Probing the Exciton Lifecycle of Single Caesium Lead Halide Perovskite Nanocrystals in Solution

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Lead-halide-based perovskites have sparked wide interest as materials for optoelectronic applications due to their high absorption cross section, high defect tolerance, excellent emission quantum yield, and low-temperature processability. These extraordinary properties have recently been translated into colloidal nanocrystals of caesium lead halide perovskites ($CsPbX_3$ X=Cl,Br,I) (P-NCs). P-NCs exhibit high emission quantum yields even in the absence of surface passivating shells, can be obtained via remarkably robust syntheses, and exhibit band-gap tunability via both composition and size tuning.[1] The thorough study of the optical physics of P-NCs is important not only for their potential real-world applicability, but also for our understanding of the effect of confinement on excitons in lead-halide-based perovskites.

Single nanocrystal emission spectroscopy promises this insight into the fundamental photo-physics of P-NCs in the absence of ensemble averaging. However, these studies have proven difficult for P-NCs due to their poor photo-stability under confocal excitation. Thus, only a few studies have reported on blinking [3], single particle spectra [3,4,2], and biexciton quantum yields (which vary widely from 6% [3,4] to 30% [2]). Moreover, all of these studies have been conducted on P-NCs of sizes comparable or larger than the exciton Bohr radius. As a result, our understanding of biexciton dynamics and emission lineshapes of single P-NCs in the confined regime is still very limited.

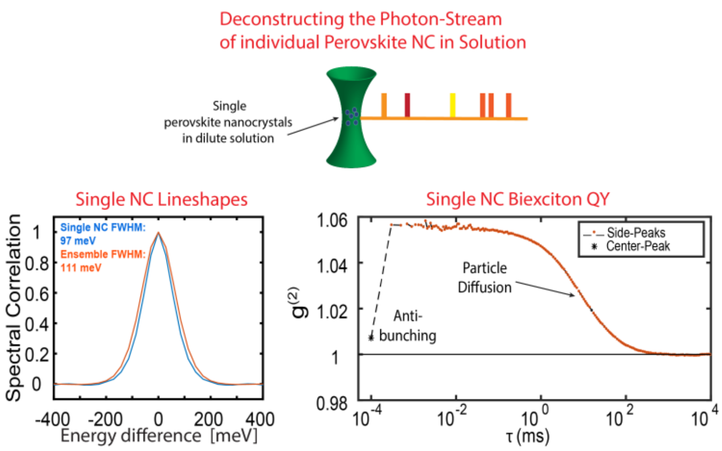

Fig. 1 We undertook a comprehensive survey of the single $CsPbX_3$ (X=Cl,Br,I) NC lineshapes and biexciton dynamics using advanced single photon-correlation based spectroscopic techniques.

To overcome the stringent sample stability requirements of conventional single NC spectroscopy, we have developed advanced solution-based photon-correlation spectroscopic techniques. With solution photon-correlation-Fourier spectroscopy (s-PCFS), we can extract the room-temperature ensemble averaged single NC linewidth from a dilute solution of NCs.[5] Anti-bunching measurements in solution (solution-$g^{(2)}$) allow for the quantification of the biexciton quantum yield.[6] The absence of user selection bias and particle photo-bleaching in our techniques allows for the batch-to-batch comparison of the single P-NCs properties and the correlation of these properties with synthetic parameters.

Using s-PCFS and solution-$g^{(2)}$, we have undertaken a broad survey of the single P-NC linewidth and biexciton quantum yield as a function of confinement degree and composition. For the first time, we have identified clear trends in the single NC linewidth and biexciton quantum yield of P-NCs. Our results provide insights into the properties of lead halide perovskite excitons in the confined regime and are directly applicable for the synthetic improvement of P-NCs.

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3) Y. Park *et al*., *J. Am. Chem. Soc*., **2015**, *10*, 10386-10393.
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