

Experimental Report

11/09/2014

Proposal:	5-51-498	Council:	4/2014
Title:	Spin and charge densities in simple water-bridged TM-dimer complexes using a combination of polarized neutrons and X-ray diffraction		
This proposal is a new proposal			
Research Area:	Chemistry		

Main proposer:	OVERGAARD Jacob
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Samples:	C40 H78 Co2 O17 C42 H72 N2 Ni2 O13 C42 H72 Co2 N2 O13
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Instrument	Req. Days	All. Days	From	To
D3 High field >1T	18	18	28/08/2014	15/09/2014
D9	6	6	15/09/2014	19/09/2014

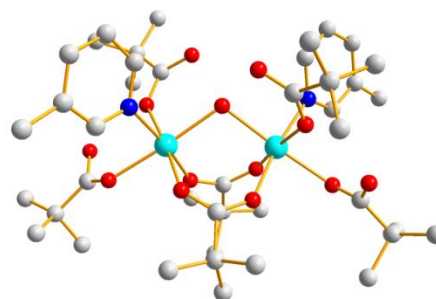
Abstract:
In this proposal our intention is to collect flipping ratios in order to obtain magnetisation densities. The group of compounds that we focus on in this work are structurally simple water- and pivalate-bridged transition metal dimer complexes. Despite their structural similarity, these compounds display fundamentally different magnetic coupling schemes such that i.e. analogues with Co may be ferromagnetically coupled while the Ni analogue is antiferromagnetic. With different ligands, the situation may be reversed. In parallel to these experiments we are already carrying out single-crystal X-ray diffraction with the same compounds at very low temperature at a dedicated synchrotron beam line at SPring8 and with the flipping ratios at hand we will co-refine multipole models of spin and charge densities (directly from measured flipping ratios and structure factors) for these compounds. The properties of these densities, derived using for instance Atoms in Molecules tools, will be correlated to the extensive magnetic properties already measured on this class of compounds, with the hope to see new connections between magnetism and spin/charge densities.

Preliminary report for the experiment entitled “Spin and charge densities in simple water-bridged TM-dimer complexes using a combination of polarized neutrons and X-ray diffraction”

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The experiment is currently being carried out and the present temporary report is primarily being written to enable the submission of a continuation proposal. The experiment was the first to be executed following the extended shutdown period of the reactor at ILL, and was therefore always in danger of being affected by initial problems. Alas, problems did materialize, as the reactor could only be run at reduced power for most of the period, and an unexpected shutdown was decided prior to the completion of our beam time.

The first experiments were carried out on D3, supplying polarized neutrons for flipping ratio measurements. The first system that was attempted is shown in the Figure to the right, which is $[\text{Co}_2(\text{H}_2\text{O})(\text{piv})_4(\text{Hpiv})_2(3\text{-pic})_2]$. The crystal



unfortunately broke irreversibly into several pieces around 45 K. It was attempted to collect data on the largest fragment, but that was not sufficiently large and it was abandoned. From this experiment, it is not possible to give any additional information of the nature of the phase transition, but this will be investigated further using in-house X-ray diffraction at Aarhus University in the near future.

Following this, the Ni-analogue was tested but similar problems surfaced and this was also abandoned. Instead, a large crystal of $[\text{Co}_2(\text{H}_2\text{O})(\text{piv})_4(\text{Hpiv})_2(\text{py})_2]$ was mounted and it was possible to cool this to 2 K and currently flipping ratios are being measured following the restart of the reactor. However, this will not be completed before the deadline for the new proposals have expired. Thus, the current report cannot contain any results from the analysis. First results do however indicate significant flipping ratios to be collected.

The final report will be uploaded as soon as a full data set has been collected and analyzed.