Towards room temperature antiferromagnetism in all-organic modular 2D materials

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2D covalent organic frameworks are an emerging class of lightweight, flexible materials built from molecular nodes connected through covalent linkers in extended 2D networks. Incorporating stable organic radicals as nodal units yields 2D covalent organic radical frameworks (2D-CORFs), whose unpaired electrons endow these systems with intrinsic magnetic functionality. In recent computational works, we have demonstrated that such materials can host antiferromagnetic (AFM) Mott insulator ground states [1] (which may become semimetallic or closed shell via mechanical perturbations [2,3]) and that they provide a robust basis for 2D spin frustration [4] and magnetoelectric response.[5]

The magnetic exchange coupling (*J*) between spins in 2D AFM materials governs many of their properties. For instance, it sets the scale for magnon transport, where large *J* values are essential for ultrafast spintronic operation. Consequently, a central challenge in developing 2D AFM materials is the controlled tuning of these exchange interactions. The modularity of 2D-CORFs offers opportunities for chemical design beyond those available in inorganic 2D magnets. However, achieving large and predictable *J* values between localized spins in stable radical centres remains nontrivial. Here, we introduce a new design principle based on a specific class of heterocyclic linkers that enables systematic chemical control of AFM exchange in 2D-CORFs (see Figure 1). Our computational modelling demonstrates that this strategy allows access to a broad range of *J* values, including regimes compatible with robust antiferromagnetic order at room temperature. By enabling precise control of exchange interactions, this approach strengthens the case for 2D-CORFs as a versatile organic alternative for spintronic technologies.

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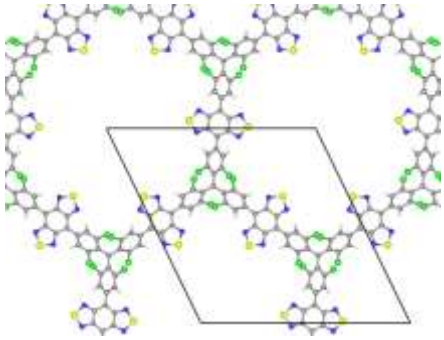

Figure 1: Honeycomb lattice-based 2D-CORF made of triarylmethyl radicals and heterocyclic linkers