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 MLF Experimental Report	提出日 Date of Report March 11, 2013
課題番号 Project No. 2012B0182 実験課題名 Title of experiment Investigation of the charge ice state by the PDF analysis 実験責任者名 Name of principal investigator Noriaki Hanasaki 所属 Affiliation Osaka University	装置責任者 Name of responsible person Dr. K.Ikeda and Dr. Ohtomo 装置名 Name of Instrument/(BL No.) BL-21 NOVA 実施日 Date of Experiment From Dec.23, 2012 to Dec.25, 2012

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.
Pyrochlore Niobates YCaNb_2O_7 Powder Pyrochlore Niobates $\text{NdCaNb}_2\text{O}_7$ Powder Pyrochlore Niobates $\text{Y}_2\text{Nb}_2\text{O}_7$ Powder Pyrochlore Niobates $\text{Pb}_{1.5}\text{Nb}_2\text{O}_{6.5}$ Powder Pyrochlore Niobates $\text{Cd}_2\text{Nb}_2\text{O}_7$ Powder

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.
<p>The sample powder (the pyrochlore niobates) and the helium gas were put into the vanadium tube, through which the neutron beam can go. This vanadium tube was set in the bottom of the cryostat. In BL-21(NOVA), the sample powder was irradiated by the pulsed neutron beam during the several hours. The neutron diffraction was observed by use of the detector set in the 90 (degree) bank. In YCaNb_2O_7, the diffraction was obtained at 50K, 300K, and 450K. In $\text{NdCaNb}_2\text{O}_7$, the diffraction was measured at 300K and 450K. In the reference sample of $\text{Y}_2\text{Nb}_2\text{O}_7$, $\text{Cd}_2\text{Nb}_2\text{O}_7$, and $\text{Pb}_{1.5}\text{Nb}_2\text{O}_{6.5}$, the diffraction was measured at room temperature. The neutron-diffraction intensity is obtained as a function of the wave number Q. By the Fourier transformation of these data, we get the neutron PDF (pair distribution function). Since the neutron absorption of Cd is very strong in $\text{Cd}_2\text{Nb}_2\text{O}_7$, the diffraction intensity is too weak to analyze the PDF.</p> <p>In the neutron PDF, one can see the peak around $r = 2(\text{Å})$, $r = 2.6(\text{Å})$ and $r = 3.7(\text{Å})$ in all the samples except $\text{Cd}_2\text{Nb}_2\text{O}_7$. The peak around $r = 2(\text{Å})$ is ascribed to the correlation between the niobium atom and the nearest</p>

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

neighbor oxygen atoms, which is consistent with the single crystal analysis by the X-ray diffraction, indicating the reliability of these PDF data.

We investigate the temperature dependence of the PDF data in YCaNb_2O_7 and $\text{NdCaNb}_2\text{O}_7$. As the temperature decreases, the peak becomes sharp. Besides this, the striking change of the peak shape cannot be seen. Since the diffuse scattering, that originates from the Nb displacement, is observed below 500°C in the X-ray diffraction, we should compare the PDF data measured below and above 500°C . However, we cannot measure neutron diffraction above 500°C , since there is a possibility that the cryostat, which is used the sample holder, can be damaged under the high temperatures. The PDF measurement above 500°C will be necessary and left for the further study.

We examine the peak around $r = 2(\text{Å})$ in YCaNb_2O_7 , $\text{NdCaNb}_2\text{O}_7$, $\text{Y}_2\text{Nb}_2\text{O}_7$, and $\text{Pb}_{1.5}\text{Nb}_2\text{O}_{6.5}$. There is a tendency about the peak position (r) around $2(\text{Å})$ that $r(\text{Pb}_{1.5}\text{Nb}_2\text{O}_{6.5}) < r(\text{YCaNb}_2\text{O}_7 \text{ and } \text{NdCaNb}_2\text{O}_7) < r(\text{Y}_2\text{Nb}_2\text{O}_7)$. This suggests that the distance between the niobium ion and the nearest-neighbor oxygen ion decreases, as the valence of the niobium ion increases. The striking splitting or the broadening of the peak around $r = 2(\text{Å})$, which are suggestive of the large charge disproportionation in the Nb site, are not observed.

In order to examine the possibility of the two-in or two-out Nb configuration and two-in-two-out Nb configuration, we try to simulate the neutron PDF on the assumption of these models. The conclusion has not been drawn yet.

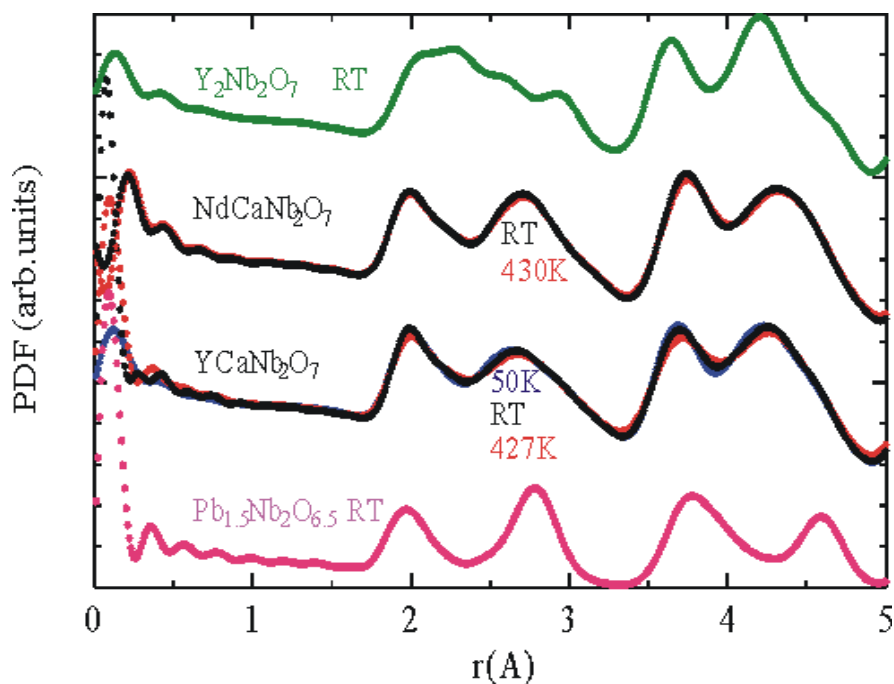


Fig. Neutron PDF of the pyrochlore niobates measured in BL-21 (NOVA).