Gap formation and magnetic ordering in URu_2Si_2 probed by high-field magnetoresistance

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We examine the phase diagram of the antiferromagnetic heavy-fermion superconductor URu_2Si_2 with resistivity experiments in continuous magnetic fields up to 25 T. The phase transition that occurs in zero field at $T_N=T_0=17.5$ K is accompanied by the formation of a tiny staggered magnetic moment of $\mu=0.04\ \mu_B$. The resistivity anomaly at T_0 reflects the formation of an energy gap Δ. The resulting phase diagram clearly distinguishes the $T_0(H)$ phase boundary and earlier observed high-field ($\mu_0 H \simeq 40$ T) field-induced phase transitions towards a large magnetic moment state. Surprisingly, the magnetic field dependence of μ, determined by previous neutron-scattering experiments, strongly differs from that of T_0 and Δ, with estimated critical fields of 14.5 T for μ and $\simeq 40$ T for T_0 and Δ. This observation suggests that there are two energy scales relevant to the magnetic phase transition of URu_2Si_2.

I. INTRODUCTION

The heavy-fermion superconductor URu_2Si_2 undergoes two consecutive phase transitions in zero magnetic field.[1] Magnetic susceptibility[1] and neutron diffraction[2] established an antiferromagnetic (AF) character of the first transition at $T_N = 17.5$ K, with a tiny ordered moment of $0.04\ \mu_B$/U atom. The ordering involves Ising-type spins coupled ferromagnetically in the ab plane, with the planes stacked antiparallel along the c axis of this tetragonal compound. The spin-wave excitation spectrum displays a gap of 115 K, and the magnetic dynamics are qualitatively well described by a singlet-singlet model.[3] This phase transition manifests itself in the electrical resistivity, $\rho(T)$, as a sharp increase at T_N and a subsequent exponential decrease to a Fermi-liquid T^2 behavior at the lowest temperatures.[4] Such behavior is associated with the opening of an energy gap over part of the Fermi surface, as evidenced in optical investigations[5] and vacuum tunneling.[6] The magnitude of this gap, 110 K in tunneling, is similar to the magnetic gap found by neutron scattering, suggesting strong coupling between spin and charge degrees of freedom.[3] Hall-effect studies show a large decrease in available charge carriers below T_N due to the gap opening.[7] At 1.5 K, these carriers are recovered at the upper field-induced transition at 39.2 T $\| c$, implying that this field closes the gap.[8]

The second transition is into a superconducting state below $T_c \simeq 1.3$ K. The excellent superconducting properties (highly sensitive to sample quality) of the single crystals used in this study have been discussed by Knetsch *et al.*[9] Here we concentrate on the phase transition at 17.5 K and its behavior in high magnetic fields.[10] The origin and nature of this transition are still not understood: although the ordered moment and the susceptibility anomaly at T_N are small, large anomalies occur in specific heat,[1] resistivity,[4] thermal expansion,[11] and nonlinear susceptibility.[12,13]

To reconcile these large anomalies with the small ordered moment, several scenarios have been put forth: the order parameter (OP) could be of nonlocal nature[12,13] (e.g., quadrupolar), or involve multispin correlators.[14] Polarized neutron experiments and a symmetry analysis[15] show that the AF peaks observed in low fields can only arise from purely dipolar order. Recent high-field neutron diffraction[16] ruled out the multispin correlator order parameters described by Barzykin and Gor'kov.[17] The type of quadrupolar ordering discussed by Santini and Amoretti[18] is in conflict with the observation of broken time reversal symmetry by polarized neutron scattering.[15] However, single-site properties of uranium in diluted $(U_xTh_{1-x})Ru_2Si_2$ $(0 < x < 0.07)$ yield compelling evidence of a doublet Γ_5 crystal-field split ground state of the $5f^2$ U ion,[19] which has the correct anisotropy and carries a quadrupole moment ($J_x^2-J_y^2$ or $J_xJ_y+J_yJ_x$). Its operators do not commute with the magnetic dipole J_z, so that the two possible order parameters compete. In case of ordering of the quadrupoles, the crystal-field level splitting can change significantly. In particular, the level splitting Δ_{CEF} itself must then be proportional to the quadrupolar OP,[20] consistent with experiment.[16] Recently, Barzykin and Gor'kov also treated the phase transition in first approximation as being of structural nature, involving U ions in a $5f^2$ configuration. The magnetic moment arises from a small admixture of a half-integer spin ($5f^1$ or $5f^3$) configuration, and time reversal symmetry is broken due to the fluctuating valence.[21] Our high-field experiments qualitatively support such a scenario.

II. RESISTIVITY IN HIGH MAGNETIC FIELDS

The $\rho(T)$ experiments were performed on two different single crystals of URu_2Si_2, grown by the Czochralski tri-arc

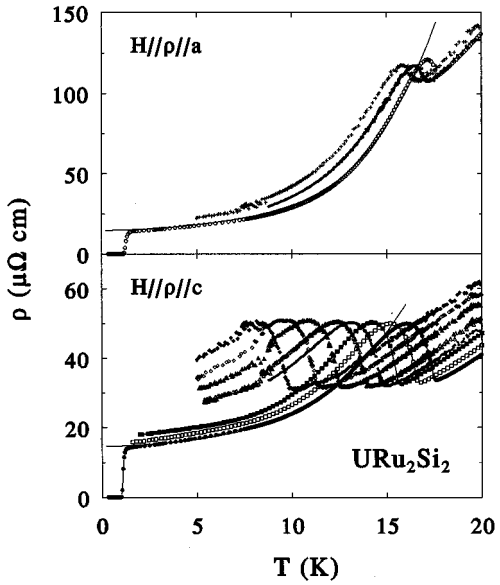

FIG. 1. Resistivity of URu_2Si_2 parallel to the a axis (upper panel) in fields of 0 (○), 16 (●), and 25 T (+) applied along a. The lower panel shows $\rho(T)$ parallel to the c axis in fields up to 25 T (from right to left, $\mu_0 H = 0$, 12, 14, 16, 18, 21, 23, and 25 T), applied along c. The solid lines are fits to Eq. (1).

method. The older crystal No. 1 (measured up to 16 T) was annealed at 900 °C for 7 days, the new crystal No. 2 (measured to 25 T) at 950 °C for 9 days. Both were slowly cooled to remove any residual stress. Earlier neutron experiments[2,16] were carried out on different, but similarly prepared crystals. Unannealed crystals are known to show anomalous temperature dependence of the ordered magnetic moment,[22] which was not observed in the crystals prepared the way described above. Both crystals show the same behavior in the experiments presented here.

We have measured the ac resistivity in constant magnetic fields, using a 18 T superconducting magnet at the University of Toronto in the temperature range from 0.3–30 K, and the hybrid 25 T magnet at the Nijmegen High Magnetic Field Laboratory, from 5–25 K. A small difference in absolute value of $\rho(T)$ and T calibration does not affect the following discussion. As we shall demonstrate, the characteristic energy for the ordered magnetic moment and the transition temperature are not the same. Therefore, we will from now on refer to the transition temperature as T_0 (following Miyako *et al.*[12]).

The zero-field $\rho(T)$ is in good agreement with previous work.[4] After an initial increase of $\rho_{\|a}$ and a near-constant $\rho_{\|c}$ upon cooling from 300 K, a large decrease below 70 K signals the formation of the coherent heavy-Fermi-liquid ground state. This evolution is interrupted by a sharp increase at $T_0 = 17.5$ K for both directions, as shown in Fig. 1. The anomaly for $\rho_{\|c}$, $\Delta\rho = \rho_{\max} - \rho_{\min} = 16.2\ \mu\Omega$ cm, is roughly twice as large as for $\rho_{\|a}$ ($\Delta\rho = 9.3\ \mu\Omega$ cm). Note that the increase $\Delta\rho_{\|c}$ is comparable to the decrease at the highest field-induced transition[24] at 39.2 T (at 1.5 K), again reflecting that the gap in the conduction band that opens at 17.5 K (and $H = 0$) closes at that field. To first approximation, $\rho(T)$ below 15 K can be well described by the expression:[23]

$$\rho(T) = \rho_0 + AT^2 + bT\left(1 + 2\frac{T}{\Delta}\right)\exp\left(-\frac{\Delta}{T}\right), \qquad (1)$$

applicable to an energy gap antiferromagnet with an additional T^2 term appropriate for Fermi-liquid behavior. In zero field, this spin-wave gap amounts to 72.7 K (51.2 K), for current parallel to the a (c) axis. The data up to 16 T, taken in 2 T intervals, allow for a similar evaluation of Δ. Given the number of fit parameters involved, a direct measurement by neutron scattering would be preferable. The resulting coefficients are given in Table I for fields along both directions.

TABLE I. Residual resistivity, Fermi-liquid T^2 coefficient A and spin-wave gap parameter Δ, derived from fits to Eq. (1), for URu_2Si_2 with current parallel to the easy c axis. The estimated error in the gap value is 0.2 K.

$\mu_0 H$ (T)	ρ_0 ($\mu\Omega$ cm)	A ($\mu\Omega$ cm/K^2)	b ($\mu\Omega$ cm/K)	$\Delta \| c$ (K)	$\Delta \| a$ (K)
0	14.7	0.0777	19.7	51.2	72.7
4	15.0	0.0768	19.5	50.1	73.4
8	15.5	0.0760	18.6	48.4	73.9
12	16.9	0.0760	18.4	46.4	73.8
16	18.4	0.0697	21.0	42.7	72.9

Figure 1 further shows the $\rho(T)$ anomaly for fields up to 25 T. For $H \| \rho \| a$, T_0 shifts slowly downwards, at a rate of −0.04 K/T. Within our accuracy, the energy gap is constant for $\mu_0 H \leqslant 16$ T. For $H \| \rho \| c$ however, the minimum in $\rho(T)$, used to define T_0, shifts downwards significantly. The magnitude of the anomaly, $\Delta\rho_{\|c}$, is constant for all fields. This suggests that the gap in the conduction band remains constant, although this should be proven by Hall-effect, tunneling, or infrared studies. The magnetoresistivity at constant T up to 25 K is given in Fig. 2 as $\Delta\rho = \rho(H) - \rho(0)$. The phase boundary determined in the constant field experiment is reproduced by the kink in $\Delta\rho(H)$ for $H \| c$.

III. MAGNETIC PHASE DIAGRAM AND DISCUSSION

Previous investigations were carried out in lower field or large pulsed fields at constant T, from 1.3–16 K. In fields around 40 T $\| c$, a three-step transition, nearly independent of T, occurs to a magnetic state with large U moments of $\approx 1\mu_B$, as demonstrated by independent pulsed-field magnetization and magnetoresistance[24,25] on different samples. We used these results to generate a more complete phase diagram, displayed in Fig. 3. The high-field magnetization process has been explained in terms of spin-reorientation transitions, using an Ising model with three exchange parameters.[25]

Where previous data in low field ($\mu_0 H \leqslant 8$ T) could be thought to directly relate to these high-field transitions, yielding only one transition into an AF state, our data up to 25 T show that these phase boundaries belong to two *different* phase transitions. This is most obvious at $T = 12$ K. The resistive transition decreases with fields parallel to c according to

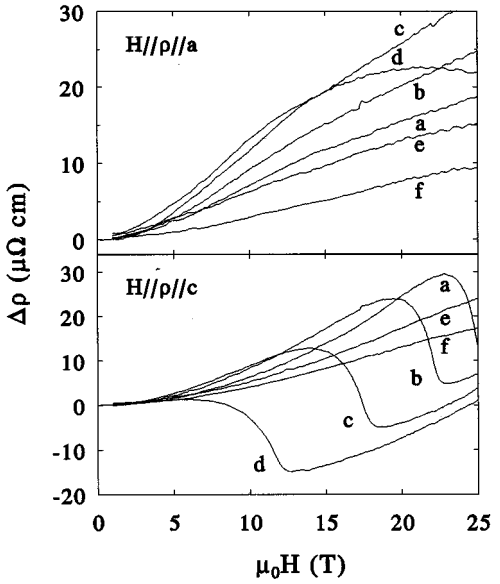

FIG. 2. Magnetoresistivity of URu_2Si_2 parallel to the *a* (upper panel) and *c* (lower panel) axes at T = 10 K (a), 12 K (b), 14 K (c), 16 K (d), 20 K (e), and 25 K (f). The break-up of the ordered phase is signalled by the minimum in $\Delta\rho$ for $H \parallel c$, that has to be contrasted with the jumps associated with the field-induced transitions (Ref. 24) around 40 T.

$$T_0(H) = T_0^0[1-(H/H_0)^2], \tag{2}$$

with $T_0^0 = 17.6(1)$ K and $\mu_0 H_0 = 40.3 \pm 1.0$ T.[26] The value for H_0 is close to the upper field-induced phase transition.[24,25] The extrapolation involved prevents us from concluding whether the high-field phase is separate from the $T_0(H)$ phase boundary or possibly degenerate as $T \to 0$. The fitted values for the spin-wave gap, obtained for $\mu_0 H \leqslant 16$ T, are also well described by a quadratic field dependence:

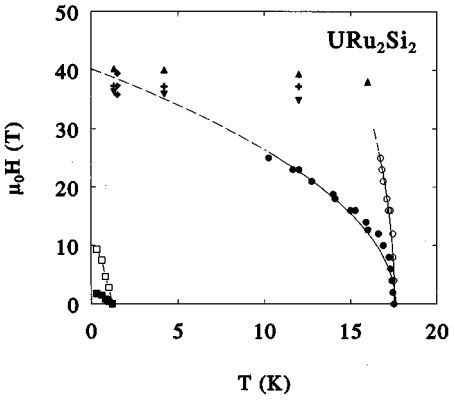

FIG. 3. Magnetic phase diagram of URu_2Si_2 with field parallel to the *a* (open symbols) and *c* axes (closed symbols), composed of isothermal magnetization data (triangles and +) taken at 1.3 K, 4.2 K, 12 and 16 K by Sugiyama *et al.* (Ref. 25), magnetoresistance data at 1.5 K (diamonds) from de Visser *et al.* (Ref. 24), and our current data for both constant field and temperature. The lines denote fits to Eq. (2). The superconducting H_{c2} is shown in the lower left corner.

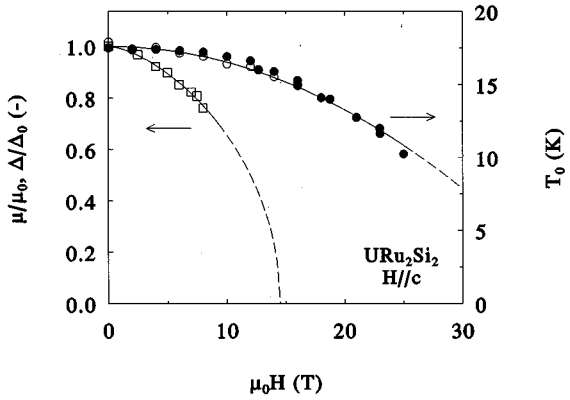

FIG. 4. Reduced staggered magnetic moment (Ref. 16) μ/μ_0 (□) at 4.3 K, reduced energy gap Δ/Δ_0 (○, left axis), and transition temperature T_0 (●, right axis) of URu_2Si_2 versus magnetic field $H \parallel c$. Note the similar field dependence of T_0 and Δ, and their large discrepancy compared with μ. If extrapolated by Eq. (4) (dashed line), μ has disappeared around 14.5 T, whereas 40 T is necessary to break up the ordered state and close the gap.

$$\Delta(H) = \Delta_0[1-(H/H_0)^2], \tag{3}$$

with $\Delta_0 = 50.4$ (2) K and $\mu_0 H_0 = 40.8$ (5) T. Note that this is the same characteristic field as found for T_0, suggesting that T_0 is determined by the magnitude of the (field-dependent) gap.

We can now discuss the relation between this phase boundary of URu_2Si_2 and the field dependence of the ordered magnetic moment, measured by neutron diffraction.[16] Figure 4 displays the phase boundary and reduced gap parameter, plotted as T_0 [with the solid line a fit to Eq. (2)] and Δ/Δ_0 versus $H \parallel c$, together with the reduced ordered moment, $\mu_{\rm ord}/\mu_{\rm ord}(0)$. At 4.3 K, $\mu_{\rm ord}(H)$ follows the relation[16]

$$\mu_{\rm ord}(H) = \mu_{\rm ord}(0)\sqrt{1-(H/H_c)^{3/2}}, \tag{4}$$

with $\mu_{\rm ord}(0) = 0.04\mu_B$ and the characteristic field $\mu_0 H_c = 14.5(3)$ T. Figure 4 shows an anomalous relation between the staggered moment and the $T_0(H)$ phase transition. $\mu_{\rm ord}$ falls off much more rapidly with field than T_0, indicating that the moment value does not determine the energy scale for the transition at T_0. Combination of Eqs. (2) and (4) gives $\mu_{\rm ord} \propto T_0^{3/8}$, which is close to the mean-field prediction ($\mu_{\rm ord} \propto T_0^{1/2}$), but the critical fields are vastly different.

We believe these observations indicate the existence of two distinct energy scales, which would imply that the zero-field phase transition at 17.5 K involves two order parameters, which respond differently to an external magnetic field. Direct proof of the existence of these two energy scales can only be obtained by neutron scattering above 15 T. Unfortunately, this field range is very difficult to access experimentally, especially given the small size of the staggered magnetic moment. Pulsed-field magnetization is insensitive for the same reason. The available data show that in a wide field range above H_c, a phase transition occurs at $T_0(H)$ with energy gap $\Delta(H)$, whereas extrapolation of the ordered moment in that field suggests $\mu_{\rm ord} = 0$. This is remarkable, since the gap was thought to be associated with the doubling of the unit cell due to the AF ordering. Therefore, an addi-

tional symmetry must be broken to explain the gap formation in high fields. A similar effect is seen in UPt_3, where pressure reduces the ordered moment, without affecting T_N.[27]

In conclusion, the magnetic field dependence of the resistive transition in URu_2Si_2 shows that this transition involves a Fermi surface reconstruction, with a field-dependent spin-wave gap, Δ, proportional to the ordering temperature, T_0. However, T_0 has a field dependence significantly different from that of the ordered magnetic moment and the high-field transitions that involve large uranium moments. This we believe is evidence for the *nonmagnetic* character of the order parameter associated with T_0, with the implication that the small ordered magnetic moment is only a by-product of this phase transition. How this can be realized theoretically is still an open question, but in the above sense, our data support the scenario put forth by Barzykin and Gor'kov.[21] Future experimental work should address the field dependence of the spin-wave and conduction band gap. Further resistance and dilatation experiments are planned to complete the intriguing phase diagram of URu_2Si_2.

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