Experimental Report

Proposal: **7-03-118** **Council:** 4/2012

Title: Oxygen mobility in a new Brownmillerite type oxide Sr2-xBax(Sc,Ga)O5

This proposal is a new proposal

Researh Area: Chemistry

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Samples: Sr2-XBaX(Sc,Ga)O5

Instrument	Req. Days	All. Days	From	To
IN6	6	4	26/10/2012	31/10/2012

Abstract:

Brownmillerite types oxides have an important role in the field of moderate temperature oxygen ion conductors.
Based on our previous studies on brownmillerite framework structures, the prerequisite for low-temperature oxygen ion conduction is a result of the underlying lattice dynamics, revealing a phonon assisted diffusion mechanism, essentially relying on a dynamic oxygen disorder scenario of the infinite MO4 chains.
In this sense the new compound Sr2-xBax(Sc,Ga)O5 can be considered a key compound to probe the influence of order-disorder phenomena for the ((Sc,Ga)O4)? chains on low temperature oxygen mobility properties. In fact, Neutron Powder Diffraction revealed a transition from an order of the ((Sc,Ga)O4)? chain at room temperature to a dynamically disorder state above 300°C.
The aim of the proposed experiment is to investigate the influence of the order/disorder transition on the oxygen mobility with respect to changes in the lattice dynamics by studying the changes of the DOS of the title compound as a function of temperature on IN6

Scientific background and aim of the experiment

The rapid growth in global energy demand in the face of dwindling resources has stimulated increasing efforts towards developing energy storage devices. Solid oxide fuel cell (SOFC) technology, for example, combines highly efficient conversion of energy stored in fossil and non-fossil fuels into electrical energy. As SOFC applications are limited by the high operating temperatures[1-3], above 800°C, intense efforts are directed at developing new SOFC electrolyte materials with higher oxide ion conductivities. The Brownmillerite structure ($ABO_{2.5}$ i.e. $A_2B_2O_5$) is a remarkable structural family relevant to this field: some of its members can undergo reversible topotactic electron-ion transfer reaction already at room temperature[4,5]. Following this concept we synthesized a new Brownmillerite phase $Sr_{2-x}Ba_xScGaO_5$, containing $3d^0$ transition-metals to serve as pure oxide ion conductors.
Previous Neutron powder diffraction (D2B@ILL) and Raman spectroscopy study show that $Sr_{2-x}Ba_xScGaO_5$ undergoes an order (*I2mb*) – disorder (*Imma*) phase transiton, approximately at 300°C.
Previous studies[6] have shown that oxygen mobility in the Brownmillerite frameworks is linked to the dynamical disorder of the tetrahedric chains. For this reason, we want to investigate the influence of the *I2mb*/*Imma* transition on the oxygen mobility with respect to the changes in the lattice dynamics. In order to follow the evolution of the phonon generalized density of states (gDOS) of $Sr_{2-x}Ba_xScGaO_5$, as a function of temperature, we performed inelastic neutron scattering (INS) experiments at IN6.

Experimental results

INS has been performed on $Sr_{2-x}Ba_xScGaO_5$ (with x=0, 0.1) to investigate the change of the lattice dynamics in these two structures. Data were collected from RT up to 700°C. Figure 1a and 1b show the experimental gDOS obtained at different temperatures of Sr_2ScGaO_5 and $Sr_{1.9}Ba_{0.1}ScGaO_5$

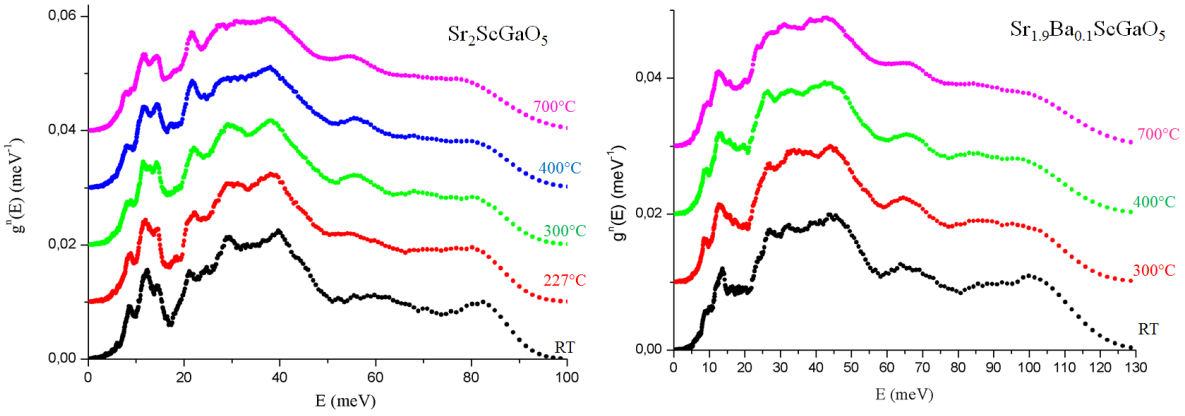

Figure 1- Phonon density of states as function of the temperature (RT-750°C) of Sr_2ScGaO_5 and $Sr_{1.9}Ba_{0.1}ScGaO_5$

The most significant contribution of the spectrum consists in the phonon mode about 9meV (see Figure 2 for an enlarged view) since the low energy phonon mode was found to be linked to the oxygen mobility[6].

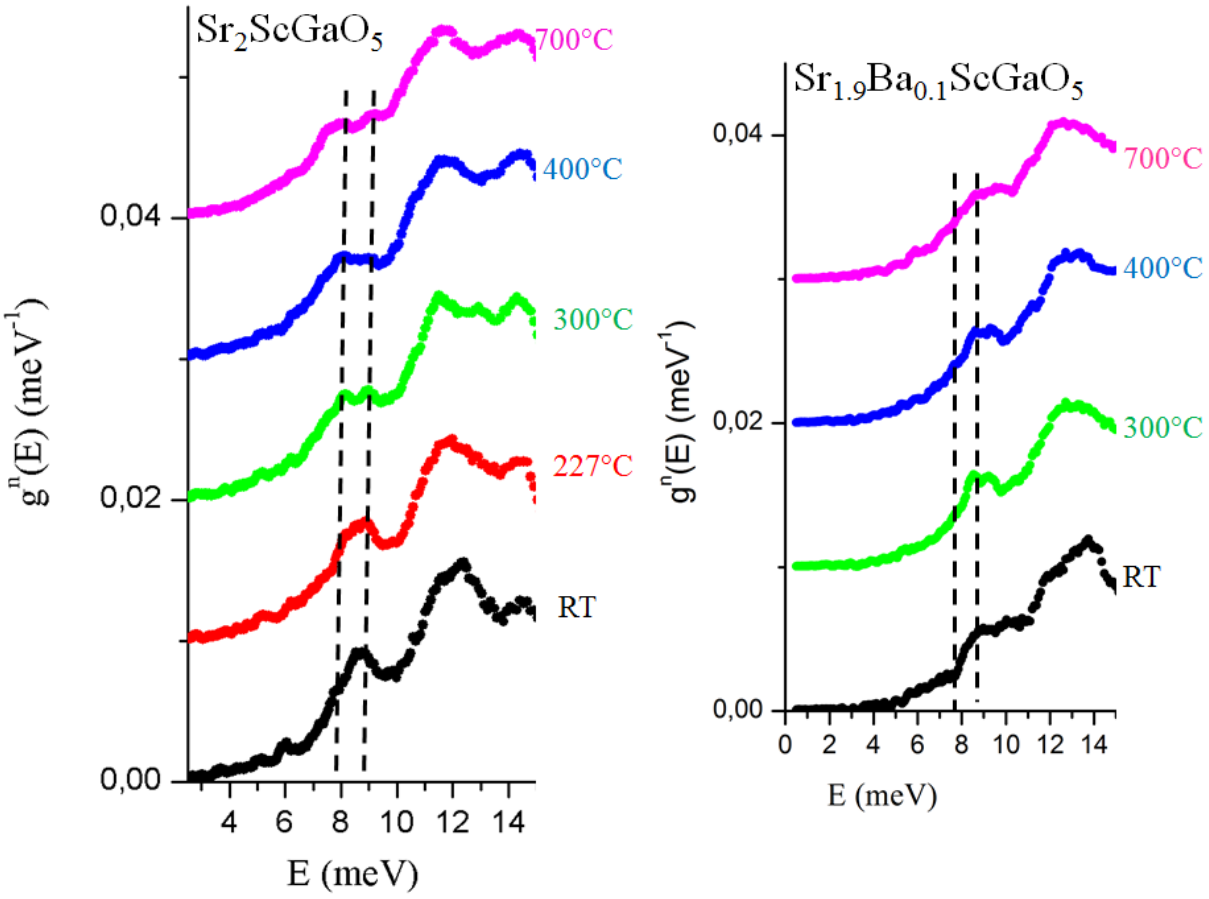

Figure 2- The low-energy region of the phonon density of states of Sr_2ScGaO_5 and $Sr_{1.9}Ba_{0.1}ScGaO_5$

In Sr_2ScGaO_5 the mode at 9 meV splits in two different modes and shifts to 8 meV at 300°C. This shift could be linked to an increase of the dynamical disorder of the GaO_4 chains in the structure. Instead, the doping with Ba does not affect the lattice dynamics as the low frequency mode at 9 meV looks to be temperature-independent in this case.

Reference

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