

*Supporting Information for*

**Achieving Switchable UV/Vis Circularly Polarized  
Luminescence by Varying Excitation Wavelength in Chiral  
Indium Halides**

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## 1. Experimental section

### Methods:

**Materials:** (*R*)-(-)-2-methylpiperazine and (*S*)-(+)-2-methylpiperazine (*R/S*-MP, 98%+, Adamas Shanghai, China); 2,2'-bipyridine (2,2'-bpy, 99%, Beijing Huawei Ruike Chemical, Beijing, China); indium(III) chloride tetrahydrate ( $\text{InCl}_3 \cdot 4\text{H}_2\text{O}$ , 99.99%, 9dingchem, Shanghai, China); acetonitrile ( $\text{CH}_3\text{CN}$ , AR, Sinopharm Chemical Reagent Co., Ltd., Shanghai, China); hydrochloric acid ( $\text{HCl}$ , AR, Sinopharm Chemical Reagent Co., Ltd., Shanghai, China). All purchased reagents are used directly without further purification.

**Synthesis of *R*-1 single crystal:** Dissolve  $\text{InCl}_3 \cdot 4\text{H}_2\text{O}$  (0.52 mmol, 0.1533 g), 2,2'-bpy (0.5 mmol, 0.0793 g), and *R*-2-MP (2 mmol, 0.2008 g) in a mixed solution of  $\text{HCl}$  (1 mL) and  $\text{CH}_3\text{CN}$  (2 mL). The mixture was then heated in an oven at 80°C for 10 minutes and removed. After cooling to room temperature (RT) under ambient conditions, the solution was evaporated, yielding colorless block crystals (0.1400 g). The yield was calculated to be nearly 54% based on In atom. EA: Calcd (%): C, 32.70; H, 3.79; N, 10.18. Found (%): C, 32.36; H, 3.29; N, 10.23.

**Synthesis of *S*-1 single crystal:** Dissolve  $\text{InCl}_3 \cdot 4\text{H}_2\text{O}$  (0.50 mmol, 0.1474 g), 2,2'-bpy (0.5 mmol, 0.0799 g), and *S*-2-MP (2 mmol, 0.2060 g) in a mixture of  $\text{HCl}$  (1 mL) and  $\text{CH}_3\text{CN}$  (2 mL). The mixture was heated in an oven at 80°C for 10 minutes, then removed and allowed to cool to RT before being evaporated at room temperature, yielding white colorless crystals (0.1631 g). The yield was calculated to be nearly 65.6% based on In atom. EA: Calcd (%): C, 32.70; H, 3.79; N, 10.18. Found (%): C, 31.23; H, 3.24; N, 10.98.

### Characterization

**X-ray crystallography.** A single crystal suitable for single-crystal X-ray diffraction (SCXRD) was selected under an optical microscope. Single crystal X-ray diffraction data were collected with graphite-monochromated  $\text{MoK}\alpha$  ( $\lambda = 0.71073 \text{ \AA}$ ) using an XtaLAB Synergy X, HyPix diffractometer at 100 K. The structures were solved by direct methods and refined with full-matrix least squares on  $F^2$  by the *SHELX*2018

package.<sup>1</sup>

**Powder X-ray diffraction (PXRD):** The experimental PXRD patterns were measured by Rigaku Miniflex-II diffractometer by utilizing  $\text{CuK}\alpha$  radiation ( $\lambda = 1.54178 \text{ \AA}$ ) in the angular range of  $2\theta = 5 - 65^\circ$ . The simulated PXRD patterns were calculated by using the single crystal X-ray structural data at 100 K via Mercury software.

**Thermogravimetric analyses (TGA):** TG curve were recorded on a NETZSCH STA 449F3 instrument with a heating rate of  $10 \text{ K}\cdot\text{min}^{-1}$  under the  $\text{N}_2$  atmosphere.

**UV-visible absorption spectroscopy (UV-Vis):** Steady-state UV-Vis spectra were recorded on a Shimadzu 2600 UV/vis spectrometer at RT in the range of 800-200 nm. The  $\text{BaSO}_4$  plate was utilized as a standard which possesses 100% reflectance. The absorption data were then obtained from the reflectance spectra by using the Kubelka-Munk function  $\alpha/S = (1 - R)^2/2R$ , where  $\alpha$  refers to the absorption coefficient,  $S$  refers to the scattering coefficient, and  $R$  refers to the reflectance.<sup>2</sup>

**Photoluminescence Characterization:** Photoluminescence excitation (PLE), photoluminescence emission (PL) spectra, PL decay spectra and photoluminescent quantum yields (PLQYs) were measured on FLS1000 UV/V/NIR fluorescence spectrometer. Temperature-dependent PL spectra were measured on an FLS980 UV/V/NIR fluorescence spectrometer with temperatures ranging from 77 K to 300 K.

**Elemental analysis (EA)** was conducted on a German Elementary Vario MICRO instrument.

**Circular dichroism (CD)** measurements were carried out on a JASCO J-1500 CD spectrometer.

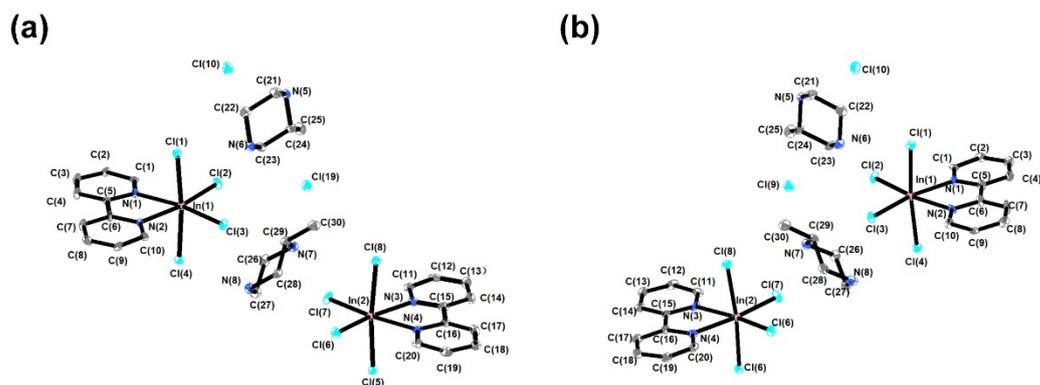
**Circularly polarized luminescence (CPL)** spectra were recorded on a JASCO CPL-300 spectrometer.

#### **Density Functional Theory (DFT) Calculations.**

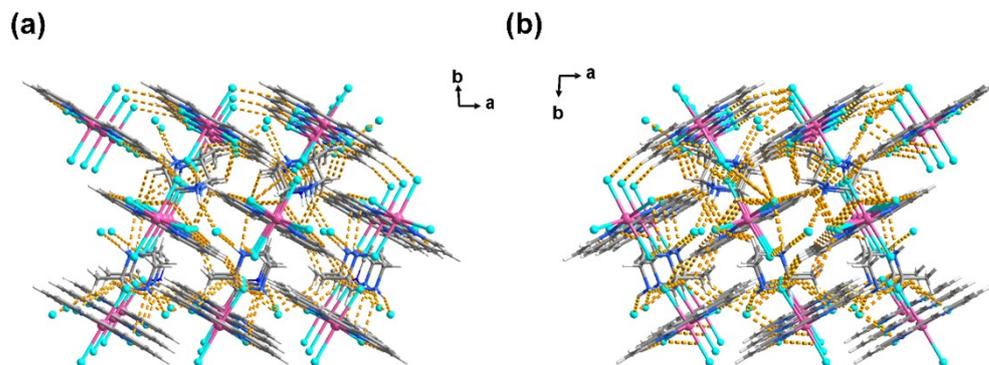
According to the single-crystal structure refinement results, DFT calculations of **R-1** were implemented in the Vienna ab initio simulation package (VASP).<sup>3</sup> The generalized gradient approximation (GGA) for the exchange-correlation term with the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional was applied for electron-electron exchange correlation processes.<sup>4</sup> Projected augmented wave (PAW)

potentials were used with the valence states 2s and 2p for C and N, 5s and 5p for In, 3s and 3p for Cl. To ensure sufficient accuracy, the cut-off energy of 500 eV for the plane wave expansion was chosen, self-consistent field (SCF) computations were set to a convergence criterion of  $1 \times 10^{-5}$  eV, and the force criterion was  $0.02 \text{ eV \AA}^{-1}$ .

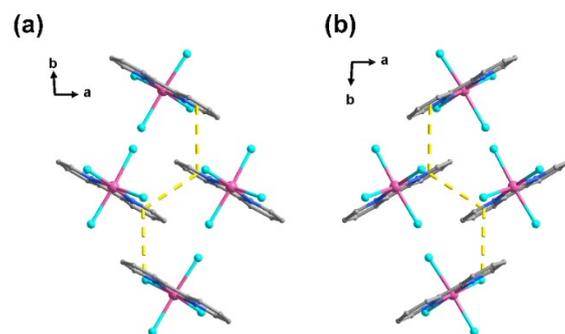
## 2. Figures



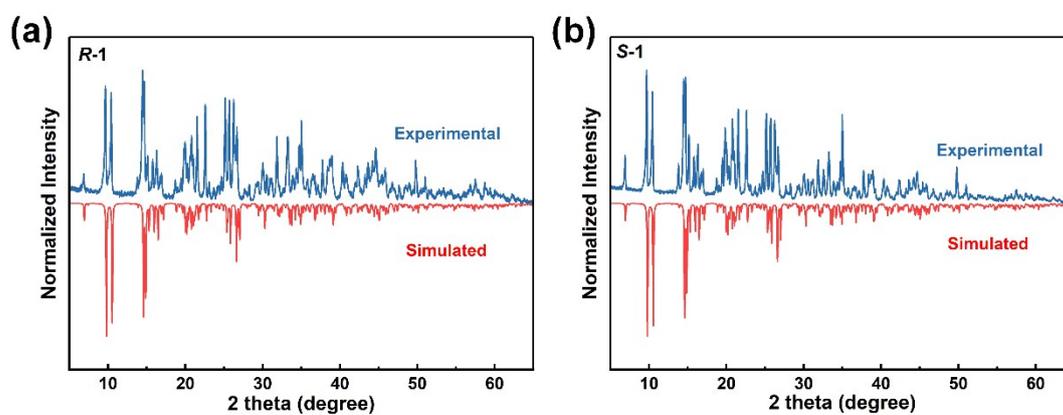
**Figure S1.** ORTEP diagrams of asymmetric units *R*-1 (a) and *S*-1 (b) (50% elliptical probability). Hydrogen atoms are omitted for clarity.



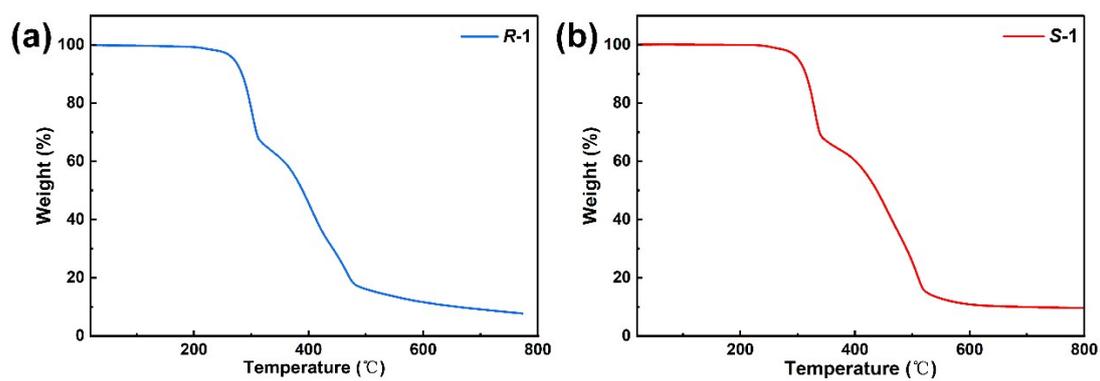
**Figure S2.** Structural stacking diagrams of *R*-1 (a) and *S*-1 (b). Hydrogen bonds are indicated by yellow dashed lines.



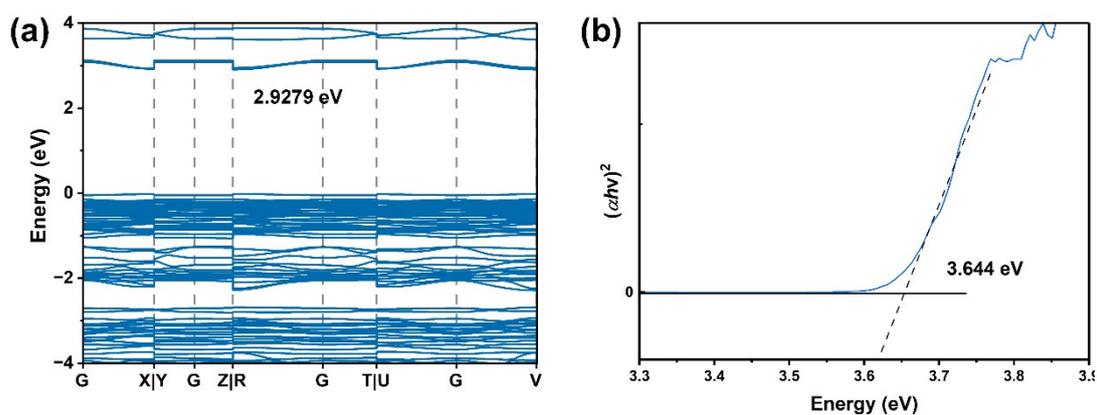
**Figure S3.** View of  $\pi$ - $\pi$  interactions (bright yellow dashed lines) in *R*-1 (a) and *S*-1 (b). Hydrogen atoms are omitted for clarity.



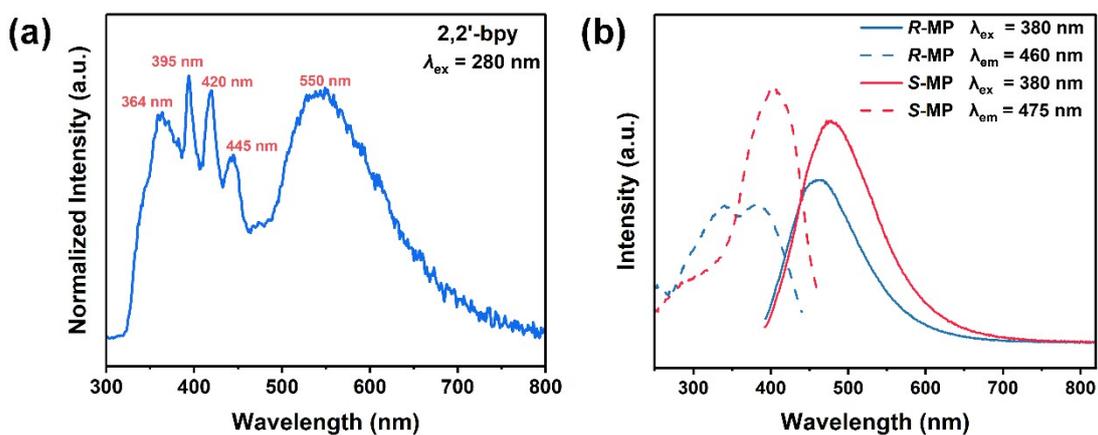
**Figure S4.** Experimental and simulated PXRD patterns of *R*-1 (a) and *S*-1 (b).



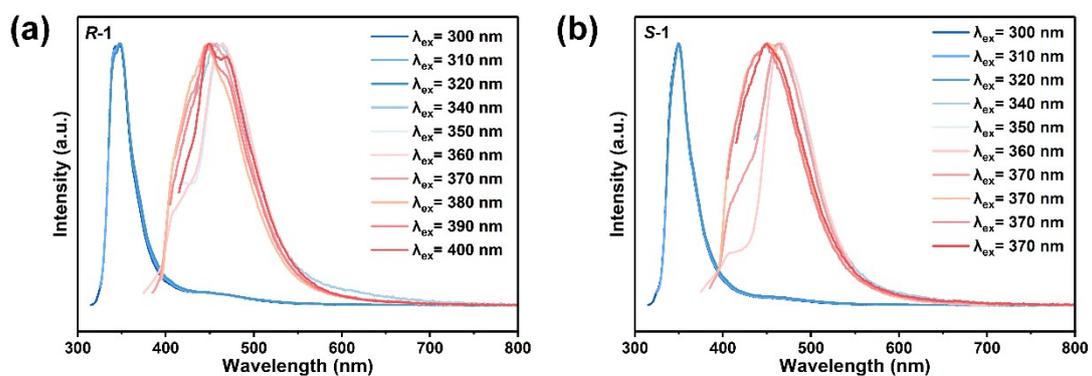
**Figure S5.** TG curves of *R*-1 (a) and *S*-1 (b).



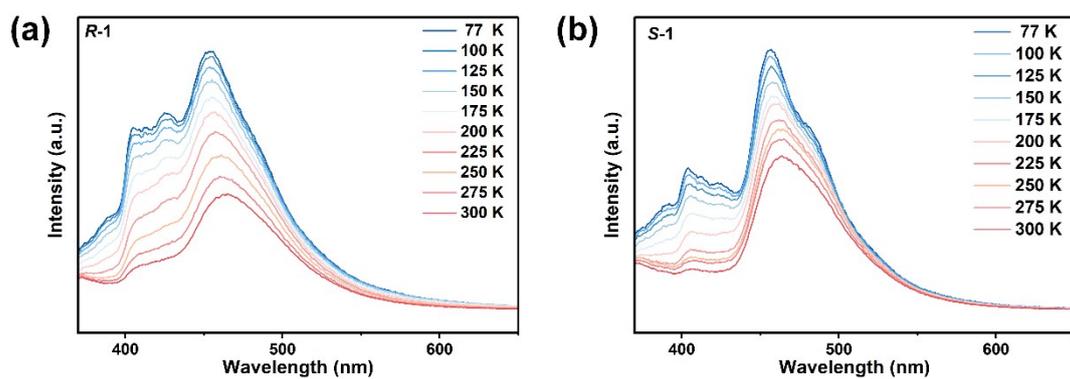
**Figure S6.** (a) Electronic band structures of *R-1*. The calculated bandgap of *R-1* is 2.9279 eV. (b) Tauc plot showing the experimental indirect band gap of *R-1*.



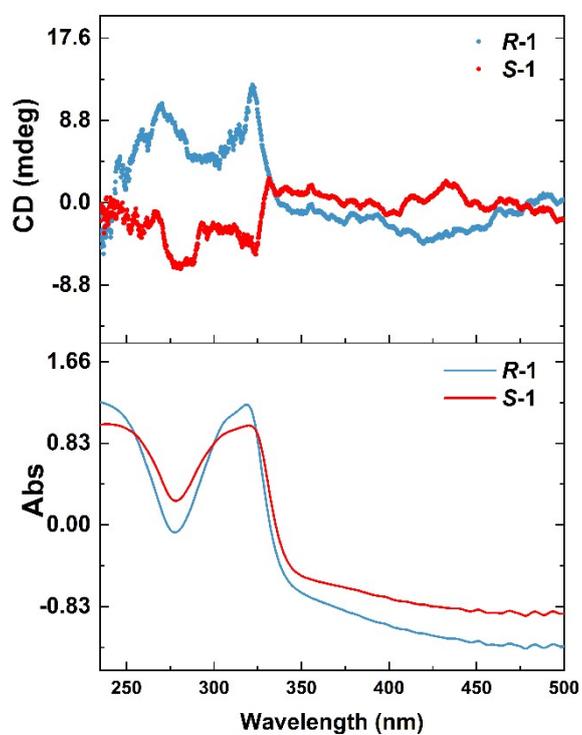
**Figure S7.** (a) Emission spectra of 2,2'-bpy. (b) Excitation and emission spectra of *R/S-MP*.



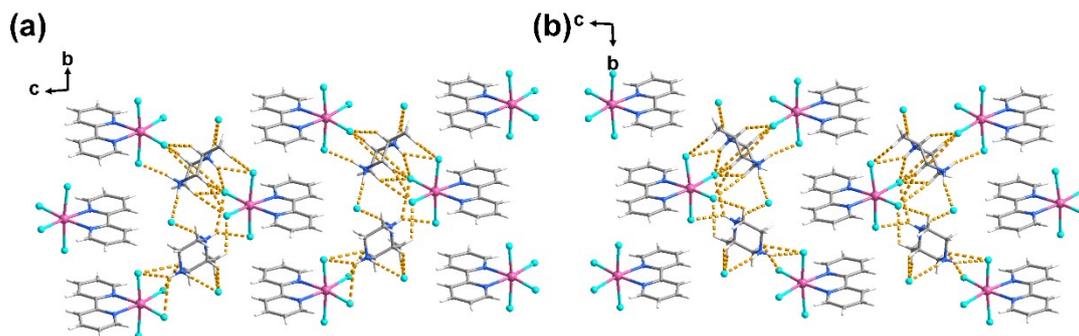
**Figure S8.** Emission spectra of *R-1* (a) and *S-1* (b) at different excitation wavelengths.



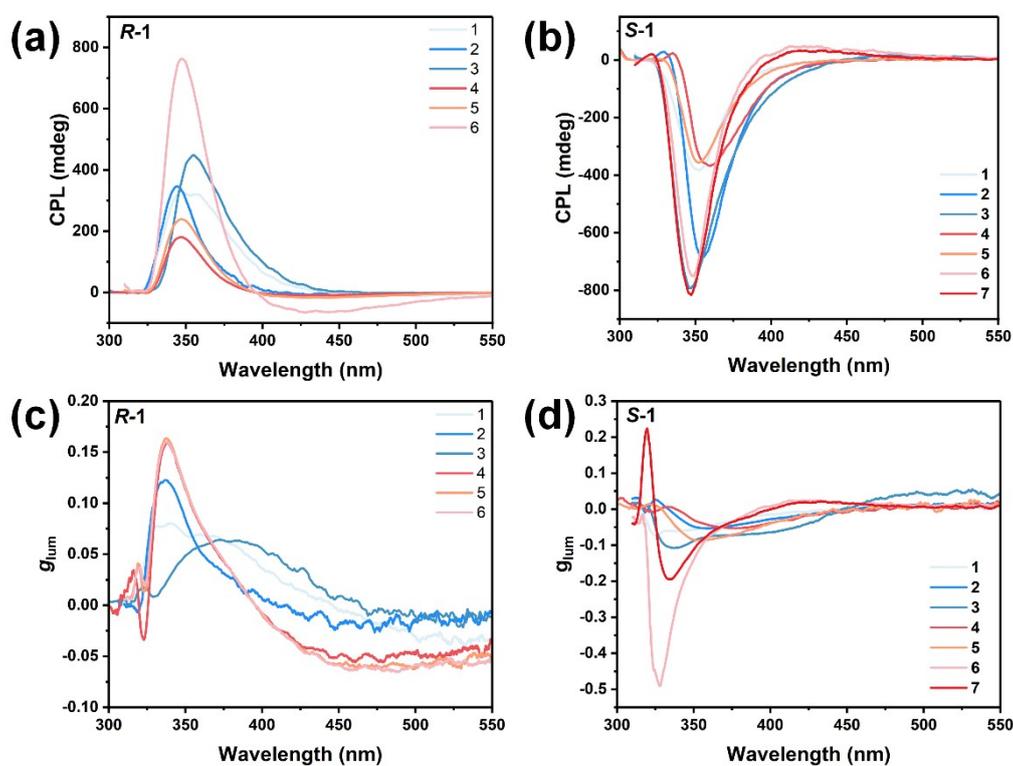
**Figure S9.** Temperature-dependent emission spectra of *R*-1 (a) and *S*-1 (b).



**Figure S10.** Top: Circular dichroism spectra of *R/S*-1. Bottom: Absorption spectra corresponding to the circular dichroism spectra of *R/S*-1.



**Figure S11.** Hydrogen bonds between anions and cations in **R-1** (a) and **S-1** (b).



**Figure S12.** CPL spectra and  $g_{lum}$  plots of synthetic samples from different batches ( $\lambda_{ex} = 280$  nm). (a) CPL spectra of **R-1**; (b) CPL spectra of **S-1**; (c)  $g_{lum}$  values corresponding to **R-1**; (d)  $g_{lum}$  values corresponding to **S-1**.

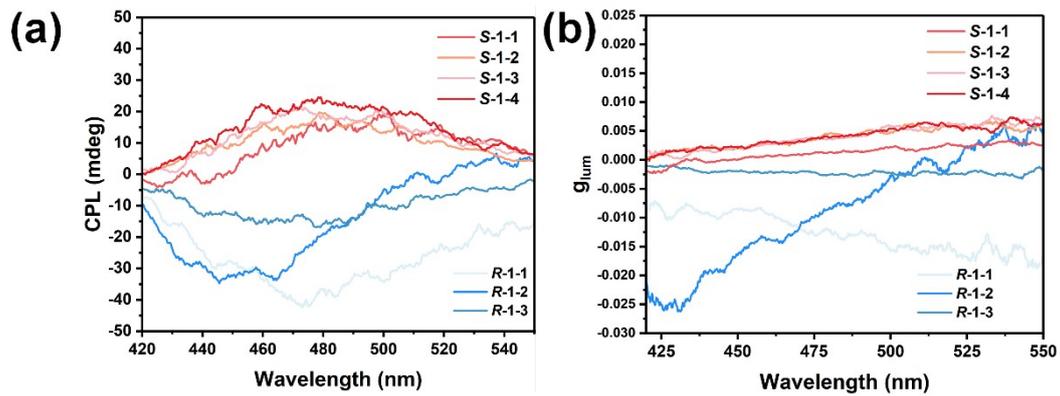


Figure S13. CPL spectra (a) and  $g_{lum}$  plots (b) of synthetic *R/S-1* from different batches ( $\lambda_{ex} = 365$  nm).

### 3. Crystallographic data

**Table S1** Crystallographic data and structure refinement details for **R/S-1**.

Identification code	<b>R-1</b>	<b>S-1</b>
Empirical formula	C <sub>15</sub> H <sub>22</sub> Cl <sub>5</sub> InN <sub>4</sub>	C <sub>15</sub> H <sub>22</sub> Cl <sub>5</sub> InN <sub>4</sub>
Formula weight	550.43	550.43
Temperature/K	100.0	100.0
Crystal system	triclinic	triclinic
Space group	<i>P</i> 1	<i>P</i> 1
<i>a</i> /Å	6.83710(10)	6.8432(2)
<i>b</i> /Å	11.9389(2)	11.9451(3)
<i>c</i> /Å	12.7775(2)	12.7802(3)
<i>α</i> /°	94.1920(10)	94.182(2)
<i>β</i> /°	90.9660(10)	90.902(2)
<i>γ</i> /°	93.0330(10)	93.030(2)
Volume/Å <sup>3</sup>	1038.52(3)	1040.25(5)
<i>Z</i>	2	2
$\rho_{\text{calc}}$ /g/cm <sup>3</sup>	1.760	1.757
$\mu$ /mm <sup>-1</sup>	1.788	1.785
<i>F</i> (000)	548.0	548.0
Crystal size/mm <sup>3</sup>	0.15 × 0.10 × 0.08	0.20 × 0.15 × 0.15
Radiation	MoK $\alpha$ ( $\lambda$ = 0.71073)	MoK $\alpha$ ( $\lambda$ = 0.71073)
Theta range for data collection /°	4.51 to 63.042	3.196 to 61.434
Index ranges	-9 ≤ <i>h</i> ≤ 9, -17 ≤ <i>k</i> ≤ 16, -17 ≤ <i>l</i> ≤ 18	-9 ≤ <i>h</i> ≤ 9, -16 ≤ <i>k</i> ≤ 16, -18 ≤ <i>l</i> ≤ 17
Reflections collected	46902	23573
Independent reflections	10736 [ <i>R</i> <sub>int</sub> = 0.0478, <i>R</i> <sub>sigma</sub> = 0.0457]	9596 [ <i>R</i> <sub>int</sub> = 0.0420, <i>R</i> <sub>sigma</sub> = 0.0528]
Data/restraints/parameters	10736/31/478	9596/37/478
Goodness-of-fit on <i>F</i> <sup>2</sup>	1.042	1.038
Final <i>R</i> indexes [ <i>I</i> ≥ 2σ( <i>I</i> )	<i>R</i> <sub>1</sub> <sup>[a]</sup> = 0.0314, w <i>R</i> <sub>2</sub> <sup>[b]</sup> = 0.0637	<i>R</i> <sub>1</sub> = 0.0375, w <i>R</i> <sub>2</sub> = 0.0916
Final <i>R</i> indexes [all data]	<i>R</i> <sub>1</sub> <sup>[a]</sup> = 0.0377, w <i>R</i> <sub>2</sub> <sup>[b]</sup> = 0.0656	<i>R</i> <sub>1</sub> = 0.0438, w <i>R</i> <sub>2</sub> = 0.0941
Largest diff. peak/hole / e Å <sup>-3</sup>	1.22/-0.58	1.67/-0.59
Flack parameter	-0.05(2)	-0.05(3)
CCDC number	2488488	2488489

$$[a] R_1 = \frac{\sum \|F_o\| - \|F_c\|}{\sum \|F_o\|}, [b] wR_2 = \left[ \frac{\sum w(F_o^2 - F_c^2)^2}{\sum w(F_o^2)^2} \right]^{1/2}$$

**Table S2** Selected bond lengths (Å) and bond angles (°) for **R-1**

<b>R-1</b>			
In(1)-Cl(1)	2.5283(18)	In(2)- Cl(5)	2.5304(19)
In(1)-Cl(2)	2.4585(18)	In(2)- Cl(6)	2.4343(18)
In(1)- Cl(3)	2.4622(19)	In(2)- Cl(7)	2.453(2)
In(1)- Cl(4)	2.5391(19)	In(2)- Cl(8)	2.5221(19)
In(1)- N(1)	2.260(6)	In(2)- N(3)	2.274(6)
In(1)- N(2)	2.272(6)	In(2)- N(4)	2.274(7)
Cl(1)-In(1)-Cl(4)	173.01(6)	N(3)-In(2)-Cl(7)	96.42(18)
Cl(2)-In(1)-Cl(1)	94.47(6)	N(3)-In(2)-Cl(8)	86.29(16)
Cl(2)-In(1)-Cl(3)	98.17(7)	N(4)-In(2)-Cl(5)	89.16(16)
Cl(2)-In(1)-Cl(4)	91.93(6)	N(4)-In(2)-Cl(6)	94.30(17)
Cl(3)-In(1)-Cl(1)	90.42(6)	N(4)-In(2)-Cl(7)	169.55(16)
Cl(3)-In(1)-Cl(4)	91.46(6)	N(4)-In(2)-Cl(8)	87.67(16)
N(1)-In(1)-Cl(1)	89.43(17)	N(4)-In(2)-N(3)	73.1(2)
N(1)-In(1)-Cl(2)	94.50(16)	C(11)-N(3)-In(2)	123.3(5)
N(1)-In(1)-Cl(3)	167.30(15)	C(15)-N(3)-In(2)	117.0(5)
N(1)-In(1)-Cl(4)	87.26(17)	C(16)-N(4)-In(2)	116.6(5)
N(1)-In(1)-N(2)	73.3(2)	C(20)-N(4)-In(2)	122.9(5)
N(2)-In(1)-Cl(1)	85.54(16)	Cl(6)-In(2)-Cl(5)	96.24(6)
N(2)-In(1)-Cl(2)	167.80(16)	Cl(6)-In(2)-Cl(7)	96.15(7)
N(2)-In(1)-Cl(3)	94.03(16)	Cl(6)-In(2)-Cl(8)	90.52(6)
N(2)-In(1)-Cl(4)	87.61(16)	Cl(7)-In(2)-Cl(5)	90.03(7)
C(1)-N(1)-In(1)	123.7(5)	Cl(7)-In(2)-Cl(8)	91.91(7)
C(5)-N(1)-In(1)	116.6(5)	Cl(8)-In(2)-Cl(5)	172.73(7)
C(6)-N(2)-In(1)	116.1(5)	N(3)-In(2)-Cl(5)	86.53(16)
C(10)-N(2)-In(1)	123.3(5)	N(3)-In(2)-Cl(6)	167.13(18)

**Table S3** Selected bond lengths (Å) and bond angles (°) for **S-1**

<b>S-1</b>			
In(1)-Cl(1)	2.527(2)	In(2)-Cl(5)	2.533(2)
In(1)-Cl(2)	2.456(2)	In(2)-Cl(6)	2.438(2)
In(1)-Cl(3)	2.463(2)	In(2)-Cl(7)	2.454(3)
In(1)-Cl(4)	2.540(2)	In(2)-Cl(8)	2.523(2)
In(1)-N(1)	2.254(8)	In(2)-N(3)	2.279(7)
In(1)-N(2)	2.266(8)	In(2)-N(4)	2.273(8)
Cl(1)-In(1)-Cl(4)	172.98(8)	N(3)-In(2)-Cl(7)	96.3(2)
Cl(2)-In(1)-Cl(1)	94.44(8)	N(3)-In(2)-Cl(8)	86.55(19)
Cl(2)-In(1)-Cl(3)	98.23(8)	N(4)-In(2)-Cl(5)	89.05(19)
Cl(2)-In(1)-Cl(4)	91.97(8)	N(4)-In(2)-Cl(6)	94.7(2)
Cl(3)-In(1)-Cl(1)	90.48(8)	N(4)-In(2)-Cl(7)	169.2(2)
Cl(3)-In(1)-Cl(4)	91.44(8)	N(4)-In(2)-Cl(8)	87.75(19)
N(1)-In(1)-Cl(1)	89.7(2)	N(4)-In(2)-N(3)	73.0(3)
N(1)-In(1)-Cl(2)	94.2(2)	C(11)-N(3)-In(2)	123.5(7)
N(1)-In(1)-Cl(3)	167.55(19)	C(15)-N(3)-In(2)	115.8(6)
N(1)-In(1)-Cl(4)	86.99(19)	C(16)-N(4)-In(2)	117.5(6)
N(1)-In(1)-N(2)	73.4(3)	C(20)-N(4)-In(2)	122.1(7)
N(2)-In(1)-Cl(1)	85.94(19)	Cl(6)-In(2)-Cl(5)	96.32(8)
N(2)-In(1)-Cl(2)	167.5(2)	Cl(6)-In(2)-Cl(7)	96.11(9)
N(2)-In(1)-Cl(3)	94.2(2)	Cl(6)-In(2)-Cl(8)	90.38(8)
N(2)-In(1)-Cl(4)	87.18(19)	Cl(7)-In(2)-Cl(5)	89.97(8)
C(1)-N(1)-In(1)	124.5(6)	Cl(7)-In(2)-Cl(8)	91.96(8)
C(5)-N(1)-In(1)	115.9(6)	Cl(8)-In(2)-Cl(5)	172.79(8)
C(6)-N(2)-In(1)	117.3(6)	N(3)-In(2)-Cl(5)	86.34(18)
C(10)-N(2)-In(1)	123.3(6)	N(3)-In(2)-Cl(6)	167.4(2)

**Table S4** Hydrogen bonds for **R-1**.

D-H...A	d(D-H)/Å	d(H...A)/Å	d(D-A)/Å	<(DHA)/°
C(1)-H(1A)···Cl(1) <sup>1</sup>	0.95	2.93	3.631(7)	131.9
C(1)-H(1A)···Cl(2)	0.95	2.80	3.466(8)	127.8
C(9)-H(9A)···Cl(8) <sup>2</sup>	0.95	2.97	3.550(7)	120.9
C(10)-H(10A)···Cl(3)	0.95	2.78	3.450(7)	128.2
C(11)-H(11A)···Cl(7)	0.95	2.89	3.540(8)	127.0
C(19)-H(19A)···Cl(1) <sup>3</sup>	0.95	2.98	3.587(9)	123.3
C(20)-H(20A)···Cl(6)	0.95	2.78	3.441(8)	127.4
N(5)-H(5A)···Cl(5)	0.948(19)	2.46(5)	3.186(5)	134(5)
N(5)-H(5A)···Cl(7)	0.948(19)	2.87(5)	3.600(5)	134(5)
N(5)-H(5B)···Cl(10) <sup>1</sup>	0.948(19)	2.18(2)	3.106(5)	164(5)
N(6)-H(6A)···Cl(1) <sup>1</sup>	0.951(19)	2.41(4)	3.202(5)	141(5)
N(6)-H(6A)···Cl(3) <sup>1</sup>	0.951(19)	2.95(5)	3.519(5)	119(4)
N(6)-H(6B)···Cl(9)	0.951(19)	2.20(3)	3.103(5)	159(5)
C(23)-H(23A)···Cl(1) <sup>1</sup>	0.99	2.97	3.484(6)	113.1
C(23)-H(23A)···Cl(2) <sup>1</sup>	0.99	2.96	3.480(6)	113.7
C(24)-H(24A)···Cl(9)	1.00	2.70	3.494(6)	136.7
C(25)-H(25A)···Cl(6) <sup>1</sup>	0.98	2.81	3.391(5)	119.0
C(25)-H(25C)···Cl(9) <sup>1</sup>	0.98	2.76	3.561(6)	138.8
N(7)-H(7C)···Cl(7) <sup>4</sup>	0.948(19)	2.89(5)	3.574(7)	130(5)
N(7)-H(7C)···Cl(8) <sup>4</sup>	0.948(19)	2.50(4)	3.283(7)	140(5)
N(7)-H(7D)···Cl(9)	0.950(19)	2.10(3)	3.027(6)	164(6)
N(8)-H(8C)···Cl(4)	0.949(19)	2.25(3)	3.166(6)	163(6)
N(8)-H(8D)···Cl(10) <sup>4</sup>	0.949(19)	2.12(3)	3.036(6)	162(5)
C(26)-H(26A)···Cl(7) <sup>4</sup>	0.99	2.84	3.411(8)	117.4
C(26)-H(26B)···Cl(3) <sup>1</sup>	0.99	2.93	3.434(8)	112.3
C(27)-H(27B)···Cl(6) <sup>4</sup>	0.99	2.96	3.824(8)	145.8
C(27)-H(27B)···Cl(7) <sup>4</sup>	0.99	2.87	3.550(9)	126.3
C(28)-H(28A)···Cl(6) <sup>4</sup>	0.99	2.79	3.701(8)	152.5
C(28)-H(28A)···Cl(7) <sup>5</sup>	0.99	2.84	3.366(8)	114.3
C(28)-H(28B)···Cl(3)	0.99	2.82	3.383(8)	116.4
C(29)-H(29A)···Cl(3)	1.00	2.75	3.439(8)	126.2
C(30)-H(30A)···Cl(9) <sup>6</sup>	0.98	2.90	3.775(6)	149.1
C(30)-H(30C)···Cl(8) <sup>4</sup>	0.98	2.77	3.505(6)	131.9

Symmetry transformations used to generate equivalent atoms: #1 -1+x, +y, +z; #2 1+x, -1+y, 1+z;

#3 +x, +y, -1+z; #4 +x, -1+y, +z; #5 1+x, -1+y, +z; #6 1+x, +y, +z

**Table S5** Hydrogen bonds for **S-1**.

D-H···A	d(D-H)/Å	d(H···A)/Å	d(D-A)/Å	<(DHA)/°
C(1)-H(1A)···Cl(1) <sup>1</sup>	0.95	2.94	3.641(9)	132.0
C(1)-H(1A)···Cl(2)	0.95	2.80	3.461(10)	127.7
C(9)-H(9A)···Cl(8) <sup>2</sup>	0.95	2.96	3.548(9)	121.4
C(10)-H(10A)···Cl(3)	0.95	2.79	3.453(9)	128.0
C(11)-H(11A)···Cl(7)	0.95	2.88	3.540(10)	127.2
C(19)-H(19A)···Cl(1) <sup>3</sup>	0.95	2.97	3.581(11)	123.0
C(20)-H(20A)···Cl(6)	0.95	2.78	3.443(10)	127.6
N(5)-H(5A)···Cl(5)	0.95(3)	2.44(6)	3.179(7)	135(7)
N(5)-H(5A)···Cl(7)	0.95(3)	2.91(6)	3.594(6)	130(6)
N(5)-H(5B)···Cl(10) <sup>1</sup>	0.95(3)	2.20(3)	3.118(7)	164(7)
N(6)-H(6A)···Cl(1) <sup>1</sup>	0.95(3)	2.30(4)	3.203(7)	157(7)
N(6)-H(6B)···Cl(9)	0.95(3)	2.26(5)	3.101(7)	147(7)
C(21)-H(21B)···Cl(6)	0.99	2.98	3.456(8)	110.9
C(23)-H(23B)···Cl(1) <sup>1</sup>	0.99	2.97	3.477(8)	113.1
C(23)-H(23B)···Cl(2) <sup>1</sup>	0.99	2.97	3.478(8)	113.3
C(24)-H(24A)···Cl(9)	1.00	2.69	3.491(8)	137.2
C(25)-H(25A)···Cl(6) <sup>1</sup>	0.98	2.87	3.398(7)	114.7
C(25)-H(25B)···Cl(9) <sup>1</sup>	0.98	2.76	3.556(8)	139.0
N(7)-H(7C)···Cl(7) <sup>4</sup>	0.95(3)	2.91(7)	3.582(8)	129(6)
N(7)-H(7C)···Cl(8) <sup>4</sup>	0.95(3)	2.49(5)	3.280(8)	141(6)
N(7)-H(7D)···Cl(9)	0.95(3)	2.10(4)	3.024(7)	163(7)
N(8)-H(8C)···Cl(4)	0.95(3)	2.26(4)	3.171(9)	160(8)
N(8)-H(8D)···Cl(10) <sup>4</sup>	0.95(3)	2.11(3)	3.033(8)	165(8)
C(26)-H(26A)···Cl(3) <sup>1</sup>	0.99	2.95	3.447(9)	112.4
C(26)-H(26B)···Cl(7) <sup>4</sup>	0.99	2.84	3.414(9)	117.7
C(27)-H(27A)···Cl(6) <sup>4</sup>	0.99	2.96	3.821(11)	146.0
C(27)-H(27A)···Cl(7) <sup>4</sup>	0.99	2.89	3.560(11)	126.0
C(28)-H(28A)···Cl(3)	0.99	2.83	3.386(10)	116.4
C(28)-H(28B)···Cl(6) <sup>4</sup>	0.99	2.80	3.703(10)	152.6
C(28)-H(28B)···Cl(7) <sup>5</sup>	0.99	2.83	3.358(10)	114.4
C(29)-H(29A)···Cl(3)	1.00	2.75	3.441(9)	126.6
C(30)-H(30A)···Cl(9) <sup>6</sup>	0.98	2.89	3.776(7)	150.7
C(30)-H(30B)···Cl(8) <sup>4</sup>	0.98	2.81	3.517(8)	129.9

Symmetry transformations used to generate equivalent atoms: #1 1+x, +y, +z; #2 -1+x, 1+y, -1+z;  
#3 +x, +y, 1+z; #4 +x, 1+y, +z; #5 -1+x, 1+y, +z; #6 -1+x, +y, +z

**Table S6** The  $\pi$ - $\pi$  interaction data for **R-1**.

<b>Cg(I)···Cg(J)</b>	<b>ARU(J)</b>	<b>Cg···Cg(Å)</b>	<b><math>\alpha</math> (°)</b>	<b><math>\beta</math> (°)</b>	<b><math>\gamma</math> (°)</b>
Cg (2) → Cg (6)	1556.02	3.877(4)	1.4(3)	30.6	30.9
Cg (5) → Cg (6)	1455.02	3.772(4)	2.2(3)	27.2	27.9
Cg (6) → Cg (2)	1554.01	3.877(4)	1.4(3)	30.9	30.6
Cg (6) → Cg (5)	1655.02	3.772(4)	2.2(3)	27.9	27.2
Cg(2): N(2) → C(6) → C(7) → C(8) → C(9) → C(10); Cg(4): N(3) → C(11) → C(12) → C(13) → C(14) → C(15)					
Cg(5): N(4) → C(16) → C(17) → C(18) → C(19) → C(20); Cg(6): N(5) → C(21) → C(22) → N(6) → C(23) → C(24)					
[1655] = 1+x, y, z; [1455] = -1+x, y, z; [1556] = x, y, 1+z; [1554] = x, y, -1+z					

**Table S7** The  $\pi$ - $\pi$  interaction data for **S-1**.

<b>Cg(I)···Cg(J)</b>	<b>ARU(J)</b>	<b>Cg···Cg(Å)</b>	<b><math>\alpha</math> (°)</b>	<b><math>\beta</math> (°)</b>	<b><math>\gamma</math> (°)</b>
Cg (2) → Cg (6)	1554.02	3.883(5)	0.9(4)	30.7	30.9
Cg (5) → Cg (6)	1655.02	3.778(5)	2.2(4)	26.9	28.1
Cg (6) → Cg (2)	1556.01	3.883(5)	0.9(4)	30.9	30.7
Cg (6) → Cg (5)	1455.02	3.778(5)	2.2(4)	28.1	26.9
Cg(2): N(2) → C(6) → C(7) → C(8) → C(9) → C(10); Cg(4): N(3) → C(11) → C(12) → C(13) → C(14) → C(15)					
Cg(5): N(4) → C(16) → C(17) → C(18) → C(19) → C(20); Cg(6): N(5) → C(21) → C(22) → N(6) → C(23) → C(24)					
[1455] = -1+x, y, z; [1655] = 1+x, y, z; [1554] = x, y, -1+ z; [1556] = x, y, 1+z					

**Table S8** Summary of chiral OIHMs with luminescent switching properties

Material	Emission Peak (Off- State)/nm	Emission Peak (On- State)/nm	Switching Method	CPL Switching	Referenc e
<i>(R/S-LH<sub>2</sub>)ZnBr<sub>4</sub></i>	505	599	pH	Yes	5
<i>(R/S-LH1)<sub>2</sub>ZnBr<sub>4</sub></i>		645			
<i>(R-/S-Br-MBA)<sub>3</sub>MnBr<sub>5</sub></i>	520	654	Temperature	Yes	6
<i>R/S-PbSnBr·H<sub>2</sub>O</i>	580	640	Humidity	NO	7
<i>R/S-PbSnBr</i>					
<i>(R/S-2-HMM)<sub>3</sub>SbCl<sub>6</sub></i>	590	630	Temperature	NO	8
<i>(R,R/S,S-DCDA)<sub>3</sub>Sb<sub>2</sub>Cl<sub>12</sub></i>	596	703	Temperature	NO	9
<i>(R/S-3AP)<sub>4</sub>SbCl<sub>11</sub>·DMF</i>	627	667	Solvent Polarity	Yes	10
<i>(R/S-3AP)<sub>4</sub>SbCl<sub>11</sub>·ACN</i>					
<i>R/S-Tb<sub>0.83</sub>Eu<sub>0.1</sub></i>	548	612	Excitation wavelength	Yes	11
<i>RMPZMB@RED</i>	525	625	Humidity	Yes	12.
<i>RMPZMB@GREEN</i>					
<i>R/S-O-H<sub>2</sub>O</i>	515	540 635	Solvent Polarity	Yes	13
<i>R/S-1</i>	345	460	Excitation wavelength	Yes	This Work

## 4. References

1. G. M. Sheldrick, *Acta Crystallogr. Sect. C-Struct. Chem.*, 2015, **71**, 3-8.
2. W. M. Wendlandt and H. G. Hecht, *New York*, 1966.
3. G. Kresse and J. Hafner, *Phys. Rev. B*, 1993, **48**, 13115-13118.
4. G. Kresse and J. Furthmuller, *Phys. Rev. B*, 1996, **54**, 11169-11186.
5. D.-Y. Liu, L.-Y. Xiong, X.-Y. Dong, Z. Han, H.-L. Liu and S.-Q. Zang, *Angew. Chem. Int. Ed.*, 2024, e202410416.
6. T. Zhang, H. Kang, B. Li, J. Zhou, P. Zhao, T. Zhao, X. Li and S. Jiang, *J. Mater. Chem. C*, 2023, **11**, 5461-5468.
7. Y. Wei, C. Li, Y. Li, Z. Luo, X. Wu, Y. Liu, L. Zhang, X. He, W. Wang and Z. Quan, *Angew. Chem. Int. Ed.*, 2022, **61**, e202212685.
8. W. Wang, C.-D. Liu, X.-B. Han, C.-Q. Jing, C.-Y. Chai, C.-C. Fan, M.-L. Jin, J.-M. Zhang and W. Zhang, *ACS Mater. Lett.*, 2023, **6**, 203-211.
9. C.-Y. Chai, X.-B. Han, C.-D. Liu, C.-C. Fan, B.-D. Liang and W. Zhang, *J. Phys. Chem. Lett.*, 2023, **14**, 4063-4070.
10. Min Tao, Rui Cao, Shuo Sun, Hongxu Li and Y. Xiao\*, *Chem. Eur. J.*, 2025, e202404470.
11. C. Chen, Z. Luo, M. Zhang, X. Li, T. Zhang and J. Chen, *Dalton Trans.*, 2025, **54**, 12658-12666.
12. X. Yu, S. Zhong, Z. Guo, J. Guan, H. Tang, X. He, Y. Chen and S. Pan, *J. Mater. Chem. C*, 2025, **13**, 2190-2197.
13. C. Li, Y. Wei, Y. Li, Z. Luo, Y. Liu, M. He, Y. Zhang, X. He, X. Chang and Z. Quan, *Small*, 2024, 2400338.