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Studies of alloying effects on $URu_2(Si, X)_2$; X = Al or Ge

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Abstract. We have studied alloying effects of Al and Ge on the Si site of URu_2Si_2 using resistivity and magnetization measurements. In contrast to previous studies of substitutions on the U and Ru sites, we observe that the substitutions of Al or Ge produce only a simple dilution effect on the itinerant antiferromagnetism transition at 17.5 K in URu_2Si_2. We discuss the alloying effects on T_N, and in particular resistivity, in terms of d–f and sp–f hybridization.

1. Introduction

Among four possible ground states for heavy-fermion compounds, a superconducting ground state attracts by far the most attention, perhaps because its origin is believed by some to be related to that of high-temperature superconductivity. Other possible ground states are a non-magnetic and non-superconducting state, a magnetically ordered state, and a state with low carrier density. Having $T_N = 17.5$ K and $T_c \simeq 1.2$ K [1], URu_2Si_2 reveals interesting aspects of its ground state, some of which are shared by other heavy-fermion superconductors. First of all, the antiferromagnetic order parameter continues to develop almost linearly below T_N and does not disappear below T_c [2]. Secondly, it has a tiny ordered moment [3], 0.03 μ_B, along the c axis; interestingly enough, such a tiny ordered moment has been also found in other heavy-fermion superconductors except UPd_2Al_3 (where μ_{ord} is 0.85 μ_B) [4].

In addition, the antiferromagnetic transition at 17.5 K is itself unique in that although the size of ordered moment is as small as 0.03 μ_B, the entropy released below T_N is as great as 0.17 R ln 2 [1]. Because of such interesting features, there have been many alloying experiments [5, 6] on both the U and Ru sites, shedding light on the antiferromagnetic and superconducting transitions. However, little work has been done in alloying studies on the Si sites; only one group [7] has studied the effects of Ge doping primarily using specific heat measurements. Since those show surprisingly little effect of Ge doping on T_N, we feel it is desirable to study the alloying effect on the Si site more thoroughly. Here we present electrical and magnetic studies of substitutions of Al or Ge on the Si site.

2. Experiment

All samples have been made by arc melting constituent elements on a water cooled Cu hearth under Ar atmosphere; after melting, less than 0.5% weight loss was observed. All samples except for pure URu_2Si_2 were subsequently annealed, first at 600 °C, and then at 800 °C for five days. After the annealing process, samples were checked by metallographic examination. All samples presented here turned out to be single phase. Further x-ray diffraction studies using Cu Kα radiation (Phillips PW1700) on some of samples confirmed

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the conclusion. According to our x-ray diffraction studies of two end compositions of URu_2Al_2 and URu_2Ge_2, however, they do not crystallize into the tetragonal structure, $ThCr_2Si_2$. A similar report has been made on URu_2Ge_2 by other researchers [8]; $U_4Ru_7Ge_6$ is claimed to be the nearest metallurgical phase. It is therefore expected that both doping experiments will show only limited solid solution regions; Ge is found to be single phase up to 40%, but for Al, this shrinks down to 10% or so.

Resistivity measurements have been made with a home-made DC probe. Magnetization has been measured on a vibrating sample magnetometer (VSM, Oxford Instruments, model 3001).

3. Results and analysis

3.1. Resistivity

Resistivity data for Al and Ge alloys are presented in figure 1(*a*) and (*b*), respectively. As far as high-temperature behaviour is concerned, the alloying effects of Al and Ge are comparatively less striking than those produced by alloying on the U site [5]. The single-impurity Kondo behaviour at higher temperatures remains almost intact up to 20% Ge, the highest doping concentration of our alloys that is single phase. At the same time T_{max}, where the resistivity shows a maximum behaviour, decreases slightly.

What is more interesting is found in the low-temperature resistivity (see insets in figure 1). Compared with the previous studies [5, 6] on the U and Ru sites, it is extraordinarily striking that the Cr-like anomaly becomes weak very gradually so that even for 20% Ge the resistivity still shows a clear sign of the Cr-like anomaly. Furthermore, T_N, indicated by the minimum in the resistivity, hardly changes with doping. For comparison, 5% Ir on the Ru site nearly kills the Cr-like anomaly in the resistivity. Such a contrasting effect on T_N may well be due to possibly less effective sp–f hybridization than d–f hybridization. The difference between sp–f and d–f hybridization effects will be further addressed later in section 4.

To allow quantitative analysis of the resistivity, we have used a resistivity formula [9] for gapped (Δ) antiferromagnetism together with a T^2 dependence below T_N

$$\rho = \rho_0 + bT(1 + 2T/\Delta)\exp(-\Delta/T) + AT^2.$$

Fitting results are given in table 1. From the analysis, we have three physically meaningful parameters: the residual resistivity ρ_0, the gap parameter Δ, and the electron–electron interaction term A. Since our URu_2Si_2 is not annealed, ρ_0 for our URu_2Si_2 is twice that for single-crystal URu_2Si_2. However the gap parameter Δ is reasonably in between values for the a and c axes of single-crystal URu_2Si_2. What is most interesting to us is that the gap parameter Δ decreases slightly with substitutions of Al and Ge. Despite some uncertainty over ρ_0 values, we can see ρ_0 increase with doping, but much more slowly than for substitutions on the U site.

3.2. Magnetization

Magnetization curves for pure URu_2Si_2, 5% Al, 7.5% Al, 5% Ge and 20% Ge are given in figure 2. As seen for the resistivity, the curves for the substituted alloys show very little change from that for pure URu_2Si_2. The temperature where magnetization for the substituted alloys shows a maximum behaviour locates near that for pure URu_2Si_2. Not

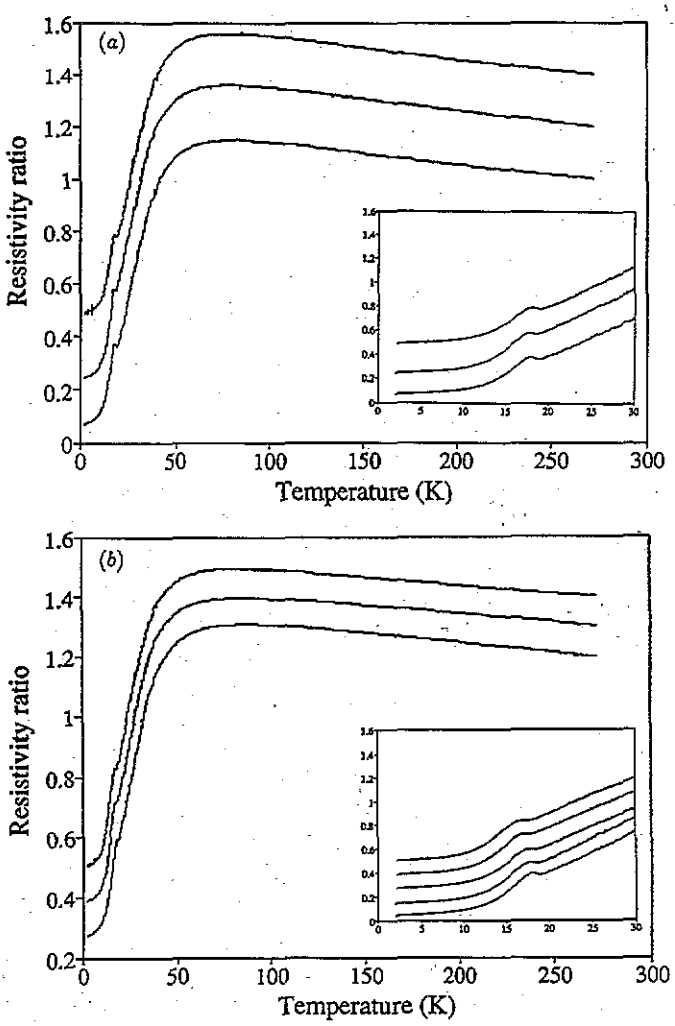

Figure 1. (*a*) Resistivity ratio, normalized to the values at room temperature, for 7.5% and 2.5% Al doped alloys and URu_2Si_2 (from top to bottom). The inset shows the low-temperature resistivity for 7.5% and 2.5% Al doped alloys and URu_2Si_2 (from top to bottom). (*b*) Resistivity ratio for 20%, 15% and 10% Ge doped alloys (from top to bottom). The inset shows the low-temperature resistivity for 20%, 15%, 10%, 5% and 2% Ge doped alloys (from top to bottom). All the data, except for the pure URu_2Si_2 and for 2% Ge at low temperatures, have been displaced upwards.

surprisingly but surely interestingly, the magnetic transition moves slightly towards lower temperatures but is still clearly visible. Therefore both resistivity and magnetization suggest without doubt that the magnetic transition never loses its character even with substitution of 20% Ge. In connection with this observation, it is worthwhile noting that according to specific heat measurements on Ge alloys [7] a peak corresponding to T_N is still visible in the specific heat for the 30% Ge alloy, although slightly broadened. It may be significant to recall that for 5% La doping such a magnetic transition is hardly seen in magnetization although an enhanced Cr-like anomaly is seen in the resistivity and a quite broad structure in the specific heat measurements. For 5% and 20% Ge alloys, the magnetization shows a

Table 1. Curve fitting results, using the resistivity formula for gapped (Δ) antiferromagnetism together with a T^2 dependence, to the low-temperature resistivity (see text).

Systems	ρ_0 ($\mu\Omega$ cm)[a]	b ($\mu\Omega$ cm K^{-1})	Δ (K)	A ($\mu\Omega$ cm K^{-2})
URu_2Si_2 [b]	64±7	340±40	68	0.36±0.04
UrU_2Si_2 [c]	33	800 (a axis)	90	0.17
		52 (c axis)	68	0.10
1% Al	30±3	230±23	67	0.22±0.02
2.5% Al	33±3	370±37	68	0.29±0.03
5% Al	29±3	150±15	62	0.18±0.02
7.5% Al	67±7	210±21	64	0.28±0.03
2% Ge	40±4	340±34	66	0.35±0.04
5% Ge	31±3	360±36	70	0.27±0.03
10% Ge	50±5	310±30	67	0.27±0.03
15% Ge	60±6	220±22	58	0.26±0.03
20% Ge	81±8	190±20	53	0.31±0.03

[a] Uncertainties arise from the irregular shape of our samples.
[b] Our URu_2Si_2 is not annealed.
[c] Data for a single crystal of URu_2Si_2 are taken from [9].

small increase in signal at low temperature which we think originates from a small impurity phase as seen in some single crystals [10].

4. Conclusions

In systematic studies [8] of UT_2Si_2 (T represents transition metals) it is pointed out that the strength of d–f hybridization and thus the degree of delocalization of f electrons are closely related to the number of d electrons at the Fermi level. Therefore systems of transition metals at the right hand side of the periodic table, e.g. UCu_2Si_2, show more localized magnetic character while UFe_2Si_2 etc show no magnetic ordering at all.

In the light of this observation, we would like to address the differences among the effects of substitutions on three different sites of URu_2Si_2, on the resistivity in particular. According to previous studies of alloying on U and Ru sites [5, 6], the Cr-like anomaly is enhanced (as in Cr alloys) by less than 5% of U site doping and is seen up to 20% La, which can be understood if the main role of substitutions on the U site is to induce impurity scattering in the f band responsible for the Cr-like anomaly. However, the Cr-like anomaly is wiped out by approximately 5% Ir or other elements on the Ru site, so it seems to be most sensitive to doping on the Ru site. In contrast to this, doping on the Si site does not produce any comparable effect, except perhaps a dilution effect. These observations seem to imply that the itinerant antiferromagnetic transition occurs by a gap opening in an f band severely delocalized by d–f hybridization, and the strength of such d–f hybridization present in pure URu_2Si_2 is clearly enough to make such a gap possible over the Fermi surface. Alloying on the Ru site modifies this hybridization most effectively, while alloying on the U site merely produces an enhanced Cr-like anomaly by impurity scattering in the f band. However alloying on the Si site modifies the situation in an sp part of the band, which plays little part in the hybridization of the 5f states and thus affects the system only by a simple dilution effect. This is an interesting contrast to Ce compounds in the same structure, e.g. $CeRu_2(Si, Ge)_2$ where a small amount of Ge doping stabilizes an antiferromagnetic transition [11].

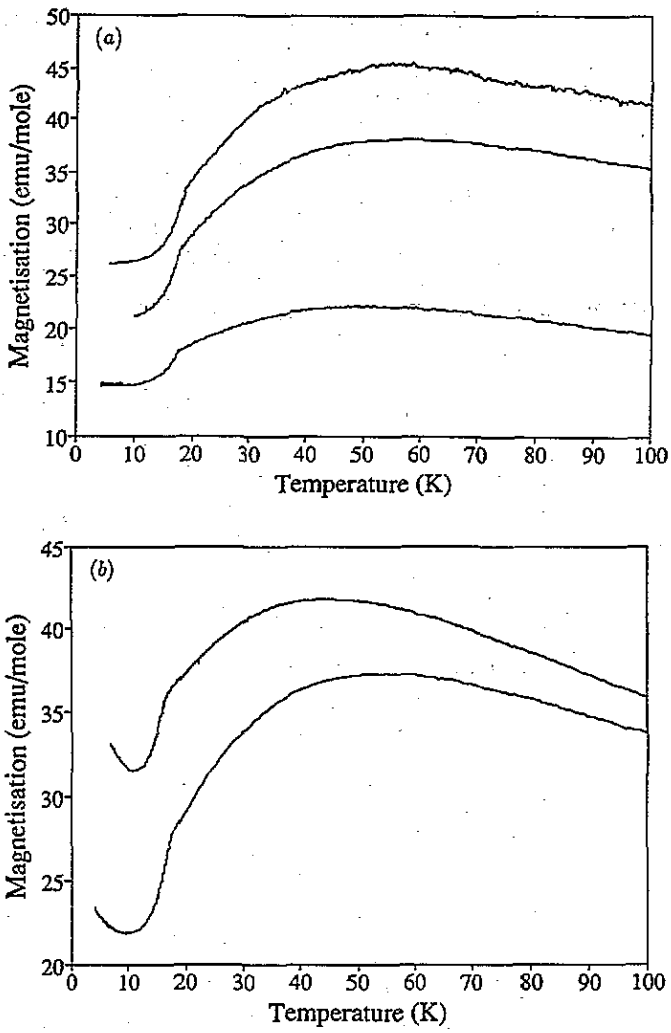

Figure 2. The temperature dependence of magnetization (*a*) for URu_2Si_2 and 5% and 7.5% Al doped alloys (from top to bottom) and (*b*) for 20% and 5% Ge doped alloys (from top to bottom). All data are for a field of 1 T.

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