Negative two-step two-photon-absorption photocurrent induced by infrared photons in quantum dots: dot density dependence

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Novel solar cell concepts are expected to play an important role in the future high-efficiency solar-cell market. The quantum dot (QD) intermediate band (IB) solar cell extends the classical conversion efficiency limit by additional photocurrent generation in QDs, which are embedded in a bulk junction.[1–3] Additional photocurrent contribution from QDs is achieved by step-wise excitation of the electron from the QD ground state to its excited state, and then upconversion to the bulk conduction band by absorption of infrared (IR) photons; an upconversion process via so-called two-step two-photon-absorption (TS-TPA).

In general the additional IR illumination is expected to enhance photocurrent. However, recently, multiple-beam excitation experiments at low temperatures have shown that these IR photons can also lead to negative photocurrent response.[4,5] This complex behavior indicates that the role of the QD layer in the IB as upconversion site as well as recombination site has to be reconsidered. Analysis of the new carrier distribution upon IR excitation, which leads to enhanced recombination, is important to design layers unaffected by photocurrent quenching through IR. The Dominant trapping and recombination in different states in the InAs layer have been proposed as one of the possible reasons for quenching.[4,6] A suited InAs layer morphology should therefore provide one way to improve the QD IB solar cell. Measurements on InAs layers with different morphologies can be used to infer about the origin of the photocurrent quenching.

In this work we analyze three undoped InAs/GaAs samples grown by molecular beam epitaxy. The growth conditions were chosen to obtain three samples with high, intermediate and low QD densities. Since QD formation is preceded by formation of disk structures, even the high density QD samples contained some disk structures, as confirmed by low-temperature photoluminescence excitation spectroscopy. Two-beam photocurrent measurements with a wide-range single tunable beam and an infrared laser were used to probe the IR responses at room temperature.

The results for excitation of the QD showed that the sample with high QD density exhibits negative photocurrent response on additional IR illumination. Intermediate and low QD-density samples show a zero and positive photocurrent increase, respectively. Interestingly, for excitation of the disk states, no photocurrent reduction has been observed in any of the samples. The results indicate the importance of the InAs layer morphology and provides further details about the origin of the negative IR response. We conclude that higher QD densities can lead to higher photocurrent reduction through the IR illumination. For further improvement of the TS-TPA efficiency, it should be considered that other InAs layer morphologies, such as those found in the low-density QD sample, may be more beneficial for IB solar cells.

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