Experimental report

14/09/2016

Proposal:	**4-01-1504**	**Council:**	4/2016
Title:	Magnetic interactions in the molecular multiferroic compound (NH4)2[Fe Cl5 (H2O)]		
Research area:	Physics		

This proposal is a resubmission of 4-01-1463

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Samples: (ND4)2[FeCl5Â·(D2O)]

Instrument	Requested days	Allocated days	From	To
IN5	7	3	05/09/2016	08/09/2016

Abstract:

The recent observation of multiferroic behaviour in the compound (NH4)2[FeCl5(H2O)] has stimulated the interest in the study of the mechanisms governing the magnetic interactions in this system. This material orders magnetically at TN= 7.25 K and multiferrocity occurs below ca. 6.9 K with the onset of ferroelectric order. Recently, we have studied by neutron diffraction the magnetic structure of the deuterated compound, (ND4)2[FeCl5·(D2O)]. At low temperature it has been observed a cycloidal magnetic structure propagating in the c-axis and with the magnetic moments lying in the ac plane. In the region between 7.25 K and 6.9 K a sinusoidal spin configuration is likely taking place. Determining experimentally the possible magnetic interactions will help us to precisely understand the origin of the magnetic structure and ultimately the multiferroic behaviour. Also, the possible observation of low energy magnons can allow us to identify those modes which may directly couple to the ferroelectric polarization. Therefore, the purpose of this proposal is to investigate by inelastic neutron scattering magnon spectrum of (ND4)2[FeCl5(D2O)].

Magnetic interactions in the molecular multiferroic compound $(NH_4)_2[FeCl_5(H_2O)]$

$(NH_4)_2[FeCl_5(H_2O)]$ has awakened a significant interest owing to to the recent observation of multiferroic behaviour.[1] This compound orders antiferromagnetically at T_N = 7.25 K and multiferroicity arises below *ca.* 6.9 K, as a consequence -- via the spin-current mechanism -- of an incommensurate cycloidal magnetic structure propagating in the *c*-axis with **k** = (0, 0, 0.23) and with the magnetic moments lying in the *ac* plane (Figure 1 left).[2] In the region between T_N= 7.25 K and the ferroelectric phase at 6.9 K (Figure 1 left), a sinusoidal spin configuration is likely taking place.[2] The magnetic structure at low temperature can be explained by a simple model using four exchange interactions J_1-J_4 (Figure 1 right)[2] The purpose of this experiment was to investigate, by inelastic neutron scattering, these magnetic interactions in order to precisely understand the origin of the magnetic structure and ultimately the multiferroic behaviour. 7 days were requested to obtain full maps of the ground state in two orientations plus an exploration of the temperature evolution, but the subcommittee allocated only 3 days in order to demonstrate the feasibility of the experiment.

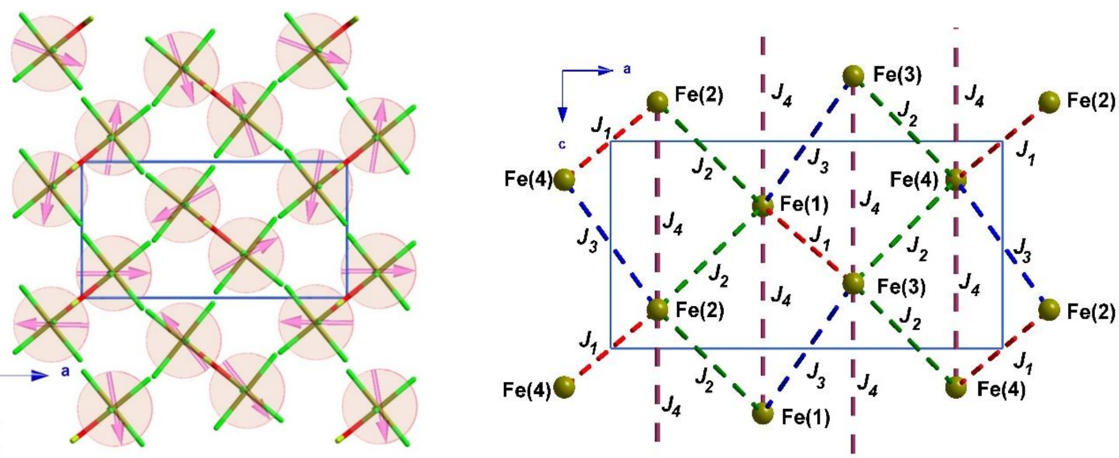

Figure 1: (left) View along the *b*-axis of the superposition of the nuclear and magnetic structures of $(ND_4)_2[FeCl_5{\cdot}(D_2O)]$. (right) View along the *b*-axis of the four magnetic interactions used to model the cycloidal magnetic structure.

A high-quality fully deuterated large single crystal of ca. 8 x 4 x 3 mm was pre-oriented in D9 and mounted with the *a*-axis vertical. The crystal orientation on IN5 was done at λ= 4.8 Å and was followed by an exploratory data collection at low temperature in order to adjust the maximum energy. The rest of the experiment was therefore performed with λ= 6 Å (E_{max} = 1.6 meV) . A full map (in a single orientation, with *a*-axis vertical) was recorded at 1.6K in ~48 h (Figure 2) in steps of Δω = 0.5º. Additionally, a map was recorded at 7 K with a coarser step Δω = 1º in order to compare the cycloidal and the sinusoidal phases.

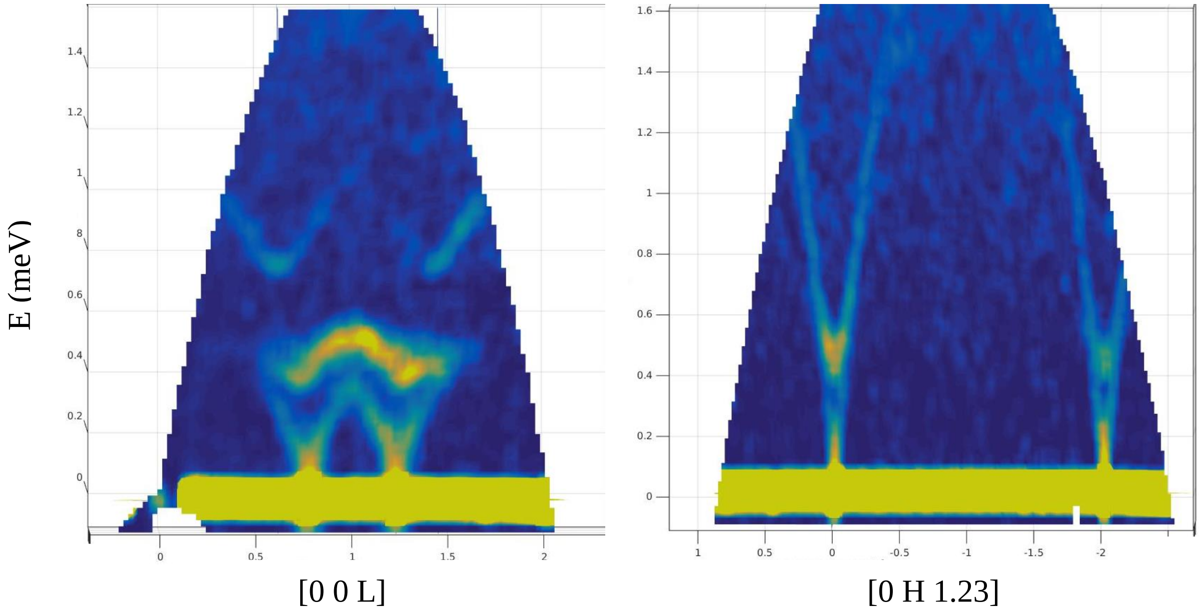

Figure 2: Two cuts of the inelastic intensity map measured at 1.6 K at IN5: views along the [0 0 L]* (left) and [0 H 1.23]* (right) directions

The data are of good quality and three branches are clearly observable (Figure 2), allowing a preliminary analysis consistent with our qualitative hypothesis about the relation between magnetic interactions. However, in order to have a full picture, a second orientation will be necessary, together with a full data collection in the sinusoidal phase.

References

[1] Ackermann M, Brüning D, Lorenz T, Becker P and Bohatý L **2013** New J. Phys. *15* 123001
[2] Rodríguez-Velamazán, J.A.; Fabelo, O.; Millán, A.; Campo, J.; Johnson, R.; Chapon. L. *Scientific Reports* **2015,** 5:14475