

# Impurity effects in superconducting $\text{UPt}_3$

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Superconducting  $\text{UPt}_3$  is characterized by a novel and complex magnetic field-temperature phase diagram, with two superconducting transitions at  $T_{c1}$  and  $T_{c2}$  in zero field. We have studied the effects of Pd and Y impurities on the zero field superconducting properties of  $\text{UPt}_3$ . Resistance measurements show that both dopants increase the residual resistivity and decrease the spin fluctuation temperature in the normal state.  $T_{c1}$  is depressed by both dopants, but more effectively by Pd.  $|T_{c1} - T_{c2}|$  is essentially unaffected by Y doping, but increases dramatically with Pd doping.

## INTRODUCTION

Recently, much interest has centered on the determination and understanding of the complex field and temperature dependent phase diagram of superconducting  $\text{UPt}_3$ .<sup>1</sup> An extraordinary feature of the phase diagram is the existence of two distinct field dependent transition temperatures  $T_{c1}(H)$  and  $T_{c2}(H)$  observed in specific heat,<sup>2</sup> thermal expansion,<sup>3</sup> ultrasound,<sup>4</sup> and mechanical<sup>5</sup> measurements. However, there are qualitative variations between the results obtained for different samples as well as for polycrystalline and single crystal samples. This observations suggests that impurities or perhaps their related strains and magnetic properties may play a central role in superconducting  $\text{UPt}_3$ . In order to address this issue, we report here the effects of impurities on the low temperature resistivity and specific heat of superconducting  $\text{UPt}_3$ . We have investigated not only a substitution on the U-site,  $\text{U}_{1-x}\text{Y}_x\text{Pt}_3$  ( $0 < x < 0.005$ ) but also a substitution on the Pt-site,  $\text{U}(\text{Pt}_{1-x}\text{Pd}_x)_3$  ( $0 < x < 0.002$ ). For the latter substitution, we reported earlier<sup>6</sup> a rapid suppression of  $T_{c1}$ .

## EXPERIMENTAL DETAILS

The polycrystalline samples used in these experiments were prepared by arc-melting in an argon atmosphere. Master alloys of  $\text{YPt}_3$  and pure and doped  $\text{UPt}_3$  were used to carefully control both the stoichiometry and the dopant levels present in the finished samples. The palladium-doped material (prepared in Amsterdam) was cast in a copper crucible to form cylinders suitable for resistance measurements and then annealed at 950 °C for 7 days. The yttrium-doped samples (prepared in Los Alamos) were spark-cut from the slowly cooled as-cast button and then annealed at 950 °C for 50 h. The resistivity measurements were performed in the four-probe configuration using a self-balancing ac bridge. The specific heat measurements were accomplished by the relaxation technique, and corrected for the presence of spurious relaxation times.

## EXPERIMENTAL RESULTS

The results of resistance measurements on the series of Y-doped samples are summarized in Fig. 1. In agreement with our previous report,<sup>6</sup> qualitatively similar results are obtained for the Pd-doped series. Although we find that both dopants reduce the upper superconducting transition temperature  $T_{c1}$  [see Fig. 2(a)], Fig. 1 demonstrates that the low temperature resistivity  $\rho(T)$  is well described by the Fermi liquid expression  $\rho(T) = \rho_0 + AT^2$  at the lowest temperatures. The residual resistivity  $\rho_0$  rises quickly with impurity level as shown in Fig. 2(b), with a rate of 11.5  $\mu\Omega$  cm per at. % Y, and  $\sim 9.5$   $\mu\Omega$  cm per at. % Pd. The increase in the coefficient A is small, amounting to 0.4 and 1.4  $\mu\Omega$  cm/K<sup>2</sup> per atomic percent Y and Pd impurities, respectively. Note that the values for  $\rho_0$  and A are given here after normalizing the measured resistivity to a room temperature value of 240  $\mu\Omega$  cm, appropriate for resistivity in the basal plane.<sup>7</sup> This is reasonable since preferential orientation of the crystallites occurs in the cast Pd-series and the spark-cut Y-series. The different values of  $T_{c1}$  mea-

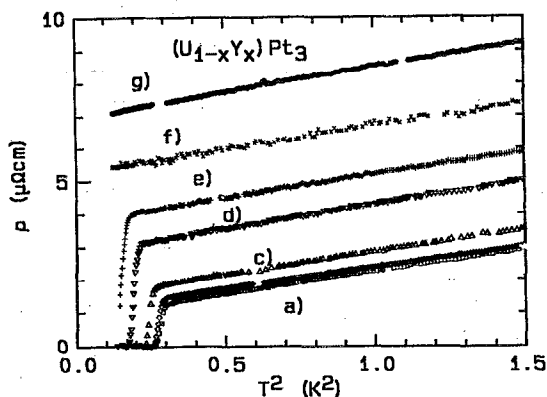


FIG. 1. Quadratic temperature dependence of the electrical resistivity of polycrystalline  $\text{U}_{1-x}\text{Y}_x\text{Pt}_3$ . (a)  $x=0$ , (b)  $x=0.00022$ , (c)  $x=0.00047$ , (d)  $x=0.0016$ , (e)  $x=0.0026$ , (f)  $x=0.0037$ , (g)  $x=0.0053$ . Resistivity normalized to room temperature value of 240  $\mu\Omega$  cm.

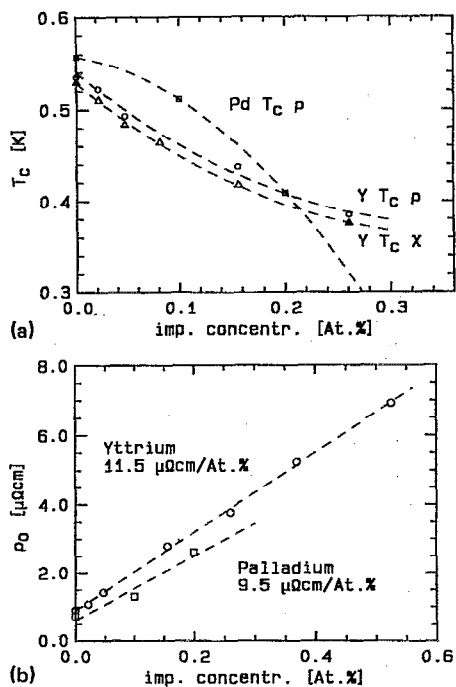


FIG. 2. (a) Depression of the superconducting onset temperature  $T_{c1}$  by Y and Pd doping.  $T_c$  taken from resistivity measurements (squares for Pd, circles for Y) and ac susceptibility measurements (triangles for Y only). (b) Dependence of the residual resistivity  $\rho_0$  on Y and Pd dopant levels. Dotted lines are guides for the eye.

sured in the two undoped  $\text{UPt}_3$  samples are likely the result of differences in the purity of the starting materials for the two series.

The results of specific heat measurements on both the Y and Pd doped series are presented in Figs. 3(a) and 3(b) respectively. The double transition is clearly evident in each of the samples measured (except the highest Y concentration), but the transitions are substantially modified by the introduction of impurities. First, both transitions are broadened, making resolution of the double peak structure increasingly difficult as the impurity level is increased. Second, the magnitude of the specific heat jump at both transitions is reduced. Finally, in agreement with the resistivity results on  $T_{c1}$ , both  $T_{c1}$  and  $T_{c2}$  drop monotonically with Y and Pd doping. However, it is striking to note that although Y impurities decrease  $T_{c1}$  and  $T_{c2}$  at approximately equal rates,  $T_{c2}$  decreases much more rapidly than  $T_{c1}$  with Pd doping.

## DISCUSSION

The double transition observed in the specific heat is thought to result from the presence of a symmetry breaking field which lifts the degeneracy of the superconducting order parameter.<sup>8</sup> Our observation that the splitting of the transition depends sensitively on the number and type of impurities present in  $\text{UPt}_3$  agrees with theoretical suggestions that the source of the symmetry breaking field is internal strain or the antiferromagnetic order observed near 5 K. Previously, large ordered moments were observed<sup>9</sup> in  $\text{U}(\text{Pt}_{1-x}\text{Pd}_x)_3$  for Pd concentrations

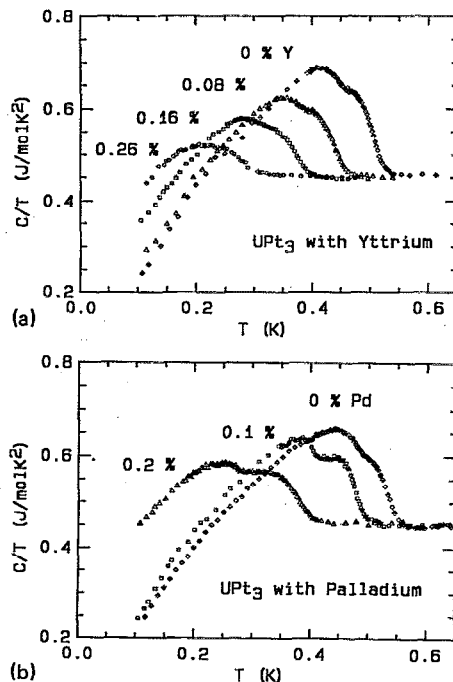


FIG. 3. Evolution of the electronic contribution of the specific heat  $\gamma = C/T$  in  $\text{UPt}_3$  as a function of (a) Y concentration and (b) Pd concentration.

$0.02 \leq x \leq 0.10$ . No indication of magnetic order was found at a comparable Y concentration.<sup>10</sup> Naively, these two observations together suggest that Y acts like a nonmagnetic impurity and Pd a magnetic impurity in  $\text{UPt}_3$ . If antiferromagnetism provides the symmetry breaking field, the zero field splitting is expected to be proportional to the square of the ordered moment,<sup>9</sup> and would increase with "magnetic" Pd doping but be insensitive to "nonmagnetic" Y impurities. This is in qualitative agreement with our experimental results, but does not completely rule out the possibility that the symmetry breaking field is related to impurity-induced strains. What is more, the presence of a double transition in nominally undoped  $\text{UPt}_3$  suggests that trace impurities or perhaps unrelaxed strains also make important but less quantifiable contributions to the symmetry breaking field.

In any case, the experimental results presented here point out that a theoretical investigation of the role of impurities in the development of antiferromagnetism and superconductivity in  $\text{UPt}_3$  is clearly required if the microscopic character of the split superconducting transition is to be fully understood.

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<sup>1</sup>For a general discussion, see L. Taillefer, *Physica B* **163**, 278 (1990).

<sup>2</sup>R. Fisher *et al.*, *Phys. Rev. Lett.* **62**, 1411 (1989); K. Hasselbach, *Phys. Rev. Lett.* **63**, 93 (1989); T. Vorenkamp, *Physica B* **163**, 564 (1990).

<sup>3</sup>K. Hasselbach *et al.* (unpublished).

<sup>4</sup>V. Muller *et al.*, *Phys. Rev. Lett.* **58**, 1224 (1987); G. J. C. L. Bruls *et al.* (unpublished).

<sup>5</sup>R. N. Kleiman *et al.*, *Phys. Rev. Lett.* **62**, 328 (1989).

<sup>6</sup>A. de Visser *et al.*, *Phys. Lett. A* **113**, 489 (1986); A. de Visser *et al.*, *J. Magn. Magn. Mater.* **76-77**, 112 (1988).

<sup>7</sup>A. de Visser *et al.*, *J. Magn. Magn. Mater.* **43**, 43 (1984).

<sup>8</sup>R. Joynt, *Supercond. Sci. Tech.* **1**, 210 (1988); D. W. Hess *et al.*, *J. Phys. Condens. Mater.* **1**, 8135 (1989); K. Machida and M. Ozaki, *J. Phys. Soc. Japan* **58**, 2244 (1989).

<sup>9</sup>A. de Visser *et al.*, *Physica B* **147**, 81 (1987).

<sup>10</sup>K. Kadowaki *et al.*, *Jpn. J. Appl. Phys.* **26-3**, 1243 (1987).