

# Deterministic Generation of a Triexciton in a Quantum Dot

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**Abstract:** We demonstrate deterministic generation of a quantum dot-confined triexciton in a well-defined coherent state using a sequence of three laser pulses.

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Single semiconductor quantum dots (QDs) are known sources of both single photons [1] and polarization-entangled photon pairs. [2] Deterministic generation of higher-order multiexcitons is a conceptual way to increase the number of quantum correlated photons that a single QD emits. Here, we deterministically generate the neutral triexciton ( $XXX^0$ ) which contains three electron-hole (e-h) pairs using a sequence of three laser pulses. The triexciton is generated in a well defined coherent state of its two eigenstates. Its decay by a three photon radiative cascade is demonstrated elsewhere [3].

The QD-confined  $XXX^0$  contains three electron-hole (e-h) pairs. The population of these carriers in their ground state can be approximately described as a pair of electrons and a pair of heavy holes occupying their respective ground energy level forming an antisymmetric spin configuration (a singlet). In addition, there is one unpaired electron and heavy hole in their respective second energy level. It follows, therefore, that the triexciton fine structure fully resembles that of the exciton ( $X^0$ ) where the unpaired e-h exchange interaction fully removes the degeneracy between the four possible e-h spin configurations, as schematically described in Fig. 1(a).

Fig. 1(b) also presents the allowed optical transitions between the ground state biexciton and the triexcitonic states. The splitting between the two optically-accessible triexcitonic states is such that a single laser pulse, wider than the energy splitting between the two states, can write an arbitrary coherent superposition of these two states in a manner similar to that used for writing the bright exciton  $X^0$  in an arbitrary coherent state [5].

The two absorption transitions to the triexcitonic states were identified using three-laser photoluminescence excitation (PLE) spectroscopy (Fig. 2(b)). The three laser pulse sequence is shown in the inset to Fig. 2(b). The first laser is tuned to an excitonic absorption resonance at 29 meV above the  $X^0$  emission energy (Fig. 2(b)). The electron relaxes quickly ( $\sim 7$ ps) to the ground  $X^0$  state in a spin-preserving process [4, 5]. The second pulse is resonantly tuned to the  $X^0 - XX^0$  transition (Fig. 2(a)) and the third laser energy is scanned while the  $XXX^0$  emission line is monitored (Fig. 2(a) [3]). A triexciton absorption resonance is clearly visible (Fig. 2(b)) at  $\sim 34$  meV above the exciton energy, corresponding to the addition of a  $p$ -level e-h pair to the  $XX^0$  and the formation of  $XXX^0$ , as described in Fig. 1(b). The solid red (blue) line in Fig. 2(b) shows the same PLE measurement as that described by the green line, but without the first (second) pulse, verifying that the triexciton PL indeed results only from the three pulses together. Fig. 2(c)

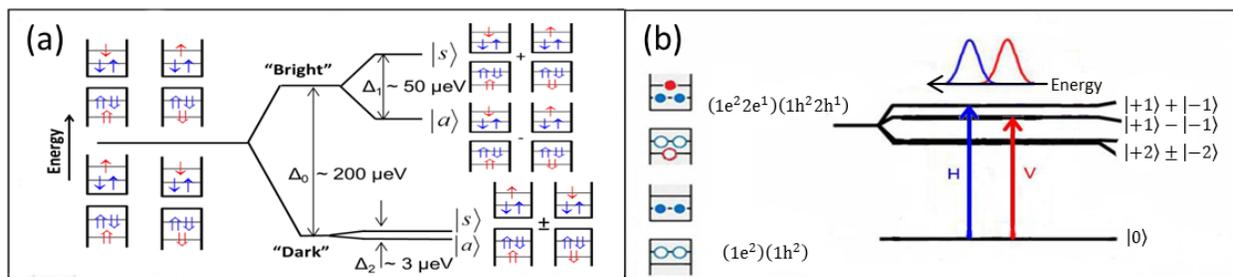


Fig. 1. Schematic of the  $XXX^0$  energy levels and allowed optical transitions from the ground-state biexciton.

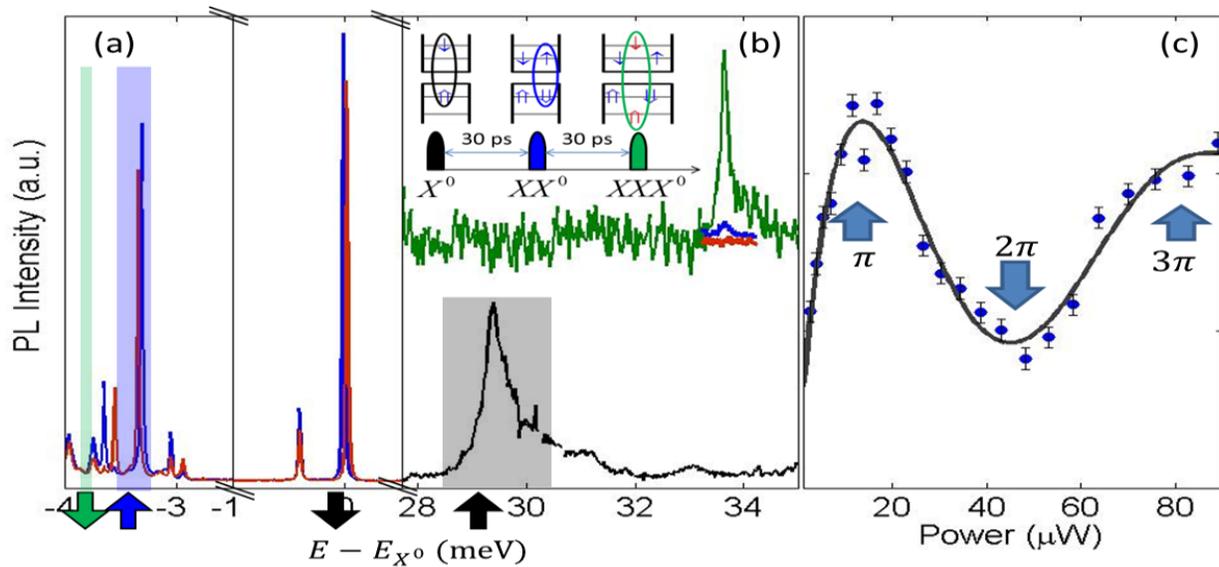


Fig. 2. (a) Rectilinear polarization sensitive PL spectra from a strongly excited quantum dot. The triexcitonic (biexcitonic) [excitonic] emission lines are shaded in green (blue) [black]. (b) Black (green) curve is the PLE spectrum of the  $X^0$  ( $XXX^0$ ) spectral line marked by a downward black (green) arrow in (a). The first (second) laser pulse is tuned to the  $X^0$  ( $XX^0$ ) absorption resonance indicated by the upward black (blue) arrow. The red (blue) curve is the PLE measurement in the absence of the first (second) pulse. The inset shows the three pulse sequence. (c) Measured (marks)  $XXX^0$  PL intensity as a function of the average power of the third pulsed laser tuned to the resonance found in (b). The solid line describes a theoretical fit to the expected Rabi oscillation behavior.

presents the  $XXX^0$  PL intensity as a function of the third resonantly tuned pulse. Rabi oscillations are clearly visible, demonstrating that our three pulse sequence deterministically generates the triexciton. The polarization of the final pulse in the sequence determines the spin configuration of the unpaired  $p$ -level e-h pair [5]. Thus, the  $XXX^0$  can be deterministically generated in any *a priori* well-defined spin configuration.

In conclusion, we have demonstrated, for the first time, deterministic generation of the QD-confined triexciton via a three laser pulse sequence.

## References

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