Tailoring the magnetic order in mesoscopic spin systems

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Mesoscopic spin systems can be designed and fabricated using modern nano-fabrication techniques. These systems can contain large numbers of patterned ferromagnetic elements, for which the shape will generally determine their effective mesospin dimensionality. The lateral arrangement of these mesospins can be further used to tune the interactions between them.

With an appropriate choice of material, it is possible to define a temperature range where thermal fluctuations of these mesospins are experimentally accessible. To actively define this range, we use δ-doped Palladium, a three-layer system of Palladium—Iron—Palladium, for which the Curie-temperature scales with the Iron layer thickness. The patterned mesoscopic elements used in this work have a stadium-like shape that promotes a single magnetic domain state, thus making these islands behave as one-dimensional Ising-like mesospins that can be observed using magnetic imaging techniques.

We investigate the impact on the magnetic order resulting from modifications of the square spin ice geometry. By adding, removing and merging elements in the square artificial spin ice architecture, energy-landscape variations can be realized. Firstly, an added interaction modifier is used to equilibrate the interactions between the mesospins at the vertex level, which can restore the degenerate ground state of the square spin ice model. Secondly, the removal of elements can lead to topologically frustrated spin systems, as not all building blocks can simultaneously be in their lowest energy state. Furthermore, the merging results in multiple element sizes in the mesospin system. As the magnetization reversal barrier is dependent on the element size, these mesospin systems have different energy barriers. The thermal ordering process in such a system differs from a single-size element system with its unique energy barrier. Using reciprocal space analysis tools like the magnetic spin structure factor we show that systems with multiple element sizes achieve a higher short-range order then their single-size element references. The magnetic order in mesoscopic spin systems could successfully be tailored by modifications of the lattice geometry.

Keywords: artificial spin ice, magnetic nano-structures, mesoscopic spin systems, mesospins, model systems, Shakti, Saint George, interaction modifier, engineering of energy-landscape, multiple energy-scales
Dedicated to my Family
List of papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.

I  A new look on the two-dimensional Ising model: thermal artificial spins
   Unnar B. Arnalds, Jonathan Chico, Henry Stopfel, Vassilios Kapaklis, Oliver Bärenbold, Marc A. Verschuuren, Ulrike Wolff, Volker Neu, Anders Bergman and Björgvin Hjörvarsson
   New Journal of Physics 18, 023008 (2016)

II Thermally induced magnetic relaxation in square artificial spin ice
   Mikael S. Andersson, Spyridon D. Pappas, Henry Stopfel, Erik Östman, Aaron Stein, Per Nordblad, Roland Mathieu, Björgvin Hjörvarsson and Vassilios Kapaklis
   Scientific Reports 6, 37097 (2016)

III The importance of the weak: Interaction modiers in artificial spin ices
   Erik Östman, Henry Stopfel, Ioan-Augustin Chioar, Unnar B. Arnalds, Aaron Stein, Vassilios Kapaklis and Björgvin Hjörvarsson
   Submitted Nature Physics

IV Magnetic order and energy-scale hierarchy in artificial spin ice
   Henry Stopfel, Erik Östman, Ioan-Augustin Chioar, Unnar B. Arnalds, David Greving, Thomas P. A. Hase, Aaron Stein, Björgvin Hjörvarsson and Vassilios Kapaklis
   Manuscript

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Comments on my participation

The following is a brief description of my involvement in the publications:

I Participated in the data analysis and interpretation and contributed to the manuscript.
II Performed the sample growth and nano-fabrication. Contribution to the data analysis, discussion of the results and writing of the manuscript.
III Responsible for sample growth and nano-fabrication. Participated in the PEEM-XMCD measurements and contributed to the analysis and theory development, discussed the results and commented on the manuscript.
IV Responsible for sample growth and nano-fabrication. Participated in the PEEM-XMCD measurements. Analyzed the data and co-wrote the manuscript.

Papers not included in this thesis

V Magnetic properties of amorphous Fe$_{93}$Zr$_7$ films: Effect of light ion implantation
Atieh Zamani, Reda Moubah, Martina Ahlberg, Henry Stopfel, Unnar B. Arnalds, Anders Hallen, Björgvin Hjörvarsson, Gabriella Andersson and Petra E. Jönsson

VI Spin transfer torque ferromagnetic resonance induced spin pumping in the Fe/Pd bilayer system
A. Kumar, S. Akansel, Henry Stopfel, M. Fazlali, J. Åkerman, R. Brucas and P. Svedlindh
Physical Review B 95, 064406 (2017)
1. Introduction

The advances in nano-fabrication techniques allow the design and creation of various two-dimensional structures consisting of differently sized and shaped elements. Sub-micrometer sized magnetic objects can be treated as one mesoscopic magnetic entity, being in a single-domain state [1]. This single domain has no restrictions on its magnetization direction in the case of three-dimensional magnetic particles with low or zero crystalline-anisotropy. Spatial confinement results in a shape-anisotropy, defining the magnetization direction of the ferromagnetic object depending on its extension in the different dimensions. Thin disc-like objects are free to rotate their magnetization direction within the disc plane and can be viewed as two-dimensional XY-spin rotors[2]. Thin needle-like structures have one preferable magnetization axis and can therefore be treated as one-dimensional mesoscopic Ising-like spins[3]. Close proximity between these mesospins enables the investigation of different interaction schemes and their resulting collective magnetic order at the mesoscale. This magnetic order can be directly observed with magnetic imaging tools. These properties are qualifying mesospin arrays as model systems for interactions in many-body systems, which can result in unconventional and exotic states.[4, 5, 6, 7]

The phase space of artificially produced spin systems can be explored using thermally active mesospins[8, 9]. Ground state ordering in square artificial spin ice[10] can be achieved by utilizing thermodynamics during the growth. Studies in thermally active artificial spin ices span from order in building blocks[11, 12] or extended arrays[13, 14, 15, 16] to the observation of relaxation dynamics[17, 18] and thermodynamic phase transitions[19].

The possibility of accessing the magnetic states thermally initiated the quest to modify the energy-landscape of these two-dimensional systems to achieve and investigate new and emergent order[20, 21, 22]. The pyrochlore spin ices with their degenerate ground state are characterized by an isotropic nearest-neighbor interaction. The planar arrangement of magnetostatically interacting mesospins in square artificial spin ice breaks this symmetry, leading to a stronger coupling of perpendicular mesospins. Altering the planar geometry of the square artificial spin ice[20, 22] can restore the full degeneracy of the square spin ice model[23], as Perrin et al.[24] have successfully presented in their study. A different way of modifying the energy-landscape was proposed by Morrison et al.[21], where new topologies are produced by removing and merging spins in the square artificial spin ice lattice. Gilbert et al.[15] realized one of these structures and showed the theoretically predicted behavior of the
Shakti lattice[25]. The Shakti lattice is a geometry composed of two element sizes. Due to the use of thick Permalloy, Gilbert et al.[15] assumed only one energy-scale for the differently sized elements. Cowburn and Welland[26] showed that different energy barriers for the elements of one system can be used to preserve a certain magnetic state in the larger elements (higher energy barrier), while the smaller elements are changing their magnetic orientation due to external energy input.

In this thesis the modification of the energy-landscape in two-dimensional magnetic mesospin systems is in the main focus. Additionally to the geometrical modification approach proposed by Morrison et al.[21], interaction modifiers are introduced in the square artificial spin ice geometry to alter the energy relations in these systems. This added magnetic element enables the variation of the coupling strength between the elements in the system and therefore modifies the energy-landscape as well. Throughout this thesis differently sized and shaped mesoscopic magnetic elements are used to modify the energy-landscape of artificial magnetic structures. The different modifications were used to create new emergent ordered phases as well as "pinning" centers for local magnetic order. Especially for the latter part we are using thermally active mesospins of varying sizes to investigate the influence of different length- and energy-scales on the thermally induced magnetic order in these systems. The impact of these different sized magnetic objects on the thermal ordering of such mesoscopic magnetic spin systems is vastly unexplored and a major part of this work. The mesoscopic spin systems are locally investigated by photoemission electron microscopy using the X-ray magnetic circular dicroism, to visualize the magnetic order within the arrays. Reciprocal space techniques such as the magnetic spin structure factor are optimal tools to investigate global ordering in these mesospin systems.

We present here the variation of the magnetic order in square artificial spin ice based structures, triggered by modified energy-landscapes. A combination of these modifications gives rise to a whole new world of thermally active multi-scale mesospin systems.


2. Mesospins

The spontaneous magnetization direction of ferromagnets can be determined by two factors: the shape- and the crystalline-anisotropies. These anisotropies result in energy barriers, which effectively reduce the degrees of freedom the spins in a ferromagnet can access. The shape-anisotropy is originating from the extension and shape of the ferromagnet, as illustrated in figure 2.1, while the crystalline-anisotropy is material dependent and originates from the overlap of the electron orbitals of neighboring atoms.[27] This leads to energetically more favorable magnetization directions in crystals, defining soft- and hard-magnetic axes.[28] In poly-crystalline materials the shape-anisotropy defines the preferred magnetization direction of a ferromagnetic object (illustrated in figure 2.1).

![Figure 2.1. Illustration of the spin dimensionality (a) and the respective extensions of the ferromagnet (b). A three-dimensional ferromagnetic single-domain particle, without crystalline-anisotropy, is free to rotate its magnetization direction in three-dimensions. A thin ferromagnetic disc is effectively reduced to two-dimensions, the shape-anisotropy forces the magnetic orientation in the remaining sample plane. The spatial reduction to an elongated ferromagnetic object, can be interpreted as being one-dimensional and the shape-anisotropy leads therefore also to an effectively one-dimensional Ising-like spin.](image-url)
The shape-anisotropy can be defined by nano-fabrication techniques. In the absence of crystalline-anisotropy, the shape-anisotropy of a magnetic thin film confines the direction of the magnetization in the sample plane. In the thin film plane the direction of the magnetization is free to rotate, but energetically unfavorable to rotate out of the sample plane. Depending on the material properties, sub-micrometer sized ferromagnets are in a single-domain state[1, 29] and can therefore be seen as mesoscopic spins. The spin dimensionality of mesoscopic spins are dependent on their nano-patterning. Thin magnetic discs will result in two-dimensional mesoscopic spinrotors[2], while thin stadium- or needle-like magnetic objects are effectively one-dimensional Ising-spins. Patterned magnetic thin films are therefore suitable as mesospin model systems, with their mesospin dimensionality controlled by the nano-fabrication and the choice of material.

Depending on the lateral placement and the used magnetic material, different coupling strengths between these mesospins can be facilitated and therefore the interactions in mesospin systems can be designed. Larger mesospin distances, for example, reduce the interactions between the mesospins and make the mesospin system more susceptible to excitations.

Especially thermally active mesospins are of great interest as they allow the thermodynamic exploration of different spin model systems. The magnetic orientation of thin magnetic discs is intrinsically free to rotate (as illustrated in figure 2.1) and is restricted by the interaction with neighboring mesospins. Thin stadium-like magnetic objects on the other hand have an energy barrier for the rotation of the magnetization direction as illustrated in figure 2.2. Due to the shape-anisotropy only two magnetization directions are energetically favorable for these mesospins. Therefore these mesospins are comparable to the spins Ernst Ising proposed in his publication in 1925[3] and being commonly called Ising-like spins. The energy barrier is dependent on the magnetic material but also on the size of the element as shown by Cowburn and Welland[26]. In their pub-

Figure 2.2. Schematic illustration of the energy barrier for stadium-like nanopatterned islands assuming a ridged spin model. The magnetization direction in such islands has two energy minima at 0° and 180°. These magnetization directions correspond to the long axis of the islands and result from the shape-anisotropy. Depending on the size of the magnetic islands the energy barrier is higher (long island – blue) or lower (short island – red).
lication Cowburn and Welland investigated chains of magnetic discs cornered on one end with an elongated long-mesospin. The magnetic discs are generally free to rotate their magnetization direction within the plane, but an energy barrier results from the close proximity to the neighboring discs. Similar to the illustration in figure 2.2 the magnetic discs exhibit a smaller energy barrier than the elongated long-mesospin. When applying an external magnetic field the magnetization direction of the magnetic discs is varied, but the magnetic field is not big enough to overcome the energy barrier of the elongated long-mesospin, which therefore preserves the magnetic information.

In thermally active magnetic systems the thermal energy is big enough to overcome the magnetization reversal energy barrier for a wide temperature range. Therefore the magnetization direction of thermally active mesospins can be easily altered in this temperature range. This creates energetically more favorable mesospin arrangements or thermal fluctuations. Simple spin systems with similarly oriented one-dimensional mesospins (section 2.1) or more elaborate geometries, where the one-dimensional mesospins are arranged to each other under certain angles (section 2.2), can be produced. Depending on the lateral arrangement of the mesospins different interactions and couplings are promoted.

2.1 Ising model systems

Magnetic model systems can be designed using one-dimensional mesoscopic Ising magnets in a close lateral arrangement.[4] The theoretical description of a one-dimensional chain was given by Ernst Ising[3] in 1925. The two solutions depend on the coupling between the spins, a positive coupling (figure 2.3a) gives rise to ferromagnetic ordering of the elements and a negative coupling (figure 2.3b) of these spins results in an anti-ferromagnetic coupling between the neighboring elements. The spatial orientation of magnetic islands, used as mesoscopic spins, results in the different coupling regimes. Head to tail aligned mesospins couple preferably ferromagnetically and parallel mesospins anti-ferromagnetically.

![Figure 2.3. Illustration of 1D and 2D Ising systems. Ferromagnetic (a) and anti-ferromagnetic (b) one-dimensional Ising chain. c), AFM measurement of an experimentally realized two-dimensional Ising system, with an illustration of the magnetic ground state configuration (PAPER-I).](image)
Figure 2.4. Representation of the as-grown state of an experimental realization of the 2D-Ising system (PAPER-I). The over 10000 mesospins of the two different ground state domains are illustrated with yellow and blue arrows, which have been measured by magnetic force microscopy.

In the 2D-Ising lattice (figure 2.3c) the coupling between the one-dimensional Ising-like mesospins is therefore different for the vertical and horizontal neighbors. This system has a well defined ordered ground state of ferromagnetically aligned chains, anti-parallel aligned to their neighboring chains. This ground state is a result of the different coupling in the two main lattice directions. Lars Onsager[30] solved the 2D-Ising system analytically and opened up the pathway to investigations on frustrated spin systems.

An experimental realization of a 2D-Ising system is presented in PAPER-I, "A new look on the two-dimensional Ising model: thermal artificial spins", using thermally active one-dimensional mesospins (as illustrated in figure 2.3c). The arrested thermalized as-grown state has been measured with magnetic force microscopy and results of these measurements are illustrated in figure 2.4. This preserved state resembles a higher temperature state locked in at a time when the array was undergoing high thermal fluctuations. Monte Carlo simulations[31], using nearest neighbor interaction energies, can reproduce the arrested state in agreement with Onsager’s[30] analytical solution.

As a model system the 2D-Ising system is a simple realization with a well defined ordered state. Modern Electron-Beam-Lithography techniques allow more elaborate structures, for further details on the patterning process see section 3.2. The used magnetic material makes the produced array athermal and therefore easily measurable by magnetic force microscopy, as the thermalization process only takes place in a short time period during the sample growth. For real thermally active magnetic mesospin systems other materials need to be employed, see more details concerning thermal active magnetic materials in section 3.1.
2.2 Artificial spin ice

Ising-like mesospins can be used in more advanced geometries to mimic and investigate certain physical phenomena. One major topic is the research on frustrated systems. Linus Pauling[32] raised in 1935 the question about the high degeneracy of water ice, pursuing earlier investigations on the configurations of water molecules in water ice[33]. Water ice inhabits even at very low temperatures residual entropy, due to its highly degenerate ground state configuration. A magnetic analogy to the disorder among the hydrogen atoms in water ice was found later in pyrochlore spin ices[34, 35]: A three-dimensional magnetic system in which the atomic spins are located at tetrahedral sites and their magnetic orientation is restricted to either point into or out of the tetrahedron (see figure 2.5).

The ground state manifold is given by the so called "ice rule", describing in water ice the relative displacement of the four hydrogen atoms around an oxygen atom (see Fig. 2.5a). In this tetrahedral geometry two of the hydrogen atoms are covalent bonded to the oxygen and the other two hydrogen atoms are connected via hydrogen bonds to the oxygen. The stronger covalent interaction leads to a displacement vector pointing into the tetrahedron. The other two hydrogen atoms have a displacement vector pointing out of the tetrahedron, since these hydrogen atoms are covalent bond to the oxygen atoms in the adjacent tetrahedrons. This two in and two out "ice rule" can directly be transferred to the magnetic equivalent. In the spin ice pyrochlores the "ice rule" describes if the spin, located at the corners of the tetrahedron, points in or out of the tetrahedron. An obeyed "ice rule" describes the highly degenerate spin ice ground state manifold and a violation of the "ice rule" results in effective magnetic charges, also called magnetic monopoles[37]. The research on such atomistic three-dimensional systems is challenging and relies on reciprocal space investigations at very low temperature[38]. To observe the local spin arrangements in such systems, two-dimensional model systems can be

Figure 2.5. a) Water ice and b) pyrochlore spin ice representations from Castelnovo et al.[36], illustrating the frustrated three-dimensional systems. c) Square and d) Kagome artificial spin ice geometries as two-dimensional realization of frustrated systems.
used. In particular artificial spin ice (ASI) structures in which the physics of frustrated and highly disordered systems can be achieved and simultaneously locally probed.

The square artificial spin ice[39, 10] (figure 2.5c) and the kagome artificial spin ice[40, 41, 11] (figure 2.5d) have been explored as two-dimensional frustrated spin model systems. As a consequence of the reduction from three- to two-dimensions both ASI systems cover different aspects of the spin ice pyrochlores.

The so called vertex is the point in the lattice where the mesospins meet (highlighted in figure 2.5c-d). In the kagome ASI the vertex inhabits three mesospins (coordination number three) while in the square ASI four mesospins create a vertex (coordination number four). As the kagome ASI has only vertices with coordination number three the "ice rule" is adjusted to two-in and one-out or vice versa. The odd coordination number of the kagome vertex leads to a residual magnetic charge at any time, resulting in new charge ordered spin ice phases. The square ASI preserves the "ice rule", but the two-dimensional nature of the lattice and the magnetostatic interaction between the elements makes the coupling between parallel and perpendicular islands different, destroying the degeneracy of the six "ice rule" obeying states. Since all investigations carried out here are based on the square ASI lattice geometry, section 2.2.1 is highlighting the properties of the square ASI.

\[ \begin{array}{cccc}
\text{Type-I}_4 & \text{Type-II}_4 & \text{Type-III}_4 & \text{Type-IV}_4 \\
\text{z} = 2 & \text{z} = 4 & \text{z} = 8 & \text{z} = 2 \\
\text{q}_m = 0 & \text{q}_m = 0 & \text{q}_m = \pm 2 & \text{q}_m = \pm 4 \\
\end{array} \]

Figure 2.6. A graphical representation of the four energetically different vertex states in the square ASI. They are ordered accordingly to their energies, starting by the lowest energy level, Type-I$_4$, up to the highest excitation, Type-IV$_4$. The degeneracy of each vertex state is given by z, resulting in a total of sixteen different mesospin arrangements. The effective magnetic charge, q$_m$, is given for each vertex state. Type-I$_4$ and Type-II$_4$ are the "ice rule" obeying vertex states and therefore free of vertex charges.
2.2.1 Square artificial spin ice

Since Wang et al. [39] there have been numerous investigations on square ASI structures. The four islands of a vertex are placed in such a way that two of them are parallel and the other two islands are perpendicular. Due to the two orientations for each of the mesospins, a total of sixteen possible vertex states can be obtained. The four different energy states are represented in figure 2.6. The broken symmetry of the interactions between parallel and perpendicular mesospins lead to an energy split between the "ice rule" obeying Type-I$_4$ and Type-II$_4$ vertices. This lifted degeneracy in the square ASI geometry results in a well defined ground state with alternating degenerate Type-I$_4$ vertices. While Type-I$_4$ vertices are magnetic charge free and have no residual magnetic dipole field, Type-II$_4$ vertices are charge free but have a residual magnetic field. Type-III$_4$ excitations violating the "ice rule" and inhabiting therefore a residual magnetic charge, $q_m$, commonly called magnetic monopoles. Additionally to the magnetic charge, Type-III$_3$ vertices also have a magnetic orientation as the Type-II$_4$ vertices. Type-IV$_4$ vertices have a residual magnetic charge as all islands are pointing in or out of the vertex and due to this the vertex has no magnetic dipole field.

The standard islands in this thesis have the following spatial constraints. The rounded ends of the islands are created by a half-circle with the diameter $w$. In between the two half-circles a rectangle with a width of $w$ and the length of $2 \cdot w$ is placed, creating an island with a stadium-like shape and a width of $w$ and a length $l = 3 \cdot w$. The aspect ratio $w : l$ of 1:3 is chosen to ensure the Ising-like behavior of the islands. The lattice parameter, $p$, is defined as the center to center distance between two parallel islands. Therefore the edge to edge distance can be calculated as

$$d_{\parallel} = p - l$$

for parallel islands. The edge to edge distance for perpendicular islands is then

$$d_{\perp} = \frac{p - l + 2w \cdot (1 - \sqrt{2})}{\sqrt{2}}.$$ 

As the mesospins used in this thesis are extended magnetic islands, the stray-field from the edge of the island is causing the interaction between the mesospins. Therefore the edge to edge distance is an important parameter for the interactions in these structures.
2.2.2 Modified square artificial spin ice lattices

The modifications of square ASI can be used to design completely new meso-spin systems, resulting in very different order and dynamics. As a base for comparisons and to investigate the magnetic orientation easily with photoemission electron microscopy (explained in section 3.4.2) we use the square ASI lattice as a foundation for our new lattices[21]. Perrin et al.[24] demonstrated impressively how the degeneracy between Type-I_4 and Type-II_4 vertices can be recovered by elevating certain islands[20, 22] and therefore restoring the equal interactions between perpendicular and parallel islands. Here we only implement planar modifications of the two-dimensional lattices. One possibility is to add magnetic material into the array to pin or mediate the magnetic stray field of the nano-patterned islands. Other modifications are removing and merging of islands in the square ASI topology. In the scope of this work we used all of these mentioned approaches, illustrated in figure 2.7. In this way we can modify the energy-landscape in square ASI, create master–slave relations between certain magnetic entities and construct lattices with mixed coordination numbers as well as different length- and energy-scales (within one array). Another aspect of removing islands from the square ASI
geometry was investigated by Drisko et al.[42], they could show the influence of topological defects on the magnetic ordering and the resulting frustration in a generally unfrustrated system. This interesting effect has not been looked upon in this thesis, as the modifications carried out here are periodic and symmetric throughout the entire lattice.

The interaction modifier

The distance difference between parallel and perpendicular islands in the square ASI is the reason for the loss of the degeneracy between the Type-I$_4$ and Type-II$_4$ vertex states. Perrin et al.[24] achieved this recovery of the degenerated six-vertex model[23] by elevating one line of mesospin. In this thesis we are using a magnetic disc, XY-mesospinrotor, in the center of each vertex to change the energy landscape in the square ASI. The additional magnetic element changes the magnetostatic interactions between the mesospins and unify their interaction with the magnetic disc, as illustrated in figure 2.8a). Depending on the lattice parameter ($p$), the disc diameter ($D_{\text{disc}}$) can be varied and different disc-islands edge to edge distances, $d_{\text{disc}-\text{island}}$, can be achieved. This distance can be calculated as

$$d_{\text{disc}-\text{island}} = \frac{p - l - D_{\text{disc}}}{2}$$

and is equidistant to all four surrounding islands. Depending on this distance, $d_{\text{disc}-\text{island}}$, and the disc diameter, $D_{\text{disc}}$, the interaction between island and disc varies. While small discs are generally in a master-slave relationship, where the Ising islands will dominate the magnetic ordering, the relation changes as the disc diameter increases. For our Ising islands with $w = 150$ nm and $l = 450$ nm a disc of $D_{\text{disc}} = 150$ nm has only 28% of the magnetic material of an island. A disc with $D_{\text{disc}} = 200$ nm on the other hand has already 50% and a disc of $D_{\text{disc}} = 250$ nm has 78% of the magnetic material of one Ising-island. The magnetic orientation of a small magnetic disc will be dictated by the magnetization direction of the surrounding islands. A larger magnetic disc will also influence the magnetic orientation of the islands and not just be dictated by its neighbors. The energy-landscape of the lattice is modified either way. A micromagnetic calculation[43] of the vertex-energies depending on the disc diameter is shown in figure 2.8b) and illustrates how the energy-landscape in square ASI is changed by the interaction modifier. Depending on the thickness of the magnetic layer, the lattice parameter and the disc size, it is possible to engineer the square ASI in a way that the degeneracy of Type-I$_4$ and Type-II$_4$ vertices can be recovered in a planar geometry. Furthermore in lattices where the Type-II$_4$ vertices have the lowest vertex energy, a new class of ASI with residual magnetic field contributions for the lowest vertex-energy is created. With even larger disc sizes a cross over between Type-I$_4$ and Type-III$_4$ can be created. This energy-landscape is especially interesting focusing
on the excitations created throughout the array. Magnetic monopoles (Type-III₄) are more likely to be created and the charge and stray-field free vertices (Type-I₄) are a highly excited state, magnetic charge order could be a result of such an energy-landscape.

Figure 2.8. a), Schematic representation of the interaction distances between the islands in a square ASI and the change promoted by the introduction of an interaction modifier in the center of the vertex. b), Vertex energy evolution depending on the diameter size of the interaction modifier. Two crossings are detectable and representing the total change of the magnetic ordering in the mSASI. First the Type-I₄ crosses the Type-II₄ leaving Type-II₄ as the lowest energy state. For even larger disc sizes the Type-I₄ vertex energy becomes even higher then the Type-III₄ vertex energy, creating a lattice with residual magnetic fields for the two lowest vertex energies.
Removing islands

The opposite modification of adding elements into the square array is the removal of certain islands. Depending on which islands are removed vertices with different coordination numbers will be created. The coordination number defines how many mesospins are present in a vertex, therefore by removing islands from a square ASI lattice the coordination number of certain vertices is reduced from coordination number four to three and in some cases even to coordination numbers of two. A vertex with four mesospins is more strongly coupled then a vertex with only three or even two mesospins, creating stronger and weaker sides in the array. Morrison et al.[21] described several of these structures and how the removed mesospins will influence the magnetic ordering and even create frustrated systems. By removing 25% of the islands out of the square ASI lattice one can create the Shakti lattice geometry. This lattice was initially introduced by Chern et al.[25] as a possible way to create an emergent six-vertex model[44], more details about this lattice are described in section 2.2.3. Lattices with the same building blocks, vertices with coordination number four, three and two, can be created by the same approach. However, the altered arrangement of the islands will lead to different magnetic order, one of these examples is the so called Saint George lattice, further explained in section 2.2.4.

Merging islands

As already mentioned, Morrison et al.[21] proposed new artificial topologies derived from the square ASI. Additionally to the removal of islands certain islands in the lattice can also be merged. Vertices with coordination number two, in particular parallel islands, can be merged into one long-island. This process reduces the degrees of freedom in the lattice and therefore adds new constraints to the magnetic ordering of the mesospins. Looking into the Shakti lattice geometry, all vertices with coordination number two will be merged into long-islands. This lattice will be further referred to as the regular Shakti (SH) lattice, while the Shakti lattice with vertices with coordination number two is called the modified Shakti (mSH) lattice, for illustration see figure 2.7. A similar analogy holds for the Saint George lattice.

Due to the differently sized elements in the lattice, different energy barriers (as illustrated in figure 2.2) and therefore different thermally active temperature regions for the respective elements will be present. The thermal ordering processes in lattices with unique or multiple element sizes will vary. More details about thermally active magnetic mesospin systems will be discussed in section 3.3.
2.2.3 The Shakti lattice

Figure 2.9. SEM images of the mSH (left) and the SH (right) lattice. The vertex types of each lattice are marked with their respective color. The SH lattice inhabiting only vertices with the coordination number three (blue) and four (green), while the mSH lattice consists of vertices with the coordination number two (yellow), three (red) and four (green). The four energy levels of the four-fold coordinated vertex are illustrated in figure 2.6, resembling the square ASI vertices and being the same for the mSH as well as the SH lattice. The remaining vertex states are illustrated in figure 2.10.

Morrison et al.[21] introduced the Shakti lattice in their publication about frustrated systems created by variations of the square ASI. Chern et al.[25] described the Shakti lattice more in detail, showing that the real nature of the frustration in this systems comes not only from the pair-wise frustration, it is a topological induced frustration. The two vertex types, coordination number four and three, have both well-defined lowest energy states (see figure 2.10), but due to their placement in the Shakti lattice the array can not satisfy all of these vertices at the same time. Chern et al.[25] introduced a plaquette, a configuration of four vertices with coordination number four as well as four three-fold coordinated vertices, see illustration figure 2.11. The ground state configuration is given by all four-fold coordinated vertices being in their lowest energy level and two of the vertices with coordination number three in their lowest and two in their first excited state, as illustrated in figure 2.11. Since there are six different combinations of the positions at which this two excited states can occur in a plaquette, the Shakti lattice in its ground state resembles an emergent six-vertex model[44]. This Shakti ground state manifold inhabits a large degeneracy.

Gilbert et al.[15] presented the first experimental study of such an exotic ASI structure. In their thermalized Permalloy Shakti lattice they could recover to a large extent the predicted behavior. Interestingly, the modified Shakti
with non-merged two-fold coordinated vertices shows the same behavior and is even slightly more in the Shakti ground state. But since the islands are made out of Permalloy the thermalization process between short- and long-islands are not that different. Therefore we want to explore these lattices with a more thermally active material like the δ-doped Palladium, see section 3.1 for more details.

Figure 2.11. Definition of a plaquette and different ways of illustrating the Shakti ground state manifold in a plaquette. a), Mesospin arrangement of the islands shared by a plaquette to illustrate the schematic plaquette description in b). Chern et al.[25] defined the SH ground state as all four-fold coordinated vertices in their lowest energy state, Type-I₄, forcing the three-fold coordinated vertices to inhabit two excitations, Type-II₃, illustrated with a circle in the plaquette nomenclature. b), Illustration of the six different defect arrangements in a plaquette, for either horizontal or vertical long-mesospin in the middle of the plaquette. c), Representation of b) focusing on the two degenerated Type-I₄ vertices and their placement in the plaquette. Type-I₄(A) is represented in magenta and Type-I₄(B) in cyan, the six plaquette illustrations can be also completely spin flipped, changing all Type-I₄(A) into Type-I₄(B) and vice versa.
2.2.4 The Saint George lattice

In the Saint George lattice, the high symmetry of the Shakti lattice is broken, as the long-mesospins are brought in close proximity. The 90° symmetry axis in the Shakti lattice is changed to a 180° symmetry in the Saint George lattice. Even though there are, as in the Shakti lattice, 25% of the islands removed and another 25% are merged to long-islands, the chosen islands are distributed differently throughout the lattice. The motivation behind the Saint George lattice is the study of order among the long-mesospins resulting from their close proximity and the implications of that ordering on the collective magnetic configuration in the whole lattice. A long-mesospin ordering would influence the short-mesospins drastically and major differences between modified Saint George (mSG) and regular Saint George (SG) lattice should be detectable.

Figure 2.12. A candidate for a ground state arrangement in the mSG and SG lattice. While the four-fold coordinated vertices are in their lowest energy state, Type-I₄, the three-fold coordinated vertices inhabit 50% excitations, Type-II₃. By allocating the mesospins to the islands, closed loops are created, indicated with a blue or red background, in the red region the bordering mesospins have a clock wise alignment and in the blue an anti-clock wise. The magnetic orientation of the center spin of these arrangement is frustrated. Energetically it does not matter in which of the two directions its magnetization points, but the choice of the direction defines the position of the Type-II₃ excitation.

Neglecting the different energy-scales for long- and short-islands and just considering the different vertex energies, the Saint George lattice is a topological frustrated lattice similar to the Shakti lattice, see figure 2.12. Of course the different island sizes in these lattices are influencing the thermodynamical response, more details about the thermodynamic nature of our systems can be found in section 3.3.
3. Experimental realization of mesospins

This chapter describes the sample production, explains the material choice and highlights the thermal behavior of our material, as well as it gives an overview of the used experimental methods and analysis tools.

3.1 δ-doped Palladium thin films

The so called δ-doped Palladium is in fact a three layer system consisting of Palladium-Iron-Palladium. The Iron layer thickness defines the Curie temperature, $T_C$, of the material. Previous studies have shown that the $T_C$ of these systems can be scaled with 200 K per mono-layer of Iron.[45] As a consequence of this we are able to create magnetic thin films which have a $T_C$ in temperature regions suitable for our desired experiments. This is achieved by the control of the Iron-layer thickness. In the presented studies the Iron thickness is in the order of 1.8 to 2.2 mono-layers, resulting in magnetic thin films with a $T_C$ of 360 K to 440 K.

The thin films were produced by magnetron sputtering on a Magnesium Oxide (MgO) substrate, with a seeding layer of Vanadium (V). For practical reasons the following Palladium (Pd) layer has a thickness of roughly 40 nm. This thick Pd layer ensures a smooth surface for the Iron (Fe) layer and will be employed as a conductive seeding layer during the experiments (for more details see section 3.4.2). The magnetization defining Fe layer is then capped by 2 nm Pd, protecting the Fe from oxidation and enabling surface sensitive measurement methods like Photoemission Electron Microscopy. Magnetic nanostructures produced out of δ-doped Pd(Fe) have a wide temperature range in which the islands undergo thermal fluctuations. By the correct choice of the
Fe-layer thickness, the thermal active temperature range can be adjusted to the requirements of the experiment. Therefore \( \delta \)-doped Pd(Fe) is a material by design, which fits our intention to create thermally active artificial magnetic systems.

### 3.2 Patterned nano-structures

This work relies on the research of artificial magnetic spin systems, the nanopatterning process is therefore an essential part of this thesis. There are different processes which can be used to create nano-structures, ranging from self-assembled particles\[46\] to Electron Beam Lithography\[47\] (EBL). The process itself can be separated in two different categories: pre- and post-patterning. The illustration of the two processes in figure 3.2 represent the ways in which the magnetic structures discussed throughout the thesis are made. PAPER-I utilized pre-patterning tools to achieve the 2D-Ising system (see section 2.1), while in the other publications PAPER-II, III and IV a post-patterning process using positive electron beam resist was applied, to create the artificial spin ice structures.

During the pre-patterning process a mask of the desired structure is produced on top of a substrate (figure 3.2a step 1-2) and the magnetic material is then deposited in the holes (figure 3.2a step 3-5). The resist is removed by a chemical lift-off process (figure 3.2a step 6), leaving the magnetic structure behind. A post-patterning process on the other hand requires the production of a magnetic thin film first, on top of which the resist is deposited (figure 3.2b step 1) and then the desired structure will be written into this resist (figure 3.2b step 2). If negative resist is used, an etching mask of resist will be present on top of the thin film and the structure needs to be imprinted into the film by milling/etching. For all the investigated artificial spin ice structures we were using only positive resist. Holes are created (figure 3.2b step 2), after the resist was developed, on top of which we have to deposit a Chromium (Cr) etching mask (figure 3.2b step 3). After the lift-off process, the Cr etching mask remains on top of our magnetic thin film (figure 3.2b step 4). Similar to the negative resist etching mask the structure can now be imprinted in the thin film by milling/etching (figure 3.2b step 5). As a final step the Chromium mask has to be removed by a wet etch process (figure 3.2b step 6), resulting in our desired magnetic structures.

**Post-patterning**

Here are some detailed information about the actual patterning process used for the magnetic nano-structures investigated throughout this thesis. All of which where patterned at the Center for Functional Nanomaterials (CFN) at the Brookhaven National Laboratory in Upton, New York. As already men-
Figure 3.2. Schematic representation of the used patterning techniques. a), The pre-patterning process, used for PAPER-I and b) illustrate the post-patterning process applied in PAPER-II, III and IV. In step 1 the resist is spin coated on top of the substrate(a) / magnetic thin film(b). Step 2, the desired structure is written in the resist and the resist was developed. The following steps differ for pre- and post-patterning. In pre-patterning(a) during step 3-5 the magnetic material is sputtered on top of the sample, resulting in a deposition of the magnetic material on top of the resist as well as into the holes. The final step 6 in the pre-patterning process(a) is the lift-off, here the resist is removed in a chemical etching process, with the consequence that the material which was sputtered in the holes is the only remaining structure on top of the substrate. In the post-patterning process(b), step 3 is the deposition of the Chromium mask, on top of the resist as well as in the holes. In step 4 the resist is resolved in a chemical etching process (similar to step 6 in a), leaving the Chromium mask on top of the magnetic thin film. In step 5 the structure is imprinted in the thin film by Argon-milling, removing the material in the non-covered parts of the sample, resulting in Iron and therefore magnetic material only present in the covered structures. In step 6 the Chromium mask is removed by a wet etch.

mentioned above, on top of the sputtered δ-doped Palladium thin film a positive resist, called ZEP520A, is spin coated (figure 3.2b step 1). ZEP520A is a solution of Methyl Styrene and Chloromethyl Acrylate Copolymer, desolved in Anisole[48, 49]. At the CFN we used ZEP520A(1:1), this is a solution of equal parts of ZEP520A and Anisole. After spin coating ZEP520A(1:1) for 45 sec at 4000 rpm, we bake the thin film with the spin coated resist for 3 min at 180°C on a hot plate. This recipe ensures a resist thickness of roughly 150 nm[50, 51].

The intended nano-structure will be written in the resist by EBL, employing the JEOL JBX6300FS at the CFN. All kinds of structures can be defined with a CAD-program and then written by the Electron Beam Lithograph, achieving gaps between the structures of as little as 15 nm. After the writing process the resist needs to be developed, therefore we put the sample first for 90 sec in Amyl Acetate and then for another 45 sec in Isopropanol. The resist which was exposed to the electron beam alters during this development, leaving holes in the resist layer (figure 3.2b step 2).
On top of this a Chromium layer will be deposited, with a thickness of 10-30 nm (figure 3.2b step 3). Afterwards a lift-off process with the Remover 1165 (1-Methyl-2-Pyrrolidinone)[52] takes place. The sample is placed in a bath of Remover 1165, which is heated up to 80°C, leaving the sample there for at least 10 min will resolve the resist completely and leaving the Chromium structure on top of our magnetic thin film (figure 3.2b step 4).

To imprint the desired structure in the magnetic film, Argon-milling is utilized (figure 3.2b step 5). The sample surface is bombarded by Argon atoms removing the surface atoms slowly over time. This process has to be done continuously until the Iron is removed from all the parts which were not protected by the Chromium mask. After 10-15 min enough material from the none covered sample area is removed and the Chromium-layer still protects the covered parts of the sample.

The final step is a wet-etch to remove the Chromium from the sample (figure 3.2b step 6), this step is especially important for the PEEM investigations as this technique is surface sensitive (see section 3.4.2). Chromium etch 1020 is a Ceric Ammonium Nitrate system[53], which is heated up to 40°C on a hot plate, the sample is placed for 1 min in the acidic liquid and rinsed thoroughly afterwards with deionized water. The resulting sample has magnetic material (mono-layers of Iron) only in the parts of the sample which were not milled by the Argon due to the Chromium protection mask. All artificial spin ice structured discussed in this thesis are produced in this way. For PAPER-II the width, $w = 150$ nm, and the length, $l = 330$ nm, of the patterned nano-islands did not follow the 1:3 ratio, as mentioned in section 2.2.1, and was approaching rather an 1:2 ratio. For PAPER-III and PAPER-IV the aspect ratio is as intended 1:3 with $w = 150$ nm and $l = 450$ nm. Of course in PAPER-III and PAPER-IV we did not investigated a square ASI lattice, we intend to tailor the magnetic order in square ASI, the applied modifications are described in section 2.2.2.

3.3 Thermally active mesospins

Recently the field of ASI switched its focus more towards thermally active systems[8] and all the possibilities and difficulties this approach carries with it. As many other groups try to achieve thermal activity by reducing the thickness of Permalloy[17, 12, 16, 54] we use δ-doped Palladium, to achieve a wide temperature range of thermal fluctuations[55, 11, 2, 14]. The thermally active temperature region for nano-patterns is defined as the temperatures in which the mesospins are in a superparamagnetic state. In this state the magnetization direction is changing rapidly, due to the fact that the magnetization reversal energy barrier is smaller or at least in the order of the thermal energy, $k_B T$. Therefore the energy provided by the ambient temperature is enough
to switch the magnetization direction. The magnetization reversal energy is
mainly attributed to the shape-anisotropy of the object and therefore can be
found as equation (3.1)[56]:

\[ E_B = \frac{1}{2} \cdot \mu_0 \cdot M(T)^2 \cdot V \cdot \Delta N \] (3.1)

The volume, \( V \), is given by the patterning and the thickness of our mag-
netic layer. \( \mu_0 \) is the vacuum magnetic permeability and therefore constant.
\( \Delta N \) is the difference of the in-plane demagnetization factors, which can be
calculated depending on the shape of the magnetic islands with the Osborn
methodology[57]. The temperature dependent magnetization, \( M(T) \), can be
calculated for our \( \delta \)-doped Pd(Fe) with equation (3.2):

\[ M(T) = M_0 \cdot \sqrt{1 - \frac{T}{T_C}} \] (3.2)

Where \( M_0 \) is the magnetization at 0 K and \( T_C \) the Curie temperature.

**Demagnetization factor**

In his publication "Demagnetizing Factors of the General Ellipsoid"[57] from
1945, Osborn calculates the demagnetizing factors of ellipsoids with different
relations for the ellipsoidal axes. He provides numerical as well as analytical
expressions for different geometries. Our patterned nano-structures, which
are in fact stadium shaped (see section 2.2.1), can be roughly considered as an
ellipsoid, with the ellipsoidal semi-axes \( a \), \( b \) and \( c \). While \( a = \frac{l}{2} \), \( b = \frac{w}{2} \) and
\( c = \frac{z}{2} \) are pre-defined by our lithography, the following relation between the
semi-axes of the ellipsoid is given:

\[ a \geq b \gg c \]

Corresponding to:

\[ \text{length} \geq \text{width} \gg \text{height} \]

of the magnetic islands. The demagnetization factors in the corresponding
directions are:

\[ N_a = \frac{c}{a} \sqrt{1 - x^2} \frac{E_{int}^I - E_{int}^II}{x^2} \] (3.3)

\[ N_b = \frac{c}{a} \frac{E_{int}^II - (1 - x^2)E_{int}^I}{x^2 \sqrt{1 - x^2}} \] (3.4)

\[ N_c = 1 - \frac{c}{a} \frac{E_{int}^II}{\sqrt{1 - x^2}} \] (3.5)
Where $E_{int}^{I;II}$ are the complete elliptic integrals of the first and second kind. $x$ is the argument for these integrals, given as:

$$x = \sqrt{1 - \frac{b^2}{a^2}}$$

The elliptic integral of the first kind can be written as:

$$E_{int}^{I}(x) = \int_{0}^{\frac{\pi}{2}} \frac{d\theta}{\sqrt{1 - x^2 \cdot \sin^2 \theta}} = \int_{0}^{1} \frac{dt}{\sqrt{(1 - t^2) \cdot (1 - x^2 \cdot t^2)}} \tag{3.6}$$

and the elliptic integral of the second kind as:

$$E_{int}^{II}(x) = \int_{0}^{\frac{\pi}{2}} \sqrt{1 - x^2 \cdot \sin^2 \theta} \cdot d\theta = \int_{0}^{1} \frac{\sqrt{1 - x^2 \cdot t^2}}{\sqrt{1 - t^2}} \cdot dt \tag{3.7}$$

Applying the island size, used for most of our investigations, with $2a = l = 450 \text{ nm}$, $2b = w = 150 \text{ nm}$ and $2c = z = 2 \text{ nm}$ to equations (3.3)-(3.5) we calculate the following demagnetizing factors:

$$N_a = 0.0024$$
$$N_b = 0.0125$$
$$N_c = 0.9852$$

These values indicate to some extend the restrictions provided by the shape anisotropy. As the magnetic height of our islands is roughly two orders of magnitude smaller than their length and width, the magnetization is highly unlikely to rotate out of the XY-plane. Considering this restriction the demagnetization factor $\Delta N$ for the reversal energy barrier (3.1) only takes $N_a$ and $N_b$ into account, providing the magnetization reversal processes will only rotate the magnetization in the XY-plane and therefore the demagnetizing factor is given as:

$$\Delta N = |N_a - N_b| = 0.0101 \tag{3.8}$$

**Blocking temperature of patterned islands**

In 1949 Néel[58] and later Brown[59] developed the idea of a relaxation time $\tau$ for single particles[60, 61]:

$$\tau = \tau_0 \cdot e^{\frac{E_B}{k_B \cdot T}} \tag{3.9}$$

where $E_B$ is the energy barrier from equation (3.1), $k_B$ the Boltzmann constant, $T$ the temperature and $\tau_0$ the inverse attempt frequency. This inverse attempt
frequency represents the relaxation time at the Curie temperature of the material. At $T_C$ the energy barrier (3.1) will be zero, since the magnetization (3.2) drops to zero, therefore equation (3.9) reduces to

$$\tau(T \geq T_C) = \tau_0$$

The value of $\tau_0$ is generally given as $10^{-10}$ s, prompting from the gyromagnetic resonance frequency for a single-domain particle[59].

If the relaxation time is equal to the observation time

$$\tau \approx \tau_{\text{observation}}$$

then is a single-domain particle changing its magnetic configuration during the observation time, $\tau_{\text{observation}}$. This means that the thermal energy

$$k_B \cdot T \approx E_B$$

is in the order of the reversal energy barrier (3.1). The respective temperature is then assumed to be the blocking temperature, derived from equation (3.9):

$$T_B = \frac{E_B}{k_B \cdot \ln\left(\frac{\tau_{\text{observation}}}{\tau_0}\right)} \quad (3.10)$$

### 3.3.1 Blocking temperature of δ-doped Pd(Fe) mesospins

To determine the blocking temperature (3.10) a graphical solution can be applied. Therefore the thermal energy, $k_B T$, and the reversal energy barrier, $E_B$, are plotted over the temperature. The thermal energy has to be scaled by the observation time like it is done in equation (3.10):

$$E_{\text{thermal}} = \ln\left(\frac{\tau_{\text{observation}}}{\tau_0}\right) \cdot k_B \cdot T \quad (3.11)$$

The observation time is taken to be 300 s, as this is the measurement time for a PEEM-image. $E_B$ is taken from equation (3.1) and the intersection with (3.11) defines the blocking temperature, see figure 3.3a). To calculate the energy barrier we assume our standard island size, with $l = 450$ nm, $w = 150$ nm, $z = 2$ nm and 2.0 mono-layer of Iron resulting in a $T_C = 400$ K. The only unknown parameter from (3.1) is $M_0$. The magnetization at 0 K, is given by the magnetic moment at 0 K per volume, which has been determined by SQUID measurements on a reference sample and is $M_0 = 6.98 \cdot 10^5$ A/m.

As one can see in figure 3.3a) (blue line) the blocking temperature we calculate with the given values is around 285 K. The image 3.3b) clearly shows, that islands with exactly the same specifications used for the calculation of the blue line in 3.3a) are mainly in their superparamagnetic phase (at a measuring
Figure 3.3. a) Graphical solution for the blocking temperatures of two different island sizes, with their full (line) and by 50% reduced (doted line) magnetization. b) Short-islands measured with PEEM using the XMCD at 135 K. From 295 uncoupled islands in the picture only 56 islands could be identified (as black or white), therefore 81% of the islands are flipping their magnetization during the 5 minutes acquisition time. c) Long-islands measured with PEEM using the XMCD at 135 K. From 192 uncoupled islands in the picture 141 islands could be identified (as black or white), therefore only 27% of the long-islands are flipping during the 5 minutes measurement time.

temperature of 135 K). To scale our calculated blocking temperature with the actual observation, we need to reduce the magnetic moment (infect scaling the magnetization) of the islands by 50%, achieving a blocking temperature of 150 K. There are multiple reasons for the failure of the Osborn approach, first of all the whole demagnetization factors rely on ellipsoids, what is a rough estimate of our stadium like islands. Furthermore this approach assumes a uniform magnetization direction (rigid spins), as it does not take into account any kind of curling of the magnetization of an island, or other inner island excitations. Therefore all dynamics in the magnetization reversal process within the island are neglected. While the scaling of the magnetic moment by 50% can capture the $T_{B2}$ of the short-islands, the same method applied to a long-island ($l_{long} = 1110$ nm) provides a blocking temperature of $T_{B1} = 265$ K. The experimental observation in 3.3c) on the other hand shows already thermal activity among the long-islands at a measurement temperature of 135 K. These experimental results in 3.3b)-c) illustrate clearly the difference in the energy barrier between short- and long-islands. The Osborn methodology overestimate these differences and therefore this theoretical description fails to capture the thermodynamic response of our system.
3.3.2 Two blocking temperatures in one array

In arrays like the Shakti (see section 2.2.3) or the Saint George (see section 2.2.4) lattices, two differently-sized elements co-construct the lattice, their different lengths result in different energy-scales for these two elements. While the magnetization of the two islands does not differ, the demagnetization factors do, resulting in two different blocking temperatures. As the Osborn methodology fails to account for this difference accurate, only a phenomenological description of this effect can be given, see illustration 3.4.

Assuming the SH and mSH lattices at high temperatures (right side of figure 3.4), both lattices and both island types are in their paramagnetic state. Reducing the temperature and crossing the Curie temperature, $T_C$, both short- and long-islands are in their superparamagnetic state. At this stage the islands act as mesospins, rapidly changing their magnetization direction. Lowering the temperature until the first blocking temperature, $T_{B1}$, slows down this rapid magnetization reversal constantly. At $T_{B1}$ the magnetization reversal energy barrier of the long-islands becomes larger then the thermal energy, resulting in a blocked magnetization direction of the long-mesospins. While the long-mesospins are thermally stable, the short-mesospins are still in their superpara-

![Figure 3.4](image-url)
magnetic state, as experimentally shown with the PEEM-images in figure 3.3. Reducing the temperature below the blocking temperature of the short-islands, $T_{B2}$, the magnetization direction of the short-mesospins is blocked as well, resulting in a thermally stable magnetization for all island sizes. Taking these different length- and especially energy-scales into account, the magnetic ordering processes in SH and mSH lattices should result in different ordering schemes.

### 3.4 Measurements and analysis

Different measurement and analysis tools have been employed throughout this work. To verify the Curie temperature of the $\delta$-doped Pd(Fe) the Magneto-Optical Kerr-Effect (MOKE) has been used to measure temperature dependent hysteresis loops of the thin films. The patterned nano-structures were structurally characterized by means of Atomic-Force-Microscopy (AFM) and Scanning-Electron-Microscopy (SEM). The magnetic characterization of the patterned arrays is more challenging, while athermal systems like the one in PAPER-I can be rather easily measured by Magnetic-Force-Microscopy (MFM), thermal systems investigated throughout the rest of this work are in need of measurement techniques which are not altering the magnetic order in the system. MFM measures the force acting on the magnetic tip, due to the magnetic interactions between the magnetic sample and the tip. While athermal systems need rather high energies to overcome the reversal energy barrier ($E_B$), thermally active systems can easily alter their magnetic order with the stray field of the magnetic tip. Low temperature MFM could provide a possibility of imaging the frozen state of a thermal system, but low temperature MFM require large experimental efforts and the scanning area in low temperature MFM instruments is restricted to a few micro-meters. Therefore large statistics are hard to achieve with low temperature MFM systems and other measurement techniques need to be explored.

To investigate the magnetic relaxation of a square ASI, PAPER-II, we employed a custom built SQUID-magnetometer (see section 3.4.1). The main investigation technique used throughout this work (PAPER III-IV) is Photoemission Electron Microscopy (PEEM) using the X-Ray Magnetic Circular Dicroism (XMCD) to visualize the magnetization direction of the individual islands, further explained in section 3.4.2.

For energy calculations, for the different vertex states, we used a micromagnetic calculation software called MUMAX3[43]. Beside standard statistical tools, the spin structure factor (SSF) was applied to identify magnetic ordering in our nano-arrays, further details about the SSF are explained in section 3.4.3.
3.4.1 Magnetic relaxation

To measure the magnetic relaxation of nano-structures like the ones discussed in PAPER-II, "Thermally induced magnetic relaxation in square artificial spin ice", it is important to have large arrays of the desired structures. We investigate extended arrays of about $2 \times 2 \, \text{mm}^2$ with two different lattice parameters for the square ASI lattice. Since the magnetic signal of our $\delta$-doped Pd(Fe) is very low, we employed a custom built SQUID-magnetometer, which was developed for studies of static and dynamic magnetic properties of various materials[62].

The square ASI, used for the relaxation measurement, need to be in a well defined state from which the array can relax into its ground state. An illustration of the experiment is given in figure 3.5. Starting at room-temperature (position I in figure 3.5), the array is in its superparamagnetic regime, under a magnetic field of 800 A/m applied in the [11]-direction of the array the sample is cooled down. When crossing the blocking temperature ($T_B$), the magnetization of the islands aligns within the external magnetic field, resulting in a dressed state out of Type-II$_4$ vertices (position II in figure 3.5). The sample is cooled down with 5 K/min until the desired measurement temperature and there the temperature is stabilized (waiting time $t_0$ until $t_s$ is 300 s). At the moment the magnetic field is switched off (position III in figure 3.5) the time dependent measurement of the magnetization starts. Due to internal processes the first measurement point is taken after 0.3 s. The whole relaxation experiment, at one temperature, is taken for 3000 s. In the lower graph in figure 3.5 the yellow region identifies this measurement window. The first magnetization reversals take place during the first 0.3 s of the measurement and therefore they will not be detected. The first mesospin flip (position IV in figure 3.5) creates two Type-III$_4$ vertices out of two Type-II$_4$ vertices, therefore an energy barrier needs to be overcome to start the relaxation process into an energetically more favorable state, Type-I$_4$. From this first reversals the system is energy driven by the formation of Dirac-strings[17, 41] (position V in figure 3.5), which are in fact Type-I$_4$-chains and therefore lowering the energy of the array. From here the measured magnetization of the square ASI array is reducing further and further (position VI in figure 3.5) until the magnetization is zero (position VII in figure 3.5). Position VII in figure 3.5 does not mean the square ASI array has reached its ground state, but 50% of the islands which were aligned with the external field have flipped their magnetization resulting in an overall spin arrangement of 50% of the islands still pointing in the [11]-direction, while the other 50% pointing in the opposite ([1-1]) direction.
Figure 3.5. The upper graph represents the temperature and field evolution during the measurement protocol. Three vertex states are shown in the top right corner. The lower graph represents the relaxation dynamics of the magnetization, M, of such an ASI array. Panels (I)-(VII) represent the different stages of the array during the experiment. The magnetization of the array decays over time until the overall magnetization is zero and the array goes from a dressed magnetic state (III) into a relaxed state. The relaxation curve represents a measurement temperature of 200 K and the yellow region indicates the measurement window. The magnetic configuration of the array relaxes by forming strings[17, 41] (V). Over the time evolution the magnetic configuration of the array relaxes more and more (VI) until its magnetization reaches zero (VII). As indicated in (VII) the array is not fully ordered at this time, but no further information can be acquired from the relaxation measurement since the average magnetization is zero.
Repeating this measurement for different temperatures and for two different lattice parameters, allows us to plot time versus temperature maps, indicating the magnetization of the array by colors, see figure 3.6.

Figure 3.6. Time and temperature dependent relaxation of the magnetization of square ASI arrays. The different lattice parameters of 380 nm (a) and 420 nm (b) show clear shifts of the relaxation processes. The gray dotted line identifies the measurements at 190 K. The gray dashed line highlights the magnetization after 300 s for each relaxation measurement.

The two different lattice parameters represent an altered coupling between the islands in the two arrays, a larger lattice parameter leads to less coupled islands. The coupling on the other hand reduces the ability of an island to reverse its magnetic orientation. One could view this as adding an island interaction term to $E_B$ (equation (3.1)) and by doing so increasing the blocking temperature of the non-coupled islands. A stronger coupling leads therefore to a higher blocking temperature which is clearly visible comparing figure 3.6a) and 3.6b). In figure 3.6a) the relaxation process advances earlier in time and temperature. Looking at 190 K (gray dotted line) in 3.6a) the magnetization varies from 0.9 to 0.6 during the measurement interval, leaving a reduction of the magnetization of about 30%. While in 3.6b) the magnetization drops from 0.8 to 0.4, in the same measurement interval. On the other hand if we focus on a certain relaxation time, like the gray dashed line in figure 3.6, the magnetization of the array has dropped down to below 0.3 at roughly 225 K for the stronger coupled array, figure 3.6a), while the weaker coupled, figure 3.6b), reaches this magnetization stage already at roughly 205 K for a relaxation after 300 s.

Figure 3.6 presents the wide temperature range of thermal activity in magnetic nano-structures produced out of $\delta$-doped Pd(Fe). The Iron thickness as well as the lattice geometry will define the actual temperature range in which the arrays are thermally accessible.
3.4.2 Photoemission Electron Microscopy

To investigate magnetic order and frustration in magnetic nano-structures, local magnetic imaging tools are useful. As our magnetic nano-islands are in the sub-micrometer regime, optical light microscopy like Kerr-microscopy are failing in capturing the local magnetic orientation of the islands, due to the diffraction limit[28]. While MFM has the resolution we need, the interaction between magnetic tip and sample is a crucial parameter. Especially in thermally active systems, which are studied here, small perturbations like the tip-sample interaction can lead to variations in the investigated magnetic state. Therefore only at temperatures far below the blocking temperature of the islands, a measurement with MFM would be possible. As our statistical analysis methods require large areas to be investigated, low temperature MFM is limited in its field of view and makes it very difficult to acquire substantial data. Therefore the choice of a local probe measurement is the Photoemission Electron Microscopy (PEEM) using the X-Ray Magnetic Circular Dichroism (XMCD).

PEEM imaging

PEEM is a technique developed in the 1930s[63] as a laboratory instrumentation, using ultra-violet lamps as a radiation source. The PEEM measurements on which this work is based are done at Synchrotron facilities. The general idea of PEEM is that the incident radiation excites core electrons and sufficient electron optics detect the created secondary electrons. As the energy

![Figure 3.7. PEEM image indicating the areas where the two spectra were acquired. The red and blue rectangle represent the area where the respective spectra was taken. The blue region is on a marker of the sample and therefore inhabits 2.0 monolayer of Fe, while the red area is a milled region where no Fe should be left. The blue graph indicates clearly the presence of Fe, with the element specific peak maximum at 707.5 eV. The red graph on the other hand lacks this peak, indicating no Fe is in this part of the sample.](image)
of the incident radiation is the key for the emission of the electrons, an energy selective incident radiation allows for element specific microscopy. Parts of the sample, inhabiting a specific element, will emit more electrons than parts where this element is absent and therefore elemental contrast can be achieved using PEEM[64, 65, 66]. With the broad energy ranges provided by a Synchrotron, element specific investigations can be carried out easily. As the L\textsubscript{3}-edge of Fe corresponds to 706.8 eV[67] and lays within the provided energy range of the Synchrotrons our δ-doped Pd(Fe) is a suitable candidate for this kind of investigations, as shown in figure 3.7. But of course an element specific PEEM measurement does not provide the magnetic orientation of the measured islands. Therefore XMCD has to be applied and the incident radiation needs to be polarized.

**XMCD at the Fe L\textsubscript{3}-edge**

For the XMCD effect the incident radiation needs to be circularly polarized. The polarization of the radiation defines the probability of excitation of electrons from magnetic domains parallel or anti-parallel to the incident beam direction. Therefore the main magnetization direction needs to be parallel to the incident X-ray direction. As our square ASI-based magnetic nano-structures have 90° between the two magnetization directions, an alignment of the lattice in the [11]-direction is recommended. Therefore all islands have a magnetic component in the X-ray direction. Depending on the magnetic orientation of the islands, the intensity value will differ between the two polarization states, circular left and circular right polarized X-rays. These information is saved in each pixel of the corresponding PEEM images. To achieve magnetic contrast the following operation has to be executed with the left circular polarized, \( I^l \), and right circular polarized, \( I^r \), PEEM-image:

\[
I_{\text{magnetic}} = \frac{I^l - I^r}{I^l + I^r}
\]  

(3.12)

![Figure 3.8. PEEM-images of the SH lattice with a lattice parameter of 600 nm. 50 averaged PEEM images measured with circular left, a), and circular right, b), polarized X-rays at an energy of 707.2 eV. c), The magnetic contrast resulting from equation (3.12) applied on image a) and b). The magnetic orientation of all measured mesospins can be identified uniquely, d.](image-url)
The results are presented in figure 3.8. The magnetic orientation of all islands can uniquely be identified. PEEM using XMCD suits as a perfect local imaging tool to determine the magnetic ordering in our nano-arrays, without any perturbation of the investigated magnetic configuration.

3.4.3 Spin Structure Factor

The local ordering, like the abundance of the different vertex states in a lattice, are best to identify with local probing techniques like PEEM. However global ordering, expanding over several lattice parameters is often difficult to identify with local probe experiments. Global measurements like X-ray or neutron scattering on the other hand can be used to investigate global ordering. The experimental realization of scattering experiments on magnetic nano-structures as we investigate here are very demanding and therefore have not been carried out in the scope of this thesis. But reciprocal space analysis tools like the magnetic spin structure factor (SSF) are a suitable way to apply scattering analysis techniques to our real space, locally measured, data. The SSF simulates a neutron scattering experiment on a given spin arrangement. As our PEEM-measurements represent rather large statistics with over 4000 investigated mesospins per array, the application of the SSF on this data allows us to identify specific magnetic ordering among the different lattices. The general equation to calculate the SSF of a given spin arrangement is:

$$I(q) = \frac{1}{N^2} \sum_{i,j=1}^{N} S_i^\perp \cdot S_j^\perp \cdot e^{iq(r_i-r_j)}$$  \hspace{1cm} (3.13)

With $I(q)$ being the intensity value for a certain scattering vector $q$, $N$ is the number of all spins over which the SSF is calculated, $S_i^\perp$ represents the spin component perpendicular to the $q$ vector and $r_i$ is the position of the corresponding spin in the lattice.

**SSF calculation simplification**

As equation (3.13) consists of a product of two sums, for each $q$ vector in the SSF map, the computations can be rather heavy depending on the size of the spin system and the $q$ resolution. To shorten the numerical calculations a few simplifications can be applied to reduce a $N^2$ problem to a problem with the complexity of $N$. Firstly the exponential can be separated using

$$e^{iq(r_i-r_j)} = e^{iqr_i} \cdot e^{-iqr_j}$$

leading to equation (3.14)

$$I(q) = \frac{1}{N^2} \sum_{i=1}^{N} \sum_{j=1}^{N} S_i^\perp \cdot S_j^\perp \cdot e^{iqr_i} \cdot e^{-iqr_j}$$  \hspace{1cm} (3.14)
In equation (3.15) the two sums are separated

\[ I(q) = \frac{1}{N^2} \sum_{i=1}^{N} S_i^\perp \cdot e^{i q \cdot r_i} \cdot \sum_{j=1}^{N} S_j^\perp \cdot e^{-i q \cdot r_j} \quad (3.15) \]

Using now Euler’s formula:

\[ e^{i \phi} = \cos(\phi) + i \cdot \sin(\phi) \quad (3.16) \]
\[ e^{-i \phi} = \cos(\phi) - i \cdot \sin(\phi) \quad (3.17) \]

we can find equation (3.18).

\[ I(q) = \frac{1}{N^2} \left[ \sum_{i=1}^{N} S_i^\perp \cdot \cos(q \cdot r_i) + i \cdot \sum_{i=1}^{N} S_i^\perp \cdot \sin(q \cdot r_i) \right] \cdot \left[ \sum_{j=1}^{N} S_j^\perp \cdot \cos(q \cdot r_j) - i \cdot \sum_{j=1}^{N} S_j^\perp \cdot \sin(q \cdot r_j) \right] \quad (3.18) \]

The choice of the variables \( i \) and \( j \) are arbitrary and therefore all \( j \) can be written as \( i \). With the following substitutions:

\[ A = \sum_{i=1}^{N} S_i^\perp \cdot \cos(q \cdot r_i) \quad (3.19) \]
\[ B = \sum_{i=1}^{N} S_i^\perp \cdot \sin(q \cdot r_i) \quad (3.20) \]

equation (3.18) will transform to:

\[ I(q) = \frac{1}{N^2} [A + i \cdot B] \cdot [A - i \cdot B] \quad (3.21) \]

With trivial algebraic operations, equation (3.21) can be simplified to

\[ I(q) = \frac{1}{N^2} [A^2 + B^2] \quad (3.22) \]

Using the substitutions (3.19) and (3.20) the intensity values for every desired \( q \) values can be calculated, resulting in the typical spin structure factor maps.
SSF of square ASI

Applying this technique to our two-dimensional mesospin systems gives us a great insight into the order in the different mesospin architectures. The SSF map of the ground state arrangement in a square ASI systems is shown exemplary in figure 3.9.

![Figure 3.9](image)

**Figure 3.9.** a) Square ASI mesospin arrangement for 5x5 vertices in a ground state configuration (60 mesospins). The lattice parameter is assumed to be \( p = 600 \text{ nm} \) and the real space lattice vectors are \( \mathbf{a}_{1,2} = p \cdot \mathbf{e}_{x,y} \), where \( \mathbf{e}_{x,y} \) is the unit vector in the x- or y-direction. The reciprocal lattice vector is therefore \( \mathbf{b}_{1,2} = \frac{2\pi}{p} \mathbf{a}_{1,2} = \frac{2\pi}{p} \mathbf{e}_{x,y} \). b) The resulting spin structure factor map, calculated from this limited spin arrangement.

The SSF for the degenerate square artificial spin ice system[23], with degenerate Type-I\(_4\) and Type-II\(_4\) vertices, is changing its appearance and is characterized by the so called pinch points at \( \mathbf{q} = (1,1) \)[24]. Real space analysis on ASI systems is mainly based on vertex statistics and therefore it is useful to look into the SSF maps of the different vertex states, illustrated in figure 3.10. The vertices can be seen as unit cells of the extended lattice and each vertex configuration has its own characteristic SSF map.

The Type-I\(_4\) and Type-IV\(_4\) vertices are rotational invariant, while the SSF maps of Type-II\(_4\) and Type-III\(_4\) have two different orientations, depending on the direction of the residual magnetic field of the vertex.
SSF of all four different vertex states. The SSF was calculated on the above shown mesospin arrangement, the four mesospins resemble the different vertex energies of the square ASI vertices. The SSF maps of these four mesospins are calculated following the same real space and reciprocal space lattice vectors described in the caption of figure 3.9.

SSF of square ASI based nano-structures

Due to the assumed point-like spins for the calculation of the SSF all SSF maps of square ASI based mesospin structures are unique, because of the different positions of the mesospins in the lattice. In the mSASI the interaction modifier is added in the middle of the vertex, for the SH and SG lattices the position of the long-mesospin is altered in comparison to the mSH or mSG lattices. The value and position of the interaction modifier or long-mesospin are the major reason for the differences in the SSF of these lattices, as illustrated in figure 3.11 for the Shakti and Saint George lattices. The artifacts observed here can not uniquely identified to be originated from specific order among the Ising-mesospins, it is mainly created from the different positions of the mag-

Figure 3.10. SSF of all four different vertex states. The SSF was calculated on the above shown mesospin arrangement, the four mesospins resemble the different vertex energies of the square ASI vertices. The SSF maps of these four mesospins are calculated following the same real space and reciprocal space lattice vectors described in the caption of figure 3.9.

Figure 3.11. SSF maps calculated for square ASI (p = 660 nm), mSH, SH, mSG and SG (all p = 600 nm). The real space lattice parameters for the square ASI are $a_{1:2} = p \cdot e_{xy}$ and being four times larger for all the other lattices ($a_{1:2} = 4p \cdot e_{xy}$), resulting in the reciprocal lattice units (r.l.u.) used for the normalization of the $q$ axes. A direct comparison between these different SSF maps is challenging.
netic elements. To be able to compare the SSF of different lattices, especially SH and mSH or SG and mSG lattice, investigations of identical sub-lattices are a major tool used in this thesis.

**SSF of sub-lattices**

While the choice of a sub-lattice in the mSASI is straightforward, for the SH and SG lattices multiple possibilities are available. As the square ASI is a well known reference system, the SSF sub-lattice for the mSASI is just the square ASI lattice, ignoring the interaction modifier for the calculations of the SSF. We compute the SSF for our mSASI on the square ASI bases, which can be easily compared to the square ASI and therefore square ASI analysis schemes can also be applied to the mSASI. A similar approach was chosen for the investigation of the Shakti lattices, by calculating the SSF for only short-mesospins in the SH lattice, which resemble the four-fold coordinated vertices. We can compare the results to the same sub-lattice in the square ASI and even more important to the same sub-lattice in the mSH lattice. The comparison between the SSF maps of the mSH and the SH lattice reveal the influence of the long-mesospins on the ordering of the short-mesospins around them. The same analogy was used for the Saint George lattices, but due to the geometrical arrangement of the mesospins, the SSF map of the short-mesospins in the SG lattice is a sub-lattice of three-fold coordinated vertices. Therefore a direct comparison to the square ASI is not feasible. But the comparison between mSG and SG lattice presents again the influence of the long-mesospins on the ordering of the short-mesospins and the thermodynamically induced differences of these two lattices. The calculated sub-lattices are indicated in figure 3.12 for all the investigated lattices.

*Figure 3.12. SEM images of the investigated lattice types, from left to right: mSASI, mSH, SH, mSG and SG. The respective sub-lattices, used for the SSF, are marked with red. The real space lattice vectors which are used for the respective lattice are indicated in each lattice as well. While the mSASI has the same lattice vectors as the square ASI (described in the caption of figure 3.9), all the other lattices have four times as long real space vectors, resulting in reciprocal lattice vectors $b_{1,2} = \frac{2\pi}{a_{1,2}} = \frac{\pi}{2p} e_{x,y}$ for these lattices.*
**Reduced real space input**

SSF maps of large spin arrangements are just one snapshot of one possible microstate. Multiple snapshots should be averaged to eliminate microstate specific ordering artifacts. As we can not repeat our PEEM-XMCD measurements several times, we use a different approach. Instead of making multiple images, we produce one large PEEM-image of our sample, as this image expand over tens of micrometer we are able to calculate multiple SSF with smaller real space input out of this one microstate. For the SH lattice for example, we calculate the SSF using the short-mesospin sub-lattice. Instead of calculating the SSF of all 900 four-fold coordinated vertices and their respective 3600 mesospins, we reduce the real space size to one, four or nine four-fold coordinated vertices. Using this approach we increase the number of measured microstates, by reducing the spatial extension we investigate with one SSF. An averaged SSF of a real space input of just one vertex, four mesospins, resembles the vertex statistics of these four-fold coordinated vertices. The averaged SSF for four or nine four-fold coordinated vertices defines the real space distances up till which magnetic ordering is investigated.
4. Results and Discussion

The results presented in this section are obtained by PEEM-XMCD imaging and the analysis of these measurements. The SSF as a reciprocal space analysis tool was additionally used. In section 4.1 results concerning PAPER-III are presented and in section 4.2 the main findings of PAPER-IV are shown, together with additional informations. Section 4.3 presents unpublished results investigating the Saint George lattice.

4.1 Modified square artificial spin ice

We investigated four mSASI systems and their respective square ASI references for three different lattice parameters. These investigations give us an overview on the influence of the magnetic interaction modifier on the ordering in square ASI systems. As the square ASI can be viewed as a mSASI lattice with a dot size of zero, we will further use this nomenclature. The fifteen investigated lattices are, for the lattice parameter \( p = 660 \) nm lattices including interaction modifier of the following diameters \( D_{dot} = [0 \text{ nm}, 120 \text{ nm}, 130 \text{ nm}, 150 \text{ nm}, 180 \text{ nm}] \) and for \( p = [720 \text{ nm}, 800 \text{ nm}] \) the interaction modifier has diameters of \( D_{dot} = [0 \text{ nm}, 130 \text{ nm}, 150 \text{ nm}, 180 \text{ nm}, 200 \text{ nm}] \). Our goal is to tailor the magnetic order in the designed structures, therefore we view the interaction modifier as an interaction bridge and not as a separate magnetic element. We are not interested in the magnetization state of the modifier per se, our interest lays in the ordering of the Ising mesospins. Therefore a mapping of the vertex states can be done accordingly to the square ASI states, illustrated in figure 2.6.

Vertex statistics

Imaging the frozen state of a mSASI with different sized interaction modifiers reveals the change in the energy relations between the different vertex types. While the Type-III\(_4\) and Type-IV\(_4\) vertex counts stay roughly the same for each lattice parameter and changing interaction modifier diameter, the vertex counts for Type-I\(_4\) and Type-II\(_4\) vertices alter drastically. The statistics for all three lattice parameters are presented in figure 4.1.
The general characteristics between different lattice parameters can be identified as in previous work[14]. Due to weaker coupling, the amount of higher excitations increased while the lattice parameter raises. The crossover from a Type-I₄ dominated state to a Type-II₄ dominated state, depending on modifier diameter is indicated in all graphs with a dashed line and varies depending on the lattice parameter. One might assume the gap between the modifier and the mesospins would characterize this crossover, but the vertex statistics reveal that the crossover takes place at different gap sizes for the different lattice parameters. This indicates the influence of the magnetization of the interaction modifier on the Ising mesospins. Vertex statistics only reveal the local ordering on the vertex level, but do not reveal medium- or long-range order in our lattices.

Figure 4.1. Vertex abundance for the different lattice parameters, $p$, depending on the diameter of the interaction modifier. The square ASI at diameter zero represents the Type-I₄ dominated state. While increasing the diameter of the interaction modifier, the distribution of the vertex states is changing, accordingly to the change in the energy relations between the different vertex states. The crossover of Type-I₄ and Type-II₄ vertex energies is indicated with a dashed line for each lattice parameter.
Vertex maps

A real space representation of the measurements is given in figure 4.2, representing all measured vertices in colors, depending on their vertex state.

![Figure 4.2](image)

**Figure 4.2.** Vertex maps of the measured arrays. The respective vertex states are illustrated by colored diamonds, green - Type-I\_4, blue - Type-II\_4, yellow - Type-III\_4 and red - Type-IV\_4. An increase of Type-IV\_4 vertices is present for larger lattice parameters, while Type-I\_4 domains vanish by adding larger interaction modifier in the arrays.

Similar conclusions as from the statistics in figure 4.1 can be followed up. Additionally to that the spatial placement of the different vertex states can be identified. An important parameter in this regard is the domain size of Type-I\_4 and Type-II\_4 vertices. As the energies change from a Type-I\_4 dominated state to a Type-II\_4 dominated state the Type-I\_4 domains are dropping constantly in size and the Type-II\_4 domain sizes spike up after the energy crossover, as
shown in figure 4.3. But to identify further ordering in this arrays real space analysis are rather inefficient. The use of reciprocal space methods like the SSF is needed and can give a better understanding of the arrangement of the mesospins in these lattices.

Figure 4.3. For the arrays with a lattice parameter of 660 nm the change from a Type-I₄ to a Type-II₄ dominated state is the most drastic. The investigation of domain sizes indicates this change as well. While the Type-I₄ domain size decreases by increased interaction modifier diameter, the Type-II₄ domain size spikes up after the indicated energy crossover.

Spin structure factor

The crossover between the two dominant magnetic ordering regimes can also be identified by the changes in the SSF, shown in figure 4.4. The general SSF map appearance can be recapped by calculating the averaged SSF of the vertices of the lowest energy level, illustrated in figure 4.5. As these lowest energy vertices appear as the most common ordering schemes in these lattices, the general appearance of the SSF can roughly be recovered by the averaged SSF of the lowest energy vertices. But for ordering throughout the lattice more then just the vertex SSF maps need to be calculated.

Looking at the SSF of all different lattices clearly presents a crossing between the Type-I₄ and the Type-II₄ dominated state. But this crossing is actually not an abrupt change in the ordering, as it might seem by the domain size transition in figure 4.3. It is more of a continuous change from a Type-I₄ dominated state to an actually degenerated Type-I₄ – Type-II₄ state to a Type-II₄ dominated state. The intermediate degenerated state is a very interesting spin state, since it recovers the degeneracy of the six-vertex model[23]. The Type-II₄ dominated state represents a completely new class of ASI in which the lowest energy level inhabits a residual magnetic field on the vertex level, therefore new ordered phases emerge. The residual field from a Type-II₄ vertex can be arranged in a flux lattice in different ways, to minimize the total...
Figure 4.4. Representation of the SSF maps calculated from the PEEM-XMCD measurements on the 15 different arrays. A clear transition in the SSF from a Type-I4 dominated states ($D_{dot}$ diameter small) over a degenerated Type-I4 – Type-II4 state ($D_{dot} = 150$ nm for $p = 660$ nm) to a Type-II4 dominated vertex state ($D_{dot} = 180$ nm for $p = 660$ nm) can be seen.

energy of the array. In figure 4.6 a SSF of four simulated Type-II4 tilings is illustrated. The comparison to the experimental results ($D_{dot} = 180$ nm for $p = 660$ nm) reveals that in our measured lattice all these simulated spin ar-
Figure 4.5. The first SSF is an average of the two SSF maps of the two degenerated Type-I\textsubscript{4} vertices and resembles the general appearance of the SSF of the mSASI with small interaction modifier. The second SSF map represents the average of the SSF of the two Type-I\textsubscript{4} and the four Type-II\textsubscript{4} vertices, the averaged SSF illustrates the appearance of a degenerate spin system, without the characteristic long-range ordering peaks. While the last SSF is an average of all four Type-II\textsubscript{4} vertices, showing remarkable similarities to the SSF of the array with $D_{dot} = 180$ nm and $p = 660$ nm. 

...rangements are present, as the simulated SSF reproduces the peak intensities from the measured SSF.

All investigations carried out in this section are ignoring the magnetic state of the interaction modifier, as we only threat it as a channeling object what stays in a slave relation to the dominating Ising mesospins around it. This omitting of an element in our spin system leaves a certain degree of inaccuracy on the attempt of understanding the ordering phenomena in these lattices, therefore spin systems of only well known Ising-like mesospins will be investigated in the following sections.

Figure 4.6. The simulated SSF of four Type-II\textsubscript{4} tilings is shown in a). The simulated tilings are: Type-I\textsubscript{4}-tiling on the emergent flux lattice (b), vortex/anti-vortex pairs (c) and polarized states, forming both straight domains (d) or a herringbone structure (e). f), For the green marked area the simulated SSF (a) captures the experimental peaks of the SSF for the array with $D_{dot} = 180$ nm and $p = 660$ nm.
4.2 Shakti lattice

In the SH and mSH lattices all mesospins are uniquely identifiable and therefore the spin arrangement and their respective ordering can be mapped out by direct imaging techniques like PEEM-XMCD. As explained in section 3.3 the differently sized mesospins, in the SH lattice, have different magnetization reversal energies and therefore different blocking temperatures. The magnetization direction of the long-islands will freeze in at higher temperatures than for the short-islands. Therefore the long-mesospins can be thermally stable while the short-mesospins are still in a superparamagnetic state (figure 3.4). Acknowledging for this fact, the long-mesospins freeze into their magnetization direction while being in a bath of superparamagnetic short-mesospins. A correlation between the long-mesospins seem possible and is the first parameter to investigate.

Long-mesospin correlation

Analyzing the spin arrangements of long-mesospins via the magnetization direction of the short-mesospins, domain structures can be found, see figure 4.7. As the long-mesospins are arranged perpendicular to each other, the two main directions of the long-mesospins need to be investigated.

![Figure 4.7](image)

*Figure 4.7.* Long-mesospin domains in their two orientation directions. The colored background indicates that two ferromagnetically aligned long-mesospins can be connected through the magnetization direction of the short-mesospins in between.

It is obvious in figure 4.7 that long-mesospins are connected with each other via the magnetization direction of the short-mesospins. The colored background between two parallel aligned long-mesospins represents ferromagnetically aligned short-mesospins in between these two long-mesospins. This domain structure implies a long-island correlation, but by calculating the corre-
lation of parallel/anti-parallel aligned long-mesospins, their random magnetic alignment get revealed.

Figure 4.8. Long-mesospin correlations for the two main directions of the long-mesospins. Surprisingly already the first neighbor of a long-mesospin is randomly, either ferromagnetic or anti-ferromagnetic, aligned. The misleading domain representation in figure 4.7 represents actually a random alignment of long-mesospins.

These surprising results set the baseline for further investigations. Especially the comparison between SH and mSH lattice is of high interest, since the short-mesospins align themselves in the proximity of randomly arranged long-mesospins in the SH lattice. The mSH lattice explores its whole ground state manifold without these restrictions.

Figure 4.9. Vertex statistics for mSH (red/orange) and SH (blue/light blue). There are no significant differences between mSH and SH lattice whether in the four-fold (square symbols) or three-fold (triangular symbol) coordinated vertices. With an increased lattice parameter the Type-I vertices decrease and the excited vertex states increases, due to the reduced coupling between the islands, this holds for both subsets of three- and four-fold vertices.
Vertex statistics

As described in section 2.2.2, the Shakti lattices are modifications of the square ASI, with removed and in the SH lattice case merged islands. The Shakti lattices consist therefore out of different vertices. The four-fold coordinated vertex and the three-fold coordinated vertex, for both of lattices and additionally the two-fold coordinated vertex for the mSH lattice. The abundance of the different vertex types for the three- and four-fold coordinated vertices is shown in figure 4.9.

The general behavior of ASI systems can be observed in both of the subsets. With decreased interaction between the mesospins (increased lattice parameter), the lattice is in a higher energy state and therefore less Type-I vertices and more excited vertices are registered, in both the three- and four-fold coordinated vertices. Comparing the abundance of the different vertex types across the SH and mSH lattice there is no significant difference between these two lattices. The only trend which might be identified, is a higher ordering in the three-fold coordinated vertices in the SH lattice, since the Type-I3 counts are slightly higher then in the mSH lattice for the same lattice parameters. Comparing this results with literature data from Gilbert et al.[15], we can note that due to the use of δ-doped Pd(Fe) our samples are in the weakly coupled regime, leading to numerous of excitation in our lattice, which we expect regarding previous work[14].

As the two-fold coordinated vertices are merged to one long-mesospin in the SH lattice, a ferromagnetic alignment of the two-fold coordinated vertices would reproduce the ordering of the SH lattice. Therefore it is noteworthy that over 1/3 of the two-fold coordinated vertices are in their excited anti-ferromagnetic state, see figure 4.10.

On one side, three- and four-fold coordinated vertices resemble the same vertex distributions within their errorbars for SH and mSH lattices, but at the same moment over 1/3 of the two-fold coordinated vertices are in their excited energy state. This raises a legitimate question about the ordering in these two lattices and how these states have been accomplished. The SH and mSH lattices are similar on the three- and four-fold coordinated vertex level. But showing major differences on the two-fold coordinated vertex sides, as such an excited anti-ferromagnetic alignment is not present in the SH lattice.

Figure 4.10. High amount of anti-ferromagnetic two-fold coordinated vertices, as an indication for the different ordering processes taking place in the mSH.
Charge screening or neutralization

As our weakly coupled arrays exhibit various Type-III4 excitations, a way of promoted ordering in these lattice could be through magnetic charge ordering. A charge ordered phase is known from the Kagome spin ice[68, 69], in which all vertices possess a residual charge. In the experimental realization of the Shakti lattices by Gilbert et al.[15], an observed charge screening is reported. Opposite charges are present at all three-fold coordinated vertices around a Type-III4 excitation on the four-fold coordinated vertex. This charge screening is an interesting discovery, taking into account that the charge of the three-fold coordinated vertex is dependent on different mesospins. In the case of a Type-I3 vertex the magnetic charge is dependent on the magnetic orientation of the short-mesospin, which is shared with the four-fold coordinated vertex. For a Type-II3 vertex the charge is defined by the magnetic orientation of the long-mesospin (or one of the mesospins of the two-fold coordinated vertex), which only interacts with the Type-III4 vertex via the stray-field of one of its mesospins. In the case of a Type-III4 excitation three mesospins point in or out of the vertex and the fourth one in the opposite direction, depending on the charge of the magnetic monopole. For a positively charged Type-III4(+) excitation three mesospins point into the vertex and one out of the vertex. Therefore Type-I3(-) vertices, fulfilling charge screening around the Type-III4(+) excitation, can only be located at the three mesospins pointing into the four-fold vertex, while Type-II3(-) vertices can be placed on all three-fold coordinated vertices, since their magnetic charge is defined by the long-mesospin orientation. This fact could lead to the conclusion that a charge screening is an indication for longer range ordering since the charge of a vertex would affect the magnetic orientation of a mesospin not belonging to this vertex.

But in our investigated lattices, due to the weak coupling regime, the charge screening can not be seen, as presented in figure 4.11. Furthermore a local charge minimization is found, as the total magnetic charge of the four-fold

Figure 4.11. The combined effective magnetic charges on the three-fold coordinated vertices neighboring Type-III4 excitations in the SH (blue) and mSH (red) lattice. The magnetic charges on the three-fold coordinated vertices are accounted with respect to the sign of the charge of the Type-III4 vertex. Indicating that the combined charge is normal distributed around the opposite charge of the four-fold coordinated excitation. Resulting in no effective charge for the Type-III4 vertex and its four three-fold coordinated vertex neighbors.
coordinated Type-III4 vertex together with its four surrounding three-fold coordinated vertices sums up to a total magnetic charge of zero. As this is seen in the mSH as well as in the SH lattice the charge screening or charge neutralization is an indication for strong or weakly coupled arrays. As this charge screening is somehow an investigation of vertex neighbors, we want to follow up on this in more detail and not only for the Type-III4 vertices.

Vertex neighbors

The three- and four-fold coordinated vertex statistics are identical for the mSH and SH lattice, but the two-fold coordinated vertices indicate a variation in the ordering between this lattices. Therefore we want to investigate the proximity of the different vertices in our systems. Each of the mesospins in a four-fold coordinated vertex shares a mesospin with a three-fold coordinate vertex, while a three-fold coordinated vertex shares its two parallel mesospins with two different four-fold vertices and the long-mesospin (or the two short-mesospins in the mSH case) with another three-fold coordinated vertex. An-

Figure 4.12. Investigation of the four surrounding three-fold coordinated vertices dependent on the six invariant four-fold coordinated vertex states (from up to down in the following order: Type-I4, Type-II4, Type-III4(+), Type-III4(-), Type-IV4(+), Type-IV4(-)) for the SH (left) and the mSH (right) lattice. No differences between the SH and mSH vertex neighbors are identifiable, geometrical restrictions for Type-III4 and Type-IV4 and their Type-I3 neighbors are noteworthy. There were no Type-IV4 excitations in the SH lattice.
alyzing the vertex neighbors of the four-fold coordinated vertices for both the SH and the mSH lattices reveals no differences between the two lattices, see figure 4.12.

Investigating the neighbors depending on the vertex state of the four-fold coordinated vertex reveals a polarity dependence for the neighbors of the Type-III4 vertices. Depending on the residual effective magnetic moment at the vertex the surrounding three-fold coordinated vertices, Type-I3, have a preferable magnetic charge as well. This result is purely driven by the geometric constraints imposed by the lattice itself. A positive magnetic charge at the four-fold coordinated vertex indicates that three of the mesospins point into the vertex and only one of the mesospins out of the vertex. Since the short parallel mesospins of a three-fold coordinated vertex define the charge of the Type-I3 vertex, the magnetic charge of the four-fold coordinated vertex dictates the charges of the surrounding three-fold coordinated Type-I3 vertices. For the Type-IV4 vertices this effect is even more drastic, as Type-I3 neighbors of a Type-IV4 vertex can only be opposite in charge.

**Figure 4.13.** Investigation of the two surrounding four-fold coordinated vertices dependent on the six invariant three-fold coordinated vertex states (from up to down in the following order: Type-I3(+), Type-I3(-), Type-II3(+), Type-II3(-), Type-III3(+), Type-III3(-)) for the SH (left) and the mSH (right) lattice. No differences between the SH and mSH vertex neighbors are identifiable, geometrical restrictions can be seen for Type-I3 and Type-III3 vertices and their Type-III4 neighbors.

On the other hand the Type-II3 vertex charge is only dependent on the magnetization direction of the perpendicular mesospin, which is not shared with
any of the four-fold coordinated vertices and therefore no correlation between
the charge of the Type-II$_3$ vertex and the charge of the four-fold coordinated
vertex can be seen.

These effects are of course also seen in the four-fold coordinated neighbors
of the three-fold coordinated vertices, as illustrated in figure 4.13. And once
more there are no differences between the mSH and SH lattice. But investigat-
ing the three-fold coordinated vertex neighbor of a three-fold coordinated
vertex a variation in the abundance of the different vertex neighbor states can
be identified, see figure 4.14. The long-mesospin prevents a positive charged
Type-I$_3$(+), meaning the two parallel mesospins pointing in and the perpen-
dicular mesospin (long-mesospin) points out of the vertex, to be neighboring
another positively charged Type-I$_3$(+) vertex in the SH lattice, this restriction
is not given in the mSH lattice. A Type-I$_3$(+) vertex can neighbor another
Type-I$_3$(+) vertex if the two-fold coordinated vertex in-between is in its ex-
cited state, Type-II$_2$(+). Therefore the high percentage of Type-II$_2$ states in the
mSH lattice promotes the differences in the three-fold coordinate neighbors of

![Figure 4.14](image_url)

*Figure 4.14.* Striking differences between the three-fold coordinated vertex neigh-
bor of the other three-fold coordinated vertex can be seen between the SH (left) and
mSH (right) lattice. As this two three-fold coordinated vertices are connected via the
long-mesospin (the two-fold coordinated vertex in mSH case), the effective magnetic
charge of the Type-I$_3$ and Type-II$_3$ vertex neighbor is defined in the SH lattice, due to
the shared long-mesospin. This restriction is lifted in the mSH lattice and the charge
preference almost vanishes in the mSH lattice.
the three-fold coordinated vertices. To further identify differences between the SH and the mSH lattice we need to apply other analysis tools.

Spin structure factor

As shown in figure 3.11 the SSF of the SH and mSH cannot easily be compared, because of the different elements in the base of the arrays. However, by choosing the correct subset of mesospins (as described in 3.4.3) differences between the ordering in the SH and the mSH lattice can be presented. All short-mesospins in the SH lattice and therefore all mesospins belonging to the four-fold coordinated vertices are investigated for the mSH and the SH lattice. This subset of mesospins is identical for the two lattices and a difference in the resulting SSF would identify different ordering among the mesospins of the four-fold coordinated vertices.

Figure 4.15. SSF of the mSH (left) and SH (right) lattices, calculated from the PEEM-XMCD measurements on the samples with a lattice parameter of 600 nm. The used sub-lattice of mesospins is indicated in the inset of the SSF maps. The real space lattice vectors are indicated as well, being the base for the reciprocal lattice units (r.l.u.) which are used for the $q_{x,y}$ axes.

The peak position of the SSF for both lattices is identical, see figure 4.15 and can be traced back to the SSF of the square ASI lattice (see figure 3.11), on which foundation the investigated SSF maps are based. Remarkable differences in the peak shape of the mSH and the SH lattice can be observed, revealing the different magnetic ordering among the mesospins of the four-fold coordinated vertices. An interesting question is therefore raised, on which length-scale this different ordering occurs. As the vertex distribution for fourfold coordinated vertices is similar between the two lattices the difference in the SSF has its origin in longer range ordering, extending over multiple ver-
The SSF in figure 4.15 are calculated SSF of the sub-lattice of the whole measured arrays, therefore these SSF maps are one snapshot of one possible microstate. Higher intensities can occur to specific arrangements which are unique for this certain microstate. This so called speckle information will vanish by measuring multiple microstates of the same sample and averaging them. The resulting SSF represents then a picture of the most probable magnetic ordering features common in all averaged microstates. As our statistics are large and the measured arrays extend over thousands of mesospins, we use a trick, defining the upper length-scale on which we are probing the magnetic order. The large image can be reduced to several smaller images, reducing the lateral extension of the array, but increasing the number of investigated microstates. The extension of the investigated spin-system defines the upper boundary on the ordering length-scale. As the chosen sub-lattice is a lattice out of four-fold coordinated vertices, a natural choice for the investigated spin-system size can be characterized by the number of vertices investigated. Therefore probing one vertex, meaning the magnetic ordering among the four mesospins of the four-fold coordinated vertex are investigated. Calculating the SSF for all four-fold coordinated vertices in our measured PEEM-XMCD image provides us

Figure 4.16. Averaged SSF maps for the mSH, a)-c), and the SH, d)-f), lattice for different sized real space input. The SSF for four mesospins, resembling one four-fold coordinated vertex, is similar for the mSH, a), and SH, d), lattice, as they resemble the vertex distribution on the four-fold coordinated vertices, which is similar for this two lattices. The SSF for the real space input of four (b, e) or nine (c, f) four-fold coordinated vertices, sixteen or thirty-six mesospins, represents the distinct differences we detected in figure 4.15.
with a large number of microstates which are investigated. We only probe one vertex at a time, therefore we have no information about the correlation or ordering between different vertices. The SSF of a mesospin-system of four mesospins (creating a four-fold coordinated vertex) represents the vertex distribution of the investigated array. As this vertex statistic is similar for mSH and SH lattice the averaged SSF of one four-fold coordinated vertex is similar as well. The averaged SSF for one, four and nine four-fold coordinated vertices is presented in figure 4.16.

As expected the averaged SSF for one vertex does not differ for the mSH and SH lattice, but calculating the SSF for mesospin-systems of four as well as nine four-fold vertices shows the same differences we already identified in the SSF presented in figure 4.15.

Investigating line cuts through these SSF maps the differences between mSH and SH lattice can be made clearly visible, while the lineprofile of the SSF maps for the averaged SSF of one four-fold coordinated vertex is exactly

Figure 4.17. a), A diagonal line cut through the SSF maps of the SH (blue) and mSH (red) (figure 4.15). To reduce the speckle information nine lines are averaged and a bin size of five values is used. b), Single line cuts through the averaged SSF maps from figure 4.16 for the SH (blue – left) and mSH (red – right), the number of real space input four-fold coordinated vertices is indicated on the graphs. Additionally the line cut for the not plotted SSF maps for sixteen vertices is added.
the same for the mSH and the SH lattice, see figure 4.17. Different modulations appear by including further vertices in the investigated spin-system, implying that there is different magnetic order on these length-scales.

The investigated subset of mesospins consists of mesospins belonging to the four-fold coordinated vertices, therefore we are probing the difference in the ordering among the mesospins in this special sub-lattice. Since we could identify the major differences already for a spin-system size of four four-fold coordinated vertices, we can identify the origin of this ordering. Four four-fold coordinated vertices define the edges of a plaquette, see section 2.2.3. In the middle of a plaquette is the position of the long-mesospin (SH) or the two short-mesospins (mSH). Therefore the variation of the SSF represents the influence of the long-mesospin on the ordering in the Shakti lattice.

Spin arrangement

To identify the influence of the long-mesospin on the ordering in the SH lattice in contrast to the mSH lattice, we investigate the four adjacent mesospins of the long-mesospin, as shown in figure 4.18. In the mSH lattice the long-mesospin consists out of mesospins III and IV. In the SH lattice mesospins III and IV are one long-mesospin, resulting in their magnetic alignment always being ferromagnetic. The mesospins I, II, V and VI are the first neighbors of the long-mesospin and each of them belong to a different four-fold coordinated vertex, illustrated by the red islands. While I and II are on one side of the long-mesospin (ferromagnetic two-fold coordinated vertex, Type-I$_2$, in the mSH case), mesospin V and VI are on the other side. The spin arrangements we investigate are illustrated in figure 4.19a). Among these nine spin arrangements there are three different classes to be identified. In the first spin arrangement class, there is at least one of the mesospins I or II ferromagnetically coupled to the long-mesospin and additionally to that at least one of the mesospins V or VI is also ferromagnetically coupled to the long-mesospin. For the mSH lattice the requirements for the first class are that at least one of the mesospins I or II has to be ferromagnetically coupled with mesospin III and at least one of the mesospins V or VI has to be ferromagnetically coupled with mesospin IV. But additionally to that, mesospin III need
to be ferromagnetically aligned to mesospin IV. The latter requirement can be expressed in the two-fold coordinated vertex type. If mesospins III and IV are ferromagnetically coupled they are in their lowest energy state, Type-I\textsubscript{2}, and an anti-ferromagnetic coupling represents the excited state, Type-II\textsubscript{2}.

Another way of interpreting the spin arrangements, is that the head to tail arrangement of the spins across the long-mesospin (Type-I\textsubscript{2} vertex) correlates the respective four-fold coordinated vertices to each other. The coupled four-fold coordinated vertices are indicated with light green in figure 4.19a). Spin arrangements C\textsubscript{1} to C\textsubscript{4} represent the four cases for which one four-fold coordinated vertex on one side of the long-mesospin (Type-I\textsubscript{2} vertex) is correlated to another four-fold coordinated vertex on the other side of the long-mesospin (Type-I\textsubscript{2} vertex). The next class, spin arrangements C\textsubscript{5} to C\textsubscript{8}, investigates spin arrangements which couple one four-fold coordinated vertex on one side of the long-mesospin (Type-I\textsubscript{2} vertex) to two four-fold coordinated vertices on the other side of the long-mesospin (Type-I\textsubscript{2} vertex). The final spin arrangement, C\textsubscript{9}, represents the lowest energy configuration for these five (six in the mSH case) mesospins and couples all four four-fold coordinated vertices via the long-mesospin (Type-I\textsubscript{2} vertex).

\textbf{Figure 4.19. a),} An illustration of the nine investigated spin arrangements, C\textsubscript{1}-C\textsubscript{9}. The spin arrangements represent a ferromagnetic coupling of the mesospins I, II, V and/or VI to the long-mesospin (ferromagnetic two-fold coordinated vertex in the mSH case). Four-fold coordinated vertices, to which these coupled mesospins belong to, are indicated in green in a). The spin arrangements can be classified in three classes, the coupling of one four-fold coordinated vertex with at least another four-fold coordinated vertex on the other side of the long-mesospin (ferromagnetic two-fold coordinated vertex), C\textsubscript{1}-C\textsubscript{4}, C\textsubscript{5}-C\textsubscript{8} couples one four-fold coordinated vertex to two four-fold coordinated vertices on the other side of the long-mesospin (ferromagnetic two-fold coordinated vertex). The lowest energy configuration for this set of five/six mesospins is represented by the spin arrangement C\textsubscript{9}. The fractions of these spin arrangements are shown in b) for the SH and mSH lattices, an distinct drop in the coupling via the two short-islands can be found in the mSH lattice.
Besides the reduced abundance between the three different classes in both lattices, the distinct drop in the appearance of the spin arrangements is clearly visible comparing mSH and SH lattice. The abundance is for each spin arrangement higher for the SH lattice, therefore coupling between four-fold coordinated vertices is much more favorable in the SH lattice as it is in the mSH. The origin of these disturbed couplings of four-fold coordinated vertices are the excitations in the two-fold coordinated vertices. As these mesospins are anti-ferromagnetically aligned, the chain of nearest-neighbor ferromagnetically coupled mesospins, including the two-fold coordinated vertex, is destroyed in the mSH lattice.

Regarding the ground state manifold of the Shakti lattice, the four-fold coordinated vertices are all in their lowest energy configuration, Type-I4. Two Type-I4 vertices which are coupled across a long-mesospin (Type-I2 vertex) are therefore always in their opposite degenerate vertex state. Resulting in a higher possibility of the coupling of Type-I4(A) vertices across the long-mesospin with Type-I4(B) vertices in the SH lattice. A coupling of exactly inverted mesospins can be seen as an anti-parallel alignment, which leads in scattering to a hollow peak modulation, as observed in the SSF of the SH lattice (see figure 4.15).

Bolzmann distribution

The Bolzmann distribution quantifies the thermalization of a system. In square ASI systems these investigations are carried out by fitting the vertex populations with their respective energies. The distribution of states in a system can therefore be described after Boltzmann:

$$P_x = A \cdot e^{-\frac{E_x}{k_B \cdot T_{eff}}} \quad (4.1)$$

![Figure 4.20. Illustration of the fits of equation (4.2) through the different vertex distributions for the three- and four-fold coordinated vertices for all Shakti lattices.](image)
The population of the vertex states, \( P_x \), need to be degeneracy corrected and normalized to follow the Boltzmann distribution in equation (4.1), with \( E_x \) being the energy of the vertex state. As we plot the natural logarithm of the degeneracy corrected vertex counts the Boltzmann distribution can be fitted by a linear function:

\[
fit(E) = C_1 \cdot E + C_2
\]

(4.2)

\[C_2 = \ln(A)\] is the pre-factor of the Boltzmann distribution, \( C_1 \) is the important parameter with:

\[
C_1 = -\frac{1}{k_B \cdot T_{eff}} \quad \Rightarrow \quad T_{eff} = -\frac{1}{k_B \cdot C_1}
\]

(4.3)

Where \( k_B \) is the Boltzmann constant and \( C_1 \) the fitted parameter with which we can calculate the effective temperature, \( T_{eff} \), for the investigated array. As the Shakti lattice inhabits different vertex types (coordination number three and four), we can calculate these effective temperatures for the two sub-lattices. The respective fits are shown in figure 4.20 and the resulting effective temperatures are presented in figure 4.21.

The Boltzmann fits for the different vertex types, for the different lattices and lattice parameters do not seem to differ much from each other. But a

![Figure 4.21](image)

Figure 4.21. Effective temperature for three- (triangle) and four-fold (square) coordinated vertices. Large error bars for the effective temperature at the three-fold coordinated vertices and a lattice parameter of 600 nm indicates an out of thermal equilibrium distribution for these vertices.
closer look on the actual fitted effective temperature identifies clearly certain differences (figure 4.21).

The effective temperature for a certain array and a certain lattice parameter shows no significant differences for the vertices with coordination number three and four. A slight drop in the effective temperature of both vertex types can be seen by increasing the lattice parameter. But the most striking observation is the increase of the error bars for the vertices with coordination number three for both the mSH and SH lattice. This increase can only be seen for the stronger coupled array and is not detectable in the weaker coupled arrays. One might even identify a slight enhancement of the error bars of the effective temperature of the vertices with coordination number four, but it is not as striking as in the stronger coupled case with the three-fold coordinated vertices.

Figure 4.22. Possible excitation relaxation in the mSH lattice. a), Representation of the vertex excitations in a plaquette. Two scenarios, b)-c), are exemplary presented, in which a Type-III$_4$ excitation translate to a three-fold coordinated vertex, to reduce the total energy of the plaquette.

The increased error bars are actually an indication that our array or in this case the sub-lattice of three-fold coordinated vertices does not follow the Boltzmann distribution. At the same time the four-fold coordinated vertices can be normally fitted with the Boltzmann distribution, we can explain this differences with an effective dumping of excitations on the three-fold coordinated vertices. A vertex with four mesospins, in the four-fold coordinated vertex, is stronger coupled then the vertex with three mesospins, in the three-fold coordinated vertex. Therefore the array favors the four-fold coordinated vertices to be in a thermal equilibrium, at the cost of the three-fold coordinated vertices. This effect is much stronger in the SH lattice, as the mSH lattice has the opportunity to relax excitations, which translated from the four-fold coordinated vertices into the three-fold coordinated vertices. This higher excitation at the three-fold coordinated vertex is able to relax via the two-fold coordinated vertex. Exemplary situations are graphically shown in figure 4.22.
4.3 Saint George lattice

In the Saint George lattice the same elements are used as in the Shakti lattice. The high symmetry of the Shakti lattice is destroyed to bring the long-mesospins in close proximity to each other. These lattices stem also from the square ASI, similar tools can be applied as in the Shakti analysis. The investigated Saint George lattices are produced on the same substrate as the Shakti lattices and have therefore the same island specifics, here we investigate one SG and one mSG lattice with a lattice parameter of 600 nm.

Long-mesospin chains

The arrangement of the long-mesospins in the Saint George lattice was supposed to result in ferromagnetic chain formations, as a result of the two different blocking temperatures of long- and short-mesospins. During the cooling process the interaction between the long-mesospins should have lead to these chain formations. Investigations on the correlation of the long-mesospins along one line indicates a different behavior. Surprisingly the first neighbor does not align ferromagnetically with the initial long-mesospin, its magnetic orientation is overall more anti-ferromagnetic, as one can see in figure 4.23. While the second or third neighbor is randomly oriented (correlation number of zero). The first neighbor has a small negative value, resulting from a preferred anti-ferromagnetic alignment of the first neighbor for all the investigated

![Figure 4.23. Long-mesospin correlation for the mesospins along one chain in the SG lattice (blue) and mSG (red) lattice. For the SG (left) the first neighbor is preferable anti-ferromagnetically aligned, the inset shows that the error bar does not reach the random arrangement at zero. Further neighbors are randomly arranged. For the mSG (right) as expected the first mSG-neighbor is preferably ferromagnetically aligned (> 0) while the second neighbor is preferably anti-ferromagnetically (< 0) aligned. The long-mesospin correlation (blue) of the SG lattice is plotted for comparison, at their respective neighbor position (first SG-neighbor is similar to second mSG-neighbor).](image-url)
mesospins. Investigating the same mesospins in the mSG, a different behavior was expected, since two short-mesospins represent a long-mesospin.

We identify that the first neighbor is preferable in a ferromagnetic alignment and the second, forth, sixth and eights neighbors behave like the long-mesospin neighbors, see figure 4.23. These surprising results show that the magnetic alignment of the mesospins does not differ significantly between the mSG and SG lattice, increasing the importance of the superparamagnetic background at the moment the long-mesospins freeze into their final magnetic orientation.

**Vertex statistics**

We can not identify significant differences between the SG and the mSG lattice by extracting the vertex populations. Comparing the Saint George lattices with the Shakti lattices the similarities are obvious. Small trends within the error bars can be noted. In the Shakti investigations, the mSH lattice shows a slightly higher ground state ordering among the four-fold coordinated vertices and the SH lattice is slightly higher for the coordination number three vertices. The trend in the Saint George lattice seem to show a stronger ground state ordering for the mSG lattice in both vertex types. At the same time still 33 % of the two-fold coordinated vertices are in their highest energy state, Type-II$_2$.

![Figure 4.24](image)

*Figure 4.24.* Distribution of the different vertex states for four- (a) and three-fold (b) coordinated vertices. The SG (red) and mSG (magenta) lattice distribution is shown together with the distribution of the SH (blue) and mSH (light-blue) lattice. All measured lattices have a lattice parameter of 600 nm. There are no significant differences between these four lattices, only trends within the uncertainties can be identified.
The statistics for the three- and four-fold coordinated vertices can be found in figure 4.24.

Spin structure factor

The magnetic ordering in the SG lattice is a two stage process as in the SH lattice, therefore we expect to see differences in the SSF between the mSG and SG lattice. The lattice geometry differs from the Shakti lattice, the investigated sub-lattice of mesospins is different and can therefore not directly be compared. For the SG lattice the sub-lattice consists of all short-mesospins, representing all three-fold coordinated vertices of this lattice. The same sub-lattice of mesospins is used for the SSF of the mSG lattice. Therefore all differences between the SSF maps of the SG and mSG sub-lattices identify the different ordering in this sub-systems as a result of the influence of the long-mesospin.

The SSF map of the mSG has large similarities to the SSF of the mSH (see figure 4.15), but the SSF of the SG lattice differs. Since the SG lattice has not the high symmetry of the Shakti lattice and the long-mesospins are all aligned along the vertical axis, the different peak modulation can be explained. In the SH lattice the four-fold coordinated vertex tiling had a 90° rotational symmetry, the same mechanism with the three-fold coordinated vertices in the SG lattice has only a 180° rotational symmetry. This explains the transition from a donut shape to the two elongated peaks in the SSF of the SG lattice.

![Figure 4.25. SSF of the mSG (left) and SG (right) lattice, calculated from the PEEM-XMCD measurements on the samples with a lattice parameter of 600 nm. The used sub-lattice of mesospins is indicated in the inset of the SSF maps. The real space lattice vectors are indicated as well, being the base for the reciprocal lattice units (r.l.u.) which are used for the q_{xy} axes.](image)

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Spin arrangement

To complete the analysis we investigate also the spin arrangements around the long-mesospins in the same way we did it in section 4.2. In the SG lattice the four adjacent short-mesospins belong to four different three-fold coordinated vertices, as illustrated in figure 4.26. The spin arrangements A1 to A9 investigate how many of these vertices are coupled via the long-mesospin (Type-I2).

Figure 4.26. Schematics of the investigated spin arrangement for the mSG and SG lattice. Mesospin I, II, V and VI belong each to a separate three-fold coordinated vertex, indicated by the red islands. Mesospin III and IV are one long-mesospin in the SG lattice and can therefore only be in a ferromagnetic alignment.

Figure 4.27. a), An illustration of the nine investigated spin arrangements, A1-A9. The spin arrangements represent a ferromagnetic coupling of the mesospins I, II, V and/or VI to the long-mesospin (ferromagnetic two-fold coordinated vertex in the mSG case). Three-fold coordinated vertices which belong to these coupled mesospins are indicated in green in a). The spin arrangements can be classified in three classes, the coupling of one three-fold coordinated vertex with at least another three-fold coordinated vertex on the other side of the long-mesospin (ferromagnetic two-fold coordinated vertex), A1-A4. A5-A8 couple one three-fold coordinated vertex to two three-fold coordinated vertices on the other side of the long-mesospin (ferromagnetic two-fold coordinated vertex). The lowest energy configuration for this set of five/six mesospins is represented by the spin arrangement A9. The fractions of this spin arrangements are shown in b) for the SG and mSG lattice and a distinct drop in the coupling via the two short-islands can be found for the mSG lattice.
Figure 4.28. Comparison of the spin arrangements of mesospin I and II or V and VI to each other, in the anti-parallel alignment, mesospin III was taken into account as well to create an Type-$I_3$ vertex arrangement of the investigated spins. For the parallel alignment mesospin III only defines the sign of the charge of the Type-$II_3$ vertex formation. In the SH lattice anti-parallel alignments are more favorable, while parallel are more favorable in the mSH lattice. The Saint George lattices behave different, the SG and mSG lattice show no significant differences and overall prefer the anti-parallel alignment.

The spin arrangements $A_1$ to $A_9$ behave exactly like the ones from the mSH and SH lattice, even though in the SG and mSG lattice the mesospins I, II, V and VI are not just adjacent to one long-mesospin (or two short-mesospins in the mSG case), they are actually connected to two long-mesospins at the fourfold coordinated vertex. Additionally to the spin arrangements in figure 4.27 we also investigated how mesospin I and II or V and VI are oriented to each other, see figure 4.28. In this case, while SH and mSH lattices differ in their behavior quite drastically, the mSG and SG lattices exhibit the same behavior.
5. Conclusions

Throughout this thesis we have successfully shown how to tailor the magnetic order in square ASI based magnetic mesospin systems by adding, removing, and merging magnetic elements.

The interaction modifier at the center of each vertex in the mSASI lattice recovers the degeneracy of Type-I₄ and Type-II₄ vertices and restores the highly degenerate ground state manifold in the square ASI. Additionally to the degenerated six-vertex mSASI we also achieved a Type-II₄ dominated lattice, in which the lowest energy state inhabits a residual magnetic field, leading to emergent flux-lattices on the next length scale. The influence on the magnetic ordering in these lattices strongly depends on the size of the interaction modifier and the lattice parameter. We only view the interaction modifier as the submissive part in a master–slave relation between the Ising mesospins and the interaction modifier. Therefore the gap between modifier and mesospin should define the interaction strength and therefore the magnetic ordering should be the same for similar gap sizes. Our experimental results clearly show that the gap does not define the magnetic state in these arrays. Only the interaction modifier diameter together with the lattice parameter uniquely defines the magnetic state. These results lead to the conclusion that the size of the interaction modifier is an important parameter. While the interaction modifier with \( D = 120 \text{ nm} \) is just 18 % of the size of an Ising-island the modifier with \( D = 150 \text{ nm} \) is already 28 % of the island size and the interaction modifier \( D = 200 \text{ nm} \) corresponds to 50 % of an island. Therefore the assumed slave (interaction modifier) to master (Ising mesospin) relationship is changing to a more competing interaction with increased interaction modifier diameter. Since the state of the interaction modifier is not in all cases uniquely identifiable the interaction modifier stays as an unknown variable for this mesospin system. Especially larger interaction modifiers result in an altering of the vertex state as well as the vertex state alters the magnetic state of the interaction modifier.

The Shakti lattices reveal at a first glimpse no differences between the unique (mSH) and multiple (SH) sized element systems. The disordered long-mesospins result in the same vertex statistics for the SH as well as the mSH lattice. Counter-intuitively the SH lattice shows even higher order among elements in a short-range distance, promoted by the disordered long-mesospins. Therefore the induced global disorder of the long-mesospins results in a non-altered ordering on the vertex level and promotes higher short-range order, on plaquette sizes and above, in comparison with the comparable unique sized...
mesospin system. Additionally, both systems show that removing mesospins out of a four-fold vertex reduces the coupling on this vertex point and therefore creates weaker sites in the array. These sites enable the array to relax the higher excited vertices over these weak links. With this mechanism the mesospin system ensures the thermal equilibrium on the stronger coupled vertices at the expense of the weaker coupled vertex types.

The Saint George lattice was invented to promote long-mesospin interactions and pre-defined order among the long-mesospins. However, the chosen lattice geometry promoted anti-ferromagnetic ordering between the nearest long-mesospin neighbors. Additionally the same short-range order phenomena as in the Shakti lattice could be observed. The conclusion we can draw from this lattice is that the superparamagnetic nature of the short-mesospins at the blocking temperature for the long-mesospins affects the ordering of the latter according to the lattice geometry.

These results successfully illustrate how we are able to modify the magnetic ordering in square ASI based nano-structures on different levels. Combinations of these modification approaches open up the road to new exotic ordering phenomena in highly degenerate and/or frustrated multi-scale mesospin systems.
6. Outlook

The extent of this thesis could only scratch the surface of all the possible lattice realizations for artificial magnetic mesospin systems and therefore new questions have opened up during this work.

A major follow up of the presented work is the investigation of these magnetic mesospin structures in their thermally active regime, to directly map out the different thermal response. Throughout the main part of this work frozen magnetic configurations have been investigated, but with the background provided by PAPER-II more temperature dependent measurements and analysis need to be applied to this field.

Also the thorough investigations in PAPER-III should be extended deeper into the Type-II$_4$ dominated vertex regime, especially to achieve the next vertex energy crossing (shown in figure 2.8), to achieve a mesospin array with energy relations for the different vertex states as follows:

\[ \text{Type} - \text{II}_4 < \text{Type} - \text{III}_4 < \text{Type} - I_4 < \text{Type} - IV_4 \]

One aspect of this lattice is that unlike in square ASI or the other mSASI lattices investigated in this thesis, the second vertex energy level is achieved by one simple spin flip out of the lowest energy level. Additionally, both states inhabit a residual magnetic field direction in their vertices, the lowest energy state as well as the first excitation. Emergent flux ordering and their dynamics might be a result of such an energy-landscape.

Considering extreme cases, with interaction modifier diameters of $D > 300$ nm, the interaction modifier has more magnetic material than an Ising-island. The master – slave relation between them is changed in such a way that the Ising mesospins will act as pinning centers for the large magnetic discs.

The Shakti ground state manifold[25], as described in figure 2.11, exhibit a certain tiling of the Type-I$_4$ vertices, so that 50% of the three-fold coordinated vertices are in Type-II$_3$ excitations. This tiling inhabits the high degeneracy in the Shakti lattice geometry and resembles the emergent six-vertex model[44] on a plaquette level[25]. But especially in the mSH lattice a long-range ordered ground state with all four-fold coordinated vertices being in either Type-I$_4$(A) or Type-I$_4$(B) configuration is just slightly less favorable than the Shakti ground state manifold. Engineering the gap in the two-fold coordinated vertex (shortening these mesospins) could promote a transition from a Shakti ground state manifold to a long-range ordered system.
The two energy-scales are a defining feature of the SH and SG lattices, investigations on partially or completely field-cooled samples could show the influence of a frozen-in bias in these structures. In a careful temperature series the magnetic relaxation of the short-mesospins could be seen at some elevated temperatures, while the long-mesospins are still in their biased orientation.

Of course new structures can always be designed, but also a combination of interaction modifier in Shakti or Saint George lattices is a promising pathway for future investigations. Interaction modifiers introduced in some parts of the sample and not in other parts could help to channel excitations or promote certain magnetic configurations.

All the investigations comprising this thesis are real space measurements, but as we presented especially in PAPER-III and IV reciprocal space methods like the SSF are useful tools for investigating the ordering of these systems. Therefore a logical step would be to use these two-dimensional mesospin systems and measure them, additionally to the real space investigation, in scattering experiments with neutrons or polarized X-rays[70].
Magnetiska nanostrukturer är ett utmärkt sätt att undersöka en rad olika fysikaliska fenomen både vad det gäller ordnade och oordnade system samt frustration i generell mening. I detta arbete har en mängd olika strukturer och deras underliggande fysikaliska beteenden undersökts. Speciellt fokus har legat på termiskt aktiva strukturer där temperaturen agerar som drivkraft för förändring strukturerna.

En speciell typ av magnetiska nanostrukturer har benämningen artificiell spinn-is, varvid den med kvadratisk geometri är den mest studerade och välkända. Genom en förändring i denna struktur, där vi har placerat en cirkulär magnetisk ö i mitten av varje enhet som bygger upp strukturen, har vi kunnat modifiera energin för de olika tillstånden i strukturen och därmed den övergripande magnetiska ordningen. Ett degenererat grundtillstånd, i analogi med exempelvis is (av vatten) kunde på detta sätt påvisas. Men även nya, helt okända tillstånd, med en emergent ordning observerades.

Huvuddelen av detta arbete har handlat om strukturer där elementen som bygger upp strukturen har olika aktiveringsenergier på grund av storleksskillnader, se figur 7.1.

En analys av korrelationen för de långa magnetiska elementen i den så kallade Shakti-strukturen avslöjar ett slumpmässigt magnetiskt tillstånd och

\[\text{Figure 7.1. Temperaturberoende magnetiskt ordning i magnetiska nanostrukturer.}\]

Sammanfattningsvis så visar arbetet vikten av hur olika längd- och energiskalar påverkar den magnetiska ordningen i termiskt aktiva strukturer samt hur den magnetiska ordningen kan manipuleras.
8. Summary in German


![Diagram](image)

*Figure 8.1.* Dargestellt ist das temperaturabhängiges magnetisches Verhalten im modifizierten Shakti und Shakti lattice.


In ähnlichen Gittern mit verschiedenen Element Größen konnte der selbe Effekt beobachtet werden.

Wir haben gezeigt wie wichtig die unterschiedlichen Längen- und Energieskalen für die thermischen Anordnungsprozesse in magnetischen Strukturen sind und wie magnetische Ordnung in diesen Strukturen manipuliert und verändert werden kann.
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References


[27] C. Kittel, Einführung in die Festkörperphysik (R.Oldenbourg Verlag, 1983).


[49] A. Gusman, S. Chandu, and F. Yaghmaie, ZEP520A - New resist for Electron


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