Experimental report

29/07/2024

Proposal: EASY-1194 **Council:** 4/2023

Title: Crystal and magnetic structures of high pressure Mn2-xCoxScSbO6 solid solutions

Research area: Chemistry

This proposal is a new proposal

Main proposer: Kunlang JI

Experimental team:

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Samples: Mn2-xCoxScSbO6

Instrument	Requested days	Allocated days	From	To
D20	12	12	07/09/2023	08/09/2023

Abstract:

Transition metal oxides continue to be of great important for magnetism and other properties, and double perovskite (DPv) and double corundum cation orderings are frequently used to induce and tune physical properties. We propose Mn2-xCoxScSbO6 solid solutions x = 0.5, 1.5 where both have two polymorphs, double perovskite and double corundum. x = 0 and 2 materials are NTO-type, but Mn/Co disorder stabilizes the double perovskite (DPv) structure with x = 0.5 and 1.5. Incorporation of small Co2+ at A-sites in (double) perovskites is very unusual. Rietveld refinements shows that all these solid solutions have the P21/n DPv structure, with phase purities up to ~90%. High temperature Synchrotron X-ray data of x = 1 shows a structural phase transition from DPv to a R3 corundum type structure above 270 ° C. Magnetic measurements confirmed different magnetic properties for DPv and double corundum. D20 is needed to determine the crystal structures in high-resolution mode (wavelength = 1.54 Å), given small monoclinic distortions, as well as low temperature magnetic structures and their thermal evolution in high flux mode (wavelength = 2.41 Å).

Crystal and magnetic structures of the high-pressure $Mn_{2-x}Co_xScSbO_6$ solid solutions

Transition metal oxides continue to be of great important for magnetism and other properties, and double perovskite (DPv) and double corundum cation orderings are frequently used to induce and tune physical properties. Cation ordered $A_2BB'O_6$ phases can adopt the NTO (Ni_3TeO_6)-type structure, with acentric *R*3 symmetry giving interest as polar magnets with potential multiferroic properties, e.g. in $(Co,Ni)_2ScSbO_6$ where interesting lock-in spins orders occur,[1] or as double perovskites (DPv), usually with monoclinic $P2_1/n$ symmetry, of spintronics interest. Recently the use of A= Mn^{2+} has been demonstrated to induce interesting structural features and magnetic properties in $Mn_2BB'O_6$ materials. Mn_2ScSbO_6 has both structures at different pressures (NTO at 5.5 GPa and DPv at 12 GPa).[2] Here we propose to study $Mn_{2-x}CoxScSbO_6$ solid solutions x = 0.5, 1.5 where both have two polymorphs, double perovskite and double corundum. x = 0 and 2 materials are NTO-type at 6GPa, but Mn/Co disorder stabilizes the double perovskite (DPv) structure with x = 0.5 and 1.5. Incorporation of small Co^{2+} at A-sites in (double) perovskites is very unusual. Rietveld refinements show that all these solid solutions have the $P2_1/n$ DPv structure, with phase purities up to ~90%. High temperature Synchrotron X-ray data of x = 1 shows a structural phase transition from DPv to a *R*3 corundum type structure above 270 ° C. Magnetic measurements confirmed different magnetic properties for DPv and double corundum.

In this experiment, around 200mg of the DPv $Mn_{2-x}Co_xScSbO_6$ solid solutions with x = 0.5 and 1.5 were loaded at D20 using V-foil can. Both samples were first heat up to 573 K in a cryofurnace and annealing to room temperature to obtain the NTO phase. D20 High resolution mode with wavelength of 1.54 Å (take-off angle of 42 ˚) was used to confirm NTO phase at 300 K for x = 0.5 and 1.5. Low temperature scans for both samples were collected at 100 K and 1.5 K using high flux mode (wavelength 2.41 Å and take-off angle of 42 ˚) with each counting 2 hour. Thermal scans between 1.5 K and 100 K were collected every 5 K. Neutron diffraction show long range magnetic ordering for both x = 0.5 and 1.5. 300 K Neutron diffraction patterns in Figure 1 confirmed both samples transfer from DPv to NTO phases after annealing at 573 K. Figure 2 shows the magnetic signal below transitions and indicates [00 0] magnetic structures for both samples. Further analysis will be carried out to solve the ground state spins structures and to accomplish data publication.

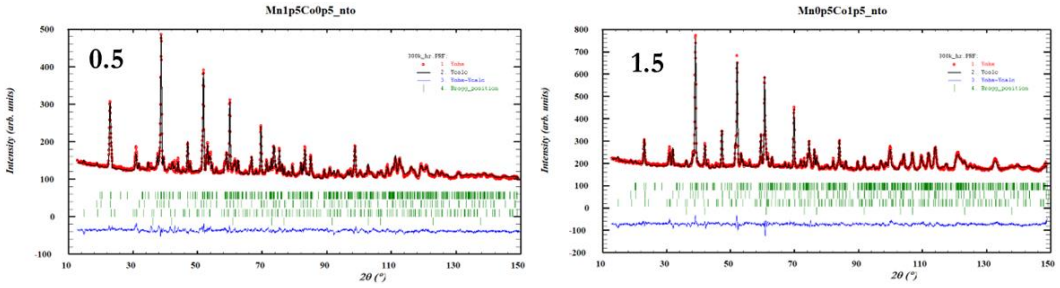

Figure 1. NPD patterns of $Mn_{2-x}Co_xScSbO_6$ (x = 0.5 and 1.5) at 300 K show clearly NTO structure phases with small remaining DPv phases and tiny impurities.

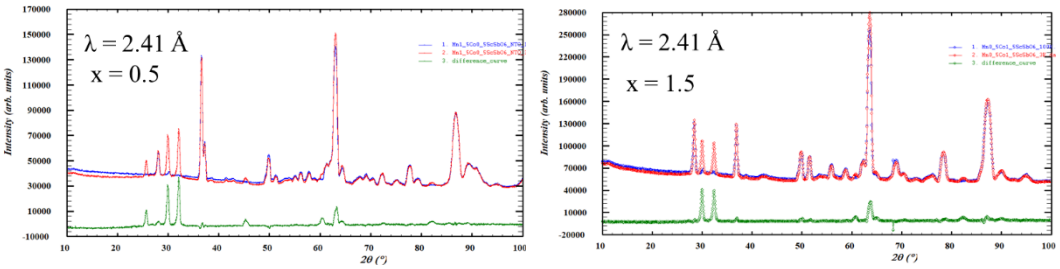

Figure 2. 100 K and 1.5 K NPD data and difference pattens of x = 0.5 and 1.5 sample show clear magnetic peaks at around 30 ˚ and 31 ˚ for NTO phase. The magnetic peak at around 26 ˚ in x = 0.5 is from the remaining DPv phase.

[1] K. Ji et al., Chem. Commun., 2018, 54, 12523.
[2] E. Solana-Madruga et al., Dalton Trans., 2015, 44, 20441.