

ENGINEERING QUANTUM SPIN LIQUIDS AND SINGLE-MOLECULE MAGNETS VIA STRUCTURAL MODULARITY IN 2D COORDINATION NETWORKS

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Molecule-based coordination polymers (oxalates, anilates) and two-dimensional metal-organic frameworks (2D MOFs) provide highly tunable platforms for engineering exotic quantum magnetic states [1-3]. Here, the structural modularity of these systems is utilized to engineer two distinct magnetic behaviors: Quantum Spin Liquids (QSLs) and Single-Molecule Magnets (SMMs).

To realize QSL candidates, we constructed geometrically frustrated lattices (triangular and kagomé) populated by $S_{eff} = 1/2$ ions. These include the bimetallic networks (PNP)[CoRh(ox)₃] and (PNP)[ZnRu(ox)₃] (PNP = bis(triphenylphosphine) iminium cation, ox = oxalate), as well as the 2D conductive MOF Cu₃(HHTT)₂ (HHTT = 2,3,7,8,12,13-hexahydroxytetraazanaphthotetraphene) [4]. Across these series, DC/AC magnetometry and EPR spectroscopy reveal a complete absence of long-range magnetic ordering down to 2 K. Further thermodynamic validation via heat-capacity measurements down to 50 mK exhibits no λ -type anomalies, confirming persistent spin fluctuations, with μ SR measurements planned to benchmark the low-temperature spin dynamics [3].

Conversely, to exploit SMM and potential qubit behavior, we utilized the cavities of these expanded networks to host anisotropic molecular cations. The intercalation of the SMM-active decamethylferrocenium ([FeCp₂^{*}]⁺) into a diamagnetic [ZnRh(ox)₃] and [ZnGa(AnCl₂)₃]⁻ (AnCl₂ = Chloroanilate) frameworks provide rigorous magnetic isolation, yielding field-induced slow relaxation of magnetization and single-ion qubit characteristics. Furthermore, by substituting the diamagnetic host with paramagnetic or ferrimagnetic anilate-based networks (e.g., Mn/Cr), we systematically investigate the modulation of the guest's SMM behavior via host-guest local internal magnetic fields.

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[2] Burzurí, E. *et al.* A quantum spin liquid candidate isolated in a two-dimensional CoIRhIII bimetallic oxalate network. *Chem. Sci.* **14**, 3899–3906 (2023).

[3] Pratt, F. L. *et al.* Properties of the frustrated spin 1/2 kagome material Cu₃(HOTP)₂. *Phys. Rev. Res.* **7**, (2025).

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