Supplementary Information for:

Controlled Magnetic Reversal in Permalloy Films Patterned into Artificial Quasicrystals

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We have patterned novel permalloy thin films with quasicrystalline Penrose P2 tilings and measured their DC magnetization and FMR absorption. Reproducible anomalies in the hysteretic, low-field data signal a series of abrupt transitions between ordered magnetization textures, culminating in a smooth evolution into a saturated state. Micromagnetic simulations compare well to experimental DC hysteresis loops and FMR spectra, and indicate systematic control of magnetic reversal and domain wall motion can be achieved via tiling design, offering a new paradigm of magnonic quasicrystals.

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I. Experimental Methods

A. Thin Film Patterning

1. Pattern Creation: Deflation Method

Penrose tilings can be constructed by a deflation method [1], implemented with a modified Mathematica (Wolfram Research) code. The tiling is based upon two building blocks: kites and darts. The kite is a quadrilateral with four interior angles of 72°, 72°, 72°, and 144° degrees; the dart is a quadrilateral with four internal angles of 36°, 72°, 36°, and 216° degrees (see Fig. SI-1a, below). The ratio \(d_1/d_2\) of the long to the short edges of kites and darts is the “golden ratio” = 1.618…. (irrational number). Bisection of a kite along its symmetry axis creates a pair of acute (type +1) Robinson triangles having interior angles 36°, 72° and 72° degrees. Alternatively, bisection of a dart along its symmetry axis yields a pair of isosceles (type -1) Robinson triangles having interior angles 36°, 36° and 108° degrees. Robinson triangles have the important property that they can be dissected into smaller triangles, one of each type: A kite can be dissected into two kites and one dart; whereas a dart can be sliced into a kite and two half-darts, as shown in Fig. SI-1a. Fiducial green and red circular arcs positioned on the kites and darts facilitate the application of the matching rule: The circular arcs of each color must smoothly connect when two kites or darts share an edge, as shown in Figs. SI-1b and SI-1c.

Higher-generation tilings can be constructed by repeated dissection of the kites and darts. The starting point is a tiling of five kites joined at a common vertex, which is denoted a Penrose sun, or 0th generation tiling (Fig. SI-1b). Kites joined in such a manner follow the matching rule. Next, each kite is dissected into two smaller kites and a dart, and rejoined according to the matching rule to obtain the 1st generation tiling. One can rescale the 1st generation tiling to make it the same size as the 0th generation tiling; the kites and darts of the 1st generation will then be smaller in size, compared to those of the 0th generation. Continued dissection and rescaling yields higher-generation Penrose P2 tilings (P2T), and the deflation process can be terminated at some chosen, finite generation (Fig. SI-1c).

The next step is to convert the final P2T tiling into an Autocad (Autodesk, Inc.) file format that can be imported into commercial electron beam lithography (EBL) software. Raith EBL software (Raith U.S.A.) enables one to rescale the entire P2T tiling in order to obtain the desired lengths of the long and short edges of the kites and darts. Note that the deflation process maintains the golden ratio of lengths of the long and short segment edges; but one can independently assign a desired segment width \(W\) (of the edges of kites and darts) to the P2T (Fig. SI-1d). The final, finite P2T exhibits five-fold rotational symmetry about the center point of the 0th generation Penrose sun, contrasting the ten-fold symmetry extrapolated for a theoretical, infinite P2T; however, there is no periodic translational symmetry for either finite or infinite P2T. A quasicrystal tiling can continuously fill all space, but the lack of periodic translational symmetry implies that a shifted copy of the tiling will never match with its original.
Fig. SI-1: Deflation Method for Penrose P2 Tiling: Dissection and Matching Rules

(a) **Dissection Rule**: Each kite converts into two kites and one dart, and each dart transforms into one kite and two half-darts. **Matching Rule**: The fiducial green and red arcs must separately match at edges.

(b) Schematic of 0th generation P2T, with matching green and red arcs. Images in (a), (b) and (c) are after Glassner [2].

(c) Schematic of 3rd generation P2T, with matching green and red arcs.

(d) Digital map of 3rd generation P2T with short and long edges of 500 and 810 nm, resp., and $W = 100$ nm. Black denotes permalloy film, and white denotes empty regions (approximating non-magnetic substrate). Note definition of the x- and y-axes used herein.
2. Electron Beam Lithography and Electron Beam Deposition

Sample films were patterned using a Raith E-Line EBL System (Raith U.S.A.). A 100-nm-thick layer of ZEP520A positive resist (ZEON Chemicals) was spun on a Si substrate. Key process steps are shown in Fig. SI-2. After electron beam exposure, the resist mask was developed in xylene and isopropanol.

A permalloy film of thickness $t = 25$ nm was then deposited on the masked Si substrate using electron beam evaporation (18 inch working distance), followed by lift-off in N-methyl pyrilidone solution and a rinse in isopropanol.

An SEM image of Sample III134E is shown in Fig. SI-3a; note this finite-area, 3rd generation tiling has five-fold rotational symmetry about the central “Penrose star”. An SEM image of 8th generation Sample III130C is shown in Fig. SI-3c. Note that the apical width (76 microns) of III130C is much greater than that of III134E (6.8 microns).
Fig. SI-3: SEM Images of 3rd, 5th and 8th Generation P2T

(a) SEM image of 3rd generation P2T Sample III134E with apical width $D = 6.8$ microns (blue double-arrow). Bright regions correspond to permalloy, and dark regions to Si substrate. Segment lengths are $d_1 = 810$ nm and $d_2 = 500$ nm, and width is $W = 85$ nm.

(b) SEM image of 5th generation P2T Sample III134A with apical width $D = 18.3$ microns (white double-arrow). Bright regions are permalloy, and dark regions are Si substrate. Note segment dimensions $d_1 = 810$ nm, $d_2 = 500$ nm, and width $W = 60$ nm.

(c) SEM image of an 8th generation P2T Sample III130C (one write field) with permalloy thickness $t = 25$ nm, and apical width $D = 76$ microns (double-arrow). Applied DC field $H$ makes angle $\phi$ with respect to the x-direction (white arrow is $\phi \equiv 0^\circ$) along a decahedral base.

(d) Blown-up SEM image of the P2T of (c), with color-coded lines showing the long and short segment lengths $d_1 = 810$ nm and $d_2 = 500$ nm. The segment width $W = 90$ nm.
### Table SI-1: Parameters of Sample P2 Tilings

<table>
<thead>
<tr>
<th>Sample</th>
<th>Generation</th>
<th>$0^{th}$ Gen $d_1$, $d_2$ (microns)</th>
<th>Final $d_1$, $d_2$ (nm)</th>
<th>Final W (nm)</th>
<th>Decagon Apical Width $D$ (microns)</th>
<th>No. Decagons in Write Field $N$</th>
<th>No. Write Field Copies $P$</th>
<th>Decagon Spacing $d$ (microns)</th>
<th>Write Field Spacing $d_W$ (microns)</th>
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<td>8</td>
<td>37.7, 23.5</td>
<td>810, 500</td>
<td>135</td>
<td>76</td>
<td>1</td>
<td>400</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>III130C SQUID</td>
<td>8</td>
<td>37.7, 23.5</td>
<td>810, 500</td>
<td>90</td>
<td>76</td>
<td>1</td>
<td>400</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>III133C SQUID</td>
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<td>200.5, 123.7</td>
<td>1618, 1000</td>
<td>107</td>
<td>402</td>
<td>1</td>
<td>25</td>
<td>420</td>
<td>420</td>
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<tr>
<td>III133E NBFMR SQUID</td>
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<td>34.3, 21.2</td>
<td>8100, 5000</td>
<td>1100</td>
<td>69</td>
<td>1</td>
<td>400</td>
<td>100</td>
<td>100</td>
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<tr>
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<td>9.15, 5.66</td>
<td>810, 500</td>
<td>60</td>
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<td>4</td>
<td>625</td>
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<td>100</td>
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<td>810, 500</td>
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<td>6.8</td>
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<td>100</td>
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<tr>
<td>Simulate</td>
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<td>810, 500</td>
<td>100</td>
<td>6.8</td>
<td>NA</td>
<td>NA</td>
<td>NA</td>
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</tr>
</tbody>
</table>

### B. Special Patterning Procedures for Magnetic Measurements

Sample patterns were initialized with long and short kite edges $d_1$ and $d_2$, respectively, which were reduced during deflation to a final generation, while maintaining the “golden ratio” $d_1/d_2 = 1.618$ for P2T. There are $N$ decagons of apical width $D$ and spacing $d$ (Fig. SI-3) in a write field that was copied $P$ times onto a square array of spacing $d_W$ to yield a total pattern area ($\approx 4$ to $6$ mm$^2$) containing $N \times P$ decagons suitable for magnetometer and FMR measurements. All pattern parameters are summarized in Table SI-1, and illustrated in an SEM image of a Penrose P2 tiling (Sample III134E) with multiple write fields shown in Fig. SI-4.

### C. DC Magnetometry

Static magnetization measurements were performed using a Quantum Design MPMS5 SQUID Magnetometer (Quantum Design, U.S.A.) with a field range of -50 kOe $< H < +50$ kOe.
Fig. SI-4: SEM of Write Fields Comprising P3T Sample III134E

SEM image of EBL write fields of 3rd generation P2T, Sample III134E, based upon square arrays of decagonal P2T with spacing $d = 13.21$ microns. Note the 20 micron scale bar at lower left. Each EBL write field has 49 P2T decagons. The write fields are arrayed on a square lattice of spacing $d_w = 100$ microns. Permalloy film is gray, and dark color is the Si substrate.
D. NB FMR Setup

Narrow-band (NB) FMR experiments were performed at fixed $f = 9.6$ GHz and fields up to 10 kOe using a Bruker EMX EPR Spectrometer. The Bruker EMX Spectrometer uses a microwave cavity tuned to resonate at a fixed frequency of interest in order to amplify the weak FMR signal that originates in the sample film under study. Figure SI-5a shows a block diagram of our NB FMR system. Our spectrometer operates in the reflection mode; thus, it measures the amount of power coming out of a resonant cavity containing a sample. A DC magnetic field $H$ directed along the $x$-axis is supplied by an electromagnet shown in Fig. SI-5a. Figure SI-5b shows the main components of the microwave bridge (MB), which include a microwave source (MS), attenuator, reference arm, and diode detector. The MS operates at a single frequency of 10 GHz. Since the power level of the MS cannot be changed, an attenuator is located in front of the MS, allowing control of the amount of power entering the cavity resonator (sample space). Since the spectrometer operates in the reflection mode, one uses a circulator to direct incident power into the cavity (sample), and reflected power toward a diode detector, so that diode detector does not see incident radiation from the MS. The diode detector then converts reflected microwave power from the cavity (sample) into electrical current, which is then further post-processed by Bruker proprietary electronics and software to yield the microwave power absorption versus DC field data exported to a computer. Our resonant cavity operates in the TE102 mode, with the DC magnetic field $H$ applied along horizontal $x$-axis (Fig. SI-5a); the microwave AC field is applied along the vertical $z$-axis. The sample film lies in the $xy$-plane and can be rotated about the $z$-axis (Fig. SI-5c). The sample absorbs microwave power at a particular DC magnetic field corresponding to a sample FMR mode; consequently, the cavity Q-value (energy stored in the cavity) goes down. This means the diode detector registers less microwave power at the applied field value for the mode.

E. BB FMR Setup; Angular Studies

Broad-band (BB) FMR measurements were performed at room temperature using a meander line method described elsewhere [3]; a schematic layout is shown in Fig. SI-6. The microwave frequency $f$ could be varied continuously from 10 MHz up to 20 GHz, and the DC applied magnetic field $H$ could be swept continuously from +8 kOe to -8 kOe. Note that in all our NB and BB FMR experiments, the magnetic field direction could be effectively rotated within the film plane, as defined by the angle $\phi$ between $H$ and the horizontal $x$-axis of the ADL (see Fig. SI-5c).
Fig. SI-5: NB FMR Set Up; Sample Rotation Geometry (NB and BB FMR)

(a) (upper left) Schematic diagram of Bruker EPR setup. Sample was placed in xy-plane, DC magnetic field $H_{DC}$ was along horizontal x-axis, whereas the microwave magnetic field was along negative z-axis, and the y-axis is into the page, as shown in (c) and Fig. SI-6.

(b) (above) Schematic diagram of microwave bridge exhibiting its main components.

(c) (left) Schematic of rotation of a sample film in an external DC applied magnetic field $H_{DC}$, which remains in-plane. The microwave field $h_{ac}$ is directed perpendicular to the film plane. These (nonstandard) definitions are equivalent to a rotation of the DC field away from the x-axis (sample reference edge) by angle $\phi$ (Figs. SI-3). See Figs. SI-15 and SI-16 for representative data.
Fig. SI-6: BB FMR Diagram

(a) Schematic of the BB FMR measurement cell, which consists of a meander line (0.1 mm diameter, varnish-insulated Cu wire) sandwiched between a Cu ground plane and a sample with the film side facing the meander line, which has around 20 turns covering a 5 mm x 5 mm area. The sample and meander line are enclosed within a graphite cup to reduce coupling to the environment. (b) Block diagram of measurement set-up for BB FMR. A microwave signal was generated by a *HP Model 83629A Synthesizer* with a frequency range of 10 MHz to 20 GHz, and a maximum power output of 25 dBm, and fed through Cu coax to the sample cell positioned in an electromagnet. The output of the meander line is amplified and fed into a diode detector, whose rectified output has a large DC offset that is compensated and fed into a DC amplifier for readout. After [3].
II.  Simulation Methods

A. Static Magnetic Simulations

Simulations were carried out using the Object Oriented Micromagnetic Framework (OOMMF) Code [1]. The permalloy parameters used in simulations were: exchange constant $A = 1.3 \times 10^{-11}$ J/m, saturation magnetization $M_S = 8.6 \times 10^5$ A/m, magnetocrystalline anisotropy constant $K = 0$, gyromagnetic ratio $\gamma = 1.9 \times 10^9$ Hz/T, and damping coefficient $\alpha = 0.01$. First, a bitmap image file of a permalloy film lying in the x-y plane was discretized on a $10 \times 10 \times 25$ nm mesh (note the exchange length $L_E \approx 6$ nm $\approx$ DW width for permalloy [4]), and imported into a code simulating the equilibrium magnetization in an applied DC field $H = Hx$ at temperature $T = 0$ K. Initially, the individual grid moments are randomly oriented and then allowed to relax in an ambient field of 12 kOe ($-12$ kOe); then $H$ is decreased (increased) in steps of 10 Oe until it reaches $-12$ kOe ($+12$ kOe), completing the hysteresis loop. The resulting data are reported in: 1) a DC magnetization map (see Fig. SI-9) whose color scale indicates the magnetization direction of each pixel on the $10 \times 10 \times 25$ nm mesh, with small arrows that indicate the direction of the local magnetization (averaged over approximately 100 pixels); 2) a magnetization curve (see Fig. SI-9c) obtained by summing the local magnetization (projected onto the film plane) over the entire P2T at each DC field value.

B. Dynamic Magnetic Simulations

Once the equilibrium magnetization configuration was determined for a given applied DC field, the dynamic magnetic response was simulated. First, a spatially uniform gaussian pulse of amplitude 200 Oe and full width at half maximum (FWHM) of 2.5 ps was uniformly applied perpendicular to the film plane along the z-axis. The magnetization response (out-of-plane tilt) was recorded at time steps (e.g., 8 ps for total elapsed times up to 8.192 ns, depending on frequency requirements). Then a fast Fourier transform (FFT) in time was applied to the tilt magnetization value at each film pixel. The absorbed power for each pixel was modeled as the square of the tilt amplitude, and the complex phase was determined from the imaginary part of the tilt magnetization. The analysis yields an area map of the tilt amplitude of the pixel moments at a given frequency $f$ (see Fig. SI-11). To obtain the global power spectrum measured in a FMR experiment (see Fig. SI-10a), the total absorbed power at a given frequency was found by summing the power over all pixels.

III. DC Magnetization Results

A. Observation of “Knees” in Experimental and Simulated $M(H,T)$

Highly reproducible knee anomalies are observed in both the experimental and simulated ($T = 0$) $M(H,T)$ data in the low-field, reversal regime that is normally dominated by hysteresis and disordered textures of magnetic domain walls (DW) in unpatterned thin films. The knees are
Fig. SI-7: Experimental Hysteresis Loops for P2T Samples at \( T = 310 \) K

(a) Experimental horizontal DC magnetization \( M_X \) normalized to saturation value \( M_S \) versus applied DC magnetic field \( H \) at temperature \( T = 310 \) K for Sample III134E (3\(^{rd}\) generation, \( W = 85 \) nm). One clear “knee” anomaly is observed at \( H \approx \pm 60 \) Oe; and a broader anomaly is seen at \( H = \pm 300 \) Oe. See Table 1 for details of sample dimensions.

(b) Experimental horizontal DC magnetization \( M_X \) normalized to saturation value \( M_S \) versus applied DC magnetic field \( H \) at temperature \( T = 310 \) K for Sample III130C (8\(^{th}\) generation, \( W = 90 \) nm). Two sharp “knees” are observed at \( H = 0 \) Oe and \( \pm 60 \) Oe; and a broader anomaly is seen at \( H = \pm 280 \) Oe. See Table 1 for details of sample dimensions.

(c) Experimental horizontal DC magnetization \( M_X \) normalized to saturation value \( M_S \) versus applied DC magnetic field \( H \) at \( T = 310 \) K for Sample III133C (12\(^{th}\) generation, \( W = 107 \) nm). Two clear “knees” are visible at \( H = \pm 50 \) Oe and \( \pm 340 \) Oe; a weak knee is seen at \( H = 0 \) Oe. See Table 1 for details of sample dimensions.

(d) Experimental horizontal DC magnetization \( M_X \) normalized to saturation value \( M_S \) versus applied DC magnetic field \( H \) at temperature \( T = 310 \) K for Sample III133E (3\(^{rd}\) generation, \( W = 1100 \) nm). Two “knees” are observed at \( H = \pm 15 \) Oe and \( \pm 20 \) Oe. See Table 1 for details of sample dimensions.
**Fig. SI-8: Simulated Knees in \( M(H) \)**

(a) Simulated horizontal DC magnetization \( M_X \) normalized to saturation value \( M_S \) versus applied DC magnetic field \( H \) at temperature \( T = 0 \) K. “Knee” anomalies are observed at DC field values indicated by arrows. Note the “simulated” pattern parameters are similar to those of several Samples listed in Table 1.

(b) Simulated susceptibility \( \chi = dM/dH \) vs. \( H \) swept from positive (+12 kOe) to negative saturation (-12 kOe). Arrows correspond to knees in the hysteresis in (a). The first small peak (no arrow) at \( H = -0.29 \) kOe corresponds to a very small knee in (a) and the reversal of the first horizontal P2T segment in Fig. SI-9a.

**Fig. SI-9: Relation of Simulated Knees in \( M(H) \) to Magnetization Textures**

(a) Simulated magnetization map of 3rd gen. P2T at \( H = -0.29 \) kOe, corresponding to small \( M(H) \) knee (arrow) in (c). Right box marks first horizontal segment to switch from red to turquoise; left box marks one “wheel” shown in (b). Note mirror symmetry of magnetization texture about the equatorial \( x \)-axis.

(b) Blow-up of (a) showing pinning of DW at vertices joining near-uniformly polarized segments near a “wheel”. Note the “two-in, three-out” segment polarizations at the wheel axle. Nonuniformly polarized segments (infrequently observed) are visible around the rim of the wheel.

(c) Simulated hysteresis of horizontal magnetization \( M_x \) normalized by saturation value \( M_S \) for three P2T of \( W = 50, 100 \) and 250 nm. Applied DC field \( H \) was swept from (12 kOe)x to (-12 kOe)x, and back. A black arrow marks the first (very small) knee observed at \( H = -0.29 \) kOe for \( W = 100 \) nm, corresponding to (a).
fewer and/or more pronounced in the finite-temperature experimental data (see Figs. SI-7, SI-8 and SI-9c).

B. Interpretation of Knees in $M(H,T)$

Our simulations show the knees represent abrupt changes in $M(H,T)$ of the P2T within the low-field, hysteretic regime. The size and sharpness of the knees, which are quite reproducible, depend on the amount of the P2T area that simultaneously reorients at the characteristic field of a given knee. The reproducibility and temperature dependence of the size and number of the knee anomalies suggest that these events are highly cooperative in nature, and may be related to symmetry breaking of the magnetization in the presence of an applied DC field.

The finite P2T under study have five-fold rotational symmetry about the pattern center, and mirror symmetry about their equatorial x-axis. Examination of simulated magnetization maps reveals that domain walls are nearly always located at vertices of the P2T “wire network” (see Figs. SI-9a, SI-9b), and the pattern segments tend to be uniformly polarized over a wide range of applied DC field. The simulated magnetization maps consequently exhibit a high degree of mirror symmetry over a range of field in the hysteretic regime. The hysteretic nature of the low-field regime nevertheless demands that disorder among the segment magnetizations will be present. The low-field reversal is dominated by abrupt reversals of individual segments; but the order of segment reversals strongly depends upon the segment orientation with respect to the applied field. One can discern five distinct subsets of film segments oriented at angles of $0^\circ$, $72^\circ$, $144^\circ$, $216^\circ$, and $288^\circ$ with respect to the x-axis (see Figs. SI-3 and SI-9). Detailed analyses of the complex low-field reversal process are underway.

IV. BB FMR Results

A. Simulated Effects of Film Patterning on FMR Modes at Saturation

Dynamic simulation results for a P2T of $W = 100$ nm are simplified in the case of saturating DC fields where FMR modes are better dispersed and have simple spatial topologies. The frequency dependence of the integrated power for $H = (12$ kOe)$x$ (Fig. SI-10a) exhibits five prominent mode peaks. Note that an unpatterned permalloy film of thickness $t = 25$ nm exhibits only a single uniform mode at $f = 46.33$ GHz. We expect that the influence of pinning of the local magnetization by film segment edges will increase as $W$ decreases [1], as shown in Fig. SI-10b, where simulated mode frequencies exhibit noticeable dependence on $W \approx 100$ nm.

The P2T is completely saturated for $H = 12$ kOe, as shown in the DC magnetization map of Fig. SI-11f. A local power map shown in Fig. SI-11a for Mode 1 at $f = 47.91$ GHz and $H = 12$ kOe involves uniform absorption within single, or two (adjacent) segments oriented at $\phi = 0^\circ$. A local power map shown in Fig. SI-11b for Mode 2 at $f = 46.69$ GHz involves nonuniform
absorption within one or two (adjacent) segments oriented at $\phi = 0^\circ$, as well as activity among 144$^\circ$ and 36$^\circ$ segments. A power map shown in **Fig. SI-11c** for Mode 3 at $f = 43.5$ GHz exhibits absorption exclusively along the edges of 144$^\circ$ and 36$^\circ$ segments. The power map shown in **Fig. SI-11d** for Mode 4 at $f = 41.08$ GHz involves uniform absorption among short chains of segments oriented at $\phi = \pm 72^\circ$ and $\pm 108^\circ$. Mode 5 at $f = 35.2$ GHz, is similar to Mode 4, but tightly confined to edges of film segments oriented at $\phi = \pm 72^\circ$ and $\pm 108^\circ$. The lower frequency and higher resonance field for Mode 5, as compared to Modes 2 and 3 for 144$^\circ$ and 36$^\circ$ segments, appears due to higher demagnetizing fields present in the 72$^\circ$ case. Similar observations can be made for additional modes active in near-saturated fields (e.g., for 2 kOe $< |H| < 12$ kOe).

**Fig. SI-10: Effect of P2T Segment Width W on Mode Frequencies at Saturation**

(a) Simulated global power vs. frequency for 3$^{rd}$ generation P2T with $W = 100$ nm in applied field $H = (12$ kOe)$x$. Note the P2T segment dimensions shown. (b) Simulated FMR mode frequencies $f$ as functions of segment width $W$. Lines are guides to the eye; and the numbers correspond to modes defined in (a).
Fig. SI-11: FMR Power Absorption Maps for Symmetric Modes at Saturation

(a) Local power map for Mode 1 (Fig. SI-10) at \( f = 47.91 \) GHz and \( H = 12 \) kOe. Color scale indicates the magnetization tipping amplitude (red is greatest, dark blue is smallest).

(b) Local power map for Mode 2 (Fig. SI-10) at \( f = 46.69 \) GHz and \( H = 12 \) kOe. Color scale indicates the magnetization tipping amplitude (red is greatest, dark blue is smallest).

(c) Local power map for Mode 3 (Fig. SI-10) at \( f = 43.5 \) GHz and \( H = 12 \) kOe. Color scale indicates the magnetization tipping amplitude (red is greatest, dark blue is smallest).

(d) Local power map for Mode 4 (Fig. SI-10) at \( f = 41.08 \) GHz and \( H = 12 \) kOe. Color scale indicates the magnetization tipping amplitude (red is greatest, dark blue is smallest).
(e) Local power map for Mode 5 (Fig. SI-10) at $f = 35.2$ GHz and $H = 12$ kOe. Color scale indicates the magnetization tipping amplitude (red is greatest, dark blue is smallest).

(f) Simulated DC magnetization map for $H = (12 \text{ kOe}) \times$. Color scale ($\phi = 0^\circ$, red; $\phi = 180^\circ$, turquoise) and small arrows indicate a saturated magnetization at start of a negative field sweep.
B. Observations of Asymmetric Modes in Low-Field Reversal Regime

1. Experimental Definition of Asymmetric Modes

Representative BB FMR spectra for Sample III129A for frequencies near 12 to 15 GHz are shown in Figs. SI-12a through SI-12f. These data were acquired in fields \( H = H_x \) that span the strongly hysteretic (e.g., \( |H| < 1 \text{ kOe} \)) and near-saturated (\( |H| > 1 \text{ kOe} \)) regimes. In the near-saturated regime, “symmetric” mode signatures repeat in both sweep directions, in spite of finite hysteresis (see Fig. SI-12a). We also observe “asymmetric” modes defined by their presence on only one side of the field origin in a given sweep; nevertheless, the asymmetric signatures quantitatively reproduce in opposite field sweeps. Remarkably, asymmetric modes are only observed in the experimental FMR power absorption in the lower-field regime where the knees in \( M(H,T) \) are observed.

Figure SI-13 shows a frequency-field dispersion plot for Sample III129A, which summarizes experimentally observed modes that span both the low-field, reversal and near-saturated regimes. We have identified distinct mode “Branches” in the data shown in Fig. SI-13 (note asymmetric Branches 10 and 11).

Fig. SI-12: BB FMR Absorption Derivative in Low-Field Regime

(a) Experimental BB FMR absorption derivative vs. DC field \( H \) for Sample III129A at frequency \( f = 15 \text{ GHz} \). Note reproducible, symmetric spectra for \( |H| > 1 \text{ kOe} \), and strong hysteresis for \( |H| < 1 \text{ kOe} \). Inset defines angle \( \phi = 0^\circ \) between central Penrose star and \( H \parallel x \)-axis.

(b) Blow-up of low-field regime of (a), showing an asymmetric anomaly near \( H = -0.20 \text{ kOe} \), which is very near Branch 10 in Fig. SI-13 (however, this anomaly is not a clearly defined resonant signature, and is therefore omitted in Branch 10).
(c) Experimental BB FMR absorption derivative vs. DC field $H$ for Sample III129A at frequency $f = 13.5$ GHz. Note reproducible, symmetric spectra for $|H| > 1$ kOe. Arrows mark clear asymmetric mode signatures near $|H| = 0.1$ kOe, corresponding to Branch 11 in Fig. SI-13.

(d) Experimental BB FMR absorption derivative vs. DC field $H$ for Sample III129A at frequency $f = 13.0$ GHz. Note reproducible, symmetric spectra for $|H| > 1$ kOe. Arrows mark clear asymmetric mode signatures near $|H| = 0.1$ kOe, corresponding to Branch 11 in Fig. SI-13 and the mode in (e), which is dispersing toward $H = 0$ with decreasing frequency.

(e) Experimental BB FMR absorption of Sample III29A at low fields for $f = 10$ GHz and $\phi = 0^\circ$. Note reproducible, symmetric spectra for $|H| > 1$ kOe. Asymmetric modes only occur on one side of the field origin for $|H| < 1$ kOe.

(f) Blow-up of low-field regime of (a). At least one asymmetric mode is visible over the interval $50 \leq |H| \leq 200$ Oe. This group of unresolved asymmetric modes extends well below Branch 11 in Fig. SI-13.
(g) Simulated global power vs. frequency for 3\textsuperscript{rd} generation P2T with parameters shown in applied fields $H = (\pm 1.0 \text{ kOe})\mathbf{x}$. Note the near-perfect overlap of the FMR response for symmetric modes.

(h) Simulated global power vs. frequency for 3\textsuperscript{rd} generation P2T with parameters shown in applied fields $H = (\pm 0.46 \text{ kOe})\mathbf{x}$. Note the clear hysteresis of the FMR response for asymmetric modes.

It is important to note that \textit{simulated global power spectra also exhibit the phenomenon of asymmetric modes} quite clearly (see Fig. SI-12g and SI-12h). Other asymmetric modes exist at frequencies below 10 GHz; however, the dispersion of the modes with DC field shown in Fig. SI-13 makes it hard to resolve the lower frequency behavior. Additional high-resolution BB FMR experiments are underway to yield better-resolved BB FMR spectra for the low-field, low-frequency regimes.

2. Origin of Asymmetric Modes: Comparisons of Dynamic Simulations with $M(H,T)$ Knees

Comparisons of simulated power absorption and static magnetization maps, and global power spectra (see Fig. SI-14) provide interesting insights into the origins of low-field asymmetric modes that are sufficiently separated in their resonance fields for $f > 10$ GHz. It is also important to relate the knees observed in $M(H,T)$ to sharp, reproducible peaks visible in the low-field BB FMR absorption derivative.

For example, we focus here on Branch 11 in Fig. SI-13, which can be associated with a mode localized within or near 0$^\circ$ segments: The FMR absorption for Branch 11 frequencies near 13.5 GHz is localized within the first few 0$^\circ$ segments to switch in negative field sweeps, as shown in Figs. SI-14a through SI-14c and Fig. SI-9a. The amplitude of this Branch increases rapidly as more 0$^\circ$ segments reverse their magnetization; and as additional horizontal segments reverse with decreasing DC field, a more complex, “nonlocal” FMR response in segments surrounding switched segments is apparent, as shown in Figs. SI-14d through SI-14f. A similar
nonlocal response is evident in other low-field modes, as shown in Fig. SI-14g through SI-14i. These results imply that the low-field dynamics of the P2T segments is collective and nonlocal in character, rather than dominated by single-segment effects (e.g., segment orientation with respect to applied field and demag fields). Additional analyses that consider the role of P2T vertices and near-neighbor effects in reversal are underway.

**Fig. SI-13: Experimental Frequency-Field Dispersion**

Experimental resonance frequency $f$ vs. DC field $H$ obtained from BB FMR absorption data taken during field sweeps at fixed frequencies. Numbers denote mode Branches. Asymmetric Branches 10 and 11 are represented by only negative field sweep data for clarity. Note that at least two additional Branches (not numbered) appear below $f = 12$ GHz; and the dispersion of modes toward $H = 0$ complicates extension of this plot to lower frequencies (see Figs. SI-12g and 12h). (The numbering scheme for the “Branches” is not the same as for the “Modes” shown in Fig. SI-10).
**Fig. SI-14: Simulated Magnetization Maps, Power Absorption Maps, and FMR Power Spectra in the Low-Field, Reversal Regime**

(a) Simulated DC magnetization map for $H = (-0.33 \text{ kOe})\hat{x}$. Color scale indicates the magnetization direction; horizontal ($\phi = 0^\circ$, red) segments begin switching (to $\phi = 180^\circ$, turquoise) at $H = (-0.29 \text{ kOe})\hat{x}$ during a negative field sweep starting at $H = +12 \text{ kOe}$.

(b) Simulated power absorption map for $f = 13.43 \text{ GHz}$, corresponding to (a) and (e). Color scale indicates magnetization tipping amplitude (red is greatest). Note strong correlation between switched (turquoise) segments in (a) and resonant (red) segments in (e).

(c) Simulated global power vs. frequency for $H = (-0.33 \text{ kOe})\hat{x}$. A peak at $f = 13.43 \text{ GHz}$, which first emerges for $H = -0.29 \text{ kOe}$, is indicated by a red arrow.

(d) Simulated DC magnetization map for $H = (-0.46 \text{ kOe})\hat{x}$. Color scale indicates the magnetization direction; horizontal ($\phi = 0^\circ$, red) segments continue switching (to $\phi = 180^\circ$, turquoise) during negative field sweep starting at $H = +12 \text{ kOe}$.

(e) Simulated power absorption map for $f = 13.72 \text{ GHz}$, corresponding to (d) and (f). Color scale indicates magnetization tipping amplitude (red is greatest). Note “standing wave” response within switched (turquoise) $0^\circ$ segments in (d), and more uniform response among nearby segments of other orientations.

(f) Simulated global power vs. frequency for $H = (-0.46 \text{ kOe})\hat{x}$. A peak at $f = 13.72 \text{ GHz}$, which first emerges for $H = -0.29 \text{ kOe}$, is very intense at this field, as indicated by a red arrow.
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<th>(g) Simulated DC magnetization map for $H = (-0.70 \text{ kOe}) \mathbf{x}$: Color scale indicates the magnetization direction ($\phi = 180^\circ$ is turquoise) during negative field sweep starting at $H = +12 \text{ kOe}$.</th>
<th>(h) Simulated power absorption map for $f = 12.35 \text{ GHz}$, corresponding to (g) and (i). Color scale indicates magnetization tipping amplitude (red is greatest). Note the resonant (red) response is extended over many segments of various orientations.</th>
<th>(i) Simulated global power vs. frequency for $H = (-0.70 \text{ kOe}) \mathbf{x}$. A peak at $f = 12.35 \text{ GHz}$ is very intense at this field, as indicated by red arrow.</th>
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### C. Ten-Fold Rotational Symmetry of Symmetric Modes

We have observed ten-fold rotational symmetry of the experimental FMR spectra in the near-saturated regime, as shown in the BB FMR data of Fig. SI-15. Ten-fold symmetry is expected for an infinite P2T; and this behavior could indicate that the demagnetizing fields of the segments on the edges of the finite sample decagons are too weak to break the ten-fold symmetry expected for an infinite P2T. Alternatively, a rotation of our finite P2T by $180^\circ$ causes segments at field angles $\phi = \pm 36^\circ$ ($\pm 72^\circ$) to make angles of $\pm 144^\circ$ ($\pm 108^\circ$), respectively; however, a simple counting exercise shows the number of segments with uniform polarizations oriented at particular angles with respect to $H$ remains unchanged. If this is the main factor controlling the sample FMR response in a near-saturated regime, the total power absorption will exhibit ten-fold rotational symmetry.

Additional rotation studies were carried out on 3rd generation Sample III133E, which has the same aspect ratio $d_1/d_2$, but segment lengths and width increased by $\sim 10$ times, as compared to 8th generation Sample III129A (see Table SI-1). These dimensions yield a very low coercive field $H_C = 72 \text{ Oe}$ (compared with 710 Oe for 8th generation III130C), which places the spectra shown in Fig. SI-16 well into the saturated regime.

We have performed dynamic simulations for the saturated state ($H = 12 \text{ kOe}$) of a 3rd generation P2T ($W = 100 \text{ nm}$) to investigate the effect of in-plane rotations of the P2T with respect to applied field ($H = H_x$). The FMR spectra exhibited ten-fold rotational symmetry, although the finite P2T tiling is only five-fold rotationally symmetric. Additional rotation studies of other P2T generations are clearly of interest.
Fig. SI-15: BB FMR Spectra versus DC Field Angle Exhibiting 10-Fold Symmetry

BB FMR absorption derivative versus applied DC magnetic field \( H \) of 8th generation Sample III129A (\( W = 135 \text{ nm} \)) in negative field sweeps at room temperature for microwave frequency \( f = 15.5 \text{ GHz} \). Different colors of curves correspond to data taken at different angles \( 0^\circ \leq \phi \leq 40^\circ \) between the horizontal reference direction of the sample (see Fig. SI-3) and \( H \); adjacent curves correspond to field angles incremented by \( \Delta \phi = 4^\circ \). These data correspond to the near-saturated regime, and exhibit ten-fold rotational symmetry (black, blue curves reproduce for \( \Delta \phi = 36^\circ \)).
Fig. SI-16: NB FMR Spectra versus DC Field Angle Exhibiting 10-Fold Symmetry

![NB FMR Spectra Chart](chart.png)

NB FMR spectra of Sample III133E at fixed $f = 9.61$ GHz for $0^\circ < \phi < 180^\circ$. Note the ten-fold rotational symmetry for $H > 20H_C$ (near-saturated regime).

V. References


