Artificial spin ice: Frustration by design

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Funding: ARO, NSF

OUTLINE

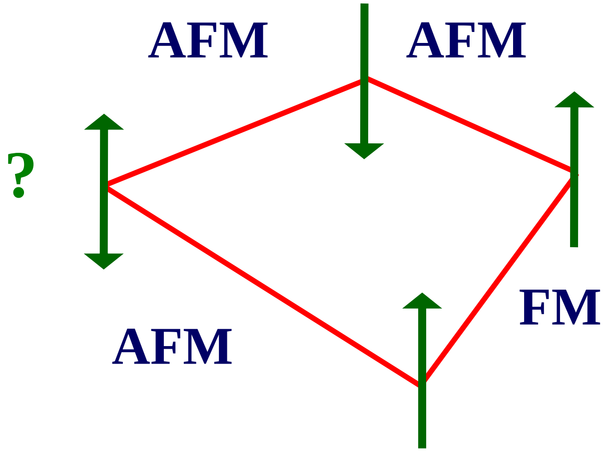
Introduction to frustration and spin ice

1. **How does spin ice freeze?**
2. **New class of materials: Stuffed Spin Ice**
3. **New way to be frustrated: Artificial Spin Ice**

Frustration

The common case of disordered magnets:

Random FM and AFM interactions

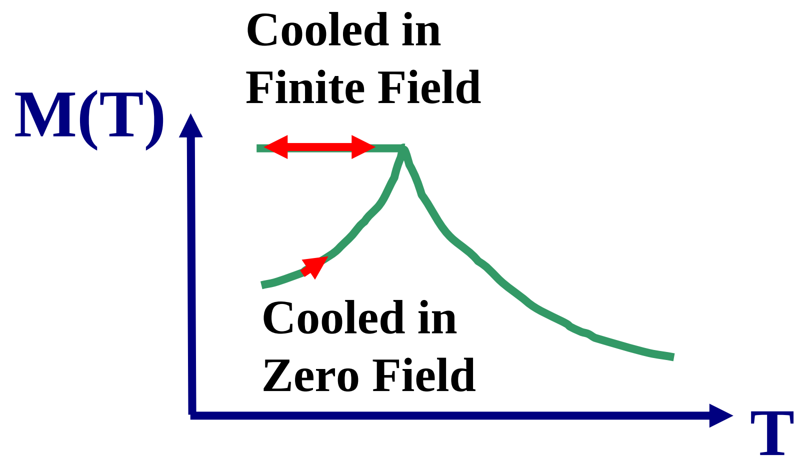


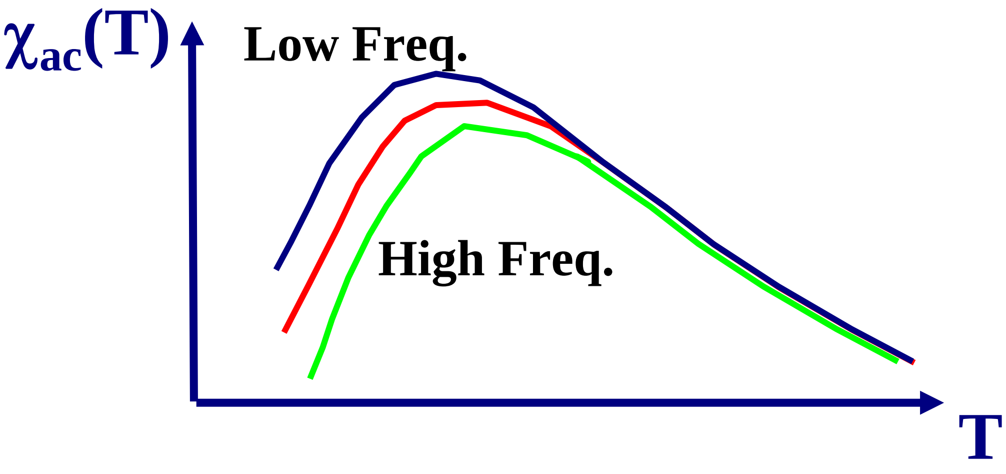
__Generic definition of Frustration:__ A system's inability to simultaneously minimize all of the interaction energies between its components resulting in multiple ground states

Disorder and frustration cause spins to freeze in random configuration at low T → “Spin Glass”

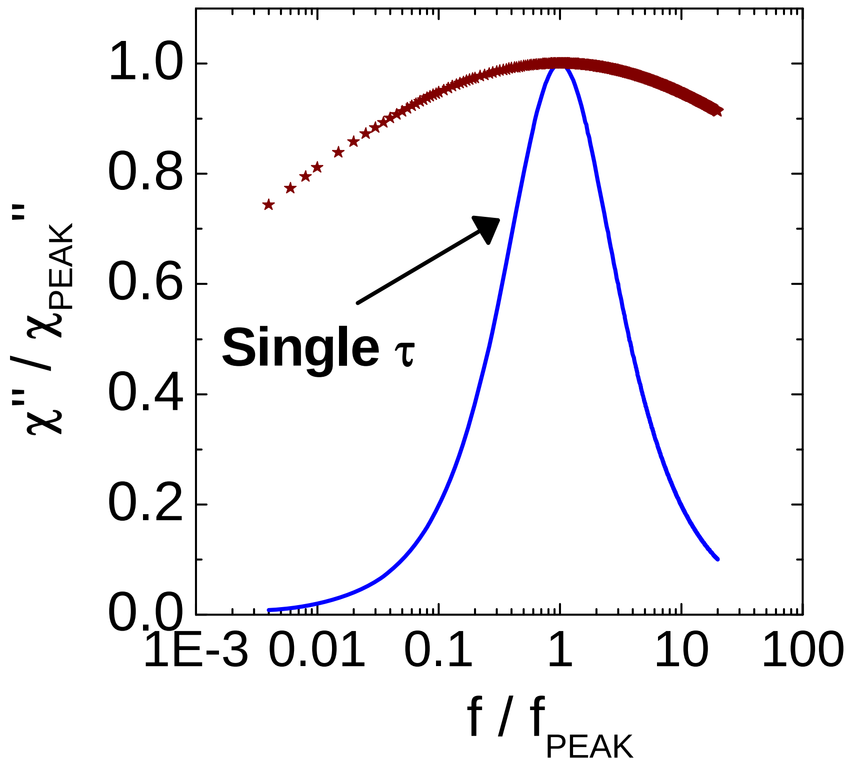
Spin Glasses: A few characteristics

Behavior both history and time dependent





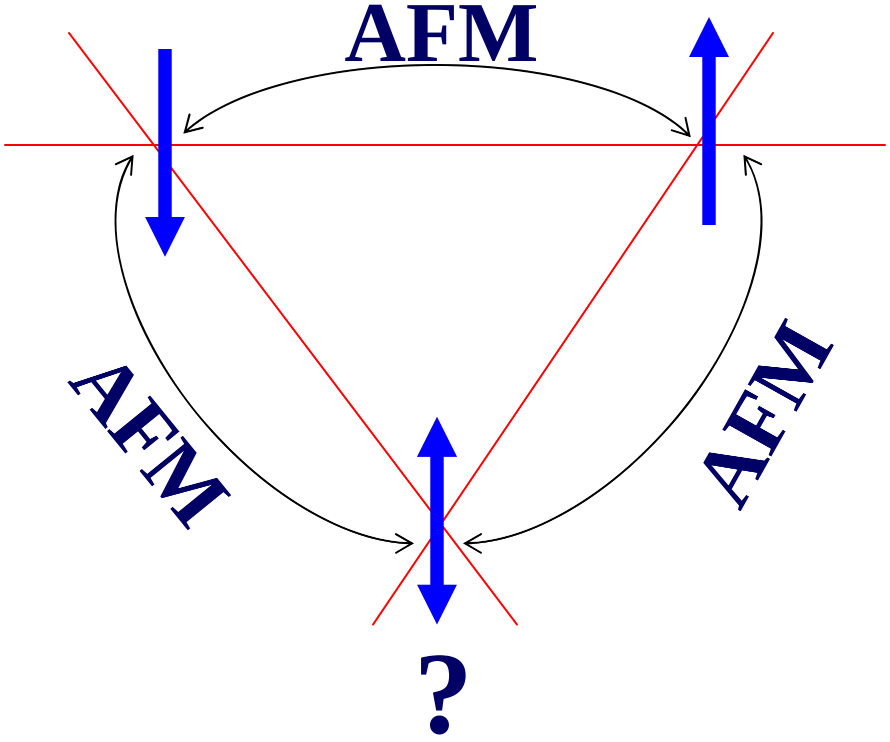
Broad range of length scales and relaxation times

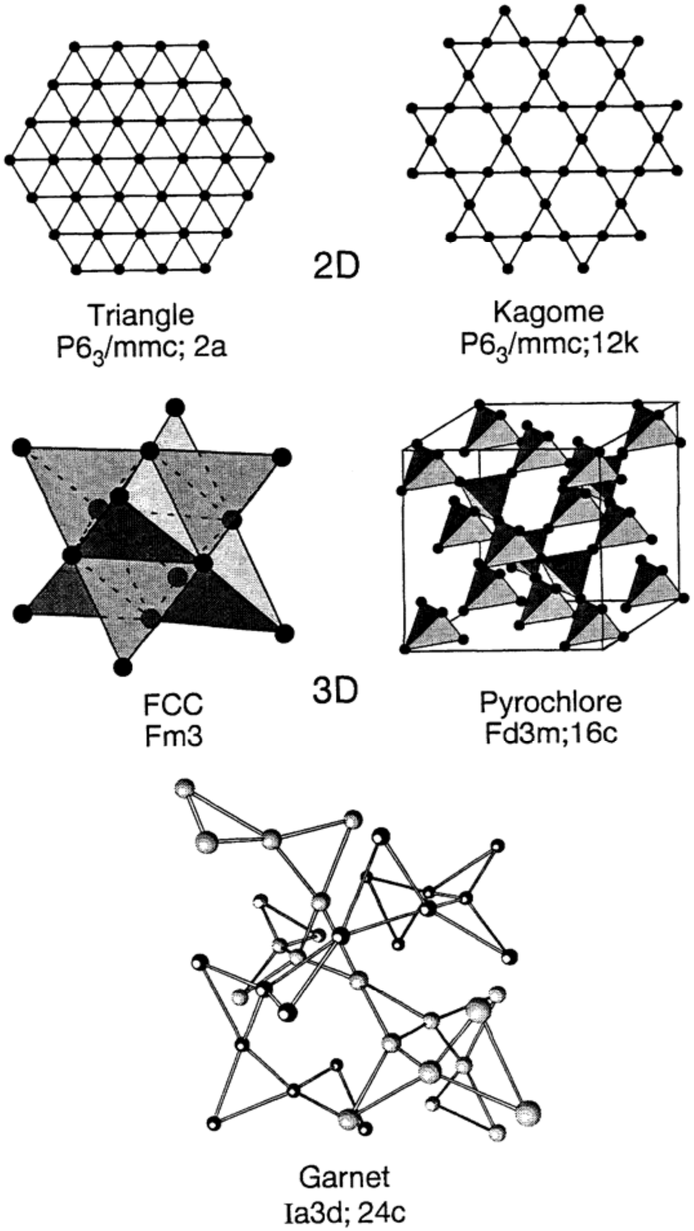


Geometrical magnetic frustration: <u>not based on disorder</u>

Competition of local interactions between spins on a regular lattice

→ high degeneracy of states from geometry of lattice

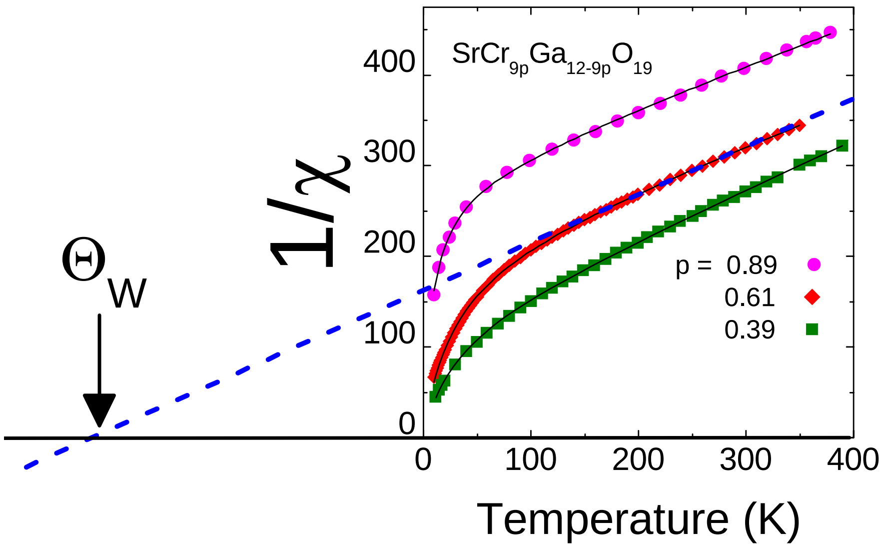


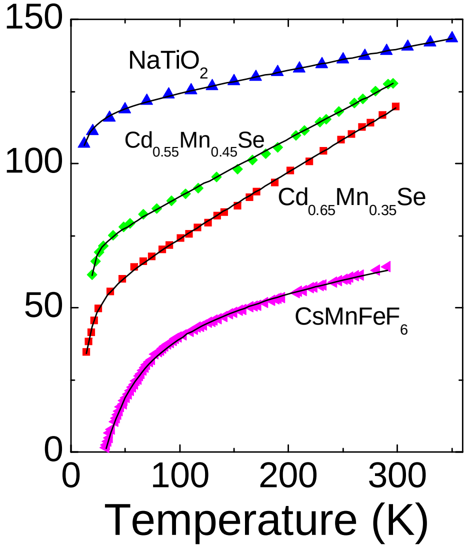


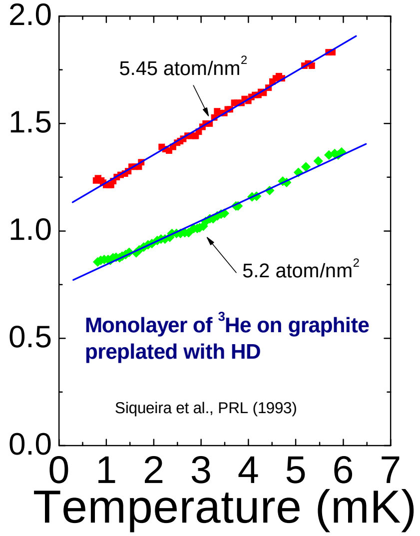
What’s interesting about geometrically frustrated magnets?

Continuum of energetically equivalent states

→ spins do not order at T ~ $|\Theta_W|$







Monolayer of ^{3}He on graphite preplated with HD

Siqueira et al., PRL (1993)

New ground states theoretically expected

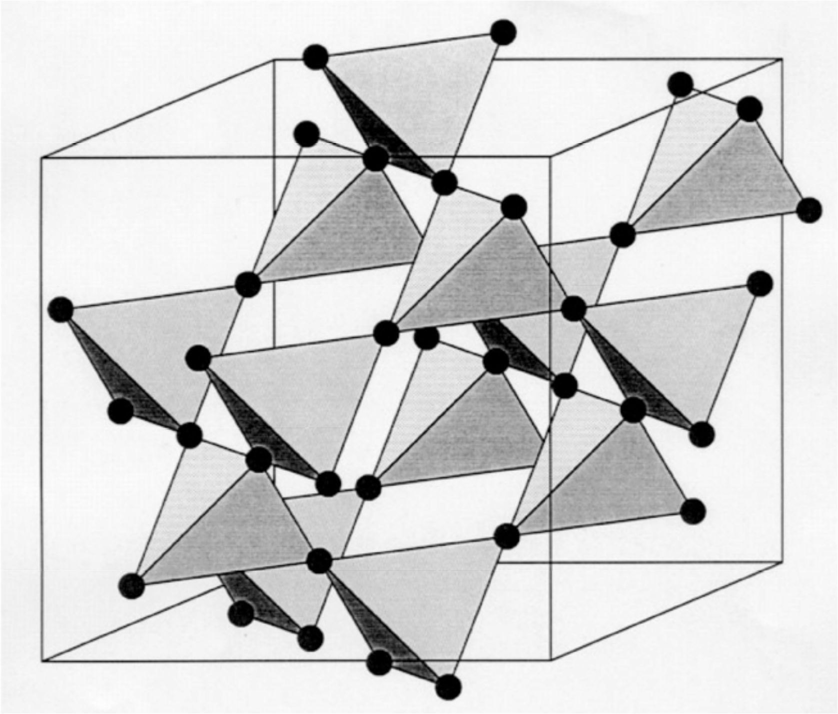
Anderson, Villain, Chandra, Shender, Berlinsky, Moessner, Chalker, LaCroix, Henley, Gingras, Lhuillier…

New ground states seen experimentally

Spin liquids, Spin glasses without disorder, Spin ice…

The “Spin Ice” Materials

Pyrochlore Lattice:
corner-sharing tetrahedra of spins



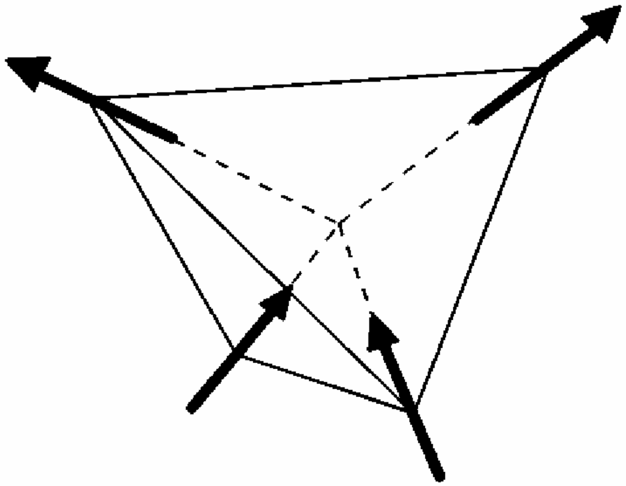
Spin ices: $Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$, $Ho_2Sn_2O_7$…..
Insulators with big rare-earth moments

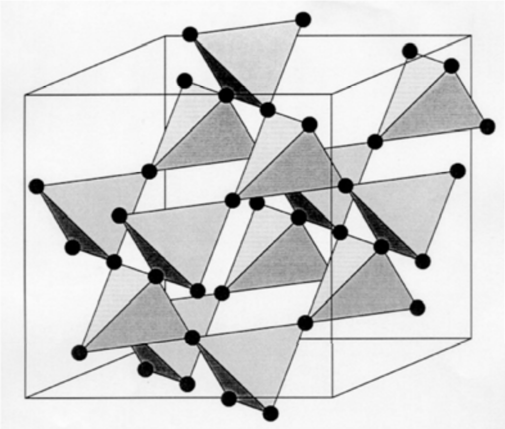
M. J. Harris *et al.* (1997)

What makes a spin ice: "Two-in/Two out"

- Crystal fields cause the rare-earth spins to be uniaxial along <111> directions (200 K energy scale)

- FM and dipole interactions cause spins to align two-in/two-out on each tetrahedron (2 K energy scale)



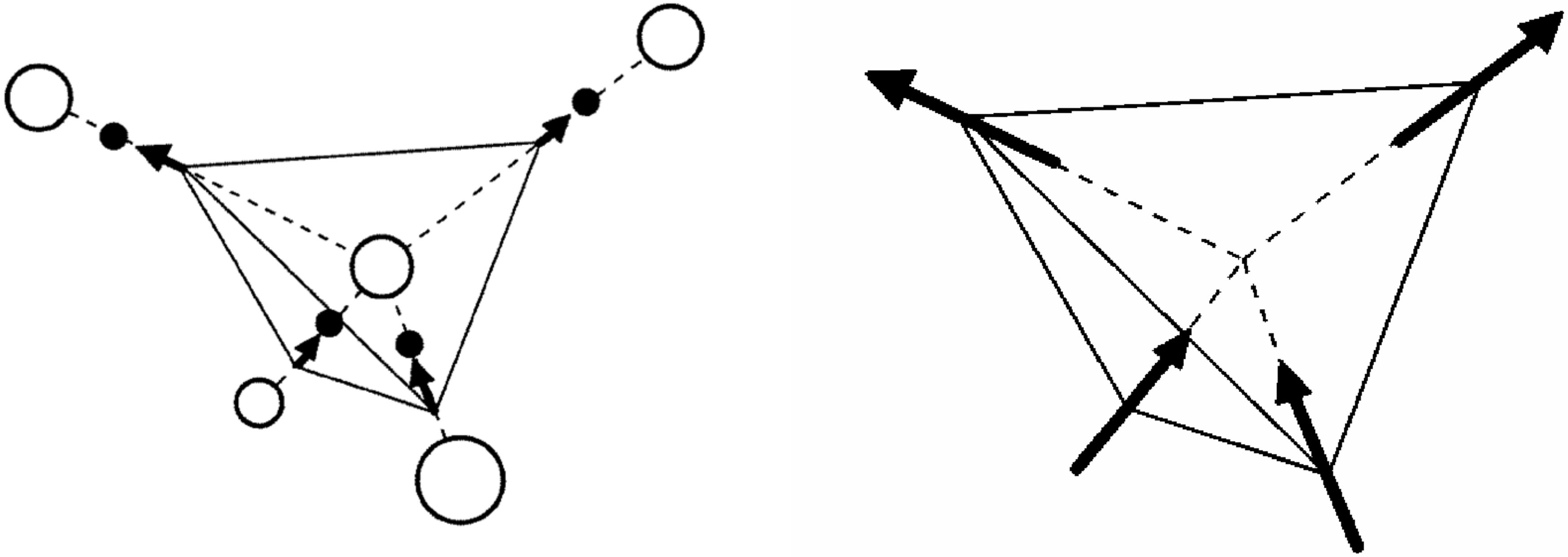


High degeneracy of possible states on pyrochlore lattice

Why call it a spin ice?

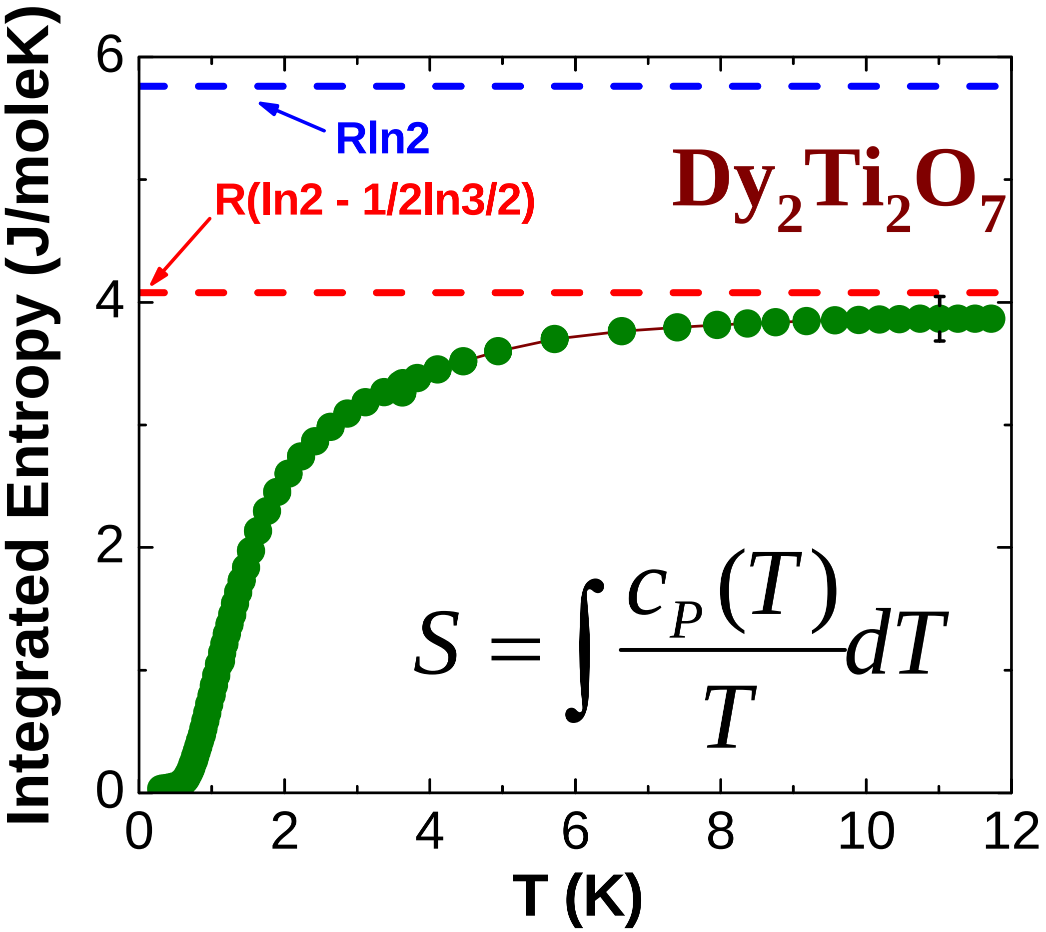
In frozen water (H_2O), each oxygen is surrounded by four hydrogens. Two are close to it, and two are closer to another oxygen ion.

Large degeneracy of states (Pauling 1945) invoked to explain the observed “ground state entropy” in ice



“Spin Ice” residual entropy seen in heat capacity experiments

Ramirez *et al.* (1999)



Ground State has predicted residual entropy in zero field, same as ordinary ice

Possibly spins are in metastable state (not in equilibrium) -- glassy ground state with onset T ~ 3K

Part 1: How does spin ice freeze?

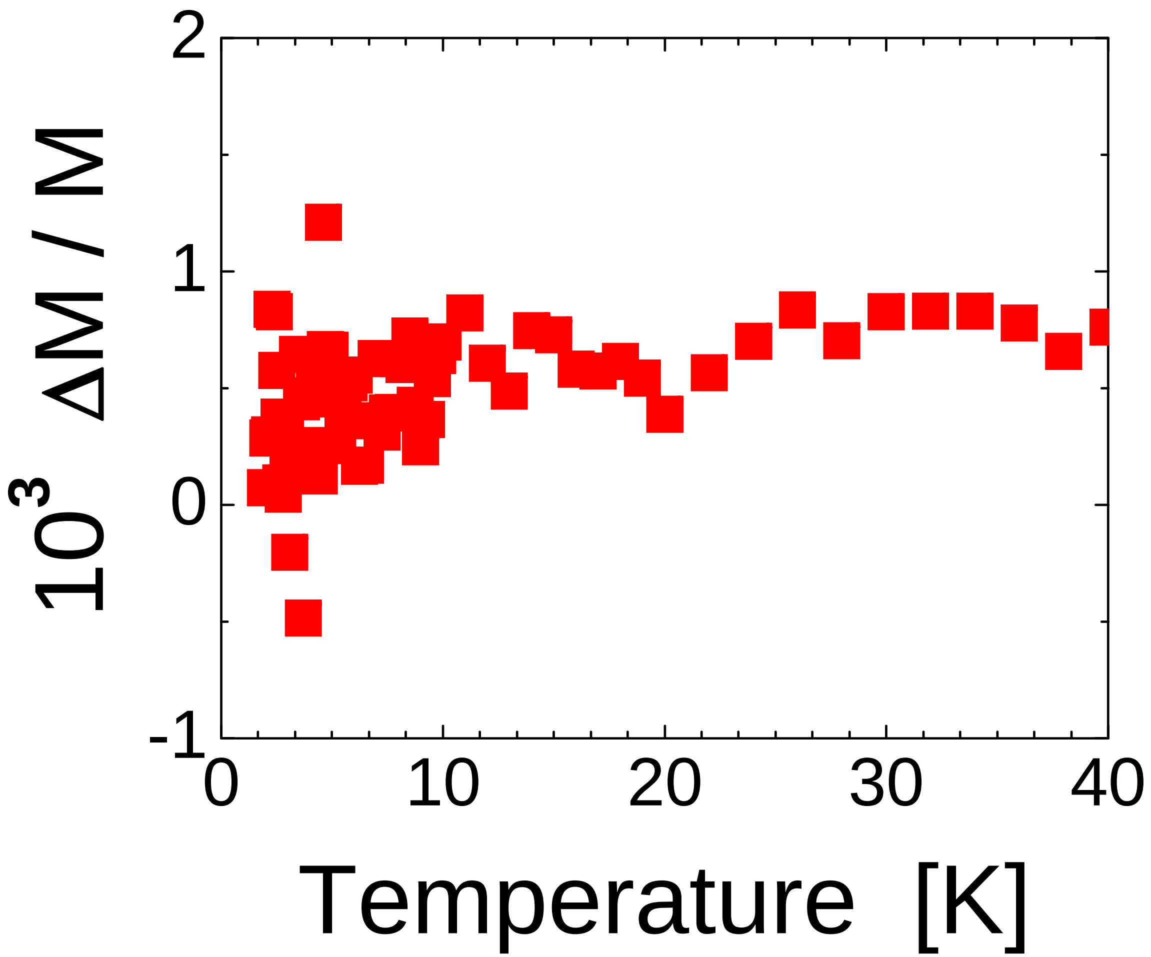
Measure a.c. and d.c. magnetic susceptibility
look for spin-glass-like freezing in $Dy_2Ti_2O_7$

Snyder *et al.* Nature 2001
similar work done by Matsuhira *et al.*
J. Phys. Cond. Mat. 2001

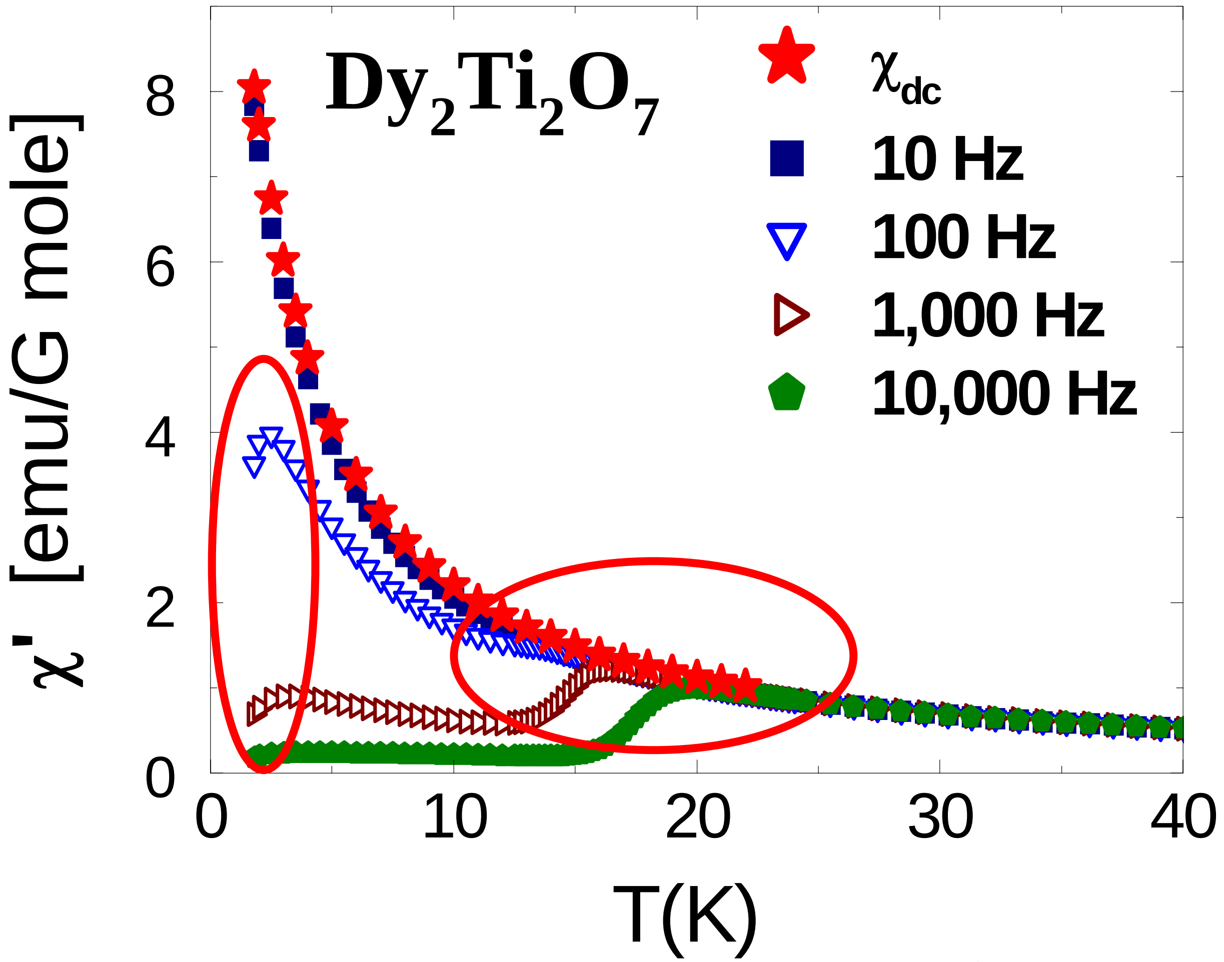
Snyder *et al.* Phys. Rev. Lett. 2003
Snyder *et al.* Phys. Rev. B 2004 (two papers)

Magnetization data: No sign of freezing above 1.8 K

No difference between field-cooled and zero-field-cooled data



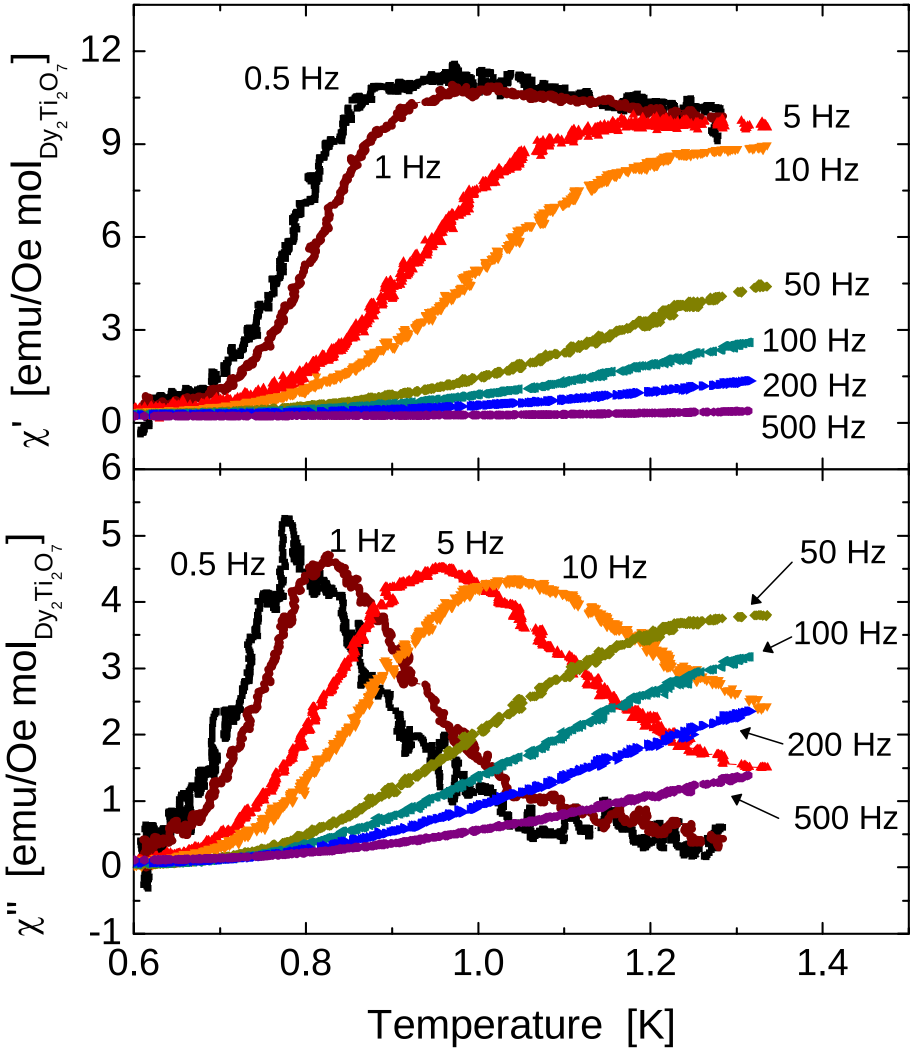
AC susceptibility – much more interesting



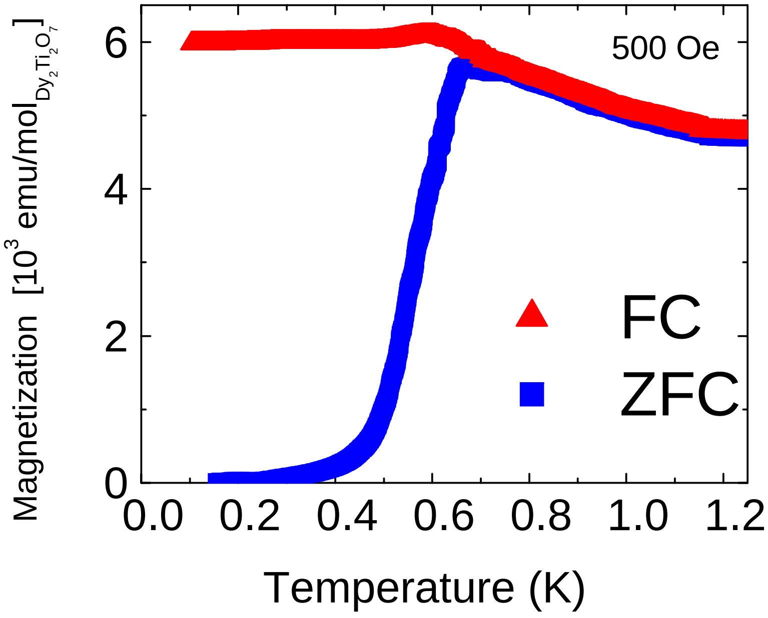
Spin freezing at T ~ 16 K and second feature at T ~ 3K

Freezing is strongly frequency dependent (Arrhenius law)

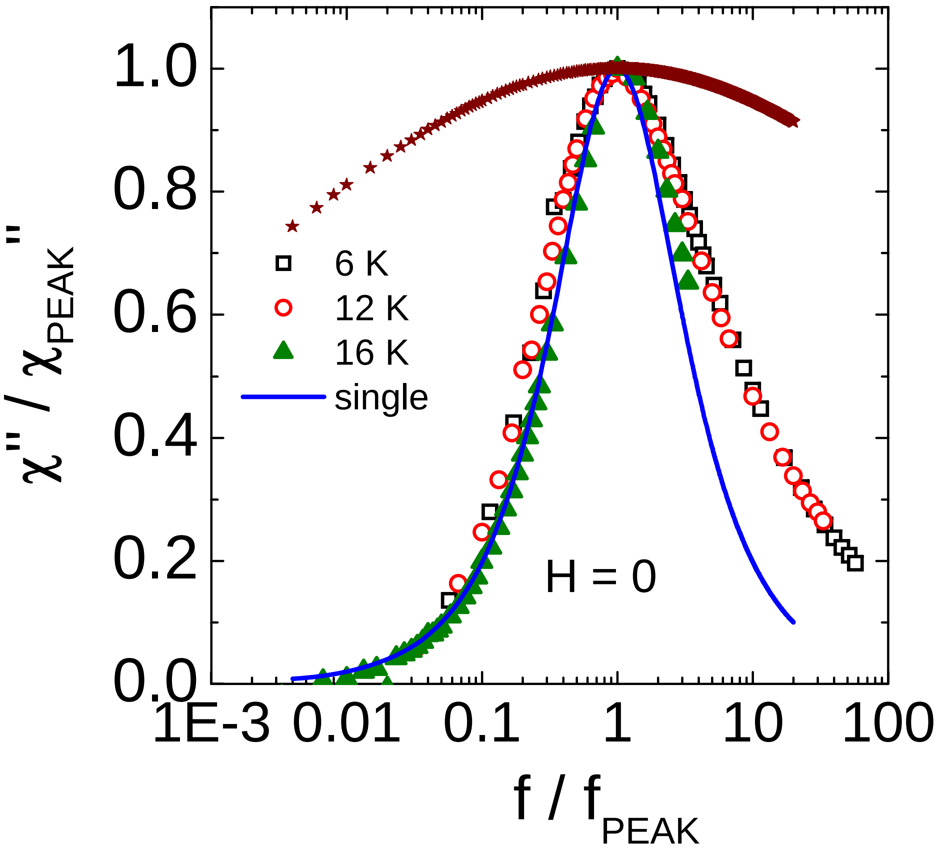
Lower temp. feature in $\chi(T)$ is another freezing

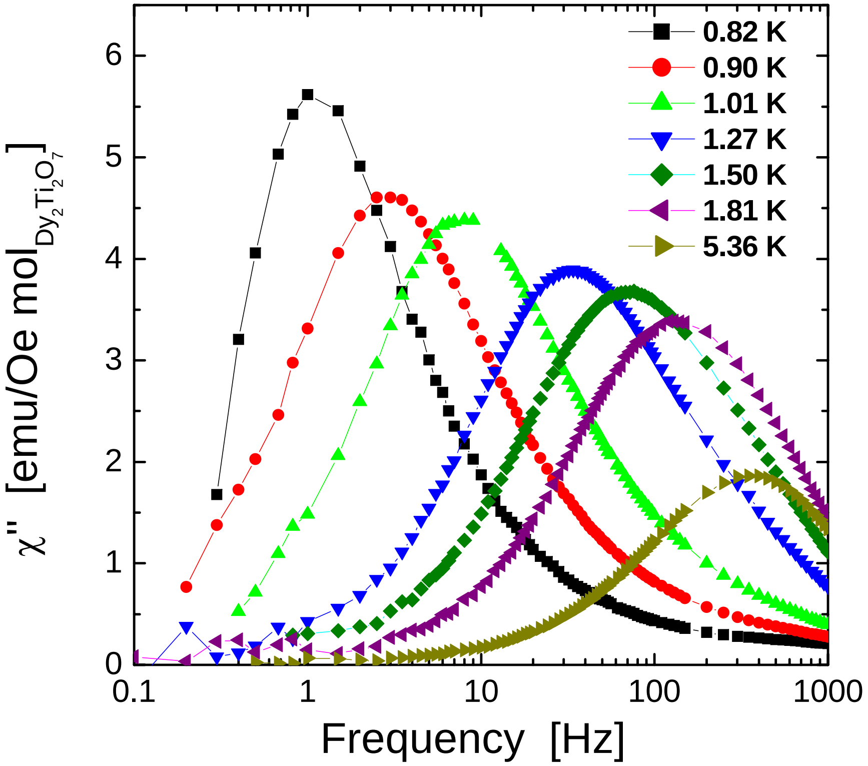


Snyder *et al.* PRB 2004



Spin ice freezing involves very narrow range of relaxation times



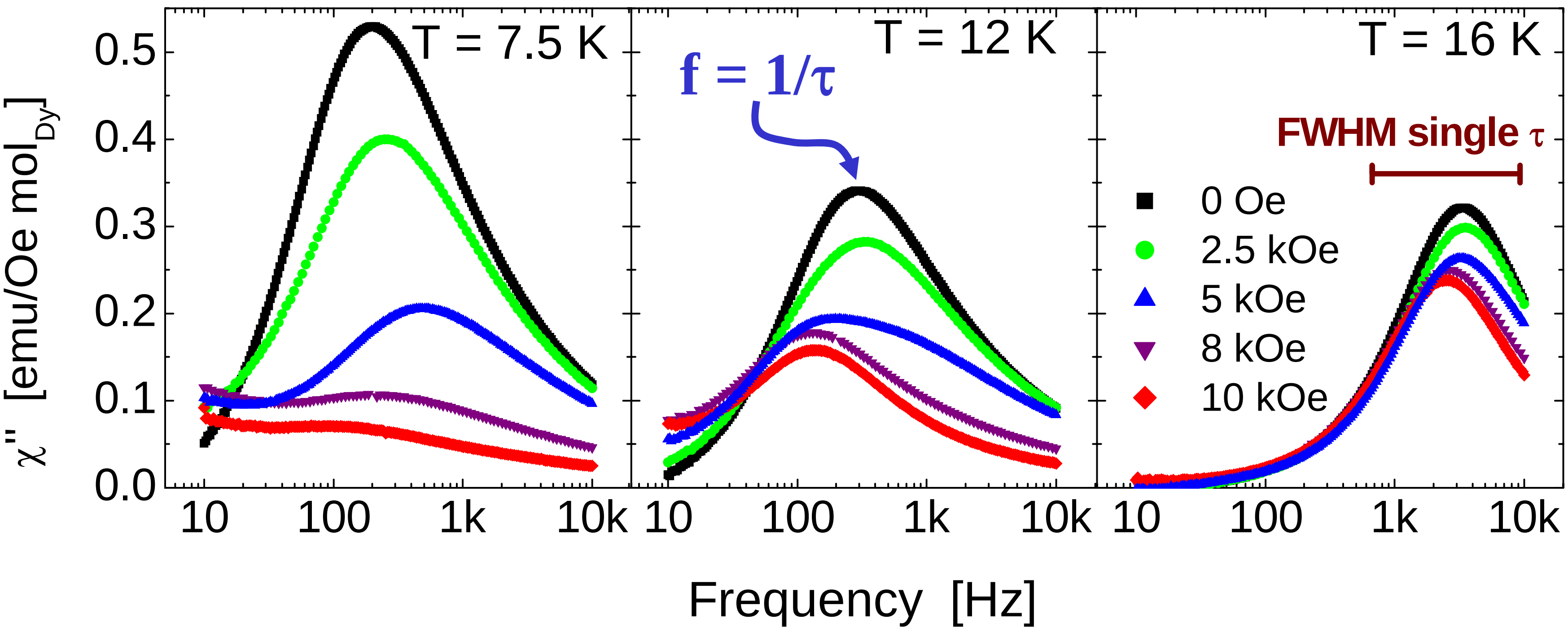


No broad range of time-scales typical of spin glasses:

<u>This freezing is something very different!</u>

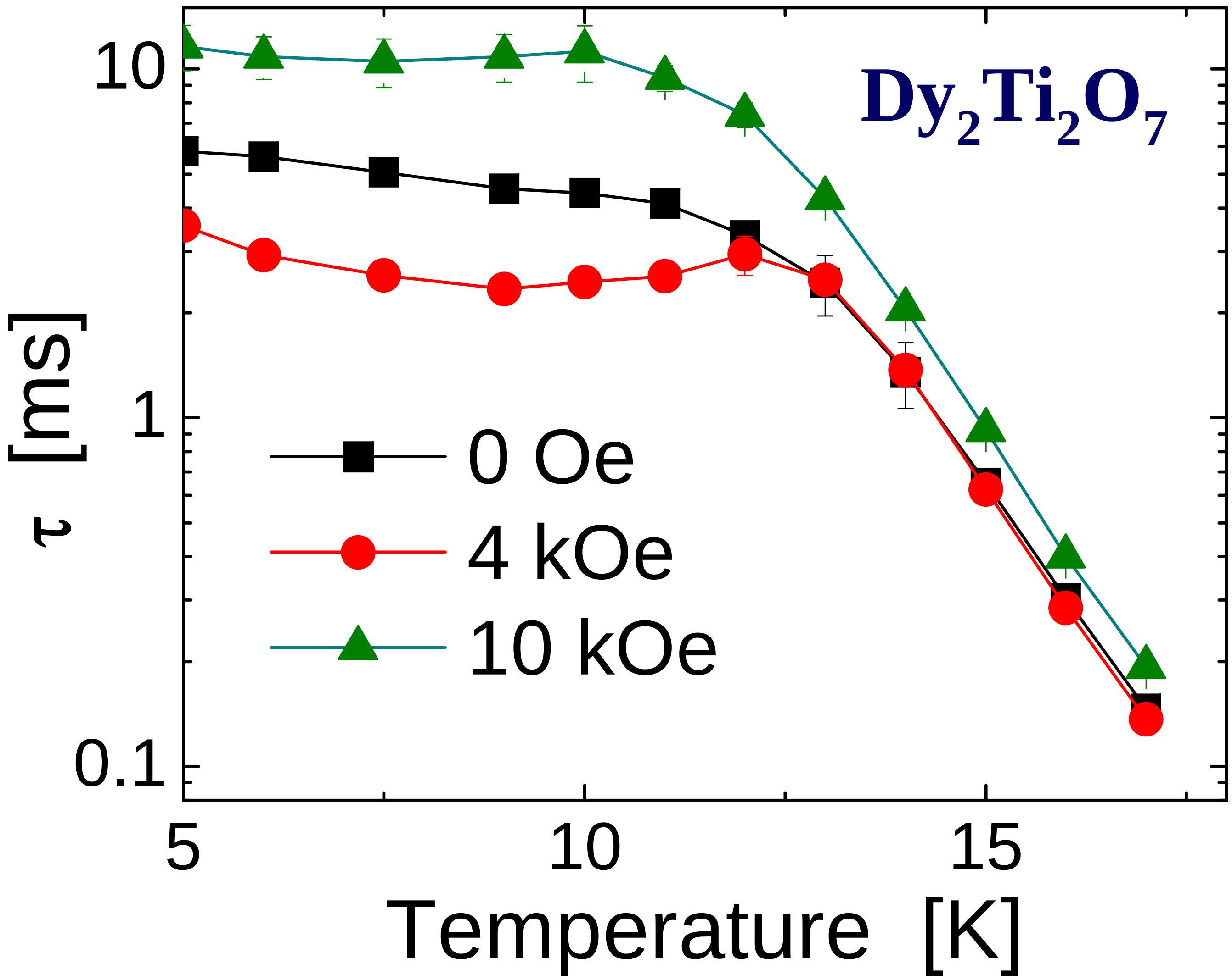
Understand double-freezing in $Dy_2Ti_2O_7$ from spin relaxation time

Maximum in χ"(freq) gives the relaxation time, τ

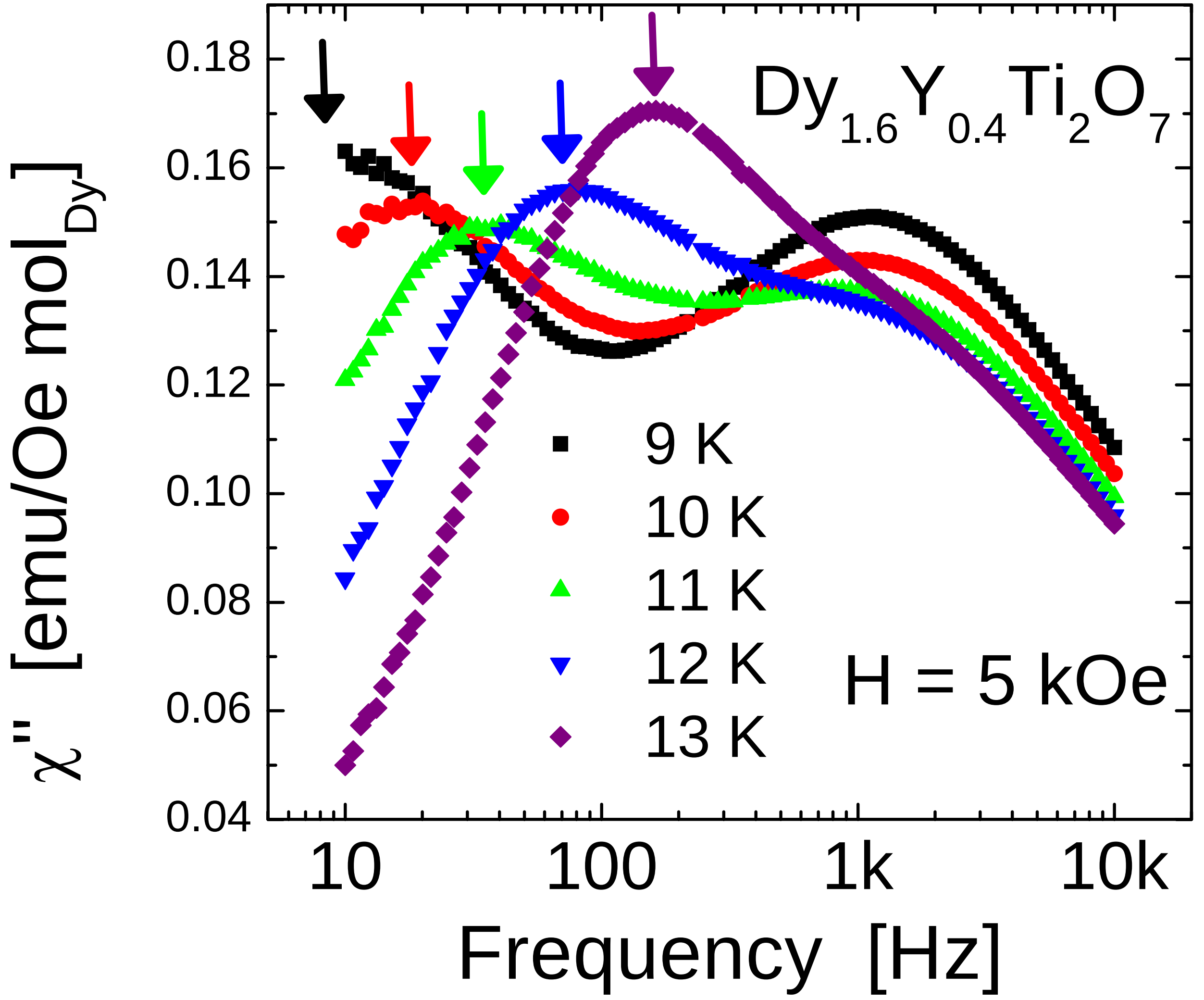


Crossover from thermal to quantum spin relaxation at *T* ~ 13 K

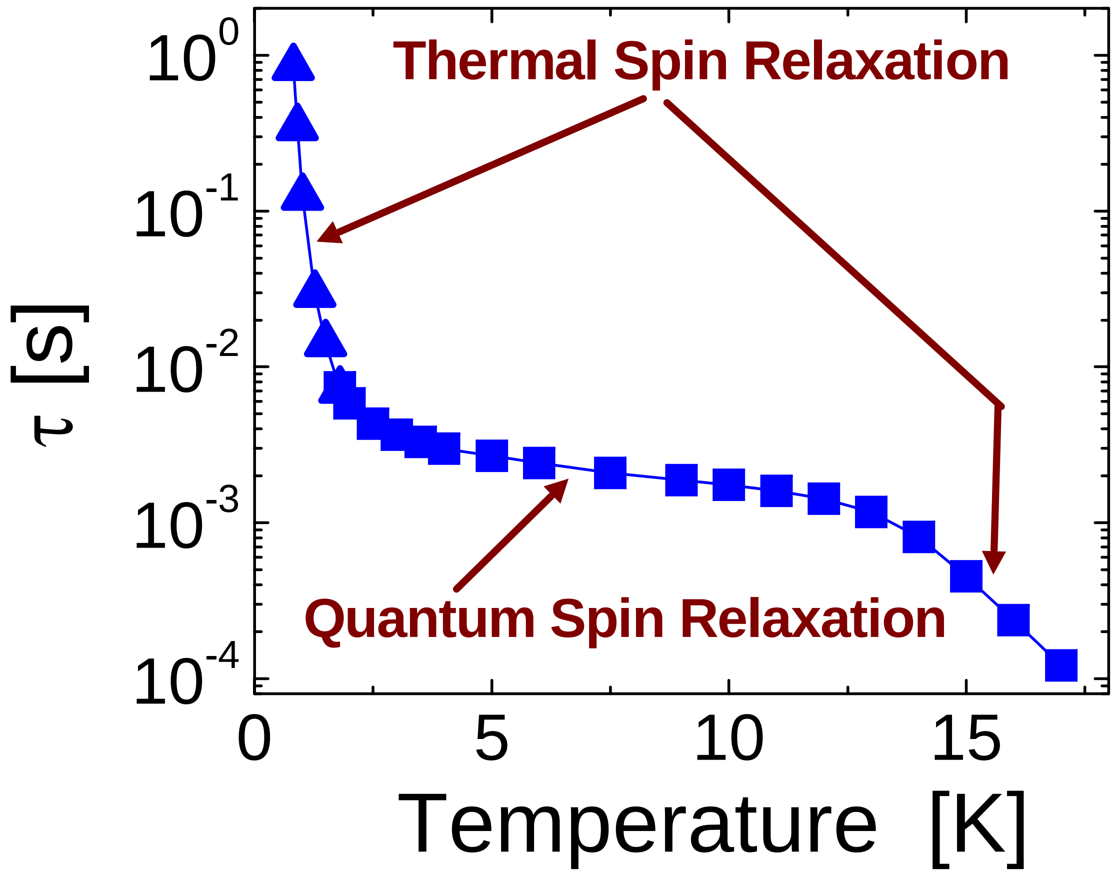
Snyder *et al.*, PRL 2003



Two types of relaxation clearly seen in a field

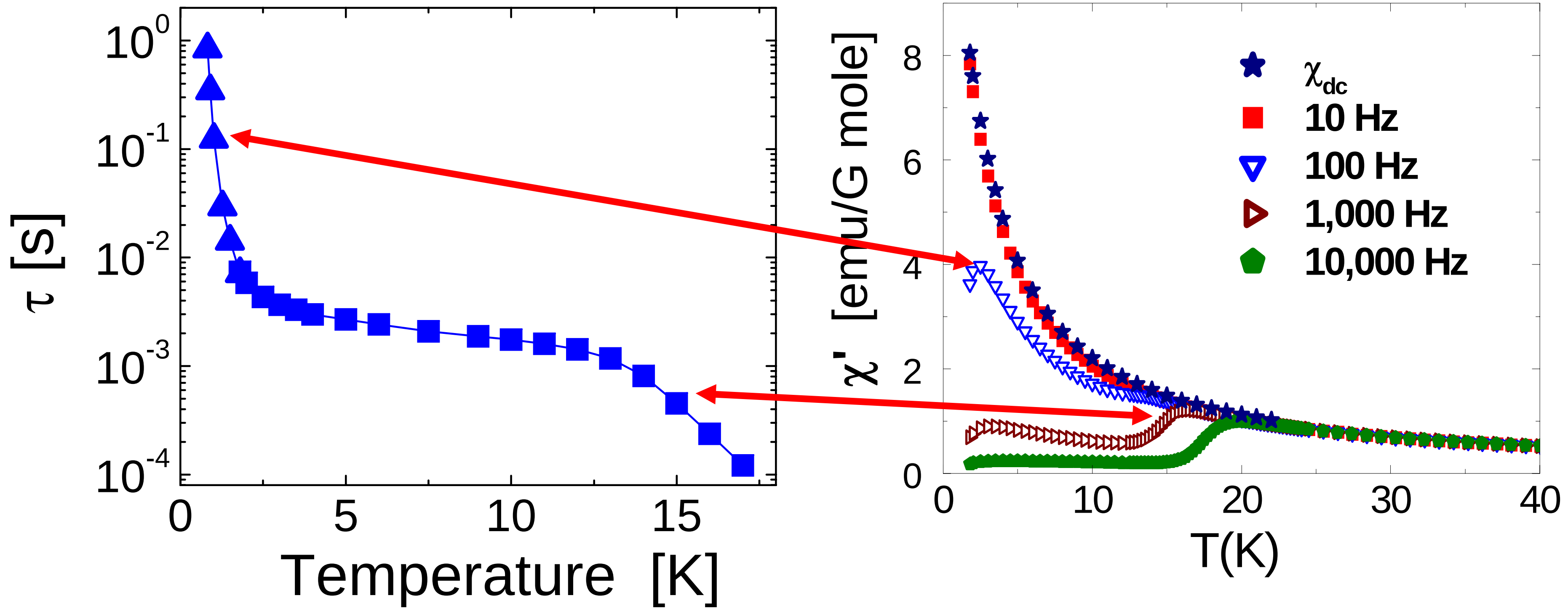


Thermal relaxation re-emerges at low temp.



Low temperature spin-correlations are apparently suppressing the quantum relaxation

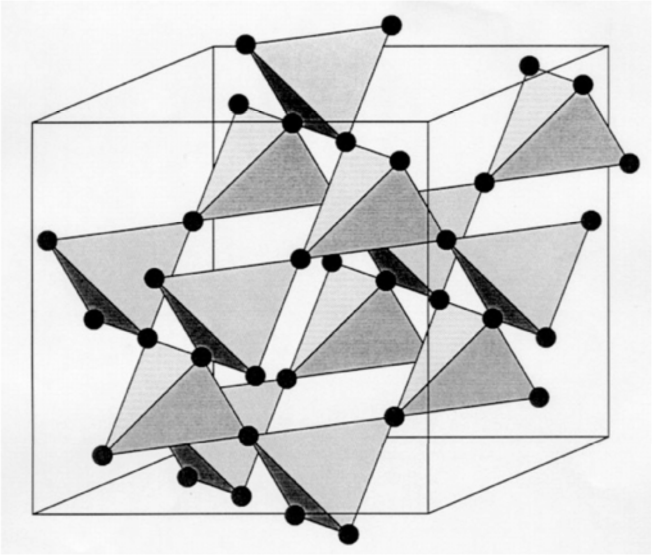
Re-entrant thermal spin relaxation explains double freezing in $Dy_2Ti_2O_7$



- **Thermal fluctuations freeze out**
  (single ion process, Ehlers et al.)
- **Quantum fluctuations until spins get correlated**
- **Correlated spins relax very slowly**

Part 2: Stuffed Spin Ice

What happens if we add more magnetic ions to the lattice?



Ho lattice only

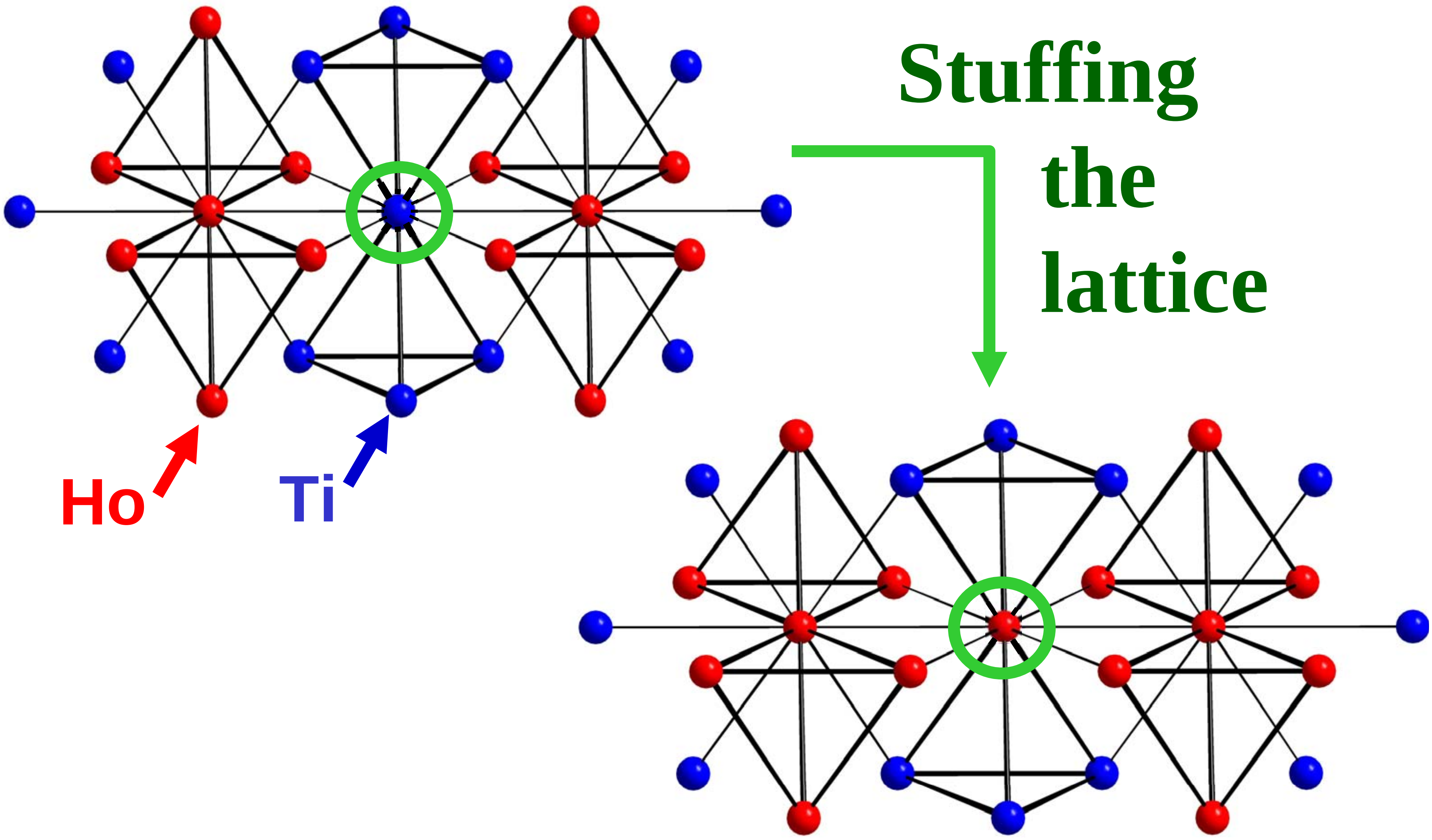
Start with $Ho_2Ti_2O_7$ and replace Ti with Ho ions

get $Ho_2(Ti_{2-x}Ho_x)O_{7-x/2}$

Lau *et al.* (Nature Physics, in press)

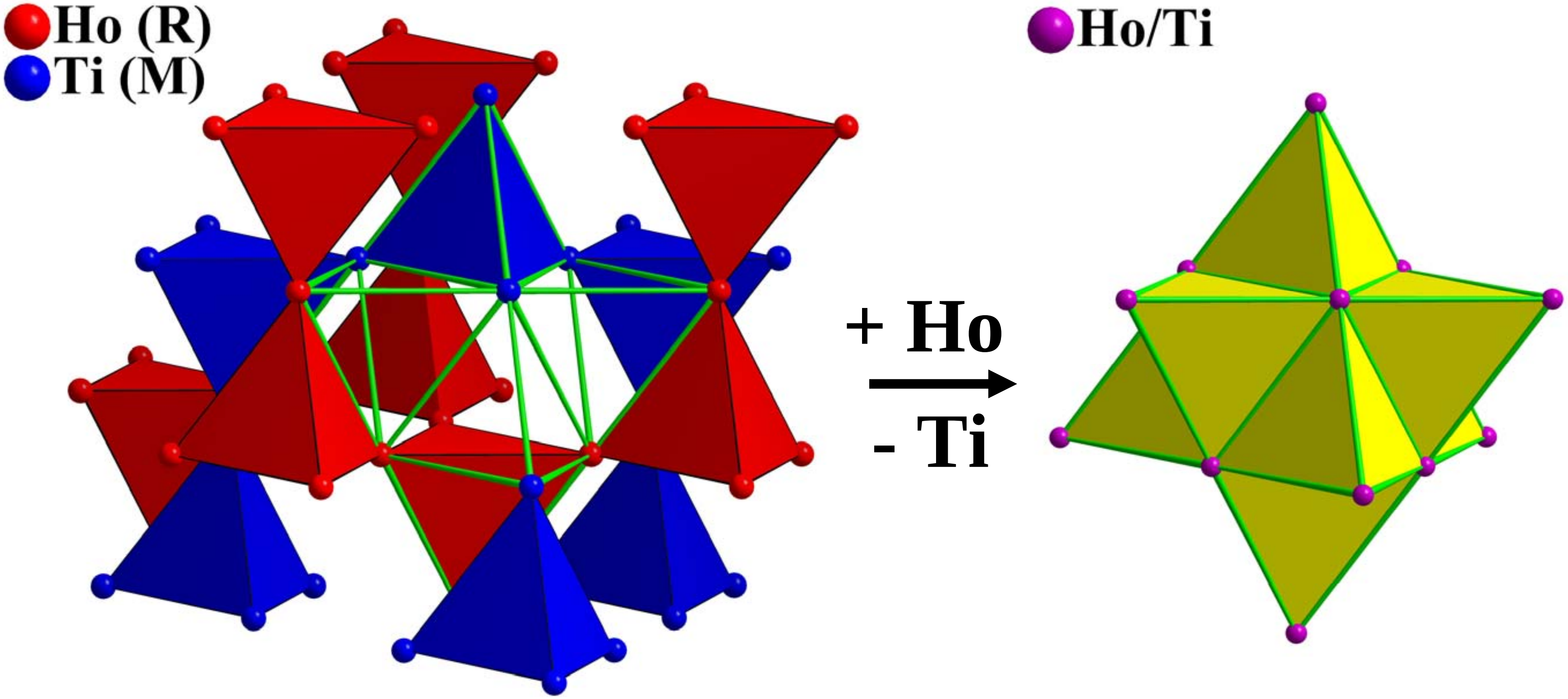
Stuffing the lattice: $Ho_2(Ti_{2-x}Ho_x)O_{7-x/2}$

Randomly replace some Ti^{4+} with Ho^{3+}



Stuffing the lattice

Stuffing changes the connectivity of spins

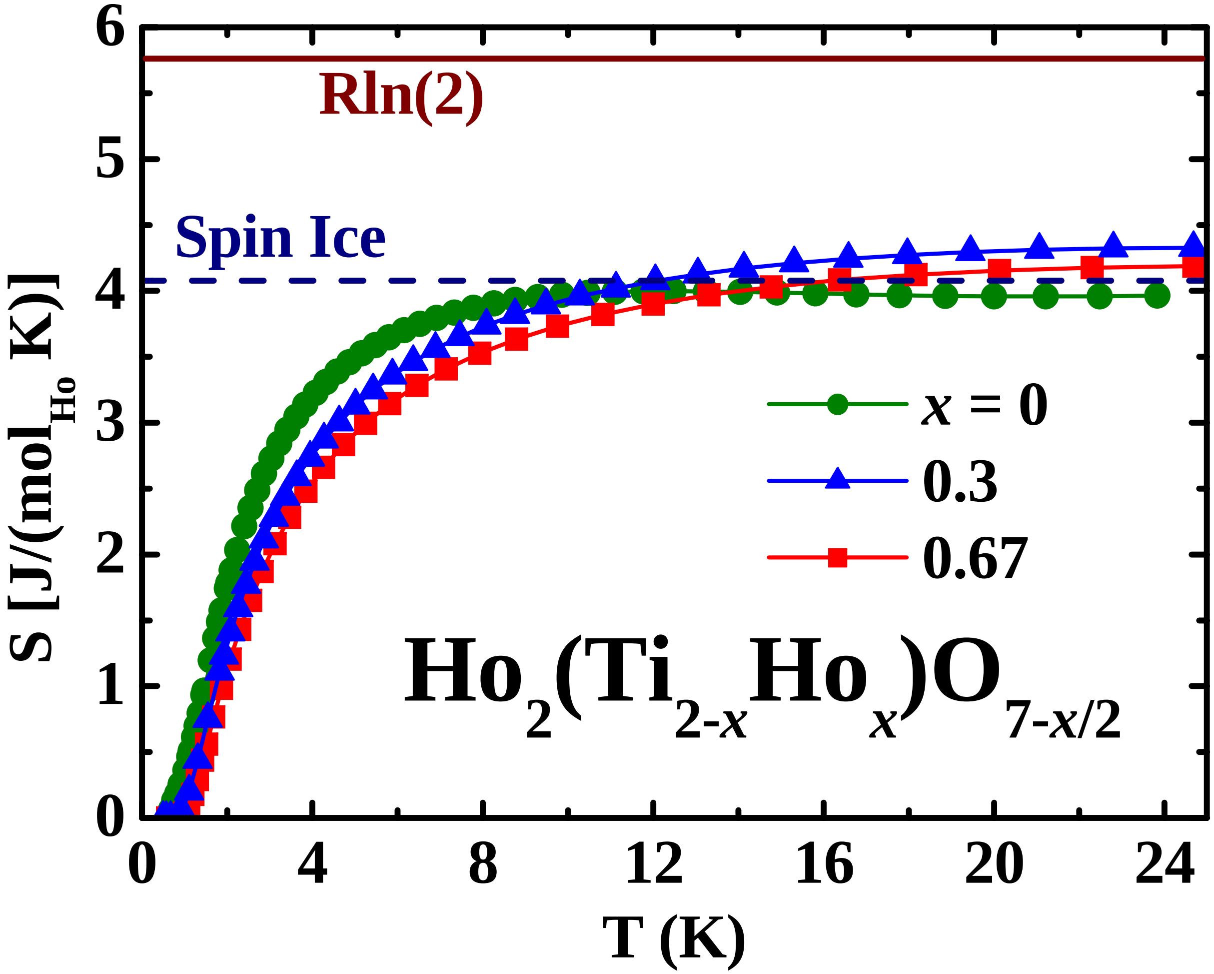


x = 0

x = 0.67

corner sharing → edge sharing

Magnetic entropy of the stuffed spin ice

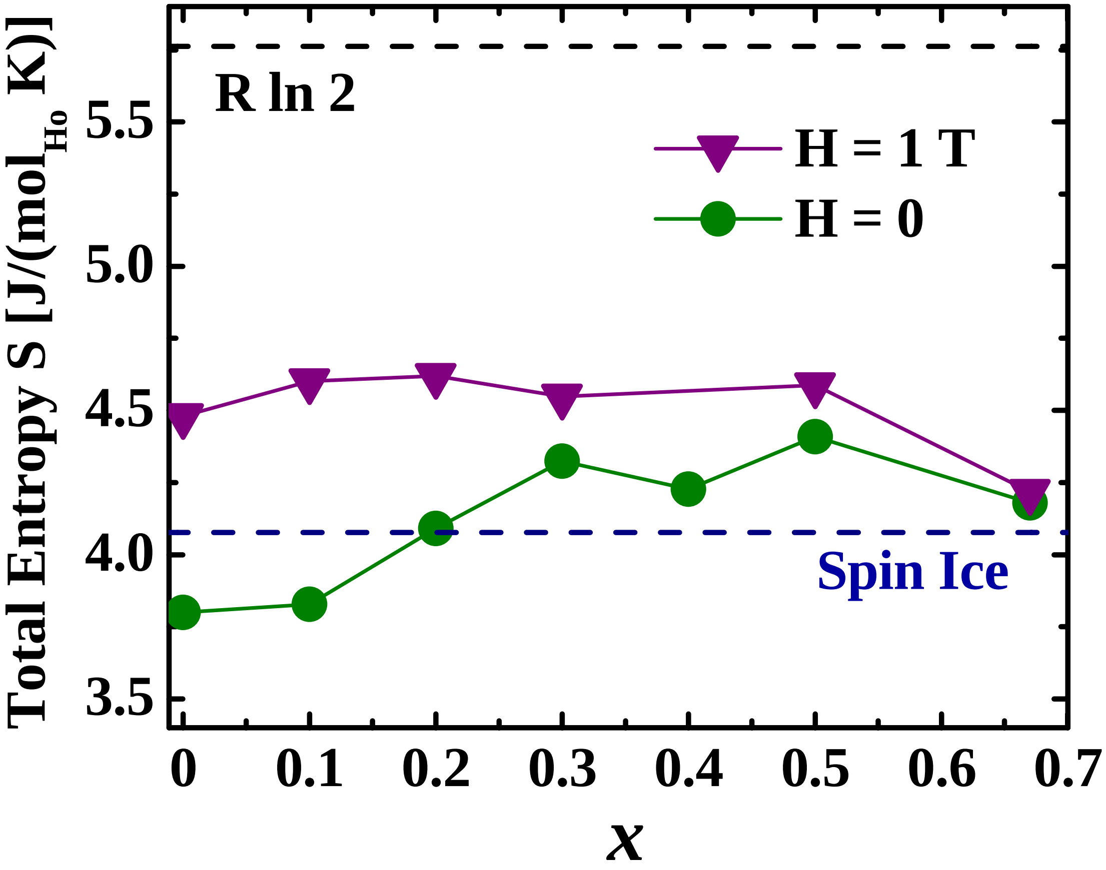


$$S = \int \frac{c_P(T)}{T} dT$$

Entropy recovery is similar for all x!!

Ground state entropy also for stuffed case

Total Entropy vs. Stuffing



Entropy is unchanged within measurement uncertainty

Open Questions:

- **What macroscopic state exists as approach fcc lattice?**
- **How common is the “ground state” entropy?**

Part 3: Artificial Spin Ice

Wang *et al.*, Nature 2006

A major limitation in the study of magnetic frustration is that we are limited by the materials

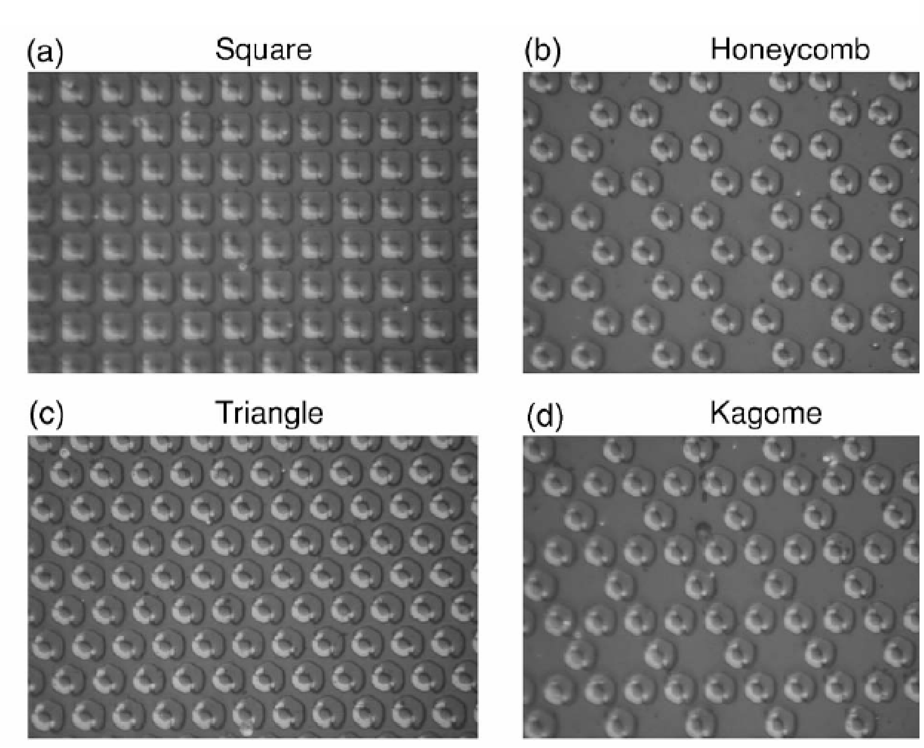
- **Cannot easily change lattice spacing/geometry**
- **Cannot control defects locally**
- **Cannot image individual spins**

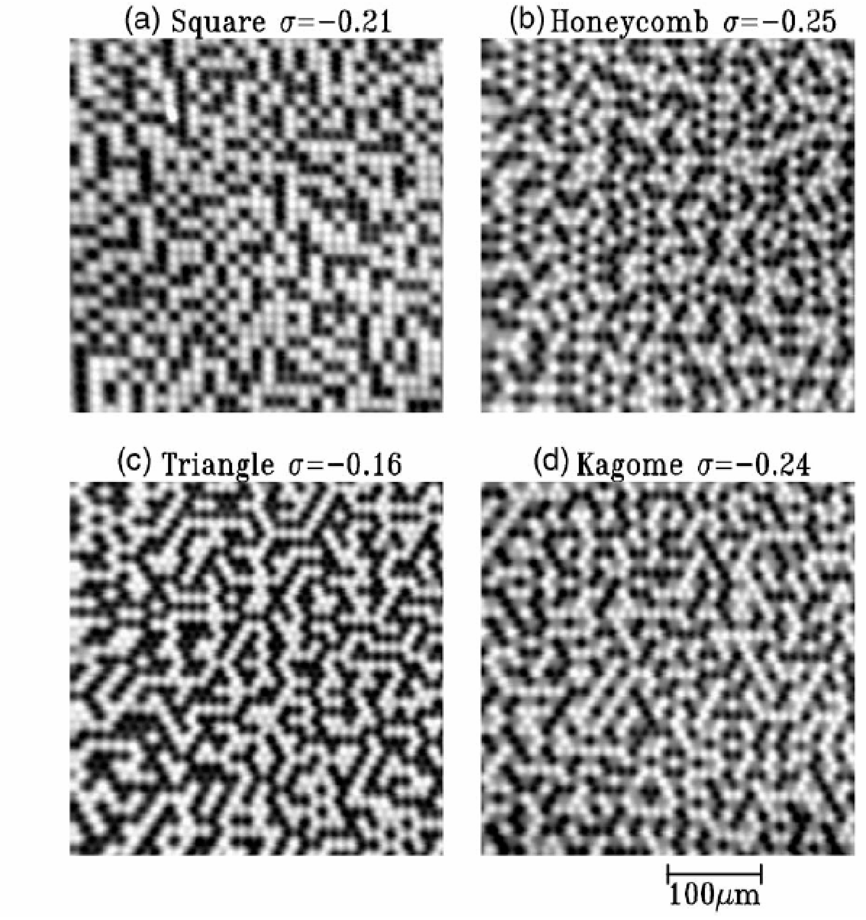
What if we use modern nanometer-scale lithography to create an artificial frustrated system?

Artificial frustrated systems with superconductors

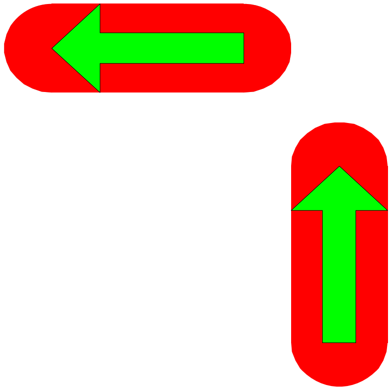
Superconducting rings or Josephson junction loops with flux trapped

Davidovic *et al.* 1996, 1997
Hilgenkamp, Kirtley, Tsuei *et al.* 2003, 2005





What if we make "artificial spins" using ferromagnetic material?



- Etch islands out of a ferromagnetic film
- If islands are small enough, they will be single-domain (like big spins)
- Shape anisotropy results in Ising-like spins with direction given by island shape
- Islands interact through local magnetic fields

Do ferromagnetic islands really behave like big spins? Yes!

Choose shape such that islands are single-domain

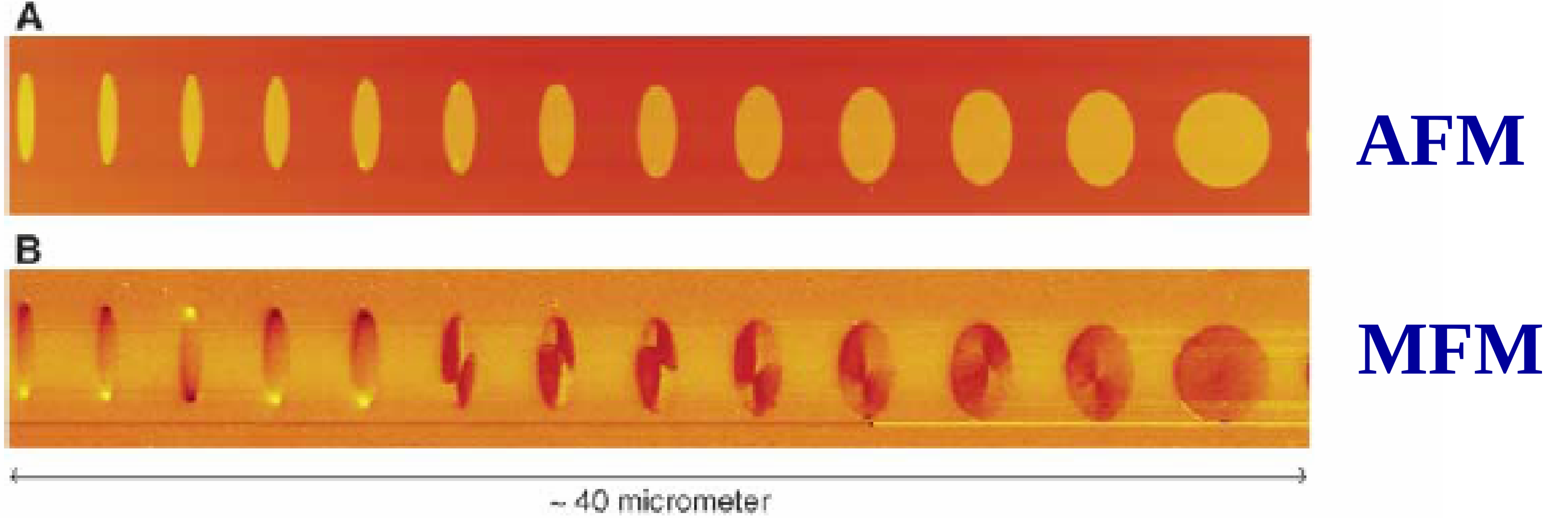
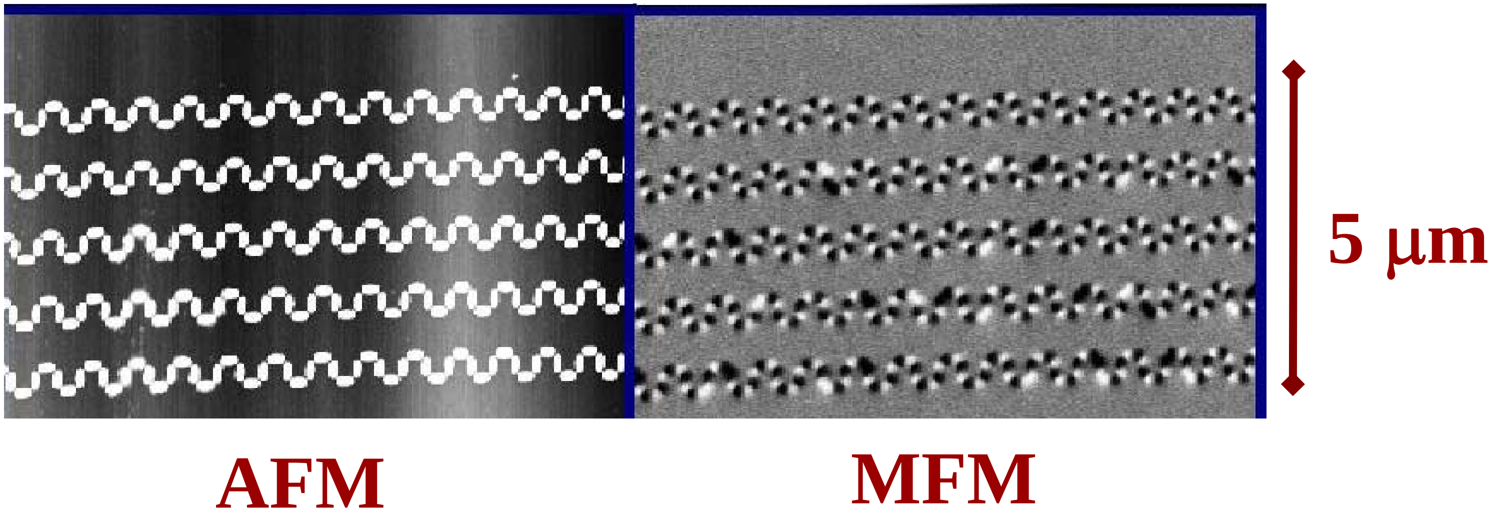
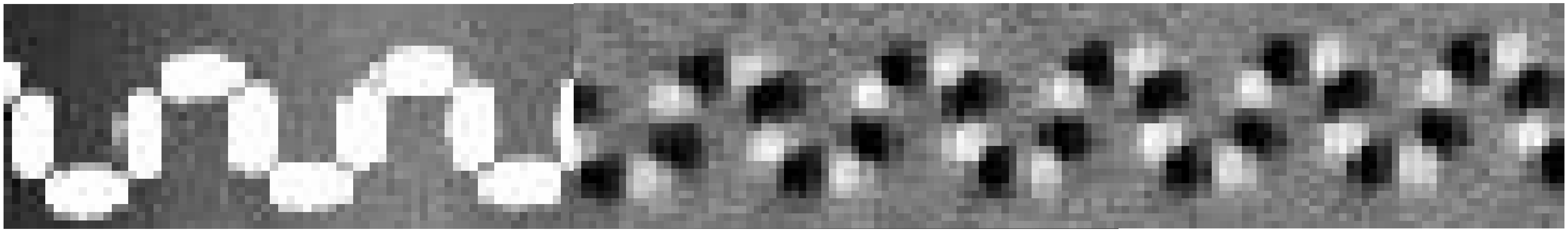


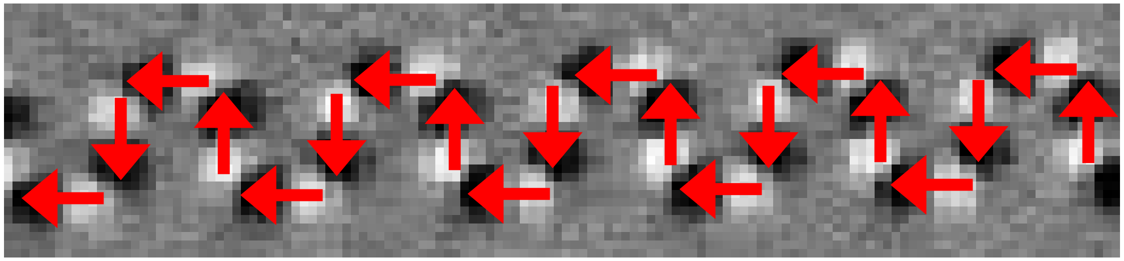
Fig. 1. Magnetic states of patterned ferromagnetic elements. (**A**) Topographic atomic force microscope (AFM) image of thin-film, polycrystalline NiFe magnetic elements in a row, made by electron-beam lithography and lift-off. While one axis of the oval elements is kept the same, the aspect ratio of the axes is varied between 7.5 (on the left side) to 1 (on the right side). (**B**) MFM image of remanent magnetization. Strong contrast in the MFM images of one-domain magnets represents the location of magnetic poles. This image reveals one-domain, two-domain, and vortex configurations, depending upon the size and shape (aspect ratio) of the elements.

Imre *et al.* 2006

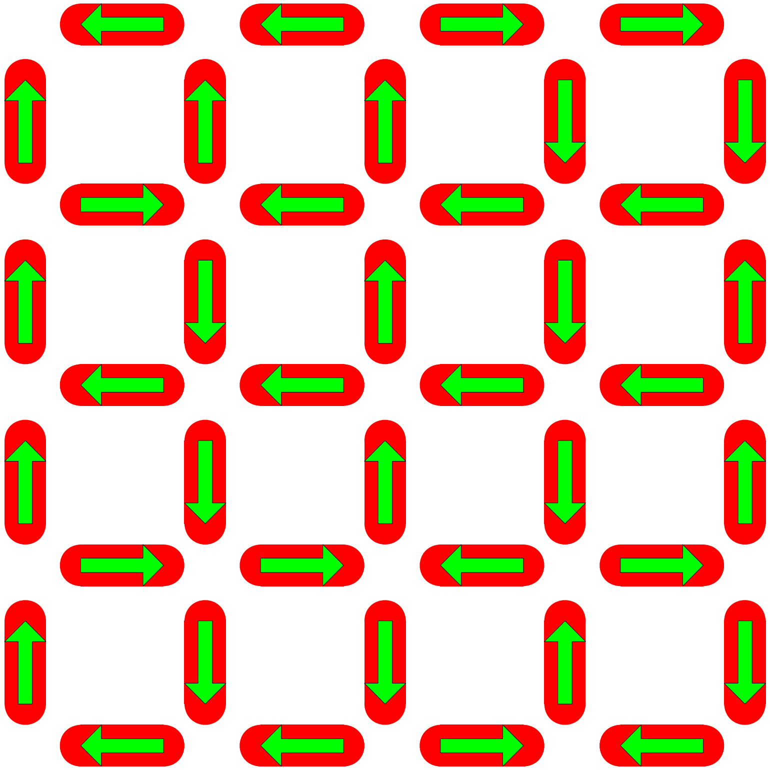
Are island moments controlled by interaction? Yes!

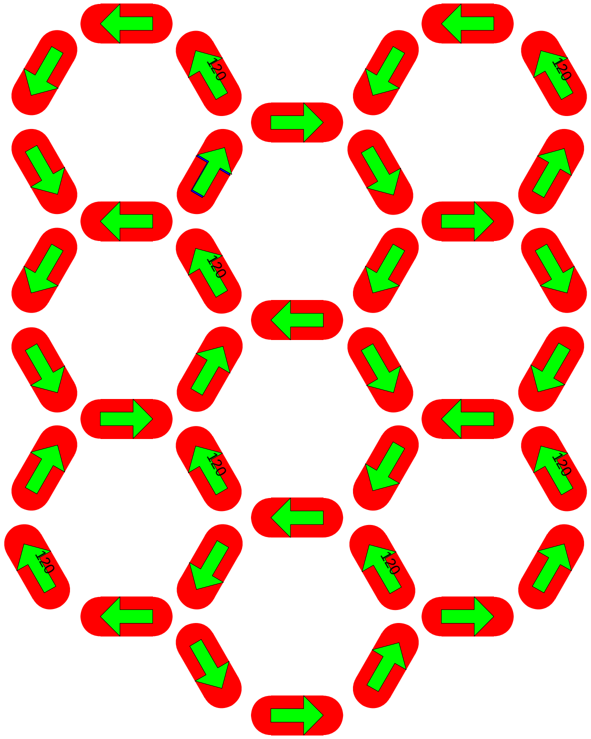






Making frustrated 2-D network out of ferromagnetic islands

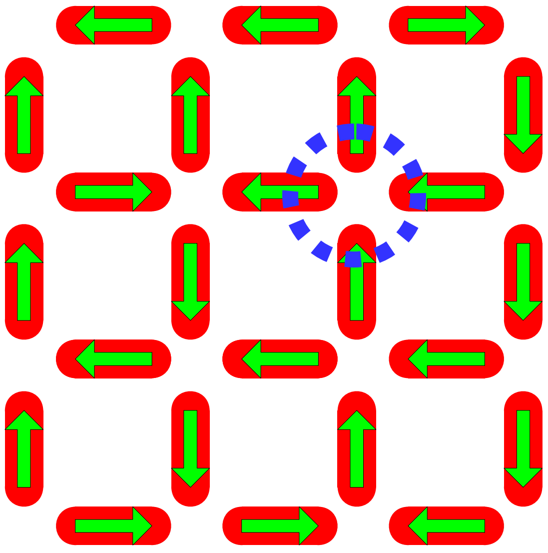


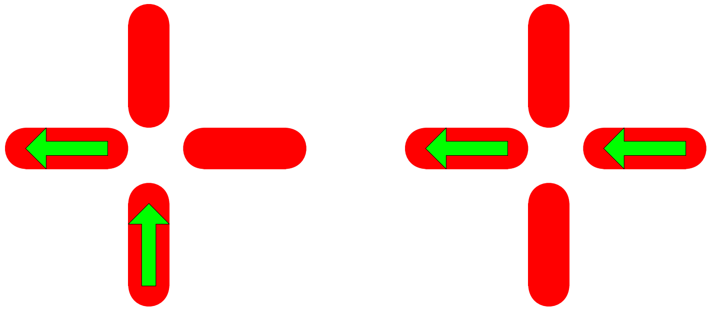


Advantage of lithographically defined network:

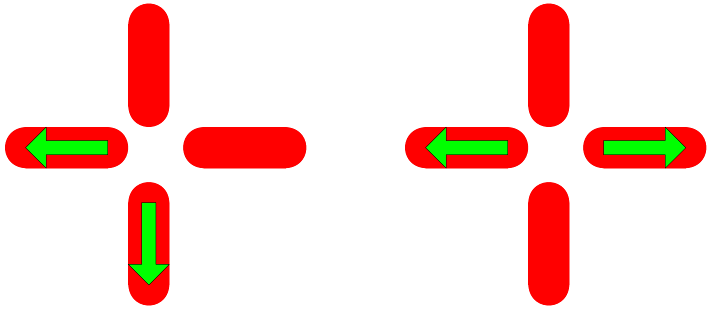
- **Fine control of island shape, size, and spacing**
- **Direct investigation of individual islands by MFM**
- **Flexibility in introducing defects**

Dipolar interactions in one vertex of the perpendicular square lattice

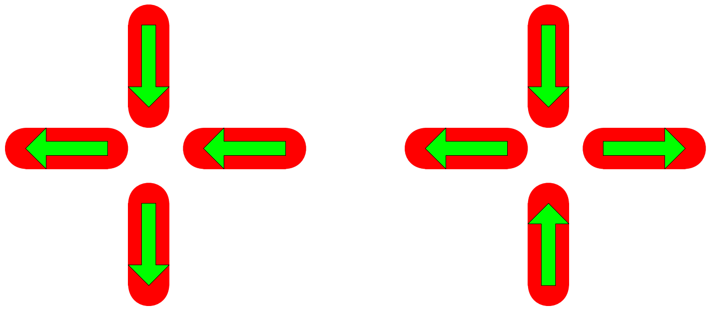




Favorable pair configuration



Unfavorable pair configuration

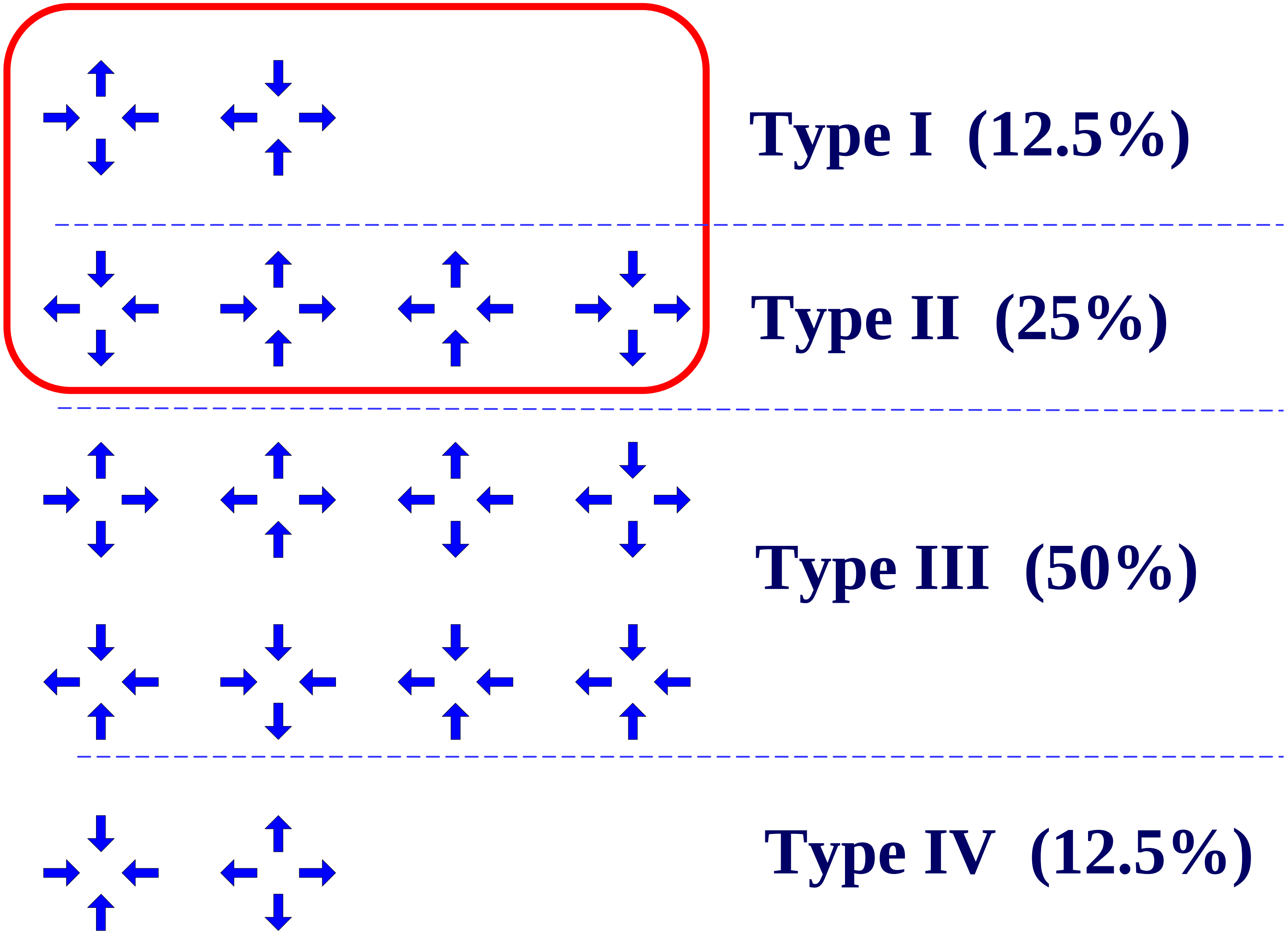


The six pairs of dipole interactions cannot be minimized simultaneously: <u>FRUSTRATION!</u>

Magnetization configurations on single vertex

Expect certain distribution if orientations are random

Lowest energy states are two-in/ two-out: LIKE SPIN ICE!!



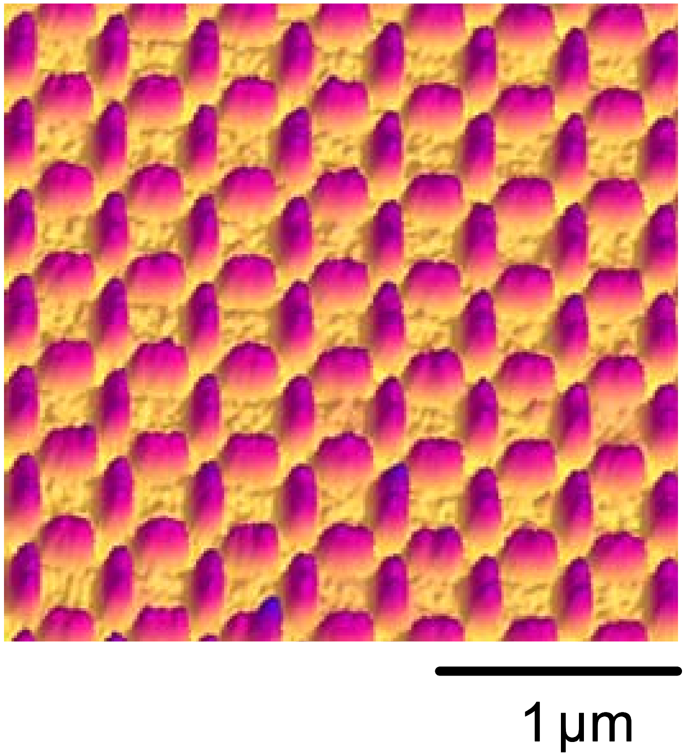
Type I (12.5%)

Type II (25%)

Type III (50%)

Type IV (12.5%)

Sample details

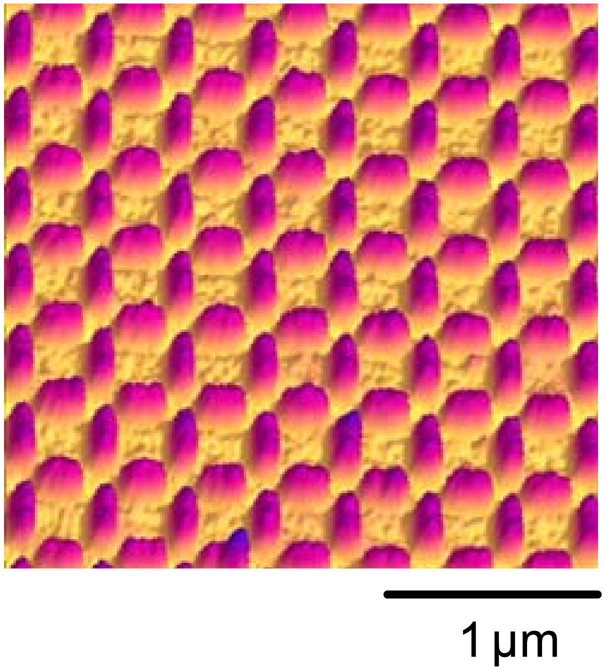


AFM image

- Patterned with e-beam lithography and lift-off technique.
- Composition: Permalloy (80% Ni + 20% Fe)
- Island size: 80nm * 220nm, 25nm thick
- Lattice parameter of 320 – 880 nm (center to center)

Important energy scales of islands

- Island size: 80nm * 220nm, 25nm thick

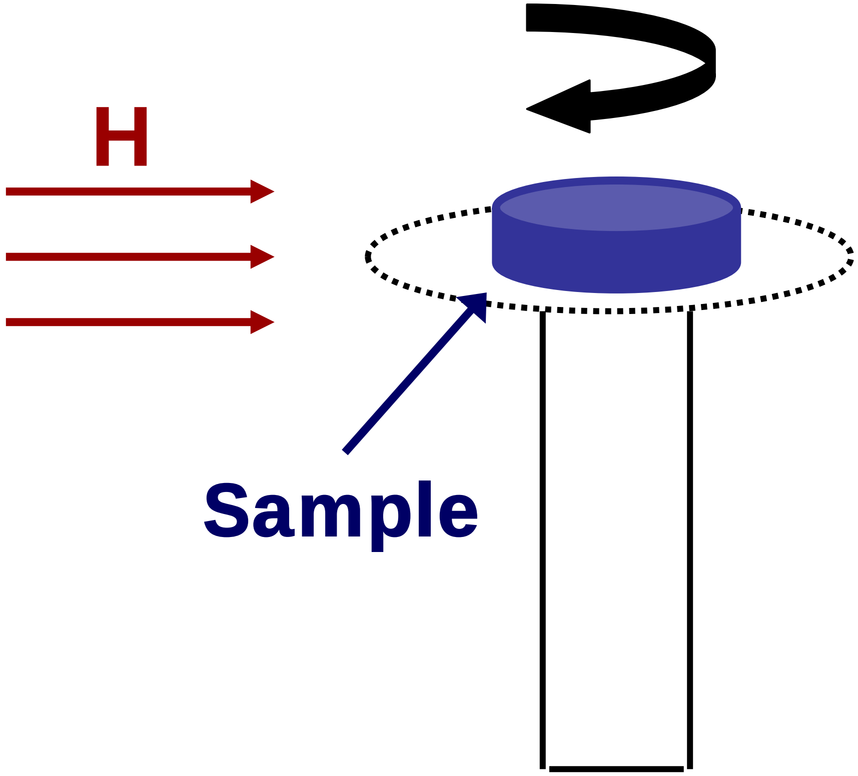


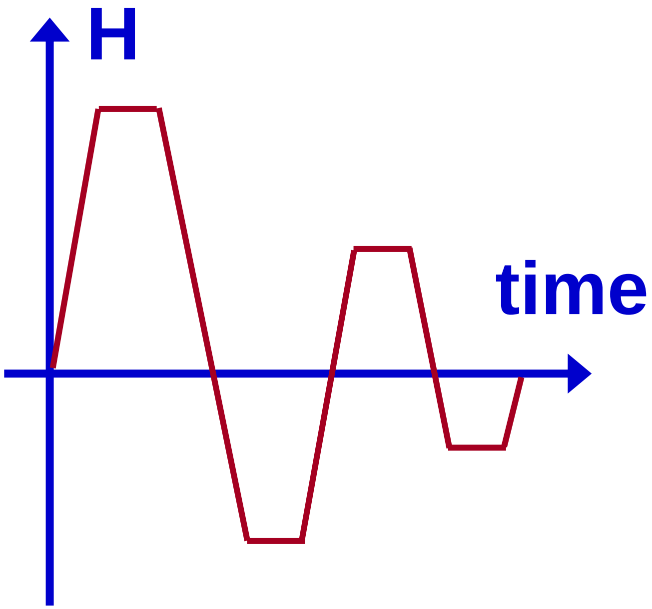
- Island moment: ~ 3×10^7 Bohr magnetons
- Zeeman energy of single island: ~ $2 * 10^4$ K in external field of 10 Oe
- Shape anisotropy energy of each island: ~ $4 * 10^6$ K
- Dipole-Dipole energy of two nearest neighbors: ~ 10^4 K depending on separation

Sample Preparation: How prepare magnetic state

Cannot use thermal energy to randomize magnetic moments (would need > 10^4 K)

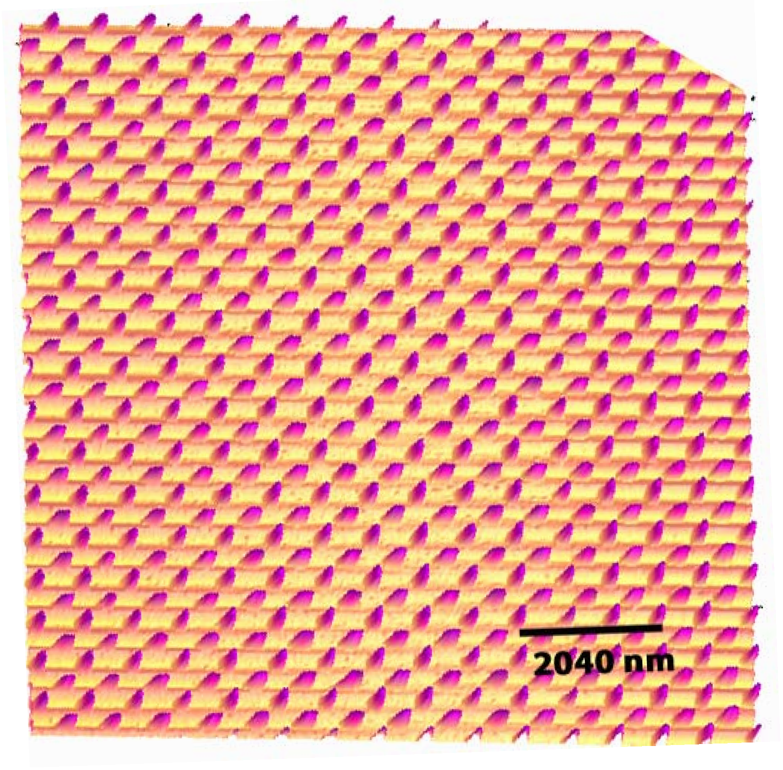
Instead rotate sample in a magnetic field which is stepped down in magnitude with switching polarity

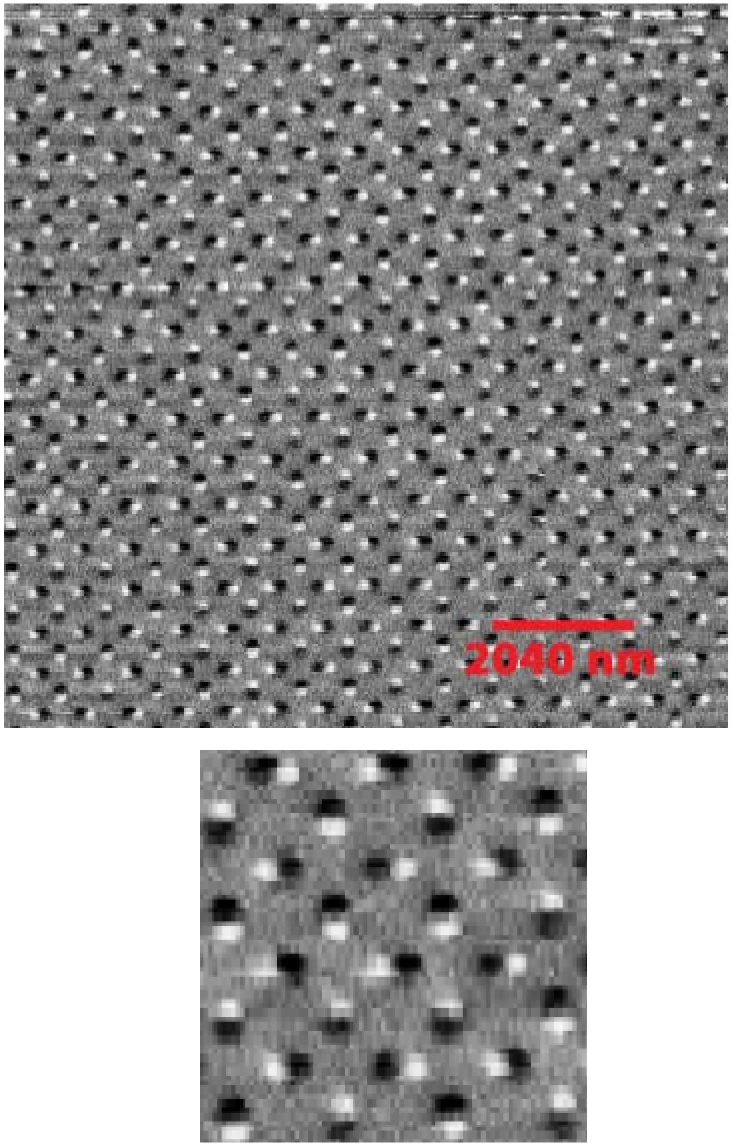




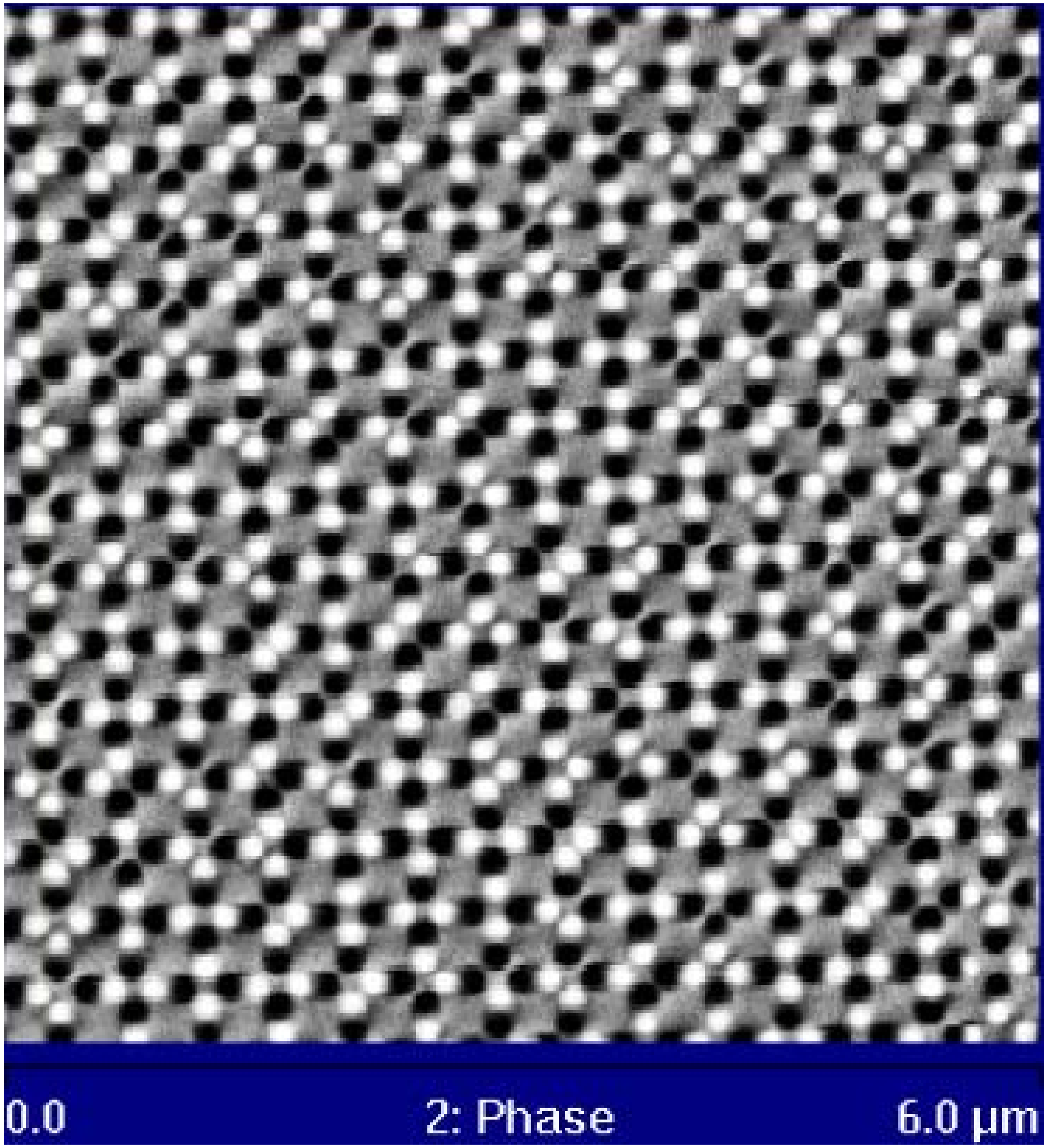
Use MFM to scan the arrangement of island moments

Can clearly see individual islands and single domain moments



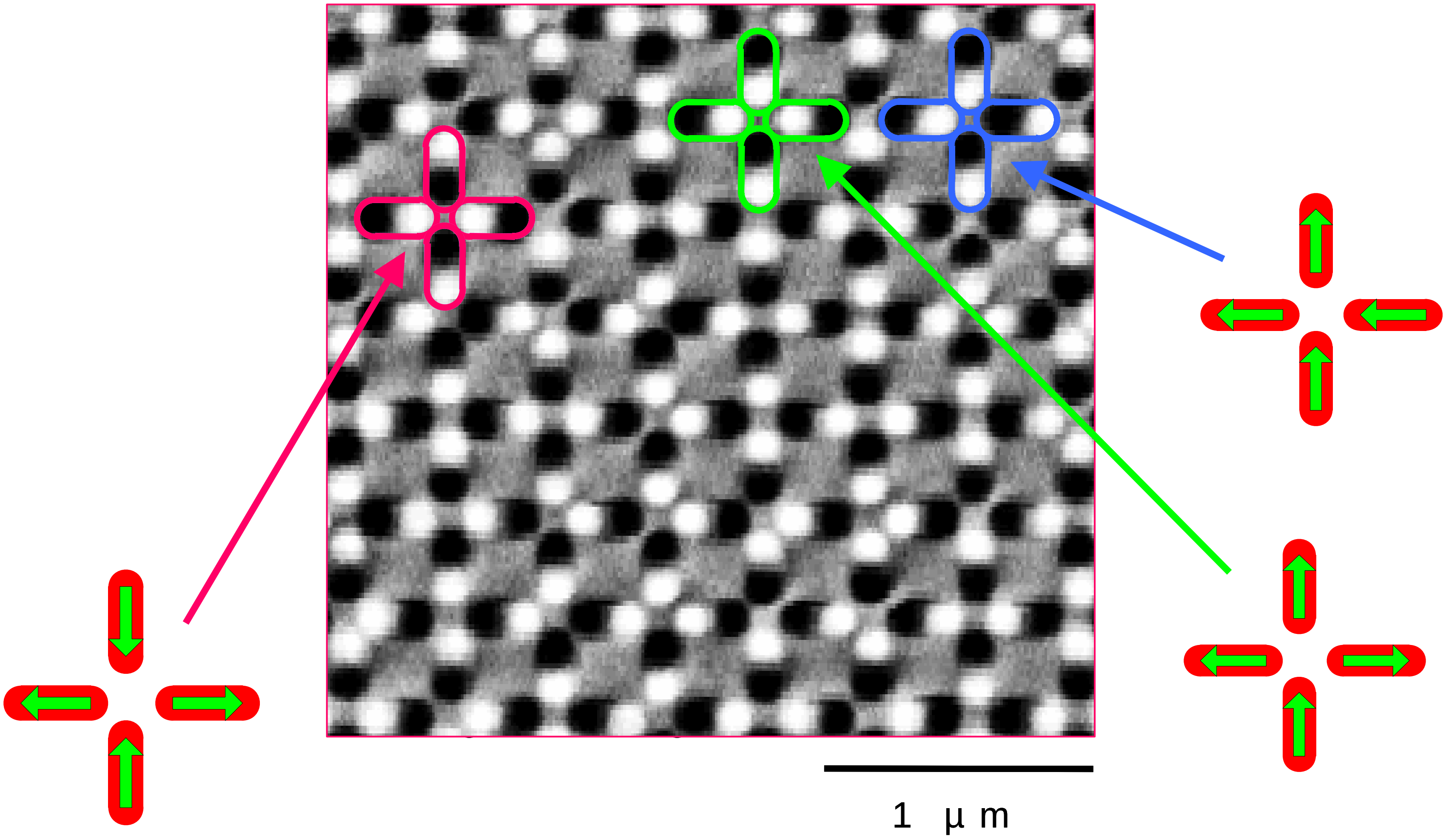


When islands are closely spaced, can clearly see disordered state



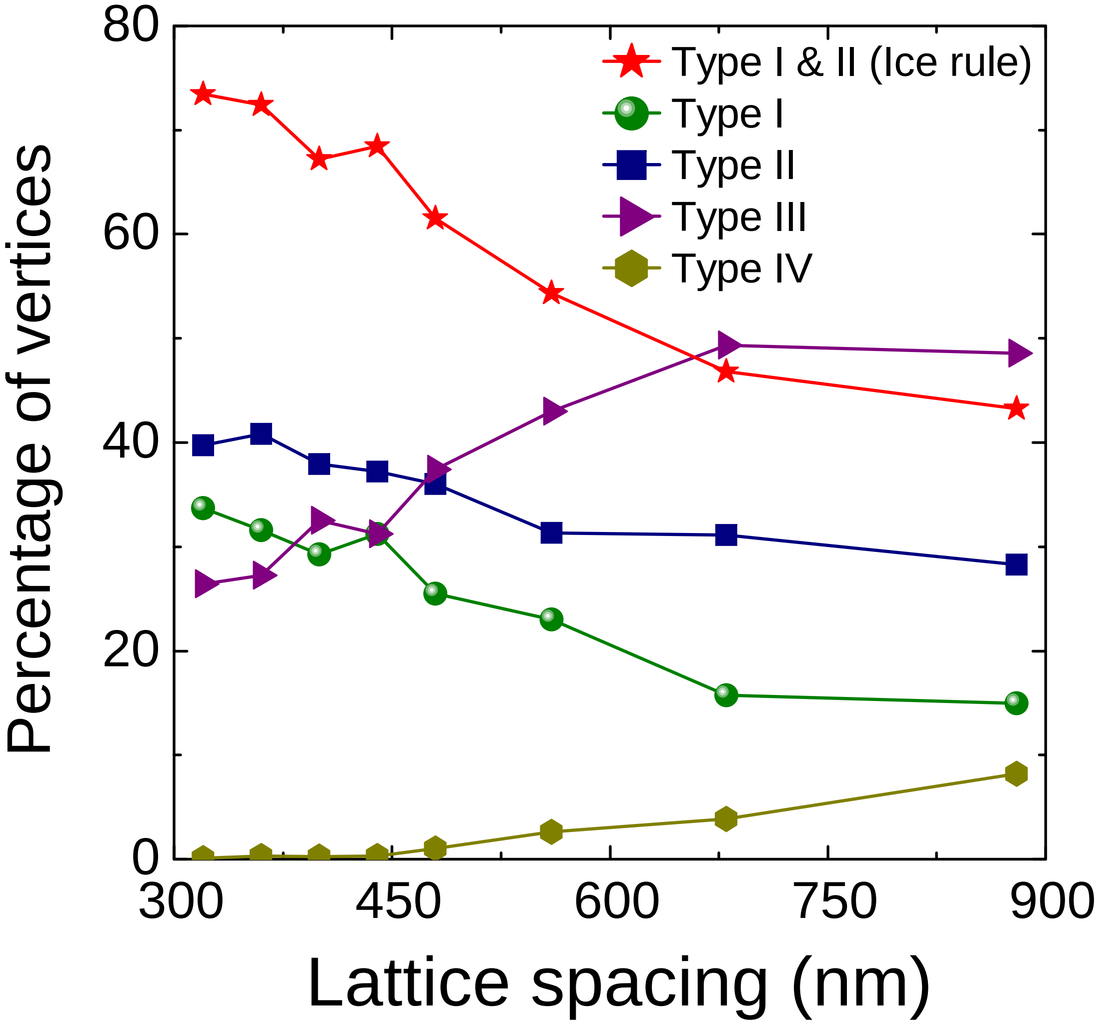
Use MFM to determine the arrangement of island moments

Can clearly see local moments and vertex types on the lattice

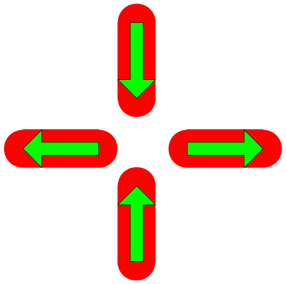


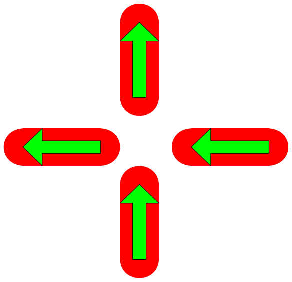
Distribution of vertices depends on island spacing

Count percentages of different vertex types



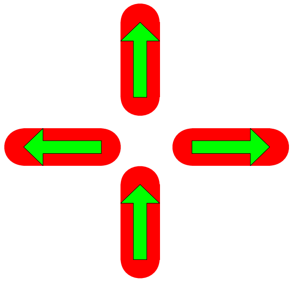
Type I (12.5%)

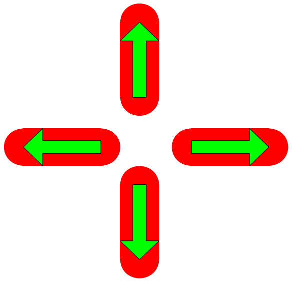




Type II (25%)

Type III (50%)

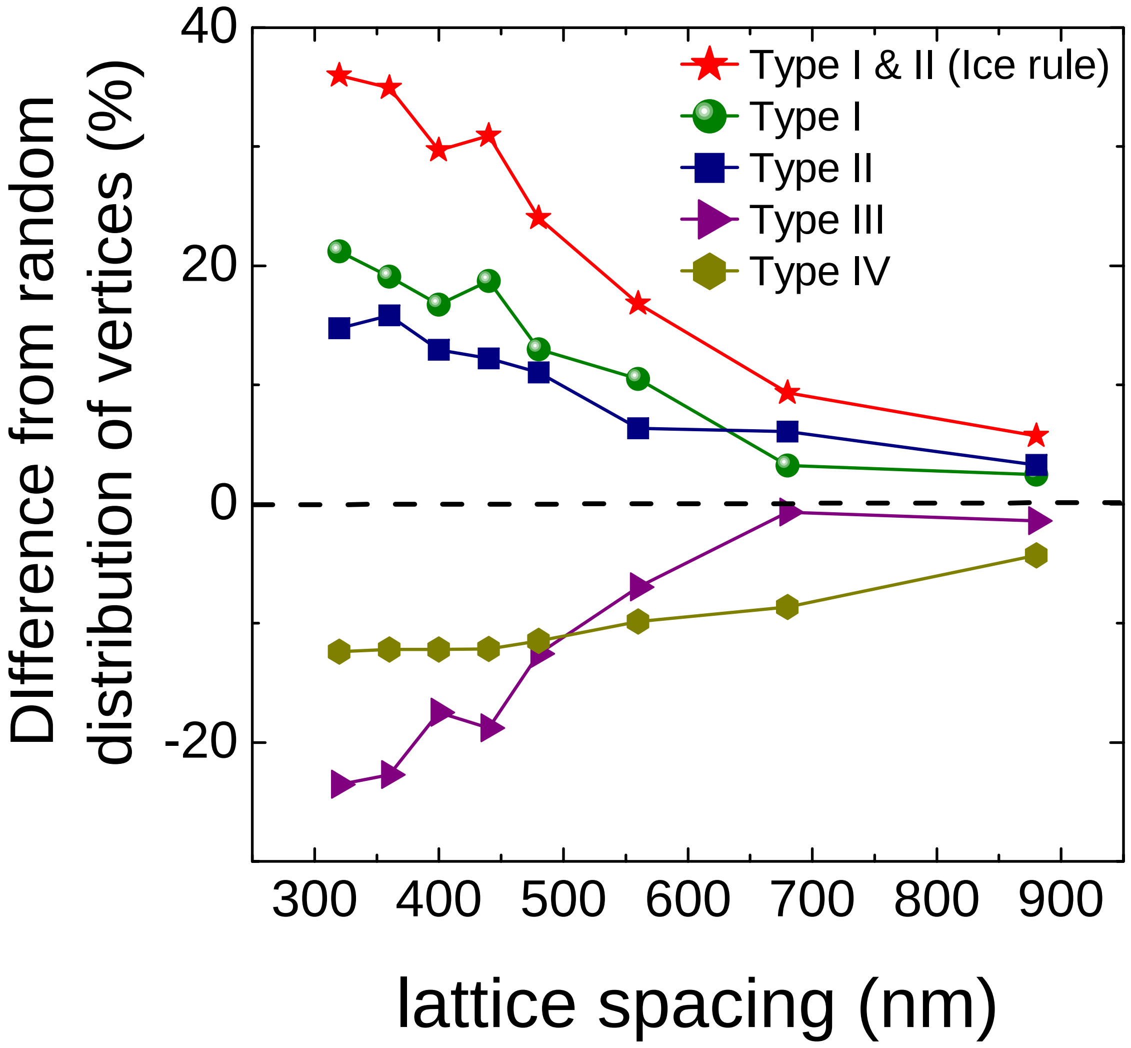




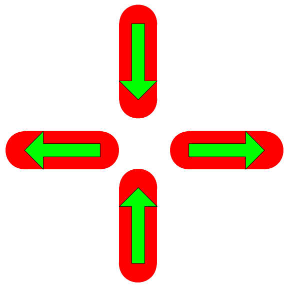
Type IV (12.5%)

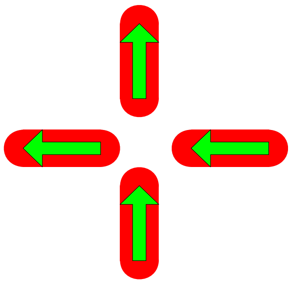
More ice-like vertices seen for closely spaced islands

As spacing increases, the percentages of vertex types converge to those expected for random moments



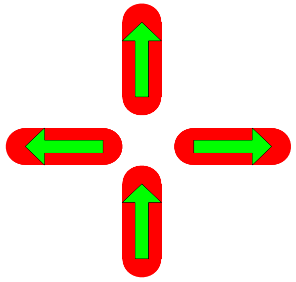
Type I (12.5%)

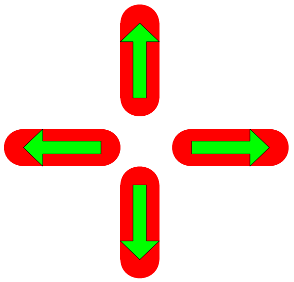




Type II (25%)

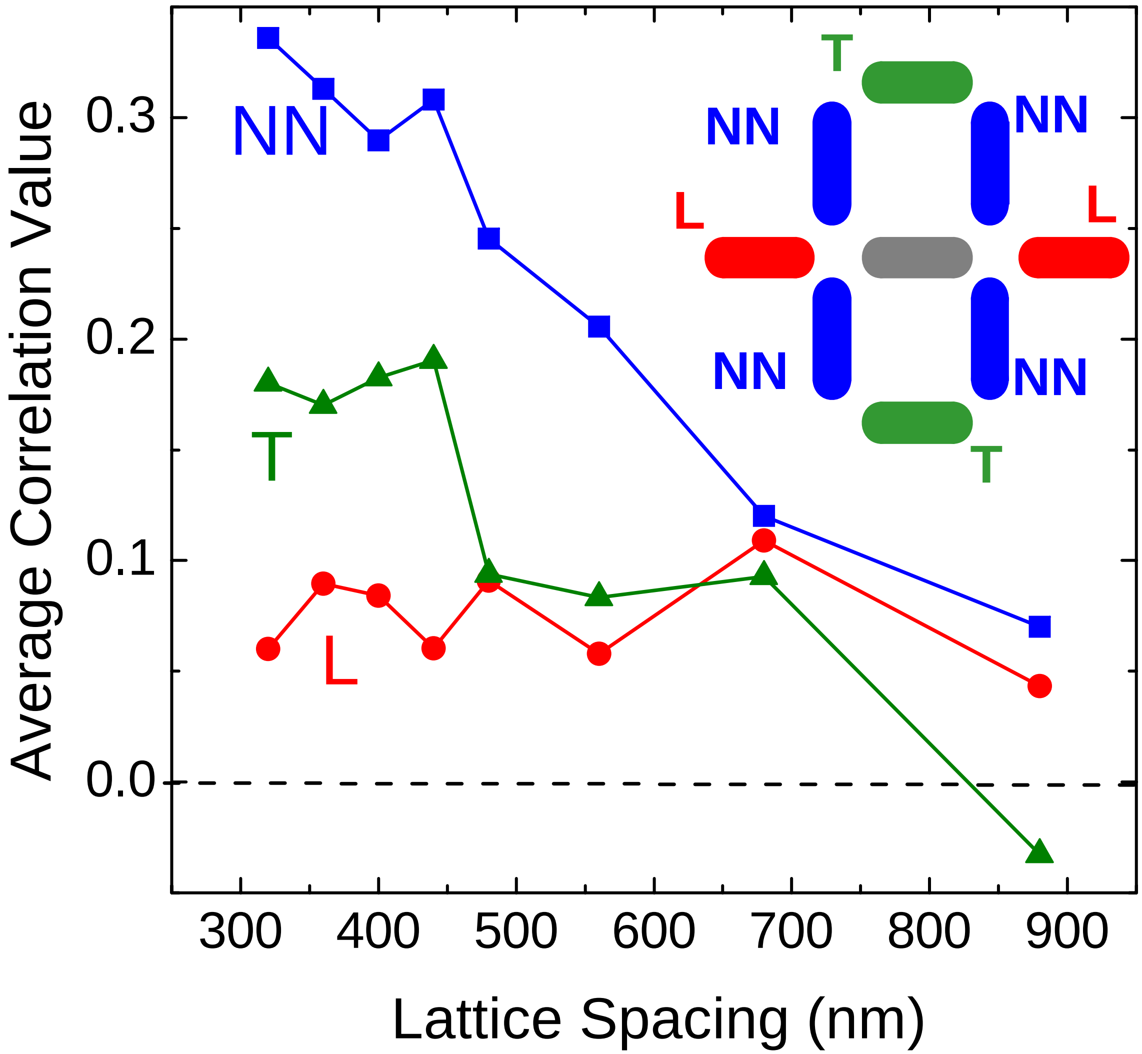
Type III (50%)





Type IV (12.5%)

Pair correlations decrease with increasing lattice spacing



Define correlation as:

+1 if pair minimizes dipole energy

→ →

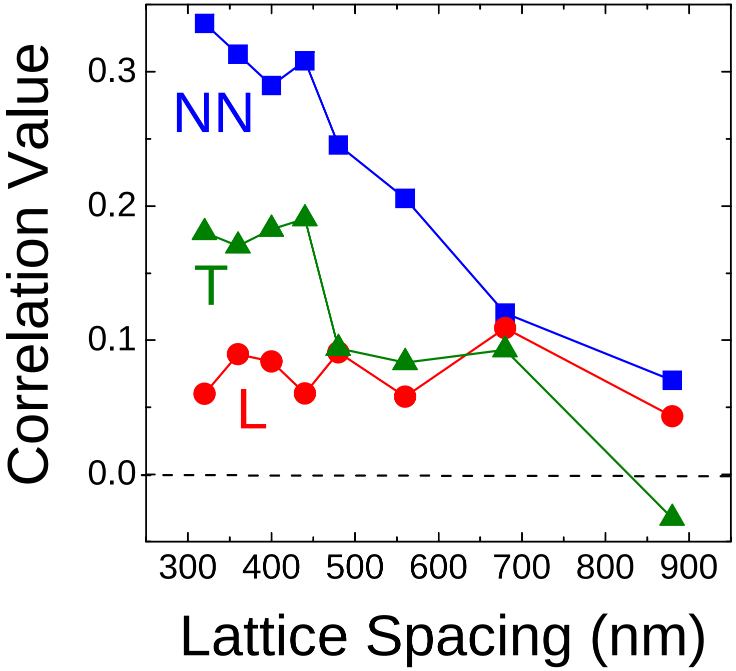
-1 if pair maximizes dipole energy

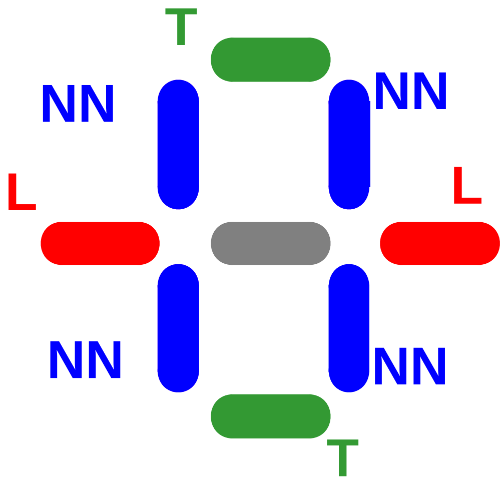
→ ←

Essentially no longer range correlations

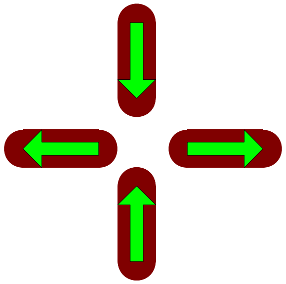
JUST LIKE PYROCHLORE ICE

Why correlations are stronger with transverse neighbors

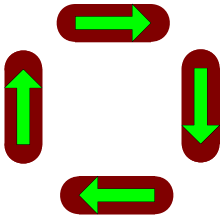




“L” neighbors are frustrated by NN interactions



“T” neighbors are not frustrated by NN interactions



Technology from interacting arrays: recent work suggests device possibilities

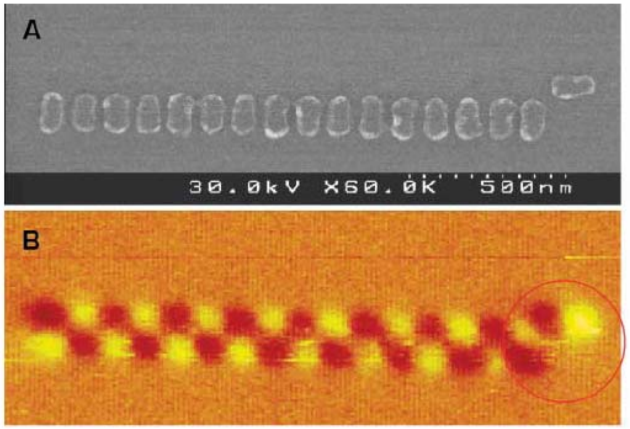


Fig. 2. Antiferromagnetic ordering in a line of nanomagnets. (**A**) Scanning electron microscope (SEM) image of a chain of 16 coupled nanomagnets of size 70 nm by 135 nm and 30-nm permalloy thickness. The separation between the vertically elongated magnets is 25 nm. The antiferromagnetic ordering along the chain is controlled by an additional, horizontally oriented elongated driver magnet. (**B**) MFM image of the same chain shows alternating magnetization of the magnets as set by the state of the horizontal driver magnet (circled).

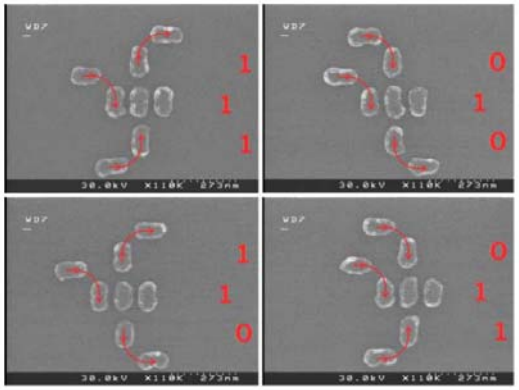


Fig. 3. Majority gates designed for testing all input combinations of the majority-logic operation. The arrows drawn superimposed on the SEM images illustrate the resulting magnetization direction due to a horizontally applied external clock-field. The magnetic state of the AFC inputs has the opposite effective votes on the central magnet as compared with the FC inputs, so AFC and FC inputs are assigned the same logical value for opposite magnetizations. The bit value "0" is assigned to magnetization direction down along the vertical axis of FC input magnets and the central magnet, and value "1" is assigned to magnetization up. The AFC input is defined oppositely.

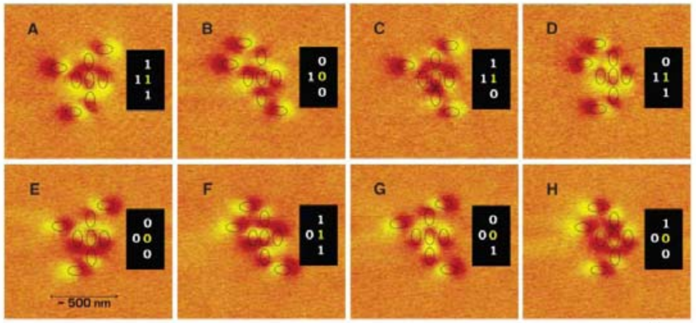


Fig. 4. MFM images of functioning majority gates. The location of the magnets is drawn superimposed on the MFM data. (**A** to **D**) Clock-field applied horizontally to the right and (**E** to **H**) to the left. Bit values assigned to the magnetization directions can be determined by the MFM contrast. Bit values shown in (**I**) are for FC inputs and central magnet. (AFC inputs are designated with the inverse logical values.) The black insets show alignment of magnetic dipoles, accounting for antiferromagnetic and ferromagnetic coupling, and demonstrate correct MQCA majority logic gate functionality.

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Notre Dame group

Imre *et al.*

Science, 2006

Artificial frustrated systems:
the fun is just beginning…

New control of frustrated lattices is possible

- make different lattices
- control strength of interactions
- put in defects

New measurements

- look at dynamics in magnetic field
- push to superparamagnetic limit
- measure entropy to compare to “real” spin ice

Perhaps relate to recording on patterned media….