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Dynamical susceptibility and magnetic-field effect at the quantum critical point in $CeCu_{6-x}Au_x$ from Cu NQR-NMR relaxation

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^{63,65}Cu NQR-NMR relaxation measurements in $CeCu_{6-x}Au_x$ for $x=0$, $x=0.1$, and $x=0.8$ are used to derive insights on temperature (T) and magnetic-field (H) dependences of the spin dynamics around the quantum critical point (QCP). The relaxation rate $2W$ is related to the $\mathbf{k}$ integrated, low energy, generalized susceptibility $\chi(\mathbf{k},\omega,H,T\rightarrow 0)$. For $x=0$ a Fermi-liquid behavior is detected, while for $x=0.8$ the temperature dependence of $2W$ is the one expected for a nearly antiferromagnetic metal ordering at $T_N=2.2$ K. Instead, for $x=0.1$, around the QCP, a response function of the form suggested by neutron scattering, namely of two-dimensional character with anomalous exponent and (ω/T) scaling, is found to explain the main experimental findings. An effect is observed in the low-temperature range for $H\gtrsim 1$ T, with a crossover to a gapped phase for the spin excitations at a field-dependent temperature.

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One of the most fascinating issues in recent years has involved the spin dynamics and the low-energy excitations in strongly correlated metals undergoing a quantum (zero temperature) phase transition.[1,2] Heavy-fermion metals, in particular, have been found to be driven through a quantum critical point (QCP) at the crossover between a Fermi-liquid (FL) paramagnetic metal and an antiferromagnetic (AF) metal. The intermetallic alloy $CeCu_{6-x}Au_x$ is considered the prototype of these systems. In fact $CeCu_6$ is a heavy-fermion paramagnet, with a Kondo temperature $T_K\simeq 6$ K, which in the low-temperature range displays FL behavior.[3] For $x>0.1$, instead, the compound is a weakly correlated AF metal which orders at a temperature T_N increasing linearly with increasing x, up to $T_N\simeq 2.3$ K for $x\simeq 1$. In $CeCu_{5.9}Au_{0.1}$ the FL and long-wavelength paramagnons scenario is abandoned and the QCP is attained.[4] The quantum phase transition is believed to result from the competition between the Kondo effect, which tends to screen the Ce magnetic moments and the long-range RKKY interaction favoring an ordered magnetic state. In principle, also an external magnetic field H can be used to tune the system around the QCP and to suppress the AF state for $x>0.1$.

Relevant insights on the magnetic fluctuations near the QCP have been derived from inelastic neutron scattering.[5,6] In particular, a response function of two-dimensional (2D) character, with rods in the reciprocal space for energy around $\hbar\omega=0.1$ meV, with anomalous critical exponent and (ω/T) scaling, has been conceived. However, the role of a strong magnetic field and, in particular, the low-energy spin excitations ($\hbar\omega<0.1$ meV) hardly detectable by neutron scattering have not been explored. This is the main aim of the present report, where insights on the generalized susceptibility $\chi(\mathbf{k},\omega,H,T\rightarrow 0)$ are obtained by means of ^{63,65}Cu NQR-NMR relaxation measurements in $CeCu_{6-x}Au_x$, for $x=0$, $x=0.1$, and $x=0.8$, in the low-temperature range, in external magnetic fields from $H=0$ (NQR) up to $H=12$ T. The spin-lattice relaxation of the Cu nuclei is driven by the time dependence of the magnetic hyperfine interaction with the electrons and in this way one has access to the magnetic response function over all the Brillouin zone.

The samples have been prepared by argon arc-melting the elements on a water cooled copper hearth with a tungsten electrode, with zirconium as getter. To grant good homogeneity the buttons were turned over and remelted several times. Weight losses were generally smaller than 0.5%. The alloys were then annealed in a furnace at 1000 K for 170 h and finally water quenched. The samples have been carefully characterized by x-ray diffraction, SQUID magnetization measurements, and by the search of a possible broadening and/or shift of the NQR lines. All the data confirmed the excellent quality of the samples and the absence of detectable magnetic impurities. To ensure a better penetration of the rf the samples were crushed to powders.

In Fig. 1(a) the ^{63,65}Cu NQR spectrum between 5 and 12 MHz in $CeCu_6$ is reported. Figure 1(b) shows the effect of Au for Cu substitution on the ^{63}Cu NQR line at about 11.3 MHz. In the $x=0$ sample the full width at half intensity of this line is about 26 kHz (for comparison see Refs. 7 and 8). It should be remarked that in spite of the broadening due to lattice stresses and to the electric-field gradients distribution induced by Au at the Cu sites, the resonance line at 11.3 MHz (used in all NQR relaxation measurements) still remains well resolved with respect to other lines, thus granting lack of contamination in the relaxation recovery laws (see later on). The recovery of the ^{63}Cu NQR echo signal after saturation of the resonance line was found strictly single exponential, from which the relaxation rate $1/T_1=2W$ was extracted. In Fig. 2 the temperature dependence of the NQR relaxation rates is reported. The data for $T\lesssim 1$ K in $CeCu_6$ are from Ref. 7, while the results for the sample at $x=0.1$ below 1 K are taken from the recent paper by Walstedt *et al.*[9]

One should remark that in this latter report[9] the relaxation rate $1/T_1$ is defined as $6W$. Furthermore the measurements have been carried out by the above authors on the NQR line

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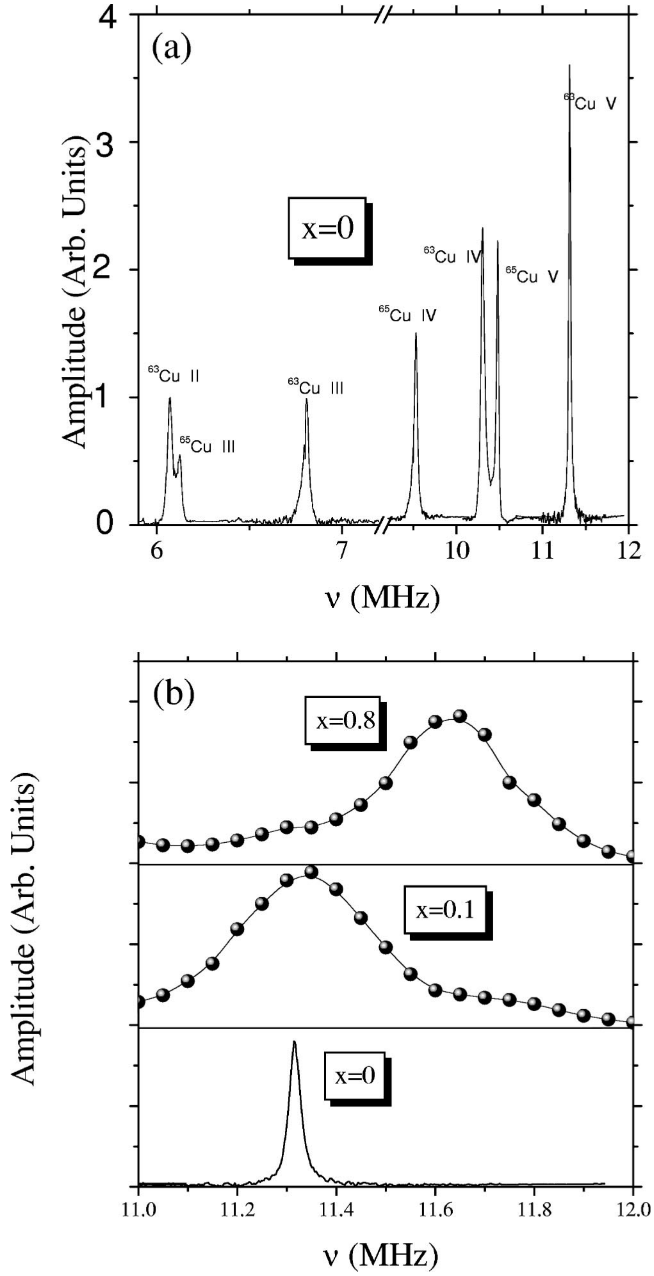

FIG. 1. ^{63,65}Cu NQR spectrum in $CeCu_6$ (a). The different Cu sites have been labeled according to Ref. 8. In part (b) of the figure the modification of the resonance line around 11.3 MHz upon Au for Cu substitution is evidenced. For $x=0.1$ and $x=0.8$ the spectrum is obtained from the envelope of the spin echo amplitude recorded at different irradiation frequencies, while for $x=0$ the direct Fourier transform of half of the echo is reported.

at 6.25 MHz [see spectrum in Fig. 1(a)] and therefore a different hyperfine factor $\mathcal{A}^2$ has to be taken into account in comparison of the data.[10] Moreover, in comparing our data with those in Ref. 9, it should be stressed that these authors have observed at short times spin-diffusion effects and nonexponential recoveries. Most likely this phenomenon[9] is due to a simultaneous irradiation of two NQR lines [see spectrum in Fig. 1(a)], the broadening in the Au substituted samples preventing the separation of the two components. On the

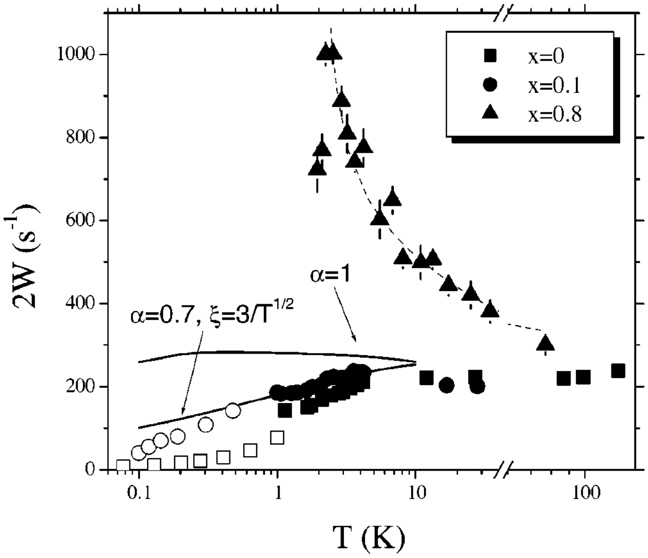

FIG. 2. ^{63}Cu NQR relaxation rates $1/T_1=2W$ in $CeCu_{6-x}Au_x$ for $x=0$, $x=0.1$, and $x=0.8$. The low-temperature data for $x=0$ (△) are from Ref. 7, while those for $x=0.1$ (○) from Ref. 9. The dashed line is the best fit behavior according to a power law $2W \propto (T-T_N)^n$ with the critical exponent $n=-0.2$. The solid lines correspond to Eq. (3) in the text for $\alpha=1$ and for $\alpha=0.7$.

other hand, thanks to the high sample quality, our measurements could be carried out on the single line around 11.3 MHz, with full irradiation by the rf sequence saturating the NQR levels, thus granting a recovery described by a single exponential, with no spectral diffusion at short times.

The effect of the external magnetic field H on $2W$, on approaching the QCP in $CeCu_{5.9}Au_{0.1}$, is reported in Fig. 3. In the presence of the field the recovery laws are modified

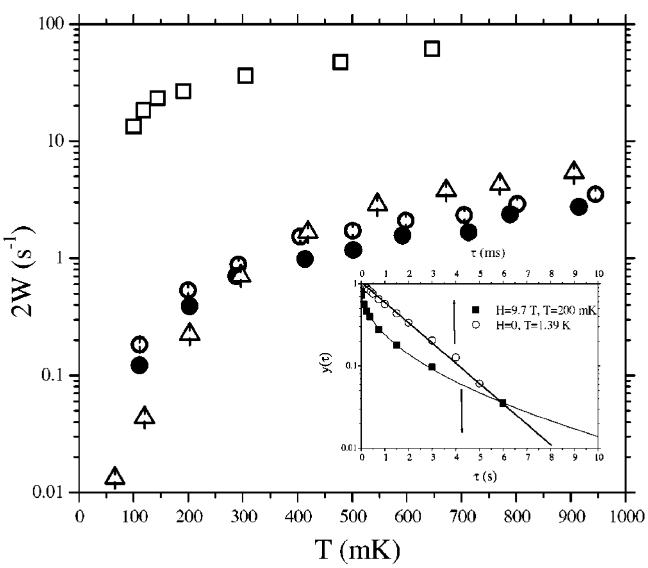

FIG. 3. Temperature dependence of the relaxation rates in $CeCu_{5.9}Au_{0.1}$ in the presence of the magnetic field H for (□) $H=0$, from Ref. 9; (△) $H=6.7$ T; (○) $H=9.8$ T; (●) $H=11.2$ T. In the inset two typical NMR and NQR recovery plots of the nuclear magnetization, after a saturating rf pulse sequence, are reported. Notice the different horizontal scale for the two plots.

and in order to extract the relaxation rates we used the laws for Zeeman perturbed NQR (for field such that $^{63}\gamma H/2\pi < 11.3$ MHz) or for quadrupole perturbed NMR (in the opposite limit and then by taking into account the superposition of adjacent lines). Some modifications in the hyperfine coupling term $\mathcal{A}^2$ (see later on) have to be expected. These changes do not have any effect on the temperature dependence.

Now we analyze the experimental results for $2W$ in term of the generalized spin susceptibility. One has[11,12]

$$2W = \frac{\gamma^2}{2N} k_B T \mathcal{A}^2 \sum_{\mathbf{k}} \left[\frac{\chi''(\mathbf{k},\omega_m)}{\omega_m} \right], \qquad (1)$$

where the hyperfine coupling term $\mathcal{A}$ has been assumed **k** independent (as appropriate for itinerant pseudoparticles) and scalar, while ω_m is the measuring frequency.

For the $x=0.8$ sample the behavior of $2W$ as a function of temperature (Fig. 2) is not far from the one expected[11] for weakly coupled AF metal. A power law of the form $2W \propto (T-T_N)^n$ with $T_N = 2.2$ K and critical exponent $n=-0.2$ describes rather well the experimental data, in correspondence to the critical slowing down of the spin dynamics on approaching the ordering temperature. For $CeCu_6$ the FL behavior, expected for $T \ll T_K$, is practically obeyed, as noticed in a preceding work.[7] Those results, including echo dephasing and temperature dependences up to 300 K, will be discussed elsewhere.[13] Here we focus on the temperature and field dependences of $2W$ for $x=0.1$, in the low-temperature range.

From inelastic neutron scattering data, combined with heuristic arguments[2,4–6] a response function of 2D character has been envisaged,

$$\chi_{2D}^{-1}(\mathbf{k},\omega,T) = k_B \left[\frac{(T - i\omega/a)^\alpha}{c} + f^\alpha(\mathbf{k},T) \right], \qquad (2)$$

with an anomalous exponent $\alpha \neq 1$, (ω/T) scaling and renormalized Curie-Weiss constant c. From Eq. (1), by using Eq. (2) with an expansion of $f^\alpha(\mathbf{k},T)$ in even powers of **q** starting from the critical wave vector, under the condition that all the excitation frequencies $\Gamma_{\mathbf{q}} \geq \Gamma_{AF}$ remain larger than ω_m, one derives the relaxation rate. First we refer to the case of zero external field and one has

$$2W = \frac{\gamma^2}{2N} k_B T \mathcal{A}^2 \sum_{2D}^{BZ} \frac{c}{a^\alpha} \frac{[1+(q\xi)^{2\alpha}]^{-2}}{k_B T^{\alpha+1}}, \qquad (3)$$

where $(aT)^\alpha = \Gamma_{AF}$ is the critical frequency and $\xi(T,H=0)$ is the correlation length. In the assumption $\alpha=1$, from 2D integration of Eq. (3) over the Brillouin zone one would have

$$2W = \frac{\gamma^2}{8\pi} \mathcal{A}^2 \frac{S(S+1)}{aT} \frac{q_D^2}{1+\xi^2 q_D^2} \qquad (4)$$

(q_D a Debye-like wave vector) with the correlation length given by[14,15] $\xi(T,H=0) = (1/\sqrt{T}) \ln(T_K/T)$ (in lattice units). In Fig. 2 the solid line for $\alpha=1$ corresponds to Eq. (4), for $a=10^{10}$ rad s^{-1} K^{-1} and $\mathcal{A}=3.8$ kOe (according to Ref. 8).

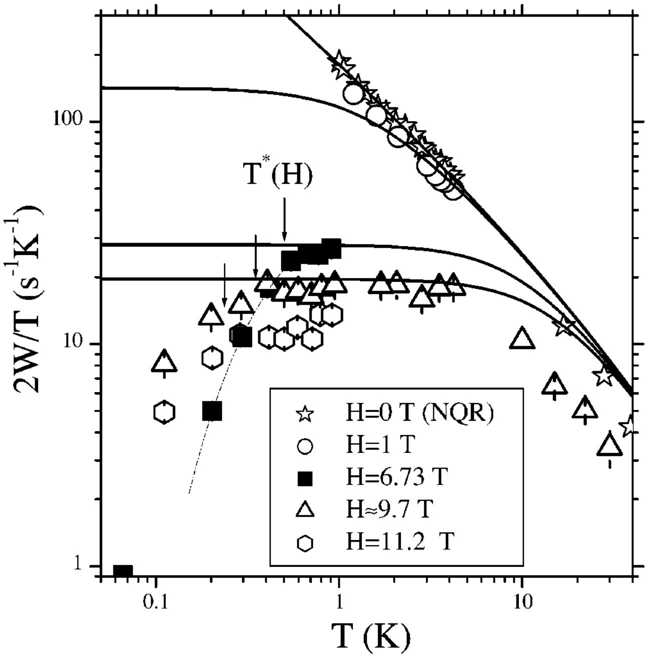

FIG. 4. ^{63}Cu relaxation rates, scaled by T, for $CeCu_{5.9}Au_{0.1}$ approaching the QCP at different external fields [the raw data are in Fig. 3. A normalization of the hyperfine factor has been performed in order to account for the irradiation of different lines related to the various sites (Ref. 8)] and comparison with the theoretical behaviors according to Eq. (3) in the text, with an effective temperature given by Eq. (5) (solid lines for representative values of the field $H=0$, $H=1$ T, $H=6.73$ T, and $H=9$ T). One should remark the departure of the data for $T<T^*(H)$ (arrows). The dash-dotted line tracks the behavior $W \propto \exp(-\Delta/T)$ for $\Delta(H=6.73\text{ T})=0.6$ K (see text). The occurrence of the gapped phase is confirmed also by the data at 9.7 and 11.2 T.

It is evident that the experimental results for $T \to 0$ indicate $\alpha \neq 1$, as already noticed from neutron scattering.[3,5]

For $\alpha \neq 1$ a numerical integration is required to derive $2W$ from Eqs. (1)–(3). The line in Fig. 2 has been obtained in correspondence to $\alpha=0.7$, again $\mathcal{A}=3.8$ kOe and $\Gamma_{AF} = (1.5\times 10^{10} T)$rad s^{-1} and for $\xi=3$ at $T=1$ K. This value of the correlation length is close to the one derived from neutron scattering, where it was found[6] $\xi^2 = (\omega_0/k_B T q_0^2)$ with $(\omega_0/q_0^2) = 10$ meV Å^2, yielding $\xi = 10/\sqrt{T}$ Å. Furthermore it is noted how below 1 K the critical frequency could hardly be detected by means of neutron scattering, because of resolution limits.

Now we discuss the effect of the external magnetic field. Since close to quantum criticality the only scale, for both energy and field dependence of the response function, is temperature, one can reformulate the derivation leading to Eqs. (3) and (4) through Eq. (2), by substituting T with an effective temperature $T_{eff}(H)$. The most straightforward generalization for the effective temperature around a critical point, including the effect of the field is[2]

$$T_{eff}(H) = [T^2 + (g\mu_B H/k_B)^2]^{1/2} \equiv [T^2 + (T_{mag})^2]^{1/2}. \qquad (5)$$

By considering first the case $\alpha=1$ (for simplicity of discussion), in the presence of the field instead of Eq. (4) one derives

$$(2W/T)\propto[(T^2+T_{mag}^2)\xi^2(T,H)]^{-1}. \qquad (6)$$

This equation, and therefore the choice of T_{eff}, is qualitatively supported by the experimental data. The trend of the results reported in Fig. 4 indicates practically no field dependence for $T\gg T_{mag}$. For strong field Eq. (6) yields (W/T) temperature independent[16] and decreasing as $1/H$, approximately as observed in Fig. 4 in the temperature range 0.5–2 K.

The solid lines reported in Fig. 4 are the theoretical behaviors obtained from numerical integration of Eq. (1) having used for T, including in $\xi(T,H)$, the effective temperature given in Eq. (5). As it appears from the figure the experimental results are rather well justified by the behaviors derived for representative values of H, for temperature above a given temperature $T^*(H)$. Below this temperature (about 200 mK for $H=11$ T and 600 mK for $H=6.7$ T) a drastic departure of the data from the theoretical behavior is observed, with a sudden decrease of $2W/T$ on cooling. As it appears from Fig. 4 below $T^*(H)$ W takes a temperature dependence approximately of the form $W\propto\exp[-\Delta(H)/T]$, typical of a system with a gap in the spin excitations. As an order of magnitude one has $\Delta(H=6.7\text{ T})\simeq 0.6$ K.

The breakdown below T^* of the quantum critical susceptibility in the form given by Eq. (2) suggests an intriguing three-dimensional (T,x,H) phase diagram around the QCP, since for $T\rightarrow 0$ and $H\rightarrow 0$ quantum criticality again holds. One could speculate that the gap originates from a field effect on the spectral density reminiscent of the one yielding a decrease in magnetoresistivity.[17] In this scenario the bell-shaped form of $T^*(H)$ would result from the competition between the increase of the splitting of the spin-up and spin-down subbands and the decrease with H of the Kondo temperature.

Summarizing, from ^{63}Cu NQR-NMR spin-lattice relaxation measurements around the quantum critical instability in $CeCu_{5.9}Au_{0.1}$different insights in the low-energy spin excitations and correlated spin dynamics have been achieved. In general the 2D character of the magnetic fluctuations, the anomalous critical exponent, and the energy/temperature scaling suggested by neutron scattering are confirmed by our data, based on the **k**-integrated low-energy response function. Different aspects involving the effect of an external magnetic field in the low-temperature range, where the critical frequency is below neutron scattering resolution limit, are pointed out by our results.

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