Nuclear Spin Patterning Controls Electron Spin Coherence

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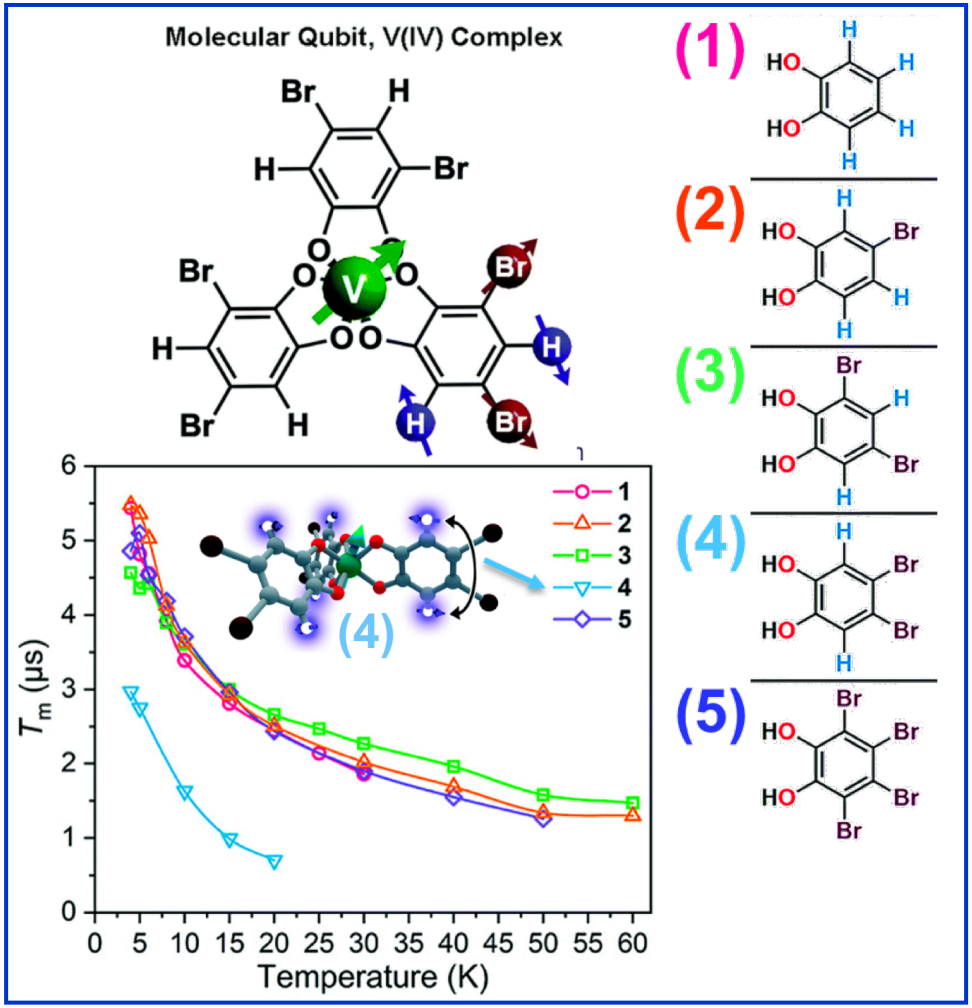
Achieving control of electron spin coherence in transition metal complexes, as measured by the quantum phase memory time, T_m, is an important goal in molecular spintronics.

In this study, users from Colorado State University provide the first evidence that nuclear-spin patterning in the ligand shell is an important handle to modulate T_m in magnetic molecules containing transition metal ions that are potential targets for next-generation quantum technologies. They synthesized and studied a series of five V^{IV} complexes with brominated catecholate ligands, $[V(C_6H_{4-n}Br_nO_2)_3]^{2-}$ (with n = 0, 1, 2, and 4), where the $^{79/81}Br$ and ^{1}H nuclear spins are arranged in different substitutional patterns (see top and right-hand figures, where the top figure has the substitution pattern "3" as labeled in the figures on the right-hand side). High-field/frequency (120 GHz) pulsed electron paramagnetic resonance spectroscopic analysis of this series reveals a pattern-dependent variation in T_m for the spin-½ V^{IV} ion (see Figure, lower-left). *Notably, it is shown that it is possible for two molecules (with substitutional patterns "3" and "4") to have values of T_m that are markedly different (by a factor of two) despite having the same chemical composition.*

NMR analyses of the protons on the ligand shell suggest that relative chemical shift, controlled by the patterning of nuclear spins, is an important underlying design principle. Having multiple protons with nearly identical chemical shifts will, ultimately, facilitate proton spin diffusion, giving rise to a shorter T_m for the bound metal ion.

Facilities and instrumentation used: EMR program, 12.5 Tesla Pulsed EPR quasi-optic heterodyne spectrometer.



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