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Mapping Exciton Spin States in CdSe Nanocrystals with Spin-Polarized, Resonant Photoluminescence Spectroscopy MADALINA FURIS, PATRICK ROBBINS, TODD BARRICK, SCOTT CROOKER, National High Magnetic Field Laboratory-Los Alamos National Laboratory, New Mexico, USA, SER-GUEI GOUPALOV, MELISSA PETRUSKA, VICTOR KLIMOV, Los Alamos National Laboratory, New Mexico, USA, ALEXANDER EFROS, Naval Research Laboratory, Washington DC, USA — We report on high-resolution, spin-polarized resonant photoluminescence (PL) studies of exciton spin states in CdSe nanocrystals in high magnetic fields up to 33 T. Optically allowed spin-up or spin-down "bright" (spin-1) excitons are resonantly pumped using a circularly-polarized, narrowband, tunable dye laser. Quasi-resonant PL from the optically forbidden "dark" (spin-2) excitons is analyzed as a function of polarization and magnetic field, allowing us to map the evolution of both "dark" and "bright" excitons with magnetic field. Strikingly, the PL develops a sharp, circularly- polarized "spin flip-like" peak for B >10 T, which we associate with the splitting of the "bright" exciton state. This energy splitting scales inversely with nanocrystal size and extrapolates to a finite value at zero magnetic fields. This large, zero-field splitting of the "bright" exciton (1-2 meV) likely originates from the anisotropy of the nanocrystal confining potential.

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