Journal of Alloys and Compounds, 207/208 (1994) 254–256
JALCOM 3042

Magnetic properties of TbNiAl

H. Maletta
Hahn-Meitner-Institut, Berlin (Germany)

V. Sechovsky
Charles University, Prague (Czech Republic)

Abstract

The intermetallic compound TbNiAl which crystallizes in the hexagonal ZrNiAl structure type has been studied by means of neutron powder diffraction. Its magnetic phase diagram turned out to be more complex than reported previously. The antiferromagnetic order below $T_N = 44.6$ K can be described by two propagation vectors $\mathbf{k}_1 = (1/2,0,1/2)$ and $\mathbf{k}_2 = (1/2,1/2,1/2)$. A second transition temperature $T_1 = 23.5$ K is found where, most probably, the frustrated spins change direction and/or the magnitude of the moments.

1. Introduction

This work is part of an extended program focused on the understanding of relations between the electronic structure of 4f and 5f metals in intermetallic compounds and their magnetic and related electronic properties. An interesting question arises about the symmetry of the magnetic ground state in RTX (R = rare earth) and UTX compounds crystallizing in the same structure. Surprisingly, much more systematic information in this respect is available for UTX (T = transition metal, X = p-metal) [1]. Therefore, we are now studying RTX compounds, starting with TbNiAl.

2. Experimental details

A polycrystalline sample of TbNiAl was prepared at HMI by inductive levitation melting under an argon atmosphere. The button was turned over and remelted several times to improve homogeneity. X-Ray powder diffraction using Co Kα radiation showed single-phase material, crystallizing in the hexagonal ZrNiAl-type structure.

Magnetization and susceptibility measurements were performed using a SQUID magnetometer (DC) and an AC- susceptometer. Here, we report mainly on our neutron diffraction experiment on the powder sample of TbNiAl carried out at wavelength of $\lambda = 2.3869$ Å on the E6 diffractometer at HMI, Berlin.

3. Results and discussion

Both the X-ray and neutron diffraction patterns show TbNiAl to crystallize in the hexagonal ZrNiAl-type structure (space group *P*-62*m*), an ordered ternary derivative of the Fe_2P structure. Systematic structural investigations of various RTX compounds were first carried out by Dwight [2].

Analyzing our neutron diffraction spectra by means of the Rietveld refinement method, we obtain the lattice constants $a = b = (689.94 \pm 0.08)$ pm and $c = (397.50 \pm 0.09)$ pm. The unit cell contains three Tb atoms at the positions $x,0,1/2$; $0,x,1/2$; $-x,-x,1/2$ (Wyckoff notation = 3g) and three Al atoms (3f) at $x,0,0$; $0,x,0$; $-x,-x,0$. The three Ni atoms per unit cell are distributed in different layers: two Ni atoms (2c) at 1/3,2/3,0; 2/3,1/3,0, and one Ni atom (1b) at 0,0,1/2. The parameter x is found to be $x = 0.5849 \pm 0.0012$ for the Tb position and $x = 0.2458 \pm 0.0030$ for Al.

Bulk measurements on our sample reveal TbNiAl as an antiferromagnet with several characteristic temperatures; at least two of them are now confirmed as magnetic transition temperatures by our neutron diffraction experiment: $T_N = 44.6$ K and $T_1 = 23.5$ K. Figure 1 displays the temperature dependence of the AC-susceptibility (the real part of χ) as an example. In addition to the anomaly in $\chi(T)$ near the Néel temperature T_N (peak in $d\chi/dT$ at T_N) and the change in slope of $\chi(T)$ near T_1, we observe two further anomalies at 7.5 K and at 51 K; both are not yet seen in the powder neutron diffraction. The existence of competing interaction effects near T_N, however, becomes even more pronounced in the curves in the lower part of

0925-8388/94/$07.00
SSDI 0925-8388(93)03042-H

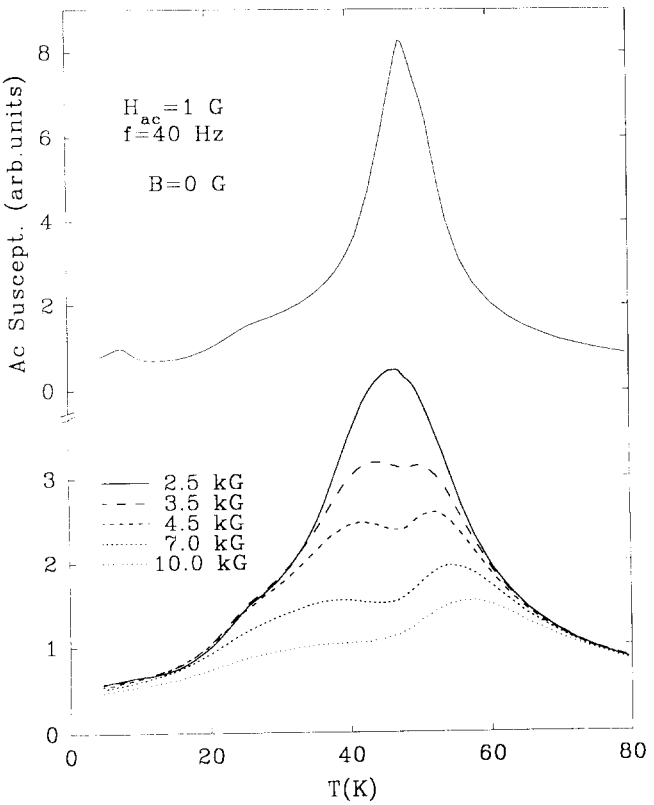

Fig. 1. Temperature dependence of the AC susceptibility (the real part of χ) of TbNiAl (a) in zero and (b) in various applied DC magnetic fields.

Fig. 1, where AC-$\chi(T)$ was measured in various applied DC magnetic fields. While the further magnetic transition near 51 K is detectable only as a small shoulder in the AC-susceptibility in $B=0$, two well pronounced peaks emerge in $\chi(T)$ when applying a static field of about 5 kG. Both peaks are shifted in opposite directions with increasing field (Fig. 1).

Many magnetic Bragg peaks were observed in the neutron diffraction experiment on TbNiAl below T_N which can be indexed as (1/2,0,1/2), (1/2,1/2,1/2), (1,1/2,1/2), (3/2,0,1/2), *etc.* Hence, the magnetic pattern indicates a magnetic unit cell of the dimensions $2a \times 2b \times 2c$, it consists of eight hexagonal crystallographic unit cells. All the magnetic Bragg peaks can be described by two magnetic propagation vectors, $\mathbf{k}_1^m = (1/2,0,1/2)$ and $\mathbf{k}_2^m = (1/2,1/2,1/2)$. It holds for the whole temperature regime of $2\,K < T < T_N$, and up to now no additional magnetic reflections could be observed in our powder diffraction experiments on E6.

The integrated intensity of the first five magnetic Bragg reflections are shown as a function of temperature in Fig. 2. All the magnetic reflections go to zero at the Néel temperature $T_N = 44.6$ K, and several anomalies are indicative of the second magnetic transition at $T_1 = 23.5$ K. Two peaks at (1/2,0,1/2) and (3/2,1/2,1/2) almost vanish at T_1 (reduction to 1/6 of the intensity

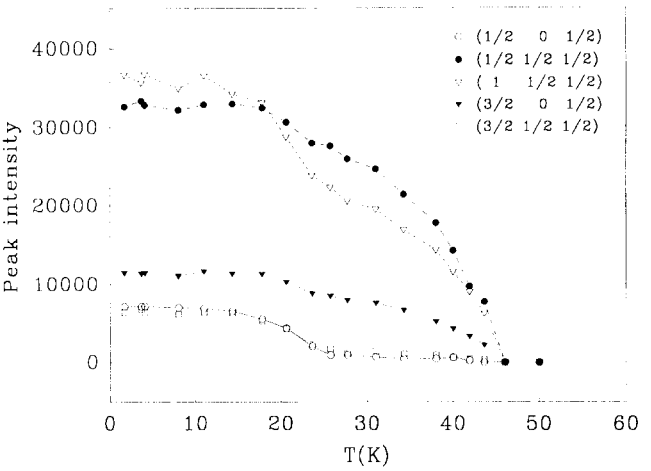

Fig. 2. Integrated intensity of various magnetic Bragg peaks in TbNiAl as a function of temperature.

at the lowest T), whereas only a smooth behavior is observed for the other two peaks at (1/2,1/2,1/2) and (3/2,0,1/2) around T_1. Intermediate behavior is measured for the Bragg peak at (1,1/2,1/2) which reduces to half the saturated intensity near T_1.

The microscopic model for the spin structure of TbNiAl is derived from the neutron diffraction spectra by simultaneous Rietveld refinement of both the nuclear and the magnetic structure. The best result is obtained by the assumptions (i) that the magnetic moments appear exclusively on the Tb atoms (no moment on the Ni) which is also verified by other experimental results, and (ii) that the magnetic moments on all the Tb atoms are aligned parallel to the c-axis.

Hence, the crystal structure which consists of two kinds of layers stacking in the sequence TbNi–NiAl–TbNi–NiAl along the c-axis, is built up of layers with magnetic Tb atoms (at $z=0.5$, 1.5, *etc.*) which are separated by non-magnetic NiAl-layers (at $z=0$, 1, *etc.*).

The analysis of the neutron spectrum at 8 K yields the lowest refinement factors R for the spin structure which is sketched in Fig. 3. The directions of the magnetic moments of Tb are parallel (+) and antiparallel (−) to the c-direction; the spin arrangement within the (a,b)-plane of the magnetic unit cell at $z=0.5$ is displayed, whereas that at $z=1.5$ is *vice versa*.

We cannot yet distinguish between two models for the spin structure at 8 K:

(1) The first model gives the refinement factor $R_m = 3.36$. Here all the Tb atoms carry the same moment of $(7.903 \pm 0.028)\ \mu_B$. At first, this assumption is rather reasonable with respect to the equivalence of all the Tb sites in this crystal structure.

(2) The second model gives $R_m = 3.14$. Here we assume the existence of two magnetically different Tb moments and obtain $(8.073 \pm 0.059)\ \mu_B$ and

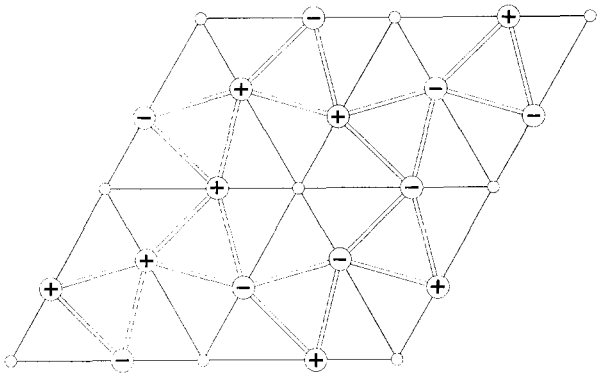

Fig. 3. Arrangement of the Tb magnetic moments in the basal plane of the magnetic unit cell ($=2a\times 2b$) of TbNiAl below T_1: parallel (+) and antiparallel (−) alignment to the c-axis.

$(7.510\pm 0.130)\ \mu_B$. The spin structure in Fig. 3 can be described as antiferromagnetically coupled Ising spins on a two-dimensional lattice of various connected triangles, which creates eight frustrated spins per magnetic unit cell; four of them at the $(-x,-x,1/2)$ crystallographic positions can be found in the $(z=0.5)$ layer in Fig. 3. These Tb atoms (+,−,+,−) have the somewhat lower magnetic moment in the second model.

It is important to note that both models break down above T_1. The analysis of the neutron spectrum at 27.7 K by means of the first model gives $R_m=22.8$ and $(5.981\pm 0.055)\ \mu_B$, whereas the second model yields $R=13.4$ with $(7.320\pm 0.078)\ \mu_B$ and $(0.2\pm 0.4)\ \mu_B$. Thus, it is clear that some changes in the spin alignments or the spin magnitude occur at $T_1=23.5$ K, most probably for the frustrated spins.

This short report on our recently started neutron diffraction study on polycrystalline TbNiAl presented a microscopic model for the antiferromagnetic order in TbNiAl at low temperature. Our studies revealed that it does not order ferromagnetically and its magnetic phase diagram is more complex than reported previously [3].

Acknowledgments

We wish to thank W. Rönnfeldt at HMI for preparation of the sample and Dr. M. Reehuis for assistance with the start up of the Rietveld program.

References

1 V. Sechovsky and L. Havela, *Phys. Scripta, T45* (1992) 99 and refs. therein.
2 A.E. Dwight *et al.*, *Trans. Metal Soc. AIME, 242* (1968) 2075.
3 H. Oesterreicher, *J. Less-Common Met., 30* (1973) 225.