Magnetic field induced non-Fermi liquid to Fermi liquid crossover at the quantum critical point of YbCu$_{5-x}$Au$_x$

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The temperature (T) dependence of the muon and $^{63}$Cu nuclear spin-lattice relaxation rates $1/T_1$ in YbCu$_{4.4}$Au$_{0.6}$ is reported over nearly four decades. It is shown that for $T \to 0$ $1/T_1$ diverges following the behaviour predicted by the self-consistent renormalization (SCR) theory developed by Moriya for a ferromagnetic quantum critical point. On the other hand, the static uniform susceptibility $\chi_s$ is observed to diverge as $T^{-2/3}$ and $1/T_1 T \propto \chi_s^2$, a behaviour which is not accounted by SCR theory. The application of a magnetic field $H$ is observed to induce a crossover to a Fermi liquid behaviour and for $T \to 0$ $1/T_1$ is found to obey the scaling law $1/T_1(H) = 1/T_1(0)[1 + (\mu_B H/k_BT)^2]^{-1}$.

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Strongly correlated electron systems with competing interactions are known to show rather rich phase diagrams, with crossovers or phase transitions which depend on the relative magnitude of the competing energy scales. A paradigmatic example is represented by heavy-fermion intermetallic compounds, where a quantum phase transition between Fermi liquid (FL) and magnetic ground-states is typically observed upon varying the single-ion Kondo coupling $J$ and the density of states at the Fermi level $D(E_F)$. The modification of these two parameters affect both the coherence temperature $T^*$, below which the f electrons delocalize and a FL behaviour is observed, and the transition temperature to a magnetic long-range order, which is determined by RKKY interaction. At the quantum critical point (QCP) $T^* \to 0$, the Fermi liquid regime is never attained and a rather peculiar behaviour of the response functions is observed down to $T \to 0$, the so-called non-Fermi liquid (NFL) regime. The QCP can be tuned by different parameters, as the chemical composition, the pressure and the magnetic field, which control the hybridization between f and s electron orbitals, i.e. $J$ and $D(E_F)$. In spite of the significant experimental efforts, an overall understanding of how the dynamical spin susceptibility behaves in the NFL regime on approaching the QCP and how it is affected by external parameters, as the magnetic field, is still missing.

YbCu$_{5-x}$Au$_x$ is a heavy-fermion intermetallic compound which has been studied in recent years mostly with techniques of macroscopic character, ranging from specific heat to magnetization and resistivity measurements. On the basis of these experimental results a tentative phase diagram as a function of $x$ has been outlined. The coherence temperature $T^*$, which for $x = 0$ was estimated around 5 K, vanishes around $x \approx 0.4$, where a quantum phase transition to a long-range magnetic order is expected. Still, it has to be pointed out that the transition temperature to the magnetically ordered phase for $x > 0.5$ has been determined just from the change of slope in the resistivity vs. temperature, raising some doubts on the accuracy of its estimate. Moreover, a careful analysis of the chemical and structural properties of YbCu$_{5-x}$Au$_x$ solid solutions have shown that homogeneous compounds with AuBe$_5$-type structure can be grown at ambient pressure only for $x \geq 0.4$, questioning some of the previous experimental observations.

Here we will show, on the basis of zero and longitudinal field muon spin relaxation ($\mu$SR), nuclear quadrupole resonance (NQR) and magnetization measurements, that for $x \approx 0.6$ a ferromagnetic QCP is attained. Moreover, it will be shown that the T-dependence of the muon spin-lattice relaxation rate $1/T_1$ for $T \to 0$ can be suitably described within the self-consistent renormalization (SCR) theory developed by Moriya. At temperatures above 1 K both $1/T_1$ and the copper nuclear spin-lattice relax-
ation rate \(1/T_1^{\mu}\) are observed to scale with the square of the static uniform susceptibility. Hereafter, with \(1/T_1\) we shall refer both to the muon and nuclear spin-lattice relaxation rate, unless when it will be specified. Finally, it was found that the application of a magnetic field \(H\) is observed to lead to a crossover from the critical NFL to a FL behaviour and to a significant reduction in the relaxation rate.

The experiments were performed on YbCu\(_{5-x}\)Au\(_x\) powders grown according to the procedure reported in Ref. 7. \(\mu SR\) experiments were performed at PSI Swiss muon source on LTF beam line. In order to reduce the background contamination, when the decay rate of the muon polarization \(1/T_1^{\mu} \leq 1\mu s^{-1}\) the data acquisition was performed in MORE mode\(^a\). The decay of the muon polarization could be nicely fit by

\[
P_\mu(t) = A \exp[-(t/T_1^{\mu})^\beta] + B ,
\]

with an initial asymmetry \((A + B) \approx 24\%\), over all the explored \(T\) range (Fig. 1). Here \(B\), of the order of a few percent is the background contribution due to the sample holder and cryostat environment. No evidence of a transition to a magnetically ordered phase was observed down to 20 mK, at variance with previous findings based on resistivity measurements\(^b\). As we shall see from the magnetic filed dependence of \(1/T_1^{\mu}\), the muon relaxation is dynamical, namely driven by spin fluctuations and not by a static field distribution.

\[
\frac{1}{T_1^{\mu}} = \frac{\gamma^2 k_B T}{2N} \sum_q |A_q|^2 \frac{\chi^\prime(q, \omega_R)}{\omega_R} , \tag{2}
\]

where \(\gamma\) is the gyromagnetic ratio and \(|A_q|^2\) is the form factor, giving the hyperfine coupling of the muon (or nuclei) with the spin excitations at wave-vector \(q\). From Fig. 2 one notices that \(\gamma_{63}^{\mu}\) and \(\mu^+\) \(1/T_1\) differ by a factor 236. From the above equations, taking into account that \((\gamma_{63}^{\mu}/\gamma_{63}^\mu)^2 \approx 144\), one realizes that the hyperfine couplings of \(63\)Cu nuclei and of the interstitial \(\mu^+\) are quite similar.

Following Ishikagi and Moriya\(^{13}\) it is convenient to write the dynamical spin susceptibility in terms of two

\[\left|\frac{\chi^s(q, \omega_R)}{\omega_R}\right| = \left(\frac{\chi^s(\vec{q}, \omega_R)}{\omega_R}\right)\left|A_{\vec{q}}\right|^2 , \tag{2}\]
characteristic parameters $T_0$ and $T_A$ which characterize the width of the spin excitations spectrum in frequency and $q$, respectively. For ferromagnetic correlations one has\textsuperscript{13,14}

$$\chi(q, \omega) = \frac{\pi T_0}{\alpha_Q T_A} k_B 2\pi T_0 x(y + x^2) - i\omega h$$  \hspace{1cm} (3)

where $x = q/q_D$, with $q_D$ a Debye-like cutoff wave-vector, $\alpha_Q$ a dimensionless interaction constant close to unity for a strongly correlated system, and $y = N_A/2\alpha_Q k_B T_A \chi_s$. Here the susceptibility is per spin and in 4$\mu_B^2$ units and has the dimensions of the inverse of an energy, while $T_A$ and $T_0$ are in Kelvin. From the previous expression one can derive $\chi^\prime(q, \omega_R)/\omega_R$ by taking the limit $\omega_R \to 0$, since $\hbar\omega_R \ll k_B T$. Then, by integrating $\chi^\prime(q, \omega_R)/\omega_R$ in $q$, over a sphere of radius $q_D$, one derives

$$\frac{1}{T_1} = \frac{\gamma^2 A^2}{2} \frac{T}{4\pi k_B T_A T_0} \alpha_Q 2y(1 + y)$$  \hspace{1cm} (4)

Now, if $T_A \gg T$ in the T-range of interest then $y \ll 1$\textsuperscript{13,14} and one can simplify the previous expression in the form

$$\frac{1}{T_1} \simeq \gamma^2 A^2 \frac{3\hbar}{8a} \frac{T}{T_0} \frac{\chi_s}{N_A}$$  \hspace{1cm} (5)

This expression corresponds to the one derived by Ishikagi and Moriya\textsuperscript{13,14}, provided one takes into account that their hyperfine coupling constants are in kOe/$\mu_B$. This is the behaviour typically observed in the presence of a magnetic ground-state.\textsuperscript{15} For a three-dimensional system approaching a ferromagnetic QCP $\chi_s$ is expected to diverge as $T^{-4/3}$ at low-temperature and, accordingly, $1/T_1 \propto T^{-1/3}$, exactly the behaviour observed in our measurements. However, it has to be pointed out that SCR theory would predict a spin-lattice relaxation rate $1/T_1 \propto T\chi_s$, at variance with the experimental findings. Therefore, although the T-dependence of $1/T_1$ seems to agree with predictions of SCR theory we do not find a full consistency of our experimental findings with the theoretical expectations. As we shall see in the next paragraph, also the magnetic field dependence of $1/T_1$ can hardly be explained by SCR theory.

![FIG. 3: Inverse of the molar spin susceptibility in YbCu$_{5-x}$Au$_{x}$ vs. $T^{2/3}$. One notices that for $x = 0.6$ a good scaling is found and that the Curie-Weiss temperature vanishes.](image)

![FIG. 4: T-dependence of the muon spin-lattice relaxation rate in YbCu$_{4.4}$Au$_{0.6}$ at three different magnetic fields. The solid line evidences that for $H=8$ kGauss, at low-T, $1/T_1 \propto \alpha T + b$, where the small offset $b$ should be either associated with the uncertainty in the background corrections or to the fact that a certain angular dependence of the effect of the magnetic field has to be expected. In the inset the magnetic field-dependence of the muon $1/T_1$ is shown. The solid line shows the scaling law $1/T_1(H) = 1/T_1(0)[1 + (\mu B H/k_B T)^2]^{-1}$, with no adjustable parameter.](image)

Now we turn to the discussion of the effect of a magnetic field on the muon longitudinal relaxation rate $1/T_1^\mu$. From Fig.4 one notices that the magnetic field progressively reduces $1/T_1^\mu$. Remarkably the effect is significant at low T, where $1/T_1^\mu(H = 0)$ is large, while it is reduced at high T where $1/T_1^\mu(H = 0)$ is low. This behaviour is the opposite of what one would expect if the relaxation had to be associated with a static field distribution $\Delta H$. In fact, in that case one would expect a significant reduction of the relaxation when $\gamma H \approx 10/T_1^\mu(H = 0)^{16}$, at variance with the experimental findings. Moreover, the similar behaviour of $1/T_1^{\mu3}$ and $1/T_1^\mu$ suggest that the effect of the magnetic field rather has to associated with a modification in the dynamical spin susceptibility. In fact, in other intermetallic compounds as CeCu$_{6-x}$Au$_x$\textsuperscript{17} and
YbRh$_2$Si$_2$ showing an antiferromagnetic QCP a similar effect of the magnetic field has been reported. In particular, it has been observed that the magnetic field drives the system away from the QCP towards a FL ground-state. Here we observe that at $H = 8$ kGauss, $1/T_1$ decreases linearly with decreasing $T$ (Fig. 4), as expected for a FL. Our results are perfectly consistent with the experimental findings by Tsujii et al., who observed in YbCu$_{4.1}$Au$_{0.6}$ for $H \approx 1$ Tesla, a crossover in the T-dependence of the susceptibility from a $T^{3/2}$ to a $T^2$ power law, the one typical of a FL.

The effect of the magnetic field cannot be explained within SCR theory, as an initial raise in $1/T_1$ and then a decrease should be expected. This is not the case here, in fact, for $H = 1$ kGauss $1/T_1$ increases on cooling down to the lowest temperature, while for $H = 8$ kGauss $1/T_1$ is practically always decreasing on cooling. In the CeCu$_{6-x}$Au$_x$ the effect of the magnetic field was accounted for by a renormalization of the temperature scale to $T_m = T[1 + (\mu_B H/k_B T)^2]^{-1/2}$. Here, by taking into account that $1/T_1 \propto T \chi_T$ $\propto T^{-1/3}$, one would expect that $1/T_1(H) \propto [1 + (\mu_B H/k_B T)^2]^{-1/6}$, at variance with the experimental findings.

On the other hand, we find that the relaxation rate obeys the scaling $1/T_1(H) = 1/T_1(0)[1 + (\mu_B H/k_B T)^2]^{-1}$, with no adjustable parameter (Fig. 4). Accordingly, for $T \to 0$ $1/T_1(H) \propto 1/H^2$. Remarkably also in MnSi the muon relaxation rate was observed to decrease with $H^2$ on approaching the transition to the magnetic ground-state. Also in that case SCR theory could not explain the field dependence of the relaxation rate and its decrease with $H^2$ was tentatively ascribed to the progressive quenching of the helical components of the critical fluctuations and to the increase of $q = 0$ fluctuations. This explanation, of course, cannot hold here where the critical fluctuations are at $q = 0$. The scaling law experimentally found in this work should rather take into account the progressive departure from the ferromagnetic QCP, induced by the magnetic field, and the insurgence of the FL ground-state.

In conclusion, from the T-dependence of the muon and $^{63}$Cu nuclear spin-lattice relaxation rates in YbCu$_{4.1}$Au$_{0.6}$ we found that for $T \to 0$ $1/T_1$ diverges following the behaviour predicted by the self-consistent renormalization theory developed by Moriya for a ferromagnetic quantum critical point and no evidence of any phase transition could be detected down to 20 mK. On the other hand, at low-T the static uniform susceptibility $\chi_s$ was observed to diverge as $T^{-2/3}$ and, accordingly, $1/T_1 T \propto \chi_s^2$, a behaviour which cannot be explained within SCR theory. Finally, the application of a magnetic field $H$ is observed to induce a crossover to a Fermi liquid behaviour and for $T \to 0$ $1/T_1$ is found to obey the empirical scaling law $1/T_1(H) = 1/T_1(0)[1 + (\mu_B H/k_B T)^2]^{-1}$.

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