numerical solution of the time-independent Schrödinger equation in the effective mass approximation.
- We obtain $\hbar U = 4.9$ meV.
- Additionally, we take $\hbar\gamma = 1$ meV.

1. P. Harrison, *Quantum Wells, Wires and Dots: Theoretical and Computational Physics of Semiconductor Nanostructures*, Hoboken, NJ: Wiley, 2009.

Single Quantum Dot



The form of the real part (dashed curve) and the imaginary part (solid curve) of the susceptibility for a single quantum dot.

The imaginary part, which determines absorption, has a Lorentzian form . The maximum in the absorption is at $\omega = \omega_{21}$.

The real part, which determines dispersion (e.g., the index of refraction), has a regular dispersive form.

One Quantum Dot Initially Occupied

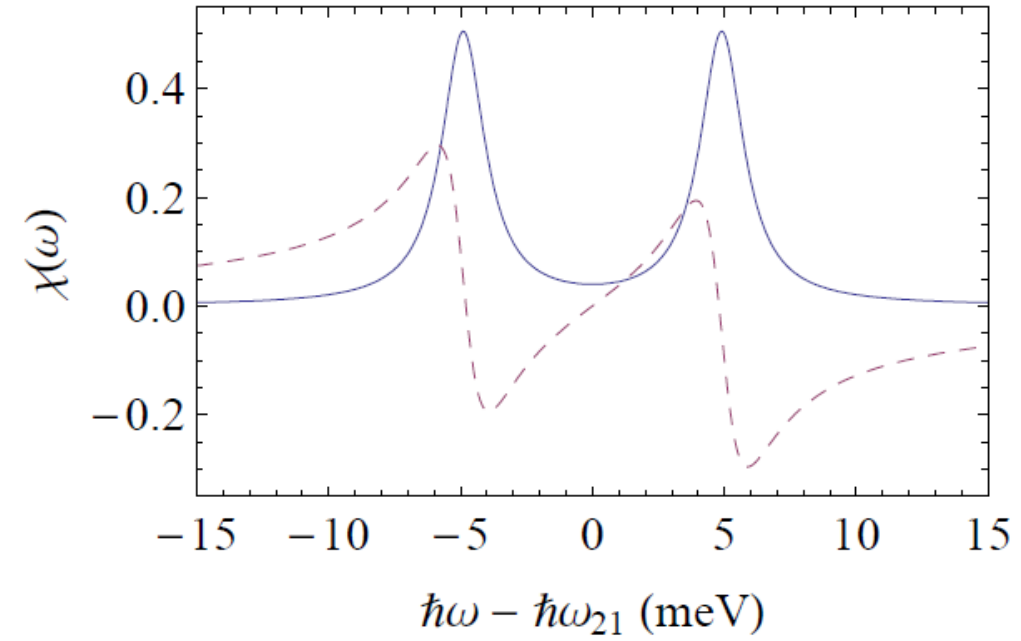
$$\alpha = 1, \beta = 0$$

$$|\psi(0)\rangle = |L_1\rangle$$

or

$$\alpha = 0, \beta = 1$$

$$|\psi(0)\rangle = |R_1\rangle$$

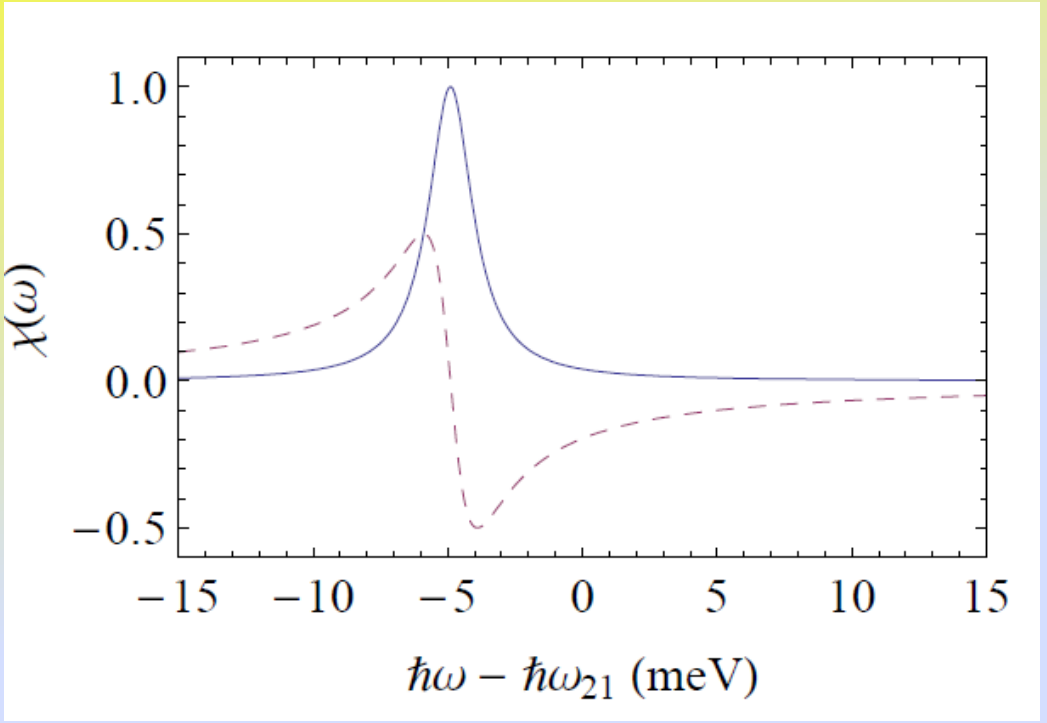


The form of the real part (dashed curve) and the imaginary part (solid curve) of the susceptibility for the symmetric double quantum dot nanostructure. Here, only one of the two quantum dots is initially occupied.

The imaginary part shows a symmetric double-peaked structure, and the peaks are obtained at the energies ε_3-ε_1 and ε_4-ε_1. For $\omega = \omega_{21}$ there is a minimum in the imaginary part of $\chi(\omega)$.

Initially Symmetric Superposition

$$|\psi(0)\rangle = \frac{1}{\sqrt{2}}|L_1\rangle + \frac{1}{\sqrt{2}}|R_1\rangle$$

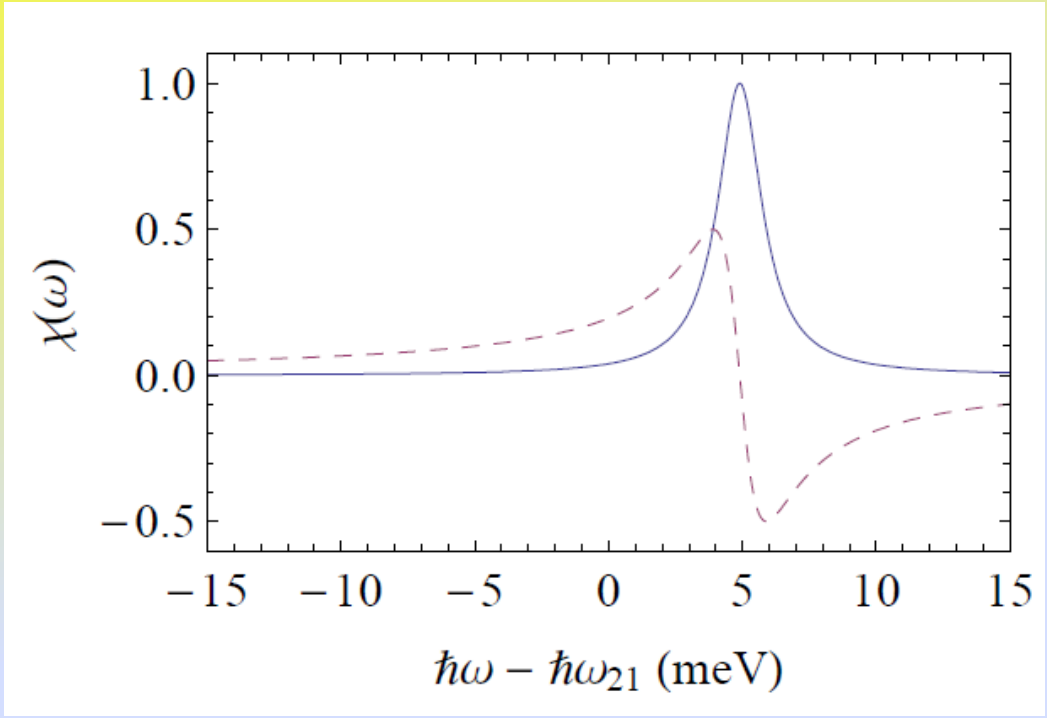


The form of the real part (dashed curve) and the imaginary part (solid curve) of the susceptibility for the symmetric double quantum dot nanostructure for an initial symmetric superposition.

The imaginary part has a single Lorentzian peak around the energy ε_3-ε_1.

Initially Antisymmetric Superposition

$$|\psi(0)\rangle = \frac{1}{\sqrt{2}}|L_1\rangle - \frac{1}{\sqrt{2}}|R_1\rangle$$



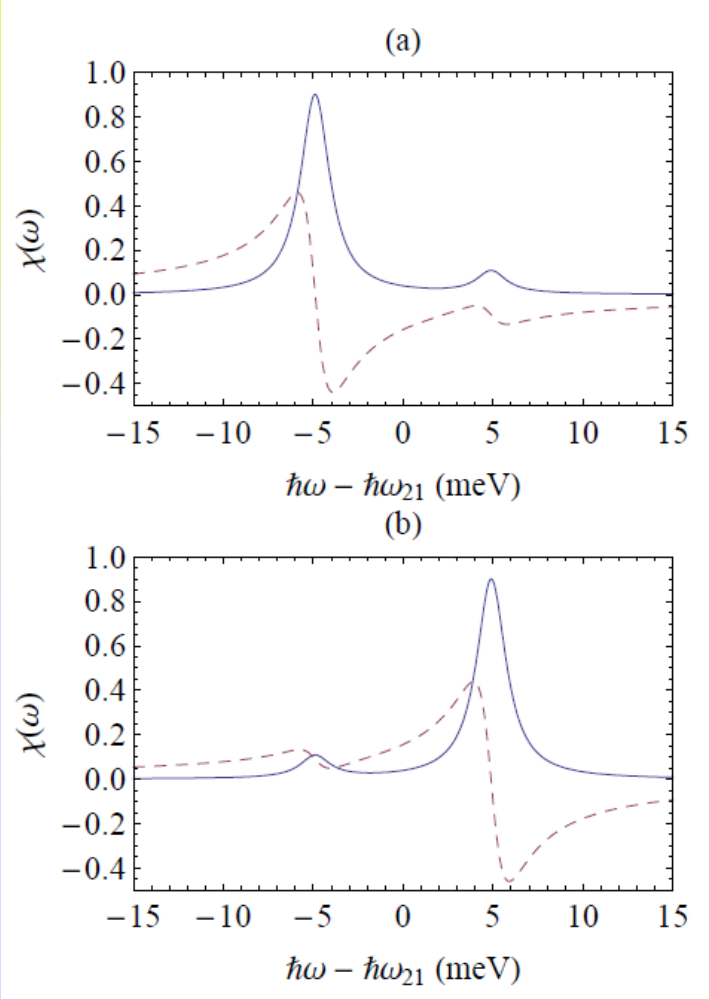
The form of the real part (dashed curve) and the imaginary part (solid curve) of the susceptibility for the symmetric double quantum dot nanostructure for an initial antisymmetric superposition.

The imaginary part has a single Lorentzian peak around the energy ε_4-ε_1.

Most of the Population in One Quantum Dot

$$|\psi(0)\rangle = \frac{1}{\sqrt{5}}|L_1\rangle + \frac{2}{\sqrt{5}}|R_1\rangle$$

$$|\psi(0)\rangle = \frac{1}{\sqrt{5}}|L_1\rangle - \frac{2}{\sqrt{5}}|R_1\rangle$$

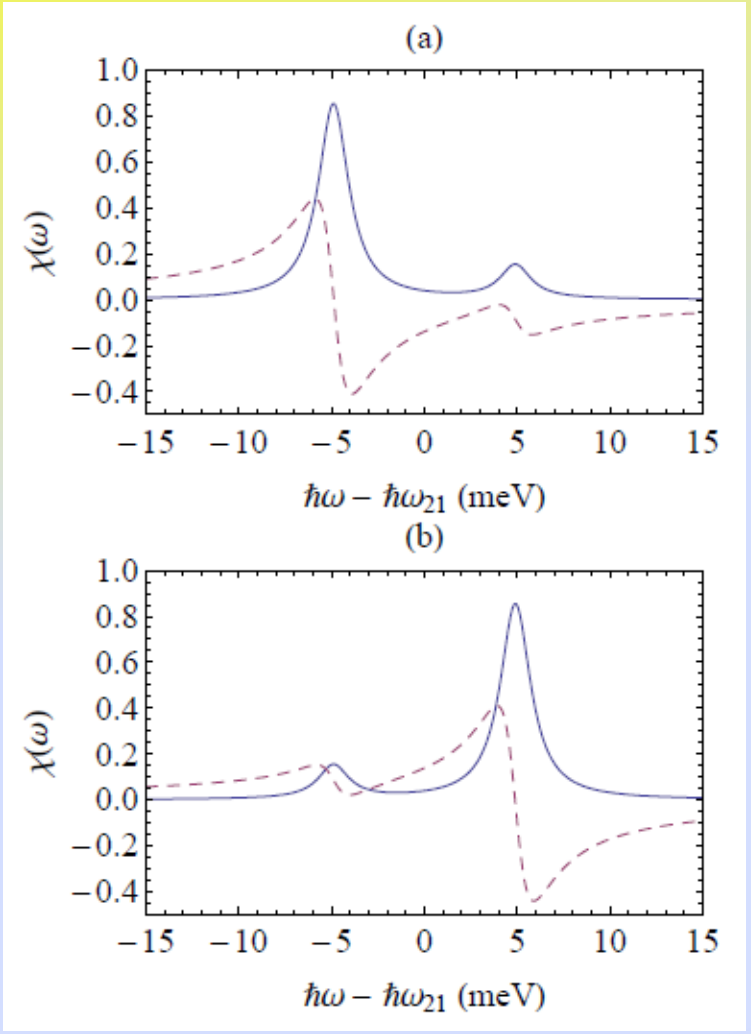


The form of the real part (dashed curve) and the imaginary part (solid curve) of the susceptibility for the symmetric double quantum dot nanostructure.

Equal Population – Complex Phase

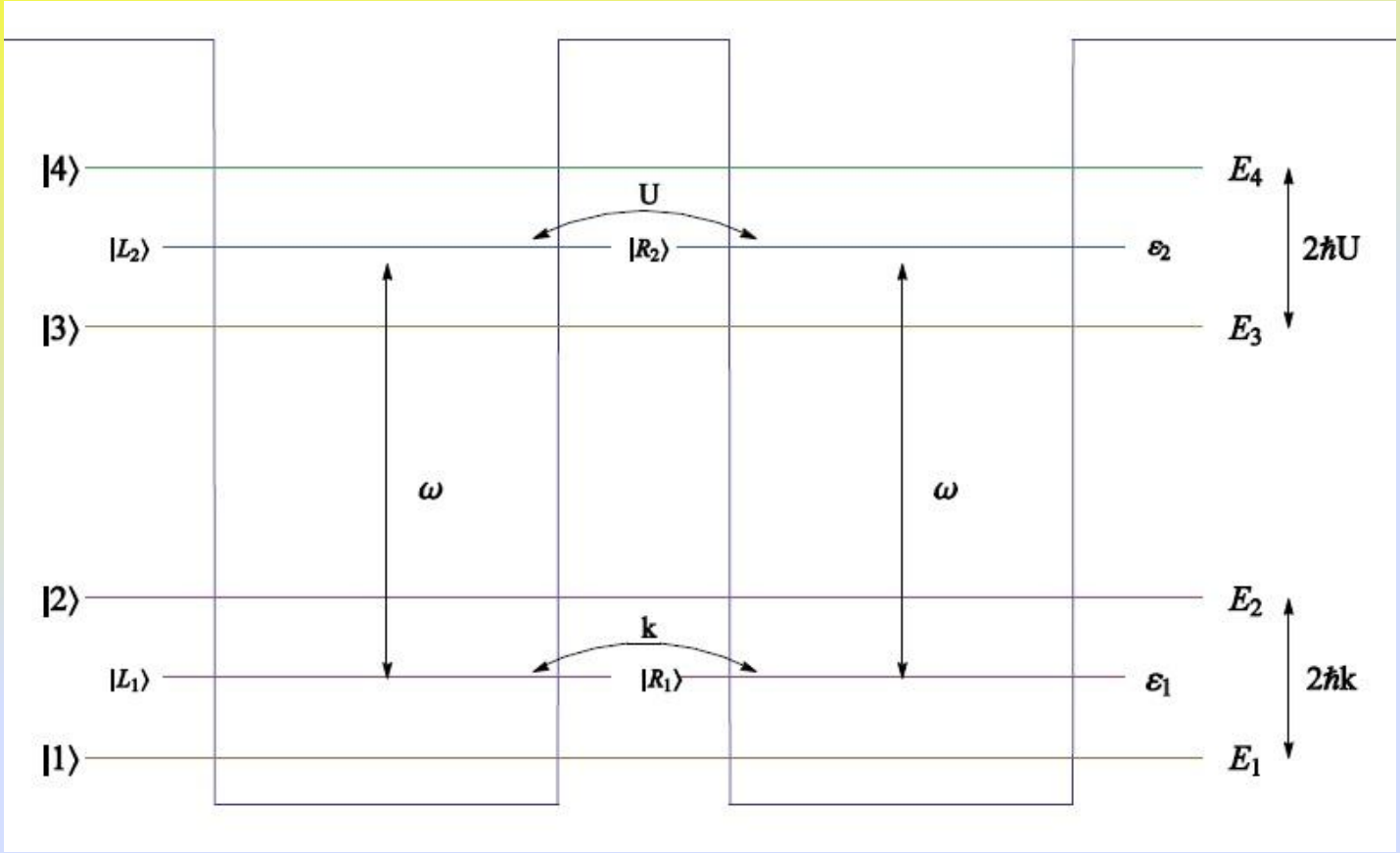
$$|\psi(0)\rangle = \frac{1}{\sqrt{2}}|L_1\rangle + \frac{e^{i\pi/4}}{\sqrt{2}}|R_1\rangle$$

$$|\psi(0)\rangle = \frac{1}{\sqrt{2}}|L_1\rangle + \frac{e^{i3\pi/4}}{\sqrt{2}}|R_1\rangle$$



The form of the real part (dashed curve) and the imaginary part (solid curve) of the susceptibility for the symmetric double quantum dot nanostructure.

Symmetric Double Quantum Dot Nanostructure
Second Model



Schematic diagram of the coupled quantum dot structure studied. We present the localized energy levels $|L_1>$, $|L_2>$, $|R_1>$, $|R_2>$ and the delocalized levels (|1>, |2>, |3>, |4>).

Hamiltonian of the System

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

- ε_n is the energy of states $|L_n\rangle$ and $|R_n\rangle$, n = 1,2.
- k is the coupling tunneling coefficient of the two lower levels.
- U is the coupling tunneling coefficient of the two upper levels.
- $E(t)$ is the applied electric field.
- μ is the electric dipole matrix element for the individual quantum dot.

Delocalized (Coupled) States

$$|1\rangle = \frac{1}{\sqrt{2}}\left(|L_1\rangle + |R_1\rangle\right)$$
$$|2\rangle = \frac{1}{\sqrt{2}}\left(|L_1\rangle - |R_1\rangle\right)$$

$$|3\rangle = \frac{1}{\sqrt{2}}\left(|L_2\rangle + |R_2\rangle\right) ,$$
$$|4\rangle = \frac{1}{\sqrt{2}}\left(|L_2\rangle - |R_2\rangle\right) ,$$

$$\epsilon_1 = \varepsilon_1 - \hbar k$$
$$\epsilon_2 = \varepsilon_1 + \hbar k$$

$$\varepsilon_3 = \varepsilon_2 - \hbar U ,$$
$$\varepsilon_4 = \varepsilon_2 + \hbar U .$$

Linear Susceptibility

Initial Condition

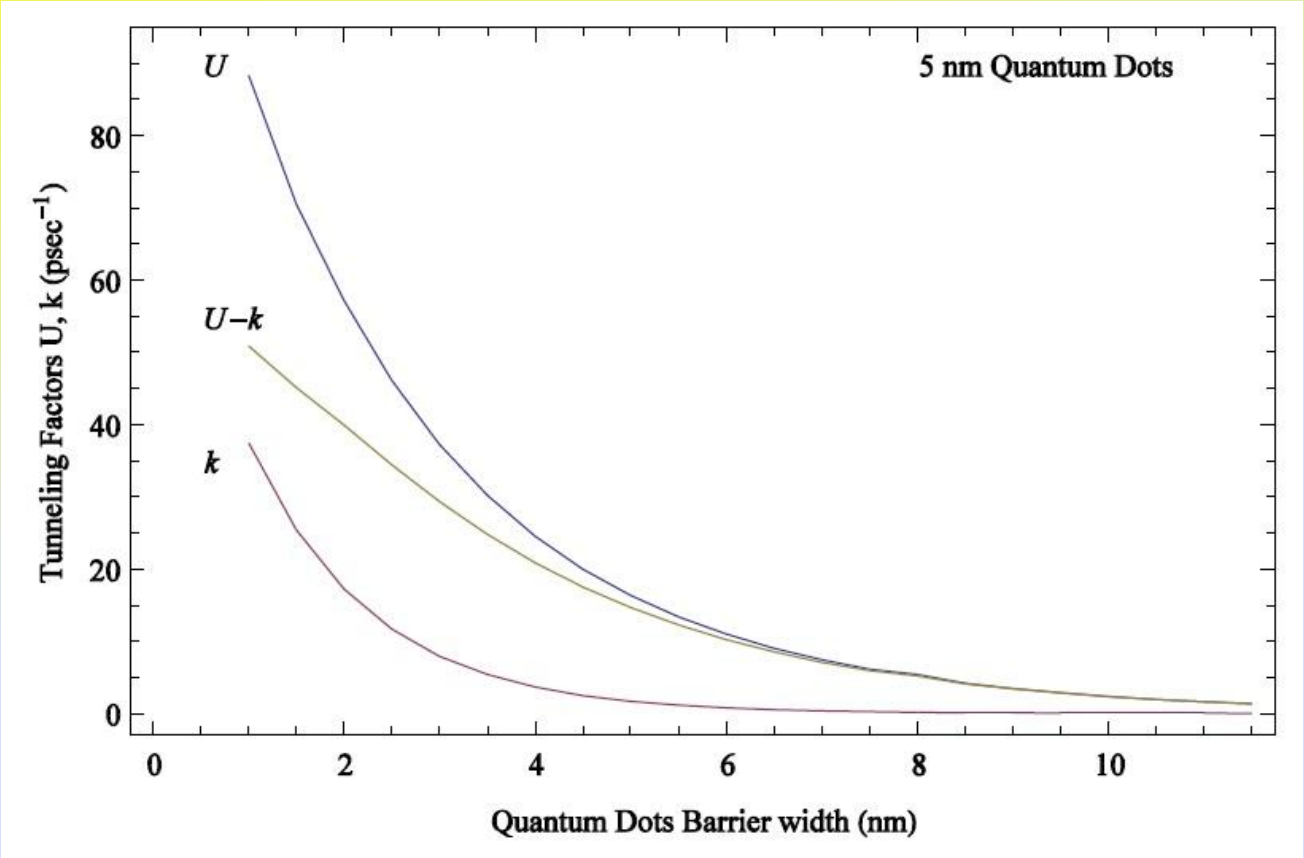
$$|\psi(0)\rangle = \alpha|L_1\rangle + \beta|R_1\rangle$$

$$|\alpha|^2 + |\beta|^2 = 1.$$

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

• This formula includes both the case of an initial superposition of the two lower states and the tunneling between the lower states as well.

Dependence of the Tunneling Coefficients on the Distance Between the Two Quantum Dots



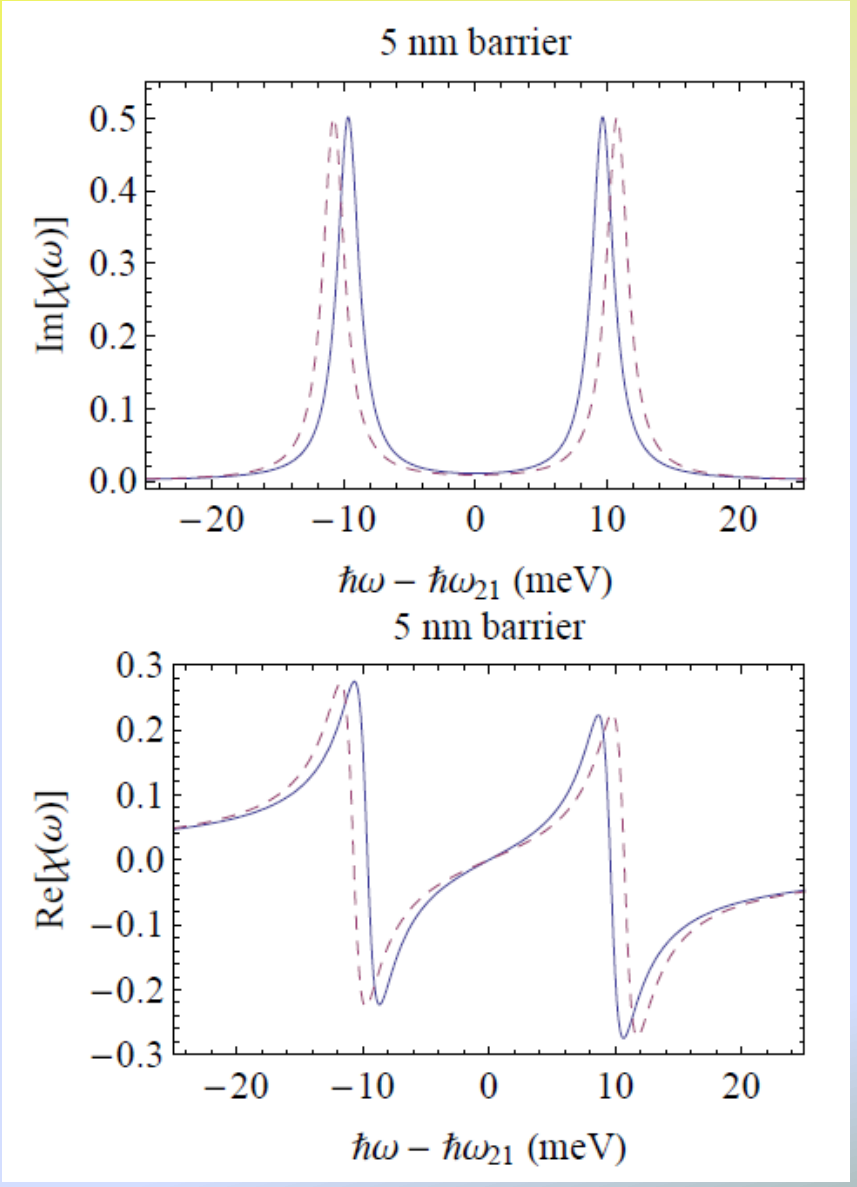
• The results have been obtained with the use of the shooting method for the numerical solution of the time-independent Schrödinger equation in the effective mass approximation.

Comparison of the Two Models (1)

$$|\psi(0)\rangle = |L_1\rangle$$

or

$$|\psi(0)\rangle = |R_1\rangle$$



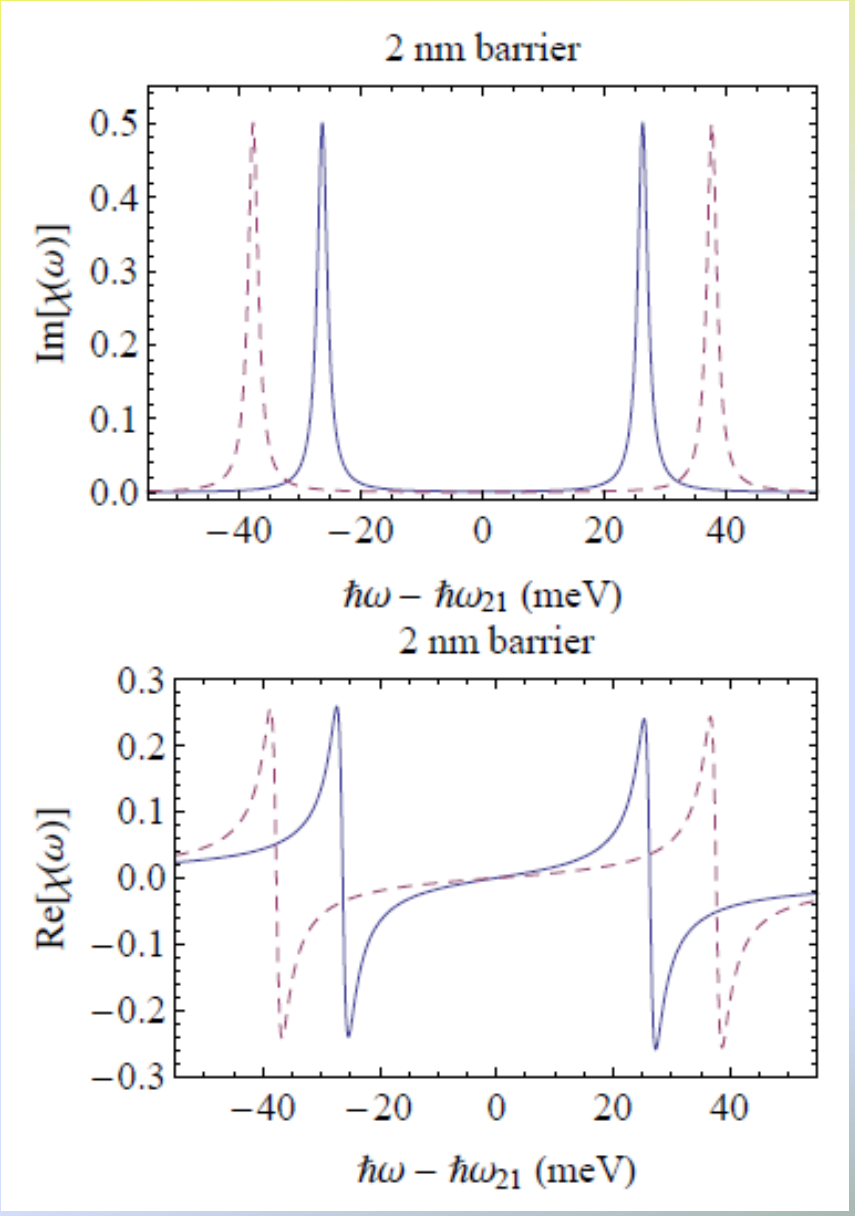
The form of the real part and the imaginary part of the susceptibility for the symmetric double quantum dot nanostructure. Dashed curve (first model), solid curve (second model) for a 5 nm GaAs quantum dot with 5 nm barrier.

Comparison of the Two Models (2)

$$|\psi(0)\rangle = |L_1\rangle$$

or

$$|\psi(0)\rangle = |R_1\rangle$$



The form of the real part and the imaginary part of the susceptibility for the symmetric double quantum dot nanostructure. Dashed curve (first model), solid curve (second model) for a 5 nm GaAs quantum dot with 2 nm barrier.

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.
- 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.
- We considered two cases: (a) the effect of tunneling is only included in the coupling of the upper levels and (b) the effect of tunneling is included in the coupling of both lower and upper levels .
- We showed that the form of the real and imaginary parts of the susceptibility depends strongly on the actual form of the initial superposition.

Acknowledgements

- This research has been co-financed by the European Union (European Social Fund - ESF) and Greek national funds through the Operational Program "Education and Lifelong Learning" of the National Strategic Reference Framework (NSRF) - Research Funding Program: Archimedes III.

- We thank Professor Sotirios Baskoutas for useful discussions and help.