## Frustration & Dynamics in Artificial Spin Ice

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#### Dynamics and hysteresis in square lattice artificial

**Spin ice** G M Wysin<sup>1</sup>, W A Moura-Melo<sup>2</sup>, L A S Mól<sup>2,3,4</sup> and A R Pereira<sup>2</sup> New Journal of Physics **10P** Institute of Physics **DEUTSCHE PHYSIKALISCHE GE** 

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Figure 1. A  $16 \times 16$  model system with  $d = k_1 = k_3 = 0.1$ , in a metastable state at temperature  $k_{\rm B}T/\varepsilon = 0.025$ , from a hysteresis scan (this is a state at  $h_{\rm ext} = 0$ ). Most of the system is locally close to the Z = +1 ground state. The upper righthand corner is locally near the Z = -1 ground state, and there is a bent domain wall connecting the two regions. For interior charge sites (junction points of four islands), there happens to be no discrete monopole charge present: all  $q_k = 0$  and the discrete  $\rho_{\rm m} = 0$ .

Artificial spin-ice. Arrays of elongated magnetic islands, dominated by anisotropy & dipole-dipole interactions.

Each arrow = one island.

Island rows are alternately aligned along x or y-axes in this artificial square ice.

This system has two degenerate ground states.

Mimics the behavior of 3D spin ices of rare earths in lattice of corner sharing tetrahedra of a pyrochlore structure. in Review article: **Advances in artificial spin ice**, Sandra Skjærvø et al. Nat. Rev. Phys. 11/08/19.

Artificial spin ices are metamaterials made up of coupled nanomagnets arranged on different lattices that exhibit a number of interesting phenomena, such as emergent magnetic monopoles, collective dynamics and phase transitions.

The ability to create thermally active artificial spin ices with fluctuating moments at room temperature makes it possible to explore the rich phase diagrams with phases that are determined by the geometry, temperature and disorder.

Signatures of the magnetic configurations are given by the specific spin-wave resonances in artificial spin ice, which offer a platform for programmable spin-wave devices, in particular magnonic crystals.

The established artificial spin ices are arranged on square and kagome lattices. New geometries include both periodic and aperiodic, different magnet shapes and anisotropies, and 3D structures.

Future work involves developments in fabrication and characterization methods, the study of artificial spin systems with new geometries and combinations of materials, and the development of applications including computation, data storage, encryption and reconfigurable microwave circuits. Example of a rare-earth pyrochlore compound.

Pr spins at corners of tetrahedrons.



The interesting properties of Pr<sub>2</sub>Ir<sub>2</sub>O<sub>7</sub> are rooted in its crystal structure, called a pyrochlore lattice: four praseodymium (Pr) ions, each of which carries a magnetic 'spin', form a tetrahedral cage around an oxygen (O) ion. At low temperatures, the spins of materials with this structure often 'freeze' into what is called a 'spin ice' (Fig. 1) because of its similarity to the way hydrogen ions form around oxygen in water ice. (phys.org/news/)



#### Realization of Rectangular Artificial Spin Ice and Direct Observation of High Energy Topology

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Atomic force microscope topography, 300 x 100 x 20 nm islands.

Artificial spin-ice. Arrays of elongated magnetic islands, dominated by anisotropy & dipole-dipole interactions.



Magnetic force microscope image showing N (bright) and S (dark) poles.

Magnetic Nano-Islands (elements of artificial spin-ice)



Approx. 50 nm - 5 µm wide but only 10 nm thick. Individual & in arrays, high-permeability soft magnetic materials. Grown with techniques of epitaxy & lithography on a non-magnetic substrate. Form arrays of particles that can interact with each other or applied fields.

Primary physics effects magnetostatics controlled by island geometry. discrete energy states for data storage. spintronics controlled by current injection. magnetic oscillators controlled by applied fields. frustration in ordered arrays of islands (artificial spin-ice).

Several principle states of a nano-island:

(1) quasi-single domain; (2) vortex; (3) multi-domains & domain walls.

~ increasing size ~

Topics for study in the islands:

I) Vortices. The static and dynamic properties of single vortices. They behave very much as particles with charges.

2) Magnetostatic anisotropy of the islands themselves. Also known as shape anisotropy because it depends mostly on the surfaces.



3) Spin-ices, frustration. Especially for elongated islands with Isinglike states, interactions within their arrays, that lead to frustrated statics and dynamics.

# Quasi-single-domain state. ĥ ĥ S pole, $\sigma_M < 0.$

Magnetization M determines an effective surface charge density:

N pole, σ<sub>M</sub>>0.

$$\sigma_M = \vec{M} \cdot \hat{n},$$

The poles produce large stray-field energy.

But ferromagnetic exchange energy is small.

#### Vortex state

Very little magnetic surface charge density. Stable only above a minimum radius



$$\sigma_M = \vec{M} \cdot \hat{n},$$

Has small poles  $(\sigma_M = \pm M_z)$  only in the core.

The stray-field energy is small.

But the ferromagnetic exchange energy is large.



Micromagnetics. Y A technique for studying a continuous system.

Each cell contains a magnetic dipole:

$$\hat{m} = \vec{M}/M_S.$$

a

a



 $\blacktriangleright Divide the sample into cells of size a x a x L.$ 

Assume that the magnetization is saturated ( $M_s$ ) inside each cell: |m|=1. Only the directions vary between cells.

The cells interact as dipoles, with exchange energy between neighbors & with the demagnetization field.

Micromagnetics Hamiltonian:  $H=H_{ex}+H_{demag}+H_B$   $m_i$ exchange:  $\mathcal{H}_{ex} = A \int dV \ \nabla \hat{m} \cdot \nabla \hat{m},$ 

magnetostatic  $\mathcal{H}_{dd} = \mathcal{H}_{demag} = -\frac{1}{2}\mu_0 \int dV \ \vec{H}_M \cdot \vec{M}$  (demagnetization):

applied field: 
$$\mathcal{H}_B = -\mu_0 \int dV \ \vec{H}_{\text{ext}} \cdot \vec{M}$$

Statics: minimize the energy  $\Rightarrow$  stable configurations. Dynamics: equation of motion  $\Rightarrow$  periodic configurations.

Difficulties:

(i) Calculating the demagnetization field  $H_M$ ;

(ii) Enforcing a desired initial position, X, of a vortex  $\Rightarrow$  E(X).

Scale energies by the exchange between cells:

$$J_{\rm cell} = \frac{2Av_{\rm cell}}{a^2} = 2AL.$$

"magnetic exchange length"

$$\lambda_{\rm ex} = \sqrt{\frac{2A}{\mu_0 M_S^2}}$$

demag. field:

 $\vec{H}_M = M_S \tilde{H}_M$ 

Hamiltonian on the grid of cells:

$$\mathcal{H}_{\rm mm} = -J_{\rm cell} \left\{ \sum_{(i,j)} \hat{m}_i \cdot \hat{m}_j + \left(\frac{a}{\lambda_{\rm ex}}\right)^2 \sum_i \left(\tilde{H}_{\rm ext} + \frac{1}{2}\tilde{H}_M\right) \cdot \hat{m}_i \right\}$$

Need 
$$\left(\frac{a}{\lambda_{ex}}\right)^2$$
 less than 1 for reliable solutions.  
(cells smaller than exchange length)

Model for magnetic anisotropy of elliptical islands. Total magnetic dipole moment =  $\mu$ . Single domain is assumed and  $\mu$  has a fixed magnitude.

$$E = E_0 + K_1 [1 - (\hat{\mu} \cdot \hat{x})^2] + K_3 (\hat{\mu} \cdot \hat{z})^2$$

Include also applied field energy: -µ\*H<sub>ext</sub>





Figure 5. Magnetic configurations for a 480 nm × 240 nm × 24 nm particle with magnetic field applied at +45° above a horizontal axis pointing to the right. The arrows are the coarse-grained averages of 9 × 9 groups of cells. In (a), the external field is h = 0.20; in (b) h = 0.0; (c) h = -0.025, just before reversal; (d) h = -0.027, just after reversal. Note the enhanced curvature of the field compared to that in the smaller particle in figure 4.







The results confirm the particle anisotropy for  $L_X \times L_y \times L_z$  particles with high aspect ratios  $L_X/L_y$ :

$$E = E_0 + K_1 [1 - (\hat{\mu} \cdot \hat{x})^2] + K_3 (\hat{\mu} \cdot \hat{z})^2$$

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**Table 1.** Values of the in-plane anisotropy constant  $K_1$  and out-of-plane anisotropy constant  $K_3$  in units of  $J = 2AL_z$  for different particle sizes and aspect ratios  $g_1 = L_x/L_y$ . All of the particles calculated have  $g_3 = L_x/L_z = 20$ .

	$L_x$								
	120 nm		240 nm		480 nm				
<b>g</b> 1	$K_1$	$K_3$	$K_1$	$K_3$	$K_1$	$K_3$			
2	6.35J	72.7 <i>J</i>	27.3J	287J	111 <i>J</i>	1140J			
3	7.32J	43.4J	31.9 <i>J</i>	169 <i>J</i>	134 <i>J</i>	670J			
5	6.96J	21.1 <i>J</i>	31.5J	79.9J	133J	311 <i>J</i>			
8	7.39J	8.30J	29.5J	33.1 <i>J</i>	118J	132 <i>J</i>			



Anisotropy constants per unit volume depend mainly on aspect ratios.



aspect ratios



**Figure 3.** The anisotropy constants  $K_1$  (solid curves) and  $K_3$  (dashed curves) scaled by elliptical particle volume, versus particle lengths, for the indicated  $g_1$  aspect ratios. All data has  $g_3 = 20$ . The values of K/V are given in units of  $A \text{ nm}^{-2}$ , where A is the exchange stiffness.  $K_1/V$  increases with aspect ratio while  $K_3/V$  decreases, and they become equal at high aspect ratio.

## What is frustration?



FM - unique ground state no frustration

Anti-FM - 2 ground states but still no frustration Anti-FM - multi ground states with frustration. Not all bond energies can acquire their minima.





FIG. 2: (a) Configuration of the ground-state obtained for L = 6a, in exact agreement with that experimentally observed. Note that the ice rules are manifested at each vertex. This is the case in which the topology demands the minimum energy (see Fig. (3)). (b) Another configuration also respecting the ice rule, but displaying a topology which costs more energy. (Mol et al 2008.)



#### quantized excitations in a vertex



#### energies of Ising-like states of a vertex



#### deviations from the ice rule

 $\Rightarrow$  higher energy and monopole "charges"



FIG. 3: The 4 distinct topologies and the 16 possible magnetic moment configurations on a vertex of 4 islands. Although configurations (a) and (b) obey the ice rule, the topology of (a) is more energetically favorable than that of (b). Hamiltonian (1) correctly yields to the true ground-state based on topology (a), without further assumptions. Topologies (c) and (d) does not obey the ice rule. Particularly, (c) implies in a monopole with charge  $Q_M$ .

How do the excitations behave as particles, interact with each other, and contribute to thermodynamics? With temperature T>0. For the movement in one cell:

$$\frac{d\hat{m}}{d\tau} = \hat{m} \times \left(\vec{b} + \vec{b}_s\right) - \alpha \hat{m} \times \left[\hat{m} \times \left(\vec{b} + \vec{b}_s\right)\right]$$
  
stochastic fields

fluctuation-dissipation theorem:

$$\langle b_s^{\alpha}(\tau) \, b_s^{\beta}(\tau') \rangle = 2\alpha \, \mathcal{T} \, \delta_{\alpha\beta} \, \delta(\tau - \tau') \qquad \qquad \mathcal{T} \equiv \frac{kT}{J_{\text{cell}}} = \frac{kT}{2AL}$$

(the stochastic fields carry thermal energy & power)

We can integrate with Heun's 2<sup>nd</sup> order algorithm:

- A. Euler predictor step.
- B. Trapezoid corrector step.



$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.01 $\approx$  ground state

$$D = \frac{\mu_0}{4\pi} \frac{\mu^2}{a^3}$$



$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.10

> ground state



$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.14 few monopoles

D_h=0,00000	kT=0,2200	0 E=-198,30	)6 n1=38 r	2=0 Z=0,57	78 Z2=0,594	1 mb=-0.00	0,000
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$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.22

 $\approx$  transition to high-T phase

ND h=0.00000 kT=0.30000 E=-115.030 n1=71 n2=4 Z=-0.008 Z2=0.456 mb=0.001 +- 0.0006
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Sus 1/1 512 Spips79 pp=75 sgl=71 multi=4State 74/103

$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.30

 $\approx$  high-T disorder

Typical thermodynamics shows a phase transition





scanning through applied magnetic field.





$$D = 0.000835$$
  
K<sub>1</sub>=0.0897  
K<sub>3</sub>=0.2000

kT=0.001

 $\neq$  ground state

(from long-time Langevin dynamics)

Note: 300 K is  $kT = 1.29 \times 10^{-5}$  $D = \frac{\mu_0}{4\pi} \frac{\mu^2}{a^3}$ 



D = 0.000835K<sub>1</sub>=0.0897K<sub>3</sub>=0.2000

kT=0.01

 $\neq$  ground state



D = 0.000835K<sub>1</sub>=0.0897K<sub>3</sub>=0.2000

kT=0.015

more monopoles

END h=0.00000 kT=0.02400 E=13.670 n1=107 n2=18 Z=-0.084 Z2=0.793 mb=0.001 +- 0.0005 ¥ 🕀 ┍⊕≪.⊕≪ (<del>+</del>)× (H) < k  $\oplus \Delta f \oplus \not \supset \ominus$ Œ ¢ ¥⊕**≠**⊖⊻⊕¥⊝ ⊝ᡧ⊕ᢆᡧ᠊⊕ <u>≯⊝≮</u> 4-⊕-? ⊝≮⊶⊕  $(-) \triangleleft$ ふ(日) \$~⊕  $\sim$ ⊕⋪⊝≁⊕ 🖉 🕀 🧷 70.27 ⊲ ⊕∿⊕∆∽ Ð 4-⊕₹ <...(⊕) ×⊖¤ \$-⊕\$~⊕ \$ Ľ æ  $\Theta \leftarrow \Theta = \Theta$ ≁⊕÷⇒⊝≁ >① 🔊 4 €₹  $\boldsymbol{\omega} \Theta$ A-⊕ € Θ ⊝๙∽⊕๙∽ œ  $\oplus$   $\rtimes \ominus$ State 80/103 Sys 1/1, 512 Spins qm=143, np=125, sgl=107, multi=18

D = 
$$0.000835$$
  
K<sub>1</sub>= $0.0897$   
K<sub>3</sub>= $0.2000$ 

kT=0.024

more monopoles

END h=0.00000 kT=0.04000 E=22.642 n1=114 n2=13 Z=-0.049 Z2=0.670 mb=0.004 +- 0.0005 ⊕ॎ∽₽ æ >⊖ \ ⊕ <del>(</del>} <u>ا</u> ⊕ 🕂 ⊝ 🛠-⊕  $\Sigma$  $(\oplus)$ Θ ≻⊝∿∼⊝  $\Theta \Delta$ Ð ⊝≁⊕ ⊝∻  $\oplus$  $\Delta s$ æ ብ 🔀 ብ ē  $\mathcal{A} \ominus$ ⋪⊝�  $\ominus \nabla \ominus \leftarrow$ 4 医 (田)  $4 - \oplus 4 +$ <u>∧⊝</u>∢- $\neg \nabla$ Ð ⅊ℰ ᡣ᠐ᡌ⊕ qm=140, np=127, sgl=114, multi=13 Sys 1/1, 512 Spins State 64/103 D = 0.000835K<sub>1</sub>=0.0897K<sub>3</sub>=0.2000

kT=0.040

highly disordered

START h=0.00000 kT=0.02000 E=-15.692 Lx=13.0 Ly=11.3 N=123 nq=66 R K > 7 A K  $\wedge$  $\Lambda$ 个 1 Ľ Ľ 4 > 7  $\Lambda$ NK 7 A K > 7 4 ース  $\wedge$ Λ  $\sqrt{}$ 个 R K X 7 ス N K 1 K A Ľ  $\wedge$ 小  $\wedge$ N K RK  $\mathbb{K} \not \subseteq \mathbb{N} / \mathbb{N}$ Ľ ース  $\wedge$ Λ Λ 个 R K 🖌 🗡 N K 7 1 Z K K  $\wedge$  $\wedge$ A K > 7 K RK ス Sys 1/1, 123 Spins State 1/4

40

D = 0.1 $K_1 = 0.1$  $K_3 = 0.5$ 

1 of 6 ground states



D = 0.1 $K_1 = 0.1$  $K_3 = 0.5$ 

1 of 6 ground states

all vertices have a monopole charge.



$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.01 (low T).

Frustrated state does not approach ground state.



$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.05



$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.1 (moderate) multi-charge poles



$$D = 0.1$$
  
 $K_1 = 0.1$   
 $K_3 = 0.5$ 

kT=0.3 (high)

many multi-charge poles

What about linearized (small amplitude) oscillations at low temperature? (work by Thomas Lasnier)



A square ice ground state. Notation for deviations around a site A. Keep only near neighbor interactions. There are four sublattices! Hamiltonian:

$$\mathcal{H} = -\frac{\mu_0}{4\pi} \frac{\mu^2}{a^3} \sum_{i>j} \frac{[3(\hat{\mu}_i \cdot \hat{\mathbf{r}}_{ij})(\hat{\mu}_j \cdot \hat{\mathbf{r}}_{ij}) - \hat{\mu}_i \cdot \hat{\mu}_j]}{(r_{ij}/a)^3} \\ + \sum_i \{K_1 [1 - (\hat{\mu}_i \cdot \hat{\mathbf{u}}_i)^2] + K_3 (\hat{\mu}_i \cdot \hat{\mathbf{z}})^2\}$$

effective field:  $\mathbf{B}_i = -\frac{\delta \mathcal{H}}{\delta \boldsymbol{\mu}_i}$  Eq. of motion:  $\frac{d\hat{\boldsymbol{\mu}}_i}{dt} = \gamma_{\rm e}\hat{\boldsymbol{\mu}}_i \times \mathbf{B}_i.$ 

$$oldsymbol{\hat{\mu}}_i \equiv \mathbf{A}$$

$$\frac{d\mathbf{A}}{dt} = \mathbf{A} \times \mathbf{F}(\mathbf{A})$$

$$\mathbf{F}(\mathbf{A}) = \kappa_1 A_x \mathbf{\hat{x}} - \kappa_3 A_z \mathbf{\hat{z}} + \delta_1 \Big\{ 3 \left[ (\mathbf{B}^{\uparrow} + \mathbf{B}^{\downarrow}) \cdot \mathbf{\hat{r}}_{xy} \right] \mathbf{\hat{r}}_{xy} - \mathbf{B}^{\uparrow} - \mathbf{B}^{\downarrow} + 3 \left[ (\mathbf{D}^{\uparrow} + \mathbf{D}^{\downarrow}) \cdot \mathbf{\hat{r}}_{\bar{x}y} \right] \mathbf{\hat{r}}_{\bar{x}y} - \mathbf{D}^{\uparrow} - \mathbf{D}^{\downarrow} \Big\}$$

Also get eqns. for dB/dt, dC/dt, dD/dt .

Linearization: Assume small deviations:

$$\mathbf{A} = \mathbf{A}_0 + \mathbf{a} = (1 + a_x, a_y, a_z), \\ \mathbf{B} = \mathbf{B}_0 + \mathbf{b} = (b_x, 1 + b_y, b_z), \\ \mathbf{C} = \mathbf{C}_0 + \mathbf{c} = (-1 + c_x, c_y, c_z), \\ \mathbf{D} = \mathbf{D}_0 + \mathbf{d} = (d_x, -1 + d_y, d_z).$$



and traveling waves:

$$b_x(\mathbf{r},t) = b_x \mathrm{e}^{\mathrm{i}(\mathbf{q}\cdot\mathbf{r}-\omega t)},$$
  
wave vector  
frequency

You get a simple 8x8 eigenvalue problem.

$$\begin{aligned} -\mathrm{i}\omega a_y &= \delta_1(6a_z + ub_z + vd_z) + \kappa_{13}a_z, \\ -\mathrm{i}\omega a_z &= \delta_1(-6a_y + \frac{3}{2}ub_x - \frac{3}{2}vd_x) - \kappa_1a_y, \\ -\mathrm{i}\omega b_x &= \delta_1(-6b_z - ua_z - vc_z) - \kappa_{13}b_z, \\ -\mathrm{i}\omega b_z &= \delta_1(6b_x - \frac{3}{2}ua_y + \frac{3}{2}vc_y) + \kappa_1b_x, \\ -\mathrm{i}\omega c_y &= \delta_1(-6c_z - ud_z - vb_z) + \kappa_{13}c_z, \\ -\mathrm{i}\omega c_z &= \delta_1(6c_y - \frac{3}{2}ud_x + \frac{3}{2}vb_x) - \kappa_1c_y, \\ -\mathrm{i}\omega d_x &= \delta_1(6d_z + uc_z + va_z) - \kappa_{13}d_z, \\ -\mathrm{i}\omega d_z &= \delta_1(-6d_x + \frac{3}{2}uc_y - \frac{3}{2}va_y) + \kappa_1d_x. \end{aligned}$$
  
interaction 
$$\mathbf{v} = 2\cos[\frac{a}{2}(q_x - q_y)] \\ \mathbf{u} = 2\cos[\frac{a}{2}(q_x + q_y)], \end{aligned}$$

Dipole

2 are <u>antisymmetric</u> across the vertex:



Mode A- has the lowest frequency at long wavelength.

Mode A+ has the highest frequency at long wavelength.

2 are <u>antisymmetric</u> across the vertex:





Mode A- has the lowest frequency at long wavelength.

Mode A+ has the highest frequency at long wavelength.

2 are <u>symmetric</u> across the vertex:



Modes S- and S+ are nearly degenerate at long wavelength.

2 are <u>symmetric</u> across the vertex:



Modes S- and S+ are nearly degenerate at long wavelength.

#### An excitation spectrum for weak island anisotropy



But should plot spectrum in 1st Brillouin zone.

The islands' unit cell is tilted at 45 degrees and the islands' lattice constant is a'=a/sqrt(2).





square lattice Brillouin zone.

Plotting spectrum in 1st Brillouin zone.



Future work & improvements

I) Dynamics in remnant state?



2) Beyond nearest neighbors. Include long-range dipole interactions.

## Summary

Shape anisotropy of magnetic islands has a strong effect on the states.

Anisotropy coefficients for islands used in artificial spin ice are found from the effective potential of the magnetic moment in an island.

A model is developed for spin-ice with continuous dynamics, based on island dipoles which can point in any direction, while constrained by easy-plane and uniaxial anisotropies.

The linearized mode spectrum around a ground state in square ice can be partitioned into symmetric and antisymmetric states; the lowest frequency mode is <u>antisymmetric</u> at long wavelength.

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