

Revealing the Exciton Fine Structure of PbSe Nanocrystal Quantum Dots Using Optical Spectroscopy in High Magnetic Fields

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- Lead-salt semiconductor nanocrystals (NCs) based on PbS, PbSe, and PbTe are actively studied due to their potential impact on important technologies including telecom-wavelength optoelectronics and enhanced solar energy conversion. However, in contrast to their counterparts that emit at visible wavelengths (e.g., CdSe NCs), the fundamental electronic structure of lead-salt NCs remains an open question, despite more than a decade of experiments and often-conflicting theoretical consideration.
- We measure the photoluminescence lifetime, τ , of excitons in colloidal PbSe NCs at low temperatures to 270 mK and in high magnetic fields to 15 T. For all NCs, τ increases sharply below 10 K but saturates by 500 mK (see Figure). In contrast to the usual picture of well-separated “bright” and “dark” exciton states (found, e.g., in CdSe NCs), these dynamics fit remarkably well to a system having two exciton states with comparable, but small, oscillator strengths that are separated by only 0.3-0.9 meV depending on NC size. Importantly, magnetic fields reduce τ below 10 K, consistent with field-induced mixing between the two states.

