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MAGNETIC SUSCEPTIBILITY, ELECTRONIC SPECIFIC HEAT AND TRANSPORT PROPERTIES
OF SOME INTERMETALLIC COMPOUNDS OF CERIUM

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Résumé. — Des mesures de résistance électrique, de chaleur spécifique électronique et de pouvoir thermoélectrique ont été faites sur les composés CeSn₃ et CeBe₁₃ pour lesquels des mesures antérieures montraient un aplatissement de la courbe 1/x(T) vers 200 °K. La chaleur spécifique est grande (≈ 50 et 100 mJ/mole/°K²) et peut s'expliquer par un état lié non magnétique 4f virtuel avec une largeur de bande habituelle (≈ 0,01 eV) et un taux d'occupation 4f égal à 1. On trouve de larges rais dans les courbes de pouvoir thermoélectrique. Les mesures de chaleur spécifique (CeAl₂ et CePb₃) et de pouvoir thermoélectrique (CePb₃) sont également indiqués par ces deux composés dont la susceptibilité suit une loi de Curie-Weiss.

Abstract. — Measurements of thermopower, electrical resistance and electronic specific heat have been made on the compounds CeSn₃ and CeBe₁₃ for which previous measurements showed a flattening off in 1/x versus T around 200 °K. The γ values are large (≈ 50 and 100 mJ/mole/°K²) and could arise from a non-magnetic 4f virtual bound state with the usual energy width (≈ 0,01 eV) and a 4f occupation number of unity. There are broad positive peaks in the thermopower curves various features of which scale as the γ values (or the energy widths). Specific heat (CeAl₂ and CePb₃) and thermopower (CePb₃) results for two compounds for which γ is essentially Curie-Weiss are also reported.

In the study of dilute magnetic alloys cerium forms an interesting link between the other rare earth impurities and the 3 d transition elements. Because the 4 f wave functions are well-localized, intermetallic compounds of Ce and the pure metal itself have several features in common with dilute alloys. For example Kondo type resistance minima have been observed in the compounds CeAl₂ [1] and CeB₆ [2] and, on the theoretical side, the Friedel-Anderson model, originally developed for isolated 3 d impurities, has been used as the starting point for a successful treatment of the anomalous phase diagram and magnetic properties of cerium metal [3].

Previous measurements [4] of magnetic susceptibility down to 77 °K on the compounds CeSn₃ and CeBe₁₃ showed a levelling off in the 1/x vs. T curves around 200 °K. This was associated with a magnetic-nonmagnetic transition of the 4 f virtual bound state of cerium. The lattice parameter of CeSn₃ is also strongly temperature dependent [5]. To investigate this transition further we have measured the thermopower, susceptibility, electrical resistance and low temperature specific heat for these two compounds, for CePb₃ (which is iso-structural with CeSn₃ but has a different (T) dependence) and for CeAl₂ (for which susceptibility [6], resistance [1], and specific heat [11], data have already been published).

The susceptibility results are shown in figure 1: the three curves are approximately parallel at high temperatures, the slope corresponding approximately to the magnetic moment of the free Ce⁴⁺ ion. The points with the large error bars below 77 °K were taken using a zero field AC method and so the low temperature rise in 1/x may not be significant.

In contrast to CeSn₃ for CePb₃, 1/x continues to fall approximately linearly with T down to 10 °K, in agreement with previous work [7].

![Graph](http://dx.doi.org/10.1051/jphyscol:19711406)
The absolute thermopowers $S$ of the three specimens are shown in figure 3. We note that:

1. In all cases there is a large positive peak ($40 \, \mu V/\mathrm{K}$) in $S$.

2. For CeSn$_3$ and CeBe$_{13}$ (and approximately for CePb$_3$) $S$ is proportional to $T$ at low temperatures, as expected for the «electron diffusion thermopower» when phonon effects can be neglected, but the initial slopes of the two curves differ by a factor of $8$.

3. At high temperatures the two curves merge and show the same linear dependence above $150 \, \mathrm{K}$ for CeBe$_{13}$ and $300 \, \mathrm{K}$ for CeSn$_3$.

The resistance results for CeBe$_{13}$ and CeSn$_3$ show no sharp anomalies but at room temperature (certainly well above $\theta_D$ for CeSn$_3$) the resistance rise is slower than the $T^2$ dependence expected. This effect, when observed in transition metals, has been attributed to narrow electron energy bands whose width is comparable with $kT$ [10].

We have tried to analyse the specific heat and thermopower results for CeSn$_3$ and CeBe$_{13}$ using the 4f density of states $\rho(E)$ from the Friedel-Anderson model referred to previously [3]. Application of the standard formula for the thermopower [10] for scattering into a narrow f-band gives the observed sign and low temperature dependence. Unfortunately the analysis is not self-consistent since the numerical values of the half-width $\Delta$ and the position of the resonance, $E_0 - E_F$, give a 4f occupation number, $n_f$, greater than unity.

However we can obtain values of $\Delta$ and $E_0 - E_F$ from the values of $\gamma$ alone, assuming $n_f = 1$. The resulting values of $\Delta$ (table I) are of the expected magnitude for Ce [3]. In figure 3 we see also that the height of the thermopower maximum scales as $1/\Delta$, the limiting dependence of $S$ is reached at $T = 2.5 \, \Delta$ and in the low temperature region the first deviations from $S/T = constant$ occur at $T = \Delta/2$ in both cases.

The susceptibility enhancement factor determined from the (approximately) temperature independent susceptibility and $\gamma$ is surprisingly small in both compounds (see table I). Possible phonon enhancement of the observed specific heat has been ignored. In contrast, the susceptibility enhancement factor in non-magnetic $\alpha$-Ce is believed to be about 20 [3].

In conclusion the high electronic density of states observed in two compounds of Ce is consistent with reasonable values of $\Delta$ together with a 4f occupation number of unity. Despite this there appears to be no localized moment at low temperatures and the susceptibility enhancement factor is low.

Various features of the observed thermopower curves scale as the ratio of these widths. In contrast to the behaviour of Ce metal under pressure no sharp

![Figure 2: Specific heat data, $C/T$ (mJ/mole/°K)$^2$ versus $T^2$ (°K$^2$). CeSn$_3$ - , CeBe$_{13}$ - , CePb$_3$ - , CeAl$_2$ - .](image)

![Figure 3: Absolute thermopower (μV/°K). Versus T(°K). CeSn$_3$ - , CeBe$_{13}$ - , CePb$_3$ - .](image)

<table>
<thead>
<tr>
<th>Compound</th>
<th>$\gamma$ (mJ/mole/°K$^2$)</th>
<th>$\rho(E_F)$ (Elec/eV/atm.)</th>
<th>$\Delta (\text{eV}) \times 10^{-2}$</th>
<th>$n_f(E_F)/n_f(E_F)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CeSn$_3$</td>
<td>53 ± 10</td>
<td>22.5 ± 4</td>
<td>1.0 ± .2</td>
<td>3.2 to 2.2</td>
</tr>
<tr>
<td>CeBe$_{13}$</td>
<td>115 ± 10</td>
<td>49 ± 4</td>
<td>.45 ± .04</td>
<td>1.67 to 1.43</td>
</tr>
<tr>
<td>CePb$_3$</td>
<td>225 ± 25</td>
<td>95 ± 11</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CeAl$_2$</td>
<td>178 ± 8</td>
<td>74 ± 3</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
anomalies are seen in either the electrical resistance or the specific heat. This reinforces the view that no discontinuous change in the 4f occupation number occurs for our compounds. The two other compounds (CePb, CeAl2) have even larger γ values and, between them, these two show all the features commonly associated with the Kondo effect.

References