Single-molecule magnets (SMMs) are individual molecules that function as single-domain nanoscale magnetic particles. Recent studies suggest that they may potentially be employed as the elementary computational unit (qubit) of a quantum computer. In order to perform large scale quantum computations, the coupling of multiple SMMs to each other will be essential. However, the interaction should be relatively weak in order to maintain the intrinsic properties of each SMM.

Weak coherent coupling of pairs of SMMs has previously been demonstrated by high-field Electron Paramagnetic Resonance (EPR) [Science 302, 1015 (2003)], albeit only in cases where the coupling was crystallographically imposed. In the present investigation, high-field EPR studies have demonstrated coherent coupling within covalently linked dimers and tetramers (supramolecular aggregates) of Mn₃ SMMs. EPR spectra reveal features that are lacking in spectra of the isolated Mn₃ SMMs, which can be attributed to the coherent coupling within the aggregates. Importantly, powder and solution spectra are essentially identical, indicating that the supramolecular aggregates remain intact in solution, thus demonstrating for the first time that their unique properties survive outside of a crystal. This work opens up a new direction in the study of exchange-coupled SMM oligomers. Their robustness in solution offers a feasible means for their deposition on surfaces for device studies.

(a) Experimental and (b) simulated temperature-dependent high-field EPR spectra of a tetramer of Mn₃ SMMs. (c) to (e) Schematics representing different basis states and couplings within the tetramer.