Probing continua of excitations in Kitaev spin liquids

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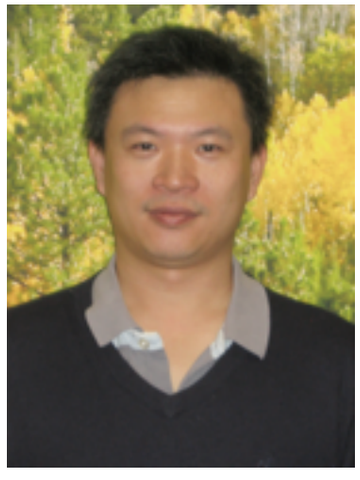
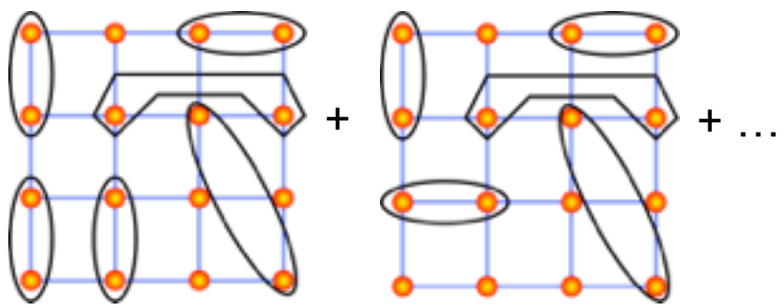


Gia-Wei Chern
University of Virginia

Quantum spin liquids

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

1973: Anderson proposes the “Resonating Valence Bond” state - a prototype of the modern QSLs

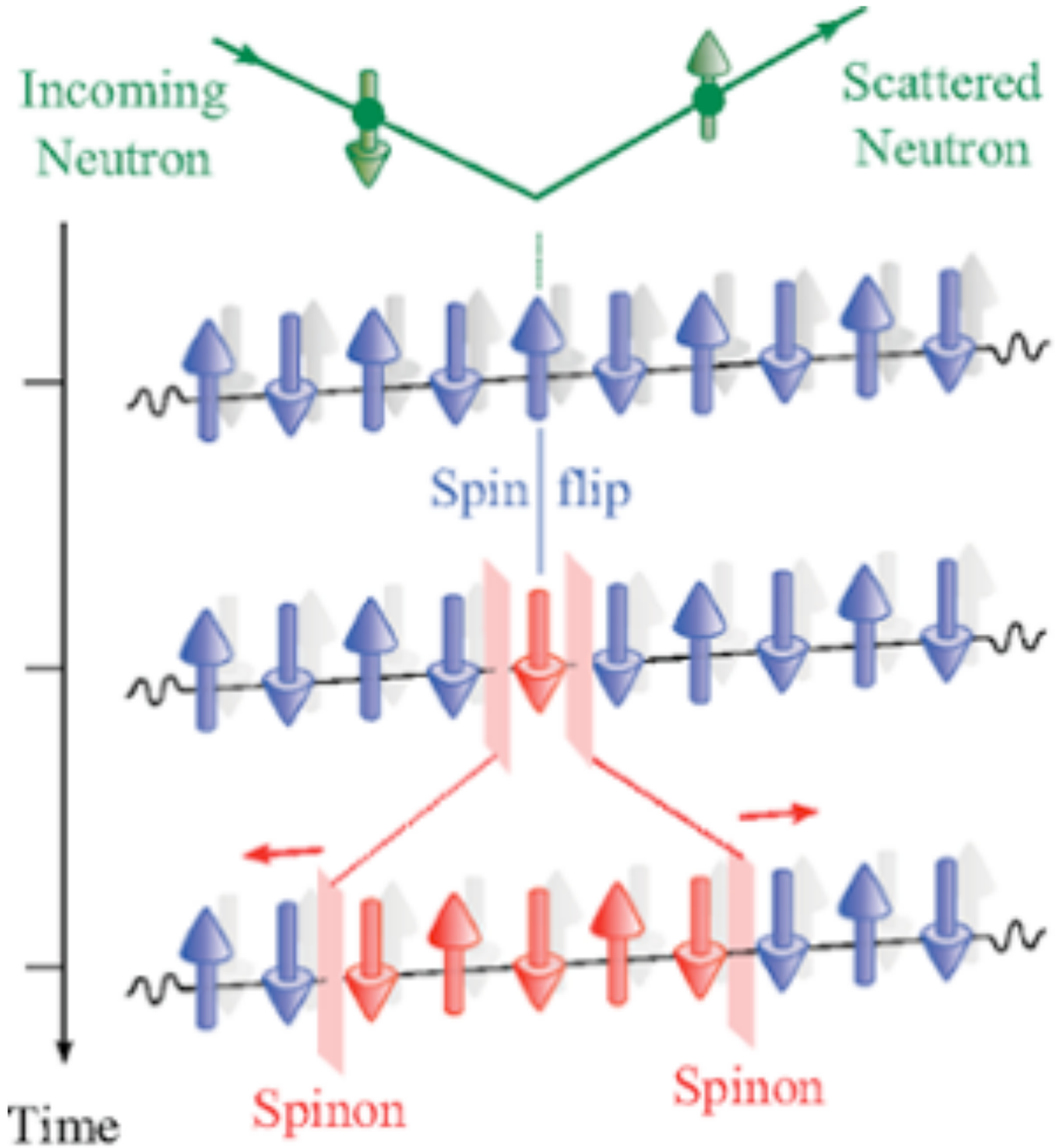
$$\Psi =$$  $+ \; + \ldots$

Unlike states with broken symmetry, QSLs are not characterized by any local order parameter.

QSLs are characterized by topological order and long range entanglement (difficult to probe experimentally).

QSLs supports excitations with *fractional quantum numbers and statistics*.

Example: Fractionalized excitations in spin-1/2 Heisenberg AFM chain



Mourigal et al, Nature Physics 2013

Many different quantum spin liquids

- Topological gapped QSLs

  Quantum dimer model, toric code model…

- Spinon Fermi surface QSLs

  Triangular lattice quantum spin liquid (YbMgGaO4)

- Variety of Kitaev gapless QSLs with nodal Majorana fermion band structures

  Hyperhoneycomb with nodal lines of Dirac cones,
  hyperoctagon with Majorana fermions Fermi surface..

- U(1) QSL with gapless emergent photon

  Quantum spin ice

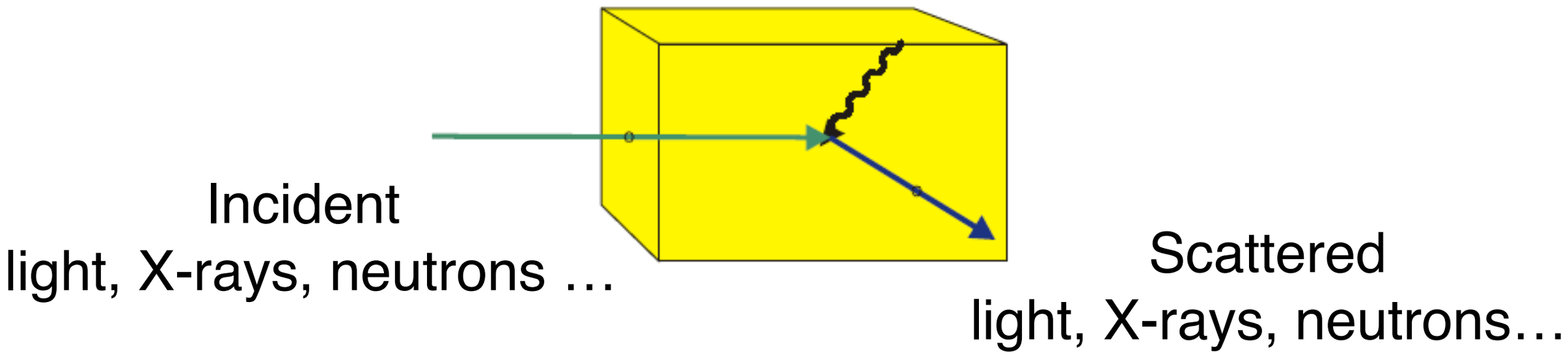
Quantum spin liquids

Main Question:

How to probe QSLs and their statistics?

Take home message:

Signatures of quantum order are mainly in the excitations

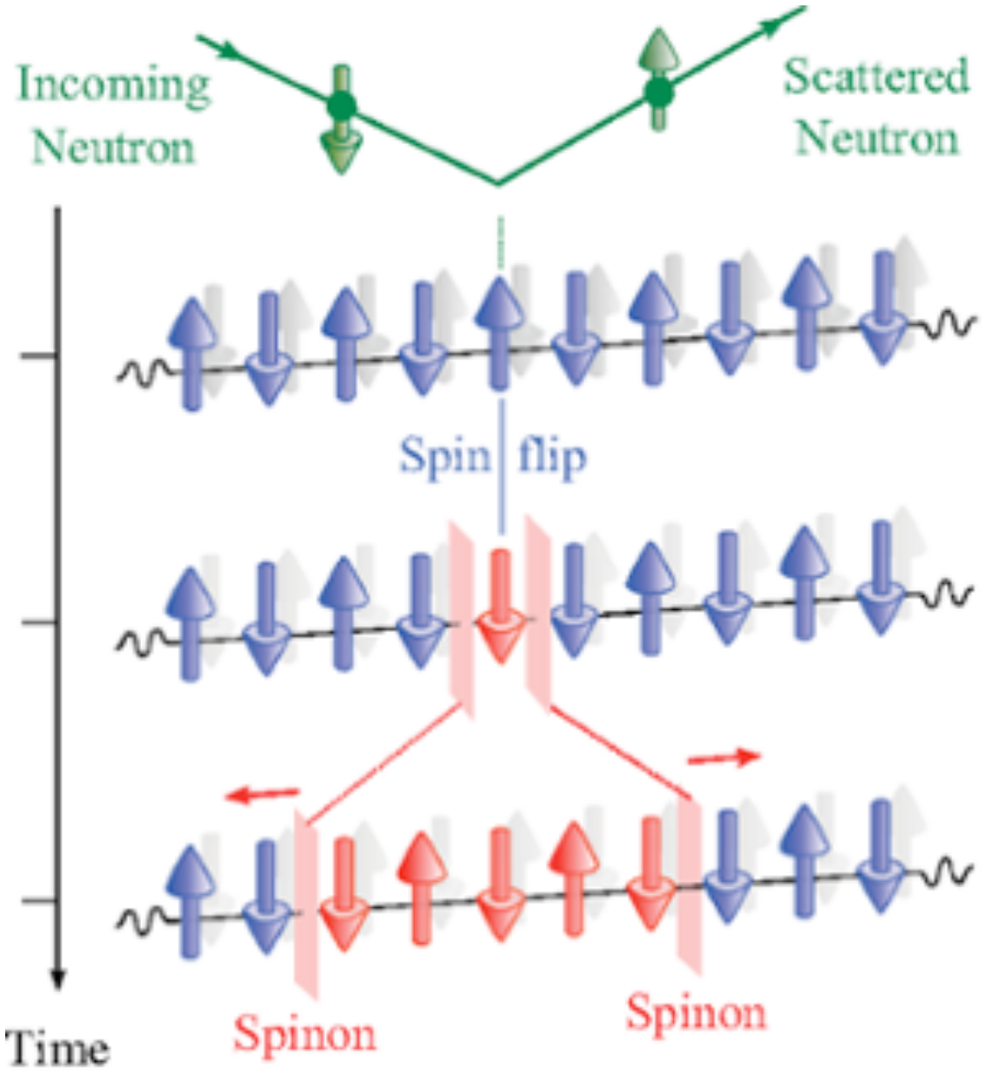


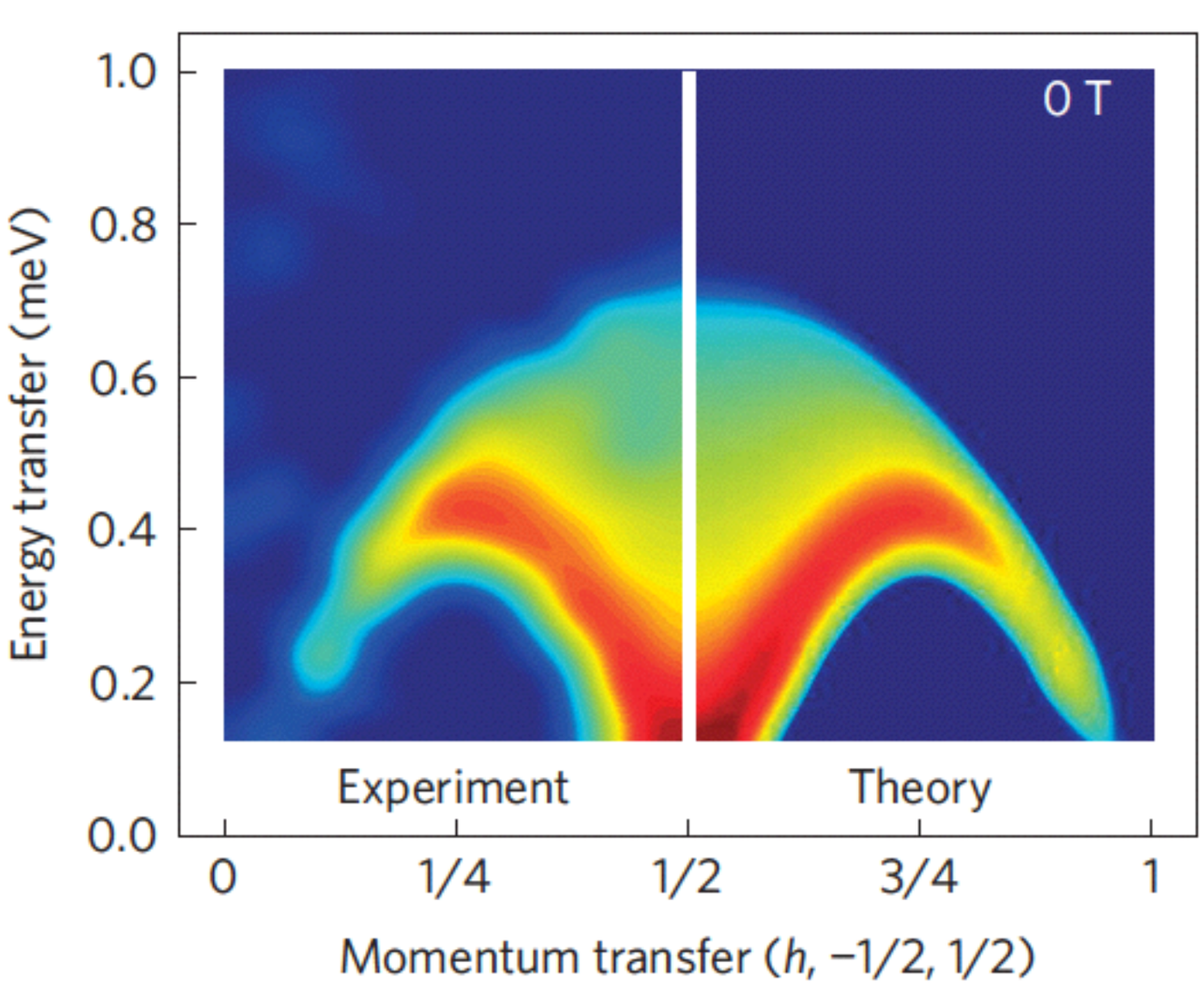
Response from QSL is always a multi-particle continuum

Since excitations carry fractional quantum numbers relative to the local degrees of freedom, only multiple quasiparticles can couple to external probes.

Spinon excitations in spin-1/2 Heisenberg AFM chain

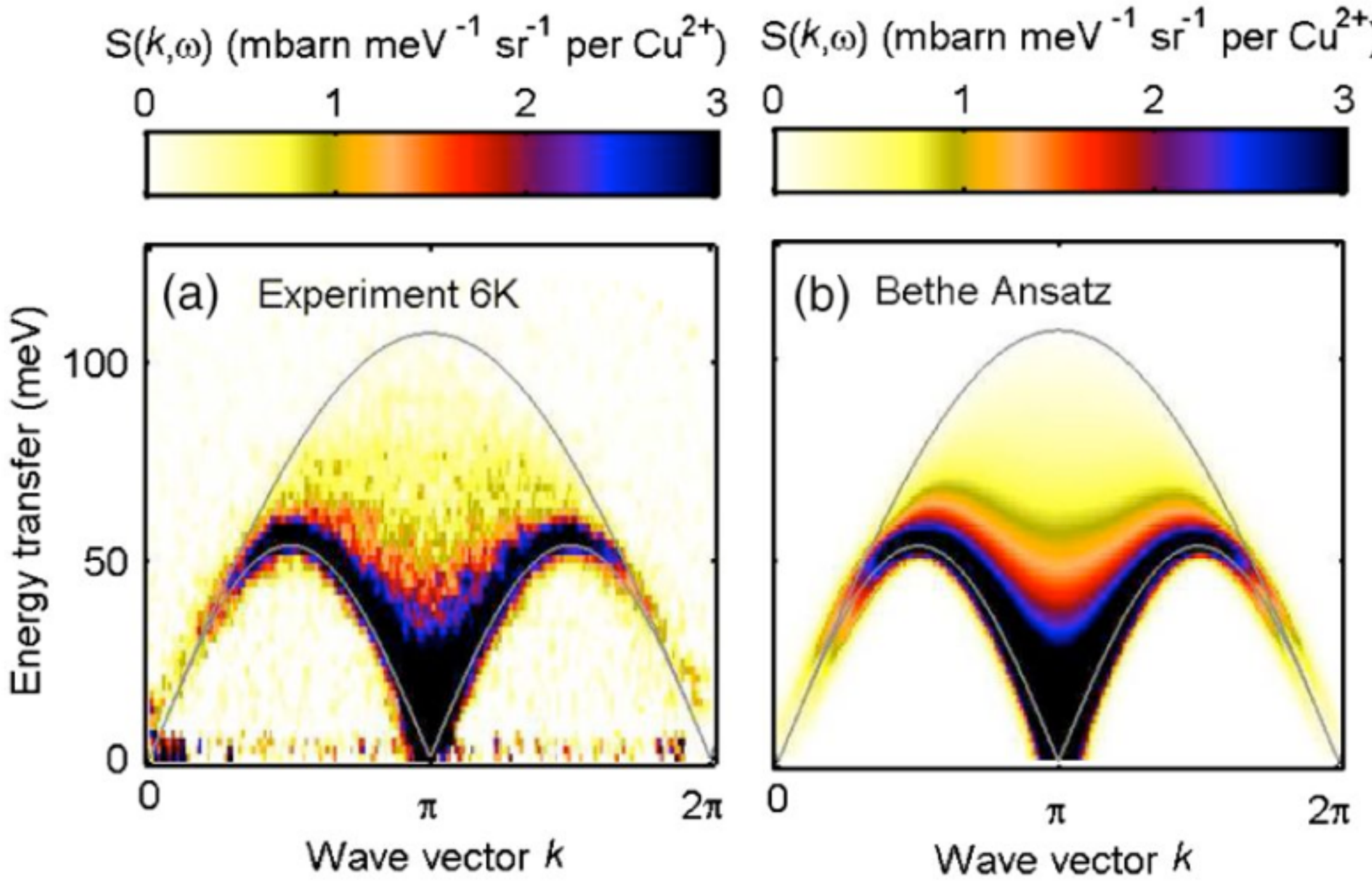
$CuSO_4 \cdot 5D_2O$,





Mourigal et al, Nature Physics 2013

Spinon excitations probed by neutrons: $KCuF_3$



The fractionalization was definitively identified by excellent **quantitative** agreement between experiments and exact calculation based on the Bethe Ansatz.

Lake, Caux, Tennant 2013

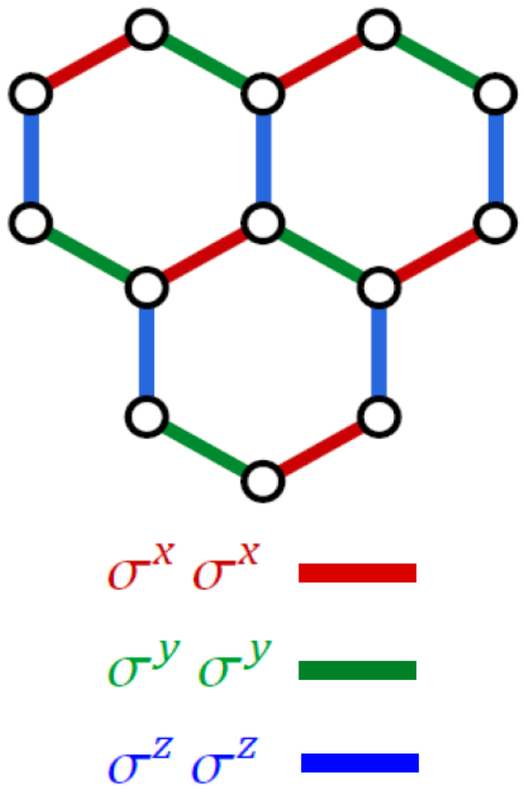
Poor understanding of strongly interacting systems beyond 1D

No exact results, only numerics or uncontrolled approximations

Kitaev Spin Liquids

Kitaev model on the honeycomb lattice

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



Exactly solvable 2D model

Spin liquid ground state

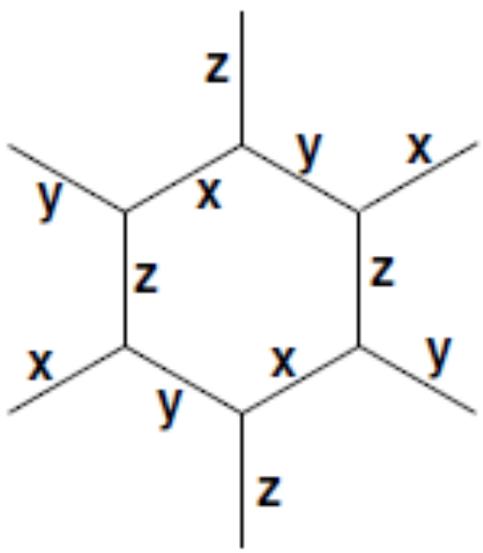
Fractionalized excitation

Mapping spins to Majorana fermions:

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

A. Kitaev, Annals of Physics **321**, 2 (2006)

Spin fractionalization and Majorana fermions



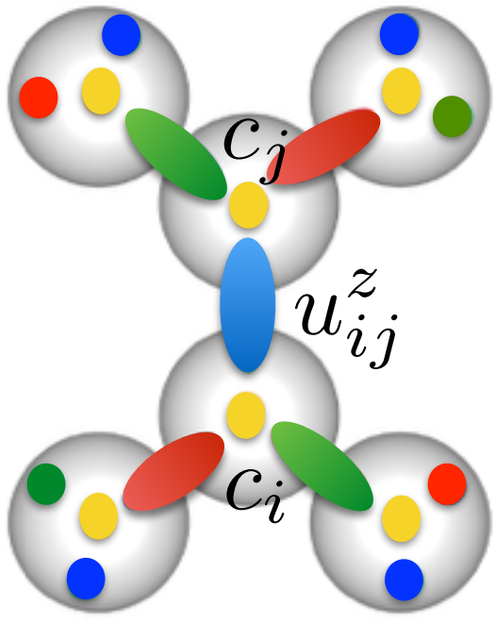
Large number of conserved quantities, local plaquette operators:

$$\tilde{W}_p = \hat{\sigma}_1^x \hat{\sigma}_2^y \hat{\sigma}_3^z \hat{\sigma}_4^x \hat{\sigma}_5^y \hat{\sigma}_6^z$$

$$[\tilde{W}_p, \hat{H}] = 0 \quad [\tilde{W}_p, \tilde{W}_p'] = 0$$

The Hilbert space can be separated into sectors corresponding to eigenvalues

$$W_p = \pm 1$$



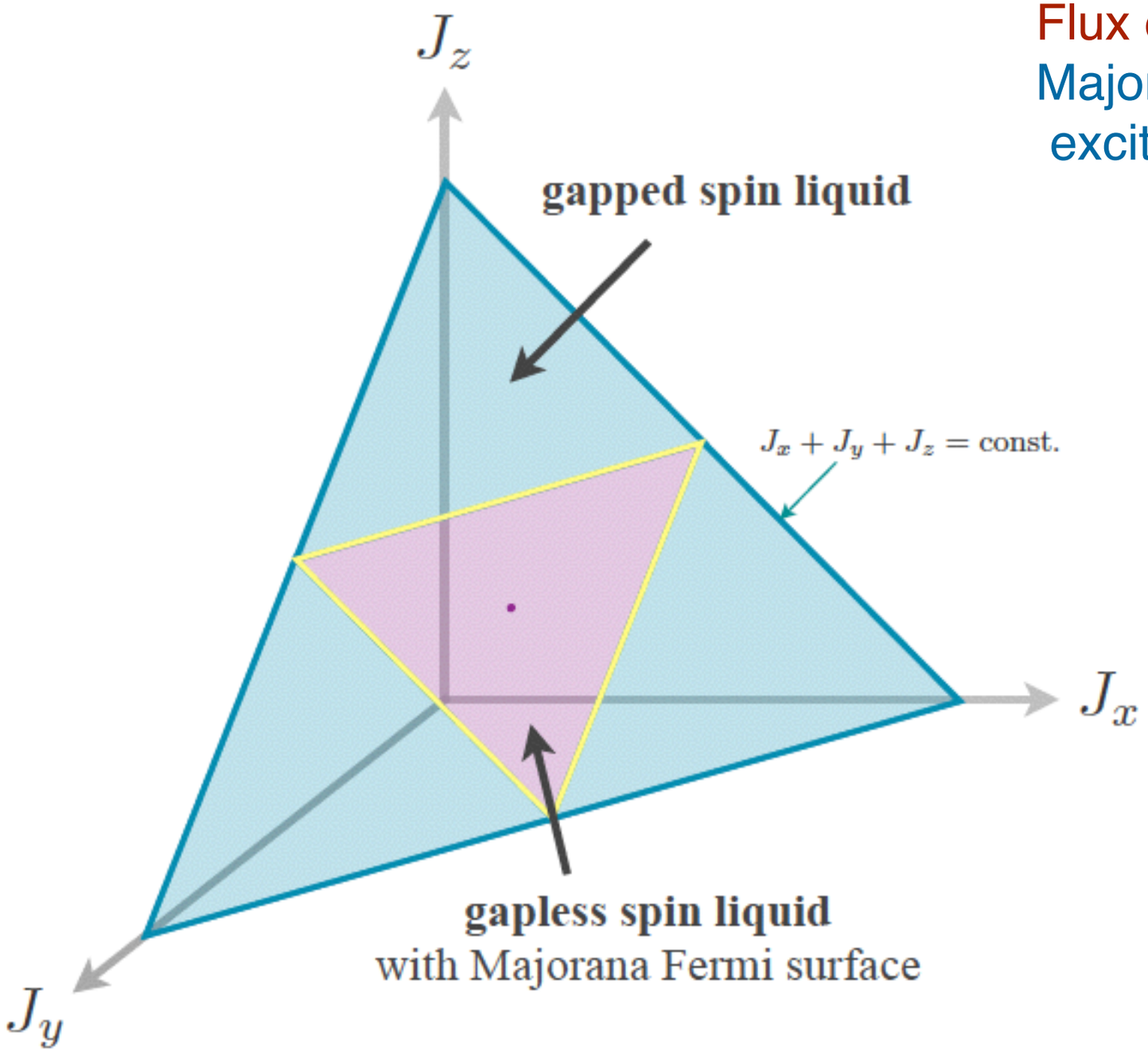
Quadratic Hamiltonian in each flux sector:

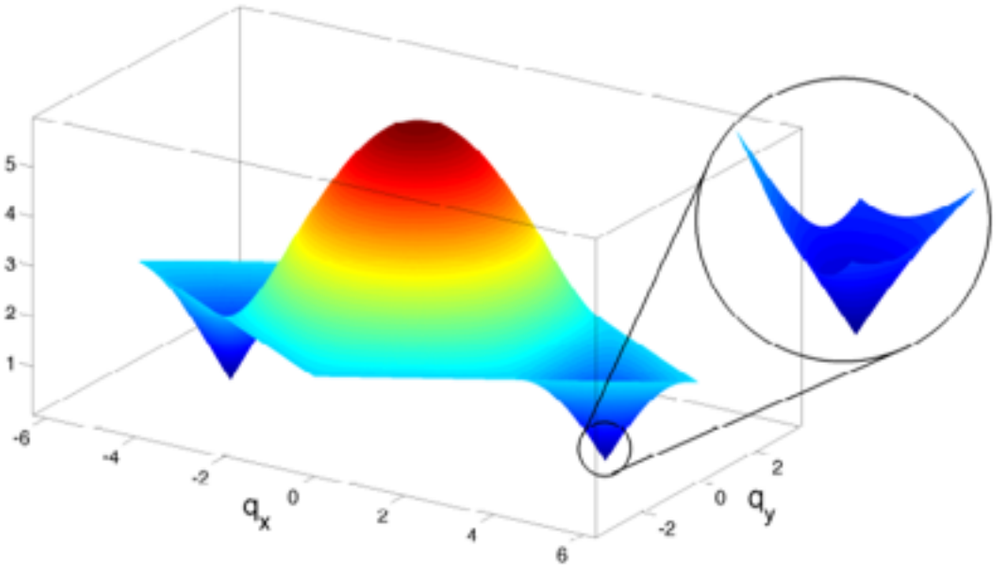
$$H = -\sum_{a=x,y,z} J_a \sum_{\langle ij \rangle_a} i c_i \hat{u}_{\langle ij \rangle_a} c_j$$

$$\hat{u}_{\langle ij \rangle_a} \equiv i c_i^a c_j^a$$

Excitations in the 2D Kitaev spin liquid

Flux excitations are always gapped.
Majorana fermion (spinon) excitations are gapped or gapless.

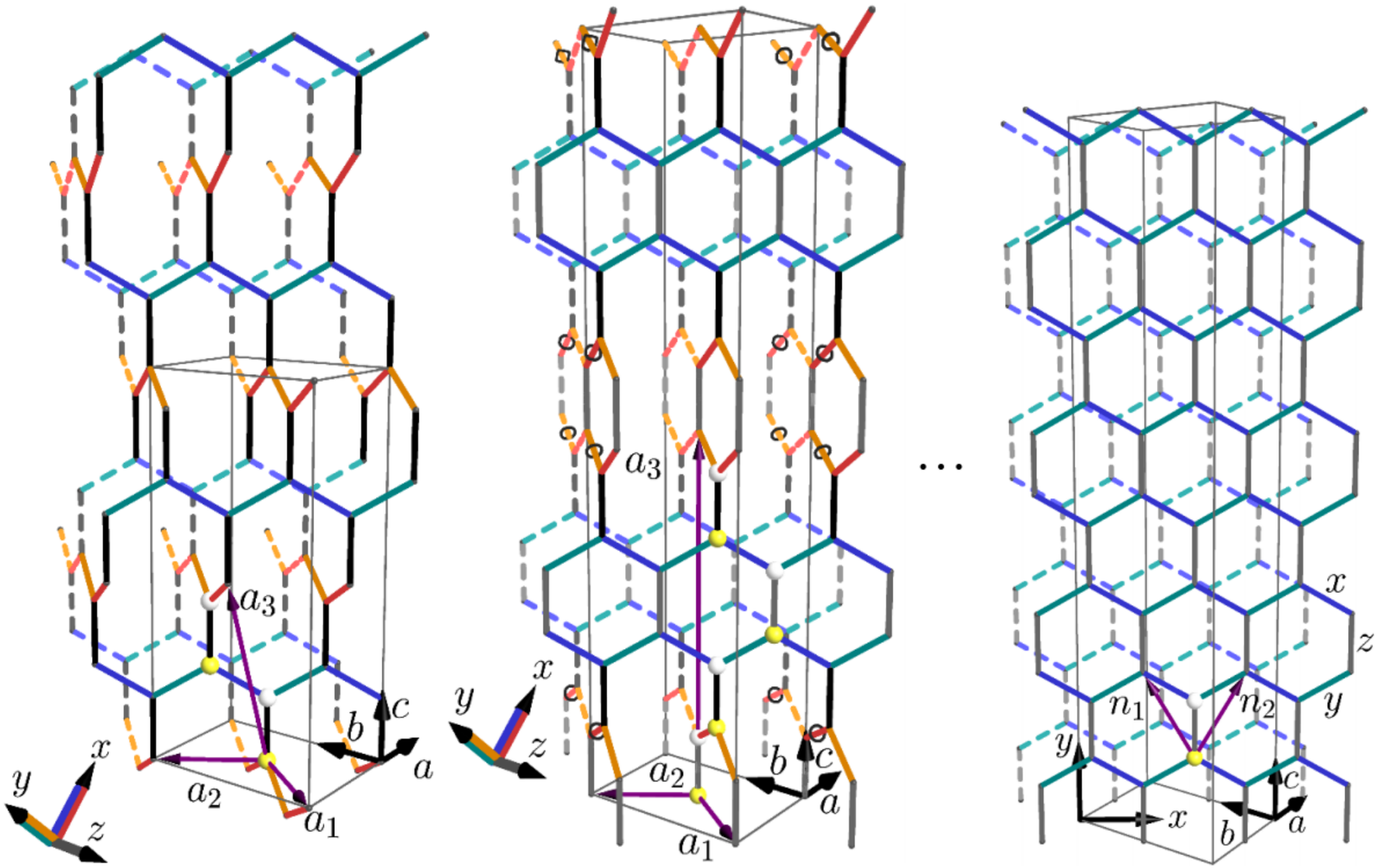




Majorana fermion excitation spectrum in the gapless phase

Fig. from M. Hermanns et al, 2014

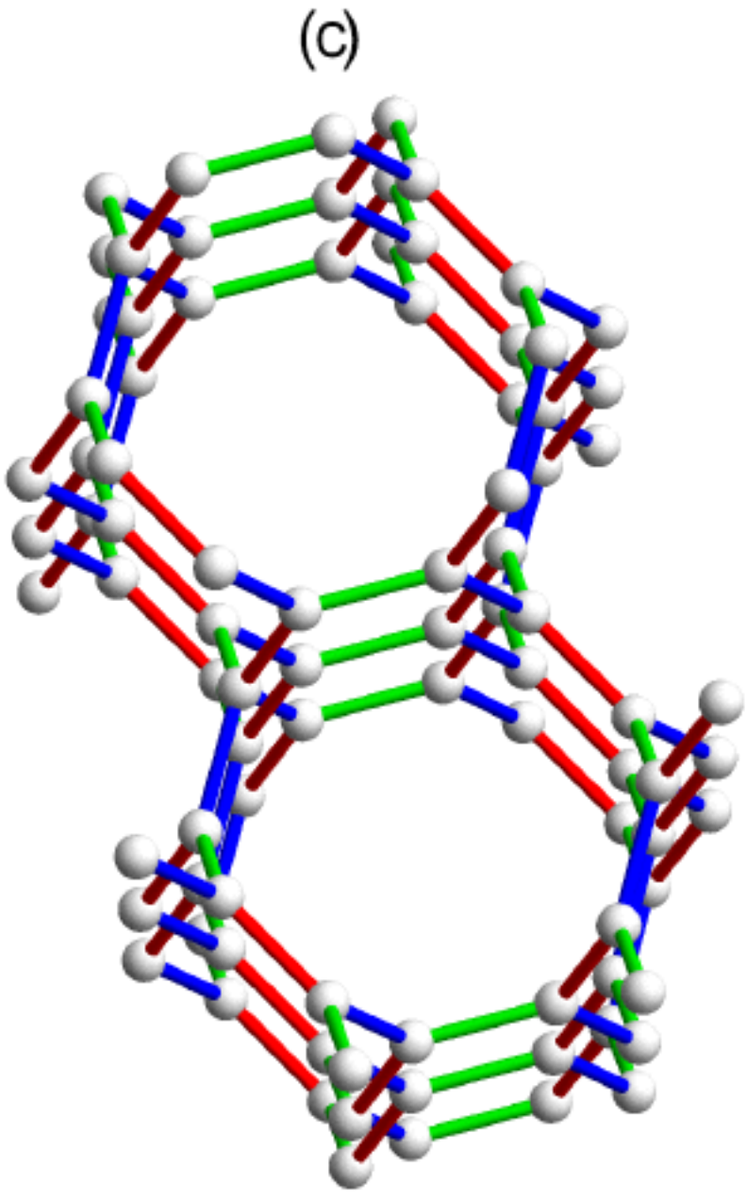
3D Kitaev family



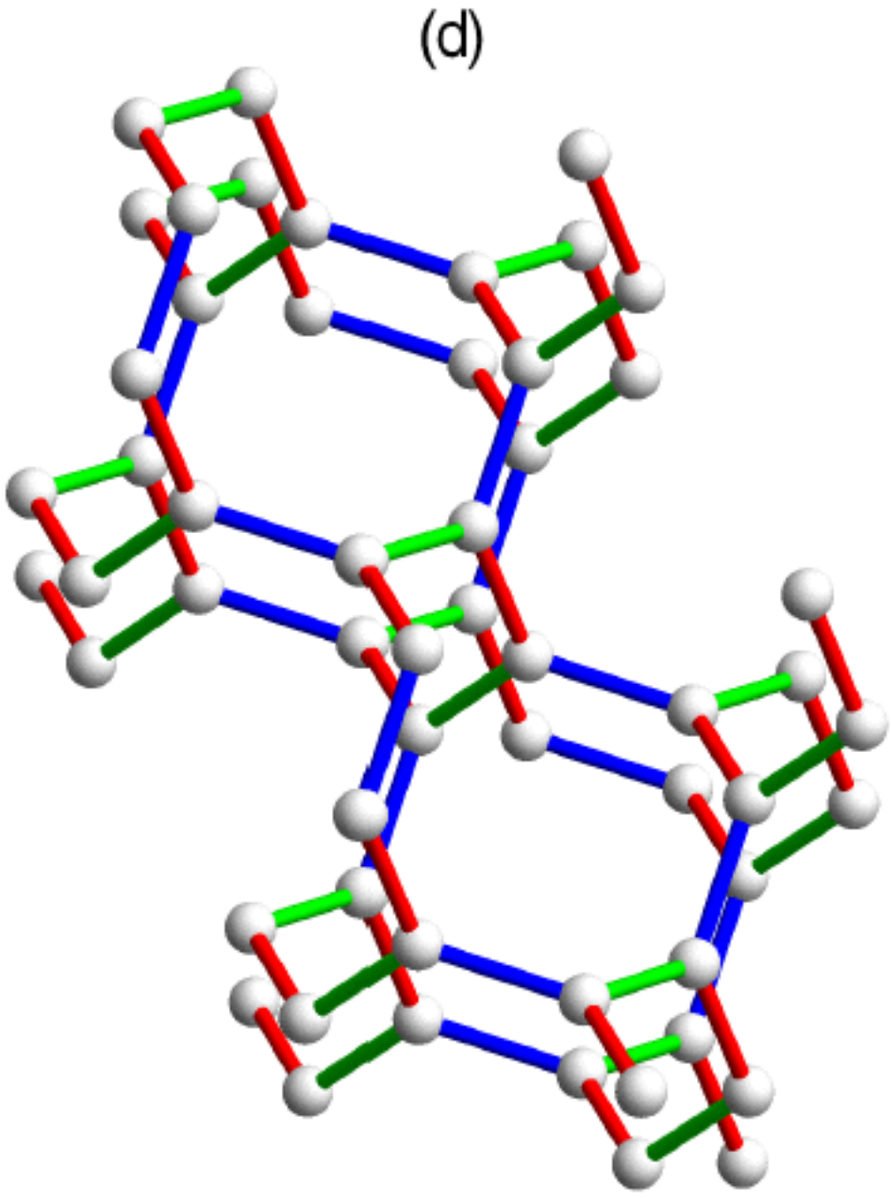
$H_{<0>}$ $H_{<1>}$ $H_{<\infty>}$

Hyperhoneycomb lattices

3D Kitaev family



Hyperhexagon

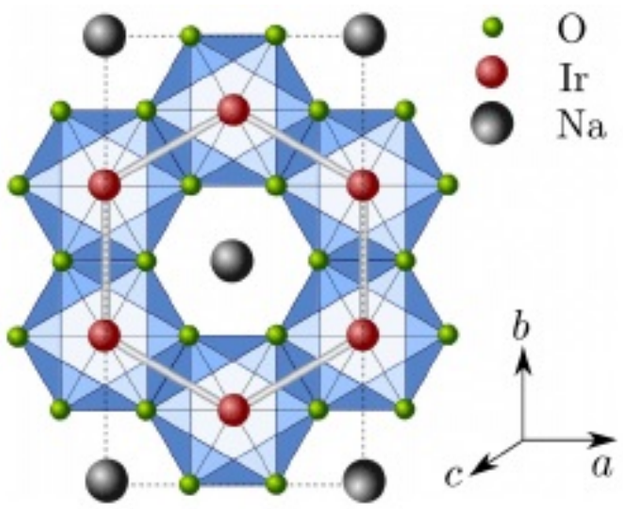


Hyperoctagon

M. Hermanns et al, 2015

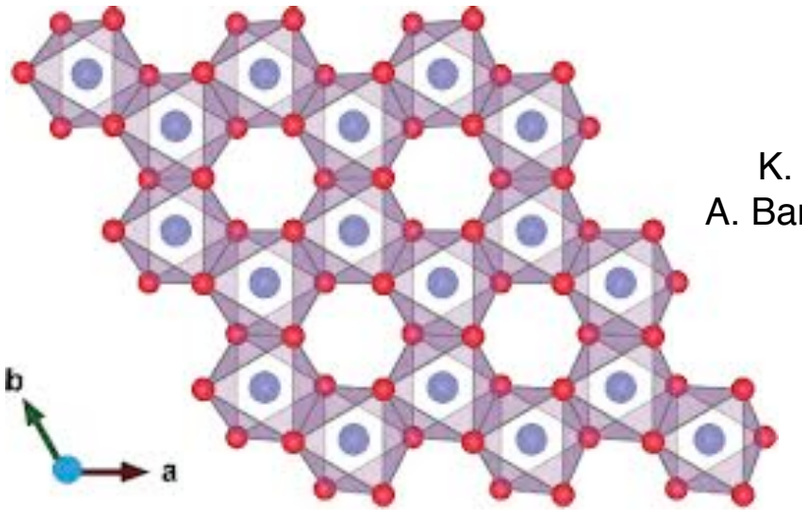
Experimental realizations

Na_2IrO_3
alpha-Li_2IrO_3



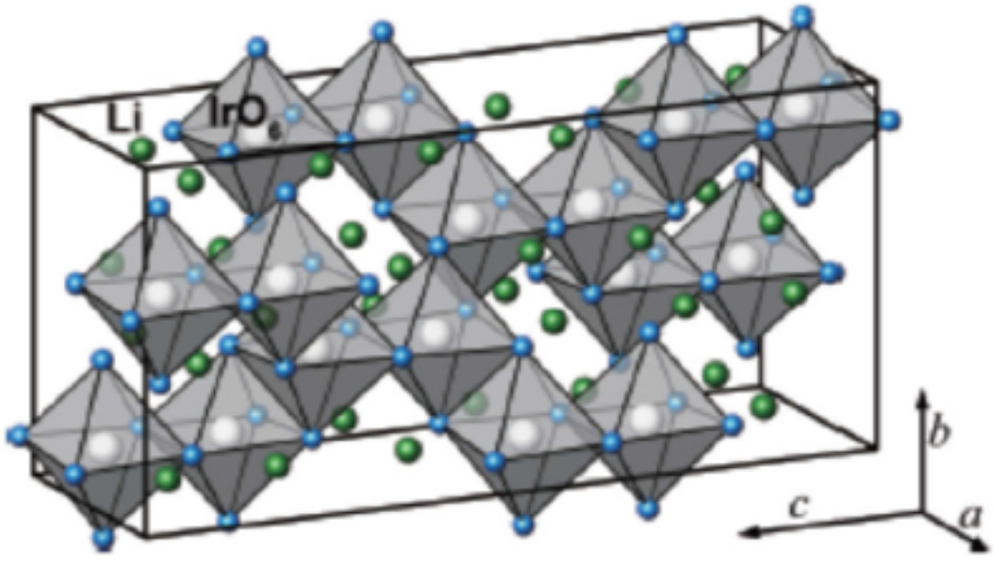
Y.Singh, P. Gegenwart,
PRL 2010, 2011

alpha-$RuCl_3$



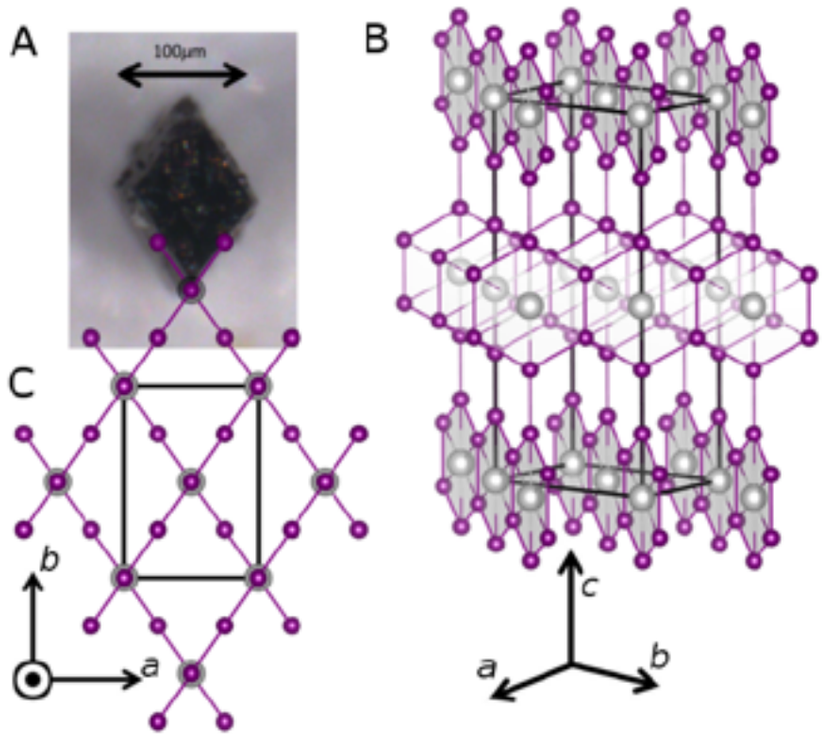
K. Plumb et al, Phys. Rev. B (2014)
A. Banerjee et al, Nature Materials (2016)

beta-Li_2IrO_3



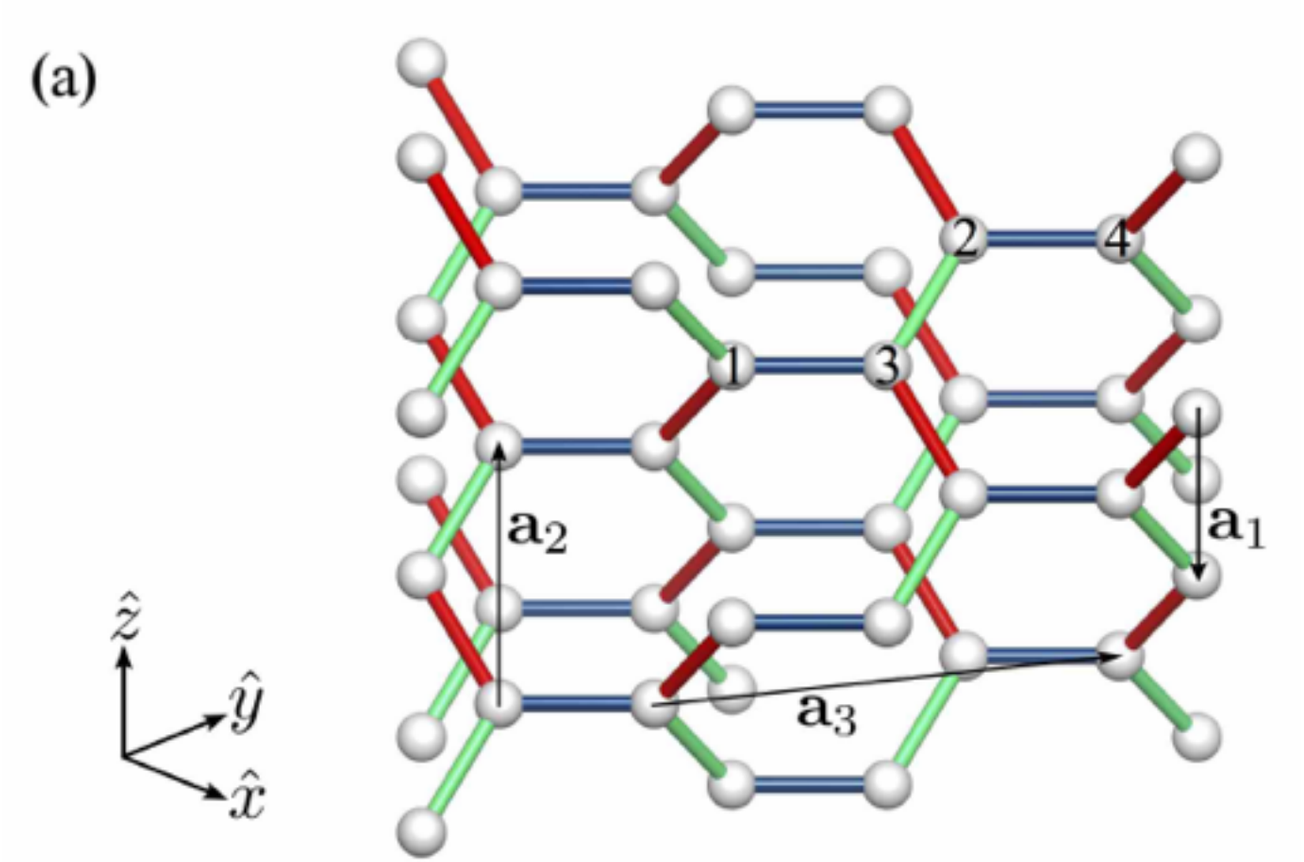
T. Takayama et al, PRL (2015)

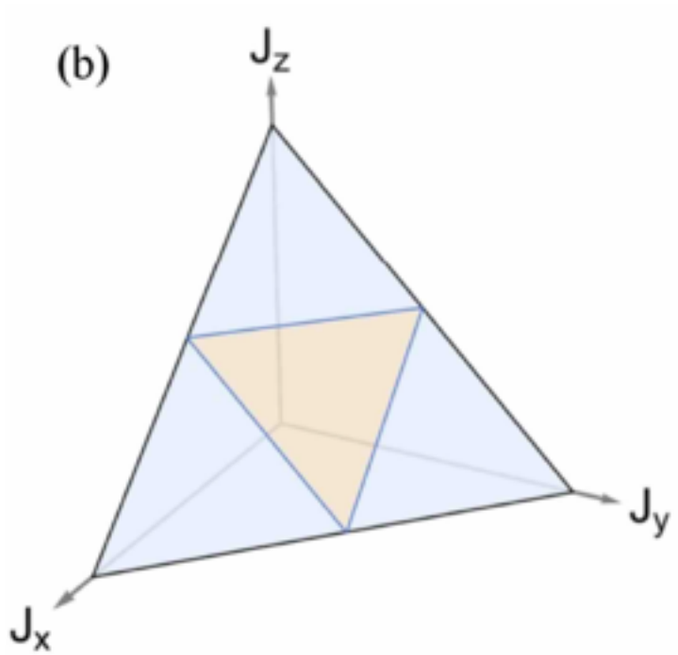
gamma-Li_2IrO_3



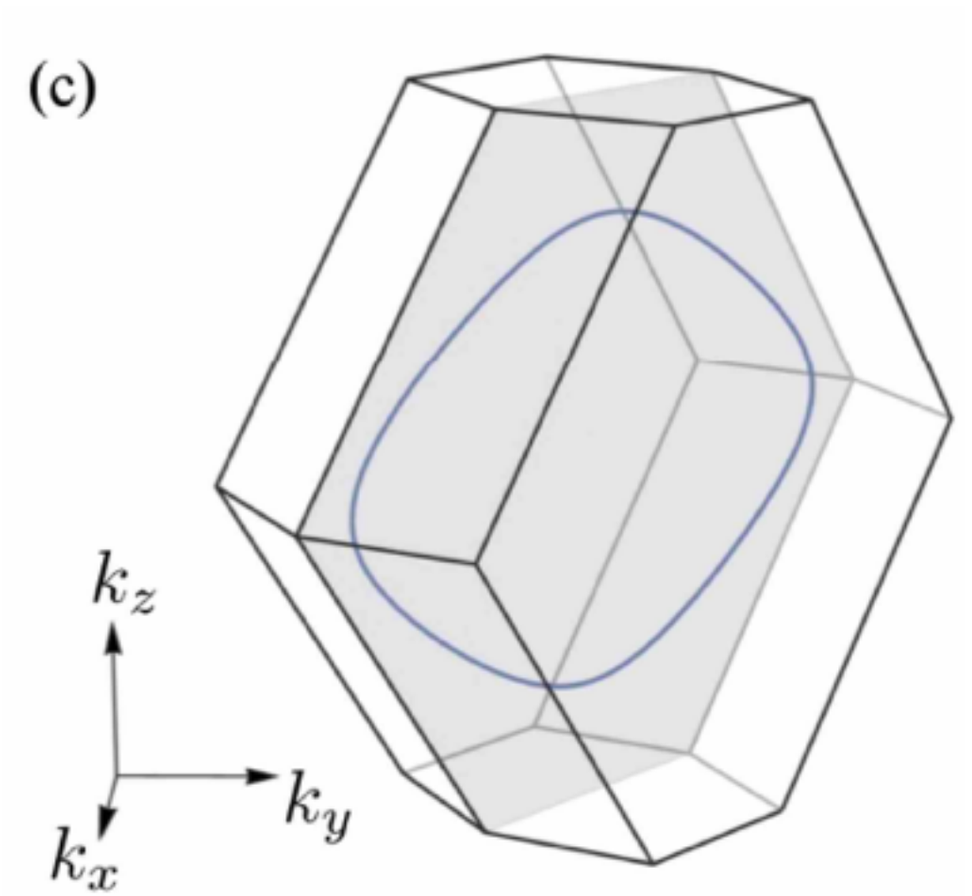
Modic, Nature Comminications **5**, 4203 (2014)

Hyperhoneycomb lattice





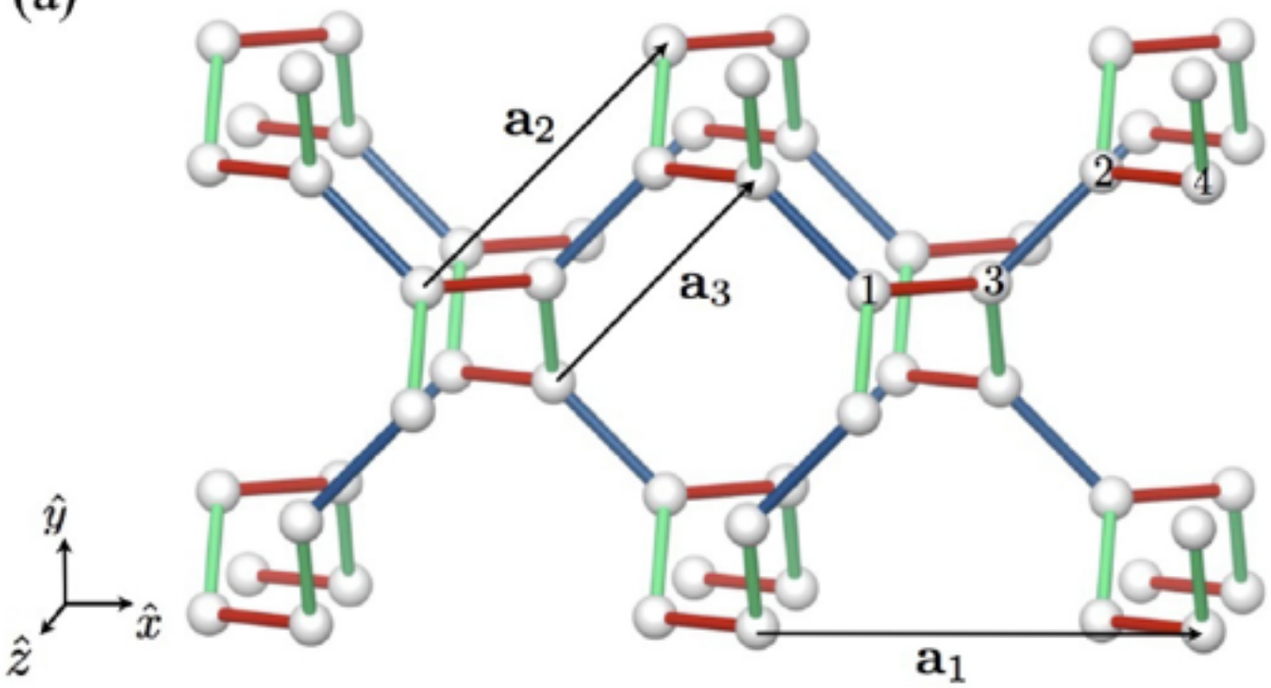
closed line of Dirac nodes



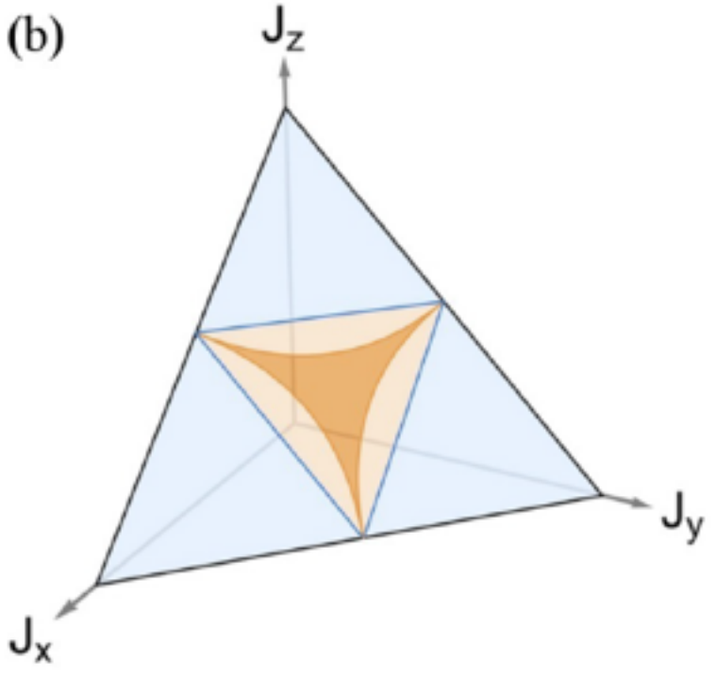
M. Hermanns et al, 2015

Hyperoctagon lattice
Majorana metal

(a)

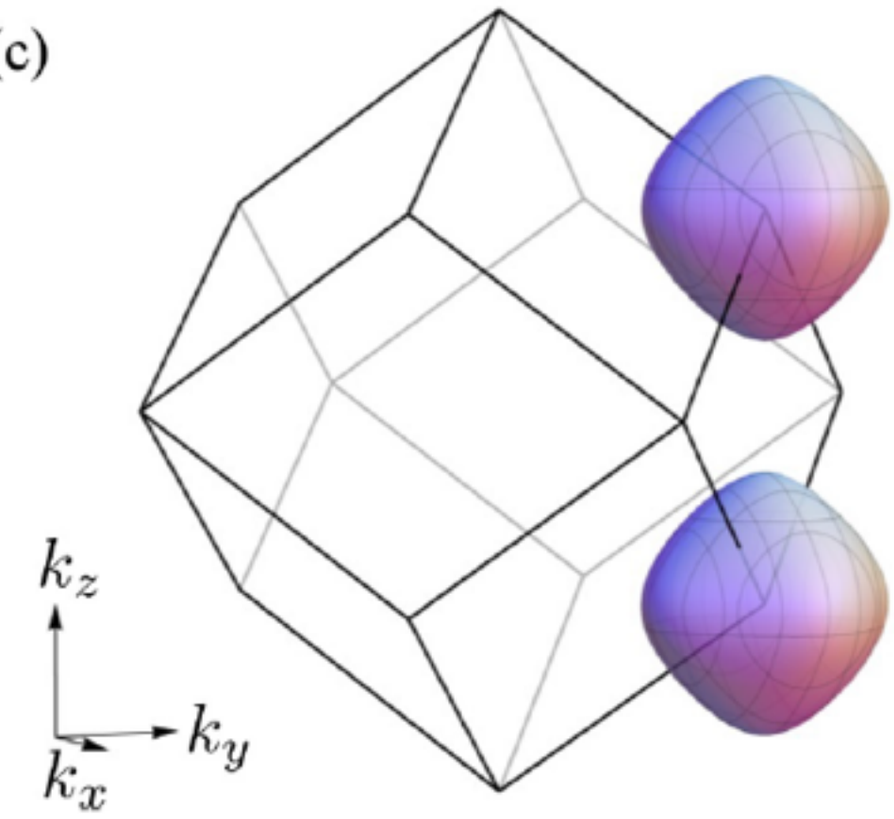


(b)



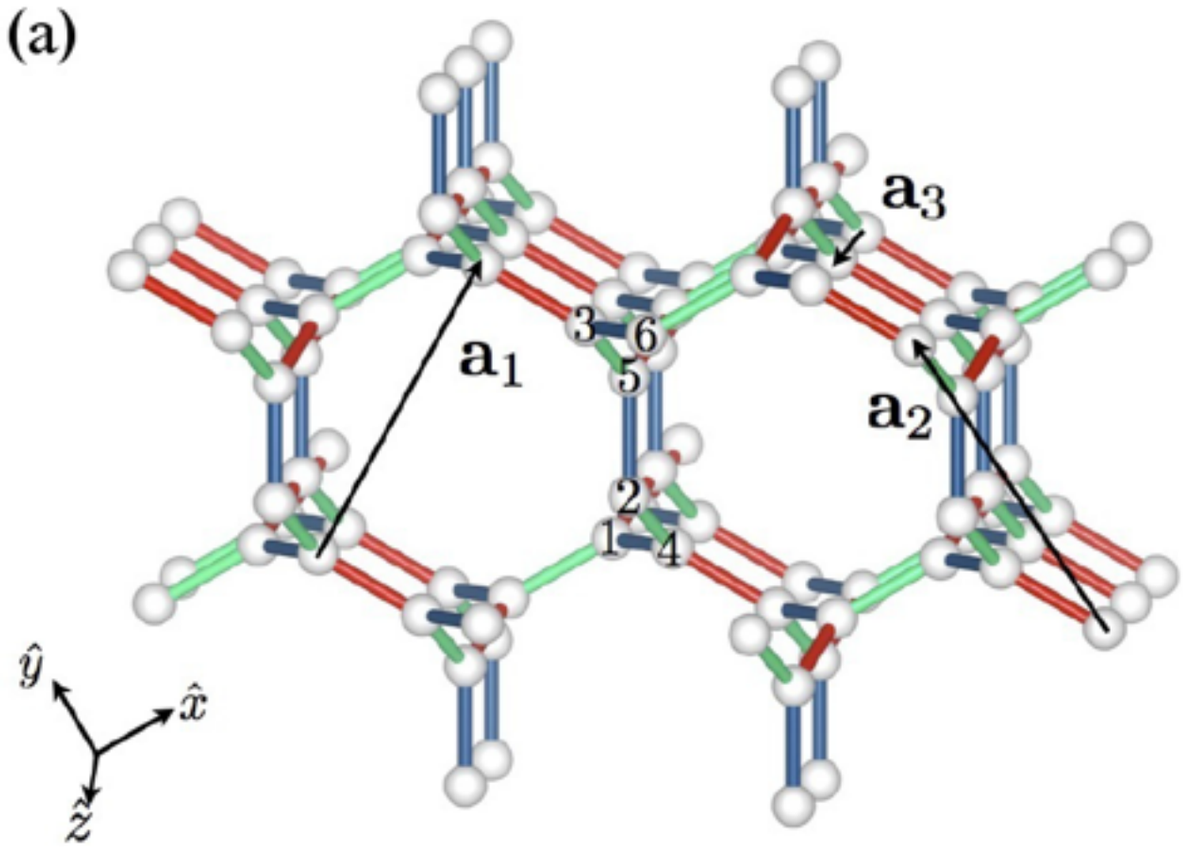
Fermi surfaces

(c)

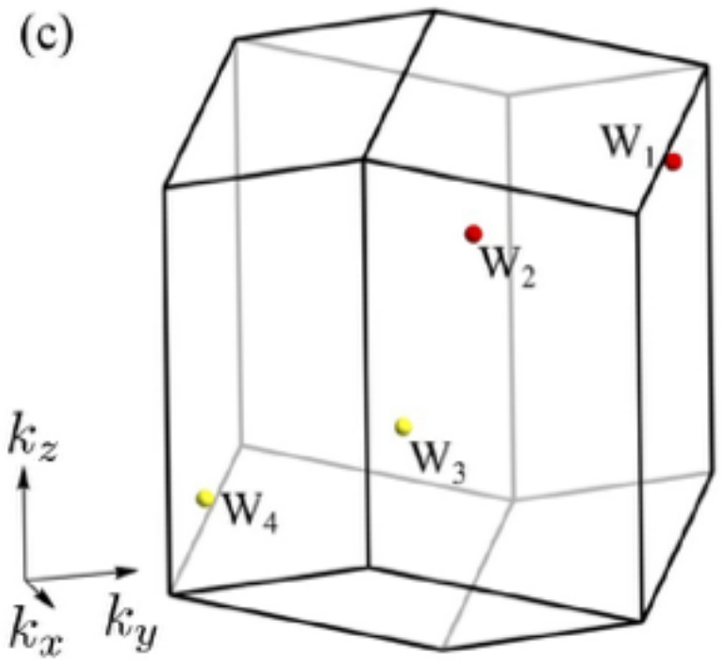


M. Hermanns et al, 2015

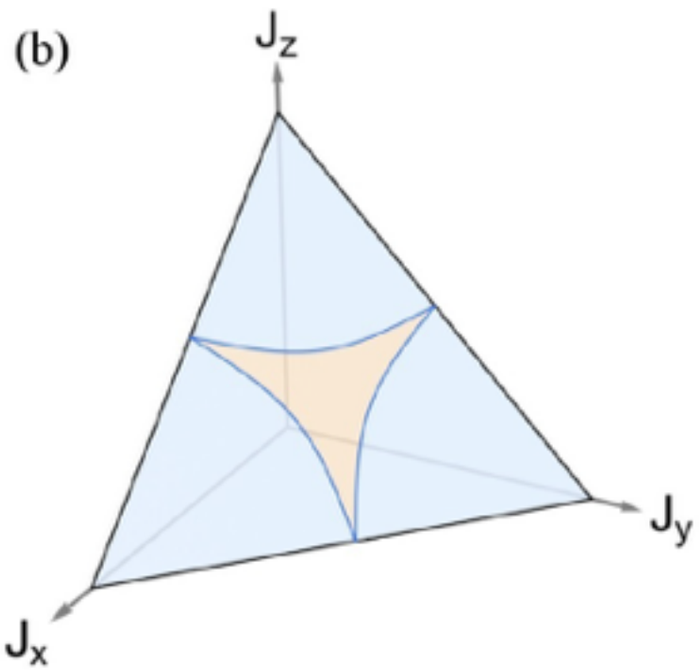
Hyperhexagon lattice



gapless Weyl points



$$\hat{H}_{2\times 2} = \mathbf{v}_0 \cdot \mathbf{q}\,\mathbb{1} + \sum_{j=1}^{3} \mathbf{v}_j \cdot \mathbf{q}\,\sigma_j$$



M. Hermanns et al, 2015

Spectroscopy of Kitaev Spin Liquids

INS of Kitaev Spin Liquids (briefly)

Raman scattering in Kitaev Spin Liquids (briefly)

RIXS scattering in Kitaev Spin Liquids

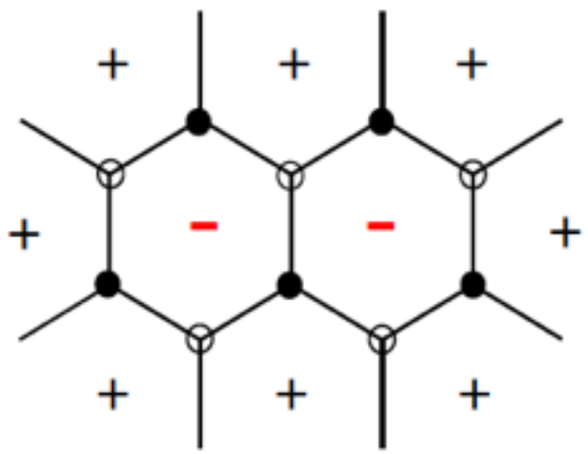
Inelastic neutron scattering of Kitaev Spin Liquids

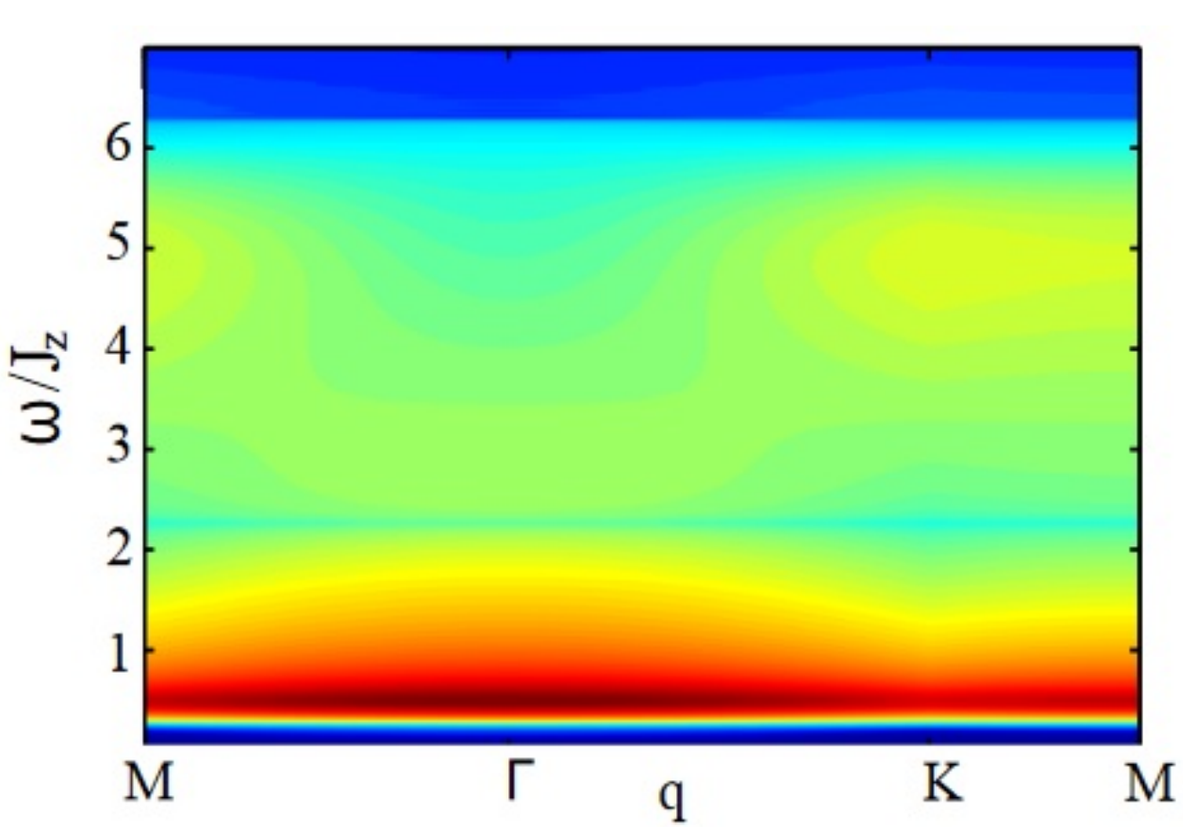
QSL: expected spin-excitation continuum vs sharp dispersive features for spin-waves

$$S^{aa}_{\mathbf{q}}(\omega) = \frac{1}{N}\sum_{ij} e^{-i\mathbf{q}(\mathbf{r}_i - \mathbf{r}_j)} \int_{-\infty}^{\infty} dt e^{i\omega t} S^{aa}_{ij}(t),$$

$$S^{aa}_{ij}(t) = \langle \hat{\sigma}^a_i(t)\hat{\sigma}^a_j(0)\rangle$$

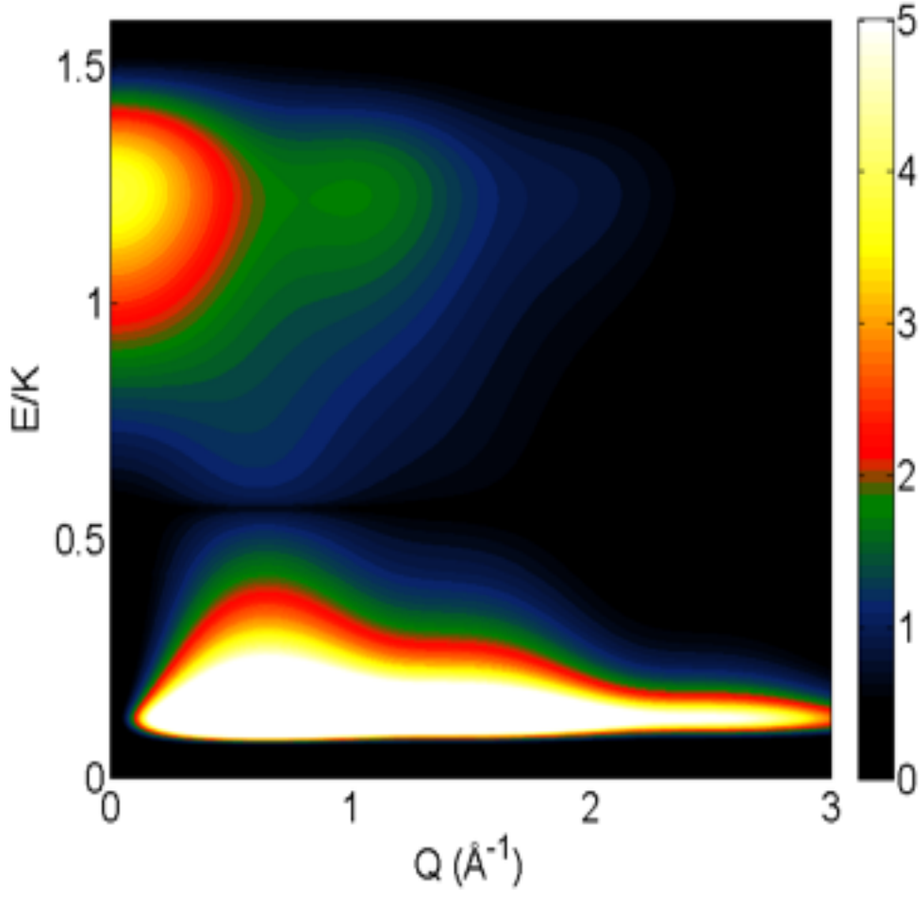
Measurement of a dynamic structure factor S(q,ω) leads to a sudden insertion of a pair of Z_2 gauge-fluxes

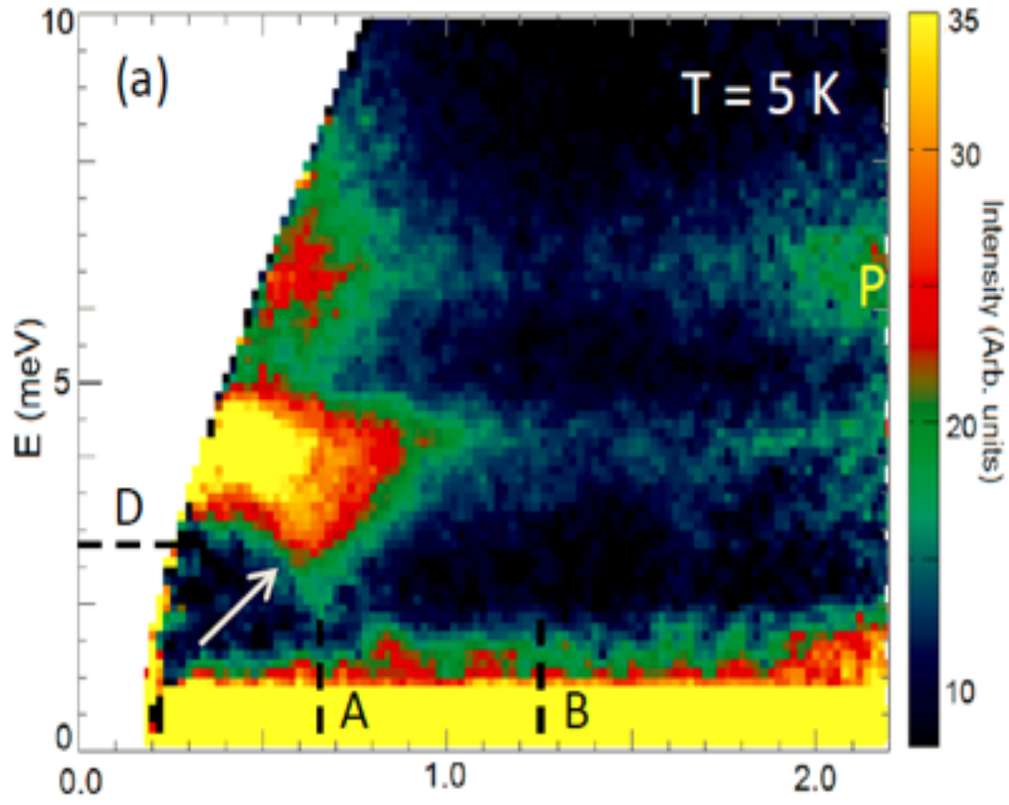




Knolle, Kovrizhin, Chalker, Moessner, *PRL* (2014)

$RuCl_3$



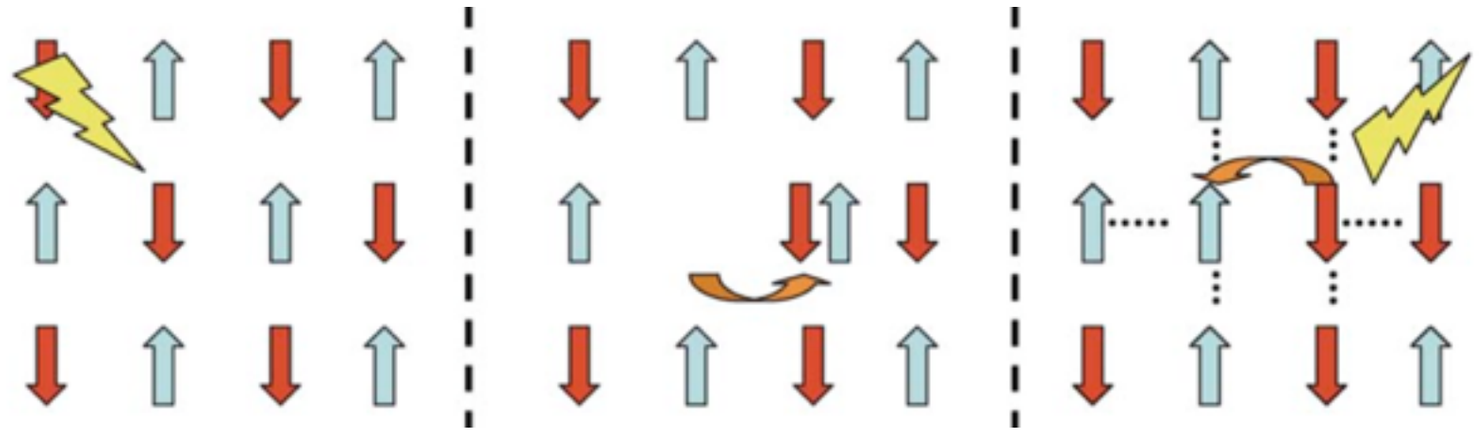


Signatures of fractionalization are visible!

Banerjee, et al., *Nature Mat.* (2016)
Banerjee, et al., Science (2017)

Raman spectroscopy of Kitaev Spin Liquids

Photon-in photon-out process

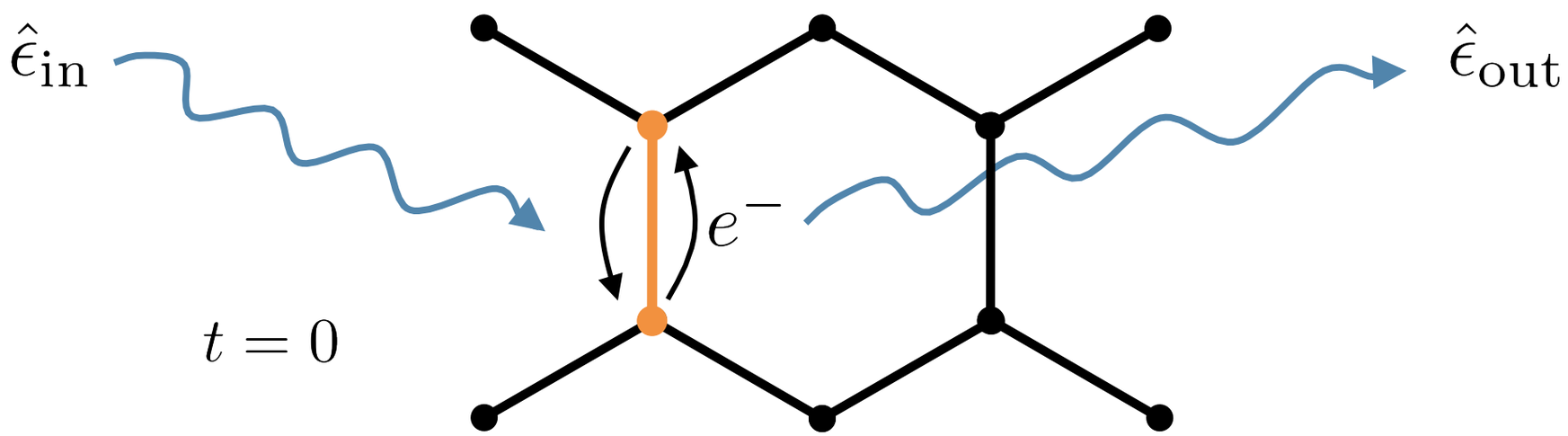


Photon induced spin exchange

Devereaux, Hackl *RMP* 2007

Raman Scattering in Kitaev model

$$I_{\mathrm{Ram}}(\omega) \propto \int dt e^{i\omega t} \langle R e^{iHt} R \rangle$$



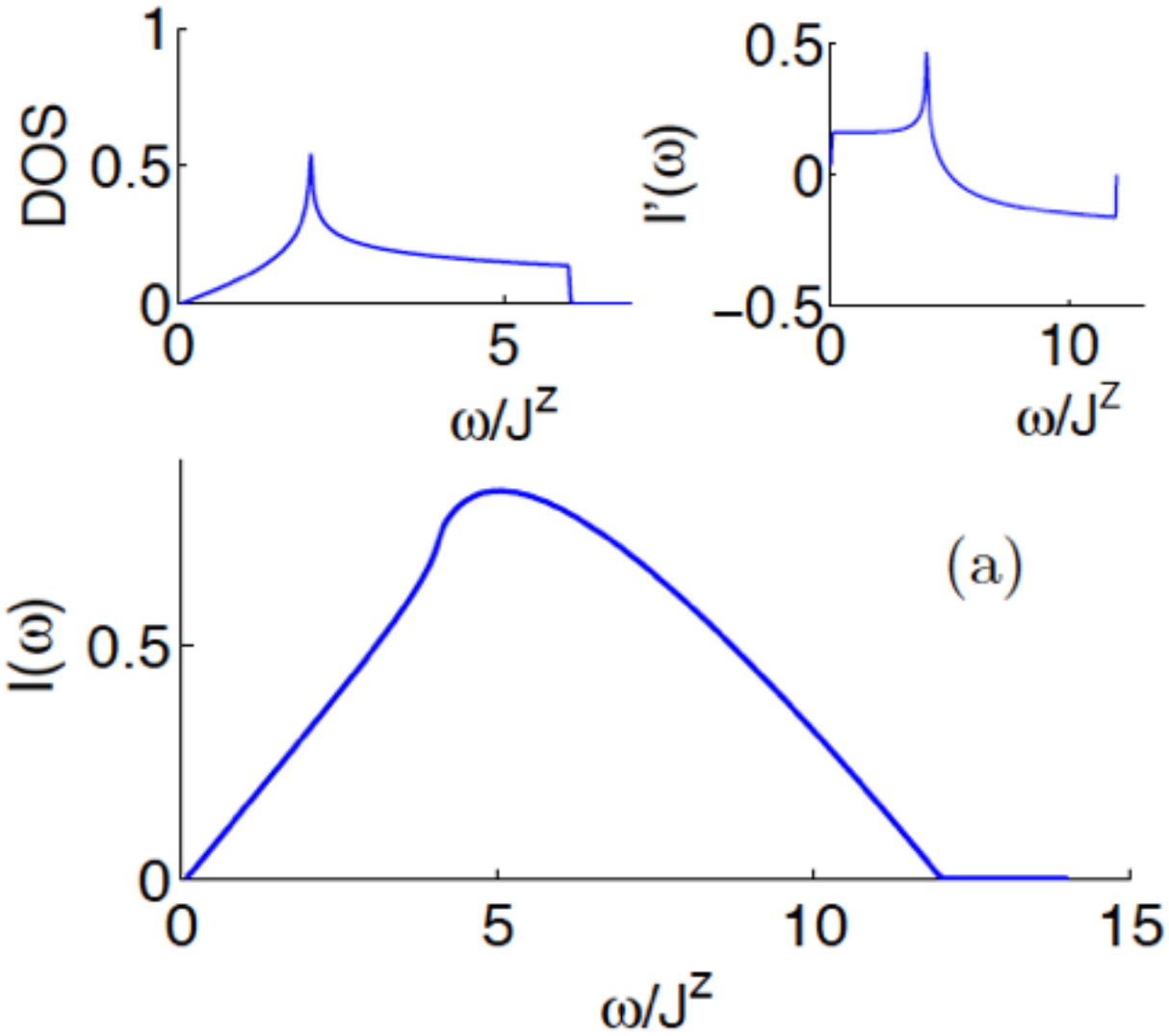
$$R = \sum_{\langle ij \rangle^\alpha} (\boldsymbol{\epsilon}_{\mathrm{in}} \cdot \mathbf{d}^\alpha)(\boldsymbol{\epsilon}_{\mathrm{out}} \cdot \mathbf{d}^\alpha) J^\alpha \sigma_i^\alpha \sigma_j^\alpha$$

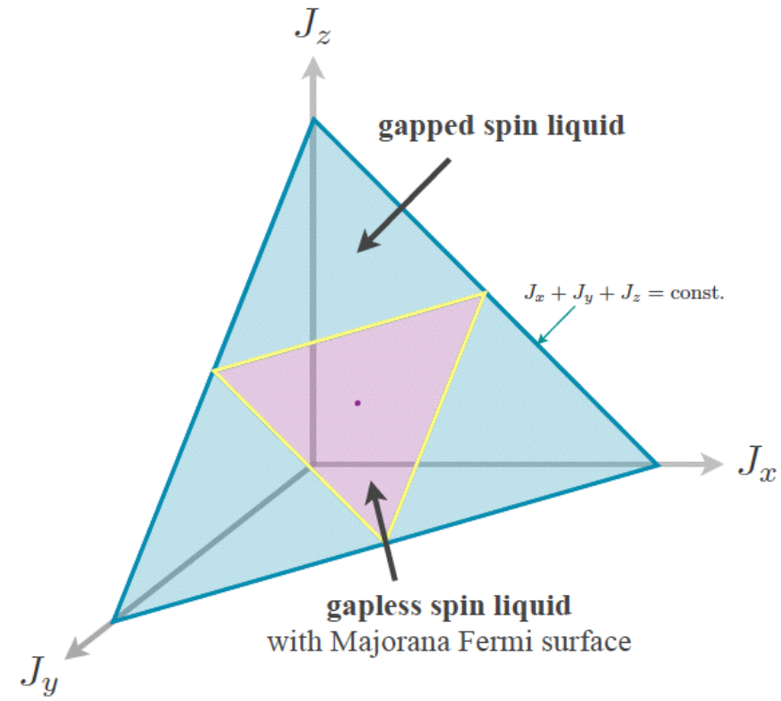
$$= i \sum_{\langle ij \rangle^\alpha} (\boldsymbol{\epsilon}_{\mathrm{in}} \cdot \mathbf{d}^\alpha)(\boldsymbol{\epsilon}_{\mathrm{out}} \cdot \mathbf{d}^\alpha) J^\alpha u_{\langle ij \rangle^\alpha} c_i c_j$$

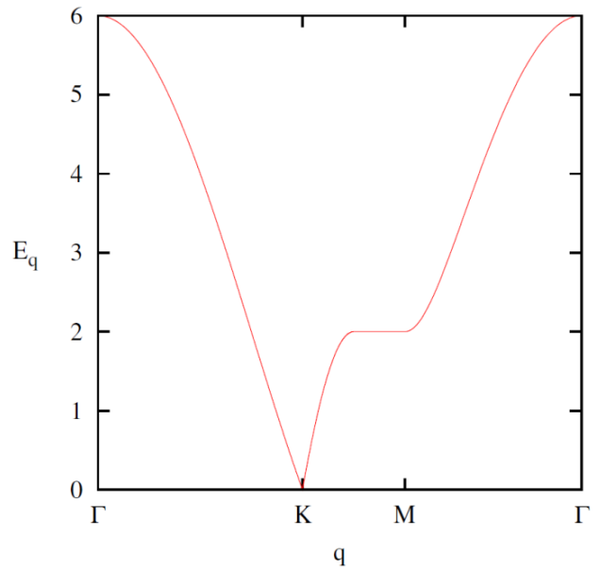
Raman vertex: diagonal in fluxes but creates two Majorana fermions

Raman Scattering results (2D)

isotropic point:
polarization independent



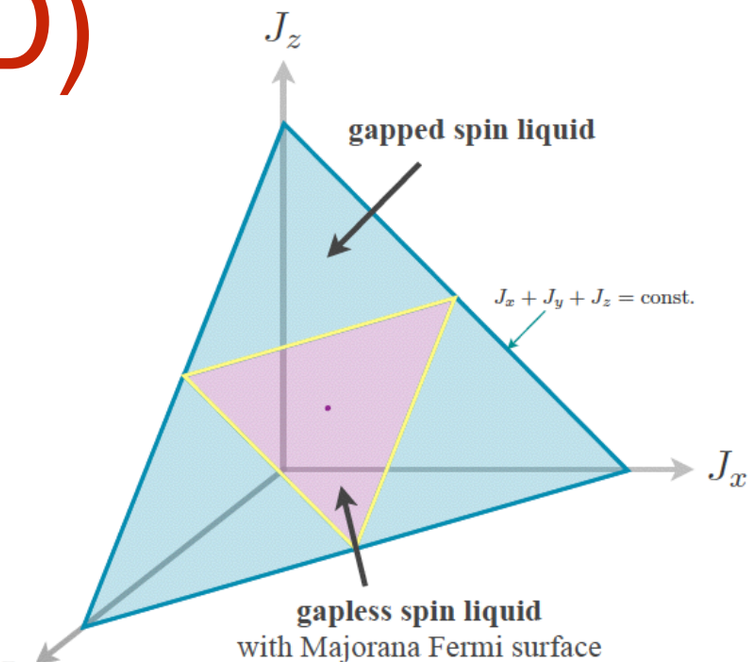


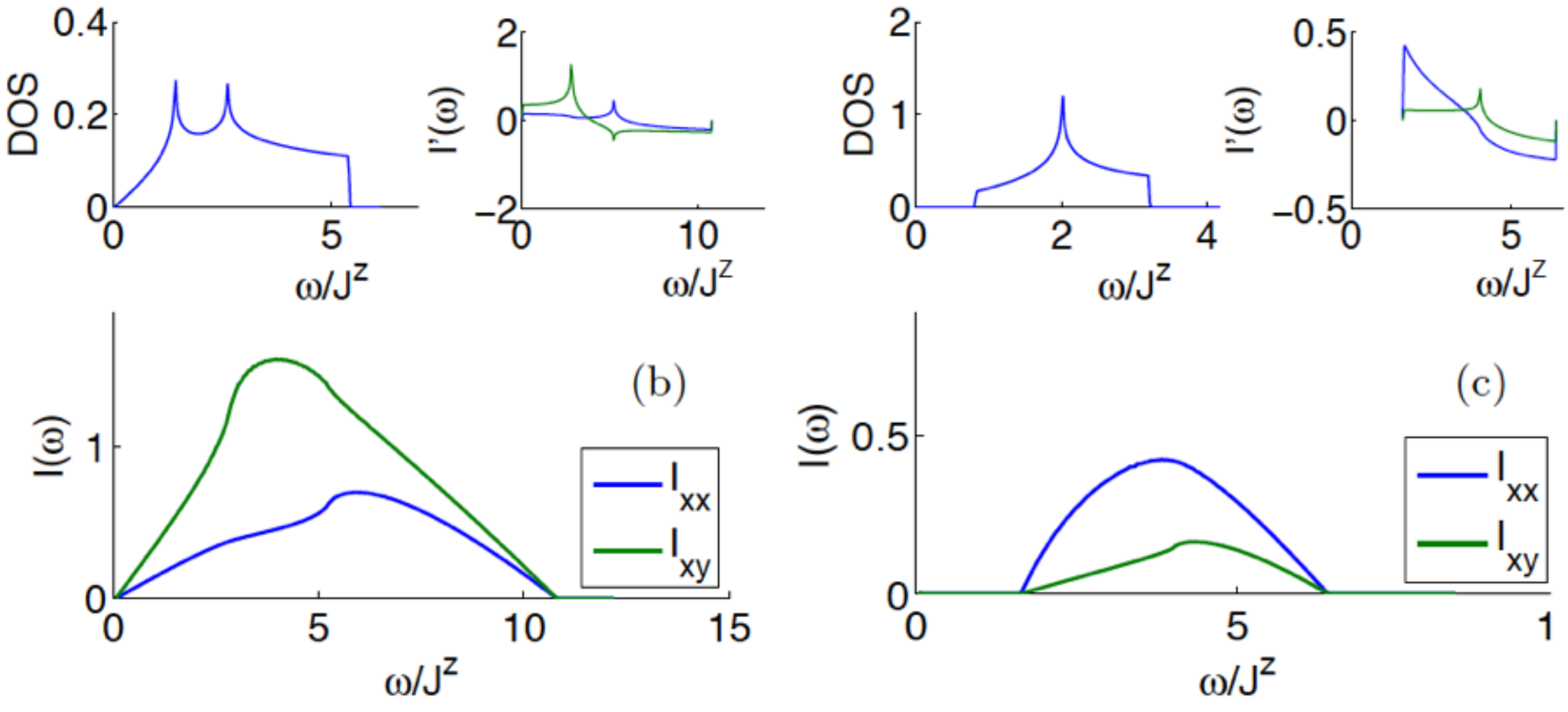


Knolle, Chern, Kovrizhin, Moessner, **NP** (PRL 2014)

Raman Scattering results (2D)

anisotropic point:
polarization dependence

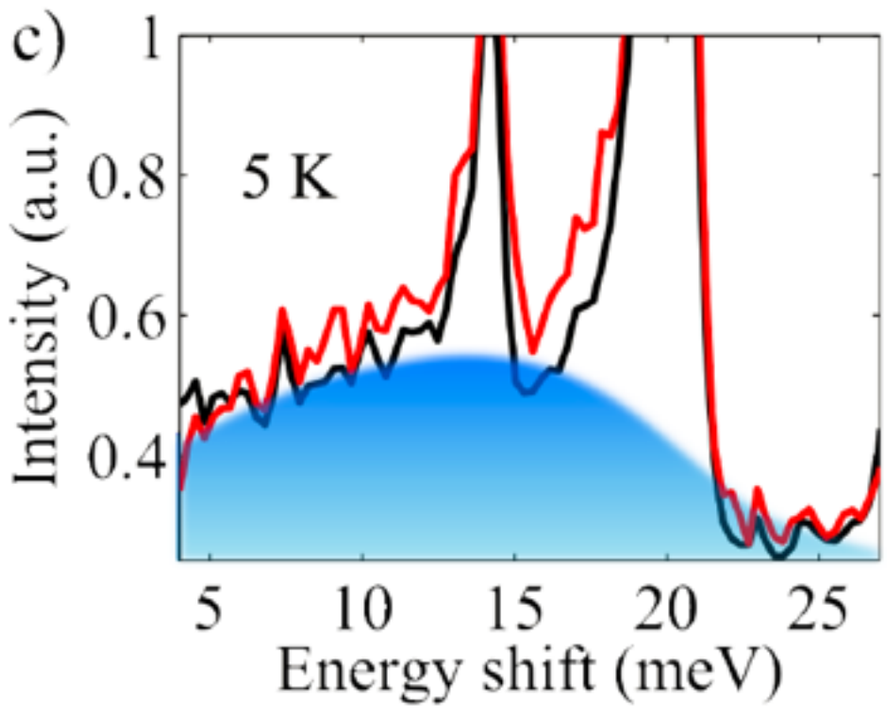




Knolle, Chern, Kovrizhin, Moessner, **NP** (PRL 2014)

$RuCl_3$

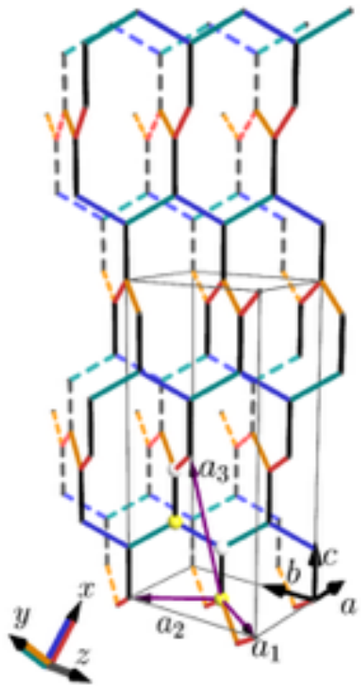
L. Sandilands,Y.J. Kim, K.S. Burch
Phys. Rev. Lett. 114 (2015)

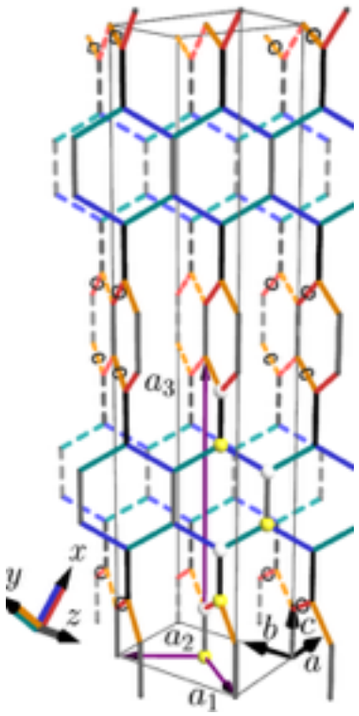


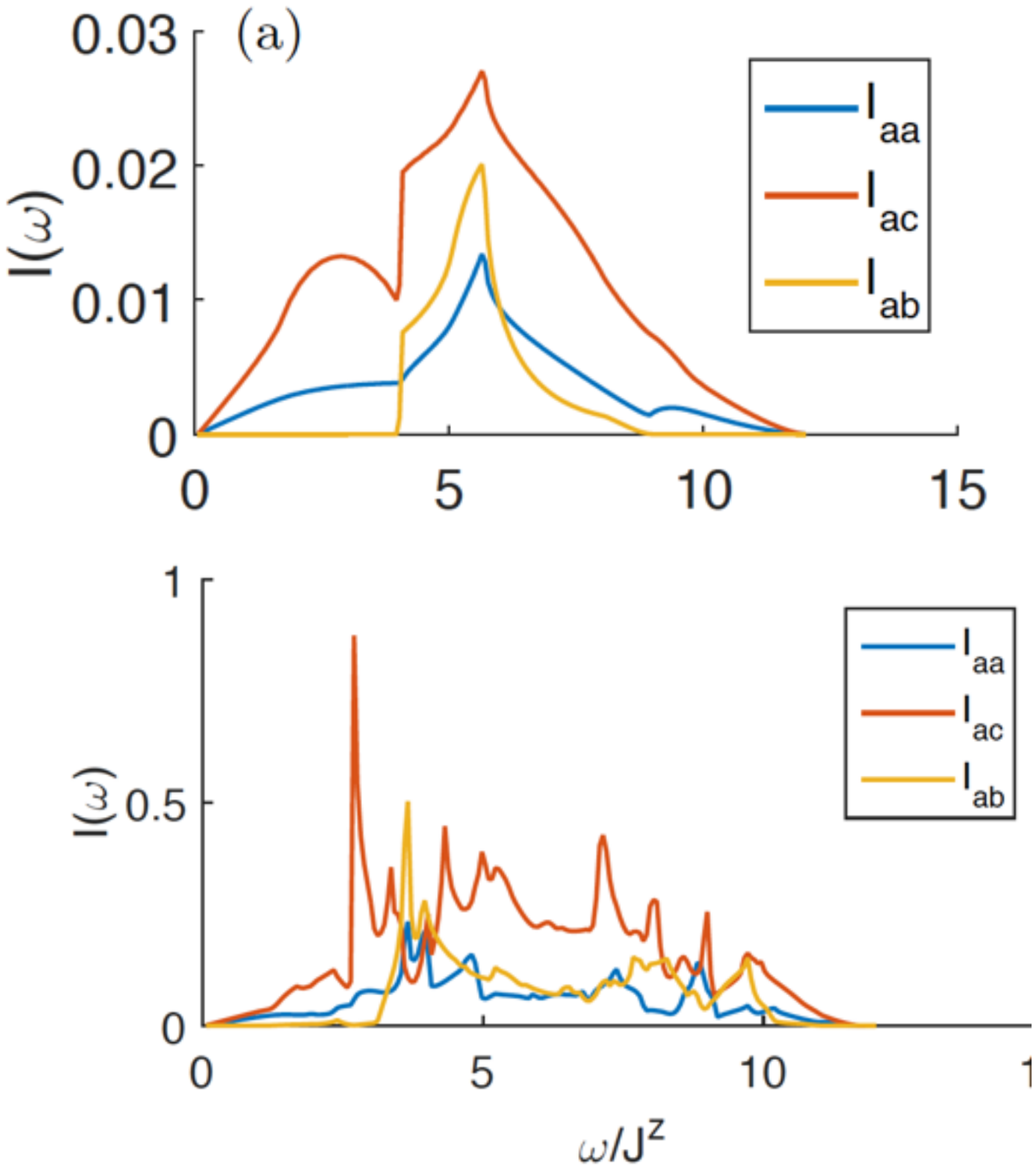
Big ‘hump’ with fine features of the Majorana DOS.
Signatures of fractionalization are visible!
(comparison gives J_K~8meV)

Raman Scattering results (3D)

The Raman response is polarization dependent!

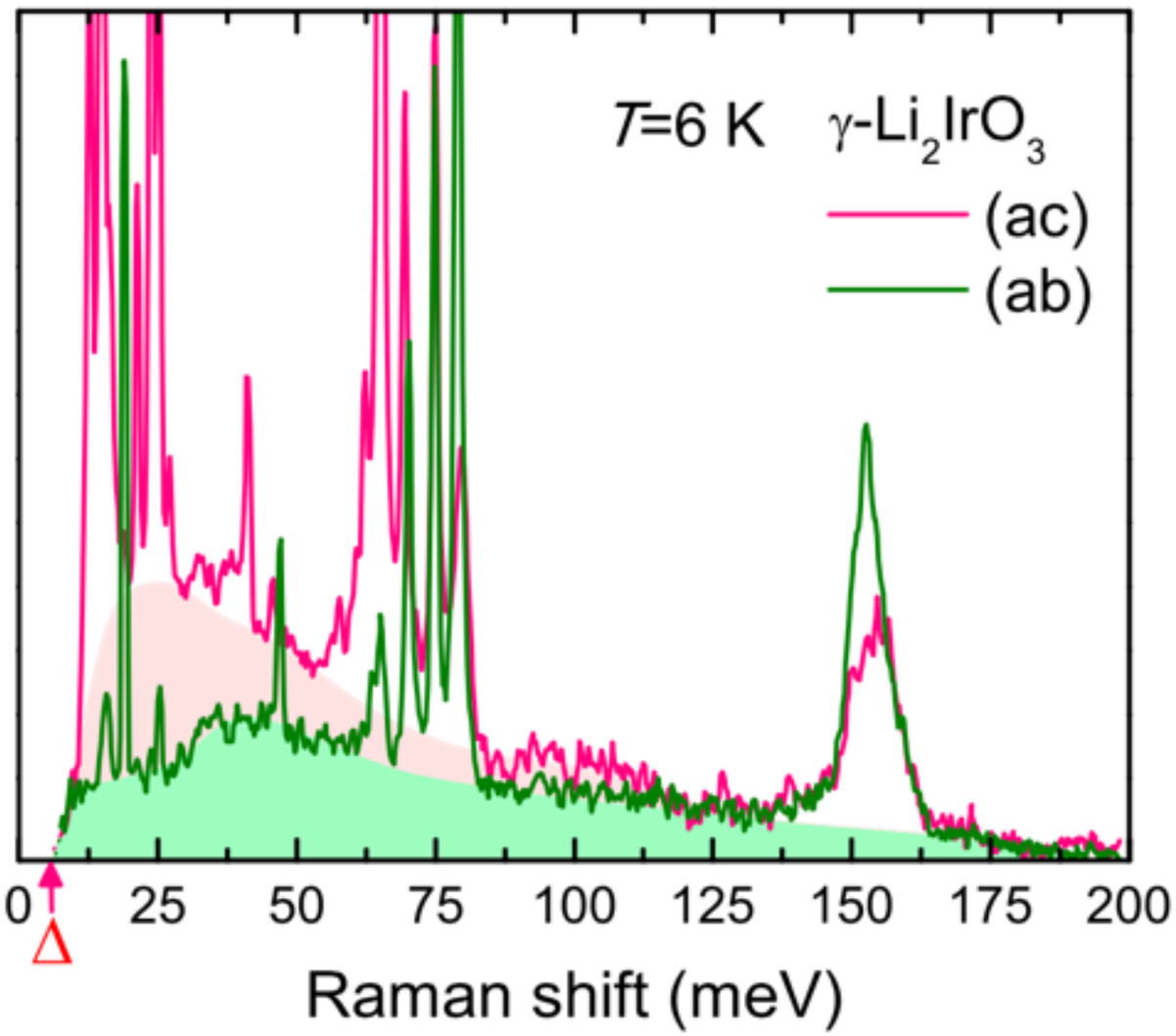


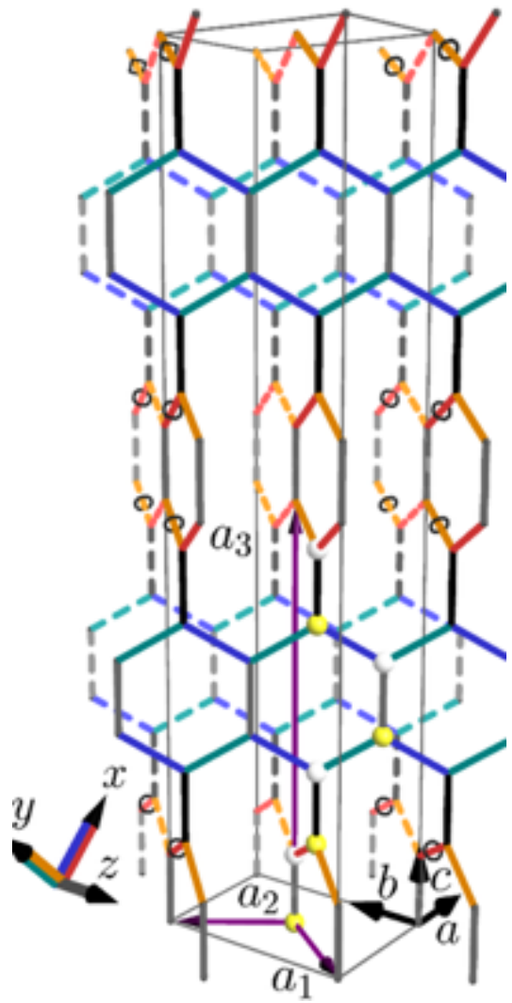




Brent Perreault, Johannes Knolle, NBP, F. J. Burnell, PRB 2015

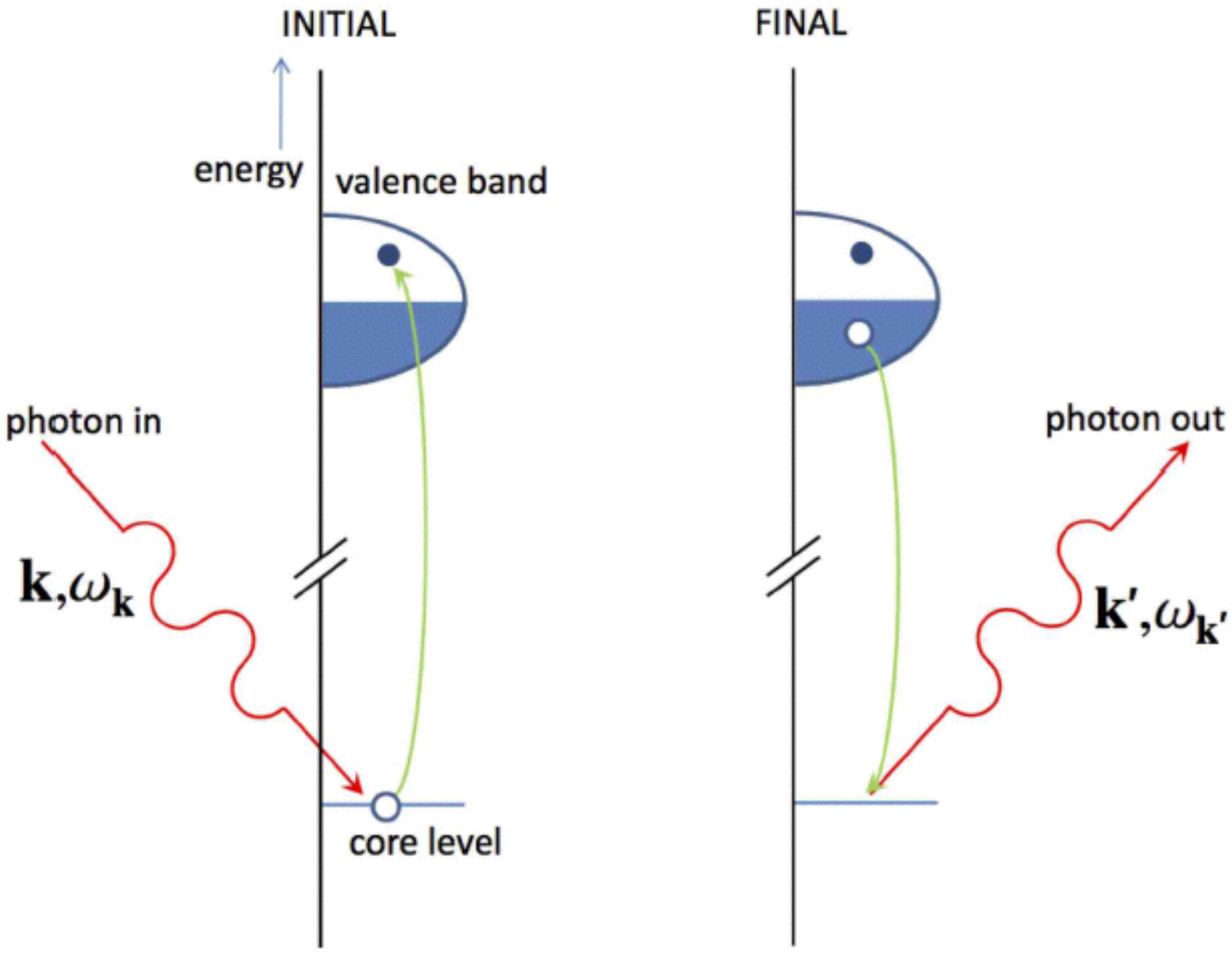
γ-Li_2IrO_3





Glamazda, Lemmens, Do, Choi, Choi, Nature Comm. 7 (2016)

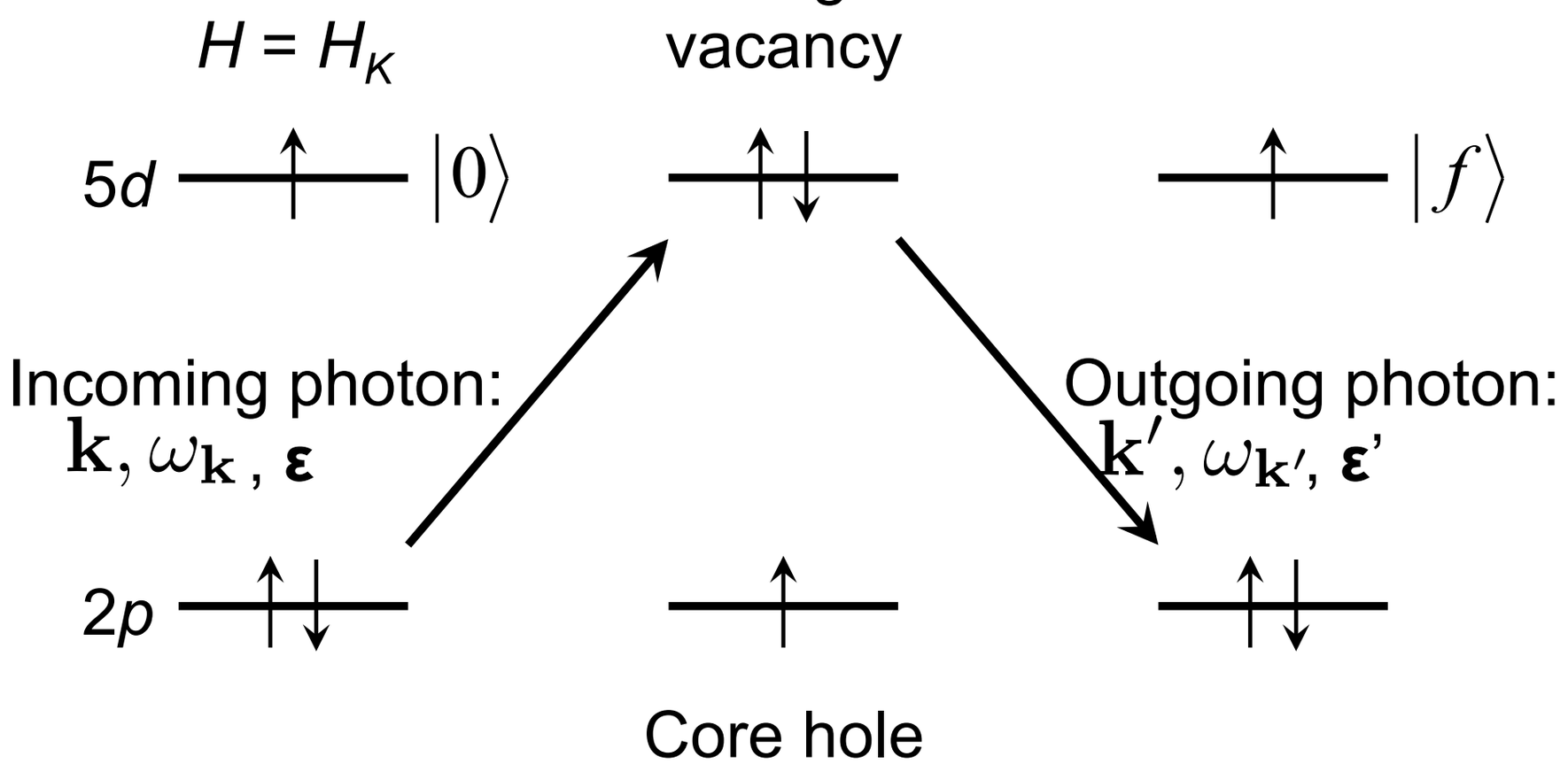
RIXS spectroscopy of Kitaev Spin Liquids



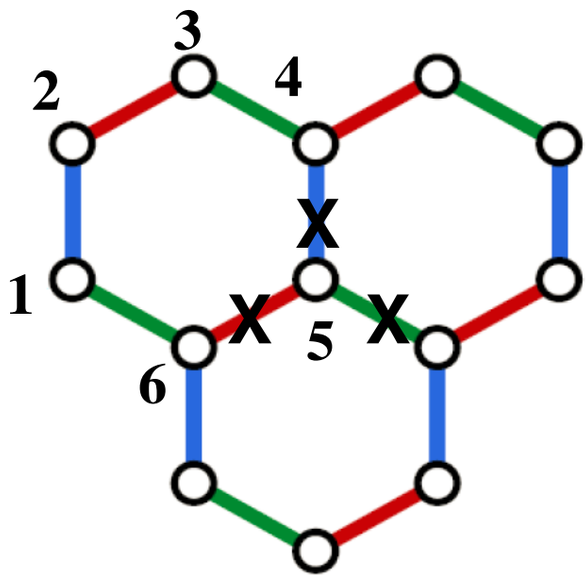
L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, J. van den Brink, RMP (2011)

RIXS from Ir^{4+}

$(Na,Li)_2IrO_3$ with Ir^{4+} in $5d^5$ configuration [L_3 edge]:



Intermediate state with a vacancy



The Kitaev model with a single vacancy at site **r**
= the original Kitaev model with switched off couplings around site **r** (exactly solvable)

Description: the vacancy is always in the spin-up state and the couplings to NN are zero.

$$d^\dagger_{\mathbf{r},\downarrow} \to \frac{1}{2}(1+\sigma^z_{\mathbf{r}}), \qquad d^\dagger_{\mathbf{r},\uparrow} \to \frac{1}{2}\sigma^x_{\mathbf{r}}(1-\sigma^z_{\mathbf{r}})$$

$$d^\dagger_{\mathbf{r},\downarrow}|\uparrow\rangle = |\uparrow\rangle \qquad d^\dagger_{\mathbf{r},\uparrow}|\uparrow\rangle = 0$$

$$d^\dagger_{\mathbf{r},\downarrow}|\downarrow\rangle = 0 \qquad d^\dagger_{\mathbf{r},\uparrow}|\downarrow\rangle = |\uparrow\rangle$$

Halász, Chalker, Moessner, PRB 2014

RIXS amplitude for the Kitaev model

$$I(\omega,\mathbf{q}) = \sum_m \left| \sum_{\alpha,\beta} T_{\alpha\beta} A_{\alpha\beta}(m,\mathbf{q}) \right|^2 \delta(\omega - E_m)$$

$$A_{\alpha\beta}(m,\mathbf{q}) = \sum_{\mathbf{r},\tilde{n}_\mathbf{r}} \frac{\langle m | d_{\mathbf{r},\alpha} | \tilde{n}_\mathbf{r} \rangle \langle \tilde{n}_\mathbf{r} | d^\dagger_{\mathbf{r},\beta} | 0 \rangle}{\Omega - E_{\tilde{n}} + i\Gamma} \, e^{i\mathbf{q}\cdot\mathbf{r}}$$

$$\mathbf{q} \equiv \mathbf{k} - \mathbf{k}'$$

Kramers–Heisenberg formula

The four fundamental RIXS channels are introduced by decomposing the polarization tensor into

$$T_{\alpha\beta} = P_\eta \sigma^\eta_{\alpha\beta}$$

(a) Spin-conserving (SC) channel with $T_{\alpha\beta} \propto \sigma^0_{\alpha\beta}$

$$P_0 = \boldsymbol{\epsilon}'^* \cdot \boldsymbol{\epsilon}$$

(b) three non spin-conserving (NSC) channels with $T_{\alpha\beta} \propto \sigma^{x,y,z}_{\alpha\beta}$

$$P_x = i(\epsilon'^*_y \epsilon_z - \epsilon'^*_z \epsilon_y)$$

and cyclic permutations

The four fundamental RIXS channels

$$A(m,\mathbf{q}) = \sum_{\nu} P_\nu A_\nu(m,\mathbf{q})$$

$$A_\nu(m,\mathbf{q}) = \sum_{\mathbf{r}} e^{i\mathbf{q}\mathbf{r}}\, e^{-\Gamma t}\, \langle m|\sigma^\nu_{\mathbf{r}}\, e^{-it\tilde{H}_{\mathbf{r}}}|0\rangle$$

Spin-conserving (SC) channel:

$$A_0(m,\mathbf{q}) = \sum_{\mathbf{r}} e^{i\mathbf{q}\mathbf{r}}\, e^{-\Gamma t}\, \langle m|e^{-itH_{\mathbf{r}}}|0\rangle$$

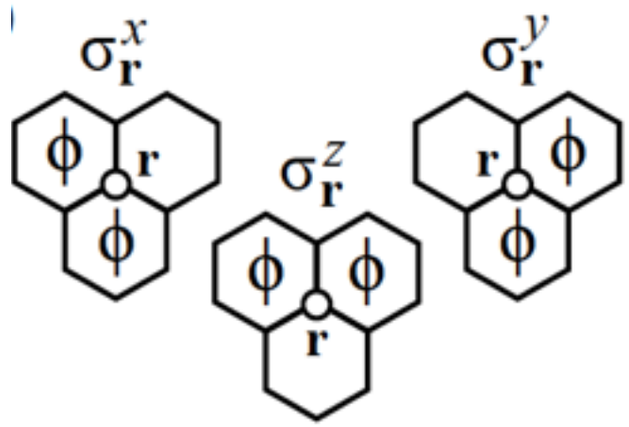
Three non spin-conserving (NSC) channels:

$$A_x(m,\mathbf{q}) = \sum_{\mathbf{r}} e^{i\mathbf{q}\mathbf{r}}\, e^{-\Gamma t}\, \langle m|\sigma^x_{\mathbf{r}} e^{-it\tilde{H}_{\mathbf{r}}}|0\rangle$$

$$A_y(m,\mathbf{q}) = \sum_{\mathbf{r}} e^{i\mathbf{q}\mathbf{r}}\, e^{-\Gamma t}\, \langle m|\sigma^y_{\mathbf{r}} e^{-it\tilde{H}_{\mathbf{r}}}|0\rangle$$

$$A_z(m,\mathbf{q}) = \sum_{\mathbf{r}} e^{i\mathbf{q}\mathbf{r}}\, e^{-\Gamma t}\, \langle m|\sigma^z_{\mathbf{r}} e^{-it\tilde{H}_{\mathbf{r}}}|0\rangle$$

create two flux excitations



Fast collision approximation

$(Na,Li)_2IrO_3$ and α-$RuCl_3$: $\Gamma / J_{x,y,z} >> 1$

$t \sim 1 / \Gamma \ << 1 / J_{x,y,z} \ \rightarrow$

The lowest order RIXS amplitude is then

$$
\begin{aligned}
A_\eta(m,\mathbf{q}) \ &\propto \ \sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}} \langle m|\sigma^\eta_{\mathbf{r}} \left[1 - \frac{iH(\mathbf{r})}{\Gamma}\right]|0\rangle \\
&= \ \sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}} \langle m|\sigma^\eta_{\mathbf{r}} \left[1 - \frac{i}{\Gamma}\sum_{\kappa} J_\kappa \sigma^\kappa_{\mathbf{r}} \sigma^\kappa_{\kappa(\mathbf{r})}\right]|0\rangle
\end{aligned}
$$

NSC channels recover INS amplitudes for infinite Γ

Flux creation: Finite gap, little dispersion

Results: SC channel
2D Kitaev model

$$A_0(m,\mathbf{q}) \propto \sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}} \langle m| \left[1 - \frac{i}{\Gamma} \sum_{\kappa=x,y,z} J_\kappa \sigma^\kappa_{\mathbf{r}} \sigma^\kappa_{\kappa(\mathbf{r})}\right] |0\rangle$$

X

Elastic response

Inelastic response

$$|m\rangle \neq |0\rangle$$

no flux and two fermion excitations

$$\omega = \varepsilon_{\mathbf{k}} + \varepsilon_{\mathbf{q}-\mathbf{k}}$$

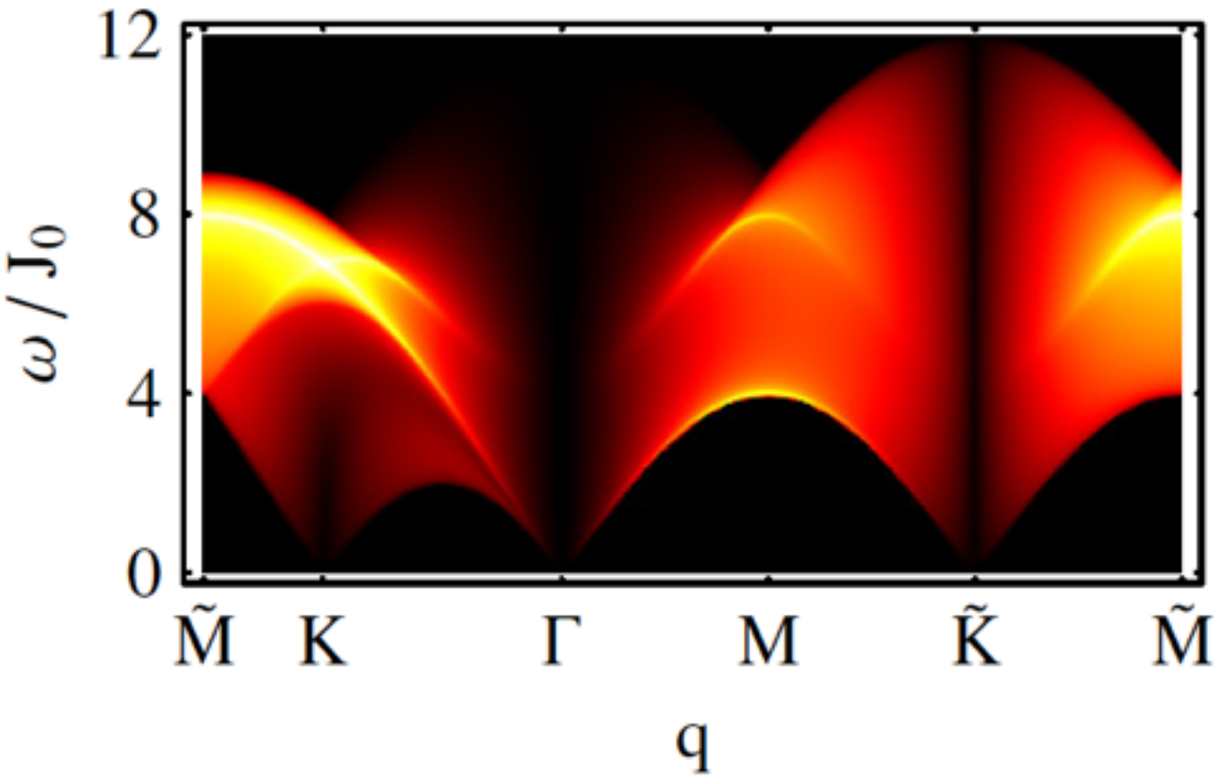
$$I_0(\omega,\mathbf{q}) = \sum_m |A_0(m,\mathbf{q})|^2 \delta(\omega - E_m)$$

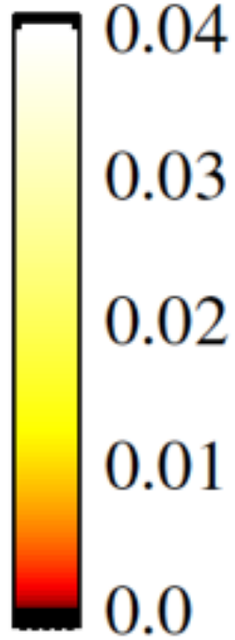
$$I_0(\omega,\mathbf{q}) \propto \int_{\mathrm{BZ}} d^2\mathbf{k}\; \delta(\omega - \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{q}-\mathbf{k}}) \left[\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{q}-\mathbf{k}}\right]^2 \left|1 - e^{i\varphi_{\mathbf{k}}}\, e^{i\varphi_{\mathbf{q}-\mathbf{k}}}\right|^2$$

A B

interference between the two sublattices

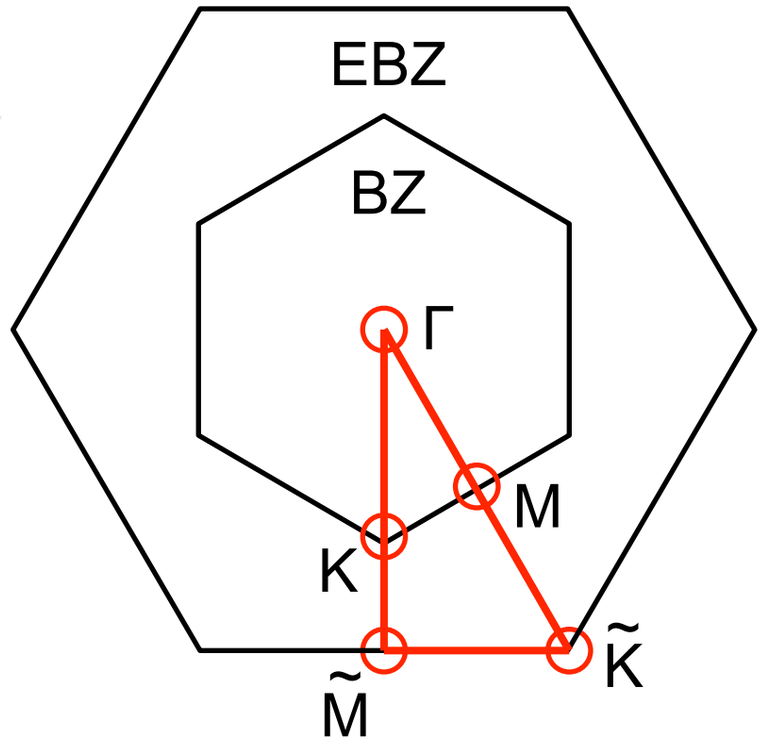
RIXS response in SC channel





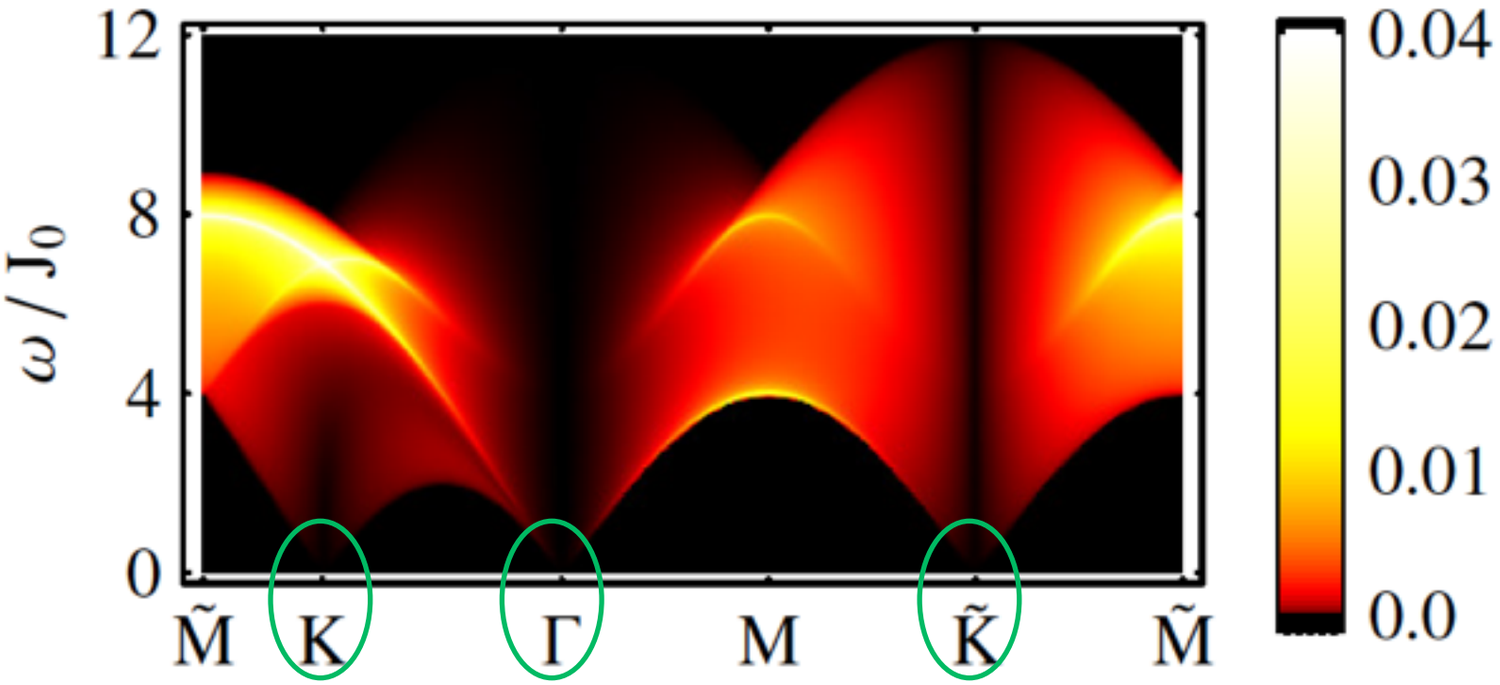
$$J_{x,y,z} = J_0$$

Reciprocal space



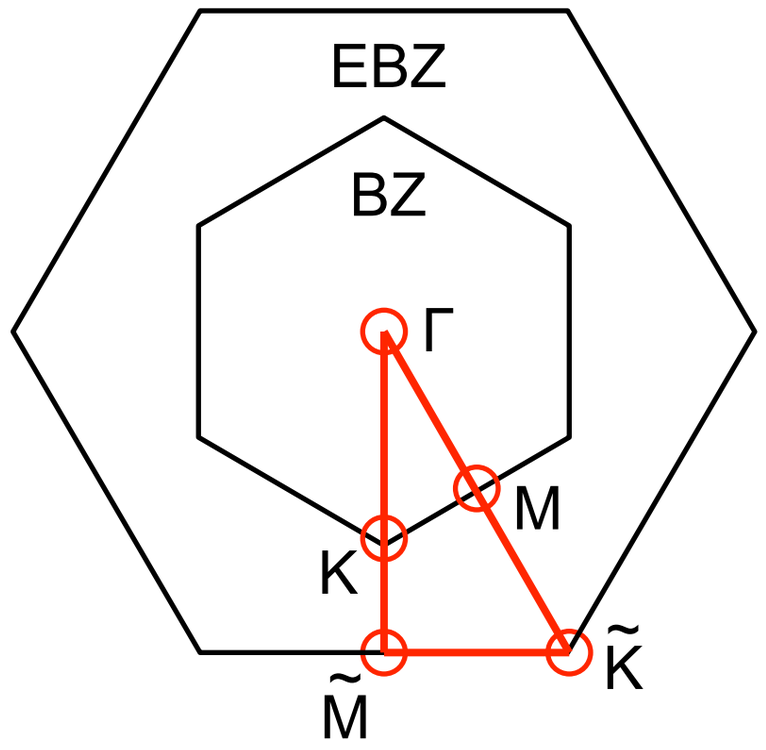
G. B. Halasz, NBP, J.van den Brink (PRL 2016)

RIXS response in SC channel



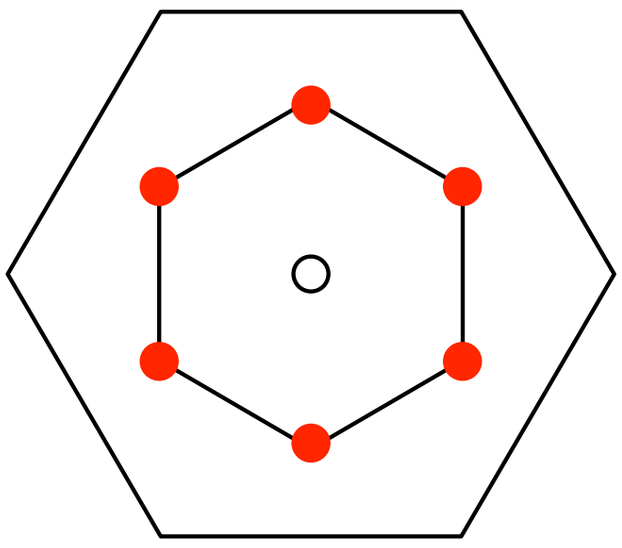
$$J_{x,y,z} = J_0$$

Reciprocal space



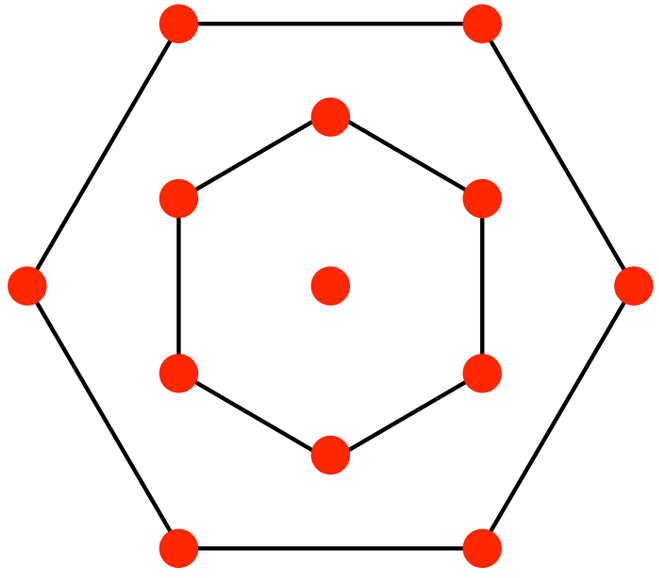
Gapless response at a finite number of discrete points

Dirac points



K points

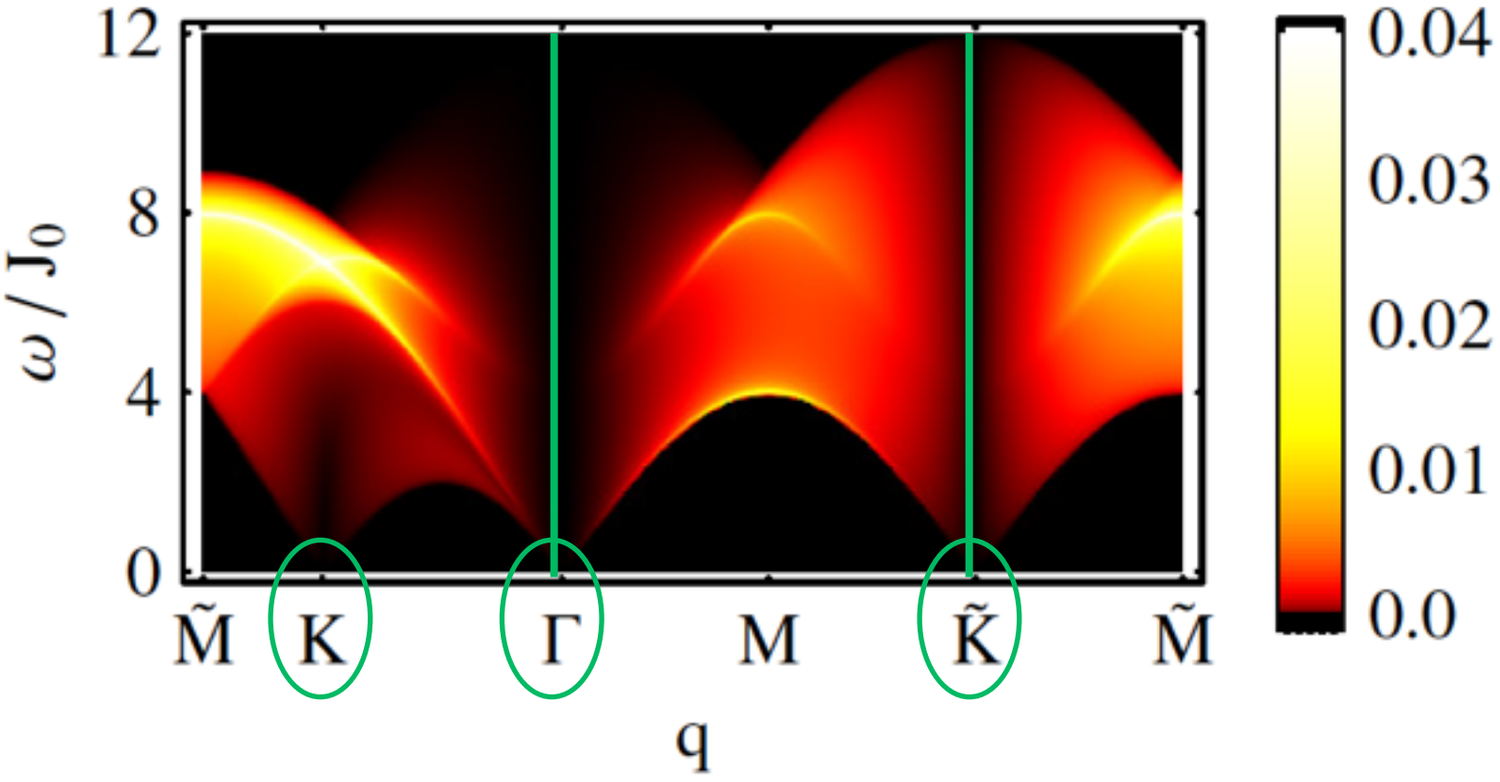
Gapless points



Γ, K, K̃ points

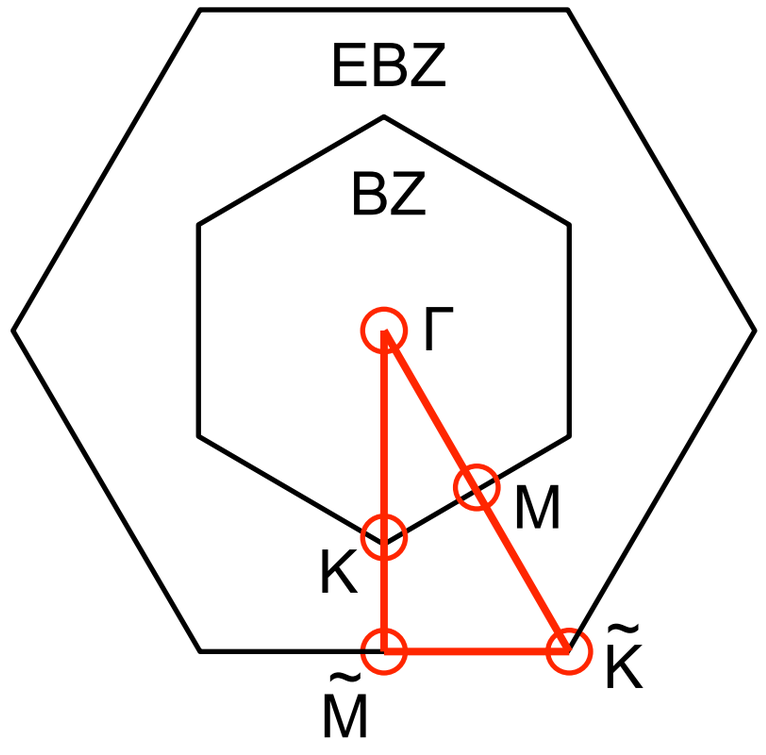
G. B. Halasz, NBP, J.van den Brink (PRL 2016)

RIXS response in SC channel



$$J_{x,y,z} = J_0$$

Reciprocal space

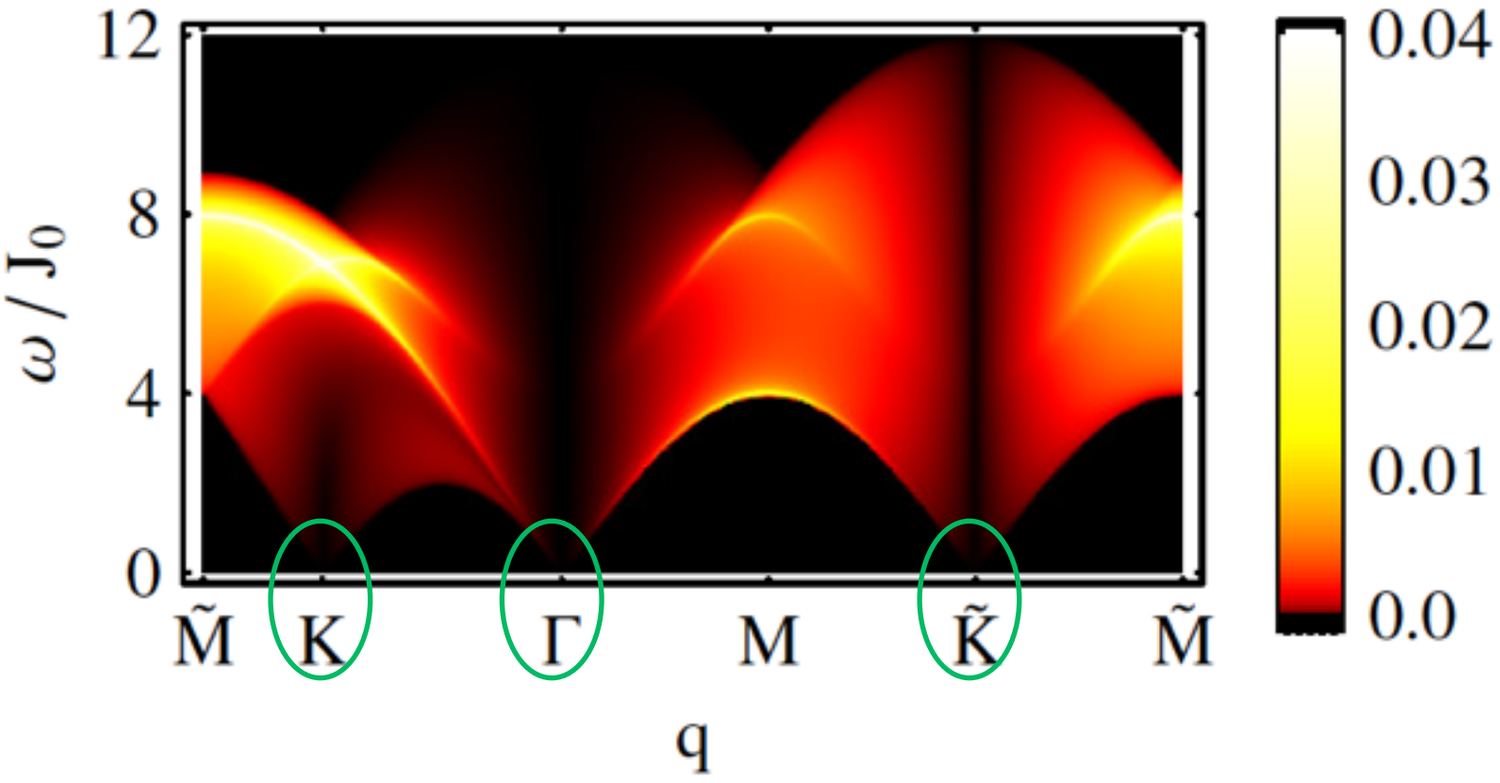


the response actually vanishes at the Γ and $\tilde{K}$ points due to the factor

$$\tilde{I}_0(\mathbf{k},\mathbf{q}) \propto [\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{q}-\mathbf{k}}]^2$$

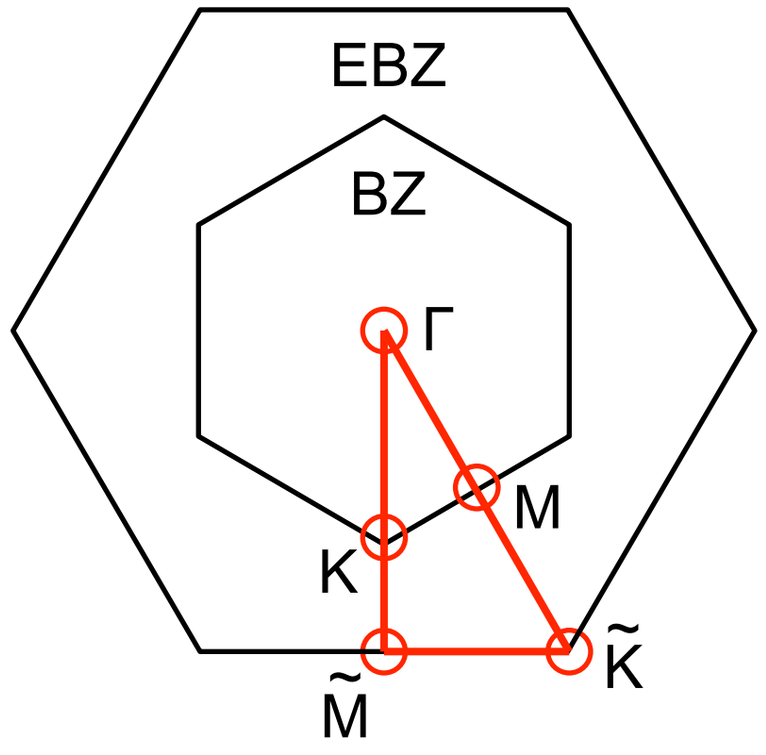
G. B. Halasz, NBP, J.van den Brink (PRL 2016)

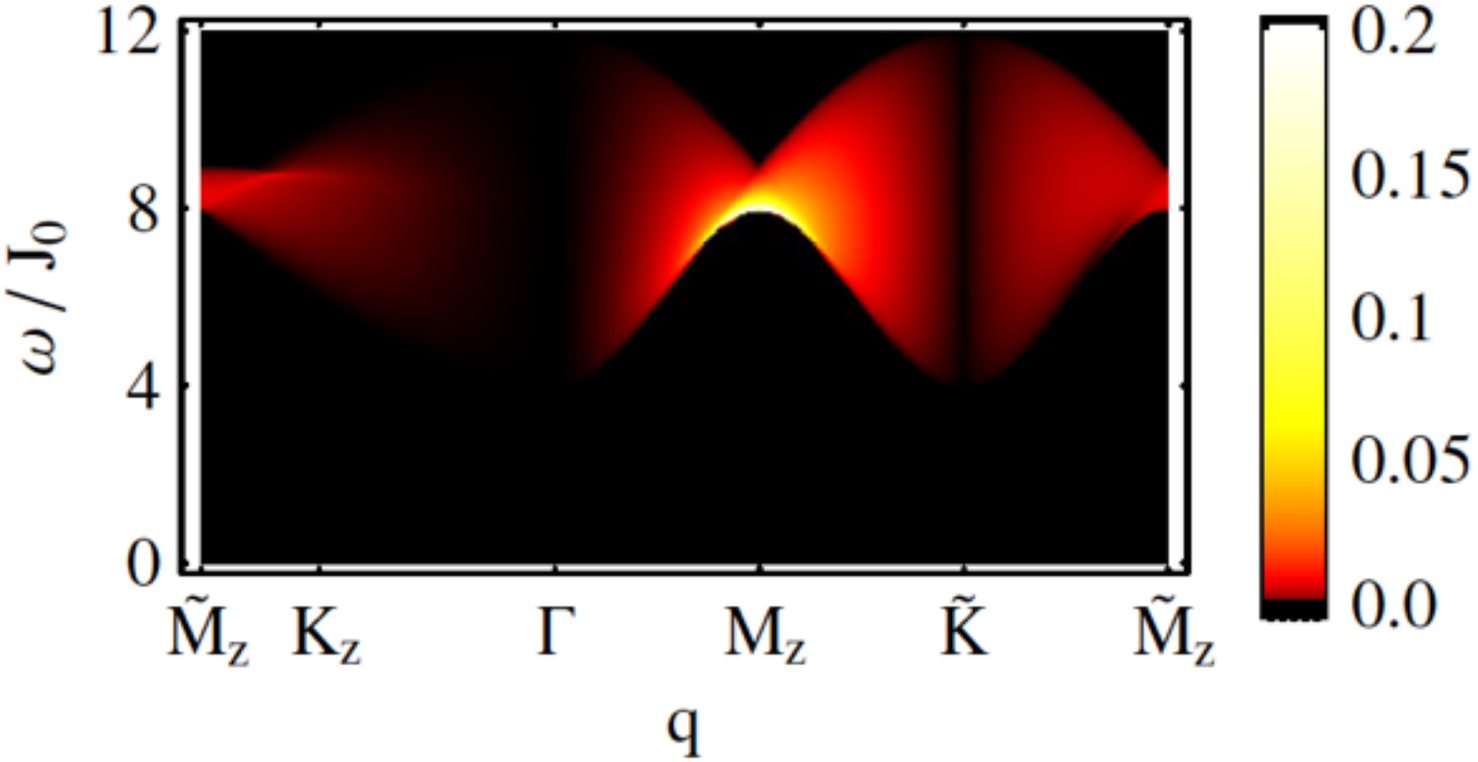
RIXS response in SC channel



$$J_{x,y,z} = J_0$$

Reciprocal space





$$J_{x,y} = J_0/2$$

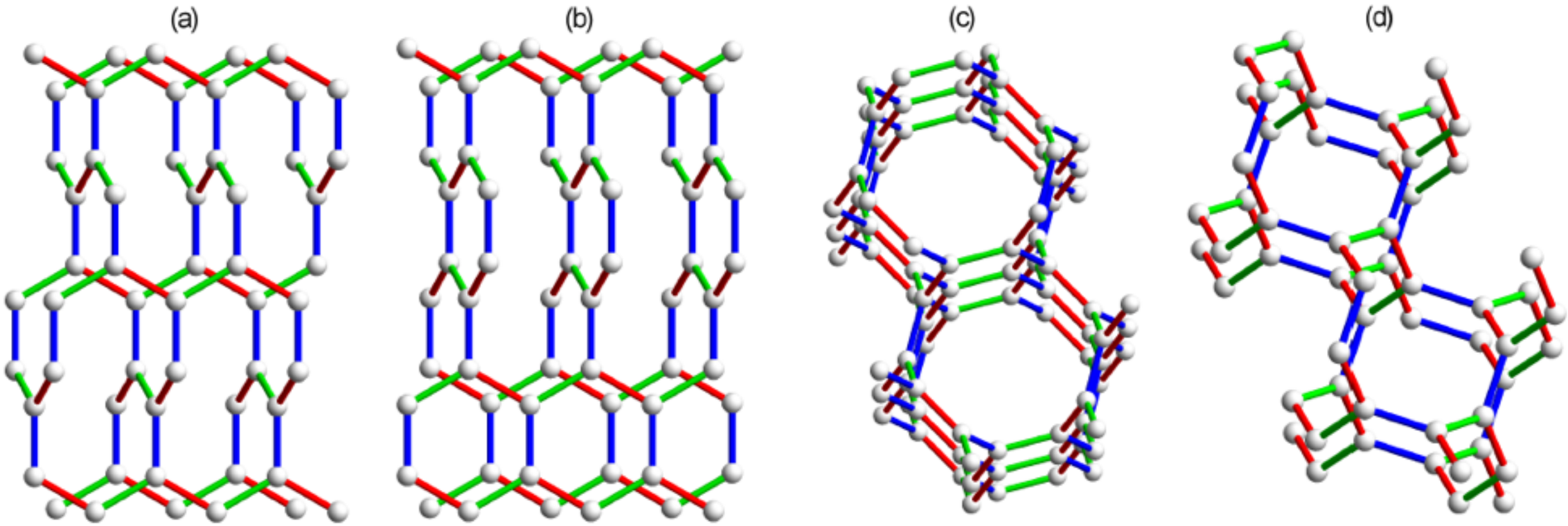
$$J_z = 2J_0$$

G. B. Halasz, NBP, J.van den Brink (PRL 2016)

Results: SC channel in 3D Kitaev models

$$I_0(\omega, \mathbf{q}) \propto \sum_{\mathbf{k},\mu,\mu'} \left|(\mathcal{A}_{\mathbf{q},\mathbf{k}})_{\mu\mu'}\right|^2 \delta(\omega - \varepsilon_{\mathbf{k},\mu} - \varepsilon_{\mathbf{q}-\mathbf{k},\mu'})$$

For each model, the low-energy(gapless) response is determined by the nodal structure of the fermions.

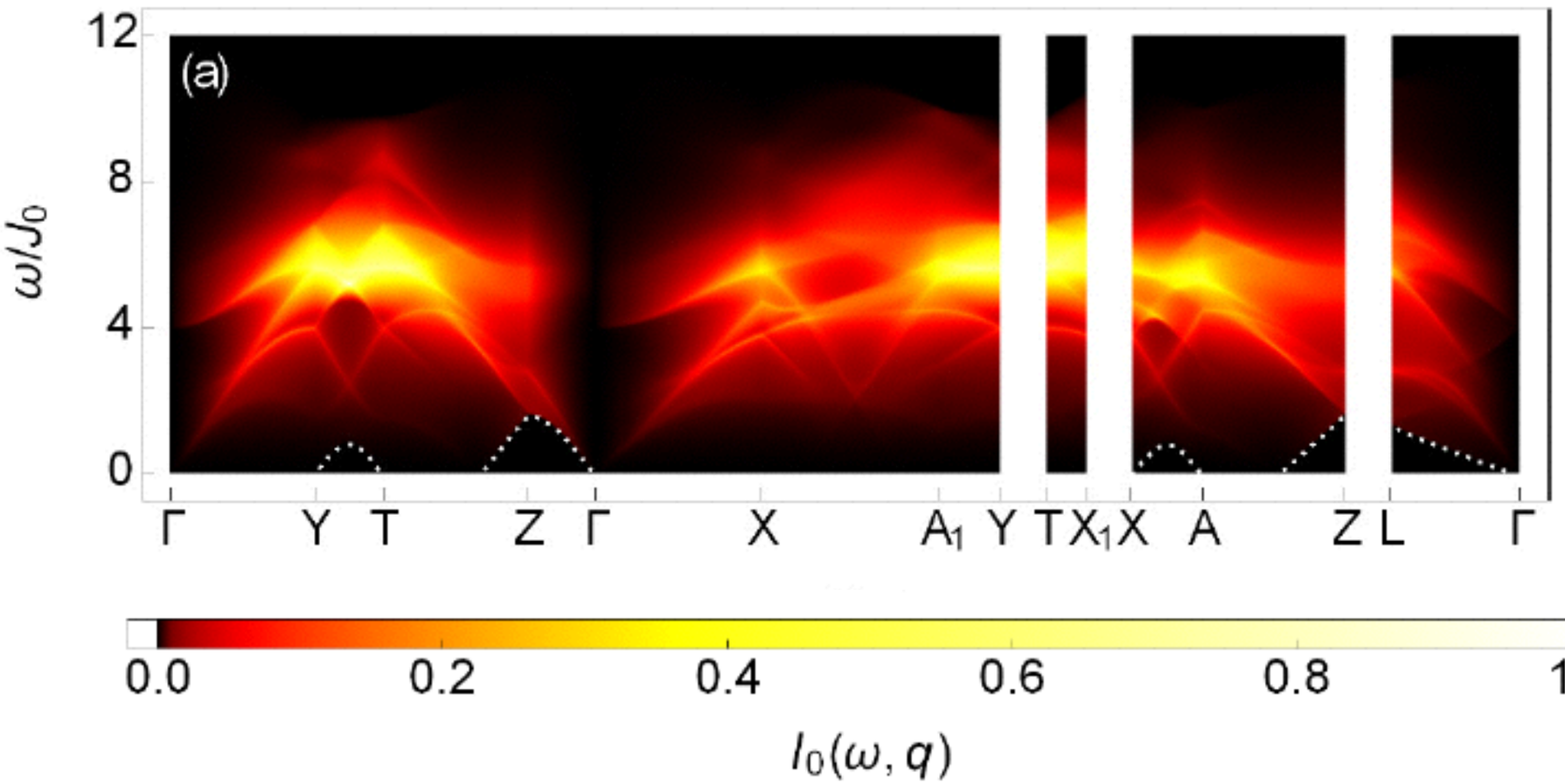


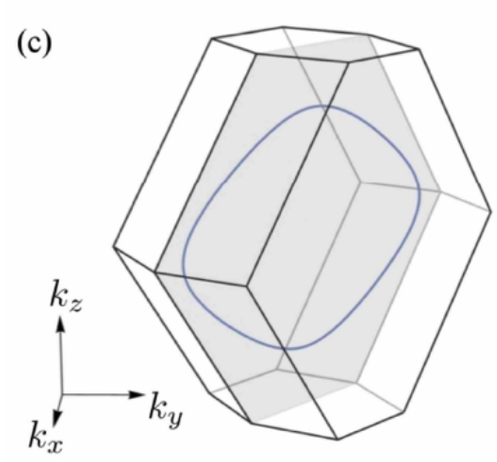
closed line of Dirac nodes

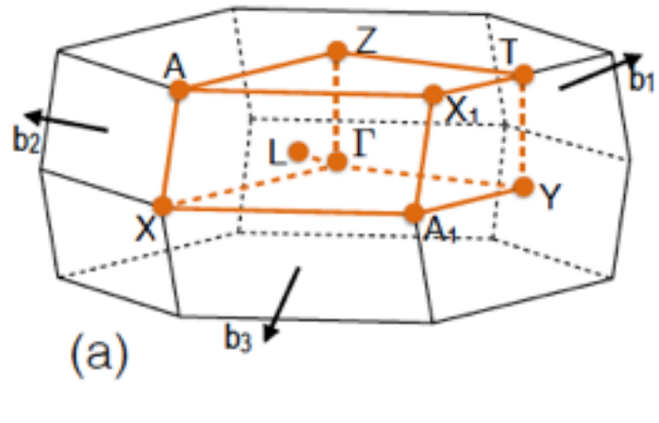
gapless Weyl points

Fermi surfaces

Hyperhoneycomb lattice





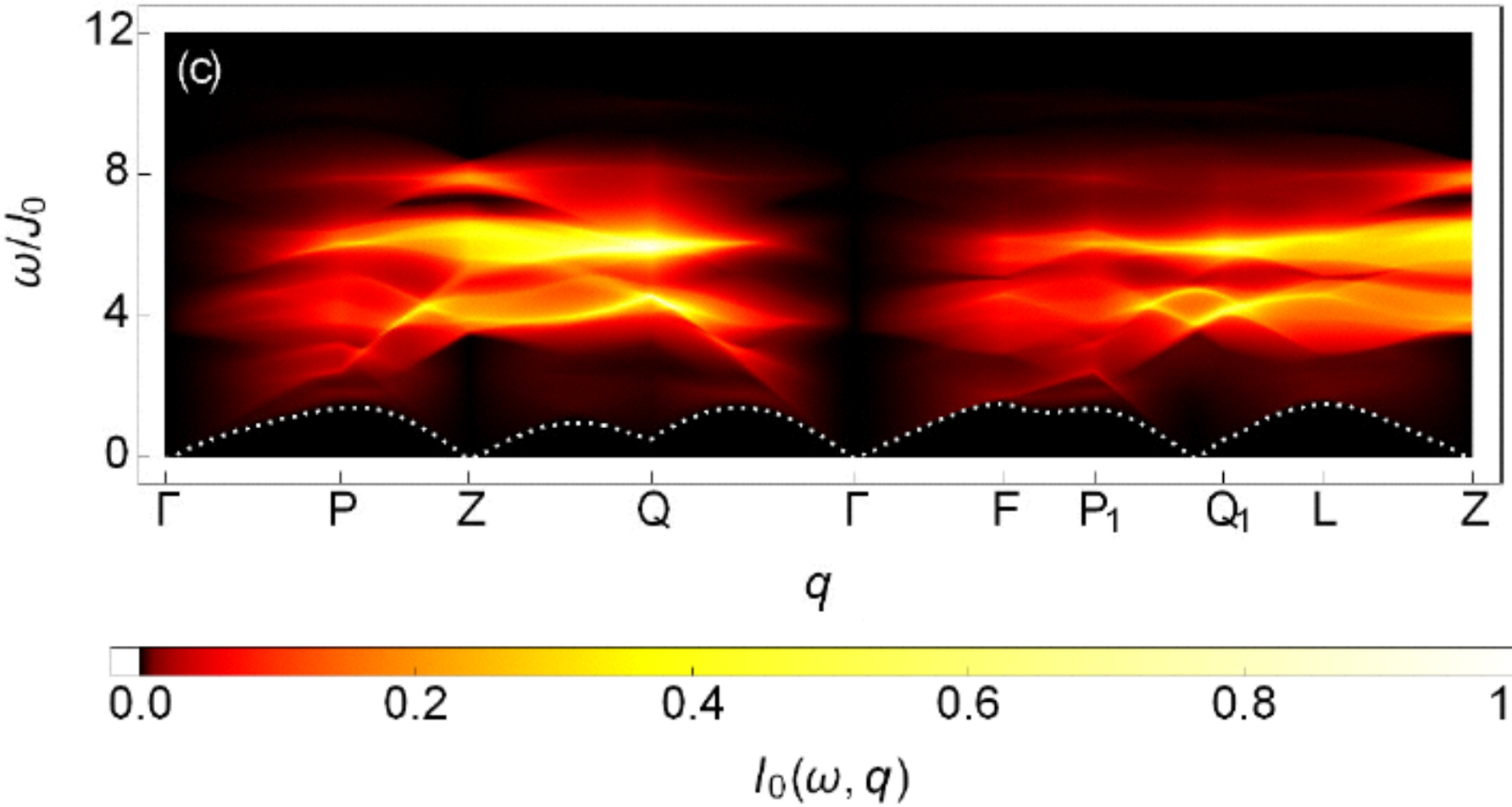


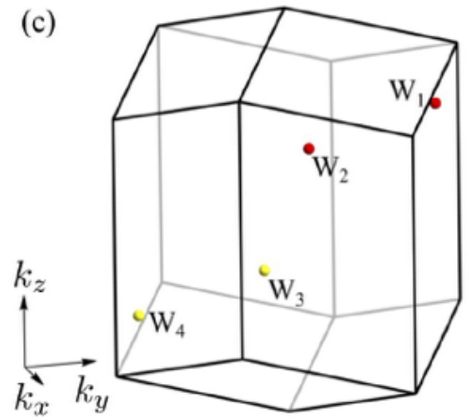
The Majorana fermions are gapless along a nodal line within the Γ-X-Y plane.

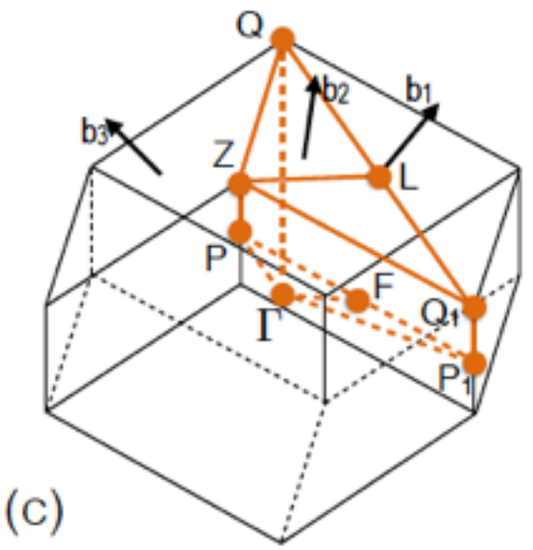
The response is thus gapless in most of the Γ-X-Y plane and also in most of the Z-A-T plane. However, it is still gapped at a generic point of the BZ.

G. B. Halasz, B. Perreault, NBP (1705.05894)

Hyperhexagon lattice





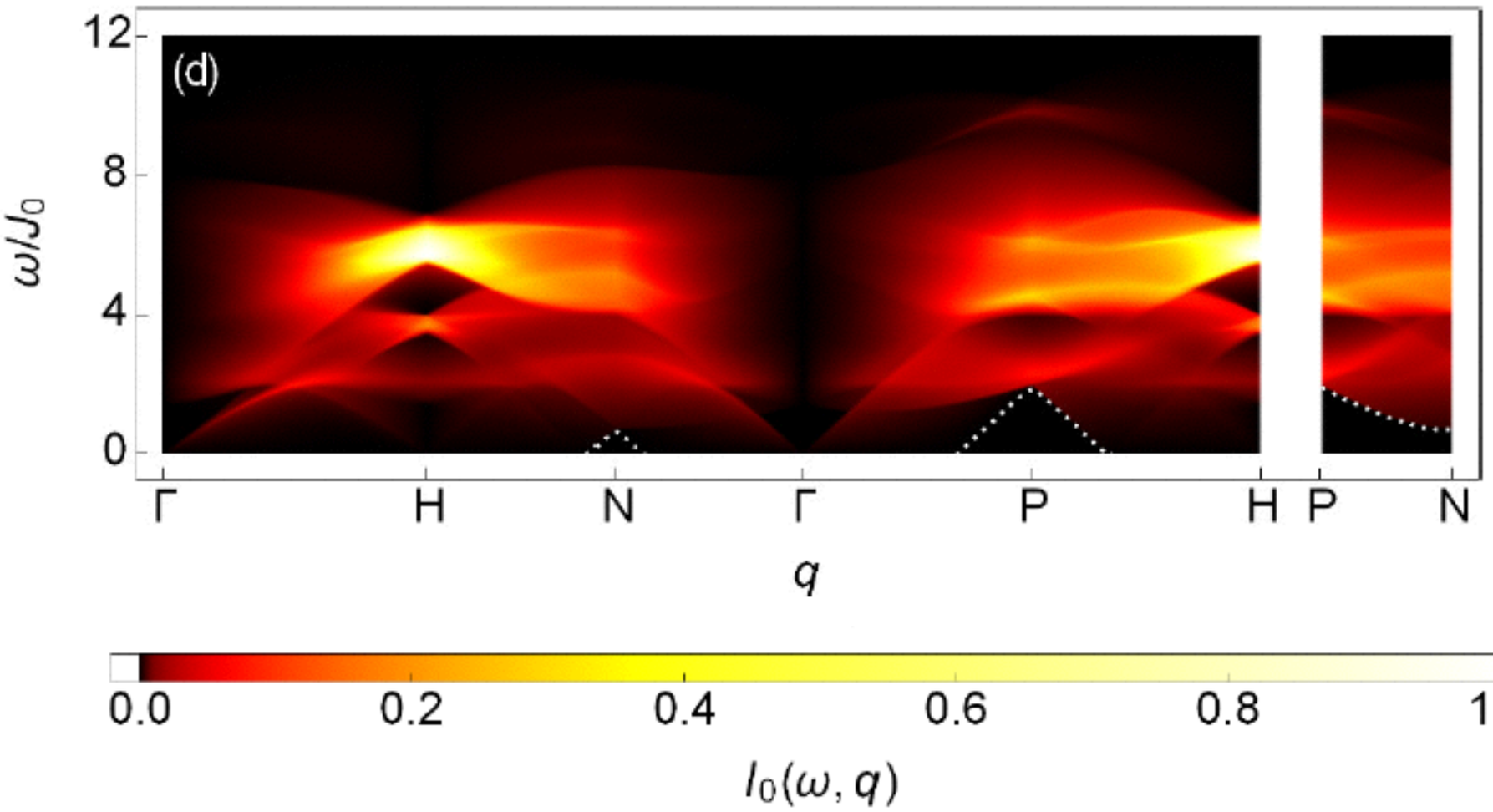


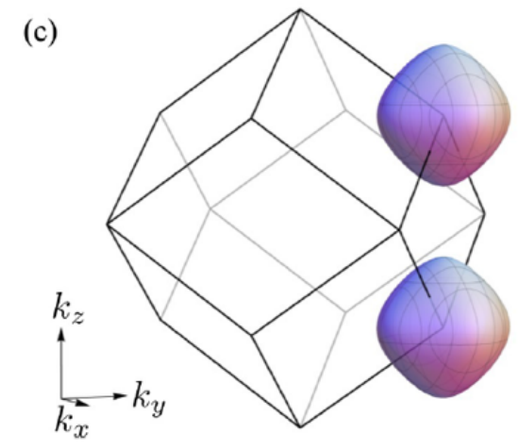
The fermions are gapless at Weyl points.

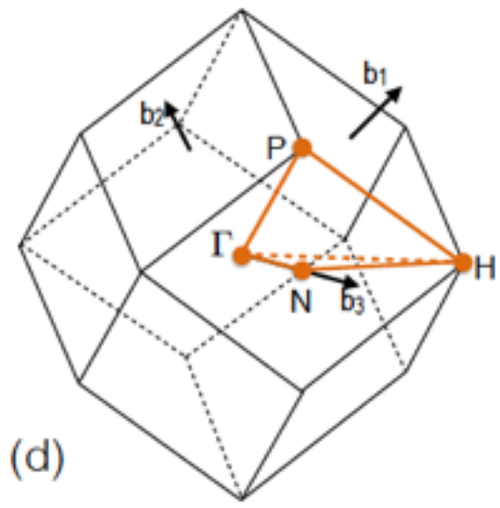
The response is thus only gapless at particular points of the BZ.

G. B. Halasz, B. Perreault, NBP (1705.05894)

Hyperoctagon lattice







The Majorana fermions are gapless on a Fermi surface.

The response is thus gapless in most of the BZ.

G. B. Halasz, B. Perreault, NBP (1705.05894)

Thank you