Electric field control and optical signature of entanglement in quantum dot molecules

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The degree of entanglement of an electron with a hole in a vertically coupled self-assembled dot molecule is shown to be tunable by an external electric field. Using atomistic pseudopotential calculations followed by a configuration interaction many-body treatment of correlations, we calculate the electronic states, degree of entanglement and optical absorption. We offer a novel way to spectroscopically detect the magnitude of electric field needed to maximize the entanglement.

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A pair of quantum dots or “quantum dot molecule” (QDM) occupied by two electrons [1,2] or by an electron-hole pair [3] have been offered [3,4] as a basis for quantum computing. The fundamental requirement for such quantum algorithm is the availability of entangled states and the ability to entangle and disentangle the quantum bits (qubits). In the context of a dot molecule, an entangled electron-hole pair can be represented by the the maximally entangled Bell state $e_{T}h_{T} + e_{B}h_{B}$, where $e$ and $h$ stand for the electron and the hole (the two qubits) and $T$ and $B$ for their localizations in top or bottom dot. The original proposal [3] and subsequent experiments [3,4] for entangled electron-hole pairs in QDMs promised a high degree of entanglement [3] based on analysis via simple models. However, later theoretical work showed [6] that electron-hole entanglement is generally low in such cases and develops a sharp maximum only at a specific interdot separation that critically depends on the size difference of the two dots. Unfortunately, it has proven to be difficult to experimentally control so precisely the interdot distance and the size difference of the two dots. The question we address here is whether the degree of entanglement can be maximized by other means, more accessible experimentally than a variation of the interdot separation. We propose and quantify theoretically that it is possible to tune and control the degree of entanglement by applying an external electric field in the growth direction [7,8,9,10].

The use of electric field has been demonstrated in quantum dots [7,8,9,10] and very recently in a single quantum dot molecules by Krenner et al. [11]. We predict that, while the entanglement at zero field is generally low (35% for our case), it can reach a high value (75% in our case) at a specific electric field $F_{S_{\text{max}}}$ (-5.4 kV/cm in our case). Moreover, precisely at this field the first two exciton lines merge, giving a well defined spectroscopic signature of the point of maximum entanglement.

In order to obtain reliable results for the correlated exciton states, it is of foremost importance to accurately account for the multi-band character of the hole states and for the correct strain dependence in the coupling region (between the dots). We have thus solved the pertinent Schrödinger equation atomistically, in a multi-band fashion. We use the Hamiltonian $H = -1/2 \nabla^2 + \sum_{n,n} v_{\alpha}(r - R_{n}) + V_{SO} + |e| F_{z}$ under an external electric field $F$ applied in [001] ($z$) direction. The atomistic pseudopotentials $v_{\alpha}$ of atom of type $\alpha$ and the non-local spin-orbit potential $V_{SO}$ are fit to reproduce InAs and GaAs bulk properties [6,12]. The atomic positions $\{R_{n}\}$ are obtained by minimizing the atomistic strain energy (via valence force field [13]) for a given shape and size of the dots. Our quantum dots have a truncated cone shape (12 nm base and 2 nm height) with a composition ranging from pure InAs at the top to In$_{0.5}$Ga$_{0.5}$As at the base, as determined in Ref. [3]. The single-particle Hamiltonian is diagonalized in a basis $\Psi = \sum_{n,k} A_{n,k}\phi_{n,k}$ of pseudopotential Bloch functions $\phi_{n,k}$ as outlined in Ref. [14], thus permitting coupling of various Bloch states. Correlations are treated via a many-body expansion in Slater determinants [15,16] where the electrons not included dynamically are represented by a model screening of the Coulomb and (long and short range) exchange [17]. The entanglement is calculated according to the von Neumann entropy of entanglement [18,19].

The bonding ($b$) and antibonding ($a$) electron molecular levels of a dot molecule will be denoted as $E_{b}$, $E_{a}$. For an idealized (mostly unrealistic) symmetric case the lowest energy molecular orbitals (MOs) develop from single-dot electron states $e_{T}$ and $e_{B}$ located on the bottom ($\bar{B}$) and top ($\bar{T}$) dots:

$$\psi[E_{b}] = \frac{1}{\sqrt{2}}(e_{T} + e_{B}); \quad \psi[E_{a}] = \frac{1}{\sqrt{2}}(e_{T} - e_{B}), \quad (1)$$

and similarly for the holes $H_{b}$, $H_{a}$. As shown previously [6,20], in reality, because of strain and random-alloy fluctuations, one does not have a symmetric bonding-antibonding behavior even if the dot molecule is made of identical (but non-spherical) dots. This is seen in Fig. 1 where both electron and hole molecular orbital wave functions are shown for zero electric field $F = 0$. We see that the (lighter-mass) electrons tunnel between dots, forming bonding-antibonding states as in Eq. (1), but the (heavier-mass) holes remain localized on the top (bottom) dot for the bonding (antibonding) MO $H_{b}$ ($H_{a}$). The single-particle molecular orbital energy levels are shown in Fig. 2. As we apply an electric field the molecular lev-
els that develop from the single-dot orbitals exhibit anti-crossing. We have indicated in Fig. 2 the major character of the molecular states $E_a$, $E_b$, $H_a$, $H_b$ in terms of the localization on individual dots ($e_T$, $e_B$, $h_T$ and $h_B$) using the calculated MO wave functions of Fig. 1. We see that for holes at positive fields $\psi(H_b) \simeq h_B$ and $\psi(H_a) \simeq h_A$ while for electrons $\psi(E_a) \simeq e_T$ and $\psi(E_b) \simeq e_B$. The opposite is true for negative fields. Thus, by applying an electric field we can tune the localization of the MO's and, for instance, compensate for size, composition or shape differences of both dots. We will see that this tuning of localization will also control the degree of entanglement.

There are four transitions between the four molecular levels shown as vertical arrows in Fig. 2. Their single-particle transition energies $\varepsilon_g$ (differences between the energies from Fig. 2) are given in Fig. 3(a) and show maxima and minima vs field. We note in Fig. 3(a) the character of the four transitions in terms of localization on single-dot orbitals. We see that at high fields, the lowest- and highest-energy transitions involve different dots: for example $E_b H_a$ is $e_B h_T$ at positive fields, and $e_T h_B$ at negative fields. Thus, the corresponding dipole transitions are expected to be weak ("dark states"). In contrast the second and third transitions at high fields involve the same dots: for example $E_b H_a$ is $e_B h_B$ at positive fields and $e_T h_T$ at negative field. Thus, the corresponding dipole transitions are expected to be large ("bright states").

The single-particle approximation underlying Fig. 3(a) is valid only in the case of large fields, where $e^{-h}$ Coulomb effects are small compared to the field-driven variation in the single-particle levels. At these large fields, the excitons are pure determinants with localization on either $e_T$ or $e_B$ or $h_T$ or $h_B$ and therefore show no entanglement. We will next see that in the interesting region of electric fields, $e^{-h}$ Coulomb interactions are crucial. In Figure 3(b) we show the calculated electron-hole Coulomb interaction $\varepsilon_{e^{-h}} + J_{e^{-h}}$, but without correlation effects. (d) Final correlated exciton results. The bidot products ($e_T h_T$, etc...) are given whenever the MO states (given on the right) are strongly dominated by one of these products.

\begin{equation}
J_{e^{-h}}[i - j] = \int \int \psi_i^*(r_a) \psi_j^*(r_b) \psi_i(r_a) \psi_j(r_b) \frac{1}{|r_a - r_b|} \, dr_a \, dr_b
\end{equation}
between the MOs \(e_i = E_a\) or \(E_b\) and \(e_h = H_a\) or \(H_b\) using the model of Resta \(17\) for the screening \(\epsilon\). This interaction \(J_{e,h}\) (Fig. 3(b)) shows the reverse behavior vs field compared with the MO energies \(e_i\) vs field [Fig. 3(a)]. For example, whereas \(J_{e,h}[E_aH_b]\) is maximal (less negative) at large positive or negative fields, and minimal at intermediate fields, the MO band gap \(\epsilon_a[E_aH_b]\) is minimal at large positive or negative fields and maximal at intermediate fields. Not surprisingly, when one calculates the Coulomb-corrected excitonic transition energies \(\epsilon_{i,j}^e + J_{e,h}^i\) between the molecular states \(i\) and \(j\) [Fig. 3(c)] one sees a partial cancellation for the two low-energy transitions, \(E_bH_b\) and \(E_bH_a\), leading to a weak dependence of the transition energy on field. In contrast, inspection of Figs. 3(a) and 3(b) for the two highest-energy transitions shows that the field-dependence of \(\epsilon_{i,j}^e + J_{e,h}^i\) reinforce each other, so the excitonic gap \(\epsilon_{i,j}^e + J_{e,h}^i\) [Fig. 3(c)] has an amplified dependence on field. We conclude that the combination of \(\epsilon_i\) and \(J_{e,h}\) bring the lowest-energy transitions closer to each other, while pushing the two higher-energy transitions apart. This will affect the correlation coupling between the MO’s, as seen next.

The Coulomb-corrected excitonic transition energies \(\epsilon_{i,j}^e + J_{e,h}^i\) neglect the interactions between the different configurations, i.e., the states from Fig. 3(c) are not allow to interact. This interaction is included in the next step via a configuration interaction (CI) \(15, 16\) calculation in which we include all Coulomb and exchange integrals from the first four electron and first four hole states (including spin). The results are shown in Fig. 3(d) as a function of electric field. The lowest energy transitions (excitons \(|1\rangle\) and \(|2\rangle\) have a very weak dependence on field, similarly to the case without correlations [Fig. 3(c)]. In contrast to the perturbative approach of Fig. 3(c) the states do not cross but anticrossing at \(-5.4\) kV/cm, as expected from interacting states. Similarly, \(|3\rangle\) and \(|4\rangle\) anticross at \(+3.6\) kV/cm but in a more abrupt fashion.

To understand the correlated CI results we next analyze \(|1\rangle, |2\rangle, |3\rangle, |4\rangle\) by decomposing the correlated excitonic states into sums of products of single-dot states \(e_T h_T, e_B h_T, e_T h_B\) and \(e_B h_B\) called “bidot products” (see Ref. 19 for details). The results of this decomposition are given in Fig. 4(a)(b) for states \(|1\rangle\) and \(|2\rangle\) as a function of the electric field. Fig. 4(c) shows the degree of entanglement calculated for the correlated CI wave functions using the von Neumann formula \(12, 13\) \(S = -\text{Tr } \rho_e \log_2 \rho_e\), where \(\rho_e\) is the reduced density matrices of the electron. When a state is made solely of a single bidot product such as \(e_B h_T\) it is unentangled, but when it is made of a coherent superposition, such as \(e_B h_B \pm e_T h_T\) it might be entangled. In the case of very strong positive fields (larger than \(20\) kV/cm), state \(|1\rangle\) is purely \(e_B h_T\) (unentangled), as the field pulls the electron to the bottom dot and the hole to the top dot. The excitons \(|2\rangle\) and \(|3\rangle\) are already, at moderate positive fields, purely \(e_B h_B\) and \(e_T h_T\), respectively, and remain unentangled states at large fields. The entanglement of \(|1\rangle\) and \(|2\rangle\) [Fig. 4(c)] reaches its maximum of around \(75\%\) at \(E_{S\text{max}} = -5.4\) kV/cm. At this field the exciton states \(|1\rangle\) and \(|2\rangle\) are mainly composed of \(e_T h_T \pm e_B h_B\) configurations, as shown in Fig. 4(a)(b). The calculated excitonic states of Fig. 4(d) are now used to calculate the absorption spectra in Fig. 5. In Fig. 5 the oscillator strength is plotted for different values of the electric field as a function of the transition energy. The plot shows a total of four transitions marked with \(|1\rangle\), \(|2\rangle\), \(|3\rangle\), \(|4\rangle\). Since the transitions \(|3\rangle\) and \(|4\rangle\) are weak, their position is marked by solid and open arrows respectively. The transitions \(|1\rangle\) and \(|2\rangle\) (\(|3\rangle\) and \(|4\rangle\) have a weak (strong) dependence on field and show an anticrossings at \(E_{S\text{max}} (E_{S\text{PT}})\). The spectra show that at \(E_{S\text{max}}\) the lowest energy exciton \(|1\rangle\) becomes dark and progressively gains oscillator strength away from the anti-crossing. The point of merging of \(|1\rangle\) and \(|2\rangle\) at the field \(E_{S\text{max}}\) reflects a “resonant conditions” with maximum entanglement (Fig. 4). The distinct spectroscopic signal of the anticrossing (where the lowest line progressively looses oscillator strength) occurs at the point of maximum entanglement and, we suggest, can give experimentalists a simple way to control a delicate quantity such as entanglement.

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![Fig. 4](image-url)
and 7 meV respectively with a gap of 1.3 eV. Generally, the energy separation between \( e_T \) and \( e_B \) and between \( h_T \) and \( h_B \) can reflect size, composition or shape differences of the two dots. These differences can be tuned by the electric field. Fig. 6(b) shows the energies of simple products of these electron and hole states. They show two closely spaced levels (3 meV apart) in the center of the spectrum and two states 7 meV lower and higher in energy. These energies are different from the MO’s energies of Fig. 3(a) that are combinations of \( e_T h_T, e_T h_B, e_B h_T \) and \( e_B h_B \) at this field. In the next step, in Fig. 6(c), the two-body Coulomb attraction is taken into account and lowers the \( e_T h_T \) and \( e_B h_B \) states in such a way that they are about 14 meV below the \( e_T h_B \) state. This is the consequence of a weak \( e - h \) binding for the dissociated excitons \( e_T h_B \) and \( e_B h_T \). Notably, the simple products \( e_T h_T \) and \( e_B h_B \) are energetically nearly degenerate at this level, this is the “resonant condition” mentioned above. In the last step, in Fig. 6(d), the excitons \( |1> \) and \( |2> \) are now created by including the effects of electron and hole hopping that effectively produce correlated states. The excitons \( |1> \) and \( |2> \) are now a bonding- and antibonding-like combination of the energetically degenerate \( e_T h_T \) and \( e_B h_B \) states. The excitons \( |1> \) and \( |2> \) are now split by a small energy of less than 1 meV. This small splitting is conceptually very similar to the Davydov splitting \([22]\) in molecular crystals. The analysis also reveals that \( |1> \) is anti-symmetric \((e_T h_T - e_B h_B)\) and therefore optically dark while \( |2> \) is symmetric \((e_T h_T + e_B h_B)\) and optically bright. The high symmetry of these states (purely symmetric and anti-symmetric) leads to the high degree of entanglement. Any deviations from \( F_{S_{\text{max}}} \) will lead to a less symmetric combinations as \( \frac{1}{2}(\alpha e_T h_T + \beta e_B h_B) \) with \( \alpha \neq \beta \) with lower entanglement and smaller oscillator strength.

In conclusion, we showed that the degree of electron-hole entanglement in coupled quantum dots can be tuned by an external electric field and that the point of maximum entanglement can be identified by measuring the photoluminescence spectra, observing the merging of two peaks. This opens new ways for experimentalists to identify the electric field needed to achieve maximum entanglement in specific dot molecules. We finally analyzed the nature of the excitons and revealed the interplay of single particle effects, direct Coulomb binding and electron and hole hopping on the many body levels. We described how these effects conspire to yield a highly entangled state.

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[23] In order for this e-h entanglement to be useful in a quantum computation scheme, the qubits need to be physically separated to be addressed individually. One possibility for such a separation seems to be the use of in plane electric fields that would coherently drive the electron and hole to different neighboring quantum dots. This would constitute the next experimental challenge.