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Grüneisen parameter inversion in $U(Pt,Pd)_3$

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An extremely large Grüneisen parameter (Γ) variation has been detected for the heavy-fermion alloys $U(Pt_{1-x}Pd_x)_3$ ($x \leqslant 0.15$). 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 Γ 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 γ-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 $U(Pt_{1-x}Pd_x)_3$ ($x \leqslant 0.15$). We report new data for the coefficient of thermal expansion of a single crystalline $U(Pt_{0.9}Pd_{0.1})_3$ 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 $U(Pt_{0.90}Pd_{0.10})_3$ was prepared by the tri-arc Czochralski technique. The sample was cut by means of a spare-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 $\alpha_v = \alpha_\parallel + 2\alpha_\perp$. The temperature variation of α_v 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 α_v of the latter sample should only be taken as approximative. 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 α_v 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

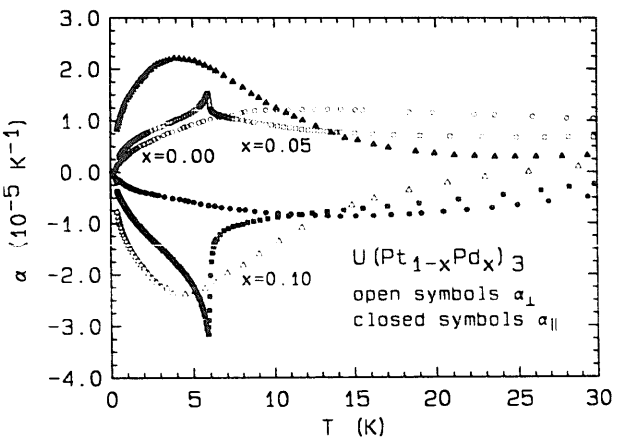

Fig. 1. Coefficient of linear thermal expansion along ($\alpha_\parallel$, closed symbols) and perpendicular ($\alpha_\perp$, open symbols) to the hexagonal axis for $U(Pt_{1-x}Pd_x)_3$ for $x = 0.00$ (○), 0.05 (□) and 0.10 (△).

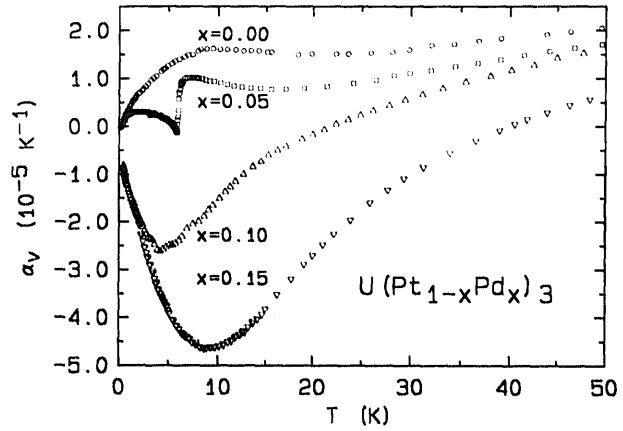

Fig. 2. Coefficient of volume expansion for $U(Pt_{1-x}Pd_x)_3$.

contracts by raising the temperature, while the c-axis expands. A large negative coefficient of volume expansion results, with a minimum at $T_{min} = 4.5$ K. For 15% Pd ($T_{min} = 9$ K) the minimum is even more pronounced (fig. 2). In fig. 3 we have plotted the effective Grüneisen parameter [11], $\Gamma_{eff}(T) = V_m\alpha_v(T)/\kappa c(T)$, where V_m is the molar volume and $\kappa = 0.48$ Mbar^{-1} is the isothermal compressibility [7]. Data for the specific heat have been obtained from refs. [1,9]. For $T \to 0$ we observe an extremely large variation of Γ_{eff} as function of Pd concentration. Note that the Grüneisen parameters are unusually large, a common feature of heavy-fermion compounds [10].

Although the $U(Pt_{1-x}Pd_x)_3$ compounds are characterized rather well experimentally, the theoretical notion of the basic interactions that bring about these peculiar low-temperature properties is far from satisfactory. The salient transition from a state ($x = 0.00$) in which strong antiferromagnetic spin-fluctuation phenomena dominate to a state where Kondo-lattice properties prevail ($x = 0.15$) can be described in terms of a competition between the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction and the Kondo-effect. The

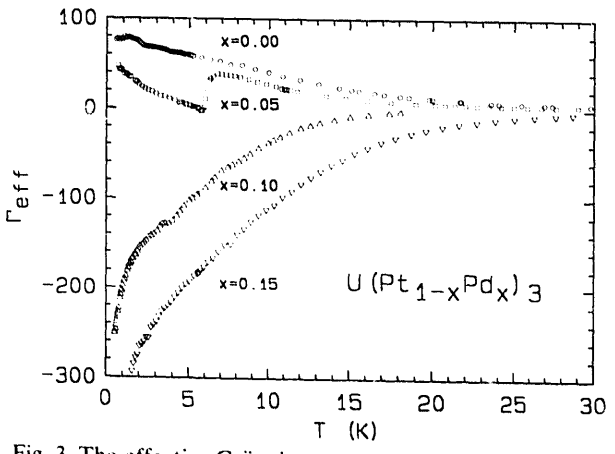

Fig. 3. The effective Grüneisen parameter for $U(Pt_{1-x}Pd_x)_3$.

general picture that emerges is that both interactions, the intersite RKKY interaction and the onsite Kondo effect, are present in pure UPt_3. Separation of these contributions is not at hand, as in general an interplay will exist. Furthermore, contributions from the crystalline electric field and the lattice are present. However, it appears that the RKKY interaction can largely by suppressed by either a strong magnetic field, i.e. a field larger than the field at which the metamagnetic transition occurs, or by substituting about 10% Pd. The suppression of the antiferromagnetic correlations in the 10% Pd compounds is evidenced by (i) the absence of the metamagnetic transition [5], (ii) the absence of a maximum in the susceptibility [11], (iii) a decrease of $c(T)$ with field [9] and (iv) a decrease of $\rho(T)$ with field [11]. This leads to the conclusion that it is mainly the remaining Kondo-lattice effect that gives rise to the negative $\alpha_v(T)$, leading to a larger ground state volume than observed for pure UPt_3. It is interesting that a similar negative $\alpha_v(T)$-curve can be induced in the analog compound $CeRu_2Si_2$ in the field region ($B > 8$ T), where the antiferromagnetic interactions are largely suppressed and the Kondo interactions remain [12]. The unusually large negative low-temperature Grüneisen parameters of 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 $\Gamma_{eff}(T \to 0)$ 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 $U(Pt_{1-x}Pd_x)_3$ ($x \leqslant 0.15$): ~ 75 for pure UPt_3 and approximately -300 for a 15% Pd compound.

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