

Pressure versus magnetic-field tuning of a magnetic quantum phase transition

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Specific heat $C(T)$ and electrical resistivity $\rho(T)$ of $\text{CeCu}_{6-x}\text{Au}_x$ at very low temperatures T show distinctly different behavior depending on whether long-range antiferromagnetic order is suppressed by hydrostatic pressure p or an applied magnetic field B . p tuning yields $C/T = a \ln(T_0/T)$ and $\rho \approx \rho_0 + A'T$, while B tuning shows $C/T = \gamma_0 - a'T^{0.5}$ and $\rho \approx \rho_0 + A''T^{1.5}$. This suggests that the spectrum of low-lying excitations that determines the behavior near these quantum phase transitions differs.

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I. INTRODUCTION

Metals can acquire distinctly different ground states, such as magnetically ordered, superconducting, or simply paramagnetic with an essentially temperature-independent Pauli spin susceptibility. The transition between magnetically ordered and paramagnetic metallic ground states at temperature $T=0$ may be continuous or discontinuous. In the former case, quantum fluctuations play an important role in determining the properties of the system around the transition. In weak itinerant magnets such as MnSi, the $T=0$ quantum phase transition may be tuned by pressure¹ and the low- T properties may be described within a quantum Ginzburg-Landau (QGL) model² of conduction electrons coupled to Gaussian spin fluctuations.³

Another class of materials where a quantum phase transition (QPT) close to a magnetic instability can be accessed easily, is provided by heavy-fermion systems (HFS) that are intermetallic compounds with nearly localized $4f$ or $5f$ electrons. In these systems, massive quasiparticles form below a characteristic temperature, due to a resonance between conduction electrons and f electrons building up at the Fermi level E_F , and the Pauli paramagnetic state is achieved through screening of the $4f$ or $5f$ magnetic moments by the conduction electrons. This scenario is well understood in dilute magnetic alloys (Kondo effect) and believed to hold also for many HFS. Yet, in HFS, short-range dynamic correlations indicate the proximity to magnetic order. A prototype of such a “nonmagnetic” HFS is CeCu_6 , where a transition to an antiferromagnetic ground state can be induced by alloying with Au. Long-range incommensurate order is found in $\text{CeCu}_{6-x}\text{Au}_x$ for $x \geq 0.15$, where the Néel temperature T_N increases linearly with x up to $x=1$.⁴ At a critical Au concentration $x = x_c \approx 0.1$, where $T_N \rightarrow 0$ in $\text{CeCu}_{6-x}\text{Au}_x$, non-Fermi liquid (NFL) anomalies are observed in the thermodynamic and transport properties,⁵ i.e., the specific heat C varies as $C/T = a \ln(T_0/T)$, and the electrical resistivity depends quasilinearly on T , $\rho \approx \rho_0 + A'T$, in stark contrast to the standard Fermi-liquid model that predicts $C/T = \text{const}$ and $\rho = \rho_0 + AT^2$, as approximately observed for pure CeCu_6 at sufficiently low T .^{6,7} The QPT can also be tuned in magnetically ordered $\text{CeCu}_{6-x}\text{Au}_x$ with $x > x_c$ when driving the Néel temperature T_N to zero by applying hydrostatic pressure. This results in the same T dependence of C/T at the

critical pressure p_c as for x_c at ambient pressure.⁸ Recently, experiments on the related system $\text{CeCu}_{6-x}\text{Ag}_x$ were reported where the antiferromagnetic order was suppressed by a magnetic field applied along the easy direction.⁹ In this case, C/T levels off towards a finite value at low T and ρ shows a $T^{1.5}$ dependence, suggestive of a different type of NFL behavior.

In order to elucidate the difference of pressure and magnetic field in tuning a QPT in f -electron systems, we have performed measurements of C and ρ using both pressure tuning and field tuning on the *same* antiferromagnetic $\text{CeCu}_{5.8}\text{Au}_{0.2}$ crystals ($T_N \approx 0.25$ K for $p=0$). Our results demonstrate a striking difference between both sets of experiments, implying that the low-lying excitations at the respective quantum critical points are different.

II. RESULTS AND DISCUSSION

A. Experiment

The experiments were carried out on different specimens cut from the same single crystal of $\text{CeCu}_{5.8}\text{Au}_{0.2}$ grown by the Czochralski method as described previously.¹⁰ Microprobe analysis and x-ray diffraction did not reveal any concentration fluctuations along the crystal. Moreover, the electrical resistivity $\rho(T)$ for several specimens from different locations of the crystal was found to be identical. The specific heat C under pressure was determined as reported earlier.⁸ For the electrical resistivity measured with a standard low-frequency four-point technique, a Cu-Be pressure cell similar to that for the C measurements was employed.

B. Neutron scattering

The long-range magnetic order as investigated by neutron diffraction (Institut Laue-Langevin, Grenoble, instrument IN 14) is characterized by sharp, resolution-limited incommensurate reflections at 50 mK, i.e., well below T_N (see inset of Fig. 1). These correspond to a magnetic ordering vector $\mathbf{Q} = (0.625 \ 0 \ 0.275)$ as reported earlier.⁴ The main frame of Fig. 1 shows that magnetic order is suppressed by a magnetic field $B_c \sim 0.42$ T as estimated from the linear extrapolation to zero of the integrated neutron-scattering intensity I of the $(2.625 \ 0 \ 0.275)$ reflection for $T=50$ mK and 180 mK. Here and in the following experiments, B was always applied parallel to the c direction, which is the easy direction in

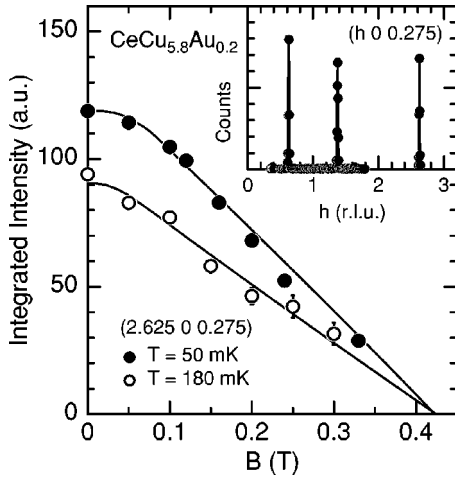


FIG. 1. Integrated elastic neutron-scattering intensity of the $(2.625\ 0\ 0.275)$ magnetic reflection for $\text{CeCu}_{5.2}\text{Au}_{0.8}$ at two different temperatures. Lines are guides to the eye. Inset shows an h scan with three resolution-limited magnetic reflections.

$\text{CeCu}_{6-x}\text{Au}_x$.¹⁰ Since the magnetic anisotropy is rather large, with $\chi_c:\chi_a:\chi_b=10:5:1$, this corresponds to a *longitudinal* field. The data are compatible with a linear $I(B)$ dependence near the critical field B_c that would correspond to $M_s \sim (B - B_c)^{1/2}$, where the exponent is different from $1/3$ expected for mean-field behavior. However, the limited statistics precludes a definite statement. Within the large scatter of the data, B_c is the same for 50 and 180 mK.

C. Specific heat

Figure 2 shows the specific heat C plotted as C/T vs T on a logarithmic scale for various hydrostatic pressures p . The magnetic transition at $p=0$ is seen as a clear kink in C/T , corresponding to a maximum in C vs T . T_N shifts to lower T , as observed previously for $x=0.5$ (Ref. 11) and 0.3 (Ref. 8). For $p=4.1$ kbar, $C/T = a \ln(T_0/T)$ is observed over nearly two decades in T signaling NFL behavior with—within the error bars—exactly the same behavior as for $x=x_c=0.1$ at ambient pressure. The slight positive deviations for $p=3.2$ kbar at low T may indicate the onset of magnetic order just below the experimentally accessible T range. At 6.9 kbar, the $x=0.2$ sample approaches a Fermi-liquid-like T dependence, i.e., $C/T = \text{const}$, as may be seen by comparing this data to C/T for pure CeCu_6 at $p=0$.

The $C/T \sim \ln(T_0/T)$ dependence at the QPT can be interpreted invoking a quasi-two-dimensional (2D) fluctuation spectrum.¹² This scenario accounts even semiquantitatively for the prefactor a of the logarithmic T dependence of C/T . The properties of $C(T)$ imply that the 2D fluctuation spectrum does not change qualitatively upon application of pressure, i.e., the logarithmic T dependence prevails. Furthermore, the coefficient a remains unchanged, hence a possible quantitative change of the spectrum appears to be compensated by a change of the quasiparticle dynamics, cf. a change of the resonance between $4f$ electrons and conduction electrons (Kondo resonance), by pressure. In fact, the identity of C/T approaching the quantum critical points (x, p)

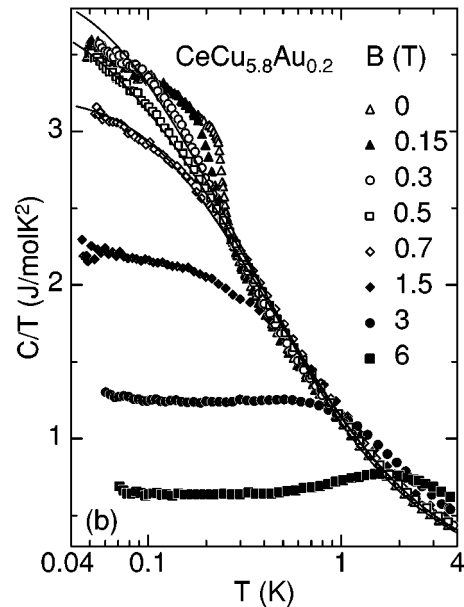
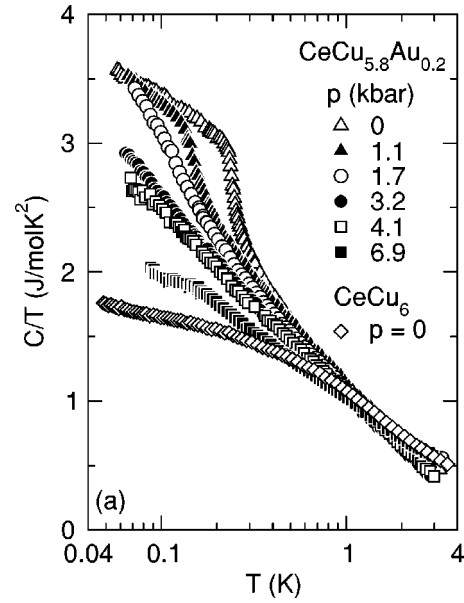


FIG. 2. (a) Specific heat C of $\text{CeCu}_{5.8}\text{Au}_{0.2}$ for different hydrostatic pressures p , plotted as C/T vs T on a logarithmic scale. Also shown are the data for CeCu_6 at ambient pressure. (b) C/T vs T on a logarithmic scale of $\text{CeCu}_{5.8}\text{Au}_{0.2}$ for different applied magnetic fields B . Solid lines indicate fits of the Moriya-Takimoto model of spin fluctuations (Ref. 13) to the data for $B=0.3, 0.5$, and 0.7 T. See text for details.

$= (0.1, 0)$, $(0.2, 4.1$ kbar), and $(0.3, 8.2$ kbar), where the $x=0.3$ data are from Ref. 8 is striking.

Figure 2(b) shows the specific heat of the $x=0.2$ sample for various applied magnetic fields B . Again, T_N is suppressed with increasing B , but the rapidly increasing rounding of C/T vs T at the transition prevents a determination of the critical field B_c where the antiferromagnetic order vanishes. However, recall that the neutron data shown in Fig. 1 clearly indicate $B_c=0.42$ T. For fields just below and above B_c , i.e., $B=0.3$ and 0.5 T, we observe a negative curvature

in C/T vs T on a logarithmic scale towards low T , distinctly different from the T dependence observed in pressure tuning the QPT. Here we have subtracted the hyperfine contribution $C_{\text{hf}} = b_N T^{-2}$ due to the Zeeman splitting of ^{63}Cu and ^{65}Cu nuclei in an effective field B_{eff} that was determined at $B = 6$ T to $B_{\text{eff}} = 1.08 B$ and scaled accordingly for lower fields. It is interesting to note that the specific-heat data at $B = 0.5$ and 0.7 T may be modeled quite accurately by the expression of the QGL model in the form given by Moriya and Takimoto¹³ as will be shown below. Here, Kondo screening is viewed to give rise to onsite (local) spin fluctuations of constant amplitude that are correlated in the spirit of the 3D intersite (nonlocal) Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Thus, the Doniach picture of the competition of the Kondo effect and RKKY interaction¹⁴ may be viewed as competition of frequency-dependent local fluctuations and q -dependent intersite fluctuations. For comparison with experiment, we introduce the effect of magnetic field as a variation of the inverse staggered susceptibility as described by the parameter y_0 . We thus assume that the isotropic model given in Ref. 13 is appropriate at comparatively small fields, ignoring the detailed field dependence of longitudinal (amplitude) and transverse fluctuations. Furthermore, the model ignores the effects of spin-orbit coupling, which in general, may be quite strong in lanthanide compounds.

By using the full finite- T expression for the specific heat C , Eq. (4.5) of Ref. 12, we obtain a good fit for $B = 0.5$ T with the parameters $y_0 = 0.01$, $y_1 = 8$, and $T_A = 2.8$ K. This expression yields a low- T asymptotic dependence $C/T = \gamma_0 - a' T^{0.5}$. Even the data for $B = 0.7$ T, may be fitted very well by $y_0 = 0.032$, an unchanged y_1 , and a slightly changed $T_A = 2.9$ K. On the other hand, the best possible fit for $B = 0.3$ T ($y_0 = 0.02$, $y_1 = 10$, and $T_A = 2.9$ K) is clearly less satisfactory. The fits are indicated in Fig. 2(b) by solid lines. It is remarkable that the agreement reaches as high as 4 K, although the range of validity, in principle, is constrained to temperatures well below the Kondo temperature. A similar analysis, though purely qualitative, has previously been performed for $\text{CeCu}_{5.2}\text{Ag}_{0.8}$, where the limiting behavior $C/T = \gamma_0 - a' T^{0.5}$ was reported to be in good agreement with experiment.⁹ Indeed, our data for $B = 0.5$ T also follow this low- T behavior for $T < 0.2$ K as expected, see Fig. 3. Again, the agreement for $B = 0.3$ T is clearly less satisfactory. However, only a model going beyond the various approximations employed here, addressing the field dependence over a large range, is expected to show if the behavior near B_c may indeed be interpreted as a field-induced *quantum* phase transition.

D. Electrical resistivity

Figure 4(a) shows the electrical resistivity $\rho(T)$ for several pressures p . Here, ρ was measured with the electrical current along the a direction. The increase below 0.22 K at ambient pressure signals the onset of antiferromagnetic order. A detailed discussion of the interplay of magnetic order and transport leading to the rise of $\rho(T)$ below T_N for current directions with a nonvanishing \mathbf{Q} component is given elsewhere.⁴ The decrease of T_N with increasing p is directly

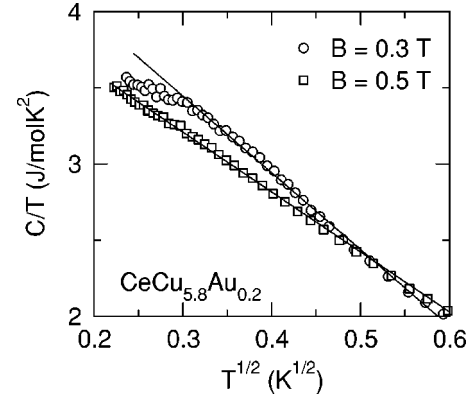


FIG. 3. Specific heat of $\text{CeCu}_{5.8}\text{Au}_{0.2}$ plotted as C/T vs \sqrt{T} for $B = 0.3$ and 0.5 T in the immediate vicinity of the field tuned QPT. Solid lines serve as guide to the eye.

visible in $\rho(T)$, with T_N vanishing around $p_c \approx 5$ kbar, in reasonable agreement with the specific-heat results. There are three further features that deserve attention: First, there is a distinct change of curvature of $\rho(T)$ above T_N as p increases. This may be attributed to a strong increase of the width of the Kondo resonance with pressure and of the concomitant $\rho(T)$ maximum observed in lattice-coherent HFS, as also seen in pure CeCu_6 .¹⁵ The associated change of curvature renders a comparison of $\rho(T)$ over an extended T range for different p difficult. However, we can nonetheless extract a linear T dependence of $\rho(T)$ over a limited T range above p_c . The lower end T_{FL} of that T range marks the onset of Fermi-liquid behavior with a T^2 dependence of ρ . T_{FL} is seen to increase linearly with increasing p (Fig. 5). Although experiments were not performed directly at the critical pressure, a careful comparison of the T dependence at 4.1 with that at 7.0 kbar allows us to draw some unambiguous conclusions for the critical pressure. For both pressures, the well-defined onset of magnetic order at $T_N \approx 30$ mK and Fermi-liquid dependence at $T_{\text{FL}} \approx 30$ mK, respectively, are close to the lower limit of our experimental range of 15 mK. On the other hand, the quasilinear variation of $\rho(T)$ in the paramagnetic regime is not sensitive to pressure over this small interval, and allows comparison with the field dependence at the critical field. Although experiments were not performed directly at p_c , the quasilinear T dependence of $\rho(T)$ for $p = 7$ kbar does resemble that of $\rho(T)$ for $x = 0.1$ at $p = 0$.

Second, the coefficient $A' = 25 \mu\Omega \text{ cm K}^{-1}$ is a factor of two smaller than that for $x = 0.1$ where $A' = 52.9 \mu\Omega \text{ cm}$, while ρ_0 is 25% larger.^{16,17} This is to be contrasted with the $C(T)$ behavior where pressure tuning leads to a quantitative recovery of the coefficient a at the quantum critical point as for $x = 0.1$ and $p = 0$. Although the quasiparticle relaxation rate at the pressure-tuned quantum phase transition is reduced by a factor of two, the qualitative consistency in $\rho(T)$ and the quantitative agreement of $C(T)$ suggest that the overall spectrum remain unchanged.¹⁸ The quasilinear $\rho(T)$ is a distinguished feature of the 2D-fluctuation scenario discussed above in conjunction with the specific heat.¹⁸ Third, the residual resistivity ρ_0 depends strongly on p , i.e., much

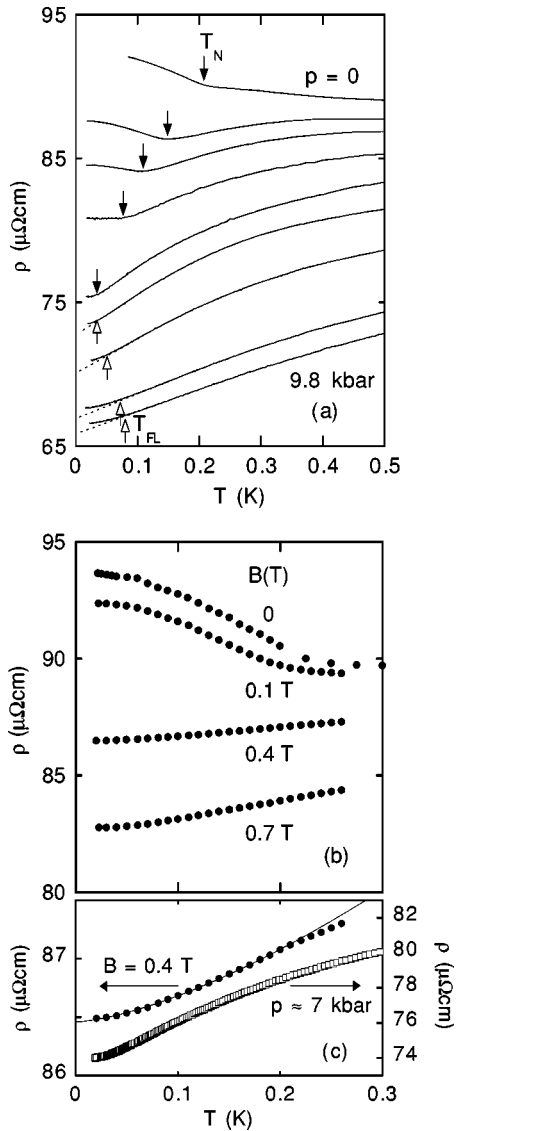


FIG. 4. (a) Electrical resistivity ρ vs temperature T of $\text{CeCu}_{5.8}\text{Au}_{0.2}$ for various hydrostatic pressures $p=0, 1.3, 2.4, 3.5, 4.1, 7.0, 8.1, 9.3,$ and 9.8 kbar (from top to bottom). Solid arrows indicate the Néel temperature T_N , open arrows the crossover temperature T_{FL} below which ρ exhibits a T^2 dependence. (b) ρ vs T of $\text{CeCu}_{5.8}\text{Au}_{0.2}$ for various magnetic fields B . (c) Comparison of the T dependence of ρ near the magnetic-nonmagnetic transition obtained by field tuning ($B=0.4$ T) and pressure tuning ($p=7$ kbar). Although in (c) p is not the critical value, data may be compared as outlined in the text.

more strongly than may be anticipated by the simple suppression of magnetic order. This indicates that the local strains and local variations of the electronic structure as introduced by alloying, which depend strongly on whether a particular Ce atom has a local environment of only Cu atoms or of Cu atoms with exactly one free atom at the Cu(2) site,¹⁹ is reduced by pressure.

Turning to the effect of a magnetic field on $\rho(T)$, we first note that its effect on ρ_0 is rather small compared to that of p [Fig. 4(b)]. Furthermore, the best fit for $\rho(T)=\rho_0+A''T^m$ yields $A''=6.8 \mu\Omega \text{ cm K}^{-m}$ and $m=1.48\pm 0.03$ where m is

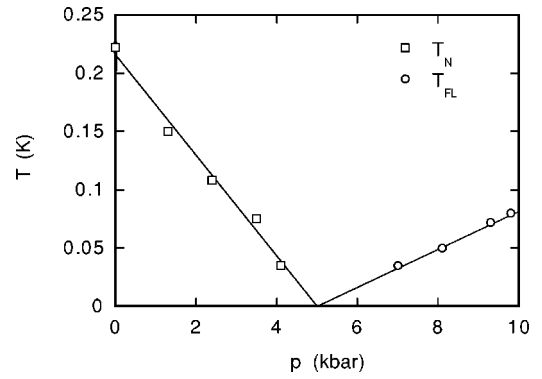


FIG. 5. Pressure dependence of the antiferromagnetic ordering temperature T_N of $\text{CeCu}_{5.8}\text{Au}_{0.2}$, and crossover to Fermi-liquid T^2 dependence, T_{FL} . The lines serve as a guide to the eye.

in very good agreement with the QGL scenario. For $B=0.7$ T, a $T^{1.5}$ fit still is satisfactory, although the data at low T are better described by a T^2 behavior leveling off towards a linear T dependence. This behavior has been observed similarly for $\text{CeCu}_{5.2}\text{Ag}_{0.8}$.²⁰ The main point, however, is the clear distinction of the resistivity $\rho(T)$ for pressure tuning vs field tuning the QPT, i.e., for p_c and B_c , respectively. This point is emphasized in Fig. 4(c) where the different T dependencies of $\rho(T)$ are clearly visible. As noted above, the T dependence in p tuning is clearly linear as seen also in concentration tuning. This difference is in line with that observed in C/T for p and B tuning, as discussed above. However, only a qualitative comparison of $\rho(T)$ with the QGL model¹³ for the $T>0$ limit is appropriate due to the presence of the coherence maximum at several K. The large values of ρ_0 , in comparison to the temperature dependent part $\Delta\rho(T)=\rho(T)-\rho_0$, may additionally indicate a large incoherent background so that the resistivity effectively only probes a tiny part of the low-lying spectrum of excitations by comparison with the heat capacity.

III. CONCLUSIONS

The different behavior of $C(T)$ and $\rho(T)$ at the QPT tuned by B or p presents strong evidence for pronounced differences in the fluctuation spectra. The pressure-tuning results suggest that the strongly anisotropic fluctuation spectrum observed for $x=0.1$ at ambient pressure that can be modeled by quasi-2D fluctuations prevails. A detailed investigation of the energy dependence of the critical fluctuations for $x=0.1$ has revealed an unexpected energy-temperature scaling of the dynamic susceptibility $\chi^{-1}(q,E)=c^{-1}[f(q)+(-iE+aT)^\alpha]$. Such a scaling is not expected for a QPT with Gaussian fluctuations. In addition, the scaling exponent $\alpha\approx 0.75$ is quite unusual, and $f(q)\rightarrow 0$ in quasi-one-dimensional regions of critical fluctuations¹² in the reciprocal ac plane. The critical dynamics exhibiting E/T scaling thereby appears to be independent of q , i.e., it emerges as a local property.²¹ Very recently, this analysis was extended to static magnetization measurements that were shown to exhibit field-temperature scaling with the same anomalous exponent α .²²

Hints why pressure tuning in $\text{CeCu}_{6-x}\text{Au}_x$ is similar to concentration tuning while field tuning is not, may be sought in the underlying microscopic mechanisms. For instance, pressure reduces the volume and thus destabilizes the magnetic moments by increasing the hybridization between $4f$ electrons and conduction electrons, compensating the effect of lattice expansion upon Au doping of pure CeCu_6 . On the other hand, a magnetic field, besides breaking magnetic order, also tends to suppress the Kondo singlet state, thus actually stabilizing the magnetic moments. Pressure and field, moreover, differ in that pressure acts as an isotropic means of tuning the QPT while magnetic field is not isotropic. The present field-tuning results suggest that the excitation spectrum might be more isotropic in a magnetic field as the data can be described within the $d=3$ spin-fluctuation scenario, if indeed the zero-field QGL scenario may be applied at finite fields. It will be interesting to check if the anomalous local scaling, which of course is not compatible with the QGL model, occurs at the longitudinal-field-induced magnetic-nonmagnetic QPT.

A further point of theoretical interest concerns the different behavior of C and ρ at the pressure vs concentration-tuned QPT, suggesting that the relaxation rate of quasiparticles is considerably smaller in the former case, while the overall low-energy excitation spectrum as determined by C remains unchanged. Inelastic neutron-scattering studies under pressure and in a magnetic field, as well as uniaxial stress studies, are highly desirable in order to qualify the findings of the present study.

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