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Magnetization and specific heat of abnormal cerium compounds


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Résumé. — Les composés anormaux de cérium CeIn₃, CeAl₂, CeAl₃ ont été étudiés sous pression par mesures d’aimantation et de chaleur spécifique. La proximité de la transition basse température magnétique-non magnétique de CeIn₃ conduit à une forte augmentation de la chaleur spécifique électronique sous pression tandis que pour CeAl₂ une décroissance est observée. Des mesures d’aimantation faites sur Ce₃Al₁₁ montrent clairement que les propriétés étranges attribuées à CeAl₃ peuvent provenir d’une faible teneur en Ce₃Al₁₁.

Abstract. — The abnormal cerium compounds CeIn₃, CeAl₂, CeAl₃ are studied by magnetization and specific heat measurements under pressure. The vicinity of the low temperature magnetic-non magnetic transition for CeIn₃ leads to a strong increase under pressure of the electronic specific heat whereas for CeAl₂ a decrease is observed. Magnetization experiments performed on Ce₃Al₁₁ clearly show that striking properties attributed to CeAl₃ may be due to a small content of Ce₃Al₁₁ in CeAl₃ samples.

The abnormal properties of cerium compounds like CeAl₂, CeAl₃ and CeIn₃ are far to be wellknown and understood. If now the persistence of a magnetic ordering is well established for CeAl₃ down to 0 K [1, 2], the low temperature state of CeAl₃ is not clear, mainly the origin of two temperatures $T_M$ of weak specific heat maxima at 2.5 and 6 K generally reported [3]. The properties of CeIn₃ has been little studied [4]; its magnetic ordering down to 5 mK is well established [2].

Magnetization experiments have been performed on these compounds and on Ce₃Al₁₁ in order to know if its presence as a parasitic phase in CeAl₃ should not explain the two temperature $T_M$ different samples of CeAl₃ have been studied. The magnetization experiments have been performed up to 150 kOe down to 1.4 K. Magnetization and specific heat measurements have been made under a pressure up to 12 kbar using a CuBe clamp. The pressure is calibrated in situ by the superconductive transition of tin. The sample specific heat is deduced by the difference between the specific heat measured with and without the sample. To give an order of magnitude, at low temperature, the specific heat of the clamp is the analog of 300 g of copper, the mean weight of the samples is 2 g. Finally temperature scales of the thermometers have been established between each run [5].

1. Ce₃Al₁₁ and CeAl₃ magnetization. — Magnetization experiments on Ce₃Al₁₁ have clearly shown:

i) at $p = 0$ that two ordering temperatures occur near 2.5 K and 6 K in agreement with older results [6, 7],

ii) at $p = 6$ kbar that only the ordering at 6 K seems to persist. The figure 1 represents the variation of the remanent $\sigma_R$ contribution measured after a cycle up to 150 kOe. The main points are at $p = 0$ the strong decrease of $\sigma_R$ below 2.5 K and at $p = 6$ kbar the persistence of $\sigma_R$ down to 1.4 K.

![Fig. 1. — Temperature variation of the remanent magnetization of Ce₃Al₁₁ at 0 and 6 kbar.](http://dx.doi.org/10.1051/jphyscol:19795111)
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of CeAl₂ and the other ten percent of Ce₃Al₁₁, the first leads to the disappearance of the specific heat anomalies at 2.5 K and 6 K and of the remanent \(\sigma_r\) whereas for the second both effects persist. In the first case, a single temperature \(T_M \sim 3.8\) K appears in good agreement with the anomaly reported for a bulk sample of CeAl₂ (below 2.5 K, both compounds give the same result). The state of CeAl₃ between 2.5 K and 6 K is far to be understood.

2. Specific heat measurements under pressure. —
An important contribution to the understanding of the abnormal behaviour of these compounds should be given by pressure measurements. We have reported in table I the zero pressure results, the low temperature data is analyzed using a convenient law:

\[ C = \gamma^0 T + \beta T^3. \]

(In ordinary antiferromagnets, the \(T^3\) law describes the spin wave excitations.) The first and second columns represent the \(\gamma^0\) and \(\beta\) values, the third the temperature of the specific heat maximum \(T_M\) if observed, the fourth the susceptibility \(\chi\) extrapolated at 0 K, the fifth an attempt to give an evaluation \(\gamma^{HT}\) of \(\gamma\) just above \(T_M\) for CeAl₂ and CeIn₃ and above 6 K for CeAl₃. According to different \(T_M\) values, the \(T^3\) term is considerably lower in CeIn₃ than in CeAl₂ (\(\beta \sim T^2\)). The 6 kbar pressure dependence is indicated in table II: the shift

\[ \Delta T_M = T(M(P) - T_M(O)) \]

is mentioned in the third column.

Table I. — Zero pressure results.

<table>
<thead>
<tr>
<th>Compounds</th>
<th>(\gamma^0) (mJ/mole/K²)</th>
<th>(\beta) (mK/mole/K⁴)</th>
<th>(T_M) (K)</th>
<th>(\chi) (10⁴ mK/mole)</th>
<th>(\gamma^{HT}) (mK/mole K²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CeAl₂</td>
<td>145</td>
<td>150</td>
<td>3.8</td>
<td>400</td>
<td>130</td>
</tr>
<tr>
<td>CeIn₃</td>
<td>136</td>
<td>15</td>
<td>10</td>
<td>129</td>
<td>144</td>
</tr>
<tr>
<td>CeAl₃</td>
<td>1640</td>
<td></td>
<td>?</td>
<td>?</td>
<td>~330</td>
</tr>
</tbody>
</table>

The susceptibility result of CeAl₂ is taken from [9] and the very low temperature \(\gamma^0\) of CeAl₃ from [8].

Table II. — 6 kbar results.

<table>
<thead>
<tr>
<th>Compounds</th>
<th>(\gamma^0)</th>
<th>(\beta)</th>
<th>(\Delta T_M)</th>
<th>(\chi)</th>
<th>(\gamma^{HT})</th>
</tr>
</thead>
<tbody>
<tr>
<td>CeAl₂</td>
<td>100</td>
<td>0.7</td>
<td>320</td>
<td>180</td>
<td></td>
</tr>
<tr>
<td>CeIn₃</td>
<td>320</td>
<td>0.5</td>
<td>129</td>
<td>200</td>
<td></td>
</tr>
<tr>
<td>CeAl₃</td>
<td>620</td>
<td>0</td>
<td>?</td>
<td>180</td>
<td>~350</td>
</tr>
</tbody>
</table>

and Wittig in this conference). From the CeIn₃ results, the experimental consequence of the vicinity of the transition from an ordered magnetic ground state to a non magnetic ground state is the strong increase of \(\gamma\) and the almost zero value of \(\beta\). The spin fluctuation of the magnetic moment with the electronic Fermi sea increases the electronic specific heat under pressure.

The \(\gamma^0\) decrease of CeAl₂ can be understood in a naive model where there is an interplay between the Kondo behaviour of one ion and the exchange coupling of the other ions which produce a fictitious molecular field \(H_m\) on the ion [10]. By increasing the pressure, the bare Kondo \(T_K\) temperature increases (a golden rule for a cerium dilute alloy), the relative molecular field \(g\mu_B H_m/k_B T_K\) parameter has an almost constant pressure dependence as reported the increase of \(T_M\); the final Kondo electronic specific heat given by:

\[ \gamma^0 \sim \frac{k T_K}{(k T_K)^2 + (g \mu_B H_m)^2} \]

(see reference [9]) decreases.

As it has been underlined, more difficult to elucidate are the CeAl₂ properties. Experimentally at \(p = 0\) the main result seems to be an increase of \(C/T\) when \(T\) decreases. Metallurgical precautions must be taken before claiming evidence of Fermi liquid spin fluctuation. At \(p = 6\) kbar \(C/T\) is nearly independent of the temperature. Recently the CeIn₃ experiments have been performed up to 12 kbar: a strong decrease of \(T_M\), \(\Delta T_M \sim -1.6\) K has been observed.

References