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M. J. Graf
Boston College

Nathanael A. Fortune
Boston University

J. S. Brooks
Boston University

J. L. Smith
Los Alamos National Laboratory

Z. Fisk
Los Alamos National Laboratory

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Specific heat of pure and thoriated UBe_{13} at low temperatures in high magnetic fields

M. J. Graf*
Department of Physics, Boston College, Chestnut Hill, Massachusetts 02167

N. A. Fortune and J. S. Brooks*
Department of Physics, Boston University, Boston, Massachusetts 02215

J. L. Smith and Z. Fisk
Center for Material Science, Los Alamos National Laboratory, Los Alamos, New Mexico 87545
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We have measured the specific heat of pure and thoriated (3.3% thorium) UBe_{13} in the temperature interval 0.3–1.8 K in applied magnetic fields up to 20 T, and find striking qualitative differences between the two systems. For pure UBe_{13} the electronic coefficient of specific heat γ_{el} $[=C_{el}(T)/T]$ is noticeably suppressed by a field of 20 T at 1.8 K; below 1 K, however, this reduction from the low-field, normal-state value tends to zero. For the 3.3% thoriated sample, the high-temperature value of γ_{el} is relatively insensitive to strong magnetic fields, but at lower temperatures the suppression of γ_{el} becomes much stronger, reaching nearly 40% at 0.35 K in a field of 20 T. This behavior is similar to that observed in most cerium-based heavy-fermion systems.

The properties of the heavy-fermion superconductor UBe_{13}, and of heavy-fermion systems (HFS) in general have raised many interesting questions in recent years.[1] At low temperatures the properties are those of a nearly localized itinerant electron system characterized by an electron effective mass several hundred times larger than the bare electron mass. Attempts have been made[2] to describe the low-temperature properties of HFS as a "Kondo lattice," that is, a periodic array of magnetic scatterers; surprisingly, many properties at low temperatures can be described by a single-ion Kondo model.[3] The electronic coefficient of specific heat γ_{el} (which is proportional to the electron effective mass) of $CeAl_3$,[4] $CeCu_6$,[5] and nonsuperconducting $CeCu_2Si_2$ (Ref. 6) is strongly reduced by the application of moderate magnetic fields (<10 T) at low temperatures [although recent measurements of the specific heat of superconducting $CeCu_2Si_2$ (Ref. 7) at 2.4 K show a much smaller reduction of γ_{el} in 23 T]. This is the expected behavior if the large effective mass of the conduction electrons is caused by Kondo-like scattering from the magnetic moments. Studies of UBe_{13} in magnetic fields[8] have shown that the normal-state specific heat is reduced by about 10% in 13 T at 2 K, but is independent of field ($H<8$ T) below 1 K. Samples doped with a few percent thorium in place of uranium have shown that in addition to the appearance of a second transition at low temperatures,[9] the specific heat below 1 K may be slightly more dependent on magnetic field.[10]

In this work we focus on the low-temperature specific heat of UBe_{13} in strong magnetic fields in order to determine if a magnetic field suppression of γ_{el}, similar to the previously mentioned examples, can be observed. We have extended the range of results on the specific heat of UBe_{13} and $U_{0.967}Th_{0.033}Be_{13}$ and $U_{0.967}Th_{0.033}Be_{13}$ to applied magnetic fields up to 20 T for temperatures between 0.3 and 1.8 K, and our results show that UBe_{13} is qualitatively much different from $U_{0.967}Th_{0.033}Be_{13}$ as well as the cerium-based HFS.

The specific heat was measured using the standard ac calorimetric technique.[11] The driving frequency ν of the sample heater was between 0.75 and 2.00 Hz, and the amplitude of the 2ν temperature oscillation was always $<0.5\%$ of the ambient sample temperature. Because the thermal diffusivity (ratio of thermal conductivity to specific heat) of the UBe_{13} is very small, thin flakes of sample (maximum thickness of 0.006 in.) were used to reduce the internal thermal time constant. The samples, with a total weight of a few tenths of a milligram, were mounted on a thin sapphire disk using either Wakefield thermal grease or silver epoxy, along with a thermometer (a small chip of Matsushita 100-Ω resistor) and a CrTi thin-film heater. The heat capacity of the addenda was always less than 15% of that of the samples, and was measured separately and subtracted from the data. Four 0.003-in.-diam brass wires provided a weak thermal link (0.7 μW/K at 1 K) to an epoxy-brass wire composite post which acted as a strong thermal link (100 μW/K at 1 K) to the ^{3}He-^{4}He mixture of a top-loading dilution refrigerator. The ambient sample temperature was controlled by applying heat to the thermally isolated end of the epoxy-brass wire post and keeping the refrigerator at a base temperature of 40 mK. The data presented here were obtained by cooling the samples in zero field to low temperatures, applying a magnetic field, then slowly warming the sample. Magnetic fields were supplied by resistive solenoids at the Francis Bitter National Magnet Laboratory.

The sample thermometer was calibrated in zero field with a calibrated germanium thermometer. To calibrate in magnetic field we have assumed that the temperature of the post at a fixed external power input (with the dilution refrigerator at base temperature) was independent of magnetic field; this assumption is only valid above approximately 0.4 K, as at lower temperatures the effects of eddy

current heating caused by magnetic field ripple are no longer negligible. This lower limit was determined by both theoretical estimates of heating and by comparison with results obtained with an 8-T superconducting magnet (which has no field ripple). Our assumption also implies that the thermal conductivity of brass is independent of field, so as a test for self-consistency we have measured the thermal conductivities of both the weak and strong thermal links and find them to be constant in magnetic field. Magnetoresistive corrections to the specific-heat data were largest at high temperatures in strong fields, where dR/dT increased by 15% between 0 and 20 T at 1.8 K, but were less than 5% below 1 K.

In Fig. 1 we show the results for the specific heat of polycrystalline UBe_{13} divided by temperature versus temperature for several values of applied magnetic field. The lattice contribution is negligible. We have pinned our results at 1 K in zero field to results for samples with a similar T_c,[12] and estimate our total mass to be 0.52 μmol, in agreement with direct measurements of the sample weight, which had an uncertainty of 20%. In zero field the normal state γ_{el} increases weakly with decreasing temperature, and a sharp superconducting transition occurs at 0.90 K. In an applied field of 9 T, the data are slightly reduced from the zero-field data above T_c, but this reduction is within a factor of 2 of the estimated relative error of 2–3%. The superconducting transition has been completely suppressed over the entire temperature range covered. For analysis we assume γ_{el} in 9 T is approximately equal to its fictitious zero-field normal-state value for $T < T_c$, and compare the higher-field data to the 9 T data. The specific heat in 15 T is reduced by 9% at 1.8 K, in fair agreement with the results of Ref. 10. However, as the temperature is lowered, this reduction *decreases,* so that below 0.7 K the 15 and 9 T data are indistinguishable within our resolution. At 20 T, the data at 1.8 K are reduced by approximately 14%, and at lower temperatures this reduction initially becomes more substantial. However, below 1 K this trend reverses itself, and γ_{el} becomes *less sensitive* to magnetic field as the temperature is lowered further. This behavior is shown in Fig. 2, where we have plotted the percent change of the specific heat in a field of 20 T relative to the 9 T data as a function of temperature. Based on the data shown in Fig. 2, and the behavior of the data at 15 T, we then infer that at some temperature lower than the range covered here the specific heat at 20 T will be the same as that at 9 T.

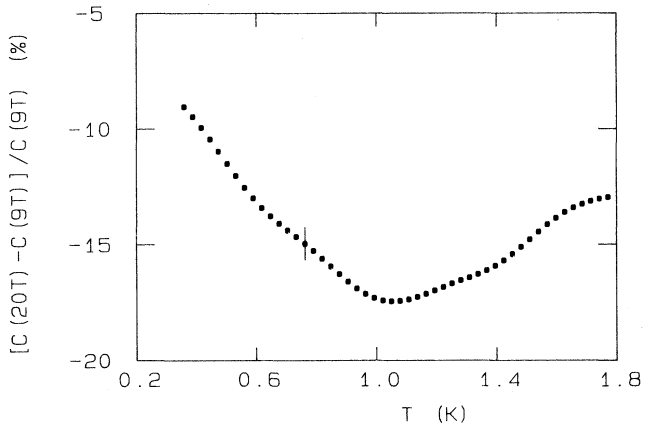

FIG. 2. The percent change of the 20 T data relative to the 9 T data vs temperature for UBe_{13}. The data have been smoothed, and the error bar shown was determined by the root-mean-square deviation of the smoothed data from the actual data.

The data taken for polycrystalline $U_{0.967}Th_{0.033}Be_{13}$ (estimated mass equals 0.31 μmol as determined by pinning our results at 0.7 K in zero field to those of Ref. 12) are shown in Fig. 3. The behavior in magnetic field is qualitatively very different from that for UBe_{13}. Two sharp transitions are observed at 0.60 and 0.42 K. There is a small reduction in γ_{el} at 1.4 K (the highest temperature studied for this sample) by the application of fields as high as 20

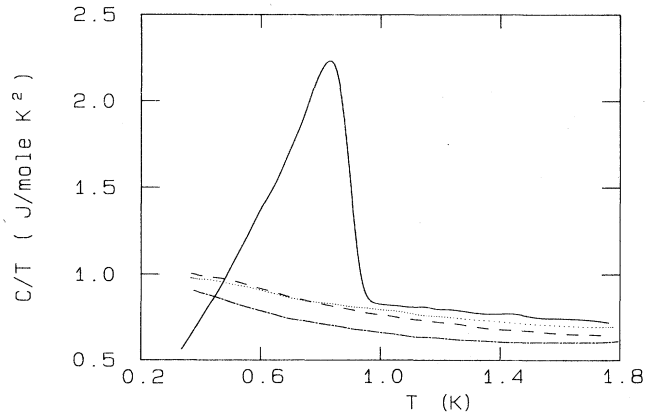

FIG. 1. Specific heat divided by temperature vs temperature for UBe_{13} in applied magnetic fields of 0 (solid), 9 (dotted), 15 (dashed), and 20 (dot-dashed) T. The absolute value of the specific heat was determined by pinning our results at 1.0 K in zero field to those of Ref. 12 for polycrystalline UBe_{13}.

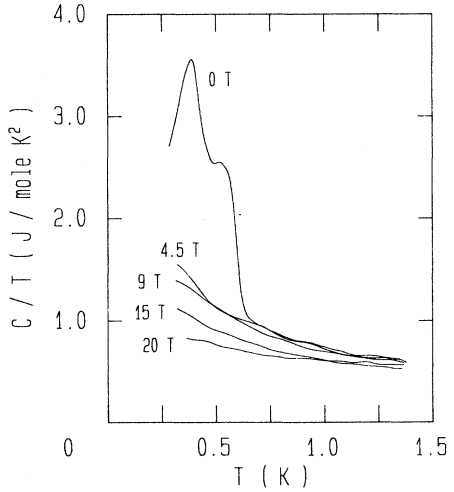

FIG. 3. Specific heat divided by temperature vs temperature for $U_{0.967}Th_{0.033}Be_{13}$ for several values of applied magnetic fields. The absolute value of the specific heat was determined by pinning our results at 0.7 K in zero field to those of Ref. 12 for a sample with a thorium concentration of 0.0331.

T. As the temperature is lowered, however, the effect of the field becomes much more pronounced—at 0.35 K, the 20-T data is reduced by nearly 40% from the 9 T data. In Fig. 4 we have replotted the data at fixed temperatures (0.35 and 0.70 K) and show C/T as a function of magnetic field. The observed reduction at 0.35 K is essentially linear with field, and we find the slope to be -46 mJ/(mol T K^2). A linear least-squares fit is shown in the figure as a solid line. At 0.70 K, the data vary more weakly with magnetic field, and only above 10 T do we begin to see a strong reduction in C/T. This linear reduction in field is similar to that observed in $CeAl_3$.[13]

Data taken for a sample with 1.5% thorium were qualitatively similar to the data for $U_{0.967}Th_{0.033}Be_{13}$, but the observed reduction of γ_{el} in field at low temperatures was nearly three times smaller. Thus the differences between the pure and thoriated samples are not extrinsic but related to the amount of thorium. More detailed measurements on the variation with thorium concentration will be presented elsewhere.

While we have no complete theoretical interpretation for these results, our observations indicate a crossover to a low-temperature state for which the correlations resulting in the large electron effective mass are more resistant to the effects of magnetic field. One possible explanation is that as the temperature is lowered below 1.8 K the UBe_{13} is crossing over from single-ion Kondo behavior to that of a coherent Kondo lattice (the resistivity peak which is usually associated with the onset of coherent behavior occurs at 2.4 K). If the renormalized Kondo temperature T_K^* below 1 K is enhanced from the single-ion value T_K, than a larger characteristic field for the suppression of spin-flip scattering occurs at lower temperatures. Variational calculations[14] indicate that the characteristic energy scale may be greatly enhanced as one enters the coherent state, although perturbation calculations[15] show the effect to be very weak. The addition of (nonmagnetic) thorium to the system would then alter the periodicity of the magnetic scatterers, and suppress (or possibly eliminate) the crossover to coherent behavior. Resistivity measurements[16] have shown that 3.3% thoriated samples have no resistance peak at 2.4 K, reflecting a suppression of coherence. This crossover picture is inconsistent with other results, however. Magnetoresistance studies[17] on UBe_{13} have been interpreted using a single-ion Kondo model with a Kondo temperature that *decreases* with decreasing temperature. Also, the incoherent-to-coherent crossover below 1 K has been well documented in specific-heat studies[4] of $CeCu_2Si_2$ and $CeAl_3$, systems which show a clear suppression of γ_{el} by magnetic fields while in the coherent state. Clearly a fundamental difference between these systems and UBe_{13} must be explained in order to ascribe the observed properties of UBe_{13} to the onset of coherence.

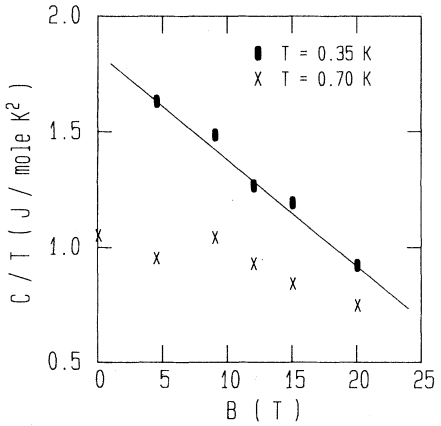

FIG. 4. Specific heat divided by temperature vs magnetic field for $U_{0.967}Th_{0.033}Be_{13}$ at temperatures of 0.35 and 0.70 K. The solid line is a linear least-squares fit to the data at 0.35 K with a slope of -46 mJ/(mol T K^2). The absolute value of the specific heat was determined as for Fig. 3.

Auerbach and Levin[18] suggest that the properties of heavy-fermion systems are dominated by a single, *temperature-independent* energy scale T_K, but we have been unable to find a field-dependent Kondo temperature $T_K(H)$ which scales our UBe_{13} data onto a single universal curve. This failure may be due to the high degree of polarization of the conduction electrons below 2 K in strong magnetic fields.

In the quadrupolar Kondo model for uranium HFS,[19] below the Kondo temperature the properties of the U sites become dominated by a proposed nonmagnetic Γ_3 doublet ground state arising from crystal-field splitting. As the temperature is lowered the effective moment approaches zero, again requiring larger fields for Zeeman splitting of the levels. This model then correctly predicts the observed temperature dependence of γ_{el} for UBe_{13} in large fields. If the presence of small amounts of thorium in the system is sufficient to appreciably alter the crystal field and suppress the formation of the Γ_3 doublet, the data for the field dependence of $U_{0.967}Th_{0.033}Be_{13}$ can also be explained. Both UBe_{13} and $ThBe_{13}$ have cubic $NaZn_{13}$ structure, and neutron scattering has shown the lattice constants for these two systems differ by only a few percent.[20] Assuming the crystal fields at the U (Th) sites are dominated by nearest-neighbor Be atoms, the quadrupolar state would have to be extremely sensitive for low concentrations of thorium to suppress its formation.

In summary, we have made the first measurements of the specific heat of pure and thoriated UBe_{13} below 1 K in magnetic fields in excess of 10 T. We find that while the specific heat of $U_{0.967}Th_{0.033}Be_{13}$ is more strongly reduced by these fields as the temperature is lowered, UBe_{13} becomes less sensitive to field at lower temperatures. More detailed work in both theory and experiment is required before this qualitative difference or the differences between the U-based and Ce-based heavy-fermion systems can be fully understood.

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*Visiting scientist: Francis Bitter National Magnet Laboratory, MIT, Cambridge, MA 02139.

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