

## Supporting Information

### Experimental detail

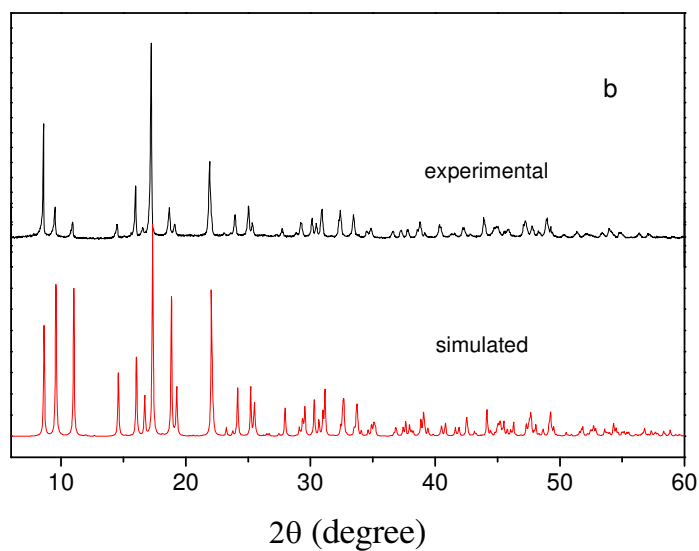
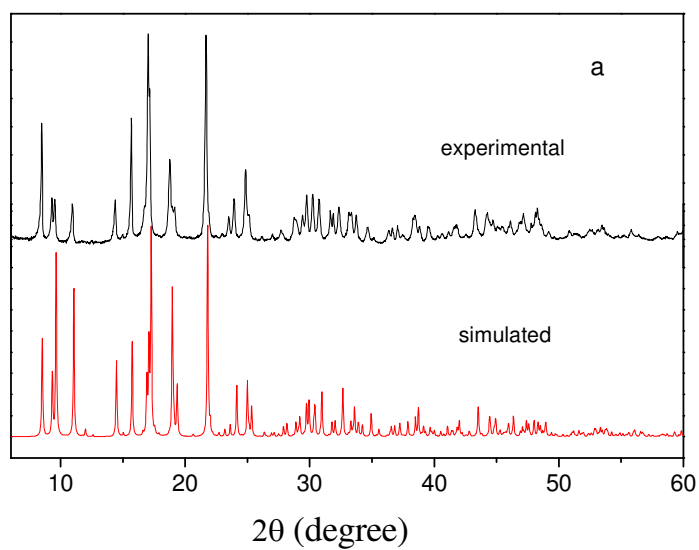
**General:** All reagents were commercially available and were used without further purification.

The codoping ratio was confirmed by ICP and elemental analysis. Elemental analyses of C, H and N were performed with an Elementar Vario EL analyzer. The IR spectra were recorded with a Nicolet Avatar 360 FTIR spectrometer by using the KBr pellet technique. The inductively coupled plasma (ICP) analysis was performed with a JY ULTIMA spectrometer. The phase purity was confirmed by powder X-ray diffraction carried out on a PANalytical X'Pert PRO MPD diffractometer for Cu K $\alpha$  radiation ( $\lambda = 1.5406 \text{ \AA}$ ), with a scan speed of  $2^\circ \cdot \text{min}^{-1}$  and a step size of  $0.02^\circ$  in  $2\theta$ . The simulated PXRD patterns were obtained from the single-crystal X-ray diffraction data. The fluorescence spectra were recorded on a SHIMADZU RT-5301PC spectrofluorophotometer at room temperature. The lifetime was measured at room temperature on LifeSpec-Red Picosecond lifetime spectrometer (Edinburgh Instruments) for complex **1**, and FLS920 Steady State & Time-resolved Fluorescence Spectrometer (Edinburgh Instruments) for complexes **3** and **5**, respectively.

**Syntheses of 1-5:** A mixture of H<sub>3</sub>imdc (0.25 mmol, 0.0390 g), LnCl<sub>3</sub>·nH<sub>2</sub>O (0.025 mmol; Ln = La, n = 7, 0.0093 g; Pr, n = 6, 0.0089 g; Eu, n = 6, 0.0092 g; Gd, n = 6, 0.0093 g; Tb, n = 6, 0.0093 g), Zn(CH<sub>3</sub>COO)<sub>2</sub>·2H<sub>2</sub>O (0.1 mmol, 0.0220 g), and HAc-NaAc buffer (pH = 5.7, 4mL) was sealed in a 25-mL stainless steel reactor with a Teflon liner and heated at 180°C for 96 h. Rod like crystals of **1** (0.023 g, 64.3%), **2** (0.016 g, 44.7 %), **3** (0.019 g, 52.7 %), **4** (0.016 g, 44.2 %), and **5** (0.018 g, 49.6 %) were obtained. Elem anal. Calcd for C<sub>64</sub>H<sub>74</sub>La<sub>2</sub>Zn<sub>8</sub>N<sub>32</sub>O<sub>48</sub>: C, 26.87; H, 2.61; N, 15.67. Found: C, 27.03; H, 2.64; N, 15.54. IR (cm<sup>-1</sup>, KBr): 3448 s, 2878 m, 1598 vs, 1474 m, 1386 s, 1249 m, 1102 m, 1075 m, 822 m, 795 m, 655 m, 534 m. Elem anal. Calcd for C<sub>64</sub>H<sub>74</sub>Pr<sub>2</sub>Zn<sub>8</sub>N<sub>32</sub>O<sub>48</sub>: C, 26.83; H, 2.60; N, 15.65. Found: C, 27.14; H, 2.33; N, 15.64. IR (cm<sup>-1</sup>, KBr): 3464 s, 2922 m, 1631 s, 1544 m, 1469 s, 1384 m, 1249 m, 1075 m, 825 m, 795 m, 654 m, 531 m. Elem anal. Calcd for C<sub>64</sub>H<sub>74</sub>Eu<sub>2</sub>Zn<sub>8</sub>N<sub>32</sub>O<sub>48</sub>: C, 26.63; H, 2.58; N, 15.53. Found: C, 26.88; H, 2.41; N, 15.62. IR (cm<sup>-1</sup>, KBr): 3448 s, 2850 m, 1589 vs, 1476 m, 1389 vs, 1251 m, 1104 m, 1071 m, 826 m, 793 m, 655 m. Elem anal. Calcd for C<sub>64</sub>H<sub>74</sub>Gd<sub>2</sub>Zn<sub>8</sub>N<sub>32</sub>O<sub>48</sub>: C, 26.53; H, 2.57; N,

15.47. Found: C, 26.86; H, 2.48; N, 15.52. IR ( $\text{cm}^{-1}$ , KBr): 3451 vs, 2940 m, 1604 m, 1544 m, 1469 m, 1384 m, 1249 m, 1103 m, 954 w, 827 m, 654 m. Elem anal. Calcd for  $\text{C}_{64}\text{H}_{74}\text{Tb}_2\text{Zn}_8\text{N}_{32}\text{O}_{48}$ : C, 26.50; H, 2.57; N, 15.45. Found: C, 26.70; H, 2.30; N, 15.41. IR ( $\text{cm}^{-1}$ , KBr): 3432 s, 2946 m, 1598 vs, 1470 s, 1385 s, 1329 m, 1251 m, 1103 m, 1074 m, 1017 w, 951 w, 827 m, 795 m, 654 m, 533 m.

No crystal was obtained when the buffer solution was replaced by water.



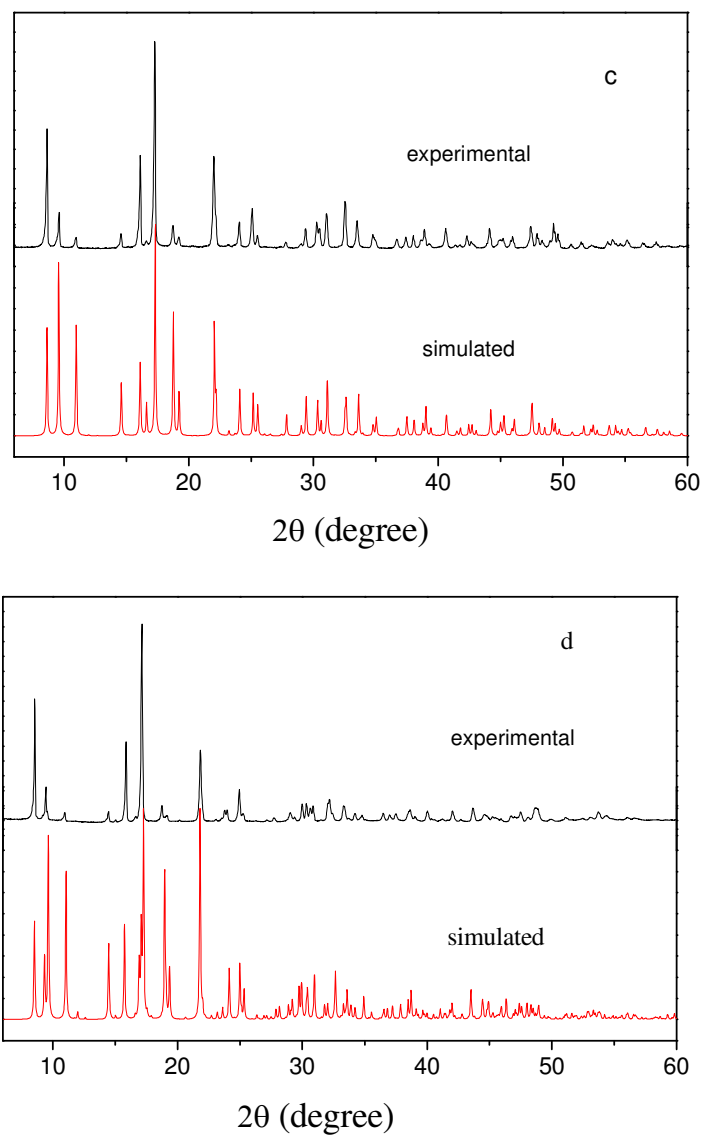


Figure S1 The PXRD patterns for complexes **1** (a), **3** (b), **5** (c) and the codoped complex (d).

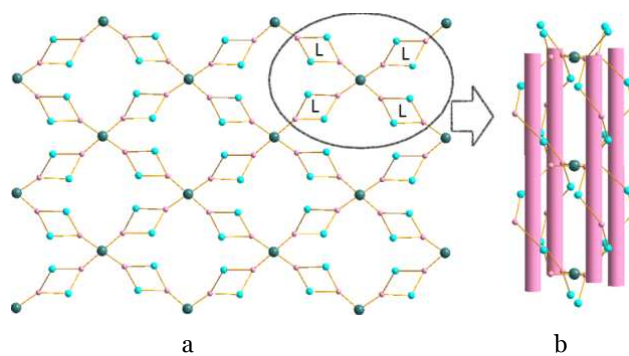


Figure S2 a) The simplified 3-D structure for complex **3**. b) Left-handed helical chains along the c-axis. cyan: Zn<sup>2+</sup>, pink: ligand, dark green: Eu<sup>3+</sup>.

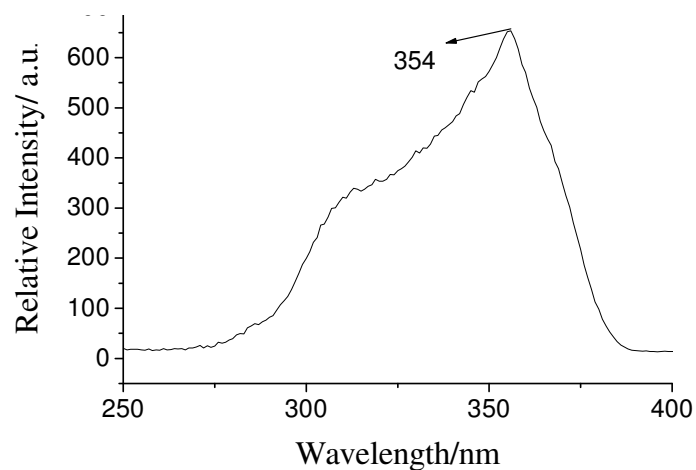


Figure S3 Excitation spectra of complex **1**,  $\lambda_{em} = 429$  nm

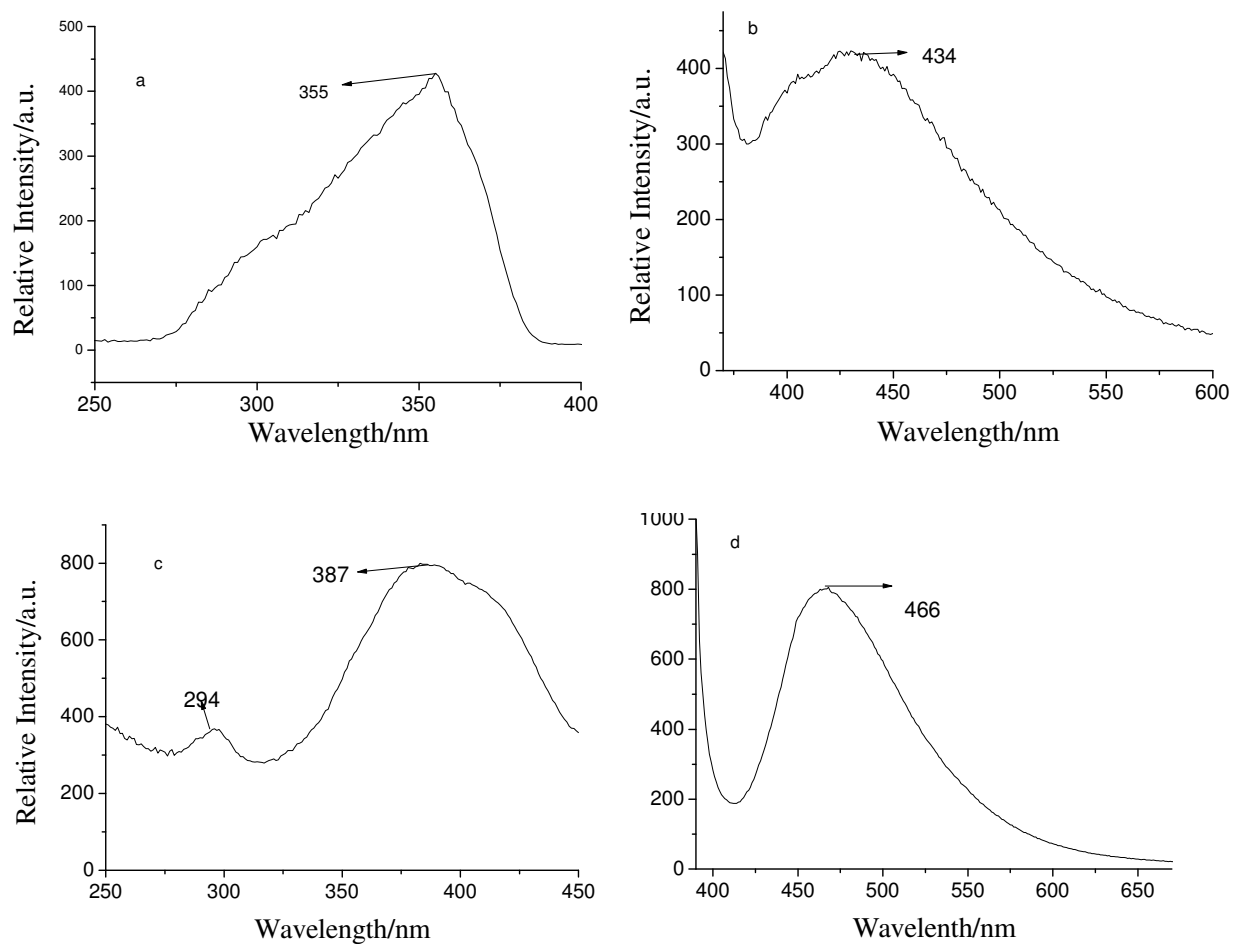


Figure S4 Excitation spectra (a, c) and emission spectra (b, d) for the ligands Him and H<sub>3</sub>imdc, respectively.

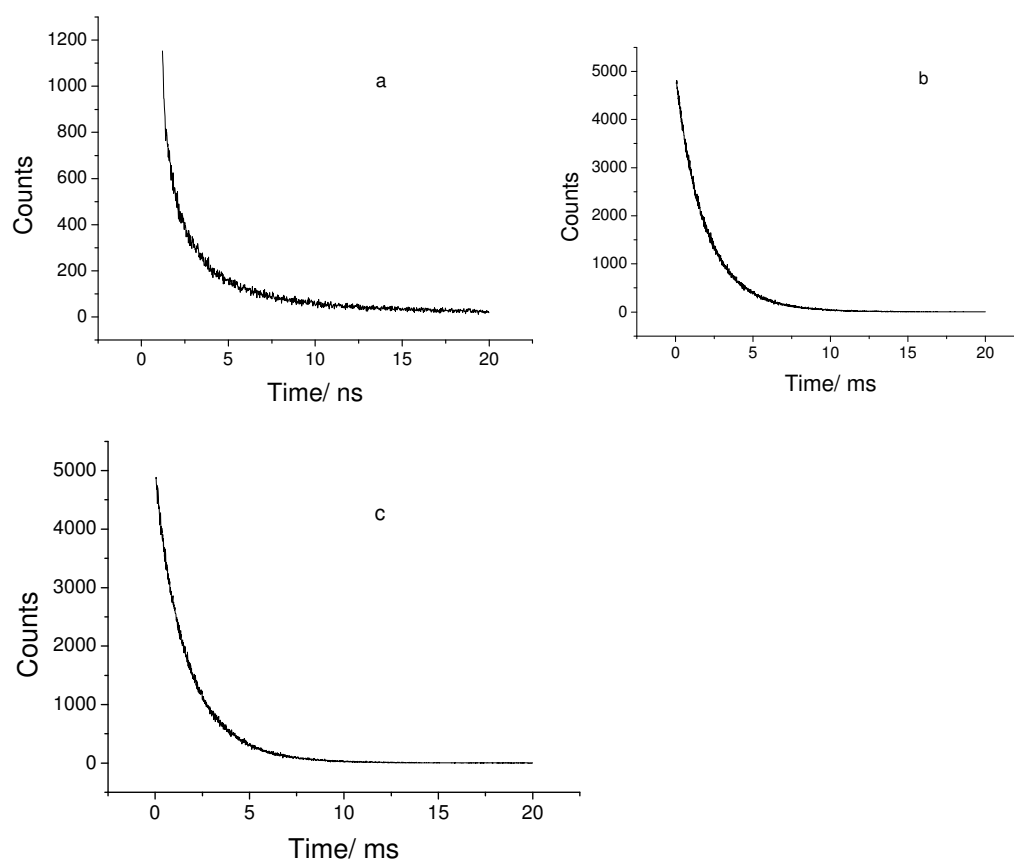


Figure S5 Decay profiles of the complexes **1** (a), **3** (b), and **5(c)**.

Table S1 CIE chromaticity coordinates for the La-Zn heterometallic compounds doped with  $\text{Eu}^{3+}$  and  $\text{Tb}^{3+}$  of different molar ratio excited at different wavelength.

molar ratio of La:Eu:Tb	CIE chromaticity coordinates	
	excited at 350 nm	excited at 294 nm
42%: 21%: 37%	(0.3116, 0.3325)	(0.3626, 0.5171)
40%: 27%: 33%	(0.3461, 0.3451)	(0.3769, 0.5100)
48%: 15%: 37%	(0.2976, 0.3345)	(0.3566, 0.5164)
46%: 22%: 33%	(0.3234, 0.3345)	(0.3682, 0.5122)
50%: 17%: 33%	(0.3134, 0.3417)	(0.3608, 0.5159)
50%: 22%: 28%	(0.3285, 0.3306)	(0.3736, 0.5096)
54%: 23%: 23%	(0.3040, 0.2881)	(0.386, 0.5048)
58%: 21%: 21%	(0.3010, 0.2850)	(0.3783, 0.5022)
70%: 15%: 15%	(0.2748, 0.2793)	(0.3739, 0.5065)
81%: 10%: 9%	(0.2507, 0.2460)	(0.3694, 0.5074)
91%: 4%: 5%	(0.2227, 0.2238)	(0.3626, 0.5041)