（ ※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 Date of Report 2014.4.
課題番号 Project No. 2013B0173 実験課題名 Title of experiment Magnetic excitation in $Ce_3Co_4Sn_{13}$: a candidate for strong electronic hybridization 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 responsible person Kenji Nakajima 装置名 Name of Instrument/(BL No.) AMATERAS (BL14) 実施日 Date of Experiment 2014.3.21 – 2014.3.25

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.

$Ce_3Co_4Sn_{13}$ polycrystalline sample (4.52 g)
$La_3Co_4Sn_{13}$ polycrystalline sample (4.52 g)

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 carried out inelastic neutron scattering experiments of magnetic excitations of f-electron states of $Ce_3Co_4Sn_{13}$, which is a new candidate material exhibiting strong electronic correlation phenonena. According to the previous studies of this material (A. L. Cornelius et al.: Physica B 378-380 (2006) 113, A. Slebarski et al.: PRB 86 (2012) 205113, E. Lyle Thomas et al.: J. Solid State Chem. 179 (2006) 1642), a value of specific heat divided by temperature reaches approximately 4 J/mol-Ce/K^2 in the lowest temperature range, indicating formation of heavy electrons. Electrical resistivity shows upturn below 160 K. This upturn is considered as a charge-density-wave (CDW) formation (C. S. Lue et al.: PRB 85 (2012) 205120), which has not clearly been evidenced by the structural study so far. Below 15 K, the resistivity increases more rapidly with decreasing temperature. There is a question whether these properties, the heavy electrons and the CDW state, can coexist. The heavy-electron state is due to a dense Kondo effect with sufficient number of carriers screening the local f-electron magnetic moments, while the CDW formation reduces the carrier number. In order to clarify whether the Ce 4f electrons contribute to the conduction band or not, magnetic excitation spectra must be measured. In the present study, we succeeded in obtaining magnetic excitation spectra of the 4f electrons of $Ce_3Co_4Sn_{13}$, which is extracted from difference in spectra between $Ce_3Co_4Sn_{13}$ and $La_3Co_4Sn_{13}$.

The compounds of $Ce_3Co_4Sn_{13}$ and $La_3Co_4Sn_{13}$ were synthesized by the molten Sn self-flux method at Tohoku University.

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

The samples for present neutron scattering measurements were powdered from the tiny single crystals. The same mass of the two samples were prepared, and both of the samples were sealed inside aluminum containers with the same manner. This procedure assures us of extraction of the 4f–electron spectrum from direct subtraction of data of the La–based compound from that of the Ce–based one. The containers were installed in the GM refrigerator of BL14. We adopted two multi–E_i modes with the incident neutron energy set of E_i = 2.732, 3.130, 41.96 meV and with that of E_i = 2.243, 4.678, 15.14 meV. The pulse–shaping mode, which provides a symmetric resolution function, was used in order to discuss the spectral components.

Figure 1 shows a contour map in the E–Q space of intensity difference between $Ce_3Co_4Sn_{13}$ and $La_3Co_4Sn_{13}$ at 6 K, which was measured with E_i = 41.96 meV. There are two distinct excitation peaks located at 7 and 29 meV,. Because the intensities decrease with an increase in Q. Such sharp excitations can be attributed to transitions between crystal–field–splitting levels of the Ce–ion 4f electron state. This result is consistent with the short report of excitation peaks at 8 and 30 meV, which are assigned to the excitations from the ground state doublet to the two excited doublet levels under the cubic symmetry at the Ce–ion site (A. D. Christianson et al.: J. Magn. Magn. Mater. 310 (2007) 266). However, in the present experiments, we found that each excitation is not composed of a single peak. Figure 2 shows a contour map of intensity difference at 6 K with E_i = 15.14 meV. Details of the spectral shape of the 7–meV peak can be seen. The intensity starts to increase rapidly at 5 meV with an increase in the energy transfer, and exhibits a maximum intensity at 6.0 meV. On the other hand, intensity tail extends over 10 meV. The distinct asymmetry of the excitation peak indicates more complicated 4f–electron level schemes, which was not taken into account in the previous study under the assumption of the single cite for the Ce–ion position. Actually, we found that our sample undergoes a structural transformation at 160 K with doubling the unit–cell length, which was evidenced by the x–ray diffraction study. Therefore, it is indispensable to identify the 4f–electron level scheme of $Ce_3Co_4Sn_{13}$ by taking into account much lower structural symmetry. This fact is also important to reveal the origin of heavy–electron–like behavior and the semiconductor–like property, which are not compatible with each other as mentioned above. We are planning further neutron studies of $Ce_3Co_4Sn_{13}$ at much lower temperatures, together with other isomorphic compounds.

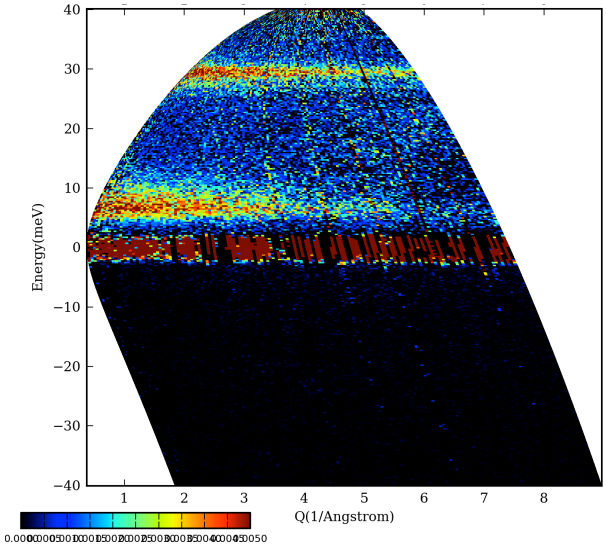

Fig.1 A contour map in the E–Q space of intensity difference between $Ce_3Co_4Sn_{13}$ and $La_3Co_4Sn_{13}$ at 6 K, which was obtained with E_i = 41.96 meV.

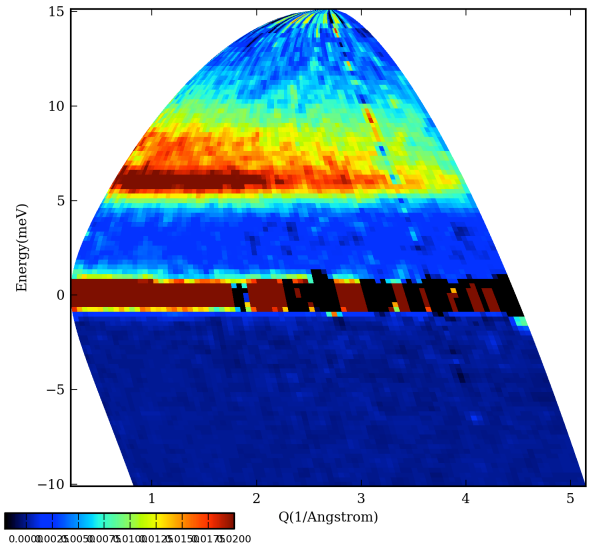

Fig.2 A contour map in the E–Q space of intensity difference between $Ce_3Co_4Sn_{13}$ and $La_3Co_4Sn_{13}$ at 6 K, which was obtained with E_i = 15.14 meV.