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DOI
10.1016/0921-4526(96)00034-8

Publication date
1996

Published in
Physica B-Condensed Matter

ELSEVIER

Physica B 223&224 (1996) 44–46

PHYSICA B

The influence of Gd and Ni impurities on the superconducting transition temperature of UPt_3

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Abstract

The effect of Gd and Ni impurities on the upper superconducting transition temperature of UPt_3 ($T_c^+ = 0.55$ K) has been studied by means of resistivity measurements. Surprisingly, we find that paramagnetic Gd impurities suppress T_c^+ at the same rate as non-magnetic Y or Th impurities, yielding further support for non-s-wave pairing. Ni impurities also suppress T_c^+, but do not dissolve homogeneously in the UPt_3 matrix.

The superconducting properties of heavy-fermion UPt_3 are extremely sensitive to substitutions on both the U and Pt sites [1, 2]. All impurity studies reported so far were directed towards non-magnetic elements: Y and Th doping on the U site, and Pd, Ir and Au doping on the Pt site. Electrical resistivity and specific-heat experiments [1, 2] yielded firm evidence that non-magnetic impurities do not simply give rise to pair-weakening, but rather give rise to effects comparable to pair-breaking, which is in strong contrast to the standard behaviour for ordinary BCS singlet superconductors. This suggests that non-magnetic impurities act as holes in the Kondo-lattice. In the course of a more comprehensive investigation, we here report on the first study [3] of a paramagnetic impurity, namely Gd (which has a free ion moment of 7.94 μ_B) on the U site, and of Ni impurities on the Pt site.

The polycrystalline samples were prepared by arc-melting on a water-cooled copper crucible in a continuously Ti gettered argon atmosphere. As starting elements we used uranium with a purity of 99.99% (supplied by JRC-EC, Geel), spectral pure Pt and Ni (Johnson Matthey) and Gd with a purity of 99.9% (Highways International, Baarn). In the case of the $U_{1-x}Gd_xPt_3$ series, we first prepared master alloys with $x = 0.000$ and $x = 0.0050$. Samples with $x = 0.0005$, 0.0010, 0.0020 and 0.0035 were obtained by diluting. In the case of the $U(Pt_{1-x}Ni_x)_3$ series, master alloys with $x = 0.000$ and $x = 0.010$ were prepared, while samples with $x = 0.001$, 0.002, 0.003, 0.005 and 0.007 were obtained by diluting. All samples were wrapped in tantalum foil and annealed in water-free quartz ampoules at a temperature of 950° C for a period of 7 days in presence of a piece of uranium that served as a getter. The annealed samples were investigated by electron probe microanalysis (EPMA). In the case of Ni doping, it was observed that not all Ni dissolves in the matrix (only about 40% of the nominal concentration) and a second phase (possibly $U_2Pt_5Ni_3$) was detected at the grain boundaries. This is attributed to the small ionic radius of Ni (0.69 Å) compared to Pt (0.80 Å). The presence of Gd in the samples could not be confirmed by EPMA, because of the limited sensitivity. The samples were shaped by means of spark erosion into dimensions suitable for electrical resistivity measurements. The experiments were performed in a ^3He system or in a dilution refrigerator, using standard probe AC-techniques.

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0921-4526/96/$15.00
PII S0921-4526(96)00034-8

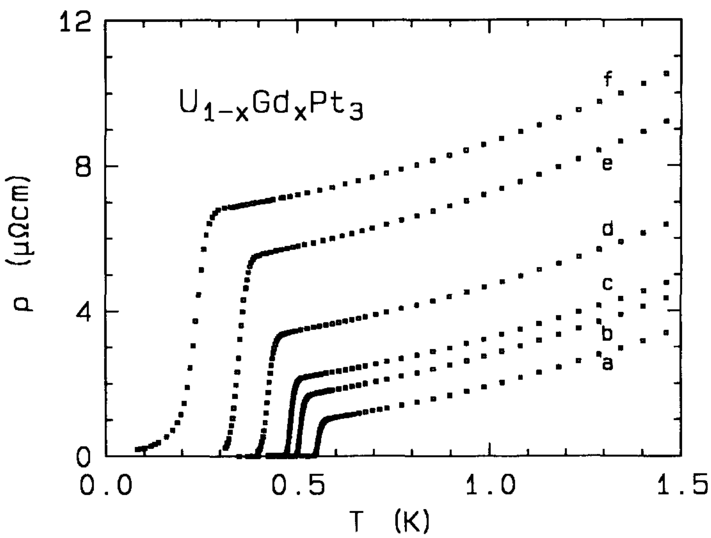

Fig. 1. Electrical resistivity versus temperature for $U_{1-x}Gd_xPt_3$ with (a) $x = 0.000$, (b) $x = 0.0005$, (c) $x = 0.0010$, (d) $x = 0.0020$, (e) $x = 0.0035$ and (f) $x = 0.0050$. The resistivity is normalized to a room-temperature value of 240 μΩ cm.

Table 1
The residual resistivity (ρ_0), the coefficient of the T^2 term (A), the upper superconducting transition temperature (T_c^+) and the width of the superconducting transition (ΔT_c^+) for $U_{1-x}Gd_xPt_3$

x	ρ_0 (μΩ cm)	A (μΩ cm/K^2)	T_c^+ (mK)	ΔT_c^+ (mK)
0.0000	0.59	1.34	555	13
0.0005	1.31	1.48	508	17
0.0010	1.81	1.45	483	17
0.0020	3.08	1.62	423	28
0.0035	5.33	1.95	349	37
0.0050	6.77	1.88	234	59

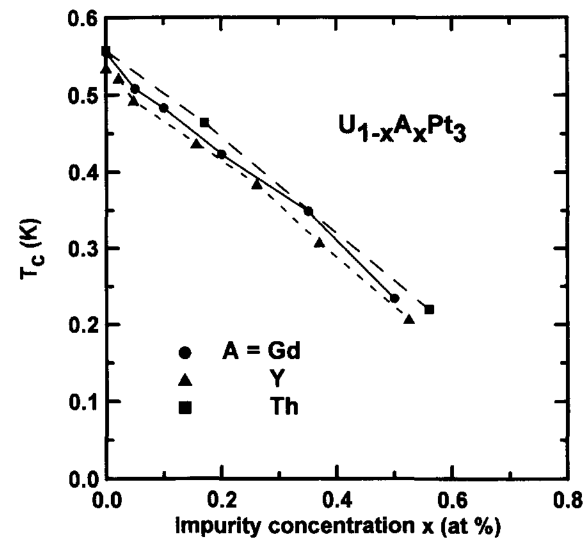

Fig. 2. T_c^+ versus impurity concentration for $U_{1-x}A_xPt_3$, where A represents Gd (●), Y (▲) or Th (■). The lines are guides to the eye.

The experimental results for the $U_{1-x}Gd_xPt_3$ series are shown in Fig. 1. The resistivity data were normalized at room temperature to a value of 240 μΩ cm (the resistivity of UPt_3 in the hexagonal plane, ρ_{ab}), because of preferred directions of the crystallites in the polycrystalline samples. Also, the effect of small concentrations of impurities ($x \leqslant 0.005$) on $\rho(300\ \mathrm{K})$ is negligible. In the normal state, for $T < 1$ K, the resistivity follows the Fermi-liquid behaviour: $\rho(T) = \rho_0 + AT^2$. Typical parameters deduced from the data in Fig. 1 are listed in Table 1. The superconducting transition temperature T_c^+ is determined by taking the average of the temperatures at which the resistivity has dropped by 10% and 90%. T_c^+ as well as ρ_0 vary linearly with Gd content: $\mathrm{d}T_c^+/\mathrm{d}x = -0.62$ K/at% Gd and $\mathrm{d}\rho_0/\mathrm{d}x = 13.5$ μΩ cm/at% Gd, hence $\mathrm{d}T_c^+/\mathrm{d}\rho_0 = -0.046$ K/ μΩ cm. The superconducting transition broadens with increasing Gd content from 13 mK for $x = 0$ up to 59 mK for $x = 0.0050$, which indicates that the samples become less homogeneous. The A parameter gradually increases with Gd content. In Figs. 2 and 3, we have plotted $T_c^+(x)$ and $\rho_0(x)$ for Gd and compare the results with data for Y and

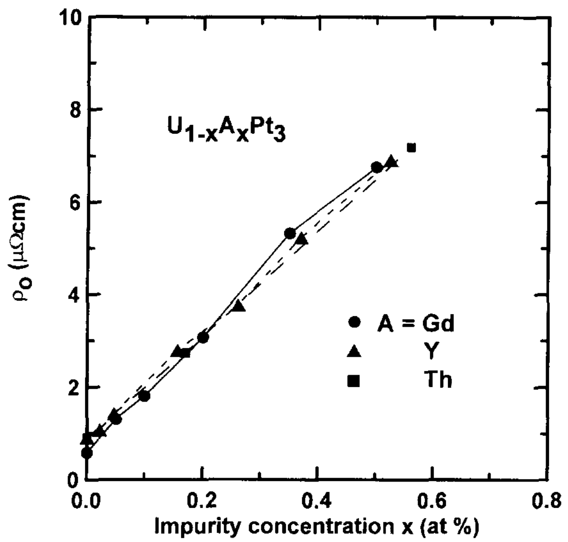

Fig. 3. ρ_0 versus impurity concentration for $U_{1-x}A_xPt_3$, where A represents Gd (●), Y (▲) or Th (■). The lines are guides to the eye.

Th [1]. Surprisingly, the data for Gd follow closely the results reported for non-magnetic Y and Th impurities [1], which is in strong contrast to the behaviour observed for normal BCS superconductors. In the case of the Ni doped samples we found a progressive suppression of T_c^+ as well. However, a finite resistivity remained for $T \to 0$ K, which we attribute to a non-superconducting second phase at the grain boundaries. This finite resistivity increased with Ni content. As only the nominal value of the Ni content is known, we will not discuss these data further in the present paper.

In general, the suppression of T_c in Kondo-lattice systems must be attributed to the combined effect of scattering by the Kondo hole [4] and scattering by the impurity spin [5], leading to the Abrikosov–Gor'kov relation

$$\ln\left(\frac{T_c}{T_c(0)}\right) = \Psi\left(\frac{1}{2}\right) - \Psi\left(\frac{1}{2} + \frac{\Gamma_{tot}}{2\pi T_c}\right) \qquad (1)$$

where Ψ is the digamma function and the total scattering rate is given by $\Gamma_{tot} = \Gamma_N + \Gamma_{mag}$. The scattering rate due to the Kondo-hole effect Γ_N is given [4] by

$$\Gamma_N = \frac{n_e x}{\pi N_0}\frac{1}{1 + \cot^2\delta} \qquad (2)$$

where N_0 is the density of states, n_e is the electron concentration and δ is the phase shift. The scattering rate due to the magnetic moment Γ_{mag} is given [5] by

$$\Gamma_{mag} = \frac{\pi}{k_B} N_0 J_{exch}^2 (g-1)^2 J(J+1)x \qquad (3)$$

where J_{exch} is the exchange parameter, g is the Landé factor and J is the quantum number for the total angular momentum. The data in Fig. 2 show that the suppression of T_c^+ is independent of the type of impurity (paramagnetic versus non-magnetic). This strongly suggests that Γ_{mag} does not contribute to Γ_{tot}. In an ordinary singlet superconductor the impurity spin (Γ_{mag}) acts in an asymmetric way on the electrons of the Cooper pair, which leads to pair-breaking. In the case of UPt_3, the most appealing implication of the fact that Γ_{mag} does not contribute to Γ_{tot}, is that the impurity spins act in a symmetric way on the electrons of the Cooper pair. This can be taken as evidence for an equal spin-pairing state in the triplet manifold ($L = 1$ and $S = 1$) in the A-phase (the high-temperature low-field phase). Of course, this relies on the assumption that the exchange coupling constant, J_{exch}, remains sizeable. Therefore, subsequent investigations must focus on the magnitude of J_{exch}. ESR experiments on UPt_3 doped with rare earth impurities [6] indicate, however, that J_{exch} is small.

In summary, by doping UPt_3 with Gd, we have investigated, for the first time, the effect of a magnetic impurity on T_c^+. We observe that the effect of magnetic and non-magnetic impurities is almost identical. Under the assumption that the exchange coupling parameter remains sizeable, the results yield strong support for non-singlet superconductivity.

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