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Normal-state and superconducting properties of the heavy-fermion compound UBe_{13} in magnetic fields

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Low-temperature measurements (200 mK$\lesssim T \lesssim$18 K) of the specific heat $C(T)$ of polycrystalline UBe_{13} show that the normal-state electronic specific heat $C_n(T_c)/T_c$ remains virtually constant at applied magnetic fields of $B=1$, 2, 4, and 8. Suggestions to explain the maintenance of the entropy balance between the normal and the superconducting state are discussed. With $B=13$ T, $C(T)$ is decreased below 3 K compared to the zero-field data, and increased above this temperature. The entropy of the electron system increases linearly above 10 K without indication of saturation. The sharp discontinuity of $C(T)$ at the transition to the superconducting state and the large dc Meissner effect measured in the same compound suggest that UBe_{13} is a very homogeneous superconductor [$B_{c2}(0)\cong 10$ T]. From isothermal magnetization measurements at $T=100$ mK, we obtain $B_{c1}\cong 4.4$ mT and, for the Ginzburg-Landau parameter, $\kappa\cong 100$.

I. INTRODUCTION

After the discovery of heavy-fermion superconductivity in $CeCu_2Si_2$,[1] UBe_{13} was the second compound in which this phenomenon was observed.[2,3] Since then, a considerable amount of experimental information on UBe_{13} has been made available, but in spite of this, the nature of its ground state is still unclear. From the similarity of the low-temperature specific heat of UBe_{13} to that of liquid ^{3}He, Ott *et al.*[4] argue that superconductivity in UBe_{13} might involve triplet pairing, a mechanism so far observed only in liquid ^{3}He. Recent measurements of the upper critical field $H_{c2}(T)$ by Maple *et al.*[5] corroborate the idea that in UBe_{13} superconductivity is of some exotic type. But the authors were unable to describe their results by current theories of either conventional or *p*-wave superconductivity. As to the normal properties underlying superconductivity, the situation is just as confusing. A pronounced low-temperature maximum in the electrical resistivity[2] may indicate a coherent Kondo-lattice state, similar to that proposed for $CeCu_2Si_2$,[6] while a strong negative magnetoresistance for temperatures below this maximum appears indicative of incoherent scattering from localized magnetic moments. Rice *et al.*[7] generalized the Brinkman-Rice theory to finite temperatures to describe the normal-state thermodynamics of heavy-mass electrons in UBe_{13}. A more conventional view is taken in a recent publication by Overhauser and Appel,[8] where most properties of UBe_{13} are explained within a one-particle description assuming singlet pairing for the superconducting state.

In this paper, we present experimental results for the specific heat of polycrystalline UBe_{13} in the temperature ranges 200 mK$\lesssim T\lesssim$4 K and 2 $\lesssim T\lesssim$18 K and for applied magnetic fields of up to 8 and 13 T, respectively. While for $T\gtrsim 2$ K, similar experiments on single crystals of UBe_{13} have already been performed by Stewart *et al.*,[9] our data are the first collected in magnetic fields below 2 K (in particular, below T_c) published so far. Moreover, we report on the first measurement of the dc Meissner effect in the same compound. With our data, we offer new experimental information on the heavy-fermion superconductor UBe_{13} which may assist in further clarifying the physics of this interesting system.

II. EXPERIMENTAL PART AND RESULTS

The samples were prepared from Be (99.96% pure) and U (99.97% pure) materials by repeated arc melting under argon, starting with a 1 at.% Be surplus to compensate for losses due to evaporation. X-ray analysis revealed polycrystalline material of homogeneous and single-phased UBe_{13}. The very sharp specific-heat discontinuity at the transition into the superconducting state at $T_c=871$ mK (see below) may serve as a further proof for the homogeneity of the samples.

Measurements of the specific heat were performed in two different calorimeters, both of the semiadiabatic heat-pulse type. Mangetic fields were applied by use of superconducting solenoids. The first calorimeter was built into a dilution refrigerator. Because of self-heating of our samples due to uranium decay, the lowest sample

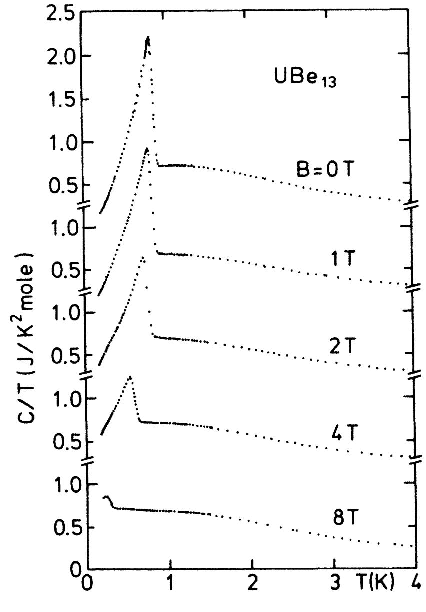

FIG. 1. Specific heat of UBe_{13} for 200 mK $< T <$ 4 K and in various magnetic fields plotted as C/T vs T.

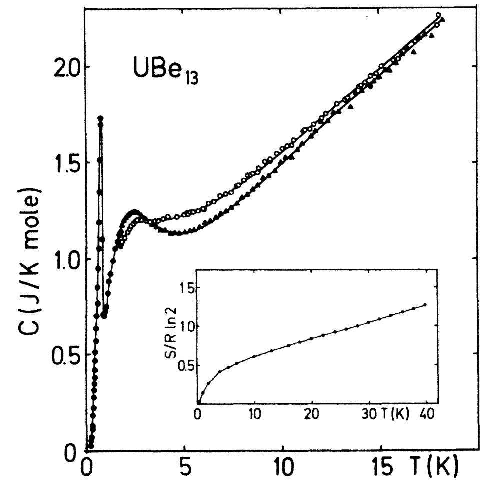

FIG. 2. Electronic specific heat of UBe_{13} for 200 mK $< T <$ 18 K in zero field (●,▲) and in a magnetic field of 13 T (○); the solid lines are guides to the eye. The inset shows the molar electronic entropy at zero field up to 40 K, including the data of Ref. 17.

temperature actually reached was 200 mK. In this facility, magnetic fields up to 8 T could be generated. The second calorimeter was installed in a ^{4}He cryostat which allowed for a temperature range between 2 and 18 K. There, a magnetic field of 13 T was applied.

In Fig. 1 we show our results for the specific heat $C(T)$ between 200 mK and 4 K and for applied magnetic fields of $B=0$, 1, 2, 4, and 8 T. The nuclear specific heat due to Zeeman splitting of nuclear states has been calculated and subtracted from the data. Since at temperatures below 4 K the lattice contribution to $C(T)$ is completely negligible, the data actually represent the electronic specific heat. Values for T_c, $C_n(T_c)/T_c$, and $\Delta C/C_n(T_c)$ are listed separately in Table I. In Fig. 2 the $C(T)$ results for $B=0$ and 13 T in the temperature ranges 200 mK $< T <$ 18 K and $2 < T < 18$ K, respectively, are put together. The lattice contribution as determined via inelastic neutron scattering[10] has been subtracted from the measured data in the temperature range $3 < T < 18$ K. In contrast to Fig. 1, where a C/T-versus-T plot is used, in Fig. 2 we preferred a C-versus-T plot in order not to obscure the S shape of the curve above 2 K. In the temperature range $2 < T < 4$ K the two sets of data for $B=0$ overlap. The agreement is excellent considering the fact that they emerged from independent measurements in different calorimeters.

III. MEISSNER EFFECT

The dc Meissner effect was measured on a 55-mg bulk sample which was cooled in various constant external fields ($B < 10^{-4}$ T) from $T > T_c$ to about 100 mK. The resulting changes in magnetization were recorded by means of a superconducting quantum-interference device. Figure 3 shows the corresponding ($-M$)-versus-B curve together with an isothermal magnetization curve (dashed line) taken at 100 mK on the same sample. One may safely assume that the initial slope of this latter curve reflects the full diamagnetic response, i.e., $-M=B/(1-D)$ (D is the demagnetizing factor of the sample). Comparison of these two $M(B)$ curves then yields a volume fraction of 25% from which the flux is expelled upon cooling (Meissner effect).

This result *does not* mean that only 25% of the sample become superconducting, since flux can be trapped at pinning centers or frozen in by ring-shaped regions of slight-

TABLE I. Transition temperature T_c^* (onset), T_c (midpoint), normal-state electronic specific heat $C_n(T_c)/T_c$, and relative jump height $\Delta C/C_n(T_c)$ in UBe_{13} for various fields B; ΔC and $C_n(T_c)$ have been extracted from our data by linearly extrapolating $C(T)/T$ from above and below T_c to T_c.

B (T)	T_c (K)	T_c^* (K)	$C_n(T_c)/T_c$ (J/mole K^2)	$\Delta C/C_n(T_c)$
0	0.871 (0.857)[a]	0.937	0.72	2.40
1	0.816 (~0.83)[a]	0.903	0.71	1.99
2	0.756 (~0.78)[a]	0.860	0.72	1.51
4	0.591 (~0.58)[a]	0.677	0.73	0.90
8	0.260 (~0.16)[a]	0.330	0.72	0.30

[a] $T_c(B)$ as taken from Ref. 5.

ly higher T_c. This has already been demonstrated by Lieke *et al.*[11] for $CeCu_2Si_2$. There, bulk samples yielded a Meissner effect of typically only a few percent, while, after powdering and annealing, the Meissner effect was raised to 60%. Thus, we consider 25% flux expulsion quite a high value and indicative of a very homogeneous material.

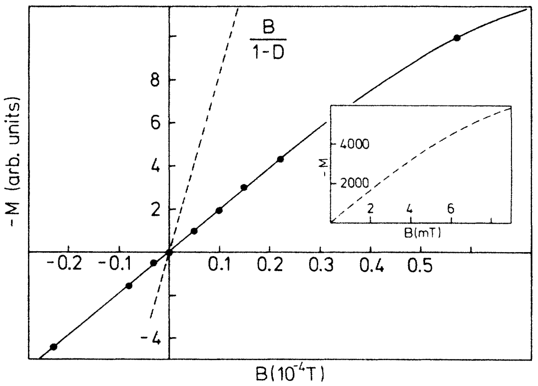

FIG. 3. Diamagnetic magnetization of a UBe_{13} bulk sample as measured in constant magnetic fields (dots) upon cooling from $T > T_c$ to 100 mK; the solid line is a guide to the eye. The dashed line represents the isothermal magnetization at 100 mK. The inset extends this isothermal curve to higher fields; note that at about 3.5 mT, $M(B)$ deviates from a straight line, thus indicating penetration of flux.

In the inset of Fig. 3 the extension of the isothermal magnetization curve at $T = 100$ mK to higher fields is shown. At $B_0 \cong 3.5$ mT, $M(B)$ starts to deviate from linear behavior. Using $D = 0.2$ as a rough guess in $B_{c1} = B_0/(1-D)$,[12] we find $B_{c1} \cong 4.4$ mT. With this value for B_{c1} and with the orbital critical field $B_{c2}^{*} \cong 25$ T,[5] the Ginzburg-Landau parameter can be estimated[12] to be $\kappa \cong 100$, which is very large.

IV. DISCUSSION

Our zero-field data below 1 K are rather similar to those of Ott *et al.*[4] A double-logarithmic plot of our results reveals, however, that our data cannot be fitted by either a T^3 or a T^2 power law (see Fig. 4). As a best fit we found $C_s(T) \propto T^{2.9}$ in the temperature range $200 \leq T \leq 850$ mK.

From the depression of T_c in magnetic fields, information upon the upper critical field $B_{c2}(T)$ may be deduced. In Table I our T_c values (defined by the midpoints of the transitions) are compared to those taken from Fig. 2 of Ref. 5 for the respective fields. For $B = 0$, 1, 2, and 4 T, they agree within a few percent, thus confirming the resistively determined data of Ref. 5 for the bulk. For $B = 8$ T, our T_c is somewhat higher, but in view of the appreciable width of this transition and the large error bars quoted for the high-field data in Ref. 5, this deviation must be considered insignificant.

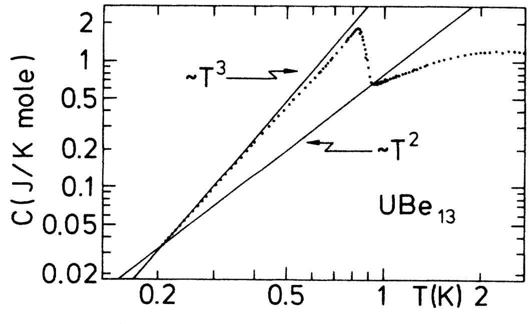

FIG. 4. Specific heat of UBe_{13} for $T < 3$ K. The zero-field data from Fig. 1 are compared in a log-log representation with simple power laws.

In the temperature range $T_c < T \leq 18$ K, the most conspicuous features of the electronic specific heat in UBe_{13} are the steep rise immediately above T_c and the S shape between 1 and 6 K (see Fig. 2). With a magnetic field of 13 T applied, $C(T)$ intersects the zero-field data at 3 K, very similar to the findings for $CeCu_2Si_2$ (Ref. 13) and $CeCu_6$ (Ref. 14). Below this point, $C(T)$ is lowered by the magnetic field, while above it, it is raised, thus leading to a weakening of the S-shaped structure. Stewart *et al.*[9] found $C(T)$ raised by a magnetic field of 11 T throughout the temperature range $2 \leq T \leq 11$ K, with a maximum increase at 3 K. Since these authors performed their measurements on a single crystal, the discrepancy may reflect possible anisotropy effects.

Defining $\gamma(B,T) = C_n(B,T)/T$, we find $\gamma(0,T_c) = 0.72$ J/mole K^2, considerably below the value of 0.86 J/mole K^2 taken from the data of Ref. 4. With increasing applied magnetic field, $\gamma(B,T_c(B))$ remains constant within the experimental errors and for $B_{c2}(0) \cong 10$ T we expect $\gamma(B_{c2},0) \cong 0.72$ J/mole K^2.

The fact that γ remains almost constant at the different transition temperatures $T_c(B)$ may be attributed to the compensation of two effects, namely an increase of γ at lower temperatures and a depression of γ in magnetic fields. The value of $\gamma(0,0)$ as measured in a fictitious experiment where superconductivity has been quenched by other means than application of a magnetic field should lie somewhat higher: If we assume that the depression of γ in the magnetic field is the same for finite temperatures and for $T = 0$, i.e., 50 mJ/mole K^2 in a magnetic field of 8 T, we arrive at $\gamma(0,0) \cong 0.77$ J/mole K^2. This would reflect an only very weak temperature dependence of γ for $B = 0$. However, such a number leads to a severe conflict regarding the entropy balance between the normal and the superconducting state. It has already been pointed out earlier[3,4] that the entropy in the superconducting state, $S_s(T_c) = \int_0^{T_c} (C/T)dT$, is significantly larger than that obtained in the normal state, $S_n(T_c)$, assuming a *constant* $\gamma(T)$ below $T_c(B=0)$. But, if the observed transition into the superconducting state is of second order, $S_n(T_c)$ must equal $S_s(T_c)$. Since there are no hints at a first-order transition in our $C(B,T)$ experiments, we have to choose between two possibilities in order to guarantee the entropy balance (see also Ref. 15): Either the positive part of $\int_0^{T_c} (1/T)(C_s - C_n)dT$ for $T \lesssim T_c$ contains other than purely electronic (e.g., magnetic) degrees of freedom, or the negative part for $T \ll T_c$ is underestimated by the weak temperature dependence in C_n/T at zero field as derived from our finite-field measurements; see above. In the latter case, an upturn in this quantity at sufficiently

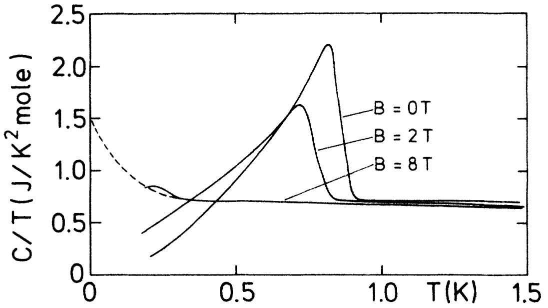

FIG. 5. Specific heat of UBe_{13} for $T < 1.5$ K at $B = 0$, 2, and 8 T. The dashed line is intended as a schematic illustration of how to conserve the entropy balance (see the text).

low temperature has to be anticipated. According to the $B \neq 0$ data, the most likely possibility, i.e., an upturn *below* $T_c(B = 8\ \mathrm{T}) = 0.26$ K, is schematically indicated in Fig. 5. Such a feature would suggest an extremely narrow peak in the heavy-fermion density of states just at the Fermi level. We note that a similar possibility has been recently discussed for the heavy-fermion superconductor UPt_3 too.[16]

We should like to mention that the magnetic susceptibility and the low-temperature specific heat in UBe_{13} were already measured by Bucher *et al.*[17] about ten years ago. Within the temperature range $2 < T < 18$ K our data follow closely these earlier results. We also determined the magnetic susceptibility in the temperature range $10 < T < 300$ K. From a χ^{-1}-versus-T fit of the data above 120 K, we found a Curie-Weiss temperature $\Theta = -100$ K and an effective moment $P_{\mathrm{eff}} = 3.34\mu_B$, again in good agreement with the values of Ref. 17 ($\Theta = -98$ K, $P_{\mathrm{eff}} = 3.52\mu_B$). Putting our results for the electronic specific heat together with those of Ref. 17 we can calculate the molar electronic entropy S for temperatures up to 40 K. Above $T = 10$ K, S increases linearly with T. It reaches a value of $1.27R \ln 2$ at 40 K and there is still no tendency to saturate.

V. CONCLUSION

Our measurements on polycrystalline samples show clearly that UBe_{13} is a rather homogeneous type-II superconductor, despite a very high Ginzburg-Landau parameter, $\kappa \cong 100$. Small but significant differences among data published in the literature (Refs. 4, 5, and 9) do, however, indicate that results for Ube_{13}, as for other heavy-fermion superconductors, may vary from sample to sample. An apparent mismatch between the entropies in the normal and superconducting states invokes interesting new effects, arising either from an extremely narrow peak in the heavy-fermion density of states at E_F or from additional (e.g., magnetic) degrees of freedom. Clarification of this point will most certainly help to understand several of the exotic properties of UBe_{13}.

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