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Ground-state crossover in $U_{1-x}Th_xBe_{13}$ ($0 \leqslant x \leqslant 0.15$)

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Abstract. By studying the temperature dependences of the electrical resistivity, magnetoresistance, nonlinear susceptibility, Hall effect and heat capacity of $U_{1-x}Th_xBe_{13}$ with $0 \leqslant x \leqslant 0.15$ we have observed a crossover between two different (magnetic and nonmagnetic) ground states which indicates the possible presence of a quantum critical point near $x \approx 0.05$.

Despite numerous experimental and theoretical efforts, the fascinating normal and superconducting properties of UBe_{13} [1] and $U_{1-x}Th_xBe_{13}$ [2] still remain unresolved. To overcome the apparent inconsistencies between experiment and the classic magnetic one-channel Kondo model, a nontrivial single-impurity two-channel Kondo (TCK) or quadrupolar Kondo-effect (QKE) model was proposed [3]. It was also found that single-ion physics could be applied to $U_{1-x}Th_xBe_{13}$ between the dilute and concentrated ($x \geqslant 0.1$) U-atom limits [4–6]. Recent experimental studies of the properties of $U_{0.9}Th_{0.1}Be_{13}$ have demonstrated that its ground state below 5–6 K is reminiscent of the one expected for the QKE model [7]. A heavy-fermion state induced by QKE fluctuations was also reported for Pr-based intermetallic compounds [8]; as for UBe_{13}, the main deviations from the QKE model are the large negative magnetoresistance [9, 10] and the nonlinear susceptibility [11], both with anomalous behaviour at low temperatures. However, a recent Monte Carlo calculation [12] has confirmed that UBe_{13} is in a mixed-valence (MV) regime [13].

In this paper, we present the results of our studies of four $U_{1-x}Th_xBe_{13}$ samples with Th content in the interval $0 \leqslant x \leqslant 0.15$. We have found that substitution of Th for U up to 5%, in agreement with reference [14], removes the small energy scale, T_{SF}, which may be related to the presence of magnetic spin fluctuations. In the region $x \geqslant 0.1$ where the nonmagnetic heavy-fermion-type ground state is formed, our results are consistent with the QKE interpretation. As indicated by the QKE scaling analysis, the new ground state may be characterized by a nonmagnetic small energy scale, T_Q, which increases with Th content. The T–x phase diagram indicates the possible presence of a quantum critical point near $x \approx 0.05$.

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The polycrystalline samples were prepared by arc melting of stoichiometric amounts of the elemental constituents in an argon atmosphere, with a slight excess of Be to compensate for losses. Uranium was used in the form of pellets after being cleaned in nitric acid and the Th was premelted before use. All samples were remelted three times to ensure good homogeneity. X-ray powder diffraction analysis proved that our samples are single phase with cubic $NaZn_{13}$-type structure. The lattice parameter a was found to correspond well to the initial linear increase with Th content as previously reported [2]. The magnetoresistance and Hall effect were measured in an Oxford magnet by using a conventional dc method. The specific heat measurements were carried out in a ^{3}He cryostat using a standard heat pulse technique. The nonlinear term χ_3 in the magnetization M along the direction of the magnetic field H ($M = \chi_1 H + (1/3!)\chi_3 H^3 + \cdots$) was determined by using a *Quantum Design* SQUID magnetometer in the way described previously [13].

Figure 1 compares the temperature dependences for $x = 0, 0.05$ and 0.15 of the normalized

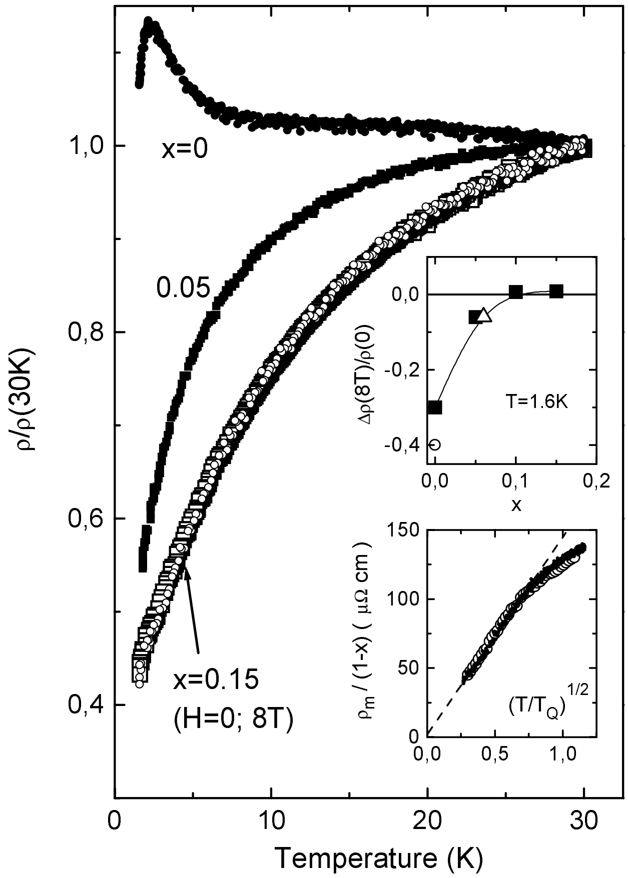

Figure 1. Temperature dependences of the normalized resistivity $\rho(T)/\rho(30\text{ K})$ for different x. The data for $x = 0.15$ show the measurements in zero field (open squares) and in the applied magnetic field of 8 T (open circles). The upper inset shows the magnetoresistance $\Delta\rho(8\text{ T})/\rho(0)$ against x. The open points correspond to the data for $x = 0$ (from reference [10]) and $x = 0.06$ (from reference [14]). The lower inset shows a scaling of the temperature-dependent part of the resistivity ρ_m with the U concentration and with the characteristic temperature T_Q, for samples with $x = 0.1$ (open circles) and $x = 0.15$ (closed circles).

resistivity $\rho(T)/\rho$ (30 K). The disappearance of the low-temperature maximum for a small Th percentage, in agreement with a previous report [15], is followed by a transition to a novel ground state at $x > 0.05$. The upper inset in figure 1 shows the dependence of the magnetoresistance $\Delta\rho(8\ \mathrm{T})/\rho(0)$, measured at $T = 1.6$ K, on x. For samples with $x = 0.1$ and 0.15, $\rho(T)$ follows a $\sqrt{T}$ law in the temperature ranges 0.8–6 K and 1.5–12 K, while at lower temperatures it varies linearly with temperature down to 0.08 K [16] and 0.3 K respectively.

The electronic specific heat, C_e, for all samples is clearly enhanced at low temperatures, as is depicted in figure 2. Below 10 K, $C_e/T(x = 0.05)$ increases as $T^{-0.75}$. For $x \geqslant 0.1$, C_e varies approximately as $T \log T$. This logarithmic dependence is almost unaffected by a magnetic field of 8 T.

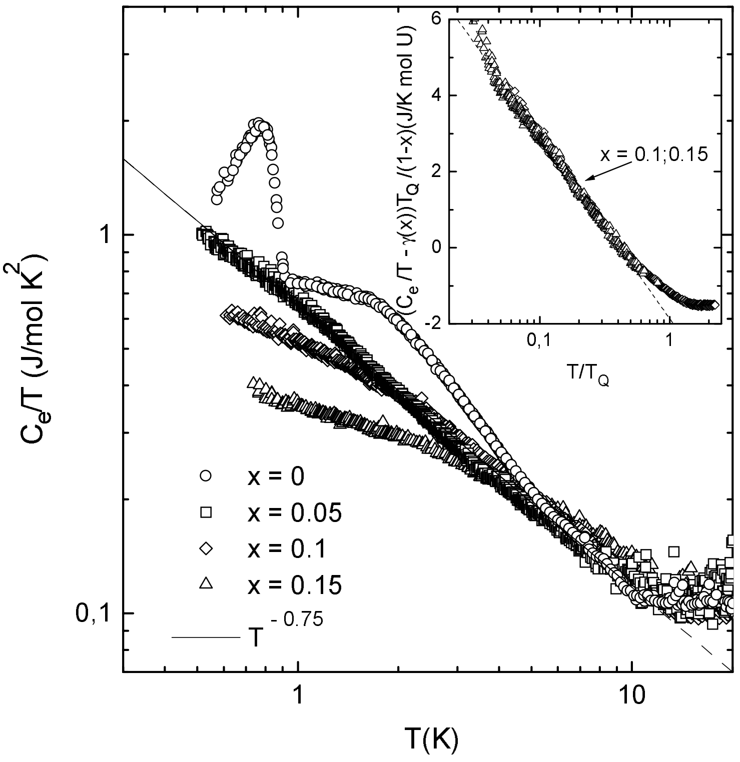

Figure 2. Temperature dependences of the electronic specific heat normalized by the temperature for $U_{1-x}Th_xBe_{13}$. The inset shows the scaling of C_e with the U concentration and T_Q for the samples with $x = 0.1$ (◇) and $x = 0.15$ (△) ($\gamma(x)T$ is the electronic contribution of the host to C_e).

Next, we monitor the transformation of the ground state by means of the Hall effect. For $x = 0$ the data (only the low-field $\rho_{xy}(H)$ measurements are shown in figure 3(a)) are similar to those reported before [17], with a maximum in the anomalous Hall effect (AHE) near $T = T_{\mathrm{max}} \approx 2.5$ K, corresponding to a low-temperature maximum in $\rho(T)$. At $T < T_{\mathrm{max}}$, the field-induced AHE results in a linear dependence $\rho_{xy}(H) \sim H$ observed below a few teslas and followed by a maximum at $H = H_{\mathrm{max}}$. As uranium is substituted for with thorium, the low-temperature behaviour $\rho_{xy}(H)$ is dramatically changed (see figure 3(a)). For $x = 0.05$ the anomalous Hall effect is observed only for sufficiently low magnetic fields, $H < 0.5$ T, and

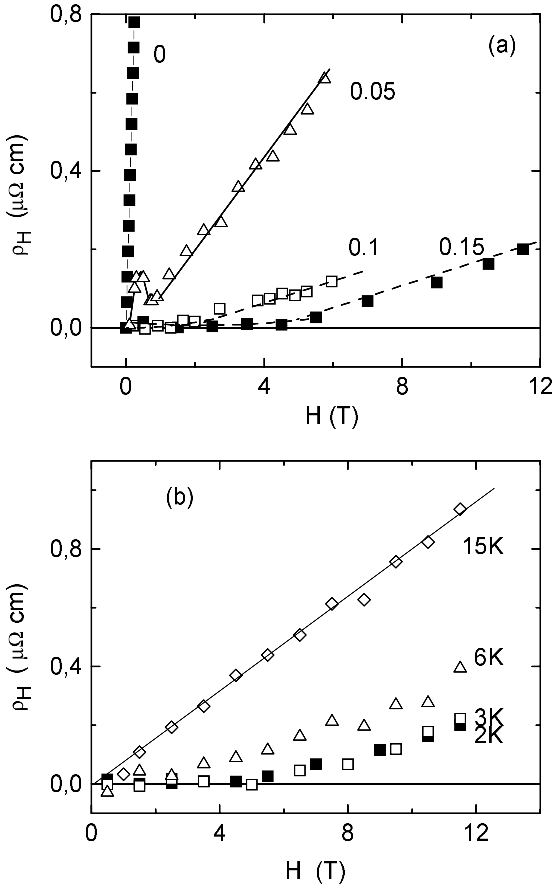

Figure 3. (a) Magnetic field dependences of the Hall resistivity for $U_{1-x}Th_xBe_{13}$ with different Th concentrations measured at $T = 2$ K. (b) Magnetic field dependences of the Hall resistivity of $U_{0.85}Th_{0.15}Be_{13}$ at different temperatures.

the value $H_{\max}$ decreases by about an order of magnitude in comparison to that for UBe_{13} [17]. A further increase in the thorium concentration qualitatively changes $\rho_{xy}(H)$. In the limit of small fields, for $x \geqslant 0.1$, a linear term in the Hall resistivity ρ_{xy}/H vanishes asymptotically and the nonzero Hall coefficient recovers only at higher magnetic fields and temperatures. This unusual behaviour is an important feature which identifies the QKE ground state [18]. The transformation of the field dependences of the Hall resistivity for $U_{0.85}Th_{0.15}Be_{13}$ with temperature is shown in figure 3(b).

In order to obtain an estimate of the magnetic energy scale from the Hall-effect data, we relate the low-temperature maxima in $\rho_{xy}(H)$ to the suppression of the spin fluctuations by the magnetic field and get $T_{\mathrm{SF}} \sim \mu_B H_{\max}/k_B \approx 2.5$ K for UBe_{13} and $T_{\mathrm{SF}} \sim 0.3$ K for $U_{0.95}Th_{0.05}Be_{13}$. We note that for $x = 0$ this rough estimate is close to the one obtained previously from the specific heat data ($T_{\mathrm{SF}} \sim 2$ K) [14].

Figure 4, which shows the temperature dependences of the nonlinear susceptibility $\chi_3(T)$ for $0 \leqslant x \leqslant 0.15$, confirms the transformation of the ground state at $x \geqslant 0.1$. For these Th contents, the nonlinear susceptibility does not show a downturn to large negative values as temperature decreases, indicating the nonmagnetic nature of the ground state.

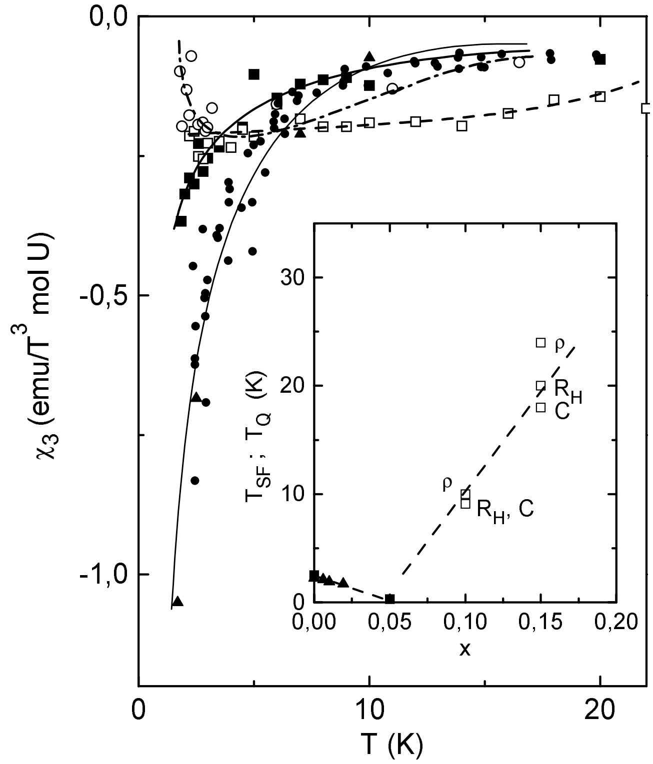

Figure 4. Temperature dependences of the nonlinear susceptibility of $U_{1-x}Th_xBe_{13}$. Closed circles and triangles correspond to the UBe_{13} data reported by Ramirez *et al* [11] and to our measurements. The closed squares, open circles and open squares are the data for $x = 0.05$, 0.1 and 0.15 respectively. The lines are guides to the eye. The inset shows the phase diagram of the ground-state properties of $U_{1-x}Th_xBe_{13}$ ($0 \leqslant x \leqslant 0.15$). Closed triangles and closed squares represent respectively the characteristic magnetic scale $T_{SF}(x)$ estimated from the specific heat (reference [14]) and from the Hall effect (reference [17] and this work). The open squares represent the nonmagnetic scale $T_Q(x)$. The signs near the data explain which experiments have been used to obtain T_Q.

Although evidence for the ground-state change between UBe_{13} and $U_{0.9}Th_{0.1}Be_{13}$ has been provided before [7], our new results obtained for four different $U_{1-x}Th_xBe_{13}$ compositions allow one to study in more detail the spin-fluctuation–nonmagnetic ground-state crossover with the aim of estimating the characteristic energies of the corresponding ground states. Let us start from the interval $x \leqslant 0.05$. The inset in figure 4 represents the energy scale evaluated above: the magnetic T_{SF} as a function of the Th content, for $U_{1-x}Th_xBe_{13}$. Closed triangles and closed squares represent respectively T_{SF} estimated from the Schottky-type anomaly in the specific heat [14, 19] and from the Hall effect (reference [17] and this work). A recent revision of the $U_{1-x}Th_xBe_{13}$ phase diagram [20] proposes that these magnetic fluctuations give rise to a true long-range magnetic order for $0.02 < x < 0.04$ with magnetic moment $\mu_S \sim 10^{-3}\ \mu_B$ at $T < 0.5$ K.

The weak dependence on applied magnetic field of the electron transport and thermodynamics at $x \geqslant 0.1$ is consistent with a nonmagnetic ground state formed by electron interaction with 'pseudospins' of nonmagnetic origin. As far as we know, the only model which describes a *nonmagnetic* heavy-fermion ground state which is insensitive to the magnetic field is the QKE theory [3]. Although the applicability of this single-ion model should be questioned for the concentrated U-atom case, we believe that this theory adequately describes our low-temperature experimental data for $x \geqslant 0.1$ in some extended low-temperature interval—similarly to how the single-ion Kondo model applies to concentrated Kondo systems above some temperature where inter-ion coherent effects become unimportant.

Scaling the temperature-dependent part of the resistivity, $\rho_m(T) = \rho - \rho_0$ (see the lower inset in figure 1), to the QKE scattering, which is supposed to be dominant at temperatures where $\sqrt{T}$-dependence holds [21], gives $T_Q(x = 0.15) \approx 22$ K for $T_Q(x = 0.1) \approx 10$ K as estimated before [22]. On the other hand, in the temperature interval where single-ion QKE scattering processes are effective, the law

$$C_e/T \propto -\left(\frac{(1-x)}{T_Q}\right)\log\left(\frac{T}{T_Q}\right)$$

is followed [23]. If we fit C_e to this relation using the numerical coefficients from reference [23], we obtain $T_Q(x = 0.1) \approx 9$ K and $T_Q(x = 0.15) \approx 18$ K. The corresponding data scaling is shown in the inset of figure 2. Finally, we relate the appearance of nonzero Hall resistivity to the crossover from a non-Fermi liquid (NFL) to a FL induced by the magnetic field. This crossover was predicted to be described by the relation $T_S \sim H_S^2/T_Q$ [24] with T_S and H_S being respectively the temperature and the field which characterize the NFL–FL transition. We estimate from Hall-effect measurements $T_Q(x = 0.15) \sim 20$ K and $T_Q(x = 0.1) \sim 9$ K. We note that quantitative scaling of the $\chi_3(x, T)$ data is complicated because they were obtained for polycrystals, while the calculations [12] were carried out for the magnetic field directed along the (100) direction. Moreover, the experimental χ_3-data were obtained from finite-magnetic-field analyses, $2\ \mathrm{T} < H < 4\ \mathrm{T}$ (see also references [11]), while the theory [12] has been constructed in the zero-field limit.

The inset in figure 4 represents the energy scales, discussed above, for $U_{1-x}Th_xBe_{13}$ ($0 \leqslant x \leqslant 0.15$). We believe that the diagram obtained here, especially for $x \geqslant 0.1$, is rather qualitative, mainly because in our scaling we proposed 100% ground-state weight and $5f^2$ configuration, an assumption which may lead to overestimation in a mixed-valence regime [12]. Near the critical region ($x_c \approx 0.05$), the characteristic energy of magnetic spin fluctuations is mostly damped and a quantum phase transition (QPT) could appear. The physical picture near $x = 0.05$ could be described by a Griffiths-phase scenario recently proposed to describe the non-Fermi liquids near magnetic–paramagnetic quantum phase transitions in the presence of disorder [25, 26]. This theory predicts a power-law variation of the electron specific heat with temperature, $C_e/T \sim T^{-1+\alpha}$ ($\alpha < 1$). Our heat capacity data for $x = 0.05$ are indeed satisfactorily described by the above relation with $\alpha \approx 0.25$.

In our view, the mixed-valence model [12, 13] may account for the transformation of the ground state of $U_{1-x}Th_xBe_{13}$ ($0 \leqslant x \leqslant 0.15$). This approach proposes the existence of the degeneracy of $5f^2$ and $5f^3$ (or $5f^1$) configurations of the U atom resulting in the valence and corresponding spin ($0 \longleftrightarrow \frac{1}{2}$) fluctuations. The coexistence of the small energy scale and valence fluctuations is understood by taking into account the excited energy levels [27]. Interestingly, the problem has two different stable fixed points depending on whether the crystal-electric-field (CEF) splitting Δ is above or below the critical value Δ_c. For the region $\Delta > \Delta_c$ the ground state is described by the two-channel Kondo model characterized by the small energy scale T^* strongly varying with Δ near Δ_c: $T^* \sim (\Delta - \Delta_c)^{7/2}$. Although for $\Delta < \Delta_c$ the low-lying excitations are different from those predicted for the TCK model, the authors anticipate the presence of another small energy crossover scale characterized, however, by a different exponent. If the ground state of UBe_{13} is close to the MV regime [12, 13, 27], with $\Delta < \Delta_c$, the following scenario could explain the experimental data. Substitution of Th for U increases the CEF splitting of the 5f^2 level, resulting in a transition between different ground states with $\Delta < \Delta_c$ and $\Delta > \Delta_c$ each characterized by small energy scales of different origin. Due to the difference in symmetry of the states involved, the above hypothesis implies a novel type of quantum critical point [28] to appear exactly when $\Delta = \Delta_c$. In order to further prove the validity of the QKE picture at $x \geqslant 0.1$, studies on $U_{1-x}Th_xBe_{13}$ single crystals are needed. Such an investigation would allow adequate comparison with theory of the nonlinear susceptibility measured to sufficiently low temperatures.

Acknowledgments

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