Exciton fine structure, reduced blinking and fast single photon emission of single cesium lead halide perovskite nanocrystals

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In recent years, single quantum emitters have attracted a lot of attention due to their wide applicability ranging from nanoscopic probes to deterministic on demand single photon sources for quantum communication. Fully inorganic cesium lead halide ($CsPbX_3$, where X = I,Br,Cl) perovskite nanocrystals exhibit narrow emission lines, ultrahigh photoluminescence quantum yields of up to 90% and are tunable over a wide energy range[1]. Moreover, perovskite quantum dots are interesting due to their facile solution processability and their potential for high-efficiency photovoltaics and light sources[2]. However, the origin of their exceptional photophysical properties as quantum light sources and their optical characteristics at the single quantum dot level are still need to be uncovered.
Here, we show that single cesium lead halide nanocrystals exhibit stable, blinking-free emission at cryogenic temperatures[3]. Furthermore, we report the absence of Auger recombination without any shell passivation.

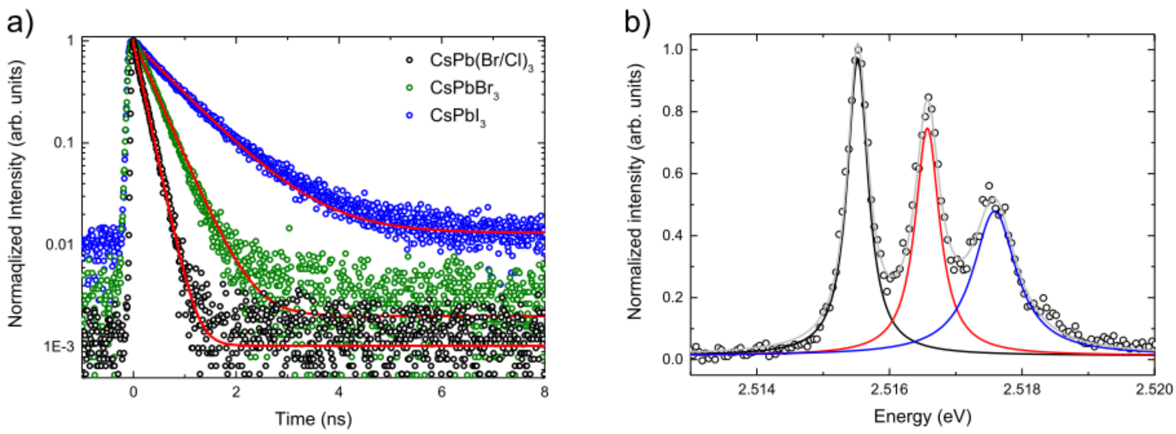

Fig. 1 : a) Time resolved photoluminescence from single cesium lead halide perovskite nanocrystals. b) High resolution PL spectrum of single $CsPb(Br/Cl)_3$ quantum dot, revealing three distinct fine structure peaks.

We investigate the origin of the composition dependent ultrafast recombination times, which can be seen in figure 1a), displaying a single exponential decay over 2 to 3 decades. For $CsPb(Br/Cl)_3$ nanocrystals the radiative lifetime is on the order of 200-250 ps, demonstrating a significant enhancement compared to other colloidal II-VI or epitaxially grown III-V quantum dots and prerequisite for efficient light sources.
Furthermore, by means of polarization dependent high resolution spectroscopy, the complex nature of the exciton fine structure splitting (Figure 1b)) and charged exciton emission has been investigated.
Their high oscillator strength, high quantum yield and broad composition and size tunability makes cesium lead halide perovskite nanocrystals superior for use as quantum light sources and pave the way towards integration into quantum communication systems.

References:

1. Protesescu, L. *et al.* Nanocrystals of Cesium Lead Halide Perovskites ($CsPbX_3$, X = Cl, Br, and I): Novel Optoelectronic Materials Showing Bright Emission with Wide Color Gamut. *Nano Lett.* **15,** 3692–3696 (2015).
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3. Raino, G. *et al.* Single Cesium Lead Halide Perovskite Nanocrystals at Low Temperature: Fast Single-Photon Emission, Reduced Blinking and Exciton Fine Structure. *ACS Nano* **10,** 2485 (2016).