Janus-like Quantum Dot Interface Charge Transfer Properties With and Without a Bond Connection Through Space

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The two materials of a Janus-like quantum dot (QD) are conjoined at an interface, with one material acting as an electron donor and one acting as an acceptor. This ability to separate charge makes Janus-like QDs useful in applications such as photovoltaic solar cells, where the ability to separate charge allows one to harvest charges before they recombine. The two interfaces of the QD also can allow for a current to move between them. The properties of the interface determine the efficiency of electron-hole recombination rates and charge transfer. Therefore, a fundamental understanding of how this interface works between the two materials can better inform industry about the construction of Janus-like QDs so that the QDs can be more efficient with charge transfer. To contribute to this understanding, we simulated and compared the properties of isolated individual nanocrystal (NC) models of $Cd_{33}Se_{33}$ and $Pb_{68}Se_{68}$ to two isolated Janus-like models which had one of each of these NCs ($Cd_{33}Se_{33}$+$Pb_{68}Se_{68}$). We also studied the interface properties of the two Janus-like QD models. The first Janus-like model has a bond through space connecting the two NCs and is approximately 16x17x29 $Å^3$ in size. The second model does not have a bond through space between the NCs and is approximately 16x17x31 $Å^3$. We use density functional theory to simulate the ground state properties of these models, including the density of states, absorption spectra, and Kohn-Sham orbital charge densities. The ground states are then excited and used as a basis for nonadiabatic on-the-fly couplings. Redfield Tensor calculations are then used to study the excited state dynamics, which yields the relaxation rates and charge transfer electrons and holes in the models. From our results, we found that the Janus-like models have smaller energy band gaps than their individual components. Our simulated absorption spectra for the Janus-like models are comparable to experimental results [i]. We also identified that having a bond connecting the two NCs at an interface reduces the hole relaxation time. Both the bonded and unbonded models have energy transitions that are very likely to occur near the highest occupied molecular orbital (HOMO) and lowest occupied molecular orbital (LUMO) of their models, which results in a comparable total charge transfer during relaxation for both models. However, the model without a bond through space also has some highly probable energy transitions that occur from orbitals that are significantly further from HOMO-LUMO. The electron-hole relaxation dynamics from these further out orbitals in the Janus-like model without a bond through space resulted in a comparably higher charge transfer.

i. Zhang, J., Chernomordik, B. D., Crisp, R. W., Kroupa, D. M., Luther, J. M., Miller, E. M., . . . Beard, M. C. (2015). Preparation of Cd/Pb Chalcogenide Heterostructured Janus Particles via Controllable Cation Exchange. ACS Nano.

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