A tale of three particles: Generation of entangled-photon pair from biexcitonic-to-exciton cascade decay in semiconductor nanoparticles

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Motivation and significance: This work aims to investigate the generation of entangled photon pairs in quantum dots. Entangled photon pairs (EPP) are important in quantum optics and are essential for quantum information, quantum teleportation, quantum key distribution, and controlled logic operations. Semiconductor nanoparticles such as quantum dot (QD) are especially well suited for EPP generation and have been proven to have high-entanglement fidelity, extraction efficiency, and photon indistinguishability.

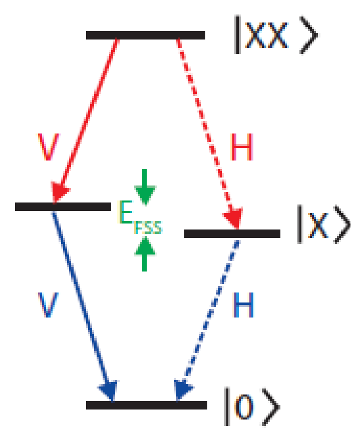

Fig 1. Entangled-photon pair generation from biexciton-exciton cascade. The photons V and H are entangled if the splitting energy (E_{FSS}) is zero.

Theoretical and computational strategy: Accurate description of biexcitonic and exciton states are crucial for accurate prediction of generation of EPPs. In this work, we have used the frequency-dependent geminal-screened electron-hole interaction kernel method (FD-GSIK) for treating the excitonic and biexcitonic states. The FD-GSIK is a first-principles, explicitly-correlated, real-space method that avoids using unoccupied orbitals to construct the electron-hole interaction kernel by performing a complete infinite-order diagrammatic summation of particle-hole excitations and deriving a renormalized R12 real-space electron-hole correlator operator. The FD-GSIK method also bypasses the computationally expensive AO-to-MO integral transformation step by computing all integrals directly in the real-space numerically using Monte Carlo integration. The frequency-dependent component of the FD-GSIK method implements the necessary quasiparticle screening needed for describing biexcitonic states. We have used a time-dependent diagrammatic approach for treating light-matter interactions. Specifically, light-matter interaction is treated using the dressed-atom representation, and the time-propagation is performed diagrammatically using Feynman-Goldstone formulation using field-dependent single-particle states.

Results and future directions: In this work, the combination of dressed-atom and FD-GISK method was applied to investigate exciton binding energies and biexciton binding energies for large quantum dots ($Pb_{140}S_{140}$, $Pb_{140}Se_{140}$, $Cd_{144}Se_{144}$). Chemical insights from biexciton binding energies, exciton binding energies, fine-structure splitting, oscillator strengths, degree of entanglement, and time-dependent electron-hole recombination probability for these quantum dots will be presented. The results from these calculations demonstrate the efficacy of the FD-GSIK method for capturing electron-hole correlation and treating 2-electron 2-hole excitation in large clusters and nanoparticles. Future and ongoing work in this direction include enhancing photon-entanglement characteristics by optimizing surface ligands and adding core/shell heterojunctions will be discussed.

Reference: McLaughlin, P.F. and Chakraborty, A., 2020. Compact Real-Space Representation of Excited States Using Frequency-Dependent Explicitly Correlated Electron–Hole Interaction Kernel. Journal of Chemical Theory and Computation, 16(9), pp.5762-5770.