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Transport properties of the heavy-fermion compound $CeCu_6$ down to 14 mK

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We report precise measurements on $CeCu_6$ of the temperature-dependent term in the resistivity $\rho(T)$ and the thermoelectric ratio G down to 14 mK and the Lorenz number above 130 mK. Below about 30 mK, G and the coefficient of the T^2 term in $\rho(T)$ are nearly independent of temperature, which implies that the coherent state is attained at about this temperature. G is positive between 14 mK and 4.2 K.

$CeCu_6$ and $CeAl_3$ are the only materials which have large effective masses and do not exhibit magnetic ordering or superconductivity down to millikelvin temperatures.[1,2] Since $CeCu_6$ can be made in single-crystal form with relatively low residual resistivity ρ_0, it is very suitable for investigating the onset of the coherent heavy-fermion state at very low temperatures.[3]

Recently Reinders *et al.*[4] observed the de Haas–van Alphen effect in this system down to 20 mK. They concluded that $CeCu_6$ has a sharply defined Fermi surface in the coherent state at this temperature and that *all* the electron masses are renormalized by many-body effects. Lacroix[5] has estimated the coherence temperature T_c using the equation $T_c = D(T_K/D)^{3/2}$, where T_K is the Kondo temperature and D is the bandwidth. If we use $T_K \sim 5$ K and $D \sim 10^4$ K, we obtain $T_c \sim 100$ mK which is roughly in agreement with the de Haas–van Alphen results. The question of interest here is whether precisely measured transport properties of this system will define more clearly the temperature below which this coherent state is attained. For example, one would expect the temperature-dependent term in the resistivity $\rho(T)$ and the thermoelectric power S to exhibit the following simple Fermi-liquid behaviors in the coherent state: $\rho(T) = AT^2$ and $S = BT$.

Only a limited number of ultralow-temperature experiments which address the above question has been performed on single crystals. Amato *et al.*[6] reported for $\rho(T)$ a T^2 behavior between 30 and 100 mK, and Sumiyama *et al.*[7] found a deviation from the T^2 dependence below 100 mK. Sumiyama *et al.*[7] and Coleridge[8] investigated the temperature dependence of the magnetoresistance and related the positive magnetoresistance to the formation of the coherent state below about 150 mK. Penny *et al.*[9] reported a sharp negative minimum in the Hall coefficient near 300 mK which they interpret as being the onset temperature of the coherent state. With regard to S, its dependence on crystal orientation and chemical composition has been investigated at higher temperatures for $Ce_xLa_{1-x}Cu_6$.[10,11] At lower temperatures, Amato *et al.*[6] have reported only that S is still positive down to 50 mK.

We report here measurements of the thermoelectric ratio G and $\rho(T)$ down to 14.5 mK along the [010] axis of one $CeCu_6$ sample. For the thermoelectric ratio G, we have

$$G = \left.\frac{j}{\dot{q}}\right|_{E=0} = \frac{S}{LT} , \qquad (1)$$

where j is the electrical current density, $\dot{q}$ is the heat current density, and L is the Lorenz number.[12] G is simpler to measure than S, since it is not necessary to measure the temperature gradient along a sample. If we have $S \propto T$ in the coherent state, then Eq. (1) implies that G will be temperature independent *only* if L is temperature independent. Since prior work on $CeCu_6$ shows that L is not constant below 1 K,[6,13] we measured L at several temperatures above 130 mK in order to convert our G measurements into S using Eq. (1).

A single crystal of $CeCu_6$ was grown by the Czochralski pulling method from a BN crucible. A planar sample of 1 mm thickness was made which had an irregular shape of maximum dimensions 4×20 mm^2. The long axis of the sample was parallel to the [010] direction, and the [100] direction was inclined about 15° from the axis of the 1 mm dimension. In order to make reliable electrical and thermal contact to this rather brittle sample, we used 1.0–1.5-mm-diameter potassium wires of 99.9% purity. Potassium cold welds beautifully at room temperature to several metals including Cu, and thus, potassium is a very suitable "soldering" material for transport measurements in the mK temperature range. The self-heating due to the naturally occurring radioisotope ^{40}K was negligible in our experiment.

The two potassium current leads were attached at opposite ends of the 20 mm axis of the sample. These leads also provided thermal contact to the refrigerator at one end of the sample and to the heater for the G measurements at the other end. For each of three runs, two potassium voltage leads were attached to the sample in different locations separated by 6–10 mm. The shape of the sample was more regular between these leads. We observed no significant dependence of ρ upon the position of these leads. The potassium leads were attached to the sample inside an Ar-filled glove box, and the assembly was placed inside a sealed vessel for transport to and mounting on a dilution refrigerator.

The techniques for making precise G measurements and very precise ρ measurements (0.02 ppm) are well established at Michigan State University and have been applied to the alkali metals for several years.[14] The four-probe reference resistor (30 $\mu\Omega$) was made from an O_2-annealed Ag(0.1% Au) alloy. The large S, which we have observed in $CeCu_6$, means that small temperature fluctuations can induce rather large voltage noise across the sample. Thus, the precision of our ρ measurements was about 50 ppm. Due to the irregular shape of the sample, it was difficult to determine its ρ at room temperature so we assumed $\rho = 70\ \mu\Omega\,\text{cm}$, as determined by others.[15]

In runs I and II we used a refrigerator which had been used extensively at MSU for alkali-metal studies.[16] It operated above 70 mK and used Ge resistance thermometers. In run II we measured directly the temperature difference ΔT along the sample in order to determine S and the thermal conductance, the latter being used with the resistance and the Wiedemann-Franz law to compute L. We attached an uncalibrated Ge resistance thermometer to each potassium voltage lead and calibrated the thermometers *in situ*. Between 0.1 and 2 K, we estimate the accuracy of the ΔT measurements to be $\pm 2\%$. In run III we used a different refrigerator which could cool samples below 15 mK. Above and below 50 mK, respectively, Ge resistance and cerium magnesium nitrate susceptibility thermometers were used. The latter was calibrated using a superconducting fixed-point device (SRM-768, U.S. National Bureau of Standards). The data from all three runs were in good agreement.

In Fig. 1(a) we show the temperature dependence of L/L_0 where L_0 is the Sommerfeld value of L. Below 2 K the estimated accuracy of these measurements is $\pm 4\%$. The point near 4 K is much less accurate, as indicated. Near 4 K our value of L is much larger than L_0 and is in rough agreement with that of Peysson, Salce, Ayache, and Bauer.[13] A similar enhancement of L/L_0 has been reported in the analysis of the point-contact spectrum in the self-heating regime.[17] This enhancement of L could be due to large energy-dependence Kondo scattering processes and also to significant heat conduction by the lattice. Bauer, Gratz, and Peysson[18] suggest that lattice conduction is the cause of this enhancement. Below about 0.6 K, we have $L < L_0$, a negative deviation which is significant on the scale of the accuracy of these data. This deviation of L from L_0 is in semiquantitative agreement with the theoretical predictions of Nakamura, Kawakami, and Okiji.[19] Although the L data (along [100]) of Amato *et al.*[6] below 0.4 K are similar to ours, they do not agree within the stated uncertainties. Our results strongly disagree with those of Peysson *et al.* who obtained $L/L_0 = 2$ even below 0.15 K. Perhaps this disagreement is due to their rather large residual resistivity (32 $\mu\Omega$ cm) which would enhance the relative contribution of lattice thermal conduction.

In Fig. 1(b) we present our resistivity measurements made in run III, where $\rho_0 = 4.9\ \mu\Omega\,\text{cm}$. These data are very similar to those of Ōnuki *et al.*[15] for samples which were also made with BN crucibles by the Czochralski method.

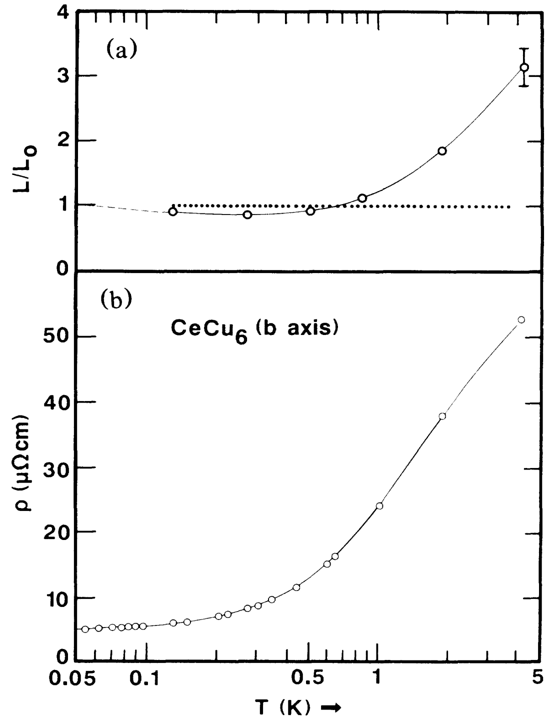

FIG. 1. (a) The Lorenz number L, normalized by L_0, vs T. The solid curve is an interpolation between the data points, and the dashed curve is an extrapolation of the data to lower T, where $L = L_0$ is assumed to occur at about 20 mK. The dotted horizontal line corresponds to $L = L_0$. (b) The resistivity ρ vs T above 50 mK. The solid curve is a guide to the eye.

In Fig. 2 we present data for $d\rho/dT$ and S above 14 mK. Using Eq. (1), we computed S from our G data and the curve drawn through the L data in Fig. 1(a). Below about 30 mK the $d\rho/dT$ data have clearly attained a linear T dependence, which corresponds to the expected T^2 behavior in $\rho(T)$. Thus, for $\rho(T)$, the coherent state is essentially fully formed below 30 mK. Likewise, S appears to have attained the expected linear T dependence below about 30 mK. Up to about 150 mK, $d\rho/dT$ and S deviate from their T^1 behaviors in a rather similar fashion. The maximum in S occurs at about the same temperature as the sharp minimum which was observed in the Hall coefficient.[9] Near 4 K the two data points represented by crosses were measured independently with a superconducting chopper amplifier and calibrated carbon resistors on a sample whose preparation, orientation, and ρ_0 were very similar.[20] Our data agree reasonably well with these two points. As mentioned earlier, Amato *et al.*[6] observed that S was positive down to 50 mK along the [100] direction. Similarly, our S data are positive above 14 mK along [010].

In order to show more clearly the temperature dependence and the reproducibility of our data, we plot $A[=(1/2T)(d\rho/dT)]$ and G vs T in Fig. 3. Clearly, A and G are very close to being constant below 30 mK, and measurements below 30 mK are necessary to obtain accurate values for these constants. The limiting values of A

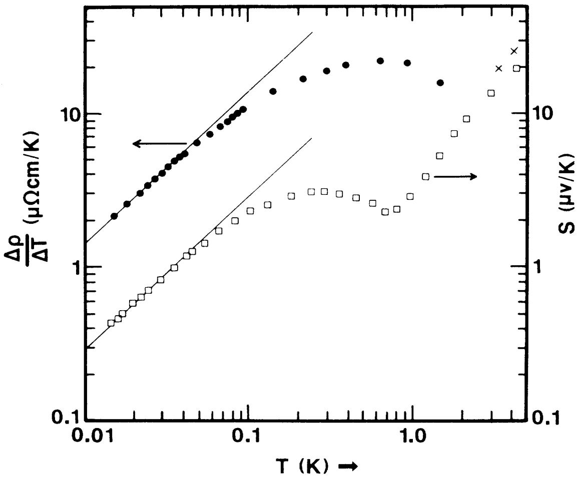

FIG. 2. Plots of $d\rho/dT$ and S vs T. Note the two ordinate scales. The straight lines correspond to a linear temperature dependence. The crosses are explained in the text.

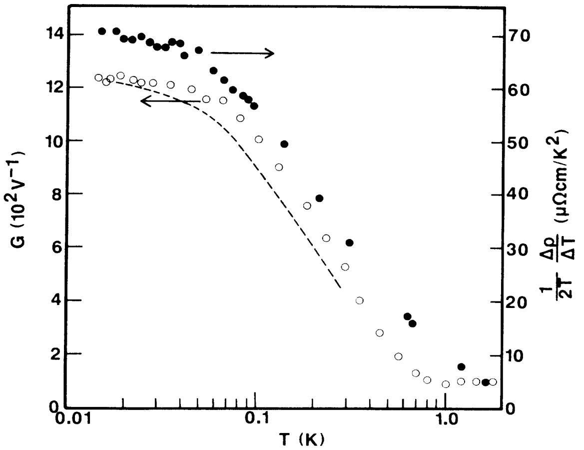

FIG. 3. Plots of $(1/2T)(d\rho/dT)$ and G vs T. Note the two ordinate scales. The dashed curve is explained in the text.

and G are 71 $\mu\Omega\,\mathrm{cm/K^2}$ and 1220 V^{-1}, respectively. For a sample of the same orientation and similar ρ_0, Sumiyama *et al.*[7] performed a T^2 fit to their ρ data below 0.1 K and obtained $A=42$ $\mu\Omega\,\mathrm{cm/K^2}$. In order to compare their results for A with ours, we also performed such a fit to our data below 0.1 K and obtained $A=62$ $\mu\Omega\,\mathrm{cm/K^2}$. This discrepancy is not understood at the present time.

As mentioned earlier, S and $d\rho/dT$ exhibit rather similar behaviors below about 150 mK in Fig. 2. This apparent similarity can be explored further by comparing A and B $(=S/T)$ in Fig. 3. Since $L\neq L_0$ for our sample, the T dependences of G and B are not the same. Using Eq. (1), we obtain that $B=GL$. The quantity B/L_0 is plotted in Fig. 3 as a dashed curve. The shape of this curve depends somewhat on the interpolations and extrapolation of the L data shown in Fig. 1(a). Below 130 mK we have assumed that L extrapolates smoothly to L_0 at about 20 mK. The ordinates for the A and B data were scaled in Fig. 3 so that their similar temperature dependences would be evident below about 0.3 K.

Although further L measurements are required below 130 mK in order to firmly establish the behavior of B, the observed behavior of A and B should place useful constraints on models which predict the temperature dependence of these transport coefficients in the coherent heavy-Fermion state. Nakamura *et al.*[19] have calculated the energy dependence of the electron scattering time $\tau(\varepsilon)$ and have obtained that $d\tau/d\varepsilon<0$ for $\varepsilon\approx\varepsilon_F$. In their calculation a gaplike structure in the density of states was placed slightly above ε_F. Our positive values of S are in agreement with their result, since $S\propto -d\tau/d\varepsilon$ at ε_F.[12] It will be interesting to see if further calculations can reproduce the temperature dependence we see for A and B.

In summary, our experiments show that the coefficient of the T^1 dependence of the positive thermoelectric power and the coefficient of the T^2 dependence of the resistivity are constant only below 30 mK. Thus, to fully establish the coherent heavy-fermion state, $\mathrm{CeCu_6}$ must be cooled below 30 mK. These coefficients exhibit similar temperature dependences below about 0.3 K.

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