Plasmon effects in the two-photon Rabi oscillations in a strongly coupled semiconductor quantum dot–metal nanoshell composite

B S Nugroho¹ and Y Arman¹

¹Physics Department, Faculty of Mathematics and Natural Sciences, Universitas Tanjungpura, Jalan Prof. Dr. Hadari Nawawi, 78115 Pontianak, Indonesia
E-mail: b.s.nugroho@physics.untan.ac.id

Abstract. We theoretically study two-photon Rabi oscillations (TPRO) in a nanocomposite comprising a semiconductor quantum dot (SQD) strongly coupled to a metallic nanoshell (MNS), which undergoes a pulsed excitation. The SQD is modeled as a three-level ladder-like quantum system (ground, one-exciton, and bi-exciton states). Its optical dynamics is described quantum-mechanically within the density matrix formalism framework, while the MNS is treated classically through its frequency-dependent polarizability. We find two effects of the presence of a MNS in close proximity to the SQD: (a) - a sufficient increase of the number of Rabi cycles as compared to the case of an isolated SQD and (b) - incoherent population of the one-exciton state giving rise to the destruction of the TPRO.

1. Introduction

TPRO is a nonlinear process that can be potentially exploited to realize a single-emitter source of pairs of entangled (with different polarizations) photons. It is of great interest for applications in quantum computing and quantum information processing [1, 2, 3, 4]. TPRO reflects a coherent superposition of two eigenstates during the system’s interaction with an intense, resonant excitation field. In a single semiconductor quantum dot (SQD), in which the multie exciton states naturally arise, this phenomenon may occur involving the ground-to-biexciton transition. The intermediate one-exciton state plays the role of a virtual state, which is unpopulated so that two photons generated during one Rabi cycle are utterly entangled.

In a previous paper [5], the TPRO of a SQD strongly coupled to a metal nanoparticle (MNP) has been theoretically studied. The optical excitations in the SQD are excitons (electron-hole pairs coupled by the attractive Coulomb interaction), while those in a MNP are localized surface plasmons (confined free electrons oscillations). It has been shown that the exciton-plasmon coupling might enhance the TPRO that leading to a significant increase in the number of entangled photon pairs per pulse. In that study, we have considered the limit where the coupling parameter’s magnitude is much smaller than the biexciton shift of the SQD.

In this communication, we examine TPRO in another limit, namely when the SQD-MNP interaction is in the order of the biexciton shift. Additionally, we complement the previous study [5] by considering not only radiative damping in the SQD but also nonradiative damping from dephasing processes. We model the SQD as a cascade three-level system [3, 6, 7, 5]. For the MNP, we use a metallic nanoshell (MNS) comprising a spherical dielectric core coated by a
metal layer [8]. This report shows that the effects of strong plasmon-exciton coupling are crucial for the nonlinear dynamics of the SQD-MNS composite.

2. Theoretical Model
We consider a nanocomposite comprising a SQD in the vicinity of a spherical MNS. The two nanoparticles are separated by a center-to-center distance \( d \) and embedded in a matrix with permittivity \( \varepsilon_d \). The MNS consists of a core of radius \( r_1 \) with the dielectric constant \( \varepsilon_1 \), coated with a metallic layer of thickness \( r_2 - r_1 \) with the dielectric function \( \varepsilon_2(\omega) \). The MNS is modelled classically and characterized by its frequency-dependent polarizability \( \alpha_1(\omega) \), which in the quasistatic limit reads

\[
\alpha_1(\omega) = 4\pi r_2^3 \frac{[\varepsilon_1 + 2\varepsilon_2(\omega)] [\varepsilon_2(\omega) - \varepsilon_b] + (r_1/r_2)^3 [\varepsilon_1 - \varepsilon_2(\omega)] [\varepsilon_b + 2\varepsilon_2(\omega)]}{[\varepsilon_2(\omega) + 2\varepsilon_b] [\varepsilon_1 + 2\varepsilon_2(\omega)] + 2 (r_1/r_2)^3 [\varepsilon_2(\omega) - \varepsilon_b] [\varepsilon_1 - \varepsilon_2(\omega)]},
\]

(1)

The SQD is considered as a three-level ladder-like quantum system with the ground \( |1\rangle \), one-exciton \( |2\rangle \) and biexciton \( |3\rangle \) states. The energies of these states are, accordingly, \( \mathcal{E}_1 = 0 \), \( \mathcal{E}_2 = \hbar \omega_2 \) and \( \mathcal{E}_1 = \mathcal{E}_2 = 2\hbar(\omega_2 - \Delta_b/2) \), where \( \Delta_b \) is the biexciton binding energy. The optical transitions, allowed by the selection rules, are \( |1\rangle \rightarrow |2\rangle \) and \( |2\rangle \rightarrow |3\rangle \) with the corresponding transition dipole moments \( \mu_{21}(\mu_{12}) \) and \( \mu_{32}(= \mu_{23}) \), respectively. The latter are assumed to be real and parallel to each other (\( \mu_{32} = \mu \mu_{21} \)) and aligned with the applied field as well. Thus, all vector quantities can be treated as scalars. Within this model, the biexciton state is dipole forbidden and can only be attained either via consecutive transitions \( |1\rangle \rightarrow |2\rangle \rightarrow |3\rangle \) or by the direct process \( |1\rangle \rightarrow |3\rangle \), absorbing simultaneously two photons, .

It is assumed that the system is excited by a pulsed external field \( \mathcal{E} = E_0(t) \cos(\omega_0 t) \), having a slowly varying amplitude \( E_0(t) \) and a carrier frequency \( \omega_0 \) which is close to frequencies of the SQD allowed transitions. We describe the optical dynamics of the SQD by means of the Lindblad master equation for the density operator \( \rho(t) \). In the reference frame, rotating with the frequency \( \omega_0 \) of the external field, this equation reads [6]

\[
\dot{\rho}(t) = -\frac{i}{\hbar} \left[ H_{\text{RWA}}(t), \rho(t) \right] + \mathcal{L}_\gamma[\rho(t)] + \mathcal{L}_\Gamma[\rho(t)],
\]

(2)

where

\[
H_{\text{RWA}}(t) = \hbar (\Delta_{21}\sigma_{22} + \Delta_{31}\sigma_{33}) - \hbar [\Omega(t)\sigma_{21} + \mu \Omega(t)\sigma_{32} + \text{H.c.}],
\]

(3)

\[
\mathcal{L}_\gamma[\rho(t)] = \frac{\gamma_{21}}{2} [\sigma_{12}\rho(t), \sigma_{21}] + [\sigma_{12}, \rho(t)\sigma_{21}], + \frac{\gamma_{32}}{2} [\sigma_{23}\rho(t), \sigma_{32}] + [\sigma_{23}, \rho(t)\sigma_{32}],
\]

(4)

\[
\mathcal{L}_\Gamma[\rho(t)] = \Gamma_2 \left( [\sigma_{22}\rho(t), \sigma_{22}] + [\sigma_{22}, \rho(t)\sigma_{22}] \right) + \Gamma_3 \left( [\sigma_{33}\rho(t), \sigma_{33}] + [\sigma_{33}, \rho(t)\sigma_{33}] \right),
\]

(5)

and

\[
\Omega = \frac{1}{\varepsilon_s} \left[ 1 + \frac{\alpha_1(\omega_0)}{2\pi d^3} \right] \Omega_0 + G (\rho_{21} + \rho_{32}),
\]

(6)

Here, \( H_{\text{RWA}}(t) \) is the Hamiltonian of the SQD in the rotating wave approximation (RWA) and the square brackets denote the commutator. \( \mathcal{L}_\gamma[\rho(t)] \) and \( \mathcal{L}_\Gamma[\rho(t)] \) are the Lindblad relaxation operators, with \( \sigma_{ij} = |i\rangle \langle j| \) with \( i,j = 1,2,3 \). \( \mathcal{L}_\gamma[\rho(t)] \) describes the radiation relaxation of the SQD states \( |2\rangle \) and \( |3\rangle \) with rates \( \gamma_{21} \) and \( \gamma_{32} \), while \( \mathcal{L}_\Gamma[\rho(t)] \) accounts for the dephasing of these states with rates \( \Gamma_{21} \) and \( \Gamma_{32} \), respectively. In Eq 3, \( \Delta_{21} = \omega_2 - \omega_0 \) and \( \Delta_{31} = \omega_3 - 2\omega_0 \) are the detunings away from the one-photon and coherent two-photon resonance. In Eq 6, \( \Omega_0 = \mu E_0/\hbar \) is the Rabi amplitude of the external field. \( G = G_R + iG_I \) is the complex-valued
feedback parameter describing the self-action of the SQD via the MNS. Accounting for multipolar contribution [9], the latter can be expressed as [6]

\[ G = \frac{\mu^2}{16\pi^3\hbar\varepsilon_0\varepsilon'_s} \sum_n \frac{n(n+1)(n+1)^2 \alpha_n(\omega_0)}{d^{2n+4}}. \]  

(7)

where \( \varepsilon'_s = (\varepsilon_s + 2\varepsilon_b)/(3\varepsilon_b) \) is the effective dielectric constant of the SQD and \( \alpha_n \) is the MNS multipolar polarizability of order \( n \), given by [10]

\[ \alpha_n(\omega) = 4\pi i^{2n+1} \left[ \varepsilon_1 + \frac{n+1}{n} \varepsilon_2(\omega) \right] [\varepsilon_2(\omega) - \varepsilon_b]^{2n+1} + \left( \frac{r_1}{r_2} \right)^{2n+1} [\varepsilon_1 - \varepsilon_2(\omega)] [\varepsilon_b + \frac{n+1}{n} \varepsilon_2(\omega)]^{2n+1} + \left( \frac{r_1}{r_2} \right)^{2n+1} [\varepsilon_2(\omega) - \varepsilon_b] [\varepsilon_1 - \varepsilon_2(\omega)]. \]  

(8)

In studying the TPRO, we consider the case when the external field is tuned exactly into the two-photon resonance \( \omega_0 = \omega_3/2 \) and choose the Gaussian shape of the external field envelope

\[ \Omega_0(t) = \frac{A}{\sqrt{\pi t_0}} e^{-\left(\frac{t-t_0}{t_0}\right)^2}. \]  

(9)

Here, \( A = \int_{-\infty}^{\infty} \Omega_0(t) dt \) is the pulse area, \( t_d \) is the time for the pulse to attain its maximum, and \( t_0 \) is the parameter determining the pulse duration.

### 3. Result and Discussion

For numerical calculations, we take a set of parameters typical for an InGaAs/GaAs quantum dot [5]: \( \varepsilon_2 = 1.34 \text{ eV}, \varepsilon_3 = 2\varepsilon_2 - h\Delta_B \) with \( h\Delta_B = 2.75 \text{ meV}, \gamma_{21} = \mu^2\gamma_{21} = \mu^2\gamma \) with \( h\gamma_{21} = 1.13 \text{ meV} \) and \( h\gamma_{32} = 0.91 \text{ meV} \). Accordingly, \( \mu = \sqrt{\gamma_{32}/\gamma_{21}} \approx 0.90 \). The dephasing rate values are taken as \( \Gamma_2 = \Gamma_3 = \gamma \). We take \( \varepsilon_s = 6 \). For the MNS, we consider silica-Ag core-shell, embedded in a silica host \( (\varepsilon_b = 2.16) \), with inner and outer radii \( r_1 = 9.00 \text{ nm} \) and \( r_2 = 9.79 \text{ nm} \). The dielectric function of silver \( \varepsilon_2(\omega) \) is taken from tabulated data [11]. Using these data and applying Eq. 1, the corresponding localized surface plasmon resonance is found to be around \( \omega_{LSP} = 1.40 \text{ eV} \), i.e., close to the two-photon resonance.

We solved Eq. 2 numerically and plot the time evolution of the SQD populations when the composite is excited by a short pulse \( t_0 = 10/\Delta_B, A = 8\pi \) in Fig. 1. Here, the pulse

![Figure 1](image-url)  

**Figure 1.** Time evolution of the SQD level populations calculated for an incident pulse of area \( A = 8\pi \), duration \( t_0 = 10/\Delta_B \) ps, and delay time \( t_d = 7 \) ps for two case: (a) isolated SQD and (b) SQD-MNS composite. The red-solid line shows the profile of the incident Gaussian pulse scaled to the figure.
spectrum, having a width approximated by $1/(2t_0)$, is considerably less (10 times narrower) than the detuning away from the one-exciton resonance, $\Delta_B/2$. In this condition, we are well inside the adiabatic limit [12, 5], where the transition to $|2\rangle$ is almost forbidden by energy conservation. In the case of isolated SQD [Fig 1(a)], it explains the absence of $\rho_{22}$ after the pulse has been switched off. Nevertheless, the maximum value of the applied pulse’s Rabi amplitude is $A/(\sqrt{\pi}t_0) = 1.42\Delta_B$, i.e., more extensive than $\Delta_B/2$, leading to noticeably elevated $\rho_{22}$ during the pulse action. Additionally, the effects of $|3\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |1\rangle$ relaxations leading to the incoherent population of $|2\rangle$ are insignificant because the pulse duration is much smaller than the radiative decay times ($t_0 = 10/\Delta_B = 2.39$ ps $\ll \gamma_{21}^{-1} = 582.49$ ps, $\gamma_{32}^{-1} = 723.31$ ps).

It is interesting to look at Fig. 1(b), when the SQD is coupled to the MNP with centre-to-center difference $d = 16$ nm. For the parameters chosen, we have $G = (1.19 + 0.04i)$ meV. As is seen, compared to the isolated SQD [Fig. 1(a)] the number of Rabi cycle is pronouncedly larger. We relate this to the enhancement effects (the external field enhanced by the induced field from plasmon excitation) felt by the SQD due to the presence of MNP. This enhancement effect can be seen qualitatively from the first term of the right-hand side of Eq. 6. Another peculiar feature reveals from the figure is $\rho_{22}$ slightly elevates after the pulse has passed, contrary to the previous case [Fig. 1(b)]. To address this issue, we recall that one of the essential effects of the SQD self-action via the MNS is renormalization of the SQD transition frequencies [6], namely $\Delta_{21} \rightarrow \Delta_{21} + G_R Z_{21}$ and $\Delta_{32} \rightarrow \Delta_{32} + 2G_R Z_{32}$, where $Z_{21} = \rho_{22} - \rho_{11}$ and $Z_{32} = \rho_{33} - \rho_{22}$. Because $G_R = 1.19$ meV is comparable with $\Delta_B/2 = 1.38$ meV, the shift of $\Delta_{21}$ brings the frequency of one-exciton transition approaching the applied field frequency leads to the excitation of state $|2\rangle$ during the pulse action and afterwards. The jiggling behaviour of the populations after the pulse action comes from a complicated interplay of dependency between the shift of transition resonances ($\Delta_{21}$ and $\Delta_{32}$) and the population differences ($Z_{21}$ and $Z_{32}$).

In Fig. 2, we present the area dependence of $\rho_{22}$ and $\rho_{22}$ for the system. Here, the parameters used as in Fig. 1. For the case of isolated SQD [Fig. 2(a)], the system clearly shows TPOR. We attribute the marginal occupation of one-exciton state $\rho_{22}$ to the direct excitation of this state due finite width of $\gamma_{21}$ as well as relaxation of $\rho_{22}$. The characteristic of the area dependence of $\rho_{22}$ and $\rho_{22}$ for SQD-MNP hybrid [Fig. 1(b)] differ significantly with the isolated SQD. The origin of this difference comes from the field enhancement (first term of the right-hand side of Eq. 6). It is also seen that there is a build-up of the one-exciton population, followed by decreasing in the two-exciton population due to the feedback effects. It implies that for the case of $G$ comparable to $\Delta_B$, the strong interaction of SQD-MNP composite may introduce a
destructive effect on the coherence of TPRO.

4. Conclusion
We have conducted a theoretical study of the TPRO of a SQD-MNS composite subject to a quasi-resonant pulsed excitation. A limit has been examined when the SQD-MNS interaction is on the order of the biexciton energy shift. Additionally to the result found previously, that is increasing the number of Rabi cycles per pulse due to the SQD-MNS interaction (positive effect) [5], we have found that this interaction may also give rise to destruction of the TPRO resulted from the incoherent population quenching of the one-exciton state (negative effect).

References
[1] Müller M, Bounouar S, Jöns K D, Glässl M and Michler P 2014 Nat. Photonics 8 224–228
[2] Gazzano O and Solomon G S 2016 J. Opt. Soc. Am. B 33 C160–C175
[3] Kaldewey T, Lüker S, Kuhlmann A V, Valentin S R, Ludwig A,Wieck A D, Reiter D E, Kuhn T and Warburton R J 2017 Physical Review B 95 161302
[4] Flamini F, Spagnolo N and Sciarrino F 2018 Rep. Prog. Phys 82 016001
[5] Nugroho B S, Iskandar A A, Malyshev V A and Knoester J 2019 Phys. Rev. B 99 075302
[6] Nugroho B S, Iskandar A A, Malyshev V A and Knoester J 2020 Phys. Rev. B 102(4) 045405
[7] Nugroho B S, Iskandar A A, Malyshev V A and Knoester J 2016 J. Opt. 19 015004
[8] Maier S A 2007 Plasmonics: fundamentals and applications (Springer Science & Business Media)
[9] Yan J Y, Zhang W, Duan S, Zhao X G and Govorov A O 2008 Phys. Rev. B 77 165301
[10] Naeimi Z, Mohammadianeh A and Miri M 2019 J. Opt. Soc. Am. B 36 2317–2324
[11] Babar S and Weaver J 2015 Appl. Opt. 54 477–481
[12] Stufler S, Machnikowski P, Ester P, Bichler M, Axt V M, Kuhn T and Zrenner A 2006 Phys. Rev. B 73 125304