Magnetization Studies on Single Crystals of Mononuclear S = 1 $Ni(HF_2)_2(3,5\text{-lutidine})_4$

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Introduction

$[Ni(HF_2)(pyz)_2]SbF_6$ (pyz = pyrazine = $N_2C_4H_4$) was recently synthesized and its crystal structure determined to consist of a tetragonal MOF-like 3D network. This network can be deconvoluted into linear Ni-FHF-Ni chains along the *c*-axis that are cross-linked by pyz ligands to form 2D square lattice propagating in the *ab*-plane [1]. Evidence provided by various magnetic property measurements establish quasi-1D magnetic exchange along the Ni-FHF-Ni chains (J_{1D}) with a much weaker coupling along Ni-pyz-Ni (J_{pyz}). Despite this, a relatively high T_N of 12.2 K was found which is the highest such temperature observed in materials of this type. Complications in data analyses stemming from the lack of single crystals, as well as crystal field splitting (D) of the octahedral Ni(II) ion, necessitated studies of model compounds that featured negligible intermolecular spin correlations between Ni(II) ions all-the-while retaining the same tetragonal NiF_2N_4 crystal field. Thus, $Ni(HF_2)_2(3,5\text{-lutidine})_4$ was synthesized and shown to contain axial (and linear) HF_2^- ligands with 3,5-lutidine ligands attached to the equatorial positions. As designed, molecules pack along the *c*-axis to form pseudo-chains with large intermolecular spacings.

Experimental

Magnetization studies were provided by a 65 T short-pulse magnet located at the Pulsed Field Facility, National High Magnetic Field Laboratory, Los Alamos, NM. The susceptometer was placed within a ^{3}He cryostat providing temperatures down to as low as 0.5 K. The field B was measured by integrating the voltage induced in a ten-turn coil calibrated by observing the de Haas–van Alphen oscillations of the belly orbits of the copper coils of the susceptometer.

Results and Discussion

Magnetization measurements (**Figure 1**) conducted on oriented single crystals of $Ni(HF_2)_2(3,5\text{-lutidine})_4$ revealed strongly contrasting behaviors. With the field applied parallel to the chain-axis a relatively sharp transition, reminiscent of a singlet-triplet excitation, occurs at ~7.5 T with the fully saturated state being achieved near 10 T. The feature is unexpected as the crystal structure does not contain spin-dimers. Rotating the field into the *ab*-plane leads to Brillouin-like behavior and disappearance of the step-like transition seen in the other orientation.

Conclusions

The data presented here are consistent with extremely weak magnetic interactions as intended. However, when the magnetic field is aligned parallel to the crystal stacking axis a sharp step-like feature is observed which vanishes by ~4 K. Such behavior is inconsistent with Bose-Einstein condensation of magnons but may originate from a different quantum magnetic state.

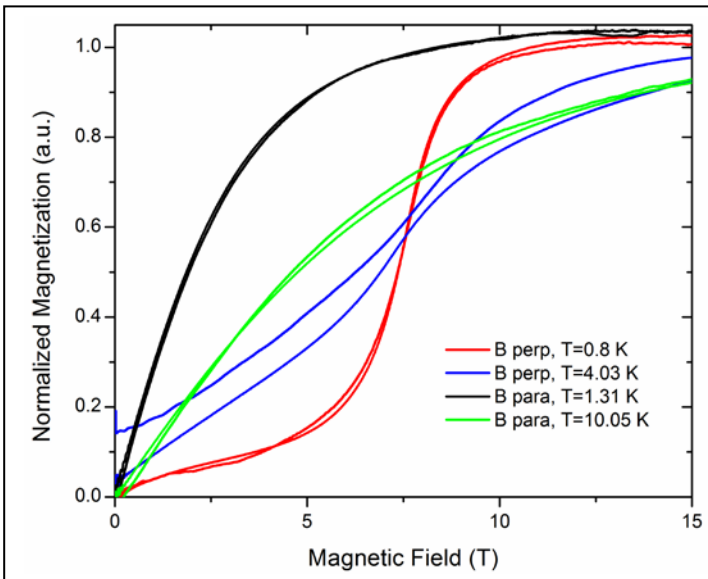

Figure 1. Magnetization data for $Ni(HF_2)_2(3,5\text{-lutidine})_4$ measured parallel and perpendicular to the *c*-axis.

Acknowledgements

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References

[1] J.L. Manson, *et al.*, *Inorg. Chem.* **2011**, *50*, 5990.
[2] J.L. Manson, *et al.*, manuscript in preparation.