Title: Pyrochlore Material Yb_2Ti_2O_7: A Model Quantum Spin Ice

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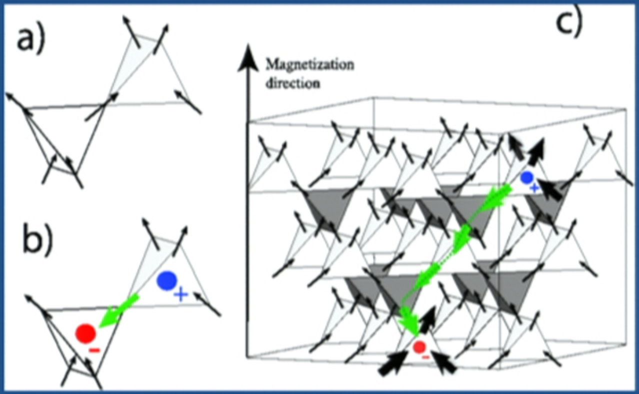
Abstract: We discuss the thermodynamic properties of the model exchange quantum spin ice material Yb_2Ti_2O_7. Using exchange parameters recently determined from high-field neutron scattering measurements, we calculate the thermodynamic properties of this model system. We find very good agreement with the heat capacity, entropy and magnetization measurements on the materials. We show that, in the weak quantum regime, quantum fluctuations lead to the selection, within the spin-ice manifold, of a conventional ordered ground state. However, the excitations above the ground state and their dynamics remain highly non-trivial. They consist of weakly bound spinon-antispinon pairs separated by long strings.

We present several pieces of indirect evidence that the low temperature phase transition observed in these materials is to this conventional ordered state.

$Yb_2Ti_2O_7$: A model Quantum Spin Ice



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

M. Gingras, T. Lin U. Waterloo

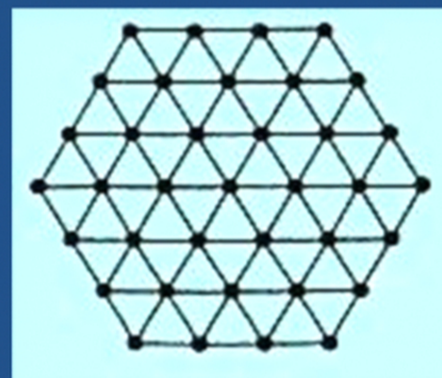
A. G. R. Day Sherbrooke

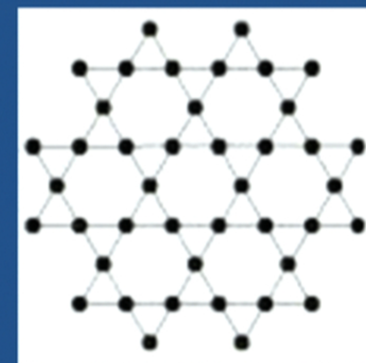
K. Ross, B. Gaulin McMaster

OUTLINE

- Introduction: Frustrated Quantum Spin Systems
- Experimental searches for Quantum Spin Liquids
- Ice rules, Spin Ice and Quantum Spin Ice
- $Yb_2Ti_2O_7$: A model system for Quantum Spin Ice
- Numerical Linked Cluster Expansions
- Perturbative Regime: Dialing Down Quantum Fluctuations
- Classical Ground State+ Composite Excitations
- 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)

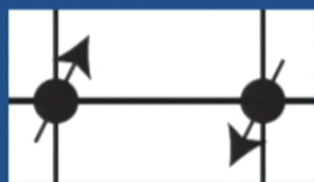
Ground state entropy under 50% of total entropy

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

Ground state entropy about 72% of total entropy

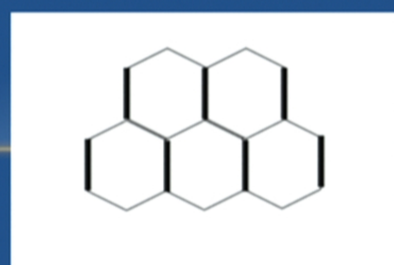
Quantum Fluctuations in a degenerate system can lead to

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

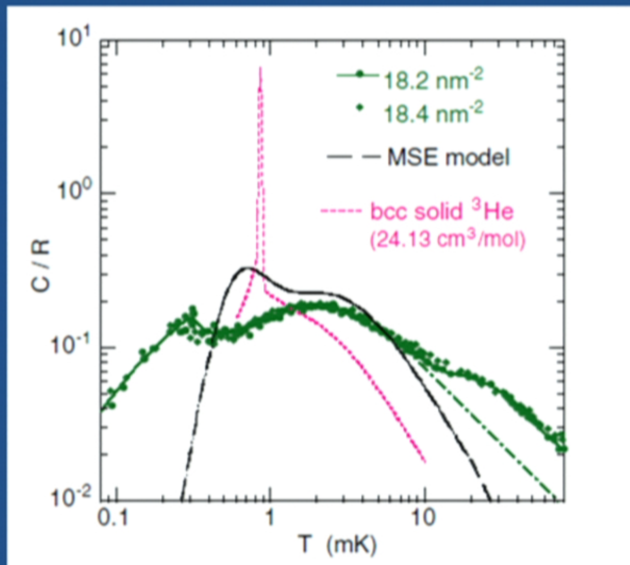
Fractional Excitations, Topological Order, Exotic Q. Criticality

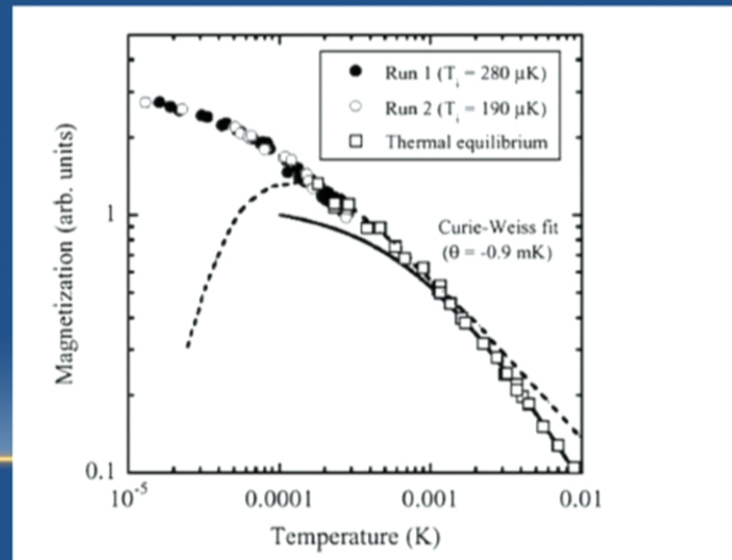
Quantum Field Theory of Many Body Systems X.G.Wen

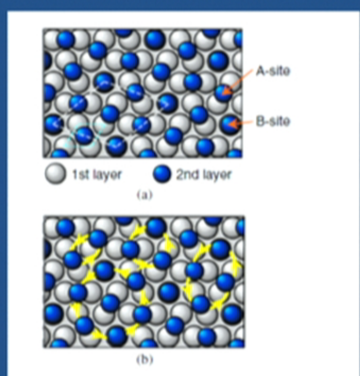
Do real materials exist which show such emergence? (exclude d=1)
What are their signatures? Entanglement Entropy, Large η (Melko)
Triangular Lattice Heisenberg Model (LRO?)
Kagome Lattice Heisenberg Model (Z2 QSL? S.R. White)
Honeycomb Lattice Hubbard Model (Intermediate U Muramatsu)
Triangular Lattice Hubbard Model (Intermediate U Motrunich)
Quantum Dimer Models (Moessner/Sondhi)
Square Lattice J1-J2 Model (Z2 QSL/VBS/Chiral SL?)

Experimental evidence: Gapless spin liquids?

Experimental Evidence: Gapless spin-liquid?





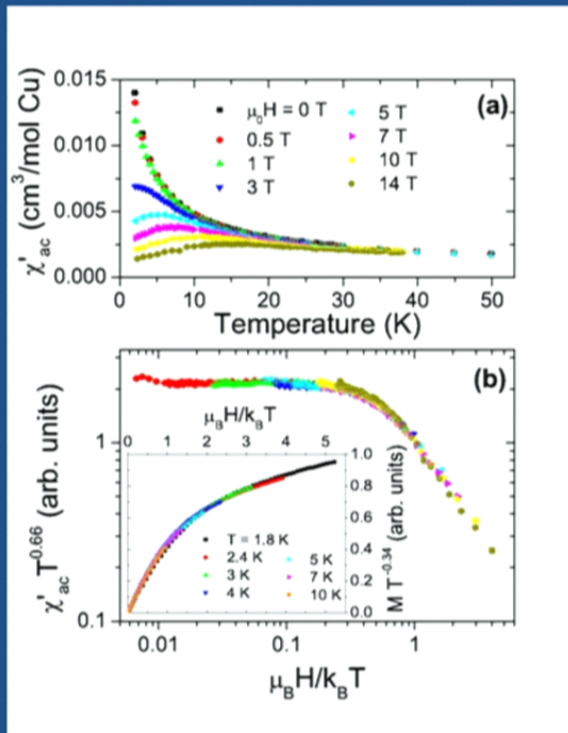


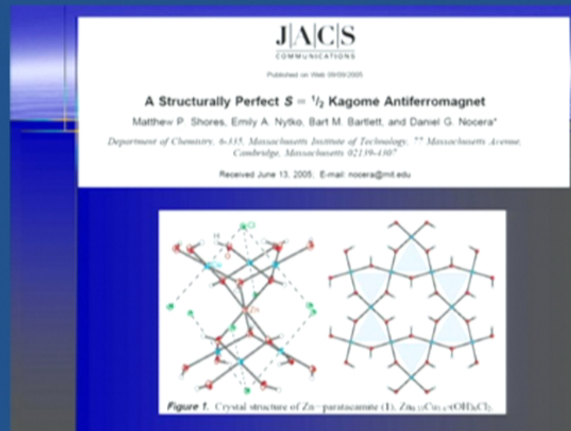
Specific Heat and Susceptibility of second layer Helium-3 on Graphite

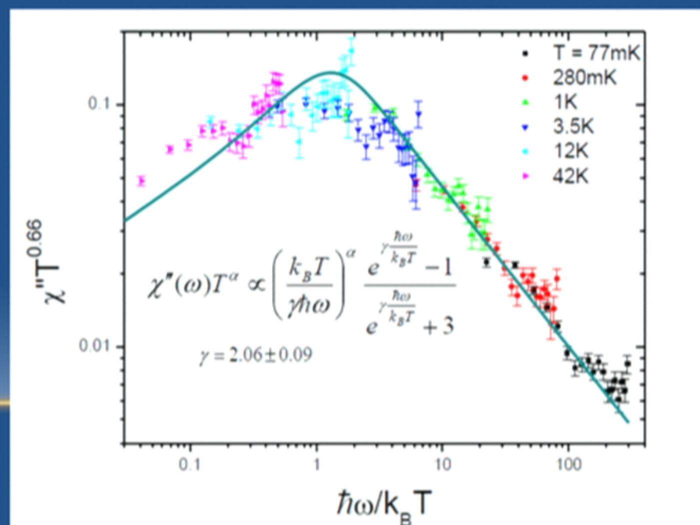
---Review by Fukuyama (JPSJ)

MSE Model Misguich et al PRL 1998

Herbertsmithites (Helton et al)





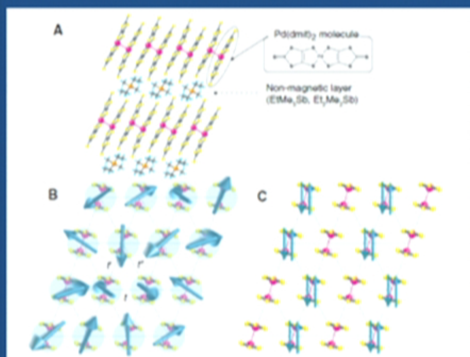


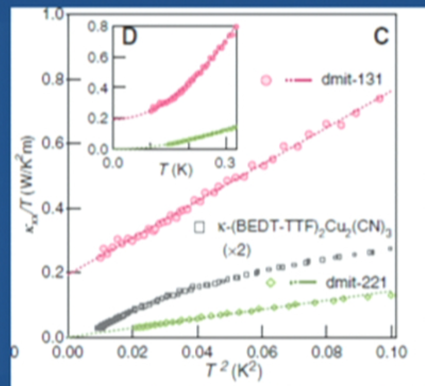
No spin gap

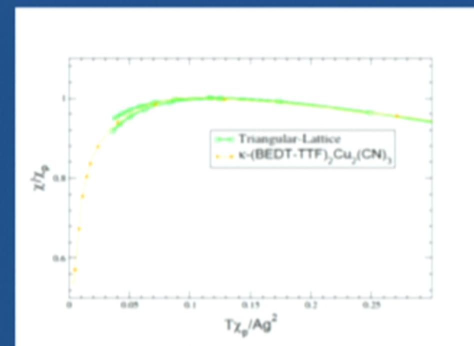
Organic Molecular Crystals

Many candidate spin-liquids

Any spin-gap is very small







Highly Mobile Gapless Excitations in a Two-Dimensional Candidate Quantum Spin Liquid

Minoru Yamashita,[1*] Norihito Nakata,[1] Yoshinori Senshu,[1] Masaki Nagata,[1] Hiroshi M. Yamamoto,[2,3] Reizo Kato,[2] Takasada Shibauchi,[1] Yuji Matsuda[1*]

Shimizu et al: Triangular Lattice
Finite susceptibility at T=0
No order for T=J/3000

Are other degrees of freedom interfering?

Organic Molecular Crystals

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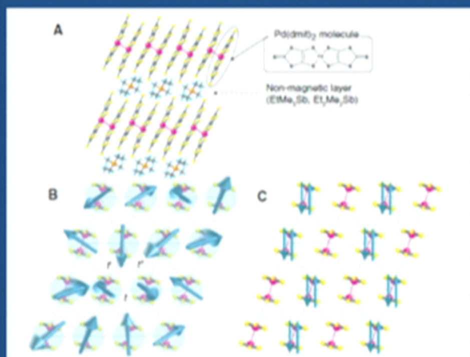

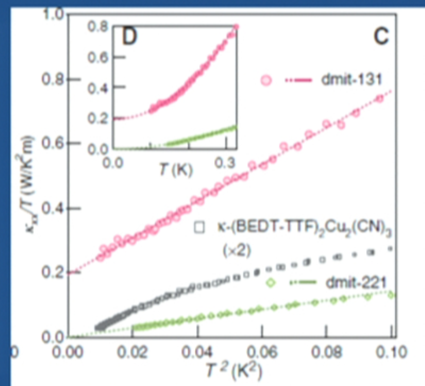

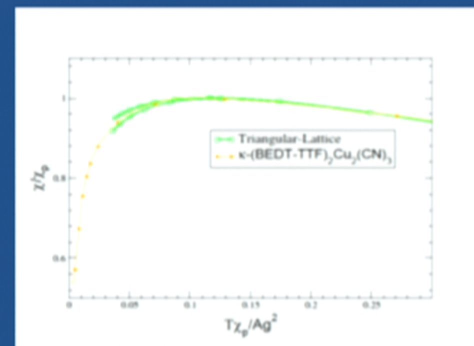

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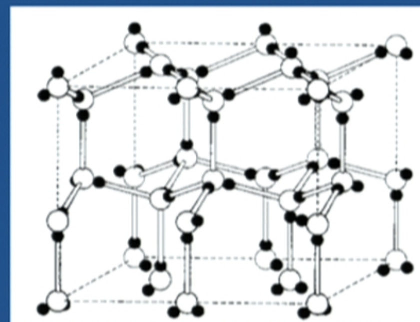
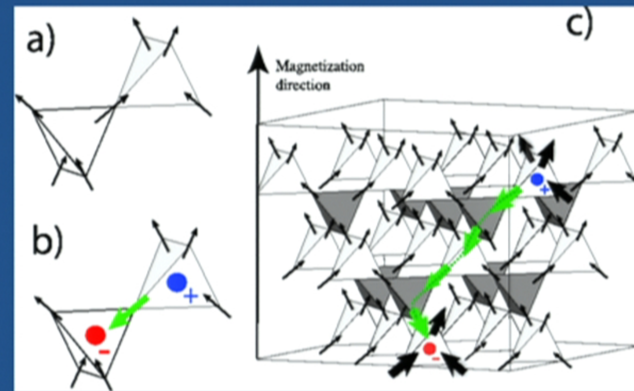
Are other degrees of freedom interfering?

ICE RULES AND SPIN ICE

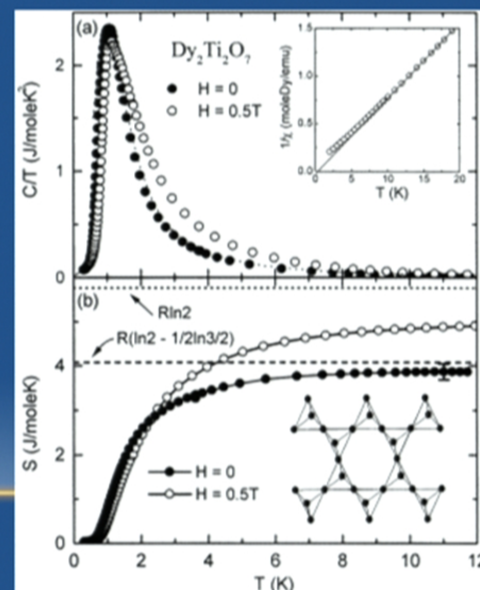




Ordinary ice has a high residual entropy (Pauling)
Exponentially Large Degenerate Configurations

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



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)

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

Second Ice Age --- Leon Balents

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

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

Degenerate Perturbation Theory in the Spin Ice subspace

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

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

Perturbation Theory: Selection at small λ

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Degenerate Perturbation Theory in the Spin Ice subspace

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

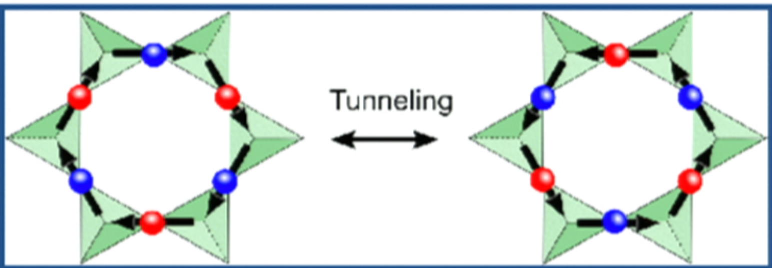
$$< \alpha | H_{eff} | \beta >= \sum_m \frac{< \alpha | H_1 | m >< m | H_1 | \beta >}{E_0 - E_m}$$

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.

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

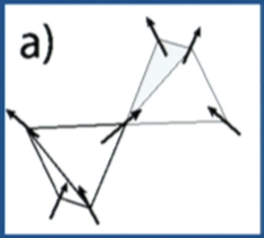


Effective Hamiltonian is Off-Diagonal
Promotes a Highly Resonating State
One can add a chemical potential for alternating ring configurations
Fine tuning leads to Kivelson-Rokhsar `equal superposition' state

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

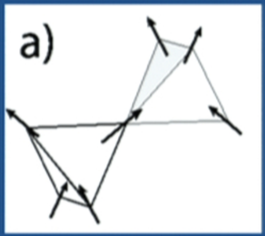
Mapping to QED: Very simplistic picture

Spin Ice subspace is a low energy subspace separated from rest.
Can disturbances travel in spin ice without an energy gap?
Just as electromagnetic waves travel in QED (Maxwell's equations)



- **Spin configurations define vector fields. Live on the original pyrochlore lattice**
- **Their sources (divergence) live on centers of tetrahedra—diamond lattice**
- **Charges/Monopoles are absent with spin-ice rules (2 in 2 out)**
- **So far all classical and called magnetic field/monopole by spin ice community**

Mapping to QED: Very simplistic picture

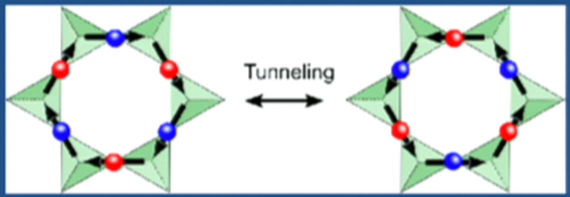


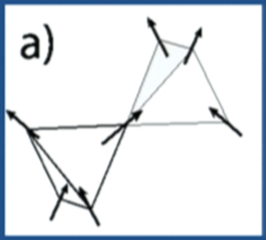
Helpful to call it Electric Field/Charge to make analogy to QED

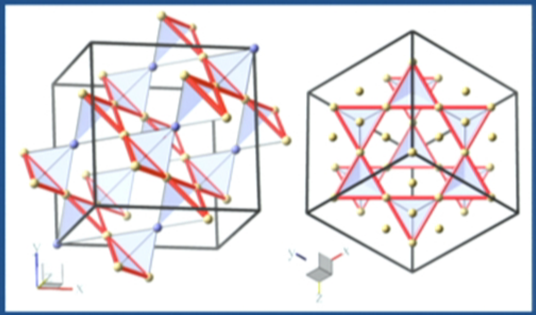
- **Quantum Mechanics allows for superposition:**
- **Phase phi determines amplitude of up and down spins at a site**
- **Phase is conjugate to spin (S+ vs Sz) – Uncertainty in one or another**
- **Plays the role of Vector potential (Dirac's QED)**

Spins define Electric Fields and Vector Potential
Live on the original pyrochlore lattice

Mapping to QED: Very simplistic picture



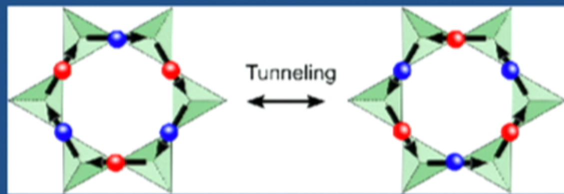


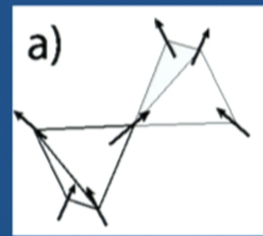


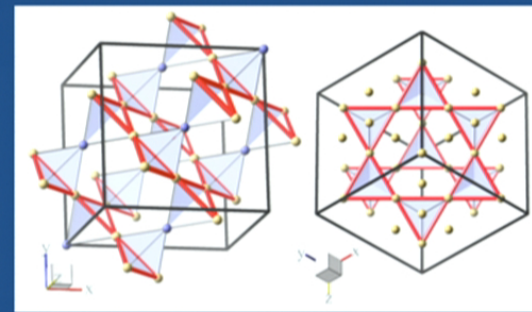
- Resonance on elementary hexagon represents tunneling from one spin ice state to another
- Curl of the vector potential on the hexagon defines a magnetic field
- It lives on the centers of hexagons
- Four types of haxagons, their centers also form a pyrochlore lattice
- Magnetic Fields live on a second pyrochlore lattice
- Their sources (magnetic charges) live on a second diamond lattice

Quantum system has dual set of fields and sources

Mapping to QED: Very simplistic picture







- **All this put together leads to QED and propagating gapless photons—**
- **Fluctuating electric field on a site, leads to changed magnetic fields on nearby hexagons**
- **Which in turn allows spins in hexagons to tunnel**
- **Which in turn changes electric fields on further away sites**
- **And the gapless collective mode (photons) can propagate in spin ice!!**
- **Only possible in an RVB (deconfining) phase with no bias in allowed configurations**

Shannon et al --- Signatures of QSL in Neutron Scattering

IS EMERGENT QED REALIZED IN REAL MATERIALS?
NON-ZERO QUANTUM TERMS ARE PRESUMABLY ALWAYS PRESENT

($Dy_2Ti_2O_7$, $Ho_2Ti_2O_7$)?

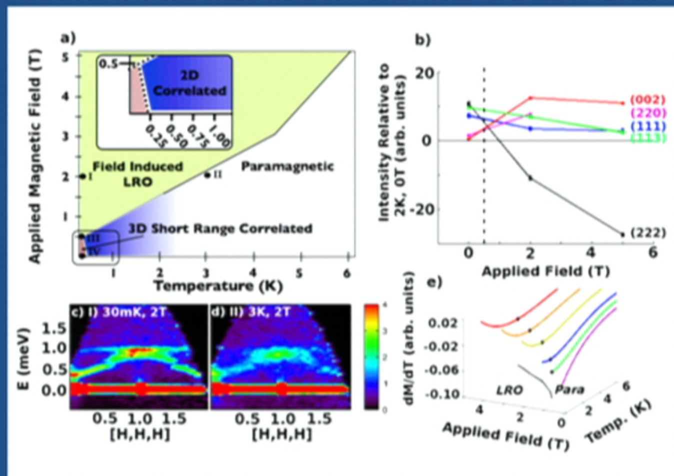
Quantum Fluctuations can not be too small
--Will be overwhelmed by classical selection (Dipolar energies)
--Time scales will diverge at low T leading to a glassy state (Ice)

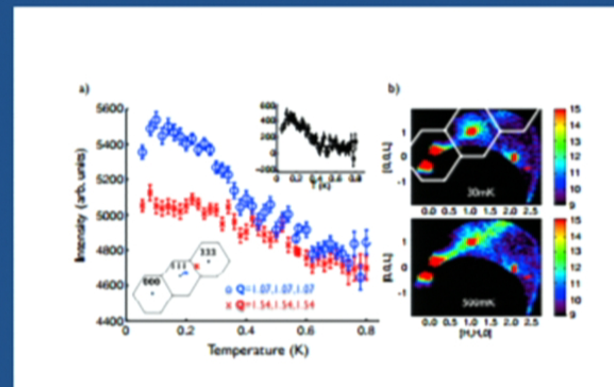
$Yb_2Ti_2O_7$ –substantial quantum fluctuations

Exchange Dominated Effective Spin-half Model (Gingras)

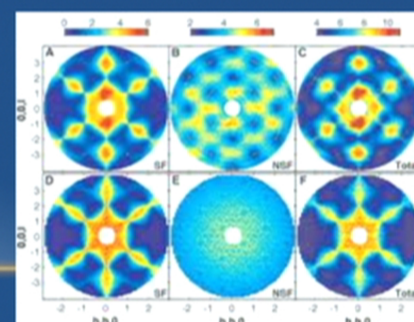
Best characterized QSI material

Yb2Ti2O7: Phase diagram proposed by Ross et al PRL 2009





Neutron Scattering in zero-field shows diffuse Rods along 111---
typical of spin ices but without pinch points
No clear evidence for magnetic long range order
No sharp spin-waves seen in zero field.
Sharp spin-waves appear in a field
Field induced ferromagnetic order
What happens at/below the 250mK peak in T=0?



Ho2Ti2O7 Fennel et al Science

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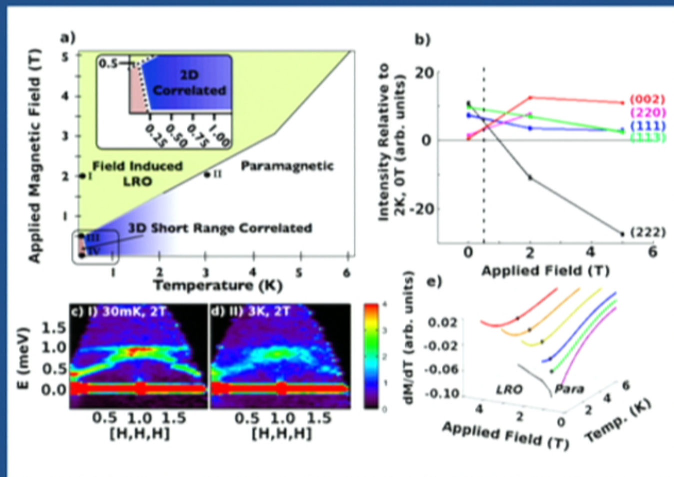

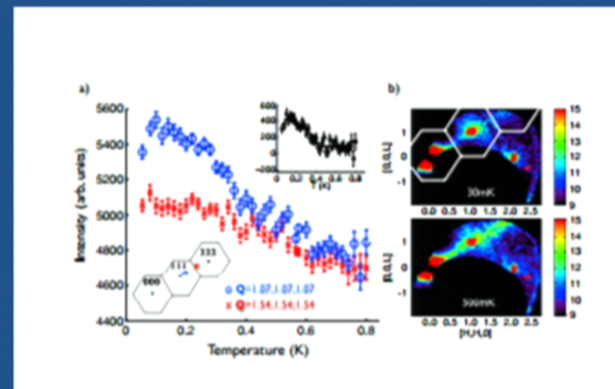

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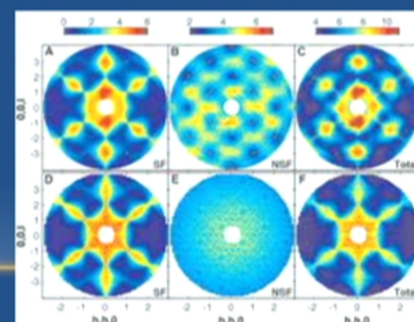
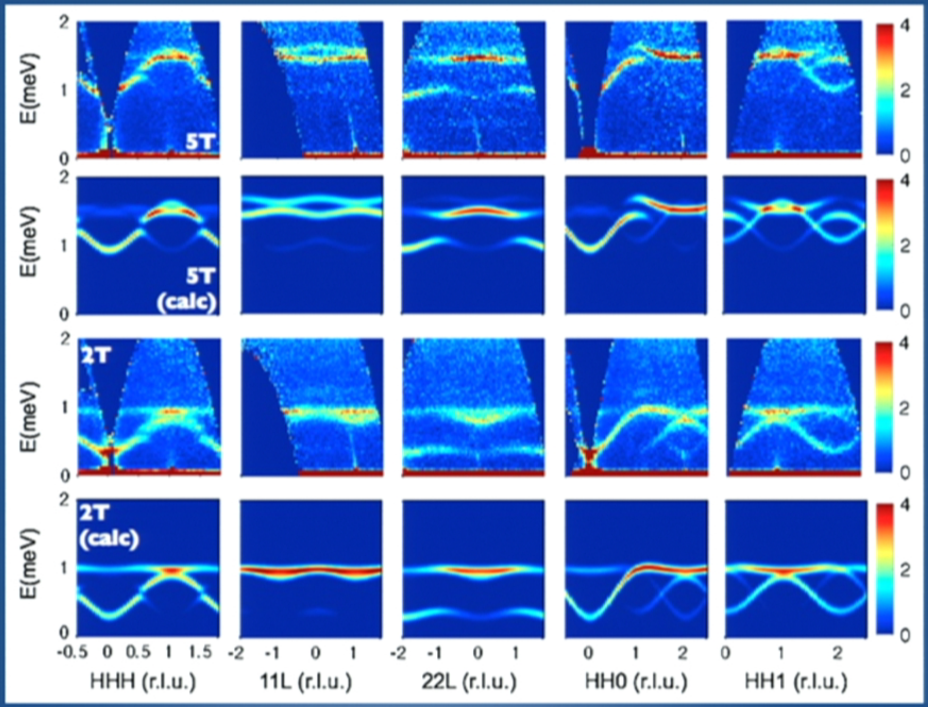


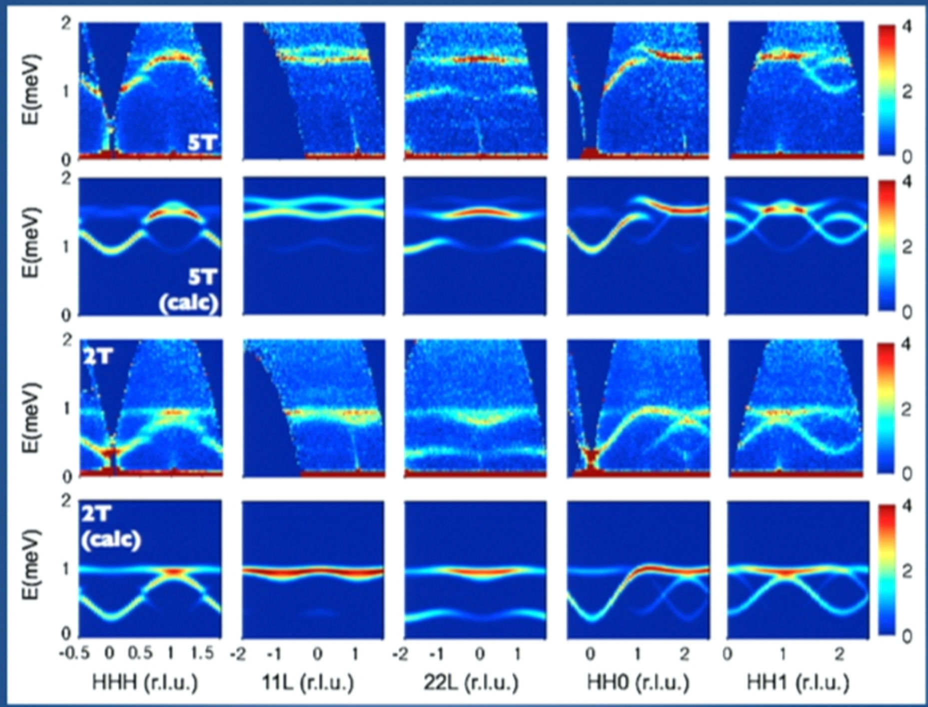
Ho2Ti2O7 Fennel et al Science

Determining Exchange Parameters



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

Determining Exchange Parameters



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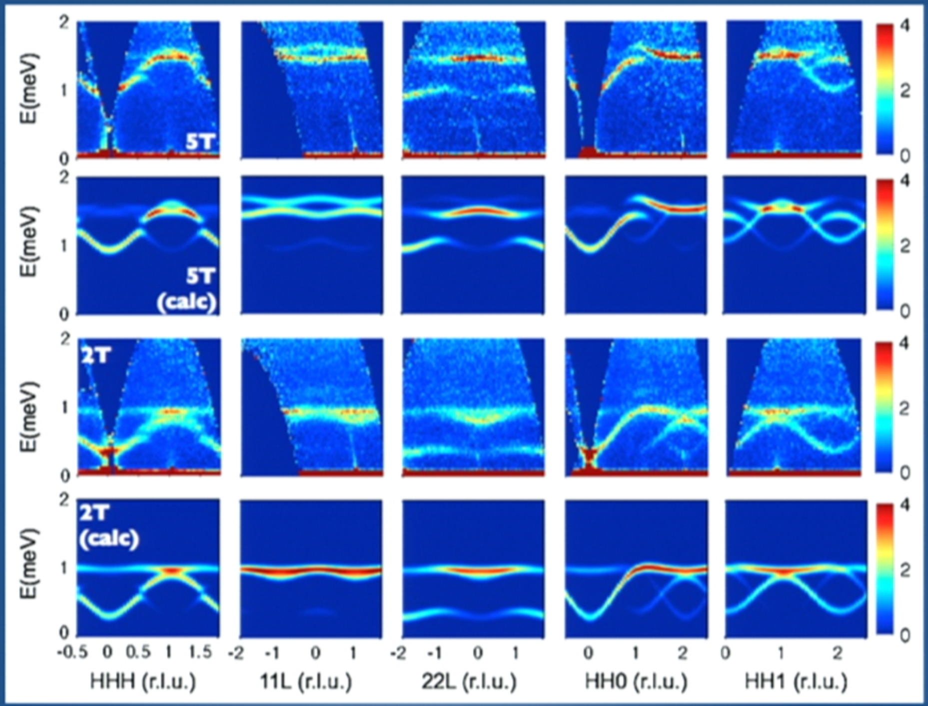

Ross et al PRX : High Field Spectra can be fit to SWT to determine exchange parameters
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Local Basis Convenient for spin-ice physics

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

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?
What is its zero-field ground state phase?
How do we calculate thermodynamic properties reliably?

HIGH TEMPERATURE EXPANSIONS

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

An extensive (intensive) property P:

$F, C_v, \chi(q), \ldots$

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

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

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

PBC introduces artifacts unless size large

(e.g. Pyrochlore Lattice 16-site cluster: Each site is its own 4^{th} neighbor 6 different ways)

Becomes Less useful with increased dimensionality and coordination number

NUMERICAL LINKED CLUSTER EXPANSION

Combines Linked Cluster + ED

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

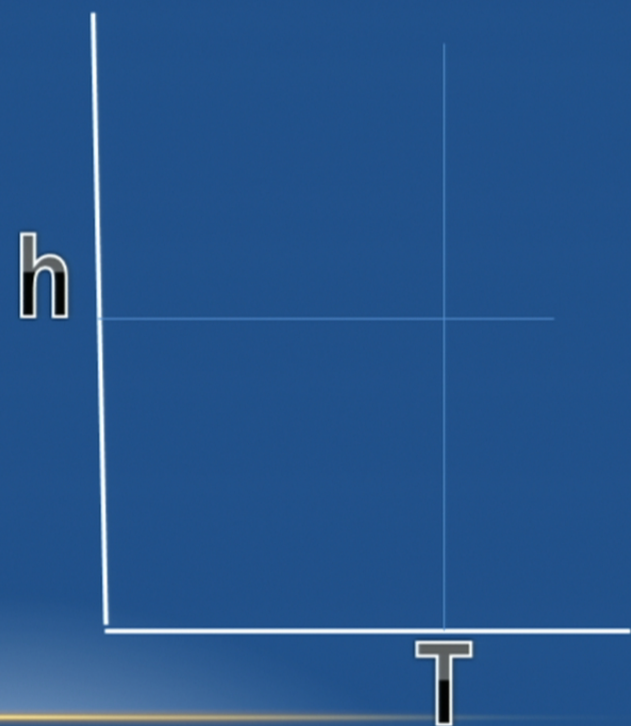
High Field Expansions:

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

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NLC TO 4^{TH} ORDER

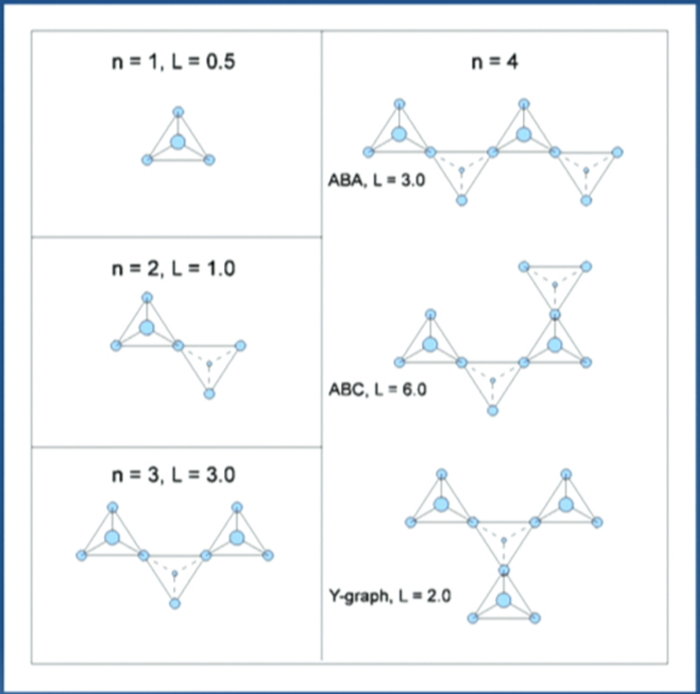
13 site clusters with no lattice symmetry
8192x8192 complex matrices

ED required 2-4 GB of memory

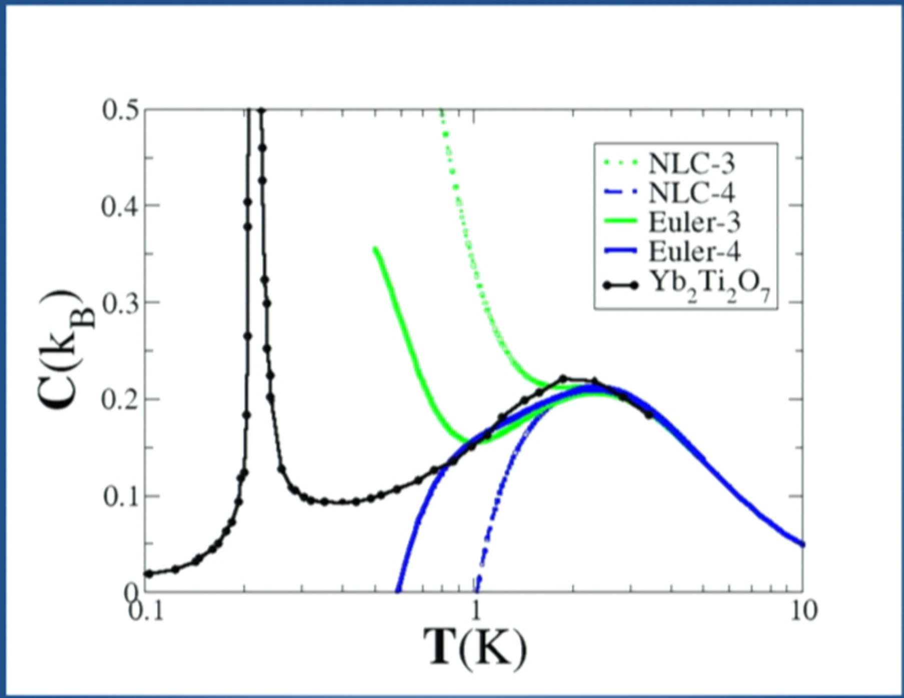
Next order: 16-site clusters memory goes up by factor of 64

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

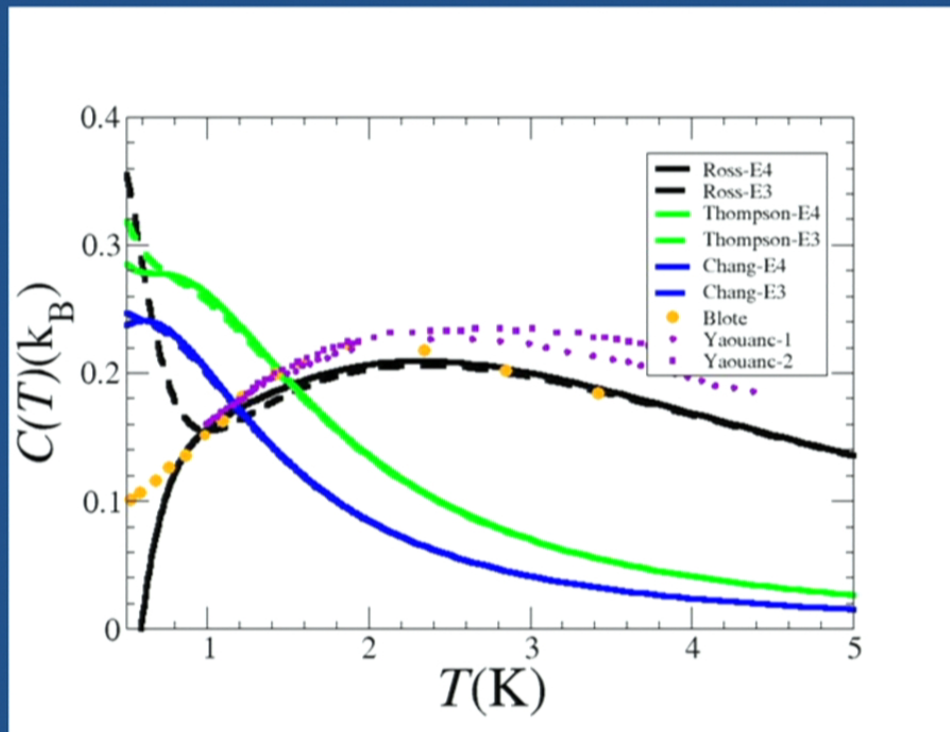
Missing: How to extrapolate for singular behavior and long-range correlations



Specific Heat: YbTO

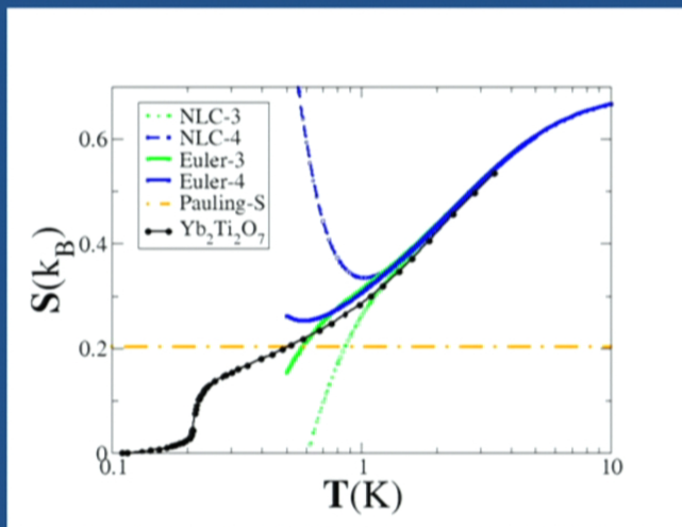


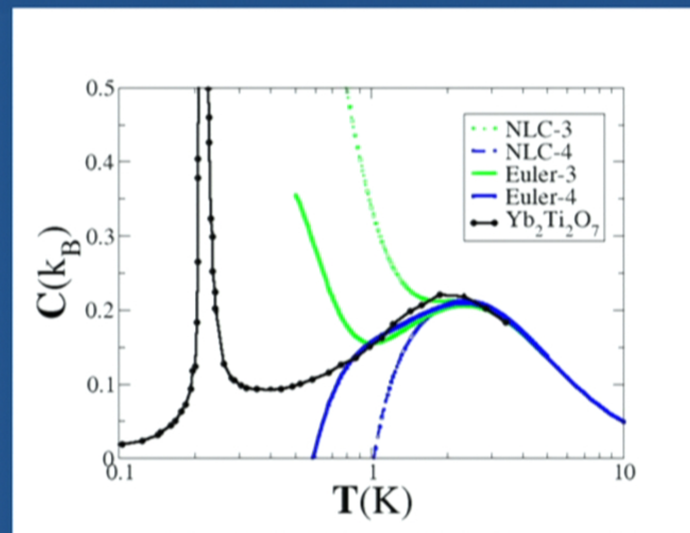
Different exchange parameters proposed for YbTO



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

Entropy and Heat Capacity of YbTO:
Regime between specific heat peaks Spin Ice?
No plateau but entropy close to Pauling value

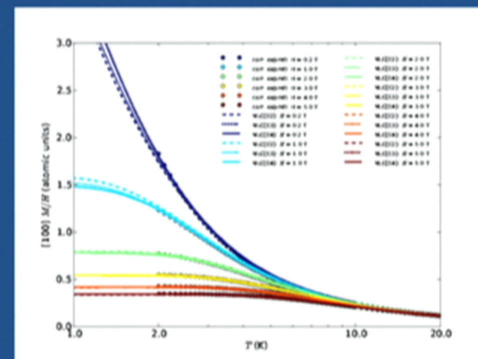


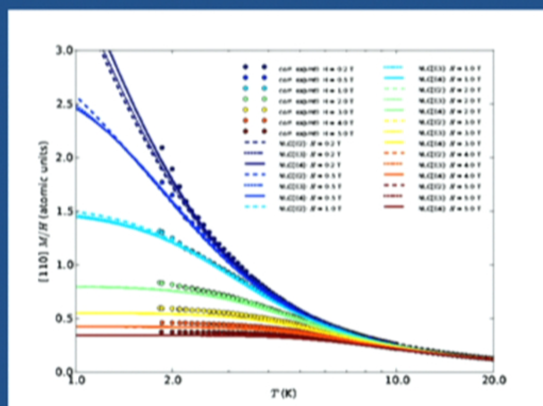


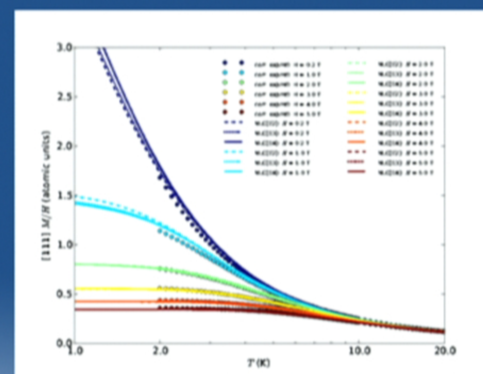
Theory: Start from k ln(2) entropy at T=infinity
Experiment: Start with zero entropy at T=100 mK
Agreement implies the model contains the phase transition at T=0.24 K

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

WHAT IS THE T=0.24 K TRANSITION? 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

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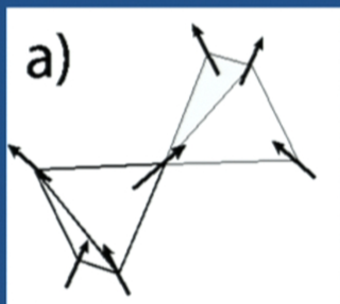
Perturbative selection with $J_{z\pm}$, $J_{\pm}$ and $J_{\pm\pm}$



$J_{\pm}$ 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.

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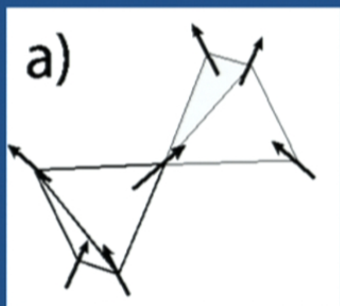

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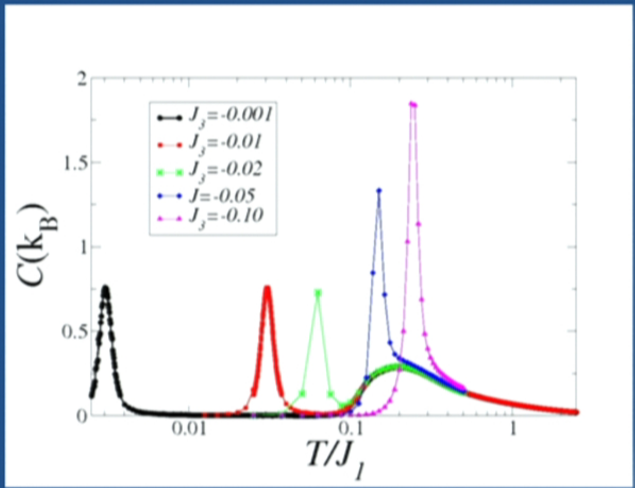
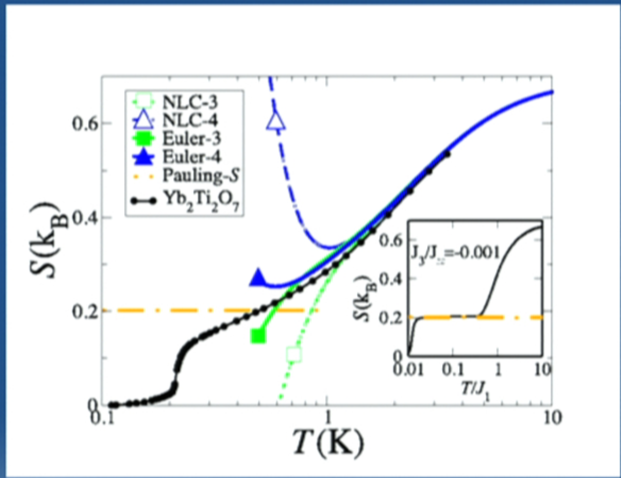
WHAT IS THE T=0.24 K TRANSITION?

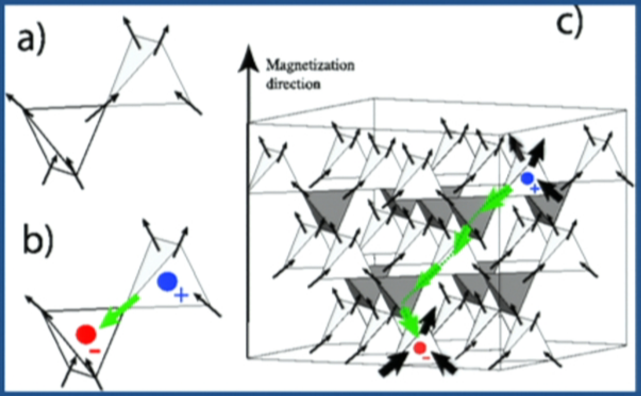
Low T peak in specific heat associated with q=0 FM order?





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

DYNAMICS REMAINS NON-TRIVIAL
SPINON-ANTISPINON PAIRS WITH STRINGS
SPINONS CAN HOP IN ORDER LAMBDA
STRING-LENGTH DIVERGES FOR SMALL LAMBDA



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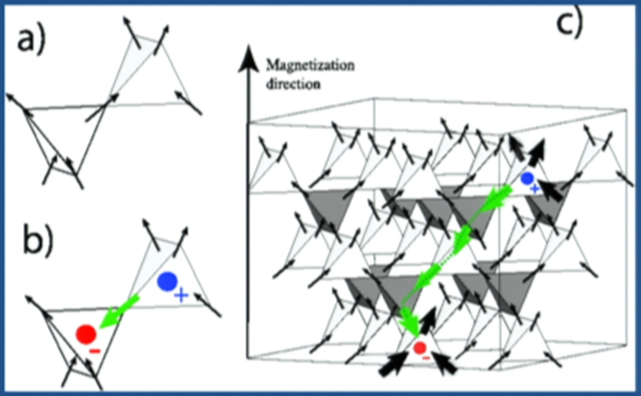
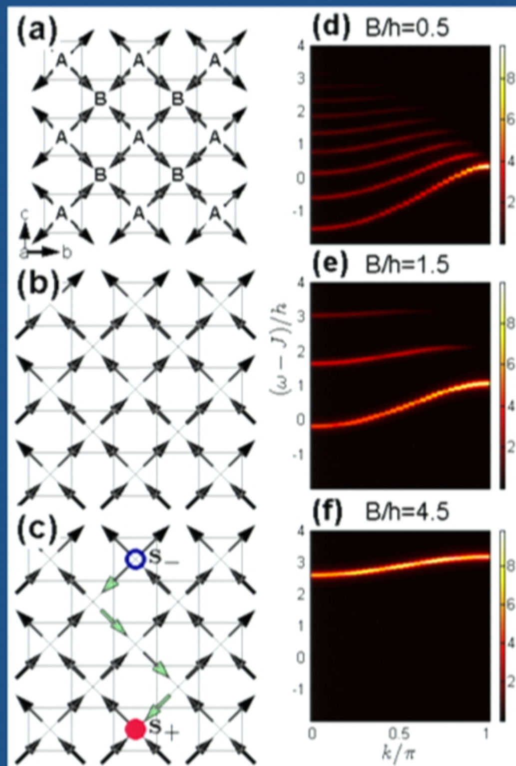
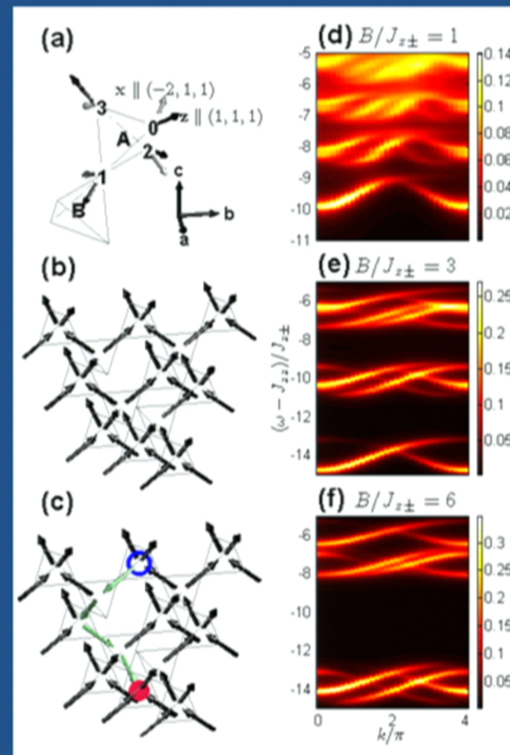


TCHERNYSHYOV: NEUTRONS SHOULD SEE MANY BRANCHES





Perturbative regime should share this physics
We are working on S(q,omega)
Experiments on YbTO?

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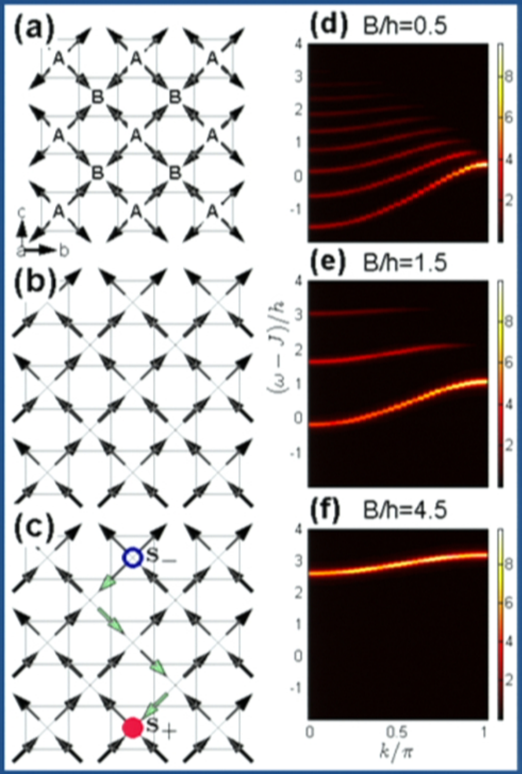

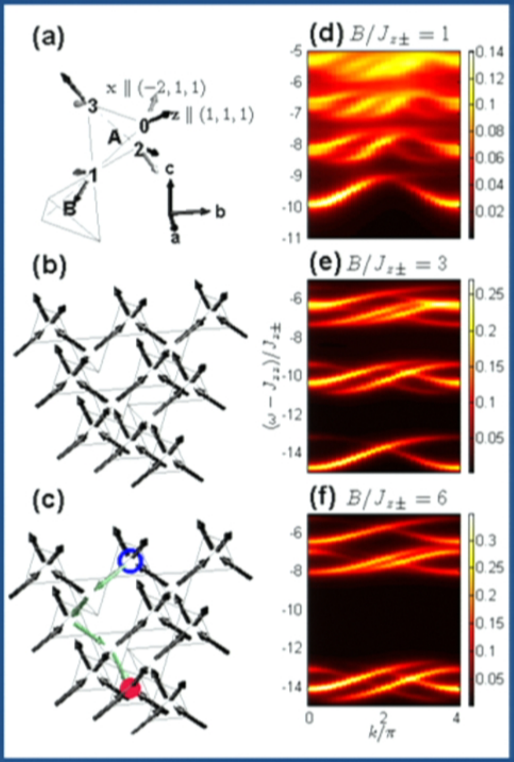

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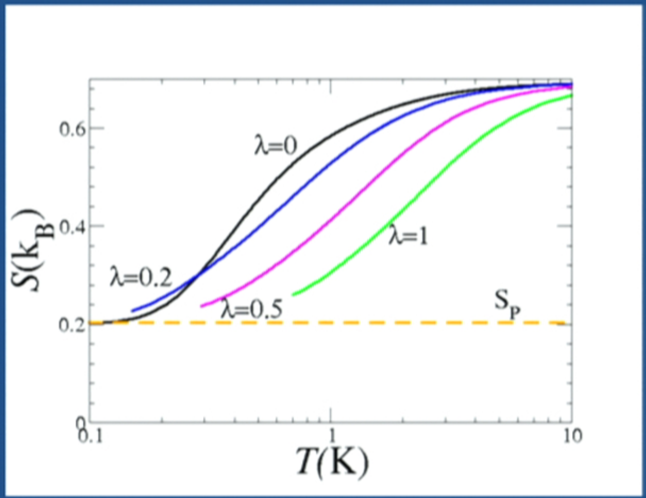
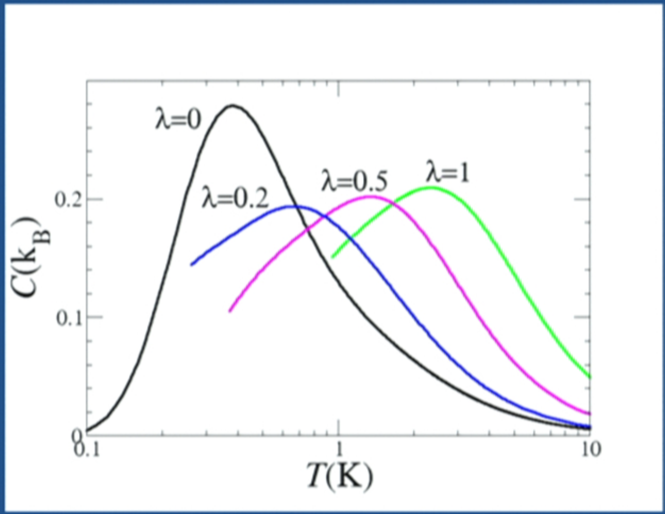
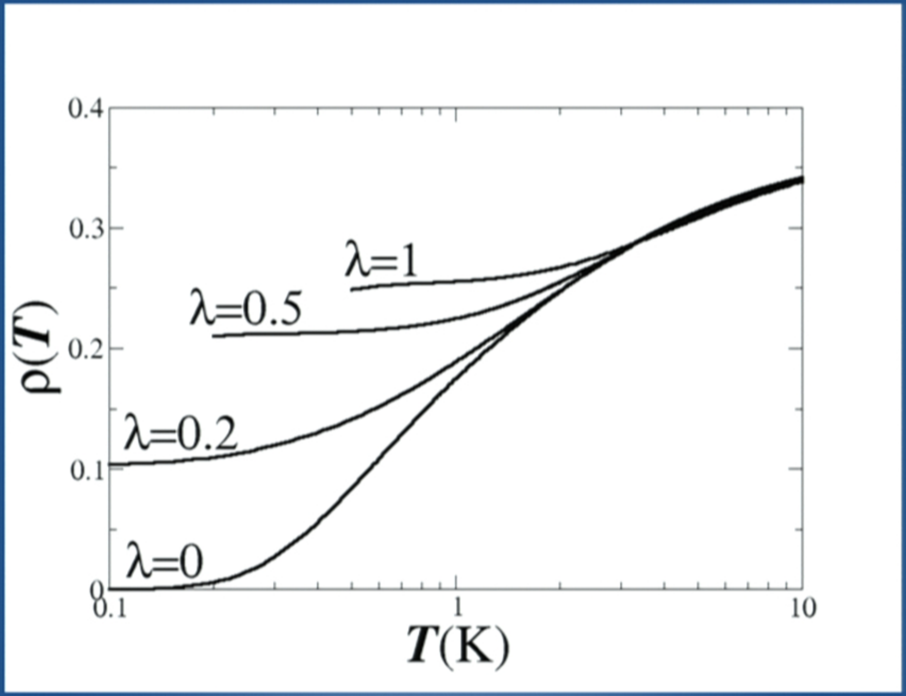
DOES THIS PICTURE CONTINUE TO LAMBDA=1?





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

DEFECT (MONOPOLE/SPINON) DENSITY (NLC)



Increasing lambda, intermediate regime is not simply classical Spin Ice
Strong Renrmalization of low energy physics

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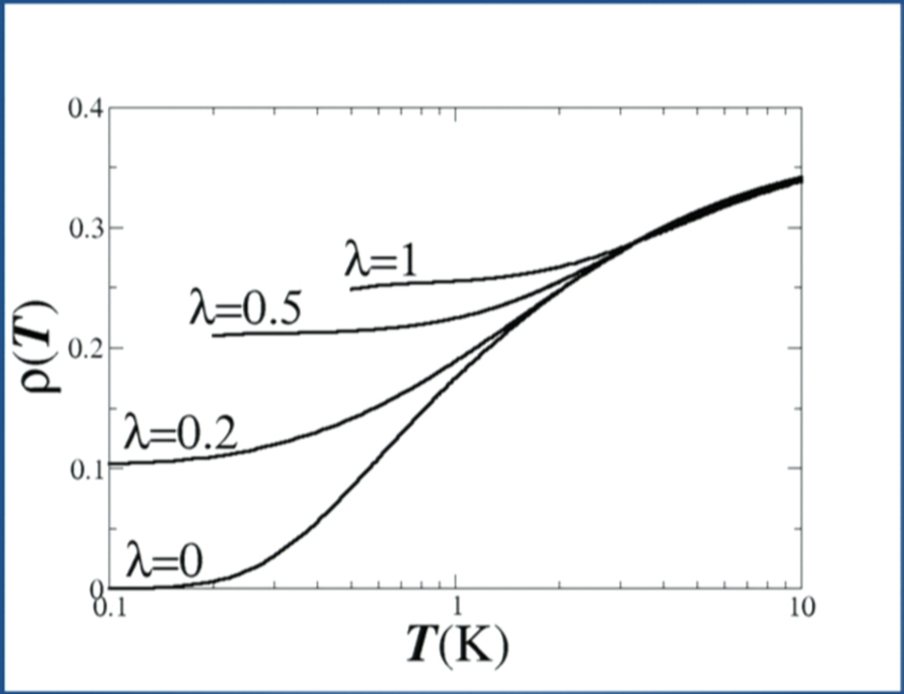

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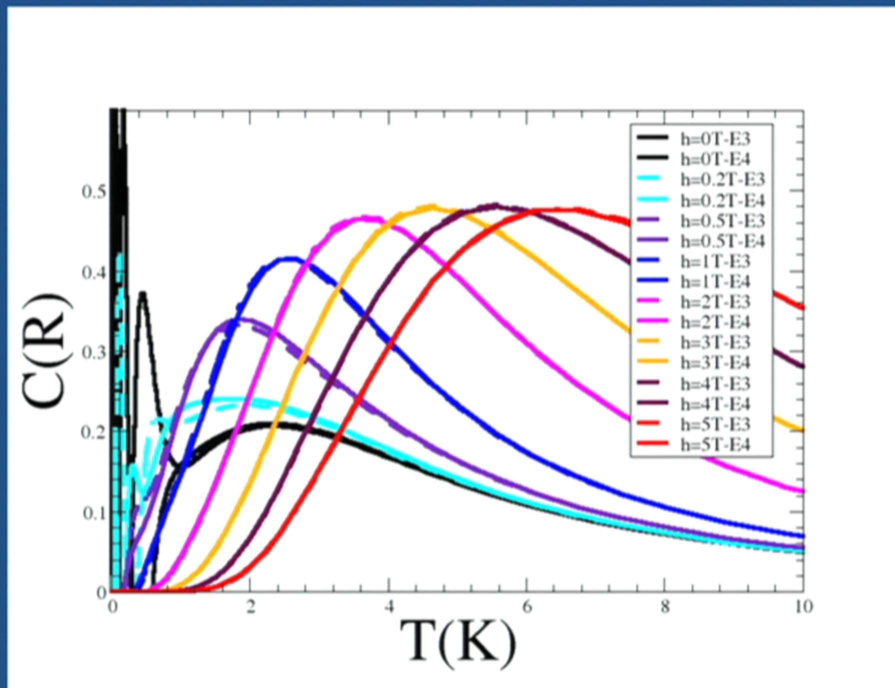
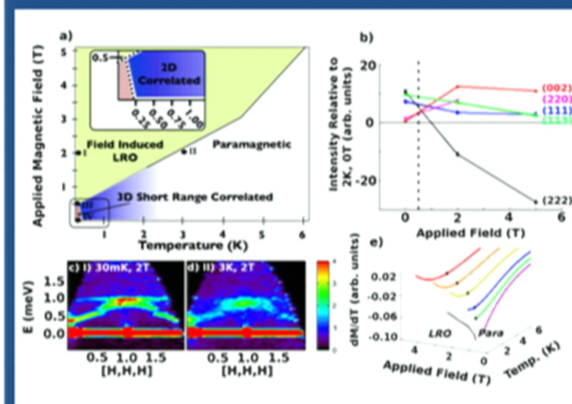
HEAT CAPACITY IN A FIELD [110]

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

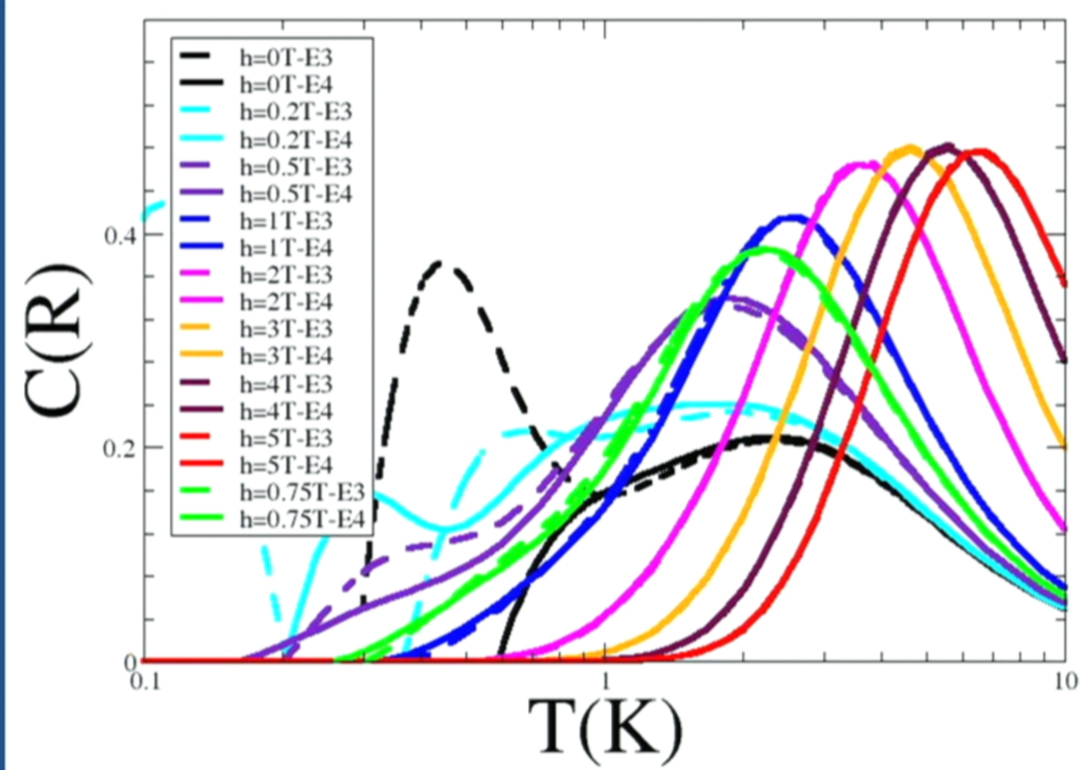




Ross et al PRL 103, 227202 (2009)

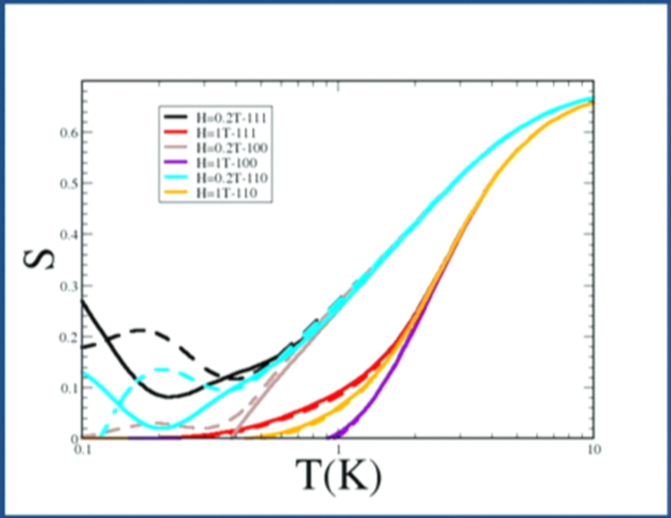
Peak tracks proposed Phase Diagram for the paramagnet/ferromagnet transition and onset of sharp excitations in high fields

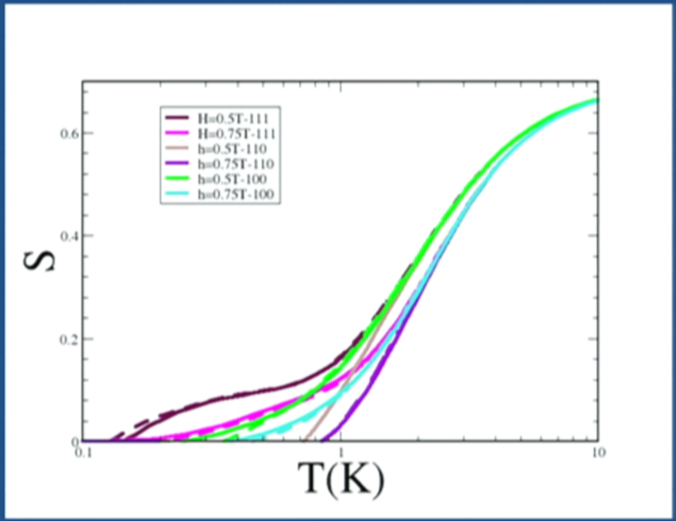
Peak splits in two near h=0.5 T (shoulder develops)



Temperature on a log scale

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 other directions.
Very suggestive of degeneracies in MFT (Phase Transition in a field except 100)
Needs more experimental study.

Novel Gauge Mean Field Theory (Savary and Balents 2012)

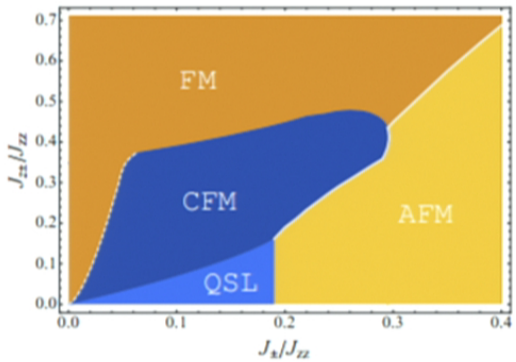
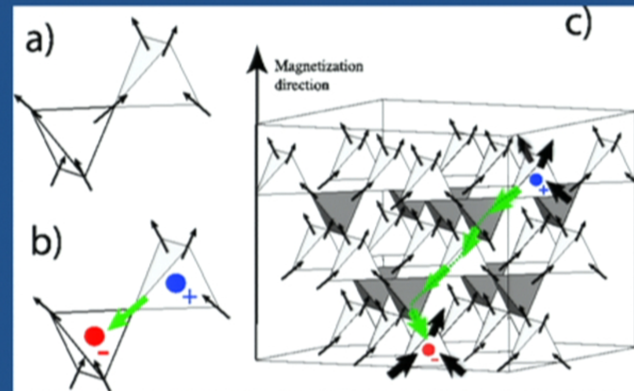
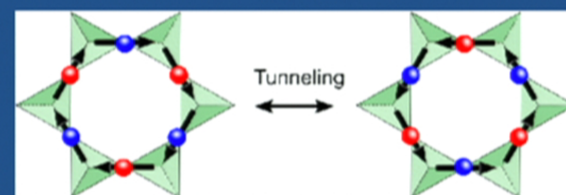


FIG. 3. Modified gMFT diagram, which takes into account the known perturbative limit $J_{z\pm}/J_{zz} \ll 1$. Note that the FM-CFM transition (white dashed line) in the latter region is a *sketch*.





Coulombic Ferromagnet and QSL Phases are both in the Phase Diagram
Jpm terms can lead to ring terms and cause string tension to become negative
(Can be studied by series expansions)
For non-Kramers Ions Jzpm=0

Summary for YbTO

- YbTO is rather well described by a nearest-neighbor anisotropic exchange QSI model
- 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**
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Conclusions and Future Directions

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