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Magnetic transitions in a heavy-fermion antiferromagnet U₂Zn₁₇ at high pressure

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Abstract

Pressure-dependent electrical resistivity and ac-calorimetry measurements on single crystals of the heavy-fermion antiferromagnet U₂Zn₁₇ at pressures to 5.5 GPa reveal that the low temperature magnetic order changes above ∼3 GPa. The Neél temperature ($T_{\text{N}} = 9.7$ K at ambient pressure) decreases slowly for pressures below 3 GPa, but above this pressure a new magnetic state develops at $T_{\text{M}} \approx 8.7$ K. This magnetic phase becomes more stable with pressure increase ($dT_{\text{M}}/dP = 1$ K GPa^{-1}, $T_{\text{M}} = 10.5$ K at 5.3 GPa). The heavy-electron state in U₂Zn₁₇ is robust against pressure—the electronic specific heat coefficient $\gamma \approx 0.4$ J mol^{-1} K^{-2} is nearly pressure independent for both magnetic phases. Magnetic ac-susceptibility measurements show that the pressure-induced state is not ferromagnetic.

1. Introduction

Among uranium compounds, U₂Zn₁₇ has attracted attention due to its unusual physical properties. It orders antiferromagnetically from a heavy-fermion state, exhibiting a very large electronic specific heat coefficient $\gamma_{\text{el}}(T_{\text{N}}) \sim 1$ J mol^{-1} K^{-2} just above $T_{\text{N}} = 9.7$ K [1–3]. Magnetic order does not completely destroy the heavy-electron state as indicated by a zero-temperature specific heat coefficient $\gamma_{\text{el}}(0) \approx 0.4$ J mol^{-1} K^{-2}. Neutron diffraction [4] finds that the ordered moment $\mu_0 \approx 0.8$ μ_{B}/U forms a simple antiferromagnetic structure, with the magnetic unit cell identical to the nuclear unit cell. This ordered moment is much smaller than the effective paramagnetic moment $\mu_{\text{eff}} = 2.25$ μ_{B}/U found from magnetic susceptibility. The ratio $(\mu_{\text{eff}} - \mu_0)^2/\mu_{\text{eff}}^2$ is equal to $\gamma_{\text{el}}(0)/\gamma_{\text{el}}(T_{\text{N}})$, as if U₂Zn₁₇ were an itinerant antiferromagnet where a gap develops over a fraction of $(1 - \gamma_{\text{el}}(0)/\gamma_{\text{el}}(T_{\text{N}}))$ of the whole Fermi surface [4]. Inelastic neutron scattering reveals spin fluctuations which persist well above the Neél temperature [5]. Analysis of these data supports a model in which U-moments with Kondo-impurity-type dynamics couple to their nearest neighbors via an effective Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction. The magnetic transition at T_{N} appears to be driven by a modification of the effective RKKY coupling with temperature [5]. On the other hand, a more complex picture of magnetic interactions below T_{N} was obtained from μ^+ SR measurements which concluded that only about half of the U 5f moments in U₂Zn₁₇ produce a dipole field compatible with the simple AF order deduced from neutron diffraction studies [6, 7]. Some of the remaining U-moments participate in a complex order below T_{N}, with a broad distribution of static internal fields; non-ordered U-moments are paramagnetic below T_{N} [7].

The magnetic ground state of U₂Zn₁₇ has been probed and modified by application of hydrostatic pressure and by introducing impurities. Even a tiny amount of impurities on either U or Zn sites rapidly suppresses magnetic order [8]. Hydrostatic pressure to 1.7 GPa, on the other hand, produces little change in the electrical resistivity and Neél temperature: the room temperature resistivity exhibits a moderate increase with pressure, and the Neél temperature initially increases slowly ($dT_{\text{N}}/dP = 0.17$ K GPa^{-1}) before saturating with $dT_{\text{N}}/dP \sim 0$ at 1.5 GPa [9]. Further pressure studies of the electrical resistivity of U₂Zn₁₇ are needed to see whether the saturation of $T_{\text{N}}(P)$ at ∼1.5 GPa is a precursor to the eventual suppression of T_{N} at higher pressures as well as to search for a possible pressure-induced change of magnetic ground state. There also have been no pressure-dependent calorimetry studies to explore the evolution of the heavy-fermion state of U₂Zn₁₇.

2. Experiment

Motivated by these needs, we have performed electrical resistivity and ac-calorimetry measurements on U_2Zn_{17} at hydrostatic pressures to 5.5 GPa generated in a miniature clamped toroid-type anvil cell [10, 11]. Single crystals were synthesized, as described earlier, by slow cooling of the melt of U and Zn components [5]. Crystals from the same growth were used for resistivity and calorimetry. A crystal, along with a piece of Pb whose superconducting transition temperature served as a measure of the pressure, was placed in a Teflon capsule filled with glycerol–water pressure-transmitting liquid, which solidifies at room temperature near 5.3 GPa and near 180 K at normal pressure. The cell assembly was cooled slowly ($\leqslant 1$ K min^{-1}) from room temperature to 1.1 K, but in the vicinity of a phase transition, the cooling rate was lower by about two orders of magnitude. The width of superconducting transition of Pb did not exceed 10 mK at the highest pressure, an indication of good hydrostatic conditions. Four-point resistivity measurements were made with an LR-700 resistance bridge. For ac-calorimetry measurements, discussed in more detail below, a flat zig-zag heater made of constantan wire ($R \sim 5$ Ω) was glued to one side of a plate-like crystal ($\sim 1 \times 1 \times 0.2$ mm^3; 1.3 mg) and electrically isolated from the sample by a thin layer of GE-7031 varnish. By measuring the heater resistance and ac-current through it, provided by the output from an SR830 lock-in amplifier, we determined the ac-power input to the sample. The amplitude of resulting temperature oscillations (($\Delta T_{ac})^{-1}$ proportional to the specific heat) of the sample was measured with a (Au0.07Fe)–chromel thermocouple, micro-spot-welded to the surface of the sample opposite the heater. The thermocouple voltage, amplified by a factor of 500 by an SR554 transformer, was detected with the SR830 lock-in amplifier at twice the frequency of the output current. The frequency of the ac-power input was varied on cooling to maintain quasi-adiabatic conditions, needed for correct ac-calorimetry measurements [12–14].

Resistivity measurements at high pressures are relatively straightforward, but this is not the case for calorimetry. Absolute measurements of the specific heat by thermal relaxation calorimetry are possible to ∼1.5 GPa in a small clamped cylinder–piston cell [15, 16]. In this method, the specific heat of the pressure cell assembly, pressure medium and sample is measured. The contribution from the pressure cell alone is typically more than 80% of the total specific heat except near a magnetic transition, where the sample specific heat is a large fraction. An alternative is ac-calorimetry, which avoids the large contribution from the pressure cell but is complicated by an unknown contribution from the pressure-transmitting medium [17–21]. As a result, the specific heat usually is reported in 'arbitrary units'. In our case, the most obvious manifestation of a contribution from the pressure medium is a decrease in specific heat at the glass transition of the glycerol–water mixture. By comparing the drop in $C(T)$ at the glass transition in the liquid surrounding the sample with that observed in independent pressure measurements of $C(T)$ of the pressure medium alone in the cell, we conclude that pressure-transmitting medium contributes at least 50%
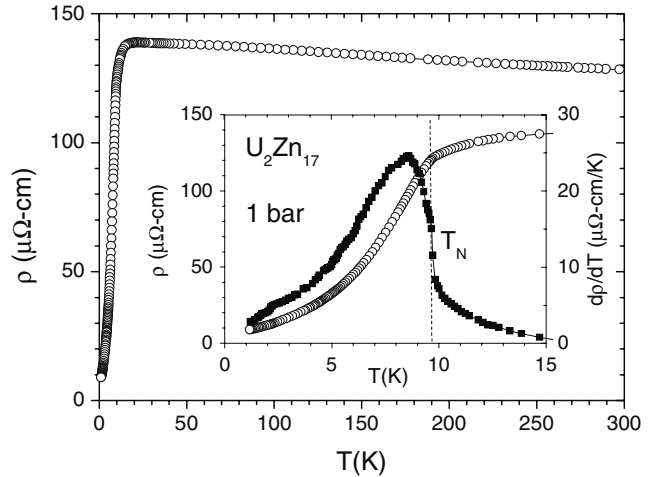

Figure 1. Electrical resistivity of U_2Zn_{17} at atmospheric pressure showing a pronounced drop of resistivity at the magnetic ordering temperature T_N. The inset compares the temperature derivative of the resistivity to the Néel temperature (vertical dashed line) determined by adiabatic specific heat.

to the total measured specific heat near the glass transition temperature. To estimate this contribution in the most important low temperature region, additional experiments were performed to map the temperature and pressure dependences of the specific heat of the pressure medium. From these studies, we found that at low temperature about 2 mg of the glycerol–water mixture contributed to the measured specific heat. This is about 40% of the total amount of a pressure-transmitting liquid in the Teflon capsule. With this information, it was possible to compare the specific heat of U_2Zn_{17} determined by ac-calorimetry at low pressures (0.05 GPa) to that obtained at atmospheric pressure by adiabatic techniques. Quantitative agreement between these two techniques gives confidence that we have established the pressure evolution of the absolute value of the specific heat of U_2Zn_{17} below ∼10 K by ac-calorimetry.

3. Results and discussion

Measurements of the electrical resistivity at atmospheric pressure (figure 1) show the behavior observed previously [1, 22].

The electrical resistivity passes through the broad maximum around 20 K and then exhibits a pronounced drop below the magnetic ordering temperature T_N. The shape of $d\rho/dT$ near T_N is depicted in the inset of figure 1. The dashed line in the inset, denoting T_N, corresponds to the peak in adiabatic specific heat. We see that T_N does not correspond to the maximum of $d\rho/dT$, but rather to the point of maximum rate of change of $d\rho/dT$. A similar shape of $d\rho/dT$ near T_N was observed earlier by Siegrist *et al* [22]. At high pressure, the temperature evolution of $\rho(T)$ depicted in figure 1 changes very little. In the temperature range between 20 and 300 K, $\rho(T)$ slowly shifts up with the increasing pressure, reaching 155 $\mu\Omega$ cm at 5 GPa at room temperature. The shape of $\rho(T)$ in the vicinity of magnetic transition shown in figure 1 also does not change significantly. To construct the P–T magnetic
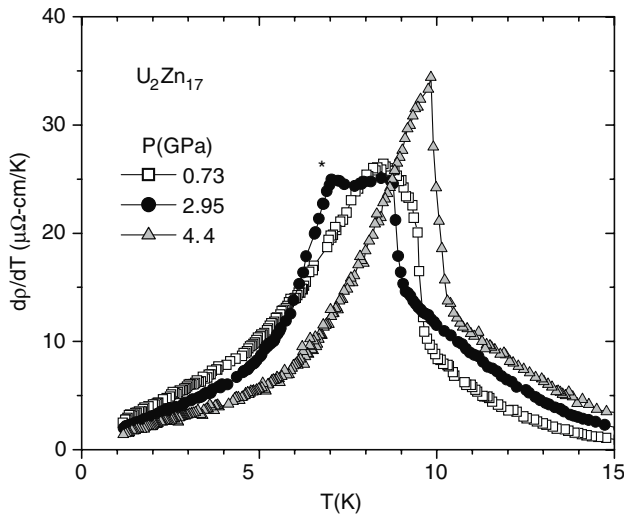
Figure 2. Dependences of $d\rho/dT$ of U_2Zn_{17} on temperature near its magnetic transitions at representative pressures. The asterisk ($*$) indicates the transition between two magnetic phases at 2.95 GPa.
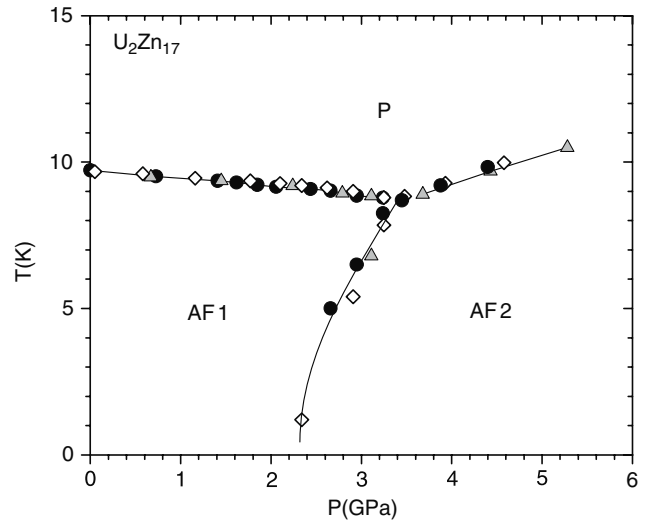

Figure 3. Magnetic pressure–temperature phase diagram of U_2Zn_{17}. The results of two resistivity experiments are shown by triangles and circles and the results of ac-calorimetry measurements are labeled by open diamonds.

diagram of U_2Zn_{17}, we used these temperature-dependent electrical resistivity data and their temperature derivatives. Representative plots of $d\rho/dT$ are depicted in figure 2 at a few pressures. Defining T_N as the temperature of maximum rate of change of $d\rho/dT$, then at low pressures $dT_N/dP \sim 0$; further increasing pressures to $\sim$3 GPa slightly shifts the Neél point down in temperature.

In the range 2.64–3.24 GPa, we observe a double peak structure in the derivative plots $d\rho/dT$, indicating the existence of two magnetic transitions in this pressure range. At pressures above 3.24 GPa, the shape of $d\rho/dT$ curves changes. A sharp peak appears (for example, the curve at 4.4 GPa in figure 2) instead of rounded one (the curve at atmospheric pressure in figure 1 and the curve at 0.73 GPa in figure 2).

The position of this peak T_M coincides closely with the temperature of maximum rate of change of $d\rho/dT$. This peak shifts to higher temperatures with increasing pressure above 3.24 GPa ($dT_M/dP = 1$ K GPa^{-1}). These observations indicate a change in the magnetic ground state of U_2Zn_{17} at $P \sim 3.0$ GPa. The magnetic P–T diagram of U_2Zn_{17} obtained from these resistivity measurements at high pressures is depicted in figure 3.

Magnetic transitions found by ac-calorimetry also are shown in the diagram and agree very well with those determined by resistivity. An example of two transitions in specific heat is illustrated by the curve at 3.25 GPa in figure 4. In the ac-calorimetry measurements, we observe two magnetic transitions in the pressure range 2.64–3.25 GPa; the upper one is from a paramagnetic to antiferromagnetic state and the lower one from the AF-phase to a new M-phase. To probe the nature of the high pressure M-phase (antiferromagnetic or ferromagnetic), ac-susceptibility measurements was performed in a separate experiment. No ferromagnetic signal was detected, and thus we conclude that the new M-phase is antiferromagnetic.

The specific heat of U_2Zn_{17} at three representative pressures before and after subtraction of the addenda
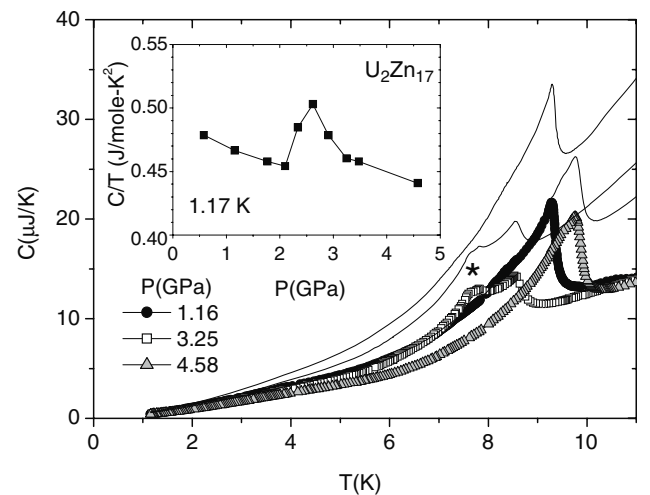

Figure 4. The specific heat of U_2Zn_{17} at three pressures (symbols). The asterisk ($*$) labels the transition between two magnetic phases at 3.25 GPa. Fine lines show the specific heat of U_2Zn_{17} before subtraction of addenda. The inset shows the pressure dependence of $\gamma = C/T$ at 1.17 K.

contribution from the pressure-transmitting liquid are shown in figure 4 as lines and symbols, respectively.

This comparison shows that at the lowest temperature the specific heat is determined by heavy electrons of U_2Zn_{17}. This contribution is dominant in both magnetic phases at low temperatures, but near 10 K an appreciable amount of the total specific heat is due to a contribution from the pressure-transmitting medium. The lines in figure 4 show a large pressure-induced variation in total specific heat above the magnetic transition. After subtracting the addenda contribution from the pressure-transmitting medium, however, the specific heat of U_2Zn_{17} just above T_N is nearly the same at different pressures (symbols in figure 4). The electronic specific heat

coefficient obtained at different pressures, after subtracting the addenda contribution, is plotted in the inset in figure 4. These values of C/T well below T_N decrease slowly with pressure but exhibit an anomaly near 2.4 GPa where the magnetic ground state changes. The value of $C/T = \gamma \sim 0.4$ J mol^{-1} K^{-2} is close to the value determined at atmospheric pressure in the earlier studies [1–3]. The magnetic entropy associated with magnetic ordering also does not change appreciably at high pressures. It is estimated from our ac-calorimetry measurements by integrating C/T up to the transition temperature as 10.3, 9.4 and 9.9 J mol^{-1} K^{-1} for pressures 1.16, 3.25 and 4.58 GPa respectively (figure 4). These values can be compared with the normal pressure value 8.5 J mol^{-1} K^{-1} obtained by integrating C/T data of [3] (relaxation calorimetry).

4. Conclusions

In conclusion, electrical resistivity, ac-calorimetry and ac-susceptibility measurements on the heavy-fermion antiferromagnet U$_2$Zn$_{17}$ at pressures to 5.4 GPa and temperatures down to 1.1 K have allowed the construction of the magnetic P–T diagram of this compound. Magnetic order is not suppressed in this pressure range; instead, a new magnetic state appears near 3.0 GPa and its ordering temperature increases weakly with pressure. The high pressure magnetic phase is a heavy-fermion antiferromagnet with an electronic specific heat coefficient $\gamma \approx 0.4$ J mol^{-1} K^{-2}, which is close to that of U$_2$Zn$_{17}$ at atmospheric pressure.

Acknowledgments

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