

Electronic Supplementary Information

**Organic Microspheres and Microcrystals Made from Pyridine-Functionalized Chromophores and a Chiral Phosphoric Acid**

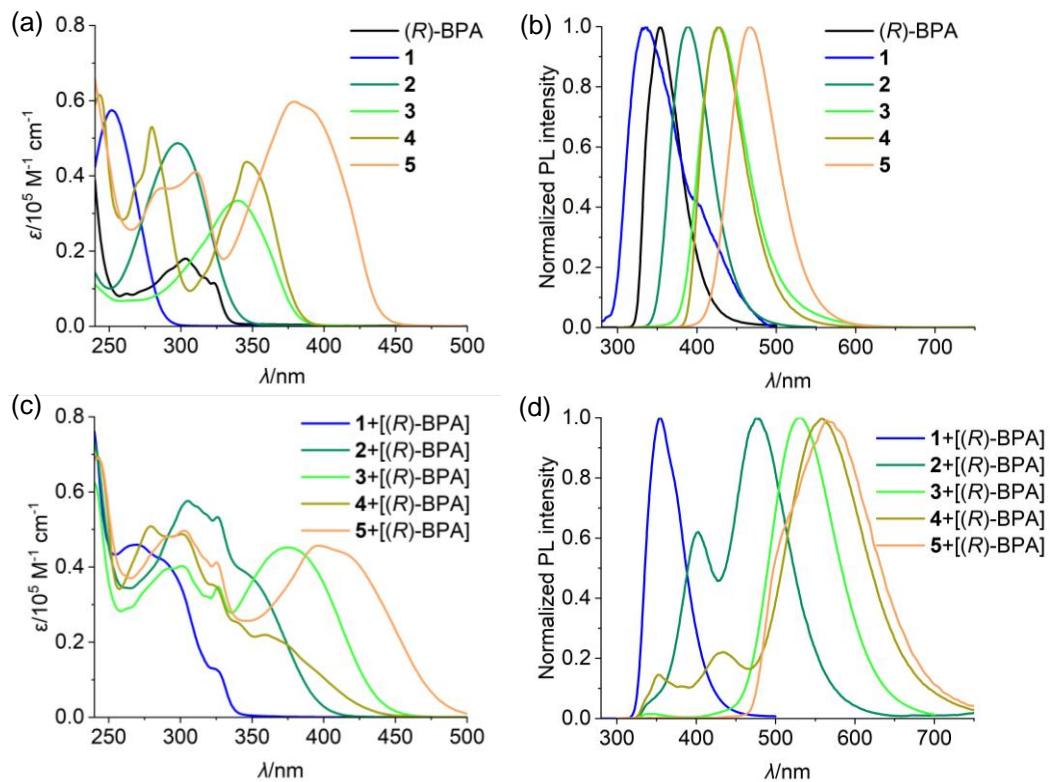
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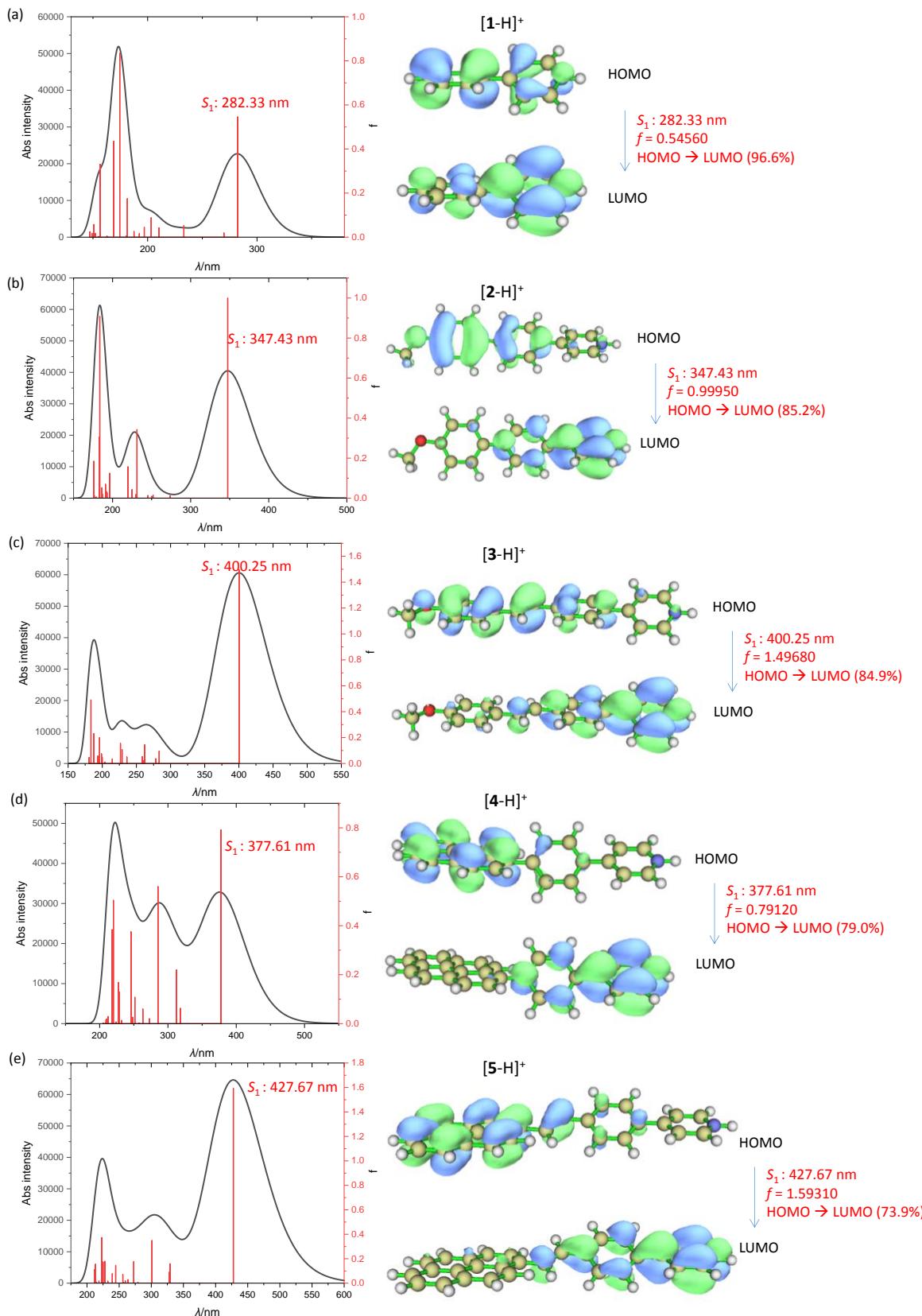
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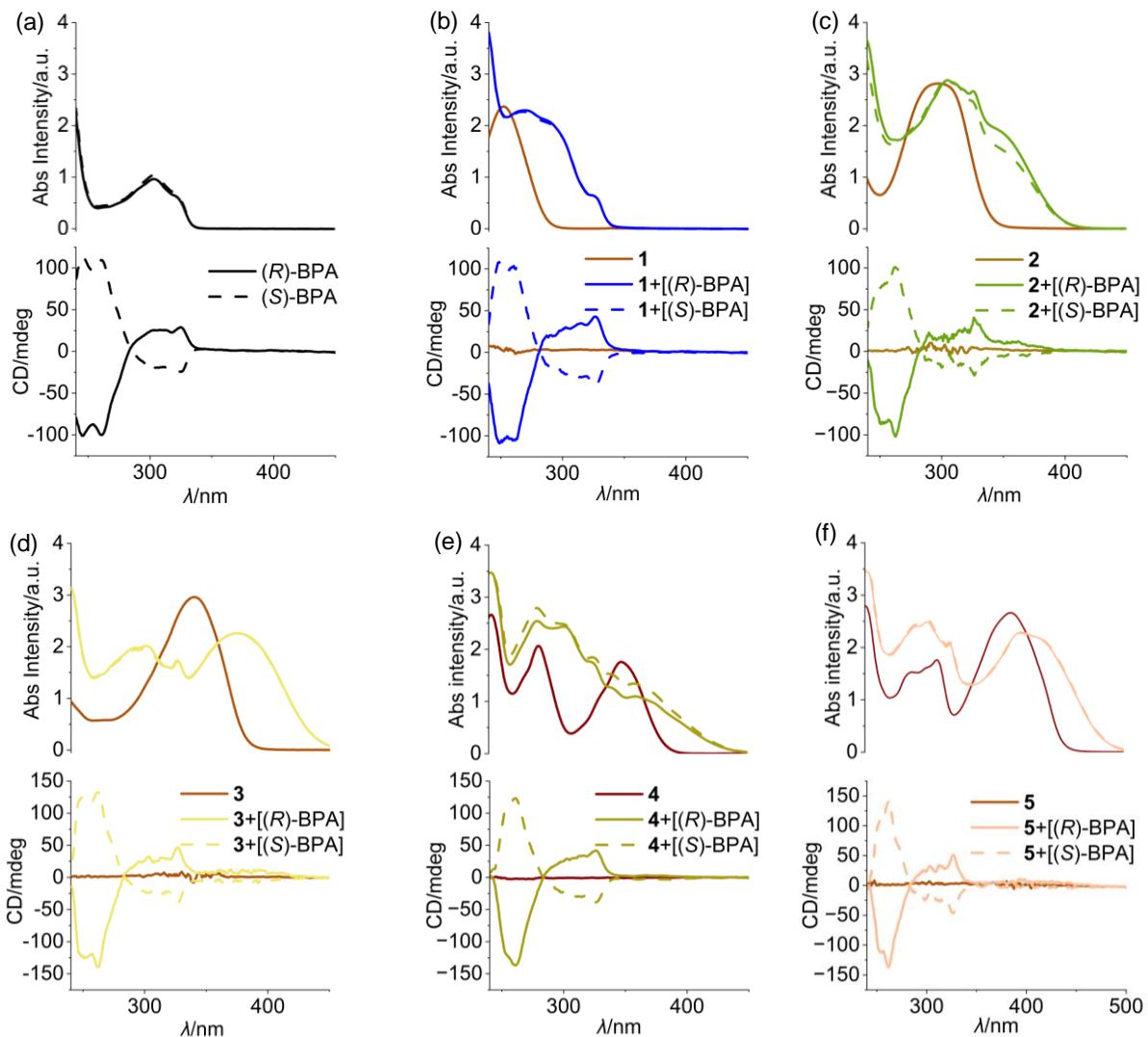
\*E-mail: [lizhongqiu@iccas.ac.cn](mailto:lizhongqiu@iccas.ac.cn) (Z.-Q.L.); [zilichen@ruc.edu.cn](mailto:zilichen@ruc.edu.cn) (Z.C.); [zhongyuwu@iccas.ac.cn](mailto:zhongyuwu@iccas.ac.cn) (Y.-W.Z.)



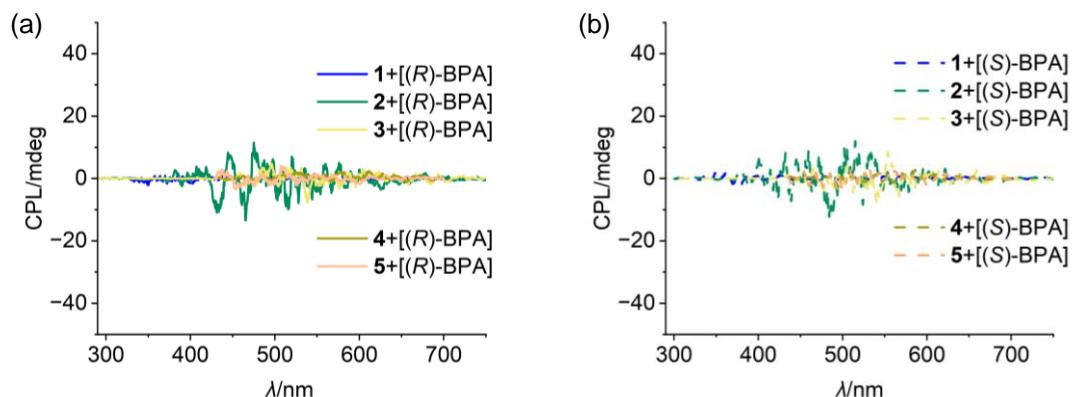
**Figure S1.** (a,c) Absorption and (b,d) photoluminescence (PL) spectra of (a,b) **1**–**5** and (R)-BPA and (c,d) **1**–**5** in the presence of 2 equiv of (R)-BPA in  $\text{CH}_2\text{Cl}_2$  ( $5 \times 10^{-5} \text{ M}$ ). Excitation wavelength: 260 nm for **1**; 280 nm for **2**–**4**; 300 nm for **5**.



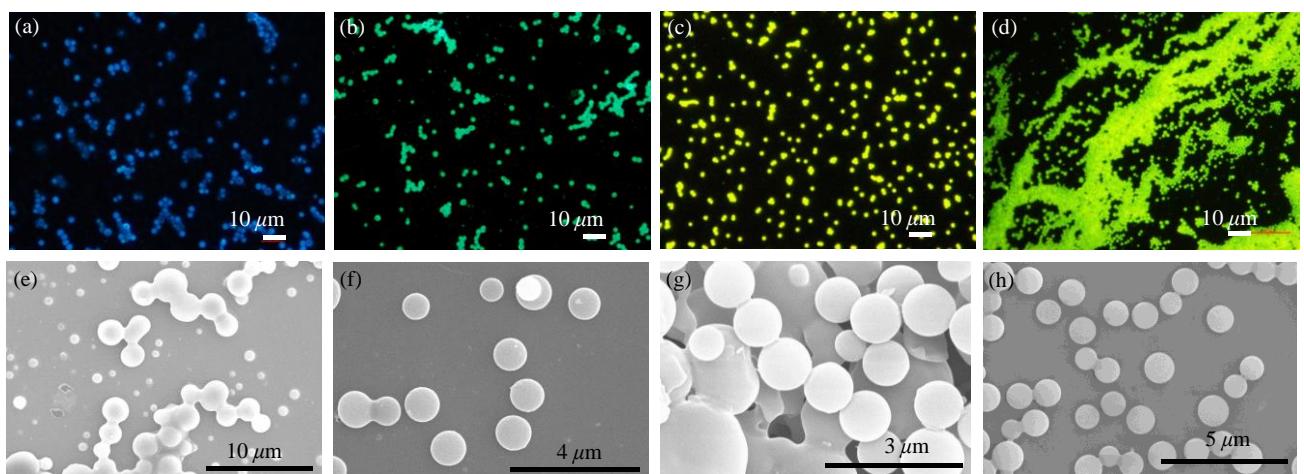
**Figure S2.** TDDFT-calculated vertical excitations of  $[1\text{-H}]^+ - [5\text{-H}]^+$ . Left: predicted excitation and simulated absorption spectrum. Right: HOMO and LUMO plots responsible for  $S_1$  excitations with indicated wavelength, oscillator strength ( $f$ ), and dominant contributions.



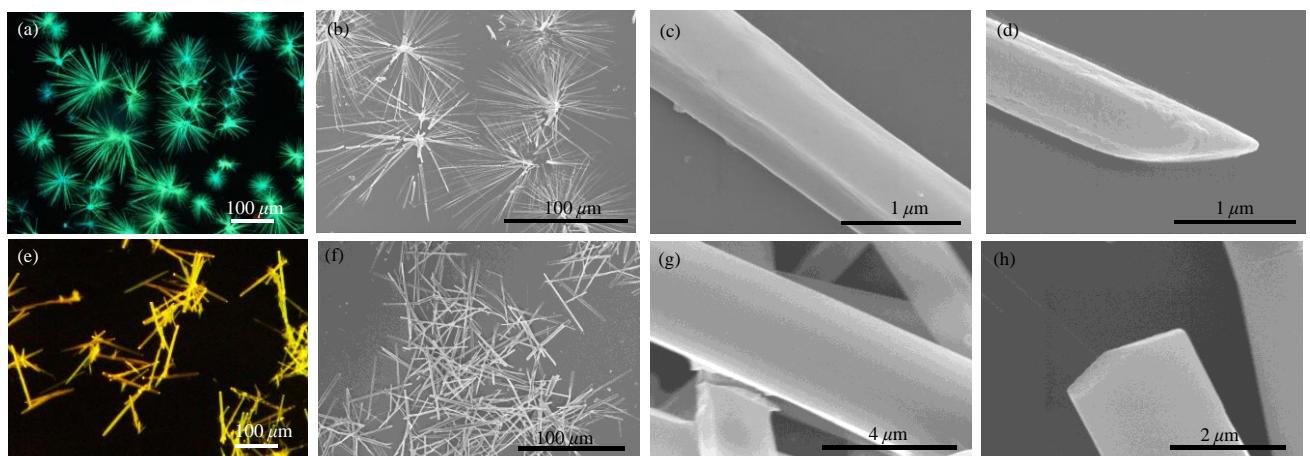
**Figure S3.** (a-f) UV-vis absorption (upper) and CD (below) spectra of (*R/S*)-BPA and **1 – 5** in  $\text{CH}_2\text{Cl}_2$  ( $5 \times 10^{-5}$  M) in the presence of 2 equiv of (*R*)-BPA and (*S*)-BPA.



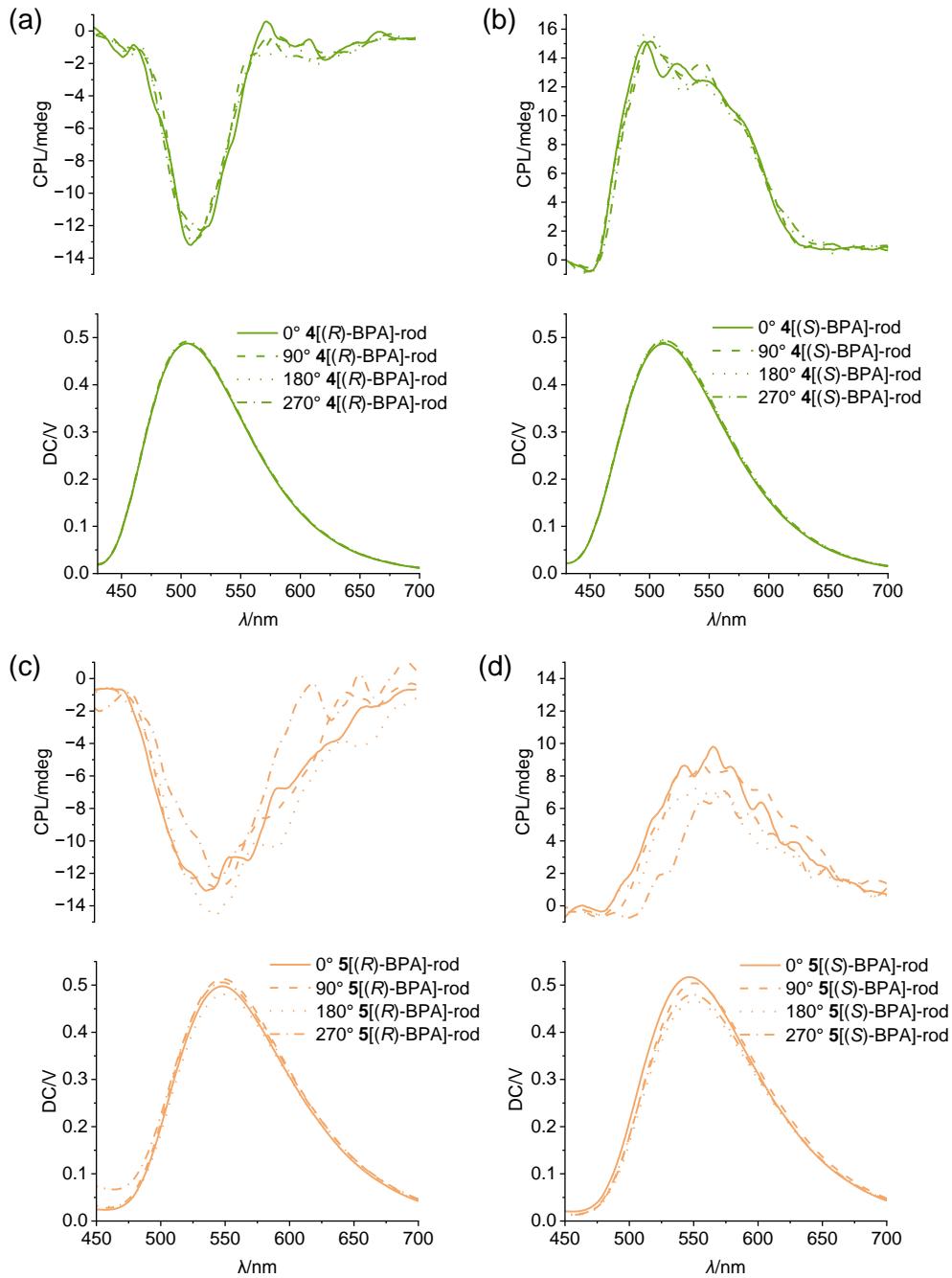
**Figure S4.** (a,b) CPL spectra of **1 – 5** in  $\text{CH}_2\text{Cl}_2$  ( $5 \times 10^{-4}$  M) in the presence of 2 equiv of (a) (*R*)-BPA and (b) (*S*)-BPA. Excitation wavelength: 260 nm for **1 – 4**; 280 nm for **5**.



**Figure S5.** (a-d) Fluorescent microscopy images of (a) **1[(S)-BPA]-sph**, (b) **2[(S)-BPA]-sph**, (c) **3[(S)-BPA]-sph**, and (d) **4[(S)-BPA]-sph**. (e-h) SEM images of (e) **1[(S)-BPA]-sph**, (f) **2[(S)-BPA]-sph**, (g) **3[(S)-BPA]-sph**, and (h) **4[(S)-BPA]-sph**.



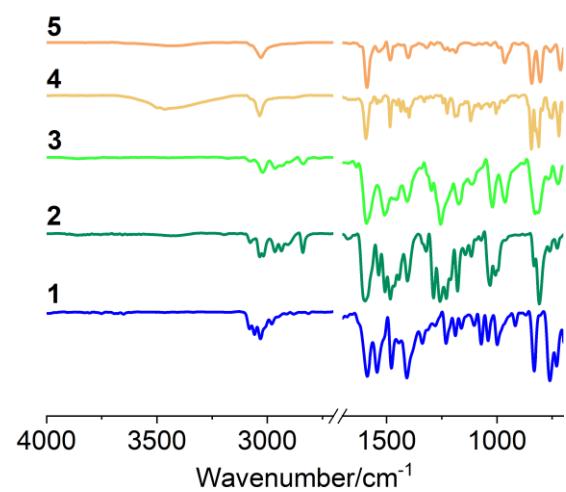
**Figure S6.** (a,e) Fluorescent microscopy images of (a) **4[(S)-BPA]-rod** and (e) **5[(S)-BPA]-rod**. (b-d,f-h) SEM images of (b-d) **4[(S)-BPA]-rod** and (f-h) **5[(S)-BPA]-rod**.



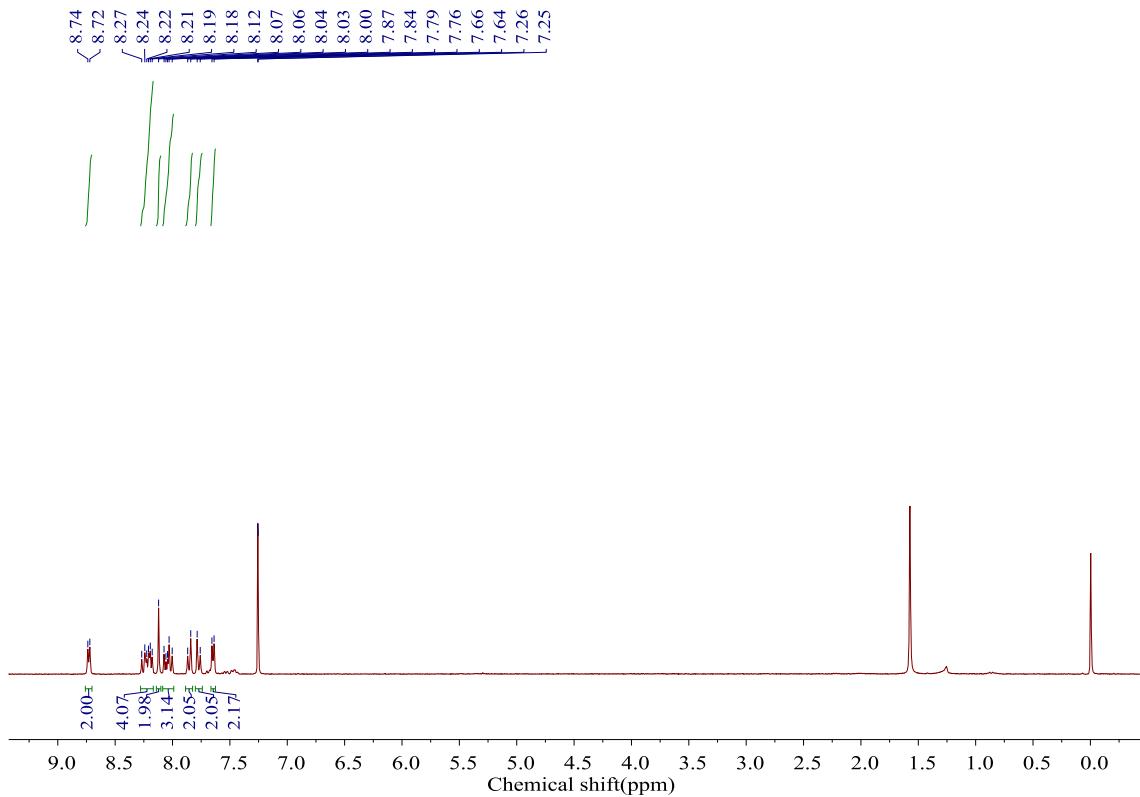
**Figure S7.** DC voltage and CPL spectra of microcrystals (a) **4[(R)-BPA]-rod**, (b) **4[(S)-BPA]-rod**, (c) **5[(R)-BPA]-rod**, and (d) **5[(S)-BPA]-rod** measured at different substrate angles from  $0^\circ$  to  $270^\circ$  (the substrate was rotated to a certain degree for repeated measurements).

**Table S1.** Crystallographic data and parameters.

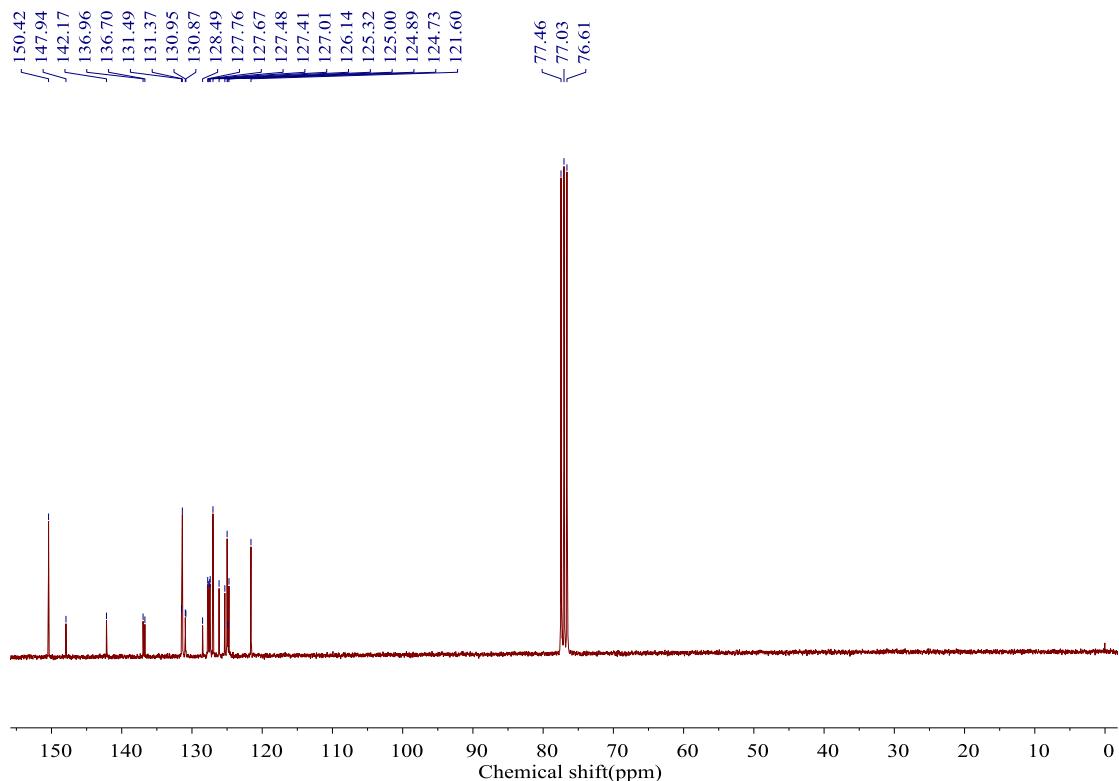
compound	<b>4[(R)-BPA]</b>	<b>5[(R)-BPA]</b>
CCDC number	2210345	2210218
empirical formula	C <sub>51</sub> H <sub>39</sub> NO <sub>5.5</sub> P	C <sub>98</sub> H <sub>64</sub> N <sub>2</sub> O <sub>8</sub> P <sub>2</sub>
formula weight	784.80	1459.45
Temperature(K)	169.99(10)	170.15
crystal system	monoclinic	monoclinic
Space group	<i>C</i> 2	<i>P</i> 2 <sub>1</sub>
a (Å)	14.37456(12)	23.1912(7)
b (Å)	17.64525(16)	6.81437(18)
c (Å)	16.12546(14)	30.7762(8)
α (°)	90	90
β (°)	96.9673(8)	103.231(3)
γ (°)	90	90
V (Å <sup>3</sup> )	4059.90(6)	4734.6(2)
Z value	4	2
Density (g/cm <sup>3</sup> )	1.284	1.024
R1 (final)	0.0630	0.0550
wR2 (final)	0.1703	0.1448
R1 (all)	0.0653	0.0702
wR2 (all)	0.1725	0.1546
Flack parameter	0.022(8)	0.080(15)



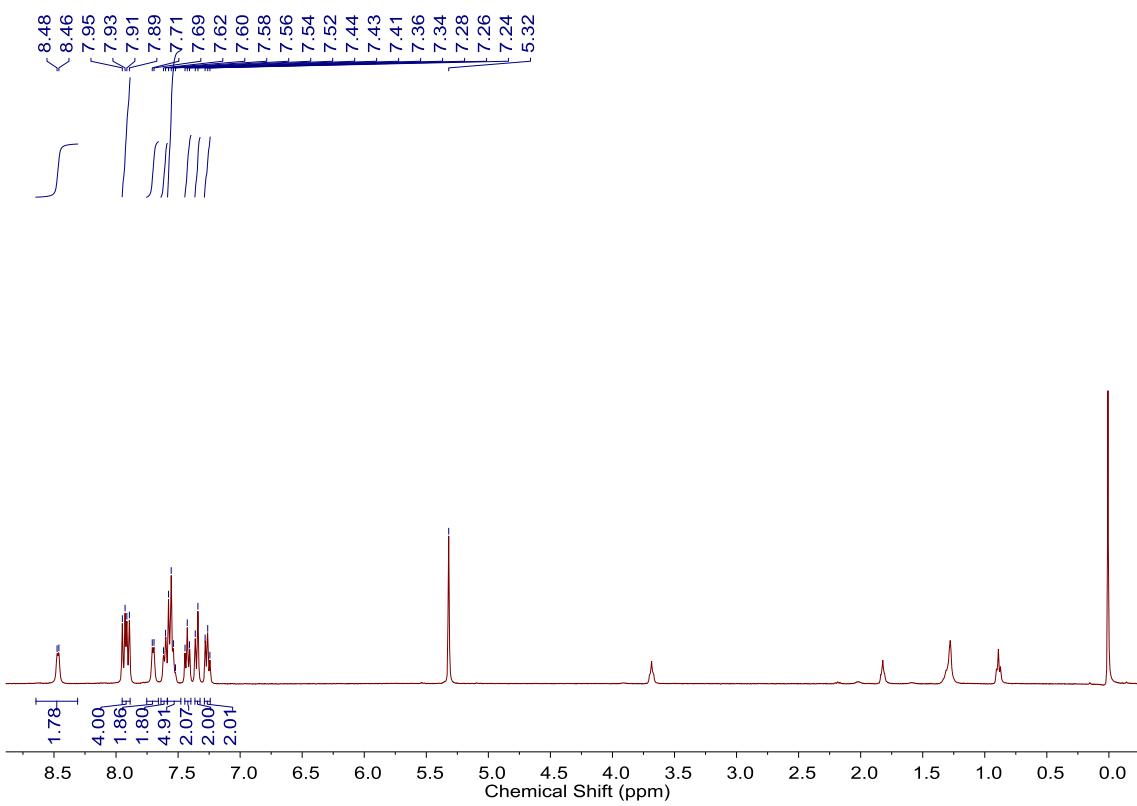
**Figure S8.** FTIR spectra of **1 – 5** as KBr pellets.



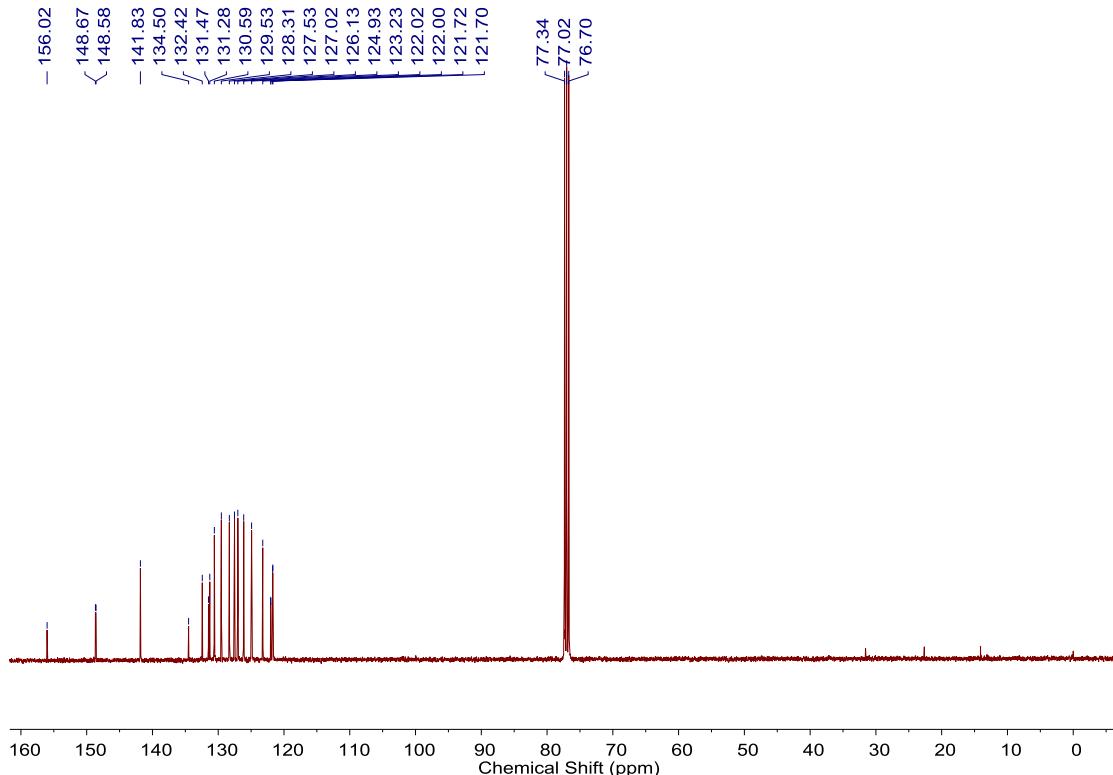
**Figure S9.**  $^1\text{H}$  NMR spectrum of **4** in  $\text{CDCl}_3$ .



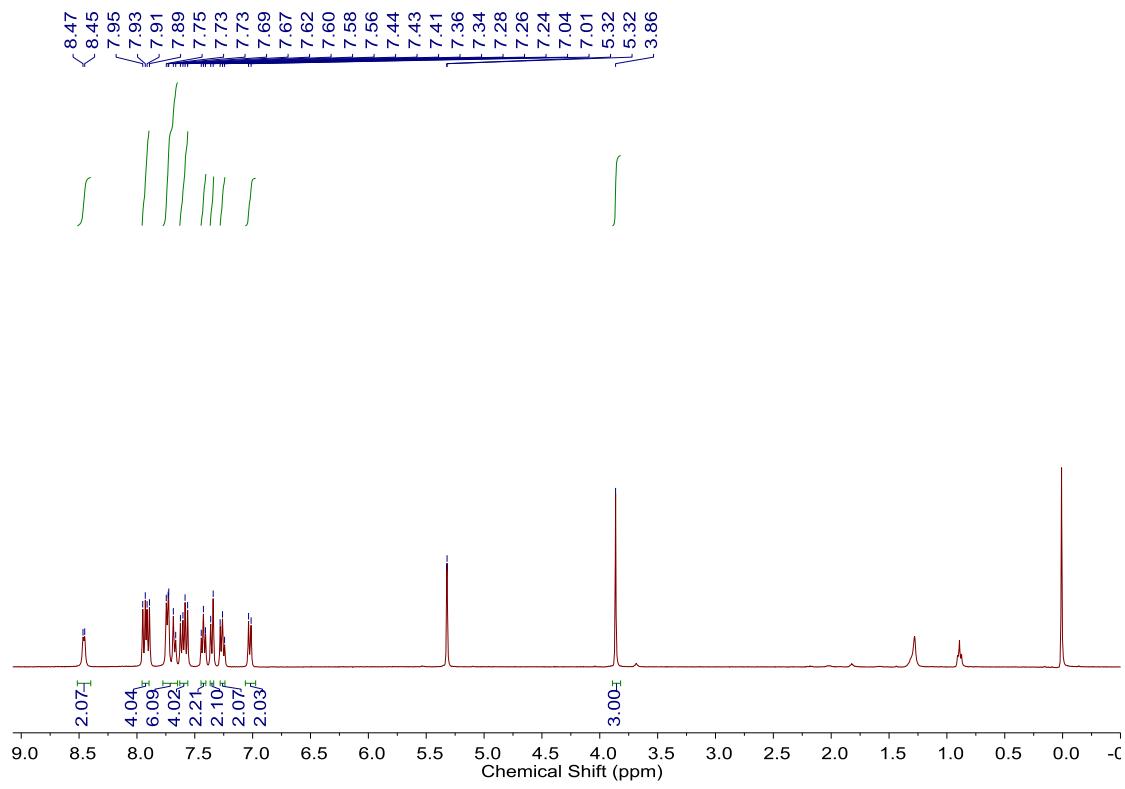
**Figure S10.**  $^{13}\text{C}$  NMR spectrum of **4** in  $\text{CDCl}_3$ .



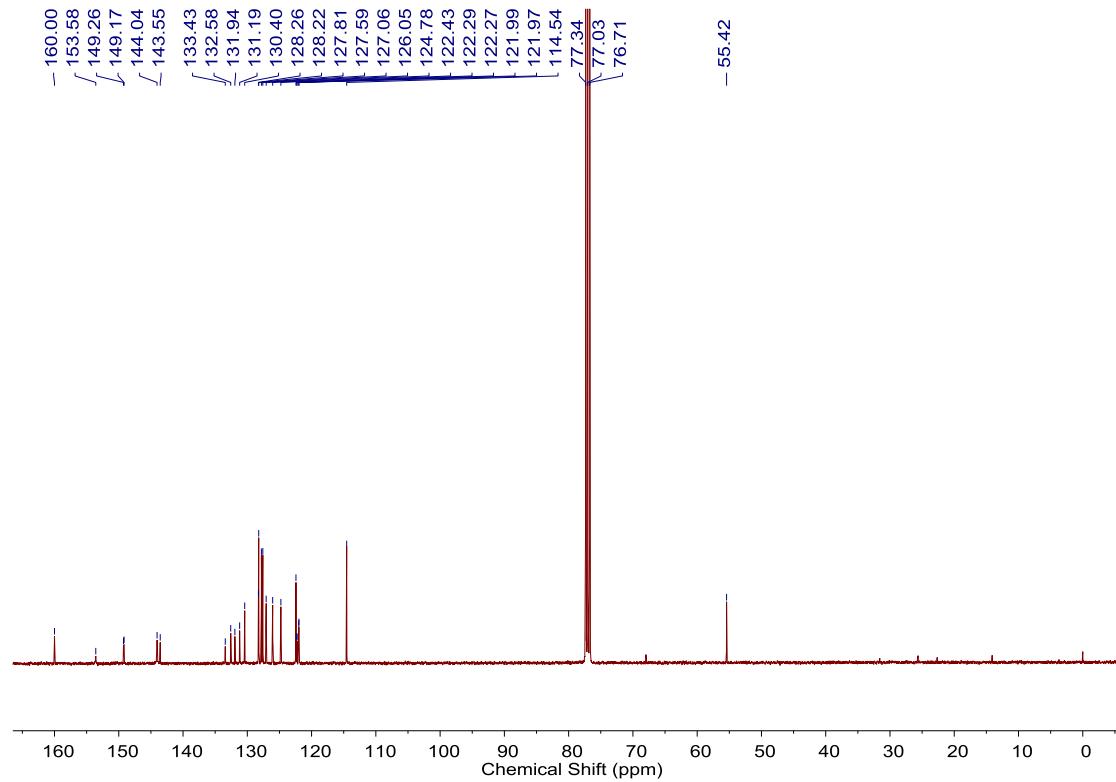
**Figure S11.**  $^1\text{H}$  NMR spectrum of **1**[(*R*)-BPA]-sph in  $\text{CD}_2\text{Cl}_2$ .



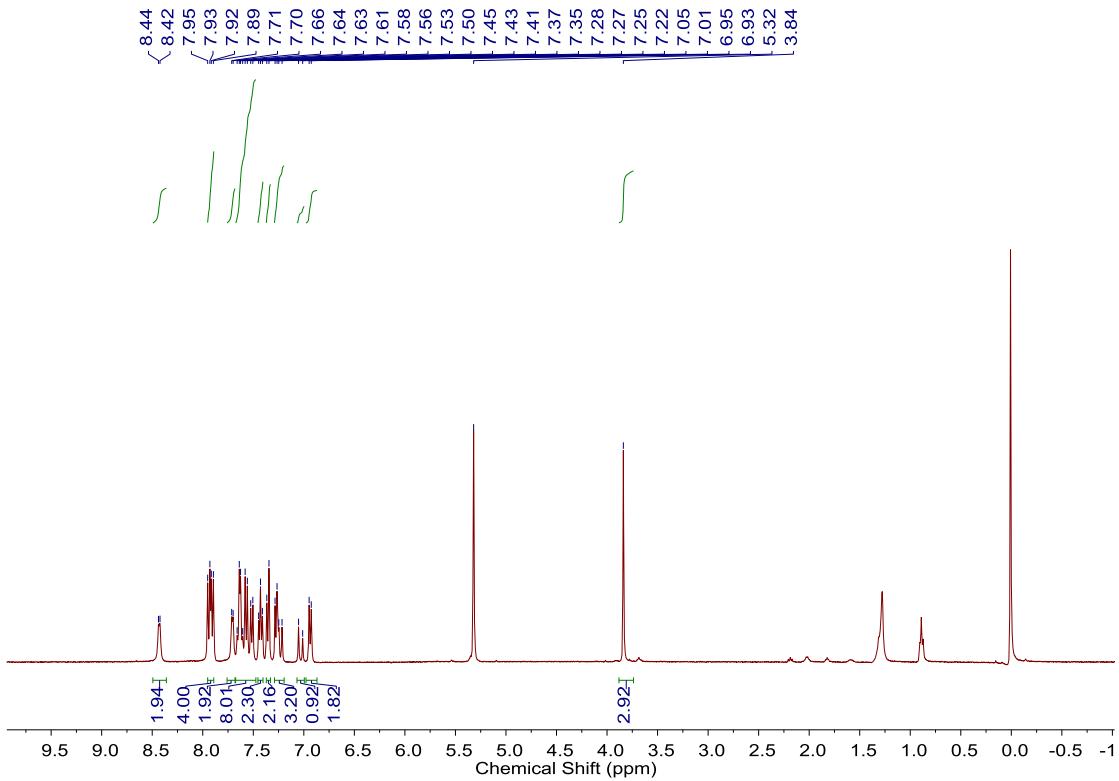
**Figure S12.**  $^{13}\text{C}$  NMR spectrum of **1**[(*R*)-BPA]-sph in  $\text{CDCl}_3$ .



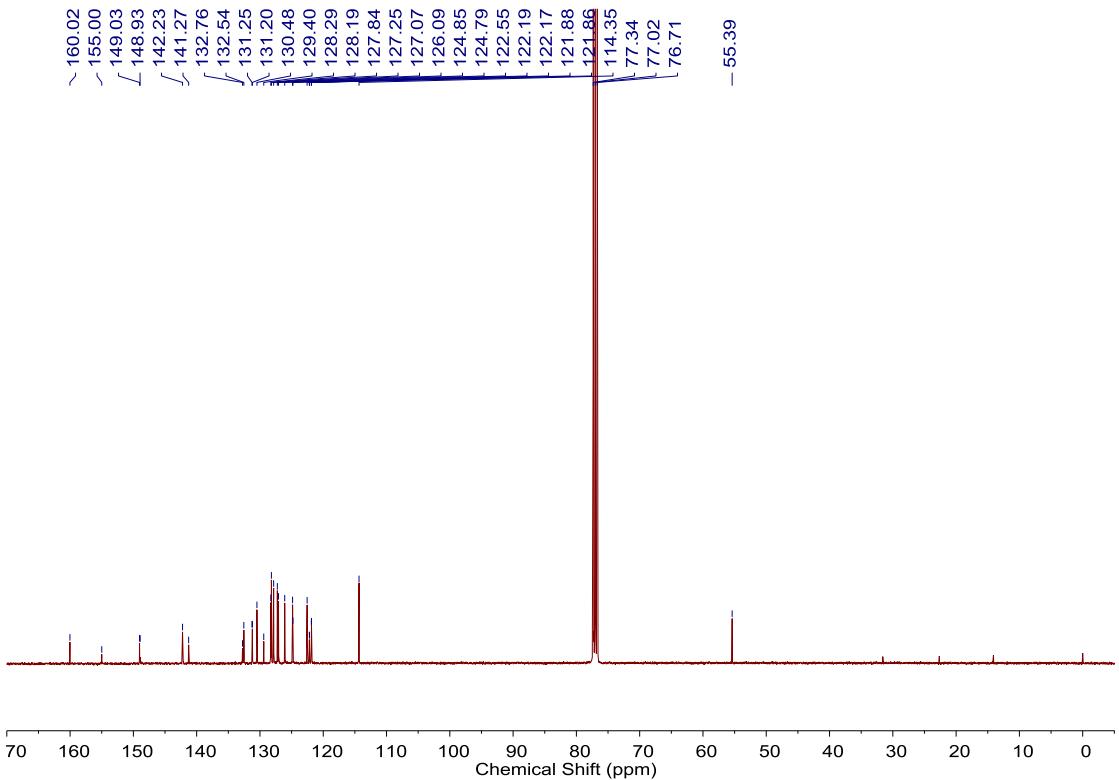
**Figure S13.**  $^1\text{H}$  NMR spectrum of **2**[(*R*)-BPA]-sph in  $\text{CD}_2\text{Cl}_2$ .



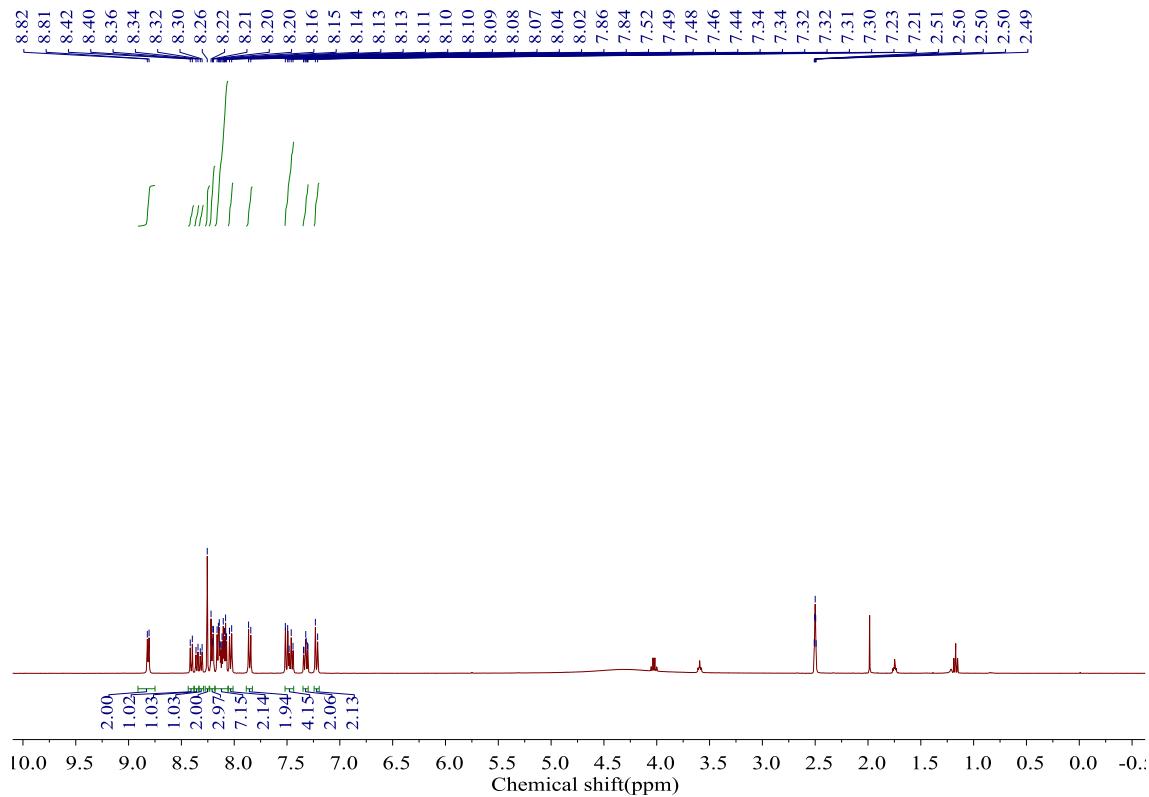
**Figure S14.**  $^{13}\text{C}$  NMR spectrum of **2**[(*R*)-BPA]-sph in  $\text{CDCl}_3$ .



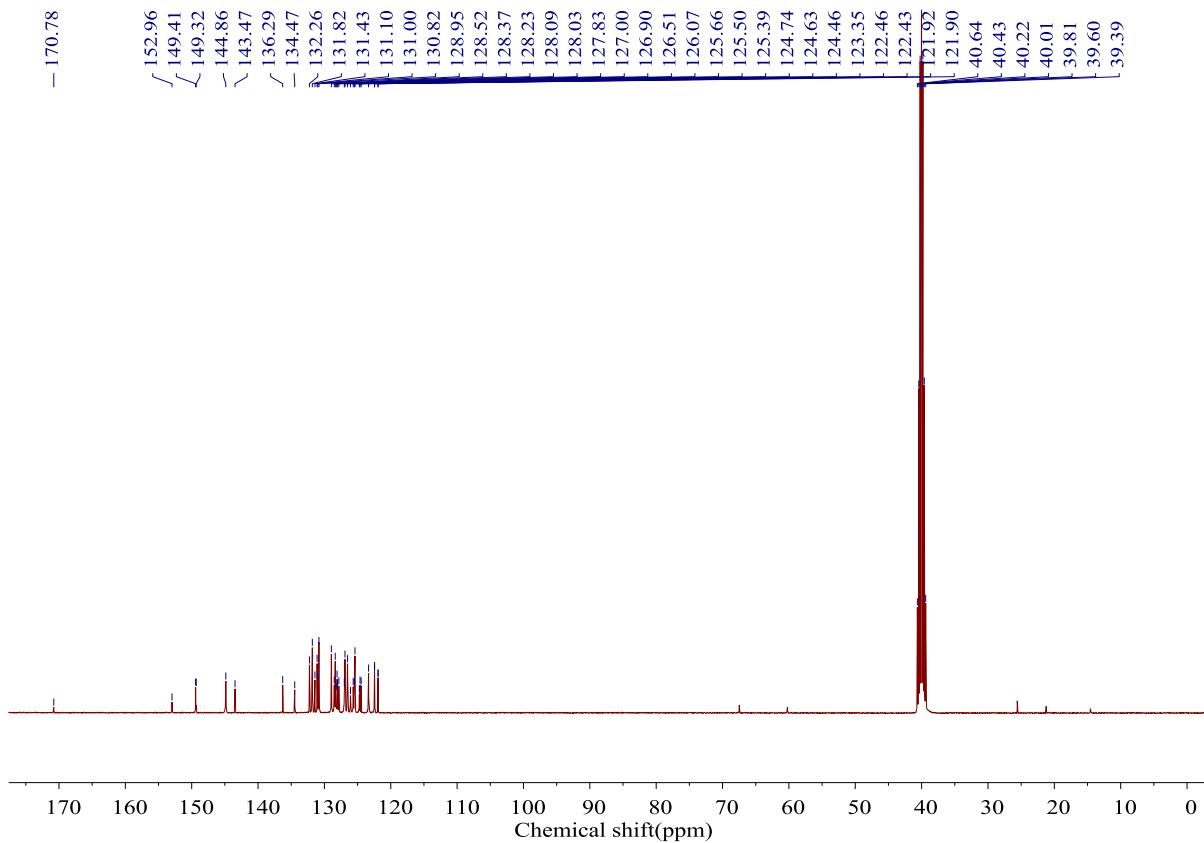
**Figure S15.**  $^1\text{H}$  NMR spectrum of **3**[(*R*)-BPA]-sph in  $\text{CD}_2\text{Cl}_2$ .



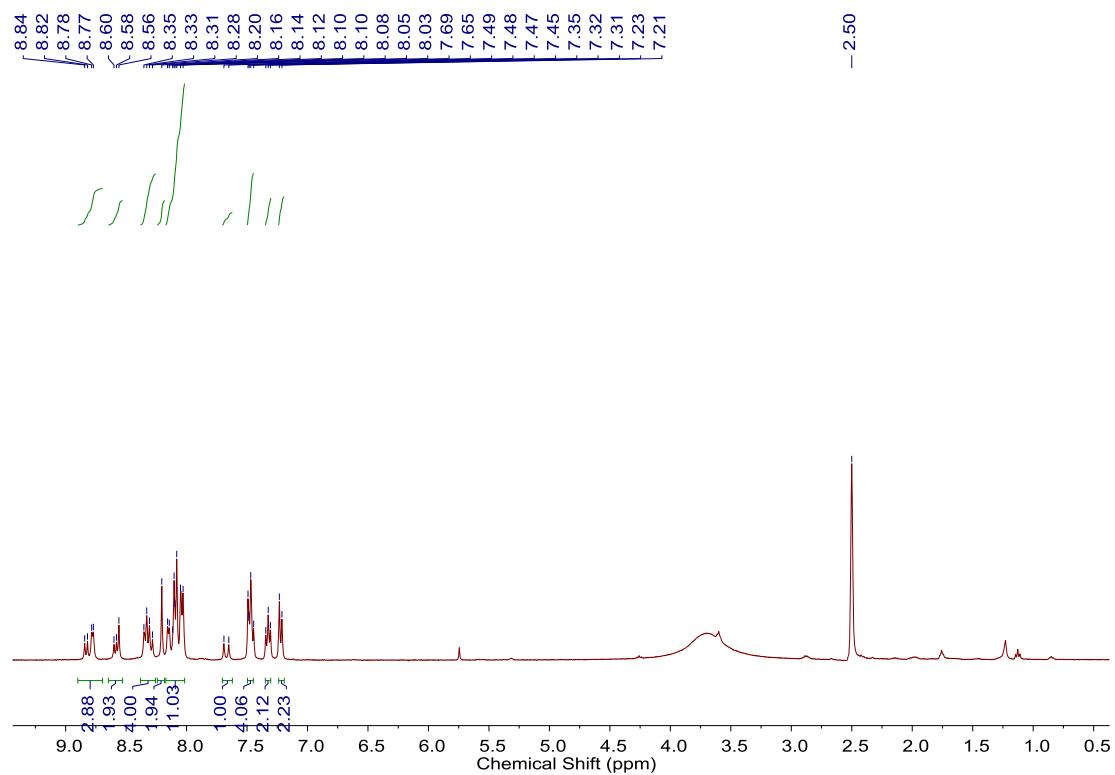
**Figure S16.**  $^{13}\text{C}$  NMR spectrum of **3**[(*R*)-BPA]-sph in  $\text{CDCl}_3$ .



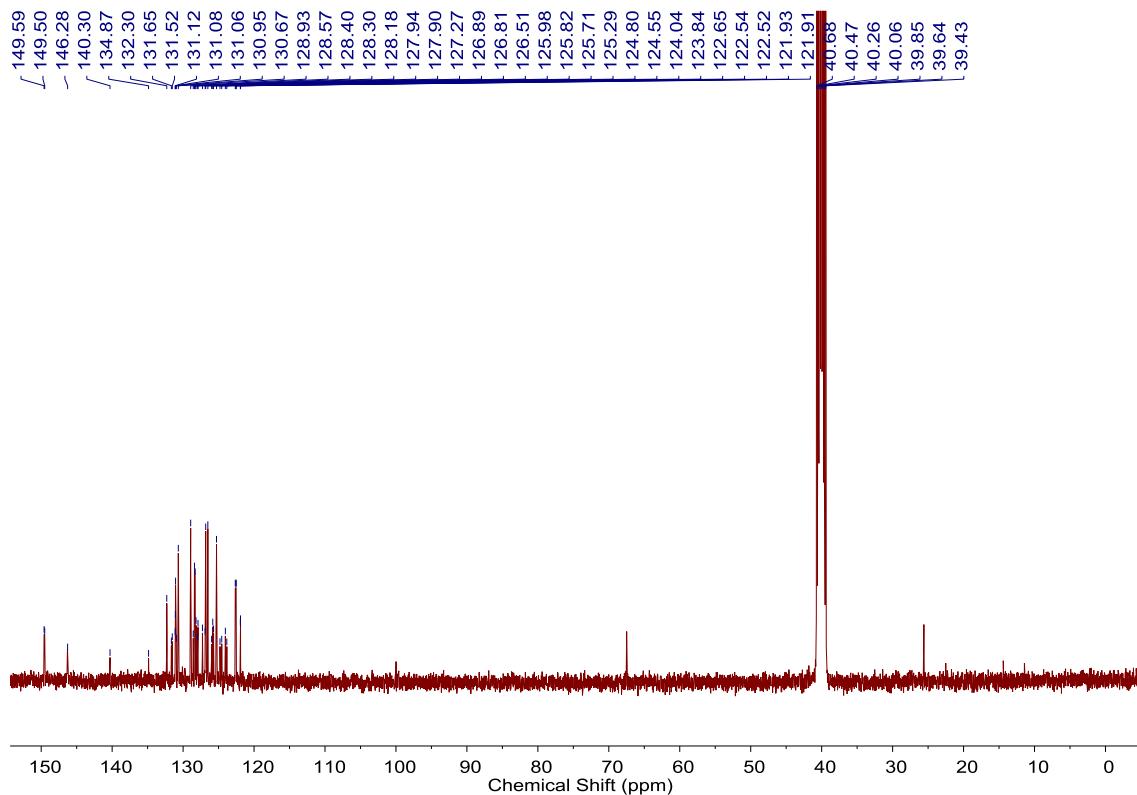
**Figure S17.**  $^1\text{H}$  NMR spectrum of **4**[(*R*)-BPA]-sph in  $\text{DMSO}-d_6$ .



**Figure S18.**  $^{13}\text{C}$  NMR spectrum of **4**[(*R*)-BPA]-sph in  $\text{DMSO}-d_6$ .



**Figure S19.**  $^1\text{H}$  NMR spectrum of **5**[(*R*)-BPA]-rod in  $\text{DMSO}-d_6$ .



**Figure S20.**  $^{13}\text{C}$  NMR spectrum of **5**[(*R*)-BPA]-rod in  $\text{DMSO}-d_6$ .