Bright-exciton fine-structure splittings in single perovskite nanocrystals

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Abstract

The electron-hole exchange interaction (e-h EI) is greatly enhanced in quantum-confined semiconductor nanostructures, leading to the energy-level splitting between bright- and dark-exciton states. In semiconductor epitaxial quantum dots (QDs), the dark excitons are generally nonemissive without applying a magnetic field and the bright-exciton state is further divided into two orthogonally- and linearly-polarized ones. A complete elimination of this bright-exciton FSS has been actively pursued to realize a polarization-entangled photon-pair source for fundamental tests in quantum mechanics and optics, as well as for practical applications in quantum communication. Interestingly, the dark excitons in colloidal semiconductor nanocrystals (NCs) are normally emissive, while the bright-exciton FSS was rarely observed.

Semiconductor colloidal perovskite NCs have just emerged as a novel type of semiconductor nanostructure capable of emitting single photons without the influence of dark-exciton emission [1]. Moreover, the suppressions of both the photoluminescence (PL) blinking and spectral diffusion effects were successfully demonstrated in single perovskite CsPbI3 NCs [2]. Here we show that the bright-exciton FSS can be easily observed in single CsPbI3 NCs at the cryogenic temperature, with an energy separation as large as hundreds of μeV between the two orthogonally- and linearly-polarized states [3]. With the laser excitation at an intermediate power, this PL doublet of neutral single exciton would switch to a single peak of singly-charged single exciton. When the laser power is further increased, PL doublets from neutral biexciton, charged biexciton and doubly-charged single exciton could be additionally observed. Based on the FSS values obtained from various exciton species, the isotropic and anisotropic e-h EI energies can be roughly estimated, which have provided valuable information on the fundamental electronic processes in these novel perovskite NCs.

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