Field induced phases in a GMR system TbNiSn

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Abstract. Results of neutron diffraction measurements on TbNiSn in magnetic fields up to 6T are reported. This material shows successive phase transitions at $T_1 = 7.6$ K, $T_2 = 6.0$ K, $T_3 = 2.0$ K and $T_N = 18.5$ K in zero field, and a giant magnetoresistance effect (GMR) near four successive metamagnetic transitions at low temperatures. Propagation vectors and main signatures of the magnetic phases appearing at elevated fields applied along the b axis at low temperatures are determined. We find large hysteresis at several phase boundaries. In some cases a co-existence of magnetic structures is detected.

1.Introduction

In the last two decades materials have been found for which the canonical definition of the magnetoresistance does not apply. Instead of an increase with the increasing field a significant decrease of the electrical resistivity by as much as -90% with increasing fields, has been found in some intermetallic compounds [1] and magnetic multilayers [2]. Such phenomenon is referred to as a giant magnetoresistance (GMR) and is usually connected with a disappearance of superzone boundaries introduced into the system by an antiferromagnetic (AF) ordering. In the transition metal oxides [3], the change in the resistance with magnetic field can be much larger so the magnetoresistance is called a colossal magnetoresistance (CMR). The materials showing GMR or CMR have been a subject of intensive studies because of potential applications to magnetic recording devices.

About a decade ago, Hori and co-workers [4] found that the rare earth ternary compound TbNiSn exhibits a GMR. It forms in the orthorhombic structure with the space group Pnma with...
lattice constants: a = 7.146 Å, b = 4.445 Å and c = 7.663 Å [5]. This material exhibits AF order below TN = 18.5 K [5,6] followed by three successive order-order magnetic phase transitions at T1 = 7.6 K, T2 = 6.0 K and T3 = 2.0 K [6]. We denote the magnetic phases stable between Tc and T1, T1 and T2, T2 and T3 and below T3 as A, B, C and D, respectively. For a magnetic field applied along the easy b-axis at low temperatures, successive metamagnetic transitions occur at Bc1 = 0.6 T, Bc2 = 1.8 T, Bc3 = 4.3 T and Bc4 = 5.3 T leading to saturated magnetization of 8.1μB/Tb [4,7] as shown in Fig. 1. A GMR is observed near these critical fields [4].

Zero field neutron diffraction using powder samples [5] and single crystals [6,8] showed that the phase A is a sine-modulated magnetic structure with a wavevector qA = (0.311, 0.325, 0.0) at 14 K followed by the B phase with qB = (0.665, 0.330, 0.0) between T2 < T < T1 [6] and an antiphase-type structure (phase C) with qC = (0.399, 0.344, 0.0) below T2 [5]. Single crystal measurements [6,8] revealed that the C phase is not sine-wave modulated but squared-up one. We note that in different studies different, but equivalent, propagation vectors are utilized and that no report has been published regarding the ground state phase D stable below T3.

2. Experimental

The single crystal of TbNiSn was grown by the Czochralski method using an induction furnace. Magnetization measurements were done with a MagLab VSM vibrating-sample magnetometer (Oxford Instruments, UK) and measured at low temperatures in fields along the principal directions up to 6 T. Neutron diffraction measurements were performed on the E4 diffractometer installed at the BER-II reactor at the Helmholtz Zentrum Berlin (formerly HMI). The incident-neutron wavelength λ = 2.44 Å was used. The E4 diffractometer was at the time of the experiment equipped with a single detector. When necessary, two λ/2 filters were used which leave a residual λ/2 contamination at a level of less than 10⁻⁴. The external magnetic field generated by a 6 T horizontal magnet was applied parallel to the b* axis of the crystal which is the easy direction of the magnetization [8]. In this geometry, which is quite restrictive due to dead regions produced by the magnet coils, reflections of the (hk0) type were in horizontal scattering plane. The individual scan profiles were analyzed by fitting to Gaussian profiles and the appropriate scattering lengths and the standard Tb⁵⁺ magnetic form factor were used in the refinement. We also followed the intensities of few reachable nuclear and magnetic reflections as a function of temperature and magnetic field by monitoring the peak intensities.

3. Results and Discussion

In Fig. 1 we show the field dependence of the magnetization measured at 1.5 K with field applied along the b axis in continuously swept applied field. As can be seen, in the present measurement, the values of critical fields, defined as the average of mid-points of sweep-up and sweep-down branches, are Bc1 = 0.6 T, Bc2 = 2.1 T, Bc3 = 4.3 T and Bc4 = 5.4 T. In the present experiment some of the transitions appear to be quite hysteretic. In the previous measurement [7], however, no appreciable hysteresis was observed at any of the critical fields. We conclude therefore that this is a signature of a non-equilibrium state caused by non-zero field sweeps being 0.1T/min. Moreover, we observe an additional transition at 0.4 T, which we denote B′c1. The saturation moment is achieved above the Bc4 and amounts to about 8.1μB/Tb. Very remarkable are the plateaus visible between the relevant critical fields showing magnetization values that are roughly 1/18, 1/9, 1/3 and 2/3 of the saturated value, respectively.

At 1.5 K in zero external field one anticipates a ground-state AF structure (phase D) in TbNiSn. A mesh scan in the vicinity of (2/3, 1/3, 0) reciprocal position shows a coexistence of two magnetic reflections that can be described by two different propagation vectors, namely by q1 = (0.685, 1/3, 0) and q2 = (0.6, 0.35, 0). This observation is new and suggests a non-trivial ground
state of TbNiSn. This observation will be discussed in detail elsewhere [9]. We want only to note that in TbNiSn different magnetic structures compete.

Fig. 2 shows the field dependence of the diffracted intensity measured at the (2,0,0) reciprocal position. Because of the restrictive geometry of the horizontal magnet, this intensity was measured by removing the filters and using $\lambda/2$ wavelength with the spectrometer set at the forbidden (1,0,0) position. Although the scaling with other reflections is a more complicated, the calculation of the magnetic moment component assumed to be aligned along the b axis is independent of this fact. It appears that above $B_{c4} = 5.3$ T one deals indeed with the ferromagnetic component close to $9.0 \mu_B$/Tb. Also the relevant critical fields are in good agreement with the magnetization measurement shown in Fig. 1. The (0,2,0) reflection measured under identical conditions is absent for all the fields. This demonstrates that the Tb magnetic moments for all the field-induced magnetic structures are most probably oriented along the b axis. Let us denote the phases between $B_{c1}$ and $B_{c2}$ as phase E, between $B_{c2}$ and $B_{c3}$ as F and between $B_{c3}$ and $B_{c4}$ as G before arriving to the field-forced ferromagnetic phase H. The $B_{c1}'$ is caused by a transition from the phase D to C that is stable up to $B_{c1}$. This phase is not discussed here.

In Figs. 3a and 3b we show the field dependence of the diffracted intensity measured at the (4/3, -1/3, 0) and (2/3, 0, 0) reciprocal positions, respectively. On increasing the field, the intensity measured at the first position shows a step-like increase at $B_{c1}$, then a slow decrease and an abrupt disappearance at $B_{c3}$. With decreasing field one observes a hysteresis around $B_{c3}$ and clearly different behavior in the low-field limit. However, within several minutes the intensity decays to its original zero field value (see the inset of Fig. 3a). At the (2/3, 0, 0) position the signal appears with increasing field at $B_{c2}$, increases steadily and then sharply decreases at $B_{c3}$, followed by an increase with a maximum around 5 T and finally decreases and vanishes at $B_{c4} = 5.4$ T. With decreasing field this signal shows a significant hysteresis around $B_{c2}$ and $B_{c3}$ and vanishes in the low field region. The described observation is highly unusual as it suggests coexistence of different propagation vectors at fields between $B_{c2}$ and $B_{c3}$. The first propagation vector is $q_E = (1/3, 1/3, 0)$ which is a unique vector for the phase E and the other $q_G = (2/3, 0, 0)$ for phase G. Phase F is described by $q_E$ and $q_G$ simultaneously. For all the field induced phases a ferromagnetic component also exists.

The symmetry group analysis considering the Pnma space group and propagation vectors $q_E$ and $q_G$ respectively allows Tb moments to be oriented either along the b axis or within the a-c plane for the $q_G$ and to have all the Cartesian components for the vector $q_E$. Consideration of the
ferromagnetic components leads to an approximate magnetic structures that are depicted to the Fig. 4. Although, in general, the main features are correct, some details remain still unclear. This applies especially to phase E. The spin arrangement shown in Fig. 4a would imply small non-zero intensity at the (0,2,0) position that is not observed. It is not clear at this moment whether this is due to domains or whether the proposed magnetic structure is incorrect. Due to complexity of the magnetic ordering in TbNiSn under influence of magnetic field, further study is needed.

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