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Anisotropic f-electron magnetism in UNi$_4$B

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The hexagonal uranium compound UNi$_4$B has been shown to exhibit strongly anisotropic hybridization of f- and d-electron states. We performed low-field (3 mT) and high-field (40 T) magnetization measurements from 1.4 K to 30 K on a single crystal of this antiferromagnetically ordering compound, $T_N$=20 K, to further investigate the magnetic anisotropy. A phase diagram, including a spin-flop transition around 9 T, is presented. The observed large values of the electronic specific heat at low temperatures are attributed to in-plane magnetic fluctuations, which persist far below $T_N$. No superconductivity was found down to 40 mK. The relevance of strong 5f–3d hybridization is confirmed by extensive experiments on a series of diluted compounds UCoxNi$_{4-x}$B (0≤x≤4).

In previous papers [1,2] we demonstrated that the hexagonal intermetallic uranium compound UNi$_4$B, a member of the growing class of "1-5"–intermetallic compounds, exhibits highly anisotropic magnetic properties. AF ordering of U-spins lying in the basal plane occurs below $T_N$=20 K. However, both susceptibility and specific heat increase below $T_N$, which can be explained by the presence of strong in-plane magnetic fluctuations, persisting down to below 1 K [2]. In this contribution we show the importance of hybridization of the f and d-electron bands by alloying UNi$_4$B with Co on the Ni-sites. As both UC$_4$B and UNi$_4$B crystallize in the same CeCo$_4$B-type structure, alloying is possible over the entire concentration range (0≤x≤4) in UC$_{x}$Ni$_{4-x}$B. In Table 1 we list the lattice parameters of this pseudo-ternary system, together with the observed AF ordering temperature. From these data, the different interatomic uranium distances can be derived: In the basal plane, $d_{U-U}$=a, while along the c-axis $d_{U-U}$ = $\frac{1}{2}$c, yielding 4.952 Å and 3.477 Å for UNi$_4$B. In general, the magnetic moment will orient perpendicular to the direction of strongest f–f hybridization, i.e. in the basal plane.

In Fig.1 we present the magnetic susceptibility ($\chi$=M/H) for polycrystalline material with $x$=1 and 2, together with low-field (3 mT) and high-field (0.5 T) data for single-crystal UNi$_4$B. While UNi$_4$B exhibits clear Curie-Weiss local-

<table>
<thead>
<tr>
<th>$a$ (Å)</th>
<th>$c$ (Å)</th>
<th>V($\AA^3$)</th>
<th>$T_N$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>UNi$_4$B</td>
<td>4.952</td>
<td>6.954</td>
<td>147.68</td>
</tr>
<tr>
<td>UC$_1$Ni$_3$B</td>
<td>4.931</td>
<td>6.964</td>
<td>146.64</td>
</tr>
<tr>
<td>UC$_2$Ni$_2$B</td>
<td>4.924</td>
<td>6.954</td>
<td>146.01</td>
</tr>
<tr>
<td>UC$_3$Ni$_1$B</td>
<td>4.910</td>
<td>6.928</td>
<td>144.62</td>
</tr>
<tr>
<td>UC$_4$B</td>
<td>4.895</td>
<td>6.933</td>
<td>143.88</td>
</tr>
</tbody>
</table>

Figure 1: Susceptibility of UCoNi$_3$B (+) and UCo$_2$Ni$_2$B (○) and single-crystal UNi$_4$B (Δ) with field parallel to the $ab$-plane and along the $c$-axis (□), all measured in $\mu_0 H$=0.5 T. Note the large anisotropy for UNi$_4$B and the saturation of $\chi$ $||$ $ab$ in 3 mT (○) below 10 K.

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moment type behavior with \( p_{eff} = 2.81 \mu_B \) above 100 K (not shown), its uranium 5f-derived moment is progressively lost upon increasing Co-concentration. This strongly suggests that the 5f-electrons are filling the 3d-band when Co is introduced in the system. As a result, the AF ordering temperature is strongly reduced, from \( T_N = 20.0 \) K for UNi4B to \( T_N = 5.0 \) K for UCoNi3B. The low-field magnetization of single-crystal UNi4B, was measured with a SQUID magnetometer in fields of 3 mT and 0.5 T. See Fig.1. In the lowest fields, \( \chi \) first increases below \( T_N \), before it saturates below 10 K. A larger field suppresses these apparent basal-plane fluctuations, yielding a maximum around 7 K.

![Figure 1: Low-field magnetization of UNi4B and UCoNi3B.](image1)

The specific heat of these samples, plotted in Fig.2 on a logarithmic temperature scale, clearly shows the reduction of \( T_N \) with increasing Co-concentration. For pure UNi4B, an increase of \( c/T \) is observed below 7 K, in accord with the susceptibility maximum. The increase in \( c/T \), which is almost field-independent [2], follows a weak \( \ln T \) dependence, reminiscent of the formation of an unusual Fermi-liquid state [3,4]. An extension towards lower \( T \) is necessary to confirm this \( \ln T \)-dependence. The extrapolated \( \gamma \)-values are 269 and 294 mJ/mol K\(^2\) for UNi4B and UCoNi3B, respectively.

![Figure 2: Specific heat of UNi4B (o), UCoNi3B (+) and UCo2Ni2B (●) plotted as c/T on a logarithmic temperature scale.](image2)

If we combine these new results with those obtained earlier [1,2], we can establish the magnetic phase diagram for UNi4B. This diagram, shown in Fig.3, is a combination of high-field magnetization, specific heat and resistivity, both in magnetic fields, for two field directions in the basal plane. The AF phase boundary lies at 20 T. A spin-flop transition is found for \( \mu_0 H = 8 \) and 11 T for the two directions, respectively. The low-T regime for in-plane fluctuations is also indicated (shaded area in Fig.3).

In conclusion, we have shown that the ordering temperature of UCo\(_{x}\)Ni\(_{4-x}\)B strongly depends on the 5f–3d hybridization strength. We have presented a detailed magnetic phase diagram for UNi4B, which incorporates a highly unusual low temperature spin state, thought to arise from large in-plane fluctuations in the antiferromagnetically ordered state.

**References**


