

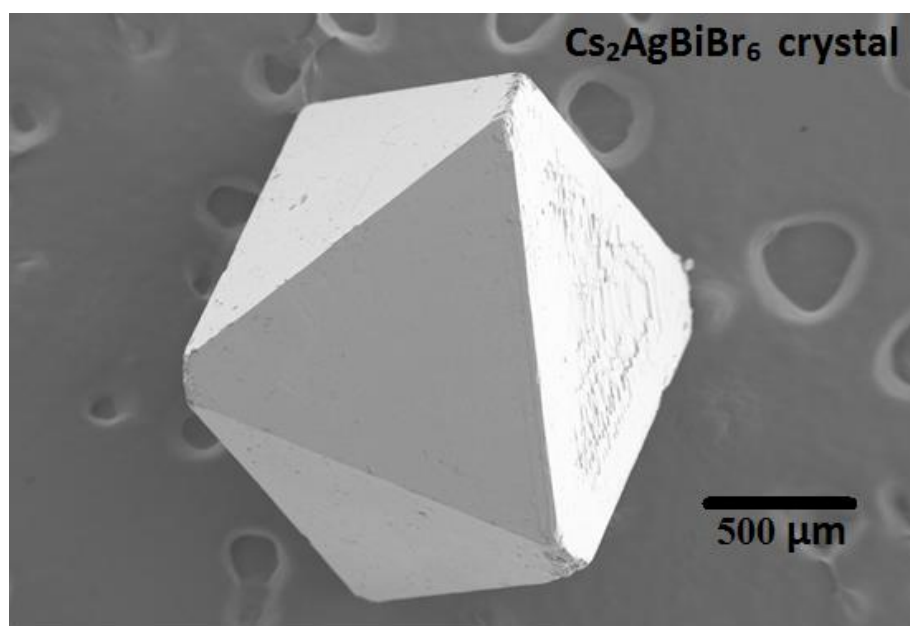
# Supporting Information

## Single-Source Vapor-Deposited Cs<sub>2</sub>AgBiBr<sub>6</sub> Thin Films for Lead-Free Perovskite Solar Cells

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**Fig. S1.** SEM image of Cs<sub>2</sub>AgBiBr<sub>6</sub> crystal with typical octahedral morphology.

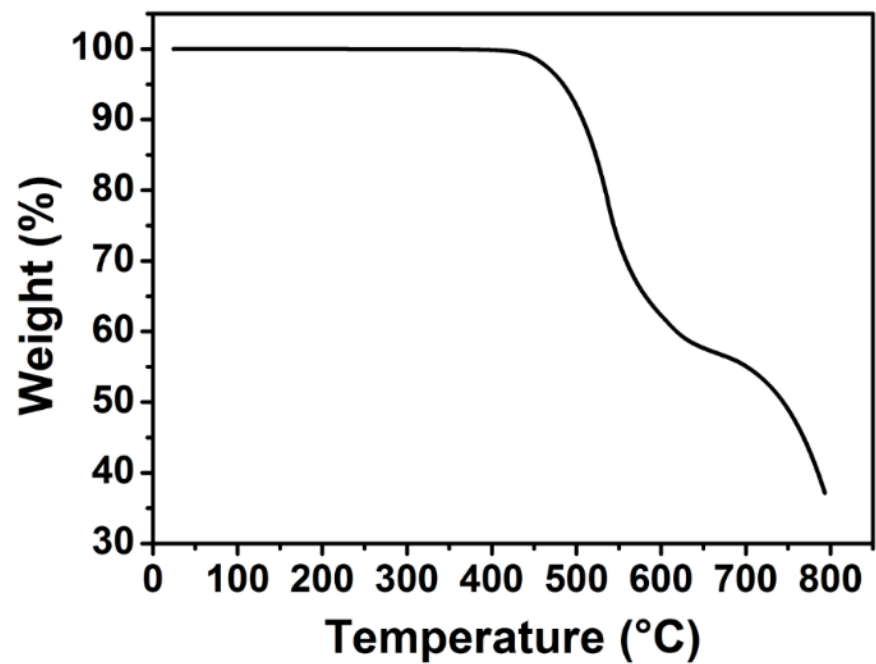


Fig. S2. Thermogravimetric analysis of Cs<sub>2</sub>AgBiBr<sub>6</sub> powder.

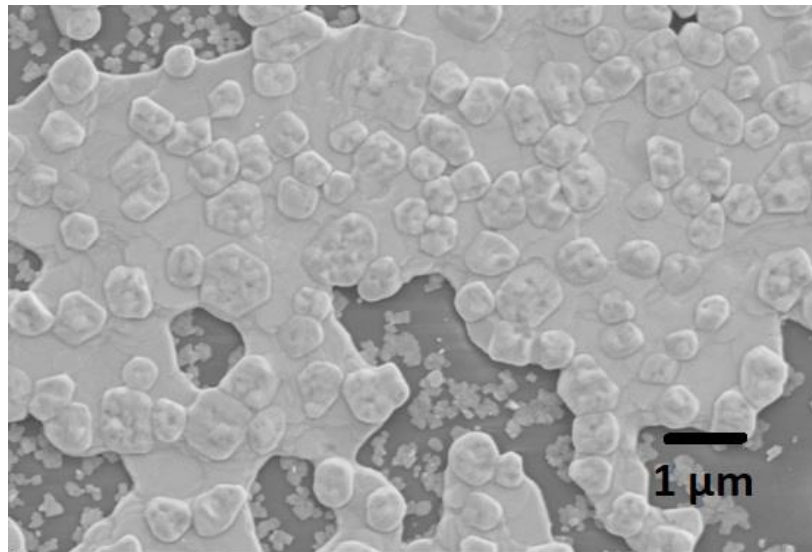


Fig. S3. SEM surface morphology of Cs<sub>2</sub>AgBiBr<sub>6</sub> film thermally annealed at 350 °C for 30 min.

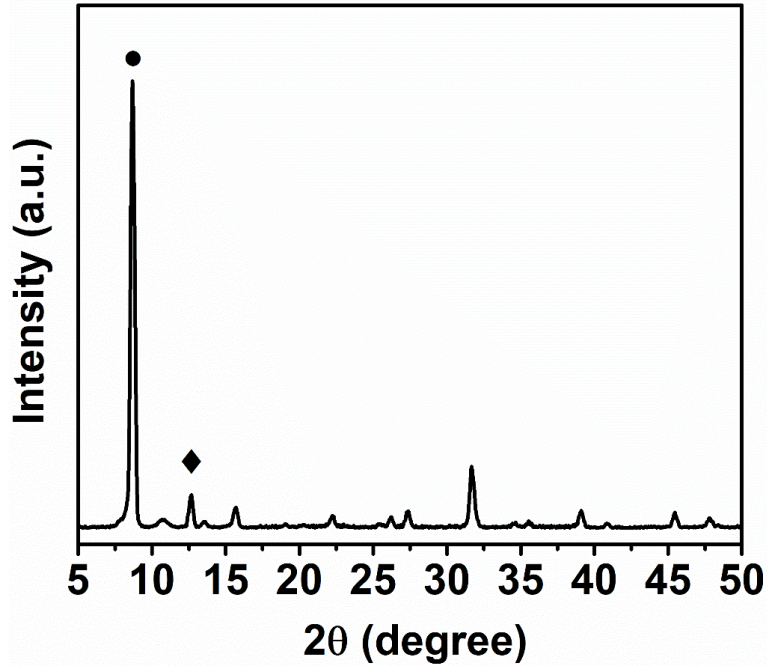


Fig. S4. XRD pattern of  $\text{Cs}_2\text{AgBiBr}_6$  film thermally annealed at  $350\text{ }^\circ\text{C}$  for 30 min. The positions of reflections labeled by circle (●) and diamond (◆) indicate the additional phases of  $\text{CsAgBr}_2$  and  $\text{Cs}_3\text{Bi}_2\text{Br}_9$  respectively.

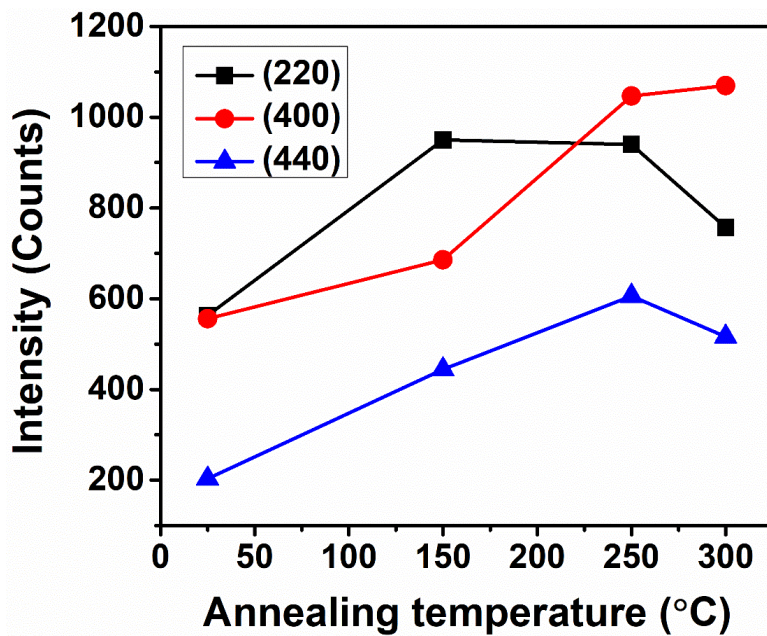


Fig. S5. The diffraction peak intensity of (220), (400) and (440) planes of  $\text{Cs}_2\text{AgBiBr}_6$  films as a function of annealing temperature, respectively.

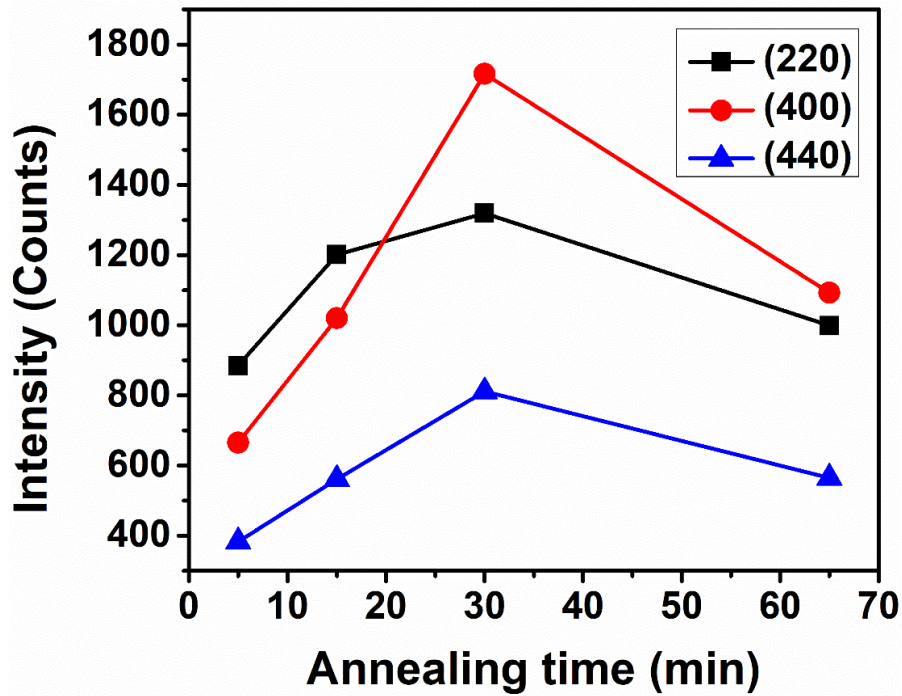


Fig. S6. The diffraction peak intensity of (220), (400) and (440) planes of  $\text{Cs}_2\text{AgBiBr}_6$  films as a function of annealing time, respectively.

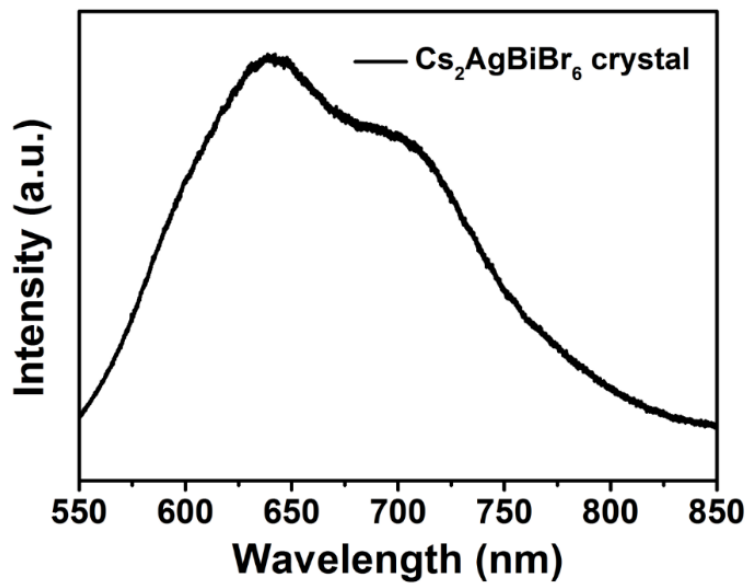
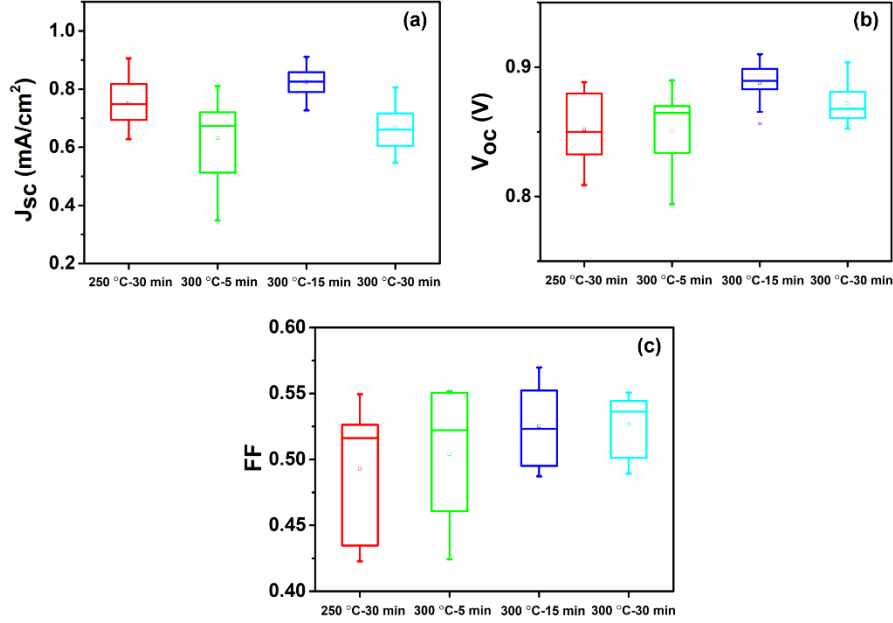


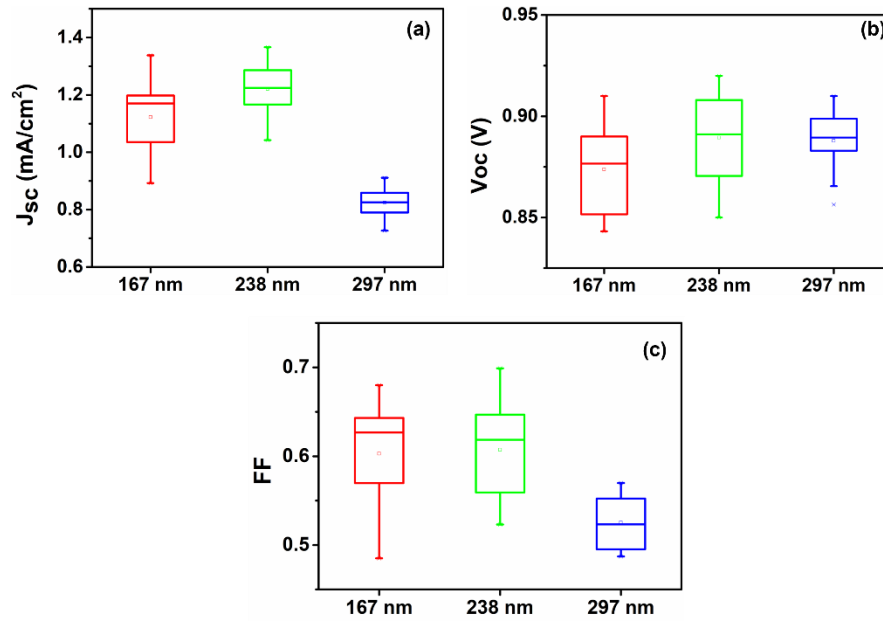
Fig. S7. Steady-state photoluminescence spectrum of  $\text{Cs}_2\text{AgBiBr}_6$  crystal.



**Fig. S8.** The statistical box charts of open-circuit voltage ( $V_{oc}$ ), short-circuit current density ( $J_{sc}$ ) and fill factor (FF) of solar cells assembled with  $Cs_2AgBiBr_6$  films (297 nm) annealed at 250 °C and 300 °C for different times respectively. The values were obtained from 16 individual devices per annealing condition.

**Table S1.** Device performance of  $Cs_2AgBiBr_6$  films with different annealing time and temperatures.

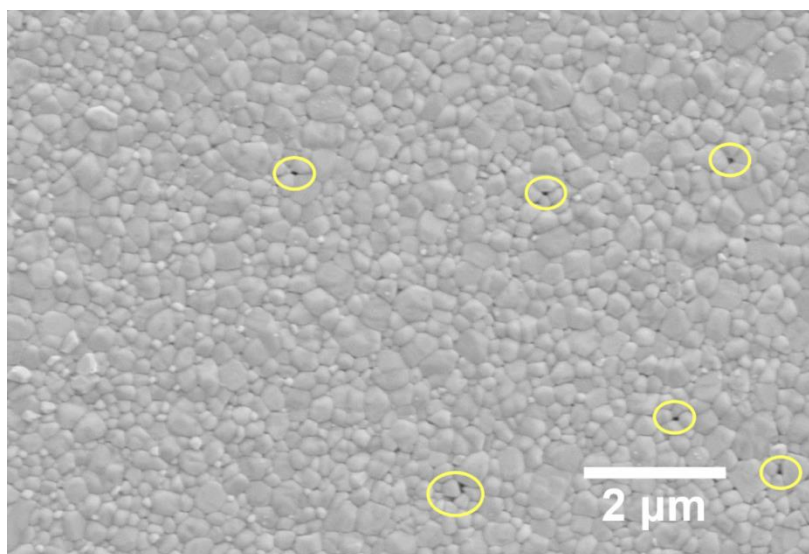
Sample	$J_{sc}$ (mA/cm <sup>2</sup> )	$V_{oc}$ (V)	FF	PCE (%)
$Cs_2AgBiBr_6$ (250 °C-30min)	$0.75 \pm 0.09$	$0.85 \pm 0.03$	$0.60 \pm 0.06$	$0.25 \pm 0.06$
$Cs_2AgBiBr_6$ (300 °C-5min)	$0.63 \pm 0.15$	$0.85 \pm 0.03$	$0.61 \pm 0.05$	$0.17 \pm 0.07$
$Cs_2AgBiBr_6$ (300 °C-15min)	$0.82 \pm 0.05$	$0.89 \pm 0.01$	$0.53 \pm 0.03$	$0.40 \pm 0.03$
$Cs_2AgBiBr_6$ (300 °C-30min)	$0.67 \pm 0.08$	$0.87 \pm 0.02$	$0.53 \pm 0.03$	$0.31 \pm 0.05$



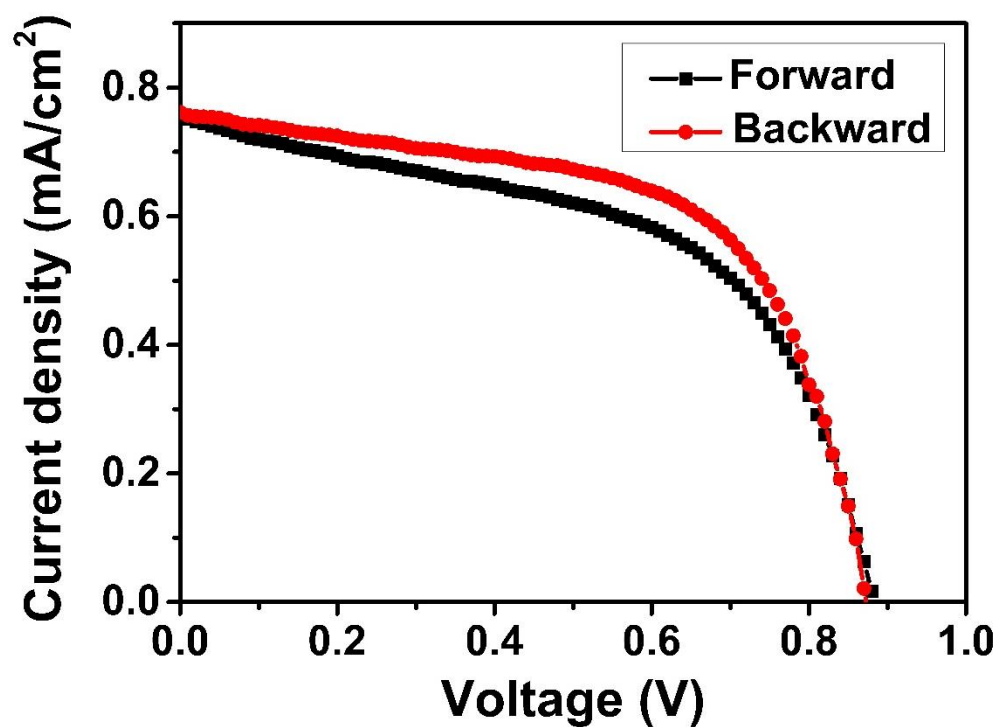
**Fig. S9.** The statistical box charts of open-circuit voltage ( $V_{oc}$ ), short-circuit current density ( $J_{sc}$ ) and fill factor ( $FF$ ) of solar cells based on  $\text{Cs}_2\text{AgBiBr}_6$  films with various thin film thickness. The values were obtained from 16 individual devices per annealing condition.

**Table S2.** Parameters of solar cell devices with different  $\text{Cs}_2\text{AgBiBr}_6$  film thicknesses.

Sample	$J_{sc}$ ( $\text{mA}/\text{cm}^2$ )	$V_{oc}$ (V)	FF	PCE (%)
$\text{Cs}_2\text{AgBiBr}_6$ (167 nm)	$1.12 \pm 0.12$	$0.87 \pm 0.02$	$0.60 \pm 0.06$	$0.53 \pm 0.10$
$\text{Cs}_2\text{AgBiBr}_6$ (238 nm)	$1.22 \pm 0.08$	$0.89 \pm 0.02$	$0.61 \pm 0.05$	$0.60 \pm 0.05$
$\text{Cs}_2\text{AgBiBr}_6$ (297 nm)	$0.82 \pm 0.05$	$0.89 \pm 0.01$	$0.53 \pm 0.03$	$0.40 \pm 0.03$



**Fig. S10.** SEM surface morphology of Cs<sub>2</sub>AgBiBr<sub>6</sub> film annealed at 300 °C for 15 min. The film thickness is approximately 167 nm. The areas marked by yellow circles indicate pinholes in the Cs<sub>2</sub>AgBiBr<sub>6</sub> film.



**Fig. S11.** J-V curves of Cs<sub>2</sub>AgBiBr<sub>6</sub> solar cell, measured by backward scan and forward scan. The Cs<sub>2</sub>AgBiBr<sub>6</sub> film was prepared at 300 °C for 30 min.