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Spontaneous and field-induced phase transitions in $TbFe_5Al_7$

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ABSTRACT

Magnetization and ultrasound measurements have been performed on a $TbFe_5Al_7$ single crystal (tetragonal crystal structure) in the temperature range from 2 to 260 K in steady magnetic fields up to 18 T and in pulsed magnetic fields up to 60 T. The compound is a ferrimagnet (T_C=242 K) having an easy-plane anisotropy. Strong anisotropy is also present within the basal plane. At 2 K, the easy magnetization direction is the [1 0 0] axis. In the vicinity of the compensation temperature, T_{comp}=84 K, $TbFe_5Al_7$ displays a spin-reorientation transition from [1 0 0] to the [1 1 0] axis accompanied by pronounced anomalies in the relative sound-velocity change and sound attenuation. Further, field-induced magnetic transitions have been observed in $TbFe_5Al_7$ by magnetization and acoustic measurements. Step-wise rotation of the magnetic moments with a wide hysteresis occurs for fields applied along the [1 0 0] axis at $T<T_{comp}$ and along the [1 1 0] axis at $T>T_{comp}$. The relative sound-velocity change displays sharp minima and the sound attenuation sharp maxima at the transitions. The critical field of the transitions tends to zero near the compensation point and grows sharply away from it reaching 19 and 33 T for fields applied along the [1 0 0] and [1 1 0] directions, respectively. The Tb–Fe inter-sublattice exchange constant has been determined directly from the high-field data and using molecular-field theory.

1. Introduction

Intermetallic compounds based on rare-earth elements and iron, crystallizing in the tetragonal crystal structure of the $ThMn_{12}$ type, form a wide group of materials with two magnetic sublattices (for a review see Ref. [1]). Some of them, e.g., $SmFe_{11}Ti$, are promising candidates for materials for permanent magnets due to their good hard magnetic properties [2–6]. On the other hand, there are compounds that display highly evolved magnetic behavior that presents a challenge for our fundamental understanding. An example is provided by materials of the type RFe_5Al_7 (R=heavy rare-earth element or Y) [7–14]. In the compounds with R=Tb, Dy, Ho, and Er a strong single-ion contribution from the rare-earth sublattice to the magnetic anisotropy was found [15–18]. They display ferrimagnetic order. At low temperatures the magnetic moment of the rare-earth sublattice exceeds that of the iron sublattice, and the opposite is observed at higher temperatures. As a result, these materials have a compensation point where the total magnetization becomes zero. In the vicinity of the compensation point the anisotropy effects in ferrimagnets are known to become most noticeable [19], and in this temperature range RFe_5Al_7 exhibit complex behavior in a magnetic field. A field-induced phase transition reflecting a step-wise rotation of magnetic moments was found in all of the materials in fields up to 14 T along the easy magnetization direction lying in the basal plane [15–18]. A common feature of this phase transition is that its critical field passes through zero at the compensation temperature, T_{comp}, and grows sharply for $T<T_{comp}$ and $T>T_{comp}$. In order to access the field-induced magnetic transition away from the compensation point in the compounds with R=Dy, Ho, and Er, pulsed magnetic fields up to 60 T were used [20–22]. An additional magnetization jump was found before the systems reached the forced ferromagnetic state.

In contrast to the RFe_5Al_7 compounds with R=Dy, Ho, and Er, $TbFe_5Al_7$ displays not only field-induced magnetic phase transitions but also a spontaneous spin-reorientation transition within the basal plane of the tetragonal lattice in the vicinity of T_{comp} [18].

High pulsed magnetic fields make it possible to obtain a more complete picture of the complex magnetization process of $TbFe_5Al_7$ and to follow the evolution of the Tb–Fe inter-sublattice exchange

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interactions in a wide temperature range. Apart from magnetization, sound propagation often displays sharp anomalies at magnetic phase transitions, both spontaneous and field-induced, due to a strong spin-lattice coupling (see Ref. [23] and references therein), and provides important information on their dynamics. In the present work, magnetization and magnetoacoustics of the $TbFe_5Al_7$ compound have been studied in the wide temperature range between 2 and 260 K and in high pulsed magnetic fields up to 60 T.

2. Experimental details

A $TbFe_5Al_7$ single crystal was grown by a modified Czochralski method in a tri-arc furnace equipped with a water-cooled Cu crucible using a tungsten rod as seed under protecting Ar atmosphere. The pulling speed was 15 mm/h. Phase purity and lattice parameters were determined using standard X-ray diffractometry on a part of the single crystal crushed into fine powder. The obtained lattice parameters of the tetragonal unit cell, $a=870.3$ pm and $c=504.4$ pm, are in good agreement with literature [9,24]. Back-scattered Laue patterns were used to orient the single crystal along the principal crystallographic directions and to cut the samples for magnetic, thermal and acoustic measurements. Typical back-scattered Laue patterns are given in Ref. [20].

The temperature and field dependence of the magnetization between 2 and 260 K were measured along the [1 0 0], [1 1 0] and [0 0 1] axes using a standard PPMS-14 magnetometer with the highest available field of 14 T.

High-field magnetization curves were measured between 2 and 145 K in pulsed magnetic fields (pulse duration 20 ms) up to 60 T at the Dresden High Magnetic Field Laboratory. The magnetization was measured by the induction method using a coaxial pick-up coil system. A detailed description of the high-field magnetometer is given in Ref. [25].

Relative changes of the ultrasound velocity and ultrasound attenuation were measured using a pulse-echo technique [26] in steady fields up to 18 T between 2 and 260 K and in pulsed fields up to 63 T between 2 and 145 K. For the measurements a pair of piezoelectric film transducers was glued to the opposite parallel surfaces of the single crystal in order to excite and detect acoustic waves. Two geometries were used for the experiment: longitudinal acoustic waves were propagated with wave vector $\boldsymbol{k}$ and polarization $\boldsymbol{u}$ along the [1 0 0] axis and along the [1 1 0] axis of the single crystal. The magnetic field $\boldsymbol{H}$ was applied along the same axes.

3. Results

Fig. 1 shows the field dependence of the magnetization along the [1 0 0], [1 1 0] and [0 0 1] axes of the $TbFe_5Al_7$ single crystal at 2 and 140 K. The magnetic moments of the compound lie in the basal plane of the tetragonal lattice, the [0 0 1] axis is the hard magnetization direction. A strong anisotropy is observed within the basal plane since the magnetization along the [1 0 0] and [1 1 0] directions displays different magnetic field dependences. At 2 K, the easy magnetization direction (EMD) is the [1 0 0] axis with the spontaneous magnetic moment $M_s=1.2$ μ_B/f.u. The spontaneous moment ratio $M_s^{1\ 1\ 0}/M_s^{1\ 0\ 0}\approx\cos 45^\circ$ corresponds well to the tetragonal symmetry. From the obtained M_s value the magnetic moment of the Fe sublattice can be calculated assuming that the Tb magnetic moment is equal to its theoretical value, 9 μ_B, and that the compound is a collinear ferrimagnet (a collinear ferrimagnetic structure for $TbFe_5Al_7$ was reported in Ref. [7]): $M_{Fe}=M_{Tb}-M_s=7.8$ μ_B/f.u. This corresponds to 1.56 μ_B per Fe atom. At 140 K, the in-plane anisotropy is still present, however, the EMD

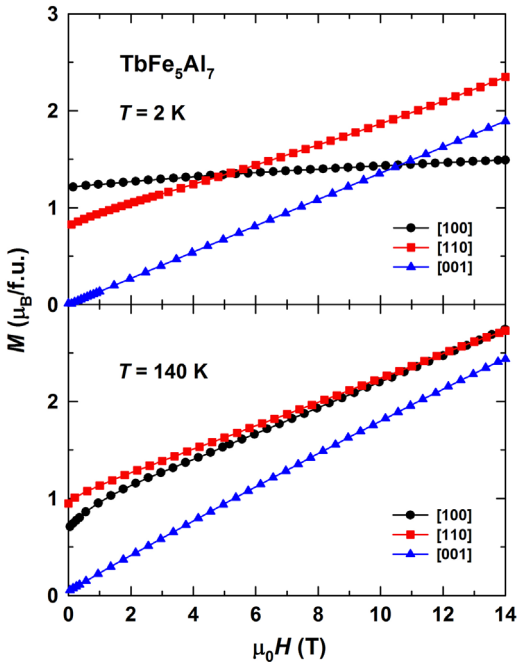

Fig. 1. Magnetization curves up to 14 T along the principal crystallographic directions of $TbFe_5Al_7$ at 2 and 140 K.

at this temperature is the [1 1 0] axis. In this case the ratio $M_s^{1\ 0\ 0}/M_s^{1\ 1\ 0}\approx\cos 45^\circ$ is fulfilled. Therefore, $TbFe_5Al_7$ displays a spontaneous spin-reorientation transition from the [1 0 0] axis at low temperature to the [1 1 0] axis at high temperature.

Fig. 2a shows the temperature dependence of the spontaneous magnetization M_s (determined from Arrott plots) for fields applied along the [1 0 0] and [1 1 0] axes. The non-monotonous temperature dependence of M_s for both directions reflects ferrimagnetic order with changing sublattice magnetizations in the compound. A compensation of the Tb and Fe sublattices is observed at $T_{comp}=84$ K. The spin-reorientation transition occurs near this compensation point. In this temperature range, the ratio $M_s^{1\ 1\ 0}/M_s^{1\ 0\ 0}\approx\cos 45^\circ$ (or $M_s^{1\ 0\ 0}/M_s^{1\ 1\ 0}\approx\cos 45^\circ$) is not fulfilled, $M_s^{1\ 1\ 0}$ values become very small. It was also found that there is no component of the spontaneous magnetic moment along the [0 0 1] axis which is the hard magnetization direction in the whole magnetically ordered state. The explanation for the low $M_s^{1\ 1\ 0}$ values is that the highest applied field of 14 T is not sufficient to induce a single domain state in $TbFe_5Al_7$ due to a considerable growth of coercivity [18]. From the magnetization measurements the spin-reorientation temperature T_{sr} could not be determined precisely.

The spin-reorientation transition affects strongly the sound propagation through $TbFe_5Al_7$. Fig. 2b shows the temperature dependence of the relative change of the sound velocity, $\Delta v/v$, and sound attenuation, $\Delta\alpha$, for a longitudinal acoustic wave propagating along the [1 0 0] axis. $\Delta v/v$ displays a broad minimum and $\Delta\alpha$ a broad maximum in the vicinity of T_{comp}. Fig. 2c shows the acoustic properties for a longitudinal wave propagating along the [1 1 0] direction of $TbFe_5Al_7$. Both, $\Delta v/v$ and $\Delta\alpha$, also display clear, but less pronounced broad anomalies in the same temperature range. The compensation point is not a phase transition since no additional symmetry is broken or new order parameter evolves at this temperature. Therefore, the observed features in the acoustic properties of $TbFe_5Al_7$ are associated with the spin reorientation. Note that similar anomalies were observed in highly magnetostrictive RFe_2 compounds near their spin-reorientation transitions