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P h y s i c s

SPECIFICATIONS OF RABI OSCILLATIONS IN THE QUANTUM EMITTERS SYSTEMS COUPLED TO THE LOCALIZED PLASMON POLARITONS

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In this work phenomenon of resonant transfer of the excited energy between quantum dipole emitters (QDE) by the localized plasmon polaritons (LPP) is investigated. In system the molecules or semiconductor quantum dots act as QDEs and the nanometer size metal particles (MNP) care LPPs. The dependence of the frequency of Rabi oscillations on system parameters (the distance between MNP and QDE, the radius of MNP and dielectric permittivity of the surrounding medium) was determined. Conditions when the period of Rabi oscillations is considerably shorter than system relaxation time were defined.

***Keywords*:** quantum dipole emitter, quantum dot, metalnanoparticle, localized plasmon polaritons.

Introduction. The resonantly coupling between quantum dipole emitters (QDE) such as molecules or quantum dots and localized surface plasmons (LSP) of metal nanoparticle (MNP) at optical frequencies allows to control the flow of electromagnetic energy and lies in the core of an explosive growth in the field of quantum plasmonics [1]. Recent advances in nano-optics, especially experiments with single molecules interacting with well-defined metal nanostructures [2–4], often referred to as nano-antennas, serve as a strong impetus for further developments in this direction [5, 6]. The most often discussed effect of QDE–MNP interaction is concerned with the modification (enhancement or quenching) of fluorescence yield determined by the balance between radiative and non-radiative decay rates, both enhanced near MNPs [3, 4, 7–9]. It is also expected that the QDE–MNP interaction can even enter the regime of strong coupling, where excitation energy is coherently transferred between QDE and MNP in the form of Rabi oscillations [10]. In [11] the entanglement between two QDE resonantly coupled to LSP of MNP are investigated. The formation time of a superposition state is much less, than spontaneous decay time of QDE, and determined by the efficiency of the resonant interaction between

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QDE and induced LSP. In this paper the conditions for the formation of the Rabi oscillations in resonantly interacting QDE systems through LSP with one another are described.

Materials and Methods. Let QDEs are located on the same axis with the center of a metal sphere at distances from the center of the sphere R_{10} and R_{01} as in Fig. 1 (a) is shown. Let the first QDE be in an excited state with energy E_1, at initial time and the second is in the ground state with energy E_0. It is assumed that the spherical MNP exhibits a dipolar LSP resonance at the frequency ω_0 of the radiative (dipole-allowed) transition $1 \to 0$ (Fig. 2, b).

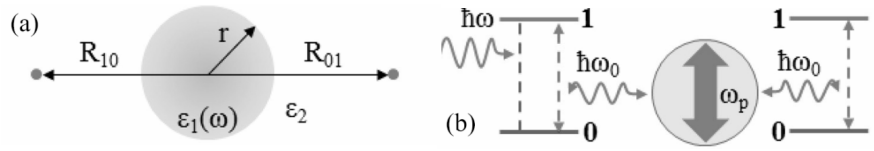


Fig. 1. Schematic of system with two QDEs placed near MNP (a) and relevant QDE energetic levels along with oscillating current associated with the LSP excitation (b).

The wave function of the system can be represented in general form

$$\Psi = a_{00}(t)\phi_{00}(\vec{r})\exp\left(-\frac{i}{\hbar}(E_0+E_0)t\right) + a_{10}(t)\phi_{10}(\vec{r})\exp\left(-\frac{i}{\hbar}(E_1+E_0)t\right) + a_{01}(t)\phi_{01}(\vec{r})\exp\left(-\frac{i}{\hbar}(E_1+E_0)t\right), \quad (1)$$

here $\phi_{00}(\vec{r}), \phi_{10}(\vec{r})$ and $\phi_{01}(\vec{r})$ are the wave functions of the system, when both QDEs are not excited, first is excited and second is in the ground state, and the first is in the ground state and the second is excited respectively, $a_{00}(t), a_{10}(t)$ and $a_{01}(t)$ corresponding probability amplitudes. Then the dipole moment of the first (second) QDE is given by the following:

$$\vec{D}_{10(01)} = a_{10(01)}a_{00}^{*}\vec{d}\exp\left[-i(\omega t - \phi_{10(01)})\right] + c.c., \quad (2)$$

where

$$\vec{d}\exp(i\phi_{10(01)}) = \int \phi_{10(01)}e\vec{r}\phi_{00}^{*}dV. \quad (3)$$

Let the vector $\vec{d}$ be directed along the axis, which connects the center MNP with QDEs, $\phi_{(10)}$ and $\phi_{(01)}$ are indefinite phases. In conditions when the wavelength of radiation λ is much larger and radius of the sphere r is much smaller than the other distances: $r \ll \min[R_{01}, R_{10}]$ and $\max[R_{01}, R_{10}] \ll \lambda$, the field on the sphere created by the dipole moments of QDEs is homogeneous and has the following form:

$$\vec{E}_{sph} = \frac{2}{4\pi\varepsilon_0\varepsilon_2}\left[\frac{\vec{D}_{10}}{R_{10}^3} + \frac{\vec{D}_{01}}{R_{01}^3}\right] = \vec{E}_0\exp(-i\omega t) + c.c., \quad (4)$$

here ε_0 and ε_2 are dielectric constants of vacuum and of environment medium. This field resonantly excites SPP mode localized on the sphere were QDE induces an electric field

$$\vec{E}_{10(01)} = \frac{2(\varepsilon_1-\varepsilon_2)r^3}{(\varepsilon_1+2\varepsilon_2)R_{10(01)}^3}\vec{E}_0\exp(-i\omega t) + c.c., \quad (5)$$

where $\varepsilon_1 = \varepsilon_1'\varepsilon_1''$ is the dielectric constant of the metal. Using the time-dependent Schrödinger equation for two-level systems in the driving field given by Eq. (5) and carrying out standard manipulations, following system of two coupled equations for the probability amplitudes would be obtained:

$$\dot{a}_{10} = \frac{i(\varepsilon_1-\varepsilon_2)|\vec{d}|^2 r^3}{\hbar\pi\varepsilon_0\varepsilon_2(\varepsilon_1+2\varepsilon_2)R_{10}^3}\left[\frac{|a_{00}|^2 a_{10}}{R_{10}^3}+\frac{|a_{00}|^2 a_{01}}{R_{01}^3}\exp(i(\phi_{01}-\phi_{10}))\right], \tag{6}$$

$$\dot{a}_{01} = \frac{i(\varepsilon_1-\varepsilon_2)|\vec{d}|^2 r^3}{\hbar\pi\varepsilon_0\varepsilon_2(\varepsilon_1+2\varepsilon_2)R_{10}^3}\left[\frac{|a_{00}|^2 a_{01}}{R_{01}^3}+\frac{|a_{00}|^2 a_{10}}{R_{10}^3}\exp(-i(\phi_{01}-\phi_{10}))\right], \tag{7}$$

$$\begin{aligned}\dot{a}_{00} = \frac{i(\varepsilon_1-\varepsilon_2)|\vec{d}|^2 r^3}{\hbar\pi\varepsilon_0\varepsilon_2(\varepsilon_1+2\varepsilon_2)R_{10}^3}&\left[\frac{a_{10}}{R_{10}^3}\exp(i\phi_{10})+\frac{a_{01}}{R_{01}^3}\exp(i\phi_{01})\right]\times\\ &\times\left[\frac{a_{10}}{R_{10}^3}\exp(-i\phi_{10})+\frac{a_{01}}{R_{01}^3}\exp(-i\phi_{01})\right].\end{aligned} \tag{8}$$

In conditions of resonance, i.e. $|\varepsilon_1'| \gg |\varepsilon_1'+2\varepsilon_2| \gg \varepsilon_1''(\varepsilon_1'<0)$, these equations can be written as follows:

$$\dot{b}_{10} = -iq[\beta_{10}b_{10}+\beta_{01}b_{01}]\beta_{10}|b_{00}|^2, \tag{9}$$

$$\dot{b}_{01} = -iq[\beta_{01}b_{01}+\beta_{10}b_{10}]\beta_{01}|b_{00}|^2, \tag{10}$$

$$\dot{b}_{00} = -iq[\beta_{10}b_{10}+\beta_{01}b_{01}]^2|b_{00}|, \tag{11}$$

where the following substitutions are done: $b_{10(01)} = a_{10(01)}\exp(i(\phi_{10(01)})$, $b_{00} = a_{00}$, $q = \dfrac{3|\vec{d}|^2 r^3}{\hbar\pi\varepsilon_0\Delta}\cdot\dfrac{R_{10}^6+R_{01}^6}{R_{10}^6R_{01}^6}$, $\Delta = \varepsilon_1+2\varepsilon_2$, $\beta_{10(01)} = \dfrac{R_{10(01)}^3}{(R_{10}^6+R_{01}^6)^{1/2}}$.

From Eqs.(9)–(11) we have

$$\dot{c}(t) = -iqc(t)|b_{00}|^2, \tag{12}$$

$$\dot{b}_{00} = -iq|c(t)|^2 b_{00}, \tag{13}$$

where $c(t) = \beta_{10}b_{10}+\beta_{01}b_{01}$.

Eqs. (12) and (13) have the following solutions:

$$c(t) = c_0\exp\left\{-iq|c_0|^2 t\right\}, \tag{14}$$

$$b_{00}(t) = c_0\exp\left\{-iq|c_0|^2 t+i\delta\right\}. \tag{15}$$

Here δ is an arbitrary phase. Inserting Eqs. (14) and (15) in Eqs. (9) and (10) respectively and considering that

$$|b_{00}(t)|^2+|b_{10}(t)|^2+|b_{01}(t)|^2 = 1;\ |b_{01}(0)|^2 = 0, \tag{16}$$

we obtain

$$b_{10}(t) = \beta_{10}c_0\left[\exp(-iq|c_0|^2 t)+\frac{\beta_{01}^2}{\beta_{10}^2}\right], \tag{17}$$

$$b_{01}(t) = \beta_{01}c_0\left[\exp(-iq|c_0|^2 t)-1\right], \tag{18}$$

where $c_0 = \beta_{10}/(1+\beta_{10}^2)^{1/2}$.

Thus, the possibilities to be in excited state the first and second atoms are determined by following equations respectively:

$$|b_{01}(t)|^2 = \frac{4\beta_{10}^2\beta_{01}^2}{1+\beta_{10}^2}\sin^2\Omega t, \quad |b_{10}|^2 = \frac{1}{1+\beta_{10}^2} - |b_{01}(t)|^2. \tag{19}$$

Here $\Omega = \dfrac{9\lambda_0^3 r^3}{16\pi^3(\varepsilon_2)^{1/2}\Delta}\cdot\dfrac{\Gamma(R_{10}^6+R_{01}^6)}{R_{01}^6(2R_{10}^6+R_{01}^6)}$, where $\Gamma = \dfrac{8\pi^2(\varepsilon_2)^{1/2}|\vec{d}|^2}{3\hbar\varepsilon_0\lambda_0^3}$.

Results. In the QDE–MNP–QDE system is realized the oscillation transfer of the excited energy from one to another QDE. The process of fluctuation of population for different R_{01}/R_{10} ratio is shown in Fig. 2.

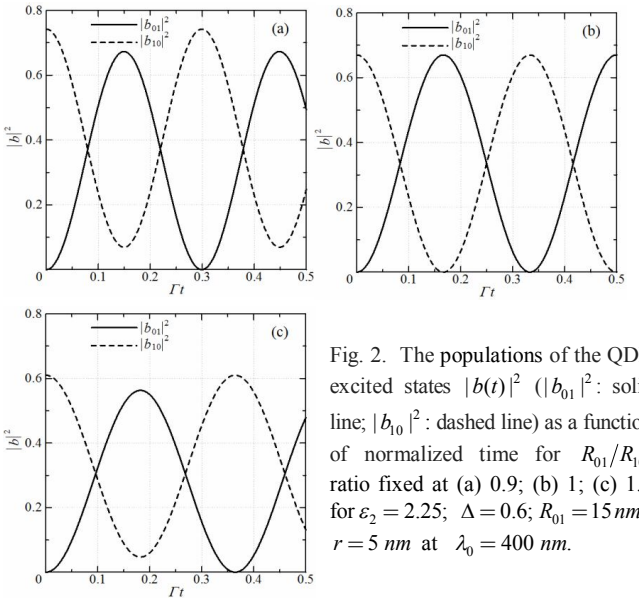
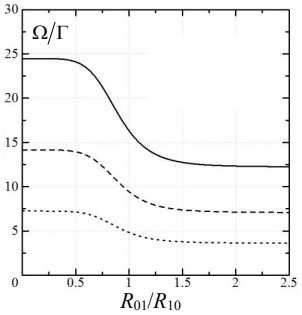


Fig. 2. The populations of the QDE excited states $|b(t)|^2$ ($|b_{01}|^2$: solid line; $|b_{10}|^2$: dashed line) as a function of normalized time for R_{01}/R_{10} ratio fixed at (a) 0.9; (b) 1; (c) 1.1 for $\varepsilon_2 = 2.25$; $\Delta = 0.6$; $R_{01} = 15nm$; $r = 5\ nm$ at $\lambda_0 = 400\ nm$.

Moreover, the process of repopulation is more completely for $R_{01} = R_{10}$ (see Fig. 2 (b)). Note that the period of energy pumping can be substantially shorter than the relaxation time of single QDE.



—— $r = 6\ nm$; - - - - $r = 5\ nm$; ····· $r = 4\ nm$

Fig. 3. Dependence of Rabi oscillations frequency on R_{01}/R_{10} ratio for MNP radius for $\varepsilon_2 = 2.25$; $\Delta = 0.6$; $R_{01} = 15nm$ at $\lambda_0 = 400\ nm$;

The frequency of Rabi oscillations decreases with increasing of R_{01}/R_{10} ratio, as well as with decreasing of MNP radius as is shown in Fig. 3.

Conclusion. In the paper was theoretically investigated the behavior of system of two molecules and MNPs in the existence of a strong connection between them. It was shown that there is a resonant transfer of excitation energy between molecules by the LPPs. The dependence of the frequency of Rabi oscillations on distance between MNP and QDE, on radius of MNP and on dielectric permittivity of the surrounding media are determined. The process can be observed in fluorescence spectroscopy through the splitting of emission peaks. Strong coupling is an important ingredient for future plasmonic-based quantum information schemes and might play a significant role in biosensor applications.

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