Evolution of Magnetism in a Jahn-Teller Driven Magnetic Dimensionality Switch

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Introduction

Of interest is the effect of pressure on polymeric magnets based on hetero-ligand Jahn-Teller (JT) active metals centers, where the transition metal ion sits in a position of octahedral symmetry, surrounded by an asymmetric ligand environment. In such systems each unique metal-organic ligand provides an additional degree of freedom on the JT-axis, meaning that small perturbations of the metal-ligand environment can be enough to switch the JT-axis and radically modify the material properties.

Recently it has been shown that $CuF_2(H_2O)_2(pyz)$ (see **Figure 1**a) undergoes two structural phase transitions as a function of pressure, accompanied by a switching of the JT-axis and orbital orientations. With the application of pressure, the elongated JT switches from the N–Cu–N to the O–Cu–O direction at 9 kbar [1]. At ambient pressure this material is a quasi-two-dimensional antiferromagnet (AFM), and so it has been proposed that this JT-axis switching and orbital reorientation may lead to a dramatic change of the magnetic behavior.

Experimental

The magnetization of single crystals of $CuF_2(H_2O)_2(pyz)$ was measured up to 35 T (NHMFL, Cell 12) using a novel radio-frequency technique [2]. All measurements were performed at 1.4 K, with the applied magnetic field parallel to the Cu–pyz–Cu chain direction (*a*-axis). A piston cylinder cell (MP35N) was used to achieve the pressure, which was calibrated in-situ using the luminescence of ruby.

Results and Discussion

$CuF_2(H_2O)_2(pyz)$ was found to be a two-dimensional AFM up to 6 kbar, with an exchange energy of approx. 11.4 K, which is reduced gradually with pressure due to the rotation of the pyrazine molecules. We observe a dramatic drop in the critical field and exchange energies at 9 kbar, a direct confirmation of a transition from two-dimensional to one-dimensional magnetism, due to the reorientation of magnetic orbitals associated with the JT-axis. This results in the primary exchange being mediated by pyrazine molecules via the Cu–pyz–Cu chain, thus allowing one-dimensional AFM to evolve. In this phase, we also observe a steady increase in the exchange energy *J* with pressure, which we attribute to a reduction in the Cu–N bond lengths.

Conclusion

We have shown that pressure can be used to drive a JT switch in $CuF_2(H_2O)_2(pyz)$, leading to an abrupt reduction in the magnetic dimensionality. It was demonstrated that pressure can be used to take control of the magnetic properties, providing the ability to tune the exchange energy *J* from 11.4 K to 5.5 K.

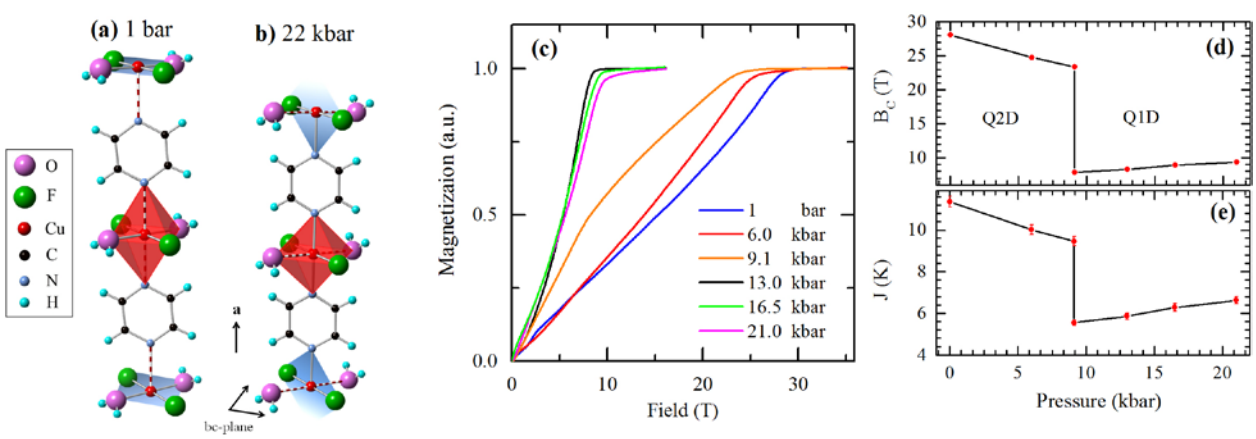

Figure 1. (a), (b) Crystal structure along the *a*-axis [1], showing the JT octahedra (red) and the plane of the magnetic orbitals (blue). Also shown is (c) the magnetization, (d) the critical field and (e) the exchange energy *J,* as a function of pressure.

References

[1] Halder, G.J., *et al*., Angew. Chem. Int. Ed., **50**, 419 (2011).
[2] Ghannadzadeh, S., *et al*., Rev. Sci. Instrum., **82**, 113902 (2011).