## Supplementary Information

## Visible light sensitized near-infrared luminescence of ytterbium via

## ILCT states in quadruple-stranded helicates

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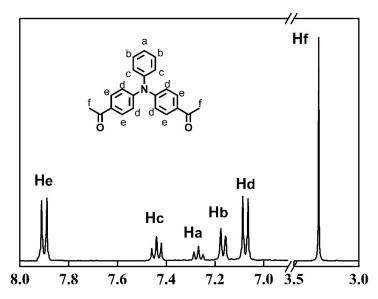


Figure S1. <sup>1</sup>H NMR spectrum of 4,4'-diacetyl triphenylamine in DMSO-*d6*.

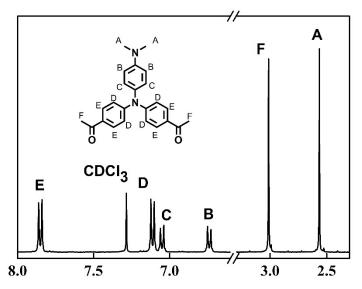


Figure S2. <sup>1</sup>H NMR spectrum of 4-(N,N-Dimethylamino)-4',4"-diacetyltriphenylamine in CDCl<sub>3</sub>

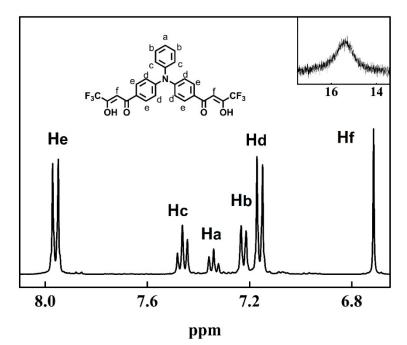


Figure S3. <sup>1</sup>H NMR spectrum of L<sup>1</sup> in DMSO-*d6*.

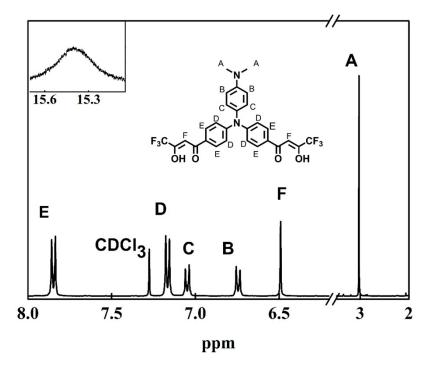


Figure S4. <sup>1</sup>H NMR spectrum of L<sup>2</sup> in CDCl<sub>3</sub>.

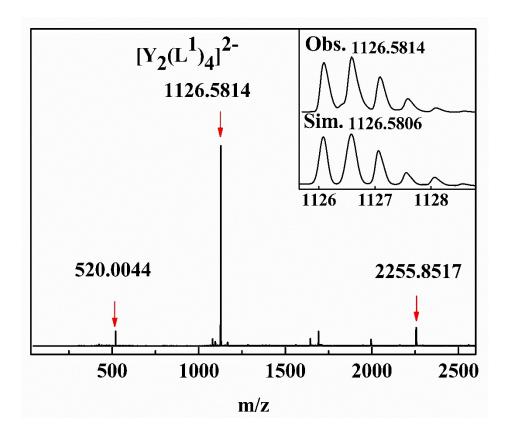


Figure S5. ESI-TOF-MS of  $[Y_2(L^1)_4]_{.}$ 

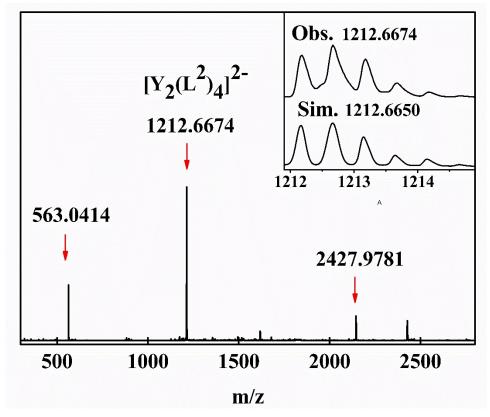
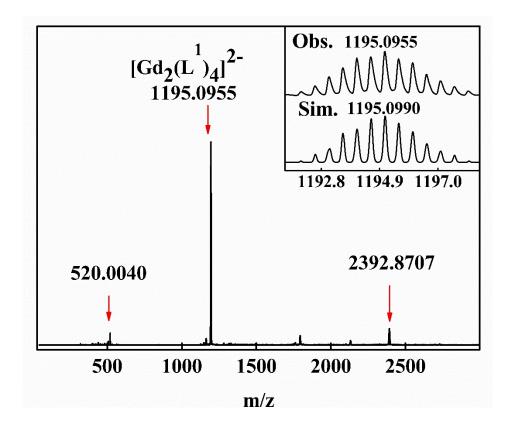


Figure S6. ESI-TOF-MS of  $[Y_2(L^2)_4]$ .





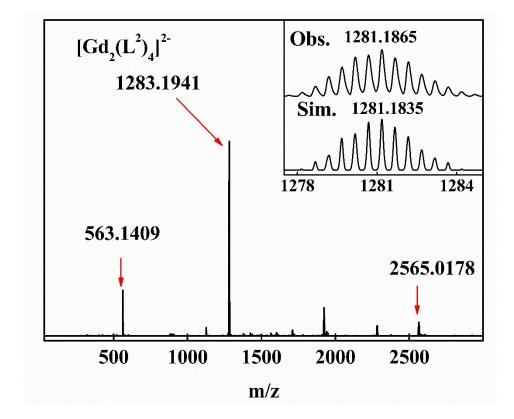
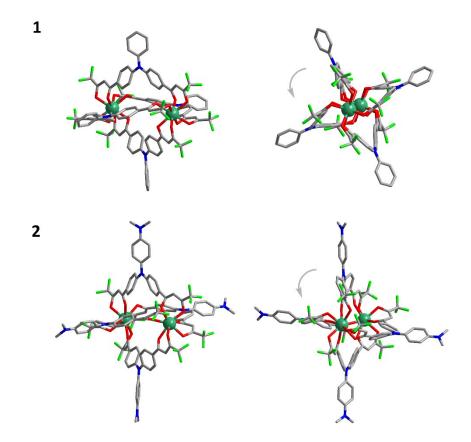
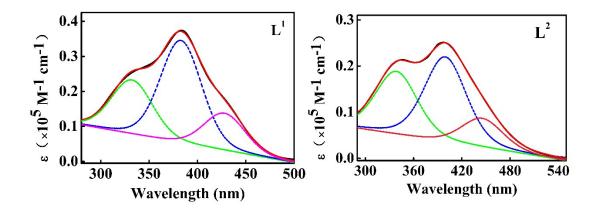


Figure S8. ESI-TOF-MS of  $[Gd_2(L^2)_4]$ .



**Figure S9.** Side view (left) and top view (right) of the structures of complexes 1 and 2 optimized by Sparkle/PM6 model (For clearance, only right-handed  $\Delta$ - $\Delta$  helix are presented).



**Figure S10**. Experimental UV/Vis absorption spectra (solid black line), respective Gaussian deconvolutions (dashed lines), and best fit (solid red line): R=0.9995 (for L<sup>1</sup>), and R=0.9999 (for L<sup>2</sup>).

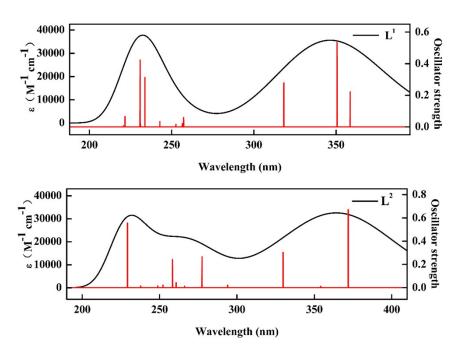


Figure S11. Theoretical UV–Visible absorption spectra of L<sup>1</sup> (up) and L<sup>2</sup> (down) and selected most pertinent lowlying electronic transitions and assignment of L<sup>1</sup>, L<sup>2</sup>.

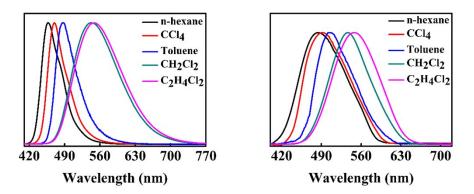
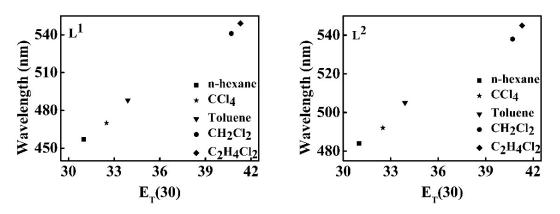


Figure S12. Normalized emission spectra of L<sup>1</sup> and L<sup>2</sup> in various solvents.



**Figure S13.** Variation of the emission wavelength maximum for  $L^1$  and  $L^2$  as a function of Reichardt's empirical solvent polarity parameters  $E_T(30)$ .

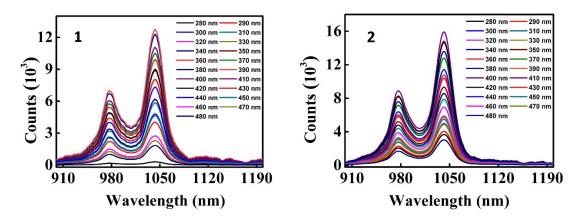


Figure S14. Wavelength dependent emission spectra of complexes 1 and 2.

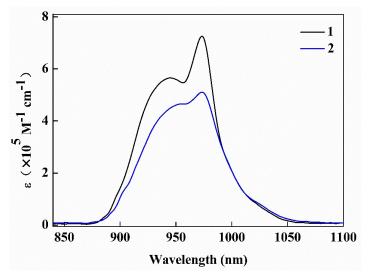


Figure S15. Near-infrared spectra of f-f absorption transition for 1 and 2 in CH<sub>3</sub>CN.

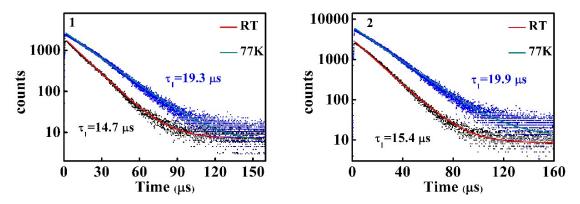


Figure S16. Luminescence decay curves of 1 and 2 in CH<sub>3</sub>CN monitored at 1042 nm.

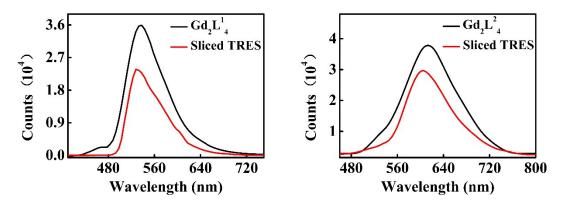


Figure S17. Emission spectra of  $Gd_2L_4$  (black) at 77 K and time delay (100  $\mu$ s) in CH<sub>3</sub>CN.

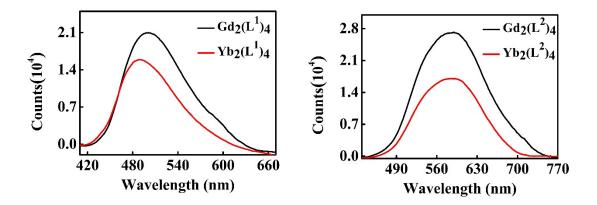


Figure S18. Emission spectra of  $Gd_2L_4$  (black) and  $Yb_2L_4$  (red) in  $CH_3CN$  at room temperature.