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# Entangled states of electron–hole complex in a single InAs/GaAs coupled quantum dot molecule

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**Abstract**

We summarize here results of calculations and experiments on electron and valence hole states in a single pair of vertically stacked and electronically coupled InAs self-assembled quantum dots. In perfectly aligned quantum dots one can relate an electron–hole complex to a pair of entangled qubits. The information carried by individual qubit is related to the quantum dot index (isospin) of individual carrier. The quality of fabricated quantum dot molecules is identified from the exciton fine structure in a magnetic field. 

*Keywords:* Quantum dot molecule; Isospin; Entanglement

The quantum processor consists of individual qubits and quantum logic gates which entangle states of two qubits [1]. Entangled states of two qubits are states which cannot be obtained by rotation of individual qubits. Quantum dots (QDs) [2–4], trapped ions [5], and nuclear spins [6] are among a number of candidates for such devices. Recently, we proposed to use coupled self-assembled QDs for quantum information processing [7]. For two vertically coupled dots, the information is carried by the dot index. For example, an electric field applied in the growth direction (cf. Fig. 1) could localize an electron in the lower dot (index 0) or the upper dot (index 1), and hence encode information. The dot indices are referred to as states of an isospin [8]. Turning off the electric field would allow the tunneling to take place and would lead to the superposition of two QD states i.e. to single qubit rotation. The simplest realization of a pair of qubits comprises of not only an electron but also a valence hole in the same pair of dots. In the electric field the two carriers are localized on opposite dots. When the electric field is turned off, the interdot tunneling and Coulomb interactions leads to the appearance of entangled states of two qubits (isospins) [3].

The investigated structures consist of two self-assembled InAs/GaAs QDs grown by the indium-flush method [3,9]. The QDs in a form of flat disks of radius $R$ (typically 8–12 nm) and height $h$ (typically 1–2 nm, cf. Fig. 1) are located on the respective wetting layers (WLs) and vertically aligned due to propagation of strain. The application of indium-flush technique allows the separation $D$ between QD layers to be $\sim$4 nm, resulting in quantum mechanical tunneling induced splitting of $\sim$ 30 meV. We show here how photoluminescence experiments [3] on such

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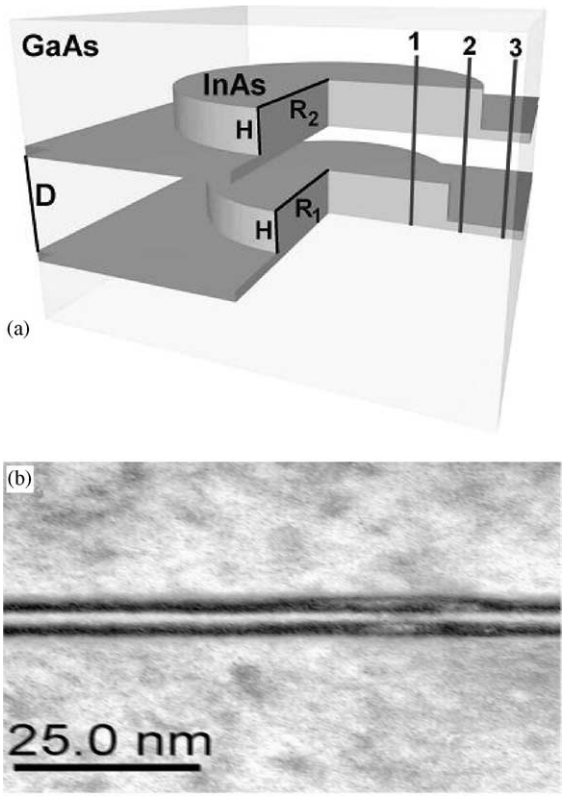


Fig. 1. (a) Schematic picture of the QD molecule. (b) TEM image of coupled InAs QDs separated by a 4-nm-thick GaAs barrier.

structures can be understood as a proof of coherent interdot tunneling and entanglement of electron–hole states. We start with only one electron. As the height of the dot is much less than the dot radius, the electronic wave function is strongly localized along the growth direction. The electronic states on each dot can be then labeled by the angular momentum $m$ and isospin. Due to strong lateral confinement it is sufficient to consider only the $m=0$ s-orbitals on each dot, so we deal only with isospin states $|0\rangle$ or $|1\rangle$. In this basis the Hamiltonian of the system can be written as [3,8]

$$H = \begin{bmatrix} E_s & -t \\ -t & E_s \end{bmatrix}, \tag{1}$$

where $E_s$ is the energy of each isospin state (identical for both states if dots are identical), and $t = \langle 0|H|1\rangle$ is the interdot tunneling matrix element. The eigenstates of this Hamiltonian are $|+\rangle = (1/\sqrt{2})(|0\rangle + |1\rangle)$ and $|-\rangle = (1/\sqrt{2})(|0\rangle - |1\rangle)$, with the corresponding eigenenergies $E_\pm = E_s \mp t$. Tunneling leads then to a rotation of the isospin and the splitting of the isospin energies to symmetric $(+)$ and antisymmetric $(-)$. Note that the Hamiltonian (1) is that of a single spin in a magnetic field, and the symmetric–antisymmetric splitting $2t$ is equivalent to the Zeeman energy. In order to quantify the parameter $t$, we compare our model to a realistic calculation of the single-particle states in coupled QDs [10]. The calculation starts with the calculation of strain using the continuous elasticity model. The strain tensor components are entered into the Bir-Pikus Hamiltonian and the effective band structure of the system is found. The strain-induced modification of the band structure is present mainly in the dots and causes the reduction of the GaAs/InAs conduction band offset to 600 meV. The heavy-hole and light-hole bands are split by $\sim 200$ meV, and the heavy-hole and light-hole InAs/GaAs band offsets are, respectively, $\sim 150$ and $\sim 200$ meV. For the application of our structures to the quantum information, a relatively easy and portable computational framework is needed. The effective mass and adiabatic approximations fulfill this requirement. Due to strong confinement in the growth direction we deal with distinct subbands $v = 0, 1, \ldots$ related to the vertical motion in the $z$-direction, on which the ladder of states corresponding to the lateral motion in the radial direction is built. Therefore, we write the total electron wave function as a product of the slowly varying with $r$ vertical part $g_r^v(z)$, and the radial part $f_m^v(r)$: $\psi(r,\vartheta,z) = (1/2\pi)e^{im\vartheta} g_r^v(z) f_m^v(r)$. For each angular momentum channel $m$ and each subband index $v$ these functions satisfy the set of equations:

$$\left[-\frac{\partial^2}{\partial z^2} + V(r,z)\right] g_r^v(z) = E_v g_r^v(z), \tag{2a}$$

$$\left[-\frac{1}{r^2}\left(r\frac{\partial}{\partial r} r \frac{\partial}{\partial r} - m^2\right) + E_v(r)\right] f_m^v(r) = E f_m^v(r). \tag{2b}$$

The potential $V(r,z) = -V_0$ in QDs and WL and 0 in the barrier. These equations are solved using the

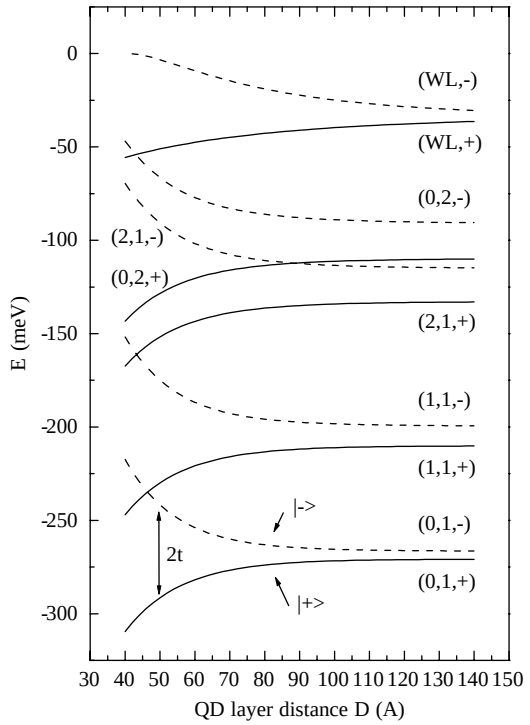


Fig. 2. Electronic energy levels of a QD molecule versus the QD layer distance $D$ calculated using the adiabatic model. States are labeled by their angular momentum and parity; wetting layer states are also shown. The s-shell is also labeled by the isospin indices.

transfer matrix method and can be easily generalized to a system of more stacked disks. The procedure requires two parameters—confinement depth $V_0$ and the effective mass $m^*$. The former is established from the strain calculation, and the latter, treated as a fitting parameter, is found by comparison to the results of a $\mathbf{k}\cdot\mathbf{p}$ simulation [11] ($m^* = 0.053m_0$). The resulting single-particle energy levels versus the QD layer distance are shown in Fig. 2. For the s-shell a symmetric–antisymmetric splitting of $\sim 5$ meV is observed at a QD layer distance of 10 nm, increasing to $\sim 80$ meV at $D=4$ nm. Also, for $D=4$–5 nm a crossing of the antisymmetric s-level and the symmetric p-level occurs. As can be seen, our simple isospin model captures the basic physics of the system. States of the s-shell are the isospin $|+\rangle$ and $|-\rangle$ states, and the tunneling matrix element $t$ depends exponentially on the QD layer distance $D$.

For holes this simple calculation is not valid due to band mixing effects [12], and the results of this calculation will be published elsewhere [13]. In order to address the PL experiment, we consider the case of an electron–hole pair localized on the QD molecule. Our Hamiltonian in this case can be written as

$$\hat{H} = \sum_i E_i^{\mathrm{e}} c_i^+ c_i + \sum_i E_i^{\mathrm{h}} h_i^+ h_i - \sum_{ijkl} \langle ij|V|kl\rangle c_i^+ h_j^+ h_k c_l, \tag{3}$$

where $c_i(h_i)$ is the electron (hole) annihilation operator, and $\langle ij|V|kl\rangle$ denotes the two-body Coulomb matrix element. In the absence of the Coulomb interaction, but in the presence of the interdot tunneling the two carriers can be represented by two rotated isospins. Thus the simplest basis set for the exciton problem is composed of four states e.g.

$$|+\rangle_{\mathrm{e}}|+\rangle_{\mathrm{h}} = \tfrac{1}{2}(|0\rangle_{\mathrm{e}} + |1\rangle_{\mathrm{e}})(|0\rangle_{\mathrm{h}} + |1\rangle_{\mathrm{h}}), \tag{4a}$$

$$|-\rangle_{\mathrm{e}}|-\rangle_{\mathrm{h}} = \tfrac{1}{2}(|0\rangle_{\mathrm{e}} - |1\rangle_{\mathrm{e}})(|0\rangle_{\mathrm{h}} - |1\rangle_{\mathrm{h}}) \tag{4b}$$

and $|+\rangle_{\mathrm{e}}|-\rangle_{\mathrm{h}}$ and $|-\rangle_{\mathrm{e}}|+\rangle_{\mathrm{h}}$. The states (4a) and (4b) can be created optically but the remaining two states are dark. Note that each basis state is a product of rotated individual qubits. Now we must diagonalize the Hamiltonian of interacting isospins in this basis set. To this end we approximate the electron and hole wavefunctions in the form $\langle \mathbf{r}|+\rangle = N_+ J_0(k_0^\rho \rho)\sum_l A_l^+ \cos(k_l^z z)$, where angular momentum $m = 0$, $N_+$ is the normalizing factor, and $J_0$ is the Bessel function. We take analogous forms for parity $(-)$. Coulomb scattering does not mix the subspace of optically active states and dark states. The exciton Hamiltonian can be simply written as

$$\hat{H} = \begin{bmatrix} E_{++} + V_{++} & V_{+-} \\ V_{+-} & E_{--} + V_{--} \end{bmatrix} \tag{5}$$

in the basis $|+\rangle_{\mathrm{e}}|+\rangle_{\mathrm{h}},\ |-\rangle_{\mathrm{e}}|-\rangle_{\mathrm{h}}$. The evolution of the resulting energy levels (bright and dark) is shown in Fig. 3a (solid lines). When the dots are far apart, there are two groups of energy levels split by Coulomb interaction. When the dots are closer, the tunneling leads to the increase of splitting between optically active states. The key result is that due to the Coulomb interaction the eigenvectors corresponding to these energy

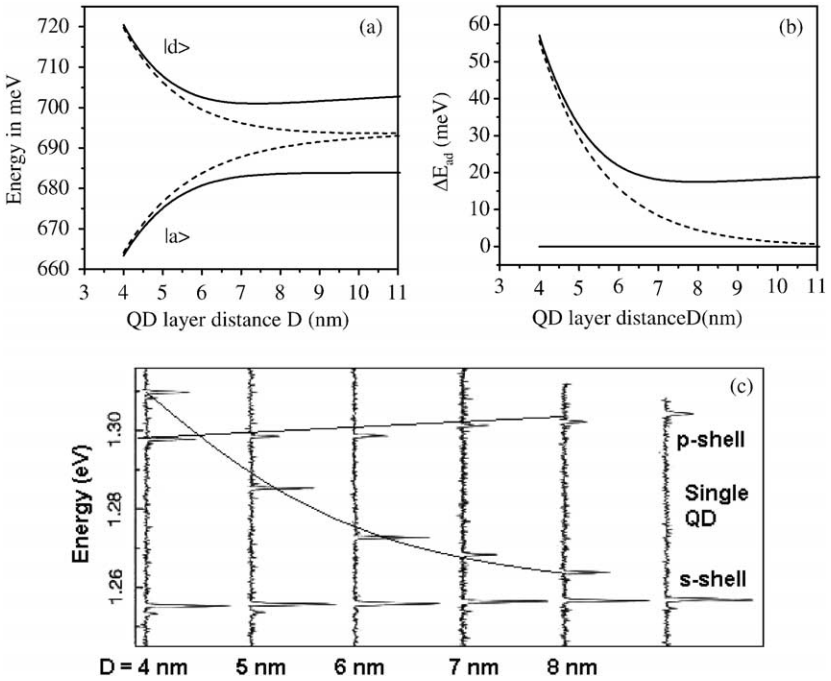


Fig. 3. (a) Energies of exciton bright states $|a\rangle$ and $|d\rangle$ calculated using the two-isospin model of the electron–hole pair with (solid lines) and without entanglement (dashed lines). (b) Splitting between the energies of optically active states as a function of QD distance with (solid lines) and without entanglement (dashed lines). (c) PL spectra for QD molecules with various interdot distances, recorded at low excitations and temperature $\sim$60 K. The PL spectrum of a single dot is shown as a reference.

levels,

$$|a\rangle = \alpha_1(|0\rangle_e|0\rangle_h + |1\rangle_e|1\rangle_h) + \beta_1(|0\rangle_e|1\rangle_h + |1\rangle_e|0\rangle_h), \quad (6a)$$

$$|d\rangle = \alpha_4(|0\rangle_e|0\rangle_h + |1\rangle_e|1\rangle_h) + \beta_4(|0\rangle_e|1\rangle_h + |1\rangle_e|0\rangle_h), \quad (6b)$$

cannot be written as simple tensor products of individual isospin states. Tunneling and Coulomb interaction lead then to the entanglement of isospins. If we create optically an electron on the dot 0, and a hole on the dot 1, then once the tunneling is allowed (the electric field is switched off), this state will evolve into an entangled state. The energies of the entangled states $|a\rangle$ and $|d\rangle$ strongly depend on the interdisk distance $D$ and in the limit of large $D$ exhibit a characteristic splitting due to the off-diagonal Coulomb matrix element $V_\pm$ (cf. Fig. 3a—solid lines). If this element is equal to zero (no Coulomb interactions), we observe no entanglement, and the behavior of the excitonic energy levels is different: the states $|a\rangle$ and $|d\rangle$ become degenerate for large $D$ (cf. Fig. 3a—dashed lines). By studying the behavior of the excitonic energy levels as a function of $D$ it should be therefore possible to distinguish the entangled isospin states from those without entanglement.

To demonstrate the validity of this model, PL spectroscopy was performed on single QD molecules located on different samples with different dot separations [3]. To suppress the inhomogeneous broadening the molecules were lithographically isolated in mesas. The measurement was taken at low excitation powers, but at temperatures $\sim$60 K, so that the emission from ground and higher states of a single exciton is expected due to thermal excitation. In Fig. 3c we show the PL spectra recorded on molecules of high geometrical symmetry (see below) where we aligned all the spectra according to the ground state emission

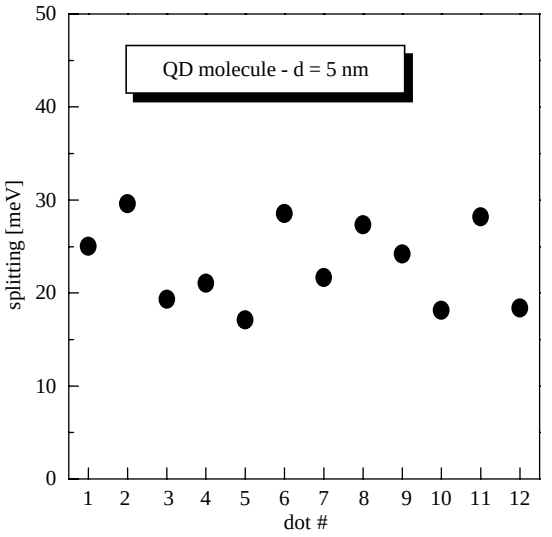


Fig. 4. Energy splitting between the two optically active isospin states of the exciton for different QD molecules all having a nominal barrier width of 5 nm.

line, treated as a reference level. We observe that the tunneling induced splitting of the s-shell is of the order of 10 meV for a QD separation of 8 nm, and increases to about 60 meV for a QD separation of 4 nm. This splitting is an order of magnitude larger than that observed in previous experiments [14]. An additional line, due to emission from the p-shell is also seen. The trends and orders of magnitude expected from model calculations (cf. Fig. 3b) are in qualitative agreement with the experimental data. Most importantly, the splitting between the optically active entangled exciton states becomes larger than 30 meV when the interdot barrier width becomes smaller than 5 nm.

The theory presented above assumes the two disks composing the molecule to be identical for all barrier widths, a situation difficult to achieve experimentally. To examine the influence of size fluctuations we performed PL measurements of a set of QD molecules with the same nominal barrier width equal to 5 nm. Fig. 4 shows the observed splitting of optically active states for several QD molecules from this set. The average value of the splitting is 23 meV, and the range of fluctuations is roughly $\pm 7$ meV. For that reason we assume that the optical spectra shown in Fig. 3c have an uncertainty of $\pm 7$ meV, a value smaller than the splitting of optically active states even for the sample with 8 nm barrier.

In our calculations we also assumed that the QDs forming a molecule are identical, which is experimentally very unlikely. However, it is important to note that the coupling-induced splitting of the electronic levels can be observed even if the dots are different. Indeed, the energies of the two isospin states will no longer be equal, i.e., $E_s^1 - E_s^0 = \Delta \neq 0$, but the coupling will be formed as long as this asymmetry correction $\Delta$ (typically of order of $\sim 5$ meV) is smaller than tunneling-induced splitting $t$ (typically of the order of $\sim 50$ meV for barrier thickness small enough). Of course, the isospin rotation, and hence also the Coulomb energies will differ from the ideal case, which will lead to a smaller degree of entanglement of the electron–hole pair. The results of the corresponding calculations will, however, be qualitatively similar to the ones presented here.

The data presented so far was obtained on structures with a rather high geometric symmetry. However, it is not absolutely clear from the experimental data whether both electron and hole tunnel. Indeed, besides structures with high symmetry we also find structures with a reduced symmetry. Insight into this symmetry can be taken from studies of the fine structure arising from the coupling of electron and hole spins by exchange and from their interaction with an external magnetic field. The different spin orientations of electron and hole result in a four-fold spin degeneracy of each isospin state. The different exciton configurations that can be formed are characterized by total angular momentum $M = S_{e,z} + S_{h,z}$ with $S_{e/h,z} = \pm\frac{1}{2}$, where we use an effective spin formalism for the hole angular momentum. The two states with $M = 0$ are optically active states, while the states with $M = \pm 1$ form an optically inactive doublet. Applying a magnetic field leads to the splitting of both doublets due to the Zeeman interaction of the carrier spins with $B$. This is the situation that we find in the experiments for the symmetric molecules that we have considered above. An example of such a molecule with $D = 8$ nm is shown in Fig. 5 (left panel). At zero magnetic field a single emission line is observed which is due to recombination of the bright excitons. The spin splitting of this emission line depends linearly on magnetic field and reaches 1.5 meV at 8 T.

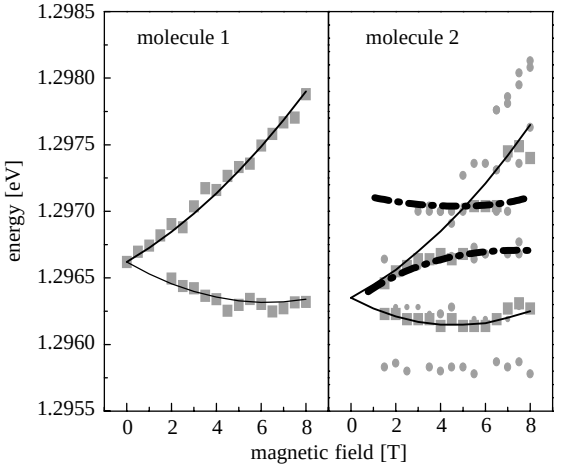


Fig. 5. Fine structure splitting of the isospin ground state as function of magnetic field. Left panel shows the splitting for a molecule of high symmetry whereas the right panel shows the splitting for a molecule structure of reduced symmetry. The barrier thickness in each case was 8 nm. Symbols give the experimental data and the thick dashed-dotted lines are guides to the eye for the anticrossing described in the text. The solid lines are the results of calculations involving the exciton fine structure Hamiltonian in an ideal QD molecule.

The right panel in Fig. 5 shows the fine structure of another QD molecule, also with a barrier of 8 nm. Here the fine structure is much more complicated. In high magnetic fields up to six emission lines appear. This can only occur when the symmetry of the molecule is reduced. Such a symmetry reduction could result from considerable compositional or geometrical variations of the QDs. Such a variation will mainly influence the hole due to its much heavier mass, while the tunneling of the electron will be unaffected as long as the variations are not too strong. In other words, the energy difference due to the asymmetry is so large for the holes that the energy reduction that is gained by the formation of a symmetric molecule state is too small for this asymmetry. The tunneling of the electron is evidenced by the observation of a higher lying state about 10 meV above the ground state. In some sense, the molecule symmetry can be restored by a magnetic field. Through the Zeeman interaction, the hole levels can be brought into resonance at which tunneling occurs. In the spectra this is observed as a pronounced anticrossing as shown in the right panel around 3 T. At lower field the spectra are dominated by a doublet. The higher lying line first shifts to higher energies. Around 3 T, however, it comes into resonance with another state and these two states exchange their character (oscillator strength and magnetic field dispersion). The anticrossing is indicated by the thick dashed-dotted lines.

To summarize these fine structure studies, the data in the left column of Fig. 5 give additional support for a tunneling of both electron and hole. We want to stress that such a demonstration also proves the entanglement of the electron–hole pair, as discussed above. Due to the restriction of space here, further data confirming the formation of entangled exciton states will be presented elsewhere [15].

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