Frustrated Magnetism

Leon Balents
ICTS winter school, December 2009

The David and Lucile Packard Foundation

Recent Collaborators

- Doron Bergman
- Gang Chen
- Sungbin Lee
- Miles Stoudenmire
- Ribhu Kaul
- Jason Alicea
- Ryuichi Shindou
- Andreas Schnyder
- Yong-Baek Kim
- Arun Paramekanti
- Michael Lawler
- Lucile Savary
- Simon Trebst
- Emmanuel Gull
- Oleg Starykh
- Masanori Kohno
- Hosho Katsura
- Dmytro Pesin

Lesson Plan*

- Introduction: basic magnetism, empirical signs of frustration
- Classical Ising systems and spin ice
- Heisenberg systems and quantum effects
- Some interesting examples
- Quantum spin liquids

*Subject to change according to my whims

The setting

- We will be discussing Mott insulators, in which electrons can be regarded as localized into specific atomic or molecular orbitals
  - In this case, the degrees of freedom are the spin and sometimes orbital state of these electrons
  - This is vast simplification over itinerant systems

Hubbard Model

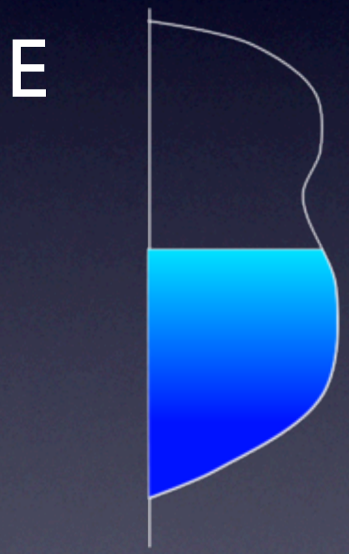
- When is the localized assumption valid?
- Useful to keep in mind a Hubbard-type model

$$H = \sum_{ij} t_{ij} c^\dagger_{i\alpha} c_{j\alpha} + U \sum_i (n_i - \overline{n})^2$$

- If average electron number per site is integer, and U/t is large enough, then the ground state is a Mott insulator

$U_c \sim W$

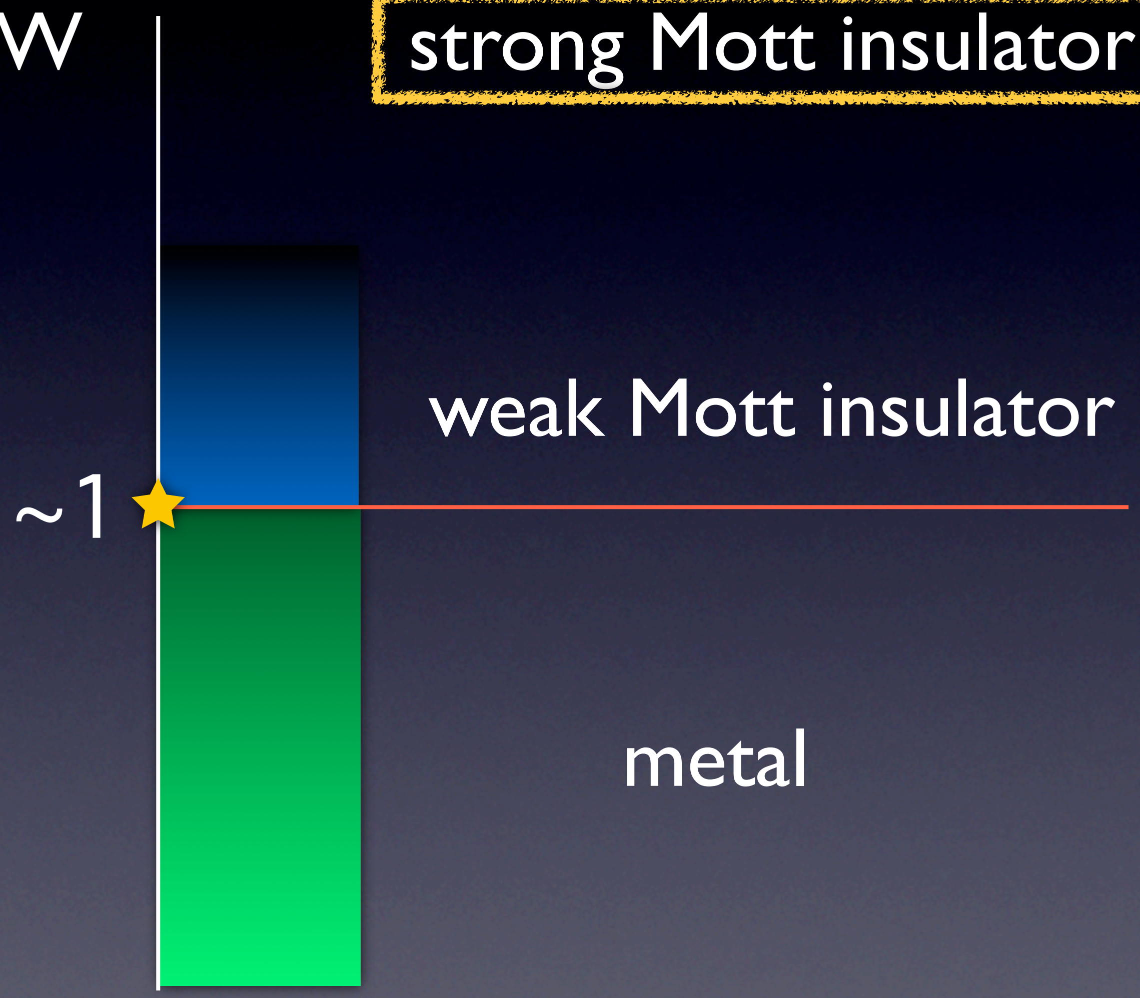
- Rule of thumb: compare U to bandwidth W



delocalized: pay Coulomb of O(U)

localized: pay KE of O(W)

Phase diagram

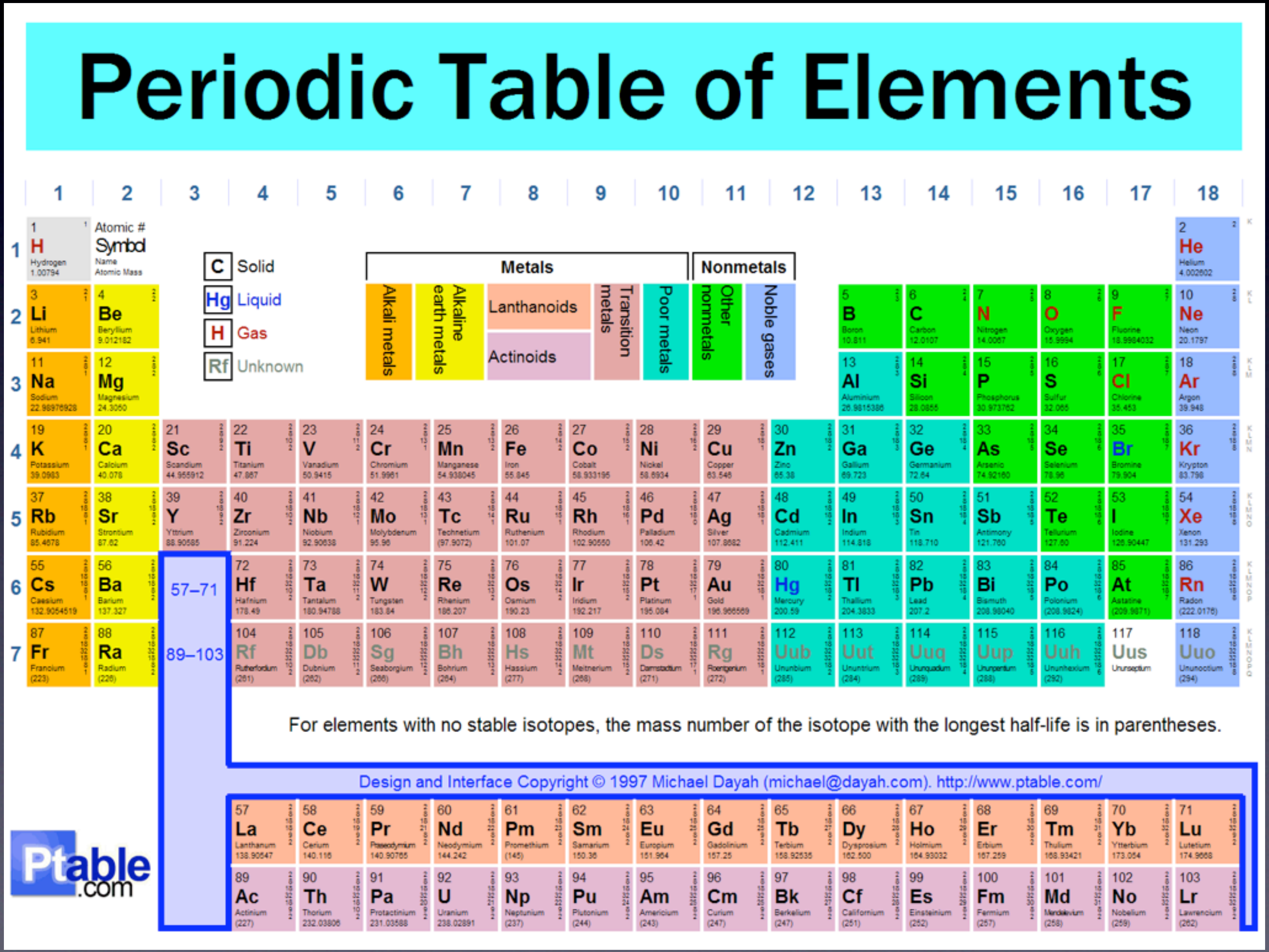


strong Mott insulator

weak Mott insulator

metal

Chemistry



Periodic Table of Elements

C Solid
Hg Liquid
H Gas
Rf Unknown

Metals: Alkali metals, Alkaline earth metals, Lanthanoids, Actinoids, Transition metals, Poor metals
Nonmetals: Other nonmetals, Noble gases

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H Hydrogen 1.00794	Atomic # Symbol Name Atomic Mass																2 He Helium 4.002602
2	3 Li Lithium 6.941	4 Be Beryllium 9.012182											5 B Boron 10.811	6 C Carbon 12.0107	7 N Nitrogen 14.0067	8 O Oxygen 15.9994	9 F Fluorine 18.9984032	10 Ne Neon 20.1797
3	11 Na Sodium 22.98976928	12 Mg Magnesium 24.3050											13 Al Aluminium 26.9815386	14 Si Silicon 28.0855	15 P Phosphorus 30.973762	16 S Sulfur 32.065	17 Cl Chlorine 35.453	18 Ar Argon 39.948
4	19 K Potassium 39.0983	20 Ca Calcium 40.078	21 Sc Scandium 44.955912	22 Ti Titanium 47.867	23 V Vanadium 50.9415	24 Cr Chromium 51.9961	25 Mn Manganese 54.938045	26 Fe Iron 55.845	27 Co Cobalt 58.933195	28 Ni Nickel 58.6934	29 Cu Copper 63.546	30 Zn Zinc 65.38	31 Ga Gallium 69.723	32 Ge Germanium 72.64	33 As Arsenic 74.92160	34 Se Selenium 78.96	35 Br Bromine 79.904	36 Kr Krypton 83.798
5	37 Rb Rubidium 85.4678	38 Sr Strontium 87.62	39 Y Yttrium 88.90585	40 Zr Zirconium 91.224	41 Nb Niobium 92.90638	42 Mo Molybdenum 95.96	43 Tc Technetium (97.9072)	44 Ru Ruthenium 101.07	45 Rh Rhodium 102.90550	46 Pd Palladium 106.42	47 Ag Silver 107.8682	48 Cd Cadmium 112.411	49 In Indium 114.818	50 Sn Tin 118.710	51 Sb Antimony 121.760	52 Te Tellurium 127.60	53 I Iodine 126.90447	54 Xe Xenon 131.293
6	55 Cs Caesium 132.9054519	56 Ba Barium 137.327	57–71	72 Hf Hafnium 178.49	73 Ta Tantalum 180.94788	74 W Tungsten 183.84	75 Re Rhenium 186.207	76 Os Osmium 190.23	77 Ir Iridium 192.217	78 Pt Platinum 195.084	79 Au Gold 196.966569	80 Hg Mercury 200.59	81 Tl Thallium 204.3833	82 Pb Lead 207.2	83 Bi Bismuth 208.98040	84 Po Polonium (208.9824)	85 At Astatine (209.9871)	86 Rn Radon (222.0176)
7	87 Fr Francium (223)	88 Ra Radium (226)	89–103	104 Rf Rutherfordium (261)	105 Db Dubnium (262)	106 Sg Seaborgium (266)	107 Bh Bohrium (264)	108 Hs Hassium (277)	109 Mt Meitnerium (268)	110 Ds Darmstadtium (271)	111 Rg Roentgenium (272)	112 Uub Ununbium (285)	113 Uut Ununtrium (284)	114 Uuq Ununquadium (289)	115 Uup Ununpentium (288)	116 Uuh Ununhexium (292)	117 Uus Ununseptium	118 Uuo Ununoctium (294)

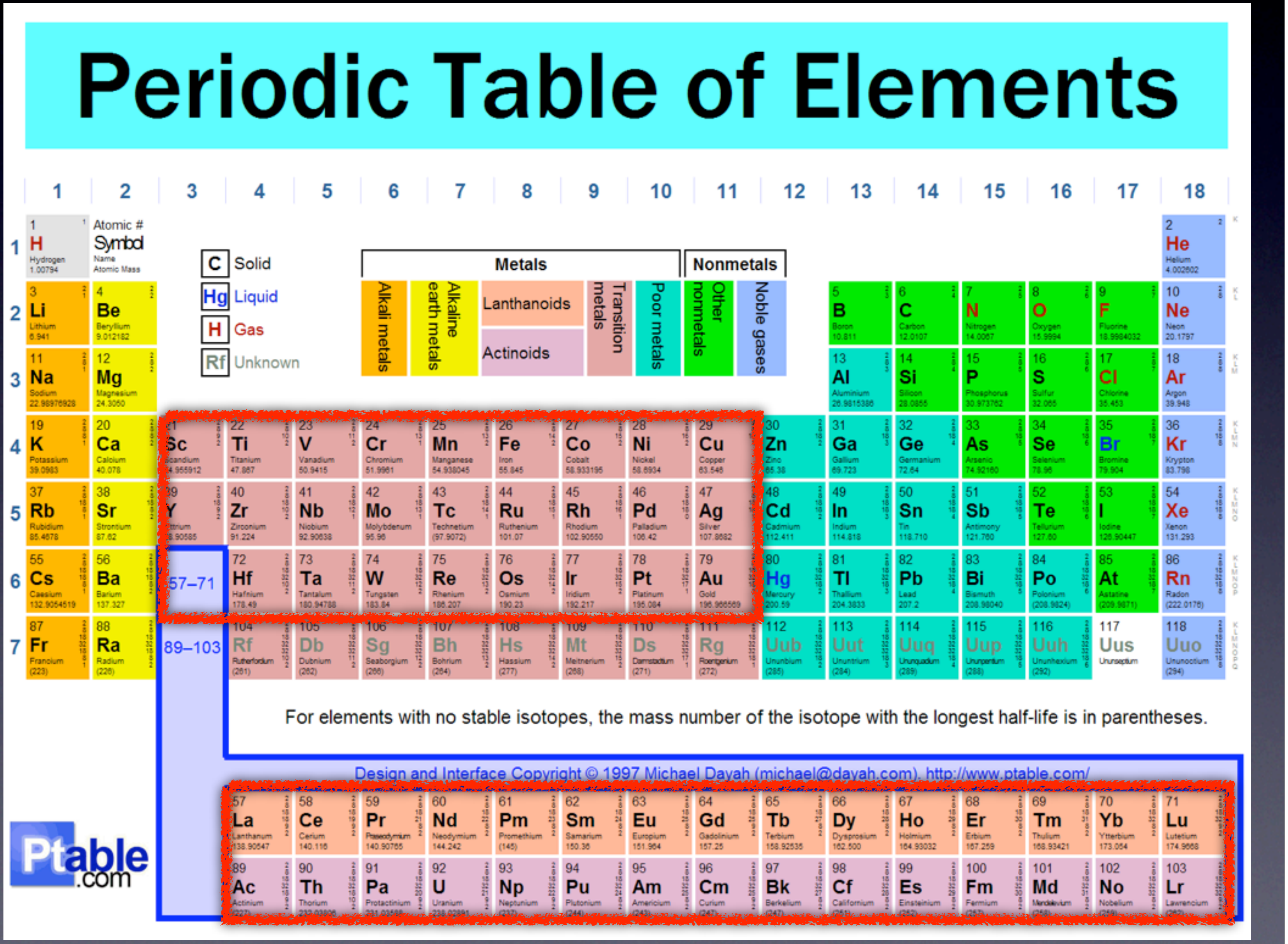
For elements with no stable isotopes, the mass number of the isotope with the longest half-life is in parentheses.

Design and Interface Copyright © 1997 Michael Dayah (michael@dayah.com). http://www.ptable.com/

57 La Lanthanum 138.90547	58 Ce Cerium 140.116	59 Pr Praseodymium 140.90765	60 Nd Neodymium 144.242	61 Pm Promethium (145)	62 Sm Samarium 150.36	63 Eu Europium 151.964	64 Gd Gadolinium 157.25	65 Tb Terbium 158.92535	66 Dy Dysprosium 162.500	67 Ho Holmium 164.93032	68 Er Erbium 167.259	69 Tm Thulium 168.93421	70 Yb Ytterbium 173.054	71 Lu Lutetium 174.9668
89 Ac Actinium (227)	90 Th Thorium 232.03806	91 Pa Protactinium 231.03588	92 U Uranium 238.02891	93 Np Neptunium (237)	94 Pu Plutonium (244)	95 Am Americium (243)	96 Cm Curium (247)	97 Bk Berkelium (247)	98 Cf Californium (251)	99 Es Einsteinium (252)	100 Fm Fermium (257)	101 Md Mendelevium (258)	102 No Nobelium (259)	103 Lr Lawrencium (262)

Chemistry

Small W:
d and f
electrons



Periodic Table of Elements

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	57–71	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89–103	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo

Atomic #
Symbol
Name
Atomic Mass

C Solid
Hg Liquid
H Gas
Rf Unknown

Metals: Alkali metals, Alkaline earth metals, Lanthanoids, Actinoids, Transition metals, Poor metals

Nonmetals: Other nonmetals, Noble gases

For elements with no stable isotopes, the mass number of the isotope with the longest half-life is in parentheses.

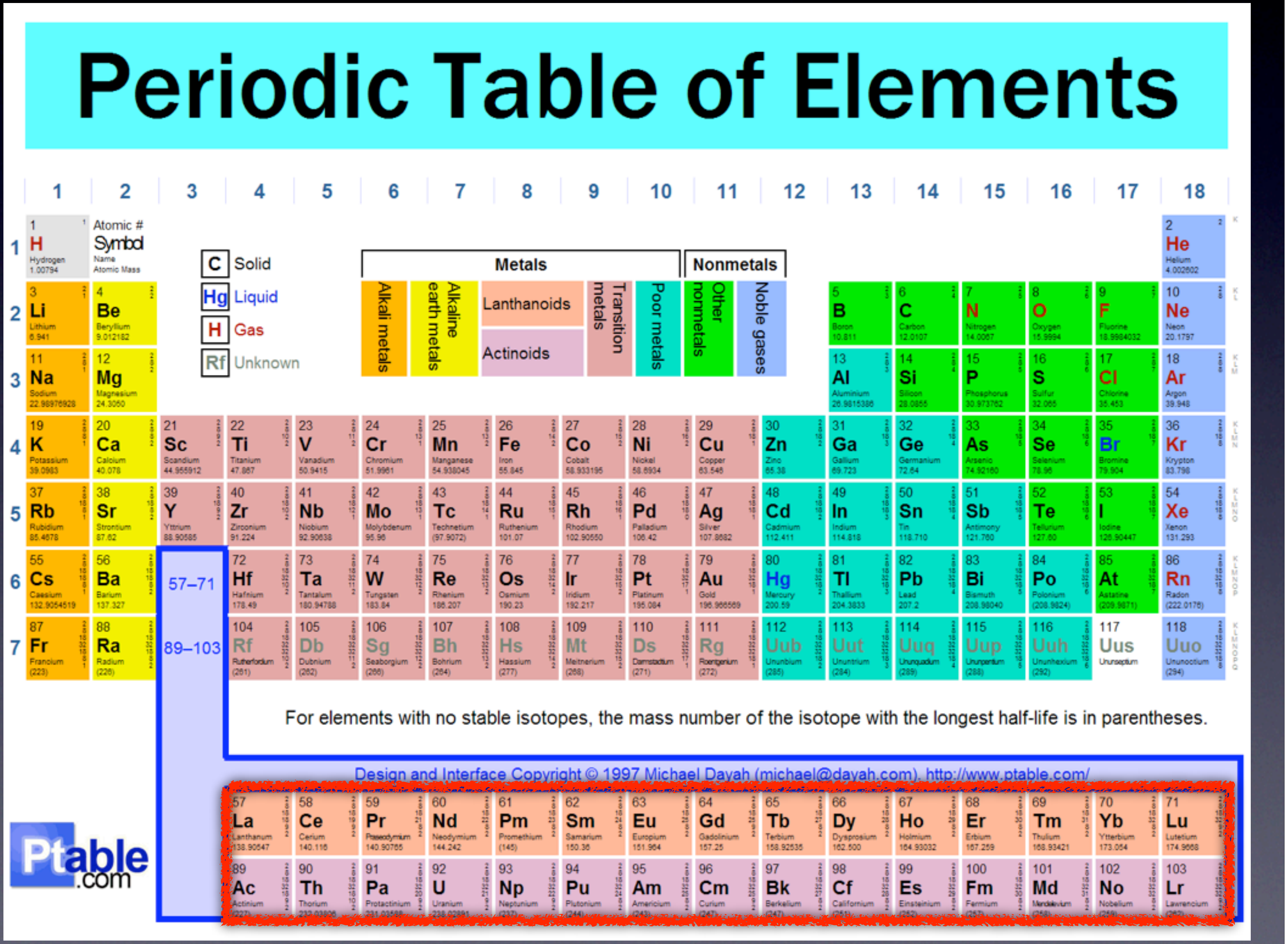
Design and Interface Copyright © 1997 Michael Dayah (michael@dayah.com). http://www.ptable.com/

57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Ptable.com

Chemistry

f electrons are nearly perfectly localized - very small W



Periodic Table of Elements

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H Hydrogen 1.00794																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	57–71	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89–103	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo

Atomic #
Symbol
Name
Atomic Mass

C Solid
Hg Liquid
H Gas
Rf Unknown

Metals: Alkali metals, Alkaline earth metals, Lanthanoids, Actinoids, Transition metals, Poor metals

Nonmetals: Other nonmetals, Noble gases

For elements with no stable isotopes, the mass number of the isotope with the longest half-life is in parentheses.

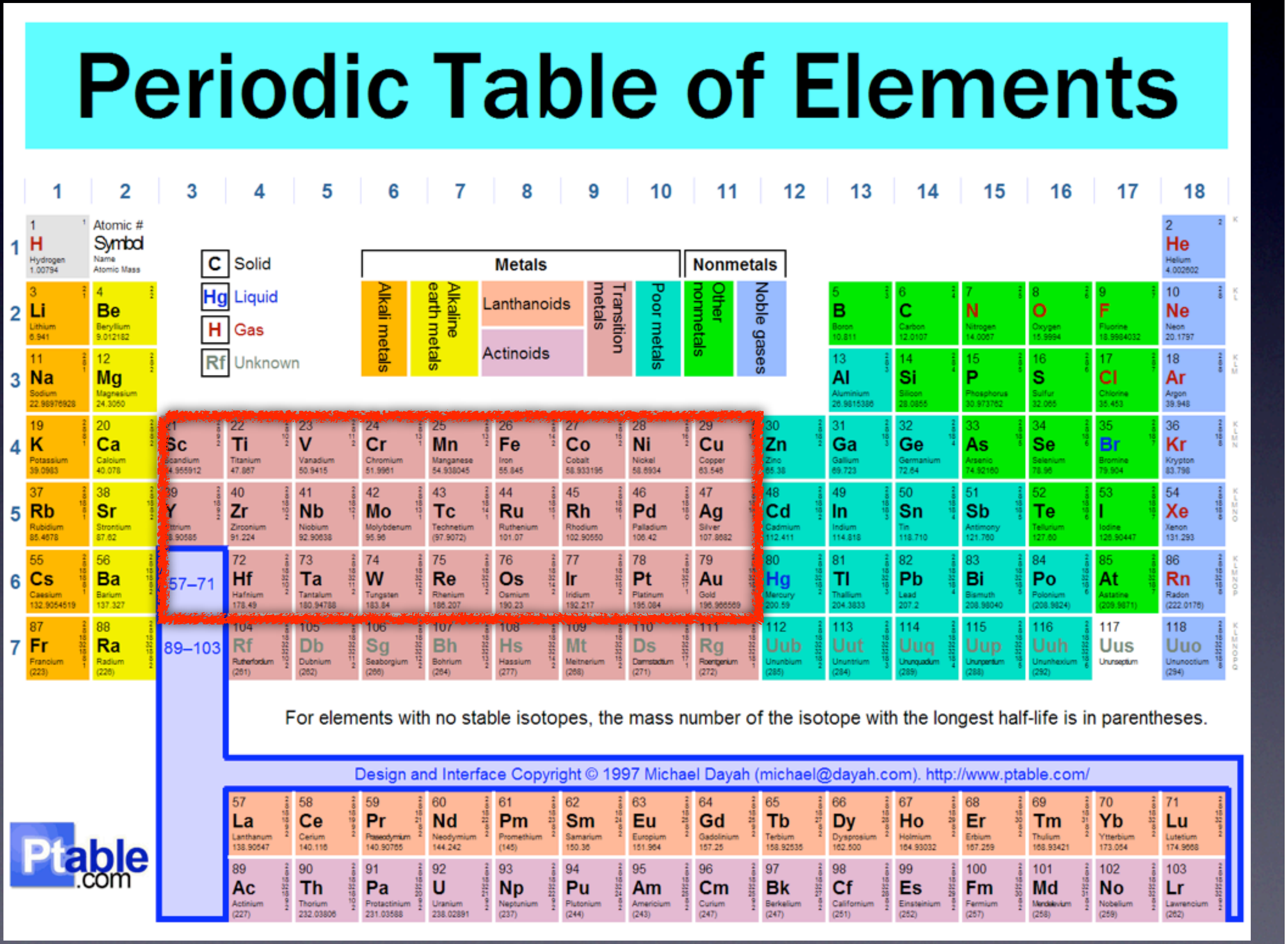
57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Ptable.com

Chemistry

Decreasing U with n: $U_{3d}>U_{4d}>U_{5d}$

Strongest correlations in 3d TMs



Periodic Table of Elements

Atomic #
Symbol
Name
Atomic Mass

C Solid
Hg Liquid
H Gas
Rf Unknown

Metals: Alkali metals, Alkaline earth metals, Lanthanoids, Actinoids, Transition metals, Poor metals

Nonmetals: Other nonmetals, Noble gases

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	57–71	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89–103	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo

For elements with no stable isotopes, the mass number of the isotope with the longest half-life is in parentheses.

Design and Interface Copyright © 1997 Michael Dayah (michael@dayah.com). http://www.ptable.com/

57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Ptable.com

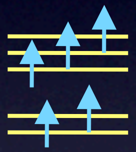
Chemistry and Structure

- Most Mott insulators have some ionic character - often oxides
  - TM or RE atoms should donate their s electrons
- Bandwidth can be substantially reduced by separating TMs by filled shell ions like O^{2-}

How to tell?

- In practice, it is often useful to rely on experiments to tell you how localized the electrons are
- resistivity - is it a good insulator? if you can measure it, it is probably not!
- optics - measure optical gap.
- are there local moments?

Local moment magnetism

- Atoms with partially filled shells
  - Hund's rules give magnetic state  e.g. Mn^{2+}
  - These moments are well-formed for $k_B T \ll U$
  - Exchange between moments $J \sim t^2/U$
- When $U \gg k_B T \gg J$, see Curie law

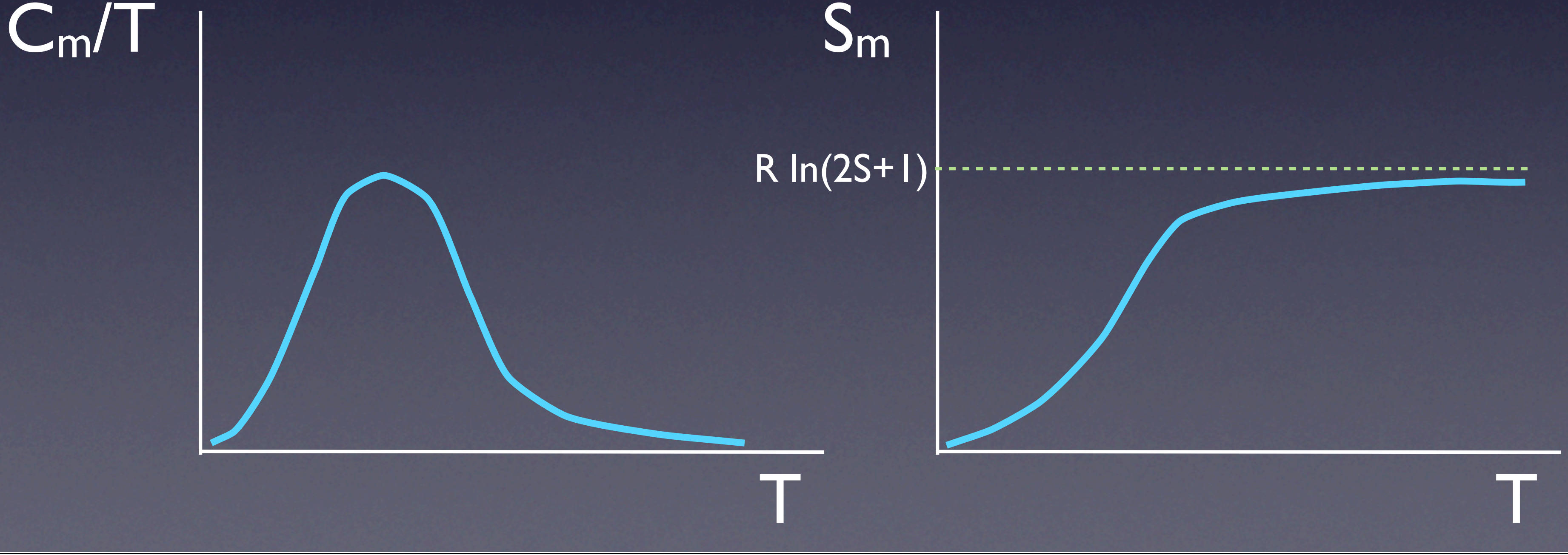
$$\chi \sim \frac{A}{T} \qquad A = \frac{N g^2 \mu_B^2 S(S+1)}{3}$$

Curie constant

Entropy

- If you can separate non-magnetic contribution, then you can count states

$$S(T) = \int_0^T dT' \frac{C(T')}{T'}$$



Frustration

Spin models

- In a strong Mott insulator, we can assume n_i is fixed and just study the spin (and perhaps orbital) state of the electrons
- e.g. Heisenberg Hamiltonian

$$H_{eff} = \frac{1}{2}\sum_{ij} J_{ij}\vec{S}_i \cdot \vec{S}_j$$

- Exchange couplings $|J_{ij}| \sim (t_{ij})^2/U$
- More complex Hamiltonians may be less symmetric, and involve orbital operators

Frustration

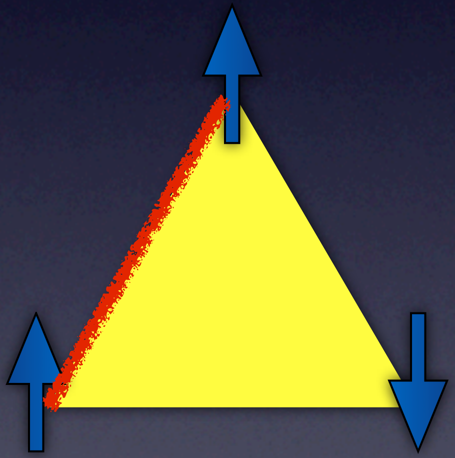
- Exchange interactions usually favor a magnetically ordered state

$$\langle \vec{S}_i \rangle \neq 0$$

- The spins act approximately classically, and align to minimize H_{eff}
- However, in some cases there is no single, simple way to do this

Frustration

- Simplest idea: pairwise exchange interactions cannot be simultaneously satisfied

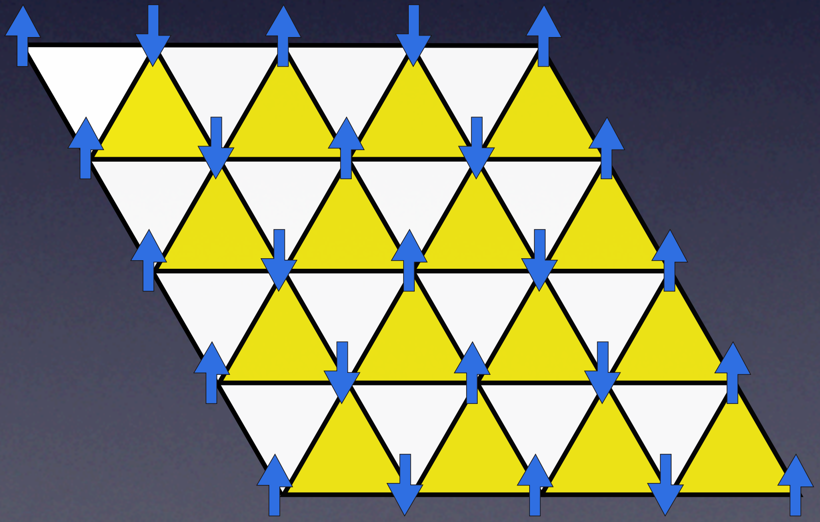


“geometric frustration”

- But this is a bit simplistic, and overstates the problem

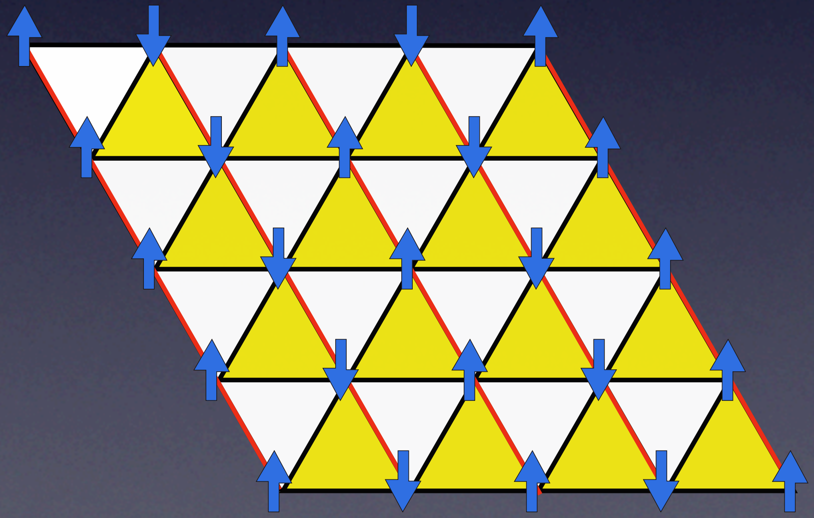
Degeneracy

- Characterize frustration by number of ground states
- Ising models $H = J \sum_{\langle ij \rangle} \sigma_i \sigma_j$ $\sigma_i = \pm 1$



Degeneracy

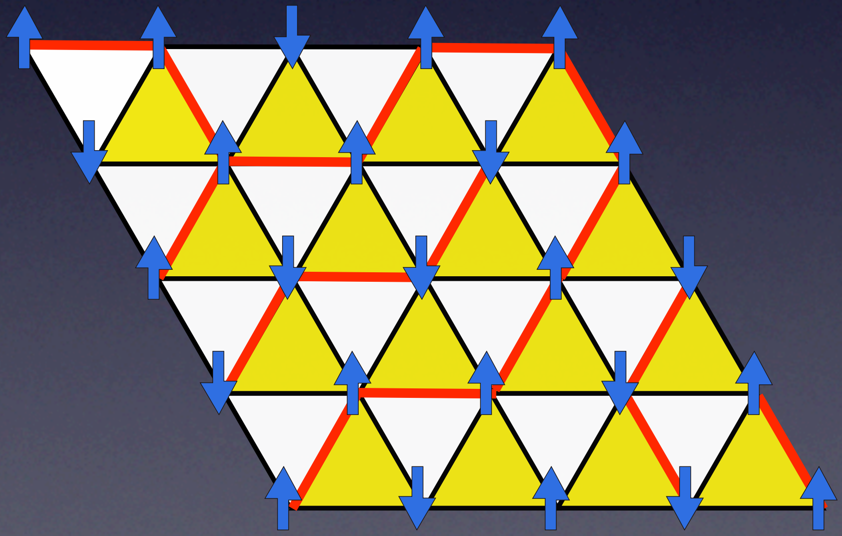
- Characterize frustration by number of ground states
- Ising models



1 frustrated bond per triangle

Degeneracy

- Characterize frustration by number of ground states
- Ising models

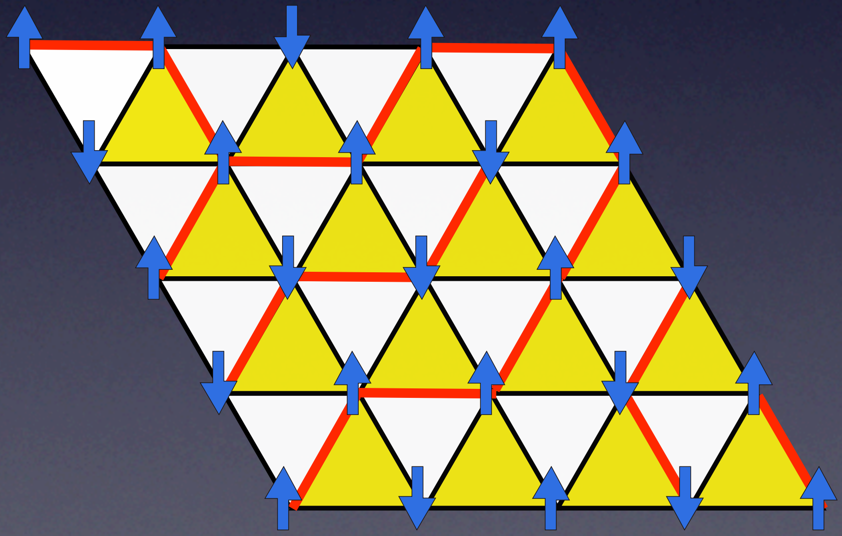


1 frustrated bond per triangle

exponentially many ground states

Degeneracy

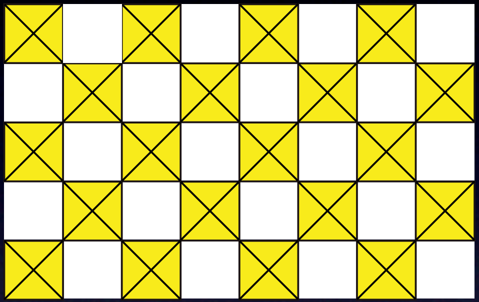
- Characterize frustration by number of ground states
- Ising models



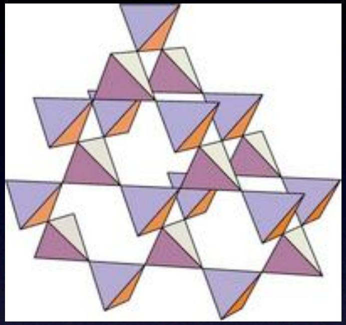
1 frustrated bond per triangle

Wannier (1950): $\Omega = e^{S/k_B}$ $S \approx 0.34 N k_B$

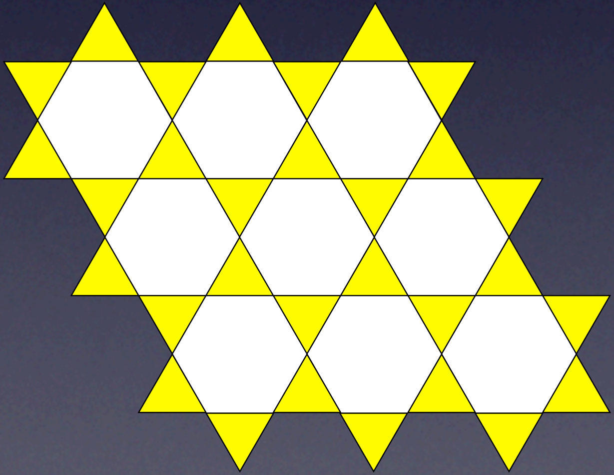
Other lattices



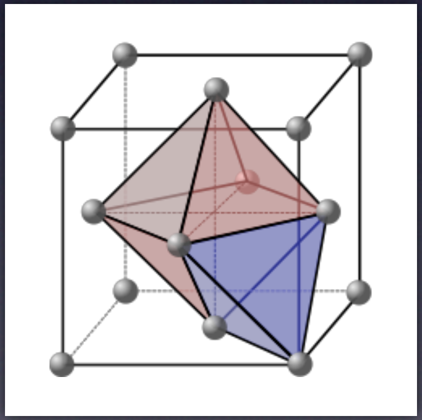
checkerboard S ~ 0.216 N k_B



pyrochlore S ~ 0.203 N k_B



kagome S ~ 0.5 N k_B



FCC: S ~ c $N^{1/3}$ k_B

But...

- Such an Ising model is very special
  - Not so common to find simple Ising spins
  - Generally there are more interactions
  - This degeneracy is very finely tuned
- In practice, we will usually need to think about more subtle models
  - How do we look for frustration if we are not sure of the model?

Looking for Frustration

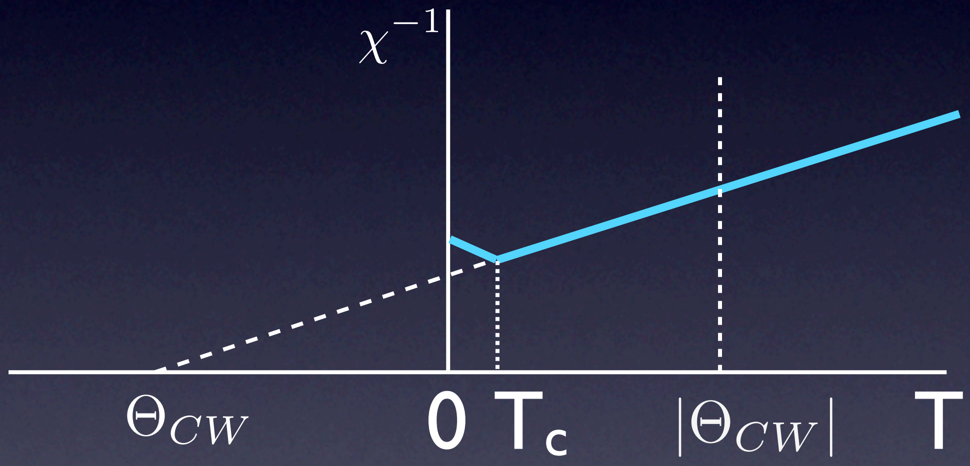
- We are looking to see that, instead of ordering, the system fluctuates amongst the many degenerate states even when $k_B T << J$
- To determine this empirically, we need to have an experimental estimate of "J" and also put an upper bound on the ordering/ freezing temperature

Curie-Weiss Law

- Antiferromagnetic exchange leads to *suppression* of susceptibility
- mean field theory/high temperature expansion
- Curie-Weiss temperature $$\chi \sim \frac{A}{T - \Theta_{CW}}$$
- $$\Theta_{CW} = -\left(\sum_j J_{ij}\right)\frac{S(S+1)}{3k_B}$$ (<0 in AFs)

Frustration "fingerprint"

- Experimental plot of inverse susceptibility:



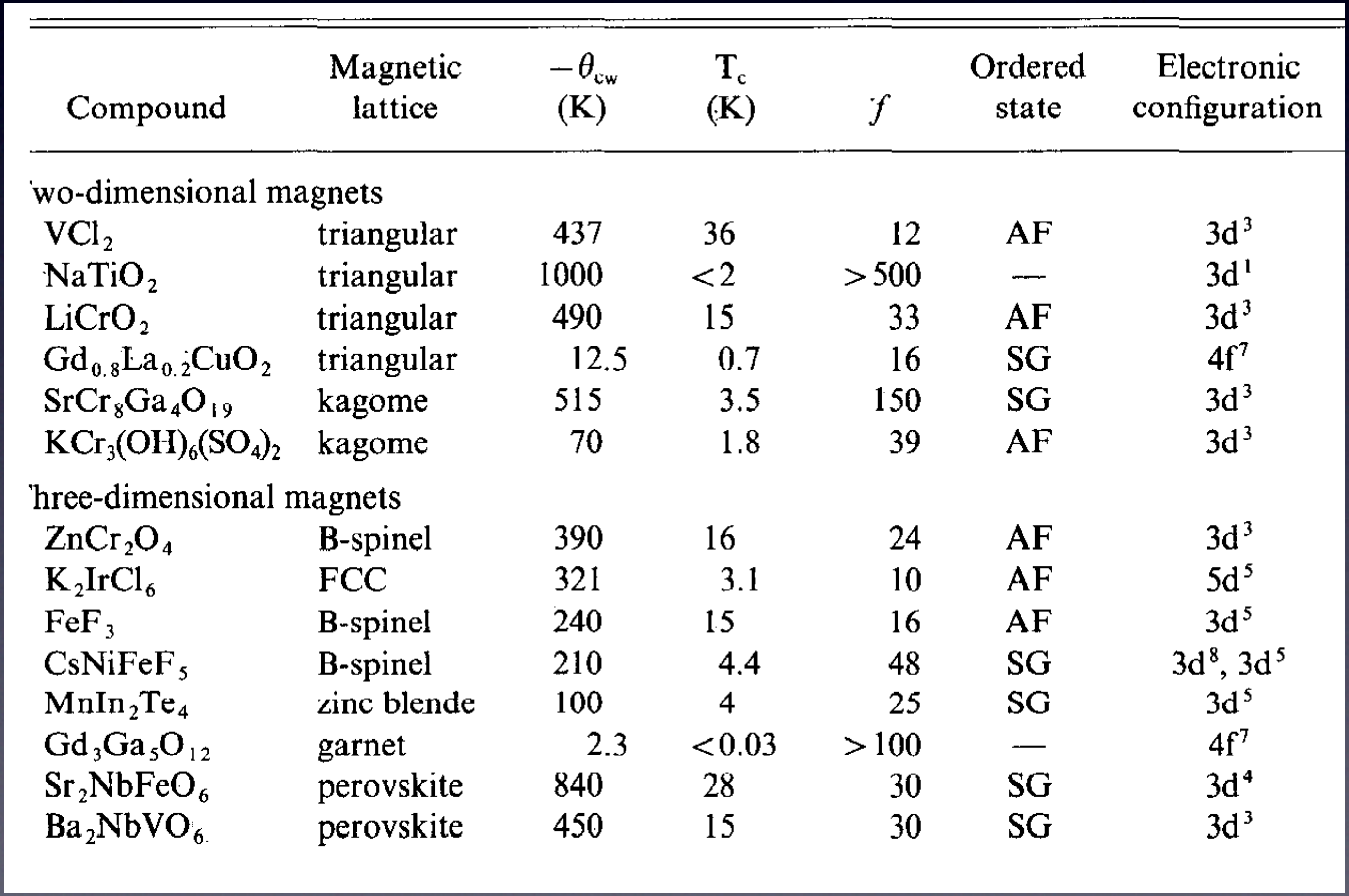
- Frustration/fluctuation parameter

$$f = \frac{|\Theta_{CW}|}{T_c}$$

>>1 indicates
suppressed ordering

Some older examples

A.P. Ramirez review, 1994



Compound	Magnetic lattice	$-\theta_{cw}$ (K)	T_c (K)	f	Ordered state	Electronic configuration
wo-dimensional magnets						
VCl_2	triangular	437	36	12	AF	$3d^3$
$NaTiO_2$	triangular	1000	<2	>500	—	$3d^1$
$LiCrO_2$	triangular	490	15	33	AF	$3d^3$
$Gd_{0.8}La_{0.2}CuO_2$	triangular	12.5	0.7	16	SG	$4f^7$
$SrCr_8Ga_4O_{19}$	kagome	515	3.5	150	SG	$3d^3$
$KCr_3(OH)_6(SO_4)_2$	kagome	70	1.8	39	AF	$3d^3$
hree-dimensional magnets						
$ZnCr_2O_4$	B-spinel	390	16	24	AF	$3d^3$
K_2IrCl_6	FCC	321	3.1	10	AF	$5d^5$
FeF_3	B-spinel	240	15	16	AF	$3d^5$
$CsNiFeF_5$	B-spinel	210	4.4	48	SG	$3d^8$, $3d^5$
$MnIn_2Te_4$	zinc blende	100	4	25	SG	$3d^5$
$Gd_3Ga_5O_{12}$	garnet	2.3	<0.03	>100	—	$4f^7$
Sr_2NbFeO_6	perovskite	840	28	30	SG	$3d^4$
Ba_2NbVO_6	perovskite	450	15	30	SG	$3d^3$

Questions

- What is the nature of the "spin liquid" regime where $T_c<T<|\Theta_{CW}|$?
  - Here spins are correlated but fluctuating
- What is the nature of the ground state, or low temperature phase if $T_c>0$?
- What are the elementary excitations of the system?

Questions

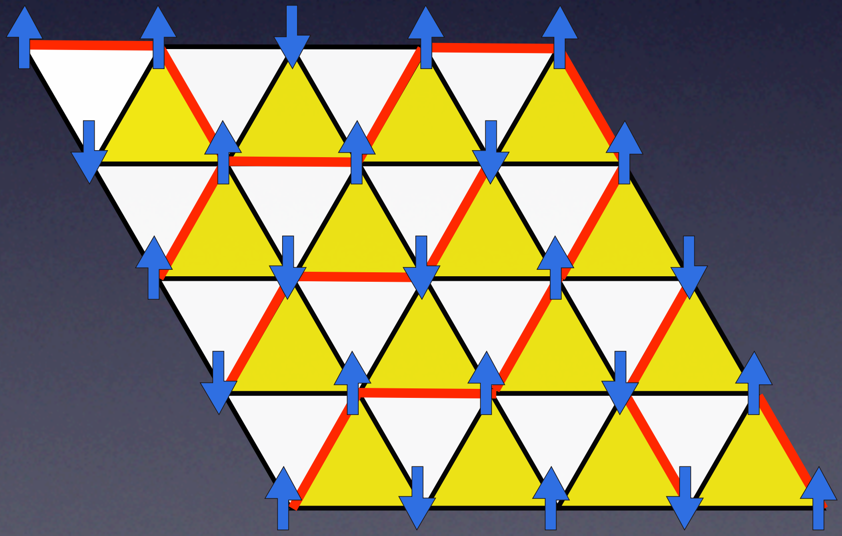
- What is the nature of the "spin liquid" regime where $T_c<T<|\Theta_{CW}|$?
  - Here spins are correlated but fluctuating
  - Do these correlations have any long-distance consequences?

For AF NN Ising Models

Lattice	Transition	Correlations ($T \ll \lvert\Theta_{CW}\rvert$)
FCC	Yes! $T_c = 1.8J$	LRO
triangular	no	power law
checkerboard	no	power law
pyrochlore	no	power law
kagome	no	very short range

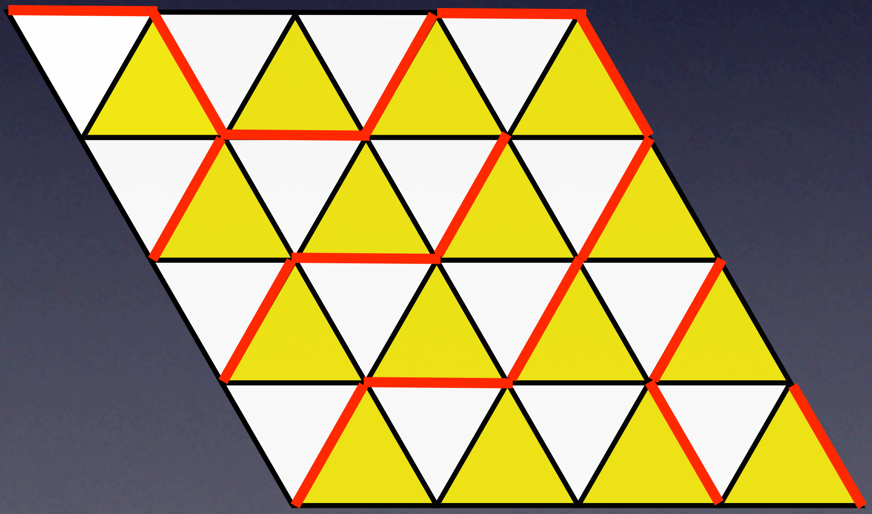
Back to the Ising model

- Correlations: we know that for T<<J, there are "no" triangles with 3 aligned spins
  - How does this induce long-distance correlations?



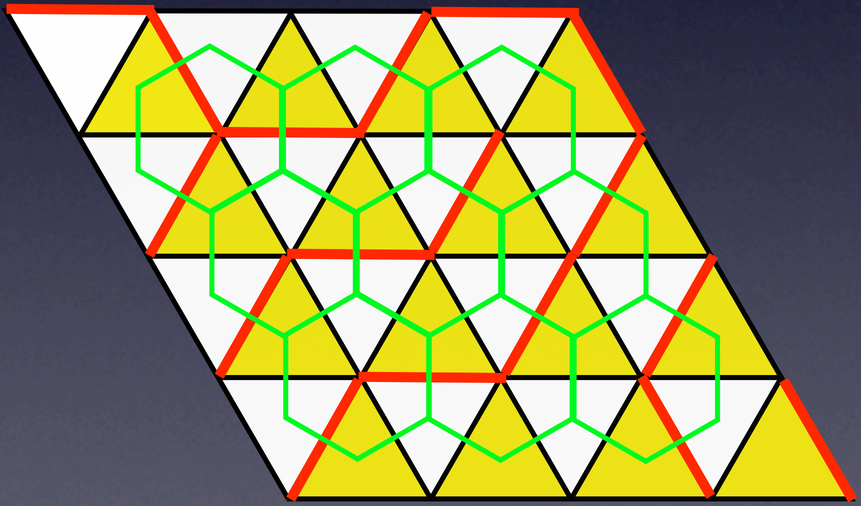
Back to the Ising model

- Dual representation
  - honeycomb lattice



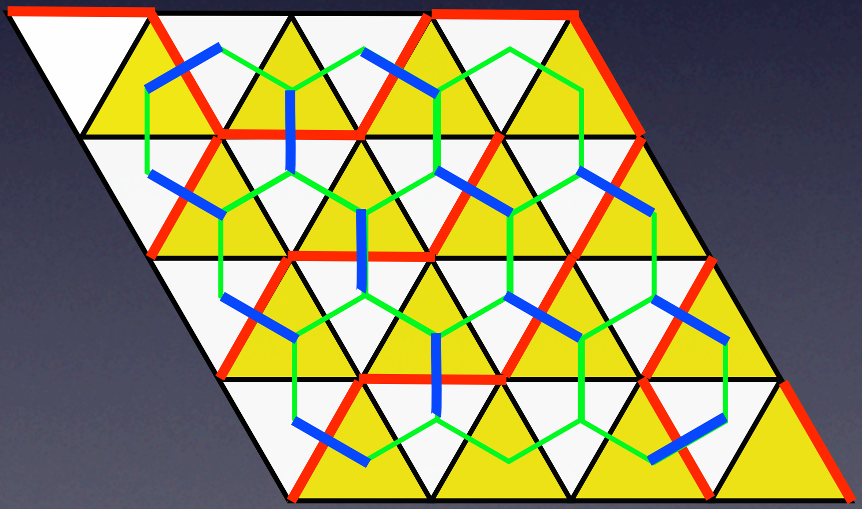
Back to the Ising model

- Dual representation
  - focus on the frustrated bonds



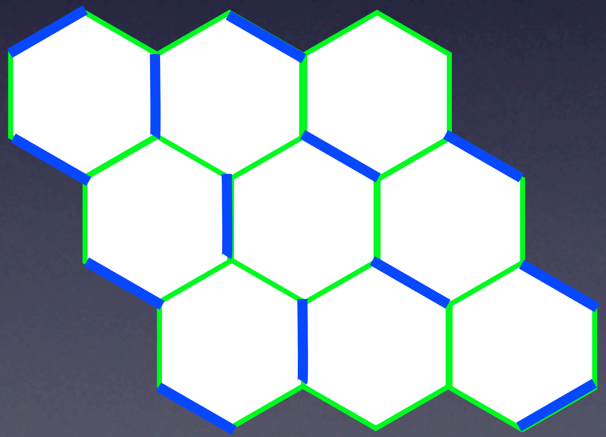
Back to the Ising model

- Dual representation
  - color "dimers" corresponding to frustrated bonds
    - "hard core" dimer covering



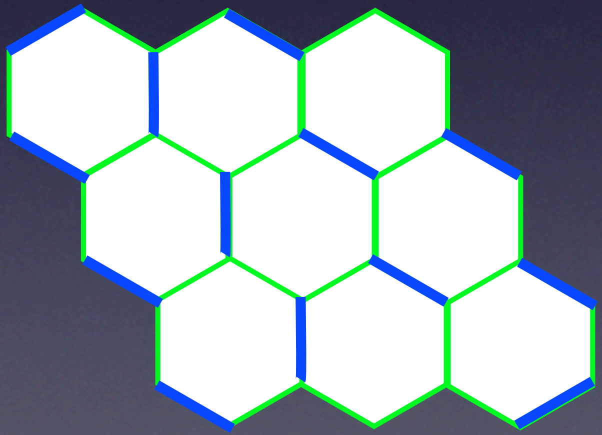
Back to the Ising model

- Dual representation
  - A 2:1 mapping from Ising ground states to dimer coverings

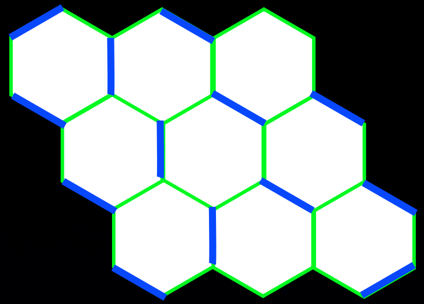


Dimer states

- First exercise: can we understand Wannier's result?
  - count the dimer coverings



Dimer states



- Consider the "Y" dual sites
  - each has 3 configurations

  - this choice fully determines the dimer covering
- But we have to make sure the Y^{-1} sites are singly covered. Make a crude approximation:
  - Prob(dimer) = 1- Prob(no dimer)= 1/3
  - Prob(good Y^{-1}) = 2/3 * 2/3 * 1/3 * 3 = 4/9
- Hence

$$\Omega \approx 3^N \left(\frac{4}{9}\right)^N = e^{N\ln(4/3)}$$

$S \approx 0.29\ N\ k_B$

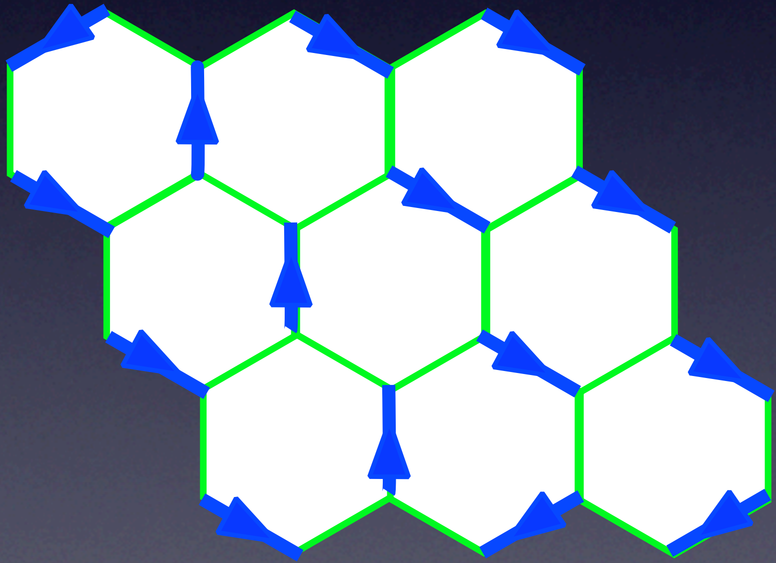
Wannier $S \approx 0.34 N k_B$

“Magnetostatics”

- Define a dimer number n_{ij}=0,1 on bond (ij)
- Turn this into a lattice “magnetic field” B_{ij}

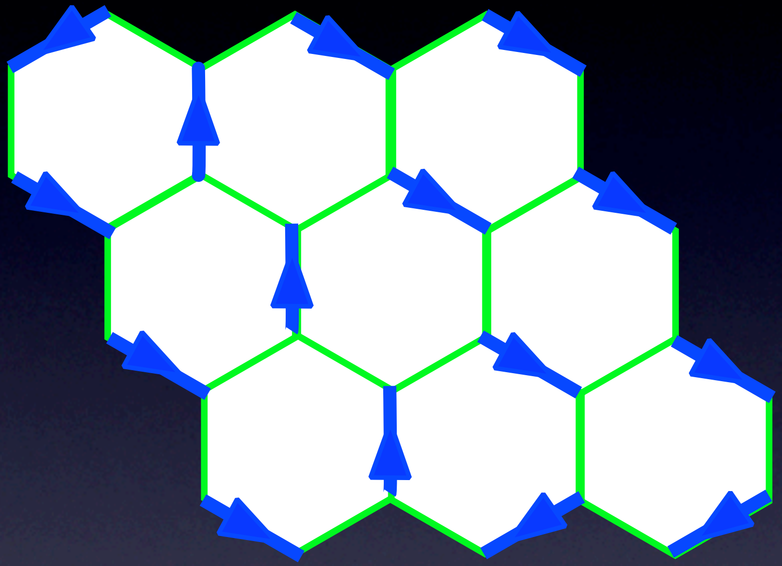
$$B_{ij} = \begin{cases} n_{ij} & i \in Y \\ -n_{ij} & i \in Y^{-1} \end{cases}$$

$$(\mathrm{div}\ B)_i = \sum_j B_{ij} = \varepsilon_i = \pm 1$$



Some magnetostatic representation exists for all the cases with power-law correlations!

“Magnetostatics”



$(\mathrm{div}\ B)_i = \sum_j B_{ij} = \varepsilon_i = \pm 1$

- Focus on fluctuations

$$B_{ij} = \overline{B}_{ij} + b_{ij}$$

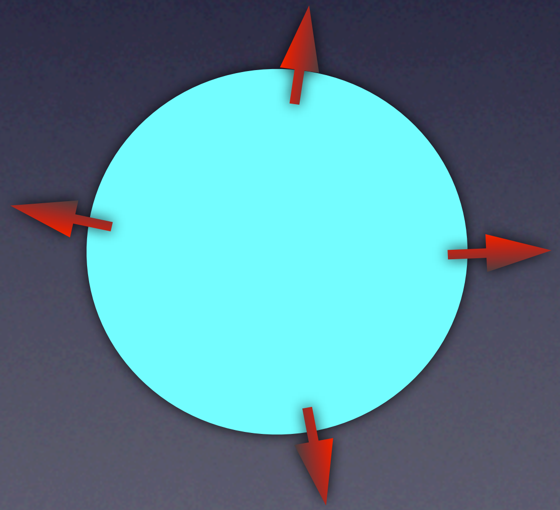
$$\mathrm{div}(\overline{B}_{ij}) = \varepsilon_i$$

$$\mathrm{div}(b_{ij}) = 0$$

- Fluctuating component b_{ij} is divergenceless

“Magnetostatics”

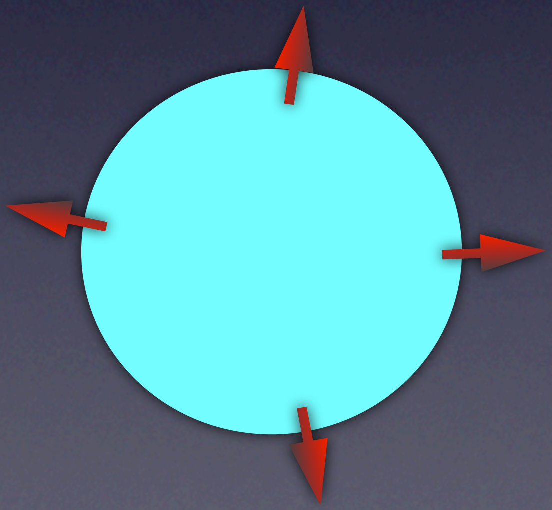
- Divergenceless condition, div b = 0, implies long-distance correlations in the fluctuations by Gauss’ law
- no monopole fluctuations



“Magnetostatics”

- Divergenceless condition, div b = 0, implies long-distance correlations in the fluctuations by Gauss’ law
- no monopole fluctuations

$$\frac{\partial}{\partial x_i}\langle b_i(x) b_j(x')\rangle = 0$$



Long distances

- For long distance correlations, we can consider a coarse-grained $b_i(x)$ field
  - Either there are no significant b fluctuations, in which case some specific ordered state is picked out
  - - or - the fluctuations are large, and hence coarse-grained b field can be regarded as a continuous variable
- The latter is true in many cases

Effective theory

- Effective free energy

$$\beta F = \int d^2x \, \frac{c}{2} |\vec{b}(x)|^2 + \mathrm{h.o.t.s}$$

- Solve divergence constraint $b_\mu = \epsilon_{\mu\nu}\partial_\nu \phi$

$$\beta F = \int d^2x \, \frac{c}{2} |\nabla \phi|^2$$

Effective theory

- Solve divergence constraint $b_\mu = \epsilon_{\mu\nu}\partial_\nu \phi$

$$\beta F = \int d^2x \, \frac{c}{2} |\nabla \phi|^2$$

- Gaussian correlation

$$\langle b_\mu(r) b_\nu(r') \rangle \sim \epsilon_{\mu\lambda}\epsilon_{\nu\gamma} \frac{\partial}{\partial x_\lambda} \frac{\partial}{\partial x'_\gamma} \left(c^{-1} \ln |r - r'| \right)$$

- 2d power-law “dipolar” form

$$\langle b_\mu(r) b_\nu(0) \rangle \sim -c^{-1} \epsilon_{\mu\lambda}\epsilon_{\nu\gamma} \frac{\hat{r}_\lambda \hat{r}_\gamma}{r^2}$$

?

copied from Lori H. Barrett Fine Art

?

- The degeneracy is probably removed by any weak perturbation
  - power-laws are not "universal" the way that those at critical points are
- It is hard to get any simple NN Ising system without substantial corrections

Magnetic Anisotropy

- Microscopically, SU(2) symmetry is broken by spin-orbit coupling $\lambda L \cdot S$
  - need to consider atomic physics
- Several effects
  - $H = H_0 + H_{e\text{-}e} + H_{\text{crystal field}} + H_{SOI}$
  - Relative magnitudes different for transition metals (d) and rare earth (f) ions

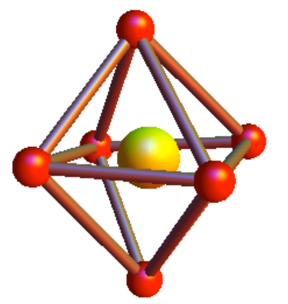
Transition Metals

- Typically, $H_{e\text{-}e}$ (responsible for Hund's rules I and II) and H_{cf} are up to of order a few eV in magnitude
- Spin-orbit is a relatively small
  - Varies as Z^4, from of order tens of meV for 3d TMs to of order 0.5-1eV in late 5d TMs
  - Except for heaviest 5d ions, it is a weak perturbation compared to crystal fields

Transition Metals

- Crystal fields split orbital degeneracy

e.g. cubic symmetry



e_g: x^2-y^2, $2z^2$-x^2-y^2

t_{2g}: yz, xz, xy

- There is always *at least* this much splitting
- The crystal field splitting reduces orbital degeneracy
    - When this results in a half-filled shell, effects of SOIs are second order ~ λ^2/Δ_{cf}

Ising TMs?

- To get an Ising spin, you need a low symmetry environment (with a singled out axis)
  - in this case, all the orbital degeneracy is usually split
  - must have a situation with some “accidental” degeneracy to allow SOIs to work, or else weak exchange $J << \lambda^2/\Delta_{cf}$
- This happens, e.g. in Co^{2+},Co^{3+} ions which show “spin state transitions”

Rare Earths

- In Ln (4f) rare earths, electrons are relatively close to the nucleus and screened from crystal fields, so typically SOIs are *larger* than crystal fields
- Since SOIs just result in a partial splitting of L+S degeneracies to a J degeneracy, the crystal fields then select anisotropic states at $O(\Delta_{cf})$
- Also, exchange interactions are weak for rare earths (typically only a few K)

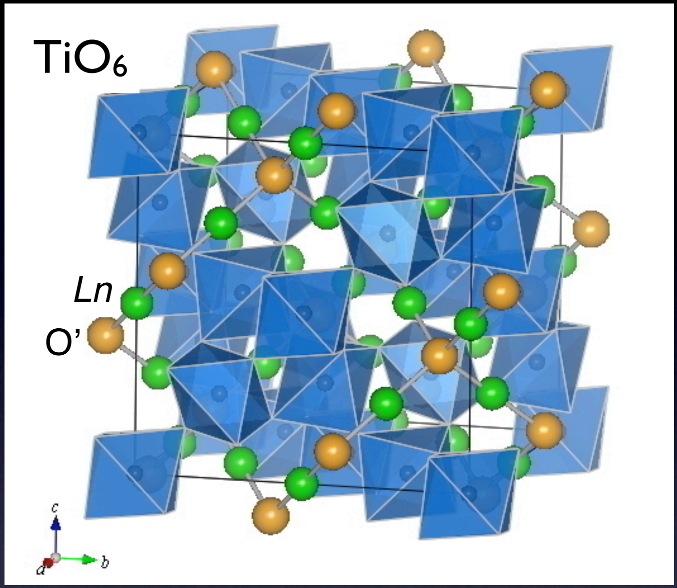
Rare Earths

- Rare earth Lns tend to exhibit anisotropic magnetism
- But...
  - dipolar forces can be comparable to exchange
  - anisotropy does not need to be so simple as an Ising model
    - usually with respect to some local axes, which can be different for different spins

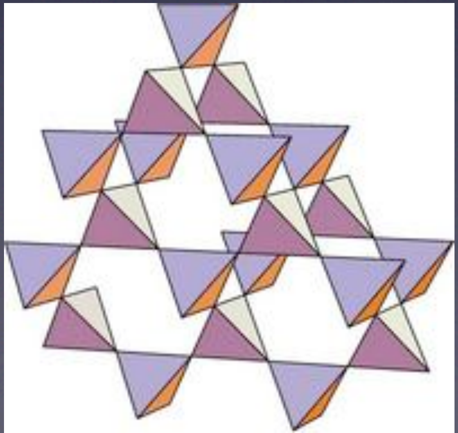
Spin Ice

Materials

- Rare earth pyrochlores
  - $Ln_2Ti_2O_7$, Ln=Dy,Ho



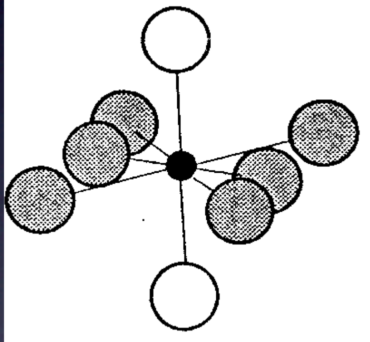
- The Ln's occupy a magnetic pyrochlore lattice
  - Strong easy-axis anisotropy oriented along <111> axes connecting tetrahedra centers



Anisotropy

- Crystal fields create a potential that depends on the spin state

$$H_{cf} \approx -D \sum_i \left(\mathbf{S}_i \cdot \hat{\mathbf{n}}_i\right)^2$$



- This leads to two Ising ground states with $S_i^z = \pm S$

Magnetic moment

- Basically the Ising anisotropy means that there are two ground states forming a doublet, such that we can define a S=1/2 “spin” from it, and associated Pauli matrices, *such that*
- The magnetic moment is nearly uniaxial

$$\vec{\mu}_i = m_0 \hat{n}_i \boxed{(\hat{n}_i \cdot \vec{\sigma})} \longrightarrow \sigma$$

- Here m_0 is a large intrinsic magnetic moment of the Ln spin, $m_0 \approx 10\mu_B$

Dipolar Interactions

- Because m₀ is so large, the dipolar interactions are relatively strong

$$H_{dip} = \sum_{i>j} \left[\frac{\vec{\mu}_i \cdot \vec{\mu}_j - 3(\vec{\mu}_i \cdot \hat{r}_{ij})(\vec{\mu}_j \cdot \hat{r}_{ij})}{|r_{ij}|^3} \right]$$

- Note that only σ enters this interaction!
  - Hence it is effectively classical
  - It is also "ferromagnetic" in the sense that the 2nd term is larger than the first

NN model

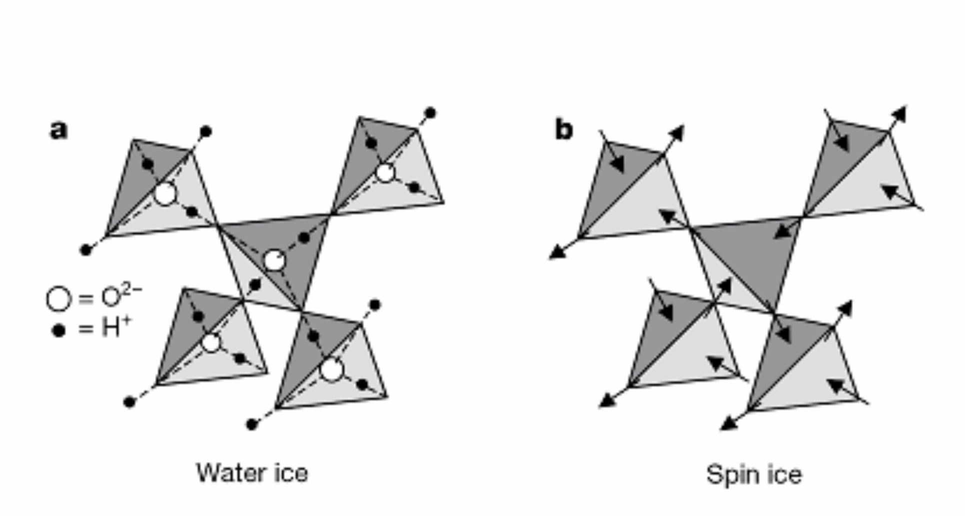
- Taking just the NN term of the dipolar interaction, one obtains an effective model

$$H_{eff} = J_{eff} \sum_{\langle ij \rangle} \sigma_i \sigma_j$$

  - with $J_{eff}>0$, i.e. like an AF Ising model.
- It is believed that this is partially compensated by some weaker exchange (not dipolar) of the opposite sign, but the net J_{eff} remains positive.

Spin ice ground states

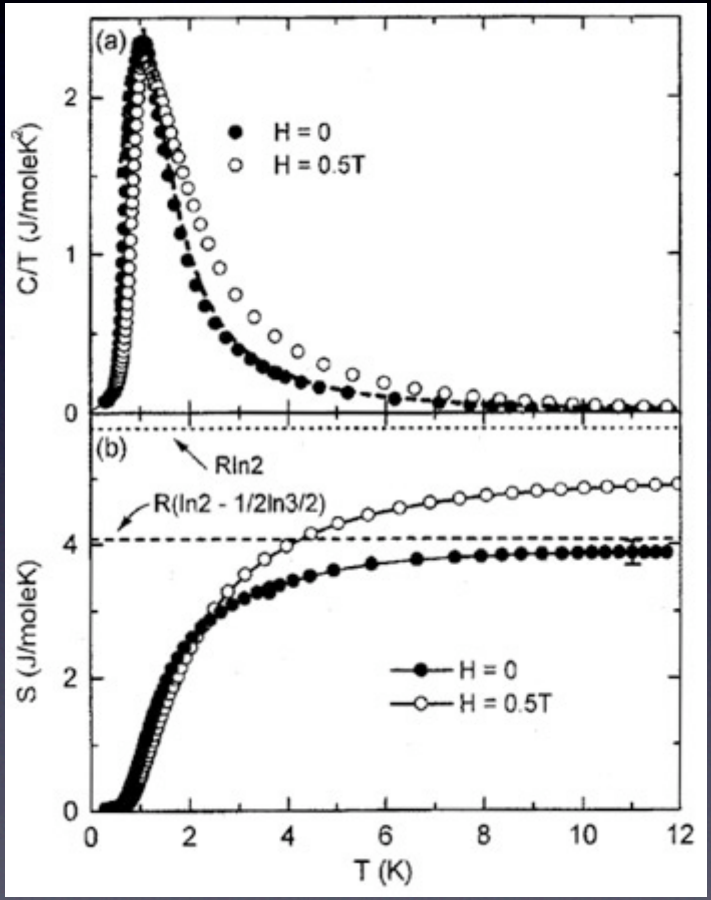
- The NN energy is minimized by making the σ_i add to 0 on each tetrahedron, so the spins point "two in/two out": the "ice rule"
- This is the origin of the name "spin ice"



M.J. Harris *et al*, 1997

Entropy

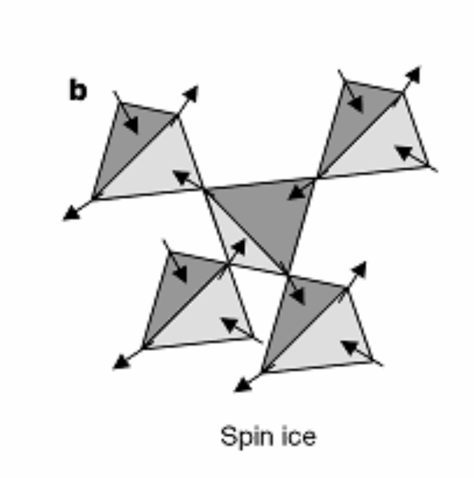
- The integrated specific heat of $Dy_2Ti_2O_7$ showed explicitly the low temperature entropy in spin ice as a "missing" part of R ln(2)
  - quantitative agreement with Pauling's 1935 estimate

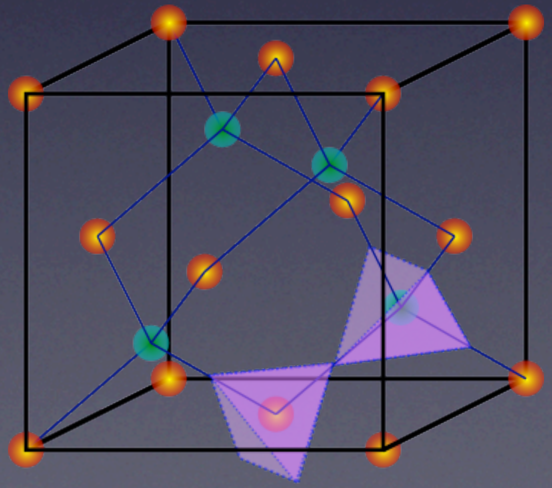


A.P. Ramirez *et al*, 1999

“Magnetostatics”

- It is clear from the picture that we can directly define a divergenceless “magnetic field” b_{ij} from the direction of the spin connecting the centers of tetrahedra i and j, which reside on a diamond lattice





Power law correlations

- Effective theory

$$H_{eff} = \int d^3 r \, \frac{c}{2} |\vec{b}|^2$$

- Using vector potential b = ∇×a

$$\langle b_\mu(r) b_\nu(0) \rangle \sim 1/c \left(\frac{\delta_{\mu\nu} - 3\hat{r}_\mu \hat{r}_\nu}{r^3} \right)$$

Power law correlations

- Effective theory

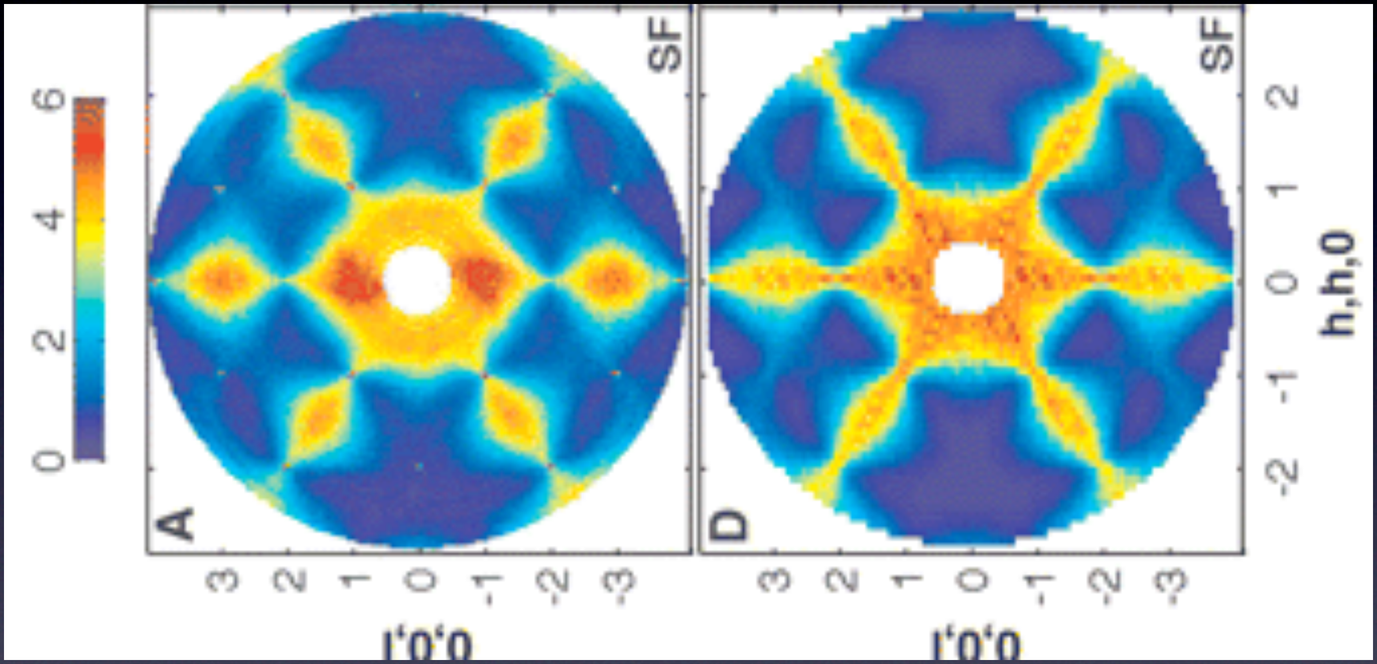
$$H_{eff} = \int d^3 r \, \frac{c}{2} |\vec{b}|^2$$

- Using vector potential b = ∇×a

$$\langle b_\mu(-k) b_\nu(k) \rangle = 1/c \left(\delta_{\mu\nu} - \frac{k_\mu k_\nu}{k^2} \right)$$

- This is directly proportional to the static magnetic structure factor measured in a neutron experiment
  - e.g. $S(\mathbf{K}_{200} + \mathbf{k}) \sim \dfrac{k_y^2 + k_z^2}{k^2}$

pinch points in $Ho_2Ti_2O_7$



experiment

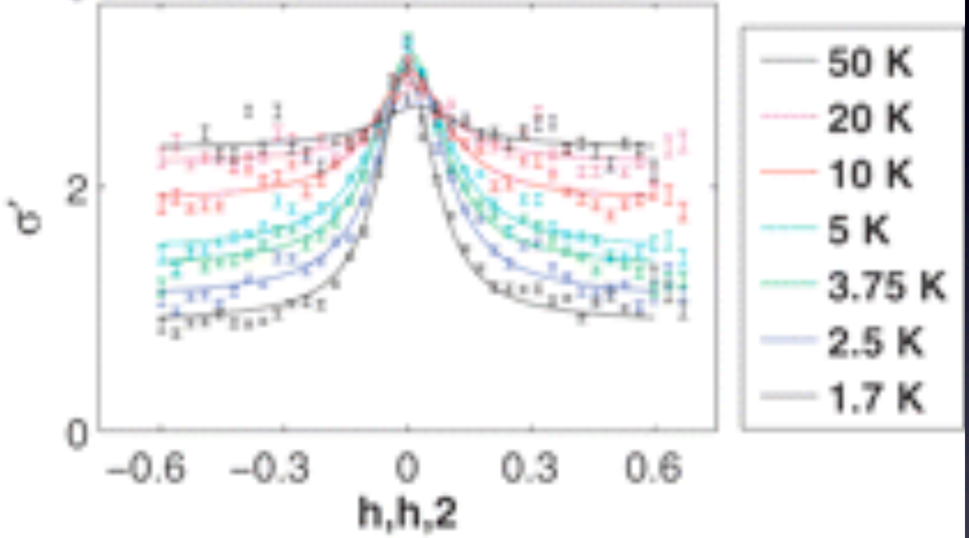
theory

T. Fennell *et al*, 2009

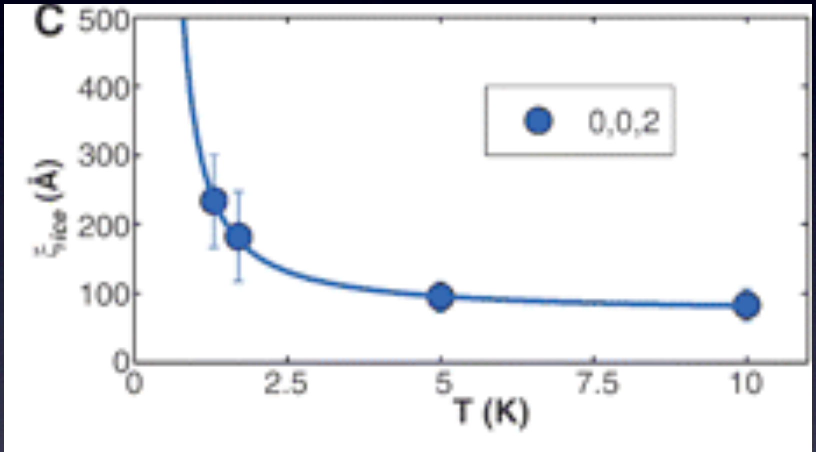
$$S(\mathbf{K}_{200} + \mathbf{k}) \sim \frac{k_y^2 + k_z^2}{k^2}$$

vanishes along lines

Quality of singularity



pinch point sharpens
with lower T

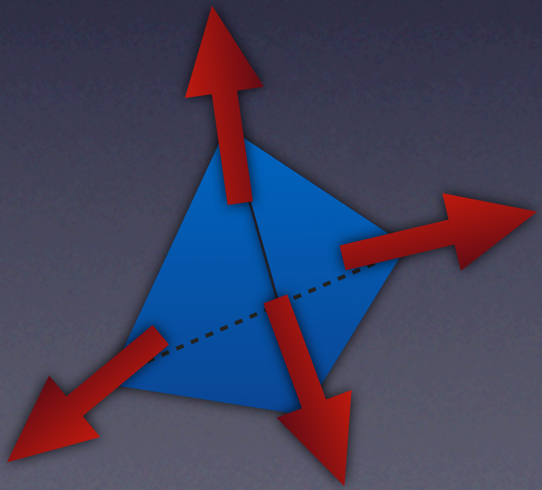


“Correlation length” for
rounding of pinch point

Roughly $\xi \sim e^{1.8K/T}$

Defects

- The ice rules constraint is not perfectly enforced at T>0
- Primitive defect is a "charged" tetrahedron with $\sum_i \sigma_i = \pm 1$.



costs energy $2J_{eff}$

What to call it?

- Consider Ising "spin"

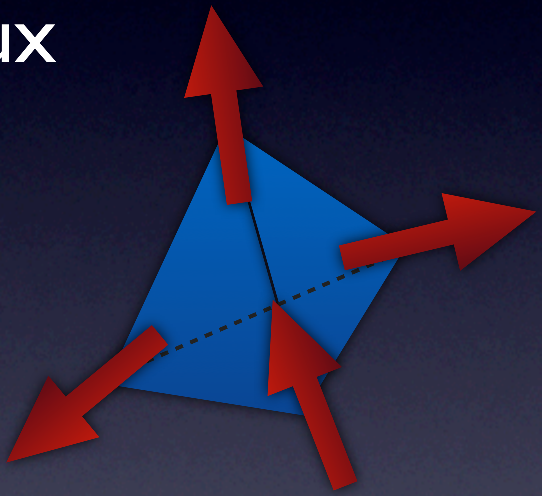
$$S^z_{\mathrm{TOT}} = \sum_i \sigma_i = \frac{1}{2}\sum_t S^z_t$$

- Single flipped tetrahedron has $S^z_{\mathrm{TOT}}=\pm 1/2$
  - "spinon"? (M. Hermele *et al*, 2004)
  - But S^z is not very meaningful in spin ice
- Use magnetic analogy: *magnetic monopole*

Magnetic monopoles

Castelnovo *et al*, 2008

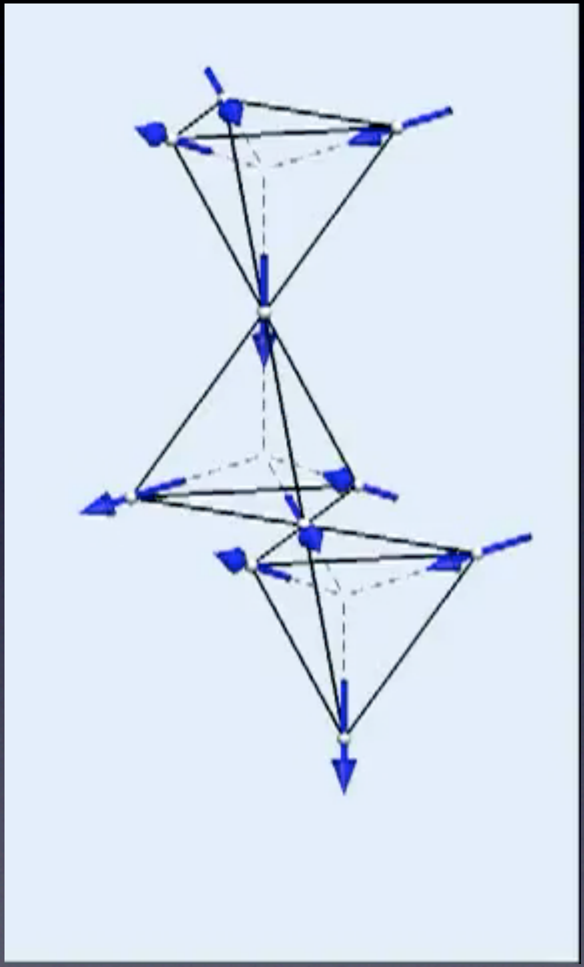
- Defect tetrahedra are sources and sinks of “magnetic” flux



div b = 1

- It is a somewhat non-local object
  - Must flip a semi-infinite string of spins to create a single monopole

String

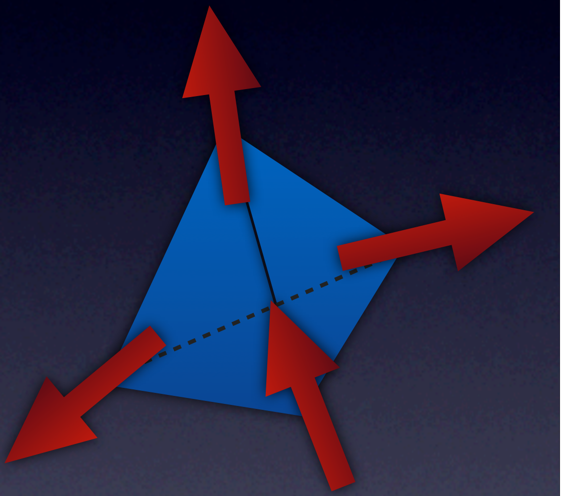


stolen (by somebody else on youtube)
from Steve Bramwell

- Note that the string is tensionless because the energy depends only on $\sum_i \sigma_i$ on each tetrahedra
  - this should be spoiled at low temperature by corrections to H
- Once created, the monopole can move by single spin flips

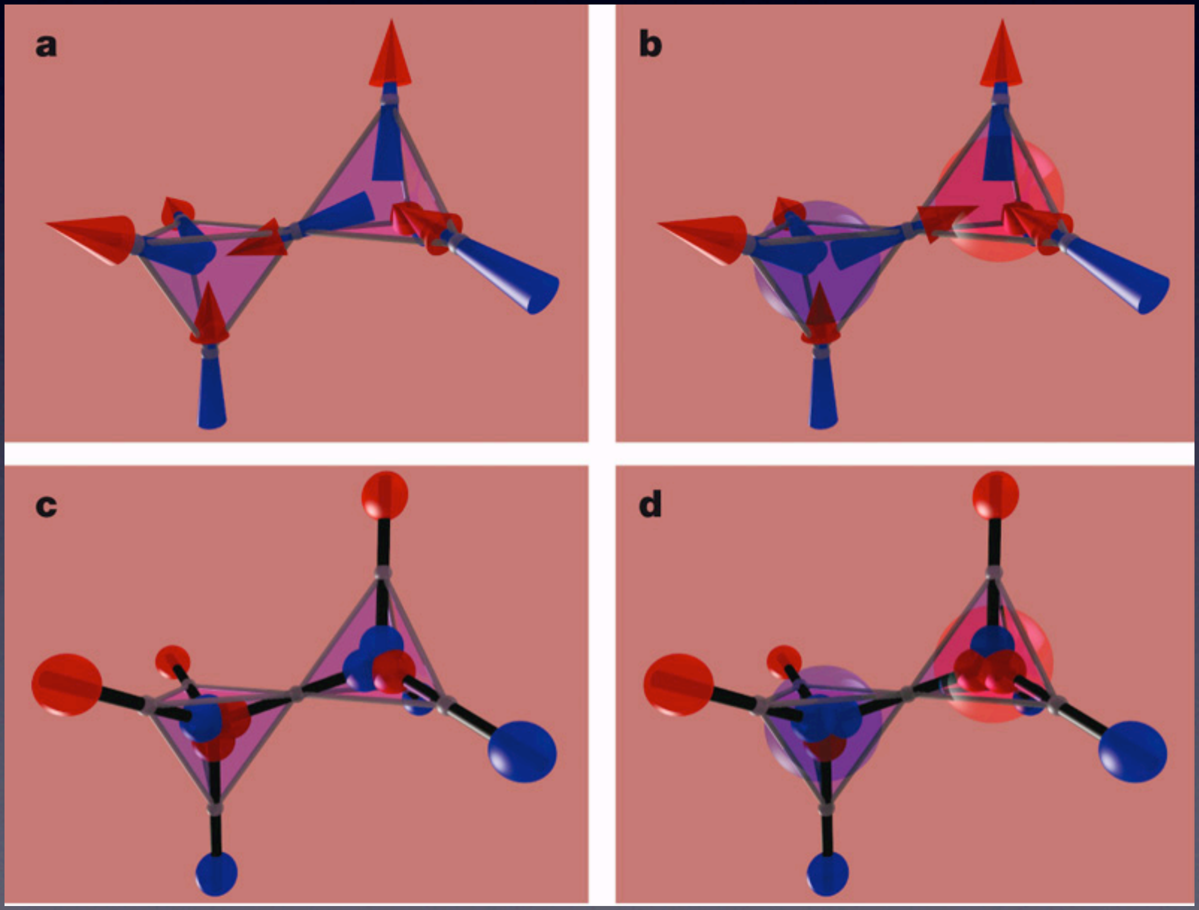
Monopoles are "real"

Castelnovo *et al*, 2008



- Monopoles actually are sources for (internal) magnetic field
    - Magnetization $M \propto b$
    - hence div M ~ div H ~ q $\delta(r)$
- Actual magnetic charge is small
    - Coulomb interaction constant is approximately 14000 times smaller than for electrons, but still $1/r^2$ forces are present and measurable at low temperature

Monopoles for dumbbells



Experimental evidence for monopoles

- Careful study of quasi-activation behavior of magnetization relaxation rate (Jaubert +Holdsworth, 2009)
  - measures the energy of a monopole
- Magnetic "Wien" effect (Bramwell et al, 2009)
  - measures a monopole's magnetic charge
- Several neutron measurements see "strings" in applied fields
- Hopefully Peter Holdsworth will discuss all these!

Kivelson's argument

"Dear Leon,

I now have evidence direct from Hollywood that you were wrong in assessing the relative importance of topological insulators vs spin ice..."

More on Ising models?

- Quantum dynamics can be introduced by transverse exchange or field

$$H = \frac{1}{2} \sum_{ij} J_{ij} \left[\sigma_i^z \sigma_j^z + \alpha (\sigma_i^x \sigma_j^x + \sigma_i^y \sigma_j^y) \right] - h \sum_i \sigma_i^x$$

  - transverse field: rather hard to find in experiment, but see talk by Ribhu Kaul
  - XY exchange: more easily realized
    - with lattice bosons (e.g. cold atoms in optical lattice)
    - Heisenberg systems in strong magnetic fields often have collinear states for which one can use such an expansion (example later?)

2d Results

- In 2d, these problems have been heavily studied
  - In a transverse field by Moessner *et al*
  - With XY exchange more recently by several groups
- Generally, the result is that Ising order develops with an infinitesimal quantum perturbation whenever the classical system has power-law correlations
  - This is related to a classic result in QFT by Polyakov that a compact U(1) gauge theory is confining in 2d due to proliferation of instantons (monopoles*)

*these are *not* anything like the spin ice monopoles

3d Results

- By contrast, in the 3d pyrochlore lattice, quantum perturbations lead to the emergence of a true quantum spin liquid state

M. Hermele *et al*, 2004
A. Banerjee *et al*, 2008

- This you can think of as analogous to the Coulomb phase of spin ice but with *quantum dynamics* added instead of just magnetostatics
- This is all possible because compact U(1) gauge theory is stable in 3 dimensions