

Experimental report

05/05/2017

Proposal: 5-31-2486

Council: 4/2016

Title: Study of crystallographic and magnetic structures of promising multiferroics: Co₂MgTeO₆, Co₂ZnTeO₆, Mn₂MgTeO₆, Mn₂ZnTeO₆

Research area: Chemistry

This proposal is a new proposal

Main proposer: Maria RETUERTO MILLAN

Experimental team: Christopher R. ANDERSEN
Maria RETUERTO MILLAN

Local contacts: Maria Teresa FERNANDEZ DIAZ

Samples: Co₂MgTeO₆
Mn₂MgTeO₆
Co₂ZnTeO₆
Mn₂ZnTeO₆

Instrument	Requested days	Allocated days	From	To
D2B	3	0		
D20	3	2	06/06/2016	08/06/2016

Abstract:

Co₃TeO₆ has been reported as a multiferroic material with a complex magnetic structure. It has a cryolite-related structure (C2/c) and several positions for the magnetic cations, which expands the degrees of freedom in the magnetic structure offering new possibilities for modifications and manipulations. It has three different magnetic transitions with different magnetic structures when the temperature is lowered below 26K. Also a strong anomaly in the dielectric constant is observed around 18K in zero magnetic field indicating the presence of an spontaneous polarization. Mn₃TeO₆ has a different crystallographic structure but very interesting magnetic properties due to an incommensurate magnetic structure below 23K. We have prepared the novel phases Co₂MgTeO₆, Co₂ZnTeO₆, Mn₂MgTeO₆ and Mn₂ZnTeO₆ and determined their structures by x-ray diffraction. Preliminary neutron diffraction measurements on Co₂MgTeO₆ show the appearance of magnetic peaks at low temperature. Therefore, this shows that, even in the presence of non-magnetic Mg and Te atoms, a long-range magnetic structure is established at low temperatures, for these promising new multiferroic phases.

Co_3TeO_6 has been reported as a multiferroic material with a complex magnetic structure. It has a cryolite-related structure ($C2/c$) and several positions for the magnetic cations, which expands the degrees of freedom in the magnetic structure offering new possibilities for modifications and manipulations. It has three different magnetic transitions with different magnetic structures, when the temperature is lowered below 26K. Also a strong anomaly in the dielectric constant is observed around 18K in zero magnetic field indicating the presence of an spontaneous polarization¹. Mn_3TeO_6 has a different crystal structure but very interesting magnetic properties due to an incommensurate magnetic structure below 23K².

We have prepared the novel phases $\text{Co}_2\text{MgTeO}_6$, $\text{Co}_2\text{ZnTeO}_6$, $\text{Mn}_2\text{MgTeO}_6$ and $\text{Mn}_2\text{ZnTeO}_6$ and determined their structures by x-ray diffraction. Preliminary neutron diffraction measurements on $\text{Co}_2\text{MgTeO}_6$ show the appearance of magnetic peaks at low temperature. Therefore, this shows that, even in the presence of non-magnetic Mg and Te atoms, a long range magnetic ordering is established at low temperatures, for these promising new multiferroic phases. In the D20 experiment, performed 6th to 8th of June, we have measured the thermal evolution of the magnetic structures of all the compounds prepared of the series. The measurements were carried out with a wavelength of $\lambda = 2.41 \text{ \AA}$ and a take-off angle of 90° . 1-2 g of each samples were prepared in V cans. The samples were cooled, while making scans of 3 min. The samples were then measured for at least 2 hours at 2 K and then heated in steps of 2 K with 30 min per step. Finally a high temperature measurement above the magnetic phase transition was made for at least 2 hours. Unexplainable peaks at the same position for all our measurements indicate an impurity present in the instrument at the time we did the experiments. See figure 1, 2 and 3. The evolution of the NPD pattern at decreasing and increasing temperatures of $\text{Co}_2\text{MgTeO}_6$ was made. A magnetic phase transition has been registered at 30 K as seen in figure 4, which has been confirmed by magnetization measurements. The magnetic structure is believed to be a commensurate antiferromagnetic ordering with a propagation vector of $k = (0 \ 0 \ 0.5)$, which has been solved with preliminary neutron diffraction measurements at D2B given in figure 5.

In the case of $\text{Co}_2\text{ZnTeO}_6$ only one peak is present together with some scattering close to the nuclear peaks between 30° and 40° . This peak at 15° indicates long range magnetic ordering, but it has not been possible to solve the magnetic structure with this single peak. In the case of $\text{Mn}_2\text{MgTeO}_6$ and $\text{Mn}_2\text{ZnTeO}_6$, long range magnetic ordering happens below 12 K, which is indicated by intense peaks increasing as temperature is lowered. A magnetic long range ordering has been seen by susceptibility measurements too. The magnetic structure is believed to be an incommensurate magnetic ordering various propagation vectors have been proposed. One among others are the propagation vector $k = (0.005 \ 0.005 \ 0.420)$, which is close to the one of Mn_3TeO_6 , $k = (0 \ 0 \ 0.4302(1))$.

1 H. Singh *et al.* arXiv:1309.6417 (2013); S. A. Ivanov *et al.* Mater. Res. Bull. 47, 63 (2012).

2 S. A. Ivanov *et al.* Mater. Res. Bull. 46, 1870 (2011).

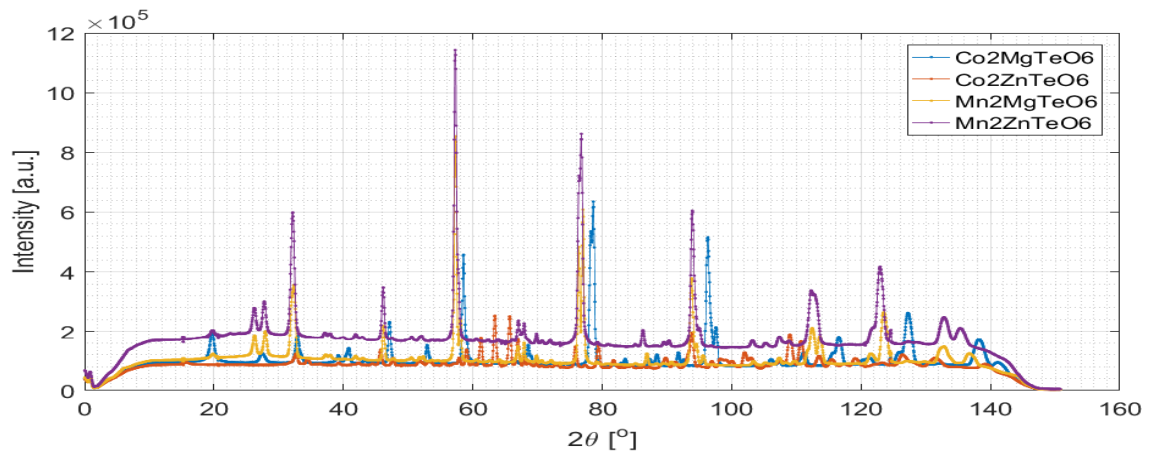


figure 1: Plot of all data. Unexplainable peaks were found for all measurements at all temperatures. This is believed to be an impurity present at the instrument at the time the measurements were made.

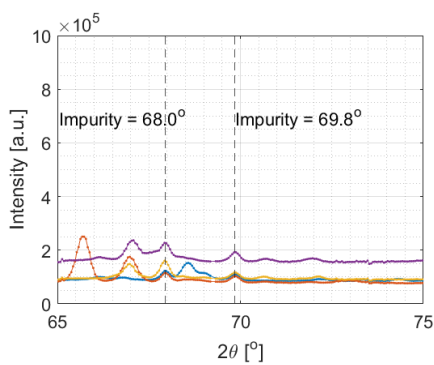


figure 2: Unexplainable peaks detected for all samples.

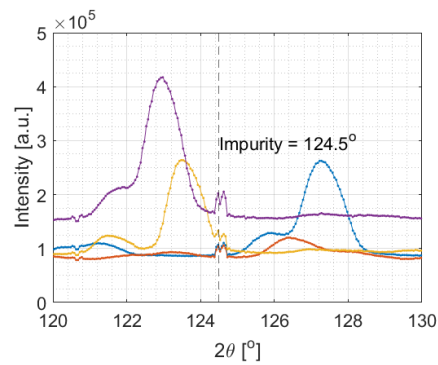


figure 3: Unexplainable peaks detected for all samples

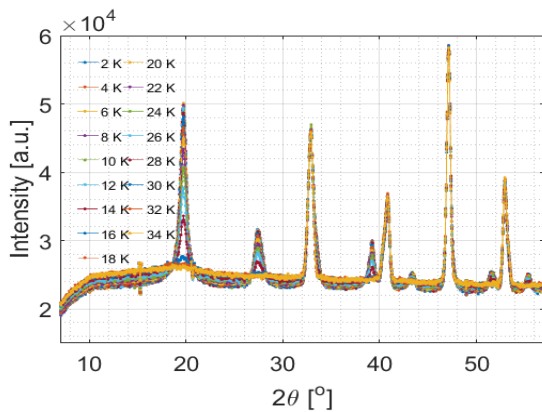


figure 4: NPD of $\text{Co}_2\text{MgTeO}_6$ as the temperature is heated. The intense peaks decrease as the temperature is

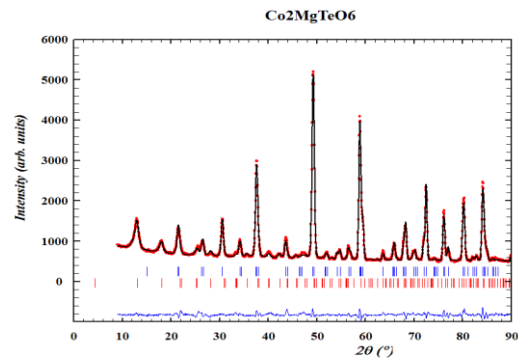


figure 5: Refinement from NPD of $\text{Co}_2\text{MgTeO}_6$ at D2B at 3 K. The propagation vector is $k = (0\ 0\ 0.5)$.