Non-Fermi Liquid States in the Pressurized $CeCu_2(Si_{1-x}Ge_x)_2$ System: Two Critical Points

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In the archetypal strongly correlated electron superconductor $CeCu_2Si_2$ and its Ge-substituted alloys $CeCu_2(Si_{1-x}Ge_x)_2$ two quantum phase transitions—one magnetic and one of so far unknown origin—can be crossed as a function of pressure. We examine the associated anomalous normal state by detailed measurements of the low temperature resistivity (ρ) power-law exponent α. At the lower critical point (at p_{c1}, $1 \leq \alpha \leq 1.5$) α depends strongly on Ge concentration x and thereby on disorder level, consistent with a Hlubina-Rice-Rosch scenario of critical scattering off antiferromagnetic fluctuations. By contrast, α is independent of x at the upper quantum phase transition (at p_{c2}, $\alpha \simeq 1$), suggesting critical scattering from local or $q = 0$ modes, in agreement with a density- or valence-fluctuation approach.

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Amongst the cerium based f-electron compounds, the superconductor $CeCu_2Si_2$ [1,2] takes a special place. The difficulty in growing high quality samples with reproducible properties and the diversity of observed low temperature states have long complicated and delayed a theoretical description of this intriguing material. After more than 25 years of intensive study, its key properties are gradually being understood. Initial confusion about the ground state properties of $CeCu_2Si_2$ samples—some magnetic, some superconducting—can now be attributed unambiguously to the delicate positioning of this material close to a magnetic quantum critical point (QCP) [3]. The precise nature of the incipient magnetism in ambient-pressure $CeCu_2Si_2$ has recently been determined as incommensurate spin density wave order [4]. Superconductivity in low-pressure $CeCu_2Si_2$ now appears amenable to an analysis along the same lines as in other Ce-based heavy-fermion (HF) compounds on the threshold of magnetism [5], in terms of magnetically mediated pairing. The evolution of $CeCu_2Si_2$ under high pressure, however, has opened up new questions.

The pressure dependence of the superconducting transition temperature T_c in $CeCu_2Si_2$ [6,7] and in its isoelectronic sister compound $CeCu_2Ge_2$ [8] is very different from that observed in other Ce-based HF compounds, such as $CePd_2Si_2$ and $CeIn_3$. In $CeCu_2Si_2$, T_c is nearly pressure independent up to about 2 GPa away from the antiferromagnetic (AFM) QCP (at p_{c1}) and then increases to a maximum value about 3–4 times that at p_{c1}.

To understand the origin of this phase diagram, we have recently performed a study on a series of partially Ge-substituted single crystals $CeCu_2(Si_{1-x}Ge_x)_2$. Because of the weakening of superconductivity by the increased impurity scattering associated with Ge substitution [9] (which widens the lattice and is counterbalanced by applying hydrostatic pressure), the broad and continuous superconducting range previously observed in the p-T phase diagram of pure $CeCu_2Si_2$ and $CeCu_2Ge_2$ breaks up into two disconnected superconducting domes [2]. The low-pressure superconducting dome occurs around an AFM QCP, suggesting magnetically mediated pairing, while the high-pressure superconducting dome straddles a weak first-order volume collapse (Fig. 1) indicative of a second quantum phase transition (QPT) at high pressure. In this Letter, we elucidate the nature of the two QPTs by studying their anomalous normal-state behavior.

Single crystals of $CeCu_2(Si_{1-x}Ge_x)_2$ have been prepared by a flux growth method in excess Cu. High sensitivity, ac four-point measurements of the electrical resistivity were carried out in Bridgman anvil ($p < 10$ GPa) and piston-cylinder ($p < 3.5$ GPa) devices down to 200 mK in an adiabatic demagnetization cooler and down to 50 mK in an Oxford Instruments dilution refrigerator. The normal-state behavior of our samples has been analyzed by fitting

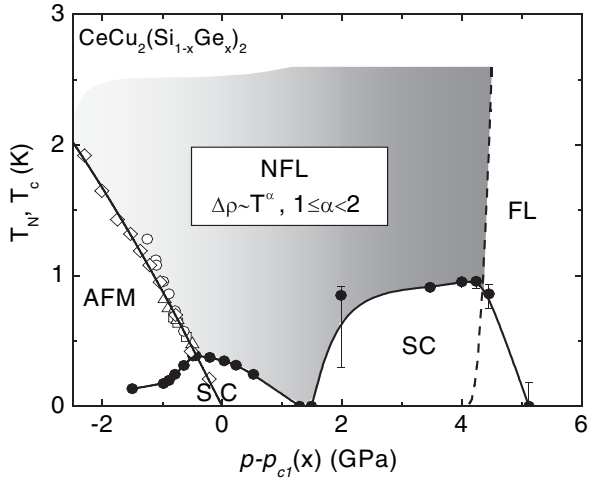

FIG. 1. The combined p-T phase diagram for $CeCu_2(Si_{1-x}Ge_x)_2$ [T_N: $x = 0.25$ (◇), 0.1 (○), 0.05 (△), 0.01 (□); T_c: $x = 0.1$ (●)].

the low temperature normal-state resistivity as $\rho = \rho_0 + AT^\alpha$ up to an adjustable maximum temperature $T_{\max}$. The resulting residual resistivity ρ_0 can be used to extract the temperature dependence of α by taking the logarithmic derivative $\alpha(T) = d\ln[\rho(T) - \rho_0]/d\ln T$, as illustrated in Fig. 2 [10]. Both methods are iterated until convergence in α and $T_{\max}$ is achieved. We note that $T_{\max}$—which represents the range of validity of the asymptotic low-T power-law behavior—depends on Ge concentration and on external pressure. It increases from about 2 K at low p to 10 K at p_{c2} (indicated by darkness of shading in Fig. 1).

Figure 1 summarizes our present knowledge of the ordered phases of the $CeCu_2(Si_{1-x}Ge_x)_2$ system. It has been constructed by shifting the pressure scale for each Ge concentration by the respective lower critical pressure, p_{c1}, at which the AFM transition temperature T_N extrapolates to zero. The critical pressure p_{c1} is about 1.4, 1.5, 1.5, and 2.4 GPa for $x = 0.01$, 0.05, 0.1, and 0.25, respectively. Because of Cu/Si site exchange and possible sample inhomogeneities, the value of p_{c1} becomes less regular for small x. Following such a pressure shift, the abscissa can be approximately regarded as a volume scale [2]. This observation is consistent with the existence of an AFM QCP in $CeCu_2(Si_{1-x}Ge_2)_2$, and indicates that the magnetic QCP exists at a unique volume of the unit cell. At very high pressures, as the system is tuned out of the HF state and into an intermediate valence state, it undergoes an isostructural first-order volume collapse, possibly analogous to the γ-α transition in elemental Ce. The likely pressure dependence of this transition is schematically indicated by a dashed line in Fig. 1. Indeed, a weak first-order volume-collapse line with an apparently low-lying critical end point has been observed around a second QPT in $CeCu_2Ge_2$, where T_c reaches a maximum value [11].

The pressure dependence of the Néel temperature T_N and the volume-collapse transition divide the phase diagram into three regions: the antiferromagnetically ordered state below p_{c1}, the intermediate valence range above p_{c2}, and the more complex region in between the two QPTs.

Focusing initially on the normal state around the low-pressure AFM QCP, we note that different low temperature states can be obtained in ambient pressure $CeCu_2Si_2$ by deliberately choosing the composition of the melt to be slightly off stoichiometry or by suitable heat treatments [12,13]. On the other hand, very similar ground states can be achieved in slightly Ge-substituted samples by applying hydrostatic pressure. This allows us to study the magnetic QCP in greater detail. As an example, Fig. 3 shows three possible cases: (a) *Magnetic* ($T_N > T_c$). At $p = 0.34$ GPa, $CeCu_2(Si_{0.9}Ge_{0.1})_2$ experiences a magnetic reorientation transition at $T_1 \simeq 1$ K (the initial AFM transition is at $T_N \simeq 1.4$ K), followed by a superconducting transition at $T_c \simeq 0.2$ K. Upon applying a magnetic field, superconductivity is quickly suppressed, but the magnetism is much more robust [inset of Fig. 3(a)]. The exponent α decreases with increasing temperature and above T_N, α remains <2 [inset of Fig. 3(a)]. The non-T^2 form of $\rho(T)$ above T_N agrees with thermodynamic properties [3], pointing at a non-Fermi-liquid (NFL) normal state above T_N. (b) *Superconducting/magnetic* ($T_N \leq T_c$). In this case, the

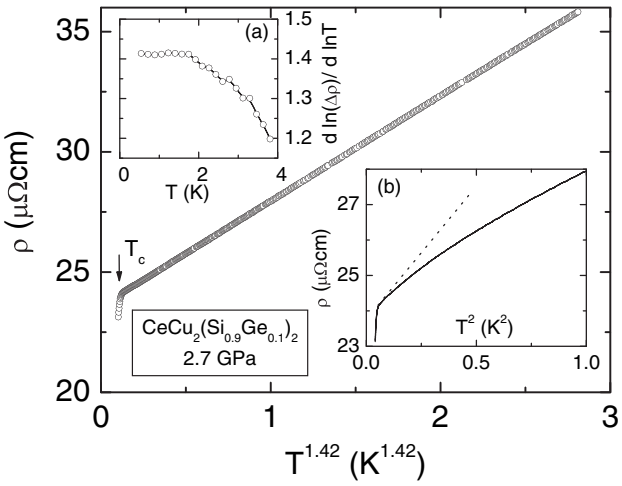

FIG. 2. The fit of the resistivity by $\rho = \rho_0 + AT^\alpha$ for $CeCu_2(Si_{0.9}Ge_{0.1})_2$ at $p = 2.7$ GPa.

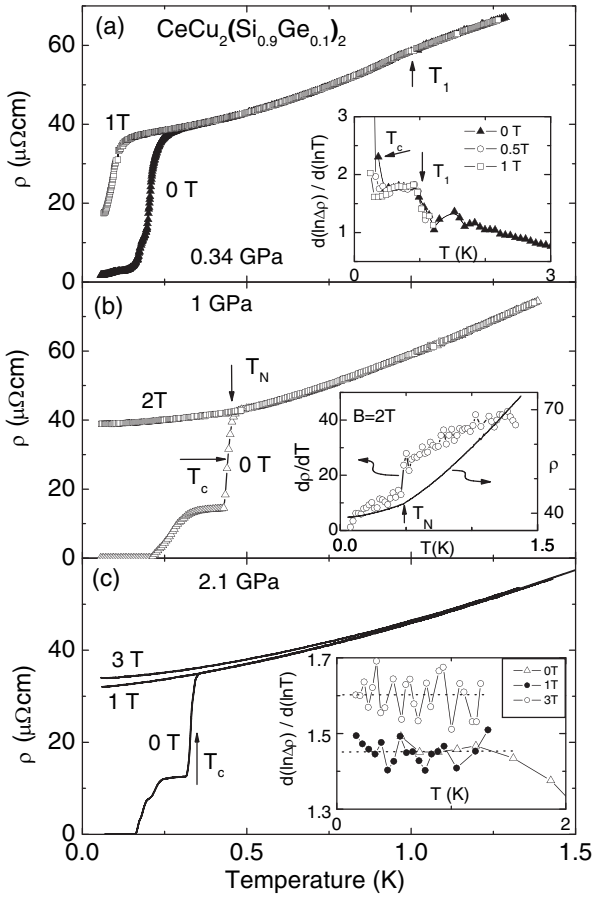

FIG. 3. The electrical resistivity $\rho(T)$ for $CeCu_2(Si_{0.9}Ge_{0.1})_2$ at various magnetic fields and hydrostatic pressures, demonstrating three types of ground states (see text).

magnetic transition is masked by superconductivity, but reappears as superconductivity is suppressed below T_N by a magnetic field. [inset of Fig. 3(b)]. (c) *Superconducting*. No magnetic transition can be observed even when superconductivity is suppressed by a magnetic field. Generally, the magnetic field has little effect on the normal state as long as the field is below the upper critical field B_{c2} [insets of Figs. 3(a) and 3(b)]. When the magnetic field exceeds B_{c2}, the exponent α gradually increases with increasing magnetic field [Fig. 3(c)].

At comparatively low pressures $p \simeq p_{c1}$, the interplay between superconductivity and magnetism in the $CeCu_2(Si/Ge)_2$ system exhibits a similar structure to what is seen in other quantum critical Ce-based HF superconductors, such as $CePd_2Si_2$. However, the question arises how the normal state develops with increasing distance from the AFM QCP, and how it connects up with the volume-collapse QPT at high pressure.

Examining the evolution of the resistivity exponent α across the p-T phase diagram [Fig. 4(a)], we note the following key points: (i) At the AFM QCP (at p_{c1}), the exponent α reaches a local minimum. The value of α at p_{c1} ranges between 1 and 1.5 and increases with increasing Ge content x. (ii) The exponent α reaches a second minimum in the high-pressure superconducting regime, approaching $\alpha \approx 1$ around the volume-collapse transition at p_{c2} ($\Delta p \sim 4$ GPa). Maximum T_c is accompanied in $CeCu_2Si_2$ and its Ge-substituted alloys by an extended T-linear form of the resistivity—*independent of Ge content (and of the associated disorder)*. Upon further increasing pressure above p_{c2}, Fermi-liquid behavior ($\alpha = 2$) is rapidly recovered. (iii) In between the two QPTs, for $p_{c1} < p < p_{c2}$, NFL behavior with $1 \leq \alpha < 2$ survives over a broad range in pressure (about 4 GPa). For small Ge concentrations (e.g., $x = 0$, 0.01 and 0.05), α is nearly pressure independent above p_{c1}. However, α goes through a local maximum at intermediate pressure for larger x ($x = 0.1$ and 0.25).

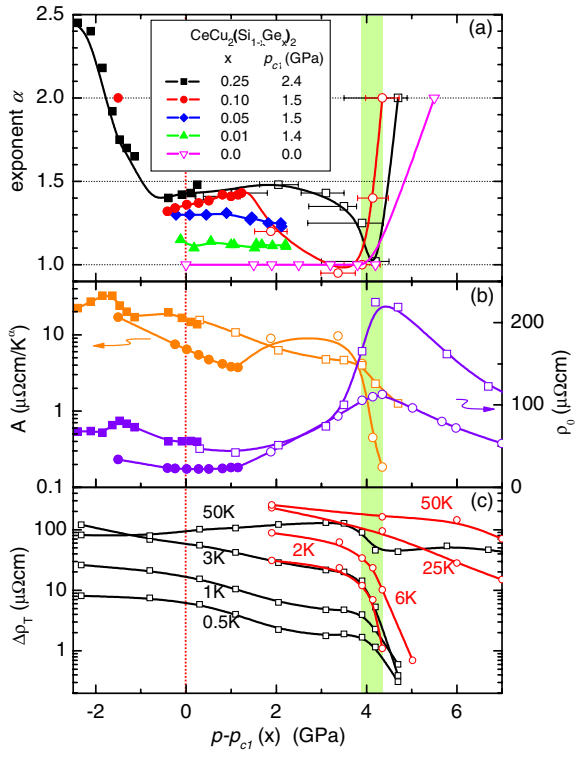

FIG. 4 (color online). The pressure dependence of (a) the resistivity exponent α ($x = 0$ is from Ref. [7]); (b) the resistivity A coefficient and the residual resistivity ρ_0; and (c) the resistivity isotherms $\Delta\rho_T(p)$ [$= \rho(p, T) - \rho_0(p)$] at various temperatures for $CeCu_2(Si_{1-x}Ge_x)_2$ [$x = 0.25$ (□), 0.1 (○), 0.05 (◇), 0.01 (△), and 0.0 (▽)]. The solid symbols represent the samples measured in clamped pressure cells and the empty ones are from Bridgman anvil cells.

As in other quantum critical HF compounds, current theories can only account qualitatively for the anomalous normal state observed in $CeCu_2(Si_{1-x}Ge_x)_2$. At the AFM QCP, spin-fluctuation theories [14–17] predict $\alpha = 1.5$ and $\alpha = 1$ for 3D and 2D spin fluctuations, respectively, while our measured exponents are sample dependent and lie between these two extremes. The observed increase of α with increasing disorder ($1 \leq \alpha \leq 1.5$) may, however, be explained within a generalized Hlubina-Rice type hot-spot/cold-spot scenario, e.g. [18]. Such an approach takes into account both the short circuiting of critical scattering at large wave vector $\mathbf{q} \sim \mathbf{Q}$ (connecting "hot" regions of the Fermi surface) by "cold" regions, and the influence of impurity scattering, which is present at all $\mathbf{q}$.

The presence of a second QPT at p_{c2} holds the key for understanding the unusual pressure dependence of the resistivity exponent in between p_{c1} and p_{c2}. In Figs. 4(b) and 4(c), the pressure dependence of the A coefficient in $\Delta\rho = AT^{\alpha}$ and the resistivity isotherms $\Delta\rho_T(p)$ [$=\rho(p, T) - \rho_0(p)$] at various temperatures are shown for the samples with $x = 0.1$ and $x = 0.25$. The collapse of $\Delta\rho_T(p)$ (at $T < 10$ K) and of $A(p)$ on crossing the upper critical pressure $\Delta p = p_{c2} - p_{c1}(x) \simeq 4$ GPa, indicates a transition from the HF state to an intermediate valence state at p_{c2}. This valence transition may be accompanied by an isostructural, weak first-order volume collapse, as suggested by x-ray diffraction experiments on $CeCu_2Ge_2$ [11]. At temperatures exceeding 10 K, the drop in the resistivity isotherms at p_{c2} weakens [Fig. 4(c)], and it vanishes below 50 K. These data suggest that the first-order transition line associated with the putative density or valence change at p_{c2} reaches its critical end point at a very low temperature, less than 50 K, explaining also why various past attempts to observe the volume collapse in $CeCu_2(Si/Ge)_2$ by high-pressure x-ray diffraction at room temperature have remained unsuccessful.

A weak volume-collapse transition at p_{c2} is expected to be accompanied by large amplitude fluctuations of the lattice density and, consequently, of the local charge distribution (i.e., the valence). Charge carrier scattering is

modified in the presence of these fluctuations, giving rise to an anomalous temperature dependence of $\rho(T)$, provided that the fluctuation relaxation rate reaches down to low enough energies. In the most detailed scenario so far, proposed by Miyake [19], nondispersive (local), but nearly critical valence fluctuations are invoked to explain the linear T dependence of $\rho(T)$ at p_{c2}, essentially as a consequence of the equipartition theorem. It is as yet unclear whether this approach can also explain the absence of a giant heat capacity or A-coefficient peak, which would be expected in the presence of very low-lying excitations spread over large portions of the Brillouin zone, as well as the occurrence of superconductivity, which usually requires a nonlocal pair-forming interaction. Density or valence fluctuations peaked at $\mathbf{q} = 0$, whether dispersive or nearly local, would however offer an explanation for the observed disorder-level independent power-law exponent at p_{c2}, because in this case the entire Fermi surface can be considered hot. In contrast to the AFM QCP at p_{c1}, where a hot-spot/cold-spot scenario accounted at least qualitatively for the impurity-level dependence of α, the T-linear resistivity obtained from a density or valence-fluctuation model should then be robust against the level of disorder—in agreement with our experimental findings.

Second to superconductivity, arguably the most dramatic phenomenon in the $CeCu_2(Si/Ge)_2$ system is the enormous enhancement of the residual resistivity ρ_0 around p_{c2} [Fig. 4(b)], which contrasts starkly with the weak *minimum* in ρ_0 at p_{c1}. The origin of this distinct peak in $\rho_0(p)$ has been proposed to lie in a strongly pressure dependent impurity scattering cross section, as p_{c2} is approached. Here, the problem lies in the computed logarithmic dependence of ρ_0 on the distance from the critical point [20], coupled with the first-order nature of the volume-collapse transition at low T. An alternative approach to the state of $CeCu_2(Si/Ge)_2$ near p_{c2} may consider the likely phase separation into low-density (HF) and high-density (intermediate-valent) domains, populated by heavy and light carriers, respectively, in distant analogy with the mechanism underlying colossal magnetoresistance. On the assumption that light quasiparticles cannot propagate in heavy-fermion domains and conversely, heavy quasiparticles scatter strongly in the intermediate-valent (high-density) domains, $CeCu_2Si_2$ is expected to turn opaque to electrical transport over a narrow region surrounding p_{c2}, leading to the observed pronounced maximum in $\rho_0(p)$.

In contrast to stoichiometric $CeCu_2Si_2$, in which a quasilinear T dependence of the resistivity extends over the entire region between p_{c1} and p_{c2}, the resistivity exponent α in Ge-substituted $CeCu_2Si_2$ single crystals reaches two distinct minima at p_{c1} and p_{c2}. These results indicate that the apparent critical *region* in the p-T phase diagram of stoichiometric $CeCu_2Si_2$ is a result of two critical *points*, each surrounded by a pressure range in which α is low. We arrive then at a picture analogous to the explanation for the wide superconducting range in stoichiometric $CeCu_2Si_2$, which is attributed to the merger of the two superconducting domes in Ge-substituted $CeCu_2(Si/Ge)_2$: the interplay of two QPTs results in the unusual pressure dependence of both superconductivity and normal-state behavior in $CeCu_2Si_2$. While the AFM critical point at p_{c1} is similar in nature to that in other Ce-based HF compounds, the precise nature and origin of the QPT at p_{c2} is still unclear. Some of its consequences—the colossal pressure dependence of ρ_0 and the linear, disorder-level independent T dependence of ρ—are, however, clearly established and invite further theoretical investigation.

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[1] F. Steglich *et al.*, Phys. Rev. Lett. **43**, 1892 (1979).
[2] H. Q. Yuan *et al.*, Acta Phys. Pol. B **34**, 533 (2003); H. Q. Yuan *et al.*, Science **302**, 2104 (2003).
[3] P. Gegenwart *et al.*, Phys. Rev. Lett. **81**, 1501 (1998).
[4] O. Stockert *et al.*, Phys. Rev. Lett. **92**, 136401 (2004).
[5] N. D. Mathur *et al.*, Nature (London) **394**, 39 (1998).
[6] F. Thomas *et al.*, Physica B (Amsterdam) **186–188**, 303 (1993).
[7] B. Bellarbi *et al.*, Phys. Rev. B **30**, 1182 (1984).
[8] D. Jaccard *et al.*, Physica B (Amsterdam) **259–261**, 1 (1999).
[9] H. Q. Yuan *et al.*, New J. Phys. **6**, 132 (2004).
[10] H. Q. Yuan, Ph.D. thesis, Technische Universität Dresden, 2003.
[11] A. Onodera *et al.*, Solid State Commun. **123**, 113 (2002).
[12] F. Steglich *et al.*, Physica B (Amsterdam) **223–224**, 1 (1996).
[13] M. Lang *et al.*, in *Electron Correlations and Materials Properties*, edited by A. Gonis and N. Kioussis (Kluwer Academic/Plenum, New York, 1999), p. 153.
[14] J. A. Hertz, Phys. Rev. B **14**, 1165 (1976).
[15] A. J. Millis, Phys. Rev. B **48**, 7183 (1993).
[16] T. Moriya *et al.*, J. Phys. Soc. Jpn. **64**, 960 (1995).
[17] G. G. Lonzarich, in *Electron*, edited by M. Springford (Cambridge University Press, Cambridge, England, 1997).
[18] A. Rosch, Phys. Rev. Lett. **82**, 4280 (1999).
[19] A. T. Holmes *et al.*, Phys. Rev. B **69**, 024508 (2004).
[20] K. Miyake *et al.*, J. Phys. Soc. Jpn. **71**, 1007 (2002).