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Ferromagnetic Kondo lattice behavior in $Ce_{11}Pd_4In_9$

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ABSTRACT

We report on the low-temperature physical properties of a novel compound $Ce_{11}Pd_4In_9$ that crystallizes with the orthorhombic $Nd_{11}Pd_4In_9$-type crystal structure (space group *Cmmm*). The compound exhibits ferromagnetic ordering at $T_C = 18.6$ K and an order-order transition at $T_t \approx 1.6$ K, as inferred from the low-temperature magnetic susceptibility, heat capacity and electrical resistivity data. In the paramagnetic region, the electrical transport in $Ce_{11}Pd_4In_9$ is dominated by Kondo effect. Below T_C, a distinct contribution due to ferromagnetic spin waves dominates the electrical resistivity data, while at the lowest temperatures, the electrical transport and thermodynamic properties are governed by strong electron-electron correlations. The features observed conjointly hint at strongly correlated ground state in $Ce_{11}Pd_4In_9$.

1. Introduction

For past few decades, investigation of cerium-based intermetallic compounds has been at the forefront of condensed matter research as it encompasses a large variety of interesting phenomena which include Kondo effect, heavy-fermion (HF) behavior, non-Fermi liquid (NFL) features, magnetic ordering, superconductivity, quantum criticality, etc. [1–5]. Hybridization between 4*f* localized electrons and conduction electrons plays a crucial role in governing the ground state properties in such materials. Of particular interest are Kondo lattice systems which exhibit magnetic ordering, as in many of them one can tune the magnetic exchange interactions by external parameter, like pressure, magnetic field or doping, to explore the possibility of suppressing the magnetic order down to absolute zero temperature at a quantum critical point (QCP). Since a few years, quantum criticality in HF systems has been one of the hot topics in condensed matter physics, owing to unusual phenomena observed in the vicinity of QCP, such as unconventional superconductivity and NFL behavior [2,5,6]. Unlike antiferromagnetic (AFM) dense Kondo systems, examples of ferromagnetic (FM) Kondo lattices are quite rare in the existing literature. Representatives of the latter group, discovered in recent years, are the FM compounds CeRuPO [7], Ce_3RhSi_3 [8], $CePd_2Al_8$ [9] and $CeCrGe_3$ [10,11].

Another subject that recently is attracting much research interest concerns Ce-based Kondo lattices that host more than one inequivalent Ce site in their crystallographic unit cell. This structural feature may bring about complex physical properties, like separate AFM order in two Ce atom sublattices in $Ce_5Ni_6In_{11}$ [12], dipolar and quadrupolar AFM orderings in $Ce_3Pd_{20}Si_6$, each associated with different Kondo ion site [13,14], or coexistence of AFM and HF superconductivity in Ce_3PtIn_{11}, where the two cooperative phenomena likely emerge in two distinct Ce atom sublattices [15].

Within the two frameworks outlined above, search for novel Ce-based intermetallics with multiple Ce sites in their crystal structure, which exhibit Kondo effect and possibly order ferromagnetically at low temperatures appears to be of particular interest. Recently, a series of rare-earth (RE) ternaries with the chemical formula $RE_{11}T_4In_9$ (T = *d*-electron transition metal) has been reported to form with orthorhombic crystal structure of the $Nd_{11}Pd_4In_9$-type (space group *Cmmm*) that hosts as many as five independent RE sites in the unit cell [16–19]. Amidst these compounds, two Ce-based materials were found. Most interestingly, $Ce_{11}Ni_4In_9$ was established to order ferromagnetically below $T_C = 16.5$ K and show an order-order phase transition at $T_t = 5$ K [20], while $Ce_{11}Ru_4In_9$ was characterized as a ferrimagnet with $T_C = 6.3$ K and weak Kondo effect [21]. Motivated by these findings, we successfully synthesized another compound from this family, namely $Ce_{11}Pd_4In_9$, the existence of which was briefly communicated in Ref. 18. In this paper, we report on the low temperature physical properties of the novel phase, studied by means of magnetic, electrical resistivity and heat capacity measurements. Remarkably, the compound was found to be a moderate HF system that orders ferromagnetically below $T_C = 18.6$ K and exhibits another magnetic phase transition in the ordered state.

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2. Experimental

Polycrystalline sample of $Ce_{11}Pd_4In_9$ was synthesized by arc-melting elemental constituents (purities: Ce-3 N, Pd-4 N, and In-6 N) on a water-cooled copper hearth in an arc furnace installed inside a glove-box filled with ultra-pure argon gas with continuously controlled partial pressures of O_2 and H_2O to be lower than 1 ppm. The ingot was flipped over and remelted several times to ensure homogeneity. The weight loss after the final melting was negligible ($< 0.4\%$). Annealing at 700 °C for two weeks resulted in producing microcracks in the sample that preserved the same crystal structure. Thus, we restricted our measurements only to the as-cast material. As a nonmagnetic reference compound, $La_{11}Pd_4In_9$ (La metal purity: 3 N) was also synthesized following the same process.

Quality of the obtained alloys was checked by x-ray powder diffraction (XRD) at room temperature on an X'pert Pro PanAnalytical diffractometer using Cu-K_α radiation. The XRD data confirmed that $Ce_{11}Pd_4In_9$ and $La_{11}Pd_4In_9$ crystallize with the orthorhombic $Nd_{11}Pd_4In_9$-type crystal structure, and the obtained lattice parameters were in agreement with the literature data [18]. Chemical composition of the samples was examined by energy dispersive x-ray (EDX) analysis using a FEI scanning electron microscope equipped with an EDAX Genesis XM4 spectrometer. The results indicated that the obtained polycrystalline samples are homogeneous and single-phase materials with the stoichiometry close to the nominal one.

Magnetic measurements were performed in the temperature range 1.72–300 K using a Quantum Design SQUID magnetometer. The electrical resistivity was measured over the temperature range 0.38–300 K employing standard ac four-probe technique implemented in a Quantum Design PPMS platform. Specific heat measurements were carried out from 0.4 to 300 K using relaxation method available in the same PPMS platform.

3. Results

3.1. Magnetic properties

The results of magnetic measurements performed on $Ce_{11}Pd_4In_9$ are summarized in Fig. 1. Above about 150 K, the reciprocal magnetic susceptibility follows the Curie-Weiss law with the effective magnetic moment $\mu_{eff} = 2.62(5)\ \mu_B$ per Ce atom and the paramagnetic Curie temperature $\theta_p = -9.0(2)$ K. The experimental value of μ_{eff} is close to the theoretical prediction for free trivalent cerium ion (2.58 μ_B). Its magnitude, calculated by averaging the separate contributions from five independent magnetic sublattices, suggests similar valence state of each Ce ion in the unit cell of $Ce_{11}Pd_4In_9$. In turn, the fairly large negative value of θ_p hints at predominance of antiferromagnetic correlations in the compound that orders ferromagnetically (see below). It seems likely that θ_p reflects significant Kondo interactions resulting in antiferromagnetic correlations in the system, alike in ferromagnetic $CePd_2Al_8$ [9], $CeIr_2B_2$ [22] and $CeTiGe_3$ [23]. Below 150 K, the $\chi^{-1}(T)$ dependence markedly deviates from a straight-line behavior evidencing strong crystalline electric field (CEF) effect.

As shown in the inset to Fig. 1, the low-temperature dependence of the magnetization in $Ce_{11}Pd_4In_9$, measured in an external magnetic field of 1 kOe (upon cooling the specimen in the applied field) exhibits a behavior characteristic of ferromagnets. The Curie temperature, defined as an inflection point on the $M(T)$ curve, equals $T_C = 18.6(4)$ K. With decreasing temperature in the ordered state, the magnetization saturates at a value of 13.7 emu/g that corresponds to the magnetic moment of 0.67(3) μ_B per Ce ion, calculated assuming equal contribution from each Ce site.

The magnetization measured at $T = 1.72$ K as a function of applied magnetic field strength (see Fig. 2) corroborates the ferromagnetic nature of the electronic ground state in $Ce_{11}Pd_4In_9$. Remarkably, in weak magnetic fields, the magnetization rapidly increases with rising field reaching about 13 emu/g in $H = 1$ kOe, and then almost saturates at this value up to 4 kOe (the inset to Fig. 2). In stronger fields, the magnetization increases gradually with a clear tendency for saturation. In the limiting field of 70 kOe, it achieves a value of about 21 emu/g yielding the average magnetic moment of 1.02(3) μ_B per Ce site. This value is only a fraction of that expected for a free trivalent Ce ion ($gJ = 2.15\ \mu_B$), and must be attributed to the doublet ground state in the CEF split $^2F_{5/2}$ multiplet. In parallel, some reduction of the ordered magnetic moment due to the Kondo interactions can be also expected (see below). As can be inferred from the inset to Fig. 2, the characteristic feature of ferromagnetic $Ce_{11}Pd_4In_9$ is a large remanence (~60%) combined with a fairly narrow hysteresis loop, roughly limited to magnetic fields $H < 1$ kOe at $T = 1.72$ K.

In order to gain better understanding of the magnetic state in $Ce_{11}Pd_4In_9$, magnetization measurements were performed in a weak magnetic field of 20 Oe, upon cooling the sample in zero (ZFC: zero field cooling) and applied (FC: field cooling) field. As can be inferred from Fig. 3, the obtained $M(T)$ data bifurcates at a temperature slightly lower than T_C, as expected for ferromagnets. However, worth noting is an unusual shape of the FC curve, which deviates at the bifurcation

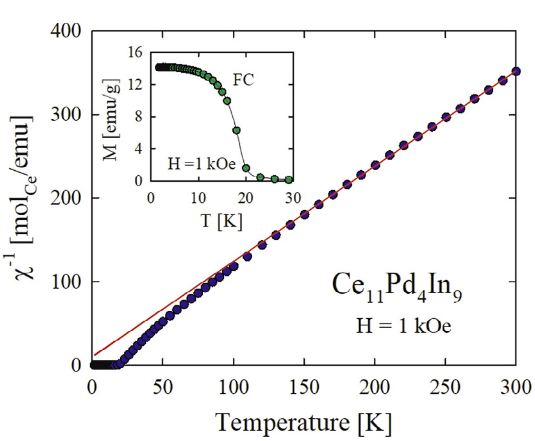

Fig. 1. Temperature dependence of the inverse molar magnetic susceptibility of $Ce_{11}Pd_4In_{11}$ measured in an applied magnetic field of 1 kOe. The solid red line represents the Curie Weiss fit discussed in the text. Inset: low-temperature variation of the magnetization in $Ce_{11}Pd_4In_{11}$ taken in a field of 1 kOe, upon cooling the sample in the applied field. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

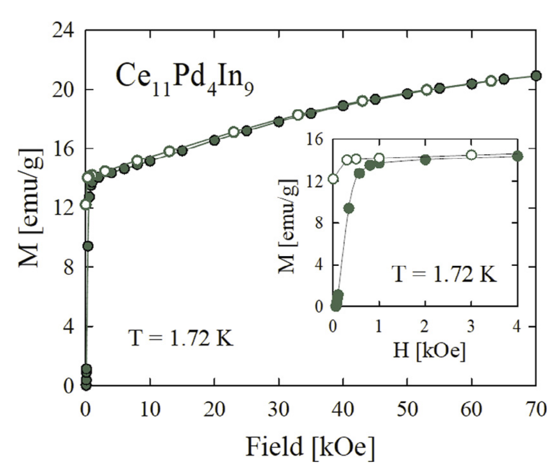

Fig. 2. Magnetic field variation of the magnetization in $Ce_{11}Pd_4In_{11}$ measured at 1.7 K with increasing (full circles) and decreasing (open circles) field strength. Inset: (in a magnified scale). Inset: zoom-in of the isothermal magnetization data up to 4 kOe.