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Relationship between resistivity and specific heat in a canonical non-magnetic heavy fermion alloy system: $UPt_{5-x}Au_x$

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Abstract. A comprehensive study of the relationship between the electronic specific heat coefficient (γ) and the temperature square coefficient (A) of the electrical resistivity for a single, cubic, heavy fermion alloy system, $UPt_{5-x}Au_x$ is presented. In this alloy system, whose low temperature properties are consistent with the Fermi-liquid behavior, γ varies by more than a factor of 10 while the corresponding A coefficient changes by a factor larger than 200. A tracks changes in γ fairly well, but A/γ^2, postulated to have a universal value for heavy fermions, is not constant and varies from about 10^{-6} ($x = 0$, 0.5) to 10^{-5} $\mu\Omega$ cm (mol K/mJ)^2 ($x > 1.1$), thus from a value typical of transition metals to that characteristic of other heavy fermion compounds. We have found a correlation between A/γ^2 and magnetic characteristics such as the paramagnetic Curie-Weiss temperature and the low temperature magnetic susceptibility divided by γ.

PACS. 71.27.+a Strongly correlated electron systems; heavy fermions – 71.10.Ay Fermi-liquid theory and other phenomenological models – 75.30.Mb Valence fluctuation, Kondo lattice, and heavy-fermion phenomena

1 Introduction

Despite more than two decades of research on heavy fermions, the understanding of the formation of heavy electrons remains unsatisfactory, especially for actinide-based systems. A notable advance in delineating the origin of the enhanced electronic mass in Ce-based systems has been achieved *via* alloying studies [1]. Such studies are able to show correlations between various low temperature characteristics of materials within a single crystal structure. Few successful alloying studies involving U-based heavy fermions have been reported so far. One of the serious obstacles in such investigations is ordering of heavy electrons occurring for a majority of U-compounds and alloys. A variety of magnetically ordered states among U-alloys make direct comparisons between different systems difficult. Thus, an ideal system for our purpose would be clearly non-magnetic (and non-superconducting) whose relevant low temperature characteristics span a large range of values upon changing the composition. Further, to avoid any complications due to anisotropy of the electronic properties associated with the crystal structure, materials crystallizing in cubic structures are preferred.

There are only few U-alloys and compounds that can be undoubtedly classified as heavy fermions and which satisfy these prerequisites. One of them, and probably the clearest case is UPt_4Au and related alloys [2]. UPt_4Au forms in a cubic $AuBe_5$-type crystal structure. Its low temperature specific heat divided by temperature is almost 700 mJ/mol K^2 at 1 K. The ratio of the magnetic susceptibility to the specific heat coefficient at low temperatures and the Wilson ratio are small in comparison with all other known heavy fermion systems. Equally small ratios have been found only for two heavy fermion superconductors, UPt_3 and UBe_{13} [3] that lead to speculations that UPt_4Au can be a heavy fermion superconductor at sufficiently low temperatures. However, magnetization measurements [4] performed down to 20 mK have not yielded any evidence of superconductivity or magnetism.

It has been reported [5] that $UPt_{5-x}Au_x$ alloys form single-phase, in a cubic $AuBe_5$-crystal structure over an extended range of concentrations x ($x = 0$ to at least $x = 2.5$). At the same time, their electronic properties, like the low temperature specific heat and magnetic susceptibility, vary a great deal, making this pseudobinary alloy system a unique and convenient system to study the development of a heavy fermion state upon varying x, and to search for possible deviations from the Landau's Fermi-liquid theory in the heavy-fermion regime.

2 Experimental and results

$UPt_{5-x}Au_x$ alloys have been prepared by arc melting. All alloys in the range $x = 0$ to 2.5 were single phase according to the X-ray diffraction analysis. Based on a previous

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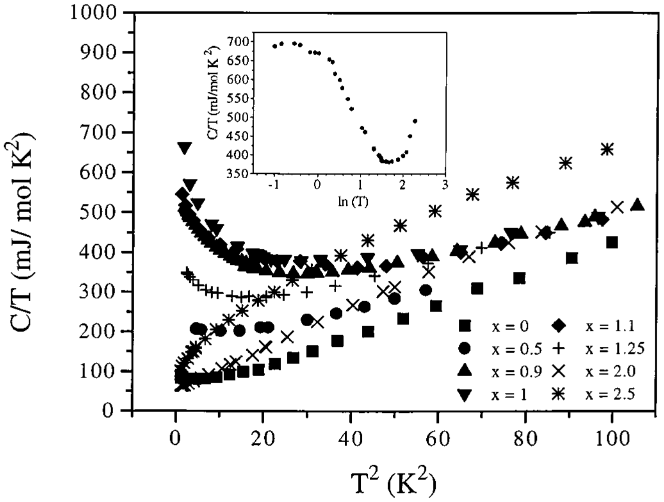

Fig. 1. C/T *versus* T^2 for $\mathrm{UPt}_{5-x}\mathrm{Au}_x$ alloys. The inset shows C/T *versus* ln$T$ for $x = 1$ and temperatures between 0.3 and 10 K.

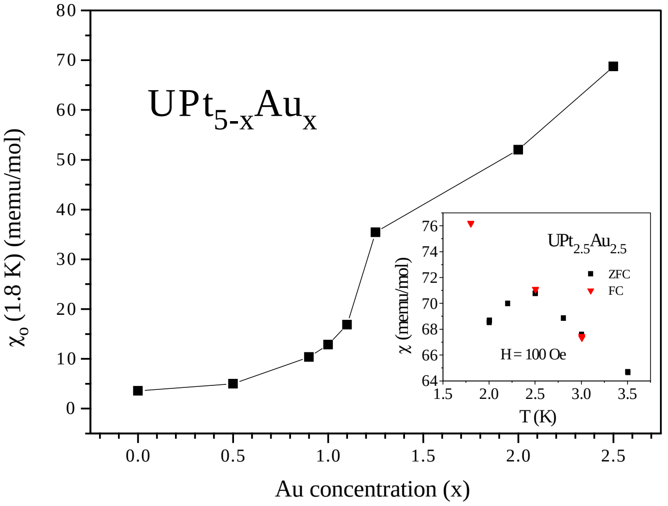

Fig. 2. χ_o *versus* x for $\mathrm{UPt}_{5-x}\mathrm{Au}_x$. The inset shows zero-field-cooled (ZFC) and field-cooled (FC) susceptibility (H = 100 Oe) for $\mathrm{UPt}_{2.5}\mathrm{Au}_{2.5}$.

study [5], which has not found any significant dependence of the low temperature properties of these alloys on annealing, all samples used were as-cast. In our present investigation, we have used some of those previously investigated unannealed samples.

Specific heat has been measured between about 1 and 10 K with the exception of $\mathrm{UPt}_4\mathrm{Au}$, which has been studied to 0.35 K. The total specific heat divided by temperature (C/T) *versus* temperature square is shown in Figure 1 for eight different alloys corresponding to x between 0 and 2.5. C/T at the lowest measured temperature, which we call further γ, spans a large range of values. It increases from about 80 mJ/mol K^2 for $x = 0$ to almost 700 mJ/mol K^2 for $x = 1$, and then drops again to about 60 mJ/mol K^2 for $x = 2$, thus changing by more than an order of magnitude.

A better estimate of γ would be C/T at 0 K obtained from the fit of the low temperature specific heat data to some theoretical function. This could be especially important for $\mathrm{UPt}_4\mathrm{Au}$ which exhibits a number of similarities to extensively studied $\mathrm{UCu}_4\mathrm{Pd}$ [6,7]. In this latter compound, also forming in the AuBe_5-type crystal structure, C/T diverges as T approaches zero. C/T of UPt_4Au, between about 1 and 10 K, has also a quasi-logarithmic temperature dependence if the phonon contribution is subtracted. However, below 1 K, the C/T data show a clear tendency towards saturation (and possibly a very weak maximum near 0.6 K which can not be resolved due to the data scattering). This saturation is shown in the inset to Figure 1 in C/T *versus* ln T format. C/T between 1.1 and 0.36 K varies by less than 5 % which is less than the absolute accuracy of the measurement, about 10 %. This result provides a justification for associating γ with the value of C/T at about 1 K.

In agreement with reference [8], we have found that the low temperature specific heat data of UPt_5 can be well described by a spin-fluctuation formula. This formula can still be applied to $x = 0.5$, but fails to describe the temperature dependence of C/T for all other investigated alloys. Interestingly, the specific heat for these other concentrations can not be associated with either Kondo-type behavior or, more generally, with any type of behavior corresponding to a single-impurity scaling, $C(T, T_\mathrm{K}) = C(T/T_\mathrm{K})$ [9]. Because of almost identical molecular weights across the series, a small phonon contribution at 1 K is expected to be approximately constant. Attempting to account for the phonon contribution, we have investigated two non-f AuBe_5 homologues: $\mathrm{ThPt}_4\mathrm{Au}$ and $\mathrm{YPt}_4\mathrm{Au}$ [10]. The specific heat for both $\mathrm{ThPt}_4\mathrm{Au}$ and $\mathrm{YPt}_4\mathrm{Au}$ has a weak spin-fluctuation temperature dependence with γ values of 11 and 17 mJ/mol K^2, respectively. Since neither of them contains $5f$-elements, these weak spin-fluctuations have to be associated with $5d$-electrons. This is an important observation suggesting that the $5d$ electrons of Pt can also be responsible for the spin-fluctuation term in the specific heat of UPt_5, and that $5f$-$5d$ hybridization effects can play a significant role in other $\mathrm{UPt}_{5-x}\mathrm{Au}_x$ alloys, especially in those corresponding to small values of x. A similar conclusion has been reached by authors of reference [8], based on a magnetic susceptibility study. They have argued that an unusually large absolute value of the Curie-Weiss temperature ($\sim$ 500 K) in the magnetic susceptibility of the pure UPt_5 compound reflects strong $5f$-delocalization effects due to the $5d$-electrons of Pt.

Magnetic susceptibility at the lowest temperature measured, χ_o (1.8 K), shown as a function of x in Figure 2, does not correlate with γ at all. χ_o increases monotonically from 3.6 to almost 70 memu/mol between $x = 0$ and $x = 2.5$ (Fig. 2). Thus the ratio χ_o/γ is not constant across the system, as expected from the single-impurity model, but increases with x. An especially strong increase in χ_o for $x > 1.25$ prompted us to perform low field magnetization measurements on these alloys. These measurements for $\mathrm{UPt}_{2.5}\mathrm{Au}_{2.5}$ revealed a maximum at about 2.5 K (T_M) in the zero-field-cooled susceptibility (ZFC) and a discrepancy between zero-field-cooled and field-cooled (FC) susceptibilities at low temperatures (Inset to Fig. 2). No corresponding maximum has been found in the specific heat data. Similar spin-glass-like anomalies have been observed in other γ-enhanced U- or Ce-based systems, with chemical formulas of the form $M(A_xB_{1-x})$, near $x = 0.5$; where M is either U or Ce, and A and B are normal or transition metals [11]. Although there is no consensus as to the nature of the ground state (or different ground states) in these alloys, it is believed that non-magnetic atom disorder (NMAD) is responsible for their spin-glass-like properties. Low field magnetization measurements performed on all other alloys, including UPt_2Au_3, have not detected any anomalies, although small discrepancies (up to 5%) between ZFC and FC susceptibility have been found for $x = 2$ below 3 K. The maximum value of T_M occurring for $x \simeq 2.5$ in the $UPt_{5-x}Au_x$ system is consistent with NMAD reaching its maximum near $x = 2.5$. Magnetic measurements indicate further that $UPt_{5-x}Au_x$ alloys are not completely free from complications associated with the nearness to magnetism.

Before addressing our resistivity results we point out the important issue of preferential site occupancy. Strong variation of the thermodynamic low temperature properties of $UPt_{5-x}Au_x$ near x=1 and their extremal values for x=1 have been attributed to possible preferential atomic occupancy of non-U sites [2,5]. Pt would preferentially occupy trigonal symmetry 16e sites while Au would go on cubic 4c sites. Additional, indirect support for this explanation is given by thermodynamic investigations of the aforementioned related U-alloy system $UCu_{5-x}Pd_x$, forming in the same crystal structure. The concentration corresponding to x=1 also exhibits extremal values of its low temperature properties in comparison to other alloys belonging to the $UCu_{5-x}Pd_x$ system. Note that the smaller atoms (Cu, Pt) would preferentially occupy majority 16e sites, while larger atoms (Pd, Au) would occupy 4c sites in both alloy systems. Previous attempts using X-ray diffraction and resistivity studies to clarify this issue for $UPt_{5-x}Au_x$ were inconclusive [5]. However, a recent elastic neutron scattering study [12] on the related $UCu_{5-x}Pd_x$ has confirmed the preferential non-U site occupancy. Thus, a similar preferential site occupancy or chemical ordering of x=1 is probable for $UPt_{5-x}Au_x$.

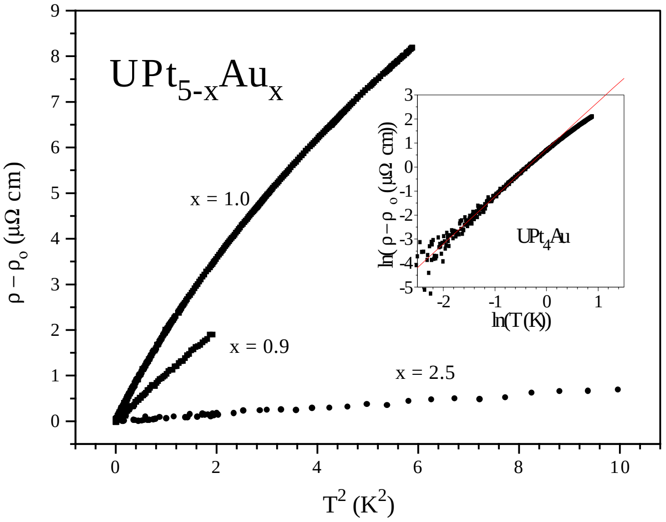

Fig. 3. $\rho - \rho_o$ *versus* T^2 for $x = 0.9$, 1, and 1.25. The inset shows the zero-temperature limit of the resistivity for UPt_4Au on a logarithmic scale.

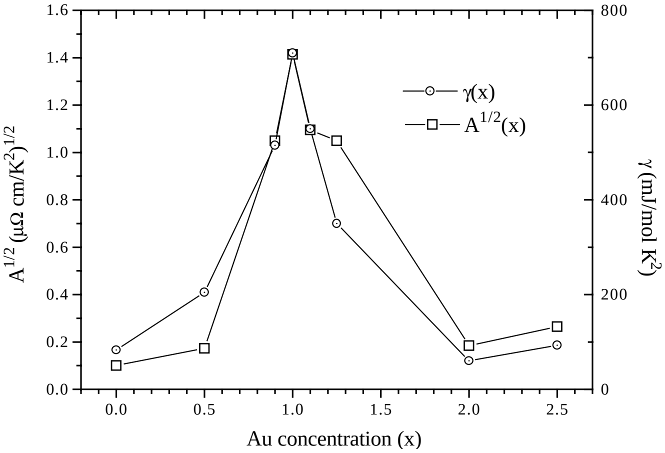

Fig. 4. $A^{1/2}$ and γ *versus* x for $UPt_{5-x}Au_x$.

The resistivity for all investigated alloys has a Fermi-liquid-like temperature dependence, $\rho = \rho_o + AT^2$, at sufficiently low temperatures. This variation is shown in Figure 3 for $x = 0.9$, 1, and 2.5 in the form of $\rho - \rho_o$ *versus* T^2. The residual resistivity ρ_o, obtained on samples derived from a single batch of U, increases with x from about 25 $\mu\Omega$ cm for $x = 0$ [8] to about 165 $\mu\Omega$ cm for $x = 2.5$. Thus, in agreement with the previous study, our resistivity results do not directly support the notion of a chemical ordering near x=1; i.e., no discernable dip in ρ_o is observed for this concentration. We have noticed some variation of ρ_o between samples obtained from U-batches having different chemical purity. One of our UPt_4Au samples had $\rho_o \simeq 40\mu\Omega$ cm, the other one (used also in Ref. 5) had $\rho_o \simeq 80\mu\Omega$ cm. Interestingly, the temperature square coefficient of the resistivity A, discussed next, was identical for both x=1 samples, within the experimental uncertainty (10 %).

The most striking feature of Figure 3 is a large variation, from concentration to concentration, in the slope of $\rho - \rho_o$ *versus* T^2. This slope corresponds to the A coefficient. In the investigated range of concentrations, A changes, in a non-monotonic fashion, from its lowest value of about 0.01 $\mu\Omega$ cm/K^2 for $x = 0$ to over 2 $\mu\Omega$ cm/K^2 for $x = 1$, thus by a factor larger than 200. The A coefficient fairly well tracks the trends for the changes in γ, as demonstrated in Figure 4. The format of this figure, $A^{1/2}$ and γ *versus* x, has been chosen based on the observation of Kadowaki and Woods [13], that for a large number of heavy fermion compounds $A \simeq 10^{-5}\gamma^2$, where A is expressed in $\mu\Omega$ cm/K^2 and γ in mJ/mol K^2. A similar relationship has been found in transition metals, but with the average ratio A/γ^2 about 20 times smaller than that in heavy fermion compounds. This proportionality between A and γ^2 in transition metals has been accounted

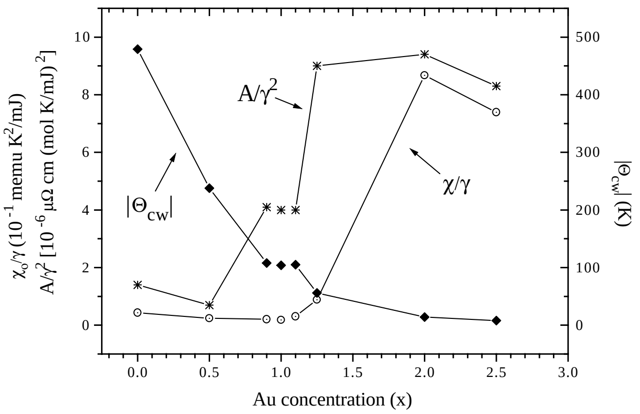

Fig. 5. A/γ^2, χ_o/γ, and $|\Theta_{\mathrm{CW}}|$ *versus* x.

for theoretically [14] in terms of Baber's model [15] describing electron-electron scattering. Also, the electron-electron scattering is believed to be the source of the enhanced values of A in heavy fermion systems [16,17]. However, the large ratio A/γ^2, in comparison with transition metals, has not yet been fully explained. Furthermore, the universality of this ratio has not been established. The ratio of A and γ^2 is not constant across the $\mathrm{UPt}_{5-x}\mathrm{Au}_x$ series (Fig. 5). This ratio is of order 10^{-6} $\mu\Omega$ cm (mol K/mJ)^2 only for the pure UPt$_5$ compound and the $x = 0.5$ alloy, thus within the range of values corresponding to many transition metals. A/γ^2 grows to 4×10^{-6} $\mu\Omega$ cm (mol K/mJ)^2 for $x = 0.9$, 1, and 1.1, and reaches a value close to the claimed universal ratio for heavy fermion compounds for $x = 1.25$, 2, and 2.5.

3 Discussion

Our results confirm the correlation between A and γ^2 in heavy fermion alloys. They also indicate a much closer relationship between A and γ than between γ and magnetic susceptibility (χ). This result, somewhat surprising, can be explained by the fact that, in general, A and γ probe low energy excitations for all values of q-vector of the Brillouin zone while χ corresponds to excitations at $q = 0$ only. Since the single impurity Kondo model predicts a simple relationship between χ_o and γ, lack of such a relationship in our data indicates importance of intersite effects, like RKKY interactions. This significance of intersite interactions has already been stressed while discussing the temperature dependence of the specific heat. There are, on the other hand, interesting correlations between the magnetic susceptibility and the ratio A/γ^2, discussed further.

There have been several theoretical investigations of this ratio [16,17] in heavy fermions. Among others, it has been postulated [16] that the disorder can influence A/γ^2 values. Both, A/γ^2 and the amount of crystallographic disorder, in general, increase with x in our alloy system, therefore implying that the disorder would have to increase this ratio. Slowing down (a plateau in Fig. 5) of an increase of A/γ^2 *versus* x near $x = 1$ (a concentration for which a partial or full chemical ordering is expected) is further consistent with this interpretation. However, identical, within the accuracy of the measurement, values of A/γ^2 obtained for two UPt$_4$Au samples with very different residual resistivities argue against the importance of disorder.

A related question of the role of the crystal symmetry on A/γ^2 has never been considered. The experimental survey published in reference [13] does not indicate such a relationship. On the other hand, the step-like increase of A/γ^2 values near $x = 1$ might be a consequence of the change of the crystal structure taking place near $x = 1$, as discussed above.

Correlations between A/γ^2 and parameters derived from the magnetic susceptibility, Curie-Weiss temperature ($|\Theta_{\mathrm{CW}}|$) χ_o/γ are illustrated in Figure 5. In view of what has been said above, one can not expect a simple interdependence between all these quantities in a Kondo-lattice system with, in general, q- and ω-dependent RKKY interactions. χ_o/γ in a single Kondo impurity limit is expected to be constant. For a Kondo-lattice with predominantly antiferromagnetic interactions, we expect an increase of χ_o/γ upon approaching the magnetic order and, in fact, the magnitude of χ_o/γ is often used in experimental studies as a measure of the proximity to magnetism. Therefore, our results seem to indicate that "more magnetic alloys" have larger values of A/γ^2. The compilation of existing experimental data suggests that systems in which heavy electrons and magnetic order (with a small value of the ordering temperature) coexist have anomalously large values of A/γ^2. *E.g.*, these values for antiferromagnetic CePb$_3$ [18], CeAl$_2$ [19], CeAuAl$_3$ [20], CePd$_2$In [21], and YbPdBi [22] (all antiferromagnets with T$_N$ of order 1 K) are (3, 5, 10, 3, and 30 10^{-5} $\mu\Omega$ cm/K^2, respectively. The effect of magnetic interactions on the A/γ^2 ratio has been studied theoretically by Takimoto and Moriyaé[17] in the framework of antiferromagnetic spin fluctuations. They have found that this ratio is essentially universal for heavy fermions except for the immediate vicinity of the antiferromagnetic order where A/γ^2 is enhanced. However, since this work does not take into account the variation of other materials characteristics, such as the conduction bandwidth (expected to be important in $\mathrm{UPt}_{5-x}\mathrm{Au}_x$ and discussed below), we can not claim the disagreement with our results.

Figure 5 shows also rather striking similarity between the x-dependence of A/γ^2 and $|\Theta_{\mathrm{CW}}|$. As it has been already argued, the very large range of values $|\Theta_{\mathrm{CW}}|$ spans (from less than 10 to almost 500 K) can be related to the evolution of the conduction-electron bands. We expect effects due to the presence of a narrow d-band and strong f-d hybridization to be particularly important for Pt-rich samples. The hybridization becomes weaker and changes its character to f-s type upon replacing Au for Pt. Therefore, it is plausible that the variation of A/γ^2 in $\mathrm{UPt}_{5-x}\mathrm{Au}_x$ reflects this evolution of hybridization parameters.

The discussed Fermi-liquid-like temperature dependence in the resistivity of $\mathrm{UPt}_{5-x}\mathrm{Au}_x$ alloys is observed at the lowest temperatures only, and the range of temperatures for whose it holds varies with x. The upper temperature limit of this variation seems to correlate with the inverse of γ; *e.g.*, it is about 1 K for $x = 0.9$, 0.7 K for $x = 1.0$, 4 K for $x = 2.0$, and about 3 K for $x = 2.5$. This observation is rather surprising since it is generally believed that the $\rho = \rho_o + AT^2$ temperature dependence is a property of the so-called coherence regime, which is a consequence of a translational periodicity of the system. The distraction of this ligand-site symmetry does not have a strong effect on the temperature width of "the coherence regime".

Finally, our results have implications on the possible explanation of the origin of the non-Fermi-liquid-like low temperature properties for the extensively studied $\mathrm{UCu}_{5-x}\mathrm{Pd}_x$ alloys [6,7]. One of the leading explanations [23] is based on the Kondo-disorder idea [24]. Despite the obvious ligand disorder also in $\mathrm{UPt}_{5-x}\mathrm{Au}_x$ alloys, their properties stay Fermi-liquid-like over a wide range of x.

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