Hyperfine-Enhanced Nuclear Spin Order of $PrPb_3$

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Abstract

The cubic compound $PrPb_3$ with the $AuCu_3$-type structure is known to undergo an antiferroquadrupolar phase transition at 0.4K. The ground state of the trivalent Pr ion in a cubic local symmetry is a nonmagnetic doublet Γ_3. This Γ_3 doublet splits into two singlet below 0.4K. We can expect that the hyperfine-enhanced nuclear moment will be differnet among the adjacent sites. We observed another phase transition at about 30 mK, which might be the anti-ferro to ferro quadrupolar transion.We will report the hyperfine-enhanced nuclear spin order in $PrPb_3$, which can be expected at about 1mK.

Key words: nuclear spin order; electronic orbital ordering; hyperfine-enhanced nuclear spin

1. Introduction

In the cubic compound $PrPb_3$ with $AuCu_3$-type structure, the ninefold degenerate ground state $^3H_4$ split into one singlet (Γ_1), one doublet (Γ_3) and two triplets (Γ4, Γ_5). The ground state of Pr^{3+}(J=4) is deduced to be the nonmagnetic Γ_3 doublet state with Γ_4 state at 19.4K, Γ_5 state at 28.5K and Γ_1 state at 46.7K [1] The wave functions of the Γ_3 are expressed as $|E_1>$=$(7/12)^2|4S>$-$(5/12)^2|0>$, $|E_2>=|2S>$. Here the nomenculture $|4S>$, etc. represents the symmetric conbinations $2^{-1/2}[|4>+|-4>|]$.

At about 0.4K, $PrPb_3$ exhibits a peak of the specific heat, which suggests the phase transition at this temperature. Since the magnetic reflection in the elastic neutron scattering experiment cannot be observed, the phase transition must be due to the Jahn-Teller type structural transition or the quadrupolar ordering. From the nonlinear magnetic susceptibility measurements and the elastic constant measurements, the phase transition might be originated in the quadrupolar ordering, especially antiferro-quadrupolar (AFQ) ordering. With a nonmagnetic Γ_3 doublet ground state, $PrPb_3$ shows the temperature independent Van Vleck magnetic susceptibility below about several K. Considering with the hyperfine interaction, the nuclear magnetic moment must be enhanced with the close of induced electronic magnetic moment. For the ground state of the Γ_3 doublet, we obtained the enhancement factor 1+K_z=28. Here $K_z=A_J(g_J/g_I)\Sigma_i |<E|J_z|G>|^2/\Delta_i$,$\Delta_i$:energy difference between the ground stste and excited ststes. Below AFQ ordering temperature, the enhancement should be different in each sublattice site of AFQ ordered structure. Due to the quadrupole ordering, Γ_3 doublet states , split into $|E_1>$ ground state or $|E_2>$ ground state in either antiqudrupole ordering site. We are interested in the hyperfine-enhanced nuclear spin ordering in this AFQ ordered $PrPb_3$. For this experiment, the magnetic susceptibility measurements in very low magnetic field are essential. Unfortunately most of all $PrPb_3$ crystal shows the superconducting trace. We tried to dimnish the superconducting phase in $PrPb_3$ crystal. By anealing the as-grown crystal at 750°C, we suc-

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ceeded to dimnish the superconducting phases. By using this nonsuperconducting crystal, we measured the temperature dependence of the dc magnetization.

2. Experimental Results and Discussions

The samples were prepared by enclosing stoichiometric amounts of Pr and Pb metals in closed Ta crucibles with anagram gas of one atmosphere to prevent from the evaporation loss of the lead. The heating temperature was 1150K. The crystallographic structure of the compounds was verified by x-ray powder diffractometry. Small Pb reflections were observed in as-grown crystals. The magnetization measurements were carried out in small magneic field of 0.1 mT between 2 and 300K by using a SQUID magnetometer. As-grown crystal showed the superconducting diamagnetism below about 8 K. The superconducting diamagnetism prevents from the observation of the hyperfine-enhanced nuclear spin order of $PrPb_3$ by the ac susceptibility measurements in zero magnetic field. The annealing at 750○ C in vacuum for more than 4 days diminished almost completely the superconducting phase.

By using this nonsuperconducting single crystal, we measured the temperature dependence of the ac magnetic susceptibility and also the magnetization in the magnetic field of 6.21 mT between 8mK and 2 K. The ac and dc magnetic susceptibility were measured by using an ac impedance bridge using a SQUID magnetometer. Just below about 0.4K, a small abrupt increase of the ac magnetic susceptibility with decreasing temperature was observed. This might be corresponding to the antiferro qudrapolar ordering [2].

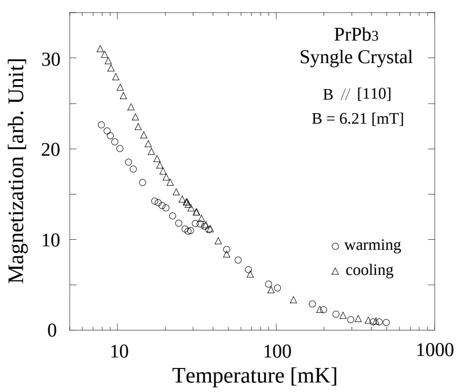

Fig. 1. Temperature depemdence of the magnetization in the magnetic field of 6.21 mT. Open circles and triangles represent the data obtained during warming and cooling the crystal respectively. The constant Van Vleck magnetic susceptibility was subtracted from the measured values.

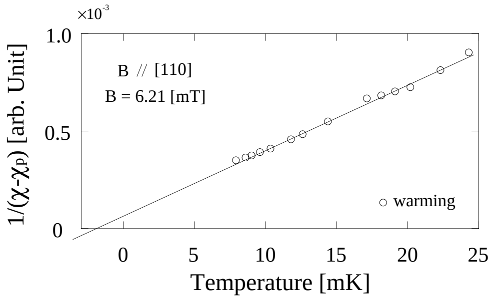

Fig. 2. The inverse of the static magnetic susceptibility in 6.21 mT against temperature. The asymptotic Curie temperature was -1.3±0.5 mK.

In the Fig.1 the temperature dependence of the static magnetizatin parallel to the [110] crystal direction is shown. In the figure, open circles and triangles represent the data measured with warming and cooling the crystal respectively. At about 30 mK, the static magnetization showed another anomally, suggesting another phase transition. Very recently Vollmer et al observed another phase transition at lower temperature below the AF quadrupolar transition in the magnetic fields between 1.5T and 4.5T[3]. They also suggested that this phase transition must be the first order. In our measurements, as shown in the figure, the large histeresis was observed at the lower transition temperature, suggesting the first order phase transition. Our measred anomally might be corresponding to this lower temperature phase transition. In nearly zero field, the transition temperature is so low. In Fig.2, the inverse of the static susceptibility (magnetization) is plotted against the temerature. From the figure, the asymptotic Curie temperaure was obtained to be -1.3 ±0.5 mK.

From these results the phase transition at about 30 mK is not the magnetic ordering. It can be another quadrupolar ordering, maybe from antiferro to ferro quadrupolar ordered state. Since in the lower temperature phase, the magnetization became smaller than the value in the higher temperature phase, the ground state in the lower temperature ferro quadrupolar oredered phase must be the Γ_{32}, which give rise to the smaller enhancement factor.

References

[1] W. Gross, K. Knorr, A. P. Murani, K. H. Buschow, Z. Physik **B37** (1980) 123.

[2] P. Morin, D. Schmitt, E. du Tremolet de Lacheisserie, J. Magn. Magn. Matter **30** (1982) 257.

[3] R. Vollmer, G. Goll, C. Pfleider, H.V.Löhneysen, M. B. Maple, P. C. canfield, Physica B **312-313** (2002) 855.