^{63}Cu Nuclear Relaxation in the Quantum Critical Point System $CeCu_{5.9}Au_{0.1}$

R. E. Walstedt,* H. Kojima, and N. Butch
Department of Physics, Rutgers University, Piscataway, New Jersey 08855

N. Bernhoeft
CEA-Grenoble, F-38054 Grenoble Cedex, France
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^{63}Cu NQR measurements of the ^{63}Cu T_1 are reported for the quantum critical point system $CeCu_{5.9}Au_{0.1}$ over temperatures ranging from 0.1 up to 4.2 K. Below ~1 K the magnetization recovery exhibits a stable, nonexponential decay function which we believe signals the onset of 2D quantum critical fluctuations, as has been noted in the literature. We find $T_1^{-1} \propto T^{0.75}$ for the region $T < 1$ K. The observed temperature dependence is in agreement with a phenomenological model of non-Fermi liquid behavior based on the uniform susceptibility but is inconsistent with calculations based on susceptibility peaks identified via neutron scattering experiments.

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Of the many f-electron systems which have been found to exhibit non-Fermi liquid (NFL) behavior characteristics at low temperatures, the $CeCu_{6-x}Au_x$ compounds have proved to be a source of extraordinarily rich and complex behavior [1]. In a simple physical picture, the f-s exchange coupling J_{fs} is responsible for the RKKY coupling which produces antiferromagnetism as well as for the Kondo condensation of the localized f moments, both effects occurring at low temperature [2]. At the boundary between the resulting heavy fermion and antiferromagnetic regions lies a quantum critical point (QCP) at $x \simeq 0.1$. In the vicinity of this composition a rich variety of NFL-type effects has been reported, including a logarithmic temperature dependence of specific heat [3,4], uniform susceptibility varying as $\chi(T) \sim 1 - \alpha T^{1/2}$ [4], resistivity linear in T [5], and anomalous dynamic susceptibility $\chi(\vec{q}, \omega)$ [6]. Neutron scattering studies have also reported magnetic fluctuation peaks with characteristic NFL behavior at various points in the Brillouin zone (BZ) [7,8].

In this Letter we report NQR studies yielding ^{63}Cu nuclear spin-lattice relaxation time T_1 data on one of the five copper sites [9] in the compound $CeCu_{5.9}Au_{0.1}$ over a temperature range from 0.1 to 4.2 K. The variation of T_1 with T offers a probe of the dynamic susceptibility at frequencies near zero for $\vec{q}$ vectors over the bulk of the BZ. The ^{63}Cu NQR frequencies in the doped compound are essentially the same as in $CeCu_6$ [10]. We find nonexponential relaxation behavior for the 6.25 MHz ^{63}Cu line, with a temperature characteristic given approximately by $T_1^{-1} \propto T^{0.75}$. This result is substantially different from the behavior reported earlier for one of the other ^{63}Cu NQR transitions in this compound [11] and also stands in sharp contrast with Fermi liquid (Korringa) behavior $T_1^{-1} \propto T$.

It is interesting and important to relate our T_1 results to other NFL effects in this system. In this Letter we endeavor to do this using a phenomenological model of $\chi(\vec{q}, \omega)$ put forward recently, wherein NFL effects are introduced in terms of a flat distribution of quasiparticle relaxation rates [12]. Using this model, we are able to show that our data for $T_1(T)$ are consistent with NFL behavior reported for $\chi_0(T)$ [4], but inconsistent with behavior found near the major fluctuation peaks [13]. A major conclusion is that the T_1 results confirm the existence of NFL behavior over regions of the BZ away from the fluctuation peaks which have been reported [7,8], which is a unique characteristic of a QCP system.

Using a conventional pulsed NMR spectrometer, we have carried out zero-field NQR measurements on a single-crystal sample from the same batch of material which was employed previously for neutron scattering studies [6]. Single-crystal boules received were crushed in an agate mortar and mixed with an approximately equal volume of 10 μm alumina powder to permit full penetration of rf fields and to minimize rf heating effects. Low frequency (16 Hz) ac susceptibility measurements on the resulting powder mixture gave results in agreement with the literature values [4] (temperature dependence only) down to $T \sim 0.15$ K, below which a sudden increase in $\chi(T)$ occurred. Since we found no corresponding effect in the NQR results, we have attributed the latter effect to a minority phase in the sample which was probably generated by the grinding process.

The sample material was placed inside of the mixing chamber of a dilution refrigerator. Temperatures were monitored with a cerium magnesium nitrate thermometer, also mounted in the mixing chamber, using a squid-based susceptibility bridge. Stable temperatures in the range 0.1 to ~2 K were routinely achieved.

A scan of the NQR frequency spectrum revealed that all of the lines reported for $CeCu_6$ [10] were present, with noticeable broadening owing to the Au doping. Initial T_1 measurements on the strong 11.8 MHz ^{63}Cu NQR line were found to be subject to significant rf pulse heating effects for $T \ll 1.0$ K. The problem was found to be

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greatly diminshed for the 6.25 MHz ^{63}Cu NQR line. For that reason and on account of its good intensity, the 6.25 MHz line was chosen for the T_1 study reported here.

T_1 was measured for the 6.25 MHz ^{63}Cu NQR line using a single π pulse for saturation followed by a $\pi/2$-π spin-echo sequence. Spin-echo data were accumulated in a signal averager in order to improve the signal-to-noise ratio. In a typical T_1 scan, a programmed sequence of echo pulse delays along with data transfer to hard disk were executed using a PC control system. The pulse sequences were well separated in order to avoid long-term heating effects. For example, at $T = 0.1$ K, where $T_1 \sim 25$ msec, the repetition frequency was 0.1 Hz. T_1 data sequences typically ran automatically for 12–15 h. Temperature and NQR spectrometer conditions were monitored for stability. Nuclear polarization (T_1) recovery curves following the saturation pulse are shown in Fig. 1.

As was reported earlier [11], the low-temperature nuclear magnetization recovery in this system was found to be nonexponential. Furthermore, owing to incomplete saturation of the entire NQR line with a single π pulse, noticeable spectral diffusion effects were present at the beginning of the magnetization recovery curves. For this reason, the rapid initial portions of the decay curves shown in Fig. 1 have been discarded. The remaining recovery waveform ($\Delta t > 0.5$ msec) has been fitted with a stretched exponential function

$$M(\Delta t)/M_0 = 1 - d\exp[-(\Delta t/T_{1s})^{\alpha}] \quad (1)$$

for temperatures ranging from 0.1 to 1.03 K (Fig. 1, solid lines), where d is the initial saturation parameter and T_{1s} is a characteristic relaxation time. The deviation of α from unity indicates the degree of nonexponential behavior. For $T < 1$ K, high quality fits are obtained with $\alpha = 0.73 \pm 0.05$ (see the inset in Fig. 1). For $T = 1$ K and above, fits gave single-exponential relaxation within experimental error.

We highlight the success of the foregoing fitting procedure with a scaled plot of all of the data from Fig. 1 shown in Fig. 2, where the time scale shown is appropriate for the data at $T = 0.1$ K. Thus, the relaxation times T_{1s} give relative rates for the T_1 relaxation over the range of temperatures covered. For comparison, the dotted line shows a simple exponential curve with the same $1/e$ point to illustrate the extent of nonexponential behavior.

We also conducted a sample heating test at $T = 0.1$ K, where the same pulses used for T_1 data were applied, except that the saturation π pulse frequency was shifted ~200 kHz away from the resonance frequency. The pulse amplitude remained approximately the same, however, so that its heating effect, if any, would be unchanged. The behavior of signal amplitude with pulse separation is shown by the filled squares at the top of Fig. 2. A

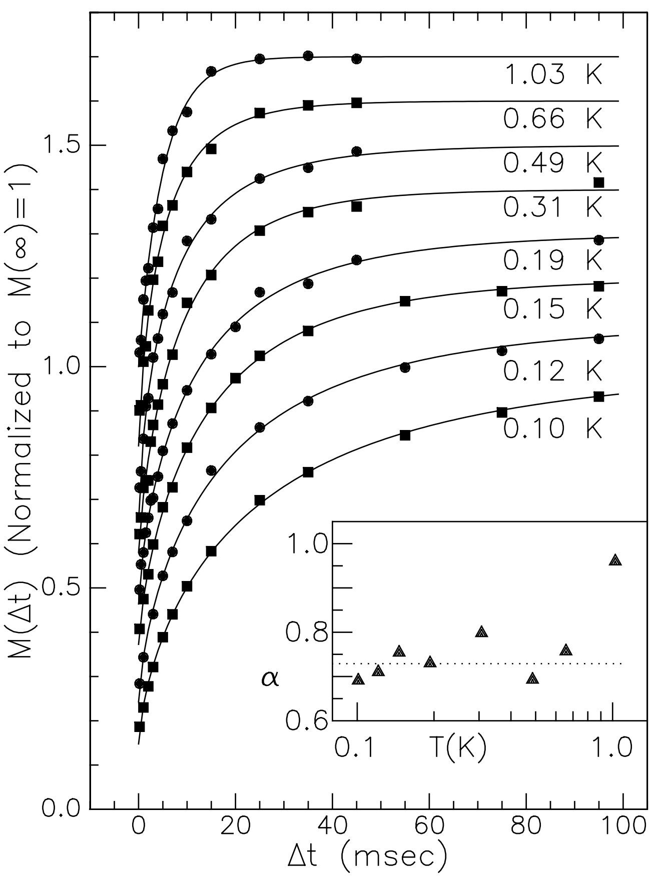

FIG. 1. ^{63}Cu T_1 magnetization recovery curves from the 6.25 MHz NQR line for temperatures ranging from 0.1 to 1.03 K. Solid lines show fits of the stretched exponential function [Eq. (1)], and the inset shows fitted values of the exponent α. The data have been scaled to approach unity as $\Delta t \to \infty$ and are displaced vertically by 0.1 from one another for clarity.

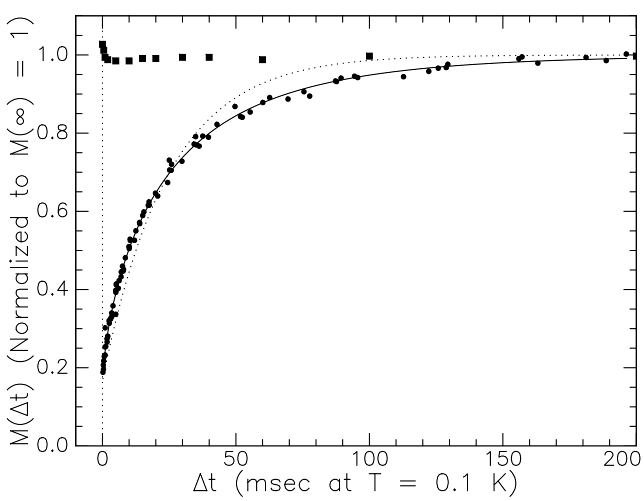

FIG. 2. The ^{63}Cu T_1 data from Fig. 1 for $T < 1$ K (closed circles) are scaled to fit a single stretched exponential function with $\alpha = 0.73$, showing that the distribution of magnetic fluctuations is stable in this temperature region. At the top is shown (filled squares) the response curve to an rf pulse sample heating test described in the text. All data are normalized to unity as $\Delta t \to \infty$. A simple exponential decay (dotted line) is shown for comparison.

temperature increase caused by the initial π pulse would result in the signal amplitude relaxing to a correspondingly lower value on roughly the time scale of T_1. Instead, we observe a nearly instantaneous drop in signal amplitude of ~5%, which is very likely either direct saturation or a spectral diffusion effect caused by the off-resonance pulse. We conclude that short-term heating effects are negligibly small in this body of T_1 data.

The temperature dependence of T_{1s}^{-1} obtained from the fits to Eq. (1) is shown in Fig. 3 (filled circles), along with the exponential T_1 data obtained between $T = 1.03$ and 4.2 K (open circles). The contrast between these two types of relaxation data is a reflection of the onset below 1 K of a fairly broad distribution of T_1 rates. Focusing on the T_{1s} data, we first note that it behaves roughly as $T_{1s}^{-1} \propto T^{0.75}$ (Fig. 3, dotted line). Further, the points above 1 K are seen to fall somewhat below the line. We note that T_1 and T_{1s} may not be directly comparable. The onset of nonexponential relaxation around $T = 1$ K suggests that below that point there develops a distribution of hyperfine couplings and/or fluctuation intensities somewhat different from those found at higher temperatures; i.e., there is an abrupt change in the scale and distribution of T_1 rates.

Regarding the nonexponential behavior of the T_1 data, we note that the ^{63}Cu quadrupolar energies ($I = \frac{3}{2}$) form a two-level system. Thus, in the Fermi liquid region where the HF coupling at this site is uniform, the T_1 process is necessarily a single exponential. At some temperature $T \ll T_K \sim 6$ K, NFL effects set in [7] and, concomitantly, the relaxation becomes nonexponential. We emphasize that this characteristic of the low-T T_1 process is not directly related to its NFL character, but to the spatially inhomogeneous fluctuation properties of a doped sample.

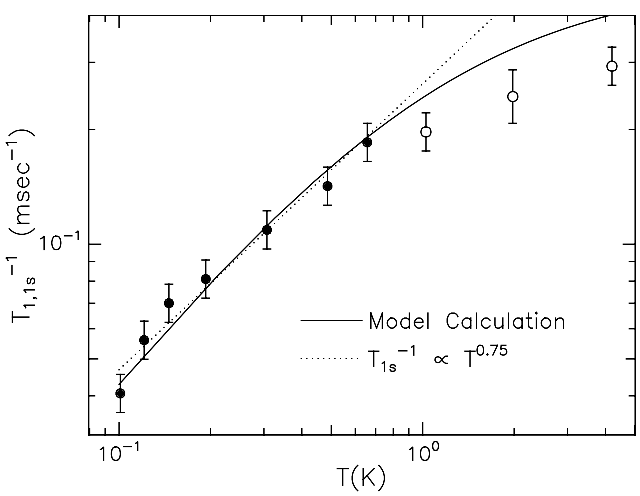

FIG. 3. A log-log plot of ^{63}Cu relaxation data T_{1s}^{-1} from Fig. 1 for $T < 1$ K (filled circles), plus exponential T_1 data between $T = 1.03$ and 4.2 K (open circles). The dotted line shows a simple power law $T^{0.75}$ and the solid line shows the behavior of the NFL model $\chi(\vec{q}, \omega)$ function [Eq. (2)], which has been fitted to the uniform susceptibility data of Ref. [4] as described in the text.

Thus, the low-temperature relaxation process for a particular 6.25 MHz ^{63}Cu site may well depend on its configuration of neighboring Au impurities. Once this fluctuation pattern is established, it appears to remain stable over a substantial range of lower temperatures.

Beyond a simple power law, we also interpret the data in Fig. 3 using the phenomenological model of Ref. [12]. In this model, the single-exponential form for magnetic fluctuations in a Fermi liquid, $\chi(\vec{q}, \omega) = \chi(\vec{q})\Gamma/(\Gamma - i\omega)$, is generalized to include a flat distribution of relaxation rates Γ extending from Γ_1 to Γ_2, giving

$$\chi(\vec{q}, \omega) = \frac{\mu_B^2 u(\vec{q})}{\Gamma_2 - \Gamma_1} \ln\left\{\frac{\Gamma_2 - i\omega}{\Gamma_1 - i\omega}\right\}, \quad (2)$$

where we have made the approximation that $\Gamma_{1,2}$ are independent of $\vec{q}$. This model has been employed to interpret NFL behavior in a variety of compounds, where Γ_2 and Γ_1 have typically been taken to be constant and linear in temperature, respectively [12]. In Eq. (2) the factor μ_B^2 is inserted so that $u(\vec{q})$ becomes a quantity of order unity for an atomic susceptibility per formula unit [14].

Here we use Eq. (2) to establish a relationship between the uniform, static ($q = 0, \omega = 0$) susceptibility $\chi_0(T)$ and the dissipative component $\chi''(\vec{q}, \omega)$, which is used to calculate the temperature dependence of T_{1s} [Eq. (1)]. From the real and imaginary parts of Eq. (2), we have $\chi_0(T) = \mu_B^2 u(0) \ln(\Gamma_2/\Gamma_1)/(\Gamma_2 - \Gamma_1)$, and $\chi''(\vec{q}, \omega) = \mu_B^2 u(\vec{q})\omega/\Gamma_1\Gamma_2$ as $\omega \to 0$. These results are used to relate the measured susceptibility [4,12] to the T_{1s} data of Fig. 3. $\chi_0(T)$ is first fitted to the above formula to determine the parameters $u(0)$ and $\Gamma_{1,2}$. In order to make contact with other data, we set $\Gamma_1(4\,\mathrm{K}) = 0.2$ meV, in agreement with neutron and specific heat results [12]. However, here we let Γ_2 be T independent. We then find that the values $u(0) = 0.441$, $\Gamma_1 = 0.0426 + 0.0394T$ meV, and $\Gamma_2 = 0.241$ meV in the above expression for $\chi_0(T)$ give an excellent fit to the experimental data [4]. It is interesting to note that $\chi_0(T)^{-1}$ has also been shown to obey a $T^{0.75}$ power law [8].

We use the above results to interpret $1/T_{1s}(T)$. Employing the foregoing expression for $\chi''(\vec{q}, \omega)$ in the fluctuation-dissipation theorem [15,16], we obtain for a particular site

$$(T_1 T)^{-1} = \frac{2u(0)\hbar\gamma_{63}^2 k_B}{g_t^2 \Gamma_1(T)\Gamma_2} \langle A_t(\vec{q})^2 \rangle, \quad (3)$$

where $\langle A_t(\vec{q})^2 \rangle = \sum_q u(\vec{q}) A_t(\vec{q})^2/u(0)$ is a weighted, squared average of the transverse HF coupling over the BZ, g_t is the transverse electronic g factor, and γ_{63} is the nuclear gyromagnetic ratio. In the NFL state of $CeCu_{5.9}Au_{0.1}$ there is a distribution of the values of $\langle A_t(\vec{q})^2 \rangle$, $[\Gamma_1(T)\Gamma_2]^{-1}$, or possibly both, which gives rise to a distribution of T_1 values, i.e., a nonexponential T_1 decay, which we have characterized with the stretched exponential relaxation time T_{1s} [Eq. (1)]. If there is a distribution of $[\Gamma_1(T)\Gamma_2]^{-1}$ values, then we presume that the foregoing evaluation based on $\chi_0(T)$ will represent average behavior which can be compared with our relaxation data. Substituting into Eq. (3), a suitable scale adjustment yields the solid line in Fig. 3. This procedure is seen to give a good account of the temperature dependence of $T_{1s}(T)$ over the range 0.1 to 1.0 K. The fitted scale factor for $T_{1s}(T)$ yields $\langle A_t(\vec{q})^2\rangle_s^{1/2} = 2.75$ kG/spin.

We compare our results with similar NQR data reported by Omuta *et al.* [11] on samples with nominally the same composition. First, our T_1 results are for the site with $\nu_Q = 6.25$ MHz, which was not studied in Ref. [11]. Different sites may differ in the strength and q dependence of the HF coupling with the Ce spin moment; thus $T_1(T)$ may differ by more than a scale factor. Nonetheless, it is something of a surprise that in Ref. [11] $T_1^{-1} \sim T^{1/2}$, in contrast with the present data. We note that at the low end of the temperature range, the slope of their data is greater than $T^{1/2}$, i.e., more in line with what we report here. Another difference with the data of Ref. [11] is that the nonexponential decay behavior extends up to $T = 2$ K or so, whereas in our case it only begins just below 1 K. One possible origin for these observed differences is a difference in sample composition.

The relaxation behavior in the region below 1 K goes very nearly as $T_1^{-1} \propto T^{0.75}$. This is clearly NFL behavior, and we believe it to be related to the 2D-like fluctuations revealed by inelastic neutron scattering [7] on this system. Nonetheless, as pointed out earlier, the behavior we observe is not compatible with detailed models of $\chi''(\vec{q}, \omega)$ which have been derived from neutron results. First, the intense fluctuations reported to occur near $\vec{q} = (1 \pm 0.2, 0, 0)$ [6] would lead to an estimate $T_1^{-1} \propto T^{0.25}$. Fluctuations have also been reported to occur near the magnetic Bragg peaks of the ordered regime [8]. The latter results lead to an expected T_1 behavior which is nearly flat over the range of our data [13] and is clearly incompatible with the present results.

We therefore conclude that the ^{63}Cu T_1 process is dominated by fluctuation behavior in regions of the BZ away from the intense fluctuation peaks which have been reported. In this connection we make several observations. First, the ^{63}Cu T_1 process is almost certainly dominated by transferred HF couplings with spin fluctuations on neighboring Ce sites. In such a case, it is possible that the q dependence of $A_t(\vec{q})$ is such that the T_1 process becomes insensitive to the fluctuation peaks noted above. Different q dependences among sites might explain the differences between our data and those of Ref. [11]. Further, Schröder *et al.* have pointed out that at the QCP composition, the $T^{0.75}$ power law behavior of $\chi_0(T)$ noted above extends also to finite $\vec{q}$ values [8]. This makes the correspondence between $\chi_0(T)$ and $\chi(\vec{q}, \omega)$ which we establish via Eq. (2) a little less mysterious, although the corresponding approximate $T^{0.75}$ power law for $T_1^{-1}(T)$ does not appear to follow from this in any simple way.

In connection with the general application of Eq. (2) to $CeCu_{5.9}Au_{0.1}$, it is important to note that results from other experimental probes, e.g., specific heat [4] and neutron scattering [6,8], appear to be dominated by large fluctuations near $\vec{q} = (\pi, \pi)$ or other special points in the BZ. Indeed, in the analysis of [12], these effects have been found to require somewhat different parameters from those which fit the uniform susceptibility $\chi_0(T)$ and $T_1(T)$. Since the latter represent behavior away from the fluctuation peaks, the T_1 results serve to verify that, unlike a 3D system with antiferromagnetic correlations, the NFL behavior for $CeCu_{5.9}Au_{0.1}$ extends over the entire BZ. This appears to be a feature uniquely characteristic of a QCP system.

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*Present address: Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken, 319-1195 Japan.

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