

Rabi oscillations in two-level semiconductor systems

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Abstract

Rabi oscillations in coherent optical excitations in bulk GaAs and quantum dot two-level systems may be converted into deterministic photocurrents, with the impurities or dots providing the tag for each qubit. Here we perform a theoretical analysis of the damping of Rabi oscillations in two-level semiconductor systems. Present calculations, through optical Bloch equations on excitonic two-level $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum-dot systems, are found in good agreement with the corresponding experimental data. Calculated results indicate that the nature underlying the dephasing mechanism associated to the damping of the measured Rabi oscillations, which has previously remained as an open question, may be associated with a field-dependent recombination rate related to the inhomogeneous broadening of the excitonic lines in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ two-level QD system.

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Since, the discovery of fast quantum algorithms adequate to handle tasks as prime factorization, searching disordered databases [1] and the development of quantum error correction codes [2], there has been a great deal of interest in finding suitable physical systems that could provide the fundamental operational base (qubits) of a reliable quantum computer (QC). Among the suggestions, are the manipulation of nuclear spin states in bulk solution using nuclear magnetic resonance techniques (NMR) [3], trapped ions [4], photons in microcavity [5], and specifically selected donor [6,7] or exciton [8–12] states in semiconductor systems as qubits to be manipulated by laser radiation. An important aspect of solid-state based QC's is scalability, i.e. the

possibility of fabricating large integrated networks as required for realistic applications. The candidates to model qubits ideally would have in common three basic characteristics: (a) a precisely defined two-fold Hilbert space, (b) operate under weak decoherence, and (c) single-qubit and two-qubit unitary operations are controlled. One should note that a QC based on any of these architectures would be effective only if the decoherence time of the two-fold Hilbert space is much longer than the time involved in the single- and two-qubit operations. In fact, this could pose a problem if these operations are controlled by switching external gate potential and/or magnetic fields since, this cannot be performed very fast. This limitation may be overcome if, instead of slow external fields, one uses laser-probe pulses to control qubit operations, which may be achieved via coherent manipulation of two-level systems through Rabi oscillations in donor [6] or exciton [8,10,11] states by applying electromagnetic fields. Cole et al. [6] and

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Zrenner et al. [10,11] have demonstrated that the coherent optical excitations in bulk GaAs and $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum dot (QD) two-level excitonic systems, respectively, can be converted into deterministic photocurrents. Recently, Li et al. [12] have reported coherent biexciton Rabi oscillations and proposed the operation of an all-optical quantum gate, through the coherent optical control of a biexciton confined in a single QD.

It is well known that Rabi oscillations are a characteristic of a quantum-mechanical two-level system driven close to its resonance frequency. In the present work, we are concerned with a proper understanding of the damping of Rabi oscillations in two-level semiconductor systems [13]. We will show that the results of the experiments performed by Cole et al. [6] in a two-level donor system and Zrenner et al. [10,11] in a two-state exciton in a QD may be interpreted within a simple model essentially describing the coupling of a discrete level (e.g. ϕ_b) to a continuum set of states ($\Psi_{E,\beta}$), $0 \leq E < \infty$, as analyzed by Cohen-Tannoudji et al. [14]. Here we briefly recall their main results on the damping terms:

$$\langle \phi_b \| V \| \phi_b \rangle = 0 \quad (1)$$

$$\langle \Psi_{E,\beta} \| V \| \Psi_{E',\beta'} \rangle = 0$$

$$\langle \Psi_{E,\beta} \| V \| \phi_b \rangle = v(E, \beta)$$

where β represents all the quantum numbers other than E , and V is considered as a perturbation. In the present situation, one may choose

$$V = V_0 + \frac{e}{m_0 c} \mathbf{A} \cdot \mathbf{p} = V_0 - \mathbf{E}_{\text{rad}} \cdot \mathbf{d} \quad (2)$$

where \mathbf{E}_{rad} is the radiation electric field, \mathbf{d} is the electric dipole moment and V_0 represents all perturbative couplings other than the electric dipole interaction, and we have

$$v(E, \beta) = v_0(E, \beta) - \langle \Psi_{E,\beta} \| \mathbf{d} \| \phi_b \rangle \cdot \mathbf{E}_{\text{rad}} \quad (3)$$

The coupling induced by V between the discrete state $|\phi_b\rangle$ and the continuum is characterized by a recombination rate

$$\gamma_b(E) = \frac{2\pi}{\hbar} \int d\beta \rho(E, \beta) |v(E, \beta)|^2; \quad E \geq 0 \quad (4)$$

$$\gamma_b(E) = 0; \quad E < 0$$

with $\rho(E, \beta)$ being the continuum density of states. In the weak-field coupling limit, one may choose $E = E_b$ and, therefore

$$\gamma_b \sim (\alpha_0 + \alpha_1 E_{\text{rad}})^2 \sim \delta_0 + \delta_1 E_{\text{rad}} + \delta_2 E_{\text{rad}}^2 \quad (5)$$

This result clearly indicates a dependence-through E_{rad} of the recombination rate γ_b on the laser-field amplitude. In the experimental work by Cole et al. [6] involving a two-level 1s and $2p_+$ donor system in bulk GaAs, they have assumed Bloch equations with recombination rates γ_2 and γ_3 representing the total dephasing rate, which includes both intrinsic dephasing

and inhomogeneous broadening of the transition energy, and the ionization rate of the $2p_+$ state, respectively, [6]. One may interpret such recombination rates as arising from (i) coupling of the 1s state with continuum states chosen as a $2p_+$ broadened impurity band or (ii) coupling of the $2p_+$ state with conduction-band states, respectively. In Fig. 1 we present the experimental data (full squares) corresponding to the γ_2 and γ_3 recombination rates [6] associated to 1s and $2p_+$ donor states in GaAs. The overall qualitative agreement between experiment and present linear fittings in the applied electric field (dotted lines) is fairly good, with the quadratic dependence (Eq. (5)) not showing up probably due to the small values of the electric field of the applied terahertz radiation. Here we note that although the damped Rabi oscillations in shallow-donor states in bulk GaAs have been previously theoretically studied [7] via a driven two-level system, the electric-field dependence of the recombination rates has not been either discussed or explained before.

Excitonic Rabi oscillations may be theoretically studied following the same approach based in the Bloch equations which are obtained from the semiconductor Hartree–Fock (HF) equations for a two-level system [15]. Zrenner et al. [10,11] have studied the Rabi-flopping of the ground state exciton in a single self-assembled $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ QD through a photocurrent (PC) technique, which enables the monitoring of the occupation probability of the exciton states via measurements of the dc tunneling current out of the QD. In the case of π -pulse excitation, they were able to perform a systematic study of the field-dependence of the Rabi oscillations, at resonance, of the ground state exciton of a single QD. The experimental measurements on the photocurrent in $\text{In}_x\text{Ga}_{1-x}\text{As}$ excitonic two-level systems [10] may be understood within a driven two-level system by considering the optical Bloch equations

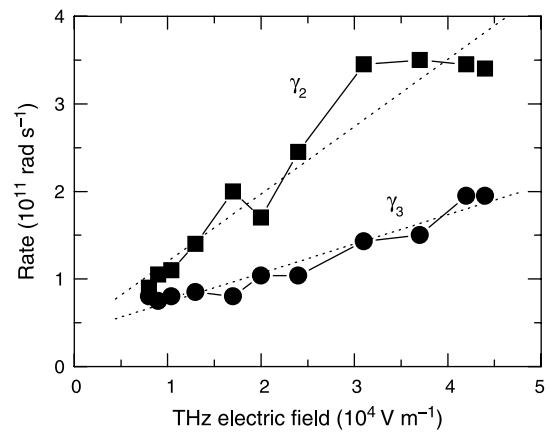


Fig. 1. Experimental γ_2 and γ_3 recombination rates (full symbols) associated to a two-level 1s and $2p_+$ donor system; γ_2 is the total dephasing rate, which includes both intrinsic dephasing and inhomogeneous broadening of the transition energy, and γ_3 is the ionization rate of the $2p_+$ state. The dotted lines are the present linear fittings (Eq. (5)).

$$\frac{d\rho_{11}}{dt} = -i\Omega_R^0 \cos(\omega_L t)(\rho_{21} - \rho_{12}) + \gamma_1 \rho_{22} \quad (6)$$

$$\frac{d\rho_{22}}{dt} = +i\Omega_R^0 \cos(\omega_L t)(\rho_{21} - \rho_{12}) - \gamma_1 \rho_{22}$$

$$\frac{d\rho_{12}}{dt} = +i\omega_{21}\rho_{12} + i\Omega_R^0 \cos(\omega_L t)(\rho_{11} - \rho_{22}) - \gamma_2 \rho_{12}$$

$$\frac{d\rho_{21}}{dt} = -i\omega_{21}\rho_{21} - i\Omega_R^0 \cos(\omega_L t)(\rho_{11} - \rho_{22}) - \gamma_2 \rho_{21}$$

where $\hbar\omega_{21}$ is the energy separation of the ground and excited states, i.e. states $|0\rangle$ and $|X\rangle$, in the notation of Zrenner et al. [10], with ρ_{11} and ρ_{22} being the corresponding number occupations of the two levels. Here Ω_R^0 is the Rabi frequency, defined as

$$\Omega_R = \frac{\mathbf{d}_{cv} \cdot \mathbf{E}}{\hbar} = \frac{\mathbf{d}_{cv} \cdot \mathbf{E}_0}{\hbar} \cos(\omega_L t) = \Omega_R^0 \cos(\omega_L t) \quad (7)$$

where \mathbf{d}_{cv} is the conduction-valence dipole matrix element and ω_L is the laser frequency. For the two-level excitonic system, γ_1 and γ_2 represent the recombination rate and the total dephasing rate, respectively. The latter includes both intrinsic dephasing and inhomogeneous broadening of the two-level exciton states (empty and one-exciton occupancy), e.g. due to $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ QD monolayer fluctuations either in composition or structure size, or related to indirect excitation of electrons or holes to or from the continuum of spatially delocalized states associated either to the InAs wetting layer or the GaAs barrier [16]. The γ_2 recombination rate may be interpreted as related to the coupling of the $|0\rangle$ state with a broadened $|X\rangle$ excitonic band, and one, therefore, would obtain the field-dependent behavior of Eq. (5), as in the previously discussed donor two-level case. By solving the density matrix Bloch equations, we have calculated a field-dependent γ_2 dephasing rate (Fig. 2(a)) which gives the appropriate Rabi oscillations found in the experimental measurements by Zrenner et al. [10]. Notice, from Fig. 2(a), that the theoretical γ_2 is essentially quadratic in the field [the Rabi frequency Ω_R^0 is linear in the field, see Eq. (7)]. Present calculations are performed by using the optical Bloch equations with γ_1 corresponding [10] to a lifetime of 1 ns. We calculate the photocurrent at resonance, for a pulse width of 1 ps, for increasing excitation amplitudes A_{exc} , which is the square root of the peak pulse intensity (coherent π -pulse excitation corresponds to $A_{\text{exc}} = 1$). Fig. 2(b) then shows the Rabi oscillations in a two-level excitonic system corresponding to the experimental measurements [10] and taken on the main resonance at about -1.04 V, together with the present theoretical results (full line). It is clear that the proposed γ_2 dependence on the laser-field amplitude leads to a very good fitting to the experimental data.

It is important to mention that Stievater et al. [9] have used transient non-linear optical spectroscopy in a study on excitons confined to single GaAs QDs, and observed oscillations which

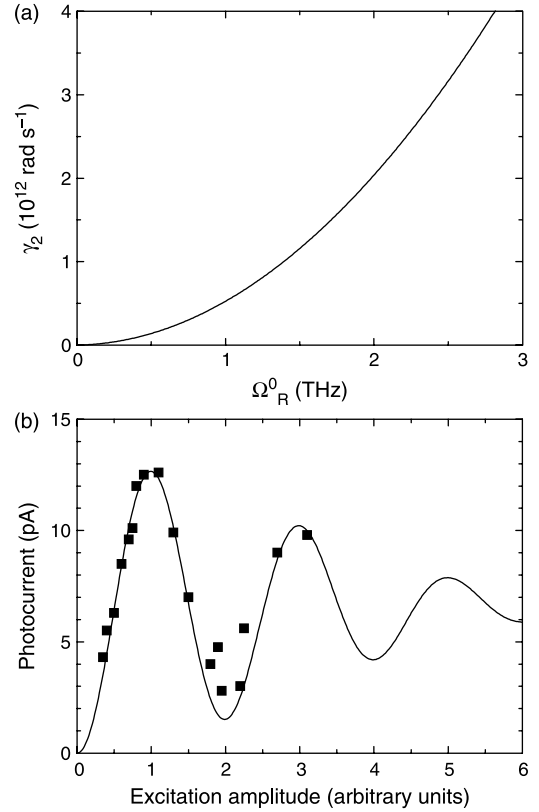


Fig. 2. (a) Calculated γ_2 total dephasing rate, associated to the coupling of the $|0\rangle$ state with a broadened $|X\rangle$ excitonic band. The γ_2 recombination rate includes both intrinsic dephasing and inhomogeneous broadening of the exciton band in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ two-level QD system; (b) Rabi oscillations in a two-level excitonic system: photocurrent at resonance for increasing excitation amplitudes A_{exc} , which is the square root of the peak pulse intensity. Coherent π -pulse excitation corresponds to $A_{\text{exc}} = 1$. Full squares are experimental data [10] for a pulse width of about 1 ps, whereas the full curve corresponds to the present theoretical results.

are analogous to Rabi oscillations in two-level atomic systems. They commented, however, that a simple two-level model would not predict the observed decay of the oscillations with increasing pulse area. In a further work, by the same authors [12], it was argued that the physical origin for an increased decay rate at higher pump power is a higher scattering rate experienced by the resonantly excited excitons and biexcitons from the nearly degenerate delocalized states. However, Wang et al. [17] have performed experiments, in self-assembled $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ QDs, with photoluminescence spectroscopy and using circularly polarized light to suppress biexciton excitation, and argued that experimental data showed almost identical intensity-dependent behavior and, therefore, ruled out the possibility of biexcitonic excitation [18]. Förstner et al. [19] have provided a theoretical description of phonon-induced dephasing and found Rabi oscillations in GaAs QDs with damping depending on the input pulse strength, although

their results do not provide agreement with the experimental measurements by Zrenner et al. [10] for short pulses, and the pulse-width dependence is inconsistent with experimental data by Wang et al. [17]. Here we note that Zrenner et al. [10] and Beham et al. [11] have pointed out that the exact nature of the underlying dephasing mechanism, which leads to the observed damped Rabi oscillations in $\text{In}_x\text{Ga}_{1-x}\text{As}$ two-level excitonic systems, remains an open question, i.e. that the reason for the observed power-dependent damping of the Rabi oscillations with increasing A_{exc} is unknown so far. Of course, an ab initio calculation of field-dependent recombination rates for a real semiconductor system would be a formidable task due to the complexity of solid-state environments.

In conclusion, the present results indicate that the nature underlying the dephasing mechanism, which remained an open question [10,11], may be associated with the inhomogeneous broadening of the exciton lines in the $\text{In}_x\text{Ga}_{1-x}\text{As}$ QD and may be explained within a simple two-level QD system. We have shown that, by considering a dephasing rate exhibiting the same kind of field-dependent (or laser intensity) of Eq. (5), it is possible to properly fit the excitonic photocurrent data. Moreover, we have suggested that the underlying mechanism is related to the fact that the broadened excitonic states may be interpreted as associated to monolayer fluctuations in composition or structure sizes, or to indirect excitation of electrons or holes to or from the continuum of delocalized states, e.g. in the wetting layer [20]. Summing up, we would like to stress that the present field-dependent recombination rate, within the approximation of a two-level system, is in quite good agreement with experimental measurements for Rabi oscillations in two-level semiconductor systems, such as donor states in bulk GaAs and excitonic QD systems.

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