Magnetic Excitations in the Pr-based Filled Skutterudite Compound PrFe₄P₁₂ Revealed by ³¹P-NMR Study

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³¹P-NMR studies have been carried out to investigate magnetic properties in Pr-based filled-Skutterudite compound $PrFe_4P_{12}$, which shows an unusual phase transition at $T_A = 6.2$ K. This is now regarded as the transition from the paramagnetic to antiferro-quadrupole (AFQ) ordering states from X-ray diffraction experiments [1]. The splitting of P-NMR spectrum, which is due to the appearance of two P sites with different hyperfine fields, was observed below T_A . The splitting, however, seems to disappear in zero magnetic field from the field dependence, indicating that the different hyperfine fields at the P sites are not due to the magnetic order, but due to the appearance of the inequivalent two P sites below T_A . This is ascribed to the distortion of the P cage surrounding a Pr ion below T_A , which gives rise to a Pr orbital ordering. Nuclear spin-lattice relaxation rate $(1/T_1)$ shows a typical behavior of the Kondo system, where the onset of the local-moment screening due to the coupling between conduction and localized Pr-4f electrons is observed below 50 K. $1/T_1$ shows a sharp decrease below T_A , but in small magnetic field less than 10 kOe, $1/T_1$ stays constant far below T_A with a relatively large value. Temperature and field dependence of $1/T_1$ reveals the presence of low-energy spin fluctuations in low-temperature and low-field region. These unconventional magnetic fluctuations are suggested to be quantum fluctuations from temperature independent behavior of $1/T_1$. These results imply that, although the static magnetism is not observed in zero magnetic field, the low-energy magnetic fluctuations which is regarded as quasi-static magnetism exist in $PrFe_4P_{12}$. Far above critical field of AFQ ordering H_A , the Korringa behavior $T_1T = const.$ is observed below 2 K in 100 kOe. The Korringa value below 2 K is one order of magnitude larger than that in LaFe₄P₁₂ without 4f electrons. These results show that PrFe₄P₁₂ is in the heavy-Fermion state far above H_A .

In this presentation, the magnetic fluctuations in the paramagnetic state of $PrFe_4P_{12}$ will be also discussed from the comparison with those in superconducting $PrOs_4Sb_{12}$ as well as in typical heavy-fermion compounds observed in Ce and Yb systems.

[1] K. Iwasa, Y. Watanabe, K. Kuwahara, M. Kohgi, H. Sugawara, T. D. Matsuda, Y. Aoki, and H. Aoki. Physica B **312-313**, 834 (2002).