Fingerprints of fractionalized excitations in scattering probes of quantum spin liquids

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Spin Dynamics in the DIRAC Systems, SPICE
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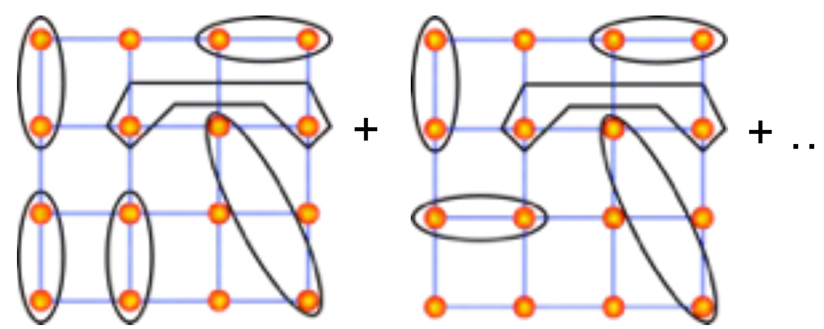
Quantum spin liquids

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

$\Psi =$ 

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.*

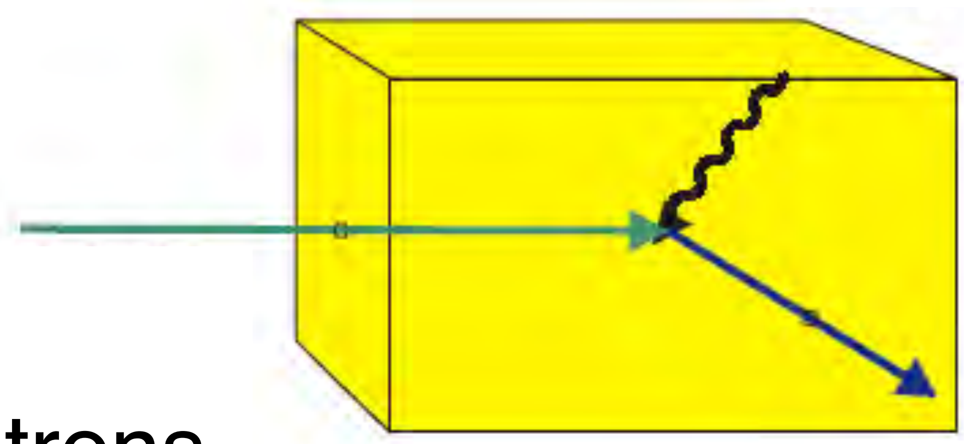
Quantum spin liquids

Main Question:

How to probe fractionalized quasiparticles in QSL and their statistics?

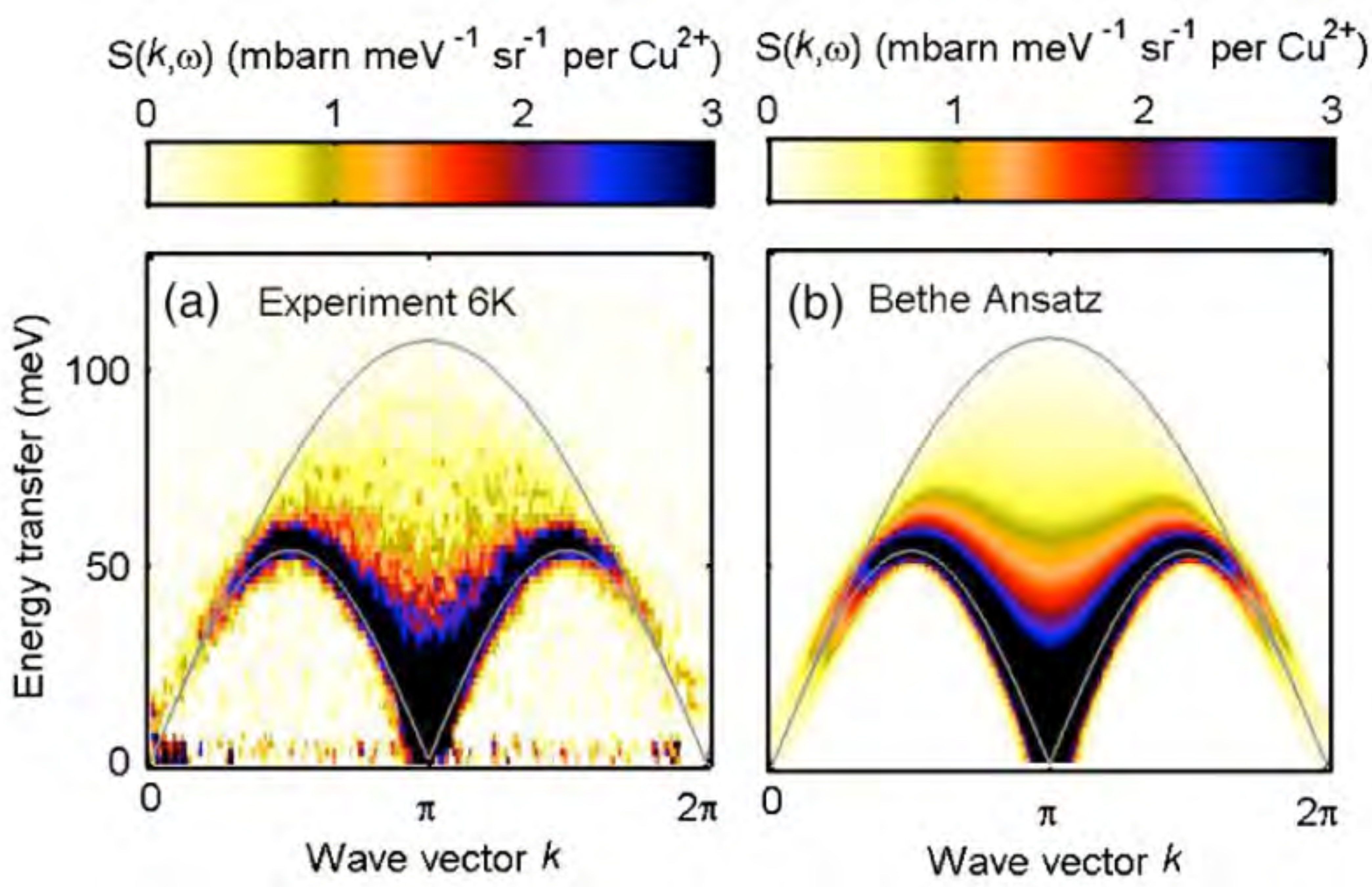
Take home message:

Signatures of quantum order are mainly in the excitations



Since excitations carry fractional quantum numbers relative to the local degrees of freedom, only *multiple quasiparticles* can couple to external probes: Response from QSL is always a multi-particle continuum.

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.

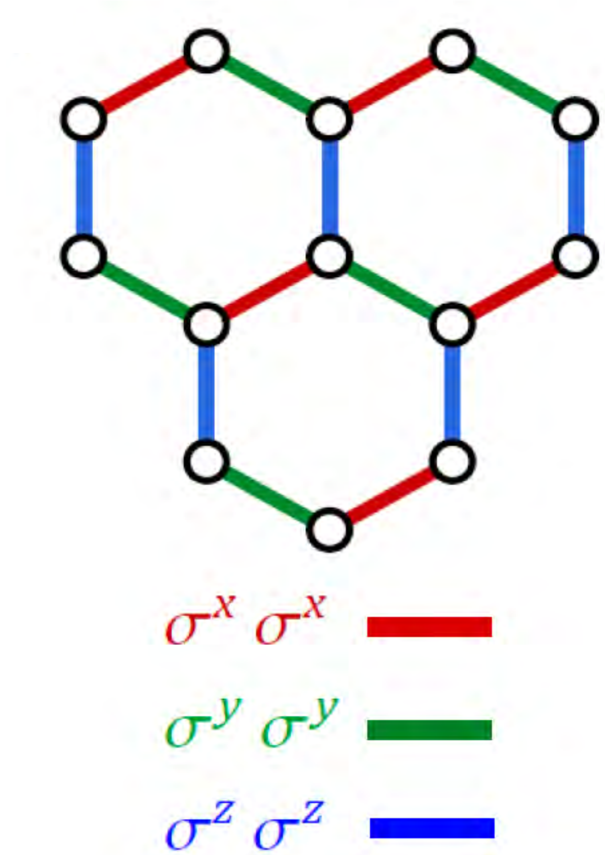
B Lake *et al*, PRL 111, 137205 (2013)

Probing continua of excitations in Kitaev spin liquids

- Spin liquid ground state and fractionalized excitations in 2D and 3D Kitaev models
- Raman response in 2D & 3D Kitaev model
- RIXS response in 2D & 3D Kitaev models
- Conclusions

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

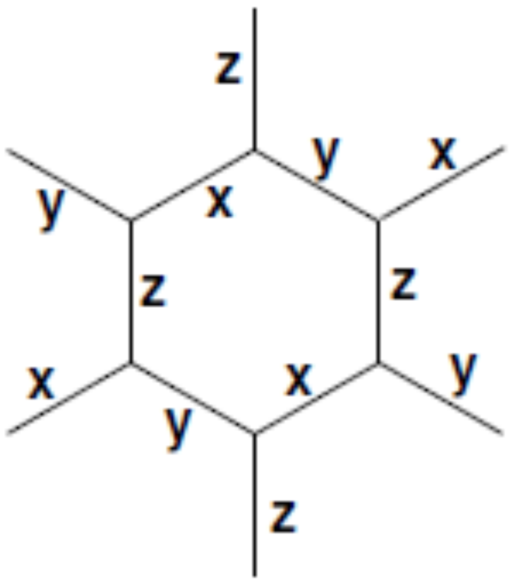
Fractionalized excitation

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)

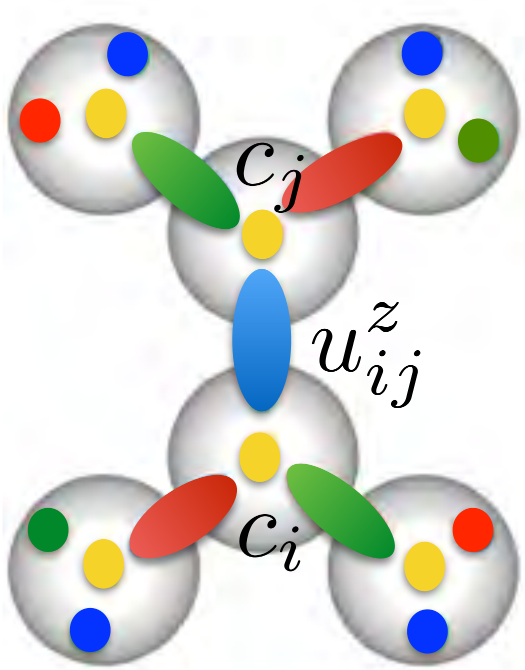
Spin fractionalization and Majorana fermions



Large number of conserved quantities, local plaquette operators:

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

$$[H, W_p] = 0$$

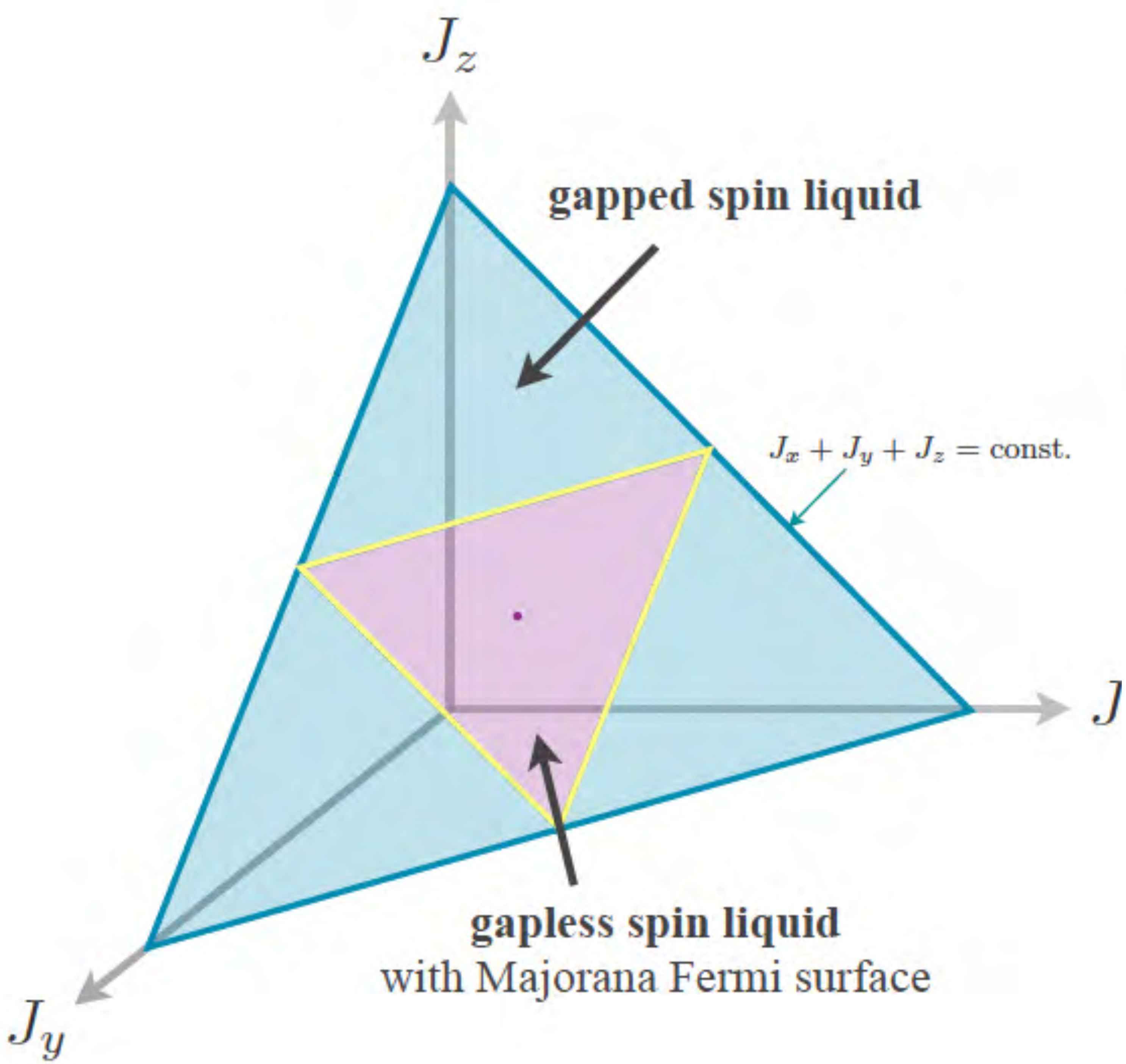


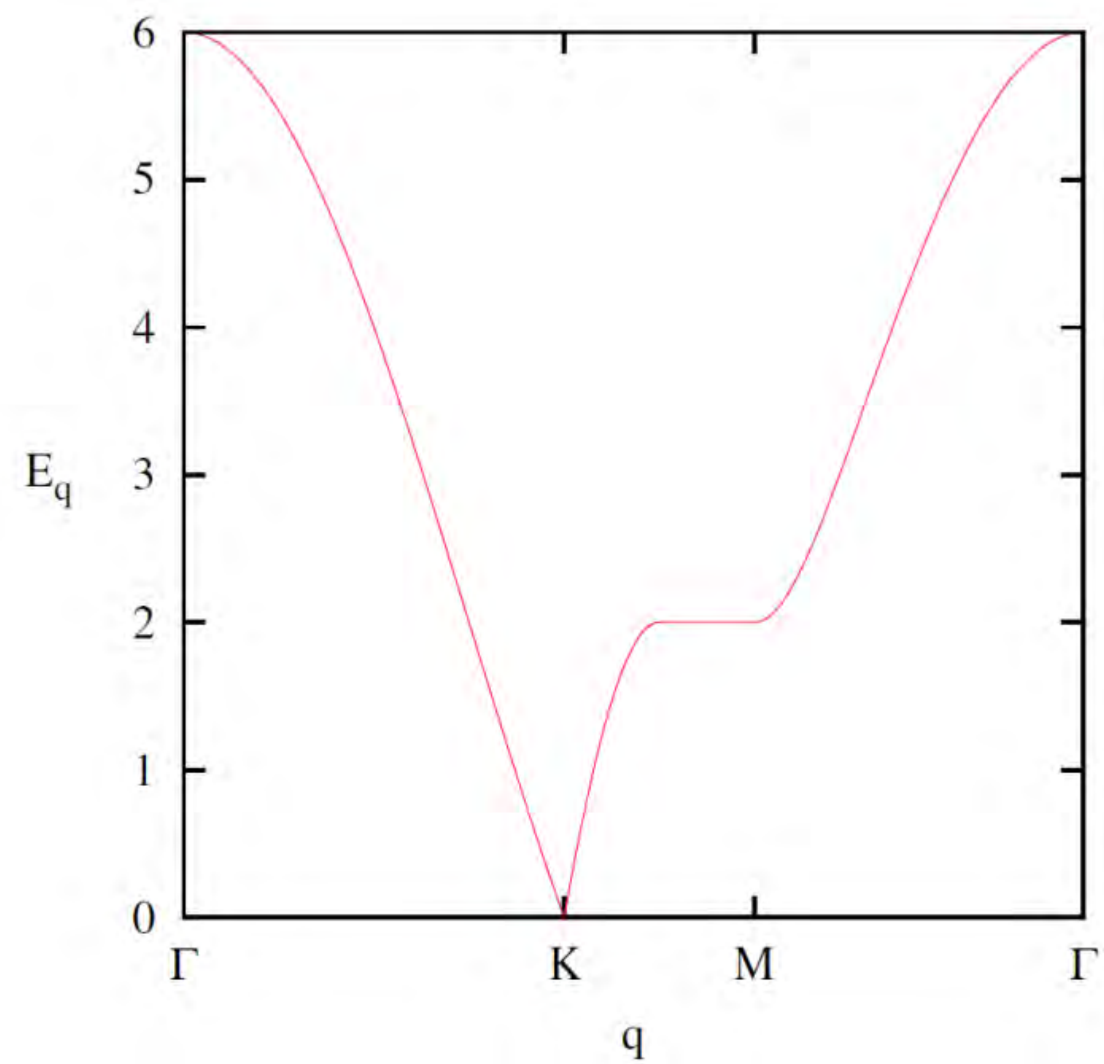
Quadratic Hamiltonian in each flux sector:

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

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

Excitations in the 2D Kitaev spin liquid





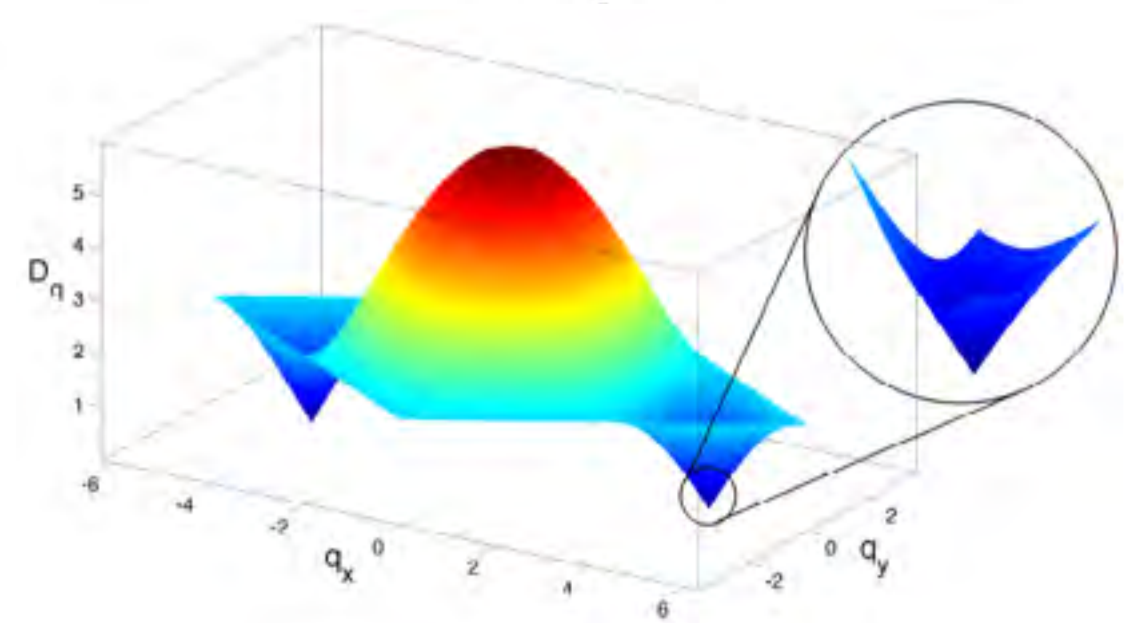
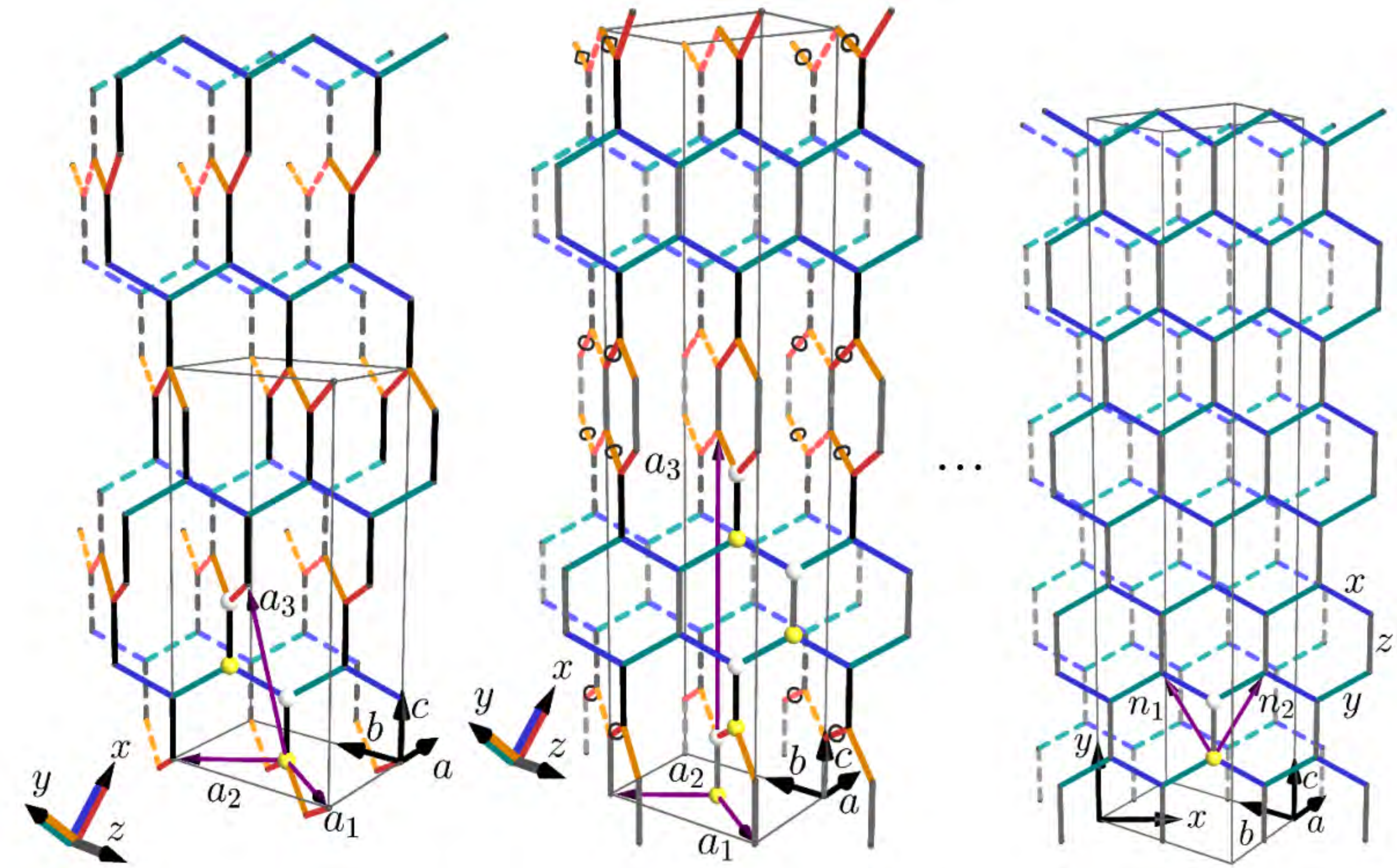


Fig. from M. Hermanns et al, 2014

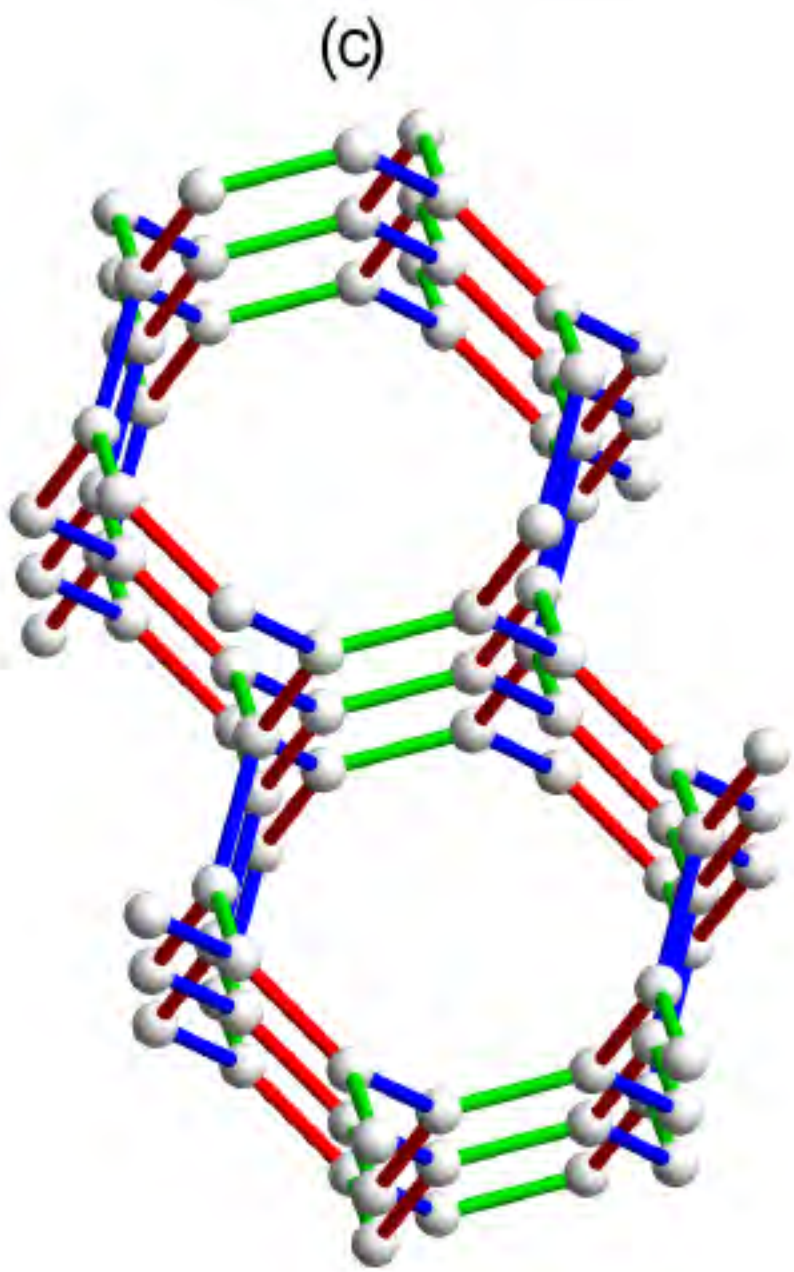
3D Kitaev family



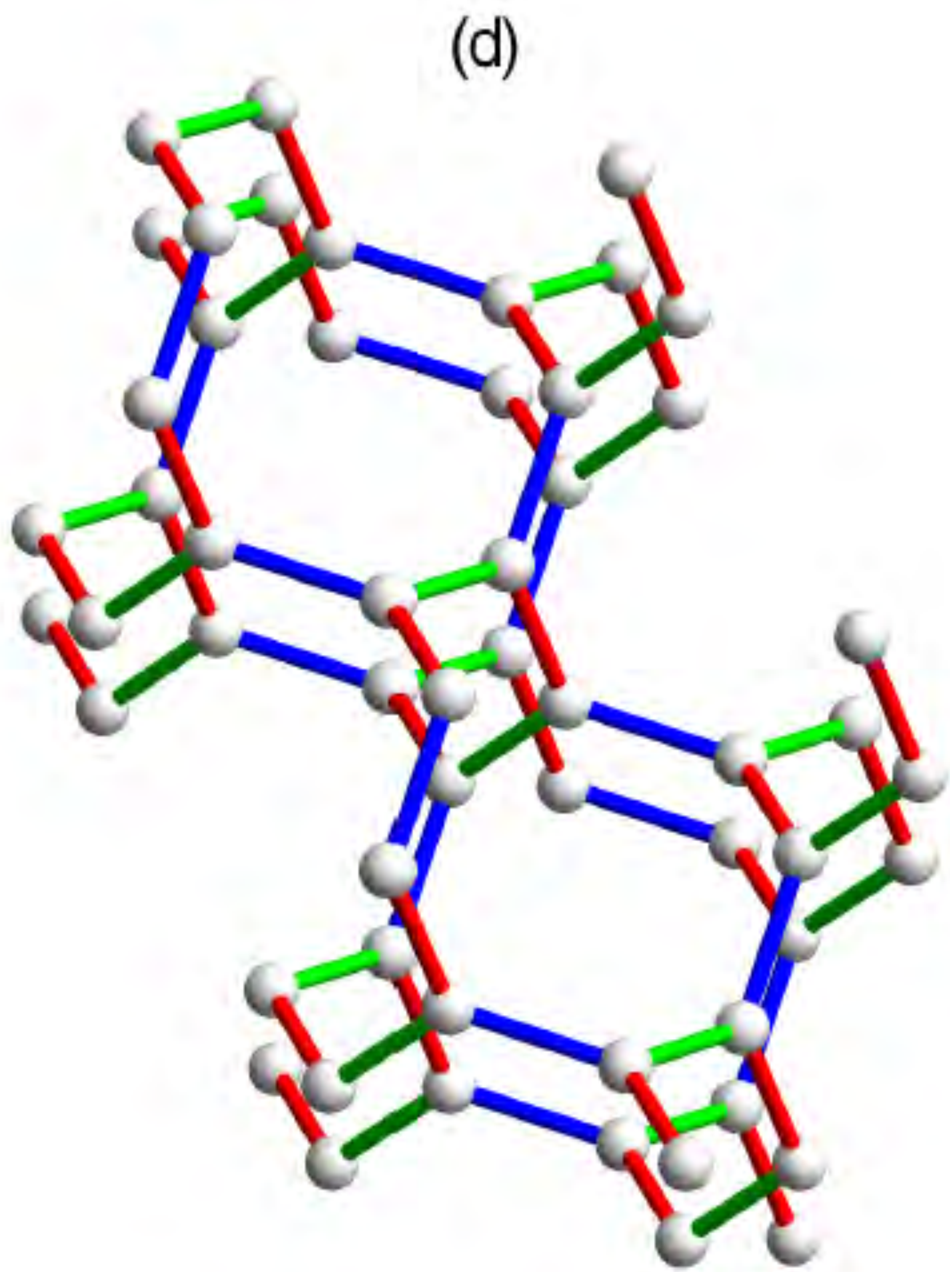
$H_{<0>}$ $H_{<1>}$ $H_{<\infty>}$

Hyperhoneycomb lattices

3D Kitaev family



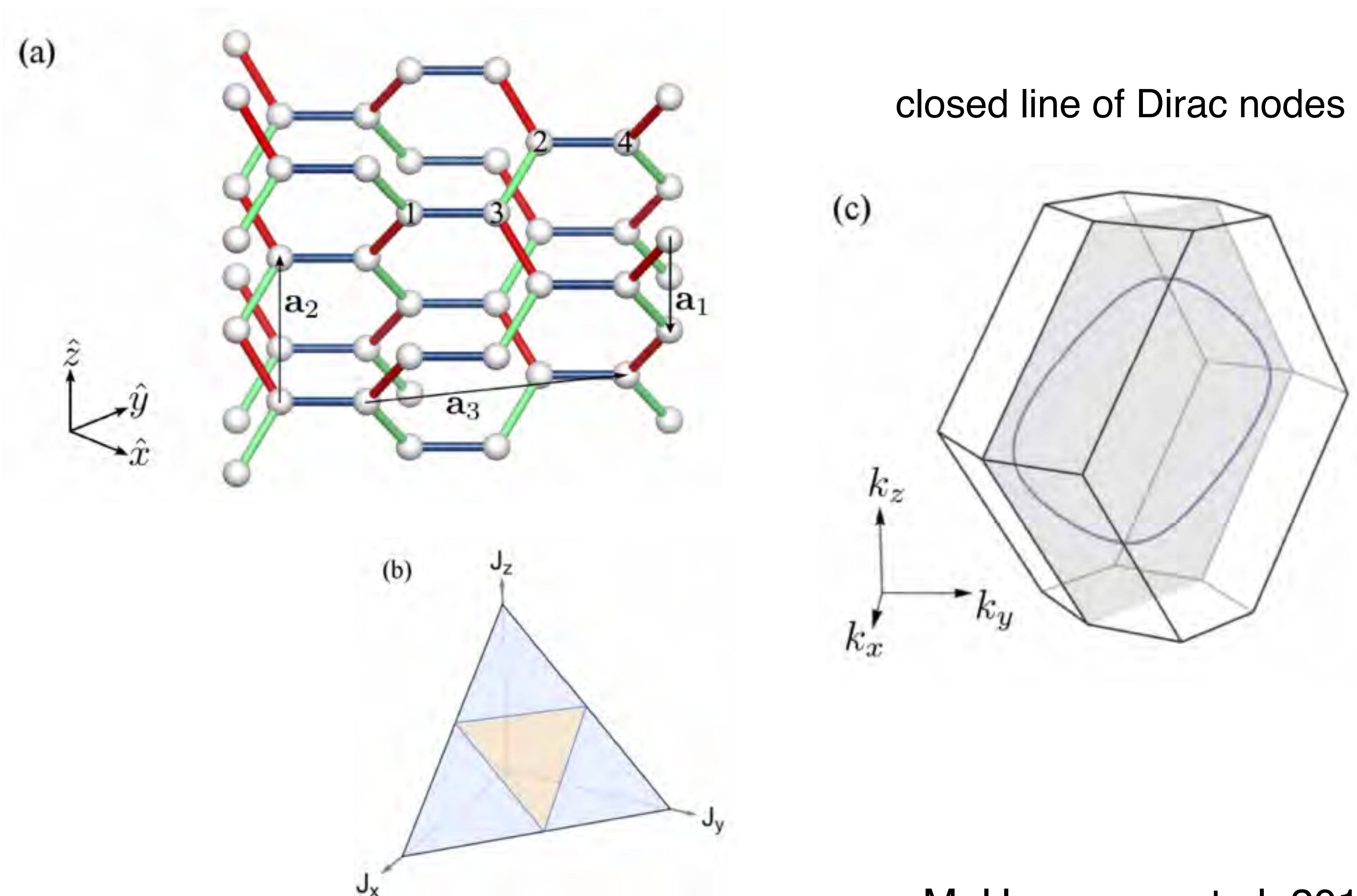
Hyperhexagon



Hyperoctagon

M. Hermanns et al, 2015

Hyperhoneycomb lattice

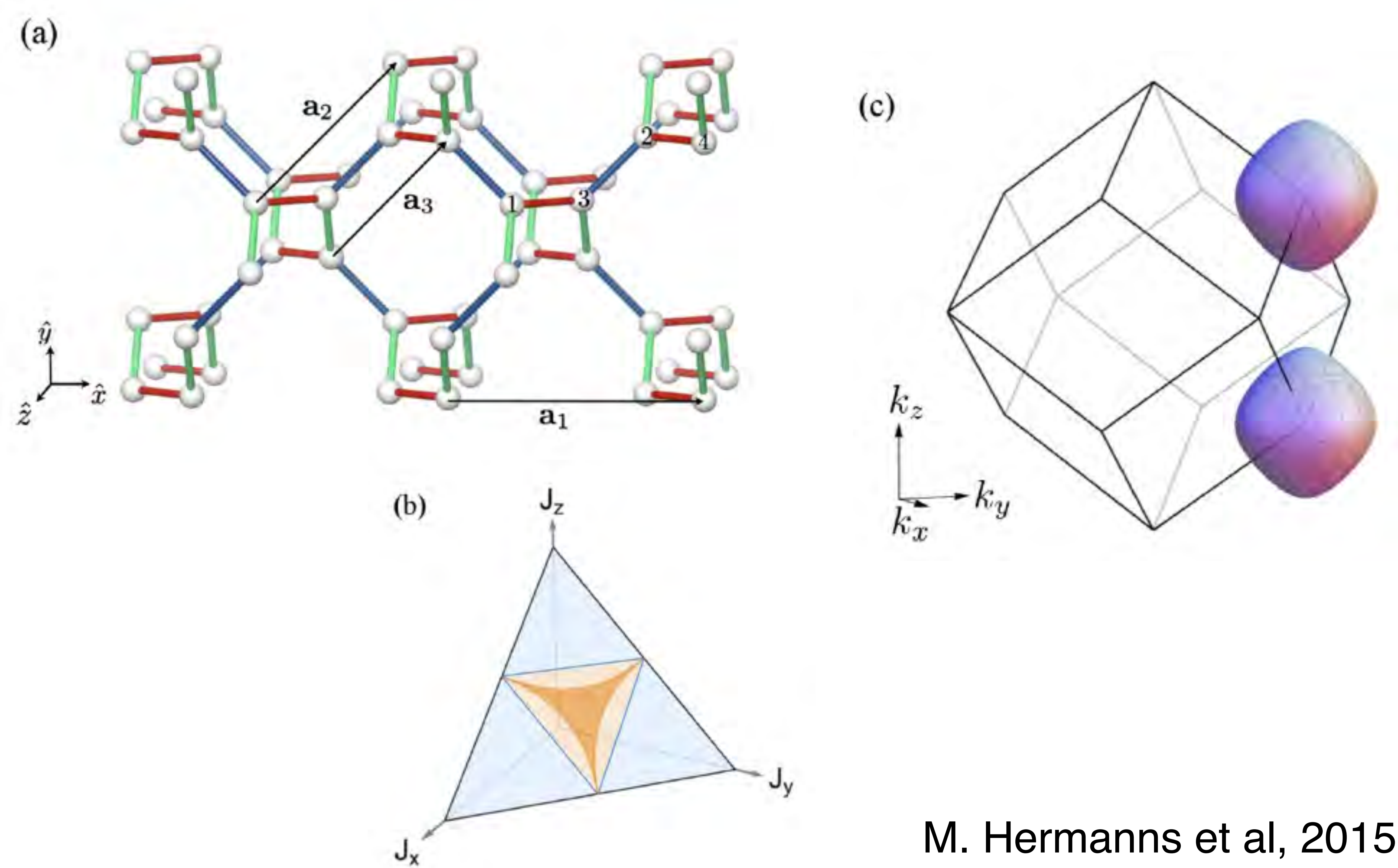


closed line of Dirac nodes

M. Hermanns et al, 2015

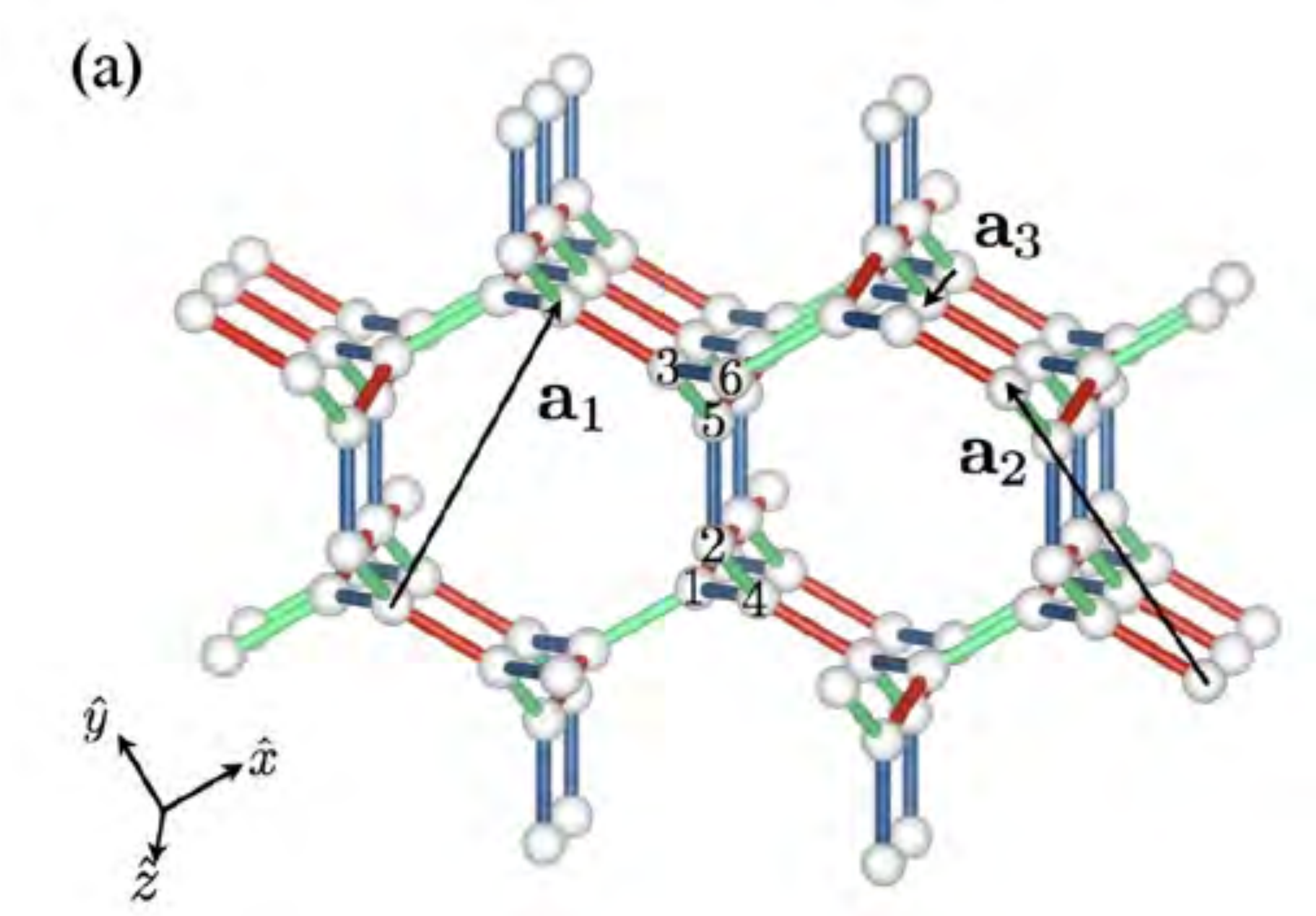
Hyperoctagon lattice
Majorana metal

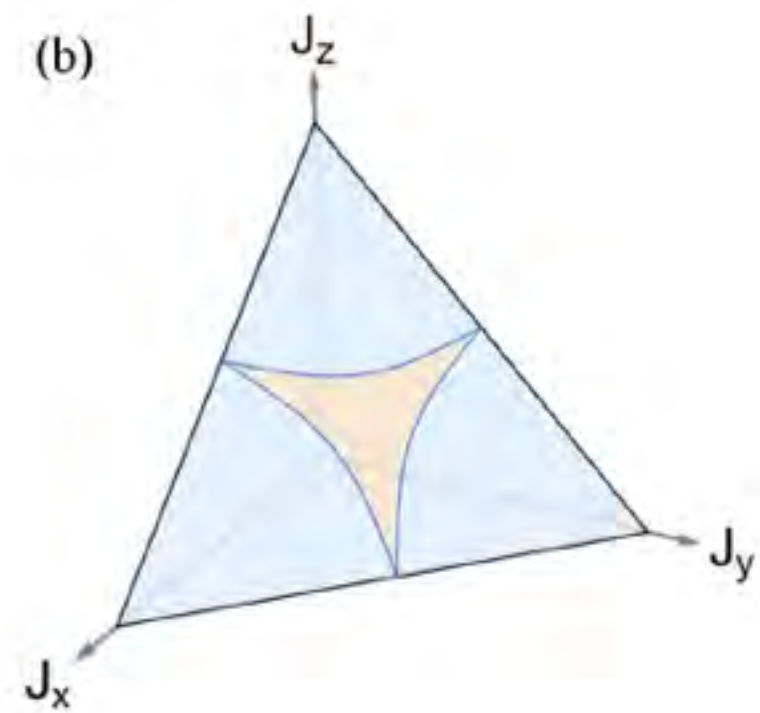
Fermi surfaces



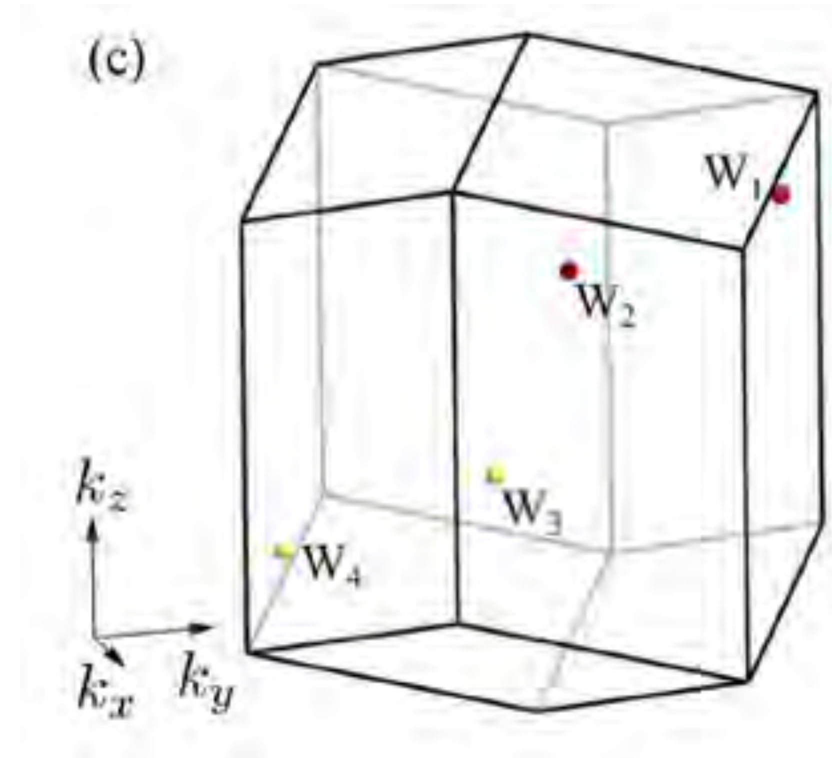
M. Hermanns et al, 2015

Hyperhexagon lattice





gapless Weyl points

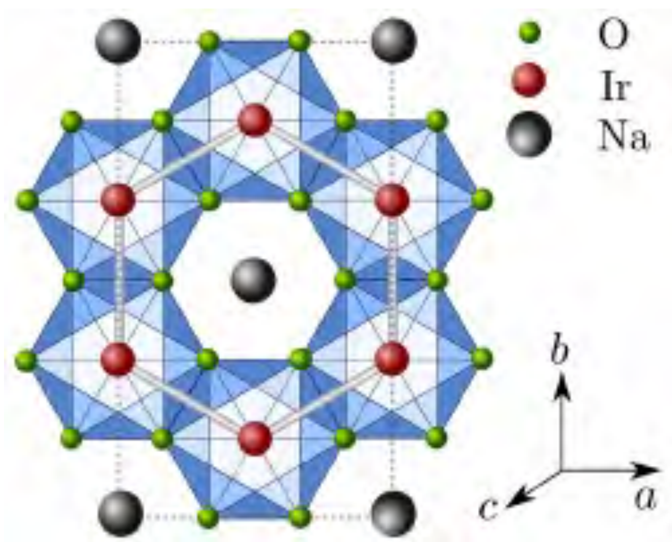


$$\hat{H}_{2\times 2} = \mathbf{v}_0 \cdot \mathbf{q}\,\mathbb{1} + \sum_{j=1}^{3} \mathbf{v}_j \cdot \mathbf{q}\,\sigma_j$$

M. Hermanns et al, 2015

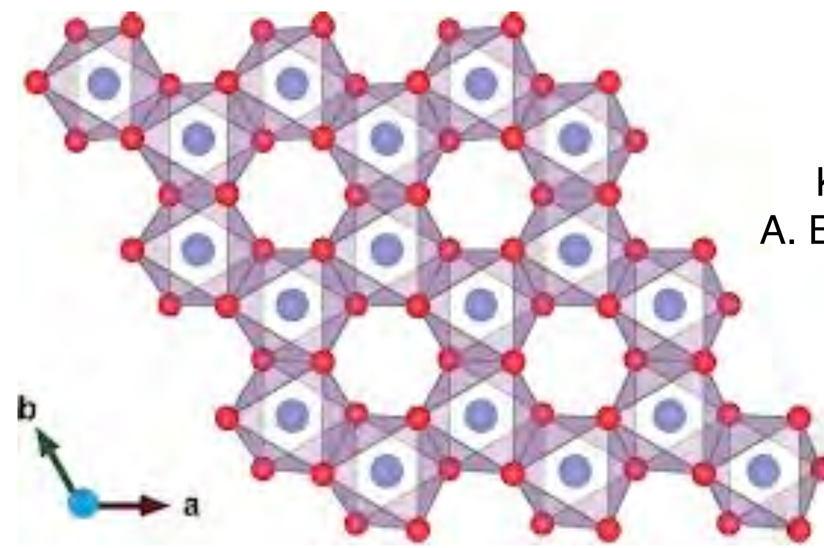
Experimental realizations

Na_2IrO_3
alpha-Li_2IrO_3



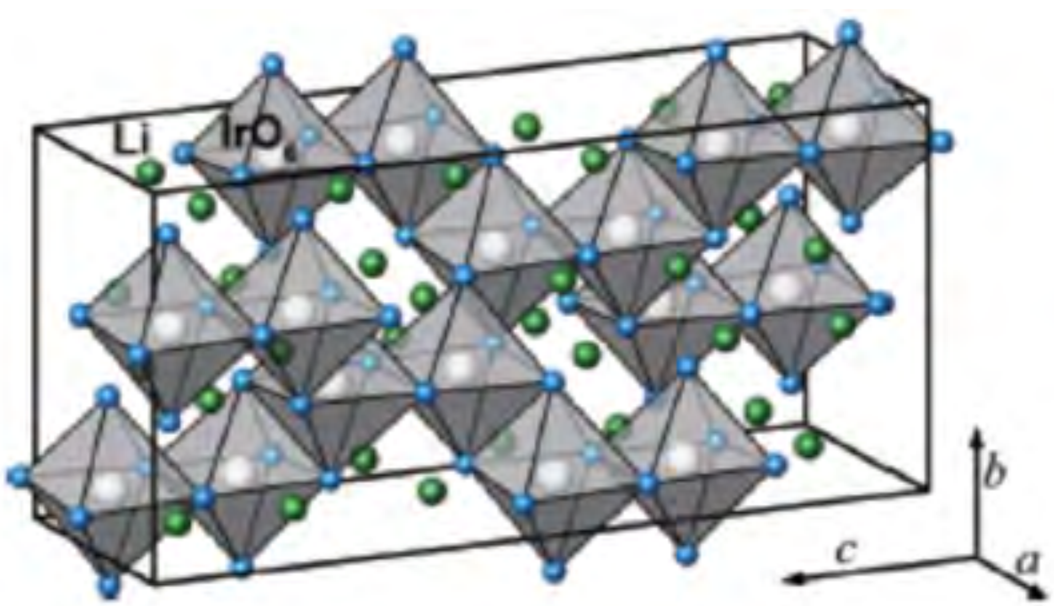
Y.Singh, P. Gegenwart,
PRL 2010, 2011

alpha-$RuCl_3$



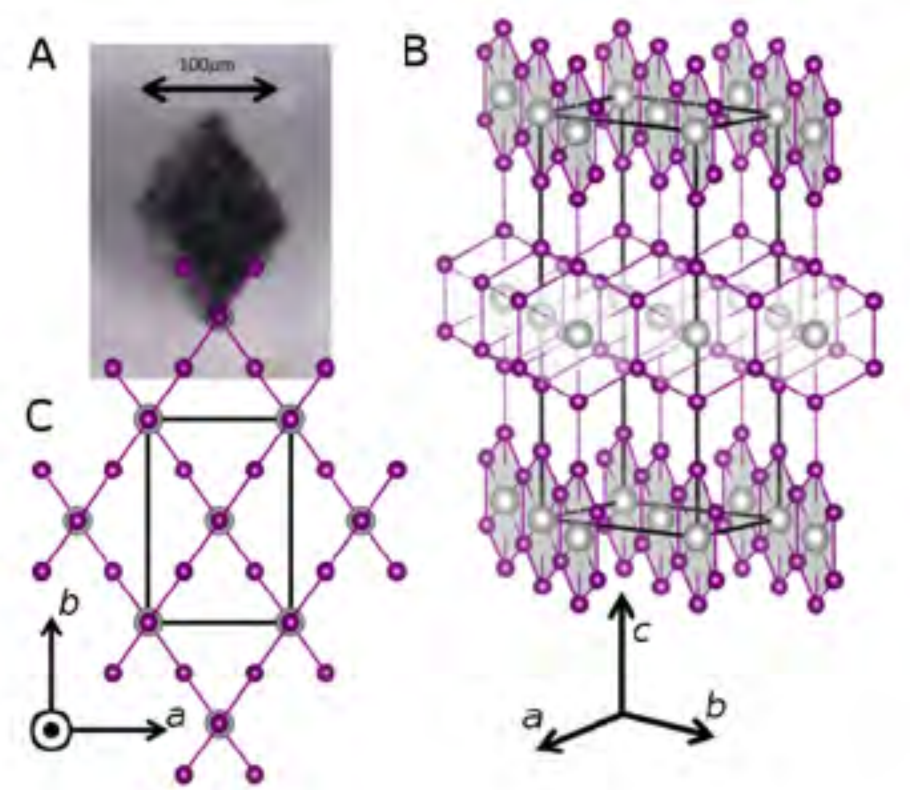
K. Plumb et al, Phys. Rev. B (2014)
A. Banerjee et al, Nature Materials (2016)

beta-Li_2IrO_3



T. Takayama et al, PRL (2015)

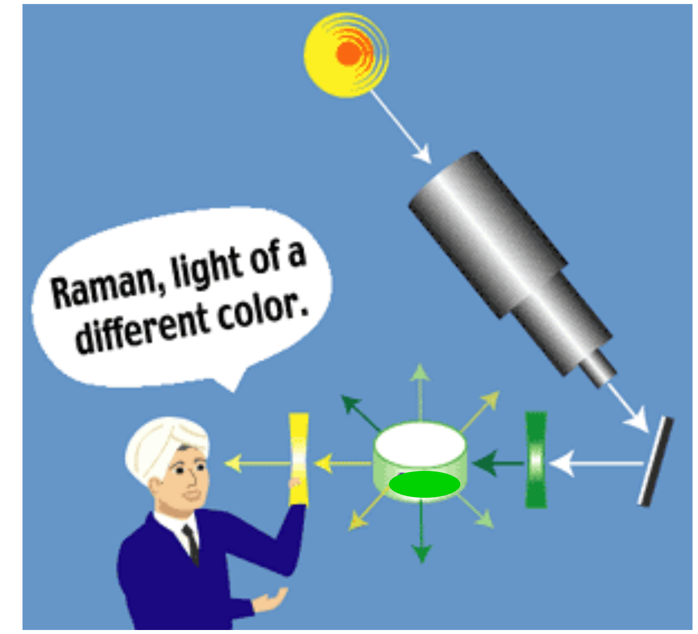
gamma-Li_2IrO_3



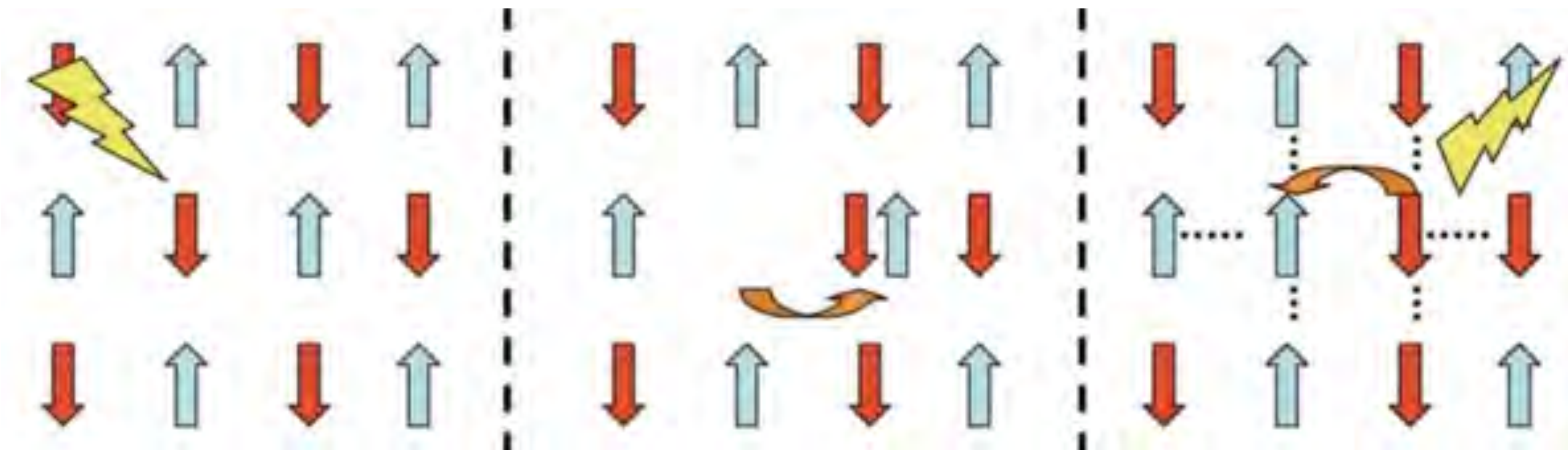
Modic, Nature Comminications **5**, 4203 (2014)

Spectroscopy of Kitaev Spin Liquids

Raman scattering in a nutshell



Photon-in photon-out process



Photon induced spin exchange

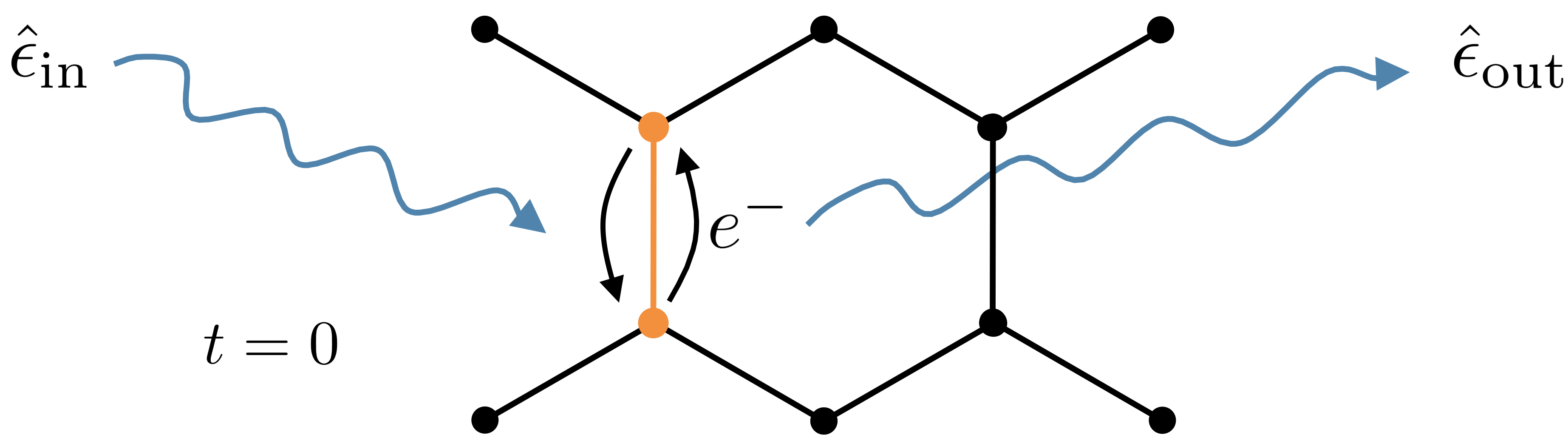
Two-magnon scattering in Mott insulators:
Loudon-Fleuri scattering vertex

$$R = \sum_{i,\pm\boldsymbol{\delta}_\mu} (\hat{\varepsilon}_{\text{in}} \cdot \boldsymbol{\delta}_\mu)(\hat{\varepsilon}_{\text{out}} \cdot \boldsymbol{\delta}_\mu)\, J_\mu\, \mathbf{S}_i \cdot \mathbf{S}_{i\pm\boldsymbol{\delta}_\mu}$$

Devereaux, Hackl *RMP* 2007

Raman Scattering in Kitaev model

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



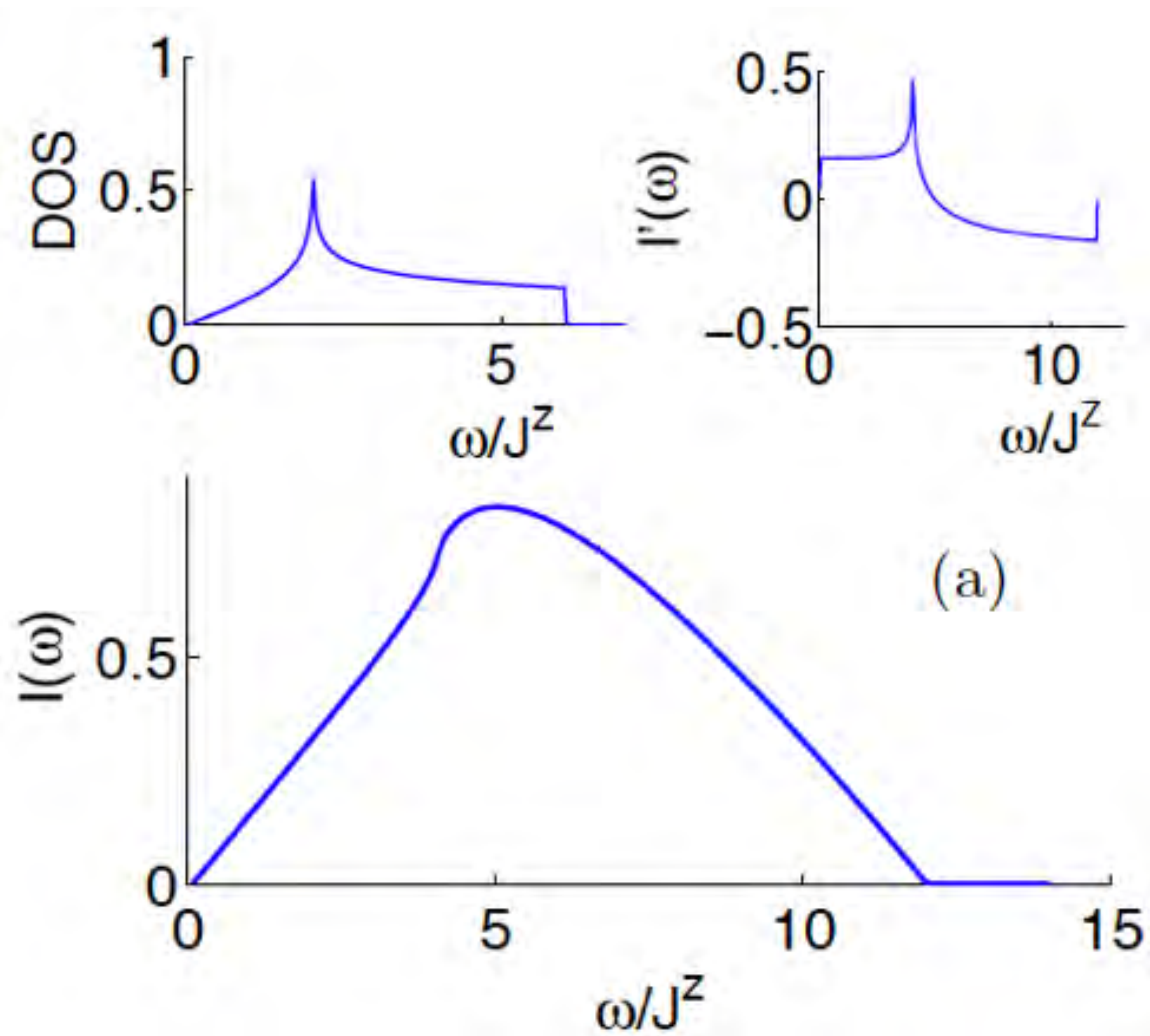
$$R = \sum_{\langle ij \rangle^\alpha} (\boldsymbol{\epsilon}_{\mathrm{in}} \cdot \mathbf{d}^\alpha)(\boldsymbol{\epsilon}_{\mathrm{out}} \cdot \mathbf{d}^\alpha) J^\alpha \sigma_i^\alpha \sigma_j^\alpha$$

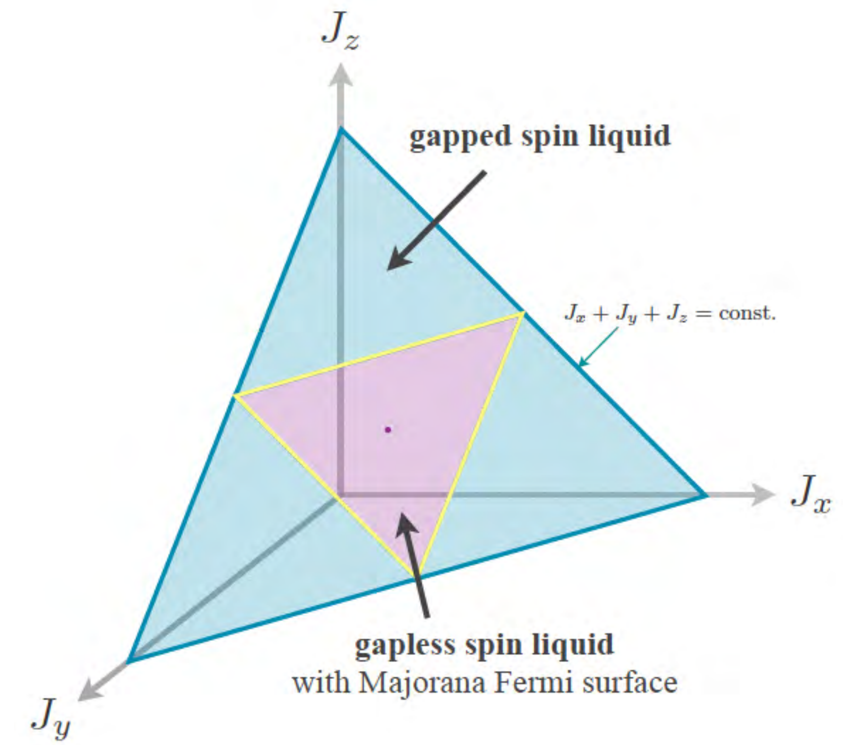
$$= i \sum_{\langle ij \rangle^\alpha} (\boldsymbol{\epsilon}_{\mathrm{in}} \cdot \mathbf{d}^\alpha)(\boldsymbol{\epsilon}_{\mathrm{out}} \cdot \mathbf{d}^\alpha) J^\alpha u_{\langle ij \rangle^\alpha} c_i c_j$$

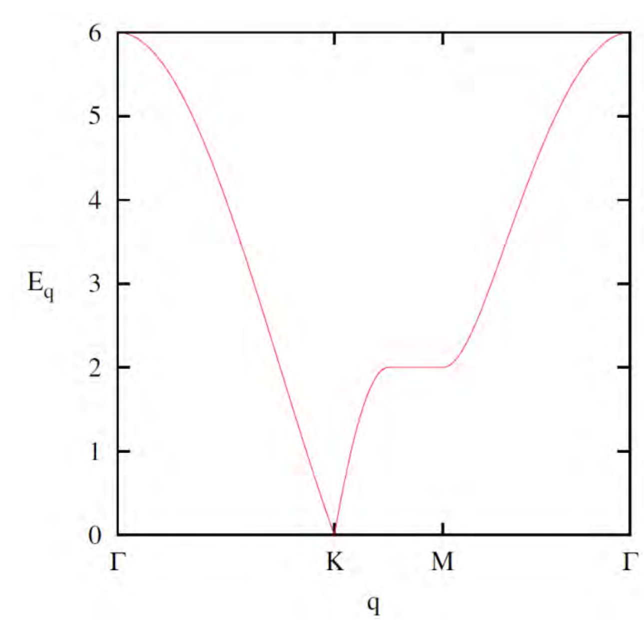
Raman vertex: diagonal in fluxes but **creates two Majorana fermions**

Raman Scattering results (2D)

isotropic point:
polarization independent

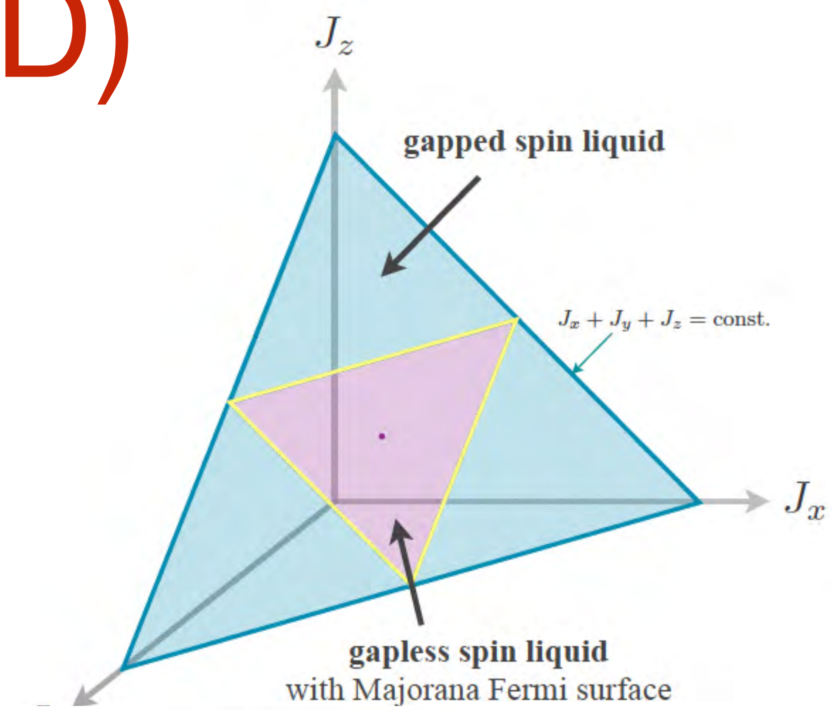




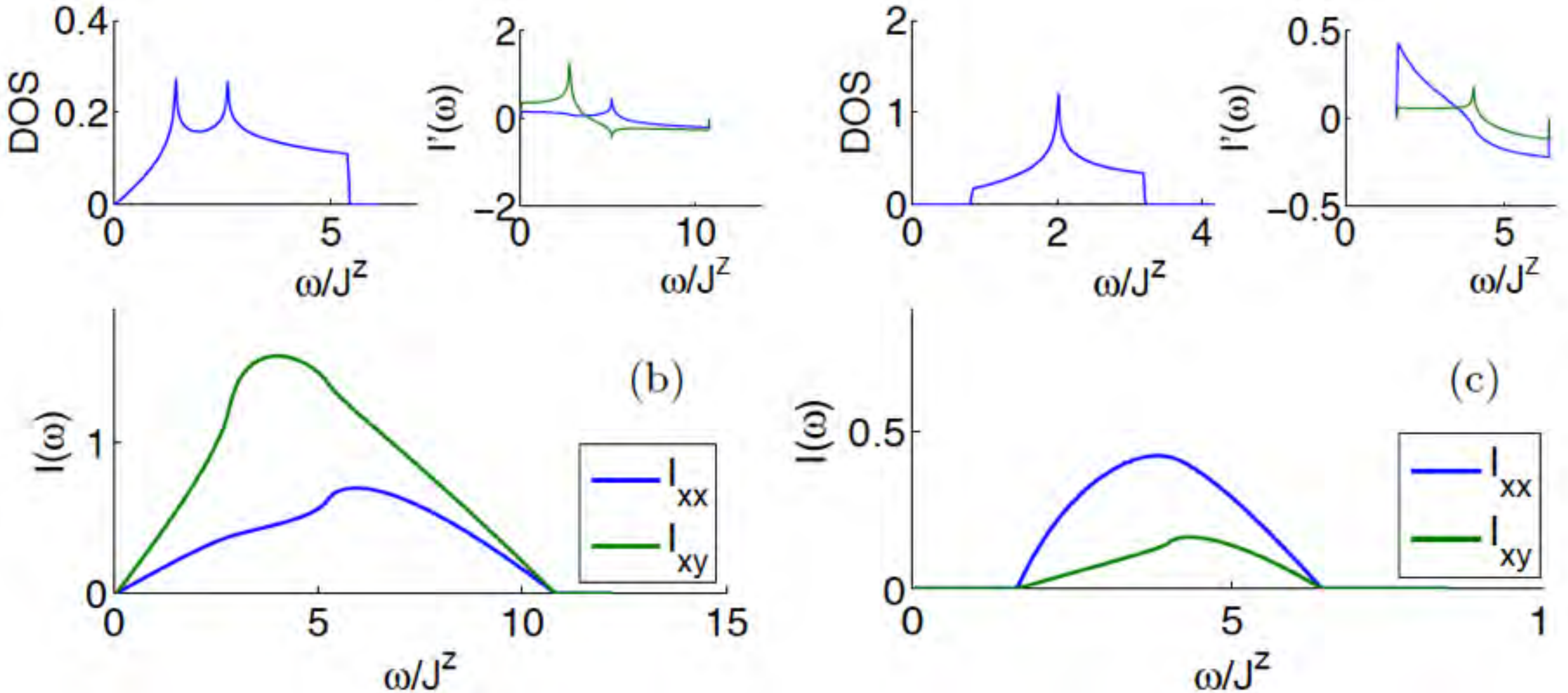


Knolle, Chern, Kovrizhin, Moessner, **NP** (PRL 2014)

Raman Scattering results (2D)



anisotropic point:
polarization dependence



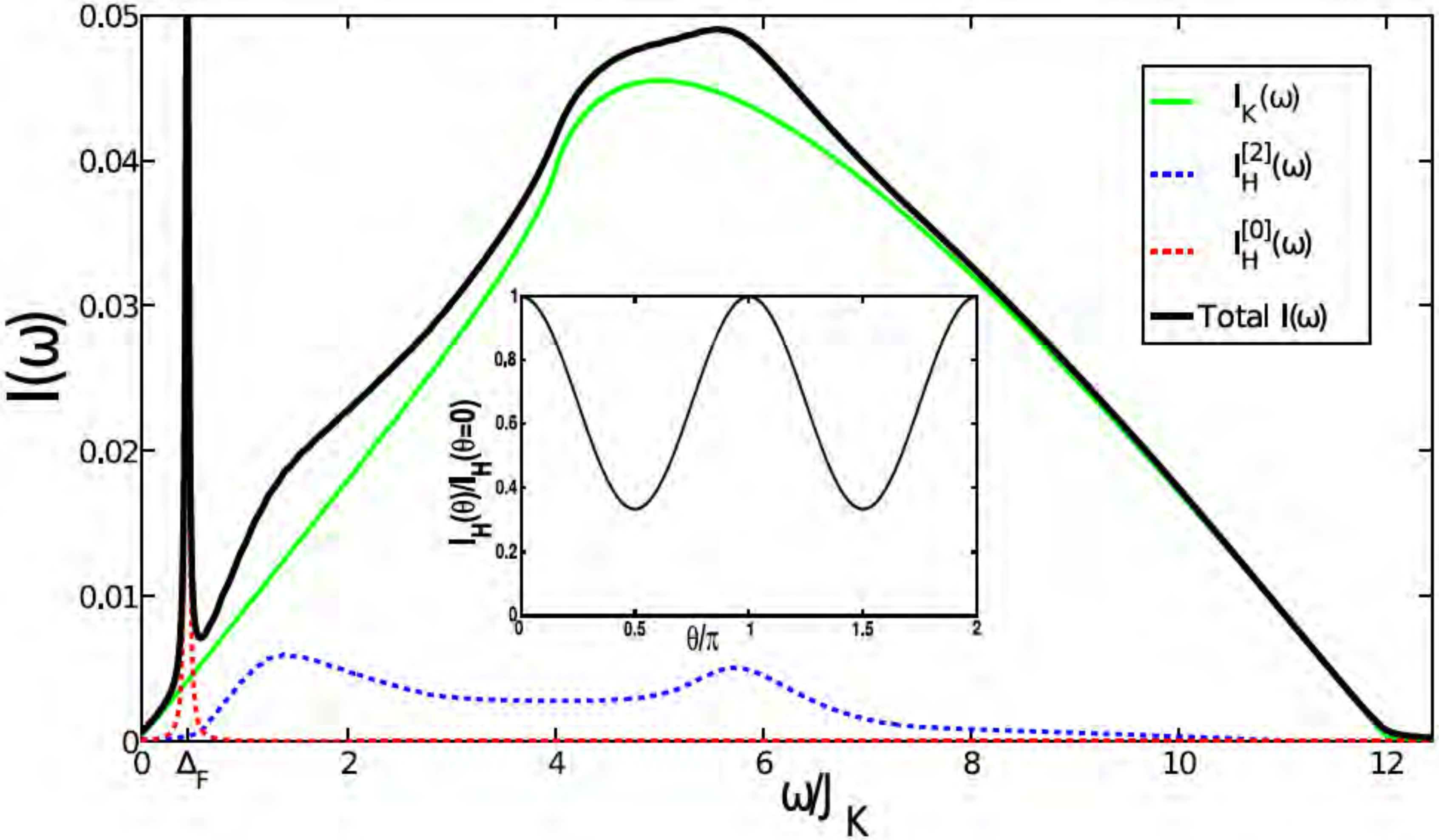
Knolle, Chern, Kovrizhin, Moessner, **NP** (PRL 2014)

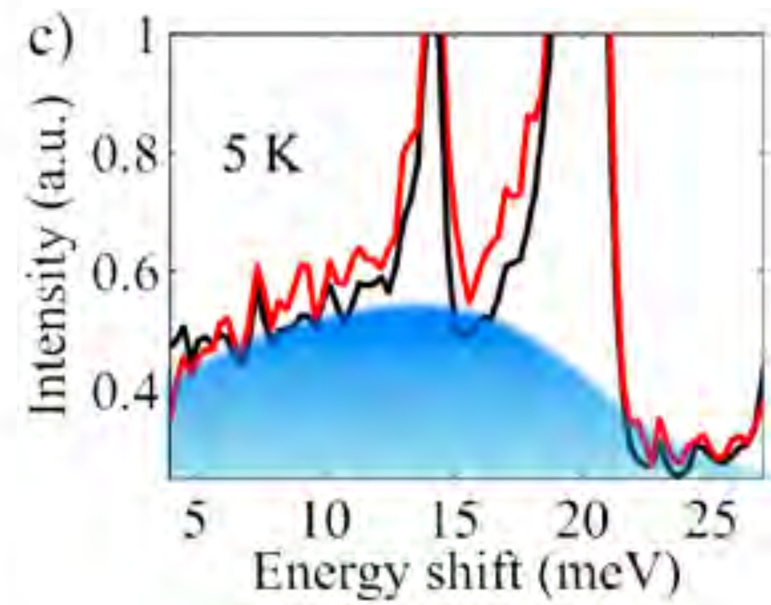
Raman Scattering results (2D)

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

computed perturbatively

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





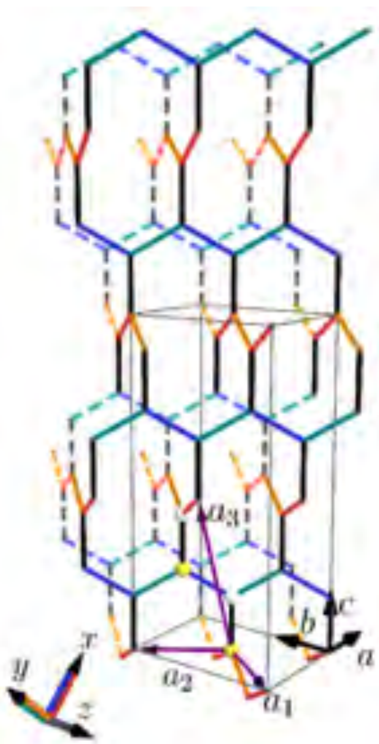
Big 'hump' with fine features of the Majorana DOS. Salient signatures of fractionalization are visible!
(comparison gives J_K~8meV)

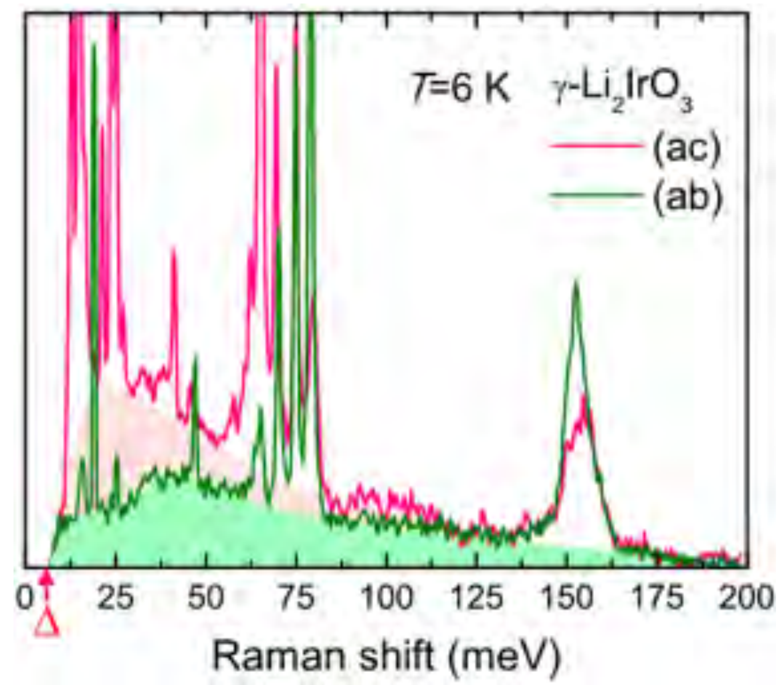
The Heisenberg contribution depends on the photon polarizations, δ-function peak at the four flux gap.

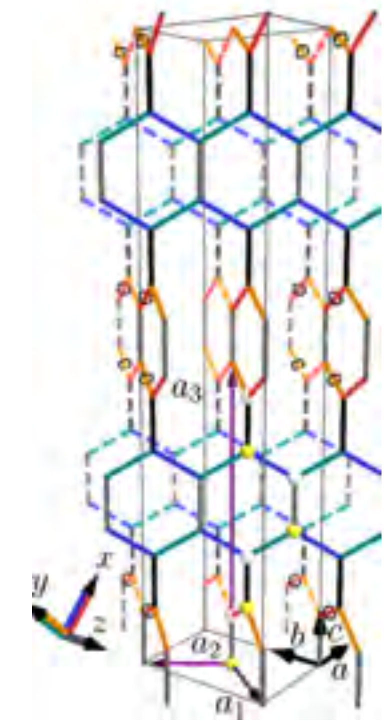
Knolle, Chern, Kovrizhin, Moessner, **NP** (PRL 2014)

Raman Scattering results (3D)

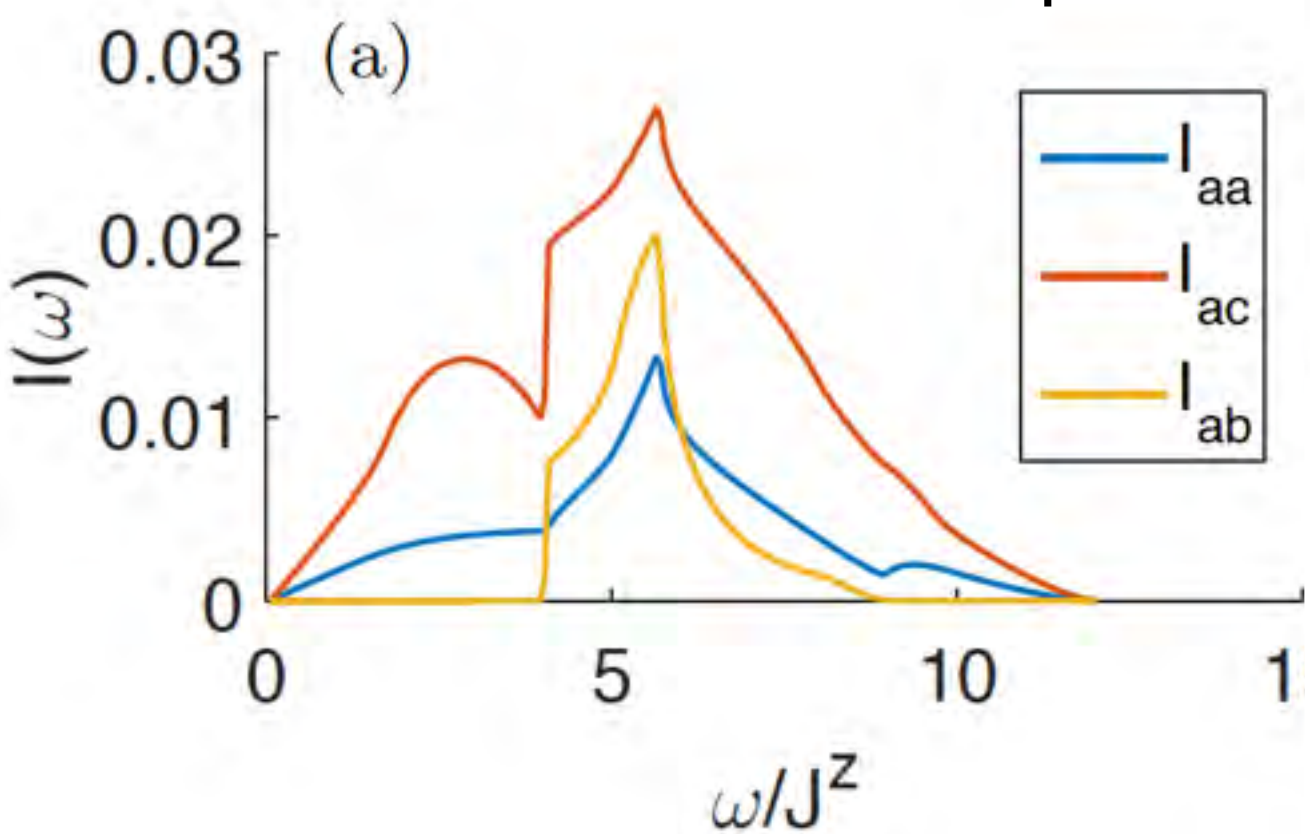
Glamazda, Lemmens, Do, Choi, Choi, Nature Comm. 7 (2016)

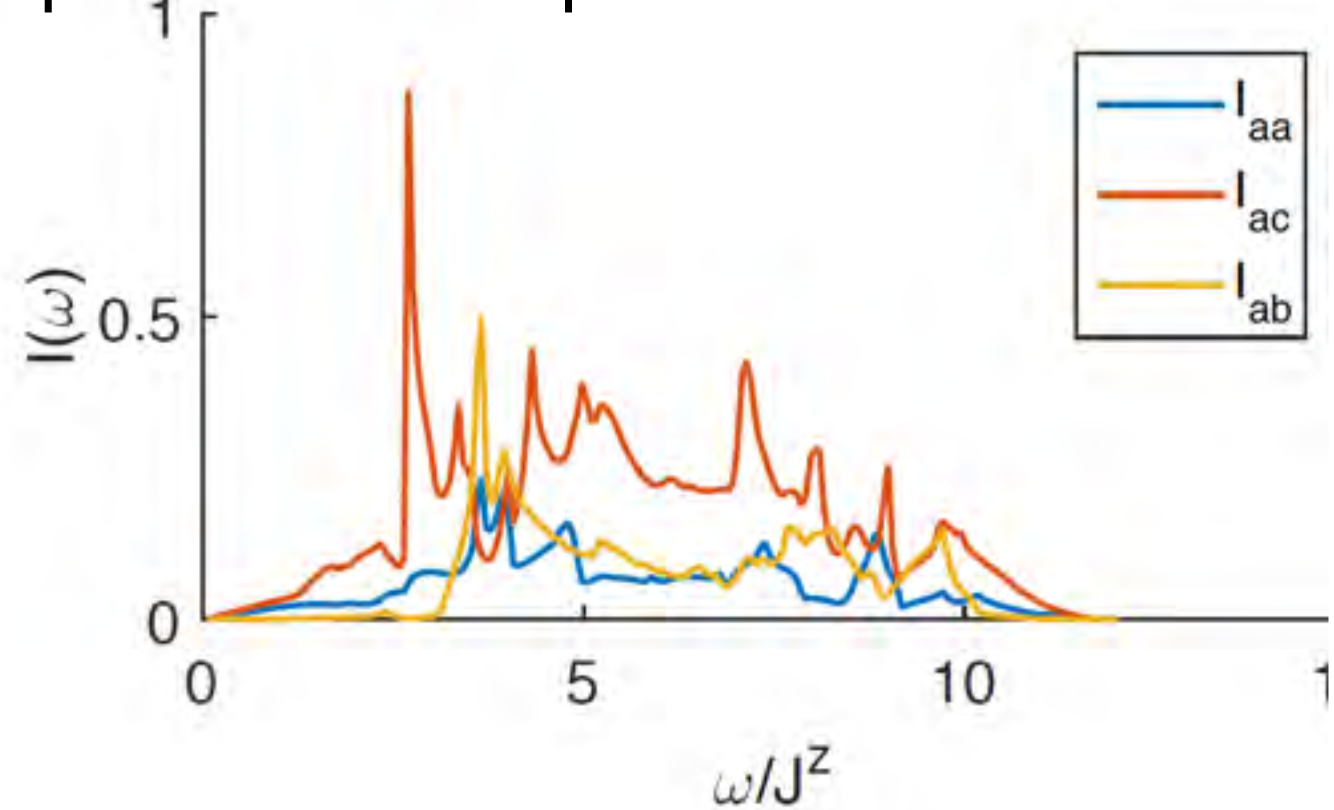






The Raman response is polarization dependent!

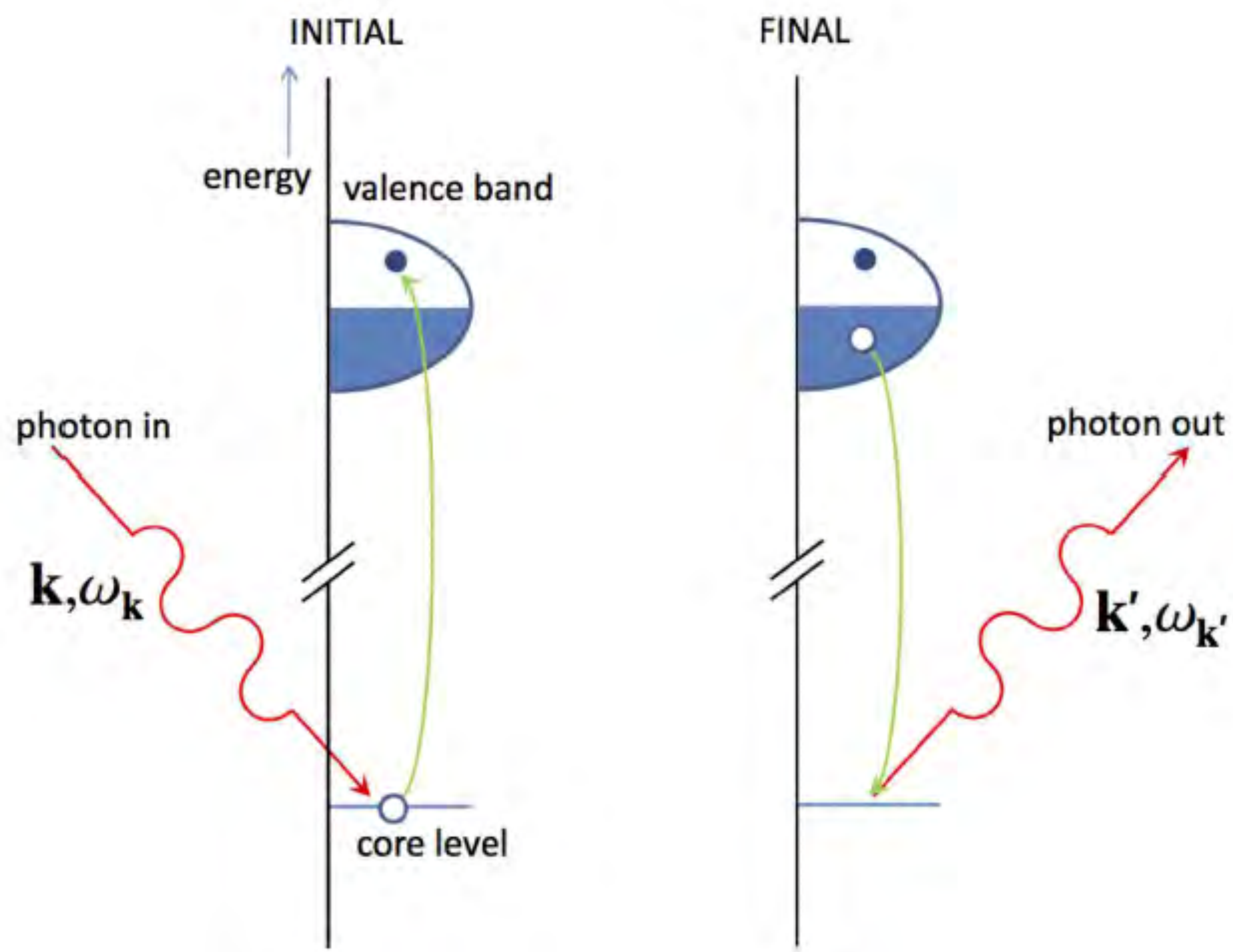




Brent Perreault, Johannes Knolle, NBP, F. J. Burnell, PRB 2015

RIXS in a nutshell

In L-edge experiments, the scattering typically, though not always, happens through a direct RIXS process.



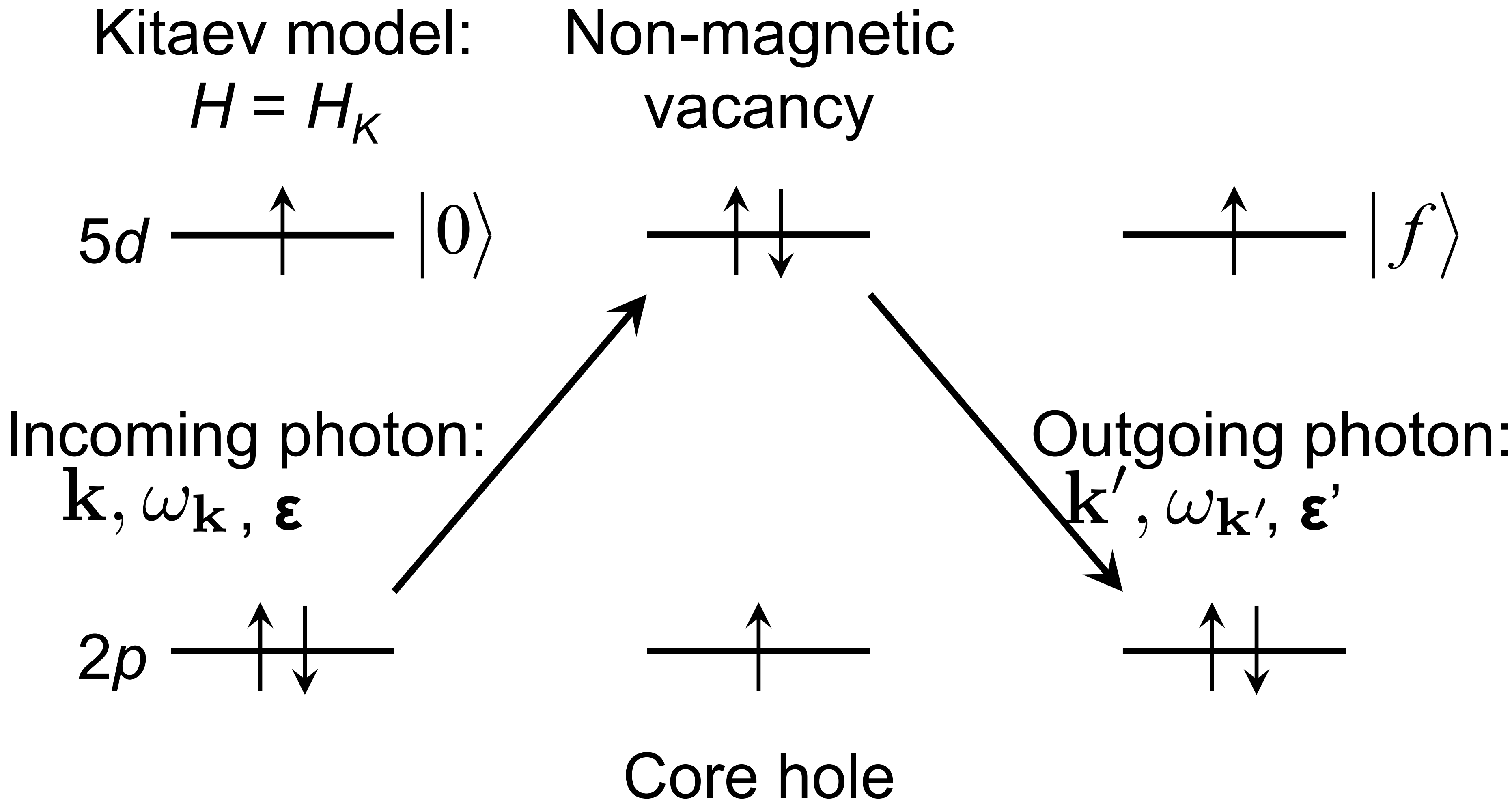
$$\mathbf{q} = \mathbf{k}' - \mathbf{k}$$

$$\omega = \omega_{\mathbf{k}'} - \omega_{\mathbf{k}}$$

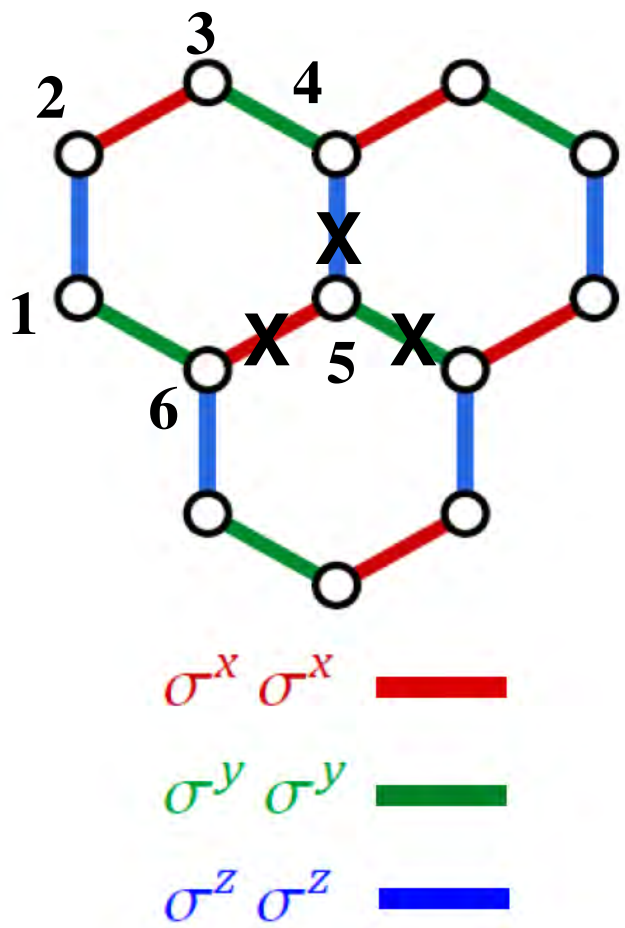
L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, J. van den Brink, RMP (2011)

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)

We demand that the vacancy is always in the spin-up state.

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

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

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

Halász, Chalker, Moessner, PRB 2014

RIXS amplitude for the Kitaev model

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

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

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

Kramers–Heisenberg formula

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

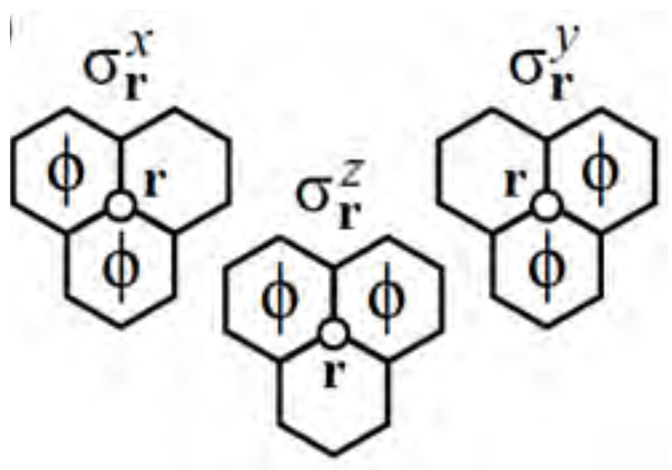
(a) Spin-conserving (SC) channel with

$$T_{\alpha\beta} \propto \sigma^0_{\alpha\beta}$$

(b) three non spin-conserving (NSC) channels with

$$T_{\alpha\beta} \propto \sigma^{x,y,z}_{\alpha\beta}$$

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

$$A_\eta(m, \mathbf{q}) \propto \sum_{\mathbf{r}} e^{i\mathbf{q}\cdot\mathbf{r}} \langle m|\sigma^\eta_{\mathbf{r}} \left[1 - \frac{i\tilde{H}(\mathbf{r})}{\Gamma}\right] |0\rangle$$

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

NSC channels recover INS amplitudes for infinite Γ

Flux creation: Finite gap, little dispersion

Results: SC channel
2D Kitaev model

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

X

Elastic response

Inelastic response

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

no flux and two fermion excitations

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

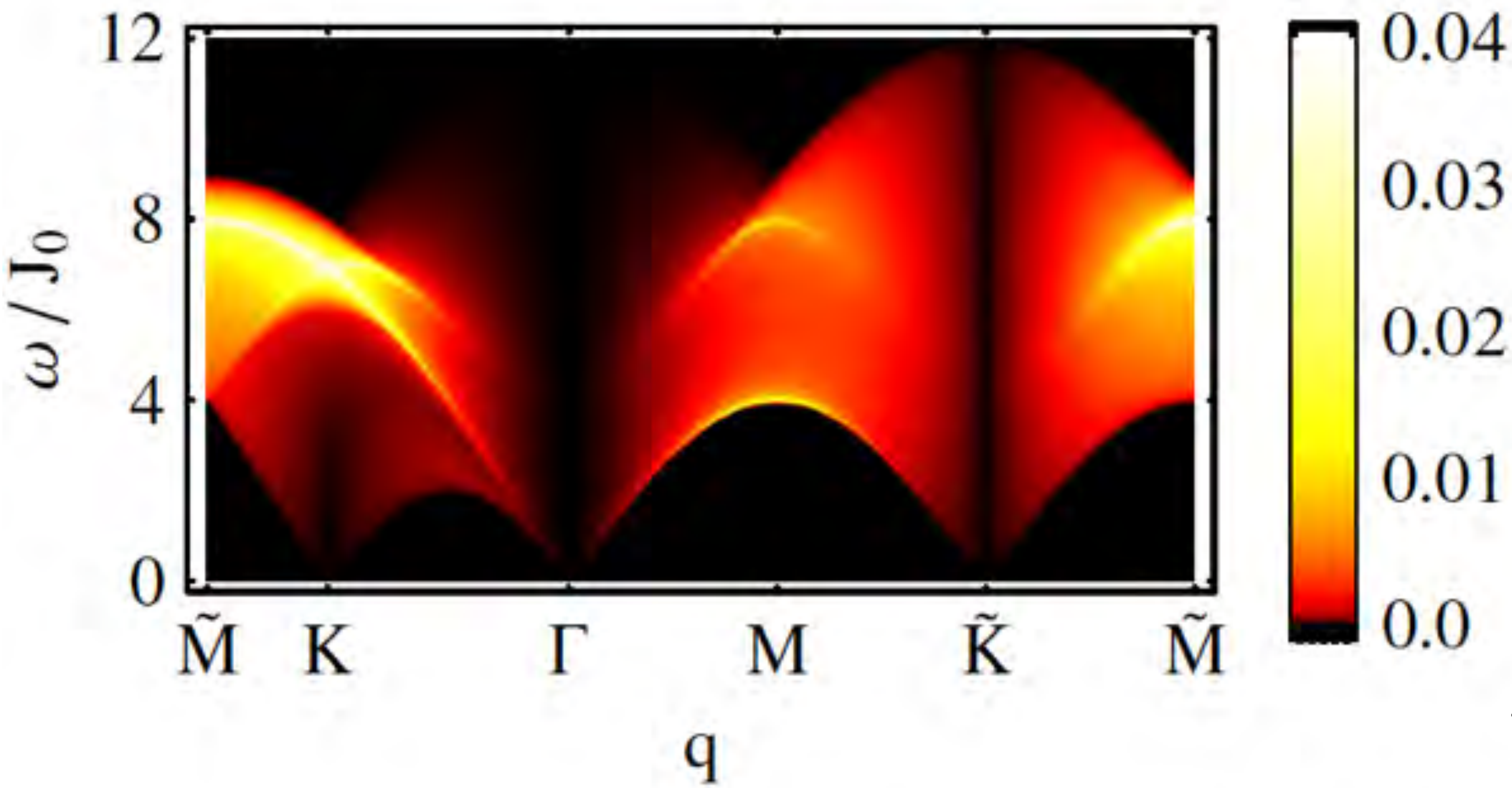
$I_0(\omega,\mathbf{q})$ as a histogram of $|A_0(m,\mathbf{q})|^2$

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

A B

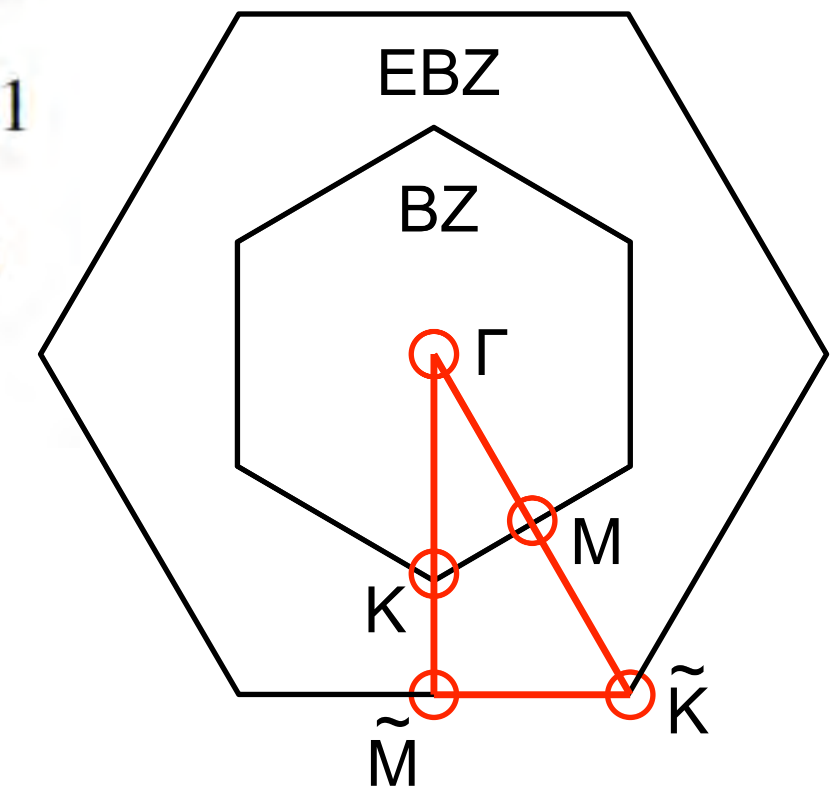
interference between the two sublattices

RIXS response in SC channel



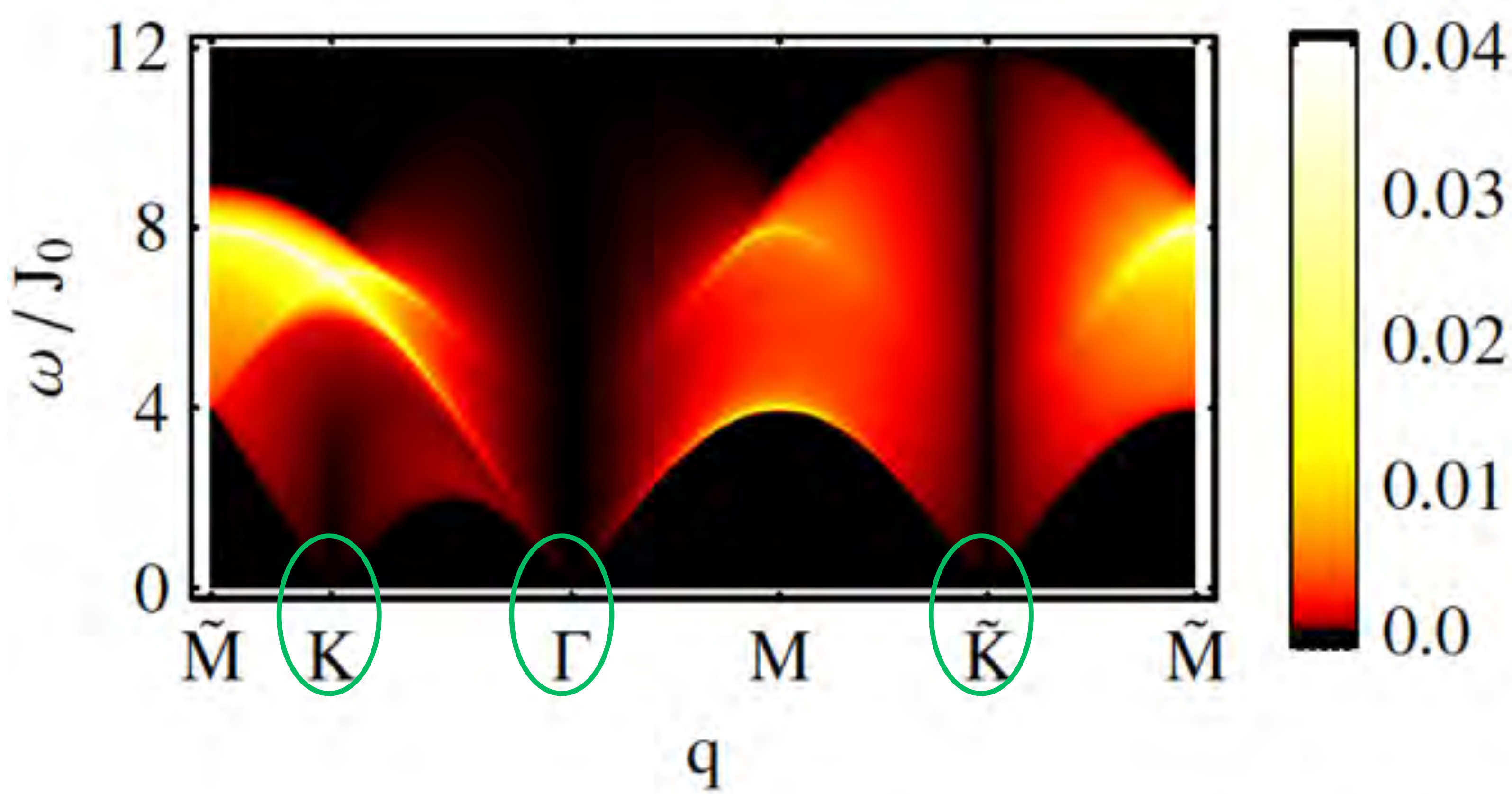
$$J_{x,y,z} = J_0$$

Reciprocal space



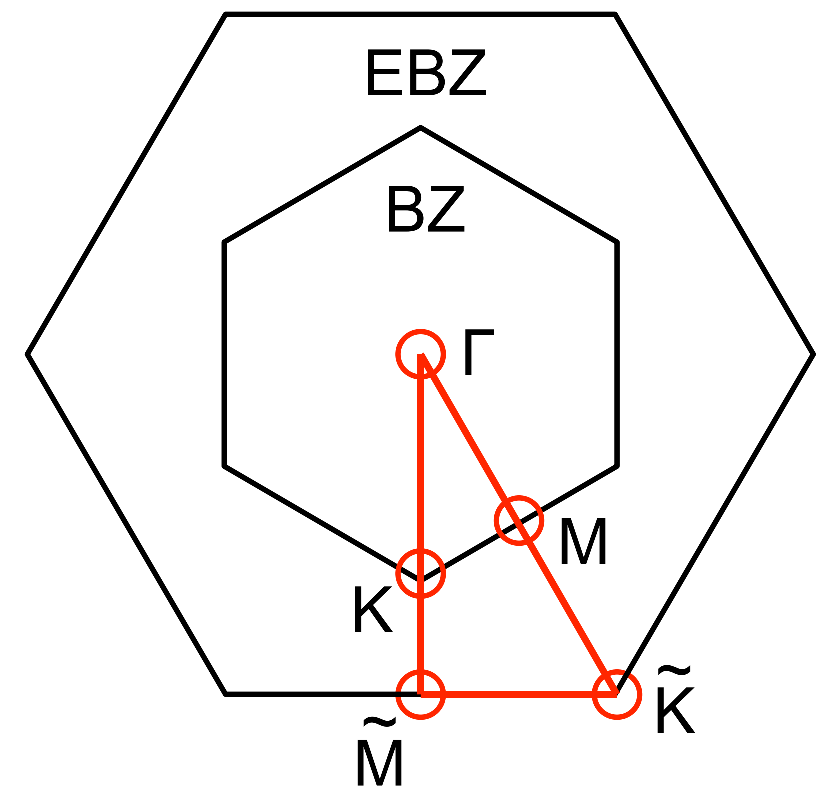
G. B. Halasz, NBP, J.van den Brink (PRL 2014)

RIXS response in SC channel



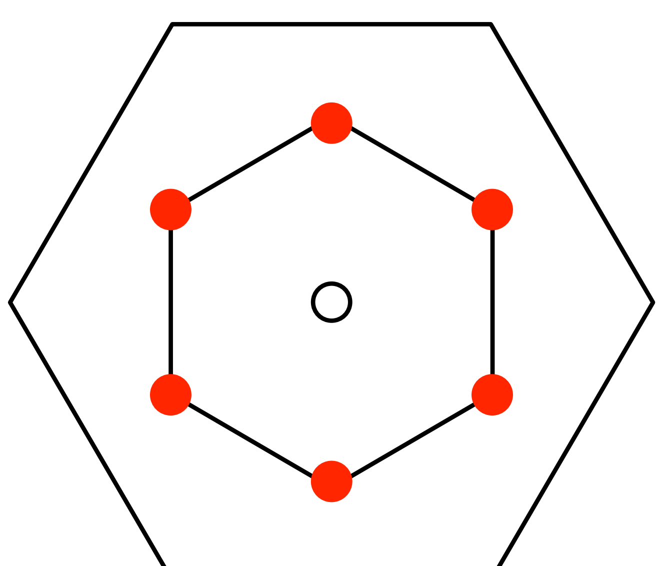
$$J_{x,y,z} = J_0$$

Reciprocal space



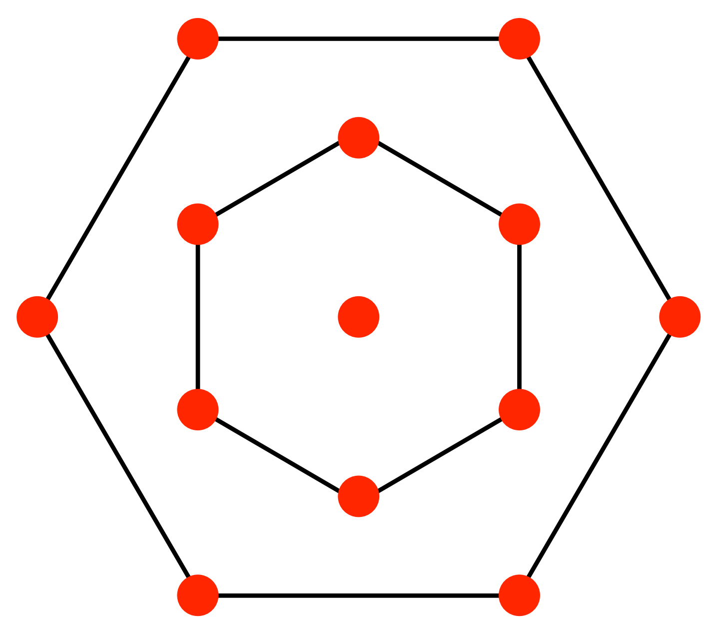
Gapless response at a finite number of discrete points

Dirac points



K points

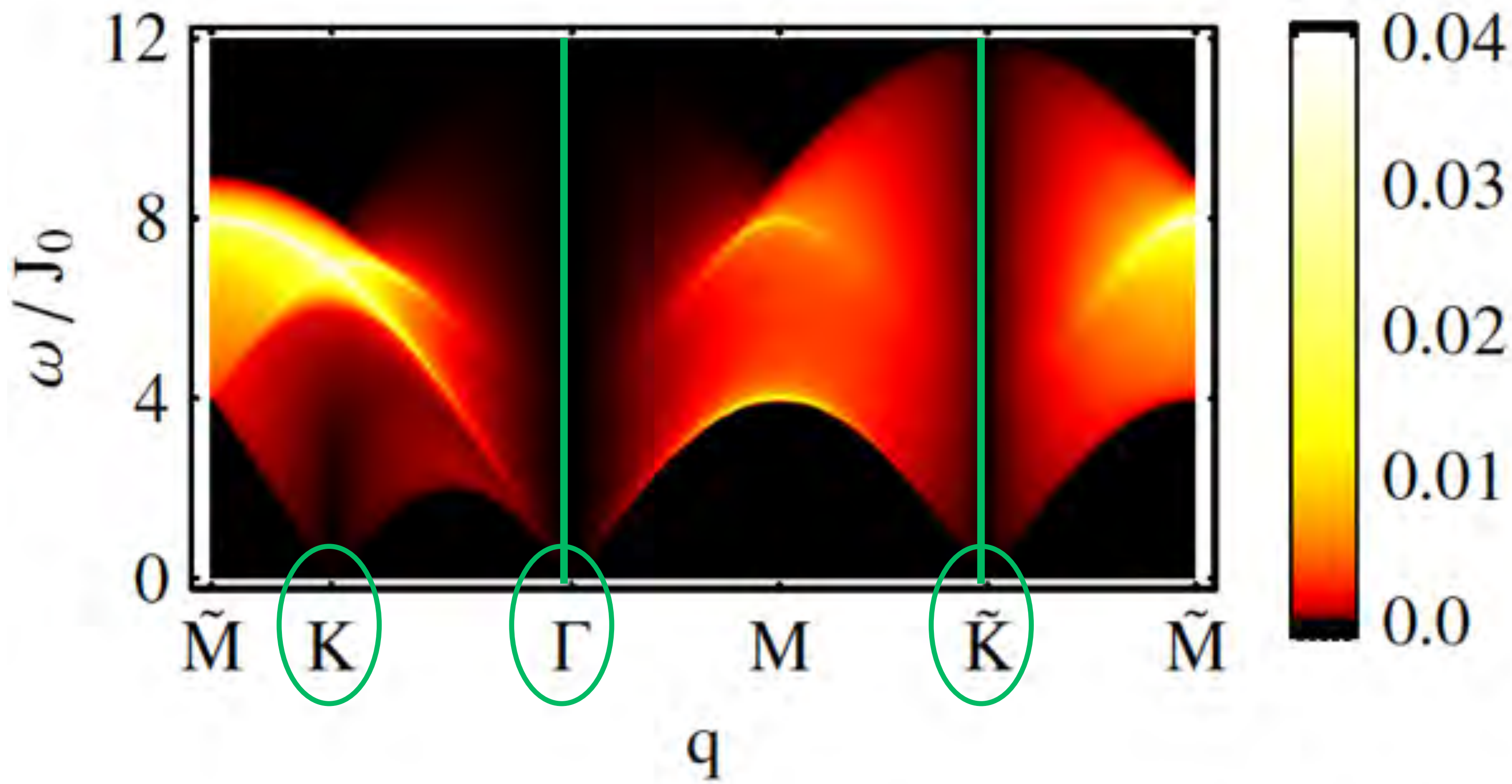
Gapless points



Γ, K, K̃ points

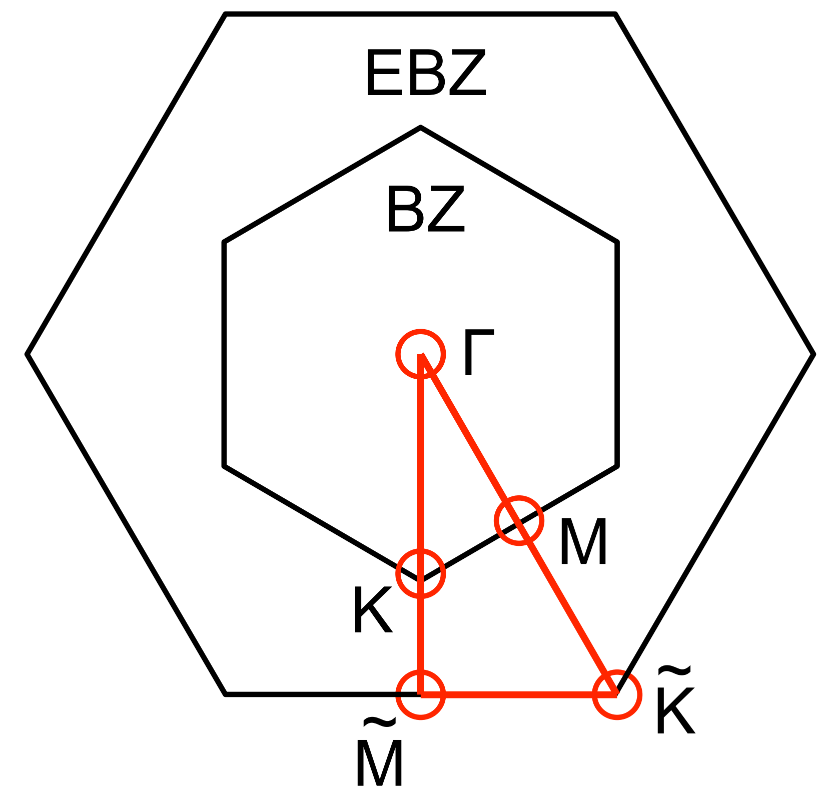
G. B. Halasz, NBP, J.van den Brink (PRL 2014)

RIXS response in SC channel



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

Reciprocal space

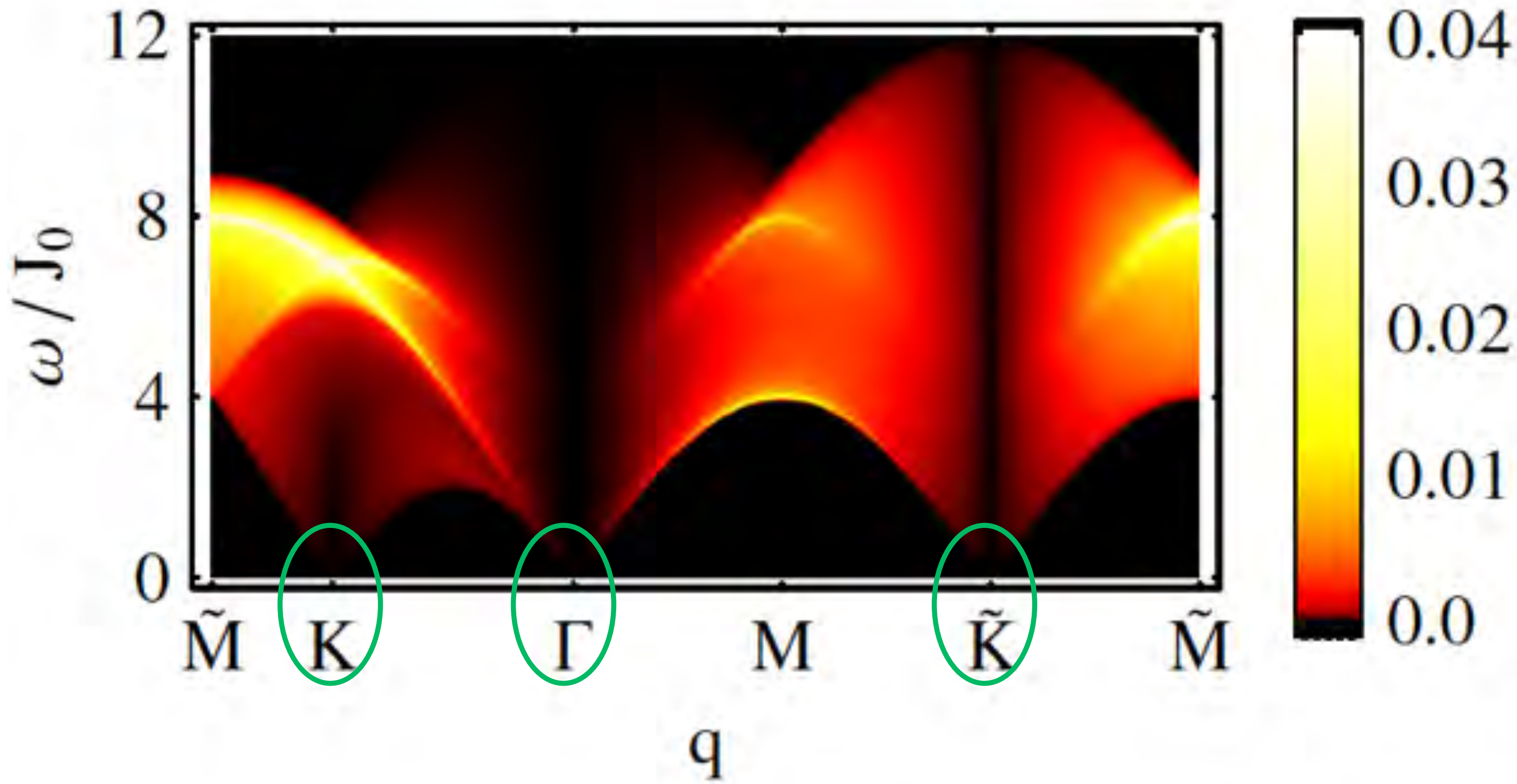


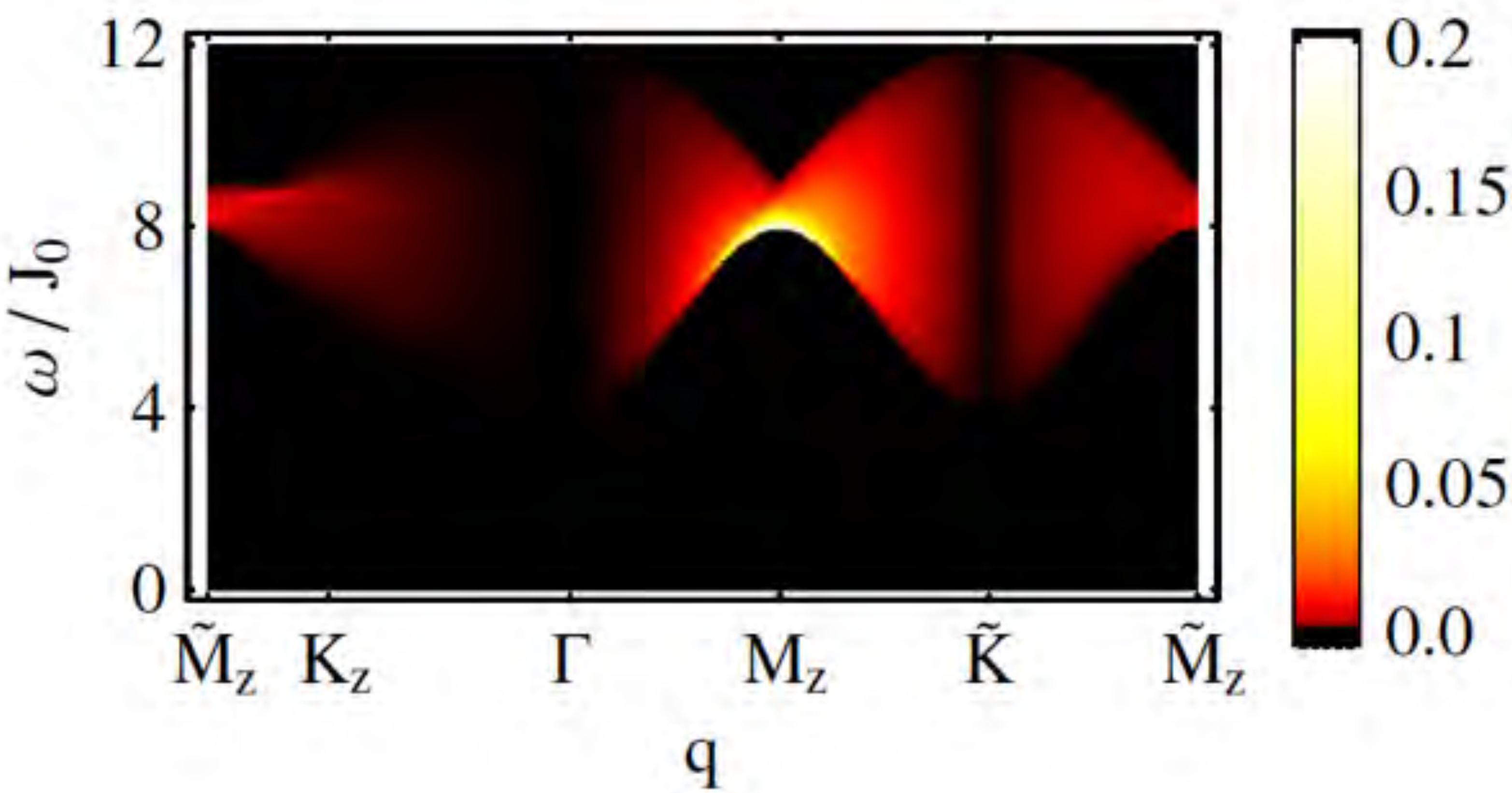
the response actually vanishes at the Γ and $\tilde{K}$ points due to the factor

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

G. B. Halasz, NBP, J.van den Brink (PRL 2014)

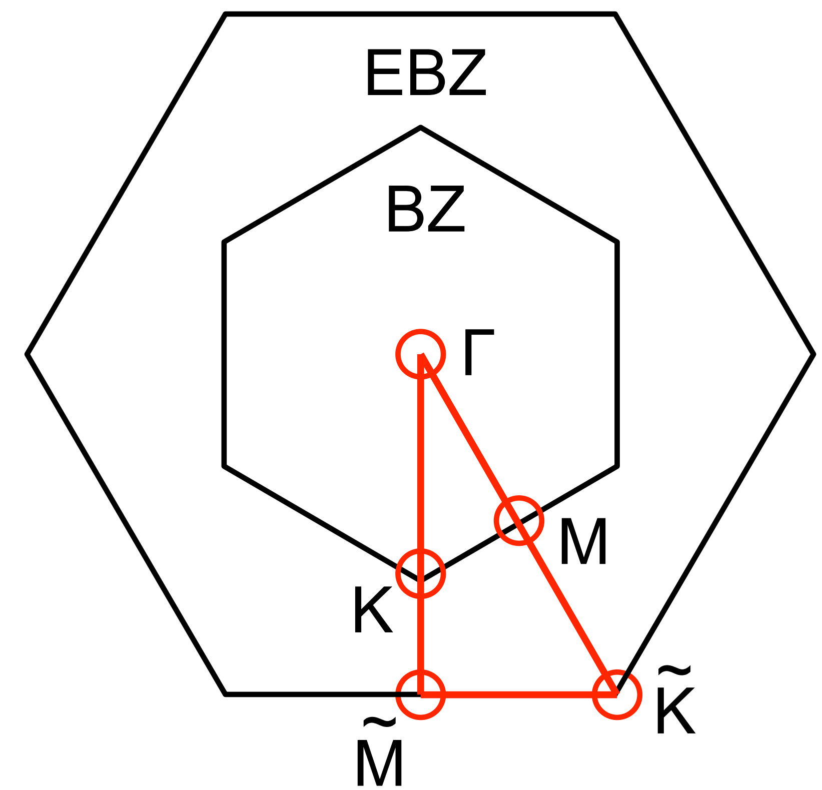
RIXS response in SC channel





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

Reciprocal space



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

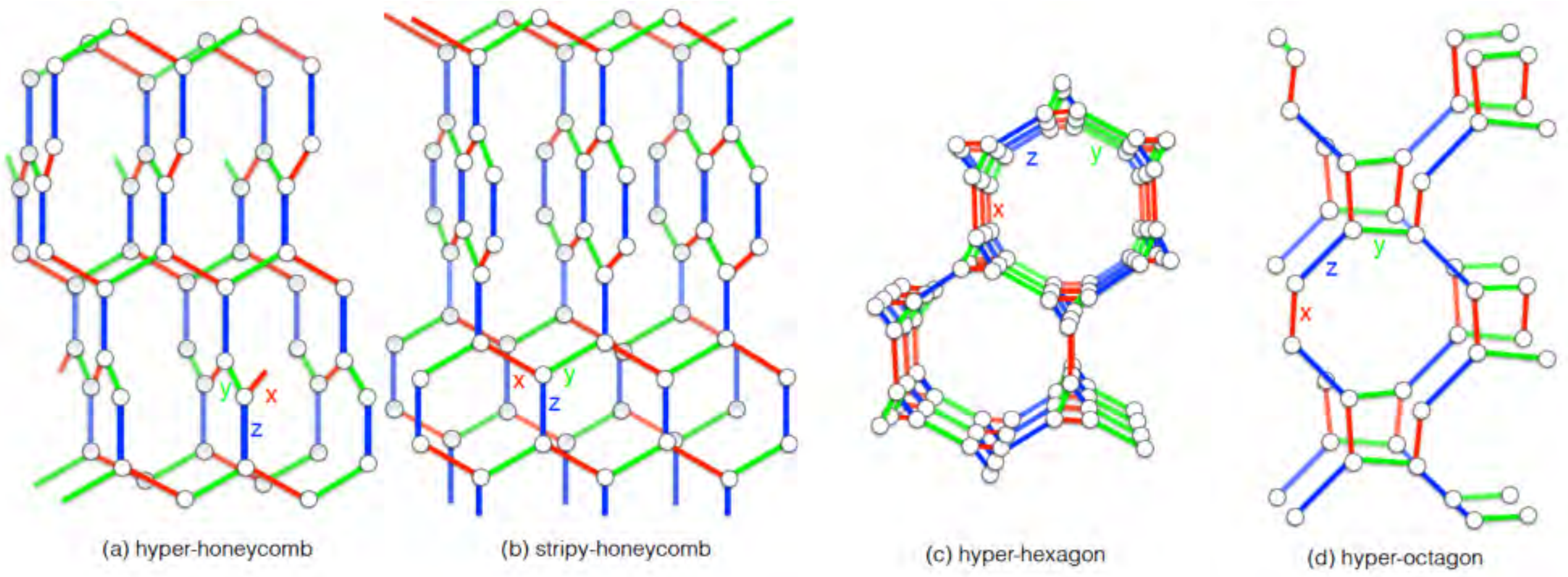
$$J_z = 2J_0$$

G. B. Halasz, NBP, J.van den Brink (PRL 2014)

Results: SC channel in3D 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.

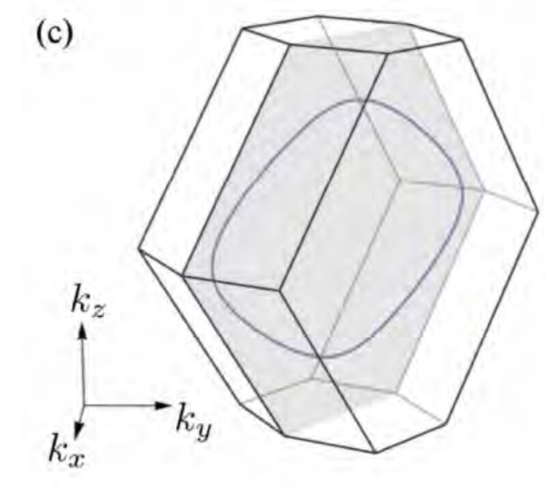


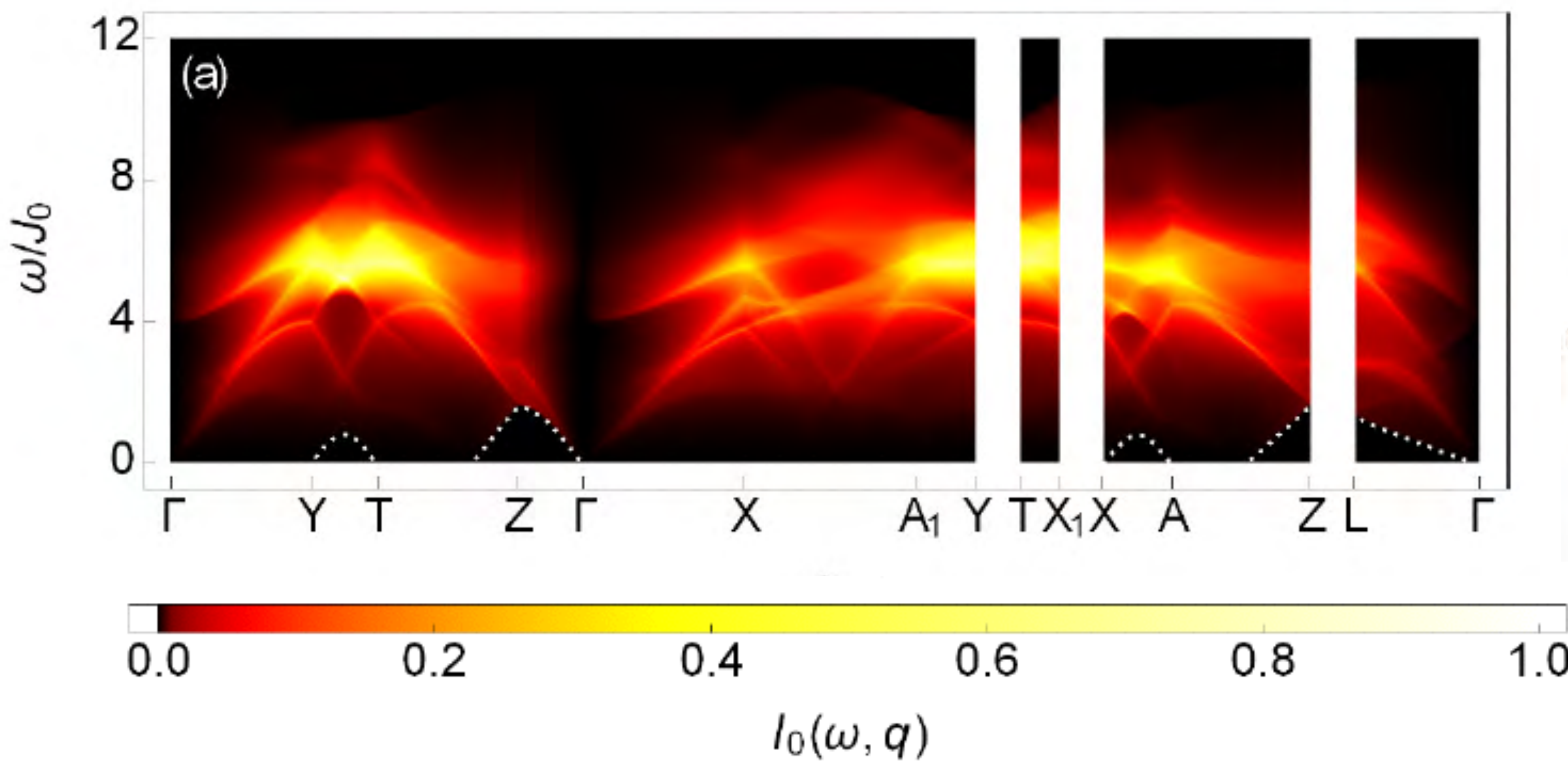
closed line of Dirac nodes

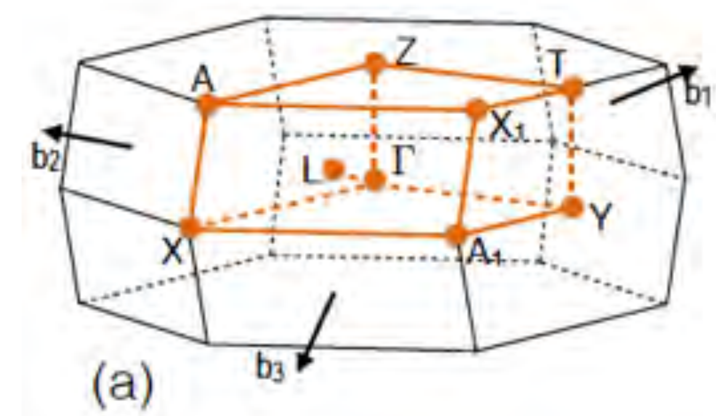
gapless Weyl points

Fermi surfaces

Hyperhoneycomb lattice





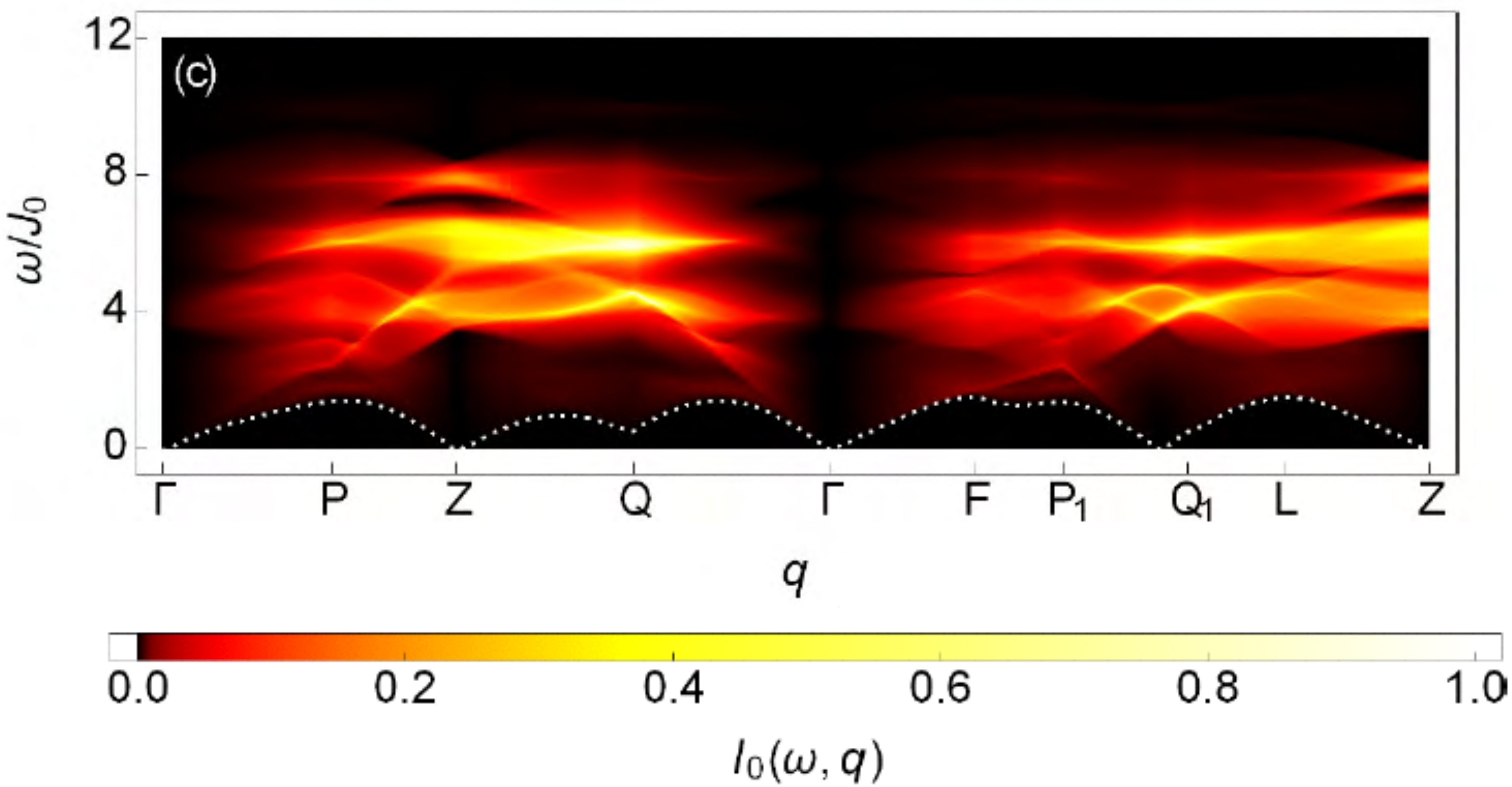


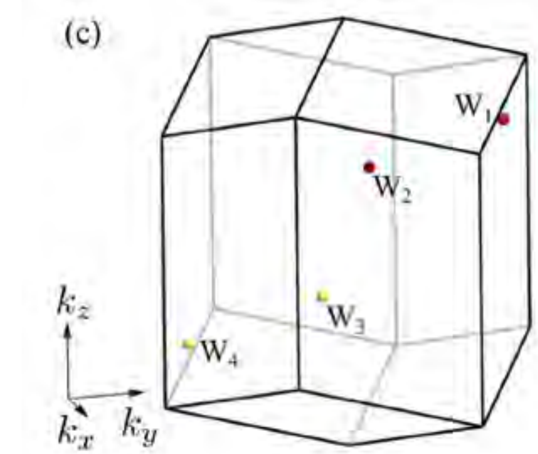
The Majorana fermions are gapless along a nodal line within the Γ-X-Y plane.

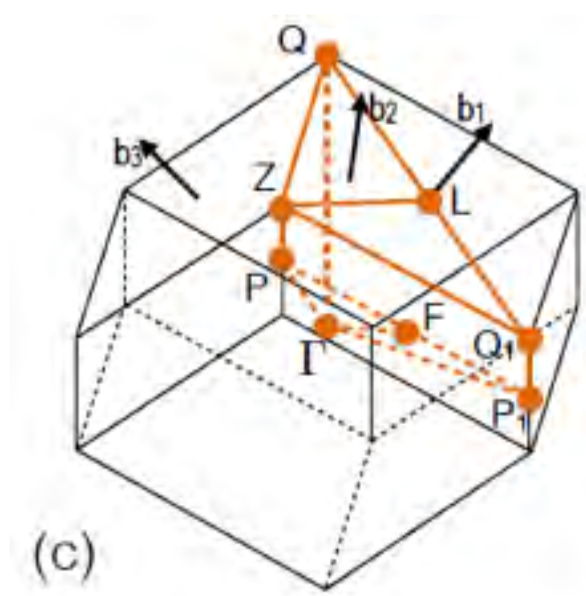
The response is thus gapless in most of the Γ-X-Y plane and also in most of the Z-A-T plane. However, it is still gapped at a generic point of the BZ.

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

Hyperhexagon lattice





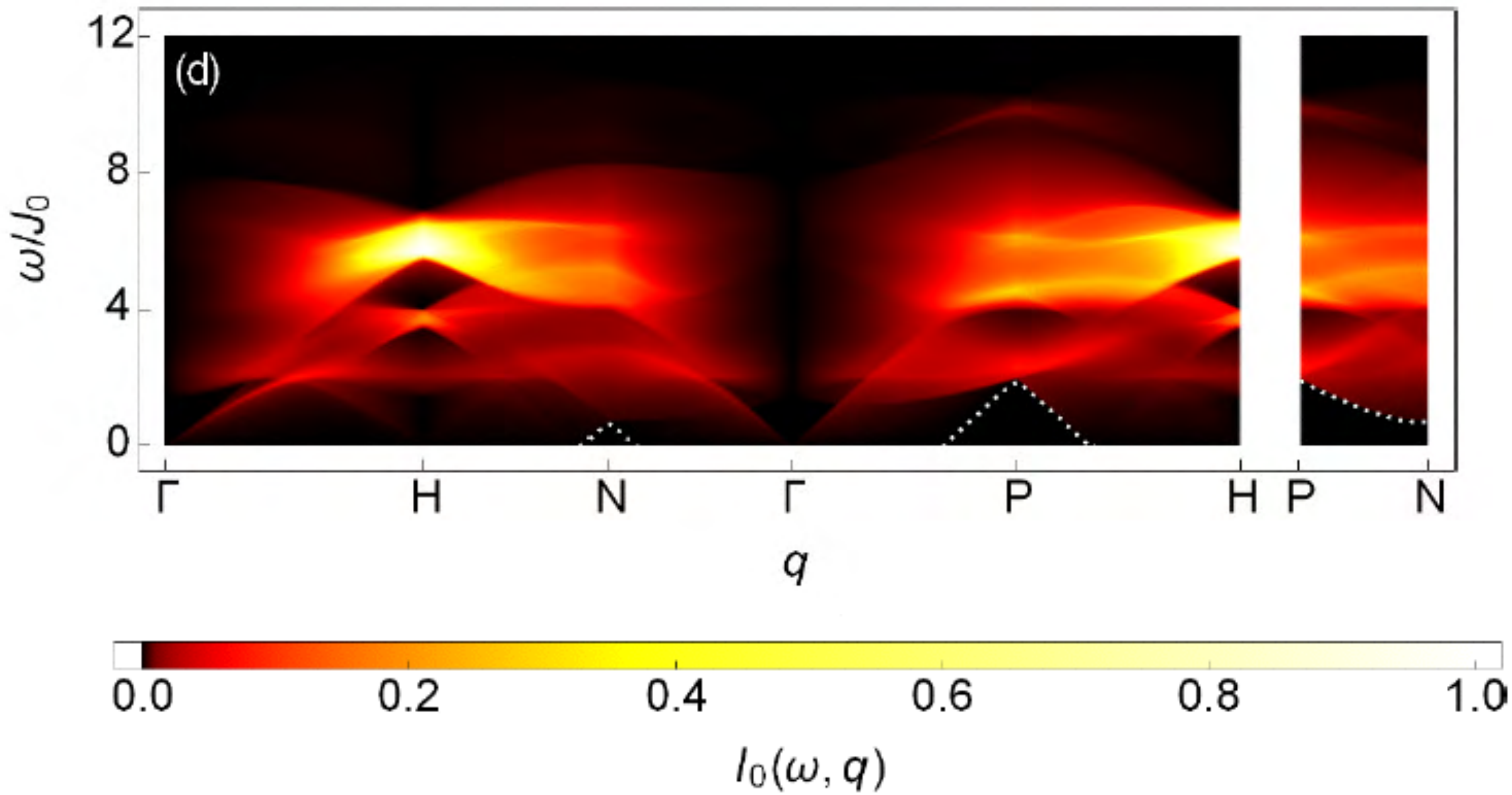


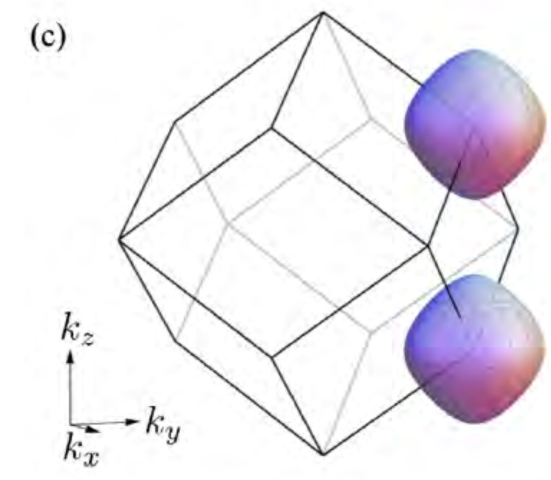
The fermions are gapless at Weyl points.

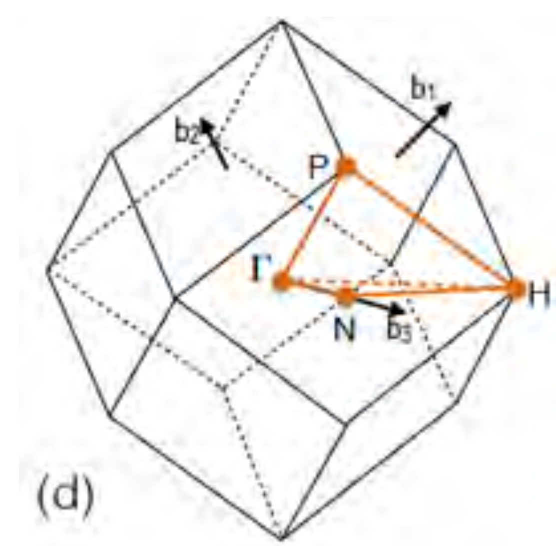
The response is thus only gapless at particular points of the BZ.

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)

Generic Kitaev spin liquids

Time-reversal-symmetric perturbations w.r.t. H_K

[Song, You, Balents, PRL 2016]

High-energy response is robust against perturbations, even beyond the phase transition into an ordered phase.

Low-energy response of a generic KSL can be completely different from that of a pure Kitaev model.

NSC channels (i.e., INS response): Gap disappears

SC channel: Two-fermion response

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

Robust

Survives only for perturbations with three-fold rotation symmetry

Robust

Thank you