Rational Synthetic Approaches to Electronic Spin Decoherence

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Introduction

In 2001, Leuenberger and Loss proposed high-spin molecules as potential qubits for quantum computation.[1] Studies inspired by this proposal have exposed generally short lifetimes for the superposition states of electronic spins.[2] The lifetimes of electronic spin qubits will ideally be extended if such systems are ultimately to find use as molecule-based processors. Decoherence can be instigated by a variety of sources; one of the most dominant arises from the qubit interaction with surrounding, fluctuating nuclear spins. In pursuit of long superposition lifetimes in molecular species, we synthesized and studied the new molecule $[Cr(C_3S_5)_3]^{3-}$ by pulsed EPR methods. The elements that compose this molecule are C, S and Cr, which have, respectively, isotopic natural abundances which are 98.9, 99.2, and 90.5 % nuclear spin-free, respectively.

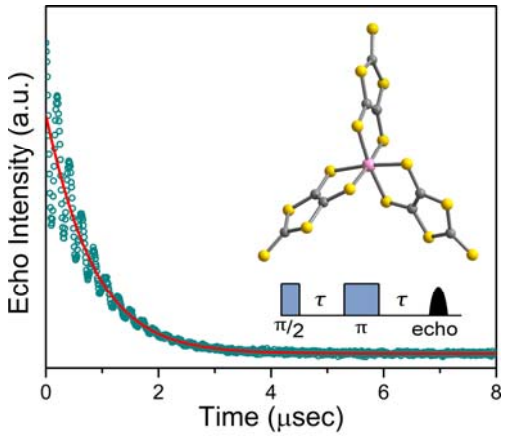

Fig. 1 Hahn-echo decay curve at X-band for a 1 mM solution of (**1**) in BuCN at 5 K and H_{dc} = 2180 G. The red line is a fit to an exponential decay with T_2 = 842 ns. Hahn-echo pulse sequence and molecular structure of $[Cr(C_3S_5)_3]^{3-}$ are depicted. Pink, yellow, and gray spheres are chromium, sulfur, and carbon atoms, respectively.

Experimental

Suitable glassing solutions of $(Me_4N)_3[Cr(C_3S_5)_3]$ (**1**) for analysis were made with butyronitrile (BuCN). Pulsed EPR studies were performed at the NHMFL EMR facility either at X-band (9.7 GHz) with a Bruker Elexsys E680 spectrometer or a custom built 120/240/336 GHz spectrometer[3] and its 12.5 T SC magnet.

Results and Discussion

The superposition lifetimes (coherence times, T_2) for **1** were determined by application of two-pulse Hahn-echo sequences at 5 K at applied fields corresponding to resonances observed in the continuous-wave and echo-detected X-band EPR spectra (see Fig. 1). Under these conditions, an oscillating decay of echo intensity yields a T_2 of 842 ns for **1**. Dilution of **1** within an isostructural diamagnetic analogue increases T_2 by a factor of two, leading to T_2 = 1.6 μs at the same temperature. The spin-lattice relaxation time (T_1) collected at 240 GHz and 1.5 K is 150 μs, while the T_2 is 1.2 μs and drops dramatically with increasing temperature, leading to a complete loss of echo intensity above 2 K. These T_1 and T_2 values, while modest in the context of other molecular qubits[2], still permitted the observation of Rabi oscillations via nutation experiments. The presence of oscillations here indicate that the pair of spin states being probed by the irradiating microwaves can assume any superposition of the two states, a requisite capability for any potential qubit.

Conclusions

The foregoing results reveal relatively short T_2s in $[Cr(C_3S_5)_3]^{3-}$ despite the scarcity of nuclear spin. The strongly temperature-dependent echo suggests that thermal mechanisms underlie decoherence to a large extent. Our future efforts are two-fold. In the near term, we will isolate **1** in a deuterated medium to explore the influence of surrounding nuclear spins on the magnitude of T_2. Longer term goals will pursue molecular fine-tuning to engineer longer T_1 times and thus longer T_2s in this and other nuclear spin-free, paramagnetic complexes.

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References

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