(※本報告書は英語で記述してください。ただし、産業利用課題として採択されている方は日本語で記述していただいても結構です。)

MLF Experimental Report	提出日 23.06.2015
課題番号 2014B0349 実験課題名 Oxygen induced soft-mode behaviour in Pr2NiO4+δ promoting phonon assisted diffusion 実験責任者名 Matthias Frontzek 所属 Paul Scherrer Institut, Laboratory for Neutron Scattering and Imaging	装置責任者 Kenji NAKAJIM A 装置名 AMATERAS / BL 14 実施日 20.-24.03.2015

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

$Pr_2NiO_{4.12}$ and $Pr_2NiO_{4.25}$ single crystals

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。)
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

The non-stochiometric oxide $Pr_2NiO_{4+\delta}$ (δ = 0 – 0.25) exhibits for δ=0.25 a long-range ordered superstructure due to order of the intercalated oxygen at room temperature. For δ=0.12 a long-range ordered, but different structure, can be observed at lower temperature (~100K), prompting the existence of a dynamic state of the intercalated oxygen at room temperature. Since these compounds are excellent conductors of oxygen with foreseeable applications in energy related research (e. g. solid oxide fuel cells), the driving mechanism of the diffusion is of utmost importance. The observation of long-range ordered superstructures lead to the postulation of a potential new mechanism of oxygen diffusion, where dynamic movement of the NiO_6 octahedron lead to a diffusion pathway of interstitial oxygen to apical oxygen. This dynamic movement should be observable as a low energy optical phonon mode.

Aim of the proposed experiment was to find evidence for the proposed phonon assisted diffusion. For this purpose we compared the inelastic excitation spectra of $Pr_2NiO_{4.12}$ and $Pr_2NiO_{4.25}$ single crystals measured in HHL scattering geometry on AMATERAS. We used the close-cycle cryostat to also be able to compare the excitation spectra of the $Pr_2NiO_{4.12}$ compound in the ordered and non-ordered state, The incident energy was

2. 実験方法及び結果（つづき） Experimental method and results (continued)

chosen to be 15.1meV which yielded additional incident energies of 7.7 meV, 4.6 meV and 3.1 mev thanks to the frame multiplication at AMATERAS. In our preliminary data evaluation, we were able to identify a weak inelastic mode in the $Pr_2NiO_{4.12}$ sample which is centered around HH = 0.5 and 1.5 r. l. u. which seems to be only weakly dispersive (see Fig. 1). The characteristics of this mode agree well with the expectations for the optical phonon connected to the oxygen diffusion, although a full ab-inito theoretical model calculation is yet missing. The next steps will be the analysis of the spectra gained at the additional incident energies and the aforementioned theoretical simulations.

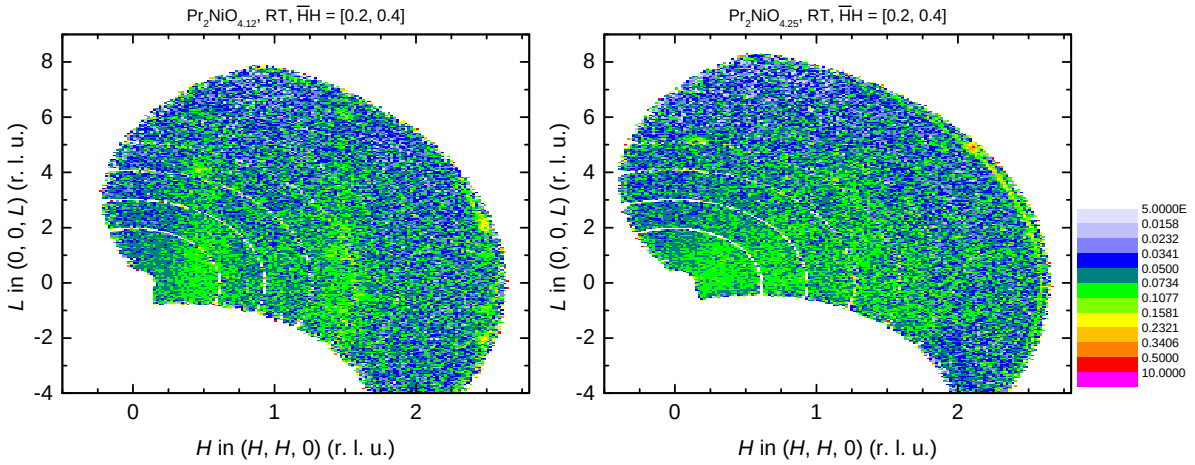

Figure 1: Constant energy maps of $Pr_2NiO_{4.12}$ (left) and $Pr_2NiO_{4.25}$ (right) at a position in −HH which is away from the strong phonons of the parent compound. The intensity scale is the same, although the $Pr_2NiO_{4.25}$ crystal was two times larger. In the left picture, diffuse inelastic scattering is centered around HH = 0.5 and 1.5 r. l. u., while in the right picture the intensity seems to be more evenly distributed.

In summary, we searched and found evidence for a low energy optical phonon mode in $Pr_2NiO_{4+\delta}$ connected to the oxygen transport properties in these compounds.