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Anomalies of Heat Capacity and Phase Transitions in $Tm_{1-x}Yb_xB_{12}$

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In the system $Tm_{1-x}Yb_xB_{12}$ the specific heat has been studied in a wide range of Yb-concentration in the vicinity of the quantum critical point $x_C \approx 0.3$. The results were obtained on high quality single crystalline samples of $Tm_{0.7}Yb_{0.3}B_{12}$ compound placed near quantum critical point, both for antiferromagnetic metals ($x < x_C$) as well as for paramagnetic insulators ($x > x_C$) within a wide temperature range of 1.9–300 K in magnetic field up to 9 T. The temperature dependence of the magnetic contribution to specific heat for $Tm_{0.74}Yb_{0.26}B_{12}$ shows a logarithmic divergence of the form $C_{mag}/T \sim -\ln T$ at $T < 4$ K, which may be attributed to the quantum critical regime, and it is suppressed by strong external magnetic field. The Schottky anomaly of the magnetic contribution to specific heat in $Tm_{1-x}Yb_xB_{12}$ has been established and analyzed in detail.

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1. Introduction

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 $Tm_{1-x}Yb_xB_{12}$ 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 $T_N \approx 0$ 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 $Tm_{1-x}Yb_xB_{12}$ 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 $Tm_{1-x}Yb_xB_{12}$ solid solutions has been studied on high quality single crystals with $0 \le x \le 0.8$ 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}, $Tm_{0.69}Yb_{0.31}B_{12}$ compound in vicinity of QCP, non--magnetic reference dodecaboride LuB_{12} (Fig. 1), and for paramagnetic solid solutions $Tm_{1-x}Yb_xB_{12}$ with $x =$ 0.23, 0.37 and 0.72 (Fig. 2). The heat capacity of LuB_{12} was measured to estimate the Sommerfeld $C_e(T)$ and lattice $C_{ph}(T)$ components in the specific heat of $Tm_{1-x}Yb_xB_{12}$ and to extract the magnetic contribution $C_{mag}(T) = C_p(T) - C_e(T) - C_{ph}(T)$. 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 $\mu_0 H = 0$, 3, 6 and 9 T. The data presentation $C_{mag}/T = f(\ln T)$ 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 $\mu_0 H \ge$ 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

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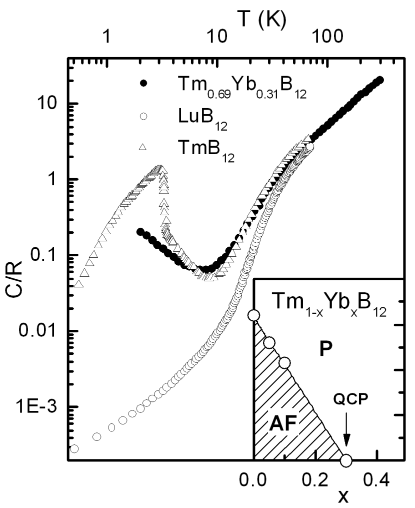

Fig. 1. Dependences of specific heat vs. temperature for dodecaborides $Tm_{1-x}Yb_xB_{12}$ with $x = 0$ and 0.31 and for non-magnetic reference dodecaboride LuB_{12}. The inset presents the T–x phase diagram.

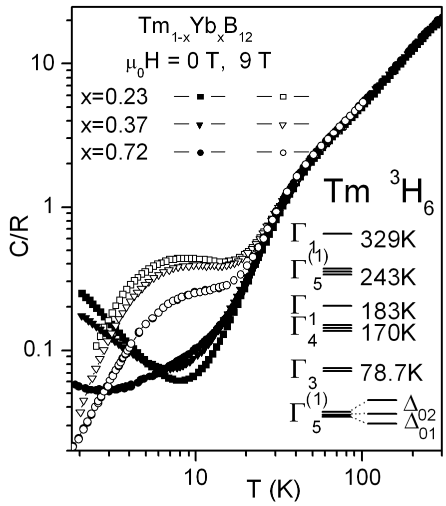

Fig. 2. Dependences of specific heat vs. temperature for dodecaborides $Tm_{1-x}Yb_xB_{12}$ with $x = 0.23$, 0.37 and 0.72 at $H = 0$ and 9 T. In the inset the scheme of crystal field splitting of the Tm^{3+} $^3H_6$ state is presented.

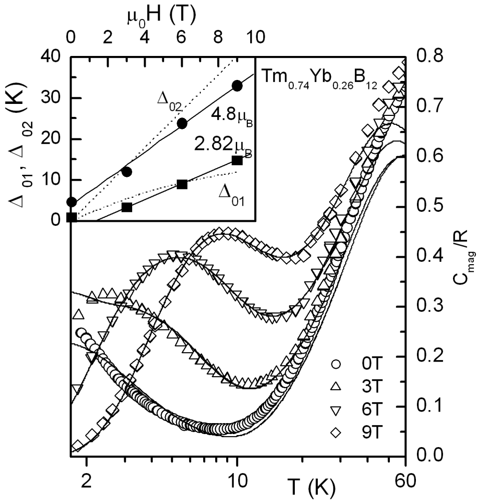

Fig. 3. Temperature dependences of the magnetic contribution $C_{\mathrm{mag}}(T)$ to specific heat of $Tm_{0.74}Yb_{0.26}B_{12}$ in external magnetic field up to 9 T. Inset shows the Zeeman splitting (Δ_{01}, Δ_{02}) of the $\Gamma_5^{(1)}$ triplet of the Tm^{3+} $^3H_6$ state in magnetic field.

detailed numerical analysis of the crystal electric field and the Zeeman splitting features of the magnetic contribution allows us both to confirm the scheme of the Tm^{3+} $^3H_6$ state splitting (inset in Fig. 2), and to estimate the effective magnetic moments ($\mu_{\mathrm{eff}} \approx 2.82\ \mu_{\mathrm{B}}$ and $4.8\ \mu_{\mathrm{B}}$, correspondingly) for the states Δ_{01} and Δ_{02} of the $\Gamma_5^{(1)}$-triplet of Tm^{3+} (see inset in Fig. 3). The results of $C_{\mathrm{mag}}(T)$ and entropy studies indicate that the Tm subsystem should be considered as a decisive factor in the formation of features of the thermal properties in the $Tm_{1-x}Yb_xB_{12}$ strongly correlated electron system.

Acknowledgments

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