Spin-Glass Transition at Nonzero Temperature in a Disordered Dipolar Ising System: The Case of $LiHo_xY_{1-x}F_4$

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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. In $LiHo_xY_{1-x}F_4$, the Ho^{3+} moments, which are well described by Ising spins with only discrete "up or down" directions, 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 nonmagnetic Y^{3+}. In this Letter, we provide compelling evidence from extensive computer simulations that a SG transition at nonzero temperature occurs in a realistic microscopic model of $LiHo_xY_{1-x}F_4$. This resolves the long-standing, and still ongoing, controversy about the existence of a SG transition in disordered dipolar Ising systems.

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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 30 years of immense theoretical effort aimed at understanding these fascinating spin-glass (SG) systems [2–4]. 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 or 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 [2–4]. 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 $LiHo_xY_{1-x}F_4$ insulator has long proven to be a remarkable material to explore collective magnetic phenomena [5–10], including SG behavior within an Ising setting [11,12].

Because of the compactness of the spin-carrying 4f orbitals, magnetic exchange and superexchange between Ho^{3+} ions is small in $LiHo_xY_{1-x}F_4$, and magnetostatic dipolar interactions are the predominant Ho^{3+}-Ho^{3+} couplings. Also, since the single-ion crystal field anisotropy is large compared to the magnetic interactions, the Ho^{3+} magnetic moments can be mapped onto effective Ising spins that can point only parallel or antiparallel to the c axis of the tetragonal crystalline structure of $LiHo_xY_{1-x}F_4$ [12]. Ignoring the small nearest-neighbor exchange, which does not qualitatively affect the physics at small x, $LiHo_xY_{1-x}F_4$ can thus be described by a model of classical Ising spins coupled by long-range dipolar interactions whose Hamiltonian is

$$H = \frac{D}{2}\sum_{i\neq j}\epsilon_i\epsilon_j\frac{r_{ij}^2 - 3z_{ij}^2}{r_{ij}^5}\sigma_i\sigma_j. \quad (1)$$

Here $D > 0$ is the scale of the dipolar interactions and $r_{ij} = |\boldsymbol{r}_{ij}|$, where $\boldsymbol{r}_{ij} = \boldsymbol{r}_i - \boldsymbol{r}_j$, with $\boldsymbol{r}_i$ and $\boldsymbol{r}_j$ the positions of Ho^{3+} ions i and j, respectively. $z_{ij} \equiv \boldsymbol{r}_{ij}\cdot\hat{\mathbf{z}}$ with $\hat{\mathbf{z}}$ parallel to the c axis. $\epsilon_i = 1$ if $\boldsymbol{r}_i$ 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 $\pm\hat{\mathbf{z}}$, respectively. Depending on the relative positions of two interacting moments, the pairwise $J_{ij} \equiv D(r_{ij}^2 - 3z_{ij}^2)/r_{ij}^5$ interaction can be either negative (ferromagnetic) or positive (antiferromagnetic). Despite the resulting geometric frustration, pure $LiHoF_4$ exhibits long-range dipolar ferromagnetic order below a critical temperature of $T_c \approx 1.53$ K [11–15]. As Ho^{3+} is progressively substituted by nonmagnetic Y^{3+}, T_c decreases, while random frustration concomitantly builds up until, for $x_c \approx 25\%$, dipolar Ising ferromagnetism disappears [9,15].

It had long been thought that a dipolar Ising SG state exists in $LiHo_xY_{1-x}F_4$ for $x = 16.5\%$ [9], while for $x = 4.5\%$ a mysterious antiglass state occurs [7,9], perhaps due to quantum effects [7]. 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 $LiHo_xY_{1-x}F_4$ for $x = 16.5\%$ [16]. Even more recent work disputes this claim [17], not without having generated a debate [18,19]. To compound this controversy, all recent numerical studies of diluted dipolar

Ising models fail to find a SG transition at nonzero temperature [15,20]. This is in sharp contrast with the 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 [21–23] 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 $LiHo_xY_{1-x}F_4$ [7,9,16–19]? If not, is the SG phase in $LiHo_xY_{1-x}F_4$ destroyed by subsidiary interactions responsible for quantum mechanical effects that may induce an exotic (e.g., antiglass) quantum disordered state [7]? Or is the expectation [24,25] that random classical dipolar Ising systems ought to exhibit a SG transition, just as occurs in the 3D Ising EA model [21–23], 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 $LiHo_xY_{1-x}F_4$. We considered a tetragonal unit [$C_{4h}^6(I4_1/a)$ space group] with lattice parameters $a = b = 5.175$ Å and $c = 10.75$ 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/a^3 was set to 0.214 K [15]. System sizes $La \times La \times Lc$, with $L = 6, 8, 10$, and an average number N of $N = x(4L^3)$ 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 [26].

Single spin-flip Monte Carlo simulations using the standard Metropolis algorithm were 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 of α. The temperatures explored for each replica are $T^{(n)} = T_{\min}\alpha^n$, where $T_{\min}$ was the lowest temperature considered and $n \in [0, N_T - 1]$; thus the highest temperature $T_{\max} = T_{\min}\alpha^{N_T-1}$ and $\alpha = \sqrt[N_T-1]{T_{\max}/T_{\min}}$. The acceptance ratio for parallel tempering swapping was maintained above 50%. At least 2×10^6 Monte Carlo steps (N_{MCS}) per spin were performed, and the last 10^6 of them were used for collecting statistics. More than 1000 realizations of disorder (N_{sample}) were considered to perform the disorder average. Table I lists the parameters used in the Monte Carlo simulations.

One way to monitor the freezing into a SG state is to calculate the overlap $q(\boldsymbol{k})$ of two replicas with the same random realization of site occupancy, with $q(\boldsymbol{k}) \equiv \frac{1}{N}\sum_{i=1,2,\ldots,N}\sigma_i^{(1)}\sigma_i^{(2)}\exp(i\boldsymbol{k}\cdot\boldsymbol{r}_i)$ 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 [21–23,29] $B = \frac{1}{2}(3 - \frac{[\langle q^4(0)\rangle]}{[\langle q^2(0)\rangle]^2})$, where $\langle\ldots\rangle$ and $[\ldots]$ denote thermal average and average over the N_{samples} realizations of random dilution, respectively.

TABLE I. Parameters of the Monte Carlo simulations.

x	L	$T_{\min}$	$T_{\max}$	N_T	N_{MCS}	N_{samples}
6.25%	6	0.032 K	0.2 K	16	2×10^6	4731
6.25%	8	0.032 K	0.2 K	20	3×10^6	4057
6.25%	10	0.032 K	0.2 K	24	5×10^6	2226
12.5%	6	0.06 K	0.3 K	16	2×10^6	2003
12.5%	8	0.06 K	0.3 K	18	2×10^6	1822
12.5%	10	0.06 K	0.3 K	24	3×10^6	1633

Figure 1 shows B vs temperature T for $x = 6.25\%$ and 12.5%. While B for different system sizes appear to eventually merge below a certain temperature, no clear crossing supporting a phase transition can be identified. Similar results were recently obtained [15], suggesting that no finite-temperature SG transition occurs in model (1). That said, unambiguous B crossings cannot be resolved in many Monte Carlo simulations, even for the 3D EA Ising model where a SG transition is believed to occur, though, in principle, crossing in B should be resolved when the system size is sufficiently large [21–23,29]. Hence, it is perhaps premature to conclude on the basis of results such as in Fig. 1 that a SG transition does not occur in model (1). Interestingly, Monte Carlo studies of the 3D EA Ising model have found that the SG correlation length ξ_L/L is a more suitable scale-invariant parameter to expose a possible finite-temperature spin freezing transition [21–23]. If a transition occurs, ξ_L/L vs temperature for different L should cross at T_{sg}. ξ_L is expected to behave asymptotically for finite L as $\xi_L/L = F((T - T_{\mathrm{sg}})L^{1/\nu})$, where F is a universal scaling function. The correlation length ξ_L above the freezing temperature can be approximately determined from the Fourier transform of the SG susceptibility $\chi_{\mathrm{sg}}(\boldsymbol{k}) \equiv N[\langle q^2(\boldsymbol{k})\rangle]$. Assuming that $\chi_{\mathrm{sg}}(\boldsymbol{k})$ follows an Ornstein-Zernike form above the SG transition tempera-

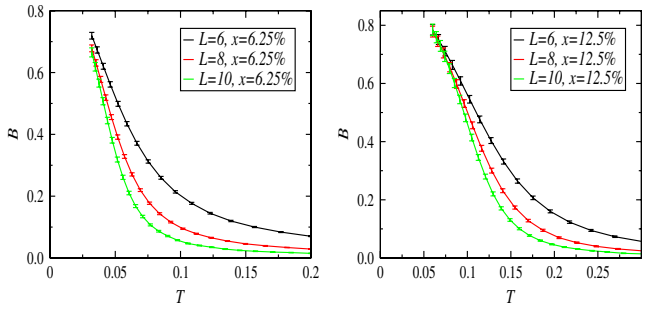

FIG. 1 (color online). Binder ratios as a function of temperature for $x = 6.25\%$ (left) and $x = 12.5\%$ (right), where B is dimensionless and T is in kelvin units.

ture T_{sg} [21–23], $\chi_{sg}(\boldsymbol{k}) \propto 1/(\xi_L^{-2} + |\boldsymbol{k}|^2)$, ξ_L/L can be determined via $\xi_L = [\chi_{sg}(0)/\chi_{sg}(\boldsymbol{k}) - 1]^{1/2}/|\boldsymbol{k}|$, with $\boldsymbol{k}$ chosen as the smallest wave vector for the finite-size system, given by $\boldsymbol{k} = 2\pi\hat{\mathbf{z}}/(cL)$. A suitable form for periodic boundary conditions is $\xi_L = [\chi_{sg}(0)/\chi_{sg}(\boldsymbol{k}) - 1]^{1/2}/[2\sin(|\boldsymbol{k}|/2)]$, which we use in the following calculations.

Figure 2 shows ξ_L/L vs T for $x = 6.25\%$ and 12.5%. A unique and well defined 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 $\xi_L(L, T)/L$: $\xi_L/L = F((1 - T_{sg}/T)(TL)^{1/\nu})$ [30]. This ESS is slightly different than the one used in Ref. [30] for the EA model with $[J_{ij}] = 0$. We parametrized the scaling function as $F(z) = \sum_{m=0,1,\ldots,4} c_m(z - z_0)^m$.

After estimating T_{sg} from the temperature at which ξ_L/L cross, the merit function Δ, defined as $\Delta = \sum_{\text{MCdata}}[F(z)L/\xi_L - 1]^2$, was minimized to obtain the coefficients c_m and z_0 and the exponent $1/\nu$. Figure 3 shows ξ_L/L vs the scaling parameter $z = (1 - T_{sg}/T)(TL)^{1/\nu}$, where $T_{sg} = 0.047$ and 0.109 K for $x = 6.25\%$ and 12.5%, respectively, determined from the temperature where the ξ_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 $(1 - T_{sg}^2/T^2)(TL)^{1/\nu}$ as a scaling parameter [30]. 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 [25]. It is likely that the simulations of model (1) have not yet entered the asymptotic finite-size scaling regime. This is, in part, because of the proximity to the ferromagnetic phase at $x > x_c$ and the highly spatially anisotropic nature of the $LiHo_xY_{1-x}F_4$ tetragonal unit cell, which would both introduce corrections to scaling not incorporated in $F(z)$.

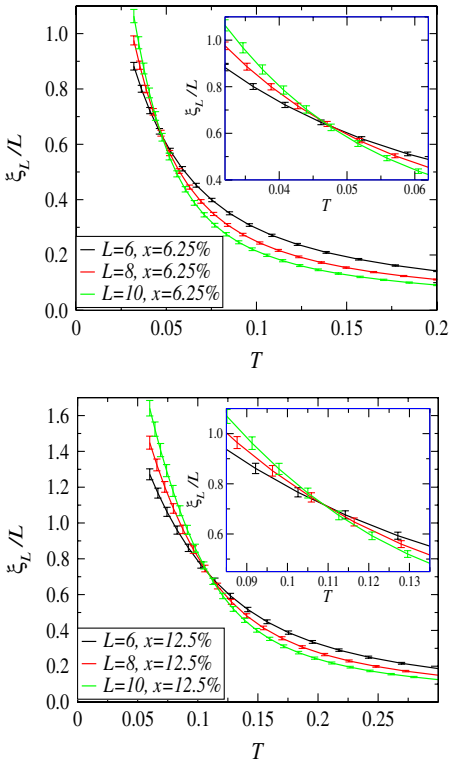

FIG. 2 (color online). Correlation lengths as a function of temperature for $x = 6.25\%$ (top) and $x = 12.5\%$ (bottom), where ξ_L/L is dimensionless and T is in kelvin units. The insets present the regions close to the crossing temperatures.

We now turn to the issue of the anisotropic unit cell of $LiHo_xY_{1-x}F_4$. The Ornstein-Zernike form for χ_{sg} is at most asymptotically correct. The smallest wave vector available in our simulation is along the c direction with $\boldsymbol{k} = 2\pi\hat{\mathbf{z}}/(cL)$. However, since $LiHo_xY_{1-x}F_4$ 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. Figure 4 shows the correlation length along the a direction, $\xi_{L,a}$, estimated from $\chi_{sg}[\boldsymbol{k} = 2\pi\hat{\mathbf{x}}/(aL)]$. 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 $\chi_{sg}[\boldsymbol{k} = 2\pi\hat{\mathbf{z}}/(cL)]$. We conjecture that, since the couplings among dipoles are stronger along the c direction than in the a direction for the $LiHo_xY_{1-x}F_4$ structure, the correlations are enhanced in the former direction. This would, for small system sizes, move the ξ_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 χ_{sg}, it is impossible to explore this anisotropy issue further.

The failure of some recent Monte Carlo studies [15,20] in identifying a $T_{sg} > 0$ transition in model (1) is mainly because the diluted dipolar system is close to its lower

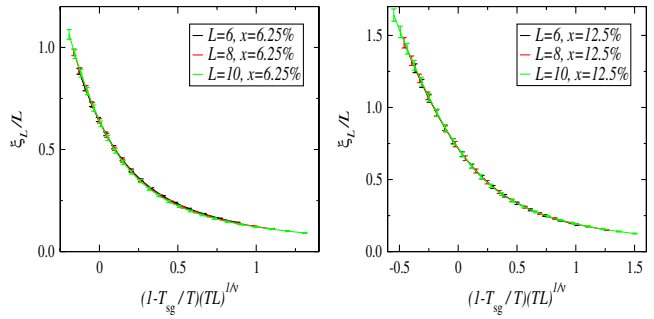

FIG. 3 (color online). Correlation lengths as a function of $(1 - T_{sg}/T)(TL)^{1/\nu}$ for $x = 6.25\%$ (left) and $x = 12.5\%$ (right), where ξ_L/L is dimensionless and T is in kelvin units.

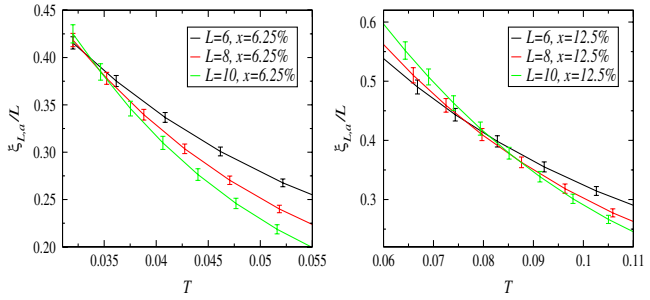

FIG. 4 (color online). Correlation lengths estimated from $\chi_{\rm sg}[2\pi\hat{\mathbf{x}}/(aL)]$, $\xi_{L,a}/L$, as a function of temperature for $x = 6.25\%$ (left) and $x = 12.5\%$ (right), where $\xi_{L,a}/L$ is dimensionless and T is in kelvin units (only data near the crossing temperatures are shown).

critical dimension, as is the 3D EA model, and because of the sole consideration [15] of B as an indicator of $T_{\rm sg} \neq 0$ as opposed to the more sensitive ξ_L/L. In addition, it is difficult to attain equilibrium down to the lowest temperature because of the exceedingly slow dynamics. In Fig. 5, we show the correlation lengths and Binder ratios as a function of $N_{\rm MCS}$ for the largest system size and the lowest temperature. From the figures, we believe that the systems considered are sufficiently equilibrated for extracting reasonably accurate data.

In summary, we studied a diluted dipolar Ising model of $LiHo_xY_{1-x}F_4$. The 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 $T_{\rm sg}$, 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

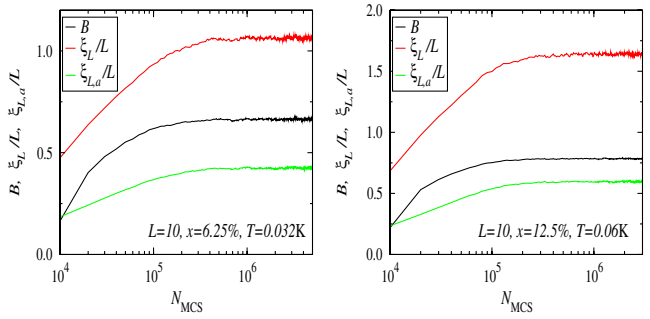

FIG. 5 (color online). Correlation lengths and Binder ratios as a function of $N_{\rm MCS}$ for $x = 6.25\%$ (left) and $x = 12.5\%$ (right) with $L = 10$ at the lowest temperatures.

occurs in the classical model (1), and for x as small as 6.25%, one may now perhaps push further the investigation of the microscopic origin of the antiglass state in $LiHo_xY_{1-x}F_4$ ($x = 4.5\%$) [7,9], assuming that it really exists [19,31].

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