Magnetic-field investigation of the 1-K transition in UCu_5

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Specific heat and electrical resistivity of UCu_5 have been measured from 0.35 to 20 K in magnetic fields up to 14 T. We report an almost tenfold drop in C/T at $T_c \sim 1$ K and $H=0$. The 2-T field does not affect T_c within the accuracy of the measurement. Magnetic fields of 5 T and larger strongly enhance T_c with an average rate of approximately of 0.24 T/K. The transition, which is hysteretic for $H=0$, is second order in high fields of 10 and 14 T. These results are discussed in relation to the most recent neutron-diffraction data, Cu NMR, and muon-spin-resonance measurements on UCu_5.

INTRODUCTION

Recently, much attention has been focused on UCu_5,[1-5] belonging to the family of heavy fermions.[6] The uniqueness of UCu_5 among all other U compounds and alloys lies in the fact that its low-temperature heavy-fermion ground state develops into a magnetic phase. Heavy electrons and antiferromagnetic order clearly coexist in this metal at low temperatures, below $T_N \approx 15$ K. The specific heat divided by temperature (C/T) reaches a value of about 400 mJ/K^2 mol at $T=1.5$ K.[1] At $T=1$ K $(=T_c)$ UCu_5 undergoes a second phase transition whose character has not been identified yet and was the subject of our investigation.

EXPERIMENT

Both T_c and the low-temperature C/T are critically dependent on sample preparation. Only high-quality, well-annealed samples have high C/T and exhibit the low-temperature transition. We have synthesized several UCu_5 samples of slightly varied composition and annealing procedures. Our best sample, described in this paper, was stoichiometric and annealed for 5 days at 900 °C.

The specific-heat results are presented in Figs. 1 and 2 in the form of the specific heat divided by temperature versus temperature. The zero-field C/T value at 1.5 K is only 320 mJ/K^2 mol, thus about 20% lower than that reported in Ref. 1. Another point of difference is the shape of C in the immediate vicinity of T_c. Our sample did not show a steep increase of C near T_c; instead, C/T increases logarithmically with decreasing T between 3 and 1.15 K and drops almost discontinuously to about 35 mJ/K^2 mol on the low-temperature side of T_c. Thus, the relative reduction of C/T at T_c ($\sim$90%) is even more spectacular than that previously observed.[1] In agreement with Ref. 1, we did observe a hysteretic behavior of the specific heat near T_c. The hysteresis (not shown) was consistent with supercooling and superheating or the first-order phase transition. However, the temperature shift of the anomaly between cooling and heating was only 60 mK versus 170 mK in Ref. 1. It is rather unlikely that all these mentioned discrepancies can be attributed to different measurement techniques: the relaxation method in our study and the semi-adiabatic method in Ref. 1. Vastly different rates of the temperature change used in both investigations produced essentially identical results. Also, a lack of the so-called τ_2 effect in our zero-field measurements argues against low thermal conductivity of the sample leading to erroneous results.

All the specific-heat data, shown in Figs. 1 and 2, were obtained while slowly raising the sample temperature. Interestingly, an application of a 2-T magnetic field did not affect the low-temperature values of C/T, nor did it change, within the accuracy of our measurement, T_c. On the other hand, fields of 5 T and larger significantly enhance T_c (temperature corresponding to C/T maximum) (see also Table I). The average rate of T_c raise between 5 and 14 T is 0.24 K/T. In addition, we have observed sharp, λ-like anomalies superimposed on the step-like change of the specific heat at T_c for $H=10$, 12, and 14 T (Fig. 2). These anomalies are also reminiscent of those observed in the zero-field specific heat by Ott *et al.*, although our high-field peaks are significantly narrower and larger in magnitude ($C/T \approx 1700$ mJ/K^2 mol at $T_c=3.6$ K in $H=14$ T). However, the measured values of C/T at T_c for $H \geq 10$ T bear sizable error bars (±

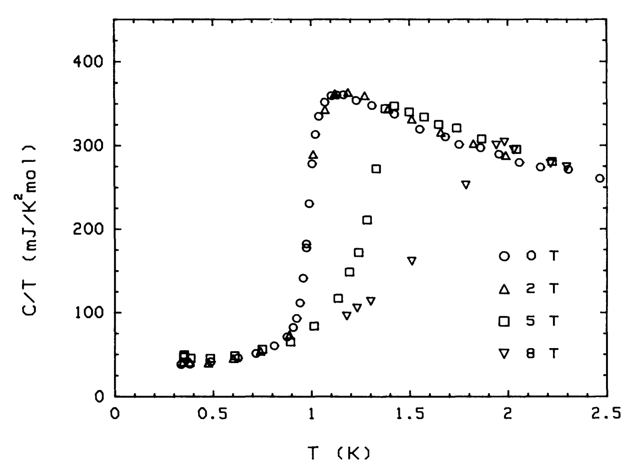

FIG. 1. Specific heat divided by temperature (C/T) vs temperature (T) of UCu_5 from 0.35 to 2.5 K and for 0-, 2-, 5-, and 8-T fields.

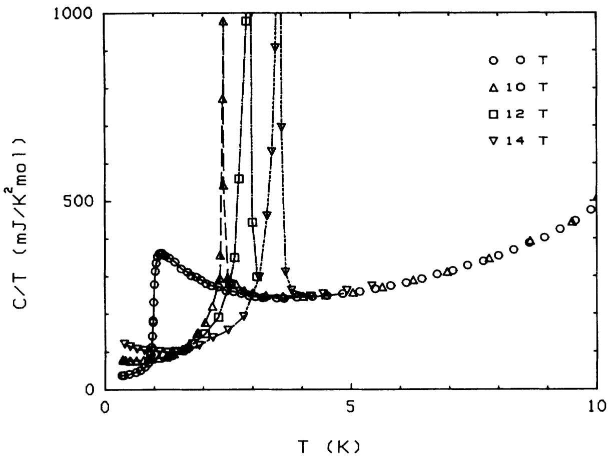

FIG. 2. C/T vs T of UCu_5 from 0.35 to 10 K and for 0-, 10-, 12-, and 14-T fields. C/T reaches 980 mJ/K^2 mol in 10 T, 1700 mJ/K^2 mol in 12 T, and 1640 mJ/K^2 mol in 14 T. The absolute error of these C/T values is ±30% vs 5% for C/T below and above T_c. Lines drawn are guides to the eye.

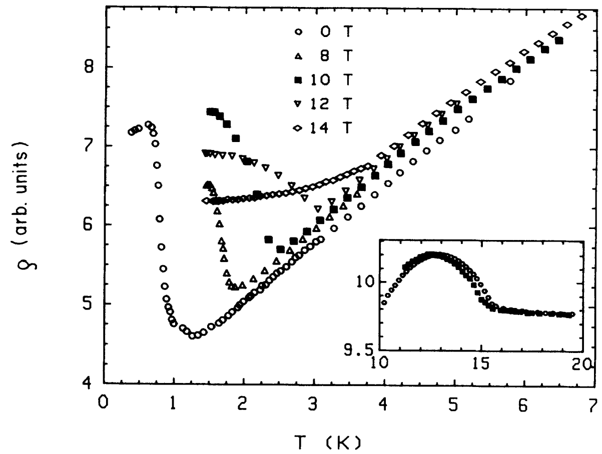

FIG. 3. Temperature dependence of the electrical resistivity of UCu_5 from 0.35 to 7 K and in 0-, 8-, 10-, 12-, and 14-T fields. The inset shows the electrical resistivity from 10 to 20 K for 0- and 10-T fields.

30%) due to an appearance of large τ_2 effects at T_c. (The large τ_2 values reflect poor thermal diffusivity of the sample.)

The specific heat above T_c is quite insensitive to magnetic fields. On the other hand, it gradually grows with H below T_c (this effect is clearly seen in Fig. 2 for $H \geq 10$ T). C/T at $T = 0.4$ K reaches 120 mJ/K^2 mol in $H = 14$ T, a fourfold increase from the $H = 0$ value. Finally, the hysteresis present in $H = 0$ seems to be absent in high magnetic fields. Several attempts to observe the hysteresis at T_c in $H = 10$ and 14 T gave a negative result.

The low-temperature transition in UCu_5 was also investigated by longitudinal magnetoresistance (ρ). Again, all the data presented in Fig. 3 were obtained while slowly warming up the sample. Similarly to the zero-field specific heat our resistivity results demonstrate some deviations from the zero-field resistivity published in Ref. 1. However, the main feature of the previously published study, a steplike increase of ρ at T_c, is also present in our $H = 0$ resistivity. The low-temperature value of ρ (for $T < T_c$) is a nonmonotonic function of H for $H < 10$ T, but decreases with H for $H \geq 10$ T. On the other hand, the ρ values above T_c are moderately enhanced by an application of magnetic fields, a behavior expected for an antiferromagnetically ordered system. There is a useful correlation between the magnetoresistance and the specific heat of our sample: the onset of the ρ increase coincides with the temperature of the C/T maximum, identified earlier as T_c (see Table I). This fact allowed us to verify the T_c versus H dependence derived from the specific-heat data.

Finally, we have checked the magnetic-field influence on the high-temperature transition in UCu_5. As can be seen from the inset of Fig. 3, the 10-T field has only a weak effect on the resistivity in the vicinity of 15 K. T_N is lowered by about 0.5 K by this field.

TABLE I. Magnetic-field dependence of the temperature corresponding to C/T maximum (T_c) and of the temperature onset of ρ increase (T_ρ). The absolute errors of T_c and T_ρ values are ±0.05 K. Data were collected while slowly warming up the sample.

H (T)	T_c (K)	T_ρ (K)
0	1.15	1.15
2	1.15	
5	1.4	
8	2.00	2.00
10	2.45	2.50
12	2.95	3.00
14	3.57	3.60

DISCUSSION

In the ongoing discussion on the origin of the 1-K anomaly in UCu_5, the most frequently asked questions are whether this transition is magnetic and how it is related to the ordering at 15 K. The neutron-diffraction work carried out by several groups [2,7] identified the magnetic structure below 15 K as type-2 antiferromagnetic with a single **q** vector. Magnetic moments in this structure are aligned ferromagnetically within (111) planes with antiferromagnetic coupling between planes with moments pointing normal to the plane. So far there has been no evidence within neutron scattering for any rearrangement of the moments below 1 K. Subsequent muon-spin-resonance[2] (μSR) and ^{63,65}Cu nuclear-magnetic-resonance[3] (NMR) measurements yield results not fully consistent with a single-**q** structure. It was noted in Refs. 3 and 4 that two other arrangements of magnetic moments, say 4-**q** and 4-**q**′, each characterized by four wave vectors, are also compatible with published neutron-diffraction data. Moreover, the 4-**q** structure interpretation agrees with both μSR and NMR results. However, the μSR data imply either lack of any change of the magnetic structure at 1K or a transition between 4-**q** and 4-**q**′ while NMR data are more consistent with a

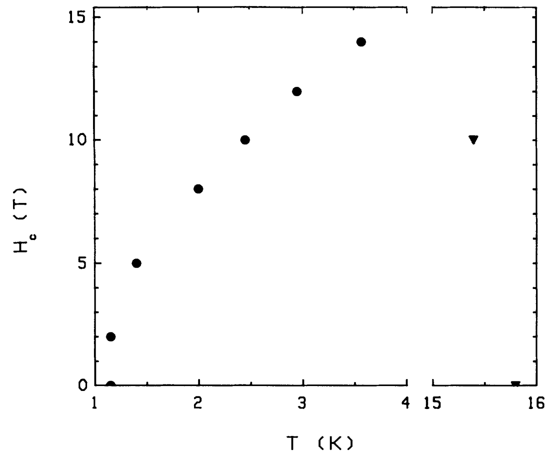

FIG. 4. Magnetic phase diagram of UCu_5 based on specific heat (●) and resistivity (▼) measurements.

4-**q** to 1-**q** rearrangement.

The magnetic phase diagram, drawn on the basis of our high-field specific-heat and electrical resistivity results (Fig. 4), certainly favors the second possibility. ($dH_c/dT>0$ necessarily implies $\delta M/\delta T|_{T=T_c}<0$, i.e., the low-temperature structure has a larger ferromagnetic component than the high-temperature structure.) This point requires further examination by theoretical and experimental work.

On the other hand, the insensitivity of the specific heat to a 2-T magnetic field, or $dH_c/dT=\infty$ for $H<2$ T, is very curious. According to the Clausius-Clapeyron equation: $dH_c/dT=-\Delta S/\Delta M$, this requires $\Delta M=0$, which is inconsistent with the first-order magnetic phase transition. Unfortunately, our experiment cannot distinguish between $dH_c/dT=\infty$ or $dH_c/dT>20$ T/K. Additional, high-resolution experiments are planned in the small-field regime. Our specific-heat data seem to imply no hysteretic effects in a field of 10 T, in contrast to zero-field results. This might imply a change in the order of the transition with a characteristic field in the interim. Further experiments will have to determine this characteristic field and whether it is related to the field in which dH_c/dT changes its slope.

The above thermodynamic equation allows us to set an upper limit on ΔM, provided that an upper boundary of $\Delta S(\Delta S_{max})$ is known. Since our method of specific-heat measurement (at discrete temperature points) is not optimized to detect any latent heat, ΔS_{max} was approximated from the specific-heat data; $|\Delta S|<400$ mJ/K mol yields $|\Delta M|<0.004\mu_B$/U atom.

A different interpretation of the 1-K anomaly was formulated in Ref. 2, i.e., a possibility of the coexistence of two independent magnetic subsystems at low temperatures, local spins that order antiferromagnetically at 15 K and heavy electrons that undergo a different kind of phase transition at 1 K. Such an interpretation also receives some support from our data. The two transitions respond very differently to large magnetic fields and sample preparation conditions. Although the 1-K anomaly is found for a well-annealed material only, the 15-K anomaly is found in even unannealed and off-stoichiometric specimens. All discussed experiments (specific heat, electrical resistivity, and NMR) suggest a formation of a gap in the electron excitation spectrum below T_c. This gap is partially filled up in high magnetic fields as seen by an increase of the low-temperature γ and a decrease of ρ for $H>10$ T.

The phase transition at $T_c(H)$ is sharper at high fields. This is similar to UCd_{11},[8] where the width of the transition is suspected to arise from the proximity of two transitions which separate at higher fields. The Hall effect and magnetoresistance data obtained for another UCu_5 sample[9] also give an evidence for the additional, magnetic-field-induced transition. (Interestingly, the virgin resistivity curve of Ref. 9, i.e., cooling in $H=0$, did not exhibit an increase of resistivity at 1 K, in disagreement with our data and Ref. 1.) These are important indications about the nature of the transition at $T_c(H)$. Our high-field measurements provide facts and observations that will have to be further explored. Our results show once again how sensitive the low-temperature state of UCu_5 is to the preparation conditions. A full accounting for the unusual properties of this compound requires comprehensive thermodynamic, transport, magnetic, spectroscopic, and structural measurements performed on a single specimen.

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