Anomalous ground state of $U_{0.9}Th_{0.1}Be_{13}$

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A study of the transformation of the temperature dependence of the specific heat in the series of solid solutions $U_xTh_{1-x}Be_{13}$ ($0.07 < x < 1$) with $x = 0.9$ at temperatures $0.8 < T < 6$ K revealed a nontrivial logarithmic increase in the electron component of the specific heat, C_e/T, as a function of the temperature T. This increase indicates the formation of a ground state which is not a Fermi-liquid state. Analysis of the effect of a magnetic field suggests that a quadrupole two-channel Kondo effect may be realized in $U_{0.9}Th_{0.1}Be_{13}$.

Despite recent progress toward an understanding of the physics of heavy-fermion compounds based on cerium as concentrated Kondo lattices,[1] the reason for the formation of a huge effective mass in the uranium-based heavy-fermion compounds remains an open question.[2] Of particular interest is the superconducting heavy-fermion compound UBe_{13}, which exhibits record values $\gamma = C_e/T \approx 1$ J/(mole·K^2), and in which the temperature dependence of the resistivity is similar to that which has been observed for the first superconducting Kondo lattice: $CeCu_2Si_2$ (see the review[1]). An attempt to reach an understanding of the nature of the ground state of UBe_{13} through a study of the transport properties[3] and the thermoelectric properties[4] of several solid solutions with the general formula $U_xTh_{1-x}Be_{13}$ ($0.07 < x < 1$) (i.e., by an approach like that which has been realized[5] for the case of $Ce_xLa_{1-x}Cu_2Si_2$) revealed a substantial difference between the impurity limit ($x \ll 1$, spin fluctuations) and the concentrated limit ($x \sim 1$, Kondo effect). A detailed further study[6] of some compounds $U_xM_{1-x}Be_{13}$ ($0.005 < x < 1$; M = Hf, Zr, Sc, Lu, Y, Pr, Ce, Th, and La) revealed a fundamental difference in the nature of the ground state of the U atom, depending on whether the nonmagnetic matrix shrank or expanded during the substitution. With regard to the dependence of $\gamma(T \rightarrow 0)$ on the lattice constant, it was found that the corresponding function is asymmetric and discontinuous[6] at the point corresponding to UBe_{13}. Such nontrivial behavior might stem from a realization in this compound of a so-called electric quadrupole Kondo effect.[2] A recent report[7] of a possible observation of this effect in dilute $Y_{1-x}U_xPd_3$ solid solutions near $x=0.2$ was explained in an alternative way,[8] however, as resulting from a magnetic phase transition at $T=0$.

In the present letter we are reporting the results of a study of the temperature dependence of the specific heat of some $U_xTh_{1-x}Be_{13}$ compounds $(0.07<x<1)$. In a previous study,[6] only the values of γ were reported for a series of compositions (x) in $U_xTh_{1-x}Be_{13}$. For the composition $x=0.9$, nearest UBe_{13}, we found that the specific heat divided by the temperature increases logarithmically with T in the interval $0.8 < T < 6$ K. That curve becomes a straight line in the coordinates $C/T=f\{\ln(T+0.3\text{K})\}$down to $T\sim 0.3$ K. The nature of the transformation of the $C_e(T)$ curve in a magnetic field suggests the possible observation of a two-channel quadrupole Kondo effect[2] in $U_{0.9}Th_{0.1}Be_{13}$.

The details involved in the preparation of the polycrystalline samples are described in Refs. 3 and 4, as is the method for determining the parameter x. The specific heat was measured in a standard helium cryostat $(2.5 < T < 25$ K) and in a ^3He cryostat $(0.3 < T < 4$ K) by a quasiadiabatic and relaxation technique. In the latter case, the τ_2 effect was taken into account.

Figure 1 shows the temperature dependence of the molar electron specific heat C_e, divided by T, for six compositions x in $U_xTh_{1-x}Be_{13}$. For a physical analysis of the experimental data, it is more convenient to calculate the specific heat introduced by a U atom. Corresponding values found at $T=1$ K and normalized to x, $\gamma_m=C_e/(T_x)$, are shown in the inset in Fig. 1. The quasilinear decrease in γ_m with decreasing x (after a sharp decrease near $x\sim 0.9$) confirms that the ground state is of a spin-fluctuation nature[3,4] for $x<0.8$ (with a change in the characteristic temperature $dT_{sf}/dx=10$ K/ x) upon substitution of Th. We recall that T_{sf} is essentially unchanged[6] when U is replaced by Y or Sc. In this case there is a single-channel Kondo effect.

We now consider the region $x\sim 1$. The fundamental result of this study, shown in Fig. 2, demonstrates a nonanalytic (logarithmic in the temperature) increase in C_e/T in the interval $0.8 < T < 6$ K. This increase reflects the formation of a non-Fermi-liquid ground state in $U_{0.9}Th_{0.1}Be_{13}$. We immediately add and stress that data[9] on UBe_{13} cannot be converted into a linear dependence over such a broad temperature range in terms of the coordinates $C_e/T=f(\log t)$. The application of a magnetic field $H=8$ T results in the observation of only a slight decrease (on the order of 10%) in C_e/T at $T<1$ K, while at $T>1.5$ K a magnetic field has essentially no effect on the specific heat.

If we assume that the Th atoms are distributed uniformly over volume (this hypothesis is supported by the absence of any indications of the superconducting transition which occurs[9] in $U_xTh_{1-x}Be_{13}$ at $0.96 < x < 1$), then the logarithmic

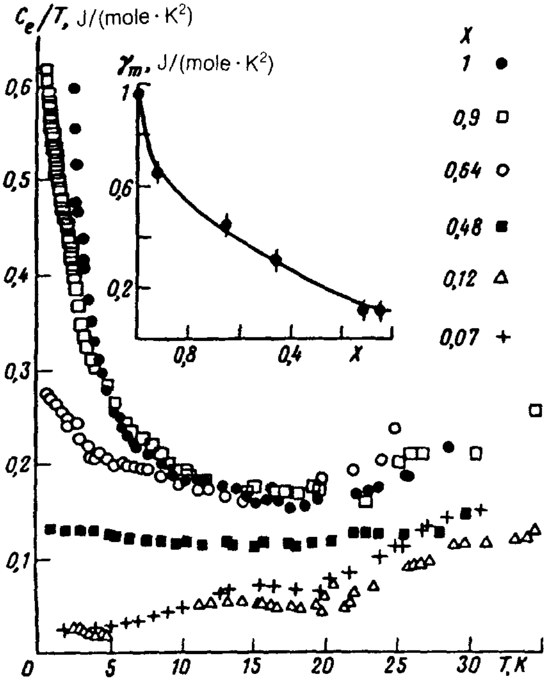

FIG. 1. Temperature dependence of C_e/T for $U_xTh_{1-x}Be_{13}$. The inset shows $\gamma_m = C_e/T_x$ as a function of x.

anomaly observed in $U_{0.9}Th_{0.1}Be_{13}$ can be explained on the basis of the existing theoretical approaches as resulting from (*i*) a two-channel Kondo effect,[10,11] (*ii*) a two-channl electric quadrupole Kondo effect,[2] or (*iii*), by analogy with Ref. 8, the occurrence of a second-order magnetic phase transition at $T=0$.

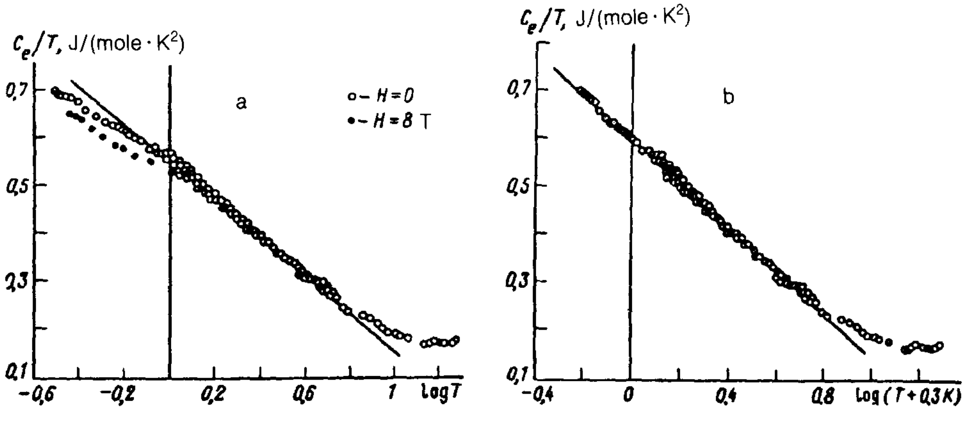

FIG. 2. a—C_e/T versus $\log T$ for $U_{0.9}Th_{0.1}Be_{13}$ in a zero magnetic field and in a field $H=8$ T; b—electron component of the specific heat divided by T, i.e., C_e/T, versus $\log(T+0.3\text{ K})$ for $U_{0.9}Th_{0.1}Be_{13}$ in a zero magnetic field.

The coexistence of a logarithmic increase in C_e/T and a phase transition (*iii*) at a low but nonzero temperature T_0 (we could then take the temperature at which $C_e/T \sim \log T$ deviates from linearity as $T_0 \approx 0.8$ K) seems improbable.[8] Furthermore, in case (*iii*) a composition deviation in either direction should probably lead to the appearance of a finite magnetic-transition temperature, and the effect of a magnetic field should strengthen sharply with decreasing T. This behavior is not seen experimentally. On the other hand, a calculation of the transformation of the specific heat for the classical two-channel Kondo model (*i*) has shown[11] that under the influence of a magnetic field one should observe a fundamental restructuring of the ground state in the direction of a normal Fermi-liquid state, and the $C_e(H)$ curves should be nonmonotonic—decreasing in a field as $T \rightarrow 0$ and increasing at intermediate temperatures, on the order of the Kondo temperature.

The two-channel quadrupole Kondo effect,[2] which was originally proposed in an effort to explain the properties of UBe_{13}, seems to us the most suitable explanation of the experimental results which have been found. An important aspect of this approach is that it presupposes the realization of a nonmagnetic doublet Γ_3 in the ground state of the U atom, while an effective spin 1/2 is created by local quadrupole degrees of freedom. The effect of laboratory magnetic fields on the thermodynamic and transport properties thus could not be strong. Furthermore, Cox[2] has predicted that it would be possible to observe a Jahn–Teller transition at an extremely low temperature, on the order of $T_{JT} \approx T_k \exp(-T_k/T_{JT0})$ (where T_{JT0} is the temperature of the JT transition in the absence of a Kondo effect). In this situation one might suggest that the electron specific heat would be more nearly linear in the coordinates $\log(T+T_{JT})$. Such a plot, as shown in Fig. 2b, can be used to find some estimates: $T_{JT} \approx 0.3$ K and $T_{JT0} \approx 1.6$ K. A more detailed study (presently in the planning stage) of the behavior of the specific heat, of the thermal expansion, and of the electrical resistance in various magnetic fields will permit a more comprehensive answer to the question of the applicability of models (*i*)–(*iii*) through a study of scaling.[12]

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