Spin-wave dynamics and symmetry breaking in an artificial spin ice

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Abstract:
Artificial spin ices are periodic arrangements of interacting nanomagnets successfully used to investigate emergent phenomena in the presence of geometric frustration. Recently, it has become clear that artificial spin ices have the potential to be used as building blocks for creating functional materials, such as magnonic crystals and ratchets, in addition to supporting a large number of programmable magnetic states. In this context, we investigate the magnetization dynamics in a system exhibiting asymmetric magnetostatic interactions owing to locally broken structural symmetry. We find a rich mode spectrum that can be tuned through an external field. We determine the evolution of the modes, starting with building blocks and evolving into larger arrays, highlighting the role of symmetry breaking in defining the mode spectrum of the system. These results contribute to the understanding of magnetization dynamics in spin ice systems beyond the kagome and square ice geometries and are particularly relevant for the realization of reconfigurable magnonic crystals based on spin ices.

Keywords: nanomagnetism, artificial spin ice, spin wave, magnetization dynamics

Over the past few years, the study of artificial spin ices has evolved from the creation of model systems designed to investigate geometric frustration with a view towards functional systems\textsuperscript{1-4}. Artificial spin ices consist of magnetostatically-coupled geometrically arranged ensembles of nanomagnets that are lithographically patterned. Each nanomagnet is in a single-domain state with the magnetization pointing in one of two orientations along the magnet long axis due to shape anisotropy\textsuperscript{5}. Spin ice geometries, such as the square and kagome ices were originally derived from crystallographic planes in rare earth pyrochlore compounds\textsuperscript{6}. Artificial spin ices with these geometries have been found to display rich collective behavior, such as phase transitions\textsuperscript{7} and well-defined mode spectra\textsuperscript{8-13} based on their large number of micromagnetic degrees of freedom\textsuperscript{14-16}. These mode spectra can be tuned by modifying the magnetic state of the system\textsuperscript{8} or through coupling with a magnetic underlayer\textsuperscript{17} and show promise for creating reconfigurable magnonic crystals\textsuperscript{4,13,18-20}. 
Meanwhile, a number of promising geometries have been proposed, beyond the square and kagome lattices. Some enable emergent frustration, rather than geometric frustration at the vertex level\textsuperscript{21}, leading to control over the dynamics of emergent charges\textsuperscript{22,23}, while other geometries have allowed the creation of arrays whose magnetic state is fully reconfigurable\textsuperscript{24}. More recently, chiral geometries have been investigated and have been shown to display e.g. ratchet behavior during thermal relaxation\textsuperscript{25} and during field-induced magnetization reversal\textsuperscript{26} as well as tunable vertex frustration\textsuperscript{27}, and tunable ferromagnetic and antiferromagnetic order\textsuperscript{28-31}.

Here, we investigate the resonant dynamics in arrays built upon a chiral pattern. The geometry, shown in Figure 1a, gives rise to asymmetric magnetostatic interactions and tunable frustration\textsuperscript{27}. While asymmetric geometries, such as chiral structures, have demonstrated rich collective behavior during thermal relaxation\textsuperscript{25} and field-induced dynamics\textsuperscript{26,28}, their GHz dynamics have not been studied so far. The geometry of the investigated structures leads to noteworthy characteristics. Firstly, the unit cell is composed of interacting horizontal and vertical nanomagnets, which leads to the asymmetric magnetostatic interactions. In comparison, square and kagome ices possess unit cells with multiple symmetry axes. Secondly, the interactions between the horizontal and vertical nanomagnets is mediated by surface charges, \(\sigma \propto \mathbf{m} \cdot \mathbf{n}\), where \(\mathbf{m}\) is the magnetization vector and \(\mathbf{n}\) a vector normal to the surface. This is in contrast to the interactions in square and kagome lattices, which are mostly due to volume charges, \(\rho \propto \nabla \cdot \mathbf{m}\), located at the extremities of the nanomagnets.

Using time-resolved magneto optical Kerr effect (MOKE), we determine that the magnetization dynamics is characterized by a rich mode spectrum, which can be tuned by an external magnetic field. Finding a complex mode distribution in large arrays, we determine the evolution of the mode spectrum with the system size and geometry, starting with building blocks. Building blocks with broken structural inversion symmetry display a large number of modes. Interestingly, while arrays larger than a unit cell exhibit mostly identical mode spectra, irrespective of their geometry, we find specific modes, whose intensity, and existence, is correlated to the size of the array. The amplitude evolution of these modes reflects the long-range nature of the interactions in the studied system. Our results unveil different
mechanisms for mode evolution, which are useful for designing artificial spin ices for applications such as magnonic crystals.

The magnetization dynamics were measured by using an all-optical time-resolved magneto-optical Kerr effect microscope based on two color collinear pump probe technique, shown in Figure 1b. The Kerr rotation of the probe laser beam (λ = 1030 nm and pulse width of ca. 50 fs) is measured after exciting the sample by a pump beam (λ = 515 nm and pulse width of ca. 50 fs). The pump and probe beams are collinear. The approximate regions covered by the pump and probe beams are schematically indicated in Figure 1a. The pump pulse modifies the local magnetization through thermal effects, thereby inducing precessional magnetization dynamics. A Fourier transform of the time resolved data is performed to obtain the mode spectra of the sample at different values of an external magnetic field applied at 45° in the sample plane (as indicated in Fig. 1a). The measured spin-wave (SW) spectra in the presence of different applied fields are shown in Fig. 2a, where distinct modifications are visible. At 100 mT, three main SW modes are present (around 8.2, 11.4 and 14.1 GHz) and, as the field is reduced, changes in the mode intensity and shifts in peak positions occur. In particular, the frequency of the mode at 8.2 GHz (labeled M₁) is red shifted, while the amplitude of the 11.4 GHz mode at 100 mT (labeled M₂) gradually decreases as the applied field is reduced. We have used micromagnetic simulations in order to elucidate the evolution of the experimentally measured modes. For comparison, in Fig. 2b the simulated FFT is performed considering a region of the array equivalent to the probe laser spot. Overall, the general distribution of the simulated modes agrees with the experimentally measured ones. The relative discrepancies in intensities and the precise positions of the peaks in the frequency domain are not always reproduced owing to the presence of a variety of effects, e.g. edge roughness, and the possible difference in the saturation magnetization and gyromagnetic ratio values in the experimental samples. We provide a simulation of the evolution of the modes as a function of the magnetic field in Fig. 2c, demonstrating how the mode spectra (frequency and amplitude) are modified. The field-dependent spectra are the result of changes in the magnetic structure: at 100 mT, the nanomagnets are almost uniformly magnetized along the applied field direction, while at 30 mT, the magnetization is mostly aligned along the long axis of the nanomagnets, displaying a curling of the magnetization at the element edges. Further insight into the nature of the modes can be obtained by
considering their spatial distribution. In Figure S1 of the Supplementary Material, we plot the simulated mode distributions for a 5x5 array at 100 mT, which exhibit complex spatial profiles, especially above ca. 8 GHz. Owing to their complexity, these modes cannot easily be analyzed. Nevertheless, we can obtain an understanding of their origin by considering building blocks of the array.

The spin-wave dynamics of three parallel nanomagnets, a 2x1 asymmetric structure and the 2x2 magnetic unit cell are simulated to determine the dependence of the mode spectra on the geometry (Figure 3a). While the spectra display similar features, they become more complex and display an increasing number of modes as the number of nanomagnets increases and their geometry is modified. The modes below 8 GHz correspond to edge modes of the individual nanomagnets, while the modes above 12 GHz correspond to quantized modes. Because both these types of modes are rather weak and, in practice, difficult to measure (except with micro-focused Brillouin Light Scattering\textsuperscript{33}, depending on the required spatial resolution), we concentrate here on the modes in the range 8 GHz – 12 GHz, and in particular on the few modes plotted in Fig. 3b, whose evolution we trace based on the geometric configuration. In the three-magnet structure, the mode at 9 GHz (highlighted by a red dot) corresponds to higher-order edge modes of the outermost elements. Interestingly, similar modes have the same frequency in the 2x1-structure configuration as well as in the 2x2 structure that corresponds to the same edge oscillations of outermost elements. The spatial distribution of these modes reflects, in part, the symmetry of the systems. The three-magnet configuration displays inversion symmetry\textsuperscript{34} about the central element (labeled $X_2$), and so does the mode distribution. The asymmetry of the 2x1 structure leads to a different mode distribution, in which the edges only oscillate in elements labeled $X_1^1$ and $Y_1^3$. While the global inversion symmetry is restored in the 2x2 structure, the mode distribution is overall symmetric, but only elements $Y_{2x1}^1$ and $Y_{2x6}^2$ display pronounced edge oscillations. The lack of oscillations in the geometrically equivalent elements $X_{2x3}^2$ and $X_{2x2}^2$ is a result of the applied field $\mu_0 H = 100$ mT, which leads to further symmetry breaking. In contrast, the mode at 9.92 GHz (highlighted by blue rectangles) is slightly blue-shifted and decreases in amplitude as the geometry of the structure becomes more complex. In the three-magnet state, it corresponds to higher-order modes in the outermost nanomagnets that retain inversion symmetry, while the mode distribution in the central nanomagnet is mostly bulk-like. In the
2x1 structure, a mode with similar features occurs at 9.96 GHz (purple square, where the mode distribution in elements $Y^T_3$ and $X^T_1$ are similar to that in $Y_3$ and $X_1$, while element $Y^T_2$ exhibits a mode distribution very similar to that found in $Y_2$. The other elements display fundamentally different oscillations. Surprisingly, similar modes are found at 10.12 GHz in central elements within the 2x2 structure: the magnetization oscillations in elements $X^{2x2}_1$ and $X^{2x2}_6$ are the same as in $X_1$ and $X_3$ (both outermost elements), respectively. The (outer) elements $X^{2x2}_3$ and $X^{2x2}_4$ equally display similar modes. Moreover, the mode distribution in element $X^T_3$ (three-magnet configuration) at 9.96 GHz is also found in at 9.96 GHz in the 2x2 structure, elements $X^{2x2}_3$ and $X^{2x2}_4$. Features of the mode at 9.96 GHz in the three-magnet geometry are thus split over multiple modes in the 2x2 structure. Finally, we consider the mode at 10.08 GHz in the 2x1-structure (highlighted by the green triangle), which has no equivalent in the three-magnet state. Features of that mode are present in the 2x2 structure. In particular, the mode distribution in $X^{2x2}_1$ and $X^{2x2}_6$ reflects that of $X^T_1$ and $Y^T_3$, respectively, while the oscillations in $Y^{2x2}_4$ have the same features as in $Y^T_1$. The oscillations of the magnetization in $X^{2x2}_3$ and $X^{2x2}_4$ also are similar to those in $X^T_1$ and $Y^T_3$ (in terms of the number of nodes and symmetry).

The observed spatial mode distributions are governed by the demagnetizing field associated with the element ensembles, which are plotted in Fig. 4. The demagnetizing field in element $Y_3$ of the three-magnet structure (Fig. 4a) and element $Y^T_3$ in the 2x1 structure (Fig. 4b) have very similar demagnetizing fields, leading to the similar mode profiles described in Fig. 3b. The demagnetizing field in the other elements of the 2x1 structure is, however, modified by the magnetostatic interactions between the elements, in particular in elements $X^T_{1,2,3}$ and $Y^T_{1,2}$, with the most substantial modifications occurring in $Y^T_1$, which strongly interacts with all three horizontal nanomagnets. These interactions break the inversion symmetry observed in the demagnetizing fields of the elements within the three-magnet structure (Fig. 4a). In addition, while the demagnetization field within $Y^T_2$ is mostly similar to that in $Y_2$, a slight asymmetry is present, which explains why the mode distribution in $Y^T_2$ at 9.0 GHz and at 9.96 GHz slightly differs from that in $Y_2$. In the 2x2 structure, the large number of magnetostatic interactions leads to a different distribution of the demagnetizing field (Fig. 4c). In particular, the demagnetizing field profiles in $X^T_3$ and $X^{2x2}_3$ are similar, resulting in similar mode profiles at 9.96 GHz (Fig. 3b-c).
Having investigated the modes of configurations up to the 2x2 structure, we now consider different sizes of nanomagnet arrays. In Figure 5, mode spectra are plotted for configurations ranging from a 2x2 structure to a 5x5 array of three-magnet groups. The eigenmodes are similar (see Supplementary Material, Fig. S2), independent of the array size and edge configuration, indicating that the dominant modes are contained within the 2x2 cell, which is the magnetic unit cell. However, a number of modes display an amplitude dependence on array size. In particular, two peaks (Fig. 5a: at 10.88 GHz and 11.04 GHz) have a distinct evolution as a function of the array size. The amplitude of the mode at 10.88 GHz (Fig. 5b) decreases with increasing array size. Plotting the mode profile in Fig. 5c demonstrates that the mode amplitude is mostly localized within edge elements. Fitting the data in Fig. 5b indicates that the mode amplitude decay follows an inverse square law, indicating the presence of significant long-range interactions in the system, in agreement with the observation in Ref. 27, where long-range ordered ground-state configurations have been observed. This is in contrast to the square ice, where mode amplitudes display a linear evolution, due to the predominance of nearest-neighbor interactions. The amplitude of the mode at 11.04 GHz increases with increasing array size because the mode profile displays significant amplitude within the bulk of the array (Fig. 5d). Fitting the data indicates a square root-like behavior, starting to level off beyond the 5x5 array.

Conclusion:
We have determined the magnetization dynamics in a spin ice geometry with asymmetric magnetostatic interactions, which give rise to a rich mode spectrum. Using time-resolved magneto optical Kerr microscopy, we find that an external magnetic field can significantly modify the spin-wave spectrum. In addition, micromagnetic simulations show that the spatial distribution of the resulting modes strongly depends on the local symmetry and distribution of the demagnetizing field. The structural asymmetry of the system, in which groups of vertical nanomagnets interact with horizontal ones, leads to a proliferation of modes with complex spatial distributions. While most modes are present in the 2x2 unit cell of the array, we have found that their amplitude varies as a function of the array size: depending on whether they are array ‘edge’ or ‘bulk’ modes, their amplitude will respectively decrease or increase, and thus indicate the presence of long-range interactions within the system.
Interestingly, a distinct mode appears around 11 GHz, whose amplitude increases with array size, providing a means of determining the array size based on the mode spectrum. These results indicate how symmetry breaking can be exploited to tune the resonant spectrum of artificial spin ices. This spectral tunability opens the way to the use of artificial spin ices as reconfigurable magnonic crystals.

Figures:

Fig. 1 (a) Scanning electron microscopy (SEM) image of the spin ice lattice made of Permalloy, with the geometry of the applied magnetic field. The edge-to-edge separation between individual nanomagnets is labeled $a$ and the separation between groups of three nanomagnets is labeled $b$. In this case, $a = 50$ nm and $b = 50$ nm. The approximate excitation region of the pump laser beam is indicated by a green ellipse, whereas the red ellipse represents the average probed region. (b) Schematic of the time resolved Kerr microscope.
setup. A linearly polarized probe beam is used to measure the magnetization dynamics after exciting the sample using a pump beam. Both pump and probe beam are focused on the sample using a Microscope objective (MO). A magnetic field (H) is applied at $45^\circ$ to the sample plane. The precessional dynamics is measured using a balanced photodetector with two photodiodes represented by A and B.

Fig. 2. Spectra of the (a) experimental and (b) simulated magnetization modes considering two nanoseconds relaxation time and a limited excited region, as sketched in Fig. 1a, for three different magnetic field strengths: $\mu_0 H_{\text{ext}}=30$ mT, 50 mT and 100 mT. While individual modes cannot be cross-correlated, the general distribution of the simulated modes agrees with the experimentally measured ones. (c) Simulated external field dependence of the spin-wave spectra.

Fig. 3: (a) Simulated spinwave spectra of three parallel nanomagnets, a 2x1 configuration and of a 2x2 magnetic unit cell with $\mu_0 H_{\text{ext}}=100$ mT applied along the direction indicated by the blue arrow. The spatial profile of the labeled modes are plotted in (b), (c) and (d) for different building block geometries.
Fig. 4 Simulated demagnetization fields for (a) three horizontal and three vertical magnet structures, (b) a 2x1 structure and (c) and 2x2 structure with $\mu_0 H_{ext}=100$ mT. The demagnetizing field is plotted along the arrow directions indicated in (a). The three magnet structure clearly displays inversion symmetry, which is broken in the T structure.

Fig. 5 (a) Mode spectra, highlighting the modes at 10.88 GHz and 11.04 GHz (indicated by the grey lines), for different square array sizes ranging between 2x2 and 5x5 groups of vertical and horizontal three-element structures. (b) Evolution of the mode amplitudes as a function of array size, with fits. (c), (d) Spatial distribution of the mode amplitudes: the power is mostly concentrated in elements at the array edges for the mode at 10.88 GHz, while it is distributed across elements within the array at 11.04 GHz.

**Methods**

**Sample preparation**

Finite arrays of Permalloy (Ni$_{83}$Fe$_{17}$) nanomagnets were prepared on a silicon (100) substrate using electron beam lithography in conjunction with thermal evaporation at room temperature and a base pressure of $2 \times 10^{-7}$ mbar followed by lift-off. The evaporation resulted in a nanocrystalline Permalloy film, which was capped by a 3 nm aluminium layer to protect against oxidation. The nanomagnets are 450 nm long, 150 nm wide and 20 nm thick.
The edge-to-edge separation between the individual nanomagnets is 50 nm whereas the edge-to-edge separation between two trident building blocks are 50 nm.

**MOKE measurements**

The magnetization dynamics was measured using a two colour optical pump-probe technique. The second harmonic ($\lambda = 515$ nm, pulse width $\approx 50$ fs) of a Fiber pulsed laser of wavelength $\lambda = 1030$ nm and pulse width $\approx 50$ fs is used to excite the sample. The fundamental laser beam ($\lambda = 1030$ nm) is used to probe the dynamics after passing through a variable time delay by measuring the polar Kerr rotation using a balanced photo diode detector. Both the pump and probe beams are made collinear and are focused by using a microscope objective of Numerical aperture N.A. = 0.65. The pump beam is slightly more defocused than the probe beam, as shown in Fig 1, which makes it easier to overlap both the pump and probe beam on the sample surface. The pump beam diameter is of ca. 3 $\mu$m. The applied magnetic field is tilted slightly out of the plane of the sample to have a finite demagnetizing field along the direction of the pump pulse. The precessional dynamics consists of an oscillatory signal on top of the exponentially decaying time resolved Kerr rotation. A fast Fourier transform (FFT) is performed after subtracting the bi-exponential background to find out the corresponding power spectra. The measurement time window used in this experiment is 2 ns.

**Micromagnetic simulations**

Micromagnetic simulations based on the Landau-Lifshitz-Gilbert (LLG) equation were performed using MuMax$^{37}$ and TetraMag$^{38}$. The parameters used for the simulations were damping ($\alpha$) = 0.006$^{39}$, exchange constant, $A = 1.3 \times 10^{-11}$ J/m, magnetocrystalline anisotropy constant $K = 0$, and saturation polarization $\mu_0M_s = 1$ T. The sample geometries and sizes used in the simulations are the same as in the experiment, as determined from SEM images. The external field is applied according to the experimental configuration and a square pulsed field of width of 20 ps and peak amplitude of 2 mT is applied at 45°, in the sample plane, as indicated in Fig. 1a, to excite the sample. For the simulated data shown in Fig. 2b, the field is applied over a region comparable to that excited by the lasering the experiments (see Fig. 1a) and the magnetization dynamics integrated for 2 ns using MuMax. The the simulated data shown in Fig. 2c, the dynamics is integrated for 10 ns. In all other figures, the magnetization was integrated for 50 ns using TetraMag. The mode spectra and the spatial profiles of the modes are obtained by calculating the Fourier transform for each cell. In MuMax, the samples
are discretized into prism-like cells with dimensions of $2 \times 2 \times 20$ nm$^3$. In TetraMag, the samples are discretized in tetrahedral elements with a side length of ca. 5 nm.

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Supplementary information

Spin-wave dynamics and symmetry breaking in an artificial spin ice

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Fig. S1: (a) Simulated mode spectrum of a 5x5 array at 100 mT with a damping of 0.006 and a 2ns relaxation time. For clarity, the modes are calculated considering a global excitation of the array. (b) The spatial profile of prominent spin-wave modes.
Fig. S2: Mode spectra for square arrays of different sizes, ranging from 2x2 to 5x5, considering a 40 nanosecond relaxation time. While a number of modes display a variable amplitude as a function of array size, only the modes in the highlighted region (grey frame) display a clear evolution while, at the same time, forming isolated peaks that are experimentally measurable. Other peaks display smaller changes in amplitude (for example at 9 GHz) and are generally nested within larger peaks (for example around 10 GHz).