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ELSEVIER



Magnetic phase diagram and low-dimensional excitations of hexagonal UNi₄B

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Abstract

The hexagonal antiferromagnet UNi₄B ($T_N = 20$ K) exhibits highly unusual magnetic properties, such as huge anisotropy and increasing electronic specific heat and susceptibility below T_N . New high-field magnetization, magnetoresistance and thermoelectric power measurements are analyzed using the recently resolved zero-field magnetic structure [1], which consists of six ordered and three paramagnetic spins.

In previous papers [1–3] we have presented the anomalous low-temperature behavior of the hexagonal intermetallic compound UNi₄B, crystallizing in a superstructure of the CeCo₄B-type structure, which orders in a unique antiferromagnetic spin arrangement below $T_N = 20$ K. This material exhibits the effects of strongly anisotropic hybridization combined with those of antiferromagnetic interactions on a triangular lattice [4,5], introducing spin frustration of magnetic uranium moments. The magnetic structure is described by a magnetic unit-cell with lattice parameters $a_{\text{magn}} = 3a$ and $c_{\text{magn}} = 1/2c$. Only six out of nine U-moments, oriented in the basal plane, are ordered in a vortex-like arrangement around the three remaining, paramagnetic moments [1]. Here we report on new high-field magnetization experiments up to 50 T, magnetoresistance and thermoelectric power measurements, on well-characterized single crystals. The magnetic phase diagram is presented, and the anomalous low-temperature magnetic and transport properties are described using the predictions for low-dimensional magnetic excitations [6].

Fig. 1 displays high-field magnetization data taken at 4.2 K on a single crystal of UNi₄B with fields along the two basal plane axes a (left panel) and b (perpendicular to a , right panel). Depending on this field orientation, three or two hysteretic spin-flop phases are observed, respec-

tively. Along a , these fields are $\mu_0 H_2 = 8.2$ T, $\mu_0 H_3 = 11.7$ T and $\mu_0 H_0 = 19.8$ T. Along b , the first spin-flop field is $\mu_0 H_1 = 9.3$ T at 4.2 K (the two spin-flop fields for $H \parallel b$ at 1.4 K are 8.1 and 18.6 T). Note that the magnetization jump is only 0.1–0.2 μ_B , in accord with the hexagonal magnetic structure determined in Refs. [1,7]. Interestingly, the magnetization still does not saturate in fields as large as 52 T. This implies antiferromagnetic nearest and next-nearest neighbor coupling between the U-moments, which in zero field are equal to $1.2(2)\mu_B$ per ordered spin. This value is exactly recovered if the magnetization per formula unit is extrapolated in a plot of M vs. $1/H$ towards $1/H = 0$, indicating that the paramagnetic spin also reaches $1.2\mu_B$ in high fields. The value of the paramagnetic spin in zero field is not known. Along the c axis, the magnetization is linear with field and reaches $0.12\mu_B/\text{f.u.}$ in 35 T [2].

The anisotropy of the *first* spin-flop field is demonstrated in a transversal magnetoresistance experiment, at

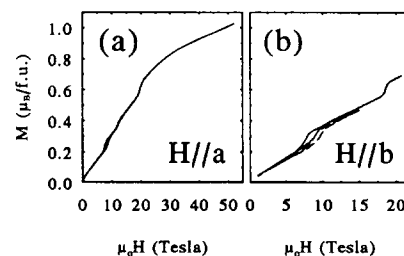


Fig. 1. High-field magnetization of UNi₄B. (a) Field parallel to the a -axis at $T = 4.2$ K. (b) Field parallel to the b -axis (perpendicular to a in the basal plane) at 1.4 K (—) and 4.2 K (---).

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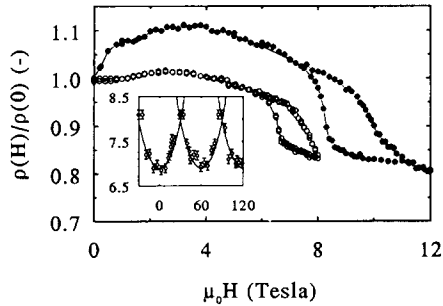


Fig. 2. Magnetoresistance of UNi_4B at $T = 1.4$ K with the current along the c -axis and the magnetic field parallel to a (\circ) and b (\bullet). The inset shows the angular dependence of the first spin-flop field, H_2 . The solid line describes the $(1/\cos\theta)$ -behavior of H_2 .

$T = 1.4$ K, with the current along the c axis and field parallel to a and b , see Fig. 2. Along a , the spin-flop field $\mu_0 H_2$ equals 7.0 T, along b $\mu_0 H_1 = 8.1$ T. The inset of Fig. 2 shows the angular dependence of this spin-flop field, which closely follows the expected $(1/\cos\theta)$ -behavior, with θ the angle between the a -axis and the magnetic field.

The huge anisotropy of UNi_4B in transport properties was further studied by thermoelectric power (S) measurements, shown in Fig. 3. Along the basal-plane direction, S is small and a pronounced, reproducible minimum is found around 215 K. No anomaly is found at T_N , reflecting the sustained very effective scattering mechanism in the ordered state. In contrast S parallel to c is rather large, and displays an upward kink at T_N , reminiscent of the formation of new magnetic superzone boundaries due to the AF ordering.

Combination of all magnetization, resistivity and specific-heat data yields the magnetic phase diagram of UNi_4B , with fields parallel to the basal plane, displayed in Fig. 4. The AF phase boundary lies at 19.8 T ($\parallel a$) and 18.6 T ($\parallel b$). The low-temperature, low-field region is indicated by the dashed line. This crossover line marks the maxima as observed in the magnetoresistance measurements. Although the zero-field magnetic structure is known, the

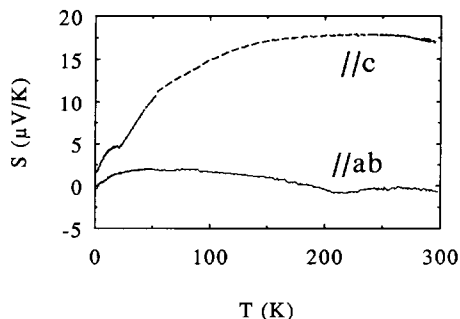


Fig. 3. Thermoelectric power of UNi_4B parallel to the basal plane (---) and along the c -axis (---).

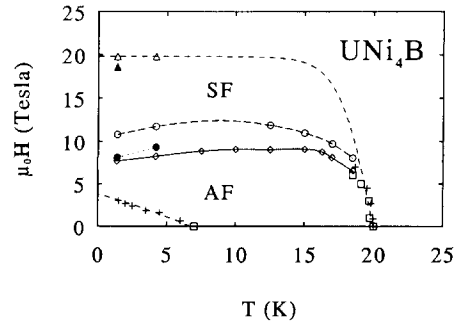


Fig. 4. Magnetic phase diagram of UNi_4B for fields parallel to the basal-plane axes a (open symbols) and b (closed symbols) as derived from magnetization, magnetoresistance and specific-heat experiments. The dashed line at low fields marks the crossover line (see text).

nature of the field-induced phases remains to be established. Of special interest again is the role the paramagnetic spins play in the high-field magnetization process.

The experiments on UNi_4B in zero and low magnetic fields were analyzed in terms of unique magnetic ordering and one-dimensional ferromagnetic (1D FM) excitations of the chains of paramagnetic U-spins [1,7]. These 1D FM excitations give rise to increasing susceptibility and electronic specific heat below T_N , and weak power-law behavior of specific heat and resistivity. Here we have demonstrated that the application of large magnetic fields in the basal-plane direction quenches the influence of these excitations, giving way to the regular behavior of an antiferromagnet. We propose that the crossover line in the phase diagram is related to this quenching. At the highest fields, the paramagnetic spins approach the same moment value as the ordered spins. Further neutron-diffraction studies on single crystals in magnetic fields are necessary to establish the nature of the spin-flop phases and possible unusual critical behavior, as expected for ordering on triangular lattices with antiferromagnetic interactions.

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