Effect of pressure on competing electronic correlations in the heavy-electron system URu_2Si_2

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The heavy-electron compound URu_2Si_2 exhibits two electronic phase transitions: superconductivity at $T_c \cong 1.5$ K and a second transition at $T_0 \cong 17.5$ K. The specific-heat anomaly associated with the transition at T_0 has a mean-field BCS-like form which suggests the formation of a charge- or spin-density wave that partially gaps the Fermi surface. Through electrical-resistivity measurements, we have studied the influence of pressure P up to ~ 15.4 kbar on these transitions. For pressures less than 12 kbar, T_0 increases linearly at a rate ~ 130 mK/kbar, while T_c decreases linearly at a rate ~ 95 mK/kbar. The nearly equal but opposite P dependences of T_0 and T_c suggest a competition for electronic density of states at the Fermi level. The resistivity, when normalized to its maximum value $\rho(T_{max})$, scales as a function of reduced temperature $T/T_{max}(P)$ from T_c to well above T_{max}, except in the immediate vicinity of T_0.

INTRODUCTION

The compound URu_2Si_2 has attracted a considerable amount of attention because it has been reported to have a moderately large low-temperature electronic specific-heat coefficient of ~ 65 mJ/mol K^2 and undergoes two low-temperature electronic phase transitions, a superconducting one at $T_c \cong 1.5$ K and another one at $T_0 \cong 17.5$ K.[1-3] It has been suggested that the phase transition at 17.5 K is due to antiferromagnetic ordering of either localized[1] or itinerant[2] U5f magnetic moments. According to a report,[4] preliminary results of neutron diffraction experiments on a single crystal specimen of URu_2Si_2 have revealed very small, antiferromagnetically ordered uranium moments of $\sim 0.01\mu_B$ below 17 K.[5] Recently, we observed that (1) the specific-heat anomaly δC associated with the 17.5-K transition in URu_2Si_2 has a shape that is reminiscent of a second-order BCS-type mean-field transition and can be described by the relation $\delta C = A \exp(-\Delta/T)$ with an activation energy $\Delta \cong 129$ K $\cong 11$ meV, and (2) the value of the electronic specific-heat coefficient γ extrapolated to 0 K (65.5 mJ/mol K^2) is substantially reduced with respect to the value of γ at temperatures above 17.5 K (112 mJ/mol K^2).[3] On the basis of these observations and the types of anomalies found in the electrical resistivity ρ and the magnetic susceptibility χ of URu_2Si_2 near 17.5 K, we proposed[3] that a charge- or spin-density wave (CDW or SDW) transition may occur at $T_0 \cong 17.5$ K that opens up an energy gap of ~ 11 meV over about 40% of the Fermi surface. Preliminary results for the pressure dependence of T_0 and T_c, inferred from ρ versus temperature T data at various pressures, were also reported[3] which are consistent with this interpretation. In the present paper, a detailed account of our measurements of $\rho(T)$ under hydrostatic pressure up to 15.4 kbar is presented. Measurements of $\rho(T)$ parallel to the a axis on a single-crystal specimen of URu_2Si_2 under hydrostatic pressure up to 4 kbar have been reported by de Boer *et al.*[6]

EXPERIMENTAL DETAILS

The polycrystalline URu_2Si_2 sample was cut from the same ingot from which samples used in our previous investigation were taken.[3] The URu_2Si_2 ingot was prepared by arc melting high-purity elements (99.97% pure U, 99.96% pure Ru, and 99.99999% pure Si) together on a water cooled copper hearth in a Zr-gettered 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 other impurity phases. The tetragonal lattice parameters had the values $a = 4.129$ Å and $c = 9.575$ Å. Electrical resistivity measurements under pressure were made in Be-Cu clamped piston-cylinder devices in a ^{4}He cryostat for temperatures between 1 and 300 K,[7] and in a ^{3}He-^{4}He dilution refrigerator between 20 mK and 1 K. A 1:1 mixture of isoamyl alcohol and n-pentane served as the quasihydrostatic pressure transmitting medium, and the pressure was inferred from the T_c of a superconducting Pb manometer.[7] The electrical resistance measurements were made using a standard four-lead and lock-in detection ac technique at a frequency of 220 Hz.[8] Temperature was determined from a calibrated carbon glass resistance thermometer in the experiments above 1 K, and from a calibrated Ge resistance thermometer in the experiments below 1 K.

RESULTS

Measurements of $\rho(T)$ between 1 and 300 K were made at six different pressures ranging from atmospheric pressure to 15.4 kbar. Typical $\rho(T)$ curves between 1 and 300 K at pressures of one atmosphere, 4.6, 10.7, and 15.4 kbar are plotted in Fig. 1. The shape of the $\rho(T)$ curve at atmospheric pressure is similar to previously reported curves for polycrystalline samples of URu_2Si_2.[1,3] However, the value of ρ at 300 K ($\sim 560\ \mu\Omega$ cm) is about 30% larger than the value reported by Schlabitz *et al.*,[1] about two times larger than the value found in single crystals by Palstra *et al.*,[4] and about four times smaller than the value we previously reported for a specimen that had been cut from the same ingot.[3] These discrepancies are apparently due to cracks and voids in the material which has a rather fibrous texture. As T is decreased from 300 K, ρ increases rather slowly, attains a maximum value at $T_{max} = 73.2$ K, and then decreases rapidly. In addition, there is a feature in the $\rho(T)$ curve near 17.5 K that is associated with the phase transition (which we presume to be a CDW or SDW transition), as well as an abrupt drop in ρ to zero near 1.5 K that is due to the onset of superconductivity. We define the critical temperature T_0 of the phase transition as the temperature of the inflection point of the anomaly in the $\rho(T)$ curve which can be readily determined from the minimum in plots of $d\rho/dT$ versus T such as the two displayed in Fig. 2 for pressures of one atmosphere and 15.4 kbar.

As revealed in Fig. 1, pressure has a relatively strong effect on the $\rho(T)$ curve for $T < 100$ K, shifting the maximum and the anomaly at T_0 to higher temperatures and the superconducting transition to lower temperatures. In contrast, pressure has a rather small effect on the $\rho(T)$ curve for $T > 100$ K, resulting in a small decrease in the slope of the part of the curve that is nearly linear in T. Shown in the inset of Fig. 1 are $\rho(T)$ data between 1 and 25 K that reveal how the anomaly associated with the phase transition and the superconducting transition evolve with pressure. Since the superconducting transition

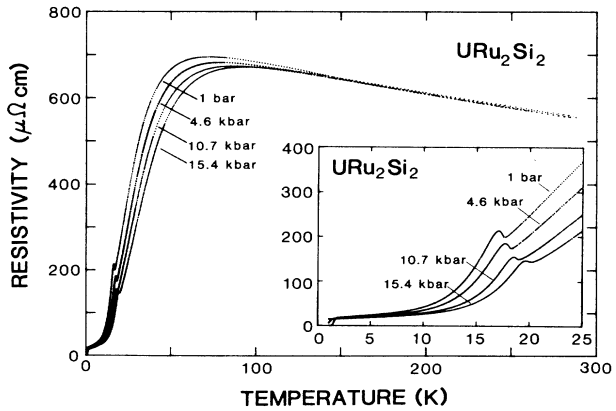

FIG. 1. Electrical resistivity of URu_2Si_2 as a function of temperature at various selected pressures. The inset shows an expanded view of the low-temperature behavior.

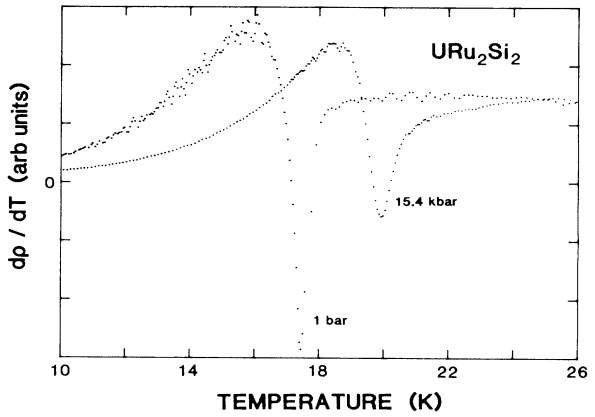

FIG. 2. Derivative $d\rho/dT$ of the resistivity of URu_2Si_2 versus temperature at ambient pressure and at 15.4 kbar. The phase transition temperature $T_0(P)$ is defined as the temperature of the sharp minimum in $d\rho/dT$.

dropped below 1 K under pressure, the $\rho(T)$ measurements were extended below 1 K in a ^{3}He-^{4}He dilution refrigerator at pressures of 9.6 and 12.6 kbar. The superconducting transitions at pressures of 0, 0.5, 3.2, 9.6, and 12.6 kbar are shown in Fig. 3. Because of experimental difficulties, the superconducting transitions determined in the dilution refrigerator were measured with a three-lead configuration, and the absolute values of the resistivity may be less accurate than for the measurements made with a four-lead configuration. The dependences of T_{max}, T_0, and T_c on pressure, derived from the data presented in Figs. 1–3 and other data not shown in these figures, are plotted in Fig. 4. The values of T_c are resistive midpoints. The transition width $\Delta T_c = T_c(90\%) - T_c(10\%)$ increases approximately linearly with pressure from ~ 0.3 K at 1 bar to ~ 0.9 K at 12 kbar. The increase of ΔT_c

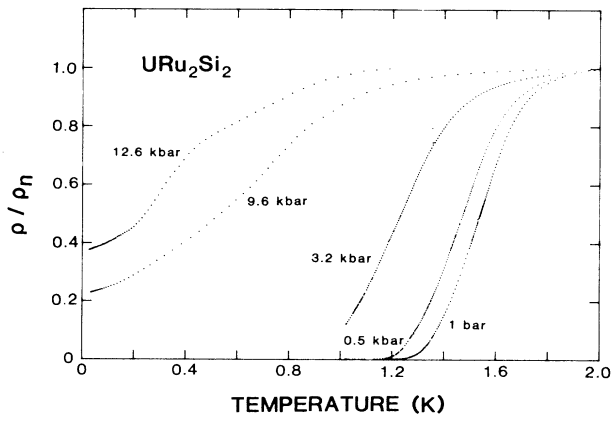

FIG. 3. Normalized electrical resistivity of URu_2Si_2 in the vicinity of the superconducting transition temperature T_c at various pressures. Note that the data shown for the two highest pressures were obtained from measurements performed with a three-lead configuration.

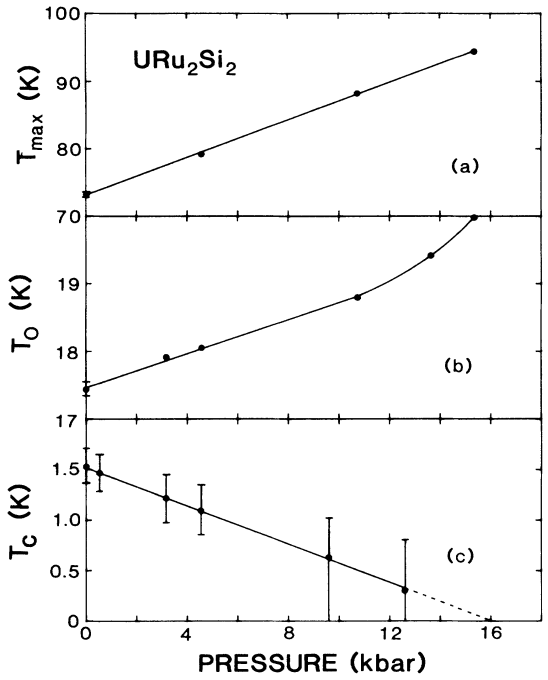

FIG. 4. Pressure dependence of T_{max}, T_0, and T_c. For (a) and (b), exemplary error bars are given at 1 bar.

with pressure may be associated with the different states of strain within the polycrystalline sample due to the distribution of orientations of the noncubic crystallites.

DISCUSSION

Apart from the feature at 17.5 K and the superconducting transition, the overall shape of the $\rho(T)$ curve for URu_2Si_2 at atmospheric pressure (Fig. 1) is reminiscent of valence fluctuation behavior displayed by many rare earth and actinide compounds. At high temperatures, the large value of ρ and the increase of ρ with decreasing T are consistent with incoherent single-ion Kondo scattering, whereas at low temperature the rapid decrease of ρ with decreasing T is presumably due to the onset of coherence leading to a low-temperature Fermi liquid ground state.

The small decrease of the slope of the $\rho(T)$ curve with pressure above ~ 100K is consistent with a small increase of the Kondo temperature T_K with pressure, in the regime $T < T_K$. The relatively larger increase of T_{max} with pressure suggests that there is another characteristic temperature scale that reflects the development of the coherent Fermi-liquid ground state, of which T_{max} is a rough measure. Evidently, this temperature scale is a much stronger function of pressure than T_K. According to the data in Fig. 4, T_{max} increases linearly with pressure at the rate $dT_{max}/dP = 1.40$ K/kbar. Shifts of T_{max} to higher temperature under pressure have been observed in a number of valence fluctuation systems such as $CeAl_3$,[9] $CeCu_6$,[10] and $CeCu_2Si_2$,[11,12] and have been attributed by others to an increase of T_K with pressure. However, recently it has been pointed out[13] that the presence of Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions may play an important role in determining T_{max} and that T_{max} is a complex function of both T_K and the RKKY interaction.

Interestingly, if $\rho(T)$ is normalized to its peak value $\rho(T_{max})$, it scales with T/T_{max} for $T_c < T < 1.2\ T_{max}$, except for the location on the scaled $\rho(T)$ curve of the anomaly due to the phase transition at T_0. The scaling behavior is illustrated in Fig. 5 where $\rho(T)$ data at 0, 4.6, 10.7, and 15.4 kbar have been plotted as $\rho(T)/\rho(T_{max})$ versus T/T_{max}. Similar scaling has been found[10,12] in other heavy fermion or mixed valence systems and appears to be a general feature associated with the development of the coherent ground state; however, the fact that scaling is excellent both above and below the transition at T_0 is especially notable.

For many valence fluctuation systems, the low-temperature properties conform to typical Fermi liquid type behavior, i.e., $\rho = \rho_0 + AT^2$ for $T \ll T_K$ with $A \alpha T_K^{-2}$ (where we have identified T_K with T_{SF}, the spin-fluctuation temperature.[14]) However, the $\rho(T)$ data for URu_2Si_2 at all pressures do not show a pure T^2 dependence over any extended temperature interval between T_c and T_0. Possible reasons for the absence of the T^2 behavior may be that it is obscured by the T dependence associated with the CDW or SDW transition below ~ 17.5 K. Recently, it was suggested[4] that the resistivity of URu_2Si_2 could be fit by the sum of a T^2 term, associated with low-temperature Fermi-liquid behavior, and an exponential contribution $\exp(-\Delta/T)$ with a temperature-dependent prefactor appropriate for an antiferromagnet with an energy gap Δ[15] in the magnetic excitation spectrum; i.e.,

$$\rho = \rho_0 + AT^2 + B(T/\Delta)[1 + 2T/\Delta]\exp(-\Delta/T) . \quad (1)$$

Nonlinear least-square fits of this equation to resistivity data between 1 and 16.5 K at all pressures investigated were made. The parameters Δ, ρ_0, A, and B derived from fits to Eq. (1) and Δ/T_0 are given in Table I. Plots of the $\rho(T)$ data between 2 and 16 K at pressures of 1 bar, 4.6 kbar, 10.7 kbar, and 15.4 kbar are compared to the fits in

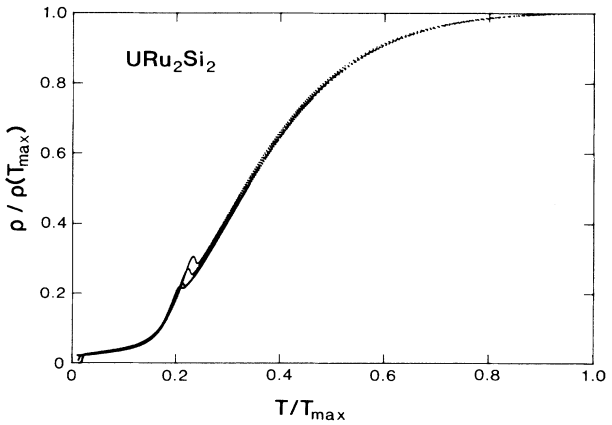

FIG. 5. Resistivity ρ normalized to its maximum value $\rho(T_{max})$ as a function of reduced temperature T/T_{max}. Data used are those given in Fig. 1.

TABLE I. Parameters Δ, ρ_0, A, and B derived from fits of Eq. (1) to the $\rho(T)$ data between 1 and 16.4 K and Δ/T_0 at various pressures P.

P (kbar)	Δ (K)	Δ/T_0	$\rho_0(\mu\Omega\,\mathrm{cm\,K^{-2}})$	$A(\mu\Omega\,\mathrm{cm\,K^{-2}})$	$B(\mathrm{m\Omega\,cm})$
0.001	71.4	4.1	19.5	0.180	30.3
3.2	77.7	4.3	14.5	0.156	37.5
4.6	82.7	4.6	16.4	0.167	49.8
10.7	110.3	5.9	15.1	0.138	196
13.6	111.5	5.7	15.1	0.121	159
15.4	115.9	5.8	14.9	0.114	180

Fig. 6. As shown in Fig. 7, the energy gap Δ increases linearly with pressure at the rate 3.1 K/kbar, while the coefficient A decreases linearly with pressure at the rate 0.0041 $\mu\Omega\,\mathrm{cm/K^2\,kbar}$. Interestingly, the value of $\Delta(P=0)$ obtained from this fitting procedure is in reasonably close agreement with the value of 5.5 meV corresponding to the onset of a gaplike magnetic excitation in the inelastic neutron scattering spectrum of URu_2Si_2 observed recently by Walter *et al.*[16]

Perhaps one of the most interesting results of this investigation is the linear decrease of T_c with pressure, which occurs at a rate of 95 mK/kbar, as illustrated in Fig. 4. This rate of depression is much larger than that found in other U-based heavy fermion superconductors. For example, $dT_c/dP=-13$ mK/kbar in UPt_3,[17] -10 mK/kbar in UBe_{13},[18] and about -53 mK/kbar in the comparably heavy electron compound U_2PtC_2.[19] The data of Fig. 4 also reveal that T_0 increases with pressure, essentially linearly between 0 and 11 kbar at a rate $dT_0/dP=0.13$ K/kbar. Above 12 kbar, dT_0/dP increases more rapidly. The inverse correlation of the pressure 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 state as a function of pressure. 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. Bilbro and McMillan,[20] Machida,[21] and Gabovich and Shpigel[22] have developed a mean-field theory of the competition between superconductivity and CDW formation involving partial gapping of the Fermi surface. This theory has been used to analyze the pressure dependence of T_c and the structural transition temperature of the $A15$ compounds[20] and Chevrel-phase compounds of the type $Eu_{1-x}Sn_xMo_6S_8$.[23] The relation between T_c and T_0 proposed by Bilbro and McMillan can be written as $T_c^n T_0^{1-n}=T_{c0}$, where T_{c0} is the superconducting transition temperature in the absence of a CDW or SDW and n is the fraction of the Fermi surface that is ungapped below T_0. Assuming that $n\cong 0.6$ for URu_2Si_2 at $P=0$ and that $dT_{c0}/dP\cong 0$ (i.e., the strength of the superconducting pairing interaction is weakly dependent on pressure), then n decreases almost linearly with P and attains a value of ~ 0.4 at 12 kbar.

In summary, electrical resistivity measurements on the

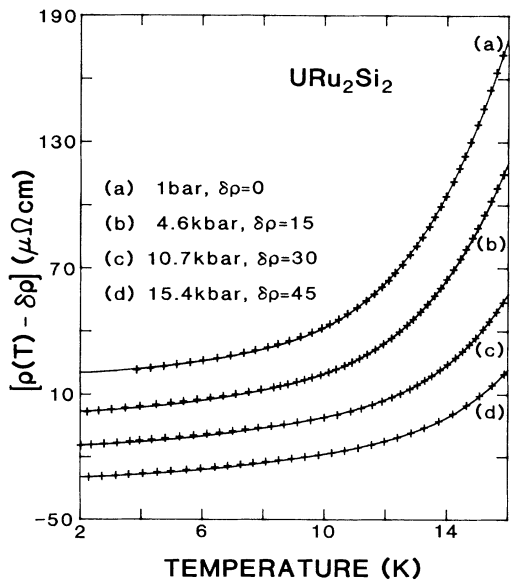

FIG. 6. Electrical resistivity $\rho(T)$—$\delta\rho$ versus temperature for URu_2Si_2 between 2 and 16 K at pressures of 1 bar, 4.6 kbar, 10.7 kbar, and 15.4 kbar. The lines represent fits of Eq. (1) to the data.

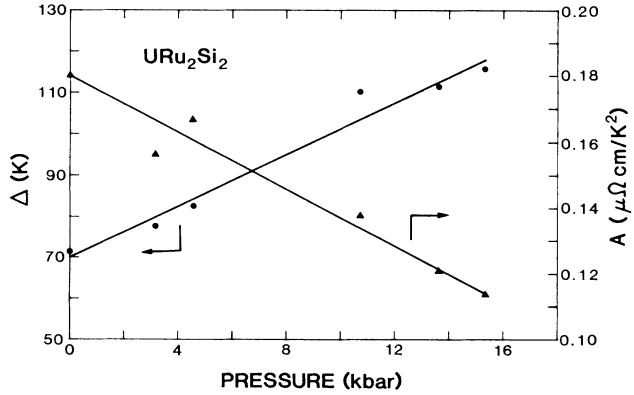

FIG. 7. Values of (a) the SDW or CDW energy gap Δ (in kelvins) and (b) the coefficient A of the T^2 contribution to $\rho(T)$ as a function of pressure for URu_2Si_2, as derived from fits of Eq. (1) to the $\rho(T)$ data.

heavy electron superconductor URu_2Si_2 reveal large inverse pressure dependences for T_c and T_0 that are consistent with the two transitions competing for regions of the Fermi surface. Scaling indicates at least two characteristic temperatures, one with little pressure dependence in the Kondo regime above 100 K and another with a strong pressure dependence corresponding to the formation of a coherent ground state. Fits to the data reveal that the energy gap which has been associated with a SDW or CDW transition increases with increasing pressure.

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