実験報告書様式（一般利用課題・成果公開利用）

（ ※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。）

CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2013/04/04 承認者 Approver Ryoichi Kajimoto 提出日 Date of Report 2013/04/03
課題番号 Project No. 2012B0116 実験課題名 Title of experiment Coupled Mode between Rattling Atomic Motions and Crystal-Field Excitations in $Pr(Os_{1-x}Ru_x)_4Sb_{12}$ 実験責任者名 Name of principal investigator Kazuaki Iwasa 所属 Affiliation Department of Physics, Tohoku University	装置責任者 Name of Instrument scientist Ryoichi Kajimoto 装置名 Name of Instrument/（BL No.） 4SEASONS (BL01) 実施日 Date of Experiment 2013.3.2 – 3.6 2013.3.19 – 2.23

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.

Single crystal samples of $Pr(Os_{0.32}Ru_{0.68})_4Sb_{12}$ and $PrRu_4Sb_{12}$.

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 aim of present experiment is to investigate the dynamical Jahn-Teller effect due to coupling between the rattling soft phonon mode and the 4*f*-electron state of the Pr ions in $Pr(Os_{1-x}Ru_x)_4Sb_{12}$. The rattling motion of filling Pr ions in the Sb cage lattice in the both end materials $PrOs_4Sb_{12}$ and $PrRu_4Sb_{12}$ appears at approximately 4 meV. Previously, the low-energy phonon dispersions in $PrOs_4Sb_{12}$ and $PrRu_4Sb_{12}$ were measured using triple-axis spectrometers. The Pr-ion rattling motions detected at approximately 4 meV exhibits anharmonic properties, which is a signature of the large amplitude motion of Pr atoms. The crystal-field splitting energies between the singlet ground state to the first triplet excited state are 0.7 and 5.4 meV for $PrOs_4Sb_{12}$ and $PrRu_4Sb_{12}$, respectively. The second excited state of another triplet appears at approximately 11 meV in both of these materials. By the mixture of the Os and Ru atoms, the crystal-field splitting energy can be tuned to the same range as the rattling motion (R. Miyazaki et al.: J. Phys.: Conf. Ser. 200 (2010) 012125). Therefore, a hybridization effect between rattling phonon and crystal-field excitations is to be enhanced in this mixture system.

The sample $Pr(Os_{0.32}Ru_{0.68})_4Sb_{12}$ is chosen for the present inelastic spectroscopy because the bare crystal-field and rattling modes converge with each other. By following previous results using of inelastic neutron scattering, we tried to observe the hybridized mode in $Pr(Os_{0.32}Ru_{0.68})_4Sb_{12}$.

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

We measured excitation spectra up to 20 meV from the co-aligned single-crystal samples of $Pr(Os_{0.32}Ru_{0.68})_4Sb_{12}$, which our collaborators synthesized recently, at two temperature points (15 and 200 K) on 4SEASONS (BL01). We also measured $PrRu_4Sb_{12}$ as a reference. The energy resolution (FWHM) of 2 meV with E_i = 30 meV. We tried to measure dispersion relations or momentum-dependence of scattering intensity in order to identify cross sections dominated by phonon or by crystal-field transitions. The spectrometer 4SEASONS (BL-01) has offered measurements by rotating single crystal samples, which is expected to be suitable for the aim of the present proposal.

Figure 1 shows the inelastic spectra of $Pr(Os_{0.32}Ru_{0.68})_4Sb_{12}$ measured at 15 and 200 K, which are integrated over the measured Q region. There is two inelastic peaks at 4 and 11 meV in the 15 K data, while these are suppressed at 200 K. These two distinct peaks arise from the crystal-field splitting levels as mentioned before. The high-temperature data show a gradual energy dependence, which may be accountable by the phonon density of states. However, it is unclear whether the rattling motion was observed or not. We also tried to measure the end material $PrRu_4Sb_{12}$ in order to compare to the result oft mixed system. Figure 2 shows data of $PrRu_4Sb_{12}$ obtained by the same measurement procedure. The clear crystal-field excitations were detected at 5.5 and 11 meV. The lower-energy excitations in the mixed and the pure systems are different in energy and intensity, which is consistent with the specific-heat study. We found also signals of the low-lying phonon for $PrRu_4Sb_{12}$, whereas the intensity is tiny. However, it is not clear in the mixed system. So far, it is not concluded whether the hybridization between ratting motion of crystal-field excitation appears in the mixed system or not.

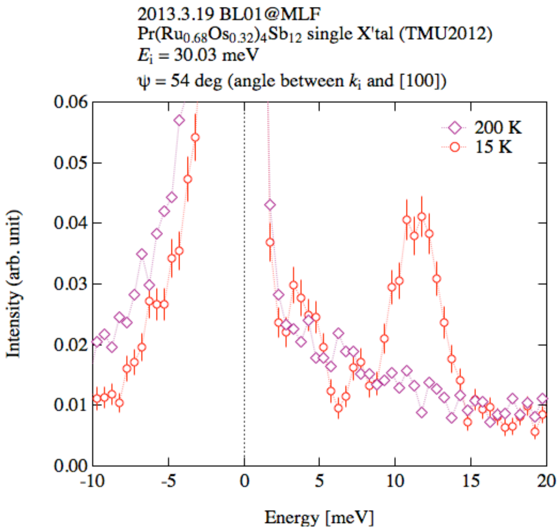

Fig. 1 Inelastic spectra of $Pr(Os_{0.32}Ru_{0.68})_4Sb_{12}$.

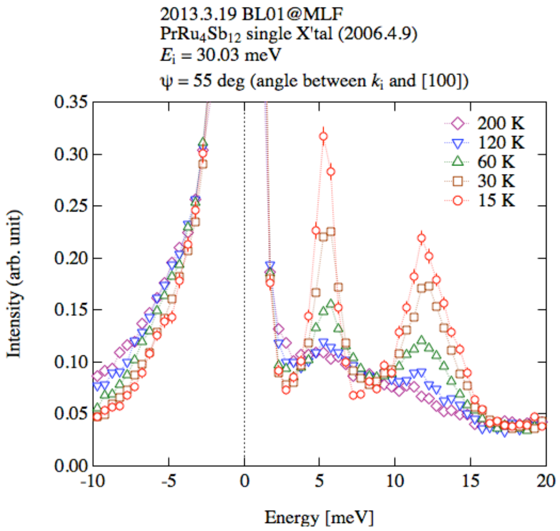

Fig. 2 Inelastic spectra of $PrRu_4Sb_{12}$.