Magnetic anisotropy of the heavy fermion state in $PrFe_4P_{12}$

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Abstract

We report the magnetic field dependence of specific heat on single crystalline $PrFe_4P_{12}$ measured in fields applied along the three main symmetry directions $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$. The electronic thermal excitation in the high-field heavy-fermion state is found to be strongly enhanced for fields along the $\langle 111 \rangle$ direction.

Key words: heavy fermion behavior; filled skutterudites; $PrFe_4P_{12}$; specific heat

Recently it has been confirmed that a filled skutterudite $PrFe_4P_{12}$ has a rare $4f^2$-based heavy fermion (HF) state in high magnetic fields where a non-magnetic ordered state (ODS), appearing below $T_A = 6.5$ K in zero field, is suppressed. The HF state is evidenced by largely enhanced values of electronic specific-heat-coefficient ($\gamma \sim 1$ J/K^{2}mol) [1,2] and cyclotron mass in dHvA studies [3]. Non-magnetic nature of the ODS has been revealed by nuclear scattering and ^{141}Pr-nuclear specific heat studies [4,5]. A possible order parameter of the ODS is an antiferro-quadrupole (AFQ) with $\mathbf{q} = [1, 0, 0]$ [6], which couples to the observed lattice distortion [7] and consequently to the expected Fermi surface instability [8]. In this scenario, a gap opening, which is suggested from the electrical resistivity $\rho(T)$ and Hall coefficient $R_H(T)$ [9], and a field-induced staggered magnetic component observed in neutron scattering experiments [10] can be naturally understood. This scenario might point to a possibility that fluctuations of the quadrupole moments are essential for the HF behavior in the high-field HF state.

In this paper, we report further detailed specific heat measurements in applied magnetic fields along the three main symmetry directions $\langle 100 \rangle$ (easy direction), $\langle 110 \rangle$, and $\langle 111 \rangle$ (hard direction). The results reveal that the electronic thermal excitation in the high-field HF state has a strong magnetic anisotropy. For all the measurements, we used the same single crystal grown by Sn-flux method (see Ref. [9]). The observation of dHvA signals on samples grown in the same batch confirms high-quality of the present sample. Specific heat $C(H,T)$ is measured by a quasiadiabatic heat pulse method described in Ref. [11] using a dilution refrigerator equipped with an 8T superconducting magnet.

Figure 1 (a) shows the magnetic field dependences of C/T at $T = 1.01$ K in applied magnetic fields along the $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$ crystallographic directions. At this temperature, the phonon contribution of the order of 10^{-4} J/K^{2}mol estimated from the specific heat data for $LaFe_4P_{12}$ [12] is negligibly small. The phase boundary of the ODS is magnetically anisotropic; the transition field $H_A = 4.0, 5.4$, and 6.5 T for $H \parallel \langle 100 \rangle, H \parallel \langle 110 \rangle$, and $H \parallel \langle 111 \rangle$. In the ODS ($H < H_A$), C/T is suppressed and weakly field dependent except for fields close to H_A; $C/T = 0.18$ J/K^{2}mol in zero field and gradually decreases with increasing field showing a shallow minimum around $2 \sim 4$ T. At $H = H_A$, a sharp jump in C/T is observed, reflecting the first-order nature of the phase transition. Because of relatively large step of the field change ($\Delta H = 0.1$ T) in the field scan measurements, sharp peak structure associated with a release of the latent heat at the phase transition was undetectable, while in the temperature-dependence measurement for $H \parallel \langle 100 \rangle$ a pronounced peak with a maximum of $C/T \simeq 25$ J/K^{2}mol at 1.31

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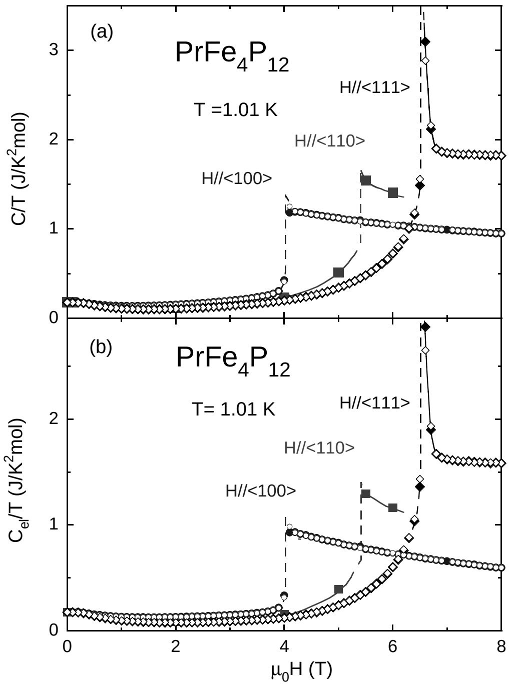

Fig. 1. (a) Magnetic field $\mu_0 H$ dependences of specific heat divided by temperature C/T in applied fields along the three main symmetry directions. The data of closed (open) symbol were obtained with increasing (decreasing) H; no appreciable hysteretic behavior is visible. The data for $H \parallel \langle 100 \rangle$ and $H \parallel \langle 110 \rangle$ are partly from Ref. [2] and Ref. [1], respectively. (b) Estimated electronic specific heat divided by temperature $C_{\rm el}/T$ as a function of $\mu_0 H$. The lines are guide to the eye.

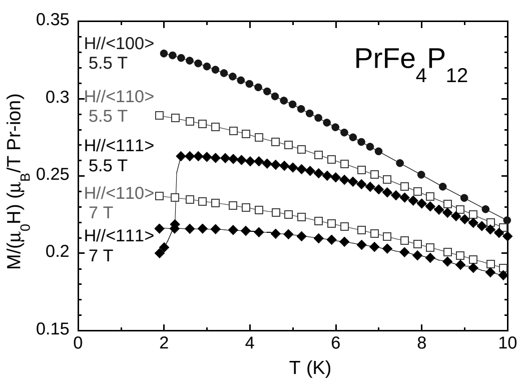

Fig. 2. Temperature dependence of magnetization divided by applied magnetic field $M/(\mu_0 H)$.

K was observed [2]. Remarkable feature in $H > H_{\rm A}$ is the substantial difference in C/T among the applied field directions.

For quantitative analyses of the electronic contribution, the nuclear contribution $C_{\rm n}$ mostly caused by ^{141}Pr nuclei (nuclear spin $I = 5/2$ for ^{141}Pr with the natural abundance of 100%) should be subtracted. We have experimentally demonstrated in Ref. [2] that the size of the Pr magnetic moment and $C_{\rm n}$ are coupled through the strong intrasite hyperfine interaction, leading to the largely enhanced Pr nuclear Schottky contribution. Using the coupling constant determined in Ref. [2], $C_{\rm n}$ can be estimated by bulk magnetization M measurements. An error of $\sim$3% in M, inevitably caused by an extrapolation to low temperatures since the present M data are limited for $T > 1.9$ K, leads to negligibly small error in $C_{\rm el}/T$.

The resulting $C_{\rm el}/T$ vs H curves are shown in Fig. 1 (b). The data in the HF state are substantially anisotropic. In 8 T, $C_{\rm el}/T$ for $H \parallel \langle 111 \rangle$ is about three times larger than for $H \parallel \langle 100 \rangle$. With increasing field, $C_{\rm el}/T$ is largely suppressed for $H \parallel \langle 100 \rangle$ but not for $H \parallel \langle 111 \rangle$. The $C_{\rm el}/T$-vs-H data are connected to the temperature dependence of M by the Maxwell relation

$$[\partial(C_{\rm el}/T)/\partial(\mu_0 H)]_T = [\partial^2 M/\partial T^2]_H. \qquad (1)$$

Qualitative agreement is found with the $M(T)$ data shown in Fig. 2; distinct $\partial^2 M/\partial T^2 < 0$ is observed for $H \parallel \langle 100 \rangle$ while M loses its temperature dependence at low temperatures for $H \parallel \langle 111 \rangle$. Therefore, the present observation suggests that the significantly-enhanced electronic thermal excitation observed in the HF state for $H \parallel \langle 111 \rangle$ (hard direction) might be of non-magnetic origin.

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