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Abstract of Master's Thesis, Submitted 8 September 2015:

Magnetic properties of the $Mn_3O(Et\text{-}sao)_3$ molecular magnet

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A Single Molecule Magnet (SMM) is a molecule that exhibits magnetic properties similar to bulk magnets, such as slow relaxation of the magnetization and magnetic hysteresis, but at the molecular level. Their magnetic bistability, due to the energy barrier to the spin-reversal, makes this class of materials interesting for the potential technological application in high-density information storage devices.

The aim of this thesis is to investigate the magnetic properties of the $Mn_3O(Et\text{-}sao)_3$ molecular magnet, that we found to possess the characteristic properties of a SMM, with the exception of low temperatures.

Oxide-centred Mn^{III} triangles represent the basic building blocks from which a plethora of polymetallic clusters with interesting physical properties are constructed. In particular, they are the building blocks for the Mn_6 SMM which held until recently the record for the highest anisotropy barrier cluster made of transition metal ions.

The most common strategy to increase the anisotropy barrier is to have a large spin ground state. It has been found that in Mn^{III} triangles the exchange can be switched from antiferromagnetic (low spin ground state S=2) to ferromagnetic (high spin ground state S=6) via targeted structural distortion. We chose to study the high-anisotropy ferromagnetic $Mn_3O(Et\text{-}sao)_3$ triangle with the objective to investigate the role played by magneto-structural correlations in its magnetic behaviour. We found that structural changes, via the application of an external pressure as a means to alter the geometry of the molecule, can tune the magnetic super-exchange interaction between Mn ions.

Inelastic Neutron Scattering (INS) technique has been used to directly access the energies of the different spin multiplets of $Mn_3O(Et\text{-}sao)_3$, allowing us to describe its magnetic excitations and to accurately estimate the parameters of the Spin Hamiltonian, i.e. the exchange parameter J and the axial anisotropy parameter D at ambient pressure. Further INS measurements, with an external pressure applied to the sample, show that structural distortions can affect the value of the dominant term of the spin Hamiltonian, i.e. the exchange interaction. By applying an external hydrostatic pressure of up to 12 kbar we were able to decrease the exchange parameter J of about 60 μeV, i.e. a sixth of its value at ambient pressure. Complementary information were extracted from magnetic DC and AC susceptibility measurements. The former suggested the presence of a long range anti-ferromagnetic order, evidenced by a peak in the low-temperature DC susceptibility, while the latter allowed us to better characterise the magnetic properties of Mn3, determining its relaxation time constant τ_0 and the effective height of the anisotropy barrier to reorientation of the magnetisation. The effective anisotropy barrier measured experimentally is considerably higher than the one deducted from the model spin Hamiltonian, giving an indication of the influence of the low lying excited spin states in the relaxation mechanism.

Neutron diffraction experiments were performed in combination with the INS study with the aim of correlating the structural changes in the geometry of the molecule with the changes in the isotropic exchange parameter. The neutron diffraction data collected above and below the temperature of the peak observed in the DC susceptibility data evidenced new emerging peaks at low temperature, a clear indication of the onset of long range anti-ferromagnetic order. The preliminary analysis of the neutron diffraction data provided additional information about the direction of AFM ordering, which comes out to be along the *c* crystallographic axis.