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Excitonic Rabi oscillations in a quantum dot: local field impact

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Abstract

The influence of local fields on the excitonic Rabi oscillations in an isolated, arbitrary shaped quantum dot (QD) has been theoretically investigated. QD interaction with both a classical electromagnetic field and quantum light has been considered. In the classical light, time harmonic and ultrashort pulse excitations are analyzed. The general formalism has been formulated for quantum light and applied to the case of a Fock qubit. Noticeable modification of the Rabi oscillation dynamics induced by the local fields is predicted to be observable in QDs exposed to both classical and quantum light. In particular, the bifurcation and anharmonism in the Rabi oscillations have been revealed under time harmonic excitation and a dependence of the Rabi oscillation period on the QD depolarization has been obtained.

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1. Introduction

Research on the properties of quantum dots (QDs)—nano-scale 3D confined narrow-gap insertions in a host semiconductor—has continued to grow unabated owing to the great potentiality for such structures in engineering applications, e.g. as the active media of double-heterostructure lasers [1,2]. Recently QDs have been proposed to serve as nodes of quantum networks that store and process quantum information [3,4]. Application of semiconductor QDs in cavity electrodynamics [5–7] and as potential quantum-light emitters [8,9] is now intensively discussed. Spontaneous emission in QDs [10] and electromagnetic fluctuations are also the focus of interest.

During the last decade, attempts have been made to establish the correspondence of and to reveal principal differences between two-level atoms and excitons in semiconductors. Many analogies have been successfully investigated, such as the excitonic optical Stark effect, photon echoes in four-wave mixing experiments, excitonic Rabi oscillations in quantum wells [11] and QDs [12–14]. A peculiarity which, among others, distinguishes the exciton from an atomic two-level system is that the exciton exists in a medium and interacts with the medium inducing its polarization (i.e., local fields). In the present paper, we investigate the role of local fields in excitonic Rabi oscillations in QDs.

Rabi oscillations are well established in a different physical systems such as cold trapped ions [15], Bose–Einstein condensates [16], semiconductor quantum wells [11]. Among these, excitonic Rabi oscillations in QDs are very promising: observed experimentally in [12–14], they correspond to one-qubit rotation which is a step towards QD application in quantum information processing [12]. Excitonic Rabi oscillations in a QD are to be distinguished from those of an ordinary atom by the following two factors: (i) the oscillator strength in a QD is essentially larger [12]; (ii) the Rabi oscillation picture strongly depends on the QD geometrical configuration. The first factor gives an opportunity to observe the Rabi oscillations in an essentially smaller field than for ordinary atoms. The second factor opens the possibility for the effect to be controlled.

In recent investigations, a significant role of electron–hole dipole–dipole interactions (local fields) in the electromagnetic response of different structures has been predicted [17–23]. General equations for the interaction between quantum states of light and condensed matter influenced by local fields have been formulated for bulk media in [23] and for confined QD excitons in [22]. However, application of these equations in the cited papers was restricted to the weak light–matter coupling regime. In our paper we aim at investigation of the local field impact on the light–QD interaction in the strong coupling regime.

2. Rabi oscillations in the QD interacting with a classical electromagnetic field

Let an isolated arbitrary shaped QD imbedded in a host semiconductor be exposed to a classical electromagnetic field. Further consideration is restricted to a two-level model which treats the QD as a spatially confined quantum oscillator. In the strong confinement regime, Coulomb interaction is assumed to be negligible for an exciton in a QD (which is considered as an electron–hole pair). Let the electron–hole pair's dipole moment be directed along the unit vector $\mathbf{e}_x$ in a Cartesian coordinate system related to the QD;

i.e., $\boldsymbol{\mu} = \mu \mathbf{e}_x$. The QD is exposed to the field $\mathbf{E}(t) = E_{0x}\mathbf{e}_x = \mathrm{Re}[\mathcal{E}(t)\exp(-\mathrm{i}\omega t)]\mathbf{e}_x$ linearly polarized along $\boldsymbol{\mu}$ ($\mathcal{E}(t)$ is the electric field amplitude, ω is the electromagnetic field carrier frequency). Accordingly to [22], in the two-level approximation the system "QD + electromagnetic field" is described by the Hamiltonian $H = (\epsilon_e a_e^\dagger a_e + \epsilon_g a_g^\dagger a_g) - V\widehat{P}_x E_{0x} + \Delta H$, where $\epsilon_{e,g}$ are the energy eigenvalues of excited and ground states, respectively; $a_{g,e}^\dagger$ and $a_{g,e}$ stand for the creation and annihilation operators of electrons in the ground and excited states; V is the QD volume; $\widehat{P}_x = V^{-1}(-\mu b^\dagger + \mu^* b)$ is the polarization operator; $b^\dagger = a_g a_e^\dagger$ and $b = a_g^\dagger a_e$ are the creation and annihilation operators for the electron–hole pair. The first term in the total Hamiltonian is the Hamiltonian of the free electron–hole pair, the second term is the Hamiltonian for the interaction of the pair with the electromagnetic field, while ΔH is the correction to the local fields. The latter is given by [22]

$$\Delta H = 4\pi N_x P_x(-\mu b^\dagger + \mu^* b), \tag{1}$$

where N_x is the depolarization coefficient of the QD, $P_x = \langle \widehat{P}_x \rangle$ is the macroscopic polarization of the QD. Under the rotating-wave approximation, the optical Bloch equations, as follows:

$$\frac{\partial u}{\partial t} = -\gamma_T u - \Omega_R w - \delta v - \Delta\omega(w+1)v, \tag{2}$$

$$\frac{\partial v}{\partial t} = -\gamma_T v + \delta u + \Delta\omega(w+1)u, \tag{3}$$

$$\frac{\partial w}{\partial t} = -\gamma_L(w+1) + \Omega_R u, \tag{4}$$

can be obtained from the foregoing Hamiltonian. The phenomenological parameters γ_T and γ_L are the dephasing and the homogeneous broadening, respectively. The parameter $\Delta\omega = 4\pi N_x|\mu|^2/\hbar V$ is the frequency shift resulting from the local field influence [19]. Detuning parameter $\delta = \omega - \omega_0 - \Delta\omega$ is corrected to the shift; ω_0 is the excitonic transition frequency. The parameter $\Omega_R(t) = \mu\mathcal{E}(t)/\hbar$ is the Rabi frequency, w is the inversion, i.e., the difference between the excitonic population in the excited and the ground states; u and v are the real and imaginary parts of the non-diagonal element of the density matrix. We assume that at $t = 0$ the QD is in the ground state. Then, the initial conditions for Eqs. (2)–(4) are given by

$$u(0) = v(0) = 0, \quad w(0) = -1. \tag{5}$$

Consider the case of an undamped system exposed to a monochromatic field in the exact synchronism regime. Note that the condition $\delta = 0$ is not the exact synchronism condition. Actually, accounting for the local field in the strong light–QD coupling regime leads to the substitution $\omega - \omega_0 \to \delta + \Delta\omega(w+1)$ Eqs. (2)–(4). Therefore, the actual physical detuning is not constant in time but oscillates together with the inversion. In the presence of local fields, a closed-form solution for the Bloch equations does not exist. Therefore, we have performed numerical integration of Eqs. (2)–(4) with initial conditions (5), assuming $\mathcal{E}(t) = \mathrm{const}$ and $\gamma_T = \gamma_L = 0$. Calculations of the inversion dynamics for different field amplitudes defined by the parameter $\xi = \Omega_R/\Delta\omega$ are shown in Fig. 1. Here, the inversion is plotted as a function of the dimensionless time $T = \Omega_R t/2\pi$. The calculations

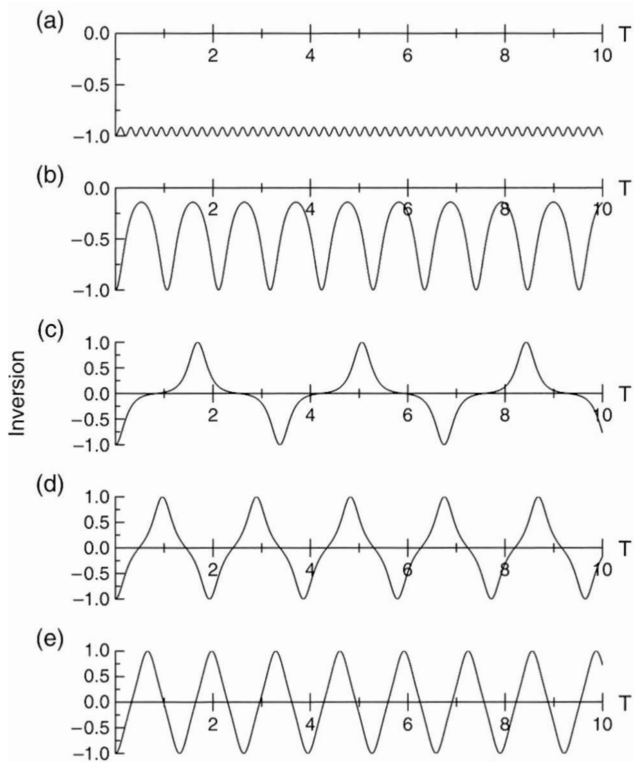

Fig. 1. Rabi oscillations of the inversion. $\delta = -\Delta\omega$; $\xi = 0.2$ (a); $\xi = 0.495$ (b); $\xi = 0.5001$ (c); $\xi = 0.51$ (d); $\xi = 0.6$ (e).

performed demonstrate that the Rabi frequency strongly depends on the depolarization parameter ξ. At $\xi \ll 1$ Rabi oscillations are practically absent: $w(t) \sim$ const, which corresponds to weak coupling between the QD and the electric field ($\Omega_R \to 0$). The increase in ξ leads to the appearance of the Rabi oscillations. The following Fig. 1 demonstrates the inversion calculations for finite values of the parameter δ. Thus, at $\xi = 0.2$, small amplitude oscillations of the inversion are observed (see Fig. 1(a)). At $\xi = \xi^{cr} = 0.5$ *bifurcation in the oscillation dynamics* is predicted (compare Fig. 1(b) and (c)), which separates two oscillatory regimes with drastically different characteristics. In the vicinity of the bifurcation, Rabi oscillations are *essentially anharmonic* (see Fig. 1(c), (d)). The anharmonism in the Rabi oscillations disappears with further increase in ξ; see Fig. 1(e).

When $\xi > 1$, the inversion behavior satisfies the approximation $w(t) \cong \cos(2\pi T)$, which describes the conventional picture of the Rabi oscillations [24]. In order to explain the bifurcation of the inversion, which results from the influence of nonlinear terms in Eqs. (2) and (3), we analyze the phase portrait of Eqs. (2)–(4). For $\gamma_T = \gamma_L = 0$ the system is conservative, and the integral of motion is given by $u^2 + v^2 + w^2 = 1$. Then using

Eqs. (2)–(4) allows us to obtain the expression

$$v(w) = \frac{1}{2\xi}(w+1)\left(w + 2\xi\frac{\delta}{\Omega_R} + 1\right), \tag{6}$$

which describes the phase portrait of the phenomenon. The period of Rabi oscillations (in units of T) is given by the integral

$$T_0 = \frac{1}{\pi}\int_{-1}^{w_0} \frac{\mathrm{d}w}{\sqrt{1 - w^2 - v^2(w)}}, \tag{7}$$

in which $v(w)$ is defined by Eq. (6) and w_0 is the root of the algebraic equation $v^2(w) + w^2 = 1$ closest to $w = -1$. For the case $\delta = -\Delta\omega$, from Eqs. (6) and (9) there follows the equation

$$T_0 = \frac{2}{\pi}\begin{cases} K(1/2\xi), & \xi > 0.5 \\ \xi K(2\xi), & \xi < 0.5 \end{cases} \tag{8}$$

where $K(\ldots)$ is the complete elliptic integral of the first kind. For the $\xi = \xi^{\mathrm{cr}} = 0.5$, one can see that $T_0 \to \infty$. At small values of ξ there are small period Rabi oscillations, while $T_0 \approx 1$ at large values of ξ ($\xi > 2$) (compare with previous results presented in Fig. 1). For a spherical GaAs QD with the radius $R_{\mathrm{QD}} \simeq 3$ nm, dielectric constant $\varepsilon_h = 12$, the estimate $\hbar\Delta\omega \simeq 1$ meV is obtained [22]. This corresponds to the value $\hbar\Omega_R \simeq 0.5$ meV for the case $\xi^{\mathrm{cr}} = 0.5$. Such a value is reachable in pump–probe and microcavity experiments [25].

Now, let us consider QD interaction with an optical pulse whose time duration τ is much less than the relaxation times in the system. An analysis for the Gaussian pulse $\mathcal{E} = \mathcal{E}_0 \exp[-(t - t_0)^2/\tau^2]$ is performed. Like in the previous section, the Rabi frequency is associated with the peak field strength of the Gaussian pulse: $\mathcal{E}_0 = \mu\Omega_{R0}/\hbar$, so that $\xi = \Omega_{R0}/\Delta\omega$. Fig. 2 shows the inversion dynamics for $\delta = -\Delta\omega$. When $\xi \geq 1$, the inversion demonstrates two different regimes. In the first regime (Fig. 2(a)), the inversion final state w_f is the excited state ($t \to \infty, w \to 1$). In the second regime (Fig. 2(b)), the inversion returns to the ground state ($t \to \infty$, $w \to -1$). When $\xi \leq 1$ (which corresponds to small Rabi frequencies) only the second regime is manifested. It should be emphasized that spontaneous radiation, which is not considered in our model, leads to the decay of the final state of inversion. However, the QD can stay in the excited state for a rather long time. Indeed, the spontaneous radiation lifetime of the spherical QD [26] can be associated, accordingly to Eq. (80) from Ref. [22], with the resonant frequency shift $\Delta\omega$ via

$$T_{sp} \simeq \frac{1}{4\tau\Delta\omega}\left(\frac{\lambda}{4\pi R_{\mathrm{QD}}\sqrt{\varepsilon_h}}\right)^3. \tag{9}$$

For the QD considered, exposure to a pulse with the parameter $\tau\Delta\omega = 15$ at the wavelength $\lambda = 1.3$ μm Eq. (9) gives $T_{sp} \sim 1.3 \times 10^2$. This result justifies the neglect in our model of the decay of the inversion final state.

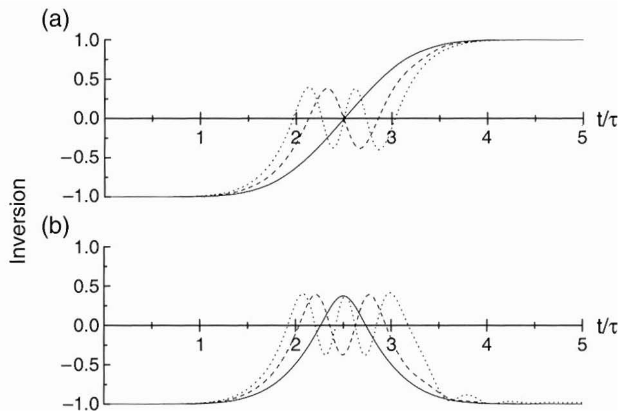

Fig. 2. Rabi oscillation dynamics of a QD interacting with the Gaussian pulse: $t_0 = 0.5\tau$, $\tau\Delta\omega = 15$. Inversion regimes: (a) $\xi = 1.0$ (solid line), $\xi = 1.26$ (dashed line), $\xi = 1.51$ (dotted line); (b) $\xi = 1.13$ (solid line), $\xi = 1.39$ (dashed line), $\xi = 1.61$ (dotted line).

3. Strong coupling of the QD with quantum light

According to [22], the model Hamiltonian for the QD interacting with nonclassical light can be written as $H = H_{\mathrm{JC}} + \Delta H$, where H_{JC} is the Jaynes–Cummings Hamiltonian [27] and ΔH is the local field correction (1). Further, we restrict the analysis to a single electromagnetic mode; therefore the mode index k can be omitted. Let the wavefunction of the system "QD + electromagnetic field" be presented by

$$|\psi\rangle = \sum_n \sum_{\sigma=\pm} (A_n^\sigma |n,\sigma\rangle + B|0,g\rangle) = \sum_n (A_n^+|n,+\rangle + A_n^-|n,-\rangle + B|0,g\rangle), \quad (10)$$

where A_n^σ and B are the unknown coefficients to be found; $|n,\sigma\rangle$ is the dressed state basis, which is as follows [27]:

$$|n,\sigma\rangle = a_n^\sigma |g\rangle|n+1\rangle + b_n^\sigma |e\rangle|n\rangle, \quad (11)$$

with parameters $a_n^+ = -b_n^+$ and $a_n^- = b_n^-$ (see, e.g., Ref. [27] for details); $|n\rangle$ denotes the field state with n photons in the given mode. Substituting wavefunction (10) into the Shrödinger equation and carrying out some standard manipulations, we come to an infinite chain of differential equations for slowly varying amplitudes for arbitrary n:

$$\begin{aligned} i\hbar\frac{\partial A_n^\sigma}{\partial t} &= \epsilon_n^\sigma A_n^\sigma + \sum_{m,\sigma'} S_{nm}^{\sigma\sigma'} + Q_n^\sigma B, \\ i\hbar\frac{\partial B}{\partial t} &= \epsilon_g B + \sum_{m,\sigma} Q_m^{\sigma *} + \mathcal{G}B, \end{aligned} \quad (12)$$

where

$$S_{nm}^{\sigma\sigma'} = \langle\sigma,n|\Delta H|m,\sigma'\rangle, \quad Q_n^\sigma = \langle\sigma,n|\Delta H|0,g\rangle, \quad \mathcal{G} = \langle g,0|\Delta H|0,g\rangle. \quad (13)$$

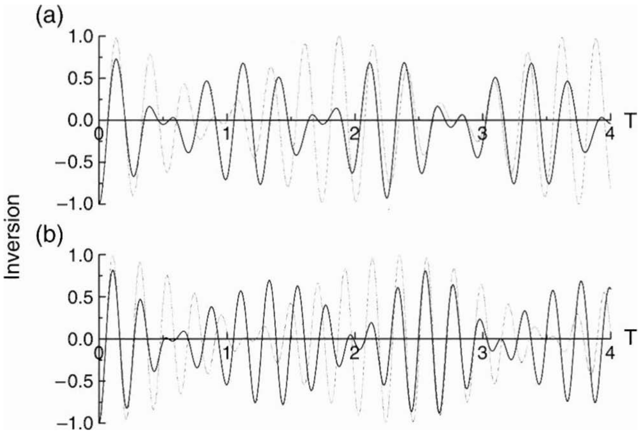

Fig. 3. Rabi oscillations of a QD interacting with a Fock qubit. Dashed lines represent the absence of local fields. $N = 3$ (a) and $N = 5$ (b).

The indexes σ, σ' in Eq. (13) denote the sum over $+$ and $-$ states of wavefunction (10);

$$\epsilon_n^\sigma = \frac{1}{2}[\epsilon_e + \epsilon_g + (2n+1)\hbar\omega] + 2\hbar\omega_n^\sigma, \quad \omega_n^\sigma = \pm\frac{1}{2}\sqrt{(\delta + \Delta\omega)^2 + \Omega_n^2};$$

Ω_n is the Rabi frequency. Note that the term in (10) resulting from the local field impact provides nonlinear coupling of all dressed states of the electromagnetic field, while the system remains uncoupled and linear when $\Delta\omega = 0$.

Let us consider as an example a QD interacting with Fock qubits. The Fock qubit is a superposition of two arbitrary Fock states which are states with fixed numbers of photons. The wavefunction of the Fock qubit is given by $\beta_N|N\rangle + \beta_{N+1}|N+1\rangle$. Here, β_N and β_{N+1} are complex-valued quantities for which the normalization relation $|\beta_N|^2 + |\beta_{N+1}|^2 = 1$ is fulfilled. Therefore, in system (12) it is enough to take the functions A_{N-1}^σ, A_N^σ and A_{N+1}^σ into account. Let the QD be in the ground state. Then a relation as follows:

$$(\beta_N|N\rangle + \beta_{N+1}|N+1\rangle)|g\rangle = \sum_n A_n^\sigma(0)|n,\sigma\rangle + B(0)|0,g\rangle$$

holds for the QD considered. From this equation, initial conditions for system (12) can be obtained:

$$A_{N-1}^\sigma(0) = a_{N-1}^\sigma\beta_N, \quad A_N^\sigma(0) = a_N^\sigma\beta_{N+1}, \quad A_{N+1}^\sigma(0) = 0. \tag{14}$$

Then, Eqs. (12) imposed on (12) describe the dynamics of the interaction between the QD and the Fock qubit.

Let us consider the case where $\delta = -\Delta\omega$; then $\omega_n^\sigma = \pm g\sqrt{n+1}$. Fig. 3 represents the numerical calculations of the inversion for the Fock qubit with $\beta_N = \beta_{N+1} = \sqrt{0.5}$, when $g = 0.5\Delta\omega$. Fig. 3(a) shows the case with $N = 3$, while Fig. 3(b) depicts the case with $N = 5$. Dashed lines denote the conventional picture of the Rabi oscillations, i.e., the case with the local field effect eliminated. One can see that the cases of Rabi oscillation dynamics are strongly influenced by local fields. As a result, the Mollow triplet [24], which characterizes atomic Rabi oscillations in the frequency domain, is expected to be transformed under the local field effect to a more complicated spectrum containing higher order satellites of the Rabi frequency.

4. Conclusions

On the basis of optical Bloch equations describing QD interactions with different states of light, the Rabi oscillation dynamics has been analyzed. As a result, we have revealed a strong dependence of the period of Rabi oscillations on the frequency shift, resulting from the local field influence as well as the bifurcation and anharmonism in the Rabi oscillations. The final state of the inversion as a function of the peak strengths of the Gaussian pulse demonstrates step-like transitions. For the QD interacting with the Fock qubit, we predict a local field induced essential modification of the Rabi oscillation picture.

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