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The low-temperature phase diagram of UCu_5 determined from magnetization and magnetoresistance measurements

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Abstract. The low-temperature phase diagram of polycrystalline UCu_5 has been determined from magnetization and magnetoresistance measurements. This system undergoes two magnetic transitions at low temperatures. The lower-temperature phase transition at $T_2 = 1.16$ K, which manifests itself as a sudden jump in the magnetization and electrical resistance with decreasing temperature, shows a temperature hysteresis of about 40 mK for fields up to 12 T. The magnetic properties of this low-temperature phase show irreversible and metastable behaviour, which becomes more dramatic at the lower fields. The critical temperature T_2 is observed to increase proportionally with the square of the magnetic field. Our results are discussed in comparison with available specific heat, neutron scattering and nuclear magnetic resonance data, with the aim of determining the actual nature of the low-temperature magnetic phase transition.

1. Introduction

Since the heavy-fermion properties of UCu_5 were discovered, its unique character amongst other uranium compounds has been recognized [1, 2]. The low-temperature heavy-fermion ground state clearly develops into a rather conventional antiferromagnetically ordered magnetic phase below a second-order transition at $T_N \approx 16$ K. Both the heavy-fermion and the antiferromagnetic states coexist below the Néel temperature. From a neutron diffraction study using a polycrystalline sample of UCu_5, Murasik *et al* [3] identified the magnetic structure below 16 K as a simple type-II antiferromagnet with a single q vector of $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$. For this type of arrangement, the U magnetic moments are aligned ferromagnetically within the (111) planes of the cubic $AuBe_5$-type crystallographic structure, with antiferromagnetic coupling between parallel (111) sheets. The moments are aligned along the ⟨111⟩ direction. Murasik *et al* [3] estimated the magnitude of the ordered moment to be $0.9 \pm 0.1\mu_B$/U-atom, but the later studies of Schenck *et al* [2] yielded a significantly larger value of $1.55 \pm 0.05\mu_B$/U-atom at 2 K.

At $T_2 \approx 1.16$ K an additional first-order phase transition is observed‖. The origin of this transition has not yet been fully understood, and it appears to be rather dependent on sample stoichiometry and annealing procedures. Various explanations for this phase

‖ T_c has frequently been used to denote this transition, but we prefer to use T_2 since it is now clear that the transition does not involve ferromagnetic ordering.

transition have been suggested, including the onset of ferromagnetism [4], or a reordering of the antiferromagnetic structure [5]. However, Schenck *et al* [2] were unable to find any evidence of a change in the magnetic structure at T_2.

A very interesting proposal was put forward by Nakamura *et al* [5], who have interpreted their recent nuclear magnetic resonance (NMR) and muon spin resonance (μSR) experiments in terms of a 4-q magnetic structure below 16 K. Below 1 K, these NMR results are in accord with a rearrangement of the magnetic moments to a multi-domain [4] 1-q structure. The authors conclude that these 1-q and 4-q structures are the only possible magnetic structures for UCu_5, assuming the following conditions.

(i) The magnetic moment distribution can be written as

$$m(r) = \sum_i m_i \exp(2\pi i q_i \cdot r)$$

with the q_i the four characteristic vectors of the type-II magnetic structure:

$$q_1 = \left(\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{1}{2}\right) \qquad q_2 = \left(-\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{1}{2}\right) \qquad q_3 = \left(\tfrac{1}{2}, -\tfrac{1}{2}, \tfrac{1}{2}\right) \qquad q_4 = \left(\tfrac{1}{2}, \tfrac{1}{2}, -\tfrac{1}{2}\right).$$

(ii) Each magnetic ion has the same magnetic moment value, i.e.

$$\sum_{i=1}^{4} m_i^2 = m_0^2.$$

The 4-q structure for a type-II antiferromagnet was first proposed by Roth [6] and discussed by Hermann-Ronzaud *et al* [7]. The 1-q structure is the arrangement proposed by Murasik *et al* [3]. The 4-q structure has cubic symmetry ($m3m$), compared to the trigonal symmetry ($\bar{3}m$) of the 1-q arrangement. The NMR results are sensitive to this symmetry difference, whilst it is very difficult to distinguish, by neutron diffraction experiments, between a multi-domain single-q and a single-domain multiple-q structure [8]. The results obtained by Schenck *et al* [2] could be explained in this way.

It has also been proposed, mainly on the basis of specific heat and electrical resistivity measurements, that the low-temperature phase transition involves the opening of a gap at the Fermi surface, associated with the development of a spin-density wave (SDW) [1, 2]. The possibility of a change in the order of the low-temperature phase transition, from the first order suggested by the hysteresis in the specific heat, to second order for fields higher than 8 T, was also suggested in a recent paper [9]. UCu_5, and isostructural compounds such as UPt_5, have been examined in order to investigate how changes in chemical composition influence the heavy-electron ground state. The reasons are the simplicity of the $AuBe_5$ structure and the large U–U distances usually found in these materials. With doping on the copper sites, the low-temperature phase transition in UCu_5 fades away, and for some dopants, including Pd, the system develops anomalous transport and thermal properties. Some of these features are indicative of marginal Fermi liquid behaviour, possibly related to the multichannel Kondo effect, whilst others are related to spin-glass behaviour [10].

In this paper we present new detailed magnetization and resistivity/magnetoresistivity measurements on UCu_5, which provide precise information on the low-temperature phase transition and particularly its field dependence. The field dependence of the Néel transition has also been examined.

2. Experimental details

Polycrystalline ingots of UCu_5 were prepared in an arc melting furnace at the School of Metallurgy and Materials, University of Birmingham, using high-purity starting materials.

The buttons were remelted several times in order to improve sample homogeneity, and were subsequently annealed for 400 hours at 900 °C in Ta sealed tubes. Attempts to make single crystals of UCu_5 have, so far, been unsuccessful.

The magnetic moment as a function of temperature or field was measured, at Birkbeck College, using a vibrating-sample magnetometer (VSM) equipped with a top-loading ^{3}He cryostat, constructed by Oxford Instruments plc. Measurements were made in the temperature range 0.4 K–40 K and for fields up to 12 T. The cryostat is operated with the sample immersed in liquid ^{3}He for $T < 1.5$ K. For $T > 1.5$ K, the sample is cooled using ^{3}He as an exchange gas with the 1 K ^{4}He bath providing the heat sink. The magnetization (M) versus temperature (T) was measured at different constant fields, both in the zero-field-cooled (ZFC) and field-cooled (FC) régimes, in order to check the occurrence of irreversible effects and/or hysteretical behaviour below both magnetic transitions. The magnetization versus magnetic field (B) was measured at various fixed temperatures, in all cases ramping the field slowly upwards and then downwards.

The electrical resistance and longitudinal magnetoresistance of UCu_5 were measured at the University of Bristol. Studies were made in a dilution refrigerator, over the temperature range 0.027 K–1.6 K and in fields up to 13.5 T, using low-frequency ac techniques. Additional measurements, from 4.2 K to 300 K, were made at Birkbeck College in a conventional immersion cryostat, using dc methods.

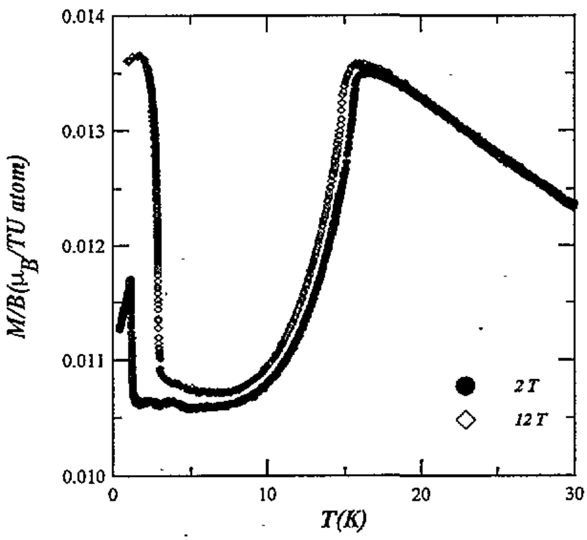

Figure 1. M/B versus T data for UCu_5 measured in fields of 2 T and 12 T. Only ZFC data are shown.

3. Results

Results from the magnetization measurements are shown as plots of M/B versus T in figures 1, 2 and 3 for various applied magnetic fields and temperature ranges. Figure 1 shows M/B versus T from 0.4 to 30 K for 2 and 12 T, showing the differing nature of the Néel and low-temperature transitions. The first-order character of the lower transition is clear. In agreement with earlier work, the Néel transition is only slightly shifted down (by $\approx$1 K) even in fields as large as 12 T (see figure 2). M/B versus T shows a thermal hysteresis of about 40 mK at the low-temperature transition (compared with 60 mK reported by Andraka *et al* [9]). However, in contrast, we observe hysteresis in all fields up to 12 T (see figure 3). Our results also indicate that the phase transition seems to broaden in high fields. At 1 T and 2 T, the ZFC data reveal a sharp peak very close to T_2, but this disappears with increasing field.

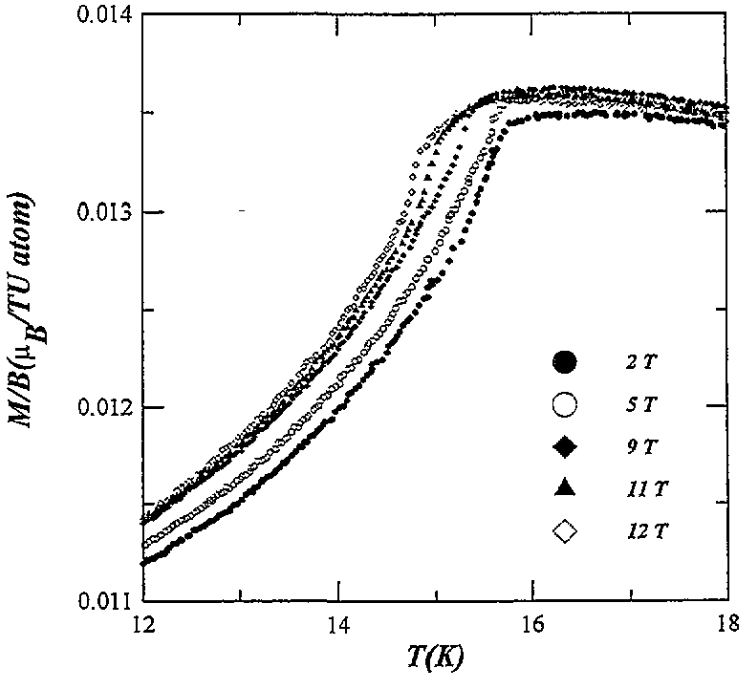

Figure 2. M/B versus T for different values of the magnetic field, close to the 16 K transition.

Another new feature was observed in the M/B versus T curves below the low-temperature transition and for $B \leqslant 9$ T: irreversibility effects are apparent from a comparison of the ZFC (irreversible) and the FC (reversible) data at temperatures below T_2. The observed effects steadily diminish with increasing field. For $B = 9$ T, the differences between the ZFC and FC results are of the same order of magnitude as the experimental error, and can no longer be resolved at 12 T. We also observed a time dependence of the ZFC moment below T_2. After cooling in zero field to 0.8 K, the field was ramped up to 2 T and held fixed. Over a time-scale of 10^4 seconds, the magnetization steadily increased, gradually approaching the value found in the field-cooled measurements.

Figure 3 illustrates the rapid increase of T_2 with magnetic field. We have defined T_2 as the onset of the jump in the magnetization versus temperature measurements, as indicated for the 6 T data in figure 3. Figure 4 shows the phase diagram deduced in this way from our results. We find a linear dependence of T_2 on B^2 (see the inset to figure 4).

The results obtained from measurements of the magnetization versus magnetic field at constant temperature are presented in figures 5 and 6. For $T > 1.16$ K, the magnetic-field-induced phase transition, hysteretic in field, is clearly revealed as a sudden change in slope (see the inset to figure 5) at a critical field consistent with the values obtained from the phase diagram constructed from the M/B versus T measurements. Below T_2, the M versus B curves show irreversible behaviour at low fields: the results for $T = 0.4$ K are presented in figure 6. The slope of the magnetization curve shows a change at $B \approx 2$ T for measurements

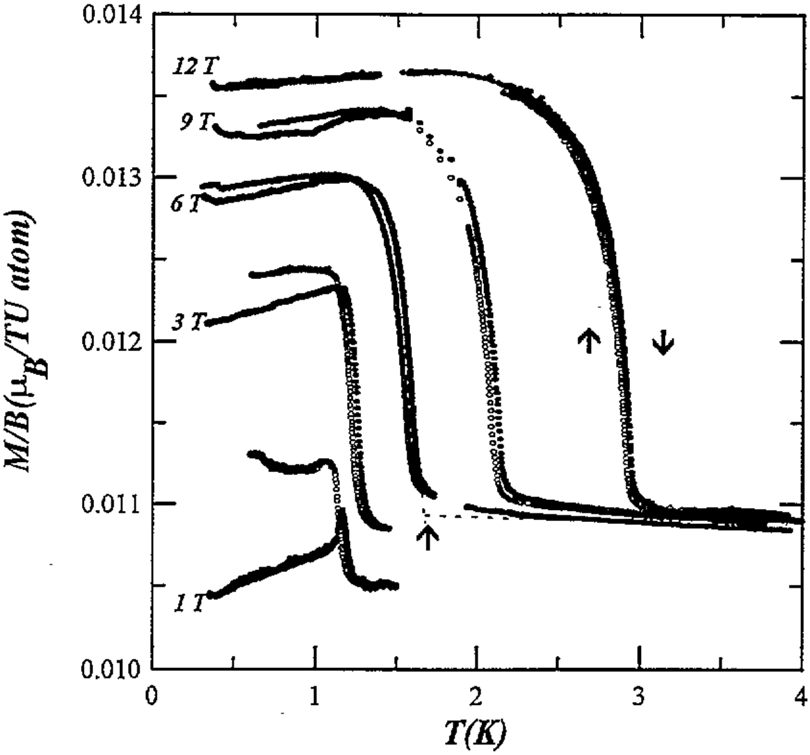

Figure 3. M/B versus T for different values of the magnetic field, showing the hysteresis between the ZFC (closed symbols) and FC (open symbols) data close to the low-temperature phase transition.

made in increasing field, but is fairly constant for the measurements performed with the field ramping downwards from 12 T. Both curves seem to coalesce for fields higher than $\approx$7 T. We note that this type of behaviour was not observed for $T_2 < T < T_N$.

In figures 7 to 9, we show results obtained from the electrical resistance measurements. Because of the irregular shape of our sample, the actual values of the resistance are given. We estimate that a resistance of 1 mΩ corresponds to a resistivity of $350 \pm 70\ 10^{-8}\ \Omega$ m. Both magnetic transitions produce significant effects in the resistance. The onset of the antiferromagnetic transition results in a sudden increase in the resistance, followed by a maximum at around 13 K, and a rapid decrease to 50% $R(T_N)$ at 4.2 K (see figure 7). Compared with previous references [1, 10–13] our results show a sharper peak and a more drastic drop of the electrical resistance. Below the second transition at 1.16 K, the electrical resistance increases by a factor of almost five (see figure 8). The size of this increase is in agreement with Ott *et al* [1], but almost twice that reported in [10–12]. Our data also display a maximum at around 0.4 K, in agreement with the recent results reported by Bernasconi *et al* [11]. Below this maximum, our experimental points, although few in number, are in accord with a T^2-dependence. Such a behaviour would be consistent with a coherent Fermi liquid state setting in at low temperatures.

An important new discovery concerns the time dependence of the resistance, when measured on cooling the sample. Figure 8 shows the first set of data, taken after cooling the sample to the base temperature of 27 mK and waiting for nine hours. The measurements were then taken whilst slowly warming the sample. In order to ensure thermal equilibrium,

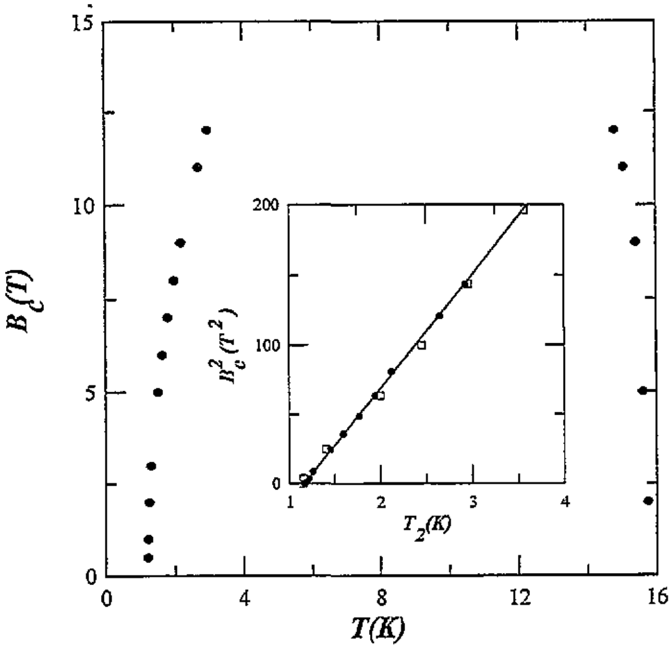

Figure 4. The magnetic phase diagram for UCu_5 determined from the field dependence of the transition temperatures. The inset shows a plot of B^2 versus T_2. We compare our results (closed circles) with those of Andraka *et al* [9], determined from specific heat studies (open squares).

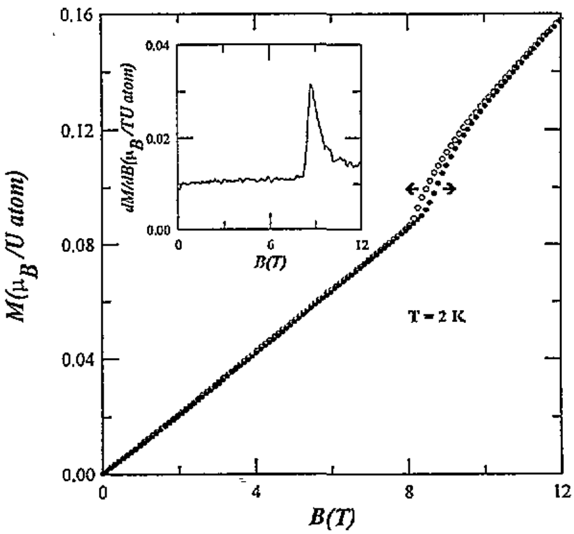

Figure 5. The magnetization of UCu_5, measured at $T = 2$ K, for fields ramping upwards (closed circles) and downwards (open circles), showing clear evidence of hysteresis. The inset shows the derivative of the magnetization.

the experimental points were recorded only after waiting for 30 minutes at each fixed temperature. After reaching 1.6 K, another data set was collected on cooling the sample. Below T_2, the resistance measured in this way was found to be time dependent. For example, figure 9 shows measurements of the resistance at 0.51 K as a function of time over a 16-

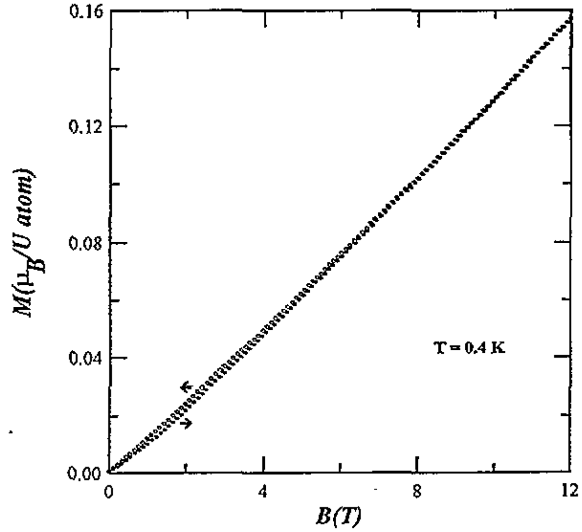

Figure 6. The magnetization of UCu_5, measured at $T = 0.4$ K, showing irreversibility at low fields.

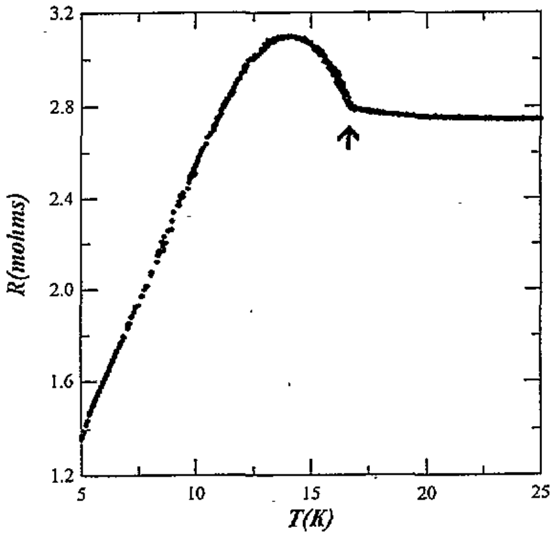

Figure 7. The electrical resistance of UCu_5, measured from 5 to 25 K.

hour period. These results follow closely an exponential behaviour with a time constant of 145 minutes. After the 16-hour waiting time, the sample was again slowly warmed and the resistance remeasured. As can be seen from figure 8, the results for the two heating runs are almost identical, reflecting the fact that they were performed after waiting times much larger than the time constant for the relaxation.

Longitudinal magnetoresistance versus field measurements in the phase below T_2 show a significant difference between the results obtained when ramping the field upwards and those for ramping it downwards (see figure 10); this is analogous to the behaviour of the magnetization in this phase. The magnetoresistance measured in increasing fields is not reversible below a characteristic field of approximately 8 T: the zero-field resistance

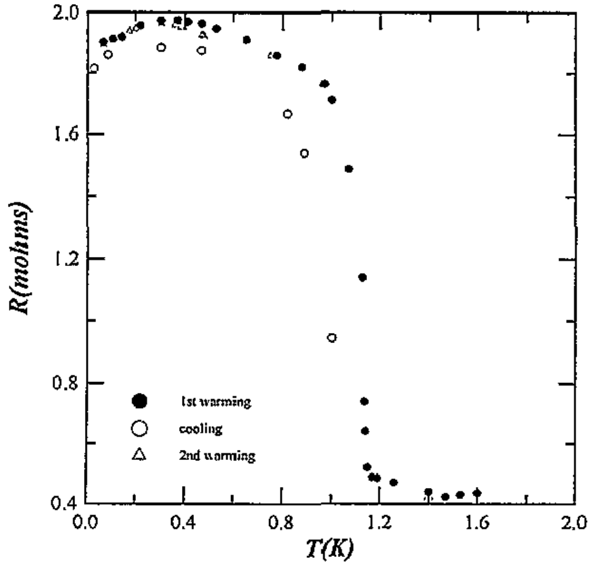

Figure 8. The resistance of UCu_5, measured on heating and cooling, as described in the text.

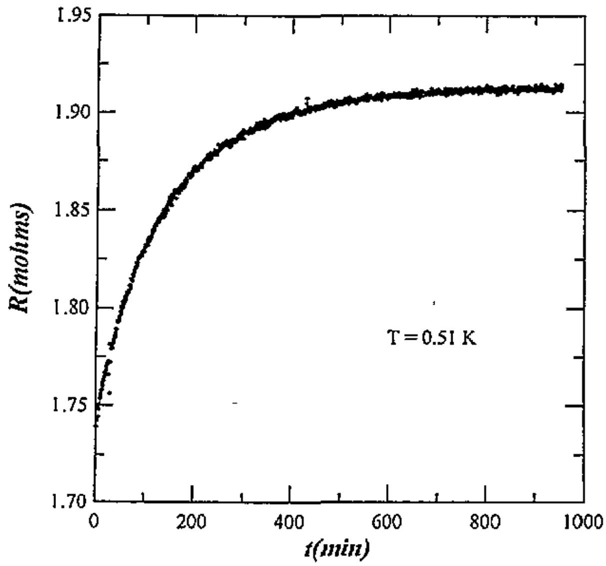

Figure 9. The time dependence of the electrical resistance of UCu_5, measured at $T = 0.51$ K after cooling from 100 K, as described in the text.

determined after ramping down the field is noticeably less than the original value. We also note from figure 10 that the magnetoresistance at $T = 27$ mK exhibits a maximum at around 2 T, and subsequently becomes large and negative (−75%). The general trend of our results is in partial agreement with previously published data [13, 14]. However, the magnetoresistance observed by us is approximately twice that reported by Ōnuki *et al* [14]. Moreover, they reported that the value of the residual magnetoresistance after ramping the field back to zero is positive, in contrast to the negative value obtained by us. The magnetoresistance appears to be approaching saturation at $B = 13.5$ T and $\Delta R/R \approx -75\%$.

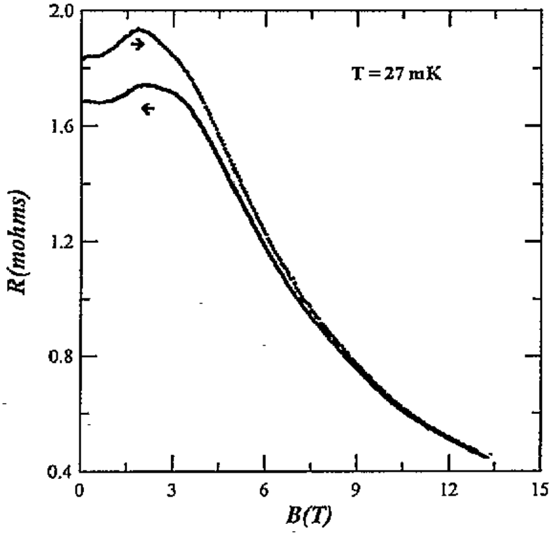

Figure 10. The magnetoresistance of UCu_5 measured at $T = 27$ mK, in increasing and decreasing fields.

The effect of a large applied field is thus to reduce the resistance to a value consistent with the zero-field resistance above T_2. This is in agreement with the observed reduction (70%) in the electronic contribution of the zero-field heat capacity at T_2.

4. Discussion

The nature of the magnetic ordering exhibited by UCu_5 below $T_2 = 1.16$ K is not fully understood. Even the order of the transition, suggested to be first order from the observed temperature hysteresis, has been proposed to change above a characteristic field, because of the apparent disappearance of the hysteresis and also the sudden observation of a lambda-like peak [9]. Although our results show clear hysteresis of about 40 mK up to 12 T, the shape of the transition changes from the very sharp jump with a clear peak superimposed, observed for fields below 3 T, to the initially very well defined, but substantially broadened transition, observed at higher fields. The observed increase in magnetization is in agreement with the entropy reduction below T_2 because, according to the Clausius–Clapeyron equation, $(\mathrm{d}B/\mathrm{d}T)_{T_2} = -\Delta S/\Delta M$. Hence T_2 shifting to higher temperatures implies $\Delta M > 0$. Thus the ΔM-values obtained for the different fields are also in agreement with Clausius–Clapeyron equation. For example, assuming $\Delta S \approx 400$ mJ K^{-1}, and $(\mathrm{d}B/\mathrm{d}T)_{T_2} \geqslant 20$ T K^{-1}, ΔM is estimated to be $<0.004\mu_B$/U-atom for fields lower than 2 T (see [9]). From our experimental results, we obtain $\Delta M \approx 0.0018\mu_B$/U-atom for 2 T and $\Delta M \approx 0.0007\mu_B$/U-atom for 1 T. This is a very small change in moment.

In the heavy-fermion antiferromagnet URu_2Si_2, the *second-order* phase transition at 17.5 K is also found [15] to shift linearly with B^2. The rare-earth element Nd is another example of an antiferromagnetic system showing a B^2-dependence of T_N, and in this case the transition is *first order* [16]. Nd shows a rich variety of magnetically ordered phases, arising from successive changes from multiple-q to single-q structures. The phase diagram and the *negative* B^2-dependence of the Néel temperature in Nd have been explained phenomenologically with a free-energy analysis [16]. This type of dependence is not uncommon in the framework of the Landau theory of phase transitions. As discussed in the introduction, the NMR and μSR experiments [5] strongly suggest that the 1.16 K transition involves a spin rearrangement from a 4-q structure below T_N to a single-q structure below T_2. The phase diagram of UCu_5 (see figure 4) is consistent with the hypothesis that the application of a magnetic field at a temperature $T_2 < T < T_N$ breaks the symmetry of the 4-q structure and leads to the development of a single-q phase.

In Nd, a transition from multi-domain single-q ($T < T_N$) to double-q structures ($T < T_2 < T_N$) is observed in zero field [16, 17], i.e. the transitions occur in the *opposite* sequence to those for UCu_5. However, the field dependence of T_2 is also reversed: in Nd a field suppresses T_2, rather than enhancing it as in UCu_5. *Thus in both systems, a magnetic field applied to the multiple-q structure favours the development of a single-q structure.* Following the analogy with Nd, we also draw attention to the similarity between the results obtained for the two systems in the magnetization measurements [18, 19] below T_2. This similarity reflects the fact that we are dealing in both cases with a magnetic-field-induced transition from multiple- to single-q magnetic structures.

The observed jump in electrical resistance at T_2 implies a drastic modification of the Fermi surface, more dramatic than the superzone scattering associated with antiferromagnetic transitions in rare-earth metals and compounds. The jump is also much larger than that observed in materials like Cr, which develops a weak first-order magnetic transition associated with the opening of a SDW gap [20]. The experimental evidence of temperature hysteresis in different properties is the primary evidence for the first-order nature of the 1.16 K transition in UCu_5. Such hysteresis, associated with supercooling and superheating effects, is not expected for a second-order transition. However, up to now it has not been possible to detect any latent heat at T_2. Moreover, the value of the temperature hysteresis appears to be sample dependent. In Cr, due to the extreme sensitivity of the SDW transition to pressure, defects and composition changes, this latent heat was detected only after major improvements in sample quality [20]. These considerations could be applied to UCu_5, in which different authors have observed an analogous dependence of the low-temperature phase transition on sample composition, annealing procedures and pressure. As we pointed out earlier, the order of magnitude of the high-field magnetoresistance (−75%) is in agreement with the observed reduction (70%) in the electronic contribution of the zero-field heat capacity at T_2, if we interpret this as the effect of removing a gap opened at the Fermi surface after applying a magnetic field.

Our results give additional experimental evidence pointing to the existence of a multi-domain metastable magnetically ordered state below T_2. This metastability is reflected in a time dependence of the electrical and magnetic properties, including the zero-field electrical resistance and the ZFC magnetization, and an irreversibility between the ZFC and FC magnetization and magnetoresistance. Similar effects are frequently observed in different kinds of magnetic material, ranging over ferromagnetic alloys, random spin systems, spin glasses, and SDW systems. However, our results can be understood in terms of the relaxation and pinning of the magnetic domains developing below the magnetic transition at T_2. In *our polycrystalline sample, the time-scale for the growth of the single-q structure domains* which have the lowest energy in an applied field is clearly of the order of 10^3 to 10^4 seconds. The magnetoresistance can be explained by a combination of domain reorientation at low fields and a progressive closing of the superzone energy gap in high fields.

The remaining problem is to establish quantitatively how the multiple-q to single-q model for the T_2 transition, which explains our magnetization measurements and the neutron diffraction, NMR and μSR studies, could explain the simultaneous opening of a gap at the Fermi level, which is the most plausible explanation for the thermal and transport properties. A good possibility would be a weak magnetoelastic distortion, responsible for the symmetry breaking below T_2 and driving the first-order transition. In rare-earth metals, magnetoelastic effects are sometimes responsible for driving magnetic phase transitions (as in Dy [21]), leading to the opening of superzone energy gaps at the Fermi surface which produce dramatic effects in the transport properties. It would appear that the energy gap in the low-temperature single-q phase must be significantly greater than in the 4-q phase. We hope that our experimental results will stimulate detailed theoretical calculations.

5. Conclusions

We have performed a detailed study of the magnetic properties of UCu_5 below its low-temperature magnetic transition. Our magnetization measurements as a function of field and temperature can be qualitatively explained in terms of a transition from a single-domain 4-q structure to a multi-domain 1-q structure below T_2. From the magnetic phase diagram, T_2 is observed to increase quadratically with magnetic field, indicating that a magnetic field favours the 1-q arrangement of the magnetic moments. The B^2-dependence indicates that the transition may be explained in terms of a mean-field theory. Below T_2, we observe irreversible behaviour and time dependence of the electrical resistivity and magnetic susceptibility. We explain these effects in terms of pinning and relaxation of domains associated with the 1-q structure. We suggest that magnetoelastic effects may be responsible for driving the first-order transition. A full resolution of this intriguing problem must await the successful growth of single crystals of UCu_5.

Acknowledgments

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