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μSR study of the "anti-glass" $LiHo_{0.045}Y_{0.955}F_4$

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Abstract

$LiHoF_4$ is a dipolar-coupled Ising ferromagnet with $T_c = 1.53$ K. If sufficient dilution is introduced into the system by substituting magnetic Ho atoms with nonmagnetic Y, it becomes glassy. Even though theoretical predictions state that the glassy state should persist down to the lowest Ho concentrations, it has been observed that the system runs into what has been called the "antiglass" phase.

The nature of the "antiglass" state is still not understood, and not much experimental data is available. We studied this system at the microscopic scale using μSR. Zero field and longitudinal field measurements were performed from room temperature down to 20 mK. We found that the Ho spins remain dynamic down to 20 mK with a fluctuation rate equal to 2.6 MHz in a longitudinal field of 16 G.

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1. Introduction

The Ising model of magnetism constitutes the simplest example of a system exhibiting a phase transition driven by thermal fluctuations. Similarly, Ising spins subject to a magnetic field applied perpendicular to the direction of the magnetic moments constitute one of the simplest systems possessing a quantum phase transition at zero temperature driven by quantum fluctuations. In this latter context, the insulating body-centered tetragonal $LiHo_xY_{1-x}F_4$ material is thought to be a physical realization of the so called transverse field Ising model.

The pure compound, $LiHoF_4$, has a ferromagnetic ground state with $T_c = 1.53$ K [1]. The ferromagnetic transition is dipolar in origin since the magnetic contribution of the antiferromagnetic exchange interaction is 1.8 times smaller than the dipolar one for the first nearest neighbors, and 20 times smaller for second nearest neighbors [1]. The ground state of the Ho atoms is an effective spin $\frac{1}{2}$ Ising doublet due to the crystal field, and has an energy gap of 9.4 K to the next excited energy levels. The effective magnetic moment of the Ising ground state is $7\mu_B$, and the Ising axis lies along the $\hat{c}$ direction of the tetragonal crystal structure.

By substituting magnetic Ho atoms with nonmagnetic Y, randomness is introduced into the system and the ferromagnetic transition temperature decreases linearly as Ho concentration is reduced. Around 50% doping, there is sufficient randomness in the system for a spin glass ground state to develop, with a maximum T_g of 0.15 K for 17% Ho concentration [2]. This behavior is in agreement with theoretical predictions for diluted Ising systems coupled by long range interactions [3]. These studies predict a critical dilution limit which marks the onset of glassy behavior. However, this work also predicts glassiness to be present down to the lowest doping levels of Ho. It has been shown that this point is not supported experimentally by measurements in $LiHo_xY_{1-x}F_4$.

Reich et al. [4,5] performed AC susceptibility measurements on $LiHo_{0.045}Y_{0.955}F_4$ and interpreted their results in terms of an evolution of the magnetic system towards a

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single energy barrier as it cools down. This behavior is opposite to that displayed by spin glasses where the distribution of energy barriers broadens when cooling, and motivated some authors to designate this state as an "antiglass". Other experiments on this compound have found coherent spin oscillations with lifetimes upto 10 s at $T = 70$ mK, making this system attractive for quantum computation [6]. The authors proposed that both phenomena, the "antiglass" state and the coherence oscillations, could be explained assuming that the individual spins are grouped into clusters of approximately 240 spins per cluster. These clusters would have zero net dipolar moment at $T = 0$ and be characterized by a single energy barrier. For $T > 0$, some clusters would be in an excited coherent state with a nonzero effective magnetic moment. Clusters would sense the field produced by other clusters, changing the size of their energy barrier accordingly. In this way, the distribution of energy barriers would broaden with temperature.

Despite the work done so far in $LiHo_{0.045}Y_{0.955}F_4$ the dynamical behavior of the magnetic lattice is still not clear. It would hence be desirable to study this system with a microscopic probe that couples directly to the magnetic field dynamics. We performed this using the μSR technique [7]. This technique is sensitive to the local magnetic field distribution as well as its dynamics within a very wide frequency window (from 10^4 to 10^{12} Hz). This frequency window is complementary to, and faster than, the frequencies used by Reich et al. in AC susceptibility measurements. In what follows we will present the experimental details followed by the analysis of the data.

2. Experimental details

The $LiHo_xY_{x-1}F_4$ family of compounds are used by the laser industry; therefore single crystals are commercially available. A high quality single crystal rod (8 mm diameter by 1cm in length) of $LiHo_{0.045}Y_{0.955}F_4$ was purchased from Tydex J.S. Co. The alignment of the crystal was performed using a Laue camera; the diffraction patterns obtained were clean and well resolved confirming the high quality of the crystal. We found that the axis of the cylinder coincided with the [310] crystallographic direction. Three slices 0.6 mm thick were cut perpendicular to the rod axis using a diamond saw. These slices were also split into two or three pieces to create a compact array of single crystals on the sample holder of the spectrometer.

The μSR measurements were performed at TRIUMF (Vancouver, Canada). Low temperature measurements, between 20 mK and 10 K, were performed using an Oxford Instruments dilution refrigerator on the M15 beam line. The samples were mounted on a silver sample holder using Apiezon N grease, keeping the crystallographic $\hat{c}$ axis perpendicular to the incoming beam of muons. High temperature measurements, between 2 and 300 K, were performed at the M20 beam line using a helium flow cryostat. Here the samples were mounted in a low background sample holder using silver tape, with the crystallographic $\hat{c}$ axis perpendicular to the incident muon beam. Measurements in the presence of an external longitudinal field (LF) (along the muon beam direction and, therefore, perpendicular to the crystal $\hat{c}$ axis) were performed on both temperature ranges and covered the range between 0 and 2 T.

3. Results and analysis

A plot of the corrected asymmetries in zero field (ZF) is shown on Fig. 1. At 200 K we observed the signal characteristic of an F-μ-F center. This phenomenon, in which the muon sits between two negative fluorine atoms, pulls them closer together and precesses in their nuclear dipolar magnetic field [8], is present in our system down to 15 K, below which low temperature relaxation dominates the μSR signal. This fact allowed us to determine a boundary between the low and high temperature regimes to be around 10 K. The F-μ-F data, as well as all the data presented in this article, was fit using the TRIUMF program "MSRFIT" which uses a χ^2 minimization routine. The function used for fitting the F-μ-F line shape is that proposed by Brewer et al. [8] and the typical χ^2 values obtained were around 1.17 per degree of freedom. From these fits we found that that the distance between F ions when the muon pulls them together is 2.38(1) Å (the distance between unperturbed nearest neighbor fluorine atoms is 2.6 Å).

At 10 K, the Ho moments start slowing down and the μSR signal relaxes so fast that the F-μ-F signal cannot be observed. Below 10 K the asymmetry is composed of a fast and a slow relaxing part. This shape of the depolarization function evolves continuously down to 20 mK. The considerable dilution of Ho atoms in this system and the fact that (see next paragraph) the fast front end is due to a quasi static field while the slow one is due to slow dynamics, motivated us to fit the raw asymmetries below

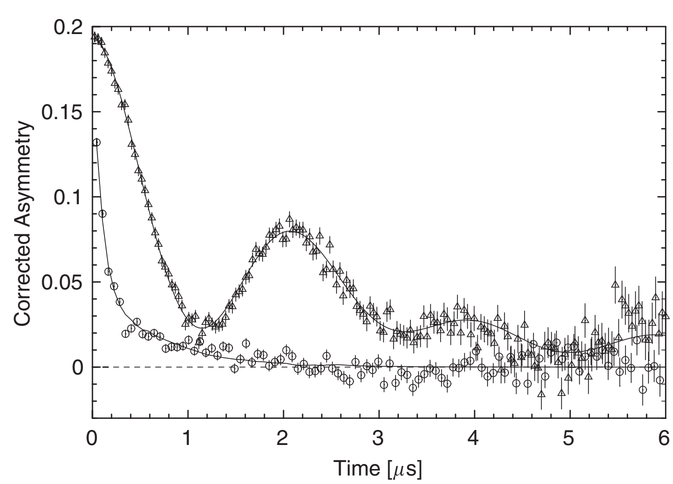

Fig. 1. μSR signals at 25 K (F-μ-F signal, in triangles) and 1 K (two component signal, in circles) in zero external field.

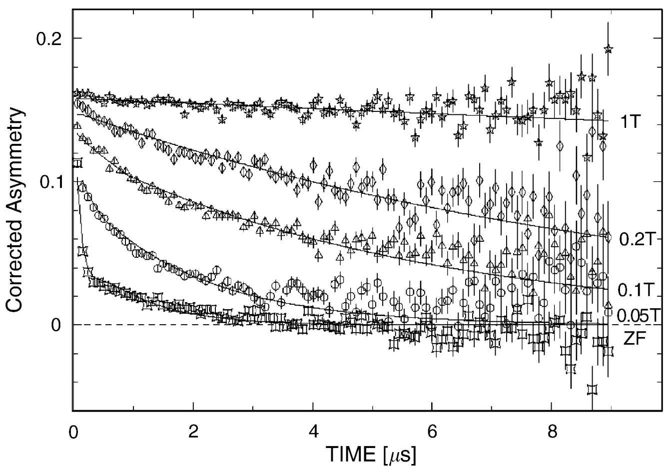

Fig. 2. Decoupling of the signal at 20 mK.

10 K to a dynamical Lorentzian Kubo-Toyabe function. The χ^2 obtained from the fitting had typical values around 1.2 per degree of freedom. Fits with other functions were tested and discarded. The fit to a dynamical Gaussian Kubo-Toyabe function produced unacceptably big χ^2 values (~4 per degree of freedom). A power exponential function was also tried without success.

Data taken in an external LF at T = 20 mK (see Fig. 2) showed that the fast front end completely decouples between 0.05 and 0.1 T while it took 2 T to fully decouple the slow one. We inferred from this observation that the fast relaxing front originates from a quasi static contribution of the magnetic field while the slow component is dynamic in origin. Fitting with a dynamical Lorentzian Kubo–Toyabe function gave good results at low fields ($\chi^2 = 1.2$ per degree of freedom at 16 G). For higher fields, the χ^2 value increased to 4.6 per degree of freedom at 200 G, and then decreased monotonically to 1.2 per degree of freedom in 2 T. This fact means that some of the physical assumptions behind the the dynamical Lorentzian Kubo–Toyabe function might not hold in fields bigger than ~100 G for this system.

4. Discussion

The results from fitting the low LF data (16 G) below 10 K are shown in Figs. 3 and 4. These figures show that the magnetic field distribution width Δ, grows from 2.81 MHz at 8.5 K up to 7.74 MHz at 0.2 K, below which it remains constant. This increase and subsequent constant behavior of Δ reflects two phenomena: first, the depopulation of the first excited crystal field levels and, second, the translation of part of the dynamic response of the system out of the μSR frequency window. This translation of the dynamic response is observed in AC susceptibility measurements [5], where the bell-shaped frequency dependence of the imaginary component shifts to lower frequencies with decreasing temperature, and moves completely out of the μSR frequency window at 200 mK.

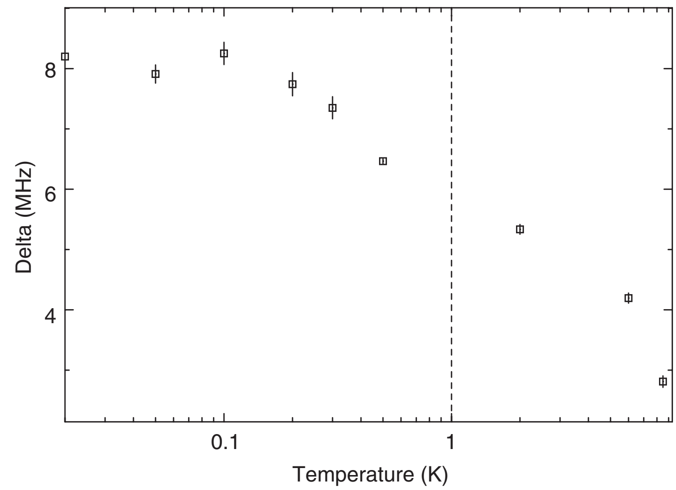

Fig. 3. Temperature dependence of Δ in a LF of 16 G. A logarithmic scale is used on the X-axis for clarity.

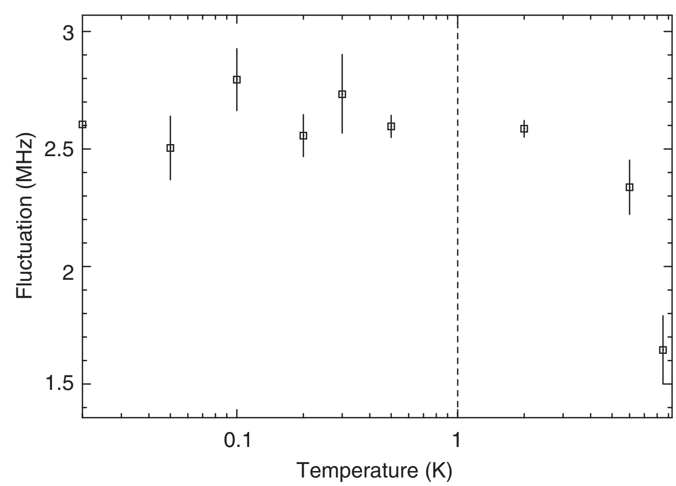

Fig. 4. Temperature dependence of the fluctuation rate in a LF of 16 G. A logarithmic scale is used on the X-axis for clarity.

Fig. 3 shows the temperature dependence of the fluctuation rate. This rate rises from 1.6 MHz at 8.5 K to 2.6 MHz at 5 K and stays constant, within error, down to base temperature. The presence of these persisting fluctuations down to base temperature is the characteristic behavior of a spin liquid ground state.

5. Conclusions

The low temperature behavior of $LiHo_{0.045}Y_{0.955}F_4$ at ZF was found to be dynamic in a frequency window that has not been accessed by previous measurements. A fluctuation rate of 2.6 MHz in a LF of 16 G is observed to be constant in the temperature range from 20 mK to 2 K. It is interesting to note that the frequency response of this system takes place over a very broad range of frequencies from 10^{-1} Hz (found in AC susceptibility experiments [6]) to 10^6 Hz (found using μSR). The temperature evolution of the magnetic field distribution width Δ has been found to track the temperature evolution of the imaginary component of the AC susceptibility [5] and with the atomic crystal field level structure.

The use of complementary probes, like μSR, which are sensitive to different timescales is clearly essential to fully explore the low temperature dynamics in this system.

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