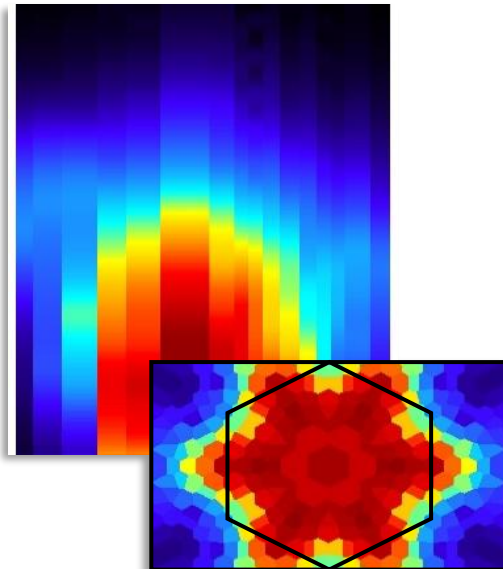
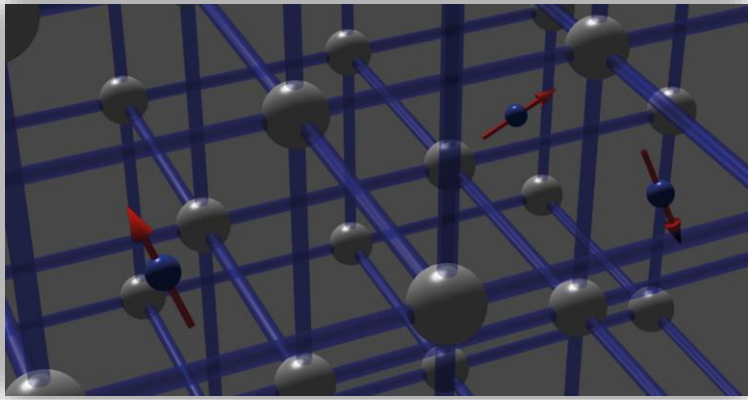


Spin-orbit driven magnetic phenomena: from first principles to experiments II



Roser Valentí
Institute of Theoretical Physics
University of Frankfurt
Germany



European School of Magnetism 2026
*Spin-orbit driven magnetic phenomena,
Monday, August 24th 2026*

Lecture 1 (DFT and magnetism) techniques

- *Motivation*
- *Microscopic description of magnetism (frustrated magnetism / spin-orbit coupling) → Kitaev frustration*

→ from first principles to spin model Hamiltonians

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \boldsymbol{\Gamma}_{ij} \cdot \mathbf{S}_j + \dots$$

Lecture 2 (Correlated electrons and magnetism) applications

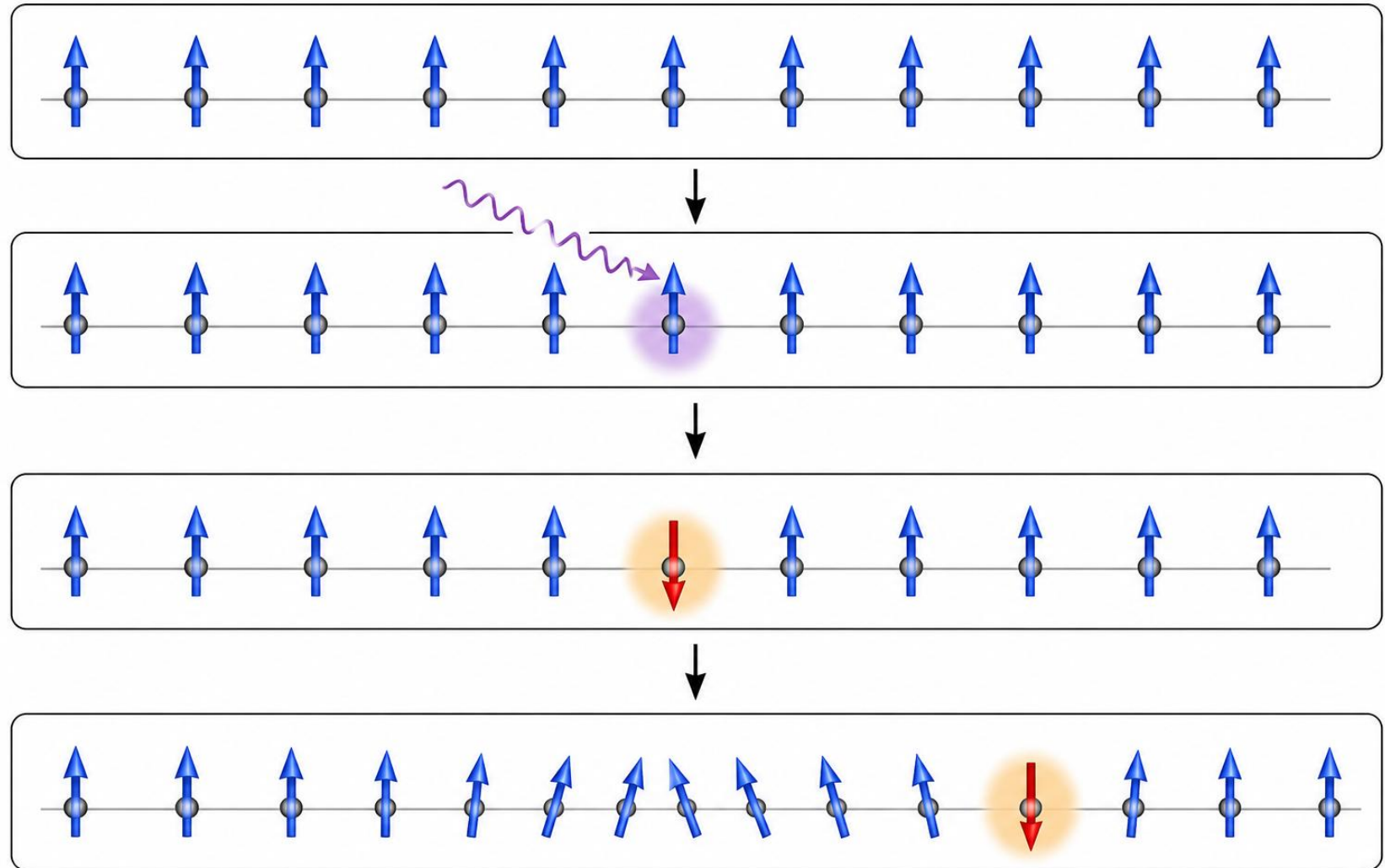
- *Excitations and dynamics in magnetic systems :*

Linear response → INS, ESR, Raman

Non-linear response → non-linear optics (2DCS)

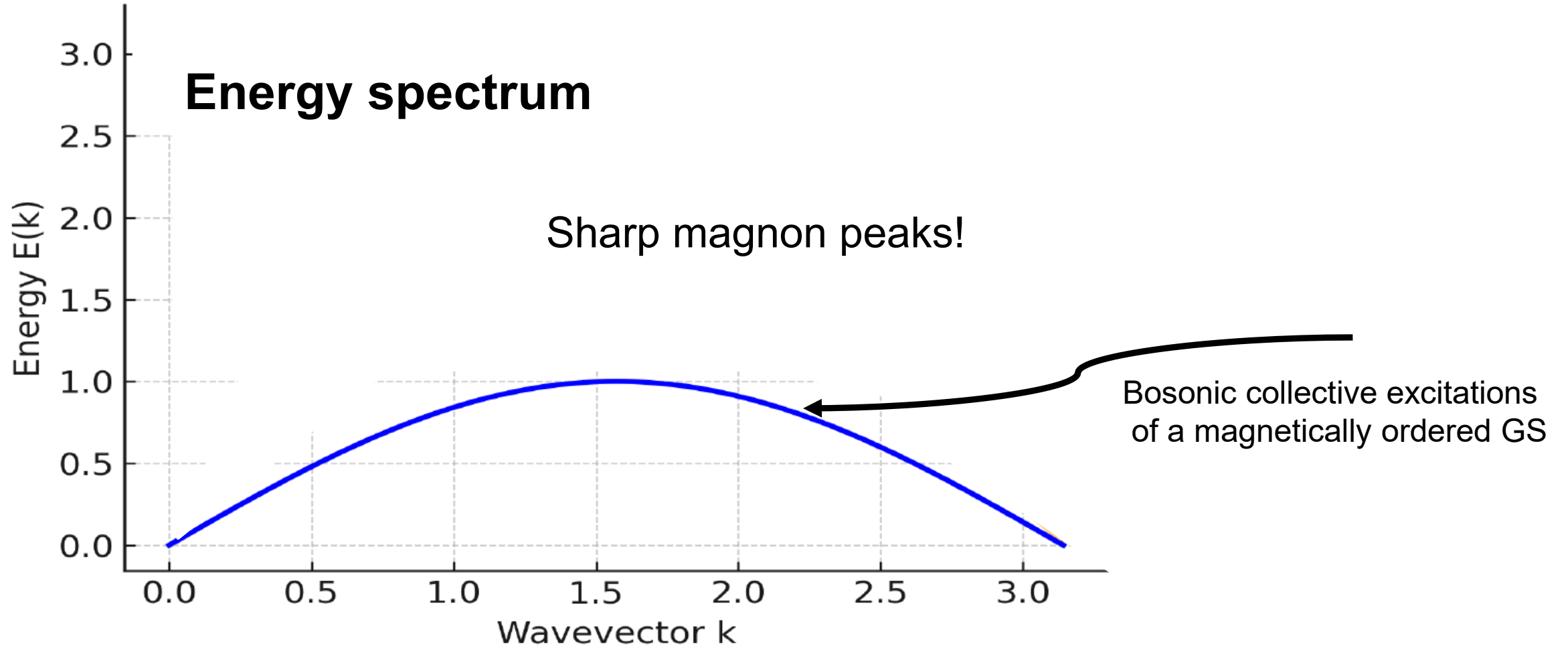
Excitations in ordered magnetic systems?

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$



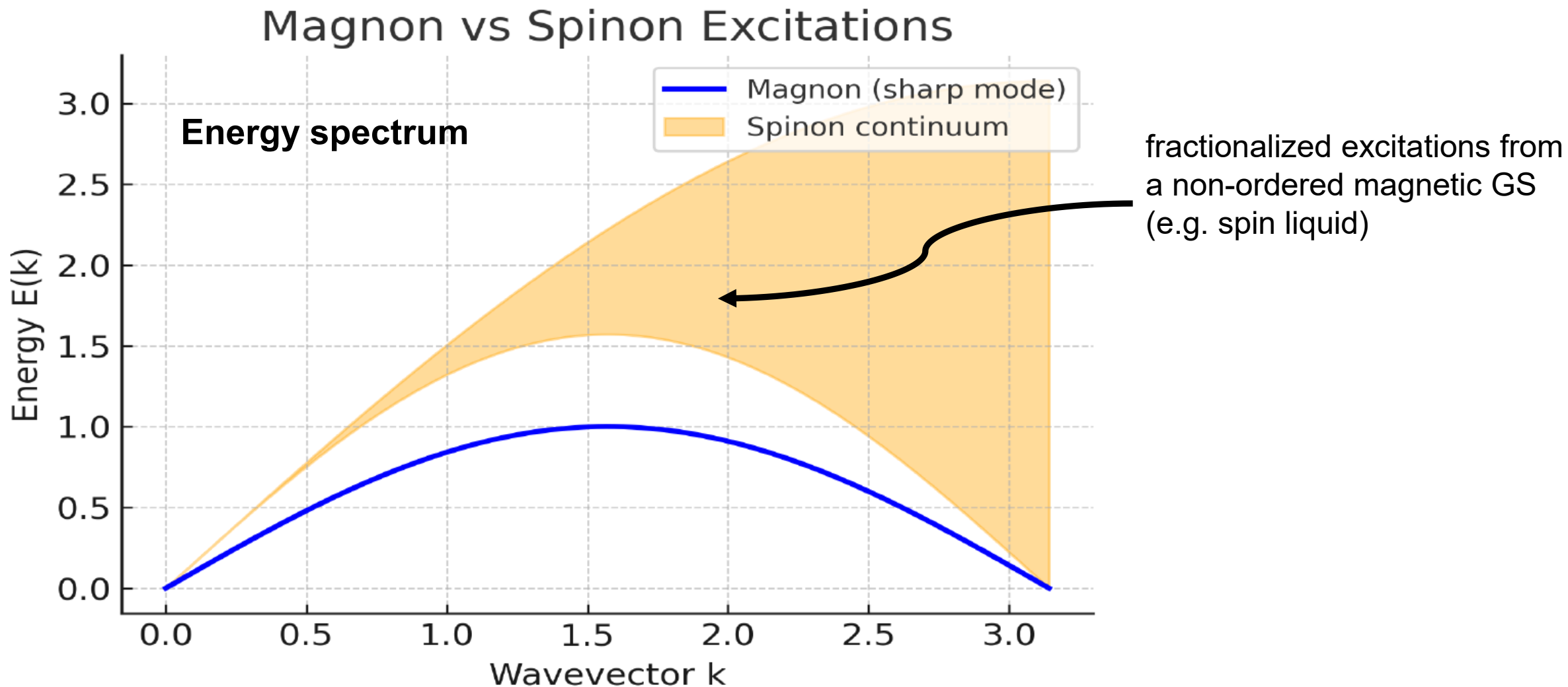
Magnons: spin density waves

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$



$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \mathbf{\Gamma}_{ij} \cdot \mathbf{S}_j$$

Excitations in quantum spin liquids? (Geometrical frustration and/or Kitaev-like interactions)



calculations of dynamical properties?

Once the spin Hamiltonian is known →
apply an appropriate many-body method to calculate dynamics

For frustrated spin Hamiltonians:

QMC (sign problem)

DMRG (low-d)

VMC (variational ansatz)

ED (finite systems)

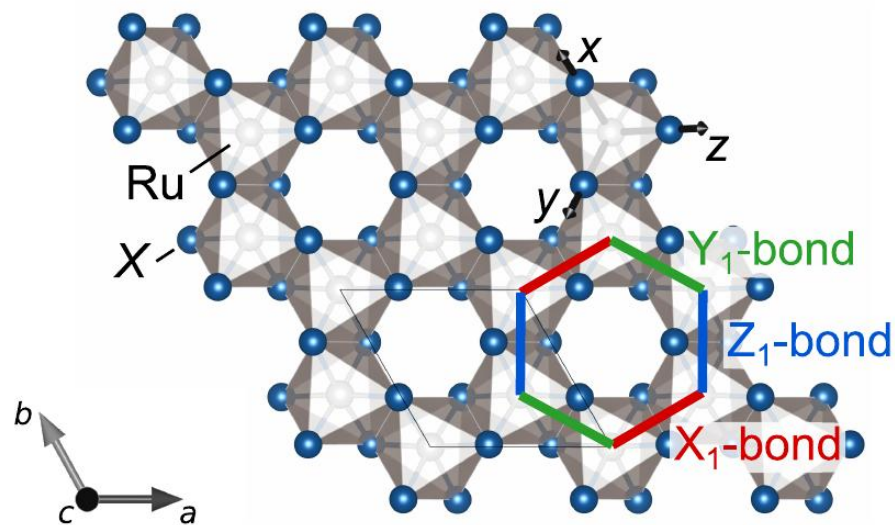
...

Example for spin-orbit-driven Hamiltonians (Kitaev-like) →

- INS, ESR, non-linear optics (2DCS), Thermal Hall...



4d⁵ Ru



Mott insulator

Kitaev candidate

$\mathbf{j}_{\text{eff}} = \frac{1}{2}$ honeycomb lattice

Ab-initio derived models: $\mathcal{H}_{\text{eff}} = \sum_{ij} \mathbf{S}_i \cdot \mathbb{J}_{ij} \cdot \mathbf{S}_j$ $\mathbf{S} = \mathbf{j}_{\text{eff}}$

$$\mathcal{H} = \underbrace{J_1 \mathbf{S}_i \cdot \mathbf{S}_j}_{\text{Heisenberg}} + \underbrace{K_1 S_i^\gamma S_j^\gamma}_{\text{Kitaev}} + \underbrace{+\Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)}_{\text{Off-Diagonal Anisotropic}}$$

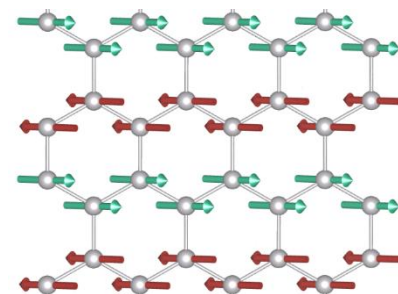
$$\underbrace{+\Gamma'_1 (S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta)}_{\text{Off-Diagonal Anisotropic}} + \underbrace{J_3 \mathbf{S}_i \cdot \mathbf{S}_j}_{\text{Heisenberg}}$$

$J_1 = -0.5, K_1 = -5.0, \Gamma_1 = +2.5, J_3 = +0.5 \text{ meV}$

Winter..., RV PRB 93, 214431 (2016)
Kaib..., RV npj Quantum Materials (2022)

Rau, Kee PRL 112, 077204 (2014)
Yamaji et al. PRL 113, 107201 (2014)

Möller, Maksimov, Jiang, White, RV, Chernyshev PRB (2025)



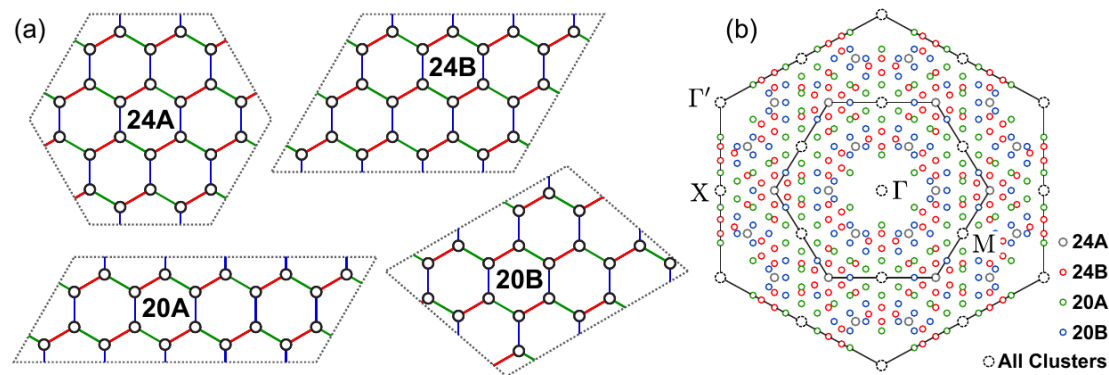
◆ Zigzag magnetic order state.

Exact diagonalization of finite clusters

Excitations –Inelastic Neutron Scattering

INS intensity

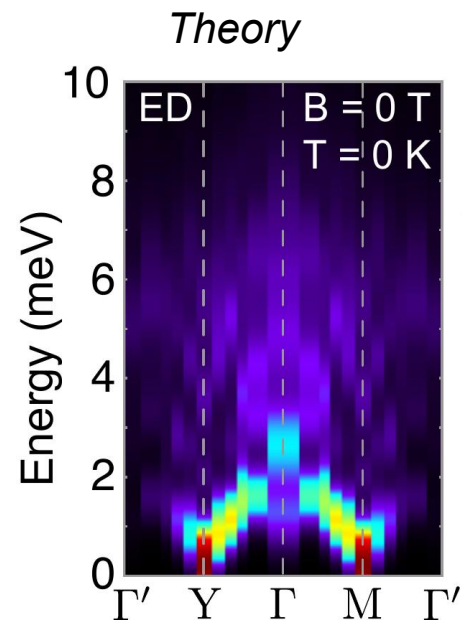
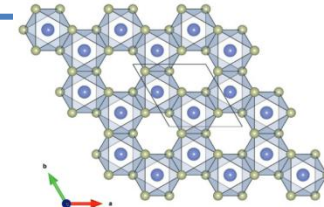
$$\mathcal{I}(\mathbf{k}, \omega) \propto f^2(\mathbf{k}) \int dt \sum_{\mu, \nu} (\delta_{\mu, \nu} - k_{\mu} k_{\nu} / k^2) \times \\ \times \sum_{i, j} \langle S_i^{\mu}(t) S_j^{\nu}(0) \rangle e^{-i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j) - i\omega t}$$



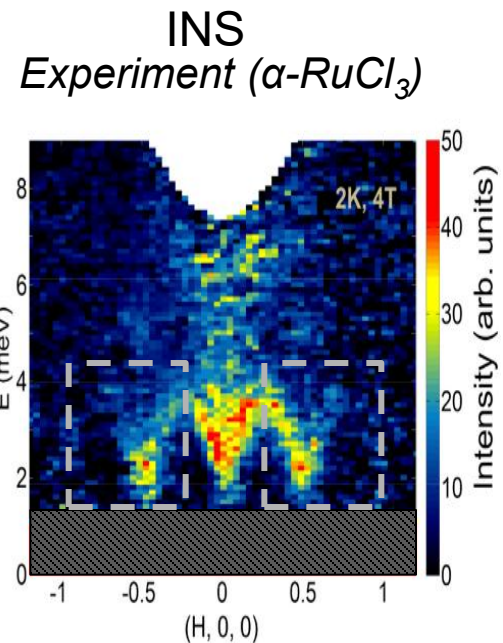
$$J_1 = -0.5, K_1 = -5.0, \Gamma_1 = +2.5, J_3 = +0.5 \text{ meV}$$

*Coexistence of magnons + continuum
majorana excitations?*

INELASTIC NEUTRON SCATTERING

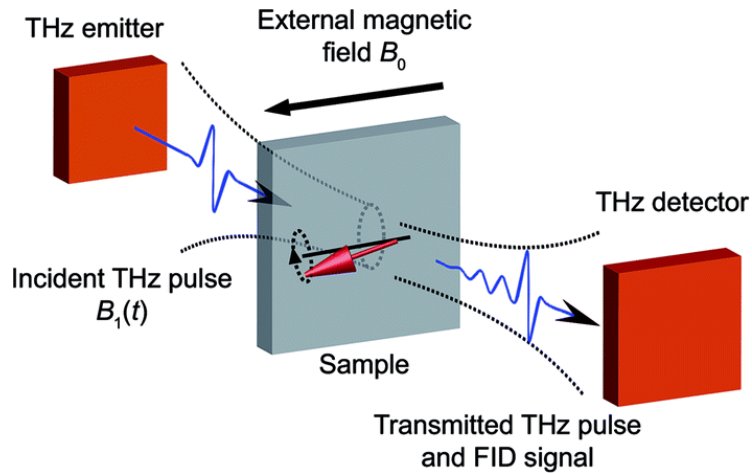


Winter, ..., RV
Nat. Comm. 8, 1152 (2017)

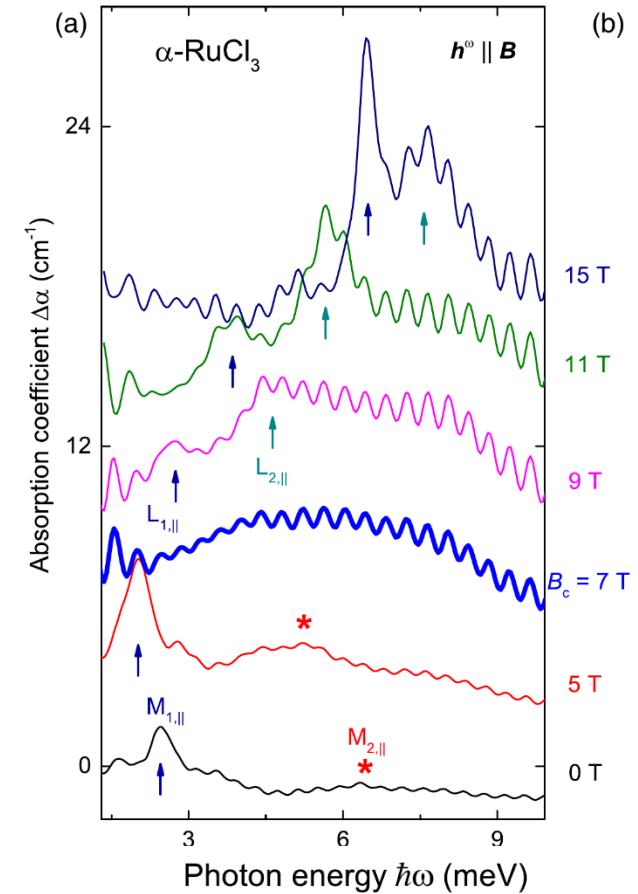


Banerjee, et al.
npj QM 3, 8 (2018)

Linear response:

THz absorption ($\sim S(\mathbf{q} = 0, \omega)$)*Chem. Sci.* **8**, 7312 (2017)

THz pulse on the system is used to probe
and excite its dynamics



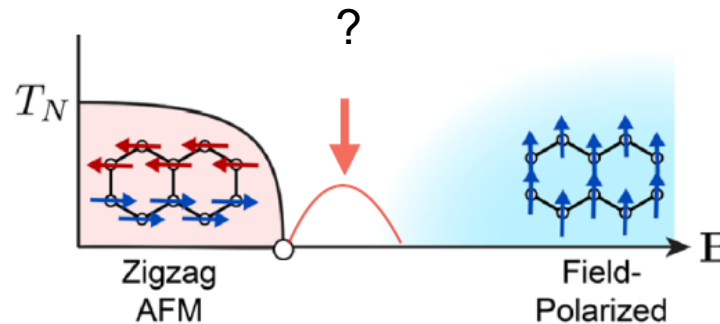
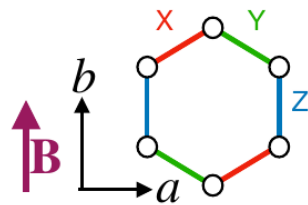
Wang et al.
PRL 119, 227202 (2017)
Winter, ..., RV
PRL 120, 077203 (2018)

Origin of continuum?

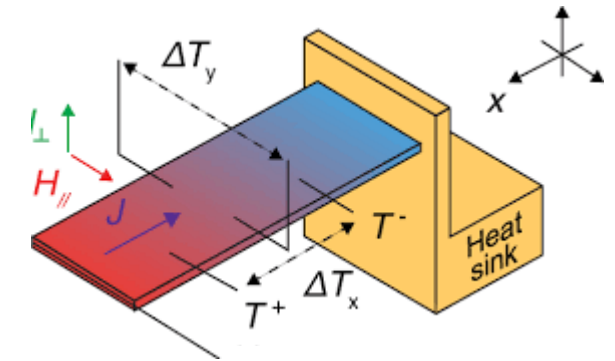
breakdown of magnons?
fractionalized excitations?

Furthermore...

➡ Controversy on possible field-induced spin liquid for $B \gtrsim B_c$



Winter, ..., RV, PRL 120, 077203 (2018)



Thermal Hall measurements

Kasahara et al. *Nature* 559, 227 (2018)

Yokoi et al., *Science* (2021)

Czajka et al., *Nat. Phys.* (2021)

Lefrancois et al., *PRX* (2022)

Bruin et al., *Nat. Phys.* (2022)

Dhokal, ..., RV, Winter arXiv:2407.0060

Nature of magnetic excitations?

**Dynamical (linear and non-linear)
spin response?**

Linear response:

$$\chi_{\mathcal{O}}^1(t_1) = i\Theta(-t_1)\langle[\mathcal{O}, \mathcal{O}(t_1)]\rangle$$

Example: **dynamical spin structure factor**

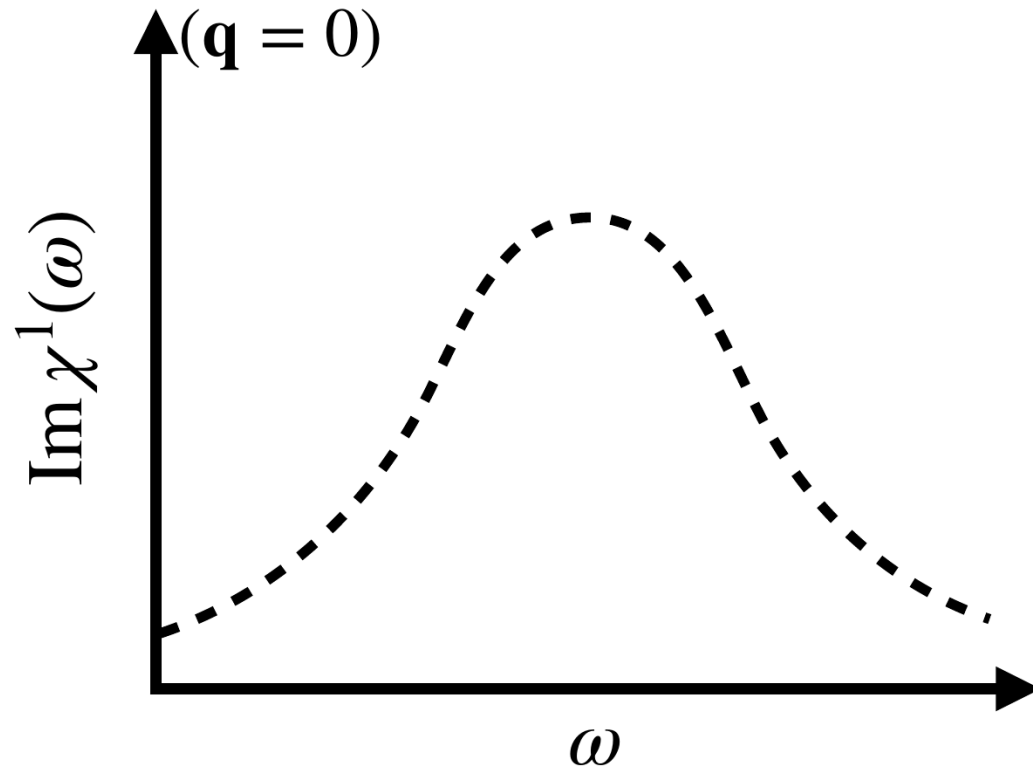
$$S^\mu(\mathbf{q}, \omega) = \text{Im} \chi_{S_{\mathbf{q}}^\mu}^1(\omega) \sim \sum_n |\langle \text{GS} | S_{\mathbf{q}}^\mu | \Psi_n \rangle|^2 \delta(\omega - E_n)$$

→ probes which excitations are accessible from the ground state

Quantum spin liquid:

Continuum in the Dynamical Structure Factor

$$S^\mu(\mathbf{q}, \omega) = \text{Im} \chi_{S^\mu_{\mathbf{q}}}^1(\omega) \sim \langle \text{GS} | \mathcal{O} | \Psi_n \rangle$$



Continuum can stem from:

- Fractionalized excitations
- Damped/decaying mode? Magnon decay?
- Disorder?

Linear response:

dynamical spin structure factor

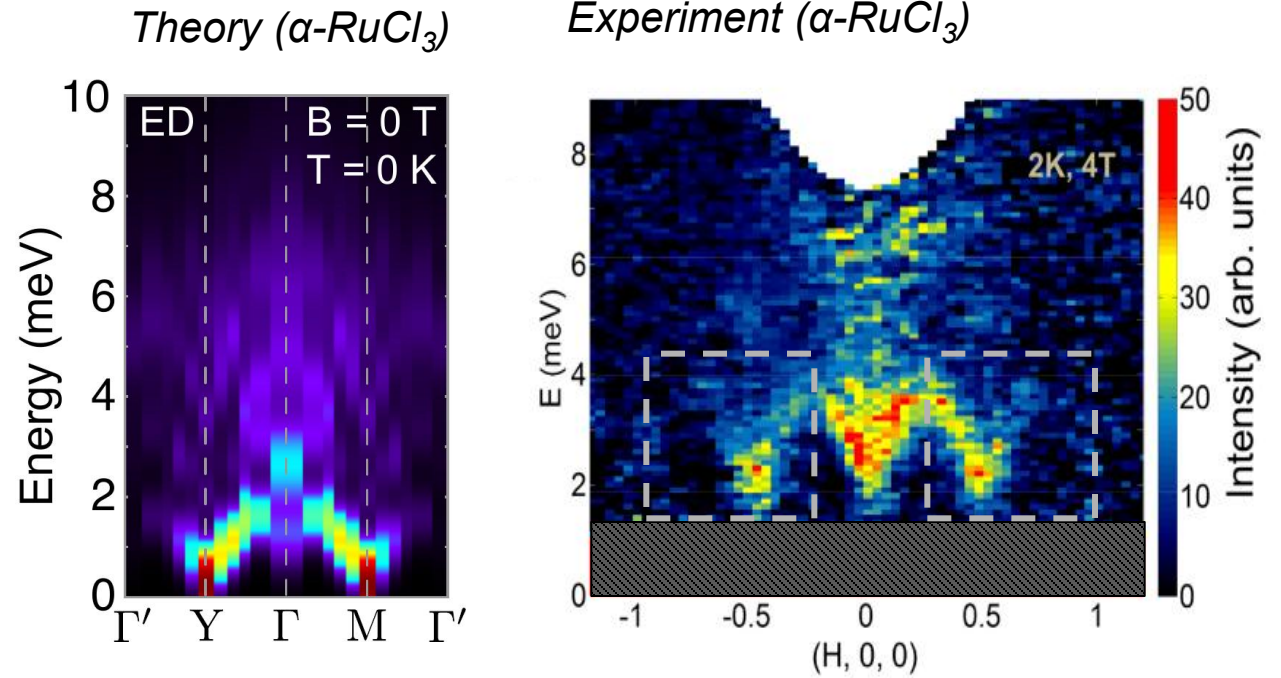
$$S^\mu(\mathbf{q}, \omega) = \text{Im} \chi_{S^\mu}^1(\omega) \sim \sum_n |\langle \text{GS} | S^\mu_{\mathbf{q}} | \Psi_n \rangle|^2 \delta(\omega - E_n)$$

$$\mathcal{H} = \sum_{\langle ij \rangle} J_1^\gamma \mathbf{S}_i \cdot \mathbf{S}_j + K_1^\gamma S_i^\gamma S_j^\gamma + \Gamma_1^\gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \\ + \sum_{\langle\langle ij \rangle\rangle} J_3 \mathbf{S}_i \cdot \mathbf{S}_j - \mu_B \sum_i \mathbf{B} \cdot \mathbf{g} \cdot \mathbf{S}_i,$$

$$\gamma \in \{x, y, z\}$$

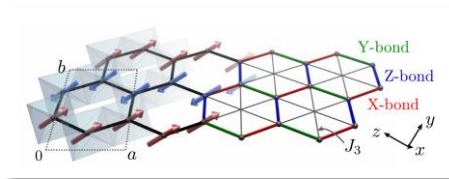
$$J_1 = -0.5, K_1 = -5.0, \Gamma_1 = +2.5, J_3 = +0.5 \text{ meV}$$

Coexistence of magnons + continuum



Winter ... RV
Nat. Comm. 8, 1152 (2017)

Banerjee, et al. npj QM 3, 8 (2018)



Origin of continuum?

breakdown of magnons?
fractionalized excitations?

Nonlinear spectroscopy

how to calculate and interpret χ ?

Higher-order responses:

challenging task!

$$\chi_{\mathcal{O}}^1(t_1) = i\Theta(-t_1)\langle[\mathcal{O}, \mathcal{O}(t_1)]\rangle \sim \langle \text{GS} | \mathcal{O} | \Psi_n \rangle$$

$$\chi_{\mathcal{O}}^2(t_1, t_2) = i^2\Theta(-t_1)\Theta(t_1 - t_2)\langle[[\mathcal{O}, \mathcal{O}(t_1)], \mathcal{O}(t_2)]\rangle \sim \langle \text{GS} | \mathcal{O} | \Psi_n \rangle \langle \Psi_n | \mathcal{O} | \Psi_m \rangle \langle \Psi_m | \mathcal{O} | \text{GS} \rangle$$

$$\chi_{\mathcal{O}}^3(t_1, t_2, t_3) = i^3\Theta(-t_1)\Theta(t_1 - t_2)\Theta(t_2 - t_3)\langle[[[\mathcal{O}, \mathcal{O}(t_1)], \mathcal{O}(t_2)], \mathcal{O}(t_3)]\rangle \sim (\dots)$$

(:) $\rightarrow$ probes which excitations accessible from the ground state

$\rightarrow$ probes relations **between excitations**

Higher-order responses:

Analytical work:

Wan, Armitage *PRL* 122, 257401 (2019) (*Transverse-field Ising chain*)

Kanega et al. *PRR* 3, L032024 (2021) (*Kitaev honeycomb*)

Kruptnitska, Brenig *PRB* 108, 075120 (2023), arXiv:2403.08867 (*Kitaev honeycomb*)

Qiang et al. *PRL* 133, 126505 (2024) (*Kitaev honeycomb*)

Numerical work:

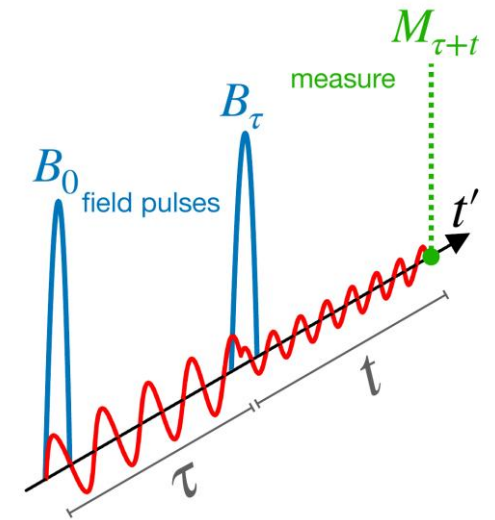
Sim et al. *PRB* 107, L100404 (2023) (*iMPS, Transverse-field Ising chain*)

Gao et al. *PRB* 107, 165121 (2023) (*DMRG Ising ladder*)

Watanabe et al. *PRB* 110, 134443 (2024) (*ED, Transverse-field Ising chain, Twisted Kitaev chain*)

Watanabe et al. *PRL* 134, 106703 (2025) (*ED, spin-1 Heisenberg+SIA*)

Two-dimensional coherent spectroscopy



- Explicit numerical time evolution for a grid t, τ
- Fourier transform obtained from χ

computationally demanding

Second-order responses:

(Exact diagonalization)

Our approach

$$\chi_{\mathcal{O}}^2(t_1, t_2) = i^2 \Theta(-t_1) \Theta(t_1 - t_2) \langle [[\mathcal{O}, \mathcal{O}(t_1)], \mathcal{O}(t_2)] \rangle$$

$$\chi^{(2)}(\omega_t, \omega_\tau) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dt d\tau e^{-i\omega_t t - i\omega_\tau \tau} \chi^{(2)}(t, t + \tau)$$

David Kaib



Our approach: (Formally) perform time-evolutions within suited M -dim. Krylov space ($M \ll \dim$)

$$\mathcal{K}_M(H, |\phi_0\rangle) = \mathcal{K}_M(H, \mathcal{O}|0\rangle) = \text{span} \left(\{ \mathcal{O}|0\rangle, H\mathcal{O}|0\rangle, H^2\mathcal{O}|0\rangle, \dots, H^{M-1}\mathcal{O}|0\rangle \} \right)$$

Result: Allows direct evaluation in frequency space:

$$\chi^{(2)}(\omega_t, \omega_\tau) \approx \sum_{n,m}^M \frac{N_{KS}^2 v_{n,0}^* v_{m,0} \sum_{j,k}^M v_{n,j}^* v_{m,k} \langle \tilde{\phi}_j | \mathcal{O} | \tilde{\phi}_k \rangle}{(\omega_t + i\eta - \tilde{E}_n)(\omega_\tau + i\eta - \tilde{E}_m)} + (\dots)$$

„Byproducts“ of Lanczos algorithm

- avoids explicit numerical time evolution and Fourier transform
- contributions from all orders $(iHt)^m$ $m < M$ are exact
- extendable to higher order susceptibilities and finite T

Kaib, Möller, RV PRR (2026)



Marius Möller

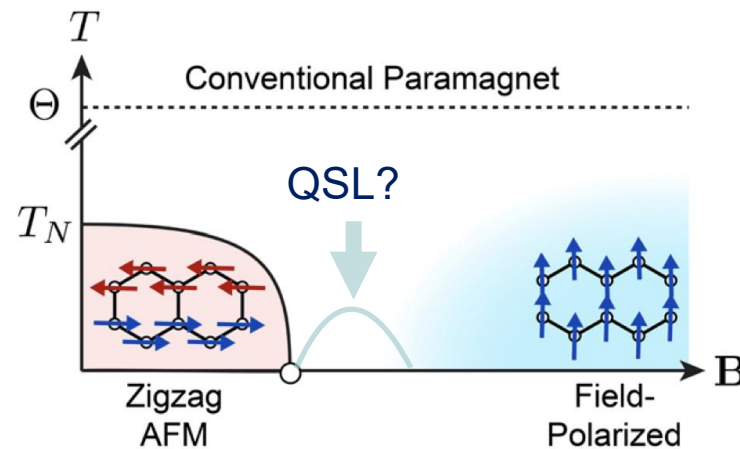
Validated with existing results so far!

Application: α -RuCl₃ (minimal model)

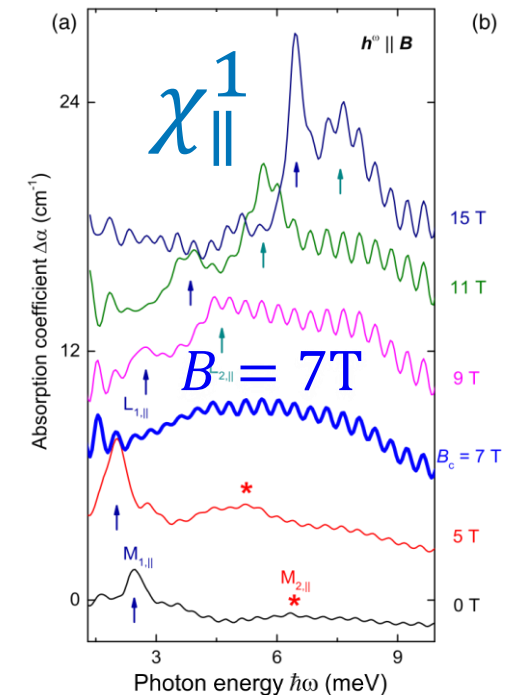
$$H = \sum_{\langle ij \rangle_\gamma} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + K_1 S_i^\gamma S_j^\gamma + \Gamma_1 \left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \right) + \sum_{\langle\langle\langle ij \rangle\rangle\rangle} J_3 \mathbf{S}_i \cdot \mathbf{S}_j - \mu_B \sum_i \mathbf{B} \cdot \mathbf{g} \cdot \mathbf{S}_i$$

$$(J_1, K_1, \Gamma_1, J_3) = (-0.5, -5, +2.5, +0.5) \text{ meV}$$

Winter et al., Nat. Comm. **8**:1152 (2017)

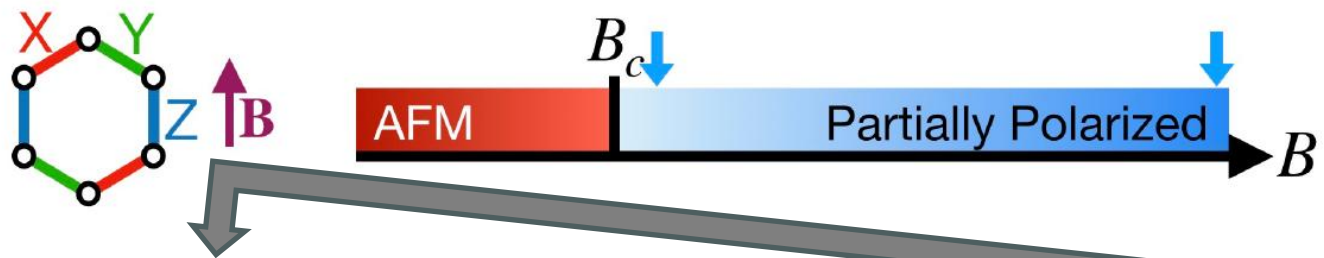


Winter, .., RV, PRL 120, 077203 (2018)



Wang et al., PRL 119, 227202 (2017)

THz absorption spectroscopy

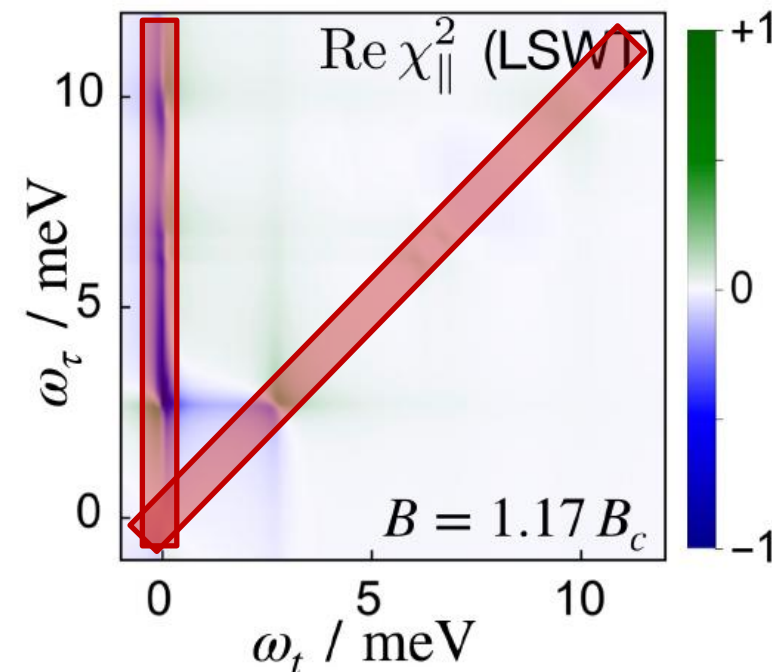
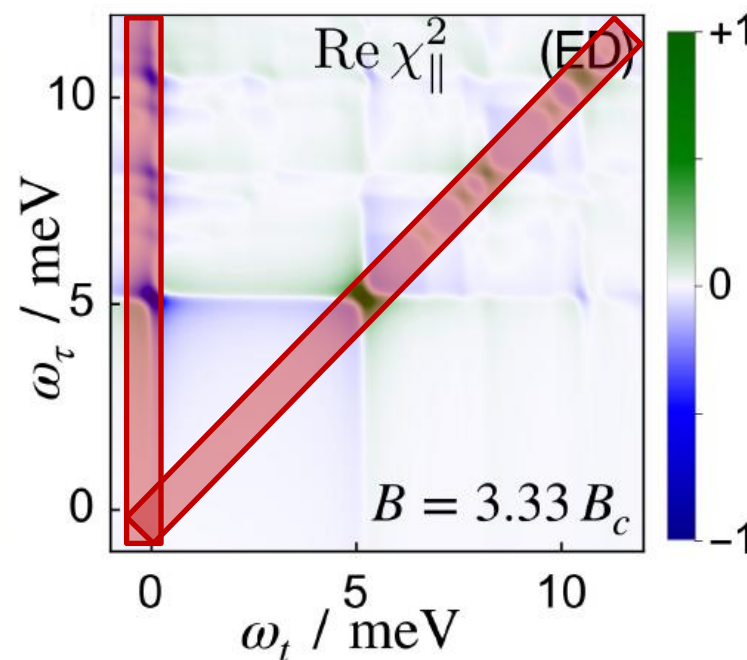
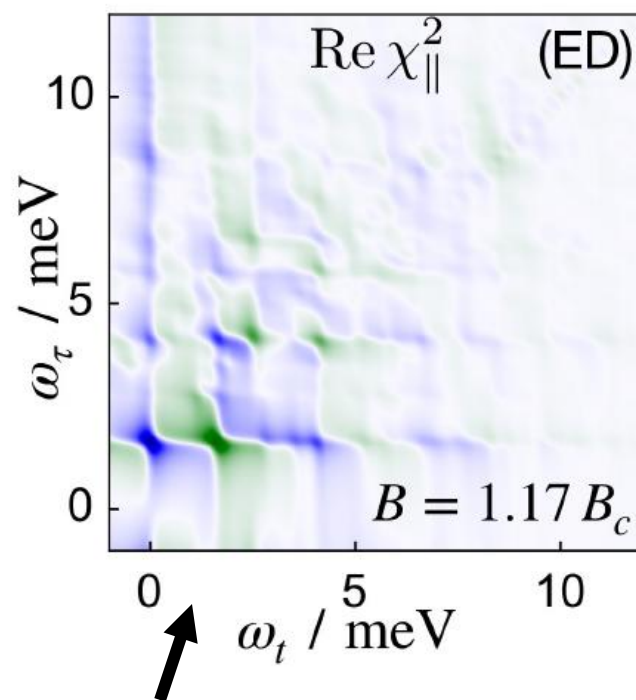


2DCS

$B \gtrsim B_c$: Magnon
breakdown

$B \gg B_c$: Conventional
magnons restored

Conventional
magnon picture



Origin: interacting fractionalized excitations, etc...

Kaib, Möller, RV PRR (2026)

→ At the LSWT level,
only F_{diag} and F_{vert} poles,
independent of model

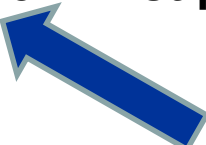
Lecture 2 (Correlated electrons and magnetism) applications

Frustrated magnets
(Kitaev-like magnets)

SUMMARY

- Derivation of microscopic (spin) models from first principles / projection method
- Nature of magnetic excitations?
- Linear response (INS, ESR, ..)
- 2D coherent spectroscopy in the THz regime
is a **powerful** technique to diagnose the nature of
(linear-response) broad spectra in **quantum magnets**
- **Efficient** computational method in **ED** to calculate non-linear
responses
 - can be extended to **generalized models**
and **higher-order** response functions

projection method



Kaib, Möller, RV PRR (2026)

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→ from first principles to spin model Hamiltonians

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \boldsymbol{\Gamma}_{ij} \cdot \mathbf{S}_j + \dots$$

Lecture 2 (Correlated electrons and magnetism) applications

- *Excitations and dynamics in magnetic systems :*

Linear response → INS, ESR, Raman

Non-linear response → non-linear optics (2DCS)