

Novel Disordering Mechanism in Ferromagnetic Systems with Competing Interactions

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Ferromagnetic Ising systems with competing interactions are considered in the presence of a random field. We find that in three space dimensions the ferromagnetic phase is disordered by a random field which is considerably smaller than the typical interaction strength between the spins. This is the result of a novel disordering mechanism triggered by an underlying spin-glass phase. Calculations for the specific case of the long-range dipolar $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ compound suggest that the above mechanism is responsible for the peculiar dependence of the critical temperature on the strength of the random field and the broadening of the susceptibility peaks as temperature is decreased, as found in recent experiments by Silevitch *et al.* [Nature (London) **448**, 567 (2007)]. Our results thus emphasize the need to go beyond the standard Imry-Ma argument when studying general random-field systems.

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Introduction.—The random-field Ising model (RFIM) plays a central role in the study of disordered systems and has been applied to problems across disciplines ranging from disordered magnets to random pinning of polymers, as well as water seepage in porous media.

At and below the lower critical dimension $d_\ell = 2$, the ferromagnetic (FM) phase is unstable to an infinitesimal random field (RF) [1,2]. At higher space dimensions the disordering of the FM phase requires the RF strength h to be of the order of the spin-spin interaction strength J . Yet, the effect of the RF on the transition between the FM and paramagnetic (PM) phases—for systems with both short-range and dipolar interactions—has been the source of vast experimental and theoretical scrutiny [3–5]. Over the past three decades the RFIM has been studied experimentally via dilute antiferromagnets in a field [6], as both the RFIM and the dilute antiferromagnets in a field seem to share the same universality class. More recently it has been shown that in anisotropic dipolar magnets the RFIM can be realized in the FM phase: By applying a transverse field to a dilute dipolar ferromagnet, such as $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$, one transforms the spatial disorder to a longitudinal *effective* RF [7–9]. This opens the doors for advancing our understanding of the RF problem [10], as well as new applications, such as tunable domain-wall pinning [12] in magnetic materials.

Silevitch *et al.* recently studied the FM-to-PM transition in the presence of RFs in $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ [13]. Remarkably, they found that T_c depends linearly on the transverse field (and thus on h [7,9]) and that the susceptibility peak diminishes and broadens as temperature decreases. In $\text{Mn}_{12}\text{-ac}$, which is a realization of the RFIM with all FM interactions, a strong suppression of T_c as a function of h was found as well [14], but with what appears to be a quite different functional dependence at small h .

Here we study the interplay between FM and spin-glass (SG) phases in a dipolar Ising model with competing interactions in the presence of a RF. We find a novel disordering mechanism of the FM phase when a RF is applied and the system is in close proximity (e.g., via dilution) to a SG phase. This disordering mechanism lies between the Imry-Ma and standard disordering mechanisms: The disordering of the FM phase occurs at a finite RF, which is considerably smaller than the typical spin-spin interaction, and the disordered phase [denoted henceforth as “quasi-SG” (QSG)] consists of not FM but glassy domains. At $T = 0$ we predict the existence of a FM-to-QSG transition and determine for $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$, analytically and numerically, the phase boundary as a function of the Ho concentration x and RF strength h . At finite temperature our theory agrees with experiments [13], suggesting that the existence of competing interactions and the proximity to the SG phase dictate the broadening of the susceptibility peaks at low temperature and the peculiar dependence of T_c on h . Our theoretical analysis of the SG phase follows the scaling approach of Fisher and Huse [15]—its validity is supported by the agreement we find with our numerical results. The nature of the SG phase in a RF, however, is controversial [16–31], but of no concern here.

Theoretical analysis.—We first study $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ at $T = 0$. For dilutions $x > x_c$ the system is FM, whereas for $x_0 < x < x_c$ the system is a SG. Below we show numerically that $x_c \approx 0.3$. To date, it is unclear if $x_0 > 0$ [32,33]. For $x \approx x_c$ we define the energy per spin of the lowest FM state of the system as $f_{\text{FM}}(x)$, and the lowest energy of the SG state as $f_{\text{SG}}(x)$. Note that $f_{\text{FM}}(x)$ is the ground-state energy of the FM phase when $x > x_c$ and $f_{\text{SG}}(x)$ represents the ground-state energy of the SG phase for $x_0 < x < x_c$.

At $x = x_c$, $f_{\text{FM}}(x_c) = f_{\text{SG}}(x_c)$, and for $x \approx x_c$, to first order in $x - x_c$, $f_{\text{SG}}(x) - f_{\text{FM}}(x) = \alpha(x - x_c) + \dots$. We consider the FM phase for $x > x_c$ in an applied RF of mean zero and standard deviation h . For small h , the FM state in three dimensions cannot gain energy from the field, because domain flips are not energetically favorable. However, for spin glasses the lower critical (Imry-Ma) dimension is infinity [15]. In particular, in 3D the energy of the system can be lowered by flipping domains, creating a QSG phase with a finite correlation length. Thus, for $x \sim x_c$ the energy of the SG state will become lower than the energy of the FM state at a finite RF, which is still considerably smaller than the typical spin-spin interaction J . More generally, any 3D Ising system with competing interactions having at zero RF a FM ground state and a SG state at a somewhat higher energy will be disordered through a transition to the QSG phase at a finite RF whose magnitude depends on the proximity to the SG phase and can be much smaller than J . Because in systems like $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ the effective RFs are a result of quantum fluctuations [7,34], this phase transition is a particular case of a quantum phase transition where the quantum fluctuations of the spins are small, involving only the spin's ground and first excited states [35], but where the collective effect of all spins is strong enough to drive the transition.

The value of the critical RF can be estimated using the short-range Hamiltonian [36] in a RF $H_{\text{EA}} = -\sum_{\langle ij \rangle} J_{ij} S_i S_j + \sum_i h_i S_i$. J_{ij} represent nearest-neighbor Gaussian random bonds between spins S_i with zero mean and standard deviation J , and h_i are Gaussian RFs of average strength h [37]. The SG ground state is unstable to an infinitesimal RF, creating domains of typical size $(J/h)^{1/(3/2-\theta)}$ ($\theta \approx 0.19$) [38]. The energy reduction per spin due to the RF is thus $f(h) = h(J/h)^{(3/2-\theta)/(3-\theta)}$. The total energy reduction per spin is of the same order, because the energy cost to flip domains is much smaller. Considering now a FM system with competing interactions, e.g., $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ at $x > x_c$ where at $h = 0$ the system is FM with $f_{\text{SG}} > f_{\text{FM}}$, the critical field $h_c(x)$ can be computed from $f(h = h_c) = f_{\text{SG}} - f_{\text{FM}}$, i.e., $f(h = h_c) = \alpha(x - x_c)$. One obtains

$$h_c(x) = \alpha'(J)(x - x_c)^{(3/2-\theta)/(3-\theta)}, \quad (1)$$

where $\alpha'(J) = \alpha^{(3/2-\theta)/(3-\theta)} J^{(3/2)/(3-\theta)}$; see Fig. 1. For $h > h_c(x)$ there are finite domains within which glassy order persists. The domain size decreases with increasing field, where at $h \approx J$ the system resembles a paramagnet. As $x \rightarrow x_c$, the disordering field approaches zero. For large $x - x_c$ and $h \approx J$ there is a crossover to the standard behavior where the disordering is a result of single-spin energy minimization; i.e., the intermediate QSG regime disappears.

We now consider finite temperatures and analyze the dependence of the FM T_c (at $x > x_c$) on the effective RF.

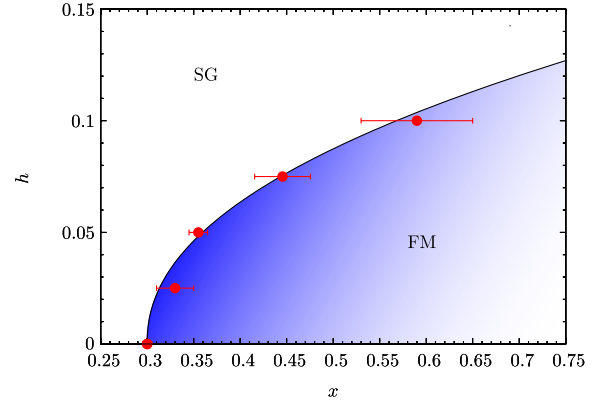


FIG. 1 (color online). Comparison of the zero-temperature numerical and analytical [Eq. (1)] h - x phase diagrams for the diluted dipolar Ising model [Eq. (2)]. $h_c(x) \sim (x - x_c)^{0.466}$, with $\theta \approx 0.19$ [38]. The analytical prediction agrees well with the numerical data (α' is a fitting parameter).

Let us denote the lowest free energies per spin of the FM phase (ordered for $T < T_{\text{FM}}$, disordered for $T > T_{\text{FM}}$) and a competing disordered QSG phase as $F_{\text{FM}}(x, T)$ and $F_{\text{QSG}}(x, T)$, respectively. Because the entropy of the QSG phase is dominated by regions at the boundaries between domains [15], the main effect of the RFs is to lower the QSG energy. Thus, $F_{\text{QSG}}(x, T) - F_{\text{FM}}(x, T) = -A(T - T_{\text{FM}}) + B(x - x_c) - h/\xi^{3/2}$ [here, for $h = 0$, $F_{\text{QSG}}(x_c, T_c) = F_{\text{FM}}(x_c, T_c)$]. For $h < h^* \equiv B(x - x_c)\xi^{3/2}$ and $T = T_{\text{FM}}$ we obtain $F_{\text{QSG}}(x, T) > F_{\text{FM}}(x, T)$, and the transition occurs between an ordered FM phase and a disordered PM phase dominated by FM fluctuations. However, for $h > h^*$ we obtain $T_c(h) - T_c(0) = A^{-1}[B(x - x_c) - h/\xi^{3/2}]$, where the FM phase is disordered by a PM phase dominated by fluctuations of domains having SG correlations over distance ξ . Thus, at $h = h^*$, $T_c(h)$ has a crossover from a roughly quadratic dependence on the RF (known for a ferromagnet in a RF [39]) to a linear dependence. This result is supported by our numerics.

Comparing to the experiments in Ref. [13], our results are consistent with $T_c(h)$ being linear when $h \ll J$, with deviations from linearity as $h \rightarrow 0$. Note that in Ref. [13] $T_c(h)$ is linear down to the lowest RFs studied if one defines T_c by the asymptotic behavior of the susceptibility at high temperatures. However, if $T_c(h)$ is defined by the peak position of the susceptibility, deviations from linearity are observed at low fields [40].

Numerical details.— $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ at low temperatures and in an external transverse magnetic field is well described by [9,41]

$$\mathcal{H} = \sum_{i \neq j} \frac{J_{ij}}{2} \epsilon_i \epsilon_j S_i S_j + \frac{J_{\text{ex}}}{2} \sum_{\langle i, j \rangle} \epsilon_i \epsilon_j S_i S_j + \sum_i h_i \epsilon_i S_i. \quad (2)$$

Here, $\epsilon_i = \{0, 1\}$ is the occupation of the magnetic Ho^{3+} ions on a tetragonal lattice (lattice constants

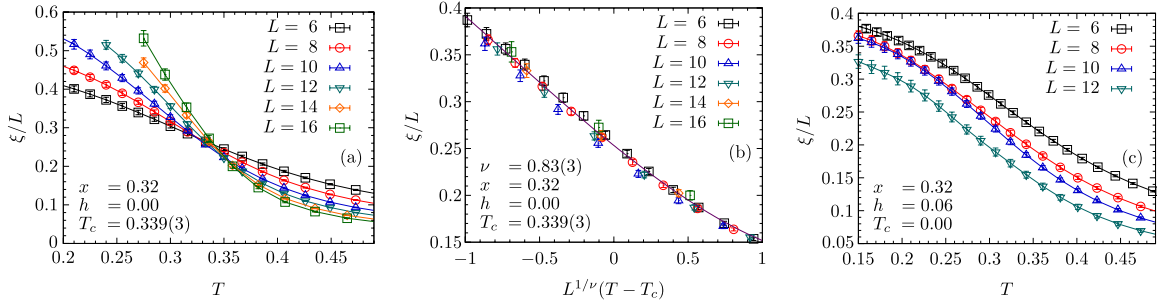


FIG. 2 (color online). Correlation length ξ_L/L as a function of T for $x = 0.32$. (a) $h = 0$. There is a clear crossing for $T_c = 0.340(3)$ [45] for different system sizes L . (b) Scaling of the data for $h = 0$. The solid line represents the optimal scaling function (polynomial approximation). (c) $h = 0.06$. There is no transition.

$a = b = 5.175 \text{ \AA}$ and $c = 10.75 \text{ \AA}$) with four ions per unit cell [42,43], $S_i \in \{\pm 1\}$, h_i represent Gaussian RFs with zero mean and standard deviation h , where h is measured in $[K]$. The magnetostatic dipolar coupling J_{ij} between two Ho^{3+} ions is given by $J_{ij} = D(r_{ij}^2 - 3z_{ij}^2)/r_{ij}^5$, where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$, \mathbf{r}_i is the position of the i th Ho^{3+} ion and $z_{ij} = (\mathbf{r}_i - \mathbf{r}_j) \cdot \hat{z}$ is the component parallel to the easy axis. $D/a^3 = 0.214 \text{ K}$ [44] and the nearest-neighbor exchange is $J_{\text{ex}} = 0.12 \text{ K}$ [42,45]. We use periodic boundary conditions with Ewald sums [43,46]. At zero field and no dilution, we find $T_c = 1.5316(2) \text{ K}$, in agreement with experimental results where $T_c = 1.530(5) \text{ K}$ [47].

For the zero-temperature simulations (Fig. 1) we use jaded extremal optimization [48,49]. Here, $\tau = 1.6, 1.8$, and 2 with an aging parameter $\Gamma = 0.05$ for at least 2^{26} steps. Ground states are found with high confidence for $L \leq 10$ and $h = 0$, and $L \leq 8$ with small $h \neq 0$. The phase boundary is identified via the Binder ratio $g = (1/2) \times (3 - [m^4]_{\text{av}}/[m^2]_{\text{av}}^2)$, where $m = (1/N) \sum_i S_i$ ($N = 4xL^3$ is the number of spins and $[\cdot]_{\text{av}}$ represents a disorder average. $g \sim \tilde{G}[L^{1/\nu}(x - x_c)]$ is a dimensionless function, allowing for the extraction of x_c and ν for a fixed h . Parameters are listed in the Supplemental Material, Table I [50].

At finite temperatures we use the parallel tempering Monte Carlo method [51]. Parameters are listed in Tables II, III, and IV in the Supplemental Material [50]. To determine the transitions for a given h and x we measure [52]

$$\xi_L = \frac{1}{2 \sin(k_{\min}/2)} \sqrt{\frac{[\langle m^2(\mathbf{0}) \rangle_T]_{\text{av}}}{[\langle m^2(\mathbf{k}_{\min}) \rangle_T]_{\text{av}}} - 1}, \quad (3)$$

where $m(\mathbf{k}) = (1/N) \sum_{i=1} S_i \exp(i\mathbf{k} \cdot \mathbf{R}_i)$. Here $\langle \cdot \cdot \rangle_T$ represents a thermal average, and \mathbf{R}_i is the spatial location of the spin S_i , and $\mathbf{k}_{\min} = (2\pi/L, 0, 0)$. $\xi_L/L \sim \tilde{X}[L^{1/\nu}(T - T_c)]$; i.e., at the transition ($T = T_c$) the argument of \tilde{X} is zero (up to scaling corrections) and hence independent of L [lines of different system sizes cross [Fig. 2(a)]]. If, however, the lines do not meet, no transition occurs [Fig. 2(c)]. To determine $T_c(h)$ we scale the data

[Fig. 2(b)]. Using a bootstrapped Levenberg-Marquardt minimization [53] allows us to determine the critical parameters with statistical errors; see Table V of the Supplemental Material [50]. Note that for a given x the critical exponent ν increases with h .

Figure 1 shows the h - x phase diagram of $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ at zero temperature. We find excellent agreement with Eq. (1), using $\theta \approx 0.19$ [38], i.e., $h_c(x) \sim (x - x_c)^{0.466}$, and α' a fitting parameter (quality of fit $Q = 0.89$) [54]. Note, however, that good fits are also possible for $0.42 \leq z \leq 0.5$ with an optimal value of $z = 0.43(4)$ ($Q = 0.82$).

Figure 3 shows finite-temperature data for different x . Figure 3(a) shows $T_c(h)$ for $x = 0.32$, i.e., $x - x_c = 0.02$ small. Our results at finite T corroborate our theoretical model with $h^* \approx 0.01$, where, for $h < h^*$, T_c is roughly independent of h (at such small fields the numerical resolution does not allow a distinction between a constant and a parabolic dependence) and for $h > h^*$, $T_c(h)$ decreases linearly. The FM phase fully disorders, at all temperatures, for $h \approx 0.055(5)$, a value slightly larger than found from the $T = 0$ simulations, yet much smaller than the interaction energy. Both the disordering of the FM phase at small fields and the linearity of $T_c(h)$ seem to persist up to $x = 0.44$ [Fig. 3(b)], the dilution used in Ref. [13], albeit with a less pronounced crossover at $h = h^*$. For $x = 0.65$ [far from the SG phase, Fig. 3(c)] the behavior of $T_c(h)$ changes to a quadratic dependence for all $h < 0.3$, suggesting a standard FM-PM transition. Critical parameters are listed in the Supplemental Material, Table V [50].

Phase diagram: Reentrance and experiment.—Our analysis for zero RF suggests that the critical concentration $x_c = 0.3$ separating the FM and SG phases depends only slightly, if at all, on temperature. Reentrance to a SG phase is either missing or limited to a small concentration regime, in contrast to previous suggestions [55].

At the same time, our results at finite RF at both zero and finite temperature for all concentrations suggest that there is a range of RFs where the system shows reentrance to a frozen QSG phase at low temperatures [56]. The RF-temperature phase diagram is shown in Fig. 3. Note also that the PM phase is characterized by distinct correlations

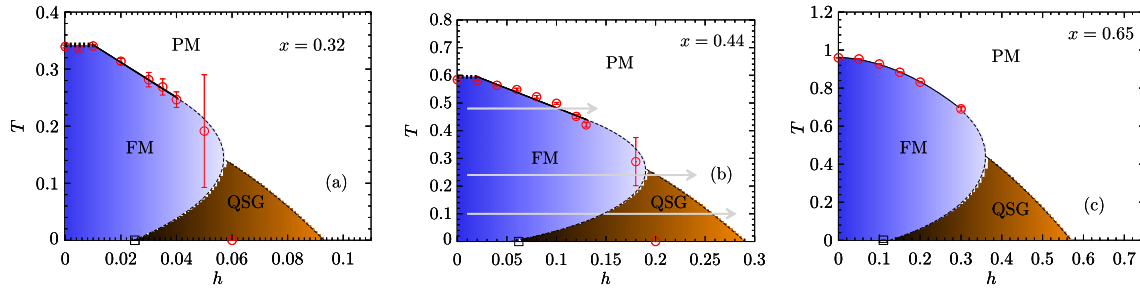


FIG. 3 (color online). T - h phase diagram of $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ for different concentrations x . Circles represent data from finite-temperature simulations, squares data from zero-temperature simulations (see Fig. 1). (a) $x = 0.32$. (b) $x = 0.44$ (dilution used in the experiments of Ref. [13]). Horizontal lines denote lines of constant temperature and varying h , as done experimentally [13] in susceptibility measurements. The top line has a direct FM-PM transition. The bottom two lines represent paths crossing the QSG phase where a broadening of the susceptibility peak and its diminishing with the enhancement of the RF at the crossover to the PM phase as temperature is decreased occur. (c) $x = 0.65$. In all panels, dotted line segments represent the conjectured phase boundary.

over the phase diagram: FM fluctuating domains close to the FM phase at $h < h^*$ [dashed line in Figs. 3(a) and 3(b)], and SG fluctuating domains close to the transition for $h > h^*$. This form of the phase diagram is strongly supported by, and provides an explanation for, the results of Ref. [13], Fig. 2. For $T > 0.3$ K [inflection point in Fig. 3(b) above] there is a direct transition from the FM to the PM [top horizontal arrow in Fig. 3(b)], as is indeed marked experimentally by a sharp cusp in the magnetic susceptibility. For $T < 0.3$ K, however, as the transverse field (and correspondingly the effective RF) is increased, the FM phase changes into a frozen QSG phase and only then to the PM phase [central horizontal arrow in Fig. 3(b)]. Experimentally, this effect is mirrored by a broad peak in the susceptibility at $T < 0.3$ K, in good agreement with the inflection point we find at $x = 0.44$. As temperature is further reduced, the crossover between the frozen QSG phase and the PM phase occurs at a larger RF [bottom horizontal arrow in Fig. 3(b)], resulting in smaller glassy domains and the experimentally observed diminishing peak of the susceptibility [7,59].

Conclusions.—We propose a novel disordering mechanism for 3D ferromagnets with competing interactions and an underlying spin-glass phase, resulting in a disordering field which is finite, yet can be much smaller than the interaction strength. We explain various aspects of the experiments of Ref. [13], including the peculiar linear dependence of T_c on the applied transverse field and the diminishing and broadening of the susceptibility peak with decreasing temperature. We further find that at smaller concentrations ($x = 0.32$, close to the spin-glass phase) the reduction of T_c with the RF becomes more pronounced. Our results strongly support the notion that it is the interplay between the competing interactions and the induced effective RF that dictate the behavior of the $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ ferromagnet at low concentrations. Our analytical results are generic to FM systems with competing interactions. It would therefore be interesting to verify these results for other types of interactions and lattice structures [60].

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