Phys. Status Solidi B **249**, No. 5, 914–917 (2012) / **DOI** 10.1002/pssb.201100100

physica status solidi b
www.pss-b.com
basic solid state physics

Electronic properties of asymmetrical quantum dots dressed by laser field

O. V. Kibis*[,1], G. Ya. Slepyan[2], S. A. Maksimenko[2], and A. Hoffmann[3]

[1] Department of Applied and Theoretical Physics, Novosibirsk State Technical University, Karl Marx Avenue 20, 630092 Novosibirsk, Russia
[2] Institute for Nuclear Problems, Belarus State University, Bobruyskaya St. 11, 220050 Minsk, Belarus
[3] Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstraße 36, 10623 Berlin, Germany

Received 16 November 2011, accepted 1 December 2011
Published online 30 January 2012

Keywords electron–photon coupling, quantum dots

* Corresponding author: e-mail oleg.kibis@nstu.ru, Phone: +7-383-3460488, Fax: +7-383-3460209

In the present paper, we demonstrate theoretically that the strong non-resonant interaction between asymmetrical quantum dots (QDs) and a laser field results in harmonic oscillations of their band gap. It is shown that such oscillations change the spectrum of elementary electron excitations in QDs: in the absence of the laser pumping there is only one resonant electron frequency, but QDs dressed by the laser field have a set of electron resonant frequencies. One expects that this modification of elementary electron excitations in QDs can be observable in optical experiments.

1 Introduction The interaction of quantum systems with strong electromagnetic fields [1, 2] is permanently in the focus of interest, due to both the high methodological value of arising problems and their direct relation to current applied projects, such as a new generation of high-efficiency lasers [3], laser cooling of atoms [2], development of a basis for quantum information processing [4], etc. One of the bright manifestations of the strong field–matter coupling is the Rabi effect [2]: oscillations of the level population in a quantum system exposed to a monochromatic electromagnetic wave. The simplest physical model leading to harmonic Rabi oscillations is a two-level symmetrical quantum system placed in a given classical single-mode electromagnetic field [2]. Incorporation into this simplest model of additional physical factors results in many non-trivial effects. For example, accounting for the quantum nature of light leads to the concept of radiation-dressed atoms [1] and the "collapse–revival" phenomenon in the population dynamics of a system exposed to coherent light. Time-domain modulation of the field–matter coupling constant [5], local-field effects in nanostructures [6, 7], and phonon-induced dephasing [8, 9] provide new possibilities for the control of the Rabi oscillation dynamics. Many interesting effects manifest themselves in more complex systems, such as two coupled Rabi oscillators [10], where the Rabi effect is observed between the ground state and a two-electron entangled state, and one-dimensional chains of Rabi oscillators, where the phenomenon of spatial propagation of Rabi oscillations – Rabi waves – is predicted to exist [11, 12]. Among others, we mention also the Rabi effect in superconducting electrical circuits [4], in oscillators coupled to a nanoantenna resonance [13], and in systems where Rabi oscillations are strongly influenced by intraband motion of quasi-particles [14, 15]. In the given paper, we investigate the role in the Rabi effect of a new physical factor – violation of the inversion symmetry. Inherent in many quantum systems (semiconductor nanostructures, asymmetrical molecules, etc.), the factor is ignored by the conventional physical model [2] of Rabi oscillations. Meanwhile, our theoretical analysis predicts a pronounced manifestation of the violation in a set of observable effects.

Let us consider a two-level quantum system with $|a\rangle$ and $|b\rangle$ as excited and ground states, respectively. Let the system interact with a classical linearly polarized monochromatic electromagnetic field (a driving field). Such a system can be described by the Hamiltonian

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{\text{int}}, \quad (1)$$

where the free-particle Hamiltonian, written in the basis of these two states, is given by

$$\hat{\mathcal{H}}_0 = \frac{\hbar\omega_0\hat{\sigma}_z}{2}, \quad (2)$$

where $\hat{\sigma}_z$ is the Pauli matrix and ω_0 is the resonant frequency of the two-level system. The interaction Hamiltonian, $\hat{\mathcal{H}}_{\rm int}$, is expressed in terms of the amplitude $\mathbf{E}_\omega$ and frequency ω of the driving field by:

$$\hat{\mathcal{H}}_{\rm int} = -\mathbf{E}_\omega \hat{\mathbf{d}} \cos(\omega t),$$

where $\hat{\mathbf{d}}$ is the electric dipole moment operator and $\mathbf{d}_{ij} = \langle i|\hat{\mathbf{d}}|j\rangle$ are its matrix elements. The critical assumption, which distinguishes the systems being considered from standard ones, is the violation of the inversion symmetry. Since in that case the states $|a\rangle$ and $|b\rangle$ do not possess a certain spatial parity, the diagonal matrix elements of the dipole moment operator prove to be non-equivalent, $\mathbf{d}_{aa} \neq \mathbf{d}_{bb}$, dictating thus the physical effects described hereafter. Confined semiconductor nanostructures with discrete energy spectrum and linear extension multiply exceeding the atomic size, i.e., quantum dots (QDs) [3], seem to be most prospective for observation of these effects. In QDs, the driving electromagnetic field can transfer electrons from the valence band into the conduction band; that is why in QDs the ground state $|b\rangle$ corresponds to the absence of free carriers while the first excited state $|a\rangle$ is the state with electrons in the conduction band and holes in the valence band. In that case, $\hbar\omega_0$ is approximately equal to the semiconductor band gap. Among a variety of different types of QDs, nitride-based confined structures seem to be most promising for the observation of the effects. Indeed, whereas the usual semiconductor III–V compounds have a cubic (zinc-blende) crystalline structure, GaN and similar group-III nitride alloys have a hexagonal (wurtzite) structure. For a consistent set of parameters of III-nitrides AlN, GaN, and InN, see Ref. [16]. As a consequence of the giant piezoelectric effect inherent in hexagonal crystals, QDs based on AlN/GaN and InN/GaN structures have a strong built-in strain-induced electric field with strength of several MV/cm [17, 18]. Due to the strong electric field, conduction-band electrons and valence-band holes in the QDs get spatially separated [19, 20], and the effective dipole moment of a III-nitride QD is given by the simple relation $|\mathbf{d}_{aa} - \mathbf{d}_{bb}| \sim el$, where l is the QD height. Then, for a typical l of several nanometres, the effective dipole moment of nitride QDs is estimated as a giant value $|\mathbf{d}_{aa} - \mathbf{d}_{bb}| \sim 10$ Debye.

In the general case, the solution of the Schrödinger equation with the Hamiltonian (1) has the form

$$|\psi\rangle = C_a(t)|a\rangle + C_b(t)|b\rangle.$$

Substituting this expression into the Schrödinger equation, we obtain the system of two equations:

$$i\hbar\dot{C}_a = \left[\frac{\hbar\omega_0}{2} - \mathbf{E}_\omega \mathbf{d}_{aa}\cos(\omega t)\right]C_a - \mathbf{E}_\omega \mathbf{d}_{ab}\cos(\omega t)C_b, \quad (3)$$

$$i\hbar\dot{C}_b = \left[-\frac{\hbar\omega_0}{2} - \mathbf{E}_\omega \mathbf{d}_{bb}\cos(\omega t)\right]C_b - \mathbf{E}_\omega \mathbf{d}_{ba}\cos(\omega t)C_a. \quad (4)$$

The solution of the system of equations Eqs. (3) and (4), which corresponds to the resonant interaction of a QD with a driving electromagnetic field, has been analyzed in Ref. [21]. Particularly, it has been shown that the resonance in the QD takes place if the driving field frequency obeys the condition

$$\omega = \frac{\omega_0}{n}, \quad (5)$$

with n an integer [21]. In the present paper, we will consider the case of non-resonant interaction between the QD and the driving field: further, we assume that the driving field frequency ω lies far away from the resonant frequencies defined by Eq. (5). In this case, the field-induced mixing of states $|a\rangle$ and $|b\rangle$ can be neglected, which reduces the system of equations Eqs. (3) and (4) to two independent equations:

$$i\hbar\dot{C}_a = \left[\frac{\hbar\omega_0}{2} - \mathbf{E}_\omega \mathbf{d}_{aa}\cos(\omega t)\right]C_a, \quad (6)$$

$$i\hbar\dot{C}_b = \left[-\frac{\hbar\omega_0}{2} - \mathbf{E}_\omega \mathbf{d}_{bb}\cos(\omega t)\right]C_b. \quad (7)$$

As can easily be verified by the direct substitution into Eqs. (6) and (7), solutions of Eqs. (6) and (7) are given by

$$C_a(t) = \mathrm{e}^{-i\omega_0 t/2}\exp\left\{\frac{i\mathbf{E}_\omega \mathbf{d}_{aa}\sin(\omega t)}{\hbar\omega}\right\}, \quad (8)$$

$$C_b(t) = \mathrm{e}^{i\omega_0 t/2}\exp\left\{\frac{i\mathbf{E}_\omega \mathbf{d}_{bb}\sin(\omega t)}{\hbar\omega}\right\}. \quad (9)$$

As a result, the time evolution of the states $|a\rangle$ and $|b\rangle$ can be described by the time-dependent wave functions

$$|\psi_a\rangle = \mathrm{e}^{-i\omega_0 t/2}\exp\left\{\frac{i\mathbf{E}_\omega \mathbf{d}_{aa}\sin(\omega t)}{\hbar\omega}\right\}|a\rangle, \quad (10)$$

$$|\psi_b\rangle = \mathrm{e}^{i\omega_0 t/2}\exp\left\{\frac{i\mathbf{E}_\omega \mathbf{d}_{bb}\sin(\omega t)}{\hbar\omega}\right\}|b\rangle, \quad (11)$$

which physically correspond to the field-induced oscillations of the band gap:

$$\varepsilon_g(t) = \hbar\omega_0 - \mathbf{E}_\omega(\mathbf{d}_{aa} - \mathbf{d}_{bb})\cos(\omega t).$$

Using the wave functions (10)–(11), we can find the optical absorption spectrum of the considered QD.

Let the QD dressed by the driving laser field be exposed to a probing field – a linearly polarized electromagnetic field with the frequency Ω and the amplitude $\mathbf{E}_\Omega$. The probing field induces optical transitions between the states (10) and (11). Though the wave functions of these states (10)–(11) are time dependent, they form a complete orthogonal basis of the QD at any point of time. Therefore, the wave function of the QD can be written as $c_a(t)|\psi_a\rangle + c_b(t)|\psi_b\rangle$. If the probing field is weak enough, we can find coefficients $c_a(t)$ and $c_b(t)$, considering the Hamiltonian

$$\hat{V} = -\mathbf{E}_\Omega \hat{\mathbf{d}} \cos(\Omega t) \quad (12)$$

as a perturbation within the conventional non-stationary perturbation theory [22]. If the QD is in the state $|b\rangle$ at the time $t = -\tau/2$, we have $c_b(-\tau/2) = 1$ and $c_a(-\tau/2) = 0$. Then the coefficient $c_a(\tau/2)$ is described by the well-known expression [22]:

$$c_a\left(\frac{\tau}{2}\right) = -\frac{i}{\hbar}\int_{-\tau/2}^{\tau/2} \langle\psi_a|\hat{V}|\psi_b\rangle \, \mathrm{d}t. \quad (13)$$

Substituting the perturbation Hamiltonian (12) and the wave functions (10)–(11) into the expression (13), we arrive at the transition probability:

$$P(\tau) = \left|c_a\left(\frac{\tau}{2}\right)\right|^2 = \frac{1}{\hbar^2}\left|\mathbf{E}_\Omega \mathbf{d}_{ab}\int_{-\tau/2}^{\tau/2} \cos(\Omega t)\, \mathrm{e}^{i\omega_0 t}\, \mathrm{e}^{-i\kappa \sin(\omega t)}\, \mathrm{d}t\right|^2, \quad (14)$$

where the parameter $\kappa = \mathbf{E}_\omega(\mathbf{d}_{aa} - \mathbf{d}_{bb})/\hbar\omega$ describes the asymmetry of the QD [21]. It is well known that:

$$\mathrm{e}^{-i\kappa \sin(\omega t)} = \sum_{n=-\infty}^{\infty} J_n(\kappa)\, \mathrm{e}^{in\omega t},$$

where $J_n(\kappa)$ is the Bessel function of the first kind. Substituting this expression into Eq. (14) and taking into account that the δ-function can be written as

$$\delta(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{e}^{i\omega t}\, \mathrm{d}t,$$

we find the transition probability (14) as

$$P(\tau) = \left(\frac{\pi \mathbf{E}_\Omega \mathbf{d}_{ab}}{\hbar}\right)^2 \times \left|\sum_{n=-\infty}^{\infty} J_n(\kappa) \times [\delta(n\omega + \omega_0 + \Omega) + \delta(n\omega + \omega_0 - \Omega)]\right|^2 \quad (15)$$

for $\tau \to \infty$. Let the frequencies of the driving and probing fields satisfy the conditions $\omega \neq 2\omega_0/m$, $\omega \neq 2\Omega/m$, and $\omega \neq 2|\omega_0 \pm \Omega|/m$ with m an integer. Then Eq. (15) takes the form:

$$P(\tau) = \left(\frac{\pi \mathbf{E}_\Omega \mathbf{d}_{ab}}{\hbar}\right)^2 \sum_{n=-\infty}^{\infty} J_n^2(\kappa) \times \left[\delta^2(n\omega + \omega_0 + \Omega) + \delta^2(n\omega + \omega_0 - \Omega)\right]. \quad (16)$$

Since for $\tau \to \infty$ we can represent the square of the δ-function as:

$$\delta^2(n\omega + \omega_0 \pm \Omega) = \delta(n\omega + \omega_0 \pm \Omega)\delta(0) = \delta(n\omega + \omega_0 \pm \Omega)\frac{1}{2\pi}\int_{-\tau/2}^{\tau/2} \mathrm{e}^{it\times 0}\, \mathrm{d}t = \delta(n\omega + \omega_0 \pm \Omega)\frac{\tau}{2\pi}.$$

Equation (16) allows us to get the probability of optical transition per unit time:

$$w = \frac{\partial P(\tau)}{\partial \tau} = \frac{\pi}{2}\left(\frac{\mathbf{E}_\Omega \mathbf{d}_{ab}}{4\hbar}\right)^2 \sum_{n=-\infty}^{\infty} J_n^2(\kappa) \times [\delta(n\omega + \omega_0 + \Omega) + \delta(n\omega + \omega_0 - \Omega)]. \quad (17)$$

From Eq. (17), it follows that the QD can absorb the probing field at the frequencies:

$$\Omega = |n\omega + \omega_0|, \quad (18)$$

where $n = \ldots -1, 0, 1, \ldots$.

Thus, elementary excitations in asymmetrical QDs, dressed by a strong non-resonant laser field, are allowed in the broad frequency range at frequencies determined by the condition (18). This novel effect is due to the strong electron–photon coupling and can be observed in asymmetrical QDs using conventional methods of optical measurements. Another type of asymmetrical two-level quantum system where the predicted effect is expected to be observable is superconducting quantum qubits. Away from the charge degeneracy point of the superconducting quantum circuits [4] formed by Josephson qubits in micro-strip resonators, the qubit symmetry is broken, and the Hamiltonian of the system takes the form analogous to that used in our analysis. Chiral nanostructures, including chiral carbon nanotubes [23–26] and nanohelices [27–33], can also be mentioned as prospective systems for the observation of the effect, since the chirality breaks the inversion symmetry. To conclude, let us stress that violation of the inversion symmetry is a common property of quantum oscillators of different physical origins and, consequently, the effect predicted is expected to manifest itself in spectral characteristics of various optical processes.

Acknowledgements The research was partially supported by the DFG (Germany) project HO 1366/23-1, the FP7 projects FP7-230778 TERACAN and FP7-266529 BY-NanoERA, the RFBR (Russia) projects 10-02-00077 and 12-02-90002, the Russian Ministry of Education and Science, the Federal goal-oriented program "Scientific and scientific-educational personnel of innovative Russia," and the BRFFI (Belarus) project F10-002.

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