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Kondo-lattice—mixed-valence resistance scaling in heavy-fermion CeCu₆ under pressure

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From measurements of the electrical resistance of heavy-fermion $CeCu_6$ subjected to hydrostatic pressures, we infer a continuous pressure-induced transition from Kondo-lattice to more strongly mixed-valence behavior. The resistance is found to scale over an appreciable temperature interval with a pressure-dependent characteristic temperature that reflects this transition. Similarities in the resistive behaviors of $CeCu_6$ and $CeAl_3$ are discussed.

There now appears to be a distinct class of materials characterized by huge electronic specific heats (γ's) and correspondingly large effective electronic masses.[1] Each of the known representatives of this class of heavy-fermion materials contain either $4f$ or $5f$ elements. Whether the large-γ enhancements are a result of the intrinsic electronic band structure or many-body effects remains an open question. What is clear, however, from thermal variations in properties such as specific heat, magnetic susceptibility, and resistivity is that electronic excitations from the heavy-mass ground state develop on a characteristic temperature scale that is small, typically much lower than phonon energies. Such observations suggest that sufficient energy could be supplied by routine high-pressure techniques to perturb noticeably the electronic spectrum of these materials.

Recently, $CeCu_6$ was discovered[2] to have an electronic specific heat $\gamma(T=0)$ of about 1.6 J/mole K^2, making it one of the largest-γ materials known. Many of the physical properties of $CeCu_6$ are strikingly similar to those[3,4] of $CeAl_3$, which for some time has been considered a proto-typical "Kondo-lattice" system. Percheron *et al.*[5] have shown that Kondo-like thermal variations in the electrical resistivity of $CeAl_3$ are strongly pressure dependent. Although a complete analysis of their data was hampered by large pressure-induced irreversibilities in the resistivity value of $CeAl_3$, they were able to conclude that the cerium $4f$ level moved closer to the Fermi level with applied pressure.

The troublesome irreversibility seen by Percheron *et al.*[5] was attributed to pressure-induced formation of an allotropic form of $CeAl_3$, with unknown properties. Because of the possibility that $CeCu_6$ might not suffer this complication and consequently represent a cleaner system to study, we have measured the effect of pressure on the electrical resistance of $CeCu_6$. As we will show, in many ways the analogy between $CeAl_3$ and $CeCu_6$ persists at high pressures. However, because the resistance of $CeCu_6$ is reversible with pressure, a somewhat different and more straightforward interpretation of the high-pressure properties of $CeCu_6$ is possible.

Four-probe ac resistance measurements were made on a single crystal of $CeCu_6$ grown by slow cooling from a melt. The direction of current flow with respect to crys-

tallographic axes was not determined. X-ray analysis showed only lines characteristic of the $CeCu_6$ orthorhombic structure. The resistance was determined over the temperature interval 1—300 K and at hydrostatic pressures up to 18 kbar, generated by a self-clamping pressure cell whose operation has been described in detail elsewhere.[6]

We show in Fig. 1 the temperature-dependent resistance of $CeCu_6$ at five fixed clamp pressures. Numbers beside each curve correspond to the order in which pressure was applied. Curve 5 fits smoothly between curves 2 and 3, indicating the absence of pressure hysteresis seen in $CeAl_3$. There are several additional qualitative features of these data that should be noted.[7] At $P=0$, the resistance decreases with decreasing temperature until it reaches a shallow minimum centered around 195 K. Near 15 K, a well-defined resistive peak develops below which the resistance falls rapidly. This behavior is similar to that of $CeAl_3$ except the resistance of $CeAl_3$ shows no minimum below room temperature and the peak for $P=0$ occurs at 35 K. With increasing pressure, the peak resistance of $CeCu_6$ becomes less prominent and is shifted to higher
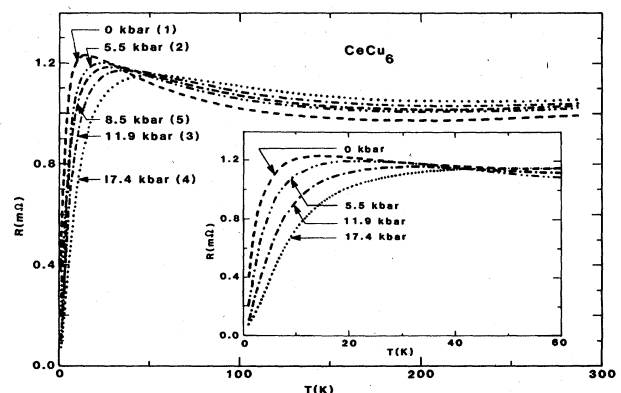

FIG. 1. Resistance vs temperature for $CeCu_6$ at five different clamp pressures. Numbers in parentheses indicate the chronological order in which the curves were acquired. The inset shows an expanded view of the low-temperature resistance. Curve 5 is not shown for clarity only.

temperatures such that the temperature difference between the peak and minimum remains essentially constant. For temperatures greater than ~ 70 K, the resistance increases monotonically with pressure; but $R(1$ K$)$ drops approximately exponentially, reaching a value at 17.4 kbar that is only about 18% of the ambient pressure resistance. Again, pressure-induced variations in the resistive behavior of $CeCu_6$ are much like those[5] in $CeAl_3$.

For $P=0$ the negative $\partial R/\partial T$ above 15 K suggests a "Kondoesque" resistivity.[4] We adopt this point of view, in addition to postulating that the cerium $4f$ level is fully occupied or nearly so and forms a very narrow band close to the Fermi level. Therefore, for pressures near ambient, the resistance for $T > 15$ K is dominated by Kondo-impurity-like scattering of the $4f$ electrons. At lower temperatures, the periodic array of cerium atoms scatters coherently, with a corresponding decrease in the resistance. Such behavior is a characteristic of "Kondo lattices."[8] With increasing pressure the cerium $4f$ level is broadened and brought more nearly into coincidence with the Fermi level, thereby driving the system into a weakly mixed-valent regime while still retaining part of its Kondo-like character.

We can estimate crudely a pressure-induced valency change by considering Friedel's expression[9] for maximum incoherent scattering:

$$\rho_{max} \leq (4\pi c\hbar/e^2 k_F v)(2l+1) , \qquad (1)$$

where l is the cerium $4f$ angular momentum, c is the concentration of cerium atoms, k_F is the Fermi momentum, and v is the number of electrons per atom. Assuming a free-electron value for k_F (as done by Andres et al.[3] for $CeAl_3$) and approximating the $CeCu_6$ unit cell as cubic, we arrive at an expression for the rate of change of ρ_{max} with pressure:

$$\partial \ln\rho_{max}/\partial P = -(\tfrac{1}{3}B + \partial \ln v/\partial P) , \qquad (2)$$

where B is the bulk modulus taken[10] to be 500 kbar. Equating ρ_{max} with the maximum value of the measured resistance R_{max}, we find $v \simeq v_0 + 0.005P$ (kbar). Therefore, we estimate the valency change induced by our highest pressure to be less than 0.09 electron. Because we have ignored coherency effects that are present even at R_{max}, this crude approximation undoubtedly overestimates the valency change. However, our results for $R_{max}(P)$ do indicate that, already at 17.4 kbar, $\partial R_{max}/\partial P$ appears to be decreasing, which would imply an approach to saturation in the maximum valency change. Such an observation is consistent with many experiments on cerium-based compounds that show evidence for a maximum cerium valency of about 3.2.[11] We emphasize, though, that arguments given here have completely neglected crystalline-electric-field (CEF) effects which might be important.[12]

As indicated, a maximum in the resistance near 15 K (at $P=0$) signals the onset of coherency[8] among the cerium atoms such that they no longer act as independent scatterers. We show in Fig. 2 the resistance normalized to its peak value (i.e., normalized to constant valency) as a function of reduced temperature $T/T_{max}(P)$, where $T_{max}(P)$ is the temperature at which the maximum resis-
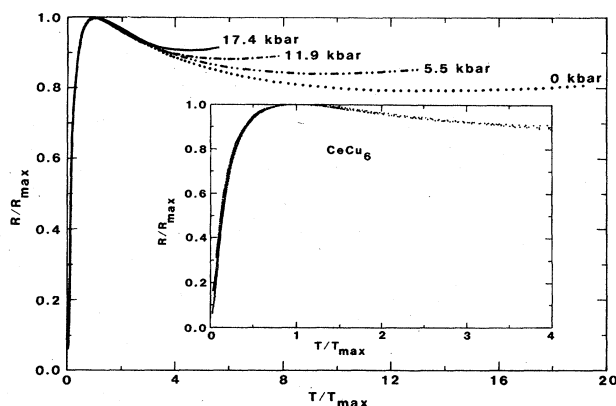

FIG. 2. Resistance normalized to its maximum value as a function of reduced temperature T/T_{max}, where T_{max} is the temperature at which the resistance peak occurs. The inset is an expanded view of the scaled resistance for temperatures near T_{max}.

tance occurs. [$T_{max}(P)$ changes by over a factor of 3, increasing approximately linearly from ~ 15 K at $P=0$ to ~ 51 K at 17.4 kbar.] The normalized resistance scales as $T/T_{max}(P)$ to temperatures in excess of $T_{max}(P)$. We attach no particular significance to the choice of T_{max} as the scaling parameter except to note that it must be proportional to some temperature that characterizes the underlying physics. The observation of scaling implies that the physical mechanisms describing the resistance at our lowest temperatures remain dominant to temperatures exceeding T_{max}, even while pressure is modifying the electronic spectrum near the Fermi energy. This suggests that a single characteristic temperature should be used to describe resistive behavior in both the Kondo-lattice and mixed-valence regimes of $CeCu_6$. Although resistive scaling was predicted by Kaiser and Doniach[13] for scattering from localized spin fluctuations in dilute alloys, their theory was criticized subsequently by Mills[14] and Jullien et al.[15] as being in general not valid. Furthermore, one would not expect the Kaiser-Doniach theory[13] to be applicable to $CeCu_6$ at low temperatures. To our knowledge there is no theory that adequately explains our results. The breakdown in scaling for $T \gg T_{max}$ might be expected,[16] since it is in this temperature range where CEF's affect the scattering. Whether similar effects would be seen in $CeAl_3$, if it were not to transform to an allotropic phase under pressure, is unknown but conceivable.

To summarize, in many respects $CeCu_6$ is analogous to $CeAl_3$, both at ambient and high pressures. However, because the resistance of $CeCu_6$ is reversible with pressure, we can identify trends which suggest that pressure enhances the $4f$ hybridization with the conduction band. We also have found that the normalized resistance of $CeCu_6$ scales over an appreciable temperature interval as a function of reduced temperature. To our knowledge, this is the first example of resistance scaling in such a system. That the scaling persists with pressure for $T \ll T_{max}$ may be attributed to the fact that at low temperatures both the Kondo-lattice and mixed-valence states may be described

as Fermi liquids and that one evolves continuously into the other as the $4f$ level approaches the Fermi energy.[8] The pressure dependence of T_{max}, then, reflects this evolution. However, an important aspect of this work, namely, that scaling continues to hold even for $T \gtrsim T_{max}$, remains theoretically unexplained.

Note added. Recently, our attention was brought to the work of Onuki *et al.*[17] and Fujita *et al.*[18] who also have discovered $CeCu_6$ to be a Kondo-lattice system. From high-temperature specific-heat measurements, Fujita *et al.* have inferred the existence of a crystal-field level in $CeCu_6$ separated from the ground state by 65 K, while their low-temperature data suggest a Kondo temperature of 3.9 K.

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[1]See, for example, G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).

[2]G. R. Stewart, Z. Fisk, and M. Wire, Phys. Rev. B **30**, 482 (1984).

[3]K. Andres, J. E. Graebner, and H. R. Ott, Phys. Rev. Lett. **35**, 1779 (1975).

[4]See, for example, J. M. Lawrence, P. S. Riseborough, and R. D. Parks, Rep. Prog. Phys. **44**, 1 (1981).

[5]A. Percheron, J. C. Achard, O. Gorochov, B. Cornut, D. Jérome, and B. Coqblin, Solid State Commun. **12**, 1289 (1973).

[6]J. D. Thompson, Rev. Sci. Instrum. **55**, 231 (1984).

[7]Thermal variations in the resistance of $CeCu_6$ shown in Fig. 1 differ from that published in Ref. 2. This difference disappears after the sample studied in Ref. 2 is given a proper anneal.

[8]R. M. Martin, Phys. Rev. Lett. **48**, 362 (1982); C. D. Bredl, S. Horn, F. Steglich, B. Luthi, and R. M. Martin, *ibid.* **52**, 1982 (1984); N. Grewe, Solid State Commun. **50**, 19 (1984).

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

[10]Because of the close similarity in physical properties between $CeCu_6$ and $CeAl_3$, we have chosen to use the measured bulk modulus of $CeAl_3$ as characteristic of $CeCu_6$. For the bulk modulus of $CeAl_3$, see M. Niksch, B. Luthi, and K. Andres, Phys. Rev. B **22**, 5774 (1980).

[11]D. Wohlleben and J. Rohler, J. Appl. Phys. **55**, 1904 (1984).

[12]B. Cornut and B. Coqblin, Phys. Rev. B **5**, 4541 (1972).

[13]A. B. Kaiser and S. Doniach, Int. J. Magn. **1**, 11 (1970).

[14]D. L. Mills, J. Phys. Chem. Solids **34**, 679 (1973).

[15]R. Jullien, M. T. Béal-Monod, and B. Coqblin, Phys. Rev. B **9**, 1441 (1974).

[16]R. D. Parks and J. D. Thompson (unpublished).

[17]Y. Onuki, Y. Shimazu, and T. Komatsubara, J. Phys. Soc. Jpn. **53**, 1210 (1984).

[18]T. Fujita, K. Satoh, Y. Onuki, and T. Komatsubara, in Proceedings of the International Conference on Valence Fluctuations, Cologne, Federal Republic of Germany, 1984 [J. Magn. Mater. (to be published)].