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Study of the Ground State Properties of $LiHo_xY_{1-x}F_4$ Using Muon Spin Relaxation

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$LiHo_xY_{1-x}F_4$ 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 ≈15 mK. Instead, below ≈ 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 $T \lesssim 2$ K and $x < 1$, $LiHo_xY_{1-x}F_4$ 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 $0.25 \lesssim x \leq 1$ the ground state of the system is a ferromagnet [3–5], and for $x \lesssim 0.25$ 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 $x \lesssim 0.25$ has been the topic of a heated debate at both the experimental and theoretical levels.

Experimentally it was found that for $0.1 \lesssim x \lesssim 0.25$ the nonlinear ac susceptibility (χ_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 χ_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 χ_{ac} 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 $x \lesssim 0.25$. 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 ether 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 (μSR) [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 μSR 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) FμF 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 μSR 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 spin glass ground state at these dilution levels [9,10], we analyzed our low LF data (<10 G) using a power-exponential fitting function: $\exp(-(\lambda t)^{\beta})$. This function has been successfully used to study μSR line shapes of spin glasses above T_g (Ref. [22], page 142).

The fit values for λ are shown in Fig. 1. We will show later that upon cooling, the increase in λ around 200 mK (most noticeable in the $x = 0.25$ data) is produced by a further slowing down of the magnetic moments of the system. This figure also shows the fit values for the power β. It can be seen that upon cooling, this parameter has a minimum at around ≈10 K, which is associated with the slowing down of the very fast fluctuating Ho moments into the μSR time window [21]. Below this temperature, β grows monotonically and it stabilizes at ≈0.85 for the $x = 0.12$, 0.08 and 0.045 systems, and at ≈1.5 for $x = 0.25$. If a glassy behavior was to be observed, this parameter should monotonically decrease upon cooling and reach a minimum of 1/3 just above the freezing temperature T_g (Ref. [22], page 142). An upper estimate for T_g in $LiHo_xY_{1-x}F_4$ can be obtained using the mean field expression: $T_{mf} \approx T_g \approx xT_c$ (T_c is the critical temperature of the ferromagnetic $x = 1$ system which is 1.54 K). These estimated temperatures are shown in Table I, together with the temperature at which the magnetic specific heat peaks [23]. Figure 1 shows that none of our samples presents a minimum of β at around T_{mf} or T_{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.

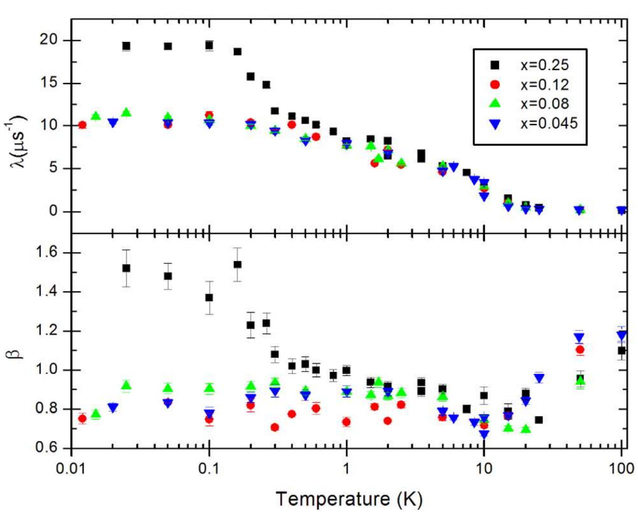

FIG. 1 (color online). Analysis of the low LF data using a power-exponential fitting function. The top panel shows the relaxation rate of the signal λ, and the lower one the exponent β.

The low temperature data from the $x = 0.018$ system could not be properly fit by a power exponential since the μSR 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 dynamical 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 β 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 β for the other systems are ≈ 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 (Δ) and ν. a (or Δ 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 ν is the fluctuation rate of this field [or the inverse correlation time of the local magnetic field, that is: $\langle B(0)B(t)\rangle \propto \exp(-\nu t)$]. We also attempted to analyze our data using other microscopic models (such as the spin glass function

TABLE I. Predicted mean field freezing temperature (T_{mf}), position of the peak of the specific heat (T_{peak}) [23], characteristic strength of the internal field (a or Δ, expressed in units of μs^{-1}, where 135.54 μs^{-1} is equivalent to 1 T) and low temperature fluctuation rate of Ho^{3+} ions (ν_0), for each of the studied samples.

x	T_{mf} (mK)	T_{peak} (mK)	a or Δ (μs^{-1})	ν_0 (μs^{-1})
0.25	390	···	17.7(2)	15.6(8)
0.12	180	···	11.8(4)	20(1)
0.08	120	120	12.6(1)	20(2)
0.045	60	130	9.6(1)	10.5(1)
0.018	30	110	4.5(2)	0.73(2)

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 (Δ for the $x = 0.25$ system) to the value found at base temperature, and then letting only the fluctuation rate ν vary as a function of temperature. The fits with the DKT produced sensible results at the qualitative level [25]. The values of a (Δ for the $x = 0.25$ 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 μ_B) [25].

Figure 2 shows the value of ν 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 ν at low temperature, in the region where it is constant, is reported as ν_0 in Table I. It can be seen that ν_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 ν_0 to flatten.

The suitability of the DKT model to describe the low temperature behavior of $LiHo_xY_{1-x}F_4$ 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 $\approx$ 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 μSR 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 FμF 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 μSR signals for LF $<$ 0.2 T are satisfactorily described by the DKT model with a fixed value of ν_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$200 G). It is possible that this is due to the fact that the internal field distribution of the system is stretched along the Ising axis ($\approx$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 ν_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

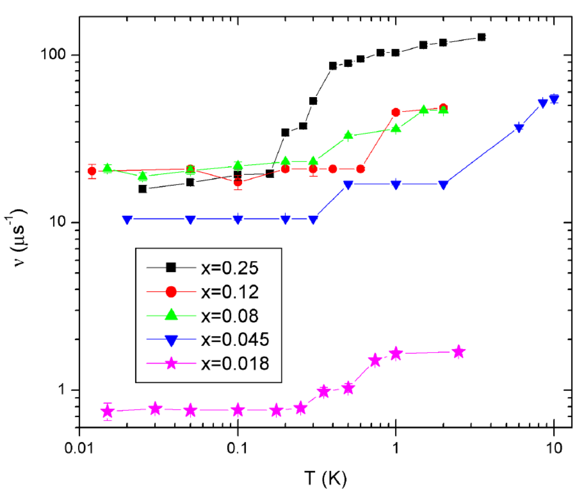

FIG. 2 (color online). Fitted values of ν as a function of temperature and doping level. Most error bars are smaller than the point size.

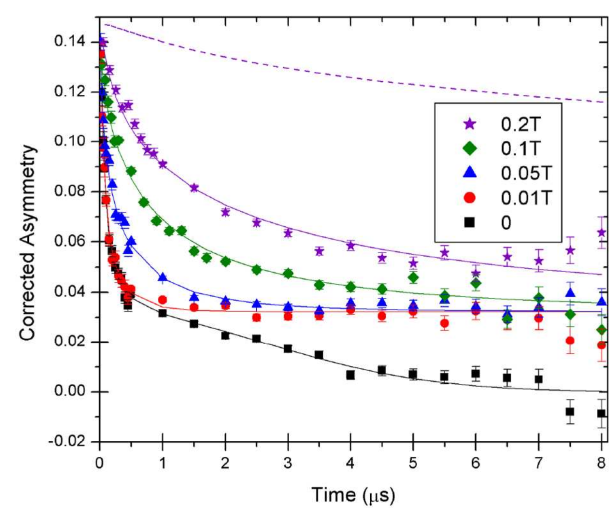

FIG. 3 (color online). Corrected asymmetry for $LiHo_{0.12}Y_{0.88}F_4$ at 12 mK and different LF. The continuous lines are fits to Lorentzian DKT functions ($a = 12\ \mu s^{-1}$, $\nu = 20\ \mu s^{-1}$). The partial decoupling for LF $\geq$ 0.01 T is due to a temperature-independent background present 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 χ_{ac} measurements in the $x = 0.198$ and 0.167 systems show a peak at $\approx$140 mK and $\approx$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 $LiHo_xY_{1-x}F_4$ is associated with the slowing of the magnetic moments down to $\approx$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 χ_{ac} measurements [6,13].

It is natural to expect a spin glass ground state in $LiHo_xY_{1-x}F_4$, 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 $LiHo_xY_{1-x}F_4$ is as surprising as the observation of spin glassiness in the pyrochlore $Tb_2Mo_2O_7$ [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 ν_0 with dilution. The importance of the hyperfine interaction for $LiHo_xY_{1-x}F_4$ 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 μSR measurements show that the ground state of $LiHo_xY_{1-x}F_4$ 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 $\approx$300 mK. Below this temperature the fluctuation rate is temperature independent down to $\approx$13 mK. The μSR 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 χ_{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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[1] M. Schechter and P. C. E. Stamp, Phys. Rev. Lett. **95**, 267208 (2005).
[2] S. M. A. Tabei *et al.*, Phys. Rev. Lett. **97**, 237203 (2006).
[3] A. H. Cooke *et al.*, J. Phys. C **8**, 4083 (1975).
[4] J. E. Battison *et al.*, J. Phys. C **8**, 4089 (1975).
[5] D. H. Reich *et al.*, Phys. Rev. B **42**, 4631 (1990).
[6] P. E. Jönsson *et al.*, Phys. Rev. Lett. **98**, 256403 (2007).
[7] C. Ancona-Torres *et al.*, Phys. Rev. Lett. **101**, 057201 (2008).
[8] Wenhao Wu *et al.*, Phys. Rev. Lett. **71**, 1919 (1993).
[9] K. -M. Tam and M. J. P. Gingras, Phys. Rev. Lett. **103**, 087202 (2009).
[10] J. J. Alonso *et al.*, Phys. Rev. B **81**, 064408 (2010).
[11] M. Schechter and N. Laflorencie, Phys. Rev. Lett. **97**, 137204 (2006).
[12] J. Mattsson, Phys. Rev. Lett. **75**, 1678 (1995).
[13] P. E. Jönsson *et al.*, arXiv:0803.1357v1.
[14] J. Snider and C. C. Yu, Phys. Rev. B **72**, 214203 (2005).
[15] A. Biltmo and P. Henelius, Phys. Rev. B **76**, 054423 (2007).
[16] A. Biltmo and P. Henelius, Phys. Rev. B **78**, 054437 (2008).
[17] S. Ghosh *et al.*, Science **296**, 2195 (2002).
[18] J. A. Quilliam *et al.*, Phys. Rev. Lett. **101**, 187204 (2008).
[19] J. Rodriguez *et al.*, Physica B (Amsterdam) **374–375**, 13 (2006).
[20] M. J. Graf *et al.*, J. Phys. Conf. Ser. **150**, 042044 (2009).
[21] M. J. Graf *et al.*, Phys. Rev. Lett. **99**, 267203 (2007).
[22] μSR *Relaxation Functions in Magnetic Materials*, edited by S. L. Lee *et al.* (Institute of Physics Publishing, Bristol, 1999).
[23] J. A. Quilliam *et al.*, Phys. Rev. Lett. **98**, 037203 (2007).
[24] Y. J. Uemura *et al.*, Phys. Rev. B **31**, 546 (1985).
[25] J. Rodriguez, Ph.D. thesis, McMaster University, 2009.
[26] R. Giraud *et al.*, Phys. Rev. Lett. **87**, 057203 (2001).
[27] The characteristic strength of the dipolar interaction becomes smaller than the hyperfine energy scale for: $(1.54\ \mathrm{K})x < 0.2$ K. Here 1.54 K is the ferromagnetic transition temperature for the $x = 1$ system, and 0.2 K is the energy scale of the hyperfine interaction.
[28] S. Ghosh *et al.*, Nature (London) **425**, 48 (2003).
[29] B. D. Gaulin *et al.*, Phys. Rev. Lett. **69**, 3244 (1992).
[30] S. R. Dunsiger *et al.*, Phys. Rev. B **54**, 9019 (1996).
[31] M. Schechter and P. C. E. Stamp, Phys. Rev. B **78**, 054438 (2008).