Supporting Information

Prolonged Phase Segregation of Mixed-Halide Perovskite Nanocrystals in the Dark

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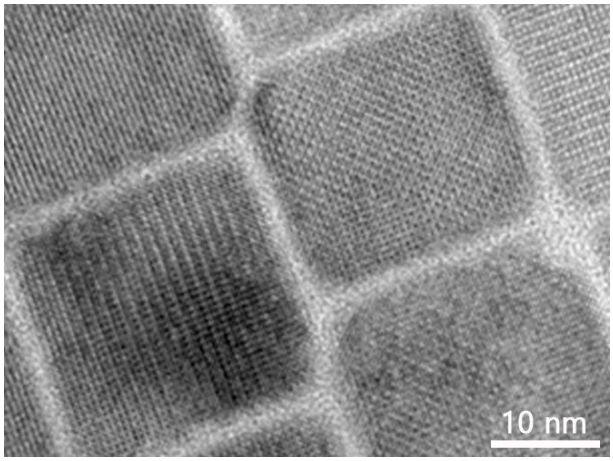

Figure S1. Transmission electron microscope image measured for the cuboid $CsPbBr_{1.2}I_{1.8}$ NCs with an average edge length of ~23 nm.

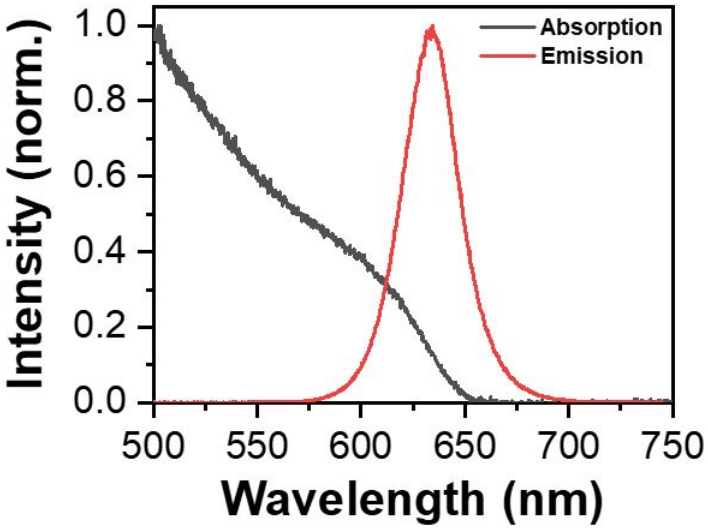

Figure S2. Absorption and emission spectra measured for the $CsPbBr_{1.2}I_{1.8}$ NCs in a hexane solution.

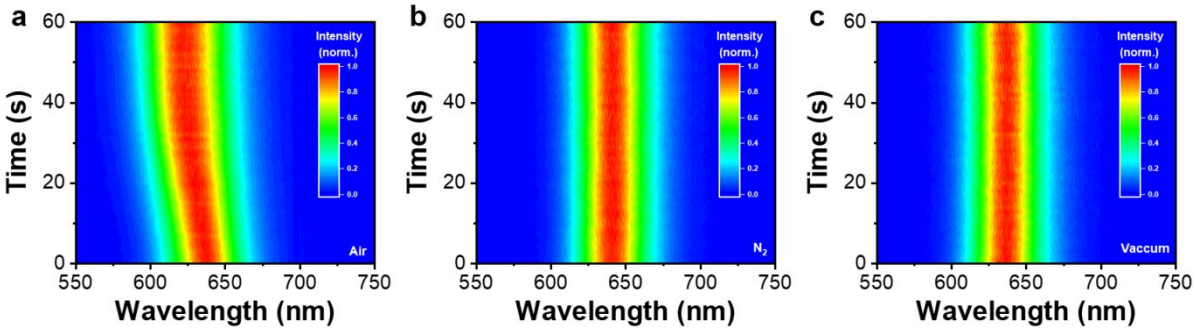

Figure S3. Time-dependent evolution of the 60 PL spectra each acquired with an integration time of 1 s for the $CsPbBr_{1.2}I_{1.8}$ NCs under the **a,** ambient, **b,** nitrogen and **c,** vacuum conditions, respectively. In the ambient air, the $CsPbBr_{1.2}I_{1.8}$ NCs show the phase segregation effect as a blue shift in the PL peak, which is completely missing under the nitrogen and vacuum conditions. In the above optical measurements, the $CsPbBr_{1.2}I_{1.8}$ NCs are excited by a 405 nm CW laser at the power density of ~50 W/cm^2.

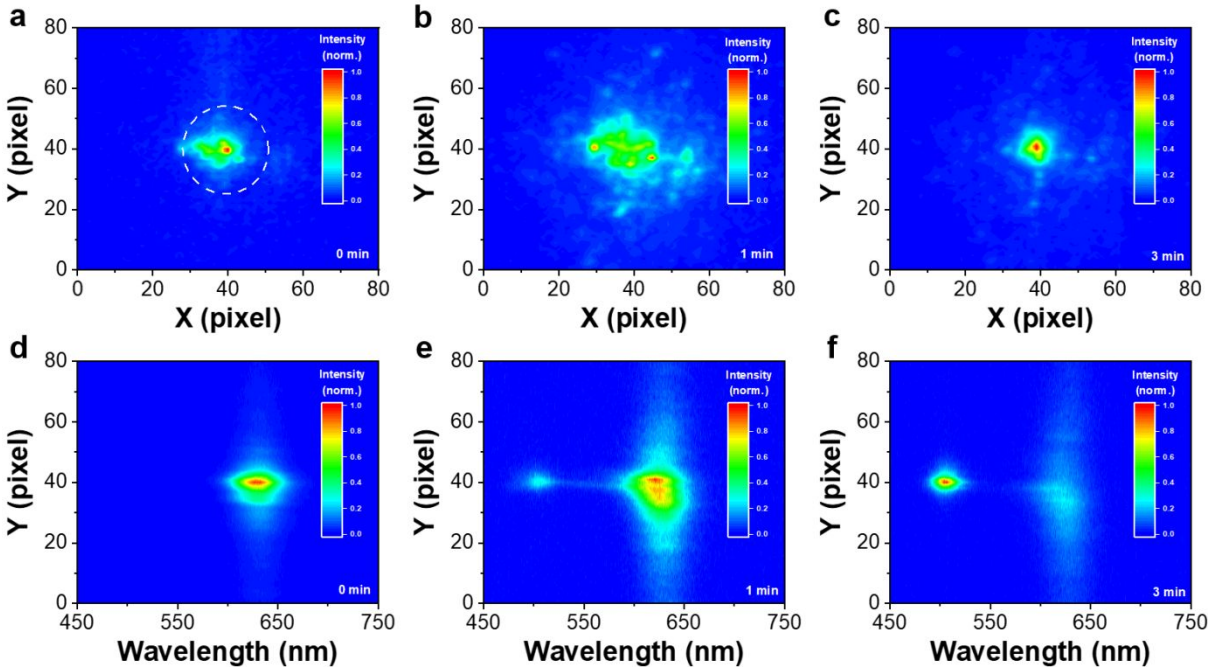

Figure S4. PL spatial images measured for the $CsPbBr_{1.2}I_{1.8}$ NCs in the **a,** beginning, **b,** middle and **c,** end of the phase segregation process, respectively. PL spectral images measured for the $CsPbBr_{1.2}I_{1.8}$ NCs in the **d,** beginning, **e,** middle and **f,** end of the phase segregation process, respectively. The dashed circle in **a** marks the ~3.8 μm FWHM (full width at half maximum) of the 405 nm CW laser spot with a Gaussian profile and a power density of ~50 W/cm^2. In **a-f**, each pixel corresponds to a spatial dimension of ~0.125 μm.

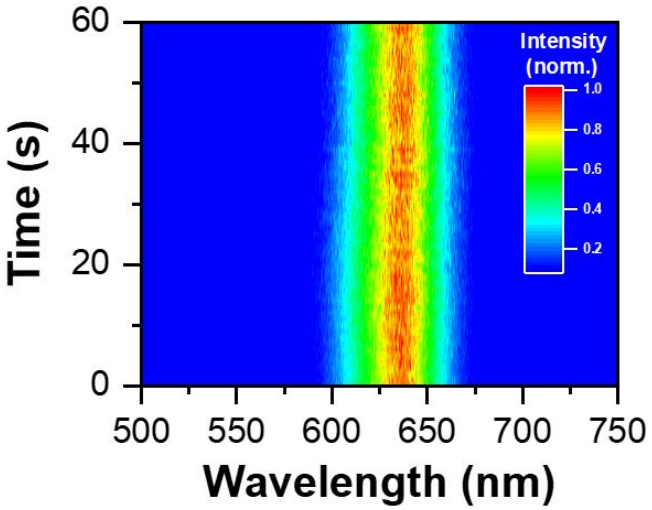

Figure S5. Time-dependent evolution of the 60 PL spectra each acquired with an integration time of 1 s for the $CsPbBr_{1.2}I_{1.8}$ NCs. These $CsPbBr_{1.2}I_{1.8}$ NCs show no sign of phase segregation under the continuous excitation of a 405 nm CW laser at the power density of ~3 W/cm^2.

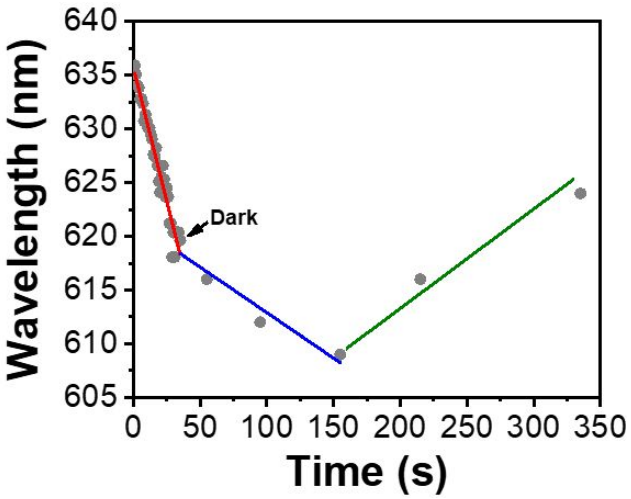

Figure S6. Time-dependent PL peak positions extracted from Figure 2a,b in the main text for the $CsPbBr_{1.2}I_{1.8}$ NCs during the light-induced phase segregation, the dark-prolonged phase segregation and the following phase remixing processes. In the above optical measurements, the $CsPbBr_{1.2}I_{1.8}$ NCs are excited by a 405 nm CW laser at the power density of ~50 W/cm^2 for only 35 s to induce the partial phase segregation. Upon laser excitation of the $CsPbBr_{1.2}I_{1.8}$ NCs, the PL peak shifts promptly to the blue side with a speed of ~30 nm/min (the solid red line). When the laser excitation is removed after 35 s, the PL peak still shifts to the blue side albeit with a slower speed of ~5 nm/min (the solid blue line). This dark-prolonged phase segregation lasts for ~115 s, and then the PL peak shifts to the red side to start the phase remixing process with a speed of ~5 nm/min (the solid green line).

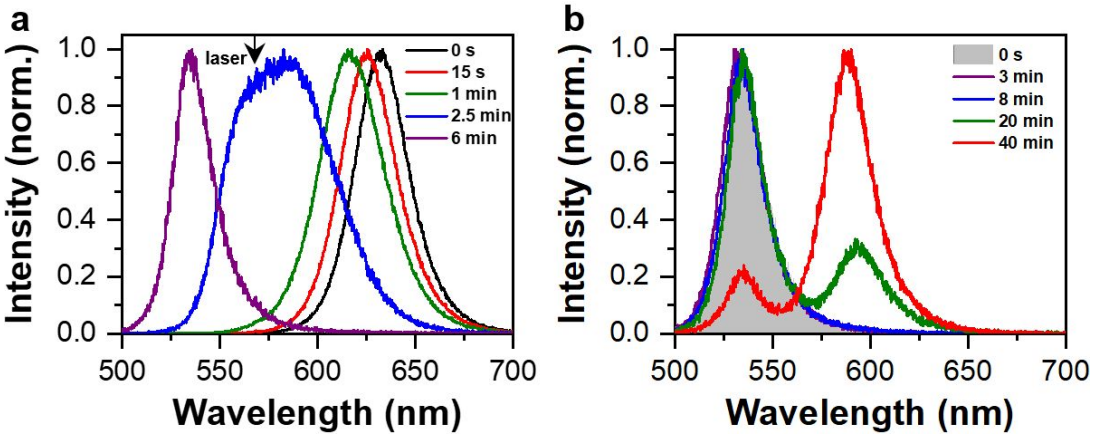

Figure S7. a, PL spectra measured for the $CsPbBr_{1.2}I_{1.8}$ NCs after being excited by a 568 nm CW laser at the power density of ~5000 W/cm^2 for 0 s, 15 s, 1 min, 2.5 min and 6 min, respectively. At each of the above time points, the 568 nm laser is blocked for 1 s while a 405 nm CW laser is employed to acquire the PL spectrum at the power density of ~3 W /cm^2. The solid black arrow on top marks the wavelength position of the 568 nm excitation laser. **b,** PL spectra measured for these $CsPbBr_{1.2}I_{1.8}$ NCs after the 568 nm laser has then been blocked for 0 s, 3 min, 8 min, 20 min and 40 min, respectively. At each of the above time points, a 405 nm CW laser is unblocked for 1 s to acquire the PL spectrum at the power density of ~3 W /cm^2.

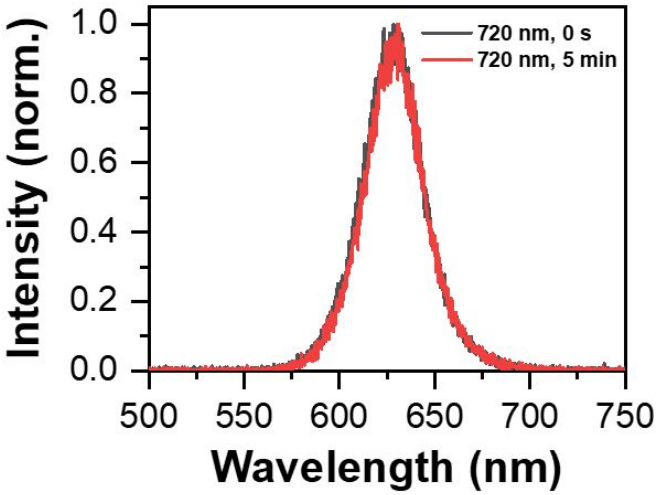

Figure S8. PL spectra measured for the $CsPbBr_{1.2}I_{1.8}$ NCs after being excited for 0 s and 5 min by a below-bandgap 720 nm pulsed laser at the power density of ~5000 W/cm^2, showing that there is no thermal effect to induced the phase segregation process. At each of these two time points, the 720 nm laser is blocked while a 405 nm CW laser is employed for 1 s to acquire the PL spectrum at the power density of ~3 W /cm^2.

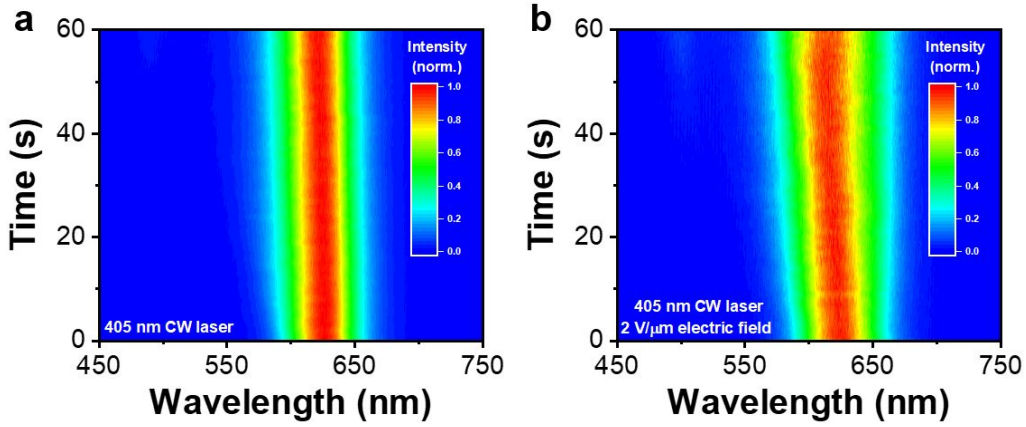

Figure S9. Time-dependent evolution of the 60 PL spectra each acquired with an integration time of 1 s for the $CsPbBr_{1.2}I_{1.8}$ NCs under **a,** the 405 nm CW laser excitation with a power density of ~50 W/cm^2 and **b,** the additional application of a 2 V/μm electric field. Under the joint influence of laser excitation and electric biasing, the PL peak of the $CsPbBr_{1.2}I_{1.8}$ NCs shifts to the blue side at a speed of ~9 nm/min, which is faster than that of ~6 nm/min measured with only laser excitation.

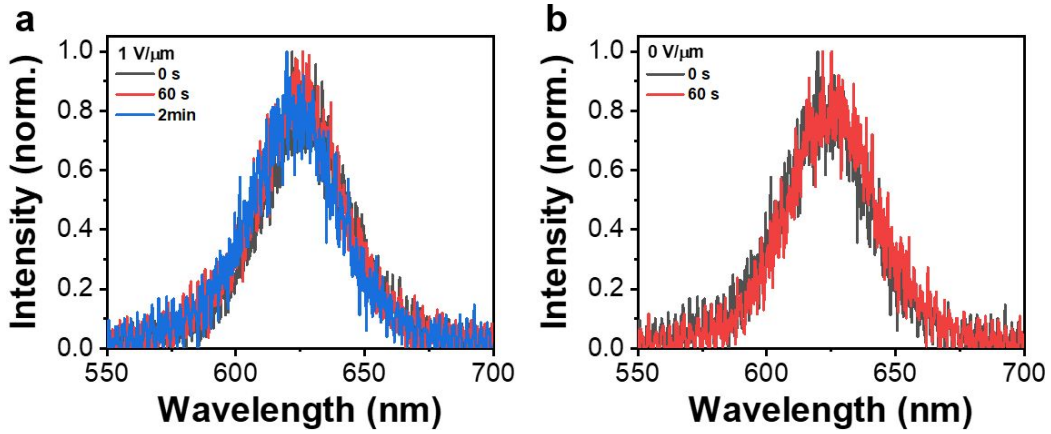

Figure S10. a, PL spectra measured for the $CsPbBr_{1.2}I_{1.8}$ NCs after an electric field of 1 V/μm has been applied for 0 s, 60 s and 2 min in the dark. **b,** PL spectra measured for these $CsPbBr_{1.2}I_{1.8}$ NCs after the electric field has then been removed for 0 s and 60 s in the dark. At each time point in **a** and **b**, a 405 nm CW laser is unblocked for 1 s to acquire the PL spectrum at the power density of ~3 W/cm^2.

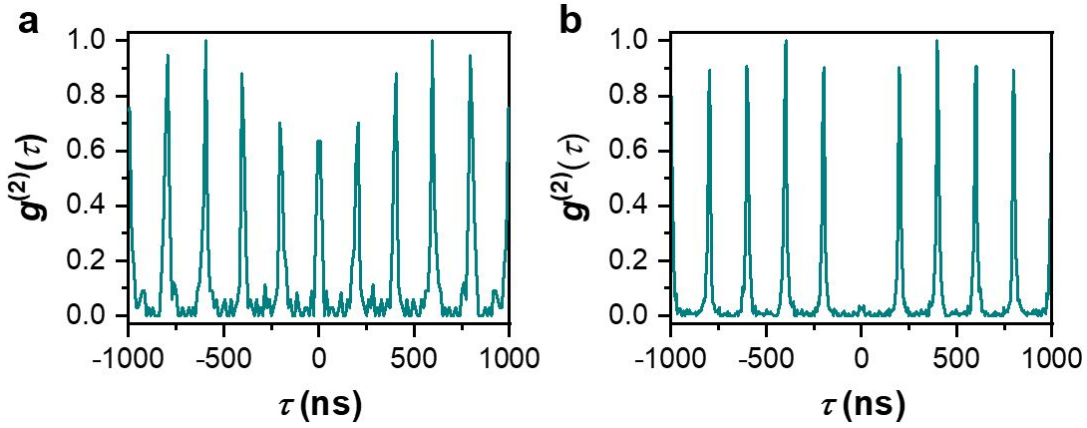

Figure S11. a, Second-order photon correlation measurement of a single $CsPbBr_{1.2}I_{1.8}$ NC with the $g^{(2)}(0)$ value of ~0.636 and the single-photon emission purity of ~36.4%. **b,** Second-order photon correlation measurement of a single $CsPbBr_3$ NC with the $g^{(2)}(0)$ value of ~0.022 and the single-photon emission purity of ~97.8%. The single $CsPbBr_{1.2}I_{1.8}$ or $CsPbBr_3$ NC is excited at room temperature by a 5 MHz 405 nm pulsed laser at the power density of ~3 W /cm^2.

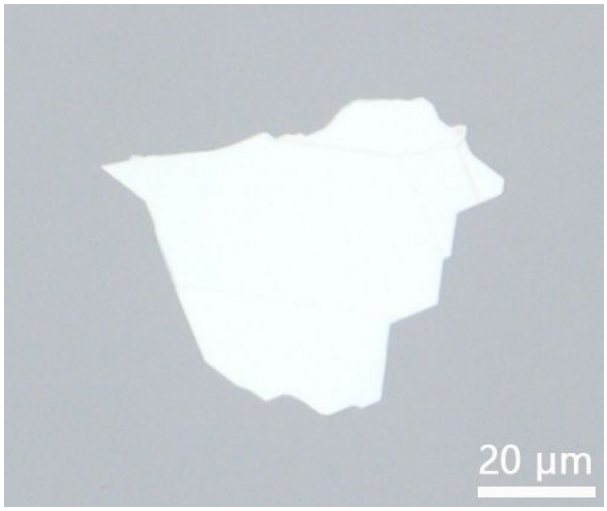

Figure S12. Optical microscope image of an hBN flake (white color) with the thickness of ~150 nm.

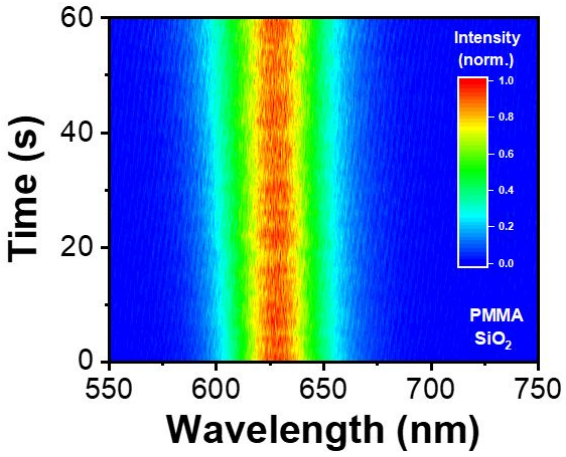

Figure S13. Time-dependent evolution of the 60 PL spectra each acquired with an integration time of 1 s for a solid film made of the $CsPbBr_{1.2}I_{1.8}$ NCs and the PMMA (polymethyl methacrylate) molecules on the SiO_2 substrate, showing that the phase segregation is almost completely suppressed under the 405 nm CW laser excitation with a power density of ~50 W/cm^2.

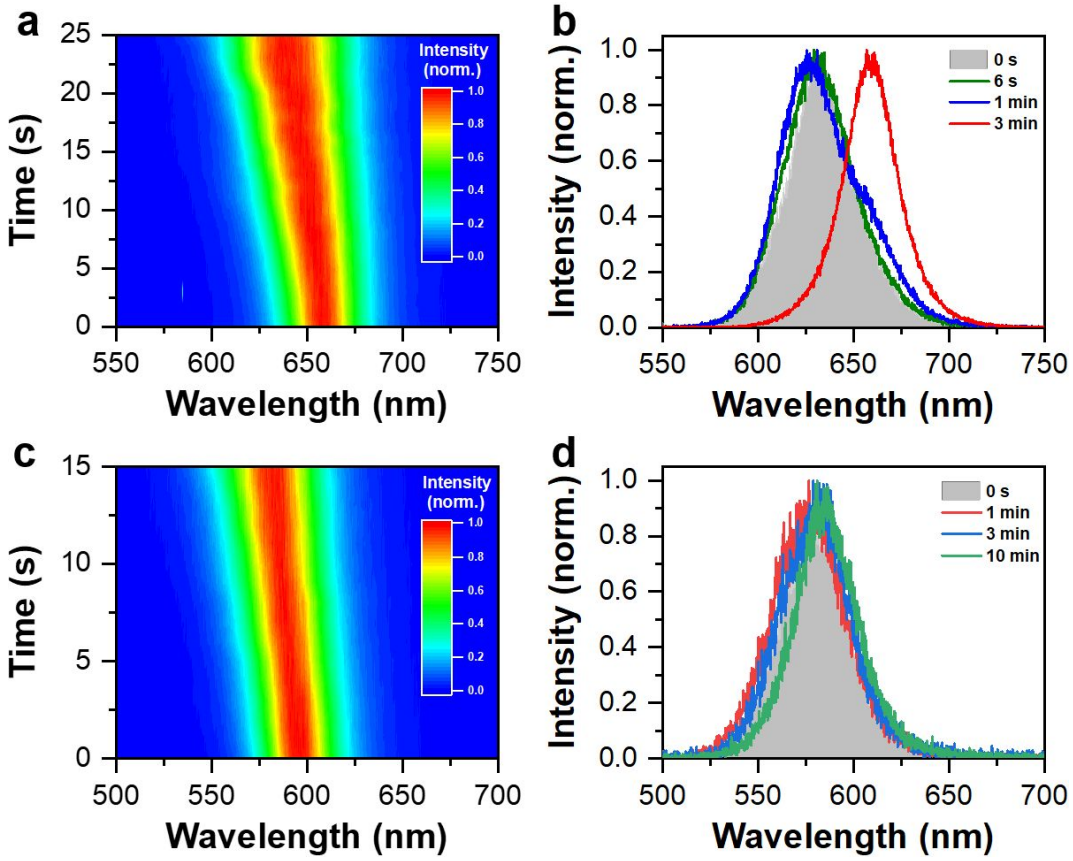

Figure S14. a, Time-dependent evolution of the 25 PL spectra each acquired with an integration time of 1 s for the $CsPbBr_{0.9}I_{2.1}$ NCs. The phase segregation process is incomplete after 25 s continuous excitation of the $CsPbBr_{0.9}I_{2.1}$ NCs by a 405 nm CW laser at the power density of ~50 W/cm^2. **b,** PL spectra measured for these $CsPbBr_{0.9}I_{2.1}$ NCs after the 405 nm laser has then been blocked for 0 s, 6 s, 1 min and 3 min, respectively. At each of the above time points, the 405 nm laser is unblocked for 1 s to acquire the PL spectrum at a power density of ~3 W/cm^2. **c,** Time-dependent evolution of the 15 PL spectra each acquired with an integration time of 1 s for the $CsPbBr_{1.5}I_{1.5}$ NCs. The phase segregation process is incomplete after 15 s continuous excitation of the $CsPbBr_{1.5}I_{1.5}$ NCs by a 405 nm CW laser at the power density of ~50 W/cm^2. **d,** PL spectra measured for these $CsPbBr_{1.5}I_{1.5}$ NCs after the 405 nm laser has then been blocked for 0 s, 1 min, 3 min and 10 min, respectively. At each of the above time points, the 405 nm laser is unblocked for 1 s to acquire the PL spectrum at a power density of ~3 W/cm^2.

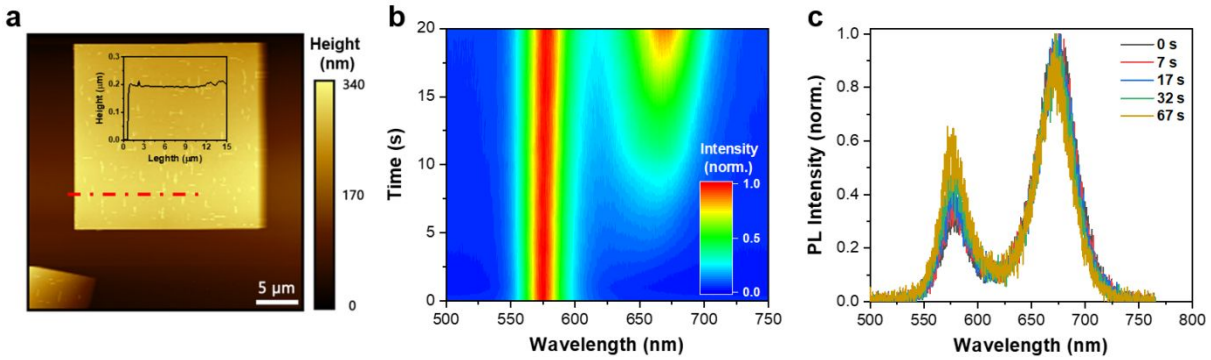

Figure S15. a, Atomic force microscope image measured for an individual $CsPbBr_{1.5}I_{1.5}$ microplate with the RMS (root mean square) surface roughness of ~10.56 nm. Inset: Height profile measured across the red dash-dot line. **b,** Time-dependent evolution of the 20 PL spectra each acquired with an integration time of 1 s for this microplate. Manifested as the appearance of a red-shifted PL peak, the phase segregation process is incomplete after this microplate has been continuously excited for 20 s by a 405 nm CW laser at the power density of ~50 W/cm^2. **c,** PL spectra measured for this partially-segregated microplate after the 405 nm laser has then been blocked for 0 s, 7 s, 17 s, 32 s and 67 s, respectively. At each of the above time points, the 405 nm laser is unblocked for 1 s to acquire the PL spectrum at a power density of ~5 W /cm^2. Normalized with respect to the red-shifted PL peak, these PL spectra demonstrate that the PL intensity of the original $CsPbBr_{1.5}I_{1.5}$ composition is instantly recovered upon removal of the laser excitation.