Excited state quantum couplings and optical switching of an artificial molecule

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We optically probe the spectrum of ground and excited state transitions of an individual, electrically tunable self-assembled quantum dot molecule. Photocurrent absorption measurements show that the spatially direct neutral exciton transitions in the upper and lower dots are energetically separated by only ~2 meV. Excited state transitions ~8–16 meV to higher energy exhibit pronounced anticrossings as the electric field is tuned due to the formation of hybridized electron states. We show that the observed excited state transitions occur between these hybridized electronic states and different hole states in the upper dot. By simultaneously pumping two different excited states with independent laser fields we demonstrate a strong (88% on-off contrast) laser-induced switching of the optical response. The results represent an electrically tunable, discrete coupled quantum system with a conditional optical response.

Quantum dot (QD) nanostructures formed by strain driven self-assembly are ideal for solid-state quantum optics experiments due to their discrete optical spectrum, strong interaction with light and robust quantum coherence for both interband polarization and spin. Embedding them into electrically active devices allows for tuning of the transition frequency and control of charge occupancy. Vertically stacking produces more sophisticated nanostructures with coherent interdot coupling due to carrier tunneling. When combined with the potential to coherently manipulate excitons over ultrafast time scales using precisely tuned laser and electrical control pulses, such systems raise exciting prospects for the operation of small-scale few qubit systems in a solid-state device. Very recently, conditional quantum dynamics for a single resonantly driven QD molecule has been observed and demonstrated a conditional optical response of an artificial molecule.

The sample consists of vertically stacked pairs of QDs separated by a 10-nm-thick GaAs spacer and embedded within the intrinsic region of a GaAs Schottky photodiode. Typical PL and PC measurements recorded at $T = 4.2$ K are presented in Fig. 1. For the PL measurement the sample was excited in the wetting layer at 1.49 eV. Typical electric-field-dependent PL from 18 to 32 kV/cm are presented in a grayscale contour plot representation in Fig. 1. The measurements show an anticrossing of two transitions arising from spatially direct and indirect excitons in the QDM where the hole is located in the upper dot. These optical transitions are depicted schematically in a single particle picture in the inset of Fig. 1: the direct exciton in the upper dot $X_{ud}$ and the indirect exciton $X_{id}$ with the hole in the upper dot and the electron in the lower dot. The indirect exciton exhibits a strong Stark shift due to the large static dipole. As the energies of the two states are tuned to resonance, electron-mediated tunnel coupling occurs that results in the formation of molecular bonding (lower-energy) and antibonding (higher-energy) orbitals and the observed anticrossing. As $F$ increases beyond ~25 kV/cm, the intensity of the luminescence reduces as charge carriers escape from the QDM via tunneling and PC measurements can be performed with resonant optical excitation. Two prominent resonances are observed in PC, examples of which are presented in the left-hand panel of Fig. 1. The energy of these peaks are plotted in the main panel of Fig. 1 for $F = 25–31$ kV/cm. The transitions observed in PC arise from charge neutral excitons and show that the QD molecule exhibits two neutral exciton absorption resonances. As $F$ decreases, these two resonances clearly evolve into the two clear peaks observed in PL, labeled $X_{ld}$ and $X_{ud}$ on Fig. 1. The state marked as $X_{ld}$ is attributed to an exciton in the lower quantum dot of the molecule (depicted in blue in the inset of Fig. 1) while $X_{ud}$ is the direct exciton in the upper dot, assignments that are confirmed by the results presented below.

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hole envelope functions.) Fits to the three anticrossings are
exciton transitions in the upper and lower dots, respectively.
resonances are shown as red squares and blue triangles for the neutral
the indirect exciton as depicted schematically in the inset. The PC
lines indicate the uncoupled neutral exciton in the upper dot and
130 cps from white to black on a logarithmic scale. The dashed red
of the applied electric field is shown as a contour plot from 0 to
dependent PL and PC measurement: The PL of the QDM as a function
of the excited state spectrum of the QDM as a
ular orbital while the hole occupies the first (\(e_1\)) or second (\(e_2\))
excited state as a function of
molecular ground states determined via PL and the first four
states and the two excited state anticrossings in Fig. 2(c).

We conducted detailed PLE measurements to track the
evolution of the excited state spectrum of the QDM as a
function of \(F\). A typical PLE scan detecting on \(X_{ud}\) is
presented in Fig. 2(a) with \(F\) fixed close to the anticrossing
(21.6 kV/cm). Several discrete electronic resonances are ob-
served in this region, the first four of which are labeled \(e_{0B-h_1}, e_{0B-h_2}, e_{0AB-h_1}\), and \(e_{0AB-h_2}\), in Fig. 2(a). This assignment an-
ticipates the nature of these excited states corresponding to
the electron being in the bonding (B) or antibonding (AB) molec-
ular orbital while the hole occupies the first \((h_1)\) or second \((h_2)\)
excited orbital state in the upper QD. These assignments are
now justified by examining the electric-field dependence of the
excited state resonances. Figure 2(b) shows the energy of the
molecular ground states determined via PL and the first four
excited states as a function of \(F\) in the range 20–25 kV/cm
measured using PLE. The first four excited states consist of
two different pairs of lines color coded by the red and blue
symbols in Fig. 2(b), each of which anticross at an electric
field close to 22 kV/cm. To analyze the observed excited state
anticrossings in more detail and compare to the anticrossing
of \(X_B\) and \(X_{AB}\) observed in PL, we plot the energy separation
(\(\Delta E\)) between the bonding and antibonding state of the ground
states and the two excited state anticrossings in Fig. 2(c).

For all three anticrossings \(\Delta E\) shows a similar hyperbolic
behavior that can be fitted with

\[
\Delta E = \sqrt{(2V_{ee})^2 + [ed(F - F_0)]^2},
\]

where \(V_{ee}\) denotes the interdot tunnel coupling strength,
\(F_0\) is the field at which the states anticross, and \(ed\) is
the equivalent static dipole moment of the indirect exciton.
\(d\) is the distance between the centers of the electron and
hole envelope functions.) Fits to the three anticrossings are
presented as lines in Fig. 2(c) and the extracted values of
\(F_0\) and \(d\) are summarized in Table I. For each anticrossing
the extracted values of \(d\) vary only slightly and are fully
consistent with the dot height of 5 nm and the separation of
10 nm. Both \(V_{ee}\) and \(F_0\) remain practically unchanged for the
transitions involving \(h_0, h_1,\) and \(h_2,\) providing evidence that
they arise from transitions between the same electron-mediated
anticrossing and different hole levels. This expectation is
confirmed by PLE measurements performed close to \(F_0,\)
detecting on either \(X_{AB}\) or \(X_B,\) respectively. Typical results
are presented in Fig. 2(a) (inset). When detecting on \(X_{AB}\)
(red curve), transitions are only observed for \(e_{0AB-h_1}\) and
\(e_{0AB-h_2}\), while \(e_{0B-h_1}\) and \(e_{0B-h_2}\) are absent. This arises
since the electron populates the lower-energy bonding level
and thermal activation into the higher-energy bonding level is
unlikely since \(2V_{ee}\gg k_B T.\) In contrast, upon exciting
states with bonding electron character, all four resonances
are observed due to phonon-mediated thermalization from
anticrossing to bonding electron states. The small differences
between \(F_0\) arise from the Coulomb interactions between the
(b) Intensity of direct resonance is quenched due to the presence of the block laser. PL from the excitation of a resonance with direct character (pump, red), indirect character (block, blue), and both lasers (black). PL from the excitation of the resonance with direct character (pump) while exciting $e_{0AB}$-h$_1$ and $e_{0AB}$-h$_2$ is expected to populate $X_{ud}$ while exciting $e_{0AB}$-h$_1$ and $e_{0AB}$-h$_2$ generates an indirect exciton $X_{ind}$. This is used to test whether the QDM exhibits a conditional optical response with a scheme illustrated in the inset of Fig. 3(a). With the condition $F < F_0$ as described above the system is resonantly excited by either one laser or two lasers simultaneously. In the two laser experiment, the first laser termed pump is resonant with $e_{0B}$-h$_1$, as indicated by the red arrow on the inset of Fig. 3(a) (middle arrow). As discussed above, an exciton created by laser absorption will primarily relax to the $X_{ud}$ ground state before PL is measured via phonon-mediated processes. A second laser, termed block, is tuned into resonance with excited states that have a predominantly indirect character in order to generate excitons with indirect character $X_{ind}$. If the absorption of the blocking laser is more efficient than that of the pump, then the QDM will be driven into an indirect exciton state and the absorption of the pump laser is suppressed since absorption

various hole orbital states. Compared to the ground state with $F_0 = 23.1 \pm 0.1$ kV/cm, the critical field of the excited state transitions are shifted by $-0.7 \pm 0.2$ and $-0.9 \pm 0.2$ kV/cm. This can be converted to an energy difference of $1.1 \pm 0.3$ and $1.4 \pm 0.4$ meV using $d = 15.8 \pm 0.2$ and $15.9 \pm 0.2$ nm, respectively. These values correspond to less than 10% of the total attractive $e$-$h$ Coulomb interaction that has been estimated to be $\sim 22$ meV for similar samples. Thus, the Coulomb shifts between the different valence-band states represent a small perturbation and we conclude that these observations provide strong evidence that the four excited states shown in Fig. 2(b) take place between an electron in either the lowest-energy bonding or antibonding levels and different hole states in the upper dot, as depicted in the inset of Fig. 2(b). The energy splitting between the lowest-energy hole state and the first two excited states is 9.5 and 10.5 meV, respectively.

Away from resonance for $F < F_0$ the bonding ground state as well as the first two excited states $e_{0B}$-h$_1$ and $e_{0B}$-h$_2$ have predominant direct character, while the third and fourth excited states $e_{0AB}$-h$_1$ and $e_{0AB}$-h$_2$ have indirect character. Therefore, exciting $e_{0B}$-h$_1$ and $e_{0B}$-h$_2$ is expected to populate $X_{ud}$ while exciting $e_{0AB}$-h$_1$ and $e_{0AB}$-h$_2$ generates an indirect exciton $X_{ind}$. This is used to test whether the QDM exhibits a conditional optical response with a scheme illustrated in the inset of Fig. 3(a). With the condition $F < F_0$ as described above the system is resonantly excited by either one laser or two lasers simultaneously. In the two laser experiment, the first laser termed pump is resonant with $e_{0B}$-h$_1$, as indicated by the red arrow on the inset of Fig. 3(a) (middle arrow). As discussed above, an exciton created by laser absorption will primarily relax to the $X_{ud}$ ground state before PL is measured via phonon-mediated processes. A second laser, termed block, is tuned into resonance with excited states that have a predominantly indirect character in order to generate excitons with indirect character $X_{ind}$. If the absorption of the blocking laser is more efficient than that of the pump, then the QDM will be driven into an indirect exciton state and the absorption of the pump laser is suppressed since absorption

| Hole state | $2V_{xx}$ (meV) | $F_0$ (kV/cm) | $d$ (nm) |
|-----------|----------------|--------------|---------|
| h$_0$     | 3.4 ± 0.1      | 23.1 ± 0.1   | 15.3 ± 0.1 |
| h$_1$     | 3.2 ± 0.1      | 22.4 ± 0.1   | 15.8 ± 0.2 |
| h$_2$     | 3.5 ± 0.1      | 22.2 ± 0.1   | 15.9 ± 0.2 |

Fig. 3. (Color online) (a) PL intensity of $X_{ud}$ for the excitation of a resonance with direct character (pump, red), indirect character (block, blue), and both lasers (black). PL from the excitation of the direct resonance is quenched due to the presence of the block laser. (b) Intensity of $X_{ud}$ as a function of the block laser energy. Whenever the block laser hits an excited state with indirect character, the PL from the upper dot is decreased.

Fig. 4. (Color online) PL of the upper and lower quantum dot as a function of the blocking laser power. Resonant excitation of an exciton in the upper dot is blocked by the presence of an exciton in the lower dot due to the Coulomb interaction. (b) Intensity of the PL of the upper and lower dot and (c) suppression ratio $\rho$ as a function of the blocking laser power.
shifts to a biexcitonic state of the system. Typical results of such a measurement are presented in Fig. 3(a), which shows the PL spectrum recorded for $X_{ud}$ for $F = 21.05$ kV/cm when the pump laser (0.5 kW/cm²) is applied (red curve). This is compared with the situation when the system is excited only by the blocking laser (5 kW/cm², blue curve) and when both lasers are applied simultaneously (black curve).

The intensity of $X_{ud}$ clearly reduces strongly when both lasers are applied simultaneously. We scanned the energy of the blocking laser over the spectrum of excited states from 1322 to 1326 meV. The result of this experiment is shown in Fig. 3(b) comparing the intensity of $X_{ud}$ as a function of the blocking laser energy for the pump laser only (red curve), the blocking laser only (blue curve), and with both blocking and pump lasers applied simultaneously (black curve). Three resonances, labeled $R_1$, $R_2$, and $R_3$ in Fig. 3(b) can be clearly seen. At these resonances PL is observed from $X_{ud}$ following excitation with the blocking laser only. The PL signal for excitation with both lasers shows a series of dips for an excitation with the pump laser only. $R_1$ and $R_2$ coincide precisely with the PLE resonances $e_{0AB}-h_1$ and $e_{0AB}-h_2$, presented in Fig. 2(b), demonstrating that the blocking laser can be used to suppress the absorption of the pump beam.

We now turn to an experimental scheme where an excitation in one of the dots forming the molecule was used to block absorption in the other. The scheme for this experiment is illustrated schematically in the inset of Fig. 4(a). An exciton with predominantly direct character can be excited in the upper dot via its first excited hole state (red arrow). If the blocking laser is tuned to a direct exciton transition in the lower dot (blue arrow) while the pump field is simultaneously on the energy laser is tuned to a direct exciton transition in the lower dot (blue dot via its first excited hole state (red arrow). If the blocking laser over the spectrum of excited states from 1322 to 1326 meV. The result of this measurement is presented in Fig. 4(a), which shows the PL intensity of $X_{ud}$ when blocking and pump lasers are switched on, $I_{01}$, and when only the blocking laser is applied, we measure $I_{00} = 9 1 ± 1$ cps (blue triangles), from which we obtain the suppression $\rho = \frac{I_{01}}{I_{00}} \sim 88 2 ± 2$. The dependence of $\rho$ on the blocking laser power is presented in Fig. 4(c). The reason why the blocking laser-induced PL from $X_{ud}$ results from the fact that the resonance used for efficiently exciting $X_{ud}$ is 22.3 meV higher than the ground state of $X_{ud}$. Therefore, relaxation from this excited state to the ground state of $X_{ud}$ is possible due to tunneling of both charge carriers.

In summary, we probed the spectrum of ground and excited state transitions in an individual, electrically tunable artificial molecule. Excited state transitions were identified between hybridized electron states having bonding or antibonding character and different excited hole states. By simultaneously pumping different discrete optical transitions, we demonstrated a conditional optical response with an on-off gating fidelity of 88 ± 2%.

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