An extremely large Grüneisen parameter ($\Gamma$) variation has been detected for the heavy-fermion alloys $U(Pt_{1-x}Pd_x)_3$, ($x < 0.15$). For pure $UPt_3$, $\Gamma(T \to 0) \approx 75$, but drops rapidly with increasing Pd content, amounting to $-300$ for $x = 0.15$. The inversion of $\Gamma$ is attributed to the weakening of the antiferromagnetic intersite correlations with increasing Pd content.

The unusual low-temperature properties of the heavy-fermion superconductor $UPt_3$ are extremely sensitive to substitutions on both the U and Pt lattice [1]. In the case of the $U(Pt_{1-x}Pd_x)_3$ alloys, a spin-density wave anomaly appears in the specific heat [2], $c(T)$, and the resistivity [3], $\rho(T)$, of the 2, 5 and 7 at% Pd samples, indicating long-range antiferromagnetic order in a small concentration range near 5% Pd, with a maximum Néel temperature ($T_N$) of 5.8 K. Neutron-diffraction experiments [4] confirmed a fairly large ordered moment of $(0.6 \pm 0.2)\mu_B/U$-atom for $x = 0.05$. The maximum in the susceptibility [2] (at 17 K for pure $UPt_3$) and the metamagnetic transition [2] (at 20 T for pure $UPt_3$) gradually disappear on alloying with Pd, and are no longer observed for a 10% compound [5]. However, the $\gamma$-value remains large up to 15% Pd [2], indicating that the heavy-fermion phenomena persist. This is furthermore supported by the appearance of a Kondo (lattice) type of resistivity curve [3] for $x = 0.10$ and 0.15. Thus, on doping with Pd, the antiferromagnetic spin-fluctuation system $UPt_3$ transforms gradually into a Kondo lattice system, while passing an intermediate antiferromagnetic long-range ordered state. Note that also pure $UPt_3$ exhibits weak antiferromagnetic order (with an ordered moment of $(0.02 \pm 0.01)\mu_B/U$-atom) [6]. It is still unclear how this small-moment state relates to the large-moment state, induced by Pd doping.

In this paper we discuss the volume effects that are related with the formation of the heavy-fermion bands in $U(Pt_{1-x}Pd_x)_3$ ($x < 0.15$). We report new data for the coefficient of thermal expansion of a single crystalline $U(Pt_{0.05}Pd_{0.10})_3$ sample, along ($\alpha_{\parallel}$) and perpendicular ($\alpha_{\perp}$) to the hexagonal axis. The data are compared with previous results, obtained for single crystalline $UPt_3$ [7] and $U(Pt_{0.95}Pd_{0.05})_3$ [8], and for a polycrystalline compound with $x = 0.15$ [1].

A single-crystalline batch of $U(Pt_{0.05}Pd_{0.10})_3$ was prepared by the tri-arc Czochralski technique. The sample was cut by means of a spar-erosion technique and shaped into a cube (edge 5 mm), in order to fit in the thermal expansion cell. The coefficient of thermal expansion, $\alpha = L^{-1} (dL/dT)$ was measured using a sensitive three terminal capacitance method in the temperature interval $0.3 < T < 100$ K. High-field specific-heat measurements on this sample can be found in ref. [9]. Another piece of the single-crystalline batch was used for high-field magnetization measurements [5].

The experimental results for $x = 0.10$ are shown in fig. 1 for $T < 30$ K, together with data for pure $UPt_3$ and $x = 0.05$. The coefficient of volume expansion is given by $\alpha_L = \alpha_{\parallel} + 2\alpha_{\perp}$. The temperature variation of $\alpha_L$ for $x = 0.00$, 0.05 and 0.10 is shown in fig. 2, where we have also plotted the data for a polycrystalline $x = 0.15$ sample. One has to bear in mind, however, that crystallites with preferred orientations are formed in polycrystalline samples. Therefore, the absolute value of $\alpha_L$ of the latter sample should only be taken as approximate. The results can be summarized as follows. For $UPt_3$, the basal plane expands on raising the temperature, while the hexagonal (c) axis contracts, resulting in a maximum in $\alpha_L$ at $T_{max} = 9$ K. For $x = 0.05$, the anisotropy is preserved and $T_{max}$ is reduced to about 6 K. The long-range order ($T_N = 5.8$ K) appears as a small negative peak superimposed on the large heavy-fermion contribution (fig. 2). However, for slightly higher Pd content (x = 0.10) the antiferromagnetic state is completely suppressed. Furthermore, a reversion of the anisotropy takes place: the basal plane...
Yosida (RKKY) interaction and the Kondo-effect. The competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the onsite Kondo interaction prevail (\(x = 0.15\)) can be described in terms of a strong antiferromagnetic spin-fluctuation phenomena that dominate to a state where Kondo-lattice properties manifest themselves. The unusually large variation of \(\alpha(T)\) as a function of pressure dependence must lead to an increase of the volume than observed for pure \(\text{UPt}_3\). It is interesting that a similar negative \(\alpha(T)\)-curve can be induced in the analog compound \(\text{CeRu}_2\text{Si}_2\) in the field region \((B > 8 \, \text{T})\), where the antiferromagnetic interactions are largely suppressed and the Kondo interactions remain. The unusually large negative low-temperature Grüneisen parameters of \(\text{UPt}_3\) alloyed with 10% and 15% Pd yield a strong decrease of the Kondo temperature with pressure, indicating a delicate Fermi-level positioning in the sharply structured density of states (the initial pressure dependence must lead to an increase of the density of states).

In conclusion \(\Gamma_{\text{eff}}(T \rightarrow 0)\) appears to be an extremely sensitive probe for the strength of the intersite and onsite interactions. It shows a dramatic variation in the heavy-fermion alloys \(\text{UPt}_{1-x}\text{Pd}_x\) \((x \leq 0.15)\) to \(\sim 75\) for pure \(\text{UPt}_3\), and approximately \(-300\) for a 15% Pd compound.

The work of one of us (AdV) has been made possible by a fellowship of the Royal Netherlands Academy of Arts and Sciences.

References