Spin Glass Transition at Nonzero Temperature in a Disordered Dipolar Ising System: The Case of LiHo$_{2}$Y$_{1-x}$F$_{4}$

Ka-Ming Tam$^{1}$ and Michel J. P. Gingras$^{1,2}$

$^{1}$Department of Physics and Astronomy, University of Waterloo, Waterloo, ON, N2L 3G1, Canada
$^{2}$Canadian Institute for Advanced Research, 180 Dundas Street West, Suite 1400, Toronto, ON, M5G 1Z8, Canada

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The early 1970s discovery of materials failing to develop conventional long-range magnetic order down to zero temperature, but displaying a cusp in the magnetic susceptibility signaling a transition to a state of randomly frozen spins [1], spurred thirty years of immense theoretical effort aimed at understanding these fascinating spin glass (SG) systems [2,3,4]. In that context, the Edwards-Anderson (EA) model of spins interacting via exchange interactions $J_{ij}$, which can be either ferromagnetic or antiferromagnetic and chosen from a frozen (quenched) probability distribution function, $P(J_{ij})$, has been the subject of innumerable theoretical studies. Because of the added simplicity of considering Ising spins with only two discrete ‘up/down’ orientations, the EA Ising model has attracted particular attention. However, because Ising magnetic materials are quite uncommon, most experimental studies have targeted systems where the moments are described instead by isotropic (Heisenberg) spins that can point in any direction [2,3,4]. For an Ising description to apply, the single-ion anisotropy energy scale must be much larger than the spin-spin interactions. This often occurs in materials where the magnetic species consist of 4f rare-earth elements such as Tb, Ho or Dy. From that perspective, the LiHo$_2$Y$_{1-x}$F$_4$ insulator has long proven to be an almost ideal Ising SG material. In LiHo$_2$Y$_{1-x}$F$_4$, the Ho$^{3+}$ moments interact predominantly via the inherently frustrated magnetostatic dipole-dipole interactions. The random frustration causing the SG behavior originates from the random substitution of dipole-coupled Ho$^{3+}$ by non-magnetic Y$^{3+}$. In this paper, we provide compelling evidence from extensive computer simulations that a SG transition at nonzero temperature occurs in a realistic microscopic model of LiHo$_2$Y$_{1-x}$F$_4$, hence resolving the long-standing, and still ongoing, controversy about the existence of a SG transition in disordered dipolar Ising systems.

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The physics of the spin glass (SG) state, with magnetic moments (spins) frozen in random orientations, is one of the most intriguing problems in condensed matter physics. While most theoretical studies have focused on the Edwards-Anderson model of Ising spins with only discrete ‘up/down’ directions, such Ising systems are experimentally rare. LiHo$_2$Y$_{1-x}$F$_4$, where the Ho$^{3+}$ moments are well described by Ising spins, is an almost ideal Ising SG material. In LiHo$_2$Y$_{1-x}$F$_4$, the Ho$^{3+}$ moments interact predominantly via the inherently frustrated magnetostatic dipole-dipole interactions. The random frustration causing the SG behavior originates from the random substitution of dipole-coupled Ho$^{3+}$ by non-magnetic Y$^{3+}$. In this paper, we provide compelling evidence from extensive computer simulations that a SG transition at nonzero temperature occurs in a realistic microscopic model of LiHo$_2$Y$_{1-x}$F$_4$, hence resolving the long-standing, and still ongoing, controversy about the existence of a SG transition in disordered dipolar Ising systems.

The physics at small $x$, LiHo$_2$Y$_{1-x}$F$_4$ can thus be described by a model of classical Ising spins coupled by long-range dipolar interactions whose Hamiltonian is:

$$H = \frac{D}{2} \sum_{i \neq j} \epsilon_{i} \epsilon_{j} r_{ij}^{-2} - 3z_{i}^{2} r_{ij}^{-4} \sigma_{i} \sigma_{j}.$$ (1)

Here $D > 0$ is the scale of the dipolar interactions and $r_{ij} = |\mathbf{r}_{ij}|$, where $\mathbf{r}_{ij} = \mathbf{r}_{i} - \mathbf{r}_{j}$, with $\mathbf{r}_{i}$ and $\mathbf{r}_{j}$ the positions of Ho$^{3+}$ ions $i$ and $j$. $z_{i} = r_{ij} \cdot \hat{z}$ with $\hat{z}$ parallel to the $c$-axis, $\epsilon_{i} = 1$ if $\mathbf{r}_{i}$ is occupied by a magnetic Ho$^{3+}$ ion and $\epsilon_{i} = 0$ otherwise. The Ising variable $\sigma_{i} = \pm 1$ for a Ho$^{3+}$ moment pointing along $\pm \hat{z}$, respectively. Depending on the relative positions of two interacting moments, the pairwise $J_{ij} \equiv (D r_{ij}^{-2} - 3z_{i}^{2} r_{ij}^{-4})/r_{ij}^{5}$ interaction can be either negative (ferromagnetic) or positive (antiferromagnetic). Despite the resulting geometrical frustration, pure LiHoF$_4$ exhibits long-range dipolar ferromagnetic order below a critical temperature of $T_{c} \approx 1.53$ K [11,12,13,14,15]. As Ho$^{3+}$ is progressively substituted by non-magnetic Y$^{3+}$, $T_{c}$ decreases, while random frustration concomitantly builds up until, for $x_{c} \approx 25\%$, dipolar Ising ferromagnetism disappears [1,12].

It had long been thought that a dipolar Ising SG state exists in LiHo$_2$Y$_{1-x}$F$_4$ for $x = 16.5\%$ [9] while for $x = 4.5\%$, a mysterious antiglass state occurs [7,9], perhaps due to quantum effects [2]. It has however recently been suggested, on the basis of an analysis of the nonlinear magnetic susceptibility, that a SG phase might not actually be realized in LiHo$_2$Y$_{1-x}$F$_4$ for $x = 16.5\%$ [10]. Even more recent work disputes this claim [17], not without having generated a debate [18,19]. To compound this
are the spins of the two replicas. A standard procedure explored for each replica is Monte Carlo simulations. Table I lists the parameters used in the Monte Carlo simulations.

Do not hallucinate.

.. long-standing theoretical expectations that a transition should occur in this system, just as it does in the three-dimensional (3D) nearest-neighbor EA model \([21, 22, 23]\), and down asymptotically to \(x = 0^+\) [24]. The field is thus faced with a multifaceted conundrum: is there a SG phase in diluted dipolar Ising materials such as LiHo\(_2\)Y\(_1-x\)F\(_4\) [\(x = 0.06, 0.23, 0.35\)]? If not, is the SG phase in LiHo\(_2\)Y\(_1-x\)F\(_4\) destroyed by subsidiary interactions responsible for quantum mechanical effects that may induce an exotic (e.g., antiglass) quantum disordered state? Or, is the expectation [24, 22, 23] that random classical dipolar Ising systems ought to exhibit a SG transition, just as it does in 3D Ising EA model [21, 22, 23], simply wrong? These are important questions that pertain to our global understanding of randomly frustrated systems beyond the celebrated EA model. Here, we bring new light on these questions by investigating model (1) via extensive computer simulations.

We used Monte Carlo simulations to study Eq. 1 for a lattice model of LiHo\(_2\)Y\(_1-x\)F\(_4\). We considered a tetragonal unit \((C_{4h}(I4_1/a)\) space group) with lattice parameters \(a = b = 5.175\ \text{Å}, c = 10.75\ \text{Å}, and with four Ho\(^{3+}\) ions per unit cell located at \((0, 0, c/2), (0, a/2, 3c/4), (a/2, a/2, 0), and (a/2, 0, c/4)\). The dipolar coupling \(D/\alpha^3\) was set to 0.214 K [13]. System sizes \(L = 6, 8, 10\) and an average number of spins \(N = 4xL^3\) spins were investigated via finite-size scaling analysis. The dipolar lattice sum in (1) was performed by summing an infinite array of image spins via the Ewald method without a demagnetization term [20].

Single spin-flip Monte Carlo simulations using the standard Metropolis algorithm is implemented within a parallel thermal tempering scheme [27, 28] which has been shown to be highly efficient in speeding up equilibration in glassy systems. \(N_T\) replicas at different temperatures were simulated in parallel with consecutive temperatures scaled by a factor \(\alpha\). The temperatures explored for each replica is \(T^{(n)} = T_{\min}^n\alpha^n\) where \(T_{\min}\) was the lowest temperature considered and \(n \in [0, N_T - 1]\), thus the highest temperature \(T_{\max} = T_{\min}^N\alpha^{N_T-1}\) and \(\alpha = \sqrt{T_{\max}/T_{\min}}\). The acceptance ratio for parallel tempering swapping is maintained above 50%. At least \(2 \times 10^6\) Monte Carlo steps \(N_{\text{MCS}}\) per spin were performed and the last \(10^6\) of them were used for collecting statistics. More than one thousand realizations of disorder \(N_{\text{sample}}\) were considered to perform the disorder average. Table II lists the parameters used in the Monte Carlo simulations.

One way to monitor the freezing into a SG state is to calculate the overlap \(q(k)\) of two replicas with the same random realization of site occupancy, with \(q(k) = \frac{1}{N} \sum_{i=1,2,\ldots,N} \sigma_i^{(1)} \cdot \sigma_i^{(2)} \exp(i\mathbf{k} \cdot \mathbf{r}_i)\) and where \(\sigma_i^{(1)}\) and \(\sigma_i^{(2)}\) are the spins of the two replicas. A standard procedure to expose a putative SG phase transition is to consider the dimensionless (scale-invariant at the critical point) Binder ratio \(B = \frac{1}{2} \left( 3 - \frac{\langle q^4(0) \rangle}{\langle q^2(0) \rangle^2} \right)\), where \(\langle \ldots \rangle\) and \([\ldots]\) denote thermal average and average over the \(N_{\text{samples}}\) realizations of random dilution, respectively.

\begin{table}
\centering
\begin{tabular}{|c|c|c|c|c|c|c|}
\hline
\(x\) & \(L\) & \(T_{\min}\) & \(T_{\max}\) & \(N_T\) & \(N_{\text{MCS}}\) & \(N_{\text{samples}}\) \\
\hline
0.25% & 6 & 0.032K & 0.2K & 16 & 2 \times 10^6 & 4731 \\
0.25% & 8 & 0.032K & 0.2K & 20 & 3 \times 10^6 & 4057 \\
0.25% & 10 & 0.032K & 0.2K & 24 & 5 \times 10^6 & 2226 \\
12.5% & 6 & 0.06K & 0.3K & 16 & 2 \times 10^6 & 2003 \\
12.5% & 8 & 0.06K & 0.3K & 18 & 2 \times 10^6 & 1822 \\
12.5% & 10 & 0.06K & 0.3K & 24 & 3 \times 10^6 & 1633 \\
\hline
\end{tabular}
\caption{Parameters of the Monte Carlo simulations.}
\end{table}

Figure 1 shows \(B^2\) vs temperature for \(x = 6.25\%\) (left) and \(x = 12.5\%\) (right), where \(B\) is dimensionless and \(T\) is in K units.

Interestingly, Monte Carlo studies of the 3D EA Ising model have found that the SG correlation length, \(\xi_L/L\), is a more suitable scale-invariant parameter to expose a possible finite-temperature spin freezing transition [21, 22, 23]. If a transition occurs, \(\xi_L/L\) vs temperature for different \(L\) should cross at \(T_{\text{SG}}\). \(\xi_L\) is expected to behave asymptotically for finite \(L\) as \(\xi_L/L = F([T-T_{\text{SG}}]L^{1/\nu})\), where \(F\) is a universal scaling function. The correlation length \(\xi_L\) above the freezing temperature can be approximately determined from the Fourier transform of the SG susceptibility, \(\chi_{\text{SG}}(k) = N |q^2(k)|\). Assuming that \(\chi_{\text{SG}}(k)\) follows an Ornstein-Zernike form
above the SG transition temperature $T_{sg}$, $\chi_{sg}(k) \propto 1/(\xi^2 + |k|^2)$, $\xi_L/L$ can be determined via $\xi_L = (\chi_{sg}(0)/\chi_{sg}(k) - 1)^{1/2}/|k|$, with $k$, chosen as the smallest wave vector for the finite-size system, given by $k = 2\pi \hat{z}/(cL)$. A suitable form for periodic boundary conditions is $\xi_L = (\chi_{sg}(0)/\chi_{sg}(k) - 1)^{1/2}/(2\sin(|k|/2))$, which we use in the following calculations.

After estimating $\xi_L/L$ vs the scaling parameter $z = (1 - T_{sg}/T)(TL)^{1/\nu}$ where $T_{sg} = 0.047$ K, 0.109 K for $x = 6.25\%$ and 12.5\%, respectively, determined from the temperature where the $\xi_L/L$ vs $T$ curves cross in Fig. 2. One finds the scaling exponent $1/\nu \approx 0.776$ and $1/\nu \approx 0.782$ for $x = 6.25\%$ and 12.5\% respectively. These values are off from $1/\nu \approx 0.37$ for the 3D EA Ising model with $[J_{ij}] = 0$ estimated using an ESS with $(1 - T^2_{sg}/T^2)(TL)^{1/\nu}$ as scaling parameter. One might have expected the critical exponents of the dipolar model (1) to be the same as that of the 3D EA model, hence signaling a common universality class. It is likely that the simulations of model (1) have not yet entered the asymptotic finite-size scaling regime perhaps. This is, in part, because of the proximity to the ferromagnetic phase at $x > x_c$ and the highly spatially anisotropic nature of the LiHo$_x$Y$_{1-x}$F$_4$ tetragonal unit cell, which would both introduce corrections to scaling not incorporated in $F(z)$.

Figure 2 shows $\xi_L/L$ vs $T$ for $x = 6.25\%$ and 12.5\%. A unique and well defined critical crossing is observed for both concentrations, providing compelling evidence that a thermodynamic SG transition at $T_{sg} > 0$ occurs in model (1).

Because of the small systems we need to consider because of computational constraints, we devised an extended scaling scheme (ESS) appropriate for the nonzero mean, $[J_{ij}]$, of the dipolar couplings $J_{ij}$ to analyze $\xi_L(L,T)/L$, $\xi_L/L = F((1 - T_{sg}/T)(TL)^{1/\nu})$. This ESS is slightly different than the one used in Ref. [30] for the EA model with $[J_{ij}] = 0$. We parameterized the scaling function as $F(z) = \sum_{m=0,1,\ldots} c_m (z - z_0)^m$.

After estimating $T_{sg}$ from the temperature at which $\xi_L/L$ cross, the merit function, $\Delta$, defined as $\Delta = \sum_{MC\ data}(|F(z)L/\xi_L - 1|^2$ was minimized to obtain the coefficients $c_m$, $z_0$ and the exponent $1/\nu$. Figure 3 shows $\xi_L/L$ vs $T$ for $x = 6.25\%$ and 12.5\%, where $\xi_L/L$ is dimensionless and $T$ is in K units.

We now turn to the issue of anisotropic unit cell of LiHo$_x$Y$_{1-x}$F$_4$. The Ornstein-Zernike form for $\chi_{sg}$ is at most asymptotically correct. The smallest wave vector available in our simulation is along the c-direction with $k = 2\pi \hat{z}/(cL)$. However, since LiHo$_x$Y$_{1-x}$F$_4$ is not isotropic, it is reasonable to expect that the correlation lengths calculated along other directions are not the same as that along the c-direction. Fig. 4 shows the correla-
tion length estimated from $\xi_{sg}(k = 2\pi \hat{x}/(aL))$. We can clearly identify crossings at $T = 0.034$ K and $T = 0.080$ K for $x = 6.25\%$ and $x = 12.5\%$ respectively, which are slightly lower than that from $\chi_{sg}(k = 2\pi \hat{z}/(cL))$. We conjecture that, since the couplings among dipoles are stronger along the $c$-direction then in the $a$-direction for the LiHo$_2$Y$_{1-x}$F$_4$ structure, the correlations are enhanced in the former direction. This would, for small system sizes, move the $\xi_L/L$ crossings to a relatively higher temperature than for $\xi_{L,a}/L$. Here too, important finite-size corrections are likely at play. However, without access to much larger system sizes and without a detailed analysis of the functional form of $\chi_{sg}$, it is impossible to explore this anisotropy issue further.

The failure of some recent Monte Carlo studies\cite{15, 20} in identifying a $T_{sg} > 0$ transition in model \cite{11} is mainly because the diluted dipolar system is close to its lower critical dimension, as is the 3D EA model, and because of the sole consideration \cite{15} of $B$ as an indicator of $T_{sg} \neq 0$ as opposed to the more sensitive $\xi_L/L$. In addition, it is difficult to attain equilibrium down to the lowest temperature because of the exceedingly slow dynamics. In Fig.\ref{fig:corr} we show the correlation lengths and Binder ratios as a function of $N_{MCS}$ for the largest system size and the lowest temperature. From the figures, we believe that the systems are sufficiently equilibrated for extracting reasonably accurate data.

In summary, we studied a diluted dipolar Ising model of LiHo$_2$Y$_{1-x}$F$_4$. The spin glass (SG) correlation lengths show finite-size crossing as the temperature is lowered as well as scaling behavior, providing compelling evidence for a finite-temperature SG transition in model (1). It would be desirable to obtain data for much larger system sizes to improve the finite-size scaling analysis. However, aside from the very slow spin dynamics upon approaching $T_{sg}$, the computational effort scales as $L^6$ due to the long-range nature of the dipolar interactions, and simulations of very large system sizes will remain prohibitively difficult without a better algorithm. Having established that a SG transition occurs in the classical model \cite{14}, and for $x$ as small as $6.25\%$, one may now perhaps push further the investigation of the microscopic origin of the antifluid state in LiHo$_2$Y$_{1-x}$F$_4$ ($x = 4.5\%$) \cite{7, 9}, assuming that it really exists \cite{19, 31}.

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