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Exotic quantum couplings in optically active quantum dot molecules

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We report detailed magneto-optical investigations of electron tunnelling and Coulomb quantum couplings of excitonic transitions in electrically tunable quantum dot (QD) molecules. Single molecule photocurrent (PC) absorption, photoluminescence (PL) emission and PL-excitation spectroscopy are combined to carefully map the entire spectrum of spatially *direct* ($e + h$ in same dot) and *indirect* ($e + h$ in different dots) excitonic transitions of a single molecule. Comparison with simulation allows us to obtain a full and consistent picture of the conduction and valence band orbital structure [1].

From PC-measurements we firmly identify the spatially direct neutral exciton transition with both e and h localized in the upper dot (X^0_{ud}). By tracking the electric field dependence of X^0_{ud}, we clearly observe the expected anticrossings arising from tunnel couplings between different electronic orbital states of the QD-molecule [2,3] (fig 1). In addition, a number of *unexpected* anticrossings are observed in PC spectroscopy and shown to arise from Coulomb mediated coupling of excitonic transitions, involving entirely *different* single particle orbitals. By examining the evolution of the tunnel and Coulomb mediated couplings with magnetic field applied in Faraday geometry, we extract new information about the orbital structure of excited hole states in the QD-molecule.

By performing ultrafast optical pump-probe spectroscopy in the PC-regime we obtain new information about ultrafast carrier and spin dynamics in the system. Close to quantum couplings we observe a strong reduction of the electron tunneling time. The results obtained elucidate the roles of ultrafast (<5ps) elastic and inelastic intra-molecular electron tunneling and allow us to directly extract the exciton - acoustic phonon coupling in the system [4]. The ability to electrically switch the system into a regime with ultrafast inter-dot tunneling enables us to optically initialize a single hole spin on timescales only limited by the width of the initializing laser pulse (<5ps) with a purity of >96% (only limited by noise). By monitoring the time evolution of a single coherent hole spin in lateral magnetic fields using optically polarized ultrafast laser pulses we observe coherent Larmor precession of a single hole spin with no observable loss of coherence within the hole lifetime [5].

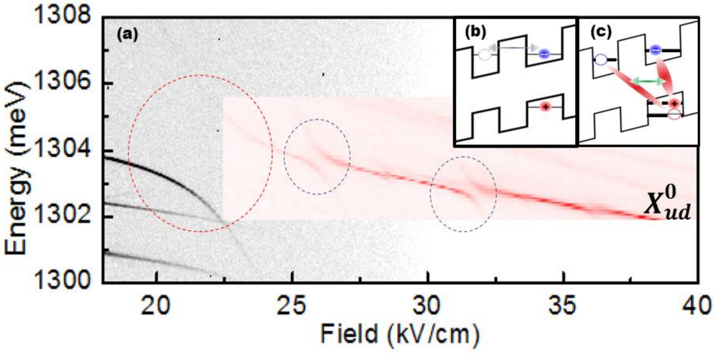

Fig.1 – (a)Typical electric field dependent PL (blue coding) and PC (red coding) spectra showing X^0_{ud} and couplings arising from tunnel and Coulomb couplings at F = 22.5 kV/cm and F=26.1kV/cm. Inset: Schematic representation of tunnel (b) and Coulomb (c) mediated couplings.

References

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