Experimental report

16/12/2022

Proposal: **CRG-2904** **Council:** 4/2021

Title: Crystal structure determination ofpotential superconducting perovskite-related oxides

Research area:

This proposal is a new proposal

Main proposer: **Regino SAEZ PUCHE**

Experimental team:

Local contacts: Ines PUENTE ORENCH

Samples: R2-xSrxNiO4 R2-xSrxNiO3

Instrument	Requested days	Allocated days	From	To
D1B	2	1	24/08/2021	25/08/2021

Abstract:

Crystal structure determination of potential superconducting perovskite-related oxides.

Over the last decades, perovskite – related oxides have been extensively studied in the search of new functional materials. Amongst several targeted properties, superconductivity has traditionally attracted much attention and fundamental research and has recently found a renewed perspective to study new potential superconducting materials with infinite layers of $Ni^{1.2+}$ cations in square-planar coordination via topotactic reduction of perovskite oxides. [1,2] A similar approach, using topotactic reduction with CaH_2 was used to produce infinite layers and chains of FeO_4 square planes from both perovskite and Ruddlesden-Popper phases. [3,4]

We recently prepared two new series of compounds with $R_{0.8}Sr_{0.2}NiO_3$ and $R_{0.8}Sr_{1.2}NiO_3$ compositions and perovskite-related structures. The first series is prepared under high pressure and high temperature conditions while the second one is obtained via topotactic reduction of their respective $R_{0.8}Sr_{1.2}NiO_4$ Ruddlesden-Popper phases. $R_{0.8}Sr_{0.2}NiO_3$ are promising precursors for reduced $R_{0.8}Sr_{0.2}NiO_2$ oxides, which along $R_{0.8}Sr_{1.2}NiO_3$ compounds are notable as potential superconducting materials due to the presence of infinite layers and chains, respectively, of $Ni^{1.2+}O_4$ square planes.

During experiment CRG-2904, we focused on the room pressure samples with Ruddlesden-Popper structure. Unfortunately, no high-pressure samples with perovskite structure were available with enough purity other than R = Sm (highly absorbing neutron radiation), while the topotactic reductions were not possible due to experimental difficulties with the vacuum system.

Among the Ruddlesden-Popper related compounds, $R_{0.8}Sr_{1.2}NiO_4$ samples with R = Nd and Pr were measured in D1B, using $\lambda = 1.28$ Å for high resolution structural determination and $\lambda = 2.52$ Å for high intensity diffraction in low angle region to detect any long-range magnetic order. Long scans were collected at room temperature, which confirmed *I4/mmm* symmetry observed from laboratory XRD. Preliminary refinements (Fig. 1) confirm disordered R/Sr in a single crystallographic position.

Short scans were collected at intermediate temperatures in ramp mode down to 1.5 K and low temperature long scans were measured for magnetic structure characterisation. No evidence of long-range magnetic interactions could however be detected, which is explained in terms of: i) the magnetic dilution of the rare earth in a disordered site with diamagnetic Sr^{2+} cations; ii) the small magnetic moment of the available R^{3+} cations (with $4f^3$ and $4f^4$ electron configurations); and iii) the low dimensionality of Ni^{2+} ($3d^8$) perovskite layers. These compounds thus show frustrated interactions justifying the absence of magnetic peaks in low temperature data, other than minor amounts of secondary NiO (3.6(2) w.t.%) in the Nd sample.

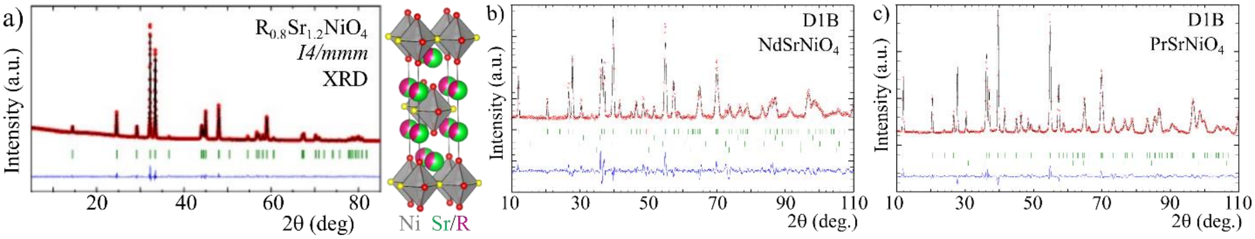

Fig. 1. Rietveld fits of the *I*4/*mmm* Ruddlesden-Popper structure against XRD (a) and NPD (b and c) data. The refined structure is shown in the right panel of figure a). Brag ticks in figures b) and c) show the main phase, secondary NiO (nuclear and magnetic) in b) and V in both b) and c).

References:

[1] D. Li, K. Lee, B. Y. Wang, M. Osada, S. Crossley, H. R. Lee, Y. Cui, Y. Hikita, H. Y. Hwang. *Nature* 2019, **572**, 624-627.

[2] M. Osada, B. Y. Wang, B. H. Goodge, K. Lee, H. Yoon, K. Sakuma, D. Li, M. Miura, L. F. Kourkoutis, H. Y. Hwang. *Nano Lett.* 2020, **20**, 5735−5740.

[3] Y. Tsujimoto, C. Tassel, N. Hayashi, T. Watanabe, H. Kageyama, K. Yoshimura, M. Takano, M. Ceretti, C. Ritter, W. Paulus. *Nature* 2007, **450**, 1062–1065

[4] Cédric Tassel, L. Seinberg, N. Hayashi, S. Ganesanpotti, Y. Ajiro, Y. Kobayashi, H. Kageyama. *Inorg. Chem.* 2013, **52**, 6096−6102.