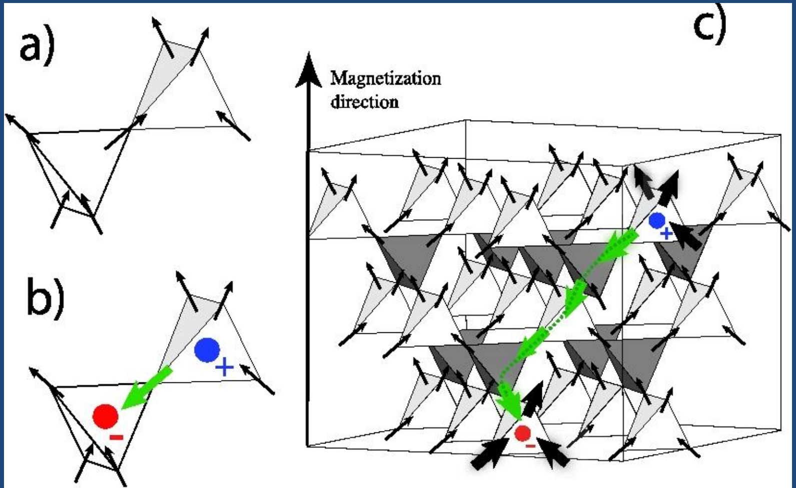
$Yb_2Ti_2O_7$: A model Quantum Spin Ice



RRPS, R. Applegate, N. R. Hayre UC Davis

M. Gingras, T. Lin, A. G. R. Day U. Waterloo

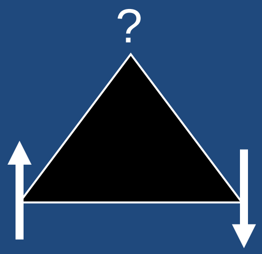
K. Ross, B. Gaulin McMaster

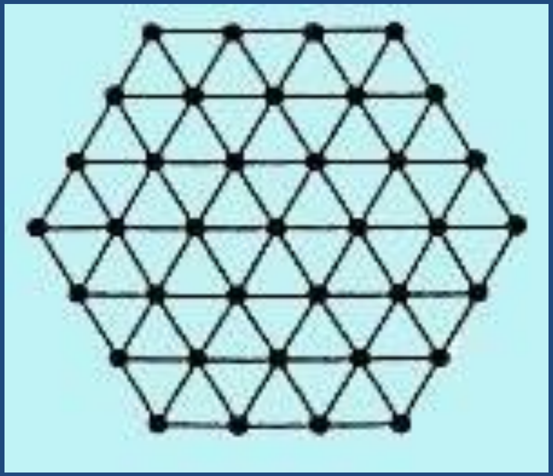
R. Applegate et al PRL 109, 097205 (2012)
N. R. Hayre et al Cond-mat arxiv:1211.5934 (in PRB)

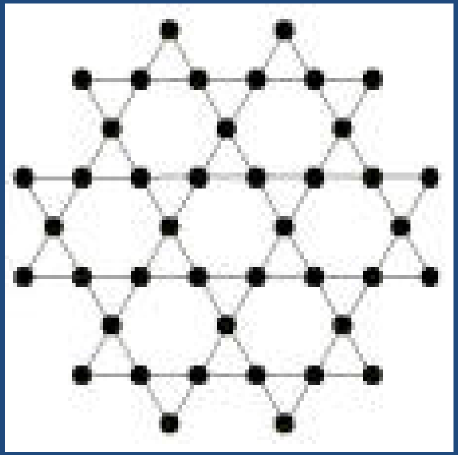
OUTLINE

- Introduction: Frustrated Quantum Spin Systems
- Ice rules, Spin Ice and Quantum Spin Ice
- $Yb_2Ti_2O_7$: A model Quantum Spin Ice
- Is it a Quantum Spin Liquid?
- Summary and Future Directions

Frustration leads to many degenerate ground states In classical spin systems







Triangle of AFM Ising spins: 6 out of 8 states are ground states

uud udu duu udd dud ddu

TLIM: T=0 critical point (Wannier)

Ground state entropy under 50% of total entropy

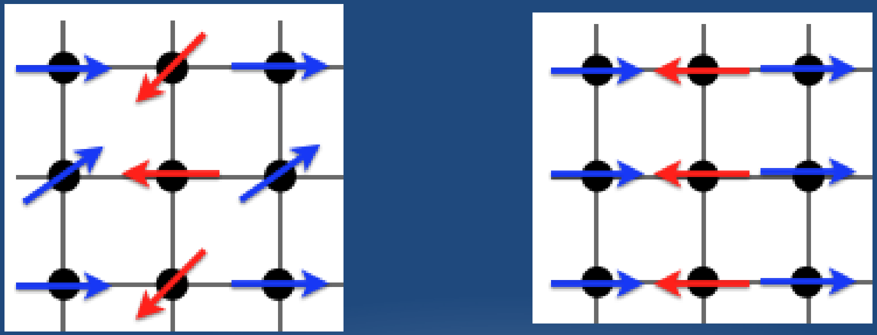
KLIM: Finite (short) correlation length even at T=0

Ground state entropy about 72% of total entropy

Quantum Fluctuations in a degenerate system can lead to

Selection of Classical Order (Order by Disorder)

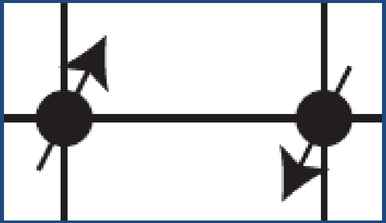
Lifting of accidental degeneracy due to fluctuations or disorder

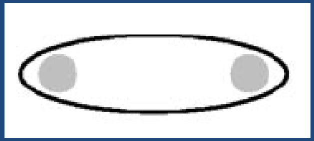


Selection of Collinear Order in J1-J2 Heisenberg Model at large J2
Relevant to the Iron Pnictide family
Selection of order in XY spin-ice Er2Ti2O7 (Savary et al, Zhitomirsky et al)

Quantum Fluctuations in a degenerate system can lead to

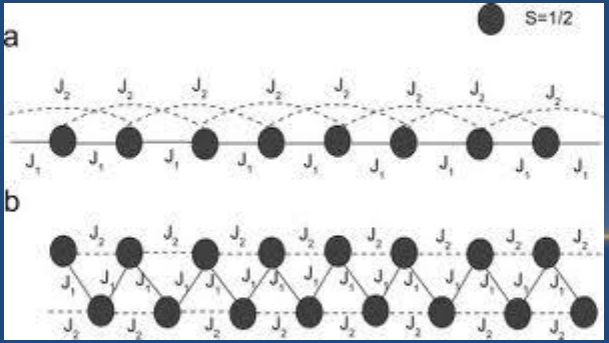
Novel Order Parameters (e.g. Valence Bond Order)

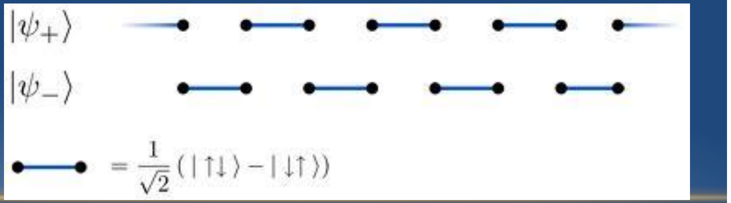


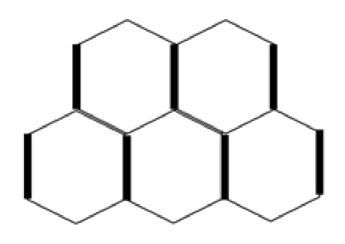


Quantum superposition allows formation of Valence Bond Singlet pairs

Valence Bond Order on a lattice breaks translational symmetry
Majumdar-Ghosh Model 1D J1-J2 model
Sandvik's J-Q Model Best studied 2D Model with VBS order





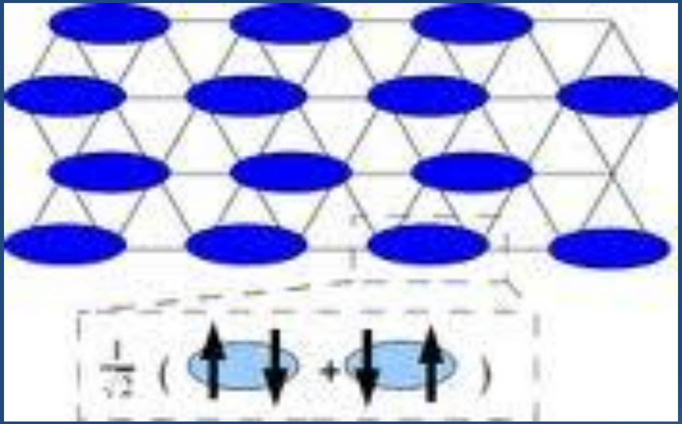


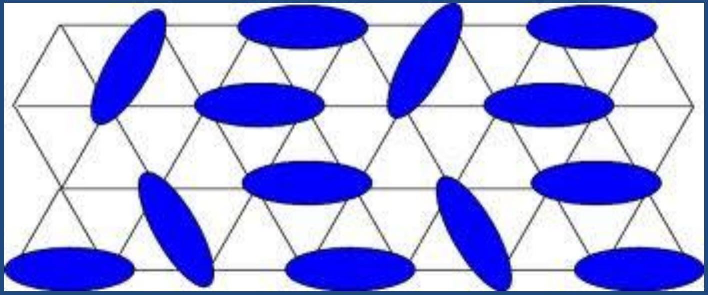
Quantum Fluctuations in a degenerate system can lead to

A highly resonating quantum superposition --- A Quantum Spin Liquid

With Exotic Emergent Properties (RVB): P. W. Anderson

Fractional Excitations, Topological Order





Absence of Broken Symmetries, Delocalized spinons

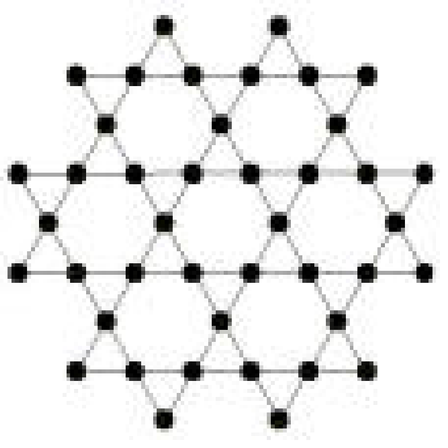
Essential Ingredients For realistic QSL (in d>1)?

Maximal Frustration?
Low dimensionality?
Low spin?
Near Metal-Insulator Transition?

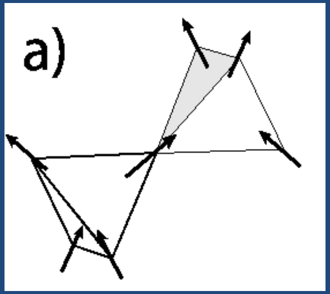
Honeycomb Lattice Hubbard Model (Intermediate U? Assaad)
Triangular Lattice Hubbard Model (Intermediate U? Motrunich)
(Spinon Fermi Surface?)

Triangular Lattice Heisenberg Model (LRO)

Kagome Lattice Heisenberg Model (Z2 QSL)
(Yan, Huse, White)



What about Quantum Spin Liquids in d=3?
Frustration not dimensionality is key for d>1

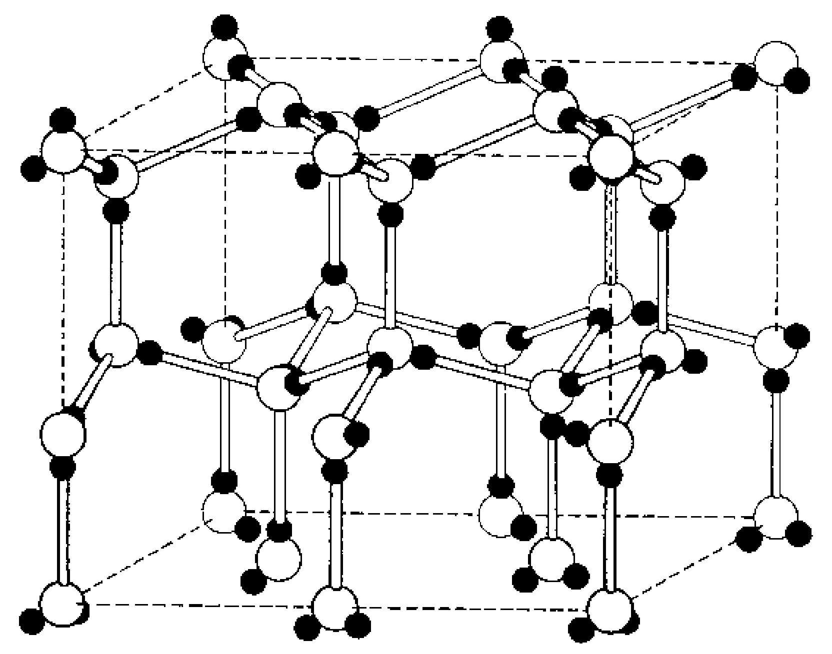


Can have fully frustrated systems in 3D
(corner sharing tetrahedra)

Large J systems can be effectively S=1/2 with strong anisotropy

Certain phases are believed to be unstable in 2D not in 3D
U(1) QSL

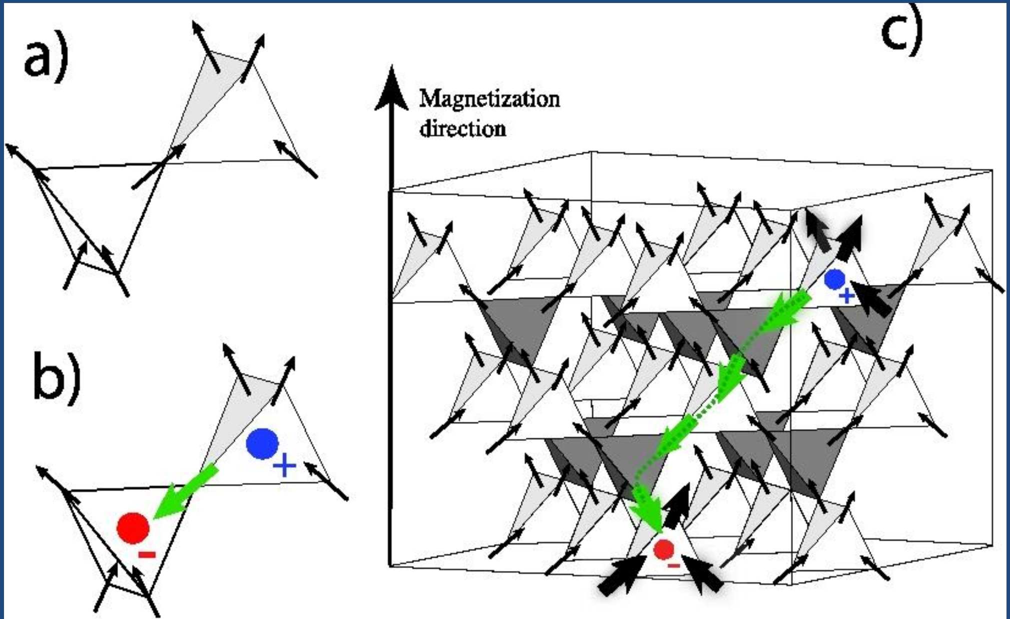
ICE RULES AND RESIDUAL ENTROPY

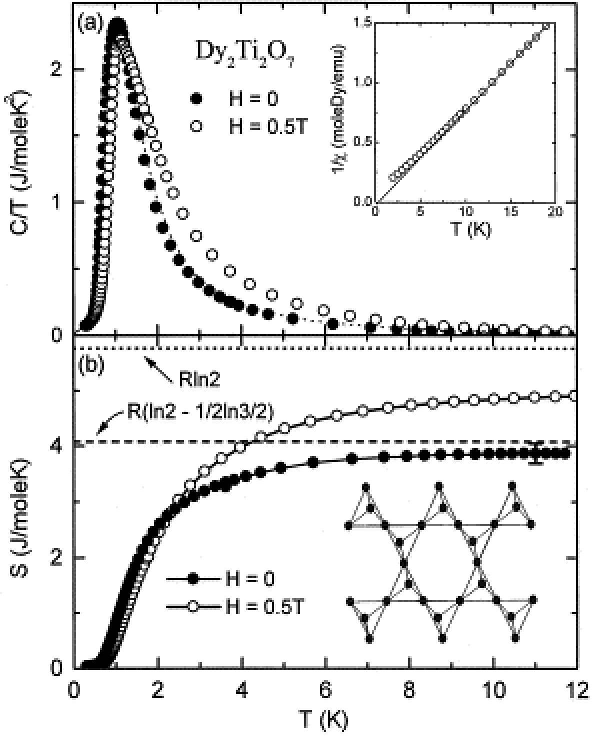


Bernal-Fowler rules for proton configurations in Ice

Leads to residual entropy   Pauling

ICE RULES AND SPIN ICE



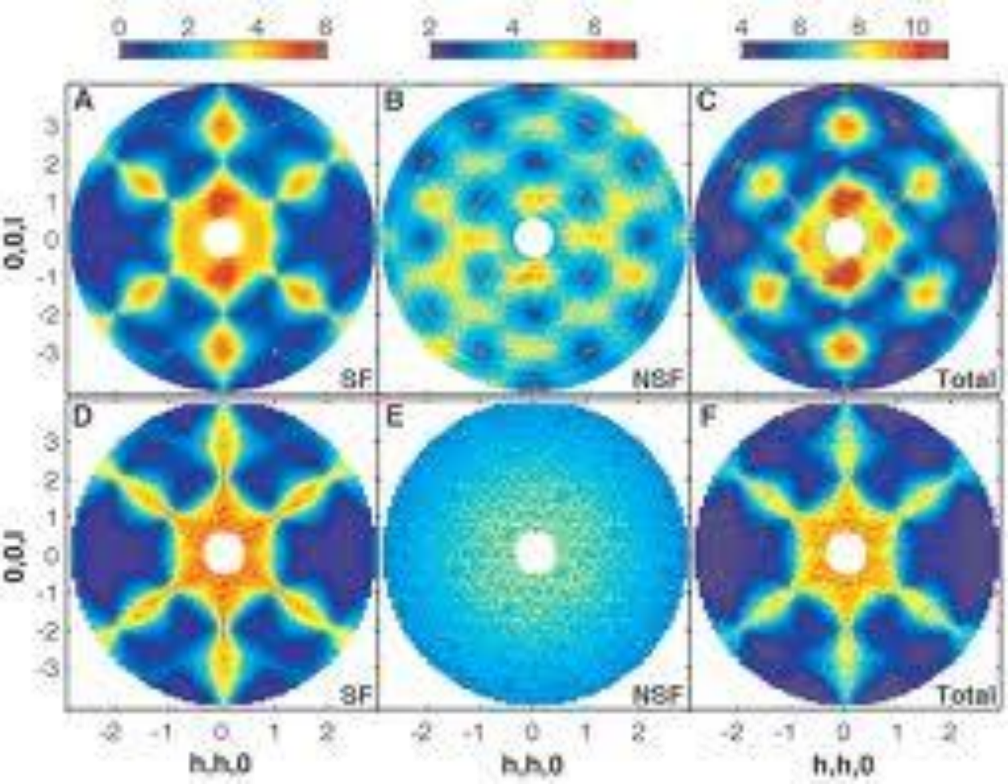


Spins (on Pyrochlore lattice) are analogs of ice with the same residual entropy (Anderson)

Ramirez et al Dy2Ti2O7 Nature 1999

Classical Spin Liquid with Monopoles/Spinons
Classical Gauge Fields
Castelnovo et al

POWER-LAW DIPOLAR CORRELATIONS LEAD TO PINCH POINTS IN NEUTRON SCATTERING



Fennel et al Science Ho2Ti2O7

ADDING QUANTUM FLUCTUATIONS TO SPIN ICE
QUANTUM SPIN ICE

Quantum Spin Liquid? (Hermele, Fisher, Balents)
(Weak quantum fluctuations on top of a highly degenerate subspace)

RVB phase is an emergent Quantum Electrodynamics
With 2 sets of conjugate vector gauge fields (E, B)

Out of an ensemble of spins can EMERGE – a novel phase with Charges, Monopoles & Photons– a full fledged fictitious Quantum Electrodynamics

Degenerate Perturbation Theory in the Spin Ice subspace

Perturbation Theory: Selection at small λ

$$\mathcal{H} = \mathcal{H}_0 + \lambda \mathcal{H}_1,$$

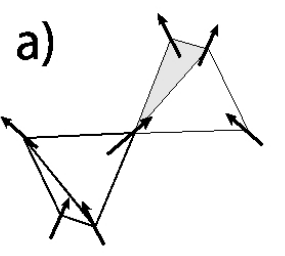
with

$$\mathcal{H}_0 = \sum_{<i,j>} S_i^z S_j^z,$$

and

$$\mathcal{H}_1 = \sum_{<i,j>} (S_i^+ S_j^- + S_i^- S_j^+)$$

No selection in first two orders



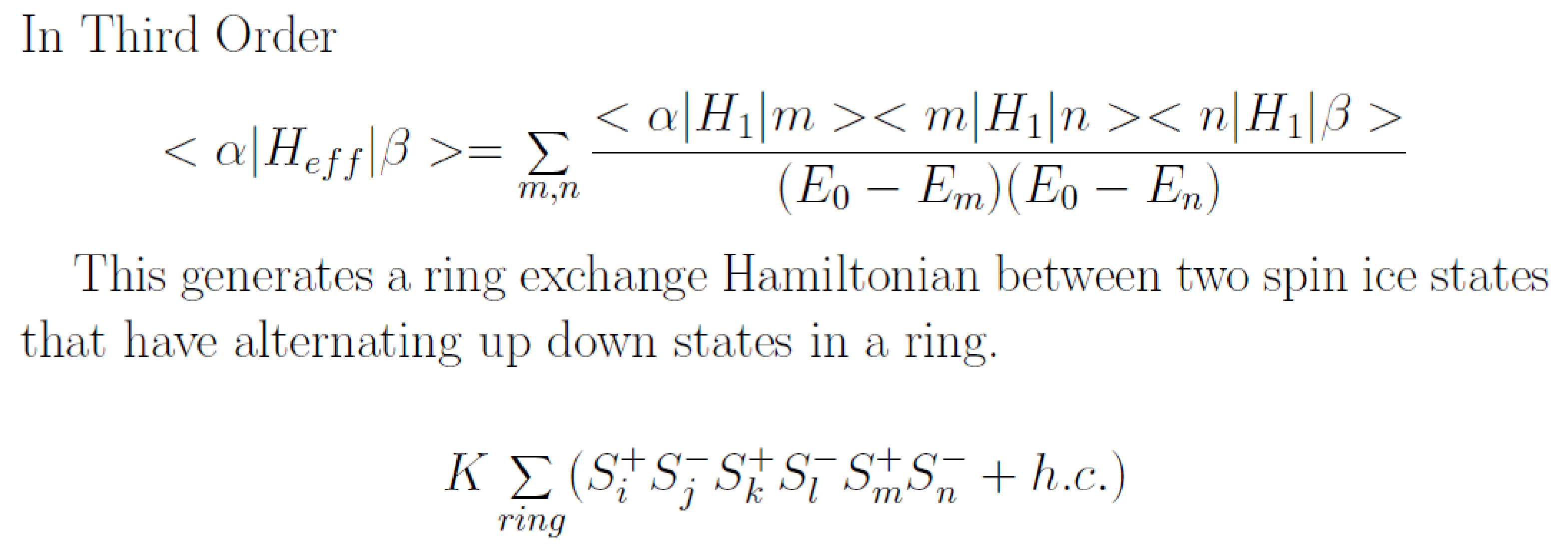
Consider a pair of spin-ice states $|\alpha>, |\beta>$

In First Order

$$<\alpha|H_1|\beta>=0$$

In Second Order

$$<\alpha|H_{eff}|\beta>=\sum_m \frac{<\alpha|H_1|m><m|H_1|\beta>}{E_0 - E_m}$$

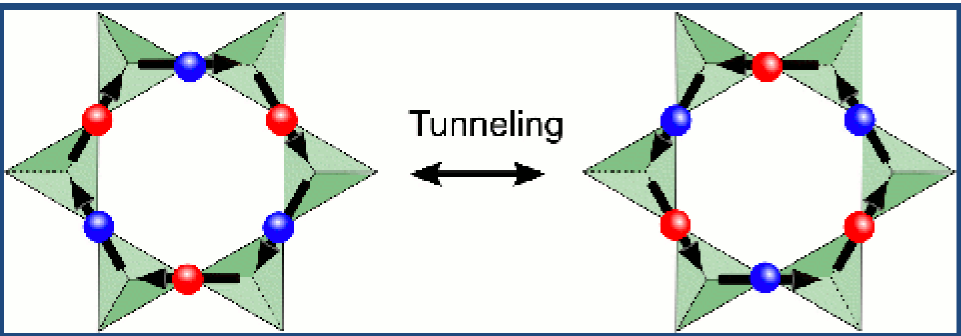


In Third Order

$$< \alpha|H_{eff}|\beta >= \sum_{m,n} \frac{< \alpha|H_1|m >< m|H_1|n >< n|H_1|\beta >}{(E_0 - E_m)(E_0 - E_n)}$$

This generates a ring exchange Hamiltonian between two spin ice states that have alternating up down states in a ring.

$$K \sum_{ring} (S_i^+ S_j^- S_k^+ S_l^- S_m^+ S_n^- + h.c.)$$



Effective Hamiltonian is Off-Diagonal
Promotes a Highly Resonating State
One can add a chemical potential for alternating ring configurations
Fine tuning leads to Kivelson-Rokhsar `equal superposition’ state

Numerical Support: Bannerjee, Isakov, Damle and Kim PRL 2008
Shannon, Sikora, Pollman, Penc and Fulde PRL 2012

IS EMERGENT QED REALIZED IN REAL MATERIALS?

NON-ZERO QUANTUM TERMS ARE PRESUMABLY ALWAYS PRESENT

($Dy_2Ti_2O_7$, $Ho_2Ti_2O_7$)?

Quantum Fluctuations can not be too small
--Will be overwhelmed by classical selection (Dipolar energies)
--Time scales will diverge at low T leading to a glassy state (Ice)

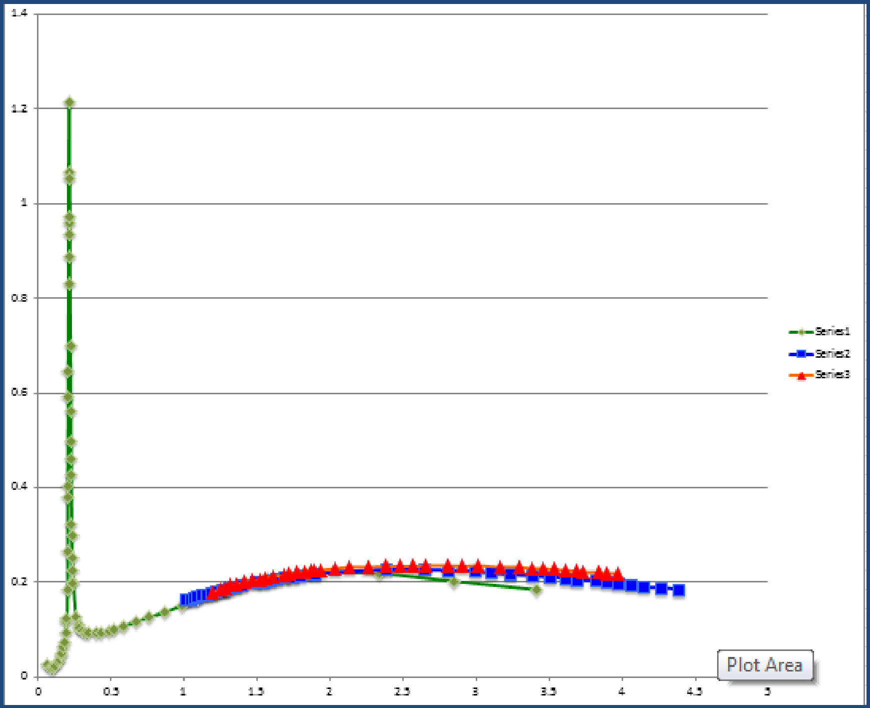
$Yb_2Ti_2O_7$ –substantial quantum fluctuations

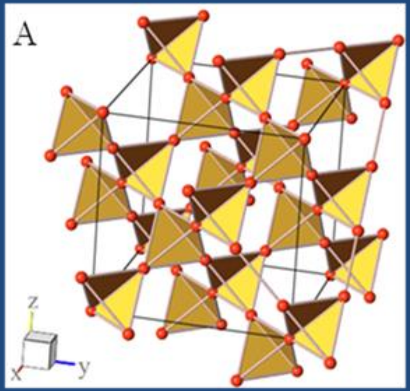
Exchange Dominated Effective Spin-half Model (Gingras)

Best characterized QSI material

Yb2Ti2O7:

Yb Spins on pyrochlore lattice (Ti is non-magnetic)
Crystal-field ground state is a Kramer's doublet well isolated from other states
Effective spin-half model

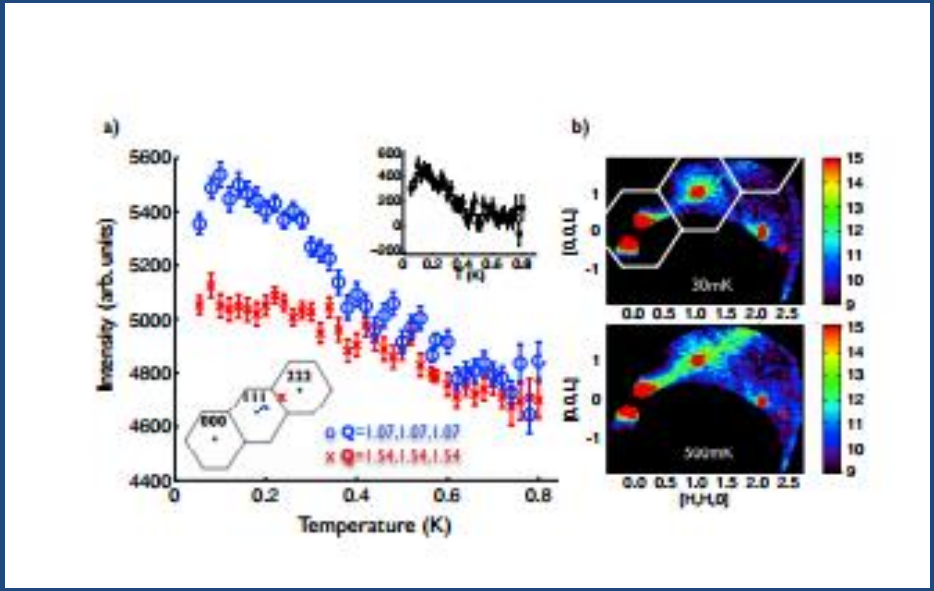


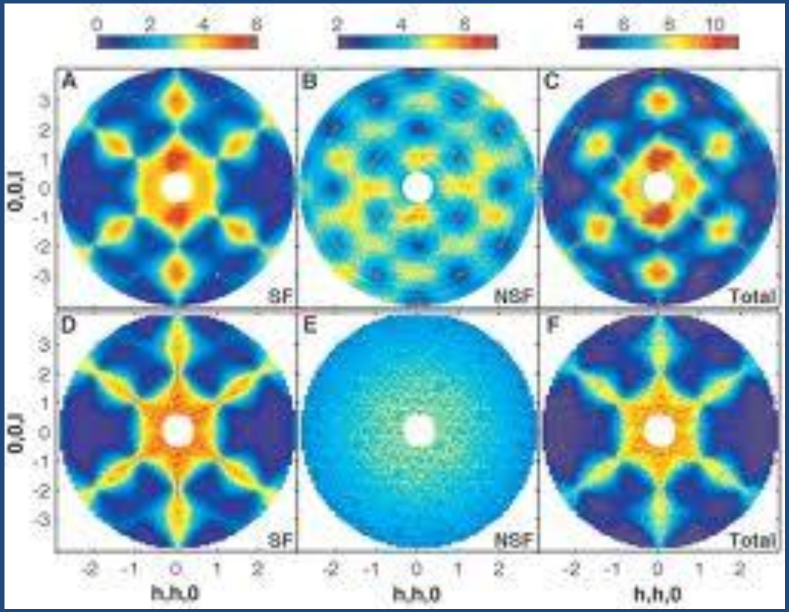


Heat capacity shows two peaks. Blote et al 1970s, Ross et al, Youanc et al
A broad hump above 2K
A sharp peak above 200 mK suggesting a first order phase transition
Data has remained largely unexplained for 40 years

Yb2Ti2O7: Neutron Scattering   Ross et al PRL 2009

Compare from Ho2Ti2O7
Fennel et al Science





Neutron Scattering in zero-field shows diffuse Rods along 111---
typical of spin ices but without pinch points
Sharpen into Bragg peaks at low T?
(Related to the 250mK peak in C?)

No sharp spin-waves at low T seen in zero field.
Sharp spin-waves appear in a high field

What is a suitable model Hamiltonian? **Kramer’s Doublet**

$$\begin{aligned}\mathcal{H}_{\mathrm{QSI}} = \sum_{<i,j>} \{ & J_{zz} S_i^z S_j^z - \lambda J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+) \\ & + \lambda J_{\pm\pm}[\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-] \\ & + \lambda J_{z\pm}[(S_i^z(\zeta_{ij} S_j^+ + \zeta_{i,j}^* S_j^-) + i \leftrightarrow j]\} \\ & - \mu_B \sum_i \{h_i^z g_{zz} S_i^z + h_i^x g_{xy} S_i^x + h_i^y g_{xy} S_i^y\}\end{aligned}$$

Nearest neighbor model that respects symmetry of pyrochlore lattice

Exchange parameters: J_{zz}, $J_{\pm}$, $J_{\pm\pm}$, $J_{z\pm}$

g-factors: g_{zz}, g_{xy}

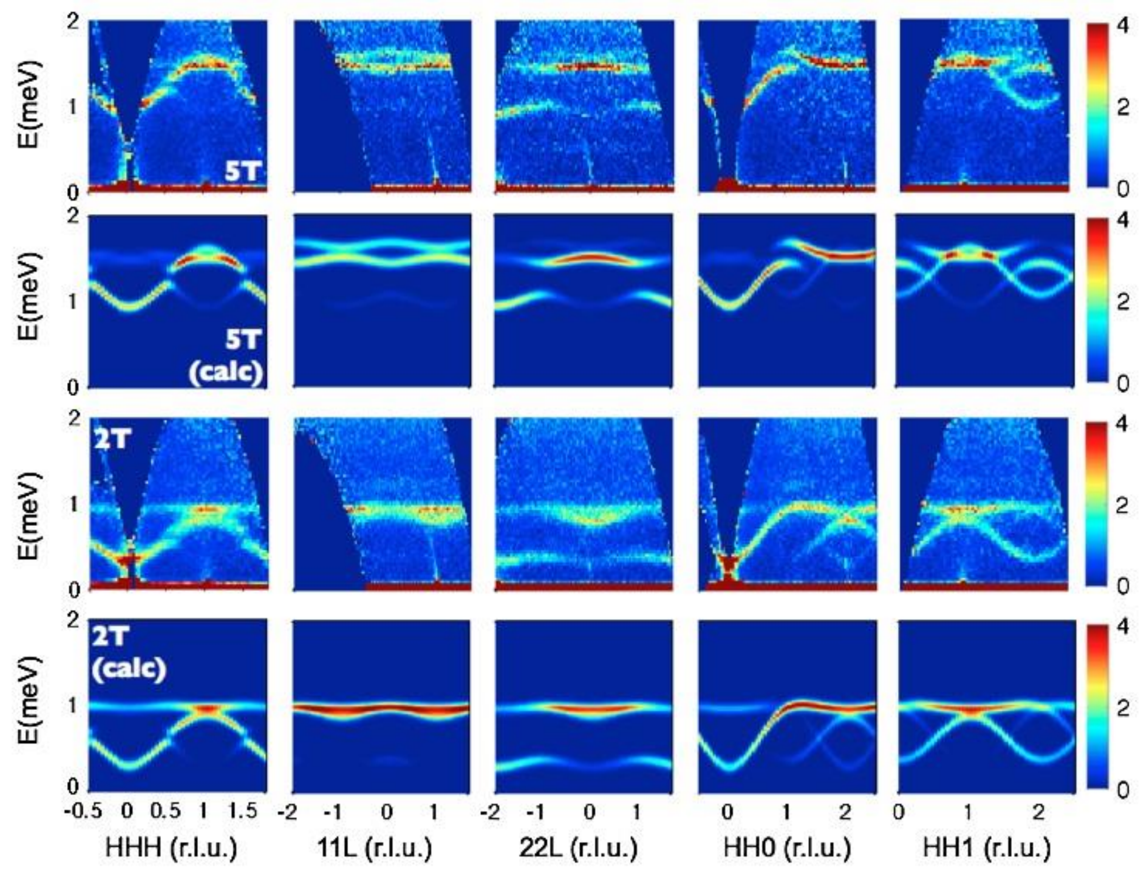
Field components at site i: h_i^z, h_i^x, h_i^y

λ dials quantum fluctuations in zero field

Does this model describe the thermodynamic behavior of Yb2Ti2O7?

How do we determine the exchange constants?

Determining Exchange Parameters
Spin-waves in a large polarizing field



Ross et al PRX : High Field Spectra can be fit to SWT to determine exchange parameters
Lack of sharp excitations in low fields suggests QSL

3 VERY DIFFERENT SETS OF EXCHANGE PARAMETERS CLAIMED

Ross et al (PRX 2012) (**High field neutron spin-wave spectra**)

Jzz=0,17 \pm 0.04, Jpm =0.05 \pm 0.01, Jpmpm=0.05 \pm 0.01, Jzpm=-0.14\pm 0.01 (meV)

Thompson et al, Chang et al (RPA based fits of zero-field structure factor)

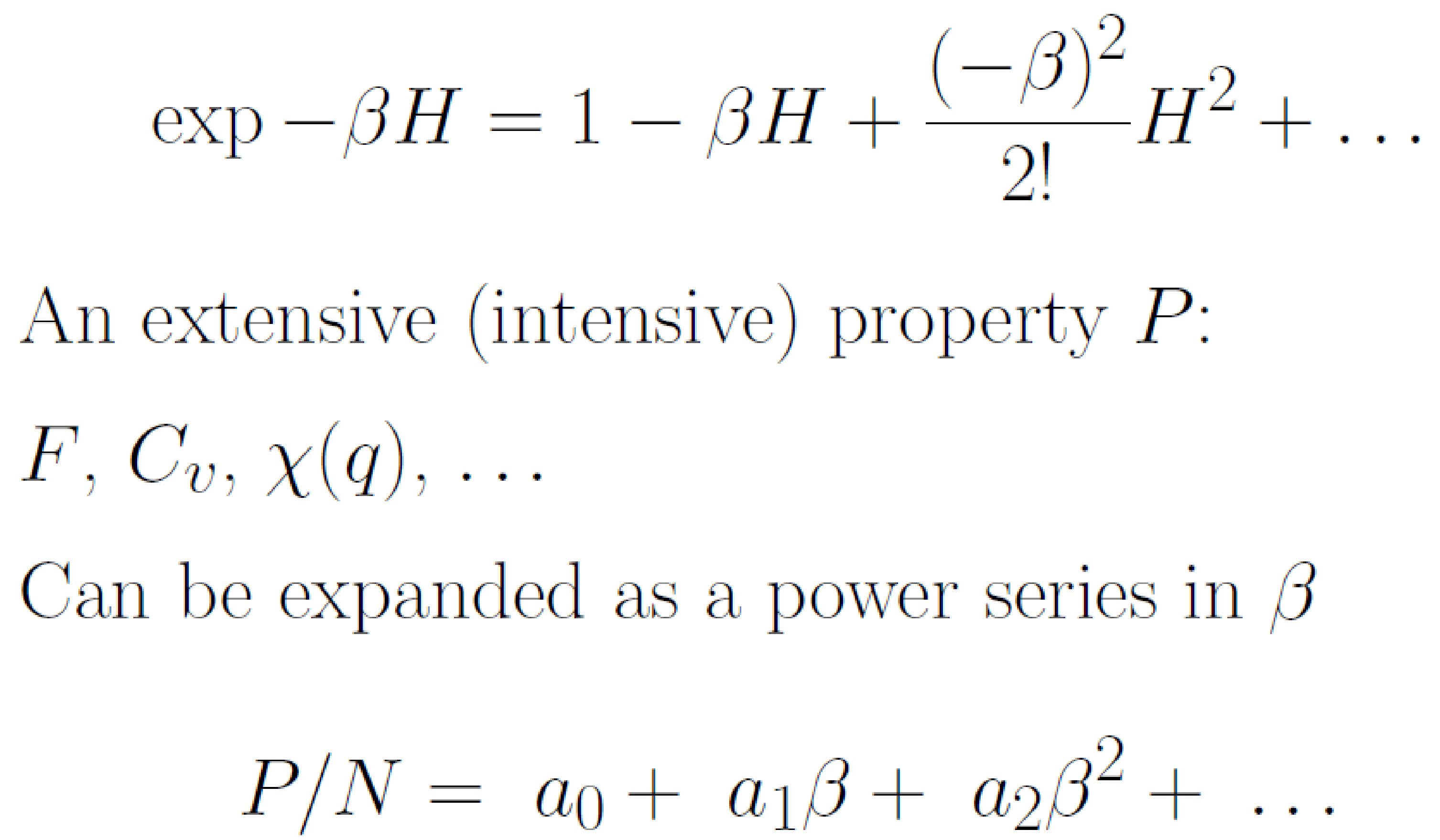
Jzz=0.01, Jpm=0.035, Jpmpm=0.01 Jzpm=-0.0424

Can any of these describe the observed thermodynamic behavior

(Note Jzz leads to classical spin ice)

How well does this model describe thermodynamic properties?

HIGH TEMPERATURE EXPANSIONS



$$\exp -\beta H = 1 - \beta H + \frac{(-\beta)^2}{2!} H^2 + \ldots$$

An extensive (intensive) property P:

F, C_v, $\chi(q)$, …

Can be expanded as a power series in β

$$P/N = a_0 + a_1\beta + a_2\beta^2 + \ldots$$

Coefficients can be calculated by Linked Cluster Method (LCM)
Oitmaa, Hamer, Zheng (Book)

HIGH FIELD EXPANSIONS

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1$$

$\mathcal{H}_0$: Field part of the Hamiltonian

$\mathcal{H}_1$: Exchange Part of the Hamiltonian

Treat H1 as perturbation. Use interaction representation to expand extensive properties
In powers of J/h (at T=0) + exponentially small corrections at low-T (exp(-c h/T))

$$P/N = a_0 + a_1\left(\frac{J}{h}\right) + a_2\left(\frac{J}{h}\right)^2 + \ldots$$

Coefficients can be calculated by Linked Cluster Methods

EXACT DIAGONALIZATION

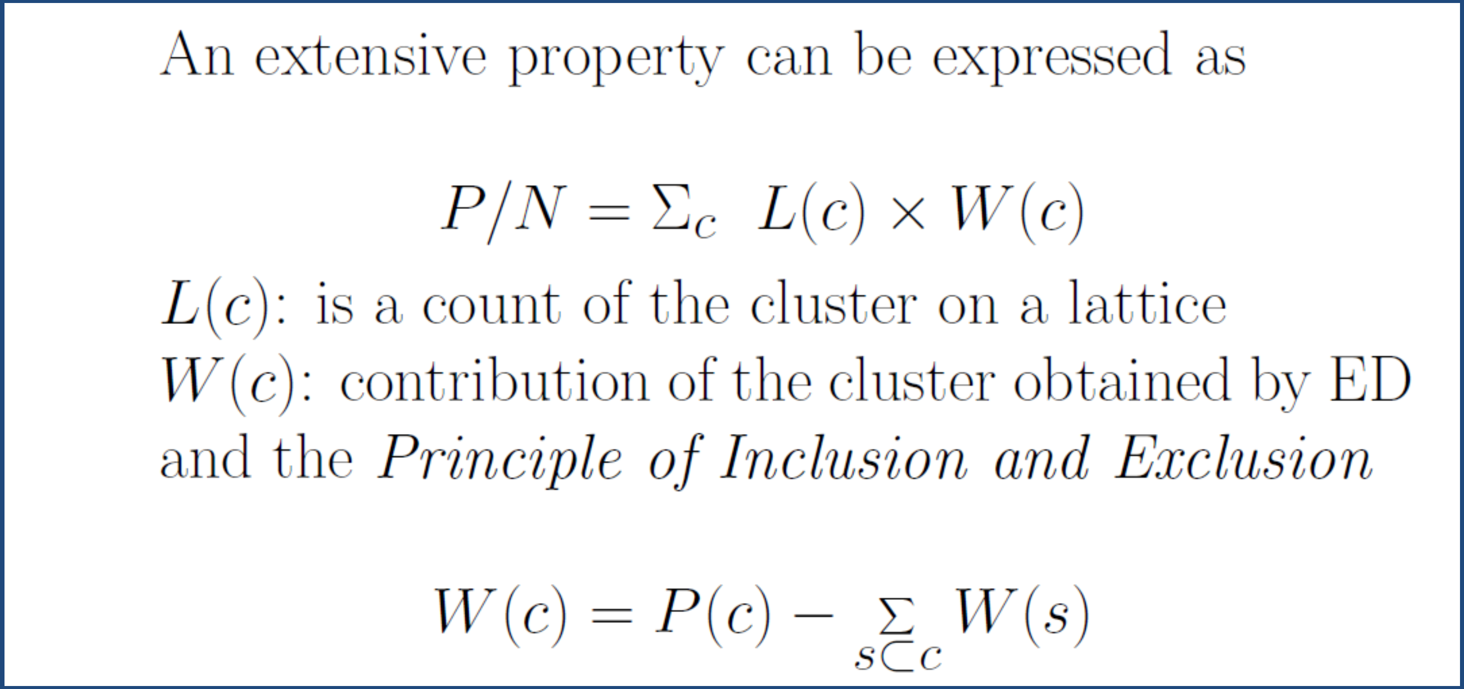
Easy if cluster size is small enough

Need PBC to avoid huge finite size effects

Becomes less useful with increased dimensionality and coordination number

NUMERICAL LINKED CLUSTER EXPANSION

Combines Linked Cluster + ED (Rigol, Bryant, RRPS PRL)



An extensive property can be expressed as

$$P/N = \Sigma_c \ \ L(c) \times W(c)$$

$L(c)$: is a count of the cluster on a lattice
$W(c)$: contribution of the cluster obtained by ED and the *Principle of Inclusion and Exclusion*

$$W(c) = P(c) - \sum_{s \subset c} W(s)$$

Obtained for any set of parameters (T, h, J, ……)
Numerically exact at high T (builds in HTE)
Numerically exact at high fields (builds in HFE)
Correct short distance Physics
Ideal for Spin-Ice (Using tetrahedral clusters)

NUMERICAL LINKED CLUSTER EXPANSION

High Temperature Expansions:

Weights of larger cluster are down by powers of 1/T

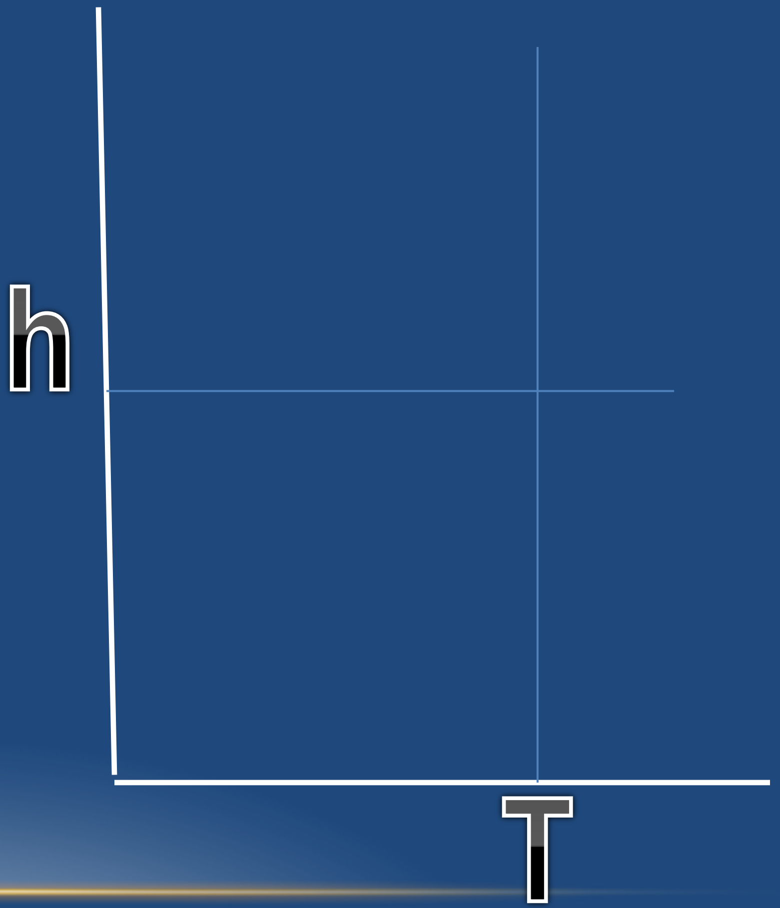
High Field Expansions:

Weights of larger clusters are down by powers of 1/h.

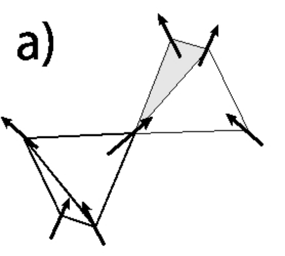
ED: Exact short distance physics

Tetrahedral Clusters: `Ice rules' always have a chance to hold.

Classical Ising Model: First Order NLC – Single Tetrahedron– Pauling Approximation



NUMERICAL LINKED CLUSTER EXPANSION
T=0 ENTROPY (ISING MODEL ON PYROCHLORE)



P = $\sum L(c) * W(c)$ (Lattice Constant L, Weight W)

Cluster 0: Single Site: S(0) = ln(2); W(0) = ln(2); L(0)=1;

S/N=ln(2)

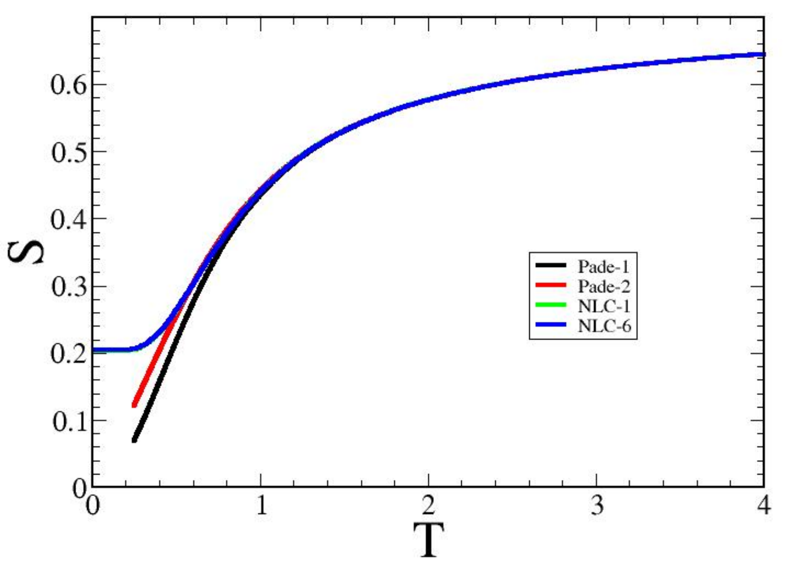
Cluster 1: One tetrahedron: S(1) =ln(6); W(1)=ln(6)-4ln(2)= ln(6/16); L(1)=1/2;

S/N=ln(2) + (1/2) ln(6/16)= (1/2) ln(3/2) (Pauling)

Ist Order NLC: Corresponds to Pauling Approx.

Accurate to a few percent down to T=0 for S, C, χ

RRPS and J. Oitmaa PRB 2012



NLC TO 4TH ORDER

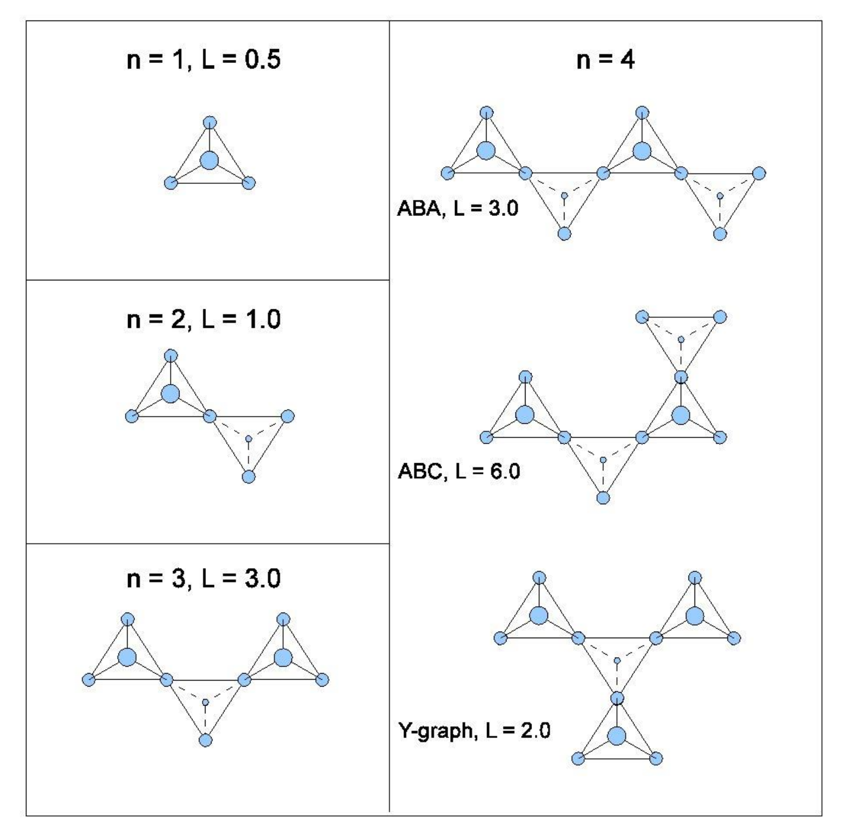
13 site clusters with no lattice symmetry
8192x8192 complex matrices

ED required 2-4 GB of memory

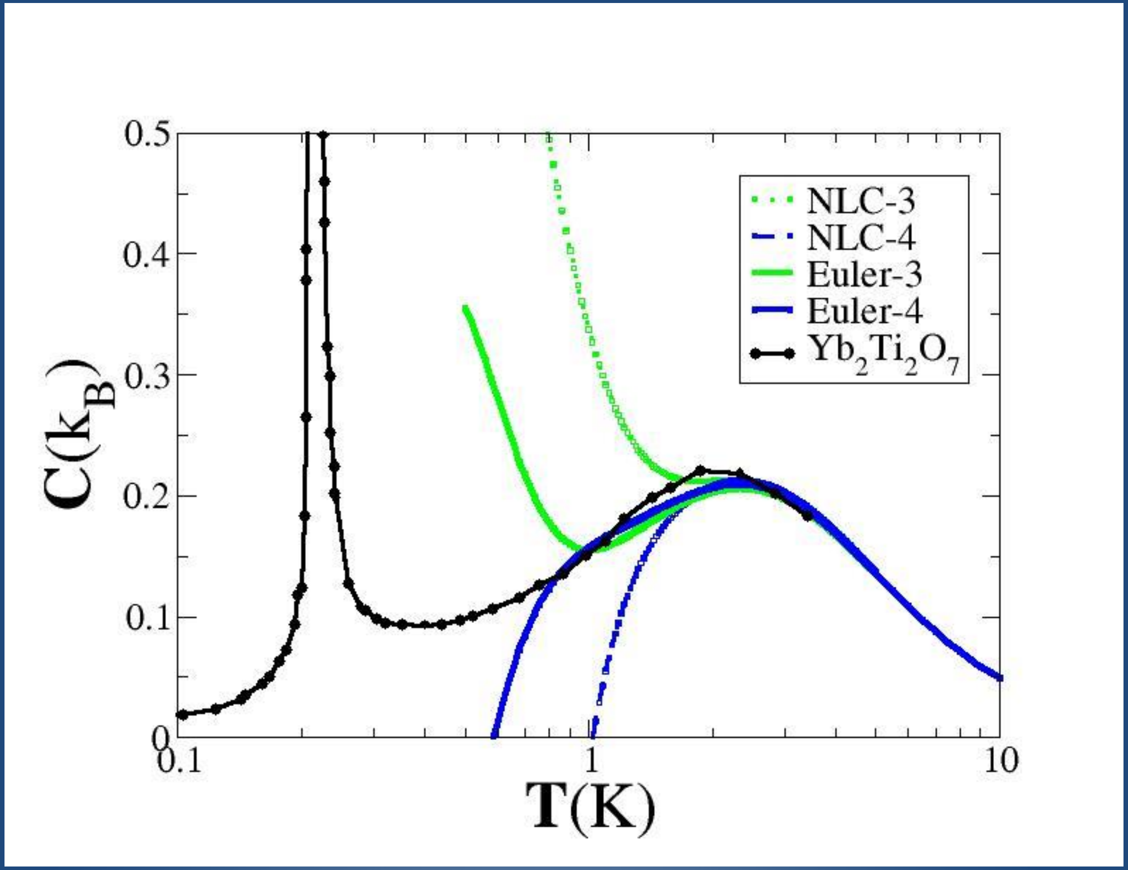
Next order: 16-site clusters memory goes up by factor of 64

Euler Extrapolation: Eliminates Leading Alternation (which sets in at low temperatures)

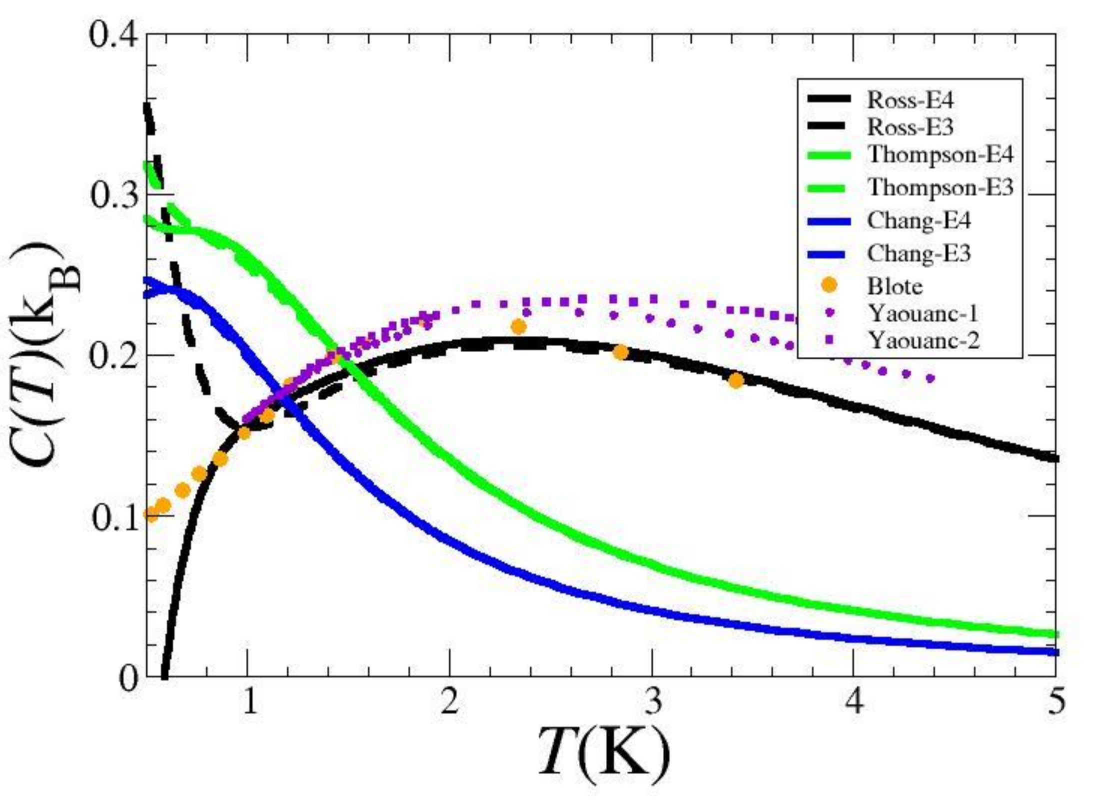
Missing: How to extrapolate for singular behavior and long-range correlations



Specific Heat: YbTO (Ross et al parameters)



Different exchange parameters proposed for YbTO



Other exchange parameter sets do not have the correct energy scale
Blote data is closest to parameters proposed by Ross et al

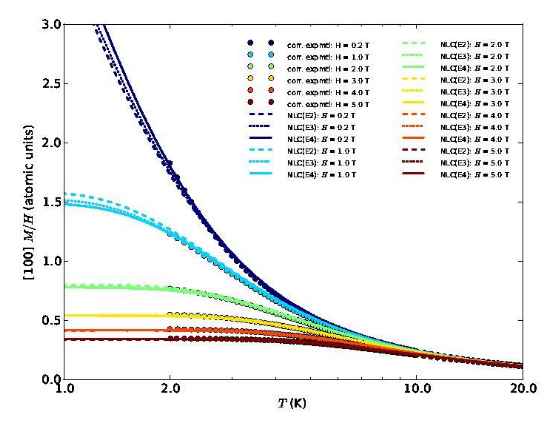
YbTO: Magnetization in a Field

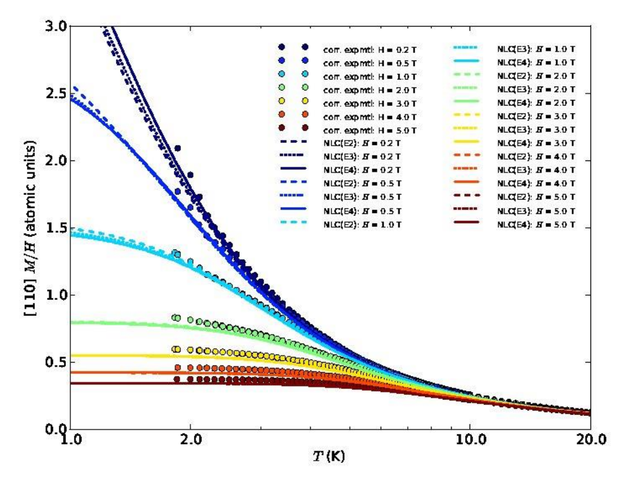
3 Different Field Directions

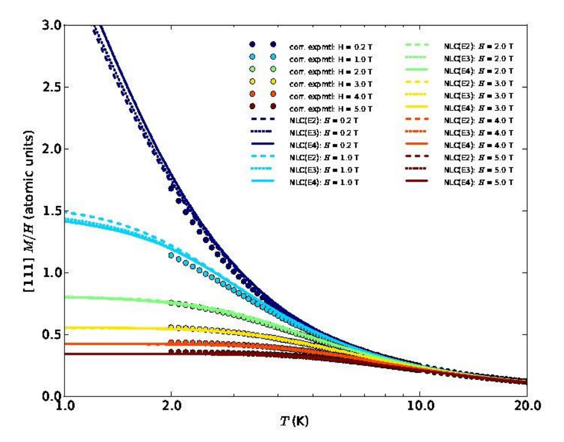
[110]   [100] [111]

No adjustment in parameters (J,g)

Demag Corrected

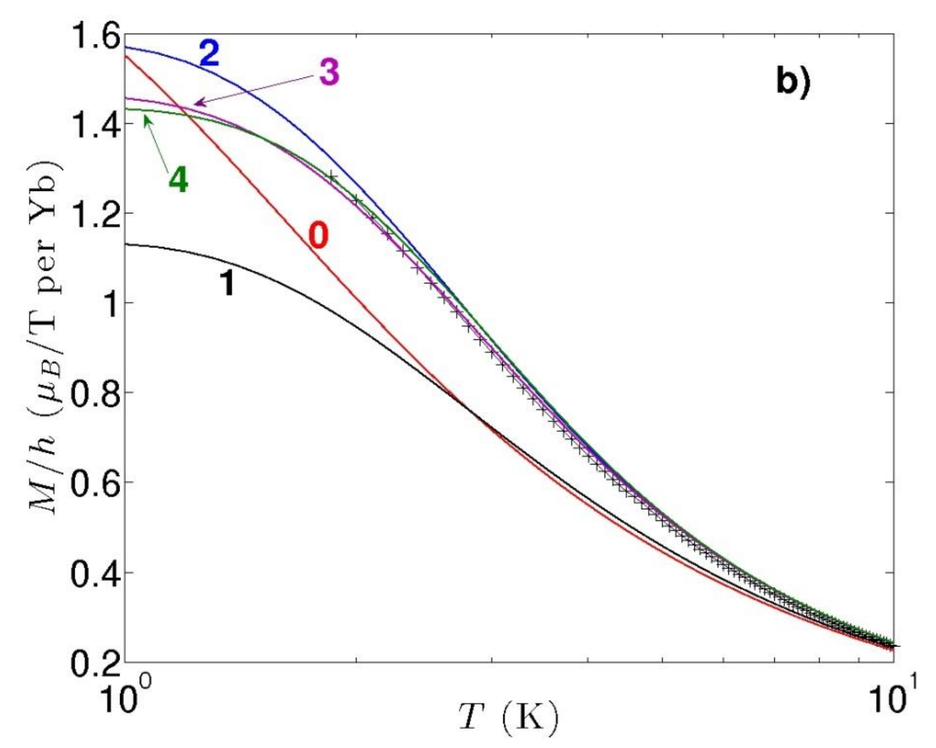






The exchange QSI model works really well for the material

DEMAG CORRECTED MAGNETIZATION

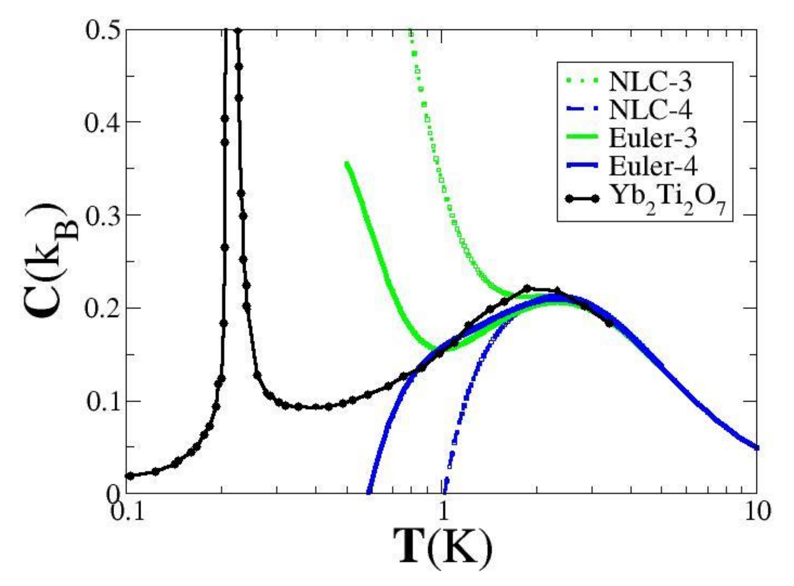


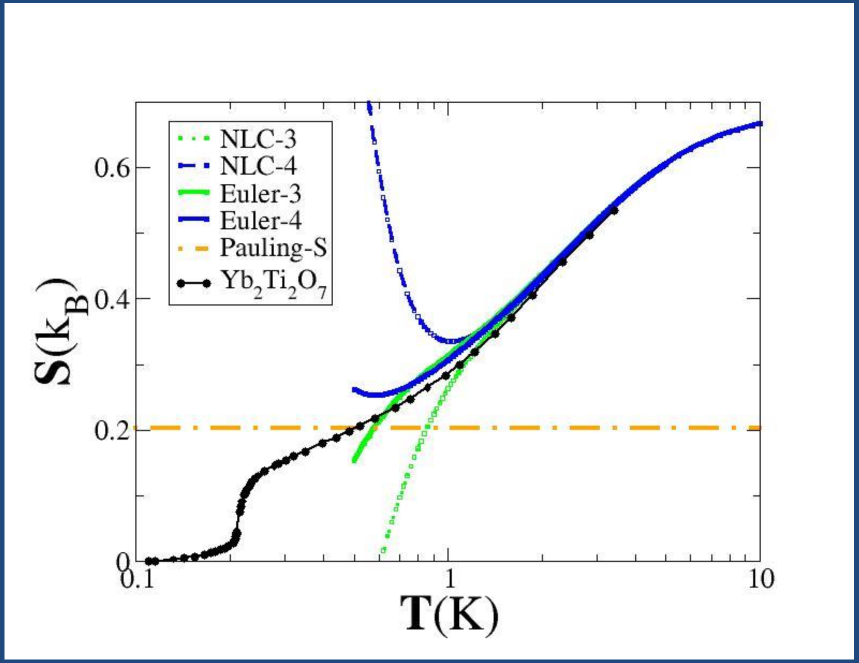
1 Tesla

Various NLC orders and experimental data

Agreement is remarkable with no adjustment of parameters

Heat Capacity and Entropy of YbTO:





Theory: Start from k ln(2) entropy at T=infinity
Experiment: Start with zero entropy at T=100 mK
Very good agreement: Regime of temperature between peaks has Pauling entropy
But, no definite plateau

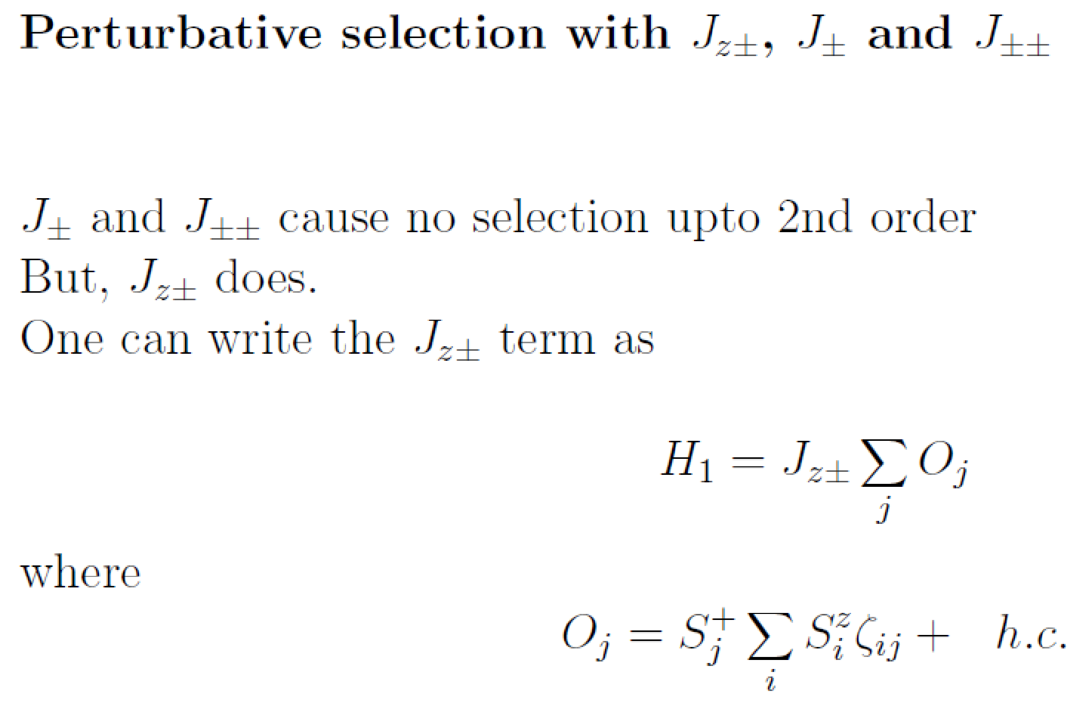
WHAT IS THE T=0.24 K TRANSITION?

NUMERICAL STUDY FAILS

DIAL DOWN QUANTUM FLUCTUATIONS

Hope the physics is smoothly connected

We have 3 quantum terms Jpm, Jpmpm and Jzpm
The largest of which is Jzpm
The latter dominates perturbation theory



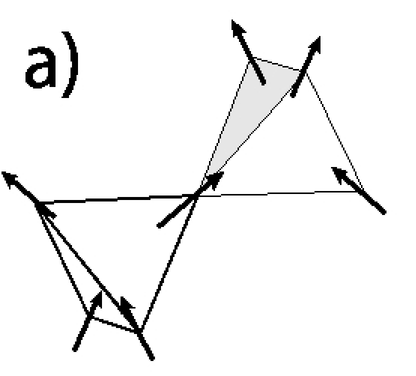
Perturbative selection with $J_{z\pm}$, $J_\pm$ and $J_{\pm\pm}$

$J_\pm$ and $J_{\pm\pm}$ cause no selection upto 2nd order
But, $J_{z\pm}$ does.
One can write the $J_{z\pm}$ term as

$$H_1 = J_{z\pm} \sum_j O_j$$

where

$$O_j = S_j^+ \sum_i S_i^z \zeta_{ij} + \quad h.c.$$



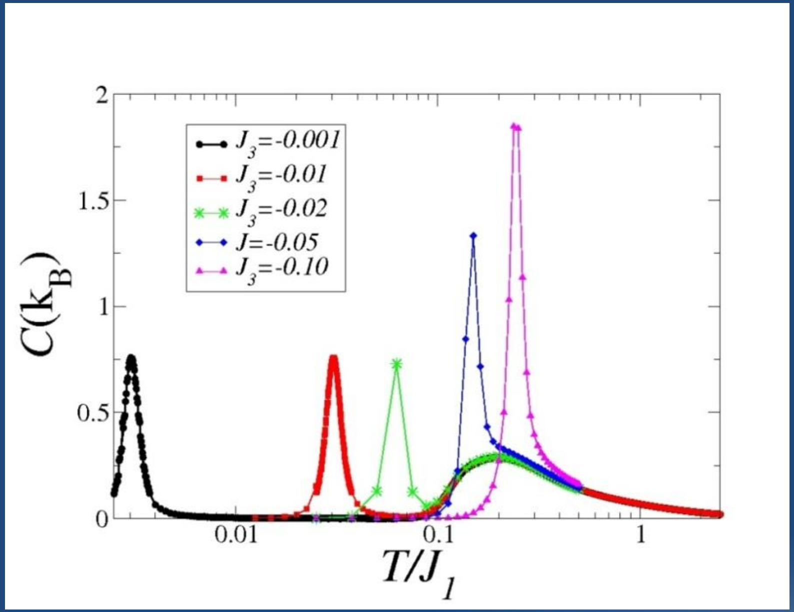
$$H_{eff} = -J_3 \sum_{<<<i,j>>>} S_i^z S_j^z$$

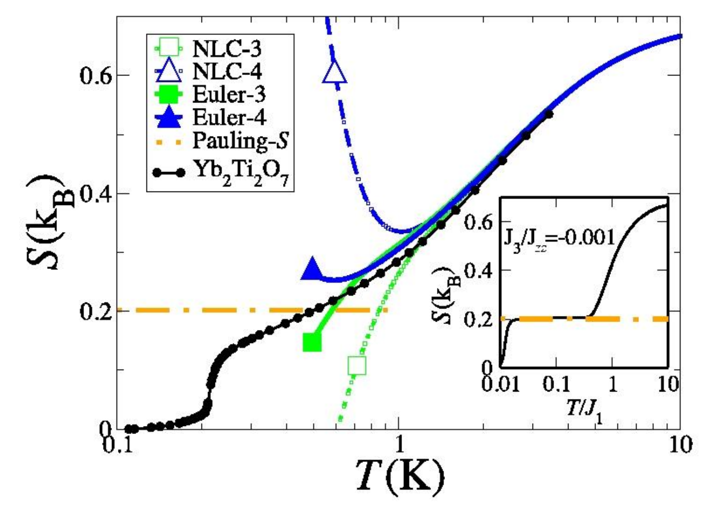
$$J_3 = 3J_{z\pm}^2 / J_{zz}$$

Interference of various terms leads to substantially enhanced FM same-sublattice coupling. It leads to selection of q=0 GS. Spin-Ice degeneracy is lifted leaving only 6 ground states. These states also cant slightly to develop a [100] moment.

WHAT IS THE T=0.24 K TRANSITION? ORDER BY DISORDER?

Low T peak in specific heat associated with q=0 FM order?

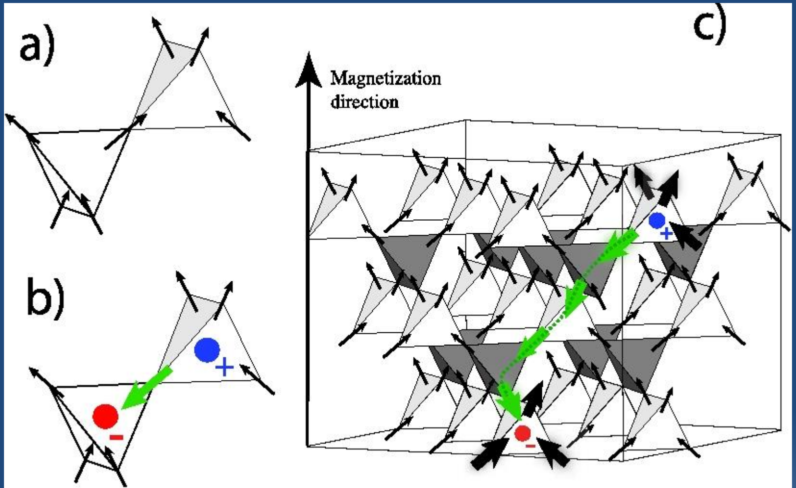




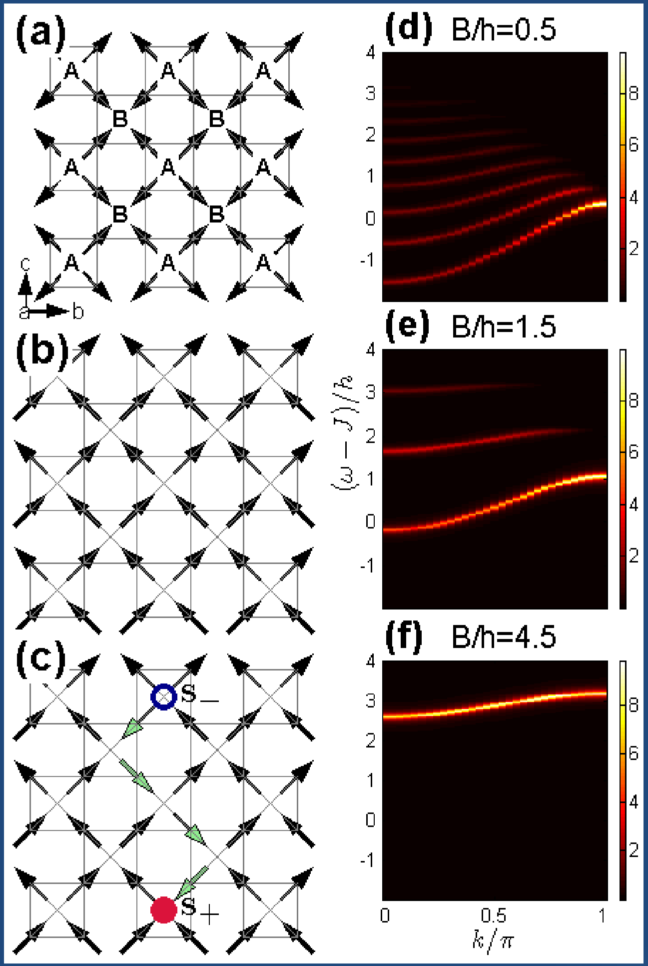
Classical Loop Monte Carlo on Effective Classical Model
First Order Transition + Clear entropy plateau for small lambda

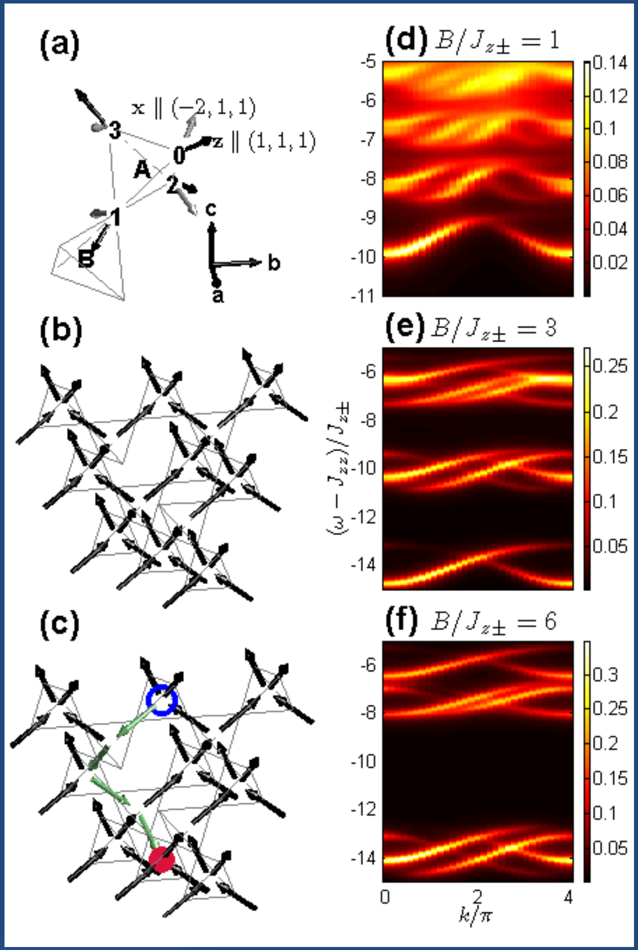
WHY NO SPIN-WAVES IN LOW FIELDS?

Despite fully ordered ground state dynamics remains non-trivial
Flipping a spin creates a spinon-antispinon pair
That can separate quite far
Hopping is order lambda while Tension is order lambda squared
Composite spin-waves : Spinon pairs with long strings



TCHERNYSHYOV: NEUTRONS SHOULD SEE MANY BRANCHES



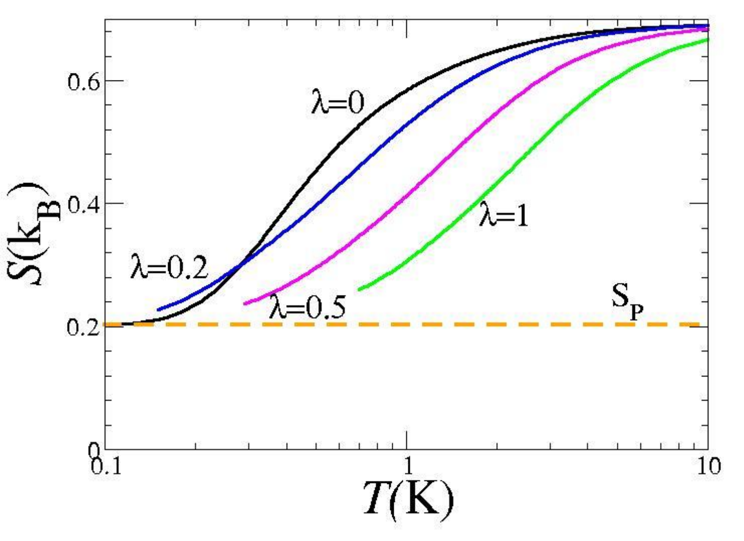


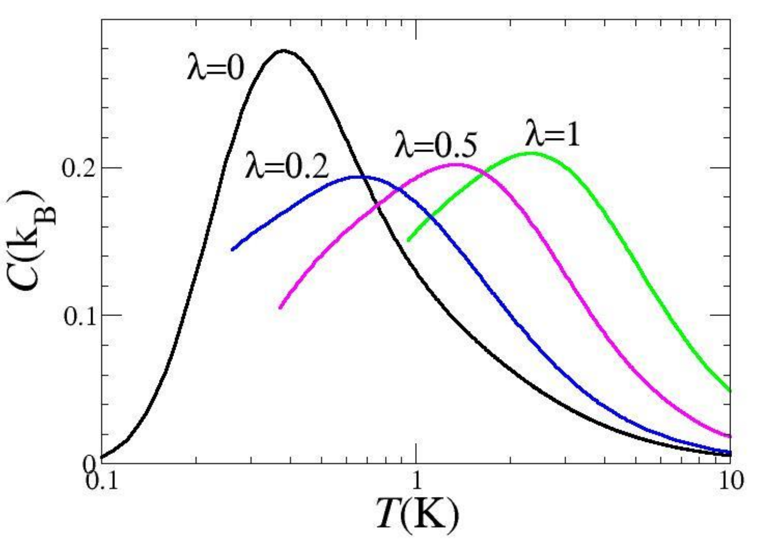
Perturbative regime should share this physics

We are working on S(q,omega)

Experiments on YbTO?

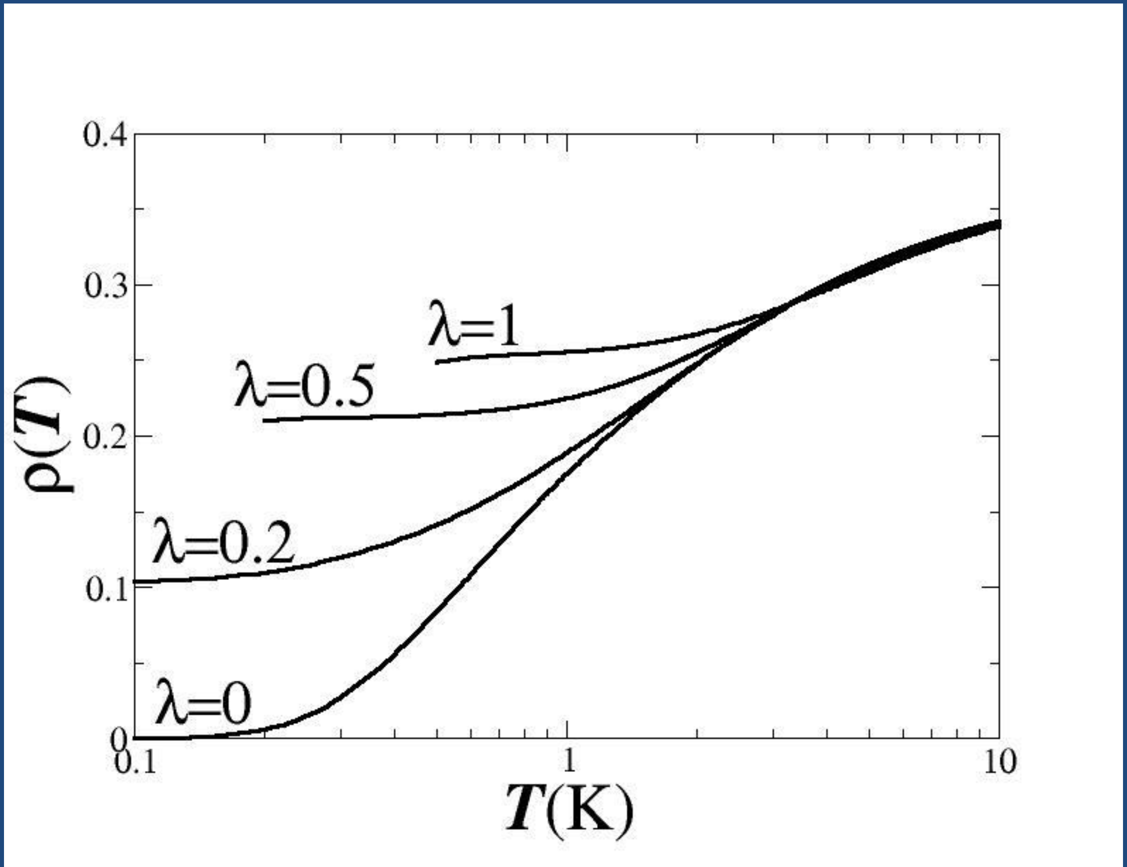
DOES THIS PICTURE CONTINUE TO LAMBDA=1?





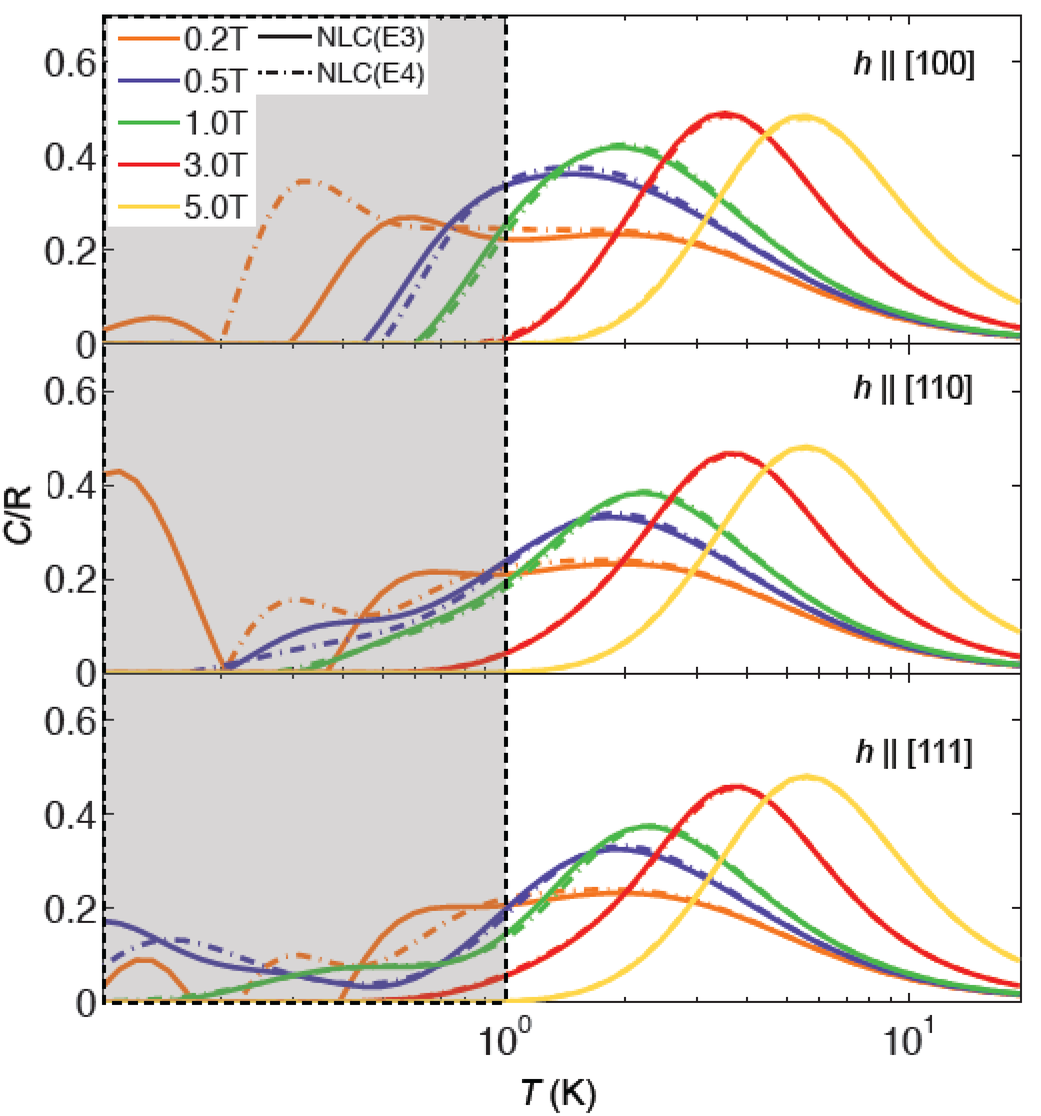
Two peaked structure but no definite separation of LRO and SRO
No clear Pauling-Like regime

DEFECT (MONOPOLE/SPINON) DENSITY (NLC)



Increasing lambda, intermediate regime is not simply classical Spin Ice
Strong Renormalization of low energy physics
Staying within the spin-ice subspace is inconsistent with quantum terms
Would pinch points arise?

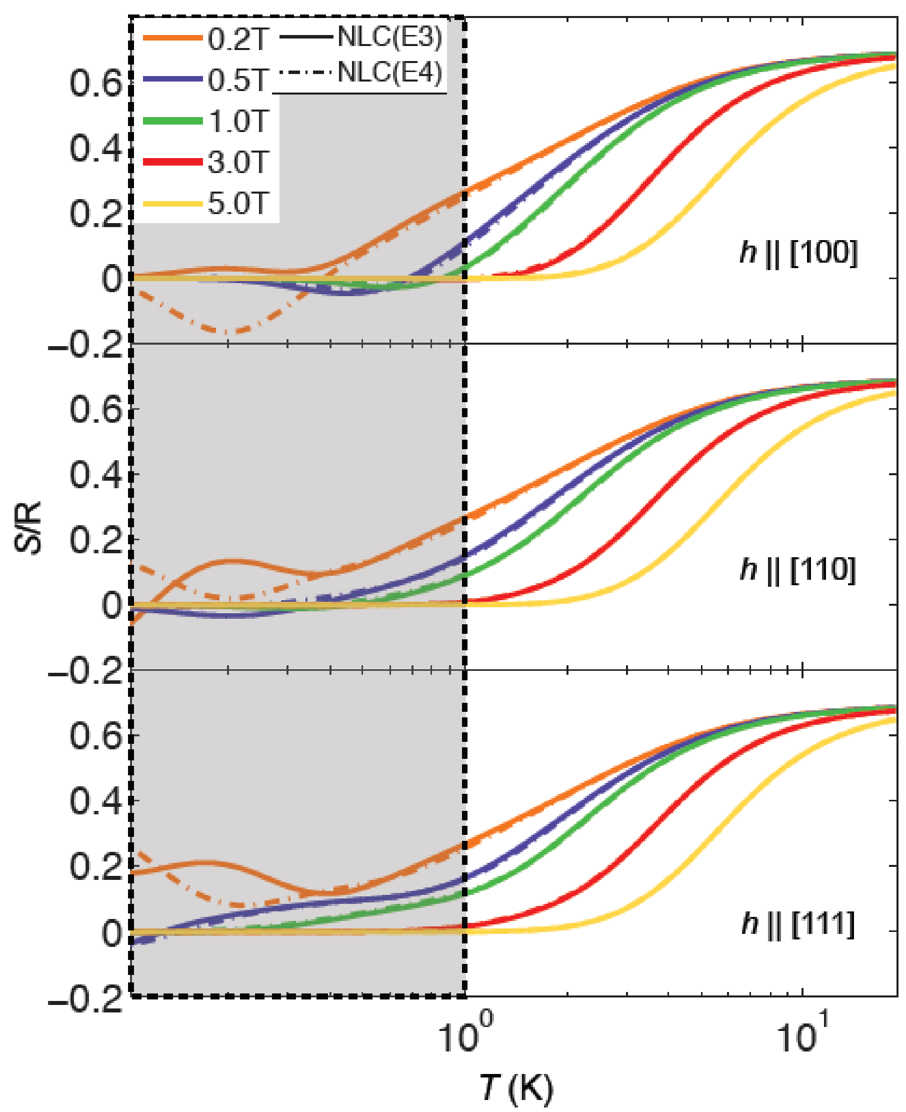
HEAT CAPACITY IN A FIELD [110]



Euler Transforms 3rd and 4th Order. At strong fields (>1T) 2nd order is good enough

Peak tracks proposed Phase Diagram for the paramagnet/ferromagnet transition and onset of sharp excitations in high fields
Ross et al PRL 2009
Peak splits in two near h=0.5 T (shoulder develops)

FIELD DEPENDENCE OF ENTROPY EVIDENCE FOR FM [100] ORDER?



Entropy removed at high T for field along 100.
Shoulder/Plateau in entropy persists for fields in some directions.
Very suggestive of degeneracies in MFT (Phase Transition in a field except 100)
Needs more experimental study.

EXPERIMENTAL SEARCHES FOR LRO

- Experimental detection of LRO remains very controversial
- Majority of experiments argue there is no LRO

Note: Its detection is masked by large nuclear peaks

Large variation from material to material (especially in

Single crystals)

Are there other weaker terms present which are only important at low temperature and low fields (lead to QSL?)?

Or is it due to domain formation? (FC along 100 should remove them)

Summary

- YbTO is rather well described by a nearest-neighbor anisotropic exchange QSI model at least at not too low temperatures and high fields
- Double-peaked Heat Capacity (SRO+LRO)
- Weak Quantum Regime: Intermediate Temperature is Spin Ice
- Low T: Conventional GS + Composite Excitations
- **Physics of YbTO is essentially the same? (Expts: String excitations?)**
- QSI but not QSL ?
- **Final answer has to come from experiments**

Looking Beyond Yb2Ti2O7

YbTO is a QSI but most probably not a QSL

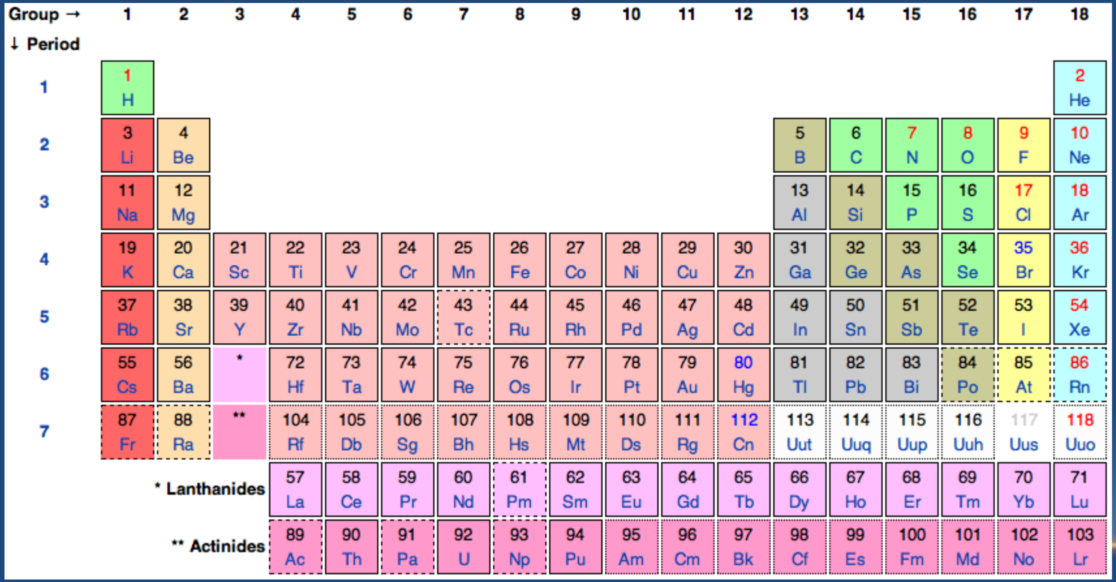
but

Theoretical methodology is in place (High field spectra/ Thermodynamics)

Experimental techniques are in place (Neutron Scattering **Broholm**)

Many variety of spin-ice materials

We should have a QSL in QSI in next two years: Leon Balents



Group → ↓ Period	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	*	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	**	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo
		* Lanthanides		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
		** Actinides		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

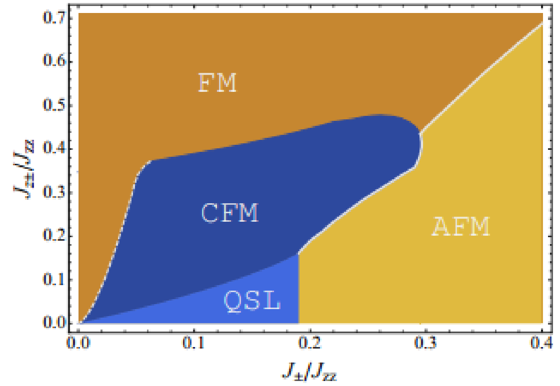


FIG. 3. Modified gMFT diagram, which takes into account the known perturbative limit $J_{z\pm}/J_{zz} \ll 1$. Note that the FM-CFM transition (white dashed line) in the latter region is a *sketch*.

Savary and Balents PRL

THE END

THANK YOU