Magnetic excitation of a heavy-fermion material $NdO_{0.5}F_{0.5}BiS_2$

Frontier Reseach Center for Applied Atomic Sciences, Ibaraki University Kazuaki Iwasa

1. Introduction

Heavy-fermion (HF) behaviors have extensively been studied for various *f*-electron systems [1]. The magnetic Kondo effect has been established in various Ce-based $4f^1$ intermetallic compounds, and the two-channel (non-magnetic quadrupole) Kondo effect has been investigated for the Pr-based $4f^2$ case [2]. Very recently, Hotta proposed theoretically the two-channel Kondo effect in the Nd^{3+} $4f^3$ configuration [3]. Therefore, an experimental attempt to search for the HF state in Nd^{3+}-based $4f^3$ systems is a next target.

Recently, $NdO_{0.5}F_{0.5}BiS_2$ has been proposed to be a HF material by the group of Tokyo Metropolitan University. This compound is a family of the Bi-based superconductor, and is a candidate material for high-performance thermoelectricity. The specific heat divided by temperature of $NdO_{0.5}F_{0.5}BiS_2$ reaches an extremely large value of approximately 7.5 J/(K^2 mol) at 0.3 K. The temperature dependence of magnetic susceptibility saturates in the same low-temperature region, which seems to be a signature of mass-enhanced Pauli paramagnetic state. The high-temperature magnetic susceptibility follows a Curie-Weiss law with an effective magnetic moment of Nd^{3+} ion, the crystal-electric-field (CEF) energy of which is estimated to be 90 K between the ground state and the first excited state. This resistivity data show an enhancement with decreasing temperature below approximately 200 K, while it drops to zero below 5 K [4]. The superconductivity is controversial, because no distinct anomaly of specific heat at 5 K was observed. However, the resistivity enhancement can be considered to indicate strong electronic correlation.

Following the aforementioned studies, we have initiated an inelastic neutron scattering (INS) study of $NdO_{0.5}F_{0.5}BiS_2$. The CEF excitation peaks were observed at 8, 13, and 23 meV for the polycrystalline sample using the chopper spectrometer HRC (BL12). Because of the local point-group symmetry, C_{4v} (*4mm*), at the Nd^{3+} $4f^3$ site, five Kramers doublets in the CEF scheme associated with the total angular momentum, $J = 9/2$, appear. The observed spectrum is consistent with such electronic state. It is expected that the ground state doublet may form a quasielastic spectrum, if the HF or Kondo state realizes. The CEF excitation spectrum obtained at HRC does not show any clear spectral broadening, and thus the hybridization between 4*f* and conduction electrons is not clear within the measurement with energy resolution of approximately 1 meV. The spectral width is estimated to be in the order of 0.1 meV or less, because this energy corresponds to the temperature range of the large specific heat.

In order to examine the validity of HF associated with hybridization between the Nd 4*f* and conduction electrons of $NdO_{0.5}F_{0.5}BiS_2$, we conducted an INS measurement using high energy-resolution spectrometer DNA (BL02), the energy resolution of which is 3.7 μeV.

2. Experiment

Polycrystalline samples of $NdO_{0.5}F_{0.5}BiS_2$ were synthesized by the group of Tokyo Metropolitan University. This sample was sealed in an aluminum can filling with helium gas. Sample temperatures were controlled down to 0.29 K using a ^{3}He cryostat installed in MLF. INS measurements were performed using the inverted geometry spectrometer DNA (BL02). Fast rotating choppers were used to obtain fine energy resolution, and several sets of chopper phase shift were applied to obtain a full spectrum between the energy –0.5 to +1.0 meV.

3. Results

Figure shows energy spectra measured at 0.29 K (red circles) and 4.0 K (black diamonds). It is noteworthy that clear excitations were detected. We are analyzing the data taking into account INS and quasielastic components. Considering a neutron scattering function, $S(E) = [n(E) + 1] \chi''(E)$, where $n(E)$ and $\chi''(E)$ are the Bose-Einstein distribution function and the imaginary part of generalized magnetic susceptibility, we see that $\chi''(E)$ is clearly enhanced with decreasing temperature. The spectral width is in the order of 0.1 meV, which is consistent with the low-temperature characteristic behaviors in the magnetic susceptibility and the specific heat. Therefore, the observed result is a clear signature of the HF formation associated with the Nd 4*f*-electron state.

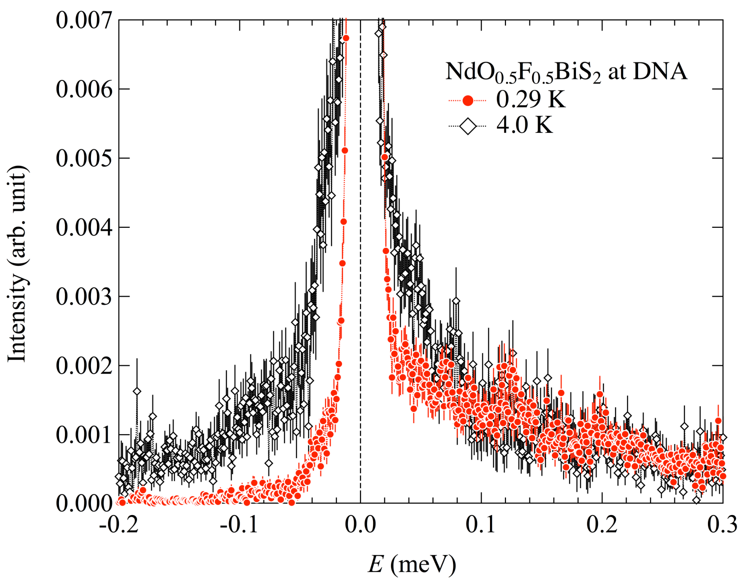

FIG. Magnetic excitation spectra of $NdO_{0.5}F_{0.5}BiS_2$ at 0.29 and 4.0 K measured at DNA.

4. Conclusion

Very fine energy-resolution INS measurement evidenced the broadening of the Nd-ion CEF ground state, which is considered to be a first-time observation of the HF state associated with the Nd 4*f*-electron state. As mentioned above, the HF or the Kondo effect in Ce-based $4f^1$ and Pr-based $4f^2$ intermetallic compounds has been established, and the possibility in the case of Nd^{3+} $4f^3$ configuration has just been discussed. The present INS study on $NdO_{0.5}F_{0.5}BiS_2$ opens an avenue to evidence experimentally the new type HF state.

Acknowledgement

This study has been conducted under the collaboration with Y. Aoki, T. D. Matsuda, and R. Higashinaka (Tokyo Metropolitan University). The neutron scattering experiment was performed with M. Matsuura and the technical staff members (CROSS and J-PARC Center). This study was supported in part by the Japan Society for the Promotion of Science, KAKENHI.

References

[1] T. Kasuya and T. Saso (Eds.), Theory of Heavy Fermions and Valence Fluctuations, (Springer, Heidelberg, 1985). [2] see for example, T. Onimaru and H. Ksunose, J. Phys. Soc. Jpn. **85**, 082002 (2016). [3] T. Hotta, J. Phys. Soc. Jpn. **86**, 083704 (2017). [4] R. Jha et al. J. Appl. Phys. **113**, 056102 (2013).