Title
VARIATION OF GROUND-STATES IN CEPT2-XAUXSI2 COMPOUNDS (X = 0, 1, 2)

Permalink
https://escholarship.org/uc/item/41k0m2pr

Journal
SOLID STATE COMMUNICATIONS, 77(5)

ISSN
0038-1098

Authors
HEEB, E
OTT, HR
FELDER, E
et al.

Publication Date
1991-02-01

DOI
10.1016/0038-1098(91)90742-E

License
CC BY 4.0

Peer reviewed
VARIATION OF GROUND STATES IN CePt$_{2-x}$Au$_x$Si$_2$ COMPOUNDS (x = 0, 1, 2)

E. Heeb*, H.R. Ott, E. Felder, F. Hulliger, A. Schilling
Laboratorium für Festkörperphysik, ETH Hönggerberg, CH-8093 Zürich, Switzerland
and
Z. Fisk
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

(Received on 11.10.90 by P. Wachter)

A comparison is made of some low-temperature properties of CePt$_2$Si$_2$, CePtAuSi$_2$ and CeAu$_2$Si$_2$. New experimental data on the specific heat, electrical resistivity and magnetic susceptibility of CePtAuSi$_2$, together with published results on the other compounds, reveal that the electronic ground state in these materials varies in a very significant way. In-between local-moment behaviour in CeAu$_2$Si$_2$ and a Kondo-type ground state in CePt$_2$Si$_2$, CePtAuSi$_2$ forms a heavy-electron state at low temperatures.

Most CeM$_2$Si$_2$ compounds, with M as a d-transition metal, crystallize in the ThCr$_2$Si$_2$ structure. Some of them are known for their particular low-temperature properties. The most spectacular features are observed in CeCu$_2$Si$_2$, a heavy-electron superconductor, and in CeRu$_2$Si$_2$, which adopts a heavy-electron ground state without a phase transition down to 0.1 K. At low temperatures this state is unstable in high magnetic fields as evidenced by a metamagnetic-type transition. A replacement of Ce by La in CeRu$_2$Si$_2$ appears to induce a magnetically ordered ground state below 10 K.

Various unusual features are observed in compounds where M is a 5d transition element. In CeOs$_2$Si$_2$ the 4f electrons appear to be delocalized in spite of a fairly large Ce–Ce interatomic distance of 4.162 Å, whereas in CePt$_2$Si$_2$ a well-defined local moment of the 4f electron is indicated by a Curie–Weiss-type temperature dependence of the magnetic susceptibility $\chi(T)$ at temperatures above 200 K, but no magnetic order has been observed above 0.3 K. CeOs$_2$Si$_2$ crystallizes in the ThCr$_2$Si$_2$ structure, while CePt$_2$Si$_2$ instead adopts the CaBe$_2$Ge$_2$ structure. If Pt is replaced by Au, the resulting compound CeAu$_2$Si$_2$ is again a member of the ThCr$_2$Si$_2$ family and its local-moment behaviour is evidenced by a magnetically ordered state at low temperatures.

In this report we compare some low-temperature properties of CePt$_2$Si$_2$, CePtAuSi$_2$ and CeAu$_2$Si$_2$. To this end we measured the specific heat $C_p(T)$ of CePtAuSi$_2$ between 1.5 and 17.5 K. The data above 10 K can be fitted very well with a sum of the type $C_p = \gamma T + \beta T^3$, where $\gamma = 2.7$ mJ/mole K$^2$ and $\beta = 1.165$ mJ/mole K$^4$. The latter value is compatible with a Debye temperature $\Theta_D = 203$ K, and the low $\gamma$ value indicates that the conduction electrons have little or no 4f character. After subtraction of this background specific heat we are left with the magnetic part of $C_p(T)$, which is shown in Fig. 1. The resulting anomaly manifests the previously reported magnetic ordering and its shape is very close to what is expected for a simple mean-field-type transition. Nevertheless, some contribution from critical behaviour above $T_N = 7.3$ K appears to be present. The Neel temperature that we find here is in-between two previously published values of 6 K and 10 K.

Although the phase transition to a magnetically ordered state of CeAu$_2$Si$_2$ seems to vary in temperature...
between different materials, the low-temperature behaviour of this compound is close to classical expectations, as we shall outline below. According to ref. 9 the temperature dependence of $\chi$ below room temperature is well described by a Curie-Weiss law with a negative paramagnetic Curie temperature $\theta_p = -12$ K. The resulting effective moment is 2.57 $\mu_B$/Ce coinciding with the value that is expected for non-interacting Ce$^{3+}$ ions. Although no distinct features for crystal-electric-field (CEF) effects are discernible in the $\chi'(T)$ data of ref. 9, it is expected that the Hund's rule $J = 5/2$ ground state is split and for symmetry reasons a ground-state doublet is expected. This is nicely confirmed by the value of the molar entropy involved in the experimentally determined magnetic anomaly shown in Fig. 1, which amounts to $R \ln 2$ within a few percent between 0 K and $T_N$. From neutron-scattering work reported in ref. 10, a simple antiferromagnetic structure of type I was deduced and the ordered moment per Ce ion was found to be 1.3 $\mu_B$/Ce, a value that may well correspond with possible wave-functions of the lowest doublet state.

The low-temperature magnetic susceptibility of CePt$_2$Si$_2$ is distinctly different from that of CeAu$_2$Si$_2$. As described in refs. 6 and 7, only at elevated temperatures above 200 K is a Curie-Weiss-type behaviour observed. With decreasing temperature, $\chi(T)$ reaches a maximum around 60 K and after a subsequent decrease is approximately constant below 20 K. These unusual features for a Ce compound are absent in CeAuPtSi$_2$. We show the result of our measurement in the form of a $\chi'(T)$ plot in Fig. 2.

We note that the data between 30 and 300 K are fitted very accurately by a straight line, implying a $(T - \theta_p)^{-1}$ dependence of $\chi$. At lower temperatures $\chi(T)$ increases more rapidly with decreasing temperature. The inset of Fig. 2 emphasizes the behaviour below 20 K. The most probable cause for the particular shape of the $\chi'(T)$ curve are crystal-field effects. A possible underlying Curie-Weiss behaviour, as indicated by the broken line in Fig. 2, is altered by the thermally induced change of occupation of the CEF-split $4f$-electron levels, again 3 doublet states in this case. The resulting effective moment is then about 2.4 $\mu_B$/Ce and the paramagnetic Curie temperature negative but close to zero. Below 10 K another region of Curie-Weiss behaviour with again a small value for $\theta_p$ may be identified. With this interpretation the effective moment of 1.6 $\mu_B$/Ce would have to be ascribed to the ground-state doublet. At the lowest temperatures a tendency towards saturation of $\chi(T)$ may be inferred from the experimental data.

In Fig. 3 we show the field dependence of the magnetization $M(H)$ for CePtAuSi$_2$ at a low temperature (2 K) and in fields up to 100 kOe. Here, a distinct difference to analogous results for CeAu$_2$Si$_2$, which were reported in ref. 9, have to be noted. Because of the antiferromagnetic order in the latter compound, the field-induced transition to the paramagnetic state between 60 and 80 kOe leads to an s-shaped curve in that case. Although our maximum field value is 100 kOe, it is difficult to estimate a saturation value for $M$. Also for CeAu$_2$Si$_2$ this limit was by far not reached even in fields of 210 kOe 9, but our data on CePtAuSi$_2$ seem to extrapolate to roughly the same $M(H)$ values for $H > 100$ kOe.

The most intriguing differences in the low-temperature behaviour of the three compounds are apparent in the temperature dependences of the specific heat. In Fig. 4 we show, on a double-logarithmic diagram, our results for $C_p(T)$ of CePtAuSi$_2$ between 0.1 and 30 K, in comparison with data for CePt$_2$Si$_2$ reported in ref. 7. Fig. 5 shows the same data on a $C_p/T$ vs. $T$ plot for temperatures below 10 K. For temperatures between 4 and 10 K, Ayache and collaborators 7 fitted their data for CePt$_2$Si$_2$ with a sum of an electronic and a Debye-type lattice contribution, whereby the electronic specific-heat parameter $\gamma = 86$ mJ/mole K$^2$ and $\theta_D = 215$ K. An analogous analysis of the data of
CePtAuSi$_2$ above 10 K leads to the values $\gamma = 118$ mJ/mole K$^2$ and $\theta = 212$ K. Comparing, in particular, the values for the Debye temperatures for all three compounds we note that their lattice excitation spectra must be nearly identical. Therefore, the visible strong differences in $C_p(T)$ at low temperatures have to be due to variations in the electronic spectrum.

As we pointed out above, CeAu$_2$Si$_2$ behaves like a classical metallic rare-earth antiferromagnet where the exchange interaction mediated by the conduction electrons is the dominating factor for the low-temperature behaviour. The electronic structure must be quite different for CePt$_2$Si$_2$, as indicated by the unusual temperature dependence of $\chi$. Its low-temperature electronic specific heat has indeed been interpreted as the lower end of a single-ion Kondo anomaly with a Kondo temperature of approximately 70 K$^7$. The situation is again drastically different for CePtAuSi$_2$ where, at temperatures above 10 K, the electronic specific heat is larger but comparable in magnitude to that of CePt$_2$Si$_2$. Below 10 K, however, a strong enhancement of the $C_p/T$ ratio in the Au-doped compound signals the formation of a heavy-electron state. With decreasing temperature, this enhancement appears to saturate but below 1 K a further enhancement and a maximum at approximately 0.3 K are observed. Extrapolating to $T = 0$, the $C_p/T$ ratio seems to approach a value of roughly 1 J/mole K$^2$. After subtracting the lattice contribution to $C_p(T)$, the entropy of the heavy electrons can be evaluated and it reaches the value of $R \ln 2$ at approximately 20 K.

As a last item, we compare the electrical resistivity $\rho(T)$ of these three compounds at temperatures below 300 K. In Fig. 6, the temperature dependence of $\rho$ for CePt$_2$Si$_2$ is shown between 1.3 K and 300 K. Inset is a logarithmic scale for CePt$_2$Si$_2$. For CePtAuSi$_2$, analogical data were reported by Murgai and co-workers$^9$. In this latter case, $\rho$ decreases with decreasing temperature below 300 K. A few Kelvin above $T_N$, $\rho$ changes sign and the magnetic ordering manifests itself by a distinct chromium-type anomaly and a maximum in $\rho(T)$ just below the ordering temperature. With further decreasing $T$, $\rho(T)$ again decreases in the usual fashion of vanishing spin-disorder scattering. For CePt$_2$Si$_2$, $\rho$ first increases with increasing negative slope on lowering $T$, passes through a maximum at about 70 K, and subsequently decreases considerably towards lower temperatures$^6$. The maxima, both in $\chi(T)$ and $\rho(T)$ for this compound at almost the same temperature, were interpreted as a sign for the increasing dominance of a Kondo-type interaction and concomittant coherence effects among conduction electrons affecting the electronic transport$^7$.

For CePtAuSi$_2$, $\rho(T)$ passes through a shallow minimum around 200 K and, with decreasing temperature, increases with increasing negative slope. At temperatures below 4 K, we note a distinct maximum and a subsequent drop in $\rho$ below $T_{\text{max}} \sim 2.5$ K. These features are very similar to those observed in other heavy-electron compounds, although for most other prominent Ce compounds in this class of substances, the onset of coherence or, in other words, a considerable decrease of $\rho$ with decreasing temperature, is usually observed at distinctly higher temperatures such as about 10 K for CeCu$_2$ or 35 K for CeAl$_3$.$^{11}$

The change of crystal structure between the two compounds CePt$_2$Si$_2$ and CeAu$_2$Si$_2$ is by itself an interesting feature. Clearly also the importance of different types of electronic interactions is varying distinctly from one substance to the other. Replacing Pt with Au leads to a decrease of the Kondo-type interaction, leaving it strong enough, however, to prevent the onset of cooperative magnetic order in the usual sense in CePt$_2$Si$_2$. The onset of a heavy-electron state in such compounds is obviously not restricted to materials crystalizing with the ThCr$_2$Si$_2$ structure, as we outlined above. Nevertheless, microscopic studies may reveal that, as in other substances$^{12}$, some unusual magnetic coherence occurs in this state possibly also in the case of CePt$_2$Si$_2$. It is especially the temperature dependence of the $C_p/T$ ratio below 1 K which leads us to this conjecture. Unfortunately, these data are not compatible with a duplicate of the well known ground state of CeCu$_2$Si$_2$ and the present experimental situation also gives no real clue for causes that determine the instabilities of heavy-electron states. In spite of this, further experiments on other materials with varying Pt to Au ratio, seeking the boundary of the crystal-structure change and investigating its influence on the magnetic behaviour might still be rewarding.

REFERENCES