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Magnetic ordering in $PrFe_4As_{12}$

T.A. Sayles[a,*], W.M. Yuhasz[a], J. Paglione[a], T. Yanagisawa[a], J.R. Jeffries[a], M.B. Maple[a], Z. Henkie[b], A. Pietraszko[b], T. Cichorek[b], R. Wawryk[b], Y. Nemoto[c], T. Goto[c]

[a] *Department of Physics and Institute for Pure and Applied Physical Sciences, University of California at San Diego, La Jolla, CA 92093, USA*
[b] *Institute of Low Temperature and Structure Research, Polish Academy of Sciences, 50-950 Wroclaw, Poland*
[c] *Graduate School of Science and Technology, Niigata Univesity, Niigata 950-2181, Japan*

Abstract

Magnetization $M(H,T)$, specific heat $C(T)$, and electrical resistivity $\rho(T)$, measurements were performed on single crystals of the Pr-based filled skutterudite compound $PrFe_4As_{12}$. $M(H)$ isotherms indicate a saturation magnetization of $2.3\mu_B$/f.u., and Arrott analysis reveals a Curie temperature $T_C = 18.2$ K and saturated moment $M_{sat} = 2.0\mu_B$/f.u. Curie–Weiss analysis of magnetic susceptibility, χ, reveals two distinct regions: for $T > 100$ K, the effective moment $\mu_{eff} = 3.98\mu_B$ and the Curie–Weiss temperature $\theta_{CW} = 4.1$ K, while for $19.5 < T < 75$ K, $\mu_{eff} = 3.52\mu_B$ and $\theta_{CW} = 17.7$ K. $C(T)$ and $\rho(T)$ show distinct features at T_C, while the ac susceptibility exhibits lower temperature features, possibly associated with changes in magnetic structure.

Keywords: Ferromagnetism; Skutterudites; Praseodymium

The interplay between crystalline electric field effects and strong hybridization of the Pr-4f localized state with conduction electron states leads to a variety of strongly correlated electron phenomena in the Pr-based filled skutterudite compounds, including: unconventional superconductivity, heavy fermion behavior, and antiferroquadrupolar order in $PrOs_4Sb_{12}$ [1–3] and multiple ordered phases, one of which is antiferromagnetic, in $PrOs_4As_{12}$ [4,5]. Presented here are thermodynamic and transport measurements on single crystals of the Pr-based filled skutterudite arsenide $PrFe_4As_{12}$.

Fig. 1 shows $M(H)$ isotherms for fields oriented along the [1 1 1] and [1 0 0] directions at 2 K. A change of the easy axis from the [1 0 0] to the [1 1 1] directions at $H \sim 0.6$ T can be seen as a change of the maxima in M along the two directions. Additional measurements of $M(H)$ at higher temperatures, $T > 10$ K, show no change in the easy-axis with field. Arrott plot analysis of $M(H)$ isotherms yields a Curie temperature $T_C = 18.2$ K.

The lower panel in Fig. 2 shows ZFC and FC dc magnetization, $M(T)$, of $PrFe_4As_{12}$ for an applied field of 5 mT as well as the imaginary part of ac susceptibility, $\chi''_{ac}(T)$, for three driving frequencies: 10, 100, and 500 Hz, respectively, with an ac field of $H_{ac} = 0.1$ mT. High temperature, $70 \leqslant T \leqslant 300$ K, Curie–Weiss fits of $\chi^{-1}_{dc}(T)$ (not shown) reveal a Curie–Weiss temperature, $\theta_{CW} = 4.1$ K, and an effective moment $\mu_{eff} = 3.98\ \mu_B$, whereas low temperature fits indicate $\theta_{CW} = 17.7$ K, consistent with the Curie temperature derived from the Arrott analysis, and $\mu_{eff} = 3.52\mu_B$. The measured saturation magnetization is larger than that expected for Pr^{3+} based on Hund's rules. However, the excess can be attributed to the spin-only moment of Fe for the low-spin configuration. The midpoint of the broadened step in $\chi''_{ac}(T)$ shown in Fig. 2 is found to be $T_C = 17.8$ K. The frequency dependence of $\chi''_{ac}(T)$ shown in Fig. 2 is possibly due to spin fluctuations related to the change in the magnetic easy-axis displayed in the 2 K $M(H)$ isotherm seen in Fig. 1. Two other distinct features are also evident in $\chi''_{ac}(T)$: at 11.7 K, there is a maximum indicative of a possible second-phase transition, while at 7.5 K there is a large peak with some frequency

*Corresponding author. Tel.: +1 858 534 2493; fax: +1 858 534 1241.
E-mail address: tsayles@physics.ucsd.edu (T.A. Sayles).

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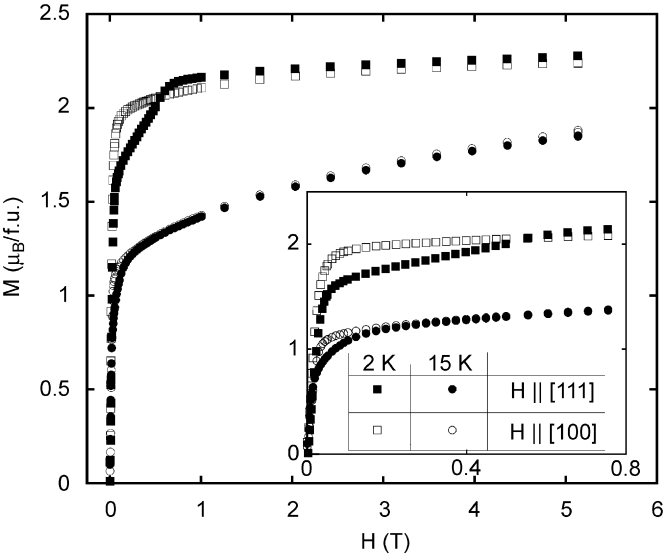

Fig. 1. $M(H)$ oriented along the [1 1 1] and [1 0 0], directions at 2 and 15 K, with the inset showing a close up of the low field data.

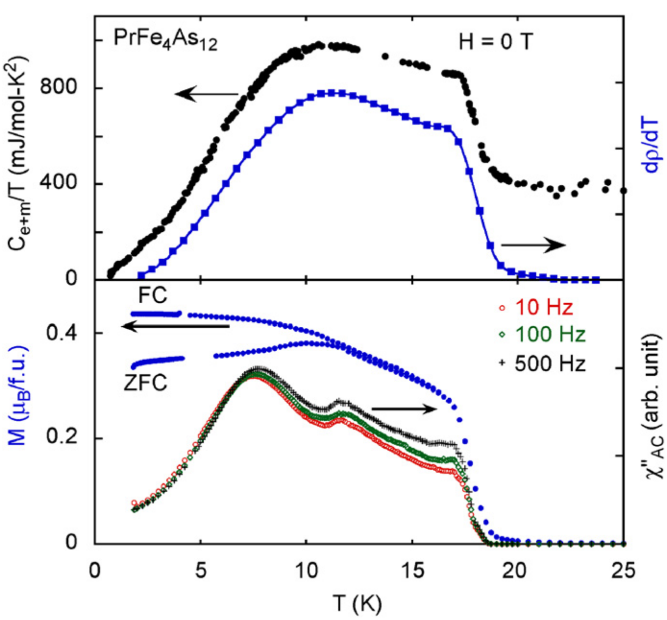

Fig. 2. Zero-field C_{e+m}/T, $d\rho/dT$, (top panel) M_{dc}, and χ''_{ac} (bottom panel) as a function of temperature, T.

dependence, which seems to be related to the change in easy-axis observed in the $M(H)$ isotherms.

The upper panel in Fig. 2 shows the magnetic and electronic contributions, $C_{e+m}(T)/T$, of $C(T)/T$ after the lattice and a low-temperature nuclear portion were removed. The lattice contribution was estimated from a Debye analysis, $C/T = \gamma + \beta T^2$ (where γ is the electronic specific heat coefficient, found to be 340 mJ/mol K^2, and β determines the lattice contribution, as well as the Debye temperature, θ_D, which is calculated to be 356 K), while the nuclear portion was analyzed using a high-temperature expansion of a Schottky anomaly—$C_{Schottky} \sim A/T^2$. Also plotted in the upper panel in Fig. 2 is $d\rho(T)/dT$. In both plots, a clear step is seen with the midpoint near 18 K indicating the onset of magnetic ordering. Additionally, a broad, Schottky like, maximum is seen in both data sets near 10.5 K. Interestingly, both plots have remarkable similarities, a possible reason being, the resistivity is dominated by magnetic scattering for temperatures below T_C.

$PrFe_4As_{12}$ has a clear transition to a ferromagnetic state at $T_C = 18$ K, followed by a possible structural change at 11.7 K, and finally a change in the easy-axis at $T = 7.5$ K. Whether the structural change drives the change in easy axis remains to be determined.

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