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Effect of interstitial C on the antiferromagnetism of UNi_4B

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Abstract

We performed measurements of the specific heat, resistance, magnetization and DC-susceptibility of UNi_4BC_x with $0 \leq x \leq 0.1$. We found that the interstitially substituted carbon strongly affects the magnetism of the UNi_4B host matrix. While UNi_4B orders antiferromagnetically below 20 K, T_N is reduced to 12 K for $\text{UNi}_4\text{BC}_{0.1}$. We discuss possible mechanisms for this tuning of the antiferromagnetism.

1. Introduction

Within the last few years the physical properties of UNi_4B , crystallizing in the hexagonal CeCo_4B -structure, have been investigated in detail [1,2]. It was found that UNi_4B undergoes an unusual antiferromagnetic transition of U moments below $T_N = 20$ K in zero field. Ni is non-magnetic. Furthermore peculiar spin-wave excitations have been seen below 7 K in the macroscopic measurements.

Recently, the magnetic structure of UNi_4B has been determined [3]. The magnetic ordering consists of two arrangements around central U atoms of (i) 6 U spins arranged in the basal plan like a vortex and (ii) 3 pairs of 2 antiferromagnetic spins. In both arrangements the net magnetic exchange interaction at the location of the central U atom is zero and its moment remains paramagnetic. Along the *c*-axis the spins are ferromagnetically coupled. Thus, at lowest temperatures ferromagnetic spin-waves appear due to the central U atoms.

Apart from the magnetic structure a crystallographic superstructure has been found in UNi_4B , and it was argued that this superstructure is related to the magnetic structure. Very recent work [4] has claimed that addition of small amounts of C to isostructural YNi_4B

destroys the crystallographic superstructure and causes superconductivity at ≈ 12 K. Therefore, we started this examination of UNi_4BC_x in order to determine how the magnetism and superstructure would be modified, and if superconductivity would occur.

2. Metallurgical details

Polycrystalline sample have been arc-melted in stoichiometric ratio (U, 3 N; B, 3 N; C, 3 N; Ni, 4 N) under an argon atmosphere. The carbon loaded samples were annealed in evacuated quartz-tubes at 900°C for 5 days. As reference sample we used polycrystalline UNi_4B annealed at 700°C for 7 days. All samples were checked by X-ray diffraction and electron probe micro analysis (EPMA). Our EPMA is sensitive to boron, whereas we can only determine carbon directly in the given phase for amounts higher than about 20 at%. Thus the C concentrations are nominal for UNi_4BC_x .

The results of the metallurgical analysis are summarized in Table 1. The carbonized samples show the presence of minority phases in a matrix of UNi_4B -stoichiometry. Only for UNiBC could we resolve the absolute amount of C. For the other second phases, as well as for the matrix, the carbon signal in the EPMA was too low to evaluate the C concentration. However, our data on physical properties and the X-ray

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Table 1

Composition and lattice parameters of UNi_4BC_x (the notation (C_y) indicates unknown amount of C)

x		a (Å)	c (Å)	V (Å ³)
0	98% UNi_4B 2% UNiB	4.953	6.964	147.95
0.05	93% $\text{UNi}_4\text{B}(C_y)$ 5% UNiBC 2% $\text{UNi}_{9.9}\text{B}(C_y)$	4.936	6.984	147.37
0.1	80% $\text{UNi}_4\text{B}(C_y)$ 10% $\text{UNi}_4(C_y)$ 8% UNiBC 2% $\text{UNi}_{9.7}\text{B}_{2.4}(C_y)$	4.934	6.985	147.26

analysis indicate that the matrix is loaded with C. Although we cannot determine the exact amount of carbon in the matrix, we note that it will be somewhat lower than the nominal x in UNi_4BC_x , for the main second phase UNiBC is carbon rich, and consequently, the remaining phases will have a lower C concentration.

X-ray diffraction was used to investigate the effect of C on the structural parameters and on the superstructure. Clearly the c/a -ratio is changed by adding C. Unfortunately, even in pure UNi_4B the superstructure reflections are quite weak. Hence, we were unable to see the superstructure in any of our samples. We therefore cannot draw a conclusion about the influence of C on the superstructure.

3. Experiments

For measurements of the physical properties we used a standard adiabatic technique to measure specific heat c_p from 2 to 30 K. Magnetization up to 12 T and DC-susceptibility up to 50 K were performed in a Foner-magnetometer. A 4-point AC-technique between 1.3 and 300 K was employed to measure resistance.

In Fig. 1 c_p/T versus T of UNi_4BC_x with $x = 0, 0.05$ and 0.1 is plotted. The main features of pure UNi_4B , the antiferromagnetism and the low-temperature anomaly due to spin-wave excitations, remain unaffected by C loading apart from the decrease in the Néel temperature. This temperature T_N decreases from 20 K at $x = 0$ to 12 K for $x = 0.1$. Also the minimum in c_p/T versus T below T_N is shifted down in temperature. Further, the electronic contribution to the specific heat γ , extrapolated from c_p above T_N , is significantly lowered with increasing x ($\gamma_{x=0} = 0.24$ J/

Fig. 1. Specific heat divided by temperature as a function of temperature for UNi_4BC_x .

mol K²; $\gamma_{x=0.05} = 0.17$ J/mol K²; $\gamma_{x=0.1} = 0.12$ J/mol K²).

Minority phases cannot account for these effects. This is evident for the shifts of T_N and the minimum in c_p/T . But also the decrease in γ cannot be due to secondary phases. Since there are no anomalies in c_p other than those of UNi_4BC_x , the contributions of the impurity phases should behave as $\gamma_{\text{imp}}T + \beta_{\text{imp}}T^3$. In the worst case γ_{imp} would be zero and γ of UNi_4BC_x higher than the above reported numbers. However, estimating the effective γ -values for this case, we only obtain an increase of 10–20% from the values given above for $x = 0.05$ and 0.1. With any non-zero γ_{imp} the real γ will be lower than this worst case scenario.

A similar shift of T_N , determined as minimum of $\partial^2 R / \partial T^2$ (see Fig. 2), which corresponds to the

Fig. 2. Normalized resistance of UNi_4BC_x as a function of temperature. In the insert $\partial^2 R / \partial T^2$ versus T for $x = 0, 0.05$ and 0.1 is plotted. The minimum in $\partial^2 R / \partial T^2$ corresponds to the antiferromagnetic kink.

position of the kink in R versus T , can be observed in the normalized resistance of UNi_4BC_x . The change in the general shape of the resistance with carbon substitution as seen in Fig. 2 probably reflects the influence of the second phases forming low resistance paths in the samples.

The tuning of T_N is also visible in the susceptibility (not shown). And in the magnetization versus field (not plotted) the first spin-flop transition reported for UNi_4B can be identified for all samples. The transition fields increase with C addition. No superconductivity was observed in any of our samples down to 1.3 K. Another sample of nominal composition $\text{UNi}_4\text{BC}_{0.2}$ showed no trace of superconductivity even down to 0.4 K.

4. Discussion

From our data we can construct a B – T phase diagram. Carbonizing UNi_4B decreases T_N , while the field of the first spin-flop transition slightly increases. This behavior probably reflects the increase in crystallographic anisotropy, as found from X-ray diffraction.

A possible model is that adding C to UNi_4B simulates the effect of pressure on the a - and b -axes. Although no data on elastic constants of UNi_4B are yet available, we can at least roughly estimate the order of magnitude of pressure. The lattice parameters $\Delta c/c$ and $\Delta a/a$ change with addition of carbon about 0.3% and –0.3%, respectively. Typically lattice parameter changes $\Delta x/x$ are about 0.1–0.01% kbar^{-1} . Therefore the shift of T_N could be the result of

chemical pressure of the order of 10 kbar. This is a comparable large, yet not impossible, pressure effect. Nevertheless, we are aware that the influence of C on the band structure should also be taken into account.

In conclusion we have presented data on UNi_4B with interstitially substituted carbon up to $x = 0.1$. By adding small amounts of carbon, the magnetism and the anisotropy of UNi_4B can be tuned. We suggest that these effects can be explained in terms of chemical pressure. But to verify this, an examination of the magnetism of UNi_4B under pressure will be necessary.

Acknowledgement

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