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MAGNETIC EXCITATIONS IN THE HEAVY-ELECTRON AND ANTIFERRO-QUADRUPOLAR-ORDERING SYSTEM $\mathbf{PrFe_4P_{12}}$* **

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$PrFe_4P_{12}$ exhibits a heavy-electron-like behavior and a phase transition to an antiferro-quadrupolar ordering state at the lowest temperature. Inelastic neutron scattering experiments for a polycrystalline sample revealed a quasielastic response and no crystal-field excitation in the heavy-electron state. It is in contrast to the observed sharp crystal-field excitations in $PrInAg_2$ considered as a candidate of a quadrupolar Kondo system. In the ordered phase, sharp peaks at 1.4 and 3.0 meV are superimposed on the broad spectrum. These sharp excitations are consistent with the localized behavior of the $4f$ electrons in the antiferro-quadrupolar ordered phase.

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1. Introduction

Among the various physical properties of the filled skutterudites RT_4X_{12} (R = rare earth, T = transition metal, X = pnictogen) crystallizing in $Im\bar{3}$ cubic structure [1], $PrFe_4P_{12}$ shows heavy-electron phenomena and an unusual phase transition. A peak of specific heat and a jump of electrical resistivity observed at $T_A = 6.5$ K at zero magnetic field indicate a phase transition [1,2]. In the region where the low-temperature phase below T_A is

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destroyed by external magnetic fields, the electronic specific-heat coefficient for the T-linear term becomes as large as 1.4 J/mol/K^2. The electrical resistivity behaves as $-\log T$ between 30 and 100 K. Cyclotron effective mass was evaluated as $81m_0$ from a de Haas–van Alphen effect experiment [3]. X-ray diffraction experiments revealed a superlattice characterized by the wave vector $\boldsymbol{q} = (1, 0, 0)$ below T_A [4]. This wave vector is consistent with the Fermi-surface nesting condition predicted by the band calculation [5]. Moreover, neutron diffraction studies revealed an antiferromagnetic component with the same $\boldsymbol{q}$ induced by applied magnetic field in the low-temperature phase [6]. This phenomenon is ascribed to the antiferro-quadrupolar (AFQ) ordering of the localized Pr^{3+}-ion $4f$ electrons. In order to investigate the $4f$-electron contribution to the heavy-electron state and the AFQ ordered phase, we performed inelastic neutron scattering experiments.

2. Experimental procedure

Polycrystalline samples of $\mathrm{PrFe_4P_{12}}$ and $\mathrm{LaFe_4P_{12}}$ as a reference system without a $4f$ electron were synthesized by a tin flux method. Inelastic neutron scattering experiments were carried out by a spectrometer LAM-D with crystal-analyzer mirrors selecting the final neutron energy of 4.6 meV installed in the pulsed-neutron facility KENS of KEK, Tsukuba, Japan. Sample temperature was controlled down to 3.5 K by a He-flow cryostat.

3. Experimental results and analysis

Figure 1 represents measured responses of $\mathrm{PrFe_4P_{12}}$ as a function of excitation energy E. These results were obtained after a correction of scattered intensity by an energy dependence of incident-neutron intensity and subtractions of backgrounds and phonon contributions estimated from the measurement of $\mathrm{LaFe_4P_{12}}$. Incoherent scattering contribution around $E = 0$ was excluded. At 60 and 15 K, we observed quasielastic spectra whose widths are much broader than the energy resolution indicated by a horizontal bar. There was no clear inelastic peak in the range up to 100 meV. The following equation with a Lorentzian relaxation function is used for analysis.

$$I = Cf^2 \frac{E}{1 - \exp(-E/k_\mathrm{B}T)} \frac{1}{\pi} \frac{\Gamma}{(E - E_0)^2 + \Gamma^2} , \tag{1}$$

where C is a scale factor, f is a magnetic form factor of Pr^{3+}. A least-squares fitting procedure based on Eq. (1) convoluted with the instrumental resolution was performed with free parameters of C and Γ and fixed $E_0 = 0$. The results shown by lines agree well with the experimental data. The obtained Γ are 4.1 and 1.8 meV for 60 and 15 K, respectively.

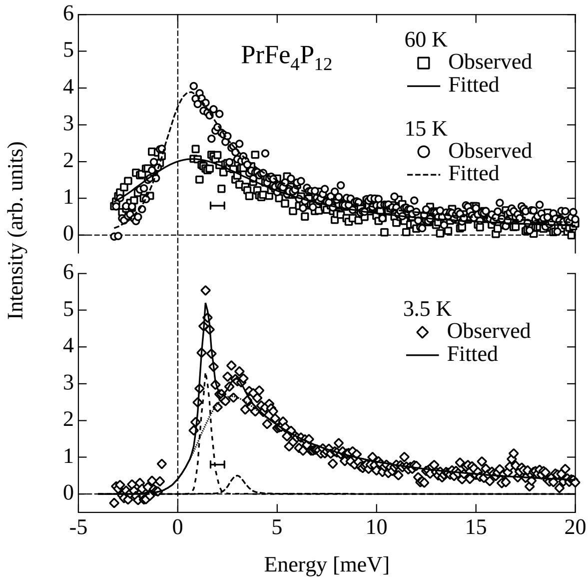

Fig. 1. Inelastic neutron scattering spectra of polycrystalline $PrFe_4P_{12}$.

At 3.5 K below T_A, as depicted in the lower part of Fig. 1, sharp peaks at 1.4 and 3.0 meV are seen together with a broad spectrum. Performing a least-squares fit, we found that three Lorentzians centered at 1.4, 1.6 and 3.0 meV reproduce well the data, as shown by a solid line and the components by broken lines. The width parameters Γ of the peaks at 1.4 and 3.0 meV are less than 0.1 meV. The peak at 1.6 meV has a large $\Gamma = 2.1$ meV. It should be mentioned that the center of the broad peak is not determined uniquely. We obtained similar fit with a set of one peak with center fixed at $E = 0$ and $\Gamma = 2.2$ meV and two inelastic peaks at 1.4 and 2.9 meV.

4. Discussion

The quasielastic spectra without apparent crystal-field excitations above T_A is similar to those of typical heavy-fermion systems [7]. Because the crystal-field ground state of Pr ions is suggested to be a non-magnetic doublet Γ_3 from the elastic constant measurement [8] and the theoretical argument [9], a quadrupole Kondo effect is expected to be important in $PrFe_4P_{12}$. However, $PrInAg_2$ studied as a candidate of the quadrupole Kondo system exhibits clear crystal-field excitations [10]. This fact is in contrast to the present result of $PrFe_4P_{12}$. Their difference in energy spectra is a key to understand heavy-electron states in Pr compounds.

The sharp peaks at 3.5 K is attributed to the localized $4f$-electrons exhibiting the AFQ ordering with the ground state Γ_3. The complicated spectrum is considered to be due to dispersions of excitation state in the ordered phase below T_A. Another possible interpretation is that the broad Lorentzian spectrum appearing above T_A is conserved even in the AFQ ordered phase. Further neutron scattering experiments for a single-crystal sample are necessary to interpret the whole spectrum of $4f$-electron state.

5. Concluding remarks

In conclusion, the present inelastic neutron scattering study of $PrFe_4P_{12}$ elucidates the broad magnetic response of $4f$-electrons above T_A in the heavy-electron state and the sharp excitations superimposed below T_A. The distinct change of the spectral shape through the phase transition is a direct evidence for the significant contribution of the $4f$-electrons to the heavy-electron state and the AFQ ordering state of $PrFe_4P_{12}$.

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