Temperature and Magnetic Field-dependent Photoluminescence Studies of Nanoscale Materials

T.D. Green, D.E. Blumling, C.A. Lowman, C. Yi, K.L. Knappenberger, Jr. (FSU, Chemistry and Biochemistry);
T. Tokumoto, S. McGill (NHMFL)

Introduction

The optical properties of spatially confined matter are of interest due to their potential applications in a variety of fields, especially solar-to-electric energy conversion. Quantum-confined semiconductors are characterized by large absorption cross sections and have band gaps that are tunable across much of the solar spectrum. In this way, devices based on the semiconductor platform can be constructed with an optical, and therefore energetic, gradient that can be tailored to optimize vectorial energy flow. Currently, these materials are limited by intrinsic non-radiative energy loss processes that degrade performance. Our group, through collaboration with McGill, has used magneto-optical data to quantify energetic barriers that mediate directed energy migration in quantum-confined Cadmium Selenide semiconductor nanocrystals. We have also investigated ways to use nanostructure morphology as a parameter for optimizing energy transfer efficiency.

Experimental

Experiments were performed at the NHMFL DC Field facility utilizing a 17.5 T superconducting magnet (SCM-3) with temperature control from 4.5 K to ambient temperature was used for this study. The 800 nm output of a regeneratively amplified Ti:Sapphire femtosecond laser system was frequency doubled to 400 nm and attenuated to 1 µJ/pulse to provide an excitation source. Photoluminescence was collected with an optical fiber and sent to a spectrometer. The signal was collected with a liquid N_2 cooled CCD for spectrally resolved data or an avalanche photodiode for time-correlated single photon counting measurements.

Results and Discussion

Our recent research collaboration with the McGill group has resulted in two publications,[1,2] while several other manuscripts are in preparation. Our published work details the quantification of the relative populations of "bright" and "dark" nanocrystals excited states, as well as a determination of the influence of each on system optical properties.[1] We have built on this work to demonstrate that applied magnetic fields can be used to control directional transfer of electronic energy through assemblies of nanocrystals.[2] The ability to control energy flow results from the ability to use magnetic field to alter the relative excited state populations (**Figure 1**). In continuing work, we are exploring the influence of magnetic fields on different nanocrystals morphologies, where field-nanocrystal orientation, and varying degrees of spatial confinement can be exploited to control nanocrystals optical properties. We have already discovered a size-dependent cross over in the energy configuration of "bright" and "dark" states in one-dimensional semiconducting nanorods. A manuscript detailing these results is currently under preparation.

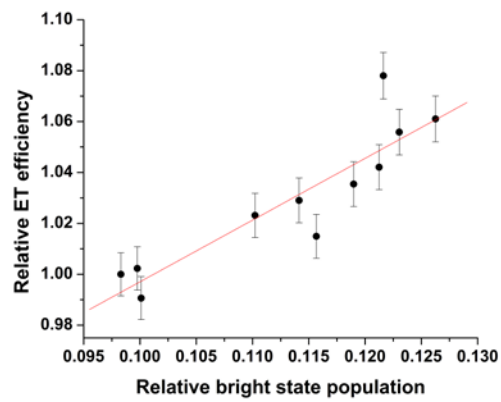

Figure 1 (from ref 2)**.** Calculated energy transfer efficiency plotted versus the relative population of the ''bright'' state of the energy donating nanocrystals within the CdSe aggregate. The red line demonstrates the linear relationship between the two parameters.

Acknowledgements

KLK gratefully acknowledges the Air Force Office of Scientific Research for financial support; grant number FA9550-10-1-0300. Work carried out at the NHMFL was supported by National Science Foundation Cooperative Agreement DMR-0084173, with the State of Florida.

References

[1] Blumling, D.B., *et al*., J. Phys. Chem. C, **115**, 14517-14525 (2011).
[2] Blumling, D.B., *et al*., Physical Chemistry Chemical Physics, **14**, 11053-11059 (2012).