Spin-dependent Rabi oscillations in single quantum dot

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Ultrafast optical pump of exciton resonance in single quantum dot by an elliptically polarized laser pulse is described by the non-Markov balance equations. Population and spin dynamics are investigated and spin-dependent Rabi oscillations are considered for the case of a flat quantum dot, with the lateral size greater than the height. The following peculiarities of temporal evolution have been found: a) a beating of Rabi oscillations under an elliptically polarized pump and b) quenching of spin orientation due to a spin-orbit splitting effect.

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Investigations of excitonic excitations in a single quantum dot (SQD) have been started in the past decade (see recent Refs. in [1]). The Rabi oscillations and spin-related effects in a QD ensemble have also been investigated, see [2] and [3] respectively. Recently the Rabi oscillations of electron population in SQD excited by ultrashort laser pulse [4, 5]. Due to this, the population redistribution. For the simplest case of a flat quantum dot, with the lateral size greater than the height, and have estimated the maximal kinetic equation (see evaluation in [6, 7]):

\[ \tilde{\delta} H_t = \frac{i e}{\omega} E w_t \begin{pmatrix} 0 & \hat{g}_e \\ \hat{g}_e & 0 \end{pmatrix} \]

(2)

Here we consider the interband photogeneration caused by the electric field \( E e^\exp(-i\omega t) + c.c.\). The averaged over polarization-dependent matrix \( \hat{g}_e = (\hat{e} \cdot \hat{v}) \) is written through the interband velocity matrix element, \( \hat{v} \), and \( \hat{g}_e \) is the non-Hermitian matrix because \( \hat{e} \neq \hat{e}^\ast \).

Below, we shall restrict our consideration to the vicinity of the excitonic resonance when \( \hat{R}_t \) can be replaced by the diagonal matrix \( \begin{pmatrix} \hat{\rho}_{e,t} & 0 \\ 0 & \hat{\rho}_{o,t} \end{pmatrix} \) with 2 × 2 matrices \( \hat{\rho}_{e,t} \) and \( \hat{\rho}_{o,t} \) describing the excited (e-) and vacuum (o-) states respectively. In agreement with the experimental conditions [4], we have neglected here the two-photon biexcitonic resonance, see [3] and Refs. therein. Thus, we transform Eqs.(1.2) into the following form:

\[ \frac{\partial}{\partial t} \begin{pmatrix} \hat{\rho}_{e,t} \\ \hat{\rho}_{o,t} \end{pmatrix} = \pm \left( \frac{e E}{\hbar \omega} \right)^2 \int_{-\infty}^{t} dt' w_{t'} e^{i\Delta \omega (t'-t)} \times (\hat{g}_e \hat{\rho}_{o,t'} - \hat{\rho}_{e,t'} \hat{g}_e) \hat{g}_e^\ast + H.c., \]

(3)

where \( \hbar \Delta \omega \) is the detuning energy with respect to the exciton peak energy. Adding these Eqs. we obtain that \( \hat{\rho}_{e,t} + \hat{\rho}_{o,t} \) is a time-independent matrix which is determined through initial conditions (see below).

To calculate the matrix \( \hat{g}_e \) we neglect the Coulomb renormalization of the interband velocity matrix element and consider

\[ \langle \lambda | \hat{\psi} \rangle \approx \sum_{jj'} v_{jj'} \int d\mathbf{x} e^{i\psi^*_j \hat{x} x} e^{i\psi_{j'} \hat{x}'}. \]

(4)

Here \( v_{jj'} \) is the 8 × 8 interband velocity matrix and \( x \) is the in-plane vector. For the flat QD model, we write
the overlap integral in (3) through the in-plane envelope functions $\psi^\lambda_{x}$. Here we have replaced the transverse contribution to the overlap integral by unit considering transitions between top $\nu$-band states and bottom $c$-band states. Neglecting the nonparabolic ($x\, p^3$) contributions, we describe the $\nu$-band states by $\varphi^{\nu(h)}_x(\sigma)$, where $\varphi^{\nu}_x$ takes into account the heavy-light hole mix $|\tilde{c}\rangle$ and $|\sigma\rangle$ corresponds to the degenerate spin state directed along $OZ$ with the spin numbers $\sigma = \pm 1$. Taking into account noticeable spin-orbit effects in $c$-band we write the eigenstate problem for spinor $\psi^{\nu(c)}_x$ as follows:

$$\begin{split}
\left\{ \frac{\ddot{p}^2}{2m} + (\sigma \cdot [v_x \times \vec{p}]) - \varepsilon \right\} \varphi^{\nu(c)}_x = 0,
\end{split}$$

(5)

where $\sigma$ are the Pauli matrices, $m$ is the electron effective mass, and the characteristic spin velocity $v_x$ is along the growth axis, $OZ$ $[9, 10]$.

We shall restrict our consideration to the case of weak spin-orbit effect, performing the unitary transformation $\psi^{\nu(c)}_x = \exp[-i\chi(\sigma \cdot [v_x \times x])]/\hbar \varphi^{\nu}_x$. Taking into account $\propto v_x^2$ contributions only, we obtain the Schrodinger equation in the form:

$$\begin{split}
\left\{ \frac{\ddot{p}^2}{2m} - 2mv_x^2[1 - (\sigma \cdot [\vec{p} \times x])]/\hbar - \varepsilon \right\} \varphi^{\nu}_x = 0.
\end{split}$$

(6)

We consider the equation with the zero boundary condition $\varphi^{\nu(c)}_x = 0$; the curve $\Gamma$ is bounded by a flat QD with the area $S$. Note that the momentum operator here, $[\vec{p} \times x]/\hbar$, is along $OZ$, so that the spinor $\varphi^{\nu}_x$ is proportional to the above introduced spinor $|\sigma\rangle$.

Next, using the two-level basis including the lower electron states with $\sigma = \pm 1$ and the upper hole states with $\sigma' = \pm 1$, we obtain the only non-zero matrix elements (2) for the transitions $|e, 1\rangle \rightarrow |o, 1\rangle$ and $|e, -1\rangle \rightarrow |o, -1\rangle$; see similar selection rules in $[9, 10]$. For the case of elliptically polarized excitation described by $\mathbf{e} = \lambda_+ \mathbf{e}_+ + \lambda_- \mathbf{e}_-$, where $\mathbf{e}_\pm$ are the polarization vectors for $\pm$ circular waves and $\lambda_\pm$ are complex numbers (moreover $|\lambda_+|^2 + |\lambda_-|^2 = 1$), we obtain the matrix $\mathbf{g}_e$ in the form:

$$\mathbf{g}_e = v_{\lambda\perp} \begin{vmatrix} \lambda_- & 0 \\ 0 & -\lambda_+ \end{vmatrix}.$$ 

(7)

The non-zero coefficients here are written through

$$v_{\lambda\perp} = \int d\mathbf{x} \varphi^{\nu(c)}_x(\sigma) \varphi^{\nu(h)}_x \simeq \mathcal{P} \left[ 1 - \alpha \left( \frac{mv_x}{\pi \hbar} \right)^2 S \right],$$

(8)

where $\mathcal{P}$ is the Kane velocity. The overlap integral here have been calculated for the cases of circular and square QDs when the numerical coefficient $\alpha$ is equal to 0.11 and 0.32 respectively. A suppression of the matrix element (7) with increasing $v_x$ also takes place for the conic QD case [11].

We describe the coherent dynamics by the use of the diagonal population numbers for the excited and ground states, $n_{e, \sigma t} = \langle e, \sigma | \rho_{e, t} | e, \sigma \rangle$ and $n_{o, \sigma t} = \langle o, \sigma | \rho_{o, t} | o, \sigma \rangle$. The system (3) is transformed into the independent equations for the states with $\sigma = \pm 1$:

$$\begin{align*}
\frac{d}{dt} \left| n_{e, \pm 1t} \right|^{\nu} = \pm \nu_{r} |\lambda_\pm|^2 & \times \int_{-\infty}^{t} \frac{dt'}{\tau_p} \Phi_{t't} (n_{o, \pm 1t'} - n_{e, \pm 1t'}).
\end{align*}$$

(9)

Here we have introduced the kernel $\Phi_{t't} = \nu_{t} \nu_{r} \cos \Delta \omega (t - t')$, and $\nu_r = 2(\varepsilon E/\hbar^2)^2 |v_{\lambda\perp}|^2/\tau_p$ is the characteristic photogeneration rate. Since $n_{e, \pm 1t} + n_{o, \pm 1t}$ are independent of time, and the oscillation strengths for $\pm$ transitions are proportional to $|\lambda_\pm|^2$, we use the phenomenological conditions in the form

$$n_{e, \pm 1t} + n_{o, \pm 1t} = |\lambda_\pm|^2,$$

(10)

which is in agreement with the particle conservation requirement $\sum_\sigma (n_{e, \sigma t} + n_{o, \sigma t}) = 1$. Using Eqs.(9), we obtain the closed equation for the population differences, $\Delta n_{\pm t} = n_{o, \pm 1t} - n_{e, \pm 1t}$, in the form:

$$\frac{d\Delta n_{\pm t}}{dt} + \nu_{r} |\lambda_\pm|^2 \int_{-\infty}^{t} \frac{dt'}{\tau_p} \Phi_{t't} \Delta n_{\pm t} = 0$$

(11)

with the initial conditions $\Delta n_{\pm t \rightarrow -\infty} = |\lambda_\pm|^2$ which are obtained from Eq.(10) and $n_{e, \pm 1t \rightarrow -\infty} = 0$.

The concentration of photoexcited electrons is given by $n_t = n_{e, \pm 1t} + n_{o, \pm 1t}$ while the spin orientation, $S_t$, is determined as follows $S_t = \sum_{\sigma=\pm} \langle e, \sigma | \sigma | e, \sigma \rangle n_{e, \sigma t}$. The only $z$-component of spin appears to be non-zero and

$$\begin{align*}
S_{zt} = k(n_{e, \pm 1t} - n_{e, \pm 1t}) & \equiv k \delta n_t, \\
k & \approx 1 - 2\alpha \left( \frac{mv_x}{\pi \hbar} \right)^2 S,
\end{align*}$$

(12)

where $k = \int d\mathbf{x} \varphi^{\nu(c)}_x(\sigma) \varphi^{\nu(h)}_x$ and $\alpha$ is the coefficient from Eq.(8). The spin orientation is proportional to the redistribution between the $\pm$ spin states, $\delta n_t$, and the factor $k$ decreases with increasing $v_x$.

For the resonant excitation case, $\Delta \omega = 0$, the kernel in Eq.(11) is not dependent on $t - t'$, one obtains the second-order differential equation

$$\begin{align*}
\frac{d^2\Delta n_{\pm t}}{dt^2} - \frac{2t}{\tau_p} \frac{d\Delta n_{\pm t}}{dt} + \frac{\nu_{r}}{\tau_p} |\lambda_\pm|^2 E^{-2(t/\tau_p)^2} \Delta n_{\pm t} & = 0
\end{align*}$$

(13)

with the use of an additional initial condition $[v_{\lambda\perp}^{-1} d\Delta n_{\pm t}/dt]_{t \rightarrow -\infty} = 0$. The solution of Eq.(13) takes the form:

$$\Delta n_{\pm t} = |\lambda_\pm|^2 \cos \left( \frac{A|\lambda_\pm|}{\sqrt{\pi}} \int_{-\infty}^{t/\tau_p} d\tau e^{-\tau^2} \right),$$

(14)

where $A = \sqrt{\pi \nu_{r}/\tau_p}$ is the excitation amplitude and $|\lambda_\pm|^2 = (1 \pm s)/2$ with the degree of circular polarization $s = |\lambda_+|^2 - |\lambda_-|^2$. 
FIG. 1: Temporal evolution of the total population, \( n_t \), and the spin redistribution, \( \delta n_t \), under the excitation amplitudes, \( A = \pi \) (a) and \( A = 3.5\pi \) (b). Solid, dashed and dotted curves correspond \( n_t \) under circular, elliptical and linear polarized pumps respectively. The dot-dashed curve presents \( \delta n_t \) under elliptical polarized pump. The pulse profile is shown as the short-dot curve.

In Fig.1 we plot the temporal evolution of \( n_t \) and \( \delta n_t \) for the cases of circular, elliptical (with \( s = 1/2 \)), and linear polarized pumps under the resonant excitation condition \( \Delta \omega = 0 \). Note that \( \delta n_t = -n_t \) and \( \delta n_t = 0 \) for the cases of circular and linear polarized pumps respectively. For the \( \pi \)-pulse excitation case (if \( A = \pi \)), one obtains monotonic increase of \( n_t \) with \( n_t \to \infty = 1 \) for the circular pump. The spin redistribution, \( \delta n_t < 0 \), varies with \( t/\tau_p \) in a similar manner. As the excitation amplitude increases, the temporal evolution becomes oscillatory, as shown in Fig.1b for \( A = 3.5\pi \). These dependencies have different periods, for the circular, linear, and elliptically polarized pumps.

Next we consider the detuning effect by calculating the solution of Eq.(11) numerically. In Fig.2 we plot the temporal evolution of \( n_t \) and \( \delta n_t \) under different \( \Delta \omega \tau_p \). The detuning results in suppression of both the Rabi flop after the pulse and the amplitude of oscillations if \( A > \pi \). The characteristics of the temporal response for different polarizations of pump do not change in comparison with the resonant case.

The oscillations of Rabi flopping population, \( n \equiv n_t \to \infty \), and the redistribution between the \( \pm \) spin states, \( \delta n \equiv \delta n_{t \to \infty} \), are shown in Fig.3 for the resonant excitation amplitudes, \( A \), are indicated by arrows.
circular case. The harmonic dependencies with periods $A = 2\pi$ and $A = 2\sqrt{2} \pi$ take place for the circular and linear polarized pumps respectively. By contrast, one can see a beating of $n$ and $\delta n$ with $A$ under the elliptically polarized pump. Moreover, the spin orientation may change a sign for this case; once again, $\delta n = -n$ and $\delta n = 0$ for the circular and linear polarized cases respectively.

Let us discuss the assumptions used in the above calculations. The main restriction is the consideration of the vicinity of the excitonic resonance without the contribution of two-photon biexcitonic resonance. Here we have applied the phenomenological initial conditions for Eq. (11). This is valid if $\hbar \Delta \omega$ and $\hbar / \tau_p$ are smaller than the exciton-biexciton splitting energy (around 3 meV in $\text{InGaAs}$ QDs) while a more detailed many-particle consideration is required for greater $\hbar \Delta \omega$ and/or $\hbar / \tau_p$. We also have described the coherent response neglecting relaxation because $\tau_p$ is shorter than the relaxation times. Next, the non-Markov equation (1) describes the interband generation of electron-hole pairs to second order accuracy, i.e. we have supposed that the energy $|e|E_{\nu R,L}/\omega$ is much lower than the energy gap. Finally, the simple model of QD has been used in order to obtain Eqs. (8,12) and more complicated numerical calculations, e.g. similar to [12], have to be performed in order to improve these simple formulas.

In conclusion, we have considered the peculiarities of Rabi oscillations in a single quantum dot due to the spin-dependent ultrafast excitation by an elliptically polarized pulse. For the simple model of SQD with the spin-flip transitions forbidden we have found both the beating of Rabi oscillations under an elliptically polarized pump and the quenching of spin orientation due to spin-orbit splitting effect. A more detailed treatment of these effects requires the consideration of complicated selection rules and to describe the exciton-biexciton resonance in the framework of a many-particle approach. What this paper does, however, is to determine the conditions for observation of the peculiarities mentioned.

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