ON THE 17.5K TRANSITION IN URu_2Si_2: THERMOPOWER AND ELASTICITY

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We report thermoelectric power (TEP) and elasticity measurements on the heavy Fermion compound URu_2Si_2. The TEP is positive and roughly independent of temperature between 300K and 120K, becomes increasingly negative below 70K, and displays a sharp change in slope at T_p=17.5K. At 15K there is a dramatic reversal in the TEP. The Young's modulus shows a strong dip near 18K, accompanied by an increase in the internal friction. The TEP and elastic anomalies are consistent with a Fermi surface instability at T_p, and we interpret our results in terms of charge or spin density wave formation.

INTRODUCTION

Of the rapidly growing class of heavy Fermion materials showing unusual electronic properties such as superconductivity, URu_2Si_2 is unique in that it shows evidence for multiple and coexisting Fermi-surface instabilities. Previous work on URu_2Si_2 has demonstrated that, in addition to exhibiting a large effective mass ($m^* \approx 25m_e$) and superconducting ground state below $T_c \approx 1.5K$, this material displays anomalous behavior in electrical resistivity, magnetic susceptibility, and specific heat near a "transition" temperature T_p=17.5K[1-5]. It has been suggested that the anomalous behavior near T_p results from antiferromagnetic ordering[3,5], or from the formation of a conduction electron charge density wave (CDW) or spin density wave (SDW)[1]. Recent neutron scattering experiments on a single crystal of URu_2Si_2 indicate that antiferromagnetic ordering occurs below T_p with a small ordered moment of $\sim 0.03\mu_B$ and a modulation along the (100) direction[6]. This antiferromagnetically ordered state appears to coexist with the superconducting state below T_c.

The low-field dc electrical resistivity of URu_2Si_2 shows a small peak just below T_p, with a functional form very similar to the resistance anomalies observed below the CDW transition in $ZrTe_3$[7] and below both CDW transitions in $NbSe_3$[8]. The strongest evidence for density wave (either CDW or SDW) formation in URu_2Si_2 comes, however, from specific heat measurements which show a BCS-like anomaly near 17.5K, consistent with a 40% destruction in Fermi surface area[1].

In this Communication, we report on careful thermoelectric power (TEP) and elastic constants measurements on URu_2Si_2 between room temperature and 4.2K. The TEP shows anomalous behavior near T_p consistent with CDW or SDW formation. Likewise, the Young's modulus Y and internal friction δ show sharp structure near T_p. From the magnitude of the elastic anomalies, the discontinuity in the specific heat at T_p can be calculated; the predicted discontinuity agrees within 7 to 17% of the experimentally determined value. Our measurements support the notion that the 17.5K transition is a density wave Fermi surface instability; however, we are not able to distinguish clearly between CDW or SDW formation.

EXPERIMENTAL

Our TEP and elastic constant measurements were performed on polycrystalline samples of URu_2Si_2 prepared by the usual arc melting method. The TEP was measured using a slow ac heating technique[9]. Small samples of URu_2Si_2 were suspended in vacuum between a pair of crystalline quartz blocks and 1-mil gold leads were attached to the sample ends with conductive silver paint. Both quartz blocks were wrapped with independent manganin heater wire which was used to ramp a temperature gradient of varying magnitude and direction across the sample. The temperature gradient was monitored with a chromel-constantan thermocouple. The thermally-induced EMF across the sample was amplified with a low noise amplifier and detected with an X-Y pen recorder.

Fig. 1 shows the TEP S(T) of URu_2Si_2 measured between 300K and 4.2K. Between room temperature and approximately 120K, the TEP is
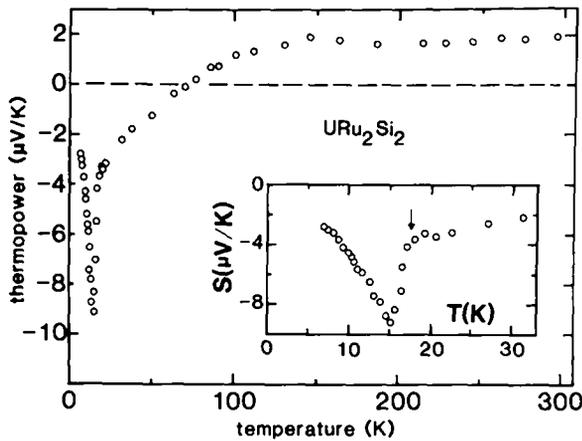
Fig. 1

Absolute thermoelectric power of URu_2Si_2 versus temperature. The inset shows in detail the behavior near the transition temperature, indicated by a vertical arrow.

temperature independent and approximately +2µV/K. Below 150K, the TEP tends toward zero at 70K and then becomes more negative with decreasing temperature. At 18K, the TEP has attained a value of -3.6µV/K. At T=17.5K, there is an abrupt change of slope in the TEP versus temperature curve, from dS/dT= 0.06µV/K at 18K to dS/dT= 2.4µV/K at 17K, signaling a thermodynamic phase transition at T_p=17.5K. Below 17K the TEP continues to increase in magnitude with decreasing temperature, until a dramatic change in direction occurs at T=15K. Below 15K, the TEP appears to smoothly approach zero with decreasing temperature. The inset in Fig. 1 shows the TEP behavior near the 17.5K transition in detail. As we shall discuss shortly, the change in slope in the TEP at 17.5K is good evidence for a Fermi surface instability where free carriers are removed at T_p. The additional sharp reversal in TEP at 15K is an apparently independent effect, not associated with any structural transition.

Young's modulus Y and internal friction δ were measured in URu_2Si_2 using a resonant vibration technique described in detail elsewhere[10]. A long, thin sample of polycrystalline URu_2Si_2 was rigidly clamped at one end and driven into vibration transverse to the long axis of the sample by a capacitively coupled electric field. A concentrated mass M of silver paint was added to the free end of the sample to produce a large-amplitude mechanical resonance in the kHz frequency range. The vibration was detected with an rf carrier signal at 373 MHz, and the drive/detection circuit was placed in a phase-lock-loop configuration that maintained the sample at the peak of the resonance and continuously monitored the resonant frequency f_r and amplitude A_r.

In the configuration used, the Young's modulus is given by[10]

$$Y = 4L^3M(2\pi f_r)^2/(t^3s) \qquad (1)$$

where L is the distance between the loading mass M and the clamped end of the sample, t is the sample thickness in the direction of flexure, and s is the sample width. A relative change in the internal friction $\Delta\delta/\delta$ is deduced from the inverse of the relative change in resonant amplitude, $\Delta A_r/A_r$.

Elastic measurements on URu_2Si_2 were performed in the temperature range 300K to 4.2K. At room temperature we estimate the absolute value of the Young's modulus to be $Y=2.5\times10^{11}$dyne-cm^{-2}, with a large uncertainty due to uncertainties in the geometrical factors associated with Eq. (1). Between room temperature and 4.2K, Y increased by a total of 5%. A broad internal friction peak, possibly due to grain boundary motion, was observed around 216K.

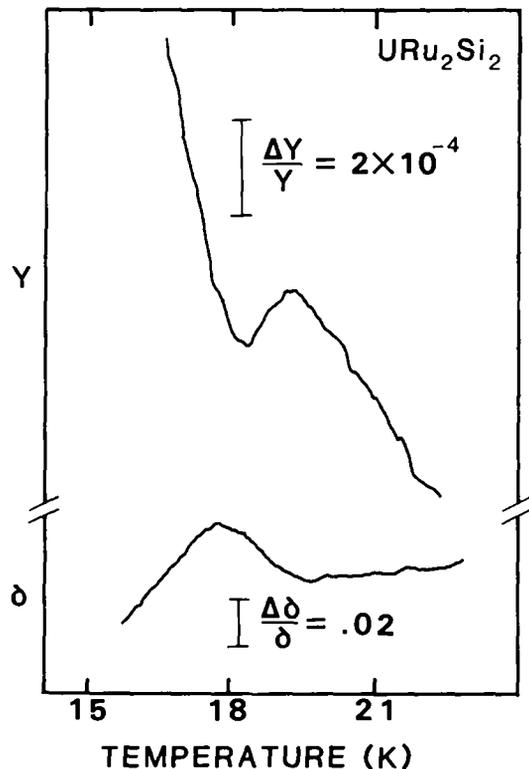Fig. 2 shows the detailed behavior of Y and

Fig. 2

Young's modulus Y and internal friction δ near T_p=17.5K in URu_2Si_2. The temperature scale refers to the sample holder temperature (see text).

δ near T_p. As is apparent from the figure, both Y and δ show well-defined anomalies near T=18K. There is some uncertainty in the temperature scale; the temperature indicated represents that of the sample holder, not the sample itself. We estimate that there might have existed a temperature differential of as much as one degree between sample and sample holder. Nevertheless, it is clear that near T_p there is

a dip in Y of magnitude $\Delta Y/Y=-2.2\times10^{-4}$ (determined by comparing Y at the bottom of the dip to the value of Y extrapolated from the high temperature behavior) and an associated maximum in δ. It is interesting to note that the maximum in δ does not correspond exactly to the minimum in Y, but is offset to a lower temperature by approximately 0.25K. No thermal hysteresis was observed in the temperature dependences of Y and δ.

DISCUSSION

The TEP of URu_2Si_2 at temperatures above the 17.5K transition is similar to that previously observed in other heavy Fermion compounds, in particular cerium based 4f systems[11,12]. In $CeCu_2Si_2$, for example, the TEP is positive at room temperature, gradually decreases with decreasing temperature and becomes negative near 80K, reaches a negative maximum near 20K, and then drops to zero as temperature goes to zero. This behavior has been attributed to the presence of an extremely narrow feature in the density of states N(E) near the Fermi level that is associated with the 4f electrons[12]. The very similar temperature dependence of the TEP in URu_2Si_2 for moderate temperatures (T>18K) would suggest a similar interpretation, i.e. the TEP reflects directly the narrow features in N(E).

In URu_2Si_2 the narrow feature in N(E) is expected to be of the order of 100K, according to transport, thermal, and magnetic measurements that have been previously reported[1-3]. As such, the resulting TEP is quite unlike that observed in conventional metals, where the bandwidth is typically several orders of magnitude larger. In the latter case temperature changes have little effect on the density of states at the Fermi level.

We analyze the TEP in terms of the semi-classical model where the diffusion TEP S_D is expressed in terms of the dc electrical conductivity σ as[13]

$$S_D = -(\pi/3)(k_B^2T/|e|)\sigma'(E_F)/\sigma(E_F) \qquad (2)$$

where the derivative is made with respcect to energy and is evaluated at the Fermi level E_F. The conductivity may be expressed in terms of Fermi surface parameters [14]

$$\sigma = e^2\tau_F v_F^2 N(E_F)/3 \qquad (3)$$

where τ_F is the relaxation time, v_F the mean Fermi velocity averaged over the Fermi surface, and $N(E_F)$ is the density of states at the Fermi level. In normal metals the band structure terms v_F and $N(E_F)$ are usually taken as independent of temperature, resulting in a TEP linear in temperature (Eq. (2)). This neglects phonon drag effects. Any deviation from linearity of the TEP in a simple metal then indicates that scattering mechanisms are coming into play which alter the TEP temperature dependence by way of $\tau_F(T)$ in Eq. (3).

The gradual change in TEP of URu_2Si_2 between 140K and 20K observed in Fig. 1 can be explained in terms of an extremely narrow resonance in N(E) near E_F. In URu_2Si_2, the origin of the resonance feature is not yet clear,

but it is probably tied to the narrow 5f electron band.

We consider the narrow resonance feature superimposed on a wide 6d band. The resonance is characterized by a maximum in the density of states at a characteristic energy Eo ($N(Eo)=Nmax$). At energies above and below Eo the density of states drops rapidly. Hence, at temperatures above the resonance width, the Fermi level will lie in the 6d band where no temperature dependent effects exist to influence the TEP; any deviation from linearity must be ascribed to a temperature dependent scattering mechanism. In particular, a temperature independent band structure and relaxation time inversely proportional to temperature would result in the flat TEP observed in Fig. 1 between 300K and 120K.

As the temperature is reduced the Fermi level will drop into the narrow resonance and the density of states at E_F will be very temperature dependent. With E_F greater than Eo, $N(E_F)$ will increase with decreasing temperature. Hence, $N'(E_F)$ in Eq. (2) will be negative, giving rise to a positive TEP. As the temperature is further reduced, $N(E_F)$ will decrease; $N'(E_F)$ then becomes increasingly positive, leading to a negative TEP.

The predicted behavior, i.e., a TEP that is constant and positive at high temperature, gradually falls to zero as T approaches $To \equiv Eo/k_B$, and gradually becomes more negative as the temperature drops further, is entirely consistent with the observed behavior in URu_2Si_2 at temperatures above 18K. Within the context of this simplified model, we may explicitly deduce the resonance width D≅140K.

Below 18K the TEP of URu_2Si_2 is dramatically different from that observed in other heavy Fermion materials. The abrupt change in slope at T_p=17.5K suggests a second order phase transition. From the anomaly in the specific heat, it has been estimated[1] that 40% of the Fermi surface is destroyed at this transition. The behavior of the TEP just below T_p indicates that a predominantly hole-like part of the Fermi surface is affected; the TEP becomes more negative as the electron-like portion of the Fermi surface becomes less compensated. Similar features in TEP are observed at T_p in materials which undergo CDW or SDW transitions[15,16]. From the behavior of the TEP, it is not possible to distinguish between CDW or SDW formation.

The behavior of the TEP at 15K is even more dramatic than that observed at T_p=17.5K. There the change in slope is extreme: $dS/dT=2.4\mu V/K^2$ just above 15K versus $dS/dT=-0.8\mu V/K^2$ just below 15K. Surprisingly, this dramatic reversal is not reflected in any other transport measurement, and we do not associate it with a secondary phase transition. Interestingly, the abrupt reversal is very similar to that observed just below the SDW transition in the Bechgaard salt $(TMTSF)_2PF_6$[16].

As mentioned previously, the low temperature phase in URu_2Si_2 is magnetic in nature. A number of other heavy Fermion materials show antiferromagnetic ordering[17]. In some cases there is no anomaly in the TEP at the Neel temperature ($CeAl_2$), in some cases there are minor changes in slope (CeB_6), and in other cases there are drastic changes in slope in the TEP at the ordering temperature ($CeRu_2Ge_2$).

We now consider the elastic anomalies near T_p. The dip in Young's modulus Y at T_p reflects the soft mode associated with the electronically driven phase transition. At a second order phase transition, Y is related to other measurable parameters by the expressions[18]

$$\partial T_p/\partial n_i = [(-\Delta Y/Y)T_p/Y\Delta C_p]^{\frac{1}{2}} \qquad (4)$$

$$\Delta \alpha_i = (\Delta Y/Y)/[Y\partial T_p/\partial n_i] \qquad (5)$$

where n_i and α_i are respectively the ith components of stress and thermal expansivity, and C_p is the specific heat. With knowledge of the uniaxial stress dependence of T_p the anomaly in Y allows a determination of the discontinuity in the specific heat at T_p. Taking for $\partial T_p/\partial n$ the values reported for the hydrostatic pressure dependence of T_p, 118 mK/kbar[4] and 125mK/kbar[1], Eq. (4) yields the values for ΔC_p=5.43 J/mole K and 4.84 J/mole K, respectively, in reasonable agreement with the measured discontinuity in the specific heat, ΔC_p=5.82 J/mole K[1]. From Eq. (5), we calculate a uniaxial thermal expansivity at T_p of $\Delta \alpha = -7.7 \times 10^{-6} K^{-1}$. It should of course be recognized that the polycrystalline nature of our samples implies that our measurements probably represent an average over all crystallographic directions.

As observed in Fig. 2, the maximum in the internal friction δ in URu_2Si_2 occurs at a slightly lower temperature than the minimum in Y. This behavior has been previously observed in materials undergoing a phase transition with an associated order parameter[19]. In general fluctuations in the order parameter lead to an internal friction peak that is symmetric about T_p, but additional relaxational processes can destroy this symmetry. The peak position of δ in URu_2Si_2 is evidence for such relaxations in the ordered state below T_p.

The observed behaviors in Y and δ near T_p in URu_2Si_2 are very similar to elastic anomalies associated with the CDW transitions in TaS_3, $NbSe_3$, and $K_{0.3}MoO_3$[10,20] and the SDW transition in Cr[21]. This, together with the TEP data discussed above, provides strong evidence for a collective mode CDW or SDW ground state in URu_2Si_2. Careful X-ray studies on single crystal samples of URu_2Si_2 should help to clarify the situation. Additional transport measurements on single crystal specimens, including Hall effect and tests for non-linear conductivity, are presently underway.

REFERENCES

1. 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)

2. 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)

3. W. Schlabitz, J. Baumann, B. Pollit, U. Rauchschwalbe,H.M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B62, 171 (1986)

4. F.R. Boer, J.J.M. Franse, E. Louis, A.A. Menovsky, J.A. Mydosh, T.T.M. Palstra, U. Rauchschwalbe, W. Schlabitz, F. Steglich,m and A. de Visser (to be published)

5. T.T.M. Palstra, A.A. Menovsky, and J.A. Mydosh, (to be published)

6. C. Broholm, J./K. Kjems, W.J.L. Buyers, P. Matthews, T.T.M. Palstra, A.A. Menorsky, and J.A. Mydosh (to be published)

7. S. Takahashi, T. Sambongi, J.W. Brill, and W. Roark, Solid State Commun. 49, 1031 (1984)

8. P. Monceau, N.P. Ong, A.M. Portis, A. Meerschaut, and J. Rouxel, Phys. Rev. Lett. 37, 602 (1976)

9. P.M. Chaikin and J.F. Kwak, Rev. Sci. Instrum. 46, 218 (1975)

10. L.C. Bourne and A. Zettl, Solid State Commun. 60, 789 (1986); L.C. Bourne and A. Zettl (to be published)

11. E. Gratz, E. Bauer, B. Barbara, S. Zemirli, F. Steglich, C.D. Bredl, and W. Lieke, J. Phys. F: Met. Phys. 15, 1975 (1985)

12. D. Jaccard, J. Flouquet, and J. Sierro, (to be published)

13. N.F. Mott, Proc. Roy. Soc. London A156, 368 (1936)

14. J.M. Ziman, in Principles of the Theory of Solids, (Cambridge University Press, London, 1972) p. 218

15. P.M. Chaikin, W.W. Fuller, R. Lacoe, J.F. Kwak, R.L. Greene, J.C. Eckert, and N.P. Ong, Solid State Commun. 39, 553 (1981)

16. K. Morentensen, Solid State Commun. 44, 643 (1982)

17. U. Gottwick, R. Held, G. Sparn, F. Steglich, K. Vey, W. Assmus, H. Rietschel, G. R. Stewart, and A.L. Giorgi, (to be published)

18. L.R. Testardi, Phys. Rev. B12, 3849 (1975)

19. M. Barmatz, L.R. Testardi, and F.J. DiSalvo, Phys. Rev. B12, 4367 (1975)

20. J.W. Brill, Mol. Cryst. Liq. Cryst. 81, 107 (1982)

21. S.B. Palmer and E.W. Lee, Phil Mag. 24, 311 (1971); K.W. Katahara, M. Nimalendran, M.H., Manghanani, and E.S. Fisher, J. Phys. F: Metal Phys. 9, 2167 (1979)