Study of the Spin-Orbit Coupling and Fluctuations in V_{15} Molecular Magnet Using High-Field Electron Spin Resonance

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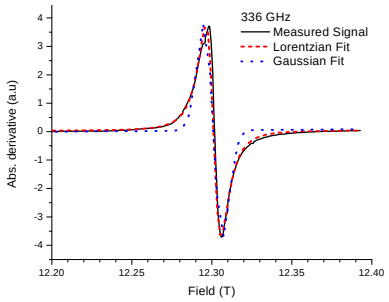
Introduction

Following the preceding publication on the precise determination of the spin Hamiltonian parameters of V_{15} magnetic molecule [1], we focused our attention on the source of spin decoherence in this system. The V_{15} molecule [2] comprises 15 spins 1/2 coupled for a total spin 1/2, which is a system that recently showed quantum Rabi oscillations of its total quantum spin [3]. This molecule is an essential system for advancing molecular structures for quantum computing and therefore understanding its spin decoherence is of paramount importance.



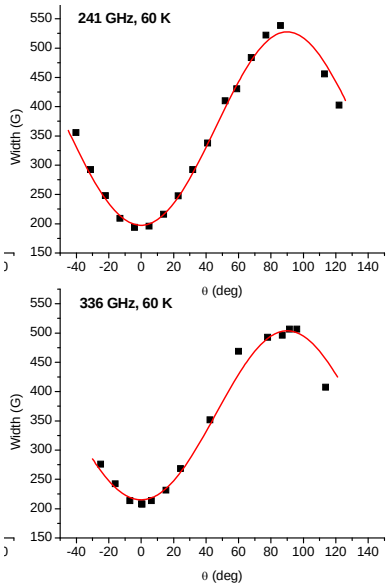


Fig. 1. Top: ESR lineshape of the V_{15} resonance. **Bottom**: Linewidth angular periodicity (squares) and simulations (in red) using a g-fluctuations based model.

Experimental

The experiments were done at the NHMFL EMR facility using high frequency ESR at 120 GHz, 241 GHz and 336 GHz and the 12.5 T SC magnet. Aside the study of resonance fields which give the spin Hamiltonian [1], we analyzed the shape and orientation dependence of the resonance linewidths. An essential aspect is that all lines have Lorentzian shapes (see Fig.1 top). The coil calibration was done by adding a small BDPA sample on the same sample holder.

Results and Discussion

We study how fluctuations in each term of the spin Hamiltonian contribute to the spin decoherence with much attention given to the spin-orbit coupling that generates g-factor anisotropy. The simulations (red lines) in Fig.1 bottom show the effect of a σ_g fluctuation on the resonance fields. Using spin-orbit theory, σ_g can be linked to the energy separation between the 3d V orbitals. The size of σ_g and thus of the spin-orbit fluctuations were studied as a function of temperature. It is observed that the calculated variations in the g-factor have a minimum value at the lowest temperatures (~3 K) and reach a saturation in our temperature range of up to ~60 K. The larger the Zeeman splitting, the higher the saturation temperature. Such behavior can be interpreted using a spin-flop model in which the thermal population of the excited Zeeman level generates spin-up / spin-down fluctuations. The energy difference due to the Zeeman effect slightly affects the energetic gap between consecutive 3d orbitals. Therefore, the g-factor fluctuations and subsequent linewidth, show the oscillatory behavior of Fig. 1 bottom, characteristic of the anisotropic g-factor observed in this system.

Conclusions

We have studied the anisotropy of the spin Hamiltonian of the molecular magnet V_{15}. The experiment achieved (i) a highly precise measurement of the g-factor anisotropy and (ii) identification of the molecule c-axis.

Acknowledgements

A portion of this work was performed at the NHMFL, which is supported by NSF Cooperative Agreement No. DMR-1157490, the State of Florida, and the U.S. Department of Energy. This work was supported by NSF grant No. DMR-1206267.

References

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[3] Bertaina, S., *et al.,* Nature, **453**, 203 (2008).