Nanoscale photovoltaics: aminoethanethiol coated CdSe quantum dots

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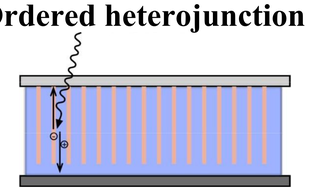
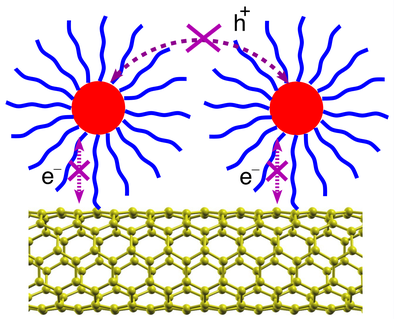
Background

Ordered heterojunction PVS



Good efficiency
Fast carrier transport
cost-effective PV cells

QD surface capping



Long capping agents form a barrier to charge transfer

QD/CNT

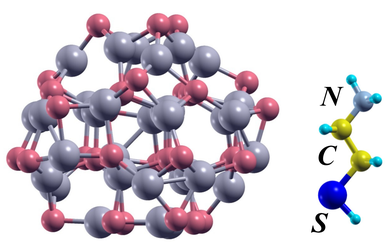
CdSe quantum dots (QDs) are decorated to long and aligned carbon nanotubes (CNTs)
QDs: tunable band gap, multiple exciton generation
CNTs: high carrier mobility

Present focus of activity

Short ligands, such as aminoethanethiol (AET) and pyridine, are current focus of activity

Motivation

CdSe QD and AET



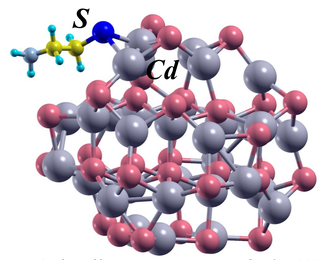
Cd, Se **$HS(CH_2)_2NH_2$**

QD, D=1.3 nm ***AET***

Using DFT, we provide insights into the atomistic details of the binding models of the AET molecule on the CdSe QD surface.

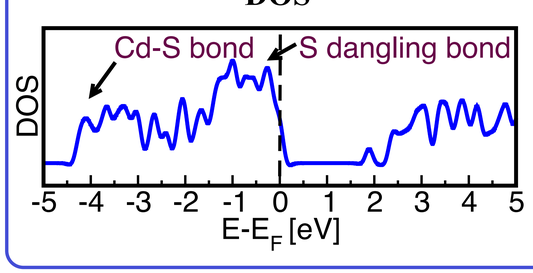
Sulfur-Cadmium Binding

Morphology

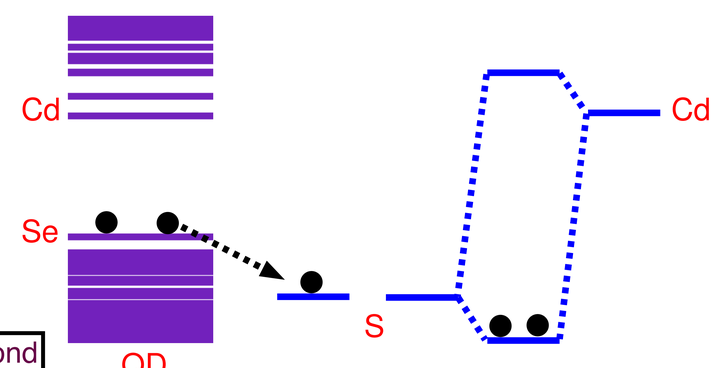


Binding energy ~2.1 eV

DOS



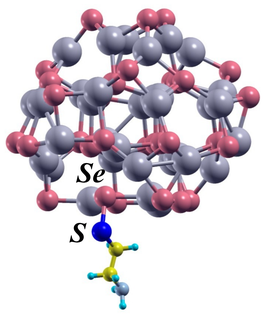
Interface molecular orbital diagram



When the S-Cd bond forms, one S π orbital and the Cd s orbital overlap to form a covalent bond. The Cd is electron poor, so S takes an electron from the Se-dominated HOMO of the QD to become closed shell.
<u>This bond hole dopes the QD</u>

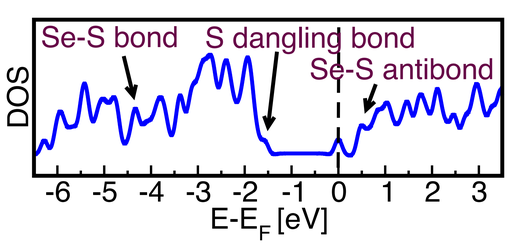
Sulfur-Selenium Binding

Morphology

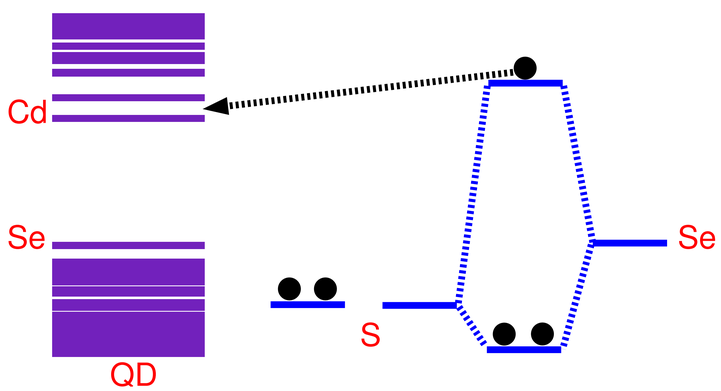


Binding energy ~1.1 eV

DOS



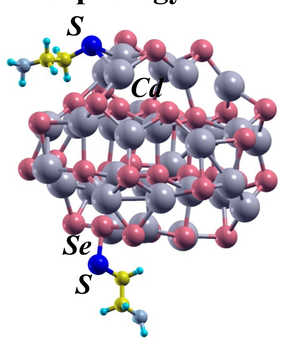
Interface molecular orbital diagram



When the S-Se bond forms, one S π orbital and one Se π orbital overlap to form a covalent bond The Se is electron rich, so the S becomes closed shell but an additional electron needs to be accommodated. Instead of occupying the high-energy anit-bonding state, the electron moves to the Cd-dominated LUMO of the QD.
<u>This bond electron dopes the QD</u>

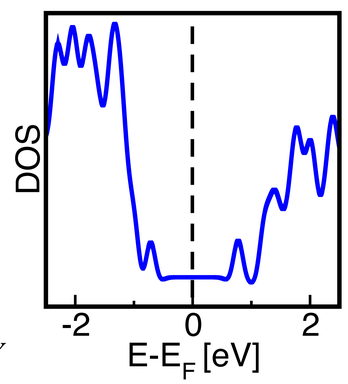
Two AET Ligands

Morphology



Binding energy ~3.7 eV

DOS



The lowest energy morphology involves one S-Cd and one S-Se bond: having one of each leads to self-compensation of the doping of QD by each ligand. Please note that when the QD is linked to a CNT via an AET molecule, it will become important to whether the AET is bound to Cd or Se as the different bindings have different band alignments which will modify the photovoltaic properties.

Conclusions

For AET coated CdSe QDs

The linking end of the AET contains a S atom, which acts as a hole donor to the QD when a Cd-S bond is formed or an electron donor when Se-S is formed.

While the S-Cd bond is stronger in isolation, the preferred binding morphology for two ligands involves both S-Cd and S-Se bonds. The preferred double binding mode is due to self-compensation, i.e., the doping electron and doped hole compensate which is always stabilizing.

when the QD is linked to a CNT via an AET molecule, it will become important to whether the AET is bound to Cd or Se.