Anomalies of Heat Capacity and Phase Transitions in Tm$_{1-x}$Yb$_x$B$_{12}$

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Quantum criticality and the associated magnetic quantum phases of heavy fermion metals on one side and the metal–insulator transition (MIT) in strongly interacting electron systems on other side are of extensive current interest [1, 2]. Moreover, the picture of a magnetic metal transformation into paramagnetic insulator state has been subjected to experimental testing in recent years. In particular, it was found very recently [3] that in the family of solid solutions Tm$_{1-x}$Yb$_x$B$_{12}$ the substitution of Tm by Yb causes a transition from antiferromagnetic metal (AF) TmB$_{12}$ ($T_N \approx 3.2$ K) through the quantum critical point (QCP) with $T_C \approx 0$ at $x_C \approx 0.3$ to the so-called Kondo insulator state in YbB$_{12}$. Additionally, the decreased dimension of the magnetic excitation spectrum was revealed both in the case of YbB$_{12}$ narrow-gap semiconductor [4] and in the well-known and most extensively studied system with the AF-type QCP — CeCu$_{5.9}$Au$_{0.1}$ [5]. In analogy with the CeCu$_{6-x}$Au$_x$ system, it is interesting to investigate in detail the behavior of specific heat $C_p(T)$ for Tm$_{1-x}$Yb$_x$B$_{12}$ compounds in a wide vicinity of AF near QCP $x_C \approx 0.3$ and within MIT. Since the magnetic field is a crucial parameter in the quantum critical region [1, 5], it seems reasonable also to perform $C_p(T)$ measurements in high magnetic fields.

2. Experimental

In this work the behavior of specific heat of Tm$_{1-x}$Yb$_x$B$_{12}$ solid solutions has been studied on high quality single crystals with $0 \leq x \leq 0.8$ within the temperature range of 1.9–300 K in magnetic field up to 9 T in a PPMS-9 (Quantum Design). For comparison the non-magnetic reference compound LuB$_{12}$ was also investigated in present study. The RB$_{12}$ single crystals were grown by vertical crucible-free induction zone melting in an inert gas atmosphere [6].

3. Results and discussion

Figures 1 and 2 show the temperature dependences of specific heat for the antiferromagnet TmB$_{12}$, Tm$_{0.60}$Yb$_{0.31}$B$_{12}$ compound in vicinity of QCP, non-magnetic reference dodecaboride LuB$_{12}$ (Fig. 1), and for paramagnetic solid solutions Tm$_{1-x}$Yb$_x$B$_{12}$ with $x = 0.23$, 0.37 and 0.72 (Fig. 2). The heat capacity of LuB$_{12}$ was measured to estimate the Sommerfeld $C_S(T)$ and lattice $C_{ph}(T)$ components in the specific heat of Tm$_{1-x}$Yb$_x$B$_{12}$ and to extract the magnetic contribution $C_{mag}(T) = C_p(T) - C_S(T) - C_{ph}(T)$. Figure 3 presents, for example, the temperature dependences $C_{mag}(T)$ of Tm$_{0.74}$Yb$_{0.26}$B$_{12}$ obtained at different values of external magnetic field $\mu_0H = 0$, 3, 6 and 9 T. The data presentation $C_{mag}/T = f(lnT)$ allows us to conclude in favor of the logarithmic divergence of the renormalized Sommerfeld coefficient at low temperatures, which is typical for systems at QCP [7], and it is attributed usually to the dramatic renormalization of the quasiparticles’ effective mass and to the issue of a non-Fermi-liquid behavior of heat capacity.

It should be noted also that the magnetic field $\mu_0H \geq 3$ T suppresses completely the quantum critical regime (Fig. 3) and that at low temperatures the $C_{mag}(T)$ curves demonstrate the Schottky anomaly of specific heat. The
Fig. 1. Dependences of specific heat vs. temperature for dodecaborides \( \text{Tm}_{1-x}\text{Yb}_x\text{B}_{12} \) with \( x = 0 \) and 0.31 and for non-magnetic reference dodecaboride \( \text{LuB}_{12} \). The inset presents the \( T-x \) phase diagram.

Fig. 2. Dependences of specific heat vs. temperature for dodecaborides \( \text{Tm}_{1-x}\text{Yb}_x\text{B}_{12} \) with \( x = 0.23, 0.37 \) and 0.72 at \( H = 0 \) and 9 T. In the inset the scheme of crystal field splitting of the \( \text{Tm}^{3+} 3H_6 \) state is presented.

Fig. 3. Temperature dependences of the magnetic contribution \( C_{\text{mag}}(T) \) to specific heat of \( \text{Tm}_{0.71}\text{Yb}_{0.29}\text{B}_{12} \) in external magnetic field up to 9 T. Inset shows the Zeeman splitting (\( \Delta_{01}, \Delta_{02} \)) of the \( I_5^{(1)} \) triplet of the \( \text{Tm}^{3+} 3H_6 \) state in magnetic field.

in the formation of features of the thermal properties in the \( \text{Tm}_{1-x}\text{Yb}_x\text{B}_{12} \) strongly correlated electron system.

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References