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Pressure-dependent resistivity studies of $(\text{Ce}_{1-x}\text{U}_x)\text{Al}_2$

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Abstract. Our studies show that hydrostatic pressure causes a substantial increase in Kondo temperature T_K and suppresses the magnetic order of the Ce-rich side of $(\text{Ce}, \text{U})\text{Al}_2$. For example, the magnetic transition of $(\text{Ce}_{0.7}\text{U}_{0.3})\text{Al}_2$ disappears above 5.5 kbar. Spin fluctuations dominant in the U-rich side become suppressed too, and simultaneously the U-rich systems are driven with pressure towards a simple metallic regime. A new scaling is found of the pressure-dependent resistivity of U-rich samples.

1. Introduction

Among the heavy-fermion compounds, CeAl_2 is a modest Kondo system with $\gamma = 135 \text{ mJ mol}^{-1} \text{ K}^{-2}$ and a Kondo temperature T_K of about 5 K [1]. It has an antiferromagnetic phase transition at around 3.9 K with an ordered moment of $0.85 \mu_B$ [2]. Thus CeAl_2 is a magnetically ordered Kondo-lattice system with a large localized moment, unusual among heavy-fermion compounds.

By comparison, UAl_2 does not show a magnetic transition or a superconducting transition down to 100 mK. UAl_2 is also a relatively moderate heavy-fermion compound with $\gamma = 142 \text{ mJ mol}^{-1} \text{ K}^{-2}$ [3]. Various physical properties, particularly the $T^3 \ln T$ behaviour of the low-temperature specific heat, characterize UAl_2 as a typical spin-fluctuation system with $T_{SF} \sim 27 \text{ K}$ [3]. UAl_2 is also distinguishable from other heavy-fermion systems in that it has very large $T = 0$ fluctuations, as seen in quasielastic neutron scattering experiments [4]. The quite large quasielastic linewidth of 25 meV for UAl_2 is one order of magnitude larger than those of most heavy-fermion compounds. Both CeAl_2 and UAl_2 have the cubic Laves MgCu_2 -type structure with lattice constants of $a = 8.059$ and 7.766 \AA respectively.

In the present work, we study the resistivity of $(\text{Ce}, \text{U})\text{Al}_2$ as a function of pressure, wherein the antiferromagnetic ground state of CeAl_2 with well localized magnetic moments changes over to a nearly magnetic spin-fluctuation ground state as in UAl_2 .

2. Experimental details

All samples were made using an arc furnace under a Ti-gettered Ar atmosphere. We prepared $(\text{Ce}_{1-x}\text{U}_x)\text{Al}_2$ alloys with $x = 0, 0.05, 0.1, 0.2, 0.3, 0.8, 0.9, 1$. Subsequently, they were subjected to heat treatments. Ce-rich samples were annealed at 800 °C for two days and then at 873 °C for five days, while U-rich samples were annealed at 850 °C for five days [5].

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The pressure-dependent electrical resistivity was measured for bar-shaped samples using the standard four-probe dc method in a temperature range from 1.5 K to room temperature. We used a liquid pressure cell with a 4:1 methanol–ethanol mixture as a pressure transmitter to generate hydrostatic pressure up to 12 kbar. The absolute value of the pressure was determined from the superconducting transition temperatures of In and Pb.

3. Experimental results

Both CeAl_2 and UAl_2 have low residual resistivity values ρ_0 of 12.1 and 14.0 $\mu\Omega$ cm respectively which increase rapidly upon substitution. For example, ρ_0 increases up to 180 $\mu\Omega$ cm for $(\text{Ce}_{0.8}\text{U}_{0.2})\text{Al}_2$. This large increase of ρ_0 is partly due to disorder caused by the Ce/U substitution, but is primarily attributed to the destruction of the *coherence* in Kondo scatterings.

$\rho(T)$ for the Ce-rich alloys shows the characteristics of a magnetically ordered Kondo lattice with a crystal-field splitting [6] and increases substantially with pressure. Figure 1 shows, as an example, the results of our measurements on $(\text{Ce}_{0.9}\text{U}_{0.1})\text{Al}_2$ and $(\text{Ce}_{0.8}\text{U}_{0.2})\text{Al}_2$. The overall increase in the resistivity with pressure is due to increased hybridization between conduction electrons and the localized f electrons of Ce ions. Since the same hybridization is responsible for both Kondo screening and magnetic RKKY interaction, we expect a strong competition between the two. The low-temperature resistivity of Ce-rich alloys is characterized by an almost pressure-independent minimum at about 15 K. Below the minimum, the resistivity shows a negative-temperature-coefficient behaviour due to the Kondo scattering of conduction electrons. Upon further cooling, all Ce-rich alloys studied become magnetically ordered and the resistivity drops steeply below the Néel temperature. Close inspection of the pressure effect on the resistivity reveals that the Kondo scattering increases with pressure: as pressure increases the negative-temperature-coefficient behaviour becomes more pronounced in the resistivity of CeAl_2 and $(\text{Ce}_{0.95}\text{U}_{0.05})\text{Al}_2$. In the case of $(\text{Ce}_{0.9}\text{U}_{0.1})\text{Al}_2$, however, the difference between the minimum and a local maximum at around 4 K increases with pressure up to 6 kbar and then begins to decrease as pressure increases (see figure 1).

Below the negative-temperature-coefficient behaviour, the resistivity of the Ce-rich alloys reaches a local maximum and then decreases rapidly due to the antiferromagnetic ordering. With increasing pressure, the maximum moves towards lower temperature; for example, with 12 kbar it moves from 5.06 to 4.46 K for CeAl_2 , from 6.03 to 4.86 K for $(\text{Ce}_{0.95}\text{U}_{0.05})\text{Al}_2$ and from 6.91 to 5.35 K for $(\text{Ce}_{0.9}\text{U}_{0.1})\text{Al}_2$. Therefore, the Néel temperature of the Ce-rich alloys decreases substantially due to increased hybridization with pressure. From the pressure dependence of the antiferromagnetic transition, we conclude that the Ce-rich alloys locate near a magnetic-to-nomagnetic transition point in the strong-coupling limit of the Doniach phase diagram [7]. It is also interesting to note that the chemical pressure effect due to U doping is opposite to the external pressure effect despite the fact that U doping also reduces the cell volume of CeAl_2 like external pressure. This apparent discrepancy between the chemical and hydrostatic pressure effects will be discussed in detail later.

Another interesting point regarding the Ce-rich compounds is the pressure effect on the resistivity contribution from the crystal-field excitation of Ce. A broad hump at 80 K is due to the effect of crystal-field excitation. As one can see in figure 1, it becomes pronounced with pressure which is consistent with a view that conduction electron scattering by the crystal-field excitation increases as hybridization between conduction electrons and f electrons becomes larger with pressure. For $(\text{Ce}_{0.9}\text{U}_{0.1})\text{Al}_2$, not only does the height of the hump increase but also the resistivity slope changes markedly (see figure 1). On the other hand, for $(\text{Ce}_{0.8}\text{U}_{0.2})\text{Al}_2$ there is only a very weak sign of crystal-field excitation at high pressure.

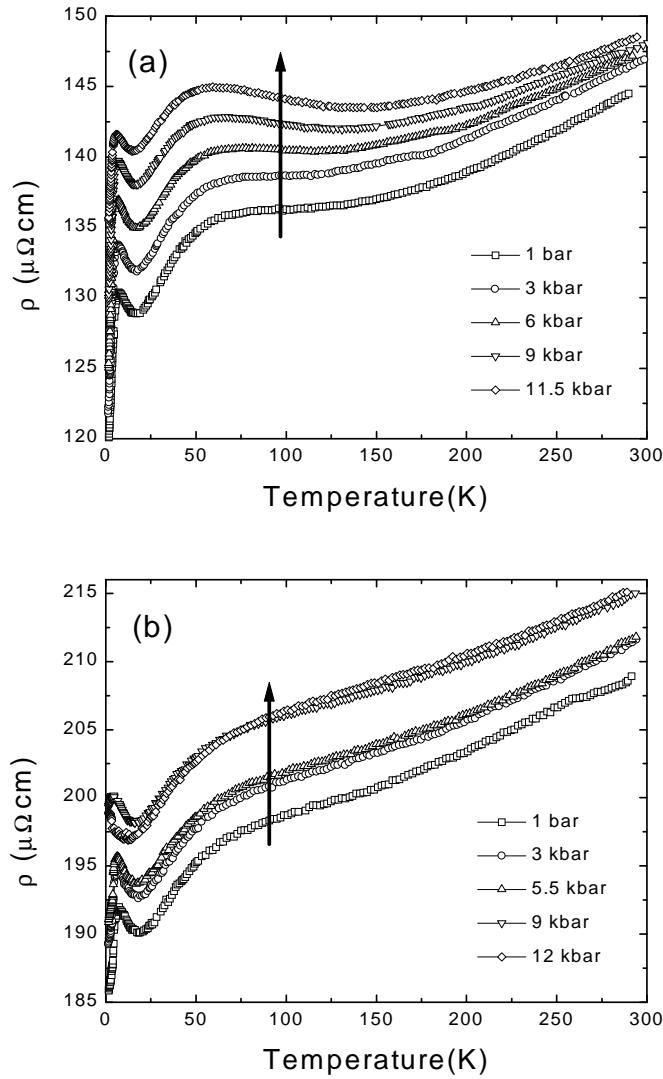


Figure 1. Resistivities of two Ce-rich alloys, (a) $(Ce_{0.9}U_{0.1})Al_2$ and (b) $(Ce_{0.8}U_{0.2})Al_2$, at different pressures. Pressure increases in the direction of the arrows.

$(Ce_{0.7}U_{0.3})Al_2$ appears to be weakly magnetic and the negative-temperature-coefficient behaviour is very modest at ambient pressure (see figure 2). On increasing the pressure above 5.5 kbar, both the magnetic ordering and the negative-temperature-coefficient behaviour become considerably reduced, indicating that the magnetic transition temperature moves toward much lower temperature. It is also noticeable that the resistivity starts to decrease with pressure for $(Ce_{0.7}U_{0.3})Al_2$. This marks most probably the pressure-induced suppression of spin fluctuations, which already seem to exist in this composition as in UAl_2 .

Figure 3 shows the pressure-dependent resistivity of a U-rich alloy, $(Ce_{0.1}U_{0.9})Al_2$. Unlike that of the Ce-rich alloys, the resistivity of the U-rich alloys decreases with pressure. As noted in the introduction, UAl_2 is a spin-fluctuation system. In such a system, magnetic interactions

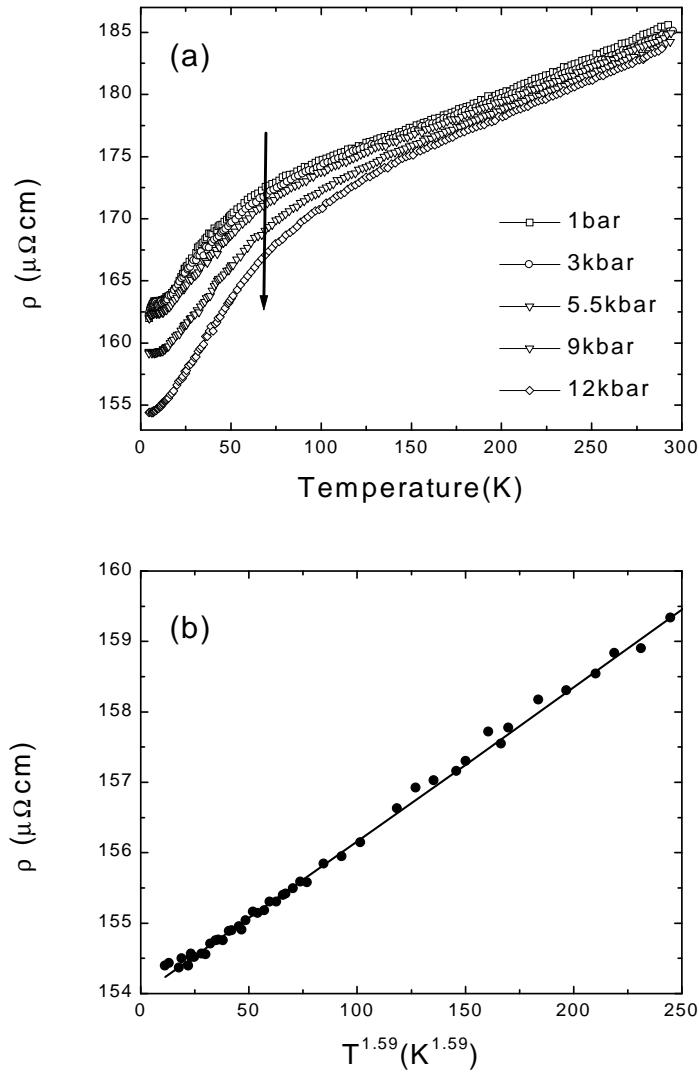


Figure 2. (a) The resistivity of $(\text{Ce}_{0.7}\text{U}_{0.3})\text{Al}_2$ at different pressures. Pressure increases in the direction of the arrow. (b) The low-temperature resistivity at 12 kbar is shown to scale with $T^{1.59}$ below 30 K.

are not strong enough to stabilize a long-range magnetic order, but they nevertheless produce some low-lying spin fluctuations. So it seems to be natural to interpret the pressure effect of $(\text{Ce}_{0.1}\text{U}_{0.9})\text{Al}_2$ in such a way that pressure suppresses the spin fluctuations and reduces resistivity. Therefore, the observed resistivity reduction with pressure can be understood as arising from the suppression of spin fluctuations. A signature for such a pressure-induced suppression of spin fluctuations can also be seen in $(\text{Ce}_{0.7}\text{U}_{0.3})\text{Al}_2$ at high pressure (see figure 2).

The resistivity of UAl_2 is characterized by an S-shaped behaviour showing a saturation behaviour near room temperature. As the Ce concentration increases, the residual resistivity value increases and reduction in the resistivity below 70 K becomes smaller, so the system eventually behaves like a simple metal, but with a quasi-linear temperature dependence at low

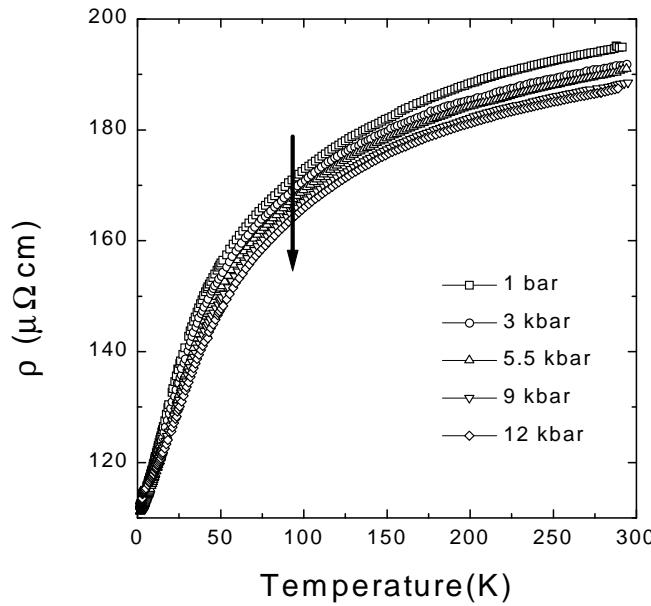


Figure 3. The resistivity of a U-rich alloy, $(Ce_{0.1}U_{0.9})Al_2$, at different pressures. Pressure increases in the direction of the arrow.

temperature. The ratio of the resistivity value at 1.5 K to the room temperature resistivity value for ambient pressure is 8% for UAl_2 while they are 58% and 67% for $(Ce_{0.1}U_{0.9})Al_2$ and $(Ce_{0.2}U_{0.8})Al_2$ respectively. The very rapid increase in ρ_0 with Ce substitutions is again due to the destruction of the *coherence* of the spin fluctuations [8]. A change is observed in the curvature of the resistivity at around 27 K (i.e., a maximum in $d\rho/dT$), which corresponds well with the spin-fluctuation temperature of UAl_2 determined from heat capacity data [3]. It becomes subdued as Ce concentration increases, in agreement with the view that Ce doping disturbs the *coherence* of the spin fluctuation in UAl_2 .

4. Discussion

A sharp drop in $\rho(T)$ below 4 K for the Ce-rich alloys is due to the antiferromagnetic ordering. With increasing pressure, Kondo screening of conduction electrons, which is destructive to magnetic ordering, gains in strength and the magnetic transition temperature T_N decreases monotonically with pressure. In the previous section, we argued that $CeAl_2$ locates near to the maximum of the magnetic transition temperature in Doniach's phase diagram. Therefore, we expect T_N to decrease with increasing hybridization on applying pressure, which is borne out in our data (see figure 4). In figure 4, two things are noteworthy. Firstly, T_N for the Ce-rich alloys increases with U doping while it decreases with pressure, although both external pressure and U doping reduce the cell volume. Secondly, the pressure dependence of T_N becomes stronger with U doping.

First of all, we think that the difference between the T_N -dependences on the U concentration and the pressure is due to the valence of U in UAl_2 . To date, U in UAl_2 has been thought to have 3+ valence, which was suggested from neutron scattering data [4]. However, with much larger lattice constants of the Ce-rich alloys, it may be possible that the U ion might have 4+

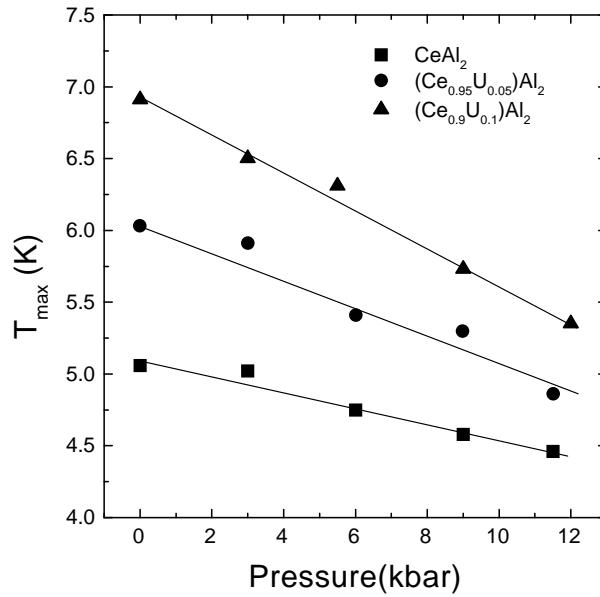


Figure 4. T_{\max} versus applied pressure for Ce-rich alloys.

valence. If this is the case, then one excess conduction electron from doped U would give rise to an increase in the Fermi level, which subsequently leads to a Fermi-level tuning effect as in $(Y, U)Pd_3$ [9]. This then can have a much stronger effect on hybridization than the changes in lattice constants due to U doping. In fact, our recent photoemission experiment on a similar composition seems to support this conjecture [10].

Secondly, in order to investigate quantitatively the change of T_N with pressure, we determined experimentally magnetic Grüneisen parameters from the graph of T_N versus pressure (see figure 4). The magnetic Grüneisen parameter Γ_N is defined as

$$\Gamma_N = -\frac{\partial \ln T_N}{\partial \ln V} = B \frac{\partial \ln T_N}{\partial P} \quad (1)$$

where the bulk modulus B was taken as the value for $CeAl_2$, 700 kbar [11]. The thus-obtained Γ_N values are -7.8 , -11.7 and -13.5 for $CeAl_2$, $(Ce_{0.95}U_{0.05})Al_2$ and $(Ce_{0.9}U_{0.1})Al_2$ respectively. Γ_N for $CeAl_2$ is in good agreement with the value obtained from previous thermal expansion and specific heat measurements using the Ehrenfest relation [12]

$$\Gamma_N = -B \frac{\delta \beta}{\Delta C} \quad (2)$$

where β is the thermal expansion coefficient and C the specific heat. Γ_N for U-alloyed $CeAl_2$ becomes larger with U concentration. This is consistent with the observation that the pressure-induced change in the resistivity becomes larger as the U doping increases up to 10%.

As can be seen in figures 1 and 2, the AF transition of $CeAl_2$ is suppressed with pressure and seems to be completely absent in $(Ce_{0.7}U_{0.3})Al_2$ at 12 kbar. Thus $(Ce_{0.7}U_{0.3})Al_2$ has a magnetic-to-nonmagnetic crossover at about 12 kbar. Interestingly, the low-temperature resistivity of $(Ce_{0.7}U_{0.3})Al_2$ shows an unusual temperature dependence in the pressure range in which the antiferromagnetic transition disappears; it has $T^{1.59}$ -dependence below 30 K (see figure 2(b)). The unconventional temperature dependence at critical pressure bears some resemblance to what is often found in other heavy-fermion compounds at similarly critical

points. For example, $Ce(Cu, Au)_6$ shows T -linear behaviour at a critical concentration [13]. Thus we think that the $T^{1.59}$ -behaviour can also be seen as a sign of non-Fermi-liquid behaviour at critical pressure.

The low-temperature resistivity of UAl_2 shows a change in curvature around a spin-fluctuation temperature T_{SF} . T_{SF} is a characteristic temperature which sets the energy scale of the spin fluctuations and is regarded as an average energy of paramagnons [14], above which temperature electron-electron correlations are destroyed. Below T_{SF} , electron correlation becomes important. If we take the inflection point in the resistivity as T_{SF} , T_{SF} at ambient pressure is 27.8 K. It is very close to the T_{SF} -value determined from other measurements, such as specific heat [3], susceptibility [15] and magnetoresistance [15].

At low temperature, the resistivity shows somewhat unusual temperature dependence: in this case, resistivity is shown to scale with $T^{1.5}$ (see figure 5(a)). We take A as the coefficient of $T^{1.5}$. With increasing pressure, T_{SF} shifts towards higher temperature. Spin-fluctuation theories predict the relation $T_{SF} \propto 1/\sqrt{A}$, which is indeed found for UAl_2 (see figure 5(b)).

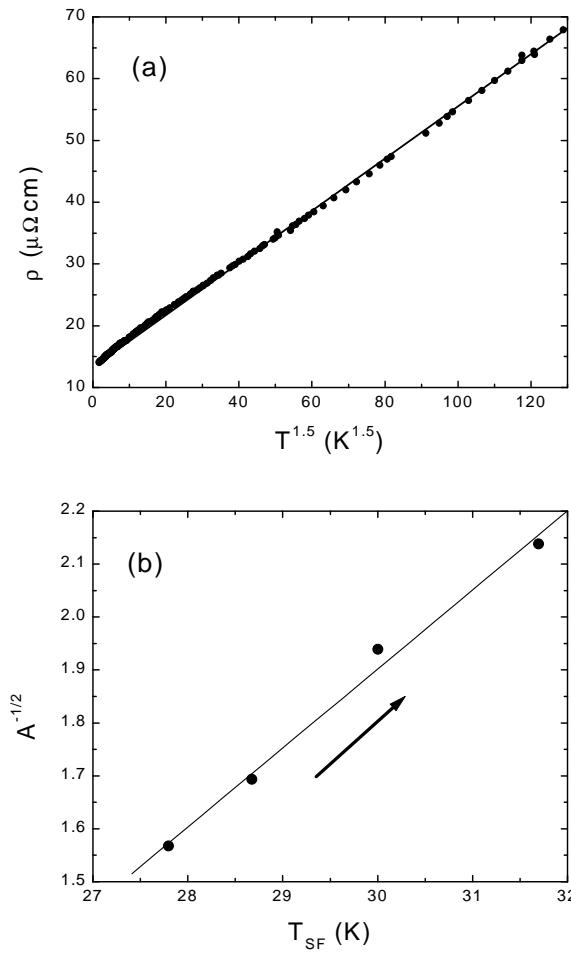


Figure 5. (a) The low-temperature resistivity of UAl_2 is shown to scale with $T^{1.5}$. (b) $A^{-1/2}$ in units of $1/\sqrt{\mu\Omega \text{ cm } T^{-1.5}}$ is given as a function of T_{SF} . Pressure increases in the direction of the arrow.

For $(\text{Ce}_{0.1}\text{U}_{0.9})\text{Al}_2$ and $(\text{Ce}_{0.2}\text{U}_{0.8})\text{Al}_2$, the inflection point locates at lower temperature and the slope of the low-temperature resistivity approaches quasi-linearity down to 10 K, revealing a less clear indication of spin-fluctuation effects in the resistivity. From this observation, we argue that about 10% Ce doping is enough to suppress a coherent state of a spin-fluctuation system like UAl_2 .

To compare the pressure dependence of the characteristic temperatures of UAl_2 and CeAl_2 , we calculated $t_{SF} = T_{SF}^{-1}(\text{d}T_{SF}/\text{d}p)_{p=0}$ for UAl_2 and $t_N = T_N^{-1}(\text{d}T_N/\text{d}p)_{p=0}$ for CeAl_2 . Surprisingly, t_{SF} and t_N turn out to have the same value of 0.011 kbar^{-1} . This is a quite interesting result when we consider that the ground states of the two materials are of very different nature. Previously, we found that the temperature and alloying dependence of the thermopower for CeAl_2 and UAl_2 are also very similar [16]. We interpreted the similarity of the thermopower data for the two systems in terms of the existence of possibly Lorentzian-type excitations near the Fermi level. Likewise, we think that low-lying excitations of both CeAl_2 and UAl_2 have similar pressure dependence, producing almost the same pressure effect on T_N and T_{SF} .

We fitted the resistivity of UAl_2 at various pressures to a scaling function of $T/T_{SF}(p)$ over a temperature range from 5 to 30 K (see figure 6). Such a scaling behaviour can be regarded as evidence strongly supporting the view that the resistivity of UAl_2 is dominated by spin-fluctuation scatterings in the pressure range studied here. The resistivities of 10% and 20% Ce-doped alloys do not show such a scaling behaviour, which is likely to be due to the fact that the *coherence* of spin fluctuations has already broken down in these compositions.

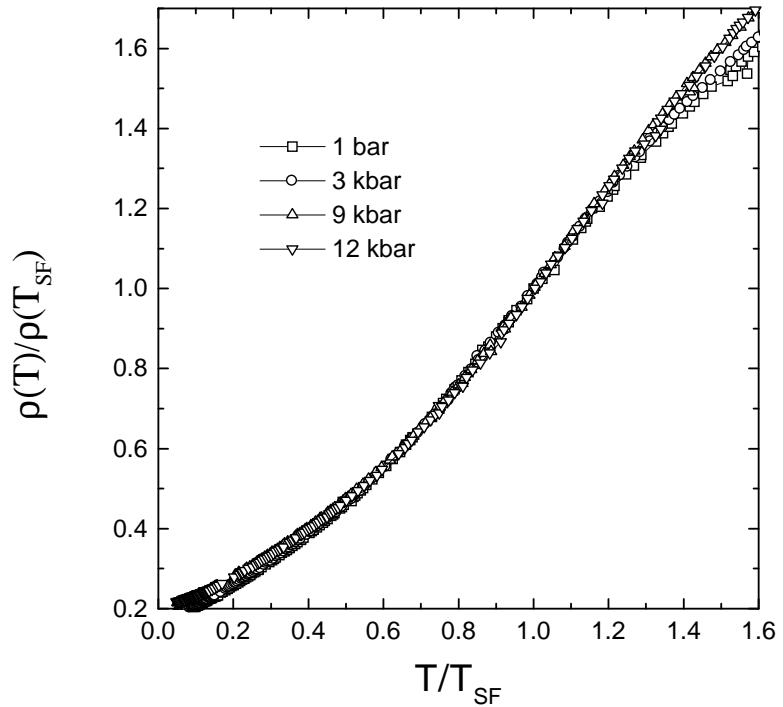


Figure 6. Scaling behaviour of the pressure-dependent resistivity of UAl_2 around T_{SF} is found. Resistivity data for different pressures collapse onto a single function of the scaled temperature $T/T_{SF}(p)$.

5. Summary

For the Ce-rich alloys, pressure causes the Kondo interaction to increase while the magnetic ordering temperature T_N decreases with pressure and while it increases with U doping. The difference seen in T_N as a function of pressure or U concentration is likely to be due to the Fermi-level tuning effect arising from the excess conduction electron of U compared with Ce. Magnetic order disappears for $(Ce_{0.7}U_{0.3})Al_2$ at high pressure. At 12 kbar, it exhibits a non-Fermi-liquid-like behaviour.

For the U-rich alloys, suppression of spin fluctuations by pressure is observed and a new scaling behaviour of the pressure-dependent resistivity is found. Suppression of spin fluctuations by pressure results in decrease of both the resistivity value and the low-temperature slope A , which is accompanied by an increase in T_{SF} . We find that the *coherence* of the spin fluctuations of UAl_2 is destroyed by more than 10% Ce doping.

Acknowledgments

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