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Transition from hexagonal to cubic structure and enhanced specific heat in $CePt_{5-x}Au_x$

A. Mielke, J.J. Rieger, E.-W. Scheidt, G.R. Stewart[1]

Institut für Physik, Universität Augsburg, Memminger Str. 6, W-86135 Augsburg, Germany

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Abstract

In our attempt to replace platinum successively by gold in $CePt_{5-x}Au_x$, we have found that with increased Au content the initial hexagonal $CaCu_5$ structure of $CePt_5$ changes into the cubic $AuBe_5$ structure for $1<x\leqslant 3$. Since this is the same lattice as in the heavy fermion system UPt_4Au, with U replaced by Ce, we can now compare the role of the two different elements in this configuration. We have investigated the influence of the Au content on the low temperature specific heat and magnetic susceptibility, which both increase with increasing Au content, and discuss to what extent this enhancement might be created by the electronic density of states or by magnetic entropy. Furthermore, we have studied the transition from the U-based heavy fermion system into the Ce-based system in $U_{1-y}Ce_yPt_4Au$.

Keywords: Hexagonal structure; Cubic structure; Specific heat

1. Introduction

Most of the known heavy fermion systems are cerium- or uranium-based metallic compounds. Thus a possible starting point to seek for the mechanisms that lead to this unconventional state is to exchange these two elements in heavy fermion systems in order to study their different influences on the system properties. For example, $CeRu_2Si_2$ with a Sommerfeld parameter $\gamma=385$ mJ mol^{-1} K^{-2} [1] shows a metamagnetic transition [2], while URu_2Si_2 reveals the coexistence of superconductivity and magnetism [3]. Since both systems crystallize in the same structure and thus possess the same ligand environment, this difference is an effect that can be traced back to the different behaviours of the two elements in this structure.

The heavy fermion compound UPt_4Au [4] has been intensively studied by doping experiments (e.g. $UPt_{5-x}Au_x$ [5], UPt_4M with $M\equiv$Au, Ag, Cu [6]). $UPt_{5-x}Au_x$, $x\leqslant 3$, crystallizes in the cubic close packed $AuBe_5$ structure (cF24) [5], which has two different Be sites [7], a fourfold and a single one ($AuBeBe_4$). The maximum in γ for $x=1$ in $UPt_{5-x}Au_x$ might lead one to expect that Au and Pt are on different atomic sites in this system, but this has not as yet been proven [8]. While we were conducting doping experiments on $CePt_5$, which crystallizes in the hexagonal $CaCu_5$ (hP6) structure [9], we discovered serendipitously that $CePt_{5-x}Au_x$, $1<x\leqslant 3$, occurs in the same $AuBe_5$ (cF24) structure as does $UPt_{5-x}Au_x$. Thus this work is another attempt to connect the different elemental properties with the properties of the respective U and Ce compounds. We also report results on $U_{1-y}Ce_yPt_4Au$.

2. Experimental details and results

The initial elements Ce (Ames), U (3N5), Pt (3N5), and Au (4N) were arc melted together under a purified argon atmosphere and remelted three times. The weight loss was less than 0.5%. The platinum-rich ($x<1.5$) and gold-rich ($x>3$) samples were annealed for 2 weeks, the others for 1 week at 900 °C.

The X-ray powder diffraction was performed with a Siemens D5000 diffractometer in Bragg–Brentano geometry with Cu radiation. The reflected beam passed a graphite monochromator permeable to Cu Kα radiation. After a numerical α_2 correction the interplanar spacings d_{hkl} were calculated based on the Cu Kα_1 radiation wavelength of 1.5409 Å. Table 1 presents the powder diffraction data of $CePt_{3.75}Au_{1.25}$ and reveals good correspondence with the cubic $AuBe_5$ structure. As demonstrated in Fig. 1, the cubic structure is stable in $CePt_{5-x}Au_x$ for $1<x\leqslant 3$ (as well as in the uranium-

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[1] Also at University of Florida, Gainesville, FL, USA.

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Table 1
Powder diffraction data of $CePt_{3.75}Au_{1.25}$, yielding a good correspondence with the cubic $AuBe_5$ structure (cF24) and no detectable second-phase peaks. The lattice parameter is $a_0 = 7.525 \pm 0.003$ Å

d_{hkl}	Relative intensity	*h k l*
4.3392	11	1 1 1
2.6582	30	2 2 0
2.2648	100	3 1 1
2.1726	47	2 2 2
1.8808	7	4 0 0
1.7237	5	3 3 1
1.5357	20	4 2 2
1.4465	36	5 1 1 / 3 3 3
1.3294	73	4 4 0
1.2702	4	5 3 1
1.1887	5	6 2 0
1.1477	13	5 3 3
1.1346	23	6 2 2
1.0865	4	4 4 4
1.0058	6	6 4 2
0.9797	32	7 3 1 / 5 5 3
0.9406	5	8 0 0
0.8867	9	6 6 0 / 8 2 2
0.8686	27	7 5 1 / 5 5 5
0.8628	22	6 6 2
0.8413	6	8 4 0
0.8259	6	9 1 1 / 7 5 3

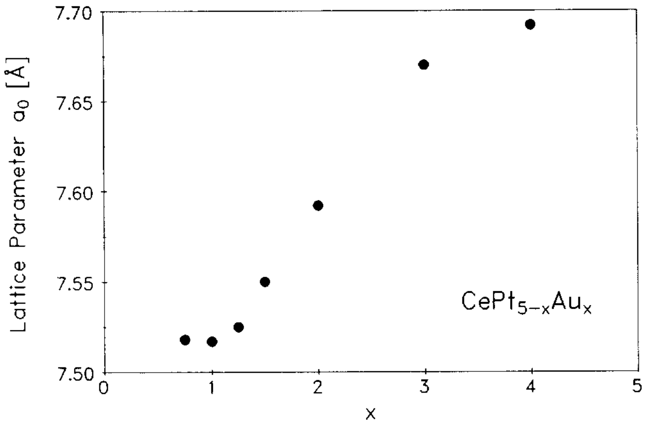

Fig. 1. Lattice parameter a_0 of the cubic structure in $CePt_{5-x}Au_x$. The saturation indicates that this structure is only stable for $1 < x \leq 3$. The accuracy is ±0.003 Å (±0.01 Å for $CePtAu_4$).

based series). This is corroborated by the absence of any second-phase peaks in this region. The fact that the ligand sites are occupied by two different elements apparently stabilizes the cubic lattice. In particular, the transition at a mole fraction of about 20% might be linked with the two different Be sites in the $AuBe_5$ structure. The cerium-based lattice is for all the different Au dopings significantly larger than the uranium-based lattice; therefore the transition from the hP6 to the cF24 structure seems to be not merely a lattice size effect. However, a gold content of exactly 20% is not sufficient to achieve a single-phase cubic structure. Despite the enhanced annealing time, we could not avoid secondary phase in $CePt_4Au$ in contrast with $CePt_{3.75}Au_{1.25}$. Furthermore, Fig. 1 shows the beginning of saturation for $x = 1$.

The measurements of susceptibility $\chi(T)$ and magnetization $M(H)$ were performed in a Quantum Design superconducting quantum interference device (SQUID) susceptometer. $\chi(T)$ was detected in a magnetic field $H = 0.5$ T for $1.65 < T < 400$ K and $M(H)$ at a temperature $T = 2$ K for $H \leq 7$ T. The accuracy is ±5%. The magnetization vs. field is strongly saturated for $CePt_{5-x}Au_x$, $x > 1$. This is an indication of unscreened Ce 4f moments in contrast with UPt_4Au where $M(H)$ is linear to at least 5.5 T. This saturation becomes more pronounced with increasing Au content in $CePt_{5-x}Au_x$ and with increasing Ce content in $U_{1-y}Ce_yPt_4Au$. The susceptibility $\chi(T)$ of the doped samples does not show Curie–Weiss behaviour up to a temperature of 400 K. The low temperature susceptibility $\chi(1.65\ \mathrm{K})$ is reduced (Table 2) with increasing Ce content in $U_{1-y}Ce_yPt_4Au$ but becomes enhanced with increasing Au content in $CePt_{5-x}Au_x$.

The specific heat measurements were performed by a relaxation method [10,11]. The absolute accuracy is ±4%. The low temperature specific heat corresponds qualitatively with the low temperature susceptibility and reveals decreasing values with increasing Ce content in $U_{1-y}Ce_yPt_4Au$ and increasing values with growing Au content in $CePt_{5-x}Au_x$ (Table 2). However, in the latter series the susceptibility increase is more pronounced than the increase in γ ($= C/T$ at $T = 1.25$ K); see Table 2. This means an enhancement of the ratio $R = \chi/\gamma$ and thus the system seems to be driven away from heavy fermion behaviour and towards more magnetic behaviour. The specific heat data of $CePt_3Au_2$ are shown in Fig. 2. The low temperature increase in the specific heat begins below 5 K. This sample revealed the highest C/T values in our series.

Now the question remains as to whether this steep increase belongs to an enhanced density of states as in the heavy fermion system UPt_4Au or whether it is an onset of a magnetic transition at lower temperatures. Since the ratio χ/γ is a factor of 10 higher than predicted for pure Pauli paramagnetic systems, and since the χ increase is stronger than the γ increase and the magnetization $M(H)$ at 2 K is distinctly saturated, we believe that magnetic correlations and magnetic entropy are the reason for the enhanced specific heat at low temperatures. The increase in the specific heat might be an onset of a low temperature magnetic transition or induced by fluctuation effects. Therefore measurements at still lower temperatures and specific heat measurements in magnetic fields would be desirable and should give further information about the origin of the high specific heat in the Ce-based system.

Table 2

Experimental data of $U_{1-y}Ce_yPt_4Au$ and $CePt_{5-x}Au_x$. The accuracy is 5% in the susceptibility χ, 4% in the specific heat C and 0.003 Å in the lattice parameter a_0. The data of UPt_4Au were taken from Refs. [4–6]. The saturation in the lattice parameter and the second phase thwart our attempt to give reliable data for $CePt_4Au$. For comparison, the ratio R for a pure Pauli susceptibility of quasi-free electrons amounts to 0.014 e.m.u. K^2 J^{-1}

Compound	a_0 (Å)	χ (10^{-3} e.m.u. mol^{-1}) at $T=1.65$ K	γ (mJ mol^{-1} K^{-2})	$R=\chi/\gamma$ (e.m.u. K^2 J^{-1})	Remarks
UPt_4Au	7.474	12.9	710	0.018	Data from Refs. [4–6]
$U_{0.8}Ce_{0.2}Pt_4Au$	7.484	9.2	430	0.021	
$U_{0.5}Ce_{0.5}Pt_4Au$	7.497	5.5	175	0.031	
$U_{0.2}Ce_{0.8}Pt_4Au$	7.510	4.23	75	0.056	
$CePt_4Au$	7.517	(25)			Second-phase impurities
$CePt_{3.75}Au_{1.25}$	7.525	1.36	17	0.08	
$CePt_{3.5}Au_{1.5}$	7.550	5.84	33	0.17	
$CePt_3Au_2$	7.592	65	360	0.18	
$CePt_2Au_3$	7.670	430	1375	0.31	

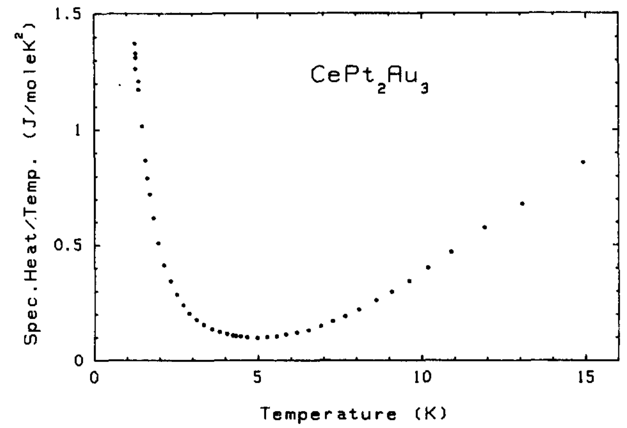

Fig. 2. Specific heat divided by temperature, C/T, vs. temperature for $CePt_2Au_3$, showing a steep increase below 5 K. The accuracy is ±4%.

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