Characterization of Optoelectronic Properties of Colloidal Quantum Dots in a Nanogap

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Abstract:

This work focused on a fundamental exploration of charge transport through a self-aligned nanogap which had been deposited with colloidal cadmium selenide (CdSe) quantum dots via solution phase processing. Characterization of charge transport through the gap, which was only a few nanometers wide, centered around measurements made at room and cryogenic temperatures, and the response of the device as a result of interaction with light.

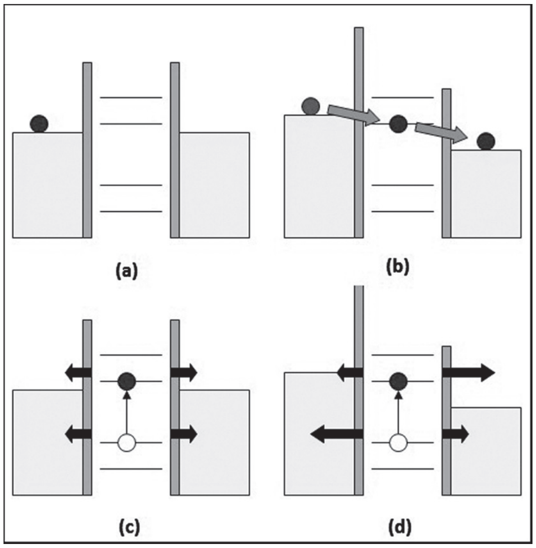

Figure 1: (a) No bias energy level structure of the device. (b) Applied bias is non zero. The source is now within tunneling distance of the unoccupied level in the island. (c) An exciton. Probabilities the charge carriers will tunnel in each direction are represented with the size of the arrows. (d) Light with applied bias. The probabilities of tunneling change and make it more likely excited electrons will contribute to the current.

Introduction:

Single electron transistors are made up of a quantum dot (or island in the following text) that sits between two metallic electrodes (source and drain). In the case under investigation, the island was weakly coupled to the two leads meaning that the level of the island did not mix with the bands of the leads and, therefore, they had a definite energy and degeneracy. This caused incoherent transport, the electron not retaining its phase as it moved between the different elements. The electrons would only be able to tunnel to the island when the bias was high enough that accessible island energy levels were within range (conduction). This was ultimately decided by the probabilities of the charge carriers to tunnel between either source or drain [1].

Charge transport was also affected by light interaction. When shining light on the devices, excitons were created and could separate, the electron and hole tunneling in either direction. But without the applied bias, there would be no net conduction through the devices as the probabilities that determined the direction of charge carrier tunneling would all be equal. However, under biased conditions, tunneling would shift in one direction and conduction would increase [1].

Methods:

Planar electrodes of different widths were fabricated using a self-alignment fabrication scheme [2]. The nanoparticles were deposited by dip-coating with ligand exchange. The sample was pretreated with short ligands and dipped into the solution where nanoparticles attached and long ligands exchanged with short ligands. This closer contact allowed for better coupling of the particles to the leads. The process was completed by dipping the chip into clean solvent to remove excess particles.

The devices were characterized empty and post deposition in a vacuum probe station. Post deposition the devices were measured in the dark and then exposed to a 545 nm laser. Analysis was done to find the best devices, which were tested at a single voltage with a chopper at varied frequencies.

In order to eradicate effects due to thermal broadening, the current-voltage responses of the devices were characterized at low temperature. The probe station was cooled down to about 10°K using liquid helium.

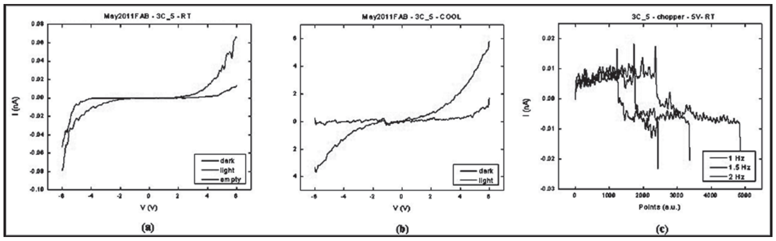

Figure 2: (a) Empty, dark and light IV curves from +/-6V at room temperature. Empty means before particle deposition while dark is after deposition without any light interaction. (b) Light and dark curves from +/-6V at 10K. (c) Chopper measurements at 5V room temperature.

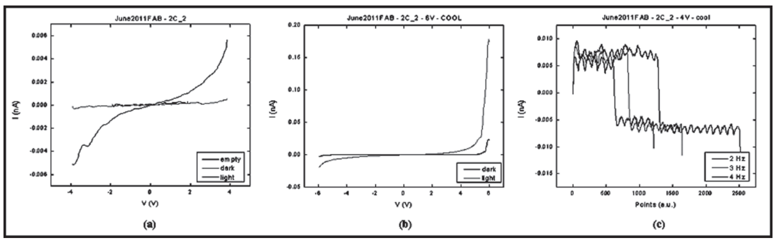

Figure 3: (a) Empty, dark and light IV curves from +/-4V at room temperature where empty is pre-deposition and dark is post-deposition without any light interaction. (b) Light and dark curves from +/-6V at 10K. (c) Chopper measurements of intensity at 5V.

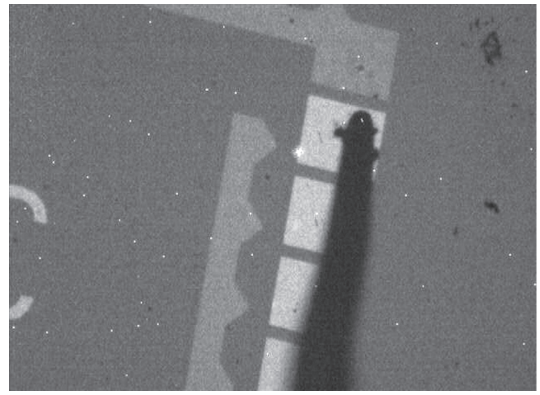

Figure 4: False-color picture of the luminescence of the device [in greyscale]. The circled area is the only light not due to noise of the camera. (See cover for full color version.)

Luminescence experiments were done using a CCD camera in a fully dark probe station. Six volts were applied to the device while the shutter of the camera was open to absorb any light emissions from the particles.

Results:

Figures 2 and 3 show the best results from the experiments with CdSe particles, showing a marked difference when the device interacted with light. The total average difference in the light response compared to the dark can be seen in the chopper graphs. For Device 1 at 5V, the average difference was about 0.015 nA and for Device 2, it was 0.014 nA at 4V. At higher voltages there was more of a response. The chopper measurements for Device 1 were taken at room temperature and Device 2 at 10°K.

The photon to electron efficiency was calculated at 4V using the definitions in equations 0.1 and 0.2 to determine the relation in 0.3. At 10°K the efficiency of devices one and two were 2.17E-8 and 1.97E-7, respectively.

$$(0.1)\ I = \frac{n_e e}{s}$$

$$(0.2)\ P = \frac{n_p h\nu}{s}$$

$$(0.3)\ \frac{n_e}{n_p} = \Delta I \frac{h\nu}{Pe}$$

The response of the devices was similar to the theory developed based on rate equations, but it is still under investigation and thus there are some discrepancies.

Luminescence experiments were performed on all working devices, but only one device generated light, seen in Figure 4. The false color red indicates areas where light was emitted. Unfortunately, this device broke before the required further tests; this also suggests more deeply investigation into the device stability at high biases.

Conclusions:

The addition of CdSe particles in this manner results in increased conductance of the devices tested. When the interaction with light is included, the conductance is increased even more, due to the intrinsic nature of the particles. More work should still be done regarding the particles to have a fuller picture of the level structure, varying power, wavelength polarization and gate voltage.

Acknowledgements:

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References:

[1] F. Prins, M. Monrabal-Capilla, E. O. Osorio, E. Coronado and H. S. J. van der Zant, Adv. Mater., 2011, 23, 1545.

[2] Thijssen, J. M.; van der Zant, H. S. J. phys. stat. sol. (b) 2008.