

Limiting exciton diffusion enhances the photoluminescence of CsPbBr₃ nanocrystal films at high excitation densities

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Nanocrystals (NCs) used as scintillators in heterostructured detector designs offer a promising route to improved performance, notably for fast-timing applications such as time-of-flight PET. Such applications require a combination of high light yield, ultrafast emission, and adequate γ -ray stopping power which are criteria that cannot be fulfilled by a single material. Lead-halide perovskite NCs, notably CsPbBr₃, have recently attracted attention in this context. A recent study demonstrated their strong scintillation potential, attributed to the multiexcitonic nature of their emission, and identified NCs with diameters exceeding 10 nm as especially favorable [1].

While ref. [1] addresses the response of isolated NCs, realistic detector implementations inevitably involve dense ensembles where interactions between excited NCs may become significant. Here, we investigate such interactions in drop-cast films of CsPbBr₃ NCs under intense femtosecond-laser excitation. By employing optical excitation, this approach enables an evaluation of the optical performance of the films at excitation densities relevant to ionizing radiation, independently of any surrounding matrix material [2]

Results from this study, shown in Fig. 1, demonstrate that the choice of surface ligand strongly influences the optical performance of drop-cast NC films. In particular, organic ligands lead to higher photoluminescence (PL) efficiency at dense excitation densities compared to inorganic passivation. To explain this behavior, we model exciton dynamics by incorporating linear decay, nonlinear bimolecular Auger recombination, and, critically, exciton diffusion between NCs. The model reproduces the experimental data by assigning different exciton diffusion rates to the two samples. Together, the experimental data and numerical modeling indicate that suppressing inter-NC exciton diffusion is a viable pathway toward improved nonlinear optical performance and provides a predictive framework for designing ultrafast luminescent nanocomposite materials.

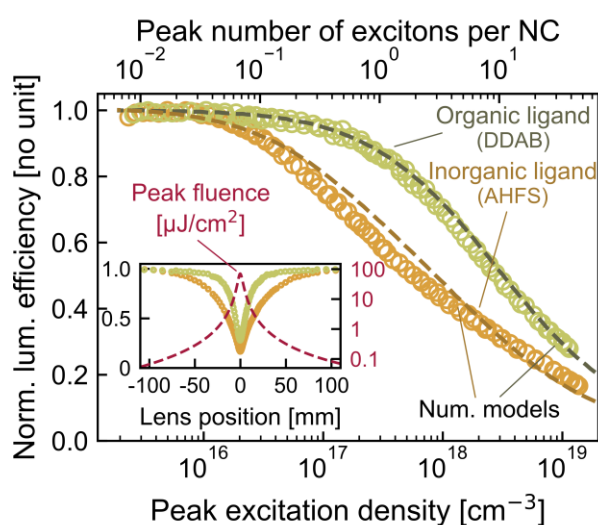


Figure 1: Z-scan luminescence measurements of two dropcast films fabricated from identical syntheses of CsPbBr₃ nanocrystals (NCs) with different surface passivation schemes. The photoluminescence (PL) intensity is recorded as a function of lens position (see inset) and converted to excitation density within the films. The data is reproduced well by numerical models (dotted lines), in which the primary difference between the samples is the exciton diffusion rate.

1. A. Fratelli et al., “Size-dependent multiexciton dynamics governs scintillation from perovskite quantum dots,” *Adv. Mater.*, **37**, 2413182 (2024)
2. S. Jessen et al., “Nonlinear quenching of excitonic emission from nanoplatelet films at high excitation densities,” *Sci. Rep.*, **23**, 23423 (2025)