Coherent Manipulation of Spin States of the Molecular Cluster V_{15}

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Introduction

The control of dynamics of spins in solid-state materials has direct implications at fundamental and applied levels. Research topics, in particular quantum computing, rely heavily on complex control techniques and long spin coherence times to achieve robust information control. In this context, molecular magnets gained significant attention in the recent years, both for their technological potential and as test-bed for quantum mechanics at macroscopic scale. The V_{15} molecule, with the chemical formula $K_6[V_{15}{}^{IV}As_6{}^{III}O_{42}(H_2O)]^{6-}\bullet 8H_2O$ and shown in **Figure 1a, b**, was among the first to show coherence via direct measurements of Rabi oscillations. The observation of spin coherence in molecular magnets is not limited to the V_{15} complex; other molecules being recently studied in systems like $CrNi_7$, Fe_8, Fe_4, which shows the high interest that these systems gained recently. We performed ESR experiments at low temperature, high fields, high frequency and in variable magnetic field orientation to study the possibility of better parametrization of the spin Hamiltonian, compared to ones done at low field, static or ac magnetic measurements.

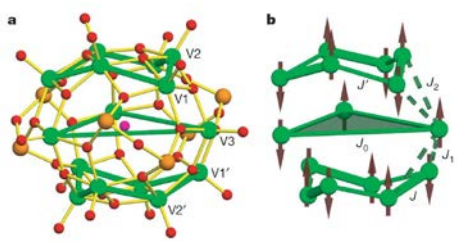

Figure 1. (a) Ball-and-stick representation of the V_{15} molecule (V-green, As-orange, O-red and in purple the O from an encapsulated H_2O group). The V ions are numbered to distinguish the different AF couplings (e.g. J_1 between V_3 and V_1 and J_2 between V_3 and V_2). (b) A sketch showing the 5 direct AF coupling and the J_0 coupling of the middle triangle.

Experimental

By means of pulsed ESR, we investigated the levels spacing and crossings of the V_{15} Hamiltonian ESR in high fields/high frequency. We used the unique setup available in Dr. J. van Tol's laboratory, where the sample can be rotated and Hamiltonian anisotropy can be explored.

Results and Discussion

The spectroscopy studies showed the high intensity signal that can be recovered from a non-diluted V_{15} single crystal. Tiny samples were used to ensure clear signals. The field dependence of the resonance signals does agree well with previous studies on this molecular complex. In addition, the setup at NHMFL made possible to have a new insight into the anisotropy of this system. At 240 GHz, the sample is rotated and the resonance field shows an oscillatory behavior, as depicted in **Figure 2**. This is the first observation of such phenomena in the molecular magnet V_{15} and is due to the site anisotropy and molecule symmetry. At the same time, the linewidth of the resonance oscillates in a very similar fashion (not shown). A more in depth theoretical analysis, using the full molecular Hamiltonian is required, for the purpose of clarifying the Hamiltonian parameters. A fit to current data may improve the knowledge on the Dzyaloshinky-Moriya coupling leading to tiny zero field splittings.

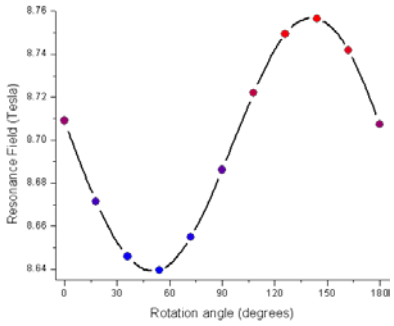

Figure 2. The figure shows oscillation of the resonance field at 240 GHz as a function of the rotation angle of the crystal by respect to the static field axis.

Conclusions

The current data give a new insight into the site and molecule anisotropy/symmetry. The future theoretical simulations/fits will hopefully bring some clarifications about the spin Hamiltonian of this molecular system.

Acknowledgements

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References

[1] Bertaina, S., *et al.,* Nature, **453**, 203-206 (2008).