Pressure Effects and Non-Linear Susceptibility in $U_{1-x}Th_xBe_{13}$

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The cubic heavy-fermion superconductor UBe_{13} [1] and its thoriated variant $U_{1-x}Th_xBe_{13}$ [2] belong to the most complex and fascinating systems containing strongly correlated electrons. Owing to simple power-law dependences discovered in, e.g., the specific heat [3] and the magnetic penetration depth [4] of pure UBe_{13}, a non s-wave, i.e., multi-component superconducting order parameter (OP), has been anticipated [5]. Hence, by lowering the crystal symmetry the degeneracy of the OP representation might be lifted: a splitting of ; 0.5 K/GPa was predicted if, under uniaxial pressure along the [100] direction, a tetragonal distortion is established.

Figure 1 displays results of specific-heat experiments performed [6] under uniaxial pressure, p, on a high-quality “H-type” UBe_{13} single crystal (T_c ; 0.93 K). The main effect of pressure is a linear p-derived depression of T_c, $|\ T_c/\ p|$ = (50±6) mK/GPa which, multiplied by 3, is close to the value reported for hydrostatic pressure (160 mK/GPa [7]). Also seen in Fig. 1 is a significant broadening of the transition at the peak of the anomaly. Nevertheless, a clear change in slope slightly above the peak temperature is resolved at the highest pressure values (inset of Fig. 1). This might be considered a possible onset of a splitting in the phase transition. However, the effect is 50 times smaller than predicted in [5]. Further investigations at higher pressures are necessary to find out if there exists a genuine split of T_c. A lack of splitting could imply: (i) the induced strain is still too small, (ii) the OP is single component rather than multi-component, (iii) depending on the details of the free energy forth-order cross-terms may pin the OP at the higher-temperature phase transition [5,8], preventing the other one even in case of a multi-component OP.

The resistivity *vs* temperature curve of normal-state UBe_{13} exhibits a characteristic maximum at T_{max} ; 2 K, which shifts to higher temperatures if hydrostatic pressure, p_h, is applied [9]. For $U_{1-x}Th_xBe_{13}$, T_{max} is found to strongly decrease with increasing p_h [10]. However, as shown for x = 0.0172, this decrease can be partially compensated by applying hydrostatic pressure, $p_h \geq 1.275$ GPa [9]. For an enhanced Th concentration (x = 0.034), however, no $\rho(T)$ maximum rather than an increase of $\rho(T)$ upon cooling was observed within the same temperature range, $T \geq$ 2K. The apparent lack of a $\rho(T)$ peak has led to speculations about the nearness of a “Kondo-insulator fixed point” [11]. To resolve this problem, we have studied $U_{1-x}Th_xBe_{13}$ samples with even higher Th concentration, i.e., x =

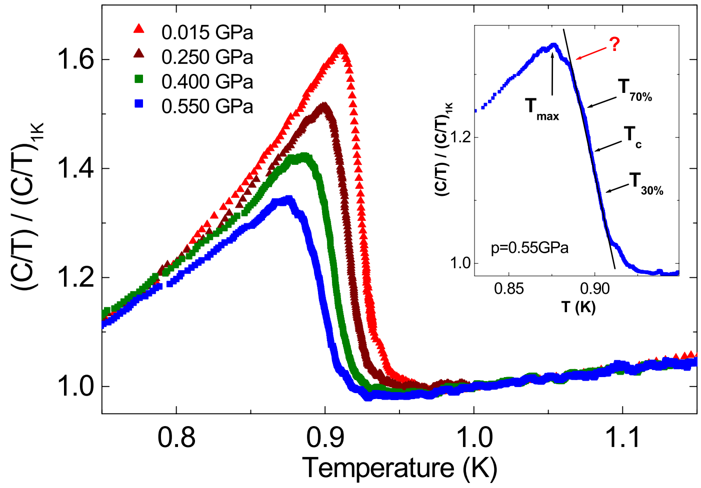

Fig. 1: Temperature dependence of the specific heat of a UBe_{13} single crystal as C/T vs T normalized to C/T at T = 1K under varying uniaxial pressures applied along the [100] direction. Inset shows a blow-up of the p=0.55 GPa data in the vicinity of the superconducting transition.

0.043 and 0.0465 [12]. As shown in Fig. 2a, a $\rho(\boldsymbol{T})$ maximum occurs in the former system for $\boldsymbol{p} \geq 2$ GPa below $\boldsymbol{T} = 1$ K and shifts upwards with increasing pressure. Taking into account the “offset pressures” to induce such a $\rho(\boldsymbol{T})$ maximum, one obtains a universal $\boldsymbol{p}_{\rm h}$-dependence for the pure compound and its two thoriated alloys (Fig. 2b). The ***effective*** negative pressure obtained this way is $\boldsymbol{p}_{\rm eff}$; -0.675 GPa/at% Th, in good agreement with $\boldsymbol{p}_{\rm eff}$; -0.7 GPa/at% Th found in [9].

Anomalies in the thermodynamic properties specific heat and thermal expansion phenomenologically related to the afore-discussed $\rho(\boldsymbol{T})$ maximum have been observed at ambient pressure for $U_{1-\boldsymbol{x}}Th_{\boldsymbol{x}}Be_{13}$ with $\boldsymbol{x} \leq 0.0455$, and were discussed in terms of an effective two-band model, consisting of localized and less localized $5\boldsymbol{f}$ states of the U^{3+} ($5\boldsymbol{f}^3$) configuration [13]. While there is experimental evidence for Uranium being almost trivalent in undoped UBe_{13} [14, 15], a gradual valence change due to the addition of Th was proposed by Aliev et al. [16]. According to their results of the non-linear susceptibility, $\chi^{(3)}(\boldsymbol{T})$, measurements, an almost tetravalent Uranium ($5\boldsymbol{f}^2$) configuration was inferred for $\boldsymbol{x} = 0.1$.

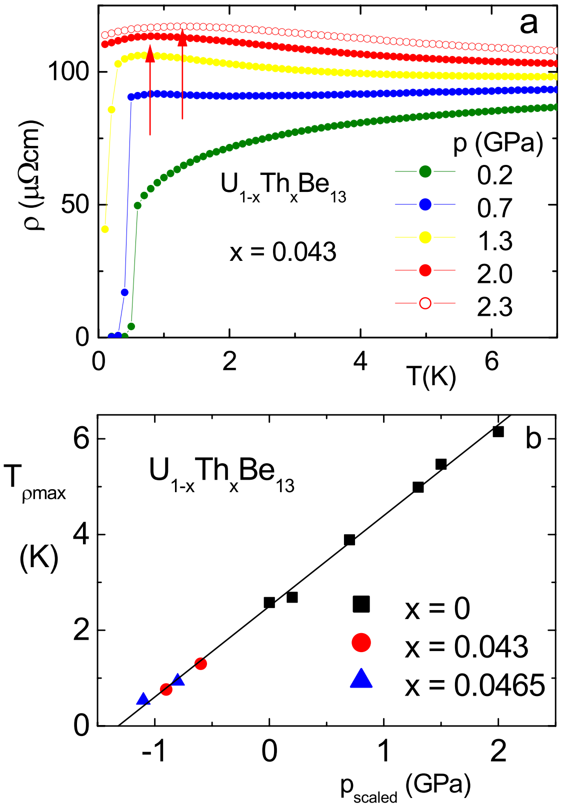

Fig. 2 a: Electrical resistivity $\rho(T)$ of $U_{0.957}Th_{0.043}Be_{13}$ at varying hydrostatic pressures. b: T_{max} as a function of scaled hydrostatic pressure [12].

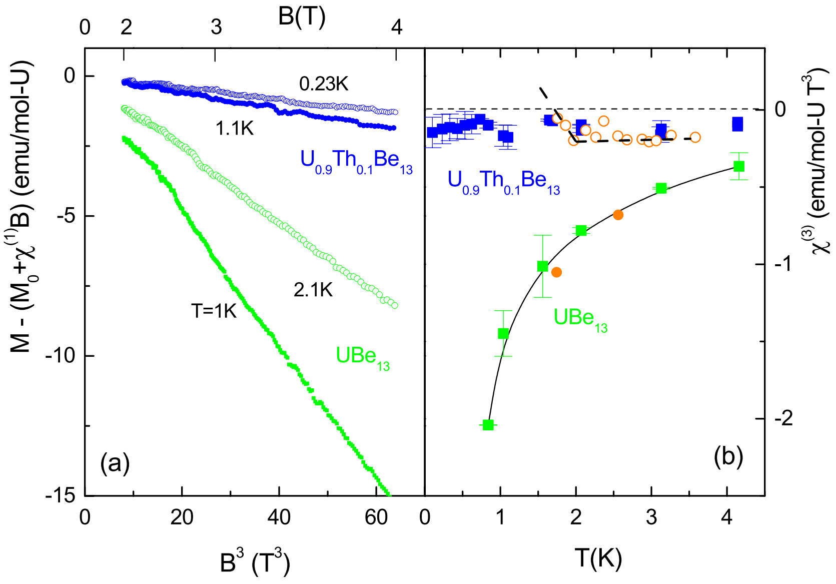

Fig. 3 a: Non-linear susceptibility, M-(M_0- $\chi^{(1)}B$) vs B^3 of single crystalline UBe_{13} and polycrystalline $U_{0.9}Th_{0.1}Be_{13}$. b: Temperature dependence of $\chi^{(3)}$ for UBe_{13} and $U_{0.9}Th_{0.1}Be_{13}$. Open symbols and broken line refer to Ref. 16.

As displayed in Figs. 3a and b, there is indeed a qualitative change in the ***T***-dependence of $\chi^{(3)}$ in going from $\boldsymbol{x} = 0$ to $\boldsymbol{x} = 0.1$, supporting [17] earlier conclusions that dipolar fluctuations are dominating in the pure compound [18], while quadrupolar ones are dominating in $U_{0.9}Th_{0.1}Be_{13}$ [16]. However, the ***T-independent*** $\chi^{(3)}$ of the latter alloy may well be ascribed to an intermediate-valence ground state of Uranium with 70% weight of the $5\boldsymbol{f}^2$ configuration [19]. A pure U-$5\boldsymbol{f}^2$ configuration as proposed in [16] is unlikely since, owing to the results of Fig. 3b, the expected quadrupolar ordering occurs – if at all – at extremely low temperatures, $\boldsymbol{T} < 50$mK [13]. If alloying with Th would have a dominating steric effect, one should expect the $5\boldsymbol{f}^3$ configuration to be stabilized by the Th-derived volume expansion. Since the opposite, i.e., a destabilization of the trivalent state, is inferred from the results of Fig. 3, we suspect that the valence change is governed by the change in the chemical potential when substituting Th^{4+} for U^{3+} ions.

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