PROBING EXCITONIC SPIN STATES IN NANOCRYSTAL QUANTUM DOTS BY RESONANT SPIN-POLARIZED PHOTOLUMINESCENCE SPECTROSCOPY

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Introduction

The excitonic energy spectrum in colloidal CdSe nanocrystal quantum dots is strongly influenced by quantum confinement as well as the electron-hole exchange interaction and wurtzite symmetry, which lifts the eight-fold degeneracy of the exciton ground state. The result is a five-state exciton structure, with a ground state characterized by a net spin projection J = 2 on the **c**-axis of the wurtzite structure, which is therefore dipole forbidden ("dark"). The study reported here focuses on probing this exciton structure in CdSe wurtzite nanocrystal quantum dots by high-resolution spin-polarized photoluminescence (PL) and Raman spectroscopy in high magnetic fields.

Experimental

The magneto-PL experiments were performed in magnetic fields to 18 T at the NHMFL facility in Los Alamos, and 33 T at the NHMFL DC field facility in Tallahassee, Florida. The measurements were carried out in the Faraday geometry (**B** parallel to **k**) for temperatures as low as 1.7 K, with the sample mounted on a fiber-coupled probe especially designed to resolve weak features that are spectrally very close to the excitation, by minimizing the inadvertent collection of scattered excitation light. The PL was resonantly excited with a narrowband tunable dye laser and, for polarization-resolved studies, thin film circular polarizers were attached onto the fiber mounts.

Results and Discussion

Typical measured resonant PL spectra vs. magnetic field are shown in Fig. 1(a). In this case, spin-up excitons were resonantly pumped into the lowest optically active J = 1 state, and emission from spin-down J = 2 excitons was detected. In addition to the expected broad features associated with "dark" excitonic recombination, the spectra measured at magnetic fields larger than 10 T exhibit a sharp peak, which moves approximately linearly with applied magnetic field, similar to a spin-flip transition. Further, the energy of this high-field peak varies systematically as a function of nanocrystal radius, as illustrated in Fig 1(b). Unlike typical electron spin-flip transitions in quantum dots, the mode energy extrapolates to a finite value at zero magnetic fields.

This observation offers an unexpected insight into the effects of anisotropy on the exciton energy levels in small nanocrystals. If the symmetry of the dot is lowered, either due to the presence of a uniaxial strain or the elongation of the nanocrystal along an axis other then the c-axis, then the degeneracy of the "bright" exciton doublet is lifted even in the absence of a magnetic field and the two resulting states are linearly polarized in orthogonal directions. The mode energy is thus associated with the splitting of the "bright" exciton state and the sharp peak is either a Raman spin-flip transition of the J = 1 exciton or PL associated with the recombination of J =1 spin down excitons. Very recent resonant time-resolved PL measurements performed on the same samples indicate that the PL decay time associated with the sharp peak is at least ten times shorter than the "dark" exciton lifetime, confirming the association of this transition with optically allowed excitons.

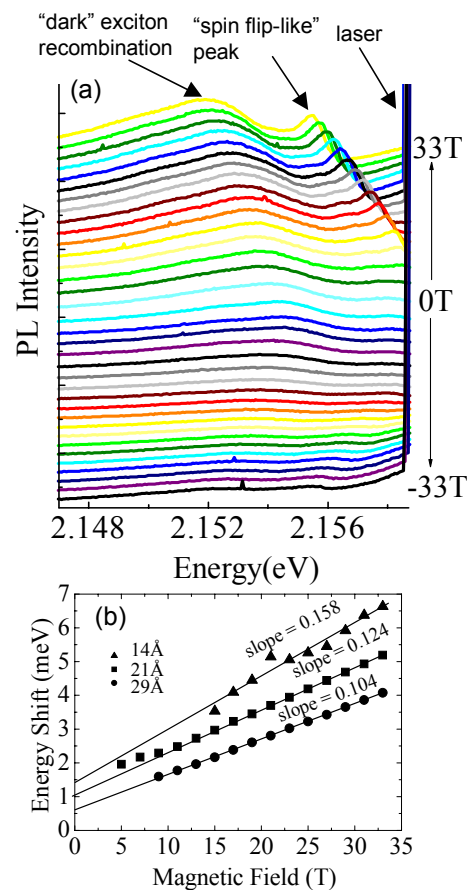

Figure 1. (a) High resolution resonant PL spectra from CdSe nanocrystals as a function of magnetic field. A "spin flip like" feature associated with the splitting of the "bright" exciton state is present for B> 10T. (b) Peak energy corresponding to the "spin flip-like" feature as a function of magnetic field for three different dot radii.