Massive Hyperfine Interaction in a Lu(II) Qubit

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Electron spin coherence, important in quantum information science, can be significantly enhanced at so-called clock transitions (CTs) – optimal operating points that are immune to magnetic noise. CTs can be generated via the hyperfine interaction (HFI) between electron and nuclear spins in atoms. Maximizing this interaction to achieve optimum coherence and a desirable operating frequency in the microwave regime requires enhancing electron spin density at the location of the nucleus, which can be achieved using coordination chemistry.

Chemists from UC Irvine prepared a family of molecules containing a single lanthanide (Ln = La or Lu) ion in the rare 2+ oxidation state. In each case, a single unpaired electron resides in a mixed 5d/6s orbital. The HFI can be controlled through choice of: (i) coordinating ligand, which tunes the s-orbital character and, hence, the spin density at the nucleus; and (ii) Ln ion, with increased orbital contraction for heavier Lu. As seen from the peak spacing in the high-field echo-detected ESR spectra (Figure), significant control of the 5d/6s mixing is achievable, with $[Lu^{II}(OAr^*)_3]^-$ exhibiting a massive 3.47 GHz HFI (c.f. < 100 MHz is typical for related molecules).

Hyperfine CTs are currently employed in more mature trapped ion quantum computers. This investigation demonstrates that the same approach is viable in molecular qubit platforms, giving rise to measurably enhanced coherence and prospects for future scalability via chemical self-assembly.

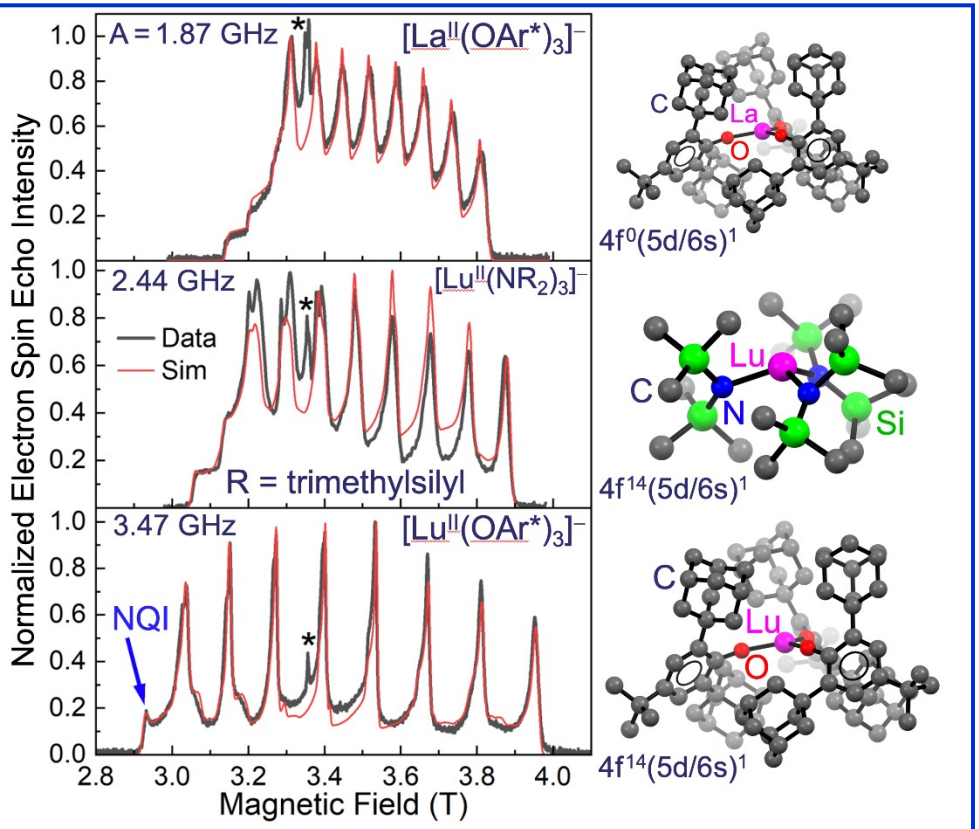

Figure. Pulsed electron spin-echo spectra recorded at 94GHz and 5K, for the three compounds shown. The eight (= 2*I* + 1) lines arise due to the hyperfine interaction (HFI) with the $I = {}^7/_2$ ^{175}Lu or ^{139}La nuclear spin; the spacing between peaks is a direct measure of the HFI strength, A, which increases from top to bottom. The asterisk marks an impurity and the "NQI" feature is attributed to a strong nuclear quadrupole interaction.

Facilities and instrumentation used: EMR (W-Band HiPER Pulsed ESR Spectrometer); **Citation:** Kundu, K.; White, J.R.K.; Moehring, S.A.; Yu, J.M.; Ziller, J.W.; Furche, F.; Evans, W.J.; Hill, S., *9.2-GHz clock transition in a Lu(II) molecular spin qubit arising from a 3,467-MHz hyperfine interaction,* **Nature Chemistry**, **14**, 1-6 (2022) doi.org/10.1038/s41557-022-00894-4 - Data Set