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SPECIFIC HEAT, MAGNETIC SUSCEPTIBILITY AND TRANSPORT PROPERTIES OF POLYCRYSTALLINE CeInAu AND CeInAu_2

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Measurements of specific heat (1.4–350 K), thermopower and thermal conductivity (1.5–300 K), electrical resistivity (40 mK–300 K) and of the magnetic susceptibility (40 mK–300 K) on polycrystalline samples of CeInAu , CeInAu_2 and of the reference compounds LaInAu and LuInAu_2 are reported. The properties of the Ce-compounds are unusual due to phase transitions (magnetic ordering, martensitic) and crystalline electric field effects. There are only weak indications of a valence instability.

1. Introduction

The ternary intermetallics YbInAu and YbInAu_2 are known as intermediate valence (IV)-compounds [1–3]. From measurements of the L_{III} -absorption edges it is found that CeInAu and CeInAu_2 are also weakly mixed valent with valence $\nu = 3.03$ and 3.05 , respectively [4]. In contrast, in a recent measurement of the L_{III} -edges CeInAu_2 has been reported to be trivalent [5]. From the lattice parameters one knows that in contrast to the Yb-compounds no volume anomaly can be detected in the two Ce-compounds. The intention of our work was to find out, whether there are typical indications of a 4f-instability of Ce in these compounds, or whether they belong to the rare class of stable trivalent intermetallic Ce-compounds.

Polycrystalline samples of REInAu and REInAu_2 (RE = La, Ce, Lu) were prepared by arc-melting stoichiometric amounts of the pure elements. X-ray patterns were obtained in agreement with other authors. CeInAu and LaInAu are hexagonal (hP9- Fe_2P), CeInAu_2 and LuInAu_2 are cubic (B2-CsCl) at room temperature [2,3,5]. CeInAu_2 undergoes a martensitic transformation at about 200 K [5]. Investigations through microsections showed that in CeInAu and CeInAu_2 minor amounts (less than 5%) of a second phase were present. Also

microcracks were visible in the REInAu samples. Annealing at 550°C diminished the second phase.

2. REInAu

The temperature dependence of the resistivity of CeInAu is quite unusual (fig. 1). Unfortunately the absolute resistivity could not be determined for the REInAu -samples, since the presence of the microcracks did not allow to determine the geometric factor. It is remarkable that annealing raised the resistivity by a factor of about 3. The residual resistivity ratio (rrr) indicates unusually strong lattice defects. This seems to be a common property of intermetallic compounds in the Fe_2P structure (similar effects are reported on YbCuAl [6]).

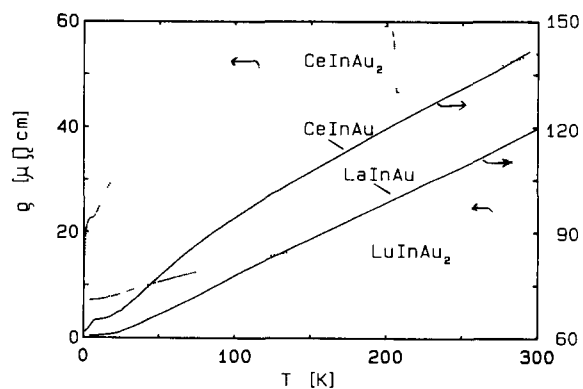


Fig. 1 Resistivity vs temperature of REInAu_2 (left scale) and REInAu (right scale). The absolute scale for REInAu was obtained by scaling via the Wiedemann–Franz law (see text).

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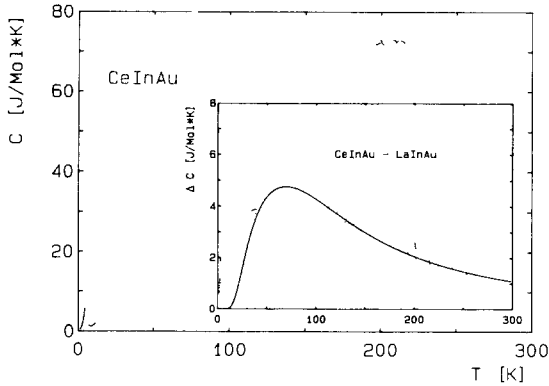


Fig 2 Specific heat C_v vs temperature of CeInAu. Inset $C_v(\text{CeInAu}) - C_v(\text{LaInAu})$ vs temperature and Schottky-fit (solid line)

The specific heat of CeInAu (fig 2) shows an anomaly at 5.7 K with an entropy of about $R \ln 2$. A plot of C/T vs T^2 is linear between 12 and 18 K and we find an electronic specific heat coefficient γ of about $30 \text{ mJ}/\text{mol K}^2$. Subtracting the data of LaInAu (inset fig 2) reveals a second anomaly with a maximum at 60 K. This anomaly can be interpreted as a Schottky-effect with a crystal field (CF)-scheme with two doublets at 120 and 290 K (solid line in fig 2)

The magnetic susceptibility (fig 3) also shows an anomaly at 5.7 K. Together with the entropy of $R \ln 2$, one can conclude that it is due to antiferromagnetic order. From 6 to 11 K and from 20 to 300 K the susceptibility shows Curie-

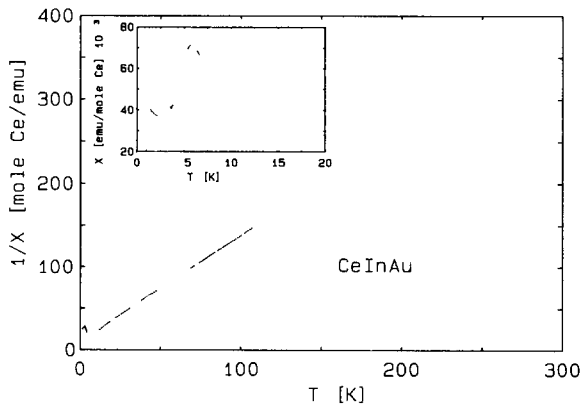


Fig 3 Inverse magnetic susceptibility vs temperature of CeInAu. Inset: Magnetic susceptibility at low temperatures

Weiss behaviour with $\theta = -1.5$ and -10 K , respectively. At low temperatures the moment is strongly reduced, with a value of $2.10 \mu_B$. At room temperature we extract a magnetic moment of $2.50 \mu_B$, which is slightly reduced with respect to that of the $4f^1$ configuration of Ce. This may be due to a weak valence instability. From $\chi(T) = (4 - v)TC/(T + T_f)$ (χ magnetic susceptibility, C Curie-constant) we get a valence of $v \leq 3.03$ and a fluctuation temperature $T_f < 20 \text{ K}$ [7]

The thermal conductivity of CeInAu and LaInAu is quite small (50 and $60 \text{ mW}/\text{cm K}$ at 300 K, respectively) and without structure. This is another indication for strong lattice defects. Assuming that the Wiedemann-Franz law $k\rho/T = L_0$ holds at room-temperature, which is reasonable for a compound with strong elastic defect scattering, we defined an absolute scale of the resistivity (fig 1, right scale)

The thermopower of LaInAu (fig 4) shows a linear temperature dependence at high temperatures. Below 50 K there is some finestructure, which, however, coincides with strong curvature of the thermopower of the contacting wires. We therefore cannot exclude that this structure is artificial. The thermopower of CeInAu changes sign at 5.7 K. The finestructure of the minimum of S below about 80 K may have the same origin as for LaInAu. Neglecting it the minimum itself may be due to phonon-drag; a phonon drag effect would be expected at $\theta_D/5$, i.e. at 40 K.

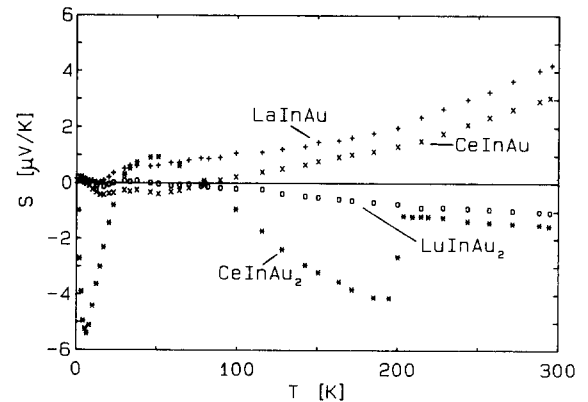


Fig 4 Thermopower vs temperature of CeInAu, LaInAu, CeInAu₂ and LuInAu₂

3. REInAu₂

The resistivity of CeInAu₂ (fig 1) shows a very structured behaviour over the whole investigated temperature range. The room-temperature value of about 54 $\mu\Omega\text{cm}$ is quite large. The martensitic transformation shows up as a step-like anomaly at 203 K.

The specific heat C of CeInAu₂ (fig 5) exhibits a very steep peak at 203 K, which is also due to the martensitic phase transition. The increase of C below 4 K suggests another anomaly. A plot of C/T vs T^2 is linear between 6 and 11 K and we extract $\gamma = 130 \text{ mJ/mol K}^2$. We note that this value is unusually large. Subtracting the data of LuInAu₂ (inset fig 5) one finds a Schottky-anomaly with a maximum at 40 K. This Schottky-anomaly yields a CF-scheme with two doublets at 85 and 600 K above the CF-groundstate. The magnetic susceptibility (fig 6) shows an anomaly at 1.2 K. The coincidence with the inflection point of the electrical resistivity points to an antiferromagnetic transition. From ref [8] we know that the susceptibility shows Curie-Weiss behaviour between 2 and 19 K and between 40 and 300 K with $\theta = -4$ and -14 K, respectively. We extract a slightly enhanced moment of $2.56 \mu_B$ at room-temperature. At low temperatures we find $2.06 \mu_B$.

The thermal conductivity of CeInAu₂ shows a

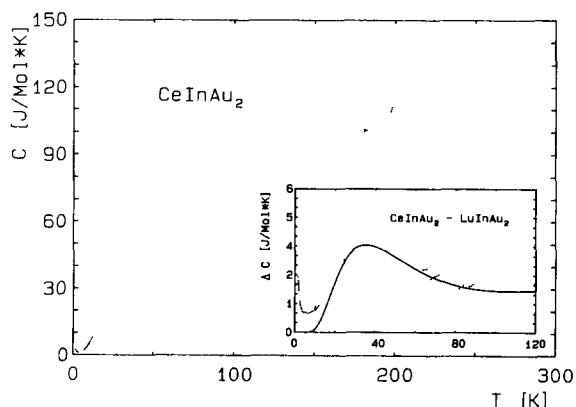


Fig 5 Specific heat C_v vs temperature of CeInAu₂. Inset $C_v(\text{CeInAu}_2) - C_v(\text{LuInAu}_2)$ vs temperature and Schottky fit (solid line)

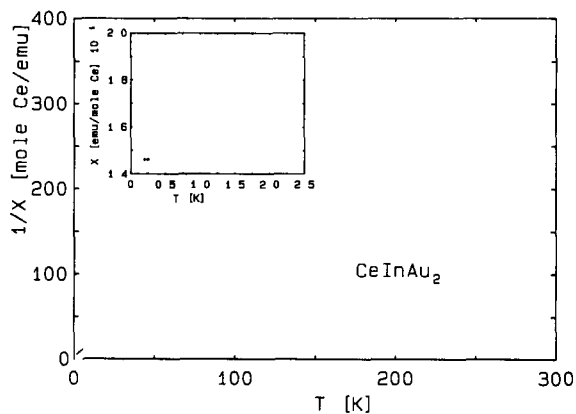


Fig 6 Inverse magnetic susceptibility vs temperature of CeInAu₂. Inset: Magnetic susceptibility at low temperatures

steep decrease at about 195 K. It has a room temperature value of about 120 mW/cm K. The thermopower of CeInAu₂ (fig 4) is very structured. The martensitic transition shows up at 200 K. The order of magnitude, however, is comparable to that of normal metals. The minimum at 8 K might be explained with CF-effects. However, theory [9] predicts the minimum to occur at a third of the temperature of the first excited CF-state, which would be at 28 K. The maximum at 50 K might be due to phonon-drag.

4. Conclusion

Except for the enhanced values of γ from specific heat and thermopower there are no indications of a valence instability of the Ce-compounds.

On the other hand the resistivity anomaly interpreted in the dynamic alloy model of Wohleben and Wittershagen [10] delivers a valence of 3.02 ± 0.01 for CeInAu, if one uses the isostructural LaInAu as reference compound. No proper reference compound exists to determine the resistivity anomaly of CeInAu₂. LuInAu₂ is cubic over the whole temperature range, LaInAu₂ shows a quite anomalous resistivity itself. However, with this proviso we extract a resistivity anomaly of about 20 $\mu\Omega\text{cm}$ at room temperature, which would also lead to a valence of about 3.02.

The valence extracted from the magnetic sus-

ceptibility of CeInAu ($\nu \approx 3.03$) is close to the one extracted from the L_{III} -measurements and from the resistivity. The magnetic ordering temperature of 5.7 K is quite high for an IV-compound.

The high temperature susceptibility of CeInAu₂ does not allow to determine the valence, since this measurement was done with an accuracy of about 3%. The magnetic ordering temperature of CeInAu₂ at 1.2 K does not obey de Gennes scaling. From the ordering temperature of the other REInAu₂ [5] one would expect an ordering at $T_N = 0.1$ K.

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