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Electrical and magnetic properties of $(U_{1-x}R_x)Ru_2Si_2$ alloys

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Abstract. Comprehensive studies have been made of the effects of substitutions at the U site in URu_2Si_2 of all the rare-earth metals, except Pm, Eu and Yb, and of Th. Attention has been focused on the changes in the magnetic character produced by La and Ce, on the different effects on the 17.5 K resistivity anomaly and on the appearance in some of the dilute alloys of a low-temperature resistivity minimum. The non-linear susceptibility of La-doped alloys has been studied. The possible roles of short-range magnetic order and crystal field splitting are discussed in connection with the maxima in the resistivity and susceptibility of URu_2Si_2.

1. Introduction

Of the heavy-fermion intermetallic compounds, URu_2Si_2 is of particular interest due to its possession of antiferromagnetic order of itinerant character below 17.5 K, coexisting below 1.2 K with superconductivity [1–3]. Although there is a strong dependence on the detailed composition of the samples, related behaviour has been reported for UPt_3 [4] and $CeCu_2Si_2$ [5], and $U_{0.97}Th_{0.03}Be_{13}$ [6] seems to develop some magnetic character at a second transition below that to superconductivity. Very recently behaviour rather like that of URu_2Si_2 has been discovered in UNi_2Al_3 and UPd_2Al_3 [7, 8], although the latter has a much larger ordered moment than the value of an order of $0.01\mu_B$ per f-element atom. The moment of $0.03\mu_B$ for URu_2Si_2 is a particular puzzle since the entropy associated with the magnetic transition approaches R, and quadrupolar character has been suggested [9] on the basis of measurements of the non-linear susceptibility. Furthermore, optical studies suggest anomalous behaviour of the charge density at this transition [10].

A number of types of measurement lead to the conclusion [2] that a spin-density wave setting in at 17.5 K opens a gap over as much as 40% of the Fermi surface. Neutron studies of Broholm *et al* [11] indicate a correlation with a charge-density wave and show a type I antiferromagnetic structure with the moments along the c-direction. It also seems that the U is a non-Kramers ion with a singlet–singlet separation that may account for the broad maximum in the susceptibility and excess specific heat. The maximum in the resistivity in the same range of temperature may also be associated with crystal field effects, but it may also be related to short-range spin correlations preceding long-range order, and such fluctuations have been observed in the neutron scattering.

Other studies of URu_2Si_2 have been concerned with the effects of pressure [12], the high-field metamagnetic transitions [13] and with the character of the superconducting state [14, 15].

A number of investigations have also been made of the effects of alloying substitutions, mainly on the Ru sites. These [16] have revealed ferromagnetic instabilities for Mn, Re

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and Tc, which are rare for heavy-fermion systems. The Cr-like anomaly in the resistivity is almost destroyed [17] by substitutions of less than 5% of these elements. This contrasts with the enhancement of this effect for substitutions of La or Ce on the U site [18], which is reminiscent of the behaviour observed in alloys of Cr [19] and α-Mn [20].

In view of this insights provided by studies of systems like (La, Ce)Cu_6, where one can move from dilute to concentrated Kondo systems and finally to a Kondo lattice, it seemed desirable to study URu_2Si_2–XRu_2Si_2 systems where ranges of solid solubility exist. This is possible for X = La, Ce and Th, although in none of them is there a complete range of solid solutions. We have also examined dilute substitutions of most of the rare-earth elements in an attempt to shed light on the anomalous behaviour of the resistivity below T_N reported for the Th alloys [21].

2. Experimental methods

Specimens were prepared by arc melting together metals of nominal 4N purity and suction chill casting into a copper mould to produce square cross section rods of 0.3 cm × 0.3 cm × 4 cm. All the La and Ce alloys were annealed first at 600 °C then for 3–5 days at 8)) °C. They were examined metallographically and by x-ray diffraction and the solubility limits are estimated to be 35% for La and 45% for Ce. The limit for Th as been quoted [21] as 10%.

Other samples were prepared with 4.4% Pr, 3.8% Nd, 5% Sm, 5% Gd, 5.3% Tb, 5.1% Dy, 7.6% Ho, 5% Er, 5.6% Tm, 2% and 5% Lu and 5% Th. These were examined only by metallographic methods, and appear (as one would expect from the atomic sizes and small doping levels) to be single phase.

Resistivity measurements were made with a conventional four-probe DC method, and because of dimensional uncertainties the absolute resistivity values are probably only correct to about ±10%. Magnetization measurements up to 8 T and DC susceptibility measurements (at 1 T) were made with an Oxford Instruments vibrating sample magnetometer, and AC susceptibilities at 320 Hz in a standard type of apparatus with primary coil fields of (0.7–1.4) × 10^{-4} T.

3. Results and analysis

Most of the data described below are new, but we include for completeness some of the results for the La and Ce alloys which have already been briefly presented [18, 22]. Rather than dealing separately with all results for a given alloy system it seems more efficient to deal in turn with the resistivity, the susceptibility and the magnetization data for all the systems.

3.1. Resistivity

In pure URu_2Si_2, four aspects of the resistivity behaviour are of interest and we have sought to explore their variation on alloying. These are the high temperature Kondo-like negative temperature coefficient (NTC), the marked fall in resistivity below a rounded maximum at around 70 K, the chromium-like anomaly associated with the antiferromagnetic ordering

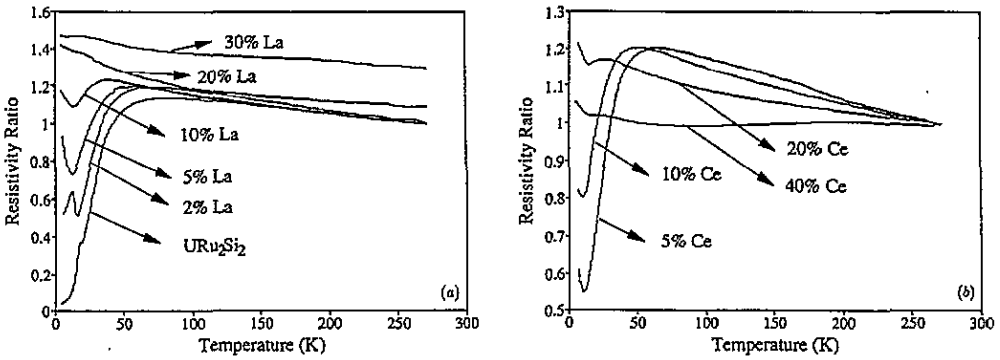

Figure 1. Resistivity ratio with respect to values at room temperature; (a) for La alloys (graphs for 20 and 30% La are displaced upwards for clarity), (b) for Ce alloys.

and the behaviour in the lowest temperature range where indications of the character of the excitations may be obtained.

Results for La and Ce alloys are shown in figures 1(a) and (b). Since all our dilute rare-earth alloys show little change in the higher-temperature resistivity, we do not present them here. However, we will discuss the low-temperature resistivity in detail later. From figure 1(a) and (b)m, it is clear that the NTC range is little affected by alloying except for the most concentrated Ce specimen, where the data shown in figure 2 suggest that crystal field splitting of Ce 4f configuration is giving the type of two-Kondo-regimes behaviour of the type discussed for other Ce systems by Cornut and Coqblin [23]. The ratio we find for the slopes of the two logarithmic regions is 3/11, which is not greatly different from 3/15 which would be appropriate for temperatures up to about room temperatures for the crystal field levels reported [24] for pure $CeRu_2Si_2$. It should be noted that the actual energy splitting between the ground state and the first excited state decreases markedly from 35 meV for $CeRu_2Si_2$ to approximately 10 meV for $(U_{0.6}Ce_{0.4})Ru_2Si_2$. (The lower temperature increase, as we shall see, can be associated with the magnetic ordering.)

The 70 K maximum and the fall below it is not capable of a simple interpretation, even for pure URu_2Si_2. There must clearly be effects associated with the sinking of the 5f configuration into its singlet ground state, with the loss of any spin disorder or inelastic scattering, but it is not clear whether, while Kondo character remains, there is some incipient onset of coherence of the type characteristic of simpler heavy-fermion compounds. Furthermore, both the susceptibility data and neutron results [11] suggest a regime of short-range magnetic order above T_N and as suggested by Broholm *et al* [11] this may be responsible for the maximum at 70 K in the resistivity data in part. This regime is little affected by small substitutions for U, but in the alloys with higher amounts of La or Ce is greatly modified. However, for the 30% La alloy (as we shall see later) the onset of magnetic order at a much higher temperature may be reducing the range in which crystal field, short-range fluctuations or coherence effects could be expected to manifest themselves in the resistivity. On balance, that the 70 K maximum decreases with substitutions of La and Ce and leads to a magnetic transition seen in magnetization is in favour of short-range fluctuations as the origin of the 70 K maximum.

The anomaly in pure URu_2Si_2 at 17.5 K ascribed to antiferromagnetic order is greatly affected by small alloying substitutions of La and Ce, and it can be used to show the variation of T_N with alloy concentrations [22]. For less than 10% of each of them, particularly La, we have seen enhancement of the sort seen in Cr [19] and α-Mn [20] alloys and which can

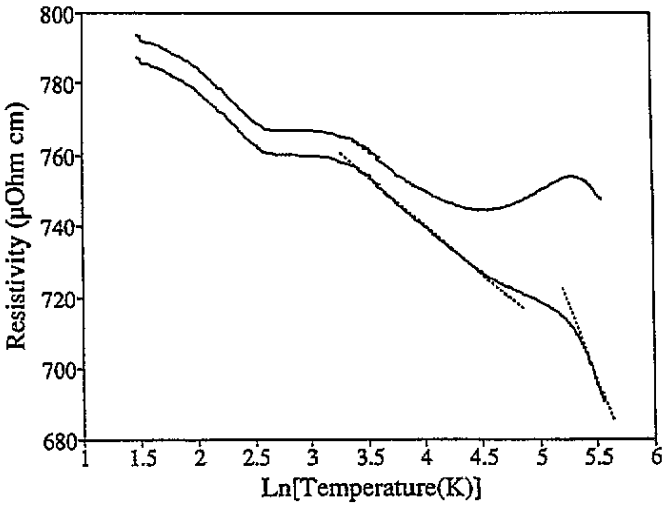

Figure 2. Blown-up picture of ρ versus ln T for 40% Ce; the upper graph is raw resistivity and the lower one resistivity subtracted off by $\rho(LaRu_2Si_2)$. Two dotted lines show Kondo behaviour regions (see the text).

be ascribed to enhancement of impurity scattering by the loss of Fermi surface. This kind of enhancement seen in the resistivity for less than 10% of La and Ce along with other measurements [10, 11] also supports the idea that the magnetic transition at 17.5 K has an itinerant character, somewhat related to the Fermi surface topology, and perhaps correlated to charge-density effects. In spite of the similarity seen in the resistivity for less than 10% of La and Ce, rather different low-temperature behaviour appears for more than 10%. With increasing concentrations, the low-temperature resistivity is modified significantly losing the Cr-like anomaly. However, the increasing resistance below the magnetic ordering temperature in the 20 and 40% Ce alloys resembles that of α-Mn with 5% Cr and 5% Fe [20].

The low-temperature resistivity for dilute rare-earth and Th alloys are shown in figure 3(a) and (b). What is interesting is that some of alloys show two anomalies rather than a single enhanced Cr-like anomaly seen for La and Ce alloys. To show this point more clearly, we have used curve fitting to $\rho(T)$ and present $d\rho/dT$ in figure 3 (c) and (d). In the alloys with heavy rare earths (from Sm on) the size of the resistivity anomaly at T_N is greatly diminished, as it is with the Th alloys [21], but examination of $d\rho/dT$ (as shown in figure 3 (c) and (d)) gives fairly clear indications of a minimum in the 14–17 K region which we ascribe to a residual T_N anomaly; the specific heat anomaly associated with T_N is significantly broadened for both Th and La [21, 25, 26]. The temperatures of this anomaly for such alloys are therefore also shown by square points in figure 4, and similar symbols are used for the minima seen for the Pr and Nd alloys, which show no $d\rho/dT$ effects in the 14–17 K region and which therefore seem to resemble alloys with 5% La and Ce.

However, in the alloys with heavy rare-earths and with Th a new type of resistivity minimum seems to be appearing at lower temperatures, and the temperature of this is shown by crosses in figure 4. It may be significant that this effect appears only in the alloys were the T_N anomaly is greatly attenuated but little changed in temperature from that in pure URu2Si$_2$, and does not appear in the La, Ce, Pr, Nd and Lu alloys where T_N is greatly depressed by approximately 5% substitutions but is very clearly seen in $\rho(T)$

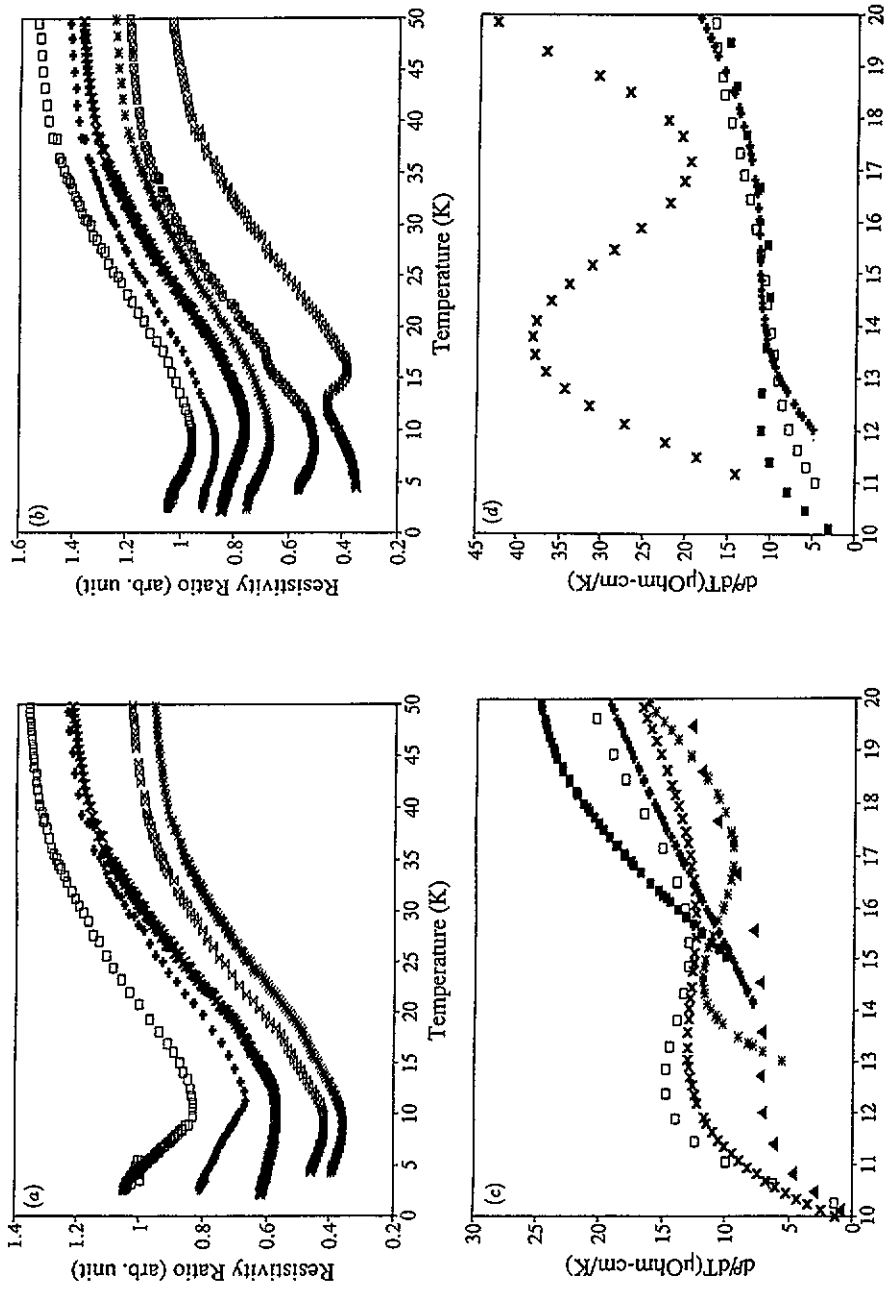

Figure 3. Blown-up picture of low-temperature resistivity. (a) from top to bottom, 4.4% Pr, 3.8% Nd, 5% Sm, 5% Gd, and 5.3% Tb, (b) from top to bottom, 5.1% Dy, 7.6% Ho, 5% Er, 5.6% Tm, 5% Th, and 2% Lu. The temperature derivative of resistivity ($d\rho/dT$) (c) 4.4% Pr (■), 3.8% Nd (+), 5% Sm (∗), 5% Gd (□), 5.3% Tb (×), and 5.1% Dy (▲), (d) 7.6% Ho (■), 5% Er (+), 5.6% Tm (□), and 5% Th (×) (see the text).

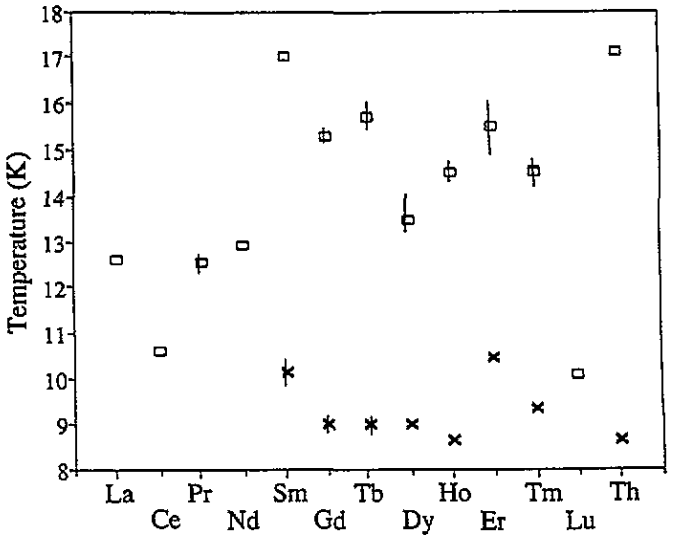

Figure 4. Transition temperatures from resistivity for 5% La, 5% Ce, 4.4% Pr, 3.8% Nd, 5% Sm, 5% Gd, 5.3% Tb, 5.1% Dy, 7.6% Ho, 5% Er, 5.6% Tm, 5% Lu, and 5% Th (see the text).

as in the Cr and α-Mn alloys. This would seem to imply that in the Th and heavy rare-earth alloys T_N is little changed but the spin ordering has a much weaker effect on the Fermi surface. The fact that in a 5% Th alloy [21] the specific heat and susceptibility continue to suggest spin ordering at a temperature little changed from T_N of pure URu_2Si_2, while the resistivity anomaly at that temperature is greatly diminished, lends support to our suggestion (supported by the analysis, see figure 3, of the $\rho(T)$ curves) that heavy rare earths are producing similar effects.

The new type of resistivity minimum at low temperatures in this group of alloys is that which, for Th alloys, was ascribed [21] to Kondo hole behaviour, that is, the effect of a missing Kondo atom in a Kondo lattice. Since the archetypal Kondo resistance minimum is associated with spin-dependent scattering from an impurity which is not yet coupled to other such atoms (and is *en route* to compensation by conduction electron antiparallel polarization) it seems surprising that a Kondo hole should produce similar effects in a lattice where some type of long-range order has already set in. However, it may be recalled that a Kondo-type resistance minimum of the sort seen for Fe in Mo [27] is seen also for Fe in iso-electronic Cr [28] in spite of the spin-density-wave ordering that the host has already undergone at a much higher temperature. The analogy cannot be pressed too far, however, since in our alloys the minimum below T_N is observed when the resistivity anomaly at T_N is greatly reduced, and not when (as for La, Ce and Lu) the latter effect is enhanced, while for the CrFe alloys the low-temperature minimum accompanies an enhancement of the T_N anomaly.

It should be recalled that dilute solutions of U in $ThRu_2Si_2$ do not show a resistivity minimum [29] but a behaviour more like the inverted Kondo effect seen in RhFe [30] alloys. However, in unpublished work of ours on dilute alloys of U in other XRu_2Si_2 (X = Y, La, and Th) hosts crystal field effects seem to be operating and at lower temperatures for concentrations above 5% U interaction effects. Amitsuka *et al* [29] interpret their susceptibility data in terms of a multichannel Kondo effect, but that will not explain the resistivity behaviour.

3.2. Susceptibility results

3.2.1. La and Ce alloys. In view of the fairly weak susceptibility of URu_2Si_2, the susceptibilities of the alloys with Ce and La were explored by DC measurements, in a field of 1 T. The results are shown in figure 5 as magnetization in that field (in emu $mole^{-1}$). Since the susceptibility of URu_2Si_2 is known [1] to be highly anisotropic, with significant temperature variations only for χ parallel to the c-axis, the absolute values for our polycrystalline samples are not very significant, but we note that the $\chi(T)$ for our pure sample is very similar to that reported for $\chi_{\|}$, so that significant preferred orientation is clearly present. Variations in the degree of preferred orientation may account for the variations with Ce or La concentration of the value we measured at 100 K, and since we are concerned only with the character of the temperature variation we have presented the results in figure 5 after adjusting the 100 K values to lie on a smooth (almost linear) curve. Only for 10% La and 40% Ce samples is there any change in the order of the 100 K values from those measured.

For the 5% and 10% alloys of both La and Ce the character of $M(T)$ is similar to that of the pure compound, with dM/dT taking its maximum value at a temperature close to that suggested for T_N by the resistivity data. The rounded maximum at higher temperatures becomes less pronounced, however, and moves to lower temperatures. For the alloys with 20% and 30% La and with 25% and 40% Ce however, the higher-temperature maximum disappears and the susceptibility has more of character of a simple antiferromagnet or perhaps a spin glass, although we have not detected field-cooling effects. For the La alloys this is in accord with the conclusion of Roy *et al* [18] and Amitsuka *et al* [26] that the antiferromagnetism has a different character in the more concentrated alloys. The specific heat data [26] for a 30% La alloy show a behaviour more like that of a spin glass, and preliminary neutron scattering results [31] on our 25% Ce alloy, while showing signs of magnetic short-range order, do not give clear indications of long-range order.

One striking feature of the results is that the temperature of the susceptibility peak is still rising with La content at 30% La, while that for 40% Ce is below that for 25% Ce. This suggests that, in spite of the moments carried by Ce (which show themselves in the larger Curie–Weiss term), the value of T_N is raised in the La system by the expansion of the lattice. This recalls the ability of La substitution to induce a long-range order in $CeRu_2Si_2$ by lattice expansion [32]. In connection to this, it is interesting to note that although there is strong similarity in alloying effects by less than 10% of La and Ce, including the behaviour of T_N and the enhanced Cr-like anomaly in ρ, resistivity for more than 10% of each of them shows rather different behaviour.

3.2.2. Non-linear susceptibilities. Attention has been drawn [9] to the usefulness of non-linear susceptibility studies as a probe of heavy-fermion materials, and we have therefore examined the deviations from linearity with H of the magnetization at low temperature (4.2 K) of some of our alloys. The effects we observe can be understood in a fairly straightforward manner, and correlate with those observed in a single crystal of URu_2Si_2 [9].

In the alloys with moment-bearing impurities the behaviour follows from the negative curvature of the Brillouin function, which gives negative values of χ_3 in the expansion of M in odd powers of H. Since URu_2Si_2 has a metamagnetic transition in high fields, the value of χ_3 is positive below T_N, and if it were a simple antiferromagnet the sign would change to negative on heating through T_N. This is, in fact, what we see in our $(U_{0.7}La_{0.3})Ru_2Si_2$ sample where, as we have seen (figure 5), the susceptibility is that of a conventional antiferromagnet. For pure URu_2Si_2 however, we have suggested that antiferromagnetic

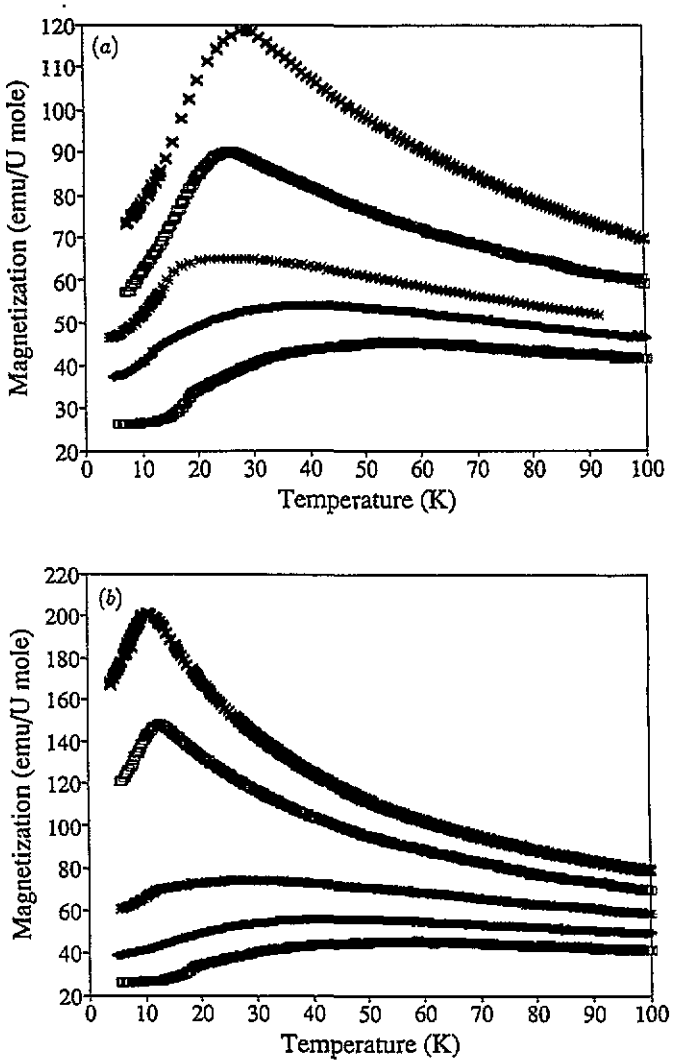

Figure 5. (a) Magnetization for La alloys: from top to bottom, 30%, 20%, 10%, 5% La and URu_2Si_2. (b) Magnetization for Ce alloys: from top to bottom, 40%, 20%, 10%, 5% Ce and URu_2Si_2. (Data have been scaled to give monotonic $\chi(x)$ at 100 K; see the text.)

correlations set in well above T_N, producing the prominent rounded maximum in the c-axis susceptibility and reducing at T_N to only 2/3 of its maximum value; in competing with these correlations an applied field will again produce a magnetization increasing more rapidly than linearly, χ_3 remaining positive above T_N as observed [9]. The results for two La alloys are shown in figure 6. It is interesting to compare the behaviour of the 10% and 30% alloys with that of the pure compound; at 10% the short-range correlations seem to have been somewhat depressed and T_{max} more than halved, but the character of the long-range order has not changed; consequently χ_3 remains positive for some way above T_N and by $2T_N$ has only just reached negative values. In the 30% alloy, however, where neither χ nor ρ suggest significant short-range correlations, χ_3 shows more conventional behaviour, becoming negative at about $1.25T_N$.

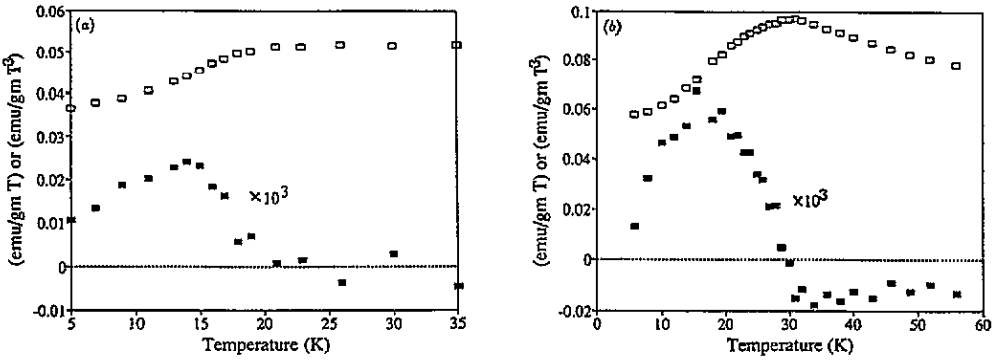

Figure 6. Linear susceptibility χ_1 (□) and non-linear susceptibility χ_3 (■) (a) for 10% La and (b) for 30% La.

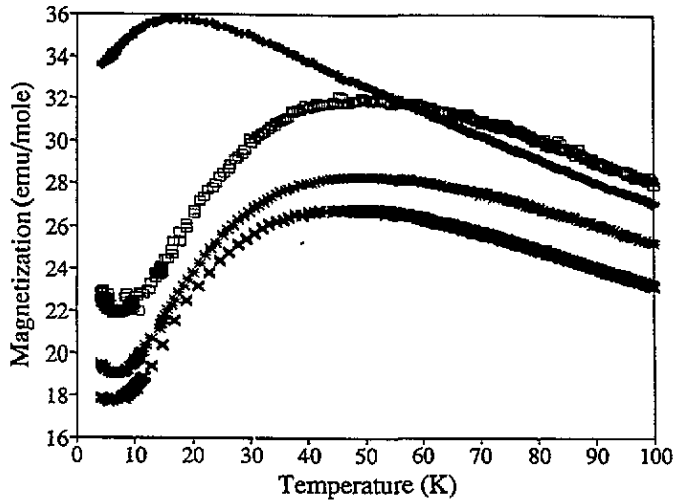

Figure 7. Magnetization for light rare-earth alloys; 4.4% Pr (+), 5% Sm (*), 5% Lu (□) and 5% Th (×).

3.2.3. Dilute alloys with rare earths and thorium. For alloys with Th and the light, low-moment rare earths the susceptibilities are again rather low and have been measured in a field of 1 T. For Sm, Lu and Th substitutions (figure 7) the general character of $M(T)$ resembles that of the pure URu_2Si_2 but the change in slope around T_N is very gradual, which is in accord with the much slighter anomaly in $\rho(T)$. While this could be understood in terms of an even further reduction in the ordered moment (perhaps by increased hybridization of the 5f levels) it is again very difficult to understand why the temperature of anomaly should be so little affected.

For the heavy rare earths, and even for Nd the low-temperature susceptibility is dominated by the solute moment, and Curie–Weiss behaviour with some indications of crystal field splitting on the solutes (especially Tb and Er) is found in the large values found for $|\Theta_p|$ in a high-temperature Curie–Weiss fit. In AC susceptibility measurements for the heavy rare earths (see figure 8), there are always changes in slope around the temperature where the resistivity shows anomalous behaviour in $d\rho/dT$, but this is also the region where

non-linearities of crystal field origin might show themselves. We have not observed any feature in our AC susceptibility measurements at temperatures corresponding to the low-temperature minima in ρ and this might conceivably suggest that the anomalies in $d\rho/dT$, not the minima in ρ, are of magnetic origin. For Gd the small negative value of Θ_p is probably associated with incipient impurity–impurity interaction effects. Large negative Θ_p values for other heavy rare earths presumably arise from single-ion crystal field effects.

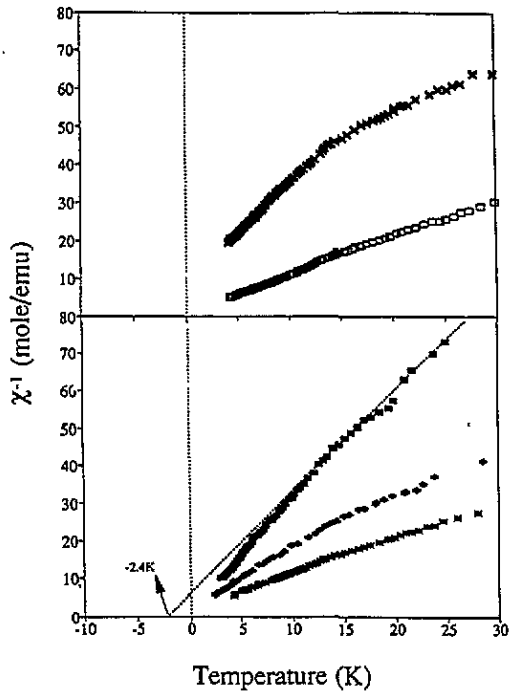

Figure 8. Inverse AC susceptibility for 5% Gd (■), 5.3% Tb (+), 5.1% Dy (∗), 7.6% Ho (□) and 5% Er (×). The dotted line represents a Curie–Weiss behaviour for 5% Ge and −2.4 K the Curie–Weiss temperature (see the text).

4. Discussion

In various physical properties, transport properties in particular, heavy-fermion compounds when pure always show coherence at low temperature. Although the coherence suggests the formation of Bloch wavefunctions out of single-impurity Kondo scattering and theoreticians have endeavoured to reproduce this from the periodic Anderson Hamiltonian, there are not many systematic studies of this [33]. Recently one of the authors [34] has tried to discuss this poorly understood concept of coherence in terms of more familiar phenomena such as magnetic ordering, short-range magnetic fluctuations, and crystal field effects. Although we recognize that the effects described as coherence cannot, in some cases like URu_2Si_2, be explained in terms of a single phenomenon, we think it worth considering, for each system, whether a novel concept is really required. At present perhaps only materials like $CeCu_6$ require it. In agreement with other workers [11, 26] we feel that much of the behaviour

of URu_2Si_2 can be understood in terms of the onset at quite high temperatures (70 K) of short-range antiferromagnetic spin correlations. In the resistivity these combine with the development on cooling of a coherent, Anderson lattice type of state with quite strong conduction electron–5f hybridization.

What is still not clear is the exact character of the ordering that sets in at 17.5 K. The resistivity anomaly, like that of Cr, suggests itinerant antiferromagnetism, and the small size of the ordered moment seems in agreement. What is difficult to understand is the magnitude of the entropy involved and the fact that alloying, especially with Th, can have such striking effects on the specific heat and the electrical resistivity with so little change in the temperature of the transition. Interestingly enough, such seemingly contrasting alloying effects have been observed in UCu_5 with 1% and 1.8% Ni doping at the Cu site [35]. Our demonstration of similar effects for alloys with moment-bearing rare-earth elements confirms a generality for this behaviour and for the ability of some, but not all, substitutions for U to produce a resistance minimum, the origin of which is not yet clear. In a single crystal of very dilute $(U_{0.99}Ce_{0.01})Ru_2Si_2$ [36], the resistivity measurements also show a low-temperature minimum for ρ in the basal plane, but in this material the ρ anomaly at T_N is still strong; such behaviour is not found for $\rho \parallel c$ or for a polycrystalline 2.5% Ce alloy, which resembles our 5% Ce alloy.

Our non-linear susceptibility studies along with magnetization suggests that the magnetic ordering at 17.5 K becomes more localized and conventional with substitutions of La and Ce.

Finally, what is most difficult to understand in present data is that heavy rare-earth and Th alloys show two anomalies at low temperature in ρ while resistivity for La, Ce, Pr, Nd, and Lu alloys present an enhanced Cr-like anomaly. In connection to this, we think it is very interesting that the magnetic ordering at 17.5 K in pure URu_2Si_2 is strongly correlated to charge-density effects [10]. To understand the two anomalies, we are planning to apply microscopic probes to the systems dealt with in this paper. We are also extending this work to systems where a complete range of solid solutions exists. Clarification of the character of the ordering in the more concentrated Ce and La alloys is needed, as is evidence for the presence or absence of quadrupolar ordering.

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