Size-dependent halide segregation of single mixed-halide perovskite nanocrystals

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It is well known that $CsPbX_3$ (X= Cl, Br, I) perovskite nanocrystal (PNC) is unstable under ambient atmosphere owing to its ionic properties. In the earlier research, single PNC exhibited photoluminescence (PL) spectrum blue-shift upon continuous photoirradiation which attributed to decreasing PNC size. [1] Moreover, halide segregation was observed in mixed halide ($CsPbBr_{1.2}I_{1.8}$) PNC due to halide migration during continuous photoirradiation.[2] However, size dependent halide segregation process in single PNCs has not been investigated, particularly in a PNCs with size beyond the quantum confinement regime. PNC size larger than the quantum confinement of approximately 11 nm allow the formation of multiple emission sites in a single PNC.

Herein, we successfully observed the photodegradation and size dependent halide segregation process at the single $CsPbBr_xI_x$ PNCs using a simultaneous atomic force microscope (AFM) and single-particle spectroscopy measurement. We obtained size dependent behavior of PL spectra, photon correlation histogram, and PL lifetime. Fig. 1 shows the blueshift of PL peaks upon continuous photoirradiation. The blueshift is attributed to the halide loss and decreasing size during photodegradation process obtained from AFM measurements. Our results show that small sized (11 nm) single PNCs exhibited single PL peak and photon antibunching behavior which attributed to quantum confinement effect. On the other hand, large (25 nm) PNCs showed multiple PL peak which attributed to the nucleation of Br^- and I^- rich region after photoirradiation induced halide segregation process. The Large PNC also exhibited multiphoton emission as shown by high $g^{(2)}(0)$ value (> 0.25) obtained from photon correlation histogram, which indicates the formation of multiple emission sites in a single large PNCs. This work shows the size dependent emission behavior and halide segregation of single PNC at the single PNC level.

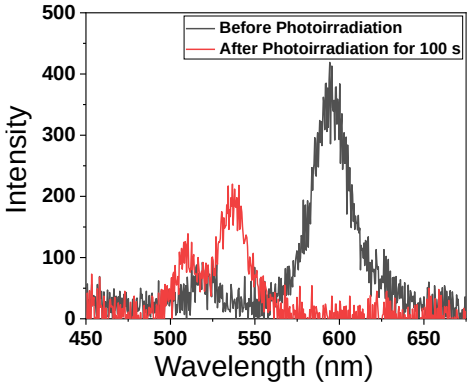

Fig. 1. PL peaks of a single PNC (25 nm) before (black) and after continuous photoirradiation for 100 s (red).

1) Y. A. Darmawan, M. Yamauchi, S. Masuo, *J. Phys. Chem. C* **2020**, *124*, 18770-18776.
2) H. Zhang, X. Fu, Y. Tang, H. Wang, C. Zhang, W. W. Yu, X. Wang, Y. Zhang, M. Xiao, *Nat. Commun.* **2019**, *10*, 1088.