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New Ce heavy-fermion system: $CeCu_6$

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We have discovered a new heavy-fermion system, $CeCu_6$, with a large susceptibility ($\chi = 0.027$ emu/mole G at 1.5 K) and a large, temperature-dependent specific heat γ below 10 K that is 840 mJ/mole K^2 at 1.8 K and extrapolates to 1.6 J/mole K^2 at $T = 0$ in analogy with $CeAl_3$. High-field specific-heat measurements agree almost perfectly with a narrow-band picture first proposed for UBe_{13}.

Since the discovery[1] by Steglich *et al.* in 1979 of bulk superconductivity in $CeCu_2Si_2$, an f electron system with highly correlated electrons having effective masses about 200 times the bare-electron mass, a great deal of interest has focused on these so called "heavy-fermion" systems. As a class of materials, the seven heavy-fermion systems known[1-7] to date all contain f electrons and have large specific-heat γ values ($C = \gamma T + \beta T^3$, from which the large effective masses are calculated), a large (> 4 Å) $4f$ or $5f$ atom separation, a high-temperature susceptibility that follows a Curie-Weiss law, a large low-temperature susceptibility, and usual temperature dependence in their resistivity. A possible division of these seven sytems at low temperatures in between those that go superconducting,[1-3] magnetic,[4-6] or neither[6,7] as is the case in $CeAl_3$ [$\gamma(T=0) = 1.6$ J/mole K^2] and $U_{0.32}Np_{0.68}Be_{13}$ [$\gamma(T=0) > 1.1$ J/mole K^2]. All but three[3-5] of these systems have γ's that are rapid functions of temperature, varying from under 200 mJ/mole K^2 at 10 K to over 1000 mJ/mole K^2 below 1 K.

We have focused a search for other heavy-fermion systems on Ce and U compounds with large f-atom separations that are not known to order magnetically at low temperatures. We report here on $CeCu_6$, an orthorhombic structure first solved[8] at Los Alamos with Ce surrounded by a cage of 19 Cu atoms and a Ce-Ce spacing of 4.83 Å. We have measured the resistivity from 1.4 to 300 K, dc susceptibility from 1.4 to 100 K, and specific heat from 1.8 to 38 K in zero and 11 T applied field on material prepared by arc melting together the pure constituents and characterized as single phase by x-ray powder diffraction. Measurements of the ac susceptibility showed no evidence of superconductivity down to 0.040 K.

The resistivity data measured by a standard four probe technique are shown in Fig. 1. The data have a minimum at about 29 K and a broad maximum centered at 11 K, with a sharp drop at lower temperatures. The dc susceptibility, shown in Fig. 2, was measured in a Faraday balance on a 69 mg piece of the arc melted button. Susceptibility data for $CeCu_6$ above 77 K have been reported[9] in the literature and agree with our data in the region of overlap. The magnitude of our measured susceptibility for $CeCu_6$ at 1.5 K, 0.027 emu/mole G is enormous—approximately twice the value[2] found for UBe_{13}. Also noteworthy in our susceptibility data is the definite feature at about 30 K, which correlates well with the resistivity minimum.

The specific heat in zero field from 1.8 to 38 K, shown in Fig. 3, was measured in a small sample calorimeter on the same piece of material used for the susceptibility measurements. At temperatures below 8 K we see the rapid increase in C/T that is characteristic of most heavy-fermion sytems. In fact, these low-temperature data are within a few percent of published specific heat results[7] for $CeAl_3$. Thus, we expect the $\gamma(T=0)$ value for $CeCu_6$ to also be about 1.6 J/mole K^2.

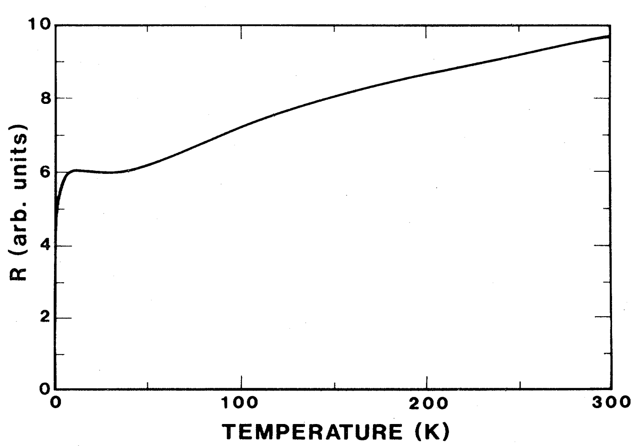

FIG. 1. Resistivity vs temperature for $CeCu_6$. Note the minimum at around 29 K.

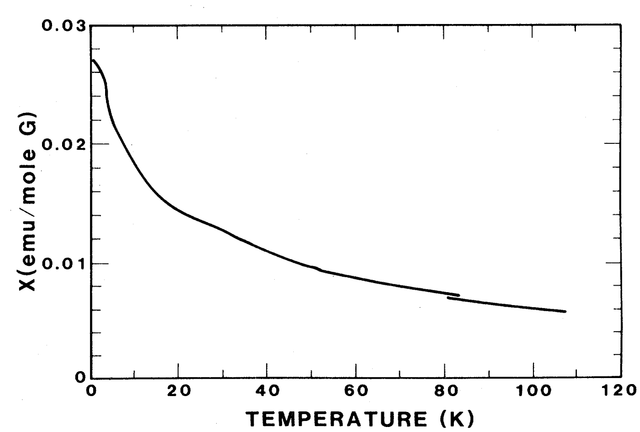

FIG. 2. Susceptibility vs temperature for $CeCu_6$. Note the slight hump at about 30 K. The mismatch around 77 K is instrumental in origin.

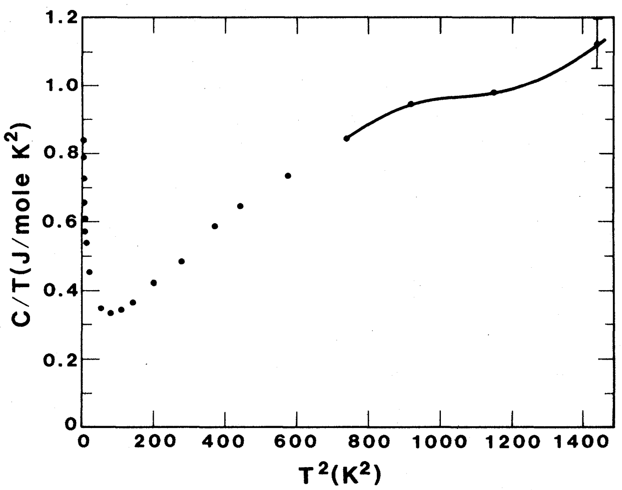

FIG. 3. Specific heat of $CeCu_6$. The rapid increase in C/T below 8 K is characteristic of most heavy-fermion systems. The data below 8 K are within a few percent of those (Ref. 7) for $CeAl_3$. The lowest-temperature point is at 1.85 K. The absolute accuracy of the data is ±3% below 20 K, and ±7% at 38 K. However, the relative precision of the data at high temperatures is ±2%, i.e., the shoulder observed around 30 K is a real feature.

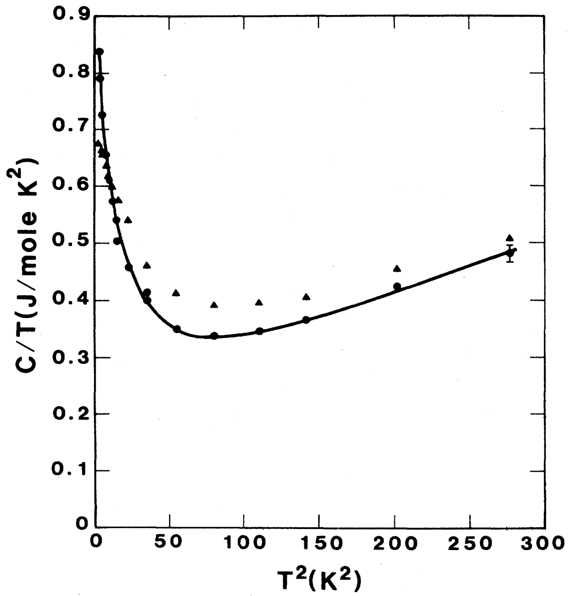

FIG. 4. Zero field (dots) and 11 T (×'s) specific heat for $CeCu_6$. The line drawn through the zero-field data is solely to serve as a guide to the eye.

When plotted as C vs T, these specific heat data for $CeCu_6$ show no evidence for a low-temperature peak, contrary to the results[1] for $CeCu_2 Si_2$ and for[10] UBe_{13}, but rather a broad shoulder below 4 K.

The higher-temperature data plotted as C/T vs T^2 in Fig. 3 do show a washed out peak, or shoulder, around 30 K that correlates with the structure seen in the resistivity and susceptibility data. A possible explanation for this anomaly is low-lying crystal field excitations of the Ce^{3+} ground state. (The assignment of the ground state as Ce^{3+} is based on high-temperature susceptibility, Ref. 9.)

In order to further understand the nature of the highly correlated electrons in heavy-fermion systems in general, and in $CeCu_6$ in particular, we have measured the specific heat from 1.8 to 17 K in an 11-T applied magnetic field. These results are shown in Fig. 4. There is a large decrease in C caused by the 11-T field at low temperatures (−19% at 1.8 K), followed by a crossover (i.e., zero field effect) at about 3.3 K to an increase in C with field at higher temperatures. This increase is also quite large (+19% at 7.4 K), but falls off at still higher temperatures (+5% at 16.7 K). These starting results follow almost perfectly what one would expect from a simple narrow-band (30–40 K wide) model. Such a model cannot only explain the temperature-dependent γ in zero field but has also been used[11] in the case of UBe_{13} in 11 T to predict, due to band splitting, an increase in C field "above 3.5 K which is a maximum around 6 K falling off at higher temperatures, with a decrease in the specific heat below 3.5 K." In the case[11] of UBe_{13}, no net decrease in C with field was found below 3.5 K; merely the observed increase became smaller in this region, while the other predicted features (maximum around 6 K and tapering off of the field-induced increase in C at higher temperatures) *were* observed.

We believe the almost perfect agreement between a narrow-band model for the specific heat and the measured data, both in zero *and* applied field, for $CeCu_6$ is strong evidence for the single-particle, narrow-band approach and contradicts the many-body, paramagnon view—at least as a first approximation. This is borne out by recent point-contact tunneling spectroscopy results on UPt_3 (Ref. 12) and $CeCu_2Si_2$,[13] where sharp features in the density of states are observed of 6 and 1.5 meV (or 70 and 20 K) widths, respectively.

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