

## Supplementary information

### Long Photocarrier Lifetimes in $\text{CsPbBr}_3$ Films Deposited by Machine-learning-assisted IR-Laser Molecular Beam Epitaxy

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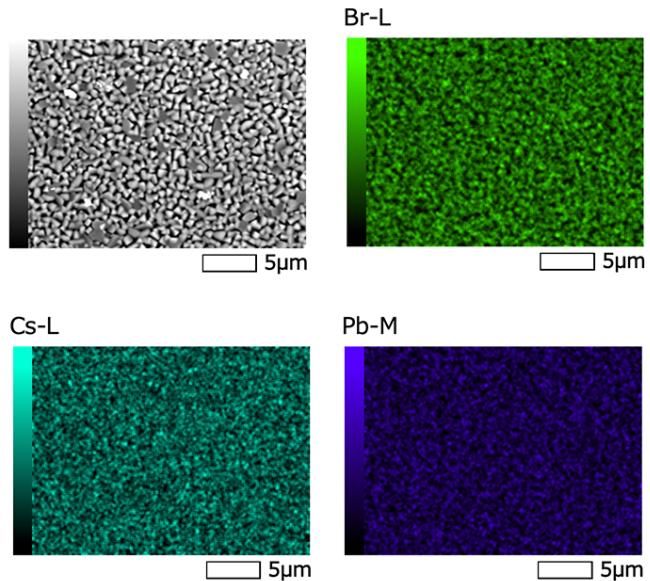
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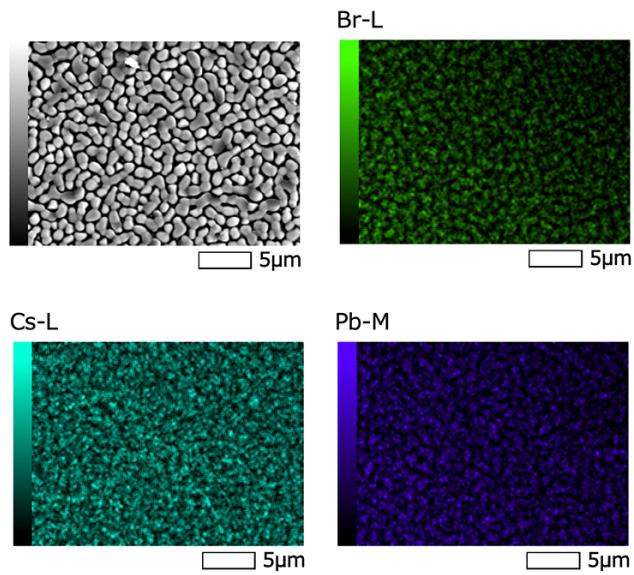
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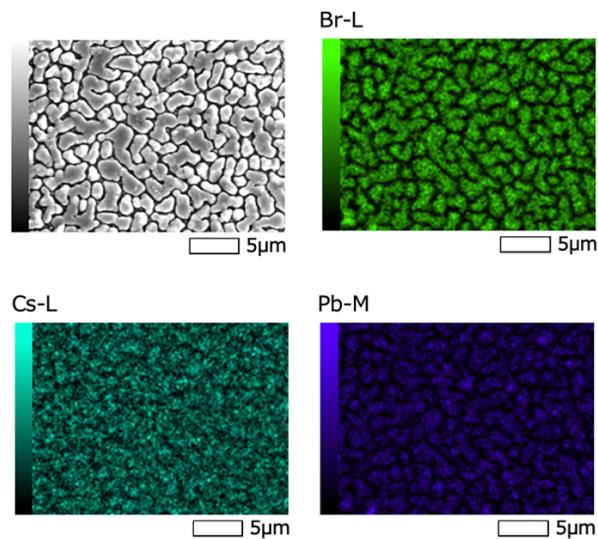
### Condition A



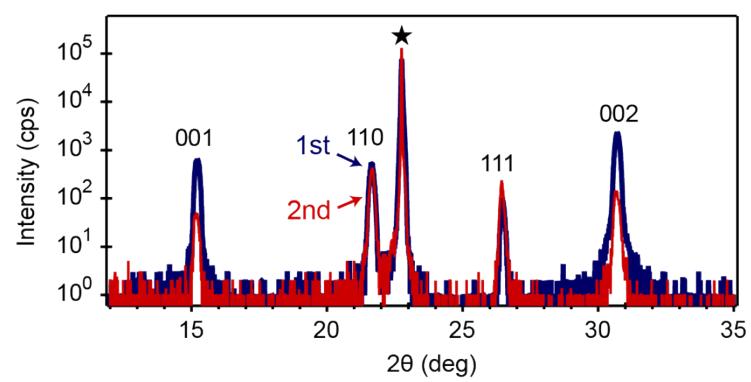
### Condition B



### Condition C

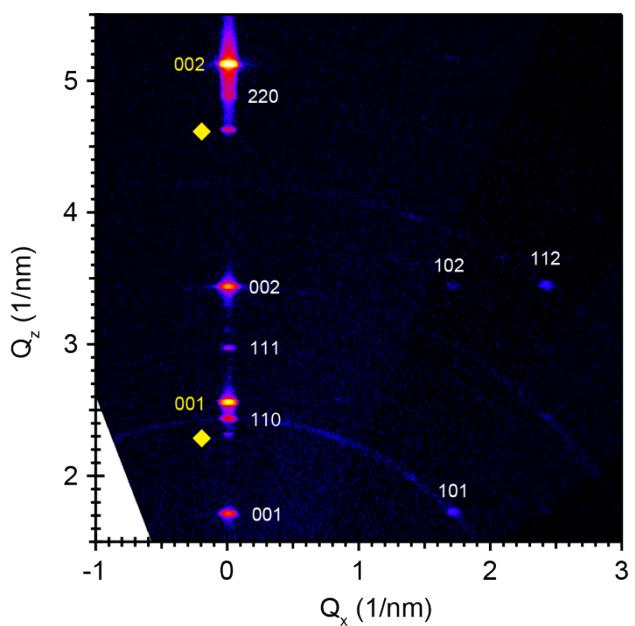


**Figure S1** : SEM-EDS mappings of  $\text{CsPbBr}_3$  films grown under condition A, B, C. The Cs/Pb ratios of  $\text{CsPbBr}_3$  films grown under conditions A, B, C were measured to be 0.997, 0.995, and 0.921, respectively.

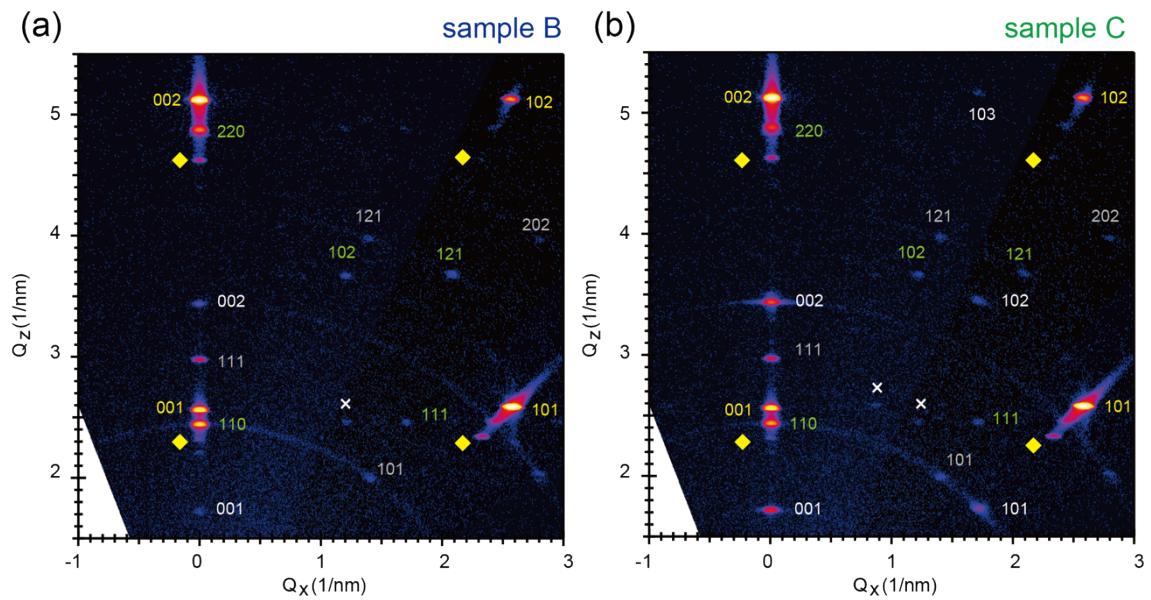


**Figure S2 :** XRD patterns to investigate the reproducibility of two  $\text{CsPbBr}_3$  films grown under condition A.

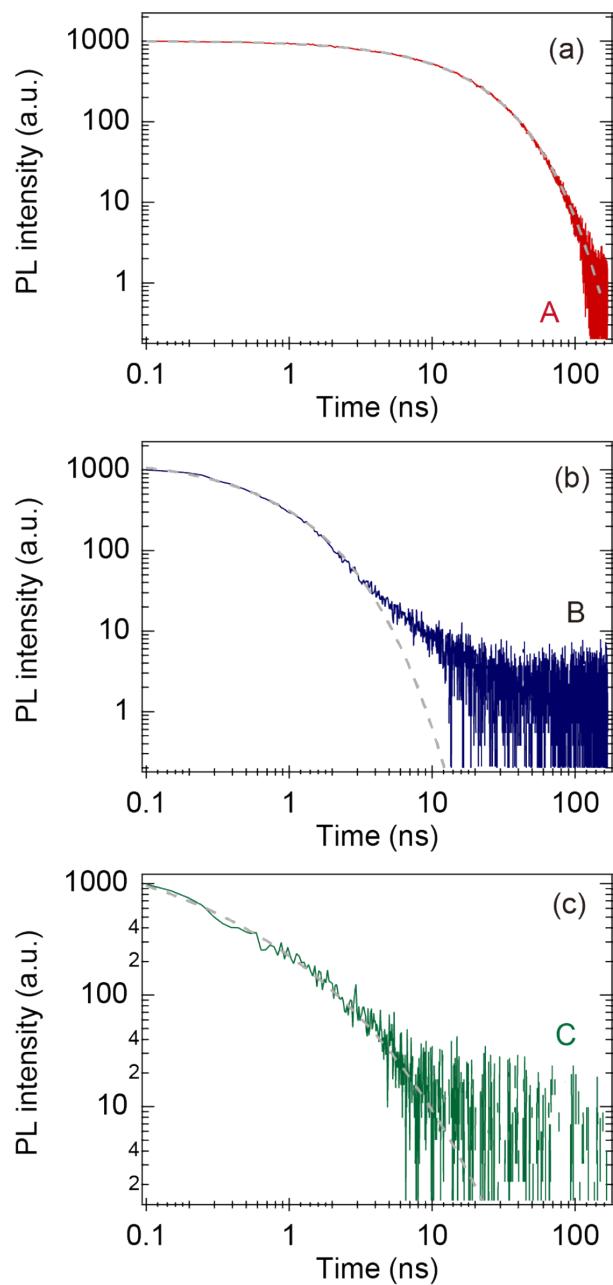
By the deposition under the growth window, no impurity peak originated from  $\text{CsPb}_2\text{Br}_5$  phase were visible.



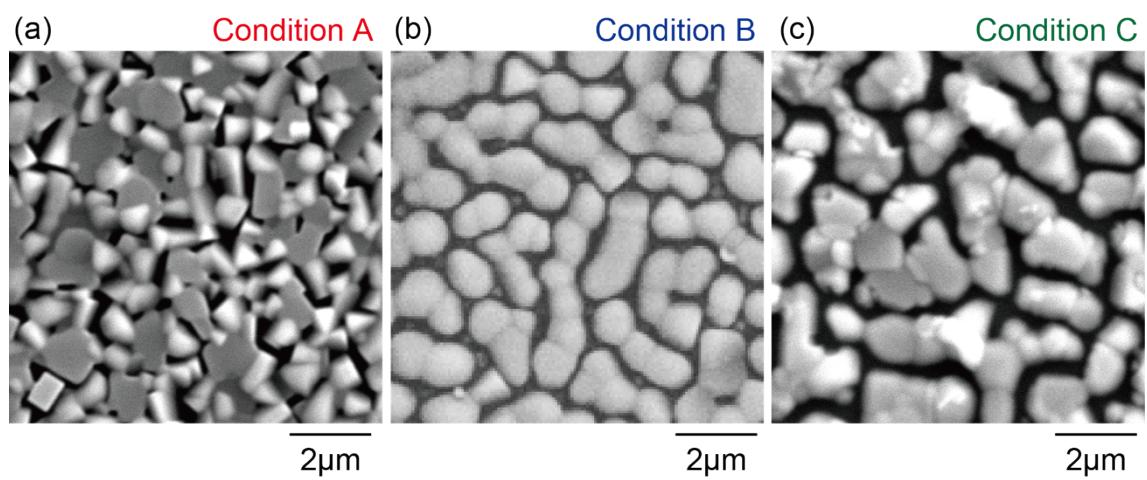
**Figure S3 :** Wide-range reciprocal space mapping along the HHL direction of a  $\text{CsPbBr}_3$  film grown under condition A on a  $\text{SrTiO}_3$  substrate. White and yellow characters denote the reflections of the film and substrate, respectively. “◆” denotes the  $K_\beta$  reflection of the substrate crystal.



**Figure S4** : Wide-range reciprocal space mappings along the HOL direction of  $\text{CsPbBr}_3$  films grown under conditions B (a) and C (b) on  $\text{SrTiO}_3$  substrates. White, green, gray, and yellow characters denote the reflections of the c-axis, (110)-oriented, (111)-oriented  $\text{CsPbBr}_3$ , and  $\text{SrTiO}_3$  substrate, respectively. “◆” and “×” denotes the  $\text{K}_\beta$  reflection of the substrate crystal and unknown phase, respectively.



**Figure S5 :** Transient TRPL profiles of a  $\text{CsPbBr}_3$  film grown at conditions A (a), B (b), and C (c), respectively. The gray dotted line denotes the fitting results.



**Figure S6 :** SEM images of  $\text{CsPbBr}_3$  films grown under conditions A (a), B (b), and C (c).