Chemical Control of Clock Frequency in a Series Molecular Lutetium(II) Qubits

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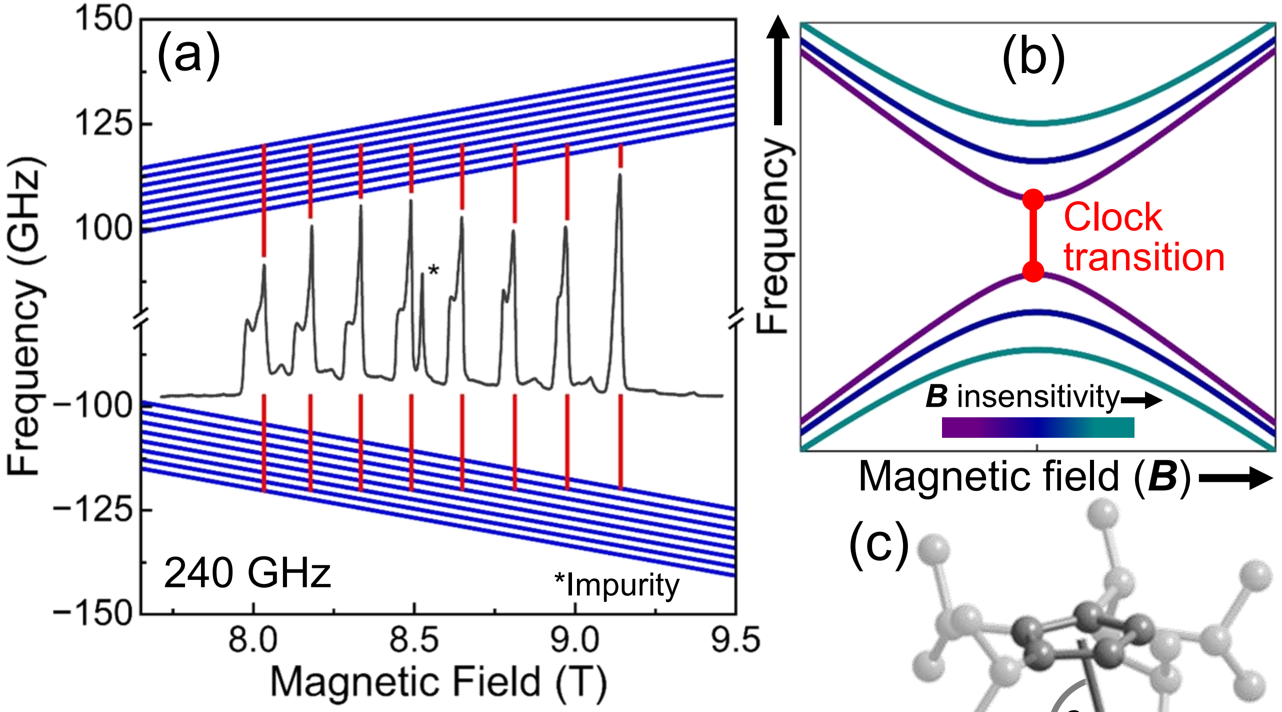
Electron spin coherence can be significantly enhanced at so-called clock transitions – optimal operating points or avoided Zeeman level crossings at which the spin resonance frequency becomes insensitive to magnetic noise. Robust spin clock transitions can be generated by enhancing the electron-nuclear hyperfine interaction. In this regard, molecular lutetium 2+ complexes are attractive because they possess a lone unpaired electron that occupies a mixed 5d/6s orbital. Chemical tuning of this mixing to maximize the 6s character and, hence, the electron spin density at the nucleus, provides a way to optimize the electron-nuclear hyperfine coupling strength.

Chemists from the Naval Air Warfare Center and UC Berkeley synthesized a family of lutetium(II) sandwich-type molecules using pairs of planar cyclopentadienyl (Cp) ligands with varying substituents on the five-membered Cp rings. In this way, they have been able to control the linearity of the chemical coordination at the lutetium ion (angle θ – see Figure) which, in turn, regulates the degree of s-orbital character. In doing so, the strongest hyperfine coupling has been achieved for any molecular lanthanide compound to date. Accurately measuring such a strong hyperfine interaction requires electron magnetic resonance measurements (EMR) to be in a high-field regime in which the Zeeman interaction dominates. Hence, measurements could only be performed using the unique spectrometers at the MagLab (see Figure).

These results outline a general strategy toward the isolation of paramagnetic molecules with strong s-orbital electronic character, leading to a massive hyperfine interaction and a simple electronic ground state with excellent coherence. Such molecules are of interest as spin qubits for next-generation quantum technologies.



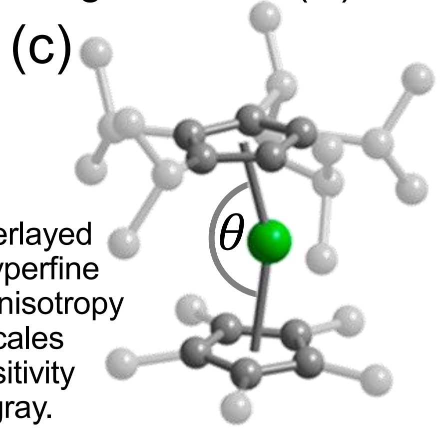


Figure. (a) High-field EPR spectrum of a powder sample overlayed on Zeeman level diagram depicting transitions between 8 hyperfine spin "up" and "down" sub-levels; fine structures are due to anisotropy in the powder. (b) Schematic low-field clock transition gap scales with hyperfine coupling strength, leading to increased insensitivity to ***B***-field. (c) $\mathrm{Lu(Cp^{Me_5})(Cp^{\mathit{i}Pr_5})}$ molecule; Lu – green, C – gray.

Facilities and instrumentation used: EMR Facility, 12.5 T Heterodyne Quasi-Optical Spectrometer.

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