MLF Experimental Report

提出日 Date of Report	
課題番号 Project No. 2016A0063	装置責任者 Name of responsible person Kenji Nakajima
実験課題名 Title of experiment Magnetic excitations of chiral-lattice semiconductors $Ce_3T_4Sn_{13}$ (T = Co, Ru, and Rh)	装置名 Name of Instrument/（BL No.） BL14 (AMATERAS)
実験責任者名 Name of principal investigator Kazuaki Iwasa	実施日 Date of Experiment 2016/11/4 – 2016/11/11
所属 Affiliation Ibaraki University	

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.

$Ce_3Rh_4Sn_{13}$ polycrystalline sample (5.04 g)

$La_3Rh_4Sn_{13}$ polycrystalline sample (5.08 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 $Ce_3Rh_4Sn_{13}$, which has been a new candidate material exhibiting strongly correlated electron phenomena. According to the previous studies [U. Köhler et al., J. Phys.: Condens. Matter **19**, 386207 (2007), M. Gamża et al., J. Phys.: Condens. Matter **20**, 395208 (2008), A. Ślebarski et al., PRB **86**, 205113 (2012)], the specific-heat value divided by temperature reaches approximately 4 J/(mol-Ce K^2) at 1 K, indicating formation of heavy electrons. The electrical resistivity of $Ce_3Rh_4Sn_{13}$ is less dependent on temperature compared with that of the superconductor $La_3Rh_4Sn_{13}$. The resistivity slightly increases with decreasing temperature below 15 K and exhibits a maximum at 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 state, we measured magnetic INS spectra. We succeeded in observing characteristic excitation spectra of the $4f$ electrons derived from difference in the spectra between $Ce_3Rh_4Sn_{13}$ and $La_3Rh_4Sn_{13}$, 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 prepared, and both of the samples 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 La-based compound from that of the Ce-based one. 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 = 3.44, 8.97, and 60.4 meV. The pulse-shaping choppers were used in order to discuss the spectral shape.

Figure 1 shows an energy spectrum obtained using the E_i = 60.4 meV setup, the intensity of which is the sum in the Q = 3 − 5 Å^{-1} region. Two distinct excitation peaks located at 7 and 39 meV were observed. This result is slightly different from the previous result showing the excitation peaks at 9 and 38 meV [D. T. Adroja et al., Physica B **403**, 898 (2008)], which may arise from the difference in the sample-synthesis method. These sharp excitations can be attributed to electronic transitions between crystalline-electric-field (CEF) splitting levels of the Ce^{3+} 4*f*-electron state, which are composed to the three Kramers doublets. However, in the measurement using E_i = 8.97 meV, we found that the low-energy excitation consists of two peaks at 6.0 and 7.2 meV. Thus, the CEF split is not uniform. Figure 2 shows a spectra measured using E_i = 3.44 meV. The data sets at 10 and 30 K show quasielastic intensities below approximately 1 meV, while that at 0.7 K shows an inelastic feature at approximately 0.2 meV. This emergence of inelastic response below 10 K indicates that the 4*f* electrons in $Ce_3Rh_4Sn_{13}$ forms spin excitation with respect to the doublet CEF ground state, which appears simultaneously with the anomalous resistivity enhancement. Moreover, the large specific-heat value at 1 K is assigned to the spin excitation. The similar spin excitation was also observed in the isomorphic compound $Ce_3Co_4Sn_{13}$, which also exhibits the upturn in electrical resistivity below 15 K and the large specific-heat value. The low-density carrier of $Ce_3Co_4Sn_{13}$ and $Ce_3Rh_4Sn_{13}$ interacts with the Ce 4*f* electrons, and the anomalous spin excitations are induced in the paramagnetic phase.

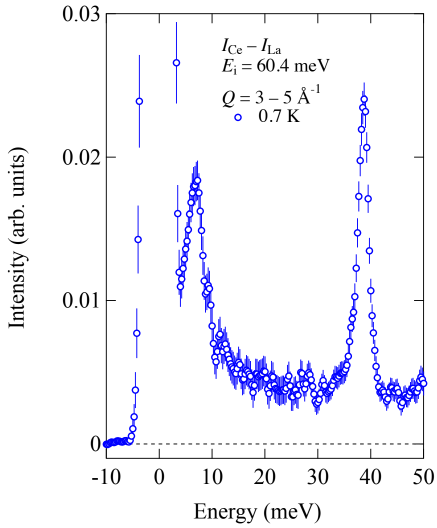

Fig. 1 Measured magnetic INS spectrum at 0.7 K using E_i = 60.4 meV.

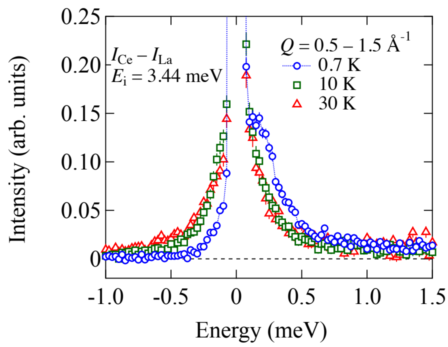

Fig. 2 Measured magnetic INS spectrum at 0.7, 10, and 30 K using E_i = 3.44 meV.

Recently, 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 [Y. Otomo et al., PRB, 075109 (2016) and K. Suyama et al., in preparation]. This structural finding supports that the electrons in these compounds are the Wyle fermions [J. L. Mañes, PRB **85**, 155118 (2012)]. The low-density carrier state is expected to originate from the *c*-*f* hybridization tuning the Fermi level near the crossing point of the linear dispersion of the Weyl electrons. This conduction electron gives rise to the observed anomalous spin excitations via the RKKY interaction.