MLF Experimental Report	提出日 Date of Report 2017/12/13
課題番号 Project No. 2017A0030 実験課題名 Title of experiment Magnetic excitations of chiral-lattice semiconductors $Ce_3T_4Sn_{13}$ (T = Co, Ru, and Rh) 実験責任者名 Name of principal investigator Kazuaki Iwasa 所属 Affiliation Ibaraki University	装置責任者 Name of responsible person Kenji Nakajima 装置名 Name of Instrument/(BL No.) BL14 (AMATERAS) 実施日 Date of Experiment 2017/10/25 – 2017/11/1

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.

$Ce_3Ru_4Sn_{13}$ polycrystalline sample (2 g)

$La_3Ru_4Sn_{13}$ polycrystalline sample (2 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 conducted an inelastic-neutron-scattering (INS) experiment of magnetic excitations of $Ce_3Ru_4Sn_{13}$, which has been reported to exhibit strongly correlated electron phenomena. According to the previous studies, the specific-heat value divided by temperature of the isomorphic compounds $Ce_3Co_4Sn_{13}$ [1-3] and $Ce_3Rh_4Sn_{13}$ [4-6] reach approximately 0.4 – 4 J/(mol-Ce K^2) at 1 K, indicating formation of heavy electrons. The electrical resistivity data of these Ce-based compounds are less dependent on temperature compared with that of the La-based superconductors. The resistivity data slightly increase with decreasing temperature below 15 – 20 K and exhibits a maximum at 1 – 2 K. In contrast, no magnetic ordering was observed, despite that the Curie-Weiss law for the Ce^{3+} $4f^1$ configuration reproduces well the magnetic susceptibility data. These anomalous behaviors are likely due to hybridization between the Ce 4f and conduction electrons (c-f hybridization), which may cause low carrier concentration. In order to clarify the 4f-electron states in these 3-4-13 systems, we measured magnetic INS spectra of $Ce_3Ru_4Sn_{13}$. We succeeded in observing characteristic excitation spectra of the 4f electrons derived from difference in the spectra between $Ce_3Ru_4Sn_{13}$ and $La_3Ru_4Sn_{13}$, which were synthesized using the molten Sn self-flux method.

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

The polycrystalline samples of these two compounds with the same mass were sealed inside aluminum containers with the same manner. This procedure assures us of extraction of the 4*f*-electron spectrum from direct subtraction of the data of the two compounds. The containers were installed in the 1-K ^{3}He refrigerator. We adopted the multi-E_i mode with the incident neutron energies, E_i = 3.43, 5.25, 8.98, 18.7, and 60.4 meV. The pulse-shaping choppers were used in order to discuss the spectral shape.

Figure 1 shows an energy spectrum obtained with E_i = 60.4 meV, the intensity of which is the sum in the Q = 3 – 5 Å^{-1} region. A distinctly broad excitation peak located at 18 meV was observed. This result is considerably different from the previous results showing the sharp excitation peaks between crystalline-electric-field (CEF) splitting levels of the Ce^{3+} 4*f*-electron state at 6.4 and 29 meV for $Ce_3Co_4Sn_{13}$ [7] (Proposal Nos. 2014P0202 and 2013B0173) and those at 7 and 39 meV for $Ce_3Rh_4Sn_{13}$ [8] (Proposal No. 2016A0063). The present Ru-based compound takes stronger hybridization effect between the 4*f* and conduction electrons. Figure 2 shows a spectra measured using E_i = 3.44 meV. The data sets at 20 K show quasielastic-like spectrum, while the data at 0.6 K show an inelastic feature at approximately 0.8 meV. This emergence of inelastic response at the low temperature indicates that the 4*f* electrons in $Ce_3Ru_4Sn_{13}$ forms spin excitation with respect to the doublet CEF ground state. The similar spin excitations were also observed in the isomorphic compound $Ce_3Co_4Sn_{13}$ and $Ce_3Rh_4Sn_{13}$, which emerge with the upturn in electrical resistivity below 15 K. However, the peak position and the spectral width of $Ce_3Ru_4Sn_{13}$ are larger than those observed for $Ce_3Co_4Sn_{13}$ and $Ce_3Rh_4Sn_{13}$. Therefore, an energy scale of the spin correlation is enhanced for $Ce_3Ru_4Sn_{13}$.

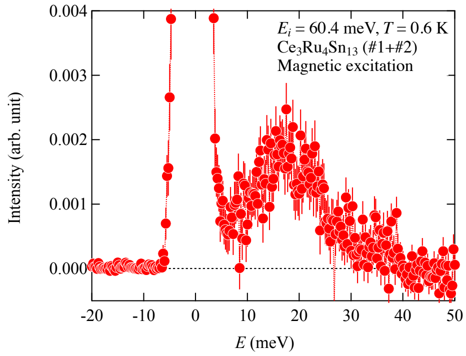

Fig. 1 Measured magnetic INS spectrum at 0.6 K using E_i = 60.4 meV.

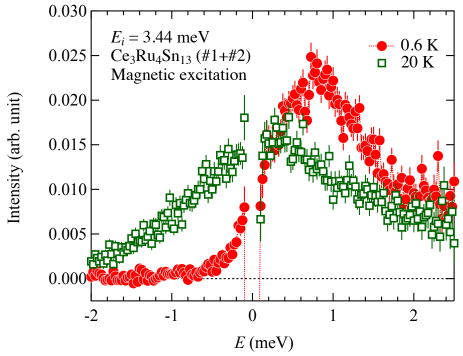

Fig. 2 Measured magnetic INS spectrum at 0.6 and 20 K using E_i = 3.44 meV.

We found that the crystal structure of $Ce_3Co_4Sn_{13}$ below 160 K and that of $Ce_3Rh_4Sn_{13}$ below 250 K are characterized by chiral symmetry [9, 10]. This structural finding supports that the electrons in these compounds are the Wyle fermions [11]. The enhancements in the *c*-*f* hybridization and the spin correlation of $Ce_3Ru_4Sn_{13}$ are also interested in terms of electronic states characteristic of the chiral-lattice system. Therefore, the structural investigation is required for understanding the electronic and magnetic properties.

References [1] A. L. Cornelius et al.: Physica B 378-380 (2006) 113. [2] A. Ślebarski et al.: PRB 86 (2012) 205113. [3] E. L. Thomas et al.: J. Solid State Chem. 179 (2006) 1642. [4] U. Köhler et al., J. Phys.: Condens. Matter **19**, 386207 (2007). [5] M. Gamża et al., J. Phys.: Condens. Matter **20**, 395208 (2008). [6] A. Ślebarski et al., PRB **86**, 205113 (2012). [7] K. Iwasa et al., PRB 95, 195156 (2017). [8] K. Iwasa et al., in preparation. [9] Y. Otomo et al., PRB, 075109 (2016). [10] K. Suyama et al., in preparation. [11] J. L. Mañes, PRB **85**, 155118 (2012).