Doping effects on UPd2Al3

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Abstract

We present first results of a doping study on the U site on UPd$_2$Al$_3$. These results give further support for a tetravalent uranium configuration and an induced type of antiferromagnetic ordering. They confirm the importance of an unperturbed electronic configuration for both antiferromagnetic long-range ordering and heavy-fermion superconductivity. Implications for the interaction between both phenomena are discussed.

1. Introduction

Since the discovery of the two heavy-fermion super-conductors (HFS) UNi$_2$Al$_3$ and UPd$_2$Al$_3$ two years ago [1, 2] many investigations have already been carried out on these two compounds, and a consistent picture of their properties is beginning to emerge [3]. From neutron-scattering [4] and μSR measurements [5], we know that they order antiferromagnetically at 4.5 and 14.5 K, respectively, with surprisingly large moments, 0.1 μ_B in the Ni compound and 0.85 μ_B in the Pd compound. Below 1.2 and 2.0 K, respectively, bulk superconductivity has been found to coexist with these large moments. There is increasing evidence from specific heat, resistivity [6, 7] and neutron-scattering results [8] that the 5f electrons are fairly well localized in UPd$_2$Al$_3$. We have proposed [9] that the peculiar temperature dependence of the magnetic susceptibility $\chi(T)$ is related to a crystal-electric-field (CEF) scheme with two lowest-lying doublets within a tetravalent uranium configuration, and that antiferromagnetic (AF) ordering occurs among induced moments. Neutron-scattering and μSR experiments as

well as specific heat measurements under pressure have also shown that the interaction between AF ordering and superconductivity is rather weak. The properties of UNi$_2$Al$_3$ are similar to those of UPd$_2$Al$_3$, however, with a characteristic energy that is a factor of 3 larger, resulting in more delocalized 5f states when compared to the Pd compound. Both systems are phenomenologically related to URu$_2$Si$_2$, which, considering its characteristic energy, seems to be intermediate between them. Despite the recent progress described above, fundamental problems, i.e. the correct description of the magnetism, the nature of the superconducting state and the interaction between both phenomena, are still unsolved. In order to get a better insight into these problems, it is useful to investigate the normal-state properties by applying an external magnetic field or external pressure or by controlled doping.

Recently we reported a study of the U(Pd$_{1-x}$Ni$_x$)$_2$Al$_3$ alloy system [10]. In the present paper we report and discuss preliminary results on U$_{1-x}$M$_x$Pd$_2$Al$_3$, where U is substituted by the following dopants: Y, Pr, Gd and Th. We will focus on the effect that small dopant concentrations have on magnetism and superconductivity, whereas Kim et al. [11], who were interested in

"non-Fermi-liquid phenomena", studied samples at higher dopant levels. Taking into account their results for La doping, we are able to discriminate different possible sources for the observed phenomena: Th is tetravalent, whereas all others are trivalent. Y and Gd have approximately the same atomic volume as U, whereas La, Pr and Th are significantly larger. Gd has a large stable local moment, $\mu_{eff} = 7.94\mu_B$, and the highest spin ($S = 7/2$) among the rare-earth elements. For Pr the values of both $\mu_{eff} = 3.58\mu_B$ and $S = 1$ are considerably lower, and Th, La and Y carry no moment at all. Samples were prepared by classical metallurgical methods and investigated by means of DC ($T > 4.2$ K) and AC (20 mK $< T < 2$ K) resistivity as well as DC ($T > 2$ K, SQUID) and AC (20 mK $< T < 2$ K) susceptibility measurements.

2. Magnetism

The most interesting among our new results is an extreme change in the susceptibility $\chi(T)$ at low Gd concentration compared to rather small changes for all other dopants. In Fig. 1 we compare $\chi(T)$ of Gd- and Pr-doped UPd$_2$Al$_3$ for 2 K $< T < 60$ K. The results for the other dopants are very similar to those for Pr. In all these latter systems increasing dopant concentration shifts the susceptibility maximum, which in the pure compound occurs around $T_m = 35$ K, to lower temperatures. Further on, the kink in $\chi(T)$ at T_N becomes more pronounced, and T_N also shifts to lower temperatures. By contrast, only 1 at% Gd induces a large additional Curie-Weiss-like contribution, by which the $\chi(T)$ maximum is masked almost completely. $\chi(T)$ taken at 4 K is increased by a factor of 8 after doping with 5 at% Gd, while a factor of 1.5 increase only is found at 5 at% Pr. From the straight line found in a plot of χ^{-1} versus T below $T = 8$ K, the "effective moment" associated with this contribution can be estimated: $\mu_{eff} = 2.0\ \mu_B$ and $3.0\ \mu_B$ for $x = 0.01$ and 0.05, respectively. These values are far larger than those expected if only Gd contributes. Consequently, a large part of this additional Curie-Weiss contribution has to originate in the uranium atoms. The same behavior was observed for Gd-doped UNi$_2$Al$_3$ [12] and appears to characterize Gd-doped URu$_2$Si$_2$ [13] as well. On the other hand, for other heavy-fermion compounds a low Gd concentration has a rather small effect on $\chi(T)$ [14]. This anomalous behavior may be related to the induced type of local U moments within a CEF scheme showing a non-magnetic lowest-lying level. In such a system the large Gd moment is likely to induce a large transferred field and, in this way, a large moment at the neighboring U sites. In fact, for the crystal-field scheme proposed for UPd$_2$Al$_3$ [9], the moment increases very sharply for $B > 30$ T [15].

Surprisingly, the long-range AF ordering appears not to be stabilized by this large Curie–Weiss contribution. On the contrary, although the anomaly at T_N is not reduced in size, it is shifted to lower temperatures when the Gd concentration is increased. In Fig. 2(a) we compare the concentration dependence of $T_N(x)$ for different dopants. All trivalent elements (La, Y, Pr and Gd) present a similar slope $\delta T_N/\delta x$. One finds -0.5 K/at% for La [11], -0.4 K/at% for Y and Pr and -0.3 K/at% for Gd. By contrast, doping with Th reduces T_N at a slower rate of -0.1 K/at%. Obviously neither the magnetic moment of the dopant nor its atomic volume, but instead its electronic configuration, appears to be essential for the T_N depression. This is in agreement with the results for U(Pd$_{1-x}$T$_x$)$_2$Al$_3$ in which Pd is replaced by either Ni [10] or Pt [11, 16], both being isoelectronic to Pd and leaving T_N nearly unchanged. This suggests that the most important parameter governing T_N is the valence of the dopant: whereas isoelectronic dopants have
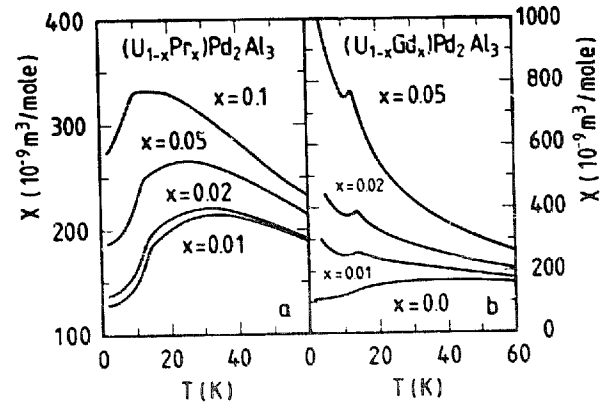

Fig. 1. Magnetic susceptibility of (a) Pr- and (b) Gd-doped UPd$_2$Al$_3$. The kinks near 14 K correspond to the AF transition.
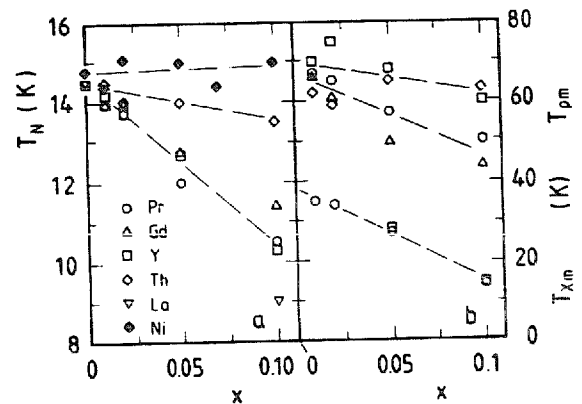

Fig. 2. (a) T_N versus x for U$_{1-x}$M$_x$Pd$_2$Al$_3$ (open symbols) and for U(Pd$_{1-x}$Ni$_x$)$_2$Al$_3$ (◆). (b) $T_{\rho m}$ and $T_{\chi m}$ versus x for U$_{1-x}$M$_x$Pd$_2$Al$_3$. Same symbols as in (a).

a minor effect on T_N, dopants with a different valence give rise to a pronounced T_N depression. A similar relation can also be established for URu_2Si_2: replacement of Si by Ge [17] leaves T_N unchanged, while replacement of Ru by Os [18] even leads to a slight increase of T_N. By contrast, if Ru is replaced by Rh, Ir or Re [18] or U by La [19], T_N is found to be strongly depressed. Thus, long-range AF ordering among U moments is found to be very sensitive to changes of the electronic configuration within the lattice.

We now analyze the composition dependence of the positions $T_{\rho m}$ and $T_{\chi m}$ of the maxima in $\rho(T)$ and $\chi(T)$ (Fig. 2(b)). For $U(Pd_{1-x}Ni_x)_2Al_3$ we previously found [10] that both $T_{\rho m}$ and $T_{\chi m}$ increase with x, while the ratio $T_{\rho m}/T_{\chi m}$ remains nearly constant in the whole concentration range. For the present case of doping on the U site the situation is more complex. Generally, both $T_{\rho m}$ and $T_{\chi m}$ decrease with increasing dopant concentration. The decrease of $T_{\chi m}$ was found to be independent of the kind of dopant. (Note that $T_{\chi m}$ cannot be determined for the Gd-doped samples.) The depression of $T_{\rho m}$ is less pronounced compared to that of $T_{\chi m}$. The non-magnetic dopants (Th, Y) appear to have a weaker effect on $T_{\rho m}$ than the magnetic ones (Pr, Gd). The results on the $U(Pd_{1-x}Ni_x)_2Al_3$ system suggested [6] the maximum in $\chi(T)$ to be related to CEF effects only while the maximum $\rho(T)$ was proposed to arise from the combined effect of CEF and Kondo interactions. Such a maximum in $\chi(T)$ can only show up for special configurations of the three lowest-lying CEF levels [9]. Already small changes of the CEF parameters would generally result in a disappearance of this feature. Thus, the $\chi(T)$ maximum is related to the lowest CEF levels only, whereas the maximum in $\rho(T)$ depends on the overall CEF scheme. In this sense, the more pronounced changes in $T_{\chi m}$ and the less pronounced ones in $T_{\rho m}$ may naturally arise from such slight changes in the crystal-field parameters which have a relatively strong effect on the low-lying CEF states, but a relatively weak one on the total CEF scheme.

3. Superconductivity

Compared to their magnetic properties, the superconducting properties of the doped samples have, so far, not been investigated as thoroughly. Up to now we have determined the transition temperatures T_c of Gd- and Y-doped UPd_2Al_3. Figure 3(a) displays an extremely rapid decrease of T_c from 2 K in the undoped samples to ~ 1 K at 1 at% Y or Gd content. It is surprising that doping with Y and Gd furnishes nearly the same effect, in view of the remarkable differences found in $\chi(T)$ (Fig. 1). This supports the idea [7] that the electronic subsystems

responsible for the respective magnetic and superconducting properties of UPd_2Al_3 are rather well decoupled. In Fig. 3(a) we also compare our results with the literature data for other HFS [20]. The T_c depression in Gd-or Y-doped UPd_2Al_3 exceeds that in Gd-doped $CeCu_2Si_2$ or UBe_{13} by approximately a factor of 3 and that in Ni-doped UPd_2Al_3 by a factor of 6 [10].

A possible explanation for such a rapid decrease of T_c is found when analyzing the residual resistivities. Since for most of the samples $\rho(T)$ has only been measured down to 4.2 K and since absolute values of ρ are very difficult to determine owing to microcracks in the samples, we shall use the resistivity ratio, $RR_{4.2} = \rho(300\ K)/\rho(4.2\ K)$, as parameter. We find that $RR_{4.2}$ decreases much more rapidly, when doping on the U site than for doping with Ni on the Pd site. Thus, in the former case the coherance of the heavy-Fermi-liquid state is found to be destroyed more rapidly. The same also holds true, apparently for superconductivity (see Fig. 3(a)). This prompted us to study the correlation between the residual resistivity and T_c. In Fig. 3(b), T_c is plotted as a function of $1/RR_{4.2}$ with the composition as an implicit parameter. $1/RR_{4.2}$ is roughly proportional to the residual resistivity. (Note that the origin of the abscissa has been shifted from 0 to 0.04: in samples with the highest resistivity ratio, $\rho(4.2\ K)$ is dominated by spin-wave scattering, constituting a lowest intrinsic value of $\sim 5\ \mu\Omega\ cm$.) In this figure, the results are shown for Y and Gd doping on the U site, for Ni doping on the Pd site and for non-stoichiometric samples [21]. Despite the large scatter, which is not surprising in view of the many parameters determining $\rho(4.2\ K)$ and T_c, a correlation between T_c and $1/RR_{4.2}$ can clearly be established: the

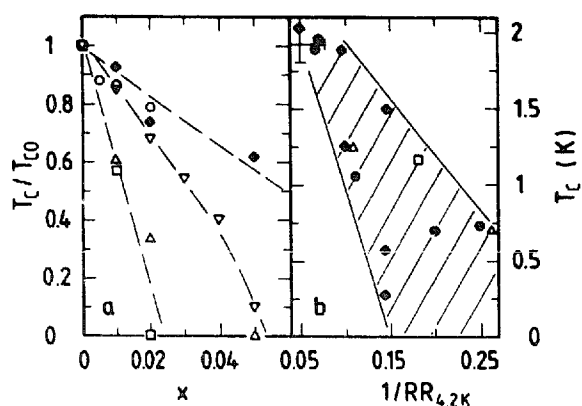Fig. 3. (a) T_c/T_{c0} versus $U_{1-x}M_xPd_2Al_3$ ($\square$: M = Y, $\triangle$: M = Gd), for $U_{1-x}Gd_xBe_{13}$ ($\bigcirc$), for $Ce_{1-x}Gd_xCu_{2.2}Si_2$ ($\triangledown$) and for $U(Pd_{1-x}Ni_x)_2Al_3$ ($\blacklozenge$). (b) T_c versus $1/RR_{4.2}$ for UPd_2Al_3 (same symbols as in (a)) and for $\bullet$: non-stoichiometric samples [21]. The cross corresponds to the mean values of both T_c and $1/RR_{4.2}$ as observed in 15 undoped samples.

larger the $1/RR_{4.2}$ value, and thus $\rho(4.2 \text{ K})$, the smaller is the value of T_c. Superconductivity disappears for $1/RR_{4.2}$ larger than a critical value of 0.15–0.3, depending on the dopant. Taking $\rho(300 \text{ K}) = 180 \pm 30$ $\mu\Omega$ cm as deduced for single crystals and crack-free samples, we estimate a residual resistivity between 30 and 60 $\mu\Omega$ cm. Within the assumption of quasi-free electrons and using a Fermi velocity of 1.2×10^4 m/s as determined for undoped UPd_2Al_3 from B_{c2} and $\gamma = 150$ mJ/K^2 mol [2], one obtains an elastic mean free path of 40–80 Å, comparable to the BCS coherence length $\xi = 85$ Å [2]. This would mean that superconductivity disappears when the elastic mean free path becomes smaller than the coherence length. Such a correlation between residual resistivity and T_c is expected for non-conventional superconductors with lines or nodes of zero gap on the Fermi surface, since an increasing potential scattering will mix different parts of the Fermi surface [22]. The same argument would also apply to a model with two subsystems of 5f quasiparticles, one being responsible for the superconducting and the other for the magnetic properties, as recently proposed by Caspary et al. [7]. In this case, an increase of the scattering between the magnetic (i.e. non-superconducting) and the superconducting electronic subsystems would strongly reduce T_c. However, one should keep in mind the preliminary character of the type of analysis presented above.

4. Conclusions

In conclusion, our doping studies have revealed interesting correlations, which show that long-range antiferromagnetic ordering and, in a much stronger way, heavy-fermion superconductivity of UPd_2Al_3 are very sensitive to changes in the electronic configuration of this compound. A partial exchange of one of the constituents by a dopant element with different valence strongly reduces both T_c and T_N. These results give further support for a tetravalent uranium configuration and an induced type of antiferromagnetic ordering. If confirmed by further investigations, the correlation between increasing residual resistivity and decreasing T_c appears to be of superior importance for the understanding of superconductivity in these compounds.

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