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Non-Markovian damping of Rabi oscillations in semiconductor quantum dots

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Abstract
A systematic investigation is performed on the damping of Rabi oscillations induced by an external electromagnetic field interacting with a two-level semiconductor system. We have considered a coherently driven two-level system coupled to a dephasing reservoir and shown that, to explain the dependence of the dephasing rate on the driving intensity, it is essential to consider the non-Markovian character of the reservoir. Moreover, we have demonstrated that intensity-dependent damping may be induced by various dephasing mechanisms due to stationary as well as non-stationary effects caused by coupling with the environment. Finally, present results are able to explain a variety of experimental measurements available in the literature.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Rabi oscillations (ROs) of an emitter's level population due to the coupling with a driving field are already well established and have been used as a universal tool for the control of the dynamics of localized emitters. The existence of ROs has been demonstrated in a variety of systems such as atoms [1], superconductor devices [2–4], quantum wells [5] and semiconductor quantum dots (QDs) [6–10]. Recently, localized semiconductor systems exhibiting few discrete energy levels, such as specially selected donor impurities and QDs (termed as 'artificial atoms'), are considered as prospective candidates to play the role of basic building blocks of quantum information processes [11]. They demonstrate typical quantum dynamical features of isolated atoms. In particular, they show ROs when coupled to a driving field. So, they can be effectively controlled by using ultra-short intensive laser pulses [6, 12]. By achieving control over the dynamics of these localized states, two-qubit and multi-qubit gate operations become real possibilities, provided that the decoherence time is long enough [13]. For example, recently, the possibility of operating two-qubit gates with excitons and bi-excitons in QDs was demonstrated [14] and a single-qubit Deutsch–Jozsa algorithm was experimentally realized [15].

In localized semiconductor systems, ROs might occur in a quite different way than for isolated atoms. In contrast with real atoms, semiconductor artificial ones couple with the surroundings in a more complicated manner. There are a variety of loss mechanisms for localized semiconductor systems, and some of them are essentially non-Markovian ones, so that one needs to take into account memory effects as well as the back-action of the dissipative reservoir on the radiating system. For example, a dephasing caused by spin–spin coupling with neighboring QDs or carriers captured in traps in the vicinity of QDs was shown to lead to non-Markovian dynamics [16–18]. Such reservoirs may have correlation times comparable with the typical decoherence time of the dephasing system. A dephasing due to coupling with phonons was also shown to lead to non-Markovian features in the system's dynamics [19–21]. The influence of electron–hole dipole–dipole interactions (local-field effects) was shown to lead to significant changes in the features of the ROs [22]. In addition, carriers and excitons in localized semiconductor systems may be coupled not only to localized neighboring states but to delocalized ones as well [23]. Exciton leakage to the wetting layer [24] and Auger capture [25] may also be a significant source of dephasing in QDs. As decoherence is one of the main obstacles to quantum information, its understanding is then crucial to the future development of quantum information processes based on semiconductor QDs.

The diversity of dissipation channels has led to a number of novel features in the observation of ROs in such semiconductor systems. In the present work we focus our attention on one peculiar phenomenon which has caused and is still causing much controversy, namely the damping of ROs due to the increase of the driving pulse area, which is an observed feature of coherently excited localized semiconductor systems [6–10]. Many conflicting attempts to explain the dephasing process behind the ROs have been suggested. For example, it has been attributed to the system's interaction with a non-Markovian reservoir of phonons [19, 20]. Nevertheless, there is evidence that the dephasing process takes place even when the coupling with phonons is negligible [8]. Another proposal was based on excitations of bi-excitons [24, 26] in the QD, although damped ROs are also observed when there is no possibility of bi-exciton excitations [8]. Recently, it has been demonstrated that the experimentally observed [7] intensity-dependent damping of ROs can be reproduced by introducing into the standard Bloch equations a dephasing rate dependent on the driving-field intensity [27, 28]. On the other hand, although there is an experimental confirmation of a driving dependence on the dephasing rate [8], an intensity-independent dephasing rate has also been measured [9].

The present investigation addresses this problem by studying a simple two-level emitter excited by a classical coherent field and coupled to a general dephasing reservoir. The present work consists of an extended version that includes details and further results of a short letter recently published [29]. Within a straightforward approach based on the interaction of the dressed system with a general reservoir, we have demonstrated that a damping dependence on the driving-field intensity may be achieved through a variety of mechanisms which are observed in different experimental set-ups, shedding some light on apparently conflicting interpretations of the phenomenon. Finally, we show that a driving-dependent damping of ROs may occur whether the reservoir is influenced or not by the external field. We organize the present study as follows: in section 2 we derive a time-local master equation by averaging over the reservoir and obtaining generalized Bloch equations. In section 3, we apply the formalism to a stationary reservoir that is not influenced by the external driving and obtain an intensity-dependent dephasing rate. In section 4 we investigate the influence of the driving over the reservoir and study the effects of non-stationary processes that occur in this case. Discussion and conclusions are given in section 5.

2. General formalism

We consider the simplest model of a two-level system (TLS) for a localized semiconductor emitter, which may represent, for instance, two low-lying states of donor impurities in a bulk semiconductor [6] or a two-level exciton system [7–9] in a QD. We assume that the system is driven by a strong resonant classical coherent field, and also that the population damping of the two-level system is negligible and that it is only affected by the coupling to a dephasing reservoir. This assumption corresponds to the real experiments carried out at low temperatures and with short intense laser pulses. This means that the population damping is negligibly slow (1 ns for experiments described in [7]) compared to the timescale of the system's evolution or to pulse widths of a few ps, and to inverse dephasing rates of a few tens of ps [30]. Note that the population damping due to exciton/carrier leakage may be controlled and be kept small on the timescale of the system's dynamics [31, 32].

In the following, we consider a rotating-reference frame with frequency, ω_L, equal to the driving-field frequency. We work in the 'rotating-wave approximation' (RWA). The function $\Omega(t)$ describes the temporal shape of the excitation pulse. Working in the interaction picture for the reservoir variables, the effective Hamiltonian describing the 'TLS + driving field + reservoir' system is given by [18]

$$H(t) = \hbar\sigma^+\sigma^-[\Delta + \mathbf{R}(t)] + \hbar[\Omega(t)\sigma^+ + \Omega^*(t)\sigma^-], \quad (1)$$

where $\sigma^\pm = |\pm\rangle\langle\mp|$ are the system's rising and lowering operators; $|\pm\rangle$ correspond to the excited and ground states of the TLS and $\Delta = \omega_0 - \omega_L$ is the detuning of the TLS transition frequency, ω_0, from the driving laser frequency ω_L, with the possible addition of a time-independent frequency-shift term originating from the interaction with the dephasing reservoir represented by the operator $\mathbf{R}(t)$. In general, $\mathbf{R}(t)$ may depend both on the dynamical and on the stochastic variables describing the reservoirs, which will be detailed below. We now assume that the reservoir correlation function $\langle\mathbf{R}(t)\mathbf{R}(\tau)\rangle$ (the averaging is assumed to be taken over the whole set of reservoir variables) satisfies the following requirement:

$$\langle\mathbf{R}(t)\mathbf{R}(\tau)\rangle \to 0, \quad (2)$$

when $t, \tau \to +\infty$ and $|t - \tau| \to +\infty$. Moreover, we suppose that the interaction of the system with the reservoir is weak and that a possible induced entanglement between the system and the reservoir disappears quickly on the timescale of the system's evolution (this is a necessary condition for using the approximations considered here). Using the Born approximation for the reservoir operator $\mathbf{R}(t)$, one may obtain a simple, local in time master equation for Hamiltonian (1), by following a scheme previously developed for the case of the interaction of a spin system with a strong driving field in the presence of a dephasing reservoir [16, 17, 33, 34]. The essence of this approach is to treat the reservoir as a perturbation acting on the TLS affected ('dressed') by the action of the driving field instead of considering the 'bare' TLS perturbed by the reservoir, as is usual. This is the most natural procedure whenever the driving field is sufficiently strong and then it is convenient to consider the TLS plus the driving field as a 'field-dressed TLS' interacting with the reservoir. The former 'bare'-TLS approach was assumed in a number of recent works on ROs in localized semiconductor systems (see, for example, the studies in [6–9, 12, 23, 24, 32]). In general, the bare-TLS approach is done by taking a set of standard Bloch equations for the description of the 'TLS + driving field' system, without a closer understanding of the procedure that originates them. As a result, some essential features of the problem have been neglected. In what follows, we hope to clear up that a straight implementation of such a standard procedure seems to be the major source of the controversy that has recently arisen.

Introducing the unitary 'dressing' transformation given by

$$U(t) = \overleftarrow{\mathbf{T}} \exp\left\{-\frac{\mathrm{i}}{\hbar}\int_{t_0}^{t} H_0(\tau)\,\mathrm{d}\tau\right\}, \tag{3}$$

where $\overleftarrow{\mathbf{T}}$ denotes the time-ordering operator, and

$$H_0(t) = \hbar\Delta\sigma^+\sigma^- + \hbar[\Omega(t)\sigma^+ + \Omega^*(t)\sigma^-], \tag{4}$$

is the non-perturbed Hamiltonian. 'Dressed' operators are defined as

$$\mathbf{S}^{\pm}(t) = U^+(t)\sigma^{\pm}U(t) = D_{+-}^{(\pm)}(t)\sigma^+\sigma^- + D_{+}^{(\pm)}(t)\sigma^+ + D_{-}^{(\pm)}(t)\sigma^- + D_{0}^{(\pm)}(t), \tag{5}$$

$$\mathbf{S}^{+}(t)\mathbf{S}^{-}(t) = U^+(t)\sigma^{+}\sigma^{-}U(t) = D_{+-}^{(+-)}(t)\sigma^+\sigma^- + D_{+}^{(+-)}(t)\sigma^+ + D_{-}^{(+-)}(t)\sigma^- + D_{0}^{(+-)}(t), \tag{6}$$

where the general forms of the 'dressing' functions $D_l^j(t)$, for arbitrary $\Omega(t)$, are given in [35]. For instance, within the RWA and for a rectangular driving pulse shape ($\Omega(t) \equiv \Omega/2 =$ const for the time interval of interest, $t \in [0, T]$, and $\Omega(t) = 0$ outside of it), one obtains

$$D_{+-}^{(+-)}(t) = \tfrac{1}{2}[1 + c^2 + s^2\cos(\Omega_{\mathrm{R}}t)], \tag{7}$$

$$D_{-}^{(+-)}(t) = \frac{\mathrm{i}\Omega}{2\Omega_{\mathrm{R}}}\{c[1-\cos(\Omega_{\mathrm{R}}t)] + \mathrm{i}\sin(\Omega_{\mathrm{R}}t)\}, \tag{8}$$

where $c = \Delta/\Omega_{\mathrm{R}}$, $s = \Omega/\Omega_{\mathrm{R}}$ and $\Omega_{\mathrm{R}} = \sqrt{\Delta^2 + \Omega^2}$.

In the interaction picture defined by the transformation (3), the Hamiltonian (1) takes the form

$$H^{\mathrm{int}}(t) = U^+(t)H(t)U(t) = \hbar\mathbf{S}^+(t)\mathbf{S}^-(t)\mathbf{R}(t), \tag{9}$$

which will be used to derive the master equation.

Then, under the Born approximation and conditions outlined above, one may use the time-convolutionless projection operator technique or a cumulant's expansion (see, for example, [16–18, 33, 34]) to obtain the general local in time master equation for the 'dressed' density matrix $\overline{\rho}(t)$ averaged over the reservoir (we assume an averaging on both quantum and classical stochastic variables and denote it by the symbol $\langle\cdots\rangle$), i.e.

$$\frac{\mathrm{d}}{\mathrm{d}t}\overline{\rho}(t) = -\mathrm{i}[\langle H^{\mathrm{int}}(t)\rangle, \overline{\rho}(t)] - \int_{t_0}^{t}\mathrm{d}\tau\,\langle[H^{\mathrm{int}}(t),[H^{\mathrm{int}}(\tau),\overline{\rho}(t)]]\rangle. \tag{10}$$

Using equation (9), one may write

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\overline{\rho}(t) = {}& -\mathrm{i}[\mathbf{S}^+(t)\mathbf{S}^-(t)\langle\mathbf{R}(t)\rangle, \overline{\rho}(t)] \\ & - \int_{t_0}^{t}\mathrm{d}\tau\,\langle\mathbf{R}(t)\mathbf{R}(\tau)\rangle\big(\mathbf{S}^+(t)\mathbf{S}^-(t)\mathbf{S}^+(\tau)\mathbf{S}^-(\tau)\overline{\rho}(t) \\ & - \mathbf{S}^+(t)\mathbf{S}^-(t)\overline{\rho}(t)\mathbf{S}^+(\tau)\mathbf{S}^-(\tau)\big) \\ & - \int_{t_0}^{t}\mathrm{d}\tau\,\langle\mathbf{R}(\tau)\mathbf{R}(t)\rangle\big(\overline{\rho}(t)\mathbf{S}^+(\tau)\mathbf{S}^-(\tau)\mathbf{S}^+(t)\mathbf{S}^-(t) \\ & - \mathbf{S}^+(\tau)\mathbf{S}^-(\tau)\overline{\rho}(t)\mathbf{S}^+(t)\mathbf{S}^-(t)\big).\end{aligned} \tag{11}$$

To 'undress' the density matrix $\overline{\rho}(t)$, the transformation inverse to the one given by equation (3) may be used to arrive at the following master equation for the 'bare' system's density matrix $\rho(t)$:

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\rho(t) = {}& -\mathrm{i}[H_0(t) + \sigma^+\sigma^-\langle\mathbf{R}(t)\rangle, \rho(t)] \\ & - \int_{t_0}^{t}\mathrm{d}\tau\,\langle\mathbf{R}(t)\mathbf{R}(\tau)\rangle\big(\sigma^+\sigma^-\mathbf{S}^+(\tau-t)\mathbf{S}^-(\tau-t)\rho(t) \\ & - \sigma^+\sigma^-\rho(t)\mathbf{S}^+(\tau-t)\mathbf{S}^-(\tau-t)\big) \\ & - \int_{t_0}^{t}\mathrm{d}\tau\,\langle\mathbf{R}(\tau)\mathbf{R}(t)\rangle\big(\rho(t)\mathbf{S}^+(\tau-t)\mathbf{S}^-(\tau-t)\sigma^+\sigma^- \\ & - \mathbf{S}^+(\tau-t)\mathbf{S}^-(\tau-t)\rho(t)\sigma^+\sigma^-\big).\end{aligned} \tag{12}$$

From the master equation (12) one obtains the following set of Bloch equations with time-dependent coefficients:

$$\frac{\mathrm{d}\rho_{++}}{\mathrm{d}t} = \mathrm{i}\left[\Omega(t)\rho_{-+} - \Omega^*(t)\rho_{+-}\right], \tag{13}$$

$$\frac{\mathrm{d}\rho_{+-}}{\mathrm{d}t} = \{\mathrm{i}[\Delta + \langle\mathbf{R}(t)\rangle] - \kappa(t)\}\,\rho_{+-} + \mathrm{i}\overline{\Omega}^*(t)[\rho_{--}(t) - \rho_{++}(t)], \tag{14}$$

where $\rho_{++} = \langle+|\rho|+\rangle$, $\rho_{\pm\mp} = \langle\pm|\rho|\mp\rangle$, and the time-dependent dephasing rate $\kappa(t)$ and the generalized Rabi frequency $\overline{\Omega}(t)$ are defined as

$$\kappa(t) = \int_{t_0}^{t}\mathrm{d}\tau\,\langle\mathbf{R}(\tau)\mathbf{R}(t)\rangle D_{+-}^{(+-)}(\tau-t), \tag{15}$$

$$\overline{\Omega}(t) = \Omega(t) - \int_{t_0}^{t}\mathrm{d}\tau\,\langle\mathbf{R}(\tau)\mathbf{R}(t)\rangle D_{-}^{(+-)}(\tau-t). \tag{16}$$

The Bloch equations ((13) and (14)) are the basis to study the coherently driven TLS coupled to a dephasing reservoir. As we shall see below, they provide an adequate qualitative description of the driving-dependent damping of ROs in a number of practical situations.

3. Undriven reservoir

In this section, the simplest situation is assumed, with the driving field interacting only with the localized TLS. In this case one may demonstrate that, if the reservoir is non-Markovian, driving-dependent damping of ROs occurs even with an unperturbed reservoir. Also, it is shown that the system will exhibit a Markovian or a non-Markovian behavior, depending on the driving-field intensity.

Firstly, one may consider the Markovian limit in which $\langle \mathbf{R}(\tau)\mathbf{R}(t)\rangle \sim \delta(\tau - t)$ and may assume that the average of the reservoir operator, $\langle \mathbf{R}(t)\rangle$, decays very fast ($\langle \mathbf{R}(t)\rangle \to 0$) on the timescale of the TLS dynamics. In the case of a rectangular driving pulse shape, equations ((7), (8), (15) and (16)) lead to $\kappa(t) = \kappa$, i.e. a constant dephasing rate independent of the driving-field intensity, and $\overline{\Omega}(t) = \Omega(t) = \Omega/2$. Substituting these values into equations ((13) and (14)), one obtains the standard system of Bloch equations:

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_{++}(t) = \mathrm{i}\frac{\Omega}{2}[\rho_{-+}(t) - \rho_{+-}(t)], \qquad (17)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\rho_{+-}(t) = (\mathrm{i}\Delta - \kappa)\,\rho_{+-}(t) + \mathrm{i}\frac{\Omega}{2}[\rho_{--}(t) - \rho_{++}(t)] \quad (18)$$

for the driven TLS with dephasing.

Typical examples of the upper level population of the TLS given by the Markovian Bloch equations ((17) and (18)) for the rectangular pulse are presented in figure 1, where one notes that the population tends to a constant value (=1/2) with increasing time (figure 1(a)). As expected, for fixed Rabi frequencies and increasing the κ dephasing rate, the oscillations decay faster. Figure 1(b) shows a π-pulse influence on the upper-state population of the TLS as a function of the Rabi frequency which is related to the pulse area as

$$\theta = \int_{-\infty}^{+\infty} \mathrm{d}\tau\, \Omega(\tau). \qquad (19)$$

For rectangular pulses ($\Omega(t) = \Omega/2$) the π-pulse area corresponds to $(\frac{\Omega}{2})T = \pi$, where T is the time needed for the upper-state population to reach its first maximum. Note that, in the Markovian limit, the population undergoes oscillations that persist for arbitrarily large Rabi frequencies (cf dashed curve in figure 1(b)).

A more general situation is found in the case of a non-Markovian reservoir, for which the reservoir correlation function is

$$\langle \mathbf{R}(\tau)\mathbf{R}(t)\rangle = K(\tau - t) + P(\tau, t), \qquad (20)$$

i.e. the sum of a stationary process, $K(\tau - t)$, tending to a non-zero value for $t = \tau$ when $t, \tau \to +\infty$, plus a non-stationary one, $P(\tau, t)$, tending to zero for $t = \tau$ when $t, \tau \to +\infty$. The non-stationary process $P(\tau, t)$ is responsible for the non-Markovian effects at the initial stage of the TLS dynamics. In section 4 we shall demonstrate that the non-stationary part of the reservoir correlation function may lead to a driving-dependent damping of ROs if the reservoir is excited by an intensive coherent pulse.

In this section we assume that $P(\tau, t)$ and the average of the reservoir's operator, $\langle \mathbf{R}(t)\rangle$, decay fast on the timescale of the TLS dynamics. Thus, $P(\tau, t)$ and $\langle \mathbf{R}(t)\rangle$ are neglected. One may represent $K(t)$ with the help of the Fourier transform $K(w)$ (which in some cases has the meaning of a spectral density of the reservoir's states, such as in the case of an electromagnetic field or in the case of a reservoir of phonons) in the following standard way:

$$K(t) = \int \mathrm{d}w\, K(w) \exp\{-\mathrm{i}(w - \omega_\mathrm{L})t\}. \qquad (21)$$

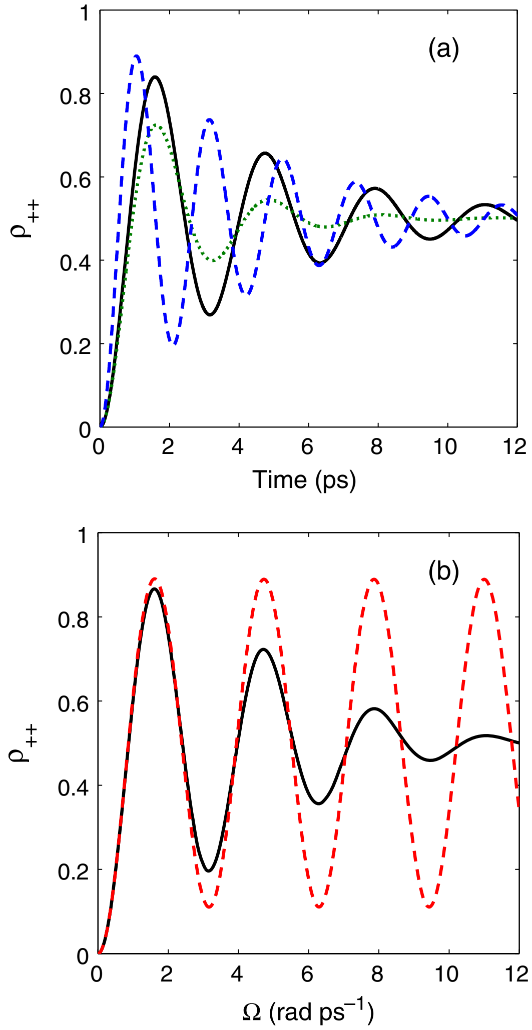

Figure 1. Examples of the TLS upper-state population time dependence at resonance: (a) the solid black line corresponds to the Markovian solution of equations (17) and (18) with $\kappa = 0.5$ rad ps^{-1}, $\Omega = 1$ rad ps^{-1}, the dashed blue line corresponds to the solution with $\kappa = 1$ rad ps^{-1}, $\Omega = 1$ rad ps^{-1} and the dotted green line corresponds to $\kappa = 0.5$ rad ps^{-1}, $\Omega = 1.5$ rad ps^{-1}; (b) the dashed red line corresponds to the Markovian solution of the Bloch equations (17) and (18) with $\kappa = 0.5$ rad ps^{-1}, whereas the solid black line is obtained for a non-Markovian reservoir and corresponds to the solution of the Bloch equations with a driving-dependent dephasing rate and generalized Rabi frequency given by equations (25) and (27), for which we have chosen $\pi K(\omega_\mathrm{L}) = 0.5$ rad ps^{-1}; $\frac{\mathrm{d}}{\mathrm{d}w}K(w)|_{w=\omega_\mathrm{L}} = 0$; $\frac{\pi}{4}\frac{\mathrm{d}^2}{\mathrm{d}w^2}K(w)|_{w=\omega_\mathrm{L}} = 0.05$ rad ps^{-1}.

For functions $K(w)$ sufficiently smooth in the vicinity of the laser frequency, ω_L, one may assume that [36]

$$\int_{t_0}^{t} \mathrm{d}\tau \int \mathrm{d}w\, K(w) \exp\{-\mathrm{i}(w - \omega_\mathrm{L})(\tau - t)\} \to \pi K(\omega_\mathrm{L}) - \mathrm{i}P \int \mathrm{d}w \frac{K(w)}{w - \omega_\mathrm{L}}, \qquad (22)$$

where P denotes the principal value of the integral; for $K(\omega)$ smooth near ω_L the second term on the right-hand side of (22) usually corresponds to a very small frequency shift (the Lamb shift in the unstructured vacuum is an example of it), which will be neglected in further considerations.

In the case of a rectangular pulse [$\Omega(t) = \Omega/2$], one obtains [16, 17] a dephasing rate given by (cf equations ((7), (8) and (15)))

$$\kappa(t) = \tfrac{1}{2}\int_{t_0}^{t} d\tau \int dw\, K(w)\exp\{-i(w-\omega_L)(\tau-t)\} \times \left(1 + c^2 + \frac{s^2}{2}\exp\{-i\Omega_R(\tau-t)\} + \frac{s^2}{2}\exp\{i\Omega_R(\tau-t)\}\right). \quad (23)$$

Note that equation (23) suggests that a sufficiently intense driving field would probe $K(w)$ far from the ω_L frequency and a single δ-function Markovian approximation would not be suitable for an adequate physical description of the system dynamics. On the other hand, if the $K(w)$ spectrum is smooth enough in the vicinity of all components of the Rabi splitting triplet ω_L, $\omega_L \pm \Omega_R$, one may perform a Markovian-like approximation for each of the triplet components (a procedure implemented by Kilin and Nizovtsev in studies on non-Markovian dephasing [16, 17], and recently used by Florescu and John [37] when dealing with the resonance fluorescence near the band edge). As the values of the $K(w)$ function are different at the triplet frequencies, it will result in an intensity-dependent dephasing rate, as shown below. Following the procedure (22), and neglecting frequency shifts, the expression (23) is now

$$\kappa(t) \equiv \kappa = \pi\frac{c^2+1}{2}K(\omega_L) + \frac{\pi s^2}{4}(K(\omega_L+\Omega_R) + K(\omega_L-\Omega_R)). \quad (24)$$

Now, if the values of the function $K(w)$ at ω_L, $\omega_L \pm \Omega_R$ are not very different, one may expand $K(w)$ around ω_L to obtain from equation (24) the following equation:

$$\kappa = \pi K(\omega_L) + \frac{\pi\Omega^2}{4}\frac{d^2}{dw^2}K(w)\Big|_{w=\omega_L} \quad (25)$$

for the κ dephasing rate, which depends on the intensity of the driving field. Here we point out that such an intensity-dependent recombination rate was used by Brandi *et al* [27, 28] to model experimental data on Rabi oscillations in localized semiconductor systems [7]. From equation (16), one has [16, 17]

$$\overline{\Omega}(t) = \frac{\Omega}{2} - \frac{i\Omega}{2\Omega_R} \times \int_{t_0}^{t} d\tau \int dw\, K(w)\exp\{-i(w-\omega_L)(\tau-t)\} \times \left(c - \frac{1+c}{2}\exp\{-i\Omega_R(\tau-t)\} + \frac{1-c}{2}\exp\{i\Omega_R(\tau-t)\}\right) \quad (26)$$

and, using the same approximation as before for equation (25), one obtains the generalized Rabi frequency

$$\overline{\Omega}(t) \equiv \overline{\Omega} = \frac{\Omega}{2} - i\pi\frac{\Omega}{2}\frac{d}{dw}K(w)\Big|_{w=\omega_L} + \frac{i\pi\Omega\Delta}{4}\frac{d^2}{dw^2}K(w)\Big|_{w=\omega_L}. \quad (27)$$

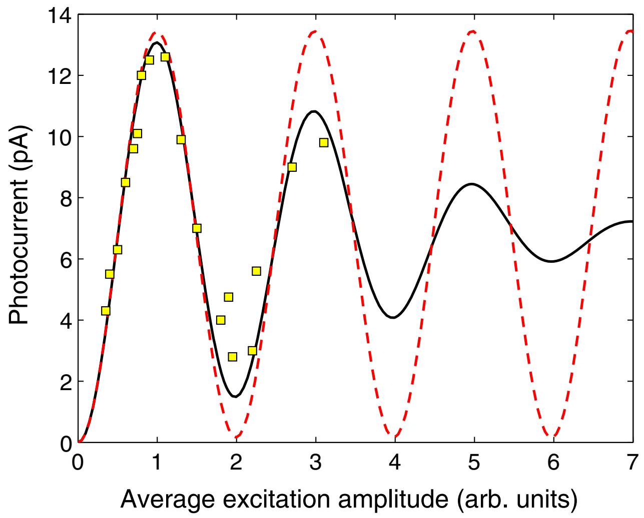

Figure 2. Rabi oscillations of the photocurrent, at resonance, as a function of the excitation amplitude. The red dashed line is the Markovian solution given by the Bloch equations with the dephasing rate independent of the driving field, whereas the solid line is obtained for a non-Markovian reservoir and corresponds to the solution of the Bloch equations with a driving-dependent dephasing rate and generalized Rabi frequency given by equations (25) and (27). Full squares represent experimental data from Zrenner *et al* [7] for a pulse width of about 1 ps. Here, a π-pulse corresponds to the unit of the excitation amplitude.

As expected (cf figure 1(b)), predictions of the Bloch equations based on the Markovian approximation are drastically different from those obtained by using equations (25) and (27), when one investigates the population values at a fixed time, while varying the driving-field amplitude (or Rabi frequency). The Markovian Bloch equations demonstrate that the population undergoes persistent oscillations (see the dashed curve in figure 1(b)), while the driving-dependent dephasing rate (cf equation (25)) leads to the decay of these oscillations. The decay rate of these oscillations increases with increasing driving pulse area, i.e. the amplitude of ROs decreases with the driving intensity and after some cycles the population tends to a constant value. As was mentioned above, such a behavior is typically found in experiments on ROs in localized semiconductor systems. In figures 2 and 3(a) we compare the present theoretical results of quantities that are proportional to the emitter's excited-state population by using a driving-dependent dephasing rate for the Bloch equations, with various available experimental data. One may notice that a quite good agreement is achieved by an appropriate choice of a couple of parameters (essentially a 'Markovian' dephasing rate, $\pi K(\omega_L)$, and $\frac{d^2}{dw^2}K(w)|_{w=\omega_L}$ in equation (25); they are fitting parameters) and by adjusting the timescale. In figure 3(b) we display, in a logarithmic scale, the theoretical amplitudes of the ROs at the $\theta = n\pi$ peaks and valleys, with results indicating, as expected, that the RO is faster damped for smaller τ pulse widths, as the laser intensity is increased (present theoretical results are in overall agreement with the experimental measurements of figure 1(d) by Wang *et al* [8]).

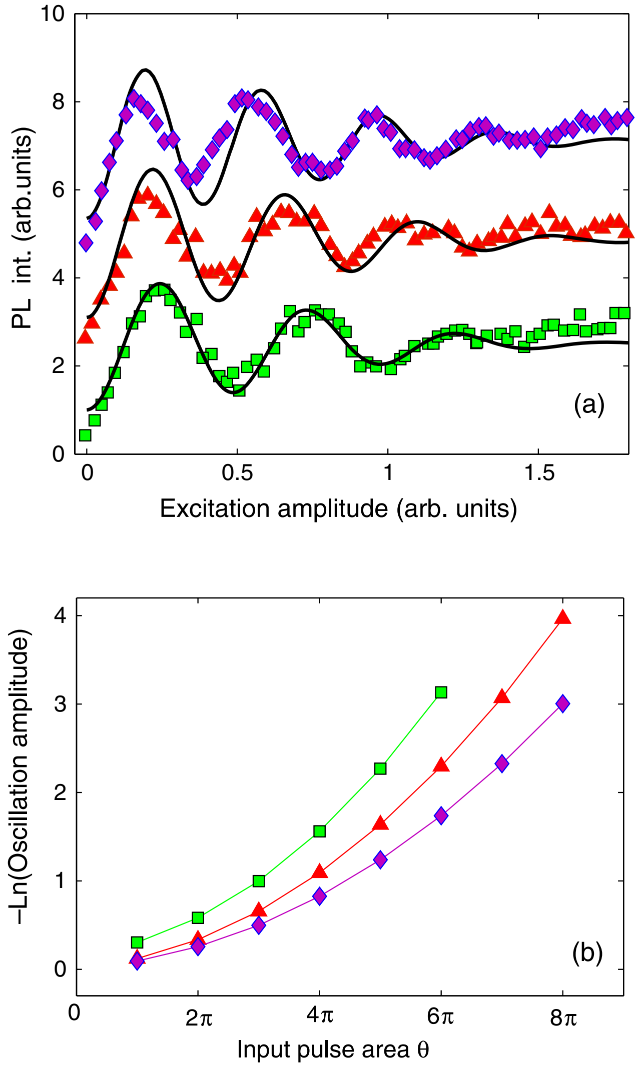

Figure 3. (a) ROs in the photoluminescence (PL) intensity, at resonance, with full theoretical curves corresponding in descending order to pulse widths of 9.3 ps, 7.0 ps and 5.4 ps, respectively. Calculations are performed for a non-Markovian reservoir and correspond to the solution of the Bloch equations with a driving-dependent dephasing rate and generalized Rabi frequency. Full symbols in (a) are the corresponding experimental data from Wang *et al* [8]; (b) negative logarithm of the RO amplitude as a function of the θ pulse area. Full symbols are the values calculated by using the corresponding theoretical values of the $\theta = n\pi$ oscillation amplitudes taken from the peaks and valleys of the theoretical ROs in (a).

To summarize the discussion presented in this section, we emphasize once more that the dependence of the dephasing rate on the driving intensity occurs as a natural consequence of the non-Markovian reservoir 'probed' by a sufficiently intense field driving the emitting system. Here we do not consider a 'full' non-Markovian reservoir, as we partially neglect effects of the reservoir (memory effects from $P(\tau, t)$ are not considered) and consider a Markovian-like approximation for the non-Markovian reservoir which only accounts for the difference between the values of the Fourier transform of the reservoir correlation function [16, 17] at different components of the Rabi triplet. It is well illustrated by the formulae in equations (25) and (27) which takes into account only the properties of the function $K(w)$ near one frequency point. We note that the particular nature of the reservoir is not specified here and it is not actually important as long as the reservoir correlation function satisfies quite general requirements.

4. Driven reservoir

In section 3 we have neglected the influence of an externally applied electromagnetic field on the localized system's surroundings. Let us now consider a more realistic situation where the coherent pulse influences both the localized TLS and its neighborhood, with effects on the wetting layer, defects, phonon excitations, etc. Effects of such an influence are twofold.

First, the stationary properties of the reservoir might be affected, i.e. the function $K(\tau - t)$ may become dependent on the Ω Rabi frequency. In this situation, one concludes that, for a driving that weakly affects the surroundings, the basic features of the system's dynamics described in section 3 will remain the same. Indeed, if one neglects the effects of the non-stationary aspects of the reservoir's correlation function ($P(\tau, t)$ in equation (20)) and assumes that its Fourier transform $K(w)$ (equation (21)) is smooth enough in the vicinity of the components of the Rabi triplet, one still ends up with equations (25) and (27). The only difference found is that the function $K(w, \Omega)$ is now dependent on the Rabi frequency. By using the expansion

$$K(w, \Omega) \approx K(w, 0) + \Omega \frac{d}{d\xi} K(w, \xi)\Big|_{\xi=\Omega}, \qquad (28)$$

and retaining in formulae ((25) and (27)) only terms up to the second order on Ω, one will find the same dependence on the Rabi frequency

$$\kappa(\Omega) \sim \kappa_0 + \kappa_1 \Omega + \kappa_2 \Omega^2, \qquad (29)$$

where the coefficients κ_i are defined by the expansion of the function $K(w, \Omega)$ and its derivatives. Of course, all the consequences of such dependence (driving-dependent damping of ROs in particular) are similar to those found in section 3.

Second, non-stationary effects, described by the non-stationary process $P(\tau, t)$ that composes the reservoir's correlation function, might come into play. Let us concentrate now on the consequences of non-Markovian effects of the system–reservoir interaction at the initial stage of the evolution, which are described by $P(\tau, t)$. We shall demonstrate that such essentially non-Markovian effects lead to the picture of the driving-dependent damping of ROs quite similar to those discussed in section 3.

To illustrate the nature and the consequences of these non-stationary effects let us consider a simple model of a bosonic reservoir driven by the same rectangular pulse that drives the TLS under investigation. Physically, it corresponds, for example, to a reservoir of phonons (or free carriers in the wetting layer, coupling to bi-excitons, a combination of mechanisms, etc) excited by the action of a driving field on the localized TLS and its neighborhood. Furthermore, we neglect the damping of the bosonic reservoir during the time intervals of interest. Describing the driving field–reservoir interaction within a rotating frame whose driving mode frequency is the same as the driving field ω_L and working in the RWA, one may write the following effective Hamiltonian for the whole system as

$$H_1(t) = \hbar\Delta\sigma^+\sigma^- + \hbar[\Omega(t)\sigma^+ + \Omega^*(t)\sigma^-] + \hbar\sigma^+\sigma^- \times \sum_j g_j(b_j^\dagger \exp\{i\omega_L(t-t_0)\} + b_j \exp\{-i\omega_L(t-t_0)\}) + \hbar\sum_j \Delta_j b_j^\dagger b_j + \hbar\sum_j \Omega_j(b_j^\dagger + b_j), \quad (30)$$

where $b_j^\dagger, b_j$ are bosonic creation and annihilation operators, respectively, for the jth mode of the reservoir, g_j are interaction constants of the TLS with the jth mode, Δ_j are the detunings of the reservoir modes from the driving field and Ω_j are the Rabi frequencies for every particular reservoir mode (here, for simplicity, chosen real).

Using the interaction picture with respect to the reservoir Hamiltonian

$$H_{res} = \hbar\sum_j \Delta_j b_j^\dagger b_j + \sum_j \Omega_j(b_j^\dagger + b_j), \quad (31)$$

one immediately finds the Hamiltonian (1) with the reservoir operator:

$$\mathbf{R}(t) = \sum_j g_j \left\{ b_j + \frac{\Omega_j}{\Delta_j} \right\} \exp\{-i(\omega_L + \Delta_j)(t-t_0)\} + \text{h.c.}, \quad (32)$$

and the TLS detuning shifted due to the interaction with the excited reservoir, i.e.

$$\Delta \rightarrow \Delta - 2\left\{ \sum_j g_j \frac{\Omega_j}{\Delta_j} \right\}. \quad (33)$$

Supposing that the initial state of the reservoir is a vacuum state, we find

$$\langle\mathbf{R}(t)\rangle = \sum_j g_j \frac{\Omega_j}{\Delta_j} \exp\{-i(\omega_L + \Delta_j)(t-t_0)\} + \text{h.c.}, \quad (34)$$

and the reservoir correlation function, $\langle\mathbf{R}(\tau)\mathbf{R}(t)\rangle$, becomes the sum of the stationary correlation

$$K(\tau, t) = \sum_j g_j^2 \exp\{-i(\omega_L + \Delta_j)(\tau - t)\}, \quad (35)$$

plus the non-stationary one:

$$P(\tau, t) = \langle\mathbf{R}(\tau)\rangle\langle\mathbf{R}(t)\rangle. \quad (36)$$

The dependence of the dephasing rate on the driving intensity that stems from the stationary correlation (35) leads to the effects already described in section 3 so that we neglect it in the following. Let us now assume that the spectrum of the reservoir's correlation function is wide and smooth enough to exhibit the stationary dephasing rate κ_s practically independent of the external driving. Then, from equation (15), one may conclude that

$$\kappa(t) = \kappa_s + \langle\mathbf{R}(t)\rangle \int_{t_0}^{t} d\tau \langle\mathbf{R}(\tau)\rangle D_{+-}^{(+-)}(\tau - t). \quad (37)$$

Clearly, the non-stationary dephasing rate $\kappa(t)$ decays in time, since $\langle\mathbf{R}(t)\rangle \rightarrow 0$ for $t \rightarrow \infty$. However the function $\langle\mathbf{R}(t)\rangle$ may decay slower than the stationary correlation function $K(t)$, considering that the influence of the external field over the different modes of the reservoir is different, and that the spectral density of the reservoir's excitation may be much narrower than the spectra of the reservoir's correlation function. Moreover, in experiments on ROs in localized semiconductor systems one deals with rather short driving pulses, so that the non-stationary dephasing rate might play a significant role in the system's dynamics. As the Rabi frequencies of the reservoir modes are proportional to the driving-field amplitude, one may take $\Omega_j \sim \Omega$ to obtain a dephasing rate of the form

$$\kappa(t) = \kappa_s + \Omega^2 f(t), \quad (38)$$

where the function $f(t) \rightarrow 0$ for $t \rightarrow \infty$. Thus here, even if one assumes $D_{+-}^{(+-)}(\tau - t) \approx 1$ (i.e. if one assumes Markovianity of the reservoir in the absence of the reservoir excitation by the driving), the non-stationary dephasing rate will be dependent on the driving-field intensity, which means that the intensity-dependent Rabi oscillation appear here as a purely non-Markovian dynamical effect. Furthermore, it leads to an intensity-dependent damping of the Rabi oscillations quite similar to the one considered in section 3. It should be noted that this effect may well be responsible for the constant value of the dephasing rate measured *after* the application of the driving pulse in the experiment by Patton *et al* [9]. We now illustrate the discussion of this section with a particular example. Let us assume a simple decaying form for the average of the reservoir operator

$$\langle\mathbf{R}(t)\rangle \sim \gamma\Omega \exp\{-\gamma(t-t_0)\}, \quad (39)$$

where γ is the decay rate of the average of the reservoir's operator. Such a decaying form of the correlation function was chosen, for example, in [16, 17]. One can see in figure 4 that even for the stationary dephasing rate κ_s independent of the driving, one finds decaying Rabi oscillations when the pulse increases. Note that, in the case that $D_{+-}^{(+-)}(\tau - t)$ is chosen as in equation (7), the κ dephasing rate displays oscillations which may be traced back to the $\cos(\Omega_R t)$ factor in the dressing function $D_{+-}^{(+-)}(t)$ (cf equation (37) and full curve in figure 4(a)). Also, as the decay time of the dephasing rate $\kappa(t)$ to reach its stationary value κ_s becomes shorter than the pulse width, experiments on the driving-damped ROs in localized semiconductor systems might as well exhibit a dephasing rate independent of the driving, when measurements are performed after the application of the excitation pulse. Moreover, it could also exhibit a decreased dephasing rate after the application of the pulse as in the experiment by Wang *et al* [8].

From the above considerations, one infers that a driving-dependent damping of ROs due to the non-stationary

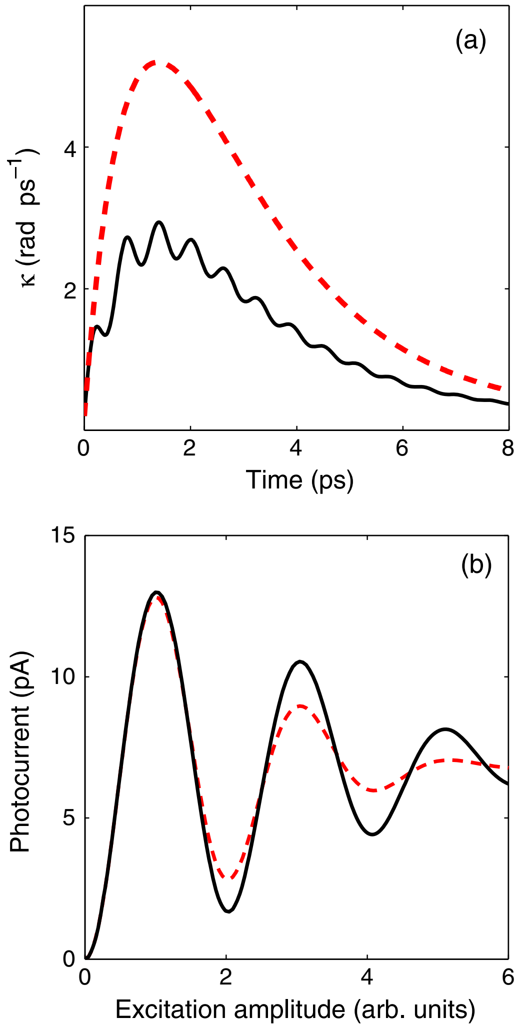

Figure 4. Examples of (a) temporal behavior of the $\kappa(t)$ dephasing rate (with $\Omega = 5$ rad ps^{-1}) and (b) upper-state population dynamics, at a time corresponding to the temporal width of the rectangular pulse, versus excitation amplitude. Calculations were performed at resonance with a dephasing rate given by equation (37) with $\kappa_s = 0.1$ rad ps^{-1} and a model reservoir correlation $\langle \mathbf{R}(t) \rangle$ defined by equation (39) with $\gamma = 0.5$ ps^{-1} (a). For (b) parameters were scaled in such a way as to fit the timescale of figure 2; $\gamma = 5$ ps^{-1} and $\kappa_s = 0.1$ rad ps^{-1} were chosen. In both figures red dashed lines correspond to $D_{+-}^{(+-)}(\tau - t) = 1$, whereas solid black lines correspond to $D_{+-}^{(+-)}(\tau - t)$ defined in equation (7).

reservoir's correlation function may take place for quite general reservoirs (an ensemble of other localized systems, traps, free carriers in a wetting layer, etc). We emphasize that non-stationary effects of a non-Markovian nature of the reservoir's excitation by the intensive coherent pulse might produce the intensity-dependent Rabi oscillations of the system's upper-state population in a similar fashion as those produced by the stationary dependence of the dephasing rate on the driving intensity.

5. Conclusions

In conclusion, we have demonstrated that the intensity-dependent damping of Rabi oscillations in localized semiconductor systems (such as QDs, systems of two-level shallow impurities in bulk semiconductors, etc) is an effect of a quite general nature. It is a consequence of the non-Markovian character of a reservoir to which the system is inevitably coupled. The exact nature of the reservoir (an ensemble of phonons, other localized systems, traps, free carriers in a wetting layer, coupling to bi-excitons or higher decaying levels, etc) is not particularly important to induce the effect. Particular details of the reservoir's structure and physics would only influence the quantitative characteristics of the system's dynamics, such as the number of ROs occurring until decay, a typical timescale of the process and the interplay between stationary and non-stationary effects of the interaction with the reservoir. The most important features for the manifestation of the driving-dependent ROs are (i) significant differences in values of the reservoir's correlation function spectrum on a scale determined by the Rabi frequency induced by the driving field and (ii) comparability of a decay time of the non-stationary correlation function of the reservoir and the temporal driving pulse width.

We have demonstrated that observable similar damping of ROs may occur as a consequence of very different physical mechanisms. The first one stems from the stationary properties of the reservoir whether or not the reservoir is affected directly by the driving. The second type of damping is purely of a non-stationary nature, occurring when the field excites the reservoir and the decay time of the non-stationary reservoir's correlation function is comparable with the driving pulse width. It is important to note when the contribution of the latter mechanism overcomes the others, so the measurement of the dephasing rate *after* the application of the driving pulse exhibits a smaller dephasing rate in comparison with the results obtained from measurements made during the action of the driving pulse. Therefore, one may even obtain the same value of the dephasing rate as in the absence of the driving. To conclude, we emphasize that the simple analysis given in the current work may be easily generalized and extended for more complicated localized systems (multi-level ones, for example), and for reservoirs of different natures.

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