4D07　Si 及び CdSe 量子ドットにおける多励起子生成・消滅のダイナミクス

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Multiple Exciton Generation and Recombination Dynamics in Si and CdSe Quantum Dots

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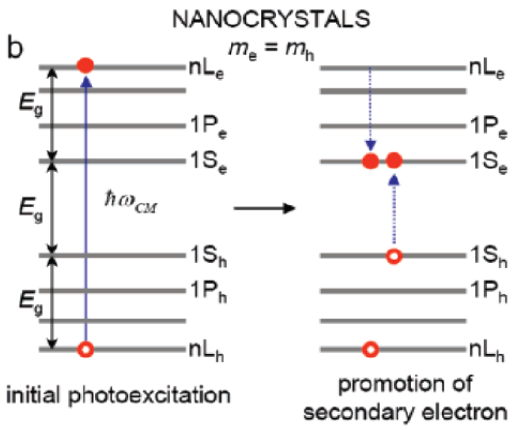
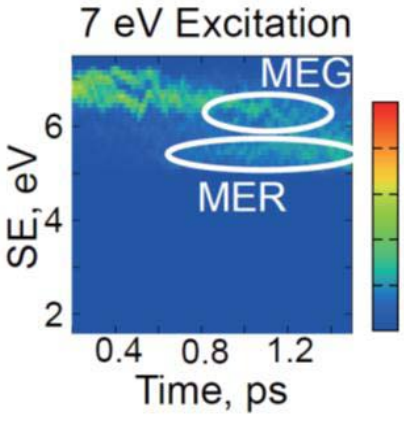
Advantages of quantum dots (QDs) in photovoltaic devices are due to the possibility of generating multiple electron-hole pairs (excitons) upon absorption of a single photon. This is called multiple exciton generation (MEG), and provides great potential for increasing solar energy conversion efficiencies relative to those in bulk systems since the MEG provides new mechanisms for utilization of excess photon energy and avoiding energy loss to heat. We have recently developed a real-time atomistic simulation method for studying the MEG and its inverse process, multiple exciton recombination (MER).[1-3] Our atomistic simulation is performed by directly solving the time-dependent Schrödinger equation based on multiple exciton bases with NA couplings and band energies obtained by the time-domain *ab initio* simulation on a nano material. Our method calculates various real-time dynamics of the MEG and MER treating NA phonon couplings non-perturbatively, and the MEG and MER are allowed to occur simultaneously. Our "atomistic" simulation method can take into account QD size, shape, defects, core-shell distribution, surface ligands, and charge trapping, which significantly influence photoexcited dynamics in nano materials. These are advantages of our method which static electronic calculations of band structures and perturbative rate theories like Fermi's golden rule cannot account for.



Schematic Depiction of MEG

We found the following important insights from our simulations on Si and CdSe QDs: MEG rapidly accelerates with initial excitation energy, reflecting strong energy dependence of double exciton (DE) density of states. At early times, MEG is Gaussian rather than exponential. The exponential dynamics, assumed in the standard rate theories, starts at a later time and becomes more important in larger QDs. Phonon-assisted MEG is observed at energies below the purely electronic threshold due to the presence of high-frequency ligand vibrations. Coupling to phonons is essential for MER since lower-energy DEs can be a main



Interplay of MEG and MER

gateway to recombine into single excitons (SEs). The MER simulated starting from a DE is significantly slower than the MER involving an optical excitation of a SE, followed by the MEG and then the MER. The latter time scale agrees with the experiments, emphasizing the importance of superpositions of many DEs for the efficient MER. The detailed description of the interplay between the MEG and MER coupled to phonon modes provides important insights for the excited-state dynamics in semiconductor QDs and in other nano materials, and also for improving the efficiency of solar energy conversion using the nano materials.

【参考文献】

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