Controlling the Pump-Probe Optical Response in Asymmetric Tunneling-Controlled Double Quantum Dot Molecule—Metal Nanoparticle Hybrids

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Abstract: In the present work, we investigate the modified nonlinear pump-probe optical properties due to the excitonic–plasmonic interaction of a double semiconductor quantum dot (SQD) molecule coupled to a metal nanoparticle (MNP). More specifically, we study the absorption and the dispersion spectra of a weak electromagnetic field in a hybrid structure with two counterparts, a molecule of two coupled SQDs, and a spherical MNP driven by a field of high intensity. We solve the relevant density matrix equations, calculate the first-order optical susceptibility of the probe field in the strong pumping regime, and investigate the way in which the distance between the two counterparts modifies the optical response, for a variety of values of the physical constants of the system, including the pump-field detuning, the tunnelling rate, and the energy separation gap associated with the excited states of the coupled SQDs.

Keywords: coupled quantum dots; equations of motion; hybrid nanostructure; metal nanoparticle; optical susceptibility; pump-probe response

1. Introduction

In current studies in photonics, coupled structures formed from semiconductor quantum dots and metal particles at the nanoscale play an important role as they give, in many cases, enhanced optical properties in comparison to individual SQDs and MNPs [1]. The coupling of the excitons to plasmons is responsible for the emergence of interesting optical effects from the SQD-MNP structures, including altered excitonic Rabi oscillations [2–4], creation and control of optical bistability and multistability [5–9], population transfer to the exciton [10–14] or biexciton state with high accuracy [15–17], controlled slow light [18], creation of gain without inversion [19–21], strong enhancement of second harmonic generation [22], and strong modification of four-wave mixing, as well as self- or cross-Kerr nonlinearity [23–26], control of resonance fluorescence [27–30], and several others [31–38]. These effects can find applications in the construction of efficient optical devices which can be implemented in the fields of nanophotonics and quantum technology.

In some of these studies, the nonlinear optical effects in an SQD-MNP hybrid structure are examined under interaction with a pump field of high intensity and a tunable probe field of much lower intensity [7,18,23,24,26,28,34,36]. A specific SQD nanostructure that exhibits optical properties of high importance is the asymmetric molecule consisting of a couple of SQDs [39–52]. The band structures of the SQDs are considered to be different from each other, while the rate at which the tunneling effects take place is determined by the value of the applied voltage. More recently, the optical properties of a hybrid structure composed of an asymmetric double SQD molecule coupled to a MNP have been analyzed [32,38]. In these studies, the construction of a diffraction grating, the special characteristics of which can be modified due to plasmonic mediated interaction [32], and the control and the energy absorption rate spectrum [38] were explored. Here, we analyze...
the response of a system of an MNP coupled to a doublet of SQDs interacting with a pump field of high intensity.

Specifically, we combine the theoretical approaches presented in Refs. [36,46] and calculate the optical susceptibility of the probe field, while a second field of much higher intensity is used for the pumping of the system. Then, we analyze the spectral profile of the real and the imaginary part of the susceptibility, for different values of the physical parameters of the structure. We find that, at the exact-resonance case with the pump field, the spectra present exact symmetry and are composed of a quartet of resonances. In the off-resonance case, the spectra exhibit six resonances and are not symmetric, for average values of the tunneling coefficient. For high interparticle distances, analytical expressions for the position of the resonances are provided based on the dressed-states analysis, which is valid in the strong intensity regime applied in the present paper. For low tunneling rates, the contribution of MNP outweighs the contribution of the coupled SQDs and it also introduces a non-negligible slope to the spectra of the total susceptibility. However, for higher values of tunneling coupling coefficient, the contribution of the double SQD molecule seems to play a more important role in the spectral profile of the total susceptibility, determining to a substantial extent the shape of its imaginary part. In every case, as the distance between the counterparts decreases, the resonances are transposed away from the center of the spectra.

This study is structured as follows. In the first part, the details of the applied methodologies are presented and the equations of motion are derived in the density matrix formalism, which should be numerically solved, in order to calculate the optical susceptibility of the probe field as it interacts with the strongly pumped hybrid nanostructure, and, in the second part, a dressed state representation of the system is introduced in the analysis. Then, in the third part of this work, the results associated with the behavior of the optical susceptibility are analyzed, for different parameters of the structure. Finally, in the last part of this study, the conclusions of the above analysis are summarized.

2. Methods

The hybrid system studied has two counterparts: a spherical MNP with dielectric function \( \varepsilon_m(\omega) \) and a molecule of two coupled SQDs with dielectric constant \( \varepsilon_S \), as seen in Figure 1. Moreover, we consider that the condition \( R > a \) is met, with \( R \) and \( a \) respectively denoting the interparticle distance and the radius of the MNP, and we assume that the size of the SQD is negligible as compared to the size of the MNP. The structure is surrounded by a material with dielectric constant \( \varepsilon_{env} \).

![Figure 1. Schematic diagram of the MNP–double-SQD hybrid system, \( a \), \( b \), and \( R \) respectively representing the radius of MNP, the radius of the SQDs, and the center-to-center distance between an SQD and the MNP. Between the SQDs, electron tunneling is present.](image)

The hybrid system interacts with two linearly polarized, oscillating electromagnetic fields, where \( E_i \) and \( \omega_i \) respectively represent the corresponding amplitude and the angular...
frequency \((i = a, \text{ for the pump field and } i = b, \text{ for the probe field})\), that excite plasmons on the surface of the MNP, providing a strong, continuous spectral response. An energy-level scheme is presented in Figure 2, where the possible transitions are also depicted. The coupling between the two SQDs with different band structures, which are typically placed in distances between 7 and 10 nm, owes its presence to the electron tunneling. The coupled SQDs are represented as a three-level quantum system \([32,38–52]\). With \(|0\rangle\) we denote the ground state, and \(|1\rangle\) (direct exciton) corresponds to the state describing a single electron excitation, in which case the excited electron, as well as the hole that is created, are both confined within the same quantum dot. On the other hand, an electron can be excited in state \(|2\rangle\) (indirect exciton) when it is conveyed from the first quantum dot to the second one, due to tunneling effects following the excitation pathway. The incident field induces an oscillating dipole moment and excites electrons from the ground state \(|0\rangle\) to the (direct) exciton state, where an electron–hole pair is bound to the same SQD. The direct exciton is also coupled to the states of the surface plasmons due to the long-range Coulomb interaction that permits the exchange of energy between the counterparts \([53–56]\). The tunneling barrier can be controlled by properly adjusting the voltage of an external gate electrode structure \([39–52]\).

![Figure 2. Schematic energy-level configuration of the hybrid system. The MNP is described as a continuum of energy states, while the asymmetric double SQD molecule is depicted as a three-level \(\Lambda\)-type system with a ground state \(|0\rangle\), a direct exciton state \(|1\rangle\) (electron-hole pair in the same quantum dot), and an indirect exciton state \(|2\rangle\) (due to the transferring of the excited electron to the excited state of the second quantum dot). The application of a threshold voltage to the double SQD induces strong tunneling between the upper levels, while, at the same time, the ground energy levels are effectively decoupled.](image)

In Equation (1), we present the Hamiltonian that describes the interaction of the hybrid system with a pump and a probe field, as depicted in Figure 2, in the dipole approximation.

\[
H = E_0|0\rangle\langle 0| + E_1|1\rangle\langle 1| + E_2|2\rangle\langle 2| + \hbar T_e (|1\rangle\langle 2| + |2\rangle\langle 1|) - \mu E_{S\!Q\!D}(|0\rangle\langle 1| + |1\rangle\langle 0|). \tag{1}
\]

The first three terms in Equation (1) describe the unperturbed system, with \(E_0\) being the energy of level \(|n\rangle\) (for the ground state, we take \(E_0 = 0\)), while the fourth term owes its presence to the tunneling effects between the SQDs, with \(T_e\) expressing the rate at which electrons tunnel within the SQD molecule. The last term in Equation (1) describes the interaction between the electromagnetic fields and the SQD molecule. Here, \(\mu\) and \(E_{\text{S\!Q\!D}}\) respectively represent the dipole moment associated to the excitation \(|0\rangle \leftrightarrow |1\rangle\) and the total field detected at the position of the SQD molecule, which is given by the formula

\[
E_{\text{S\!Q\!D}}(t) = \frac{\hbar}{\mu} \sum_{i=a,b} \left\{ [\Omega_i + G_i\rho_{10}(t)]e^{-i\omega_{10}t} + [\Omega_i^* + G_i^*\rho_{01}(t)]e^{i\omega_{10}t} \right\}, \tag{2}
\]
where

$$\Omega_i = \frac{E_i \mu}{2 \hbar \varepsilon_{\text{eff}} S} \left(1 + \frac{s_a \gamma(\Delta^2)}{R^3}\right),$$  \hspace{1cm} (3)

$$G_i = \sum_{n=1}^{N} \frac{1}{4\pi \varepsilon_{\text{env}}} \frac{s_n \gamma_n(\omega_i) \Delta^{2n+1} \epsilon^2}{\hbar^2 \varepsilon_{\text{eff}} S^{2n+4}},$$  \hspace{1cm} (4)

with $\gamma_n(\omega_i) = [\epsilon_{\text{env}} - \epsilon_m(\omega_i)] / [(n + 1) \epsilon_{\text{env}} / n + \epsilon_m(\omega_i)]$ and $\epsilon_{\text{eff}} S = (\epsilon_S + 2\epsilon_{\text{env}}) / 3\epsilon_{\text{env}}$.

In the above, we have used the quasistatic approximation, where retardation effects are omitted, following several previous works [2–38,53–56]. The first term in the expression of Equation (3) is ascribed to the direct coupling between the applied electromagnetic field and the SQDs. We expect that the MNP interacts with the applied field, as well as the field emitted by the SQDs, and as a result, it emits in its surrounding space. The coupling between the SQDs and the first (second) component of this emitted field is associated with the second term of Equation (3) (the $G_i$ parameter). The parameters $s_a$ and $s_n$, in the case of an incident field polarized along the direction of the interparticle axis, are respectively equal to 2 and $(n + 1)^2$, while, for an incident field that is polarized perpendicularly to the direction of the interparticle axis, we take $s_a = -1$ and $s_n = n(n + 1) / 2$. The $G_i$ parameter is calculated including the multipole effects for the MNP [55].

Based on the expression of the Hamiltonian that describes the system, in the rotating wave approximation, a set of nine density-matrix differential equations is derived. The number of these equations is eliminated to five, under the conditions $\sum_{i}^{n} \rho_{ii} = 1$ and $\rho_{nm} = \rho_{nm}^\dagger$, with $n \neq m$:

$$\dot{\rho}_{01}(t) = i(\Delta + i\gamma_{01})\rho_{01}(t) + iT_{r}\rho_{02}(t) - i\left[\Omega_a^\dagger + G_a\rho_{01}(t) + \Omega_b^\dagger e^{it\delta} + G_b^\dagger\rho_{01}(t)\right][\rho_{00}(t) - \rho_{11}(t)],$$  \hspace{1cm} (5)

$$\dot{\rho}_{02}(t) = -i(-\Delta + \omega_{12} - i\gamma_{02})\rho_{02}(t) + iT_{r}\rho_{01}(t) + i\left[\Omega_a^\dagger + G_a\rho_{01}(t) + \Omega_b^\dagger e^{it\delta} + G_b^\dagger\rho_{01}(t)\right]\rho_{12}(t),$$  \hspace{1cm} (6)

$$\dot{\rho}_{12}(t) = i(-\omega_{12} + i\gamma_{12})\rho_{12}(t) + iT_{r}[1 - \rho_{00}(t) - 2\rho_{11}(t)] + i\left[\Omega_a + G_a\rho_{10}(t) + \Omega_b e^{-it\delta} + G_b\rho_{10}(t)\right]\rho_{02}(t),$$  \hspace{1cm} (7)

$$\dot{\rho}_{00}(t) = -i\left[\Omega_a + G_a\rho_{10}(t) + \Omega_b e^{-it\delta} + G_b\rho_{10}(t)\right]\rho_{00}(t) + iT_{r}[\rho_{10}(t) - \Gamma_{01}\rho_{11}(t)] + IT_{r}\rho_{12}(t) - \Gamma_{01}\rho_{11}(t),$$  \hspace{1cm} (8)

$$\dot{\rho}_{11}(t) = i\left[\Omega_a + G_a\rho_{10}(t) + \Omega_b e^{-it\delta} + G_b\rho_{10}(t)\right]\rho_{11}(t) - iT_{r}\rho_{12}(t) - \Gamma_{10}\rho_{11}(t) - (\Gamma_{10} + \Gamma_{12})\rho_{11}(t),$$  \hspace{1cm} (9)

where the parameters $\delta = \omega_b - \omega_a$ and $\Delta = (E_1 - E_0) / \hbar - \omega_a$ respectively denote the difference of the angular frequencies with reference to the pump and the probe field and the detuning from the energy gap between the energy levels $|0\rangle$ and $|1\rangle$. The $|1\rangle \leftrightarrow |2\rangle$ transition corresponds to the energy difference $\hbar \omega_{12}$. The dephasing rates are symbolized with $\gamma_{nm}$, while the population decay rate for the transition $|n\rangle \rightarrow |m\rangle$ is indicated with $\Gamma_{nm}$.

To calculate the first-order susceptibility of the hybrid structure, aiming at the examination of the pump-probe optical response, we apply to the first-order Taylor expansion of $\rho_{nm}$, according to the first-order approximation, in the probe field Rabi frequency:

$$\rho_{nm} = \rho_{nm}^{(1)} + \Omega_b e^{it\delta}\rho_{nm}^{(2)} + \Omega_b e^{-it\delta}\rho_{nm}^{(3)}.$$  \hspace{1cm} (10)

In the expansions presented in Equation (10), the terms $\rho_{nm}^{(2)}$ and $\rho_{nm}^{(3)}$ are considered small, in the sense that $|\rho_{nm}^{(1)}| > |\rho_{nm}^{(2)}|$ and $|\rho_{nm}^{(3)}|$. Since the intensity of the pump field is
assumed to be four orders of magnitude higher than the intensity of the probe field \((\Omega_a \gg \Omega_b)\), we should maintain in our calculations all the orders of its interaction with the hybrid structure. By substituting the expanded density-matrix elements in Equations (5)–(9), the set of Equations (11)–(25) is derived.

\[
\hat{A}_1 = i(\Omega_a^* + G^* A_1)(E_1 - D_1) + i(\Delta + i\gamma_{10})A_1 + iT_e B_1,
\]

\[
\hat{A}_2 = -i\delta A_2 + i(\Omega_b^* + G^* A_2)(E_1 - D_1) + i(\Omega_a^* + G^* A_1)(E_2 - D_2) + i(\Delta + i\gamma_{10})A_2 + iT_e B_2,
\]

\[
\hat{A}_3 = i\delta A_3 + iG^*(E_1 - D_1)A_3 + i(\Omega_a^* + G^* A_1)(E_3 - D_3) + i(\Delta + i\gamma_{10})A_3 + iT_e B_3,
\]

\[
\hat{B}_1 = i(\Omega_a^* + G^* A_1)C_1 + i(\Delta - \omega_{12} + i\gamma_{02})B_1 + iT_e A_1,
\]

\[
\hat{B}_2 = -i\delta B_2 + i(\Omega_b^* + G^* A_2)C_1 + i(\Omega_a^* + G^* A_1)C_2 + i(\Delta - \omega_{12} + i\gamma_{02})B_2 + iT_e A_2,
\]

\[
\hat{B}_3 = i\delta B_3 + iG^*A_3C_1 + i(\Omega_a^* + G^* A_1)C_3 + i(\Delta - \omega_{12} + i\gamma_{02})B_3 + iT_e A_3,
\]

\[
\hat{C}_1 = i(\Omega_a + GA_1^*)B_1 - iT_e(1 - D_1 - 2E_1) - i(\omega_{12} - i\gamma_{12})C_1,
\]

\[
\hat{C}_2 = -i\delta C_2 + iGA_3B_1 + i(\Omega_a + GA_1^*)B_2 + iT_e(D_2 + 2E_2) - i(\omega_{12} - i\gamma_{12})C_2,
\]

\[
\hat{C}_3 = i\delta C_3 + i(\Omega_a + GA_1^*)B_3 + i(\Omega_b + GA_2^*)B_1 + iT_e(D_3 + 2E_3) - i(\omega_{12} - i\gamma_{12})C_3,
\]

\[
\hat{D}_1 = -i(\Omega_a + GA_1^*)A_1 + i(\Omega_a^* + G^* A_1)A_1^* + \Gamma_{10}E_1 + \Gamma_{20}(1 - D_1 - E_1),
\]

\[
\hat{D}_2 = -i\delta D_2 - i(\Omega_a + GA_1^*)A_2 + i(\Omega_a^* + G^* A_1)A_2^* - iGA_1A_3 + i(\Omega_a^* + G^* A_2)A_1^* + \Gamma_{10}E_2 - \Gamma_{20}(D_2 + E_2),
\]

\[
\hat{D}_3 = i\delta D_3 - i(\Omega_a + GA_1^*)A_3 + i(\Omega_a^* + G^* A_1)A_3^* - i(\Omega_b + GA_2^*)A_1 + iG^*A_1^*A_3 + \Gamma_{10}E_3 - \Gamma_{20}(D_3 + E_3),
\]

\[
\hat{E}_1 = -i(\Omega_a^* + G^* A_1)A_1^* + i(\Omega_a + GA_1^*)A_1 + iT_e(C_1 - C_1^*) - (\Gamma_{10} + \Gamma_{12})E_1,
\]

\[
\hat{E}_2 = -i\delta E_2 - i(\Omega_a^* + G^* A_1)A_2^* + i(\Omega_a + GA_1^*)A_2 - i(\Omega_b^* + G^* A_2)A_1^* + iT_e(C_2 - C_2^*) - (\Gamma_{10} + \Gamma_{12})E_2,
\]

\[
\hat{E}_3 = i\delta E_3 - i(\Omega_a^* + G^* A_1)A_3^* + i(\Omega_a + GA_1^*)A_3 - iG^*A_1^*A_3 + iT_e(C_3 - C_3^*) - (\Gamma_{10} + \Gamma_{12})E_3,
\]

where we have introduced the simplified notation

\[
\rho_{01}^{(n)} = A_n, \quad \rho_{02}^{(n)} = B_n, \quad \rho_{12}^{(n)} = C_n, \quad \rho_{11}^{(n)} = D_n, \quad \rho_{11}^{(n)} = E_n,
\]

for the coefficients defined in Equations (10) and \(G = G_a + G_b\).

For a hybrid system that is strongly pumped, the probe-field first-order absorption and the dispersion are respectively equal to \(-\text{Im}(\rho_{01}^{(2)})\) and \(\text{Re}(\rho_{01}^{(2)})\). More specifically, the first-order probe-field optical susceptibility, associated with the double SQD molecule, under the interaction with a pump field, is described by the following analytical expression:

\[
\chi_{\text{SQD}}^{(1)} = 2 \left( \frac{\Gamma}{\nu} \right) \frac{\mu \rho_{01}^{(2)}}{\epsilon_0 E_0},
\]

where the coefficient \(\rho_{01}^{(2)}(T)\) is calculated at time \(T\), which is a sufficient time interval for our system to reach its steady state. In Equation (27), \(\epsilon_0\) denotes the dielectric constant on
vacuum, and with \( V \) and \( \Gamma \) we respectively indicate the SQD volume and the optical confinement factor [7]. To perform this calculation, we numerically solve Equations (11)–(26).

The expression describing the MNP optical susceptibility at the first order [6] is given by the following expression:

\[
\chi_{\text{MNP}}^{(1)} = \frac{3\gamma_{1}r_{\text{env}}}{\epsilon_0} + \frac{3\gamma_{1}r_{\text{eff}}^{(2)*}}{2\pi\epsilon_{0}\nu_{0}\epsilon_{\text{env}}}P_{01}^{(2)*} + \frac{3\gamma_{2}r_{\text{env}}}{4\pi\epsilon_{0}\epsilon_{\text{env}}^{2}c^{2}}\chi_{\text{SQD}}^{(1)}.
\]  

(28)

At this point, if we sum the first-order susceptibilities of the individual components, we derive an expression for the optical susceptibility for the total hybrid structure at the first order:

\[
\chi_{\text{tot}}^{(1)} = \chi_{\text{SQD}}^{(1)} + \chi_{\text{MNP}}^{(1)}.
\]  

(29)

The dressed-states picture provides us with an illuminating physical insight for the results that are presented in the next section, in the specific case of a double SQD molecule non-interacting with the MNP (practically, for an interdistance above 50 nm). Under this condition, the expression of the Hamiltonian, given by Equation (1), is simplified. Under the assumption of interaction only with a strong pump field, the population amplitudes of the energy levels satisfy the expression

\[
i\hbar \frac{d}{dt} \begin{bmatrix} a_0(t) \\ a_1(t) \\ a_2(t) \end{bmatrix} = H_a \begin{bmatrix} a_0(t) \\ a_1(t) \\ a_2(t) \end{bmatrix}, \text{ with } H_a = \begin{bmatrix} E_0 & -i\hbar \Omega_{a} e^{-i\omega_a t} & 0 \\ -i\hbar \Omega_{a} e^{i\omega_a t} & E_1 & -i\hbar T_e \\ 0 & -i\hbar T_e & E_2 \end{bmatrix}.
\]  

(30)

If we apply the transformations \( a_0(t) = c_0(t), a_1(t) = c_1(t)e^{i(\Delta - i\omega_1)/\hbar} \), and \( a_2(t) = c_2(t)e^{i(\Delta - i\omega_2)/\hbar} \) in Equation (28), the Hamiltonian is written as follows:

\[
i\hbar \frac{d}{dt} \begin{bmatrix} c_0(t) \\ c_1(t) \\ c_2(t) \end{bmatrix} = H_b \begin{bmatrix} c_0(t) \\ c_1(t) \\ c_2(t) \end{bmatrix}, \text{ with } H_b = \begin{bmatrix} 0 & -i\hbar \Omega_{a}^* & 0 \\ -i\hbar \Omega_{a} & h\Delta & -i\hbar T_e \\ 0 & i\hbar T_e & h(\Delta - i\omega_1) \end{bmatrix}.
\]  

(31)

The solution to the eigenvalue/eigenstate problem \( |H_b - \lambda I| = 0 \), under the condition \( \Delta = \omega_1 \), yields the eigenvalue \( \lambda_0 = 0 \), which is associated with the dark state \( |\phi\rangle = (T_e/2) + \Omega_{a}|2\rangle/\sqrt{\Omega_{a}^2 + T_e^2} \), as well as the eigenvalues \( \lambda_{\pm} = \hbar(\Delta \pm \sqrt{\Delta^2 + 4\Omega_{a}^2 + 4T_e^2})/2 \), corresponding to the dressed states

\[
|\pm\rangle = \frac{-2\Omega_{a}|0\rangle + \left(\Delta \pm \sqrt{\Delta^2 + 4\Omega_{a}^2 + 4T_e^2}\right)|1\rangle + 2T_e|2\rangle}{\sqrt{2(\Delta^2 + 4T_e^2 + 4\Omega_{a}^2 \pm \Delta \sqrt{\Delta^2 + 4T_e^2 + 4\Omega_{a}^2})}}.
\]  

(32)

Thus, in the dressed-state description, the strong pumping splits the two SQD energy levels, creating two triplets, as presented in Figure 3. The energy gap between the initial levels and the new ones is equal to \( \Delta/2 \pm \sqrt{\Delta^2 + 4\Omega_{a}^2 + 4T_e^2}/2 \). A schematic representation of the transitions taking place between the dressed-state energy levels is presented in Figure 3a, for a positive value of the detuning of the pump field (\( \Delta = \omega_{12} > 0 \)). These transitions are associated with the following values of the pump-probe field energy mismatch: \( \lambda_{+} = |\lambda_{-}|, \lambda_{+}, |\lambda_{-}|, |\lambda_{-}| - |\lambda_{-}|, -|\lambda_{-}|, -\lambda_{+}, -\lambda_{-} - |\lambda_{-}| \). In the case of a pump field that is found at exact resonance with the \( |0\rangle \leftrightarrow |1\rangle \) transition (\( \Delta = \omega_{12} = 0 \)), the dressed states are equally separated from each other, which is the case depicted in Figure 3b. Here, the energy of the \( |\rangle \leftrightarrow |\rangle \) (\( |z\rangle \leftrightarrow |\rangle \)) transition is equal to the energy of the \( |\rangle \leftrightarrow |\rangle \) (\( |\rangle \leftrightarrow |\rangle \)) tran-
sition, and hence, a doublet of resonances, arisen around a central frequency, is responsible for the two sidebands of the FWM spectra.

\[ \lambda_{\pm} = \Delta \pm \Delta + \Omega + \Omega \]

\[ |+\rangle \]
\[ |e\rangle \]
\[ |\rangle \]
\[ |\rangle \]
\[ |\rangle \]
\[ |\rangle \]
\[ |\rangle \]

(a) \hspace{2cm} (b)

**Figure 3.** A dressed-state representation indicating the possible excitation pathways, in the case with \( \omega_{12} = \Delta \). (a) refers to the off-resonance case, with a positive pump-field detuning \( (\Delta > 0) \), while (b) refers to the exact-resonance case \( (\Delta = 0) \).

3. Results

Here, we investigate the characteristics associated with the absorption and the dispersion spectra of a probe field applied to an SQD-MNP hybrid structure, under the interaction with a strong pump field, for various interparticle distances. In the following paragraphs, we consider a linearly polarized strong pump field. Its polarization is taken parallel to the interparticle axis \( (s_a = 2) \), while its Rabi frequency \( \hbar\Omega_b \) is set equal to 3 meV. The weak probe field Rabi frequency is considered to be 100 times lower than the pump field Rabi frequency. The SQD dielectric constant is \( \varepsilon_s = 6\varepsilon_0 \), while for the modeling of the MNP we take the experimental values of the dielectric function referring to gold [57]. The radius \( a \) of the MNP is 7.5 nm. The hybrid structure is considered to be surrounded by vacuum and hence, \( \varepsilon_{env} = \varepsilon_0 \). For the SQD, we consider values typical for InAs/GaAs SQDs that have been used in previous studies [38,42,46]. As far as the dephasing and decay rates related to the double-SQD molecule are concerned, we respectively have \( \hbar\gamma_{01} = 0.22 \) meV, \( \hbar\gamma_{12} = 0.1 \) meV, \( \hbar\gamma_{02} = 0.001 \) meV, and \( \hbar\Gamma_{10} = 0.025 \) meV, \( \hbar\Gamma_{20} = 0.000025 \) meV, and \( \Gamma_{12} = 0 \). Finally, the energy gap associated with the \( |1\rangle \leftrightarrow |2\rangle \) transition is \( \hbar\omega_{12} = 1.4 \) eV and the dipole moment element \( \mu \) of the \( |0\rangle \leftrightarrow |1\rangle \) transition is equal to 0.65 · e nm.

More specifically, in Figures 4–10, we present the spectra of the asymmetric double SQD molecule susceptibility, \( \chi_{SQD}^{(1)} \), the MNP susceptibility, \( \chi_{MNP}^{(1)} \), and the total susceptibility of the hybrid structure, \( \chi_{tot}^{(1)} \), as a function of the pump-probe field detuning \( \delta \), for several values of the distance \( R \) that separates the SQD from the MNP \( (R = 100 \) nm: blue solid curve, 20 nm: red dashed-dotted curve, and 15 nm: green dashed-dotted curve). In Figure 4, the dispersion and the absorption spectra of the probe field are presented for \( \Delta = \omega_{12} = 0 \), in the case of a quite low value of the tunneling rate \( \hbar T_e = 0.01 \) meV, as seen in Figure 4a,c,e and b,d,f, respectively. First, we observe that, at exact resonance, the absorption/dispersion associated with the double SQD molecule exhibits a symmetric/antisymmetric profile for every value of the tunneling rate coefficient. In Figure 4, since the tunneling rate tends to zero, the spectra of the SQD optical susceptibility, as seen in Figure 4a,b, are identical to the ones plotted for a single SQD–MNP hybrid [36]. As the value of \( R \) decreases, the sideband resonances are transposed away from the spectral center. For the set of parameters used in Figures 4–6 \( (\Delta = \omega_{12} = 0) \), the MNP contribution outweighs the contribution of the double SQD molecule, introducing a slope to the spectra of the total susceptibility due to the presence of the first term in the expression of the MNP susceptibility, which is in accordance with the mathematical expression of Equation (28).
Figure 4. The dispersion (a,c,e) and the absorption (b,d,f) spectra for $\hbar \Omega_d = 3$ meV, $\omega_{12} = \Delta = 0$ and $\hbar T_e = 0.01$ meV, for various interparticle distances ($R = 100$ nm (blue solid curve), 20 nm (red dashed curve), and 15 nm (green dashed-dotted curve)). The rest parameters used are $\epsilon = 7.5$ nm, $\epsilon_5 = 6 \epsilon_0$, $\epsilon_{env} = \epsilon_0$, $\hbar \omega_{01} = 1.4$ eV, $\tau_a = 2$, $\hbar \Gamma_{10} = 0.025$ meV, $\hbar T_{20} = 0.000025$ meV, $\Gamma_{12} = 0$, $\hbar \gamma_{01} = 0.22$ meV, $\hbar \gamma_{12} = 0.1$ meV, $\hbar \gamma_{02} = 0.001$ meV, and $\hbar T_e = 0.01$ meV.

Figure 5. The dispersion (a,c,e) and the absorption (b,d,f) spectra for the same parameters as in Figure 4, but with $\hbar T_e = 0.5$ meV. Also, $R = 100$ nm (blue solid curve), 20 nm (red dashed curve), and 15 nm (green dashed-dotted curve).

The susceptibility spectra presented in Figure 5 refer to the case of the non-negligible electron-tunneling rate $\hbar T_e = 0.5$ meV. Here, we observe a set of inner resonances arising around the positions $\pm \hbar \sqrt{\Omega_d^2 + T_e^2}$, as well as a set of outer resonances, placed at $\pm 2\hbar \sqrt{\Omega_d^2 + T_e^2}$, which are hardly noticeable due to their low amplitude. Moreover, the transposition of the dominant inner resonances, owing its presence to the alteration of the value of $R$, is of low importance as compared to the transposition of the resonances observed in the case with $\hbar T_e = 0.01$ meV (Figure 4). However, the amplitude of the resonances is slightly decreased as the distance between the particles is increased. Finally, and most importantly, as we note by comparing Figure 4 to Figure 5, when the tunneling...
rate coefficient increases, the double SQD molecule susceptibility seems to have an even more important contribution to the spectral profile of the total susceptibility $\chi_{tot}^{(1)}$.

In Figure 6, we investigate the optical response of the two counterparts, as well as the total optical response of the system, for a high electron-tunneling rate ($hT_e = 5$ meV). Here, as the interparticle distance decreases, we note an infinitesimal transposition of the resonances, while their intensity is slightly increased, exhibiting an inverse trend with respect to the one observed in Figure 5, for $hT_e = 0.5$ meV. In addition, the distance between the inner resonances is doubled. Furthermore, the double SQD molecule contribution almost entirely defines the spectral profile of $\text{Im}[\chi_{tot}^{(1)}]$, as shown in Figure 6f, while the MNP mainly adds a shift to the spectral profile due to the contribution of the double SQD molecule, both co-shaping the profile of the Re$[\chi_{tot}^{(1)}]$ spectrum (Figure 6e).

In the next three figures, we study the off-resonant case, with $\hbar\Delta = -1$ meV, for various values of the tunneling rate coefficient (Figure 7: $hT_e = 0.01$ meV, Figure 8: $hT_e = 0.5$ meV, and Figure 9: $hT_e = 5$ meV), where we take $\hbar\omega_{12} = 1$ meV. After carefully studying all the possible cases, it becomes evident that, for $\Delta \neq 0$, the spectral profiles lose their symmetry. By comparing Figure 7a,b with the corresponding Figure 4 (for the same electron-tunneling rate, yet with $\Delta = \omega_{12} = 0$), we observe that, for $hT_e = 0.01$ meV, the general profile of the dispersion spectrum of the one figure roughly resembles the profile of the absorption spectrum of the other figure. The rest of the trends that are followed are similar to the ones presented in Figure 4, the most important of them being the significant transposition of the resonances when the center-to-center distance is modified.

In the case of an average electron-tunneling rate ($hT_e = 0.5$ meV), for $\hbar\Delta = -\hbar\omega_{12} = -1$ meV, which is presented in Figure 8, we note that the typical six-resonance pattern arises on the spectra of the SQDs’ susceptibility, as seen in Figure 8a,b. These patterns usually appear in three-level-system spectra, unless we are in a dark state with exactly resonant transitions or at two-photon resonance. Furthermore, the profile of the total susceptibility is ascribed both to the contributions of the coupled SQDs, as well to the MNP, as seen in Figure 8c,d. It becomes apparent that the contribution of MNP is responsible for the characteristic shift observed in $\chi_{tot}^{(1)}$.

Figure 6. The dispersion (a,c,e) and the absorption (b,d,f) spectra for the same parameters as in Figure 4, but with $hT_e = 5$ meV. Also, $R = 100$ nm (blue solid curve), 20 nm (red dashed curve), and 15 nm (green dashed-dotted curve).
Figure 7. The dispersion (a, c, e) and the absorption (b, d, f) spectra for the same parameters as in Figure 4, in the off-resonance case with $h\Delta = -1$ meV, $h\omega_{12} = 1$ meV, and $hT_e = 0.01$ meV. Also, $R = 100$ nm (blue solid curve), 20 nm (red dashed-curve), and 15 nm (green dashed-dotted curve).

Figure 8. The dispersion (a, c, e) and the absorption (b, d, f) spectra for the same parameters as in Figure 7, but with $hT_e = 0.5$ meV. Also, $R = 100$ nm (blue solid curve), 20 nm (red dashed curve), and 15 nm (green dashed-dotted curve).

However, if we further increase the tunneling-rate coefficient ($hT_e = 5$ meV), as seen in Figure 9, two inner resonances and two outer resonances arise again, respectively emerging at the positions $\pm h\sqrt{\Omega^2_a + \bar{T}^2_e}$ and $\pm 2h\sqrt{\Omega^2_a + \bar{T}^2_e}$, the outer being negligible regarding its magnitude. Except for the loss of symmetry, which is detected in the spectral response, for this specific set of parameters, the spectra present additional similarities to the ones presented in Figure 6 for the same values of the energy gap $h\omega_{12}$ and tunneling rate $T_e$, yet with a zero pump field detuning $\Delta$. It is important to note that, even for a comparatively high tunneling rate, the position of the inner resonance arising for $\delta > 0$ moves towards higher values of the detuning parameter as we decrease the interparticle distance, in which case the two counterparts of the hybrid structure interact more strongly...
with each other. This interesting feature was not observed in the spectra of the susceptibility at exact-resonance for the same value of the electron-tunneling rate (Figure 6).

Finally, in Figure 10, where we consider exactly the same values for the other parameters as in Figure 7, except for $h\Delta = 1$ meV, we observe that the change of the detuning sign is responsible for the inversion of symmetry with respect to the origin $(0,0)$, as far as the spectra of the double SQD molecule susceptibility are concerned (Figure 10a,b). Adding an extra shift, due to the MNP contribution (Figure 10c,d), we finally take the spectra of the total susceptibility (Figure 10e,f).

**Figure 9.** The dispersion (a,c,e) and the absorption (b,d,f) spectra for the same parameters as in Figure 7, but with $hT_e = 5$ meV. Also, $R = 100$ nm (blue solid curve), 20 nm (red dashed curve), and 15 nm (green dashed-dotted curve).

**Figure 10.** The dispersion (a,c,e) and the absorption (b,d,f) spectra for the same parameters as in Figure 4, but with $h\omega_{12} = h\Delta = 1$ meV and $hT_e = 0.01$ meV. Also, $R = 100$ nm (blue solid curve), 20 nm (red dashed curve), and 15 nm (green dashed-dotted curve).
4. Conclusions

In summary, we perform calculations in order to investigate the optical response of a strongly pumped hybrid structure made of an SQD molecule and a spherical MNP. Starting with the Hamiltonian, the density matrix equations are derived and the first-order optical susceptibility of the weak probe field is calculated, for the total system, as well as for its two counterparts, respectively. We find that, in the exact-resonance pump field regime, the spectra of the real and the imaginary part of the optical susceptibility are four-peaked and exhibit a symmetric profile, while, in the off-resonance regime, the spectra exhibit six resonances and are not symmetric for average tunneling rates. For high interparticle distances, analytical expressions for the position of the resonances are provided, based on the dressed states analysis, which is valid within a specific range of values of the Rabi frequency. For low tunneling rates, the contribution of MNP outweighs the contribution associated with the asymmetric double SQD molecule, introducing a non-negligible slope to the spectra of the total susceptibility. However, for a stronger tunneling rate, the contribution of the SQD molecule seems to play an even more important role to the spectral profile of the total susceptibility, determining to a substantial extent the shape of the spectrum of its imaginary part. In every case studied, as the distance between the counterparts is decreased, the resonances are transposed away from the center of the spectra.

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