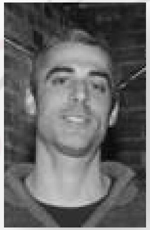
Quantum Spin Ice in Confined Geometries

Jeffrey G. Rau

University of Windsor

Lucas P. Montcalm



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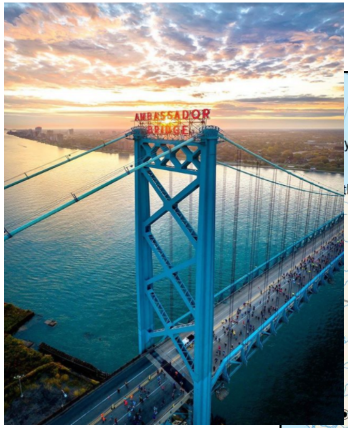


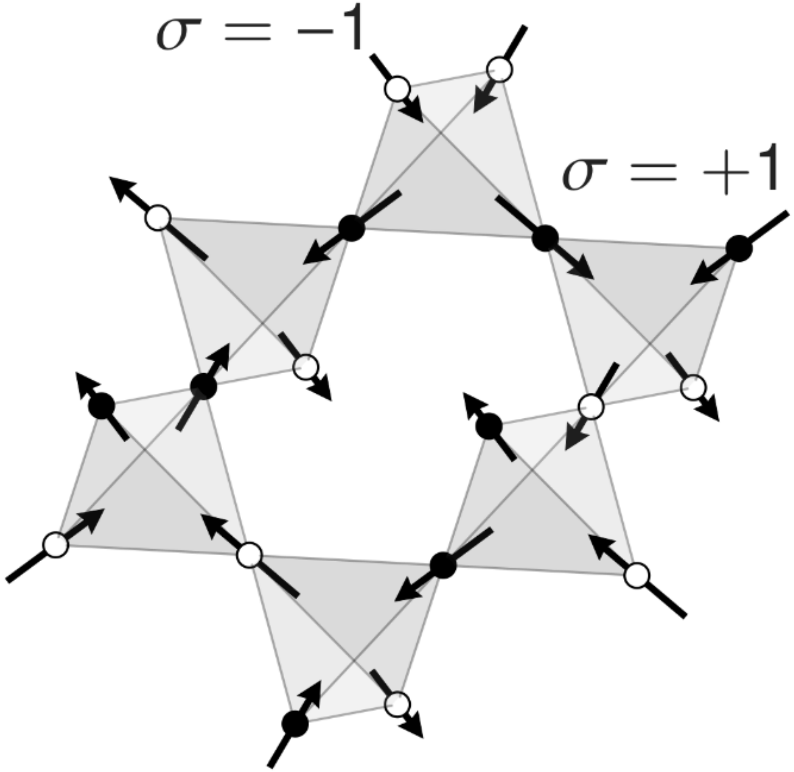
Windsor, Ontario, Canada

Classical Spin Ice

- *Simplest:* Nearest neighbour Ising model on the pyrochlore lattice

$$J \sum_{\langle ij \rangle} \sigma_i \sigma_j$$

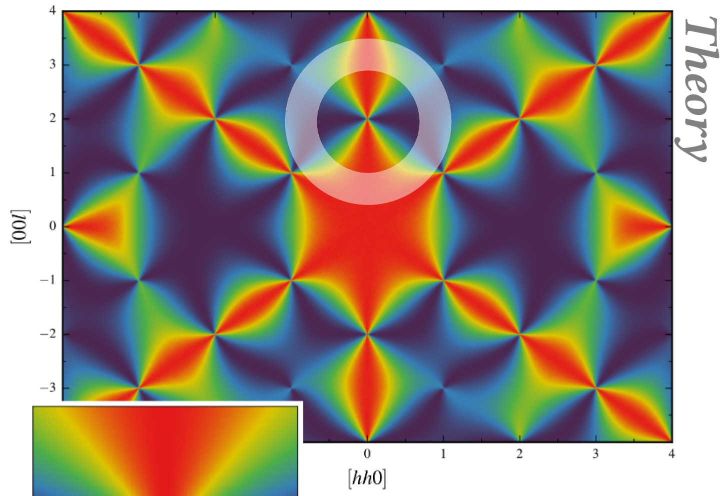
- Ground states are 2-in/2-out states
- Extensive degeneracy
- Excitations are magnetic monopoles (3-in/1-out)

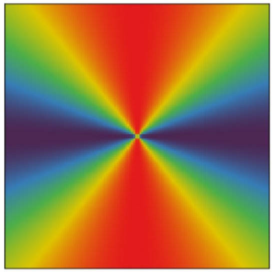


Spin Ice, Springer (2021)

Emergent Magnetostatics

Result from simplest version of



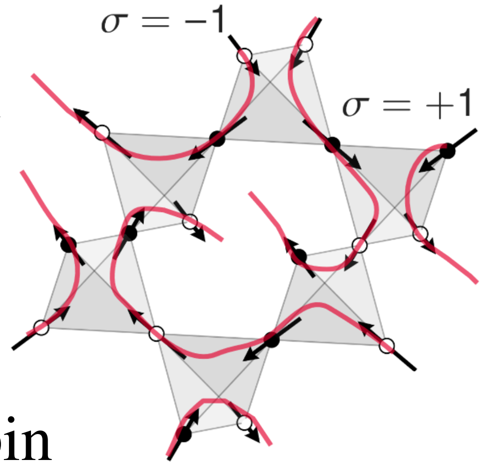


"Pinch-point"

- Ground state manifold formed of alternating spin **loops**
- Interpretation as emergent magnetostatics

$$\nabla \cdot \boldsymbol{B} = 0$$

- Implications: Algebraic spin correlations, **Pinch-point features**



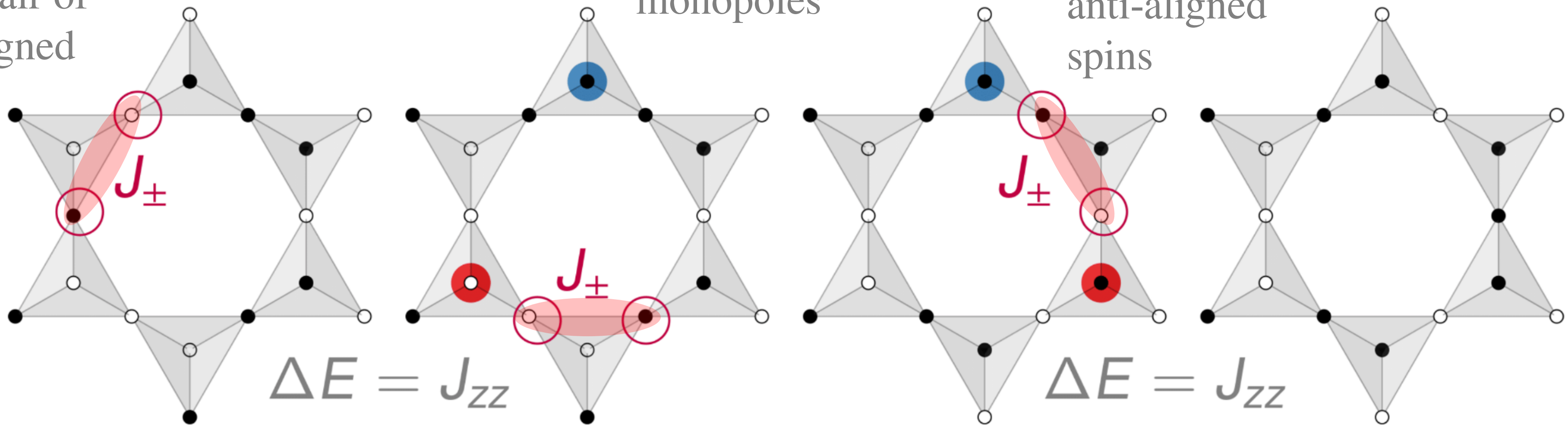
Spin Ice, Springer (2021)

Quantum Spin Ice

Swap pair of anti-aligned spins

Create pair of monopoles

Swap pair of anti-aligned spins



Swap pair of anti-aligned spins

Hop monopoles around hexagon

Return to new ice state

Hermele et al, Phys. Rev. B **69**, 064404 (2004)

Effective Hamiltonian

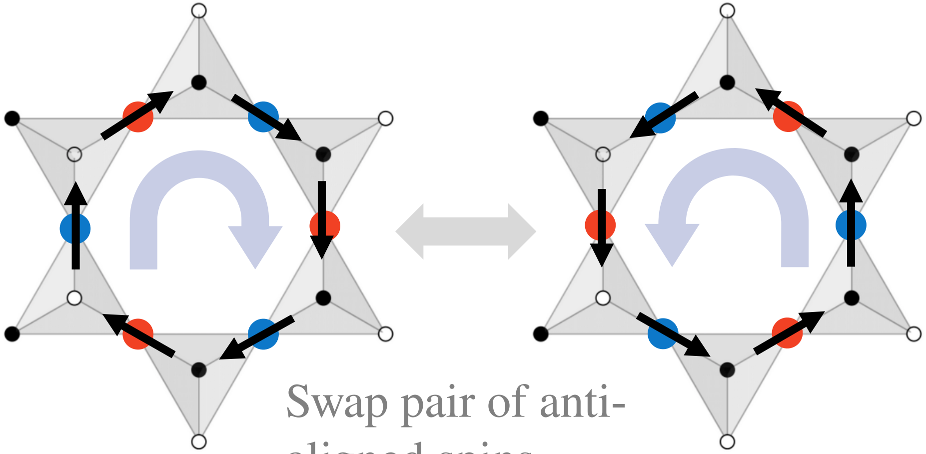
- Six-spin "loop flip" term in effective Hamiltonian

Flux lines become dynamical

$$H_{\text{eff}} = -\frac{2J_{\pm}^2}{J_{zz}}N - \frac{12J_{\pm}^3}{J_{zz}^2}\sum_{\text{hexagons}} P_{\text{ice}}\left(S_1^+S_2^-S_3^+S_4^-S_5^+S_6^- + \text{h.c.}\right)P_{\text{ice}}$$

- Energy scale of dynamics in ice

$$g \equiv \frac{12J_{\pm}^3}{J_{zz}^2} \ll J_{\pm} \ll J_{zz}$$

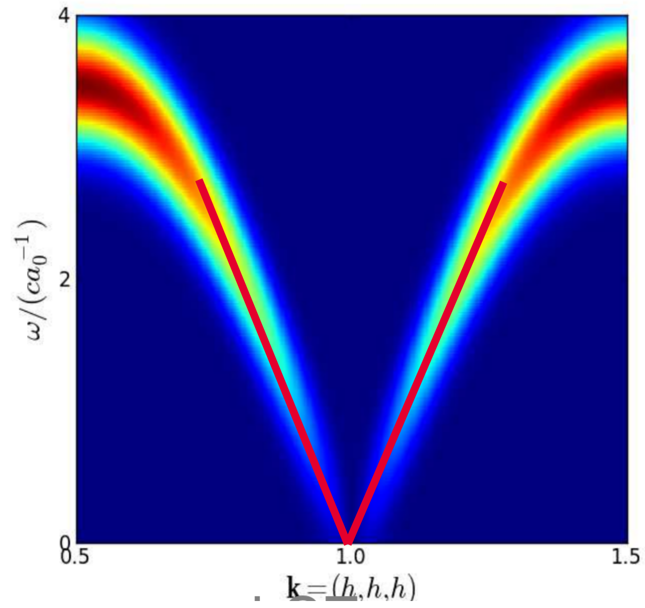


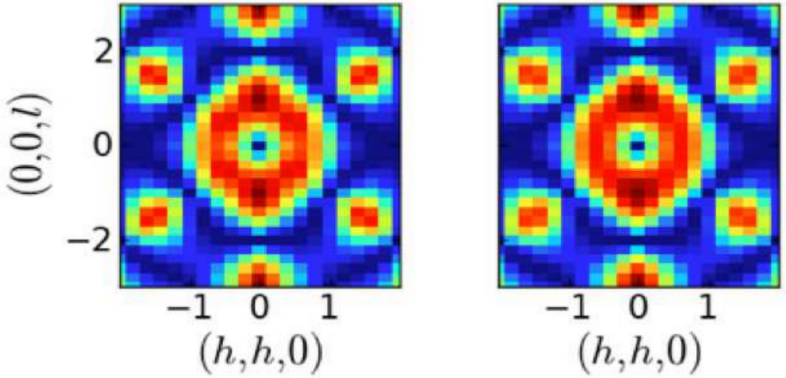
Swap pair of anti-aligned spins

Hermele et al, Phys. Rev. B **69**, 064404 (2004)

Low-energy physics

- This model has a description as a U(1) lattice gauge theory
- **Emergent quantum electrodynamics**
- Correlations power law now in space/time ("hollow" pinch-points)
- **Gapless photon excitation**
  - Visible (in principle) in dynamical structure factor

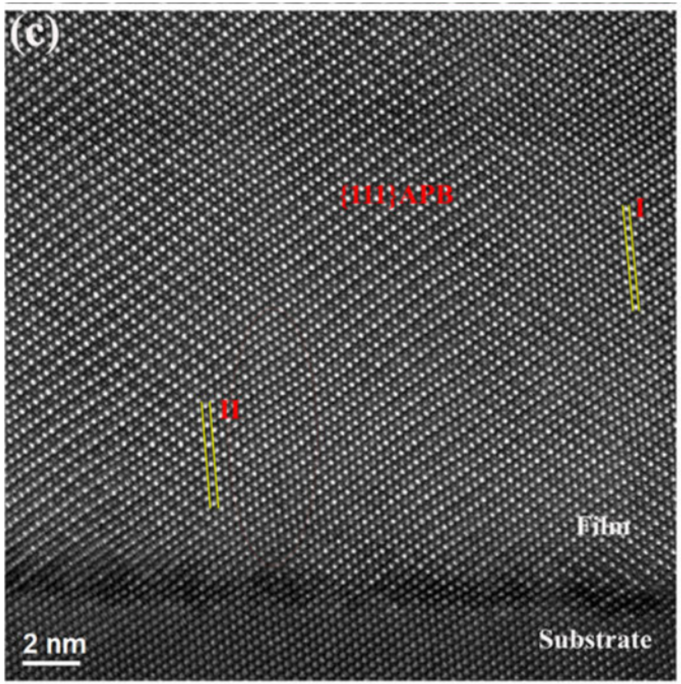




Benton, Sikora, Shannon, Phys. Rev. B **86** 075154 (2012)

Spin Ice Thin Films

- Experimentally can grow **thin films** of canonical spin ice materials
  - e.g. $Dy_2Ti_2O_7$, $Ho_2Ti_2O_7$
- Thickness is ~ tens of cubic unit cells
- Many possible geometries:
  - [001], [110], [111] directions
- Effect of substrate? Strain?

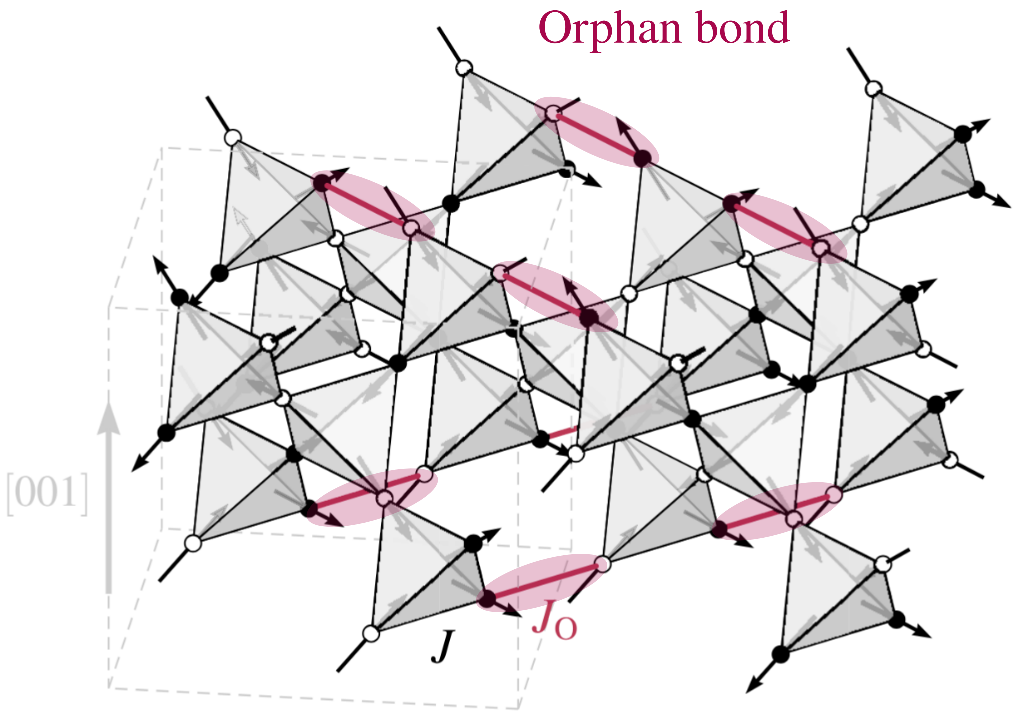


How does the physics of spin ice change in a confined geometry?

Bovo et al, Nat. Comm. **5** 3439 (2014), Leusnik et al, APL Materials 2 (2014), Bovo et al, Nat. Comm. **10** 1219 (2019), Barry et al, Phys. Rev. Mat. **3** 084412 (2019), Wen et al, J. Appl. Phys. **129**, 025302 (2021), Zhang et al, Nat. Comm. **14**, 1404 (2023), …, Xing et al, arxiv:2509.03082 (2025)

Classical Spin Ice Films

- Consider nearest-neighbour spin ice model in a thin film
- Surfaces are different than bulk; simplest termination
- Qualitatively different bonds at surfaces
    - **"Orphan" bonds** do not belong to a complete tetrahedron

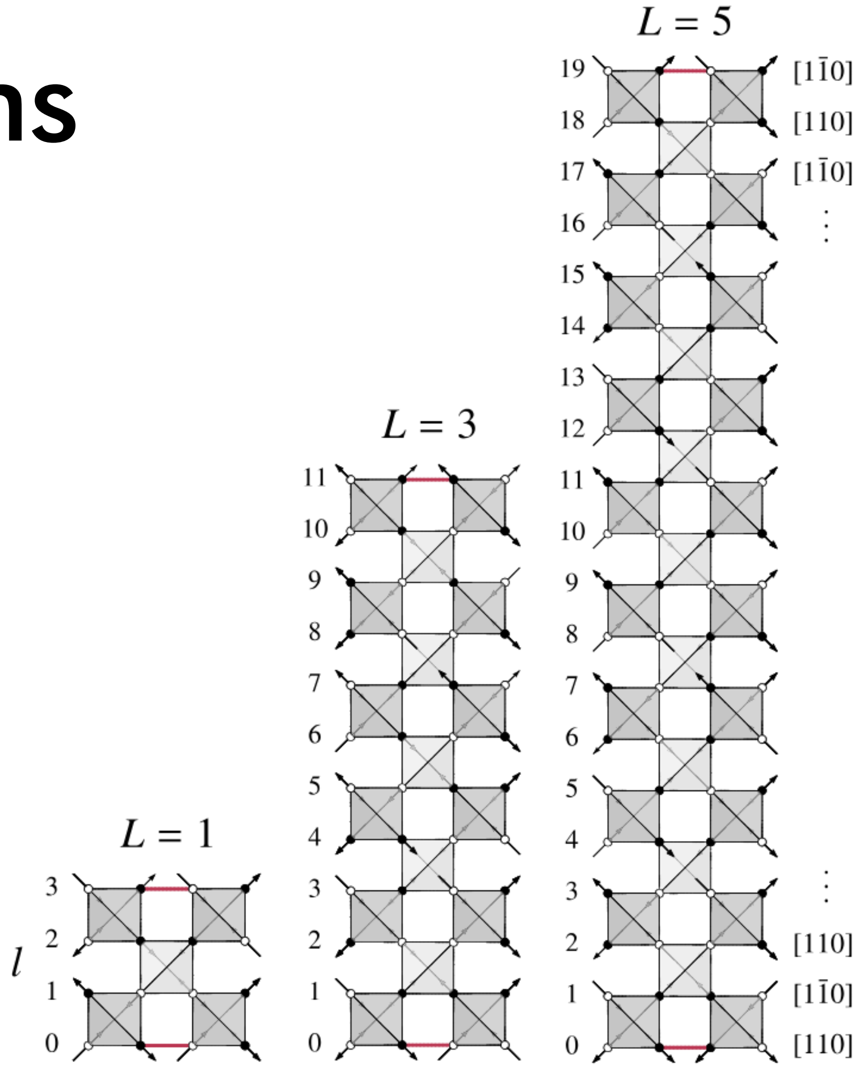


Chains along [1,1,0] on top surface, [1,-1,0] on bottom

Jaubert et al, Phys. Rev. Lett. **118**, 207206 (2017), Lantagne-Hurtubise, Rau, Gingras, Phys. Rev. X **8** 8747 (2018)

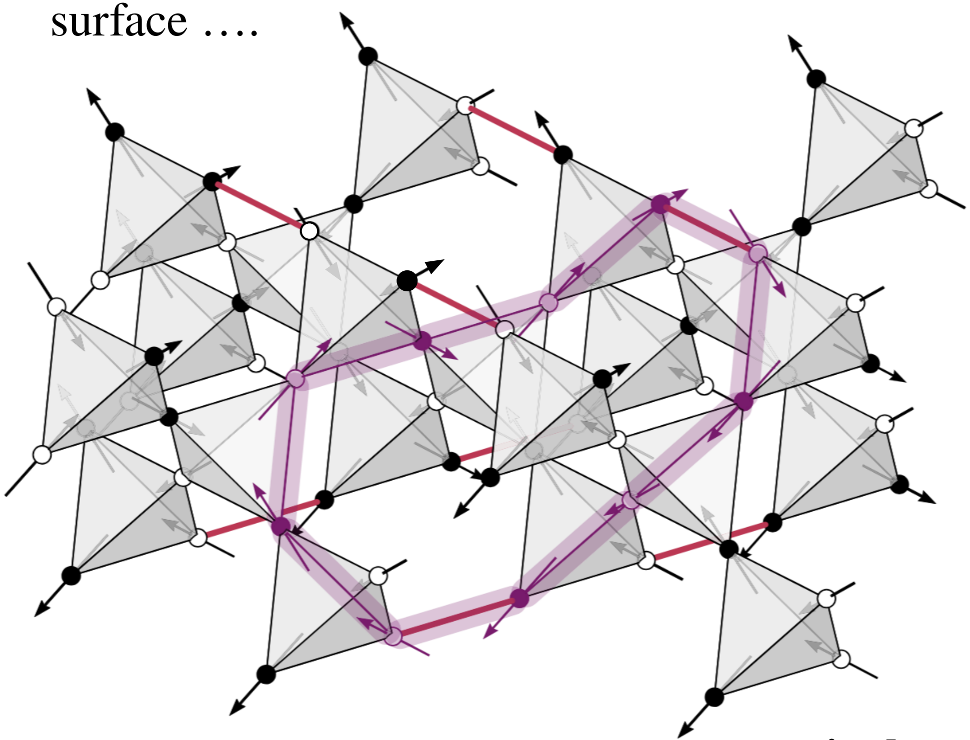
Boundary Conditions

- Since tetrahedron is incomplete, consider two possible couplings
  - **Antiferromagnetic orphans:** $J_O > 0$ matching bulk
  - **Ferromagnetic orphans:** $J_O < 0$ opposite to the bulk
- Antiferromagnetic bonds more natural
  - Details would depend on super-exchange physics, surface termination



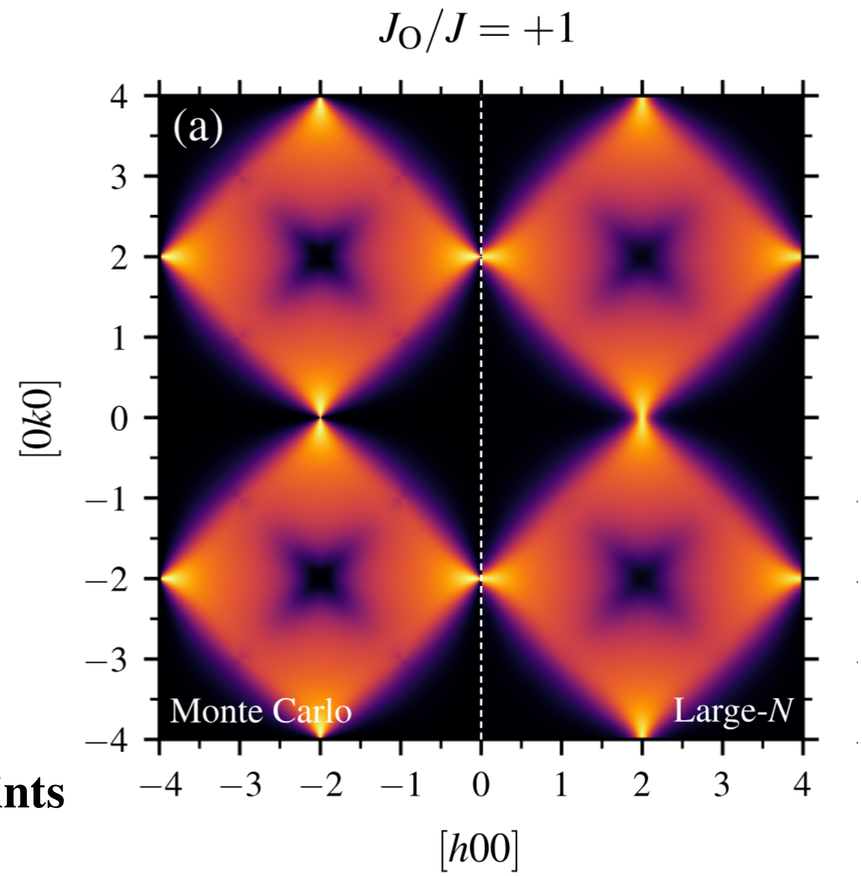
Antiferromagnetic Orphans

Loops **close** at surface ….



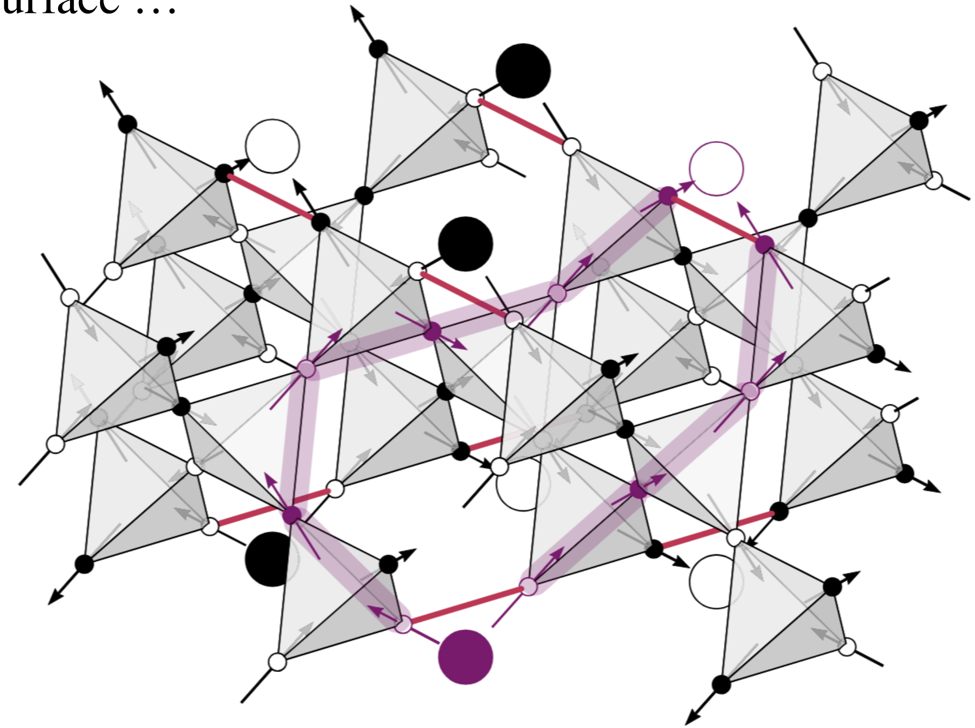
$J_O/J > 0$

… pinch-points survive!



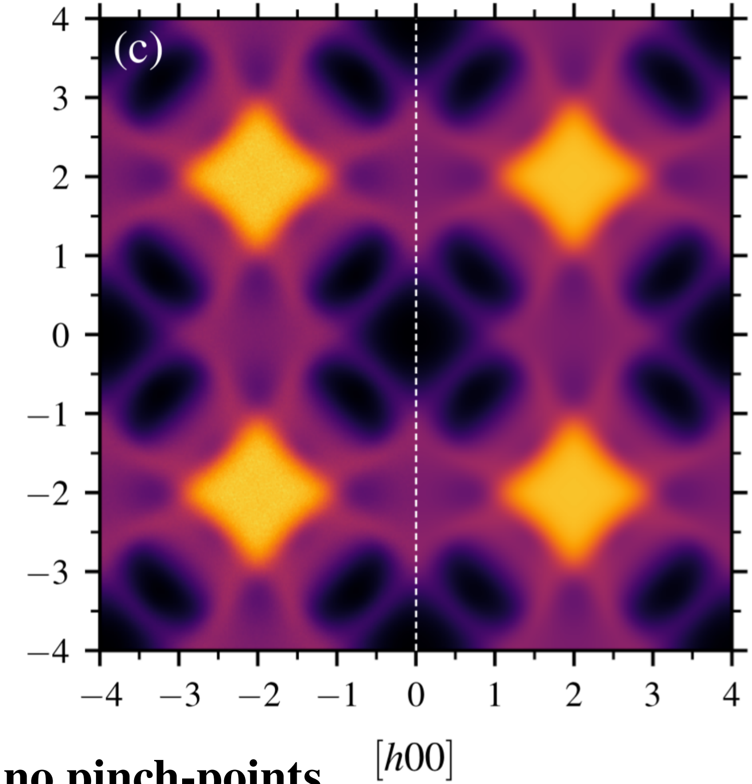
Ferromagnetic Orphans

Loops **paired** at
surface …



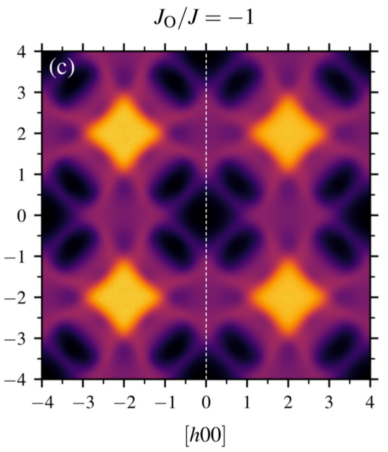
$J_O/J < 0$

$J_O/J = -1$

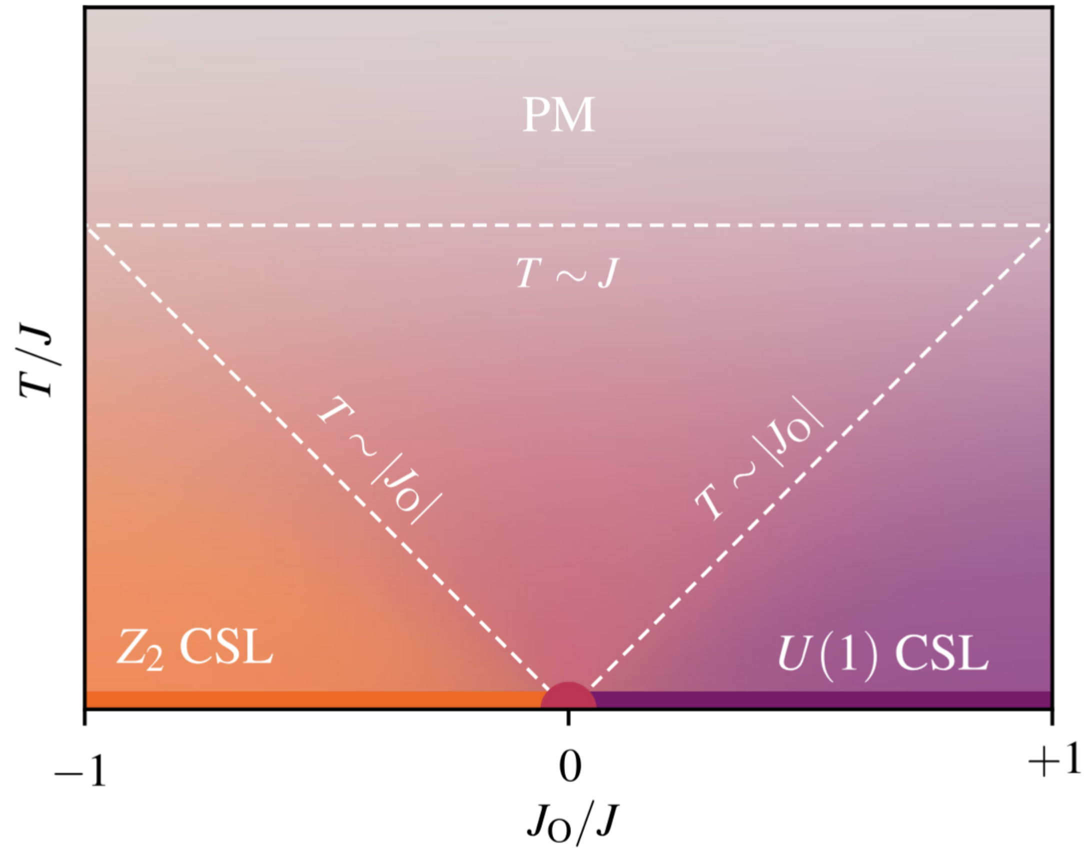


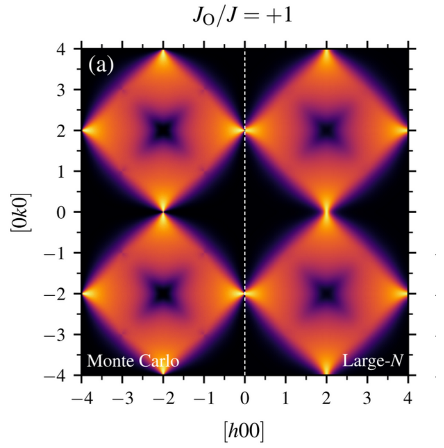
… **no pinch-points**

Phase Diagram



Classical Z_2 spin liquid

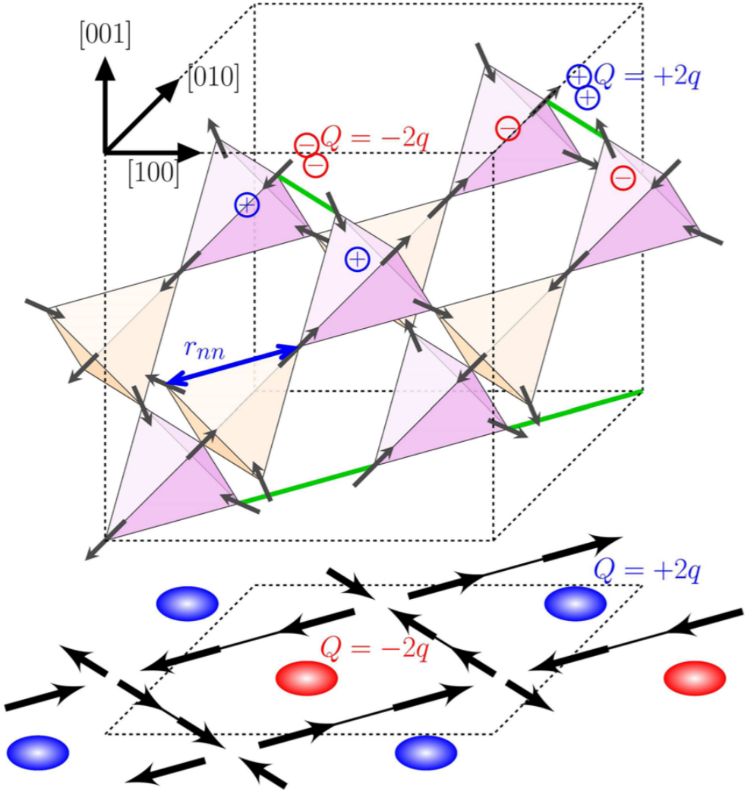




Classical U(1) spin liquid

Aside: Dipolar Spin Ice Thin Films

- Add dipolar interactions changes the surface physics
- Monopoles fluctuating at surface interact via an effective long-range Coulomb interaction
    - o "Dumbbell" picture
- **Charge ordering on surface**, checkerboard pattern

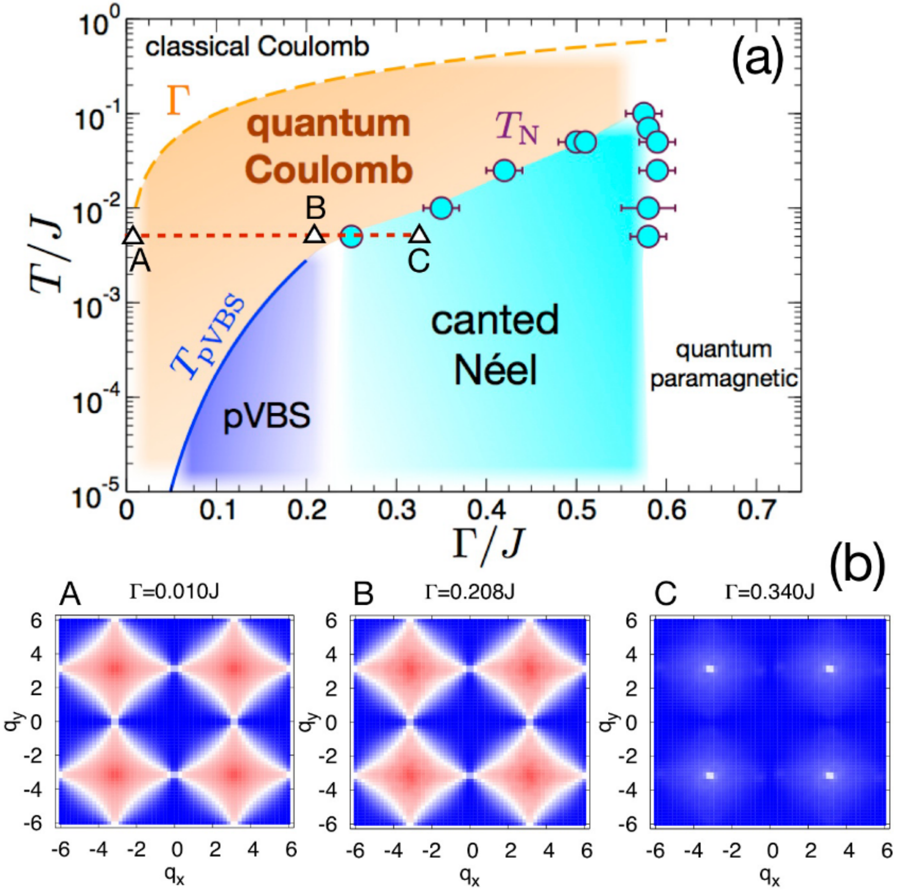


Jaubert et al, Phys. Rev. Lett. **118**, 207206 (2017)

Quantum Spin Ice Films

Aside: Instability

- 2D quantum spin ice is described by compact QED in 2+1D
- *Polyakov:* Unstable to proliferation of (gauge) monopoles
- **I'm going to ignore this:**
  - Intermediate regime in temperature where SL "survives"
  - Scale (should) vanish *exponentially* with thickness

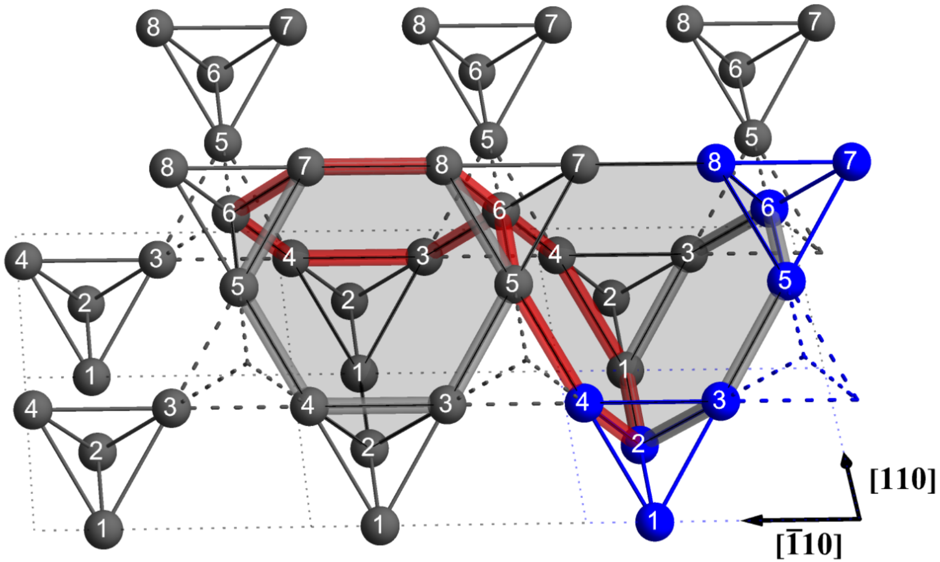


Henry & Roschilde, Phys. Rev. Lett. **113**, 027204 (2014)

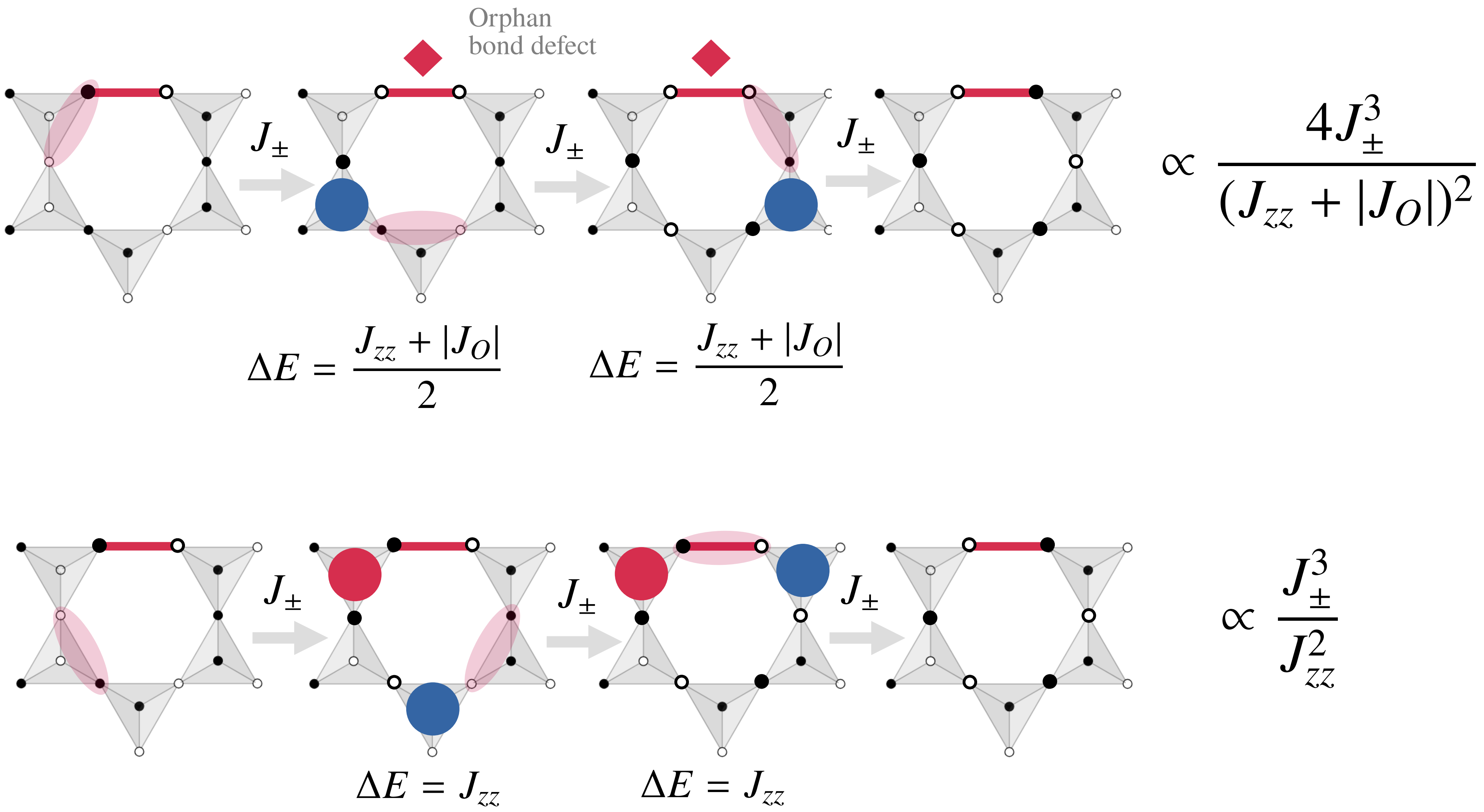
Effective Hamiltonian

- How does the effective Hamiltonian change where there are boundaries?
- At third-order still generate the ring-type tunnelling terms
  - *Thinnest film*: one type of hexagon
  - *Thicker films*: weak modulation at surface

$L = 1$



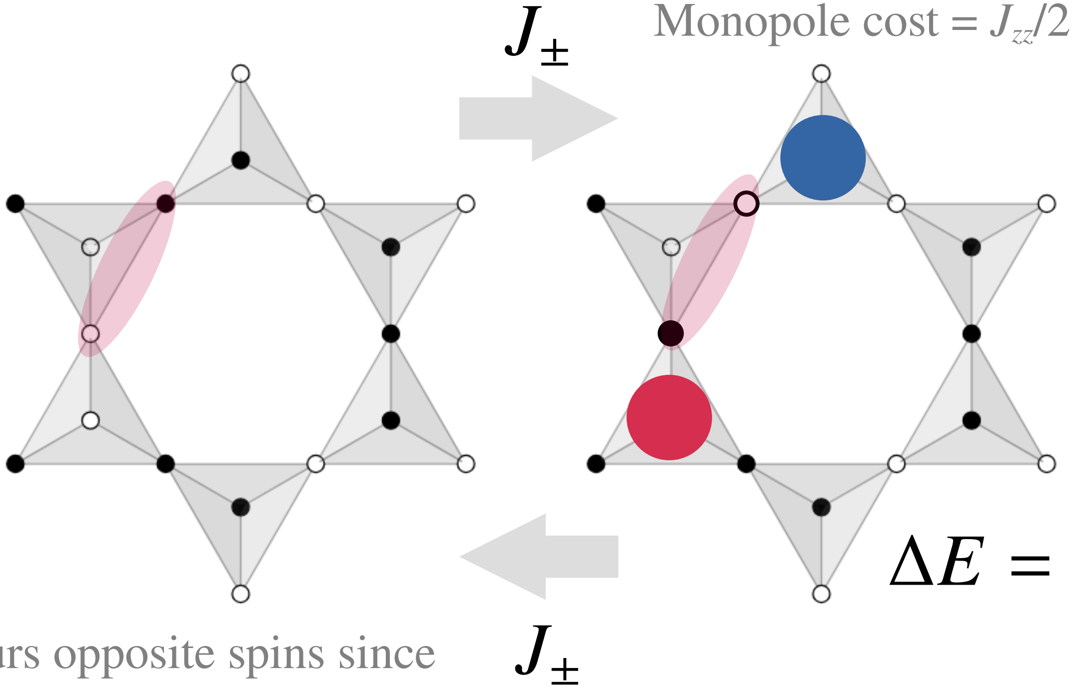
$$\approx -\frac{12J_{\pm}^3}{J_{zz}^2} P_0 \left(\sum_{\hexagon} S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- \right) P_0 + \text{h.c.}$$



Orphan
bond defect
$J_\pm$
$J_\pm$
$J_\pm$
$\propto \frac{4J_\pm^3}{(J_{zz}+|J_O|)^2}$
$\Delta E = \frac{J_{zz}+|J_O|}{2}$
$\Delta E = \frac{J_{zz}+|J_O|}{2}$
$J_\pm$
$J_\pm$
$J_\pm$
$\propto \frac{J_\pm^3}{J_{zz}^2}$
$\Delta E = J_{zz}$
$\Delta E = J_{zz}$

Surface Ising Modulations

- Second order terms *usually* benign in bulk spin ice



$\Delta E = J_{zz}$

Favours opposite spins since they allow fluctuation

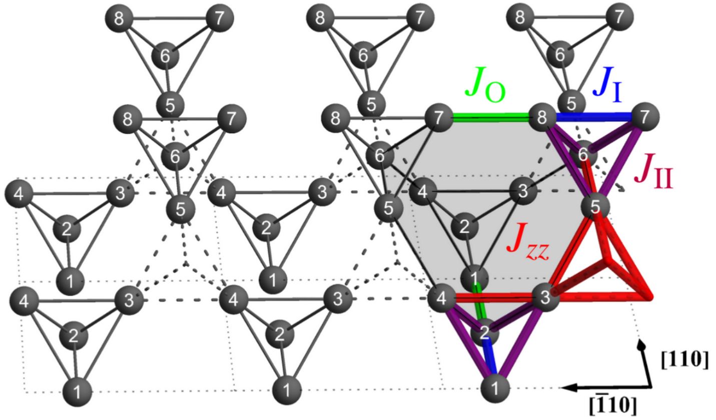
$$H_{\text{eff}}^{(2)} = \frac{2J_\pm^2}{J_{zz}} P_0 \sum_{\langle ij \rangle} S_i^z S_j^z P_0$$

$$= \text{const.}$$

Energy of ice states are all identical

Surface Ising Modulations (cont.)

- Bonds near the surface have different energy denominator
- **Not a constant** even in ground state manifold
- *Four* types of bonds



$$\delta J_{zz} = \frac{2J_{\pm}^2}{J_{zz}}$$

Bulk – Monopole pair

$$\delta J_{\text{I}} = \frac{2J_{\pm}^2}{|J_O|}$$

Surface – break two orphans

$$\delta J_{\text{II}} = \frac{4J_{\pm}^2}{J_{zz} + |J_O|}$$

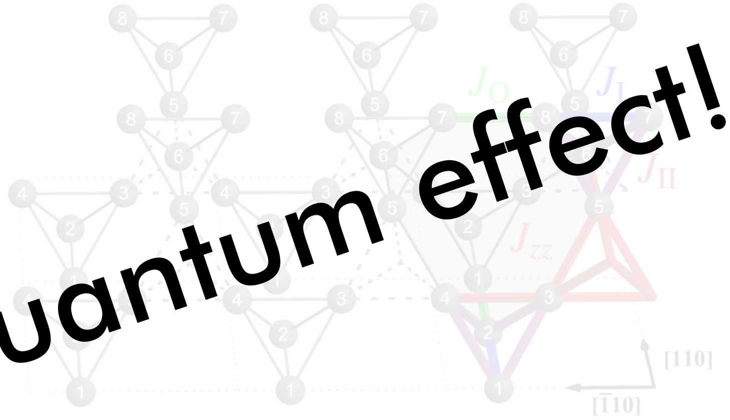
Surface – break orphan and single monopole

$$\delta J_{\text{O}} = \frac{2J_{\pm}^2}{J_{zz}}$$

Surface – Monopole pair

Surface Ising Modulations (cont.)

- Bonds near the surface have different energy denominator
- **Not a constant** even in ground state manifold
- *Four* types of bonds



$$\delta J_{zz} = \frac{2J_{\pm}^2}{J_{zz}}$$

Monopole pair

$$\delta J_{\mathrm{I}} = \frac{2J_{\pm}^2}{|J_O|}$$

Surface – break two orphans

$$\delta J_{\mathrm{II}} = \frac{4J_{\pm}^2}{J_{zz} + |J_O|}$$

Surface – break orphan and single monopole

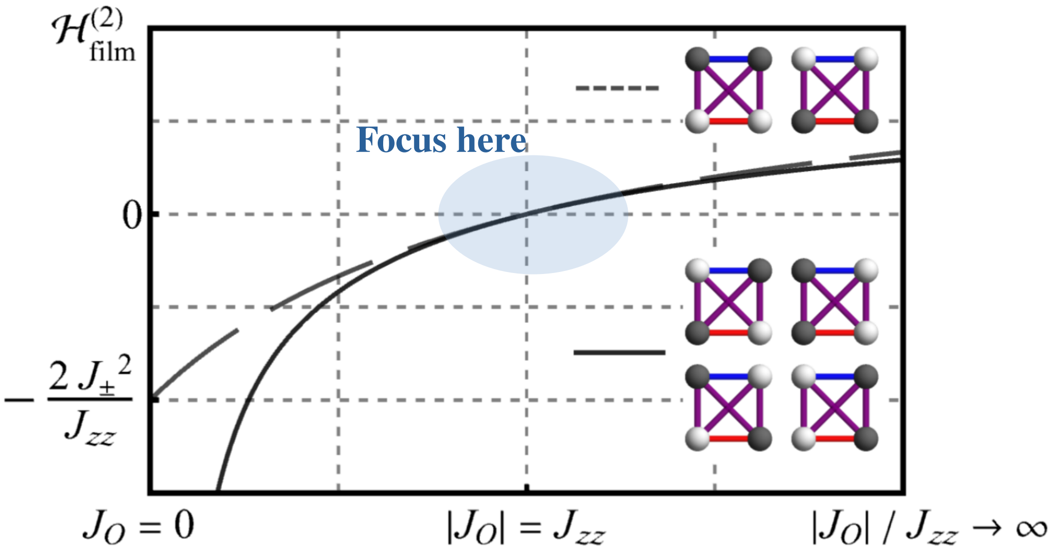
$$\delta J_{\mathrm{O}} = \frac{2J_{\pm}^2}{J_{zz}}$$

Surface – Monopole pair

Leading order quantum effect!

Surface Ising Modulations (cont)

- Bond modulations change ground state manifold for surface tetrahedra
  - **Ice states are no longer degenerate**
- Four of the ice states are selected for J_O different from J_{zz}
- **Degeneracy restored for matching $J_O = J_{zz}$**



Four states "lock" into antiferromagnetic chains along the top/bottom surfaces

Model

- Consider minimal model near point where $J_O = J_{zz}$ where *surface Ising modulations are perturbative*

Taylor expand in η

$$gP_0\left(\sum_{\text{hex}} S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + \text{h.c.}\right)P_0 + P_0\left(2\gamma \sum_{\langle ij\rangle\in\text{I}} S_i^z S_j^z + \gamma \sum_{\langle ij\rangle\in\text{II}} S_i^z S_j^z\right)$$

- Consider where $J_O = J_{zz}(1-\eta)$ and η small gives $\gamma \equiv \dfrac{\eta J_\pm^2}{J_{zz}}$
- In this limit Ising terms are a *perturbation* to QSI

Shift away the bulk and orphan corrections

How do these modulations *and* the film geometry affect QSI?

Semi-classical approach

- Use large-S approach, starting with Villain representation

$$S_i^+ = \bar{S}\, e^{iA_i/2}\,(1 - E_i^2)^{1/2}\, e^{iA_i/2}, \qquad [A_i, E_j] = i\delta_{ij}/\bar{S}$$

$$S_i^- = \bar{S}\, e^{-iA_i/2}\,(1 - E_i^2)^{1/2}\, e^{-iA_i/2}, \qquad \bar{S} = S + 1/2$$

$$S_i^z = \bar{S}\, E_i.$$

- Carry out an expansion of *effective model* about the large-S limit
- For bulk model gives good description of physics of the photon-sector of quantum spin ice without ad-hoc terms

Kwasigrosch, Doucot & Castelnovo, Phys. Rev. B **95**, 134439 (2017)

Effective Hamiltonian

- Use large-S approach, starting with Villain representation

$$\approx \; P_0\left\{ \bar{g} \sum_{\hexagon} (\mathrm{curl}_{\hexagon} A)^2 \right\} P_0 \; + \; P_0\left\{ 6\bar{g} \sum_i E_i^2 \right\} P_0 \; +$$

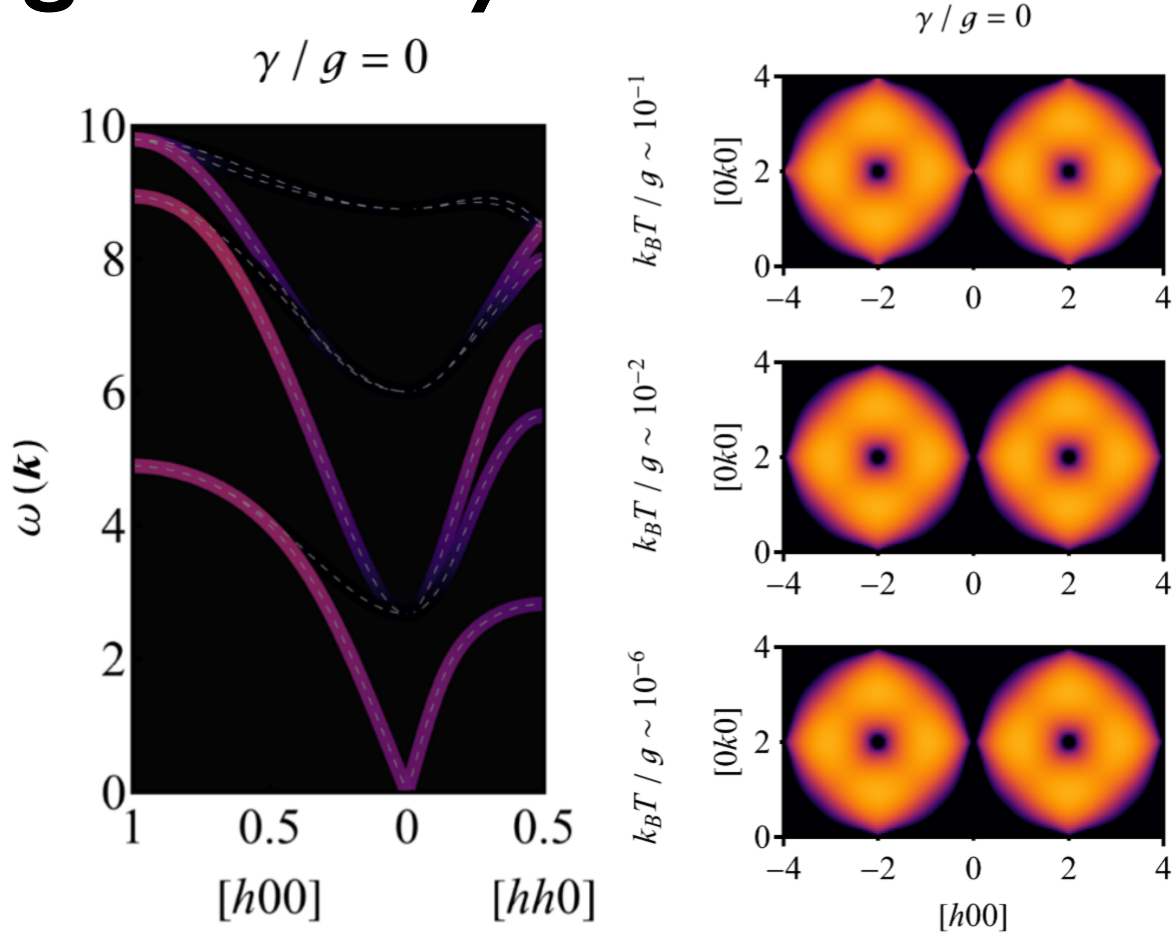
$$P_0\left(2\bar{\gamma} \sum_{\langle ij\rangle \in \mathrm{I}} E_i E_j + \bar{\gamma} \sum_{\langle ij\rangle \in \mathrm{II}} E_i E_j \right) P_0$$

- Quadratic Hamiltonian in canonically conjugate variables; solvable
- Projection to ground state manifold removes all zero modes

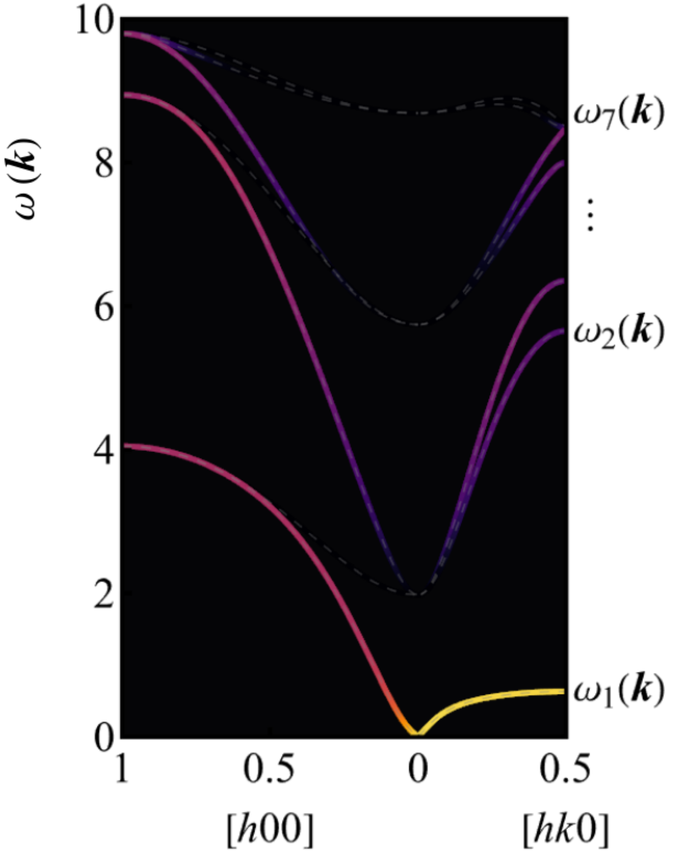
Effect of film geometry

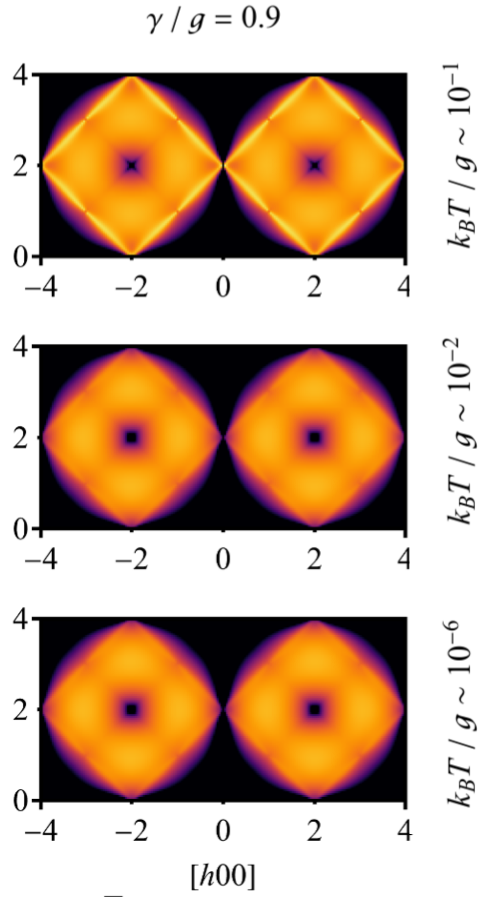
- Spectrum of emergent photons modified
  - Larger number of photon bands due to finite thickness
- Zero mode counting subtle; each tetrahedron *and* orphan bond provides constraint

$L = 2$

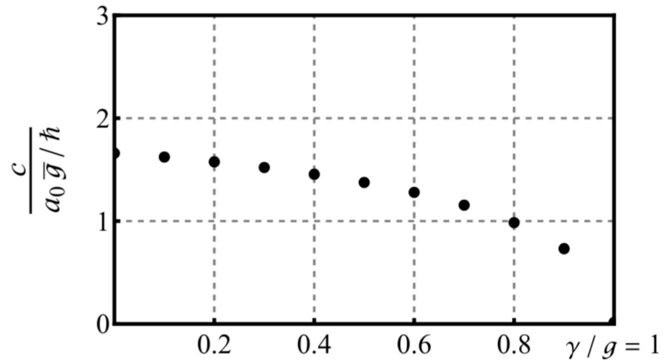


Phase Diagram

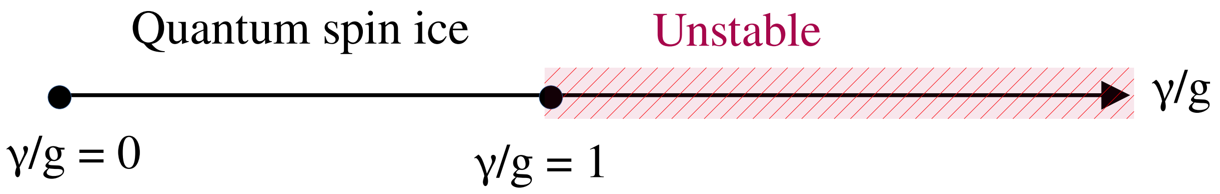




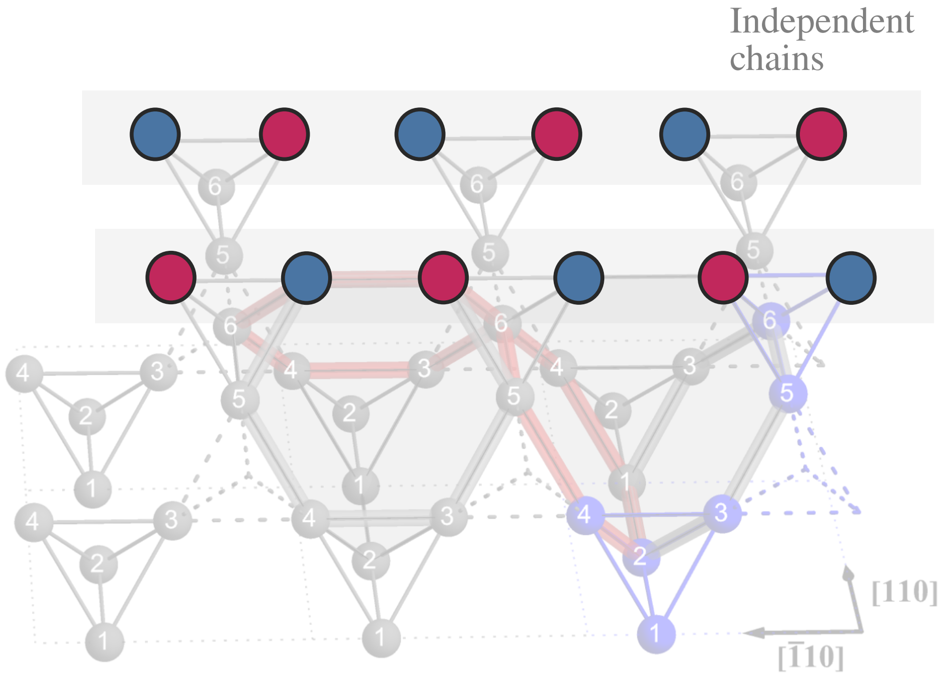
- Photon velocity drops as γ increases
- Line of modes goes soft at γ = 1
- **Instability of quantum spin ice state**



Chain Instability

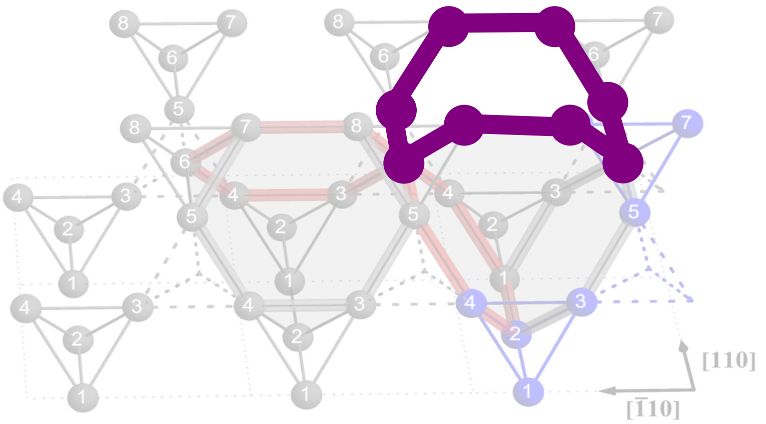


- Line of soft modes is perpendicular to surface chains
- Ising interactions fix four ice states; orphans force AF but *independent* chains
- New effective “surface”

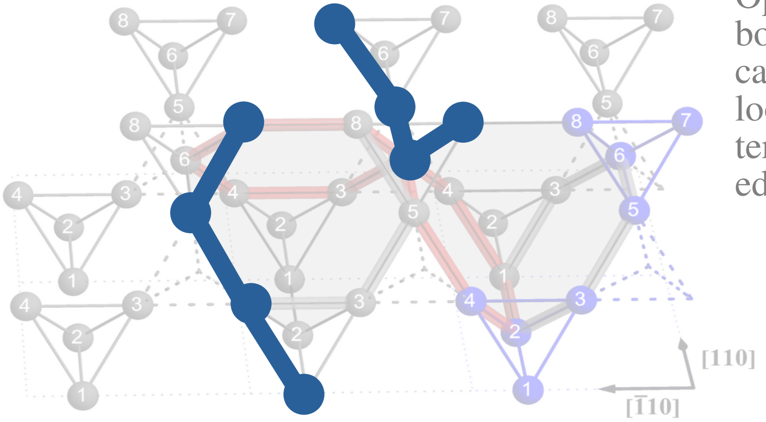


Surface Loops, Z_2, and all that

Longer than six for J_O>0 case

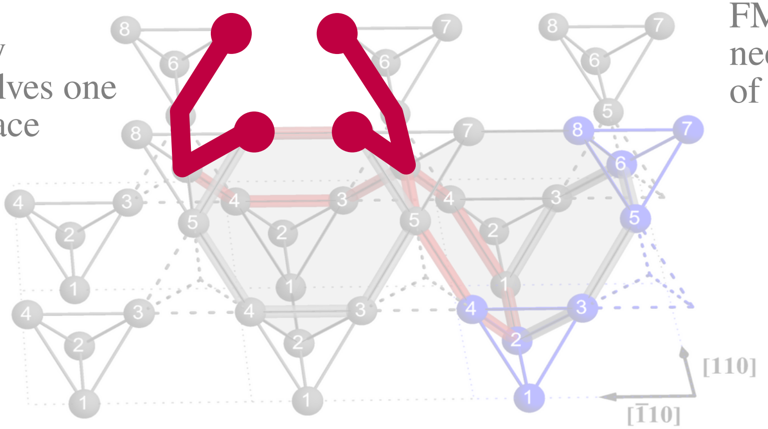


For thin films can even from top to bottom surface



Open boundary can have loops terminate at edges

Only involves one surface



FM case needs **pair** of loops

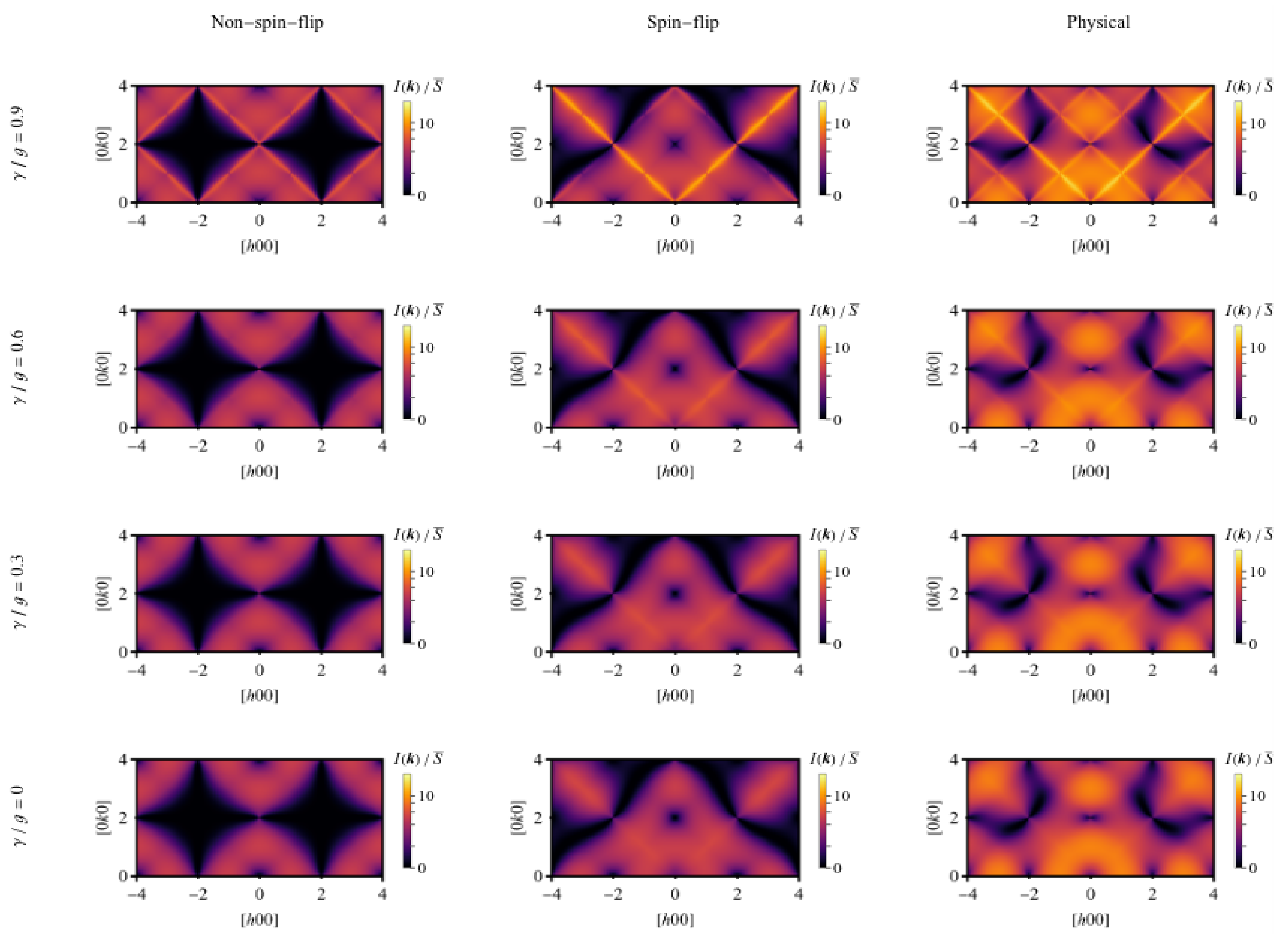
- Boundary conditions can change quantum tunnelling processes too
- Open ($J_O = 0$) vs. FM ($J_O < 0$)

Conclusions

- Considered the effect of transverse quantum exchange on spin ice thin films
  - **Leading order:** Ising interactions at surface are modulated (symmetry is reduced at the surface)
  - When AF orphan bond matches bulk, acts *perturbatively* QSI physics
  - Drives an instability toward **surface (chain) ordering**
- **Lots of directions to explore**
  - Gauge field boundary conditions, scaling of confinement in slabs
  - Z_2 spin liquid from FM orphans?
  - Other terminations [111], [110], ...

Details to appear soon (-ish)
arxiv:251x.xxxxxx

Extra Slides



Non−spin−flip
Spin−flip
Physical
γ / g = 0.9
γ / g = 0.6
γ / g = 0.3
γ / g = 0
[0k0]
[h00]
I(k) / S̄