Mechanism for the Non-Fermi-Liquid Behavior in $CeCu_{6-x}Au_x$

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We propose an explanation for the recently observed non-Fermi-liquid behavior of metallic alloys $CeCu_{6-x}Au_x$: Near $x = 0.1$, the specific heat C is proportional to $T\ln(T_0/T)$, and the resistivity increases linearly with temperature T over a wide range of T. These features follow from a model in which three-dimensional conduction electrons are coupled to *two*-dimensional critical *ferromagnetic* fluctuations near the quantum critical point $x_c = 0.1$. This picture is motivated by the neutron scattering data in the ordered phase ($x = 0.2$) and is consistent with the observed phase diagram. [S0031-9007(97)03504-7]

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Fermi-liquid (FL) theory has traditionally led to an accurate description of the low temperature properties of metals. Even in the heavy-fermion compounds, where the bare electron parameters are renormalized by up to 3 orders of magnitude by the interaction, FL behavior is observed at low temperatures T with a specific heat $C \propto T$, a magnetic susceptibility $\chi \approx$ const, and a resistivity $\rho \approx \rho_0 + AT^2$. However, in several heavy-fermion systems [1–7], pronounced deviations from FL behavior have been found in a number of physical properties.

Three main theoretical scenarios have been proposed to explain the occurrence of the non-Fermi-liquid (NFL) behavior: In the first one [4,8,9] it is assumed that disorder introduces a distribution of (one-channel) Kondo temperatures T_K in the system; this distribution can be directly related to an anomalous low temperature behavior. The second model proposes a single-impurity origin of the NFL, e.g., associated with the quadrupolar (two-channel) Kondo effect [1,3].

In $CeCu_{6-x}Au_x$, there is clear experimental evidence [5–7] for a third mechanism based on the proximity to a quantum phase transition (QPT) [10–13] near $x_c = 0.1$. For $x > x_c$ the system orders magnetically with a Néel temperature $T_N \propto (x - x_c)^\mu$ with $\mu = 1$ as shown in Fig. 1. The QPT can be interpreted as the result of the competition between the Kondo effect, which tends to screen the magnetic moments, and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which favors a magnetically ordered state. The Kondo effect is weakened by increasing doping which, as suggested experimentally, mainly leads to a volume increase with no apparent change in the number of carriers.

$CeCu_{6-x}Au_x$ remains metallic for all x with typical FL properties at low temperatures both at $x = 0$ and for $x \gg x_c$. However, at $x = x_c$, T_N vanishes and NFL behavior is observed in all accessible quantities down to the lowest temperatures, $T \ll T_K \approx 6$ K. As an example, we show in Fig. 2 the specific heat with $C/T \propto \ln(T_0/T)$ over nearly two decades. At the same doping, the static susceptibility $\chi \propto 1 - \alpha\sqrt{T}$ and the resistivity $\rho \approx \rho_0 + A'T$ show a remarkable NFL behavior over a substantial T range [5].

Up to now [5,7,14] it was assumed that the critical fluctuations of the QPT are dominated by incommensurate three-dimensional correlations. This would strongly suggest a description by a quantum-critical theory with $d = 3, z = 2$ as investigated in [10,11,14]. However, this well-established theory is in contradiction to the thermodynamic properties which are observed in the experiment, as it would suggest $C/T \propto 1 - B\sqrt{T}$ and $\rho \approx \rho_0 + A''T^{3/2}$ at low temperatures [11,14]. One could, however, argue that in the experiments only a crossover region is accessible [14]. In our opinion this is not fully convincing, and it appears not to be possible to fit both resistivity and specific heat over the observed T range by such a theory [7,15].

Below we will focus on a novel feature of the magnetic fluctuations observed by elastic neutron scattering

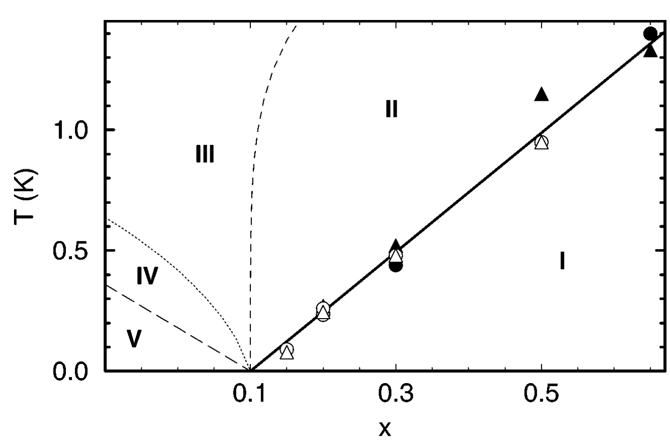

FIG. 1. Phase diagram of $CeCu_{6-x}Au_x$. The points are Néel temperatures [7] (open and closed symbols for single and polycrystals, respectively), the solid line denotes the phase transition, the dashed lines are theoretical crossover lines. The regions are described in the text.

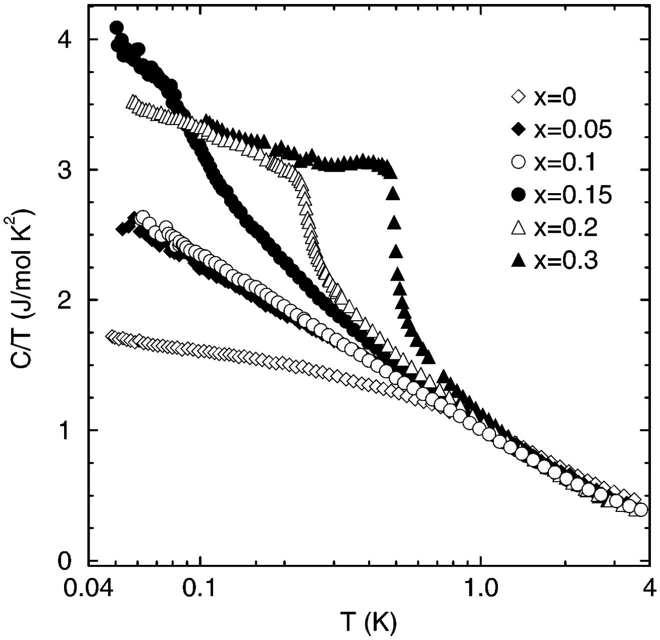

FIG. 2. The specific heat C/T of $CeCu_{6-x}Au_x$ (from [7]) on a logarithmic scale.

experiments near the quantum critical point (QCP), performed at the triple axis spectrometer TAS7 at Risø. The data taken in the ordered phase ($x = 0.2$) are displayed in Fig. 3. A scan along $(h,0,0)$ reveals besides the nuclear reflections two magnetic features. The satellite peaks at $(\pm 0.79, 0, 0)$ describe a three-dimensional incommensurate magnetic order. The magnitude of the ordered magnetic moment is extremely small and is estimated to be of the order of $0.02\mu_B$. The correlation length determined from the peak width is approximately 20 lattice constants a (orthorhombic notation), with 4 Ce atoms per unit cell.

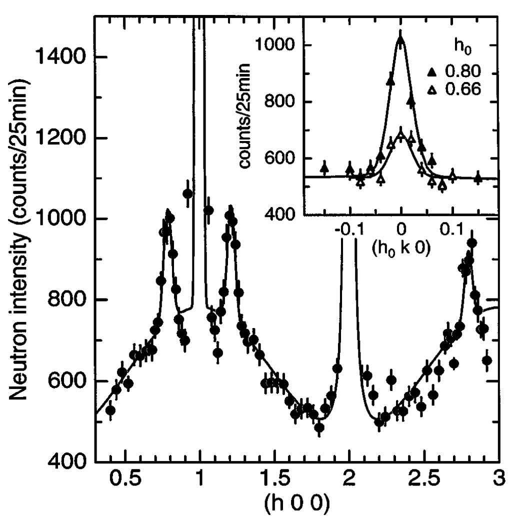

FIG. 3. Elastic neutron scattering of $CeCu_{5.8}Au_{0.2}$ along the a^* axis (reciprocal lattice direction) at 70 mK. The inset shows that all magnetic features are equally sharp in the b^* direction. The q resolution at $(1,0,0)$ is $\Delta h = 0.04$ reciprocal lattice units (rlu) and $\Delta k = 0.01$ rlu (FWHM).

The second feature is a broad q-dependent signal ranging from 500 to 700 counts/25 min, which has a factor of 3–4 higher integrated weight. It is important to stress that both features are sharp in the b^* direction with a width of order 20 unit cells as can be seen from scans along $(h_0, k, 0)$ for different h_0 (Fig. 3). The intensity of both structures decreases strongly towards T_N. However, some of the anisotropic signal remains up to 1 K, possibly due to quasielastic contributions in the experimental energy window of 0.15 meV.

In this Letter, we propose that the magnetic fluctuations associated with the second broad structure in the neutron scattering cross section dominate the critical fluctuations at the QPT in the observed T range. Starting from this assumption we will show that the logarithmic increase of C/T for decreasing T, the linear resistivity, and the phase diagram can easily be derived. We interpret this structure as arising from ferromagnetically ordered planes perpendicular to the a direction. A sine modulation as drawn in Fig. 3 would suggest that effectively two of these ferromagnetic planes with distance $a/2$ couple antiferromagnetically in the a direction, but different pairs of planes are incoherent. While it is possible to identify slightly corrugated planes of Ce atoms in the crystal structure, we do not see an obvious reason for a strongly asymmetric coupling, which could directly explain the observed two-dimensional structure. For the following, the details of the magnetic order are irrelevant—e.g., one could also fit the broad q dependence in the a^* direction with an incommensurate double structure. We will assume only that some critical two-dimensional fluctuations exist.

To derive an effective action near the phase transition in a Ginzburg-Landau-Wilson approach [10] we have to account for the damping of the critical fluctuations. To describe the critical modes, we introduce a scalar field $\Phi_{\mathbf{q}_{\|}}$; the experiments suggest a preferred direction of the magnetic moments along the c axis [7,16]. $\mathbf{q}_{\|}$ is a two-dimensional vector in the bc plane. Ordering will occur at $\mathbf{q}_{\|} = 0$. The primary damping mechanism is the coupling to particle-hole pairs. The dynamics of the quasiparticles is three dimensional; this can be inferred from the transport properties which vary at most by a factor of 2 in different directions [15]. We assume a coupling of the critical fluctuations to the heavy quasiparticles (with creation operators $c^\dagger_{\mathbf{k}}$) by the following Hamiltonian:

$$H_c = g \sum_{\mathbf{k},\mathbf{q},\alpha,\beta} (c^\dagger_{\alpha,\mathbf{k}+\mathbf{q}} \sigma^z_{\alpha\beta} c_{\beta,\mathbf{k}}) \Phi_{\mathbf{q}_{\|}} h(q_\perp) . \qquad (1)$$

$\mathbf{q} = \mathbf{q}_{\|} + \mathbf{q}_\perp$ is the transferred momentum split up in two components parallel and perpendicular to the planes. σ^i are the Pauli matrices and g is the coupling constant. $h(q_\perp)$ is some smooth function describing the magnetic structure perpendicular to the planes.

Integrating out the fermions (cf. [10] for a thorough discussion) induces damping of and interaction between the critical modes $\Phi_{\mathbf{q}_{\|},\omega}$. To quadratic order, we obtain the following contribution to the effective action in imaginary time $[\beta = 1/(k_B T)]$:

$$S_d = \frac{g^2}{\beta} \sum_{\omega_n,\mathbf{q}_{\|}} \left[\int |h(q_\perp)|^2 \chi^0(\mathbf{q}, i\omega_n)\, dq_\perp \right] |\Phi_{\mathbf{q}_{\|},i\omega_n}|^2 .$$

$\omega_n = 2\pi n/\beta$ are bosonic Matsubara frequencies. The damping is described by the imaginary part of the particle-hole bubble χ^0, $\mathrm{Im} \int dq_\perp |h(q_\perp)|^2 \chi^0(\mathbf{q}, \omega + i0) \approx \gamma\omega + O(\omega \mathbf{q}_{\|}^2)$. Near the QCP, after a proper rescaling, the effective action takes the following form:

$$S = S_2 + S_{\mathrm{int}} ,$$
$$S_2 = \frac{1}{\beta} \sum_{\omega_n} \int \Phi^*_{\mathbf{q}_{\|},i\omega_n} (\delta + \mathbf{q}_{\|}^2 + |\omega_n|) \Phi_{\mathbf{q}_{\|},i\omega_n} d^2\mathbf{q}_{\|} . \tag{2}$$

S_{int} describes the interaction between the critical modes; the leading term is given by $S_{\mathrm{int}} \approx U \int_0^\beta d\tau \int d^2\mathbf{r} |\Phi(\mathbf{r},\tau)|^4$ [10]. The distance from the QCP is measured by δ. At a critical value δ_c, determined by the strength of interaction, the system is at the QCP, i.e., $\delta - \delta_c \propto x_c - x$. The effective action corresponds to a quantum-critical theory in $d = 2$ with a dynamic exponent $z = 2$. This dynamic exponent describes the fact that if one scales momenta by $\mathbf{k} \longrightarrow \lambda\mathbf{k}$ one has to scale frequencies (or the temperature) by $\omega \longrightarrow \lambda^z \omega$.

This theory has been widely investigated by many authors (e.g., [10–12,17]), mainly in the context of the antiferromagnetic spin-fluctuation picture of high-T_c compounds. We will primarily employ the results of Millis [11], who did a careful renormalization-group study of Eq. (2). The effective dimension of this theory at $T = 0$ is $d + z = 4$; the scaling dimension of the interaction S_{int} is $4 - (d + z) = 0$, therefore it is marginal. Nevertheless, the leading behavior of the specific heat in the disordered phase can be directly calculated from the Gaussian part, S_2, of the action of Eq. (2). The free energy per volume corresponding to S_2 is

$$F = \int_0^{\Lambda_k} \frac{d^2\mathbf{q}_{\|}}{(2\pi)^2} \int_0^{\Lambda_\omega} \frac{d\epsilon}{\pi} \coth\frac{\epsilon}{2T} \arctan\frac{\epsilon}{\delta + \mathbf{q}_{\|}^2}$$
$$\propto \frac{T^2}{\omega_c} \ln\frac{\omega_c^2}{T^2 + (\delta - \delta_c)^2} . \tag{3}$$

Note that $\delta_c = 0$ in this approximation; ω_c is a typical cutoff energy of the order of the Kondo temperature in the system. Consequently, the coefficient of the specific heat $\gamma = C/T = -d^2F/dT^2$ diverges logarithmically with decreasing temperature at the QCP, as observed in the experiment. For $\delta > \delta_c$, i.e., $x < x_c$, γ stays finite. Note that the logarithmic terms do not arise from some marginal operators. They are due to a pure phase space effect, which typically happens at $d + z = 2z$ [11].

By a solution [11] of the scaling equations for the model of Eq. (2) a phase diagram emerges as displayed in Fig. 1. Region I, the low temperature phase for $x > x_c$, is the ordered phase. The behavior near T_N will depend on the structure of the order parameter, e.g., whether the ordering is of Ising type or how it is stabilized by three-dimensional coherence. The Ginzburg criterion for our model in Eq. (2) predicts [11] that T_N is proportional to the distance from the critical point $T_N \propto x - x_c$. This relation is fulfilled by the experiment over an astonishingly large range. The other crossover lines are hard to analyze quantitatively with the existing experimental data; therefore we do not attempt to fit them. The qualitative trend is, however, consistent with the phase diagram. In Fig. 1 we include the theoretical curves of [11]. Region II is dominated by the fluctuations of the finite-temperature phase transition; in region III true quantum-critical behavior with $\gamma \propto \ln(T_0/T)$ and, as we shall see, $\rho \approx \rho_0 + A'T$ can be observed. In this quantum-critical regime—in [11] it is called "classical Gaussian regime"—the temperature is the most important energy scale. The correlation length ξ depends on temperature as $\xi^{-2} \propto T$ with logarithmic corrections. In region IV, a pure crossover regime which is probably hard to observe, ξ is determined by the distance from the critical point $\xi^{-2} \propto x_c - x$, while energy fluctuations and therefore the specific heat are governed by the temperature with $\gamma \propto \ln(T_0/T)$. For region V, FL behavior is expected with a linear specific heat $C/T \propto -\ln(x_c - x)$ and a finite correlation length $\xi^{-2} \propto x_c - x$ as in region IV.

The whole scenario is in qualitative agreement with the experiments [5–7]. In particular, the logarithmic increase of the specific-heat coefficient in region III, the T-linear resistivity which we will calculate in the following, and the linear increase of T_N with x are confirmed by the experiments.

For the calculation of the resistivity we closely follow Hlubina and Rice [18], who have calculated the resistivity of electrons in two dimensions coupled to quantum-critical antiferromagnetic spin fluctuations. In our case the dynamics of the quasiparticles is three dimensional as mentioned above. Our starting point is Eq. (1) and the corresponding collision term in a Boltzmann equation:

$$\left.\frac{\partial f_{\mathbf{k}}}{\partial t}\right|_{\mathrm{coll}} = \frac{2g^2}{T} \sum_{\mathbf{k}'} \int_{-\infty}^{\infty} d\omega\, n(\omega) f^0_{\mathbf{k}'} (1 - f^0_{\mathbf{k}}) (\varphi_{\mathbf{k}'} - \varphi_{\mathbf{k}})$$
$$\times\, \delta(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'} - \omega)\, \mathrm{Im}\chi_{\mathbf{k}'_{\|}-\mathbf{k}_{\|}}(\omega) . \tag{4}$$

Here we have already linearized the collision term; the occupation of a state with momentum $\mathbf{k}$ is given by $f_{\mathbf{k}} = f^0_{\mathbf{k}} + \varphi_{\mathbf{k}} (\partial f^0_{\mathbf{k}}/\partial\epsilon)$, where $f^0_{\mathbf{k}} = f^0(\epsilon_{\mathbf{k}})$ is the usual Fermi function; $n(\epsilon)$ is the Bose function. We have omitted the factor $|h(q_\perp)|^2$, the qualitative behavior of the resistivity is not influenced by this smooth function. $\chi_{\mathbf{q}}(\omega) = \langle \Phi^*_{\mathbf{q},\omega} \Phi_{\mathbf{q},\omega} \rangle$ is the order-parameter susceptibility. In the following we use for the disordered phase

$$\chi_{\mathbf{q}_{\|}}(\omega) \approx \frac{A}{T^* + cT + \mathbf{q}_{\|}^2 - i\omega} , \tag{5}$$

with temperature-independent constants A, c, and T^*. This phenomenological form corresponds to the behavior of the correlation length described above, $\xi^{-2} \propto T$ in the quantum-critical region up to logarithmic corrections. $T^* \propto \delta - \delta_c$ measures the distance from the QCP. Equation (5) is valid in regions III–V; we are primarily interested in the quantum-critical regime III with $T^* \ll T$. Following [18], the resistivity can be determined within Boltzmann theory from the minimization of a functional of $\varphi_{\mathbf{k}}$

$$\frac{\rho}{\rho_0} = \min_{\varphi_{\mathbf{k}}} \left[\frac{\sum_{\mathbf{k}\mathbf{k}'} W_{\mathbf{k}\mathbf{k}'} (\varphi_{\mathbf{k}} - \varphi_{\mathbf{k}'})^2}{\{\sum_{\mathbf{k}} \varphi_{\mathbf{k}} \mathbf{v}_{\mathbf{k}} \cdot \hat{\mathbf{n}} (-\partial \mathbf{f}^{\mathbf{0}}_{\mathbf{k}} / \partial \boldsymbol{\epsilon})\}^2} \right], \quad (6)$$

where $\hat{\mathbf{n}}$ is the direction of the applied electric field, $\mathbf{v}_{\mathbf{k}}$ the velocity of the electrons $\rho_0 = \hbar/e^2$, and

$$W_{\mathbf{k}\mathbf{k}'} = \frac{2g^2}{T} f^0_{\mathbf{k}} (1 - f^0_{\mathbf{k}'}) n(\epsilon_{\mathbf{k}'} - \epsilon_{\mathbf{k}}) \, \mathrm{Im}\chi_{\mathbf{k}'_{||} - \mathbf{k}_{||}} (\epsilon_{\mathbf{k}'} - \epsilon_{\mathbf{k}}).$$

Note that a second contribution due to impurity scattering has to be added, which is not given here. For simplicity we assume a spherical Fermi surface. The radial part of the momentum integration can be rewritten as an energy integration. Using $\int f^0(\epsilon)[1 - f^0(\epsilon + \omega)] d\epsilon = \omega(1 + n(\omega))$ and $\int_0^\infty \omega n(\omega)[1 + n(\omega)] \, \mathrm{Im}\chi_{\mathbf{k}'_{||} - \mathbf{k}_{||}} \times (\omega) d\omega = I[\beta(T^* + cT + (\mathbf{k}'_{||} - \mathbf{k}_{||})^2)] TA$ with $I[x] \approx \pi^2/3x(x + 2\pi/3)$ [18] we can perform the energy integration. As long as the resistivity is dominated by impurity scattering—this is true in the whole range where a linear temperature dependence has been observed—we can assume [18] $\varphi_{\mathbf{k}} \approx \mathbf{v}_{\mathbf{k}} \cdot \hat{\mathbf{n}}$ and arrive at

$$\Delta\rho \propto T^2 \int \frac{g(\alpha)}{(T^* + cT + k_F^2 \alpha) \, (T^* + c'T + k_F^2 \alpha)} \, d\alpha,$$

$$g(\alpha) = \int\!\!\int d\Omega_{\mathbf{k}} \, d\Omega_{\mathbf{k}'} \, (\hat{\mathbf{n}} \cdot \hat{\mathbf{k}} - \hat{\mathbf{n}} \cdot \hat{\mathbf{k}}')^2 \delta[\alpha - (\hat{\mathbf{k}}'_{||} - \hat{\mathbf{k}}_{||})^2], \quad (7)$$

with $c' = c + 2\pi/3$. $\hat{\mathbf{k}}$ denotes a unit vector in the direction of $\mathbf{k}$, and $\hat{\mathbf{k}}_{||}$ its projection to the bc plane. For $\hat{n}$ not parallel to the planes, we find $g(\alpha) \approx$ const for small α, and therefore we get approximately

$$\Delta\rho \propto \frac{T}{c - c'} \ln \frac{T^* + cT}{T^* + c'T} \approx \begin{cases} T^2, & T < T^*/c' \\ T \, \frac{\ln c/c'}{c - c'}, & T > T^*/c \end{cases}. \quad (8)$$

For a finite T^* and small temperatures we recover the usual $\Delta\rho \propto T^2$ of a FL. However, in the quantum critical regime, i.e., for $T^* \ll cT$, the resistivity is linear in temperature as observed in the experiment [5,7,15].

For an electric field parallel to the planes, we obtain $g(\alpha) \propto -\alpha \ln \alpha$ and, accordingly, $\Delta\rho \propto T^2 \ln T$ for $T^* = 0$. This is, however, an artifact of our approximation of using a spherical Fermi surface and an isotropic s-wave scattering amplitude. In a more realistic approach, different directions would mix and give a linear increase of the resistivity with temperature in all directions, as observed [15]. Nevertheless, one would still expect that the linear increase of the resistivity at the QCP is largest in the direction perpendicular to the plane which is indeed observed [15].

The NFL character is also manifest in an anomalous self-energy of the electrons. Calculation of the lifetime in Born approximation at $T = 0$ using Eqs. (1) and (5) gives $1/\tau_{\mathbf{k}} \propto \epsilon_{\mathbf{k}} \ln 1/\epsilon_{\mathbf{k}}$ for directions parallel to the planes and $1/\tau_{\mathbf{k}} \propto \epsilon_{\mathbf{k}}$ in the a direction. Averaging over the Fermi surface results in $\langle 1/\tau_{\mathbf{k}} \rangle \propto \epsilon_{\mathbf{k}} \ln 1/\epsilon_{\mathbf{k}}$, in sharp contrast to the usual $1/\tau_{\mathbf{k}} \propto \epsilon_{\mathbf{k}}^2$ in a Fermi liquid.

In [5,7] the static susceptibility for $x = x_c$ was fitted by $\chi \propto 1 - \alpha\sqrt{T}$ from the lowest measuring temperature of 80 mK up to 3 K. However, the data can also be reasonably well described by $\chi \approx a_0 + 1/(a_1 + a_2 T)$ for temperatures up to 1.4 K. One would expect a susceptibility of the latter form for two antiferromagnetically coupled planes with an order-parameter susceptibility as given by Eq. (5). Further theoretical and experimental studies are needed to clarify this point. It will also be important to investigate further the interplay of the two- and three-dimensional order which could finally lead to a change in the observed properties at some lower temperature. We think that the explanation of the phase diagram, of the linear increase of the resistivity with temperature, and of the anomalous specific heat in $CeCu_{6-x}Au_x$ by a quantum-phase transition with $d = 2, z = 2$ is already promising. To our knowledge, this would establish the first clear experimental realization of such a theoretical scenario.

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