

Supplementary Information

**Laser Ablation Process of CsPbBr₃ Heterostructures
for LED Applications**

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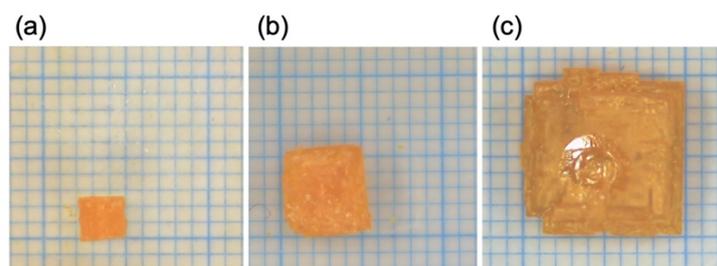


Figure S1. (a) Photograph of a CsPbBr₃ single crystal grown via the first ITC process. (a) The first preparation produced a 3 × 3 mm crystal that was used as the seed crystal for the second preparation. (b) and (c) show the photographs captured after the 2nd and 3rd preparation process, respectively.

Powders of CsBr (Tokyo Chemical Industry Co.: C2202) and PbBr₂ (Tokyo Chemical Industry Co., L0364) were dissolved in DMSO (Tokyo Chemical Industry Co., D0798) at a molar ratio of 1:2. The solution was stirred at 60 °C to prepare a supersaturated solution. The solution was filtered to remove the undissolved powder. The supersaturated solution was placed in a glass bottle, and the temperature was increased from 60 °C to 120 °C at a rate of 0.06 °C/min to precipitate CsPbBr₃ crystals. To maintain the uniform temperature of the glass bottle during heating, it was immersed in silicone oil.

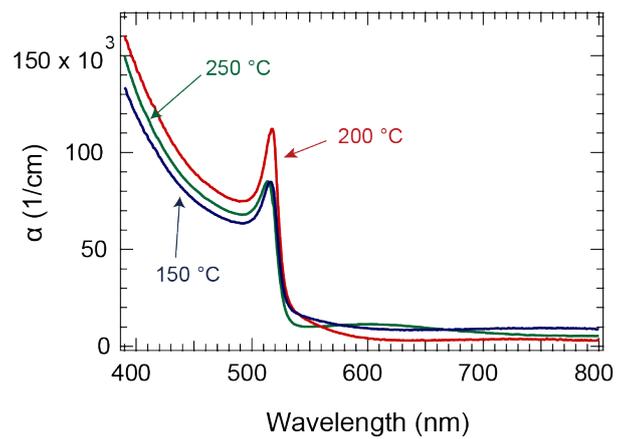


Figure S2. The optical absorption spectra of the CsPbBr₃ films grown at 150 °C (blue), 200 °C (red), and 250 °C (green).

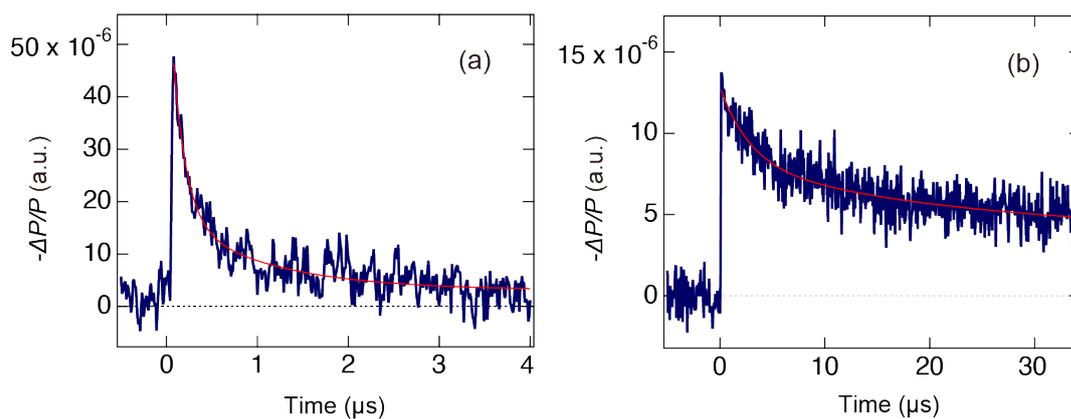


Figure S3. The transient profiles of TRMC signals for a CsPbBr₃ powder (a) and CsPbBr₃ single crystal (b). The red line denotes the fitting by using the double exponential function.

To investigate the relationship between film crystallinity and photocarrier lifetime, TRMC measurements were performed on CsPbBr₃ powder (Tokyo Chemical Industry Co.: C3569) and CsPbBr₃ single crystals prepared using the ITC method. Both the samples were placed in quartz tubes with a diameter of 5 mm. A CsPbBr₃ single crystal with an area of 1 cm² was crushed to the size of a few millimeters and inserted into a quartz tube. Figure S2 (a) and (b) show the TRMC profiles of the CsPbBr₃ powder and single crystal, respectively. The red plot shows the fitting profile used to systematically calculate the half-life and maximum intensity of the TRMC signal. The maximum value of the TRMC signal and half-life in the manuscript were calculated in a similar manner.

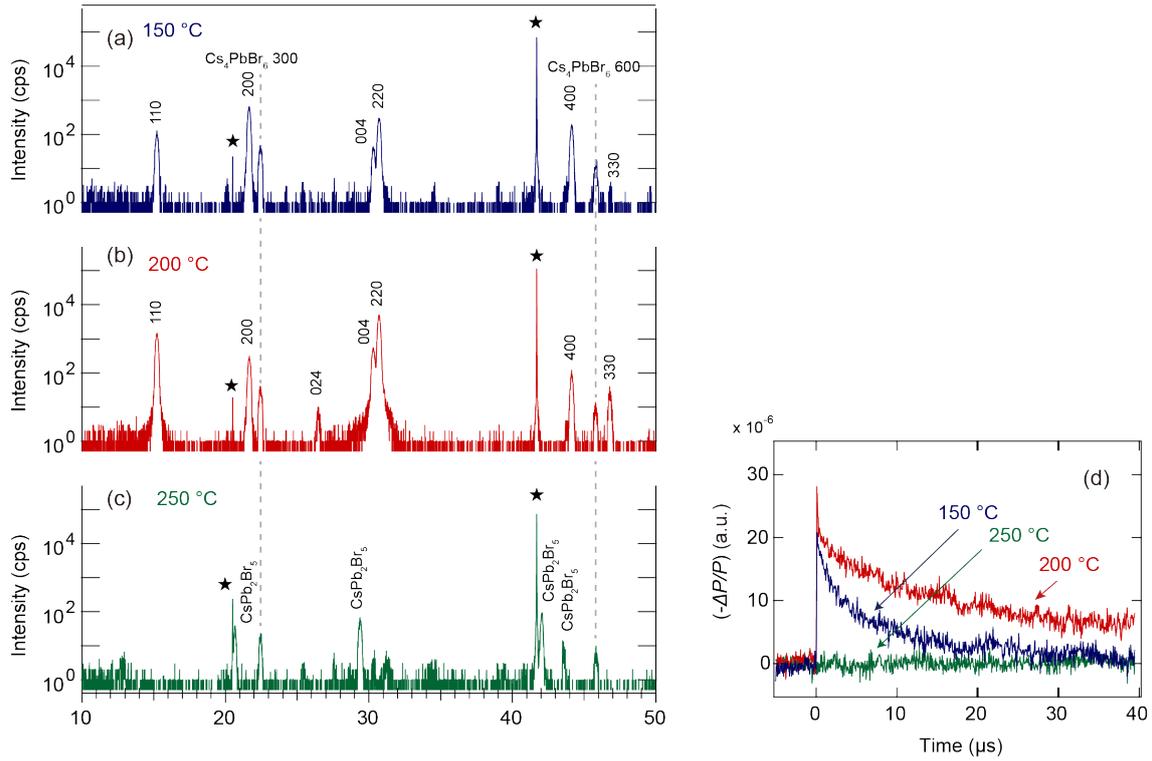


Figure S4. (a-c) XRD patterns of CsPbBr₃ films grown at 150 °C (a, d), 200 °C (b, e), and 250 °C (c, f). The symbol “★” in (a-c) denote the substrate peak. (d) Transient profiles of the $(-\Delta P/P)$ signals for CsPbBr₃ films grown at 150 °C (blue), 200 °C (red), and 250 °C (green).

To investigate the reproducibility of the growth temperature dependence on the film crystallinity and TRMC properties, CsPbBr₃ films were prepared at 150 °C, 200 °C, and 250 °C, again. Figure S4 (a-c) shows the XRD patterns of CsPbBr₃ films grown at 150 °C, 200 °C, and 250 °C. The best crystallinity was obtained from the growth at 200 °C. Figure S4 (d) shows the transient TRMC profiles of CsPbBr₃ films grown at 150 °C, 200 °C, and 250 °C. The longest photocarrier lifetime and largest TRMC signal was observed for CsPbBr₃ films grown at 200 °C. The best growth temperature of 200 °C is reproducible for the CsPbBr₃ film growth by PLD.

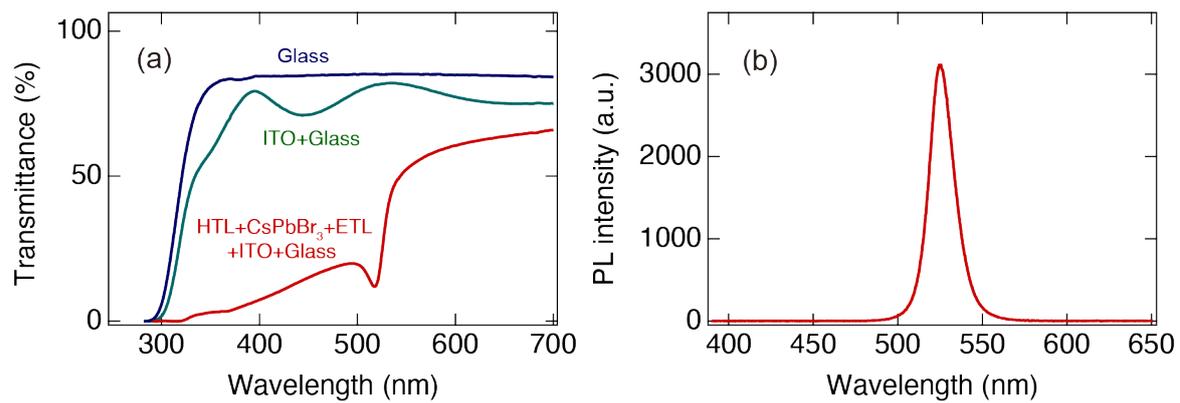


Figure S5. (a) Optical transmittance and (b) PL spectrum of a CsPbBr₃ LED device