#### Orbital Ice: p-band Mott insulator on the diamond lattice



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# Outline

- Geometrical frustration:
  - Macroscopic ground-state degeneracy.
  - Perturbations  $\rightarrow$  rich and diverse magnetic phases.
- Spin ice:
  - Fractionalized excitations: magnetic monopoles.
  - Emergent gauge structure and coulomb phase.
- Orbital ice:
  - *p*-band Mott insulators in optical diamond-lattice.
  - Exact many-body ground states forming a ice manifold.
- Conclusion and outlook.



- One frustrated bond on each  $\triangle$ .
- # of ground states  $W_{gs} = e^{\text{const} \times N}$

const =  $\frac{2}{\pi} \int_0^{\pi/3} \ln(2\cos x) \, dx = 0.323066$ 

- Residual entropy  $S = k_B \ln W_{\rm gs} \propto N$ .
- No long-range spin order down to  $T \to 0$ . power-law spin correlation  $\langle \mathbf{S}_{\mathbf{r}} \cdot \mathbf{S}_0 \rangle \sim \operatorname{const}/r^2$ .

G. H. Wannier, R.M.F. Houtappel (1950)

#### **Antiferromagnet on Bi-Simplex Lattice**



#### How many Ground States ?

Discrete Spins (e.g. Ising spin):



6 out of  $2^4 = 16$  configurations satisfy  $\mathbf{S}_{\boxtimes} = 0$ , a fraction of  $\frac{3}{8}$ . Number of ground states:  $W_{\text{gs}} \approx \left(\frac{3}{8}\right)^{N_{\text{tet}}} \times 2^N = \exp\left(\frac{N}{2}\ln\frac{3}{2}\right)$ .  $[N_{\text{tet}} = N/2]$ 

 $\implies \text{Residual entropy: } \frac{S}{N} = k_B \frac{1}{2} \ln \frac{3}{2} = 1.68 \text{ J/(mol K)}.$ [Pauling's estimate]

Experimental spin-ice  $Dy_2Ti_2O_7$ :  $S_0/N = 1.86 \text{ J/(mol K)}$ . [Bramwell and Gingras (2001)]

#### Continuous (Heisenberg) Spins





Ground states: Rigid rotation  $+(\theta, \phi)$ 

#### **Pyrochlore Lattice:**

The degenerate ground states form a continuous manifold  $\mathbf{x} = (x_1, x_2, \cdots, x_D)$ 

$$D = N_{\boxtimes} = N_{\rm spin}/2$$



Macroscopic degeneracy : Hypersensitivity to perturbations Relieving the frustration:

- Quantum fluctuations (order-by-disorder): Valence bond solid
- Further neighbor interactions: Néel order, spin nematic
- Long-range dipolar interaction: Spin ice (Ho<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub> and Dy<sub>2</sub>Ti<sub>2</sub>O<sub>7</sub>)
- Spin-Lattice coupling: Néel order (ZnCr<sub>2</sub>O<sub>4</sub>, CdCr<sub>2</sub>O<sub>4</sub>)
- itinerant electrons, double-exchange coupling: Non-coplanar magnetic order



### Spin-Lattice coupling

- magnetoelastic coupling:  $E_{ij} = J(r_{ij})\mathbf{S}_i \cdot \mathbf{S}_j = J_0 \mathbf{S}_i \cdot \mathbf{S}_j + J' \delta r_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j).$
- elastic energy:  $\frac{k}{2}(\delta r_{ij})^2$ .
- $\Rightarrow E_{\text{eff}} = \sum_{\langle ij \rangle} \left[ J \mathbf{S}_i \cdot \mathbf{S}_j K (\mathbf{S}_i \cdot \mathbf{S}_j)^2 \right]$
- Collinear spins in ground state !
- Tetragonal flattening along x, y, and z.

O. Tchernyshyov, R. Moessner, S. Sondhi, PRB (2002) G.-W. Chern, C. Fennie, O. Tchernyshyov, PRB (2006)

#### Ex: CdCr2O4

#### Staggered distortion:

J.-H. Chung et al., (2005).

• 1st-order transition at T = 7.8 K.



G.-W. Chern, C. Fennie, O. Tchernyshyov, PRB (2006)



# Spin Ice:

- Pyrochlore *ferromagnet* of rare-earth ions. (e.g.  $Di_2Ti_2O_7$  with S = 15/2)
- Strong crystal-field anisotropy  $\Delta_{\rm CF} \approx 200$  K along local [111] axis  $\Rightarrow \mathbf{S}_i \approx \sigma_i \mathbf{e}_i$  at  $T \sim 10$  K, Classical Ising spins  $\sigma_i = \pm 1$ .



• a simple Spin-ice Hamiltonian:

$$H = -J_F \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = +\frac{J_F}{3} \sum_{\langle ij \rangle} \sigma_i \sigma_j = +\frac{J_F}{6} \sum_{\boxtimes} \sigma_{\boxtimes}^2.$$

 $\Rightarrow$  Ising antiferromagnet on the pyrochlore lattice



#### Evidence of ice-rule (residual entropy):



#### Why 'dipolar' spin ice obeys ice rule ?

• A realistic Hamiltonian:  $(J \approx 1 \sim 2 \text{ K and } D \approx 1.4 \text{ K})$ 

$$H = \frac{J}{3} \sum_{\langle ij \rangle} \sigma_i \sigma_j + \frac{Da^3}{2} \sum_{ij} \sigma_i \sigma_j \begin{bmatrix} \mathbf{e}_i \cdot \mathbf{e}_j \\ \mathbf{r}_{ij}^3 \\ \mathbf{r}_{ij}^5 \end{bmatrix} - \frac{3(\mathbf{e}_i \cdot \mathbf{r}_{ij})(\mathbf{e}_j \cdot \mathbf{r}_{ij})}{r_{ij}^5} \end{bmatrix}$$
  
exchange  
interaction  $E_{\text{ex}}$  dipolar  
interaction  $E_{\text{dip}}$ 

• For spins  $\{\sigma_i\}$  satisfy ice rules:  $\sum_{i \in \boxtimes} \sigma_i = 0$  for all  $\boxtimes$ .  $\Rightarrow E_{\text{ex}}(\{\sigma_i\}) = \text{const.}$ 

However, Why is the dipolar energy also almost the same,  $E_{\text{dip}}(\{\sigma_i\}) \approx \text{const } ??$ 

#### Dumbbell Picture

Why is the long-range dipolar interaction (almost) irrelevant?







- $H \Rightarrow \sum_{\substack{\alpha \\ \boxtimes}} \frac{v_0}{2} \stackrel{\circ}{\xrightarrow{}} replace each dipole \vec{d} by two equal independent charges \pm q separation of the bond length <math>q = d/a$   $\stackrel{\circ}{\xrightarrow{}} H_{quad} + H_{quad}$ • renormalize the onsize Colomb interaction so as to give the correct neuroinhour interaction between dipoles:
  - $Q_{\mathbf{a}} \equiv \sum_{i \in \alpha} \sigma_i : \text{magnetic} \underset{v(r_{ij}) \equiv}{\text{charge}} e^{\frac{\mu_0}{4} \frac{q_i \beta_j}{O_i}}_{a} \text{tetrahedron} \quad \substack{i \neq j \\ v_o(\frac{\mu}{a})^2 = \frac{J}{3} + 4\frac{D}{3}(1 + \sqrt{\frac{2}{3}})}_{i = j},$
- Low-energy states:  $Q_{\alpha} = 0 \Rightarrow 2$ -in-2-out ice rule !
- Thanks to projective equivalence, this dumbell model reproduces the energy.
- $H_{quad} \sim \mathcal{O}(1/r_{\alpha\beta}^5)$  responsible for the low-T magnetic ordering.

#### Fractionalization: Magnetic Monopoles

(C. Castelnovo *et al.* Nature 2008) (Z. Nussinov *et al.* PRB 2006).



#### **Observing monopoles**

Dirac quantization: monopole charge:  $q_m = \frac{2\mu}{a} \approx \frac{q_D}{8000}$ .  $(eq_D = \frac{nh}{\mu_0}, n = 0, 1, 2, \cdots)$  $\Rightarrow$  Difficult (but not impossible) to observe directly.



#### Is the ice phase really 'featureless' ?



#### **Emergent Gauge structure**



#### **Emergent Coulomb phase**

Ice rule  $\Rightarrow \sum_{i \in \boxtimes} \sigma_i = 0$  (Ising spins) (Isakov *et al.* PRL 2004) (C. Henley PRB 2004)  $\Rightarrow \nabla \cdot \mathbf{B}(\mathbf{r}) = 0$  (coared-grained approx)

• emergent magnetostatics (Coulomb phase):

 $Z = Z_0 \int \mathcal{D}\mathbf{B}(\mathbf{r}) \prod_{\mathbf{r}} \delta\left(\nabla \cdot \mathbf{B}(\mathbf{r})\right) e^{-\int d^3 \mathbf{r} \, \frac{k}{2} |\mathbf{B}(\mathbf{r})|^2}$  $= Z_0' \int \mathcal{D}\mathbf{A}(\mathbf{r}) e^{-\int d^3 \mathbf{r} \frac{k}{2} |\nabla \times \mathbf{A}(\mathbf{r})|^2} \quad (\mathbf{B} = \nabla \times \mathbf{A})$ 

• Dipolar correlation:  $\langle B_i(\mathbf{r}) B_j(0) \rangle \approx \frac{4\pi}{k} \frac{3x_i x_j - r^2 \delta_{ij}}{r^5}.$   $\Rightarrow \text{Pinch-pint singularity}$ 

in structure factor:



#### Transition out of Coulomb phase

- Residual quadrupolar interaction  $\Rightarrow$  1st-order transition to a q = (001) magnetic order.
  - (R. Melko et al. 2001)
- Coupling to electrons (RKKY):  $\Rightarrow$  1st-order transition to a q = (001) magnetic order
  - (A. Ikeda & H. Kawamura, 2008)
- Magnetic field  $H \parallel [001]$   $\Rightarrow$  Kasteleyn transition to a q = 0 magnetic order.
- Uniaxial pressure  $\Rightarrow$  infinite-order transition to a q = 0 magnetic order.

(L. Jaubert et al. 2008, 2010)





#### Other 'ice' models: Klein S-1/2 model

• Klein spin-1/2 model on pyrochlore lattice:

(Z. Nussinov, C. D. Batista, B. Normand, & S.A. Trugman, PRB 2007)

$$H_K = J \sum_{\boxtimes} \mathcal{P}^{S_{\boxtimes}=2} + \cdots$$

• Ice rules  $\Rightarrow$  hard-core dimer covering:



• Fractionalized excitations: Deconfined spinons (monopoles) in 3D.

#### Other 'ice' systems: magnetic nano-arrays

• Artificial spin ice in 2D square and kagome lattices:





#### checkerboard ice



1 µm





#### More artificial ices:

A. Libál, C. Reichhardt,& C. J. Olson Reichhardt (PRL 2006)

 charged colloidal particals on arrays of optical traps





• vortices in nanostructured superconductors

A. Libál, C. J. Olson Reichhardt,& C. Reichhardt (PRL 2009)



#### Kagome vs. Pyrochlore spin ice • Minimizing magnetic charges Q: $\Rightarrow Q_{\triangle} = \pm 1$ in kagome vs. $Q_{\boxtimes} = 0$ in pyrochlore • Two-stage ordering in kagome spin ice. (G.-W. Chern *et al.*, PRL 2011) ordering of magnetic charges $Q_{\triangle}$ Rln2 1 14 $\begin{array}{c} \mathrm{S}(\mathrm{T}) & \mathrm{U} & \mathrm{I} \mathrm{Mol}^{-1}\mathrm{K}^{-1} \\ \mathrm{S} & \mathrm{U} & \mathrm{U} \\ \mathrm{S} & \mathrm{U} \end{array}$ 1.75 (a) 1.50 $12 \cdot$ 0.8 1.25 1.00 $\ln 2$ C/T (J mol<sup>-1</sup>K<sup>-2</sup>) ` 10 -0.75 0.6 0.50 8 0.25 $s_{\rm ice} \approx 0.5014$ 0.00 .ż .3 6 0.4 2'37 8 10 4 · Т $(\mathbf{K})$ 0.2 2 0 0 0.00 0.25 0.50 0.75 1.00 1.25 1.50 1.75 2.00 0.01 0.1 100 10 1 T (K) T/D magnetic magnetic crossover crossover transition transition to spin ice to spin ice

# New frustrated systems on cold-atom optical lattices



- Increasing  $U/t \Rightarrow$  Mott-insulating phase of cold atoms.
- Loading *spinless* (or polarized) fermions: 2 atoms per site.

 $\Rightarrow \text{Remaining degrees of freedom:} \\ \text{localized orbitals } (p_x, p_y, \text{ and } p_z). \end{cases}$ 



• Exchange Hamiltonian:  $|\mathbf{\hat{n}}\rangle = \hat{n}_x |p_x\rangle + \hat{n}_y |p_y\rangle + \hat{n}_z |p_z\rangle$  $H_{ij} = -J \left[ P_i^{\mathbf{\hat{n}}} (1 - P_j^{\mathbf{\hat{n}}}) + (1 - P_i^{\mathbf{\hat{n}}}) P_j^{\mathbf{\hat{n}}} \right]$ 

 $P^{\mathbf{\hat{n}}} = |\mathbf{\hat{n}}\rangle\langle\mathbf{\hat{n}}|$  : Projection operator of active orbital along  $\mathbf{\hat{n}}$ 

#### **2D** Square and honeycomb lattices

• Square lattice: AF Ising model

$$H = J \sum_{\langle ij \rangle} \tau_i \tau_j \qquad \tau_i = \begin{cases} +1 & : p_x \\ -1 & : p_y \end{cases}$$

Néel order

• Honeycomb lattice: Quantum 120° model:







- residual entropy:  $s_0 \approx 0.59 \, k_B$
- disordered orbitals.
- exponential orbital correlation:

 $C_{\tau}(L) \sim \exp(-L/\xi)$ 

#### P-band Mott insulator on diamond lattice

• there are 4 distinct n.n. bonds:

 $\hat{\mathbf{n}}_0 = [111], \ \hat{\mathbf{n}}_1 = [1\overline{1}\overline{1}],$  $\hat{\mathbf{n}}_2 = [\overline{1}1\overline{1}], \ \hat{\mathbf{n}}_3 = [\overline{1}\overline{1}1]$ 

• orbital projectors along the 4 neighbors:

$$P_m = \frac{1}{3} \left( \hat{1} + \sqrt{3} \,\vec{\mu} \cdot \hat{\mathbf{n}}_m \right)$$

• pseudovector  $\vec{\mu} = (\mu_x, \mu_y, \mu_z)$ 

 $\Rightarrow$  Quantum 'tetrahedral' Hamiltonian for pseudovectors  $\{\vec{\mu}_i\}$ :

$$H_{\text{ex}} = J \sum_{m=0}^{-} \sum_{\langle ij \rangle \parallel \hat{\mathbf{n}}_{m}} \left( \vec{\mu}_{i} \cdot \hat{\mathbf{n}}_{m} \right) \left( \vec{\mu}_{j} \cdot \hat{\mathbf{n}}_{m} \right)$$

 $([\mu_{\alpha},\mu_{\beta}] \neq 0, \text{ for } \alpha \neq \beta)$ 



$$\mu_x = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}$$
$$\mu_y = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}$$

 $\mu_z = \left( \begin{array}{rrr} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{array} \right)$ 

#### Mean-field ground states

• Gutzwiller products ansatz:

$$|\Psi\rangle = \prod_{i=1}^{N} \otimes |\theta_i, \phi_i\rangle$$

• Single-site wavefunction:

- $\begin{aligned} |\theta,\phi\rangle &= \sin\theta\cos\phi|p_x\rangle + \sin\theta\sin\phi|p_y\rangle + \cos\theta|p_z\rangle \\ &\Rightarrow \langle \vec{\mu} \rangle = \left(\sin 2\theta\sin\phi, \ \sin 2\theta\cos\phi, \ \sin^2\theta\sin 2\phi\right) \end{aligned}$
- Monte Carlo minimization of  $E_{\rm mf} = \langle \Psi | H_{\rm eff} | \Psi \rangle$ :
  - Macroscopic degeneracy of Gutzwiller ground states.
  - pseudovector  $\langle \vec{\mu}_i \rangle = \pm \vec{x}, \pm \vec{y}$ , and  $\pm \vec{z}$  in the ground state.

 $\mu^x$  -

 $\mu^z$ 

- Each bond  $\langle ij \rangle$  has exact energy  $E_{ij} = -J/3$ .

#### Ground-state structure:

• Orbitals in the ground states:

 $|\pm \vec{x}\rangle = |p_y\rangle \pm |p_z\rangle \quad |\pm \vec{y}\rangle = |p_z\rangle \pm |p_x\rangle \quad |\pm \vec{z}\rangle = |p_x\rangle \pm |p_y\rangle$ 

 $\sigma_i^3 = +1$  $\sigma_i^2 = -$ 

 $\cdot z$ 

 $\sigma_i^0 = +1$ 

 $\sigma_i^1 = -1$ 

- - Ising variables for the 4 bonds attached to site  $\mathbf{r}_i$ :

$$\sigma_i^m = \sqrt{3} \langle \hat{\mathbf{n}}_m \cdot \vec{\mu}_i \rangle = \pm 1$$
$$(m = 0, 1, 2, 3)$$

• Ground-state constraint:

 $\sigma_i^m \sigma_j^m = -1, \quad \forall \langle ij \rangle$ ('ice' rule for orbitals !)

#### Exact Eigenstates

- Ground-state conditions:
  - 1. Cubic anisotropy:  $\langle \vec{\mu}_i \rangle = \pm \vec{x}, \pm \vec{y}, \text{ and } \pm \vec{z}.$
  - 2.  $\{\sigma_i^m\}$  satisfy Ice rules:  $\sigma_i^m \sigma_j^m = -1 \quad \forall \langle ij \rangle.$



• The extensively degenerate Gutzwiller states:

 $|\Psi\rangle = |+\vec{x}\rangle_1 \otimes |-\vec{y}\rangle_2 \otimes |-\vec{x}\rangle_3 \otimes |+\vec{z}\rangle_4 \otimes |+\vec{y}\rangle_5 \otimes \cdots$ 

are *exact* eigenstates of the orbital exchange Hamiltonian ! $H_{\rm ex}|\Psi\rangle=-\frac{2}{3}NJ|\Psi\rangle$ 

also confirmed by exact diagonalization.

## Mapping to pyrochlore spin ice:

- How to characterize the degeneracy of the Gutzwiller ground states ?
- *Pyrochlore* is the "medial" lattice of the diamond structure  $\Rightarrow$  placing spins at the *bond midpoints* of diamond lattice.



• a 1-to-1 mapping of orbital ground state and spin-ice state on pyrochlore !

#### Orbital Coulomb phase (orbital ice)

• The six cubic directions of  $\langle \vec{\mu} \rangle$  are mapped to the six 2-in-2-out ice states.



- Pauling estimate of entropy density:  $s_0 = k_B \ln \frac{3}{2} \approx 0.405 k_B$
- Dipolar-like power-law orbital correlations: The pseudovector plays the role of the emergent 'magnetic field':

$$\mathbf{B}(\mathbf{r}_i) \approx \pm \langle \vec{\mu}_i \rangle \quad \Rightarrow \quad \text{ice rule: } \nabla \cdot \mathbf{B}(\mathbf{r}) = 0$$
$$\langle \mu_i(\mathbf{r}) \, \mu_j(0) \, \rangle \sim \pm \frac{3x_i x_j - r^2 \delta_{ij}}{r^5} \sim \frac{1}{r^3}$$

#### Orbital correlation functions

- Classical Monte Carlo simulations with non-local 'loop' updates on pyrochlore spin ice
- Correlation function  $C_{\mu}(r) = \langle \vec{\mu}(\mathbf{r}) \cdot \vec{\mu}(0) \rangle$



### **Conclusion and Outlook**

- A orbital analog of ice.
- a first example of orbital Coulomb phase.
- could possibly be realized in optical diamond lattice.
- *Exact* many-body ground states which form a degenerate ice-manifold of a nontrivial quantum Hamiltonian.

 $|\Psi\rangle = |+\vec{x}\rangle_1 \otimes |-\vec{y}\rangle_2 \otimes |-\vec{x}\rangle_3 \otimes |+\vec{z}\rangle_4 \otimes |+\vec{y}\rangle_5 \otimes \cdots$ 

- $\Rightarrow$  Experimental signature of orbital ice: structure factor, time-of-flight measurement ...
- $\Rightarrow (exact) Elementary excitations ?$ Monopole-like quasiparticles ?