Electronic Supplementary Information (ESI)

AAlkylammonium Halides for Phase Regulation and Luminescence Modulation of Cesium Copper Iodide Nanocrystals for Light-Emitting Diodes

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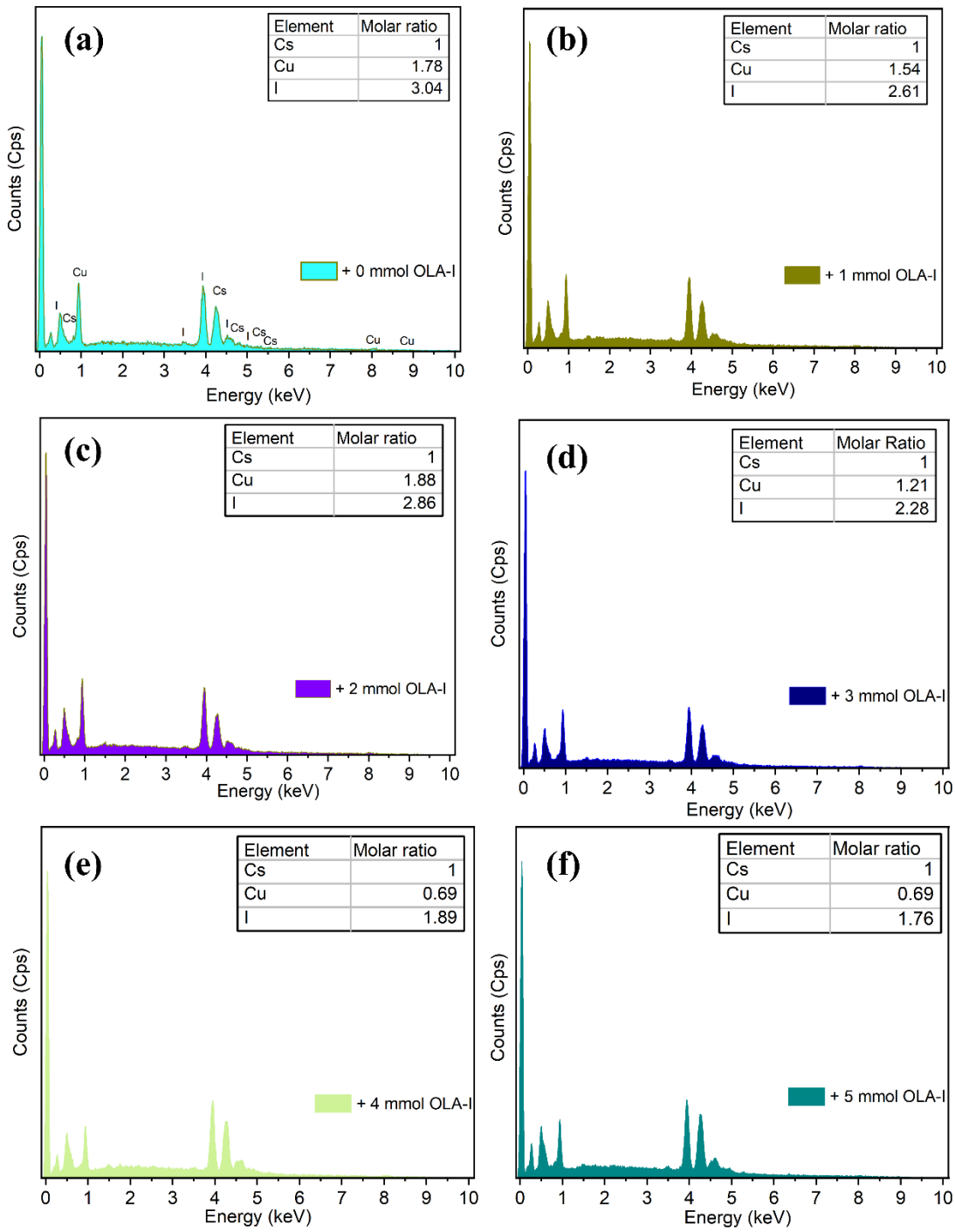

Figure S1. The Energy dispersive spectra (EDS) and corresponding elemental content of compounds obtained with different molar ratio OLA-I added: (a-f) 0 mmol; 1 mmol; 2 mmol; 3 mmol; 4 mmol and 5 mmol OLA-I.

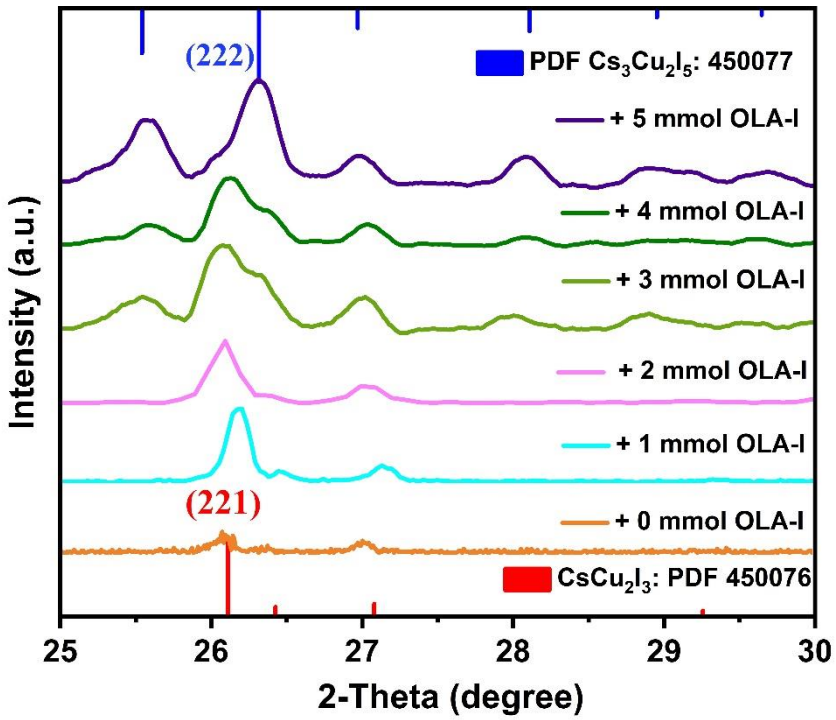

Figure S2. XRD patterns of the samples with enlarged views from 25° to 30°.

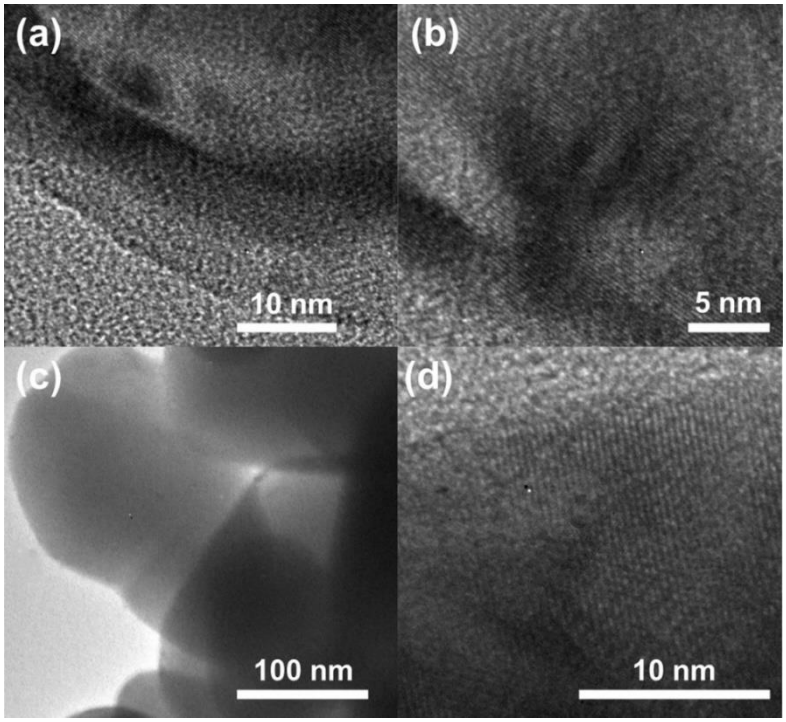

Figure S3. The transmission electron microscope (TEM) images of $CsCu_2I_3$ MRs (a) low-resolution and (b) high-resolution; TEM images of $Cs_3Cu_2I_5$ NCs (c) low-resolution image and (d) high-resolution image.

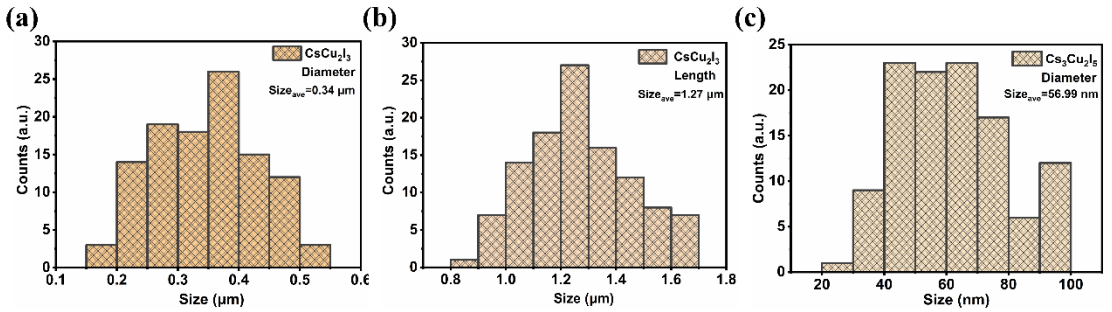

Figure S4. (a-b) The diameter and length distribution statistics of $CsCu_2I_3$ MRs obtained through 1 mmol OLA-I added. (c) The particles size of diameter distribution statistic of $Cs_3Cu_2I_5$ NCs.

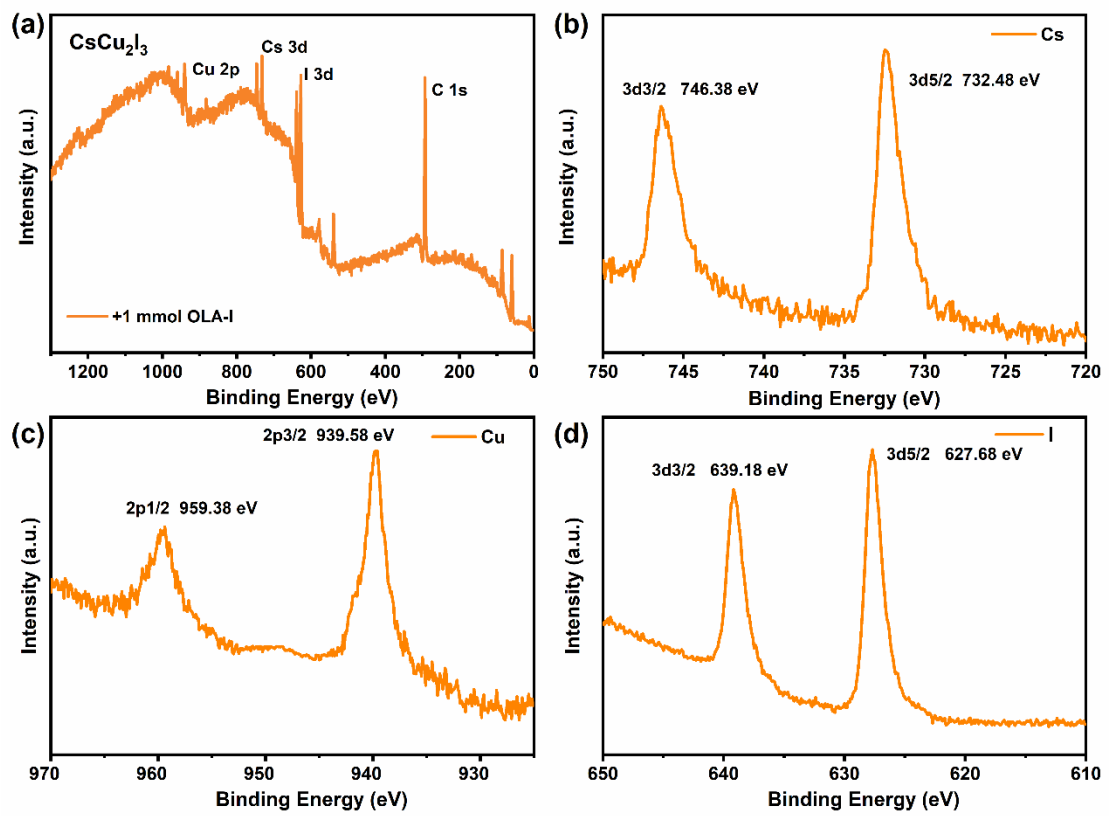

Figure S5. (a) XPS survey spectrum of $CsCu_2I_3$. And the high-resolution spectra corresponding to the curves of (b) Cs 3d, (c) Cu 2p and (d) I 3d orbitals.

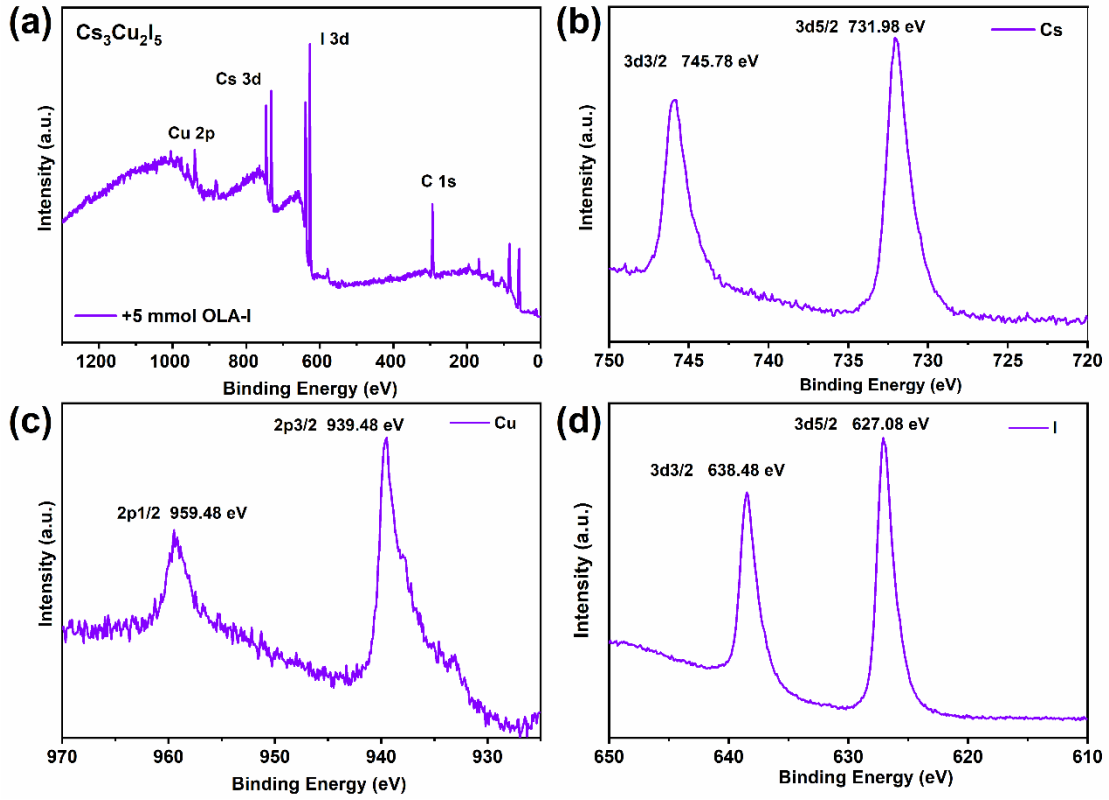

Figure S6. (a) XPS survey spectrum of $Cs_3Cu_2I_5$. And the high-resolution XPS spectra of (b) Cs 3d, (c) Cu 2p and (d) I 3d orbitals.

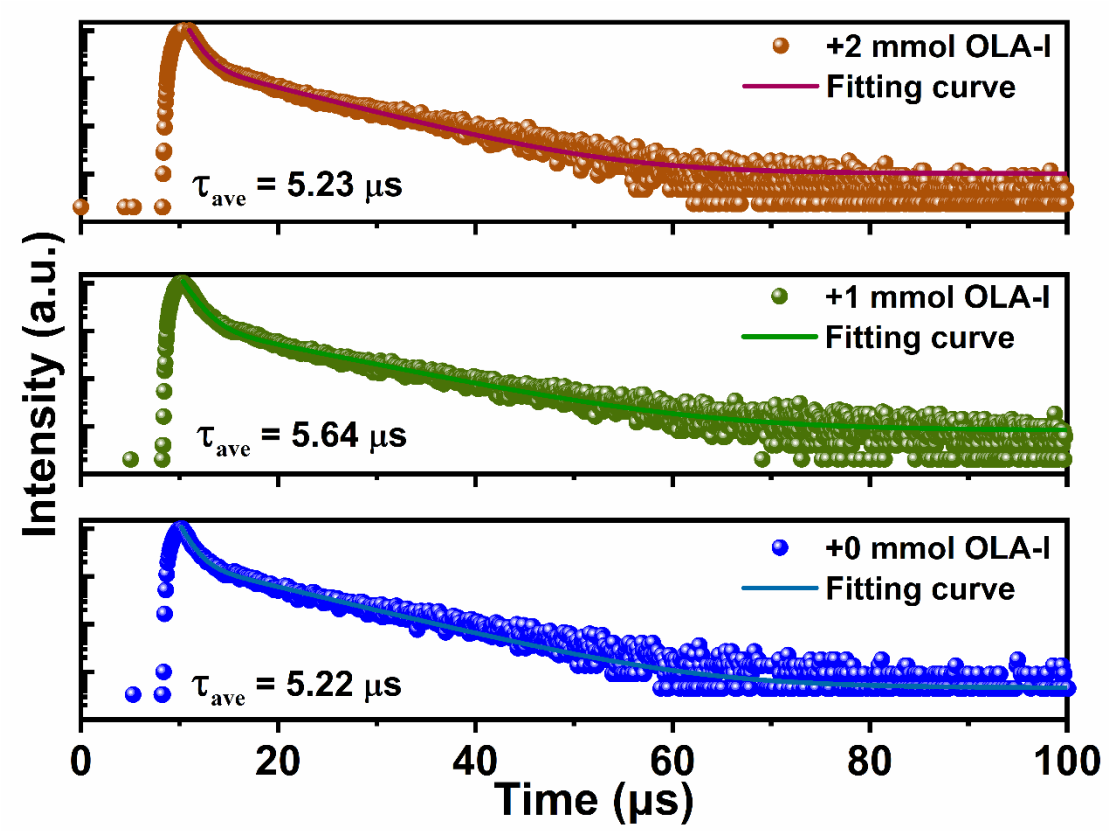

Figure S7. Time-resolved PL decay curves at room temperature for $CsCu_2I_3$ (adding 0 mmol, 1 mmol and 2 mmol OLA-I) compound and solid lines represent the fitting curves by a double exponential function.

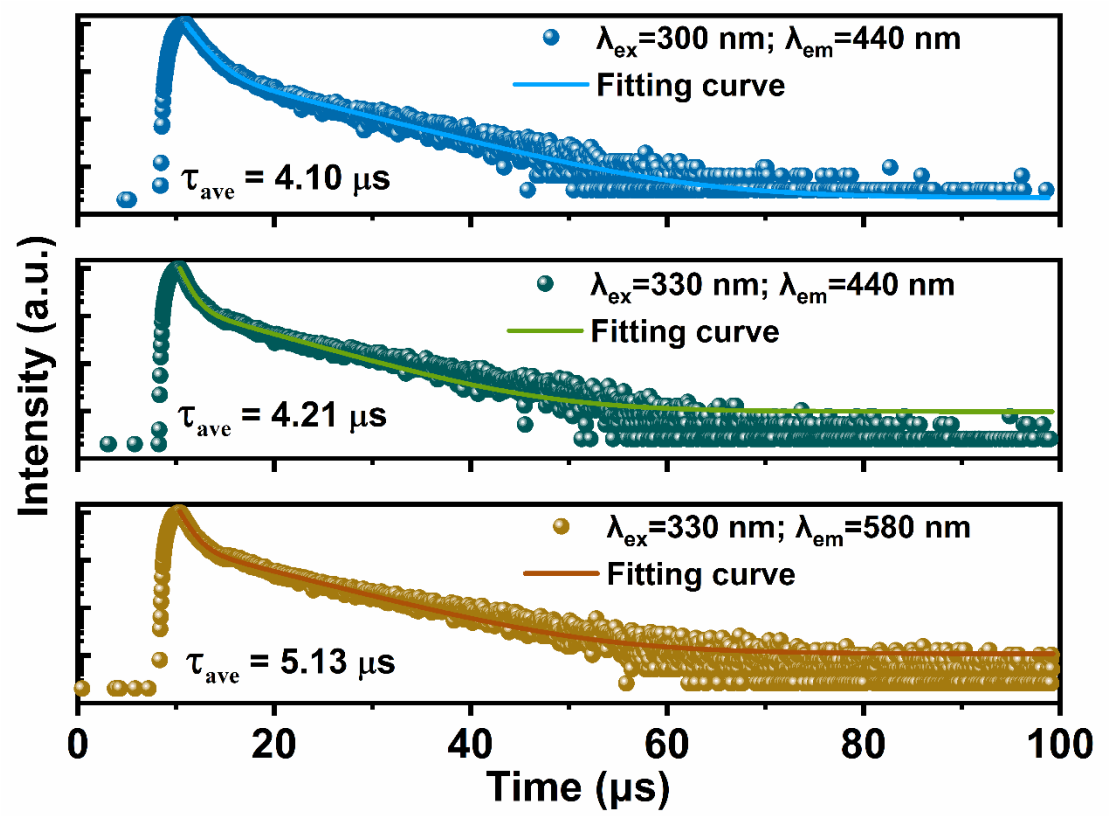

Figure S8. Time-resolved PL decay curves at room temperature for $CsCu_2I_3$/$Cs_3Cu_2I_5$ (adding 4 mmol OLA-I) compound and solid lines represent the fitting curves by a double exponential function.

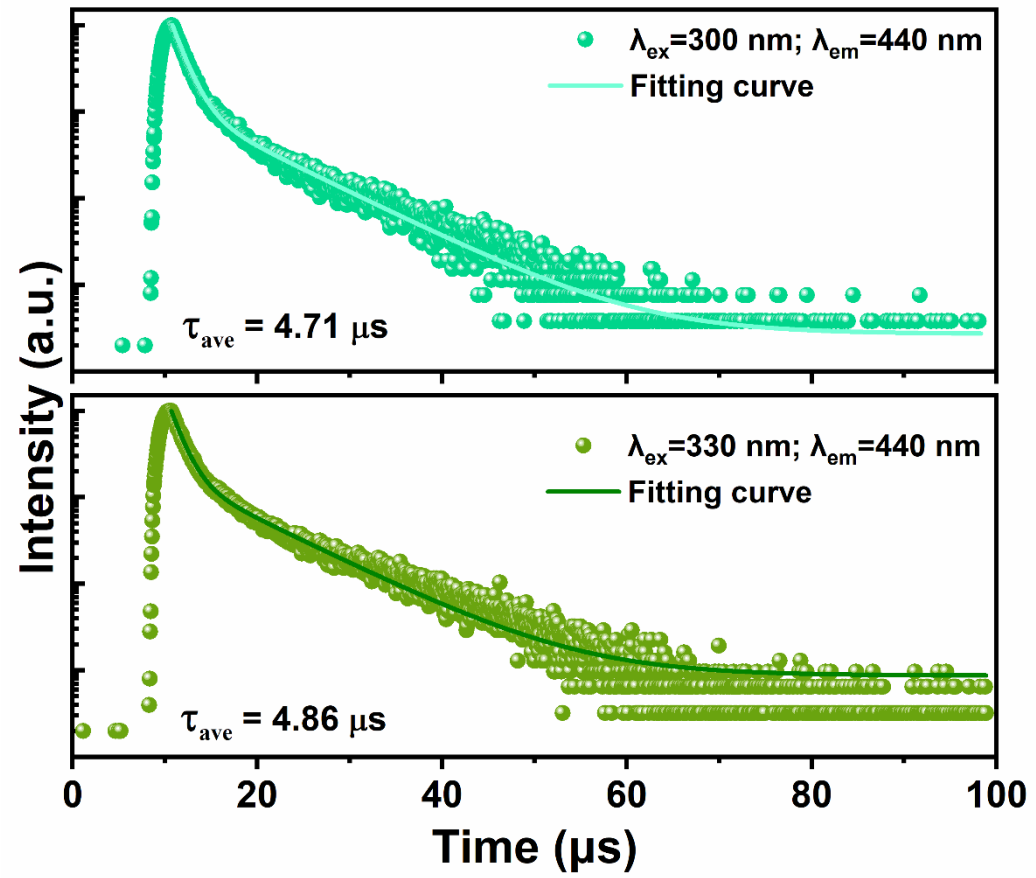

Figure S9. Time-resolved PL decay curves at room temperature for $Cs_3Cu_2I_5$ (adding 5 mmol OLA-I) compound and solid lines represent the fitting curves by a double exponential function.

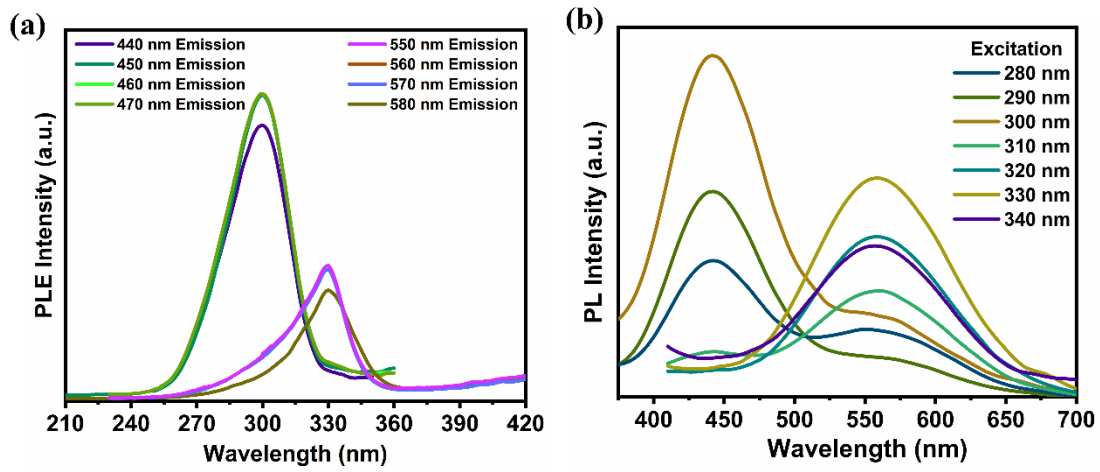

Figure S10. (a) PLE spectra of $CsCu_2I_3/Cs_3Cu_2I_5$ compound for different emission wavelengths. (b) PL spectra of $CsCu_2I_3/Cs_3Cu_2I_5$ compound upon UV irradiation with different excitation wavelengths.

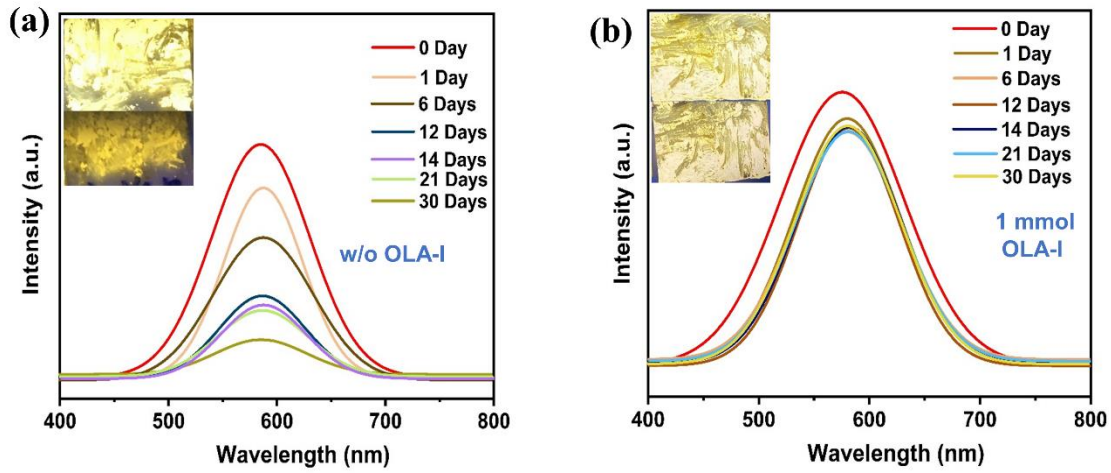

Figure S11. (a) Evolution of the PL spectra of 0 mmol OLA-I treated $CsCu_2I_3$ under 85 ℃ over 30 days for stability investigation. (b) Evolution of the PL spectra of 1 mmol OLA-I treated $CsCu_2I_3$ over 30 days for stability investigation (Inserted images representing the luminance changing under 85 ℃ environment. Above: for 0 day; Down: for 30 days).

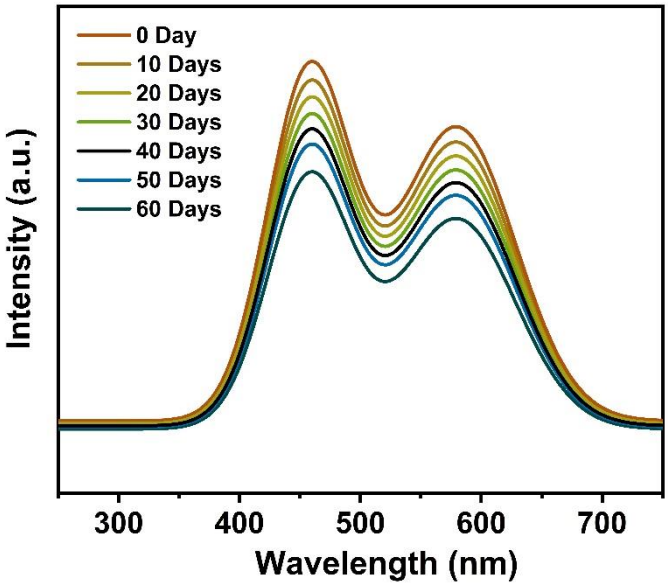

Figure S12. PL emission spectrum of the obtained compound with adding 3 mmol OLA-I exposed to air for 60 days.

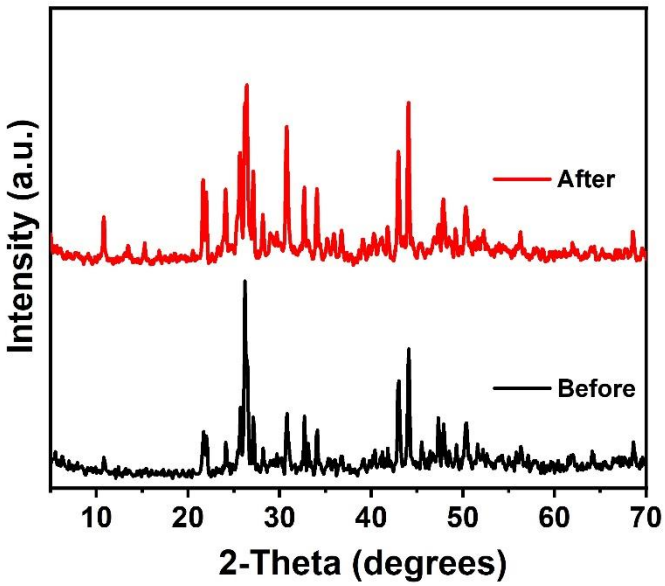

Figure S13. XRD patterns of 3 mmol OLA-I treated $CsCu_2I_3/Cs_3Cu_2I_5$ component before and after storage under the ambient condition for two months.

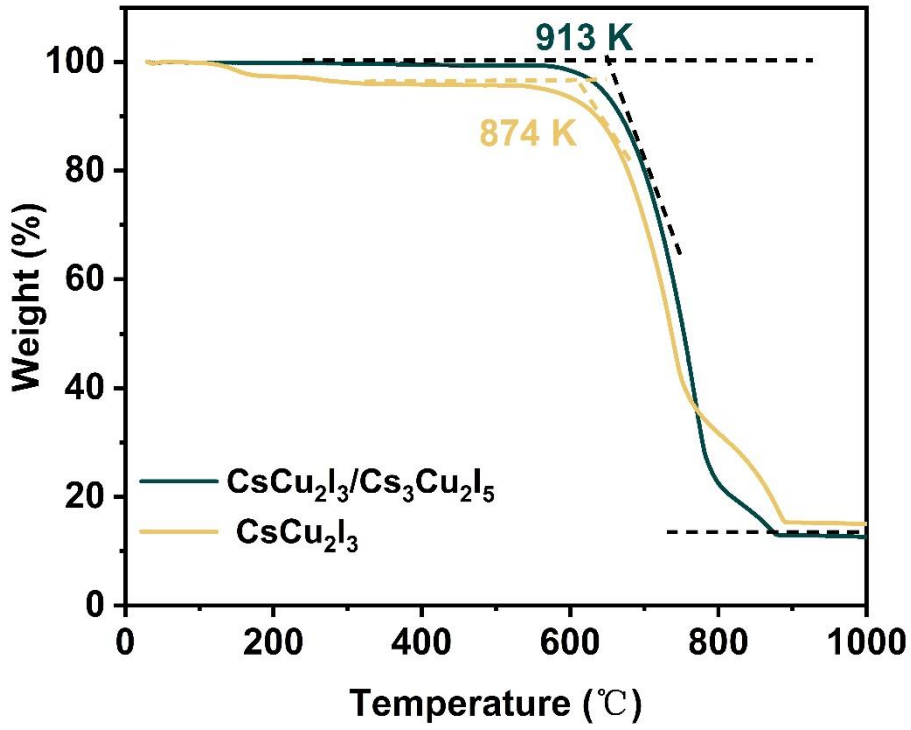

Figure S14. Thermogravimetric analysis (TGA) of $CsCu_2I_3$ and $CsCu_2I_3/Cs_3Cu_2I_5$.

Table S1. Element analysis measurement results and the calculated ratios of Cs, Cu and I for prepared samples with adding different molar ratio of OLA-I (Ratio of inputting amounts for Cs: Cu: I=3.2: 4: 4).

+ x mmol OLA-I	Cs (%)	Cu (%)	I (%)	Ratio	Component
+ 0 mmol	13.8	24.6	41.9	1:1.78:3.04	**$CsCu_2I_3$**
+ 1 mmol	14.2	21.8	36.9	1:1.54:2.61	**$CsCu_2I_3$**
+ 2 mmol	12.5	23.5	35.7	1:1.88:2.86	**$CsCu_2I_3$**
+ 3 mmol	15.7	19.0	35.8	1:1.21:2.28	**$CsCu_2I_3$** **$Cs_3Cu_2I_5$**
+ 4 mmol	22.1	15.3	41.7	1:0.69:1.89	**$CsCu_2I_3$** **$Cs_3Cu_2I_5$**
+ 5 mmol	20.2	13.9	35.6	1:0.69:1.76	**$Cs_3Cu_2I_5$**

Table S2. Summary of the synthetic strategies and optical parameters of recent reported copper-based halide compounds.

Formula	Morphology	Method	Emission peak (nm)	PLQY (%)	Ref.
$CsCu_2I_3$	NA	Solid-state reaction	576	3.23	[1]
$CsCu_2I_3$	Thin films	Spin-coating	~ 548	20.6	[2]
$CsCu_2I_3$	Micro-rods	Antisolvent infiltration	575	12.1	[3]
$CsCu_2I_3$	Nanorods	Hot injection	553	5	[4]
$CsCu_2I_3$	Nanorods	Hot injection	561	11	[5]
$CsCu_2I_3$	Wires	Antisolvent	570	NA	[6]
$Cs_3Cu_2I_5$	Nanocrystals	Hot injection	441	67	[4]
$Cs_3Cu_2I_5$	Nanocrystals	Modified hot injection	445	73.7	[7]
$Cs_3Cu_2I_5$	Powder	Solution stirring	440	NA	[8]
$Cs_3Cu_2I_5$: Mn	Microparticles	Solid-state reaction	448 and 556	57	[9]
$Cs_3Cu_2I_5$	Nanocrystals	Hot injection	441	67	[10]
$Cs_5Cu_3Cl_6I_2$	Powder	Solid-state reaction	462	95	[11]
$CsCu_2I_3$	**Micro-rods**	**Hot injection**	**560**	**47.3**	**This work**
$CsCu_2I_3$/ $Cs_3Cu_2I_5$	**Micro-rods and Nanocrystals**	**Hot injection**	**560 and 440**	**66.4**	**This work**
$Cs_3Cu_2I_5$	**Nanocrystals**	**Hot injection**	**440**	**95.3**	**This work**

Additional References

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