

# Tutorial Talk

1. Feel free to ask questions at any time
2. If anything is under/misrepresented  
- please let me know !

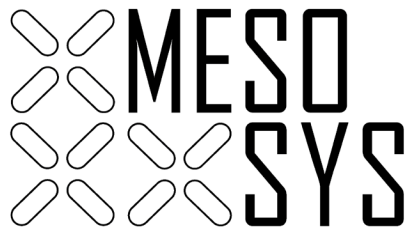
→ A review on 3D artificial spin ice, partly based on this presentation, was submitted in July 2024.

L Berchiolla, GM Macauley and LJ Heyderman (2024)

Prof. Laura Heyderman :: ETH Zurich - Paul Scherrer Institute

## Three Dimensional Artificial Spin Ice

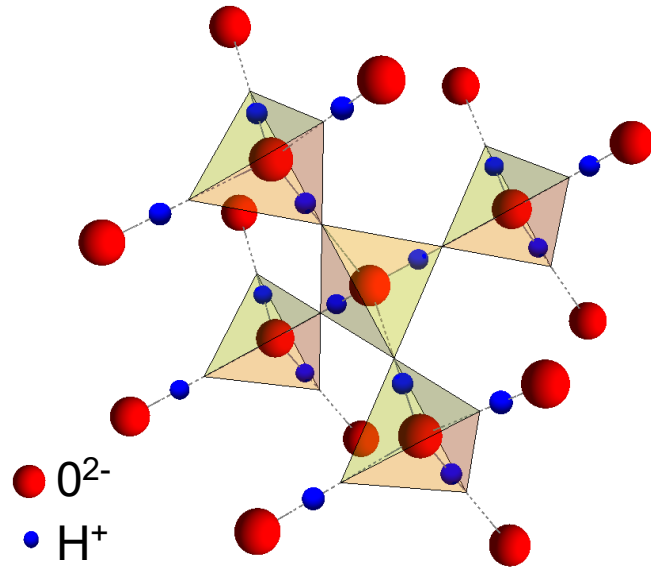
SPICE Workshop on Nanomagnetism in 3D, April/May 2024



Mesoscopic Systems

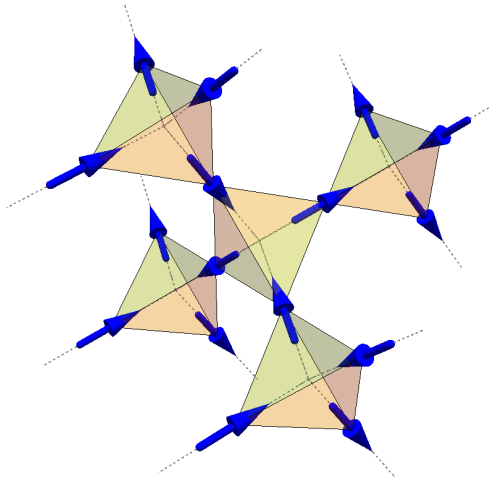
<http://www.mesosys.mat.ethz.ch>

# From Water Ice to Artificial Spin Ice



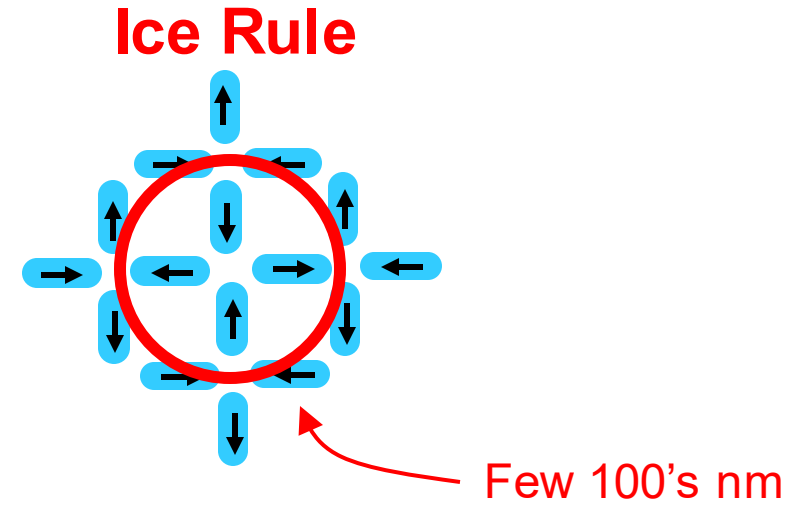
Water Ice

L Pauling  
JACS (1935)



Spin Ice

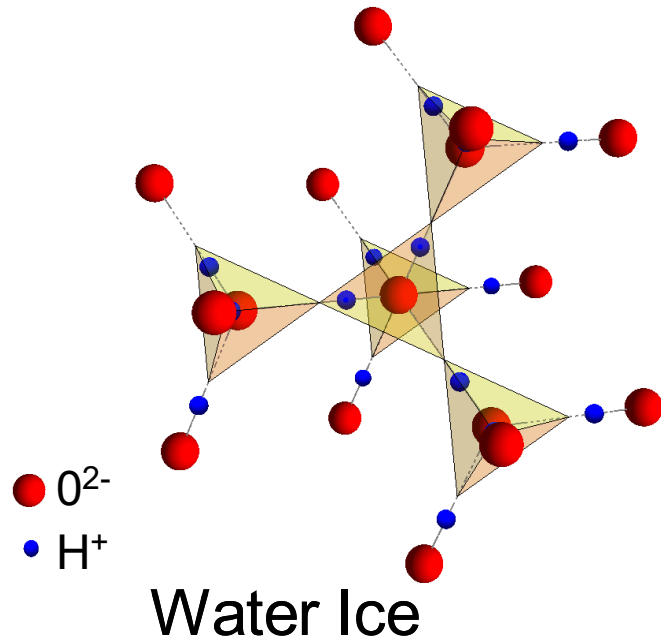
MJ Harris *et al.*  
PRL (1997)



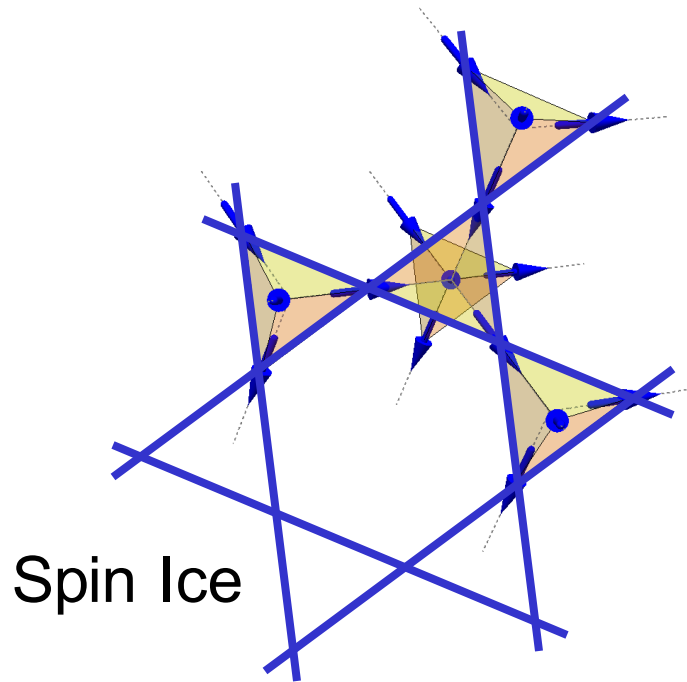
Artificial Spin Ice

RF Wang *et al.*  
Nature (2006)

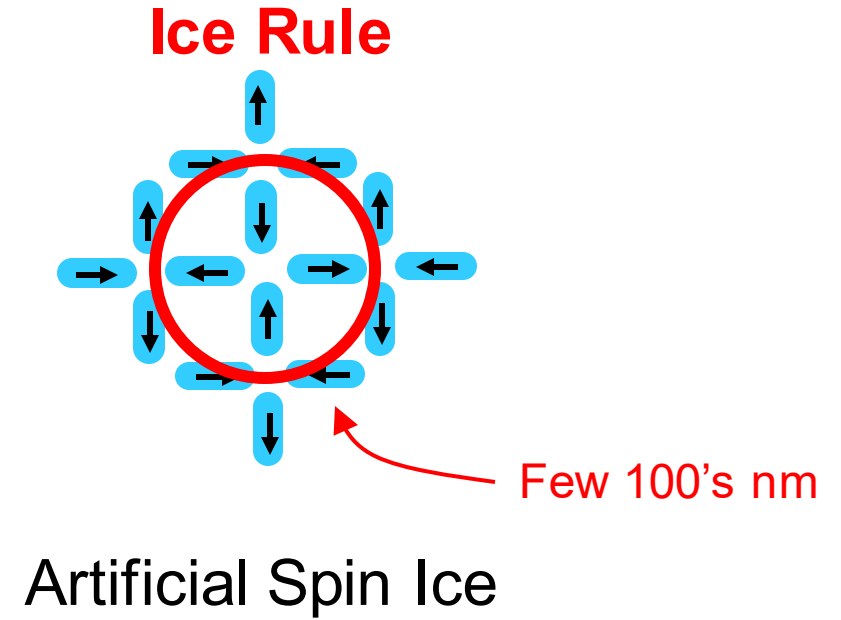
# From Water Ice to Artificial Spin Ice



L Pauling  
JACS (1935)

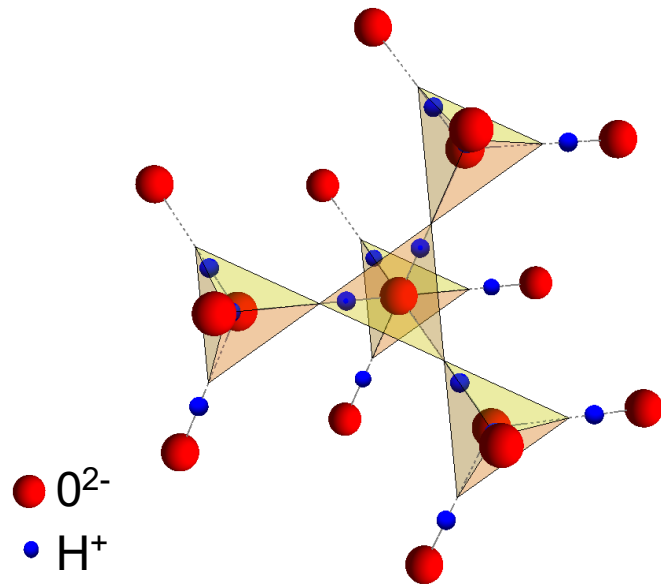


MJ Harris *et al.*  
PRL (1997)



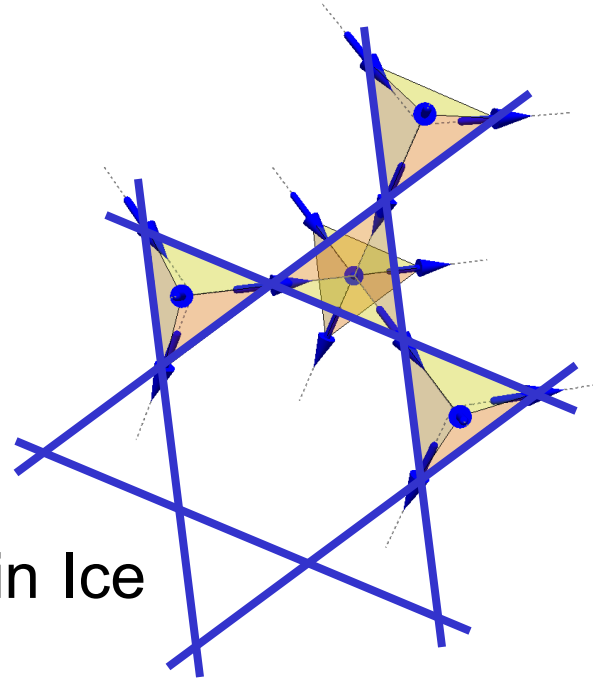
RF Wang *et al.*  
Nature (2006)

# From Water Ice to Artificial Spin Ice



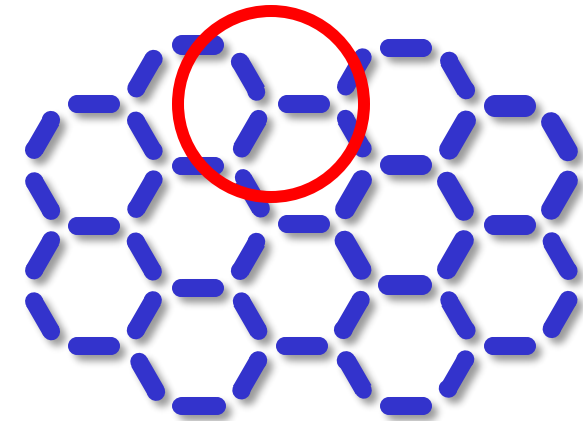
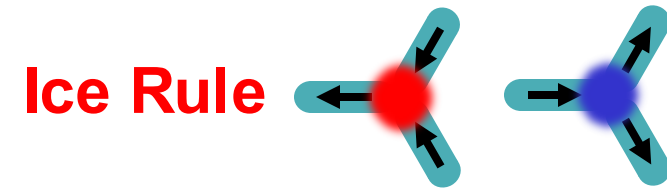
Water Ice

L Pauling  
JACS (1935)



Spin Ice

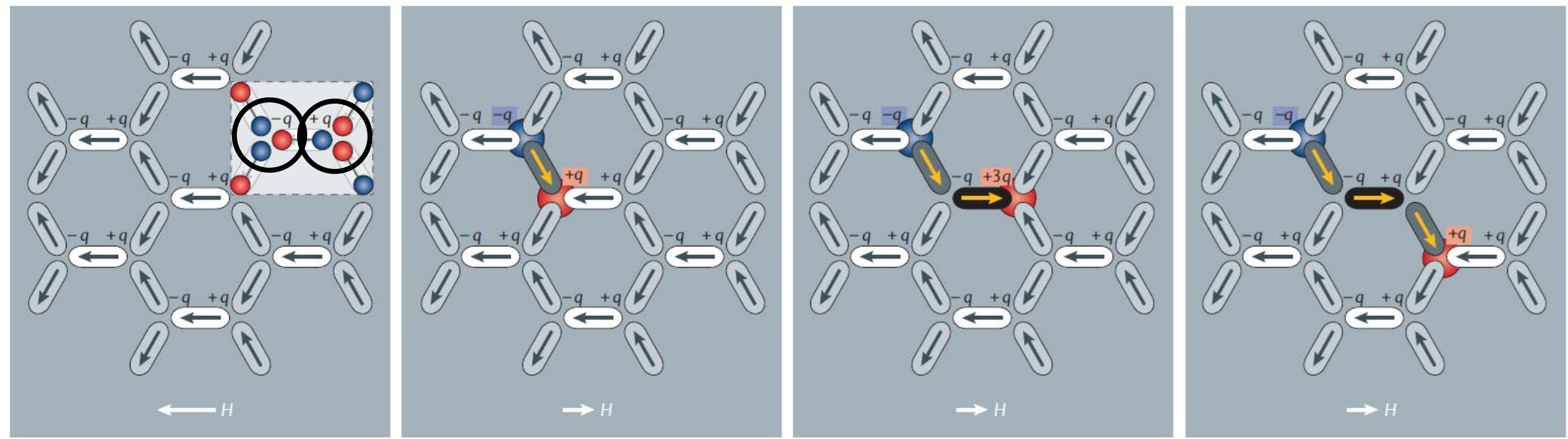
MJ Harris *et al.*  
PRL (1997)



Artificial Kagome  
Spin Ice

AS Wills *et al.*  
Phys. Rev. B (2002)

# Artificial Spin Ice

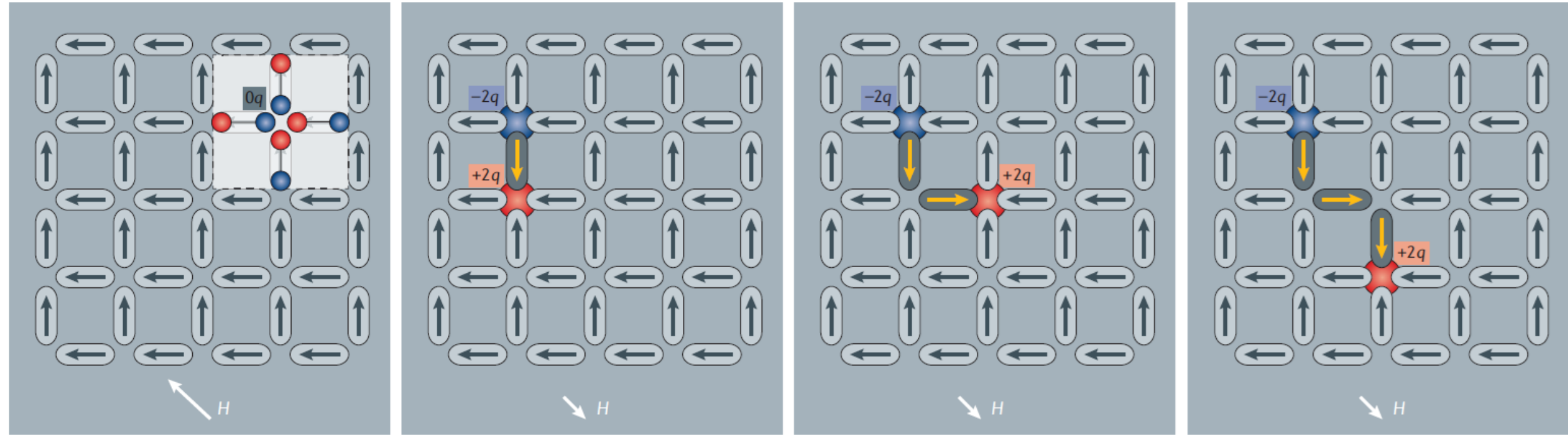


$\Delta Q = \pm 2q$   
 (as long as population is dilute)

LJ Heyderman & RL Stamps J Phys: Condens Matter (2013)

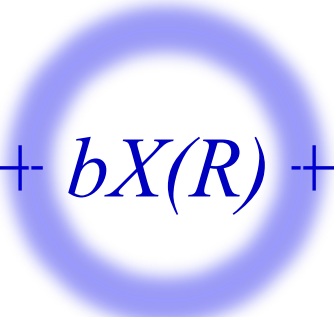
SH Skjærvø, CH Marrows, RL Stamps, LJ Heyderman, Nature Reviews Physics (2020)

# Artificial Spin Ice



$\Delta Q = Q = \pm 2q$

The potential ( $E_{\text{Excited}} - E_{\text{GS}}$ ):  $V(R) = q/R + bX(R) + c$

Coulombic-type term            monopole pair creation

stringlike excitations that bind the monopoles

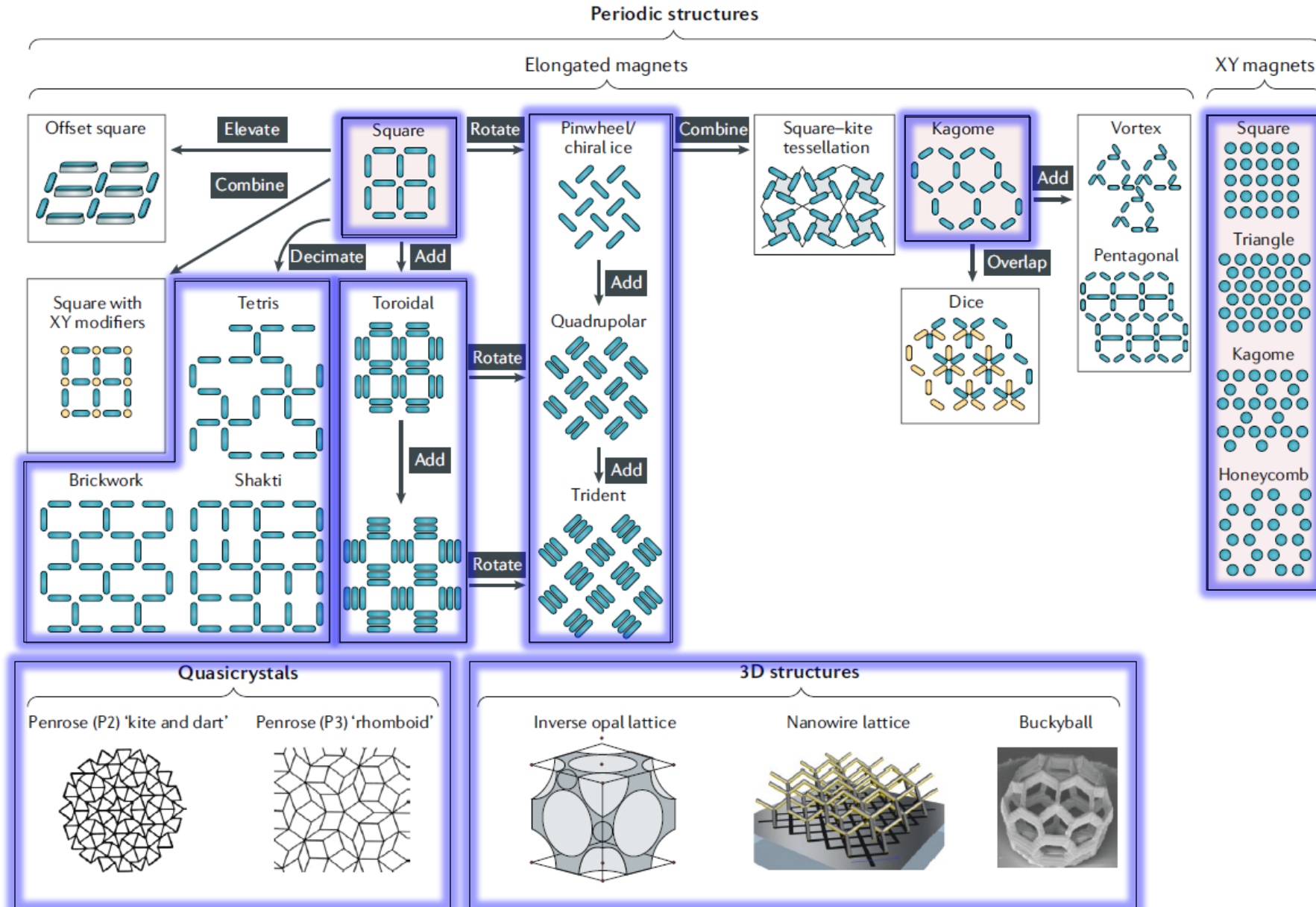
LA Mól et al. JAP (2009)

LJ Heyderman & RL Stamps J Phys: Condens Matter (2013)

SH Skjærvø, CH Marrows, RL Stamps, LJ Heyderman, Nature Reviews Physics (2020)



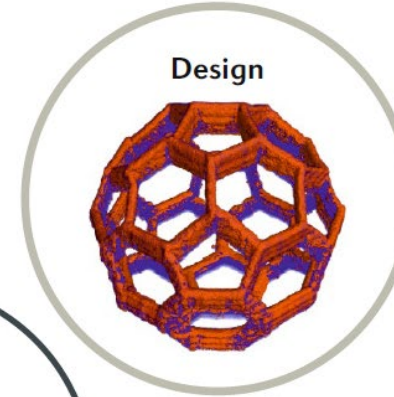
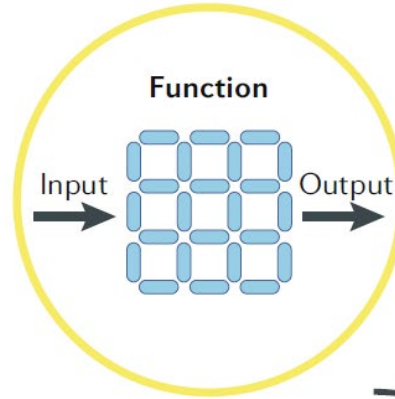
# Artificial Spin Ice Geometries



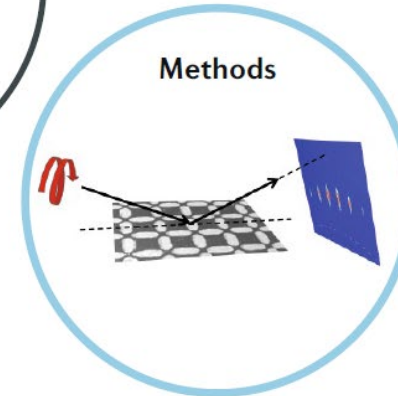
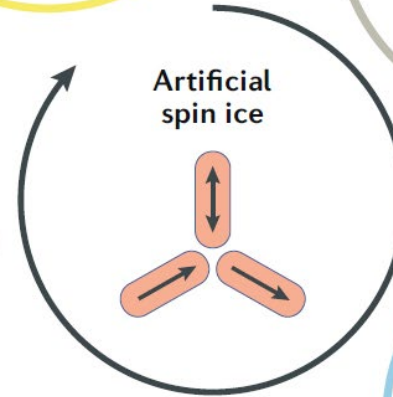
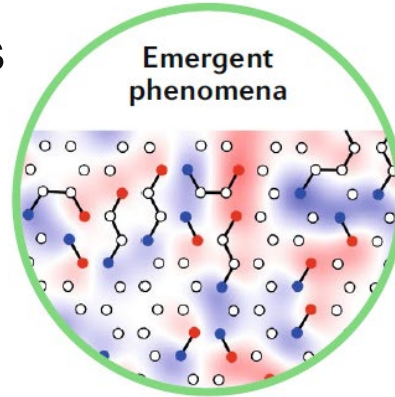
# Artificial Spin Ice Activities

- Computation
- Magnonics
- Hybrid systems

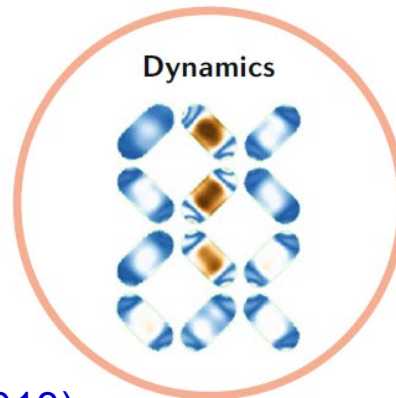
- Geometries
- Materials



- Magnetic monopoles
- Phases transitions
- Vertex frustration
- Chirality



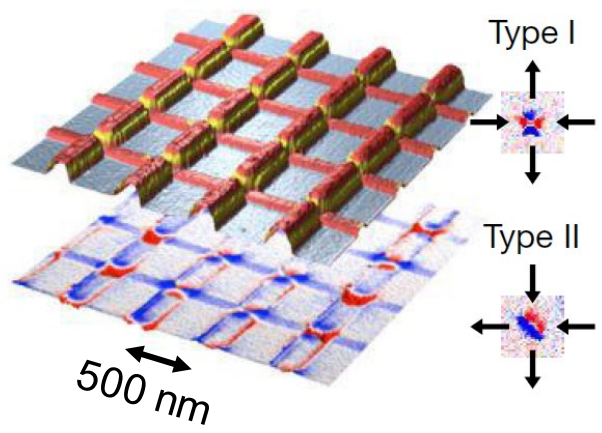
- Driven processes
- Thermal relaxation



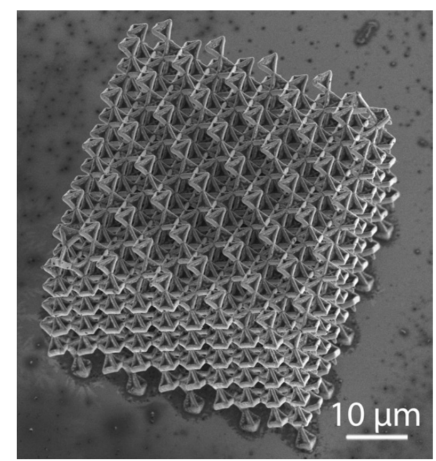
- Fabrication
- Characterization
- Manipulation
- Theory & Simulations



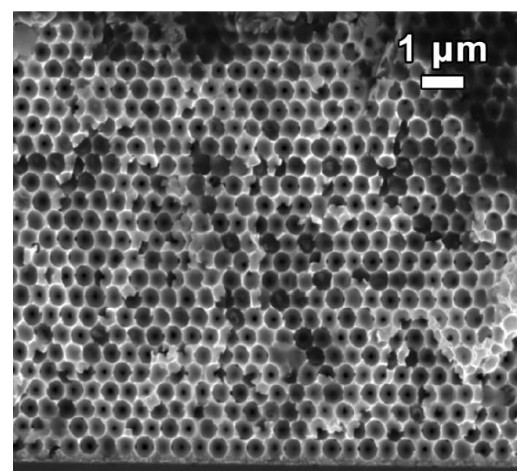
# 3D Artificial Spin Ice



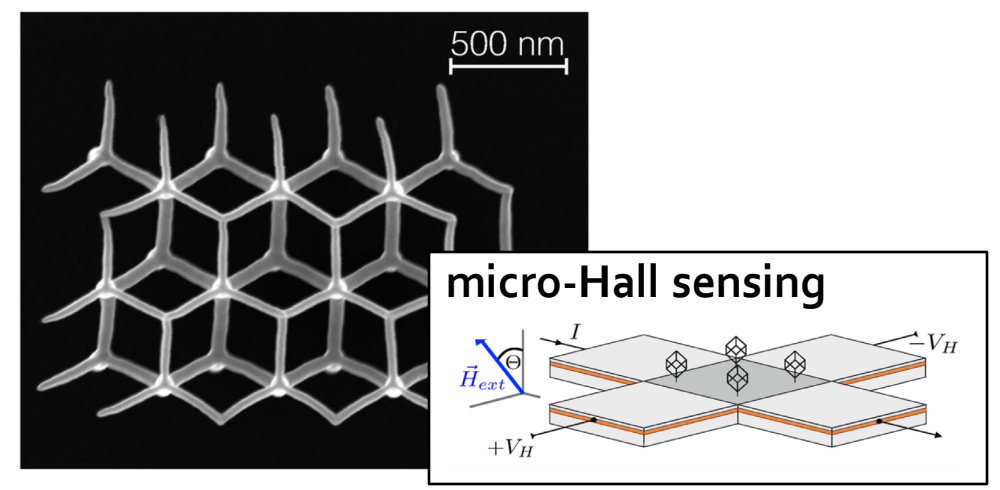
**Electron Beam Lithography**  
Y Perrin et al. Nature 2016



**Two Photon Laser Lithography**  
P Pip et al. Small 2020



**Self-Assembly Methods**  
**Inverse Opal-Like Structures (IOLS) & Gyroids**  
AA Mistonov et al. JMMM 2019

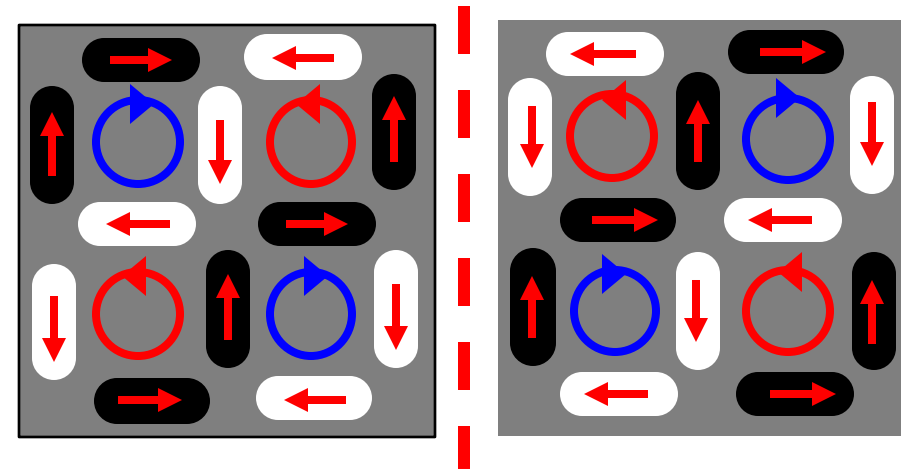
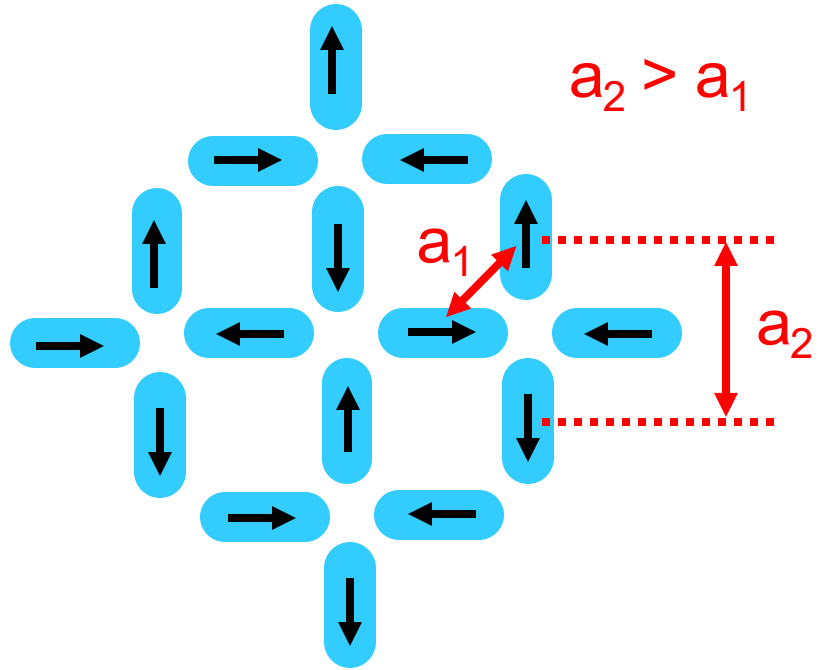


**Focused electron beam induced deposition**  
L Keller et al. Sci Rep 2018  
C Schröder et al Cond Mat ArXiv 2024



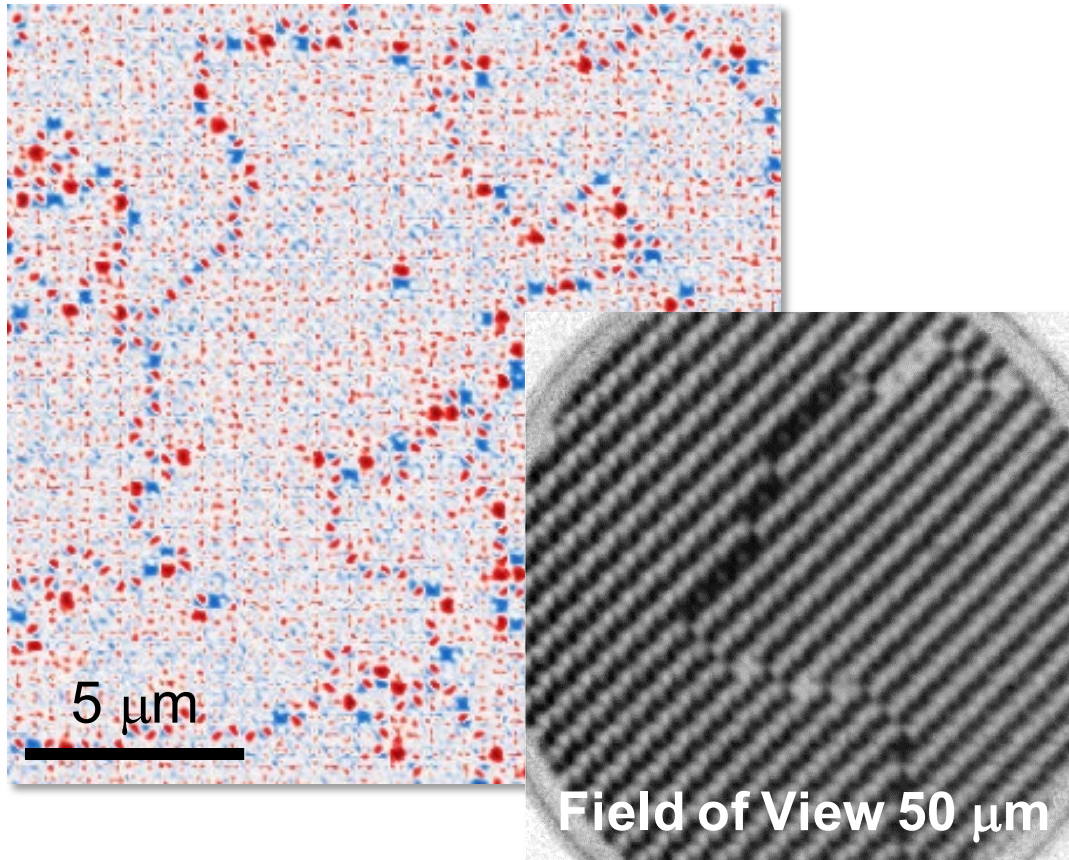
- Layered Structures
- 2.5 D

Elena Mengotti working at Vistec EPBG Electron Beam Writer

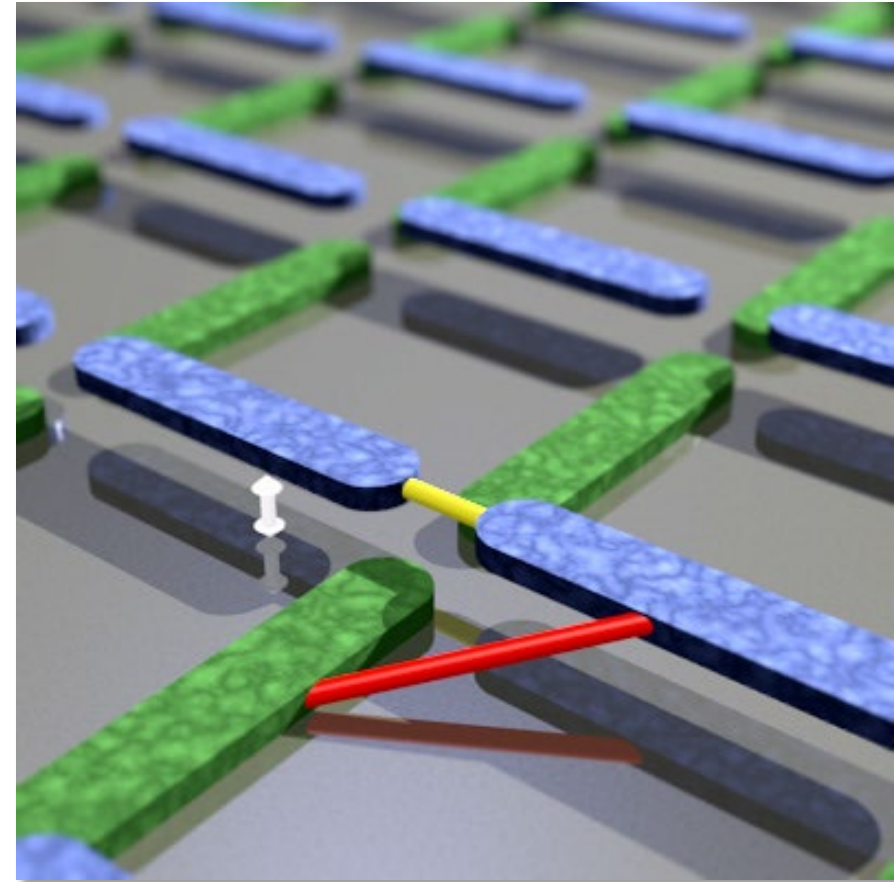




# Raising a Sub-Lattice

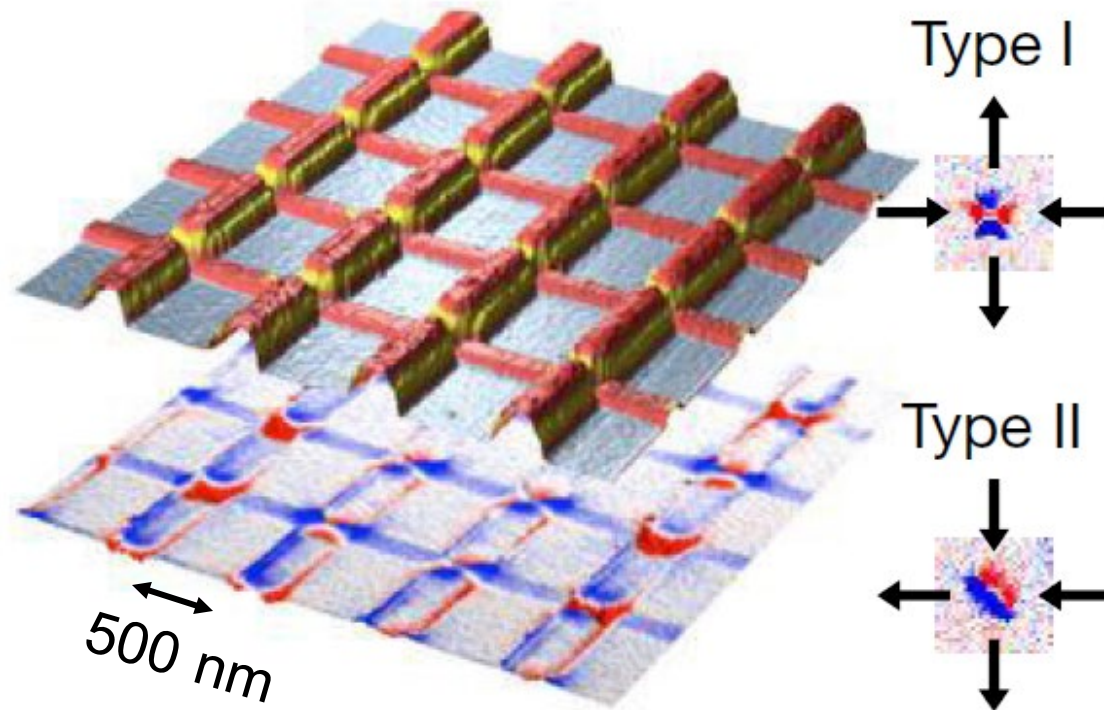


J Morgan et al. Nature Physics (2011)  
 JM Porro et al. NJP (2013)  
 S Zhang et al. Nature (2013)  
 A Farhan et al. PRL (2013)



G Moller & R Moessner PRL (2006)  
 G-W Chern et al. APL (2014)  
 Y Perrin et al. Nature (2016)  
 A Farhan et al. Science Advances (2019)

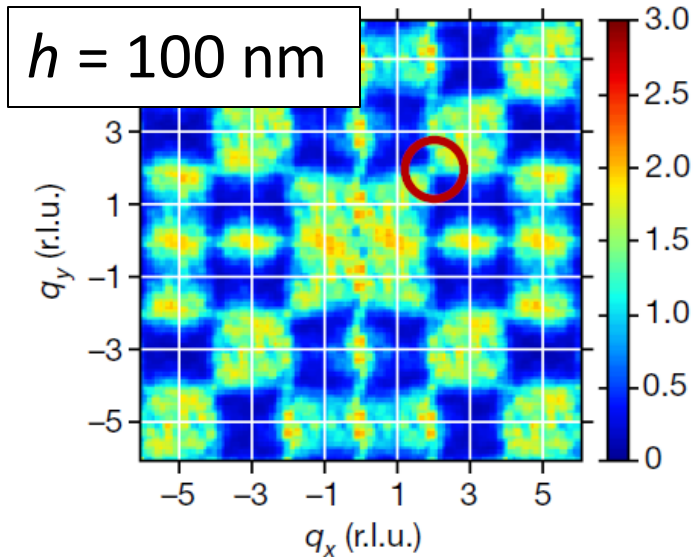
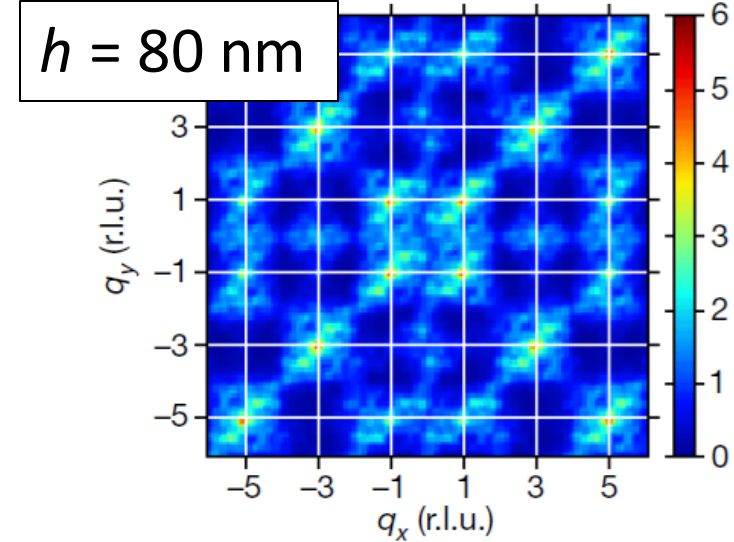
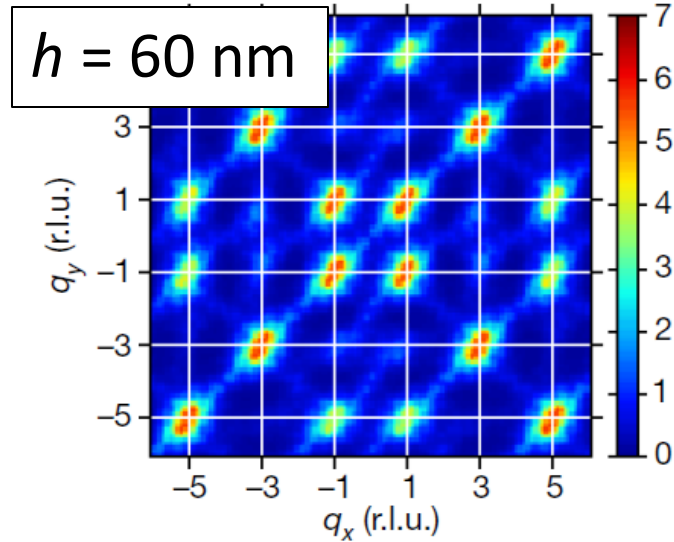
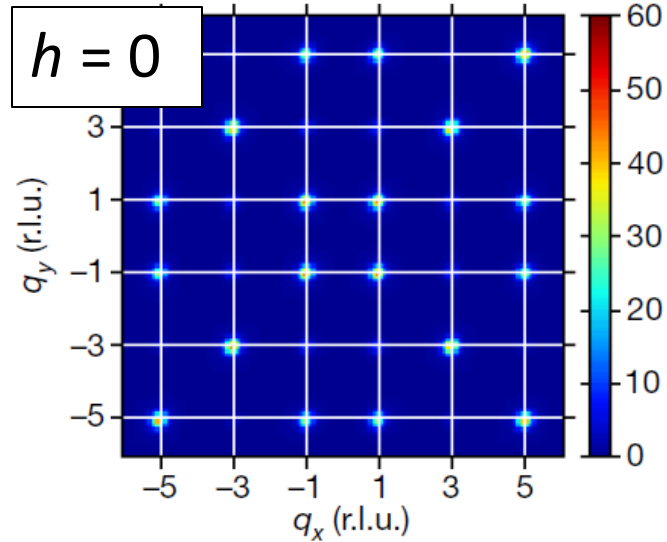
## Raising a Sub-Lattice



Height offset:  
titanium bases with  
gold capping layer

Y Perrin et al. Nature (2016)  
A Farhan et al. Science Advances (2019)

# Magnetic Structure Factors

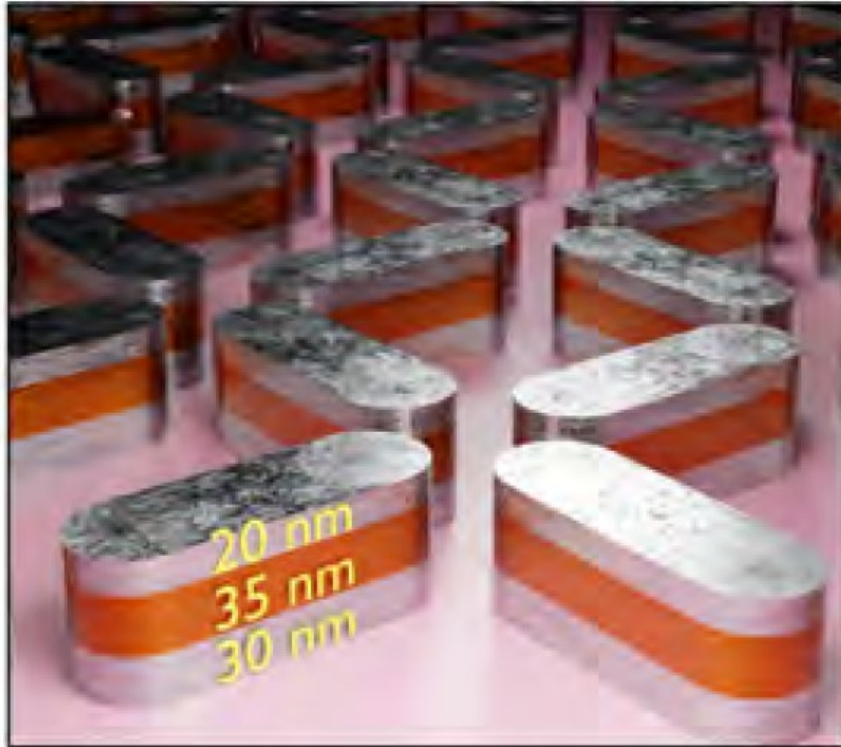


“...finite density of classical monopoles within a Coulomb phase.”  
 Y Perrin et al. Nature (2016)

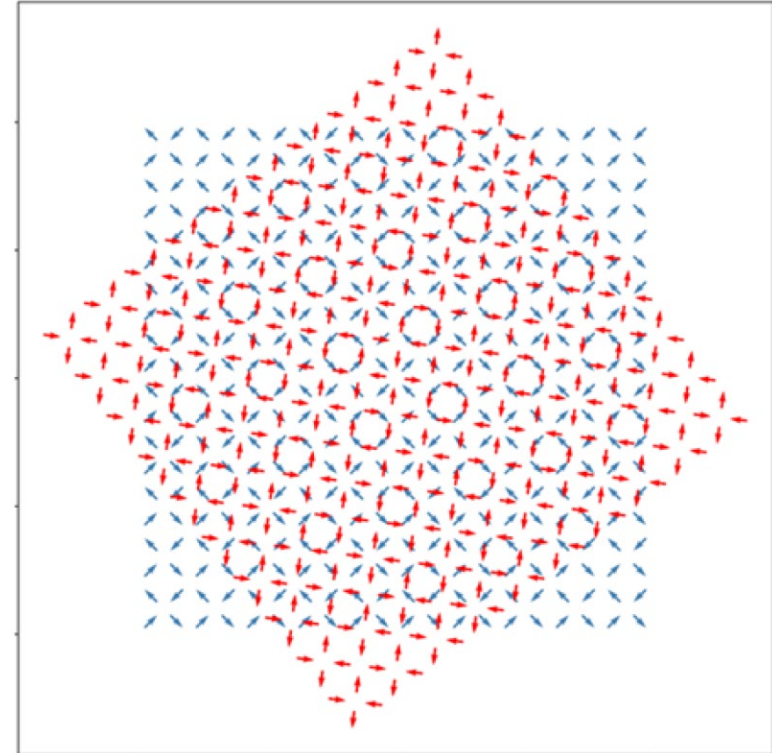
“Debye-Hückel theory - topological defects act like a plasma of Coulomb-type magnetic charges”  
 A Farhan et al. Science Advances (2019)

→ Christopher L. Henley, Annu. Rev. Condens. Matter Phys. 2010  
 The “Coulomb phase”: local constraints that can be mapped to a divergence-free “flux.” Defects at which the local constraint is violated behave as effective charges with Coulomb interactions.



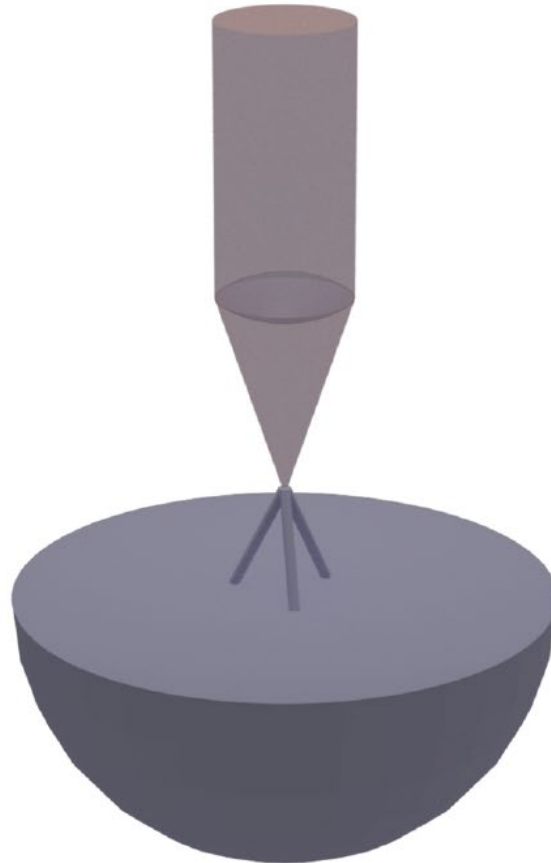


Ultrastrong Magnon-Magnon Coupling  
and Chiral Symmetry Breaking  
T Dion et al. Cond Mat ArXiv (2023)  
Nanomagnet length: 550 nm

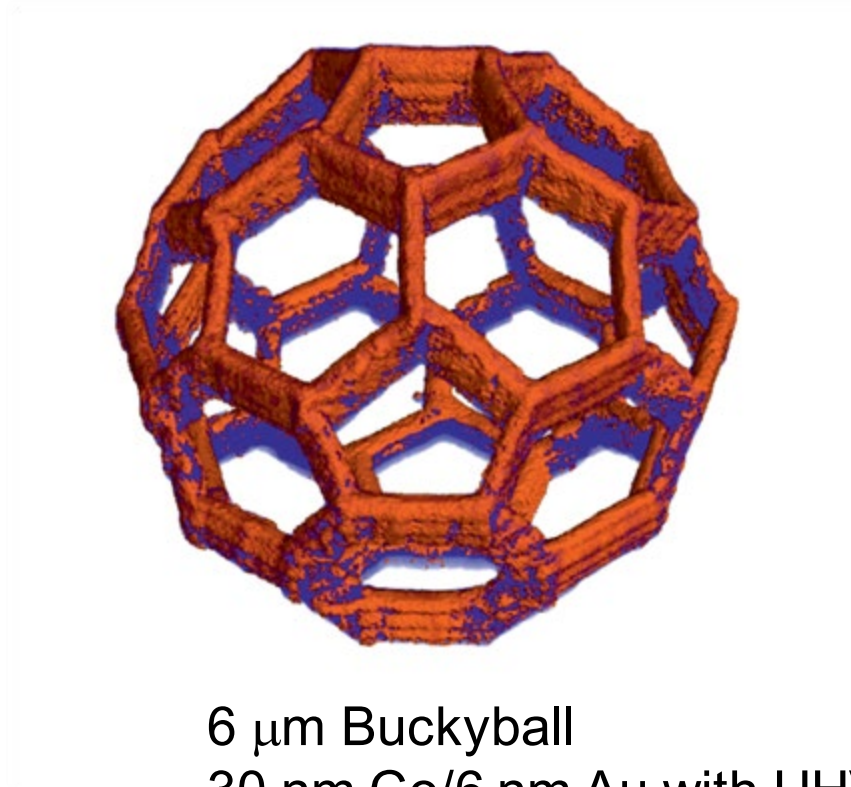


Numerical simulations of twisted  
bilayer artificial spin ice  
RB Popy et al. JAP 2022

*Fabrication of  
polymeric microscaffold*



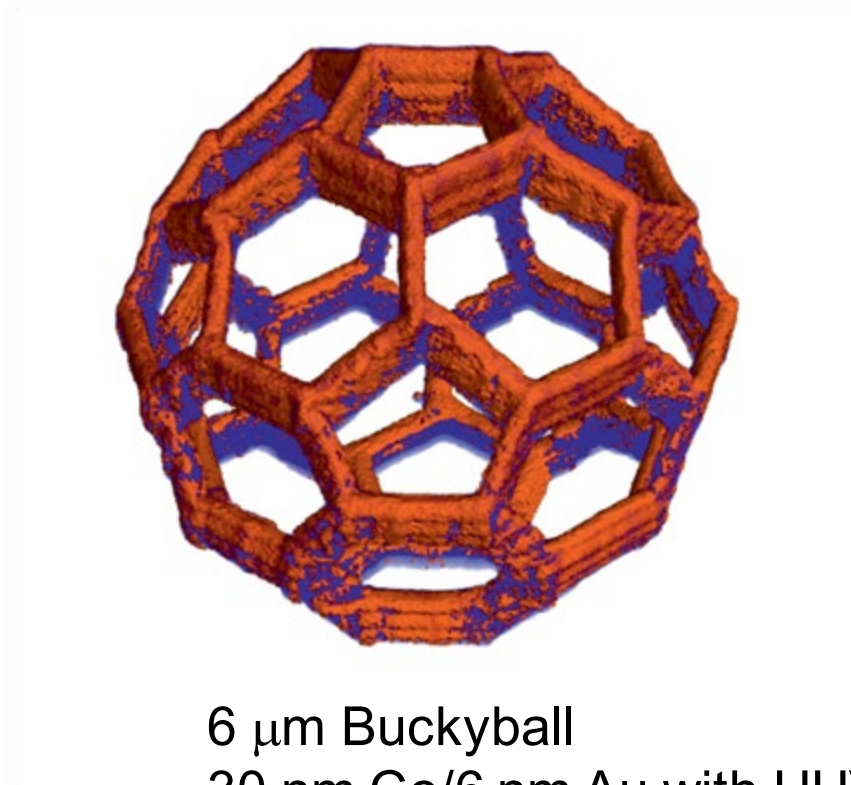
- Nanoscribe Photonic Professional GT 2 Laser Direct Writing System
- Infra-red laser with  $\lambda = 780 \text{ nm}$
- UV sensitive photoresist
- Laser is focused by an optical lens
- At focal point: two-photon absorption ( $\lambda_{\text{eff}} = 390 \text{ nm}$ ) and polymerisation
- Development in propylene glycol methyl ether acetate (PGMEA)



6  $\mu\text{m}$  Buckyball  
30 nm Co/6 nm Au with UHV sputtering

### **Resonant Ptychographic Tomography**

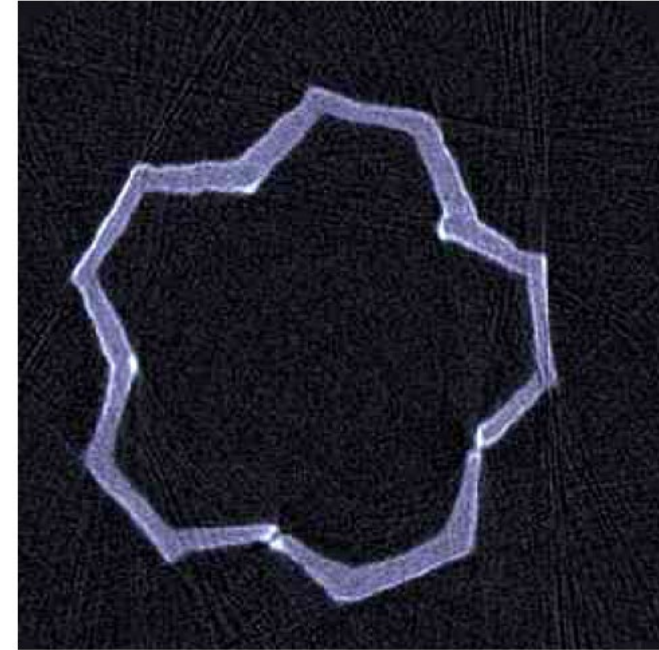
*Quantitative hard x-ray phase imaging & resonant elastic scattering  
→ element-specific 3D characterization with 25 nm spatial resolution*



6  $\mu\text{m}$  Buckyball  
30 nm Co/6 nm Au with UHV sputtering

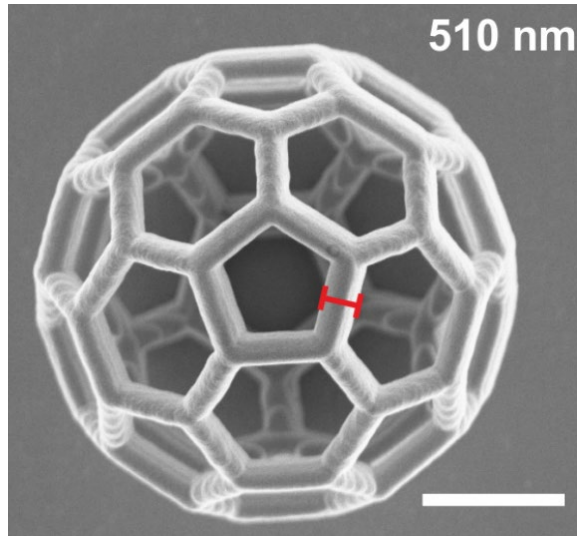
### Resonant Ptychographic Tomography

*Quantitative hard x-ray phase imaging & resonant elastic scattering  
→ element-specific 3D characterization with 25 nm spatial resolution*



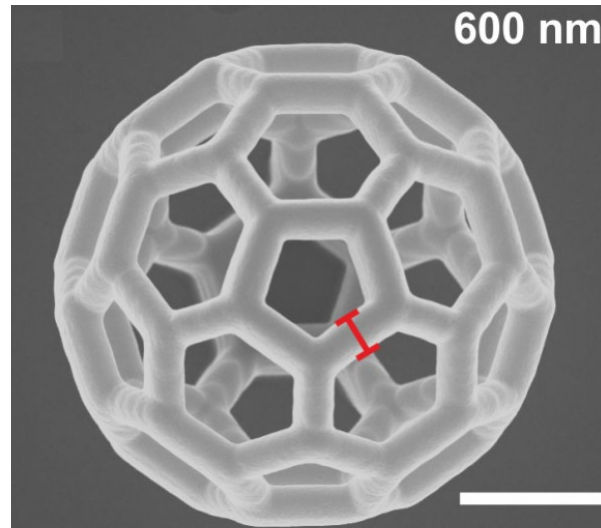


## 10 nm Al<sub>2</sub>O<sub>3</sub>/10nm Ir grown conformally with ALD

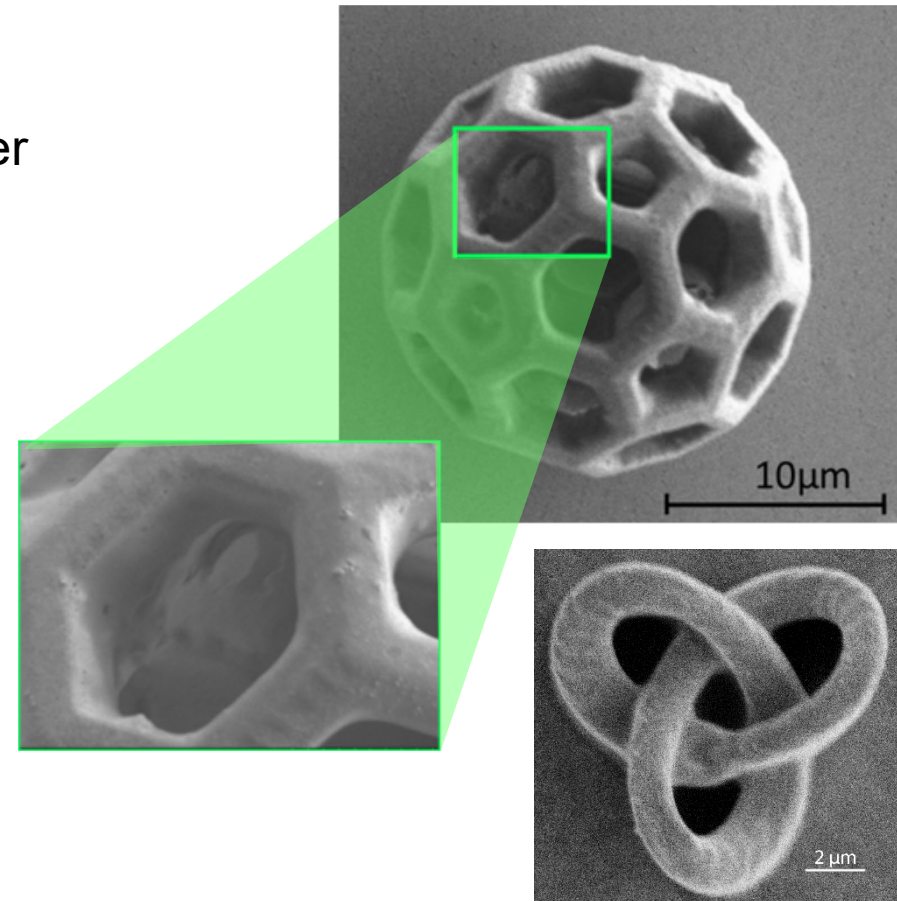


**Ir:** conductive layer  
**Al<sub>2</sub>O<sub>3</sub>:** prevents cracking of Ir film due to expansion of polymer  
**Scale bars:** 2 μm

**45 nm-thick electrodeposited Ni film**

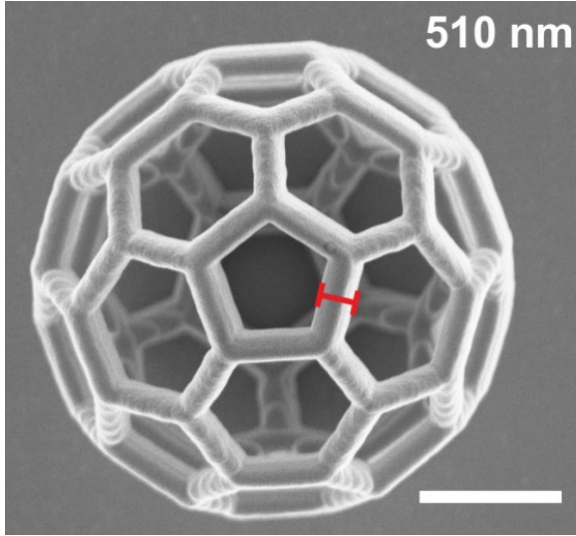


## Electroless Deposition of NiFe(B)



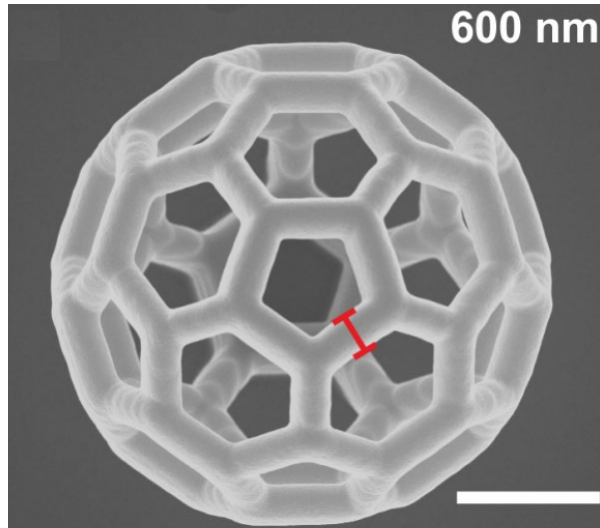
cf. Petai Pip & Laetitia Philippe, EMPA Thun  
 P Pip PhD Thesis ETH Zurich 2021  
 P Pip et al. Small (2020)

## 10 nm Al<sub>2</sub>O<sub>3</sub>/10nm Ir grown conformally with ALD

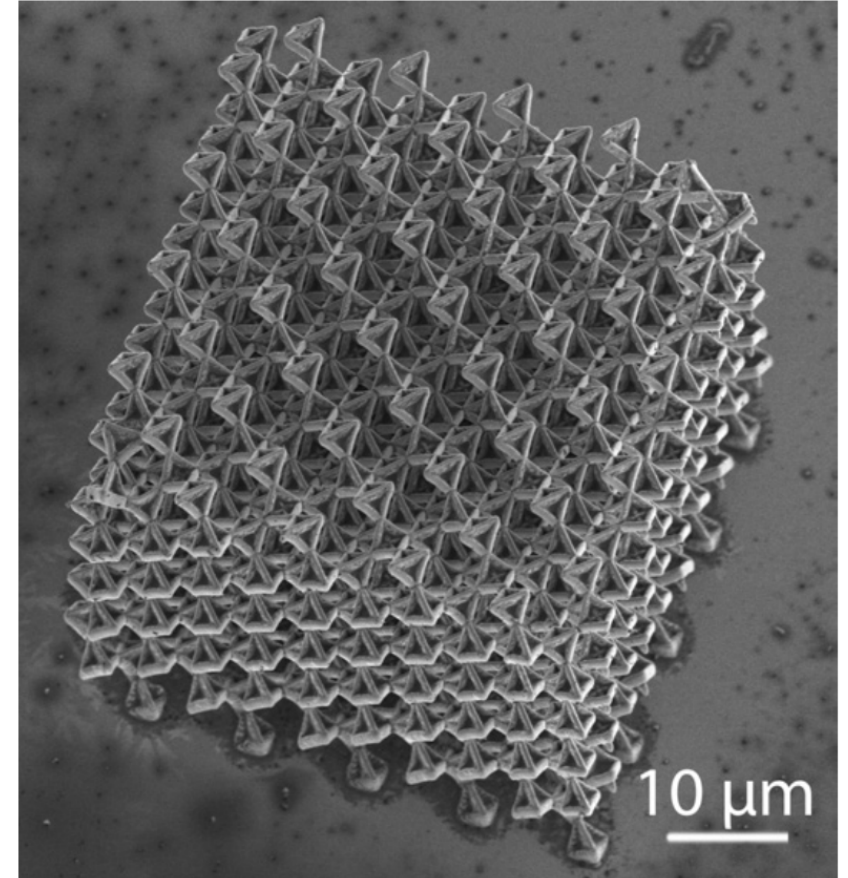


**Ir:** conductive layer  
**Al<sub>2</sub>O<sub>3</sub>:** prevents cracking of Ir film due to expansion of polymer  
**Scale bars:** 2 μm

**45 nm-thick electrodeposited Ni film**

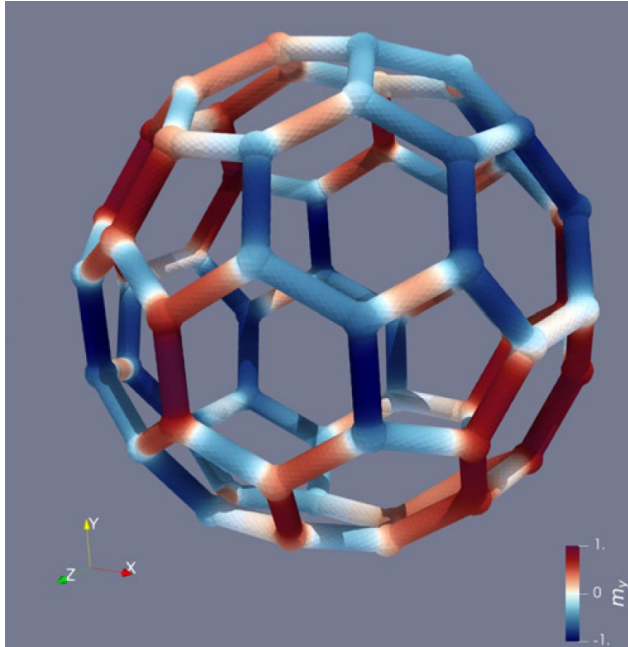


## Electroless Deposition of NiFe(B)

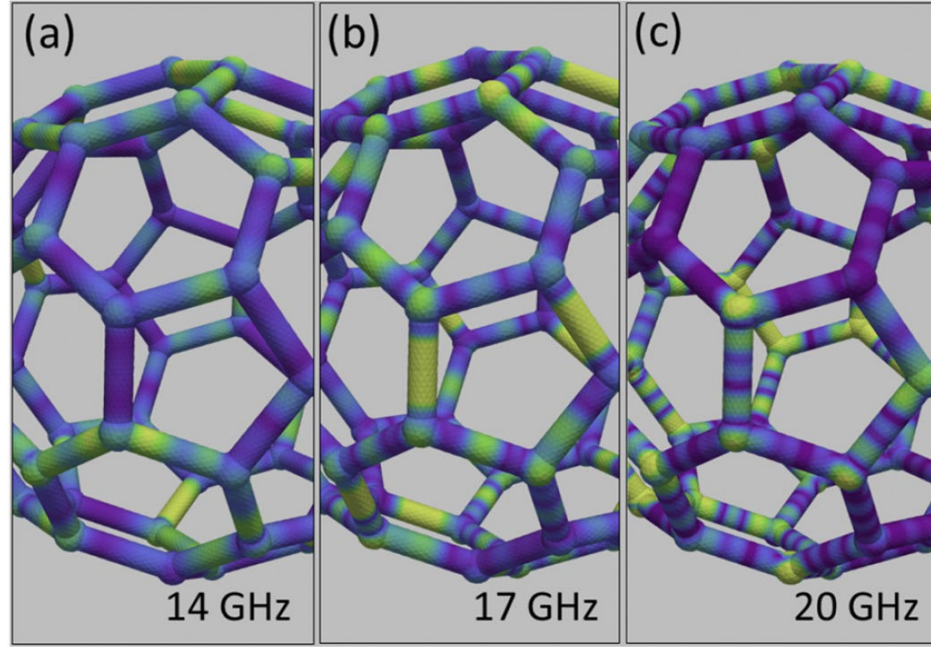


cf. Petai Pip & Laetitia Philippe, EMPA Thun  
 P Pip PhD Thesis ETH Zurich 2021  
 P Pip et al. Small (2020)

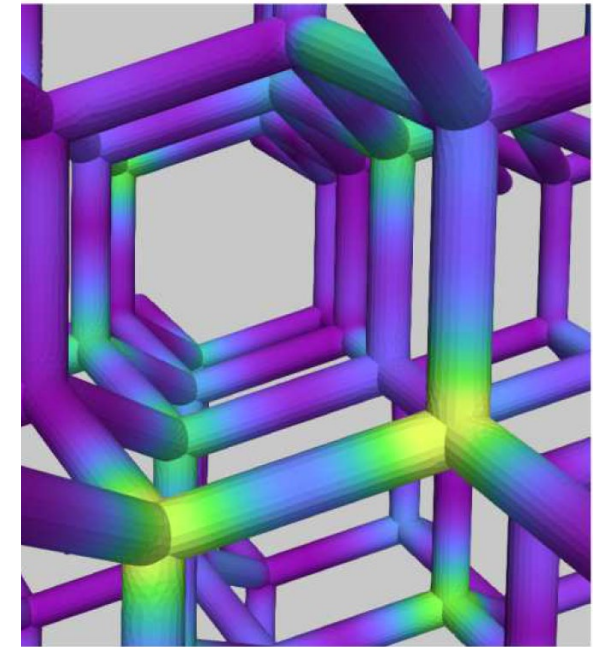




**Switching in a buckyball**  
 R Cheenikundil et al. APL (2021)  
 Nanowire Length 100 nm

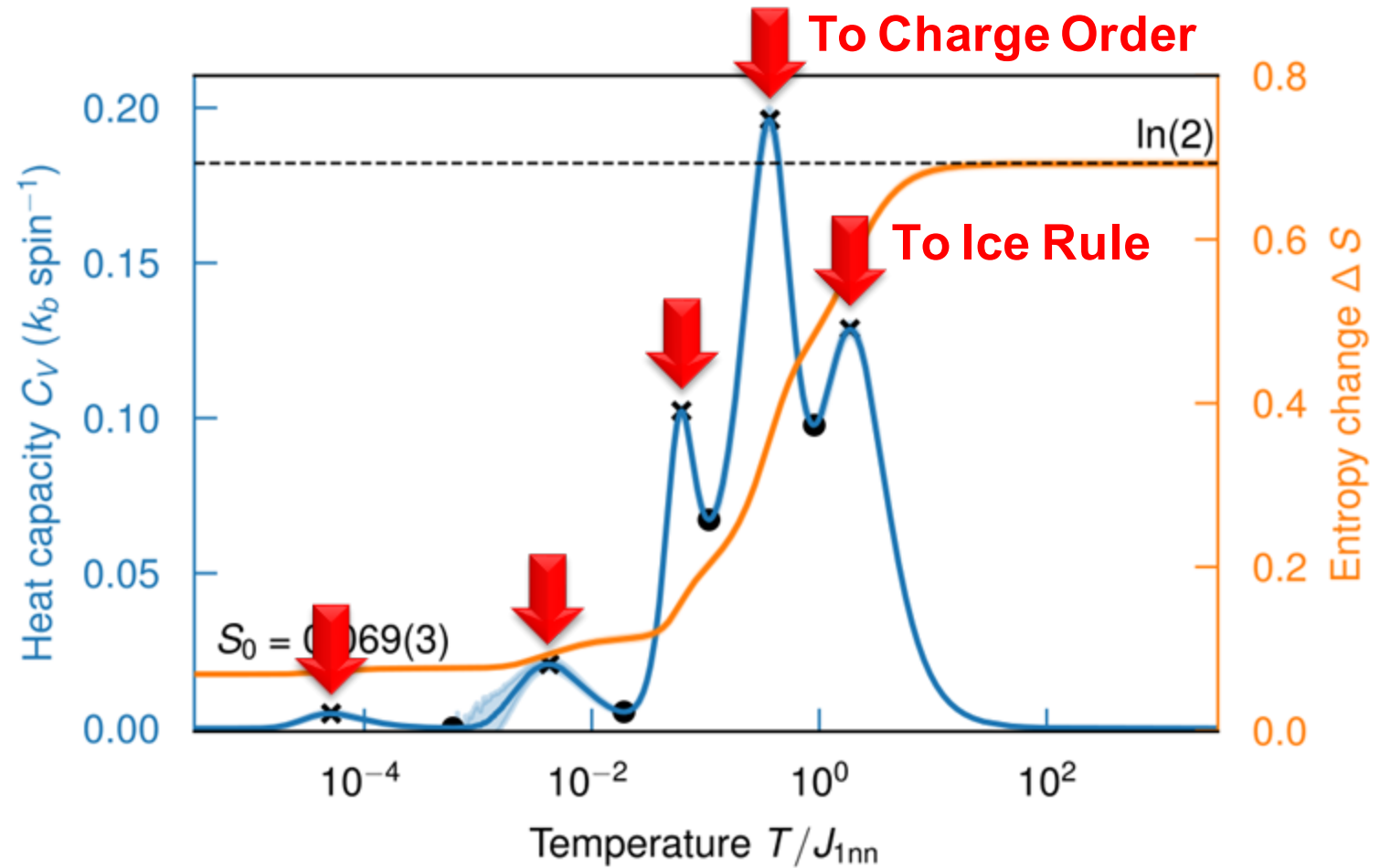
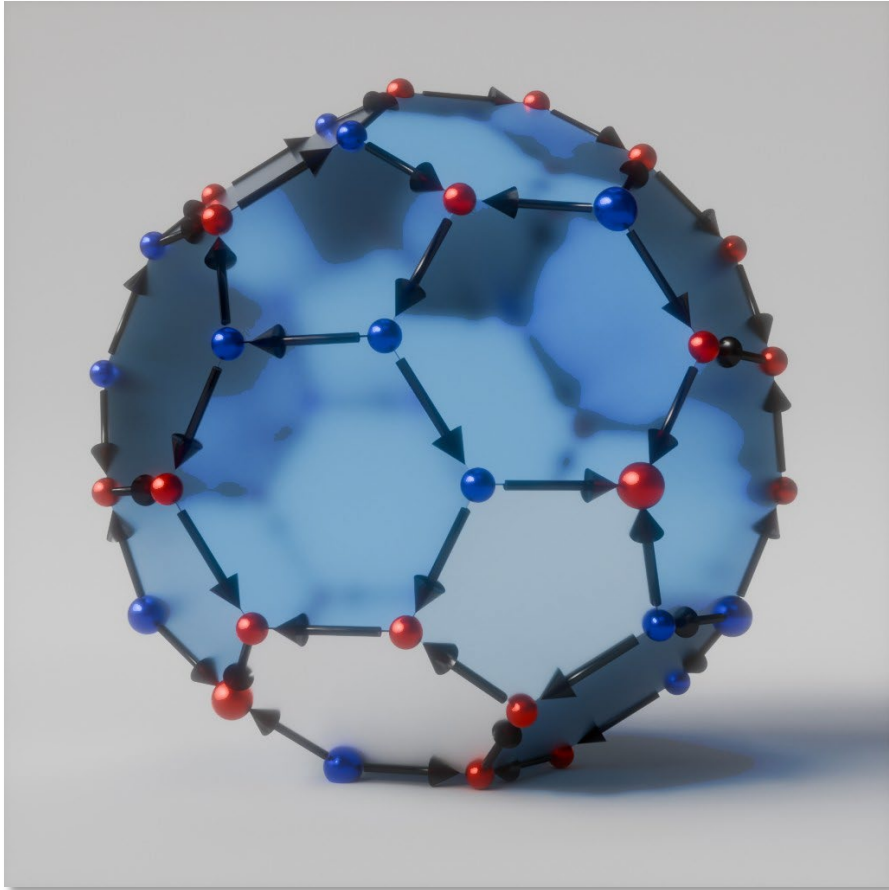


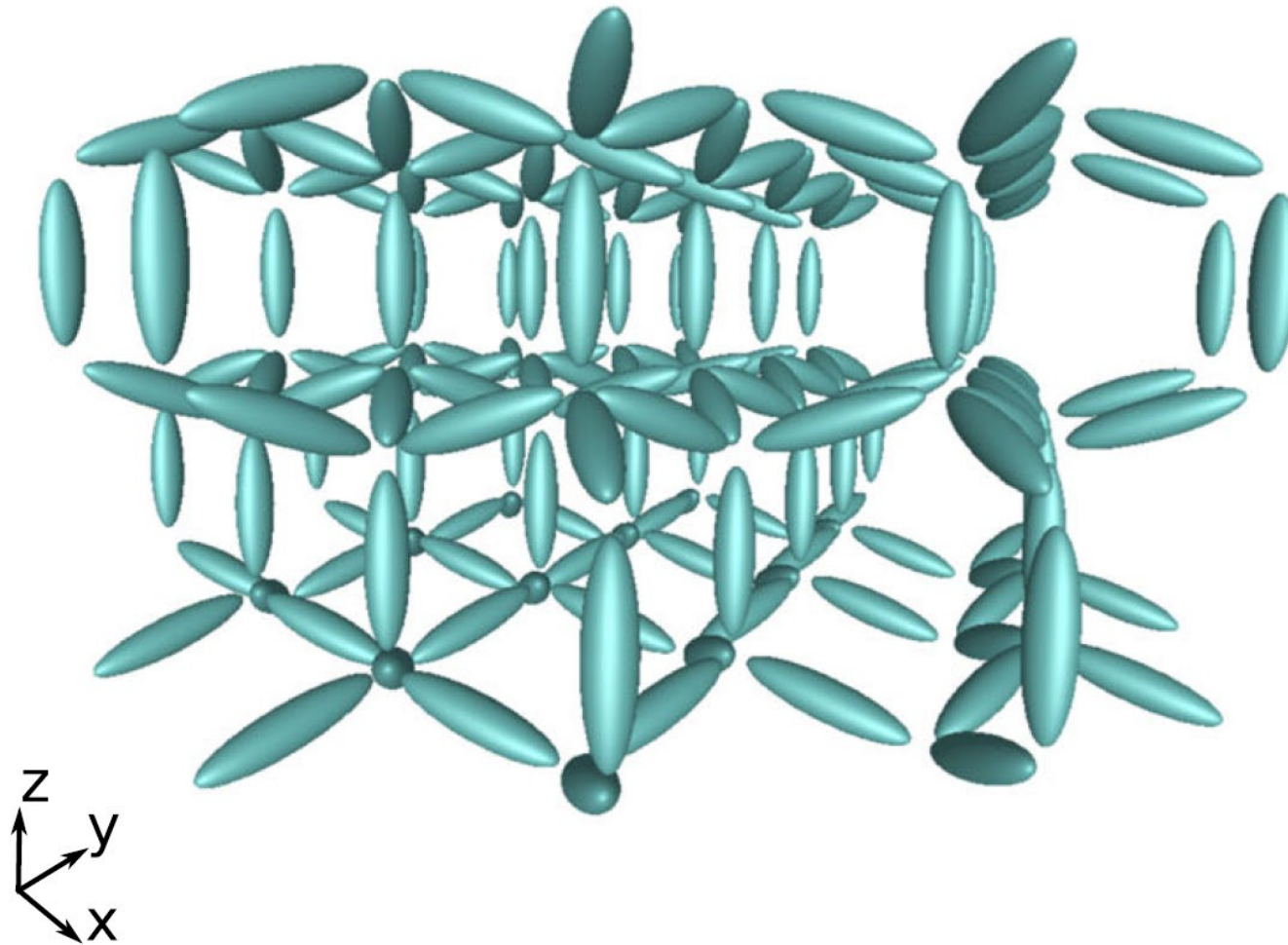
**High-frequency modes in a buckyball**  
 R Cheenikundil et al. APL Materials (2022)  
 Nanowire Length 100 nm



**Defect-sensitive High-freq. modes in 3D ASI**  
 R Hertel et al. hal-03850245 (2022)  
 Nanowire Length 70 nm

# Thermodynamics in a Buckyball



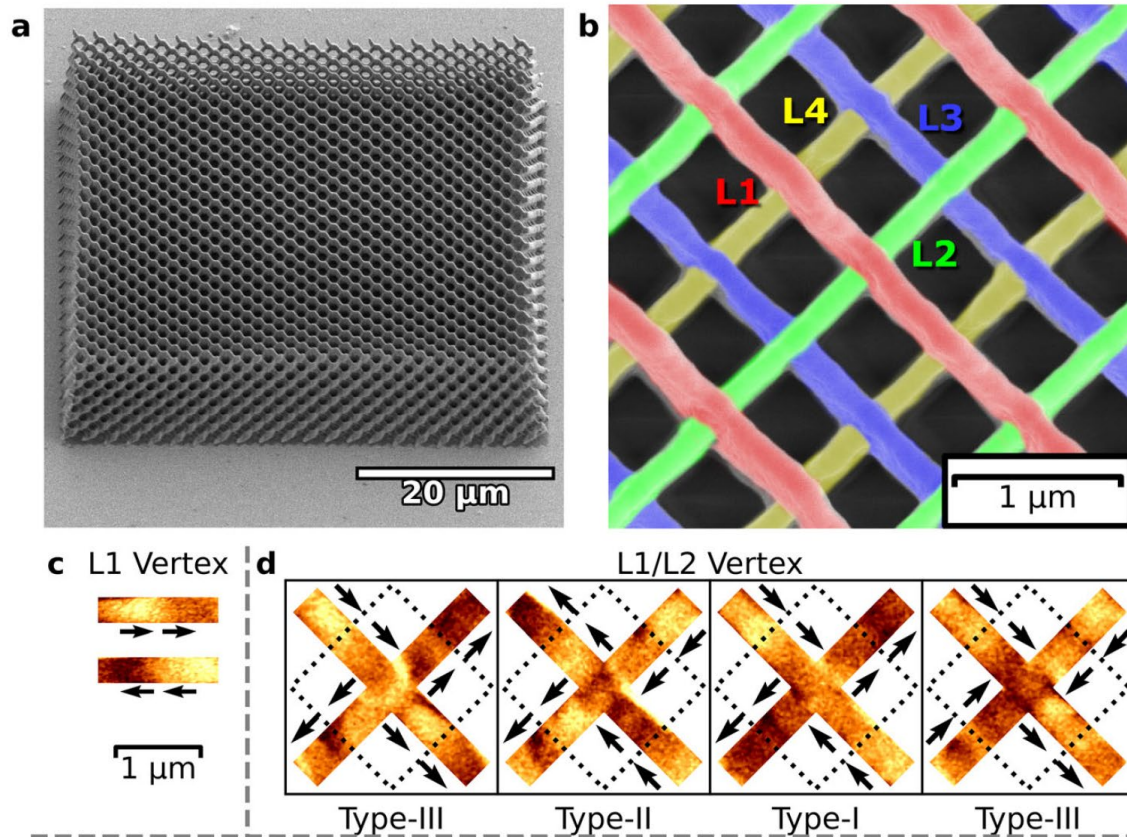


Tension-free Dirac strings and steered magnetic charges  
S Koraltan et al. NPJ Comp Matls 2021  
Ellipsoid Length 100 nm



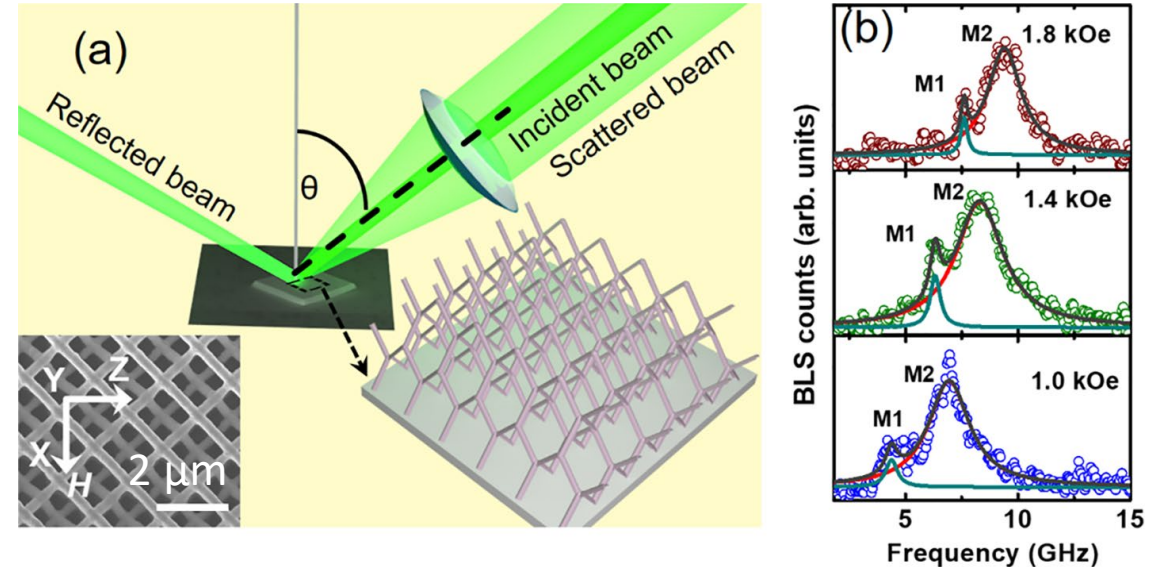
# Two Photon Laser Lithography

Magnetic Force Microscopy (MFM)



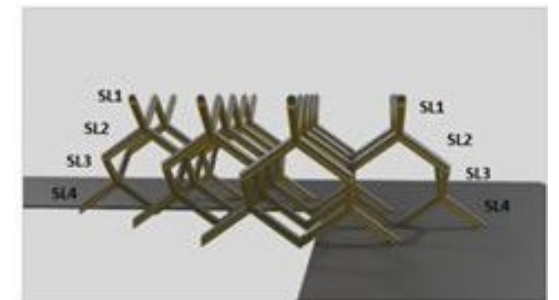
A May et al. Realisation Comm Phys 2019  
 A May et al. Charge Propagation Nat. Comm. 2021  
 M Saccone et al. ASI Phases Commun. Phys. 2023

Brillouin Light Scattering (BLS) Spectroscopy

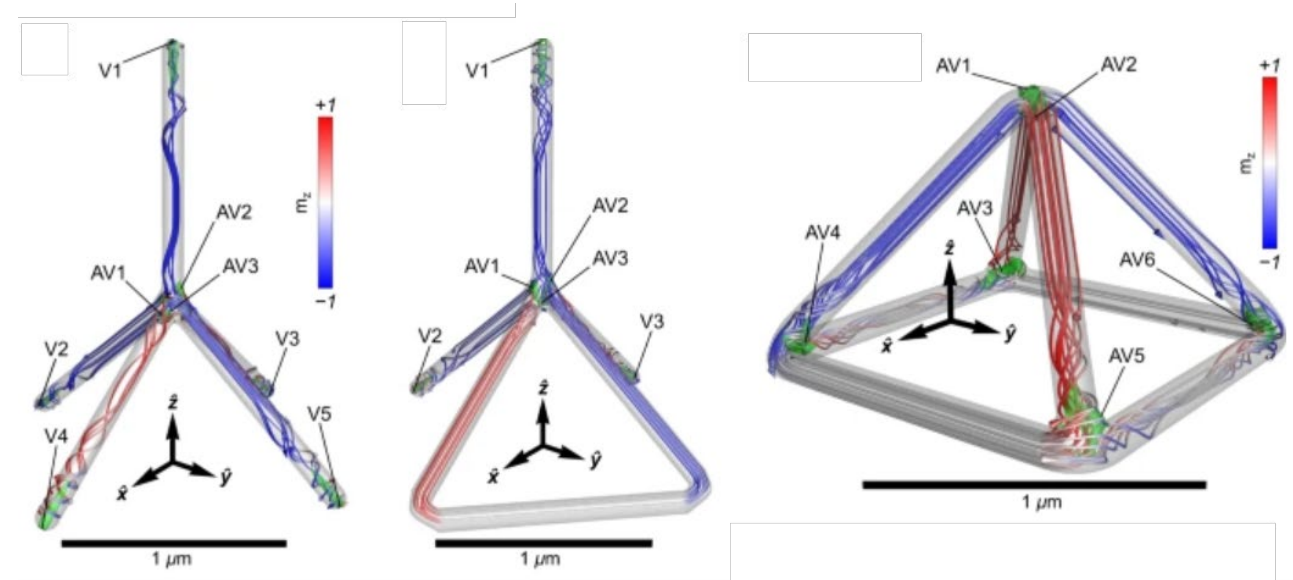
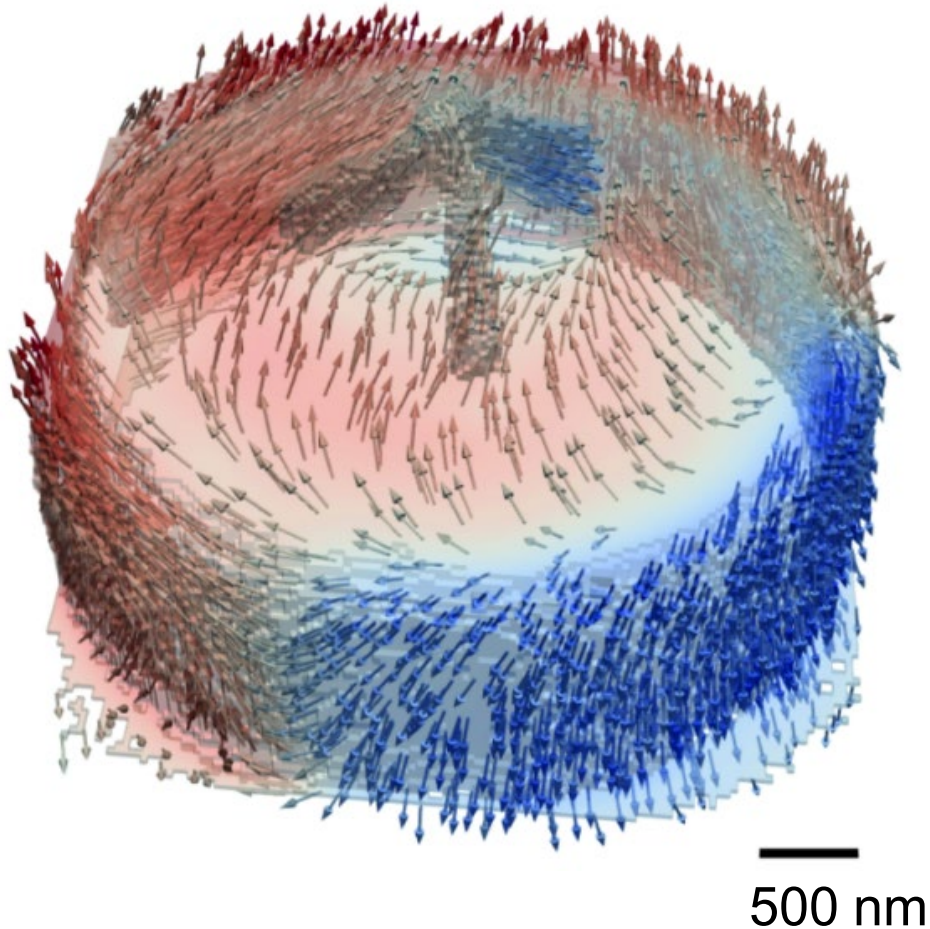


S Sahoo et al. Coherent Spin Waves Nano Lett 2021  
 S Sahoo et al. Ultrafast dynamics in a tetrapod with time-resolved MOKE microscopy Nanoscale 2018  
 H Guo et al. Bulk & surface modes in a woodpile structure Adv Mater 2023

E Harding et al. APL Mater. 2024  
 Nanowire Length 1 μm



# Artificial Spin Ice Building Block



**Topological magnetization textures with high-order vorticity in wireframes & FEBID**

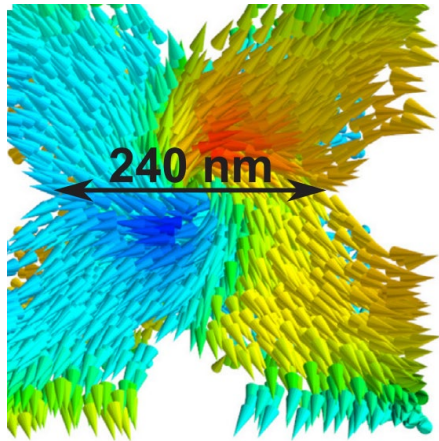
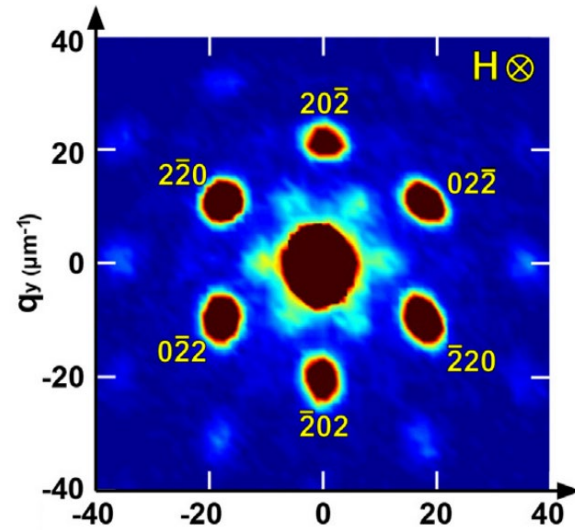
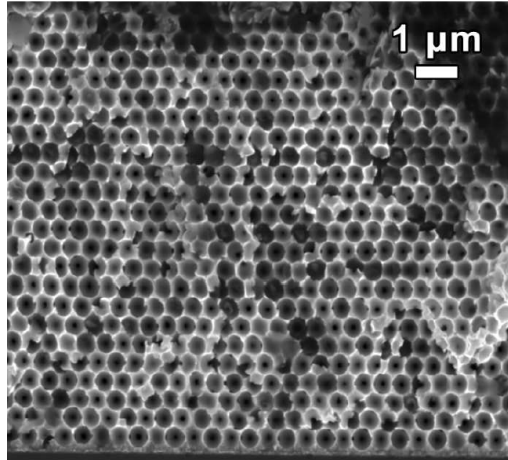
OM Volkov et al. Nature Commun 2024  
 Scalebar: 1  $\mu\text{m}$

**Soft x-ray magnetic laminography**

P Pip et al. APL Materials (2022)

In-plane reconstruction spatial resolution of  $< 100\text{nm}$

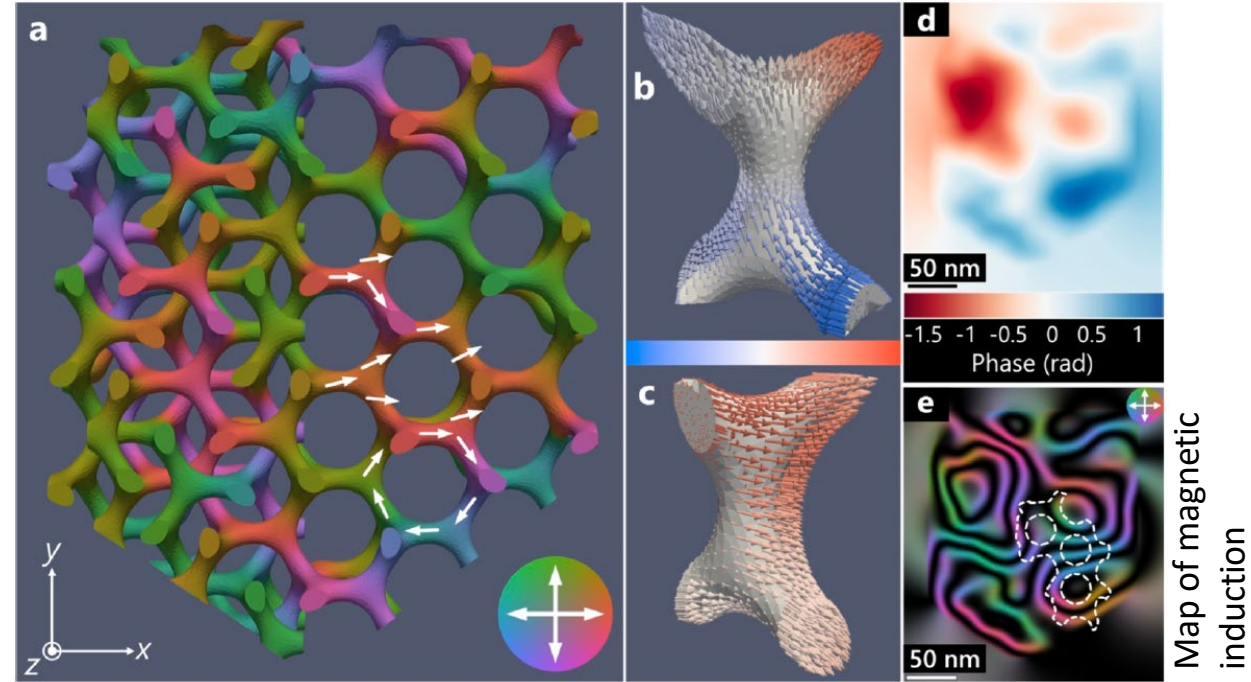




- (i) Artificial opal:  
Electric-field-assisted assembly of polystyrene microspheres  
 $D = 620 \text{ nm}$ ;  $RSD < 10\%$
- (ii) Si(100) substrate with 200 nm thick Au:  
Co Electrodeposition in voids

## Inverse Opal-Like Structures (IOLS)

IS Dubitskiy et al. Micromagnetic Simulations JMMM 2017  
 AA Mistonov et al. SANS & Micromagnetics JMMM 2019  
 AA Mistonov et al. 3D ASI PRB 2013  
 IR Shishkin et al. Geometric scaling of coercivity PRB 2016



- (i) Thermal annealing of a block copolymer template
  - (ii) Selective dissolution of the minority block
  - (iii) Electrodeposition
- NiFe double-gyroids: 42 nm unit cell & chiral triple junctions

## Gyroids

J Llandro et al. 3D Nanoscale Gyroid Nanolett 2020  
 AS Koshikawa et al. Magnetic Order PRB 2023



# Nanomagnet Robots

## Magnetic Micromachines

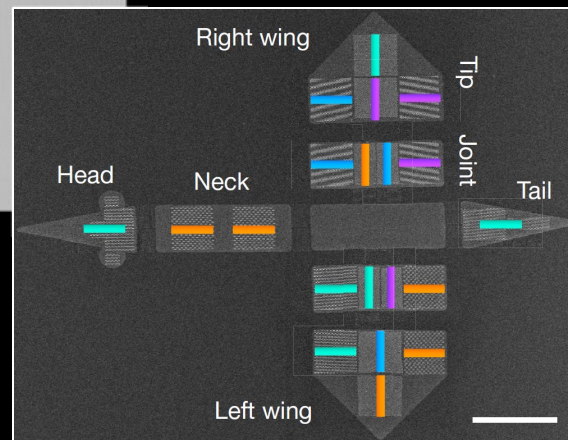
Jizhai Cui (with Bradley Nelson, Tian-Yun Huang & further group members)

### Origami Bird



J Cui et al. Nature (2019)

- Arrays of 60 nm thick Co nanomagnets
- 50 nm thick low-stress SiN Membrane panels
- Released in an organic liquid (PGMEA)
- Activated in a magnetic field:  
 $B \sim 1-10$  mT at 0-25 Hz



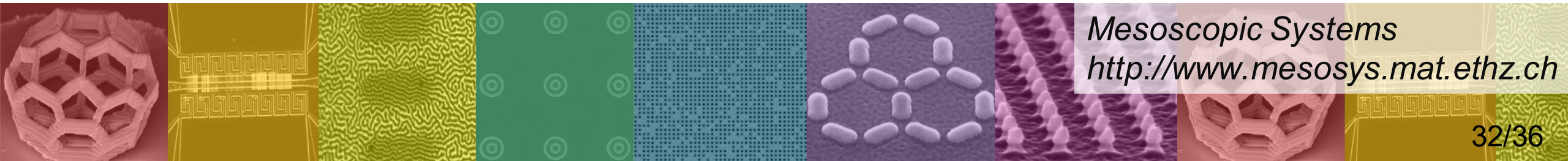
→ **3D Structures**

→ **Movement**

→ **Actuation & Robotics**

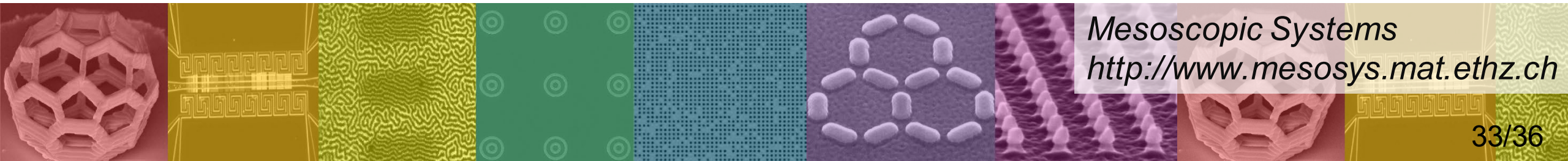
<https://marss-conference.org>  
“Manipulation, automation and robotics at small scales”

# Acknowledgements





# Thankyou to Mesoscopic Systems



Mesoscopic Systems  
<http://www.mesosys.mat.ethz.ch>

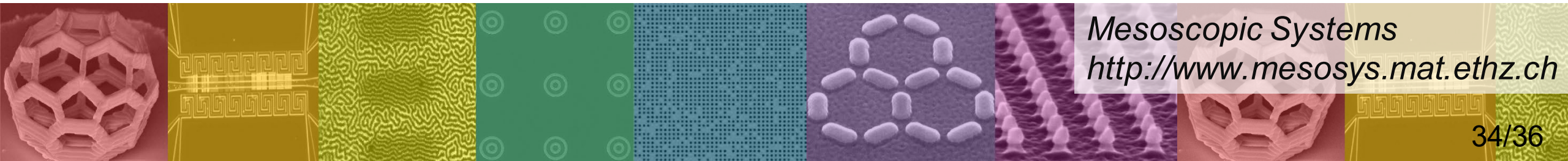
## **Research & Technical Staff, Paul Scherrer Institute**

### **Swiss Light Source**

- ❖ *Photoemission Electron Microscopy, SIM Beamline*  
Armin Kleibert, Carlos Vaz
- ❖ *Scanning Transmission X-ray Microscopy, PolLux Beamline*  
Joerg Raabe, Simone Finizio
- ❖ *X-ray Scattering, SIM Beamline*  
Urs Staub
- ❖ *X-ray Tomography and Laminography, cSAXs & PolLux Beamlines*  
Manuel Guizar Scicairos, Mirko Holler, Simone Finizio, Joerg Raabe

**Laboratory for Muon Spin Spectroscopy:** Hubertus Luetkens

**Condensed Matter Theory Group:** Peter Derlet





# Acknowledgements

- ETH Zurich:** Pietro Gambardella & Group  
Tianyun Huang, Bradley Nelson & Group
- Peking University:** Zhaochu Luo & Group
- MPI Chem. Phys. Sol:** Claire Donnelly
- Univ. St Andrews:** Machiel Flokstra, Steve Lee
- Univ. Cambridge:** Gunnar Möller
- SOLEIL Synchrotron:** Nicolas Jaouen, Jean-Marc Tonnerre,  
Jan Lüning, Maurizio Sacchi
- Adv. Light Source:** Andreas Scholl & Colleagues

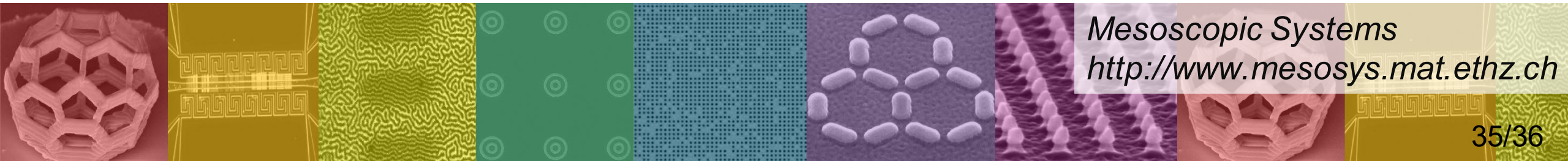


**Spin  
ENGINE**

<https://spinengine.eu>



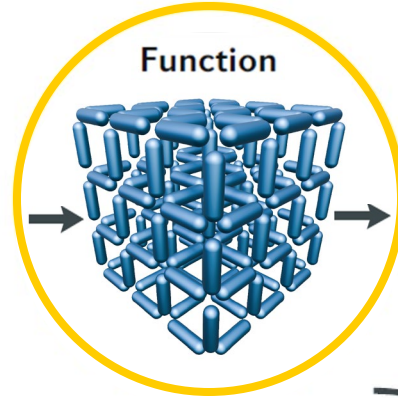
**Swiss National  
Science Foundation**



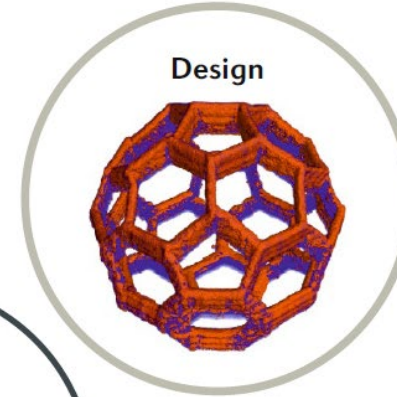
# Artificial Spin Ice Activities in Three Dimensions

- Computation
- Magnonics
- Hybrid systems

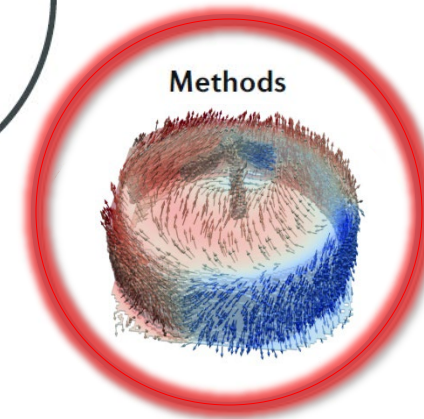
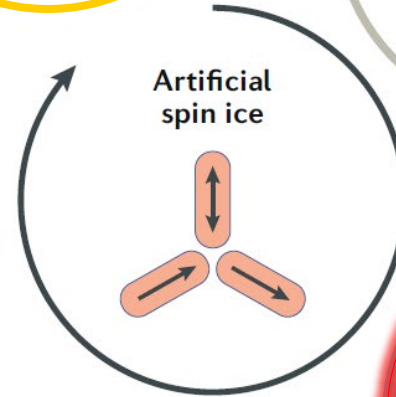
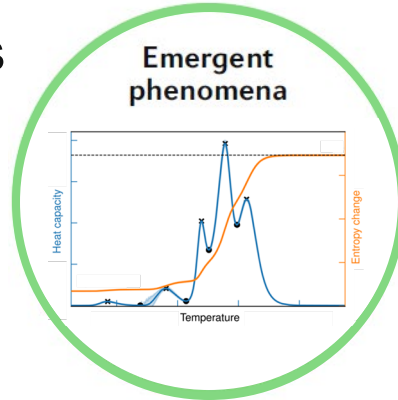
2D:  $2^{24} \sim 2 \times 10^7$   
 3D:  $2^{144} \sim 2 \times 10^{43}$



- Geometries
- Materials

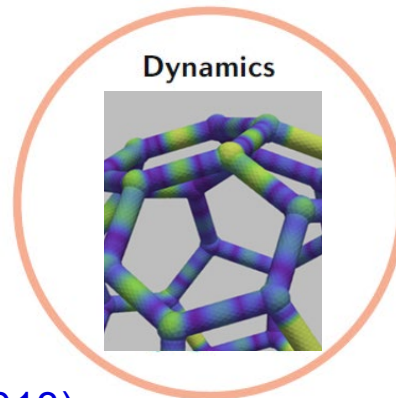


- Magnetic monopoles
- Phases transitions
- Vertex frustration
- Chirality



## Enabling Technologies

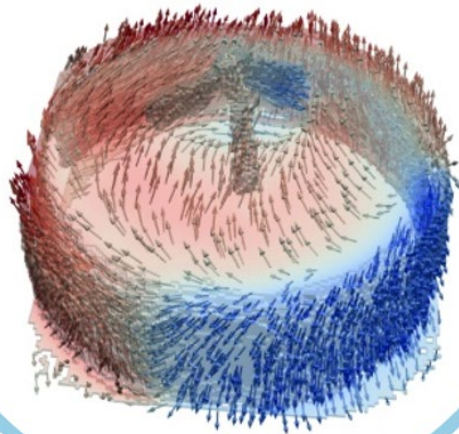
- Driven processes
- Thermal relaxation



- Fabrication ←
- Characterization ←
- Manipulation
- Theory & Simulations ←



## Methods

**Enabling Technologies**

*“3D is exciting, but we have to make it feasible”*

**I. Fabrication**

- Control of feature sizes & geometries, large volumes
- Electrodeposition & FEBID of high-quality magnetic materials

**II. Characterization****• Imaging**

- Large penetration depths
- High spatial & temporal resolution
- Synchrotron X-rays
- Electron microscopy
- Lab Based Methods: MFM & Kerr Microscopy

- **Magnetic Properties:** MOKE, SQUID VSM, Heat Capacity

- **Transport:** read & write magnetic states

- **Magnetization Dynamics**

**III. Theory & Simulations**

- Dynamics & Thermodynamics: phases & phase diagrams
- Multiscale Materials Modelling: atoms – microstructure – properties