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Thermoelectric Power of the $URu_{1-x}Pd_xGe$ System

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We report the measurements of thermoelectric power, $S(T)$, of the $URu_{1-x}Pd_xGe$ solid solutions in the temperature range 1.9–300 K. It is found that $S(T)$ of URuGe is consistent with the behaviour of Kondo lattice, characterized by a low-temperature negative minimum and a high-temperature positive maximum. On the contrary, $S(T)$ of the compositions $0.1 \leq x \leq 0.7$ is negative over the whole temperature range measured and shows only one negative minimum around 200 K. The compositions $x = 0.9$ and 1, in addition to a high-temperature negative minimum, exhibit anomalies at low temperatures, presumably associated with the magnon drag. We interpret the experimental data assuming the presence of the Kondo and crystal-electric field effects.

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1. Introduction

Previous investigations found that intermetallics URuGe and UPdGe, crystallizing in the orthorhombic TiNiSi-type structure have different magnetic ground states, i.e., paramagnetic and ferromagnetic, respectively [1, 2]. In order to search for quantum critical points nearby magnetic instability, we have investigated pseudoternary solid solutions $URu_{1-x}Pd_xGe$. Measurements of magnetization, $M(T)$, specific heat, $C_p(T)$, and electrical resistivity, $\rho(T)$, have revealed a dramatic change in their ground-state magnetic properties; from non-magnetic ($x < 0.35$), through antiferromagnetic ($x = 0.35$–0.8) to a complex magnetic state with two successive magnetic phase transitions in $x = 0.9$ and 1. It is remarkable that compositions located at the nonmagnetic-magnetic border were found to exhibit typical non-Fermi-liquid properties, such as $M(T) \sim T^{-0.48}$, $C_p(T) \propto -\sqrt{T}$ and $\rho(T) \propto T^{3/2}$ [3, 4]. To shed more light on the electron transport properties, we present in this contribution experimental data of thermoelectric power for $URu_{1-x}Pd_xGe$ and discuss them in the frameworks of the existing theories.

2. Experimental details

The samples of $URu_{1-x}Pd_xGe$ investigated in this work were the same as reported in [3]. The samples were cut into parallelepiped bars with typical dimensions $1 \times 1 \times 6$ mm^3. The measurements were made in the temperature range 1.9–300 K using a standard steady-state method, utilizing a commercial Quantum Design PPMS platform. At any measured temperature, the temperature gradient $\Delta T(T)$ of 0.5 K was established and the voltage difference between two terminals $\Delta U(T)$ was recorded. The Seebeck coefficient, hereafter called thermopower, is defined as $S(T) = -\Delta U(T)/\Delta T(T)$. Accuracy of the measurements, mainly due the heat loss is about ± 1 μV/K.

3. Results and discussion

Figure 1 shows the temperature dependence of the thermopower of URuGe. It is seen that the overall behaviour of $S(T)$ of URuGe resembles very much those of the Kondo lattices, like $CeCu_2Si_2$ [5], $CeCu_2Ge_2$ [6]. For URuGe the absolute value of S at room temperature is about 5 μV/K and steadily increases with decreasing temperature until to 100 K, where S attains its maximum value of 15 μV/K. The appearance of maximum in $S(T)$ can be understood in terms of single-ion model [7]. According to the model, thermopower should possess a maximum at $T_{\max}$, reflecting the Kondo coupling between the f- and the conduction electrons. The thermopower $S(T)$ is determined by the single scale Kondo temperature T_K, the effective spin moment J and the potential scattering phase shift, δ_v, and can be expressed as

$$S_K(T) = \frac{\pi^2 J(J+1)\sin(2\delta_v)}{\left[\ln^2\left(\frac{T_K}{T}\right) + \pi^2 J(J+1)\right]^{3/2}}. \quad (1)$$

We have fitted the experimental data for $T > 95$ K to the sum of the Kondo and diffusion terms

$$S(T) = cS_K(T) + S_d(T), \quad (2)$$

where the diffusion term $S_d(T)$ is linear to temperature αT. The parameters c and α are constants. The theoretical curve is shown as dashed line in Fig. 1. The fitting parameters are $c = 113$ μV/K, $J = 0.172$, $\delta_v = 0.85$, $T_K = 108$ K, $\alpha = 0.0024$ μV/K^2.

It is worthwhile to mention that the existence of high-temperature maximum $T_{\max}$ in $S(T)$ has been interpreted due to the interplay of the Kondo and crystal electric field (CEF) effect, characterized by a crystal-field splitting Δ [8]. The Δ parameter was also invoked in the theory developed by Zlatić and Monnier [9], who explained the thermoeletric behaviour of numbers of Ce- and Yb-based intermetallics. Due to the Kondo

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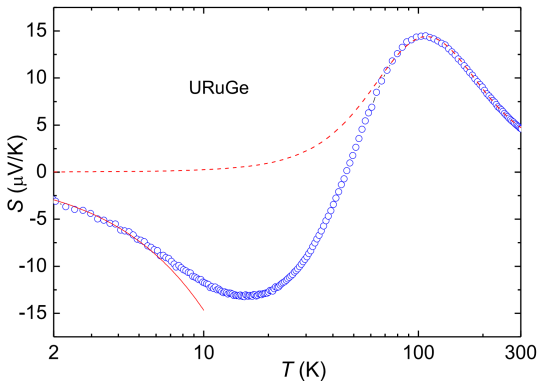

Fig. 1. The thermopower as a function of temperature for URuGe. The dashed and solid lines are theoretical curves.

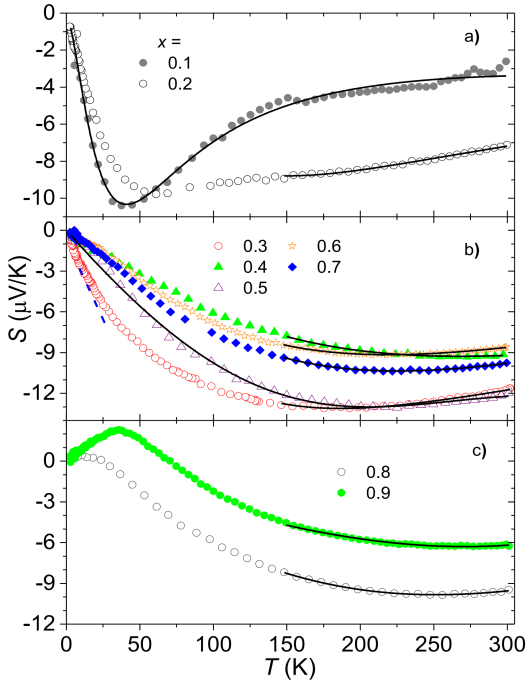

Fig. 2. The thermopower as a function of temperature for $URu_{1-x}Pd_xGe$. The dashed and solid lines are theoretical curves.

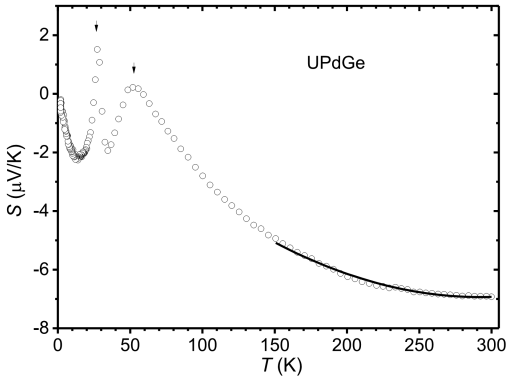

Fig. 3. Temperature dependence of the thermopower of UPdGe. The solid line is a fit of the high-temperature data to Eq. (4).

scattering off of the thermally populated CEF levels, $T_{\max}$ should locate between $\Delta/6$ and $\Delta/3$.

Interestingly, $S(T)$ changes its sign to negative at 45 K and exhibits a distinct minimum at 16 K. The mechanism of the negative $S(T)$ minimum is not quite understood. Fisher [7] assumed the presence of a resonance term due to spin interactions. This term has opposite sign to $S_K(T)$, therefore, may cause a change of sign at temperature about $0.6\,T_K$. On the other hand, negative minimum in the thermopower is frequently observed in spin fluctuators, such as UAl_2 [10], (La, Ca)Fe_4Sb_{12} [11]. Within the framework of a two-band paramagnon model, which is comprised of heavy and light electrons, Okabe [12] interpreted thermopower extremum, which occurs just below a characteristic temperature T_{sf}. The thermopower of URuGe below 5 K exhibits a linear dependence $S(T) = -1.47T$ (shown as solid line in Fig. 1), presumably due to the dominant contribution of drag of heavy electrons related with spin fluctuations.

In Fig. 2 we show the thermopowers of solid solutions with $0.1 \le x \le 0.9$. Clearly, small amounts of doped Pd up to $x \le 0.2$ (Fig. 2a), put down the high-temperature thermopowers, so the maximum around 100 K observed in URuGe vanishes. Instead, we observe a shift of the position of negative minimum to higher temperatures. The thermopowers of alloys in the concentration range $0.3 \le x \le 0.7$ (Fig. 2b) are similar to each other. $S(T)$ of these compositions is negative over the whole temperature range measured, and do exhibit only one extremum, i.e., a broad minimum around 200 K. This observation hints at a common nature of the phenomena in these alloys. There is a similarity of the data to those previous reported for Yb-based compounds, such as $YbAl_3$ [13] and $YbCu_2Si_2$ [14]. According to the theory [8, 9] the minimum can be ascribed to the Kondo scattering on the full CEF split multiplet of the magnetic uranium ions. In contrast to nonmagnetic ($0 \le x \le 0.3$) and antiferromagnetic ($0.3 < x \le 0.8$) alloys, the $x = 0.9$ composition (Fig. 2c) exhibits a positive $S(T)$ curve at low temperatures. For this alloy, we observe also a broad maximum at 36 K, nearby its magnetic phase transition ($T_C = 30$ K [3]).

In the same manner as for URuGe, the experimental data of $x = 0.1{-}0.9$ can be analyzed with Eq. (2). From the fittings we obtained values of the Kondo temperature (an illustration of the fit is shown as solid lines in Fig. 2 for $x = 0.1$ and $x = 0.5$.

Concerning the data below 5 K, we found a T-linear behaviour of thermopowers, which signals the contribution of electron diffusion. The low-temperature (LT) electron diffusion coefficient attains a maximum value of $\alpha_{LT} = -0.26\ \mu V/K^2$ at $x = 0.3$ (illustrated by dashed line in Fig. 2b).

In Fig. 3, we show thermopower of UPdGe as a function of temperature. The $S(T)$ curve is characterized by two maxima at 27 and 52 K. The position of these anomalies is found to coincide with the ferromagnetic ($T_C = 30$ K) and antiferromagnetic ($T_N = 50$ K) phase

transitions, respectively [1]. Therefore, in both the $x = 0.9$ and $x = 1$ compositions the appearance of $S(T)$ anomalies manifests the dominant contribution of the magnon drag to the thermopower. The fitting of high-temperature $S(T)$ data yield similar values to those of $x = 0.9$ (see Table).

TABLE

Position of the DOS peak of the $5f$ band in respect of the Fermi level $\varepsilon_f - \varepsilon_F$, width of the resonance peak Γ and LT and HT electron diffusion coefficients α_{HT} and α_{LT}.

x	T_K [K]	α_{LT} [μV/K]	$\varepsilon_f - \varepsilon_F$ [meV]	Γ [meV]	α_{HT} [μV/K]
0	108	−1.47	1.1	6.6	−0.009
0.1	40	−0.38	−1.3	32	0.0106
0.2	67	−0.32	−1.27	23	−0.0011
0.3	190	−0.26	−2.4	29	−0.0074
0.4	200	−0.09	−2.8	44	0.0023
0.5	235	−0.09	−3.5	38	0.0072
0.6	230	−0.08	−2.4	38	0.0042
0.7	270	−0.07	−3.1	40	0.0071
0.8	273	0.03	−5.7	56	0.0203
0.9	229	0.08	−17	100	0.048
1	223	0.21	−16.3	99	0.044

To examine any relation between the $S(T)$ and details of the electronic structure near the Fermi level, we analyzed the data with the help of phenomenological resonance model [15]. The model assumes that the dominant contribution to thermopower is issued from the scattering between electrons of a broad s-band and a narrow f-band with the Lorentzian shape, and can be expressed by

$$S_{HF}(T) = \frac{2\,(\varepsilon_f - \varepsilon_F)\,T/|e|}{\frac{3\left[(\varepsilon_f-\varepsilon_F)^2+\Gamma^2\right]}{(\pi k_B)^2} + T^2}, \tag{3}$$

where $\varepsilon_f - \varepsilon_F$ is the position of the density of state peak of f-electron band relative to the Fermi level and Γ is the width of the resonance peak. We fit the data with an equation:

$$S(T) = cS_{HF}(T) + \alpha_{HT}(T). \tag{4}$$

As an illustration of the fits, solid lines between 150–300 are shown in Figs. 2 and 3 for some selected alloys. Some of parameters obtained from fitting of the experimental data to Eq. (4) are given in Table.

4. Concluding remarks

We have performed a study of the thermopower properties of solid solutions $URu_{1-x}Pd_xGe$. The obtained results indicate the presence of the Kondo and CEF effects.

The magnon drag affects evidently the thermopower of $x = 0.9$ and 1. Using existing theories for the Kondo effect we have analyzed the experimental data. We derived some physical parameters such as the Kondo temperature, electron diffusion coefficients, position of the DOS peak of the $5f$ band in respect of the Fermi level $\varepsilon_f - \varepsilon_F$, width of the resonance peak. Two major conclusions emerge from the fitting parameters: (i) There exists close relationship between the Kondo temperature T_K and width Γ for $0.2 \leq x \leq 0.8$, i.e., with increasing x, both T_K and Γ increase. (ii) The position of the DOS peak of the $5f$ band is found to be sensitive to the composition in the series of alloys $URu_{1-x}Pd_xGe$. It appears that the $5f$ band shifts down further with increasing x, indicating increased $5f$-electron localization.

Acknowledgments

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References

[1] R. Troć, V.H. Tran, *J. Magn. Magn. Mater.* **73**, 389 (1988).

[2] V. Sechovsky, L. Havela, in: *Ferromagnetic Materials*, Eds. E.P. Wohlfarth, K.H.J. Buschow, North Holland, Amsterdam 1988, Vol. 4, and the references therein, p. 309.

[3] D. Gralak, A.J. Zaleski, V.H. Tran, *Acta Phys. Pol. A* **126**, 314 (2014).

[4] V.H. Tran, D. Gralak, *Acta Phys. Pol. A* **127**, 312 (2015).

[5] W. Franz, F. Steglich, D. Wohlleben, *J. Phys. Colloq. (France)* **40**, C5-342 (1979).

[6] P. Link, D. Jaccard, P. Lejay, *Physica B* **225**, 207 (1996).

[7] K.H. Fischer, *Z. Phys. B* **42**, 245 (1981).

[8] A.K. Bhattacharjee, B. Coqblin, *Phys. Rev. B* **13**, 3441 (1976).

[9] V. Zlatić, R. Monnier, *Phys. Rev. B* **71**, 165109 (2005).

[10] H. Armbriister, W. Franz, W. Schlabitz, F. Steglich, *J. Phys. (France)* **40**, C4-150 (1979).

[11] T. Takabatake, E. Matsuoka, S. Narazu, K. Hayashi, S. Morimoto, T. Sasakawa, K. Umeo, M. Sera, *Physica B* **383**, 93 (2006).

[12] T. Okabe, *J. Phys. Condens. Matter* **22**, 115604 (2010).

[13] D.M. Rowe, V.L. Kuznetsov, L.A. Kuznetsova, Gao Min, *J. Phys. D Appl. Phys.* **35**, 2183 (2002).

[14] D. Andreica, K. Alami-Yadri, D. Jaccar, A. Amato, A. Schenck, *Physica B* **259-261**, 144 (1999).

[15] U. Gottwick, K. Gloos, S. Horn, F. Steglich, N. Grewe, *J. Magn. Magn. Mater.* **47-48**, 536 (1985).