Metal-free magnetism in organic two-dimensional polymers

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Two-dimensional (2D) polymers with metal-free magnetism have attracted considerable research interest due to their great potential applications in organic spintronics. [1-3] However, achieving stable spin-polarization and controlling magnetic interactions in these systems remains challenging due to strong electronic coupling and the closed-shell nature of most organic monomers. In this presentation, I will introduce two strategies to induce spin-polarization and regulate magnetic interactions in 2D polymers.

The first approach leverages chemical doping to introduce unpaired spins into narrow-band 2D covalent organic frameworks (COFs). [4] We demonstrated that forming a supramolecular charge-transfer complex is crucial for localizing electronic states and stabilizing paramagnetic centers in initially diamagnetic COFs. We revealed the anisotropic magnetic interactions in these 2D COFs and enabled effective tuning of both spin transport and magnetic coupling between spin centers.

The second approach employs nanographenes with stable radicals as building blocks, specifically triangulene—the smallest polybenzenoid with a triplet ground state and inherent spin-polarization. [5] By assembling triangulene monomers into dimers and extended 2D polymers, we theoretically explore strategies to control magnetic interactions and electronic structures. We achieve enhanced magnetic coupling (*J*) up to -198 meV through rational chemical design of triangulene dimers. [5] Notably, nitrogen-centered triangulene dimers exhibit ferromagnetic interactions that is beyond the predictive power of Ovchinnikov's rule. Furthermore, triangulene-based 2D polymers exhibit unique electronic features, including a Dirac point flanked by twin flat bands. By tuning the Fermi level, we predict metal-free ferromagnetism and potential half-metallic behavior. [6]

References

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