Magnetic excitation in paramagnetic semimetal $Yb_3Ir_4Ge_{13}$

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1. Introduction

Heavy fermion (HF) behaviors are central issues in the researches on strongly correlated electrons in rare-earth-based materials [1]. In the present study, we investigated correlated electrons in the 3-4-13 class of materials, which is of special interest because of its diverse physical properties, in particular their HF like behaviors. $Yb_3Ir_4Ge_{13}$ crystallizes in a tetragonal structure categorized in the space group $I4_1/amd$ (No. 141) [2, 3]. The ground state of $Yb_3Ir_4Ge_{13}$ is paramagnetic, in contrast to a magnetic susceptibility showing an cusp-like anomaly at T_{mag} = 0.9 K and a Weiss temperature estimated to be –17 K, which indicates an antiferromagnetic interaction between the Yb ions. At T_{mag}, a specific heat divided by temperature also exhibits a peak anomaly, which moves to higher temperature side with applied magnetic fields. An electrical resistivity increases with decreasing temperature (0.3-300 K). The data above 100 K is reproduced by an activation type behavior with an energy gap of 120 K, which might be attributed to a Kondo semiconductor or semimetal state. However, the resistivity deviates downward from this activation type, and shows a plateau-like behavior near 20 K. In addition, the resistivity increases rapidly below 20 K. These unconventional behaviors remain unexplained, although the Kondo effect basically plays a role in these electronic phenomena. Microscopic investigations of the electronic state, in particular the Yb 4*f* electron magnetic state, is highly required for further understanding.

In the present study, we performed high energy-resolution neutron spectroscopy in order to extract magnetic excitation in the temperature range of the anomaly in the electrical resistivity. As a result, below approximately 20 K, we observed a drastic enhancement in the excitation below approximately 3 meV, which is closely relevant to the anomalies in the resistivity, the magnetic susceptibility, and the specific heat at T_{mag}.

2. Experiment

Samples of $RE_3Ir_4Ge_{13}$ (*RE*: Yb and Lu) were synthesized using the molten Ge-flux method [4]. Several pieces of crystals (3.84 and 4.13 g of the Yb- and Lu-based materials, respectively) were selected for inelastic neutron scattering (INS) measurements. INS experiments were performed using the AMATERAS (BL14) [5] from April 4th to 11th, 2018 (proposal No. 2018A0150). Because of the almost same sample mass of $Yb_3Ir_4Ge_{13}$ and non-magnetic $Lu_3Ir_4Ge_{13}$ specimens, the Yb 4*f* electron contribution to the spectra was extracted from the difference between the data for these two compounds. A set of incident neutron energies were selected via chopper combination; E_i = 3.439, 5.246, 8.967, 18.69, and 60.39 meV. The Utsusemi software suite was used to analyze the spectral data obtained via the pulsed-neutron scattering technique [6]. Sample temperatures in the INS measurements were controlled between 0.6 and 50 K using a ^{3}He closed-cycle refrigerator.

3. Results

Figure shows energy spectra at (a) 0.6 and (b) 20 K extracted from the measurements with E_i = 3.439 meV. The measured spectra were integrated over Q = 0.75 - 1.75 Å^{-1}, and a difference between $Yb_3Ir_4Ge_{13}$ and $Lu_3Ir_4Ge_{13}$ are plotted in the figure. It means that the data of $Lu_3Ir_4Ge_{13}$

was used for estimating phonon contributions and background counts from the sample cell filling with helium gas for thermal contact between the sample and the refrigerator. We obtained almost pure magnetic scattering from $Yb_3Ir_4Ge_{13}$ after the subtraction of the $Lu_3Ir_4Ge_{13}$ data. The spectrum at 0.6 K shows a peak at 0.4 meV and a tail up to 3 meV. At 20 K, the peak is suppressed, and the intensity in the negative E region is enhanced. Considering the temperature factor for INS spectrum, $n(E) + 1$, where $n(E)$ is the Bose-Einstein distribution function, we obtained emergence of this low energy excitation below 20 K, where the electrical resistivity shows an enhancement with decreasing temperature. We carried out a least-squares fitting analysis to the obtained magnetic scattering on the basis of the damped harmonic oscillator (DHO) model, and results are shown by solid lines in the figures. The results are in agreement with the measurement data.

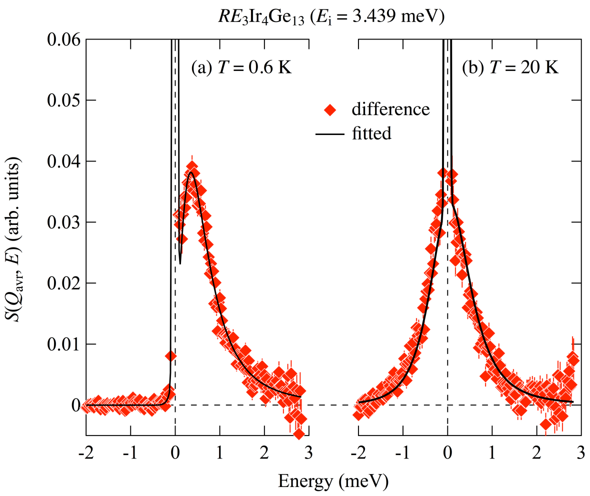

FIG. INS spectra correspond to the difference between the Yb- and Lu-based compounds at (a) 0.6 K and (b) 20 K measured using E_i = 3.439 meV. Intensities were integration in the Q range between 0.75 and 1.75 Å^{-1}.

4. Conclusion

As described above, the low-energy magnetic excitation emerges below 20 K in a paramagnetic semimetal phase of $Yb_3Ir_4Ge_{13}$. The spectrum is well reproduced by the DHO model, which indicates characteristic spin fluctuation. In contrast, no clear crystal-electric-field excitations were detected, despite that the Yb^{3+} $4f^{13}$-electron configuration should split into four Kramers doublets. These facts indicate that a strongly hybridized state between the $4f$ and conduction electrons exhibits the DHO type spin fluctuation. Thus, the electrical conductivity behaves unusual increment with the spin-fluctuation emergence. For further investigation, a single-crystal INS study is required in order to reveal spatial correlation of the spin dynamics.

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