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Electrical and magnetic studies of URu_2Si_2 with Y, Sc, and Ir substitutions

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Abstract. We report a low-temperature resistivity-minimum anomaly along with the Cr-like one in Y- and Sc-doped systems, as previously seen for heavy rare-earth and Th-doped systems. In the Y alloys, it is found that this low-temperature-minimum anomaly exists even up to 50% Y. However it is easily destroyed by alloying Ir on the Ru site of already Sc-doped samples. For Sc, the Cr-like anomaly moves towards lower temperatures before disappearing at 15% Sc, while we observe that the itinerant antiferromagnetic transition becomes a spin-glass type at 50% Y. The concentration dependence of the various effects varies for Sc, Y, La, and Ce substitutions.

1. Introduction

For some time now URu_2Si_2 has existed as one of a fascinating group of materials, a magnetically ordered and superconducting heavy-fermion compound [1–3]; T_N = 17.5 K and T_c = 1.2 K. In addition to the considerably enhanced electronic contribution to the specific heat, the subgroup of magnetically ordered and superconducting heavy-fermion compounds reveals very interesting features. Although the coexistence of magnetism with superconductivity has been discovered in some Chevrel phases and related ternary compounds [4] before, it is a unique feature of these heavy-fermion compounds that the same electrons, probably strongly hybridized f electrons, are responsible for both magnetic and superconducting transitions. Some groups have shown this clearly by ΔC at T_c, γ, and γ above and below T_N. Apart from the coexistence of magnetism and superconductivity, URu_2Si_2 shows other noteworthy anomalies related to the magnetic transition. First of all, despite an ordered moment as small as 0.03 μ_B, the entropy released below T_N reaches $0.17k_B$ ln 2 per formula unit [3]. Although as far as we know there is no clear understanding of the relationship between the entropy released and the size of the ordered moment, these values still seem to be large. Second, the nature of the magnetic transition itself is intriguing. According to both inelastic neutron scattering [5] and far-infrared reflectance measurements [6], the spin density wave responsible for the 17.5 K magnetic transition has a similar form to that of charge density effects, leading to a speculation that the antiferromagnetic transition is strongly coupled to somewhat charge-related character or is even driven by an electronic instability. In the light of this, it may not be surprising that in spite of such a small ordered moment, the associated effect in the resistivity is a strong Cr-like anomaly [1–3]. In addition, features of URu_2Si_2 worth mentioning include possible quadrupolar interactions [7] and a metamagnetic transition [8].

While most results alluded to are gathered from single-crystal work, we have also seen some useful contributions from alloying experiments to an understanding of the

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underlying physics in URu_2Si_2. To name a few, we may first mention the ferromagnetic instability [9] induced by substitutions of Mn, Re, and Tc; ferromagnetism is rare in heavy-fermion compounds. Second, Si-site alloying experiments [10] reflect that sp–f hybridization (affected most by alloying on the Si site) is not so significant in forming the antiferromagnetic ground state while related studies using Ge in $CeRu_2Si_2$ [11], another heavy-fermion compound, induce a rapid appearance of an antiferromagnetic ground state in non-magnetic $CeRu_2Si_2$. Finally, there is a number of U-site alloying experiments [12, 13] that strongly suggest that the maximum behaviour in both resistivity and magnetic susceptibility measurements originates mainly from short-range magnetic fluctuations. We would like to emphasize however that this interpretation does not rule out contributions from crystal field excitations completely. During studies of U-site alloying experiments, later extended by us to most of the possible doping elements, it has been found by two groups independently that in addition to the enhanced Cr-like anomaly another minimum behaviour appears in the resistivity at low temperatures (below the magnetic transition temperature) [14, 15]. Although microscopic measurements will be needed in order to understand the anomalous minimum in detail, it is tempting to recall that the spin density wave responsible for the 17.5 K transition is strongly coupled to charge density effects. Perhaps different alloying effects on two density waves may be plausible [16].

In this paper, we show that this anomalous minimum behaviour is also seen in Y- and Sc-doped alloys. Moreover, since this minimum is seen in Y-doped samples up to 50%, we can probably rule out the possibility that it is an impurity Kondo hole effect, as suggested by one of its discoverers [14]. In continuing efforts to elucidate the origin of this minimum behaviour, we have found that its existence is sensitive to the Ru-site doping. To make our presentation more systematic, we discuss all data for $(U, Y)Ru_2Si_2$ first and then move on to $(U, Sc)(Ru, Ir)_2Si_2$.

2. Experimental details

Constituent elements were melted together on a water-cooled Cu hearth under an Ar atmosphere. All samples, encapsulated in evacuated quartz tubes, were annealed at 600 ° for 2 days and then 800 °C for 5 days. Subsequent metallographic examinations show that all samples are single phase. X-ray diffraction studies using Cu $K\alpha$ radiation (Philips PW1710) have also been made of all Y-doped samples and 4% Sc-, 6% Sc–5% Ir-, and 15% Sc-doped samples. According to these, there appears only a very small decrease in the unit cell volume with increasing Sc concentration. For the Y-doped samples, however, we have not seen any change in the unit cell volume within experimental error while the a-axis (c-axis) lattice expands (contracts) respectively.

Resistivity measurements have been made using a home-made DC four-probe rig. A vibrating sample magnetometer (VSM3001, Oxford) has been used for magnetization measurements.

3. Experimental results

3.1. $(U, Y)Ru_2Si_2$

Resistivity data for $(U, Y)Ru_2Si_2$ are presented in figure 1. As reported before, URu_2Si_2 shows three characteristics: a negative temperature coefficient behaviour at high-temperature regions, a maximum around 80 K, and the Cr-like anomaly at 17.5 K.

With 5% Y, two interesting results appear. One is related to the Cr-like anomaly. The Cr-like anomaly, which is much more pronounced for the same amount of La or Ce [13, 15],

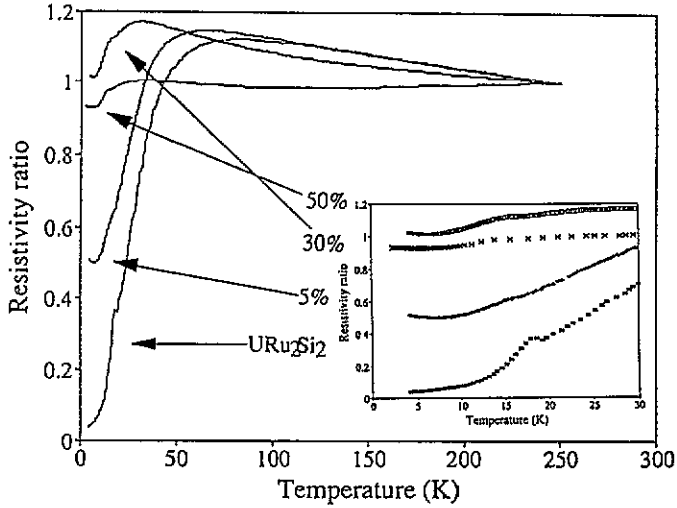

Figure 1. Resistivity ratios, $R(T)/R(270\ \mathrm{K})$, are presented for URu_2Si_2 and three Y-doped samples. The inset shows an expanded view of low-temperature resistivity ratios for the same samples.

is becoming weak and looks like a shoulder now. The apparent transition temperature has moved very slightly, compared with the La- or Ce-doped systems. Below the shoulder-like structure originating from the magnetic transition, we have an additional minimum around 6 K. It may be worth recalling that a similar minimum has been reported at almost the same temperature for heavy rare-earth and Th-doped samples [14, 15]. At higher temperatures, the negative temperature coefficient behaviour is still visible.

For a 30% Y-doped sample, the residual resistivity shows a large increase due to impurity scattering. Apart from the relatively large increase in the residual resistivity, generally speaking the resistivity for 30% Y shows little differences from that for 5% Y; both show a magnetically originated shoulder, a low-temperature minimum, and a negative temperature coefficient behaviour at high temperatures. The temperatures where the shoulder and minimum appear hardly change from those for 5% Y. It is interesting to note however that the temperature where the resistivity shows a marked fall decreases from 65 K for 5% Y to 30 K for 30% Y. It may also be worth mentioning that the negative temperature coefficient behaviour is slightly more apparent in 30% Y than in 5% Y.

On increasing the Y concentration up to 50%, neither the minimum nor the shoulder-type structure disappears. The temperature where a higher-temperature maximum appears in the resistivity is around 30 K, as in the 30% Y-doped sample. On the high-temperature side, the negative temperature coefficient effect is so much weaker that a minimum even appears around 120 K. This general trend seen in the negative temperature coefficient behaviour seems to suggest that a single Kondo temperature, T_K = 370 K, deduced from high-temperature resistivity data [17] for URu_2Si_2, may decrease enormously with increasing Y concentration. Interestingly enough, however, we have seen virtually no change in γ values, when normalized per mole U, between URu_2Si_2 and 30% Y. We will discuss this particular point in detail later.

Our magnetization data for URu_2Si_2 and all Y-doped samples are presented in figure 2. From the magnetization for pure URu_2Si_2, we can see a maximum around 55 K and a maximum in the second derivative of magnetization around the magnetic transition temperature. With 5% Y doping, there is little change except for a severely smeared feature

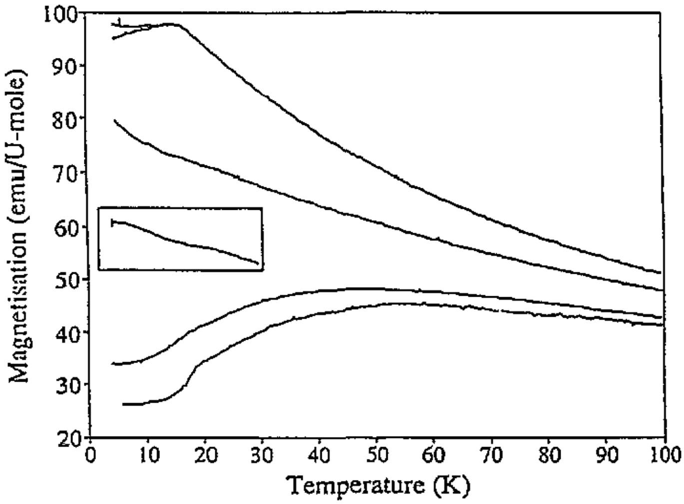

Figure 2. The magnetization measured with 1 T applied field along the sample axis as a function of temperature with URu_2Si_2 and three Y-doped samples: from top to bottom, 50, 30, and 5% Y-doped samples and URu_2Si_2. The inset shows low-temperature data of magnetization for 30% Y measured with 4 T applied magnetic field (normalized to 1 T and shifted downwards).

around the magnetic transition with a maximum around 50 K. Within the experimental resolution, we do not see any new feature around the temperature where the low-temperature minimum appears. For 30% Y in 1 T, the maximum seen in the magnetization for URu_2Si_2 and $(U_{0.95}Y_{0.05})Ru_2Si_2$ disappears. Furthermore, we do not see any feature that could correspond to the anomalies seen in the resistivity: the shoulder-type transition and the low-temperature minimum. The magnetization measured in 4 T on cooling, however, seems to show a tendency to yield plateaus in both the temperature region around 17.5 K and that where the low-temperature resistance minimum occurs, but in both regions $M(H)$ is almost linear. Therefore we think that the reason why we could observe some plateaus with 4 T but not with 1 T is not a magnetic-field-induced transition but a better signal-to-noise ratio.

For 50% Y, the magnetization shows evidence for short-range magnetic correlations at low temperatures. In fact, the short-range magnetic correlations lead to a zero-field cooling and field cooling effect around 15 K, which is where the resistivity shows a shoulder. This kind of magnetic memory effect has often been regarded as an earmark of spin-glass behaviour. Therefore our magnetic studies of Y-doped samples show that there is a cross-over from itinerant antiferromagnetism in URu_2Si_2 to a spin-glass type transition in $(U_{0.5}Y_{0.5})Ru_2Si_2$.

We have also measured $M(H)$ up to 8 T at different temperatures for three Y-doped samples. For the analysis of magnetization we have used a formula, $M = \chi_1 H + \chi_3 H^3/3!$. What is interesting to us is that for 30% Y the non-linear susceptibility, χ_3, is negative at three different temperatures (4.2 K, 14 K and 68 K) while the non-linear susceptibility for 5% Y is negative at 4.2 K but recovers to positive values at 14 K. In pure URu_2Si_2, the non-linear susceptibility along the *c*-axis is positive below well above T_N and has a sudden jump at the magnetic transition temperature, while it is almost temperature independent along the basal plane [18].

3.2. $(U, Sc)(Ru, Ir)_2Si_2$

For these studies, we have prepared several alloys with compositions such as 4% Sc, 4%

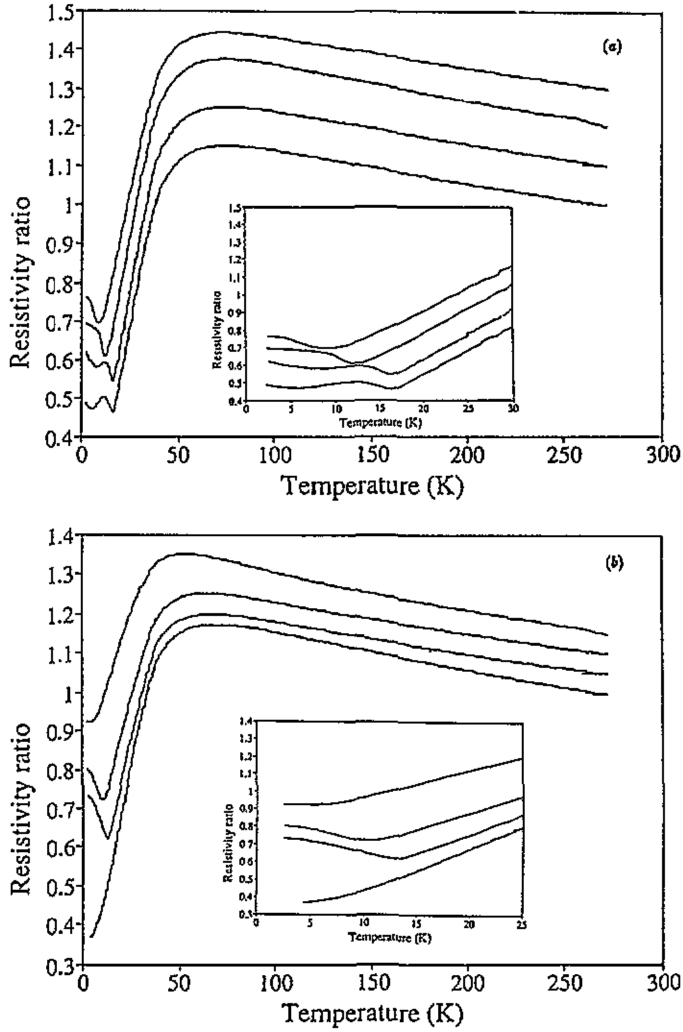

Figure 3. Resistivity ratios, $R(T)/R(270\ \mathrm{K})$, are presented (a) for 5% Sc–2.3% Ir, 4% Sc–2% Ir, 4% Sc–0.43% Ir, and 4% Sc (from top to bottom) and (b) for 6% Sc–5% Ir, 10% Sc, 7.5% Sc, and 15% Sc (from top to bottom). The insets show low-temperature data for the same samples. For the sake of clarity, some data are shifted upwards.

Sc–0.43% Ir, 4% Sc–2% Ir, 5% Sc–2.3% Ir, 6% Sc–5% Ir, 7.5% Sc, 10% Sc, and 15% Sc.

Resistivity data for the samples are presented in figure 3. Notably 4% Sc shows two clear anomalies at low temperatures: one is an enhanced Cr-like anomaly and the other a low-temperature minimum. Unlike other systems showing the two anomalies, the features around T_N looks very much like what we saw in $(U_{0.95}La_{0.05})Ru_2Si_2$ [13], instead of a weak shoulder-like structure, a clearly exaggerated Cr-like anomaly is visible. Below it, however, a small minimum appears additionally as in 5% Y but not 5% La. With increasing Sc concentration we first lose the low-temperature minimum between 4% and 7.5% Sc but still have the Cr-like anomaly. It is to be noted though that the two anomalies have been observed in even a 50% Y-doped sample. Further increase in Sc concentration to 10% moves the Cr-like anomaly towards lower temperatures, and for 15% Sc it finally disappears. Throughout the Sc doping, the temperatures where the resistivity shows a maximum behaviour change only very slightly, but the Sc concentrations do not extend up to those where the Y alloys showed marked changes.

To explore a possible correlation between the two anomalies below 20 K, we doped Ir on the Ru site of already Sc-doped samples. From the previous studies with Ir alone [19], it is known that more than 5% Ir doping washes out the Cr-like anomaly completely.

For 4% Sc–0.43% Ir, the Cr-like anomaly decreases slightly in temperature and the low-temperature minimum becomes less noticeable, compared with 4% Sc. On increasing the Ir concentrations to 2% but keeping the Sc concentration constant at 4%, the resistivity no longer shows the low-temperature-minimum behaviour. At the same time, the Cr-like anomaly moves towards lower temperatures. On increasing the Ir concentration further, the Cr-like anomaly moves gradually towards lower temperatures without losing its shape much before disappearing in the most concentrated Ir-doped sample, 6% Sc–5% Ir. This is consistent with the previous studies using primarily Ir [19].

In our magnetization measurements, we have seen the rounded maximum behaviour even with 15% Sc at almost the same maximum temperature as in pure URu_2Si_2. In the light of previous studies [12, 13, 15], this suggests that short-range magnetic fluctuations might well be present in a 15% Sc-doped sample. We did not see any clear signs of the magnetic transition (SDW) temperature in any Sc-doped sample in magnetization measurements, except for the 4% Sc sample, but this has been seen to be a subtle effect in all alloys.

4. Discussion

In our Y-doped samples, two anomalies in the resistivity at low temperatures, the low-temperature minimum and the Cr-like anomaly, are remarkably insensitive to Y concentration. In figure 4, we plot magnetic transition temperatures for Y-doped samples along with those for La-, Ce-, and Sc-doped samples. From figure 4, it is clear that the magnetic transition temperatures for Y-doped samples are almost concentration independent, compared with those for systems doped with other elements. However we should mention that the nature of the magnetic ground state changes from an itinerant antiferromagnetism in URu_2Si_2 to a spin glass at 50% Y.

The temperature where the resistivity shows a maximum, however, decreases with increasing Y. The higher the Y concentration, the less pronounced is the negative temperature coefficient behaviour at high temperatures, so for 50% Y the resistivity shows a minimum around 110 K. In connection with this, the temperature where the resistivity shows a rounded maximum seems to move down in value as the Y concentration increases, suggesting a decrease in short-range ordering effects, and the negative slope of $d\rho/dT$ above it decreases. Both of these effects could suggest a smaller contribution of Kondo scattering to the total resistivity as the U content decreases. The specific heat of the 30% Y alloys [20] suggests however that γ per mole of U above the magnetic transition is close to that of pure URu_2Si_2, so the Kondo temperature of impurity scattering for 30% Y may be the same as that for URu_2Si_2. The data are shown in figure 5 and it would be useful to have them for the 50% Y alloy. The value of C/T for the 30% Y alloy at the lowest temperatures is appreciably larger than that for pure URu_2Si_2 (it is also larger than C/T extrapolated from above T_N), suggesting a smaller loss of Fermi surface on magnetic ordering, but the temperature dependence is somewhat anomalous and one must expect complication when magnetic excitations as well as single-particle excitations are present.

Although the resistivity shows only slight changes from one Y-doped sample to another, the magnetization reveals that the itinerant antiferromagnetic ground state in URu_2Si_2 changes its character, becoming spin-glass type at 50% Y. It is also worth while mentioning

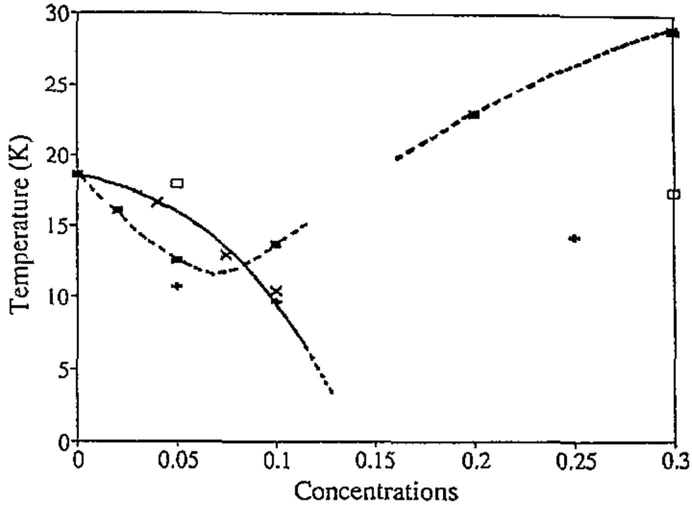

Figure 4. The temperatures where the resisitvity shows the Cr-like anomalies are presented for Y (□), Sc (×), La (■), and Ce (+). The solid and dashed lines are guides for Sc and La respectively.

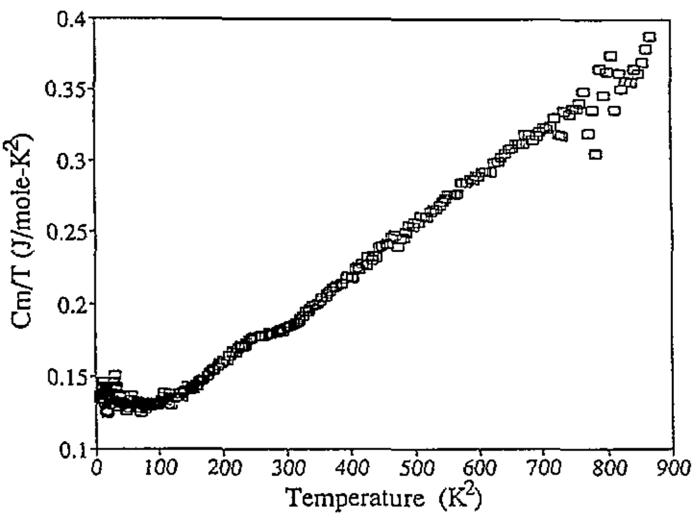

Figure 5. The magnetic specific heat is presented for 30% Y; $C_m = C((U_{0.7}Y_{0.3})Ru_2Si_2)$-$C(YRu_2Si_2)$.

that the unit cell volume hardly changes up to 50% Y while the a-axis (c-axis) expands (contracts) with increasing Y. Since it is well known that anisotropy is an important character in most physical properties of URu_2Si_2, we think Y-doped systems deserve further study. However, it is far beyond our speculation how the different behaviour of a a- and c-axes under Y doping affects magnetic ground states, leading to a spin-glass-type transition.

In alloying experiments with both Sc and Ir, the low-temperature-minimum behaviour induced by small doping of Sc (around 4%) weakens rapidly with increasing Ir, and disappears between 0.43% and 2% Ir before the magnetic transition is destroyed by more than 5% Ir doping. The persistent presence of Cr-like anomaly in the resistivity of relatively Sc-rich samples suggests that the magnetic ordering character seen in most of the Sc-doped alloys might not be very different from that of URu_2Si_2. Concerning the magnetic transition temperature on substitution of Sc, figure 4 reveals an interesting point. In contrast to the La- or Ce-doped cases, Sc doping gradually reduces the magnetic transition temperature before destroying it completely between 10% and 15% Sc doping.

To conclude, we have studied the effects of Y and Sc–Ir doping in URu_2Si_2. In these studies, the anomalous low-temperature minimum is visible up to 50% Y, which makes a 'Kondo hole' explanation rather unlikely. We have also found a spin-freezing behaviour in a 50% Y-doped sample. Additions of Ir to the Sc-doped alloys remove the low-temperature resistance minimum as well as lowering the temperature of the Cr-like anomaly still further.

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