

# Magnetic organic 2D crystals

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Magnetic organic 2D crystals have garnered significant attention for their promising role in next-generation organic spintronics, owing to their lightweight nature and chemical tunability. [1] However, realizing robust spin polarization and tunable magnetic coupling in such systems is difficult, primarily because of strong electronic delocalization and the closed-shell character typical of conventional organic building blocks.

In this presentation, we introduce a powerful approach that leverages nanographenes bearing intrinsic stable radicals—particularly triangulene, the smallest polybenzenoid hydrocarbon with a triplet ground state—as monomeric units to construct extended 2D magnetic polymers. Through systematic theoretical investigations, we examine how dimerization and polymerization of triangulene units allow precise control over magnetic exchange interactions and overall electronic properties. [2]

Rational molecular engineering of triangulene dimers yields remarkably strong antiferromagnetic coupling, with exchange constants ( $J$ ) reaching up to  $-198$  meV. Strikingly, certain nitrogen-doped triangulene dimers display ferromagnetic coupling—even in twisted geometries—challenging the predictions of Ovchinnikov's rule and highlighting unconventional spin alignment mechanisms in these systems. [2]

Extending to periodic 2D triangulene-based networks, these structures exhibit a distinctive honeycomb-kagome lattice electronic signature, featuring a Dirac point symmetrically flanked by twin flat bands. By shifting the Fermi level via heteroatom substitution or doping, we predict the emergence of purely organic ferromagnetism, including half-metallic states in certain configurations. [3-4]

Furthermore, a mixed-topology strategy—combining different sublattice patterns—enables the design of stable, purely organic 2D ferromagnetic semiconductors characterized by substantial magnetic coupling strengths ( $J$  up to  $127$  meV) and estimated Curie temperatures well above  $500$  K, offering exciting prospects for room-temperature metal-free magnetic materials. [5]

These findings underscore triangulene-based architectures as a versatile platform for engineering correlated magnetic and electronic phenomena in organic 2D crystals.

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## References

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