



Deconstructing magnetization noise: Degeneracies, phases, and mobile fractionalized excitations in tetris artificial spin ice

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Direct detection of spontaneous spin fluctuations, or "magnetization noise," is emerging as a powerful means of revealing and studying magnetic excitations in both natural and artificial frustrated magnets. Depending on the lattice and nature of the frustration, these excitations can often be described as fractionalized quasiparticles possessing an effective magnetic charge. Here, by combining ultrasensitive optical detection of thermodynamic magnetization noise with Monte Carlo simulations, we reveal emergent regimes of magnetic excitations in artificial "tetris ice." A marked increase of the intrinsic noise at certain applied magnetic fields heralds the spontaneous proliferation of fractionalized excitations, which can diffuse independently, without cost in energy, along specific quasi-1D spin chains in the tetris ice lattice.

artificial spin ice | monopoles | noise | frustrated magnetism | fractionalized excitation

While the existence of elementary magnetic monopoles remains hypothetical, spin (i.e., dipole) excitations in certain frustrated magnetic systems (1-4) can fractionalize and separate into two delocalized "monopole-like" quasiparticles that each carry an effective magnetic charge (5-12). These fractionalized excitations are topologically protected, can diffuse through the crystal lattice in thermal equilibrium, and can move in response to applied magnetic fields, motivating studies of "magnetricity" in analogy to electricity

Mobile magnetic quasiparticles have been investigated in both natural spin ice materials such as the 3D pyrochlore Dy₂Ti₂O₇ (13) and also in engineered 2D arrays of nanomagnets known as artificial spin ice (ASI) (9, 18-20). Recently, powerful detection modalities have emerged for revealing and studying these fractionalized excitations, based on direct sensing of the very small—but measurable—magnetization fluctuations that arise from their stochastic creation, annihilation, and diffusion in thermal equilibrium. In Dy₂Ti₂O₇, spontaneous magnetization noise was detected at cryogenic temperatures via SQUID (superconducting quantum interference device) magnetometry (21-23), from which the presence of monopoles and their dynamics were inferred. Separately, in archetypal square ASI lattices, magnetization noise from quasiparticle kinetics was detected at room temperature via optical magnetometry (24, 25). Crucially, in these latter studies, the sudden appearance of excess noise at certain applied magnetic fields revealed the presence of phases rich in mobile magnetic charges.

Motivated by these studies, here, we use optical magnetometry of spontaneous noise to reveal families of fractionalized excitations in the low-symmetry frustrated ASI known as "tetris ice" (26, 27). By applying small magnetic fields, tetris ice can be tuned through a variety of complex spin configurations, whose degeneracies and boundaries are directly revealed by noise. In particular, we find particularly intense and narrow bands of noise for certain directions and ranges of applied field. Using Monte Carlo simulations to deconstruct these noise signatures, these bands are shown to herald emergent regimes wherein magnetic quasiparticles delocalize and proliferate, without cost in energy, along extended quasi-1D spin chains in the lattice. These results demonstrate the power of noise-based studies to probe microscopic details of complex magnetic phenomena, in this case, specifically the equilibrium kinetics driven by fractionalized magnetic excitations.

Results

Sample. Fig. 1A shows a scanning electron microscope (SEM) image and illustration of tetris ice. The individual islands have lateral dimensions 220 × 80 nm and are made of ferromagnetic Ni_{0.8}Fe_{0.2} using established lithographic methods (27) (for details, see Materials and Methods). Each island behaves as a single Ising-like macrospin, with magnetization orientation parallel or antiparallel to its long axis. Crucially, the islands

Significance

In certain frustrated magnetic materials, flipping one spin creates a magnetic dipole excitation that can subsequently "fractionalize" and separate into two delocalized excitations that can be described as magnetic-monopole-like quasiparticles, each carrying an effective magnetic charge. These magnetic charges can diffuse through the lattice in thermal equilibrium and move in response to applied magnetic fields, motivating studies of "magnetricity," in broad analogy to electricity. Using lithographically defined arrays of nanomagnets, these results show that the presence and motion of such magnetic charges can be revealed by the intrinsic fluctuations (i.e., magnetization noise) that they generate. More generally, these studies highlight the utility of noise-based measurements to identify, in frustrated magnetic materials, regimes that are rich in mobile magnetic quasiparticles.

The authors declare no competing interest.

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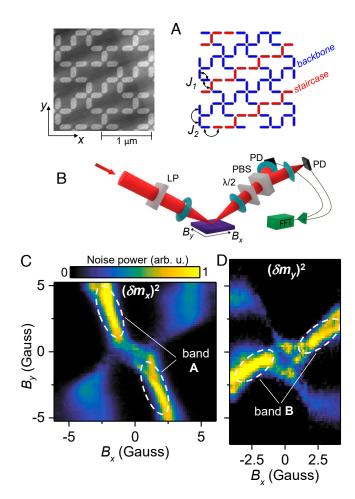


Fig. 1. (A) SEM image and diagram of tetris ice. The lattice comprises "backbones" (blue islands) and "staircases" (red islands). (B) Experiment: Magnetization fluctuations are passively detected via the polarization (Kerr rotation) fluctuations that they impart on a linearly polarized laser reflected from the ASI. (C) A map of the measured noise power from the horizontal islands, $\langle (\delta m_X)^2 \rangle$, in thermal equilibrium, vs. in-plane magnetic fields B_X and B_V . (D) Same, but for the vertical islands, $\langle (\delta m_V)^2 \rangle$. Intense noise along bands A and B herald the proliferation of fractionalized magnetic excitations.

are very thin (\approx 3.5 nm), so that at room temperature, they behave as thermally active superparamagnets, i.e., their magnetization direction thermally fluctuates in the absence of a strong biasing magnetic field (28, 29). This ensures that the ASI can efficiently explore the huge manifold of possible magnetic configurations and remain at or near its lowest energy in thermal equilibrium.

Recent studies of tetris ice demonstrated that its low-energy configuration at zero applied field (B = 0) comprises ordered "backbones" (blue islands in Fig. 1A) that include the four-fold coordinated vertices (z = 4), separated by disordered "staircases" (red islands) containing z = 3 and z = 2 vertices (27). At B = 0, the phenomenon of vertex frustration (26) prevents all the staircase vertices from achieving their lowest-energy configuration, leading to extensive degeneracy (27) and entropy-driven ordering (30, 31).

Experimental Setup. Despite the intriguing physics observed in tetris ice at B = 0, its properties have never been explored in applied magnetic fields. However, given its lower symmetry and the demonstrated emergence of complex collective behavior, a rich set of phenomena in the presence of magnetic fields can be anticipated. We investigate the field-dependent behavior of tetris ice by optically detecting its intrinsic magnetization noise, using the approach shown in Fig. 1B. This method is adapted from earlier studies of optically detected spin noise in atomic, semiconductor, and ferromagnetic systems (32–35). Here, spontaneous magnetization fluctuations in thermal equilibrium are passively detected via the Kerr rotation fluctuations that they impart on a linearly polarized laser reflected from the ASI surface. These fluctuations are detected in real time by balanced photodiodes, and the total noise power is computed via fast-Fourier transform methods. Small coils apply magnetic fields B_x and B_y in the sample plane. By reorienting the laser with respect to the sample, we measure fluctuations of either the horizontal islands, $\langle (\delta m_x)^2 \rangle$, or the vertical islands, $\langle (\delta m_y)^2 \rangle$ (see *Materials* and Methods for more details).

Experimental Results. Fig. 1 C and D show the main experimental results, which are field-dependent maps of the spontaneous magnetization noise from the horizontal and vertical islands in tetris ice. The maps reveal a complex structure, with both sharp and diffuse bands of noise appearing along certain directions and regions of B_x and B_y . The dark areas, where noise is absent, indicate regions of stable (nonfluctuating) magnetic order, which are separated by noisy boundaries. As discussed below in Fig. 2, these boundaries reveal where different magnetic configurations become energetically degenerate. Note that these maps are not related by 90° rotation, reflecting the absence of C_4 symmetry. Fig. 1D even shows an annular region of noise surrounding $B \approx 0$ (but Fig. 1C does not), which is related to fluctuations within the backbones and will be discussed later.

Of primary interest, and the main focus of this work, are the bright and narrow bands in the noise map where spontaneous fluctuations are especially strong (labeled A and B, indicated

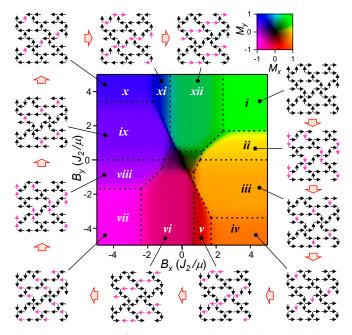


Fig. 2. Magnetic states of tetris ice, vs. B_X and B_V . The color and brightness indicate the direction and magnitude of the average magnetization M, as computed by MC simulations. Dotted lines denote boundaries between different magnetic configurations (labeled i-xii) that are stabilized when $|B_{X,Y}|$ is large. Moment configurations are shown; pink arrows indicate the subset of islands that flip when transitioning from state $i \rightarrow ii \rightarrow iii$..., clockwise around the map's perimeter. Boundaries occur when Zeeman energies balance the local interaction energies given by $J_{1,2}$. The dark central region is discussed in SI Appendix.

by dashed lines). These bands appear only for sufficiently large applied fields and follow specific diagonal trajectories in the (B_x, B_y) plane. Motivated by recent studies of archetypal square ASI, wherein the sudden onset of spontaneous noise revealed regimes of highly mobile monopoles, we therefore seek to identify the origin of these intense fluctuations in tetris ice.

Monte Carlo Simulations. Crucial insight into these complex noise maps is provided by Monte Carlo (MC) simulations of Glauber spin dynamics, following our earlier simulations of noise in square and quadrupolar ASI (24, 25). Only nearest- and next-nearest neighbor interactions were considered (J_1 and J_2 ; Fig. 1A). We used $J_1 = 1.7J_2$, and temperature $kT = 0.7J_2$ that corresponds to room temperature in our structures; both values are consistent with micromagnetic MuMax3 simulations (36) of these permalloy islands (24). These values were also validated by directly comparing measured and simulated noise maps. In particular, as discussed below, the positions of the various noise bands are determined by J_1 and J_2 , and their widths are determined in part by kT. At each value of $(B_x,$ B_{y}), the average magnetization M and the thermodynamic fluctuations $\langle [\delta M(t)]^2 \rangle$ about this mean value were determined from the computed time series (for more details, see Materials and Methods).

Fig. 2 shows a calculated map of the field-dependent magnetization, revealing a rich tableau of magnetic configurations (color and brightness indicate the direction and magnitude of **M**). Dotted black lines denote boundaries between the different static configurations that are stabilized at high field when B_x and/or B_y are large (labeled i-xii and depicted around the periphery of the map). Beginning in the upper-right corner of the map where both B_x and B_y are large and positive (region i), all horizontal and vertical islands are trivially magnetized along $+\hat{x}$ and $+\hat{y}$, respectively, by the applied field. Proceeding clockwise around the map (i.e., as $B_{\gamma} \rightarrow 0$), a subset of the vertical islands flips to $-\hat{y}$ when $B_y < J_1/\mu$, where μ is the moment of an island; that is, when the Zeeman energy balances the interaction energy and the lattice transitions to configuration ii. The pink arrows in the diagram next to region ii show which islands have flipped upon transitioning from $i \rightarrow ii$: Here, it is only the subset of vertical islands in the z = 4 vertices; these have an "unbalanced" number of horizontal neighbors. Continuing clockwise around the perimeter, from region $ii \rightarrow iii \rightarrow iv \rightarrow v$..., different subsets of islands (pink arrows) flip when crossing each boundary.

From this diagram, it is already possible to associate the experimentally measured bands of noise with boundaries between stable magnetic configurations and, more importantly, that bands A and B of exceptionally high noise occur at the internal diagonal $iii \leftrightarrow v$ boundary (or equivalently, $ix \leftrightarrow xi$), and at the diagonal $xii \leftrightarrow ii$ ($vi \leftrightarrow viii$) boundary.

This association is confirmed in Fig. 3 A and B, which show the calculated noise power from the horizontal and vertical islands $[(\delta m_x)^2]$ and $(\delta m_y)^2$, respectively. Crucially, the overall agreement with the experimental data is remarkably good (cf., Fig. 1), including capturing both narrow and diffuse bands of noise, as well as the small annulus of noise surrounding $B \approx 0$ in $(\delta m_y)^2$. Because the MC simulations are validated in this way, we have confidence that they can be explored in detail to reveal the correlated physics of the system. Most importantly, the simulations allow us to determine which subset(s) of islands give rise to the different noise signatures that are observed experimentally. In other words, we can use the validated MC simulations to deconstruct the noise maps and

tease apart the fluctuations of individual moments in ways that are experimentally intractable.

To this end, Figs. 3 C–E show the calculated noise from specific subsets of the horizontal islands (indicated by bright blue and red in the adjacent diagrams). Similarly, Figs. 3 F–H show noise from subsets of vertical islands. From these noise maps, it is clear that the horizontal and vertical bands of noise appearing at large $B_{x,y}$ originate from subsets of disconnected and uncorrelated islands that become thermally active when crossing a horizontal or vertical configuration boundary in Fig. 2.

Most importantly, these deconstructed maps reveal the origins of the intense noise that emerges in the diagonal bands A and B of the experimental data. Fig. 3 C and F, considered together, show that band A arises from fluctuations of the horizontal and vertical islands comprising the extended 1D spin chains that connect the z=4 vertices (i.e., the "spines" of the backbones). As Fig. 3I shows, it is precisely and only these connected spin chains that flip at the diagonal $iii \leftrightarrow v$ boundary (and $ix \leftrightarrow xi$, by symmetry). Along this border, configurations iii and v are energetically degenerate and the balance of Zeeman and interaction energies restores the Z2 spin symmetry of the entire chain. Similarly, Fig. 3 D and G show that noise in band B arises from the four-island chains that form the "ribs" of the backbones; only these spin chains are degenerate at the diagonal $ii \leftrightarrow xii$ (and $vi \leftrightarrow viii$) boundary (Fig. 3I).

Discussion

Fig. 4 A and B show how fluctuations at the $iii \leftrightarrow v$ boundary arise from spontaneous creation, fractionalization, and kinetics of magnetic quasiparticles along the spin chains that form the spines of the backbones. Vertices along these chains have coordination z = 2 or z = 4, and an effective magnetic charge, q, given by the balance of inward- and outward-facing moments. In region iii, all islands in the chain have stable orientation up or right, and all vertices have q = 0 (2-in, 2-out at the z = 4 vertices, and 1-in, 1-out at the z = 2 vertices). Just across the border, in region v, all islands in the chain have orientation down or left, and again, all vertices have q=0. Along the border, however, the two configurations are degenerate and Z2 symmetry of the spin chain is restored. Beginning with an ordered chain, a thermal spin-flip of any island in the chain creates a dipole excitation and a pair of adjacent vertices with $q=\pm 2$ (e.g., a z=4 vertex with 3-in, 1out and a neighboring z = 2 vertex with 0-in, 2-out). Subsequent flips of neighboring islands cause the initial dipole excitation to fractionalize into effective magnetic charges that separate and move along the 1D chain, leaving behind a string of flipped islands. Crucially, exactly along the $iii \leftrightarrow v$ border, this motion has no net cost in energy, and spin excitations can therefore readily fractionalize into quasiparticles that diffuse independently along the spin chain. The mean quasiparticle lifetime becomes very long, resulting in their rapid proliferation and formation of a regime rich in fractionalized excitations. Since the kinetics of these quasiparticles is necessarily associated with flipping of islands, this generates a telltale spike in the equilibrium noise, precisely as observed.

Similarly, Fig. 4 C and D shows that noise in band B arises from the emergence and kinetics of mobile charges along the short 4-island ribs of the backbones. Fig. 4E shows a map of the calculated density of fractionalized excitations (i.e., quasiparticles separated by more than one lattice spacing). Bands A and B of the measured noise maps therefore correspond to emergent regimes of mobile spin excitations, stabilized by the interplay between Zeeman and interaction energies.

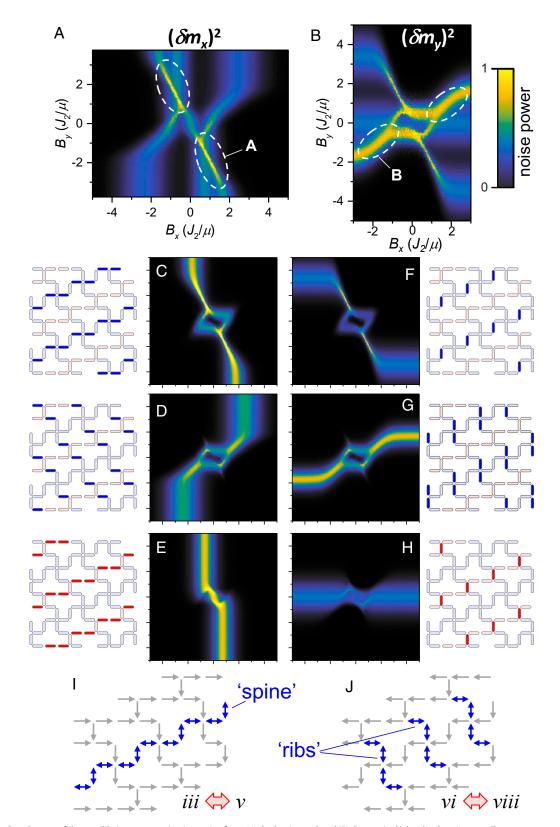


Fig. 3. Calculated maps of the equilibrium magnetization noise from (A) the horizontal and (B) the vertical islands, showing excellent agreement with experiment (cf., Fig. 1). (C-E) Deconstructing noise: Noise maps from specific subsets of horizontal islands, indicated by bright colors (blue and red denote backbone and staircase islands). (F-H) Same, but for subsets of the vertical islands. (I and J) Blue arrows indicate the 1D spin chains that flip when crossing the diagonal iii \leftrightarrow v and vi \leftrightarrow viii boundaries, respectively. These chains form the "spines" and "ribs" of the backbones. Along these boundaries, the balance of Zeeman and interaction energies restores the Z2 symmetry of the spin chains.

Finally, note that the deconstructed noise maps in Fig. 3 E and H confirm that fluctuations at B = 0 arise solely from the staircases, which at zero field are vertex-frustrated, disordered, and extensively degenerate (26, 27). In contrast, Fig. 3 C, D, F,

and G confirm that the backbones do not fluctuate at B=0, in line with the known stable type-I order of the z = 4 vertices (26, 27). However, the small annular region of noise that arises from the backbone spins reveals where the z = 4 vertices transition from

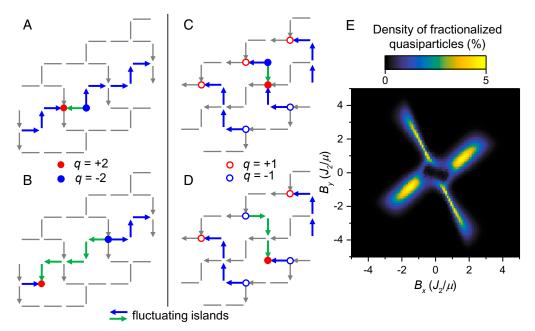


Fig. 4. (*A*) A thermal spin-flip of one island (green) in an ordered 1D "spine" generates a dipole excitation and two adjacent vertices with magnetic charge $q=\pm 2$. (*B*) Subsequent flips of neighboring islands fractionalize the excitation into two delocalized quasiparticles that move along the spine, leaving behind a string of flipped islands. For (B_X, B_Y) along the $iii \leftrightarrow v$ boundary, this motion costs no net energy, and the charges therefore diffuse independently and proliferate, leading to the excess noise observed in band A. If (B_X, B_Y) is tuned off the boundary, the ± 2 charges move preferentially in opposite directions and annihilate. (*C* and *D*) Similar phenomena along the "ribs," at the $vi \leftrightarrow viii$ boundary (band B). Note that ordered ribs end in z=3 vertices, with $q=\pm 1$; therefore excitations have $q=\mp 1$ or ± 2 . (*E*) Map of the density of fractionalized (delocalized) quasiparticles.

type-I to stable type-II order as $|B_{x,y}|$ increases (*SI Appendix*). As a final point of interest, we note that although the horizontal backbone islands in Fig. 3 C and D both generate annular regions of noise, the total noise from all horizontal islands does not (Fig. 3A). This is because, as discussed in *SI Appendix*, fluctuations of these island subsets are temporally anticorrelated, giving minimal contribution to the net $(\delta m_x)^2$, in agreement with experiment. In contrast, fluctuations of the different vertical spins in Fig. 3 F and G are temporally correlated, giving a large total $(\delta m_y)^2$ in Fig. 3B, again in agreement with experiment and further demonstrating the utility of noise decomposition in ASIs.

Summary

We have shown that tetris ice can host a rich variety of field-induced magnetic states. Comparing the configuration diagram (Fig. 2) and the experimental noise maps (Fig. 1) demonstrates that noise studies are a potent tool to directly detect and disentangle these states, where the critical boundaries are characterized by high kinetic activity. Crucially, certain noise signatures herald collective behaviors corresponding to the diffusion and proliferation of fractionalized magnetic quasiparticles along specific spin chains in the lattice. The fact that these phenomena are field-tunable may open the door to future functionalities in sensing or beyond-Turing computation (37–40). Future studies will concentrate on the detailed nature of the noise spectra and establish relations between the nontriviality of the disorder of certain phases, tied to their collective behavior, and the "color" of the noise spectrum (23, 24, 41, 42) at larger frequencies.

Materials and Methods

Sample Fabrication. Tetris lattices were fabricated by methods similar to those employed in prior work (27). Resist masks on Si/Si-N substrates were patterned by electron beam lithography for subsequent permalloy ($Ni_{0.8}Fe_{0.2}$) deposition

in ultrahigh vacuum in a molecular beam epitaxy system, followed by lift-off. The permalloy was capped with approximately 3 nm of aluminum oxide. Islands of lateral dimension 220 \times 80 nm were formed, with permalloy thickness $\sim\!\!3.5$ nm.

Optical Detection of Magnetization Noise. The tetris ASI samples were mounted face-up in the x-y plane, on a temperature-controlled micro-positioning stage. The horizontal and vertical islands were oriented along \hat{x} and \hat{y} , respectively, and two sets of small coils were used to apply magnetic fields along these directions. Following earlier noise studies of square and quadrupolar ASI (24, 25), a \sim 1 mW probe laser incident at \sim 45° in the x-z or y-z plane was linearly polarized and focused to a spot of $\sim\!4~\mu m$ on the surface of the sample. Thermodynamic magnetization fluctuations of the horizontal or vertical islands (depending on the direction of the laser incidence) imparted small Kerr rotation fluctuations on the polarization of the reflected laser, which were detected with balanced photodiodes. The signal from the photodiodes was amplified, digitized, and its power spectrum was computed in real time using fast Fourier transform (FFT) methods. The measured noise contained constant background contributions from amplifier noise and photon shot noise, which were subtracted off by also measuring the noise spectra in the presence of a large applied magnetic field ($B_X \approx B_V \approx 20$ G) where all the islands were strongly polarized and magnetization noise from the tetris ice was entirely suppressed. To obtain the maps of the frequency-integrated noise power vs. applied magnetic field (Fig. 1 C and D), for each value of the magnetic field, the noise spectrum was acquired for several seconds, which allowed us to record data in the frequency range from a few hundred Hz to a few hundred kHz.

Monte Carlo Simulations. Standard Monte Carlo (MC) simulations of Glauber spin dynamics were performed using 32×32 vertex tetris lattices with periodic boundary conditions. For each MC time step, N single islands were chosen randomly (N is the number of islands in the lattice; cluster and loop flips were not considered), and the spin flip acceptance probability was $[1 + \exp(\Delta/kT)]^{-1}$, where Δ is the energy difference associated with the spin flip and k is Boltzmann's constant. Only nearest- and next-nearest neighbor interactions were considered (J_1 and J_2), with $J_1 = 1.7J_2$, and temperature $kT = 0.7J_2$ that corresponds to room temperature in our

structures; both values are consistent with micromagnetic MuMax3 simulations (36) also validated by directly comparing measured and simulated noise maps. At each value of (B_x, B_y) , 10^6 MC annealing steps were performed to ensure thermal equilibrium, and then the magnetization was recorded for several million additional MC time steps to determine the average magnetization M and the thermodynamic fluctuations $\langle [\delta M(t)]^2 \rangle$ about this mean value.

Data, Materials, and Software Availability. All study data are included in the article and/or supporting information. Raw noise data have been deposited in Zenodo. https://zenodo.org/record/8393083 (43).

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Author contributions: M.G., C.N., C.L., P.S., and S.A.C. conceived the research; X.Z., J.R., and $J.D.W.\,fabricated\,and\,prepared\,the\,samples;\,M.G.\,performed\,the\,noise\,measurements\,and$ Monte Carlo simulations; All authors discussed the results; M.G., P.S, and S.A.C. prepared the manuscript in consultation with all authors.

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