



WIR SCHAFFEN WISSEN – HEUTE FÜR MORGEN

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

# **Artificial Ferroic Systems: from hybrid systems to magnetic metamaterials**

**Topology Matters, July 25th-28<sup>th</sup>, 2017**  
**JGU MITP/SPICE, Mainz, Germany**

L J Heyderman & R L Stamps  
J. Phys.: Condens. Matter 25 (2013)

# **ARTIFICIAL FERROIC SYSTEMS**

Dynamics

Ferroics

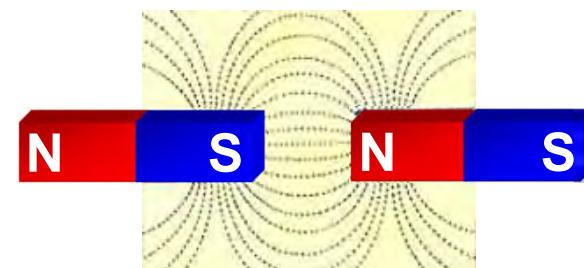
Interactions

Structure

# Artificial Ferroic Systems

Hybrid  
or  
Composite  
Systems

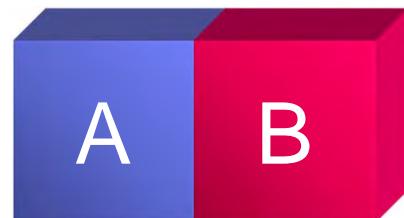
Artificial  
Spin  
Systems



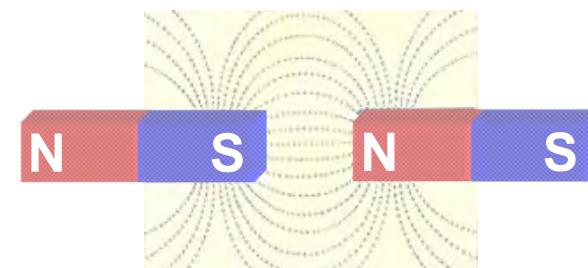
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# Artificial Ferroic Systems

Hybrid  
or  
Composite  
Systems

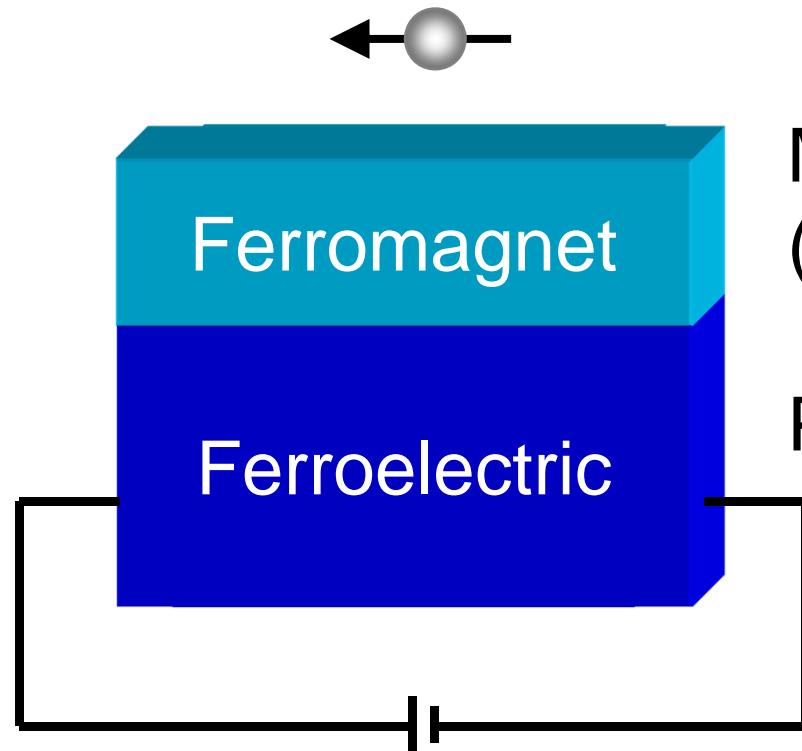


Artificial  
Spin  
Systems



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# Multiferroic Composite/Artificial Multiferroic

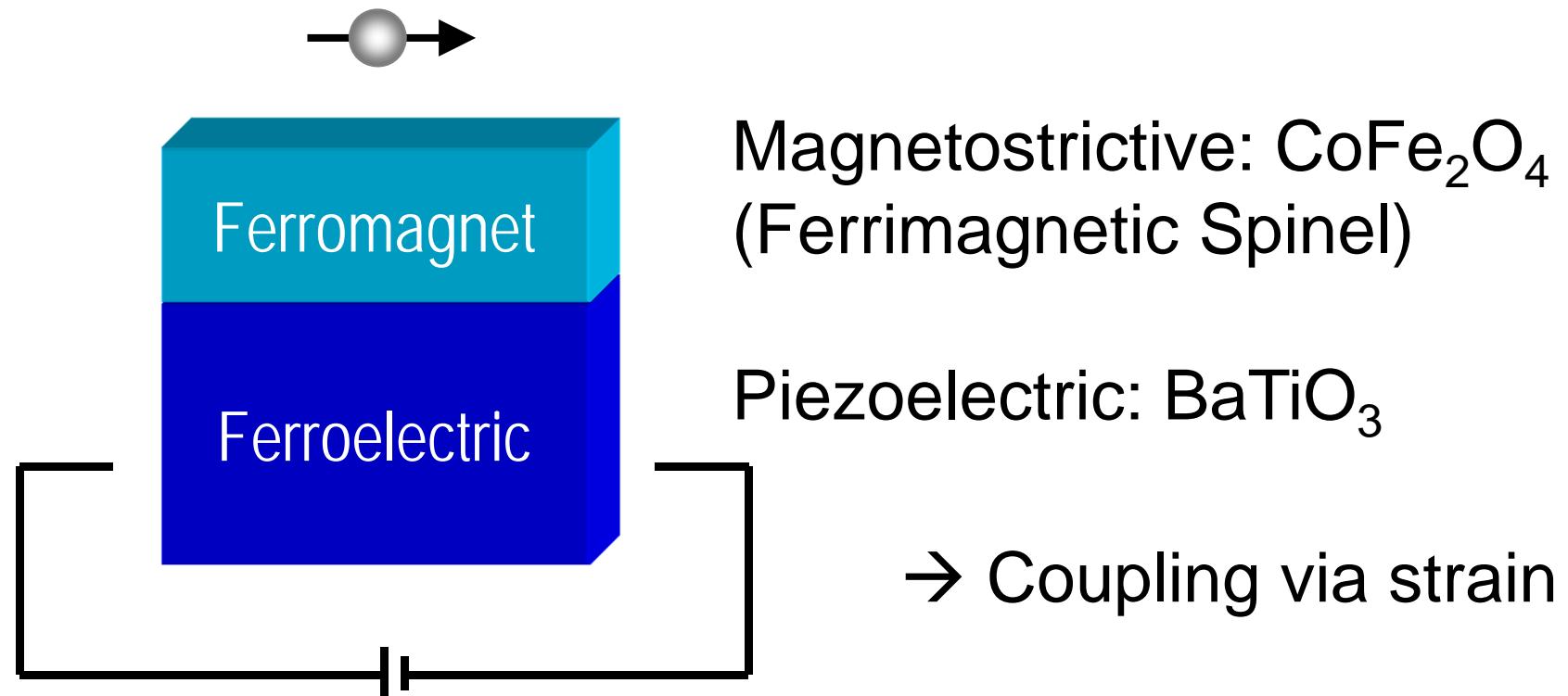


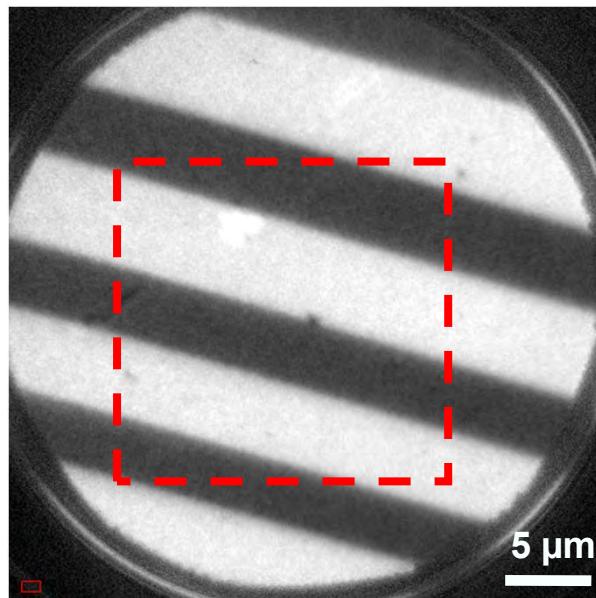
Magnetostrictive:  $\text{CoFe}_2\text{O}_4$   
(Ferrimagnetic Spinel)

Piezoelectric:  $\text{BaTiO}_3$

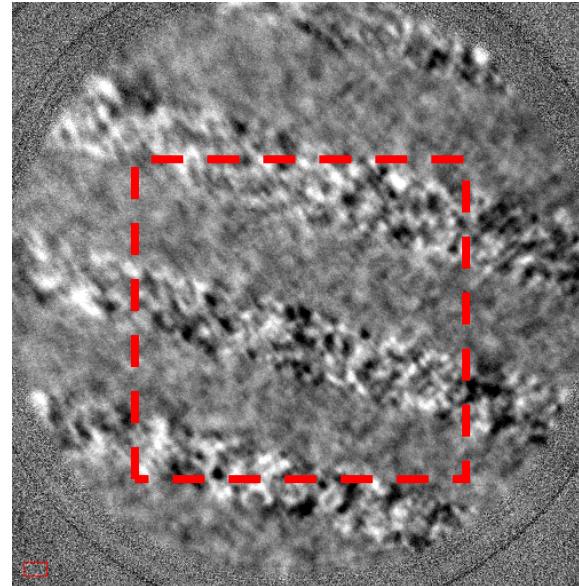
→ Coupling via strain

# Multiferroic Composite/Artificial Multiferroic

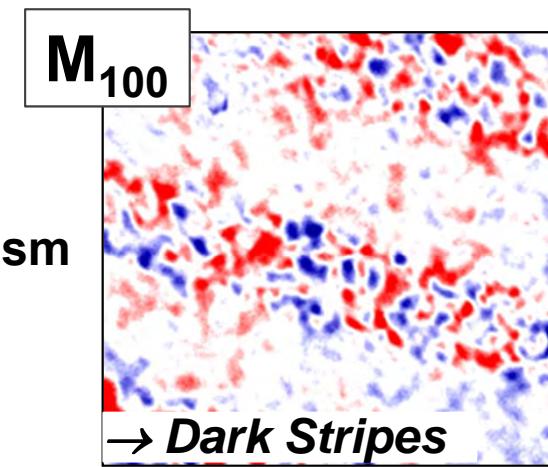
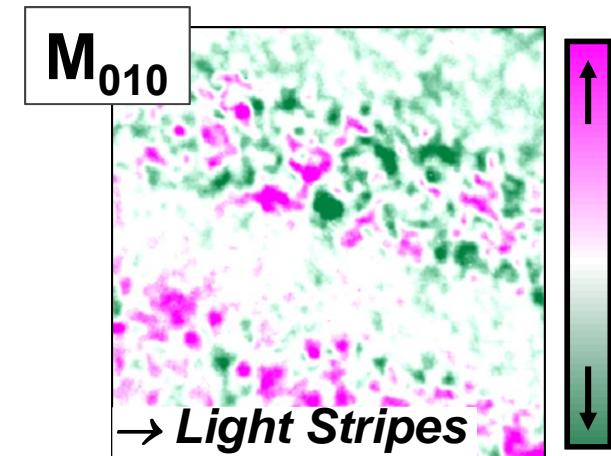




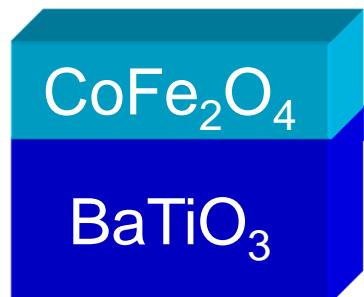
Fe X-ray Linear Dichroism



Fe X-ray Magnetic Circular Dichroism

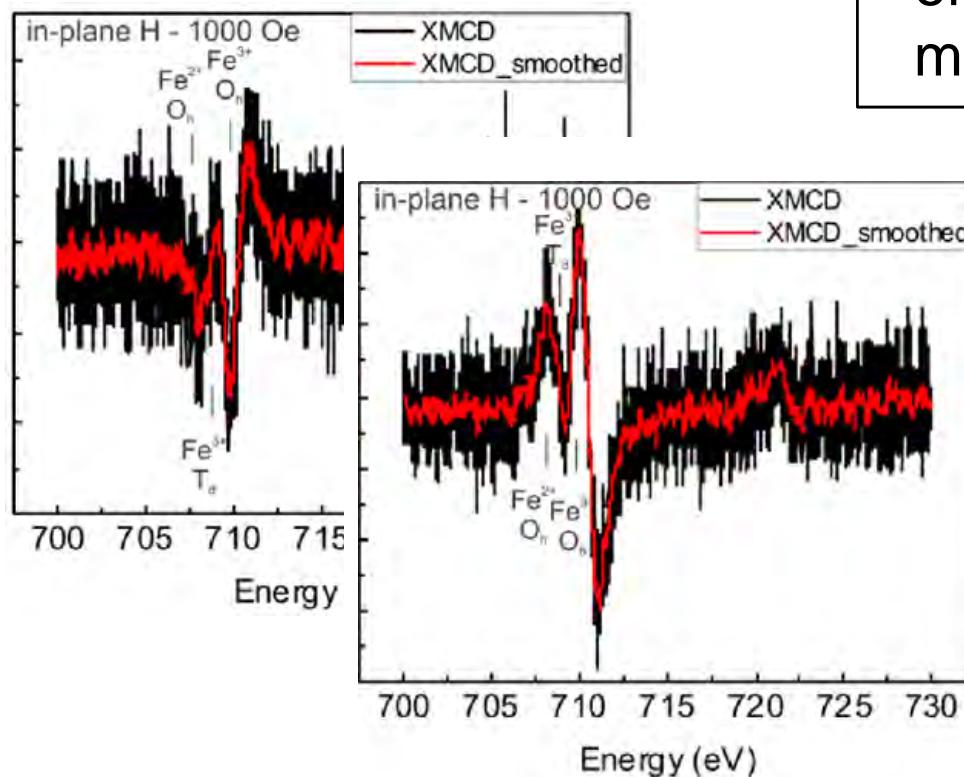
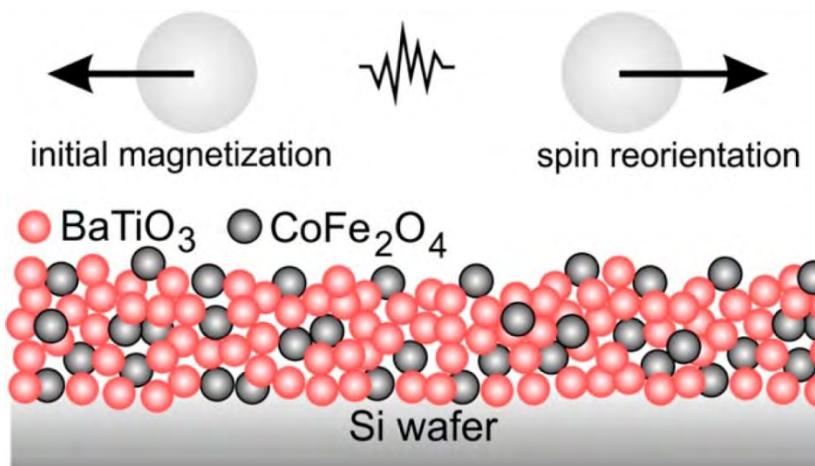


$M_{001} \sim 0$



- Fe XLD: strain domains
- Fe XMCD: magnetic domains
- Substrate-induced strain  
modifies magnetic anisotropy  
 $\rightarrow$  Strain Imprints Magnetic States

# Multiferroic Composite Thin Films



- Spin-coated nanoparticle dispersions of  $\text{BaTiO}_3$  &  $\text{CoFe}_2\text{O}_4$
- Electrical & magnetic studies → both ferroelectric & ferromagnetic order
- Demonstrated coupling between orders with voltage control of magnetism

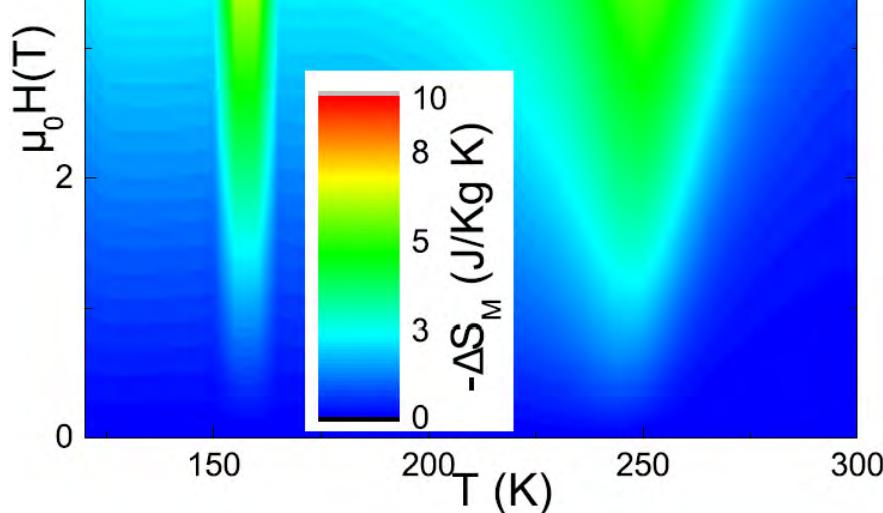
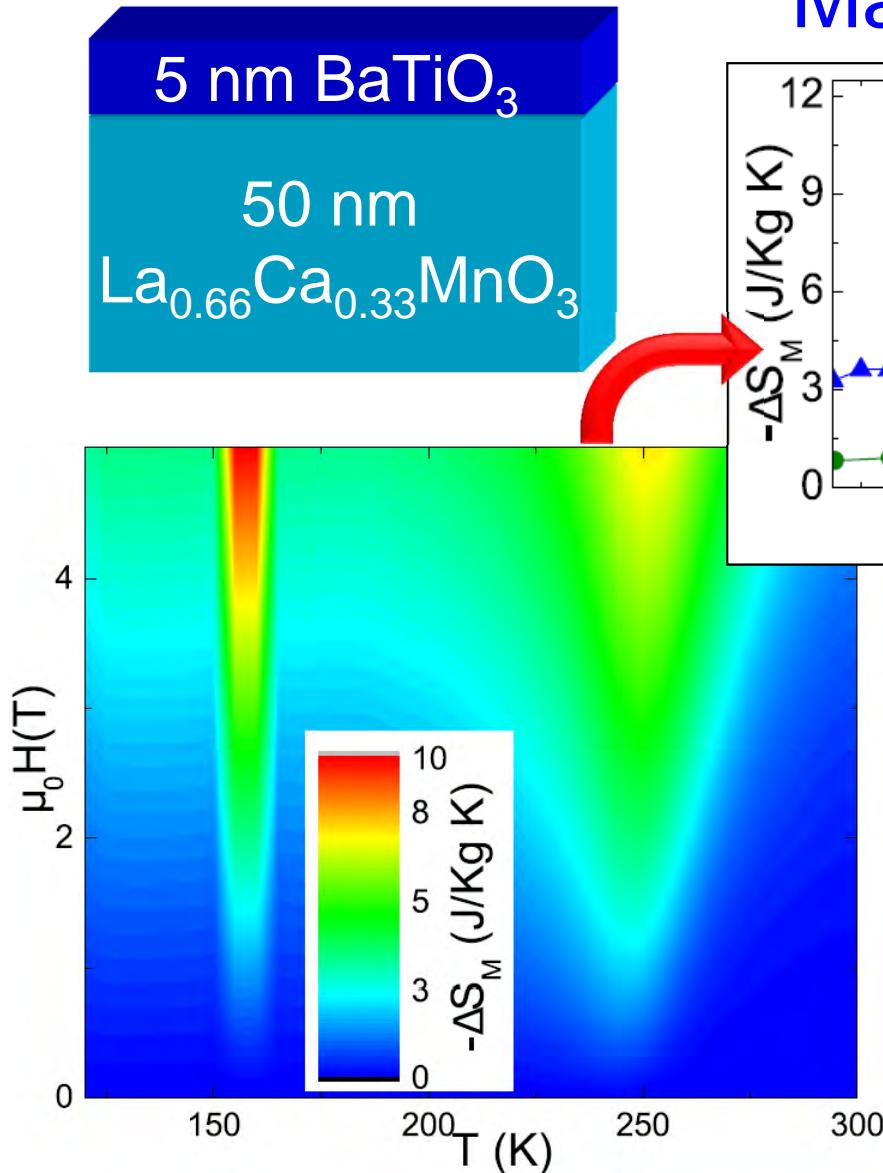
## XMCD Spectra

$\text{BaTiO}_3$  – 5 wt %  $\text{CoFe}_2\text{O}_4$  sample  
magnetization reversal after 40 V  
DC electrical poling

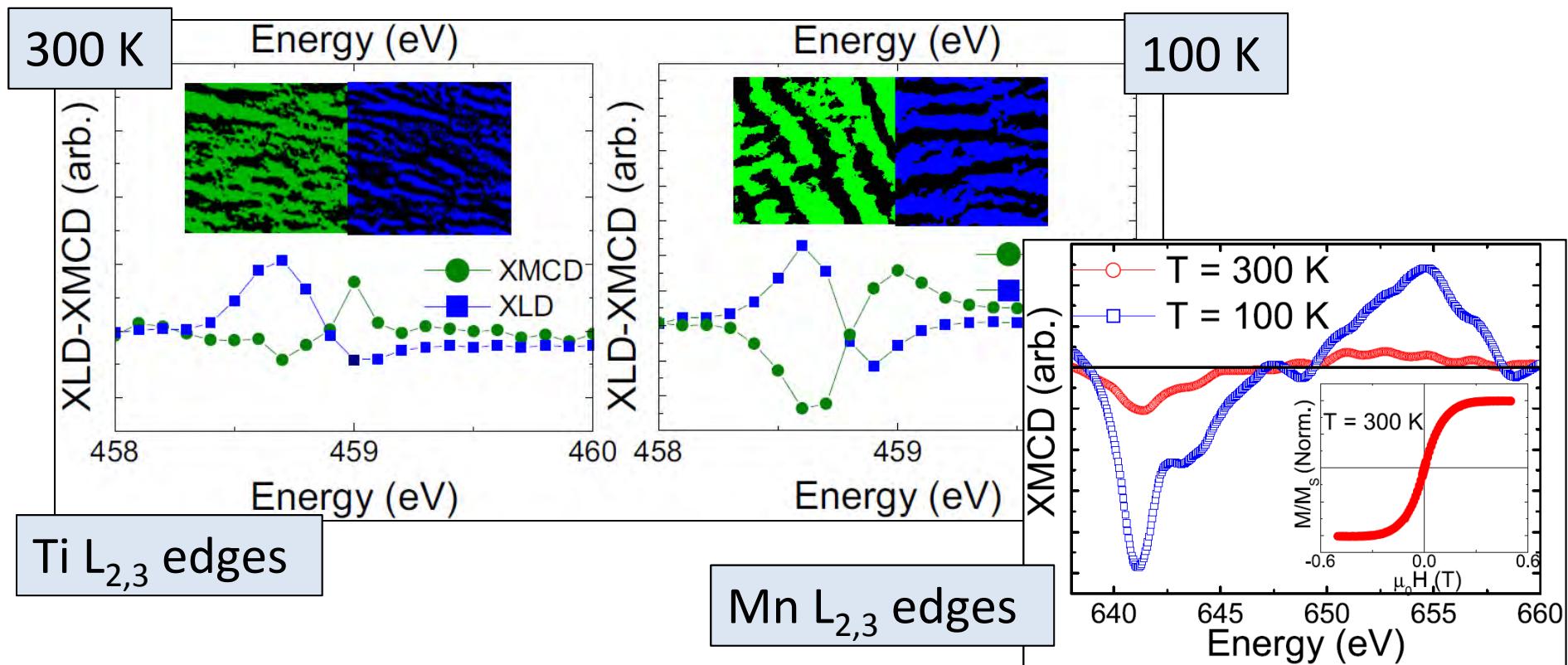
D. Erdem, N.S. Bingham, F.J. Heilitag,  
N. Pilet, P. Warnicke, C.A.F. Vaz, Y. Shi,  
M. Buzzi, J.L.M. Rupp, L.J. Heyderman,  
M. Niederberger, ACS Nano (2016)

D. Erdem et al., Adv Func Matls (2015)

# Strain-induced Enhancement of Magnetocaloric Effect

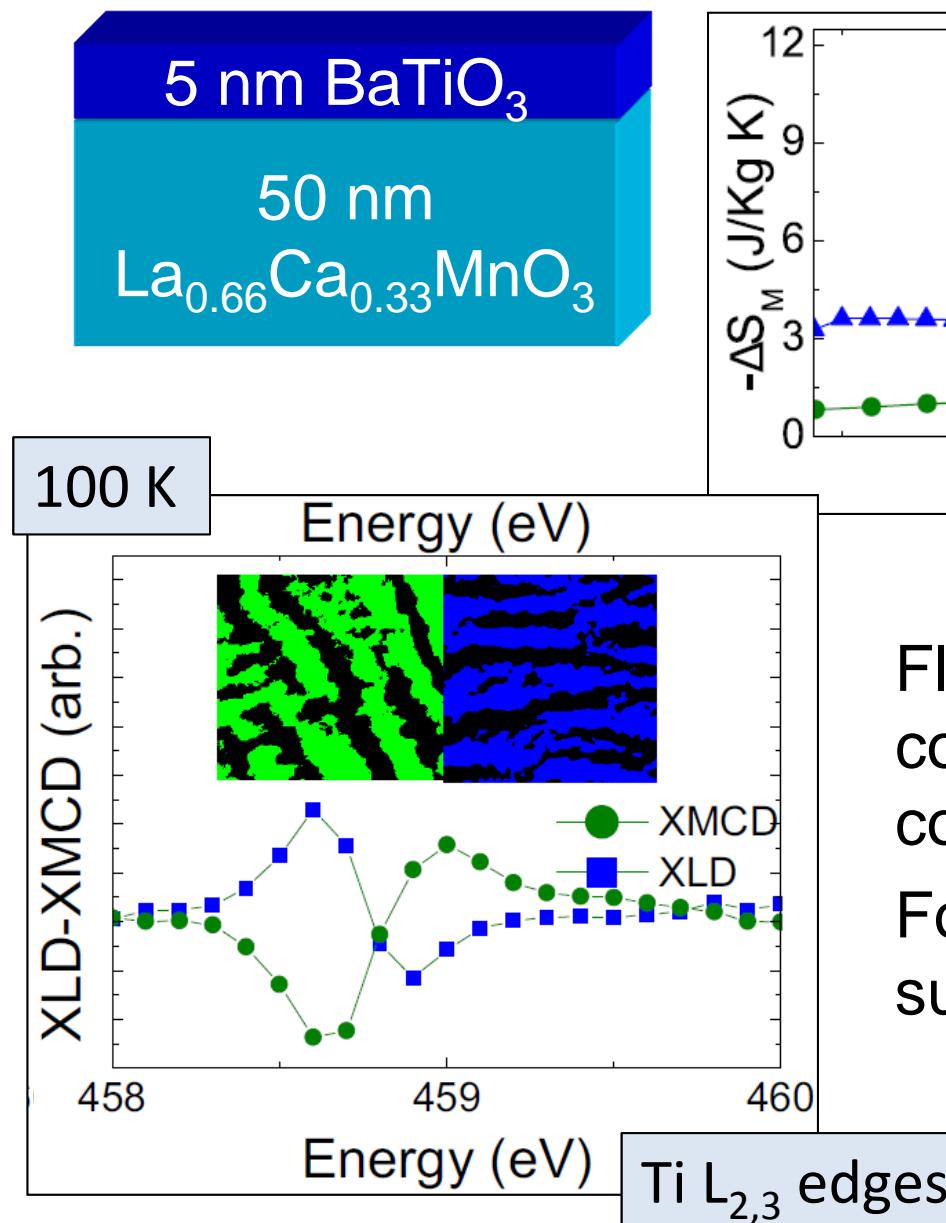


- Large MCE at 150 K
- Enhancement of MCE at 250 K
- Due to structural phase transitions in BTO:
  - orthorhombic-rhombohedral (150 K)
  - orthorhombic-tetragonal (250 K)



X-ray absorption spectroscopy measurements:

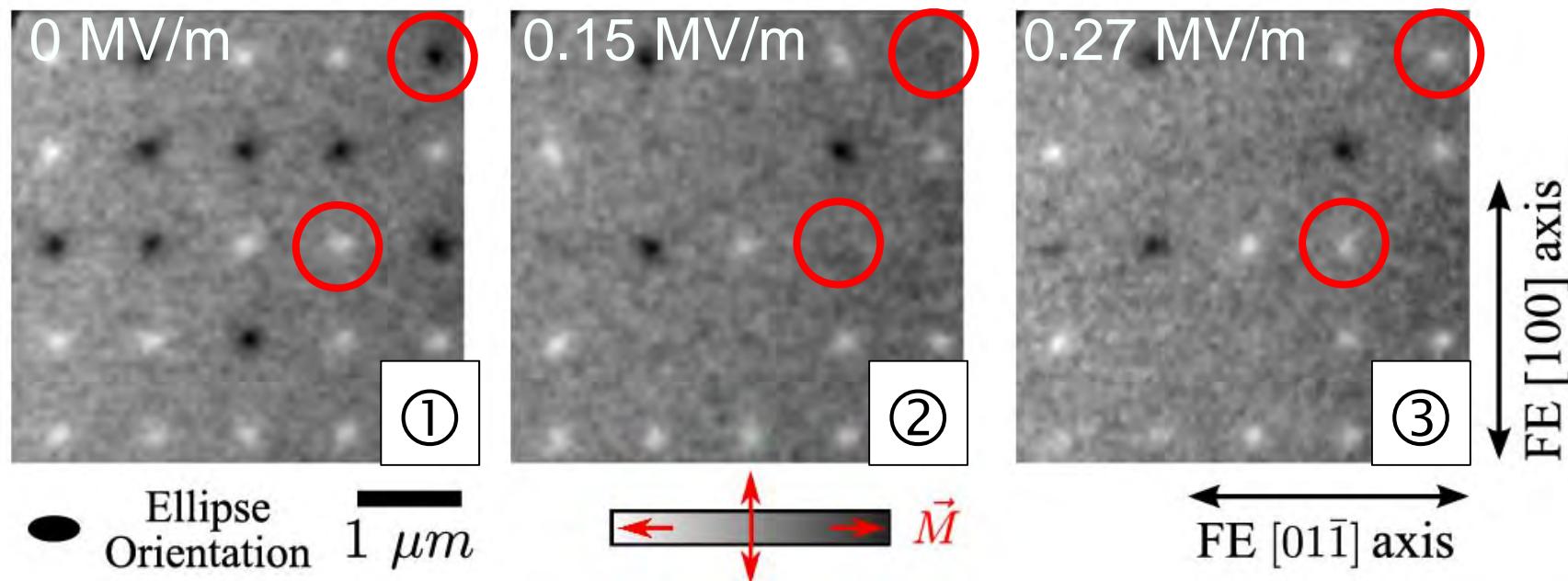
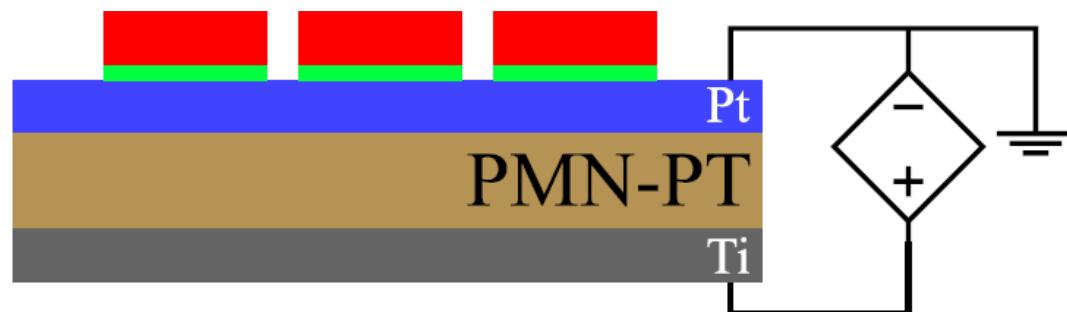
- Room temperature ferromagnetism at interface due to electronic reconfiguration of LCMO and BTO
- Indication that charge carrier transfer results from hybridization of  $\text{Mn}^{3+/4+}$  and  $\text{Ti}^{4+}$  to give  $\text{Mn}^{2+/3+}$  and  $\text{Ti}^{3+}$



Flexible approach to enhance & control further phenomena in complex oxide heterostructures  
For example:  
superconductivity & multiferroicity

M. Buzzi, R.V. Chopdekar, J. L. Hockel,  
A. Bur, T. Wu, N. Pilet, P. Warnicke, G. P. Carman  
L. J. Heyderman, and F. Nolting PRL 2013

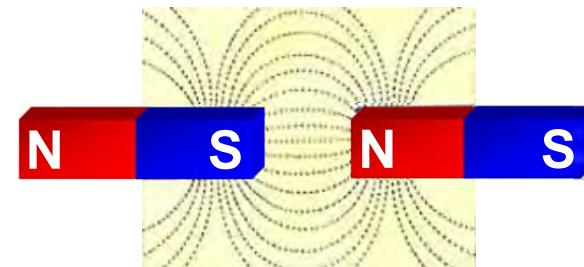
### Nickel Nanoislands



# Artificial Ferroic Systems

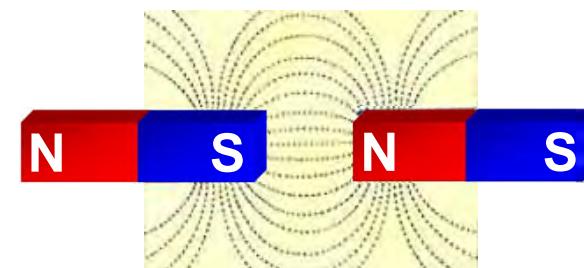
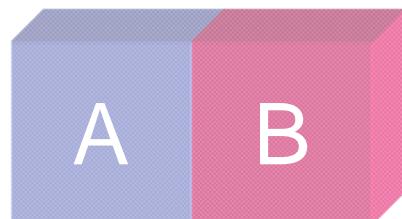
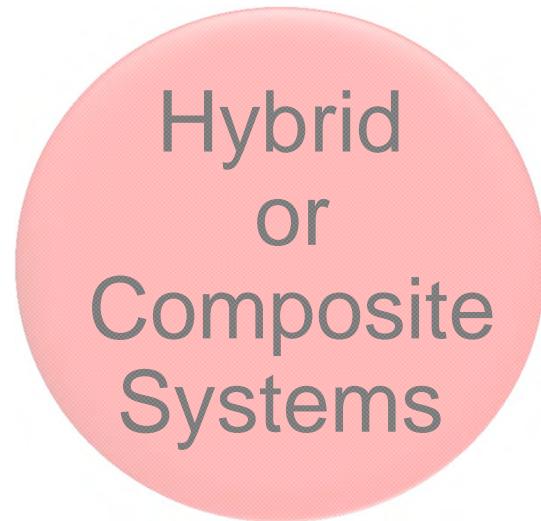
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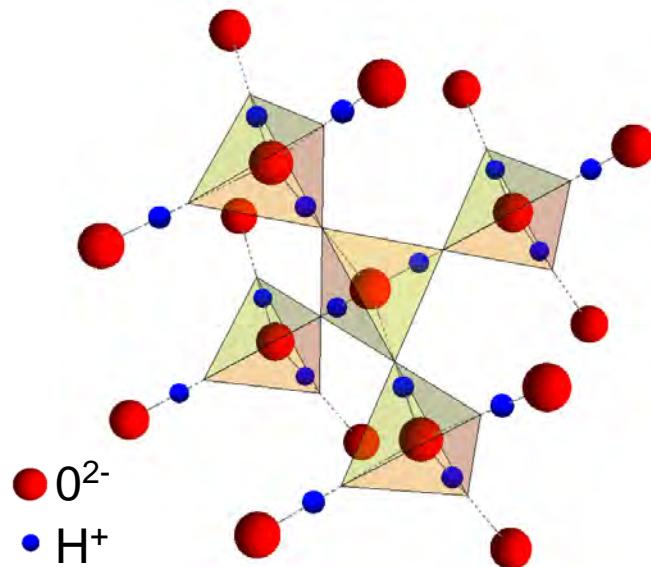


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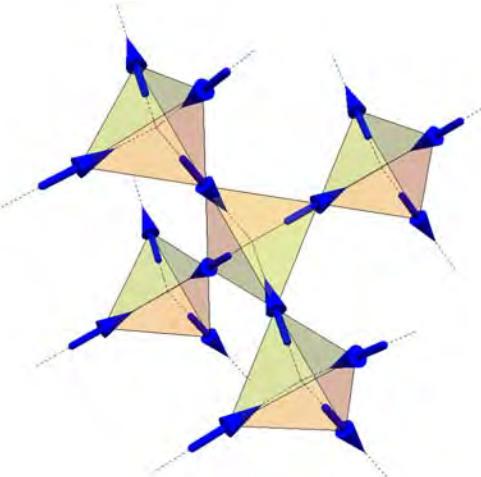
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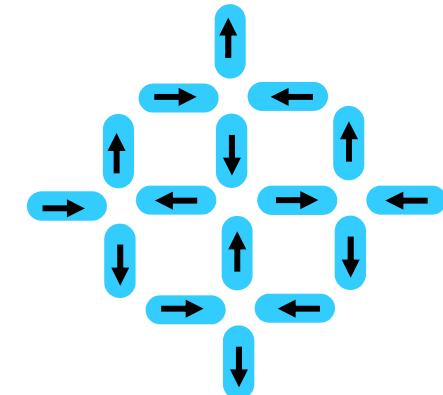
L J Heyderman & R L Stamps  
J. Phys.: Condens. Matter (2013)



Water Ice



Spin Ice

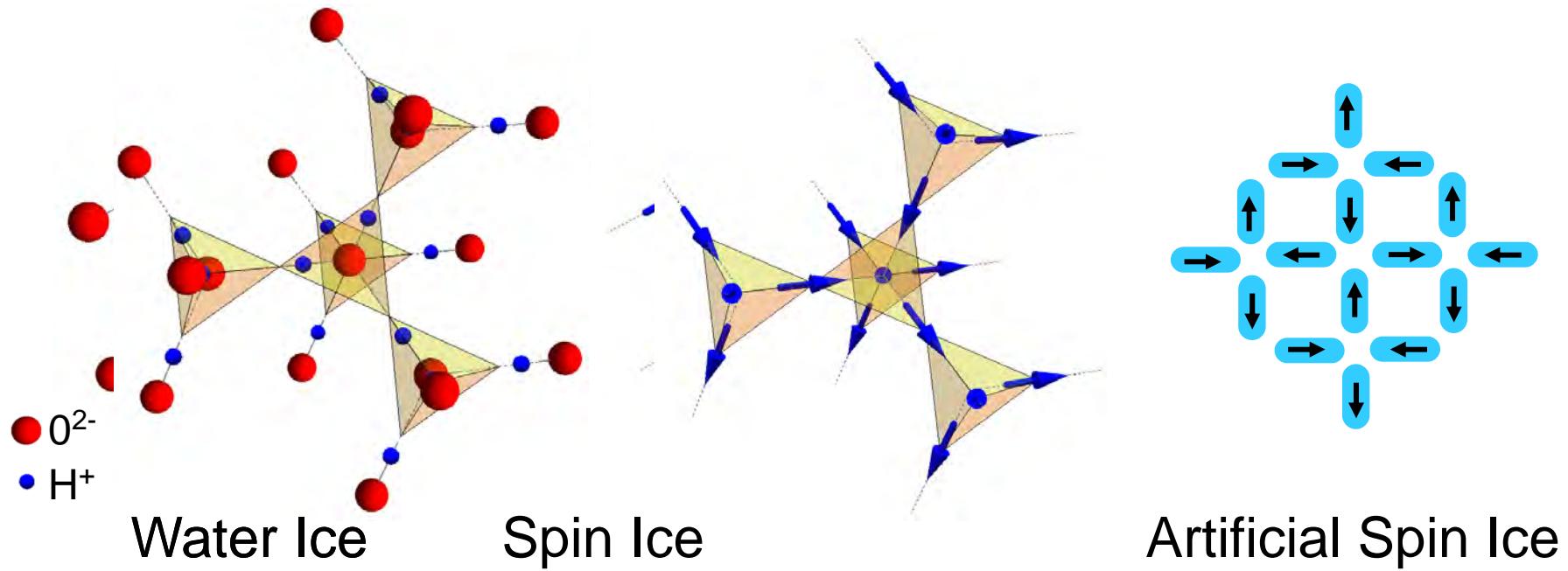


Artificial Spin Ice

MJ Harris *et al.*  
PRL (1997)

RF Wang *et al.*  
Nature (2006)

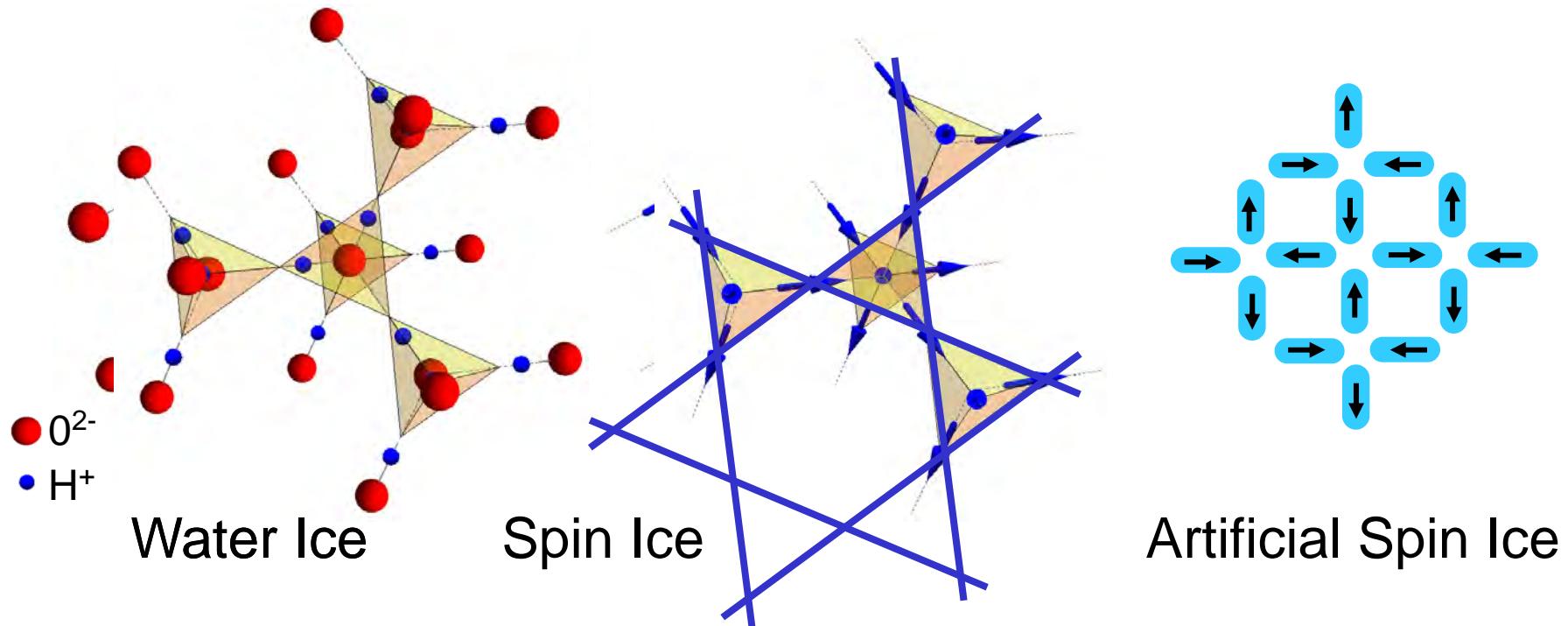
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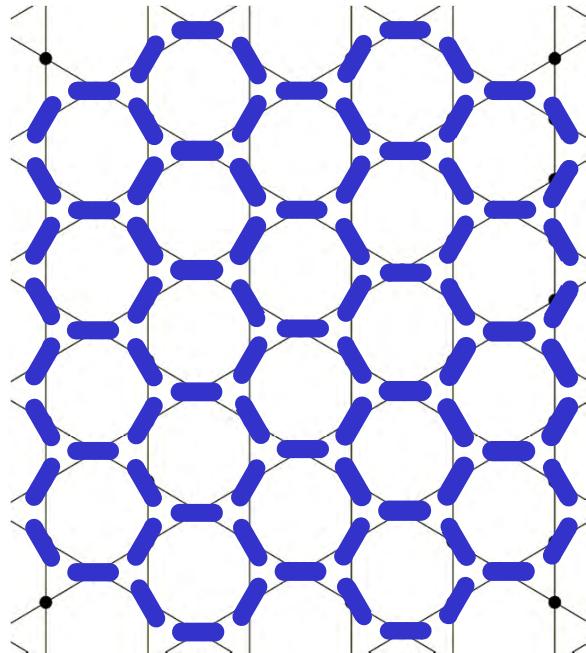


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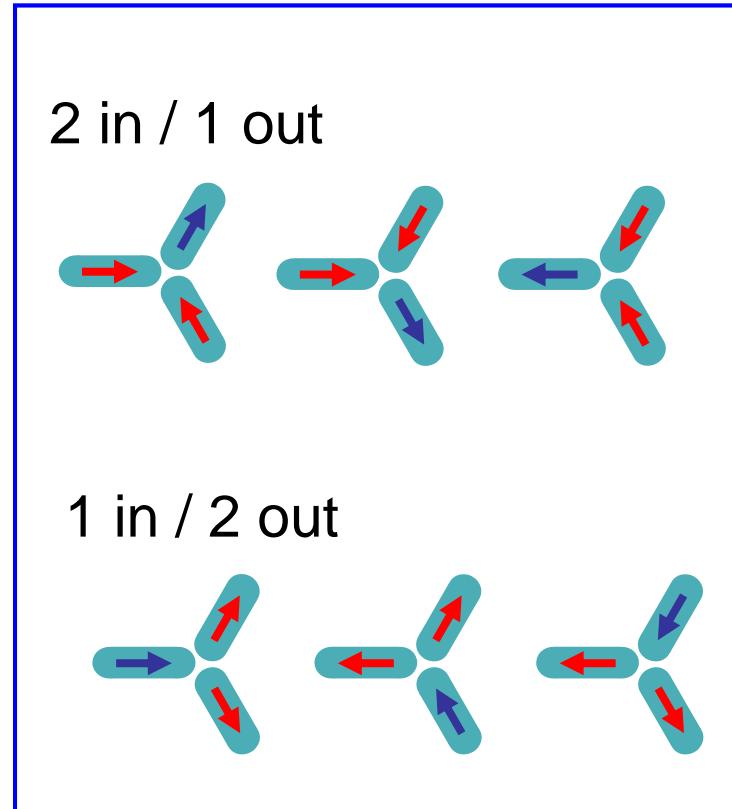
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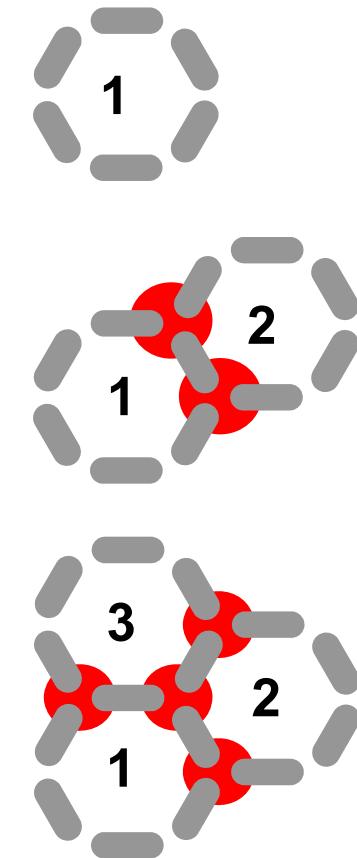
# Artificial Kagome Spin Ice



Kagome lattice

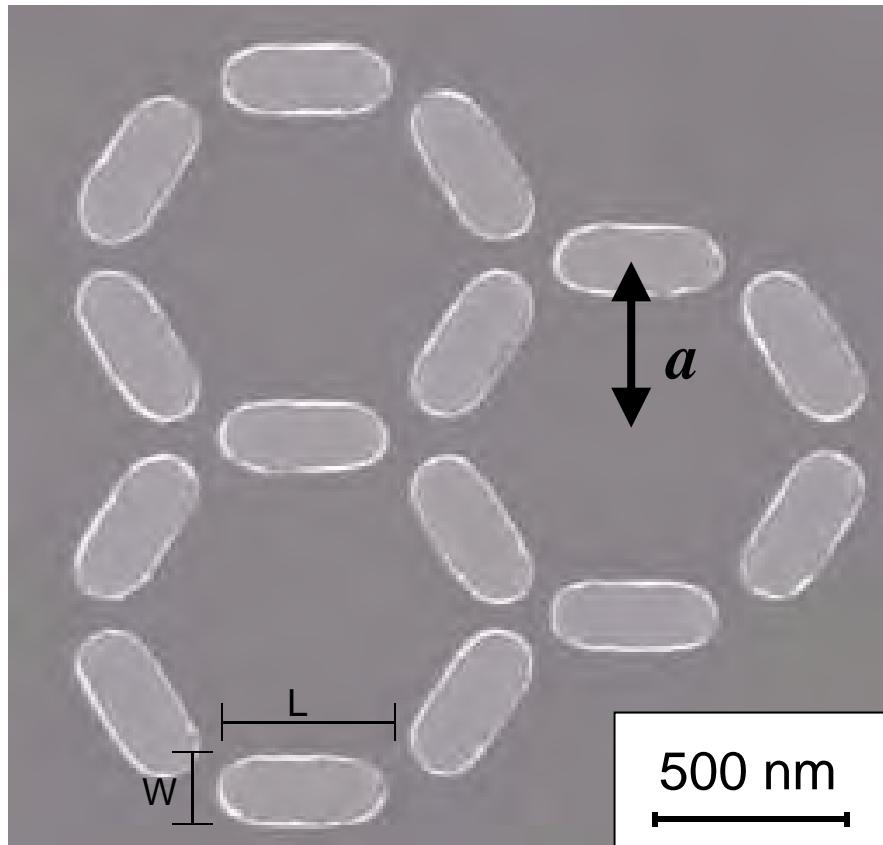


Ice rule



Building Blocks

# Fabrication



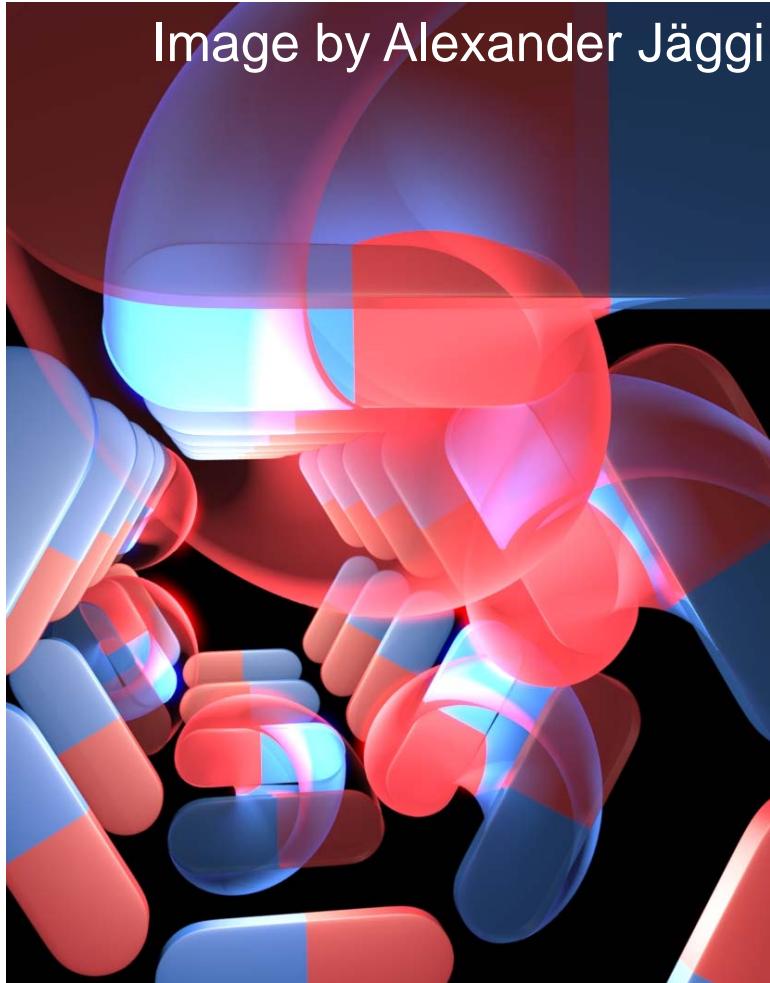
## *Electron Beam Lithography*

- Elongated ferromagnetic islands:  
e.g. Permalloy or Cobalt
- Aluminum capping layer
- Vary island size & lattice parameter

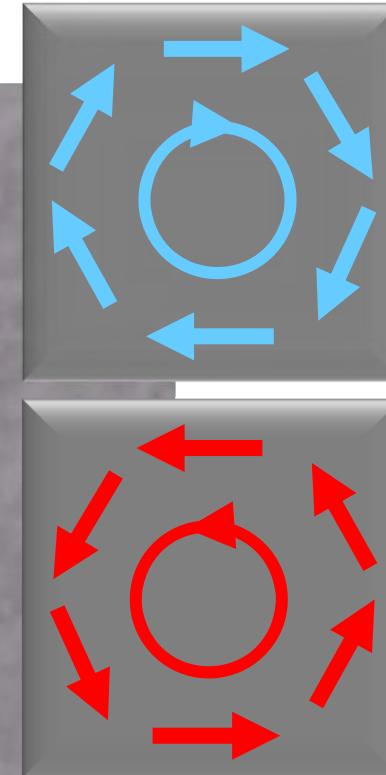
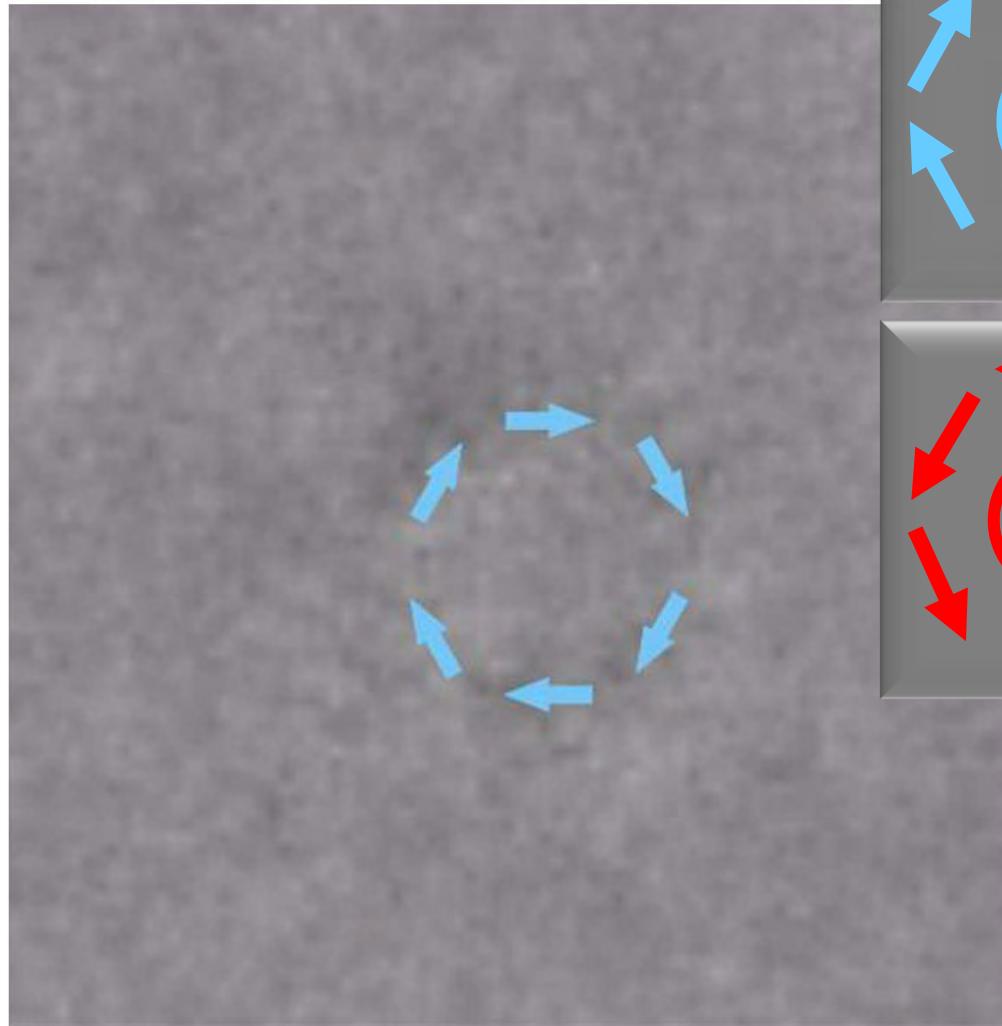
Width	= 170 nm
Length	= 470 nm
Smallest $a$	= 500 nm
Thickness	= 3-40 nm

E. Mengotti et al. Phys. Rev. B (2008)  
A. Trabesinger, News & Views, Nature Physics (2008)

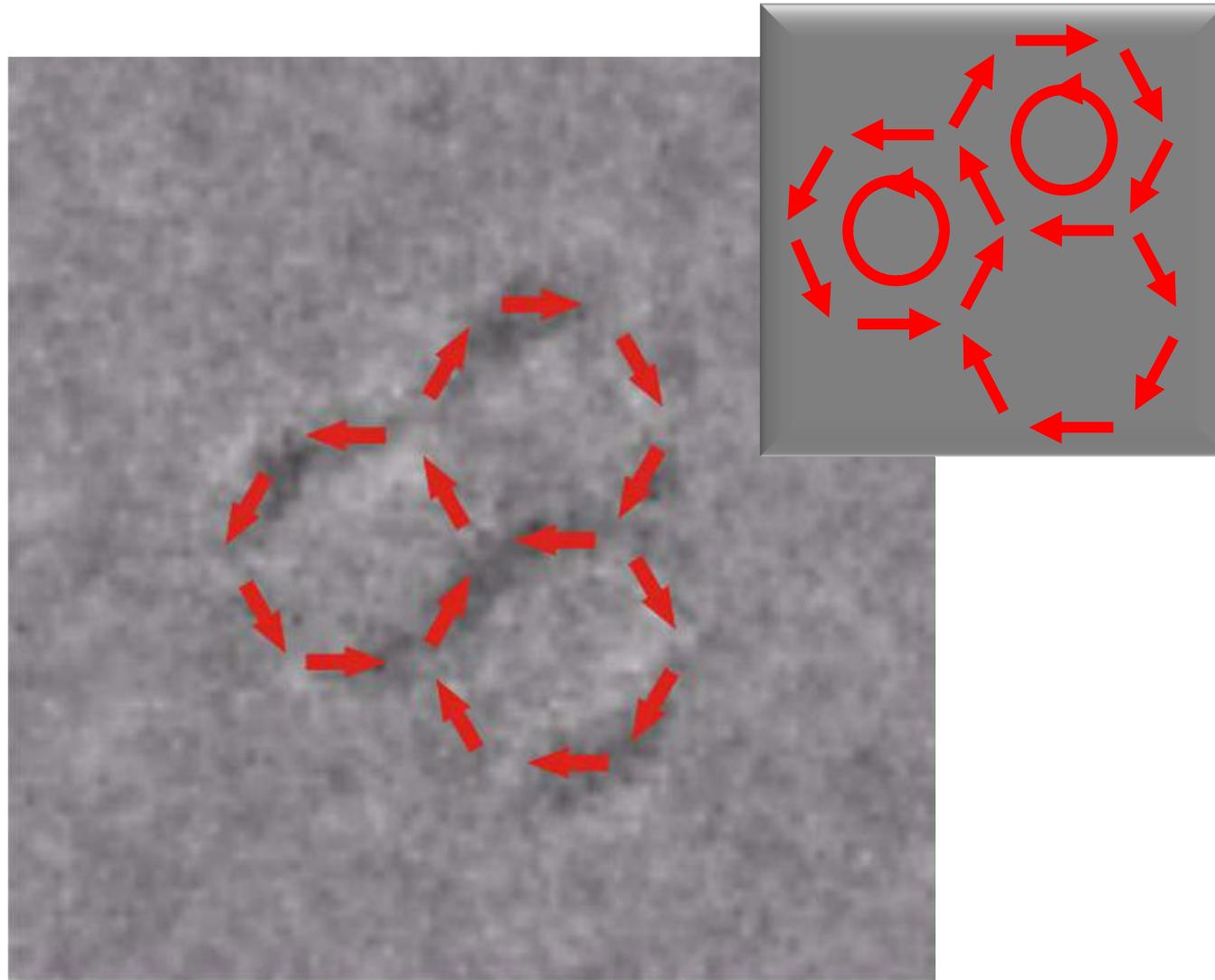
Image by Alexander Jäggi



L. Heyderman, *News & Views*,  
Nature Nanotechnology (2013)

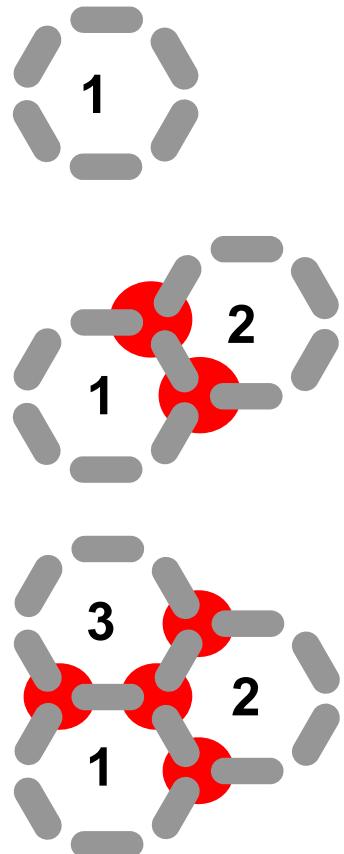


Photoemission Electron Microscopy  
Supplementary Movie from A. Farhan et al. Nature Physics (2013)

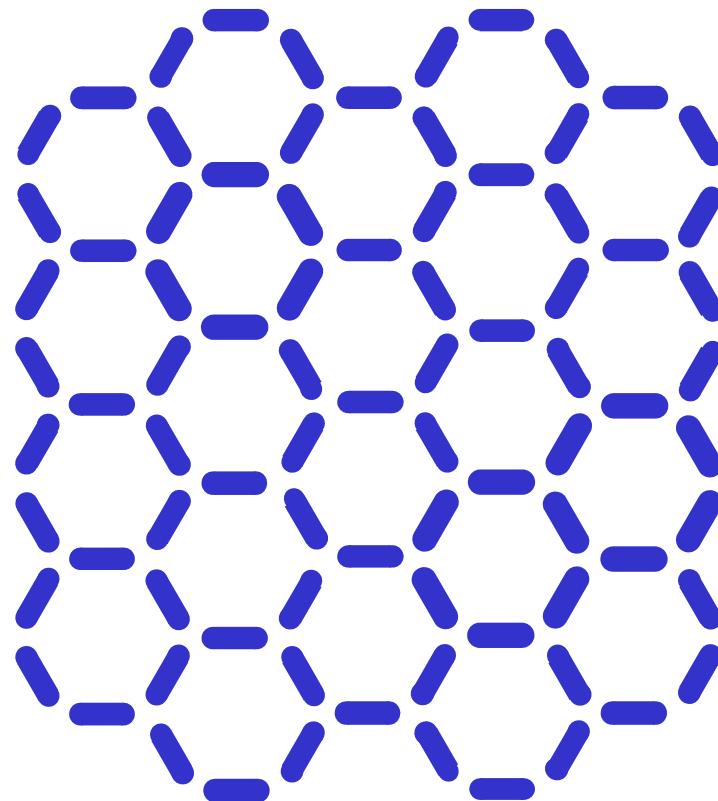


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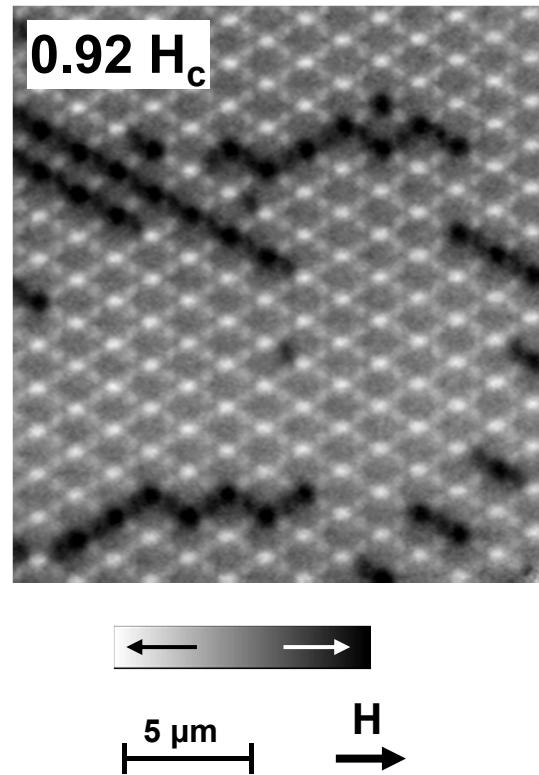
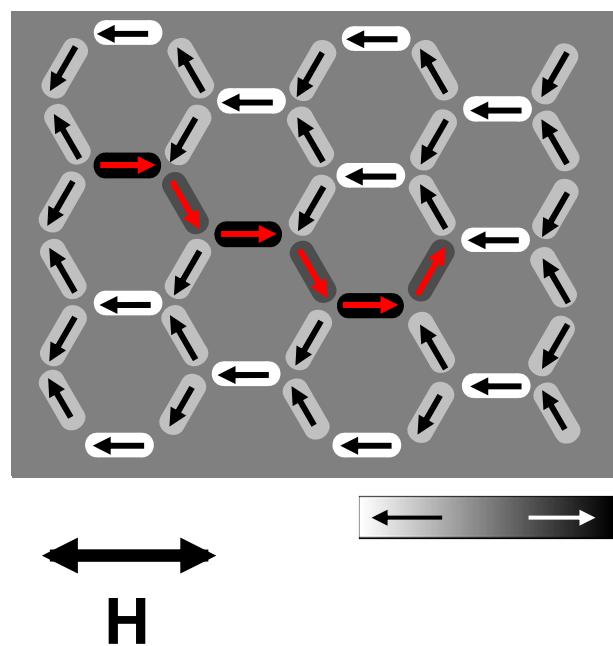


**Building Blocks**



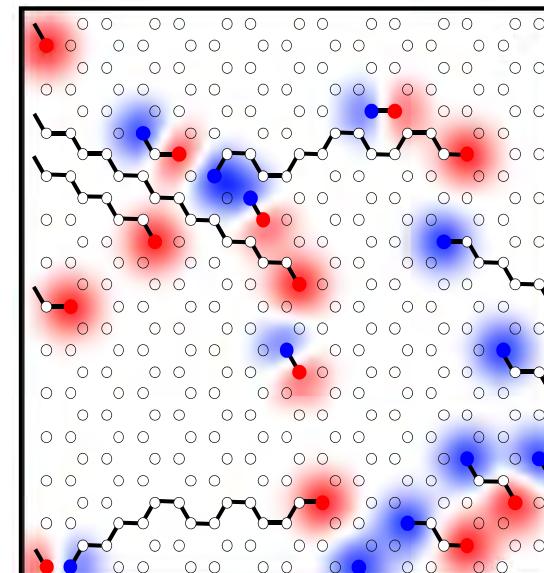
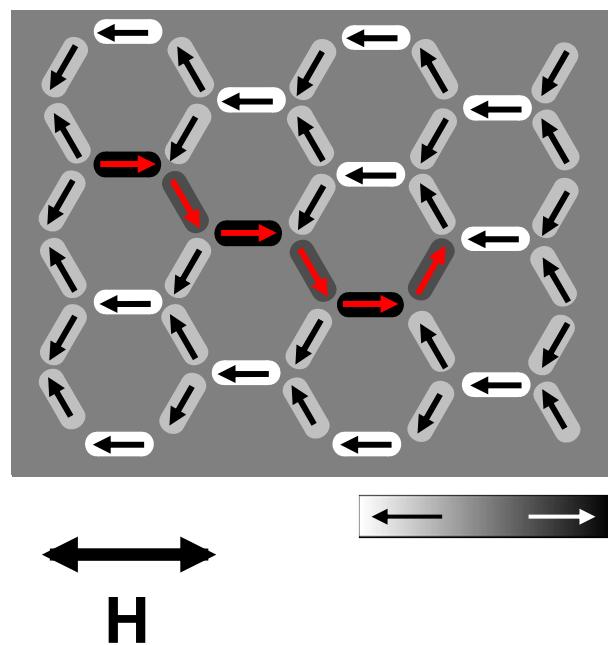
**Extended Arrays**

# Emergent Magnetic Monopoles & Dirac Strings



E. Mengotti, L. J. Heyderman, A. Fraile Rodríguez, F. Nolting, R.V. Hügli, H. B. Braun  
Nature Physics (2011)

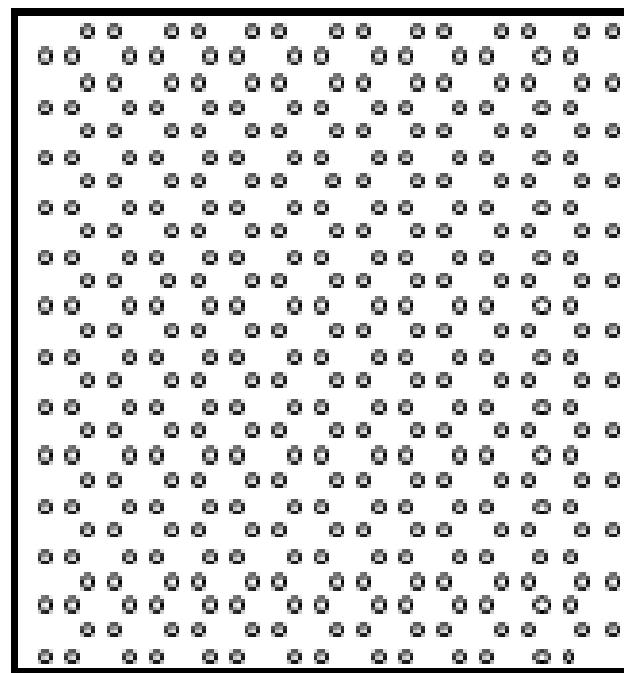
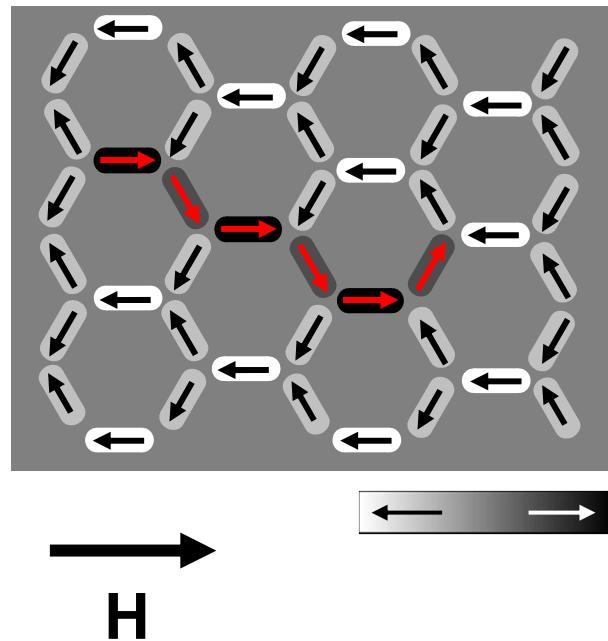
# Emergent Magnetic Monopoles & Dirac Strings



$$\begin{array}{c} \tilde{\rho}_m \\ \Delta Q/q \end{array} \quad \begin{array}{ccccc} \textcolor{blue}{\bullet} & & \textcolor{white}{\circ} & & \textcolor{red}{\bullet} \\ -2 & -1 & 0 & 1 & 2 \end{array}$$

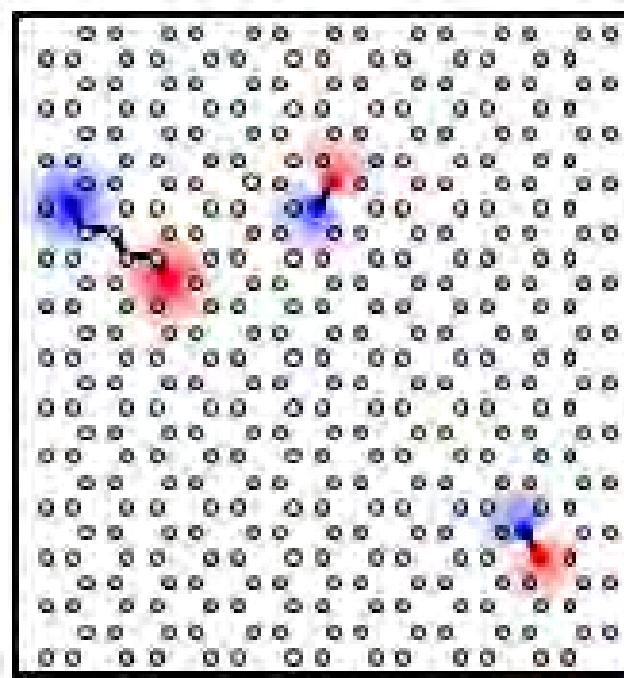
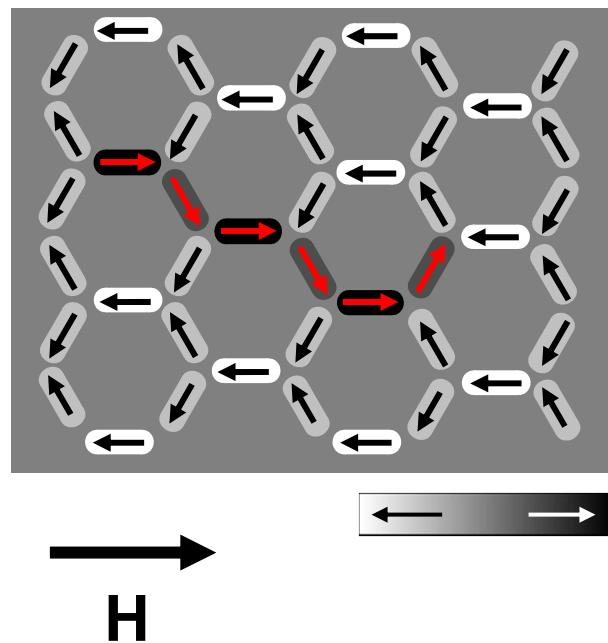
E. Mengotti, L. J. Heyderman, A. Fraile Rodríguez, F. Nolting, R.V. Hügli, H. B. Braun  
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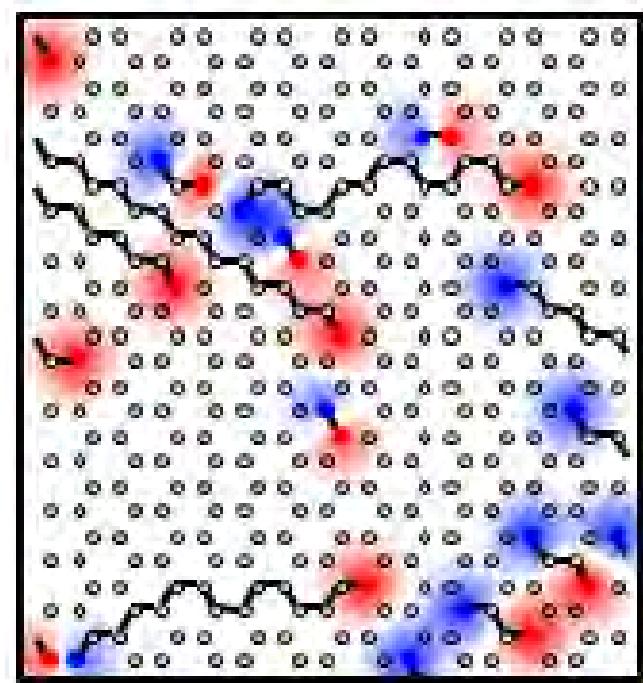
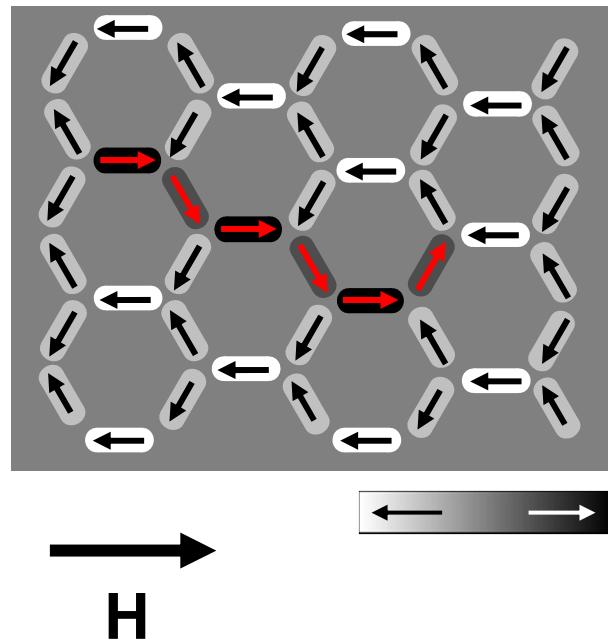
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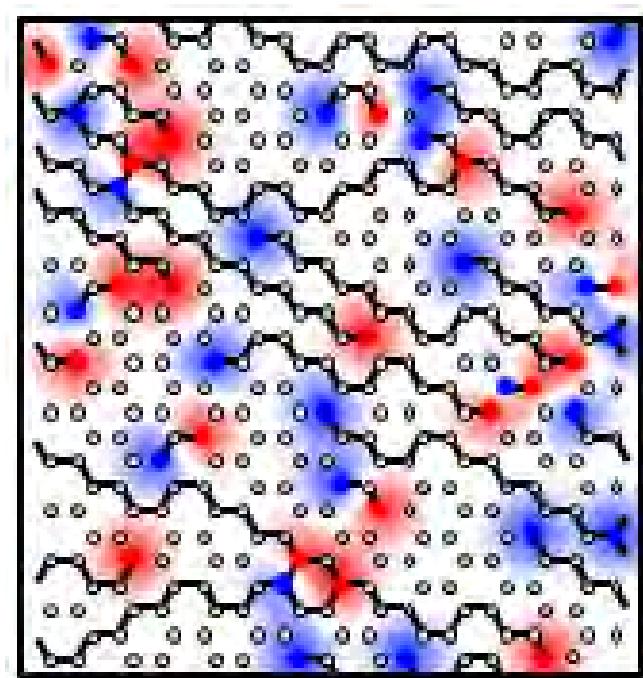
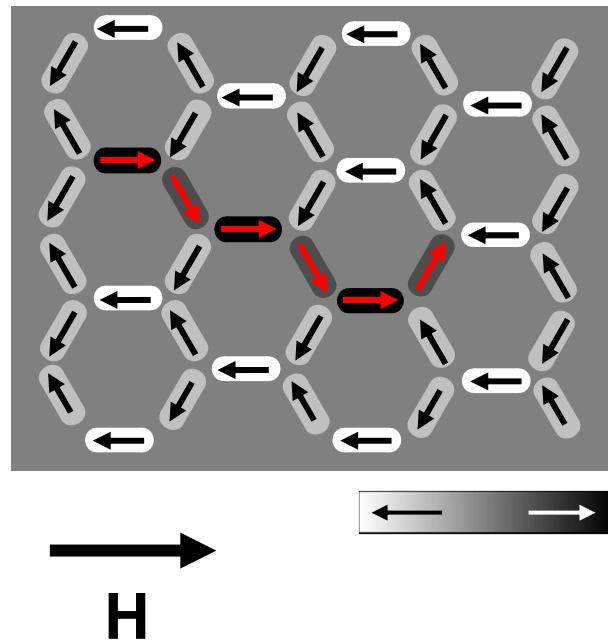
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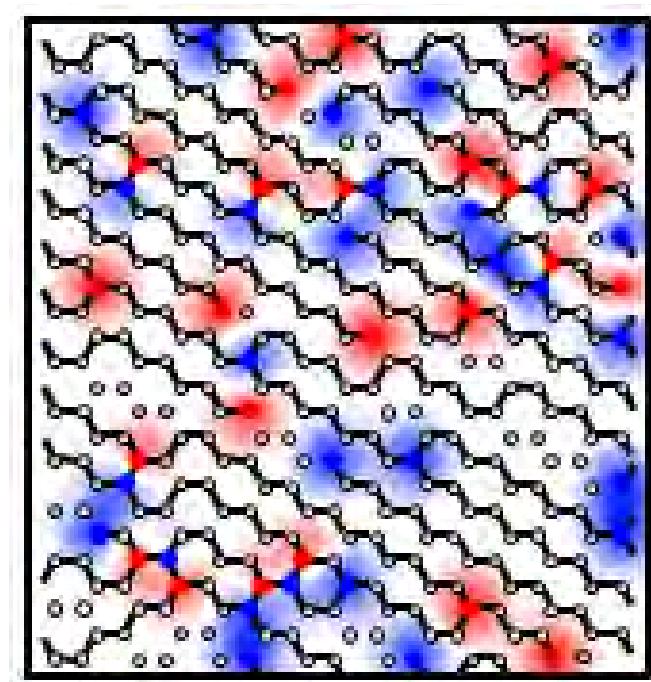
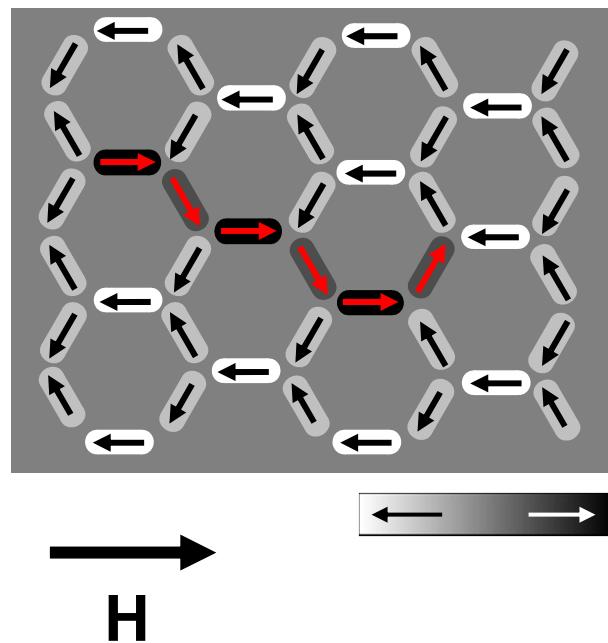
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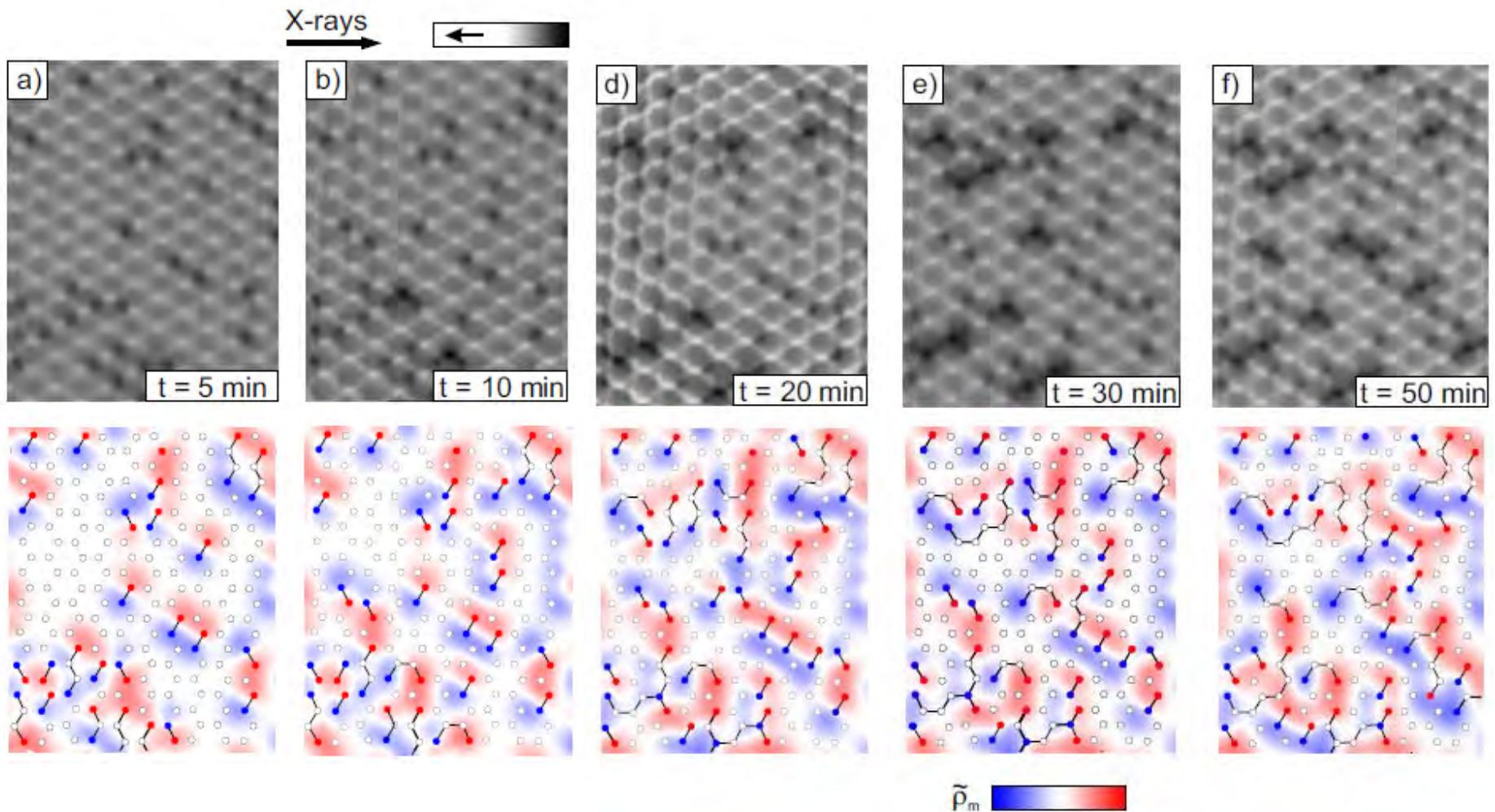
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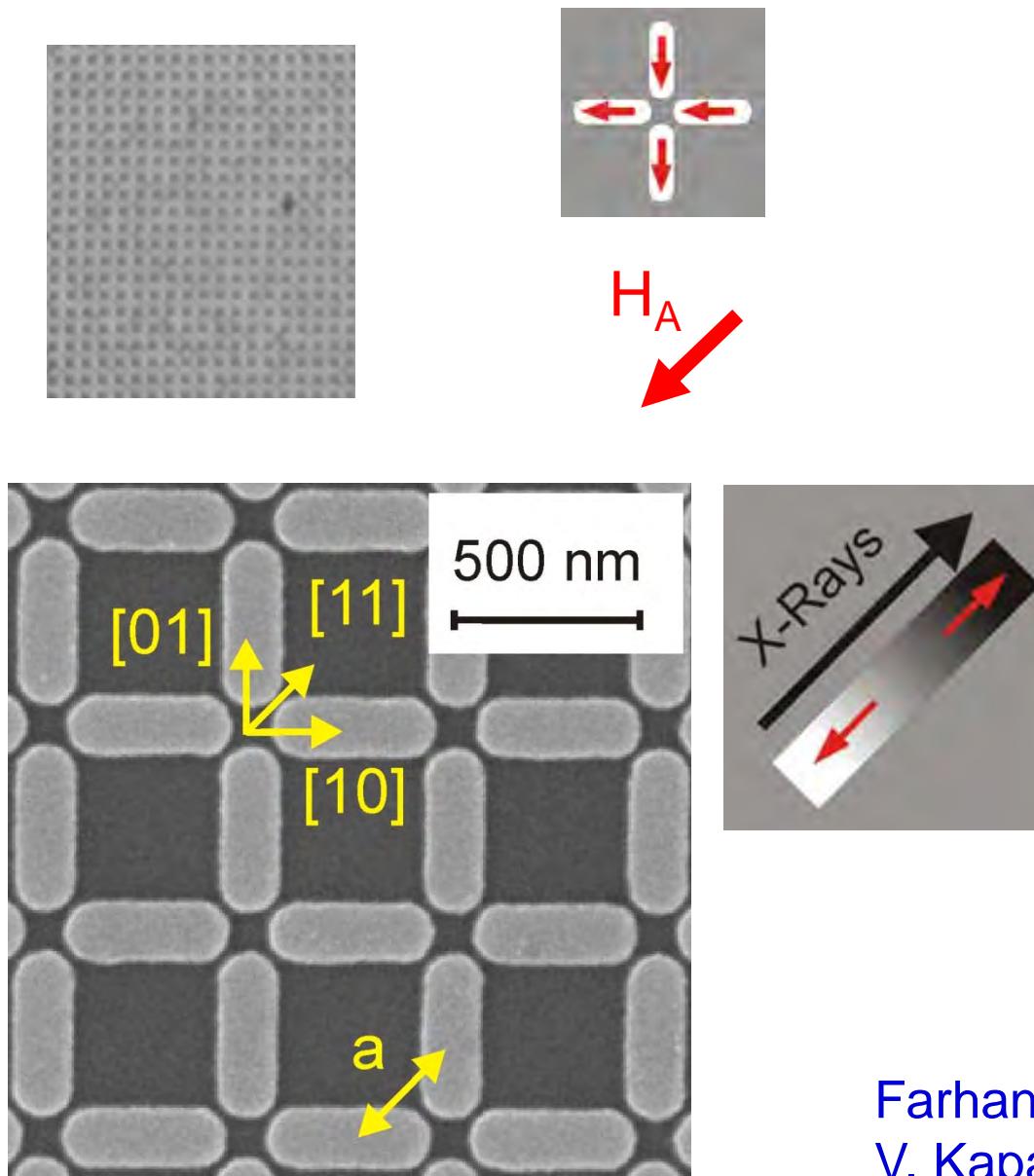


E. Mengotti, L. J. Heyderman, A. Fraile Rodríguez, F. Nolting, R.V. Hügli, H. B. Braun  
Nature Physics (2011)

# Thermally Active Artificial Kagome Ice



# Thermal Artificial Square Ice

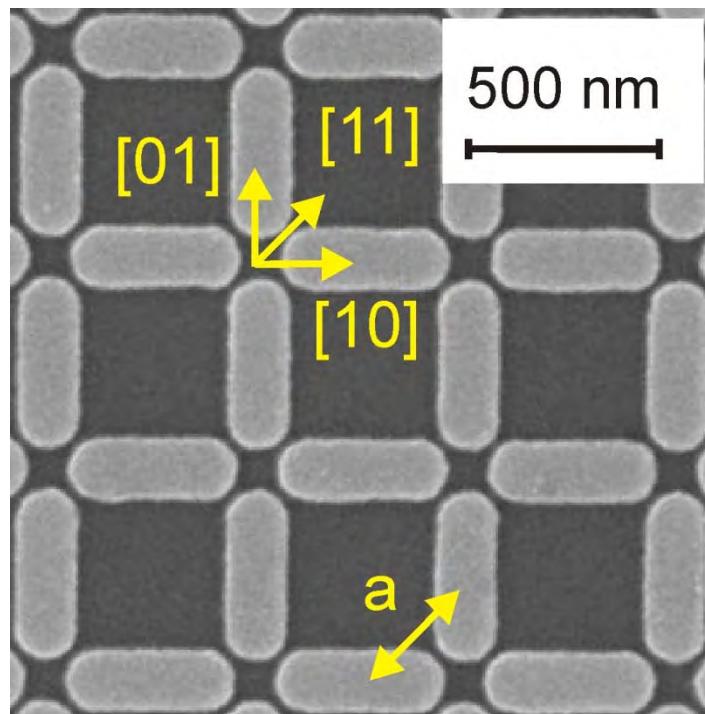
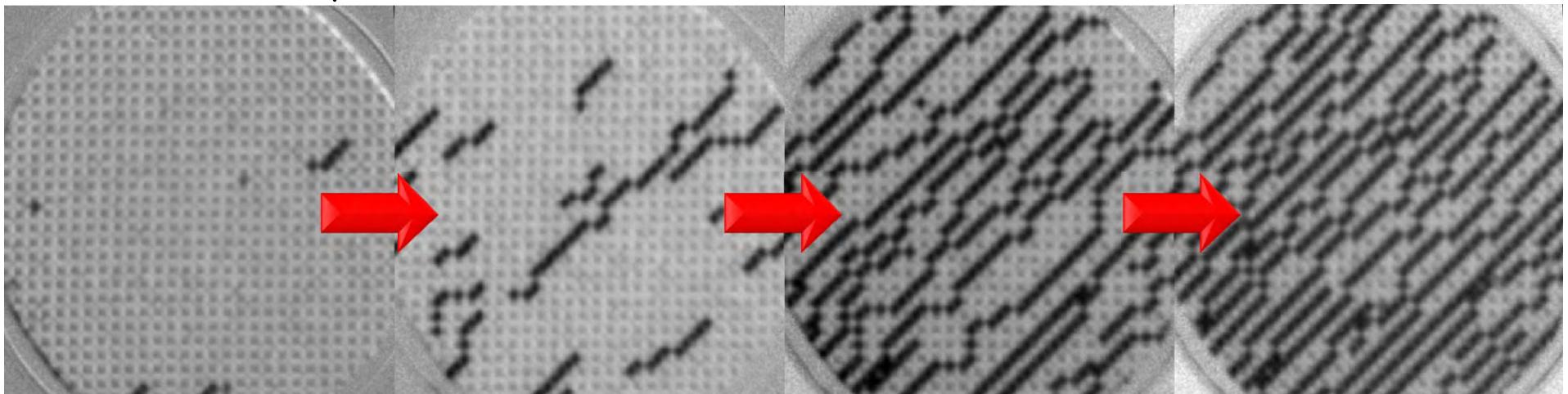


“String Regime”

Farhan et al. PRL (2013)  
V. Kapaklis et al. Nature Nanotech. (2014)

# Thermal Artificial Square Ice

Field of View 20  $\mu\text{m}$

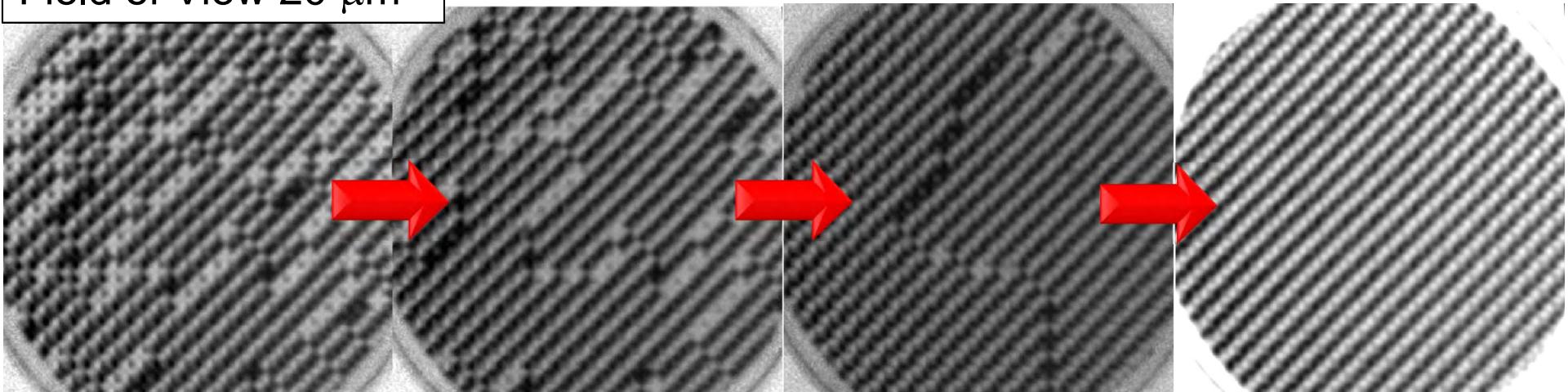


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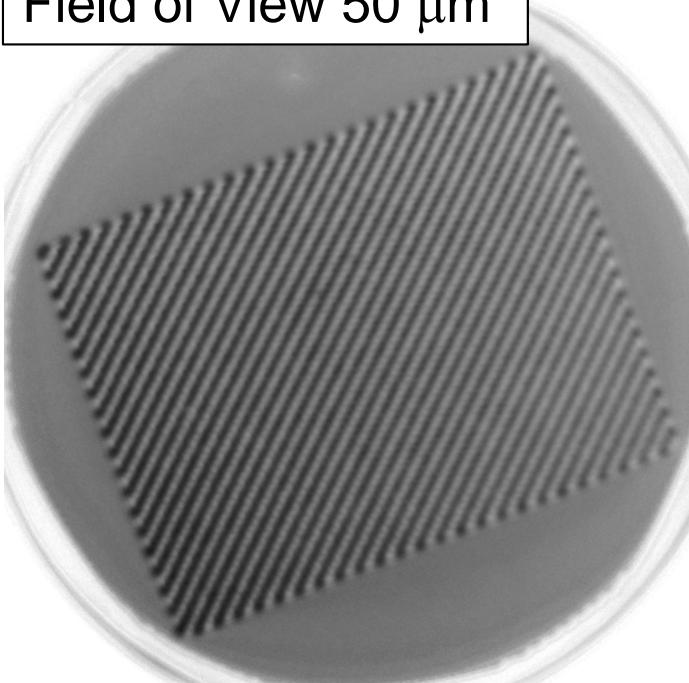
Farhan et al. PRL (2013)  
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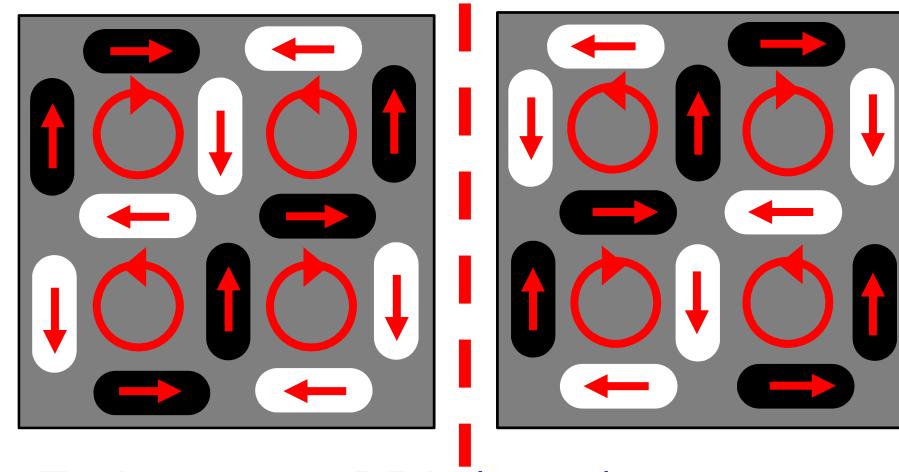
Field of View 20  $\mu\text{m}$



Field of View 50  $\mu\text{m}$



## “Domain Regime”

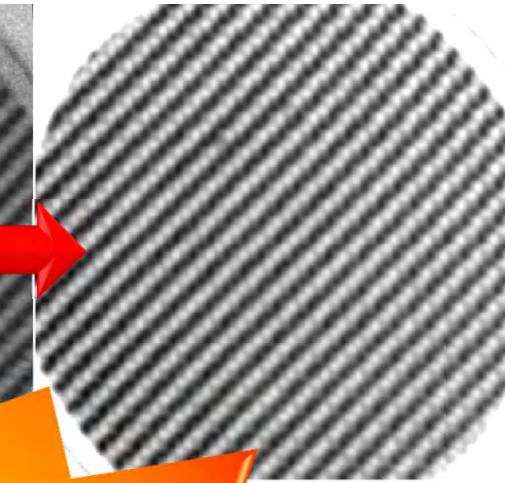
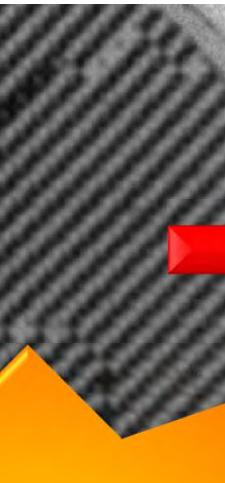
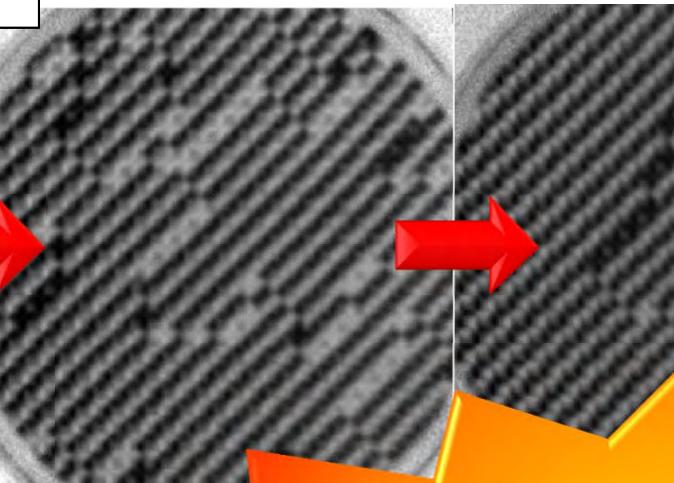
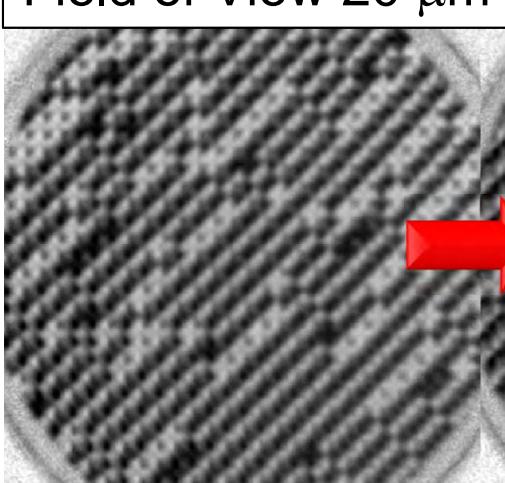


Farhan et al. PRL (2013)

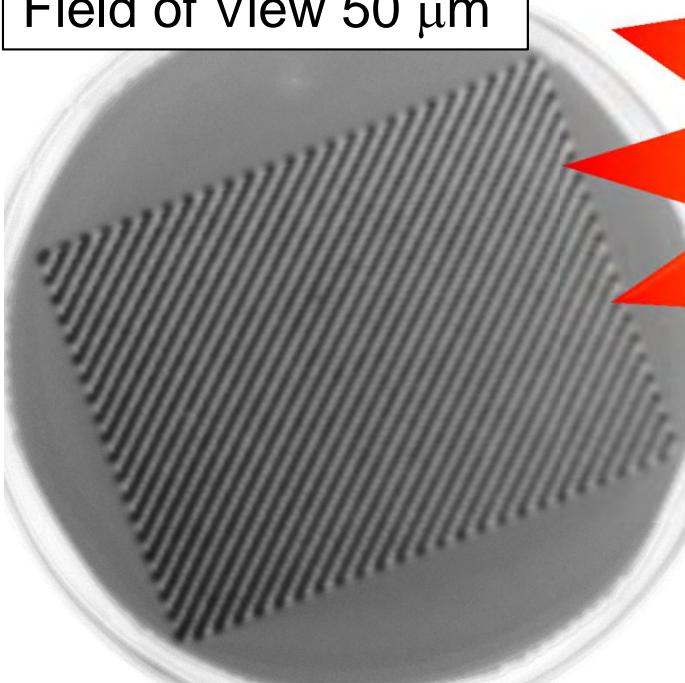
V. Kapaklis et al. Nature Nanotech. (2014)

# Thermal Artificial Square Ice

Field of View 20  $\mu\text{m}$



Field of View 50  $\mu\text{m}$

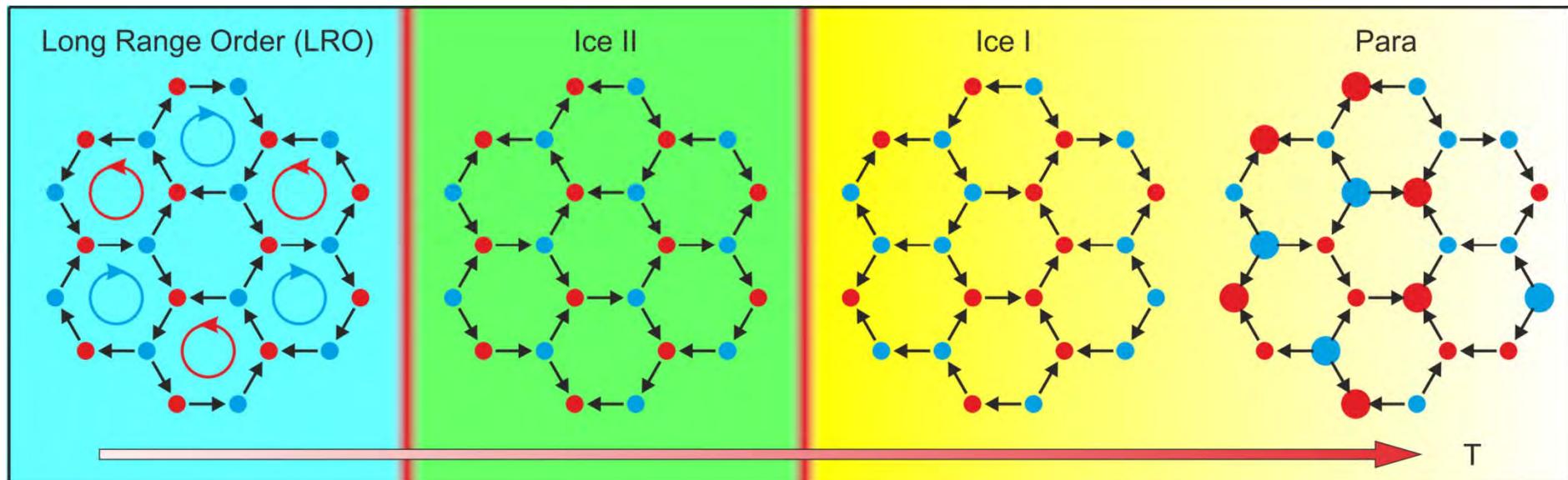


*Thermally active systems  
provide a route  
to the ground state.....*

Farhan et al. PRL (2013)

V. Kapaklis et al. Nature Nanotech. (2014)

# Kagome Spin Ice Phases



G Moller, R Moessner

*Magnetic multipole analysis of kagome and artificial spin-ice dipolar arrays*

Phys Rev B (2009)

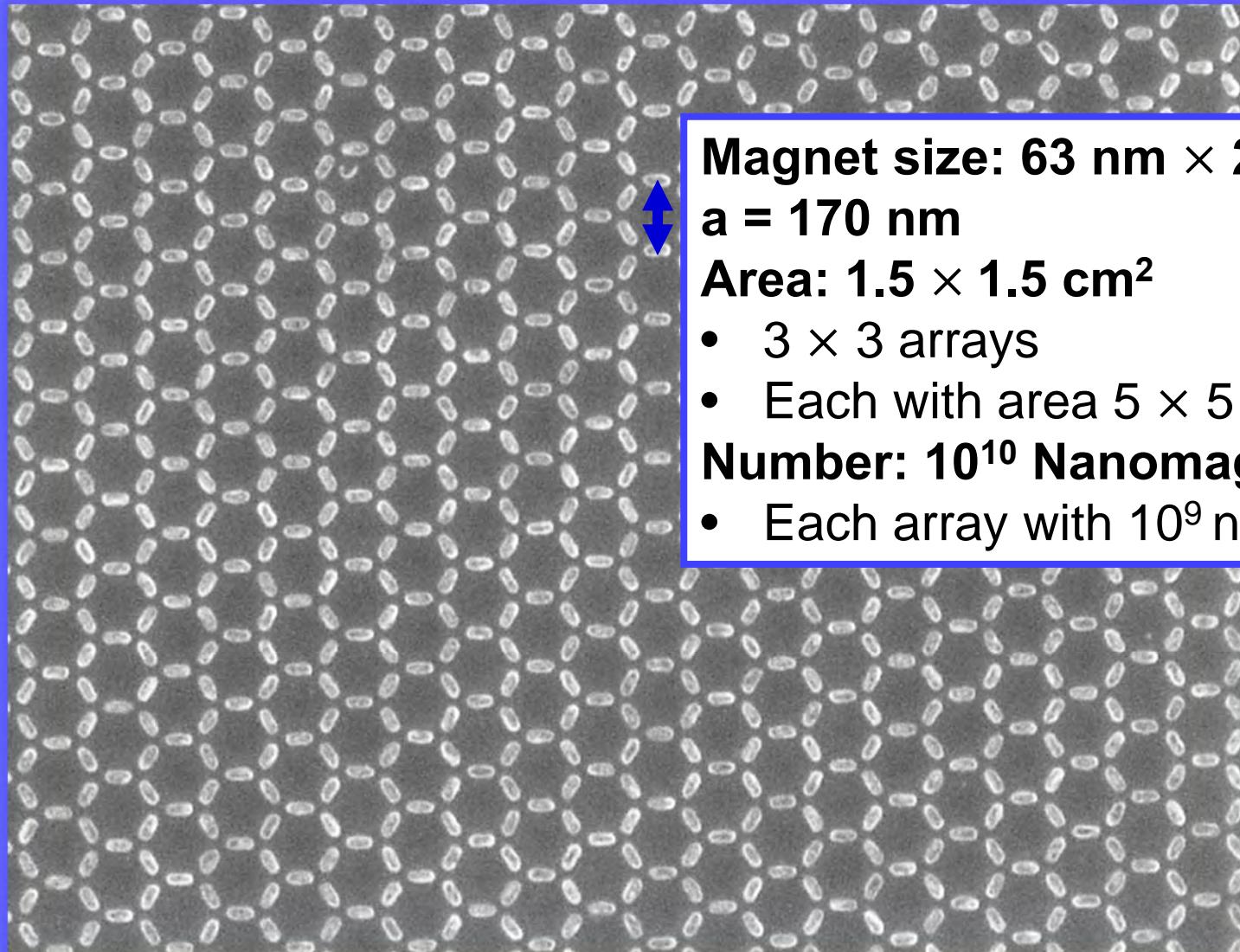
GW Chern, P Mellado, O Tchernyshyov

*Two-Stage Ordering of Spins in Dipolar Spin Ice on the Kagome Lattice*

Phys Rev Lett (2011)

L. Anghinolfi et al. Nature Communications (2015)

# Artificial Kagome Spin Ice

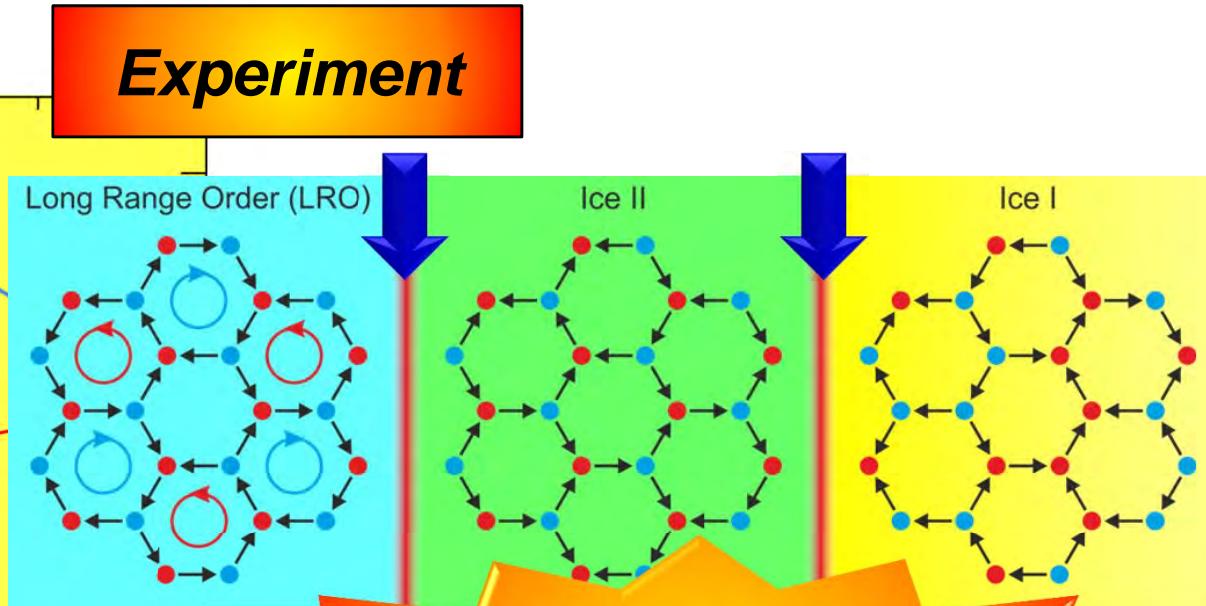
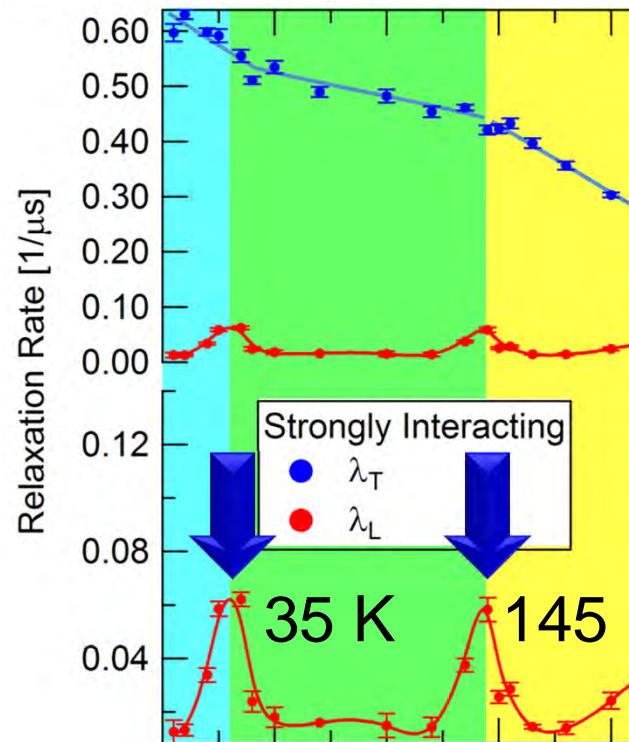
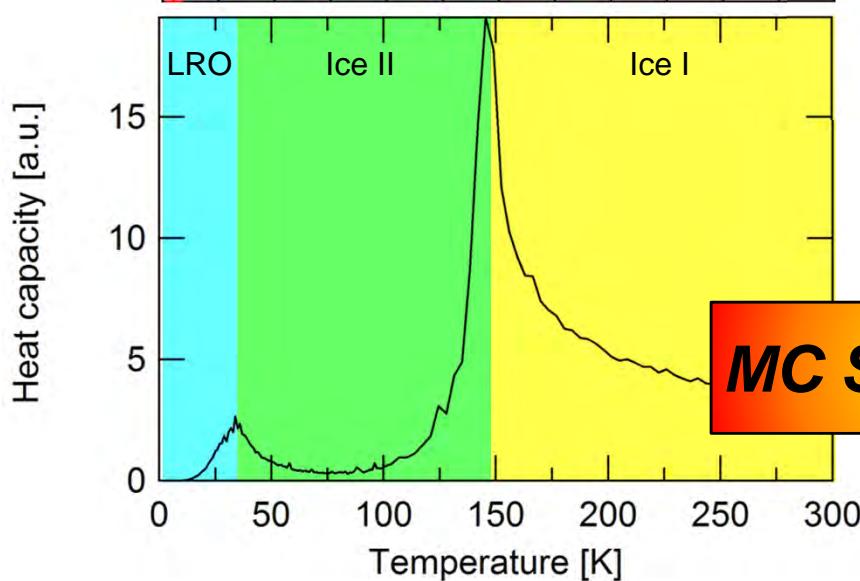


**Magnet size:**  $63 \text{ nm} \times 26 \text{ nm} \times 6 \text{ nm}$   
**a =**  $170 \text{ nm}$   
**Area:**  $1.5 \times 1.5 \text{ cm}^2$ 

- $3 \times 3$  arrays
- Each with area  $5 \times 5 \text{ mm}^2$

**Number:**  $10^{10}$  **Nanomagnets**

- Each array with  $10^9$  nanomagnets

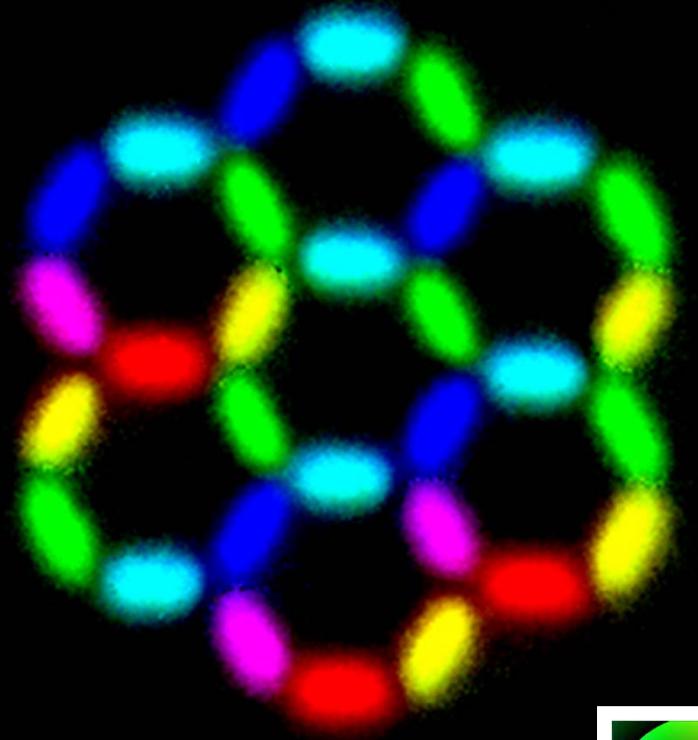
**Experiment****Frustrated Magnetic Metamaterial**

- Thermal equilibrium
- Phase transitions

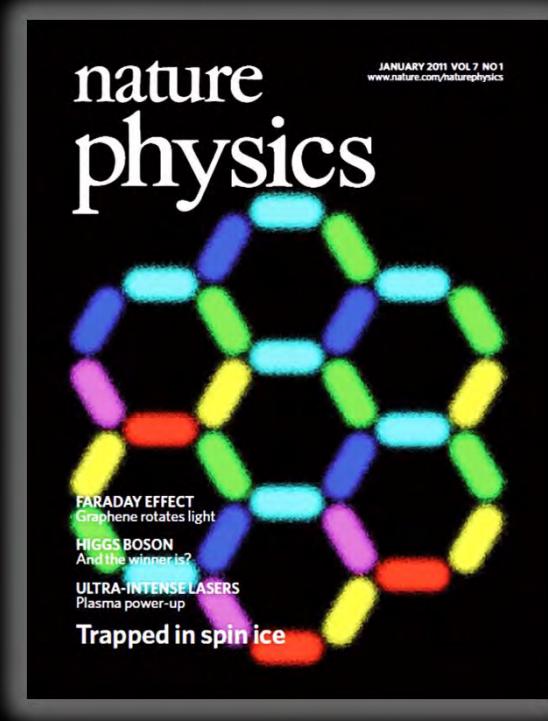
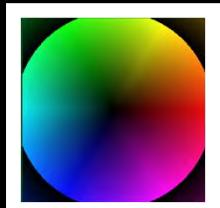
**MC Simulations**

L. Anghinolfi et al.  
Nature Communications (2015)

# Artificial Spin Ice

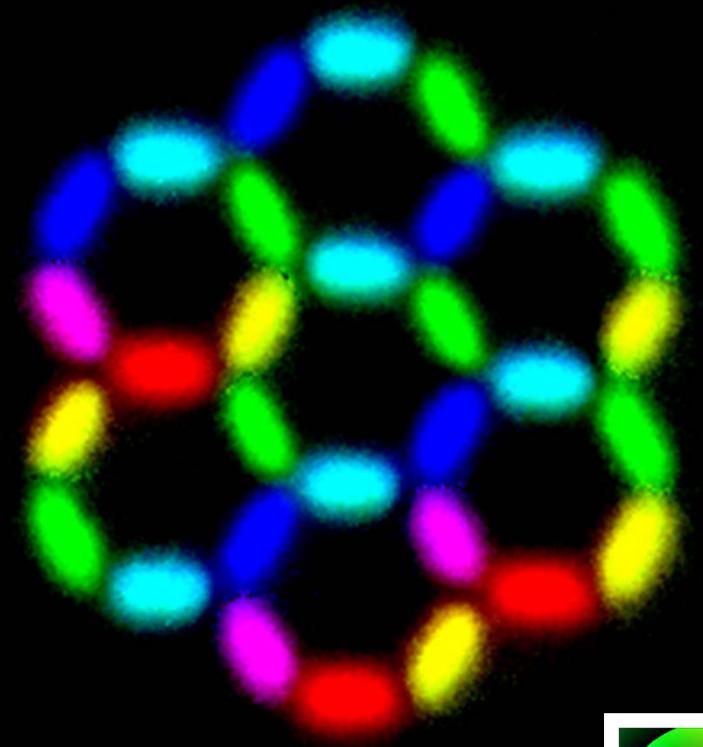


500 nm  
—

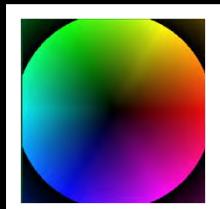


L J Heyderman & R L Stamps, **Artificial Ferroic Systems**, J. Phys. Cond. Mat. (2013)

# Artificial Spin Ice

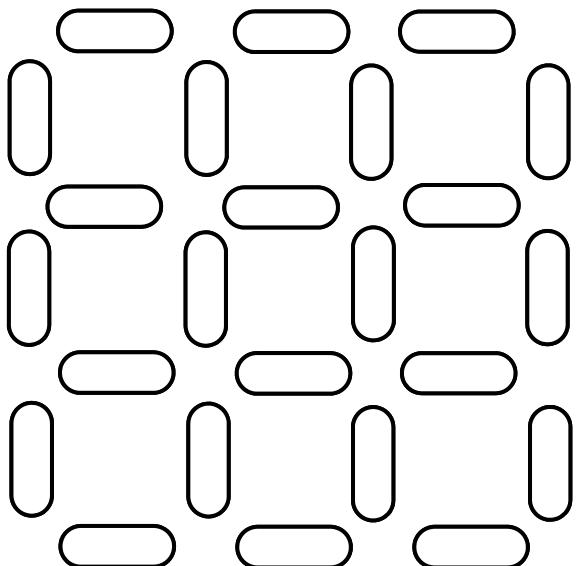


500 nm  
—



- Devices
- Materials
- Geometries
- Timescales
- Techniques

# Chiral Ice



**Square Ice → Chiral Ice**

Devices  
Materials  
Geometries  
Timescales  
Techniques

# Chiral Ice

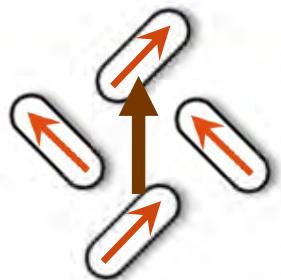


**Square Ice → Chiral Ice**

**Devices**  
**Materials**  
**Geometries**  
**Timescales**  
**Techniques**

S Gliga, G Hrkac, C Donnelly, J Büchi, A Kleibert, J Cui, A Farhan, E Kirk, R Chopdekar,  
Y Masaki, NS Bingham, A Scholl, RL Stamps, LJ Heyderman In Review (2017)

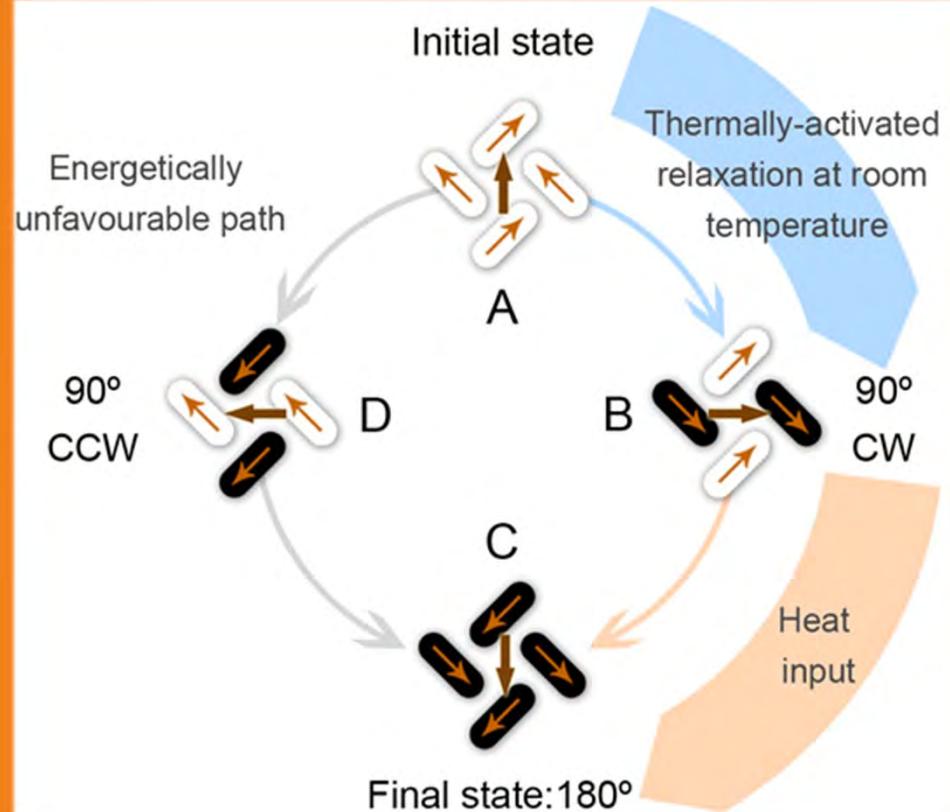
# Chiral Ice



**Square Ice → Chiral Ice**

Devices  
Materials  
Geometries  
Timescales  
Techniques

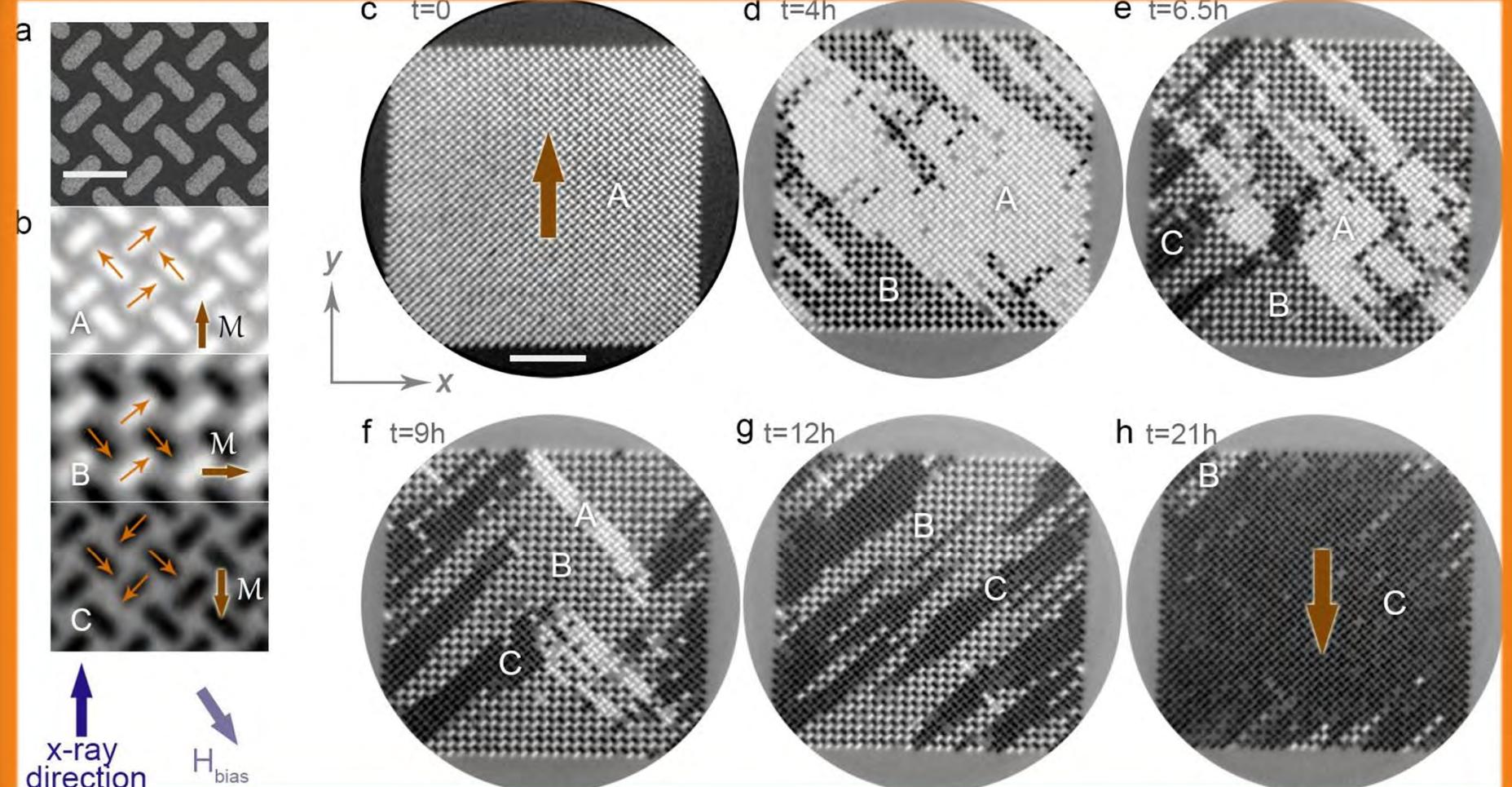
# Chiral Ice



Devices  
Materials  
Geometries  
Timescales  
Techniques

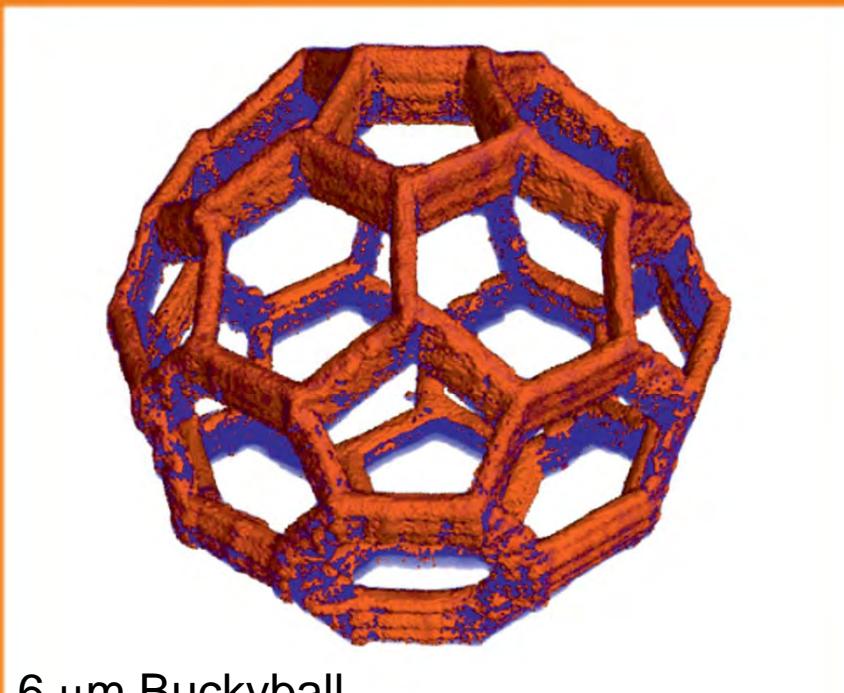
S Gliga, G Hrkac, C Donnelly, J Büchi, A Kleibert, J Cui, A Farhan, E Kirk, R Chopdekar, Y Masaki, NS Bingham, A Scholl, RL Stamps, LJ Heyderman In Review (2017)

# Chiral Ice



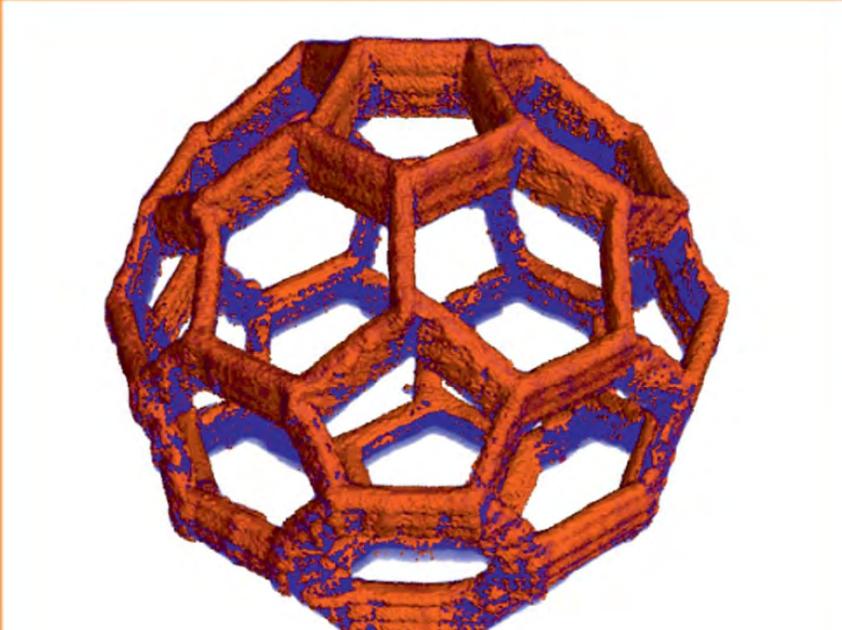
S Gliga, G Hrkac, C Donnelly, J Büchi, A Kleibert, J Cui, A Farhan, E Kirk, R Chopdekar, Y Masaki, NS Bingham, A Scholl, RL Stamps, LJ Heyderman In Review (2017)

# Artificial Spin Ice – Three Dimensions



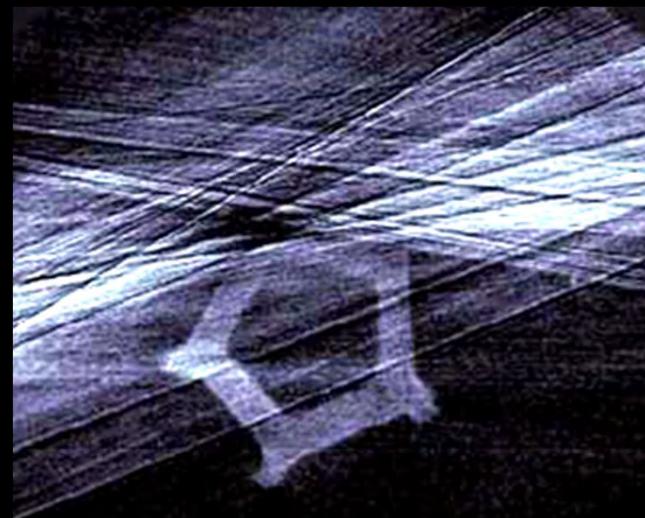
Devices  
Materials  
→ Geometries  
Timescales  
Techniques

# Resonant Ptychographic Tomography



6  $\mu\text{m}$  Buckyball

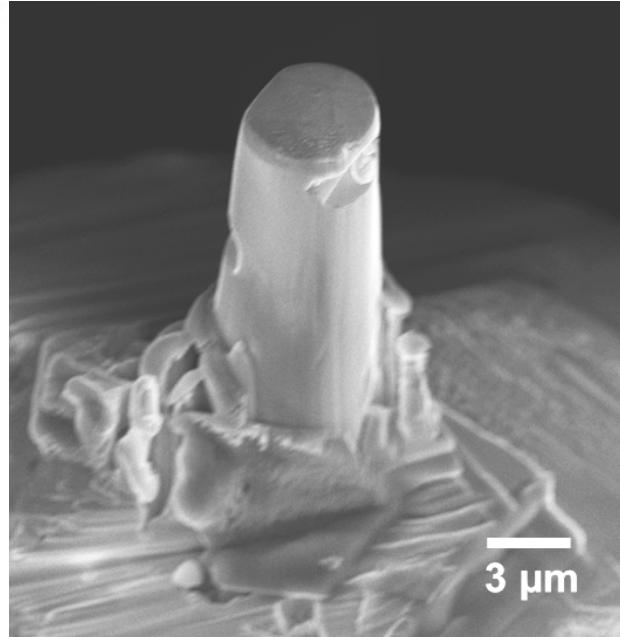
C. Donnelly et al. PRL (2015)



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

L J Heyderman & R L Stamps, **Artificial Ferroic Systems**, J. Phys. Cond. Mat. (2013)

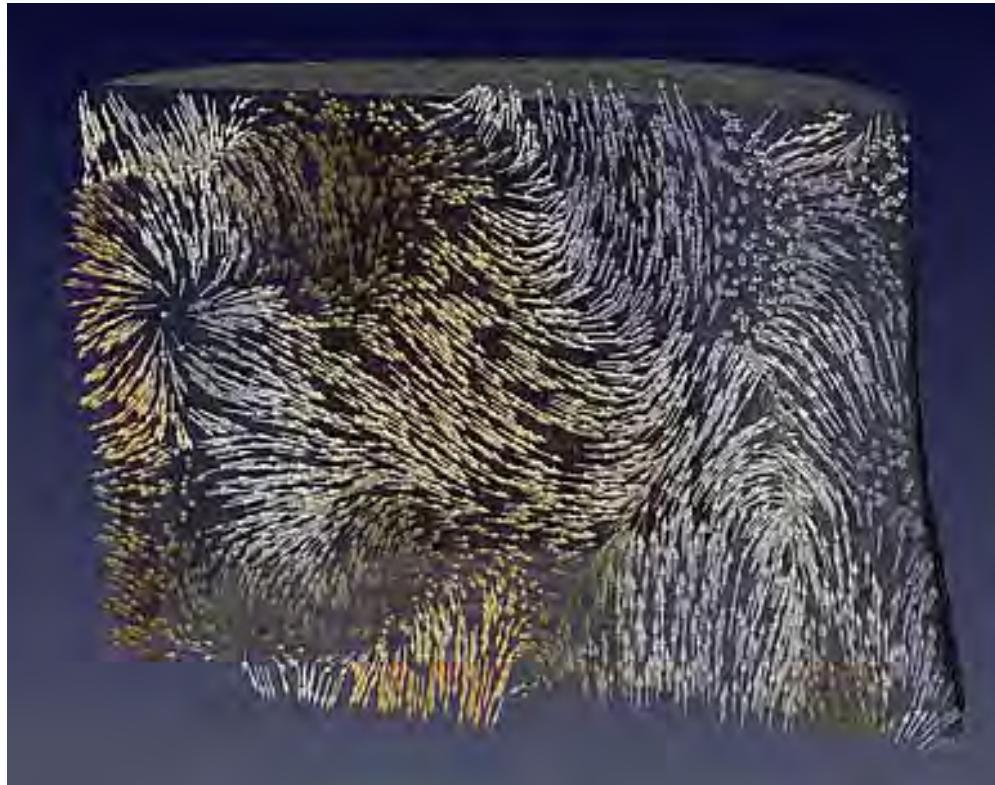
# Hard X-ray Magnetic Tomography



**GdCo<sub>2</sub> Pillar**

Cut from nugget with FIB

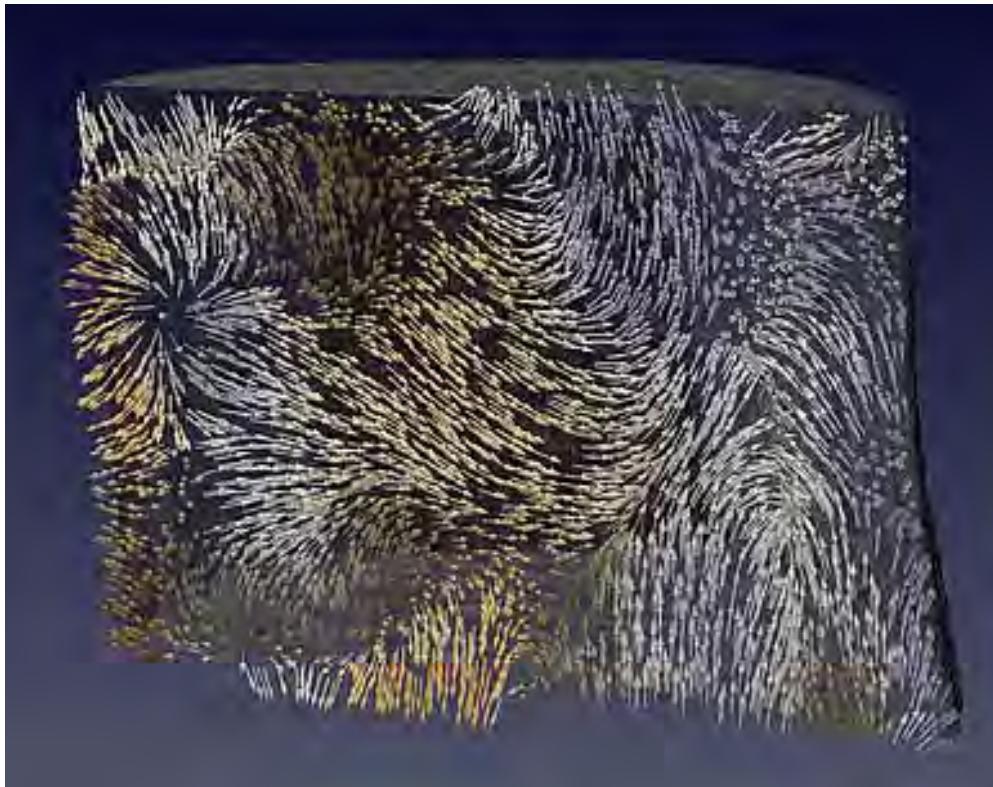
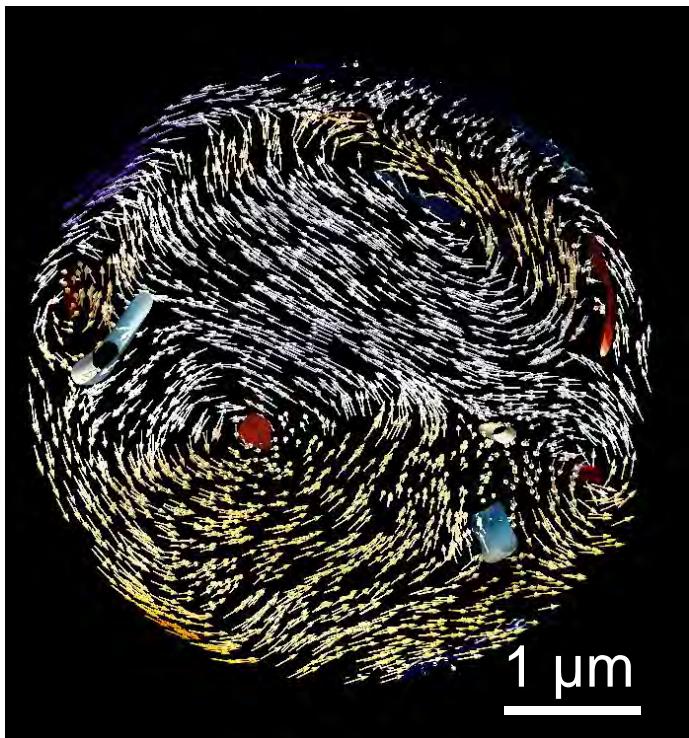
Sample from:  
R. Galera, CNRS, Grenoble



- C. Donnelly et al. Nature (2017)  
P. Fischer, *News & Views*, Nature (2017)  
C. Donnelly et al. PRB (2016)

# Hard X-ray Magnetic Tomography

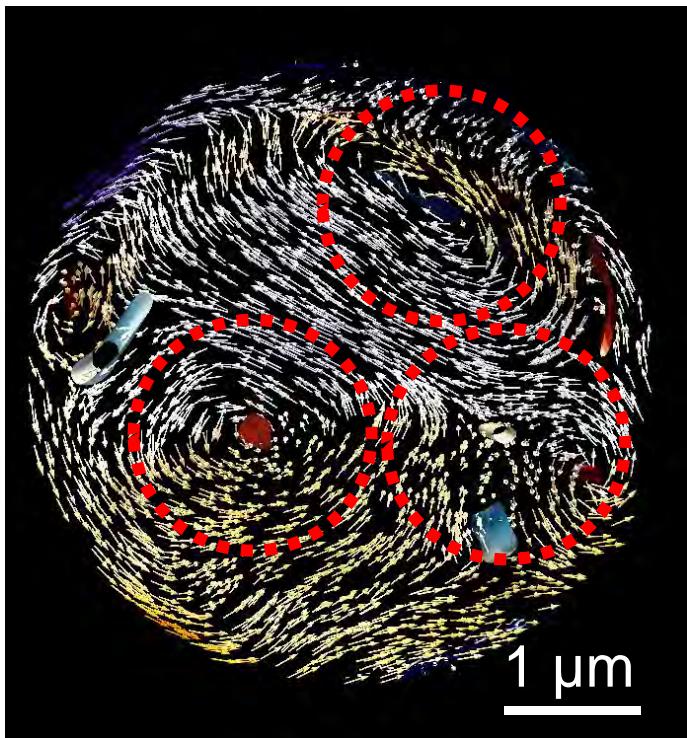
One slice:



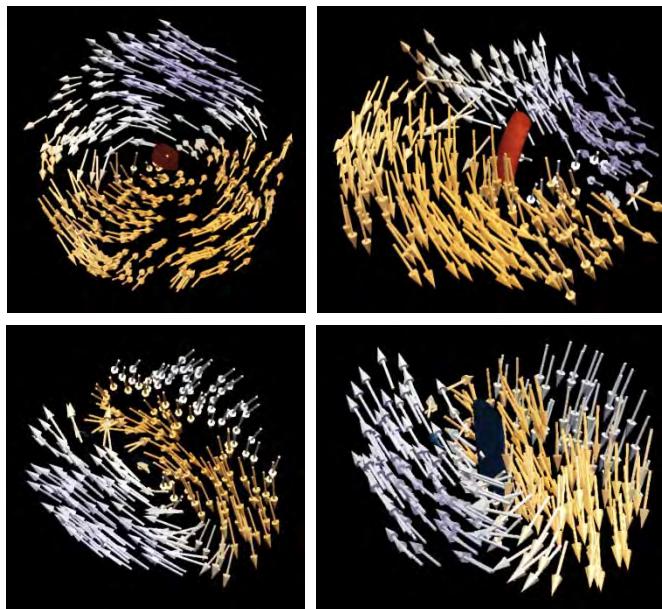
- C. Donnelly et al. Nature (2017)  
P. Fischer, *News & Views*, Nature (2017)  
C. Donnelly et al. PRB (2016)

# Hard X-ray Magnetic Tomography

C. Donnelly et al. Nature (2017)  
P. Fischer, *News & Views*,  
Nature (2017)  
C. Donnelly et al. PRB (2016)



## Vortex

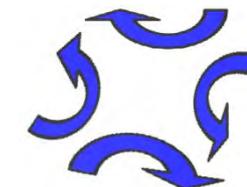


Clockwise

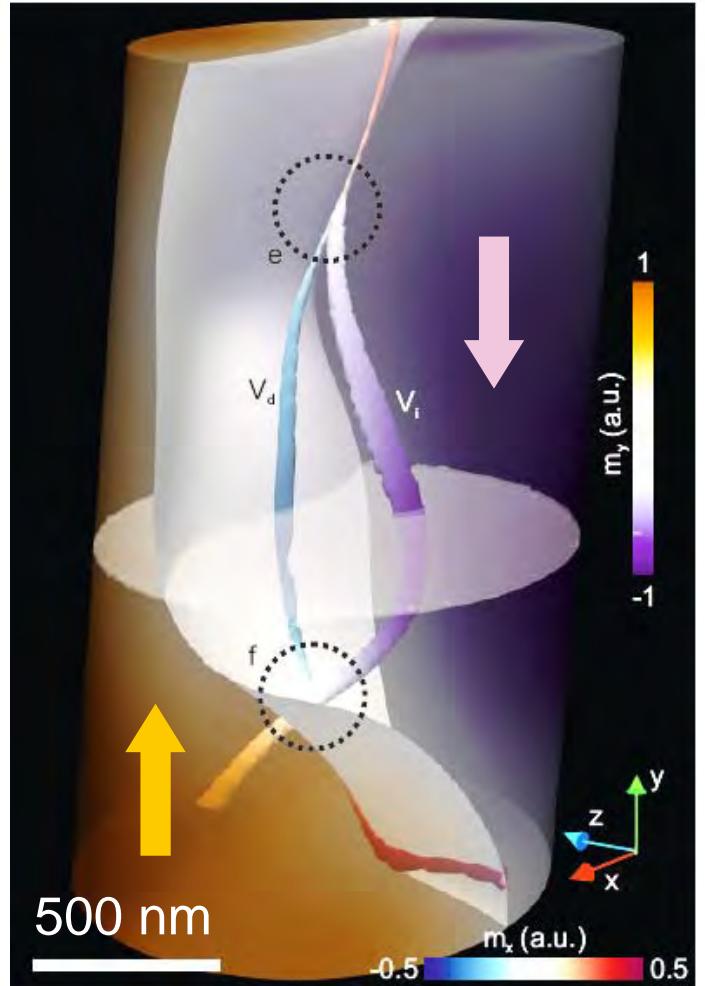


Anticlockwise

## Antivortex



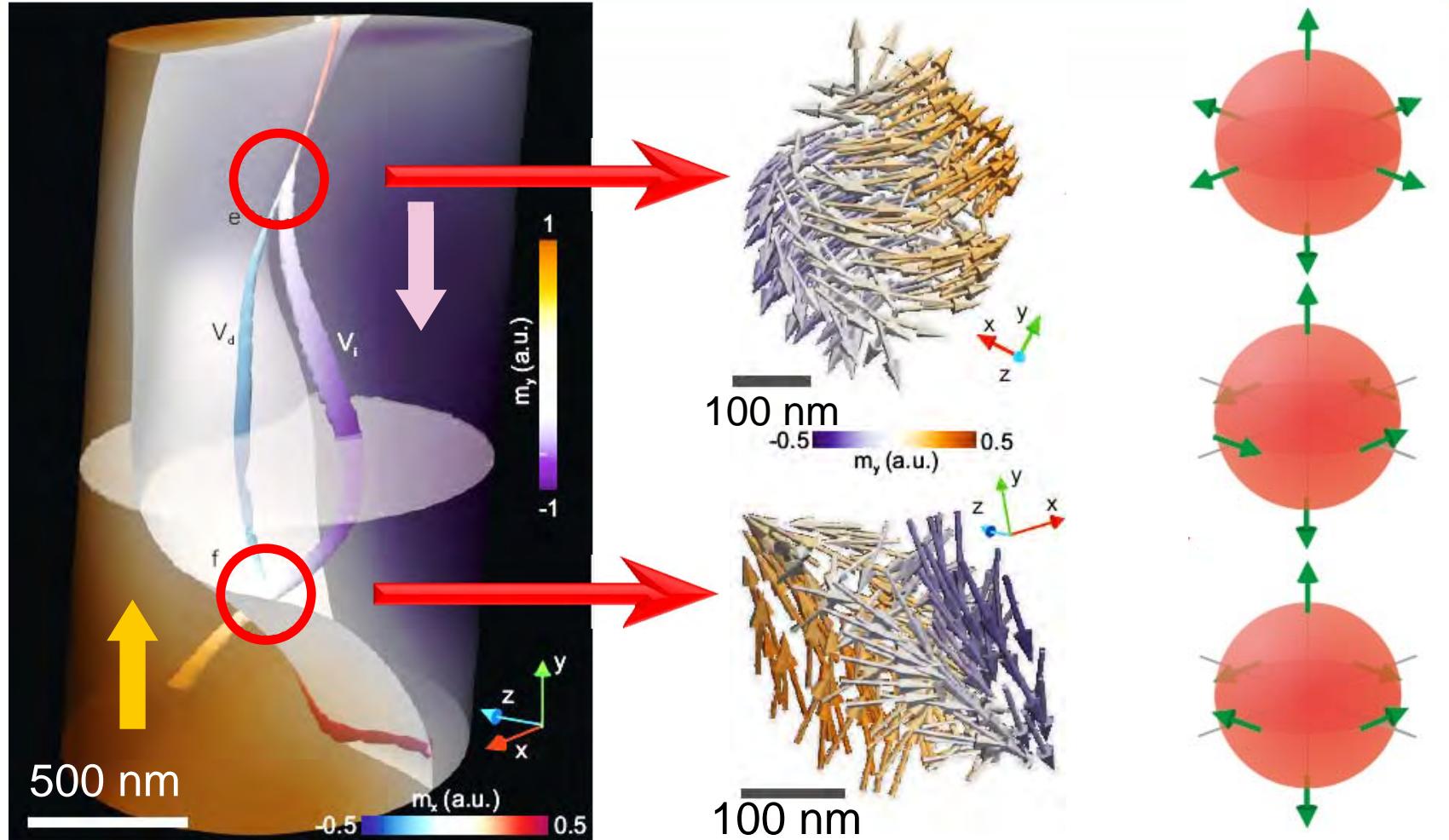
# Hard X-ray Magnetic Tomography



C. Donnelly et al. *Nature* (2017); P. Fischer, *News & Views, Nature* (2017)

C. Donnelly et al. *PRB* (2016)

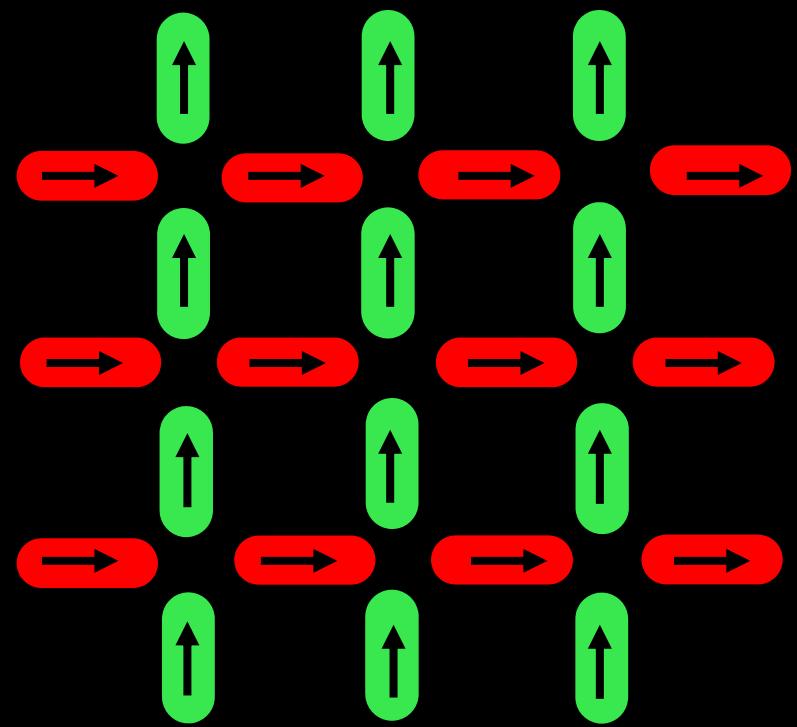
# Hard X-ray Magnetic Tomography



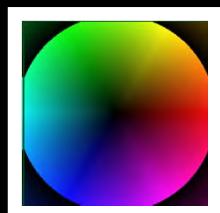
C. Donnelly et al. Nature (2017); P. Fischer, *News & Views*, Nature (2017)

C. Donnelly et al. PRB (2016)

# Artificial Spin Ice – Square Lattice

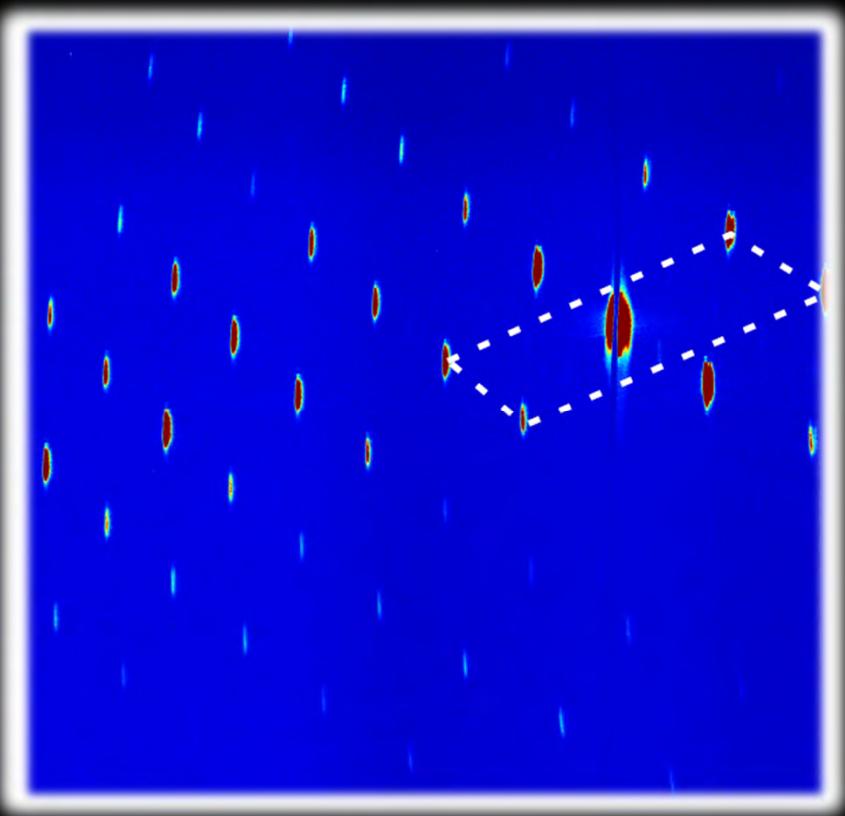
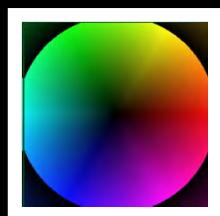
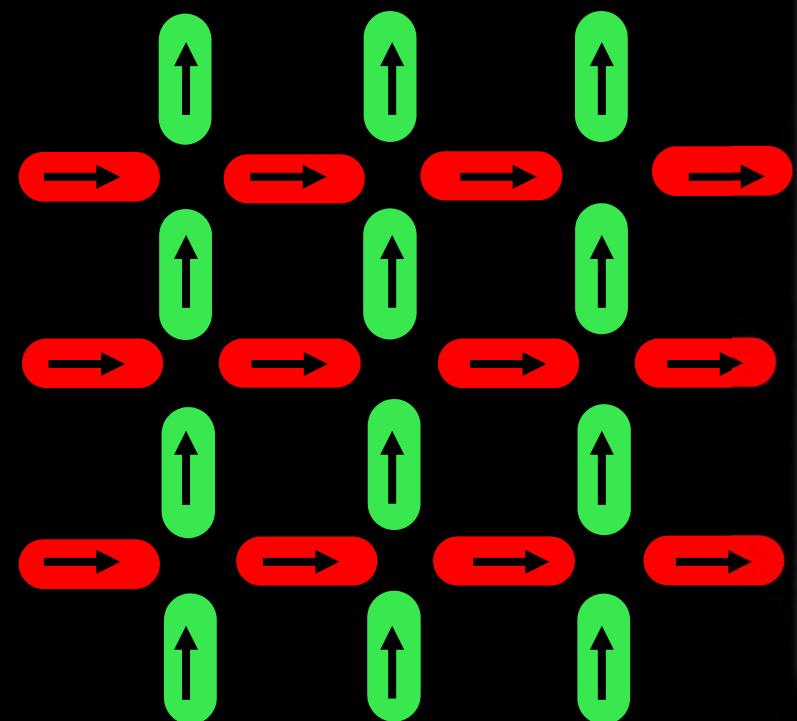


Devices  
Materials  
Geometries  
Timescales  
Techniques



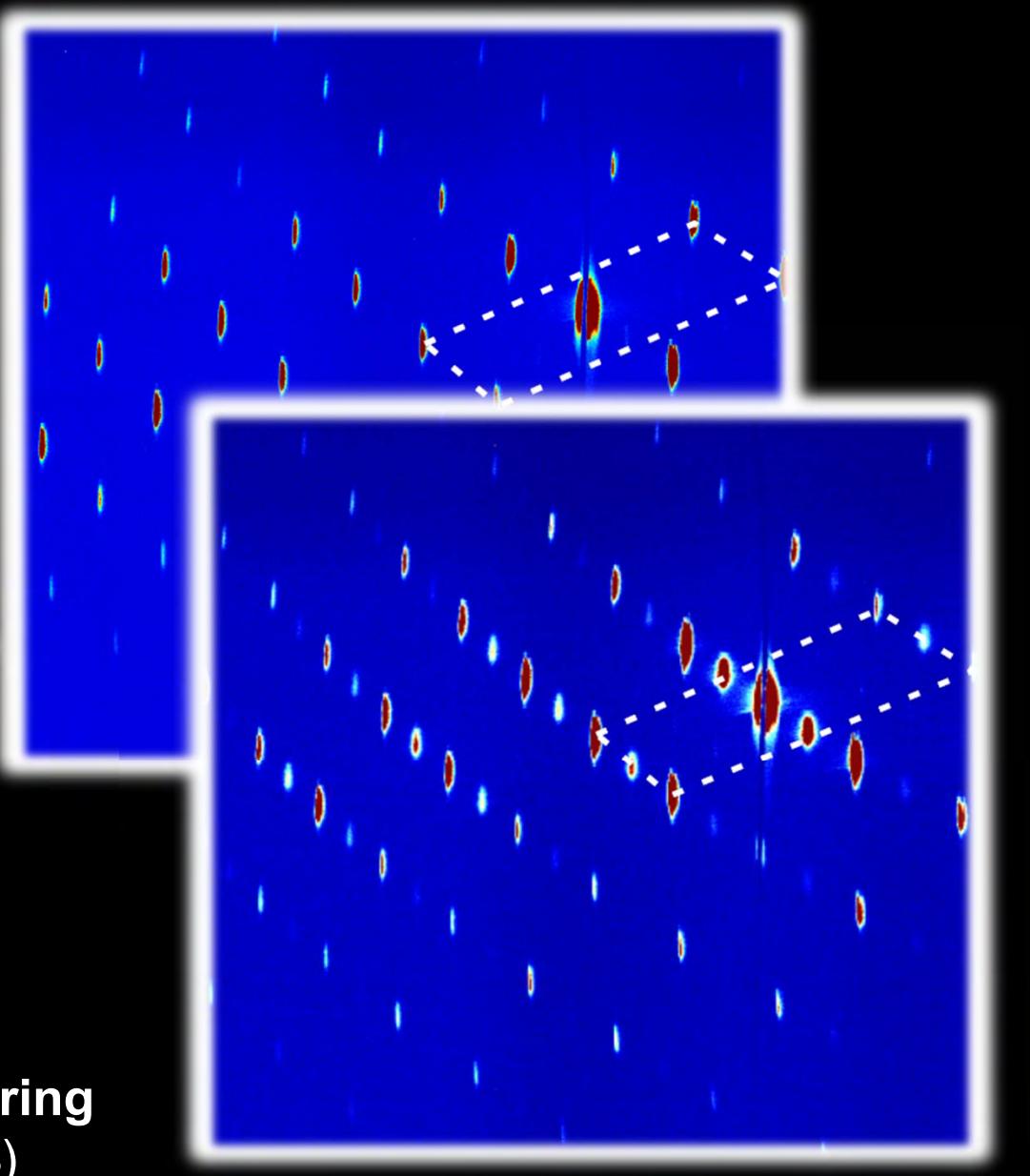
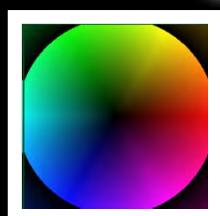
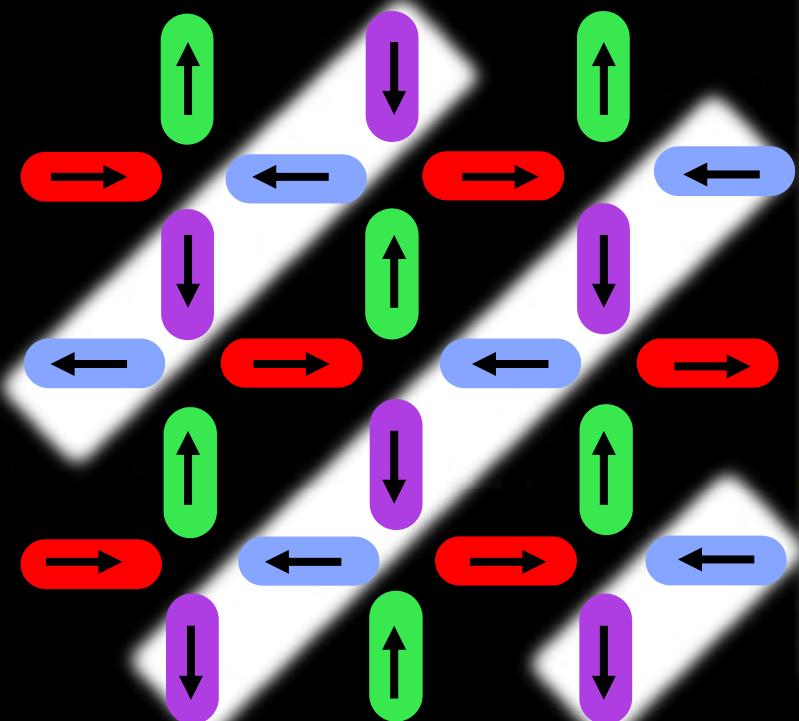
**X-ray Magnetic Resonant Scattering**  
J. Perron et al. Phys. Rev. B (2013)

# Artificial Spin Ice – Square Lattice



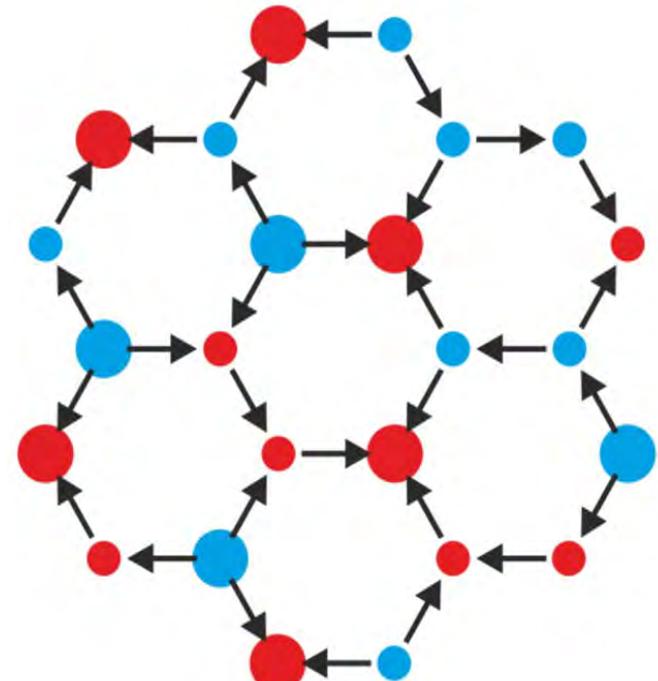
**X-ray Magnetic Resonant Scattering**  
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# Artificial Spin Ice – Square Lattice

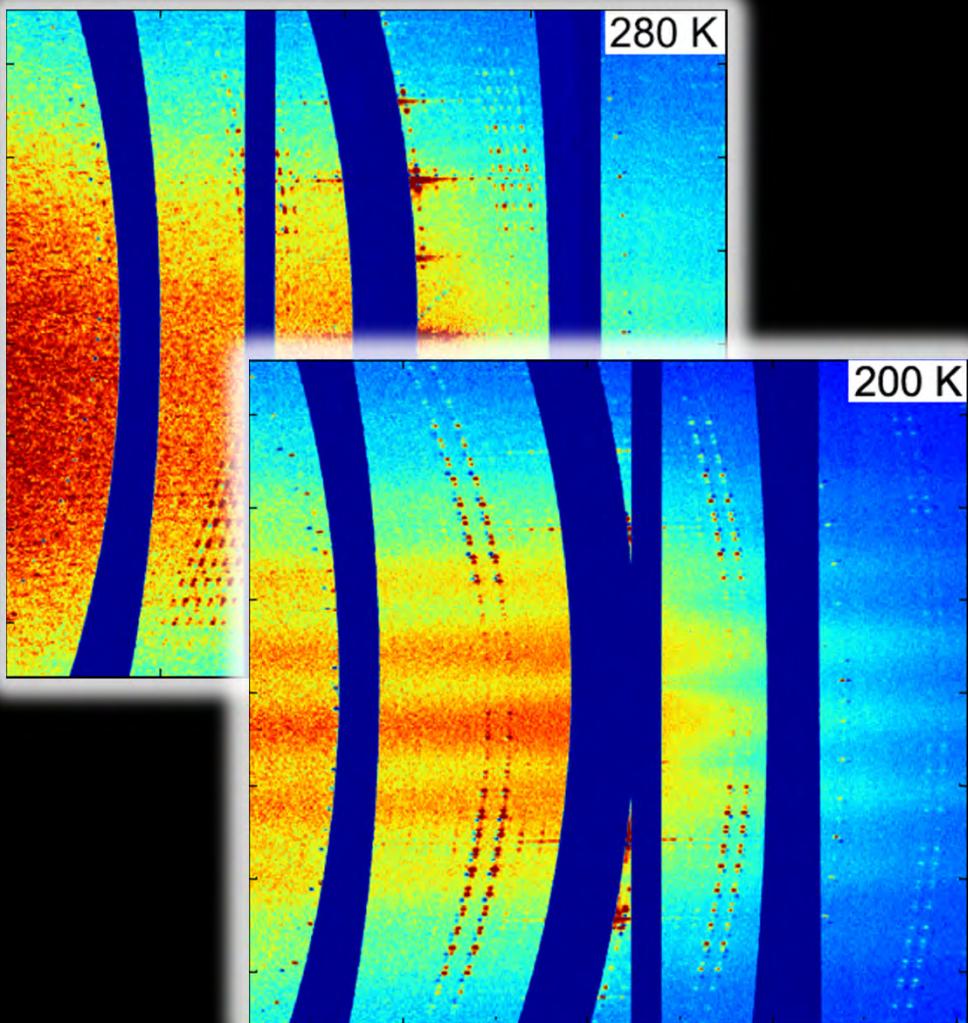


**X-ray Magnetic Resonant Scattering**  
J. Perron et al. Phys. Rev. B (2013)

# Artificial Spin Ice – Kagome Lattice



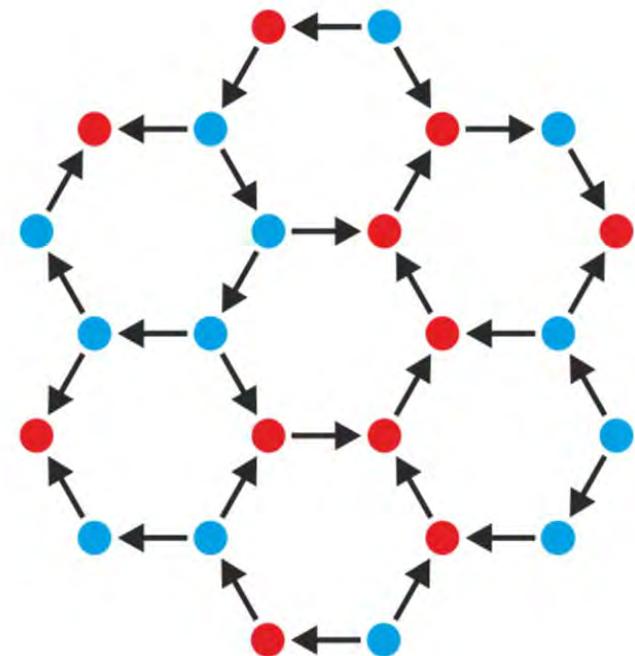
*Paramagnetic*



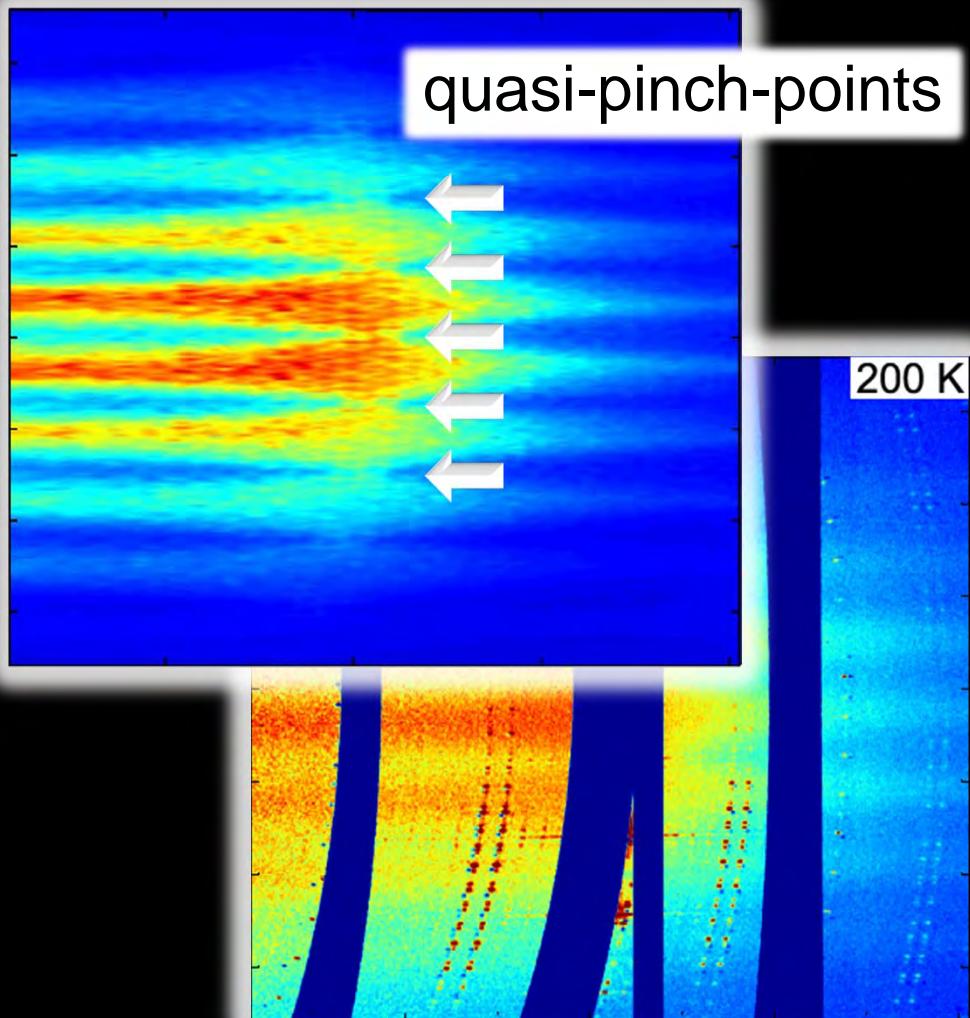
## X-ray Magnetic Resonant Diffuse Scattering

O Sendetskyi, L Anghinolfi, V Scagnoli, G Möller, N Leo, A Alberca, J Kohlbrecher, J Lüning, U Staub, LJ Heyderman Phys. Rev. B (2016)

# Artificial Spin Ice – Kagome Lattice



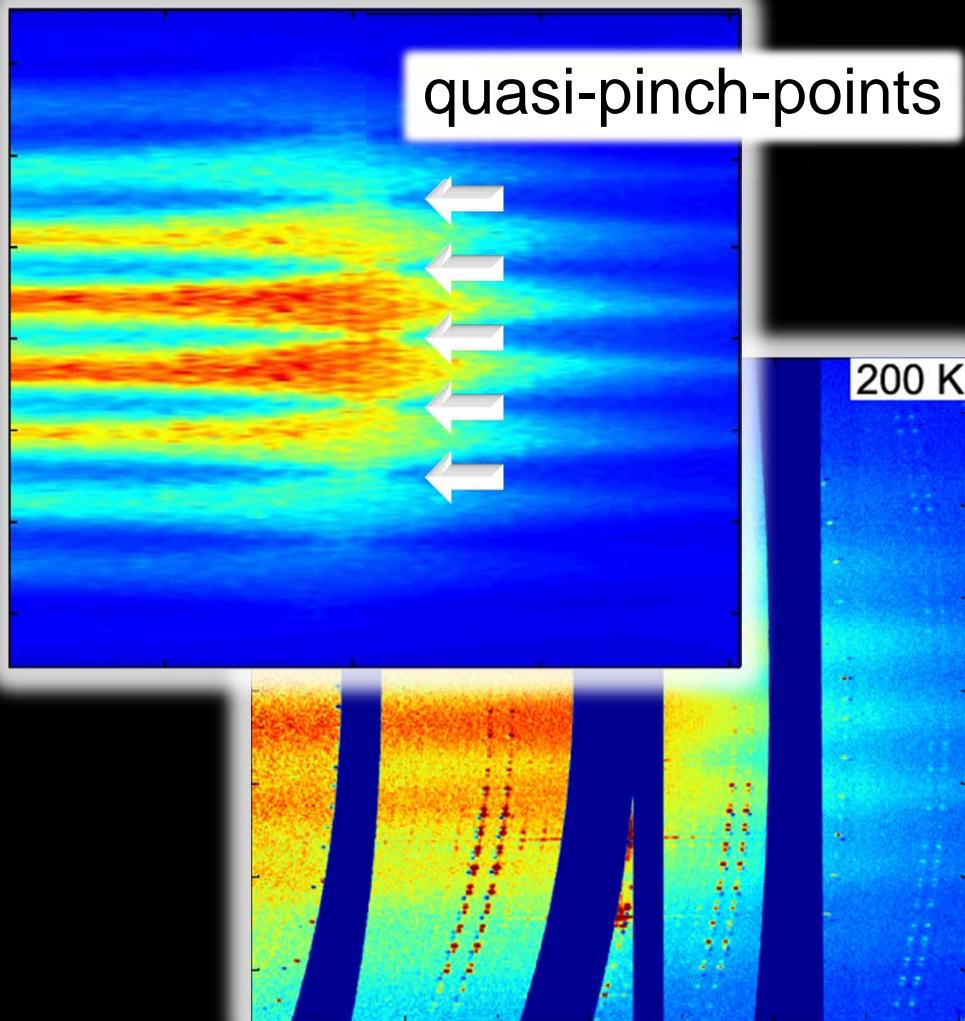
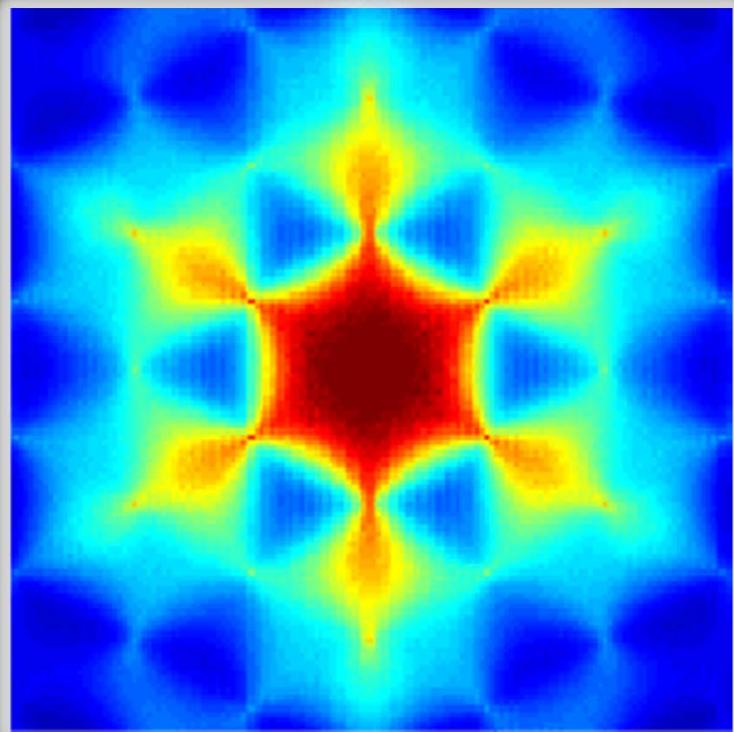
*Kagome Ice I*



## X-ray Magnetic Resonant Diffuse Scattering

O Sendetskyi, L Anghinolfi, V Scagnoli, G Möller, N Leo, A Alberca, J Kohlbrecher, J Lüning, U Staub, LJ Heyderman Phys. Rev. B (2016)

# Artificial Spin Ice – Kagome Lattice



## X-ray Magnetic Resonant Diffuse Scattering

O Sendetskyi, L Anghinolfi, V Scagnoli, G Möller, N Leo, A Alberca, J Kohlbrecher, J Lüning, U Staub, LJ Heyderman Phys. Rev. B (2016)

# **Artificial Spin Ice**

Laura Heyderman

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

***Mesoscopic Systems (ETH Zurich – Paul Scherrer Institute)***

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***Swiss Light Source***

*Photoemission Electron Microscopy:* Armin Kleibert, Ana Balan, Arantxa Fraile Rodriguez, Loic Le Guyader, Frithjof Nolting

*X-ray Scattering:* Aurora Alberca, Joachim Kohlbrecher, Urs Staub

*Hard X-ray Tomography & Imaging:* Manuel Guizar Sicairos, Andreas Menzel, Joerg Raabe, Mirko Holler, Elisabeth Müller, Thomas Huthwelker

***Condensed Matter Theory Group:*** Peter Derlet

***Laboratory for Muon Spin Spectroscopy:*** Hubertus Luetkens, Andreas Suter, Thomas Prokscha

# Artificial Spin Ice

Laura Heyderman

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Gino Hrkac (Exeter), Robert Stamps (Glasgow), Yusuke Masaki (Tokyo)

***Univ. St Andrews:***

Machiel Flokstra, Steve Lee

***UC Dublin:***

Remo Hügli, Gerard Duff,  
Hans-Benjamin Braun

***Univ. Cambridge:***

Gunnar Möller

***Uppsala University:***

Vassilios Kapaklis, Unnar Arnalds,  
Björgvin Hjörvarsson

***SOLEIL Synchrotron:***

Nicolas Jaouen, Jean-Marc Tonnerre,  
Jan Lüning, Bharati Tudu,  
Maurizio Sacchi

***ESRF Synchrotron:***

Fabrice Wilhelm, Francois Guillou,  
Andrei Rogalev, Carsten Detlefs

***Adv. Light Source:***

Andreas Scholl, Tony Young

# *Artificial Ferroic Systems*

Laura Heyderman

**HYBRID  
SYSTEMS**

**ARTIFICIAL  
SPIN  
SYSTEMS**

**3D**

## **Hybrid Systems – Artificial Multiferroics**

- ✿ Strain imprints magnetic states
- ✿ Cost effective nanoparticle dispersions
- ✿ Control:
  - ✿ with structural phase transitions of cap layer
  - ✿ magnetic state with electric field

## **Artificial Spin Ice**

- ✿ Emergent magnetic monopoles & Dirac strings
- ✿ Thermally active artificial spin ice:
  - ✿ Switching in kagome spin ice structures
  - ✿ Thermal relaxation in square ice
  - ✿ Phase Transitions → Frustrated Magnetic Metamaterial

