An Evaluation of the Shape-dependent Kinetics of Halide Vacancy Filling in Lead Halide Perovskites

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Keywords: Halide Perovskite; Photoluminescence; Halide Vacancy; Crystal Shape; Microcrystal

Lead halide perovskites (LHPs), owing to their high photoluminescence (PL) quantum yields (QY) and excellent charge carrier transport properties, are one of the most promising semiconductor materials for next-generation optoelectronic devices. However, the PL QYs of LHPs are significantly decreased due to the halide vacancies. We investigate the crystal shape- and halide vacancy- dependent recombination rates of photogenerated charge carriers in methylammonium lead bromide ($MAPbBr_3$) perovskite microcrystals.

Microcrystals [(microcuboids (Figure 1 a), microplates (Figure 1b), and microrods (Figure 1c)] of $MAPbBr_3$ with 10 to 20 μm edge lengths are prepared on glass coverslips. These crystals are immersed in a MABr solution and exposed to a picosecond (ps) laser beam to fill the Br^- vacancies. The PL decays (Figure 1d-f) of these microcrystals are recorded by using a time-correlated single-photon counting (TCSPC) system. The time evolution of the PL lifetimes of the microcrystals is shown in Figure 1g. The PL lifetime of the microcuboid in the MABr solution is increased only slightly by the ps laser irradiation (Figure 1d,g), whereas the PL lifetimes of the microplate (Figure 1e,g) and the microrod (Figure 1f,g) in the MABr solution are increased enormously by the ps laser irradiation. Nonetheless, in the case of the microrod, the PL lifetime is decreased by further ps laser irradiation. The increases in the PL lifetimes are attributed to filling of the Br^- vacancies. We discuss the differences in the photoinduced filling of the Br^- vacancy from the view point of the difference in the shapes of the microcrystals.

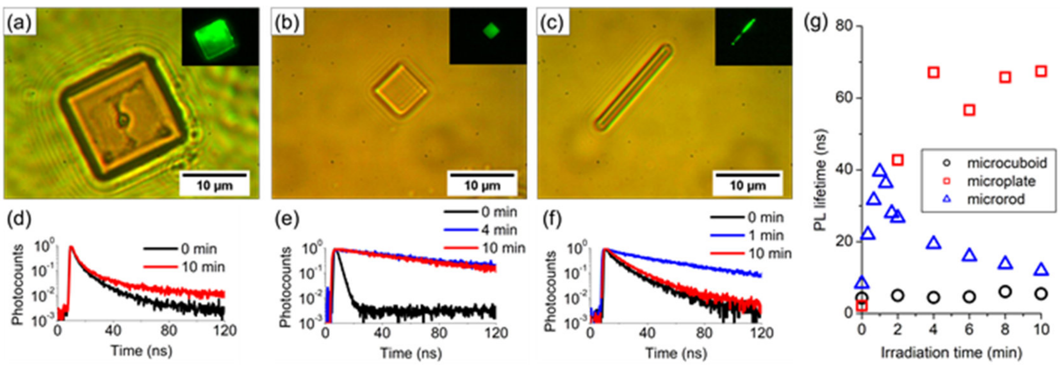

Figure 1. Optical microscope images and PL decay curves of (a, d) a microcuboid, (b, e) a microplate, and (c, f) a microrod. The insets show the PL images of the crystals. (g) The time evolutions of the PL lifetimes of the microcuboid (○), the microplate (□) and the microrod (△).