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High-field magnetization study of single-crystal URu_2Si_2

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Abstract. We report measurements of the c-axis magnetization of URu_2Si_2 single crystals, both unannealed and annealed, in fields up to 20 T. From our results, we conclude that the kink-type anomaly in the magnetization has a different high-field dependence from the dipole moment as seen in neutron diffraction experiments. Within the experimental error, we are unable to observe the effects of heat treatment on the field dependence. We discuss these new findings together with recent high-field results of resistivity and neutron diffraction experiments.

1. Introduction

Of all the heavy-fermion superconductors, URu_2Si_2 was the first shown to have both an antiferromagnetic ordering ($T_N = 17.5$ K) and subsequently a superconducting transition ($T_c = 1.5$ K) [1]. Unlike for the more recently discovered heavy-fermion superconductor UPd_2Al_3 [2], it is generally accepted that the same strongly correlated electrons participate in both the magnetic and superconducting transitions of URu_2Si_2. It is also one of the few uranium compounds to exhibit reasonably well defined magnetic excitations, which become broad with increasing temperature [3]. Although the superconductivity and its coexistence with the magnetic order have been subject to extensive theoretical and experimental study, the real mystery in URu_2Si_2 concerns the distinctly modest nature of the antiferromagnetic transition. Firstly, the ordered moment ($\mu_{ord} \approx 0.04\mu_B$/U-atom) is one of the smallest so far observed in uranium compounds [3]: the only other examples of such tiny moments are in UPt_3 [4], another heavy-fermion superconductor, and in UPd_3 [5] where the f electrons are well localized. Secondly, the transition temperature in URu_2Si_2 is rather high compared with those of the magnetic transitions in other uranium compounds. For example, UPd_2Al_3 does not order magnetically until $T_N = 14$ K, despite having an ordered moment of $\mu_{ord} \approx 0.8\mu_B$/U-atom: it is superconducting below $T_c = 2$ K [6].

However, what is most puzzling is that the entropy associated with the 17.5 K phase transition in URu_2Si_2, as measured by heat capacity studies, is about $0.2R \ln 2$, far too large to be attributed to the ordering due to the tiny moments alone. Several suggestions have been made to explain the unusual magnetic transition. One of them is that possibly quadrupolar interactions are the driving force behind the 17.5 K transition [7]. Although this approach

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is rather successful in explaining the observed susceptibility and heat capacity, there is not sufficient experimental evidence to support the idea that quadrupolar interactions drive the phase transition in URu_2Si_2. In this respect, it is very interesting to note that the order parameter of the 17.5 K transition breaks time-reversal invariance, indicating dipolar, or possibly octupolar, symmetry [8]. Therefore, it has been suggested that for a quadrupolar moment to be an order parameter, either the 17.5 K transition should be discontinuous or there must be two successive phase transitions [9]. However, there are no experimental results to support either of these two propositions. The existence of quadrupolar interactions in URu_2Si_2 has also been suggested independently by two groups [10] as an explanation for the anomaly in the non-linear susceptibility of URu_2Si_2.

In this paper, we report on our magnetization studies of single-crystal URu_2Si_2, which help to elucidate the unusual magnetic transition in this system.

2. Experimental details

Measurements have been made on two single crystals of URu_2Si_2; the dimensions of one is approximately $2 \times 2 \times 3$ mm^3 and the other is slightly smaller. They were grown by the tri-arc Czochralski method at Hokkaido University. In order to investigate possible annealing effects on the transition, one crystal was heated, in vacuum, at 900 °C for four days at Birkbeck College; the other crystal was left unannealed. We recall that Fåk *et al* found that the temperature dependence of the ordered moment varied with heat treatment [11]. Magnetization measurements up to 7 T were carried out at Birkbeck College in a Quantum Design MPMS7 SQUID magnetometer. Measurements in higher magnetic fields (up to 20 T) were made at the High Magnetic Field Laboratory of the CNRS/MPI in Grenoble, using an extraction magnetometer in a resistive magnet.

3. Experimental results and discussion

Figure 1 (left-hand panel) shows our results for the temperature dependence of the *c*-axis magnetization in a series of fields up to 7 T, plotted as M/H for ease of display. As can be seen in the figure, we found no appreciable difference in the field dependence of the transition temperature between the unannealed and the annealed samples. This is very surprising considering the neutron diffraction data of Fåk *et al* [11] mentioned in the previous section. They reported that the ordered moment of their well annealed sample showed a somewhat similar temperature behaviour to that already published. On the other hand, their unannealed sample seemed to have T_N lower (by 3.5 K) than 17.5 K, and had a plateau in the ordered moment versus temperature plot between 5 and 10 K before increasing rapidly at low temperatures to recover the value found in the annealed samples.

Before discussing our data further, we note that previous heat capacity measurements made by de Visser *et al* [12] and Fisher *et al* [13] in magnetic fields up to 7 T show no appreciable change between zero field and 7 T. This is confirmed in our low-field magnetization data which show only a small change in the transition temperature over this field range (see figure 1 (left-hand panel)). However, the measurements made in higher fields, shown in the right-hand panel of figure 1, clearly reveal a steady decrease in the transition temperature, which drops to around 14 K at 20 T. Indeed, the H^2-dependence of T_N seen before [14] is extended to at least 20 T. The field dependence of T_N is shown in

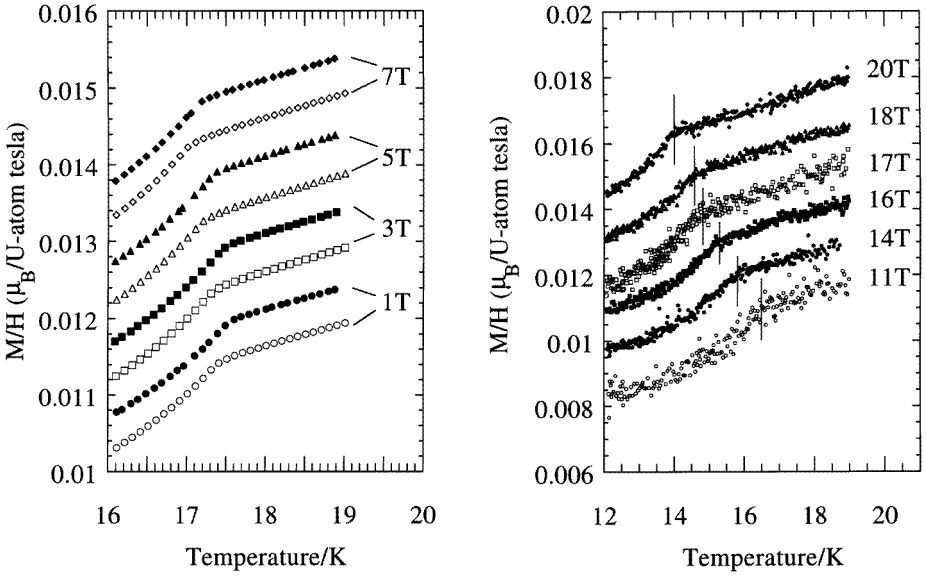

Figure 1. Magnetization, for fields applied along the *c*-axis, versus temperature for unannealed (closed symbols) and annealed (open symbols) samples of URu_2Si_2. For clarity of presentation, the data sets have been displaced vertically by a constant amount, and the low- and high-field data are shown separately.

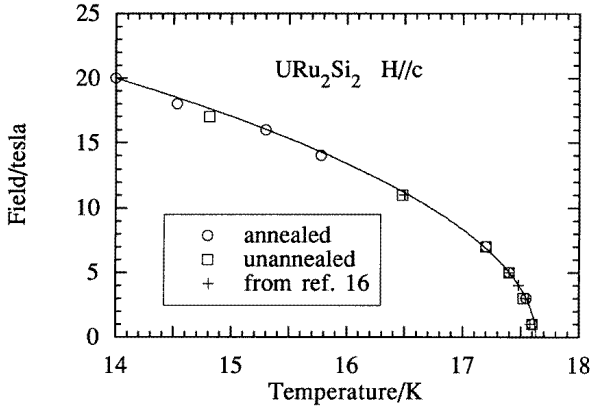

Figure 2. The magnetic field dependence of the transition temperature of URu_2Si_2 measured up to 20 T. Data up to 11 T from [16] have been reproduced here. The line shows the curve fitted using the equation given in the text.

figure 2, and our data are well fitted by the following equation:

$$T_N = T_N(0)\left\{1 - \left(\frac{H}{H_c}\right)^2\right\}$$

with $T_N(0) = 17.63$ K and $H_c = 44$ T.

The field of 44 T, which is predicted to drive the transition temperature to $T = 0$, is very close to the field at which Sugiyama *et al* [15] found metamagnetic transitions.

They observed a three-step metamagnetic transition between 36 and 40 T in the c-axis magnetization at 1.3 K. However, these metamagnetic transitions are almost temperature independent before becoming too broad to follow near 17.5 K.

The field dependence of the transition temperature should be compared with the neutron diffraction experiments in magnetic fields by Mason and co-workers [16]. They found that the measured antiferromagnetic diffraction peak is reduced to 77% of the zero-field moment at 7.5 T and by extrapolation is expected to disappear at around 15 T. Their results may be fitted to the equation

$$\mu = \mu(0)\left\{1 - \left(\frac{H}{H_c}\right)^{3/2}\right\}^{0.5}$$

with $H_c = 14.5$ T. This reveals that the ordered moment has a quite different field dependence to that of the transition temperature. We learned recently that Mentink *et al* [17] had found a similar field dependence of the transition temperature in their magnetoresistance measurements up to 25 T. Therefore the anomaly in the resistivity has the same origin as that in the susceptibility, but it is distinct from that of the antiferromagnetically ordered magnetic moments.

Whatever the origin of the anomalies in the resistivity and the susceptibility, both our results and those of Mentink *et al* show beyond doubt that there are at least two order parameters coexisting at zero field. Moreover, these order parameters may be distinguished by applying a magnetic field. If there were only a single order parameter, then the transition temperature and the antiferromagnetically ordered moment should have the same field dependence. As we have noted previously, one of the order parameters should be of dipole origin and disappears at around 15 T while the other survives even above 25 T. Hence, the transition temperature cannot be described by the same mechanism as the one for the ordered moment.

The two-order-parameter hypothesis then explains rather well why the thermal expansion coefficient [12] displays a sudden jump at the transition, whereas the dipole moment has a much slower temperature dependence [3, 11, 16, 18]. In fact, close inspection of the reported data suggests that the temperature dependence of the ordered moment observed in diffraction experiments differs from one sample to another even when they are annealed. Therefore we can argue that it is not the tiny magnetic moment of dipole origin but an order parameter, whose origin is yet to be identified, which is responsible for most of the anomalies seen around the transition.

Regarding the question of whether the order parameters are coupled to each other or not, it is appropriate to note that the unannealed sample in the studies of Fåk *et al* [11] appears to have a lower onset temperature than their annealed one. If that is indeed correct, then the fact that they appear at the same temperature in well annealed samples is likely to be only accidental. This may also explain why Ramirez *et al* [19] have found that different, but metallurgically identical, parts of their sample give rise to a double superconducting transition, whilst sectioning their original sample can lead to a single transition. It would be desirable to explore this behaviour further by detailed neutron diffraction studies. However, if the dipole moment transition is ever found to exist independently from the main anomaly, it will be intriguing that such a tiny moment can be sustained within an environment of thermal fluctuations.

In conclusion, we have found that the anomaly in magnetization has a different field dependence from the magnetic moment of dipole origin, and is independent of the heat treatment, unlike the moment itself. Combined with other results, this suggests that a new order parameter other than the dipole moment is responsible for most of the anomalies seen

at around 17.5 K. However, the origin of the other order parameter remains open, as does the question of whether the two order parameters exist independently or not.

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