CARRIER INTERACTIONS IN QUANTUM DOTS

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Introduction

Quantum confinement can increase carrier interactions. If this increases carrier multiplication yields, it has a potential application in more efficient photovoltaics based on carrier multiplication. Recent experiments on the high energy electronic dynamics of quantum dots led us to suggest that electronic excitation above the carrier multiplication threshold may produce independent carriers rather than quantum confined excitons.[1] We have carried out femtosecond experiments that probe interactions between hot carriers in lead sulfide quantum dots by systematically varying both the excitation probability and the pump-probe delay.

Experiment

The high energy absorption spectrum of PbS quantum dots contains features that exhibit quantum confinement, such as the E1 transition, which occurs at about 3x the bandgap and blue-shifts with decreasing nanocrystal size. However, the quantum confined absorption features at high energy ride on top of a background absorption spectrum that does not show confinement effects – the molar extinction coefficient of the background is proportional to nanocrystal volume. Most of the high energy absorption is attributable to this background. So far, experiments have been conducted at two photon energies, one probing a mixture of confined states and the background, and one probing the background alone.

Results and Discussion

Pumping and probing at the quantum confined E1 transition produces signals that immediately (within the 25 fs time resolution) depart from proportionality to the number of absorbed photons. This departure is readily measurable below an average of 0.3 absorbed photons per dot and attributable to the fraction of excited dots that have absorbed 2 photons. This is consistent with the immediate interaction expected between delocalized quantum dot excitons.

In contrast to experiments on quantum confined features, pumping and probing at a lower photon energy, where the molar extinction coefficient is proportional to nanocrystal volume generated signals that start out proportional to the absorbed photon number even up to an average of 7 absorbed photons per dot. These signals develop departures from proportionality, indicative of carrier interactions, on a timescale of 130 fs, which is faster than Auger recombination (120 ps) or cooling to the band edge (600 fs). The initial lack of interaction would be expected for free carriers in a bulk semiconductor.

Conclusion

The nature of the rapidly developing interaction between hot carriers excited via the background absorption is not entirely clear. An extension of the usual analysis for Auger recombination kinetics suggests the fastest interaction may be a hot Auger recombination process that is the inverse of multiple exciton generation with an excess energy threshold. Implications for carrier multiplication in photovoltaic applications will be discussed.

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Reference

(1) Cho, B.; Peters, W. K.; Hill, R. J.; Courtney, T. L.; Jonas, D. M. *Nano Letters* **2010**, 10 (*7*), 2498.

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