Charge Transport via Redox Ligands in Quantum Dots

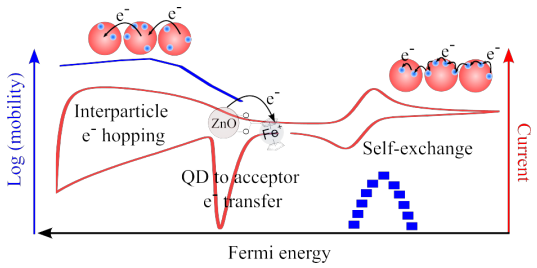
Y. B. Vogel[1], A. J. Houtepen[1]*

[1]Department of Chemical Engineering, Delft University of Technology

We present a strategy to actively engineer long-range charge transport in colloidal quantum dot (QD) assemblies by using ligand functionalities that introduce electronic states and provide a path for carrier transfer.[1] This is a paradigm shift away from the use of inactive spacers to modulate charge transport through the lowering of the tunneling barrier for interparticle carrier transfer. It has immediate implications in the design of QD lasers, light-emitting diodes, solar cells and photodetectors where charge transport determines the device performance.

The introduced states are narrow and only available at a certain Fermi energy, which makes it challenging to explore. To overcome this challenge, we turned to an electrochemical approach that allows to set the Fermi energy while simultaneously measure the rate of charge transport. We show that charge transport occurs through a succession of self-exchange reactions between the immobile redox ligands distributed at random positions in the assembly and their density is readily controllable. This allows to adjust the conductivity which is explained by a percolation model. Modulating the Fermi energy allows to switch between charge transport mechanisms, from self-exchange to electron hopping, and this is shown to be accompanied by ion transport.

Overall, the strategy presented here enables a means to actively control long-range charge transport in QD assemblies that remained until now unexplored.



[1] Yan B. Vogel, Maarten Stam, Jence T. Mulder, Arjan J. Houtepen, *ACS Nano*, **2022**, 16, 21216-21224.