Dynamics of 2- and 3-dimensional Quantum Spin Liquids

OAK, September 2015 Kolymbari

Johannes Knolle

TCM Cavendish Lab – University of Cambridge

Collaborators:

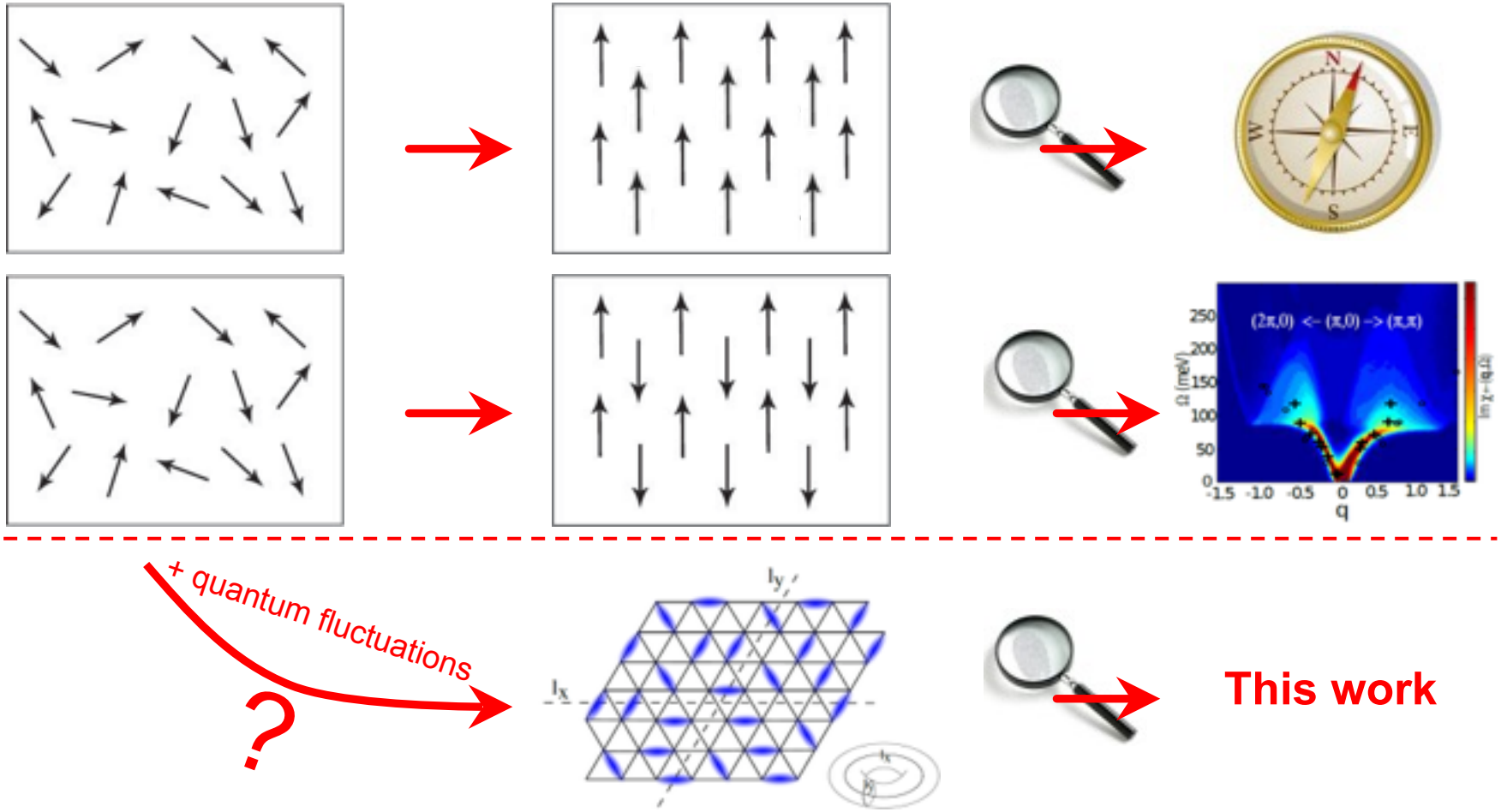
Roderich Moessner	***(MPI PKS Dresden)***
Dima Kovrizhin	***(Cambridge U)***
John Chalker	***(Oxford U)***
Gia-Wei Chern	***(Los Alamos NL)***
Natalia Perkins	***(U of Minneapolis)***
Brent Perreault	***(U of Minneapolis)***
Fiona Burnell	***(U of Minneapolis)***

Outline

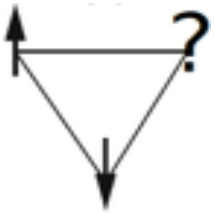
1. Introduction and Motivation
   - → New states of matter and fractionalization
   - → Spin liquids and dynamics
2. Kitaev's Honeycomb Model
   - → Exact solution
   - → 2- and 3-dimensional candidate materials
3. Exact Dynamical Correlation Functions
   - → Spin structure factor and the X-ray edge
   - → Raman scattering
4. Conclusion

1. Introduction

- Understanding phases of matter and their transitions. Democritus 500 BC
- Phases are described by symmetries and order parameters. Landau 1937



Quantum Spin Liquids



- Frustrated magnets can evade ordering down to T=0.

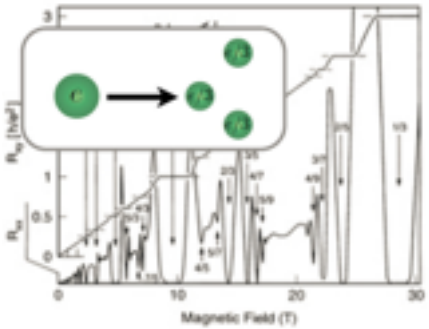
  Anderson 1973

- **QSL:** State of interacting spins that breaks no rotational or translational symmetry and has only short range spin correlations.

RVB: $\psi =$

Moessner, Sondhi 2000

- **Topologically ordered** with **fractional excitations.** Wen 1991



FQHE

Spinons in QSLs

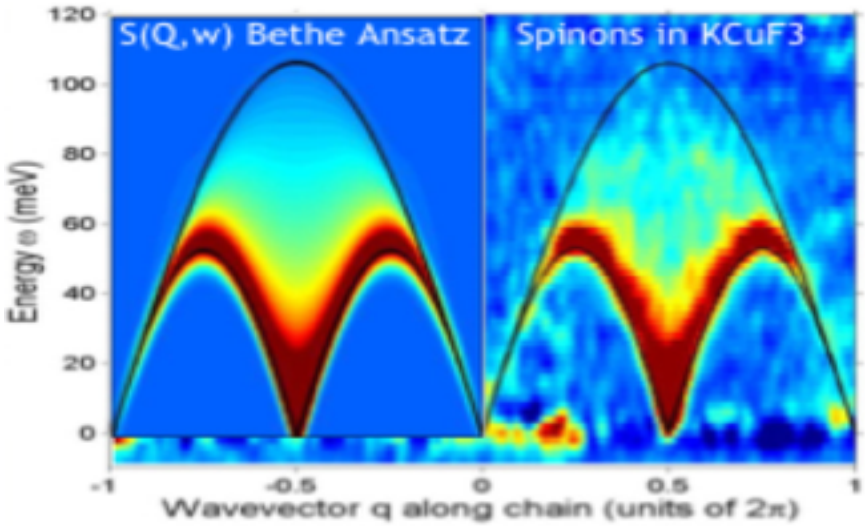
QSL and Dynamics

- Shortage of experimental signatures due to lack of local order.

- Look for signatures of QSL in the **dynamical response**!

  → Inelastic Neutron Scattering

  → Established in 1D Bethe 1931
  Caux, Tennant 2012



- **Poor understanding of strongly interacting systems beyond 1D:**

  → No exact results, only numerics or uncontrolled approximations

- Here: **First exact calculation of a dynamical correlation function for a strongly interacting 2D and 3D QSL.**

Motivation

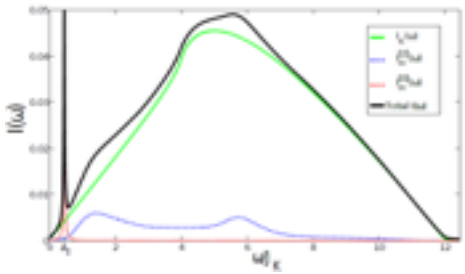
- Main Questions:

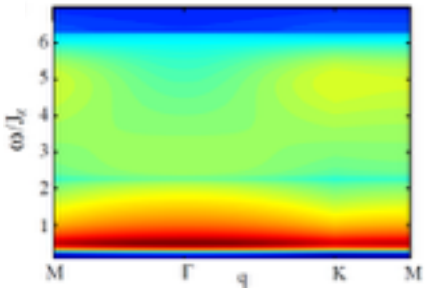
  How to diagnose a quantum spin liquid in real experiment?

  How to probe fractionalized quasiparticles?

- Take Home Message:

  Signatures of fractionalization are visible in dynamical correlation functions accessible by standard experiments.





2. Kitaev‘s Model

- Spin ½ on the honeycomb lattice with strong spin orbit coupling

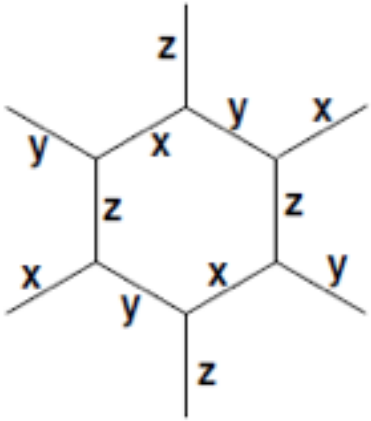
  Kitaev 2006

$$H = -J_x \sum_{x \text{ links}} \sigma_j^x \sigma_k^x - J_y \sum_{y \text{ links}} \sigma_j^y \sigma_k^y - J_z \sum_{z \text{ links}} \sigma_j^z \sigma_k^z$$

- Large number of conserved quantities, local plaquette operators:

$$W_p = \sigma_1^x \sigma_2^y \sigma_3^z \sigma_4^x \sigma_5^y \sigma_6^z$$

$$[H, W_p] = 0$$

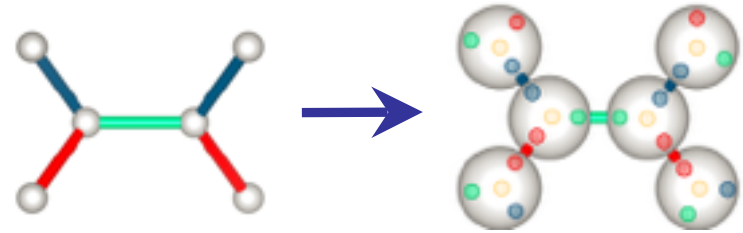


- Exactly solvable interacting 2D model!

Exact Solution

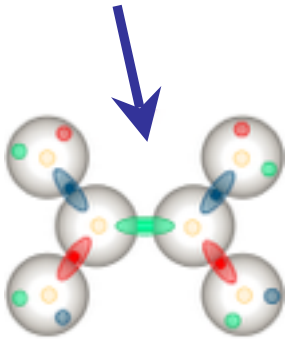
- Mapping spins to Majoranas:

$$\sigma_i^a = i c_i c_i^a, \qquad a = x, y, z$$

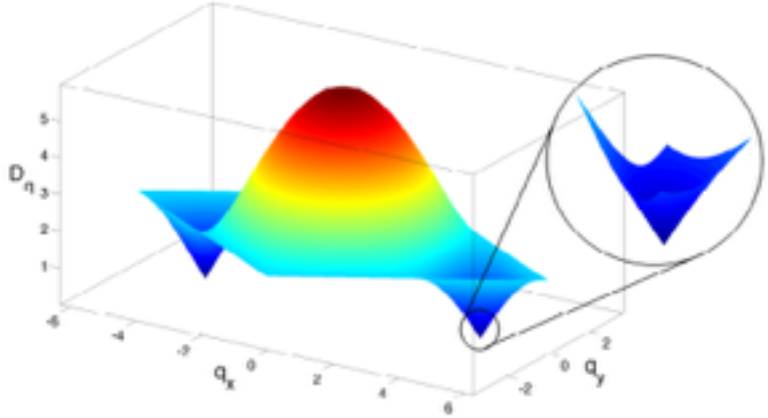


- Quadratic Hamiltonian in each flux sector:

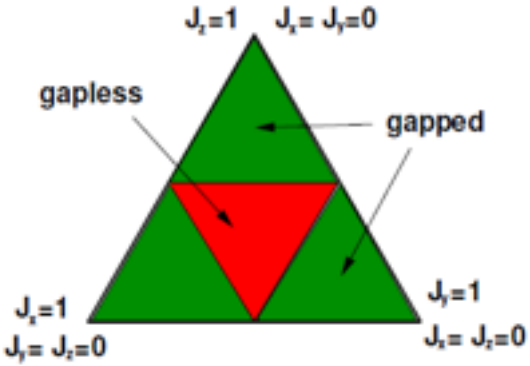
$$H = -\sum_{a=x,y,z} J_a \sum_{\langle ij \rangle_a} i c_i \hat{u}_{\langle ij \rangle_a} c_j \text{ with } \hat{u}_{\langle ij \rangle_a} \equiv i c_i^a c_j^a$$



- Spectrum:



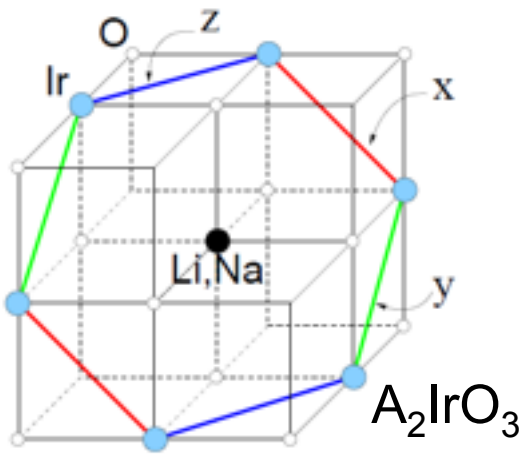
- Phase diagram:

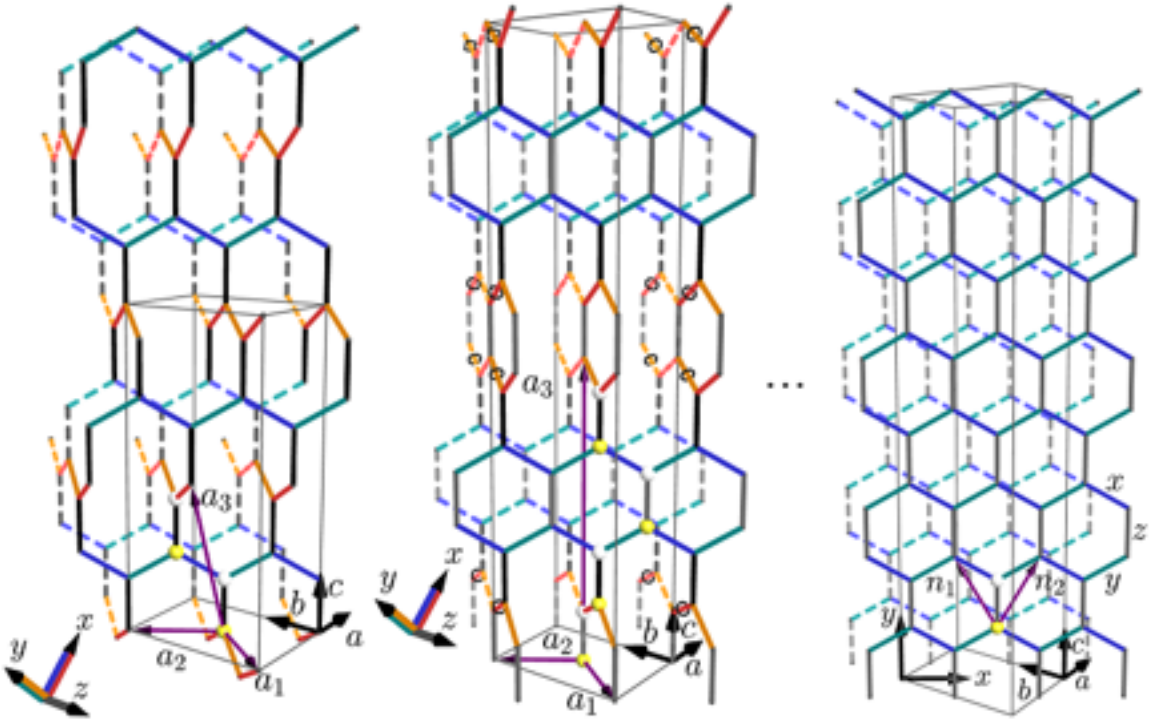


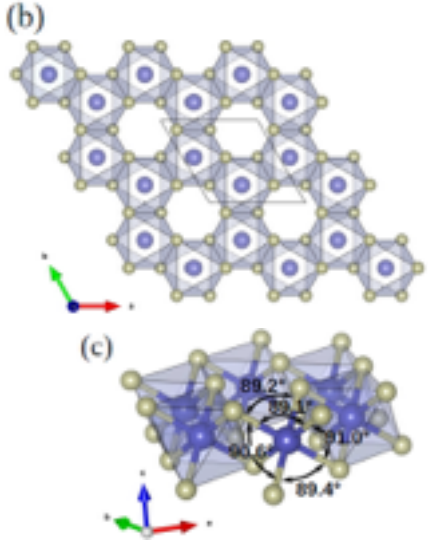
Candidate Materials

- → Strong spin orbit interaction of late transition metal ions (e.g. Ir).

  Jackeli, Khaliullin 2009, Singh, Gegenwart 2010
  Modic et al. 2014, Takayama et al. 2015, Plumb et al. 2014







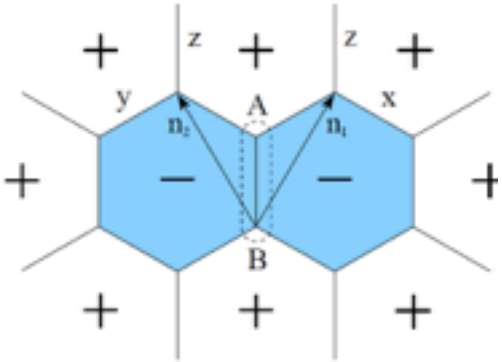
- **Recent 3D harmonic honeycomb series for Li_2IrO_3 and α-$RuCl_3$**

3. Dynamical Correlations

- Spin correlation function:

$$S^{ab}_{\mathbf{R},\mathbf{R}'}(t) = \langle M_G|\langle G|\sigma^a_{\mathbf{R}}(t)\sigma^b_{\mathbf{R}'}(0)|G\rangle|M_G\rangle$$

$$\sigma^a_i \rightarrow i c_i \hat{\Pi}^{\text{left}}_{\langle ij\rangle,a}\hat{\Pi}^{\text{right}}_{\langle ij\rangle,a}$$



- Only N.N. spin correlations from flux selection rule

Baskaran, PRL 2007

$$S^{ab}_{\mathbf{R},\mathbf{R}'}(t) = -i\langle M_G|e^{iH_0 t}c_{\mathbf{R}}e^{-it(H_0+V_a)}c_{\mathbf{R}'}|M_G\rangle\delta_{a,b}\delta_{\langle \mathbf{R},\mathbf{R}'\rangle_{N.N.}}$$

- Dynamics from the perturbed **Majorana sector** with **two extra fluxes** neighboring the N.N. bond

$$V_a = -J_a 2i c_{\mathbf{R}} c_{\mathbf{R}'}$$

→ Quadratic **non-equilibrium** problem

Spin Structure Factor

- Spin correlation function as a **local quantum quench**

$$S_{ij}^{ab}(t) = -i\langle M_0|e^{i\hat{H}_0 t}\hat{c}_i e^{-i(\hat{H}_0+\hat{V}_a)t}\hat{c}_j|M_0\rangle\delta_{ab}\delta_{\langle ij\rangle,a}$$

- Lehmann representation: lowest response and few particle contribution.

$$S_{ij}^{ab}(\omega) = -i\sum_{\lambda}\langle M_0|\hat{c}_i|\lambda\rangle\langle\lambda|\hat{c}_j|M_0\rangle \times \delta\left[\omega - (E_\lambda - E_0)\right]\delta_{\langle ij\rangle,a}\delta_{ab}$$

- Bogoliubov rotation $\hat{b}_\lambda = \sum_{\mathbf{q}} X^*_{\lambda\mathbf{q}}\hat{a}_{\mathbf{q}} + Y^*_{\lambda\mathbf{q}}\hat{a}^\dagger_{\mathbf{q}}$ diagonalizes $\hat{H}_0 + \hat{V}_a$

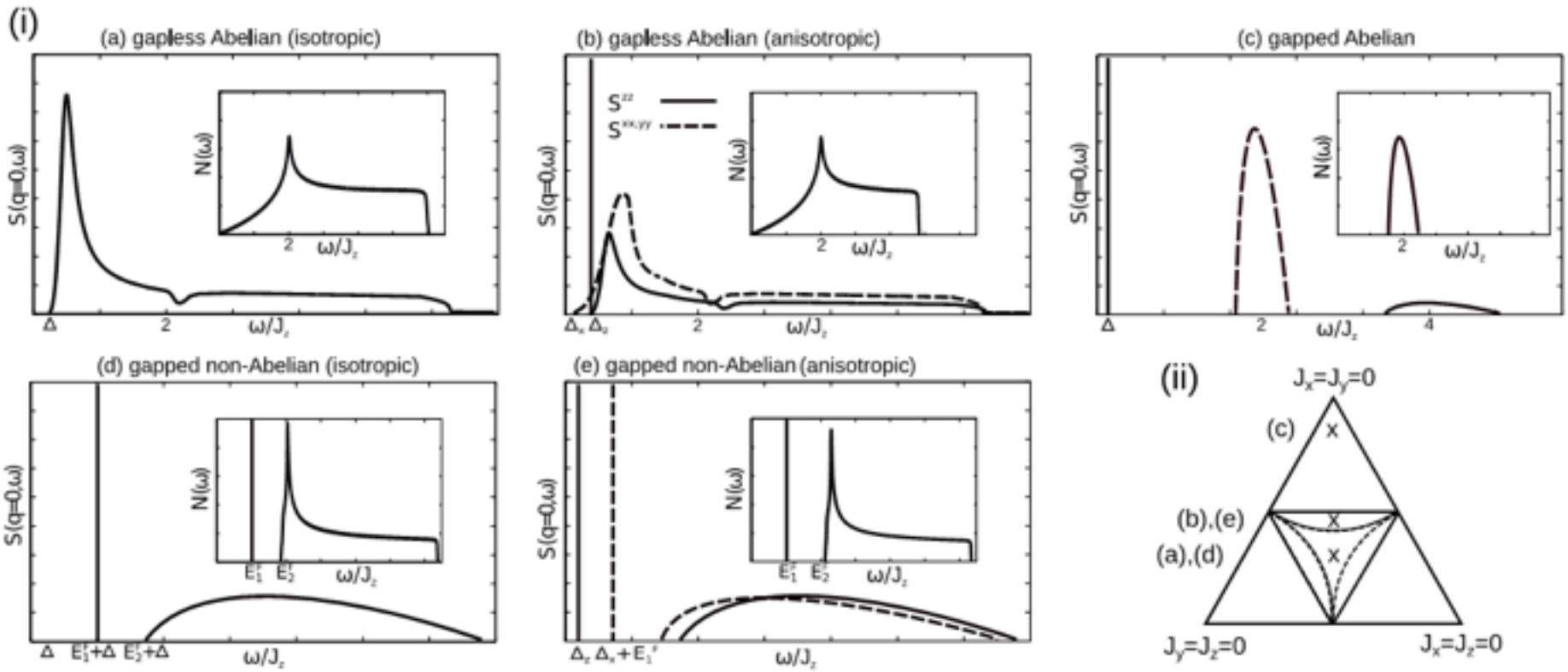
- $|\lambda_0\rangle = [X^\dagger X]^{\frac{1}{4}} e^{-\frac{1}{2}\hat{\mathbf{a}}^\dagger X^{*-1}Y^*\hat{\mathbf{a}}^\dagger}|M_0\rangle$ gives the ground state

- Single particle contribution from the s.p. states $|\lambda\rangle = \hat{b}^\dagger_\lambda|\lambda_0\rangle$

$$S^{zz}_{[1]}(\mathbf{q}=0,\omega) = \frac{2}{N}|\det X|\sum_{\lambda,\mathbf{q}\mathbf{q}'}\delta[\omega - (E^1_\lambda - E_0)]\cos\theta_{\mathbf{q}} X^{-1}_{\mathbf{q}\lambda}[X^{-1}]^\dagger_{\lambda\mathbf{q}'}\cos\theta_{\mathbf{q}'}$$

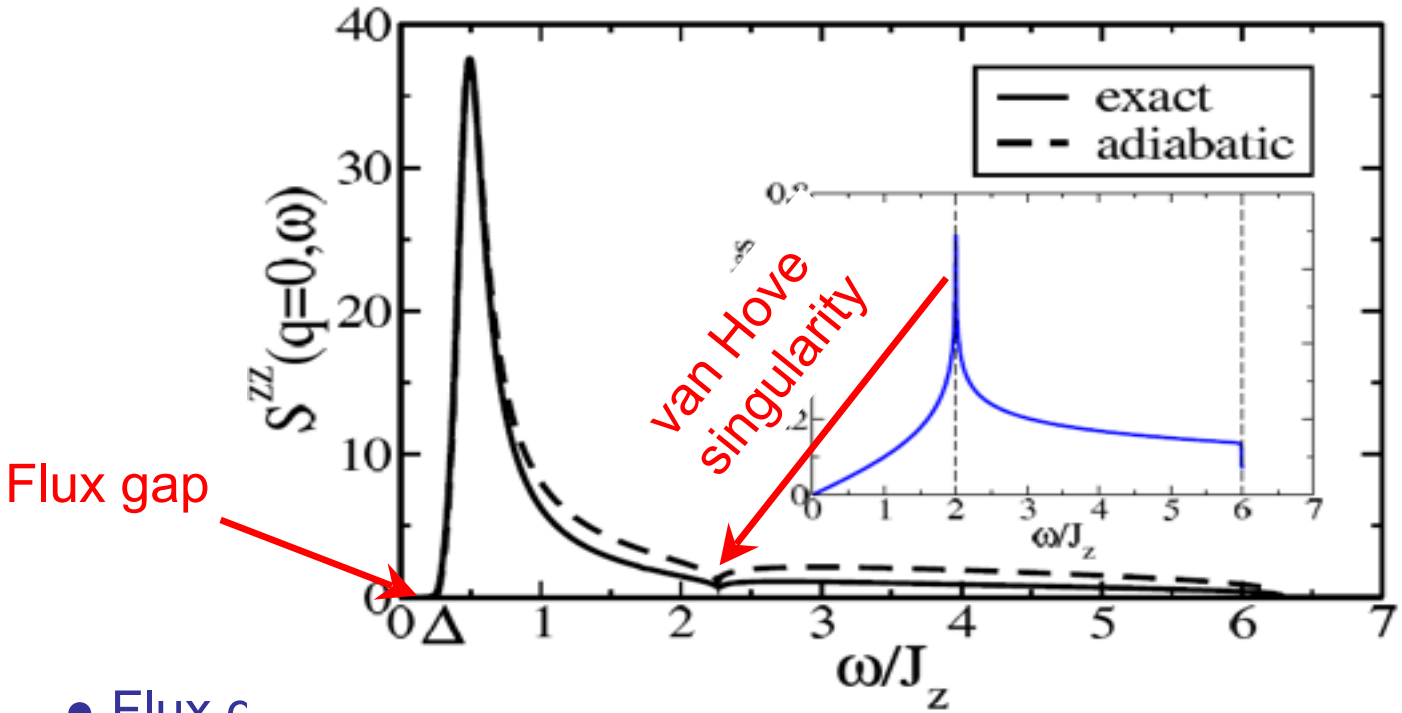
Results 2D QSL

- Response on the full phase diagram for Abelian and non-Abelian QSL
  → **Salient signatures of fractionalization are visible!**



Results Gapless QSL $J_z=J_x=J_y$

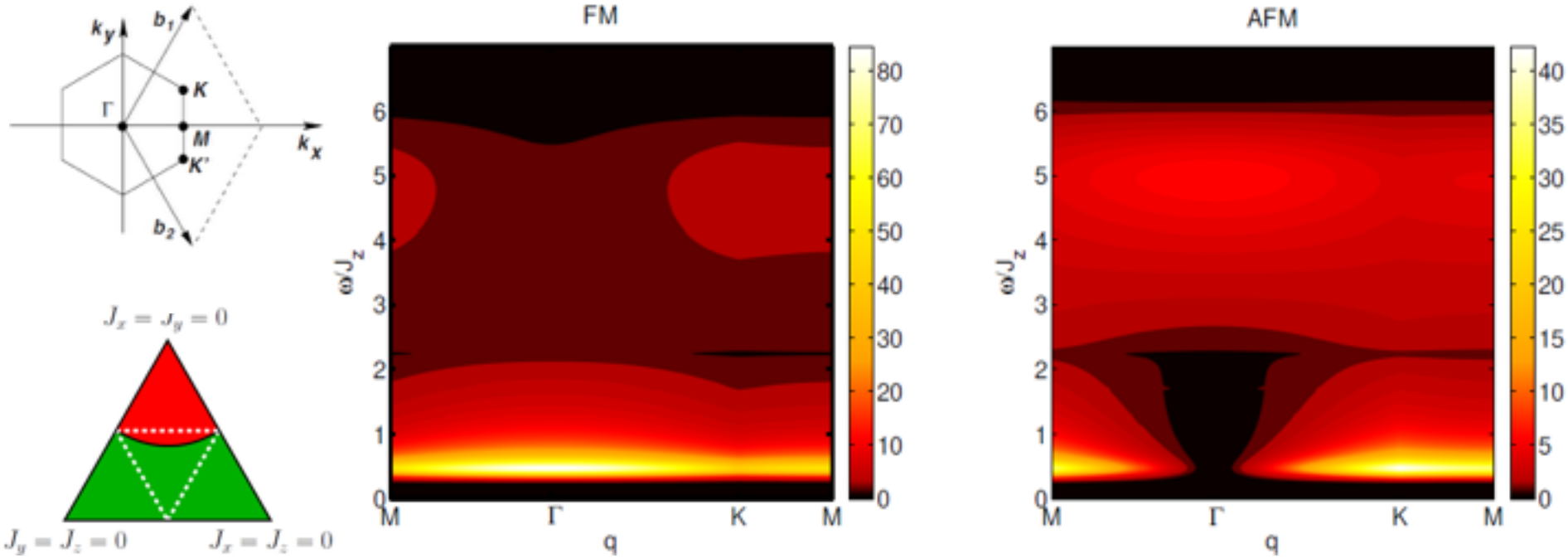
- **Exact structure factor S(q=0,ω) for a 2D quantum spin liquid:**



- Flux gap of Δ~0.26 and features of the matter fermion DOS
  → **Salient signatures of fractionalization are visible!**

Results Gapless Phase $J_z=J_x=J_y$

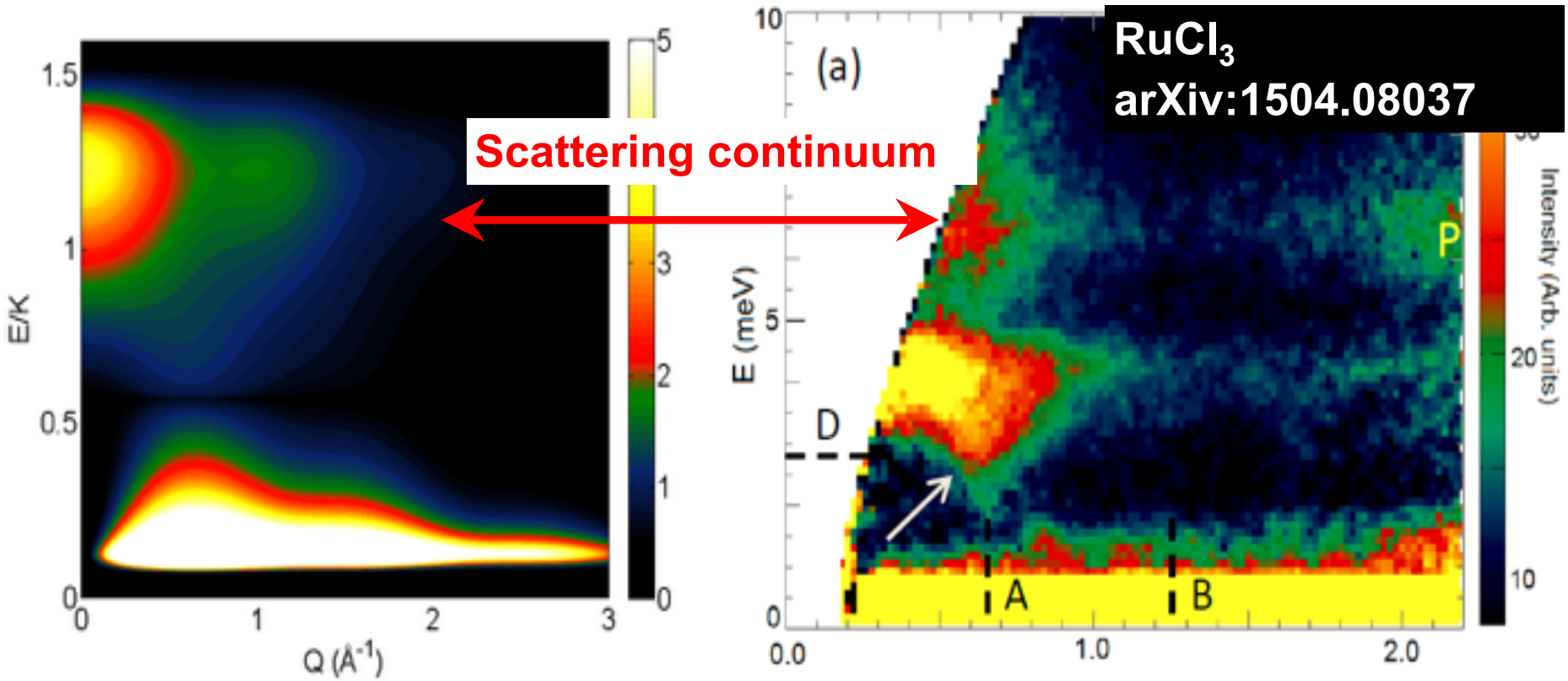
- Momentum-dependence of $\mathrm{Im}\chi(q,\omega)$ as measured by INS, FM vs AFM



- Broad features in reciprocal space → N.N. correlations only
- Results in full phase diagram → **δ-function in dimer limit!**

Results Gapless Phase $J_z=J_x=J_y$

- Powder Average as measured in INS for AFM Kitaev coupling



$$\frac{d\sigma}{dE} \propto \frac{q}{q_{\text{in}}} \int d\theta \;\; |f(\mathbf{q})|^2 \left[(1-\cos^2\theta)\mathcal{S}^{xx}(\mathbf{q},\omega) + (1-\sin^2\theta)\mathcal{S}^{yy}(\mathbf{q},\omega) + \mathcal{S}^{zz}(\mathbf{q},\omega)\right]$$

Exact Solution and the X-Ray Edge

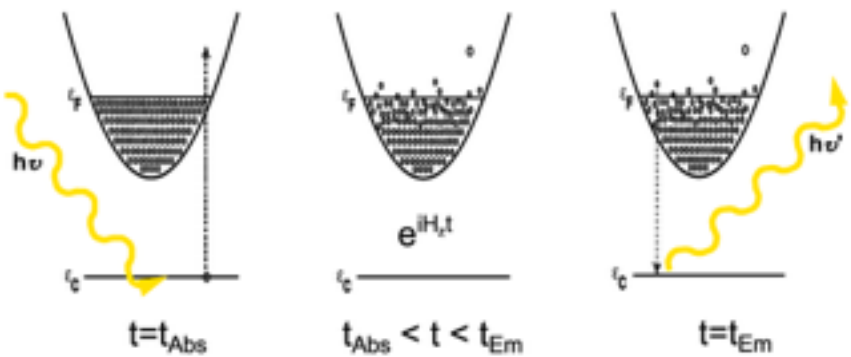
- In terms of complex matter fermions: $\hat{f}_{\mathbf{r}} = \frac{1}{2}(\hat{c}_{A\mathbf{r}} + i\hat{c}_{B\mathbf{r}})$

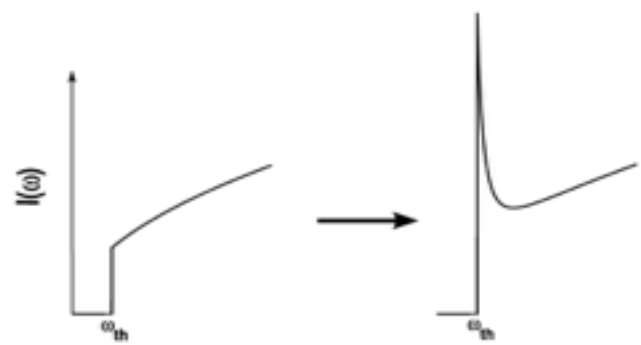
$$S^{zz}_{A0B0}(t) = i[g(t,0) + g(0,t)]$$

$$g(t,0) = -i\langle M_0|T\{\hat{f}_0(t)\hat{f}_0^\dagger(0)\exp\left[-i\int_0^t \hat{V}_z(\tau)d\tau\right]\}|M_0\rangle$$

with $\hat{V}_z(t) = v\left[\hat{f}_0^\dagger(t)\hat{f}_0(t) - \frac{1}{2}\right]$ **local impurity term**

- Local quench equivalent to a **X-ray edge** problem!



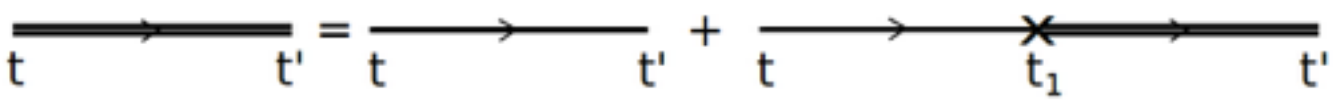


Nozieres, DeDominicis 1969

Exact Solution

- Problem reduces to solving a **singular integral equation**.

$$G_c(t,t') = G_0(t,t') - 4J_z \int_{t'}^{t} dt_1 G_0(t,t_1) G_c(t_1,t').$$



- Solved for $t \to \infty$ for a Fermi liquid with $G_0(t) \sim 1/t$

  Nozieres, DeDominicis 1969 → **not applicable** here!

- For the singular integral equation

  find a new integral operator K_1 such that

  is non-singular. Mushkelishvili 1953

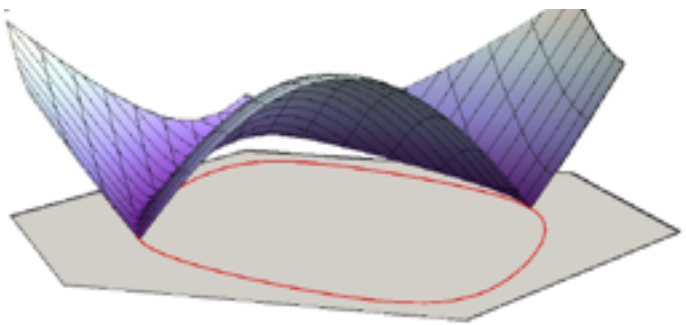
$$f = \hat{K}_2 \tilde{G}_c$$
$$\hat{K}_1 f = \hat{K}_1 \hat{K}_2 \tilde{G}_c$$

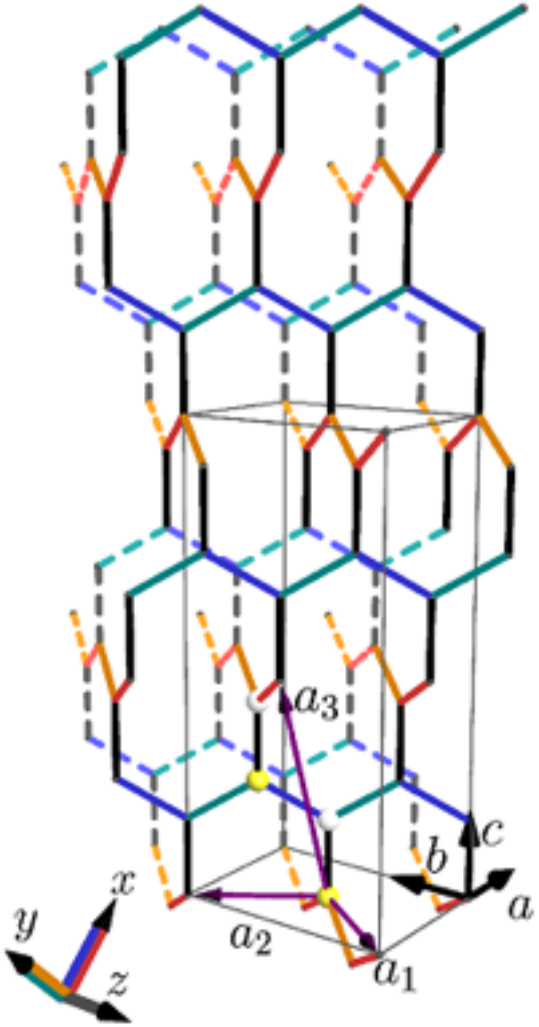
- Method can be extended to Non-Abelian and 3D generalizations of the Kitaev model.

Structure Factor 3D Kitaev QSL

- Generalization to the **hyperhoneycomb** lattice for ß-**Li_2IrO_3**

  Mandal et al. 2009,
  Lee et al. 2014/15;
  Kimchi 2014



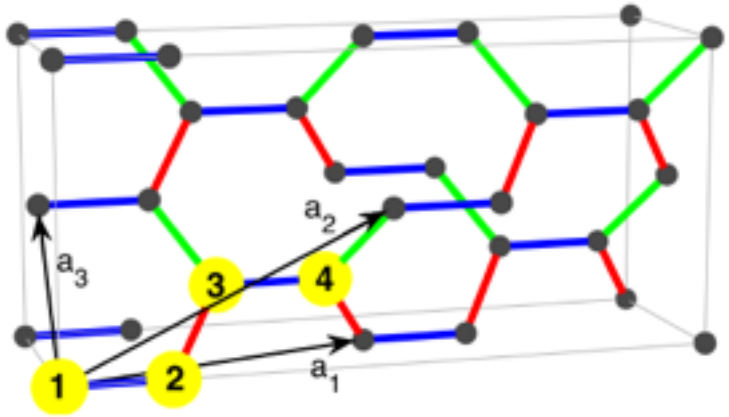


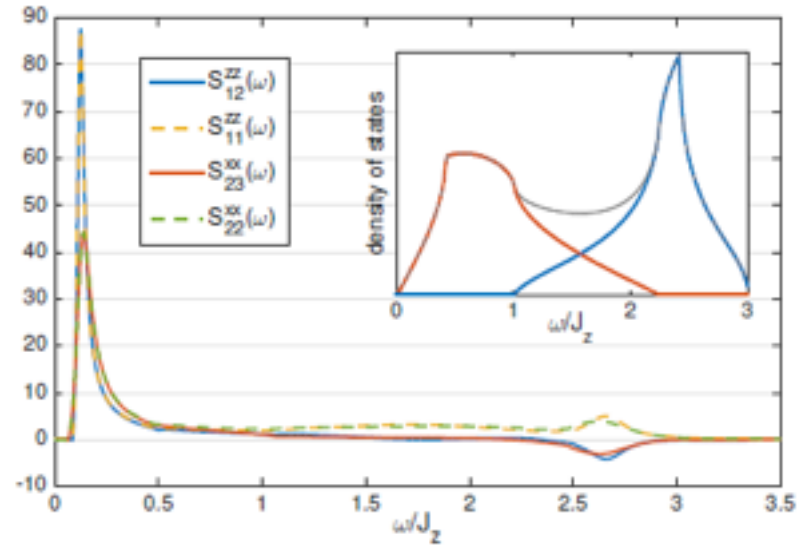
- Remains exactly solvable with fluxes replaced by flux lines.

- Spectrum has a gapless line. Additional three spin interaction induces **Weyl Spin Liquid**.

  Hermanns et al. 2015

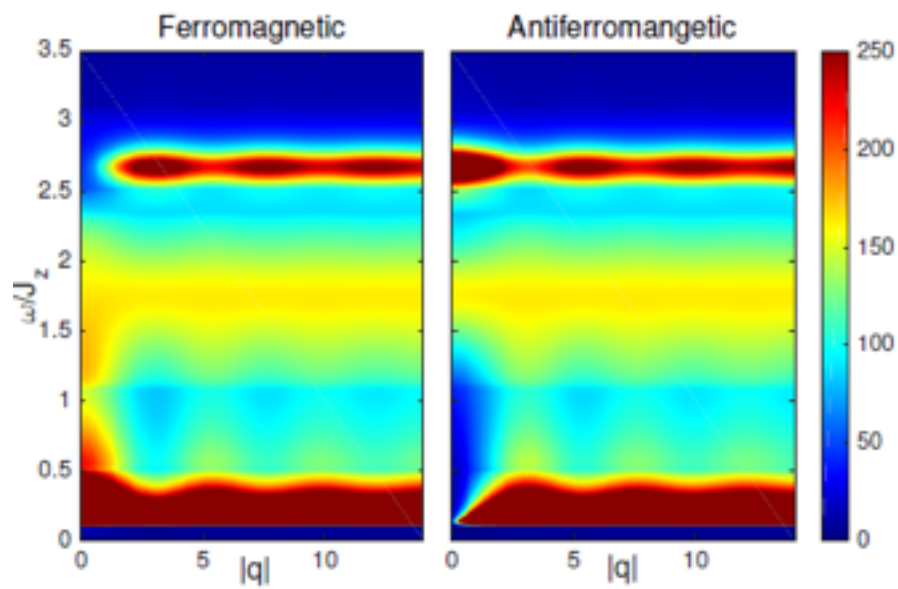
Results isotropic 3D QSL



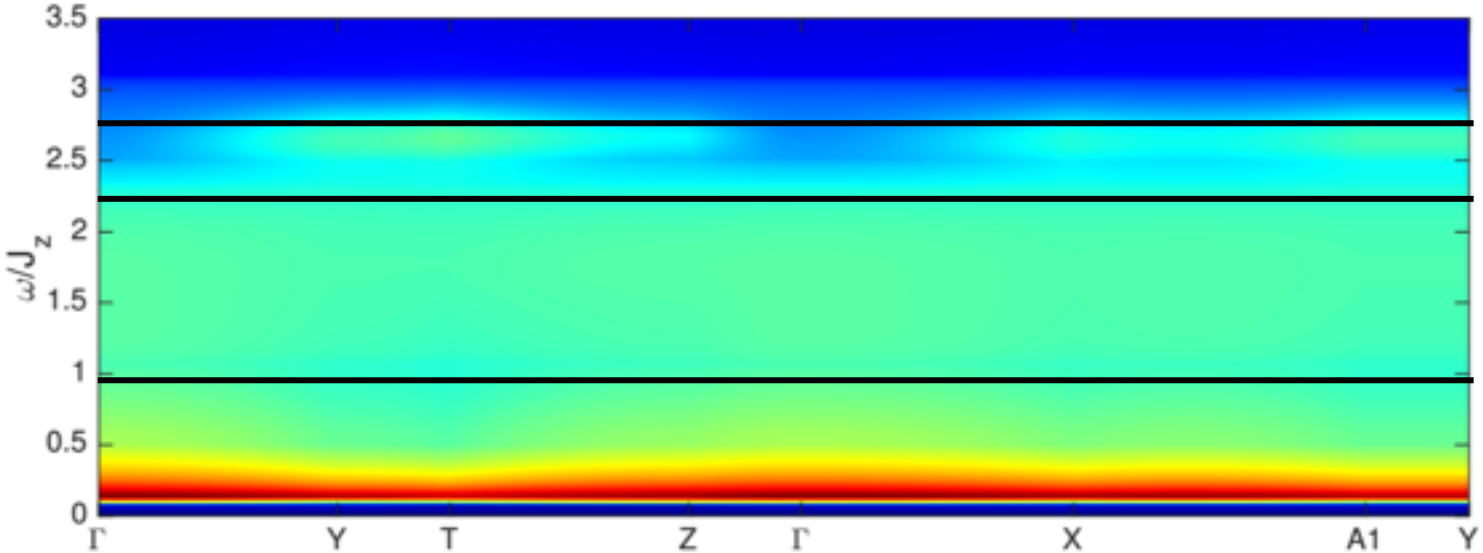


- New features from inequivalent bonds and multiple bands
  → no orthogonality catastrophe

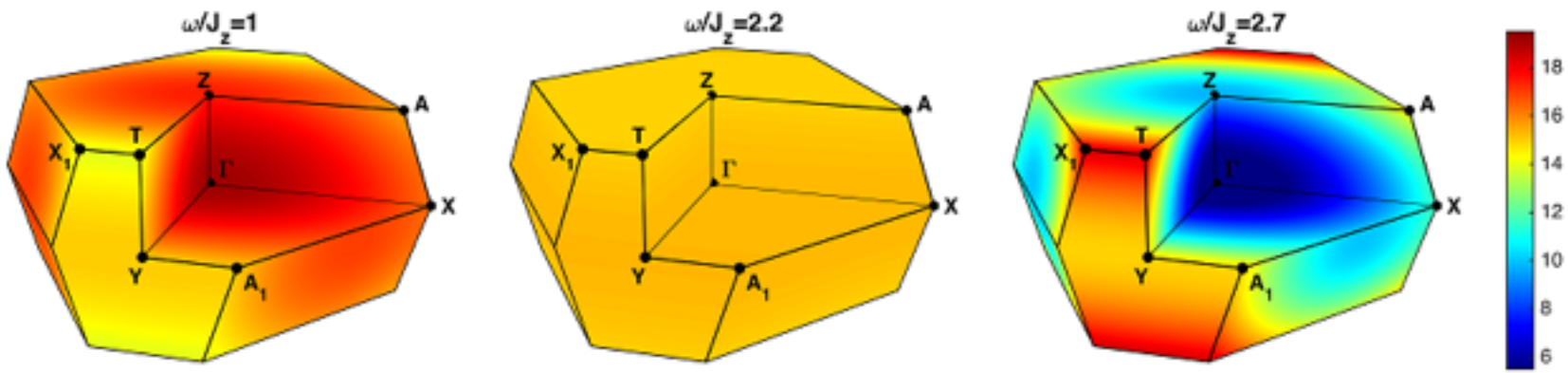
- Distinct (high-energy) features of the Majorana fermions in **powder averaged structure factor**!
  → relation to ß-**Li_2IrO_3** ?



Results isotropic 3D QSL



- Single crystal dynamical structure factor along a path through the BZ



Raman Scattering

- Photon-in photon-out process Devereaux, Hackl 2007

  → Polarization dependent

- **Polarization independence in QSLs** Cepas, Lhuillier 2008; Ko et al. 2010

- Loudon-Fleury correlation function Shastry, Shraiman 1990

$$I(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \hat{\mathcal{R}}(t) \hat{\mathcal{R}}(0) \rangle$$

- Na_2IrO_3 described by Kitaev-Heisenberg model Jackeli, Khaliullin 2009

$$\hat{\mathcal{H}} = -J_K \sum_{\langle ij \rangle_a} \hat{\sigma}_i^a \hat{\sigma}_j^a + J_H \sum_{\langle ij \rangle} \hat{\boldsymbol{\sigma}}_i \cdot \hat{\boldsymbol{\sigma}}_j$$

- Vertex: $$\hat{\mathcal{R}} = \sum_{\langle ij \rangle_a} (\hat{\boldsymbol{\epsilon}}_{\text{in}} \cdot \mathbf{d}_a)(\hat{\boldsymbol{\epsilon}}_{\text{out}} \cdot \mathbf{d}_a) \left(K_K \hat{\sigma}_i^a \hat{\sigma}_j^a + K_H \hat{\boldsymbol{\sigma}}_i \cdot \hat{\boldsymbol{\sigma}}_j \right)$$

Raman Scattering Theory

- Close to the integrable limit $\lambda = J_H/J_K = K_H/K_K \ll 1$

$$I(\omega) \approx I_K(\omega) + I_H(\omega)$$

- Exactly solvable contribution of the integrable Kitaev model:

$$I_K(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle 0| \mathrm{T}[\hat{\mathcal{R}}_K(t)\hat{\mathcal{R}}_K(0)]|0\rangle$$

$$I_K(\omega) = 4\pi \sum_{\mathbf{q}} \delta\left(\omega - 4|S(\mathbf{q})|\right) \left[\frac{\mathrm{Im}\left[h(\mathbf{q})S^*(\mathbf{q})\right]}{|S(\mathbf{q})|}\right]^2$$

with $$h(\mathbf{q}) = \sum_{\alpha} K_K \left(\hat{\boldsymbol{\epsilon}}_{\mathrm{in}} \cdot \mathbf{n}_\alpha\right)\left(\hat{\boldsymbol{\epsilon}}_{\mathrm{out}} \cdot \mathbf{n}_\alpha\right) e^{i\mathbf{q}\mathbf{n}_\alpha}$$

- **Polarization independent!**

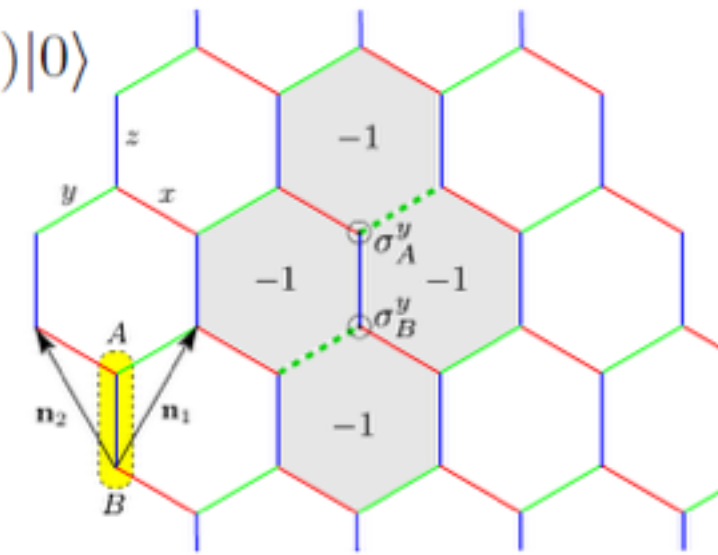
Raman Scattering Theory

- Integrability-breaking Heisenberg term:

$$I_H(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle 0| \mathrm{T}[\hat{\mathcal{R}}_H(t)\hat{\mathcal{R}}_H(0)|0\rangle$$

- Selection rule from flux sector

  e.g. for $\langle \sigma^y_{A\mathbf{r}}(t)\sigma^y_{B\mathbf{r}}(t)\sigma^y_{A\mathbf{r}'}(0)\sigma^y_{B\mathbf{r}'}(0)\rangle$



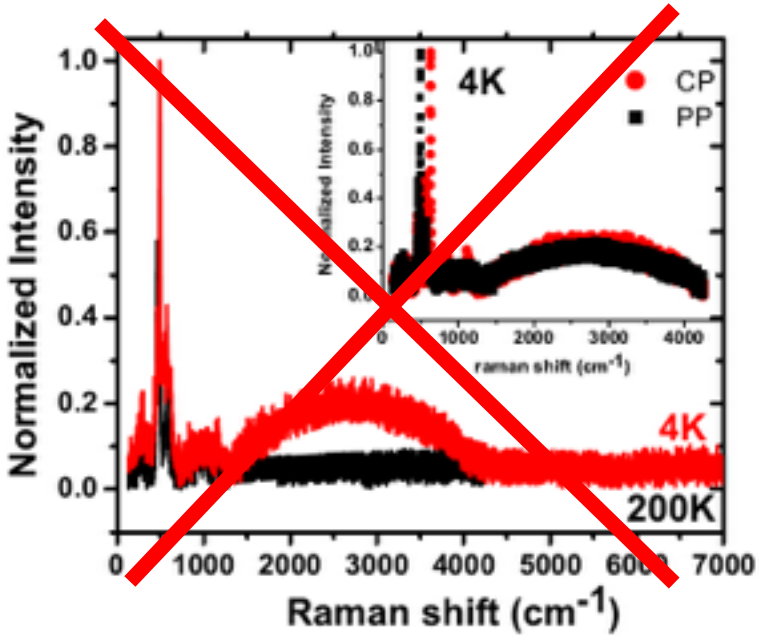
- **Quantum quench of 4 fluxes:**

$$I_H(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \sum_{a=x,y,z} \sum_{\mathbf{r}} \langle M_0| e^{it\hat{\mathcal{H}}_0} e^{-it(\hat{\mathcal{H}}_0 + \hat{V}^a_{\mathbf{r}})} |M_0\rangle$$

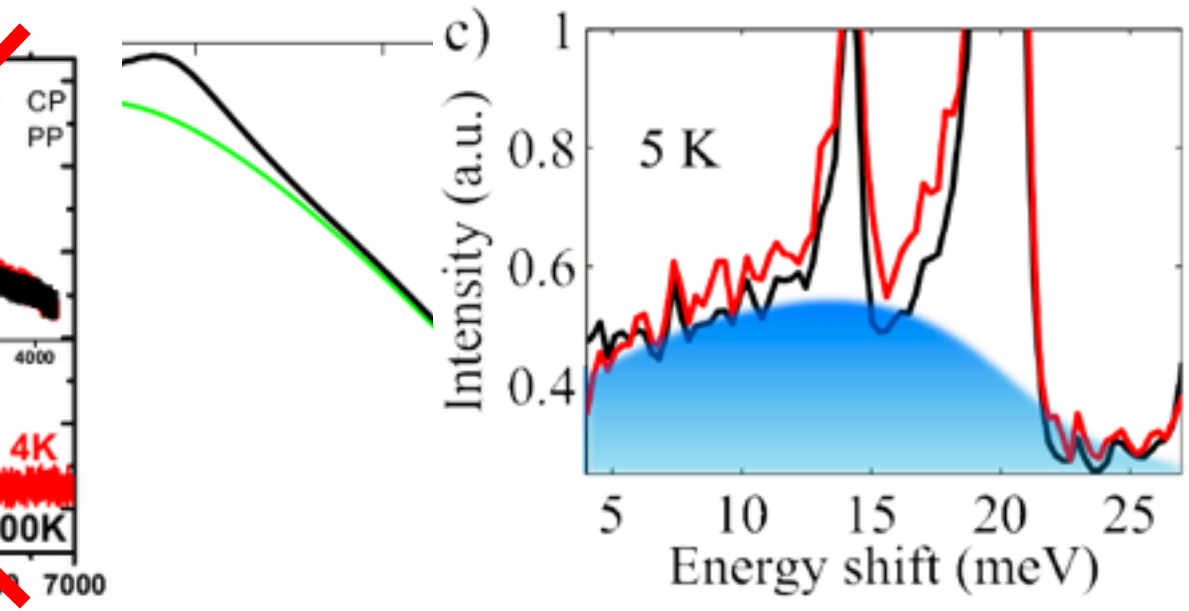
- Sharp contribution at the flux gap from g.s. contribution $|\lambda\rangle = |M_F\rangle$

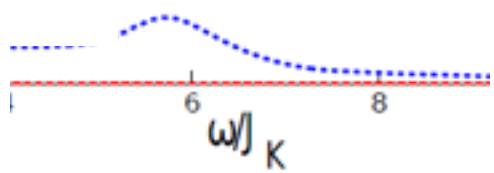
$$I_H^{[0]}(\omega) = 2\pi\delta(\omega - \Delta_F)\sqrt{\det(\mathcal{X}^\dagger\mathcal{X})}$$

Raman Scattering Results



Gupta, Singh et al. 2014





$RuCl_3$

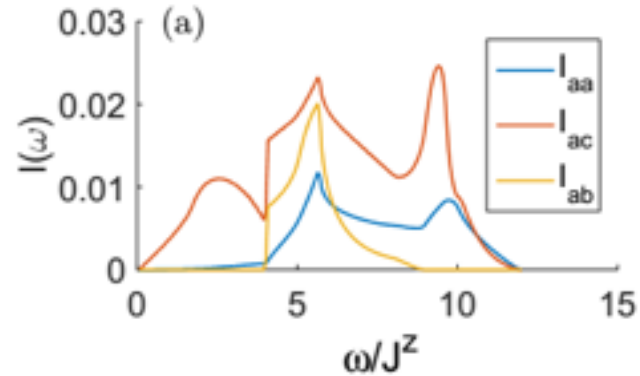
Y.J. Kim, K.S. Burch, et al., Phys. Rev. Lett. 114 (2015)

(interesting T-dependence)

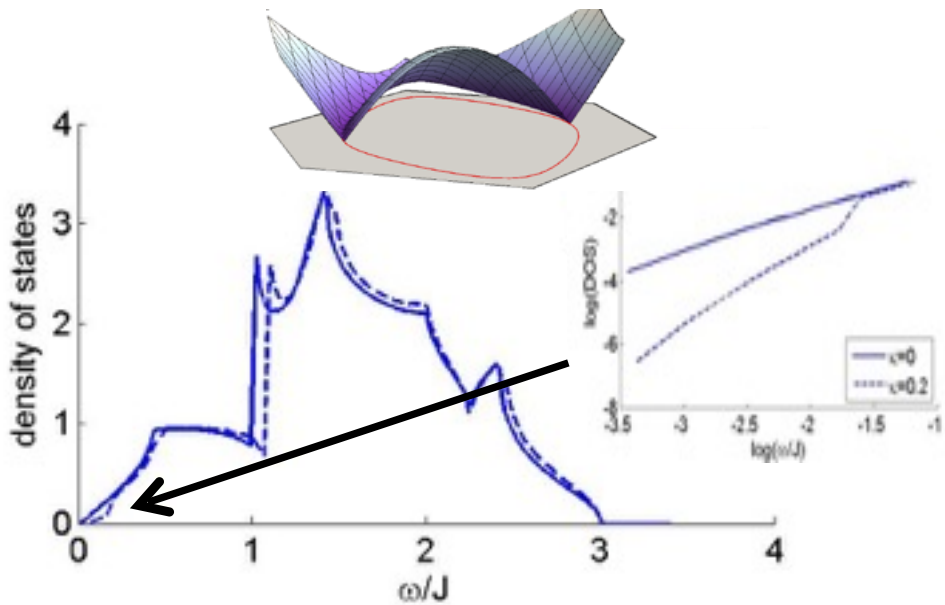
- Weak polarization dependence
- Sharp response at the four flux gap $\Delta_F = 0.446 J_K$
- Big **'hump'** with fine features of the Majorana DOS

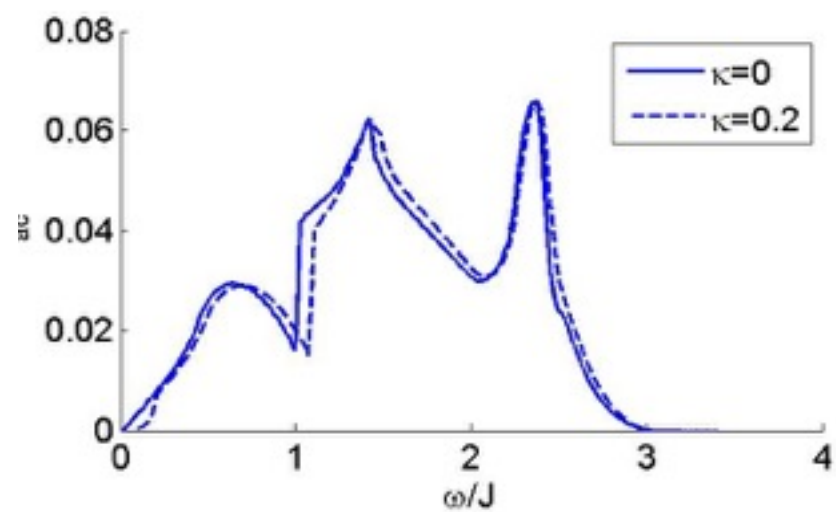
3D QSL Raman scattering

- Polarization dependence from inequivalent bonds.
  → distinguishable from phonons



- With time reversal breaking three spin interaction κ the low energy two spinon-DOS is changed → **signatures of the Weyl points**

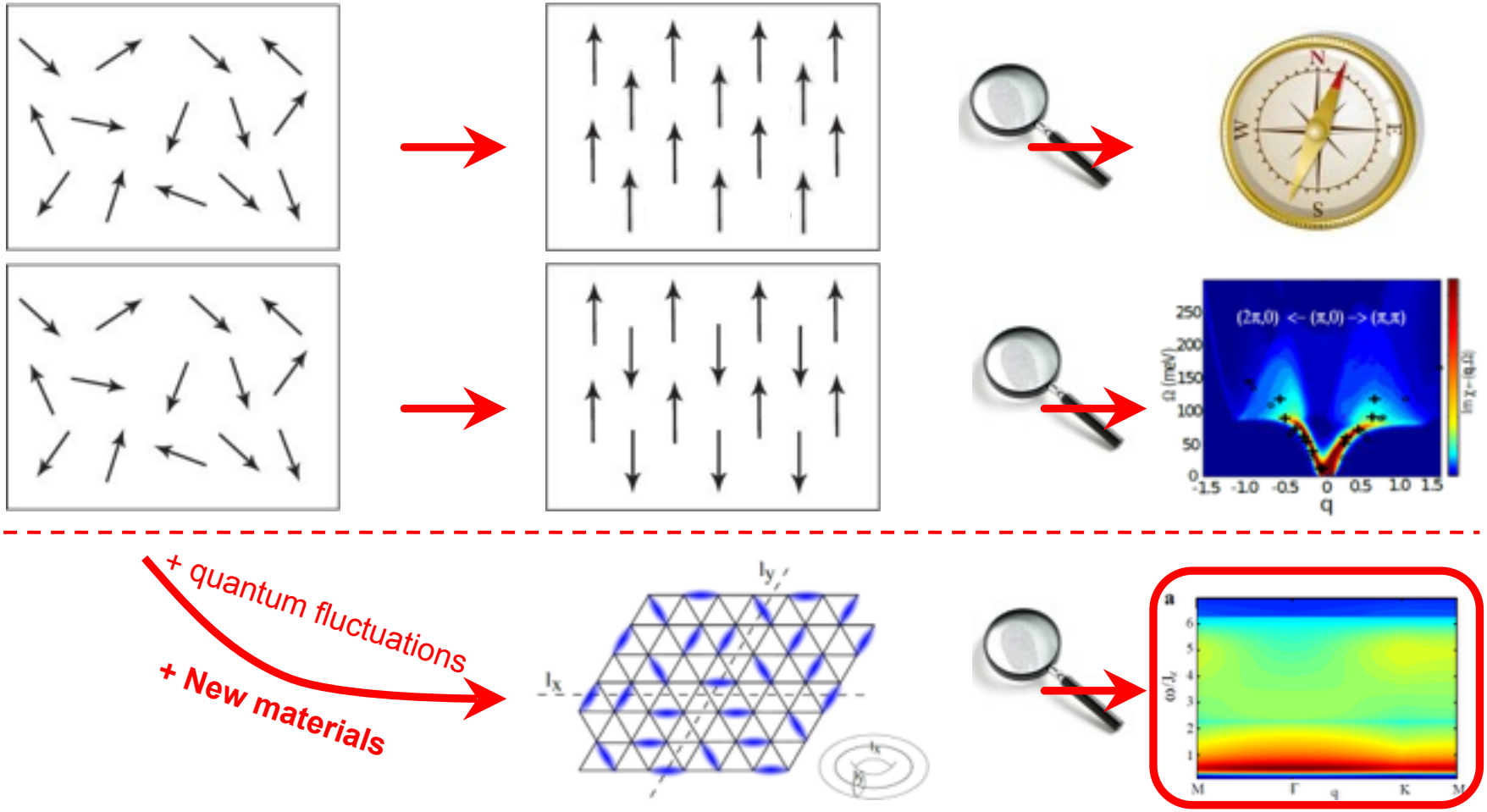




4. Conclusion

- Topologically ordered phases beyond the Landau paradigm:

  → QSLs are one example with recent 2D/3D candidate materials.



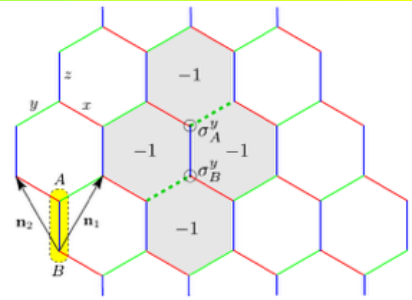
5. Conclusion

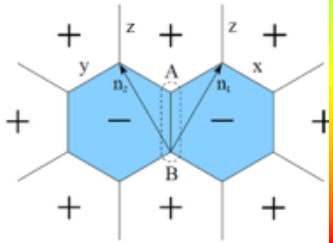
- Exact dynamical correlation functions of a 2D/3D quantum spin liquid
  - → Benchmark for numerics and approximations
  - → Connections to experiments e.g. Li/Na_2IrO_3 and $RuCl_3$

- Fractionalized d.o.f. show up as characteristic features in the response
  - → Flux gap + δ-peaks
  - → Majorana fermion DOS

- Non-equilibrium physics beyond the X-ray edge

- **INS, ESR and Raman experiments can diagnose spin liquids!**





Thank you!

Phys. Rev. Lett. 112, 207203 (2014)
featured with a *Viewpoint* in *Physics* by A. Tsvelik

Phys. Rev. Lett. 113, 187201 (2014)

arXiv:1504.08037
arXiv:1507.01639 (accepted in PRB)
arXiv:1507.02865 (accepted in PRB)
arXiv:1508.05324

more coming soon…