Time-Resolved Magneto-Photoluminescence Measurements to Determine the Role of Dark Exciton States in Semiconductor Nanocrystals

Richard D. Schaller (C-PCS, LANL); Scott A. Crooker (NHMFL-LANL); Jeffrey M. Pietryga; Victor I. Klimov (C-PCS, LANL)

Introduction

Chemically synthesized nanocrystals (NCs) exhibit broadly size-controlled tunability of the emission color and high photoluminescence (PL) quantum yields. These characteristics make NCs attractive materials for light-emitting applications ranging from bio-labeling and solid-state lighting to optical amplification and lasing. PbSe NCs can be size tuned through the near- to mid-infrared and are highly efficient emitters with reported PL quantum yields as high as 80%. [1] However, this material also exhibits an unexpectedly long excited-state lifetime at room temperature (of the order of 0.8 to 2 μs), whereas other materials such as CdSe NCs have much shorter lifetimes of ~0.02 μs.

The origin of the long excited state lifetime in PbSe NCs is not well-understood. A low energy, optically passive, dark exciton state may exist that causes PL to only take place upon thermal activation to an optically active (bright) exciton state. However, other explanations of the lifetime exist such as the role of dielectric screening. [1,2] Dielectric screening may dramatically reduce the rate of dipolar recombination in PbSe NCs as the bulk material has a very large optical dielectric constant of 23 whereas that of CdSe is 6. Some preliminary measurements by Clark et al. on PbS NCs bolster this possibility, but do not quantitatively agree and only explore a small range of dielectric environments. [3] If PbSe does emit a low-lying triplet state, application of large magnetic fields should cause an increase of the radiative rate.

PbSe NC samples, synthesized according to the method reported by Murray, [4] were diluted in an inert host material so as to minimize the effects of energy transfer between NCs and placed in the helium cryostat sample chamber of a 15T staff lab superconducting magnet. A fiber optic was used to both illuminate the sample and to detect PL signals. Excitation was provided by a short pulse laser diode operating at 635 nm and PL photons were directed to a spectrograph and InGaAs array or a fast InGaAs photomultiplier.

Although this work is still ongoing, we have been able to detect PL photons with sufficient efficiency to make measurements of reasonably high signal-to-noise. Thus far, we have measured the excited-state lifetime dependence on temperature and observe that a real increase in lifetime does occur at low temperature. Such an increase in lifetime is indicative of a contribution from a low-lying dark exciton state, but additional studies with applied magnetic field need to be performed.

This work was supported by Los Alamos LDRD funds.

References