Open-shell Singlet Diradicaloids, Polyradicaloids and Covalent Organic Radical Frameworks

Our group has conducted intensive studies on open-shell singlet diradicaloids and polyradicaloids,1 mainly including zigzag graphene fragments2 and pro-aromatic quinoidal compounds.3 In addition, global aromaticity in 2D macrocyclic polyradicaloids4 and even 3D molecular cage5 was observed. Extension of the polyradicaloid system to 2D framework leads to covalent organic radical frameworks (CORFs),6 which demonstrated interesting magnetization depending on the topological structure and inter-radical exchange interaction.



**References**

[1] See our reviews/accounts: (a) Z. Sun *et al.*, *Acc. Chem. Res.***2014**, *47*, 2582. (b) Z. Zeng *et al*., *Chem. Soc. Rev.* **2015**, *44*, 6578. (c) T. Y. Gopalakrishna *et al.*, *Chem. Commun.* **2018**, *54*, 2389. (d) C. Liu *et al.*, *Acc. Chem. Res.* **2019**, *52*, 2309. (e) W. Zeng *et al*., *Chem* **2021**, *7*, 358.

[2] See examples: (a) Z. Sun *et al*., *J. Am. Chem. Soc.* **2011**, *133*, 11896. (b) Y. Li *et al*., *J. Am. Chem. Soc.* **2012**, *134*, 14913. (c) Z. Sun *et al*., *J. Am. Chem. Soc.* **2013**, *135*, 18229. (d) R. Huang *et al*., *J. Am. Chem. Soc.* **2016**, *138*, 10323. (e) W. Zeng *et al*., *Angew.**Chem. Int. Ed.* **2016**, *55*, 8816. (f) P. Hu *et al*., *J. Am. Chem. Soc.* **2016**, *138*, 1065. (g) W. Zeng *et al*., *Chem* **2017**, *2,* 81. (h) Y. Gu *et al*., *Angew. Chem. Int. Ed.* **2018**, *57*, 6541. (i) Y. Ni *et al*., *Angew. Chem. Int. Ed.* **2018**, *57*, 9697. (j) W. Zeng *et al*., *J. Am. Chem. Soc.* **2018**, *140*, 14054. (k) J. Su *et al*., *Sci. Adv.* **2019**, 5: eaav7717. (l) Y. Zou *et al*., *J. Am. Chem. Soc.***2019**, *141*, 7266. (m) J. Su *et al*., *Nano Lett*. **2021**, *21*, 861. (n) M. Telychko *et al*., *Sci. Adv.* **2021**, *7*: eabf0269.

[3] See examples: (a) Z. Zeng *et al*., *J. Am. Chem. Soc.* **2012**, *134*, 14513. (b) Z. Zeng *et al*., *Angew. Chem. Int. Ed.* **2013**, *52*, 8561. (c) Z. Zeng *et al*., *J. Am. Chem. Soc.* **2013**, *135*, 6363. (d) Z. Zeng *et al*., *J. Am. Chem. Soc.* **2015**, *137*, 8572. (e) Y. Ni *et al*., *Angew. Chem. Int. Ed.* **2016**, *55*, 2815. (f) H. Zhang *et al*., *Angew. Chem. Int. Ed.* **2017**, *56*, 13484. (g) G. Li *et al*., *Angew. Chem. Int. Ed.* **2017**, *56*, 5012. (h) J. Wang *et al*., *Angew. Chem. Int. Ed.* **2017**, *56*, 14154. (i) G. Li *et al*., *Angew. Chem. Int. Ed.* **2019**, *58*, 14319. (j) G. Li *et al*., *Angew. Chem. Int. Ed.* **2021**, *60*, DOI: 10.1002/anie.202100606.

[4] See examples: (a) S. Das *et al*., *J. Am. Chem. Soc.* **2016**, *138*, 7782. (b) X. Lu *et al*., *J. Am. Chem. Soc.* **2017**, *139*, 13173. (c) G. Li *et al*., *Angew. Chem. Int. Ed.* **2018**, *57*, 7166. (d) A. Rana *et al*., *Angew. Chem. Int. Ed.* **2018**, *57*, 13052. (e) X. Lu *et al*., *Angew. Chem. Int. Ed.* **2018**, *57*, 13052. (f) C. Liu *et al*., *Chem* **2018**, *4*, 1586. (g) X. Lu *et al*., *J. Am. Chem. Soc.***2019**, *141*, 5934. (h) Y. Ni *et al*., *Chem* **2019**, *5*, 108. (i) Z. Li *et al*., *J. Am. Chem. Soc.***2019**, *141*, 16266. (j) L. Ren *et al*., *Angew. Chem. Int. Ed.* **2020**, *59*, 2230. (k) Y. Ni, et al., *Angew. Chem. Int. Ed.* **2020**, *59*, 7414.

[5] See examples: (a) X. Gu *et al*., *Angew. Chem. Int. Ed.* **2017**, *56*, 15383. (b) Y. Ni *et al*., *Nat. Chem.* **2020**, *12*, 242. (c) G. Li, *et al*., *Angew. Chem. Int. Ed.* **2020**, *59*, 9727. (d) Y. Ni *et al*., *J. Am. Chem. Soc.* **2020**, *142*, 12730. (e) J. Zhu *et al*., *J. Am. Chem. Soc.* **2021**, DOI: 10.1021/jacs.1c00409.

[6] See examples: (a) S. Wu *et al*., *Angew. Chem. Int. Ed.* **2018**, *57*, 8007. (b) H. Phan *et al*., *Chem* **2019**, *5*, 1223.