

Charge Transfer in Janus CdSe/PbSe Quantum Dots with N719 Dye

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We simulate the electronic and optical properties of Janus-faced quantum dots (QDs) made of two hemispheres of differing semiconductor materials CdSe and PbSe with the attached Ru(II) dye (N719). Our calculations aim to understand how the QD-to-dye charge transfer is affected by the dye attachment to different surfaces of the QD via different anchoring groups in different solvents using DFT-based non adiabatic dynamics combined with Redfield Theory. We found that the QD-dye binding energy is strongly stabilized for the deprotonated dye, favoring bonding of the dye via a single carboxylic group independent on the QD surface. The hole states originated from the dye appear in the energy gap of the host QD in vacuum and weakly polar solvents but move deep inside the QD's valence band in polar solvents. Thus, a non-polar or weakly polar environment favors thermodynamic conditions for the QD-to-dye hole transfer. The results of charge carrier dynamics induced by selected photoexcitations show that hole relaxation is much faster than electron relaxation in the QD/dye systems, with QD-to-dye hole transfer happening in a few picoseconds. In contrast, hole relaxation is slower than electron relaxation in pristine Janus QD due to slow hole transfer from the Cd-side to the Pb-side, while electron states are hybridized over the Pb-Cd interface. Obtained results provide atomistic insights for optimizing charge transfer between a QD and its environment – an important property for photovoltaic and photocatalysis applications.