

# Magnetoresistance Anisotropy and Magnetic $H$ - $T$ Phase Diagram of $\text{Tm}_{0.996}\text{Yb}_{0.004}\text{B}_{12}$

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The antiferromagnetic ground state has been studied by transverse magnetoresistance, heat capacity and magnetization measurements, which were carried out on high quality single crystals of  $\text{Tm}_{0.996}\text{Yb}_{0.004}\text{B}_{12}$  dodecaboride in strong magnetic fields at liquid helium temperatures. Both antiferromagnetic-paramagnetic (AF-P) and spin-orientation (AF1-AF2) phase transitions have been observed, and allowed to construct a complicated magnetic  $H$ - $T$  phase diagram for this compound. Strong magnetoresistance anisotropy was found both in AF states ( $\rho(H||[110])/\rho(H||[111]) \sim 1.2$  at  $H \sim 20$  kOe) and at the critical field of AF-P transition ( $H_N[100]/H_N[111] \sim 1.25$ ) in this magnetic metal with a simple *fcc* crystal structure.

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## 1. Introduction

It was shown recently that  $\text{Tm}_{1-x}\text{Yb}_x\text{B}_{12}$  solid solutions may be considered as model compounds which enable to choose between various scenarios of antiferromagnetic (AF) instability, quantum critical behavior and mechanisms responsible for metal-insulator transition in strongly correlated electron systems [1]. In these conductors with a cage-glass structure [1, 2] the antiferromagnetic state is suppressed and the Neel temperature decreases from  $T_N(\text{TmB}_{12}) \approx 3.2$  K to  $T_N(\text{Tm}_{0.74}\text{Yb}_{0.26}\text{B}_{12}) = 0.8$  K during the transition from an AF metal to paramagnetic (P) insulator  $\text{YbB}_{12}$  with strong electron correlations [3]. In studies [1–3] fast  $4f$ - $5d$  spin fluctuations were established to be responsible for the formation of heavy fermions and the emergence of spin polarons in the  $\text{RB}_{12}$  matrix. In this work we investigate the magnetic phase transitions inside the AF state as well as between AF and P phase of  $\text{Tm}_{0.996}\text{Yb}_{0.004}\text{B}_{12}$  by transverse magnetoresistance (MR), heat capacity and magnetization measurements. To shed more light on the origin of the complicated AF ground state, we concentrate here on the research of anisotropy both of magnetoresistance and of the magnetic  $H$ - $T$  phase diagram of  $\text{Tm}_{0.996}\text{Yb}_{0.004}\text{B}_{12}$ .

## 2. Experimental details

The single crystals of substitutional  $\text{Tm}_{1-x}\text{Yb}_x\text{B}_{12}$  solid solutions were grown by vertical crucible-free induction zone melting in an inert gas atmosphere. Magnetic field, temperature and angular dependences of the transverse magnetoresistance  $\Delta\rho/\rho = f(H, T, \phi)$  were measured by dc four-probe method on the unique experimental setup described in [1]. The specific heat of  $\text{Tm}_{0.996}\text{Yb}_{0.004}\text{B}_{12}$  single crystals was studied at constant pressure over a wide temperature range 2–300 K in PPMS-9 (Quantum Design), magnetization was investigated by an Oxford Instruments vibrating sample magnetometer (up to 11 T). The quality control of the samples was performed by electron microprobe and X-ray diffraction analysis.

## 3. Results and discussion

Figure 1 shows  $\Delta\rho/\rho = f(H, T_0)$  curves obtained (a) at temperatures 2–4.2 K in the AF and P phases for  $\mathbf{H}||[110]$  orientation, and (b) at  $T = 2.1$  K for  $\mathbf{H}$  along [110], [100] and [111] axis. The specific features in MR field dependences, indicated by arrows, correspond to both spin orientation ( $H_M$ ) and AF-P ( $H_N$ ) phase transitions.

Angular dependences of resistivity presented in Fig. 2 demonstrate directly the anisotropy of MR at  $T = 2.1$  K in AF and P phases of  $\text{Tm}_{0.996}\text{Yb}_{0.004}\text{B}_{12}$ . A strong

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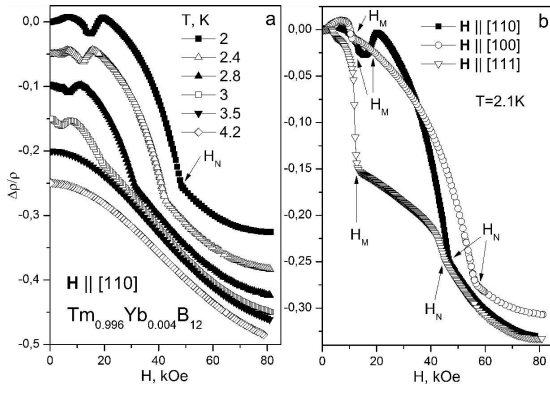


Fig. 1. Magnetic field dependencies of magnetoresistance  $\Delta\rho/\rho(H)$  in  $Tm_{0.996}Yb_{0.004}B_{12}$  (a) at various temperatures in the range 2–4.2 K and for  $\mathbf{H}||[110]$  (curves are shifted by 0.05 along vertical axis for convenience) and (b) at  $T = 2.1$  K for three main crystallographic directions.  $H_N$  and  $H_M$  are critical fields of AF-P and spin-orientation magnetic phase transitions, respectively.

anisotropy of magnetoresistance is observed in the interval 10–40 kOe (Fig. 1b and Fig. 2) and the resistivity ratio  $\rho(\mathbf{H}||[110])/\rho(\mathbf{H}||[111])$  reaches a value of  $\sim 1.2$  in the AF phase at  $H \sim 20$  kOe. Additionally, a strong difference between  $H_N$  values for  $\mathbf{H}||[100]$  and  $\mathbf{H}||[111]$  orientation can be discerned from Fig. 1b ( $H_N[100]/H_N[111] \sim 1.25$ ), pointing to an anisotropy of the AF-P phase boundary in this *fcc* metal. Further, a pronounced hysteresis, both of the angular (see Fig. 2, curves for  $H = 10$  kOe) and field dependences of MR, has been found at low magnetic fields  $H < 20$  kOe.

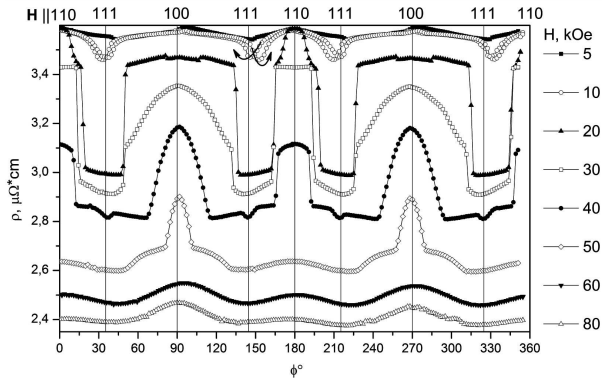


Fig. 2. Angular dependencies of resistivity  $\rho(\phi)$  in  $Tm_{0.996}Yb_{0.004}B_{12}$  at  $T = 2.1$  K for various magnetic fields.

A numerical differentiation of MR field dependences  $d(\Delta\rho/\rho)/dH = f(H, T_0)$  (see Fig. 3a) allows to detect precisely the magnetic phase transitions at  $H_M$  and  $H_N$  and, as a result, to construct in detail the magnetic  $H$ - $T$  phase diagram of  $Tm_{0.996}Yb_{0.004}B_{12}$  (see Fig. 3b). The undertaken analysis allows to interpret the data in favour of (i) an anisotropic character of the AF-P phase boundary (AF2-P in Fig. 3b) and (ii) formation of several

additional magnetic phases (AF1, AF2 in Fig. 3b) for  $\mathbf{H}||[110]$  orientation.

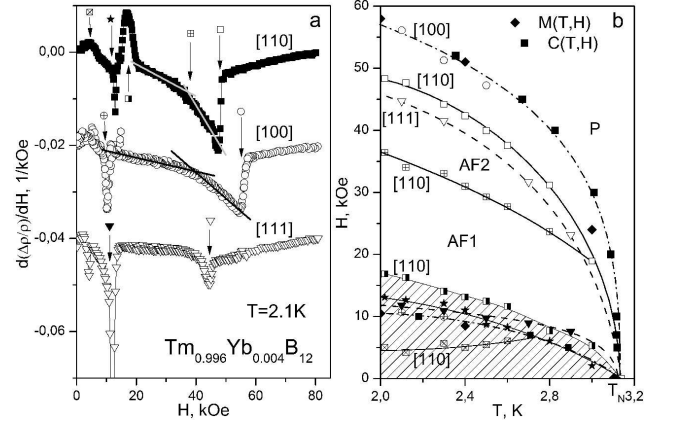


Fig. 3. (a) Derivatives of magnetoresistance  $d(\Delta\rho/\rho)/dH$  at  $T = 2.1$  K for three crystallographic directions (curves are shifted by 0.01 along vertical axis for convenience) and (b) magnetic  $H$ - $T$  phase diagram of  $Tm_{0.996}Yb_{0.004}B_{12}$ . AF1, AF2 are two AF phases, P denotes the paramagnetic phase. Points of phase transitions on panel (a) are shown with arrows and corresponding symbols, the MR hysteresis area is shaded.

#### 4. Conclusions

The magnetoresistance study of  $Tm_{0.996}Yb_{0.004}B_{12}$  in AF states and P state allowed to construct the magnetic  $H$ - $T$  phase diagram of this antiferromagnet. The anisotropy of both of the AF-P boundaries and the MR behavior at low ( $H < 20$  kOe) and intermediate (40–60 kOe) magnetic fields may be interpreted in terms of some additional magnetic interaction, which modifies the magnetic structure produced by RKKY indirect exchange.

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