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Terahertz excitation and coherence effects of two-level donor systems in GaAs quantum dots

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Abstract

The confinement effects on the electronic and donor states in GaAs–(Ga, Al)As quantum dots, in the presence of an applied magnetic field, are studied. Using the optical Bloch equations with damping, we study the time evolution of the 1s, $2p_-$, and $2p_+$ states in the presence of an applied magnetic field and of a terahertz laser. We discuss the conditions to obtain a bound $2p_+$ state in contrast to the bulk situation, where the $2p_+$ state is resonant with the continuum states (in this case, decoherence may be a serious problem for practical realizations). We show that the pronounced confining effects of semiconductor quantum dots lead to conditions such that the $2p_+$ excited donor state may lie below the continuum, originating a favourable situation concerning the coherence time of the corresponding Rabi oscillations. We also calculate the electric dipole transition moment, discuss the photosignal, and the possible experimental conditions under which decoherence is weak and qubit operations are efficiently controlled.

1. Introduction

Recently, there have been many new proposals concerning solid-state quantum computers due to their great advantage in manipulating a large number of qubits as compared to atomic and molecular systems. Among them, several studies consider quantum dots (QDs) as the basic architecture for their implementation, in which discrete electronic charge or spin states are the qubits responsible for encoding quantum information [1–12]. The qubits may be manipulated via external electric and/or magnetic fields, or by electromagnetic radiation, and the information between qubits can be transferred through electron–electron Coulomb interaction [1–3], by optical-cavity modes [6, 7] or manipulating coherent laser radiation [10]. Furthermore, the basic characteristics required for an ideal model qubit are the existence of a precisely defined twofold Hilbert space, that the model-qubit system operates under the

conditions that decoherence processes are weak and finally where single-qubit and two-qubit unitary operations are controlled. This implies that a quantum computer would be effective only if the decoherence times are much longer than the time involved in the single- and twoqubit operations. If these operations are controlled by switching external gate potential and/or magnetic fields, this could pose a problem since they cannot be performed very quickly. The use of a laser pulse to control the qubit operations may overcome this possible limitation. Cole *et al* [5] and Zrenner *et al* [8] have demonstrated that the coherent optical excitations in two-level donor systems in bulk GaAs and QD two-level excitonic systems, respectively, may be converted into deterministic photocurrents. Although Cole *et al* [5] have made a quite interesting proposal, decoherence may be a serious problem for its practical realization. In this case, the 1s and $2p_+$ donor states in GaAs (under applied magnetic fields) are the states of a model qubit coherently manipulated by laser radiation. It has been shown by Brandi et al [12] that if the excited donor state lies below the continuum, it is more robust to excitation by photons leading to a favourable situation concerning the coherence time. As is well known, low-dimensional heterostructures such as semiconductor QDs exhibit pronounced confining effects. This makes donor-doped QDs natural candidates for both theoretical [11, 12] and experimental investigations. In the present work we investigate the confinement effects of a model spherical QD in the presence of an applied magnetic field on the electronic and oncentre donor states in GaAs-(Ga, Al)As QDs. We study the conditions in which one may obtain a bound $2p_+$ state in contrast to the resonant one in the study by Cole *et al* [5], and using the optical Bloch equations with damping terms, we study the time evolution of the 1s, $2p_+$ and $2p_-$ (which lies below the continuum) donor states in the presence of an applied magnetic field and under the action of a terahertz laser. We also discuss their influence on the photocurrent, calculate the electric dipole transition moment and discuss the role of the QD electronic confinement on decoherence processes.

2. Theoretical framework

In the effective-mass approximation, the on-centre donor Hamiltonian in a spherical GaAs-(Ga, Al)As QD is given by

$$H = -\nabla^2 + \gamma l_z + \gamma^2 \rho^2 / 4 + V_{\rm b}(r) - 2/r, \tag{1}$$

where $l_z = \frac{1}{i} \frac{\partial}{\partial \phi}$, $V_b(r)$ is the QD barrier potential, $\gamma = \frac{e\hbar B}{2m^*cR^*} = \mu_B^* B/R^* = (a_0^*/l_B)^2$ is the ratio of the magnetic and Coulomb energies (for donors in GaAs, $\gamma = 1$ corresponds to an applied magnetic field of ≈ 6.9 T), $R^* \approx 5.9$ meV is the GaAs donor effective Rydberg, $l_B = (\hbar c/eB)^{1/2}$ is the magnetic length (or cyclotron radius), and a_0^* and μ_B^* are the effective Bohr radius and effective Bohr magneton, respectively. Using hydrogenic-like envelope wavefunctions, the 1s and $2p_{\pm}$ energies may then be variationally [12, 13] obtained as a function of the z-direction applied magnetic field.

The time evolution of the elements of the density matrix within a two-level model for the donor-QD system are obtained via standard procedures [11, 12, 14], from the set of optical Bloch equations, i.e.,

$$\frac{d\rho_{11}}{dt} = -i\Omega_{R}\cos(\omega_{L}t)(\rho_{21} - \rho_{12}) + \gamma_{1}\rho_{22}
\frac{d\rho_{22}}{dt} = +i\Omega_{R}\cos(\omega_{L}t)(\rho_{21} - \rho_{12}) - (\gamma_{1} + \gamma_{3})\rho_{22}
\frac{d\rho_{12}}{dt} = +i\omega_{21}\rho_{12} + i\Omega_{R}\cos(\omega_{L}t)(\rho_{11} - \rho_{22}) - \gamma_{2}\rho_{12}
\frac{d\rho_{21}}{dt} = -i\omega_{21}\rho_{21} - i\Omega_{R}\cos(\omega_{L}t)(\rho_{11} - \rho_{22}) - \gamma_{2}\rho_{21}$$
(2)

where $\omega_{\rm L}$ is the terahertz laser frequency, and ω_{21} is the energy separation of the 1s and $2p_{\pm}$ impurity levels. The parameters γ_1 , γ_2 , and γ_3 are recombination rates as introduced phenomenologically in Cole *et al* [5]. The decay parameter γ_1 gives the rate of spontaneous emission of photons due to $2p_{\pm} \rightarrow 1$ s transitions, and may be shown to be negligible as compared with γ_2 or γ_3 , in the case when the $2p_{\pm}$ state is immersed in the continuum [5]. If the bound excited state is separated from the continuum, the ionization rate will decrease significantly, giving $\gamma_3 \ll \gamma_2$ and allowing γ_3 to be neglected. The recombination rates γ_2 and γ_3 represent the total dephasing rate, which includes both intrinsic dephasing and inhomogeneous broadening of the transition energy, and the ionization rate of the $2p_{\pm}$ state, respectively [5, 11, 12]. The decay parameters may be obtained within a simple model essentially describing the coupling of a discrete level to a continuum set of states of finite width, as analysed by Cohen-Tannoudji *et al* [14]. Such a study, however, is beyond the scope of the present work.

3. Results and discussion

Results for bulk GaAs and GaAs–Ga_{0.7}Al_{0.3}As spherical QDs, for R = 220 and 400 Å, are shown in figure 1, which displays the magnetic-field dependence of the energies of donor states 1s, $2p_{-}$, $2p_{+}$, and of $\epsilon_c = \epsilon_0 + \gamma$. We have chosen the bulk GaAs conduction-band edge as the origin of energy. Notice that, in the bulk case, $\epsilon_0 = 0$, whereas for QD calculations ϵ_0 corresponds to the energy of the lowest unoccupied state of the bare QD. This figure also shows the $1s-2p_+$ (or $1s-2p_-$) transition energies corresponding to 2.52 THz, which is the free-electron laser frequency used in the experimental measurements by Cole et al [5]. In figure 1(a) it is clear that for the value of the magnetic field, ≈ 3.4 T, used to tune the terahertz radiation to the $1s-2p_+$ transition, the bulk $2p_+$ state is **resonant** with the continuum states⁵. This is a source of decoherence due to ionization by photons and phonons processes [9, 12]. As shown in figure 1(b), the confinement effects due to the QD, for R = 400 Å, are such that a magnetic field of ≈ 3.0 T tunes the terahertz radiation to the corresponding 1s-2p₊ transition, and this source of decoherence is removed. In figure 1(c) we show results for a QD of radius R = 220 Å. The confinement effects are more pronounced than in the preceding situations, as expected, and the 2p₊ state is above the first Landau level even when no magnetic field is applied. Also notice that now the 2.52 THz free-electron radiation is tuned to the $1s-2p_{-}$ transition for a magnetic field of ≈ 6.2 T.

The time evolution of the elements of the density matrix within a two-level model for the donor-QD system are obtained from the set of optical Bloch equations, with recombination rates introduced phenomenologically which may be theoretically evaluated by standard procedures [12, 14]. Once the variational calculation for the two-level donor states is performed, one may use the 1s and $2p_{\pm}$ hydrogenic-like wavefunctions to calculate both the *x*-component of the corresponding $1s-2p_{\pm}$ dipole matrix element, $d_{12}^x = \langle 1s|x|2p_{\pm}\rangle$, and the Rabi frequency $\Omega_R = E_{\text{THz}} d_{12}^x / \hbar$, where E_{THz} is the amplitude of the terahertz electric field (in the *x*-direction). Figure 2 shows the $\langle 1s|x|2p_{\pm}\rangle$ matrix elements as a function of the applied magnetic field, and figure 3 presents the terahertz electric-field dependence of the Rabi frequency for a fixed applied magnetic field, for bulk GaAs, and GaAs-Ga_{0.7}Al_{0.3}As spherical QDs of different radii. The d_{12}^x matrix-elements results for a GaAs-Ga_{0.7}Al_{0.3}As spherical QD of radius R = 1500 Å are essentially the same as for bulk GaAs, as expected. In the bulk regime, for small values of applied magnetic fields, the $\langle 1s|x|2p_{\pm}\rangle$ matrix elements increase

⁵ One should notice that the 1s and $2p_+$ energy results shown in figure 1(a) by Brandi *et al* [12] are, by mistake, shifted upwards by an energy corresponding to γ .



Figure 1. Magnetic-field dependence of the energies of donor states 1s, $2p_-$, $2p_+$, and $\epsilon_c = \epsilon_0 + \gamma$. The energy hv_L for a 2.52 THz free-electron laser is shown as $1s-2p_{\pm}$ transitions. Results correspond to the cases of (a) bulk GaAs, (b) R = 400 Å and (c) R = 220 Å Ga_{0.7}Al_{0.3}As spherical QDs.

with increasing magnetic fields which can be related to the magnetic-field confinement effects being stronger for the $2p_{\pm}$ state as compared to the 1s state. This leads to a larger overlap between 1s- and $2p_{\pm}$ -like wavefunctions and therefore to a larger value of the d_{12}^x matrix elements. One notices the existence of a maximum around 2–3 T, which may be traced back to the fact that, with increasing values of the magnetic field, the Landau magnetic length and Bohr radius (i.e., magnetic and Coulomb energies) become comparable. On the other hand, in the case of a QD, the impurity states are very much confined by the QD potential; as the magnetic field increases, the 1s- and $2p_{\pm}$ -like donor states become somewhat more localized, which leads to a smooth decrease of the $\langle 1s|x|2p_{\pm}\rangle$ matrix elements. Of course, the smaller the QD radius, the larger its confinement effects, and the smoother is the decrease, as may be noticed by a comparison between results for the R = 400 and 220 Å QDs.

The dependence of the Rabi frequencies on the terahertz electric field is shown in figure 3 for bulk GaAs, and for R = 400 and 220 Å GaAs–Ga_{0.7}Al_{0.3}As spherical QDs. Calculated



Figure 2. Magnetic-field dependence of the matrix elements $\langle 1s|x|2p_{\pm} \rangle$ for bulk GaAs (dotted lines), and different GaAs–Ga_{0.7}Al_{0.3}As spherical QDs.



Figure 3. Theoretical Rabi frequencies as functions of the terahertz electric field for bulk GaAs, and GaAs–Ga_{0.7}Al_{0.3}As spherical QDs of radii R = 220 and 400 Å. Notice that each one of the curves corresponds to different values of applied magnetic fields, i.e., results are shown for 1s–2p_± transitions tuned to a 2.52 THz free-electron laser.

results are for $1s-2p_{\pm}$ transitions tuned to a 2.52 THz free-electron laser, and correspond to different values of the magnetic fields as shown in figure 1. By following the set of optical Bloch equations associated with the two-level model we calculate the time evolution of the photosignal corresponding to the $1s-2p_{+}$ transitions. In what follows, we choose $\gamma_1 = 0$, $\gamma_2 = 2.7 \times 10^{11}$ rad s⁻¹, and $\gamma_3 = 1.4 \times 10^{11}$ rad s⁻¹, corresponding to the $1s-2p_{+}$ recombination rate, the total dephasing rate of the transition energy, and the $2p_{+}$ ionization rate, respectively. The above values essentially correspond to fit parameters obtained by Cole *et al* [5] in their GaAs bulk experiment with terahertz radiation, for a terahertz electric field of 3.0×10^4 V m⁻¹. We use a theoretical value of 2.9×10^{11} rad s⁻¹ (see figure 3) for the bulk GaAs Rabi frequency at this value of terahertz electric field. Also, we have taken



Figure 4. Theoretical 1s–2p₊ Rabi oscillations of the photosignal, at resonance, and for a fixed terahertz electric field. Results are shown for bulk GaAs (full lines), and R = 400 Å GaAs–Ga_{0.7}Al_{0.3}As spherical QDs (dot–dashed, dotted, and dashed curves) for different γ_2 total dephasing rates (see the text for details).

the photosignal as proportional to $1 - \rho_{11}$, as in the work of Cole *et al* [5]. If the excited state lies in the conduction band, the photosignal may be measured through photoconductivity experiments [5], whereas in the case when the excited state is isolated from the conduction band, one needs to use techniques such as differential transmission methods [15], where the signal is proportional to the level of excitation induced by the laser. The results shown in figure 4 are calculated, at resonance, for the GaAs bulk (solid curve, $B \approx 3.4$ T) and for an R = 400 Å GaAs-Ga_{0.7}Al_{0.3}As spherical QD ($B \approx 3.0$ T). For the case of the R = 400 Å QD, $\Omega_R = 2.5 \times 10^{11}$ rad s⁻¹ (see figure 3), and we set $\gamma_3 = 0$, as the 2p₊ level is below the first Landau level (see figure 1(b)). Different dephasing rates are chosen for the recombination rate, i.e., $\gamma_2 = 2.7 \times 10^{11}$ rad s⁻¹ (dashed curve). This last value of γ_2 is consistent with the estimate obtained from the linear spectra [5] as well as with values obtained from farinfrared measurements [16]. Of course, a smaller γ_2 dephasing rate should be expected if the excited 2p₊-like donor state lies below the first Landau level, as mentioned previously [5]. One clearly notices that the displayed Rabi oscillations are more robust as the γ_2 dephasing rate diminishes, corresponding to a favourable coherence time so that qubit operations may be efficiently controlled.

4. Conclusions

In conclusion, we have calculated the electric dipole transition moment, and discussed the possible experimental conditions under which decoherence is weak and possible qubit operations are efficiently controlled. We have shown that, using the optical Bloch equations with damping, we are able to discuss, in a phenomenological manner, the coherence effects on Rabi oscillations associated to donor states confined in GaAs–(Ga, Al)As QDs in the presence of an applied magnetic field and of a terahertz laser. Also, it was shown that the pronounced confining effects of semiconductor QDs lead to conditions such that the 2p₊ excited donor state may lie below the continuum, originating a favourable situation concerning the coherence time of the corresponding Rabi oscillations.

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