実験報告書様式(一般利用課題·成果公開利用)

MLF Experimental Report	提出日 Date of Report
課題番号 Project No.	装置責任者 Name of responsible person
2017A0029	Kenji Nakajima
実験課題名 Title of experiment	装置名 Name of Instrument/(BL No.)
Two-channel Kondo effect of PrT2X20 (T = transition metal	BL14 (AMATERAS)
elements, X = Al, Zn)	実施日 Date of Experiment
実験責任者名 Name of principal investigator	2017/4/17 - 2017/4/19
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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.

PrRu2Zn20 polycrystalline sample (2.9 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 study *f*-electron states of intermetallic compounds, $\Pr T_2 Zn_{20}$ (*T* = transition metals), which crystallize into the *Fd-3m* cubic structure. These are attractive systems because of non-magnetic multipole ordering of Pr-ion *f* electrons and characteristic electric conductivities (T. Onimaru and H. Kusunose: JPSJ 85, 082002 (2016)). $\Pr Ir_2 Zn_{20}$ exhibits a superconductivity below 0.05 K and a phase transition at 0.1 K. This phase transition is attributed to a multipole ordering originating from the non-Kramers doublet ground state of the $\Pr^{3+} f^2$ configuration (K. Iwasa et al.: PRB 95, 155106 (2017)).

In contrast, the isomorphic $PrRu_2Zn_{20}$ does not exhibit any phase transition. A previous inelastic-neutron-scattering (INS) study did not resolve the crystal-electric-field (CEF) levels (K. Iwasa et al.: JPSJ 82, 043707 (2013)). The electron diffraction measurement has suggested a crystal-structure transformation at 138 K (T. Onimaru et al.: J. Phys.: Condens. Matter 24, 294207 (2012)). Therefore, we carried out an INS measurement for investigating magnetic excitation spectra of $PrRu_2Zn_{20}$ in the low-temperature phase by using he high-resolution disk-chopper spectrometer AMATERAS (BL14).

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

The polycrystalline sample was sealed inside an aluminum container. We use a GM cryostat with the base temperature of 5 K. We adopted a multi- E_i mode with the incident neutron energy set of E_i = 2.24, 4.67, and 15.13 meV. The pulse-shaping mode was used to obtain symmetric resolution functions.

Figure 1 shows magnetic excitation spectra of $PrRu_2Zn_{20}$ measured using $E_i = 15.13$ meV. The spectrum at 5 K is composed of excitation peaks at approximately 0.84, 1.97, 3.37, 3.96, 5.04, 5.82, 7.46, and 8.37 meV, as indicated by solid line for the fitted result. These sharp excitations depend on temperatures, and are attributed to transitions between the CEF splitting of well-localized Pr-ion $4f^2$ electron configuration. The multi-peak structure between 2 and 6 meV is observed more clearly, compared to the previous measurement. The sharp peaks are suggestive of less hybridization effect between the *f* and conduction electrons. The spectrum 1t 150 K shown in the bottom part of Fig. 1 is thermally broadened, and it is difficult to identify the excitation levels.

Figure 2 shows results of low-energy spectra of $PrRu_2Zn_{20}$ by using $E_i = 2.24$ meV. The background intensities exhibit monotonic increase with increasing energy. The spectrum is also contains many peaks at approximately 0.490, 0.779, 1.047, 1.776 meV, as indicated by solid line for the fitted result. These also exhibit temperature dependencies.

These experimental results indicate that the cubic symmetry at the Pr sites is broken on the structural transition at 138 K, because the present result is not consistent with an expected four CEF levels in the original Fd-3m structure. Therefore, the degeneracy of the non-Kramers doublet ground state is lifted, which means the 4f-electron multipole fluctuation is no longer active in the low-temperature region of PrRu₂Zn₂₀. The crystal structure in the low-temperature phase is characterized by the wave vector $\mathbf{q} = (2/3, 2/3, 0)$ (T. Onimaru et al.: J. Phys.: Condens. Matter 24, 294207 (2012)). Such long-period structure is expected to contain different Wyckoff sites for the Pr ions, and the several individual CEF schemes appear below 138 K.







Fig. 1. INS spectra of $PrRu_2Zn_{20}$ measured with $E_i = 4.67$ meV on BL14.