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Vorenkamp, T.; Aronson, M.C.; Koziol, Z.; Bakker, K.; Franse, J.J.M.; Smith, J.L.

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Substitution studies and the nature of superconductivity in UPt_3

T. Vorenkamp, M. C. Aronson,* Z. Koziol, K. Bakker, and J. J. M. Franse
Van der Waals-Zeeman Laboratorium, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

J. L. Smith
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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The influence of substitutions on the superconductivity in the systems $(U_{1-x}Y_x)Pt_3$, $(U_{1-x}Th_x)Pt_3$, and $U(Pt_{1-x}Pd_x)_3$ have been studied by resistance and specific-heat measurements for $T < 1$ K. The results of yttrium and thorium substitution are discussed in terms of the models developed by Hirschfeld *et al.* and Schmitt-Rink *et al.* in which the power laws in the low-temperature behavior of several properties are calculated under the assumption of resonant impurity scattering. Both yttrium and palladium are concluded to have a pair-breaking effect in UPt_3. For palladium substitution an increase of $\Delta T_c = T_{c1} - T_{c2}$ with concentration is reported. The importance of these results for the question of the nature of the order parameter and for the hypothesis of a coupling between the superconducting and magnetic order parameter is discussed.

I. INTRODUCTION

In the 1980s a new class of intermetallics was discovered, presently known as heavy-fermion compounds.[1] At low temperatures these materials are characterized by conduction electrons with very large effective masses (m^* is 100–1000 times the free-electron mass), as follows from the specific-heat coefficient γ of the electronic specific heat ($C_e = \gamma T$). At present, six heavy-fermion compounds are known to have a superconducting ground state. Of these compounds, UPt_3 has especially attracted wide attention due to claims of unconventional superconductivity.[2] To the remarkable properties of UPt_3 belong a multicomponent superconducting phase diagram,[3,4] with two superconducting transitions at T_{c1} and T_{c2} in zero field ($T_{c2} < T_{c1}$ and $\Delta T_c = 55$ mK) (Refs. 5–7) and a kink in both $H_{c2}(T)$ (Refs. 8 and 9) and $H_{c1}(T)$.[10,11] Recently, various elaborations of the Ginzburg-Landau theory have been developed[12] in an attempt to explain the aforementioned properties. A number of these models use the assumption that a coupling exists between the superconducting order parameter and the small ($0.02\mu_B$ per U atom) antiferromagnetic moment of the ordering which has been observed[13,14] below 5 K . Although there is strong evidence for the nontrivial character of the superconducting order parameter of UPt_3, its precise nature is still unknown. The question whether the pairing is d-wave-like or p-wave-like is still subject to controversy.[2] (By p-wave-like we mean anisotropic pairing with an odd-parity order parameter of which the p wave is the analog in a system with rotational freedom such as superfluid ^{3}He. By d-wave-like we mean anisotropic pairing with an even-parity order parameter of which the d wave is the analog in a system with rotational freedom.)

Properties such as the specific heat C, thermal conduction K, and longitudinal α^L and transversal α^T ultrasonic attenuations for different directions of the polarization $\hat{\mathbf{e}}$ of the ultrasound, nuclear spin-lattice relaxation time T_1, and penetration depth λ reveal a power-law temperature dependence in the superconducting state which is characteristic[2] of an unconventional superconductor with nodes in the quasiparticle energy gap. In principle, the value of the power in these power laws provides information on the presence of line or point nodes in the gap.[2] Using a classification[15] of possible order parameters, the symmetry of a gap node can be related to the symmetry of the order parameter. A short overview of the power-law problem will be given below. In early investigations the data were compared with predictions for the polar state (having a line node in the basal plane) and the axial state (having two point nodes at the poles, where the hexagonal axis intersects the Fermi sphere). However, various experiments gave no consistency in the gap structure. The problems are illustrated with an overview in Table I.

(i) There seems to be a sample dependence for the measurements of the ultrasonic attenuation α and the penetration depth λ. For the specific-heat and thermal-conduction measurements there is no pure power law and the value of $\gamma(0)$ is sample dependent.

(ii) In the measurements of the penetration depth, there is a different result for dc and rf measurements.

(iii) The predictions for the polar state and the axial state, without impurity scattering, cannot explain the experimental data.

One possible explanation for the inconsistency is that the temperature range in some of the experiments did not extend to low enough values to derive the genuine exponents. In the case of UPt_3, this explanation is not

TABLE I. Overview of the low-temperature power laws in several properties of UPt_3. For definition of symbols see text. The experimental results (Expt. results) are compared with the predictions for a conventional gapless superconductor and with the predictions for an unconventional superconductor with a polar gap [line node at the equator (x, y plane) of the Fermi sphere] or with an axial gap (point nodes at the poles where the hexagonal axis intersects the Fermi sphere). The abbreviation "no Imp." indicates that impurity scattering has not been taken into account. The "dc" stands for dc and "rf" for rf measurements; and k denotes a constant. The measurements as well as the predictions of α are in the hydrodynamic limit ($ql \ll 1$, with q the ultrasound wave vector, l the electron mean free path).

Power laws	α^L $\hat{\mathbf{e}} \parallel c$	α^T $\hat{\mathbf{e}} \parallel c$	α^T $\hat{\mathbf{e}} \perp c$	K	C	$1/T_1$	λ dc	λ rf
Expt.	T^2 (Ref. 16)	T^3 (Ref. 17)	T (Ref. 17)	$K'(0)T + \beta T^2$	$\gamma T + \alpha T^2$	T^3	T^2 (Ref. 18)	T^4 (Ref. 19)
results	T^3 (Ref. 20)	T^3 (Ref. 20)		(Ref. 21)	(Refs. 5, 21, 22)	(Ref. 23)	T (Ref. 24)	T^2 (Ref. 24)
Conv. gapless		$c + T^2$ (Ref. 25)		T (Ref. 25)	T (Ref. 26)	T (Ref. 26)	$c + T^2$ (Ref. 25)	
Polar no imp.	T^2 (Ref. 16)			T^2	T^2 (Ref. 26)	T^3 (Ref. 26)	T (Ref. 27)	
Axial no imp.	T^4 (Ref. 16)			T^4	T^3 (Ref. 26)	T^5 (Ref. 26)	T^2 (Ref. 27)	

very appealing because in many cases the power-law dependence is clearly present over a wide range in temperature: $0.1 \leq T/T_c \leq 0.6$. A far more likely explanation is that the discrepancy can be solved by taking into account impurity scattering. Pair breaking due to impurity scattering changes the minimum excitation energy and the density of states and therefore the shape of the low-temperature behavior of several properties.[2] The influence of impurity scattering has been calculated in the Born approximation by Pethick and Pines.[28] The results reveal that the T dependences of α and K are not different from those in the normal state: A temperature-independent ultrasonic attenuation for all orientations of the wave vector and polarization and a thermal conduction proportional to T prevails. Both results are in conflict with experiment. Pethick and Pines argued that the Born approximation failed due to the presence of high scattering phase shifts which are related to resonant impurity scattering. Schmitt-Rink, Miyake, and Varma,[29] Miyake,[30] and Hirschfeld and co-workers[31,32] developed self-consistent models using the T-matrix approximation for the influence of resonant impurity scattering on the low-temperature dependence of several properties. Putikka, Hirschfeld, and Wölfle[33] discussed calculations on the influence of resonant impurity scattering on the dc and rf electromagnetic response. The results are the following: From the calculations of Schmitt-Rink, Miyake, and Varma[29] it follows that the data of C, K, and α^T are more consistent with a polar state than with an axial state. Also the $1/T_1$ data are more close to predictions for a polar state as follows from the calculations of Hirschfeld, Wölfle, and Einzel.[32] The $\alpha_L \propto T^3$ is shown by Miyake and Varma[34] to be close to the predictions of a polar state, under assumption of the occurrence of the Landau-Khalatnikov mechanism associated with the relaxation of the order-parameter amplitude. A strong argument for this assumption is that it also explains the hitherto unexplained peak in α^L near T_c observed by Müller *et al.*[20] in the same measurement. Especially the α^T data[17] of Shivaram *et al.*, $\alpha^T(\hat{\mathbf{e}} \parallel c) \propto T^3$ and $\alpha^T(\hat{\mathbf{e}} \perp c) \propto T$, and the calculations of Schmitt-Rink, Miyake, and Varma[29] and Hirschfeld, Vollhardt, and Wölfle[31] make clear that the gap structure is dominated by gap nodes in the basal plane. From the classification[15] of possible order parameters in a hexagonal lattice by Volovik and Gor'kov, which takes into account spin-orbit coupling, it follows that this can only be an order parameter with a line node in the basal plane. This leads to a E_{1g} representation, of which the order parameter which is stable at low temperatures is the $(1,i)$ type which has a hybrid gap. (The hybrid gap has a line node in the basal plane and two point nodes at the poles of the Fermi sphere.) From the classification by Volovik and Gor'kov[15] it follows that p-wave-like states with a line node do not exist in a hexagonal lattice. Therefore it is tempting to conclude that UPt_3 is a d-wave-like superconductor. However, as recently has been noted by Machida and Ozaki,[35] it cannot be excluded that a classification of order parameters should be used in which spin-orbit coupling is taken to be negligibly small. According to Miyake[36] the possibility exists that the effective spin-orbit coupling for Cooper pairs is weak, although individual quasiparticles near the Fermi level are subject to strong spin-orbit coupling. From the classification of order parameters by Ozaki and Machida[37] it follows that a gap state dominated by nodes in the basal plane is found in an order parameter of the one-dimensional A_{2u} representation, which also has a hybrid gap. We note that the polar and axial states are known from superfluid ^{3}He and represent the two most simple gap states. In fact, they do not represent states belonging to order parameters that are stable at low temperatures in a hexagonal lattice. The general conclusion from the aforementioned early calculations, that the data of C, K, α^T, α^L, and $1/T_1$ are more consistent with the polar than the axial state and that a line node in the basal plane appears to be required, excludes the possibility of a gap with only point nodes, but does not exclude the possibility of the hybrid gap, which, according to the classification of order parameters, belongs to a possible order parameter for the hexagonal lattice. In the later calculations of Hirschfeld, Wölfle, and Einzel[32] for C and K the hybrid gap state has been included. The result is that no clean power law is expected in the temperature dependence above the gapless regime ($T \geq 0.1T_c$). However, within a limited range of temperature ($0.16T_c \leq T \leq 0.6T_c$) the behavior for hybrid- and polar-gap states can fit equally well the experimentally observed dependences

$$K = K_0'T + \alpha T^2 \quad \text{and} \quad C = \gamma(0)T + BT^2. \quad (1.1)$$

The K_0' and $\gamma(0)$ in this fit are explained by resonant impurity scattering and are predicted to increase with impurity concentration. Apart from these experiments which suggest a hybrid gap, or at least are not inconsistent with a hybrid gap, there are a few experiments that clearly speak in favor of a hybrid gap: (i) The rf measurement of λ in a nominally pure sample can only lead to a T^4 dependence in case of a hybrid gap as follows from the calculations of Putikka, Hirschfeld, and Wölfle[33]. (ii) The μSR penetration-depth measurement of Broholm *et al.* can only be explained[38] by a hybrid gap.

Though not all data are yet explained (the rf λ data with a T^2 dependence, because for this experimental situation no calculations are available,[24] and the α data, because no calculations for the hybrid-gap state are available), there is a growing body of evidence that (i) the sample dependency of the power laws in the temperature dependences of several properties can be explained taking into account resonant impurity scattering. (ii) The frequency dependence of the results for the penetration depth, as derived from the measurement of the electromagnetic response, can be explained[33] with amplification of the shielding current by rf-field excitation of quasiparticles near the gap node, which is broadened due to resonant impurity scattering. (iii) The superconducting order parameter of UPt_3 has a hybrid gap.

Because of the aforementioned uncertainty which classification of order parameters is appropriate, the question whether the order parameter is d-wave-like or p-wave-like is not solved and two possibilities remain: (i) a two-dimensional E_{1g} (d-wave-like) order parameter[39,40] under the assumption of strong spin-orbit coupling and (ii) a one-dimensional A_{2u} (p-wave-like) order parameter[41,42] under the assumption of weak spin-orbit coupling.

Apart from the power laws, there are two other properties, both related to the parallel spins of the Cooper pair in the triplet state, which in principle should distinguish between p-wave-like and d-wave-like pairing. One is the paramagnetic limiting of the upper critical field. Recently Choi and Sauls[43] used the interplay of the paramagnetic effect, the strong spin-orbit coupling, and the spin of odd-parity pairs to explain the "crossing" at low temperatures of the upper critical field lines for $\mathbf{H} \perp c$ and $\mathbf{H} \parallel c$. They interpreted this "crossing" as evidence for p-wave-like pairing. The second property is the spin susceptibility in the superconducting state. The rotation of the polarized-muon spin due to the local field experienced by the muon in a μ^+SR measurement allows one to measure a muon Knight shift. This Knight shift has been observed by Luke *et al.*[44] to remain constant (within an error bar of 10%) below T_c. The authors use this measurement as argument for p-wave-like superconductivity. However, the explanations of the "crossing" by Choi and Sauls and of the constant muon Knight shift by Luke *et al.*, in terms of p-wave-like pairing, are not consistent with each other. This has the following reason: For the explanation of Choi and Sauls, using the anisotropy in the paramagnetic limiting of H_{c2}, it is necessary that the spins of the state have a direction in the basal plane (which is the case for the triplet E_{1u} and B_{1u} states assuming strong spin-orbit coupling). In the Knight-shift experiment of Luke *et al.* the direction of the field was along the c axis. Therefore, the spins of p-wave-like states, with spin direction in the basal plane, cannot contribute to the susceptibility and thus cannot explain a constant Knight shift below T_c.

Since one cannot decide on basis of the presently available experimental evidence, further experiments are needed that give direct information about the symmetry of the order parameter.

In the present paper we report on substitution studies with yttrium and palladium. Yttrium is a nonmagnetic substitution at the uranium site. Palladium is a substitution on the platinum site, which is expected to induce a more localized character in the $5f$ electrons of uranium. Moreover, for concentrations between 1 and 5 at.%, palladium is known to increase the antiferromagnetic-ordered moment at the uranium site.[45] Substitution studies are of interest for the understanding of the superconductivity in UPt_3 for the following reasons.

(i) Nonmagnetic impurities have a pair-weakening effect in conventional superconductors[46] and a pair-breaking effect in d-wave-like and p-wave-like superconductors.[2] Moreover, the depression of T_c is of qualitatively different character for conventional superconductors and superconductors with anisotropic pairing.[47] Therefore, substitution with nonmagnetic impurities is a direct probe for anisotropic pairing.

(ii) If the substitution is at the uranium site, it can give information about the hypothesis of nonmagnetic resonant impurity scattering and about the validity of the predictions of the Hirschfeld-Wölfle-Einzel model.[32]

(iii) Until now, the general attention on the effect of impurities in UPt_3 has been focused exclusively on nonmagnetic impurities. Magnetic impurities however, are of prominent interest. As became clear from the work of Maki[25] and de Gennes,[48] pair breaking results from a perturbation that breaks time-reversal symmetry. This is valid in the case of a standard BCS superconductor for which the Cooper pair consists of two electrons which are, with respect to each other, in a time-reversed state. When we reformulate this for the more general case which includes anisotropic pairing, the principle is that pair breaking results from a perturbation that breaks the symmetry of the Cooper-pair wave function. The Hamiltonian for magnetic pair breaking,[49] $H = -J\mathbf{S} \cdot \mathbf{s}$, breaks the spin symmetry of the Cooper pair in the case of a singlet superconductor but not in the case of a triplet superconductor. Because of the Pauli principle, the spin symmetry of the Cooper pair is directly coupled to the parity of the order parameter. From this, it is clear that pair breaking due to magnetic impurities might be useful as a probe of the symmetry of the order parameter. The importance of such a probe has been stressed above.

(iv) In the case that substitution increases the antiferromagnetic-ordered moment at the uranium site, the impurity study directly probes the hypothetical coupling between the superconducting order parameter and this ordered moment.

We note that for points (iii) and (iv), apart from results

of substitution studies, also a more precise knowledge of the magnetism in UPt_3 is needed.

II. EXPERIMENTAL RESULTS

The superconductivity of a series of $(U_{1-x}Y_x)Pt_3$, $(U_{1-x}Th_x)Pt_3$, and $U(Pt_{1-x}Pd_x)_3$ compounds has been studied by means of resistance and specific-heat measurements for temperatures below 1 K.

Polycrystalline samples have been prepared by arc melting the appropriate amounts of the constituents in a titanium-gettered argon atmosphere. In view of the low impurity levels that were needed, special care has been taken to guarantee impurity homogeneity. This has been done by making use of master alloys (with a high impurity concentration) which were diluted to obtain the low concentrations. The buttons were turned over and remelted several times to ensure homogeneity. In the case of the Y samples the buttons were slowly cooled in high vacuum to reduce strains. Afterwards the samples were cut from the button by the spark erosion technique. In the case of the Pd and Th samples the melt was cast into a water-cooled copper crucible to obtain a cylindrical shape. In this case, the samples were annealed at 950 °C for a period of 7 days to reduce strains. To prevent reactions of the sample with air or container material, the sample was wrapped in tantalum foil and put in an evacuated quartz ampoule which was gettered with a piece of uranium. Resistivity has been measured using an ac technique with a transformer-coupled resistance bridge, and the specific heat has been measured using a relaxation method.

The results of the resistance measurements are summarized in Fig. 1 for the yttrium-doped samples. The curves for the palladium-doped samples are similar. The figure demonstrates that the temperature dependence of the resistivity for $T <\sim 1.2$ K is well described by a Fermi-liquid behavior:

$$\rho(T) = \rho_0 + A\,T^2. \tag{2.1}$$

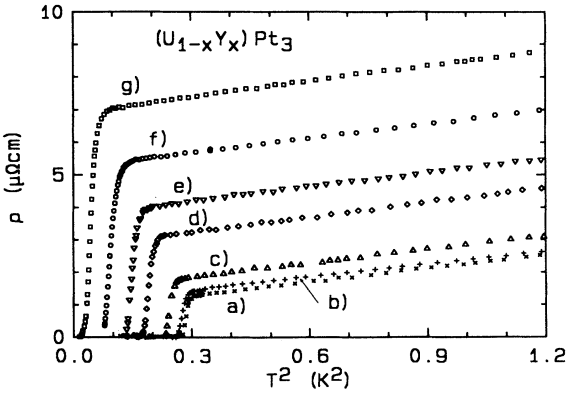

FIG. 1. Temperature dependence of the resistivity of polycrystalline $(U_{1-x}Y_x)Pt_3$ in a plot of ρ vs T^2 with (a) $x = 0.00$, (b) $x = 0.00022$, (c) $x = 0.00047$, (d) $x = 0.0016$, (e) $x = 0.0026$, (f) $x = 0.0037$, (g) $x = 0.0053$. The resistivity is normalized to a room-temperature value of 240 $\mu\Omega$ cm.

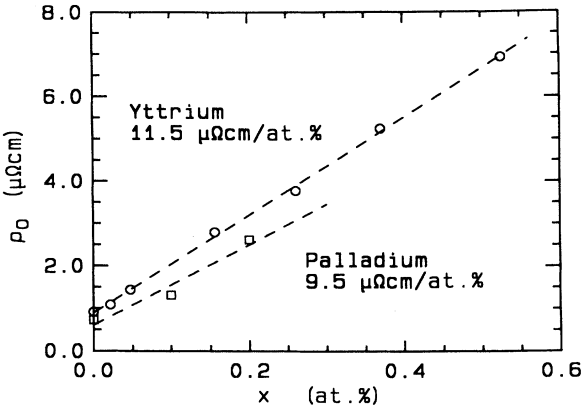

FIG. 2. Dependence of the residual resistivity ρ_0 on dopant concentration x for yttrium (○) and palladium (□). Lines are guides to the eye.

Note that, to eliminate the uncertainty in ρ_0 and A which arises from estimating the geometrical factor, the resistivity value at room temperature is normalized to $\rho(300\text{ K}) = 240\ \mu\Omega$ cm, appropriate for resistivity in the basal plane.[50] This is reasonable because of the preferential orientation of the crystallites in the samples of the cast palladium series and the spark-cut yttrium series. As shown in Fig. 2, for both dopants the residual resistivity ρ_0 increases strongly with impurity concentration at a rate of 11.5 $\mu\Omega$ cm/at. % yttrium and 9.5 $\mu\Omega$ cm/at. % palladium. The dopant concentration dependence of the coefficient A of the quadratic term of $\rho(T)$ is shown in Fig. 3. For yttrium the increase in A is very small: 0.4 (1) $\mu\Omega\,\text{cm}\,\text{K}^{-2}$/at. %. For palladium the increase in the A coefficient is larger: 1.4 $\mu\Omega\,\text{cm}\,\text{K}^{-2}$/at. %.

The superconducting transition temperature is rapidly depressed by doping with yttrium or palladium. In Fig. 4, the transition temperature T_c (obtained from the midpoint of the resistive transition) as a function of impurity concentration is shown. For the 0.5 at. % palladium alloy the susceptibility below 1 K has been measured by de

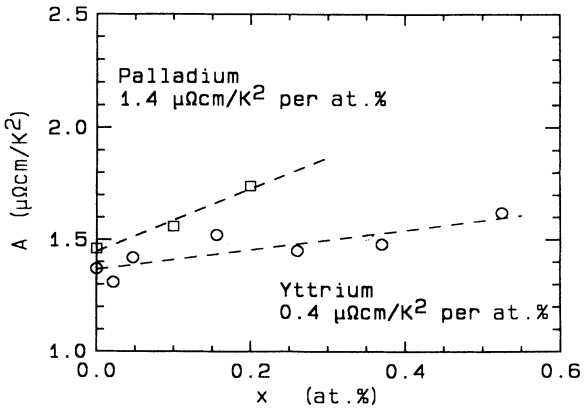

FIG. 3. Dependence of the A coefficient of the resistivity $[\rho(T) = \rho_0 + AT^2]$ on dopant concentration x for yttrium (○) and palladium (□). Lines are guides to the eye.

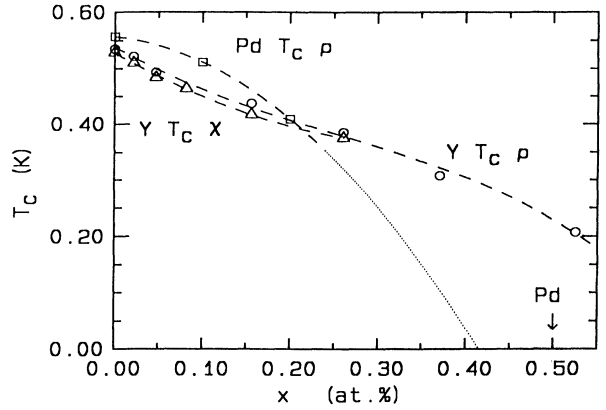

FIG. 4. Depression of the superconducting transition temperature for Y and Pd doping. T_c obtained from resistivity (ρ) measurements for Pd (□) and for Y (○) and ac-susceptibility (χ) measurements for Y (△).

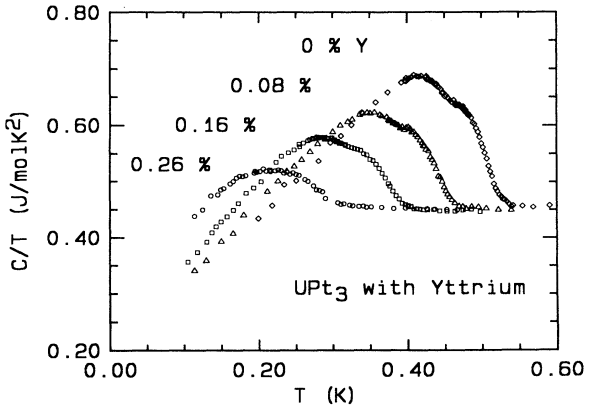

FIG. 5. Evolution of the temperature dependence of the electronic specific heat C/T in $(U_{1-x}Y_x)Pt_3$ as a function of Y concentration, with $x = 0$ (◇), $x = 0.0008$ (△), $x = 0.0016$ (□), $x = 0.0026$ (○).

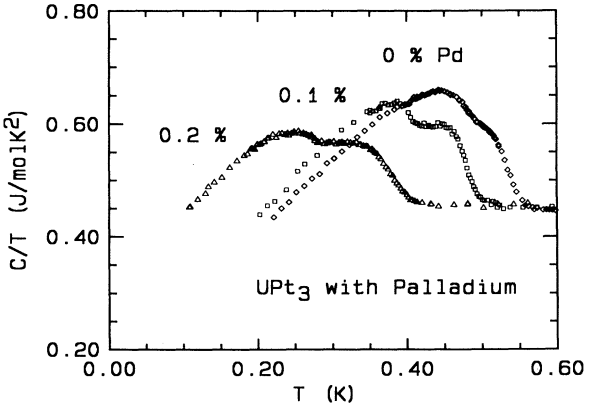

FIG. 6. Evolution of the temperature dependence of the electronic specific heat C/T in $U(Pt_{1-x}Pd_x)_3$ as a function of Pd concentration, with $x = 0$ (◇), $x = 0.001$ (□), $x = 0.002$ (△).

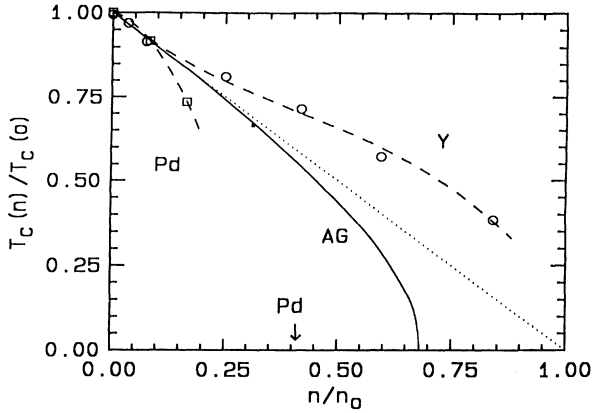

FIG. 7. Reduced transition temperature T_c/T_{c0} vs the reduced dopant concentration n/n_0. T_c is obtained from the resistivity measurements with T_{c0} the value for the undoped sample. The value of n_0 is determined by the condition that for each curve the same linear depression at low concentrations is obtained. The solid line (AG) denotes the Abrikosov-Gor'kov behavior. The dashed lines are guides to the eye; the dotted line denotes the slope of the linear depression at low concentration.

Visser *et al.*[45] The arrow in Fig. 4 indicates that for this concentration of palladium the superconducting transition is not observed down to 40 mK. For yttrium concentrations up to 0.26 at. % additional T_c values are presented, as obtained from ac-susceptibility measurements performed at Los Alamos. The slightly different values of T_c and ρ_0 for the two undoped samples of UPt_3 are attributed to differences in the purity of the starting materials for the two series. The depression of T_c is clearly of different character for yttrium and palladium.

Preliminary results of the specific-heat measurements

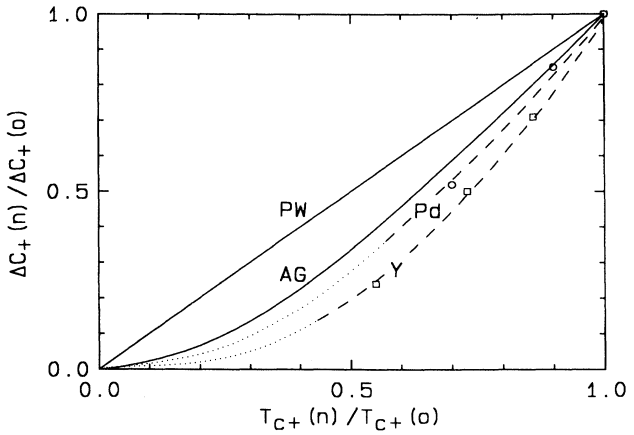

FIG. 8. Reduced jump $\Delta C/\Delta C_0$ in the specific heat as a function of the reduced transition temperature T_c/T_{c0}. ΔC_0 and T_{c0} are the values for the undoped sample. The solid line PW denotes the behavior for pair weakening, and the solid line AG represents Abrikosov-Gor'kov behavior. The dashed lines are guides to the eye, and the dotted lines are extrapolations to 0 K.

TABLE II. Values of the parameters which are derived from analysis of the specific-heat data with the idealized double-peak construction as described by Vorenkamp *et al.* (Ref. 55), for different concentrations of yttrium or palladium dopant.

Compound	$(U_{1-x}Y_x)Pt_3$				$U(Pt_{1-x}Pd_x)_3$		
x	0.00	0.08	0.16	0.26	0.00	0.10	0.20
T_{c*} (mK)	429	357	287	201	451	387	254
T_{c+} (mK)	502	434	364	277	528	477	371
ΔT_c	73	77	77	76	76	90	117
$\Delta C_*/T_{c*}$	0.284	0.218	0.173	0.103	0.259	0.234	0.177
$\Delta C_+/T_{c+}$	0.207	0.171	0.142	0.092	0.182	0.172	0.134

were presented[51] in an earlier paper. Since then, in the data for 0 and 0.08 at. % Y and 0 and 0.1 at. % Pd, for T below 200 mK, a systematic error in the measurement of the thermal conduction of the heat link was found. The 0.08 at. % Y sample therefore has been remeasured. For 0 at. % Y and 0 and 0.1 at. % Pd, the data for T below 200 mK are not taken into account for further analysis.

The specific-heat data are presented in Fig. 5 for the yttrium-doped samples and in Fig. 6 for the palladium-doped samples. From these figures it is clear that for both dopants with increasing concentration the transition temperatures and the jump in the specific heat ΔC at T_c are reduced. The specific heat in the normal state for $T < 5$ K can be expressed[52] as

$$C_n = \gamma_n T + \delta T^3 \ln T + \varepsilon T^3. \tag{2.2}$$

The specific heat in the superconducting state can be expressed as

$$C_s = \gamma_0 T + BT^2 \qquad \text{or} \qquad \gamma_s = \gamma_0 + BT. \tag{2.3}$$

Within the absolute accuracy of the measurement, no important change in γ_n with concentration is observed. The value for γ_0, however, is clearly increasing with concentration. The double transition (T_{c1} and T_{c2}) is clearly visible for each of the samples, except for the highest yttrium concentration. The most remarkable outcome is that $\Delta T_c = T_{c1} - T_{c2}$ remains constant for yttrium doping, whereas, in contrast, for palladium doping it increases with concentration. The character of pair breaking can be studied by plotting the relative depression of ΔC as a function of the relative depression of T_c and the relative depression of T_c as a function of the relative concentration variation.[53,54]

Using the values of T_{c+} and ΔC_+ that are obtained from an idealized double-peak construction,[55] these plots have been made, and a comparison is made with the curves for pair weakening and for pair breaking according to Abrikosov-Gor'kov theory.[49,56] The results are presented in Fig. 7 and Fig. 8. The values obtained from the constructions are summarized in Table II.

III. DISCUSSION

A. Substitution of yttrium or thorium on the U site

As mentioned before, yttrium is assumed to be a nonmagnetic impurity at the uranium site and thus a "Kondo hole." Yttrium does not have a partially filled f-electron shell, and yttrium substitution, therefore, amounts to removing the moments related to the $5f$ electrons of replaced uranium.

Figure 8 shows the relative depression of ΔC as a function of the relative change in T_c for yttrium substitution. The straight line indicates the situation that ΔC is depressed by the same amount as T_c, which is characteristic for pair weakening. For the yttrium impurities it is clear that ΔC is depressed at a higher rate than T_c, which is the signature of pair breaking.[53,54] As is well known from the Anderson theorem,[46] nonmagnetic impurities cause only pair weakening in s-wave-like superconductors. As shown by Chi and Carbotte,[57] there is one exception, namely, an s-wave-like superconductor with a strongly anisotropic gap, which is "mimicking" an anisotropic pairing superconductor. However, Maki and Huang[47] have shown in this case, for nonmagnetic impurities, even in the limit of resonant scattering, that the T_c depression remains limited and that T_c remains nonvanishing at high concentrations. This behavior is clearly in contrast with the observed depression of T_c with yttrium, which has a negative curvature at higher concentrations (see Fig. 4). Therefore, it is concluded that the character of pair breaking by yttrium impurities evidences unconventional superconductivity.

The models of Hirschfeld, Wölfle, and Einzel[32] and Schmitt-Rink, Miyake, and Varma[29] for the low-temperature behavior of the specific heat and the transport properties of UPt_3 are based on the assumption of resonant impurity scattering. This has been justified with the following argument:[29] In the Kondo lattice of the heavy-fermion compound, each magnetic ion leads to a scattering phase shift $\delta_0 = \frac{1}{2}\pi$. The net effect, however, is zero due to the periodicity, because the resistivity of a periodic lattice is zero at zero temperature. Therefore, a nonmagnetic ion in such a lattice (a "Kondo hole") would appear to offer a phase shift of $\frac{1}{2}\pi$ with respect to the background.

The increase in residual resistance ρ_0 as a function of the concentration of impurities is very high for Y dopants (11.5 $\mu\Omega$ cm/at. %). To investigate whether this can be explained by s-wave scattering in the unitarity limit (scattering phase shift $\delta_0 = \frac{1}{2}\pi$) we make use of the standard phase-shift expression for the resistivity.[58] As has been argued by Abrikosov[59] in his account on the Fermi-liquid theory of the Kondo problem at low temperatures, for $T \ll T_k$ the Kondo-ion spin is completely shielded; i.e., the total spin of the electron Kondo-ion complex is practically equal to zero. Therefore, the scattering interaction with this complex may be considered as a point interaction. According to Landau and Lifshitz,[60] in the

case of a point interaction, it is sufficient to take into account only s scattering. This gives us, for the residual resistivity in the unitarity limit,

$$\rho_0 = \frac{12\pi^3\hbar n_i}{e^2 k_F^4}, \tag{3.1}$$

with n_i the impurity concentration. In the derivation of this formula a spherical Fermi surface (FS) has been assumed. Using the value $k_F = 0.75$ Å^{-1} as follows from optical reflectivity measurements of Marabelli *et al.*[61] we obtain

$$\rho_0^{\text{limit}}(\text{spherical FS}) = 6.5\ \mu\Omega\,\text{cm/at.}\,\%, \tag{3.2}$$

which is much smaller than the value of 11.5 $\mu\Omega$ cm/at. % measured for yttrium substitution. This might be due to anisotropy, because the Fermi surface of UPt_3 is anisotropic with anisotropic effective masses $m^{\parallel c}$ and $m^{\perp c}$ and with $\rho_0^{\parallel c} < \rho_0^{\perp c}$. Therefore

$$\rho_0^{\text{limit}}(I \parallel c) < \rho_0^{\text{limit}}(\text{spherical FS}) < \rho_0^{\text{limit}}(I \parallel ab). \tag{3.3}$$

Though the unitarity limit for $I \parallel ab$ for the case of an anisotropic Fermi surface cannot be calculated, an upper and lower limit can be estimated because the anisotropy in ρ can be measured. From de Visser, Menovsky, and Franse[50] we obtain $\rho_0(I \parallel ab) = 1.7\rho_0(c)$. Further, we obtained above $\rho_0^{\text{measured}} = 1.77\rho_0^{\text{limit}}(\text{spherical FS})$. With these values and Eq. (3.3) it follows that

$$0.56\rho_0^{\text{measured}} < \rho_0^{\text{limit}}(I \parallel ab) < 0.96\rho_0^{\text{measured}}. \tag{3.4}$$

The result is illustrated in Fig. 9. From this we conclude that the difference between the calculated unitarity limit and measured concentration dependence of ρ_0 might be accounted for by anisotropy, and the present data for yttrium dopants cannot negate the assumption of s-wave scattering in the unitarity limit.

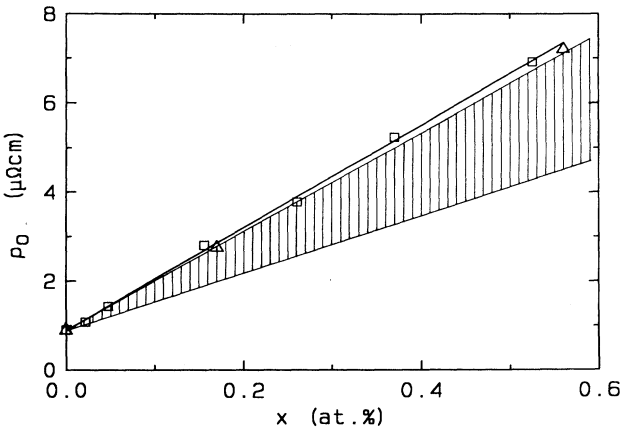

FIG. 9. Residual resistivity as a function of dopant concentration for yttrium (□) and thorium dopant (△) compared with the unitarity limit which, for current in the basal-plane direction, is within the shaded area. The solid line denotes the slope for yttrium dopants.

Because of the anisotropy of the Fermi surface, which has not been taken into account in the Hirschfeld-Wölfle-Einzel theory, the quantitative values of specific-heat jumps cannot be taken as the basis of a realistic comparison with experiment.[32] Instead of this, a comparison can be made of the qualitative behavior of the superconducting specific heat for several impurity concentrations. The experimental data of the specific heat in the superconducting state within the measured temperature range can be represented by

$$\gamma_s = \gamma_0 + BT. \tag{3.5}$$

This linear function has been fitted to the data and extrapolated to 0 K to obtain a γ_0 value. The result is shown in Fig. 10 where the data are plotted on a normalized scale. The model predicts, for data of different impurity concentrations plotted with a normalized scale, a crossing of slopes and an increase of γ_0 with increasing impurity concentration.[32] The observed increase of γ_0 with yttrium concentration gives support to the Hirschfeld-Wölfle-Einzel model and supports the assumption that the sample dependence of γ_0, which has been observed before,[5] is correlated with the content of impurities. We note, however, that the saturation of γ_s predicted by Hirschfeld, Wölfle, and Einzel[32] occurs at temperatures too low to be observable in the present experiments.

As has been explained in the beginning of this section, we conclude from Fig. 8 that yttrium causes pair breaking. The T_c depression as a function of yttrium concentration is plotted in Fig. 7. The behavior deviates clearly from the Abrikosov-Gor'kov curve which is predicted by the Hirschfeld-Wölfle-Einzel theory for resonant impurity scattering on nonmagnetic "Kondo holes." This type of deviation consisting of a positive curvature followed by a negative curvature is, however, well known from the two-band model of Entel[62] for superconductors with an

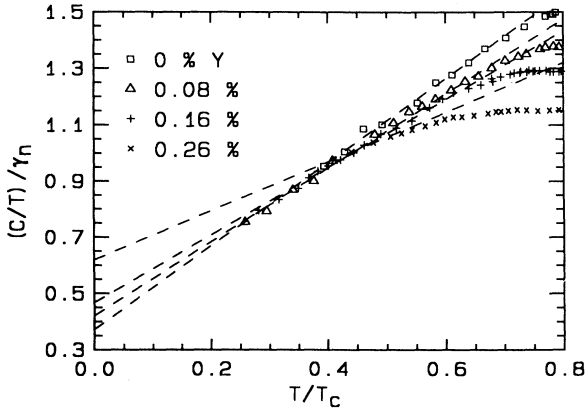

FIG. 10. Superconducting specific heat C/T normalized by $\gamma_n = C_n/T_c$ as a function of the reduced temperature T/T_c for different concentrations of yttrium dopant in $(U_{1-x}Y_x)Pt_3$, with $x = 0$ (□), $x = 0.0008$ (△), $x = 0.0016$ (+), $x = 0.0026$ (×). The dashed lines denote the slope of the fitted behavior $C/T = \gamma_0 + CT$, which is extrapolated to 0 K.

anisotropic Fermi surface. It has been shown by Entel that the occurrence of this two-band anomaly (positive curvature followed by negative curvature) is very sensitive to the amount of interband scattering. This might explain that the same type of behavior is not observed in the case of palladium dopants.

The depression of ΔC as a function of T_c for yttrium impurities is shown in Fig. 8. The data for yttrium are well below the Abrikosov-Gor'kov (AG) curve. A comparison with the Hirschfeld-Wölfle-Einzel model cannot be made, because the depression of $\Delta C/\Delta C_0$ has not been calculated.

It is of interest to compare the influence of yttrium and thorium impurities. Thorium replaces uranium in UPt_3 with phase purity for concentrations up to at least 10 at.%. The atomic volumes in a metallic environment of thorium and yttrium agree within a few percent. Because thorium does not have a partially filled f shell, just as in the case of yttrium, the $5f$ moment of uranium is removed by substitution. Thorium and yttrium differ in the sense that thorium has one more valence electron.

In Fig. 11 specific-heat measurements are shown for 0 at.% and 0.17 at.% thorium. The behavior is very similar to that for yttrium impurities and clearly different from that of palladium impurities, which show an increase of ΔT_c. Comparison of the concentration dependence of ρ_0 for thorium and for yttrium in Fig. 9 shows that they are nearly identical. Also the depression of T_c by thorium, presented in Fig. 12, is similar to that of yttrium and different from that of palladium, for which a concentration of 0.5 at.% suppresses the superconductivity.

Apparently, at low concentrations (< 1 at.%) replacement of uranium by thorium and replacement of platinum by palladium have a different effect, while for concentrations between 1 at.% and 5 at.% both palladium and thorium induce antiferromagnetic order.[63]

The fact that the depression of the specific-heat anomaly and the depression of T_c with concentration are so similar for yttrium and thorium, despite the difference of one valence electron, proves that, in this small

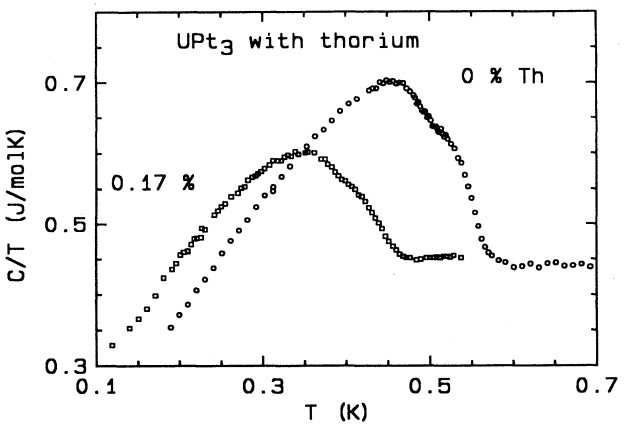

FIG. 11. Temperature dependence of the specific heat C/T for polycrystalline $(U_{1-x}Th_x)Pt_3$ for $x = 0.00$ and $x = 0.0017$.

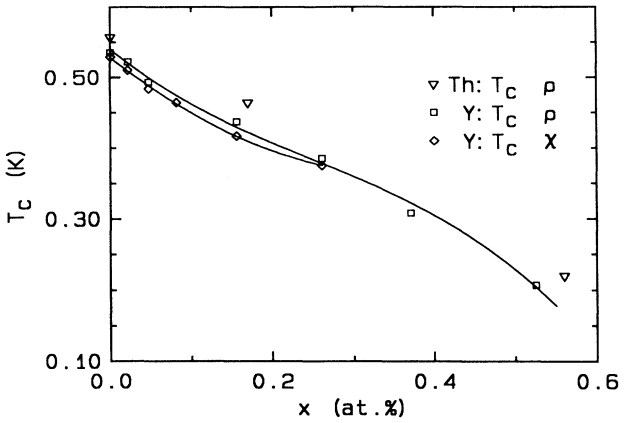

FIG. 12. Depression of T_c with dopant concentration for thorium substitution compared with that for yttrium substitution. T_c obtained from measurements of the resistivity (ρ) for Th (△) and for Y (□), and from measurements of the ac susceptibility (χ) for Y (◇).

concentration regime, there must be a strong common factor that determines the pair breaking. The most obvious common factor is that yttrium and thorium both remove the $5f$ moment of uranium by substitution. The observed results therefore support the assumption that the pair breaking is caused by resonant scattering from "Kondo holes."

B. Substitution of palladium on the Pt site

Palladium is an almost ideal substitute for platinum. First of all, palladium is isoelectronic. Moreover, the atomic volumes of palladium and platinum in a metallic environment agree within a few percent. The only clear difference is that palladium has a partially filled $4d$ shell instead of the partially filled $5d$ shell of platinum. (We note, however, that recently polycrystalline UPt_3 has been observed to contain regions of stacking defects[64–66] with a typical dimension of 25–30 Å. The influence of palladium doping on the formation of stacking faults and the relationship between these regions and the magnetic and superconducting properties is at present unknown.) From the spatial variation of the $4d$ and $5d$ orbitals one might expect that the hybridization between uranium $5f$ electrons and palladium $4d$ electrons will be smaller than that between the hybridization between uranium $5f$ electrons and platinum $5d$ electrons. From this we expect that a uranium atom with a palladium neighbor (due to substitution) has $5f$ electrons with more localized character.

From Fig. 8 it is clear that for palladium ΔC is depressed at a higher rate than T_c, which is the signature[53,54] of pair breaking. From Fig. 7 we conclude that in the case of palladium impurities the depression of T_c as a function of concentration is stronger than for the AG curve which represents the prediction[32] for resonant potential scattering by nonmagnetic impurities for anisotropic pairing. The Hamiltonian of the pair-breaking interaction can either work on (i) the orbital

part of the wave function, giving rise to pair breaking due to potential scattering, or (ii) the spin part of the wave function, giving rise to pair breaking due to magnetic scattering.

Pair breaking by nonmagnetic impurities cannot be stronger than the AG curve, since this is already in the limit of resonant scattering. Therefore, it is tempting to conclude that, in the case of palladium impurities, magnetic pair breaking must be involved. In the case that magnetic scattering has a pair-breaking effect, the pair-breaking interaction should work in an antisymmetric way on both spins of the Cooper pair, as was argued in the Introduction. This would imply an even-parity order parameter and, because isotropic (s-wave-like) pairing is excluded, points to d-wave-like pairing. However, in view of a possible spin-fluctuation-mediated pairing mechanism, it cannot be excluded that palladium substitution depresses the superconductivity by depression of the spin fluctuations. In the case that the Fermi-liquid behavior of the resistivity [$\rho(T) = \rho_0 + AT^2$] is analyzed with a spin-fluctuation model[67,68] the increase of the A coefficient with palladium concentration indicates that the spin fluctuations still increase. However, because a theoretical description of the heavy-fermion state is still lacking, it is not clear whether the Fermi-liquid behavior can be attributed to spin fluctuations. Therefore, a final conclusion cannot be drawn until more information is obtained from neutron measurements about the influence of small amounts (less than 1 at. %) of palladium on the part of the spin-fluctuation spectrum that might be attributed to the pairing mechanism. Provided that this information is available, the results of palladium substitution might give a clue to the nature of the pairing.

C. Analysis of the data with the E-representation model

In the recent work of Midgley *et al.*[69] it is shown that the presence of an incommensurate structural modulation in UPt_3, which is visible after annealing the sample at 1200 °C for 6 days, is correlated with the presence of a sharp double-peak structure in the specific heat. In an unannealed sample the modulation is not developed and the double-peak structure is not visible. We note that (i) according to our experience, in a sample without a visible double-peak structure, the double transition in the specific heat is only hidden due to the broadness of the transitions. The sample can still have a kink in H_{c2} and in H_{c1} and, derived from this, a distance between T_{c1} and T_{c2} of around 60 mK. (ii) Using the formation temperature of the modulation and the compressibility one can deduce a critical pressure that destroys the modulation. As has been claimed by Midgley *et al.*,[69] because of the high formation temperature, it is likely that the structural modulation still exists above pressures that were observed to destroy both the splitting of T_c and the ordered antiferromagnetic moment.[70,71]

We conclude that the presence of a modulation is not necessarily correlated with the presence of a splitting of T_c. Models which give an explanation of the splitting of T_c in terms of a breaking of symmetry caused by the ordered antiferromagnetic moment are still relevant.

A remarkable outcome of the specific-heat data is the increase of $\Delta T_c = T_{c1} - T_{c2}$ for palladium impurities presented in Fig. 13. One of the models proposed to explain the double superconducting transition is the E-representation model, developed in parallel by Hess, Tokuyasa, and Sauls[39,40] and Machida, Ozaki, and Ohmi.[41,42] The model makes use of a two-dimensional superconducting order parameter belonging to the E_1 or E_2 representation (using the classification for order parameters of Volovik and Gor'kov[15]). In this model the orbital degeneracy for the two components of the order parameter is lifted by a small perturbation so that two slightly different transition temperatures result. This perturbation is assumed to be due to a coupling between the superconducting order parameter and the small ($0.02\mu_B$ per U atom) antiferromagnetic moment.

In a previous paper[55] it has been shown that the parameters of the E-representation model derived from the data of (i) the double transition in $C(T)$, (ii) the kink in $H_{c1}(T)$ for $\mathbf{H} \parallel b$, and (iii) the kink in $H_{c2}(T)$ for $\mathbf{H} \parallel b$, all three measured on the same sample, are consistent with each other. It is therefore of interest to discuss the present data in terms of this model. We use the notation of Hess, Tokuyasa, and Sauls[39] with T_{c+} denoting the higher transition temperature and T_{c*} the lower one. The ΔT_c ($= T_{c+} - T_{c*}$) is given by the expression[39]

$$\Delta T_c = \left(\frac{1 + \frac{\beta_1}{\beta_2}}{\alpha_0} \right) \varepsilon, \tag{3.6}$$

with the Ginzburg-Landau (GL) parameters α_0 given by[41] $\alpha_0 = N(0)/T_{c+}$ and the ratio of GL parameters,[39]

$$\frac{\beta_2}{\beta_1} = \left(\frac{\Delta C_*}{T_{c*}} \right) \left(\frac{T_{c+}}{\Delta C_+} \right) - 1. \tag{3.7}$$

The parameter ε describes the coupling between the superconducting order parameter and the symmetry-breaking field, which is assumed to be the antiferromag-

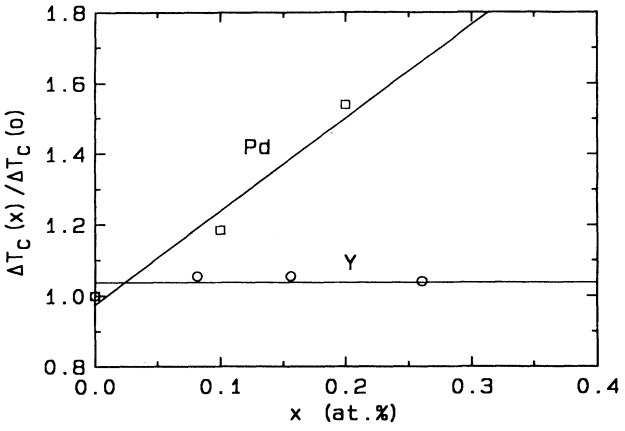

FIG. 13. Evolution of the reduced distance between the transition temperatures $\Delta T_c/\Delta T_{c0}$ (as deduced from the specific-heat measurements) with dopant concentration. ΔT_{c0} denotes the value for the undoped sample.

netic moment. With this assumption, ε is given by[41]

$$\varepsilon = cM^2, \tag{3.8}$$

with c a constant and M the spin-density-wave amplitude. $N(0)$ is the density of states at E_F in the normal state. Because no change in γ_n is observed within the limits of accuracy, we conclude that for the reported concentrations $N(0)$ remains constant. However, because of pair breaking, T_c and the peak height ΔC are strongly depressed. As a result of this, the calculated values of β_2/β_1 and α_0, and thus of ε, can vary largely for samples with different amount of impurities. This is illustrated in Table III. Of course, we note that the E-representation model does not take into account the effects of pair breaking.

The important conclusion is that the purity of the sample is important to obtain the correct values for GL-model parameters. Equation (3.6) can be written as

$$\Delta T_c = k\,\varepsilon, \tag{3.9}$$

with k being influenced by pair breaking. To apply this formula for the samples with impurities, it is possible to correct for the effect of pair breaking by keeping k constant, under the condition that ε and ΔT_c are not influenced by pair breaking.

From the definition given above it is clear that ε is not influenced by pair breaking. The ΔT_c is not influenced by pair breaking because T_{c+} and T_{c*} are depressed in the same way. This follows from the fact that at the second transition the Hamiltonian of the pair-breaking interaction remains the same, and the superconducting order parameter does not change its parity. Moreover, it is confirmed by the data for yttrium-doped samples. First of all, as was concluded from Fig. 8, yttrium impurities cause pair breaking; second, the ΔT_c value remains constant with yttrium concentration as is shown in Fig. 13. We therefore conclude from Eq. (3.9) that within the E-representation model an increase of ΔT_c with palladium concentration is directly related to an increase of ε. Let us assume that the ordered moment, which is $0.02\mu_B$/U atom in UPt_3 and $0.6\mu_B$/U atom in UPt_3 doped with 5 at. % palladium, increases proportionally to the palladium concentration. Then the prediction of Eqs. (3.9) and (3.8) that an increase of cM^2 leads to an increase of ΔT_c is consistent with the experimental observation that ΔT_c increases with palladium concentration. How reliable is this assumption? It is generally assumed that between 0 at. % and 1 at. % palladium, a spin-density-wave ordering is present, with short-range correlations.[13,41] Above 1 at. % Pd a long-range spin-density-wave ordering occurs.[45,52] Between 1 and 5 at. % palladium the peak in the specific heat at the magnetic ordering temperature increases with concentration. For a spin-density-wave ordering, ΔC at T_N is proportional to the amplitude M of the spin-density wave. Because M, and thus the ordered moment, increases with palladium concentration between 1 and 5 at. % and because the ordering in pure UPt_3 is interpreted to be a spin-density-wave ordering, it is likely that M also increases with concentration between 0 and 1 at. %.

Therefore, we conclude that the results of palladium substitution might give an important clue to the question whether a coupling exists between the superconducting and magnetic order parameter, provided that more information is obtained from neutron measurements about the development of the ordered moment with concentration of palladium, for concentrations up to 1 at. %.

IV. CONCLUSION

Results are presented of substitution studies within the systems $(U_{1-x}Y_x)Pt_3$ and $U(Pt_{1-x}Pd_x)_3$. From analyses of the relative depression of ΔC and T_c, both yttrium and palladium are concluded to have a pair-breaking effect in UPt_3. An important finding is the increase of ΔT_c with palladium concentration. This can be analyzed within the E-representation model,[39–42] which gives a possible explanation for the double superconducting transition. Within this model we conclude that the increase of ΔT_c is directly connected to an increase of the magnitude of the symmetry-breaking field which is regarded to be the antiferromagnetic moment. The quantitative behavior of the T_c and ΔC depression is not yet described by a model for UPt_3. In the Introduction, substitution studies were placed in the broader context of questions with regard to the nature of the superconductivity in UPt_3 and with regard to existing models and hypotheses. We can conclude the following.

(i) The question of the existence of anisotropic pairing in UPt_3. From the analysis of the depression of ΔC and T_c with yttrium concentration we find evidence for anisotropic pairing (unconventional superconductivity). This is consistent with the conclusions that have been drawn[2] from the observation of other unusual properties in the superconducting state.

(ii) The hypothesis of resonant impurity scattering. The observed increase of γ_0 with yttrium concentration gives support to this hypothesis and thus to the Hirschfeld-Wölfle-Einzel[32] model. Also, the observed increase of ρ_0 with yttrium concentration is consistent with resonant impurity scattering. For yttrium a remarkable depression of T_c with concentration is observed, which starts with positive curvature at low concentration and changes into negative curvature at higher concentration. This is in contrast with the prediction of the Hirschfeld-

TABLE III. Values of the parameters β_2/β_1 and ε/α_0 from the E-representation model, as derived from analysis of the specific-heat data for different concentrations of yttrium or palladium dopant.

Compound	$(U_{1-x}Y_x)Pt_3$				$U(Pt_{1-x}Pd_x)_3$		
x	0.00	0.08	0.16	0.26	0.00	0.10	0.20
β_2/β_1	0.38	0.28	0.22	0.12	0.49	0.36	0.32
ε/α_0 (mK)	20	17	14	8	25	23	28

Wölfle-Einzel model. There might, however, be an explanation for this since a qualitative similar behavior for a different type of superconductor has been explained by Entel[62] with a two-band model for an anisotropic Fermi surface.

(iii) As has been argued, palladium substitution might give information about the parity of the order parameter. The depression of T_c with palladium concentration, which is stronger than what is expected for pair breaking due to potential scattering in the resonant limit, suggests that additional pair breaking due to magnetic scattering is present. The Hamiltonian for magnetic pair breaking breaks the symmetry of the Cooper pair in case of a singlet superconductor. This suggests an even-parity or d-wave-like order parameter. However, in view of a possible spin-fluctuation-mediated pairing mechanism it cannot be excluded that the superconductivity is depressed by depression of the spin fluctuations. Therefore, a final conclusion cannot be made unless more information is obtained (from neutron measurements) about the development of the spin fluctuations with small concentrations (0–1 at.%) of palladium.

(iv) The hypothesis of a coupling between the superconducting order parameter and the magnetic order parameter. Under the assumption that M increases with palladium concentration for concentrations of 0–1 at.%, our data supports this hypothesis. However, more information from neutron measurements is needed to verify this assumption.

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*Present address: Randall Laboratory, Dept. of Physics, University of Michigan, Ann Arbor, MI 48109-1120.

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