第253回 OPERA研究交流セミナー
第244回 ISIT有機光エレクトロニクス研究特別室セミナー
第311回 未来化学創造センターセミナー

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「Tuning the closed/open shell character of surface-generated molecules by their coupling motifs」

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On-surface synthesis (OSS) has enabled the fabrication and characterization of a myriad of π-magnetic molecular materials.1 Iconic examples include triangulenes, which are magnetic due to sublattice imbalance, and acenes, which have increasing diradical character with size. Recently, we showed that substituting a N in the C network of triangulene, to make aza-triangulene, enriches its surface chemistry, leading to opposite charge transfer directions on Au(111) and Ag(111).2 We illustrate a new strategy to expand the chemical space of such structures: by forming dimeric derivatives with various coupling motifs. For aza-triangulene, forming a symmetric dimer led to a structure without magnetic fingerprints, while asymmetric coupling motifs led to structures that display Kondo resonance (with associated detection of SOMO/SUMO).3 Meanwhile, for acenes, we show an opposite trend with pentacene,4 the largest acene widely accepted to have a closed-shell character. A symmetric dimer, with a four-membered ring as a coupling motif, displayed a low HOMO-LUMO gap typically associated with diradicaloid molecules. Meanwhile, asymmetric dimers with five-membered rings as coupling motifs showed a considerably higher gap despite having the same extent of pi-conjugation. Understanding these structure–property relations is desirable not only for fundamental reasons but also for designing new complex and functional molecular structures. We illustrate this with DFT results on prospective new molecules, which illustrate further fine-tuning of magnetic properties.

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