Quantum Spin Glass Relaxation in UCu₄Ni

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Abstract. Strong non-exponential relaxation due to magnetic disorder in UCu₄Ni is observed in zero- and longitudinal-field µSR measurements in this material. Zero-field experiments reveal no evidence for spin freezing down to 20 mK. Qualitatively the shape of the muon asymmetry curves in longitudinal fields is strongly affected as the field is increased from zero and less so as the field is increased towards kOe values. Analysis of the time/field scaling \((t/H)\) for the asymmetry data, performed without reference to any specific decay function or model, together with the absence of spin freezing enables one to classify this system as a quantum-spin-glass; i.e., similar to UCu₄Pd. However, in contrast with the behavior of the Pd material and similarly to the more conventional spin glass AgMn, UCu₄Ni follows \((t/H)\) scaling only above applied fields of order 100 Oe. Unlike UCu₄Pd which has been discussed extensively as partially ordered, disorder ought to be randomly distributed in UCu₄Ni.

1. Introduction

We found previously by muon spin relaxation (µSR) experiments that UCu₄Pd shows glassy dynamics but no finite spin-freezing point, we called it: a quantum spin glass (QSG) [1]. The main attributes of this type of material behavior are (1) the lack of evidence for a phase transition or crossover into a spin-frozen state, which would be easily revealed by a noticeable change in the value of the muon-spin relaxation rate in a longitudinal field, as well as (2) a time/field scaling law in which the muon-asymmetry decay curves follow a functional scaling form of the type \((t/H)\), with \(\gamma\) an exponent related to the type of spin autocorrelation function of the underlying spin distributions [2].
The experiments reported here concern UCu$_4$Ni, a similar material to UCu$_4$Pd in the sense that both are obtained from the dilution of Cu in the parent compound UCu$_5$, but different in the sense that the Ni ions are substantially smaller than Pd and Cu ions and, therefore, are expected to sit with equal probability in the two available sites of Cu in the crystal structure of UCu$_5$ [3]. It has been thought for a long time that Pd ions sit preferentially in one of the two inequivalent Cu-sites of this structure and tend to produce an ordered compound in UCu$_4$Pd [4] (see also e.g., references [1–3] and references therein). However, more recent work on UCu$_{5-x}$M$_x$ (M = Ni, Ag) by Chaboy et al. [5] seems to suggest that Pd ions too are randomly distributed. This group reported an analysis of x-ray data that is consistent with Ni/Cu site interchange without any preferential site assignments for Ni or Cu, as one could expect. However, this was also the case for Ag, which like Pd, was thought due to its larger ionic radius to occupy only the bigger site in the UCu$_5$ structure. The question of whether UCu$_4$Ni presents quantum spin glass dynamics similar to UCu$_4$Pd is therefore very timely.

We report here that UCu$_4$Ni does indeed display quantum spin glass dynamics below 1 K. On the other hand, we also find that the Ni and Pd materials differ in their behaviour under applied longitudinal fields. In fact the Ni material resembles more a normal spin glass (i.e., AgMn) in terms of the time/field scaling range than it does the Pd system. These findings reveal differences in the density of spin fluctuations of the two QSGs which will require further experimentation to understand.

2. Zero Field Measurements

The muon spin depolarization in UCu$_4$Ni under zero-field conditions is effected by two mechanisms; the temperature-independent static magnetism of the Cu nuclei (Kubo-Toyabe, or K-T) and the dynamic spin fluctuations of the U moments. We therefore, fit the asymmetry decay curves to the product of a K-T function [6] and a stretched exponential exp\{-$(\Lambda t)^K$\}. The latter is a convenient way to parameterize the decay function by a single time constant even for cases where there is a distribution of relaxation rates [the uniform cases of a clear exponential (K = 1) or Gaussian (K = 2) are, of course, allowed in the fit].

The temperature-independent K-T root-mean-squared value of the relaxation was obtained from the asymmetry at $T > 1$ K where the value of $\Lambda$ becomes negligible and kept constant for the lower temperature fits at 0.23 $\mu$s$^{-1}$. The remaining two parameters ($\Lambda$ and $K$) as functions of temperature are plotted in Figure 1; $\Lambda$ (triangles) and $K$ (circles). The lack of strong enhancement of $\Lambda$ at lower temperatures and the range of $K$ (i.e., $0.5 < K < 0.8$) for all temperatures suggest a distribution of relaxation rates without a crossover or transition into a spin-frozen state down to 20 mK. We have already reported a distribution of static rates at higher temperatures (300 to 2.5 K) from $\mu$SR experiments in transverse fields [7]; here we extend the temperature range in which an inhomogeneous magnetic environment persists down to 20 mK.

![Figure 1](image-url)  
**Figure 1:** Parameters obtained from fitting the zero-field muon decay asymmetry to a Kubo-Toyabe (K-T) function [6] times exp\{-$(\Lambda t)^K$\}. Triangles: relaxation rate $\Lambda$. Circles: Exponent $K$. The K-T rate was kept at 0.23 $\mu$s$^{-1}$ for all temperatures.
3. Longitudinal Field Measurements

In Figure 2(a) we show the “raw” asymmetry data at 20 mK in UCu$_4$Ni for a set of applied magnetic fields as indicated. To check for QSG dynamics, we show in Figure 2(b) the results of plotting the same curves as functions of $t/H$, with $t$ in $\mu$s and $H$ in Oe. Notice how scaling in the form $[t/H^{0.75(5)}]$ is clearly followed for about three orders of magnitude for a range of fields between 100 Oe and 3 kOe. This is similar to AgMn alloys [7] but different from UCu$_{4-x}$Pdx (x=1, 1.5) [1,2], where the scaling starts at fields as low as 13 Oe. Notice also that the long-time part of the low-field curves continues to scale extending the scaling range to 4 orders of magnitude. Similarly the short-time regions of the curves extend the scaling at high fields (see e.g., the ~5 kOe plot). These two limiting cases correspond to $(t/H) \rightarrow 0$ and $\infty$ respectively. Although (given that the asymmetry at $t = 0$ is the maximum possible for all fields) one would expect the high field limit to continue to scale at short times as seen here for the highest available field, it is not clear why the long-time part of the curves would extend the scaling as demonstrated here. This effect, which does not seem to happen in either UCu$_{4-x}$Pdx (x=1, 1.5) [1,2] or AgMn [8], requires further investigation.

4. Discussion/Conclusion

In UCu$_{5-x}$Pdx (x = 1 and 1.5) the asymmetry decay curves can be fit to a stretched exponential function and one finds that the value of $K$ is independent of field. The materials are also found to present $(t/H)$ scaling starting at fields as low as 13 Oe. Using the general character of a stretched exponential
function: \( \exp\{ -[At]^K \} \) as a guide, we see for the Ni case that qualitatively the low-field curves in Figure 2(a) seem consistent with \( K \approx 1 \). (We actually know from Figure 1 that \( K \approx 0.8 \) in zero field, but here the Kubo-Toyabe part of the relaxation, which has not been extracted, makes the curve closer to exponential. Recall also that for fields of 10 Oe or above the K-T term is negligible, “decoupled” from the relaxation by the applied field [1,2,6].) As the field increases however, positive curvature develops indicating a deviation from exponential form and a change in the value of \( K \), such that \( K \) would tend to become smaller. This seems to happen for field values up to those were the full \( (t/H) \) scaling begins. For field scaling to hold in a stretched exponential scenario, \( K \) would become independent of field at the scaling field threshold. Thus we expect \( K \) to be field independent above 100 Oe. In an actual fit of the raw data in Figure 2(a) to a stretched exponential function, the values of \( K \) and \( \Lambda \) start to drop with increasing field. Above 100 Oe or so \( K \) and \( \Lambda \) become increasingly correlated in the fit and their values more uncertain. We therefore only describe the graphs qualitatively and reserve the quantitative study of the shape of the asymmetry curves as functions of field for a future report. Qualitatively, we find some similarities and differences between the Ni and Pd systems. The glassy dynamics and lack of spin freezing allow one to classify both as quantum spin glasses. There is on the other hand a difference in the field dependences of the asymmetry curves and the time/field scaling ranges. This implies that the details of the spectral density of spin fluctuations for the two materials are necessarily different. The Ni system presents a much higher threshold-field for field-scaling and resembles more the conventional spin glass AgMn in this regard. The main implication appears to be in the frequency cutoff of the power law that describes a spin glass system with \( \gamma < 1 \) [8]. In UCuPd this cutoff occurs at a lower frequency than it does in the Ni case. An interesting potential correlation comes into play if one compares the magnetic susceptibilities, i.e., the Ni system also has a higher average Kondo temperature than the Pd one [9].

The similarities between these two systems would suggest that quantum criticality is insensitive to the details of the disorder. However, based on the differences one could ask new questions: how is the time/field-scaling range related to the quantum critical behavior of these materials? Could differences in the nature of the disorder explain the observed differences? Could the frequency cutoff be related in any way to the high temperature energy scale of the spin glass? In order to get some answers to these questions more experiments are underway.

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