Tunneling and Electric-Field Effects on Electron-Hole Localization in Artificial Molecules

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Abstract

I theoretically investigate the Stark shift of the exciton groundstate in two vertically coupled quantum dots as a function of the interdot distance. The coupling is shown to enhance the tunability of the linear optical properties, including energy and oscillator strength, as well as the exciton polarizability. The coupling regime that maximizes these properties results from the detailed balance between the effects of the single-particle tunneling, of the electric field and of the carrier-carrier interaction. I discuss the relevance of these results to the possible implementation of quantum-information processing based on semiconductor quantum dots: in particular, I suggest the identification of the qubits with the exciton levels in coupled- rather than single-dots.

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Semiconductor quantum dots (QDs) are quasi-zero-dimensional heterostructures, where the strong spatial confinement of electrons and holes gives rise to discrete optical spectra [1]. As compared to natural ones, such “artificial atoms” show a wide tuneability of the structural, electronic and optical properties, of the coupling with each other and with external fields. This makes them an ideal laboratory for the many-body physics, where the relative importance of the different kinds of interaction can be modified in order to produce regimes which are precluded to atomic and molecular physics (see, e.g., Refs. [2, 3], and references therein). In particular, the application of an electric field on single and coupled QDs and the resulting quantum confined Stark effect (QCSE) has been theoretically [4, 5, 6] and experimentally [7, 8, 9, 10] investigated, in order to gain insight into the spatial distribution of the carriers and into the effects of possible strain-induced or built-in electric fields. In recent years an increasing attention has been devoted to QDs as candidates for a solid-state implementation of quantum-information processing (QIP). Many possible schemes have been proposed, involving the use of either spin [11] or orbital [12] degrees freedom as qubits. The latter include those that identify the computational degrees of freedom with specific exciton levels and propose to manipulate them on the ps timescale by means of coherent-carrier control techniques [13, 14, 15]. In each of these QD-based schemes the coupling between dots is a crucial issue and is generally supposed to be tuneable by means of external electric or magnetic fields. In particular, the use of the electric field has been suggested in order to induce relevant dipoles in the exciton states, which would result in a dipole-dipole interaction between excitons (qubits) localized in neighbouring dots [14]. The more critical aspects of similar proposals are related to the need of spectrally resolving each single qubit within a large array and to the efficient decohering phenomena, such as the radiative recombination of the electron-hole pairs [16], that result in decoherence times between tens and hundreds of ps. In these two respects an increased tuneability of the exciton energy and of the oscillator strength might represent a relevant improvement: one of the main goals of the present work is to demonstrate that this can be achieved by switching from single to coupled QDs. In particular, I will focus on the range of electric-field values that is compatible with finite exciton oscillator strength, as required in order to allow optical manipulation of the exciton states. As a first step, I compute the single-particle eigenfunctions and eigenvalues for electrons and holes within the envelope-function and effective-mass approximations [1]. The external
confinement of the carriers in the double dot is described by a prototypical confinement potential, which is double-box-like along \( z \) and parabolic in the \((x,y)\)-plane; an additional term in the single-particle Hamiltonian \( H_{SP} \) accounts for the electric field \( F \), directed along \( z \):

\[
H_{SP} = \sum_{\alpha=e,h} \int \tilde{\psi}_\alpha^\dagger(r) \left[ -\frac{\hbar^2}{2m^*_\alpha} \nabla^2 + V_{DW}^\alpha(z) + \frac{1}{2} m^*_\alpha \omega_\alpha^2 (x^2 + y^2) - q_\alpha F z \right] \tilde{\psi}_\alpha(r) \, dr,
\]

(1)

where \( \hat{\psi}_e(r) \) and \( q_e \) (\( \hat{\psi}_h(r) \) and \( q_h \)) are the electron (hole) field operators and electric charge respectively, \( V_{e,h}^{DW}(z) = V_{e,h}^0 \left[ \theta(|z| - d/2 - l) + \theta(d/2 - |z|) \right] \), \( l = 10 \text{ nm} \) and \( d \) are the width of the wells and the interdot barrier respectively, \( V_{e,h}^0 = 400,215 \text{ meV} \) are the well depths for electrons and holes.

Starting from these ingredients, I compute the interacting electron-hole states by means of a full configuration interaction (CI) calculation \[17\]. For each number of electron and holes, I generally truncate the Hilbert space by keeping only the \( N_H \) configurations of lowest single-particle energy, \( N_H \) being determined on the grounds of a convergence criterion. Within such subspace, I diagonalize the Hamiltonian matrix \( H_{SP} + H_C \), where

\[
H_C = \frac{1}{2} \sum_{\alpha,\beta=e,h} \int \int \frac{dr \, dr'}{(\kappa |r - r'|)} \hat{\psi}_\alpha^\dagger(r') \hat{\psi}_\beta^\dagger(r) \hat{\psi}_\beta(r) \hat{\psi}_\alpha(r')
\]

(2)

accounts for the carrier-carrier Coulomb interactions and for the resulting correlation effects (\( \kappa \) is the semiconductor dielectric constant, \( r \neq r' \)). In the investigation of the single exciton ground state 20 SP states for both electrons and holes are kept and no further truncation of the Hilbert space is performed (therefore \( N_H = 400 \) configurations).

As a preliminary step, let me consider the effects of the interplay between tunneling and electric field on the SP states, which is described by the \( z \)-dependent part of \( H_{SP} \). In Fig. 1 I plot the distributions along \( z \) corresponding to the electron and hole SP groundstates as a function of \( F \) and of the interdot distance \( d \). These eigenstates result from a competition between the coherent coupling, that tends to delocalize the wavefunctions over the double dot, and the field, that instead tends to localize them in one dot or in the other. While the tunneling-induced energy splitting between the bonding and antibonding states is conventionally denoted by \( 2t \), the one that the field induces between states localized in two different dots is approximately given by \( 2\Delta \equiv e(d + l)F \). The ratio \( x \equiv \Delta/t \) thus provides a good
estimate of the relative strength of such two effects and of the resulting degree of localization of each carrier: this is found to increase from electrons to holes (typically \(m_e^* < m_h^*\) and thus \(t_e > t_h\)) and with increasing \(d\) (see Fig. 1(a-d)). In the same range of values of \(F\) the SP eigenstates and eigenfunctions corresponding to a single dot (not reported here) are hardly affected by the field. Within the present picture, such a difference can be understood by thinking of the single QD as a double dot in the in the limit \(d \to 0\): the values of \(x \equiv \Delta/t\) corresponding to an electric field \(F \leq 8\) kV/cm turn out to be very small both for electrons \((x_e \leq 0.038)\) and for holes \((x_h \leq 0.18)\).

The structural parameters which determine the carrier confinement in each dot, namely \(l\) and \(\hbar \omega_{e,h}\), are kept constant throughout the following calculations: while a detailed understanding of their role would require a full exploration of the parametric space, a few comments can be done on the grounds of the present results. In particular, the possible reduction of the well width \(l\) would mainly result in an enhancement of the carrier penetration in the barrier and therefore of the interdot coupling and of \(t\), whose role and interplay with the electric field are discussed above. The in-plane confinement, instead, does not directly affect the interdot coupling (and therefore \(t\) or \(x\)). A possible increase of \(\hbar \omega_{e,h}\) would essentially produce an enhancement of the intradot Coulomb matrix elements and therefore of the Coulomb-induced effects (see the discussion below).

The properties of the exciton states result from the interplay between the abovementioned features of the SP states and the electron-hole Coulomb interaction. In Fig. 2 I plot the Stark shifts and the oscillator strengths of the exciton groundstate \(|X_0\rangle\) as a function of \(F\) and \(d\) (panel (a)) and the contributions to the total energy arising from \(H_{SP}\) and \(H_C\) (Fig. 2(b-c)); the results for the single dot are also reported for a comparison. Quite generally, the magnitude of the QCSE is greatly enhanced and the wavefunctions are much more affected by the field in coupled dots as compared to single ones. While in single QDs the Stark shift \(\Delta E \equiv E(F) - E(0)\) typically shows a quadratic dependence on \(F\), due to the perturbative nature of the contribution from the field, a nonparabolic behaviour is expected in the case of “artificial molecules” [5, 6]. The field induces a monotonic decrease of the single-particle energy \(E_{SP}\), which determines the negative sign of the Stark shift, and a progressive spatial separation of the carriers, as demonstrated by the increasing Coulomb energy \(E_C\) and oscillator strengths (Fig. 2(a-c)).

A closer inspection of the plots reveals the occurrence of two regimes. In the strong-field
region \((F \gtrsim 6 \text{ kV/cm})\) the overlap between the electron and hole distributions rapidly decreases and \(\Delta E\) depends quasi linearly on \(F\) (as in the case of a field-rigid dipole interaction): both trends are enhanced at larger values of the interdot distance \(d\). In the weak-field region \((F \lesssim 6 \text{ kV/cm})\) the interplay between tunneling, Coulomb interaction and electric field is more balanced: rather counterintuitively, the energies \(E, E_{SP}\) and \(E_C\) get closer to the single-dot value as \(d\) is increased. As discussed in the following, this can be attributed to the fact that the strength of the interdot coupling determines to which extent the carrier-carrier Coulomb interaction can oppose the effects of the electric field.

As discussed in Ref. [5], the possible presence of strain effects is expected to meaningfully affect the valence-band states and thus the optical properties, e.g., in self-assembled InAs/GaAs QDs. More specifically, the authors show how the biaxial components of the strain field decrease the height of the interdot barrier, increase the energy levels of the confined holes, and induce a stronger confinement of the holes in the upper dot as compared to the lower one. The first two features result in an effective enhancement of the interdot coupling for the holes, the latter implies an asymmetry in the QCSE with respect to the direction of the applied electric field. According to the present analysis, neither of these two effects is required for the anomalous Stark shift to occur in coupled QDs: I thus expect it to be observed also in case of softened or negligible strain fields in and around the dots.

The spatial distribution of the carriers provides some more physical insight in the exciton groundstate \(|X_0\rangle\). In Fig. 3(a)(b) I plot the probability of finding the electron (hole) in the upper dot for \(d = 1, 2\) and \(3\) nm (red, green and blue lines), and in the upper part of the single dot (black dotted line). As expected from the previous discussion on the SP states, the hole is highly localized in the upper dot already at small values of the field and especially for large values of \(d\). On the other hand, the behaviour of the electron again reflects the existence of the abovementioned two regimes. In the strong-field region the electron is seen to approach the lower dot, under the direct influence of the field; as observed in Fig. 2 for the energies, the derivatives of the curves in this region \((|\partial N_{e,h}^{ud}/\partial F|)\) increase with increasing \(d\). In the weak-field region the Coulomb interaction with the hole dominates on the interaction with the field and the electron tends to be localized in the upper dot. This spatial arrangement is also reflected by the small changes in this region of the oscillator strength and Coulomb interaction (Fig. 2(b-c)). The curves plotted in Fig. 3(a) cross at \(F \approx 7 \text{ kV/cm}\): both in the weak- and in the strong-field regimes the degree of localization of the electron, in the upper
or in the lower dot respectively, is opposed by the tunneling. For \( d = 1 \text{ nm} \) the electron is basically frozen on a delocalized bonding orbital and \( N_{ud}^e \) does not deviate significantly from \( 1/2 \) in the considered range of \( F \) values; at large interdot distances (\( d = 3 \text{ nm} \)) \( t_e \) is small enough for the antibonding orbital to be also occupied in order to produce the localization of the electron in either dot, depending on the relative strength of the external- and of the hole-induced electric field.

The polarizability of the exciton groundstate reflects this behaviour. While the value of \( D \) is on average greatly enhanced in coupled QDs as compared to the single one (black dotted vs. coloured lines in Fig. 3(c)), the sign of \( \partial D / \partial d \) actually depends on the range of electric-field values of interest. For \( F \lesssim 6 \text{ kV/cm} \), where the electron tends to follow the hole in the upper dot, the larger degree of localization resulting from the larger values of \( d \) tends to suppress the dipole; for \( F \gtrsim 6 \text{ kV/cm} \), where the electron interaction with the field dominates on that with the hole, a larger degree of localization results in a larger induced dipole. Depending on the detailed balance between SP and Coulomb terms in the Hamiltonian, either the strongly or the weakly coupled QDs show the highest polarizability.

Let me finally give a rough estimate of the dipole-dipole interaction that a field of 8 kV/cm could induce between two pairs of vertically coupled dots, given that \( E_{\text{int}} \approx |\vec{D}_1 \cdot \vec{D}_2 - 3(\vec{D}_1 \cdot \vec{r}_{12})(\vec{D}_2 \cdot \vec{r}_{12})|/\kappa r_{12}^3 \), and with \( \vec{D}_1 \parallel \vec{D}_2 \perp \vec{r}_{12} \), \( |\vec{D}_{1,2}| = 10 \text{ e}\times\text{nm} \) and \( r_{12} = 20 \text{ nm} \). The energy shift produced by the exciton-exciton interaction is \( E_{\text{int}} \approx 1.4 \text{ meV} \), i.e. the same order of magnitude of the exciton-exciton interactions in a single QD of the same sizes.

In the perspective of the QD-based QIP schemes, the large enhancements of the exciton polarizability and of the optical tuneability suggest the identification of the computational degrees of freedom with exciton levels in vertically-coupled dots rather than in single ones, as proposed in previous works \cite{13,14}. The tuneability of the exciton levels by means of local electric fields can be exploited in order to favour the spectral resolution of specific transitions (qubits) within an array of neighbouring dots of similar dimensions and to bring specific transitions in and out of resonance with the laser fields used for the gate implementation. The polarizability of the exciton states, instead, is required in order to induce a relevant dipole-dipole interaction \( E_{\text{int}} \) between excitons that are localized in different and (quantum-mechanically) decoupled dots. In fact, the value of \( E_{\text{int}} \) determines a lower bound to the duration \( \tau_g \) of the required laser pulses (\( \tau_g \gtrsim \hbar/E_{\text{int}} \)): an exciton-exciton interaction of the order of one meV, as the one estimated above, allows in principle to perform the quantum
gates on the ps timescale.

The suggested geometry can be integrated in a planar array of vertically coupled dots: each double dot would couple to its nearest neighbours only by means of the field-induced dipole-dipole interaction, while the in-plane SP tunneling should be suppressed in order for the required tensor-product structure of the Hilbert space to be preserved. As compared to the ones that consider an in-plane electric field, the present geometry has a twofold advantage: (i) it avoids the breaking of the optical selection rules related to the cylinder symmetry of the dot and the resulting thickening of the optica spectra, which interferes with the identification and selective addressing of the single absorption lines; (ii) it does not require the etching of a mesa structure and thus a strong limitation on the number of qubits that compose the quantum hardware.

To summarize, I have investigated the effects of a vertical electric field on the exciton groundstate and on the related optical properties of two vertically coupled QDs. The present results suggest the identification of each qubit with the exciton groundstate in a pair of coupled dots rather than in a single one, in view of the higher degree of tuneability of the linear optical properties, including the energy levels and the corresponding oscillator strength. Besides, the polarizability of the lowest exciton states in artificial molecules is enhanced by one order of magnitude or more as compared to the one in artificial atoms, resulting in relevant dipole-dipole interactions between excitons localized in different pairs of coupled dots. More specifically, due to the interplay between tunneling, carrier-carrier Coulomb interactions and electric field, both the tuneability of the optical properties and the polarizability are maximized either in the strong- or in the weak-coupling regime, depending on the range of $F$ values.

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FIG. 1: Distribution $\rho_{e,h}(z)$ of the electron (a,c) and hole (b,d) eigenstates as a function of the applied field $F$ for $d = 1$ nm (a,b) and $d = 3$ nm (c,d), where $\rho_{e,h}(z) = \iint |\psi(\mathbf{r})|^2 \, dx \, dy$. The degree of localization depends on $x \equiv \Delta/t$, which is proportional to $F$, ranging from 0 to 8 kV/cm. The values of the remaining physical parameters of the physical parameters are the following: $\hbar \omega_{e,h} = 20, 3.5$ meV; $l = 10$ nm for the well width; $V_{0}^{e,h} = 400, 215$ meV for the well depth (band offsets). The effective masses and the dielectric constant are the ones of GaAs (wells) and AlGaAs (barrier).

FIG. 2: (a) Quantum confined Stark effect as a function of the applied field for vertically coupled QDs with interdot distances $d = 1, 2$ and 3 nm (red, green and blue squares respectively) and for the single dot (white squares). The sides of the squares are proportional to the intensity of the corresponding optical transitions and thus to the square moduli of the oscillator strengths. The contributions to the exciton energy arising from the single-particle ($\langle X_0| H_{SP}| X_0 \rangle$) and from the Coulomb terms ($\langle X_0| H_{C}| X_0 \rangle$) in the Hamiltonian are plotted in the (b) and (c) panels respectively, with the same convention for the colours.

FIG. 3: Probability of finding the electron (a) and the hole (b) in the upper dot for $d = 1, 2$ and 3 nm (red, green and blue lines respectively), where $N_{ud}^{e,h} = \langle X_0 | \int \hat{\psi}^{\dagger}(\mathbf{r}) \hat{\psi}(\mathbf{r}) \, d\mathbf{r} | X_0 \rangle$; the black dotted line is the probability of finding the carriers in the upper half of the single dot. (c) Dependence on $F$ of the induced dipole $D = \langle X_0 | \int z \, (\hat{n}_h - \hat{n}_e) \, d\mathbf{r} | X_0 \rangle$, with the same convention for the colours.
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