**SPIN ICE**

Dynamical fractal and anomalous noise in a clean magnetic crystal

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Fractals—objects with noninteger dimensions—occur in manifold settings and length scales in nature. In this work, we identify an emergent dynamical fractal in a disorder-free, stoichiometric, and three-dimensional magnetic crystal in thermodynamic equilibrium. The phenomenon is born from constraints on the dynamics of the magnetic monopole excitations in spin ice, which restrict them to move on the fractal. This observation explains the anomalous exponent found in magnetic noise experiments in the spin ice compound Dy$_2$Ti$_2$O$_7$, and it resolves a long-standing puzzle about its nature. In this work, we identify an emergent dynamical fractal in a disorder-free, stoichiometric, and simple topological many-body systems.

The current intense research efforts on the behavior of topological matter attempt to provide an understanding of these systems that is on the same level of both generality and detail as is available for conventional systems (1, 2). One particular frontier concerns the dynamical properties, especially those of topological systems that host exotic, fractionalized excitations, such as Laughlin quasiparticles with anyonic statistics in the quantum Hall effect (3) or emergent magnetic monopoles in the topological spin liquid known as spin ice (4) (Fig. 1). The dynamical behavior of the latter has been an enigma since its discovery (5–7). Most recently, ultrasensitive, low-temperature superconducting quantum interference device (SQUID) experiments have identified another puzzle: The magnetic noise spectral density exhibits an anomalous power law as a function of frequency, with the low-temperature exponent $\alpha = 1.5$ deviating strongly from the well-known $\alpha = 2$ of a paramagnet (8, 9) [see also (10–12)]. Within the generally successful framework of what we refer to as the standard model (SM) of spin ice dynamics (13, 14), this behavior cannot be accounted for using broadly accepted model Hamiltonian parameters (9).

In this work, we identify the missing element: Besides the constraints imposed by the emergent gauge field in spin ice, there is a further dynamical bottleneck on account of the local (transverse) field distribution, which suppresses the dynamics of another quarter of the spins (15). Both restrictions reflect the random yet correlated orientation of the spins in the spin ice ground states and force the monopoles to move on an effectively disordered cluster in real space, even in the absence of quenched disorder.

Crucially, this cluster is close to a percolation transition. It therefore exhibits a well-developed fractal structure on short and intermediate length scales, which we characterize in detail. We show that it is through hosting effectively subdiffusive monopole motion that the fractal structure bequeaths anomalous exponents to the magnetic noise. Our numerical modeling of this process allows us to quantitatively reproduce the experimental noise curves, with only a single global fitting parameter for a microscopic time scale. In the process, we also shed light onto a further, long-standing puzzle in spin ice—the steeper-than-expected rise of the macroscopic relaxation time upon cooling. Our theory explains this phenomenon naturally, in a clean (i.e., stoichiometric and uniform) system, as a reflection of the sparseness and structure of the dynamical fractal. The fractal geometry in spin ice thus influences the dynamics in a qualitative and experimentally observable way while leaving no signatures in the thermodynamics.

Spin ice is a topological magnet (4, 16) with fractionalized quasiparticles in the form of mobile magnetic monopoles (17), as illustrated in Fig. 1. The low-temperature behavior of spin ice in and out of equilibrium can largely be recast in terms of the dynamics of a dilute gas of monopoles (18) and their interactions with the background spin configuration. Specifically, the widely used SM of incoherent spin ice dynamics forbids spin flips that create—rather than hop—monopoles (Fig. 1). For a monopole in a tetrahedron, this constraint systematically blocks one direction out of four (14). This model has successfully described important features of the dynamics, especially the exponentially divergent relaxation time at low temperatures. However, it has failed to account for the large energy scale in the leading exponential growth of this time scale (18, 19), and it has particularly been challenged by susceptibility (20–22) and anomalous magnetic noise experiments (8, 9). These puzzles and their resolutions are discussed below.

The beyond the standard model (bSM) dynamics introduced here incorporate the observation that the internal field distribution on spins across which monopoles hop is peculiarly bimodal (15). In particular, one-third of the flippable spins experience a near-vanishing transverse field (23). We model these spins as flipping at a lower rate, $1/t_{\text{slow}}$. By contrast, the other spins experience a finite transverse...
field and flip at some reference rate, $1/\tau_{\text{fast}}$. (In the SM, all spins attempt to flip at a single rate, $1/\tau_0$) We take $\tau_0$, $\tau_{\text{fast}}$, and $\tau_{\text{slow}}$ to be independent of temperature. The scale $\tau_{\text{fast}}$ defines our unit of time, and it is in fact the only fitting parameter in our analysis.

Regarding the interaction parameters, we use

$$H_{\text{op}} = D \gamma \sum_{i,j} \left[ \frac{S_i \cdot S_j}{r_{ij}^2} - 3 \frac{(\langle S_i \cdot \vec{r}_{ij} \rangle)}{r_{ij}^2} \right] + J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{\langle i,j \rangle} S_i \cdot \vec{S}_j \\
+ J_3 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_4 \sum_{\langle i,j \rangle} S_i \cdot \vec{S}_j$$

an extension of the conventional dipolar spin ice Hamiltonian that was previously obtained from a combined fit to neutron scattering, magnetic susceptibility, and specific heat measurements (24). It comprises long-range dipolar interactions and first, second, and third nearest-neighbor exchange terms with strengths $D \gamma = 1.3224$ K, $J_1 = 3.41$ K, $J_2 = 0.0$ K, $J_3 = -0.00466$ K, and $J_4 = 0.04390$ K. For details, see (23), where a comparison is drawn to the maximally simple nearest-neighbor spin ice model.

Fig. 2A shows the magnetic noise measured using SQUID magnetometry on a single crystal of Dy$_2$Ti$_2$O$_7$ (9) in the temperature range 0.64 K $\leq T \leq 1.04$ K, which is high enough to be above the freezing of spin ice (7) and low enough for monopoles to be sparse, weakly interacting quasiparticles. The noise is expressed in terms of the power spectral density (PSD), $S(v)$, defined as the temporal Fourier transform of the magnetization, $M(t)$, autocorrelation function: $S(v) = \mathcal{F}[\langle M(0)M(t) \rangle]$.

Comparison with our simulations using $H_{\text{op}}$ shows that bSM dynamics reproduce experiments over four orders of magnitude in frequency and six orders of magnitude in noise power (Fig. 2). The only fitting parameter is $\tau_{\text{fast}} = 85$ µs [we estimate $\tau_{\text{slow}}/\tau_{\text{fast}} \approx 10^6$ (23), indistinguishable from $\tau_{\text{slow}} = \infty$ in these plots]. By comparison, SM dynamics with fitting parameter $\tau_0 = 200$ µs are unable to describe the experimental data at low temperature.

The extracted relaxation time from the bSM dynamics (Fig. 2B) also agrees well with the experiment (9), especially when compared with the SM, which yields much too short a relaxation time at low temperature. This has been a puzzle in the community for many years (6, 7, 13, 18, 21, 22, 25–27): In a gas of freely moving monopoles, the relaxation time of the magnetization scales with their inverse density $\rho$, $1/\rho$ $\times \exp(\Delta_m/T)$, which is set by the energy cost $\Delta_m$ of an isolated monopole. Increasing the energy in the Arrhenius law to $\Delta_v > \Delta_m$ is largely precluded by basic statistical mechanics, whereas estimates suggest that a $\Delta_v$ in excess of twice $\Delta_m$ is actually required to fit the experimental growth of the relaxation time (25). Previous theories of the steep rise of the relaxation time upon cooling invoked extrinsic contributions caused by open boundary effects, disorder, and an autonomously temperature-dependent microscopic time scale (21, 22, 28–30); the identification of an intrinsic mechanism leading to a parametrically faster growth of the relaxation time compared with $1/\rho$ has been lacking.

To explain the anomalous behavior in the magnetic noise and susceptibility and the corresponding strongly diverging relaxation time, we consider the motion of an isolated monopole in spin ice with bSM dynamics. This has between zero and three choices of sites to move to, with the statistical average being two. Linking the sites reachable by successive monopole hops, as in Fig. 3, yields a fractal cluster. Its fractal exponents are then picked up in the experimental anomalous noise signal, thus altering the relaxation properties of the system.

In more detail, to understand the fluctuations of the magnetization, we need to analyze the statistical properties of the monopole motion because (i) their motion proceeds through the flipping of spins and (ii) they are natural (sparse, weakly interacting) quasiparticles in the regime $T \leq 1$ K. The motion of the monopoles takes place on a dynamical cluster in real space, defined by excluding the spins that are not flippable energetically, or because of a small local transverse field.

For $\tau_{\text{slow}} = \infty$, this defines a percolation problem close to the critical point so that the fractal structure is visible on small and intermediate scales (Fig. 3). In this regime, the number of sites that the structure contains grows anomalously slowly with the chemical distance $n$—i.e., the minimum number of steps on the lattice needed to join two sites. We find that the growth agrees very well with $n^{1.85}$, the predicted exponent from critical percolation in $d = 3$ (32, 33) (Fig. 3), in contrast to the conventional $n^4$. The sparseness of the fractal is notable: Up to $n_{\text{sp}} = 14$, it contains only ~130 of the 2071 sites available on the diamond lattice. Around $n_{\text{sp}}$, the system crosses over to conventional three-dimensional (3D) behavior at longer length scales. Although our dynamical rules yield a nonstandard correlated percolation problem, this does not affect its critical

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**Fig. 2. Magnetization dynamics in Dy$_2$Ti$_2$O$_7$.** (A) PSD of magnetization fluctuations extracted from SQUID measurements on Dy$_2$Ti$_2$O$_7$ (filled circles) (9). Monte Carlo results for $H_{\text{op}}$ with SM dynamics (gray) and bSM dynamics (black) are shown with solid lines. The curves have been shifted vertically (23). Overall time scale factors of $\tau_{\text{fast}} = 85$ µs and $\tau_0 = 200$ µs were applied for the bSM and SM results, respectively, to visually match the experimental data, with $\tau_{\text{slow}} = \infty$ for simplicity. The deviation at high frequencies is a known feature of Monte Carlo dynamics, and other minor discrepancies are to be expected in a model with a necessarily sharper parameter distribution than the experimental system (23). (B) Relaxation times $\tau$ were extracted from fits of the PSD curves to the function $A(1 + (2\pi v)^{n_{\text{sp}}})^{-1}$ for both experimental and numerical data.
behavior, and the properties of the cluster are in fact accurately reproduced by a random walker on a random percolation cluster on the diamond lattice at filling fraction $p = 0.43$, which is only slightly above its critical value $p_c = 0.39$ (23, 34).

Our discussion so far has largely been based on the behavior of a single monopole. Beyond this, the motion of other monopoles can change the local spin and transverse field configurations, thereby endowing the percolation cluster with slow dynamics of its own (23). A higher monopole density (alongside a finite $\tau_{\text{slow}}$) therefore diminishes the crossover value $n_c$.

It is the crossover in real space that terminates the anomalous regime in the PSD toward low frequencies in the time domain. At even lower frequencies, a plateau appears on account of the trajectories of different monopoles overlapping. From the theory of random walks on percolation clusters above the percolation threshold (35–37), the PSD of a monopole should show anomalous decay $S \sim v^{-(1+\alpha)}$ at high frequency and conventional decay $S \sim v^{-2}$ at low frequencies. Exact enumeration in the random percolation problem was previously used to obtain $\alpha = 0.50 \pm 0.01$ in $d = 3$ (38). This largely explains the experimentally observed anomalous power law, which hovers near $v^{-1.85}$ in the regime under consideration (9).

From the point of view of percolation, we have discovered a purely dynamically generated fractal object in a uniform, stoichiometric, and disorder-free bulk crystal. Its existence is predicated on the emergence of point-like mobile objects—the magnetic monopoles—in a 3D topological spin liquid that remains fluctuating down to low temperatures (17). The monopole motion is subject in turn to a two-fold set of constraints. One, imposed by the emergent gauge field represented by the spin background, reflects the large-scale topological nature of the spin ice state. The other is purely microscopic in origin, resulting from an interplay of the geometric spin arrangement and the short-range statistical spin correlations imposed by the ice rules. It is the combination of these constraints that makes the monopoles move on a fractal structure. This in turn is the origin of the previously puzzling anomalous magnetic noise and rapidly diverging relaxation time. Conversely, this highly nontrivial phenomenology provides a validation of a concrete model of the microscopic dynamics, in particular the bimodal distribution of local transverse fields.

Notably, this is all accessible by probing magnetization response and fluctuations—specifically using ac susceptibility or noise measurements—whose frequency dependence reflects spatial information, linked by the (sub)-diffusive motion of the monopoles. The emergence of a dynamical fractal in a disorder-free crystal presents a distinct mechanism for the existence of anomalous noise—a subject studied in many other contexts within materials science (39–42) and elsewhere (43–46).

Some of the above elements can be manipulated in a controlled manner, all of which provide promising avenues for future experiments. Trivially, different spin ice compounds have different interaction parameters, resulting in broadly different transverse field distributions. Moreover, the microscopic dynamics can also be altered by applying uniaxial strain (47) or through the crystal field scheme by switching from (Kramers) Dy-based spin ice to, for example, (non-Kramers) Ho-based spin ice. For example, Ho$_2$Ti$_2$O$_7$ is predicted to have a smaller ratio $\tau_{\text{slow}}/\tau_{\text{fast}}$ (35).

Our effective percolation problem has turned out to be at a sweet spot near criticality—reducing constraints would eliminate the anomalous nature of the signal, whereas increasing them would likely eliminate equilibration. It will be interesting to see whether doing so (e.g., by the controlled introduction of some form of quenched or dynamical disorder) may shed light on the so-called glassy physics below 650 mK (48). Clearly the advent of highly tunable noisy intermediate-scale quantum (NISQ) platforms opens up a previously unexplored set of directions, especially in two dimensions (49, 50), where anomalous noise has already been seen in the classical nanomagnetic artificial spin ice (51). This includes questions related to quantum diffusion of monopoles and the role of increasingly coherent many-body quantum dynamics (49). The latter could alter the noise in a characteristic way (23) and therefore be used as...
evidence for the presence of so-called ring-exchange processes in candidate quantum spin ice compounds.

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**SUPPLEMENTARY MATERIALS**

science.org/doi/10.1126/science.add1644 Materials and Methods

Supplementary Text

Figs. 51 to S8

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