# Experimental report

26/10/2020

**Proposal:** 5-41-1019 **Council:** 4/2019

**Title:** Crystal and Magnetic structure determination on the (NH4)2[MoCl5(H2O)] compound

**Research area:** Physics

**This proposal is a new proposal**


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**Samples:** (NH4)2[MoCl5(H2O)]

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| D9 | 15 | 10 | 23/09/2019 | 03/10/2019 |


## Abstract:

The A2[FeX5(H2O)] erythrosiderite-type compounds, where A stands for an alkali metal or ammonium ion and X for a halide ion, have recently awakened interest due to the occurrence of multiferroicity or magnetoelectricity. We have recently investigated by means of single crystal neutron diffraction the influence on the nuclear and magnetic structures of the chemical substitution in the A-site. The influence on the macroscopic behavior has been studied on two of these compounds, (ND4)2[FeCl5(D2O)] and (Cs)2[FeCl5(D2O)]. In the current work, we want to study the nuclear and magnetic structure when the Fe(III) is replaced by Mo(III), with very similar properties but with differnt spin. The crystal structure at RT was solved on the orthorhombic space group Pnma. At low temperature the specific heat shows a clear lambda peak at 7.8 K, which is related with the occurrence of long range magnetic order. Moreover the magnetic structure changes with magnetic fields above 3 T, due to a possible spin-flip transition. In order to elucidate the nuclear and magnetic structure, at zero and under magnetic-field, we kindly apply to 15 days of beam time on D9 single crystal neutron diffractometer.

# EXPERIMENTAL REPORT

EXPERIMENT N° **5-41-1019** INSTRUMENT **D9**

DATES OF EXPERIMENT **23/09/2019 to 03/10/2019**

TITLE

Crystal and Magnetic structure determination on the $(NH_4)_2[MoCl_5 \cdot (H_2O)]$ compound

EXPERIMENTAL TEAM (names and affiliation)

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LOCAL CONTACT **Dr. José RODRÍGUEZ VELAMAZÁN, Dr. Óscar Ramón FABELO ROSA**

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We proposed in this experiment to determine with neutron diffraction techniques the magnetic structure of $(NH_4)_2[MoCl_5 \cdot (H_2O)]$ and $(NH_4)_2[MoBr_5 \cdot (H_2O)]$ compounds in the ground state at low temperatures (T~2 K). The goal was to obtain the magnetic structure under zero magnetic field and under an applied magnetic field above 3 T. In addition, we planned to follow some magnetic reflections with the external applied field.

With this experiment we would have all the needed data to understand the complex magnetic phase diagram of this compound and understand the influence of the Fe(III) substitution by Mo(III).

To do these tasks we received 10 days of beam time in the D9 single crystal diffractometer working at wavelength $\lambda$ = 0.8363 Å. The crystal was mounted onto specific aluminum sample holders that produce a low background and sealed into a closed-cycle cryostat. A vertical cryomagnet was necessary in order to increase the magnetic field up to 6 Tesla. We oriented the single crystal with the **a** crystallographic axis parallel to the vertical magnetic field. The crystal alignment for these measurements was obtained by a previous orientation of the crystal using the neutron Laue diffractometer Orient Express.

In order to determine the crystal and magnetic structure at low temperature and zero magnetic field data collections consisting in a combination of omega- and omega-2θ-scans of each individual reflection were carried out at RT and 2 K for each sample. Additionally, q-scans around the (0 -3 ±q), (-1 0 -1±q), (-2 0 ±q) and (-3 0 ±q) (q = 0.5) reflections were performed for the Cl compound at 2 K.

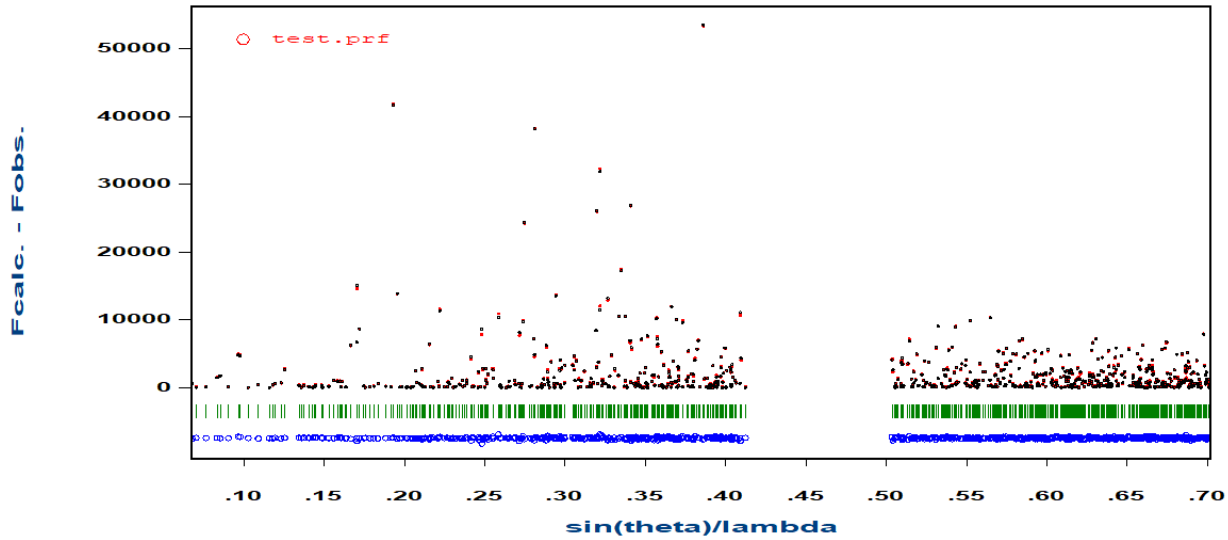
To determine the evolution of the magnetic structure with the applied magnetic field at 2 K and 4 T for the Cl compound data collections consisting in a combination of omega-scans of each individual reflection and q- scans around the (0 -3 ±q), (0 0 1±q), (0 ±q 3) and (±q 0 3) (q = 0.9) reflections where carried out.

Finally, the evolution at 2 K of 6 different reflections ((0 -3 0), (0 0 -3), (0 -2 -1), (2 -2 0), (2 3 1) and (0 1 2)) was followed in the magnetic field range from 0 to 4 T. A complete evolution of 2 reflections ((0 -3 0) and (0 -2 -1)) in the temperature range 2 to 9.5 K and magnetic field range 0 to 4 T was also performed.

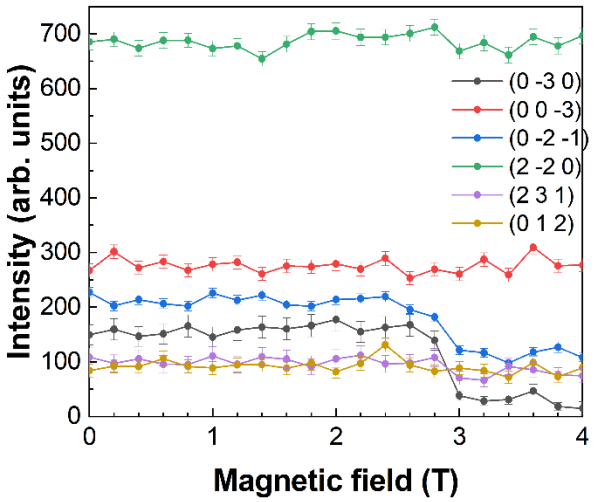
The program RACER45 was used to integrate the omega- and omega-2-scans and to correct them for the Lorentz factor.

A preliminary analysis using the program FullProf Suite suggests that the magnetic structure at zero magnetic field can be described by the space group Pn'm'a' (62.449) (propagation vector k = (0; 0; 0)), in which the Mo magnetic moments align along the a-axis, due to a ferromagnetic interaction in a-axis and an antiferromagnetic interaction in b-axis. In this case the modulus of the magnetic moments at 2 K takes a value of 2.7(9) and 3.1(2) $\mu_B$ for the Cl and Br compounds, respectively.

In the case of the applied magnetic field (B = 4 T), the magnetic structure can be described by the space group Pna'21' (No 33.147) (propagation vector k = (0; 0; 0)). Here the magnetic moments re-orientate from a-axis to b-axis mainly, while the intensity of the (0 0 -3) reflection remains constant: the direction of the magnetic moments changes but is still in the a-b plane.




Single crystal diffraction pattern at 2 K showing the intensities of different nuclear and magnetic reflections. Experimental data (red circles) are fitted using the theoretical model proposed (black circles). $(NH_4)_2[MoBr_5{\cdot}(H_2O)]$ compound.




Evolution at 2 K of 6 different reflections: (0 -3 0), (0 0 -3), (0 -2 -1), (2 -2 0), (2 3 1) and (0 1 2) in the magnetic field range from 0 to 4 T. $(NH_4)_2[MoCl_5{\cdot}(H_2O)]$ compound.