## **Supporting Information**

## **0D** Hybrid Indium Halides with Highly Efficient Intrinsic Broadband Light Emissions

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## **Experimental Section**

**Materials.** The chemical materials and regents were commercially purchased from Aladdin chemical company and directly used in the preparation reaction without any further purification or other physical process. InCl<sub>3</sub>·4H<sub>2</sub>O (99.99%), InBr<sub>3</sub> (99.99%), N,N'-Dimethylpiperazine (DMP,  $C_6H_{16}N_2$ , 99%), hydrochloric acid (HCl, 37%), hydrobromic acid (HBr, 50%), acetone (80%), *n*-Propanol (99%).

Synthesis of  $[H_2DMP]_2InCl_7 2H_2O$ . Mixture of  $InCl_3 4H_2O$  (1 mmol, 0.2932 g) and N,N'-Dimethylpiperazine (1 mmol, 0.116 g) was dissolved in a mixed solvent of hydrochloric acid (1 mL), acetone (4 mL) and *n*-propanol (1 mL) with constant magnetic stirring for one hour to obtain clear solution. Then, the mixed solution was sealed in a 25 mL glass vial and reacted at 80 °C for 5 days. After cooling to room temperature, colorless layer-shaped crystals were obtained and subsequently determined as  $[H_2DMP]_2InCl_7 2H_2O$  ( $C_{12}H_{36}N_4O_2InCl_7$ ) by single-crystal X-ray diffraction. The crystals were then washed with ethanol, dried, and stored in a vacuum (75% yield based on In). Elemental analysis calculated for  $C_{12}H_{36}N_4O_2InCl_7$ : C, 22.82%; H, 5.74%; N, 8.87%; found: C, 22.90%; H, 5.75%; N, 8.92%.

Synthesis of  $[H_2DMP]_2InBr_7 2H_2O$ . InBr<sub>3</sub> (1 mmol, 0.354 g) and N,N'-Dimethylpiperazine (1 mmol, 0.116 g) were mixed and dissolved in a mixed solution of hydrochloric acid (1 mL) and acetone (3 mL) with constant magnetic stirring and heating at about 25 °C leading to a clear solution. Then, the mixed solution was sealed in a 25 mL glass vial and reacted at 80 °C for 3 days. After cooling to room temperature, colorless layer-shaped crystals were obtained and subsequently determined to be  $[H_2DMP]_2InBr_7 2H_2O$  ( $C_{12}H_{36}N_4O_2InBr_7$ ) by single-crystal X-ray diffraction. The crystals were then washed with ethanol, dried, and stored in a vacuum (78% yield based on In). Elemental analysis calculated for  $C_{12}H_{36}N_4O_2InBr_7$ : C, 15.29%; H, 3.84%; N, 5.94%; found: C, 15.35%; H, 3.88%; N, 5.98%.

Single-Crystal X-ray Crystallography. The single crystal data of  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  and  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  were collected on the Bruker Apex-II CCD diffractometer with Cu-K $\alpha$  radiation ( $\lambda = 1.5378$ ) at room temperature. The crystal structures were solved by direct method and refined based on  $F^2$  using SHELXTL-2018 program. All the non-hydrogen atoms were refined with anisotropic thermal parameters, and hydrogen atoms of organic molecules were positioned

geometrically and refined isotropically. Structural refinement parameters of  $[H_2DMP]_2InBr_7 \cdot 2H_2O$ and  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  were summarized in Table S2 and important bond parameters were listed in Tables S3-S6. The Cambridge Crystallographic Data Centre (CCDC) 2166146 (**DMP-Br**; 273.15 K) and 2166147 (**DMP-Cl**; 273.15 K) contain the supplementary crystal-lographic data for this paper. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre.

**Powder X-ray Diffraction.** The powder X-ray diffraction (PXRD) analysis was performed on Bruker D8 ADVANCE powder X-ray diffractometer equipped with Cu-K $\alpha$  radiation at a voltage of 40 kV and a current of 40 mA. The diffraction patterns were scanned over the angular range of 5-60° (2 $\theta$ ) with a step size of 5 °/min at room temperature. Simulated PXRD data was calculated from the single crystal data in Mercury software.

**Common Characterizations.** Elemental mapping was conducted on a scanning electron microscope (SEM, Zeiss Merlin Compact). The thermal stability was tested by using the thermogravimetric analysis (TGA), which was carried out on a Mettler TGA/SDTA 851 thermal analyzer in the temperature range of 30-800 °C under the constant protection of N<sub>2</sub> atmosphere flow. The solid state UV-vis absorption optical spectrum was collected at PE Lambda 900 UV/Vis spectrophotometer at 300 K in wavelength range of 200-800 nm with BaSO<sub>4</sub> serving as a reference standard. The Raman spectrum measurement was performed on powder sample in the range of 100-4000 cm<sup>-1</sup> by using Horiba Scientific LabRam HR Evolution under 365 nm excitation wavelength.

**Photoluminescence Property Characterization.** The PL spectrum was performed on an Edinbergh FLS980 fluorescence spectrometer. The photoluminescence quantum yield (PLQY) was achieved by incorporating an integrating sphere into the FLS980 spectrofluorometer. The PLQY was calculated based on the equation:  $\eta_{\text{QE}} = I_{\text{S}}/(E_{\text{R}}-E_{\text{S}})$ , which  $I_{\text{S}}$  represents the luminescence emission spectrum of the sample,  $E_{\text{R}}$  was the spectrum of the excitation light from the empty integrated sphere (without the sample), and  $E_{\text{S}}$  was the excitation spectrum for exciting the sample. The time-resolved decay data was carried out using the Edinburgh FLS980 fluorescence spectrometer with a picosecond pulsed diode laser. The average lifetime was obtained by exponential fitting. The corresponding Commission Internationale Eclairage (CIE) chromaticity coordinates were calculated based on PL emission spectrum.

The thermal activation energy  $(E_a)$  can be evaluated by fitting the temperature-dependent integral PL emission intensity according to the Arrhenius-type function:

$$I_{PL} = \frac{I_0}{1 + Aexp(-\frac{E_a}{k_B T})}$$

where  $I_{PL}$  and  $I_0$  belong to the integrated emission intensity at *T* and 0 K, respectively,  $k_B$  is the Boltzmann constant, and  $E_a$  represents the energy barrier for exciton to be bound from radiative to nonradiative centers.

The temperature-dependent FWHM can be fitting based on the follow equation:

$$\Gamma(T) = \Gamma_0 + \Gamma_{phonon} (e^{E_{LO}/k_B T} - 1)^{-1} + \Gamma_{inhomo} e^{-E_b/k_B T}$$

where  $\Gamma_0$  is the FWHM of 0 K,  $E_{\rm LO}$  and  $E_{\rm b}$  represent the energy of the longitudinal-optical phonon and trap states,  $\Gamma_{\rm phonon}$  and  $\Gamma_{\rm inhomo}$  are the relative contributions of electron-phonon coupling and trapped states induced inhomogeneous broadening, respectively.

**Fabrication of White LED Lamp.** The white LED lamp was fabricated by combining a 365 nm UV LED chip with the yellow emissive  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  and blue phosphor (BaMgAl<sub>10</sub>O<sub>17</sub>:Eu<sup>2+</sup>). The phosphors were mixed with epoxy resin and stirred continuously for 10 min. Then the mixture was coated on the surface of the UV LED chip and dried for 30 min under vacuum condition. The electroluminescence spectrum of the fabricated white LED was evaluated by a temperature-programmed LED optoelectronic analyzer with an integrating sphere (EVERFINE HAAS-2000).

**Theoretical Calculation on Electronic Band Structure.** The single crystal data of  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  and  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  were used to calculate the electronic band structure in CASTEP software based on density functional theory (DFT) and Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation. The interactions between the ionic cores and the electrons were described by the norm-conserving pseudopotential. Hence, the C- $2s^22p^2$ , N- $2s^22p^3$ , H- $1s^1$ , In- $5s^25p$ , Br- $4s^24p^5$  and Cl- $3s^23p^5$  orbital were adopted as valence electrons. The number of plane wave included in the basis sets was determined by a cutoff energy of 320 eV and numerical integration of the Brillouin zone is performed using Monkhorst-Pack k-point sampling of  $2 \times 2 \times 2$ . Other calculating parameters and convergence criteria were set by the default values of the CASTEP cod.



**Fig. S1** Experimental and simulated XRD patterns of  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (**DMP-CI**) (a) and  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (**DMP-Br**) (b).



**Fig. S2** The SEM photo images SEM elemental mapping of single crystal of [H<sub>2</sub>DMP]<sub>2</sub>InCl<sub>7</sub>·2H<sub>2</sub>O (**DMP-Cl**).





![](_page_7_Figure_0.jpeg)

**Fig. S4** The thermogravimetric analysis (TGA) curves of  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (**DMP-C1**) and  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (**DMP-Br**).

![](_page_8_Figure_0.jpeg)

**Fig. S5** The solid-state UV-vis absorption spectra and Tauc plots based on the assumptions of direct transitions of [H<sub>2</sub>DMP]<sub>2</sub>InCl<sub>7</sub>·2H<sub>2</sub>O (**DMP-Cl**) (a-b) and [H<sub>2</sub>DMP]<sub>2</sub>InBr<sub>7</sub>·2H<sub>2</sub>O (**DMP-Br**) (c-d).

![](_page_9_Figure_0.jpeg)

Fig. S6 The solid-state UV-vis absorption spectrum of  $[H_2DMP]Cl_2$ .

![](_page_10_Figure_0.jpeg)

Fig. S7 The PL emission spectrum (a) and decay curve (b) of organic salt [H<sub>2</sub>DMP]Cl<sub>2</sub> at 300 K.

![](_page_11_Figure_0.jpeg)

Fig. S8 The PL decay curve monitoring at 441 nm of DMP-Cl at 300 K.

![](_page_12_Figure_0.jpeg)

Fig. S9 The PLQY values of  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (DMP-Cl) (a) and  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (DMP-Br) (b).

![](_page_13_Figure_0.jpeg)

Fig. S10 The excitation wavelength dependent PL excitation spectra (a and b), and emission wavelength dependent PL excitation spectra (c) for  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (DMP-Cl).

![](_page_14_Figure_0.jpeg)

Fig. S11 The excitation wavelength dependent PL excitation spectra (a), and emission wavelength dependent PL excitation spectra (b) for  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (DMP-Br).

![](_page_15_Figure_0.jpeg)

Fig. S12 Comparisons of PL emission spectra of bulk crystals and microscale powders for  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (DMP-Cl) (a) and  $[H_2DMP]InBr_7 \cdot 2H_2O$  (DMP-Br) (b).

![](_page_16_Figure_0.jpeg)

Fig. S13 Raman spectra of  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (DMP-Cl) (a) and  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (DMP-Br) (b).

![](_page_17_Figure_0.jpeg)

**Fig. S14** Calculated band structure (a) and density of states (b) for [H<sub>2</sub>DMP]<sub>2</sub>InBr<sub>7</sub>·2H<sub>2</sub>O (**DMP-Br**) (b).

![](_page_18_Figure_0.jpeg)

Fig. S15 PXRD patterns and PL spectra of  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (DMP-Cl) and

 $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (**DMP-Br**) after immersing in various organic solvents (a-d) and constant heating in various temperature (e-h) (ACN = acetonitrile, DCM = dichloromethane, EAC = acetylacetone, CP = acetone, EtOH = ethonal, *i*-PrOH = isopropyl alcohol).

**Note:** After immersion in various organic solvents for one day, there are almost no change in PXRD patterns indicating the unvaried crystal lattice, and the corresponding PL emission spectra also show almost identical spectral profiles with sufficient emission intensity, which proves highly stable PL performance of **DMP-Cl** and **DMP-Br**. Under constant heating at high temperature, **DMP-Cl** and **DMP-Br** also show identical structural lattice and sufficient emission intensities.

![](_page_19_Figure_2.jpeg)

**Fig. S16** Characterizations of a fabricated white LED: (a) EL spectrum at 20 mA drive current (inset: photograph of white LED), (b) EL spectra under different current.

**Note**: The white light emitting diode (LED) was prepared by coating the mixture of **DMP-Cl** and blue fluorescent (BaMgAl<sub>10</sub>O<sub>17</sub>:Eu<sup>2+</sup>) on an UV chip. The white LED displays bright "warm" white light emission under the drive of 20 mA current with perfect CIE coordinates of (0.34, 0.34) and color temperature of 5152 K. The color rendering index (CRI) of 91.2 is obviously higher than the value of commercial available WLED based on YAG: Ce<sup>3+</sup> (< 80). Under the drive of increased currents from 20 to 120 mA, the steadily enhanced emission intensity demonstrates the potential application in high-power optoelectronic device (Fig. S16b).

Material	Emission	PLQY	FWHM	Lifetime	Ref.
[H <sub>2</sub> DMP] <sub>2</sub> InCl <sub>7</sub> ·2H <sub>2</sub> O	568 nm	58.53%	158 nm	6.5972 µs	This work
Cs <sub>3</sub> InBr <sub>6</sub>	440 nm	46%	80 nm		1
[DAPEDA]InCl <sub>6</sub> ·Cl·H <sub>2</sub> O	520 nm	40.40%	116 nm	3.3150 µs	2
(PMA) <sub>3</sub> InBr <sub>6</sub>	610 nm	35%	132 nm	1.260 µs	2
[DPA] <sub>3</sub> InCl <sub>6</sub>	510 nm	34.01%	108 nm	3.1819 µs	3
Cs <sub>2</sub> InBr <sub>5</sub> ·H <sub>2</sub> O	695 nm	33%		1.65 μs	4
[H <sub>2</sub> DMP] <sub>2</sub> InBr <sub>7</sub> ·2H <sub>2</sub> O	610 nm	28.73%	149 nm	1.33 µs	This work
Cs <sub>2</sub> InCl <sub>5</sub> ·H <sub>2</sub> O	610 nm	18%	164 nm		5
Cs <sub>3</sub> InBr <sub>6</sub>	375 nm	17.1%	75 nm		6
RInBr <sub>4</sub>	437 nm	16.36%	95.3 nm		7
(C <sub>8</sub> NH <sub>12</sub> ) <sub>6</sub> InBr <sub>9</sub> ·H <sub>2</sub> O	475 nm	8.85%	240 nm		8
$(C_4H_{14}N_2)_2In_2Br_{10}$	670 nm	3%		3.2 µs	9
(C <sub>6</sub> H <sub>8</sub> N) <sub>6</sub> InBr <sub>9</sub>	544 nm	2.72%	100 nm		10
Rb <sub>2</sub> InCl <sub>5</sub> ·H <sub>2</sub> O	408 nm	<1%			11

**Table S1.** Summary of the PL properties of 0D indium perovskites.

PMA = phenylmethylammonium; DAPEDA = 1,2-bis(3-aminopropylamino)ethane; DPA = dipropylamine; R = trimethyl(4-stilbenyl)methylammonium; C<sub>4</sub>H<sub>14</sub>N<sub>2</sub> = 2-(dimethylamino)ethylamine; DMP = N,N'-Dimethylpiperazine; C<sub>6</sub>H<sub>8</sub>N = aniline.

Compound	$[H_2DMP]_2InCl_7 \cdot 2H_2O$	$[H_2DMP]_2InBr_7 \cdot 2H_2O$
chemical formula	$C_{12}N_4H_{36}O_2InCl_7$	C <sub>12</sub> N <sub>4</sub> H <sub>36</sub> O <sub>2</sub> InBr <sub>7</sub>
fw	631.42	942.64
Temp (K)	296.15	296.15
Space group	P21212	$P2_{1}2_{1}2$
<i>a</i> (Å)	9.301(6)	11.7222(10)
<i>b</i> (Å)	11.500(8)	12.5328(11)
<i>c</i> (Å)	12.069(8)	9.6047(8)
α (°)	90	90
eta (°)	90	90
γ (°)	90	90
$V(Å^3)$	1291.0(15)	1411.0(3)
Z	2	2
$D_{\text{calcd}}(\text{g}\cdot\text{cm}^{-3})$	1.624	2.219
$\mu$ (mm <sup>-1</sup> )	1.655	10.763
F (000)	640	892
Reflections collected	10241	16742
Unique reflections	2911	3306
GOF on $F^2$	1.069	0.952
$^{a}R_{1}, wR_{2} (I > 2\sigma(I))$	0.0435/0.1166	0.0273/0.0526
${}^{b}R_{1}, wR_{2}$ (all data)	0.0474/0.1197	0.0361/0.0545

**Table S2.** Crystal Data and Structural Refinements for compounds  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (**DMP-Cl**)and  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (**DMP-Br**).

 ${}^{a}R_{1} = \sum ||F_{o}| - |F_{c}|| / \sum |F_{o}|. {}^{b}wR_{2} = [\sum w(F_{o}^{2} - F_{c}^{2})^{2} / \sum w(F_{o}^{2})^{2}]^{1/2}.$ 

In1-Cl2	2.794(3)	In1-Cl3 <sup>1</sup>	2.673(3)
In1-Cl2 <sup>1</sup>	2.794(3)	In1-Cl4	2.426(2)
In1-Cl3	2.673(3)	In1-Cl4 <sup>1</sup>	2.426(2)
Cl2 <sup>1</sup> -In1-Cl2	62.70(9)	Cl3 <sup>1</sup> -In1-Cl3	65.78(10)
Cl3-In1-Cl2 <sup>1</sup>	177.70(6)	Cl4-In1-Cl2 <sup>1</sup>	92.13(7)
Cl3 <sup>1</sup> -In1-Cl2	177.70(6)	Cl4-In1-Cl2	83.14(7)
Cl3-In1-Cl2	115.79(7)	Cl4 <sup>1</sup> -In1-Cl2	92.13(7)
Cl3 <sup>1</sup> -In1-Cl2 <sup>1</sup>	115.79(7)	Cl4 <sup>1</sup> -In1-Cl2 <sup>1</sup>	83.14(7)
Cl41-In1-Cl31	89.36(7)	Cl4 <sup>1</sup> -In1-Cl3	95.29(7)
Cl4-In1-Cl31	95.29(7)	Cl4-In1-Cl3	89.36(7)
1+X,1/2-Y,+Z			

**Table S3**. Selected bond lengths (Å) and bond angles (°) for  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (**DMP-CI**).

D-H···A	d(D-H)	$d(H \cdots A)$	$d(D \cdots A)$	<(DHA)
N2-H5…Cl2	0.98	2.48	3.386(7)	153
N1-H14…O2	0.98	2.58	3.405(12)	142
N1-H14…Cl3	0.98	2.81	3.457(6)	124
O2-H21…Cl1	0.86	2.22	2.906(8)	138
C6-H1…Cl4	0.96	2.10	2.982(7)	152
C7-H6…Cl4	0.97	2.45	3.366(6)	156
C5-H10…Cl1	0.96	2.71	3.197(10)	112
С3-Н18…О2	0.97	2.12	2.973(10)	145

**Table S4.** Hydrogen bonds data for  $[H_2DMP]_2InCl_7 \cdot 2H_2O$  (**DMP-Cl**).

In1-Br2	2.6298(7)	In1-Br3 <sup>1</sup>	2.7096(6)
In1-Br2 <sup>1</sup>	2.6298(7)	In1-Br4	2.7101(7)
In1-Br3	2.7096(6)	In1-Br4 <sup>1</sup>	2.7101(7)
Br2-In1-Br2 <sup>1</sup>	90.86(3)	Br2-In1-Br4 <sup>1</sup>	177.24(2)
Br2-In1-Br3 <sup>1</sup>	90.22(2)	Br2 <sup>1</sup> -In1-Br4 <sup>1</sup>	177.24(2)
Br2 <sup>1</sup> -In1-Br3 <sup>1</sup>	92.30(2)	Br3 <sup>1</sup> -In1-Br3	176.41(3)
Br2-In1-Br3	92.30(2)	Br31-In1-Br4	85.25(2)
Br2 <sup>1</sup> -In1-Br3	90.22(2)	Br3-In1-Br4 <sup>1</sup>	85.25(2)
Br2-In1-Br4	90.413(18)	Br31-In1-Br41	92.17(2)
Br2 <sup>1</sup> -In1-Br4 <sup>1</sup>	90.413(18)	Br3-In1-Br4	92.17(2)

**Table S5**. Selected bond lengths (Å) and bond angles (°) for  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (**DMP-Br**).

1+X, 1/2-Y,+Z

D-H···A	d(D-H)	$d(H \cdots A)$	$d(D \cdots A)$	<(DHA)
N2-H5…O2	0.98	1.84	2.764(6)	156
N1-H13…Br4	0.98	2.78	3.566(5)	137
N1-H13…Br3	0.98	2.65	3.412(4)	135
O2-H18…Br3	0.85	2.49	3.324(4)	169
O2-H19…Br1	0.85	2.54	3.277(5)	146
C3-H17…Br2	0.97	2.92	3.636(5)	131

**Table S6.** Hydrogen bonds data for  $[H_2DMP]_2InBr_7 \cdot 2H_2O$  (**DMP-Br**).

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