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Non-Fermi-Liquid State in $URu_{0.68}Pd_{0.32}Ge$

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We report the measurements of dc-magnetization (M), specific heat (C_p) and electrical resistivity (ρ) of an intermetallic $URu_{0.68}Pd_{0.32}Ge$, that lies at the border between nonmagnetic and magnetic regimes in the magnetic phase diagram of solid solutions $URu_{1-x}Pd_xGe$. The studied composition shows enhanced dc-magnetic susceptibility $\chi(T)$ and the Sommerfeld ratio $C_p(T)/T$ at low temperatures. Below 4 K $\chi(T)$, $C_p(T)/T$ and $\rho(T)$ can be described by $\chi(T) \propto T^{-0.48}$, $C_p(T)/T \propto \sqrt{T}$ and $\rho(T) \propto c\ln(T) + AT^{3/2}$, respectively. These observations provide an evidence that $URu_{0.68}Pd_{0.32}Ge$ is a moderately heavy-fermion system with an electronic ground state of non-Fermi liquid character. We found non-linear effect of the magnetization and a large value of the Wilson ratio, which are consistent with the interpretation in terms of magnetic instability.

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1. Introduction

Amongst equiatomic ternaries UTX (T = d-electron transition metal, X = Si or Ge), crystallizing in the TiNiSi-type structure, URuGe is a nonmagnetic, whereas UPdGe undergoes two successive magnetic phase transitions: an antiferromagnetic at 50 K and a ferromagnetic at 30 K [1, 2]. In order to investigate magnetic phase diagram as well as to search for a quantum critical point we have synthesized and performed measurements of X-ray diffraction, dc-magnetization M, ac-susceptibility $\chi_{ac}(T)$, specific heat $C_p(T)$ and electrical resistivity $\rho(T)$ of solid solutions $URu_{1-x}Pd_xGe$ [3]. We found that the alloys crystallize in the orthorhombic TiNiSi-type structure (space group $Pnma$). The measurements of the physical properties have revealed that the compositions with $x \leq 0.32$ are nonmagnetic down to 2 K, while these with $0.35 \leq x \leq 0.8$ are antiferromagnetic. It was found that for $x < 0.5$ the Néel temperature T_N varies with concentration as $T_N \propto (x - 0.32)^{2/3}$ and it reaches the maximum value of 32 K at $x = 0.8$. The alloy $x = 0.9$, like UPdGe, manifests two magnetic phase transitions: antiferromagnetic at $T_N = 20$ K and ferromagnetic-like at $T_C = 30$ K. In this contribution we will focus on the low-temperature properties of the $x = 0.32$ alloy. Since this composition is placed at the border between nonmagnetic and magnetic regimes, we anticipate interesting phenomena caused by magnetic instability, including e.g., non-Fermi liquid behaviour or quantum criticality.

2. Experimental details

Sample $URu_{0.68}Pd_{0.32}Ge$ was prepared and characterized with the same procedure reported previously [3]. X-ray diffraction analysis indicated the sample to be a single phase with the room temperature lattice parameters $a = 0.619$ nm, $b = 0.4357$ nm and $c = 0.7548$ nm. Measurements of magnetization, specific heat and electrical resistivity in this study were conducted in the temperature range 0.4–400 K and in magnetic fields up to 9 T, using a Quantum Design MPMS magnetometer and PPMS platform.

3. Results and discussions

In Fig. 1a we show the temperature dependence of the inverse magnetic susceptibility, $\chi(T)^{-1} = H/M(T)$, of $URu_{0.68}Pd_{0.32}Ge$ measured at a magnetic field of $\mu_0 H = 0.5$ T. In the temperature range 70–400 K, the susceptibility follows a modified Curie–Weiss law:

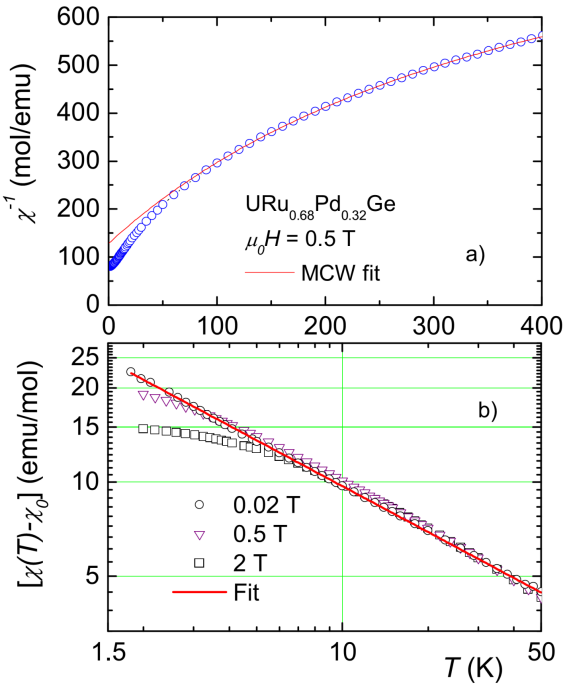

Fig. 1. (a) Temperature dependence of the inverse dc-susceptibility of $URu_{0.68}Pd_{0.32}Ge$ at 0.5 T. (b) A log-log plot of $[\chi(T) - \chi_0]$ vs. T of $URu_{0.68}Pd_{0.32}Ge$ in several fields up to 5 T.

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$\chi(T) = N_A\mu_{eff}^2/3k_B(T-\Theta_p)+\chi_0$, where N_A is the Avogadro number, μ_{eff} is the effective moment of the uranium ions, Θ_p is the paramagnetic Curie temperature and χ_0 is the temperature independent susceptibility.

A fit of the data yields $\mu_{eff} = 1.71\ \mu_B$, $\Theta_p = -54$ K and $\chi_0 = 0.5\times10^{-3}$ emu/mol. The obtained μ_{eff} value is situated in between those of URuGe and UPdGe [1]. A large value of Θ_p implies a strong antiferromagnetic coupling between f electrons. The value of the temperature independent susceptibility χ_0 was used to calculate the difference $[\chi(T)-\chi_0]$ shown in Fig. 1b. The log–log plot of $[\chi(T)-\chi_0]$ vs. T clearly illustrates a power law of the temperature dependence of low-field susceptibility, $[\chi(T)-\chi_0]\propto T^{-n}$, which hints at a non-Fermi liquid behavior. We found a relation $[\chi(T)-\chi_0]\propto T^{-0.48}$ for data collected in the temperature range 1.7–10 K and in a field of 0.02 T. This non-Fermi liquid character slowly vanishes as external magnetic field increases and for fields above 0.5 T the susceptibility apparently recovers the Fermi liquid property. It should be recalled that the power-law dependence of the susceptibility, $[\chi(T)-\chi_0]\propto T^{-n}$ with $n\sim0.5$ has previously been found in some non-Fermi liquid alloys, showing instability due to nearness to antiferromagnetic $Th_{1-x}U_xPt_2Si_2$ [4] or ferromagnetic $Th_{1-x}U_xCu_2Si_2$ [5] and $CePt_{1-x}Rh_x$ [6] ordering. For $URu_{0.68}Pd_{0.32}Ge$, a large magnetic susceptibility at 1.7 K ($\approx$ 23 emu/mol) provides additional backing of the closeness to a magnetic order. In such a system, non-linear effects are expected to be sizeable. We evaluated non-linear susceptibilities based on a relation: $M(H) = M_0+\chi_1H+\frac{\chi_3}{3!}H^3+\frac{\chi_3}{5!}H^5$, where M_0 is spontaneous magnetization, χ_1 is the linear susceptibility and χ_3, χ_5 are nonlinear ones.

The results of a fit of data are shown as solid lines in Fig. 2. We found $\chi_3 = -1.1\times10^{-3}\ \mu_B/T^3$ and $\chi_5 = 4.7\times10^{-7}\ \mu_B/T^7$ from the fitting of the 2 K magnetization. The absolute values of χ_3, χ_5 decrease with increasing temperature and at 10 K are negligibly small. This fact means that the non-linear effect no longer exists at temperatures above 10 K, where the magnetization is practically linear with applied fields.

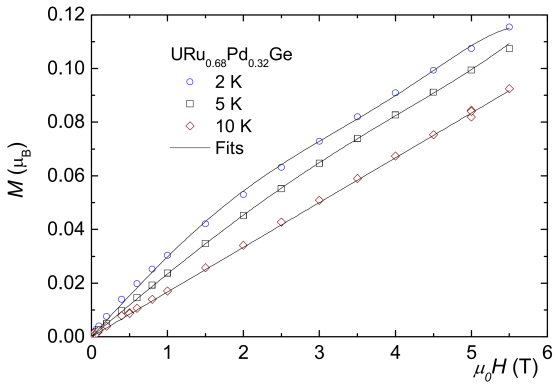

Fig. 2. Isotherms of $URu_{0.68}Pd_{0.32}Ge$. The solid lines are theoretical curves.

The specific heat $C_p(T)$ data of $URu_{0.68}Pd_{0.32}Ge$ are depicted in Fig. 3. The low-temperature $C_p(T)$ data show a tendency to rise, indicative of a large influence of electron correlations. We tried to analyze low-temperature data with a sum: $C_p(T)/T = \gamma+\beta T^2-\delta T^2\ln(T/T_{sf})$, predicted for nearly ferromagnetic Fermi liquids [7]. In the equation, the first term is the electronic specific coefficient, the second is associated with the crystal lattice and the last one denotes the contribution from the spin fluctuations. However, the agreement between the experimental and theoretical data is not satisfactory.

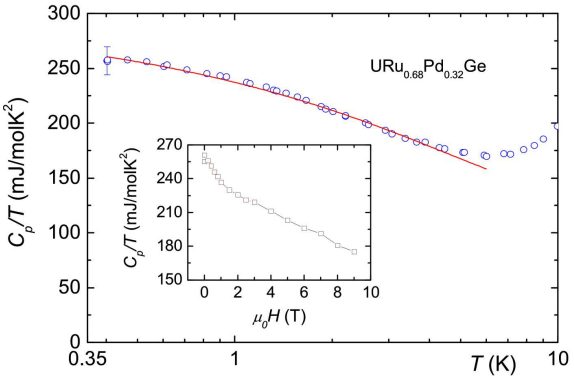

Fig. 3. The specific heat of $URu_{0.68}Pd_{0.32}Ge$ plotted as C_p/T versus $\ln T$. The solid line is a theoretical fit. The inset shows field dependence of the ratio C_p/T at 0.4 K.

It appears that for a good description a power law variation $T^{1/2}$ has to be introduced. The best fit of data between 0.4 K and 4 K to a sum: $C_p(T)/T = \gamma+\beta T^2-b\sqrt{T}$, yields $\gamma = 301(2)$ mJ/(mol K^2), $\beta = 0.43$ mJ/(mol K^4) and $b = 65(1)$ mJ/(mol K$^{5/2}$). Interestingly, similar $C_p(T)$ behaviour has been observed in many intermetallics alloys with magnetic instabilities, e.g., U_2Co_2Sn [8], $(U_{0.8}La_{0.2})_2Zn$ [9], $YbCu_{3.5}Al_{1.5}$ [10] and $U_{0.07}Th_{0.93}(Ru,Pt)_2Si_2$ [11]. We notice that the $C_p(T)/T\sim-\sqrt{T}$ dependence may account for non-Fermi liquid behavior. According to Hertz [12], Millis [13], and Moriya and Takimoto [14], the square root temperature dependence of C_p/T is predicted for antiferromagnetic spin fluctuations at $d=3$, contrary to ferromagnetic coupling, where a dependence $\propto C_p(T)/T-\ln(T/T_0)$ was anticipated.

A field-study of $C_p(T)$ reveals dramatic change of the $C_p(T)$ dependence. An application of magnetic fields rapidly suppresses the value of C_p/T at 0.4 K. Above 1 T the Fermi-liquid behavior of $C_p(T)$ is recovered and the Sommerfeld ratio decreases linearly with field (see inset of Fig. 3). It is worthwhile to emphasize that even in a high field of 9 T, the Sommerfeld ratio C_p/T remains still large enough, which certainly points to a moderate heavy-fermion behaviour. We estimated the Wilson ratio [15] $R_W = \pi^2k_B^2\chi(0)/[\mu_B^2\mu_{eff}^2\gamma(0)]$ to amount to 7.7, if one assumes the values $\chi(0) = \chi(2)$ K) and $\gamma(0) = C_p/T$ at 2 K. Such a large value of R_W is observed in systems with strongly magnetic correlations.

The electrical resistivity $\rho(T)$ of $URu_{0.68}Pd_{0.32}Ge$ is plotted in Fig. 3. Due to the presence of microcracks in the sample, the resistivity normalized to a room temperature value $\rho(T)/\rho_{300K}$ is given. With decreasing temperature the $\rho(T)/\rho_{300K}$ curve shows deviations from an ordinary metallic feature. An observed negative derivative $d\rho(T)/dT$ suggests the single-ion Kondo-type interaction, while the maximum around 150 K notifies the development of coherence state. Below 3.5 K, we observe a $\rho(T)$ upturn. The reason may be an electron–electron correlation or Kondo-like spin dependent scattering. Consequently, we fitted the low-temperature data to equations: $\rho(T)/\rho_{300K} = \rho_{res} - cT^{1/2}$ and $\rho(T)/\rho_{300K} = \rho_{res} - c\ln(T)$, respectively. However, the experimental data could not well satisfy above equations. Instead, the data between 0.4 and 10 K are well represented by $\rho(T)/\rho_{300K} = \rho_{res} - c\ln(T) + AT^{3/2}$, where ρ_{res} is a renormalized residual resistivity, c and A are constants (see solid line in Fig. 4).

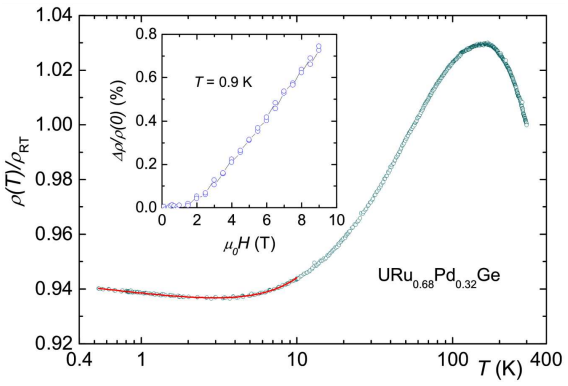

Fig. 4. Temperature dependence of the electrical resistivity of $URu_{0.68}Pd_{0.32}Ge$. The solid line is a theoretical curve. Inset shows field dependence of the magnetoresistance at 0.9 K.

The magnetoresistance, MR, defined as $\Delta\rho/\rho(0) = [\rho(H,T) - \rho(0)]/\rho(0)$, measured in fields up to 9 T (see inset of Fig. 4) is positive. There are two well-known mechanisms responsible for a positive magnetoresistance; i.e., Lorentz force and antiferromagnetic correlation. The first mechanism exists in all metallic systems. Owing to the reduction of the effective mean free path of the conduction carriers, the MR follows H^2-dependence in low fields and saturates in high fields. In the studied sample, the mechanism may be different than the conventional Lorentz force, since neither $\Delta\rho/\rho(0) \sim H^2$ nor a saturation is not found. So, we rather suspect $URu_{0.68}Pd_{0.32}Ge$ to be a system with dominating antiferromagnetic correlation.

In summary, we investigated low-temperature properties of $URu_{0.68}Pd_{0.32}Ge$. The observed power divergent resistivity and specific heat in good agreement with the theories for spin fluctuations [12–14]. However, the finding $\chi(T) \propto T^{-0.48}$ is in disagreement with any of the predictions from above models, where for antiferromagnetic spin fluctuations $\chi(T) \propto T^{-3/2}$ should be expected. The departure is not very surprising because of a nearness to magnetic order, which may give additional contribution to the magnetic susceptibility.

For discussion, it is interesting to compare the behaviour between two related solid solutions $URu_{1-x}Rh_xGe$ [16, 17] and $URu_{1-x}Pd_xGe$. In both systems, the long-range order can be induced just at a critical value of valence electrons/f.u., i.e., approximately 8.65 e/f.u. This proves that the number of sd -electrons of T-metals plays a role in establishing magnetic order. Further, the occurrence of non-Fermi liquid behaviour in $URu_{0.68}Pd_{0.32}Ge$ is very reminiscent of the case of $URu_{0.32}Rh_{0.68}Ge$ [16, 17]. In particular, the latter composition has been regarded as a f-electron system with a ferromagnetic quantum critical point at ambient pressure [16]. Thus, a question relating to possible quantum criticality in $URu_{0.68}Pd_{0.32}Ge$ remains open and it encourages further investigations.

Acknowledgments

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