RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 33, NUMBER 9 1 MAY 1986

Anisotropic electrical resistivity of the magnetic heavy-fermion superconductor URu_2Si_2

T. T. M. Palstra, A. A. Menovsky,* and J. A. Mydosh

Kamerlingh Onnes Laboratorium der Rijks-Universiteit Leiden, 2300 RA Leiden, The Netherlands

(Received 24 December 1985)

Electrical-resistivity measurements $\rho(T,H)$ on *single-crystal* URu_2Si_2 have been performed between 0.3 and 300 K in fields up to 7 T. $\rho(T)$ is highly anisotropic with the 300-K values parallel to the a and c axes differing by a factor of 2. Both directions exhibit a $d\rho/dT < 0$ down to 80 K, followed by a steep decrease in ρ below 50 K. An upper antiferromagnetic transition is clearly discerned by a "Cr-like" anomaly at ≈ 17 K which also marks the onset of a strong, positive $\rho(H)$. Below 4 K, $\rho(T)$ is isotropic and goes to zero at the superconducting transition $T_C = 0.8$ K.

Very recently we have shown that the heavy-fermion system URu_2Si_2 has both a magnetic phase transition at 17.5 K and a superconducting transition at 0.8 K.[1] The upper phase transition was indicated by a λ-like anomaly in the specific heat and a maximum in the slope of the magnetization versus temperature. Superconductivity was detected by ac susceptibility, magnetization, and specific-heat measurements.[2] Our results on single-crystal samples demonstrated that both the normal and superconducting state properties were highly anisotropic. The c axis is the easy magnetic axis and very little magnetization was measured parallel to the a axis. With respect to the superconductivity, we observed no anisotropy within our experimental accuracy for the initial temperature slope of the critical field $(-\mu_0 dH_{c2}/dT|_{T\to T_C} = 4.4$ T/K) between the a and c axes. However, anisotropy does arise in H_{c2} for $T < 0.97T_C$, namely, the c axis shows a standard $H_{c2}(T)$ behavior (decreasing slope with decreasing temperature), while parallel to the a axis the slope $-\mu_0 dH_{c2}/dT$ continuously increases with decreasing temperature up to 14.5 T/K at 0.53 K.[1]

In order to gather additional information about this highly unusual heavy-fermion behavior we have studied the electrical and magnetoresistivity $\rho(T,H)$ of URu_2Si_2. All measurements were performed on high-quality single crystals between 0.33 and 300 K in magnetic fields up to 7 T. The electrical resistivity is highly anisotropic with its room-temperature value parallel to the a axis almost twice as large as parallel to the c axis. Above ≈ 20 K the overall $\rho(T)$ behavior for URu_2Si_2 is generic to many heavy-fermion systems with the exception of UPt_3.[3] The magnetic and superconducting transitions are clearly illustrated by a sharp jump in ρ at 17 K and $\rho \to 0$ at 0.8 K, respectively.

The single-crystal samples were grown with a specially adopted Czochralski triple-arc method.[4] No further heat treatment was given. Cylindrical samples of typical dimensions $\phi = 1$ mm, $l = 5$ mm were spark cut, parallel to the a and c axes, out of the same single crystal on which magnetization measurements were reported.[1] The electrical resistivity was measured with a standard four-point method using a dc current of 5 mA. The absolute value of the resistivity was determined at room temperature to better than 2% by measuring the diameter of the cylinders and the voltage drop at various distances over the entire length of the sample. The temperature was measured with calibrated carbon-glass and platinum thermometers. A dc magnetic field up to 7 T could be applied perpendicular to the current direction via a superconducting solenoid.

Figure 1 shows the overall temperature dependence of the electrical resistivity parallel to the a and c axes. The room-temperature resistivity is 330 $\mu\Omega$ cm parallel to the a axis and 170 $\mu\Omega$ cm parallel to the c axis. These values are about a factor 10 smaller than that reported by Maple *et al.* and a factor 1.5 smaller than reported by Schlabitz *et al.* on polycrystalline material.[2] Such differences illustrate the need of using single-crystal samples. The temperature coefficient $d\rho/dT$ is negative in both directions down to 80 K, but much "larger" along the a axis. Below 50 K the resistivity decreases rapidly to a residual resistivity of 32 $\mu\Omega$ cm—the same for both a and c directions.

Two distinct anomalies are observed in the resistivity behavior at low temperatures. In Fig. 2 we show these anomalies on an expanded scale. The inset of Fig. 2 clearly elucidates the superconducting transition $\rho \to 0$. The 50% point of the resistivity transition is at 0.70 K with a transition width between the 10 and 90% points $\Delta T_c = 0.2$ K. The second anomaly which is strongly anisotropic in magnitude occurs around 17 K and is reminiscent of the Néel temperature anomaly for $\rho(T)$ in pure Cr (Ref. 5)—a spin-density-wave (SDW) antiferromagnet. To better describe this critical behavior we have computer calculated the temperature derivative $d\rho/dT$ and present our results in Fig. 3. Note the negative divergence of $d\rho/dT$ at 17 K.

The temperature dependence between 1 and 17 K can accurately be described by using the formula appropriate for

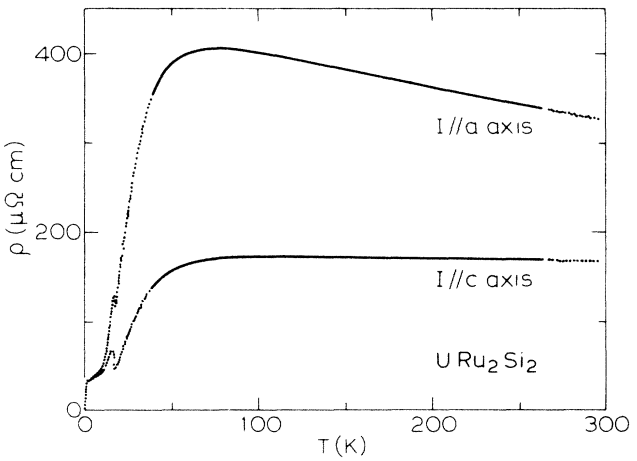

FIG. 1. Temperature dependence of the electrical resistivity of unannealed, single-crystal URu_2Si_2 parallel to the a and c axes.

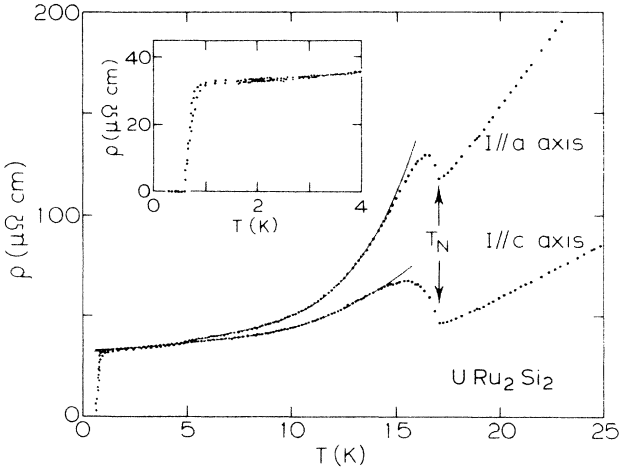

FIG. 2. Low-temperature resistivity of single-crystal URu_2Si_2 parallel to the a and c axes, showing the magnetic (T_N) and superconducting (T_C) phase transitions. The solid lines illustrate a best fit to Eq. (1). The inset shows an enlargement of the superconducting phase transition.

an energy gap (Δ) antiferromagnet[6] with an additional T^2 term appropriate for Fermi-liquid behavior,

$$\rho-\rho_0=bT(1+2T/\Delta)\exp(-\Delta/T)+cT^2 \ . \qquad (1)$$

Best fitting (see Fig. 2) gives $\Delta=90(68)$ K, $b=800(52)$ $\mu\Omega$ cm/K, $c=0.17(0.10)$ $\mu\Omega$ cm/K^2, and $\rho_0=33$ $\mu\Omega$ cm, parallel to the a (c) axis, respectively. Just above T_N, the resistivity has a power-law behavior $\rho-\rho_0\propto cT^2$ with $c=0.35(0.126)$ $\mu\Omega$ cm/K^2 parallel to the a (c) axis.

In Fig. 4 we plot the relative resistivity change $[\rho(T,H)-\rho(T,0)]/\rho(T,0)$, versus magnetic field parallel to the a and c axes at several fixed temperatures. Below about 20 K a large, positive magnetoresistivity emerges in both directions. As the temperature is reduced to 4 K, $\Delta\rho/\rho$ becomes larger reaching 10% in fields of 7 T. Above 25 K, the relative resistivity change is much smaller ($<\frac{1}{2}\%$). By fitting these curves to a parabolic field dependence, $\Delta\rho/\rho=aH^2$, a temperature-dependent coefficient $a(T)$ is obtained and shown in the insets of Fig. 4. From this coefficient, a characteristic temperature T_0 can be extrapolated (see Fig. 4) resulting in $T_0=15$ and 18 K parallel to the c and a axes, respectively. T_0 is in close correspondence with the magnetic transition temperature $T_N=17.5$ K determined from other measurements.

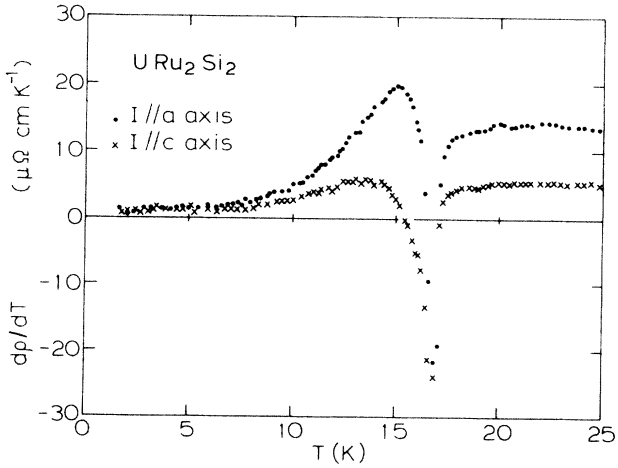

FIG. 3. Temperature dependence of the temperature coefficient $d\rho/dT$ of URu_2Si_2 parallel to the a and c axes.

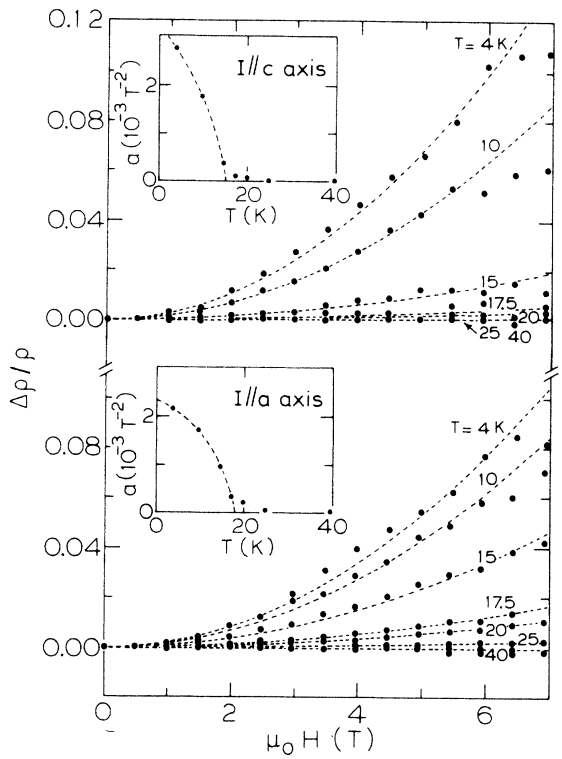

FIG. 4. Magnetic field dependence of the resistivity change $\Delta\rho/\rho$ of URu_2Si_2 parallel to the a and c axes at several fixed temperatures. The dotted lines are a fit to parabolic field dependences. The insets show the temperature dependence of the fit coefficients $a(T)$ defined as $\rho-\rho_0=a(T)H^2$.

Combining these resistivity results with our earlier measurements,[1] we now can calculate some microscopic parameters. Using the BCS relations given in Ref. 7, we need four independent parameters to form a self-consistent description of the superconducting state. We have chosen these parameters as the isotropic residual resistivity $\rho_0=31\times10^{-8}$ Ω m, $\gamma=50$ mJ/molK^2, $T_C=0.78$ K, and the isotropic initial slope of the upper critical field $-\mu_0(dH_{c2}/dT)_{T\to T_C}=4.4$ T/K. All were measured on various unannealed single-crystal samples. Accordingly, the relation

$$-\mu_0(dH_{c2}/dT)_{T\to T_C}=1.26\times10^{35}\gamma^2T_C/S^2+4780\gamma\rho_0 \qquad (2)$$

yields a Fermi surface area $S=1.88\times10^{20}$ m^{-2}. The "dirt parameter" $\lambda_{tr}=0.52$ indicates that we are neither in the pure nor dirty limit. This results[7] in a Fermi velocity $v_F=8.84\times10^3$ m/s, a mean free path $l_{tr}=2.62\times10^{-8}$ m, a BCS coherence length $\xi_0=1.56\times10^{-8}$ m, a London penetration depth $\lambda_L(0)=8.60\times10^{-7}$ m, and a Ginzburg-Landau parameter $\kappa_{GL}=73$.

Thus far no anisotropy is involved in the above calculation since (I) the initial slope of the upper critical field and the residual resistivity are isotropic and (II) we have used formulas[7] which are valid independent of the shape of the Fermi surface, i.e., they depend only on the total area S. The anisotropy does indeed affect the determination of the

Fermi momentum k_F because the spherical Fermi-surface approximation $S = 4\pi k_F^2$ is not valid for this highly anisotropic compound. Furthermore, we cannot even estimate k_F from S because of the anisotropically gapped Fermi surface due to the antiferromagnetic ordering at 17.5 K (see below). This gap will reduce S drastically without necessarily inducing large changes in k_F.

Therefore, in order to proceed a bit further, we have attempted two other approaches which are frequently used in heavy-fermion systems[8] to evaluate k_F. According to Friedel,[9] k_F can be determined by the number of conduction electrons per formula unit Z,

$$Z = \left[\frac{2(2l+1)\hbar x}{e^2\rho_{\max}}\right]^{3/4}\left[\frac{\Omega}{3\pi^2}\right]^{1/4} . \quad (3)$$

Here the angular momentum is $l=3$, the fraction of U atoms is $x=\frac{1}{5}$, $\rho_{\max}$ is the maximum resistivity, and $\Omega = 8.17\times10^{-29}$ m^3 is the volume per U atom. Using maximum resistivities parallel to the a and c axes of 400 and 170 $\mu\Omega$ cm, we calculate $Z=2.02$ and 3.83, respectively. This gives a Fermi momentum $k_F=(3\pi^2 Z/\Omega)^{1/3}$ being 0.90 Å^{-1} parallel to the a axis and 1.12 Å^{-1} for the c axis. These values are reasonable when compared to our second approach, the fully isotropic, free-electron case of three conduction electrons per U atom ($Z=3$), yielding $k_F=1.03$ Å^{-1} and $S=13.3\times10^{20}$ m^{-2}. This value of S, a high-temperature one, is much larger than the value calculated above from the BCS relation which gives a low-temperature limit. The difference suggests that only about 15% of the Fermi-surface area contributes to the superconductivity and is not removed by the antiferromagnetic order. Our result of $\approx 15\%$ remaining Fermi-surface area is somewhat smaller than the estimate based on the ratio of the electronic specific-heat coefficients $(\gamma)_{T\to T_c}/(\gamma)_{T\to T_N}\approx 28\%$.[1] Similarly, the Ginzburg-Landau parameter $\kappa_{GL}=73$ obtained above is larger than $\kappa_{GL}=33$ measured in an arbitrary direction.[1] The enhancement of the effective mass m^* relative to the bare mass m_0 can be determined by $m^*/m_0=\hbar k_F/v_F m_0$. As this enhancement is governed by the actual value of k_F, we cannot use the BCS relations to calculate k_F, as only a minor part of the Fermi surface is involved with the superconductivity and so no conversion can be made from the Fermi-surface area S to the Fermi momentum k_F. Here the estimates for k_F based upon the approaches of Friedel or the free-electron gas are perhaps more appropriate, providing there are no dramatic changes of the conduction electron density ($n\propto k_F^3$) in the high- and low-temperature limits. Thus, using an average value of $k_F=1.0$ Å^{-1}, we obtain $m^*/m \approx 130$.

It should be briefly mentioned here that a number of theoretical proposals[10] exist for the coexistence of an itinerant, SDW antiferromagnet and superconductivity. Such models usually consider part of the Fermi surface to be gapped by the SDW and the remaining portion to be available for the superconductivity.

In conclusion, rather than speculate on the causes or models of such behavior we simply summarize our experimental findings as follows: (i) At high temperatures ($T>150$ K) the resistivity is very large and highly anisotropic.[11] Also the temperature coefficient $d\rho/dT$, even when the phonons are not taken into account, is negative and hence Kondo-like. (ii) At 75 K a broad maximum appears in $\rho(T)$ and as the temperature is further reduced there is a dramatic decrease in the resistivity. This is usually associated with coherence effects in the Kondo lattice.[12] (iii) At 17.5 K both the a and c axes resistivities show a remarkable maximum/minimum behavior whose $d\rho/dT$ character closely resembles that of the SDW antiferromagnet Cr.[13] (iv) The $\rho(T)$ data below $T_N=17.5$ K can be nicely described by the theory of an energy-gap antiferromagnet with the addition of a T^2 term from Fermi-liquid theory. This T range also marks the onset of a strong, positive magnetoresistance proportional to H^2.[14] (v) Below 5 K the anisotropy in $\rho(T)$ completely disappears and superconducting order sets in at ≈ 1 K. We conclude from the U form factors found in the neutron measurements,[13] the size of the discontinuity in the specific heat at T_c, and the large value of the initial slope of the upper critical field, that both the magnetism and the superconductivity are carried by the same hybridized $5f$ electrons. The above represents a rather remarkable collection of experimental effects and is badly in need of a theoretical explanation.

We wish to acknowledge a series of stimulating discussions with G. J. Nieuwenhuys, P. H. Kes, and K. Bedell in Leiden at the Kamerlingh Onnes Laboratory, The Netherlands, and J. Kjems, C. Broholm, N. Hessel Andersen, and P. Lingard at the Risø National Laboratory, Denmark. M. Vlastuin has competently assisted us with the measurements. This work is part of the research program of the Nederlandse Stichting voor Fundamenteel Onderzoek der Materie (FOM).

---

*Also at Natuurkundig Laboratorium der Universiteit van Amsterdam, Amsterdam, The Netherlands.

[1] T. T. M. Palstra, A. A. Menovsky, J. van den Berg, A. J. Dirkmaat, P. H. Kes, G. J. Nieuwenhuys, and J. A. Mydosh, Phys. Rev. Lett. **55**, 2727 (1985).

[2] Similar results on polycrystalline samples have been obtained by W. Schlabitz, J. Baumann, B. Pollit, U. Rauchschwalbe, H. M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B **62**, 171 (1986); and by M. B. Maple, J. W. Chen, Y. Dalichaouch, T. Kohara, C. Rossel, M. S. Torikachvili, M. W. McElfresh, and J. D. Thompson, Phys. Rev. Lett. **56**, 185 (1986).

[3] See for an overview G. R. Steward, Rev. Mod. Phys. **56**, 755 (1984).

[4] A. A. Menovsky and J. J. M. Franse, J. Cryst. Growth **65**, 286 (1983).

[5] Ö. Rapp, G. Benediktsson, H. U. Åström, S. Arajs, and K. V. Rao, Phys. Rev. B **18**, 3665 (1978).

[6] N. Hessel Andersen, in *Crystalline Field and Structural Effects in f-Electron Systems*, edited by J. E. Crow, R. P. Guertin, and T. W. Mihalism (Plenum, New York, 1980), p. 373.

[7] T. P. Orlando, E. J. McNiff, S. Foner, and M. R. Beasley, Phys. Rev. B **19**, 4545 (1979).

[8]See, for example, H. R. Ott, H. Rudigier, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **50**, 1595 (1983); J. W. Chen, S. E. Lambert, M. B. Maple, Z. Fisk, J. L. Smith, G. R. Stewart, and J. O. Willis, Phys. Rev. B **30**, 1583 (1984).

[9]J. Friedel, Nuovo Cimento Suppl. **7**, 287 (1958).

[10]K. Machida, J. Phys. Soc. Jpn. **50**, 2195 (1981); **53**, 712 (1984); E. W. Fenton and G. C. Psaltakis, in *Superconductivity in d- and f-Band Metals,* edited by W. Buckel and W. Weber (Kernforschungszentrum Karlsruhe, Karlsruhe, 1982), p. 531.

[11]The resistivity is much larger along the *a* axis, yet the *c* axis is the easy axis for magnetization and along which the moments order.

[12]For some further discussion, see J. S. Schiling, Phys. Rev. B **33**, 1667 (1986).

[13]Preliminary results of neutron diffraction experiments at the Risø National Laboratory on a single-crystal sample of URu_2Si_2 have revealed very small, antiferromagnetically ordered uranium moments of order $0.01\mu_B$ below 17 K. J. Kjems, C. Broholm, W. L. Buyers, T. T. M. Palstra, A. A. Menovsky, and J. A. Mydosh (unpublished).

[14]In general, antiferromagnetically ordered materials also have a quadratic field dependence of their magnetoresistivity. See, for example, K. A. McEwen, in *Handbook of the Physics and Chemistry of Rare Earths,* edited by K. A. Gschneider, Jr. and L. Eyring (North-Holland, Amsterdam, 1978), Vol. I, p. 479.