MLF Experimental Report	提出日 Date of Report 2014 September 16
課題番号 Project No. 2012B0024 実験課題名 Title of experiment Isotopic Contrast Experiment for the Local Structural Investigations of Quasicrystal $Ti_{45}Zr_{38}Ni_{17}$ 実験責任者名 Name of principal investigator Toyoto Sato 所属 Affiliation Tohoku University	装置責任者 Name of responsible person Toshiya Otomo 装置名 Name of Instrument/(BL No.) NOVA BL-21 実施日 Date of Experiment 2012 December 15 – 16 2012 December 18

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.

Quasicrystal Titanium Zirconium Nickel, $Ti_{45}Zr_{38}Ni_{17}$, bulk

Quasicrystal Titanium Zirconium Nickel, $Ti_{45}Zr_{38}{}^{58}Ni_{17}$, bulk

Quasicrystal Titanium Zirconium Nickel, $Ti_{45}Zr_{38}{}^{60}Ni_{17}$, bulk

Approximant Titanium Zirconium Nickel, $Ti_{45}Zr_{38}Ni_{17}$, bulk

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.

Crystallographically, quasicrystals are interesting materials because of 5, 8, 10 and 12 fold symmetries and lack of translational order, both of which are incompatible with structural rules of crystalline materials [W. Steure, Z. Kristallogr. **219** (2004) 391]. For the reasons, they are considered to take a unique position in between crystal and amorphous materials.

In particular, some quasicrystals absorb and desorb hydrogen atoms substantially.

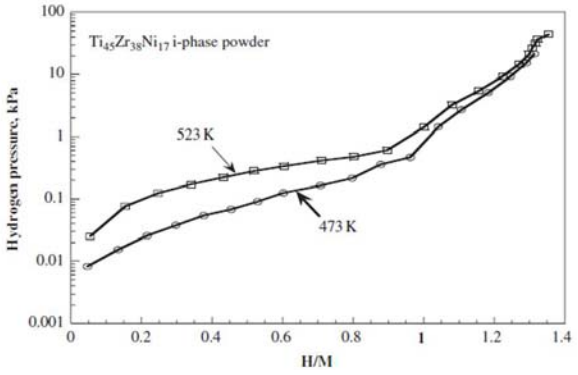

Fig. 1 Hydrogen absorption pressure–composition isotherm for $Ti_{45}Zr_{38}Ni_{17}$ at temperature of 473 and 523 K

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

For example, a hydrogen gravimetric density reaches up to 2.8 wt.% for quasicrystal of Ti–Zr–Ni system (Fig. 1) [A. Takasaki, et al., Mater. Res. Soc. Symp. Proc. **805**, 351 (2004).], which is better than a practical crystalline hydrogen storage materials (e.g $LaNi_5$ (1.4 wt.%)).

In order to improve the hydrogen storage properties and understand the storage mechanism, determination of atomic sites is required. However, in contrast to crystalline materials, it is more difficult to uniquely determine an atomic structure in the absence of a space group.

Since qusicrystals do not follow crystallographic symmetry rules, general method like Rietveld refinement cannot be employed. Then, the pair distribution function (PDF) analysis performed by a total neutron scattering experiment up to high Q region can be a suitable method because it directly provided atomic pair distributions [T. Egami, S.J. Billinge, Undernearth the Bragg Peaks Structural Analysis of Complex Materials, Pargamon: Amsterdam, 2003.].

In the total neutron scattering, the amplitudes are governed by neutron scattering lengths *b*. This is that different amplitudes are obtained on samples with the same structures and compositions if they were substituted by isotope elements with different *b*. This is very distinctive neutron scattering experiment (so–called isotopic contrast) and often employed for more accurate evaluation of $g_{ij}(r)$ [E. Sváb et al., J. Non–Cryst. Solids **104** (1988) 291.].

Therefore, we had total neutron scattering experiments on quasicrystals $Ti_{45}Zr_{38}Ni_{17}$ with natural Ni (*b* = 10.3 fm), ^{58}Ni (*b* = 14.4 fm) and and ^{60}Ni (*b* = 2.8 fm) and the approximant $Ti_{45}Zr_{38}Ni_{17}$ with natural Ni (*b* = 10.3 fm), which adopt a crystal structure with similar local structures to quasicrystals $Ti_{45}Zr_{38}Ni_{17}$. Approximately 1.0 – 2.0 g of samples were placed in a cylindrical null–scattering $V_{96}Ni_4$ alloy sample container with outside diameter of 6.00 mm and thickness of 0.10 mm.quartz container with an inside diameter of 6 mm for the neutron diffraction experiments.

Figure 2 shows S(Q)s and G(r)s for the quasicrystal and the approximant $Ti_{45}Zr_{38}Ni_{17}$. Although they show different S(Q), they show similar G(r). This indicates that they adopt the same short range atomic arrangements. Presently, the crystal structure of the approximant $Ti_{45}Zr_{38}Ni_{17}$ is being determined in order to obtain local atomic arrangements for the quasicrystal.

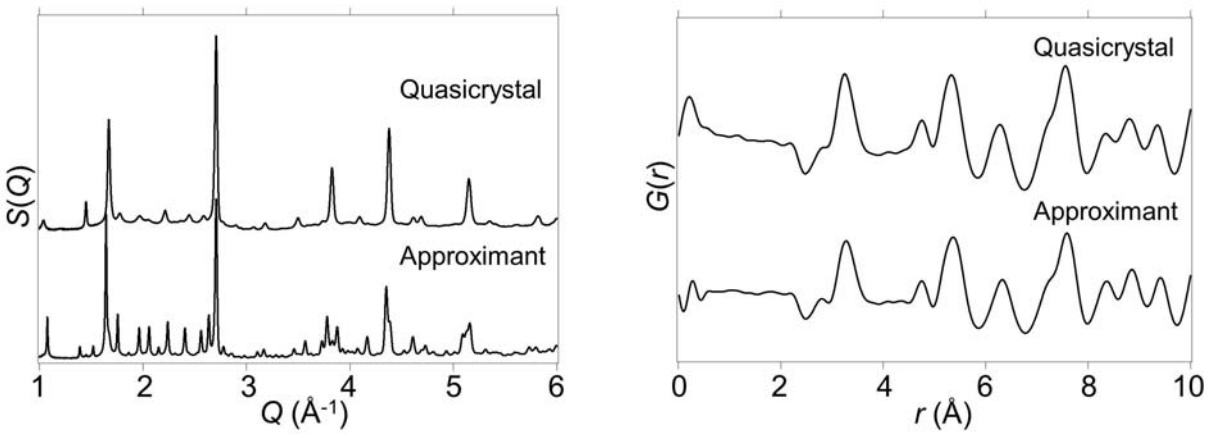

Fig. 2 (left) S(Q)s and (right) G(r)s for the quasicrystal and the approximant $Ti_{45}Zr_{38}Ni_{17}$