# Raman Circular Dichroism of Chiral Quantum Spin Liquids

*SPICE conference "Quantum Geometry and Transport of Collective Excitations in (Non-)Magnetic Insulators" 2025, May 6-8 2025, Ingelheim*

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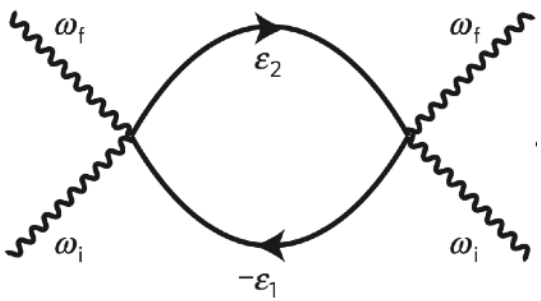
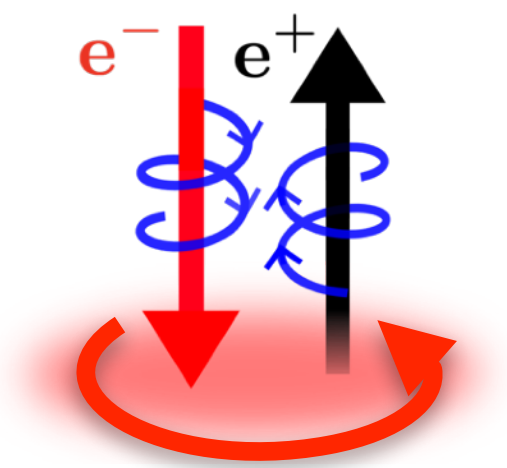


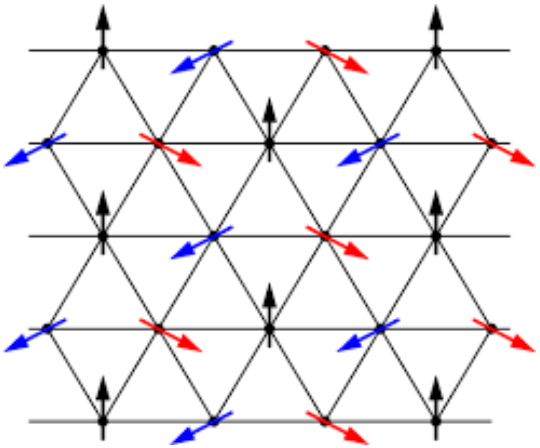

arXiv:2503.14091



# Triangular lattice magnets

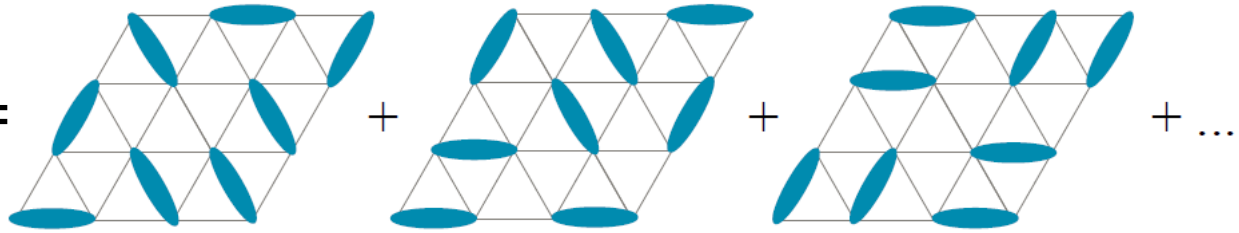
- **Geometric frustration**

$$H = J_1 \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} \vec{S}_i \cdot \vec{S}_j$$



- **Quantum fluctuations: long-range entangled QSLs**

RVB: $\Psi =$



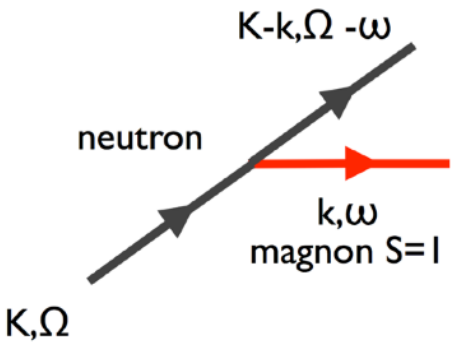
$$= \frac{1}{\sqrt{2}} \left( |\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right)$$

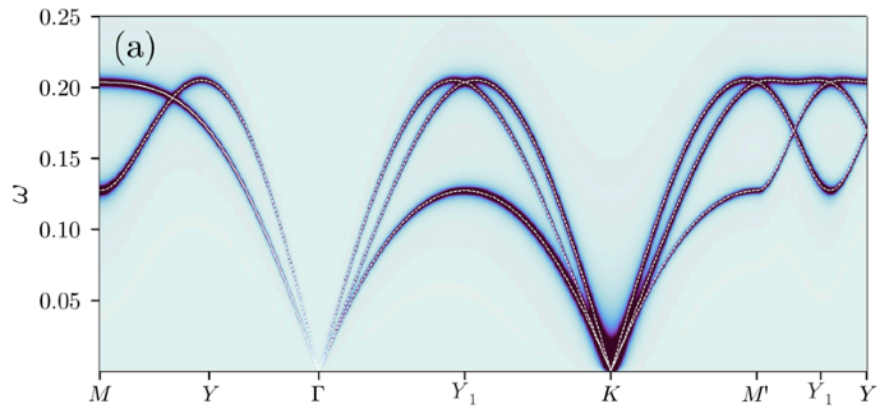
Anderson 1973

- **Fractional excitations with Anyonic statistics** Wen 1990

# How to identify QSLs?

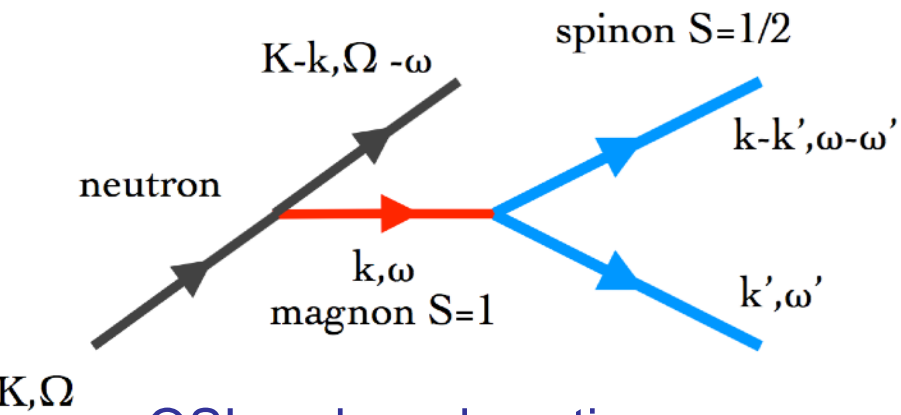
- **Inelastic neutron scattering**

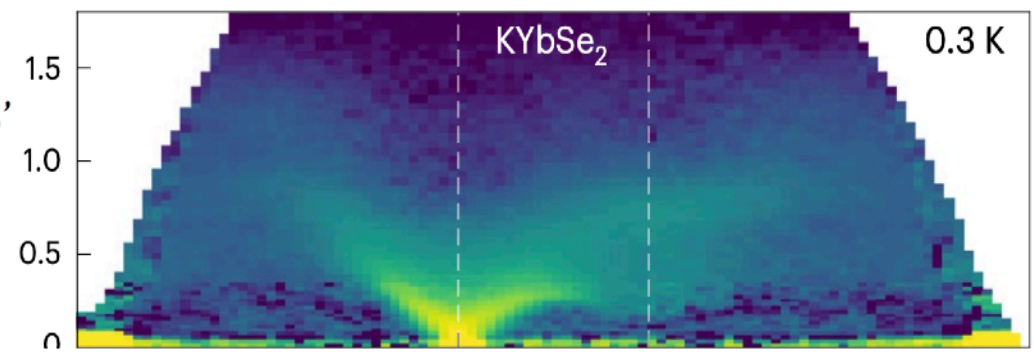



LRO → sharp dispersion

Willsher, Jin, Knolle PRB 2024



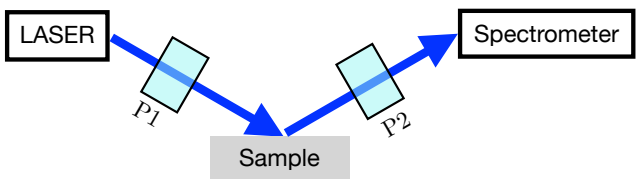



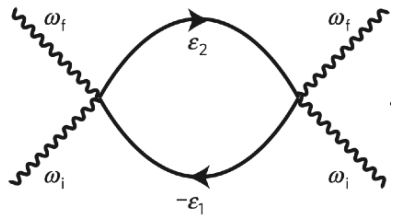

QSL → broad continuum

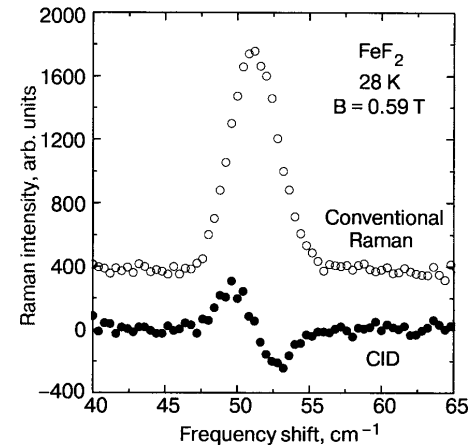
Scheie et al., Nature Physics 2024

# How to identify QSLs?

- **Raman scattering**



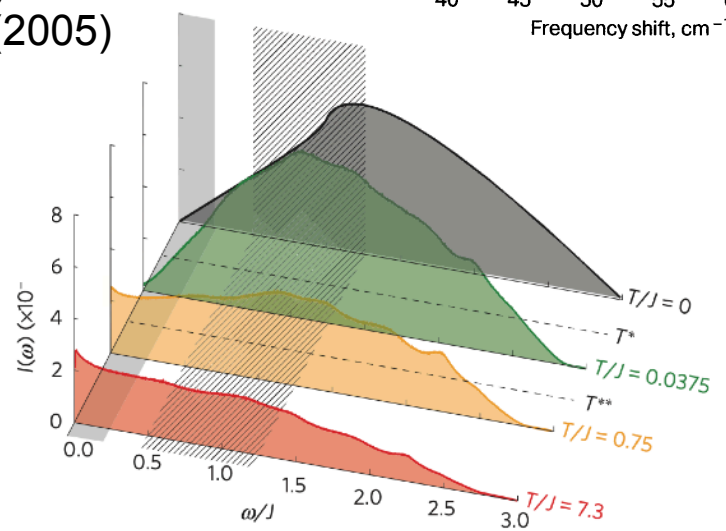




Devereaux, Hackl, Rev Mod. Phys. (2007)
Hoffman et al., Low Temp. Phys. 31, 786 (2005)

- **Probe of QSL excitations**

→ measure spinon DOS

→ thermal broadening



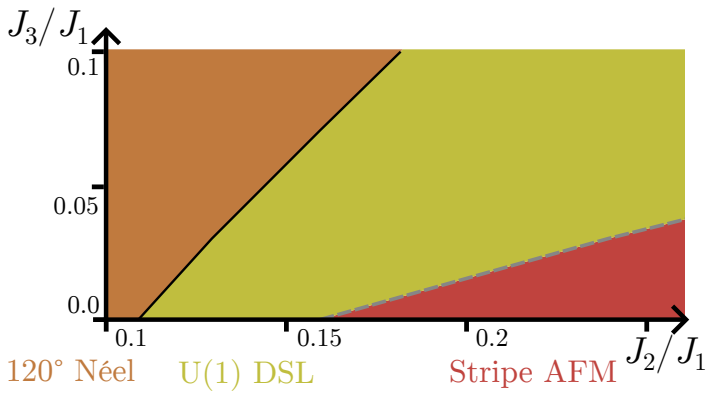

Knolle, G.-W. Chern et al. 2014
Nasu, Knolle et al., Nature Physics 2016

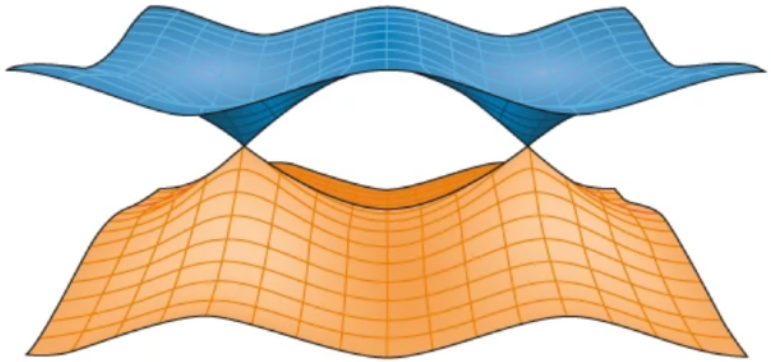
**Question:**

**Is there a more direct probe of the spinon topology?**

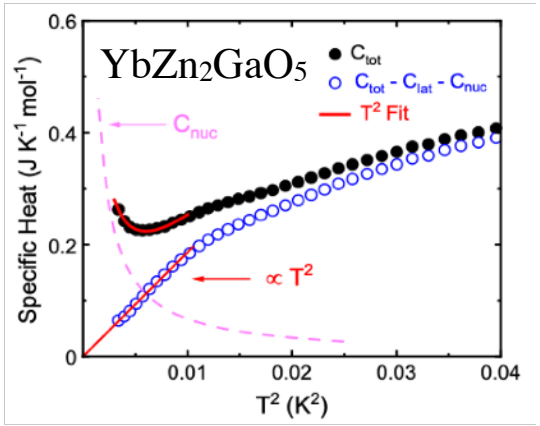
# Triangular lattice QSLs

- **A stable critical spin-disordered phase**

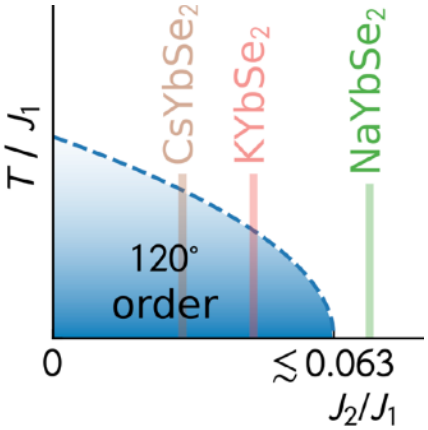


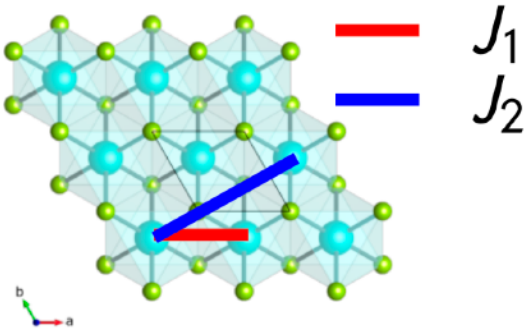



- **New candidate materials**




Bag, Xu, …, Haravifard PRL 2024




Scheie, …, Tennant, arXiv:2406.17773

# Effective Description I

- **O$^{th}$ order parton mean-field theory** Xiao-Gang Wen PRB 2002

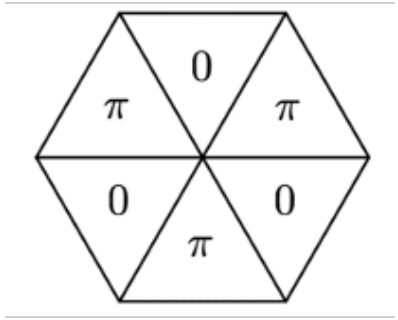
spin to fermion mapping $\rightarrow$ $\vec{S}_i = \frac{1}{2} f^\dagger_{i,\alpha} \vec{\sigma}_{\alpha\beta} f_{i,\beta}$

$$\mathcal{H}_{J_1 J_2} = \sum_{\langle ij \rangle} J_{ij}\, \vec{S}_i \cdot \vec{S}_j = -\frac{1}{2} \sum_{ij} J_{ij} \left( f^\dagger_{i\alpha} f_{j\alpha} f^\dagger_{j\beta} f_{i\beta} + \frac{1}{2} f^\dagger_{i\alpha} f_{i\alpha} f^\dagger_{j\beta} f_{j\beta} \right)$$

bond singlet decoupling $\rightarrow$ $t_{ij} = J_{ij} \langle f^\dagger_{i\alpha} f_{j\alpha} \rangle$ with $t_{ij} = t e^{i a_{ij}}$

$$\mathcal{H}_0 = \sum_{ij} t_{ij} f^\dagger_{j\alpha} f_{i\alpha} + \text{h.c.}$$

staggered pi-flux Ansatz



$\rightarrow$ **4 Dirac cones for spinous (2 valley x 2 spin)** Iqbal et al. PRB 2016

# Effective Description II

- **1st order parton mean-field theory** Song, …, He, Nature Comm 2018

local U(1) gauge redundancy $f^\dagger_{i\alpha} \to e^{i\theta_i} f^\dagger_{i\alpha}$ & $a_{ij} \to a_{ij} - \theta_i + \theta_j$

- **Analyse stability w.r.t. gapless phase fluctuations**

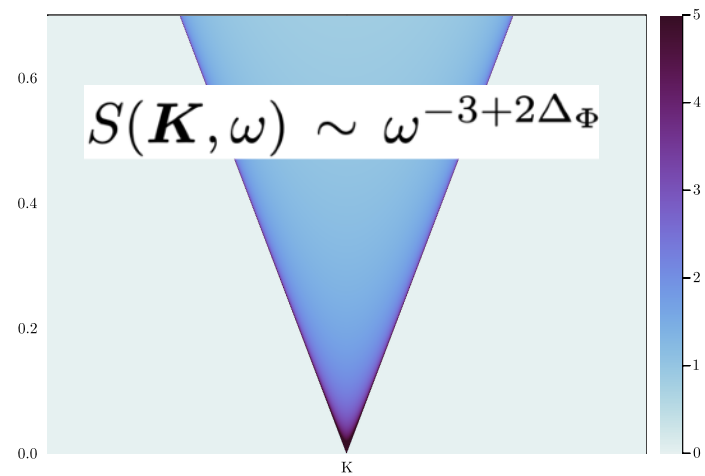
$$\mathcal{L}_{\mathrm{QED}_3} = \sum_{i=1}^{N=4} \left[-\bar{\psi}_i \gamma^\mu (\partial_\mu - \mathrm{i}a_\mu)\psi_i\right] + \frac{1}{g^2} f_{\mu\nu} f^{\mu\nu} + \ldots$$

- **Novel type of excitation**

→ **monopole** can tunnel $2\pi$ flux

→ strongly coupled CFT

→ **continuum at K-point**




→ 2+1D analog of Luttinger liquid Seifert, Willsher, … Knolle Nature Comm 2024

# Effective Description III

**Key Questions:**

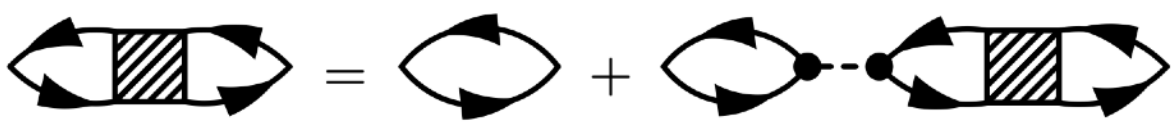
**What is the scale of universal low-energy physics?**

**What about non-universal finite frequency response?**

- **Study stability w.r.t. to gapped longitudinal fluctuations** $t_{ij} = t e^{i a_{ij}}$

$$\mathcal{H}_0 = \sum_{ij} t_{ij} f^\dagger_{j\alpha} f_{i\alpha} + \text{h.c.} - \frac{1}{2} \sum_{ij} J_{ij} \left( f^\dagger_{i\alpha} f_{j\alpha} f^\dagger_{j\beta} f_{i\beta} + \frac{1}{2} f^\dagger_{i\alpha} f_{i\alpha} f^\dagger_{j\beta} f_{j\beta} \right)$$

→ dynamical structure factor within RPA approximation

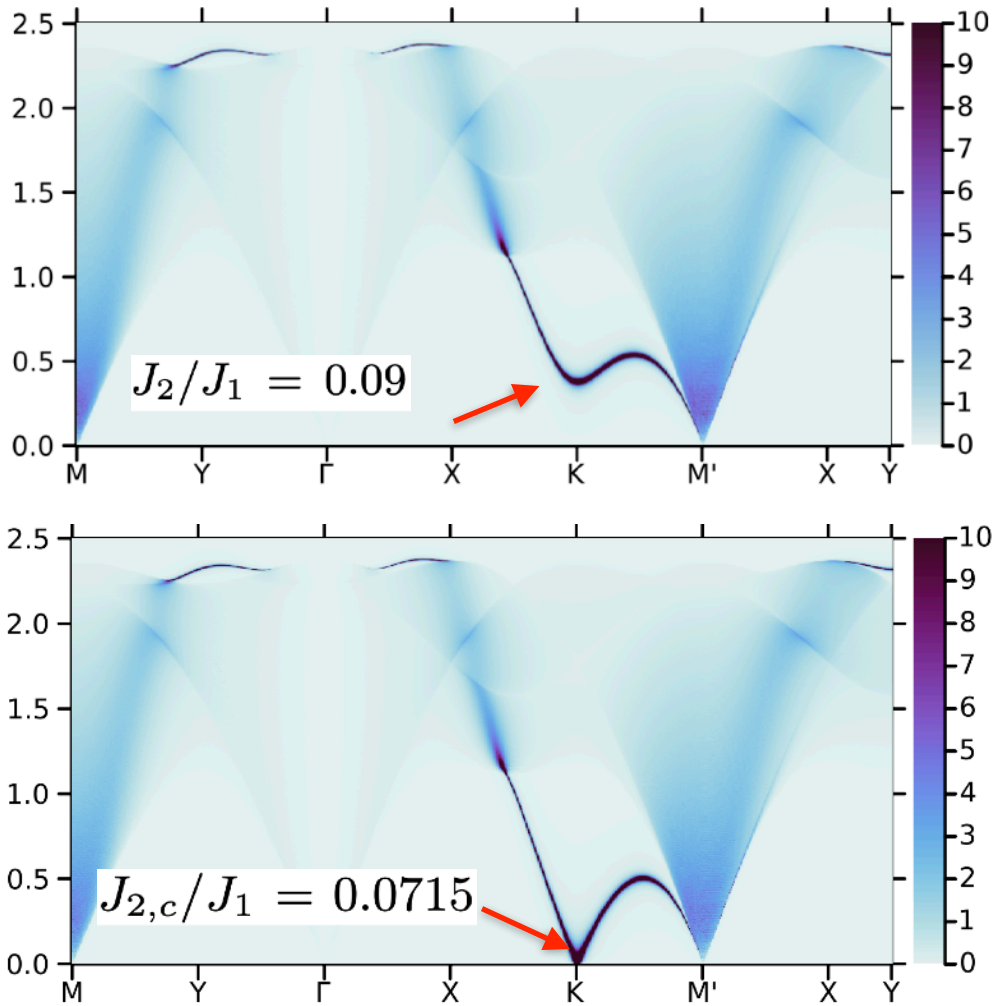


$$[\chi(\boldsymbol{q},\omega)]^{\mu\nu} = [\chi^0(\boldsymbol{q},\omega)]^{\mu\nu} + [\chi^0(\boldsymbol{q},\omega)]^{\mu\rho} J^{\rho\sigma}_{\boldsymbol{q}} [\chi(\boldsymbol{q},\omega)]^{\sigma\nu}$$

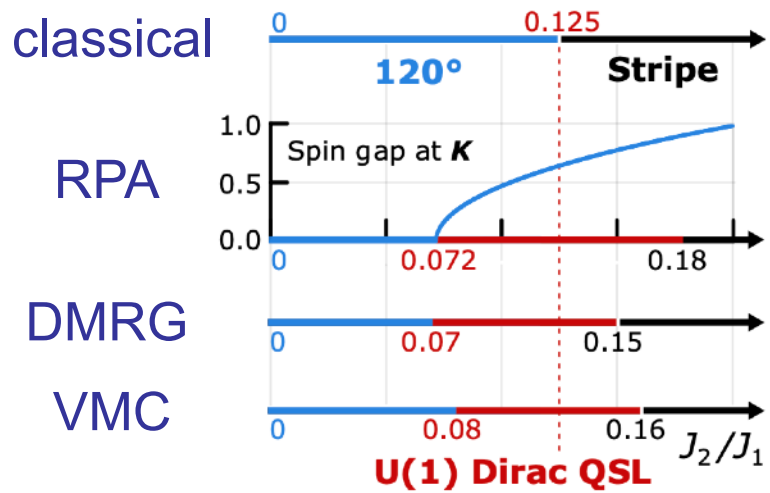
→ **no free parameters (fully self-consistent)**

# INS Response

- **Sharp 'spinon exciton' coexisting with broad continua**



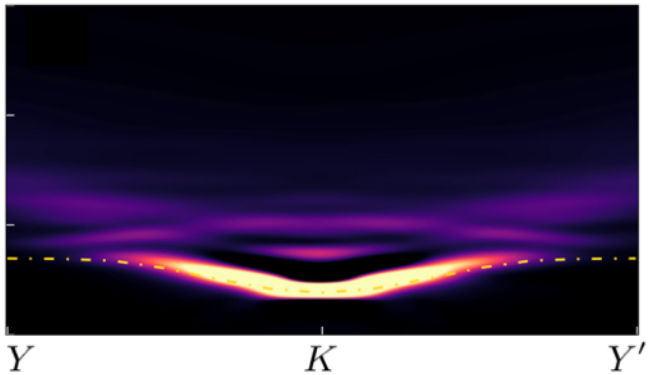

**Phase diagram**




**Almost quantitative agreement with state-of the art numerics !**

# INS Response

- **Comparison to numerical methods**

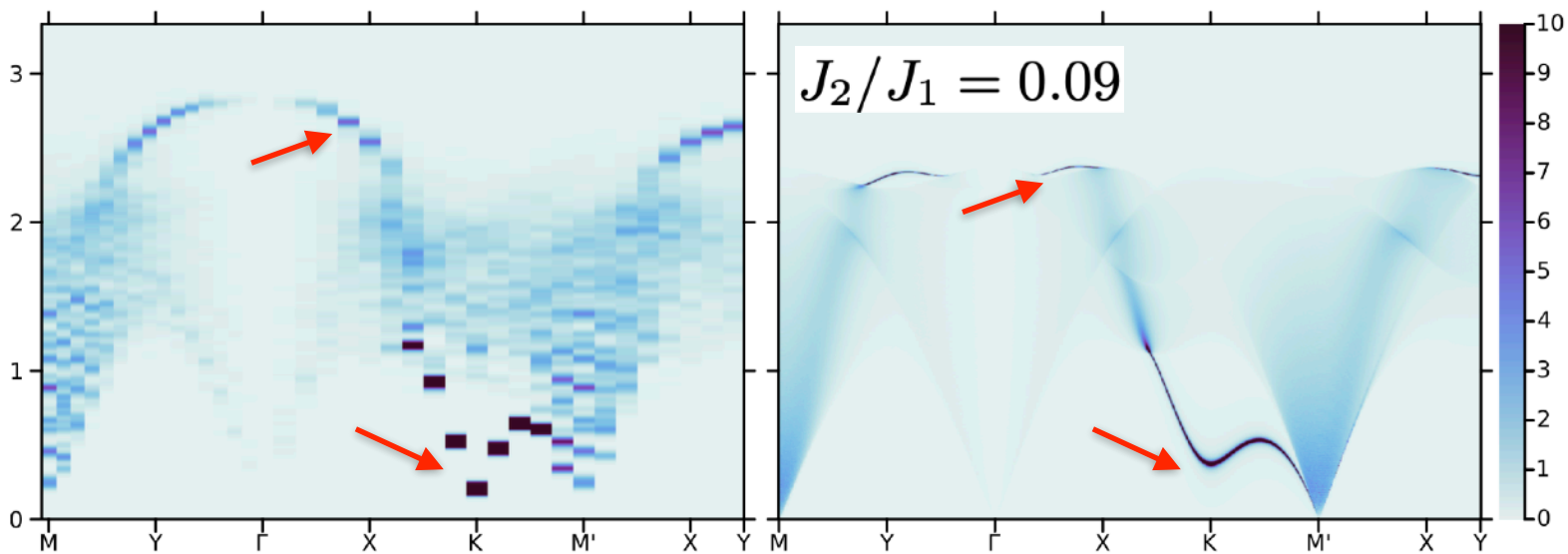



→ sharp mode at K point in tDMRG

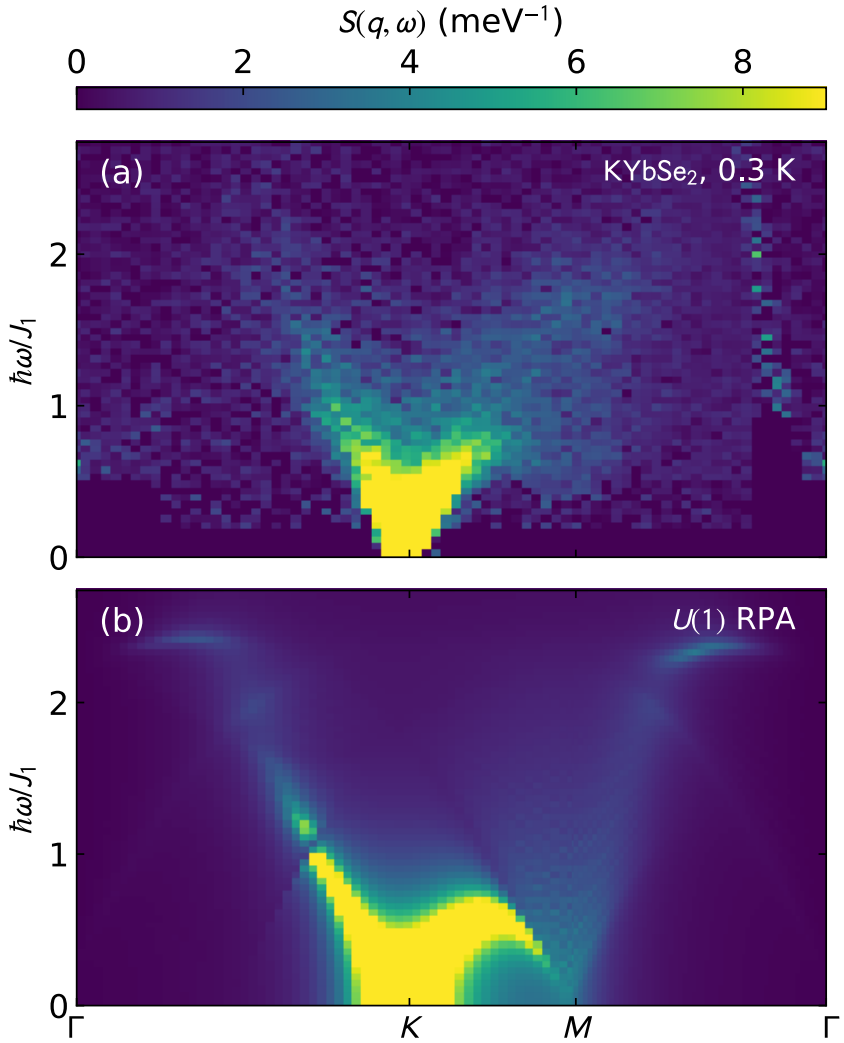
→ ‘gapped spin wave’ in QSL

Drescher, …, Pollmann, PRB 2023

- **RPA accounts almost quantitatively for VMC** Ferrari, Becca, PRX 2019



# Comparison to experiment




Scheie et al., Nature Physics 2024

[Thanks to Allen Scheie!]

→ **Minimum at M point but is it gapless?**

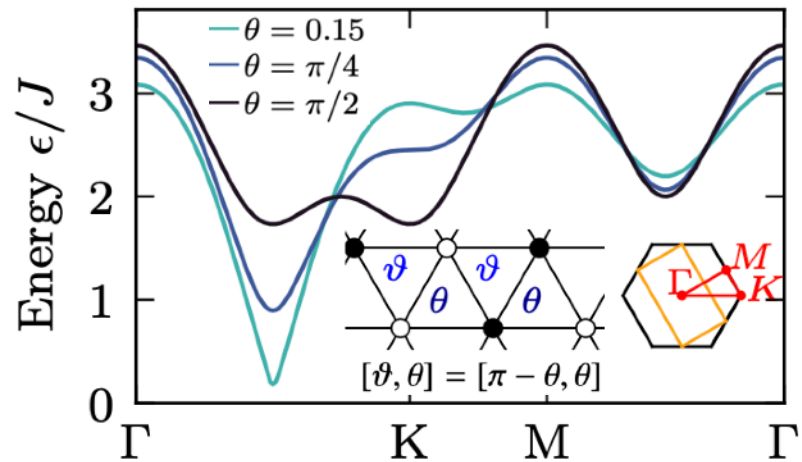
# Chiral QSL

- **TRS breaking gaps Dirac cones**

$$\mathcal{H}_{J_1 J_2} + J_\chi \sum_{ijk \in \triangle, \triangledown} \vec{S}_i \cdot (\vec{S}_j \times \vec{S}_k)$$

→ Chern bands akin to FQHE

Kalmeyer, Laughlin PRL 1987
Comment: Kivelson & Rokhsar

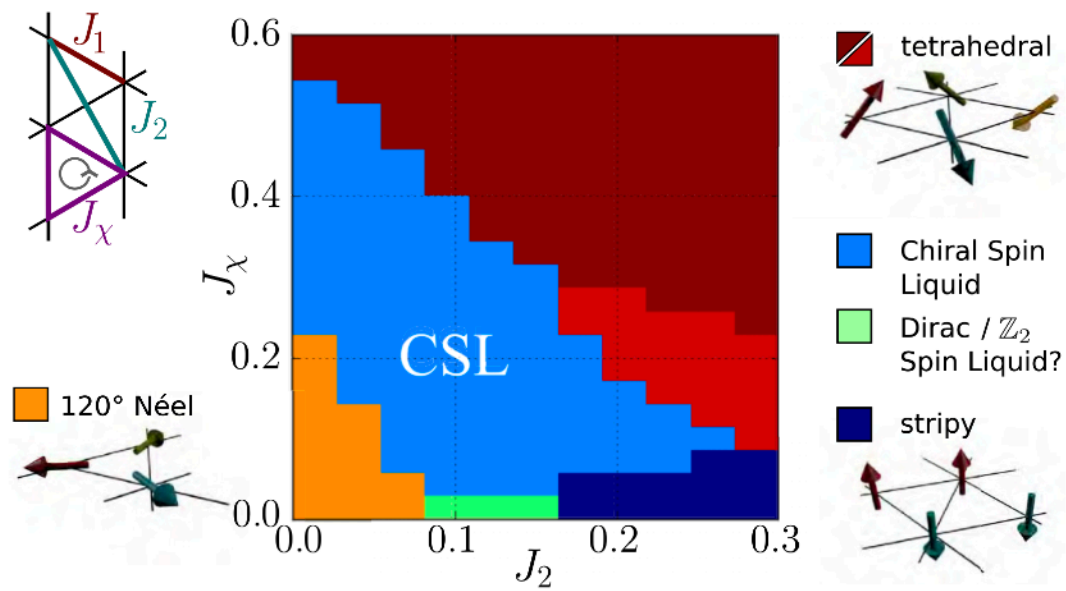



- **Phase diagram**

→ stable CSL phase

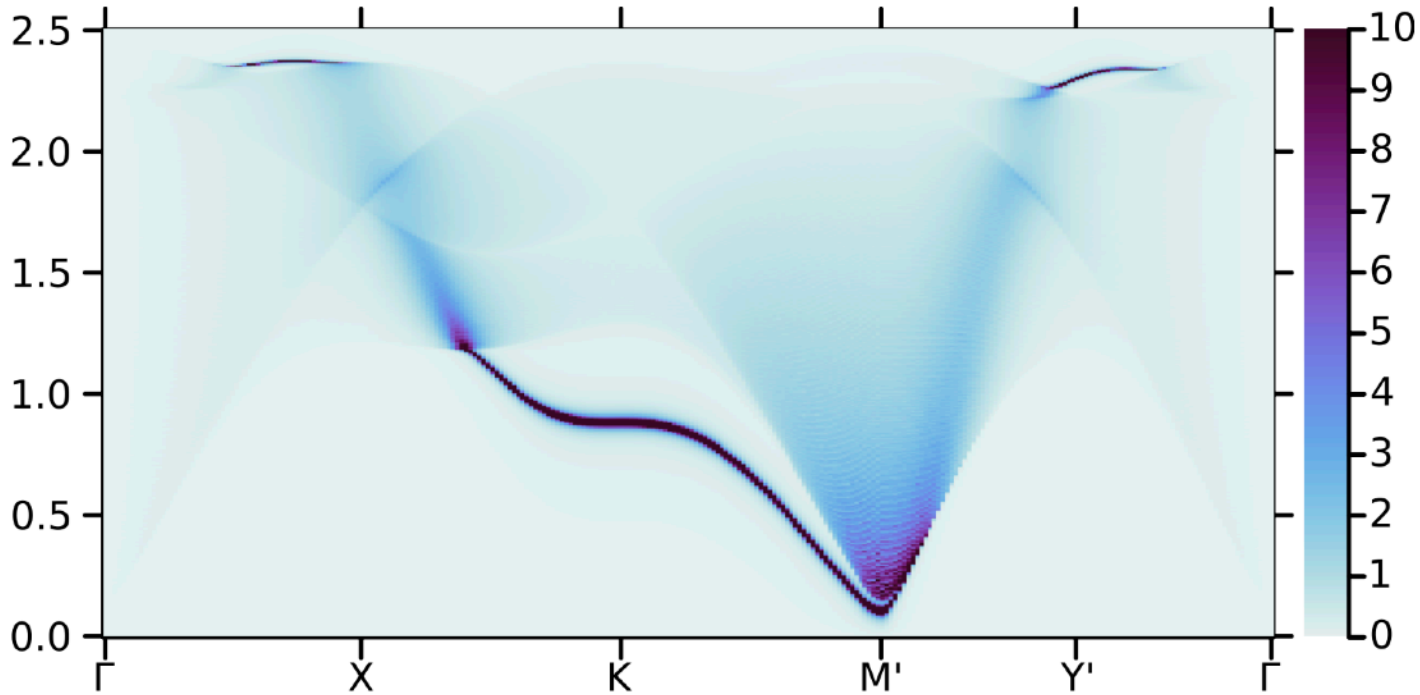
→ non-coplanar orders

Wietek, Läuchli PRB 2017



# Chiral QSL

- **Sharp mode at M point within self-consistent RPA scheme**




→ Condensation at M leads to tetrahedral orders

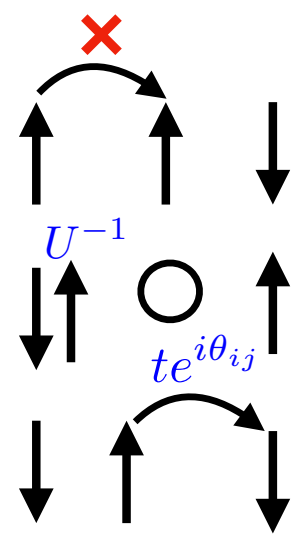
# Raman scattering

- Coupling of EM field to charged electrons:

$$h(\boldsymbol{k}+e\boldsymbol{A}) = h(\boldsymbol{k}) + e\sum_{\mu} l^{(1)}_{\mu}(\boldsymbol{k})A_{\mu} + \frac{e^2}{2}\sum_{\mu,\nu} l^{(2)}_{\mu\nu}(\boldsymbol{k})A_{\mu}A_{\nu} + \ldots$$

→ Raman vertex: $R^{ss'}_{\mathrm{LMC}} = \sum_{\mu,\nu}\sum_{m,n} e^{s}_{\mu} e^{*s'}_{\nu} \sum_{\boldsymbol{k}} \psi^{\dagger}_{m}(\boldsymbol{k}) l^{(2)}_{mn,\mu\nu}(\boldsymbol{k}) \psi_{n}(\boldsymbol{k})$

with LMC: $l^{(2)}_{mn,\mu\nu}(\boldsymbol{k}) = \partial_{k_\mu}\partial_{k_\nu} h_{mn}(\boldsymbol{k})$




- **Leudon-Fleury mechanism in Mott insulators**

$$R^{ss'}_{\mathrm{LF}} = \sum_{i,j} (\boldsymbol{e}^{s}\cdot\boldsymbol{d}_{ij})(\boldsymbol{e}^{*s'}\cdot\boldsymbol{d}_{ij}) \sum_{\alpha,\beta} S^{\alpha}_{i} J^{\alpha\beta}_{ij} S^{\beta}_{j}$$

→ Raman vertex as light-induced exchange

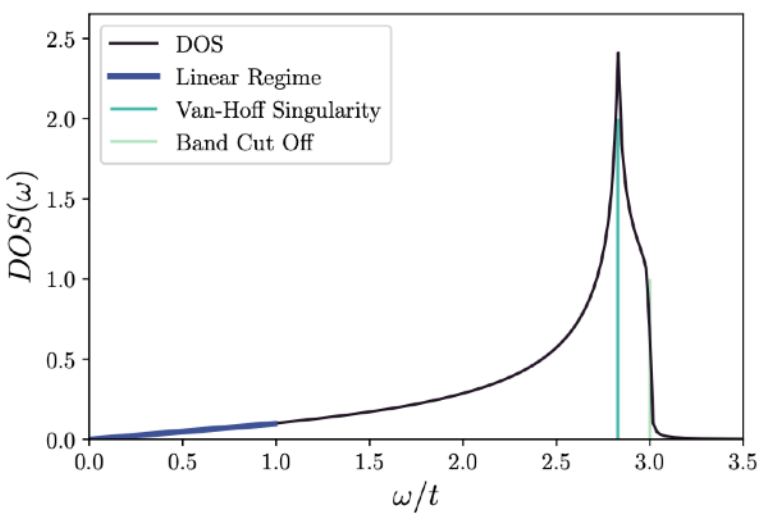
Fleury, Leudon PR 1968

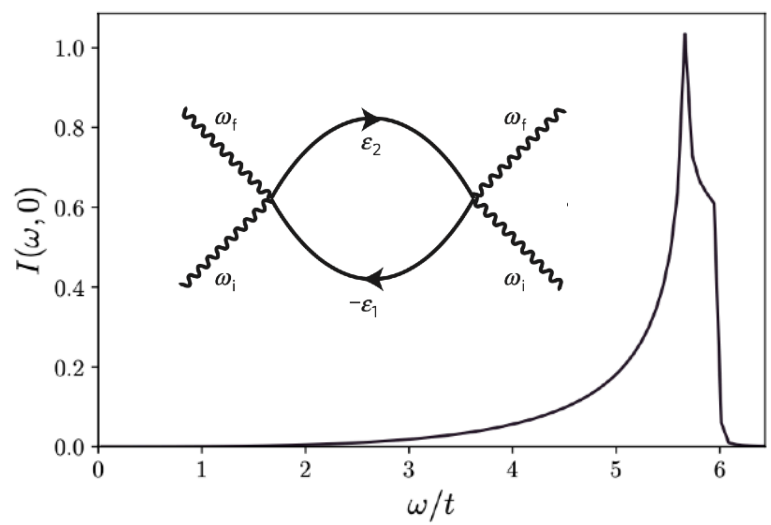
# Raman scattering

- Raman response from basic Kubo formula

$$I(\omega) = \int dt e^{i\omega t} \langle \mathcal{T}_t R^\dagger(t) R(0) \rangle$$

- **Raman scattering in QSL probes spinon DOS**




**What about topology of spinon bands?**

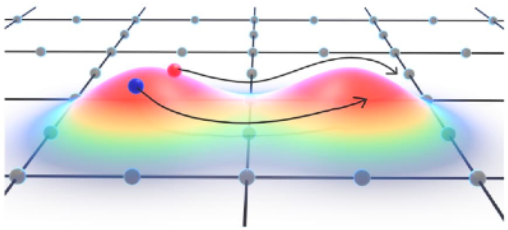
# Quantum Geometry

- **Quantum Geometric Tensor** of single particle excitations

$$Q_{\mu\nu} = \langle \partial_\mu n | 1 - \mathcal{P}_n | \partial_\nu n \rangle = \sum_{n \neq m} \langle \partial_\mu n | m \rangle \langle m | \partial_\nu n \rangle$$

$$Q_{\mu\nu} = g_{\mu\nu} - \frac{i}{2} F_{\mu\nu}$$

Quantum geometry  Berry curvature



→ **determines light-matter coupling in electronic systems**

Topp, Eckhardt, Kennes, Sentef, Törmä, PRB 104, 064306 (2021)

Review: Yu, Bernevig, Queiroz, Rossi, Törmä, Yung arXiv:2501.00098

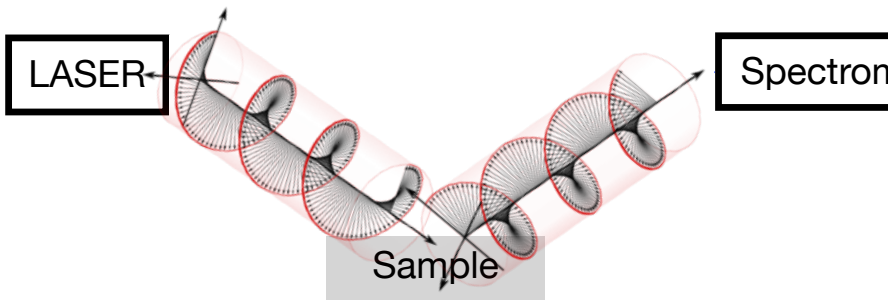
- **For quadratic spinon (and magnon) Hamiltonians:**

$$\mathcal{R}^{ss'}_{mn} = L^{(2)}_{\mu\nu,mn} \epsilon^s_\mu \bar{\epsilon}^{s'}_\nu$$

Boström, Parvini, McIver, Rubio, Kusminskiy, Sentef, PRL 130 (2023)

# Circular Dichroism

- **Probes TRS symmetry breaking**

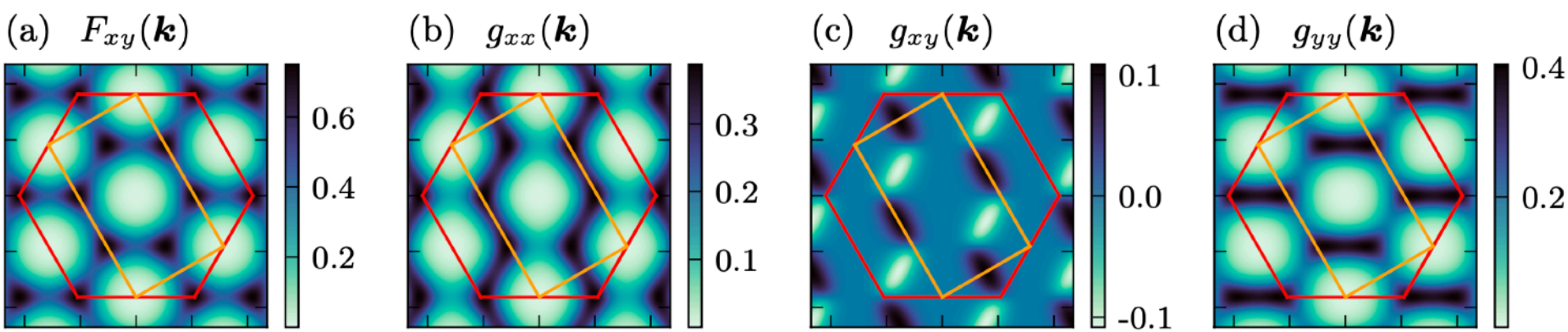



$$I_{\mathrm{RCD}}(\omega) = I^{+-}(\omega) - I^{-+}(\omega)$$

$$I_{\mathrm{RCD}}(\omega) = \sum_{\boldsymbol{k}} 2\mathrm{Im}\left[\left(l^{(2)}_{12,xx}(\boldsymbol{k}) - l^{(2)}_{12,yy}(\boldsymbol{k})\right) l^{*(2)}_{12,xy}(\boldsymbol{k})\right] \delta\left(\omega - 2\epsilon(\boldsymbol{k})\right)$$

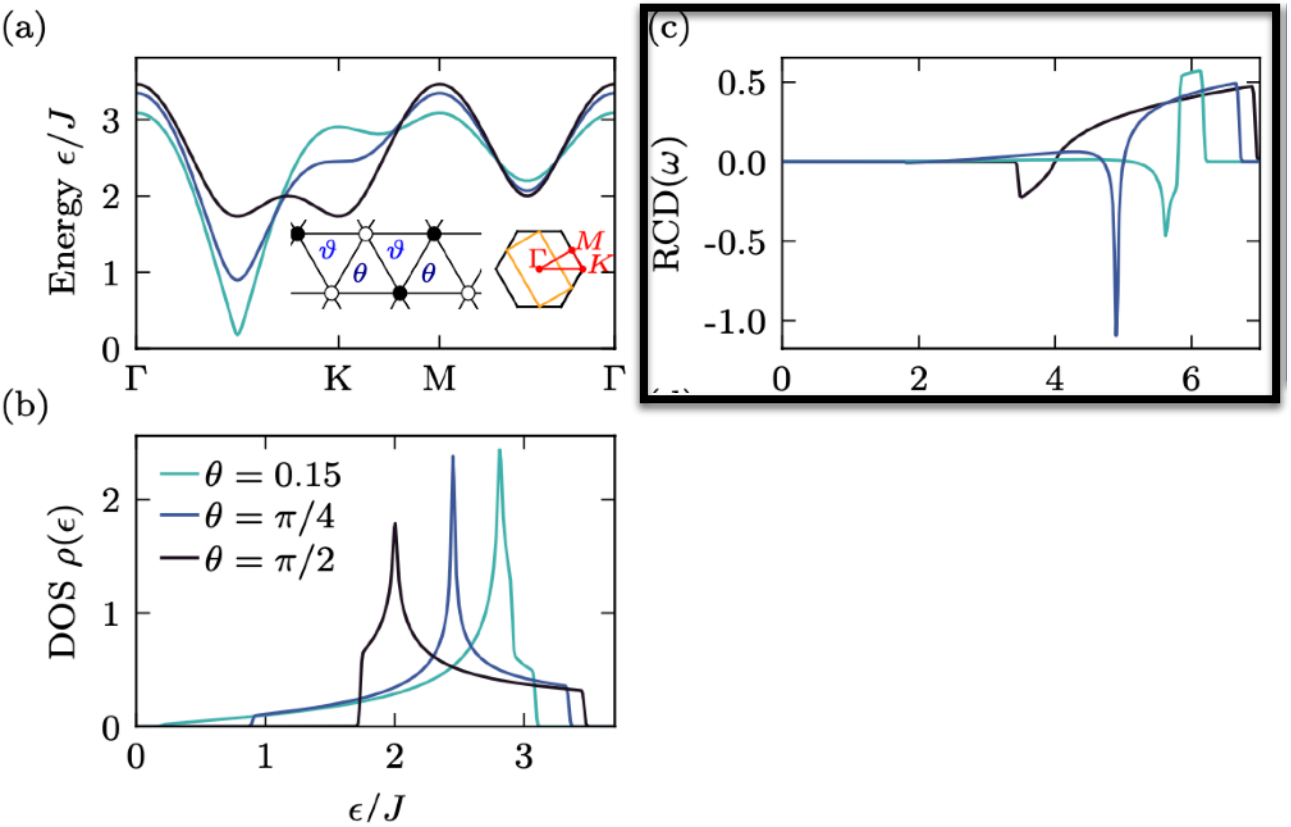
- **RCD probes quantum geometry of spinons**

$$I_{\mathrm{RCD}}(\omega) = \sum_{\boldsymbol{k}} \left[\boxed{h_{\mathrm{B}}(\boldsymbol{k}) F_{xy}(\boldsymbol{k})} + \boxed{h_{\mathrm{G}}(\boldsymbol{k})\left(g_{xx}(\boldsymbol{k}) + g_{yy}(\boldsymbol{k})\right)} + h_{\mathrm{S}}(\boldsymbol{k})\right] \delta\left(\omega - 2\epsilon(\boldsymbol{k})\right)$$



# Circular Dichroism

$$I_{\mathrm{RCD}}(\omega) = \sum_{\boldsymbol{k}} \left[ h_{\mathrm{B}}(\boldsymbol{k}) F_{xy}(\boldsymbol{k}) + h_{\mathrm{G}}(\boldsymbol{k})\left(g_{xx}(\boldsymbol{k}) + g_{yy}(\boldsymbol{k})\right) + h_{\mathrm{S}}(\boldsymbol{k}) \right] \delta\left(\omega - 2\epsilon(\boldsymbol{k})\right)$$



# Summary

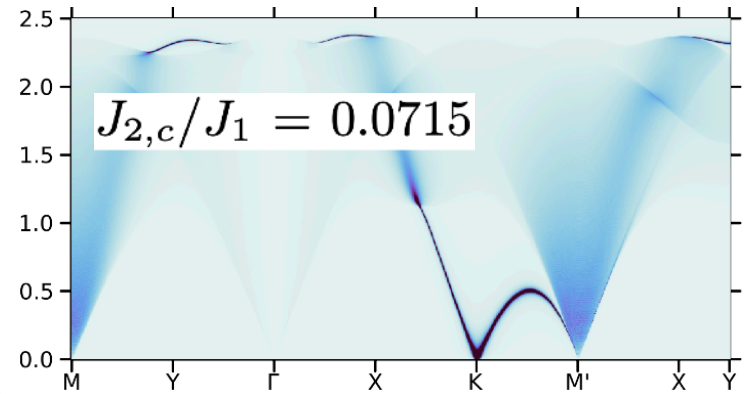
- **Self-consistent RPA for dynamical response**

  → sharp modes and continua

  → quantitative phase diagrams (no free parameter)

  Josef Willsher, Knolle, arXiv:2503.13831

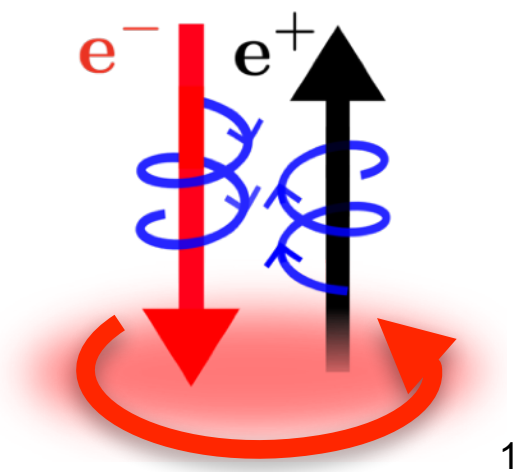
  Peng Rao, Moessner, Knolle, arXiv:2503.10330




- **Raman circular dichroism**

  → probes Quantum Geometry of spinons

  Eduard Koller, …, Knolle arXiv:2503.14091




- **Many open directions**

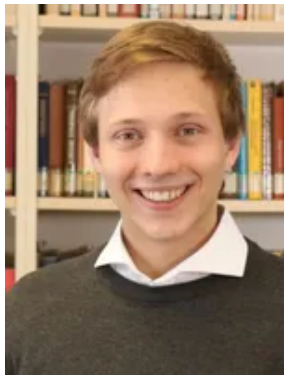
  → include fluctuations beyond pMFT

  → other non-equilibrium optical probes

Imperial College London

MCQST

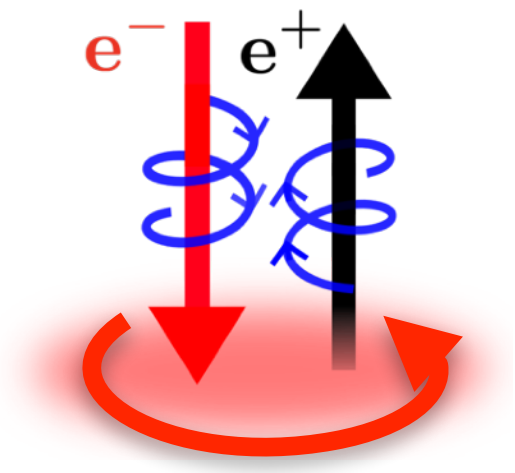
TUM

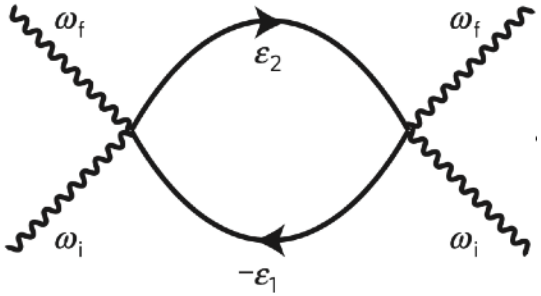


# Thank you!

**Josef Willsher**, Knolle, arXiv:2503.13831

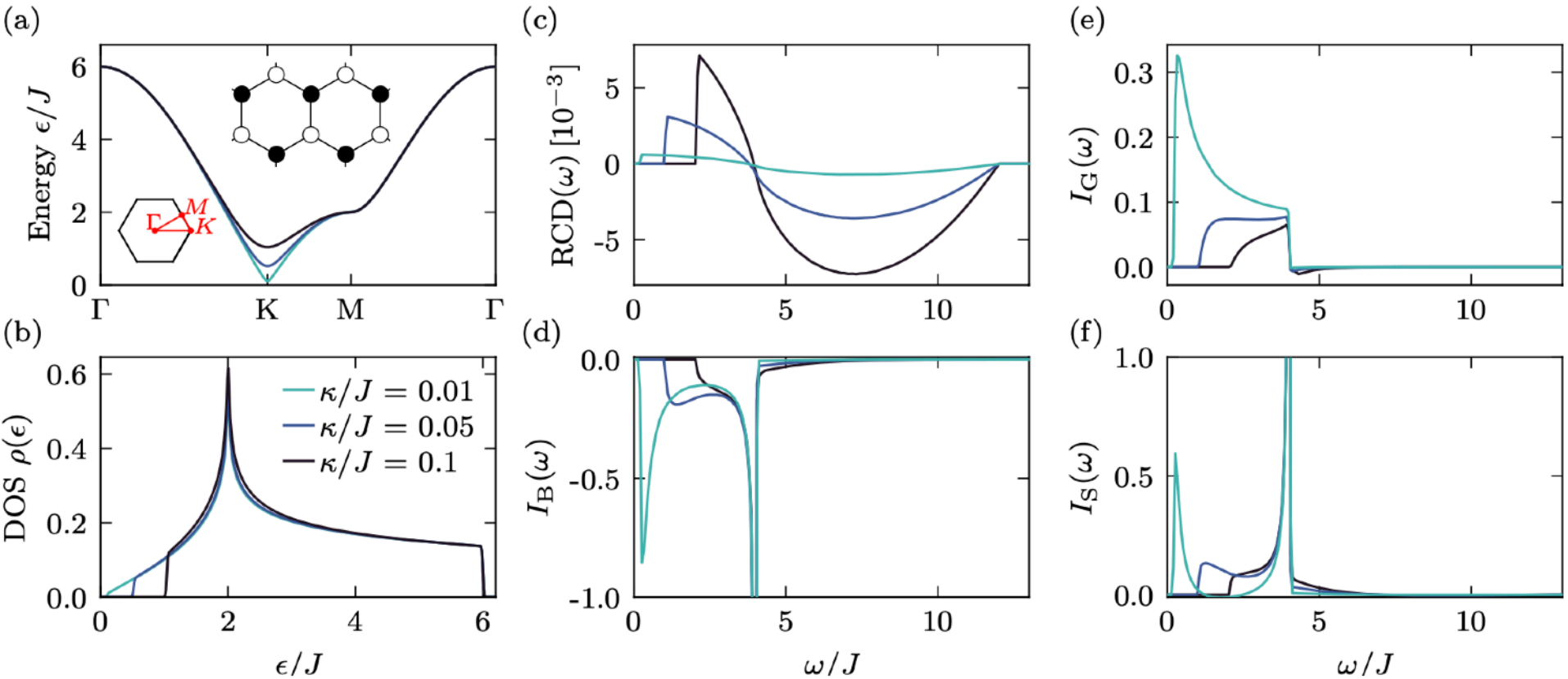
**Eduard Koller**, Valentin Leeb, Natalia Perkins, Knolle arXiv:2503.14091



# Chiral Kitaev QSL

$$I_{\mathrm{RCD}}(\omega) = \sum_{\boldsymbol{k}} \left[ h_{\mathrm{B}}(\boldsymbol{k}) F_{xy}(\boldsymbol{k}) + h_{\mathrm{G}}(\boldsymbol{k})(g_{xx}(\boldsymbol{k}) + g_{yy}(\boldsymbol{k})) + h_{\mathrm{S}}(\boldsymbol{k}) \right] \delta\Big(\omega - 2\epsilon(\boldsymbol{k})\Big)$$

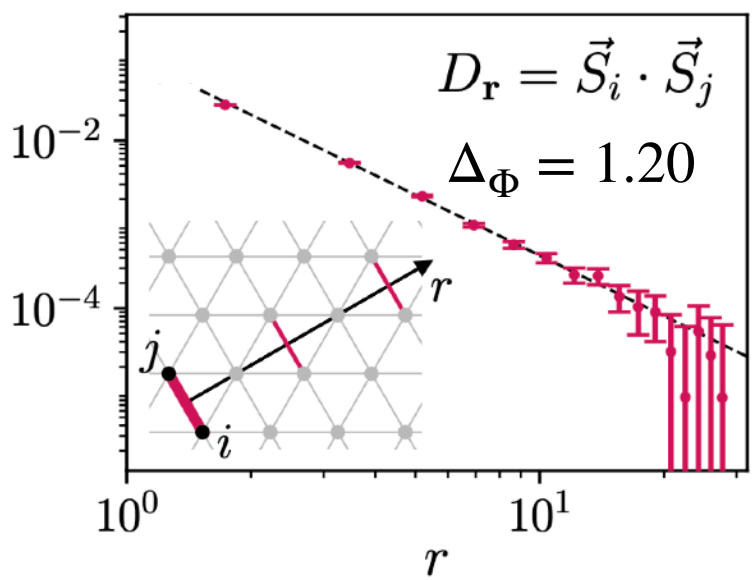


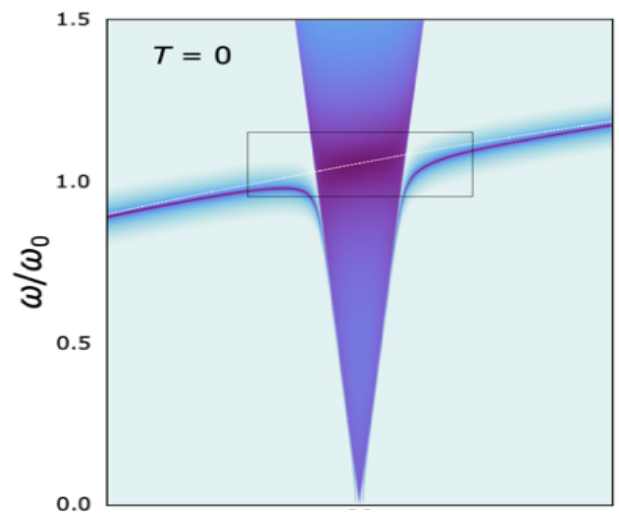
# Monopole versus Spinons

- **Full theory with transverse gauge field fluctuations missing**

  **→ singlet monopole at K/2 couples to bond-dimer correlations**

$$\langle D_o D_r \rangle_c \propto \langle \Phi_0 \Phi_r \rangle \propto r^{-2\Delta_\Phi}$$




**K/2**

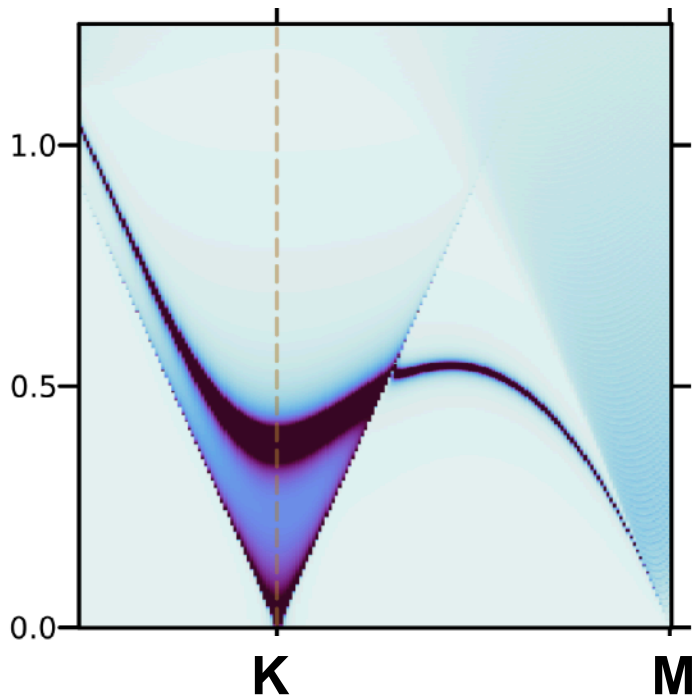
Ferrari, Willsher, … Knolle arXiv:2410.16376

Seifert, Willsher, … Knolle Nature Comm 2024

- **bond-correlations couple to phonons**

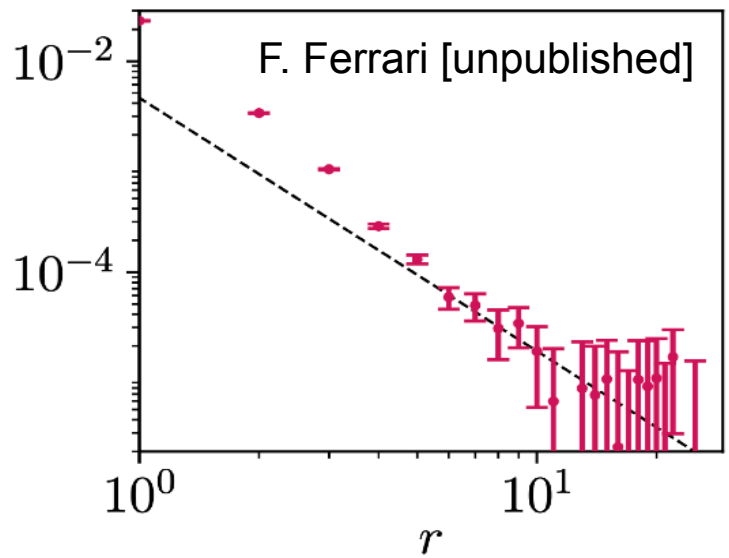
# Monopoles versus Spinons

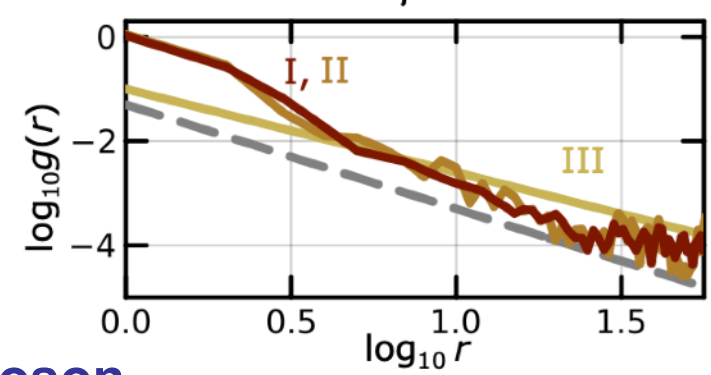
- **Triplet monopole at K**




→ couple continuum to spinons

$$\langle S_o S_r \rangle_c \propto \langle \Phi_0 \Phi_r \rangle \propto r^{-2\Delta_\Phi}$$




- **Gauge field coupled to critical boson**

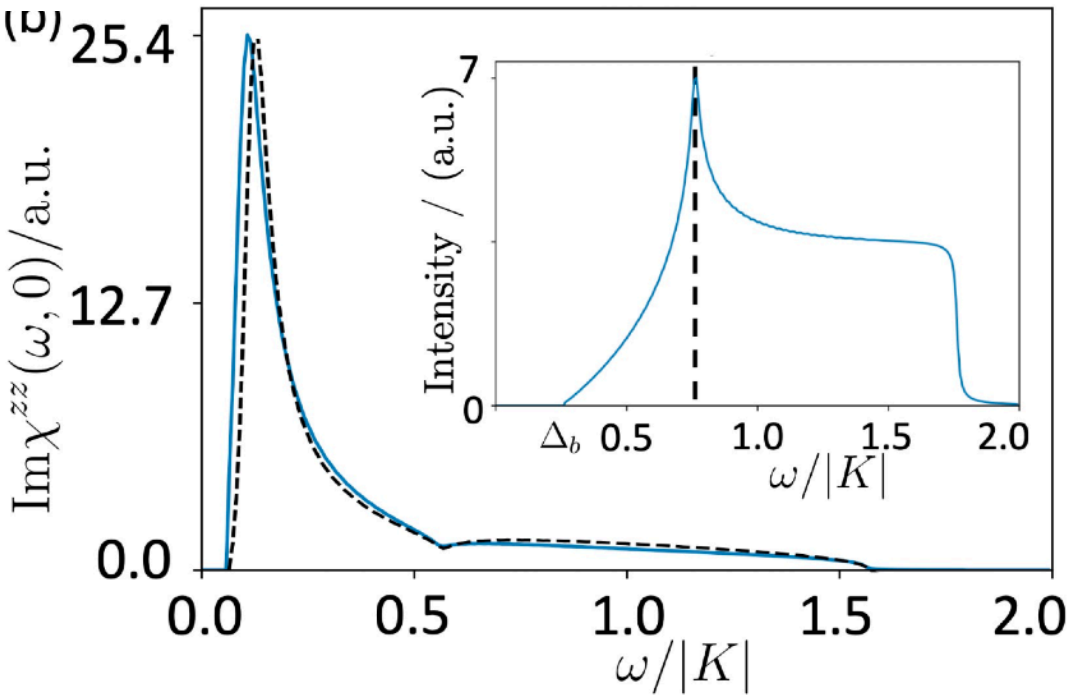
Heisenberg-Gross-Neveu theory

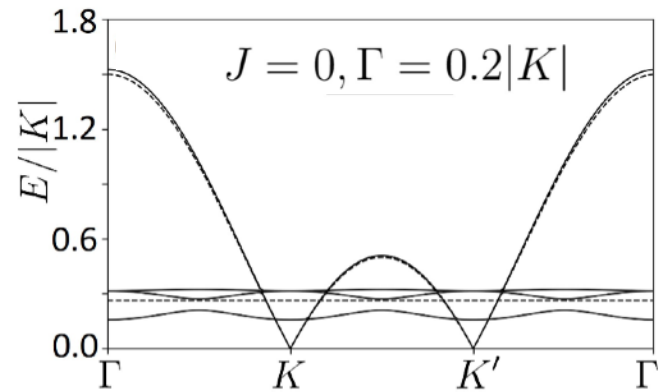
Dupuis, Paranjabe, Witczak-Krempa PRB 2019

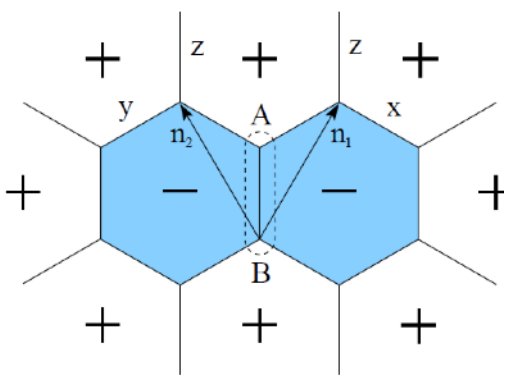
# Unreasonable Effectiveness of RPA

- **Benchmark with only known exact results in 2D - Kitaev QSL**

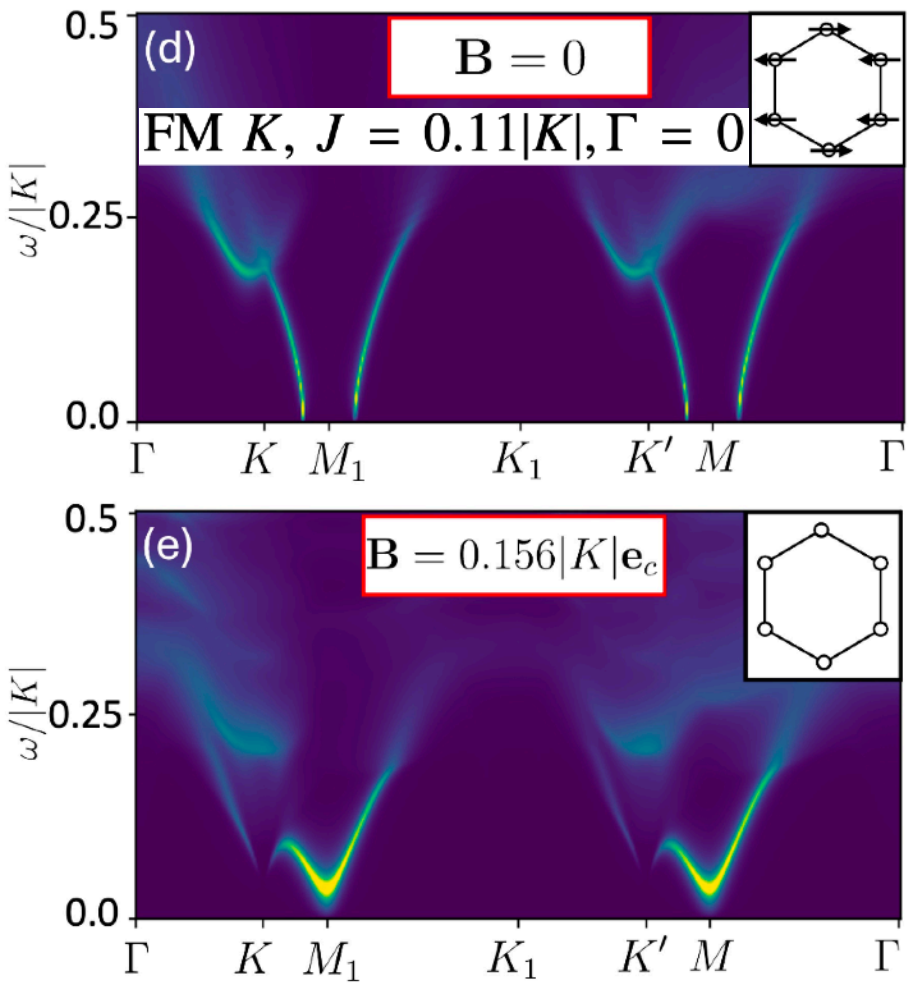
Knolle, Kovrizhin, Chalker, Moessner PRL 2014




→ **interactions mimik $Z_2$ flux excitations!**

# Kitaev QSL in a field

- **Phase diagram of extended Kitaev model and field-induced QSL**