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Tuning magnetic ordering in a dipolar square-kite tessellation

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The potential application of artificial spin ice in magnetic nanodevices provides a strong drive to
investigate different lattice geometries. Here, we combine components of a recently investigated
artificial spin ratchet with components of the prototypical square lattice to form a geometrically
frustrated artificial spin ice system where Ising-type nanomagnets are arranged onto a two-
dimensional square-kite lattice. Using synchrotron-based photoemission electron microscopy, we
explore moment configurations achieved in this lattice geometry. Following thermal annealing, we
image how a variation of the relevant lattice parameter affects magnetic ordering in four-island
squares and four-island vertices during cooling through the Blocking temperature. Depending on
lattice spacing, both nearly uniform and disordered spin configurations are accessible in our sam-
pies. We show that the relative energies of the building blocks of the system, which are typically
used to classify lattice configurations, are not predictive of the low energy states adopted by the
experimental system. To understand magnetic ordering in the square-kite lattice, longer range inter-
actions must be considered. Published by AIP Publishing. https://doi.org/10.1063/1.5014041

Frustration generally involves interactions that cannot be
simultaneously minimized, leading to extensive ground state
degeneracies and the emergence of exotic states. In magne-
tism, intriguing phenomena reported in systems incorporating
frustration1–4 have traditionally been explored using macro-
scopic techniques such as neutron scattering,5 Mössbauer spec-
troscopy,6 or heat capacity and susceptibility measurements.2,4
The introduction of artificial spin systems has provided a pow-
erful means to investigate frustration microscopically using
real-space imaging techniques.7–12 One particular interest has
focused on the real-time thermodynamics of these systems.13–15
In addition, artificial spin ice lattices have shown
potential for applications in the fields of spintronics16–18 and
energy storage.19 Most recently, a frustration-by-design20
concept involving two-dimensional geometries has been proposed.
These lattices exhibit exotic emergent phenomena that do not
necessarily occur in nature as a consequence of geometrical or
topological frustration.21–28 Some of these systems show great
potential as functional materials in magnetic nanomotors or
actuators. An example is the artificial spin ratchet which can
convert energy to unidirectional dynamics in a simple lattice.27

Here, we combine the fundamental components of the
artificial spin ratchet and the extensively studied square lat-
tice. The artificial spin ratchet consists of square building
blocks and shows chiral behavior. We take these squares and
couple them with vertices, with the goal of influencing the
ordering within the squares by varying the strength of the
coupling. The resulting geometry, a square-kite lattice, is
realised through lithographically arranged single-domain
Ising-type nanomagnets occupying the centers of the lattice
sites (see Fig. 1). The nanomagnets possess a lateral dimen-
sion of 450 × 150 nm and a thickness of 2.7 nm. As these
FIG. 1. Schematic of the dipolar square-kite spin ice. The nanomagnets
occupy the site centers of a square-kite tessellation. While the nanomagnet
dimensions (length $L = 450$ nm, width $W = 150$ nm, and thickness $d = 2.7$ nm)
are kept constant, the lattice parameter $a$ is varied between 700 nm and
900 nm.

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nanomagnets are coupled via dipolar magnetic interactions, we refer to this system as the dipolar square-kite spin ice. The thus created lattice consists of two sublattices with horizontal and vertical four-nanomagnet vertices (green circles in Fig. 1). The square components are coupled via four-nanomagnet vertices by interactions \( J_1 \) and \( J_2 \). The strength of this coupling is directly tunable by lattice parameter \( a \), which we vary from 700 nm to 900 nm in this study. We demonstrate that the low-energy configurations and ordering of the system sensitively depend on parameter \( a \). Besides, our observations reveal that a whole set of low-energy building blocks is suppressed in favor of moment configurations that minimize second nearest neighbor interactions.

For this work, a 2.7 nm thick permalloy (Ni81%Fe19%) film was patterned using electron beam lithography. Three sets of square-kite lattices were created. Parameter \( a \) of the lattices was set to be 700 nm, 800 nm, and 900 nm, respectively. A 2 nm thick Al capping layer was added to avoid fast oxidation. Magnetic configurations were directly visualized using synchrotron-based photoemission electron microscopy (PEEM),\(^29\) employing x-ray magnetic circular dichroism (XMCD) at the Fe \( L_3 \) edge.\(^30\) Similar to previous work,,\(^13,23,24\) a film thickness of 2.7 nm was chosen to ensure that thermally driven moment fluctuations can be resolved with XMCD imaging (7–9 s per image) at experimentally accessible temperatures. In our particular case, the blocking temperature \( T_B \) of the nanomagnets was 280 K.

In analogy to other artificial spin ice systems,,\(^8,22–24,31\) moment configurations achieved in the dipolar square-kite lattice are characterized by its building blocks, four-island squares, and four-island vertices in this geometry. Possible square- and vertex types are listed with increasing dipolar interactions (see Fig. 1). The Type III and Type IV states are shown with their corresponding dipolar energies \( E \) and degeneracies \( g \).

![FIG. 2. Square- and vertex types of the dipolar square-kite spin ice. They are shown with their corresponding dipolar energies \( E \) and degeneracies \( g \).](image)

To experimentally measure low-energy configurations, we placed our spin ice lattices (\( a = 700, 800, \) and 900 nm) into the PEEM. There, the arrays were first heated to 330 K, and after a waiting period of 2–3 h, the sample was cooled down below the blocking point to 250 K, so that low-energy configurations could be imaged [see Figs. 3(a), 3(b), and 3(c)]. The obtained magnetic configurations are characterized by plotting the square- and vertex type populations as a function of lattice parameter \( a \) [see Figs. 3(d) and 3(e)]. To obtain good statistics, we repeated this annealing protocol 5 times. As expected, Type I vertices dominate the configurational landscape. As \( a \) changes from 700 nm to 900 nm, the population of Type I shows only a moderate decrease, while the Type IIa vertex population rises, correspondingly. All other vertex types (Type IIb, III, and IV) are almost never observed, as they are energetically unfavorable. The square type populations show a more pronounced variation, as the Type A population rapidly drops with the increasing lattice parameter, while the populations of Type C and Type D squares rise accordingly until a highly disordered moment configuration is achieved at \( a = 900 \) nm. In this disordered state, the populations of Type A, C, and D squares are almost

\[
H_{ij} = \frac{\mu_0 |\mathbf{m}|^2}{4\pi L^2} \left[ \frac{1}{|\mathbf{r}_{ij} - \mathbf{r}_{kl}|} - \frac{1}{|\mathbf{r}_{ij} - \mathbf{r}_{kl}|} - \frac{1}{|\mathbf{r}_{kl} - \mathbf{r}_{ij}|} + \frac{1}{|\mathbf{r}_{kl} - \mathbf{r}_{ij}|} \right],
\]

where \( \mathbf{r}_{ij} \) and \( \mathbf{r}_{kl} \) are the locations of the positive and negative charges on nanomagnet \( i \), respectively. \( \mu_0 = 4\pi \times 10^{-7} \) NA\(^{-2} \) is the magnetic permeability, and \( |\mathbf{m}| = M = MV/V \) is the magnetic moment of a nanomagnet, with \( M \) being the saturation magnetization of the patterned magnetic film and \( V \) the volume of the nanomagnet. The energies in Fig. 2 are calculated for a lattice with \( a = 700 \) nm and \( M = 486 \) kA/m.

Based on these calculations, we see that Type A and Type B squares are almost identical in energy, while Type I vertices have significantly lower energy compared to other possible vertex configurations, similar to artificial square ice.\(^14\) In contrast to artificial square ice, where the so-called Type II vertices are four-fold degenerate,\(^8\) the Type II vertex configurations in the dipolar square-kite lattice are split into two different types, with Type IIa being lower in energy than Type IIb. This can be easily understood by considering that Type IIa configurations minimize the stronger \( J_1 \) vertex interactions, while Type IIb states satisfy the weaker \( J_2 \) vertex interactions (see Fig. 1). The Type III and Type IV states exhibit the same degeneracies as in artificial square ice.

configuration and degeneracy \( g \) of the nanomagnet.

\[
E = \frac{\mu_0 |\mathbf{m}|^2}{4\pi L^2} \left[ \frac{1}{|\mathbf{r}_{ij} - \mathbf{r}_{kl}|} - \frac{1}{|\mathbf{r}_{ij} - \mathbf{r}_{kl}|} - \frac{1}{|\mathbf{r}_{kl} - \mathbf{r}_{ij}|} + \frac{1}{|\mathbf{r}_{kl} - \mathbf{r}_{ij}|} \right],
\]

where \( \mathbf{r}_{ij} \) and \( \mathbf{r}_{kl} \) are the locations of the positive and negative charges on nanomagnet \( i \), respectively. \( \mu_0 = 4\pi \times 10^{-7} \) NA\(^{-2} \) is the magnetic permeability, and \( |\mathbf{m}| = M = MV/V \) is the magnetic moment of a nanomagnet, with \( M \) being the saturation magnetization of the patterned magnetic film and \( V \) the volume of the nanomagnet. The energies in Fig. 2 are calculated for a lattice with \( a = 700 \) nm and \( M = 486 \) kA/m.

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equal. The Type D square is generated in the artificial spin ratchet through application of a magnetic field. Here, we observe it with no applied field, at large values of \(a\).

Interestingly, Type B squares are never observed for all lattice spacings, despite having the same energy as Type A squares (see Fig. 2). At first sight, this observation seems surprising, as both Type A/Type I and Type B/Type I configurations are possible and minimize the relevant vertex interactions \(J_1\) and \(J_2\) in Fig. 1). We investigated this curiosity by including a varying number of interactions in computer simulations of the lattice. We perform Monte Carlo simulations using the Hamiltonian given in Eq. (1). The saturation magnetization was selected to fit the experimental results. We used a temperature of \(T = 250\) K, as in the experiments. The simulated lattice consisted of 14 400 nanomagnets. The results from simulations taking into account all interactions are included as lines in the plots of Fig. 3. In this case, the experiments and simulations are consistent. However, if only nearest neighbor interactions in a nanomagnet’s own vertex or square are considered, we find different populations compared to the experiments. In this case, a significant population of Type B squares is present in the simulations [see Fig. 4(b)]. Only if second nearest neighbor interactions are included in the simulations, do we qualitatively reproduce the experimental populations, with almost no Type B squares [see Fig. 4(d)]. This implies that second nearest neighbor interactions favor the formation of Type A squares and, consequently, that the energies of the vertex and square building blocks are not sufficient to predict low energy configurations of the dipolar square-kite lattice.

To further probe the long-range behavior of the lattice, we calculated the spin correlations between islands (Fig. 5). The presented data are based on simulation. Experimental values (not shown) are in good agreement with the simulations [see also Figs. 3(d) and 3(e)] but have less statistics. Spins in the same direction \((S_1 \cdot S_2 > 0,\) where \(S_1\) and \(S_2\) are the spin vectors\) are assigned a correlation value of 1, while spins in opposite directions are assigned \(-1\).

![FIG. 3.](image1)

![FIG. 4.](image2)
(marked with a white circle), averaged over all equivalent pairs in the lattice. We clearly observe strong long-range ordering in the $a = 700$ nm system. At $a = 900$ nm, the ordering of squares is drastically reduced, while nearest neighbor islands within a vertex remain strongly correlated by the ice rule.

In summary, we introduced an artificial frustrated system whose low energy moment configurations can be tuned by a change in lattice parameter $a$. With decreasing coupling (increasing $a$), we see a transition from long-range to short-range order. We find that it is not possible to predict the low energy configurations of the system by considering only the building blocks of the lattice. A good agreement between experiments and simulations is only attained when longer range interactions are taken into account, highlighting their importance in the magnetic ordering of this and similar types of artificial spin ice systems. While the dipolar square-kite lattice offers an interesting artificial frustrated system from a fundamental aspect, as it combines features from the artificial spin ratchet with artificial square ice with an interesting capability of tuning the ground state preferences, it also offers an interesting prospect for researchers interested in exploring its dynamic response. So far, such studies focused on vertex-dominated artificial spin ice geometries, and it will be interesting to see how tunable square-type preferences in the square-kite lattice influence the dynamic properties of respective systems. Artificial spin ices can also be used as programmable metamaterials for spin waves, where the magnon band structure is sensitive to the moment configuration. The ability to tune the degree of long-range ordering in the square-kite lattice makes it a promising candidate for future explorations in magnonics and spintronics.

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