Controlling Spin-Spin Relaxation T_2 of a Triatomic Single Molecule Magnet: $Cr_3(dpa)_4Cl_2 \cdot CH_2Cl_2$

J. Wang, Z. Wang, N.S. Dalal (FSU, Chemistry & NHMFL); J. Van Tol, A. Ozarowski (NHMFL)

Introduction

Controlling spin decoherence time, T_2, in molecular magnetic systems is of high interest for quantum information processing [1]. Using a single crystal containing two or more symmetry related sites in the unit cell, we demonstrated a method for controlling T_2 by optimizing the crystal orientation in the Zeeman field in a high field pulsed electron paramagnetic resonance measurement. The sample employed is a trichromium(II) complex $Cr_3(dpa)_4Cl_2 \cdot CH_2Cl_2$, with total spin S = 2 [2,3], studied at 1.5 K, at 240 GHz. T_2 could be varied via change in the crystal orientation from < 0.5 μs to up to 2.6 μs in *ab* plane, as shown in **Figure 1**(b). The observations are in good agreement with an analysis based on spin-exchange between dipole-coupled neighboring molecules in an electron spin bath. To further confirm this conclusion, spin-spin relaxation time T_2 in another plane such as *ac* plane is desired, since two site chains are equivalent in this plane and no angular dependence of T_2 is expected if it's aligned exactly in *ac* plane.

Experimental

Spin-spin relaxation time T_2 measurements were made using home-built pulsed superheterodyne EPR spectrometer with a superconducting magnetic field up to 12 T at the National High Field Magnetic Laboratory, Tallahassee, Florida.

Results and Discussion

A single crystal of $Cr_3(dpa)_4Cl_2 \cdot CH_2Cl_2$ was face-indexed by XRD and was mounted in a way so that the Zeeman field is parallel to *ac* plane when the crystal is rotated. Indeed, much weaker angular dependence of T_2

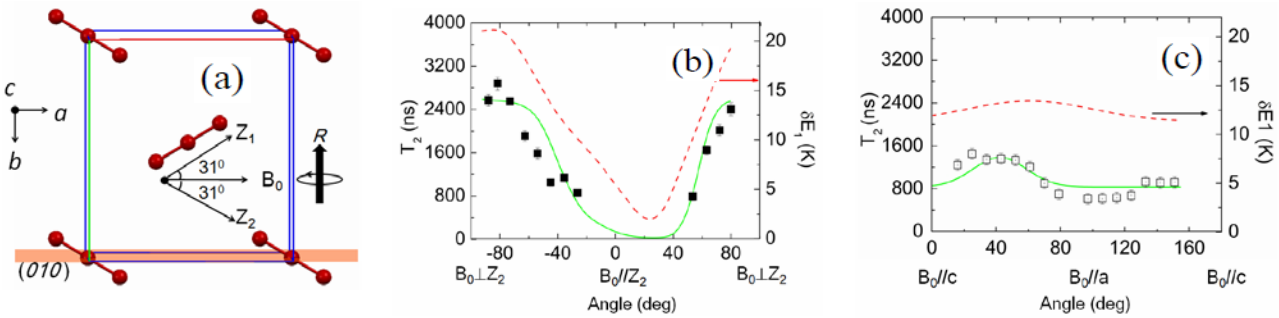

Figure 1. (a) unit cell packing of $Cr_3(dpa)_4Cl_2$; b) angular dependence of T_2 resonant in *ab* plane; (c) ac plane.

was observed in this rotation plane (Fig. 1 (c)), even though the rotating plane was about 10^0 off the *ac* plane in the actual experiment. T_2 varied from ~1.4 μs to 0.7 μs when the crystal was rotated, as compared to the T_2 range of ~2.6 μs to less than 0.5μs in *ab* plane (Fig. 1(b)). The energy difference of the non-resonant spins also displayed much less angular dependence, as shown in the red dashed curve in both (b) and (c).

Conclusions

The comparison of T_2 time between *ab* and *ac* planes provides further support to the possibility of tuning T_2 relaxation in *ab* plane by changing the orientation of magnetic field relative *Z* axes of *D* tensor. These results point to a novel way of controlling the spin decoherence time.

Acknowledgements

This work is supported in part by FSU. The National High Magnetic Field Laboratory is funded by the NSF through Cooperative Agreement DMR-0654118, by the State of Florida and by the DOE.

References

[1] Leuenberger, M.N., *et al.*, Nature, **410**, 789-793 (2001).
[2] Berry, J.F., *et al.*, J. Am. Chem. Soc., **126**, 7082-7096 (2004).
[3] Wang. J., *et al.*, Polyhedron, **30**, 3058-3061 (2011).