A quantum-dot antenna layer coupled to a thin-film solar cell.

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The minimum thickness of the light-absorbing layer in a thin-film solar cell is decided by the absorption depth of the light. Inventive photonic structures have been proposed to trap the incoming light in the light absorbing layer. Another route to effective light harvesting is the use of a light-absorbing antenna layer coupled to the active electron-hole separating layer in a cell. Photons are absorbed in the antenna layer and travel as excitons towards the interface with the cell. The excitons are injected *non-radiatively* in the electron-hole separating layer of the cell. The loss of energy quanta in the antenna layer and at the interface of the antenna layer with the cell by *non-radiative* recombination should be avoided.

At present, we attempt to couple an antenna layer of CdSe or CdTe quantum dots to the thinfilm gold/TiO₂/Ti photovoltaic device proposed by McFarland and Tang [1]. The principle of photonto-current energy conversion in this device is schematised below.



There are 4 essential steps: (1) Photon absorption by the quantum dot solid (indicated in gray) and excitonic decay. (2) Exciton energy transfer from QD-dot to QD-dot (3) Ballistic electron transfer of hot electrons in the TiO₂/Ti device through a thin layer of gold (20 - 100 nm). (4) Hole scavenging by electrons from the gold. Due to the specific injection of high-energy photo-excited electrons in the gold/TiO₂/Ti layer (over the Schottky barrier), and photoexcited holes in the gold layer, a photovoltage is measured between the gold metal layer and the Ti. In this cel, light harvesting and electron-hole pair separation are spatially separated. In contrast to a conventional solar cell, the specific electron and hole membranes are NOT spatially separated. McFarland used a sub-monolayer of dye-molecules as light absorber, and reported a photovoltage 600 to 800 meV. The short-circuit photocurrent density was $10 - 20 \,\mu\text{A/cm}^2$.

We have prepared a gold/ TiO_2/Ti device and studied its opto-electric characteristics (see below). We also have prepared CdTe solutions and solids, and studied the exciton dynamics in this system (see below). At present we are trying to couple a CdSe quantum dot solid to a gold/ TiO_2/Ti device.

[1] E. W. McFarland and Jing Tang, Nature 421, 616 (2003).

Preparation and opto-electrical characteristics of the gold/ TiO2/Ti device.

Ti substrates (99.96 %) were polished until a mirror-like surface was obtained. A TiO₂ layer (50 – 100 nm) was grown by anodization in 0.5 M H₂SO₄ at a slowly increasing potential between 0 and 50 V. Alternatively, TiO₂ layers were thermally grown at 500 C. The resulting layers were light blue with mirror-like appearance. Gold layers 20 –30 nm in thickness were sputtered on the TiO₂. The resulting gold/ TiO₂/Ti interface showed rectifying I,V behaviour in the dark, a clear photocurrent and a photovoltage of 600 mV when the the device was illuminated with a Xe UV-VIS source through the thin gold layer.

Exciton energy transfer in a CdTe quantum dot solid.



Exciton energy transfer in a molecular system is well studied. An exciton can be transferred from a donor molecule to an acceptor molecule if there is spectral overlap, i.e. if the emission spectrum of the donor overlaps with the absorption spectrum of the acceptor. If there is a strong electronic coupling between the donor and acceptor orbitals, energy transfer can occur by super-exchange. If the electronic coupling is weak, but there is a strong electrostatic coupling between the optical transition dipoles of the donor and acceptor, Förster-energy transfer can occur. The probability of this type of energytransfer decays with 1/d⁶, d being the distance between the transition dipoles. In quantum dot assemblies and solids, exciton energy transfer does also occur. In the group of my colleague Meijerink, energy transfer in an assembly of CdTe quantum dots has been studied. Colloidal solutions of quantum dots show a high luminescence quantum yield (50%) and a mono-exponential photoluminescence decay with a decay time of about 20 ns at room temperature, typical for a purely radiative excitonic decay. The left figure shows the absorption and luminescence spectra of two solutions of CdTe quantum dots. One solution consists of CdTe quantum dots emitting in the green, the second solution consists of larger CdTe quantum dots which emit in the red. In both cases, the photoluminescence decay at room temperature is mono-exponential with an excitonic life time of about 20 ns. However, with a concentrated solution or with an assembly of such quantum dots the exciton luminescence yield decreases indicating non-radiative losses. This is explained by the fact that exciton energy transfer occurs towards a defect CdTe quantum dot in which the exciton is trapped. The photoluminescence of assemblies consisting of a mixture of red and green emitting quantum dots have been studied. If a small amount of red-emitting CdTe quantum dots is mixed with the green-emitting quantum dots, the observed photoluminescence is predominantly arising from the red-emitting dots. This indicates effective exciton energy transfer between the green quantum dots, and efficient irreversible trapping of the exciton in the larger red-emitting dots. Energy transfer from the green to the red quantum dots is nicely demonstrated in the right figure which shows accelerated decay of the green quantum dots and retarded decay of the luminescence of the red quantum dots.