MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2016B0239 実験課題名 Title of experiment Magnetic Structure of Ru_2O_9 dimer in $Ba_3Co_{1-x}M_xRu_2O_9$ (M=Zn, Ca) 実験責任者名 Name of principal investigator Yasui, Yukio 所属 Affiliation Meiji University	装置責任者 Name of responsible person Ishigaki, Toru 装置名 Name of Instrument/（BL No.） iMATERIA (BL 20) 実施日 Date of Experiment 2017/2/1 ～ 2017/2/5 2017/3/21

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.

$Ba_3Co_{1-x}Ca_xRu_2O_9$ (x=0, 0.1, 0.2, 0.3)

$LaAl_{1-x}Zn_xO3$ (x=0, 0.01, 0.02)

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.

We investigated the crystal structure and the magnetic structure of $Ba_3Co_{1-x}Ca_xRu_2O_9$ (x=0, 0.1, 0.2 and 0.3) from 3.6K to 150 K particularly focusing on how the magnetic moment on Ru^{5+} begins to fluctuate with x. The compounds with the general formula $Ba_3MRu_2O_9$ (M=Co, Ca, Zn) have a hexagonal barium titanate structure type, where the structure consists of units of two face-shared RuO_6 octahedra (Ru_2O_9 dimers) interconnected by perovskite-type corner sharing with MO_6 octahedra. $Ba_3CoRu_2O_9$ exhibits an antiferromagnetic transition at T_N=98K. The magnetic structure of $Ba_3CoRu_2O_9$ is already reported by Ref. 1, where the Co^{2+} and Ru^{5+} spins in the ordered state have the ordered moments of 2.7 μ_B and 1.5 μ_B, respectively. Recently, we have discovered a novel type of quantum spin liquid in $Ba_3ZnRu_2O_9$, where any traces of the Curie tail or glassy behavior of magnetic susceptibility have not been detected down to 50 mK [2]. We have controlled the magnetic ground state from an antiferromagnetic order to the gapless spin liquid state by making a solid solution between Zn^{2+} and Co^{2+}. The magnetic behavior of $Ba_3CaRu_2O_9$ is also investigated already by Ref. 3, and the spin system is in the nonmagnetic singlet state with the Ru^{5+} dimers.

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

We have also controlled the magnetic ground state from an antiferromagnetic order to nonmagnetic state by making a solid solution system $Ba_3Co_{1-x}Ca_xRu_2O_9$. With increasing the Ca-concentration, the antiferromagnetic transition temperature T_N decreases such as T_N=88K (x=0.1), 82K (x=0.2), and 76K (x=0.3), respectively. We investigated the crystal structure and the magnetic structure of $Ba_3Co_{1-x}Ca_xRu_2O_9$ (x=0, 0.1, 0.2 and 0.3) by the neutron powder diffraction measurements using the iMATERIA at MLF.

Figure 1 shows examples of neutron diffraction profiles of $Ba_3CoRu_2O_9$ taken at various temperatures. We can see the growth of intensities of the magnetic reflections, which is indicated by arrows. As a result of preliminary magnetic structure analysis, the obtained results are almost similar to that of the repoted structure by ref. 1, which is collinear magnetic structure of both Co^{2+} and Ru^{5+} spins along *b*-crystal axis. However, we observed 010 magnetic reflection, which can not be explained by the repoted structure. Then, the magnetic structure of $Ba_3CoRu_2O_9$ seems to be canted structure from the reported collinear one. We are going to continue analyzing neutron diffraction data.

Figure 2 shows the neutron diffraction profiles of $Ba_3Co_{1-x}Ca_xRu_2O_9$ with x=0, 0.1, 0.2, and 0.3 taken at *T*=3.6K. In Fig.2, arrows show the magnetic reflections, and we can see the sharp magnetic peak even in x=0.3. The intensities of several magnetic peaks decrease with increasing x. On the other hand, the intensities of remaining peaks do not change by Ca-doping. Then, the change of the magnetic structure by Ca-substitution for Co is not simple. We are going to carry out the magnetic structure analysis. Moreover, the results of the crystal structure analysis of obtained data give us the important information to understand the origin of different magnetic ground states between $Ba_3CoRu_2O_9$ and $Ba_3CaRu_2O_9$.

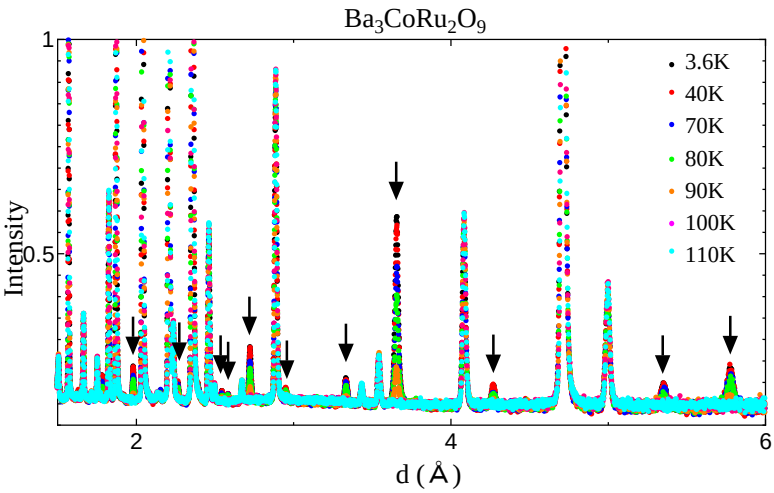

Fig. 1: Profiles of the neutron diffraction of $Ba_3CoRu_2O_9$ taken at various temperatures. Arrows indicate the magnetic reflections.

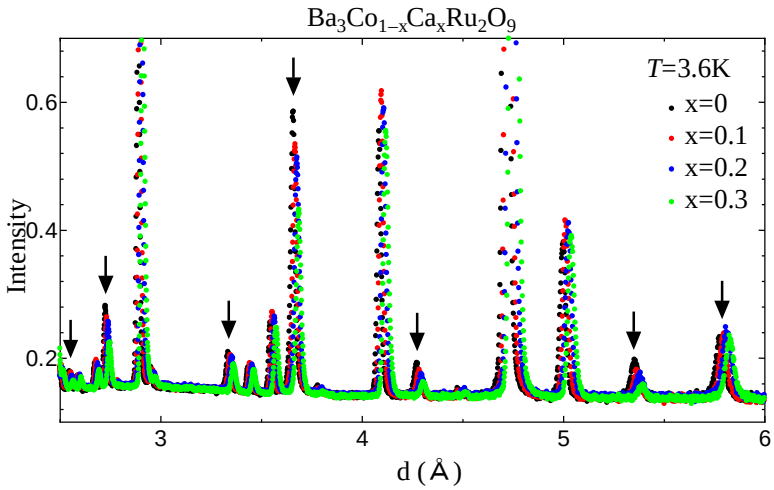

Fig. 2: Profiles of the neutron diffraction of $Ba_3Co_{1-x}Ca_xRu_2O_9$ with x=0, 0.1, 0.2, and 0.3 samples taken at *T*=3.6K. Arrows indicate the magnetic reflections..

[1] Lightfoot and D. Battle: J. Solid State Chem.**89** (1990) 174.

[2] I. Terasaki *et al.*: J. Phys. Soc. Jpn. 86 (2017) 033702.

[3] M. Senn *et al.*: J. Phys.: Condens Matter **25** (2013) 496008.