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Coulomb-interaction driven anomaly in the Stark effect for an exciton in vertically coupled quantum dots

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Abstract

The effect of the electric field on an exciton confined in a pair of vertically coupled quantum dots is studied. We use a single-band approximation and a parabolic model potential. As a result of these idealizations, we obtain a numerically solvable model, which is used to describe the influence of the electron–hole interaction on the Stark effect for the lowest-energy photoluminescence lines. We show that for intermediate tunnel coupling between the dots this interaction leads to an anomalous Stark effect with an essential deviation of the recombination energy from the usual quadratic dependence on the electric field.
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Self-assembled InAs/GaAs quantum dots grown on subsequent layers stack one above the other [1] which results in the spontaneous formation of artificial molecules. Tunnel coupling between the quantum dots leads to a shift of the photoluminescence lines as a function of the interdot distance [1–3]. Photocurrent measurements of the Stark effect [4] provides information on the electronic structure of the buried quantum dots. Measurements of the electric-field dependence of the photoluminescence spectrum of the coupled dots should provide data useful for the description of the interdot coupling. Experimental work [5] in this direction is currently under way. Preliminary data [5] show that the interdot coupling essentially changes the character of the Stark effect.

In this paper, we investigate the role of the electron–hole interaction in the Stark shift of the photoluminescence lines. For this purpose we use a model of coupled quantum dots, which—due to its

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simplicity—allows for an exact numerical solution of the Schrödinger equation in the single-band approximation. Previously, the ground-state Stark shift was studied within the Hartree approach [6]. The ground-state Stark shift has also been calculated within the $\mathbf{k}\cdot\mathbf{p}$ method [7], which however did not include the electron–hole interaction. For the intermediate barrier thickness the Coulomb interaction leads to a deviation of the dependence of the ground-state photoluminescence line on the external electric field (F) from its usual quadratic form [8] $\Delta E = pF + \beta F^2$, where p is the electric dipole moment and β the polarizability of the exciton. A physical understanding of this deviation requires a calculation of the exciton excited states.

We assume a parabolic lateral confinement potential with equal electron and hole confinement energy ($\hbar\omega$). Under this assumption, the problem is solved numerically after the center-of-mass separation. In self-assembled quantum dots the assumption of separable center of mass is not realistic, however it should not essentially perturb the susceptibility of the carriers to the electric field oriented vertically. The Hamiltonian of the system is a sum of the lateral center-of-mass (H_{cm}) and relative Hamiltonians (H_{rel}), which are given by

$$H_{\mathrm{cm}} = -\frac{\hbar^2}{2M}\nabla^2_{\rho_{\mathrm{cm}}} + \frac{M\omega^2}{2}\rho^2_{\mathrm{cm}} \tag{1}$$

and

$$H_{\mathrm{rel}} = -\frac{\hbar^2}{2\mu}\nabla^2_{\rho_{\mathrm{eh}}} - \frac{\hbar^2}{2m_{\mathrm{e}}}\frac{\partial^2}{\partial z_{\mathrm{e}}^2} - \frac{\hbar^2}{2m_{\mathrm{h}}}\frac{\partial^2}{\partial z_{\mathrm{h}}^2} + \frac{\mu\omega^2}{2}\rho^2_{\mathrm{eh}} + V_{\mathrm{e}}(z_{\mathrm{e}}) + V_{\mathrm{h}}(z_{\mathrm{h}}) - \frac{e^2}{4\pi\varepsilon\varepsilon_0 r_{\mathrm{eh}}} + eF(z_{\mathrm{e}} - z_{\mathrm{h}}), \tag{2}$$

where $M = m_{\mathrm{e}} + m_{\mathrm{h}}$, $\mu = m_{\mathrm{e}}m_{\mathrm{h}}/(m_{\mathrm{e}} + m_{\mathrm{h}})$, m_{e} (m_{h}) is the electron (hole) effective band mass, F is the external electric field, ∇^2_ρ stands for the Laplacian in the $x-y$ plane, z_{e} (z_{h}) is the vertical coordinate of the electron (hole), subscripts cm and eh correspond to the center-of-mass and relative electron–hole coordinates, ρ is the lateral radial coordinate, r_{eh} is the electron–hole distance in the three-dimensional space, and ε is the dielectric constant. Vertical confinement potentials ($V_{\mathrm{e}}(z_{\mathrm{e}})$ and $V_{\mathrm{h}}(z_{\mathrm{h}})$) for the electron and the hole are double potential wells of depth V^0_{e} for the electron and V^0_{h} for the hole and of width 6 nm separated by a barrier of thickness b. Isolated quantum dots may possess a built-in strain-induced electric field [4]. However, in coupled dots, the built-in electric field has the opposite orientation [5]. Therefore, this intrinsic field is neglected in the present calculations (in fact such a build in electric field can be also interpreted as a shift of our applied field). The eigenstates of Hamiltonian (2) have definite z components of total angular momentum and for $F = 0$ also have definite parity with respect to a change of signs of the z coordinates [3]. We are interested in the optically active exciton states that—in III–Vs—correspond to the envelope function of zero angular momentum. The eigenfunctions of Hamiltonian (2) are calculated by the imaginary time technique. We use the material parameters for an In_xGa_{1-x}. As quantum dot embedded in a GaAs matrix with a uniform concentration of indium in the quantum dot $x = 0.66$ [3]. Then, $\varepsilon = 12.5$, $m_{\mathrm{e}} = 0.037m_0$, $m_{\mathrm{h}} = 0.45m_0$, where m_0 is the free electron mass, $V^0_{\mathrm{e}} = -0.508\,\mathrm{eV}$, $V^0_{\mathrm{h}} = -0.218\,\mathrm{eV}$, and we take for the lateral confinement $\hbar\omega = 20\,\mathrm{meV}$. We note, that in the limit of $\hbar\omega = 0$ the present problem reduces to the Stark effect for exciton in coupled quantum wells [9].

Fig. 1 shows the energy spectrum of even-parity zero-angular-momentum exciton states as a function of the barrier thickness. The energy is measured with respect to the energy gap of the barrier material. The area of each dot is proportional to the absorption/recombination probability for state μ calculated as $p_\mu = |\int \mathrm{d}^6\mathbf{r}\Psi_\mu(\mathbf{r}_{\mathrm{e}},\mathbf{r}_{\mathrm{h}})\delta^3(\mathbf{r}_{\mathrm{e}} - \mathbf{r}_{\mathrm{h}})|^2$. The arrows in Fig. 1 show the pairs of energy levels, which are degenerate in the weak interdot coupling limit, i.e., if the electron–hole interaction can be neglected. For large b the lower (upper) energy level, which is split by the electron–hole interaction, corresponds to the electron and the hole in the same quantum dot (different quantum dots). In the pair of states marked by γ the hole is excited in the vertical direction within the dot, which results in a zero recombination probability also in the lower of the two γ energy levels in which the

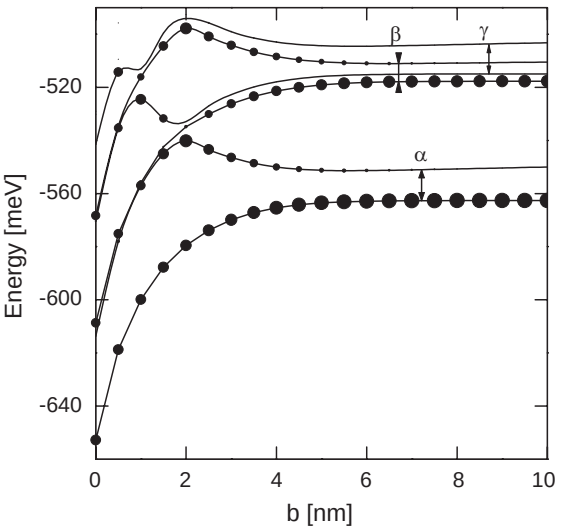

Fig. 1. Even parity energy levels of an exciton with zero angular momentum as function of barrier thickness b. The area of each dot is proportional to the recombination probability. The arrows link the pairs of states that are degenerate for large b, i.e., for negligible electron–hole interaction.

particles occupy the same dot (the odd parity hole wave function is orthogonal to the even parity electron wave function). In this state, both the particles occupy the same quantum dot; however, the odd parity hole wave function is orthogonal to the even parity electron wave function. The energy levels marked by β in absence of the interaction correspond to lateral excitation. For $b<4$ nm the upper energy levels of the couples marked by α and β become bright due to the tunnel coupling between the quantum dots. In the energy spectrum around $b = 2$ nm, anticrossings between the energy levels are clearly visible. The present results for the bright energy levels are in good quantitative agreement with the results of the calculations [3], performed under the assumption that the electron and hole angular momenta are conserved due to the strong lateral confinement (cf. Fig. 4 in Ref. [3]).

The dependence of the energy spectrum on the external electric field for $b = 2$ nm is plotted in Fig. 2a. At zero electric field the first excited state (of odd parity) corresponds to the hole excitation. The electric field breaks the parity symmetry of the system and the excited state becomes optically

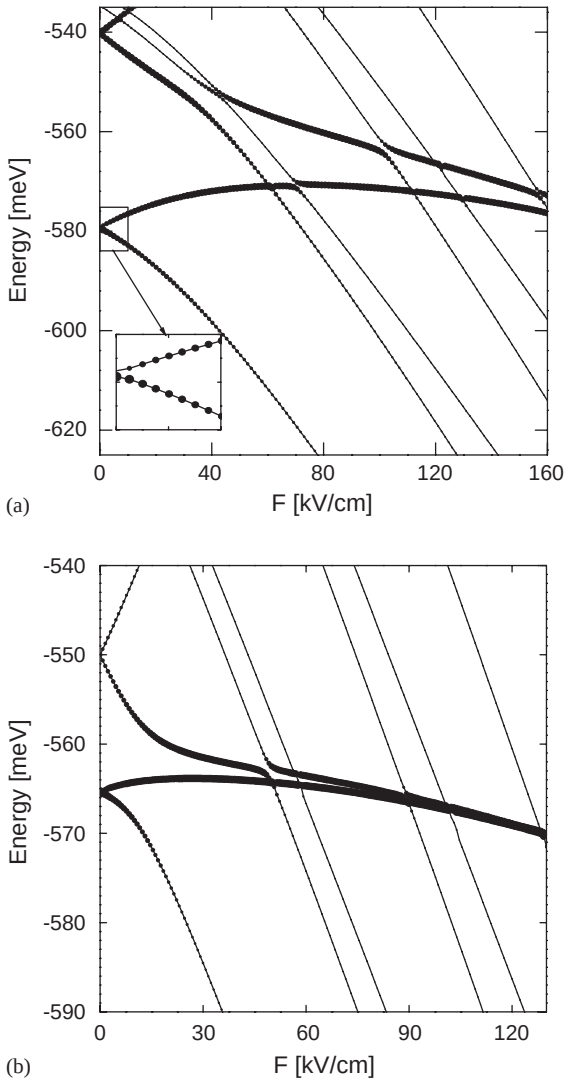

Fig. 2. Exciton energy spectrum as a function of external electric field F for barrier thickness $b = 2$ nm (a) and $b = 4$ nm (b). The area of the dots (thickness of the curves) is proportional to the recombination probability. The inset in (a) shows the low-energy low-field part of the spectrum.

active. The energy dependence of the ground state on the electric field is typical for the normal, i.e., quadratic, Stark effect. The polarizability of the ground state is related to the formation of the induced dipole moment due to the localization of the electron and the hole in the lower and upper quantum dot, respectively, which eventually leads

to the extinction of the recombination. In the excited part of the spectrum one observes two bright energy levels which tend to degeneracy at high electric field. In these two energy levels the electron and the hole occupy the same quantum dot. For the lower (higher) of the two energy levels the carriers are localized in the lower (upper) quantum dot which is favorable for the electrostatic energy of the electron (hole). In the lower-energy (higher-energy) excited bright state, the hole becomes localized in the energetically unfavorable (favorable) quantum dot already for relatively weak electric field and the electron enters the same quantum dot with a pronounced delay. For this reason the lower-energy (higher-energy) level increases (decreases) with increasing electric field. These two bright energy levels exhibit avoided crossings (anticrossings) with the dark energy levels, which correspond to both the carriers separated by the electric field in the same way as in the ground state. For weaker tunnel coupling, i.e., for $b = 4\,\mathrm{nm}$ [cf. Fig. 2(b)] the two bright energy levels become degenerate at lower electric fields.

The most interesting spectrum is obtained for larger barrier thickness, i.e., for weak interdot coupling. Fig. 3(a) displays the electric-field dependence of the exciton energy spectrum for $b = 7\,\mathrm{nm}$. For $F = 0$ the twofold degenerate ground state corresponds to both carriers in the same quantum dot, while in the nearly degenerate excited state the carriers occupy different dots. The degenerate ground state energy is not affected by the electric field, since the electrostatic energy gained by the electron is lost by the hole and vice versa). Near the avoided crossing, i.e., around $F = 9\,\mathrm{kV/cm}$, the state with both the charge carriers in the upper dot is replaced by the state with carriers separated by the external electric field (hole in the upper dot and electron in the lower dot). The state with energy independent of F is orthogonal to the states with anticrossing levels since in this state the hole is localized in the lower dot.

The dependence of the ground-state energy on the electric field is clearly different from a simple parabolic form, which is typical for the well-known quadratic Stark effect. The unusual Stark effect, shown in Fig. 3(a), should be visible in low-excitation photoluminescence spectroscopy [2].

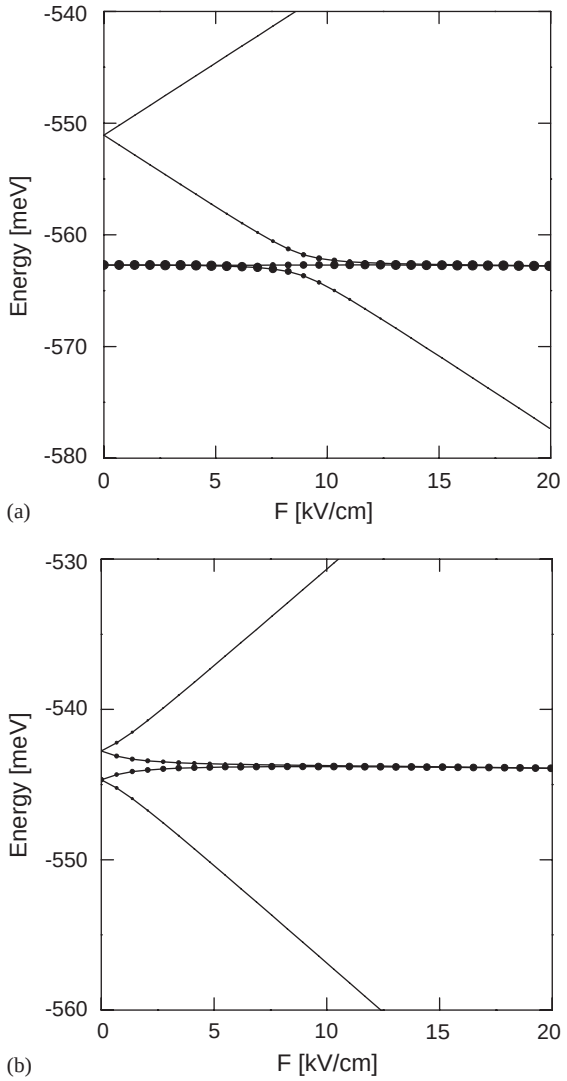

Fig. 3. Exciton energy spectrum as a function of external electric field F for barrier thickness $b = 7\,\mathrm{nm}$ with (a) and without (b) the electron–hole interaction.

The predicted anomaly of the Stark effect is due to the electron–hole interaction which stabilizes the carriers in the same quantum dot for a quite wide range of electric field. Fig. 3(b) shows the dependence of the spectrum on the electric field, calculated without electron–hole interaction. The low-energy spectrum consists of two pairs of the energy levels, which are almost degenerate with respect to the parity of the hole and split by the

electron tunnel coupling. For $F = 0$ in all the states, the probabilities of finding the electron and the hole in the same or different quantum dot are equal to $\frac{1}{2}$. In the ground state the electric field has to overcome only the weak electron tunnel coupling between the dots to separate the carriers.

In summary we have found an unusual non-parabolic Stark effect for the exciton in vertically coupled quantum dots. The effect appears for relatively thick barriers and is a consequence of the electron–hole interaction. The separation of the carriers appears via an avoided crossing which forms a characteristic structure with a third energy level independent of the field.

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