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Massive 9 GHz Hyperfine Clock Transition in a Molecular Spin Qubit[1] KRISHNENDU KUNDU, Post-doctor, JESSICA WHITE, SAMUEL MOEHRING, JASON YU, Student, JOSEPH ZILLER, facility manager, FILIPP FURCHE, WILLIAM EVANS, STEPHEN HILL, Professor — Spins in molecules have been proposed as potential qubits in quantum computers, enabling chemical tunability of their quantum nature and potential for scaleup via self-assembly. We demonstrate chemical control on the degree of s-orbital mixing into the spin-bearing d-orbital associated with a series of spin-La(II) and Lu(II) molecules. Increased s-orbital character reduces spin-orbit coupling and enhances the electron-nuclear Fermi contact interaction. In one particular Lu(II) complex, we have observed an enormous hyperfine interaction for a molecular system, Aiso = 3467 MHz (more than 1200 G), which, in turn, generates a 9 GHz clock transition. The large magnitude of this hyperfine interaction necessitated high-field W-band EPR in order to fully characterize the electron-nuclear spin Hamiltonian parameters. Meanwhile, pulsed X-band EPR studies reveal an order of magnitude increase in phase memory time, Tm, at the 9 GHz clock transition. These findings suggest new strategies for the development of molecular quantum technologies, akin to trapped ion systems.

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