Oxidation Effect in Single Dot Quantum Junctions

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Abstract:

Three-terminal devices with a monolayer of quantum dots (QD) between nanometer-spaced gold electrodes allows for the study of single electron quantum transport. Indeed, IV measurements for a 50 nm device show characteristic Coulomb staircase under dark condition and with 543 nm and 670 nm light. However, lead selenide (PbSe) devices of various sizes exhibited current fluctuations on the order of nanoAmperes (nA) over time, both under dark condition and with light. Also, a slow relaxation after shining light was observed. With increasing oxidation, the devices stabilized and the relaxation time decreased, which suggests the surface state of the QD governs transport through the device.

Device Fabrication:

The devices had high aspect ratios: while the width of the devices varied from 10 μm to 20 nm, the gap between the electrodes was kept at about 6 nm to fit the QD monolayer. In order to achieve this, a layer of chromium (Cr) was evaporated on top of titanium (Ti) adhesion and first gold (Au) electrode layers. The growth of Cr_xO_y led to an overhang over the edges of the first layer. The overhang served as a mask for the second Au electrode layer, producing 6 nm gaps when the Cr and oxide were etched away [1]. A monolayer of PbSe colloidal QD was deposited via the dipcoating method [2]. Ligand substitution, which increases coupling to the electrodes, was performed with 1,2-ethanedithiol (EDT).

Theory:

Device behavior was modeled on single electron resonant transport, which assumed weak coupling (though coupling may be much stronger because EDT is short). The QD can be considered to have discrete quantum levels (Figure 1).
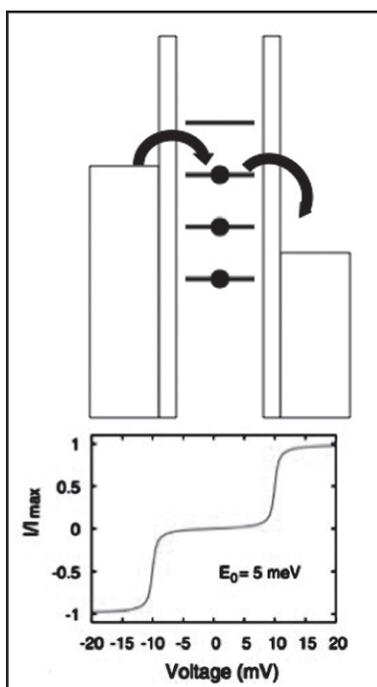
Figure 1: Schematic of chemical potentials of QD and electrodes; Coulomb staircase.

Tunnel barriers separated the QD from the electrodes on each side, wherein the electrons obeyed the Fermi-Dirac distribution. To observe quantum behavior, low temperature was required to ensure that the energy difference between quantum levels in the QD (~ 1 meV) and the charging energy of the device ($e^2/(2C)$, where C is the capacitance of the device, ~ 25 meV) were greater than the thermal energy of the charge carriers.

In these devices, current was driven by the chemical potential difference between the charged states in the QD and the electrodes. For resonant transport, the chemical potential of the QD charged transition state must lie within the bias window, which is the energy range between the chemical potentials of the electrodes. Electrons with energy resonant with the QD level tunneled from occupied states in the source to the QD, then ended up in empty states in the drain. A sharp increase in current was observed as a step when the bias window enclosed a resonance.

At low bias, current was suppressed as there were no transitions that were within the bias window. For the QD to undergo a charge transition (by adding or removing an electron to or from the QD), a charging energy penalty was required. This region of high differential resistance is known as a Coulomb blockade and is observed as a flat region in the IV curves.

Photoconductance depended on the formation of excitons and extraction of charge carriers to the electrodes. Formation of excitons depended on the absorption rate, which depended on laser power. Photoconductance varied monotonically with the laser power density; therefore, increased current should be observed with increased laser power.
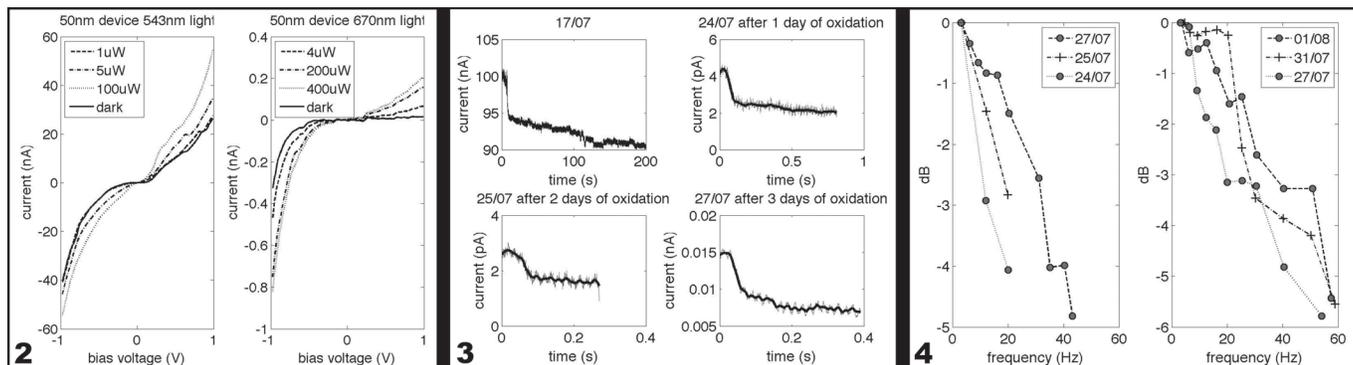
Figure 2, left: Coulomb staircase in 50 nm device at 10K.

Figure 3, middle: Continuous-wave measurement of current decay at room temperature.

Figure 4, right: Time-resolved measurement with modulated light intensity at room temperature.

Results:

IV measurements were performed from -1V to 1V in vacuum and at cryogenic temperatures. Figure 2 shows a 50 nm device that exhibited step behavior characteristic of single or few QDs. Photoconductance was observed for both 543 nm and 670 nm. The Coulomb blockade region became smaller with increased laser power as well. For the larger devices with more QDs, the IV curves were smoother, indicating that the presence of many QDs smoothed out the staircase behavior of a single QD. However, the reference dark IVs changed by a few nA over days, with no apparent trend for the change in Coulomb blockade region size or magnitude of current. Measurements of current over hours at 0.5V showed fluctuations of order nA as well.

The devices were exposed to air in room temperature one day at a time. After each day of oxidation, continuous-wave and time-resolved measurements were performed. For the continuous-wave measurement, after shining 670 nm 100 μW light, the devices were then covered and the decay of the current was observed over time. For the other measurements, the intensity of the light was modulated at increasing frequencies in order to observe when the device stopped exhibiting photoresponse. The value of 3dB was a metric for the cutoff of photoresponse.

Figure 3 clearly shows the relaxation time decreased with increased oxidation, especially dramatically after one day. Figure 4 shows that with the frequency for the cutoff of photoresponse increased with increased oxidation. These both indicate that the device showed clearer photoresponse with increased oxidation.

Future Work:

Oxidation reduces the time dependence of current and the relaxation time in these three-terminal devices. However, the measurements performed were only preliminary measurements and oxidation was not performed in a completely controlled fashion. Future work includes oxidation of different systems in a more controlled manner, with variation of QD type (CdSe, for example) and device width.

Acknowledgments:

References:

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[3] Thijssen, J.M., et al., Charge transport and single-electron effects in nanoscale systems. Phys. Stat. Sol. (b). 2008, 245, 1455-1470.