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Interaction modifiers in artificial spin ices

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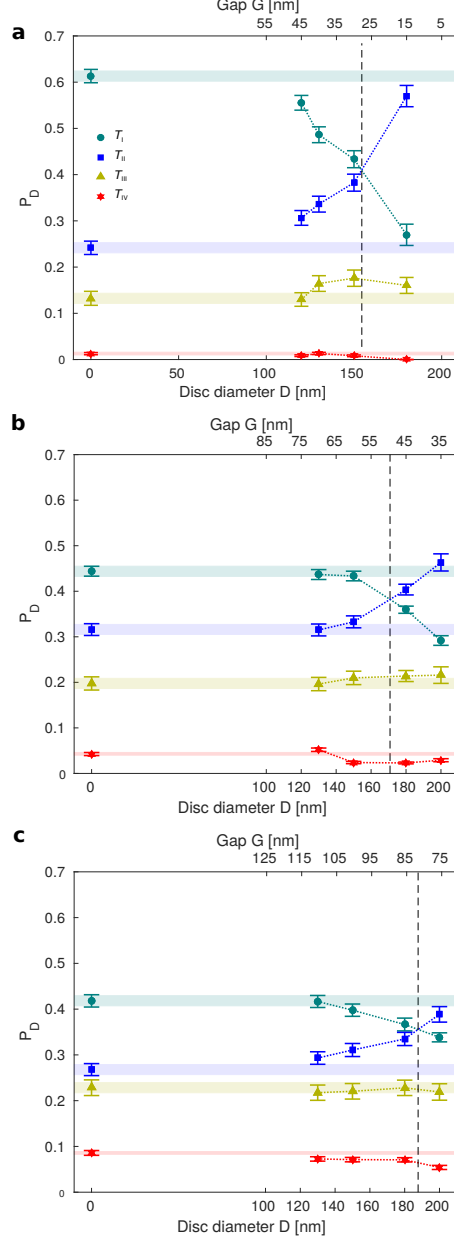
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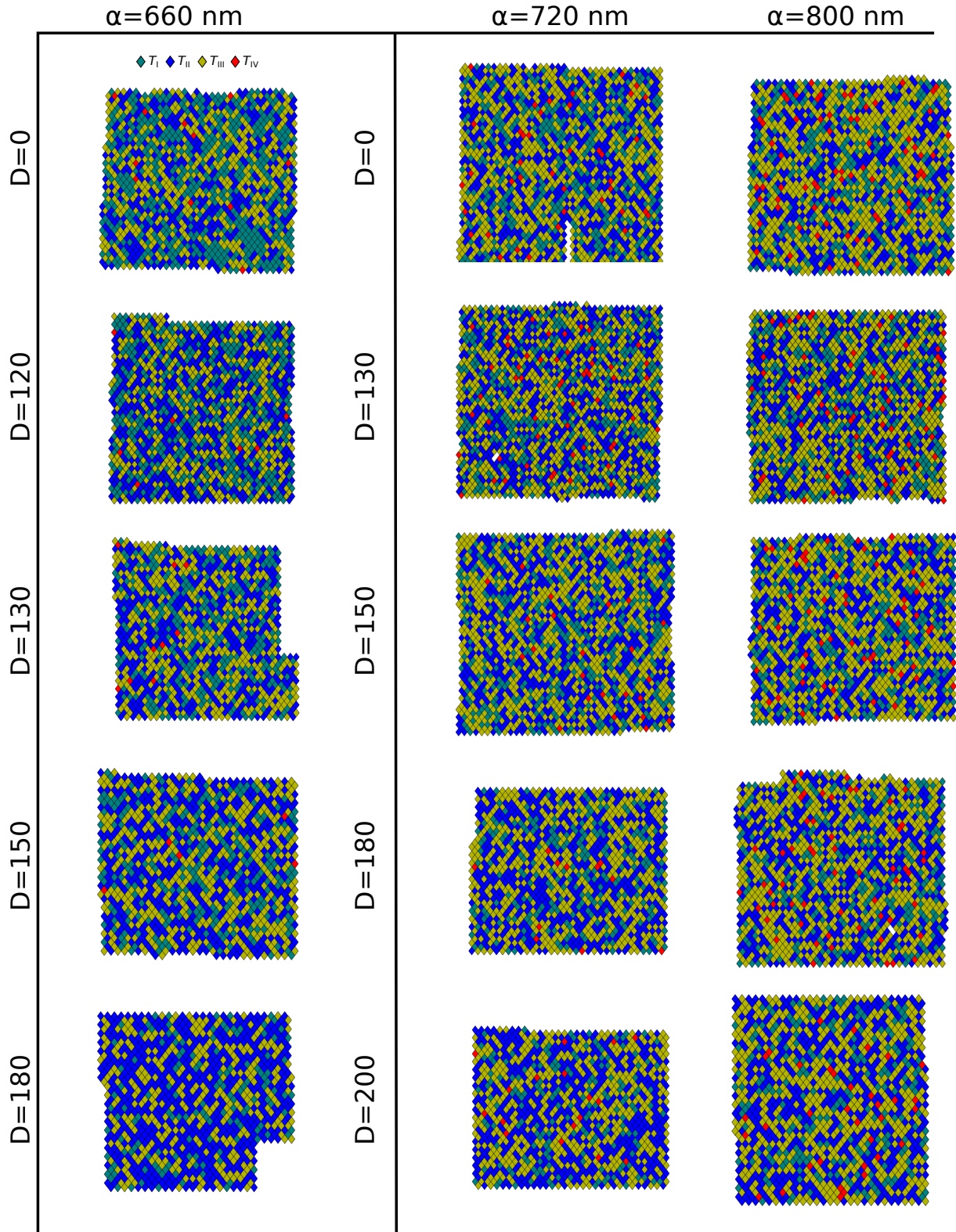
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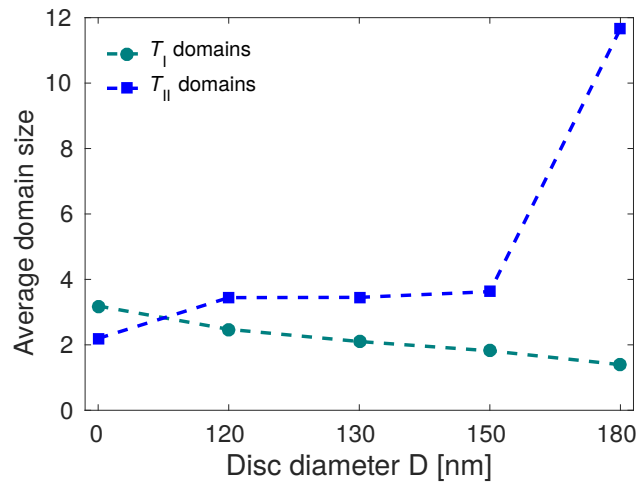
(Dated: January 17, 2018)



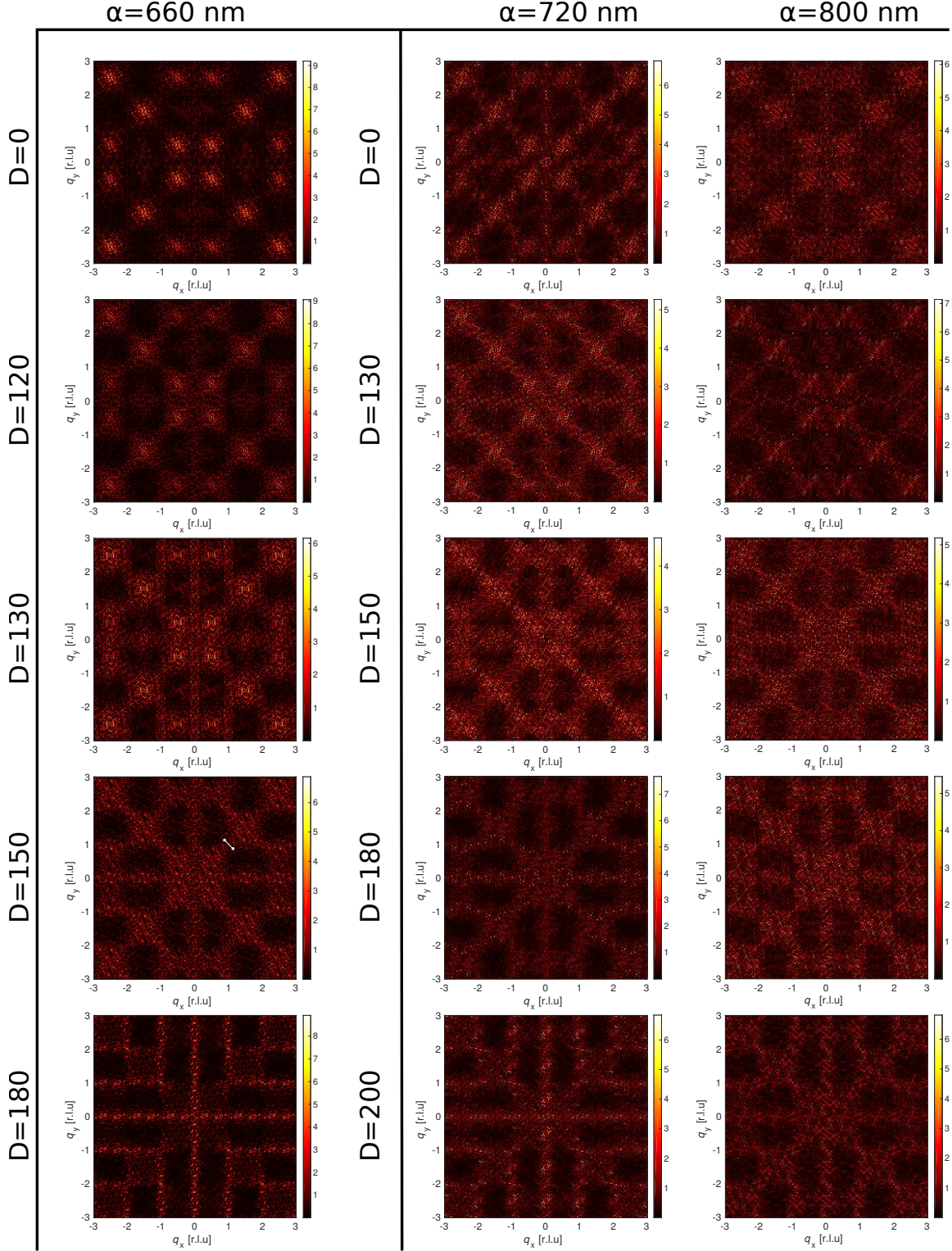
Supplementary Figure 1. Degeneracy corrected and normalized vertex populations for $\alpha = 660$ nm **a**, $\alpha = 720$ nm **b**, and $\alpha = 800$ nm **c**. Vertical dashed lines indicate the intersection between T_I and T_{II} vertices drawing straight lines between the data points. The horizontal colored stripes refer to the populations for $D=0$ and serve as a guide to the eye along with the dotted lines, highlighting the vertex population changes while varying D . All three groups with different lattice parameters undergo a transition from a majority of T_I vertices to a majority of T_{II} vertices with an increase in disc diameter. The increase in lattice parameter decreases the coupling strength between the interacting elements evident from the increase of charges for the $\alpha = 720$ nm and the $\alpha = 800$ nm lattices. All error bars represent one standard deviation.



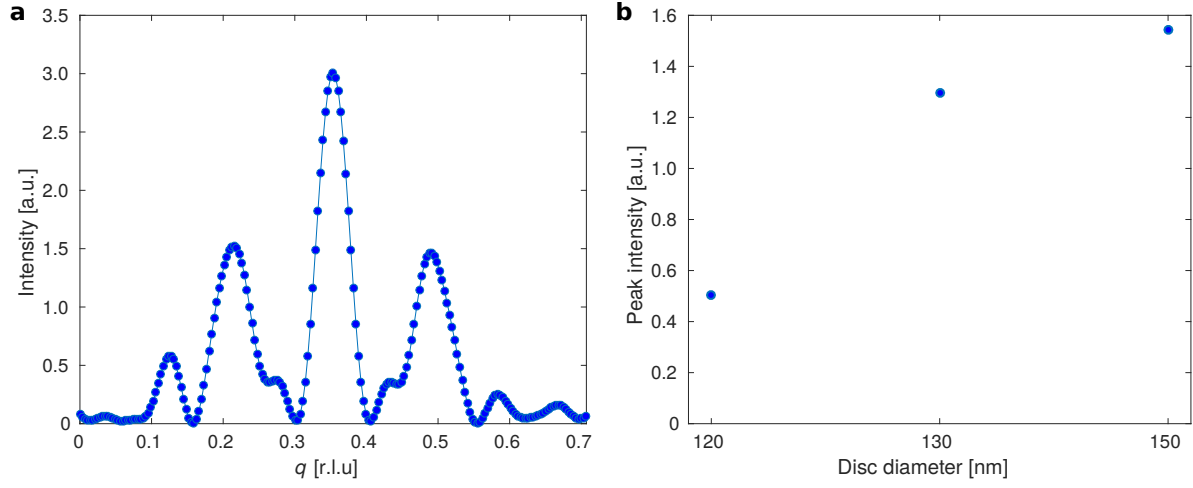
Supplementary Figure 2. Vertex maps for all measured arrays. The lattice parameter, α is defined for each vertical line, the diameter, D , is defined for every lattice horizontally.



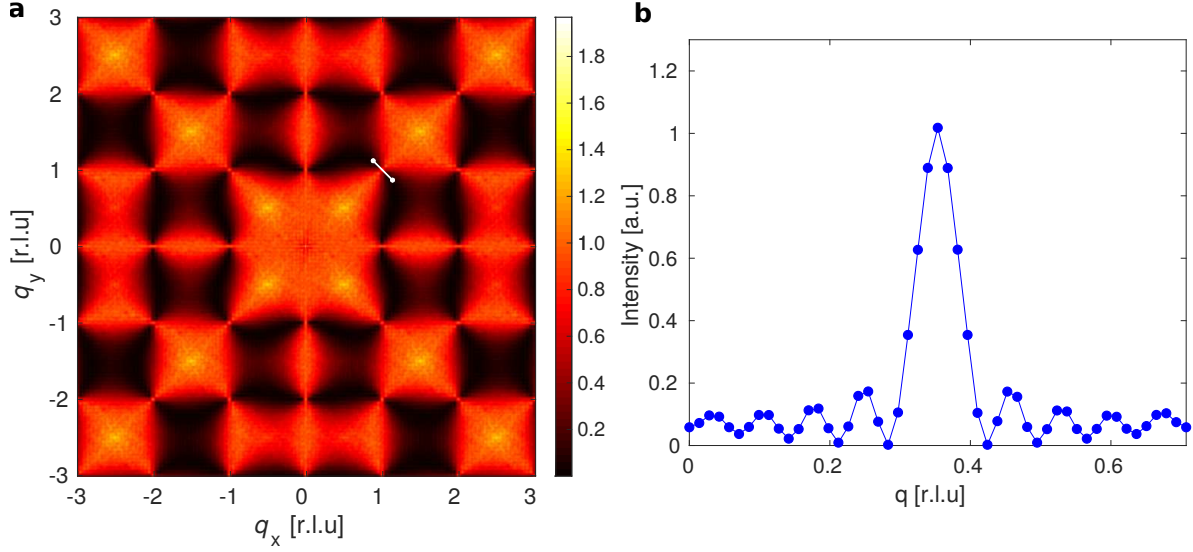
Supplementary Figure 3. Average domain size evolution for T_I and T_{II} vertices with increasing disc diameter.



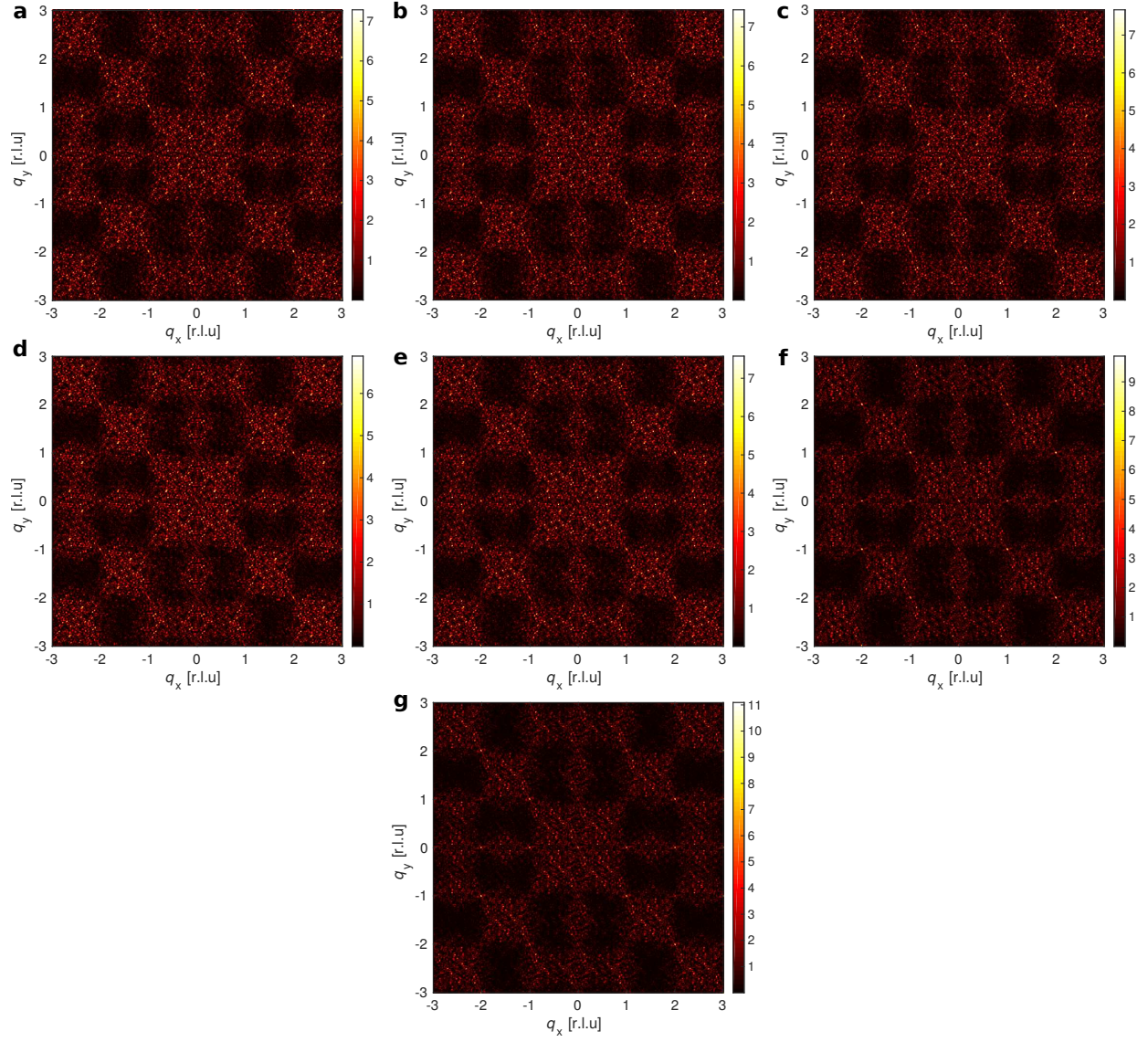
Supplementary Figure 4. Magnetic spin structure factor for all measured arrays. The white line in $\alpha = 660 \text{ nm}$ $D = 150 \text{ nm}$ illustrates the cut in reciprocal space where from the pinch point data in Supplementary Fig. 5 a is taken.



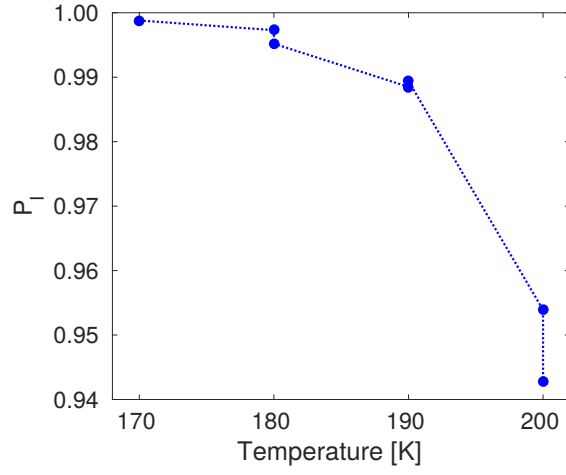
Supplementary Figure 5. **a**, Intensity profile of the $\alpha = 660$ nm, $D=150$ nm array going from $[3/4, 5/4]$ to $[5/4, 3/4]$ r.l.u passing through the pinch point at $[1,1]$ r.l.u in reciprocal space, see Supplementary Fig. 4. **b**, Evolution of peak intensity values at the pinch points while increasing the disc diameter. Peak values are averaged over all four positions $[\pm 1, \pm 1]$ r.l.u.



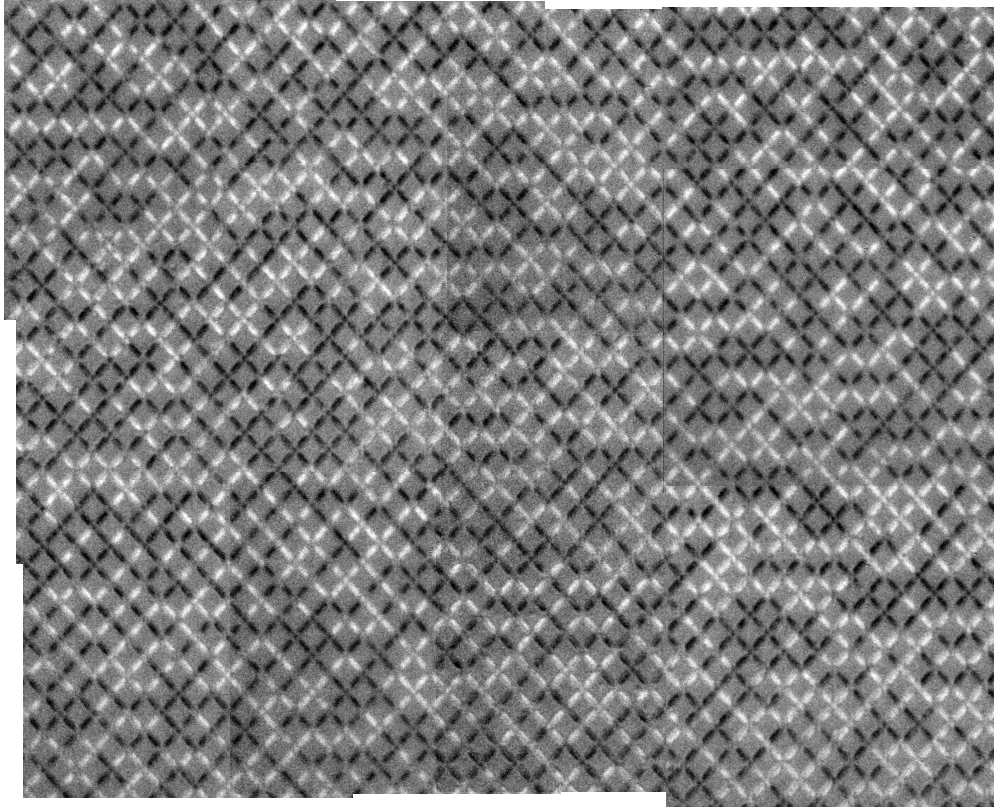
Supplementary Figure 6. **a**, Computed magnetic spin structure factor averaged over 1000 de-correlated spin configurations satisfying the ice rule. In order to calculate a theoretical spin map for a fully compensated lattice we start with a spin configuration containing $N=800$ spins polarized diagonally with respect to the major axis of the square lattice and employ the standard loop algorithm¹⁻³ using periodic boundary conditions. To fully de-correlate the spin configurations 30N loops are flipped. For each loop the first spin is chosen at random in the lattice, the next spin is chosen from one of the six neighbors at random with the condition that the spins in the loop must align ferromagnetically after the spin reversal. As soon as a closed loop is found all spins contained within the loop reverses and a new loop is started. The loops can either be closed, contained within the lattice, or open, wrapped around the lattice. In this way all vertices will obey the ice rule and only the ice rule manifold will be explored. **b**, Intensity profile over $[3/4, 5/4]$ to $[5/4, 3/4]$ r.l.u. passing through the pinch point at $[1,1]$ r.l.u., as indicated by the white line in (a).



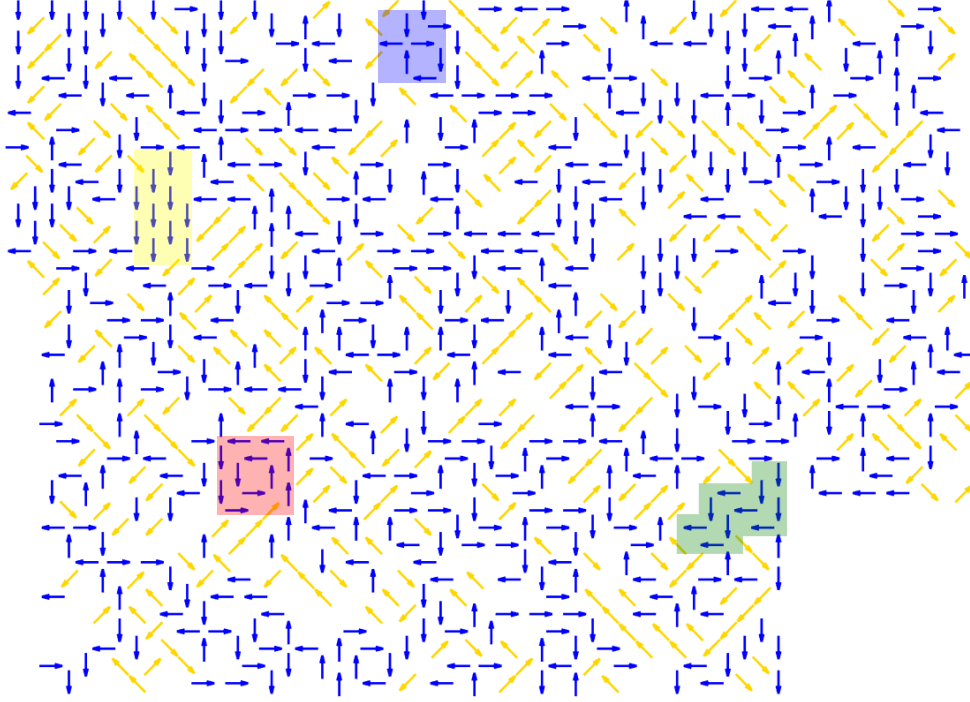
Supplementary Figure 7. Computed magnetic spin structure factor for the time-temperature series. **a**, corresponds to 170 K $t=t_0$, **b**, 180 K $t=1047$ s, **c**, 180 K $t=1531$ s, **d**, 190 K $t=2371$ s, **e**, 190 K $t=2864$ s, **f**, 200 K $t=3753$ s, **g**, 200 K $t=4242$ s.



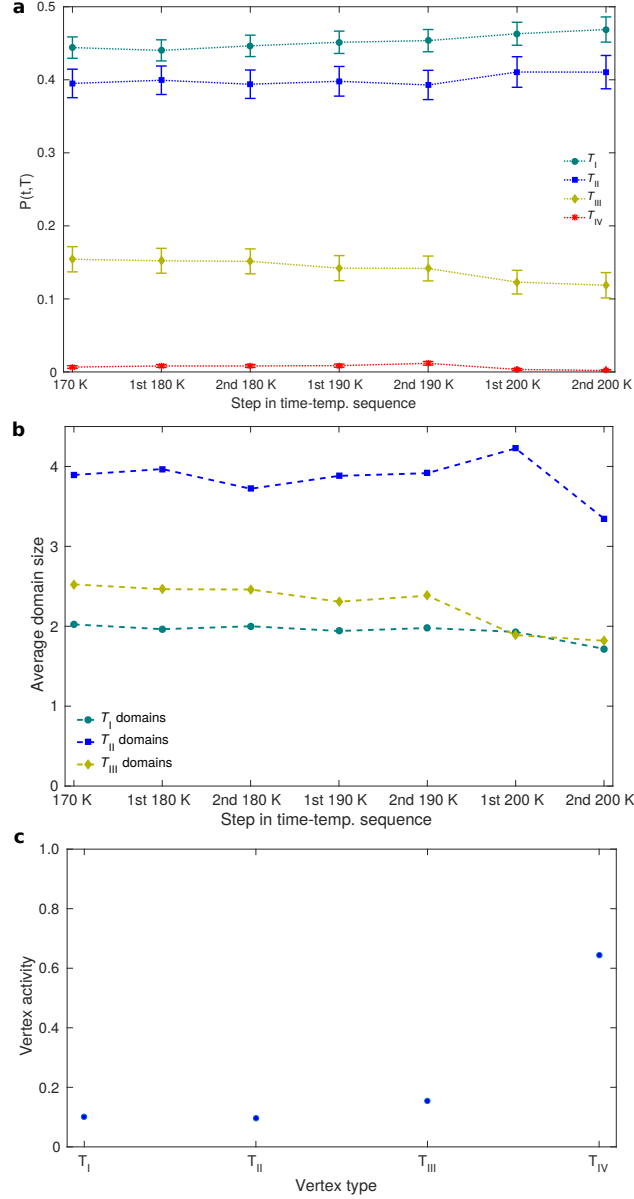
Supplementary Figure 8. Population of islands with a distinct magnetization direction, P_I , for each time-temperature step. An island spending equal, or near equal, amount of time in each of its two possible directions during the measurement will not show any magnetic contrast.



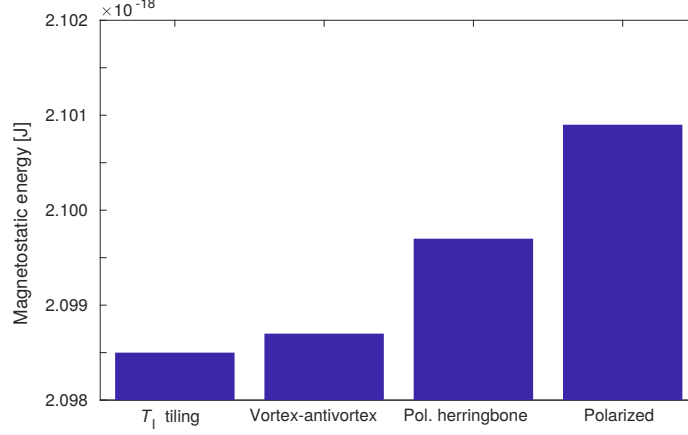
Supplementary Figure 9. PEEM-XMCD image of the $\alpha = 660$ nm $D=150$ nm lattice obtained at 120 K. The islands orientation is rotated 45 degrees with respect to the image shown in Fig. 2 in the article. The image is merged from 12 individual PEEM-XMCD images. The quality of the image is representative for all lattices measured.



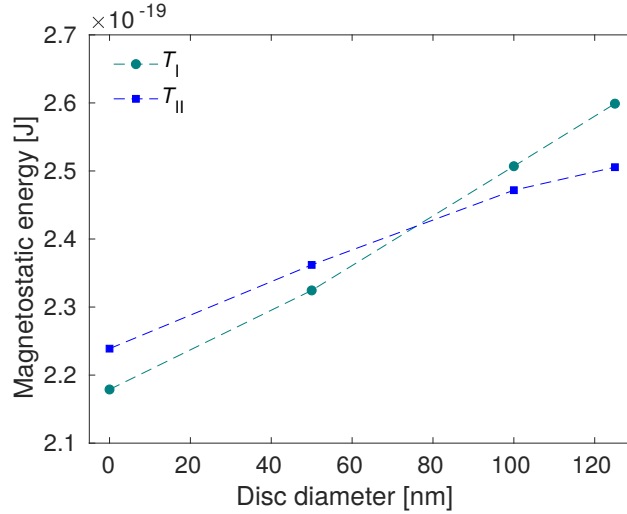
Supplementary Figure 10. Magnetic flux map of the $\alpha = 660$ nm $D=180$ nm lattice with residual flux from T_{II} , blue, and T_{III} , yellow, vertices. The map is rotated 45 degrees in comparison to the schematic maps in Fig. 4 in the article. Different T_{II} flux ordering are marked out. Ferromagnetic order, yellow box, herringbone structure, green box, T_I tiling, blue box and larger scale vortex, red box.



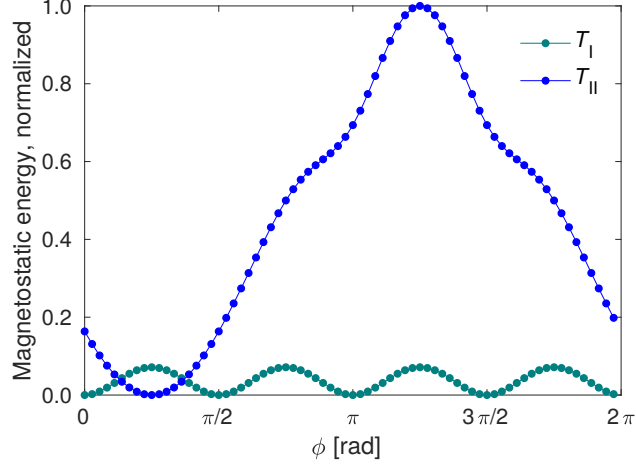
Supplementary Figure 11. **a**, Evolution of the degeneracy corrected, normalized, population during the time-temperature sequence. The populations remain almost constant for the measurement interval. **b**, Evolution of domain sizes for the different vertex types during the time-temperature sequence. The slight decrease in domain sizes is attributed to the decreasing value of islands with a distinct direction of magnetization, see Supplementary Figure 8. **c**, Cumulative vertex activity, where the activity for each vertex type is defined as the relative observed change in the vertex count between consecutive time steps. All error bars represent one standard deviation.



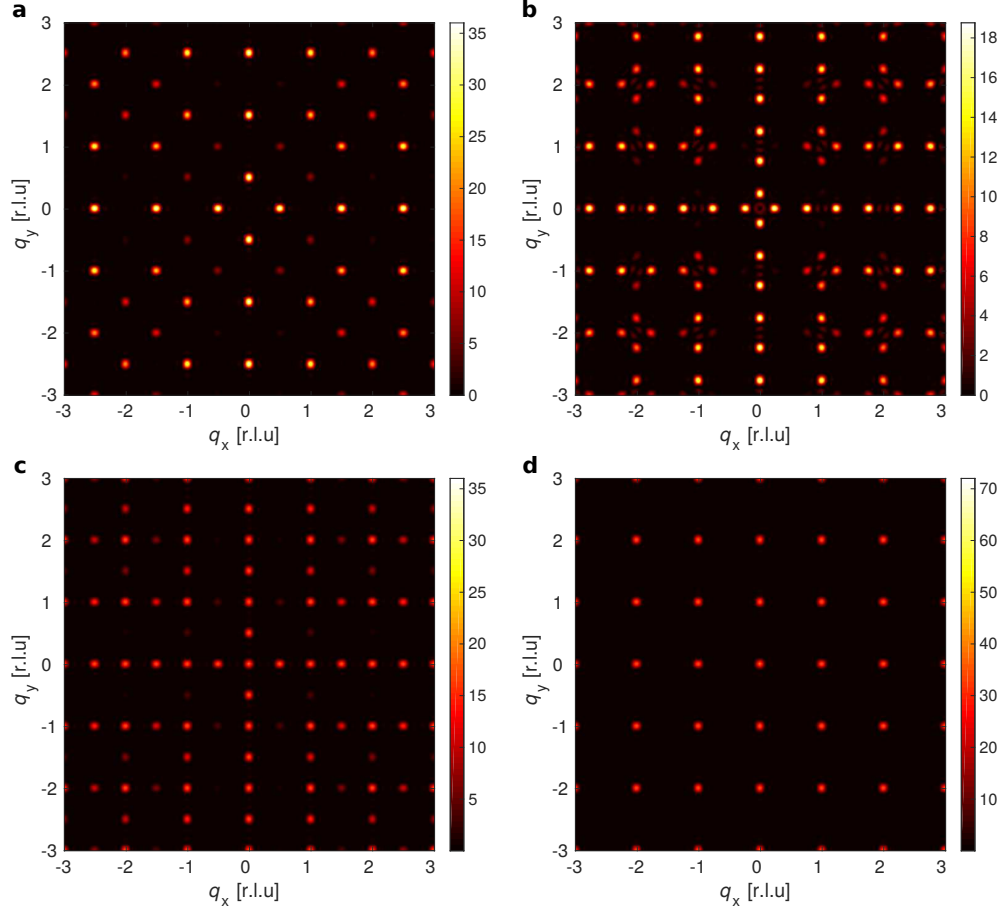
Supplementary Figure 12. Energy comparison between the different all- T_{II} states, as defined in the paper (see Fig. 4), calculated using MuMax3. The absolute energy of each state depend on the size and shape of the domains, therefore it is the relative energies which are of importance in a comparison.



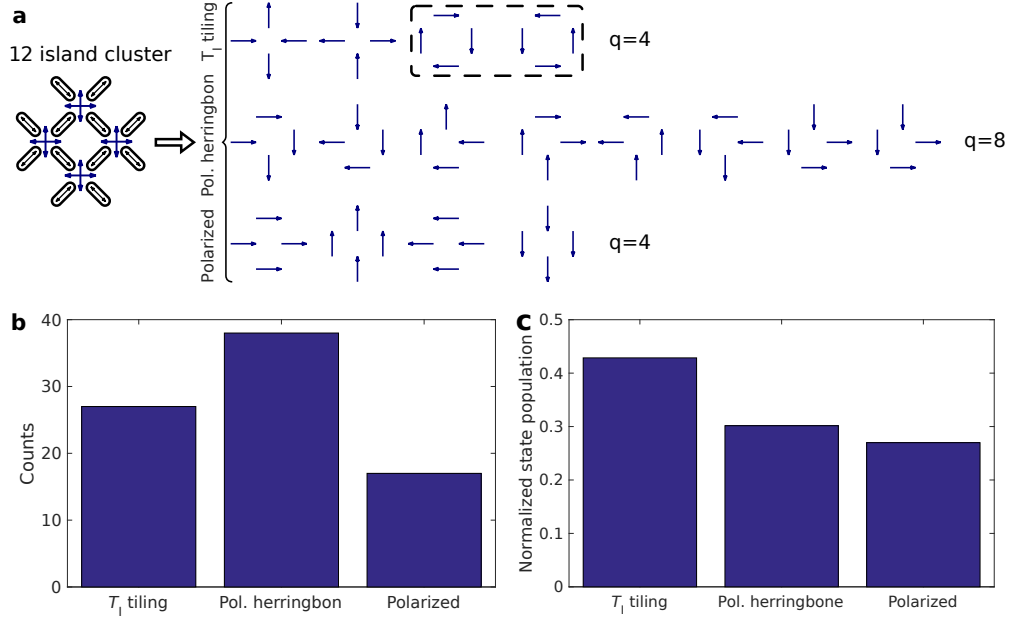
Supplementary Figure 13. Energy comparison between T_I and T_{II} vertices for $\alpha=660$ nm with increasing disc diameter calculated using MuMax3. The crossover in energy happens at smaller disc diameters than what is suggested by the population inversion see Supplementary Figure 1. It is not feasible for a micro-magnetic simulation to capture all the physics contained in a thermal system and it is therefore not surprising that the 0 K simulations do not quantitatively match the experiment to full extent.



Supplementary Figure 14. Energy for T_I and T_{II} vertices in a purely collinear XY-spin approximation for the disc's magnetization while rotating the magnetization of the disc. The energies are normalized to the maximum energy of the T_{II} vertex. The energy minima for the disc in T_I vertices is four fold (the same is true for T_{IV}) vertices). For T_{II} and T_{III} vertices (not shown) there is one energy minima along the combined flux of the islands. Compare to Fig. 1 in the paper. $\phi=0$ rad lies along the long axis of one island.



Supplementary Figure 15. Computed magnetic spin structure factors for the different all- T_{II} states, see Fig. 4 in the paper. **a**, T_1 -tiling, **b**, vortex-antivortex, **c**, herringbone structure, and **d** is the polarized state. The points at $[q_x, q_y] = [n \pm 0.25, 0]$ or $[q_x, q_y] = [0, n \pm 0.25]$, where n is an integer, are specific to the vortex-antivortex state, **b**. Peaks at integer values are shared between **c** and **d**. Points at half integer values are shared between **a** and **c**.



Supplementary Figure 16. In order to estimate the amount of flux ground state, i.e. the T_I -tiling, we consider 12 islands (four flux) as a unit and simply count how many of these units there are in the $660 \text{ nm} \times 180 \text{ nm}$ array, see Supplementary Figure 10, which satisfy the condition shown in **a**. The flux closure circles, dashed box in **a**, are included in the T_I -tiling tiling count since they are a part of the overall structure of an extended T_I -tiling. In absolute counts for the different states there are: 27 Type T_I -tiling, 38 polarized herringbone, and 17 polarized collinear states. **b**, The normalized and degeneracy corrected state population is shown in **c**, where the T_I -tiling is the most abundant.

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- [3] Y. Perrin, B. Canals, and N. Rougemaille, Extensive degeneracy, Coulomb phase and magnetic monopoles in an artificial realization of the square ice model, *Nature* **540**, 410–413 (2016).