Glassy Spin Dynamics in Non-Fermi-Liquid UCu_{5-x}Pd_{x}, x = 1.0 and 1.5

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Local f-electron spin dynamics in the non-Fermi-liquid heavy-fermion alloys UCu_{5-x}Pd_{x}, x = 1.0 and 1.5, have been studied using muon spin-lattice relaxation. The sample-averaged asymmetry function \( G(t) \) indicates strongly inhomogeneous spin fluctuations, and exhibits the scaling \( G(t, H) = G(t/H^\alpha) \) expected from glassy dynamics. At 0.05 K \( \gamma(x=1.0) = 0.35 \pm 0.1 \), but \( \gamma(x=1.5) = 0.7 \pm 0.1 \). This is in contrast to inelastic neutron scattering results, which yield \( \gamma = 0.33 \) for both concentrations. There is no sign of static magnetism \( \gtrsim 10^{-3} \mu_B/U \) ion in either material above 0.05 K. Our results strongly suggest that both alloys are quantum spin glasses.

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Magnetic resonance \cite{1} and other \cite{2,3} experiments have demonstrated the importance of structural disorder in the breakdown of Landau’s Fermi-liquid theory in certain f-electron intermetallic compounds and alloys. Disorder-driven mechanisms have been considered for the non-Fermi-liquid (NFL) properties of some of these systems \cite{4,5}, and it is natural to consider the possibility of extremely disordered or “glassy” behavior. On theoretical and experimental grounds it is known that glassy dynamics lead to long-time correlations with distinct signatures as the freezing or “glass” temperature \( T_g \) is approached from above \cite{6}. In a spin glass the spin autocorrelation function \( q(t) = \langle S_i(t)S_i(0) \rangle \) is theoretically predicted to exhibit power-law \( q(t) = ct^{-\alpha} \) or “stretched-exponential” \( q(t) = c \exp[-(\Delta t)^\beta] \) behavior \cite{7}. Power-law correlation has been found in spin-glass AgMn using muon spin relaxation (\( \mu \)SR) \cite{8}.

This Letter describes evidence from \( \mu \)SR experiments that spin correlations in the NFL alloys UCu_{5-x}Pd_{x}, x = 1.0 and 1.5, are indicative of glassy spin dynamics. The sample-averaged muon relaxation function \( (\mu \)SR) \( G(t) \) is strongly sub-exponential, indicating a quenched inhomogeneous distribution of relaxation rates, and obeys the field-time scaling relation \( G(t, H) = G(t/H^\alpha) \) for applied magnetic field \( H \) between \( \sim 15 \) Oe and \( \sim 1 \) kOe. The field dependence corresponds to a measurement of the Fourier transform of \( q(t) \) over the frequency range \( \gamma_H/2\pi \approx 200 \) kHz–14 MHz, where \( \gamma_H = 2\pi \times 13.55 \) kHz/Oe is the muon gyromagnetic ratio. Power-law behavior of \( q(t) \) is implied by the observation \( \alpha < 1 \) \cite{9}, and also by the temperature-frequency scaling \( \gamma \) \cite{10}, although the possible connection with glassy dynamics has to our knowledge not been previously noted. The present measurements extend by three orders of magnitude the frequency range over which power-law correlations are observed in UCu_{5-x}Pd_{x}. Zero-field \( \mu \)SR above 0.05 K shows no sign of static magnetism or spin freezing in UCuPd; this together with the glassy scaling points to some form of quantum spin-glass behavior \cite{11,12,13}.

In \( \mu \)SR experiments spin-polarized positive muons are implanted into the sample, and the subsequent time evolution of the muon polarization is monitored by detecting the asymmetric distribution of positrons from the muon decay. Muon relaxation in a magnetic field applied parallel to the muon spin direction (longitudinal field) is due to thermally-excited f-electron spin fluctuations that couple to the muons. A muon at a given site experiences a time-varying local field \( \mathbf{H}_{loc}(t) \) due to fluctuations of neighboring f moments. Following Keren et al. \cite{14}, under motionally narrowed conditions the local muon asymmetry \( G(t, H) \) relaxes exponentially:

\[
G(t, H) = \exp \left[ -2\Delta^2 \tau_c(H)t \right],
\]

where \( \Delta^2 = \gamma_H^2 \langle |\mathbf{H}_{loc}|^2 \rangle \) is the time-averaged mean-square coupling constant in frequency units, and the local correlation time \( \tau_c(H) \) is given by

\[
\tau_c(H) = \int_0^\infty dt \gamma(t) \cos(\gamma_H t) = cu_c(H).
\]

We consider \( \Delta \) and the prefactor \( c \) but not the functional form of \( u_c(H) \) to vary from site to site in a disordered material. Then the sample-averaged asymmetry \( G(t, H) \) is given by

\[
G(t, H) = \int \int d\Delta dc \rho(\Delta, c) \exp \left[ -2\Delta^2 cu_c(H)t \right],
\]
where $\rho(\Delta, c)$ is the joint distribution function for $\Delta$ and $c$. It can be seen that the field and time dependence enter Eq. (3) in the combination $u_c(H)t$. This means that $\mathcal{G}(t, H)$ scales as this combination independently of the form of $\rho(\Delta, c)$.

For both the power-law and stretched-exponential forms of $q(t)$ this time-field scaling results in

$$
\mathcal{G}(t, H) = \mathcal{G}(t/H^{\gamma})
$$

(4)

after Fourier transforming $q(t)$. Here $\gamma = 1 - \alpha < 1$ for power-law correlation, and $\gamma = 1 + \beta > 1$ for stretched-exponential correlation as long as the muon Larmor frequency $\omega_\mu = \gamma_\mu H$ is much greater than $\lambda [9]$. If Eq. (3) is obeyed a plot of $\mathcal{G}(t, H)$ versus $t/H^{\gamma}$ will be universal for the correct choice of $\gamma$. The sign of $\gamma - 1$ distinguishes between power-law and stretched-exponential correlations.

Samples of UCu$_{3.5}$Pd and UCu$_{3.5}$Pd$_{1.5}$ were prepared as described previously [12]. Arc-melted ingots were crushed into powder under acetone. The powder was passed through a 90 micron sieve, and pressed with GE 7031 varnish into pellets 13 mm dia. $\times$ $\sim$1 mm thickness. $\mu$SR data were taken at the LTF facility of the Paul Scherrer Institute, Villigen, Switzerland, for temperatures between 0.05 and 1.1 K and for magnetic fields between zero and 10 kOe applied in the direction of the muon spin polarization.

Figure 1 shows $\mathcal{G}(t)$ in UCu$_{3.5}$Pd$_{1.5}$ for $T = 0.05$ K and values of applied field $H$ between 13 Oe and 2.5 kOe. The relaxation slows with increasing field. For low enough fields we expect the field dependence to be due to the change of $\omega_\mu$ rather than an effect of field on $q(t)$; a breakdown of scaling would occur for high fields where this ceases to be true. The same asymmetry data are plotted in Fig. 2 as a function of the scaling variable $t/H^\gamma$. For $\gamma = 0.7 \pm 0.1$ the data scale well over more than three orders of magnitude in $t/H^\gamma$ and for all fields except 2.5 kOe. Fields $\mu_B H \lesssim k_B T$ would be expected to affect the spin dynamics, and indeed the static susceptibility of UCu$_4$Pd is suppressed by fields $\sim$1 kOe below $\sim$0.5 K (Vollmer et al., Ref. [10]). The scaling exponent $\gamma$ is less than 1, implying that $q(t)$ is well approximated by a power law (or a cutoff power law [6]) rather than a stretched-exponential or exponential. From our data $q(t) \approx ct^{-0.3\pm0.1}$. We note again that no specific form for the muon asymmetry function has been assumed.

A scaling plot is given in Fig. 3 for UCu$_4$Pd, $T = 0.05$ K. Here the scaling exponent $\gamma = 0.35 \pm 0.1$ is significantly smaller than in UCu$_{3.5}$Pd$_{1.5}$. Scaling again breaks down for high enough fields; the data for 2 kOe clearly fall above the low-field scaling curve. Data taken in UCu$_4$Pd at $T = 0.5$ K (not shown) scale with the same exponent. This is in contrast to the scaling behavior of spin-glass $Ag_{0.995}Mn_{0.005}$ [6], where $\gamma$ varies strongly with temperature as the glass temperature $T_g = 2.95$ K is approached from above.

The fluctuation-dissipation theorem [13] relates $\tau_c(H)$ to the imaginary component $\chi''(\omega)$ of the local ($q$-independent) $f$-electron dynamic susceptibility:

$$
\tau_c(H) \approx \frac{k_B T}{\mu_B^2} \left( \frac{\chi''(\omega)}{\omega} \right)
$$

(5)

for $h\omega \ll k_B T$. INS experiments [14] show scaling of the sample-averaged $\chi''(\omega, T)$ as $\omega^{-\gamma}F(h\omega/k_BT)$, with $\gamma = 0.33$ and $F(x) = \tan(x/1.2)$ for both UCu$_4$Pd and UCu$_{3.5}$Pd$_{1.5}$. Using this form $\chi''(H)$ obtained from Eq. (5) is independent of $T$ and proportional to $H^{-\gamma}$; the latter is in accord with the $\mu$SR scaling. The INS value of $\gamma$ agrees with $\mu$SR data for UCu$_4$Pd ($\gamma = 0.35$) but not for UCu$_{3.5}$Pd$_{1.5}$ ($\gamma = 0.7$), suggesting that in the latter sample the behavior of $\tau_c$ changes in the unexplored region between the highest muon frequencies ($\sim$10 MHz) and the lowest INS frequencies ($\sim$10 GHz). Clearly high-
resolution INS studies (neutron spin echo or backscat-
tering) would be desirable between these frequencies.

To go further one must fit the \( \mu \)SR data to an appropriate functional form for the asymmetry. We have chosen the stretched-exponential

\[
\bar{G}(t) = \exp[-(\Lambda t)^K],
\]

where \( K < 1 \) gives sub-exponential relaxation corresponding to a distribution of relaxation rates. This function is purely empirical. It is used because it characterizes an \textit{a priori} unknown relaxation-rate distribution, and because the rate \( \Lambda \) conforms with a general definition of a characteristic rate by the time \( 1/\Lambda \) where \( \bar{G}(t) \) decays to \( 1/e \) of its initial value. Equation (6) fits the data in applied field to within the statistical error.

For \( H = 0 \) the data were fit to the product of Eq. (6) and the zero-field Kubo-Toyabe (K-T) function [18] characteristic of static relaxation by nuclear dipolar fields at muon sites. This form is expected when the muon local field has both static nuclear dipolar and dynamic \( f \)-moment contributions. A nuclear dipolar field \( \sim 2.3 \text{ Oe} \) was measured in both alloys for \( T \gg 1 \text{ K} \), where the contribution of U-moment fluctuations to the zero-field muon relaxation rate vanishes. Nonzero values of \( H \) were chosen large enough to “de-couple” the muon relaxation from the nuclear dipolar field [18] leaving only the dynamic U-moment contribution, so relaxation data for these fields were fit to Eq. (6) without the K-T function. Curves giving these fits are plotted in Fig. 3.

Figure 4 gives \( \Lambda(T) \) and \( K(T) \) for UCu\(_4\)Pd at three values of \( H \). Below 1.1 K \( \Lambda \) increases slowly and saturates to a constant below 0.1–0.2 K. As noted previously a temperature dependence of \( \Lambda(T) \) is not in agreement with the temperature-independent relaxation predicted from INS scaling. The exponent \( K \) is approximately 0.7 at 0.05 K, indicative of a broad distribution of relaxation rates [6], and decreases slightly with increasing temperature. Similar behavior is exhibited by \( \Lambda(T) \) and \( K(T) \) in UCu\(_{3.5}\)Pd\(_{1.5}\) (data not shown), with rates slower than in UCu\(_4\)Pd by \( \sim 30\% \) at low fields and \( \sim 100\% \) at 100–300 Oe due to the larger scaling exponent.

No anomaly is found in the zero-field data at temperatures \( \sim 0.1–0.2 \text{ K} \), where specific heat and ac susceptibility (in different samples) suggest spin-glass-like freezing [16]. The muon–\( f \)-moment coupling in UCu\(_{5-x}\)Pdx is predominantly dipolar [3] with a coupling field \( 0.55 \pm 0.05 \text{ kOe/} \mu_B \). Randomly-frozen moments of the order of 1 \( \mu_B/\text{U ion} \) would result in a muon relaxation rate \( \sim 50 \mu\text{s}^{-1} \), two orders of magnitude larger than the observed rate. This result places an upper bound of \( \sim 10^{-3} \mu_B/\text{U ion} \) on any frozen moment in UCu\(_4\)Pd or UCu\(_{3.5}\)Pd\(_{1.5}\). We believe that the discrepancy with the results of Refs. [16] results from differences in annealing conditions; \( \mu \)SR experiments to explore this question are currently underway. It is noteworthy that the saturation of \( \Lambda \) and \( K \) occurs in the same temperature range as the reported spin-glass-like freezing [16].

Our results can be compared with existing theories of disorder-driven NFL behavior. Preliminary analysis has indicated that the order of magnitude of the experimental rates at these very low temperatures cannot be directly accounted for by the simple single-ion Kondo disorder model of NFL behavior [19]. In an early version of the “Griffiths-phase” theory [8], which treats the effect of \( f \)-moment clustering, there is no dissipation in the \( f \)-electron spin dynamics, and the local cluster dynamic susceptibility is sharply resonant at a distributed charac-
teristic tunneling energy $E$:
\[
\chi''(\omega, E) \propto \delta(\omega - E) \tanh(E/2T).
\]
Together with the distribution function $P(E) \propto E^{-1+\lambda}$, where $\lambda < 1$ is a nonuniversal scaling exponent, this immediately yields
\[
\chi''(\omega) = \int dE P(E) \chi''(\omega, E) \propto \omega^{-1+\lambda} \tanh(\omega/2T),
\]
in agreement with the INS data for $\lambda \approx 0.7$ and reminiscent of our $\mu$SR results.

But the observed time-field scaling of $G(t, H)$ demonstrates that it is the local $\chi''(\omega)$ itself, not just the average $\overline{\chi''}(\omega)$, which scales as $\omega^{-\gamma}$ [cf. Eqs. (3, 4)]. This is not a property of the Griffiths-phase model, in which the scaling is found only after the average has been taken, and only for a sharply resonant (and nonscaling) form of $\chi''(\omega, E)$. A recent form of this theory [20] considers dissipative effects, which broaden $\chi''(\omega, E)$ but do not give it a scaling form. Furthermore, if the width of $\chi''(\omega)$ is much greater than $\omega$ it is not hard to show that $\overline{\chi''}(\omega)$ no longer follows $P(\omega=E)$ so that this mechanism for scaling of $\overline{\chi''}(\omega)$ is lost. It is also difficult to see how the dynamic susceptibility of Ref. [20] would yield the observed temperature dependence of $\Lambda(\omega, T)$. Thus the Griffiths-phase theory does not seem to account for our results for a number of reasons.

Muon relaxation in UC$_{6-x}$Pd$_x$ clearly indicates glassy dynamics, and the increase of $\Lambda(T)$ at low temperatures (Fig. 1) suggests a low-temperature critical point. The increase in $K(T)$ with decreasing temperature might be due to more efficient averaging over the disorder associated with a growing correlation length. But $\Lambda(T)$ saturates below $\sim 0.2$ K and there is no evidence for a true phase transition. The relaxation rates are broadly distributed ($K \lesssim 0.7$); we are never dealing with critical behavior in a homogeneous system. The distribution of rates is itself static, at least over the time scale of the experiment ($\sim 10 \mu$s), otherwise $\overline{G}(t)$ would tend to be averaged to an exponential with the average rate.

The faster relaxation (i.e., slower spin fluctuations) for $x = 1.0$ than for $x = 1.5$ implies that the relevant critical point is near the concentration for which the Néel temperature $T_N$ is suppressed to zero ($x \approx 1$). But in spin-glass AgMn $\gamma \rightarrow 1$ from below as $T \rightarrow T_g$ [4], whereas here $\gamma(x=1.5) = 0.7$, $\gamma(1.0) = 0.35 < \gamma(1.5)$. A mean-field model of a disordered Kondo alloy at a quantum critical point [13] predicts $\gamma = 1/2$ at $T = 0$, suggestive of our results but not in detailed agreement with them.

By definition NFL behavior is a property of the lowest-lying excitations of a metal, to which a low-frequency probe such as $\mu$SR is extremely sensitive. Many aspects of the spin dynamics in UC$_{6-x}$Pd$_x$ are poorly understood, but the strong disorder, glassy behavior, and absence of a phase transition strongly suggest that these alloys are quantum spin glasses.

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[1] O. O. Bernal, D. E. MacLaughlin, H. G. Lukefahr, and B. Andraka, Phys. Rev. Lett. 75, 2023 (1995).
[2] D. E. MacLaughlin, O. O. Bernal, and H. G. Lukefahr, J. Phys.: Condens. Matter 8, 9855 (1996).
[3] O. O. Bernal et al., Phys. Rev. B 54, 13000 (1996).
[4] B. Ambrosini, J. L. Gavilano, P. Vonlanthen, and H. R. Ott, Phys. Rev. B 60, R11249 (1999).
[5] C. H. Booth et al., Phys. Rev. Lett. 81, 3960 (1998).
[6] M. C. de Andrade et al., Phys. Rev. Lett. 81, 5620 (1998).
[7] E. Miranda, V. Dobrosavljević, and G. Kotliar, J. Phys.: Condens. Matter 8, 9871 (1996).
[8] A. H. Castro Neto, G. Castilla, and B. A. Jones, Phys. Rev. Lett. 81, 3531 (1998).
[9] A. Keren, P. Mendels, I. A. Campbell, and J. Lord, Phys. Rev. Lett. 77, 1386 (1996).
[10] R. G. Palmer, D. L. Stein, E. Abrahams, and P. W. Anderson, Phys. Rev. Lett. 53, 958 (1984).
[11] M. C. Aronson et al., Phys. Rev. Lett. 75, 725 (1995).
[12] S. Sachdev, Philos. Trans. R. Soc. London, Ser. A 356, 173 (1998).
[13] D. R. Grempel and M. J. Rozenberg, Phys. Rev. B 60, 4702 (1999).
[14] A. Schenck, Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics (A. Hilger, Bristol & Boston, 1985).
[15] B. Andraka and G. R. Stewart, Phys. Rev. B 47, 3208 (1993).
[16] E.-W. Scheidt et al., Phys. Rev. B 58, R10104 (1998); R. Vollmer et al., Phys. Rev. B 61, 1218 (2000).
[17] The fluctuation-dissipation theorem may be invalid in the frozen spin-glass state because of broken ergodicity, but holds in the high-temperature paramagnetic state considered here. See, e.g., K. H. Fischer and J. A. Hertz, Spin Glasses (University Press, Cambridge, 1991), pp. 138–9.
[18] R. S. Hayano et al., Phys. Rev. B 20, 850 (1979).
[19] D. E. MacLaughlin, J. Phys. Soc. Jpn. 69, Suppl. A, 33
[20] A. H. Castro Neto and B. A. Jones, Phys. Rev. B 62, 14975 (2000).