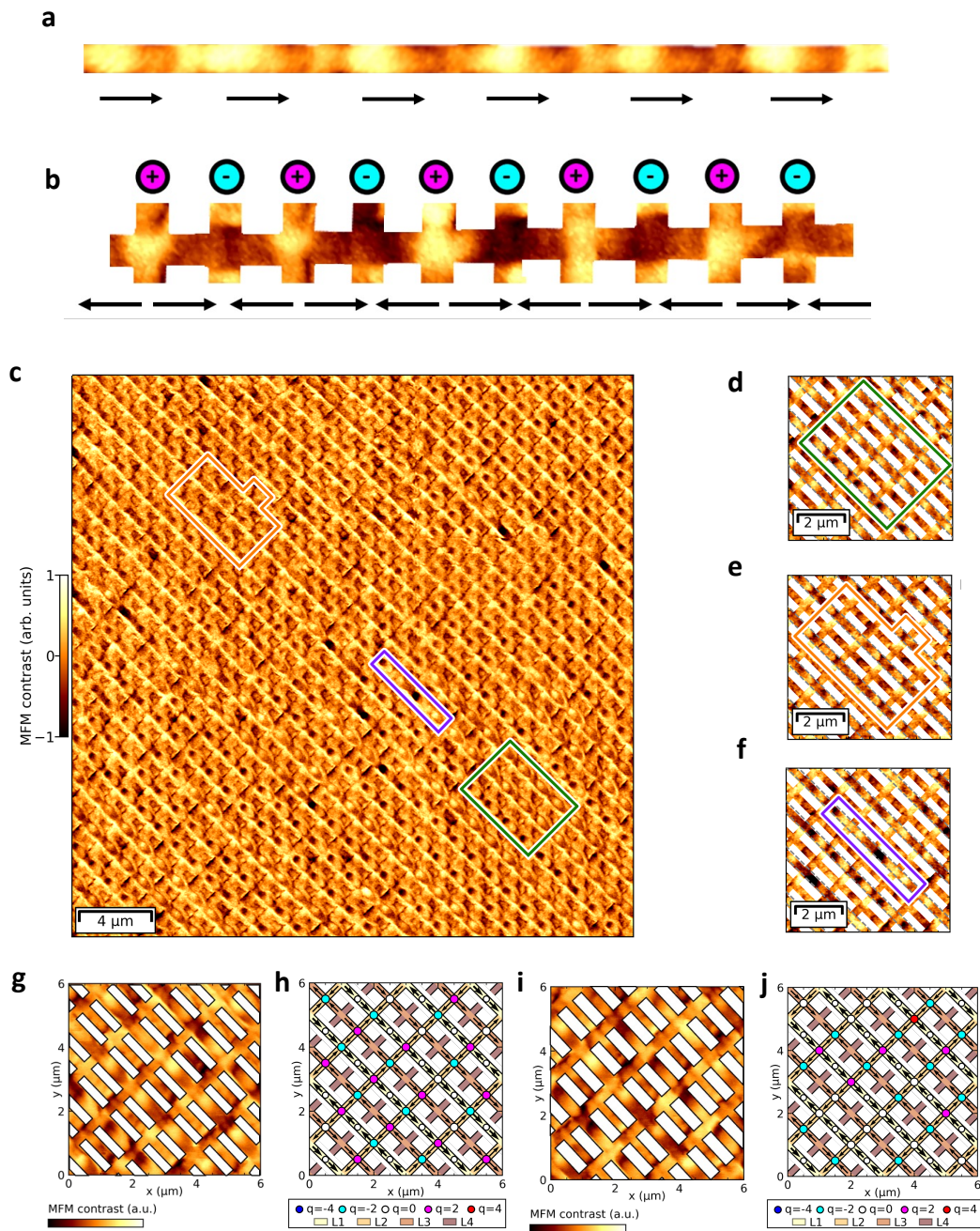


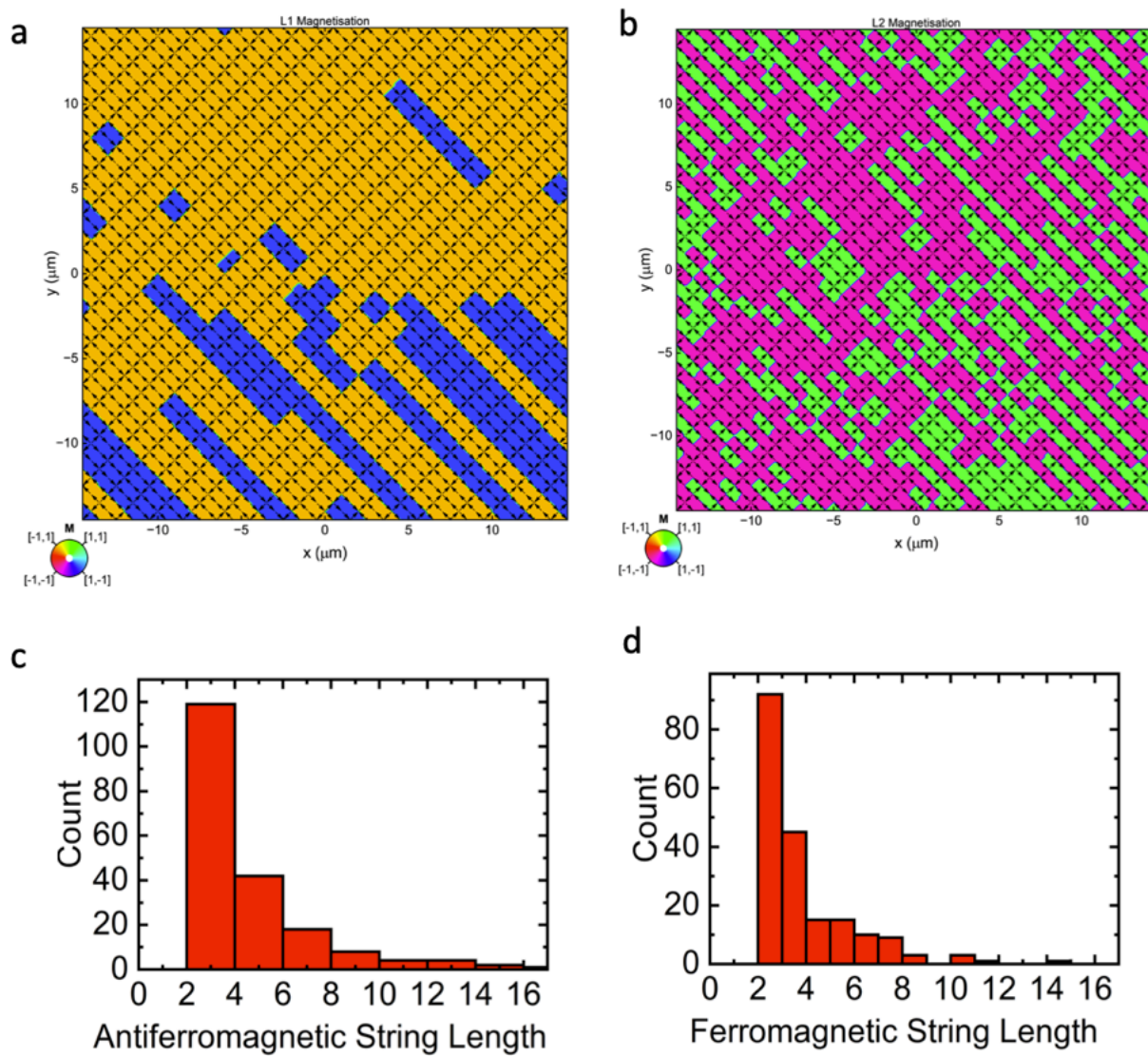
Exploring the phase diagram of 3D artificial spin-ice

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and Sam Ladak^{2,†}

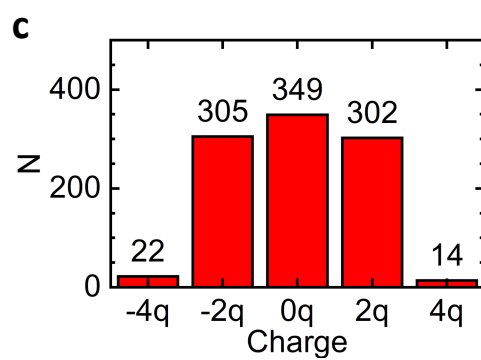
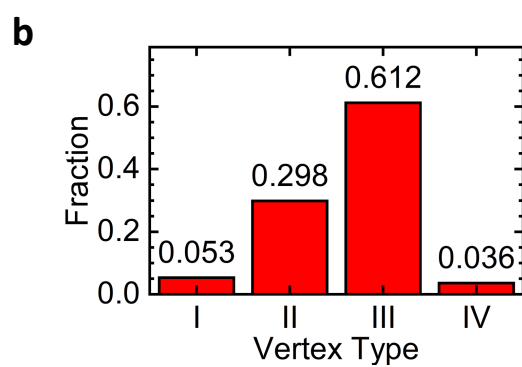
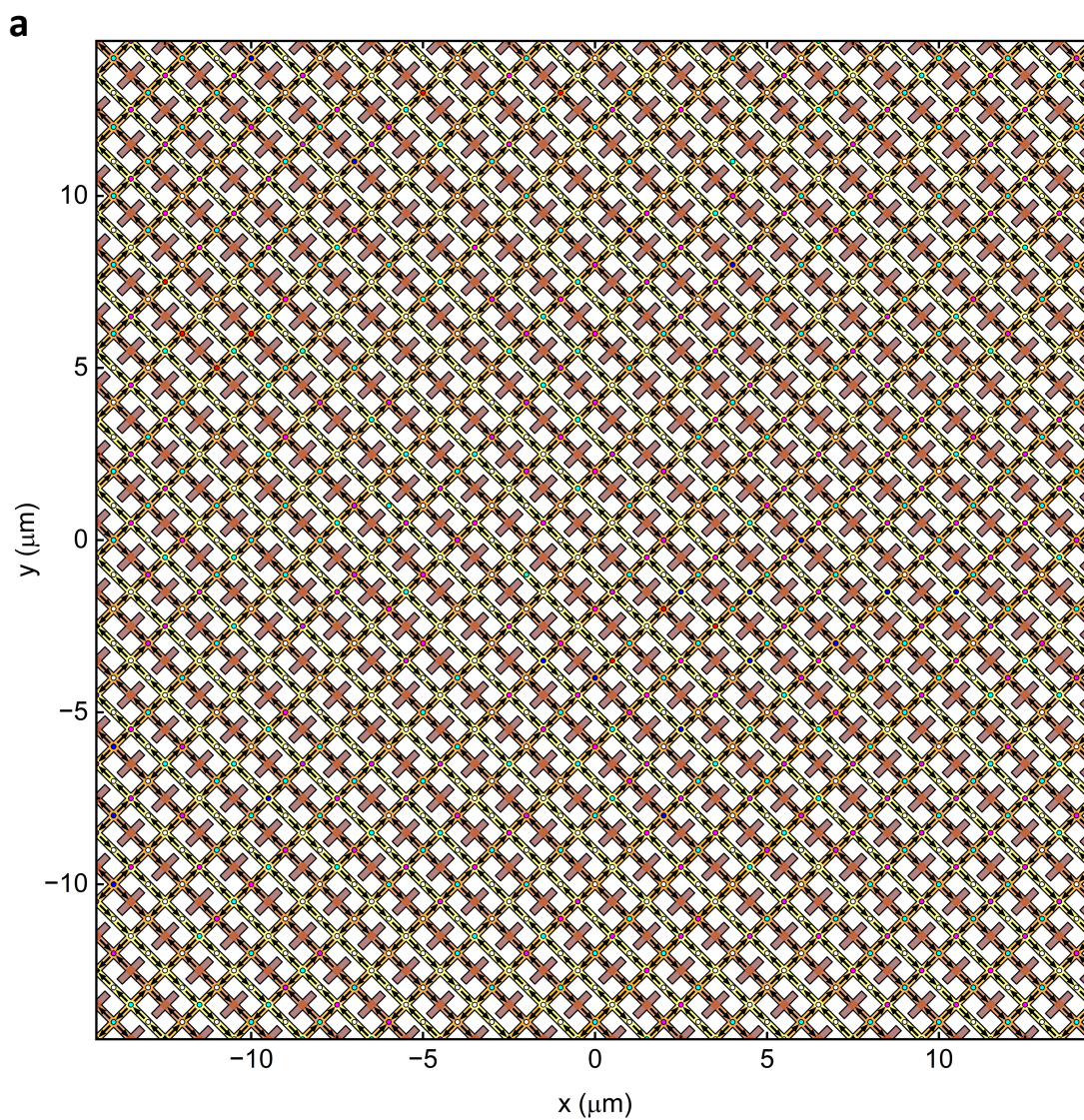
1. Center for Nonlinear Studies and Theoretical Division, Los Alamos National Laboratory, Los Alamos, NM, USA.
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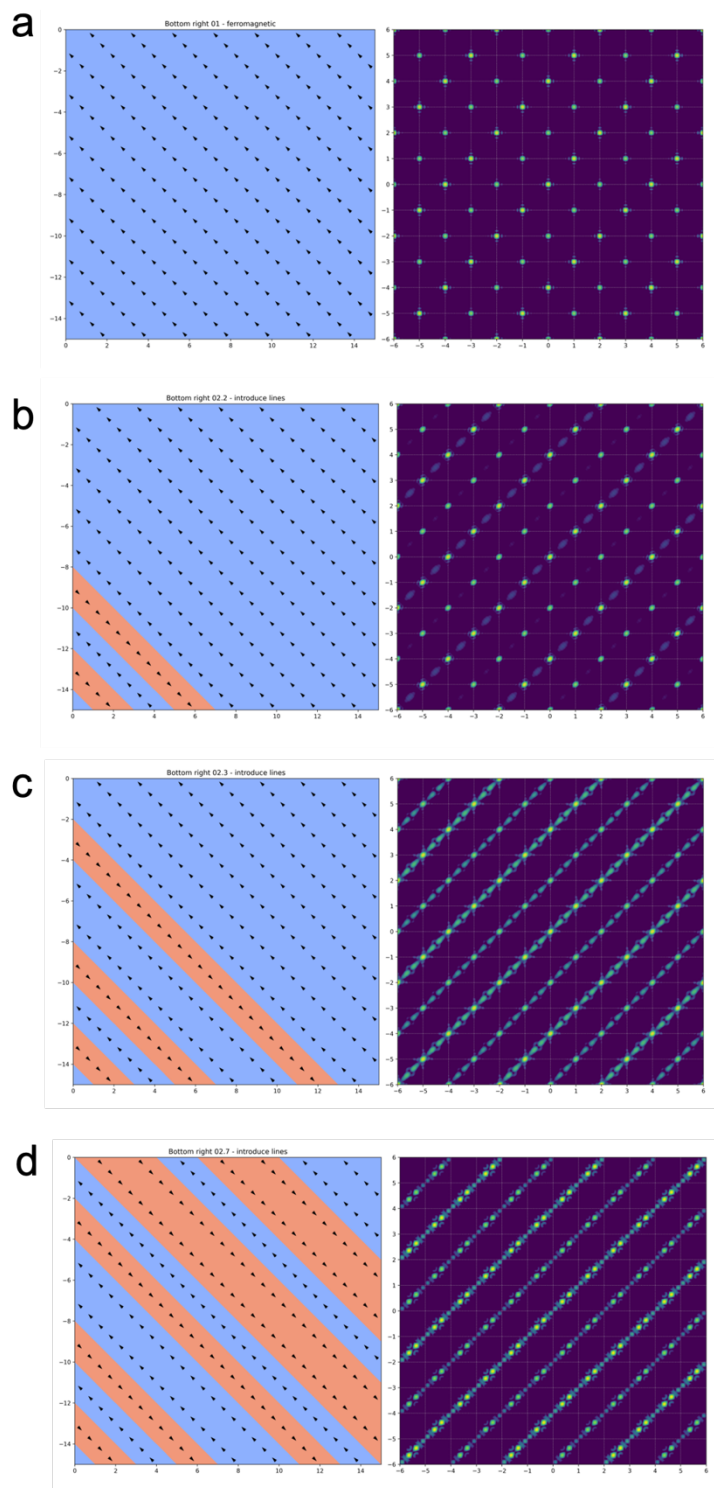
Supplementary Figure 1: (a) Magnetic force microscopy (MFM) image showing typical ferromagnetic ordering observed on the L1 sub-lattice. Arrows indicate direction of magnetisation. (b) MFM image showing antiferromagnetic ordering observed within magnetic charge crystallites on L2. Arrows indicate magnetisation and vertex charges are shown above the image. (c) Large area MFM image of a 3DASI sample. (d) MFM image of an area showing magnetic charge crystallite, (e) the ice phase and (f) a small region showing the double magnetic charge phase. (g-j) Further charge crystallite areas from other nominally identical samples, with associated arrow maps.



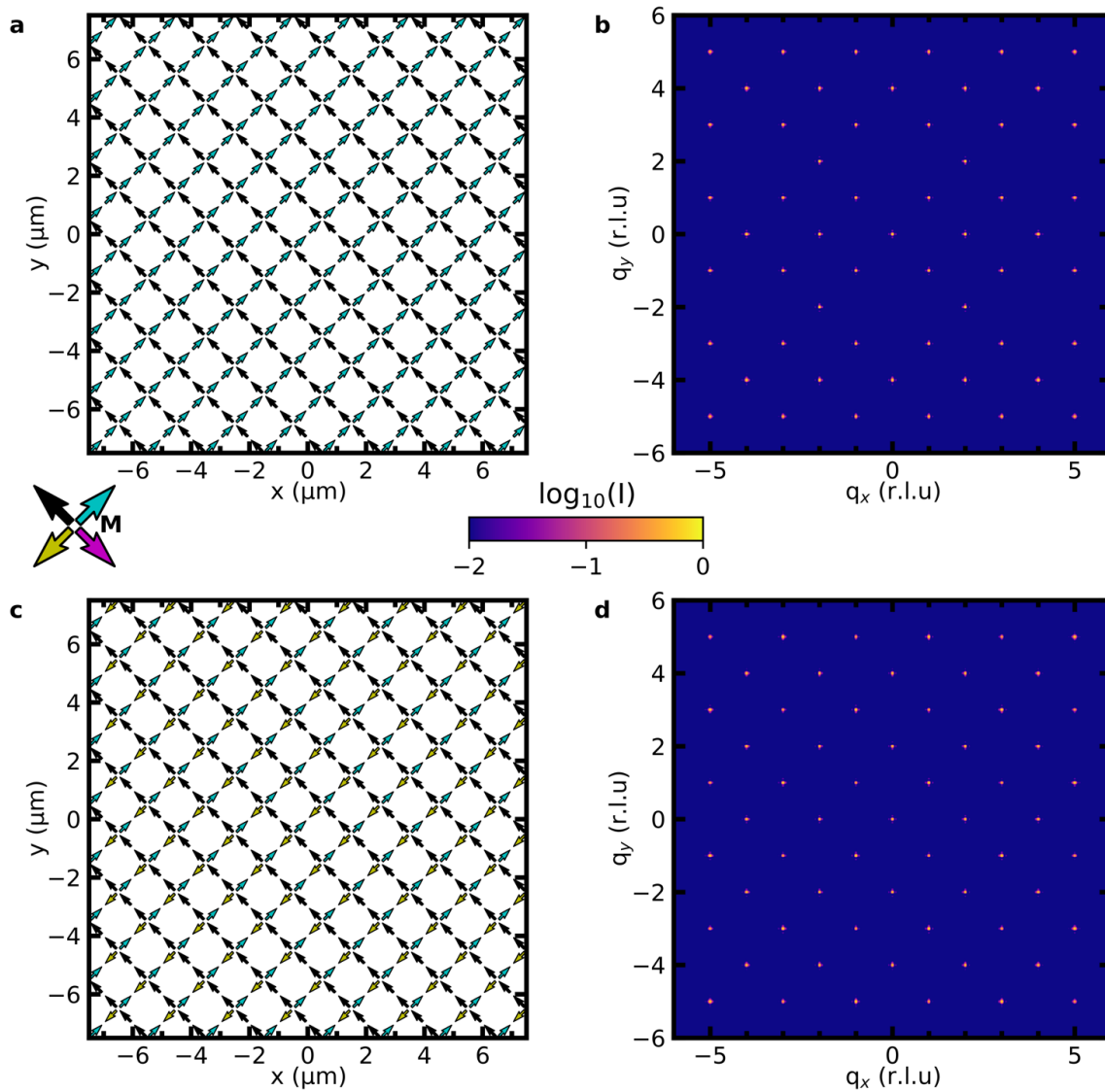
Supplementary Figure 2: (a) Arrow map showing the magnetisation upon the L1 sub-lattice, which is seen to largely be composed of long ferromagnetic stripes. (b) Arrow map showing the magnetisation upon the L2 sub-lattice. (c) Histogram of L2 antiferromagnetic string frequency. (d) Histogram of L2 ferromagnetic string frequency.



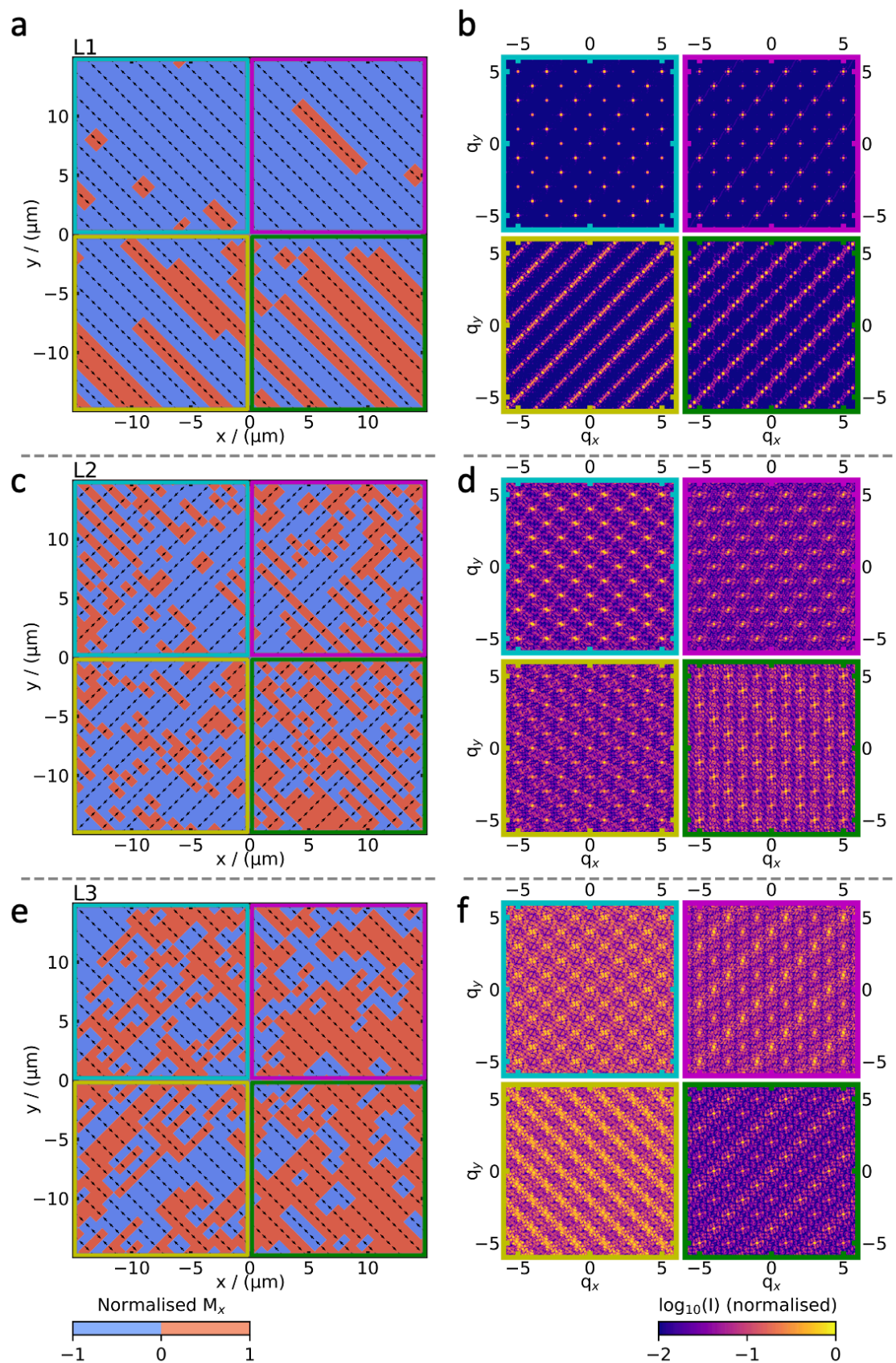
Supplementary Figure 3: (a) Full magnetisation map of the $30\ \mu\text{m} \times 30\ \mu\text{m}$ sample area measured by magnetic force microscopy. (b) Vertex populations for the measured area. Coordination two vertices are not included. (c) Distribution of vertex charge across the measured area.



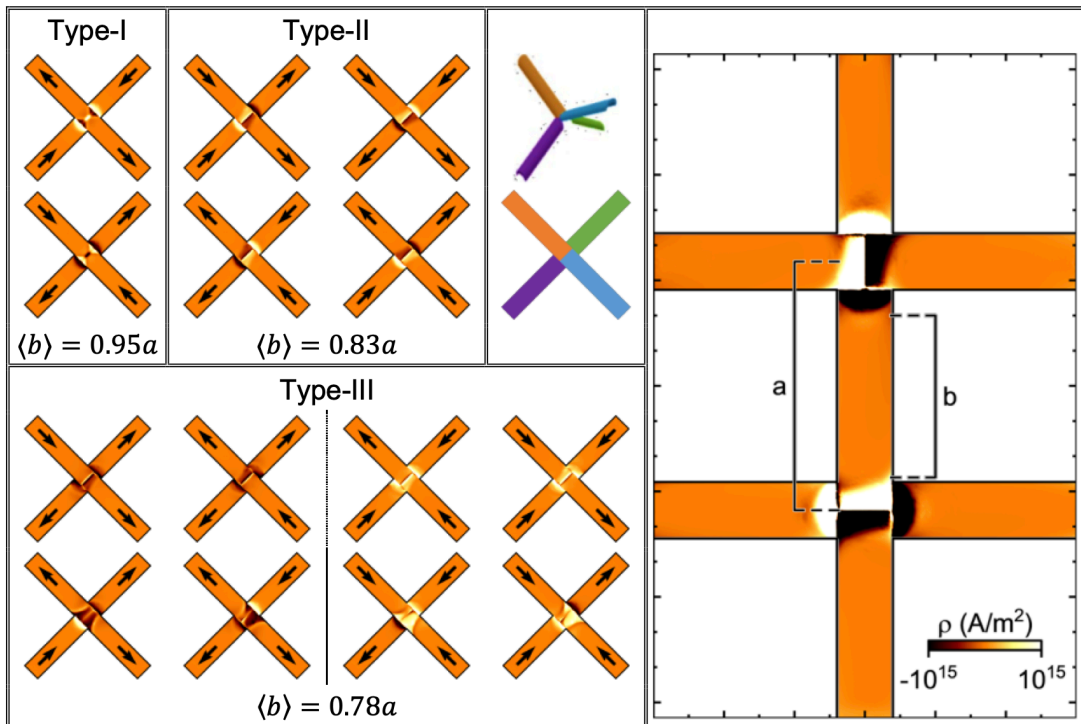
Supplementary Figure 4: Evolution of L1 structure factor, starting with **(a)** perfect ferromagnetic order and then **(b-d)** implementing domains seen in experimental arrow map.



Supplementary Figure 5: Arrow map and magnetic structure factor for (a,b) uniformly tiled type II vertices and for (c,d) a perfectly ordered experimental charge crystal (C_6I).



Supplementary Figure 6: Spin maps and associated structure factor for experimental data, separated by region and layer. (a) Arrow map for the L1 sub-lattice with (b) the associated structure factor. (c) Arrow map for the L2 sub-lattice with (d) the associated structure factor. (e) Arrow map for the L3 sub-lattice with (f) the associated structure factor. For each sub-lattice, regions as identified by square outline have corresponding structure factors as shown in associated panel.

a**b**

Vertex	Type I	Type II	Type III
b/a	0.95	0.83	0.78

Supplementary Figure 7: (a) Magnetic charge density for vertex types computed using the NMag micromagnetic code. The lattice constant, a , is measured from vertex centre to vertex centre. The needle length, b , is measured across the region where the magnetisation is uniformly magnetised. Colour contrast shows the magnetic charge density as indicated in legend. **(b)** Table showing scaled dipolar needle length for range of vertices.