High-frequency EPR Study of Coherence Times in Oxovanadyl TPP Single Crystals

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Introduction

Molecular spin ensembles have shown great promises for applications in quantum information processing due to their scalability which has led to multidisciplinary efforts to prolong their coherence times at high temperatures [1,2]. Single ion nuclear spin free ligand vanadyl-based samples are some of the most well-known molecular spin qubits showing long coherence times [3,4]. Here we investigate the oxovanadyl tetraphenyl porphyrin (VOTPP), a molecular derivative of recent synthesis diluted in an isostructural TiOTPP diamagnetic analogue.

Experimental

Single crystals of VOTPP in TiOTPP with 1% and 10% concentration were investigated using pulsed EPR spectroscopy at low temperatures (1.5 - 10 K) and at frequencies of 120 and 240 GHz. The crystals were aligned with the static magnetic field parallel to the VO bond (**Fig.1a**). The measurements were carried out using the heterodyne quasi-optical spectrometer and its 12.5 T SC magnet at the EMR facility.

Results and Discussion

The field-swept echo detected spectra collected at 240 GHz (**Fig.1 a**) show eight resonances corresponding to the hyperfine coupling of the *S*=1/2 electron spin associated with the VO^{2+} group, and the *I*=7/2 nuclear spin of the vanadium [3,4]. The coherence times (T_2) measured for resonance #A are above 1 μs across the investigated temperature range, and reach ≈8 μs at 240 GHz and 1.5 K (**Fig.1b**). Similar coherence times are measured for resonance #B at 120 and 240 GHz (not shown). As shown in **Fig.1c,** increasing the concentration by a factor of 10 does not alter the relaxation times significantly which can be attributed to the high degree of spin polarization induced by the large applied fields.

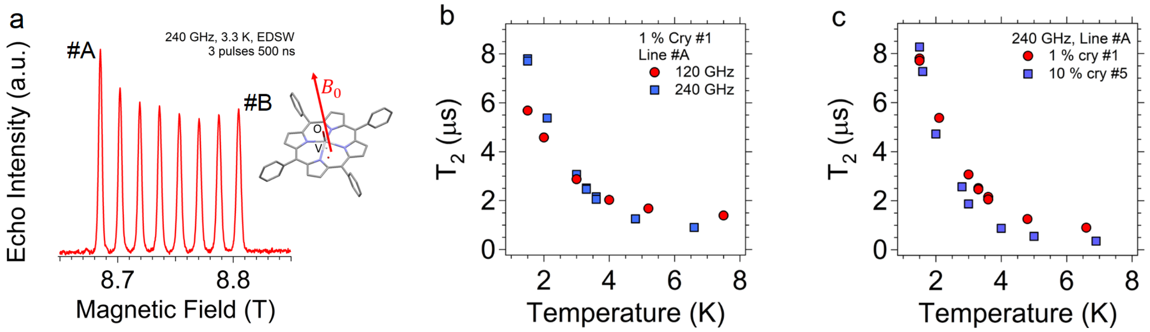

Fig.1 (a) Field-swept echo detected spectra for the 1% VOTPP sample at 240 GHz and 3.3 K. The inset shows the VOTPP molecule with the static magnetic field (B_0) parallel to the VO bond. Coherence times are studied at the two resonances marked with #A and #B. (b) Temperature dependence of T_2 for the 1% sample at 120 and 240 GHz for resonance #A. (c) Temperature dependence of T_2 for the 1% and 10% concentrations at 240 GHz for resonance #A.

Conclusions

The vanadyl compounds studied here show remarkably long spin-spin relaxation times at low temperatures, despite the relatively high concentration of vanadium in the investigated samples.

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