

Supporting Information

Highly Efficient Luminescent and Color-tunable White Emission from 0D $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ Perovskites via Al^{3+} and Sb^{3+} Codoping

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1. Experimental Materials.

Caesium chloride (CsCl, Aladdin, 99.99%), Indium oxide (In_2O_3 , Alfa Aesar, 99.99%), Antimony oxide (Sb_2O_3 , Macklin, 99.9%), Aluminum acetate ($\text{C}_6\text{H}_9\text{AlO}_6$, Aladdin, 99%), Hydrochloric acid (HCl, 36–38wt.% in water, Beijing Chemical Works), and ethanol (AR, Beijing Tongguang Fine Chemical Co., Ltd., China) were purchased and used as received without further purification.

2. Experimental Synthesis.

2.1 Synthesis of $\text{Sb}^{3+}/\text{Al}^{3+}$ Co-Doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$:

$\text{Sb}^{3+}/\text{Al}^{3+}$ Co-Doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ chalcogenides were synthesized by solvothermal reaction. Typically, 2 mmol of CsCl, 0.5 mmol of In_2O_3 and 0.05 mmol of Sb_2O_3 and y mmol of $\text{C}_6\text{H}_9\text{AlO}_6$ ($y=0.03, 0.05, 0.1, 0.15, 0.2, 0.3, 0.4$) were placed into a 25mL polytetrafluoroethylene autoclave, and 5 ml HCl was added, followed by lining into a stainless-steel Parr autoclave, and heat the reaction in an oven at 180 °C for 12 hours. After the heating reaction is over, it is naturally cooled to room temperature. Then the prepared perovskite powder was filtered out, washed with absolute ethanol three times, and dried in a vacuum drying oven at 60°C for 8 hours. Finally, the synthesized samples were obtained in different doses.

2.2 Synthesis of Sb^{3+} Doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ and Al^{3+} Doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$:

For synthesizing Sb^{3+} doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$, only x mmol Sb^{3+} ($x=0, 0.005, 0.01, 0.05, 0.1, 0.2, 0.3$) was used under otherwise identical conditions. To synthesize Al^{3+} doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$, only y mmol $\text{C}_6\text{H}_9\text{AlO}_6$ ($y=0, 0.03, 0.05, 0.1, 0.15, 0.2, 0.3, 0.4$).

2.3 Fabrication of WLED device:

The crystal sample of Sb^{3+} - Al^{3+} co-doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ was poured into a mortar and fully ground, then ground into a fine powder and transferred to a centrifugal tube for preservation. Take out a slide, pour a small amount of ground perovskite powder onto the slide, dip a certain proportion of UV-curable glue with the tip of the pipette gun, and evenly stir the perovskite powder to make it fully mixed with UV-curable glue. Then, the perovskite powder mixed with UV-curable adhesive is coated on the 365 nm LED chip and 310 nm-chip, and after coating, the ultraviolet lamp is irradiated for 3-5 minutes to solidify the UV-curable adhesive, and then the packaged calcium tablet is packaged and characterized accordingly. There is no need to mix phosphors.

3. Material characterization.

SEM-EDS Measurements: Sigma 500 field emission scanning electron microscope, and the proportion of elements was characterized by energy dispersive X-ray spectroscopy from Oxford instruments.

Powder X-ray Diffraction (XRD): The crystallographic phase structure was determined by X-ray diffraction (XRD, Bruker D8 Discover) The powder X-ray diffraction pattern was measured using an X-ray diffractometer (model: SMARTLAB 3 KW) manufactured by Rigaku Corporation in the 2θ range of 5° - 70° .

X-ray photoelectron spectroscopy (XPS): The chemical state and element content were identified by X-ray, and samples was measured by via SHIMADZU AXIS Ultra DLD. instrument.

Absorbance: UV–visible absorption spectra were collected by Lambda 750 spectrophotometer (PerkinElmer Instruments) at a range of 240 to 830nm.

Photoluminescence Measurements: The photoluminescence (PL) and PL excitation (PLE) spectrum were carried out with the HORIBA multifunctional fluorescence spectrometer test system to collect room temperature of the samples.

Photoluminescence quantum yield Measurements: The photoluminescence quantum yield (PLQY) was measured by HORIBA multifunctional fluorescence spectrometer test system, and the data were collected by placing the powder on a glass slide inside the integrating sphere.

Lifetime Measurements: For emission lifetime measurement, the pulsed excitation light (338 nm) was generated by a μ F2 60W xenon flash lamp. The detector signal was set below a threshold of 3000 counts per second. The lifetime of emission was determined by reconvolution fit with the instrument response function using the Edinburgh F1000 software. In all cases, emission decay was satisfactorily fitted with a multiple-exponential function until reconvolution .

Temperature dependent emission spectra were also recorded by HORIBA Fluorolog-QM instrument with liquid nitrogen used for temperature reduction.

Raman Measurements: The DXR Raman Microscope (Thermo Fisher Scientific) and a fast and confocal Raman imaging system (WITec alpha 300R) was used to characterize the Raman spectrum of the sample.

Powerdependent PL spectra were collected by Alpha 300R spectrometer.

4. Computational Methods.

All calculations using density functional theory (DFT) were carried out using the Vienna Ab initio Simulation Package (VASP).¹ The generalized gradient approximation of the Perdew–Burke–Ernzerhof (PBE) parameterization with the projector-augmented wave method was performed for the exchange and correlation functional.²⁻³ The kinetic-energy cutoff of 400 eV and a $2 \times 5 \times 7$ Monkhorst–Pack k-mesh for the wavefunction basis set is employed.

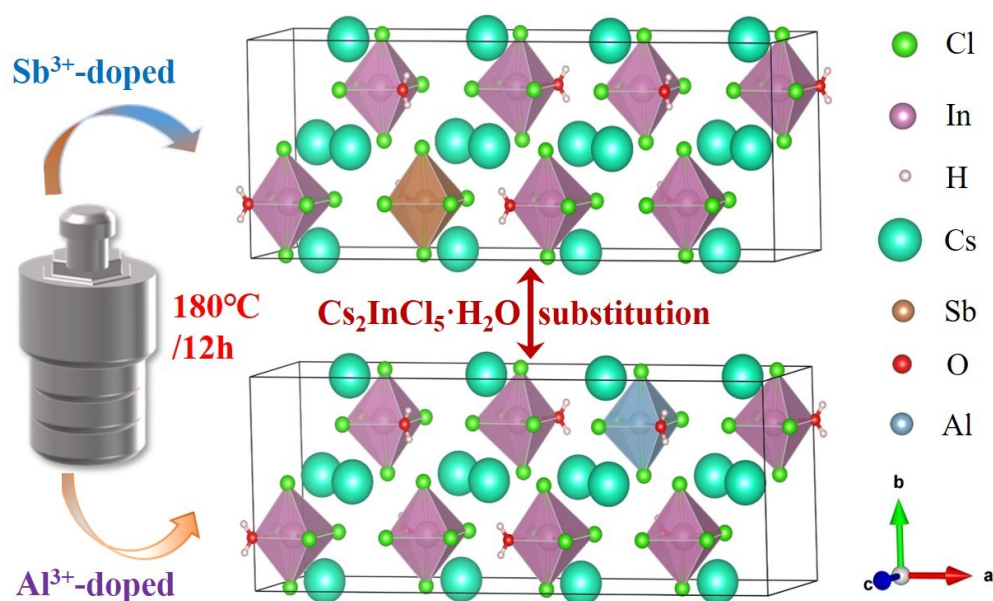


Fig. S1 Schematic representation of individually Sb³⁺ and Al³⁺ doped Cs₂InCl₅·H₂O synthesized using hydrothermal method.

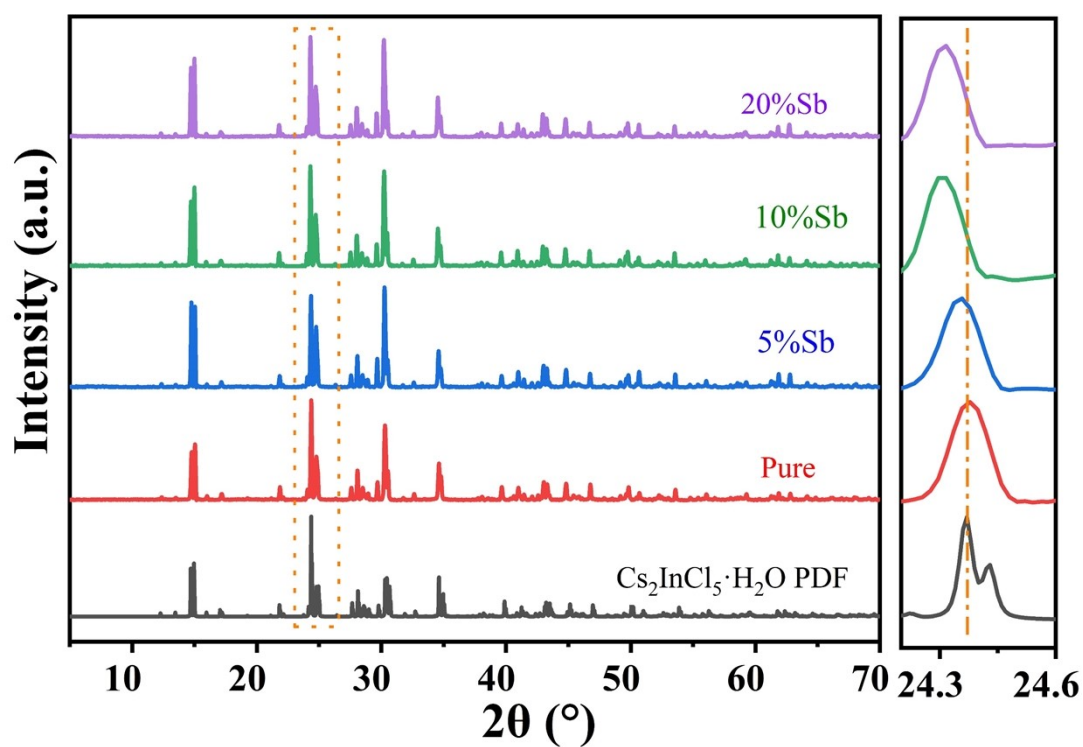


Fig. S2 XRD patterns of xSb³⁺ doped Cs₂InCl₅·H₂O with different Sb³⁺ doping concentrations. (x=0,0.05,0.1,0.2) The XRD pattern is shifted around 23.4°.

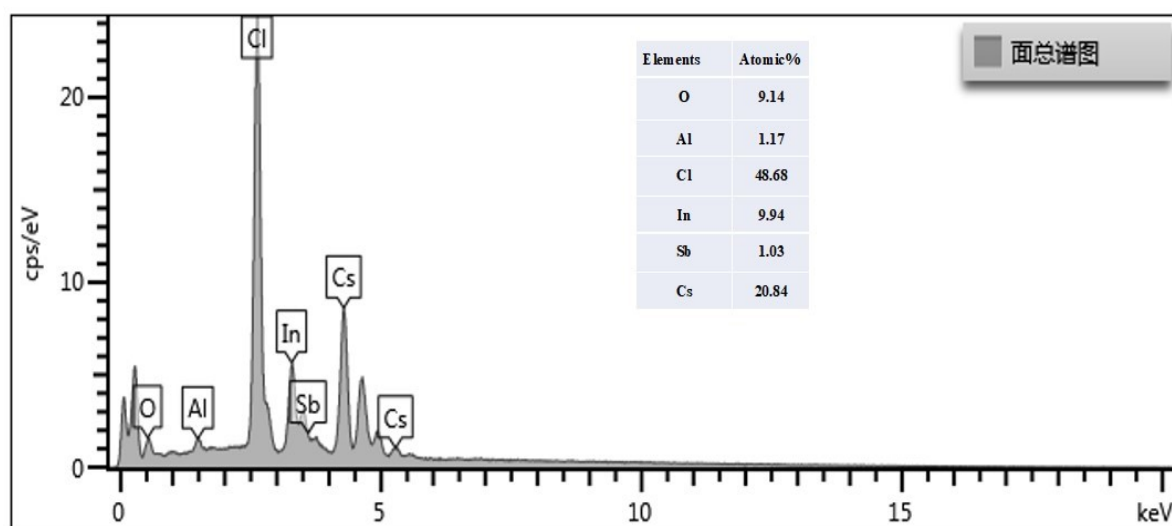


Fig. S3 The corresponding EDS spectrum of $x\text{Sb}^{3+}$ - $y\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ ($x=0.1$, $y=0.1$).

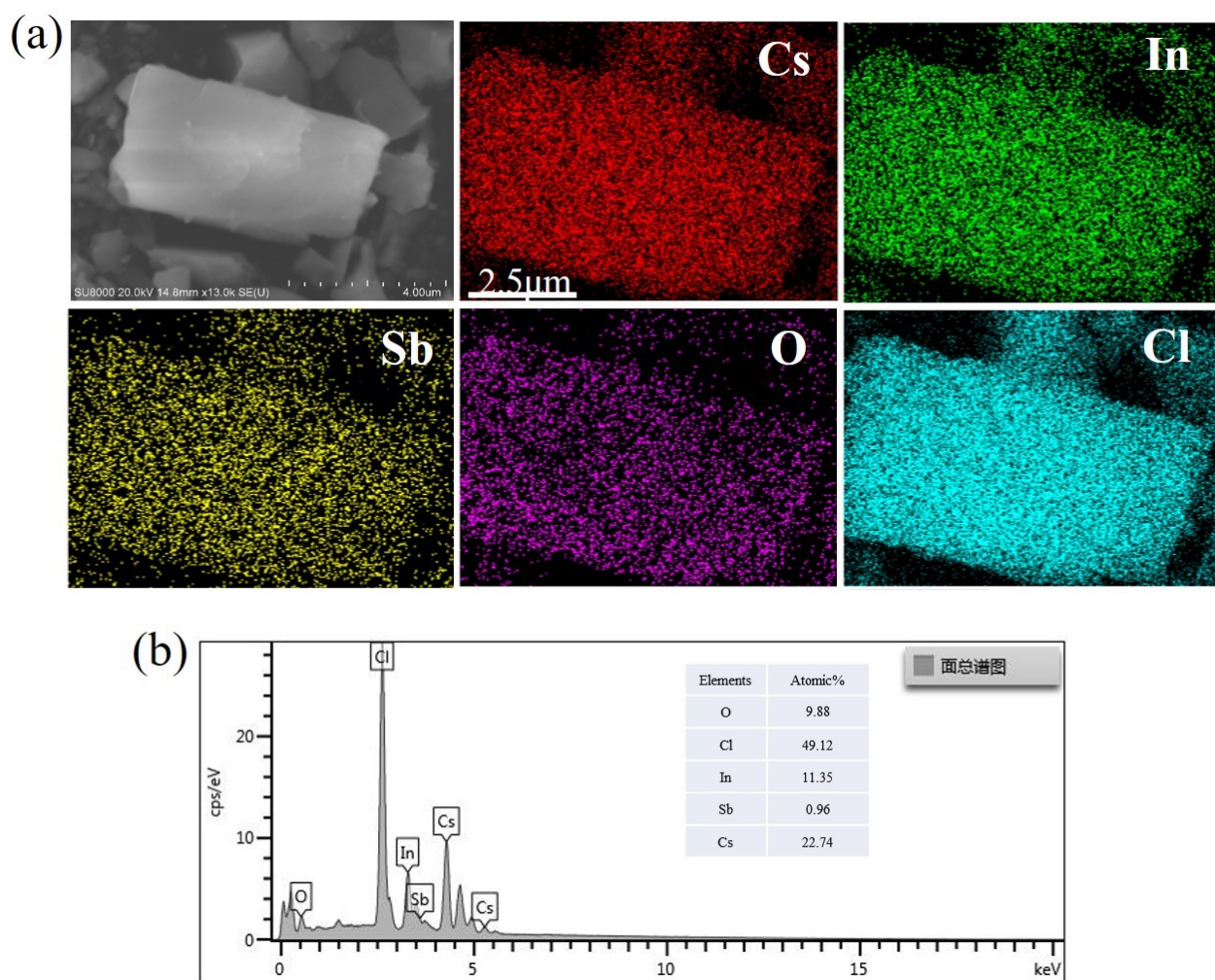


Fig. S4. (a) SEM image of $x\text{Sb}^{3+}$ doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ ($x=0.1$) and EDS element diagram of Cs, In, Sb, O and Cl. (b) The corresponding EDS spectrum.

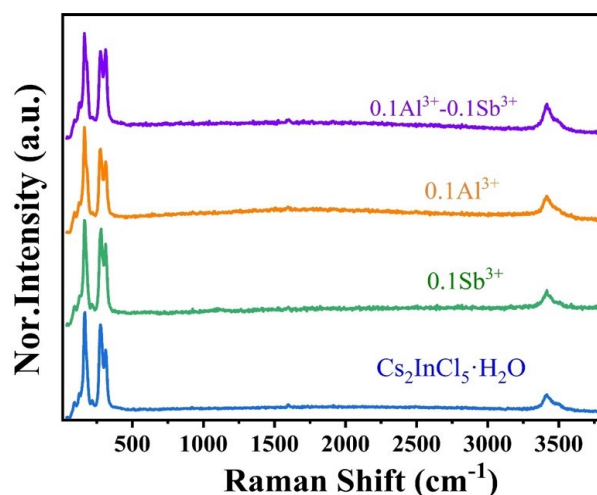


Fig. S5 Normalized Raman spectra of different doping.

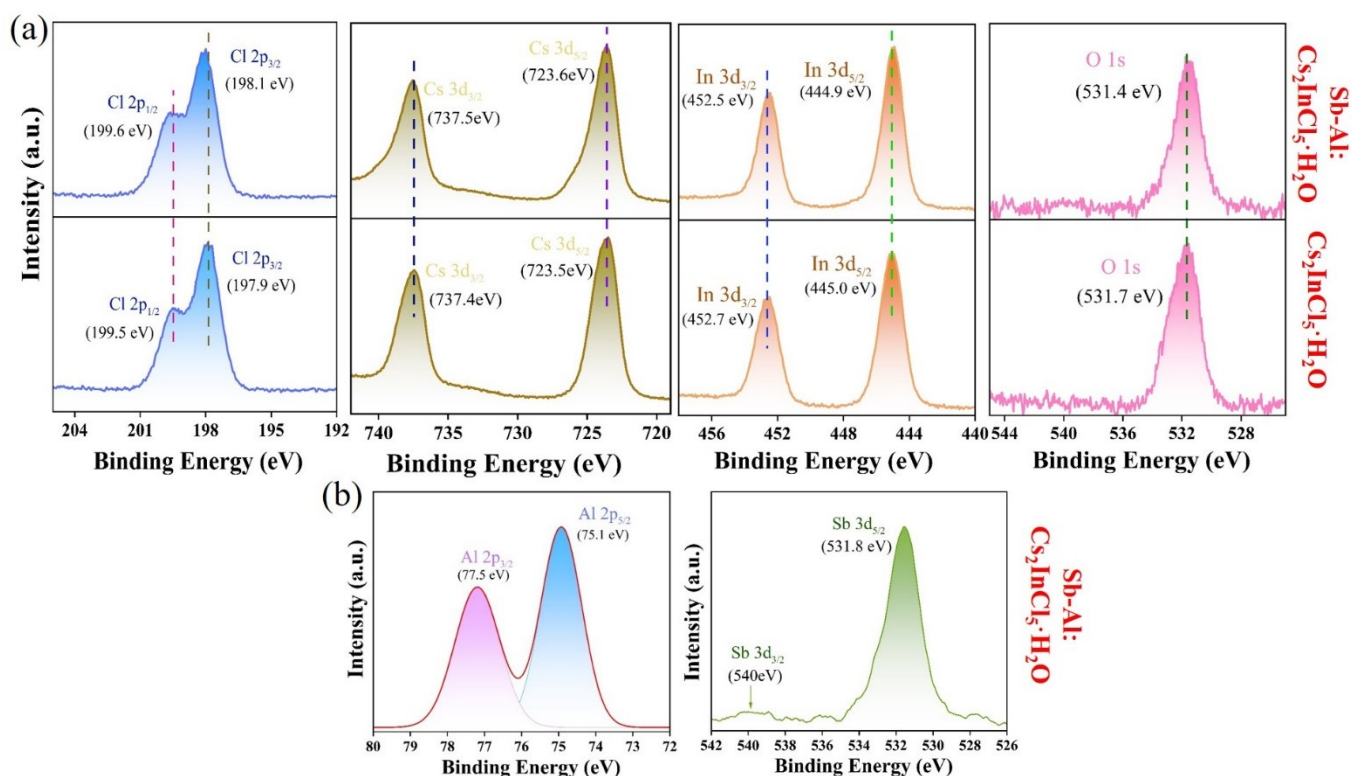


Fig. S6 (a) Comparison of XPS spectra of individual elements of $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ and $0.1\text{Sb}^{3+}-0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$. (b) Characteristic peaks of Al and Sb in $0.1\text{Sb}^{3+}-0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$.

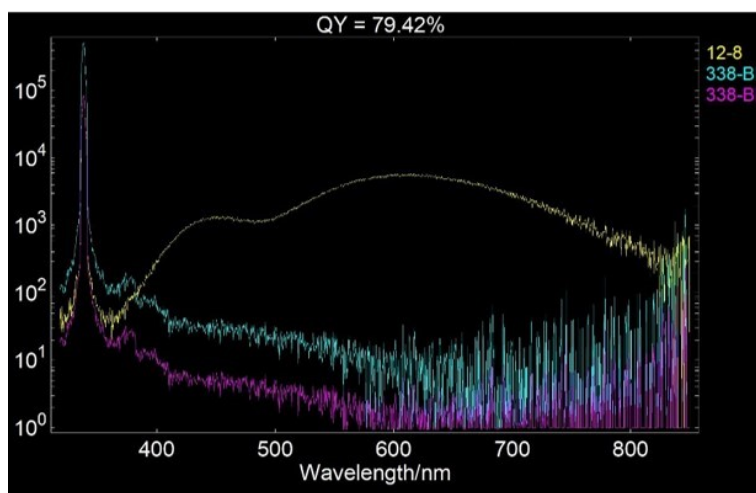


Fig. S7 The PLQY measurement cross-sectional view of $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ sample (the yellow curve) shows that the PLQY is as high as 79.42%. The excitation wavelength was 338 nm.

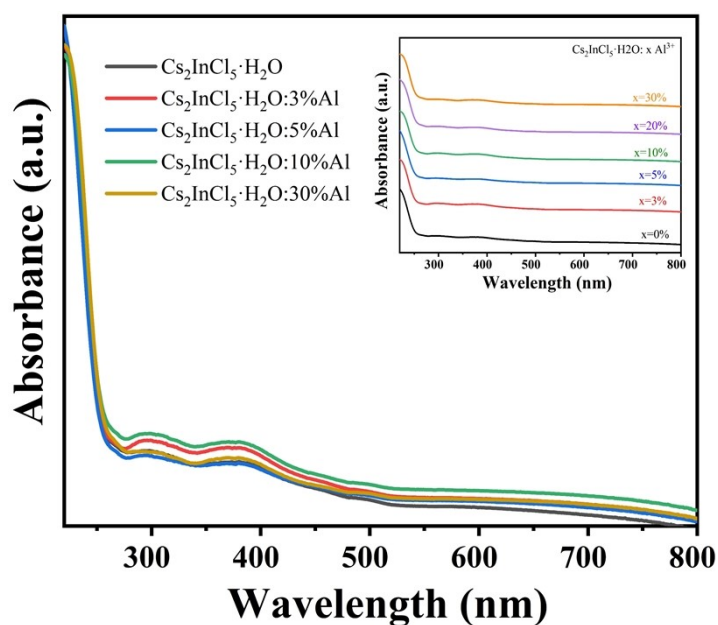


Fig. S8 Absorption spectra of Al^{3+} doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ with different Al^{3+} doping concentrations. The inset is an absorption stacking diagram showing that Al^{3+} doping does not change the structure of the absorption spectrum.

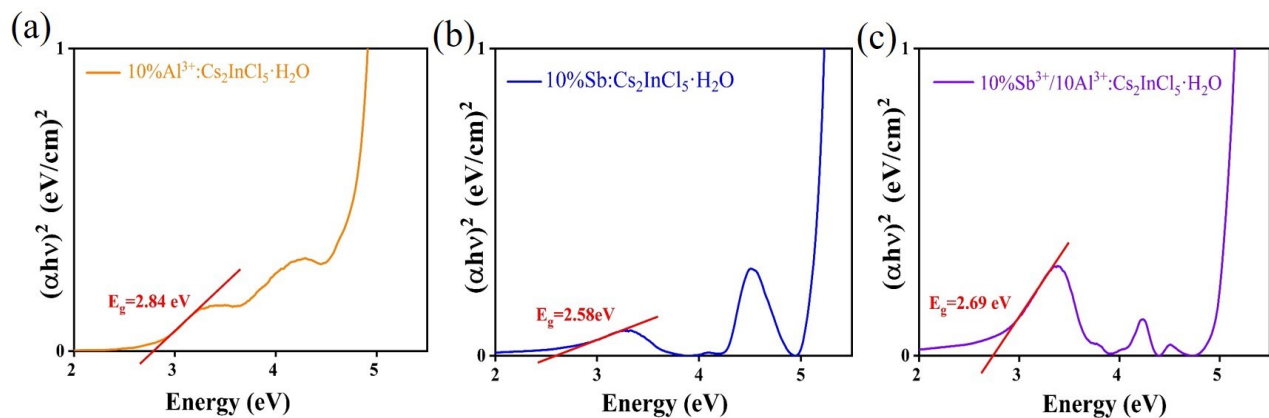


Fig. S9 Tauc-plot of the UV–Vis absorption spectrum of (a) 0.1Al³⁺ doped Cs₂InCl₅·H₂O; (b) 0.1Sb³⁺ doped Cs₂InCl₅·H₂O; (c) 0.1Sb³⁺-yAl³⁺ co-doped Cs₂InCl₅·H₂O.

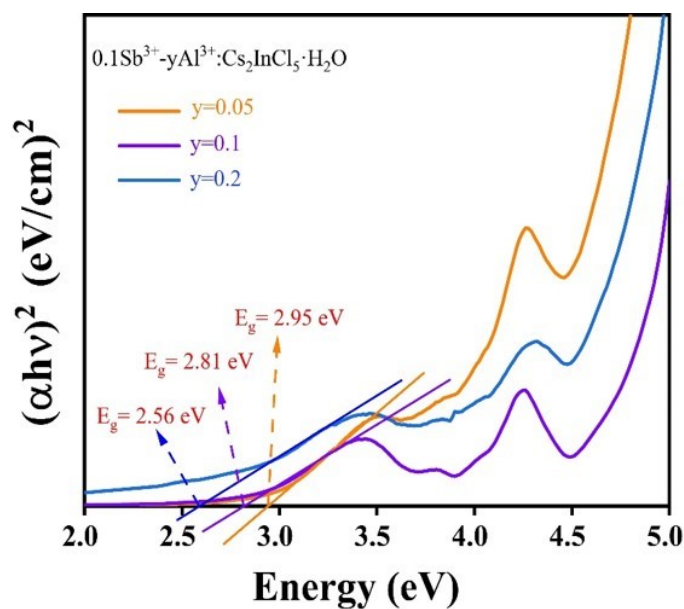


Fig. S10 Tauc-plot of the UV–vis absorption spectrum of 0.1Sb³⁺-yAl³⁺ co-doped Cs₂InCl₅·H₂O. (y=0.05,0.1,0.2). The band gap gradually decreases with increasing Al³⁺ doping concentration.

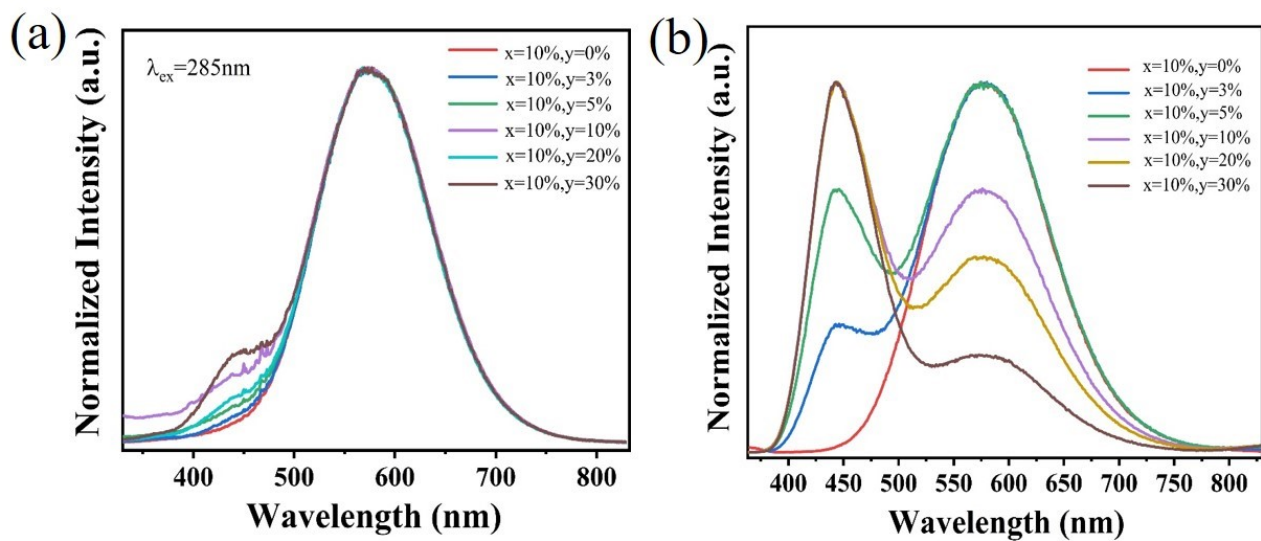


Fig. S11 Normalized emission spectra of $0.1\text{Sb}^{3+}\text{-yAl}^{3+}\text{: Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ at excitation wavelengths of (a) 283 nm and (b) 338 nm. ($y=0,0.03,0.05,0.1,0.2,0.3$).

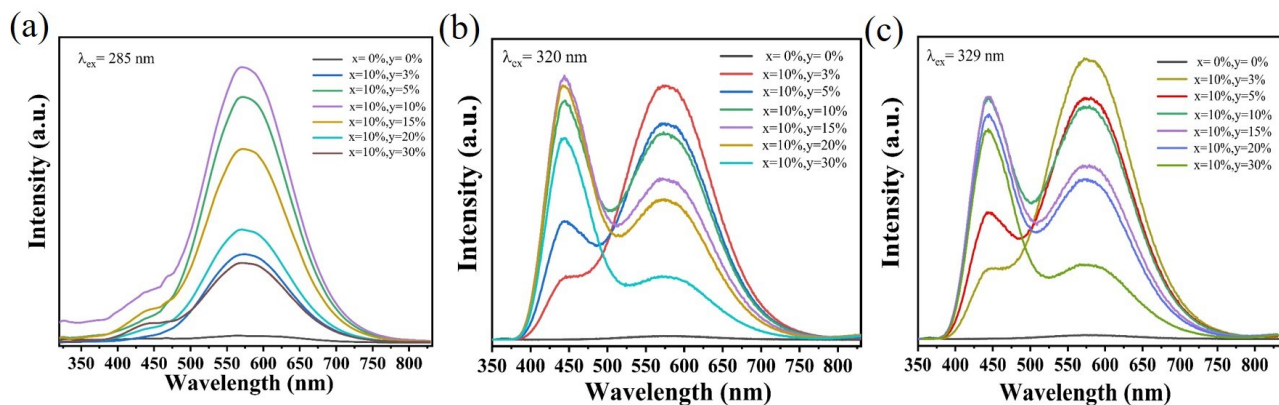


Fig. S12 Emission spectra of $x\text{Sb}^{3+}\text{-yAl}^{3+}\text{: Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ at excitation wavelengths of (a) 285 nm; (b) 320 nm and (c) 329 nm. ($x=0,0.1$; $y=0,0.03,0.05,0.1,0.15,0.2,0.3$).

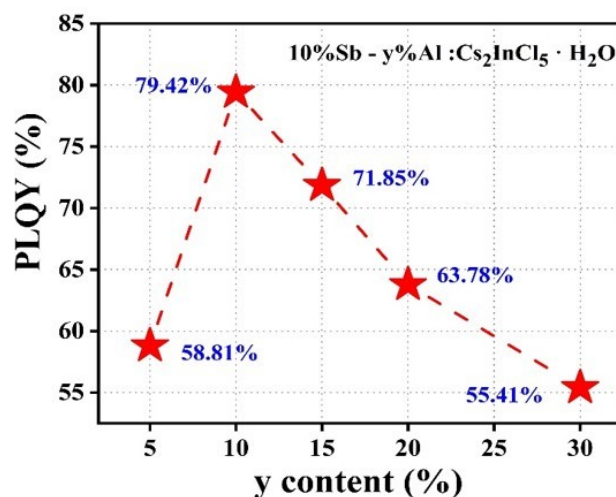


Fig. S13 Corresponding PLQY values for $0.1\text{Sb}^{3+}\text{-yAl}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ at different Al^{3+} doping concentrations.

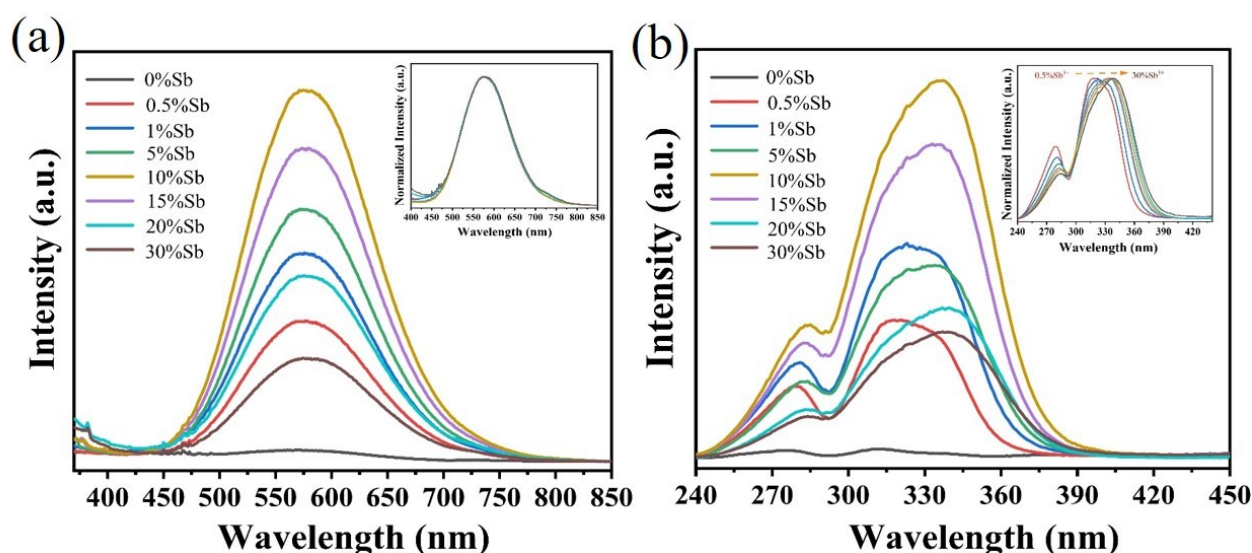


Fig. S14 (a). Emission spectra of $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ doped with different concentrations of Sb^{3+} under 330nm excitation. The inset shows its normalized spectrum, which exhibits the same spectral shape. (b). Excitation spectra of $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ doped with different concentrations of Sb^{3+} under 580nm emission. The normalized spectra of the upper right inset show that the excitation spectra are gradually red-shifted as the Sb^{3+} doping concentration gradually increases from 0.5% to 30%.

Note: the two insets of Fig..S14 showing the respective normalized PL spectra of their counterparts all show the same spectral shapes, and the almost exact agreement of all the PLE excitation spectra also suggested that the emission came from the same excited state out of Sb triplet STE, mainly due to the lowest triplet state of Sb-Cl in the system.

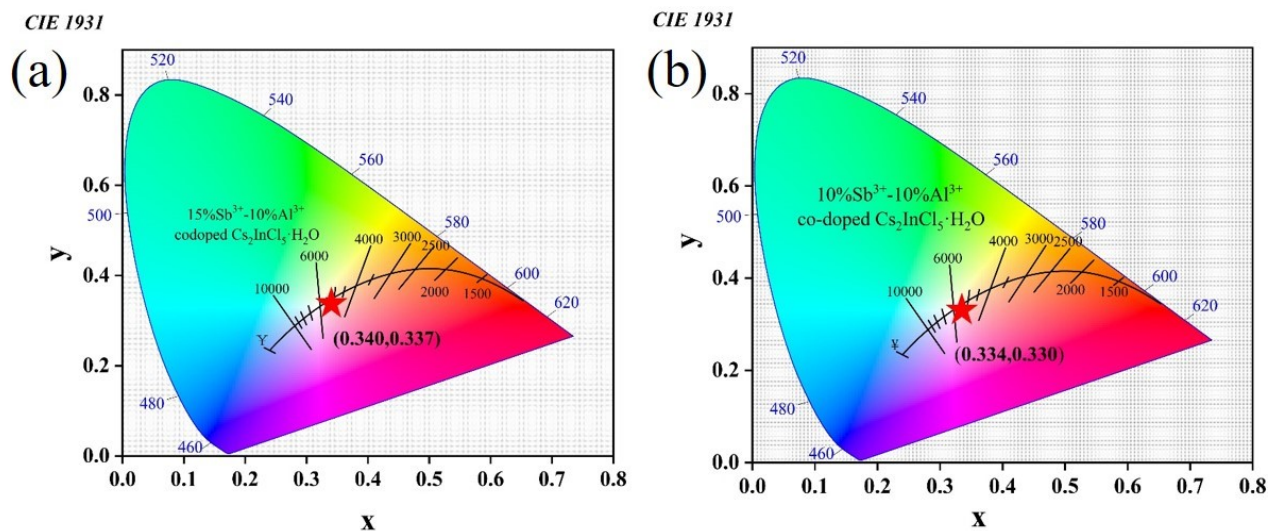


Fig. S15 The CIE coordinates of (a) $0.15\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$. under 338 nm excitation and (b) $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$. under 330 nm.

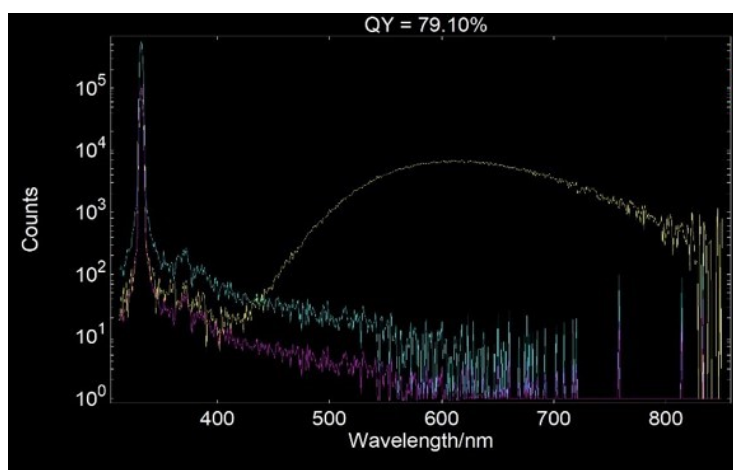


Fig. S16 The PLQY measurement cross-sectional view of 0.1Sb^{3+} doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ sample (the yellow curve) shows that the PLQY is as high as 79.10%. The excitation wavelength was 338 nm.

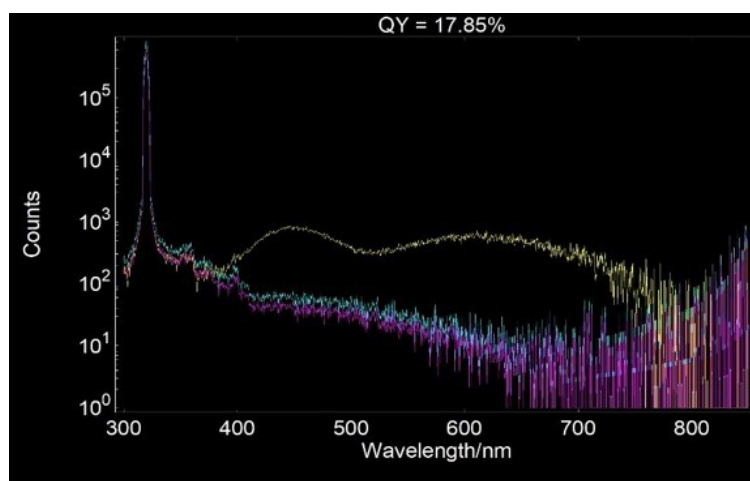


Fig. S17 The PLQY measurement cross-sectional view of 0.1Al^{3+} doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ sample (the yellow curve) shows that the PLQY is only 17.85%.

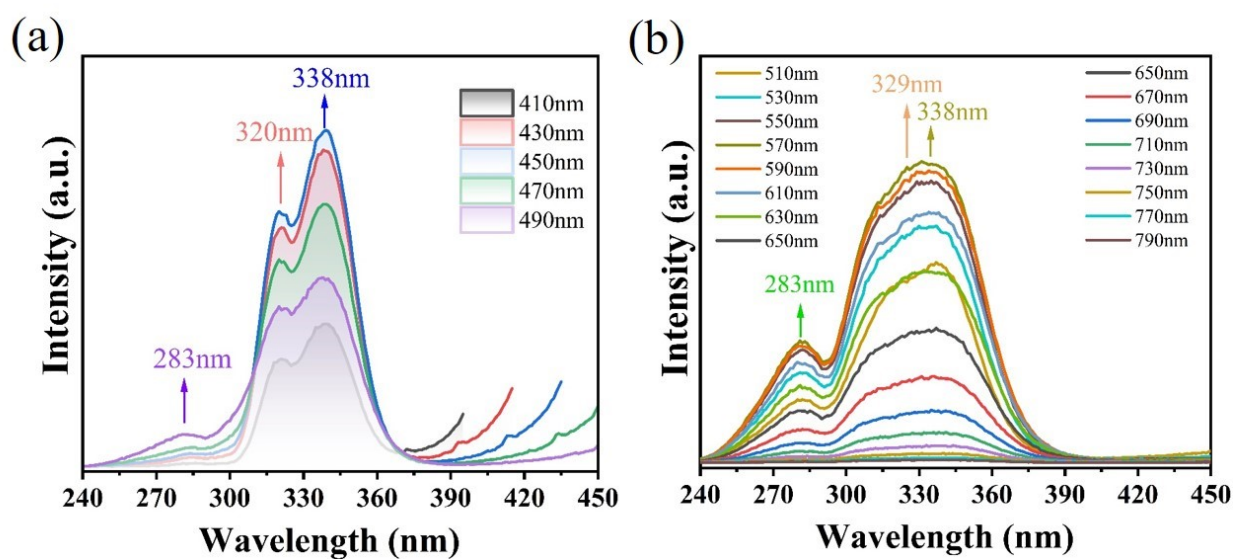


Fig. S18 PLE spectra of 0.1Sb^{3+} - 0.1Al^{3+} co-doped $\text{Cs}_2\text{InCl}_5 \cdot \text{H}_2\text{O}$ (a) from 410 to 490nm emission; (b) from 510 to 790nm emission.

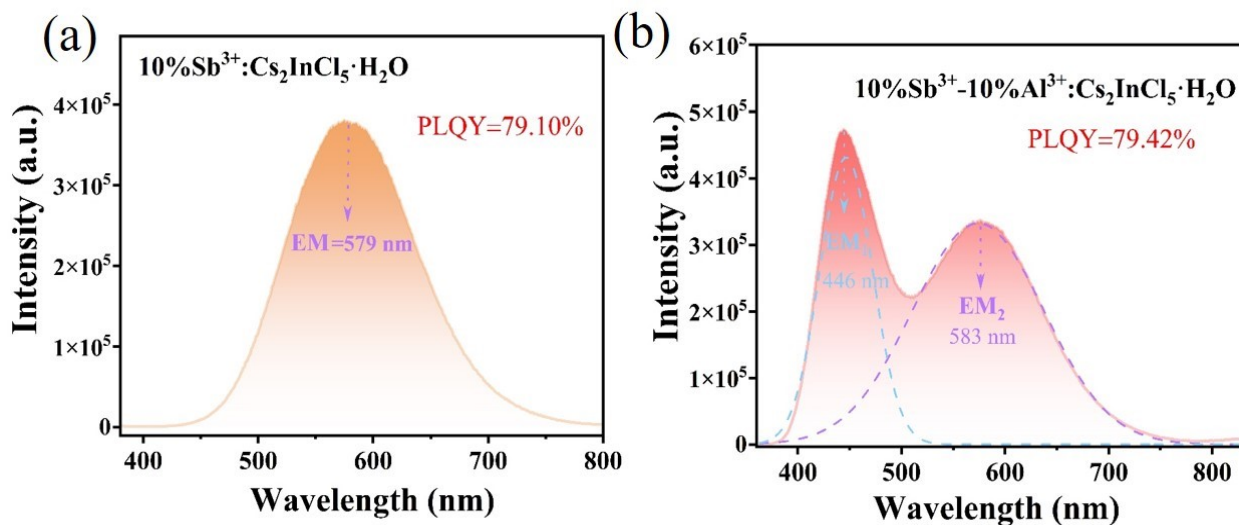


Fig. S19 PL spectrum at 335nm excitation of (a) 0.1Sb³⁺ doped Cs₂InCl₅·H₂O at 0.7 slits and (b) 0.1Sb³⁺-0.1Al³⁺ co-doped Cs₂InCl₅·H₂O at 0.7 slits.

Note: Due to the high luminous intensity of 0.1Sb³⁺-0.1Al³⁺ co-doped Cs₂InCl₅·H₂O and 0.1Sb³⁺ doped Cs₂InCl₅·H₂O, the slit is selected as 0.7 to protect the instrument. The PL spectra with EM₁(446nm) and EM₂(583nm) emission centers were obtained by peak fitting of the co-doped samples.

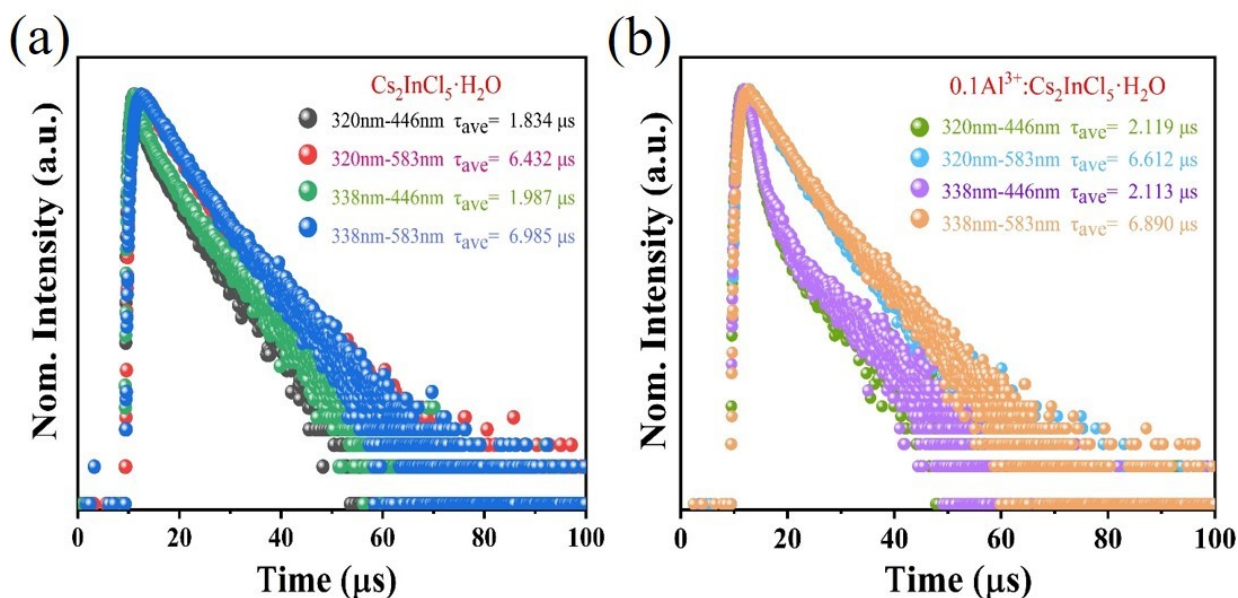


Fig. S20 The PL lifetime decay curves of (a) Cs₂InCl₅·H₂O and (b) 0.1Al³⁺ doped Cs₂InCl₅·H₂O.

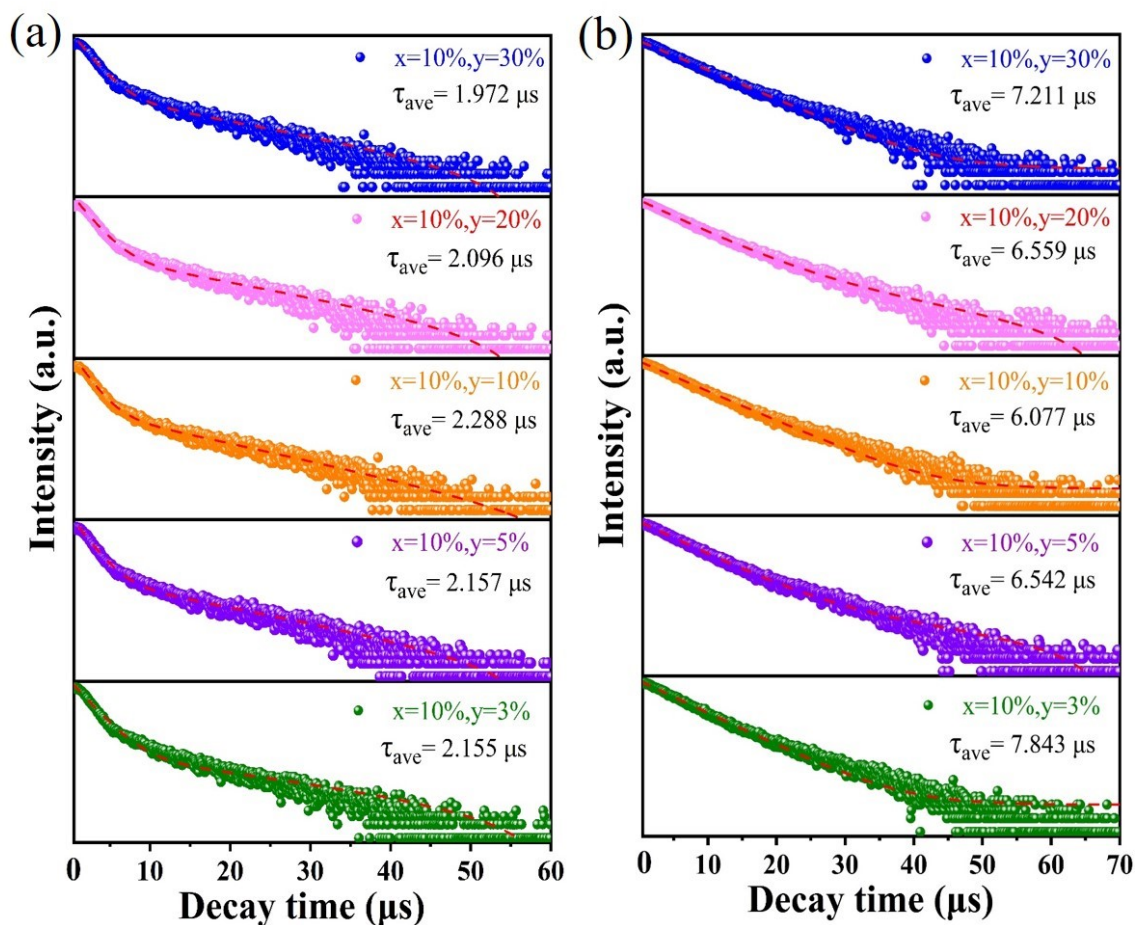


Fig. S21 The PL lifetime decay curves of 0.1Sb³⁺-yAl³⁺ co-doped Cs₂InCl₅·H₂O (a) under 338 nm excitation, monitored at 446 nm and (b) under 338 nm excitation, monitored at 583 nm. (y=0.03,0.05,.01,0.2,0.3)

Note: The average lifetime of the emission at 446 nm increases and then decreases with increasing Al³⁺ doping content, while the average lifetime of the emission decay at 583 nm decreases and then increases, which also confirms the previous results of possible energy transfer from high level In state to the lower Sb STE state emissions. This is due to the fact that the emission properties of Sb³⁺ are affected by Al³⁺ doping, which enhanced the high level In state lifetime. For the 446 nm decay center, the increase in Al³⁺ concentration results in more carrier population on In state and increased its lifetime. This high energy state at excessive Al³⁺ doping may lead to an increase in non-radiative complexation processes for more defects, which reduces the PL decay lifetime (Table S3). For the decay center at 583 nm, as the Al³⁺ concentration increases, the excited state of Sb³⁺ may be reduced by Al confinement and blocked energy transfer, leading to a decrease in its own lifetime. At even higher Al³⁺ doping concentration, the exciton leaps from the STE excitation energy level of In site to the lower STE of Sb site, resulting in a gradual increase of the PL decay lifetime at 583 nm. (Table S2).

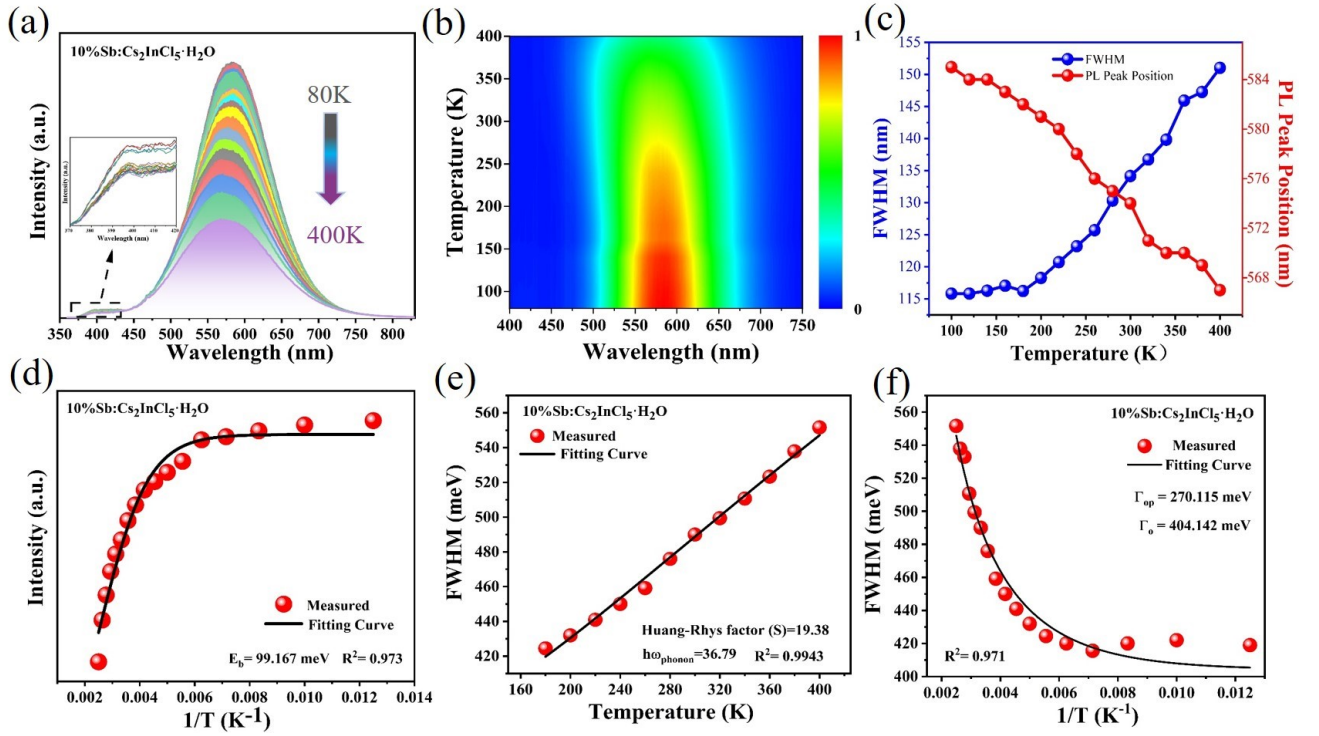


Fig. S22 0.1Sb³⁺ doped Cs₂InCl₅·H₂O of (a) temperature-dependent PL band at 583 nm from 80 K to 400 K (The inset is a local magnification along with the PL 365 nm–430 nm range.). (b) PL/Temperature correlation map upon excitation at 335 nm. (c) Variation of PL peak position and FWHM of emission with temperature. (d) The relationship between the photoluminescence intensity and temperature. (e) Fitting results of the FWHM (meV) as a function of temperature to obtain the Huang–Rhys factor (S). (f) Fitting results of the FWHM (meV) as a function of temperature inverse to obtain the phonon coupling energy.

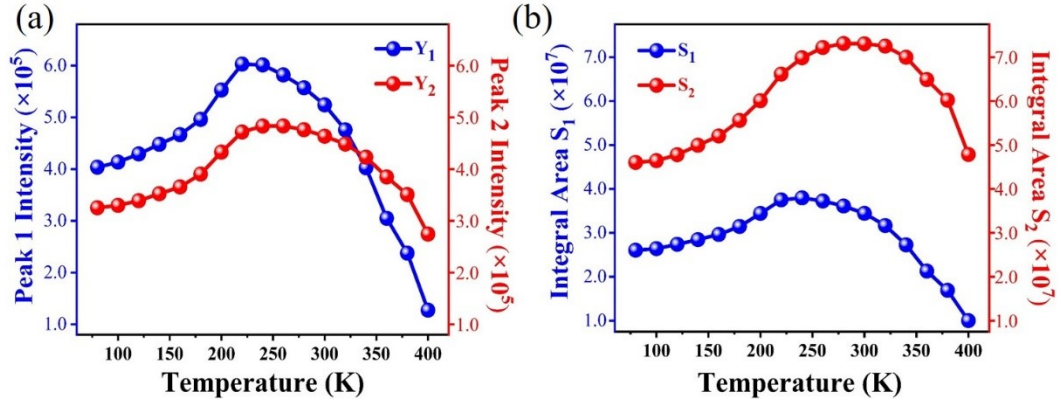


Fig. S23 (a) Temperature dependence curves of the intensity of emission peaks 1 and 2 after peak splitting of $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$; (b) Temperature dependence curves of the integration area of $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$.

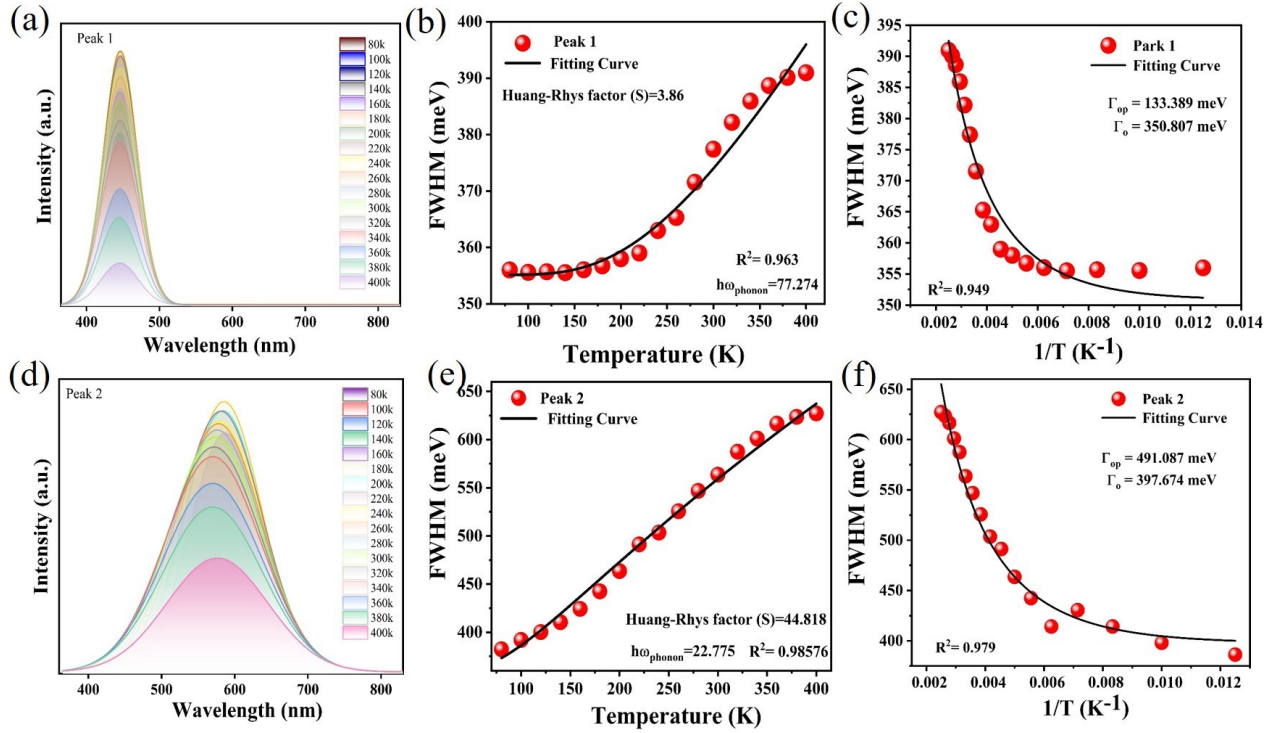


Fig. S24 Peak 1 of $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ after split-peak fitting for (a) temperature-dependent PL spectra from 80 K to 400 K; (b) fitting results of the FWHM (meV) as a function of temperature to obtain the Huang–Rhys factor (S); (c) fitting results of the FWHM (meV) as a function of temperature inverse to obtain the phonon coupling energy. Peak 2 of $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ after split-peak fitting for (d) temperature-dependent PL spectra from 80 K to 400 K; (e) fitting results of the FWHM (meV) as a function of temperature; (f) fitting results of the FWHM (meV) as a function of temperature inverse.

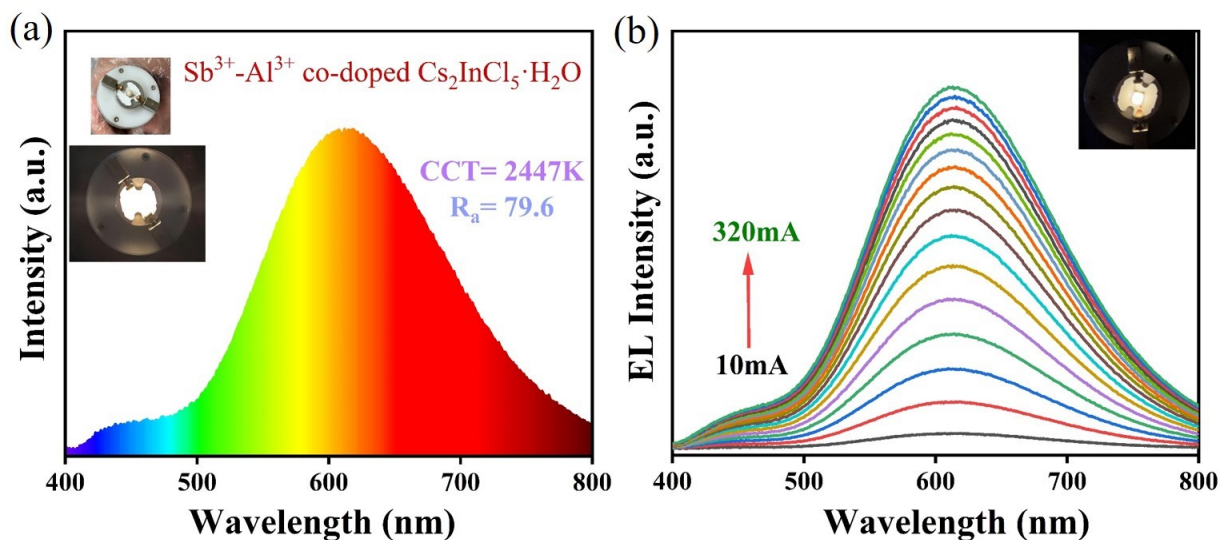


Fig. S25 LED were made by coating a sample powder of $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ on a 310-nm chip. (a) Emission spectrum under 20 mA drive current and the inset shows the WLED before and after operating. (b) Emission spectra of the device under various drive currents.

Note: The emission spectrum at 20 mA drive current, and we found a weak emission peak at about 450 nm, which is different from the LEDs fabricated with a 365 nm chip. This is mainly attributed to the different PL spectral profiles of the co-doped sample under different excitations, corresponding to the previous variable excitation spectrum (Fig.3a). The inset in Fig.S25a shows the LEDs before and after operation, showing a bright yellow light under power-up, corresponding to a color rendering index (R_a) of 79.6 for and a color temperature of 2447 K. The EL spectra at different driving currents ranging from 10 mA to 320 mA show the same profile (Fig. S25b).

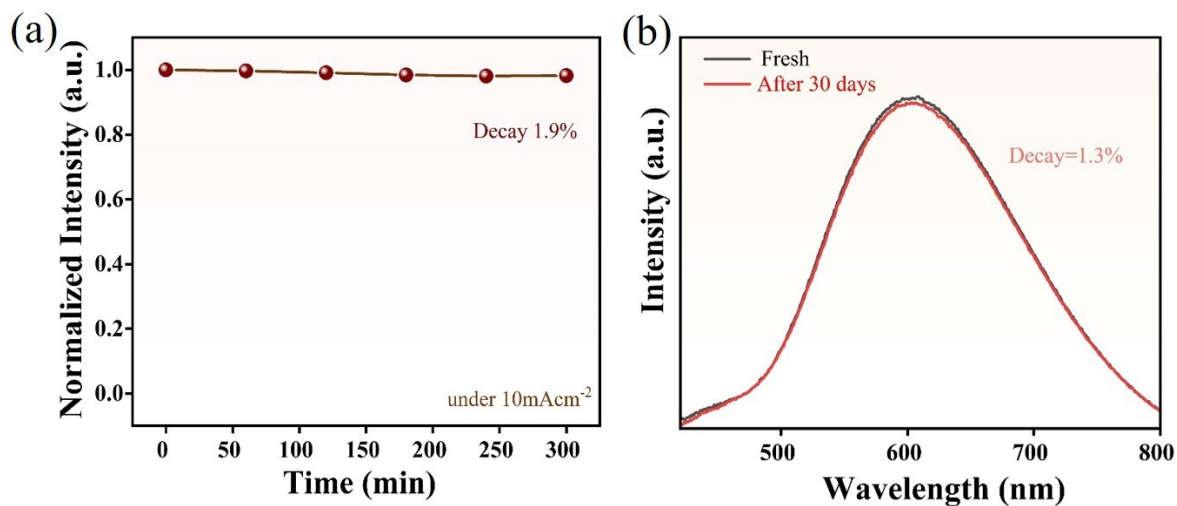


Fig. S26 LED were made by coating a sample powder of $0.1\text{Sb}^{3+}\text{-}0.1\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ on a 365-nm chip. (a) Long-term stability of the WLED device within 300 min. (b) Decay of emission intensity after 30 days of placement.

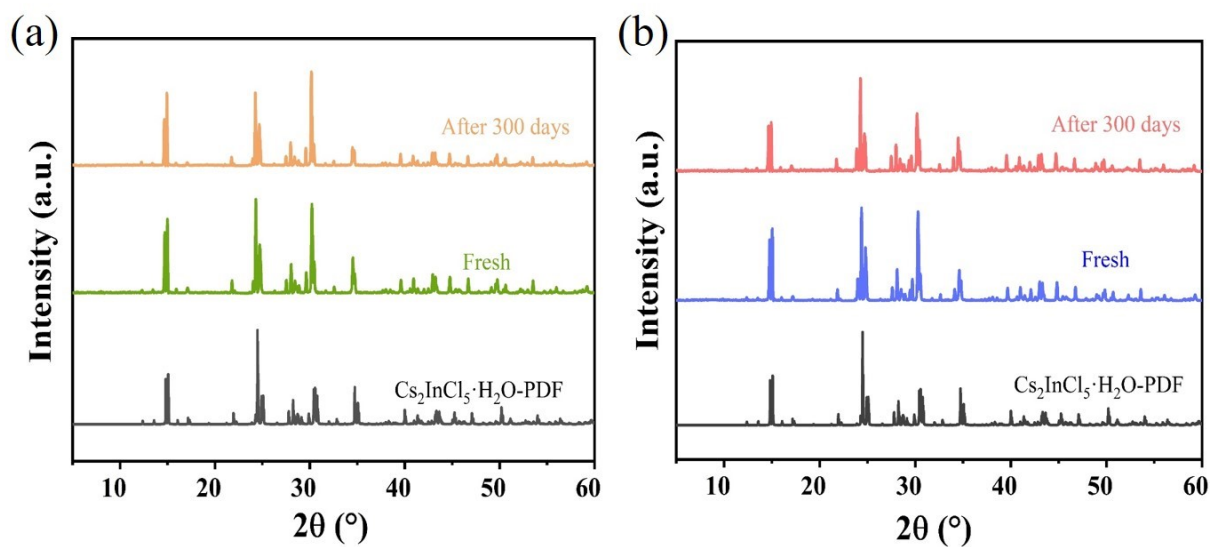


Fig. S27 $x\text{Sb}^{3+}\text{-}y\text{Al}^{3+}$ co-doped $\text{Cs}_2\text{InCl}_5\cdot\text{H}_2\text{O}$ of (a) $x=0.1, y=0$; and (b) $x=0.1, y=0.1$ for XRD image after five months of storage.

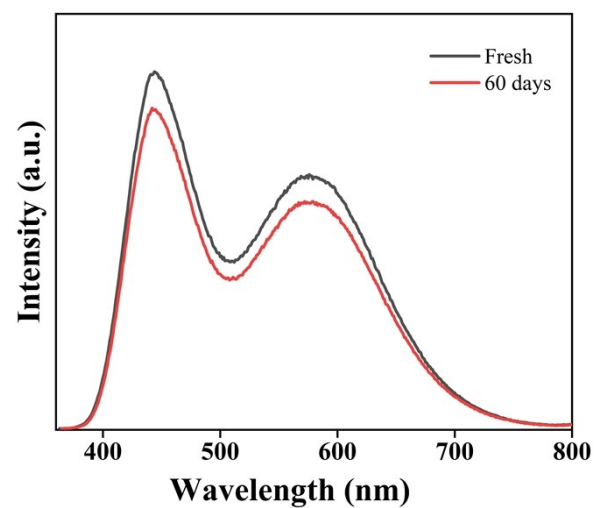


Fig. S28 Spectral comparison of 0.1Sb³⁺-0.1Al³⁺ co-doped Cs₂InCl₅·H₂O before and after storage for 60 days (excitation at 338 nm).

Table S1 Photophysical Properties of Cs₂InCl₅·H₂O related-doped materials.

Composition	Excitation Peak	Emission Peak	FWHM	PLQY	Refs
0.3%Te ⁴⁺ : Cs ₂ InCl ₅ ·H ₂ O	405 nm	660 nm	180 nm	13.2%	4
0.07Bi ³⁺ : Cs ₂ InCl ₅ ·H ₂ O	310 nm	410 nm;652 nm	—	46%	5
0.25Cr ³⁺ : (Cs _{0.7} K _{0.3}) ₂ Cl ₅ ·H ₂ O	360 nm	910 nm	175 nm	75.1%	6
40%Mn ³⁺ : Cs ₃ InCl ₆	302 nm	440 nm;520 nm;620 nm	—	32.18%	7
0.01%Sb ³⁺ -0.01%Bi ³⁺ : Cs ₂ InCl ₅ ·H ₂ O	260 nm	420 nm;610 nm	—	—	8
10%Pb ²⁺ : Cs ₂ InCl ₅ ·H ₂ O	275 nm	390 nm;430 nm;520 nm	120 nm	58%	9
40%Sb ³⁺ : Rb ₂ InCl ₅ ·H ₂ O	365 nm	600 nm	200 nm	62.1%	10
0.1Sb ³⁺ -0.1Al ³⁺ : Cs ₂ InCl ₅ ·H ₂ O	338 nm	446 nm;583 nm	235 nm	~80%	This work
10%Sb ³⁺ : Cs ₂ NaInCl ₆	310 nm	443 nm	100 nm	75.89%	11
0.5%Sb ³⁺ : Cs ₂ InCl ₅ ·H ₂ O	340 nm	580 nm	135nm	~95%	12
Sb ³⁺ : Rb ₃ InCl ₆	320 nm	497 nm	155nm	95%	13

Table S2 PL decay parameters of the 0.1Sb³⁺-yAl³⁺ co-doped Cs₂InCl₅·H₂O excited at 338 nm and monitored at 583 nm.

x	y	τ_1 (μ s)	A ₁ (%)	τ_2 (μ s)	A ₂ (%)	τ_{ave} (μ s)
0.1	0.03	5.893	99.766	74.077	0.233	7.843
0.1	0.05	5.866	99.502	31.648	0.497	6.542
0.1	0.1	5.671	99.291	21.255	0.708	6.077
0.1	0.2	5.973	99.631	34.206	0.368	6.559
0.1	0.3	6.057	99.746	56.211	0.253	7.211

Table S3 PL decay parameters of the 0.1Sb³⁺-yAl³⁺ co-doped Cs₂InCl₅·H₂O excited at 338 nm and monitored at 446 nm.

x	y	τ_1 (μ s)	A ₁ (%)	τ_2 (μ s)	A ₂ (%)	τ_{ave} (μ s)
0.1	0.03	17.067	0.033	2.074	99.966	2.115
0.1	0.05	17.694	0.034	2.113	99.968	2.157
0.1	0.1	23.554	0.031	2.217	99.968	2.288
0.1	0.2	19.072	0.024	2.058	99.975	2.096
0.1	0.3	14.44413	0.0303	1.9446	99.96968	1.972

Table S4 PL decay parameters of the 0.1Al³⁺ doped Cs₂InCl₅·H₂O under different excitation and emission.

x=0 , y=0.1	τ_1 (μ s)	A ₁ (%)	τ_2 (μ s)	A ₂ (%)	τ_{ave} (μ s)
338nm-583nm	6.36821	98.73566	19.92819	1.26434	6.890
338nm-446nm	15.01436	0.04277	2.07339	99.95723	2.113
320nm-583nm	6.16337	99.36734	24.42454	0.63266	6.612
320nm-446nm	17.56166	0.03052	2.08019	99.96948	2.119

Table S5 PL decay parameters of the 0.1Sb³⁺ doped Cs₂InCl₅·H₂O under different excitation and emission.

x=0.1 , y=0	τ_1 (μ s)	A ₁ (%)	τ_2 (μ s)	A ₂ (%)	τ_{ave} (μ s)
338nm-583nm	5.81391	99.641	37.33849	0.359	6.527
338nm-446nm	3.34684	96.438	9.34342	3.562	3.907
320nm-583nm	5.78425	62.714	5.78389	37.286	5.784
320nm-446nm	2.54856	98.881	9.16292	1.119	2.807

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