Title: Spin Slush in an Extended Spin Ice Model

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Abstract: We introduce a new classical spin liquid on the pyrochlore lattice by extending spin ice with further neighbour interactions. This disorder-free spin model exhibits a form of dynamical heterogeneity with extremely slow relaxation for some spins while others fluctuate quickly down to zero temperature. We thus call this state "spin slush", in analogy to the heterogeneous mixture of solid and liquid water. This behaviour is driven by the structure of the ground state manifold that extends the two-in/two-out ice states to include branching structures built from three-in/one-out, three-out/one-in and all-in/all-out defects. Distinctive liquid-like patterns in the spin correlations serve as a signature of this intermediate range order. Finally, we discuss possible applications to materials as well the effects of quantum tunneling.

Spin slush in an extended spin ice model

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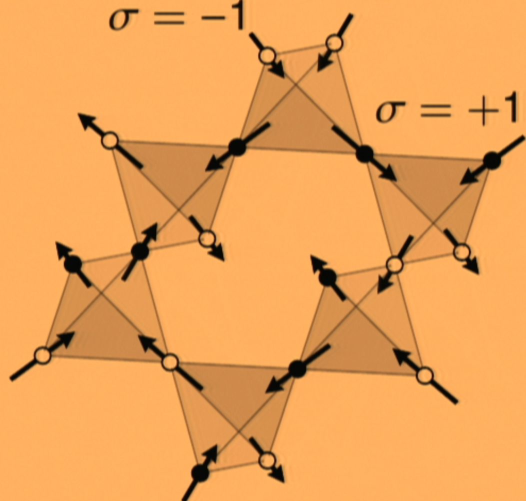
Spin ice (simplest model)



- Macroscopic ground state degeneracy, residual entropy $S/k_B \sim 0.202$ per spin
- Classical spin liquid down to $T = 0^+$

- Nearest-neighbour anti-ferromagnetic Ising model on pyrochlore lattice

$$H = J \sum_{\langle ij \rangle} \sigma_i \sigma_j = 2J \sum_{\Phi} Q_{\Phi}^2 + \text{const.}$$

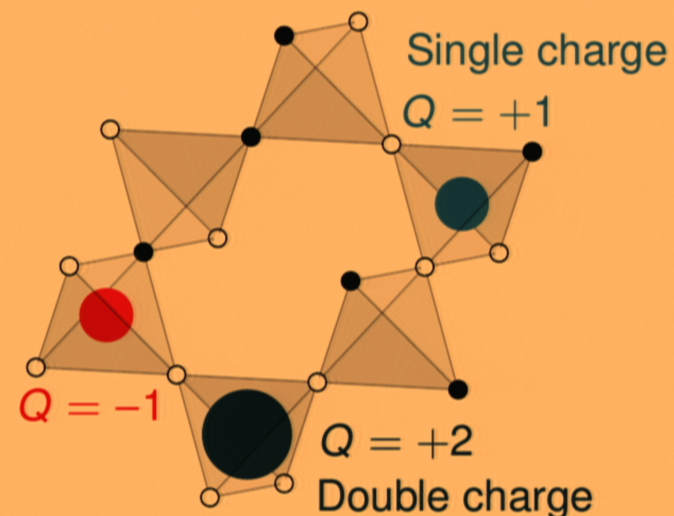
- Charge defined on tetrahedron

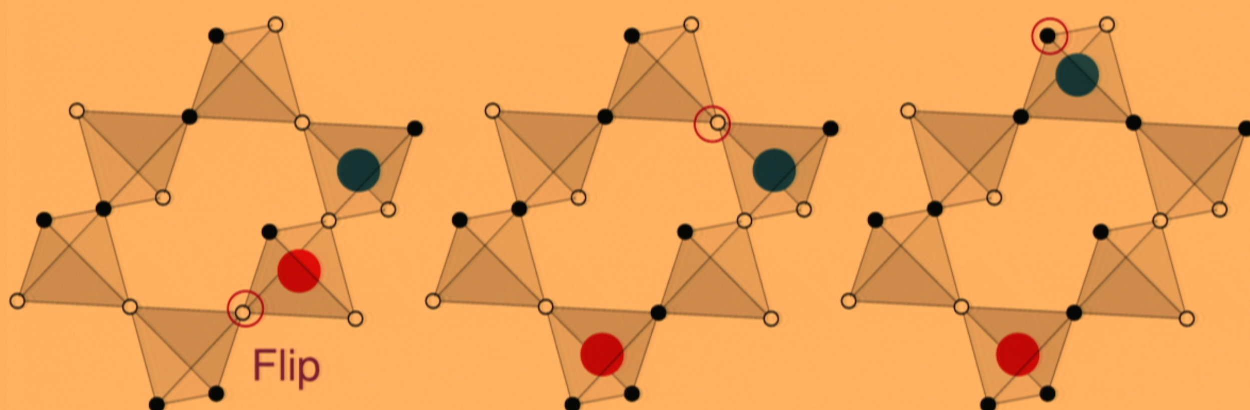
$$Q_{\Phi} = \frac{1}{2}(-1)^{\Phi} \sum_{i \in \Phi} \sigma_i$$

- Ground states with $Q = 0 \rightarrow$ ice rules (two-in,two-out)

Defects

- Lowest energy defects are single charges $Q = \pm 1$
- Cost $\sim 4J$ to create pair; free to move (deconfined)
- Leaves behind Dirac string of flipped spins





Magnetic correlations

- Coarse-grained description in terms of magnetization **B**

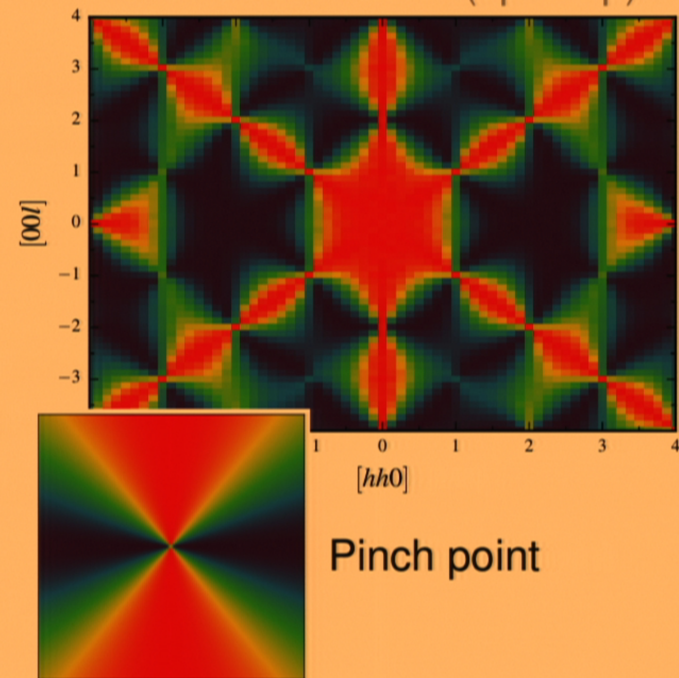
$$F \sim \frac{K}{2}|\mathbf{B}|^2 + \frac{J}{T}(\boldsymbol{\nabla}\cdot\mathbf{B})^2$$

- As $T \to 0$ one has constraint $\boldsymbol{\nabla}\cdot\mathbf{B} = 0$ enforcing the ice rules
- Power-law spin correlations:

$$\langle B_\mu(0) B_\nu(\mathbf{r})\rangle \sim \frac{1}{|\mathbf{r}|^3}(\delta_{\mu\nu} - 3\hat{r}_\mu\hat{r}_\nu)$$

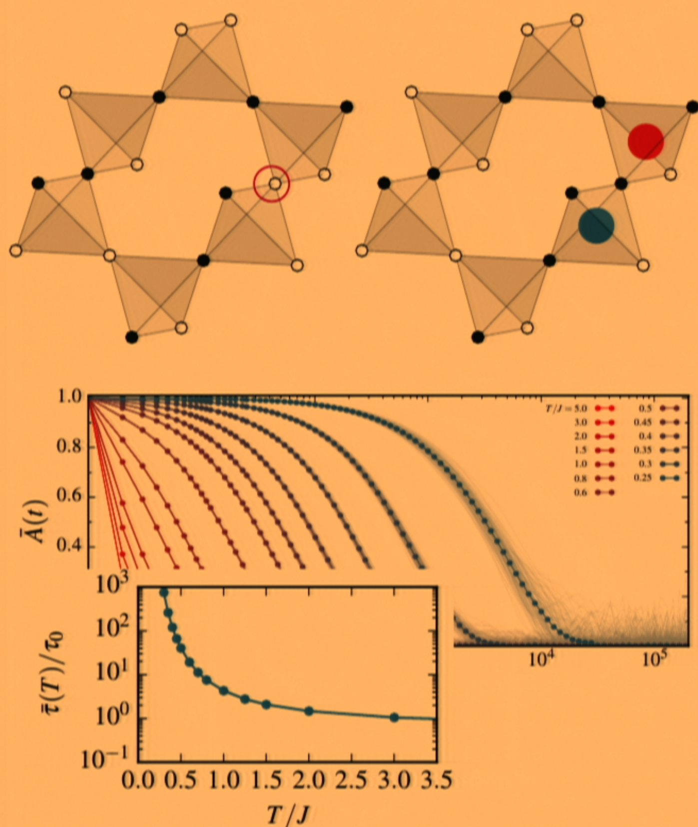
- Manifests as pinch-points in structure factor $\sim (\delta_{\mu\nu} - \hat{k}_\mu\hat{k}_\nu)$

Structure factor (spin-flip)



- **B**-field mediates entropic Coulomb interaction ($\propto T$) defect charges

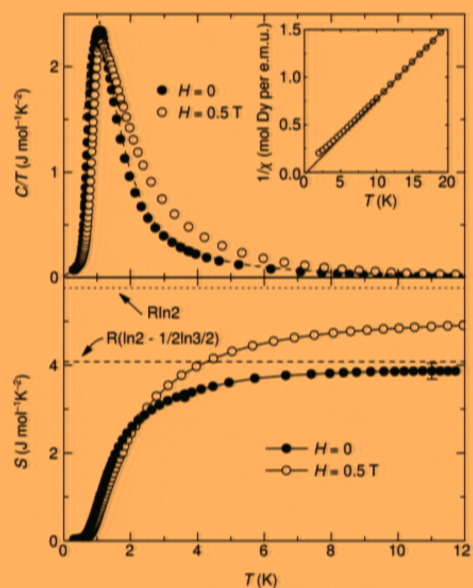
Dynamics

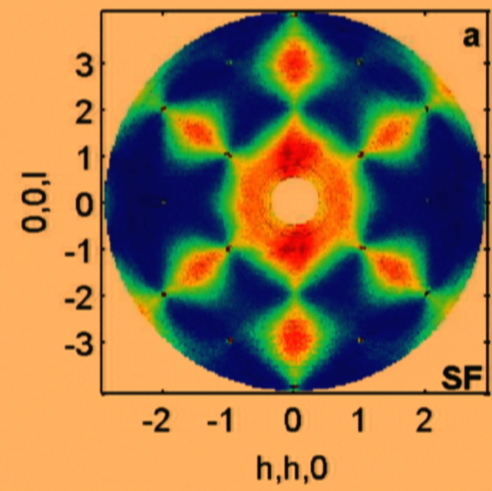


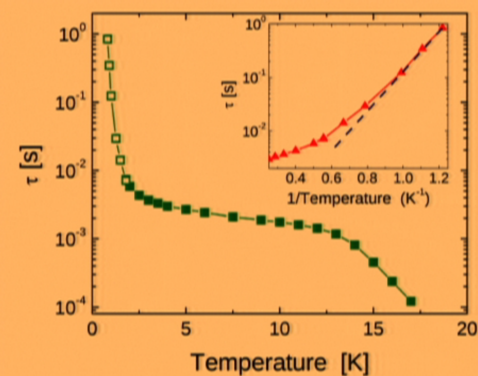
- Energy barriers between ice states
- A spin-flip always creates a pair of opposite charges pairs at cost $\sim O(J)$
- Expect exponential slow down in dynamics at low temperature
- Characteristic time scale $\tau \sim e^{2J/T}$ (Arrenhius)
- Homogeneous freezing

Experiments

- Two canonical examples: $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$
- Long-range dipolar interactions important







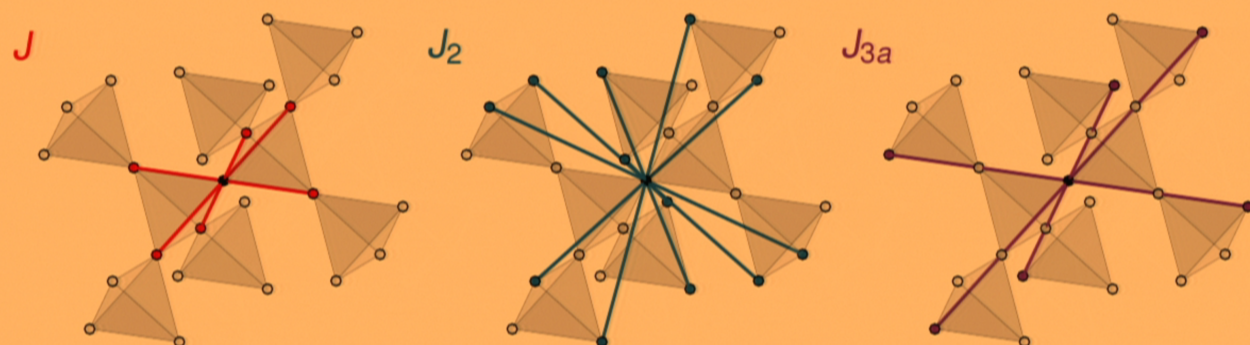
Fennell (2009), Ramirez (1999), Snyder (2004), ...

Extending spin ice

- Add second- and third-neighbour interactions (two nearest neighbour bonds)

$$H = J \sum_{\langle ij \rangle} \sigma_i \sigma_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \sigma_i \sigma_j + J_{3a} \sum_{\langle\langle\langle ij \rangle\rangle\rangle_a} \sigma_i \sigma_j$$

- Can be natural: super-exchange, virtual crystal field, etc

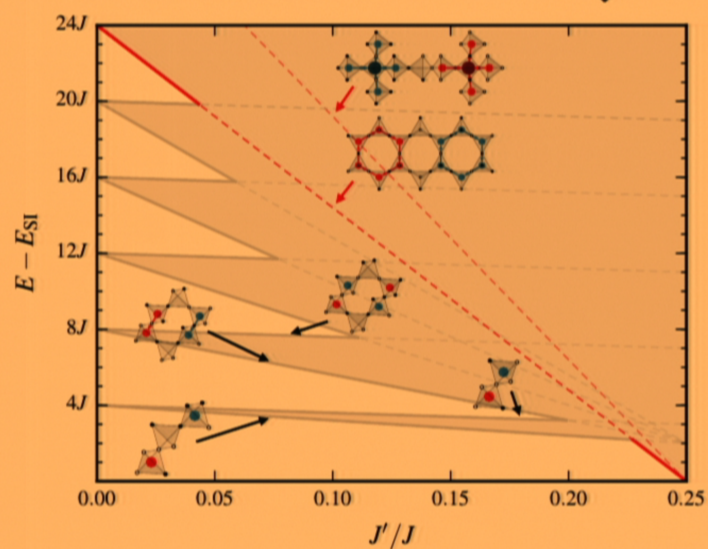


Generically this lifts the ice degeneracy

Extending spin ice (cont.)

- Special line: if $J_2 = J_{3a} \equiv J'$ the ice degeneracy is *preserved* (extended spin ice)

$$H_{\mathrm{ESI}} = 2(J - 2J') \sum_{\diamond} Q_{\diamond}^2 - 4J' \sum_{\langle \diamond, \diamond' \rangle} Q_{\diamond} Q_{\diamond'} - N(J - J')$$



- Excited states are modified: nearest-neighbour attraction of like charges
- Collapse of infinite set of excited states at $J' = J/4$.

What happens there?

End point

- Will study the $J' = J/4$ end point
- Model is square of total spin on *pairs* of tetrahedra

$$H = \frac{J}{2} \sum_{\Diamond\!\!\Diamond} \left(\frac{1}{2} \sum_{i \in \Diamond\!\!\Diamond} \sigma_i \right)^2 + \text{const.}$$

- Odd number of spins per tetrahedron pair, so ground states satisfy

$$P_i \equiv \frac{1}{2} \sum_{j \in \Diamond\!\!\Diamond_i} \sigma_j = \pm \frac{1}{2}$$

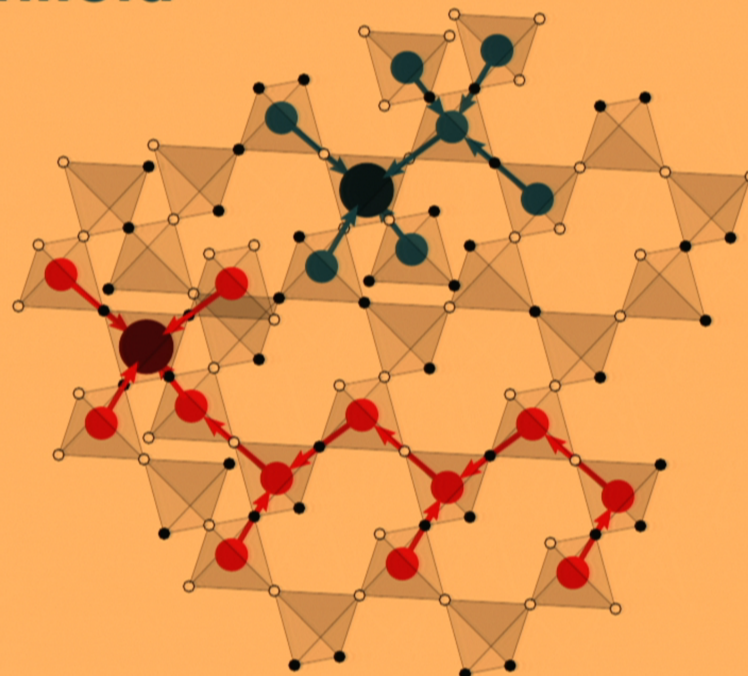
- Constraint $P_i = \pm\frac{1}{2}$ satisfied for all spin ice states $\rightarrow$ extension of spin ice manifold

How to construct these states?

Ground state manifold

Rules: Specify all non-ice parts tetrahedra:

- Single charge rule: The minority spin of a single charge, $Q = \pm 1$, must be connected to a charge of the same sign.
- Double charge rule: A double charge $Q = \pm 2$ must be surrounded by single charges of the same sign.

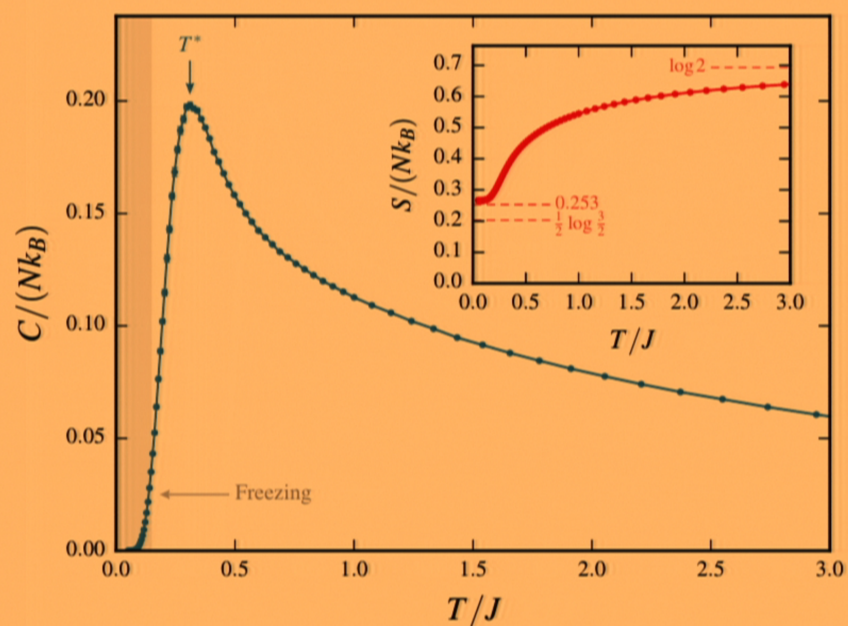


- Neighbour rule: A single charge, $Q = \pm 1$, cannot neighbour single charges of opposite sign

Fill remainder with compatible ice states

Finite temperatures?

Thermodynamics

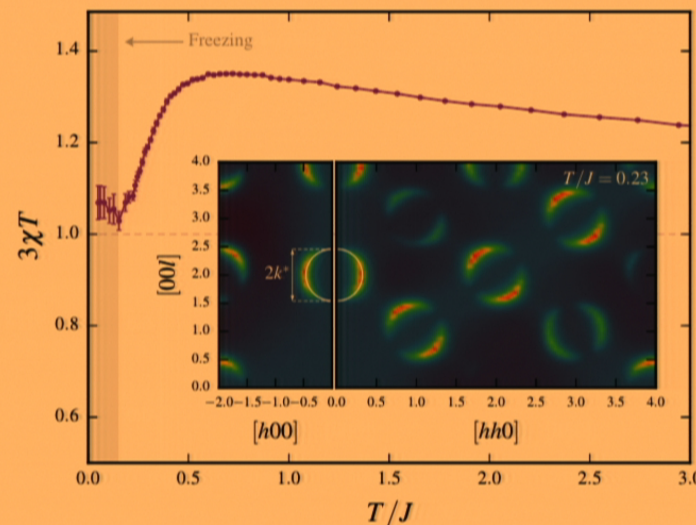


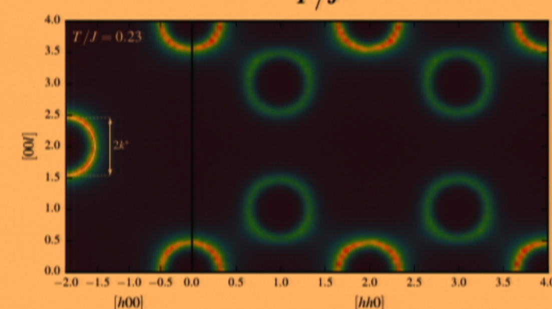
Magnetic properties?

- Simulate using classical Monte Carlo methods
- Cooperative paramagnet down to low temperature
- *No* evidence of any ordering
- Higher residual entropy than spin ice, $S/k_B \sim 0.253$
- Strong dynamical freezing below $T \sim 0.1J$.

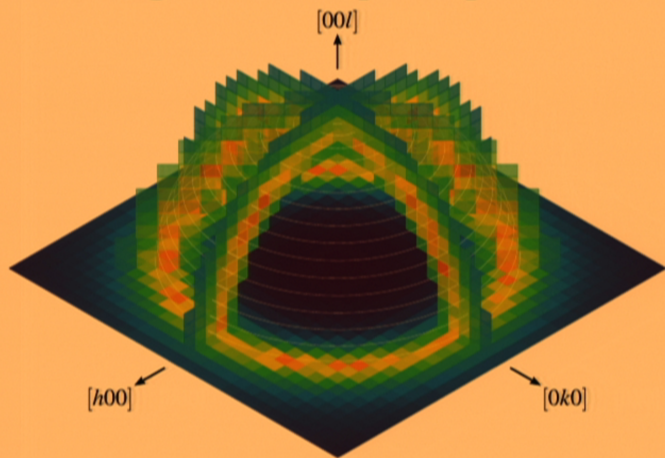
Magnetic properties

- "Curie"-like susceptibility, $3\chi T \sim 1$ as $T \to 0$
- Striking spherical patterns in structure factor (spin and charge)
- Centered on locations of pinch-points in spin ice
- Define intermediate scale magnetic correlations with wave-vector $|\mathbf{k}| \sim 0.5(2\pi/a) \equiv k^*$





Magnetic properties (cont.)



- Roughly, at end point (in **k** space):

$$\sim -\frac{1}{2}\beta J(\mathbf{k}\cdot\mathbf{B})^2 + \beta A|\mathbf{k}|^4|\mathbf{B}|^2 + \cdots$$

- Minimum in **k** is nearly spherical surface

- Charge-charge correlations are maximal on spheres in reciprocal space
- Can be partially understood in coarse-grained theory

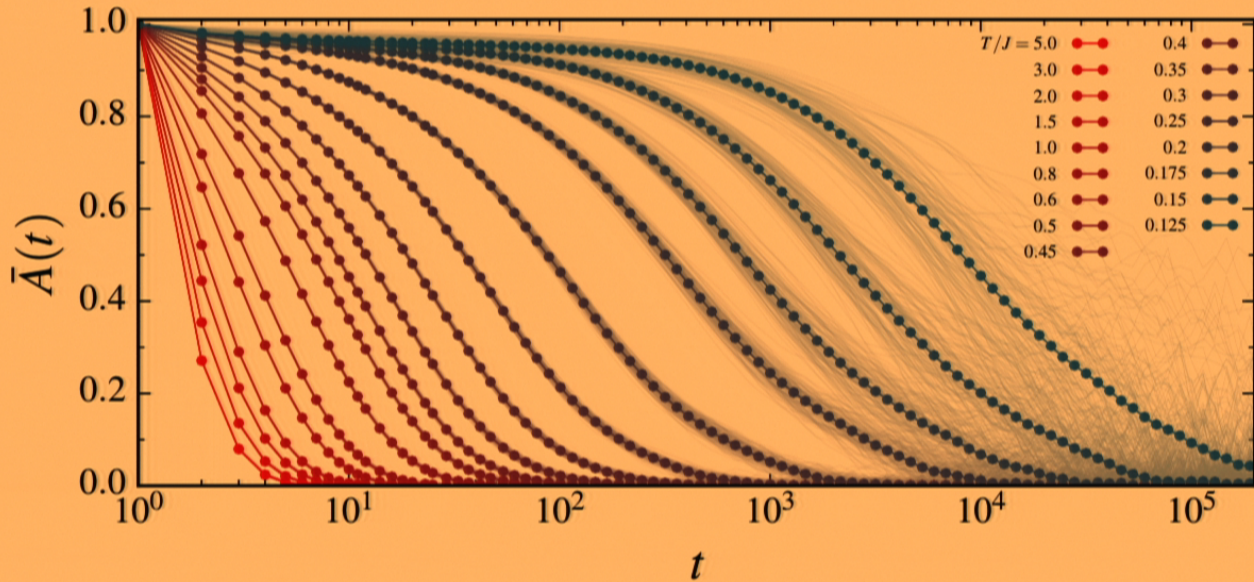
$$\sim \beta J(\nabla\cdot\mathbf{B})^2 - 6\beta J'(\nabla\cdot\mathbf{B})^2 + O(\nabla^4)$$

- For $J' < J/6$ one has $\nabla\cdot\mathbf{B} = 0$ (spin ice)
- For $J' = J/6$ has *Lifshitz transition*

Nature of freezing?

Heterogeneous dynamics

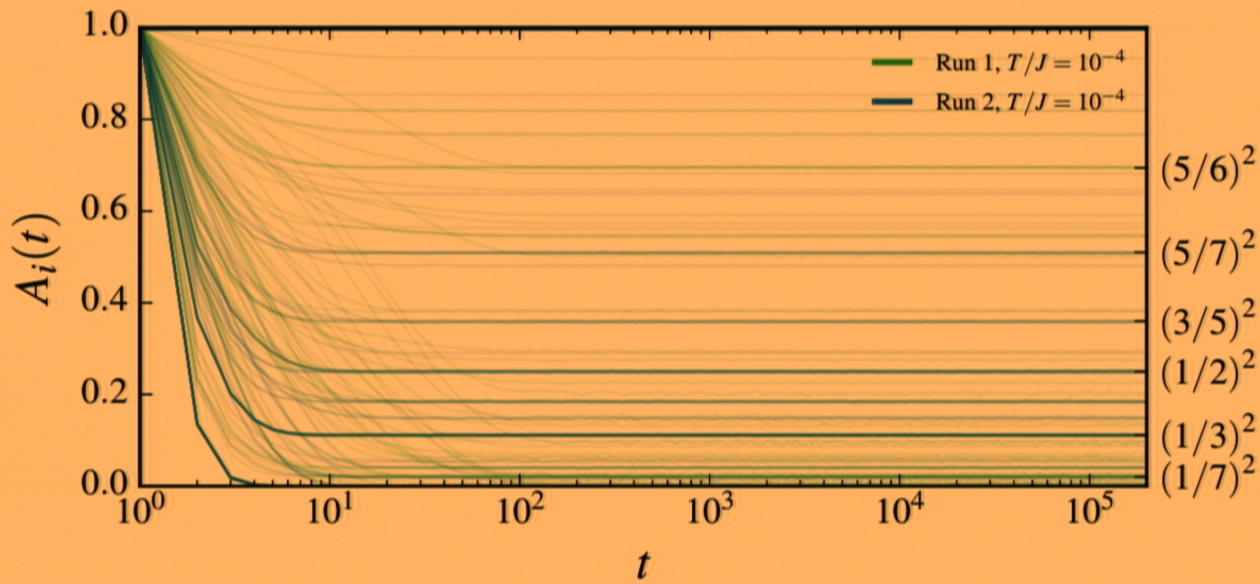
- Consider auto-correlation function $A_i(t) \equiv \langle \sigma_i(t_0)\sigma_i(t+t_0) \rangle$



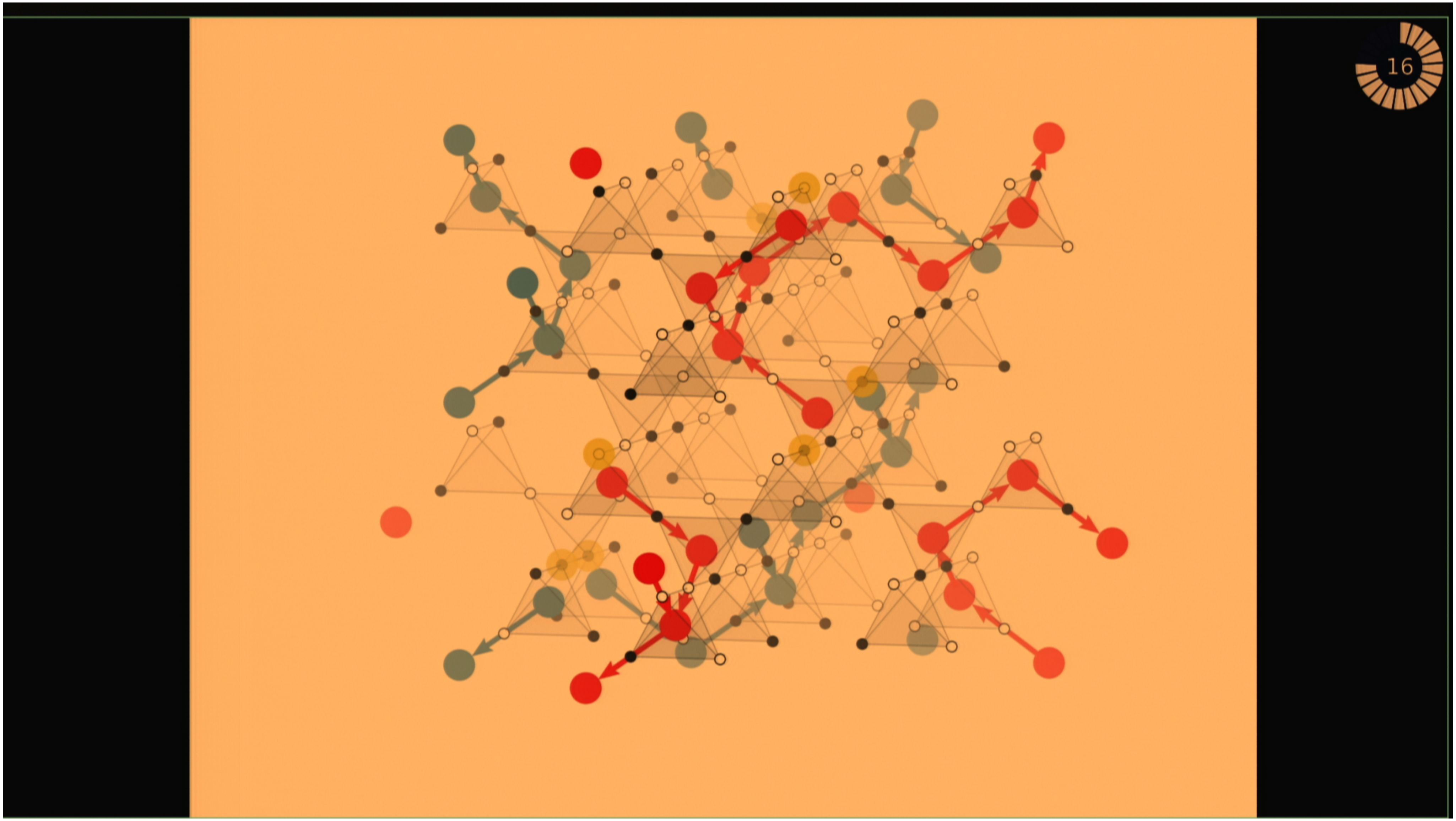
- Energy barriers slow down dynamics as $T \to 0$
- Not simple exponential $\sim e^{-t/\tau}$
- Slowing down is spatially heterogeneous

Heterogeneous dynamics (cont.)

- To see this clearly to go $T = 10^{-4}J$ – anneal to ground state, then look at dynamics



- Auto-correlations grow exponentially on some sites, remain $O(1)$ on others – "spin slush"
- Long-time values cluster on squares of simple fractions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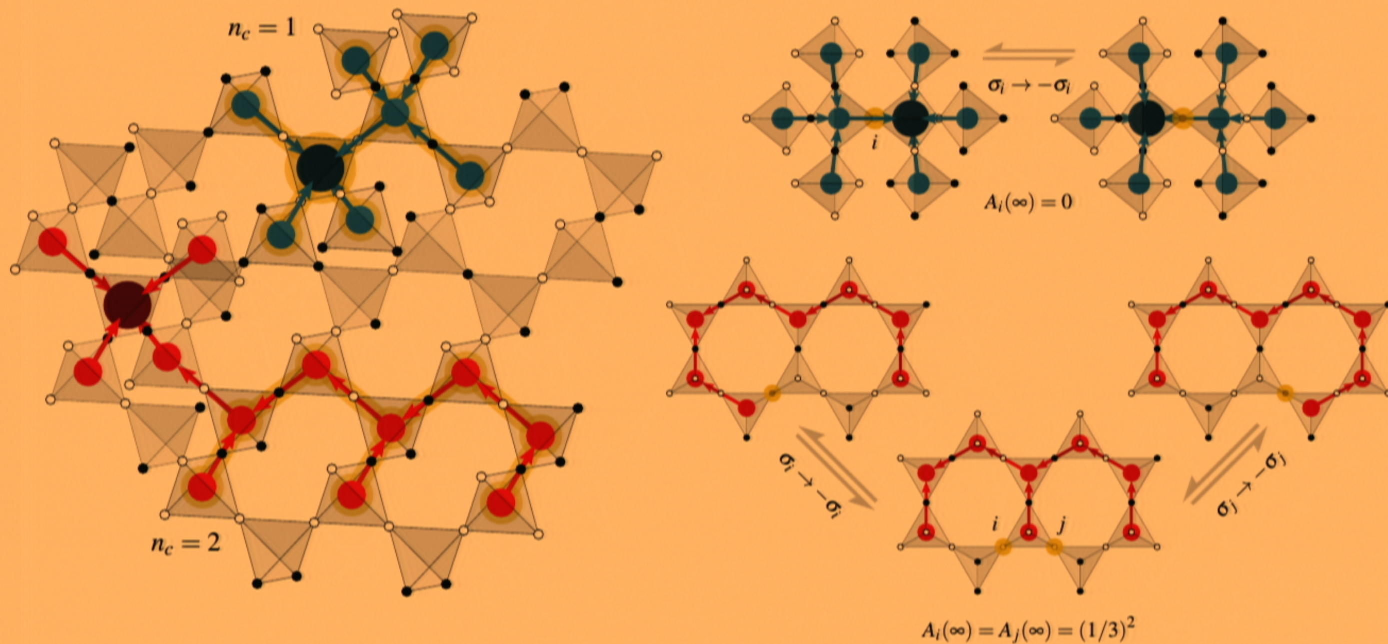
Dynamical clusters

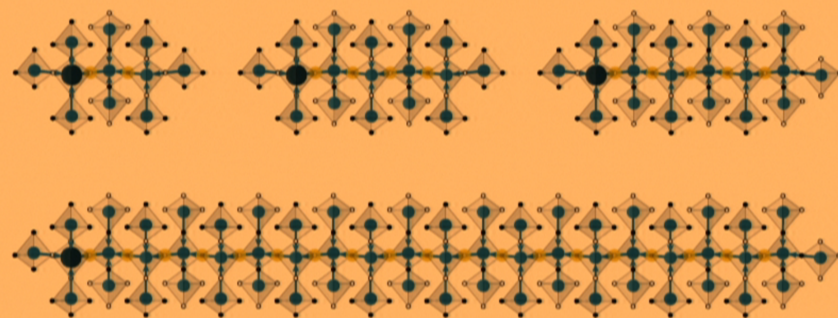
- Localized clusters with single-spin flip dynamics in spin-slush manifold



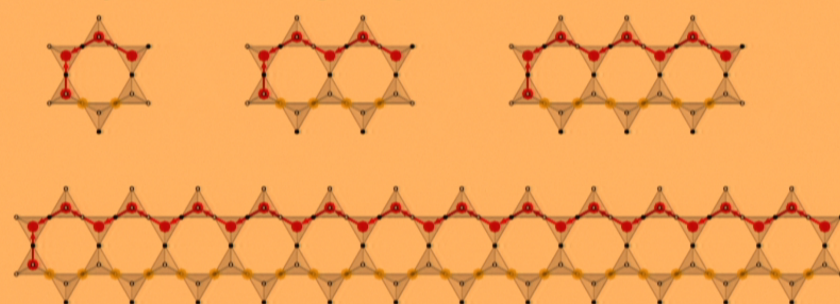
$$A_i(\infty) \sim \left(\frac{1}{n}\sum_{m=1}^{n}\sigma_i^{(m)}\right)^2 = \text{(time avg. magnetization)}^2$$

Dynamical clusters (cont.)

- Can be made arbitrarily large, can build from single- *and* double-charge structures

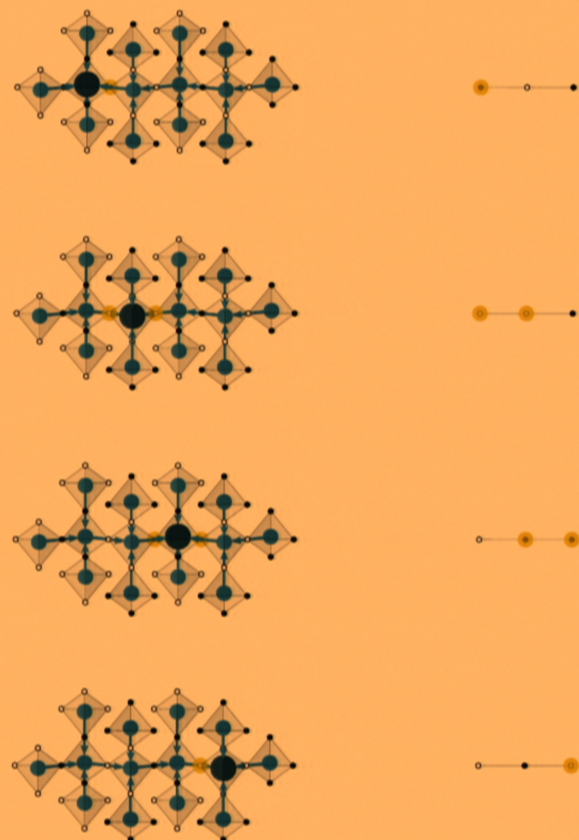


- Also exist for spin-exchange dynamics



Quantum spin slush?

- Add transverse field $\Gamma \sum_i S_i^x$ or exchange $J_\pm \sum_{\langle ij \rangle} (S_i^+ S_j^- + \mathrm{h.c.})$
- Spin ice → third or sixth order effect (weak)
- Spin slush → first order quantum effects
- Follows directly from presence of dynamical clusters
- A new *quantum* spin liquid?



Conclusions

- New classical spin liquid on pyrochlore lattice
- Ground states built from complex branching structures of spin ice charge – intermediate scale magnetic correlations
- Exhibits heterogeneous dynamics at low temperature
- New quantum spin liquid?
- Nearby phase space? Dipolar interactions? J_{3b}, J_4, etc?
- Nearby $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ phase, related to $Tb_2Ti_2O_7$?

Thank you for your attention

For details:

Jeffrey G. Rau and Michel J. P. Gingras, *Spin slush in an extended spin ice model*, arxiv:1603.02683 (should appear in Nat. Comm. soon)

See also related work: M. Udagawa et al. arXiv:1603.02872