

## Supplementary Information

### **A Theoretical exploration on second-order NLO properties of linked sandwich double-layered metallocarborane: magic charge transfer caused by linked groups**

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### **Content**

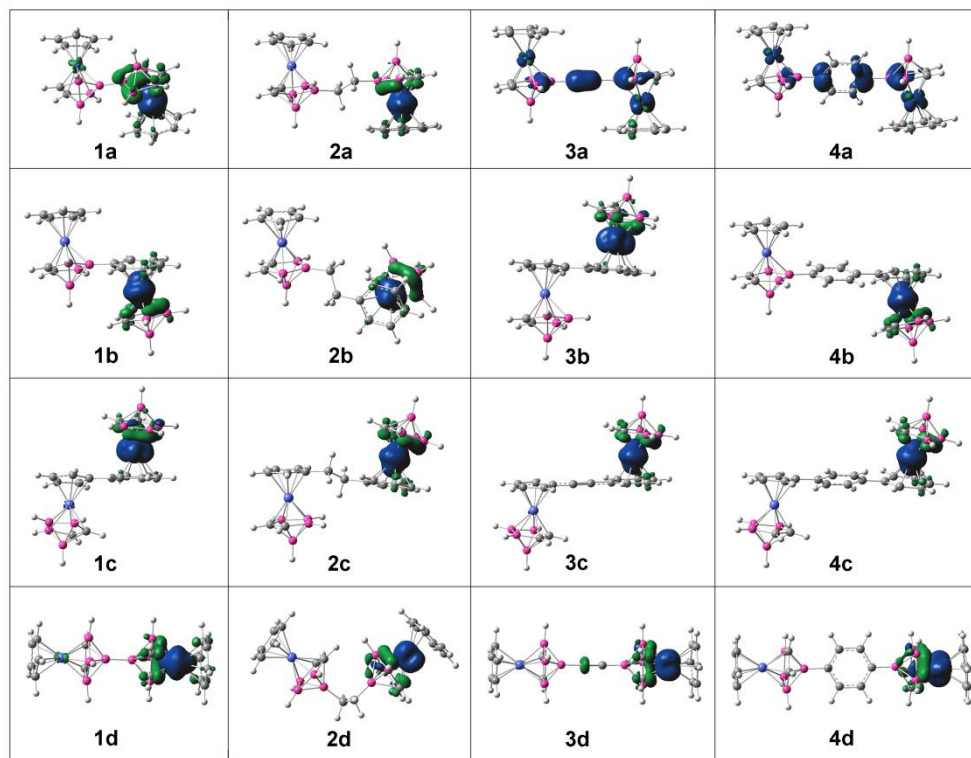
**Table S1.**  $\beta$  values (a.u.) calculated by CAM-B3LYP and  $\omega$ B97XD functionals with 6-31+G(d)/SDD basis set for redox states.

**Fig. S1.** Spin density of oxidation states for all the studied systems.

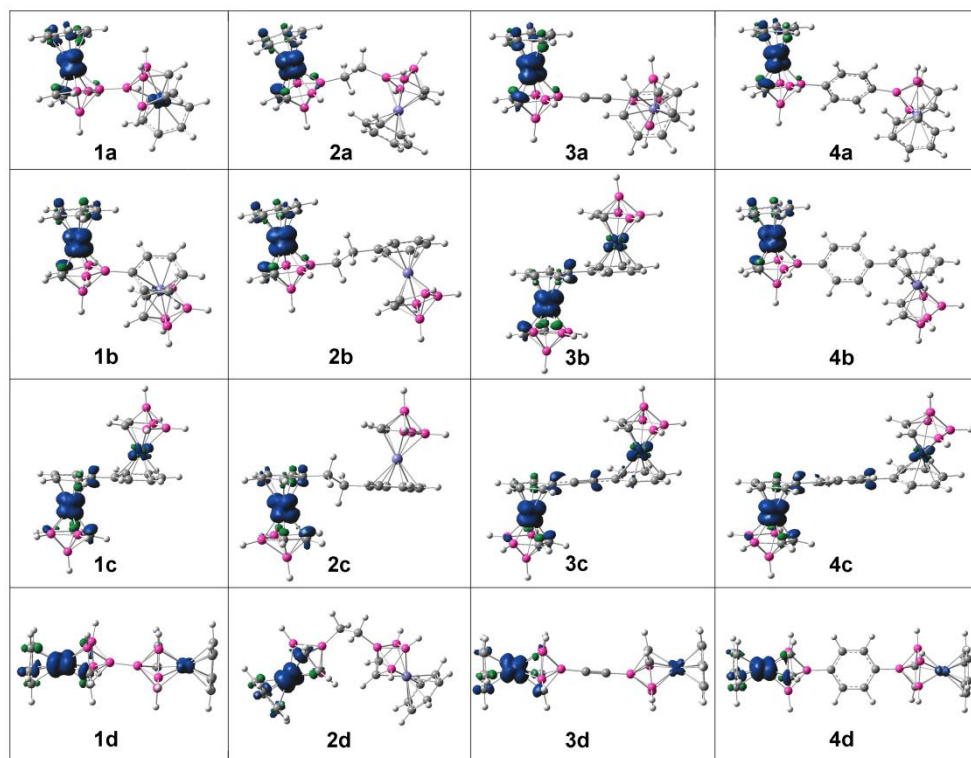
**Fig. S2.** Spin density of reduced states for all the studied systems.

**Table S1.**  $\beta$  values (a.u.) calculated by CAM-B3LYP and  $\omega$ B97XD functionals with 6-31+G(d)/SDD basis set for redox states.

Comp.	CAM-B3LYP/6-31+G(d)/SDD			$\omega$ B97XD6-31+G(d)/SDD		
	oxi	0	red	oxi	0	red
<b>1a</b>	40.4	4.6	19.1	39.5	4.5	16.6
<b>1b</b>	8.8	7.0	11.7	7.6	7.0	10.7
<b>1c</b>	29.1	7.6	70.4	31.9	7.5	72.1
<b>1d</b>	69.8	5.7	47.3	54.8	5.6	40.1
<b>2a</b>	10.1	4.5	4.0	9.3	4.6	3.9
<b>2b</b>	5.2	2.5	5.6	4.7	2.6	1.9
<b>2c</b>	55.3	5.0	2.7	67.1	4.7	13.6
<b>2d</b>	19.4	6.0	13.2	46.8	5.8	18.1
<b>3a</b>	48.6	9.3	29.0	12.7	8.9	25.1
<b>3b</b>	16.3	5.5	63.7	19.0	5.9	59.2
<b>3c</b>	98.9	10.5	397.7	90.3	10.5	382.3
<b>3d</b>	171.0	2.0	93.0	146.5	2.2	77.7
<b>4a</b>	57.7	8.0	46.9	42.3	7.6	39.5
<b>4b</b>	72.0	4.6	102.7	62.4	4.9	94.3
<b>4c</b>	97.9	6.3	444.9	83.1	6.3	530.8
<b>4d</b>	317.5	2.7	109.4	235.5	3.0	108.3



**Fig. S1.** Spin density of oxidation states for all the studied systems.



**Fig. S2.** Spin density of reduced states for all the studied systems.