実験報告書様式（一般利用課題・成果公開利用）

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# MLF Experimental Report	提出日 Date of Report 2014.6.28
課題番号 Project No. 2014A0261 実験課題名 Title of experiment Ways to release entropies at low temperatures in PrT2Zn20 (T = Ru, Rh, Os, Ir) 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 2014.4.25 – 2014.5.1 2014.5.23 – 2014.5.28

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.

$PrOs_2Zn_{20}$ polycrystalline sample (1.2 g)
$PrIr_2Zn_{20}$ polycrystalline sample (2.5 g)
Empty cells

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, PrT_2Zn_{20} (T = Ir, Ru, Rh), 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 et al.: JPSJ 79 (2010) 033704, PRL 106 (2011) 177001 K. Wakiya et al.: J. Korean Phys. Soc. 62 (2013) 2143). $PrIr_2Zn_{20}$ exhibits superconductivity below 0.05 K. There is another phase transition at 0.1 K found by specific heat measurement. This phase transtion is attributed to a quadrupole ordering originating from the non-Kramers doublet ground state of f^2 configuration of Pr^{3+}. It should be noted that entropy value at the transition temperature of 0.1 K is approximately 1/3 of Rln2 [J/K/mol], and it reaches Rln2 [J/K/mol] at 1 K. Such gradual release of the magnetic entropy cannot be explained by splitting of the degenerated levels of well-localized *f* electrons on the phase transition. In order to understand this phenomenon, we examine an electronic hybridization effect or orbital Kondo effect, which might be investigated by measurement of low-energy magnetic spectrum in lower-temperature range. $PrOs_2Zn_{20}$ does not exhibit any phase transition down to 0.4 K, despite that specific heat data suggests a degenerated ground state. There is a question why the Os-based compound does not exhibit any electronic ordering at low temperature. It is probable that the electronic hybridized state is stable in $PrOs_2Zn_{20}$. Therefore, we carried out two experiments for investigating magnetic excitation spectra of $PrIr_2Zn_{20}$ and $PrOs_2Zn_{20}$.

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 polycrystalline crystals. These were sealed inside aluminum containers. In order to avoid strong absorption effect of Ir nuclei, the sample of $PrIr_2Zn_{20}$ was formed into a pipe-like shape. We use a carry-in ^{3}He cryostat with the base temperature of 0.6 K. We adopted a multi-E_i mode with the incident neutron energy set of E_i = 2.24, 4.68, and 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 magnetic excitation spectra of $PrOs_2Zn_{20}$ measured at 4 K (solid line) together with those of $PrIr_2Zn_{20}$ at 0.6 K (solid circles with dotted line) and $PrRh_2Zn_{20}$ at 5 K measured previously (broken line, K. Iwasa et al. J. Phys. Soc. Jpn. 82 (2013) 043707). The spectra of $PrIr_2Zn_{20}$ and $PrRh_2Zn_{20}$ are composed of two sharp excitation peaks at approximately 2.5 and 5.5 meV, which are explained by the crystal-field splitting of well-localized Pr-ion $4f^2$ electron configuration under the cubic symmetry. In contrast to these results, the spectrum of $PrOs_2Zn_{20}$ is apparently broadened. This fact is suggestive of hybridization between *f* and conduction electrons. In addition, we observed three excitation peaks of $PrOs_2Zn_{20}$ located at 1.4, 3.5, and 5.8 meV. It means that there are four splitting levels which is more than expected for cubic symmetry. Although we do not fully solve a crystal-filed level scheme, the spectrum of $PrOs_2Zn_{20}$ can be reproduced by a hexagonal symmetry with the doubly degenerated ground state and the spectral broadening effect.

Figure 2 shows results of low-energy spectra of $PrIr_2Zn_{20}$ measured by using E_i = 2.24 meV. The background intensities obtained from the empty-cell data are subtracted from the data of the samples. There is no clear excitation between 0.1 and 1 meV at any temperature between 0.6 and 6.5 K. This result seems to indicate no electronic density-of-state in this energy region, and to be inconsistent with the gradual release of magnetic entropy up to 1 K described above. The same high-resolution measurement was also performed for $PrOs_2Zn_{20}$, and the result is similar to $PrIr_2Zn_{20}$. These results of the two materials are explained by the ground state doublet that does not hold dipolar transition probability detected by neutron scattering. This fact is important for the low-temperature state dominated by non-dipolar magnetic fluctuation, expected for the orbital Kondo effect.

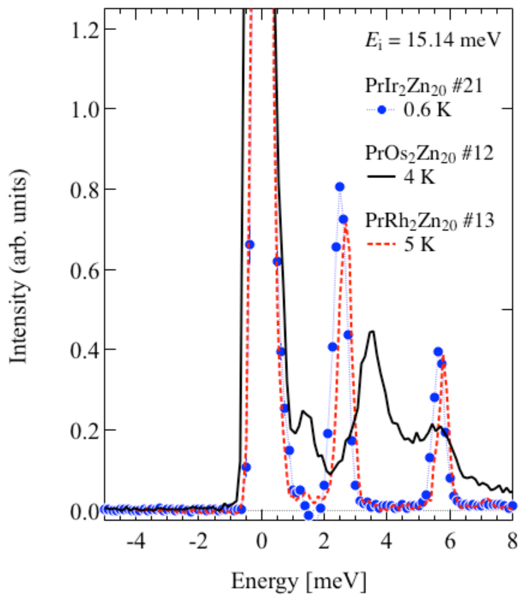

Fig. 1. Inelastic neutron scattering spectra of PrT_2Zn_{20} (T = Ir, Os, and Rh) measured with E_i = 15.14 meV on BL14.

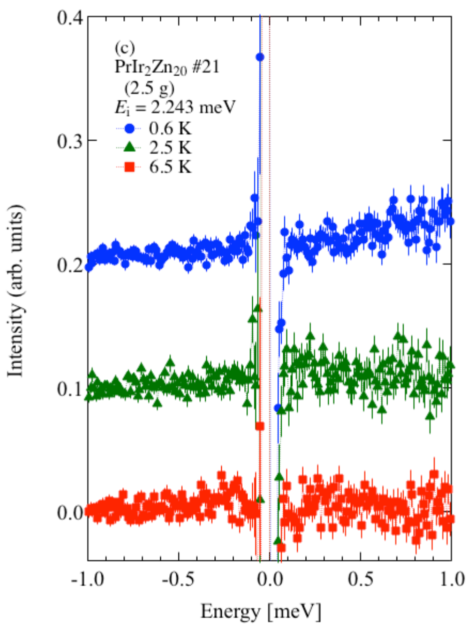

Fig. 2. Inelastic neutron scattering spectra of $PrIr_2Zn_{20}$ measured with E_i = 2.243 meV on BL14.