

Coulombic Quantum Liquids in Spin-1/2 Pyrochlores

Lucile Savary



Collaborators



$\text{Yb}_2\text{Ti}_2\text{O}_7$ project



Leon Balents
(KITP)



Kate Ross



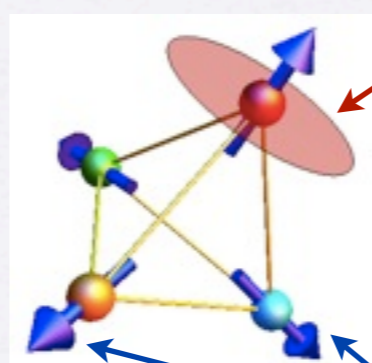
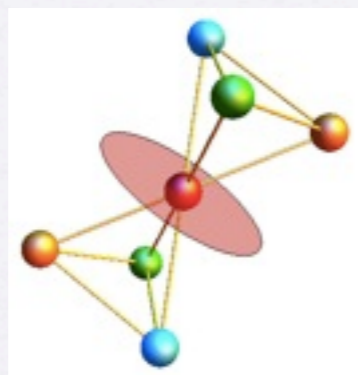
Bruce Gaulin

(experiments, McMaster)

Special thanks to Benjamin Canals and Peter Holdsworth.

Reminder

- the pyrochlore lattice



local XY-plane

local z-axes

- spin ice (cf. Michel's talk)

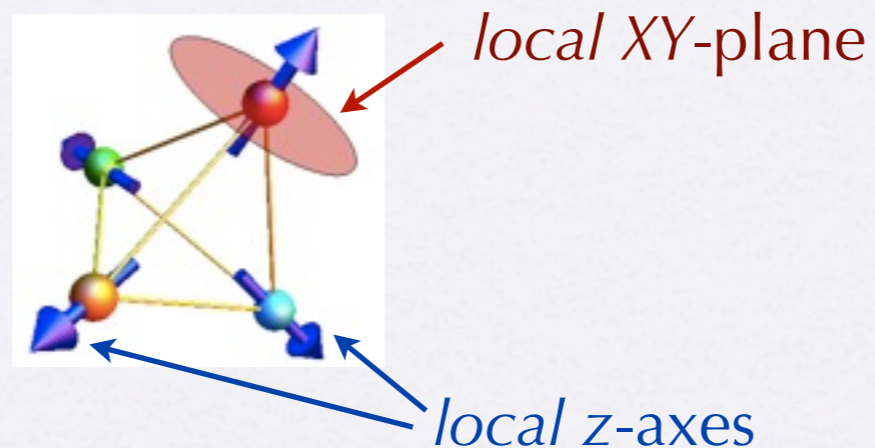
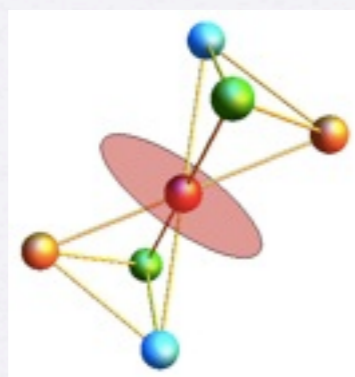
$$H_{\text{SI}} = J_{zz} \sum_{\langle i,j \rangle} S_i^z S_j^z \quad J_{zz} > 0$$

in local bases
↓

$$H_{\text{SI}} \sim \frac{J_{zz}}{2} \underbrace{(S_0^z + S_1^z + S_2^z + S_3^z)^2}_{= 0}$$

Reminder

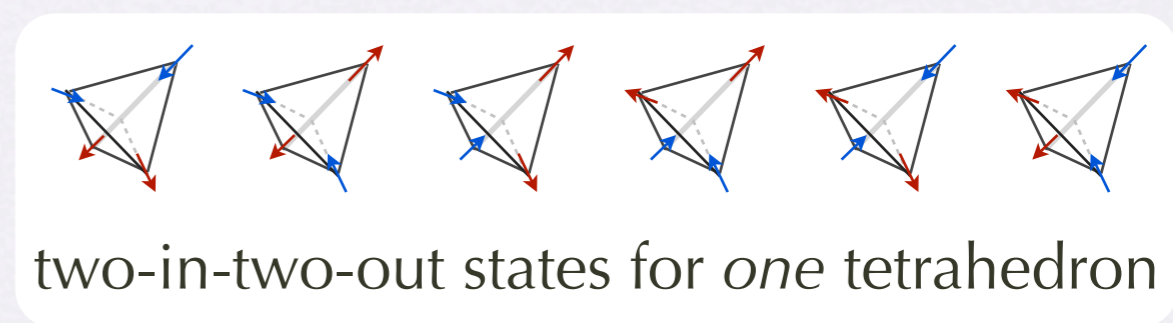
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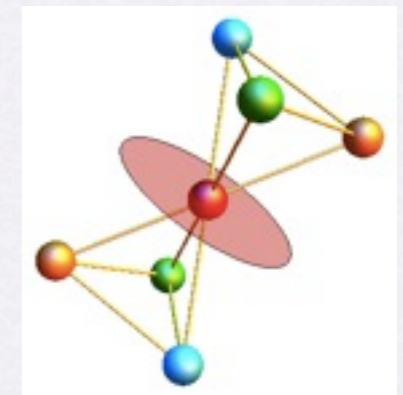
extensive degeneracy \longrightarrow classical spin liquid with monopole excitations

Rare-earth pyrochlores



- grown rare-earth pyrochlores: $\text{Ho}_2\text{Ti}_2\text{O}_7$, $\text{Dy}_2\text{Ti}_2\text{O}_7$, $\text{Ho}_2\text{Sn}_2\text{O}_7$, $\text{Dy}_2\text{Sn}_2\text{O}_7$, $\text{Er}_2\text{Ti}_2\text{O}_7$, $\text{Yb}_2\text{Ti}_2\text{O}_7$, $\text{Tb}_2\text{Ti}_2\text{O}_7$, $\text{Er}_2\text{Sn}_2\text{O}_7$, $\text{Tb}_2\text{Sn}_2\text{O}_7$, $\text{Pr}_2\text{Sn}_2\text{O}_7$, $\text{Nd}_2\text{Sn}_2\text{O}_7$, $\text{Gd}_2\text{Sn}_2\text{O}_7$, ...
- grown rare-earth B-site spinels: CdEr_2S_4 , CdEr_2Se_4 , CdYb_2S_4 , CdYb_2Se_4 , MgYb_2S_4 , MgYb_2S_4 , MnYb_2S_4 , MnYb_2Se_4 , FeYb_2S_4 , CdTm_2S_4 , CdHo_2S_4 , FeLu_2S_4 , MnLu_2S_4 , MnLu_2Se_4 , ...

lots of room for diverse behaviors!



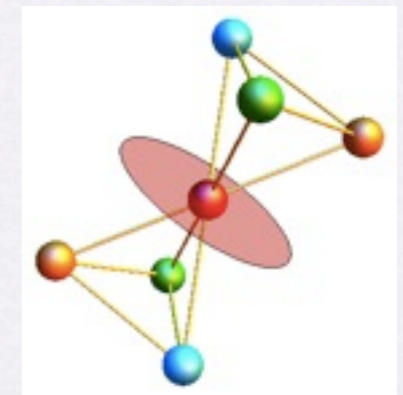
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spin ices

- grown rare-earth B-site spinels: CdEr_2S_4 , CdEr_2Se_4 , CdYb_2S_4 , CdYb_2Se_4 , MgYb_2S_4 , MgYb_2S_4 , MnYb_2S_4 , MnYb_2Se_4 , FeYb_2S_4 , CdTm_2S_4 , CdHo_2S_4 , FeLu_2S_4 , MnLu_2S_4 , MnLu_2Se_4 , ...



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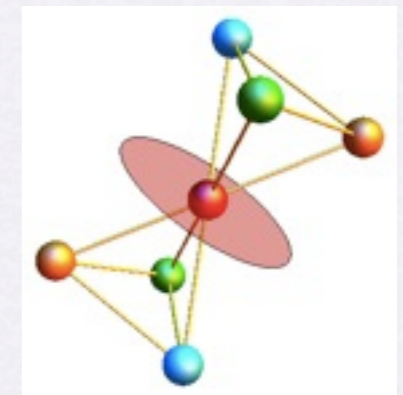


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quantum AFM,
order by disorder

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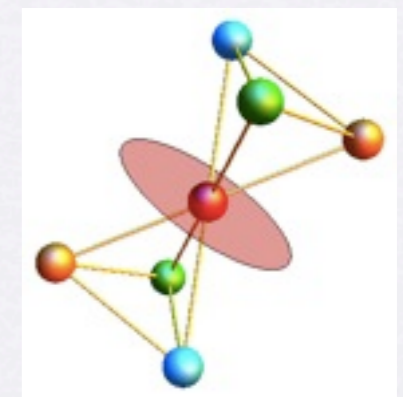
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quantum spin liquids ?

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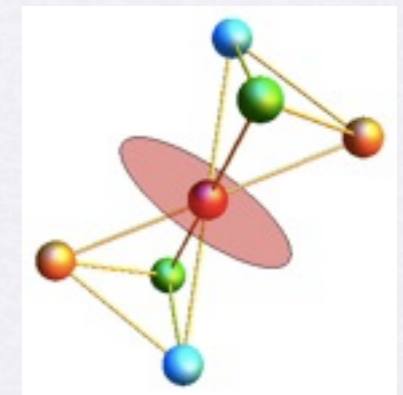
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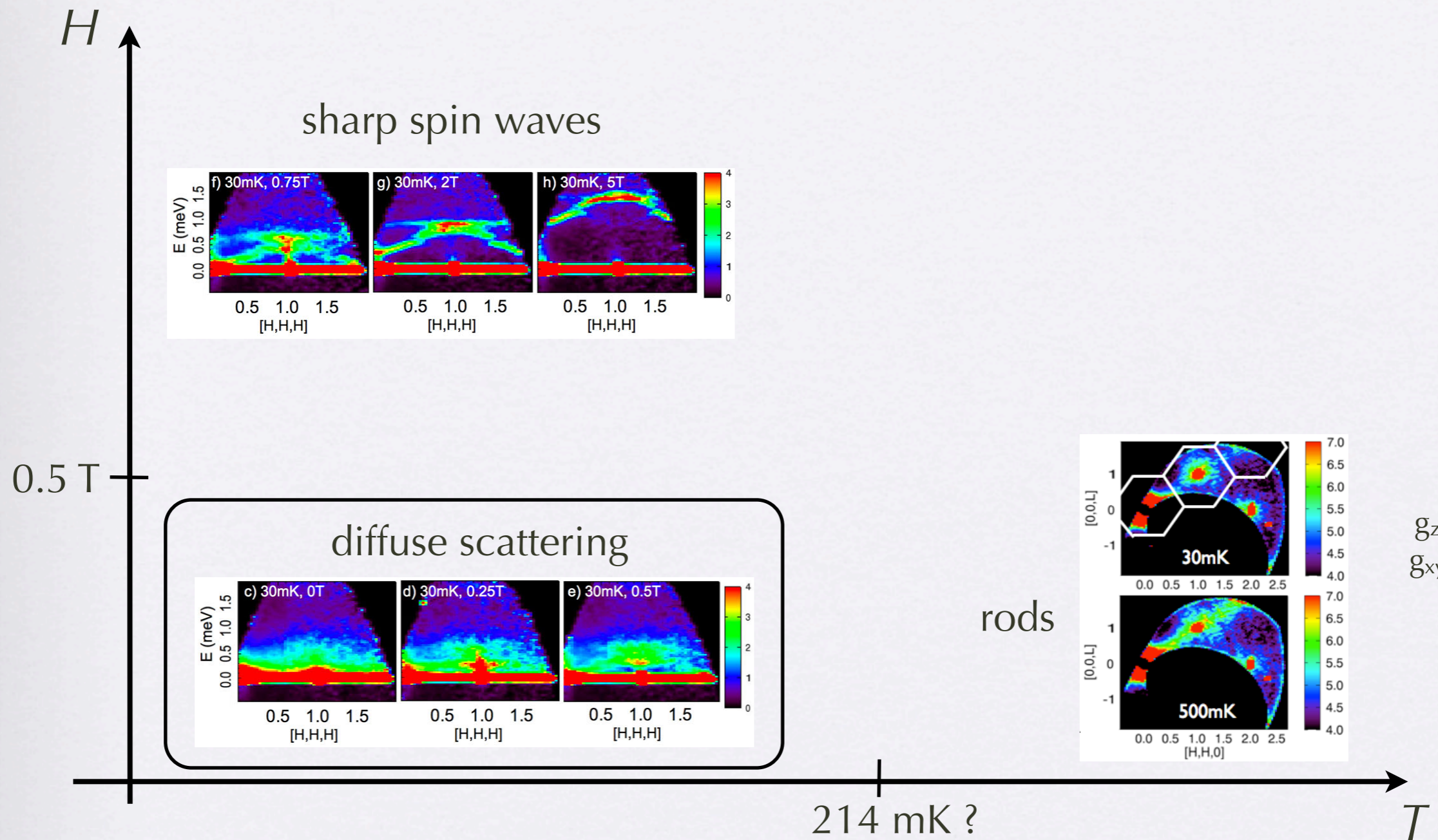
spin ice ?

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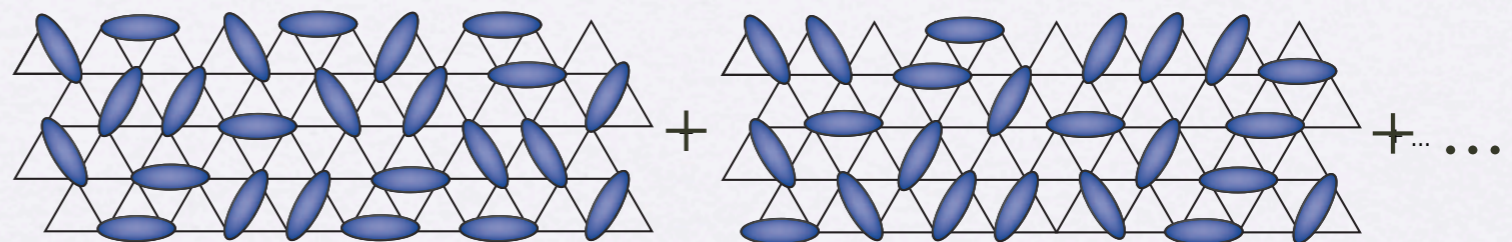
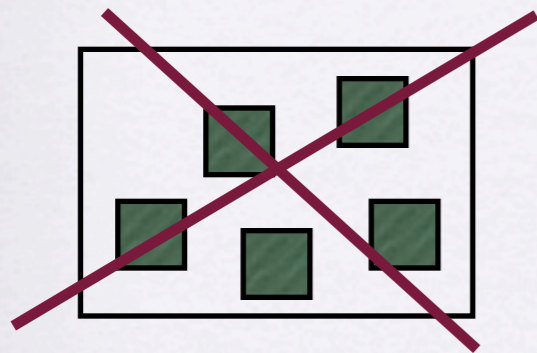
Yb₂Ti₂O₇: puzzling experimental features



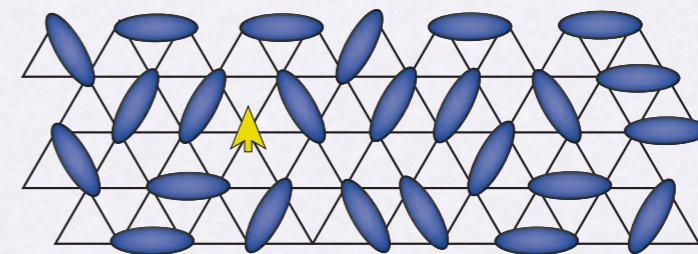
quantum spin liquid regime?

What Is a Quantum Spin Liquid (QSL)?

- history: $\langle \mathbf{S} \rangle = \mathbf{0}$ - too restrictive
- **long-range entangled**: a state which cannot be written or approximated as a product of any finite blocks



- consequence: **exotic properties**, e.g. **fractional particles**



note: $\{\langle \mathbf{S} \rangle = \mathbf{0} \text{ \& \ exotic properties}\}$ requires long-range entanglement

What Is a Quantum Spin Liquid?

- first prediction: Anderson 1973
- would be *really* nice to find one in nature!
- good place to look: frustrated magnets
 - organics (triangular lattice), J_1 - J_2 models, pyrochlores...

Outline

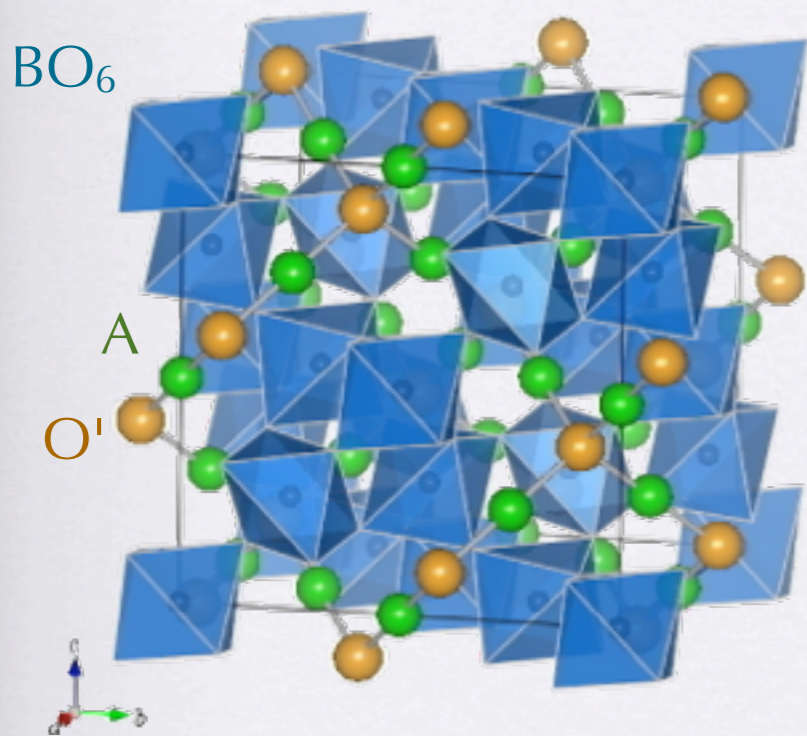
- method
- results
- experimental signatures
- materials

Symmetries of the Hamiltonian

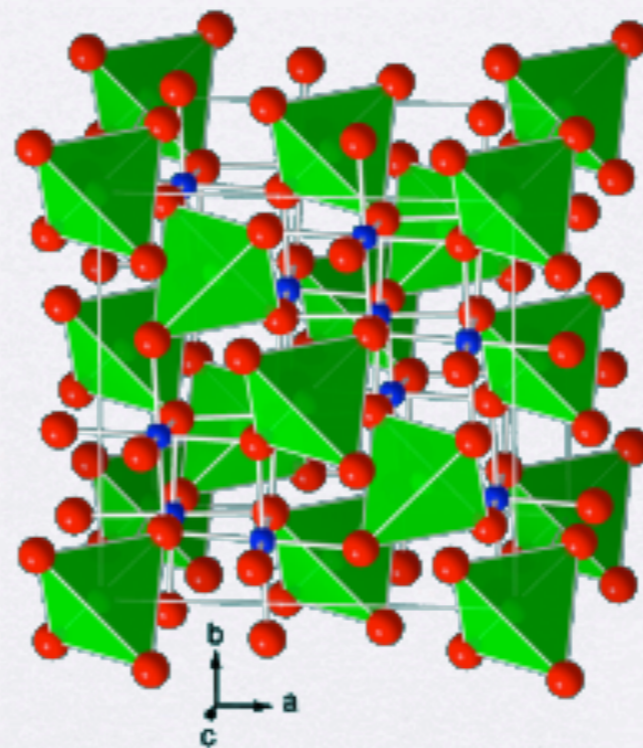
rare-earths : intrinsic strong spin-orbit coupling

→ *discrete cubic symmetries only*

space group: $Fd-3m$, i.e. #227 :



$A_2B_2O_7$
"pyrochlore oxides"



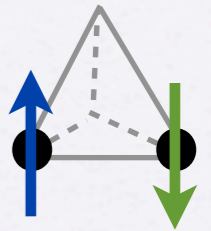
AB_2X_4
spinel

strong crystal fields

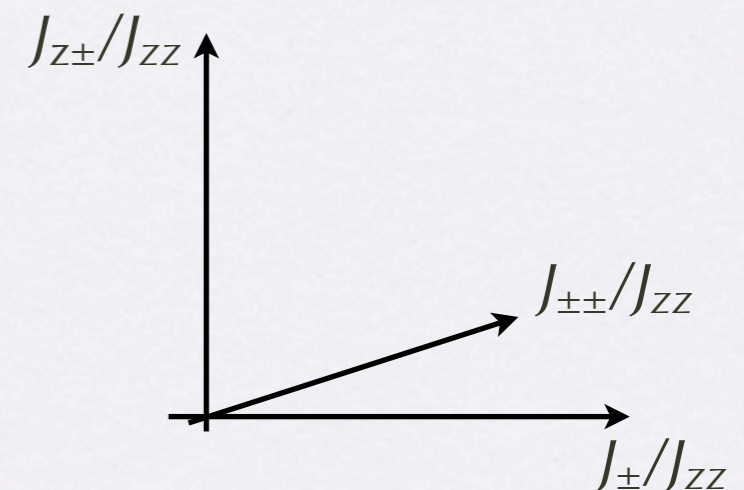
A
B
X

General NN exchange Hamiltonian for effective spins 1/2

$$\begin{aligned}
 H = \sum_{\langle ij \rangle} & \left[J_{zz} S_i^z S_j^z \right. \\
 & - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \\
 & + J_{z\pm} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_j^-) + i \leftrightarrow j] \\
 & \left. + J_{\pm\pm} [\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-] \right]
 \end{aligned}$$



to each material corresponds a set of J 's

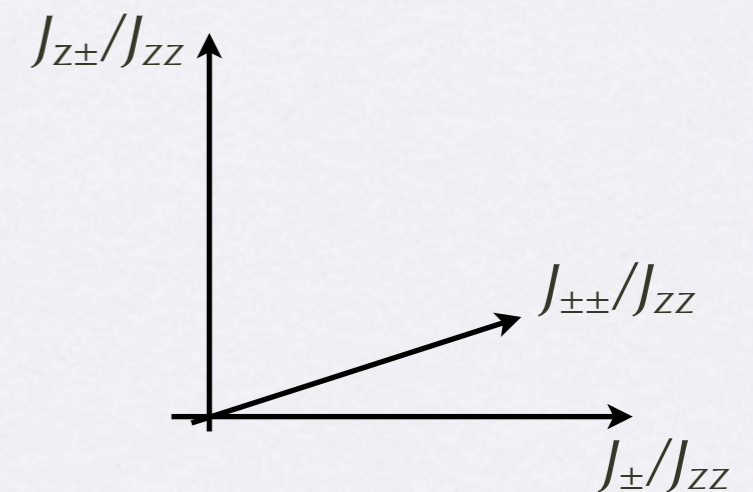
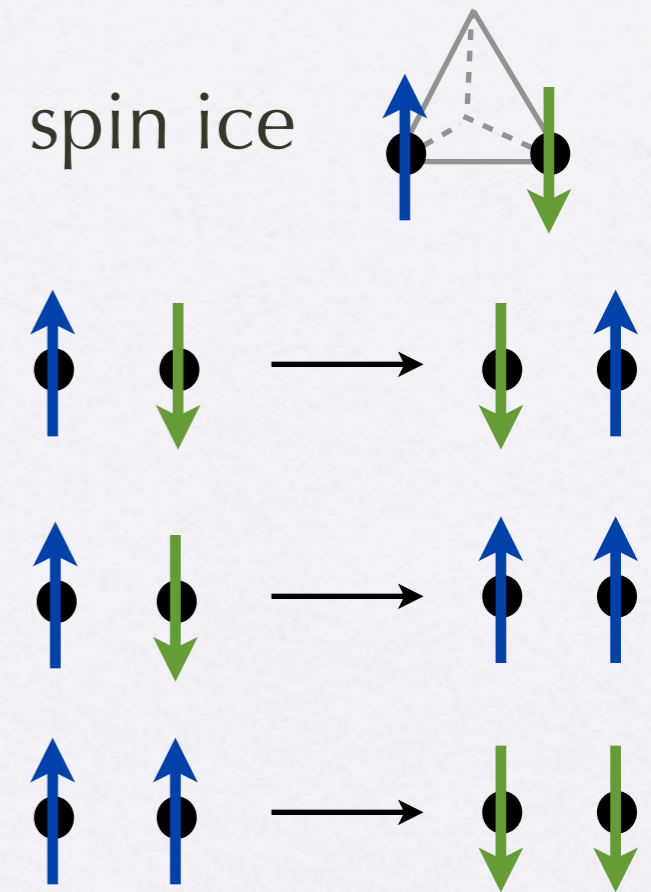


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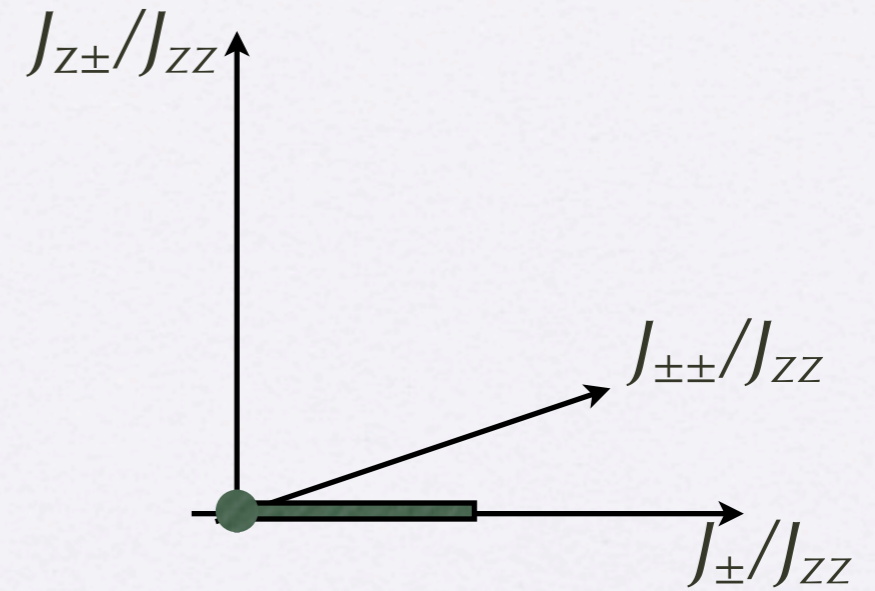
to each material corresponds a set of J 's

What is the phase diagram ?
 Are there any exotic phases there ?

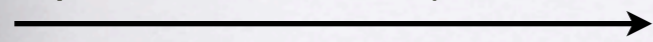


The Hermele *et al.* QSL

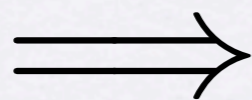
$$H = \sum_{\langle ij \rangle} \left[J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \right]$$



perturbation theory in J_{\pm}/J_{zz}



quantum electrodynamics $H \sim H_{\text{QED}} \sim E^2 + B^2$



photon (gapless and linear)
particle-hole excitations (gapped)

Relation to classical spin ice

classical spin ice

U(1) quantum spin liquid

thermal spin liquid

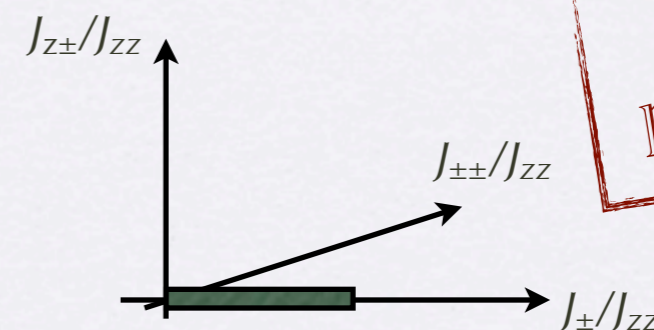
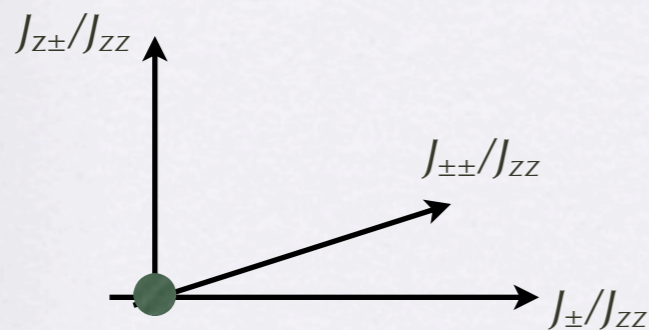
quantum spin liquid

extensively many degenerate ground states

one entangled ground state (= vacuum)

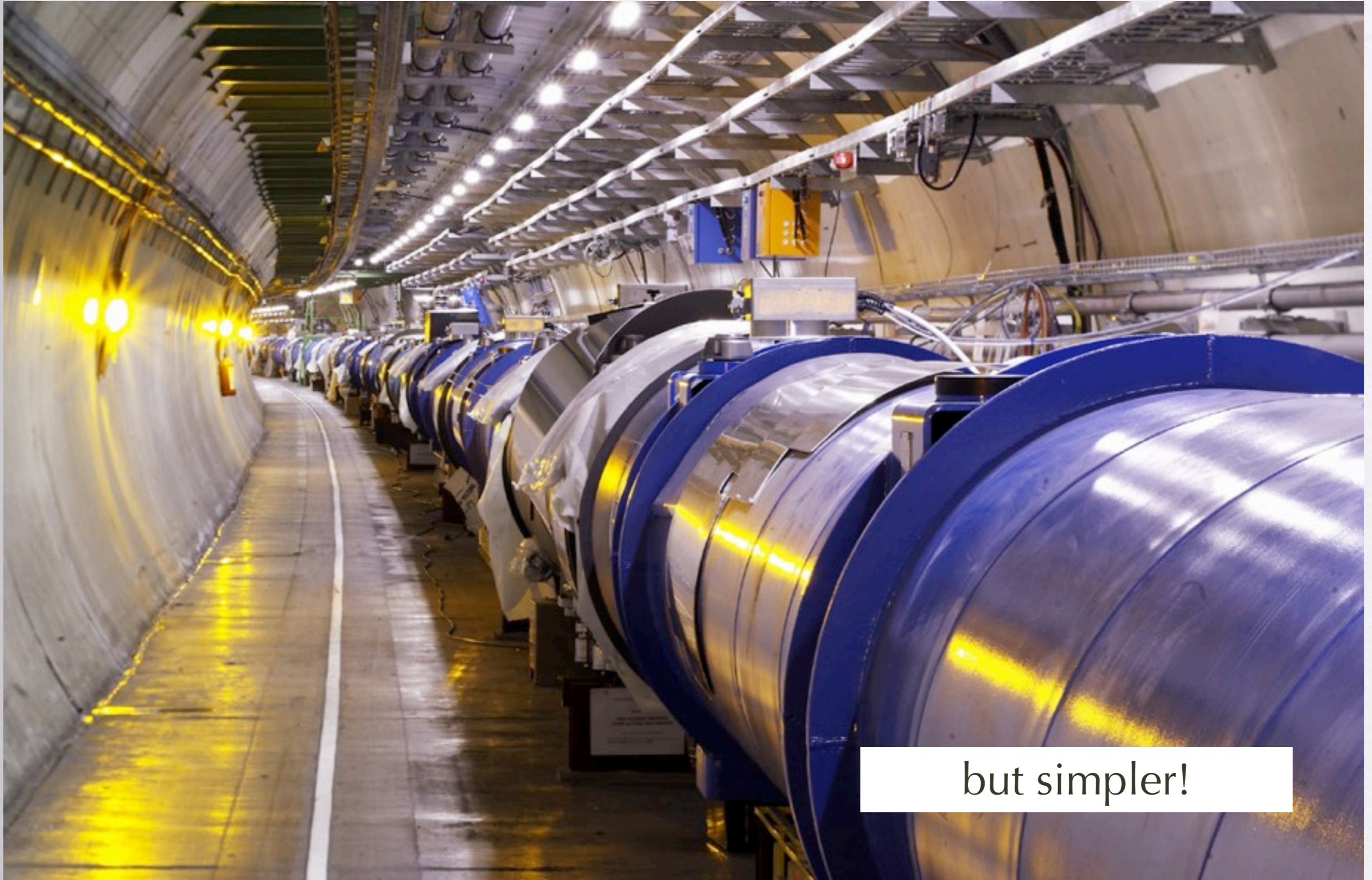
magnetic monopoles = spinons

spinons
"electric" monopoles
gapless photon



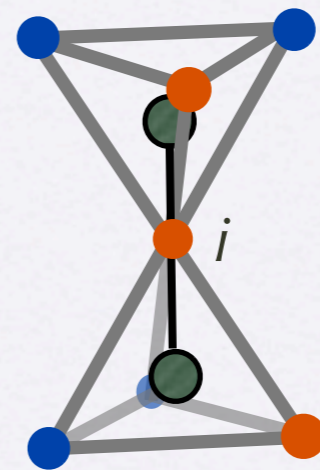
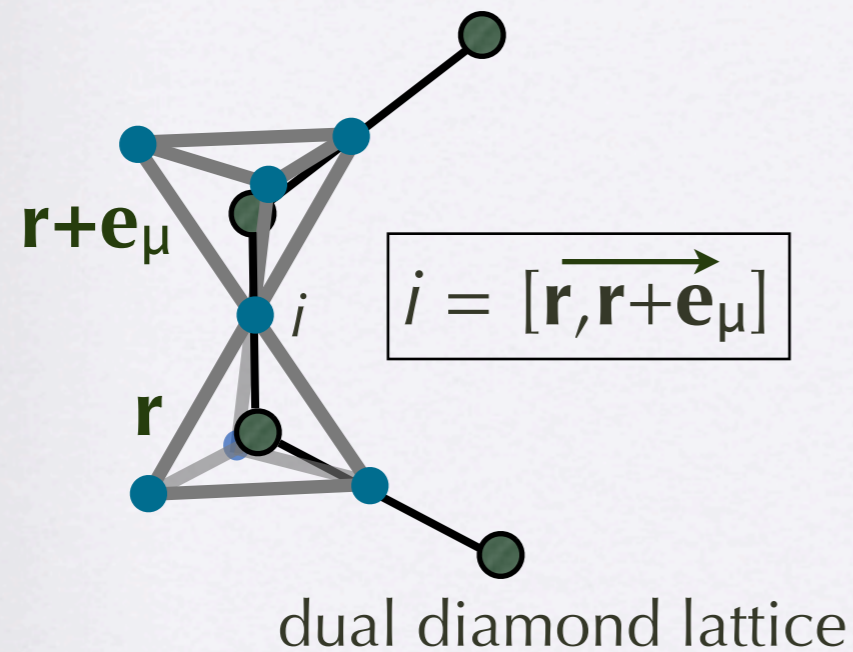
stable to all perturbations

How we do this: compact abelian lattice Higgs theory

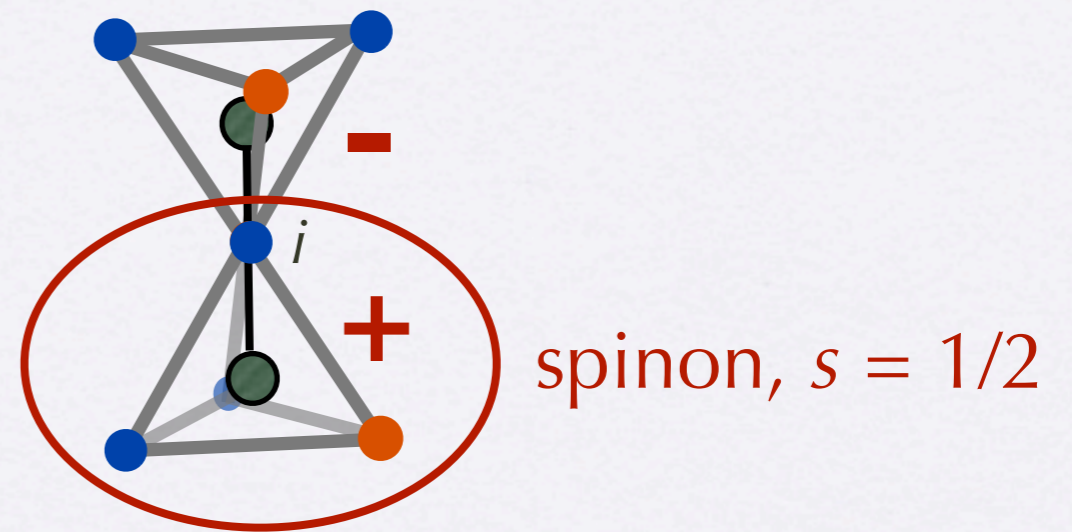
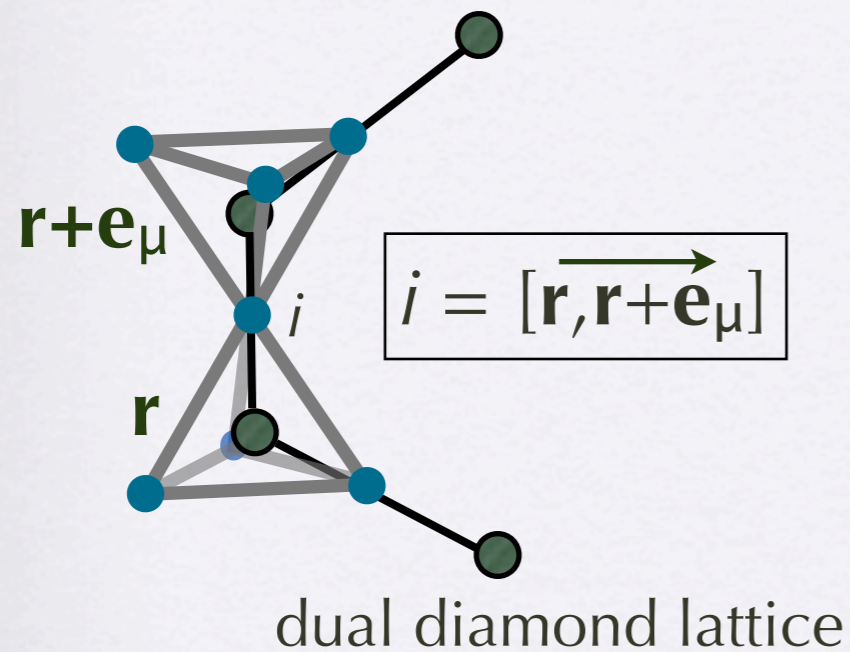


but simpler!

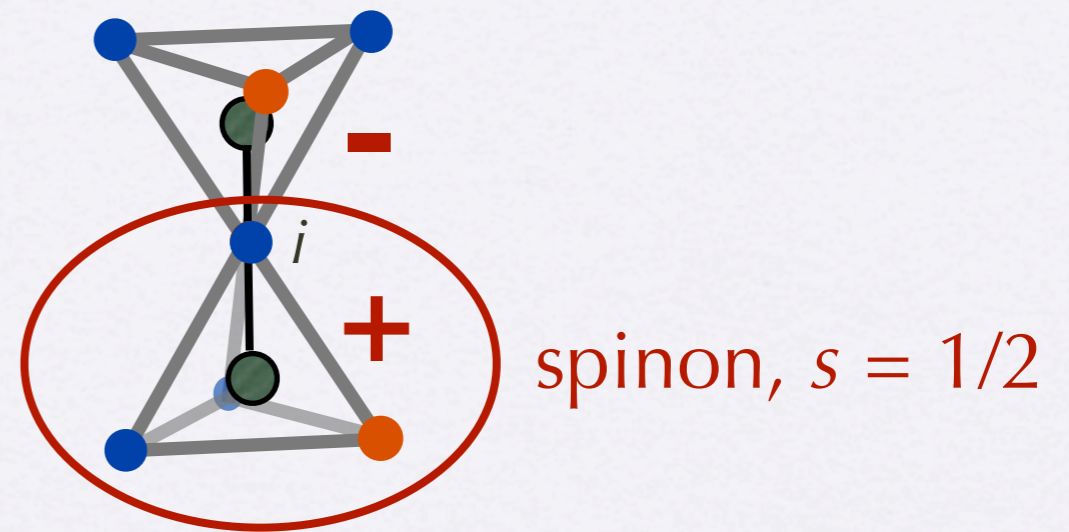
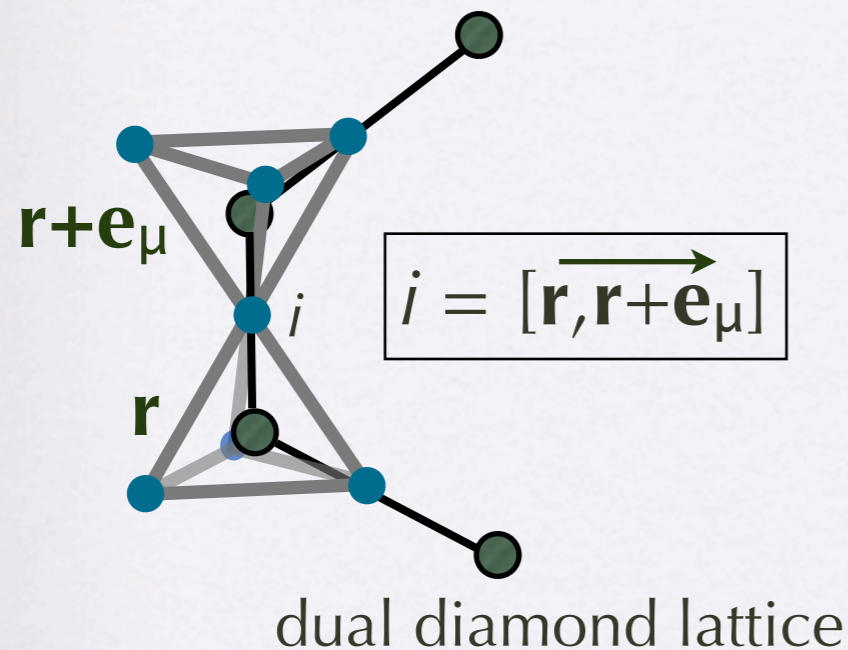
How we do this: compact abelian lattice Higgs theory



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How we do this: compact abelian lattice Higgs theory



$$S_{\mathbf{r}, \mathbf{r} + \mathbf{e}_\mu}^+ = \Phi_{\mathbf{r}}^\dagger s_{\mathbf{r}, \mathbf{r} + \mathbf{e}_\mu}^+ \Phi_{\mathbf{r} + \mathbf{e}_\mu}$$

$$S_{\mathbf{r}, \mathbf{r} + \mathbf{e}_\mu}^z = s_{\mathbf{r}, \mathbf{r} + \mathbf{e}_\mu}^z$$

$$\begin{cases} \Phi_{\mathbf{r}} \rightarrow \Phi_{\mathbf{r}} e^{-i\chi_{\mathbf{r}}} \\ s_{\mathbf{r}\mathbf{r}'}^\pm \rightarrow s_{\mathbf{r}\mathbf{r}'}^\pm e^{\pm i(\chi_{\mathbf{r}'} - \chi_{\mathbf{r}})} \end{cases}$$

U(1) gauge symmetry

the slave particles have a simple interpretation

How we do this: compact abelian Higgs U(1) lattice gauge theory

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$$S_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu}^z = S_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu}^z$$

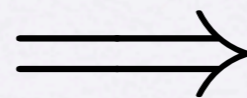
$$|\Phi_{\mathbf{r}}| = 1$$

$$Q_{\mathbf{r}} = \pm \sum_{\mu} S_{\mathbf{r},\mathbf{r}\pm\mathbf{e}_\mu}^z$$

$$S_{\mathbf{r}\mathbf{r}'}^z = E_{\mathbf{r}\mathbf{r}'}$$

$$S_{\mathbf{r}\mathbf{r}'}^\pm = e^{\pm i A_{\mathbf{r}\mathbf{r}'}}$$

our spinons are bosons



they can condense

$\langle \Phi \rangle$	phase
$\neq 0$	conventional
$= 0$	exotic

How we do this: compact abelian Higgs U(1) lattice gauge theory

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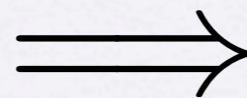
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$$H = \sum_{\mathbf{r} \in \text{I,II}} \frac{J_{zz}}{2} Q_{\mathbf{r}}^2 - J_{\pm} \left\{ \sum_{\mathbf{r} \in \text{I}} \sum_{\mu, \nu \neq \mu} \Phi_{\mathbf{r}+\mathbf{e}_\mu}^\dagger \Phi_{\mathbf{r}+\mathbf{e}_\nu} S_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu}^- S_{\mathbf{r},\mathbf{r}+\mathbf{e}_\nu}^+ + \sum_{\mathbf{r} \in \text{II}} \sum_{\mu, \nu \neq \mu} \Phi_{\mathbf{r}-\mathbf{e}_\mu}^\dagger \Phi_{\mathbf{r}-\mathbf{e}_\nu} S_{\mathbf{r},\mathbf{r}-\mathbf{e}_\mu}^+ S_{\mathbf{r},\mathbf{r}-\mathbf{e}_\nu}^- \right\}$$

$$- J_{z\pm} \left\{ \sum_{\mathbf{r} \in \text{I}} \sum_{\mu, \nu \neq \mu} \left(\gamma_{\mu\nu}^* \Phi_{\mathbf{r}}^\dagger \Phi_{\mathbf{r}+\mathbf{e}_\nu} S_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu}^z S_{\mathbf{r},\mathbf{r}+\mathbf{e}_\nu}^+ + \text{h.c.} \right) + \sum_{\mathbf{r} \in \text{II}} \sum_{\mu, \nu \neq \mu} \left(\gamma_{\mu\nu}^* \Phi_{\mathbf{r}-\mathbf{e}_\nu}^\dagger \Phi_{\mathbf{r}} S_{\mathbf{r},\mathbf{r}-\mathbf{e}_\mu}^z S_{\mathbf{r},\mathbf{r}-\mathbf{e}_\nu}^+ + \text{h.c.} \right) \right\} + \text{const.}$$

$$J_{\pm\pm} = 0$$

How we do this: compact abelian Higgs U(1) lattice gauge theory

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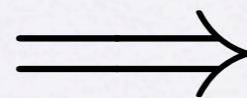
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vacuum: quantum superposition of two-in-two-out states

H = hopping Hamiltonian for spinons in fluctuating background

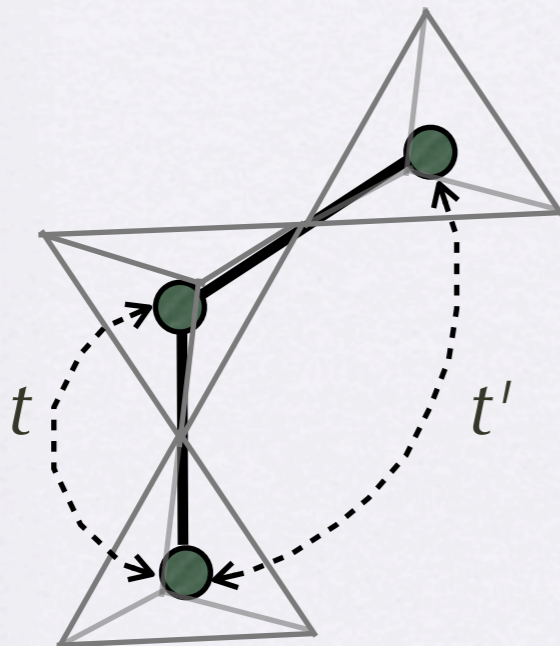
gauge Mean Field Theory (gMFT)

$$\Phi^\dagger \Phi s s \rightarrow \Phi^\dagger \Phi \langle s \rangle \langle s \rangle + \langle \Phi^\dagger \Phi \rangle s \langle s \rangle + \langle \Phi^\dagger \Phi \rangle \langle s \rangle s - 2 \langle \Phi^\dagger \Phi \rangle \langle s \rangle \langle s \rangle$$

$$H_s^{\text{MF}} = - \sum_{\mathbf{r}} \sum_{\mu} \vec{h}_{\text{eff},\mu}^{\text{MF}} \cdot \vec{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_\mu} \quad \text{free (but self-consistent) "spins"}$$

$$H_\Phi^{\text{MF}} = - \sum_{\mathbf{r}} \sum_{\mu \neq \nu} \left[t_\mu^{\text{MF}} \Phi_{\mathbf{r}}^\dagger \Phi_{\mathbf{r}+\mathbf{e}_\mu} + t'_{\mu\nu}^{\text{MF}} \Phi_{\mathbf{r}}^\dagger \Phi_{\mathbf{r}+\mathbf{e}_\mu-\mathbf{e}_\nu} + \text{h.c.} \right]$$

hopping Hamiltonian for spinons in fixed (but self-consistent) background

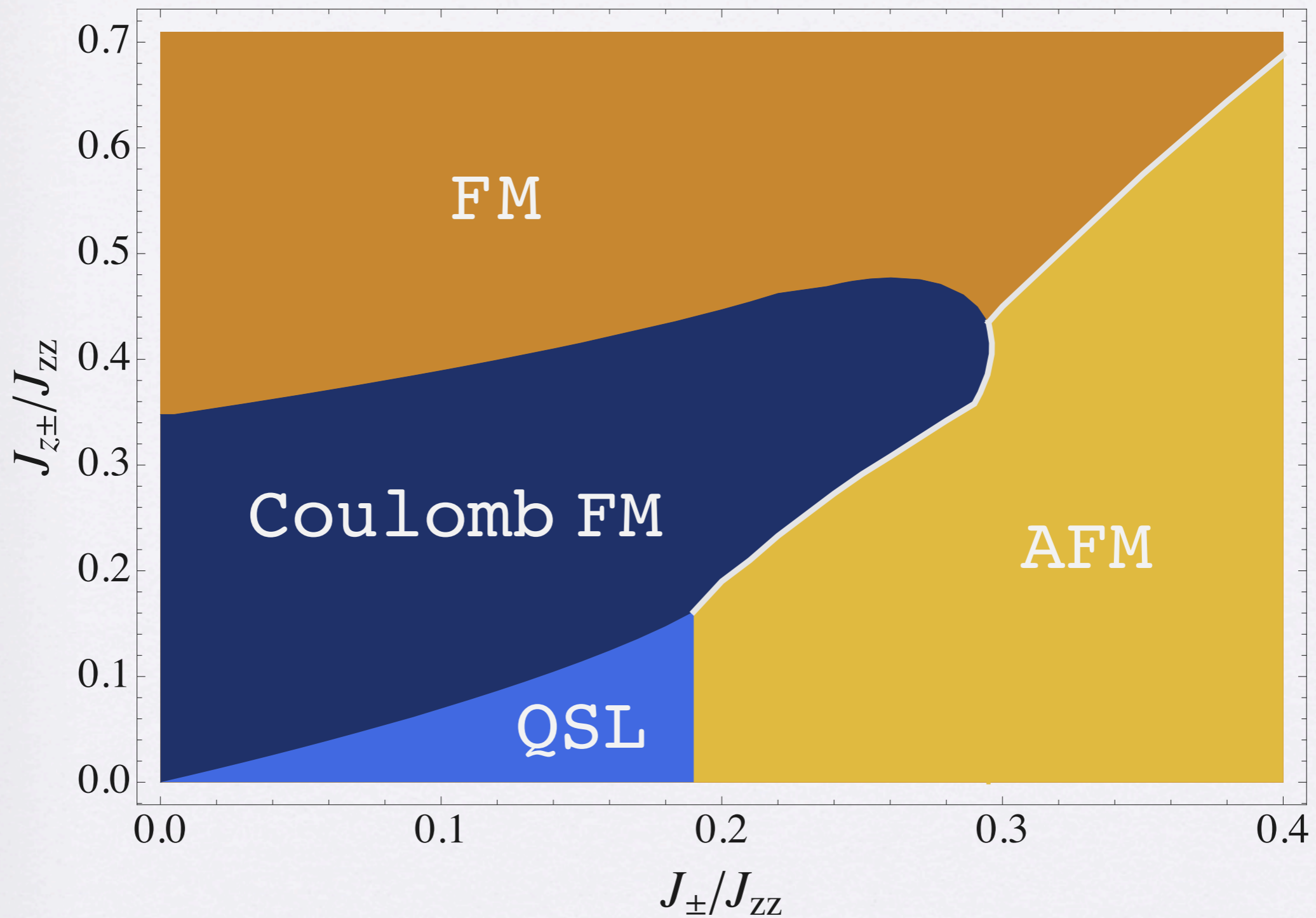


Solve the consistency equations



Quantum Spin Liquids in Quantum Spin Ices

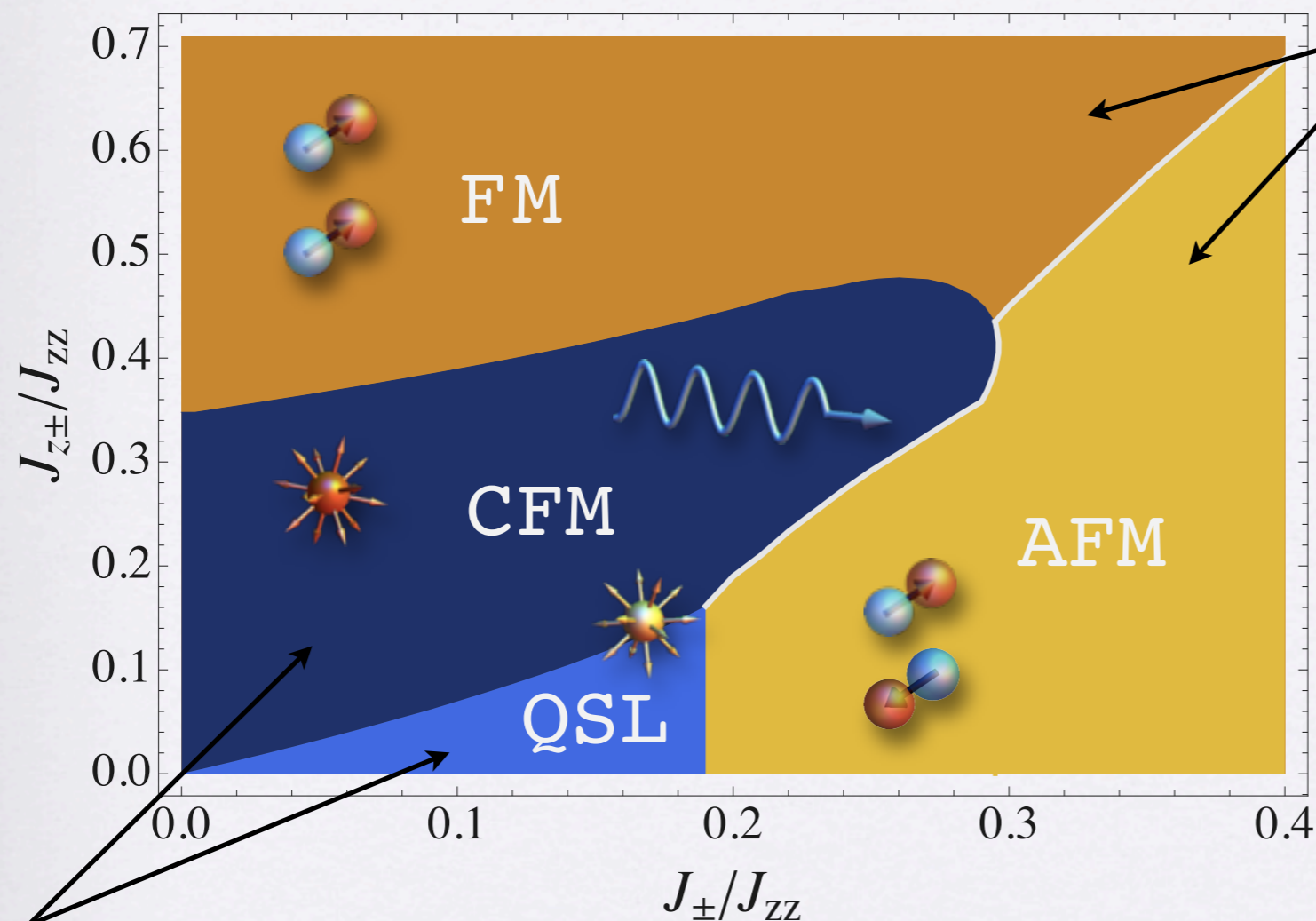
Phase diagram



$$J_{\pm\pm} = 0$$

Phase diagram

$$J_{\pm\pm} = 0$$



Higgs
 = gauge symmetry
 breaking
 = condensed
 = **conventional phases**

$$\langle \Phi \rangle \neq 0$$

deconfined
 = uncondensed
 = **exotic**

$$\langle \Phi \rangle = 0$$

$\langle \Phi \rangle$	$\langle S^z \rangle$	phase
$\neq 0$	$= 0$	AFM
$\neq 0$	$\neq 0$	FM
$= 0$	$= 0$	QSL
$= 0$	$\neq 0$	CFM

Insight into the exotic phases

- superposition of states

$|\psi\rangle \sim$ equal-weight quantum superposition of 2-in-2-out states

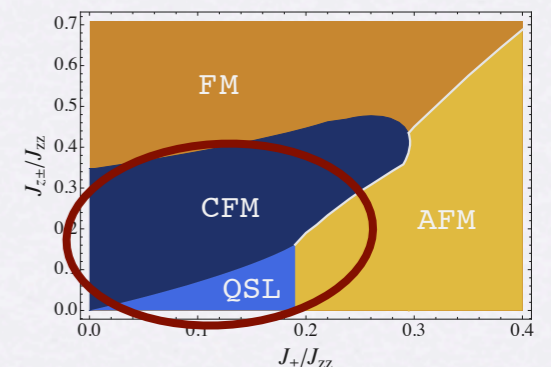
- inelastic structure factor $\mathcal{S}(\mathbf{k}, \omega) = \sum_{\mu, \nu} \left[\delta_{\mu\nu} - (\hat{\mathbf{k}})_{\mu} (\hat{\mathbf{k}})_{\nu} \right] \sum_{a, b} \langle m_a^{\mu}(-\mathbf{k}, -\omega) m_b^{\nu}(\mathbf{k}, \omega) \rangle$

$\langle S^z S^z \rangle$ contribution \longleftrightarrow photon mode

$\langle S^+ S^- \rangle$ contribution \longleftrightarrow spinon mode

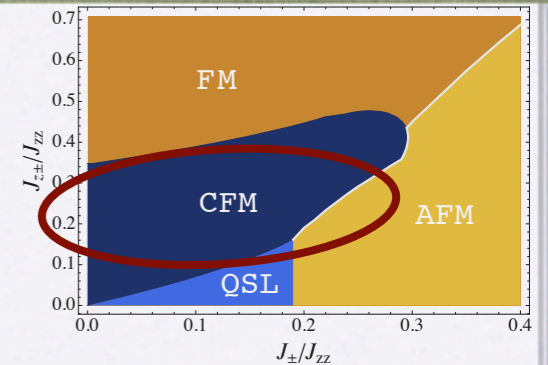
$$S^z |\psi\rangle = |1 \text{ photon} + \text{vacuum}\rangle$$

$$S^+ |\psi\rangle = |2 \text{ spinons} + \text{vacuum}\rangle$$



The Coulomb ferromagnet

(secretly a quantum spin liquid!)



- magnetized

$$\langle S^z \rangle \neq 0$$

$$\langle S^z \rangle < 1/2$$



spins with non-zero expectation value

$\langle \Phi \rangle$	$\langle S^z \rangle$	phase
$\neq 0$	$= 0$	AFM
$\neq 0$	$\neq 0$	FM
$= 0$	$= 0$	QSL
$= 0$	$\neq 0$	CFM

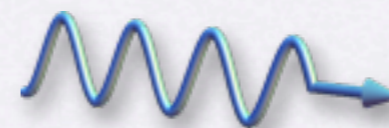
- supports exotic excitations

$$\langle \Phi \rangle = 0$$

$$\langle S^\pm \rangle = 0$$



spinon



gapless photon



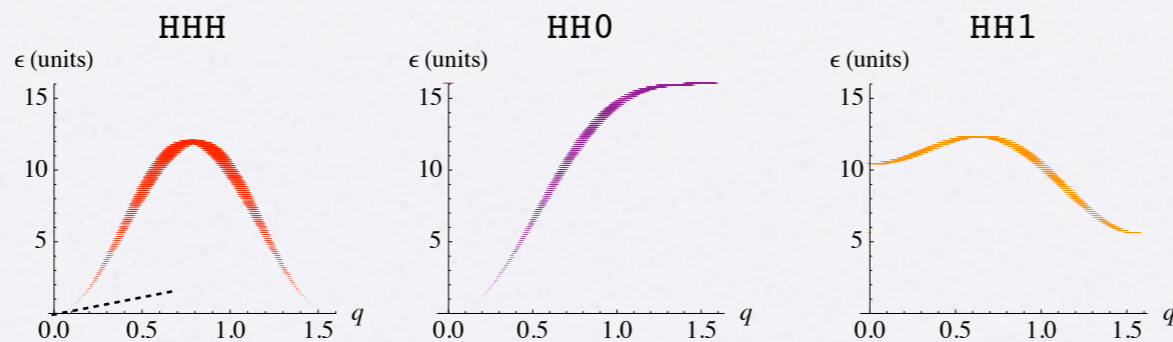
"electric" monopole

Signatures of the deconfined phases



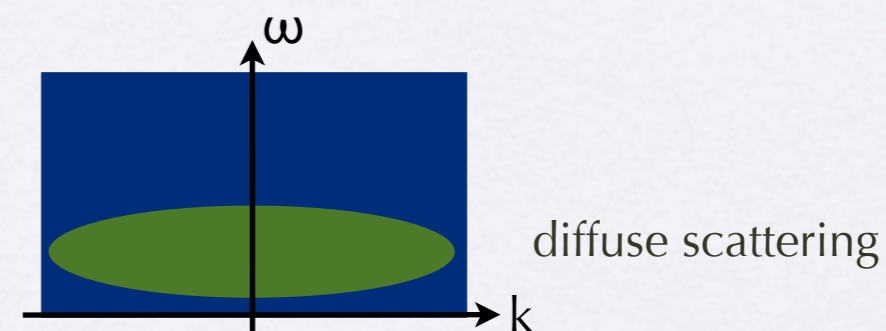
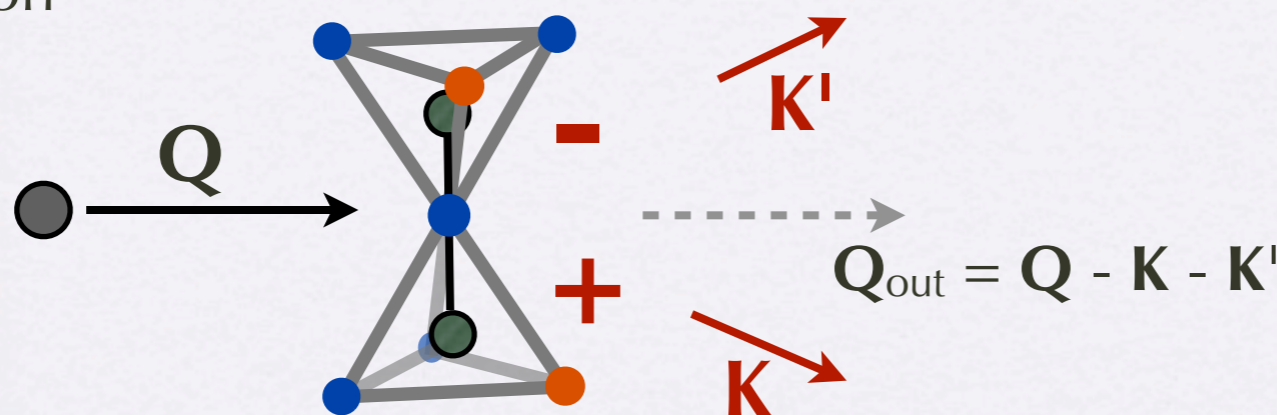
- inelastic neutron scattering:

- photon



$$\mathcal{S}(\mathbf{k} \sim \mathbf{0}, \omega) \sim \omega \delta[\omega - v|\mathbf{k}|]$$

- spinon

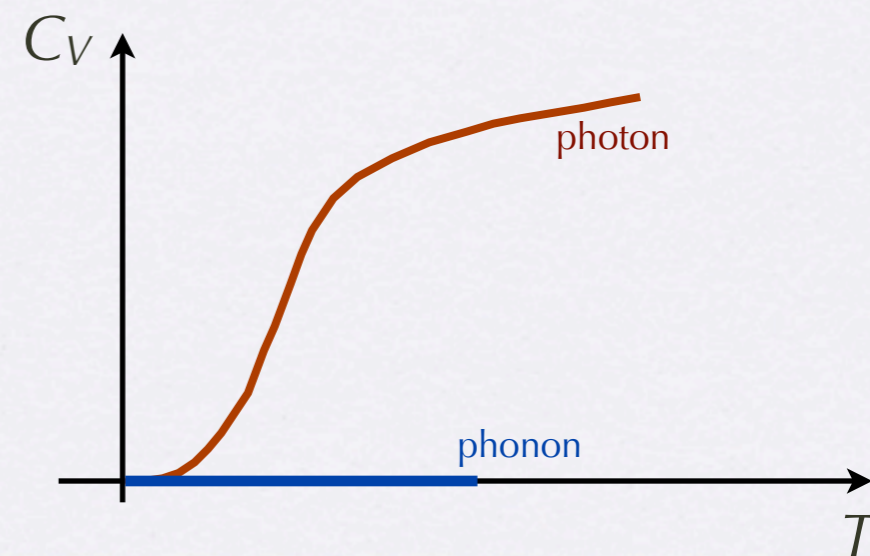


- specific heat:

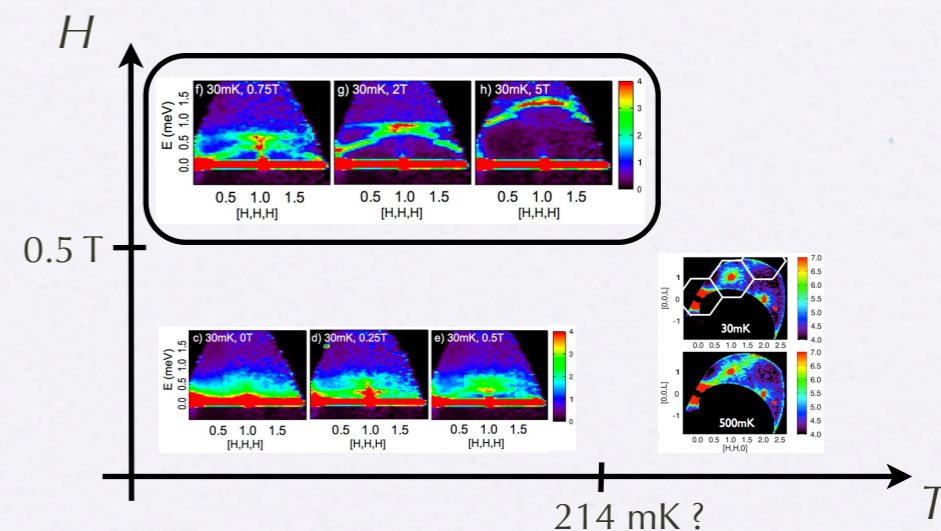
- photon

$$C_v^{T \approx 0} \sim B_{\text{photon}} T^3 + B_{\text{phonon}} T^3$$

$$B_{\text{photon}} \sim 1000 B_{\text{phonon}}$$

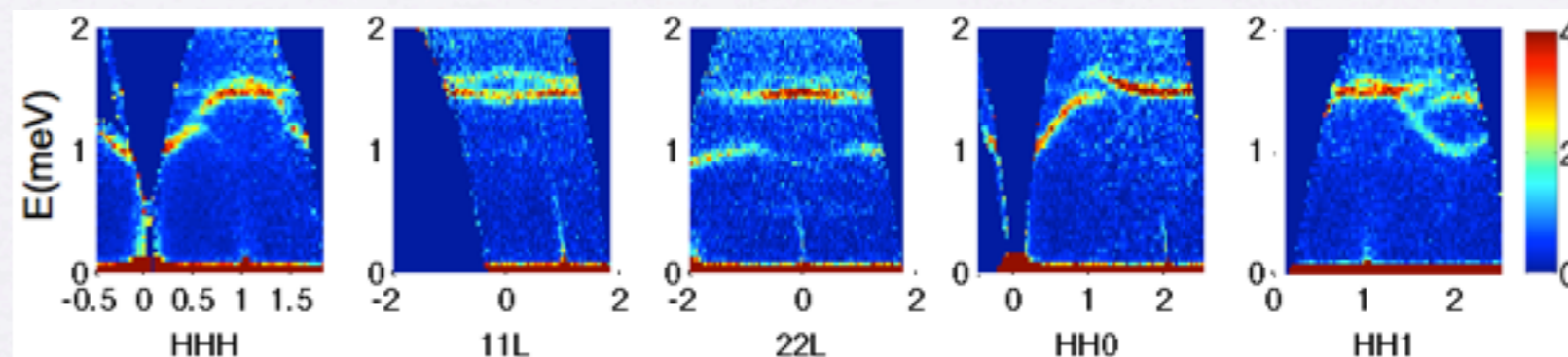


Yb₂Ti₂O₇



high field $H = 5T$

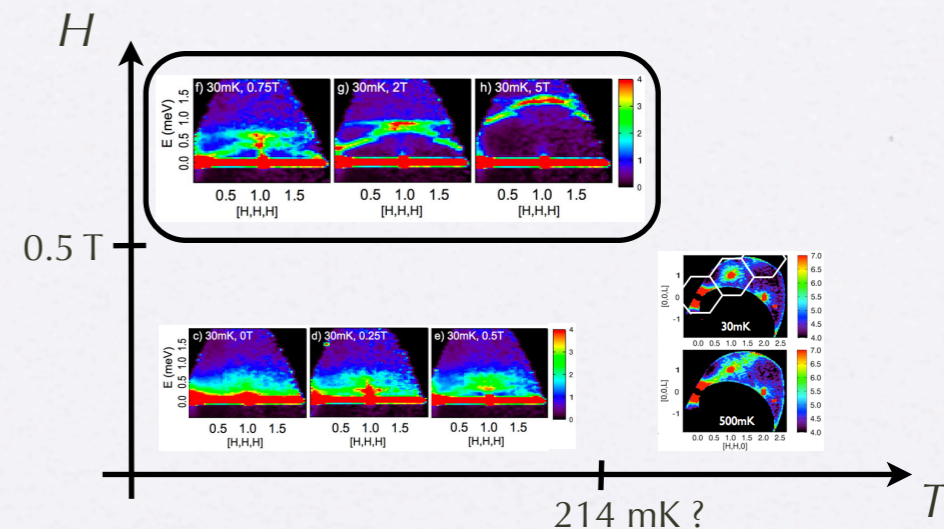
experiment



spin wave theory

1. classical high-field ground state
2. Holstein-Primakoff bosons in the spirit of large s
3. calculation of the inelastic structure factor

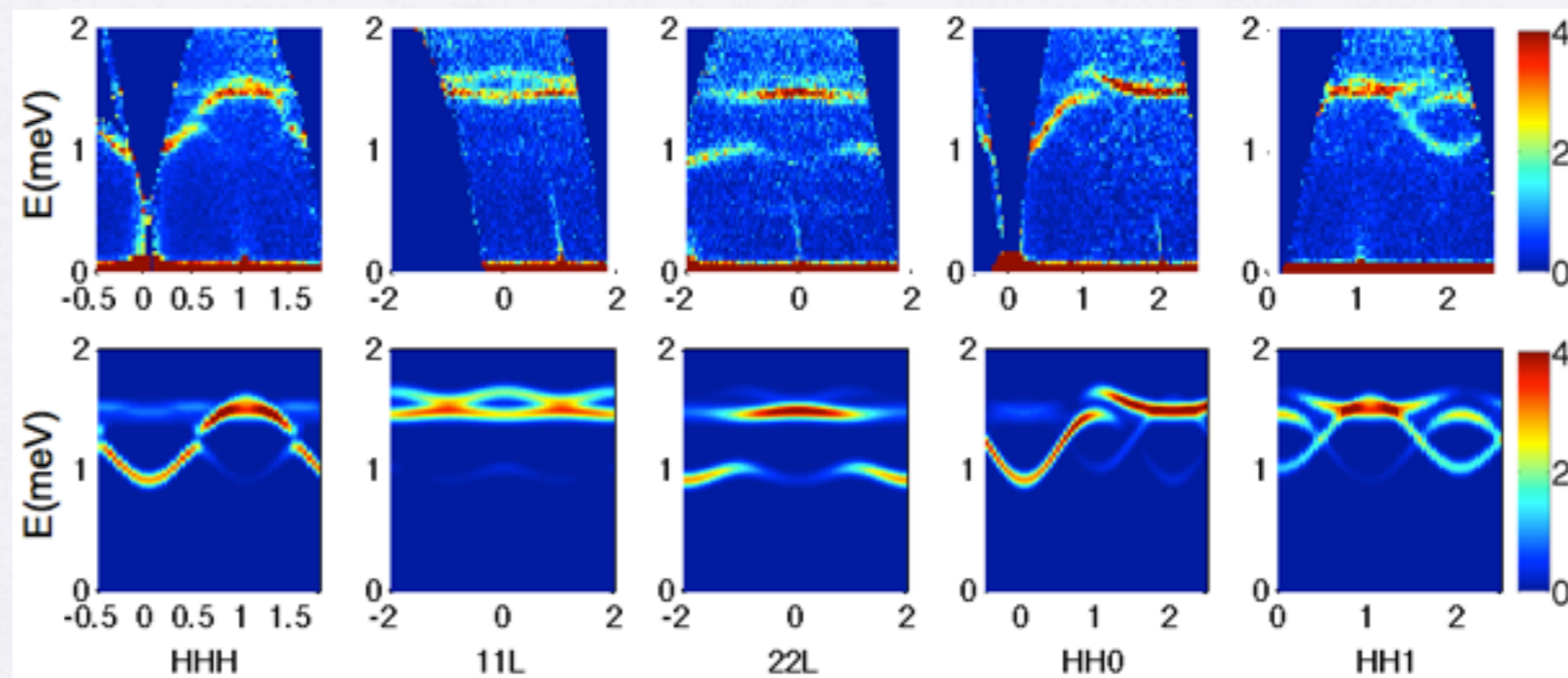
Yb₂Ti₂O₇



high field $H = 5T$

experiment

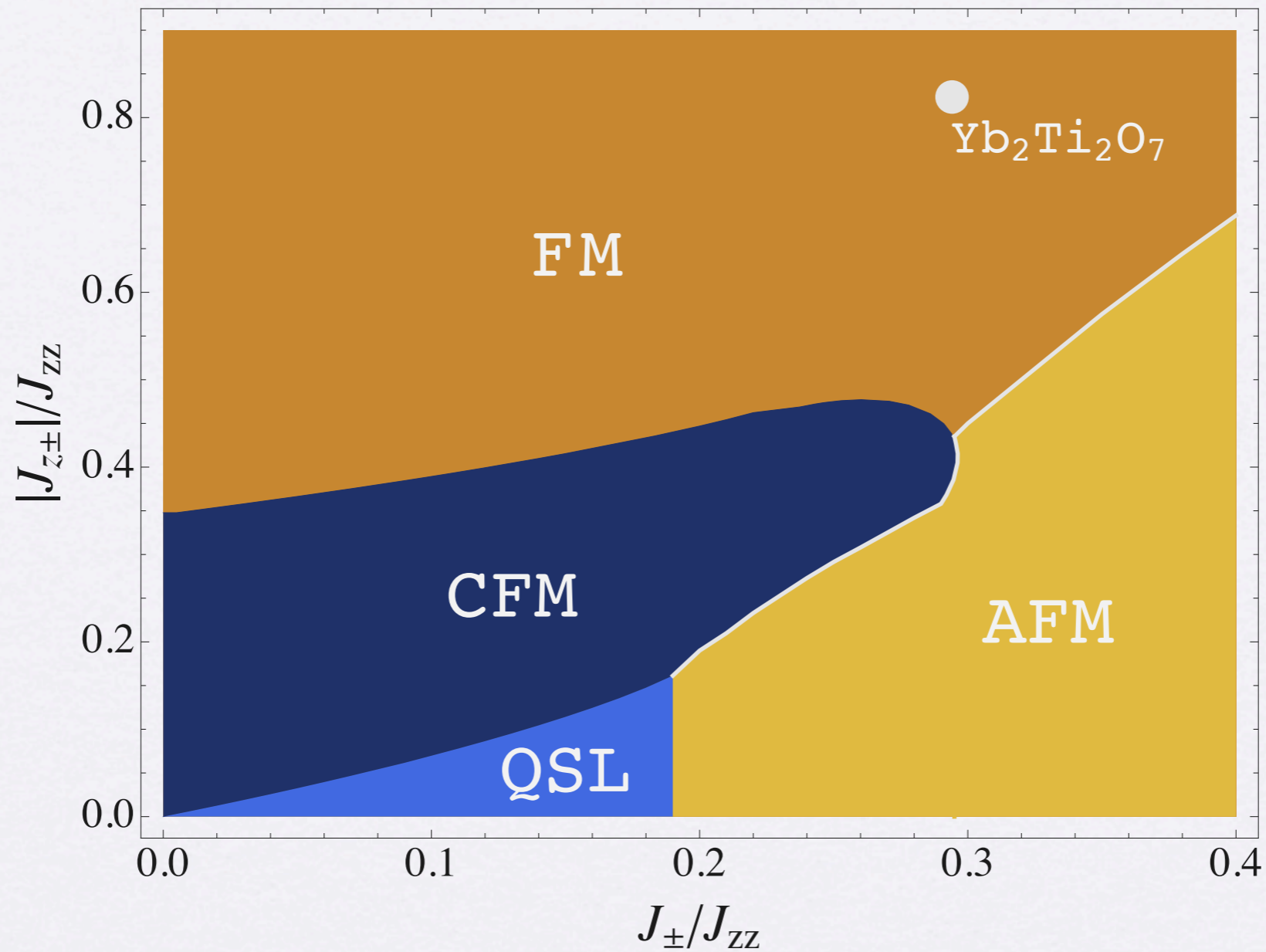
spin wave theory



$$J_{zz} = 0.17, \quad J_{\pm} = 0.05, \quad J_{z\pm} = -0.14, \quad J_{\pm\pm} = 0.05 \quad \text{meV}$$

Yb₂Ti₂O₇

$$J_{zz} = 0.17, \quad J_{\pm} = 0.05, \quad J_{z\pm} = -0.14, \quad J_{\pm\pm} = 0.05 \text{ meV}$$

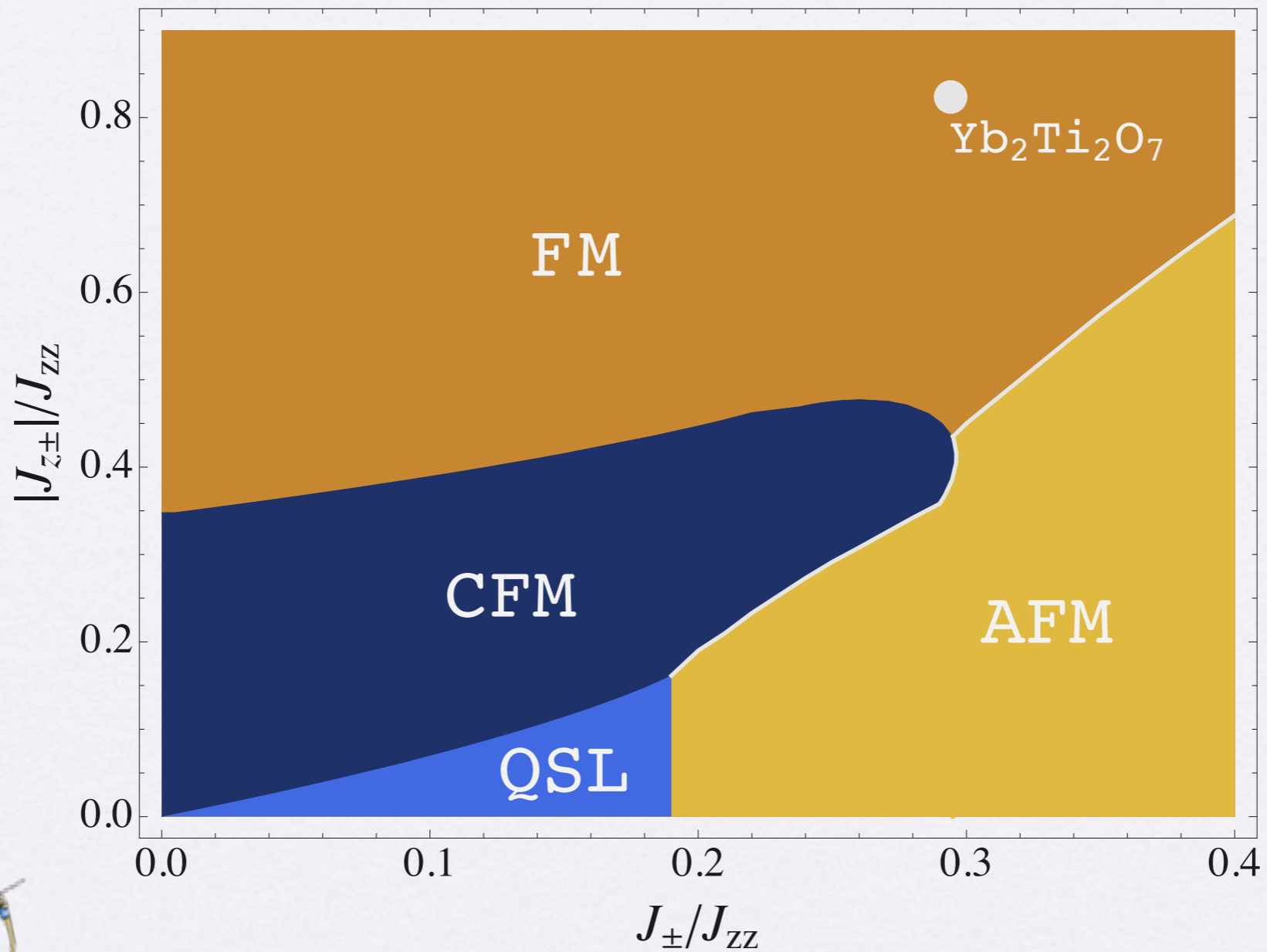


some
uncertainties

follow-up work: Ross *et al.* (2011), Chang *et al.* (2011), Applegate *et al.* (2012)

Yb₂Ti₂O₇

$$J_{zz} = 0.17, \quad J_{\pm} = 0.05, \quad J_{z\pm} = -0.14, \quad J_{\pm\pm} = 0.05 \text{ meV}$$

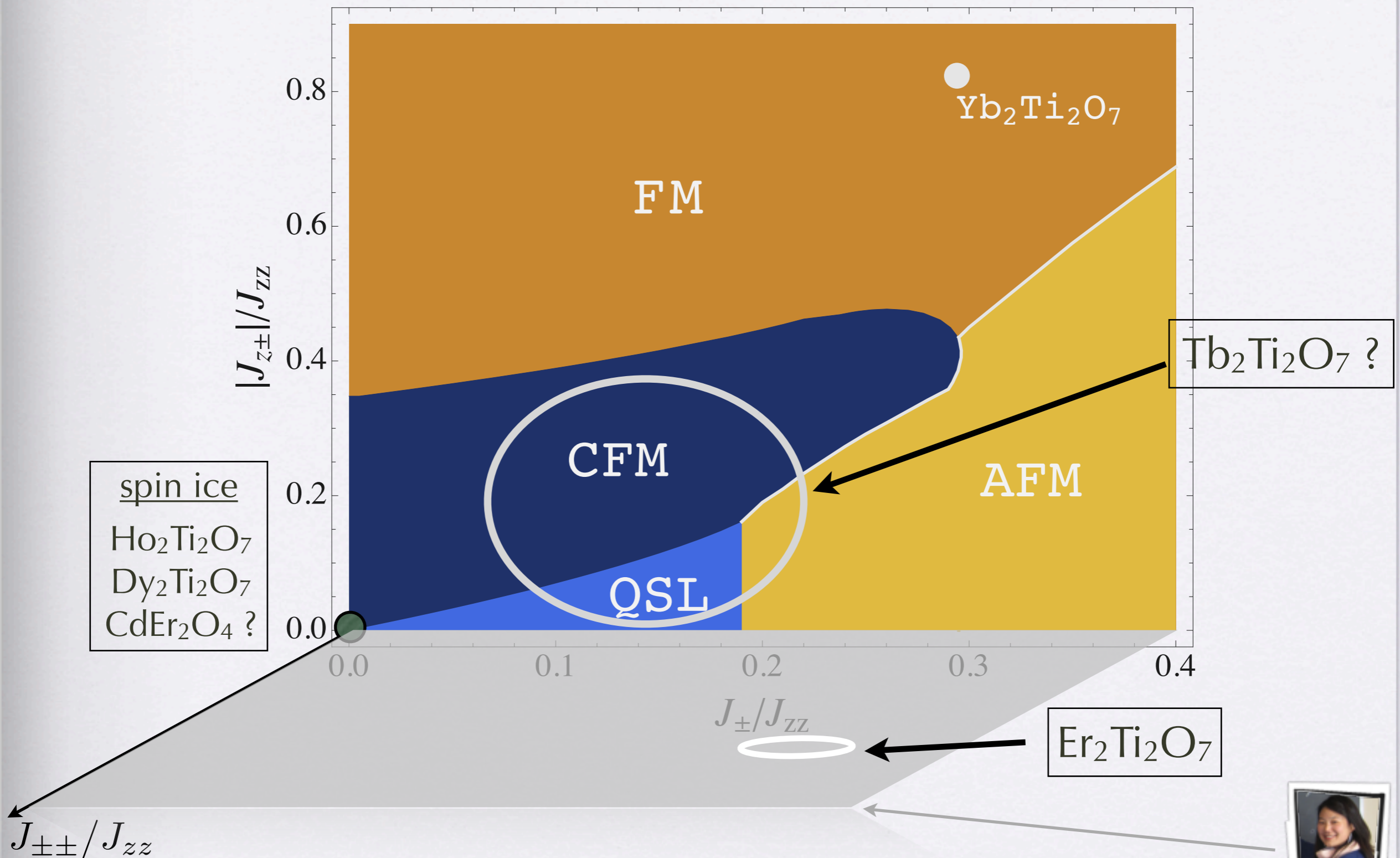


some
uncertainties



Tb₂Ti₂O₇ ? Er₂Ti₂O₇ ? CdEr₂O₄ ? ...

Materials



many other materials under investigation (cf. HFM 2012)

SungBin Lee



Conclusions and perspectives

- Model and phase diagram which should apply to a **wide spectrum of materials**
- Realization of the **U(1) QSL** in a phase diagram for **real materials**
- Existence of a **new phase of matter: the Coulomb FM**
- Need **numerics**
- Need exchange constants of **more materials**
- Need more **low temperature specific heat** data
- Effects of **disorder**
- Effects of **temperature**
- **Longer range** interactions...



Thank you for your attention