

# Magnetic Phase Diagram of TmB<sub>4</sub> under High Pressure

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TmB<sub>4</sub> is a Shastry-Sutherland frustrated system which exhibits very complex magnetic properties. In this contribution the phase diagram of magnetic field vs. temperature of TmB<sub>4</sub> under hydrostatic pressure up to 26.5 kbar is investigated using sensitive ac-resistance measurements. Temperature and magnetic field dependences of resistance at various pressures were carried out in a piston cylinder pressure cell between 1.7 and 14 K and in magnetic fields up to 6 T. The obtained results exhibit shifts of ordering temperatures  $T_N$  as well as shifts of boundaries between different magnetic phases. The observed pressure dependences of  $T_N$  can be described by the relation  $d \ln T_N / dp = +(0.16 \div 0.18) \% / \text{kbar}$ . The effect of pressure on various interactions between magnetic ions in this compound is discussed.

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## 1. Introduction

Properties of spin systems with frustrated antiferromagnetic (AF) interactions have attracted widespread interest in the last years due to discoveries of various new types of complex quantum ground states. Among them one can mention e.g. the spin liquid dimer phase, observed in SrCu<sub>2</sub>(BO<sub>3</sub>)<sub>2</sub> [1] (a 2D magnet on the Shastry-Sutherland lattice (SSL)), or the spin ice state in pyrochlore [2]. The interest in such systems is, in particular, related with magnetization plateaus at fractional values of saturation magnetization, as observed in SrCu<sub>2</sub>(BO<sub>3</sub>)<sub>2</sub> [3], or with fractionalized excitations in spin ice.

Recently it was shown that rare earth tetraborides (REB<sub>4</sub>) represent other 2D frustrated magnets on the SSL which are relatively easy and fully accessible for experiments up to the saturation magnetic field [4–6]. REB<sub>4</sub> compounds crystallize in a tetragonal lattice, and their chemical and physical properties are very similar for RE = Ho, Er, Tm. . . For example, regarding their electronic structure, one of the three valence electrons of RE ions goes to the conduction band, and therefore all these compounds are good metals, and the RKKY interaction is thought to be the relevant exchange mechanism between magnetic moments. The RE ions arrangement in  $a - b$  planes of tetraborides maps a regular array of squares and triangles, which are almost equilateral, and thus maps exactly the SSL.

Probably the most studied among tetraborides, TmB<sub>4</sub>, exhibits a rich phase diagram as a function of temperature and field. In zero field the magnetic order, in which

the magnetic moments are oriented parallel with the  $c$  axis, sets in at  $T_N = 11.8$  K, the basic “AF low temperature Néel phase” becomes stable below 9.9 K. Based on magnetization experiments in magnetic field  $B$  oriented along the  $c$  axis, two other phases can be observed. The so called “high field” phase (above  $\sim 2$  T) is ferrimagnetic with a plateau at  $M/M_{sat} = \frac{1}{2}$ , where  $M_{sat}$  denotes the saturation magnetization. In the “intermediate phase”, magnetization plateaus with  $M/M_{sat} = 1/7, 1/8, 1/9 \dots$  are seen at temperatures below 4 K, and their observation is subject to hysteresis. Close to  $T_N$  the intermediate phase splits into more complex phases as identified in specific heat and resistivity measurements [5, 6]. TmB<sub>4</sub> neutron diffraction measurements point to various unique stripe structures of Tm magnetic moments [7]. Up to now a few models were proposed to describe the observed phenomena. In [7] it has been suggested that the observed plateaus and spatial structures may be described by a model based on fractional Quantum Hall Effect. Another model is based on the coexistence of two subsystems, namely the spin subsystem, described by the Ising model, and the electronic subsystem described by the Falicov-Kimball model on the SSL. Moreover, both subsystems are coupled by an anisotropic spin-dependent interaction of the Ising type [8]. A complete and exact solution of the ground-state problem for the Ising model on the SSL in an applied field was found recently in [9].

In order to shed more light onto this very complex problem, in this contribution the influence of hydrostatic pressure on TmB<sub>4</sub> is investigated and discussed.

## 2. Experimental details

Single crystal of TmB<sub>4</sub> was grown by an inductive, crucible-free zone melting method. The residual resistivity ratio of the samples was larger than 100, documenting their very high quality. Sensitive ac-resistance measure-

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ments under hydrostatic pressure up to 26.5 kbar were carried out in a piston cylinder pressure cell. The temperature and magnetic field dependences of resistance at various pressures were performed between 1.7 and 14 K and in magnetic fields up to 6 T.

### 3. Results and discussion

The obtained results of  $ac$ -resistivity  $\rho(T)$  as a function of temperature  $T$  is shown for selected pressure values in Fig. 1. It shows that under increasing hydrostatic pressure the shape of  $\rho(T)$  dependences and the positions of anomalies, which indicate transitions between various magnetic phases, slightly shift to higher temperatures. The observed pressure dependence of  $T_{N1}$  and  $T_{N2}$  in zero field can be described by relations  $d \ln T_{N1}/dp \approx +0.16 \text{ \%/kbar}$  and  $d \ln T_{N2}/dp \approx +0.18 \text{ \%/kbar}$  (see inset of Fig. 1). On the other hand, the zero-temperature critical field  $B_c(0)$  shifts at pressure of 26.5 kbar to higher  $B_c$  values by about  $\approx 100 \text{ mT}$ , providing thus a positive pressure change of  $dB_c/dp \approx +3.8 \text{ mT/kbar}$ .

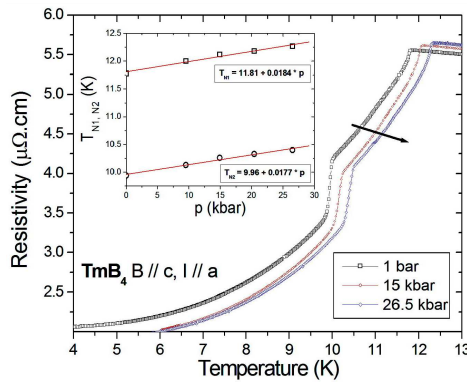


Fig. 1. Zero field resistivity of  $TmB_4$  for selected pressure values. The inset shows pressure dependences of Néel temperatures.

These results show that the  $B$  vs.  $T$  magnetic phase diagram (see Fig. 2 for 1 bar and 26.5 kbar) of  $TmB_4$  under pressure does not change its topology. Only the shifts of phase boundaries to higher temperature and magnetic field values are observed, which could be described also as an inflation of the magnetic phase diagram. This points to the fact that hydrostatic pressure does not change the basic essence of this complex anisotropic frustrated system. High pressure just seems to lead to an increase of the exchange interaction between magnetic RE ions due to the reduction of lattice constants under its influence. This causes consequently the truncation of interaction paths and leads to the fact that magnetic order sets in at higher temperatures and persists in higher magnetic field. Therefore, as  $TmB_4$  has a tetragonal lattice which exhibits a SSL arrangement in the  $a - b$  plane with magnetic moments oriented parallel to the  $c$  axis, in order to be able to distinguish between the roles of interaction paths in various directions (along the  $c$  axis or along the

axes in the  $a - b$  plane), further investigations, above all under high uniaxial pressure (e.g. along the  $c$  axis), are needed.

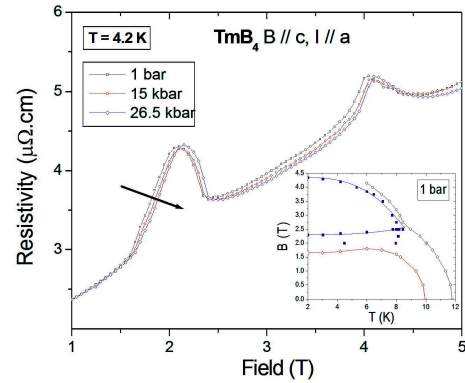


Fig. 2. Magnetic phase diagram of  $TmB_4$  for 1 bar (black lines) and 26.5 kbar (red lines).

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