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## Surface Defects Affect Fluorescence of Caesium Lead Halide Perovskite Supercrystals

Advances in the self-assembly of colloidal nanocrystals (NCs) from solution into three-dimensional arrays with long-range order have enabled the design of microscopic “supercrystals” that approach the structural precision of atomic single crystals. The individual NCs, which are the building blocks of a supercrystal, are often regarded as “artificial atoms”, and hence analogies between atomic crystals and such supercrystals have been made. In view of the recent progress in exploiting the massive structural coherence in NC supercrystals to generate collective optoelectronic properties, a critical question remains whether this artificial atom analogy can be extended towards the optical properties of NC supercrystals.

In this work, we show that in close analogy to atomic crystals,  $\text{CsPbBr}_2\text{Cl}$  and  $\text{CsPbBr}_3$  NC supercrystals exhibit structural distortions near their surfaces which significantly alter their fluorescence properties.[1] Confocal fluorescence microscopy of individual  $\text{CsPbBr}_3$  supercrystals displayed spatial variations in the fluorescence peak wavelength and intensity, indicating that local structural inhomogeneities may substantially affect the fluorescence properties of the entire supercrystal. Our approach is based on simultaneous WAXS and SAXS with a nano-focused beam to probe the structural defects and crystallographic orientation of the supercrystal and the constituting NCs on a local level, respectively. By correlation with diffraction-limited confocal fluorescence microscopy and modeling with density functional theory (DFT), we present a proof that compressive strain, a loss of structural coherence, and an increasing atomic misalignment between adjacent nanocrystals at the edges of perovskite NC supercrystals are responsible for a blue-shifted emission and decrease of the fluorescence lifetimes.

References:

1. D. Lapkin et al., “Spatially resolved fluorescence of caesium lead halide perovskite supercrystals reveals quasi-atomic behavior of nanocrystals”, *Nature Commun.* 13 (2022), 892

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