

RESEARCH ARTICLE OPEN ACCESS

Pyrochlore Artificial Spin Ice: Advances in Fabrication and Characterization of a 3D Magnetic Metamaterial

Luca Berchialla^{1,2} | Arjen van den Berg³ | Gavin M. Macauley^{1,2} | Evangelos Goliás⁴ | Yuran Niu⁴ | Tianyue Wang^{1,2} | Armin Kleibert⁵  | Sam Ladak³ | Laura J. Heyderman^{1,2} 

¹Laboratory for Mesoscopic Systems, Department of Materials, ETH Zurich, Zurich, Switzerland | ²PSI Center for Neutron and Muon Sciences, Villigen PSI, Switzerland | ³School of Physics and Astronomy, Cardiff University, Cardiff, UK | ⁴MAX IV Laboratory, Lund University, Lund, Sweden | ⁵PSI Center for Photon Science, Villigen PSI, Switzerland

Correspondence: Luca Berchialla (l.berchialla@gmail.com) | Arjen van den Berg (vandenberga@cardiff.ac.uk) | Gavin M. Macauley (gm3291@princeton.edu) | Laura J. Heyderman (laura.heyderman@psi.ch)

Received: 26 February 2026 | **Revised:** 14 July 2026 | **Accepted:** 10 August 2026

Keywords: 3D artificial spin ice | magnetic force microscopy | magnetic metamaterials | photoemission electron microscopy | two-photon lithography

ABSTRACT

Artificial spin ices are magnetic metamaterials consisting of arrays of magnetically coupled nanoscale magnets arranged in various geometries. They display a variety of fascinating phenomena including emergent magnetic monopoles, phase transitions and frustration. While the nanomagnets are often arranged in planar arrays, the development of artificial spin ices in three dimensions would allow the exploration of new fundamental phenomena. 3D artificial spin ices are also of interest for neuromorphic computing because of the rich magnetic state space, enhanced connectivity, and increased areal density. Here, a 3D artificial spin ice based on the pyrochlore lattice found in a spin ice crystal is presented. In this Pyrochlore artificial spin ice, disconnected nanomagnets, inclined at an angle to the substrate, are located at four different heights. The nanomagnets are held up by a polymer scaffold fabricated using a combination of two-photon lithography and pyrolysis. The scaffold consists of sloped nanomagnet supports on tailored hemispherical decoupling structures that ensure uniform shrinkage during pyrolysis. Imaging with magnetic force microscopy indicates that the nanomagnets adopt a variety of magnetic states including single-domain, s-, and vortex states, while the necessity for further advances in synchrotron X-ray imaging techniques tailored to these 3D magnetic architectures is highlighted.

1 | Introduction

Artificial spin ices (ASIs) are arrays of magnetically coupled single-domain nanomagnets arranged in geometries that are often designed to enforce frustration where it is not possible to minimize all pairwise nanomagnet interaction energies simultaneously [1, 2]. They display a variety of fascinating phenomena such as emergent magnetic monopoles [3–5], phase transitions [6, 7], and vertex frustration [8]. Early ASIs were designed to emulate the spin geometry of the titanate pyrochlores whose crystal-field anisotropy constrains spins to point along the local $\langle 111 \rangle$ axes, yielding low-energy ice-rule-obeying configurations

with two spins pointing into and two pointing out of each tetrahedron [9]. In such spin ice crystals, the spins on the corner-sharing tetrahedra are associated with a large number of low-energy degenerate states, which is a signature of geometric frustration. In a low-energy state, reversing a single spin on a tetrahedron, which can happen as a result of thermal excitation, leads to a violation of the ice rule, producing three-in/one-out or one-in/three-out defects that behave as effective magnetic monopoles interacting via a Coulomb-like law [10].

ASIs are typically fabricated in two dimensions with the nanomagnets lying in a single plane and, in order to capture the

Luca Berchialla and Arjen van den Berg contributed equally to this work.

This is an open access article under the terms of the [Creative Commons Attribution](https://creativecommons.org/licenses/by/4.0/) License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

© 2026 The Author(s). *Small Structures* published by Wiley-VCH GmbH.

geometry of the pyrochlores with a coordination number of four, a square lattice arrangement of nanomagnets is often adopted. However, this lifts the degeneracy of the low-energy magnetic configurations through unequal interactions between neighboring nanomagnets, leading to additional energy terms in the monopole physics [3]. Nevertheless, modern lithographic techniques offer precise control over lattice geometry, as well as nanomagnet shape, size, separation, and material, thus enabling fine tuning of interactions between the nanomagnets, which can be exploited to restore the degeneracy of the magnetic configurations. On the whole, the study of two-dimensional (2D) ASIs remains an effective approach for exploring fundamental phenomena, and ASIs also show great promise for computing applications, including logic gates [11] and neuro-morphic devices [12, 13].

Recent advances in three-dimensional (3D) nanostructuring, particularly with two-photon lithography and focused electron beam-induced deposition, have enabled a wide range of 3D magnetic architectures to be fabricated, from simple domain wall conduits [14, 15] to chiral systems that host novel spin textures [16, 17], as well as conformally coated lattices that allow fine tuning of spin-wave spectra [18]. Central to these developments has been the drive to realize 3D ASIs [19], which have up to now been made from connected magnetic nanowires and include frustrated buckyball geometries [20–23], diamond-bond-like connected nanowire networks that mimic the pyrochlore lattice [24], and the constituent building blocks [25, 26].

Connected 3D ASIs have quickly demonstrated their potential, opening possibilities to observe nucleation and propagation of monopole-like defects [27], charge-ordered states [28], topological textures in the stray field [29], and magnetization dynamics [30, 31]. However, in such connected systems, the magnetic charges take the form of domain walls, and the nanomagnet interactions have additional contributions associated with the junctions where the nanowires meet. In addition, the voxel shape of the laser used for two-photon lithography results in nanowires with an elliptical cross-section, and this leads to a lifting of the degeneracy for ice-rule-obeying vertices [27] as well as among the monopoles themselves [29]. These complexities enrich the associated physics and can lead to several useful behaviors, for example, fading memory and nonlinearity, that can be exploited in reservoir computing [32], but they also move the system away from a purely dipolar analog of natural spin ice.

3D ASIs, such as inverse opal-like or gyroid structures, can be fabricated using bottom-up or self-assembly-based methods [33, 34]. These fabrication methods can be used to create large areas of small regular 3D magnetic nanostructures suitable, for example, for investigations with neutron scattering [33]. However, these methods lack the flexibility to finely tune the geometry, for example, in terms of the number of nanomagnets that meet at a vertex and nanomagnet angle, and again provide nanowire networks rather than arrays of disconnected nanomagnets.

It is therefore desirable to realize 3D ASIs in which the nanomagnets are physically separated and interact exclusively through dipolar coupling. With this, the accessible state space can be more simply tuned through geometric parameters such as nanomagnet orientation, size and separation, without unwanted modifications

to the energy landscape arising from connectivity. Creating such 3D ASIs not only offers better analogs to the crystal spin systems than planar arrays of nanomagnets but also paves the way to designer magnetic metamaterials with tunable bond angles, thus providing bespoke static and dynamic magnetic properties.

Here, we present a method to fabricate disconnected 3D ASIs through a combination of two-photon lithography, pyrolysis, and thin-film deposition. These magnetic metamaterials consist of physically separated nanomagnets arranged in a similar manner to the spins on the pyrochlore lattice, with different orientations and heights. We characterize this Pyrochlore ASI with magnetic force microscopy (MFM) and synchrotron-based X-ray photoemission electron microscopy (XPEEM), demonstrating both the opportunities and challenges associated with the determination of the magnetic configuration of 3D ASIs with physically separated nanomagnets.

2 | Design of the 3D Pyrochlore Artificial Spin Ice

The 3D ASI in this work comprises physically separated stadium-shaped nanomagnets that have lateral dimensions of $\sim 250 \times 650 \text{ nm}^2$. The nanomagnets are placed on a polymer scaffold, with the nanomagnets oriented at an angle of 45° to the substrate and located at four different heights, as illustrated in the Graphical Abstract Figure and Supplementary Movie. While the nanomagnets could have been oriented at approximately $\pm 35^\circ$ to the horizontal, which is analogous to the spin orientation in the titanate pyrochlores, an angle of 45° is chosen because it lies midway between the planar and perpendicular limits, making fabrication straightforward and yielding dipolar interactions with comparable lateral and vertical field components. This highlights the flexibility of this design—the nanomagnets can be fabricated at any tilt angle so that different geometries, which are inaccessible in natural spin systems, can be systematically explored.

The nanomagnets are located on top of sloped polymer platforms, shown in orange in the Graphical Abstract Figure and Figure 1a, which are raised to the required height with polymer support structures shown in turquoise. The support structures have four different heights; the lowest support has a height of $\sim 3 \mu\text{m}$ (650 nm) before (after) pyrolysis and the heights of the other three supports are set according to the design (see Figure S1). In this way, two unit cells are realized in the vertical direction. Having only two unit cells ensures that nanomagnets are not stacked on top of each other. This not only facilitates their fabrication but also means that all nanomagnets are visible from the top, as shown in Figure 1b, which is important for both deposition of the magnetic material onto every platform and subsequent determination of the full magnetic configuration with microscopy methods. In Figure 1c, a side view of the design is shown, which further illustrates how the tilted nanomagnets are placed at four different heights with respect to the substrate.

Magnetic functionality and interactions can then be obtained by depositing permalloy—a soft magnetic NiFe alloy typically used for ASIs—on the top of the polymer scaffold with thermal evaporation (gray coating shown in the Graphical Abstract Figure).

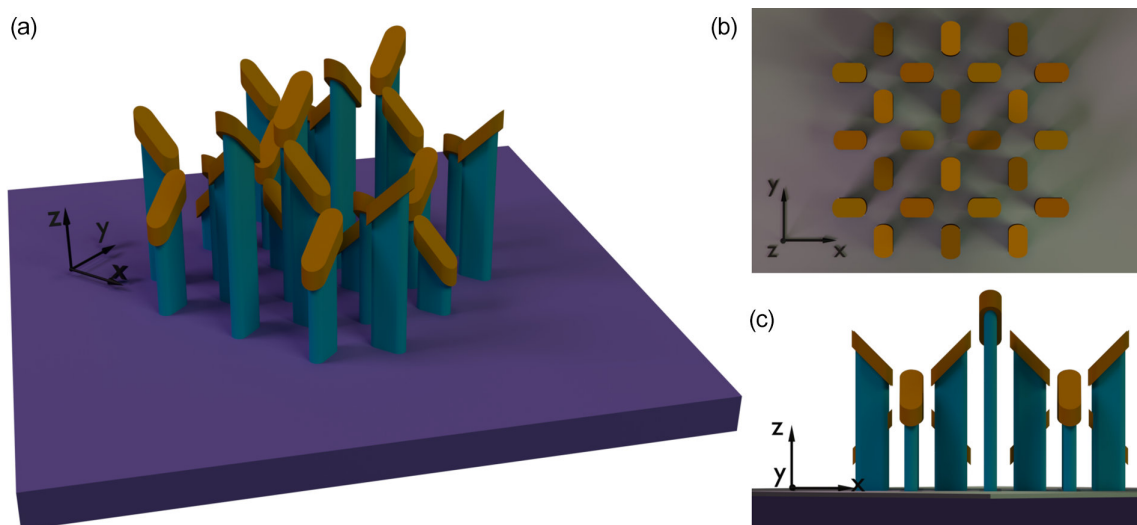


FIGURE 1 | Design of the polymer scaffold for the Pyrochlore ASI. (a) Tilted view, (b) top view, and (c) side view. The sloped platforms, shown in orange, are printed using piezo mode. The support structures, shown in turquoise, are fabricated using galvo mode. This combination of printing strategies enables the realization of the complex 3D geometry required to approximate a 3D ASI based on the pyrochlore lattice.

In this way, disconnected nanomagnets with the required height and orientation are fabricated on top of the sloped platforms. There is also a thin film of permalloy on the lower surface where the turquoise support structures meet the substrate, as shown in the Graphical Abstract Figure. However, since the thickness of the permalloy nanomagnets is usually in the range of a few nm to a few tens of nm, interactions between the permalloy nanomagnets on top of the support structures and the film on the substrate can be straightforwardly mitigated by implementing support structures with sufficient height, in this case with a height of more than 650 nm above the substrate surface.

3 | Fabrication Using Two-Photon Lithography and Pyrolysis

Realizing the disconnected 3D ASI based on the pyrochlore lattice shown in the Graphical Abstract Figure requires the ability to create a scaffold on which to place the nanomagnets, oriented at 45° to the substrate plane, at four different heights and four different orientations. These geometrical conditions can be easily met by printing a polymer scaffold with two-photon lithography, as demonstrated by previous works on nanowire lattices [24, 27, 28]. Other additive manufacturing techniques, such as focused electron beam-induced deposition, could also be used to fabricate such lattices [16, 35–38]. However, up to now, only structures of limited extent have been produced using this method. Moreover, in order to obtain disconnected nanomagnets, two different materials would need to be deposited in the same structure, which poses an additional challenge. One could also imagine nanostructuring a silicon substrate in such a geometry using electron beam lithography, but this would be extremely challenging as it would require many lithography and etching steps to produce the four different heights and the four differently oriented sloped platforms.

In addition to the geometrical requirements, each nanomagnet should ideally have a stadium shape with lateral dimensions not

much larger than $200 \times 500 \text{ nm}^2$ so that they are single domain as in traditional 2D ASIs. While it is possible to print features of 200 nm or smaller with two-photon lithography by scanning the laser in a single line, the voxel associated with the laser printing is an ellipsoid, which would result in the platforms having a curved surface. This should be avoided in a 3D ASI because placing the nanomagnets on a curved rather than a flat surface can alter their magnetic properties in undesired ways [39, 40]. Therefore, in order to mitigate the curvature produced by the voxel shape of the laser, each nanomagnet platform can be realized with the laser focus traveling over multiple overlapping lines. This creates a flat surface onto which the magnetic material can be deposited, confining the curvature to the nanomagnet edges. However, to create platforms small enough to support nanomagnets, it is first necessary to shrink such a polymer scaffold printed with two-photon lithography through pyrolysis.

In the following sections, we give details regarding the fabrication of the sloped supports for the nanomagnets, as well as the design of the decoupling structures used to give uniform shrinkage during pyrolysis. Further details concerning the two-photon lithography and pyrolysis are given in the Methods section and Figures S1–S3.

3.1 | Optimization of the Printing of the Sloped Supports

The design of the polymer scaffold for the Pyrochlore ASI is shown in Figure 1, with the nominal dimensions of the printed structure before pyrolysis given in Figure S1. Each nanomagnet lies on top of a sloped polymer platform, depicted in orange in the Graphical Abstract Figure and Figure 1. The orange slope is raised to the required height by a polymer support structure illustrated in turquoise. The sloped platform should have a flat surface with the side walls as vertical as possible in order to minimize the curvature of the nanomagnet and prevent magnetic material from being deposited on the support side walls.

A Nanoscribe Photonic Professional GT2 tool is used for two-photon lithography. To ensure a smooth and flat surface of the platforms, the so-called piezo mode—a precise writing mode for the fabrication of 3D structures with high resolution and smoothness—is used to expose a photoresist in parallel sloped lines (see Figure S2a). This ensures that the surface is smoother compared with the one produced using the high-speed galvo mode.

In Figure 1a, it can be seen that the lateral dimensions of the platforms (shown in orange) are designed to be slightly larger than those of the support structures (shown in turquoise). This is to ensure that, when the permalloy is deposited on top, disconnected thin-film nanomagnets are created on top of the platforms.

The support structure is printed as a woodpile structure using galvo mode, in which each layer of polymer resist is printed as a set of lines orthogonal to the sets of lines in neighboring layers, as shown in Figure S2a. This gives mechanically strong support structures.

In order to ensure symmetrical printing of the supports and platforms, and therefore even structures without unintended gradients in the surfaces, both the supports and platforms are printed with a sequence of lines that starts from the center and progressively reaches the edges. This printing sequence, illustrated in Figure S2b, allows for the reliable printing of large numbers of sloped supports with small distances between them.

3.2 | Design of the Decoupling Structures for Pyrolysis

The pyrolysis process shrinks the polymer scaffold printed with two-photon lithography to the required size. However, because the substrate does not shrink, it is desirable to decouple the printed structure from the substrate. Therefore, it is necessary to not only shrink the individual sloped supports for the nanomagnets but also the polymer surface on which they are placed. For this reason, a polymer decoupling structure is developed that has a flat top surface that can freely shrink, while the decoupling structure remains safely attached to the nonshrinking substrate. The decoupling structure is printed in galvo mode.

A decoupling structure with a straightforward geometry could be a tall cylinder (see Figure S4a), in which the part closer to the substrate deforms to mechanically decouple the upper portion of the cylinder from the substrate. However, a tall cylinder has a large volume that substantially increases the printing time. Therefore, in order to reduce the amount of material needed for the decoupling structure for a given top surface size, the area in contact with the substrate can be reduced. One approach to achieve this is to implement hemispheres as decoupling structures. Hemispheres have a very small area in contact with the substrate, which makes them very effective decoupling structures. However, for the same reason, they are also not firmly attached to the substrate and can detach or tilt, as can be seen in Figure S4b. This is not satisfactory, taking into consideration that, for XPEEM imaging, a flat surface parallel to the substrate is important. MFM also requires a horizontal, flat surface with surface tilts below $\sim 5^\circ$.

By imposing a minimum diameter at the lower part of the hemisphere, as shown in Figure S4c, a cylindrical base (colored orange in the inset of Figure S4c) is created that increases the area in contact with the substrate, thus giving better adhesion. However, this strategy reduces the decoupling of the hemisphere because part of it is now transformed into a cylinder. This can result in a highly concave top surface, as seen in Figure S4c.

In order to mitigate this problem, an additional short cylindrical section, which extends below the hemisphere, is introduced, as illustrated in blue in the inset of Figure 2b. With this approach, the total height of the decoupling structure is increased. When the diameter and height of this additional cylindrical section are properly optimized, one can then obtain a decoupling structure with a flat surface before and after pyrolysis. This decoupling structure design, with the nominal dimensions before pyrolysis given in Figure S1, is used to fabricate the 3D ASI based on the pyrochlore lattice. A top and side view of the whole structure before and after pyrolysis is presented in Figure 2 where, after pyrolysis, the platforms have dimensions of $\sim 250 \times 650 \text{ nm}^2$.

There is a low level of geometric disorder in the Pyrochlore ASI, as quantified in Figure S5, with a variation in inter-element separation of $\sim 3\%$. Furthermore, the variation in the nanomagnet aspect ratio is found to be less than 5% . Several hundreds of these structures were fabricated, and all displayed a similar precision in terms of the location of the nanomagnets, demonstrating that the fabrication of the near-perfect structures is highly reproducible. In 2D ASIs, disorder that is typically on the order of 10% can lead, for example, to a finite size of Dirac string avalanches [5].

At the center of the Pyrochlore ASI shown in Figure 2c, a defect with a reduced spacing between the nanomagnet supports can be seen, which is due to the fact that the top surface of the decoupling structure is slightly concave. The reason for this is that this sample was used for XPEEM measurements, for which the support structure should be as small as possible to avoid charging, and a decoupling structure with a small hemisphere diameter, here shrinking from $\sim 50 \mu\text{m}$ down to $\sim 15 \mu\text{m}$ during pyrolysis, typically results in a concave surface. While such a central defect will not change the overall behavior of the Pyrochlore ASI, similar defects have been shown to change the local magnetic energy landscape, for example, acting as pinning centers for domain boundaries [41] and nucleation centers for emergent magnetic monopoles [42]. Perfect uniformity of the nanomagnet supports may be achieved with further optimization of the design of the decoupling structure in terms of its overall diameter, the size of the bottom cylinder on which the hemisphere stands, and the design of nanomagnet supports to eliminate such a defect. Indeed, for the sample used for MFM measurements, the diameter of the hemispherical decoupling structure is $\sim 60 \mu\text{m}$ (after shrinking down from $\sim 150 \mu\text{m}$ during pyrolysis), so it is much larger. This means that its surface is naturally slightly convex and, instead of having a single Pyrochlore ASI at the center of the decoupling structure, as is the case for the XPEEM samples, there is room for several Pyrochlore ASIs located away from the center so that they do not have a central defect (see Figure S6).

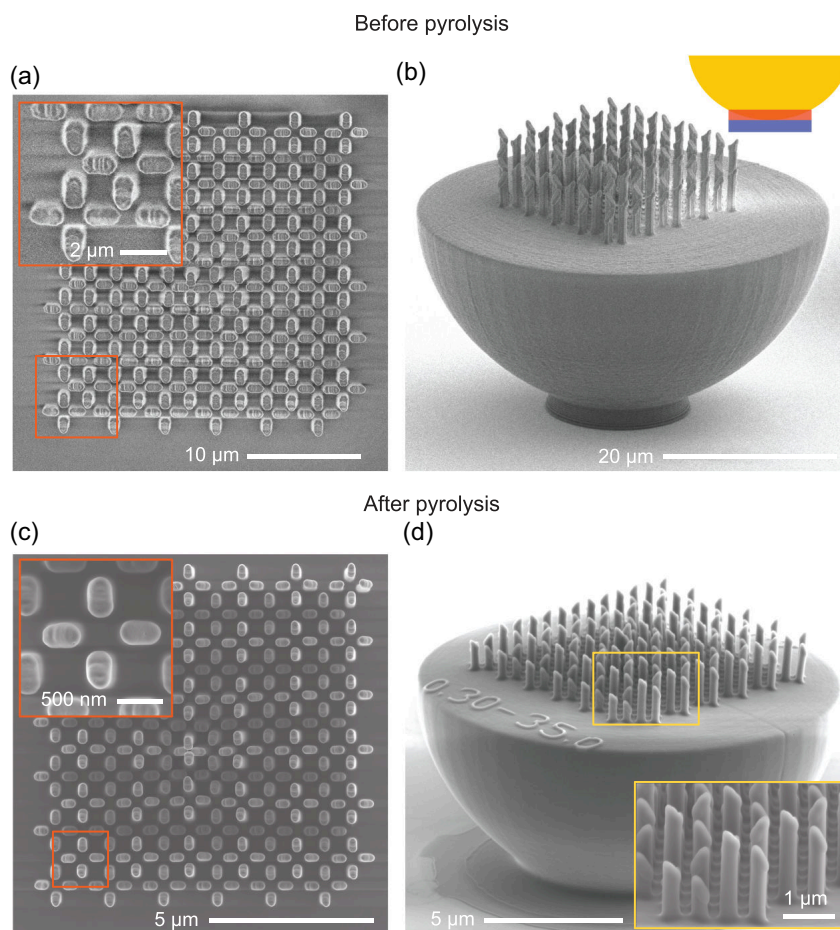


FIGURE 2 | Scanning electron micrographs of the polymer scaffold for Pyrochlore ASI before (a,b) and after pyrolysis (c,d). (a,c) Top view of the polymer scaffold and (b,d) side view of the polymer scaffold taken at a 70° sample tilt angle. A schematic of the design of the optimized decoupling structure is provided in the top right corner of panel b and consists of a hemisphere (yellow), a widening of the base into a cylinder (orange) and a cylindrical base (blue). The insets in panels a and c are zoomed-in images of regions in the lower left corner of the polymer scaffold as indicated. The zoomed-in image of the region in the inset of panel d further highlights the fidelity of the support structures after pyrolysis.

4 | Magnetic Imaging Using Magnetic Force Microscopy

MFM measurements on 3D nanostructures are challenging, which is particularly true in the case of the Pyrochlore ASI because of the high aspect ratio of the support structures. A major concern is tip strike, which can damage both the MFM tip and the structure being characterized, especially given the large height difference between the nanomagnets on the support structures and the top surface of the decoupling structure. Precise tip-position calibration is therefore carried out to ensure reliable landing of the tip, and slow scan speeds are implemented to ensure that the proportional-integral-derivative controller loop can respond if the tip encounters the edge of the 3D ASI. After acquiring a low-resolution atomic force microscopy (AFM) image to locate the 3D ASI, the scan window is refined to keep the MFM tip within the structure and enable safe acquisition of a high-resolution image.

The nanomagnets possess a nonzero out-of-plane magnetization component that enhances dipolar coupling to the MFM tip. While this facilitates interpretation of the MFM images, this tip-sample interaction increases the likelihood of switching induced by

the MFM tip, resulting in spurious contrast. To avoid this, an ultralow-moment tip (NANOSensors SSS-MFMR) was used. The low moment of the tip suppresses tip-sample interactions but also gives a reduction in the signal-to-noise ratio, making clear magnetic contrast difficult to obtain. In addition, the MFM images of the nanomagnets contain a mixture of a genuine magnetic signal and substantial topographical contamination arising from electrostatic forces. Since the latter often dominates, a least-squares fit of the AFM height map to the MFM contrast is performed, and the remaining contrast is interpreted as the magnetic contribution.

For a Pyrochlore ASI with 30 nm-thick nanomagnets, the topography obtained with AFM is shown in Figure 3a, where it can be seen that the individual nanomagnets can be resolved. We find that the root-mean-square surface roughness extracted from all measured nanomagnets is 14.9 nm. This can contribute to increased energy barriers to nanomagnet switching and a broader distribution of energy barriers, both of which can be reduced through optimization of the nanofabrication conditions. The corresponding corrected MFM data is shown in Figure 3b, where a variety of magnetic textures are revealed for MFM measurements performed on the as-deposited Pyrochlore ASI. The measurement

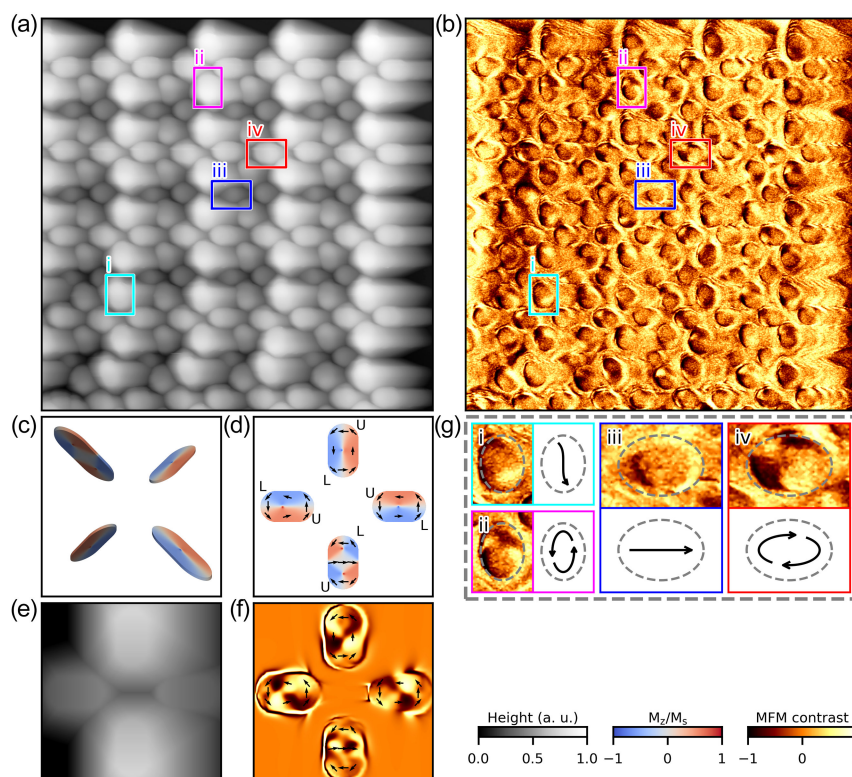


FIGURE 3 | MFM images and micromagnetic simulations of Pyrochlore ASI with 30 nm-thick permalloy nanomagnets. (a) Topography of the Pyrochlore ASI and (b) corresponding magnetic contrast with selected nanomagnets outlined to highlight the variety of observed magnetic states. The size of the scanned area is $10 \times 10 \mu\text{m}^2$. (c,d) Micromagnetic simulation of an idealized building block of the Pyrochlore ASI with four nanomagnets (c) viewed from the side and (d) viewed from the top. In panel d, it can be seen that the upper three nanomagnets are in a vortex state and the lower nanomagnet is in a double-vortex state, with arrows indicating the magnetization direction, and the letters U and L indicating the upper and lower ends of the magnets. An idealized atomic force microscopy height profile (e) is then used to generate the simulated MFM contrast shown in (f), again with arrows indicating the magnetization direction. (g) Zoomed-in MFM images of the nanomagnets indicated with boxes of the same color in panel b, together with schematics of their magnetic states.

is repeated with the tip magnetization reversed, which inverts the magnetic contrast while leaving nonmagnetic contributions unchanged, as shown in Figure S7. This enables the clear separation of magnetic signals from uncompensated electrostatic contributions or processing artifacts.

Micromagnetic simulations of the magnetic configurations at a single vertex where four nanomagnets meet are performed to help interpret the observed MFM contrast (see the Methods section for details). Since the dimensions of the experimental magnets depend on the detailed design and variations in processing conditions, we set the nanomagnets in the simulations to representative values of width $\times$ length $\times$ thickness = $200 \times 500 \times 40 \text{ nm}^3$, with the distance between the nanomagnets and the center of the vertex set to 250 nm. These dimensions deviate somewhat from those of the experimental nanomagnets ($250 \times 650 \times 30 \text{ nm}^3$). Nevertheless, since these particular simulations were only carried out to predict the MFM contrast that would result from the different possible nanomagnet states, this is of little consequence.

A simulated vertex in which three nanomagnets relax into vortex states and one relaxes into a double-vortex state is shown in Figure 3c,d. Across ten independent simulations, none of the nanomagnets relaxed into a single-domain macrospin state, indicating that nonuniform magnetization configurations are favored

for these nanomagnet dimensions. Simulated MFM contrast is typically computed at a constant lift height above the sample. However, the large variation in height of this Pyrochlore ASI produces a nontrivial imaging surface. To account for this, an AFM measurement is simulated to approximate the imaging surface (Figure 3e). Subsequently, the MFM contrast is computed at a constant height above this surface (Figure 3f).

The simulations of magnetic configurations in Figure 3f are now compared with the zoomed-in MFM images of the as-deposited magnetic states of the nanomagnets shown in Figure 3g. The locations of the nanomagnets in the Pyrochlore ASI are indicated with frames of the corresponding colors in the larger image shown in Figure 3b. Starting with the uppermost element with a long axis oriented in the vertical direction (Figure 3g,i), there is opposing bright and dark contrast at the nanomagnet ends, suggesting a near-uniform state. However, the observed bright and dark contrast is more localized along one edge, indicating some nonuniformity more characteristic of an s-state seen in thin-film planar elements [43–45]. This s-state is schematically shown to the right of the MFM image in Figure 3g,i and is typically seen for nanomagnets that are large enough for the magnetostatic energy to give rise to a curling of the magnetization but not large enough to host a fully-formed vortex state. The fact that the type of state present in the nanomagnets depends intimately on their

dimensions and saturation magnetization explains why the s-state is seen in the MFM images but not in the simulations.

The MFM contrast for the nanomagnet shown in Figure 3g,ii is indicative of a vortex state, with four lobes of alternating black and white contrast similar to the calculated contrast of the uppermost nanomagnet shown in Figure 3f, indicating a counterclockwise circulation of the magnetization. As a guide, simulated MFM contrast for nanomagnets with two opposite tilts and hosting vortices with the two different magnetization circulations is given in Figure S8. Focusing now on the MFM contrast observed in horizontal nanomagnets, one of them exhibits symmetric opposing contrast at the ends (Figure 3g,iii), suggesting a uniform single-domain state. For the nanomagnet in Figure 3g,iv, the contrast is consistent with a vortex state, but now with the magnetization circulating clockwise. The preference for flux-closed vortex states in the experimental data can be understood in the context of micromagnetic simulations as shown in Figure S9a. Here, it can be seen that, when employing the experimental nanomagnet lateral dimensions ($250 \times 650 \text{ nm}^2$) with a nanomagnet thickness of 30 nm, the double-vortex state has the lowest energy, followed closely by the single-vortex state, while the single-domain state is much higher in energy.

Overall, despite the challenges presented by the Pyrochlore ASI with its high surface topography, we have been able to reliably extract the magnetic contrast with MFM and identify multiple magnetic states. These magnetic states are often nonuniform because of the substantial nanomagnet thickness of 30 nm, which was chosen to give a strong MFM signal. We expect that a reduction in the nanomagnet thickness will give a preference for single-domain states. This is supported by our micromagnetic simulations, which demonstrate that single-domain states are favored for a permalloy nanomagnet with lateral dimensions of $250 \times 650 \text{ nm}^2$ and a thickness below $\sim 7 \text{ nm}$ (see Figure S9a). If we instead consider nanomagnets with a smaller width of 200 nm, the transition to a single-domain state occurs at a thickness of $\sim 21 \text{ nm}$ (see Figure S9b).

Our simulations demonstrate the feasibility of realizing a disconnected 3D ASI with single-domain, Ising-like macrospins. Nevertheless, nonuniform magnetic states increase the richness of magnetic states available, which has been shown to be beneficial for spin-wave reservoir computation. For example, in ref. [12], the authors demonstrate that nanomagnets with different dimensions can support different nanomagnet states, either Ising-like macrospins with strong interactions and more weakly coupled vortices. These different states display very different signatures in the spin wave spectra that can be used as an output. The enhanced microstate space gives rise to history-dependent nonlinear fading memory on application of a magnetic field cycle, which is important for reservoir computing. In ref. [46], the authors demonstrate a vortex spin-torque nano-oscillator for reservoir computing where pinning sites act as random reservoirs that can be used to emulate neuronal functions. In ref. [47], a task-adaptive reservoir computing is demonstrated in a chiral magnet where different magnetic phases display distinct resonances, and give rise to various key properties that can be tuned with magnetic field and temperature, including memory capacity, nonlinearity, and complexity.

5 | Magnetic Imaging Using Synchrotron X-ray Photoemission Electron Microscopy

The imaging of 3D ASIs using synchrotron XPEEM presents its own distinct set of challenges for sample preparation and characterization. Thick nanomagnets are not required for this surface-sensitive technique. Therefore, for these measurements, a thickness of $\sim 5 \text{ nm}$ was chosen, which should give uniform single-domain nanomagnets, as confirmed by the micromagnetic simulations shown in Figure S9a for nanomagnets with lateral dimensions of $250 \times 650 \text{ nm}^2$.

An initial XPEEM experiment was conducted at the Surface/Interface: Microscopy (SIM) beamline of the Swiss Light Source. The measurements at this beamtime revealed a critical issue—the polymerized photoresist used in the polymer scaffold had insufficient electrical conductivity, severely limiting the quality of XPEEM images. As a result, the signal-to-noise ratio was low, the image was distorted, and the magnetic contrast was difficult to perceive. An XPEEM image of the structure is shown in Figure 4a, where it is possible to see the structural outline of the Pyrochlore ASI and to deduce the location of certain four-nanomagnet vertices, with one of the vertices indicated with a blue rectangular frame. However, the detailed structure of the Pyrochlore ASI is difficult to discern. In addition, only a subset of nanomagnets (approximately half) appear in the image, which can be attributed to the distortion of the electric field by the high-aspect-ratio support structures, with a local field enhancement at the sharp tips. Also, the illumination of the sample with grazing-incidence synchrotron X-rays, with the X-ray propagation direction indicated with the blue arrow in Figure 4a, further enhanced the nonuniformity of the contrast due to shadowing. This explains why one side of the image appears brighter than the other, further impeding the interpretation of the contrast.

To address the issue of surface charging, in addition to using small decoupling structures, conductive coatings were introduced via two approaches: (i) oblique-incidence platinum sputter deposition and (ii) the growth of an iridium layer using atomic layer deposition. These coatings were applied directly onto the polymer scaffold before deposition of permalloy and successfully established a conductive path, thus mitigating charging effects. Additionally, in order to address the shadowing associated with shallow-angle X-ray illumination, a subsequent beamtime was performed at the photoemission electron microscopy end station (MAXPEEM) at the MAX IV synchrotron, where the synchrotron X-rays are at normal incidence to the sample surface. This instrument geometry, combined with a conductive coating and a high start voltage, significantly improved the image quality, eliminating both the shadowing issue and surface charging, as seen in Figure 4b. A four-nanomagnet vertex in which the individual nanomagnets can be clearly distinguished is highlighted with a blue frame. Nevertheless, some electron path distortions remained, with the nanomagnets at the right side of the Pyrochlore ASI in Figure 4b seen more clearly than those at the left side. As before, only a subset of nanomagnets appear in the image due to the distortion of the electric field by the support structures.

With the optimized imaging conditions at the MAXPEEM end station, an attempt was made to measure the magnetic contrast. For this, the magnetic state of all nanomagnets was first set by

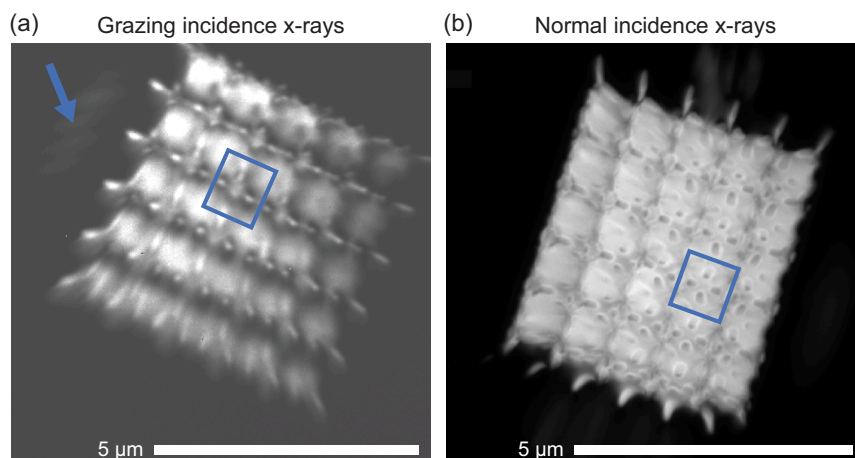


FIGURE 4 | Synchrotron XPEEM images of Pyrochlore ASI. The images are taken in single polarization mode, in which the structure of the Pyrochlore ASI is visible. The image in (a) is obtained at the Swiss Light Source. Here, the polymerized photoresist had insufficient electrical conductivity so that the signal-to-noise ratio is low and the image is distorted. In addition, the synchrotron X-rays are at a grazing incidence, impinging on the sample at an angle of 16° , with the X-ray propagation direction indicated with a blue arrow. This introduces shadowing effects and a nonuniform illumination of the sample. The image in (b) is obtained at the MAX IV synchrotron. Here, conductive coatings are introduced to mitigate charging effects, the synchrotron X-rays have normal incidence on the sample surface, which eliminates shadowing, and a high start voltage is used. This significantly improved the image quality although some electron path distortions can still be seen. The blue box in each panel outlines four nanomagnets at a vertex, which can be seen more clearly in panel b. For these samples, the permalloy thickness is ~ 5 nm.

applying an out-of-plane magnetic field of approximately $+70$ mT. The expectation was that this would give a uniform magnetic contrast across all nanomagnets so that they would appear all bright or all dark, depending on whether the applied magnetic field points into or out of the plane. The resulting X-ray magnetic circular dichroism (XMCD) contrast is shown in Figure 5a where all nanomagnets appear bright compared to the background.

As a next step, the polarity of the magnetic field was reversed, applying an out-of-plane magnetic field of approximately -70 mT to switch the magnetization direction of all nanomagnets. This should have led to an inverted magnetic contrast relative

to the previous image, with all nanomagnets appearing dark. However, the expected reversed, dark XMCD contrast of the nanomagnets was not clearly observed. Instead, the contrast associated with the nanomagnets was similar to the background (see Figure 5b).

The origin of this anomalous behavior in the contrast remains unclear—it could be an artifact associated with the imaging or something inherent in the magnetic behavior of the Pyrochlore ASI. It is unlikely to be due to X-ray-induced chemical reactions between the permalloy and the polymer scaffold, which could render the nanomagnets nonmagnetic over time. This is because

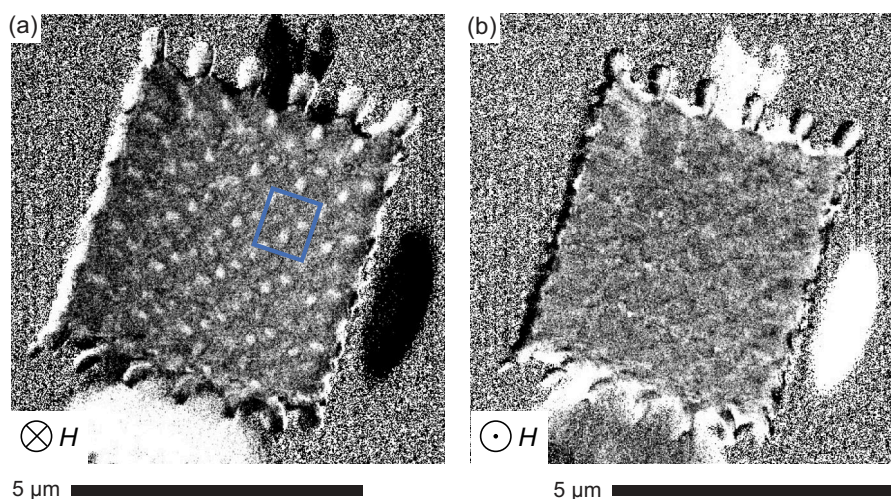


FIGURE 5 | XMCD contrast of the Pyrochlore ASI taken with synchrotron XPEEM following the application of an out-of-plane magnetic field H with the direction indicated. (a) After applying an out-of-plane magnetic field of approximately $+70$ mT, all nanomagnets exhibit bright contrast, consistent with uniform magnetization. The blue box outlines four nanomagnets at a vertex. (b) After applying an out-of-plane magnetic field of approximately -70 mT, the expected inverted dark contrast is not observed. Instead, the XMCD signal from the nanomagnets appears weak and comparable to the background. For these samples, the permalloy thickness is ~ 5 nm.

there is a conductive coating between the polymer scaffold and the nanomagnets. In addition, the bright contrast of the nanomagnets is recovered on reapplying an out-of-plane magnetic field of approximately +70 mT. Irrespective of the origin, this anomaly in magnetic imaging hindered our ability to reliably determine the magnetic state of the nanomagnets.

While the sample conductivity and the shadowing effects could be addressed, the present experiments reveal that the following issues still need to be overcome for XPEEM imaging of 3D ASIs: (i) local electric field distortion due to the 3D nature of the sample, which results in a deformation of the image, and (ii) the tilt of the nanomagnets with respect to the microscope optical axis, preventing high-resolution imaging in XPEEM. While XPEEM instruments allow mechanical rotation of the sample stage to compensate for a small sample tilt, the sample rotation is limited to only a few degrees, and, in any case, the nanomagnets have four different tilts that cannot be simultaneously compensated. With the nanomagnets tilted at 45° to the optical axis in the Pyrochlore ASI, there is a severe degradation in spatial resolution. Future developments such as vector potential photoelectron microscopy, which makes PEEM suitable for real-world samples with surface roughness [48], or novel XPEEM lens systems [49] might provide solutions for these issues.

6 | Conclusions

We have presented advances in the fabrication and characterization of a 3D ASI based on the pyrochlore lattice composed of physically separated nanomagnets. This required substantial developments in both the design of such a magnetic metamaterial and the fabrication. The fabrication of the Pyrochlore ASI consisted of two-photon lithography to fabricate the initial polymer scaffold, followed by pyrolysis to shrink the scaffold to a suitable size, and then deposition of a magnetic thin film by thermal evaporation to create the nanomagnets. The two-photon lithography process was optimized by printing multiple parallel lines for the sloped supports, starting from the center and moving outward, and by combining mechanically robust woodpile support structures with sloped platforms that have smooth surfaces. A central challenge was to maintain the fidelity of the polymer scaffold during pyrolysis while allowing the entire structure to shrink uniformly. This required the development of decoupling structures, consisting of a hemisphere with a cylindrical base, which gave mechanical isolation of the polymer scaffold from the rigid substrate.

Two complementary magnetic imaging techniques, MFM and XPEEM, were employed to characterize the magnetic configuration of the Pyrochlore ASI. With careful implementation of the MFM measurements, which included precise calibration of the position of the MFM tip and slow scan speeds, it was possible to obtain interpretable magnetic contrast and identify a range of magnetic states within the nanomagnets, including single-domain, s-, and vortex states. If single-domain nanomagnets are required, the nanomagnet volume can be reduced, either by reducing the nanomagnet thickness, lateral dimensions, or both. At the same time, if strong dipolar coupling between the

nanomagnets is required, the nanomagnets should have both a sufficiently large volume and a small separation.

A significant improvement in XPEEM imaging was obtained with the implementation of normal-incidence synchrotron X-rays and conductive coatings, to reduce shadowing and surface charging, respectively. Nevertheless, the 3D profile of the Pyrochlore ASI resulted in a nonuniform electric field that distorts the trajectory of the emitted photoelectrons, indicating the need for further developments in synchrotron X-ray imaging of such systems. One possibility is to use synchrotron X-ray laminography, which has shown promise for the magnetic imaging of small ASI building blocks [26]. Its implementation for the imaging of the magnetic configurations in Pyrochlore ASI presents its own challenges, particularly with the need to mount the sample on a membrane substrate and ensure sufficient X-ray transparency, although encouraging progress has been made in nanowire systems [50].

If single-domain nanomagnets are implemented, the magnetic states can be represented as macrospins or magnetic charge dumbbells [5]. Such a Pyrochlore ASI would allow the investigation of the motion of monopoles and the realization of a Coulomb-like phase in three dimensions [51]. In addition, we can now create a 3D ASI where the magnetic configuration is not only influenced by a magnetic field applied in the substrate plane, as is the case for 2D systems, but also with a field applied perpendicular to it. This would, for example, allow the creation of Type I vertices at will (see Figure S10 for the definition of the different vertex types) and also the investigation of the influence of the magnitude of the applied out-of-plane magnetic field on the resulting magnetization reversal on application of an in-plane magnetic field. With careful tuning of the in-plane and out-of-plane magnetic fields, the Pyrochlore ASI could be systematically modified from a 3D system to an effective 2D Kagome ice. See, for example, refs. [52] and [53].

Such a disconnected 3D ASI also offers a powerful platform to explore the physics associated with statistical vertex models [54]. To highlight this, we have used a point dipole model (see the Methods section) to determine the vertex energies as a function of the nanomagnet tilt angle (see Figure S10). At an angle of approximately 35°, Type I and Type II vertex configurations, with two macrospins pointing in and two pointing out, are degenerate as expected. Increasing the angle to 45° yields a preference for Type II vertex configurations, thus becoming more like a KDP vertex model [55]. Conversely, decreasing the angle below 35° yields a preference for Type I vertex configurations, as expected for conventional 2D square ASI and the F-model [56].

Determining the populations of vertex configurations in an ASI is a useful way to ascertain, for example, how close the system is to the ground state. However, since the nanomagnets in the Pyrochlore ASI presented here are not all single domain, there would be a myriad of magnetic states to analyze, making such an analysis impractical.

Continued progress in both fabrication and imaging will therefore be crucial for fully accessing and understanding the magnetic microstates in these architected 3D ASIs. This will, in turn, spur the discovery of novel phenomena and development of applications associated with such functional magnetic metamaterials.

7 | Methods

7.1 | 3D Artificial Spin Ice

The Pyrochlore ASIs are fabricated on $10 \times 10 \text{ mm}^2$ silicon substrates with a $\langle 100 \rangle$ orientation. The first step involves printing the polymer scaffold with two-photon lithography using the focused laser of a Nanoscribe Photonic Professional GT2 tool. The two-photon lithography is performed on an IP-Dip negative photoresist, which then serves as the polymer scaffold on which the nanomagnets are deposited. The IP-Dip negative photoresist is drop-cast on the substrate. Following laser exposure, the polymer scaffold is developed in PGMEA solvent for 15 min, rinsed in isopropanol, and critical-point dried to prevent damage from the surface tension of the isopropanol. This step is crucial in maintaining the integrity and shape of the polymer scaffold. The polymer scaffold consists of vertical support structures. On top of the support structures are platforms with stadium-shaped sloped surfaces, which are designed to host the nanomagnets at specific orientations. The sample is then subjected to pyrolysis, a heat treatment that is detailed below. This causes the polymer scaffold to shrink down to approximately one-third of its original size. This reduction in size is necessary to obtain nanomagnets that are $\sim 250 \text{ nm}$ wide and 650 nm long after pyrolysis.

The geometry of the polymer scaffold means that the magnetic material can be deposited on top, resulting in arrays of disconnected nanomagnets that are $\geq 650 \text{ nm}$ above the flat surface of the decoupling structure, which itself is also coated with a magnetic thin film. The deposited magnetic layer is permalloy, which is up to 40 nm thick, with $\text{Ni}_{83}\text{Fe}_{17}$ pellets used as the source material. However, it should be noted that, since the nanomagnets are sloped, the actual maximum thickness is then given by $40 \text{ nm} \times \cos(45^\circ) \sim 30 \text{ nm}$. The magnetic layer was deposited on top of a 10 nm -thick aluminum layer and capped with a 2 nm -thick aluminum layer to prevent oxidation. The lower aluminum layer also avoids the presence of an interfacial Dzyaloshinskii–Moriya interaction that would result from direct contact with the platinum conductive coating [57], thus simplifying interpretation of the magnetic behavior. The permalloy and aluminum layers are deposited via thermal evaporation at a base pressure of $1 \times 10^{-6} \text{ mbar}$. For the samples investigated with XPEEM, the permalloy is deposited as a wedge with the thickness going from 0 to 7 nm over a distance of $\sim 8 \text{ mm}$. The XPEEM images are taken at the thicker end of the wedge, at a permalloy thickness of $\sim 5 \text{ nm}$.

Since the nanomagnets are created with thermal evaporation, the atomic flux distribution is heavily centered around the direction perpendicular to the target material [58]. In addition, since the material is deposited at a base pressure of $1 \times 10^{-6} \text{ mbar}$, the large mean free path of evaporated atoms ($\gg 30 \text{ cm}$) means that negligible interatomic collisions take place [59]. The thermally evaporated atoms will therefore travel in a straight line, normal to the substrate, and, given that the platforms extend beyond the support structures, we expect negligible deposition of material on the support structure sidewalls. We are also confident that, with such tall support structures, the nanomagnets are not in physical contact with the magnetic thin film on the surface of the hemispherical decoupling structure.

7.2 | Printing the Polymer Scaffold With Two-Photon Lithography

The polymer scaffold of the Pyrochlore ASI is printed using two-photon lithography. This method exploits the higher probability of multiphoton absorption at the focal point of the laser beam, thereby altering the chemical properties of the photoresist in an ellipsoidal voxel. By moving the focal point of the laser beam in the photoresist, it is possible to expose arbitrary 3D shapes.

For the Nanoscribe Photonic Professional GT2 tool used for two-photon lithography, there are two main modes in which the focal point of the laser can be moved in the photoresist. The first mode keeps the focal point in a fixed position and uses a three-axis piezoelectric stage to move the sample. This mode, called the piezo mode, provides the writing of arbitrary 3D trajectories. The second mode relies on two high-speed electrically controlled mirrors (galvo mirrors) to move the laser beam and thus the focal point, in a plane parallel to the sample surface. This mode, called the galvo mode, only allows the writing of arbitrary trajectories in a plane. In galvo mode, 3D structures are therefore exposed plane by plane, exploiting the piezoelectric stage to only change the position of the focal plane with respect to the substrate. Thanks to the high-speed galvo mirrors, the galvo mode can achieve writing speeds that are 100 times faster than those of the piezo mode.

The printing process is controlled by a script, where the printing parameters are specified along with a list of points or segments that need to be printed. Alternatively, a 3D design, contained in a .stl file, can be sliced into planes that can be printed according to the specified slicing settings. However, the latter approach does not allow the user to fully tailor the printing process to the printed structure and substructures. For this reason, customized scripts were developed in order to print the various parts of the polymer scaffold in a reliable, repeatable, and tunable fashion. In order to create support structures with small lateral dimensions and high aspect ratios, the polymer scaffold is printed using the IP-Dip negative photoresist and a $\times 63$ magnification immersion objective. Among the various configurations available, this combination allows for the fabrication of the smallest printable features with the Nanoscribe Photonic Professional GT2 tool.

To achieve the best print quality, many printing parameters have been optimized for each different substructure of the polymer scaffold. These include the choice of the printing mode, the printing speed, the laser power, the laser beam acceleration in galvo mode, the trajectory fidelity in piezo mode, the settling time for each movement of the piezoelectric stage, and the settling time between each line. In addition to the optimized printing parameters, the following ensure a high repeatability of the results:

- **Waiting 10 min after each large mechanical stage movement.** Because the objective is immersed in the photoresist, every large sample movement creates unwanted polymer flow in the photoresist. These polymer currents can be detrimental for the printing process and can be minimized by implementing a waiting time after each large-stage movement.

- **Reducing the mechanical stage speed.** A smaller stage speed reduces the polymer currents generated as a result of the motion of the objective in the photoresist.
- **Calibrating the laser power multiple times during a long exposure.** The laser power of the Nanoscribe system tends to decrease over time. For this reason, the power is recalibrated every hour.
- **Cleaning the substrate surface with plasma before applying the photoresist.** This treatment is applied using a portable atmospheric air plasma pen, which produces ozone in the air. This procedure ensures better adhesion of the printed structures to the substrate surface.

7.3 | Pyrolysis

The pyrolysis process involves the thermal decomposition of polymeric or other organic materials in an inert environment. In a microfabrication context, pyrolysis can be used to reduce feature sizes and increase the strength of negative photoresist structures [60]. In the present work, pyrolysis is conducted in a nitrogen atmosphere using a small tube furnace. The nitrogen atmosphere, with a total volume of approximately 2500 cm³, is continuously refreshed from a bottle with a nitrogen purity higher than 99.999% and a flow rate of 200 sccm, ensuring a complete purge every 12.5 min.

To create the Pyrochlore ASI, pyrolysis is used to reduce the dimensions of the polymer scaffold printed using two-photon lithography. The nominal lateral dimensions of the nanomagnet platforms are 600 × 1900 nm² (see Figure S1), but, since the scanned laser voxel is at least 100 nm wide, this would give platform dimensions of 700 × 2000 nm. Shrinking of the polymer scaffold by a factor of three is therefore required to obtain nanomagnet platforms with lateral dimensions of ~250 × 650 nm² after pyrolysis. Because the final physical properties of the 3D ASI are determined by the polymer scaffold geometry, it is important that the shrinkage is isotropic, thus giving a uniform reduction in the structure dimensions that preserves the original layout defined by the polymer scaffold design. Achieving such an isotropic shrinking requires precise control of the pyrolysis heating protocol. In particular, a slow increase in temperature with a carefully controlled temperature ramp allows uniform decomposition of the polymer and controlled outgassing throughout the structure, minimizing internal stresses and shape distortions. Fast heating can cause uneven contraction and surface defects, especially in combined structures with different surface-to-volume ratios that pyrolyze and shrink at different rates. By slowly raising the temperature and including holding steps at key points, we therefore make sure that the pyrolysis at each temperature is close to completion before increasing the temperature further, thus ensuring uniform shrinkage and smooth surfaces with low roughness and porosity.

Our heating protocol, shown in Figure S3, follows these principles: the temperature is increased at a slow ramp rate with holding steps of at least 45 min every 50 °C. The longer holds at 250 and 350 °C, adapted from Bauer et al. [60], serve distinct purposes. The first hold allows thorough degassing while avoiding damage to the polymer, which retains its shape up to ~250 °C. The second

hold enables controlled and uniform volume reduction at the temperature where most of the shrinkage occurs. The maximum pyrolysis temperature is limited to 550 °C as higher temperatures lead to increased surface porosity and roughness that degrade polymer scaffold quality. This upper temperature limit results in a final polymer scaffold size that is roughly one third of the original.

It is essential to decouple the polymer scaffold from the silicon substrate, since the substrate does not appreciably change in size during pyrolysis. This is achieved by printing the polymer scaffold on cylindrical or hemispherical decoupling structures that physically isolate it from the substrate, enabling uniform 3D contraction. The most successful decoupling structure consisted of a hemisphere with an extended cylindrical structure at its base. For MFM and XPEEM, the diameter of the decoupling structures before pyrolysis was 150 μm and 50 μm, respectively, which reduced to approximately a third during pyrolysis.

7.4 | Micromagnetic Simulations

Micromagnetic simulations employing a finite element method are performed using the publicly available Nmag micromagnetic simulation code. Simulation geometries are defined using Blender and meshed to 5 nm using Netgen. Standard material parameters for permalloy for saturation magnetization ($M_s = 0.86 \times 10^6 \text{ Am}^{-1}$) and the exchange stiffness ($A_{\text{ex}} = 1.3 \times 10^{-11} \text{ Jm}^{-1}$) are implemented, with the mesh size informed by the exchange length $\lambda_{\text{ex}} = \sqrt{2A_{\text{ex}}/\mu_0 M_s^2} \approx 5 \text{ nm}$. The lowest energy configuration is computed by performing a time (t) evolution of the Landau–Lifshitz–Gilbert equation

$$\frac{dM}{dt} = -\gamma M \times H_{\text{eff}} + \frac{\alpha}{M_s} M \times \frac{dM}{dt}$$

where M is the magnetization, H_{eff} is the effective field, γ is the gyromagnetic ratio, and α represents the phenomenological damping parameter of the material. The simulations consider equilibrium states only, so that an artificially high damping parameter ($\alpha = 1$) can be used to reduce computation time.

To obtain the simulated magnetic states shown in Figures 3 and S8, each nanomagnet cell was given a random magnetization vector, and then energy minimization was achieved using a damped time-evolution method based on the numerical integration of the Landau–Lifshitz–Gilbert (LLG) equation in the absence of an external field. No magnetic field protocol was applied.

7.5 | Scanning Probe Microscopy Simulations

Outputs from micromagnetic simulations are used to compute the corresponding MFM contrast. First, an imaging surface is calculated with a binary search algorithm using two 3D surfaces representing (i) the simulation object and (ii) an MFM tip based on a Bruker MESP-V2 MFM tip that has a similar shape to the NANOSensors tips used in this work. For each in-plane coordinate, the binary search algorithm determines the smallest out-of-plane offset to be applied to the simulated tip such that the two simulation objects do not intersect. The offsets calculated for

each in-plane coordinate provide the imaging surface. The sample stray field is then calculated at 99, 100, and 101 nm above the imaging surface such that $\partial_z^2 H_z$, where H_z is the out-of-plane stray field component, can be determined numerically at 100 nm above the imaging surface.

While the fabricated nanomagnets have a 45° tilt to the substrate plane, the simulations are performed on nanomagnets that have their long axis parallel to their local $\langle 111 \rangle$ axes, which means that the nanomagnets are oriented at approximately $\pm 35^\circ$ [$\pm \arccos(2/3)^{1/2}$] to the substrate plane. We do not expect this discrepancy in the tilt angle to significantly affect the magnetic states predicted with finite-element simulations and the corresponding MFM contrast.

7.6 | Magnetic Force Microscopy

MFM measurements are performed on the Pyrochlore ASI with 30 nm-thick nanomagnets using a Bruker Dimension ICON SPM system with ultralow moment NANOSENSORS SSS-MFMR tips. Atomic force microscopy (AFM) measurements are captured in the tapping mode, and MFM images are captured on a second pass at a lift height of 100 nm. The tip is magnetized using a permanent magnet before image capture, and each image capture is repeated after reversing the tip magnetization to differentiate magnetostatic and electrostatic contributions to the contrast.

In the stadium-shaped nanomagnets, the MFM contrast is dominated by stray fields across the width of the nanomagnets. We can use this fact to deduce the circulation of the observed vortex states. With dark contrast corresponding to a positive stray-field gradient and bright contrast corresponding to a negative stray-field gradient, when the magnetization is parallel to the nanomagnet short axis, we can assign the magnetization direction to point from dark to bright contrast. This is confirmed in Figure S8 with micromagnetic simulations of individual stadium-shaped nanomagnets and the corresponding simulated magnetic force microscopy images for different combinations of nanomagnet tilt (positive or negative) and circulation of the magnetization (clockwise or counterclockwise).

7.7 | Synchrotron X-ray Photoemission Electron Microscopy

Magnetic contrast in XPEEM imaging is obtained by making use of the X-ray magnetic circular dichroic (XMCD) effect. Images are acquired with left and right circularly polarized light at the Fe L_3 edge with a photon energy of 708.1 eV. From these images, the XMCD contrast I_{XMCD} , which is proportional to the projection of the magnetization along the propagation direction of the X-rays, is calculated from

$$I_{\text{XMCD}} = \frac{I_{\text{left}} - I_{\text{right}}}{I_{\text{left}} + I_{\text{right}}}$$

The XMCD contrast follows a $\cos\theta$ dependence, where θ is the angle between the local magnetization vector and the wave vector of the helical X-rays.

Synchrotron XPEEM imaging of the Pyrochlore ASI is performed at both the Surface/Interface:Microscopy (SIM) beamline of the Swiss Light Source and the MAXPEEM beamline of the MAX IV synchrotron. At the SIM beamline of the Swiss Light Source, the X-rays impinge on the sample surface at an angle of 16°. At the MAXPEEM beamline, the incident X-rays are orthogonal to the sample surface. XMCD images using the latter arrangement are particularly sensitive to the out-of-plane component of magnetization. In addition, the normal incidence angle is useful for reducing shadowing that occurs at a lower incident angle where one part of a 3D structure prevents the X-rays from reaching another part of the structure.

The XPEEM image shown in Figure 5a,b is taken after applying a magnetic field with the variable field holder of the MAXPEEM with a current in the coil of +1 A (−1 A) corresponding to a magnetic field of approximately +70 mT (−70 mT).

The conductive coatings introduced to prevent surface charging are (i) a 5 nm-thick layer of platinum, which is sputter deposited at a base pressure of $\sim 3 \times 10^{-8}$ mbar while rotating the sample, with the angle between the source and sample of $>45^\circ$, and (ii) plasma-enhanced atomic layer deposition of a 10 nm-thick iridium coating at 370 °C. These coatings were applied directly to the polymer scaffold before depositing the aluminum/permalloy/aluminum trilayer used for the nanomagnets.

7.8 | Point Dipole Model

Vertex energies as a function of nanomagnet tilt angle were calculated using a point dipole model comprising four dipole moments with positions given in spherical polar coordinates as $(0, \theta)$, $(\pi/2, -\theta)$, (π, θ) , and $(3\pi/2, -\theta)$. Here, θ is defined as the elevation angle above/below the xy -plane.

The direction of the dipoles was set according to the appropriate vertex types, and dipolar interaction energies (U) were calculated according to

$$U = \frac{\mu_0}{4\pi} \left[\frac{\mathbf{m}_1 \cdot \mathbf{m}_2}{r^3} - \frac{3(\mathbf{m}_1 \cdot \mathbf{r})(\mathbf{m}_2 \cdot \mathbf{r})}{r^5} \right]$$

where μ_0 is the permeability of free space, and $\mathbf{m}_1$ and $\mathbf{m}_2$ are the dipole moments of two interacting point dipoles with r being the distance between them.

Author Contributions

Luca Berchialla, **Gavin M. Macauley**, and **Laura J. Heyderman** conceived the project. **Luca Berchialla**, **Arjen van den Berg**, and **Laura J. Heyderman** wrote the manuscript with contributions from all coauthors. With the support of **Sam Ladak**, **Arjen van den Berg** acquired and processed the MFM data. **Luca Berchialla**, **Gavin M. Macauley**, and **Tianyue Wang** imaged the samples with XPEEM with assistance from **Armin Kleibert**, **Evangelos Golias**, and **Yuran Niu**.

Acknowledgments

L.B., G.M.M., and L.J.H. would like to acknowledge funding from the Swiss National Science Foundation (Project No. 200020_200332). S.L.

acknowledges funding from the EPSRC (EP/L006669/1, EP/R009147) and Leverhulme Trust (RPG-2021-139). We acknowledge the MAX IV Laboratory for beamtime on the MAXPEEM beamline under proposal 20221216. Part of this work was performed at the Surface/Interface: Microscopy (SIM) beamline of the Swiss Light Source (SLS), PSI, Villigen, Switzerland. We are very grateful to Celestino Padeste for donating the oven for pyrolysis and for the technical support from the staff at the cleanroom facilities at the Paul Scherrer Institute, in particular Anja Weber. We would also like to acknowledge Peter Derlet for inspiring discussions about 3D ASIs.

Open access publishing facilitated by ETH-Bereich Forschungsanstalten, as part of the Wiley - ETH-Bereich Forschungsanstalten agreement via the Consortium Of Swiss Academic Libraries.

Funding

This work was supported by the Swiss National Science Foundation (200020_200332), EPSRC (EP/L006669/1, EP/R009147), the Leverhulme Trust (RPG-2021-139), and the MAX IV Laboratory for beamtime on the MAXPEEM beamline under proposal (20221216).

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data underlying this study will be openly available in <https://doi.org/10.5281/zenodo.18787961>.

References

1. R. F. Wang, C. Nisoli, R. S. Freitas, et al., "Artificial 'spin Ice' in a Geometrically Frustrated Lattice of Nanoscale Ferromagnetic Islands," *Nature* 439 (2006): 303–306.
2. S. H. Skjærø, C. H. Marrows, R. L. Stamps, and L. J. Heyderman, "Advances in Artificial Spin Ice," *Nature Reviews Physics* 2 (2020): 13–28.
3. L. A. Mól, R. L. Silva, R. C. Silva, A. R. Pereira, W. A. Moura-Melo, and B. V. Costa, "Magnetic Monopole and String Excitations in Two-Dimensional Spin Ice," *Journal of Applied Physics* 106 (2009): 063913.
4. S. Ladak, D. E. Read, G. K. Perkins, L. F. Cohen, and W. R. Branford, "Direct Observation of Magnetic Monopole Defects in an Artificial Spin-Ice System," *Nature Physics* 6 (2010): 359–363.
5. E. Mengotti, L. J. Heyderman, A. F. Rodríguez, F. Nolting, R. V. Hügli, and H.-B. Braun, "Real-Space Observation of Emergent Magnetic Monopoles and Associated Dirac Strings in Artificial Kagome Spin Ice," *Nature Physics* 7 (2011): 68–74.
6. L. Anghinolfi, H. Luetkens, J. Perron, et al., "Thermodynamic Phase Transitions in a Frustrated Magnetic Metamaterial," *Nature Communications* 6 (2015): 8278.
7. O. Sendetskyi, V. Scagnoli, N. Leo, et al., "Continuous Magnetic Phase Transition in Artificial Square Ice," *Physical Review B* 99 (2019): 214430.
8. I. Gilbert, Y. Y. Lao, I. Carrasquillo, et al., "Emergent Reduced Dimensionality by Vertex Frustration in Artificial Spin Ice," *Nature Physics* 12 (2016): 162.
9. S. T. Bramwell and M. J. Harris, "The History of Spin Ice," *Journal of Physics: Condensed Matter* 32 (2020): 374010.
10. C. Castelnovo, R. Moessner, and S. L. Sondhi, "Magnetic Monopoles in Spin Ice," *Nature* 451 (2008): 42–45.
11. H. Arava, P. M. Derlet, J. Vijayakumar, et al., "Computational Logic With Square Rings of Nanomagnets," *Nanotechnology* 29 (2018): 265205.
12. J. C. Gartside, K. D. Stenning, A. Vanstone, et al., "Reconfigurable Training and Reservoir Computing in an Artificial Spin-Vortex Ice via Spin-Wave Fingerprinting," *Nature Nanotechnology* 17 (2022): 460–469.
13. A. Kurenkov, J. Maes, A. Pac, et al., "Perpendicular-Anisotropy Artificial Spin Ice With Spontaneous Ordering: A Platform for Reservoir Computing With Flexible Timescales," *Communications Engineering* 4 (2025): 183.
14. A. van den Berg, M. Caruel, M. Hunt, and S. Ladak, "Combining Two-Photon Lithography With Laser Ablation of Sacrificial Layers: A Route to Isolated 3D Magnetic Nanostructures," *Nano Research* 16 (2023): 1441–1447.
15. D. Sanz-Hernández, R. F. Hamans, J.-W. Liao, A. Welbourne, R. Lavrijsen, and A. Fernández-Pacheco, "Fabrication, Detection, and Operation of a Three-Dimensional Nanomagnetic Conduit," *ACS Nano* 11 (2017): 11066–11073.
16. C. Donnelly, A. Hierro-Rodríguez, C. Abert, et al., "Complex Free-Space Magnetic Field Textures Induced by Three-Dimensional Magnetic Nanostructures," *Nature Nanotechnology* 17 (2022): 136–142.
17. M. Xu, M. Deenen A.J., H. Guo, et al., "Geometry-Induced Spin Chirality in a Non-Chiral Ferromagnet at Zero Field," *Nature Nanotechnology* 21 (2026): 58.
18. H. Guo, A. J. Deenen, M. Xu, M. Hamdi, and D. Grundler, "Realization and Control of Bulk and Surface Modes in 3D Nanomagnonic Networks by Additive Manufacturing of Ferromagnets," *Advanced Materials* 35 (2023): 2303292.
19. L. Berchialla, G. M. Macauley, and L. J. Heyderman, "Focus on Three-Dimensional Artificial Spin Ice," *Applied Physics Letters* 125 (2024): 220501.
20. C. Donnelly, M. Guizar-Sicairos, V. Scagnoli, et al., "Element-Specific X-Ray Phase Tomography of 3D Structures at the Nanoscale," *Physical Review Letters* 114 (2015): 115501.
21. G. M. Macauley, L. Berchialla, P. M. Derlet, and L. J. Heyderman, "Five-Stage Ordering to a Topological-Defect-Mediated Ground State in a Buckyball Artificial Spin Ice," *Communications Physics* 9 (2026): 288.
22. R. Cheenikundil and R. Hertel, "Switchable Magnetic Frustration in Buckyball Nanoarchitectures," *Applied Physics Letters* 118 (2021): 212403.
23. R. Cheenikundil, J. Bauer, M. Goharyan, M. d'Aquino, and R. Hertel, "High-Frequency Modes in a Magnetic Buckyball Nanoarchitecture," *APL Materials* 10 (2022): 081106.
24. A. May, M. Hunt, A. Van Den Berg, A. Hejazi, and S. Ladak, "Realisation of a Frustrated 3D Magnetic Nanowire Lattice," *Communications Physics* 2 (2019): 13.
25. G. Williams, M. Hunt, B. Boehm, et al., "Two-Photon Lithography for 3D Magnetic Nanostructure Fabrication," *Nano Research* 11 (2018): 845–854.
26. P. Pip, S. Treves, J. R. Massey, et al., "X-Ray Imaging of the Magnetic Configuration of a Three-Dimensional Artificial Spin Ice Building Block," *APL Materials* 10 (2022): 101101.
27. A. May, M. Saccone, A. van den Berg, J. Askey, M. Hunt, and S. Ladak, "Magnetic Charge Propagation Upon a 3D Artificial Spin-Ice," *Nature Communications* 12 (2021): 3217.
28. M. Saccone, A. Van den Berg, E. Harding, et al., "Exploring the Phase Diagram of 3D Artificial Spin-Ice," *Communications Physics* 6 (2023): 217.
29. A. van den Berg, P. Rickhaus, F. Barrows, C. Nisoli, and S. Ladak, "Direct Visualization of the Magnetic Monopole Field in a 3D Artificial Spin Ice," arXiv: 2511.04877 (2025).
30. S. Sahoo, A. May, A. van Den Berg, A. K. Mondal, S. Ladak, and A. Barman, "Observation of Coherent Spin Waves in a Three-Dimensional Artificial Spin Ice Structure," *Nano Letters* 21 (2021): 4629–4635.

31. M. Gołębiewski, H. Reshetniak, U. Makartsou, et al., "Spin-Wave Spectral Analysis in Crescent-Shaped Ferromagnetic Nanorods," *Physical Review Applied* 19 (2023): 064045.
32. I. B. Yildiz, H. Jaeger, and S. J. Kiebel, "Re-Visiting the Echo State Property," *Neural Networks* 35 (2012): 1–9.
33. A. Mistonov, I. Dubitskiy, I. Shishkin, et al., "Magnetic Structure of the Inverse Opal-Like Structures: Small Angle Neutron Diffraction and Micromagnetic Simulations," *Journal of Magnetism and Magnetic Materials* 477 (2019): 99–108.
34. J. Llandro, D. M. Love, A. Kovács, et al., "Visualizing Magnetic Structure in 3D Nanoscale Ni-Fe Gyroid Networks," *Nano Letters* 20 (2020): 3642–3650.
35. C. Schröder, B. Ghebretinsae, M. Lonsky, et al., "Origin of Magnetic Switching Cascades in Tetrahedral CoFe Nanostructures," *Applied Physics Letters* 126 (2025): 222402.
36. O. M. Volkov, O. V. Pylypovskiy, F. Porrati, et al., "Three-Dimensional Magnetic Nanotextures With High-Order Vorticity in Soft Magnetic Wireframes," *Nature Communications* 15 (2024): 2193.
37. F. Meng, C. Donnelly, C. Abert, et al., "Non-Planar Geometrical Effects on the Magnetoelectrical Signal in a Three-Dimensional Nanomagnetic Circuit," *ACS Nano* 15 (2021): 6765–6773.
38. L. Skoric, C. Donnelly, A. Hierro-Rodriguez, et al., "Domain Wall Automation in Three-Dimensional Magnetic Helical Interconnectors," *ACS Nano* 16 (2022): 8860–8868.
39. D. D. Sheka, "A Perspective on Curvilinear Magnetism," *Applied Physics Letters* 118 (2021): 230502.
40. D. Makarov, O. M. Volkov, A. Kákay, O. V. Pylypovskiy, B. Budinská, and O. V. Dobrovolskiy, "New Dimension in Magnetism and Superconductivity: 3D and Curvilinear Nanoarchitectures," *Advanced Materials* 34 (2022): 2101758.
41. J. Drisko, T. Marsh, and J. Cumings, "Topological Frustration of Artificial Spin Ice," *Nature Communications* 8 (2017): 14009.
42. R. Puttock, I. M. Andersen, C. Gatel, et al., "Defect-Induced Monopole Injection and Manipulation in Artificial Spin Ice," *Nature Communications* 13 (2022): 3641.
43. W. Rave and A. Hubert, "Magnetic Ground State of a Thin-Film Element," *IEEE Transactions on Magnetics* 36 (2000): 3886–3899.
44. R. Hertel, "Thickness Dependence of Magnetization Structures in Thin Permalloy Rectangles," *Zeitschrift für Metallkunde* 93 (2002): 957–962.
45. L. J. Heyderman, S. Czekaj, F. Nolting, et al., "Photoemission Electron Microscopy Study of Remanent Magnetic Domain States in Ferromagnetic Wedge Films Deposited on Substrates With Micrometer-Sized Square Plateaus," *Journal of Applied Physics* 99 (2006): 063904.
46. S. Shreya, A. S. Jenkins, Y. Rezaeiyan, et al., "Granular Vortex Spin-Torque Nano Oscillator for Reservoir Computing," *Scientific Reports* 13 (2023): 16722.
47. O. Lee, T. Wei, K. D. Stenning, et al., "Task-Adaptive Physical Reservoir Computing," *Nature Materials* 23 (2024): 79–87.
48. R. Browning, "Vector Potential Photoelectron Microscopy," *Review of Scientific Instruments* 82 (2011): 103703.
49. O. Tkach and G. Schönhense, "Multimode Objective Lens for Momentum Microscopy and XPEEM: Theory," *Ultramicroscopy* 276 (2025): 114167.
50. E. Harding, T. Araki, J. Askey, et al., "Imaging the Magnetic Nanowire Cross Section and Magnetic Ordering Within a Suspended 3D Artificial Spin-Ice," *APL Materials* 12 (2024): 021116.
51. C. L. Henley, "The "Coulomb Phase" in Frustrated Systems," *Annual Review of Condensed Matter Physics* 1 (2010): 179–210.
52. Y. Tabata, H. Kadowaki, K. Matsuhira, et al., "Kagomé Ice State in the Dipolar Spin Ice $\text{Dy}_2\text{Ti}_2\text{O}_7$," *Physical Review Letters* 97 (2006): 257205.
53. T. Fennell, S. T. Bramwell, D. F. McMorrow, P. Manuel, and A. R. Wildes, "Pinch Points and Kasteleyn Transitions in Kagome Ice," *Nature Physics* 3 (2007): 566–572.
54. R. J. Baxter, *Exactly Solved Models in Statistical Mechanics* (Academic Press, 1982).
55. E. H. Lieb, "Exact Solution of 2-Dimensional Slater KDP Model of a Ferroelectric," *Physical Review Letters* 19 (1967): 108.
56. E. H. Lieb, "Exact Solution of F Model of an Antiferroelectric," *Physical Review Letters* 18 (1967): 1046.
57. K. Hofhuis, A. Hrabec, H. Arava, et al., "Thermally Superactive Artificial Kagome Spin Ice Structures Obtained With the Interfacial Dzyaloshinskii-Moriya Interaction," *Physical Review B* 102 (2020): 180405.
58. T. Karabacak, "Thin-Film Growth Dynamics With Shadowing and Re-Emission Effects," *Journal of Nanophotonics* 5 (2011): 052501.
59. Z. Cui, *Nanofabrication Principles, Capabilities and Limits* (Springer, 2024).
60. J. Bauer, A. Schroer, R. Schwaiger, and O. Kraft, "Approaching Theoretical Strength in Glassy Carbon Nanolattices," *Nature Materials* 15 (2016): 438.

Supporting Information

Additional supporting information can be found online in the Supporting Information section. This movie highlights the geometry of the Pyrochlore ASI. At the start of the movie, a top-down view of the Pyrochlore ASI is shown with four different colors indicating the nanomagnets at four different heights. The colors are then removed, and the design is rotated to display the Pyrochlore ASI from different perspectives. At the end of the movie, four nanomagnets are shown in pink, once again to highlight the four different nanomagnet heights.