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Anomalous low-field magnetic behaviour of polycrystalline URu_2Si_2

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Abstract. A low-field magnetization study of polycrystalline URu_2Si_2 reveals a magnetic structure with substantial irreversibility around $T = 30$ K, with applied fields $H \leqslant 50$ mT. Possible origins of such phenomena are discussed.

1. Introduction

Schlabitz and co-workers [1] first reported that the compound URu_2Si_2 (with a $ThCr_2Si_2$ structure) shows antiferromagnetic ordering below $T_N \sim 17$ K, which is followed by a superconducting transition around 1.5 K. Although URu_2Si_2 is now generally considered as a heavy-fermion (HF) system, many of its bulk properties are quite different from other uranium-based HF systems. It shows an enhanced electronic specific heat which is approximately linear in T just above $T_N \sim 17$ K and which corresponds to $\gamma \sim 100$–180 mJ mol^{-1}K^{-2} [2, 3]. Below T_N the heat capacity, apart from an exponential term which suggests the presence of a gapped spectrum, has a linear term $\gamma \sim 50$–65 mJ mol^{-1}K^{-2} [2, 3]. This latter γ value, although much larger than that of a normal metal, is nearly an order of magnitude smaller than those observed in other HF systems [4]. Neutron measurements [5] detected an antiferromagnetic ordering below T_N with an extremely small moment ($0.03\mu_B$). Given this small moment, the value of $T_N \sim 17$K and also the jump in the specific heat at T_N, ΔC, appear to be surprisingly large. The electrical resistivity (ρ) against temperature (T) behaviour of URu_2Si_2 is qualitatively similar to that of many hybridized f-electron rare-earth and actinide compounds [6], particularly the rapid decrease in ρ with decreasing temperature below ~ 50 K, which is presumed to be associated with the freezing-out of some sort of charge- and/or spin-disorder scattering. The antiferromagnetic transition is indicated by the distinct Cr-like anomaly around 17 K. Such an anomaly is expected for a charge-density wave (CDW) or spin-density wave (SDW) transition, or perhaps a structural transition. However, no structural transition has been detected in the vicinity of 17 K with x-ray diffraction measurements [3]. In comparison with the resistivity and specific heat, the signature of the antiferromagnetic transition in the bulk magnetic susceptibility is quite subtle. It is visible only as a change in slope around 17 K [1–3]. There is a broad maximum in the susceptibility around 50 K, which is associated with short-range magnetic fluctuations [1]. All these magnetic measurements were performed using an external field $H \geqslant 50$ mT. Here we would like to report our low-field ($H \leqslant 50$ mT) magnetization studies in URu_2Si_2, revealing some new features which, to our knowledge, have not been reported before. We believe that the present result, in conjunction with the recent μSR study [7] on URu_2Si_2 (to be described below), has implications for the normal-state properties of URu_2Si_2.

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2. Experimental details

The samples were prepared by argon arc melting from metals of at least nominal 99.99% purity and subsequently annealed at 800 °C for seven days. The samples were subjected to x-ray and metallographic analysis to investigate the possible presence of an impurity phase. The resistivity studies made on a sample from this batch revealed all the characteristic behaviour of URu_2Si_2 reported in the literature, and a specific heat study showed a sharp peak at 17.5 K [8]. Magnetization measurements to be reported here were performed using a commercial SQUID magnetometer (MPMS5, Quantum Design). We used a scan length of 4 cm, and for each measurement an average over three scans (each scan containing 32 data points) was taken. This relatively short scan length ensures that a relatively small field inhomogeneity is experienced by the sample during the measurement. The sample used here is in the shape of a square cross-section rod of dimension 5 mm × 1 mm × 1 mm. The sample was mounted firmly in the sample holder with its long axis parallel to the applied field, and the same sample position and configuration were strictly adhered to during all the measurements. This latter step is required to avoid any orientational effect due to anisotropy. Before starting each experiment care was taken to minimize the trapped field (which is about ±0.3–0.4 mT) in the superconducting magnet of our magnetometer. This was done by ramping down a high applied magnetic field to zero in an oscillatory mode, as recommended in the Quantum Design MPMS5 instruction manual.

3. Results and discussion

In figure 1 we present magnetization (M) against temperature (T) plots of URu_2Si_2 in external fields $H = 2$ mT, 4 mT, 10 mT, 50 mT and 0.5 T in both the zero-field-cooled (ZFC) and field-cooled (FC) conditions. Data also exist for $H = 1$ mT, 6 mT, 8 mT, 20 mT, 30 mT and 0.1 T, but are not shown here for the sake of clarity. The M against T plot with $H = 0.5$ T (see figure 1 (bottom right)) is very similar to what has been reported earlier [3]. (Although Maple and co-workers [3] reported the results of magnetic susceptibility, this should be similar to the magnetization behaviour because M against H is linear in URu_2Si_2 up to at least 30 T [9]. It is to be noted here that non-linearity in magnetization in a subtle form has been observed recently around T_N [10].) Also there is no difference at all between the ZFC magnetization (M_{ZFC}) and the FC magnetization (M_{FC}). In comparison, the $H = 2$ mT plot (see figure 1 (top left)) is drastically different. From the M_{ZFC} against T plot it appears that a ferromagnetic-like transition is taking place around 30 K, and at 17 K there is a sharp change in the slope of the magnetization. The FC magnetization shows a sharp structure around 17 K, which is also quite different from the behaviour in $H = 0.5$ T. However, there is no transition observed in the FC data at 30 K. In fact, the ZFC and FC data match perfectly above 30 K, and a pronounced irreversibility sets in only below that temperature. With an increase in H, the FC magnetization quickly acquires the well known shape [1, 3] of M against T plots (see figure 1 (top right)) but the behaviour of M_{ZFC} remains quite different. With a further increase in field, M_{ZFC} gradually approaches the FC behaviour and irreversibility decreases, but it takes a field of 0.1 T to erase the irreversibility completely.

It is known from the single-crystal study of URu_2Si_2 [2] that the easy axis of magnetization is the c axis, while magnetization perpendicular to the c axis is practically independent of temperature. Since our sample is polycrystalline, it is expected that the easy axis will not be aligned with the applied field direction, and hence there will be a longitudinal

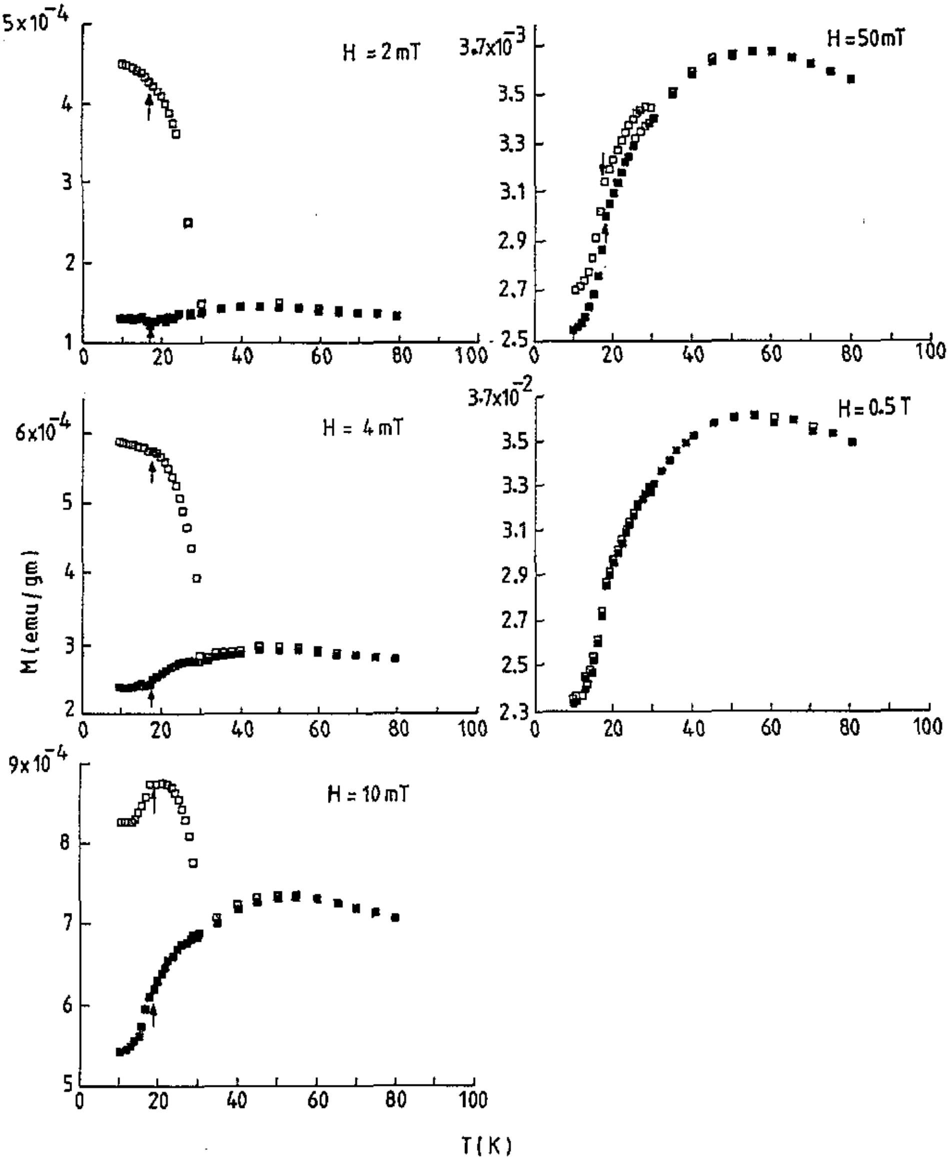

Figure 1. Magnetization (M) against temperature (T) plots for URu_2Si_2 in various external magnetic fields (H). The open squares represent zero-field-cooled (ZFC) magnetization and the full squares represent field-cooled (FC) magnetization. Above $T \sim 30$ K, for $H = 2, 4, 10$ and 50 mT and at all temperatures for $H = 0.5$ T there is no difference between ZFC and FC magnetization, and the respective symbols merge.

as well as a transverse component (with respect to the applied field) of magnetization. In our SQUID magnetometer we have the facility to study magnetization, with a transverse SQUID detector. Our preliminary study of the transverse SQUID response in URu_2Si_2 indicates that with fields $H \leqslant 0.2$ mT, only a longitudinal component of magnetization exists. It is as if in this field regime anisotropy has no role to play, and the magnetization is easily aligned with the applied field. With an increase in field, the transverse response gradually starts appearing and with $H = 10\,\mathrm{mT}$ one observes the expected transverse response. Such a behaviour was found only with the ZFC samples in the temperature regime below 35 K. Magnetization below 35 K in the transverse SQUID configuration also shows pronounced non-linearity in the field regime $0.1\,\mathrm{mT} < H < 50\,\mathrm{mT}$, while preliminary studies do not reveal such non-linearity with the standard longitudinal configuration. A detailed study in this regard is in progress.

Our studies indicate the existence of an interesting field-dependent magnetic structure around $T \sim 30\,\mathrm{K}$. The question now arises: what is the origin of such a magnetic structure? One possibility is that a small amount (about 1%) of impurity can always be present in the sample and go undetected in the x-ray study. Such a small impurity phase containing U is capable of carrying an effective moment of about $0.1\mu_B$, and can dominate the paramagnetic response of $\mathrm{URu_2Si_2}$ in the low-field regime $H \leqslant 50\,\mathrm{mT}$. (However, a simple ferromagnetic ordering of this impurity phase will not explain the observed magnetic structure with strong thermomagnetic irreversibility). This interpretation in terms of the contribution from a small magnetic impurity phase precipitate seems extremely unlikely in the light of recent μSR experiments. (This latter microscopic measurement presumably is not sensitive to such a small impurity precipitate.) μSR studies involving Knight shift measurements revealed the appearance of a small isotropic K O below 35 K. This is indicative of the availability of f-electron states which can be polarized by the external field, leading to field-induced moments at the U sites [11]. On the other hand, there is a marked change in the relaxation rate at 17.5 K (which seems to have set in already at temperatures above 17.5 K), which can be related to the establishment of small-moment antiferromagnetic order. It was suggested that the small-moment magnetic ordering below 17.5 K in $\mathrm{URu_2Si_2}$ is embedded in a strongly paramagnetic background [11]. Stronger support regarding the possibility of a magnetic ordering in $\mathrm{URu_2Si_2}$ around 30 K comes from a very recent μSR study by Knetsch and co-workers [7]. They have observed the presence of a weak ferromagnetic signal around 30 K in aged samples of $\mathrm{URu_2Si_2}$. (We would here like to mention that our present sample is also more than a year old). It is to be noted that in the same temperature regime a long-range ferromagnetic order could be established in $\mathrm{URu_2Si_2}$, on doping with Mn, Re and Tc at the Ru sites [12]. The results of μSR measurements in conjunction with our low-field magnetization study thus suggest the presence of a magnetic ordering in $\mathrm{URu_2Si_2}$ around 30 K, and this ordering can be ascribed to a single metallurgical state.

We shall now discuss whether any hints of such a structure exist within the already available experimental results on $\mathrm{URu_2Si_2}$. In specific heat measurements a broad maximum in the electronic specific heat (which was thought to be reminiscent of a Schottky anomaly) was indeed observed at about 30 K [1]. This feature has often been taken as support for the importance of a crystal field effect in $\mathrm{URu_2Si_2}$ [13]. Even in the neutron measurements of a good-quality single crystal of $\mathrm{URu_2Si_2}$, the presence of elastic magnetic scattering has been reported above 17.5 K [13]. This behaviour was tentatively associated with the stacking faults which might cause the relative orientation of the adjacent (101) ferromagnetic planes parallel or antiparallel at random. If a stacking fault is present in a good-quality single crystal, it is to be expected that our polycrystalline sample, as well as that of Schlabitz and co-workers [1], will contain similar stacking faults as well. (The importance of stacking faults in the study of HF physics has been pointed out recently by Smith [14]). Is it then possible that the metallurgical defects control the zero- and low-field properties of $\mathrm{URu_2Si_2}$ giving rise to the maximum in the low-temperature specific heat around 30K [1], and the interesting magnetic behaviour around 30 K reported in the present communication? If so, one need not then invoke the idea of crystal-field effects to explain various normal-state properties of $\mathrm{URu_2Si_2}$. On the other hand, the importance of anisotropy in U 1–

2–2 compounds with $ThCr_2Si_2$ is now well known [15]. Field anisotropy can also arise from an additional term in the RKKY interaction which is of the Dzyaloshinski–Moriya (DM) type having its origin in the spin–orbit interaction [16]. Weak ferromagnetism (or canted antiferromagnetism) can occur from a combination of RKKY and DM interactions in certain crystal symmetries, for example 2-Fe_2O_3, MnSi and $CrFe_3$ [17, 18]. Whether the $ThCr_2Si_2$ structure meets the required condition is not known to the present authors. In some quarters [16, 19] it is also believed that the presence of such DM anisotropy is essential for the occurrence of macroscopic irreversibility in metallic spin-glasses.

Regarding the splitting of low-field ZFC and FC magnetization below 35 K and the signature of the magnetic transition in the ZFC (but not in the FC) magnetization of URu_2Si_2, there is an uncanny resemblance to the diluted antiferromagnet $(Fe_{0.46}Zn_{0.54})F_2$ [20]. This latter system, which is a representative of random-field Ising systems, shows very similar behaviour around 25 K. Although anisotropy is an important feature in both systems, based on the present study alone it is too premature to conjecture about a deeper connection (if any) with underlying physics. (It should be noted here that long-range ferromagnetic order, in general, is not expected in random-field Ising systems [21].)

4. Conclusions

In conclusion, we would like to say that we have observed interesting structure, around 30 K, in a low-field magnetization study of polycrystalline URu_2Si_2, which is accompanied by thermomagnetic hysteresis. This, along with the μSR result of Knetsch and co-workers [7], is indicative of a magnetic phase transition which, even if it turns out to be of metallurgical origin (i.e. due to stacking faults), is likely to have interesting implications for the existing puzzle concerning the normal-state properties of URu_2Si_2. Our results will also then indicate that we can have a system where a combination of anisotropy and intrinsic disorder, such as stacking faults, can give rise to magnetic behaviour very similar to spin-glasses and/or random-field Ising systems. Such a system is simpler than the random magnetic alloys, which are the traditional hunting grounds for spin-glass-like properties.

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