Study of the V15 Molecular Magnet Spin Hamiltonian Using High-Field Electron Spin Resonance

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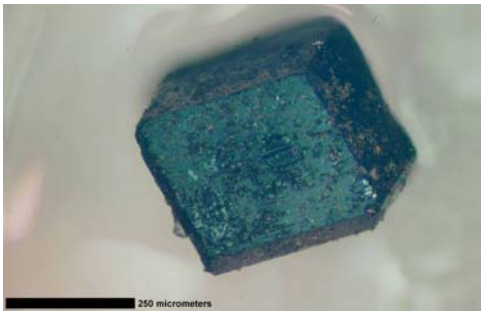
Introduction

We have studied molecular magnets using high field/high frequency Electron Spin Resonance. Such molecular structures contain many spins linked by exchange interactions and consequently their energy structure is complex and requires a good understanding of the molecular spin Hamiltonian. In particular, we studied the V_{15} molecule [1], comprising 15 spins 1/2 with a total spin 1/2, which is a system that recently showed quantum Rabi oscillations of its total quantum spin [2]. This molecule is an essential system for advancing molecular structures for quantum computing.



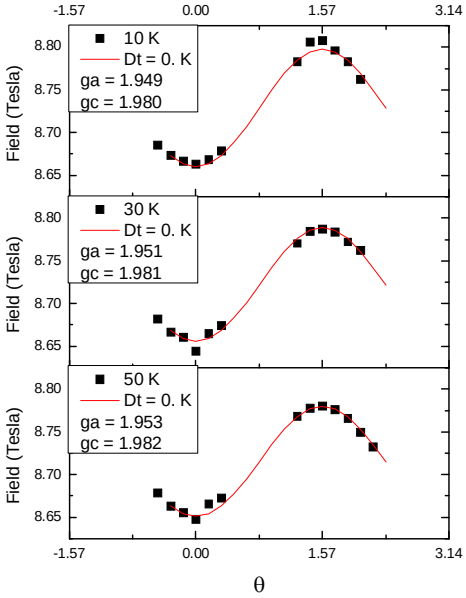


Fig 1. Top: Photograph of the V_{15} single crystal used in measurements. **Bottom**: Resonant fields (squares) vs rotation angle (radians) and simulations (in red) using an anisotropic spin Hamiltonian.

Experimental

We used high frequency characterization techniques (of hundreds of GHz) to gain insight into the anisotropy of the exchange interactions, crystal field and of anti-symmetric interactions [3] in this system. Experiments were done in the EMR Facility using the 12.5 T heterodyne instrument. We analyzed the data using a detailed numerical analysis of spin interactions. The Hamiltonian comprises g-factor anisotropy and a Dzyaloshinky-Moriya interaction term.

Results and Discussion

The precise nature of the high field experiments allowed us to measure the resonance fields of the molecule with high resolution. The coil calibration was done by adding a small BDPA sample on the same sample holder. The crystal used had a well defined shape (see Fig.1 top) and we thus could search for the orientation of the molecule's c-axis by respect to the general shape of the crystal. The V_{15} molecule is formed by 2 hexagons sandwiching a triangle and the anisotropy c-axis is perpendicular to the triangle plane. Resonance fields measured at 240 GHz are shown in Fig.1 bottom, for 3 temperatures. The red line shows the results of our simulation. We measured the temperature dependence of the minimum and maximum resonance field and our simulations are able to reproduce the experimental data.

Conclusions

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

Acknowledgements

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References

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