Two-dimensional fluctuations at the quantum-critical point of CeCu_{6-x}Au_x

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The heavy-fermion system CeCu_{6-x}Au_x exhibits a quantum critical point at \( x_c \approx 0.1 \) separating nonmagnetic and magnetically ordered ground states. The pronounced non-Fermi-liquid behavior at \( x_c \) calls for a search for the relevant quantum critical fluctuations. Systematic measurements of the inelastic neutron scattering cross section \( S(q,\omega) \) for \( x = 0.1 \) reveal rod-like features in the reciprocal \( ac \) plane translating to two-dimensional (2d) fluctuations in real space. We find 3d magnetic ordering peaks for \( x = 0.2 \) and 0.3 located on these rods which hence can be viewed as 2d precursors of the 3d order.

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Continuous quantum phase transitions which occur in a strict sense only at temperature \( T = 0 \) are driven by quantum fluctuations instead of thermal fluctuations as for ordinary classical phase transitions [1,2]. This leads to unusual and rich behavior even at finite temperatures in the neighborhood of the critical point. Because of the uncertainty principle the energy scale of fluctuations introduces a time scale which leads to an intricate coupling of static and dynamic critical behavior. For instance, the critical behavior of the specific heat will depend on the dynamical critical exponent \( z \) relating the typical lifetime \( \xi \) and correlation length \( \xi \) of critical fluctuations, \( \xi \sim \xi^z \). Such a quantum phase transition can be achieved by changing a coupling parameter which plays a role analogous to temperature in ordinary phase transitions. In recent years, many physical realizations of quantum phase transitions have been found. The case of a magnetic-nonmagnetic transition in heavy-fermion metals is particularly interesting because of the involvement of itinerant electrons.

Excitations of a system of interacting itinerant electrons in a metal, i.e. quasiparticles, are usually described within the Fermi-liquid theory, with the specific heat \( C \propto T \), a Pauli susceptibility independent of \( T \), and an electrical resistivity contribution \( \Delta \rho \propto T^2 \) due to quasiparticle-quasiparticle scattering. Interactions renormalize the quasiparticle masses with respect to the free-electron mass \( m_0 \). Even in heavy-fermion systems with quasiparticle masses as high as several 100 \( m_0 \), Fermi-liquid behavior is the rule rather than the exception [3]. In heavy-fermion systems, the coupling parameter tuning the magnetic-nonmagnetic transition is the (antiferromagnetic) exchange interaction \( J \) between \( 4f \) or \( 5f \) magnetic moments and conduction electrons [3]. If it is strong, a local singlet state is formed via the Kondo effect around each \( 4f \) or \( 5f \) site, leading to a nonmagnetic ground state. On the other hand, a weak (but non-zero) exchange interaction leads to a Rudermann-Kittel-Kasuya-Yosida coupling between moments and hence to magnetic order. In the exemplary system CeCu_{6-x}Au_x doping of CeCu_6 with the larger Au atom leads - via lattice expansion - to a weakening of the Kondo effect and hence to long-range antiferromagnetic order for \( x > x_c \approx 0.1 \), with a linear increase of the Néel temperature \( T_N \propto (x-x_c)\mu \), i.e. \( \mu = 1 \).

At \( x_c \) where \( T_N \) vanishes, i.e. around the quantum critical point, pronounced deviations from Fermi-liquid behavior occur. This non-Fermi-liquid (NFL) behavior is seen, e.g. in the specific heat where \( C/T \propto -\ln(T/T_0) \) over nearly two decades and in the resistivity where \( \Delta \rho \propto T \). It is precisely this NFL behavior at the quantum critical point that stirred a lot of interest [4] since it cannot be explained in terms of a transition driven by three-dimensional (3d) fluctuations, because for an antiferromagnet with \( d = 3 \) and \( z = 2 \), \( C/T \propto 1 - B\sqrt{T} \) and \( \Delta \rho \propto T^{3/2} \) would be expected [4].

A step forward towards the solution of the NFL puzzle in CeCu_{6-x}Au_x was to realize [7] that 2d critical fluctuations coupled to quasiparticles with 3d dynamics will indeed lead to \( \gamma = C/T \propto -\ln(T/T_0) \), \( \Delta \rho \propto T \) and \( \mu = 1 \) as experimentally observed. Elastic neutron scattering experiments at 0.07 K on CeCu_{6.8}Au_{0.2} with \( T_N = 0.25 \) K showed, in addition to peaks attributed to (short-range) antiferromagnetic order, broad maxima along the \( a^* \) direction that were much sharper in the \( b^* \) direction [7]. This latter feature was interpreted in terms of ferromagnetic planes perpendicular to the \( a \) direction (orthorhombic notation) and thus provided a possible scenario of the \( d = 2 \), \( z = 2 \) universality class [7]. Without any direct evidence it is certainly hard to believe that 2d correlations are dominating an intrinsically 3d alloy, even if the thermodynamics strongly support such a picture. Therefore it is essential to investigate the quantum critical fluctuations directly by inelastic neutron scattering.
ordered state for the \( x = 0.3 \) meV in CeCu

where \( T < 100 \) mK at an energy transfer \( \hbar \omega = 0.1 \) meV derived from Fig. 1 and from further measurements. To corroborate the rod-like nature of \( S(q, \omega) \) for \( x = 0.1 \), further scans across the peaks were performed along independent directions, one in the \( a^*c^* \) plane parallel to the rod-like feature. They, too, reveal a width of comparable magnitude as can be seen from Fig. 3b and c. In order to interpret \( S(q, \omega) \) of CeCu5.9Au0.1, we recall that a rod-like feature in the plane given by next-nearest neighboring Ce atoms. Thus,

\( (h_0 0 l) \) along \( c^* \) for fixed \( h = h_0 \). The peak at \( (1.2 \ 0 \ 0) \) splits when moving away from the \( a^* \) axis. The solid lines present Lorentzian fits with a width of \( (0.24 \pm 0.02) \) Å\(^{-1}\) for all scans shown. However, the main point is that the peak height remains roughly constant across the whole Brillouin zone (cf Fig. 3a). The width along \( c^* \) is comparable to the width of the \( (1.2 \ 0 \ 0) \) maximum along \( (h 0 0) \) (cf. Fig. 1a). This suggests a rod-like feature of the dynamical magnetic response. It does not, however, extend along the \( a^* \) axis as previously assumed but in an oblique direction.

Fig. 1b shows that for \( x = 0.1 \) there is a very rich structure of \( S(q, \omega) \) in the \( a^*c^* \) plane, as derived from scans \( (h_0 0 0) \) along \( c^* \) for fixed \( h = h_0 \). The peak at \( (1.2 \ 0 \ 0) \) splits when moving away from the \( a^* \) axis. The solid lines present Lorentzian fits with a width of \( (0.24 \pm 0.02) \) Å\(^{-1}\) for all scans shown. However, the main point is that the peak height remains roughly constant across the whole Brillouin zone (cf Fig. 3a). The width along \( c^* \) is comparable to the width of the \( (1.2 \ 0 \ 0) \) maximum along \( (h 0 0) \) (cf. Fig. 1a). This suggests a rod-like feature of the dynamical magnetic response. It does not, however, extend along the \( a^* \) axis as previously assumed but in an oblique direction.

\[ S(q, \omega) \] in the reciprocal \( ac \) plane in two perpendicular directions at very low \( T < 100 \) mK at an energy transfer \( \hbar \omega = 0.1 \) meV. The \( (h 0 0) \) scan (Fig. 1a) reveals a broad double maximum at \( (0.8 \ 0 \ 0) \) and \( (1.2 \ 0 \ 0) \). This double maximum is only resolved at small \( \hbar \omega \). For instance, for \( \hbar \omega = 0.25 \) meV only a single broad feature centered at \( (1 \ 0 \ 0) \) is seen. Hence it may be thought of as developing from the broad maximum observed at the same \( q \) for \( \hbar \omega = 0.3 \) meV in CeCu6 [8]. Upon entering the magnetically ordered state for the \( x = 0.2 \) alloy, the double-peak structure appears as a (quasi-)elastic feature for \( x = 0.2 \) that represents short-range ordering evidenced by a width in \( q \) that is considerably larger than the \( q \) resolution [9].

The experiments were carried out at the triple-axis spectrometer IN14 at the Institut Laue-Langevin, Grenoble with a fixed final neutron energy \( E_f = 2.7 \) meV \( (k_f = 1.15 \) Å\(^{-1}\)), giving an energy resolution (FWHM) of 0.07 meV. The CeCu6-xAu\(_x\) single crystals were grown with the Czochralski method in a W crucible. The specific heat of the sample with \( x = 0.1 \) exhibits the NFL behavior \( C/T \propto -\ln(T/T_0) \) as measured down to 60 mK, in agreement with previous samples of the same Au concentration.

Fig. 1 shows \( q \) scans of the dynamic structure factor \( S(q, \omega) \) in the reciprocal \( ac \) plane in two perpendicular directions at very low \( T < 100 \) mK at an energy transfer \( \hbar \omega = 0.1 \) meV. The \( (h 0 0) \) scan (Fig. 1a) reveals a broad double maximum at \( (0.8 \ 0 \ 0) \) and \( (1.2 \ 0 \ 0) \). This double maximum is only resolved at small \( \hbar \omega \). For instance, for \( \hbar \omega = 0.25 \) meV only a single broad feature centered at \( (1 \ 0 \ 0) \) is seen. Hence it may be thought of as developing from the broad maximum observed at the same \( q \) for \( \hbar \omega = 0.3 \) meV in CeCu6 [8]. Upon entering the magnetically ordered state for the \( x = 0.2 \) alloy, the double-peak structure appears as a (quasi-)elastic feature for \( x = 0.2 \) that represents short-range ordering evidenced by a width in \( q \) that is considerably larger than the \( q \) resolution [9].

Fig. 2 shows the peak positions in the \( a^*c^* \) plane for \( x = 0.1 \) and \( \hbar \omega = 0.1 \) meV derived from Fig. 1 and from further measurements. To corroborate the rod-like nature of \( S(q, \omega) \) for \( x = 0.1 \), further scans across the peaks were performed along independent directions, one in the \( a^*b^* \) plane along \( b^* \) and one in the \( a^*c^* \) plane perpendicular to the rod-like feature. They, too, reveal a width of comparable magnitude as can be seen from Fig. 3b and c. In order to interpret \( S(q, \omega) \) of CeCu5.9Au0.1, we recall that a rod-like feature in the plane given by next-nearest neighboring Ce atoms. Thus,
the observed quasi 2d correlations strongly support the proposed scenario of 2d spin fluctuations coupled to quasiparticles with 3d dynamics, although the quasi 2d correlations are not ferromagnetic as initially supposed.

The 2d fluctuations apparently are the precursor of the 3d magnetic ordering. Indeed, the Bragg points for samples not too far from the magnetic instability, e.g. \(x = 0.2\) and 0.3, are located on the rods for \(x = 0.1\). For \(x = 0.2\) in addition to the rather broad double maximum at \(q = (0.8 \ 0 \ 0)\) and \((1.2 \ 0 \ 0)\) [7] we find resolution-limited peaks at \((0.625 \ 0 \ 0.275)\) and at lattice-equivalent positions in reciprocal space. The inset in Fig. 2 displays a (1,375,0,1) scan of such a Bragg peak. The main frame of Fig. 2 shows that its position is indeed on one of the rods as is the position of the short-range order peaks along \(a^*\). However, we have not observed a 3d precursor for \(x = 0.1\), i.e. enhanced scattering intensity around the Bragg peak for \(x = 0.2\). This is an important point in favor of the 2d scenario. For \(x = 0.3\), the Bragg position remains almost unchanged while no short-range order peaks on the \(a^*\) axis were detected. For \(x = 0.5\) a sudden reorientation of the magnetic ordering vector is observed, with incommensurate order along \(a^*\) with \(\tau = (0.59 \ 0 \ 0)\) [11] which is then roughly constant up to \(x = 1\) [11]. The reorientation of \(\tau\) occurring between \(x = 0.3\) and 0.5 deserves further study.

Returning to the quantum-critical point at \(x = 0.1\), we recall that in the \(d = 2, z = 2\) scenario we expect the following generic form of the \(\omega\)– and \(q\)-dependent susceptibility describing magnetic fluctuations in the plane [12,2]:

\[
\chi^{2d}_{\parallel}(\omega) \approx \frac{1}{q_0^2} \left( \frac{1}{\xi^2 + q_0^2} - i \omega/\omega_0 \right)^{-1}
\]

(1)

The imaginary part of the susceptibility is directly proportional to the magnetic structure factor \(S(q, \omega)\) measured with inelastic neutron scattering, \(S(q, \omega) = (1 + n_B(\omega)) \text{Im} \chi^{2d}_{\parallel}(\omega) f(q_\perp)\), where \(n_B(\omega)\) is the Bose-function. The smooth function \(f(q_\perp)\) describes the weak \(q\)-dependence perpendicular to the planes, i.e. along the rod-like structures shown in Fig. 2. \(q_\parallel\) is the momentum in the plane, i.e. perpendicular the rods. \(q_0\) and \(\omega_0\) are constants that vary only slightly with temperature \(T\) and depend only weakly on the momentum along the rods. We expect that the above equation is valid (up to logarithmic corrections) for small momenta and frequencies \(\omega, \omega_0 q_0^2/q_0^2 \ll k_B T_K\), where \(T_K \approx 6 \text{K}\) is the Kondo temperature in the system. We have neglected the small anisotropy within the planes. The effective correlation length is given by \(\xi\). It is expected to vary strongly with \(T\), \(\omega_0/(q_0 \xi)^2 \approx A k_B T\) with \(A\) being a constant of order one varying only logarithmically with \(T\) [33].

The \(q\)-scans were performed with an energy transfer of 0.1 meV (=1.2K-kB) which is large compared to the temperature of 70 meV. Therefore we expect that the width of the peaks shown in Fig. 2 and 3 does not measure the true correlation length \(\xi\) but determines the ratio

\[
\frac{q_0^2}{\omega_0} \approx (0.1 \pm 0.02) \text{meV A}^{-1}
\]

(2)

The important question arises whether the observed magnetic fluctuations can be related to the NFL behavior of the thermodynamic quantities at the quantum critical point, i.e. to the logarithmically diverging specific-heat coefficient. Actually, the prefactor of the specific-heat coefficient per area of a plane is fully determined by the quantum critical theory [4] \(\gamma^{2d} = (n/12)(q_0^2/\omega_0) \ln(T_0/T)\) where \(n\) is the number of spin components. We use \(n = 1\), as the magnetic anisotropy [4] suggests an Ising system. \(T_0\) is an unknown temperature scale of the order of \(T_K\). To calculate the specific heat per volume one has to know the distance \(L\) of the planes – note that especially in an incommensurate structure such a distance is only an effective quantity. Then the molar specific heat is given by

\[
\gamma \approx 2 \frac{V_M \gamma_{\text{plane}}}{L} \ln(T_0/T) \approx (1.3 \pm 0.2) \frac{J}{\text{mol K}^2} \text{ A ln}(T_0/T)
\]

(3)

\(V_M\) is the volume per mol of CeCu5,9Au0,1, the factor 2 takes into account that the correlations show up in two different directions (see Fig. 2). This value has to be compared to the measured [4] specific heat coefficient.
\( \gamma = 0.6 \frac{1}{\ln(T_0/T)} \) which is indeed of the same order of magnitude as our estimate \(^3\). From the crystal structure one would expect \( L \) to be of the order of 4 to 10\( \text{Å} \) while from \(^3\) we obtain \( L \approx 2 - 3 \text{Å} \) which is somewhat too small. However, one has to take into account the considerable theoretical uncertainty, e.g. arising from the definition of \( L \) in an incommensurate system or the unknown effective number of spin-components in this anisotropic system. In addition, the momenta and frequencies used in our analysis are quite large.

Therefore, we think that the semi-quantitative agreement of the width of the rods in \( q \) space compared to the specific heat gives strong support for the idea that the 2\( d \) fluctuations are responsible for the observed NFL behavior.

As a final point, we discuss the energy dependence of the critical modes. Fig. 4 shows energy scans at exactly the \( q \) value of the magnetic order in CeCu\( _{5.8} \)Au\( _{0.2} \) (cf. inset of Fig. 2). The solid lines indicate a fit comprised of a Gaussian and a quasi-elastic Lorentzian (convoluted with the resolution) with full-width \( \Gamma = 0.15 \text{Å}^{-1} \) for different temperatures: \( \circ \) 70 mK, \( \bullet \) 500 mK and \( \blacksquare \) 5 K. For comparison an energy scan at \( q = (1.8, 0, 0) \) for \( T = 70 \text{mK} \) is displayed \(^5\). The inset shows energy scans at \( (1.2, 0, 0) \) in CeCu\( _{6-x} \)Au\( _x \) for \( x = 0 \) \((T = 80 \text{mK})\) and \( x = 0.1 \) \((T = 50 \text{mK})\), \( k_f = 1.15 \text{Å}^{-1} \). The background is subtracted for all scans.

In conclusion, we have identified the critical two-dimensional fluctuations leading to non-Fermi-liquid behavior in CeCu\( _{5.9} \)Au\( _{0.1} \) with a systematic study of quasielastic neutron scattering. From the observed dynamic susceptibility a semi-quantitative agreement with the prefactor of the logarithmic increase of the specific-heat coefficient is found.

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