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Study of the Ground State Properties of LiHo$_x$Y$_{1-x}$F$_4$ Using Muon Spin Relaxation

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LiHo$_x$Y$_{1-x}$F$_4$ is an insulator where the magnetic Ho$^{3+}$ ions have an Ising character and interact mainly through magnetic dipolar fields. We used the muon spin relaxation technique to study the nature of its ground state for samples with $x \leq 0.25$. In contrast with some previous works, we did not find canonical spin glass behavior down to $\sim 15$ mK. Instead, below $\sim 300$ mK we observed temperature-independent dynamic magnetism characterized by a single correlation time. The 300 mK energy scale corresponds to the Ho$^{3+}$ hyperfine interaction strength, suggesting that this interaction may be involved in the dynamic behavior of the system.

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Ising models play a central role in our understanding of magnetic systems and their phase transitions. Their importance stems from their simplicity with respect to other models, and in the fact that they reproduce many observed physical phenomena (e.g., glassiness and quantum phase transitions). For $T \ll 2$ K and $x < 1$, LiHo$_x$Y$_{1-x}$F$_4$ is thought to be a physical realization of the random transverse field Ising model with dipolar-magnetic interactions (plus a smaller nearest neighbor antiferromagnetic exchange interaction) [1,2]. For $0.25 \leq x \leq 1$ the ground state of the system is a ferromagnet [3–5], and for $x \approx 0.25$ enough randomness is introduced in the system such that long range ferromagnetic order is destroyed [5–7]. It is natural to expect that in this most diluted regime, the long ranged dipolar interactions (which can be antiferromagnetic for many bonds) together with the quenched chemical disorder produce a spin glass ground state. Surprisingly, the nature of the ground state for $x \leq 0.25$ has been the topic of a heated debate at both the experimental and theoretical levels.

Experimentally it was found that for $0.1 \leq x \leq 0.25$ the nonlinear ac susceptibility ($\chi_3$) peaks as a function of temperature, and that this peak gets rounded upon the application of an external magnetic field perpendicular to the Ising axis [7,8]. These measurements were interpreted by the authors as a transition to a low temperature spin glass state. This interpretation is supported by some numerical calculations [9,10], and the rounding of the peak in the presence of an external field was proposed to be a consequence of field-induced random fields [2,11]. Other researchers though, pointed out that a critical analysis of $\chi_3$ using only data above the peak (in equilibrium) indicates that there is no transition into a spin glass state at any finite temperature and transverse field [6,12,13], in agreement with previous numerical calculations [14–16]. At a lower doping ($x = 0.045$), one research group observed that the frequency response of the linear ac susceptibility is narrower than that of a spin glass, and this was interpreted as a splitting of the system into clusters of spins which behave as single harmonic oscillators [17]. In contrast, this narrowing was not observed in the measurements from another group which used a different sample with the same doping level. Instead, the temperature dependence of this $\chi_3$ data was shown to be compatible with that of a spin glass with a transition temperature lower than that attained in those measurements [18].

At this time there is no consensus on the nature of the ground state for $x \leq 0.25$. One of the main reasons for this is the lack of experimental data using different probes. Most data available are on magnetic susceptibility and specific heat, and data from microscopic probes is either very limited [5,19,20] or performed in extremely diluted systems where the average dipolar interaction is very weak [20,21]. We report in this Letter muon spin relaxation ($\mu$SR) [22] measurements in a series of samples which span the whole diluted regime ($x = 0.25, 0.12, 0.08, 0.045$ and 0.018).

Our samples are single crystals purchased from TYDEX J.S.Co. (St. Petersburg). Pieces from the main crystals were placed in the sample holder in such a way that the externally applied magnetic field was perpendicular to the Ising axis. The $\mu$SR measurements were performed at the M15 and M20 beam lines of TRIUMF (Canada) in the longitudinal field (LF) configuration. In this configuration the
initial muon spin direction is along the external magnetic field and therefore perpendicular to the Ising axis. For the measurements at M15, the samples were mounted on a silver sample holder using “Apiezon N” grease for thermal contact. This holder was then screwed to the mixing chamber of a dilution refrigerator. In this device the temperature of the samples was typically varied between 15 mK and 3 K, while the external field was scanned up to 0.2 T. In the M20 beam line the temperature was controlled with a helium flow cryostat in the range from 1.8 K to 100 K, and the samples were mounted using a low background sample holder.

At high temperature (>20 K) F\(\mu\)F bond formation is observed and upon cooling, the relaxation of the signal increases due to slowing down of the magnetic Ho\(^{+3}\) ions into the \(\mu\)SR time window [19,21]. The increase of the relaxation upon cooling is monotonic down to base temperature. Since some theoretical works expect to observe a glassy behavior was to be observed, this parameter should monotonically decrease upon cooling and reach a minimum at around \(T_{\text{mf}}\) or \(T_{\text{peak}}\). This fact together with the observation that no change in the shape of the signals is observed upon further cooling from \(100\) mK, indicates that these systems do not have a canonical spin glass ground state.

The low temperature data from the \(x = 0.018\) system could not be properly fit by a power exponential since the \(\mu\)SR signal develops a shoulder at low temperatures. This type of behavior indicates that fluctuations of the magnetic moments in the \(x = 0.018\) system are slow (compared with the precession rate of the muon in the average local field). In order to get quantitative information about the dynamic behavior of the system, we analyzed our data using the polarization functions derived from the stochastic dynamical Kubo-Toyabe (DKT) model (Ref. [22], page 89). This model is able to reproduce the low temperature shoulder observed in the \(x = 0.018\) system and, as it will be shown later, it accounts satisfactorily for our LF data. The value of \(\beta\) for the \(x = 0.25\) system at low temperature is approximately 1.5 (Fig. 1) which indicates that, magnetically speaking, the system is rather dense. Thus, we used Gaussian DKT functions to fit this data. On the other hand, the low temperature values of \(\beta\) for the other systems are \(\approx 0.85\), indicating that these systems are magnetically diluted. Thus, we used Lorentzian DKT functions to fit the data from the \(x = 0.12, 0.08, 0.045\) and 0.018 systems. The fitting parameters of the Lorentzian (Gaussian) model are \(a\) (\(\Delta\)) and \(\nu\), \(a\) (or \(\Delta\) in the Gaussian model) represent the typical strength of the magnetic field at the muon site (which is largely dominated by the Ho moments), while \(\nu\) is the fluctuation rate of this field [or the inverse correlation time of the local magnetic field, that is: \(\langle B(0)B(t) \rangle \approx \exp(-\nu t)\)]. We also attempted to analyze our data using other microscopic models (such as the spin glass function

\[\lambda(t) = \exp(-t^{\beta})\]

where \(\beta\) is the fluctuation rate of this field [or the inverse correlation time of the local magnetic field, that is: \(\langle B(0)B(t) \rangle \approx \exp(-\nu t)\)].

**TABLE I.** Predicted mean field freezing temperature \((T_{\text{mf}})\), position of the peak of the specific heat \((T_{\text{peak}})\) [23], characteristic strength of the internal field \((a\) or \(\Delta\)\), expressed in units of \(\mu\text{s}^{-1}\), where 135.54 \(\mu\text{s}^{-1}\) is equivalent to 1 T) and low temperature fluctuation rate of Ho\(^{+3}\) ions \((\nu_0)\), for each of the studied samples.

<table>
<thead>
<tr>
<th>(x)</th>
<th>(T_{\text{mf}}) (mK)</th>
<th>(T_{\text{peak}}) (mK)</th>
<th>(a) or (\Delta) ((\mu\text{s}^{-1}))</th>
<th>(\nu_0) ((\mu\text{s}^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.25</td>
<td>390</td>
<td>⋯</td>
<td>17.7(2)</td>
<td>15.6(8)</td>
</tr>
<tr>
<td>0.12</td>
<td>180</td>
<td>⋯</td>
<td>11.8(4)</td>
<td>20(1)</td>
</tr>
<tr>
<td>0.08</td>
<td>120</td>
<td>120</td>
<td>12.6(1)</td>
<td>20(2)</td>
</tr>
<tr>
<td>0.045</td>
<td>60</td>
<td>130</td>
<td>9.6(1)</td>
<td>10.5(1)</td>
</tr>
<tr>
<td>0.018</td>
<td>30</td>
<td>110</td>
<td>4.5(2)</td>
<td>0.73(2)</td>
</tr>
</tbody>
</table>
in Ref. [24]) but none of them produced sensible (or physical) results [25].

The fits of the low LF data using the DKT functions were performed by fixing \( a (\Delta \text{ for the } x = 0.25 \text{ system}) \) to the value found at base temperature, and then letting only the fluctuation rate \( \nu \) vary as a function of temperature. The fits with the DKT produced sensible results at the qualitative level [25]. The values of \( a (\Delta \text{ for the } x = 0.25 \text{ system}) \) are shown in Table I. These values were found to roughly follow the \( a = (24 \ \mu s^{-1}) \times \sqrt{x} \) trend expected at low values of \( x \) (using a a Ho magnetic moment of 7 \( \mu_B \)) [25].

Figure 2 shows the value of \( \nu \) as a function of temperature for all our samples. It can be seen that as the temperature is lowered the fluctuation rate of the ions decreases, and below a temperature \( T^* \) this fluctuation rate becomes temperature independent. \( T^* \) does not seem to have a clear dependence on \( x \), and has a typical value of 300 mK. We note that this temperature is approximately the same size as the hyperfine interaction energy scale of the Ho\(^{3+} \) ions (\( \approx 200 \) mK) [26]. The average value of \( \nu \) at low temperature, in the region where it is constant, is reported as \( \nu_0 \) in Table I. It can be seen that \( \nu_0 \) increases with Ho concentration until approximately 0.08, above which it levels off. It is interesting to notice that the point of the \( x - T \) phase diagram where the characteristic strength of the dipolar interaction is smaller than that of the hyperfine interaction [27] is 0.13, which is close to 0.08, the point where we observed \( \nu_0 \) to flatten.

The suitability of the DKT model to describe the low temperature behavior of LiHo\(_{0.12}\)Y\(_{1-x}\)F\(_4\) was tested with the high LF data (see Fig. 3). Using the low LF fit parameters, we only increased the field in the DKT model and the resulting functions were observed to follow satisfactorily the experimental data up to \( 0.2 \) T for the \( x = 0.25, 0.12, 0.08 \) and 0.045 samples. The LF scan in Fig. 3 clearly shows that the system is dynamic at low temperature. If the magnetic environment was static (frozen), it is expected that a LF of 0.2 T would decouple the \( \mu \)SR signal by about 85\% (corresponding to an asymmetry of 0.14 in the figure). Instead, at this field the signal relaxes much below this point, providing evidence for a dynamic environment. Furthermore, the dashed line in the figure shows that the system is not even quasistatic. The LF data also rule out a scenario where the Ho moments are static and the dynamical field comes from a smaller contribution of dynamic nuclear moments (the fact that the \( \mu \)F signal is wiped out below \( \approx 10 \) K provides evidence for the magnetic field of the Ho ions being much bigger than that from the nearest fluorine nuclei, which provide the biggest contribution to the nuclear field), since in this case the decoupling would also be faster. The fact that the \( \mu \)SR signals for LF < 0.2 T are satisfactorily described by the DKT model with a fixed value of \( \nu_0 \), indicates that the external field does not have a big effect on this parameter (as expected since the Ising levels are not coupled for LF \( \ll 2 \) T [1]).

The DKT prediction in the \( x = 0.018 \) system did not follow closely the experimental data in the intermediate LF regime (\( \approx 200 \) G). It is possible that this is due to the fact that the internal field distribution of the system is stretched along the Ising axis (= 30\% longer in the Ising direction for the \( x = 0.08 \) system [25]), instead of the spherical one that the DKT functions assume. A deformation affects the way in which the static signal is decoupled, and therefore its effect is most appreciable at this doping due to its low value of \( \nu_0 \) compared to the parameter \( a \) (see Table I).

It should be remarked that our data shows the same qualitative behavior for all the samples over the explored temperature-field space. This is in contrast with the claim

FIG. 2 (color online). Fitted values of \( \nu \) as a function of temperature and doping level. Most error bars are smaller than the point size.

FIG. 3 (color online). Corrected asymmetry for LiHo\(_{0.12}\)Y\(_{0.88}\)F\(_4\) at 12 mK and different LF. The continuous lines are fits to Lorentzian DKT functions (\( a = 12 \ \mu s^{-1}, \nu = 20 \ \mu s^{-1} \)). The partial decoupling for LF \( \approx 0.01 \) T is due to a temperature-independent background present in the zero field data. The dashed line is the expected DKT line shape in 0.2 T if the system was quasistatic (\( a = 12 \ \mu s^{-1} \) and \( \nu = 1.2 \ \mu s^{-1} \)).
that the $x = 0.045$ system has a physically different ground state from the $x = 0.167$ one [5]. We should note though that specific heat and ac susceptibility measurements of our $x = 0.045$ sample [18,23] are different from those reported in Refs. [5,17,28], opening the door to the possibility that the difference lies at the sample level. More measurements with other samples are needed to clarify this point.

It is interesting to compare our results with those from other experimental techniques. Nonlinear $\chi_{ac}$ measurements in the $x = 0.198$ and 0.167 systems show a peak at $=140$ mK and $=130$ mK, respectively [7]. These temperatures, as well as those at which the magnetic specific heat peaks (Table I), are in the range where we observe the onset of temperature-independent fluctuations, between 100 mK and 600 mK. Then, it is possible that the spin glass behavior of LiHo$_x$Y$_{1-x}$F$_4$ is associated with the slowing of the magnetic moments down to $=300$ mK, but note that the temperature-independent fluctuations at low temperature indicate that the freezing is not completed (Fig. 2). This is in agreement with the absence of a spin glass transition obtained from a critical analysis of other nonlinear $\chi_{ac}$ measurements [6,13].

It is natural to expect a spin glass ground state in LiHo$_x$Y$_{1-x}$F$_4$, since it possesses the required characteristics: frustration, introduced by the dipolar interactions, and quenched disorder from the random dilution. As shown before, the ground state of the system is dynamic, and therefore is not a classical spin glass. In this sense, the observation of a dynamic ground state in LiHo$_x$Y$_{1-x}$F$_4$ is as surprising as the observation of spin glassiness in the pyrochlore Tb$_2$Mo$_2$O$_7$ [29,30]. Since the diluted dipolar Ising model does seem to have a spin glass ground state [9], it is tempting to believe that other terms in the Hamiltonian have to be considered to account for the observed low temperature behavior. In particular, we have found that the hyperfine energy scale coincides with the onset of the temperature-independent dynamic ground state, and it might also appear in the dependence of $\nu_0$ with dilution. The importance of the hyperfine interaction for LiHo$_x$Y$_{1-x}$F$_4$ has already been pointed out in the literature [1,31], but it is not clear how this interaction can be responsible for the observed dynamical behavior since it has the effect of preventing fluctuations between the Ising levels instead of promoting them [1].

In conclusion, our $\mu$SR measurements show that the ground state of LiHo$_x$Y$_{1-x}$F$_4$ for $x \leq 0.25$ is not that of a canonical spin glass. We have observed that the low temperature state of the system can be described by the stochastic Kubo-Toyabe model (which assumes a single correlation time scale). Using this model we have determined that the fluctuation rate of the Ho magnetic moments decreases as the temperature is lowered until $=300$ mK. Below this temperature the fluctuation rate is temperature independent down to $=13$ mK. The $\mu$SR signals from all our samples exhibit the same qualitative behavior as a function of temperature and doping which stands in contrast to the observation of an additional “anti-glass” phase inferred from some $\chi_{ac}$ measurements. The hyperfine energy scale seems to manifests in the measured dynamical properties, suggesting that the hyperfine interaction may be involved in the interesting dynamic behavior of this system.

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