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Structural and electronic properties of the highly concentrated $U_xY_{1-x}Ru_2Si_2$ alloy system

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ABSTRACT

We report on the resistivity and thermopower measurements together with the microstructural investigations of two highly concentrated alloys of the $U_xY_{1-x}Ru_2Si_2$ alloy system, for $x = 0.5$ and $x = 0.67$, nominally. Microstructural analysis shows the existence of two different crystalline phases at the room temperature: the solid solution $U_cY_{1-c}Ru_2Si_2$ ($c < x$) phase and the phase which can be represented by chemical formula URu_2Si. We show that the uranium single ions dissolved in the YRu_2Si_2 matrix exhibit the ordinary or single-channel Kondo effect described by the Coqblin–Schrieffer Hamiltonian. In the light of the obtained results, the URu_2Si_2 compound is discussed.

1. Introduction

Already for about twenty five years, URu_2Si_2 has been one of the most intensively studied compounds because of the phase transition at $T_0 = 17.5$ K [1]. Although the superconducting transition at about 1.5 K, for which is/was taken to coexist with antiferromagnetism, is a very interesting subject of matter, the phase transition and particularly the nature of the low temperature (LT) phase, have drawn much more interests. The phase transition has been attributed to a type-I antiferromagnetic ordering with very small magnetic moment of about $0.03\mu_B$ per unit cell [1]. But this tiny moment is incompatible with large entropy change at the magnetic transition of about 300 mJ/K^2 mol. One would expect such entropy change if the moment of $0.5\mu_B$ was magnetically ordered. This long standing problem of Kondo physic has been known as the searching of the hidden order (HO) parameter of the LT phase. Today, some high pressure experiments suggest that the HO phase is not connected to the ordered magnetic moments, although the small-moment antiferromagnetism actually exists below T_0 at ambient pressure [2,3].

In order to understand underlying physics of the LT phase, quite different theoretical models were proposed. Some models take localized [4,5] and the other ones take itinerant character of the 5f electrons as a starting point [6]. Although some physical properties of URu_2Si_2 are considered within crystalline electrical field (CEF) picture, i.e., within the localized model, there is no unique interpretation of the experimental data coming from neutron scattering, magnetic susceptibility and specific heat measurements. In particular, essentially quite different CEF energy spectra are taken to explain the experimental data. Moreover, today, as seems, the opinion that the localized view is not valid prevails because there are no clear evidences of the CEF split energy levels of the U 5f level [7].

In order to try to resolve these controversies, we looked upon the U-impurity side of an alloy system to reveal experimentally if the U-single ion is characterized by the CEF levels. So far some investigations have been made on the alloys containing small amount of U dissolved in a nonmagnetic matrix like YRu_2Si_2 or $ThRu_2Si_2$. However, neither of these alloy systems shows a resistivity upturn at low temperatures, a hallmark of the Kondo interaction on the lowest doublet of a CEF levels, although one takes the U ion is a Kondo ion, like Ce or Yb are. Because of the absence of the sign of the one-channel Kondo scattering, the low temperature anomalous features were explained within multi-channel Kondo scattering, quadrupolar interactions [8,9] or by intersite interactions, which eventually lead to spin glass behaviour [10–13]. A clear spin-glass-like behaviour for a sample with nominally $x = 0.08$ concentration of U was found in $U_xY_{1-x}Ru_2Si_2$. The existence of two crystalline phases even in such low concentrated alloy was revealed. It was shown that only $c = 0.035$ of U was in the solid solution in this investigated sample [12].

Yet in the alloys with higher concentrations of U ion, one can observe the low temperature upturn in the resistivity. In addition, a striking characteristic of $U_xY_{1-x}Ru_2S_2$ is the sign of a transition, like

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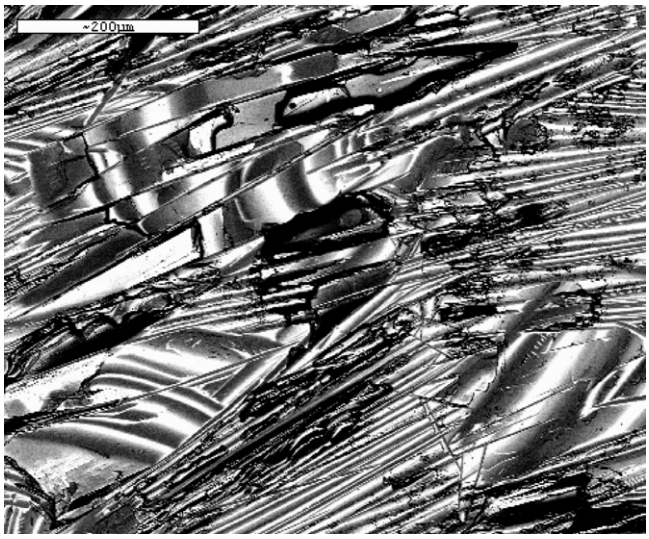

Fig. 1. One of the micrograph images of the $U_{0.50}Y_{0.50}Ru_2Si_2$ sample. Magnification: 200×. The "grey" phase, solid solution phase – smaller grains. The "light" phase – larger grains.

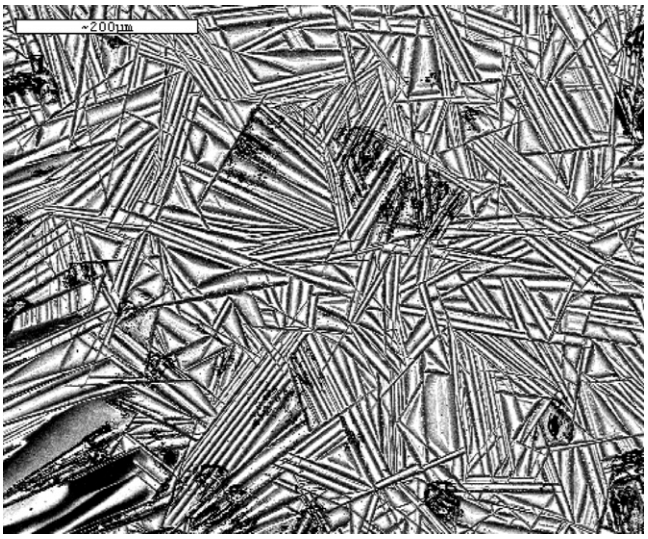

Fig. 2. One of the micrograph images of the $U_{0.67}Y_{0.33}Ru_2Si_2$ sample. Magnification: 200×. The "grey" phase, solid solution one – smaller grains. The "light" phase – larger grains. It is obvious that there is more "light" phase in the $U_{0.50}Y_{0.50}Ru_2Si_2$ sample.

it exists in URu_2Si_2, which persists even down to $x = 0.30$ [14]. An additionally intriguing characteristic of the transition in this alloy system is its temperature independency on the U content: it happens at about 17.5 K as in URu_2Si_2. These interesting findings have not been thoroughly investigated because it seems as this effect could come from URu_2Si_2 as a secondary phase in these samples of $U_xY_{1-x}Ru_2S_2$.

In this paper, we give an analysis of the resistivity and thermopower data combined with structural investigations of the $U_xY_{1-x}Ru_2Si_2$ alloy system for $x = 0.50$ and 0.67 nominally. In spite of complexity of the alloys investigated, we succeeded to extract the characteristics of the single U-ion in the YRu_2Si_2 matrix and revealed the spectra of the CEF energy levels of the U-ion. This is the main point of this paper, but we discuss also the impact of this result, and the other facts found investigating these alloys, on understanding of the long standing problem of URu_2Si_2. In addition, we can add that, as far as we know, it is the first extraction of crystalline electrical field energy levels from the resistivity data.

2. Preparation of the samples and microstructural investigations

Constituent elements were melted together on a water-cooled Cu hearth under Ar atmosphere. Samples, encapsulated in evacuated quartz tubes, were annealed at 600 °C for 2 days and then at 800 °C for 5 days. Here and after, we denote our investigated samples with their nominal concentrations, $U_{0.50}Y_{0.50}Ru_2Si_2$ and $U_{0.67}Y_{0.33}Ru_2Si_2$, although the actual structure was rather different and more complex.

Microstructure and elemental composition of the alloy have been analyzed using scanning electron microscope (SEM, Jeol JSM 5800) and energy dispersive X-ray spectroscopy (EDXS, Oxford – Link ISIS 300). Both SEM and EDXS analyses have shown that both samples were two-phase systems. The micrographs are shown in Fig. 1 ($U_{0.50}Y_{0.50}Ru_2Si_2$) and Fig. 2 ($U_{0.67}Y_{0.33}Ru_2Si_2$). It can be seen that in the samples prevails "grey" phase and that there is more "light" phase is in the $U_{0.50}Y_{0.50}Ru_2Si_2$ sample than in the $U_{0.67}Y_{0.33}Ru_2Si_2$ one. This qualitative overview is supported with quantitative analysis of the EDXS spectra. The quantitative analysis will be exposed and discussed later.

We also note that the Debye–Scherrer method of X-ray diffraction shows that both samples consist of solid solution phase (grey phase), which is the body-centered tetragonal $ThCr_2Si_2$-type structure, and of traces of a secondary phase (light phase).

Standard four-probe technique was used for the dc resistivity measurement from 2 to 300 K. The thermopower was measured in situ against a high-T_c YBCO material at low temperatures and against Pb at high temperatures. For further experimental details, see Ref. [15].

3. Results and discussion

The resistivity data of $U_{0.50}Y_{0.50}Ru_2Si_2$ and $U_{0.67}Y_{0.34}Ru_2Si_2$ together with the data of the nonmagnetic parent YRu_2Si_2 compound are shown in Fig. 3. Both resistivities exhibit a faint low-temperature upturn below a minimum at about 6 K which could be attributed to the hallmark of the ordinary one-channel Kondo effect. At about 17.5 K, one can observe a clear kink, which is seen much clearer in Figs. 5 and 6. This kink is of the same form as the one existing in URu_2Si_2 [1]. The resistivity exhibits a maximum at about 36 K and decreases with further temperature increase for $x = 0.67$, while for $x = 0.5$ the resistivity forms a minimum at about 150 K.

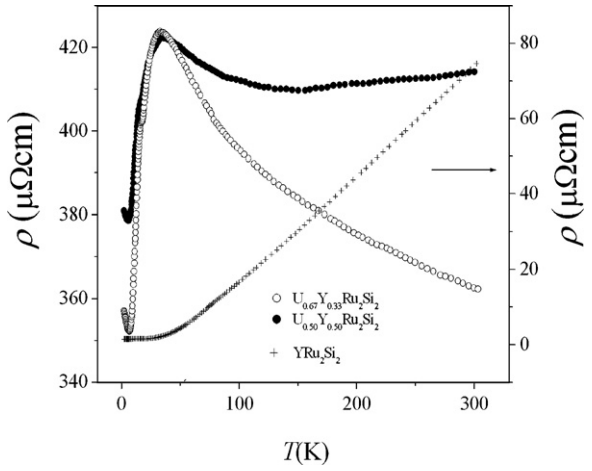

Fig. 3. The resistivities of $U_{0.50}Y_{0.50}Ru_2Si_2$, $U_{0.67}Y_{0.34}Ru_2Si_2$ and YRu_2Si_2.

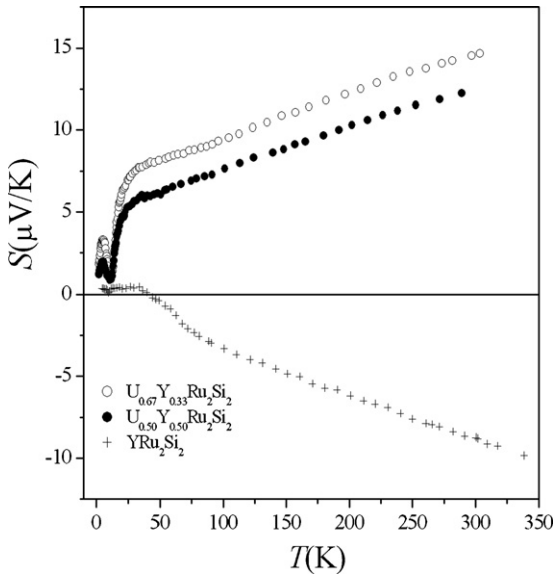

Fig. 4. The thermopowers of $U_{0.50}Y_{0.50}Ru_2Si_2$, $U_{0.67}Y_{0.34}Ru_2Si_2$ and YRu_2Si_2.

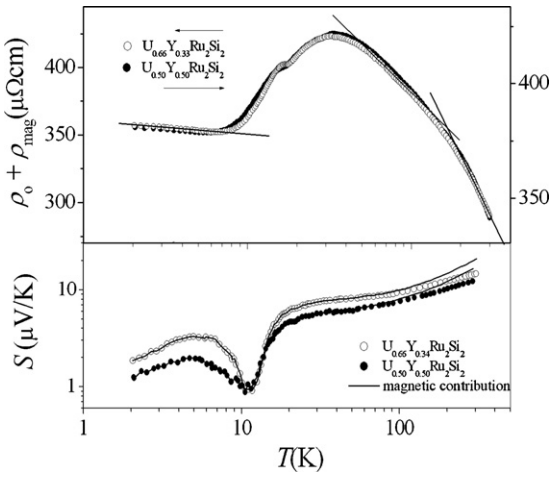

Fig. 5. (a, upper part) The magnetic resistivities of $U_{0.50}Y_{0.50}Ru_2Si_2$, $U_{0.67}Y_{0.34}Ru_2Si_2$ and YRu_2Si_2. (b, upper part) The magnetic thermopowers of $U_{0.50}Y_{0.50}Ru_2Si_2$, $U_{0.67}Y_{0.34}Ru_2Si_2$ and YRu_2Si_2.

The maximum at 36 K could arise from various reasons (for example, from small gap in semimetals [16]), but we ascribed it to the electron scattering on the excited CEF energy levels of the U-ion.

The resistivity of nonmagnetic YRu_2Si_2 (Fig. 1) exhibits temperature dependence typical for a phonon scattering in the case of ordinary metals.

The thermopower (Fig. 4) is positive; it increases with temperature and exhibits a maximum at about 5 K. An extreme in thermopower in the same temperature range where the resistivity exhibits logarithmic upturn, is another clear hallmark of the one-channel Kondo effect. With further temperature increase, the thermopower attains a minimum, and then increases up to the room temperature showing a faint maximum just about 36 K, where the resistivity shows a maximum indicating again that CEF excitations are responsible for these peaks. The same feature in thermopower can be observed in La doped URu_2Si_2 for the higher doping where coherence is removed [17]. The gross features of the thermopower and the resistivity data resemble the thermopower and resistivity typical for a Kondo system, like, e.g., in the $Ce_xLa_{1-x}Cu_{2.05}Si_2$ alloy system [18], although there is no such huge hump in the thermopower at high temperatures.

The magnetic resistivity of $U_xY_{1-x}Ru_2Si_2$, ρ_{mag}, is obtained by subtracting the resistivity of the nonmagnetic counterpart YRu_2Si_2, ρ_{nonmag}, from the measured resistivity ρ. Such procedure is the usual one and was also used in Refs. [18,19] (and in references therein):

$$\rho_{mag} = \rho - \rho_{nonmag} \quad (1)$$

The magnetic resistivities for the both samples exhibit the same temperature dependence in the whole temperature range which can be described by the equation

$$\rho_{mag} = \rho_o - c \cdot \rho_{mag1} \quad (2)$$

where ρ_{mag1} is a temperature-dependent single ion magnetic resistivity, while c is a concentration of the magnetic ions and ρ_o residual resistivity. In a simple Kondo system ρ_o is the unitary limit of resistivity and is proportional to the concentration of magnetic ions [20]. However, in a real and complex system there is scattering on various lattice obstacles and imperfections (vacancies, grain boundaries, cracks and so on). Hence, ρ_o is certainly sample-dependent in our multicomponent systems. In order to eliminate this parameter, we make a derivative of the $\rho_{mag}(T)$. The results are displayed in the inset of Fig. 6: the open circles for $U_{0.67}Y_{0.34}Ru_2Si_2$, and the line for $U_{0.50}Y_{0.50}Ru_2Si_2$. If the derivative of $\rho_{mag}(T)$ for the $U_{0.50}Y_{0.50}Ru_2Si_2$ sample is multiplied by scaling factor 1.86 (filled circles), the curves coincide. This value is too large in comparison to the nominal uranium concentration ratio 67/50 = 1.34. Interestingly enough, the quantitative analysis of the EDXS spectra from the grey phase in both samples gives that the alloy of nominally $x = 0.67$ can be represented just by $U_{0.67}Y_{0.34}Ru_2Si_2$ formula (13.54 at% U, 8.17 at% Y, 39.82 at% Ru and 38.47 at% Si) as expected. But the alloy of nominally $x = 0.50$ can be represented well by $U_{0.33}Y_{0.67}Ru_2Si_2$ formula (6.75 at% U, 14.07 at% Y, 40.34 at% Ru and 38.84 at% Si). If this is so, the scaling factor between these two curves, taking into account the real concentrations of magnetic ions, should be 2 (and not 1.34). The difference to the experimental result, 1.86, of about 8% is certainly within the cumulative errors in determining the magnetic resistivity curve. As a conclusion of this part, we may say that we successfully extracted the magnetic contribution to the resistivity of the single U dissolved in the YRu_2Si_2 matrix. Just this scaling by using the concentrations of the U ions found experimentally in the grey, i.e., in the solid solution phase of the two samples: $c = 0.33$ and 0.67, allows us to conclude that we reveal single ion property. The result of this scaling is displayed in Figs. 5 and 6.

The magnetic contribution to the thermopower is calculated by using Nordheim–Gorter rule [21]:

$$S_{mag} = \left(\frac{\rho}{\rho_{mag}}\right) S - \left(\frac{\rho_{nonmag}}{\rho_{mag}}\right) S_{nonmag}, \quad (3)$$

where S_{nonmag} is the thermopower of YRu_2Si_2 taken for the nonmagnetic contribution. The result is displayed in Fig. 5b with full thin lines. According to the theory, if there are no intersite interactions, as can be concluded from the scaling of the magnetic resistivities, the magnetic contribution to thermopower does not depend on concentration. As can be seen from Fig. 5b, the magnetic thermopowers for the two different U-concentrations have almost the same form in the whole temperature range measured. The difference in their values is not large and can rather be prescribed to the numerous sources of errors when calculating the magnetic contribution than to the non-single-ion effects. Note the four different measurement data sets are used in Eq. (3). One can also see that almost all thermopower consists of the magnetic part. The main reason is the high magnetic contribution to the resistivity which is mach higher than the nonmagnetic one.

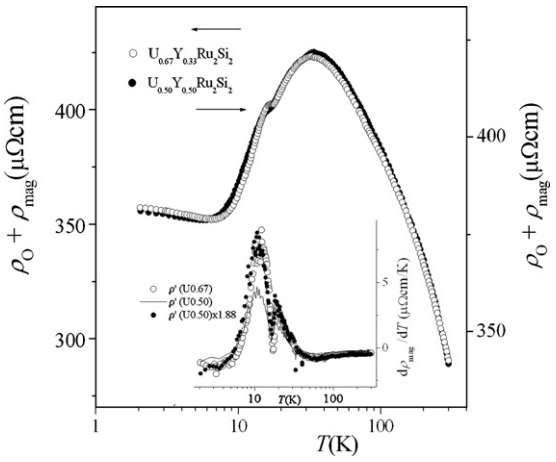

Fig. 6. The magnetic resistivities per mol U extracted from $U_{0.50}Y_{0.50}Ru_2Si_2$ and $U_{0.67}Y_{0.34}Ru_2Si_2$. Inset: the derivatives of magnetic resistivities of $U_{0.67}Y_{0.34}Ru_2Si_2$ (open circles) and of $U_{0.50}Y_{0.50}Ru_2Si_2$ (full line). By closed circles is denoted the magnetic derivative of $U_{0.50}Y_{0.50}Ru_2Si_2$ multiplied by 1.86.

The obtained magnetic resistivities (Fig. 5, or Fig. 6) are typical for scattering on a Kondo ion in the presence of CEF [22] and we shall discuss the results according to this theory. Based on the Coqblin–Schrieffer Hamiltonian (Eq. (112) from Ref. [22]), in the limit of weak potential scattering, the theoretical expression for the magnetic resistivity can be written as:

$$\rho_{mag} = 2\Re^* \frac{\lambda_n^2 - 1}{2J+1} \Gamma^3 g^2 \cdot \ln \frac{k_B T}{D^{(n)}}, \quad (4)$$

where Γ is the exchange coupling constant between conducting electron and f-electron, J is the total angular momentum of f-electron, g is the density of the electronic states at the Fermi level, D is the band width, $\Re^*$ is a constant, and k_B is the Boltzmann constant. The constant $\lambda_n = \sum_{i=1}^{n} \beta_i$ is a sum of the degeneracies β_i of the n lowest-lying CEF levels. The main features of the theoretical result (Eq. (4)) are few intervals with logarithmic temperature dependence due to the Kondo scattering of conducting electrons on the excited CEF levels.

Close inspection of the magnetic resistivity vs. $\log T$ in Figs. 5 and 6 reveals three almost linear parts which exist between 2 and 6 K, 70 and 140 K and 220 and 310 K. The slopes of the drown lines by the computer, when normalized to the highest temperature slope, are 0.0329 ± 0.0012, 0.429 ± 0.098, and 1.00, respectively. We compare the measured slopes with the theoretical ones $(\lambda_n^2 - 1)/4J(J+1)$ given by Eq. (4). For the U^{4+} ion, $J = 4$, there are $2J + 1 = 9$ of the 5f levels. The best match with the experiment data we obtain by taking a doublet ground state $\lambda_1 = 2$, four levels ($\beta_2 = 4$), $\lambda_2 = 6$, for the second slope and then λ_3 is 9. The theoretical normalized slopes are then 0.0375, 0.4375, and 1.00, respectively, and, as seen, they are very close to the corresponding experimental ones given afore. Note that the ground state doublet is in accordance with the presence of the Kondo interaction. If we assume that the U^{3+} ions having $J = 9/2$ exist in solid solution of $U_xY_{1-x}Ru_2Si_2$, the agreement with theory cannot be obtained whatever the choice of CEF energy scheme one takes. Hence, our analysis of the magnetic resistivity data supports that the $5f^2$, or the U^{4+} electronic configuration of the U-ion with a doublet ground state, exists in the $U_xY_{1-x}Ru_2Si_2$ alloy system. According to the resistivity and thermopower data (Fig. 5) the first excited CEF level is at about 36 K, indicated by the maximum in the magnetic resistivity and in the thermopower. The overall CEF splitting is about 300 K.

The low-temperature magnetic thermopower exhibits a low temperature maximum at about 6 K. It seems obvious that this maximum together with the upturn in the resistivity corresponds to the low temperature Kondo temperature, T_{KI}, due to the scattering at the ground-state doublet. The high temperature Kondo temperature, T_{Kh}, is about or higher than 300 K because all possible 9 levels are within 300 K.

It is interesting to note that although the effective concentrations of the two alloys considerably differ, their Kondo temperatures are almost the same. This can be understood by the fact that the lattice parameter depends only slightly on concentration and, therefore, there is no chemical pressure effect which could change the hybridization and consequently the Kondo temperatures.

From Figs. 5 and 6, we found that the lowest-temperature logarithmic slope of the Kondo upturn is 8.3 μΩ cm/mol-U. This is rather low value compared to the logarithmic resistivity slopes for some other Kondo alloys with doublet ground state; 185 μΩ cm/mol-Ce for Ce dissolved in the $LaCu_{2.05}Si_2$ matrix [14], and 53 μΩ cm/mol-Ce for Yb dissolved in $YInCu_4$ [19]. Within the Cornut–Coqblin theory [22], this slope is proportional to the product $\Gamma^3 g^2$ (Eq. (4)). Under a reasonable assumption that the density of electronic states g does not vary significantly for the above mentioned compounds, we conclude that hybridization Γ in the $U_xY_{1-x}Ru_2Si_2$ alloys is weak. This could explain why Kondo interaction is not observed for the lower concentrations of U [8–13]. In addition, the influence of the secondary phase, which exists also even for the lowest concentrations investigated [12] on the transport properties, is probably more significant for the low U concentrations. Thus, the single ion and, in addition, the one-channel Kondo behaviour for the lowest concentration has not been observed.

In what follows, we discuss the obtained results with respect to corresponding data found in literature. Before all, it should be said that these results are done mainly on URu_2Si_2 and there is only few data on highly concentrated U alloys and, moreover, these data were not analyzed thoroughly. Magnetic measurements, which are usually used to reveal the ionic state of a magnetic ion, are not able to make a clear distinction between the ionic state of the U ion because the theoretical values of the magnetic moments of the isolated ions U^{3+} and U^{4+} are almost the same: $3.578\mu_B$ and $3.68\mu_B$, respectively.

Analysing their resistivity data of URu_2Si_2, Schoenes et al. [23] assume that U^{3+} exists in URu_2Si_2. But in many other papers, the U^{4+} ion is taken to explain some experimental results. However, there are great and essential differences in the spectra of the CEF levels used to explain some results or are inferred from experimental data. Theory of crystal electrical field splitting for body centered tetragonal lattice gives that the corresponding Hund's rule $^3H_4$ (U^{4+}) state is ninefold degenerate and splits into five singlets and two doublets. To explain their neutron scattering data obtained on URu_2Si_2 at low temperatures, Brocholm et al. [4] assume that the first three levels up to 13 meV are singlets. These three levels are very dispersive and it could be taken that the other levels at higher energies are not observable. Similarly, to fit the susceptibility data of URu_2Si_2, Santini et al. [5] assume that the lowest lying levels are singlets. To explain and to fit the susceptibility of URu_2Si_2 measured in 50 T field, Sugiyama et al. [24] assume a doublet as a ground state. These essentially contradictory results for the CEF scheme of URu_2Si_2 together with the fact that neutron experiments give only three (out of nine) very dispersive spectral lines up to 13 meV, which are supposed to be singlets, lead to conclusion that there are no clear evidences of existence of CEF split energy levels in URu_2Si_2 [7]. However, we think that these results are not mutually quite contradictory regarding the question of the ground state. Moreover, they are in accordance with our results obtained on U doped YRu_2Si_2 ($c = 0.33$ and 0.67). The main and the most important question is: what is the ground state of the U ion. If the ground state is doublet, the one-channel Kondo interaction is possible and then it is

involved in the formation of the low temperature phase of URu_2Si_2. On contrary, if the ground state of the U ion is a singlet, Kondo interaction is excluded. To discuss this item, we assume a rather well established experimental fact: the spectrum of CEF levels of a magnetic ion does not change essentially and significantly with concentration going from impurity towards Kondo lattice regime in a Kondo alloy system thoroughly discussed in Ref. [18] and the references therein. Our experimental results of the resistivity and thermopower and the very good accordance to the theory undoubtedly show that the ground state of the U ion doped in the YRu_2Si_2 matrix is a doublet. Therefore, one expects that the ground state of U in URu_2Si_2 is a doublet, as well. Hence, the observed singlet as the ground state [4,5], we ascribe not to the U ion itself but to a dynamical or Kondo singlet formed of the magnetic moment of a Kondo ion and surrounding conduction electrons. Such singlets exist in URu_2Si_2 due to hybridization enhanced likely by coherency. In alloys there is no coherency while hybridization is weaker. The Gold–Schmidt radius of the Y^{3+} ion is 1.06 au and of U^{4+} is 1.05 au. Hence, for the higher concentration of U, the lattice parameter is lower and, therewith, the chemical pressure is stronger producing exponentially stronger hybridization between magnetic ion and conducting electrons. Besides the formation of singlets, a stronger interaction together with coherency destroys the sharpness and lowers the intensity of the lower lying CEF levels.

All these effects can be seen in the resistivity. We note that the maximum in the resistivity of URu_2Si_2 is at about 60 K [1,25] but not at 36 K as it is in the investigated alloys. This means that in URu_2Si_2 the maximum is due to interplay of the CEF levels and coherency, but in the alloys investigated, the maximum is due to excited CEF levels only. Going to lower temperatures, the resistivity does not show resistivity upturn because URu_2Si_2 tends to coherent heavy fermion state characterized by the Fermi liquid laws which is evidenced with the $+T^2$ dependence of the resistivity [1,25] and relatively large (negative) thermopower [26].

The measurement of susceptibility of URu_2Si_2 in the high field of 50 T [24] strongly supports our view. In high magnetic field, Kondo interaction is not effective and singlets do not form. Then the susceptibility assumes the characteristics of impurity regime and one has to introduce a doublet as the ground state of URu_2Si_2 to fit the susceptibility data. Above the ground state doublet the authors got a singlet at 39 K. For the other levels, a doublet and four singlets, it was obtained that they are above 3000 K. Our CEF scheme above the ground state doublet differs from the one in Ref. [24]. Four levels are between 36 K (where the maximum in the resistivity is) and 140 K and three levels are between 220 K and 310 K. In a conclusion of this part, we can say that, except the low concentrations where the one-channel single inpurity Kondo characteristics are not observed and except the kink at 17.5 K, the evolution of the transport properties with concentration in the $U_xY_{1-x}Ru_2Si_2$ alloy system resembles the evolution in the $Ce_xLa_{1-x}Cu_{2.05}$ alloy system – a Kondo alloy system [18]. Precisely speaking, these conclusions are worth for the solid solution phase, the grey phase of the $U_xY_{1-x}Ru_2Si_2$ alloy system because it changes with concentration.

In what follows, we discuss the influence of the secondary phase – the light one. The concentration analysis of the EDXS spectra from the light phase gives that there is: 22.44 at% U, 52.86 at% Ru and 24.70 at% Si for the $U_{0.50}Y_{0.50}Ru_2Si_2$ sample and 27.43 at% U, 48.52 at% Ru and 23.75 at% Si for the $U_{0.67}Y_{0.33}Ru_2Si_2$ sample. One can see that this crystalline phase can be well represented by the URu_2Si chemical formula in both samples. Moreover, we found that this phase, as a secondary phase, exists even in the alloy with the lowest concentrations investigated, $x = 0.08$ [12], but also for $x = 1$, i.e., in the case where the primary phase is URu_2Si_2 [25]. There is relatively much more U in URu_2Si than in URu_2Si_2 and one can expect that this crystalline phase has magnetically ordered ground state. Thus, it is naturally to assume that the small-moment antiferromagnetism is settled in the light phase, but not in the solid solution phase, i.e. in the grey phase. This idea is supported by our finding that the solid solution phase is characterized by an on-site interaction – the Kondo interaction, as we discuss afore. It is hard to reconcile the coexistence of Kondo interaction with magnetic ordering, i.e., it is hard to except that both effects, Kondo interaction and antiferromagnetism, coexist within the same phase.

For $x = 0.5$ and 0.67, we do not observe some effects of the light phase in the transport properties, expect, probably, in the residual resistivity. We point out that we have succeeded to extract the single impurity characteristics in the solid solution phase regardless the antiferromagnetism certainly exists at low temperatures as it exists in URu_2Si_2. This supports strongly the view that antiferromagnetisam does not exist within the solid solution phase.

Going further, we conclude that our experiments oppose the long standing opinion that the small-moment antiferromagnetism is inherent to the so called hidden ordered (HO) phase bellow T_0. This view is in accordance with investigations in Refs. [2,3] where the authors suspect that antiferromagnetism is connected to the so called HO phase. Our investigations show that the magnetically ordered phase, the light one, is separated from the solid solution phase, the grey one.

But there is still one interesting point for the alloys we investigate. It was found, for $x = 0.50$ and $x = 0.67$ (this work) and even for $x = 0.3$ [13] of the $U_xY_{1-x}Ru_2Si_2$ alloys system, that the drop of the resistivity exists at about $T_0 = 17.5$ K like it exists for $x = 1$ [1,25]. This rather strange and interesting point was not investigated thoroughly and was not discussed much. Namely, one could regard this effect as it arises from some small traces of URu_2Si_2 present in the alloys as secondary phase. Our microstructural investigations (Section 3) show that secondary phase does exist, but it is not URu_2Si_2. The first idea that seems naturals is that the transition happens in the light phase, i.e., in the URu_2Si phase, and that it is the antiferromagnetic transition. Namely, the chemical formula of this phase is independent on x as it is the temperature of the phase transition T_0. However, in that case one would expect larger effect for $x = 0.5$, where there is larger amount of the light phase, than in $x = 1$.

4. Summary

In order to understand the physics of the URu_2Si_2 compound, a long standing problem of the Kondo physics, we have sought to find the properties of the single U ion in the YRu_2Si_2 nonmagnetic matrix. There is astonishingly small number of investigations of U dissolved in a nonmagnetic matrix. One of the reasons can be attributed to the following: our investigations [12,13,25] together with this work and some other ones [2,3] indicate that it is difficult or even impossible to obtain pure solid solution if U is one of the constituent elements.

Here we present our investigations on higher concentrations of the $U_xY_{1-x}Ru_2Si_2$ alloy system: $x = 0.50$ and $x = 0.67$. For both samples, microstructural analysis showed the existence of two different crystalline phases; the solid solution phase $U_cY_{1-c}Ru_2S_2$, where $c < x$ ($c = 0.33$ and $c = 0.67$) and the URu_2Si phase. It was shown that the extracted magnetic contribution to the resistivity scales with c in the whole temperature range measured. Therefore, in spite of complexity of the systems consisting of two different crystalline phases, and in spite of the high U concentrations, we may say that we succeeded to reveal the magnetic contribution to the resistivity of the single U ion in the solid solution phase. General feature of the magnetic resistivity is explained by the ordinary one-channel Kondo scattering mechanism in the presence of CEF split energy levels of the U^{4+} ion using the Coqblin–Schrieffer theory [22]. The CEF scheme inferred from the magnetic resistivity consists of the doublet ground state, the first excited state roughly at about 36 K,

and the overall CEF splitting of about 300 K. The Kondo temperature, T_{K1}, due to the electron scattering on the ground state is about 6 K and is clearly indicated by the logarithmic upturn in the resistivity and the maximum in the thermopower. The logarithmic dependences at higher temperatures reveal the existence of Kondo scattering on the excited CEF levels.

Although our transport properties data can be explained within the single-channel Kondo theory, our work has wider significance. If one tries to explain the experimental results on URu_2Si_2 by localized scenario, for example taking quadrupolar of even octupolar character of f electrons, one should assume the existence of CEF excitations. But, as stated by Janik et al. in 2009, the CEF excitations are not clearly observed [26]. We think that this work can help in understanding the long standing problem of URu_2Si_2.

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