#### Coulombic Quantum Liquids in Spin-1/2 Pyrochlores

#### Lucile Savary









Natal, December 13th, 2011

#### Collaborators





#### Leon Balents (KITP, UCSB)

#### Yb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> project



Kate Ross

Bruce Gaulin

(experiments, Mc Master)

Special thanks to Benjamin Canals and Peter Holdsworth.

# Yb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>: puzzling experimental features



Ross et al., PRL 2009, PRB 2011

# Yb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>: puzzling experimental features



#### What we know

• What spin ice is -- Michel's talk this morning

$$H_{\rm SI} = J_{zz} \sum_{\langle i,j \rangle} \mathsf{S}_i^z \mathsf{S}_j^z \qquad J_{zz} > 0$$



two-in-two-out states for one tetrahedron

- What the generic definition of a quantum spin liquid is (and that it would be very nice to find one in nature) -- Leon's talk yesterday afternoon
  - looks trivial:  $\langle \vec{S} \rangle = \vec{0}$  \*history
  - but non-trivial correlations & fractional excitations!
- Good place to look for QSLs: frustrated magnets

quantum

entanglement



- grown rare-earth pyrochlores: Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Ho<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, Dy<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, Er<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Yb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Tb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>, Er<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, Tb<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, Pr<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, Nd<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, Gd<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, ...
- grown rare-earth B-site spinels: CdEr<sub>2</sub>S<sub>4</sub>, CdEr<sub>2</sub>Se<sub>4</sub>, CdYb<sub>2</sub>S<sub>4</sub>, CdYb<sub>2</sub>Se<sub>4</sub>, MgYb<sub>2</sub>S<sub>4</sub>, MgYb<sub>2</sub>S<sub>4</sub>, MnYb<sub>2</sub>S<sub>4</sub>, MnYb<sub>2</sub>Se<sub>4</sub>, FeYb<sub>2</sub>S<sub>4</sub>, CdTm<sub>2</sub>S<sub>4</sub>, CdHo<sub>2</sub>S<sub>4</sub>, FeLu<sub>2</sub>S<sub>4</sub>, MnLu<sub>2</sub>S<sub>4</sub>, MnLu<sub>2</sub>Se<sub>4</sub>, ...

lots of room for diverse behaviors!



Gardner, Gingras, Greedan, RMP 2010, Lago et al. PRL 2010

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spin ices



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- grown rare-earth B-site spinels: CdEr<sub>2</sub>S<sub>4</sub>, CdEr<sub>2</sub>Se<sub>4</sub>, CdYb<sub>2</sub>S<sub>4</sub>, CdYb<sub>2</sub>Se<sub>4</sub>, MgYb<sub>2</sub>S<sub>4</sub>, MgYb<sub>2</sub>S<sub>4</sub>, MnYb<sub>2</sub>S<sub>4</sub>, MnYb<sub>2</sub>Se<sub>4</sub>, FeYb<sub>2</sub>S<sub>4</sub>, CdTm<sub>2</sub>S<sub>4</sub>, CdHo<sub>2</sub>S<sub>4</sub>, FeLu<sub>2</sub>S<sub>4</sub>,  $MnLu_2S_4$ ,  $MnLu_2Se_4$ , ...

lots of room for diverse behaviors!







• grown rare-earth pyrochlores:  $H_{02}Ti_2O_7$ ,  $Dy_2Ti_2O_7$ ,  $H_{02}Sn_2O_7$ ,  $Dy_2Sn_2O_7$ ,  $Er_2Ti_2O_7$ ,  $Yb_2Ti_2O_7$ ,  $Tb_2Ti_2O_7$ ,  $Er_2Sn_2O_7$ ,  $Tb_2Sn_2O_7$ ,  $Pr_2Sn_2O_7$ ,  $Nd_2Sn_2O_7$ ,  $Gd_2Sn_2O_7$ , ...

#### quantum spin liquids ?

 grown rare-earth B-site spinels: CdEr<sub>2</sub>S<sub>4</sub>, CdEr<sub>2</sub>Se<sub>4</sub>, CdYb<sub>2</sub>S<sub>4</sub>, CdYb<sub>2</sub>Se<sub>4</sub>, MgYb<sub>2</sub>S<sub>4</sub>, MgYb<sub>2</sub>S<sub>4</sub>, MnYb<sub>2</sub>S<sub>4</sub>, MnYb<sub>2</sub>Se<sub>4</sub>, FeYb<sub>2</sub>S<sub>4</sub>, CdTm<sub>2</sub>S<sub>4</sub>, CdHo<sub>2</sub>S<sub>4</sub>, FeLu<sub>2</sub>S<sub>4</sub>, MnLu<sub>2</sub>S<sub>4</sub>, MnLu<sub>2</sub>Se<sub>4</sub>, ...

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#### Outline

- method
- results
- experimental signatures
- materials

based on Savary and Balents arXiv 1110.2185 - to appear in PRL (Dec. 2011)

### Symmetries of the Hamiltonian

rare-earths : intrinsic strong spin-orbit coupling

discrete cubic symmetries only

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



A<sub>2</sub>B<sub>2</sub>O<sub>7</sub> "pyrochlore oxides"



AB<sub>2</sub>X<sub>4</sub> spinels

#### General NN exchange Hamiltonian for effective spins 1/2

$$\begin{split} H &= \sum_{\langle ij \rangle} \left[ \begin{array}{c} J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z \\ &- J_{\pm} (\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+) \\ &+ J_{z\pm} \left[ \mathsf{S}_i^z (\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_j^-) + i \leftrightarrow j \right] \end{split}$$

$$+ J_{\pm\pm} \left[ \gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^- \right] \right]$$

 $J_{z\pm}/J_{zz}$  $J_{\pm\pm}/J_{zz}$  $J_{\pm\pm}/J_{zz}$ 



local axes, specific local bases

to each material corresponds a set of J's

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local axes, specific local bases

to each material corresponds a set of J's

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

Curnoe PRB 2008, Ross Savary Gaulin Balents PRX 2011

#### The Hermele et al. QSL



#### The Hermele et al. QSL





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



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

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



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

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

 $\implies$   $\stackrel{\mathsf{p}}{\longrightarrow}$ 

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

magnetic monopoles = spinons one entangled ground state (= vacuum)

> spinons "electric" monopoles gapless photon

z±/Jzz ↑	
	$J_{\pm\pm}/J_{ZZ}$
	→ / <sub>±</sub> // <sub>zz</sub>

J<sub>z±</sub>/J<sub>zz</sub>  $|_{\pm\pm}/|_{77}$ ► |+/|77

Gingras, Introduction to Frustrated Magnetism (2011), Hermele Fisher Balents, PRB 2004











spinon, s = 1/2





$$\begin{cases} \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} = \Phi_{\mathbf{r}}^{\dagger} \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}} \\ \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} = \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} \end{cases}$$





$$\left( \begin{array}{l} \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} = \Phi_{\mathbf{r}}^{\dagger} \, \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}} \\ \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} = \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} \end{array} \right)$$

$$\begin{cases} \Phi_{\mathbf{r}} \to \Phi_{\mathbf{r}} e^{-i\chi_{\mathbf{r}}} \\ \mathsf{s}_{\mathbf{rr}'}^{\pm} \to \mathsf{s}_{\mathbf{rr}'}^{\pm} e^{\pm i(\chi_{\mathbf{r}'} - \chi_{\mathbf{r}})} \end{cases}$$

U(1) gauge symmetry



$$\begin{split} \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} &= \Phi_{\mathbf{r}}^{\dagger} \, \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}} \\ \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} &= \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} \end{split}$$

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U(1) gauge symmetry

the slave particles have a simple interpretation

$$\begin{split} \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} &= \Phi_{\mathbf{r}}^{\dagger} \, \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{+} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}} \\ \mathsf{S}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} &= \mathsf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{z} \end{split}$$

$$|\Phi_{\mathbf{r}}| = 1$$
  
 $Q_{\mathbf{r}} = \pm \sum_{\mu} \mathsf{s}^{z}_{\mathbf{r},\mathbf{r}\pm\mathbf{e}_{\mu}}$ 

$$s_{\mathbf{rr'}}^{z} = E_{\mathbf{rr'}}$$
$$s_{\mathbf{rr'}}^{\pm} = e^{\pm iA_{\mathbf{rr'}}}$$

our spinons are bosons

$$\begin{split} S^{+}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} &= \Phi^{\dagger}_{\mathbf{r}} \, \mathbf{s}^{+}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}} \\ S^{z}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} &= \mathbf{s}^{z}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} \end{split}$$

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$$s_{\mathbf{rr'}}^{z} = E_{\mathbf{rr'}}$$
$$s_{\mathbf{rr'}}^{\pm} = e^{\pm iA_{\mathbf{rr'}}}$$

they can condense

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

$$\begin{split} S^{+}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} &= \Phi^{\dagger}_{\mathbf{r}} \, \mathbf{s}^{+}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}} \\ S^{z}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} &= \mathbf{s}^{z}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}} \end{split}$$

our spinons are bosons

$\Phi_{\mathbf{r}} =1$	
$Q_{f r}=\pm\sum{\sf s}^z_{{f r},{f r}\pm{f e}_\mu}$	
$\overline{\mu}$	

$$\mathbf{S}_{\mathbf{rr}'}^{\sim} = E_{\mathbf{rr}'}$$
$$\mathbf{S}_{\mathbf{rr}'}^{\pm} = e^{\pm iA_{\mathbf{rr}'}}$$

they can condense

$\langle \Phi  angle$	phase		
$\neq 0$	conventional		
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$$H = \sum_{\mathbf{r}\in\mathbf{I},\mathbf{II}} \frac{J_{zz}}{2} Q_{\mathbf{r}}^{2} - J_{\pm} \left\{ \sum_{\mathbf{r}\in\mathbf{I}} \sum_{\mu,\nu\neq\mu} \Phi_{\mathbf{r}+\mathbf{e}_{\mu}}^{\dagger} \Phi_{\mathbf{r}+\mathbf{e}_{\nu}} \mathbf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\mu}}^{-} \mathbf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\nu}}^{-} + \sum_{\mathbf{r}\in\mathbf{II}} \sum_{\mu,\nu\neq\mu} \Phi_{\mathbf{r}-\mathbf{e}_{\mu}}^{\dagger} \Phi_{\mathbf{r}-\mathbf{e}_{\nu}} \mathbf{s}_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\nu}}^{-} \mathbf{s}_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\nu}}^{-} \right\}$$
$$-J_{z\pm} \left\{ \sum_{r\in\mathbf{I}} \sum_{\mu,\nu\neq\mu} \left( \gamma_{\mu\nu}^{*} \Phi_{\mathbf{r}}^{\dagger} \Phi_{\mathbf{r}+\mathbf{e}_{\nu}} \mathbf{s}_{\mathbf{r},\mathbf{r}+\mathbf{e}_{\nu}}^{+} + \mathrm{h.c.} \right) + \sum_{\mathbf{r}\in\mathbf{II}} \sum_{\mu,\nu\neq\mu} \left( \gamma_{\mu\nu}^{*} \Phi_{\mathbf{r}-\mathbf{e}_{\nu}}^{\dagger} \Phi_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\nu}}^{-} + \mathrm{h.c.} \right) \right\} + \mathrm{cont} \left\{ e^{\mathbf{II}} \sum_{\mathbf{r}\in\mathbf{II}} \left( \gamma_{\mu\nu}^{*} \Phi_{\mathbf{r}-\mathbf{e}_{\nu}}^{\dagger} \Phi_{\mathbf{r},\mathbf{r}-\mathbf{e}_{\nu}}^{-} + \mathrm{h.c.} \right) \right\}$$

 $J_{\pm\pm} = 0$ 

$ \begin{cases} 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} \end{cases} $	$\begin{vmatrix} \Phi_{\mathbf{r}} \end{vmatrix} = Q_{\mathbf{r}} = \pm$	$\frac{1}{\sum_{\mu} s^{z}_{\mathbf{r},\mathbf{r}\pm\mathbf{e}}}$	${f s}_{{f r}{f r}'}^{z}={f s}_{{f r}{f r}'}^{\pm}={f s}_{{f r}{f r}}^{\pm}={f s}_{{f r}{h}}^{\pm}={f s}_{{f r}{f r}}^{\pm}={f$	$= E_{\mathbf{rr'}}$ $= e^{\pm iA_{\mathbf{rr}}}$
our spinons are bosons	$\Longrightarrow$	they ca	n condense	
		$\langle \Phi \rangle$	phase	
		$\neq 0$	conventional	
		=0	exotic	

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

H = hopping Hamiltonian for spinons in fluctuating background

 $\Phi^{\dagger}\Phi\,\mathsf{s}\,\mathsf{s}\to\Phi^{\dagger}\Phi\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\mathsf{s}\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\mathsf{s}-2\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle$ 

 $\Phi^{\dagger}\Phi\,\mathsf{s}\,\mathsf{s}\to\Phi^{\dagger}\Phi\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\mathsf{s}\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\mathsf{s}-2\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle$ 

$$H_{\rm s}^{\rm MF} = -\sum_{\bf r} \sum_{\mu} \vec{{\sf h}}_{{\rm eff},\mu}^{\rm MF} \cdot \vec{{\sf s}}_{{\bf r},{\bf r}+{\bf e}_{\mu}}$$

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

 $\Phi^{\dagger}\Phi\,\mathbf{s}\,\mathbf{s} \to \Phi^{\dagger}\Phi\langle\mathbf{s}\rangle\langle\mathbf{s}\rangle + \langle\Phi^{\dagger}\Phi\rangle\mathbf{s}\langle\mathbf{s}\rangle + \langle\Phi^{\dagger}\Phi\rangle\langle\mathbf{s}\rangle\mathbf{s} - 2\langle\Phi^{\dagger}\Phi\rangle\langle\mathbf{s}\rangle\langle\mathbf{s}\rangle$ 

$$H_{\rm s}^{\rm MF} = -\sum_{\rm \mathbf{r}} \sum_{\mu} \vec{\mathsf{h}}_{{\rm eff},\mu}^{\rm MF} \cdot \vec{\mathsf{s}}_{{\bf r},{\bf r}+\mathbf{e}_{\mu}}$$

free (but self-consistent) "spins"

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

 $\Phi^{\dagger}\Phi\,\mathsf{s}\,\mathsf{s}\to\Phi^{\dagger}\Phi\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\mathsf{s}\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\mathsf{s}-2\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle$ 

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



 $\Phi^{\dagger}\Phi\,\mathsf{s}\,\mathsf{s}\to\Phi^{\dagger}\Phi\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\mathsf{s}\langle\mathsf{s}\rangle+\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\mathsf{s}-2\langle\Phi^{\dagger}\Phi\rangle\langle\mathsf{s}\rangle\langle\mathsf{s}\rangle$ 

$$H_{\rm s}^{\rm MF} = -\sum_{\rm \mathbf{r}} \sum_{\mu} \vec{\mathsf{h}}_{{\rm eff},\mu}^{\rm MF} \cdot \vec{\mathsf{s}}_{{\bf r},{\bf r}+\mathbf{e}_{\mu}}$$

free (but self-consistent) "spins"

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

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



Solve the consistency equations

# Now is when you tune back in for




 $J_{\pm\pm} = 0$ 

 $J_{\pm\pm}=0$ 



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

 $J_{\pm\pm}=0$ 



= 0

 $\neq 0$ 

= 0

= 0

QSL

CFM

 $J_{\pm\pm}=0$ 



= 0

 $\neq 0$ 

= 0

= 0

QSL

CFM

 $J_{\pm\pm}=0$ 



CFM

 $\neq 0$ 

= 0

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

 $J_{++} = 0$ 



 $\neq 0$ 

= 0

# Insight into the exotic phases

superposition of states

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

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

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

 $\mathsf{S}^{z}|\psi\rangle = |1 \text{ photon} + \text{vacuum}\rangle$  $\mathsf{S}^{+}|\psi\rangle = |2 \text{ spinons} + \text{vacuum}\rangle$ 



# The Coulomb ferromagnet (secretly a quantum spin liquid!)

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

FΜ

CFM

0.1

QSL

0.2 $J_{\pm}/J_{zz}$ 

# The Coulomb ferromagnet (secretly a quantum spin liquid!)

magnetized

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

FΜ

CFM

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QSL

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#### The Coulomb ferromagnet (secretly a quantum spin liquid!)

magnetized

 $\langle \mathsf{S}^z \rangle \neq 0$ 



spins with non-zero expectation value

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

0.6

FΜ

CFM

0.1

QSL

0.2

 $J_{\pm}/J_{\rm ZZ}$ 

0.3

#### The Coulomb ferromagnet (secretly a quantum spin liquid!)

magnetized

 $\langle \mathsf{S}^z \rangle \neq 0$ 

 $\langle \mathsf{S}^z \rangle < 1/2$ 



spins with non-zero expectation value

$\langle \Phi  angle$	$\langle S^z  angle$	phase
$\neq 0$	= 0	AFM
$\neq 0$	$\neq 0$	FM
= 0	= 0	QSL
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FΜ

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= 0	= 0	QSL
= 0	$\neq 0$	CFM

0.6

FΜ

CFM

0.1

QSL

0.2  $J_{\pm}/J_{\rm ZZ}$ 

supports exotic excitations

gapless photon  $\left< \Phi \right> = 0$  $\left< \mathsf{S}^{\pm} \right> = 0$ spinon "electric" monopole

#### Signatures of the deconfined phases

- inelastic neutron scattering:
  - photon

• spinon

• <u>specific heat:</u>

#### • photon



• <u>specific heat:</u>

#### • photon



- <u>specific heat:</u>
  - photon



- <u>specific heat:</u>
  - photon



- <u>specific heat:</u>
  - photon



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



 $B_{\rm photon} \sim 1000 \, B_{\rm phonon}$ 





Ross, Savary, Gaulin, Balents PRX 2011



high field H = 5T

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high field H = 5T

experiment



spin wave theory

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experiment

spin wave theory

- E(meV) 1 2 0 -0.5 0 0.5 1 1.5 0. 0 2 0 2 2 -2 2 0 1 1 0 HHH 11L 22L HH0 HH1
  - 1. classical high-field ground state
  - 2. Holstein-Primakoff bosons in the spirit of large s
  - 3. calculation of the inelastic structure factor



high field H = 5T

#### experiment

#### spin wave theory





high field H = 5T

spin wave theory

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

0

11L

2 -2

0

22L

2

0

2

0

1

HH0

0 -2

-0.5 0 0.5 1 1.5

HHH

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2

1

HH1

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











 $Tb_2Ti_2O_7$ ?  $Er_2Ti_2O_7$ ?  $CdEr_2O_4$ ?...

#### Materials 0.8 $Yb_2Ti_2O_7$ FM 0.6 $2Z r I/J_{ZZ}$ $Tb_2Ti_2O_7$ ? CFM AFM spin ice 0.2 $Ho_2Ti_2O_7$ $Dy_2Ti_2O_7$ QSI $CdEr_2O_4$ ? 0.0 0.1 0.3 0.4 0.0 0.2 $J_{\pm}/J_{ m ZZ}$ $Er_2Ti_2O_7$ ? $J_{\pm\pm}/J_{zz}$ 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

# extra slides for questions



# Order parameters

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

$$\begin{cases} \hat{\mathbf{e}}_0 = (1, 1, 1)/\sqrt{3} \\ \hat{\mathbf{e}}_1 = (1, -1, -1)/\sqrt{3} \\ \hat{\mathbf{e}}_2 = (-1, 1, -1)/\sqrt{3} \\ \hat{\mathbf{e}}_3 = (-1, -1, 1)/\sqrt{3}, \end{cases}$$

$$\begin{cases} \mathbf{\hat{a}}_0 = (-2, 1, 1)/\sqrt{6} \\ \mathbf{\hat{a}}_1 = (-2, -1, -1)/\sqrt{6} \\ \mathbf{\hat{a}}_2 = (2, 1, -1)/\sqrt{6} \\ \mathbf{\hat{a}}_3 = (2, -1, 1)/\sqrt{6} \end{cases}$$

$$\mathbf{\hat{b}}_i = \mathbf{\hat{e}}_i imes \mathbf{\hat{a}}_i$$

# Curie-Weiss temperature

$$\Theta_{\rm CW} = \frac{1}{2k_B(2g_{xy}^2 + g_z^2)} \left[ g_z^2 J_{zz} - 4g_{xy}^2 (J_{\pm} + 2J_{\pm\pm}) - 8\sqrt{2} g_{xy} g_z J_{z\pm} \right]$$

#### parameters

$$J_{zz} = -\frac{1}{3}(2J_1 - J_2 + 2(J_3 + 2J_4))$$

$$J_{\pm} = \frac{1}{6} (2J_1 - J_2 - J_3 - 2J_4)$$

$$J_{z\pm} = \frac{1}{3\sqrt{2}}(J_1 + J_2 + J_3 - J_4)$$

$$J_{\pm\pm} = \frac{1}{6}(J_1 + J_2 - 2J_3 + 2J_4)$$

$$J_{1} = \frac{1}{3} \left( -J_{zz} + 4J_{\pm} + 2\sqrt{2}J_{z\pm} + 2J_{\pm\pm} \right)$$

$$J_{3} = \frac{1}{3} \left( -J_{zz} - 2J_{\pm} + 2\sqrt{2}J_{z\pm} - 4J_{\pm\pm} \right)$$

$$J_{2} = \frac{1}{3} \left( J_{zz} - 4J_{\pm} + 4\sqrt{2}J_{z\pm} + 4J_{\pm\pm} \right)$$

$$J_{4} = \frac{1}{3\sqrt{2}} \left( -\sqrt{2}J_{zz} - 2\sqrt{2}J_{\pm} + 2J_{z\pm} + 2\sqrt{2}J_{\pm\pm} \right)$$

$$J_{01} = \begin{pmatrix} J_{2} & J_{4} & J_{4} \\ -J_{4} & J_{1} & J_{3} \\ -J_{4} & J_{3} & J_{1} \end{pmatrix}$$



#### Other diagrams





corrected gMFT diagram



staggered magnetization for  $J_{z\pm} = 0$ 



## "Electric" monopole condensate?



### Field



#### Classical *T-H* MFT diagram for Yb<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>



### Rare-earth pyrochlores



Gardner, Gingras, Greedan, RMP 2010





 $J_{\pm} < 0$ 



