Correction to Enhanced Multiple Exciton Generation in Quasi-One-Dimensional Semiconductors

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7 AUTHORS, INCLUDING:

Paul D Cunningham
United States Naval Research Laboratory

Janice E. Boercker
United States Naval Research Laboratory

Joseph G. Tischler
United States Naval Research Laboratory

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Correction to Enhanced Multiple Exciton Generation in Quasi-One-Dimensional Semiconductors

Paul D. Cunningham,* Janice E. Boercker, Edward E. Foos, Matthew P. Lumb, Anthony R. Smith, Joseph G. Tischler, and Joseph S. Melinger

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In our initial report of multiple exciton generation (MEG) in PbSe nanorods,[1] we reported enhanced MEG in PbSe nanorods compared to PbSe nanocrystals as characterized by an increase in MEG efficiency to $\eta = 0.81$ from 0.41, and a reduction in threshold energy for the MEG process to $(2.23 \pm 0.03)\ E_g$ from $3.4\ E_g$

We recently discovered that a malfunctioning lock-in amplifier (LIA) affected our measurements by limiting the signal-to-noise ratio. This limited the lowest fluences that could be reached and introduced significant noise into the exciton dynamics data. As a result, the exciton quantum yields were overestimated and determined with large uncertainty. Nevertheless, our main finding, that MEG in PbSe nanorods is more efficient than in PbSe nanocrystals, remains qualitatively correct.

In Figure 1, we compare two sets of data obtained from fresh PbSe nanorods using the malfunctioning LIA and a correctly functioning LIA. The data taken with the malfunctioning LIA agree within experimental error with the measurements in our initial report. However, when a correctly functioning LIA is used, the exciton quantum yields are measured to be lower than we reported. These new results agree with those recently reported by Sandberg et al.[2] To quantify the overestimation we use an empirical model from Beard et al.[3] This model is characterized by the threshold parameter P, which describes the competition between the MEG and carrier cooling rates. Results obtained with the malfunctioning LIA can be fitted with $P = 4.8 \pm 0.4$, which yields a biexciton generation threshold energy of $h\nu_{th} \sim 2.21$. Results obtained with a correctly functioning LIA yield $P = 3.0 \pm 0.1$ and $h\nu_{th} \sim 2.33$. For reference, best fits to MEG measurements of PbSe nanocrystals yield $P = 1.5$ and $h\nu_{th} \sim 2.7$.[3] Therefore, our finding of a reduced MEG threshold in nanorods still holds.

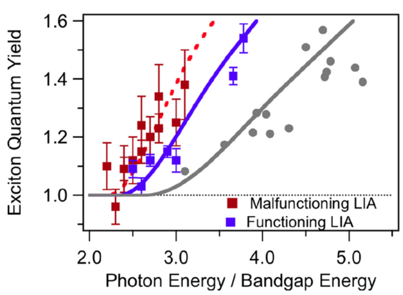

Figure 1. A comparison of exciton quantum yield as a function of photon energy for the case of a correctly functioning and malfunctioning LIA. The solid and dashed lines are fits to the model in Beard et al.[3] The gray data are literature measurements of PbSe nanocrystals.[3]

Our overestimated MEG efficiency also affected our predictions for the power conversion efficiencies possible in photovoltaic cells. Using the corrected values, we now find that for a band gap energy larger than 0.4 eV the maximum obtainable PCE is only slightly above the Shockley–Queisser limit, Figure 2.

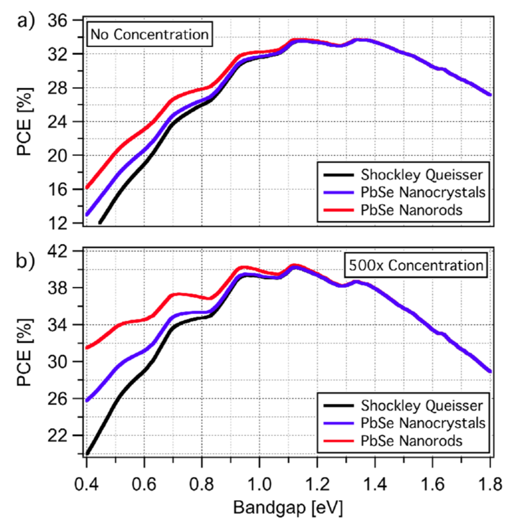

Figure 2. The power conversion efficiency versus band gap energy under (a) AM1.5 and (b) 500× solar concentration for PbSe nanocrystals (blue) nanorods (red). The Shockley–Queisser limit (black) is included for reference.

In summary, the qualitative findings of our initial report remain valid. MEG in PbSe nanorods shows a reduced threshold energy and an increased efficiency compared to PbSe nanocrystals. However, the magnitude of this enhancement was overestimated.

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