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Lateral excitonic switching in vertically stacked quantum dots

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We show that the application of a vertical electric field to the Coulomb interacting system in stacked quantum dots leads to a 90° in-plane switching of charge probability distribution in contrast to a single dot, where no such switching exists. Results are obtained using Path Integral Quantum Monte Carlo with realistic dot geometry, alloy composition and piezo-electric potential profiles. The origin of the switching lies in the strain interactions between the stacked dots hence the need for more than one layer of dots. The lateral polarization and electric field dependence of the radiative lifetimes of the excitonic switch are also discussed.

I. INTRODUCTION

Even the simplest circuits need a switch to stop, start or redirect the flow of current, photons or bits. Switching the state of a system by interaction of the matter with a single photon or an external fields opens new possible applications in measurements1 and quantum computing.2 Switches based on an electron-hole pair system (exciton) have been successfully realized in coupled quantum wells and implemented into an exciton system (exciton) have been successfully realized in coupled quantum dots using realistic dot geometry, alloy composition and piezo-electric potential profiles. The origin of the switching lies in the strain interactions between the stacked dots hence the need for more than one layer of dots. The lateral polarization and electric field dependence of the radiative lifetimes of the excitonic switch are also discussed.

II. QUANTUM DOT MODEL

The switching effect we report arises from the interaction of the two-body excitonic state with piezoelectric fields in the strained GaAs layers between dots. To perform a quantitative study of the switching effect, it is necessary to have a numerical technique that includes both detailed modeling of the materials properties of the dots and accurately solves for the correlated two-body excitonic state. We use an atomistic model of the self-assembled dots to accurately model the strain fields. Strain-modified band offsets are used to construct a detailed, three-dimensional effective mass Hamiltonian for the electron and hole.

The model of the quantum dot used is shown in Fig. 1. The single structure Fig. 1(a) is a 4-nm-tall In0.5Ga0.5As truncated-conical dot of a base radius 10.2 nm and top radius 7.2 nm, located on a 0.9 nm thick In0.3Ga0.7As wetting layer. A typical simulation con-
tains approximately $1.7 \times 10^7$ atoms placed in a random alloy structure as described in Ref. 14, and immersed in a GaAs matrix. The triple dot system, Fig. 1 (c), is formed by stacking single structures in the vertical direction with a separation $3.4 \text{nm}$ between the top and bottom of the adjacent dots. The figure also shows iso-surfaces of the piezo-electric fields in the strained GaAs matrix surrounding the dots. Triple dot structures have been simulated in a similar fashion.

Lattice mismatch strain generates strong induced piezoelectric potentials. The valence force field method (VFF)\textsuperscript{15} is used to relax the atoms from their initial zinc-blende lattice distribution to their equilibrium positions, and the resulting relaxed positions and residual forces are then used to compute the stress tensor for grid. The contribution to the stress tensor is calculated for each atom from the VFF potentials. We approximate the local strain field from the stress tensor averaged over eight-atom cubic cells and bulk elastic constants.\textsuperscript{16} The piezo-electric fields are then calculated from the strain field including both linear and non-linear terms.\textsuperscript{17}

\begin{equation}
 p_\mu = \sum_{i=1}^{6} e_{\mu i} \eta_i + \frac{1}{2} \sum_{i,k=1}^{6} B_{\mu ik} \eta_i \eta_k ,
\end{equation}

where $e_{\mu i}$ is the third-rank proper piezoelectric tensor of the unstrained compound, $B_{\mu ik}$ describes the first-order change of the piezoelectric tensor due to strain and $\eta_i$ represents six independent components of the strain tensor (in the Voigt notation).\textsuperscript{17} We calculate the piezoelectric charge densities by taking the divergence of these fields, then calculate the resulting piezoelectric potential, $\Phi_{pz}$. While detailed comparisons are hard to make given the number of material parameters involved the piezoelectric potentials thus obtained are in good agreement with the analytic approach of Davies.\textsuperscript{18}

Typical piezo-electric potentials for single and triple dots are shown in Fig. 1 and these, as will be shown, significantly perturb the electronic states in the structure. The strong piezoelectric field in the GaAs barrier region is also crucial to the lateral switching operation.

Electrons and holes experience different confining potentials in the $[110]$ and $[1 \bar{1} 0]$ directions. Similarly in coupled structures confinement varies not only on two perpendicular diagonals but also from dot to dot. In Fig. 2 the valence band profile across the single and triple dots structure is plotted. The LH energy calculated from the biaxial strain shift is also indicated and compared to the ground and first excited state of the HH in the single and triple nanostructure.

For clarity the lateral confinement potential in Fig. 2 is averaged over the vertical extent and over $[110]$ and $[1 \bar{1} 0]$ directions. In the single dot the mean value on two diagonals is plotted. It is evident that in the triple dot the biaxial strain drives the LH states towards the bottom of potential well. In the single structure the impact of the LH is also weakened as the LH branch is localized in the surrounding matrix away from the dot. It is worth mentioning that in both types of structures the spin-orbit splitting energy band $\Delta = 0.3 \text{eV}$\textsuperscript{19} greatly exceeds the range of confining potential and similarly to the LH branch, do not affect our calculations. We conclude, in agreement with\textsuperscript{7,8} that the HH exciton dominates in InGaAs dot structures.

In order to illustrate the profile of the valence and conduction band in the single and triple quantum struc-

FIG. 1. Cross-section of model quantum dots with piezoelectric potentials of truncated-conical dot. The calculated piezoelectric potential iso-surfaces are also shown.
The band profile is plotted in the growth direction [001] along the axis of the dot in the inset to Fig. 2.

![Lateral valence band profile](image)

**FIG. 2.** Lateral valence band profile of (a) single dot and (b) triple dot structures averaged over the vertical extent, and over [110] and [110] directions. Also indicated are the ground state (gs) and the first excited state (1st es) of the single particle heavy-hole in the single and vertically stacked structure. Insets shows (c) single dot and (d) triple dot conduction (c.b.) and valence band (v.b.) profile in the growth direction along the axis of the dot.

These confining and piezoelectric potentials are used in a two-band effective mass Hamiltonian,

\[
H = \frac{p_e^2}{2m_e} + \frac{p_h^2}{2m_h} + V_h(r_h) + V_e(r_e) - \frac{e^2}{4\pi\varepsilon_0\varepsilon_r |r_{eh}|} + e\Phi_{pz}(r_e) - e\Phi_{pz}(r_h) - eE \cdot r_{eh},
\]

where the effective masses are \(m_e = 0.067m_e\) and \(m_h = 0.144m_e\), \(r_{eh} = r_e - r_h\) and the dielectric constant \(\varepsilon_r = 12.5\). The uniform external electric field, \(E\), is used to drive the excitonic switching.

### III. THE PATH INTEGRAL METHOD

The switching behavior relies on (i) details of the three dimensional geometry, (ii) the strong piezoelectric fields inside and outside the dots, and (iii) electron-hole correlation from strong electron-hole Coulomb interaction. The PI-QMC method allows an essentially exact treatment of the model Hamiltonian, Eq. 2. As there is no basis-set expansion or trial-wavefunction needed, shifting of the quantum state between dots, in the inter-dot barrier region, and the cusp as the electron and hole approach each other are all represented without variational or basis-set bias. This allows an accurate prediction of experimentally observable excitonic polarizabilities and radiative recombination rates, quantities which are known to require treatment of quantum correlations.

The essence of the PI-QMC method is to write the excitonic thermal-density matrix as a path integral,

\[
\rho(\{r_e, r_h; \beta\}) = \frac{1}{Z} \int D\{r'_e, r'_h\} e^{-S_E[R(\tau)]/\hbar},
\]

where \(R(\tau)\) is the imaginary-time trajectory of the electron and hole, \(S_E\) is the Euclidian action for the model Hamiltonian, Eq. 2, and \(\beta = 1/k_bT\) with temperature \(T\). This path integral is evaluated with Monte Carlo sampling as described in Ref. 16. We perform the calculations at a low temperature (12 K) so that the thermal density matrix describes the ground state excitonic wavefunction.

#### A. Calculation of exciton energy, probability density, and pair correlation functions

Properties of the exciton wavefunction are calculated from the thermal density matrix as the path integral is sampled. We give a brief description here, and further details can be found in Ref. 16.

The energy is calculated from the thermodynamic energy estimator, \(E = -(1/\beta)(dZ/d\beta)\). Potential energy contributions arise from the physical location of the electron and hole trajectory. Kinetic energy contributions come from quantum confinement, the extent to which the confining potential and Coulomb forces prevent the trajectories from extending as much as they would for free particles.

The probability density and pair correlation functions can be calculated directly from the electron and hole trajectories in imaginary time. This is very similar to how the quantities would be calculated in a classical...
simulation, yet the path integral includes all quantum confinement and quantum correlation effects. This intuitive aspect of path integrals is particularly appealing for studies of quantum nanostructures.

B. Calculating polarizability

The effect of an external electric field may be treated by the direct inclusion of an electric field in the Hamiltonian, in which case energies, charge densities, pair correlation functions, and radiative rates may be collected as a function of applied electric field. There are two difficulties with the direct application of an electric field. First a separate PI-QMC calculation must be performed for each electric field choice. Second, the estimation of the polarizability requires an accurate calculation of the total energy of the exciton, since the polarizability is the quadratic dependence of the recombination energy with the applied field.

Linear response theory may also be used to compute polarizabilities of exciton in the presence of modest electric fields\textsuperscript{17,20}. In linear response theory, the polarizability is given by the imaginary-time fluctuation of the electron-hole dipole moment. This allows the polarizability tensor to be calculated from a single PI-QMC simulation without an applied field. Further, linear response theory uses the electric dipole moment, which has much less Monte Carlo noise than energy differences, allowing a faster determination of accurate polarizabilities. One limitation of linear-repose theory is that it may miss non-linear effects of stronger electric fields.

In this work, we combine both approaches. We explicitly include the strong vertical ([100]) electric fields that drive the switching. This is important because the interesting switching behavior is non-linear. This also allows us to calculate charge densities, pair correlation functions, and radiative recombination rates as a function of the vertical field strength. For comparison to experiments, we also calculate the lateral polarizability of the exciton in its two different switched states. We use dipole-dipole fluctuations and linear response theory to calculate the lateral polarizability. Thus, we obtain the response of the exciton to a weak, probing electric field as a for a range of strong vertical fields.

C. Calculating radiative recombination rates

We calculate radiative recombination rates as described in Ref. 14. The mathematical formalism follows directly from Fermi's golden rule for spontaneous photon emission. For excitonic recombination in quantum dots, there are two physical aspects to the radiative recombination rate: (1) the electron and hole must meet each other to recombine, and (2) quantum coherence of the electron-hole contact across a large volume enhances the radiative recombination. The first consideration leads to larger rates when the electron and hole have higher overlap, for example, in a small dot, or because of strong electron-hole correlation. The second consideration leads to larger rates if electron-hole correlation allows the exciton to maintain coherence across a larger nanostructure. For the present stacked-dot geometry with external fields, the asymmetry of the electron and hole confining potential diminishes the second effect, so radiative recombination rates are primarily affected by the amount of electron-hole contact in the wavefunction.

The radiative rate is sampled by allowing the path integral, Eq. 3, to include two configurations: (1) a direct contribution with closed electron-hole paths that that samples the trace of the density matrix, and (2) a radiating contribution where the electron and hole connect at a single point at imaginary times $\tau = 0$ and $\tau = \beta \hbar$. The rate follows directly from the ratio $\bar{\tau}$ and $\beta \hbar = 0$ of the path integral for the two configurations. Further details of the radiative recombination rate calculations are given in Ref. 14.

IV. RESULTS

We begin with a discussion of the charge distribution in a single-dot structure, with and without an applied electric field. Next, we discuss the switching effect in the triple-dot structure, which is much richer due to both the the additional vertical structure as well as the strong piezoelectric fields between the dots. To gain further insights into the switching we show pair correlation functions for the excitonic states in single and triple dots, as a function of applied electric field. We have also completed calculations for double dots (not shown) whose behaviour is found to lie between those of the single and triple dots described below. Finally, we present calculations of radiative recombination rates for the correlated excitonic states.

A. Charge distribution in single and stacked dots

The simplest way to understand the switching behavior in stacked dots is to visualize at the charge distributions of the electron and hole. As will be shown, for single dots, these charge distributions are unremarkable and serve as a baseline for the absence of switching. For stacked dots, the charge distributions are interesting in both the vertical and lateral distribution. The switching behavior can be seen as a 90° rotation of the lateral charge density, driven by a vertical applied field.

1. Charge distribution in single dots: Absence of switching

Electrons and holes are subject to different confining potentials in the growth direction, Fig. 3(a) and (b). This results in a spatial separation of electrons and holes producing a permanent dipole moment. The dipole couples to any external electric field which will
pull (push) the electron-hole system apart (together) in the vertical direction and affects its oscillator strength. The size of the dipole depends strongly on the dot composition profile, in particular on the In concentration.\textsuperscript{25}

We theoretically study the influence of two opposite orientations of a static electric field, $E$, along the growth direction on the bound states in the quantum dot.

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The slice through the centre of a single dot in the [1\overline{1}0] direction, Fig. 4(g)-(i), shows the charge density profile of the heavy-hole exciton as a function of the applied field. It can be seen that the quantum dot has an small inverted (electron-over-hole) dipole with 0 kV/cm applied electric field. The wider lateral confinement near the bottom of the conical pyramid is more effective in localizing the heavy hole than the light electron inducing this small dipole. Stronger dipoles induced by In gradients and the resulting strain profiles are well-known\textsuperscript{25}. The piezoelectric potential adds to the strain fields and perturbs the valence band which enhances localisation of the hole wave function in the [110] direction, Fig. 3(c). The conduction band is only weakly affected by biaxial strain allowing the electron charge distribution to be delocalised in the structure and even tunnel into the barrier material. The piezoelectric potential interacts with conduction band, but as shown in Fig. 3(a) the minima are shallower and appear to not directly overlap with the confining potential in the structure. The vertical electric field applied to the system stretches the electron-hole pair reducing the exciton binding energy. The orientation of the electric field has also a significant impact on the polarity of the exciton in the quantum dot as shown in Fig. 4(g),(i). In the lateral direction there is little effect of switching the electric field as seen in Fig. 4(a)-(f).

In summary, and by way of context setting for the triple dot case presented below, we have shown that the interplay between electric field and piezoelectric potential with an exciton in a single, self-assembled quantum dot is insufficient to induce spatial localisation of the charge distribution in the plane perpendicular to applied field. This contrasts with the case of a single ring which does exhibit switching\textsuperscript{13}.

\section{Charge distribution in stacked dots: Lateral switching}

In contrast to single dots, a stacked system made of two or three, vertically stacked dots demonstrates the interesting phenomena of lateral switching of charge distribution as we now show.

In Fig. 5 the charge distribution of electron, hole and exciton in a triple quantum system is presented, to contrast with Fig. 4 for the single dot. In contrast to the single dot, the most striking feature of triple quantum system is a strong, diagonal localisation of the charge distri-
bution which we can accurately switch in the plane of the quantum dot by changing the direction of the perpendicular electric field, Fig. 5(a)-(f). It is important to note that probability distribution in the top dot for the electron and the hole is 90° rotated with respect to the bottom one. This affects the picture and dilutes the sharp diagonal distribution. In fact we are looking down through all three quantum structures from the top, therefore we see the residues of the distribution in middle and bottom quantum dots.

The much larger volume of alloy in the coupled structure compared to the single quantum dot induces a stronger piezoelectric potential. The piezoelectric field perturbs the confinement for the electron and hole, Fig. 3(c) and (d). An interesting characteristic of the triple dot system is that the piezoelectric field originating from the central dot is canceled by fields from the top and bottom structures. This leads to the symmetry along the [110] and [110] diagonals for the middle dot. The piezoelectric potential penetrates into the top and bottom structures creating local minima and maxima in the dot confining potential, Fig. 3(c) and (d).

In Fig. 5(b) the top view of the electron charge probability distribution can be compared to the electron distribution in the vertical cross-section Fig. 5(h) in the absence of external electric field. We see strong delocalisation of the electron probability density in the whole dot volume and uniformly in all three quantum dots. The hole charge distribution however, is strongly confined to the central region of each of the dots, Fig. 5(e). In Fig. 5(h) it is also apparent that the hole charge distribution is shifted toward the base of each dot with highest probability to be found in top and bottom dots. As already indicated the confinement differs in all three structures and also along diagonals in the plane of the dots. The top and bottom dots are characterised by deeper wells compared to the middle one. The repelling action of positive piezoelectric lobes and different strength of confinement on the diagonals naturally favours localisation of the hole in an outer dot, as much more energy is required to overcome the barrier and transit to the central structure. This is clearly visible in Fig. 5(h).

### B. Electron-hole pair correlation functions

Further insight into the electron-hole distribution in the single and coupled quantum dot systems can be obtained from the pair correlation function \( g(r) \). In Fig. 6 the pair correlation data are plotted for the single and triple dot, with and without an electric field. We study two opposite direction of the electric field at ±30 kV/cm. As before, we start with the single dot system without an electric field and compare it to the coupled structure. In Fig. 6(b) the pair correlation function (PCF) for the single and triple system is plotted in the absence of an electric field. The exciton in the single dot is strongly bound by the Coulomb interaction and the particle separation is close to physical height of the structure. This separation can be slightly greater, as the electron is free to tunnel into the surrounding material, and the hole favors the wider bottom of the structure. An increase of the electric field pulls the exciton apart, this weakens the Coulomb interaction and confinement, allowing the charge distribution to spread across and even outside the dot.

The PCF of the triple dot structure, Fig. 6(b), shows some additional features not present in the single quantum dot. One can see two distinguishable maxima at 4 nm and 8 nm, Fig. 6(b). The first, lower maximum, corresponds to the spatially direct exciton when the particles occupy the same structure. The second one indicates the spatially indirect exciton when the electron and the hole occupy neighbouring dots.

The application of a vertical electric field to the system affects the single and multiple structures in a completely different ways. The plot of the PCF, Fig. 6(a) for the single quantum dot hardly changes when the external field is applied. The strong confining potential of the structure keeps the exciton localised in the dot. The hole, which is much heavier than the electron, localises towards the base of the quantum dot. The electron on the other hand is delocalised in the whole
structure, including tunnelling into the barrier material. The positive hole and the negative electron form the electric dipole. Application of an external electric field stretches/compresses the electron-hole pair in the vertical direction. Changing the orientation of the field flips the system’s overall polarity, however the electron-hole separation is only marginally affected.

The coupled quantum dots are far more sensitive to the change of environment compared to the single one, Fig. 6(a). We see that even small changes in applied field, yields substantial variations in the charges’ relative position. For $E = \pm 30 \text{kV/cm}$ the most energetically favourable configuration is the one in which the electron and hole are confined to neighboring dots. The charge carriers tunnel between the middle and outer quantum dots, however the lowest to middle transition dominates. The electric field creates a dipole by stretching the exciton towards the top and base of opposite dots, promoting inter-dot tunnelling. The electron and the hole tend to occupy neighboring structures, Fig. 6(c), in this case the bottom and the middle dot(see Fig. 5(i)). When the sign of $E$ is negative (Fig. 6(a)) the occupancy of the middle and top dots dominates.

In the above section we have shown that an exact treatment of the electron-hole Coulomb potential, as well as a full description of the strain and piezoelectric fields, in the PI-QMC simulations, demonstrates that lateral switching, hitherto only predicted to occur in quantum rings, may also be found in stacked quantum dots. The charge distributions clearly demonstrate the switching between two perpendicular diagonals of the couple quantum dot systems in response to the change of orientation of the vertical electric field. This effect is absent in the single quantum dot due to the much weaker piezoelectric potential within the dot. The localised charge distribution functions for the electron and hole are in excellent agreement with the pair correlation results.

C. Lateral polarizability as a probe of switching

One possibility to measure the lateral switching effect described here would be to monitor the lateral polarizability of the quantum dot. This could be achieved e.g. by use of surface contacts to apply a small fixed lateral electric field and monitor the shift in the photoluminescence as a function of this field. The shift will be proportional to the lateral polarizability hence will change with the exciton switch described earlier. This would be accompanied by an increase in the exciton lifetime as the electron and hole are separated. (see Section D) The observation of these two effects at the same vertical field value would confirm the lateral switching. An second strong indicator of the lateral switching would be to observe a switching in the optical polarization of the photoluminescence emission however, because of band-mixing effects, a quantitative estimate of the efficiency of such switching is beyond the single valence band calculations adopted here.

As the exciton transition goes from spatially direct to indirect, the oscillator strength varies for different values of the external field. Due to the different potentials on planes [110] and [110] the measured oscillator strength is not symmetric around the $0 \text{kV/cm}$ point, as the exciton is attracted by local minima and maxima in top and bottom structure in a different way. Changing the direction of the electric field in the vertical direction lowers the energy on one of the diagonals and promotes the lateral switching between the [110] and [110] directions, (Fig. 5). As a result, an increase in the lateral polarizability tangential to the direction of the confining potential is observed as shown in Fig. 7.

It is important to note that the lateral switch operates only for values of electric field in the range $\pm 30 \text{kV/cm}$. Stronger fields, more than $\pm 40 \text{kV/cm}$, break the exciton. The unbound charge carriers are then attracted and trapped by the piezoelectric extrema and the lateral polarizability remains constant in the triple dot structure. Orientation of vertical electric field still swaps their spatial distribution in the coupled quantum dot, however the lateral switching is not possible anymore, due to its Coulombic nature.

The situation is a little different in a double dot, in which the lateral switching is preserved even for higher values of an electric field. The smaller volume of the alloy in double dot creates a weaker piezoelectric potential which attracts the charges. The electron and the hole are able to tunnel between the top and bottom structures as the dots are relatively close. From a different point of view a strong electric field tries to pull the electron-hole system apart and extend the electric dipole in the surroundings forcing it out of the structure. The sum of the piezoelectric and confining potentials is however strong enough to preserve the Coulomb interaction and sustain the lateral switching. When the orientation of the external electric field is changed the polarizability in the [110] direction is much larger. It is clear, Fig. 7, that the response of the exciton to the electric field in a single dot is extremely weak compared
to the multiple-dot structures. This is caused by the electron and the hole occupying the same structure. The small vertical separation between the electron and the hole creates a much weaker dipole, which in addition is tightly bound by the confining potential of the single quantum dot, and is perturbed less by the associated weaker piezoelectric fields of the single dot. A coupled structure's bright states can be examined experimentally by measuring the lateral polarizability of the exciton, which determines the sensitivity of excitonic energy to an electric field, Fig. 7, and can be used to confirm the lateral switching phenomenon.

D. Effect of lateral switching on photoluminescence

We have also calculated the emission energy (Fig. 8) and radiative recombination rate (Fig. 9) of the single and triple dots. Such calculations are important as if the switched exciton is optically dark due to the spatial confinement. By contrast in the triple dot the electron-hole radiative recombination rate drops off as the applied field is increased and the exciton is essentially dark at fields higher than ±40 kV/cm. For the stronger fields e.g.: ±80 kV/cm, the fields overcome the Coulomb attraction splitting the exciton apart. At this point the electron and hole occupy different structures, wave functions do not overlap any more and the oscillator strength drops essentially to zero.

V. CONCLUSIONS

We have used Path Integral Quantum Monte Carlo calculations to study a heavy-hole exciton in single and multiple quantum dots. We found that the lateral switching of the charge distribution in the vertical electric field, previously reported for quantum rings, is also clearly present and highly tuneable in the system of stacked quantum dots. On the other hand it cannot be observed in a single quantum dot structure. We have also investigated the lateral switching in dots of different shapes, sizes and composition profiles. It was found that this generic phenomena is caused by the interplay of piezoelectric field deformation of conduction and valence bands and the coulomb interaction. Switching was observed for applied fields sufficiently small that exciton dissociation was not a factor hence an optical observation of this phenomenon should be feasible.

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