Extensive degeneracy, Coulomb phase and magnetic monopoles in artificial square ice

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Artificial spin-ice systems are lithographically patterned arrangements of interacting magnetic nanostructures that were introduced as a way of investigating the effects of geometric frustration in a controlled manner1–4. This approach has enabled unconventional states of matter to be visualized directly in real space5–18, and has triggered research at the frontier between nanomagnetism, statistical thermodynamics and condensed matter physics. Despite efforts to create an artificial realization of the square-ice model—a two-dimensional geometrically frustrated spin-ice system defined on a square lattice—no simple geometry based on arrays of nanomagnets has successfully captured the macroscopically degenerate ground-state manifold of the model19.

Instead, square lattices of nanomagnets are characterized by a magnetically ordered ground state that consists of local loop configurations with alternating chirality1,20–26. Here we show that all of the characteristics of the square-ice model are observed in an artificial square-ice system that consists of two sublattices of nanomagnets that are vertically separated by a small distance. The spin configurations we image after demagnetizing our arrays reveal unambiguous signatures of a Coulomb phase and algebraic spin-spin correlations, which are characterized by the presence of ‘pinch’ points in the associated magnetic structure factor. Local excitations—the classical analogues of magnetic monopoles27—can be seen to evolve in an extensively degenerate, divergence-free vacuum. We thus provide a protocol that could be used to investigate collective magnetic phenomena, including Coulomb phases and the physics of ice-like materials.

To recover the true degeneracy associated with the square-ice model, we fabricated a series of artificial square-ice systems inspired by a previous theoretical proposition29. The main idea behind that proposition is to reduce the coupling strength between perpendicularly oriented nanomagnets ($J_1$) while keeping the coupling strength between colinear nanomagnets ($J_2$) unchanged by vertically shifting one of the two sublattices of the square array (Fig. 1a). Such a height offset $h$ makes it possible to finely tune the $J_1/J_2$ ratio. If $h = 0$, then the system is a conventional artificial square-ice system, characterized by $J_1 > J_2$ and a magnetically ordered ground state (Fig. 1b). If $h$ is continuously increased, then $J_1$ can become infinitively small compared to $J_2$ until a situation is reached where the horizontal and vertical lines of the square lattice are magnetically decoupled ($J_1 = 0$; Fig. 1c). Therefore, there is a critical height offset $h_c$ at which the two coupling coefficients $J_1$ and $J_2$ are equal (Fig. 1d). On the basis of a dumbbell description of the nanomagnets, it was found28 that $h_c = 0.207a$ for $l/a = 0.7$, where $l$ is the length of the nanomagnets and $a$ is the lattice parameter. A critical height offset of $h_c = 0.27a$ was calculated29 by incorporating dipolar interactions over the entire volume of uniformly magnetized nanomagnets. However, both of these approaches neglected key experimental ingredients: the geometric properties and the micromagnetic nature of the nanomagnets were not taken into account. Here, we determine $h_c$ from a set of micromagnetic simulations that describe the real shape and internal micromagnetic configuration of the nanomagnets used experimentally (Methods). The main result of our calculation is that $h_c$ strongly depends on the gap left between neighbouring magnetic elements, and is qualitatively similar to the estimate deduced from the dumbbell description (Fig. 1e and Extended Data Fig. 1), but quantitatively very different.

To recover the degeneracy of the square-ice model, we need to lithographically pattern arrays of nanomagnets in which the third dimension now plays a key part, extending artificial spin-ice systems from two to three dimensions. This additional dimension makes the fabrication and imaging of spin-ice architectures much more challenging. Our shifted artificial square-ice systems were fabricated using a two-step electron-beam lithography process (Methods and Extended Data Fig. 2). The first step is dedicated to the design of non-magnetic bases that are used to lift one sublattice of nanomagnets. The thickness of the bases determines the final height offset $h$ (the base thicknesses used here are 60 nm, 80 nm and 100 nm). The second step
consists of depositing the nanomagnets on a square lattice in such a way that one sublattice is grown atop the non-magnetic bases and the other is grown on the substrate. On each sample, a reference square lattice with $h = 0$ is patterned for direct comparison with the shifted arrays. Magnetic images were obtained using magnetic force microscopy (Fig. 2a) after demagnetizing the arrays in an in-plane oscillating magnetic field with slowly decaying amplitude (Methods). All of the arrays were demagnetized simultaneously to ensure identical field history between samples.

The three shifted arrays we studied were demagnetized four times to improve the statistics and to test the reproducibility of the experimental observations, and we systematically imaged the reference array ($h = 0$) present on each sample to check the efficiency of the field demagnetization protocol. For these 12 realizations, the reference arrays were always found in a magnetic configuration close to the ordered antiferromagnetic ground state (Fig. 1b). A typical magnetic state is shown in Fig. 2b, in which a domain boundary separating two anti-phase domains is observed. Consequently, type-I vertices are present everywhere, except in the domain wall formed by type-II vertices (for definitions of type-I and -II vertices, see the inset of Fig. 1a).

Our demagnetization protocol is therefore efficient and brings the system into a low-energy manifold, with large patches of the ground-state configuration, similar to what is found in thermally active artificial spin ice\textsuperscript{10,21,24–26}. For the reference arrays, the density of type-I, -II, -III and -IV vertices are, on average, 86%, 12.5%, 1.5% and 0%, respectively (Fig. 2c; type-III (-IV) vertices refer to vertices with three (four) in or three (four) out spin configurations\textsuperscript{3}), and the mean size of type-I domains is about 87 vertices. Consequently, the residual magnetization is low, typically 3% in both the vertical and horizontal directions. The computed magnetic structure factor (Methods and Extended Data Fig. 3), averaged over the 12 different reference arrays, shows clear magnetic Bragg peaks located at the corners of the Brillouin zone (Fig. 3a).

Figure 2c shows the variation in vertex density $\rho$ when $h$ is increased, revealing a clear trend: the density of type-I vertices continuously decreases whereas the density of type-II vertices increases. For $h = 60$ nm, the physics is essentially unchanged from the $h = 0$ case: type-I vertices are the most prevalent and form patches of the antiferromagnetic ground state, although the average size of the ordered domains decreases to 15 vertices—6 times smaller than for the reference arrays. The corresponding magnetic structure factor shows a spreading of the magnetic Bragg peaks associated with antiferromagnetic ordering, but the peaks remain located at the corners of the Brillouin zone (Fig. 3b). This result is consistent with the predictions from micromagnetic simulations (Fig. 1e), which indicate that the ground state is expected to be the antiferromagnetic ordered configuration when $h = 60$ nm.

For $h = 80$ nm, the population of type-II vertices (52%) becomes higher than that of type-I vertices (39%); type-II patches start to form and the spatial extent of type-I domains is further reduced. The magnetic Bragg peaks in the magnetic structure factor have almost disappeared, which is an indication that the spin configurations have started to be disordered. If the background intensity in the magnetic structure factor becomes more diffuse, then it develops a structure with geometric features that resemble those expected from the square-ice model (Fig. 3c, e and Methods). This result is consistent with the micromagnetic simulations: although the ground state is expected to be the antiferromagnetic ordered configuration, the magnetic configuration is disordered after demagnetizing the array, as $h$ approaches $h_e$ ($J_1$ starts to compare with $J_2$).

The similarity to the square-ice model becomes more evident for $h = 100$ nm (Fig. 3d, e). Contrary to all previous results, which demonstrate that square lattices of nanomagnets are magnetically ordered in their low-energy manifold, we show that our artificial square ice is highly disordered. The magnetic Bragg peaks in the magnetic structure factor have totally disappeared for $h = 100$ nm and the diffuse background is strongly structured. However, on the basis of the micromagnetic simulations presented above, the ground state of artificial square ice with $h = 100$ nm is expected to be ordered. We interpret this difference between observation and prediction as a consequence of the kinetics associated with the spin dynamics when the sample is demagnetized under a rotating magnetic field. During the demagnetization protocol, spins are reversed via an avalanche process that favours the formation of straight lines (Methods and Supplementary Videos 1 and 2). Type-II vertices are then stabilized by the external magnetic field at the expense of type-I vertices, even though type-I vertices have a slightly lower energy. In other words, our protocol shifts the critical value $h_c$, at which the transition to the disordered phase is expected.

![Figure 2](image-url)

**Figure 2 | Experimental results.** a, Topography (atomic force microscopy; top) and magnetic (magnetic force microscopy; bottom) images of our artificial realization of the square-ice model. In the topography image, the nanomagnets appear red, the bases are yellow and the substrate is grey. In the magnetic image, the magnetic contrast appears in blue and red for negative and positive magnetic charges, respectively. Typical contrasts obtained on type-I and -II vertices are shown in the inset. Type-I (-II) vertices correspond to local two-in/two-out spin configurations carrying zero (net) magnetic moment. b, Magnetic images (raw data on the left and the corresponding analysis on the right) for a height offset of $h = 0$ (top) and $h = 80$ nm (bottom). For $h = 0$, most of the vertices are type-I (blue), and a domain boundary separating anti-phase domains is clearly visible (type-II vertices are shown in red). For $h = 80$ nm, the magnetic state appears disordered; type-III vertices are coloured in green. c, Analysis of the vertex density of type-$i$ vertices $\rho_i$ as a function of the height offset $h$. The points represent the mean and the error bars represent the standard deviation calculated from the four demagnetizations.
Whereas the theoretical scan reveals a sharp peak associated with a correlation length of $\xi_{\text{theo}} = 5.2a \pm 4\%$, the experimental scan displays a broader peak associated with a shorter correlation length of $\xi_{\text{exp}} = 4.4a \pm 12\%$. The broader peaks indicate the presence of local excitations, that is, a finite density of classical monopoles within a Coulomb phase.

Experimentally, we observe a similar density of $+2$ and $-2$ monopoles for all of the arrays (although they do not systematically obey charge neutrality owing to their finite size), with the density increasing as we retrieve the degeneracy of the square-ice model (see $h$ dependence of type-III vertices in Fig. 2c). The variation in the monopole density we measure is not random and appears to be robust when comparing successive demagnetization protocols. The

Figure 3 | Magnetic structure factors and pinch-point analysis. a–d, Magnetic structure factors deduced from the experimental images for $h = 0$ (a), $h = 60$ nm (b), $h = 80$ nm (c) and $h = 100$ nm (d). The colour scale refers to the intensity at a given point $(q_x, q_y)$ of reciprocal space. e, Computed magnetic structure factor averaged over 1,000 random, decorrelated spin configurations that satisfy the ice rule (Methods). f, Experimental (main plot) and theoretical (inset) intensity profiles across the pinch point highlighted by a red circle in d. The points represent the mean and the error bars represent the standard deviation calculated from the four demagnetizations for the experimental data and the 1,000 random ice-rule configurations for the theoretical data. The red curves are single-peaked Lorentzian fits of the pinch points. The $q$ axis corresponds to a scan from $(3/2, 5/2)$ to $(5/2, 3/2)$ in reciprocal space. r.l.u., reciprocal lattice unit; a.u., arbitrary units.

Figure 4 | Magnetic monopoles in square-ice systems. a–c, Monopole/anti-monopole pair (red and blue circles) in a magnetically saturated background (type-II background; a), in the antiferromagnetic ground state (type-I background; b) and within a disordered manifold (disordered background; c). Blue, red and green squares indicate type-I, -II and -III vertices, respectively. The black arrows in a illustrate a chain of reversed spins. d, Experimental spin configurations for $h = 100$ nm showing two pairs of oppositely charged monopoles. e, Analysis of the configuration in d. Monopoles appear as red and blue circles on top of a green square.
densities are fairly high when approaching the spin-liquid phase (Fig. 2c), meaning that we do not bring the system into its massively degenerate ground-state manifold. Instead, the imaged spin configurations are characteristic of excited states embedded within a Coulomb phase. The monopoles that we observe in our arrays differ substantially from those that have previously been visualized in artificial square ices.\textsuperscript{2,20–26} All of the monopoles reported so far are high-energy local configurations evolving in an uncharged, but magnetically ordered, vacuum (Fig. 4a, b), characterized by a magnetic structure factor that contains only magnetic Bragg peaks (Methods).

Our system has very distinct behaviour: the monopoles we observe are free to move into a spin-liquid state, that is, a massively degenerate, disordered low-energy manifold (Fig. 4c). They are therefore particle-like objects present in a diffuse but structured magnetic structure factor, free of any Bragg peaks (Fig. 3d). An example experimental configuration is shown in Fig. 4d and is schematized in Fig. 4e. Two pairs of oppositely charged monopoles are present in this disordered magnetic configuration containing type-I and -II vertices. It is not possible to determine the path that these monopoles have followed during the demagnetization process, because the trace of reversed spins (often referred to as a Dirac string) has been erased by the magnetic disorder. This is in contrast to the aforementioned cases for which monopoles evolve within an ordered spin configuration and for which the influence of the field or temperature can be unambiguously visualized (Fig. 4a, b). Here, there is no way of knowing the trajectory of the magnetic monopoles and, consequently, it is not even possible to pair two oppositely charged monopoles.

This result raises interesting, crucial questions. We envision that, if similar artificial, shifted square-ice systems could be made thermally active, the dynamics of these de-confined, interacting quasi-particles could be investigated in real space and time. We then wonder whether a typical distance between oppositely charged monopoles would be established at thermodynamic equilibrium and whether this distance could be linked to the correlation length that was deduced from the analysis of the width of the pinch point (Extended Data Fig. 6 and Methods). It would also be interesting to study how these quasi-particles nucleate, propagate and annihilate with their anti-particle, and to directly observe how their interactions affect the disordered background.

We have shown that shifted magnetic square lattices offer the possibility to tune the nearest-neighbour coupling strength and, in particular, to experimentally realize the semial square-ice model. The fabrication of thermally active, shifted square ice in the future would enable the thermodynamics and dynamics of the low-energy manifolds and the recombination of their topological excitations to be investigated (Methods). Finally, our work demonstrates that artificial loop models, such as the square-ice model, are not beyond reach, thanks to lithography engineering. These models have numerous extensions in very different fields of research, including polymer physics, topological quantum computing, self-avoiding random walks and Schramm–Loewner evolution. Implementing loop models is therefore of broad interest in physics and chemistry, and our contribution illustrates that magnetic versions of these loop models are experimentally accessible.

**Online Content** Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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to evolve even after several hours of demagnetization, consistent with what has been observed previously\(^3\),\(^4\),\(^5\). It seems that, in general, allowing a large number of spin-flip events is the key ingredient to reaching low-energy manifolds in artificial spin systems. For example, to demagnetize Kagome lattices, 110-h demagnetization protocols helped us to reach an effective temperature of about 0.06\(T_{\text{room}}\) (where \(T_{\text{room}}\) is the nearest-neighbour coupling strength) and to observe spin fragmentation\(^14\). With shorter protocols, fragmentation was not observed because the associated (effective) temperature remained too high. For our square arrays, we find that three days is the optimum protocol length; we do not see substantial differences when applying longer demagnetization protocols.

**Numerical demagnetization.** To interpret our experimental data, we performed numerical simulations of the field demagnetization protocol. We consider a square array of magnetic point dipoles carrying a magnetic moment \(\mu\). The interaction energy \(E_{ij}\) between two spins \(i\) and \(j\) separated by a distance \(r_{ij}\) is of dipolar-type, with a cut-off radius \(r_c\):

\[
E_{ij} = \begin{cases} \frac{1}{r_{ij}^3} \mu_i \cdot H_i - \frac{3}{r_{ij}^5} (\mu_i \cdot r_{ij}) (\mu_j \cdot r_{ij}) & \text{if } r_{ij} < r_c \\ 0 & \text{otherwise} \end{cases}
\]

The local field \(H_{ij}^1\) felt by the dipole \(\mu_i\) is the sum of all dipolar fields coming from the surrounding spins plus the external applied magnetic field. Each dipole \(\mu_i\) behaves as an Ising pseudo-spin with its own switching field \(H_{sw}^i\). Then, if the spin \(i\) is flipped. Following previous work\(^22\), we introduce a Gaussian distribution of the switching field \(H_{sw}^i\) to allow the system to approach its low-energy manifold. The probability \(P(H_{sw}^i)\) for a spin \(i\) to have a switching field \(H_{sw}^i\) is

\[
P(H_{sw}^i) = \frac{1}{\sigma \sqrt{2\pi}} \exp\left(\frac{(H_{sw}^i - E_{sw})^2}{2\sigma^2}\right)
\]

where \(E_{sw}\) is the average switching field and \(\sigma\) the spreading (standard deviation) of the distribution. For lattices with \(h = 0\) (it being the height offset), the ground state is ordered and \(\sigma\) provides a control on the density of nucleation sites during the demagnetization protocol. Our experimental results are well reproduced if \(\sigma\) is set to 0.1\(H_{sw}\). In the simulations, the field ramp decreases linearly through 10\(^6\) steps and 10\(^4\) turns. Numerical demagnetizations showing the reversal of spin chains and the complete protocol are provided as Supplementary Videos 1 and 2.

The same code is used here: the four vertex types (I–IV) are represented by squares coloured in blue, red, green and yellow, respectively. As in the experiments, type-IV vertices are never observed in the simulations.

**Magnetic structure factor.** We define the magnetic structure factor as in neutron scattering experiments, in which the spin correlations perpendicular to the diffusion vector \(\mathbf{q}\) are measured. We therefore define a perpendicular spin component \(S_{\mathbf{q}}^\perp\):

\[
S_{\mathbf{q}}^\perp = S_{\mathbf{q}} - (\mathbf{q} \cdot S_{\mathbf{q}}) \hat{\mathbf{q}}
\]

where \(\hat{\mathbf{q}}\) is the unit vector along the diffusion vector \(\mathbf{q}\).

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Extended Data Fig. 3a shows the geometric construction of the vectors involved in equation (1). The intensity \(I(q)\) scattered at location \(q\) in reciprocal space is defined as

\[
I(q) = \frac{1}{N} \sum_{\alpha, \beta} 2 S_{\alpha} \cdot S_{\beta} \exp(i \mathbf{q} \cdot \mathbf{r}_{\alpha,\beta})
\]

in which \(i\) and \(j\) scan all \(N/2\) unity cells, and \(\alpha\) and \(\beta\) the two sites of each cell. However, obtaining a more convenient form for equation (2) would enable a direct calculation starting from a magnetic configuration. \(I(q)\) can be split into two parts \(I = I^+ - I^-\) with

\[
I^+(q) = \frac{1}{N} \sum_{\alpha, \beta} \rho_\alpha(q) \rho_\beta^*(q)
\]

\[
I^-(q) = \frac{1}{N} \sum_{\alpha, \beta} \rho_\alpha(q) \rho_\beta^*(q)
\]
where 
\[g_i(q) = \sum_{r} \sigma_i \exp(iq \cdot r)\]
and \(\sigma_i\) is the Ising variable \((\pm 1)\) of the site \(i\). In this split form, \(f\) is a real quantity. To compute \(I(q)\) diagrams in reciprocal space, we calculate the quantity \(I = g_i^2 + g_j^2 - (p_i + p_j)^2\) at several \(q\) locations. The magnetic structure factor reported in Extended Data Fig. 3b is composed of a matrix of 120 × 120 points covering an area of \(q_r, q_i \in [-6\pi, 6\pi]\). This area is 36 times larger than the first Brillouin zone.

**Generating low-energy magnetic configurations.** The magnetic structure factor of the square-ice model was computed by averaging 1,000 low-energy spin configurations that satisfy the ice rule everywhere. To do so, we start from a magnetically saturated configuration and then flip a number \(N\) of randomly chosen spin loops. These loop flips are necessary to ensure that all of the generated spin configurations satisfy the ice rule. Because our lattice has free boundary conditions, the procedure leads to open (crossing the array) or closed loops (Extended Data Fig. 4). Both loops are used to generate a low-energy spin configuration. To decorrelate the initial (saturated) and final spin configurations, we take \(N\) to be of the order of the number of spins present in the array \((840)\).

**Pinch points and correlation length.** The magnetic structure factor shown in Extended Data Fig. 3b is averaged over 1,000 ice-rule configurations. Pinch points located at the centre \(\Gamma\) of the Brillouin zone are clearly visible, indicating the existence of a Coulomb phase and algebraic spin–spin correlations, that is, a correlated, disordered magnet within which spin–spin correlations decay like point-dipole interactions\(^{36}\). The finite size of our arrays has consequences for the magnetic structure factor, in particular for the width of the pinch points. Extended Data Fig. 5 shows the influence of the lattice size \(L\) on the width of the pinch points, which narrow as the lattice size is increased. This width of the pinch points can be linked to a correlation length \(\xi\) in the system\(^{37}\). This correlation length can be extracted from a Lorentzian fit to the intensity profile passing through a pinch point:

\[I(q) = A - \frac{\xi^2}{(q - q_0)^2 + \xi^2 + B}\]

where \(A\) and \(B\) are constants, \(q_0\) is the location of the pinch point in reciprocal space, and \(q\) is the diffusion vector. The correlation lengths deduced for different lattice sizes are reported in Extended Data Table 1. Uncertainties represent the variability within the 1,000 sampled spin states.

**Long-range dipolar interactions.** Although the square-ice model is a vertex model, here we are interacting dipoles with long-range effects. It is not clear whether the infinite range of the dipolar interaction affects the physics of the model, here we have interacting dipoles with long-range effects. It is not clear whether the infinite range of the dipolar interaction affects the physics of the model, here we have interacting dipoles with long-range effects. It is not clear whether the infinite range of the dipolar interaction affects the physics of the model, here we have interacting dipoles with long-range effects. It is not clear whether the infinite range of the dipolar interaction affects the physics of the model, here we have interacting dipoles with long-range effects.
Extended Data Figure 1 | Dumbbell description of the nanomagnets.
Map of $J_1/J_2$ as a function of $l/a$ and $h/a$ for an isolated vertex.
The condition $J_1 = J_2$ is indicated by the dark line. Our results perfectly reproduce those reported in ref. 29. The white dots indicate the values that correspond to the different samples studied here.
Extended Data Figure 2 | Illustration of the two-step electron-beam lithography process. a, Schematic of the gold bases subsequently used to shift the vertical sublattice. b, Schematic of the permalloy magnets on the vertical and horizontal sublattices.
Extended Data Figure 3 | Magnetic structure factor of the square-ice model. a, Sketch of the vectors involved in equation (1). b, Magnetic structure factor for an ideal square-ice model, computed for 1,000 low-energy states made of $N = 840$ spins. Red circles indicate the regions of interest for the intensity profiles in Fig. 3f and Extended Data Fig. 5.
Extended Data Figure 4 | Loop flips in the square lattice. Schematic illustrating the open (red arrows) and closed (green arrows) spin loops used to generate a low-energy configuration that is representative of the massively degenerate ground-state manifold of the square-ice model\textsuperscript{19}. The lattice contains 840 spins and the number of loops that are flipped between two decorrelated configurations is set to \( N = 840 \). \( L \) corresponds to the linear size of the square lattices.
Extended Data Figure 5 | Analysis of the pinch points. a–d, Maps of the pinch points indicated by red circles in Extended Data Fig. 3b (left) and associated intensity profiles along the \( q_v = 0 \) direction (right), for different lattice sizes \( L \): \( L = 10 \) (a), \( L = 20 \) (b), \( L = 40 \) (c), \( L = 80 \) (d). The colour scale refers to the intensity at a given point of reciprocal space. The coordinates \( (q_u, q_v) \) are relative to the intensity profile and do not correspond to the real axes of reciprocal space. The red curves are single-peaked Lorentzian fits; the points represent the mean and the error bars represent the standard deviation calculated from 1,000 random ice-rule configurations.
Extended Data Figure 6 | Magnetic monopoles in artificial square ice.
Experimental spin configuration for $h = 100$ nm. Type-I and -II vertices appear as blue and red squares, respectively. Monopoles appear as red and blue circles. Their associated pairing is represented by black ellipses.
Extended Data Table 1 | Correlation lengths extracted from the intensity profiles

| $L$ | 10   | 20   | 40   | 80   |
|-----|------|------|------|------|
| $\xi$ | 3.19$a$ ± 2% | 5.2$a$ ± 4% | 8.2$a$ ± 7% | 13$a$ ± 14% |