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Phase Diagram of TmB_4 Probed by AC Calorimetry

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TmB_4 is a frustrated system based on the Shastry–Sutherland lattice that exhibits complex magnetic properties. In this contribution the magnetic field B vs. temperature T phase diagram of TmB_4 has been studied by ultrasensitive AC calorimetry in the temperature range between 2.9 and 12 K and in magnetic fields up to 8 T. Apart from already known phases our measurements have recognized several new phase transitions suggesting that the phase diagram of TmB_4 is even more complex and deserves further studies.

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1. Introduction

The problem of 2D geometric frustrated Shastry–Sutherland lattice (SSL) attracted considerable attention in recent years, above all due to observation of magnetization plateaus at fractional values of saturation magnetization [1]. It was surprising that tetragonal TmB_4 revealed such properties, and neutron diffraction measurements pointed out to various stripe structures of Tm magnetic moments [1]. The developed theoretical model [2] suggests that TmB_4 is related to a magnetic analogy of the fractional quantum Hall effect. Therefore further studies of TmB_4 and other tetraborides are important.

In this paper the phase diagram of TmB_4 is addressed via the specific heat measurements in the temperature range between 2.9 and 12 K and in magnetic fields up to 8 T. The temperature and magnetic field dependences of the specific heat consistently indicate the existence of different phases in the system below its Néel temperature $T_{\mathrm{N}} = 11.2$ K. While already known phases are approved also new transitions are indicated, requiring further studies.

2. Experimental

A large single crystal of TmB_4 was prepared by a floating zone melting technique. The c-axis and a-axis of the crystal were determined by the Laue diffractions. Specific heat measurements down to 2.9 K and up to 8 T, $C(T, B)$, were made by AC calorimetry technique as described elsewhere [3]. The basics of the method consist of applying a periodically modulated power and measuring the resulting temperature oscillations of the sample. In a proper frequency regime, the heat capacity of the sample is inversely proportional to the amplitude of the temperature oscillations. Although AC calorimetry is not capable to measure the absolute values of heat capacity it is a very sensitive technique for measurements of relative changes on very small samples. Also it enables continuous measurements during either temperature or magnetic field sweeps (changing temperature while keeping the magnetic field constant and vice versa) and thus it is suitable to detect phase transitions in detail.

All measurements were performed in magnetic field parallel to the c-axis of the crystal. Temperature oscillations of the sample were recorded by a sensitive thermocouple. Corrections of the Cernox thermometer and of the thermocouple in magnetic field were carefully inspected and included in the data treatment.

3. Results

In Fig. 1 we present magnetic field sweeps of the specific heat of TmB_4 at selected temperatures. All curves are non-monotonic and reveal several maxima. We attribute these maxima to phase transitions of unknown character that occur in this material. To get a general picture of how the phases evolve with magnetic field and temperature we marked each maximum as a point in the phase diagram.

Figure 2 summarizes the obtained values of the phase transitions derived not only from the field sweep measurements shown in Fig. 1, but also from temperature sweeps measurements (not shown here). We used open symbols for the results from the temperature sweeps and closed

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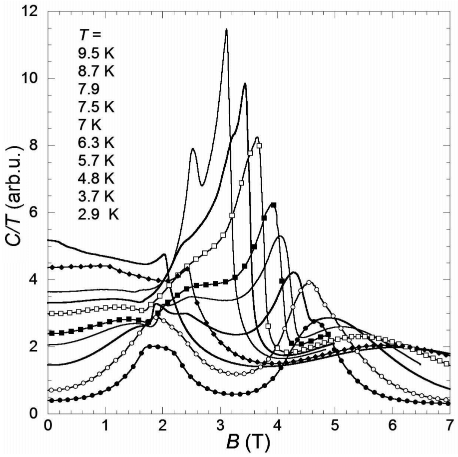

Fig. 1. Field dependences of specific heat of TmB_4 at different temperatures for $B \parallel c$. The lowest curve is the one measured at 2.9 K.

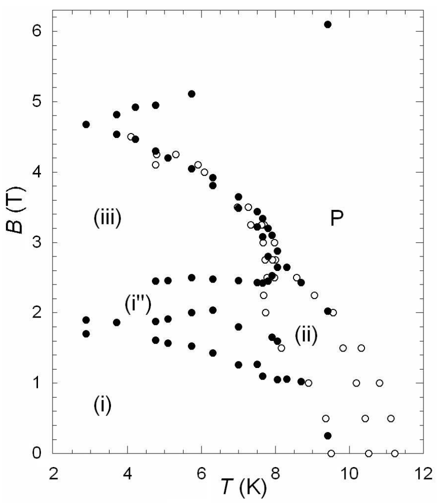

Fig. 2. Magnetic field vs. temperature phase diagram of TmB_4 for $B \parallel c$. Phases: P — paramagnetic, (i) — intermediate, (i″) — plateau regime, (ii) — antiferromagnetic, (iii) — ferrimagnetic. Open symbols — temperature sweeps, full symbols — field sweeps.

symbols for the ones from the field sweeps for clarity. Let us note that in certain parts of the phase diagram points from both kinds of measurement show a very good overlap confirming the consistency of received results. On the other hand, there are some parts of the diagram where phase transition were detectable only in one way, especially when a line in the diagram is close to being parallel with the abscissa or the ordinate.

The obtained phase diagram (Fig. 2) confirms the existence of major phases and is in agreement with those observed from other measurements. For example, in [4, 5] based on magnetization experiments, at low temperatures several phases were observed: a "high" field ferrimagnetic phase (iii), an intermediate phase with magnetization plateaus (i) and the "low" field antiferromagnetic phase (ii).

Results based on AC calorimetry measurements indicate that in zero field, magnetic order sets in at T_N = 11.2 K, and the transition from paramagnetic state is gradually shifted towards lower temperatures with increasing magnetic field. The antiferromagnetic low temperature Néel phase is stable below 9.5 K. Moreover several additional transitions *appear*. For example, the line in the diagram close to the paramagnetic (P) phase (between approximately 2.5 and 4.5 T) splits into two. This splitting is consistently observed on the both field sweep and temperature sweep measurements. Also the intermediate phase splits into more complex phases. Finally probably a new phase is detected at high magnetic fields above around 4.6 T. The nature and the properties of the observed phases call for further studies of this and related compounds.

4. Conclusions

A complex magnetic phase diagram of TmB_4 was obtained from the AC specific heat measurements. Several new phase transitions were observed. The nature of the magnetic order in separate parts of the diagram demands further studies.

Acknowledgments

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