Partially Gapped Fermi Surface in the Heavy-Electron Superconductor URu_2Si_2

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Transport, thermal, and magnetic data for the heavy-electron system URu_2Si_2 inicate that a charge- or spin-density-wave transition opens an energy gap of ~ 11 meV over a portion of the Fermi surface below $T_0 \approx 17.5$ K and demonstrate that bulk superconductivity occurs below $T_c \approx 1.5$ K. The pressure dependences of T_0 and T_c support this interpretation. The unusually large initial slope of the upper critical magnetic field (9.2 T/K) is consistent with the high values of the electronic-specific-heat coefficient and the electrical resistivity.

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Last year, Schlabitz and co-workers[1] reported that the compound URu_2Si_2 becomes superconducting at ~ 1.5 K and has unusual superconducting and normal-state physical properties similar to those exhibited by heavy-electron superconductors. These properties include a large slope of the upper critical magnetic field H_{c2} at the superconducting critical temperature T_c, $(-dH_{c2}/dT)_{T_c} = 7.4$ T/K,[2] and a moderately large electronic-specific-heat coefficient, $\gamma = 75$ mJ/mole-K^2.[1] In addition, features in the specific heat, electrical resistivity, and magnetic susceptibility at $T_0 \sim 17.5$ K were interpreted as evidence for itinerant-electron antiferromagnetic ordering.[1] To our knowledge, the occurrence of magnetic order below T_0 has not been verified by elastic-neutron-scattering measurements. However, inelastic-neutron-scattering measurements on URu_2Si_2 have revealed a sharp inelastic excitation at about 7.5 meV at low temperatures, which has been attributed to a gap produced by the hybridization of U $5f$ states with conduction-electron states.[2]

In this Letter, we present transport, thermal, and magnetic data on the compound URu_2Si_2 that characterize the 17.5-K phase transition and demonstrate that bulk superconductivity occurs below 1.5 K. The phase transition appears to involve the development of a charge- or spin-density wave (CDW or SDW) out of the heavy-electron system, which opens an energy gap of ~ 11 meV over a portion of the Fermi surface. The energy gap is comparable in magnitude to the gap inferred from the inelastic-neutron-diffraction measurements, suggesting that they may be one and the same. Measurements of the pressure dependence of T_0 and T_c, the superconducting critical temperature, indicate that the fraction of the Fermi surface removed by the CDW or SDW increases under pressure.

The polycrystalline URu_2Si_2 sample was prepared by the arc melting of high-purity elements (U, 99.97%; Ru, 99.96%; Si, 99.99999%) together on a water-cooled copper hearth in an argon atmosphere, followed by annealing at 900 °C for one week. X-ray diffraction measurements revealed a high-quality diffraction pattern with sharp lines that could be indexed to the body-centered tetragonal $ThCr_2Si_2$ structure and no evidence of any other impurity phases. The tetragonal lattice parameters had the values $a = 4.129$ Å and $c = 9.575$ Å.

Electrical resistivity ρ versus temperature T data between ~ 80 mK and 300 K for URu_2Si_2 are displayed in Fig. 1(a). The measurements were made on a bar-shaped specimen with a standard four-lead ac technique at a frequency of 16 Hz. The large magnitude of ρ (2.46 mΩ-cm at 300 K) is particularly noteworthy and appears to be intrinsic as discussed below. The shape of the $\rho(T)$ curve is qualitatively similar to that of many hybridized f-electron rare-earth and actinide compounds, particularly the rapid decrease in ρ with decreasing temperature below ~ 50 K which is presumed to be associated with the freezing out of charge and/or spin disorder scattering. The abrupt drop in ρ below 1.7 K is due to the onset of superconductivity, with the midpoint of the resistive transition curve occurring at 1.5 K. In addition, there is a small but sharp peak in ρ at 17.2 K which resembles the type of anomaly that would be expected for a CDW or SDW transition or, perhaps, a structural transition. However, we were unable to detect a structural transition in the vicinity of 17.5 K from x-ray diffraction measurements made at various temperatures between 4 and 300 K. The anomaly in $\rho(T)$ at 17.2 K is relatively insensitive to applied magnetic fields, shifting downwards in temperature by only ~ 0.1 K in a field

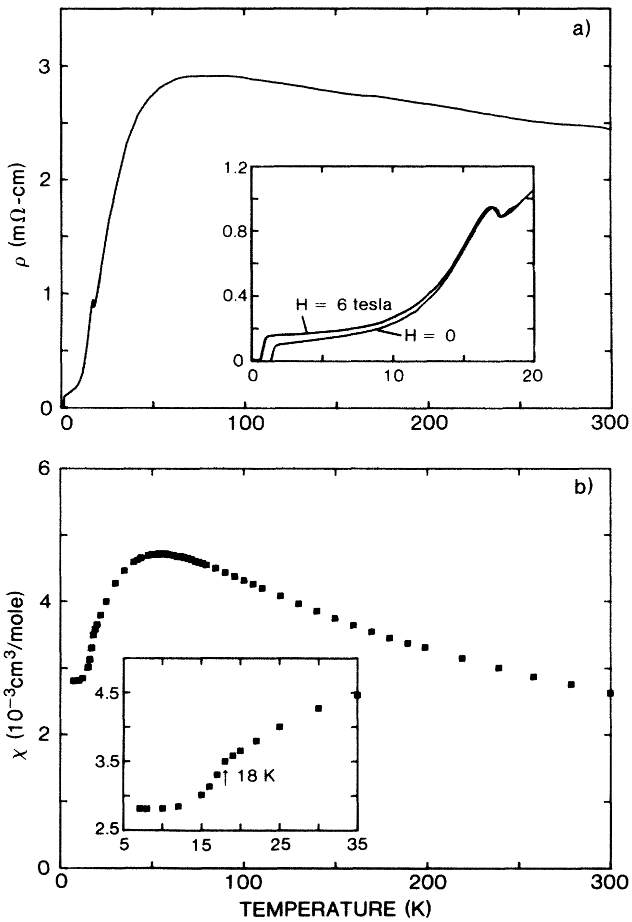

FIG. 1. (a) Electrical resistivity ρ vs temperature for URu_2Si_2 between 80 mK and 300 K. Inset: ρ vs temperature between 80 mK and 20 K in magnetic fields of 0 and 6 T. (b) Magnetic susceptibility χ vs temperature between 2 and 300 K. Inset: χ vs temperature in the vicinity of the phase transition at ~ 17.5 K.

of 6 T [see inset of Fig. 1(a)]. Between T_c and ~ 8 K, ρ varies approximately linearly with T, and at 2 K, a positive and linear magnetoresistance of $\sim 60\%$ at 6 T is observed.

The magnetic susceptibility χ vs T of URu_2Si_2, measured in a field of 0.5 T with a SHE SQUID magnetometer between 2 and 300 K, is displayed in Fig. 1(b). There is a rounded maximum near 55 K in the $\chi(T)$ data which correlates with a similar maximum in the vicinity of 70 K in the $\rho(T)$ data. A small but distinct change in slope in $\chi(T)$ at 18 K [see inset of Fig. 1(b)] accompanies the phase transition. Both the increase in ρ and the decrease in χ as T is decreased through ~ 18 K could result from a change in the Fermi-surface topology associated with the formation of a CDW or SDW.[3] At various temperatures between 5 and 60 K, the magnetization was found to be linear in applied magnetic field up to 4 T.

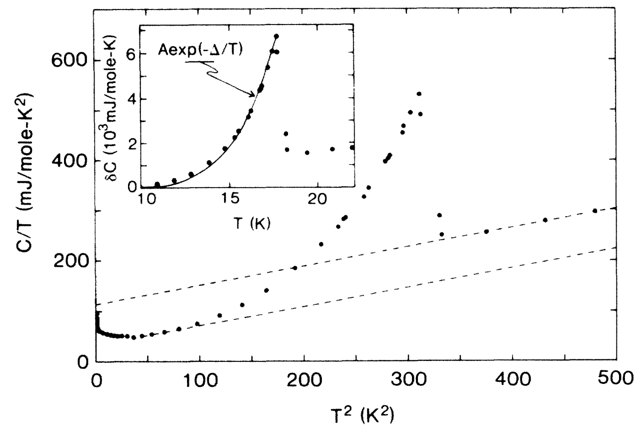

FIG. 2. Specific heat C divided by temperature T vs T^2 for URu_2Si_2 between 0.6 and 500 K^2. The meaning of the dashed lines is explained in the text. Inset: Estimated specific heat δC vs T associated with the apparent CDW or SDW transition at ~ 17.5 K. The solid line represents the function $A\exp(-\Delta/T)$ with values for A and Δ given in the text.

The strongest evidence for the formation of a CDW or SDW over a portion of the Fermi surface in URu_2Si_2 is provided by low-temperature specific-heat measurements. Data on the specific heat C, taken in a semiadiabatic calorimeter with a standard heat-pulse method, are plotted as C/T vs T^2 in Fig. 2 between 0.6 and 500 K^2. There is a striking feature in the $C(T)$ data between ~ 9 and ~ 18 K whose shape is reminiscent of a second-order BCS-type mean-field transition which would be expected for the formation of a CDW or SDW. An analysis of this $C(T)$ feature, discussed below, yields an estimate of the energy gap and the fraction of the Fermi surface removed by the CDW or SDW. Below 1.5 K, there is another broadened BCS-type specific-heat anomaly that is associated with the superconducting transition. A linear extrapolation to 0 K of the C/T vs T^2 data in the normal state above 1.5 K yields a value of ~ 65.5 mJ/mole-K^2 which presumably represents the 0-K value of the electronic-specific-heat coefficient γ in the absence of superconductivity. This value for γ is comparable to the value quoted by Schlabitz and co-workers (75 mJ/mole-K^2),[1] and the C/T vs T^2 data have the same characteristic low-temperature upturn found in many of the heavy-electron systems such as $CeCu_2Si_2$[4] and UBe_{13}.[5] This moderately large γ value can be used to estimate the electron effective mass m^* following the method described by Maple *et al.*,[6] yielding the value $m^* \sim 25 m_e$, where m_e is the free electron mass. The portion of the Fermi surface that is not gapped by the CDW or SDW is removed by the superconductivity that occurs below 1.5 K, resulting in an energy gap $\Delta_S \sim k_B T_c \sim 0.1$ meV over the remainder of the Fermi surface.

The specific-heat anomaly associated with the 17.5-K phase transition may be analyzed in the following manner. Above 18 K, the $C(T)$ data can be described by the expression $C=\gamma T+\beta T^3$, where γT and βT^3 are the electronic and lattice contributions, respectively, with $\gamma=112$ mJ/mole-K^2 and $\beta=0.382$ mJ/mole-K^4 (corresponding to a Debye temperature $\theta_D=294$ K). If we now estimate the specific heat of the remaining ungapped electrons between ~ 9 and 18 K by assuming that the lattice term is unchanged and C/T is equal to the experimental value below 9 K (see lower dashed line in Fig. 2), then the difference $\delta C=C-C'$, where C' is the estimated specific heat of the ungapped electrons plus the lattice, presumably represents the contribution to C of the electrons that are involved in the formation of the charge or spin ordered state. The δC vs T data obtained in this manner are shown in the inset of Fig. 2. We find that the jump in δC at 17.9 K is equal to 5.82 J/mole-K$=2.9\gamma T_0$, where $T_0=17.9$ K is the phase transition temperature, while δC can be described by

$$\delta C=A\exp(-\Delta/T), \qquad (1)$$

with $A=9890$ J/mole-K$=4900\gamma T_0$ and $\Delta=129$ K$=7.2T_0$ (solid line in the inset of Fig. 2). Thus, the specific-heat data appear to be consistent with a second-order phase transition that involves the formation of an energy gap of the order of 129 K $\sim$ 11 meV. It is interesting to note that the value of the energy gap is comparable to the energy gap that was inferred from the inelastic-neutron-diffraction measurements of Walter *et al.*[2] The entropy $\int_0^{T_0}(\delta C/T)dT$ is estimated to be 979 mJ/mole-K$=0.170k_B\ln2$ per formula unit. From the ratio of the ungapped γ, estimated by linearly extrapolating the C/T vs T^2 data from above 18 K to $T=0$, to the low-temperature γ, obtained by linearly extrapolating the C/T vs T^2 data in the normal state above 1.5 K to $T=0$, we find that the CDW or SDW removes about 40% of the Fermi surface.

Shown in Figs. 3(a) and 3(b), respectively, are H_{c2} vs T and C/T vs T data that pertain to the superconducting state. The H_{c2} vs T data were determined from resistively measured superconducting transition curves at 16 Hz in a ^{3}He-^{4}He dilution refrigerator in magnetic fields up to 6 T. The H_{c2} vs T curve is very steep with an initial slope of $(-dH_{c2}/dT)_{T_c}=9.2$ T/K, somewhat higher than the value of 7.4 T/K quoted in Ref. 2. In the dirty limit, the Werthamer-Helfand-Hohenberg-Maki theory[7,8] of type-II superconductivity gives $(-dH_{c2}/dT)_{T_c}=4.44\gamma\rho$, where H_{c2} is in teslas, γ is in ergs/cm^3-K^2, and ρ is in ohm centimeters.[9] Using the values $\gamma=65.5$ mJ/mole-K$^2=1.34\times10^4$ ergs/cm^3-K^2 and $\rho(2\text{ K})=1.04\times10^{-4}$ Ω-cm, we calculate $(-dH_{c2}/dT)_{T_c}=6.2$ T/K, in reasonable agreement with the measured value. This indicates that the

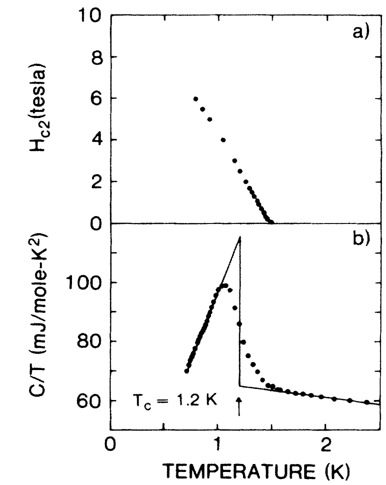

FIG. 3. (a) Upper critical magnetic field H_{c2} vs temperature for URu_2Si_2. (b) Specific heat C divided by temperature T vs T for URu_2Si_2.

large value of ρ might well be intrinsic to the specimen. The C/T vs T data show that the superconductivity of URu_2Si_2 is a bulk phenomenon. The jump ΔC at T_c is equal to 60 mJ/mole-K, $\sim$ 56% of the value expected from the BCS theory $\Delta C_{BCS}=1.43\gamma T_c=108$ mJ/mole-K.[10]

The pressure dependences of T_0 and T_c of URu_2Si_2 are also consistent with the occurrence of a CDW or SDW transition at T_0. Measurements of $\rho(T)$ at various hydrostatic pressures P between 0 and 16 kbar reveal that T_0 increases linearly with P at the rate $dT_0/dP=125$ mK/kbar up to 11 kbar, and at higher rates above 11 kbar, while T_c decreases at the rate $dT_c/dP=-97$ mK/kbar. The inverse correlation between the P dependences of T_0 and T_c is consistent with an increase of the fraction of the Fermi surface that is removed by the formation of the CDW or SDW. If the increase in T_0 is associated with an increase in the fraction of electron states involved in the formation of a CDW or SDW, rather than an increase in the coupling constant, then a smaller fraction of electron states would be available for superconducting pairing, leading to a decrease in T_c. A mean-field treatment of the competition between superconductivity and CDW formation involving partial gapping of the Fermi surface was developed by Bilbro and McMillan[11] and applied to the pressure dependence of T_c and the structural transition temperature of the $A15$ compounds[11] and Chevrel-phase compounds of the type $Eu_{1-x}Sn_xMo_6S_8$.[12] The proposed relation between T_c and T_0 can be written as $T_c^nT_0^{1-n}=T_{c0}$, where T_{c0} is the superconducting transition temperature in the absence of a gap and n is the fraction of the Fermi surface which is ungapped.

In summary, transport, thermal, and magnetic measurements on the heavy-electron superconductor URu_2Si_2 indicate that a phase transition occurs at $T_0 \sim 17.5$ K which opens an energy gap of ~ 11 meV over about 40% of the Fermi surface. The identification of this transition with either a CDW or a SDW requires further investigation. The rather high value of the electrical resistivity of this material seems to be intrinsic and correlates well with the unusually large initial slope $(-dH_{c2}/dT)_{T_c}$. We anticipate that a better understanding of the physics of this material will emerge from studies on high-quality single crystals where the effects of disorder and anisotropy can be assessed.

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Note added.—After submission of this paper for publication, we received a preprint of a paper by Palstra *et al.*[13] reporting specific-heat and magnetic-susceptibility data on single-crystal and polycrystal specimens of URu_2Si_2. In addition, we were sent a copy of the Ph.D. thesis of Baumann.[14] Experimental results on URu_2Si_2 presented in this thesis are in general agreement with those reported in the present paper.

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