Evolution of Magnetism in a Series of Pressure-Induced Jahn-Teller Axis Switches

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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.

$CuF_2(H_2O)_2$(pyz) (see Fig. 1a) undergoes two structural phase transitions as a function of pressure, accompanied by a switching of the JT-axis and orbital orientations [1]. At ambient pressure this material is a quasi-two-dimensional antiferromagnet (AFM). Previous work at the NHMFL by us [2] has shown that the JT-axis change and orbital reorientation leads to a dramatic change of the magnetic behavior at 9 kbar, causing a sudden switch from two-dimensional to one-dimensional magnetism. It has been proposed that the second structural transition at 31 kbar may also lead to a similar change in magnetic properties.

Experimental

The magnetization of single crystals of $CuF_2(H_2O)_2$(pyz) was measured in a Physical Properties Measurement System, using a radio-frequency magnetometer. All measurements were performed at 1.8 K, with the applied magnetic field parallel to the Cu–pyz–Cu chain direction (*a*-axis). A diamond anvil cell [3] was used to achieve pressures of up to 56 kbar, 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 30.9 kbar, with an exchange energy of approximately 6 K, which is increased gradually with pressure due to the reduction of the Cu–N bond lengths. We observe a dramatic increase in the critical field and exchange energies at 30.9 kbar, at the same pressure as that of the structural transition and JT-axis reorientation. The reason for this sudden increase in *J*, in light of the relatively small change in bond lengths of the primary exchange pathway Cu–pyz–Cu (*a*-axis) at the structural transition, is still under investigation.

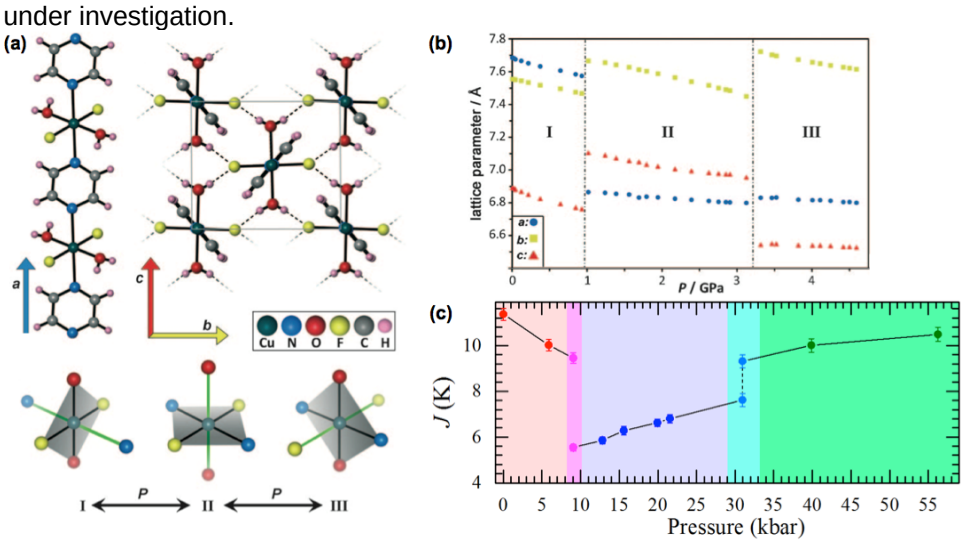

Figure 1. (a) The crystal structure and (b) the lattice parameters as a function of pressure [1]. Also shown is (c) the evolution of exchange energy *J* with pressure.

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References

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