Emergent ice rule and magnetic charge screening from vertex frustration in artificial spin ice

Ian Gilbert¹, Gia-Wei Chem², Sheng Zhang³, Liam O’Brien⁴, Bryce Fore¹, Cristiano Nisoli², and Peter Schiffer¹*

1. Department of Physics and Frederick Seitz Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA
2. Theoretical Division, and Center for Nonlinear Studies MS B258, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
3. Department of Physics and Materials Research Institute, Pennsylvania State University, University Park, PA 16802, USA
4. Department of Chemical Engineering and Materials Science, University of Minnesota, Minneapolis, Minnesota 55455, USA
5. Thin Film Magnetism Group, Department of Physics, Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, UK

* Corresponding author: Peter Schiffer, pschiffe@illinois.edu.

Simulation Details

In our Monte Carlo simulations, the magnetic state of the spin ice is described by a set of Ising variables \( \{ \sigma_i \} \); the magnetization of \( i \)-th island is then \( m_i = \sigma_i e_i \). The effective Hamiltonian in terms of the Ising spins is \( H = \sum_{i,j} J_{ij} \sigma_i \sigma_j \). Although the long-range part of the interaction is well approximated by a dipolar form \( J_{ij} \sim 1/r_{ij}^3 \), the interaction between the first few neighboring spins depends on microscopic details of the island and lattice geometry. Since the long-range tail of the dipolar interaction does not play an important role in the temperature regime of our interest, we consider a simplified model with interactions up to only a few neighbors. First, the nearest-neighbor interactions \( J_{NN} \) are characterized by the three parameters \( \alpha, \alpha', \beta \). Here \( \alpha \) and \( \beta \) are interaction energy between a pair of perpendicular and parallel short islands, while \( \alpha' \) denotes the coupling between a pair of orthogonal short and long islands. Interactions \( J_{ij} \) beyond the nearest neighbors and up to sixth-nearest-neighbors are subsumed into magnetic charge interactions of the form: \( K \sum_{\langle m,n \rangle} q_m q_n \), where \( q_m \) denotes the magnetic charge at vertex \( m \), since the main effect of interactions beyond the first-neighbor spin pairs is to produce the nontrivial charge-charge correlations observed in our experiments. The magnetic charge \( q_m \) measured in natural units is obtained by summing all the Ising spins converging in the vertex. In our simulations, we introduce two parameters: \( K_1 \) denotes the charge-charge couplings between a four-moment and a three-moment vertex, while \( K_2 \) is the coupling between a pair of nearest-
neighbor three-moment vertices. We find that reasonably good agreement with the experimental results can be achieved with $\alpha = 1.6\beta$, $\alpha' = 2.4\beta$, $K_1 = 0.2\beta$, and $K_2 = 0.1K_1$. We used the standard Metropolis algorithm, which is very efficient for the temperature regime of our interest, in our Monte Carlo simulations. For each temperature, we first thermalize the system with $10^6$ sweeps over the lattice, and each data point is averaged over $10^4$ different configurations (we use 1000 Monte Carlo sweeps to obtain independent configurations in the average). The results shown in Figures 5 and 6 in the main text and Figure S1 are obtained from simulations of a lattice containing $5 \times 30^2$ spins. However, comparison with those obtained from larger lattices shows negligible finite-size effects. In order to compare the experimental data to the simulations, we note that $kT/\beta \propto kT a^3$ (since the interaction energy $\beta$ is proportional to $1/a^3$). We can convert the effective temperature $kT/\beta$ from simulation to lattice constant $a$ by finding the correct proportionality constant $C$ such that $kT/\beta = Ca^3$. We pick $C$ such that $kT/\beta = 7.5$ corresponds to $a = 880$ nm.
neighbor three-moment vertices. We find that reasonably good agreement with the experimental results can be achieved with \( \beta = 6.1 \), \( \beta = 4.2 \), \( \beta = 2.01 \), and \( \beta = 1.0 \). We used the standard Metropolis algorithm, which is very efficient for the temperature regime of our interest, in our Monte Carlo simulations. For each temperature, we first thermalize the system with 10^6 sweeps over the lattice, and each data point is averaged over 10^4 different configurations (we use 1000 Monte Carlo sweeps to obtain independent configurations in the average). The results shown in Figures 5 and 6 in the main text and Figure S1 are obtained from simulations of a lattice containing 5×30^2 spins. However, comparison with those obtained from large \( r \) lattices shows negligible finite-size effects. In order to compare the experimental data to the simulations, we note that \( \frac{3}{kT} \) \( \propto \beta \) (since the interaction energy \( \beta \) is proportional to \( \frac{3}{a} \)). We can convert the effective temperature \( \frac{kT}{\beta} \) from simulation to lattice constant \( a \) by finding the correct proportionality constant \( C \) such that \( \frac{3}{C} a \frac{1}{kT} = \beta \). We pick \( C \) such that \( \frac{5.7}{kT} = 880 \) nm.

**Fig. S1.** Vertex population fractions for the short-island shakti (a-c), long-island shakti (d, e), and square (f) lattices compared to the Monte Carlo simulations described in the text. Note that the Type I fraction in the square lattice goes to nearly unity at small lattice constants, indicating good thermalization. Also note that the fractions of Type I_3 and II_3 vertices go to 0.5 at small lattice constants—half of the excited Type II_3 are topologically protected and cannot be eliminated from the lattice.

**Six-vertex Model Correspondence**

Figure S2 below gives a more detailed description of mapping between the shakti lattice and the six-vertex model.
**Fig. S2.** (a) and (b) reproduce the MFM image and spin map of the short-island shakti lattice shown in Fig. 2e and h. Panel C marks the defect (Type II₃) vertex sites with solid dots and ground state (Type I₃) vertices with open dots. The many possible arrangements of defects on the possible sites give the shakti lattice its ground state degeneracy. Panel (d) shows the defect sites mapped to the arrows usually used to visualize the six-vertex model. The correspondence is laid out in (e): each plaquette with vertical (horizontal) central islands has arrows placed pointing toward (away from) the defects and away from (toward) the empty defect sites. Note that the red arrows in (d) and (e) correspond to the defect sites and *not* to the blue arrows representing island moments in (b).

**Disorder**

Figure S3 shows the vertex populations from Monte Carlo simulations without disorder and with 20% disorder in vertex energies. The vertex fractions are not affected by the presence of disorder at this level.
**Fig. S2.** (a) and (b) reproduce the MFM image and spin map of the short-island shakti lattice shown in Fig. 2e and h. Panel C marks the defect (Type II) 3 vertex sites with solid dots and ground state (Type I) 3 vertices with open dots. The many possible arrangements of defects on the possible sites give the shakti lattice its ground state degeneracy. Panel (d) shows the defect sites mapped to the arrows usually used to visualize the six-vertex model. The correspondence is laid out in (e): each plaquette with vertical (horizontal) central islands has arrows placed pointing toward (away from) the defects and away from (toward) the empty defect sites. Note that the red arrows in (d) and (e) correspond to the defect sites and not to the blue arrows representing island moments in (b).

**Fig. S3.** (a) Four-island vertex populations from Monte Carlo simulations with no disorder (solid lines) and 20% disorder in vertex energies (dashed lines). (b) Three-island vertex populations, again comparing results from simulations with no disorder (solid lines) and 20% disorder (dashed lines). The differences between the vertex populations from disorder-free simulations and simulations including disorder are negligible, indicating that the shakti lattice is robust to weak disorder.
Charge excitation screening in the short-island shakti lattice

When the quantities $\langle Q_{nn} \rangle$ and $P(Q_{nn} = -4)$, as defined in the main text, for the short-island shakti lattice are compared with Monte Carlo simulations including or excluding long-range charge-charge interactions, we find results similar to those in the main text for the long-island shakti lattice, as shown in Fig. S4.

Fig. S4. Screening of Type III$_4$ charge excitations in the long- (a, b) and short- (c, d) island shakti lattice. Simulation MC 1 does not include interactions between magnetic charges, while simulation MC 2 does; it is clear that MC 2 better describes the data, demonstrating the importance of interactions between magnetic charges in understanding the behavior of this system.