Transport and Magnetic properties of PrCoIn$_5$

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Structural, electrical and magnetic measurements of 115 single crystals of PrIn$_5$ are reported. It has a tetragonal structure and has slightly lower cell volume than its isomorphic counter part CeCoIn$_5$. The resistivity saturates for $T \geq 10K$. Analysis of the resistivity for $10K < T < 60K$ indicates a regular fermi liquid behavior. It does not exhibit superconductivity down to $T \sim 1K$. The magnetic susceptibility analysis yielded the moment to be $4.00\mu_B$ indicating that the magnetism of PrCoIn$_5$ is dominated by Pr$^{3+}$ free ions with some admixture of the magnetic moment of the Co sublattice. The paramagnetic Curie temperature $\theta \sim -40K$. At low temperatures the susceptibility follows a broad maximum around $T_N \sim 14.5K$, and increases as the temperature is lowered. The disappearance of superconductivity for $T > 1K$ is attributed to chemical pressure effects and magnetic pair breaking.

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I. INTRODUCTION

A typical metal is expected to exhibit fermi liquid behavior in that at low temperatures the electronic specific heat coefficient, $\gamma$ and the magnetic susceptibility, $\chi$ are constants, and the resistivity, $\rho = \rho_0 + AT^2$. Instead the normal state properties of CeCoIn$_5$ are characterized by a non fermi liquid behavior where $C/T \sim \ln T$ and $\rho = \rho_0 + AT^n$ ($n < 2$) and $\chi \sim T^{-n}$ ($n < 1$). Its superconducting transition temperature $T_c \sim 2.3K$ is significantly influenced by application of hydrostatic pressure and chemical substitution. In the system CeMIn$_5$ ($M=Co, Rh, Ir$), it has been shown that the superconductivity can be induced by tuning Neel temperature, $T_N$ to zero. Substitution of In with Sn[1] and Ce with La[2] produced pair breaking effects attributed to chemical pressure[3]. In this work we synthesized good quality PrCoIn$_5$ single crystals to study its structure, thermal and magnetic properties. The structurally similar PrCoIn$_5$ is non superconducting, its resistivity saturates below 10K. These differences in properties may allow researchers to study the evolution from superconducting non fermi liquid ground state exhibited by CeCoIn$_5$ to a non superconducting fermi liquid state exhibited by PrCoIn$_5$. Single crystals of PrCoIn$_5$ were synthesized from indium flux by combining stoichiometric amounts of Pr and Co with excess indium. The charge is put inside an alumina crucible, which is sealed in an evacuated quartz cylinder. It is then heated to 1150 C at the rate of 2 C per minute, followed by cooling to 750 C at 3C/minute, rapid cooling to 450C at 2C/minute. The charge is then removed, and quickly put in centrifuge to remove the excess flux. Each good crystal is etched with concentrated hydrochloric acid for several hours and then it is rinsed with ethanol in ultrasonic environment. The resulting crystals are mostly plates with dimensions $2mm \times 2mm \times 1mm$.

FIG. 1: x-ray powder diffraction patterns of PrCoIn$_5$

The lattice parameters were obtained from single crystal x-ray diffraction pattern with Mo-K$_\alpha$ radiation. The x-ray intensity as a function of $2\theta$ is shwon in Figure 1 above. The reflections were indexed for the tetragonal unit cell. Shown on Table 1 are the lattice parameters and the cell volumes of PrCoIn$_5$ and CeCoIn$_5$. The $c/a \sim 1.62$ clearly indicates that the structure of PrCoIn$_5$ is tetragonal and it is structurally isomorphic with CeCoIn$_5$.

II. RESISTIVITY OF PRCoIN$_5$

Resistivity was measured using a standard 4- wire technique at temperatures ranging from 300 K to 1.8 K in helium cooled cryostat. Excitation currents of

| Sample | $a(\text{Å})$ | $b(\text{Å})$ | $c(\text{Å})$ | $c/a$ | Cell volume ($\text{Å}^3$) |
|--------|--------------|--------------|--------------|-------|-----------------|
| PrCoIn$_5$ | 4.61 | 4.61 | 7.50 | 1.62 | 161.09 |
| CeCoIn$_5$ | 4.62 | 4.62 | 7.56 | 1.63 | 161.36 |

TABLE I: Lattice parameters of PrCoIn$_5$
500\mu A – 1000\mu A were applied parallel to the c-axis at frequencies of 16 Hz. Each sample was mounted on a resistivity puck with GE varnish. \#40 gauge copper wires were attached to the samples with silver epoxy. The magnetic susceptibility and magnetization measurements were carried out with a PPMS system at NHMFL-Los Alamos.

As shown in Figure 2 the resistivity shows metallic behavior above about 150K. Since for typical metals such as copper, the deviation from linearity begins at \( T \sim 0.7\theta_D \). We estimated \( \theta_D \sim 241 \) K. Below 150K the data follow a broad maximum near \( T \sim 72 \) K accompanied with slow decrease as the temperature is lowered. The broad maximum is attributed to crystalline electric field effects. There is no evidence of superconductivity within the accessible temperature \( T > 1K \). Shown in Figure 3 is our fit of the data for \( 10K < T < 60K \) using a polynomial of order five. Assuming that the contribution of the higher order terms diminishes as the temperature is lowered, we retained the fit parameters, \( \alpha = 0.034\mu\Omega\cdot\text{cm}, \beta = -0.005\mu\Omega - cm/K \) and \( A = 4.94 \times 10^{-4}\mu\Omega - cm/K^2 \). While the negative linear term can be an artifact of the fit, the \( T^2 \) term is suggestive of a normal fermi liquid behavior. Extrapolated from the high temperature linear component we obtain the residual resistivity \( \rho_0 \sim 0.021\mu\Omega - cm \) and the residual resistivity ratio (\( RRR = \rho_H/\rho_0 \)) was 200. This ratio indicates that this crystal is of reasonable quality. As shown in Figure 4 the resistivity saturates for \( T < 10K \) with \( \rho_{sat} \sim 0.034\mu\Omega - cm \). \( \rho_{sat} \) is lower than the residual resistivity, \( \rho_0 \). Whether or not the origin of this saturation is of fundamental nature or caused by inclusions and surface effects is to be determined.

III. MAGNETIC SUSCEPTIBILITY OF PrCoIn_5

Shown in Figure 5 is the magnetic susceptibility of PrCoIn_5 measured at 1kOe. The data showed a rapid upturn at low temperature. The inset shows the susceptibility below 30K, the broad maximum near \( T_N \sim 14.5K \). This structure coincides with the resistivity saturation for \( T < 10K \) and we believe it is associated with antiferromagnetic ordering of the Pr ions. While similar behaviour has observed in other Pr based alloys such as PrBa_2Cu_3O_7, a non superconducting cuprate[4] and Pr_3In[5] the presence of multiphases cannot be ruled out. Shown in Figure 6 is the plot of the inverse susceptibility.

| Table II: Physical parameters of PrCoIn_5 |
|------------------|------------------|------------------|
| PrCoIn_5         | \( T_K(K) \)     | \( T_c(K) \)     |
|                  | 72               | 0                |
|                  | \( T_N(K) \)     | \( \theta(K) \)  |
|                  | 14.5K            | -40              |
|                  | \( \mu(\mu_B) \) | 4.00             |
$\chi^{-1}$ versus temperature.

At high temperatures ($T \geq 100K$), $\chi^{-1}$ is linear and follows Curie-Weiss behavior. We obtained effective moment of $4\mu_B$. This value is larger than the $3.73\mu_B$ that is expected for a free Pr$^{3+}$. The higher magnetic moment could be attributed to an admixture of moments from both Co ions and Pr$^{3+}$. The Co sub lattice can be magnetized because of f-d exchange as in $RCO_3$ [6]. Obviously the moment is dominated by Pr$^{3+}$ in the J = 4 Hunds rule ground state indicating that the 4f electrons are almost localized within the Pr atoms. Similar results are obtained by others except that our analysis shows a lower paramagnetic Curie temperature $\theta \sim -40K$ compared to $-56K$ [7].

In summary we prepared clean single crystals of PrCoIn$_5$. The structure is tetragonal it has a $c/a \sim 1.62$ slightly smaller volume than its counterpart CeCoIn$_5$. Superconductivity is not observed down to 1K. In addition the behavior of the low temperature resistivity appears to have originated from a fermi liquid behavior. Pair breaking phenomenon were observed when CeCoIn$_5$ is alloyed by the non magnetic La further demonstrating unconventional superconductivity in this system [2]. PrCo$_5$ has slightly lower ($C/a \sim 1.62$) than that of CeCoIn$_5$ ($C/a \sim 1.63$). Whether or not this difference has such detrimental effect on superconductivity is not determined. However the high value of the magnetic moment and the presumed magnetic order below $T_N \sim 14.5K$ may have led to magnetic pair breaking.

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