Anomalously weak ferromagnetism of $U_xTh_{1-x}Be_{13}$ (0.48<x<1) compounds

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A study of the temperature dependence of the magnetic susceptibility of UBe_{13} and of the solid solutions $U_xTh_{1-x}Be_{13}$ ($0.07<x<1$) in weak magnetic fields ($H<1$ kOe) indicates that weak ferromagnetic correlations arise for compositions with $x>0.5$ at temperatures below 150–200 K. © *1994 American Institute of Physics.*

The heavy-fermion superconductor UBe_{13} has been the subject of very active research over the past decade. The electron specific heat of this compound is the highest among uranium compounds,[1] and the stability of the superconducting state with respect to magnetic fields near T_c is the record high.[2] The most systematic theory (the Cox model[3]) which has been offered in an effort to explain the properties of the nontrivial ground state of UBe_{13} is based on the assumption that the levels of the $J=4$, f^2 multiplet, split by the cubic crystal field, are in an unusual arrangement for Kondo lattices, with the ground state corresponding to a Γ_3 doublet (not a Kramers doublet). In this model, the low-temperature magnetic susceptibility is of a purely Van Vleck nature and should exponentially reach saturation as a function of the temperature as T is lowered. According to the Cox model,[3] however, a finite hybridization should be manifested in a quantum filling of excited magnetic energy levels, even at temperatures $T<\Delta_{CF}$ (Δ_{CF} is the minimum splitting of the $5f$ level of U by the crystal field), and thus in a strengthening of the magnetic response of the system at low temperatures and in an asymptotic saturation of the magnetic susceptibility as $T\to 0$, which is a square-root function of the temperature.[4] The second of these properties has recently been confirmed experimentally[4] for $U_{0.9}Th_{0.1}Be_{13}$.

With regard to the temperature dependence of the magnetic susceptibility of UBe_{13} over a broad temperature range, all the studies which have been published to date have been carried out in magnetic fields of 5 kOe, to the best of our knowledge.[5] A Curie–Weis law with an effective magnetic moment approximately equal to the free moment of the U^{3+} and U^{4+} atoms has been observed at temperatures above 150 K. Below 150 K, all that has been observed is some deviation from the Curie–Weis law.

In this letter we are reporting the first experimental study of the magnetic susceptibility of UBe_{13} and of solid solutions $U_xTh_{1-x}Be_{13}$ ($0.07<x<1$) over the temperature range $1.7<T<300$ K, in magnetic fields from 100 to 5×10^4 Oe (preliminary results were reported at an international conference on the physics of strongly correlated electron systems[4]). The experiments were carried out on a single-crystal UBe_{13} sample and also on polycrystalline $U_xTh_{1-x}Be_{13}$ samples ($x=1$, 0.9, 0.64, 0.48, and 0.07; data on the

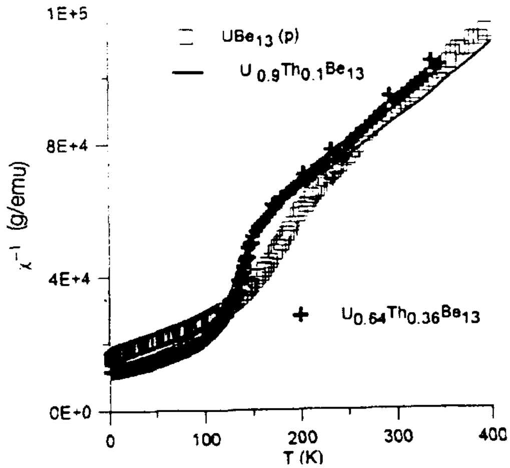

FIG. 1. Temperature dependence of the reciprocal magnetic susceptibility per U atom of polycrystalline samples of $U_xTh_{1-x}Be_{13}$ ($x=1$ at $H=100$ Oe; 0.9 and 0.64 at $H=1000$ Oe).

preparation of the samples and their analysis have been published previously[7,8]). We used a commercial (Quantum Design) SQUID. The UBe_{13} samples were superconductors with a transition temperature of about 0.9 K.

Figure 1 shows the temperature dependence of the reciprocal magnetic susceptibility, normalized to the U concentration, of polycrystalline U_x $Th_{1-x}Be_{13}$ samples ($x=1$ for $H=100$ Oe; 0.9 and 0.64 for $H=1000$ Oe). The fact that the high-temperature asymptotes of the $\chi^{-1}=f(T)$ dependence are linear in T and essentially coincident is evidence that the energy levels of the split $5f$ ground state do not change and that they correspond to the presence of an effective magnetic moment on the order of $3.5\mu_B$ per uranium atom at $T>200$ K. At temperatures $T<200$ K for UBe_{13} and $T<150$ K for $U_xTh_{1-x}Be_{13}$ ($x=0.9$, 0.64, 0.48), there is a substantial deviation in weak magnetic fields. The direction of the deviation corresponds to an increase in the value of the magnetic susceptibility (Fig. 1). A magnetic field $H>10^4$ Oe suppresses this effect.

Figure 2 shows results of measurements of the magnetic susceptibility of single-crystal UBe_{13} in magnetic fields of 10^3, 10^4, and 5×10^4 Oe. Again, as in the case of the polycrystalline samples with $x>0.48$, we observe the appearance of correlations of an electromagnetic type at a temperature $T<150$ K in weak magnetic fields. These correlations can be suppressed by a magnetic field. The effect of a magnetic field on the magnetic susceptibility is seen most clearly below two characteristic temperatures, $T<T_1=\Delta_{CF}=150$–180 K and $T<T_2=T_K$ [$T_K=10$ K (Ref. 4) is the Kondo temperature].

The most convincing argument in favor of the presence of ferromagnetic correlations at $T<150$ K is the slight hysteresis on the field dependence of the magnetic moment (Fig. 3). This figure shows data obtained on single-crystal UBe_{13}. Similar results were found for the compounds $U_xTh_{1-x}Be_{13}$ with $x>0.48$. The residual magnetic moment corresponds to an effective moment of less than $10^{-2}\mu_B$ per uranium atom. Another important experimental result is the complete absence of any indications of ferromagnetic correlations in the compound $U_{0.07}Th_{0.93}Be_{13}$, with a low concentration of uranium at-

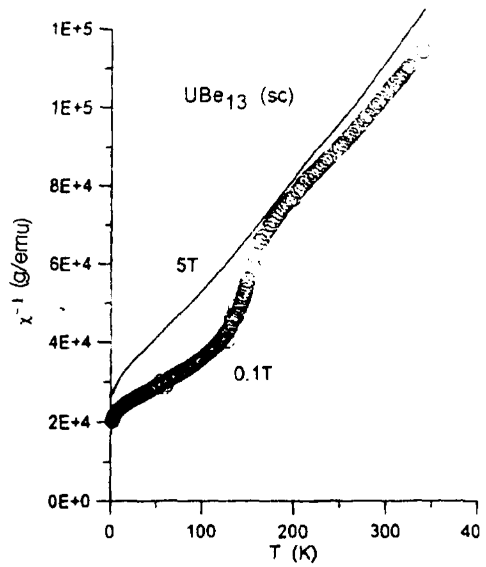

FIG. 2. Temperature dependence of the reciprocal magnetic susceptibility of single-crystal UBe_{13} in magnetic fields of 10^3 and 5×10^4 Oe.

oms. In this case, in contrast, the effect of a magnetic field on an isolated atomic uranium ion is at a maximum at high temperatures, $T>100$ K, and is negligible at low temperatures.

We should begin a discussion of the results with a possible effect of ferromagnetic impurities. For the most likely candidates here—uranium oxides—the temperature of the ferromagnetic transition does not exceed 50 K. Another possible impurity, UH_3, with a transition temperature of about 180 K, is extremely unstable and quickly decomposes. Although we cannot completely rule out a possible effect of impurities, the possibility that the observed effect is of a nonimpurity nature is suggested by the following facts: 1) the ferromagnetic correlations, in both the polycrystalline samples and the single-crystal

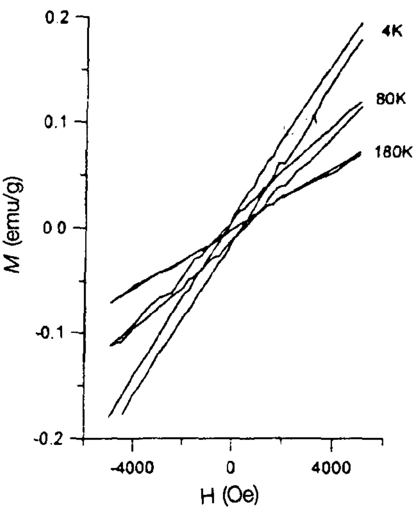

FIG. 3. Magnetic moment versus the field for a single-crystal UBe_{13} sample at various temperatures.

UBe_{13}; 2) the circumstance that the temperature below which ferromagnetic correlations are observed in polycrystalline UBe_{13} is greater than 200 K; 3) the absence of correlations in dilute solid solutions $U_xTh_{1-x}Be_{13}$. There are also several additional, indirect experimental results, discussed below.

Weak ferromagnetic correlations are exceedingly unusual for heavy-fermion systems, in which one customarily considers only the possibility of a realization of antiferromagnetic correlations. Interestingly, the temperature below which the correlations are observed corresponds to an energy $\Delta_{CF}=150-180$ K of the splitting of the ground state by the crystal field.[9] It is thus the change in the nature of the magnetism of the uranium atoms, which occurs at $T<\Delta_{CF}$ according to the Cox model,[3,10] that can induce the onset of a weak ferromagnetism at these temperatures.

The presence of weak ferromagnetic interactions, which can be suppressed by a field of a few kiloersteds, leads to an explanation of several anomalous experimental properties of UBe_{13}. For example, the scaling of the magnetoresistance which has recently been found for single-crystal UBe_{13} can be explained only on the basis that weak ferromagnetic correlations are present.[11] In addition, recent studies have revealed that the intensity of quasielastic neutron scattering depends weakly on the momentum transfer Q (Ref. 12). This result may be indirect evidence in favor of ferromagnetic correlations. Finally, the presence of weak ferromagnetic correlations can explain why the superconducting transition temperature of UBe_{13} is essentially independent of the magnetic field at fields[2,7] $H<3-5$ kOe: in this case, the temperature of the superconducting state decreases because of a local magnetic field in the absence of an external magnetic field. The suppression of correlations by an external field then "amplifies" the superconductivity and compensates for the energy loss of the superconducting condensate due to the application of an external magnetic field.

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