Tutorial

Inelastic Neutron Scattering in the Quasiparticle Zoo

Magnons, Spinons, Bound States, and Their Interactions

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DE-SC-0018660

Spice Workshop

Quantum Geometry and Transport of Collective Excitations in (Non-)Magnetic Insulators

May 7, 2025

Outline

- Heavy Introduction
    - Quantum Magnetism Big Picture
    - Research Approach
    - Inelastic Neutron Scattering
    - Modeling

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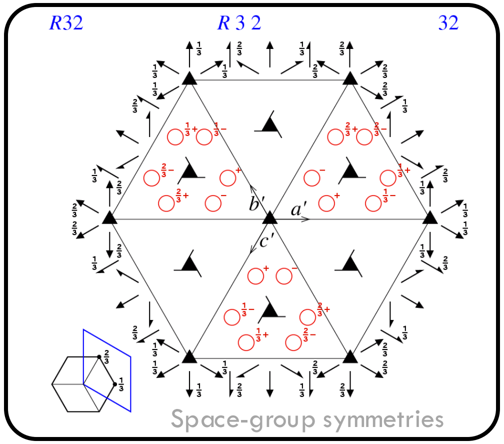
- ❑ Heavy Introduction
  - Quantum Magnetism Big Picture
  - Research Approach
  - Inelastic Neutron Scattering
  - Modeling
- ❑ Examples: Greatest Hits 2012-2025
  - Spinons: Heisenberg spin-1/2 chains in copper sulfate
  - Magnons: Square-lattice problems
  - Bound-states: Triangular uniaxial spin-1 system FeI_2
  - **C**ontinuous excitations does not always mean fractionalization/decay
- ❑ Conclusion

Quantum magnetism and the Heisenberg model

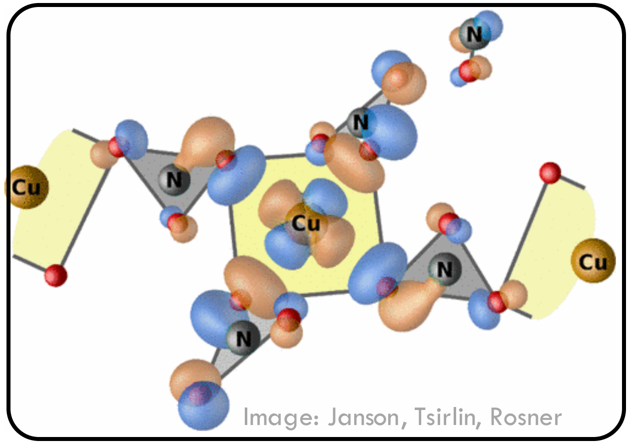
- Magnetic insulators are important for quantum physics and applications

$$\hat{\mathcal{H}}_{\text{el}} =$$

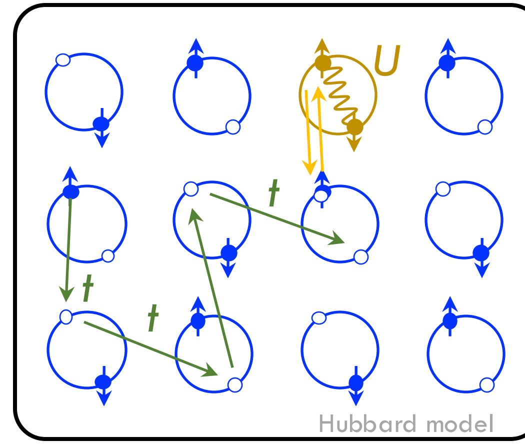
Crystallography



Quantum Chemistry

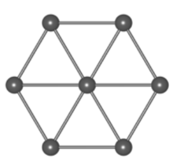


Electronic Correlations



- Emergence of a new low-energy scale ***J*** (quantum spin exchange)

J_{ij}

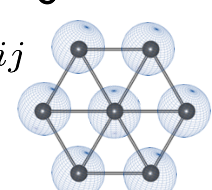


lattice-space
(extended)

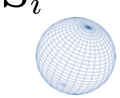
$$\hat{\mathcal{H}}_{\text{mag}} = \sum_{ij} J_{ij}\, \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + \cdots$$

~~Heisenberg Model (1928)~~

J_{ij}



$\hat{\mathbf{S}}_i$

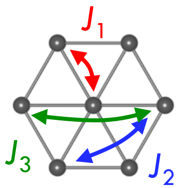


$$= \frac{\hbar}{2}\{\hat{\sigma}_i^x, \hat{\sigma}_i^y, \hat{\sigma}_i^z\}$$

spin-space
(local)

Quantum magnetism and the Heisenberg model

- Effective magnetic Hamiltonians are usually more complicated:



lattice-space

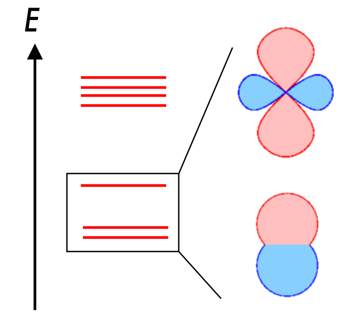
$$\hat{\mathcal{H}}_{\mathrm{mag}} = \sum_{ij} \hat{\mathbf{S}}_i \underbrace{\begin{bmatrix} \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots \end{bmatrix}_{ij}}_{\mathcal{J}_{ij}} \hat{\mathbf{S}}_j$$

anisotropic bond-dependent exchange

+ local anisotropies

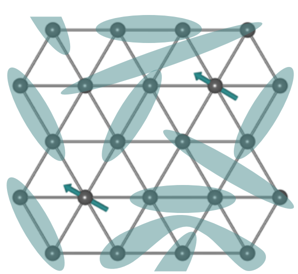
+ multipolar exchange

+ disorder

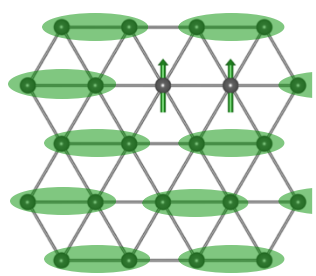


spin-space

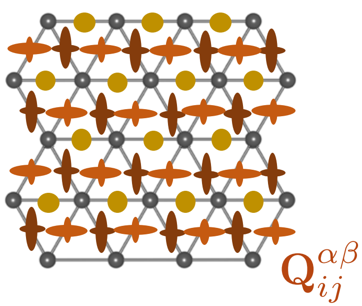
- What is the correlated physics at (and below) the spin exchange scale?



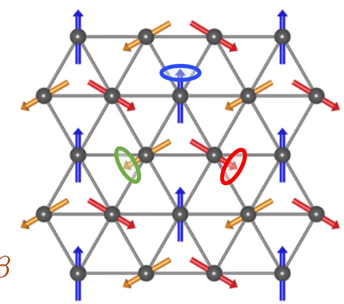
Quantum spin liquid



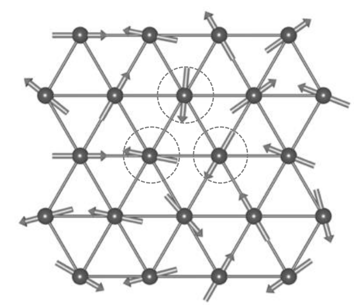
Quantum paramagnet



Multipolar Order



Dipolar Order

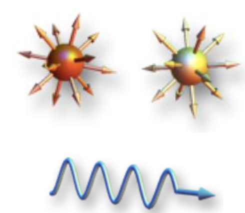


Classical paramagnet

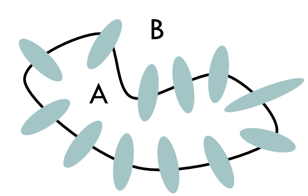
entanglement

- Why do we care?

New states of matter, perhaps new forms of QFT



exotic quasiparticles



entanglement entropy

Quantum magnetism and the Heisenberg model

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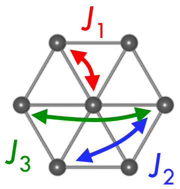

lattice-space

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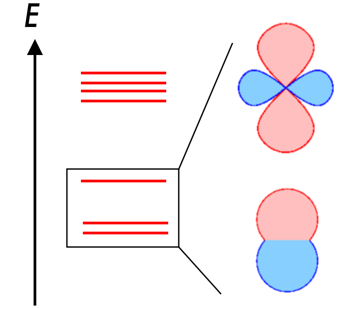
\+ disorder



spin-space

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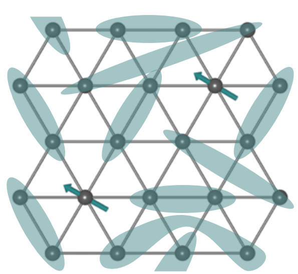
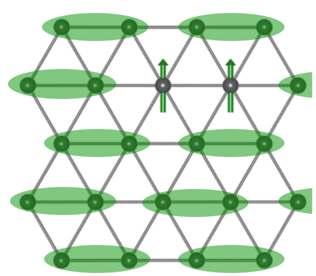
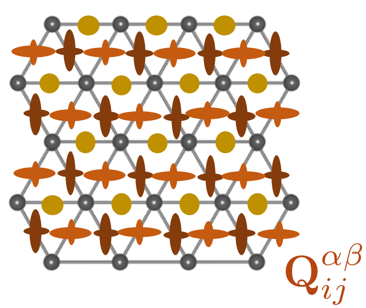


Quantum spin liquid



Quantum paramagnet



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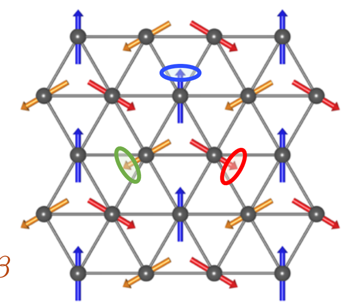

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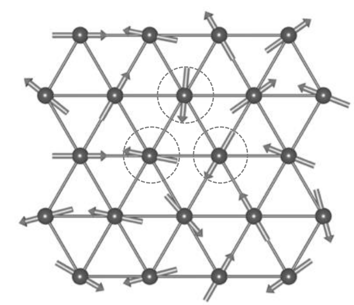
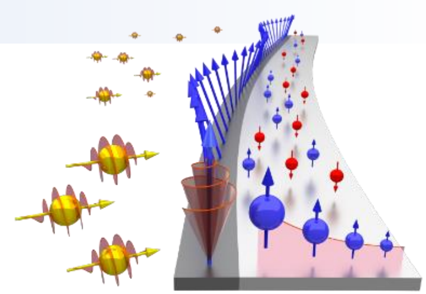


Classical paramagnet

entanglement

- Why do we care?

Understand low-energy excitations (perhaps for applications?)



Schultheiß Group (Dresden)

Quantum magnetism and the Heisenberg model

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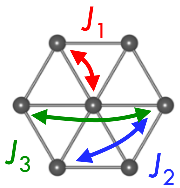

lattice-space

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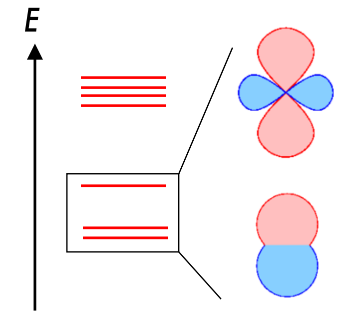

spin-space

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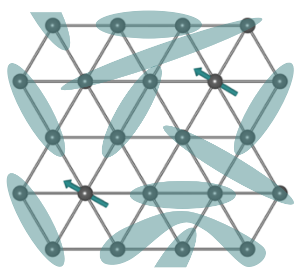
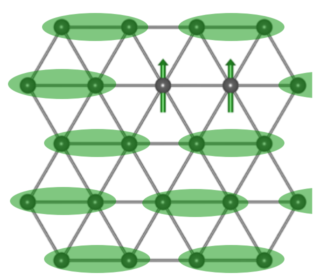
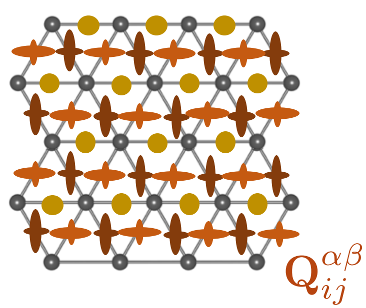


Quantum spin liquid



Quantum paramagnet



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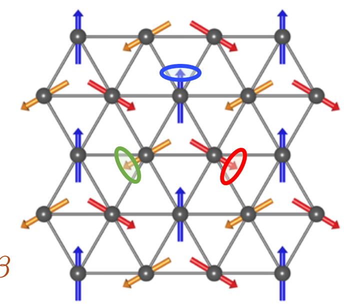
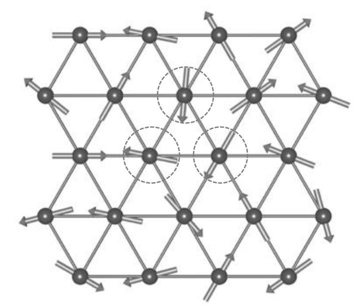


Dipolar Order

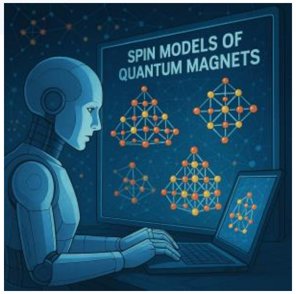


Classical paramagnet

entanglement

- Why do we care?

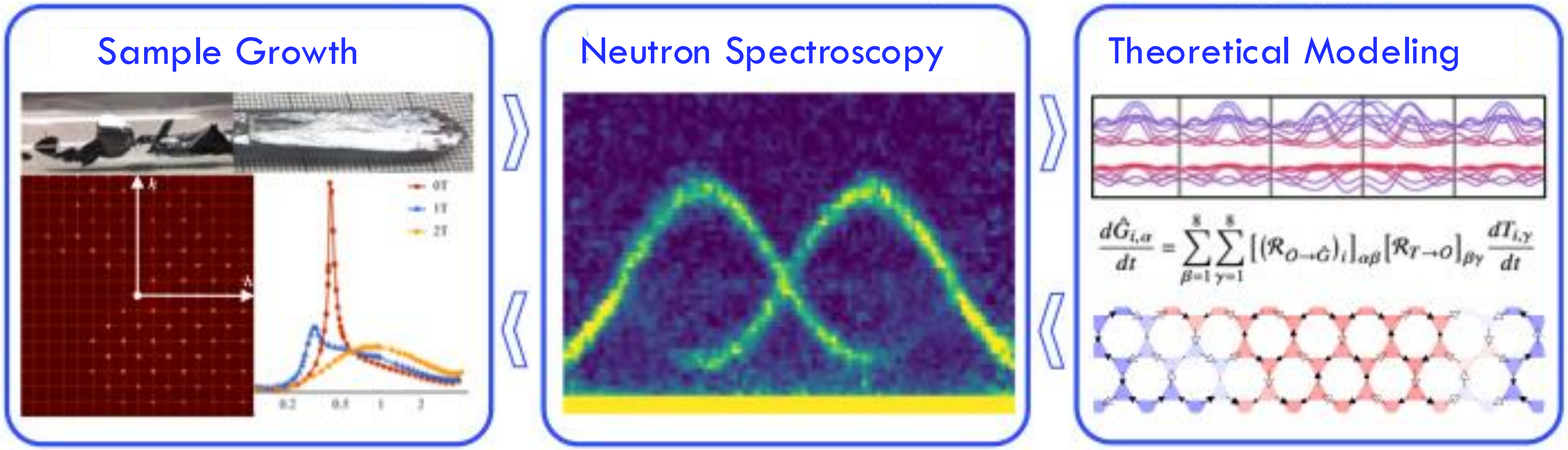
Accurate effective **models** of quantum magnets: “ground truth”



Chat GPT

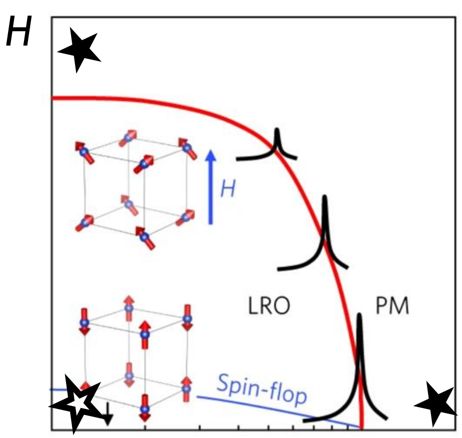
Research approach adopted by most groups

- Feeback between sample discovery/growth, spectroscopy and modeling



Other spectroscopies are complimentary and often superior: ESR, FIRMS, TDTS, 2DS

- For understanding/model extraction, often need semi-classical regime

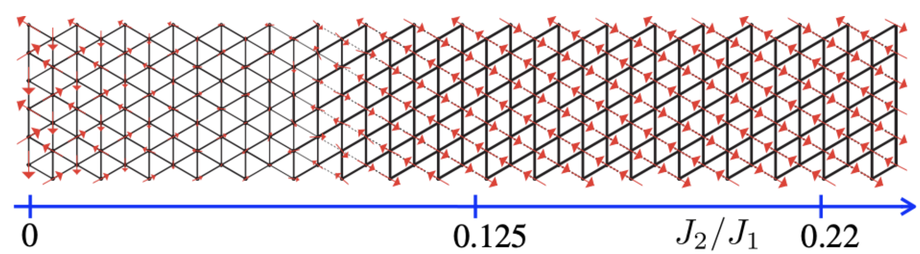


High Field

Ordered State

High Temp

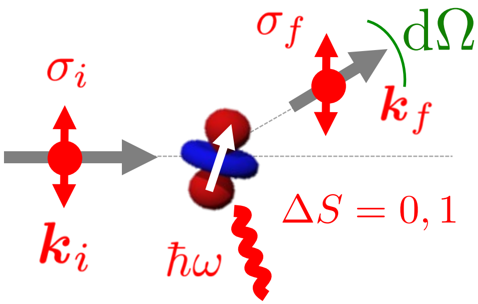
But things are changing (DMRG, MPS)



*Zhu, PRL **119**, 157201 (2015)*

Inelastic magnetic neutron scattering

- Inelastic scattering cross-section for unpolarized neutrons:



$$\boxed{\left.\frac{\mathrm{d}^2\sigma}{\mathrm{d}E_f\,\mathrm{d}\Omega}\right|_{\mathrm{mag}}} = \frac{k_f}{k_i} r_0^2 \;|gf(\mathbf{Q})|^2 \boxed{\sum_{\alpha\beta}(\delta_{\alpha\beta} - \hat{Q}_\alpha\hat{Q}_\beta)} \boxed{\mathcal{S}^{\alpha\beta}(\mathbf{Q},\omega)}$$

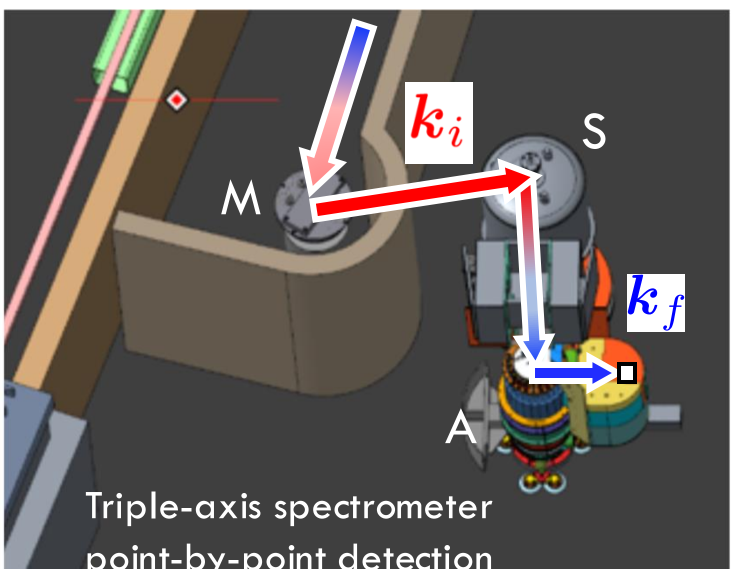
measured cross-section — magnetic form-factor — dipole factor — dynamic spin structure-factor

"Probing spins with spins"

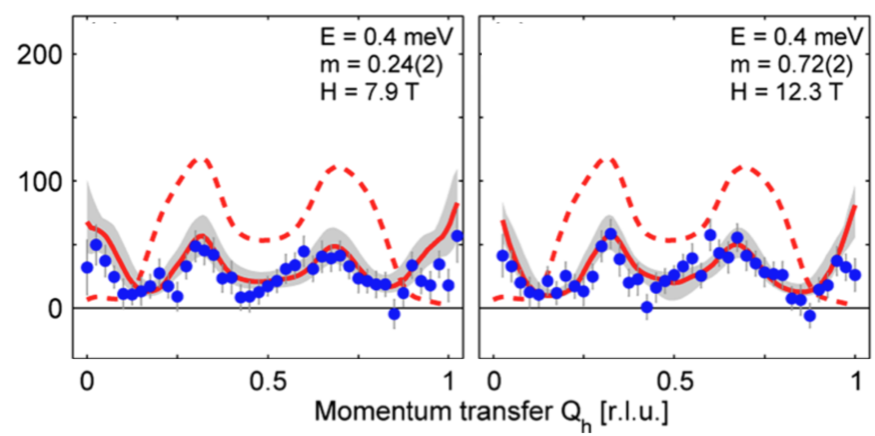
$$\mathcal{S}^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar}\int \mathrm{d}t e^{-i\omega t}\frac{1}{N}\sum_{\mathbf{r}\mathbf{r}'} e^{-i\mathbf{Q}\cdot(\mathbf{r}-\mathbf{r}')} \boxed{\langle \hat{S}^\alpha(\mathbf{r},t)\hat{S}^\beta(\mathbf{r}',0)\rangle_{Q,T}}$$

dynamic quantum pair correlations

- Instrumentation advances have transformed quantum magnetism



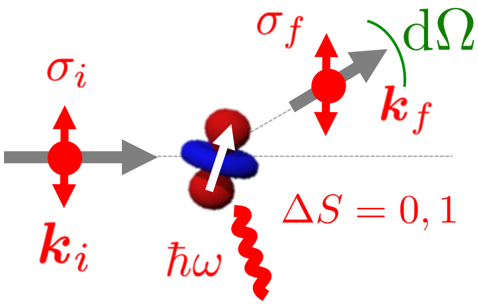
✓ ideal for restricted sample environment



Thielemann, PRL (2009)

Inelastic magnetic neutron scattering

- Inelastic scattering cross-section for unpolarized neutrons:



$$\boxed{\left.\frac{d^2\sigma}{dE_f\, d\Omega}\right|_{mag}} = \frac{k_f}{k_i} r_0^2 \; |gf(\mathbf{Q})|^2 \boxed{\sum_{\alpha\beta}(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)} \boxed{\mathcal{S}^{\alpha\beta}(\mathbf{Q},\omega)}$$

cross-section in absolute units — magnetic form-factor — dipole factor — dynamic spin structure-factor

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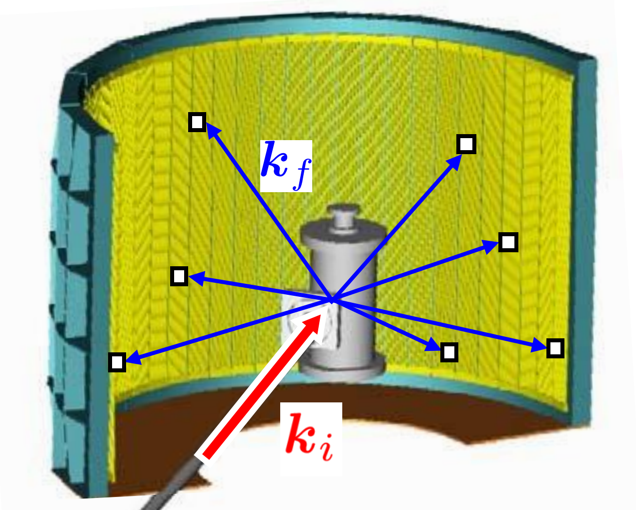
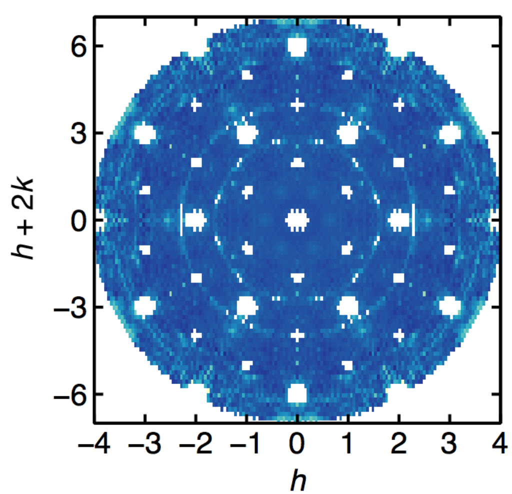
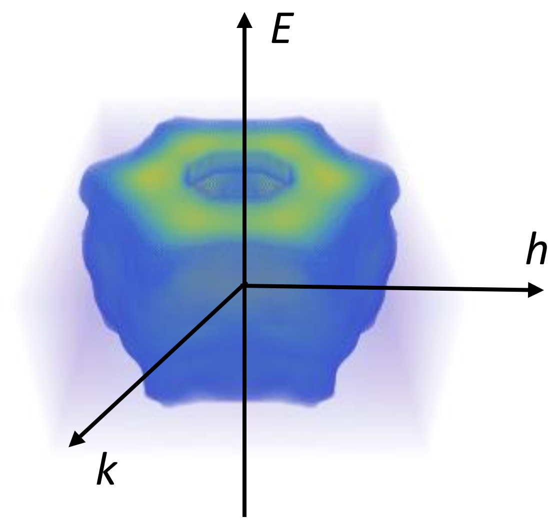


Time-of-flight spectrometer
parallelized detection





✓ allows wide surveys of momentum-energy space

Additional Neutron Scattering Tricks

- Model-free interpretation and sum-rules stem from quantitative x-section

Zeroth Moment Sum-Rule

$$\sum_{\alpha}\int_{-\infty}^{\infty} d\omega \int_{\mathrm{BZ}} d\mathbf{Q}\, \mathcal{S}^{\alpha\alpha}(\mathbf{Q},\omega) = N_{\mathrm{BZ}}\boxed{S(S+1)}.$$

Quantum Expectation Value

New

Entanglement Witnesses

Scheie PRB (2023); Laurell AQT Review (2025)

For Heisenberg systems, energy-integrated quantities directly yield expectation values

$$\mathcal{S}(\mathbf{Q}) = \int_{-\infty}^{+\infty} \mathrm{d}E\, \mathcal{S}(\mathbf{Q},E)$$

Instantaneous

$$= \frac{2}{3N}\sum_{ij}\boxed{\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle}\cos\left(\mathbf{Q}\cdot\mathbf{r}_{ij}\right)$$

Correlator

$$\mathcal{K}(\mathbf{Q}) = \int_{-\infty}^{+\infty} \mathrm{d}E\, E\, \mathcal{S}(\mathbf{Q},E)$$

First moment

$$= -\frac{1}{3N}\sum_{ij}\boxed{J_{ij}\,\langle \mathbf{S}_i \cdot \mathbf{S}_j\rangle}\left[1-\cos(\mathbf{Q}\cdot\mathbf{r}_{ij})\right]$$

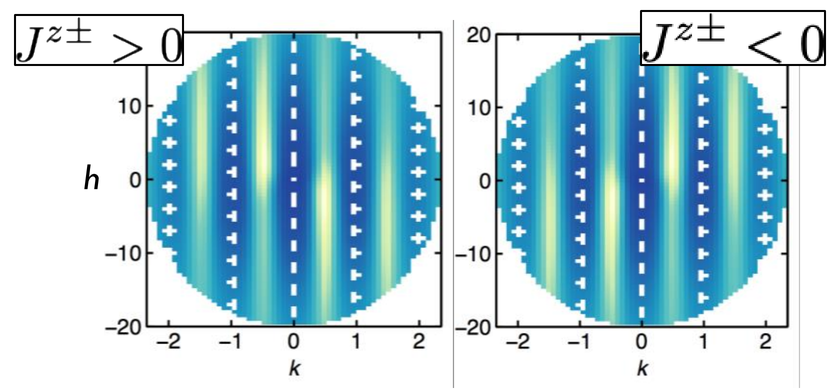
Bond Energy

Hohenberg PRL (1974)
Broholm PRL (2001)

- Polarization and dipole factor

$$\boxed{\sum_{\alpha\beta}(\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta)}\ \mathcal{S}^{\alpha\beta}(\mathbf{Q},\omega)$$

Dipole factor: couples spins and momentum



Paddison PRL (2020)

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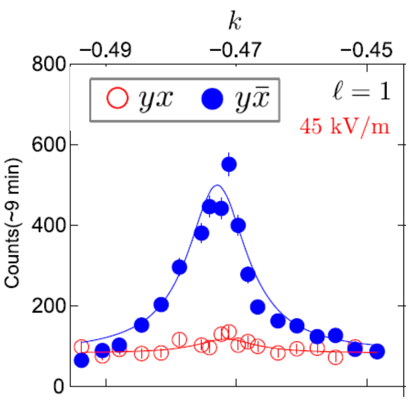
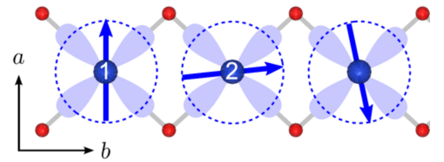
Dipole factor: couples spins and momentum

Polarized neutrons: can measure any component





Mourigal PRB (2011)

The energy resolution gap

- Energy resolution

Typical energy resolution is $\delta E / E_{i(f)} \approx \boxed{2-5\%}$ $E_i = 3\ \mathrm{meV} \rightarrow \boxed{\delta E = 60(0) - 150(0)\mu\mathrm{eV}}$

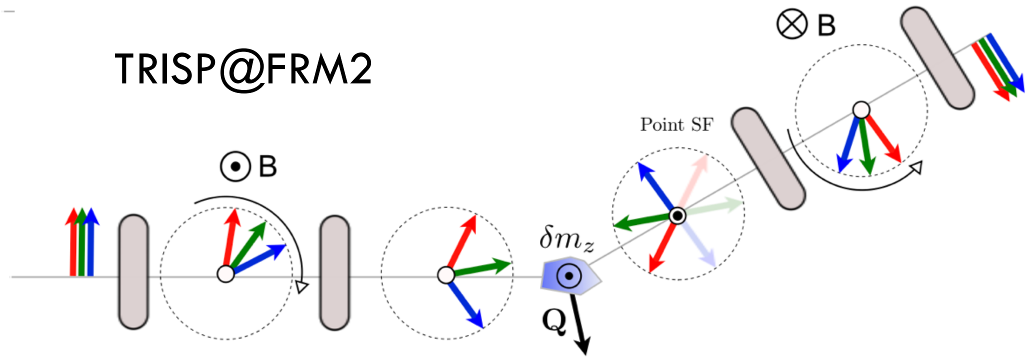
It's a problem

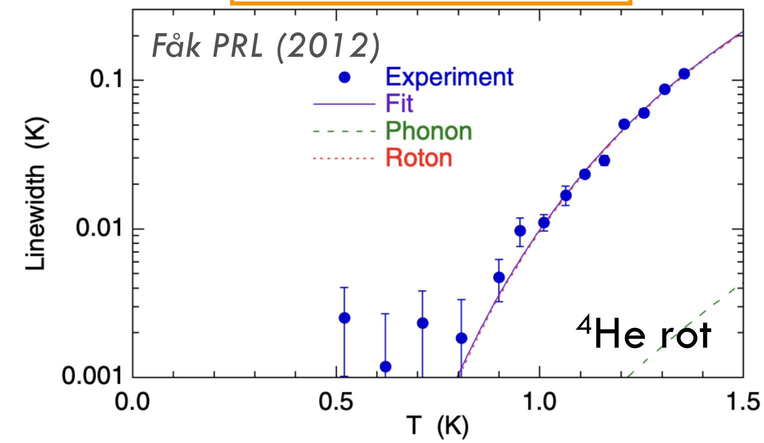
resolution gap

$\boxed{\delta E = 0.1 - 10\mu\mathrm{eV}}$

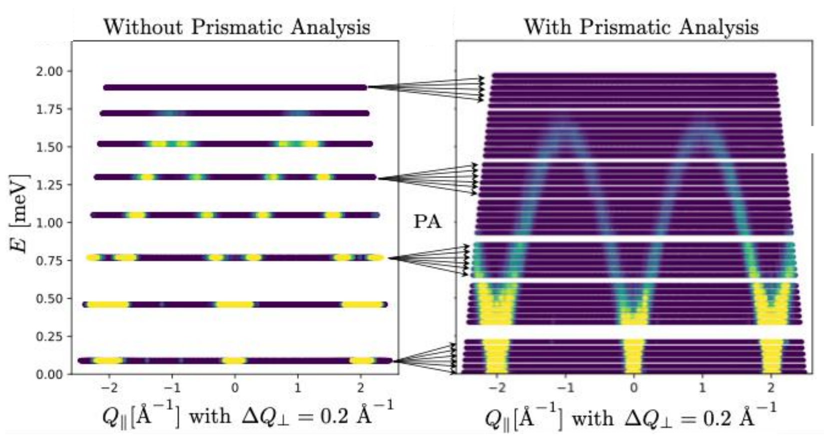
- Resonant spin-echo

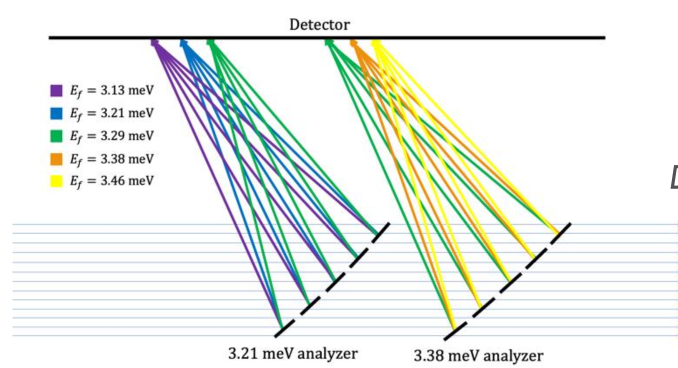
TRISP@FRM2





- Prismastic-analysis spectrometers





CAMEA@PSI

Desai RSA (2023)
Desai NIMA (2024)

Modeling

- Semi-classical approaches

Spin-wave theory

$$\sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} b^\dagger_{\mathbf{k}} b_{\mathbf{k}} + \frac{1}{3!}\sum_{\{\mathbf{k}_i\}} V_{31}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3)\left[b^\dagger_{\mathbf{k}_1} b^\dagger_{\mathbf{k}_2} b_{\mathbf{k}_3} + \mathrm{h.c}\right] + \ldots$$

cubic vertex

$$+ \frac{1}{4}\sum_{\{\mathbf{k}_i\}} V_{40}(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4)\left[b^\dagger_{\mathbf{k}_1} b^\dagger_{\mathbf{k}_2} b_{\mathbf{k}_3} b_{\mathbf{k}_4}\right] + \ldots$$

quartic vertex

Good: quasiparticle-based, interpretability

Bad: need right spin configuration, finite temperature

Classical Dynamics in a Thermal Bath

$$\frac{d\mathbf{S}}{dt} = -\mathbf{S} \times \left[\xi + \frac{dH}{d\mathbf{S}} - \lambda\left(\mathbf{S} \times \frac{dH}{d\mathbf{S}}\right)\right]$$

noise   coupling to bath

Good: finite temperature, classical non-linearities

Bad: stochastic nature, classical thermal bath

This has been generalized to multipolar degrees of freedom

Su(n)ny

arXiv:2501.13095

- Full quantum approaches

  Matrix Product States

  Quantum Monte-Carlo, Exact Diagonalization

- Emerging/Exotic

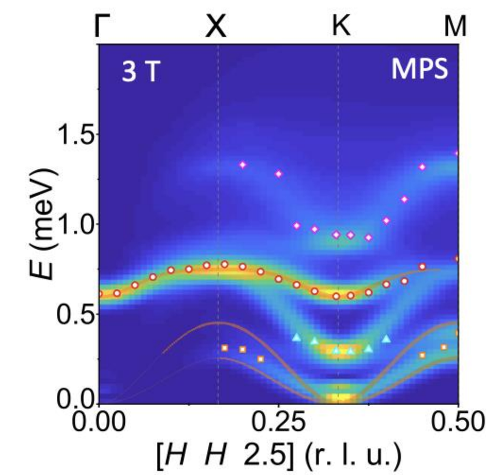
  Truncated Hilbert Space Exact Diagonalization

  LSWT-Kernel Polynomial Method

  Lane Scipost (2025)

  Beyond Mean-Field Parton Decomposition

Ghioldi PRB (2018), Zhang (2022), Willsher (2025)



Nikitin, Läuchli npj QM (2023)

Outline

- ❑ Heavy Introduction
  - Quantum Magnetism Big Picture
  - Research Approach
  - Inelastic Neutron Scattering
  - Modeling

2

- ❑ Examples: Greatest Hits 2012-2025
  - Spinons: Heisenberg spin-1/2 chains in copper sulfate
  - Magnons: Square-lattice problems
  - Bound-states: Triangular uniaxial spin-1 system FeI_2
  - **C**ontinuous excitations does not always mean fractionalization/decay

- ❑ Conclusion (maybe)

Outline

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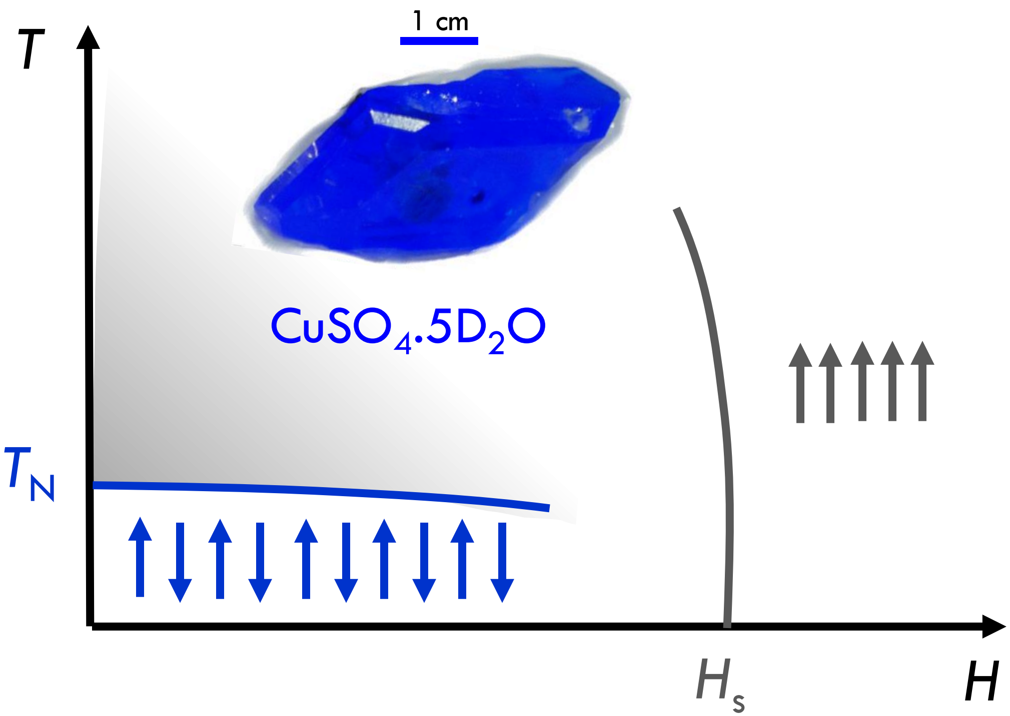
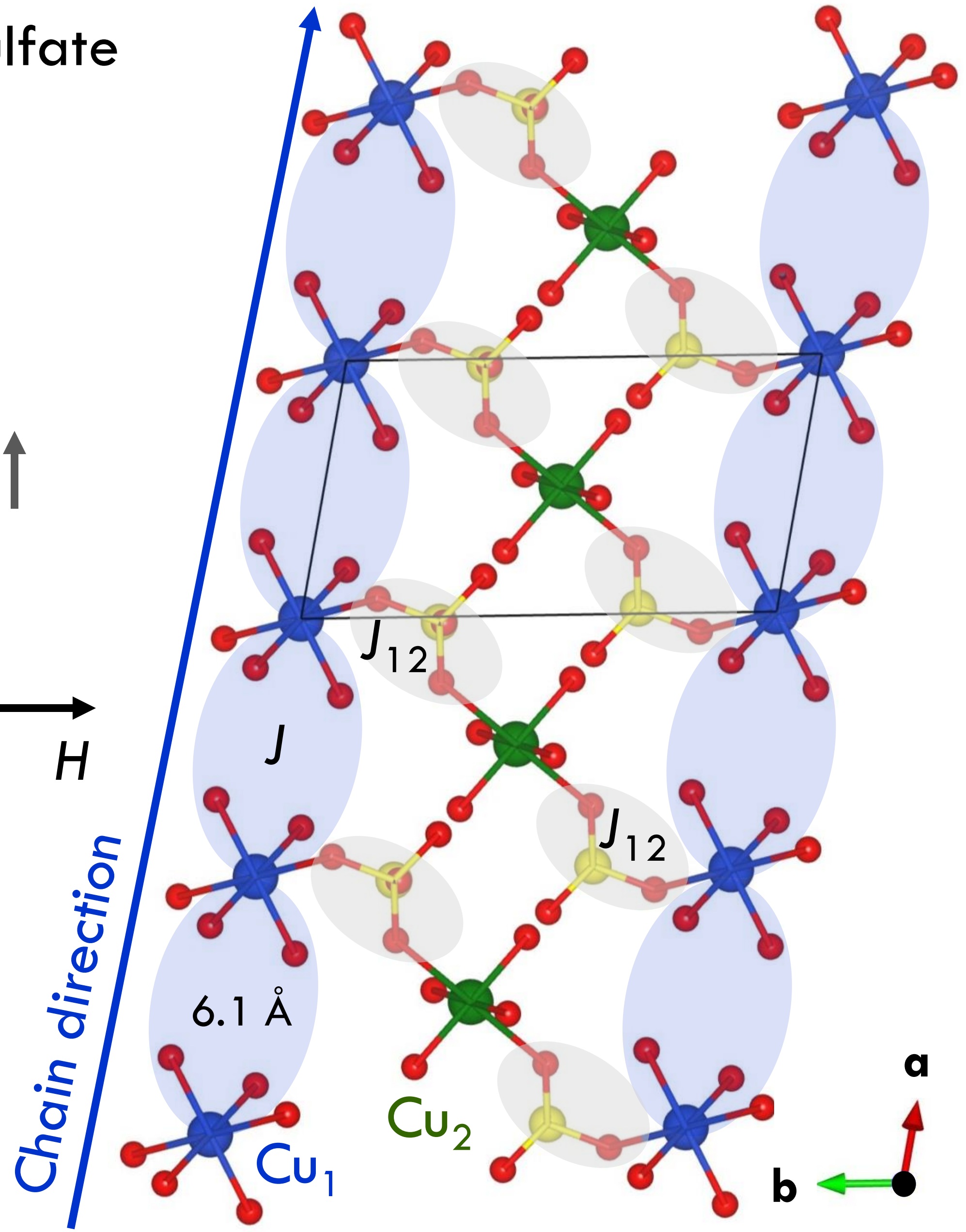
- ❑ Examples: Greatest Hits 2012-2025
  - Spinons: Heisenberg spin-1/2 chains in copper sulfate

Mourigal, Enderle, Rønnow, Caux, Nature Physics **9**, 435-441 (2013)

Weakly coupled Heisenberg spin-1/2 chains

- Thermo-magnetic props. of Cu-sulfate



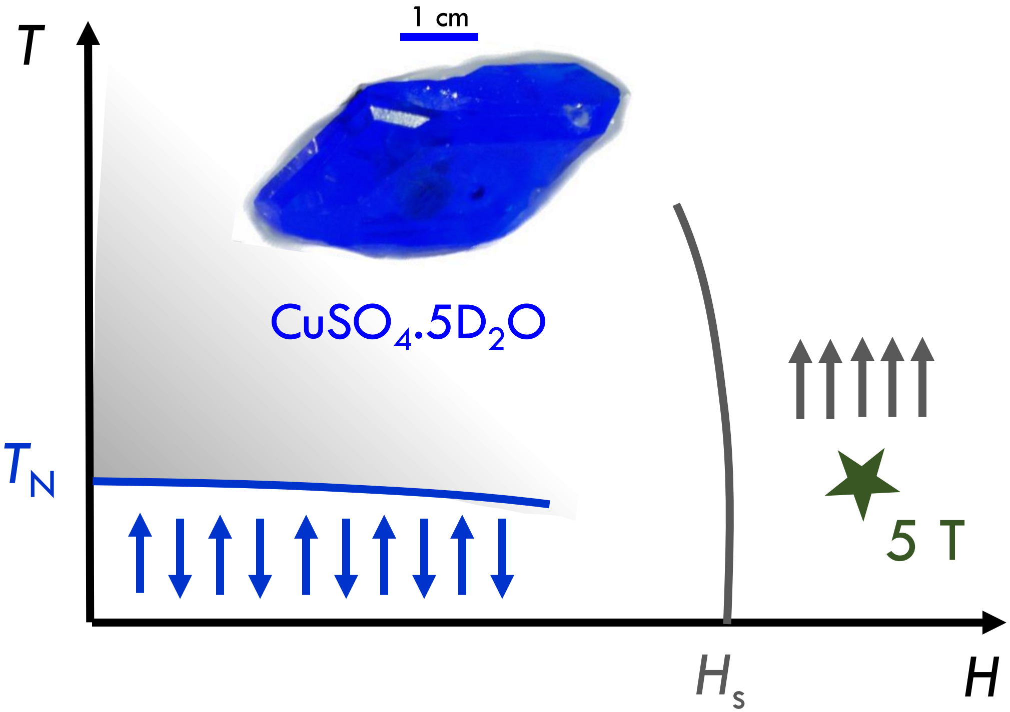


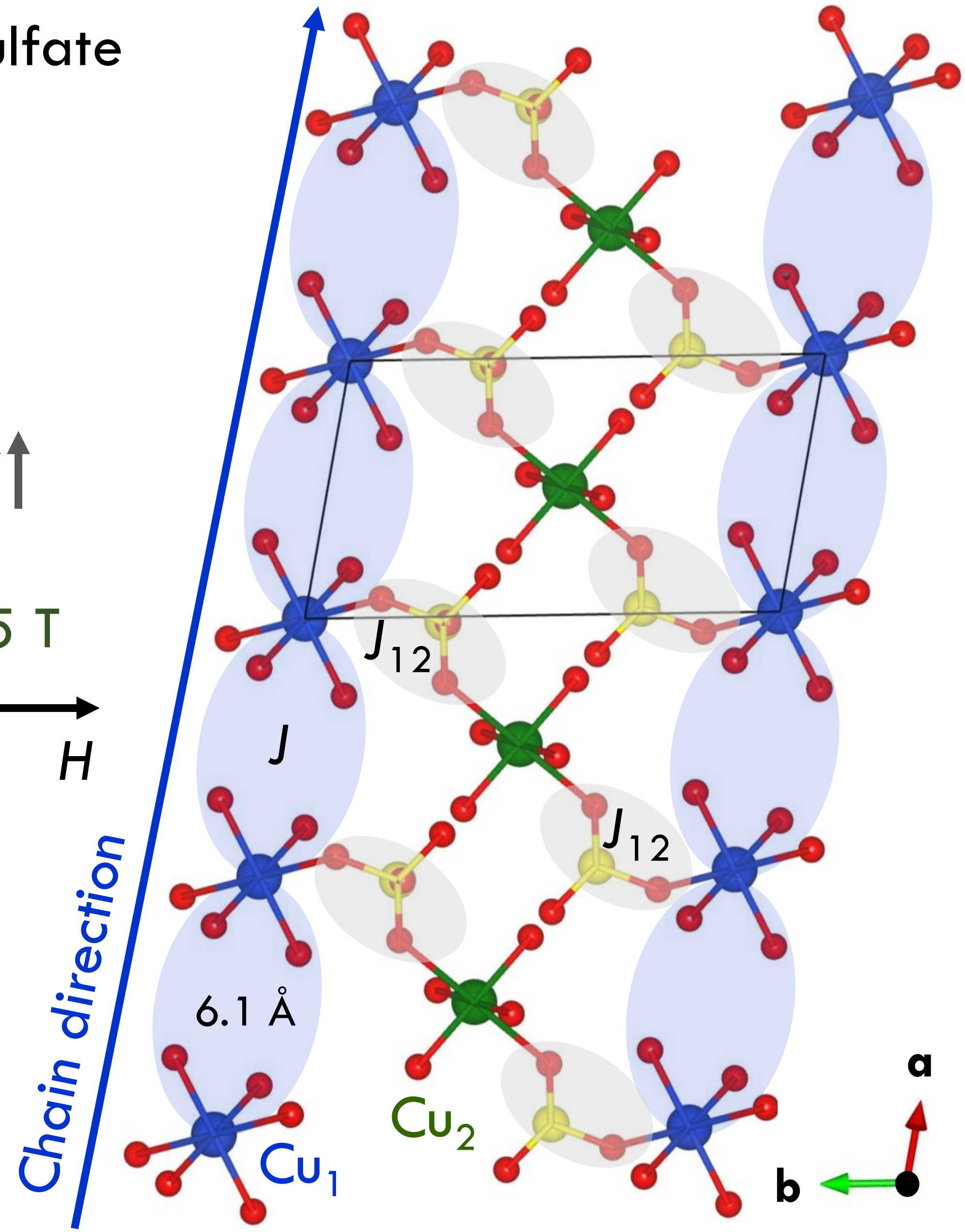
Spin ordering $T_N = 0.092$ K

Spin saturation $H_s = 3.62$ T

Weakly coupled Heisenberg spin-1/2 chains

- Thermo-magnetic props. of Cu-sulfate



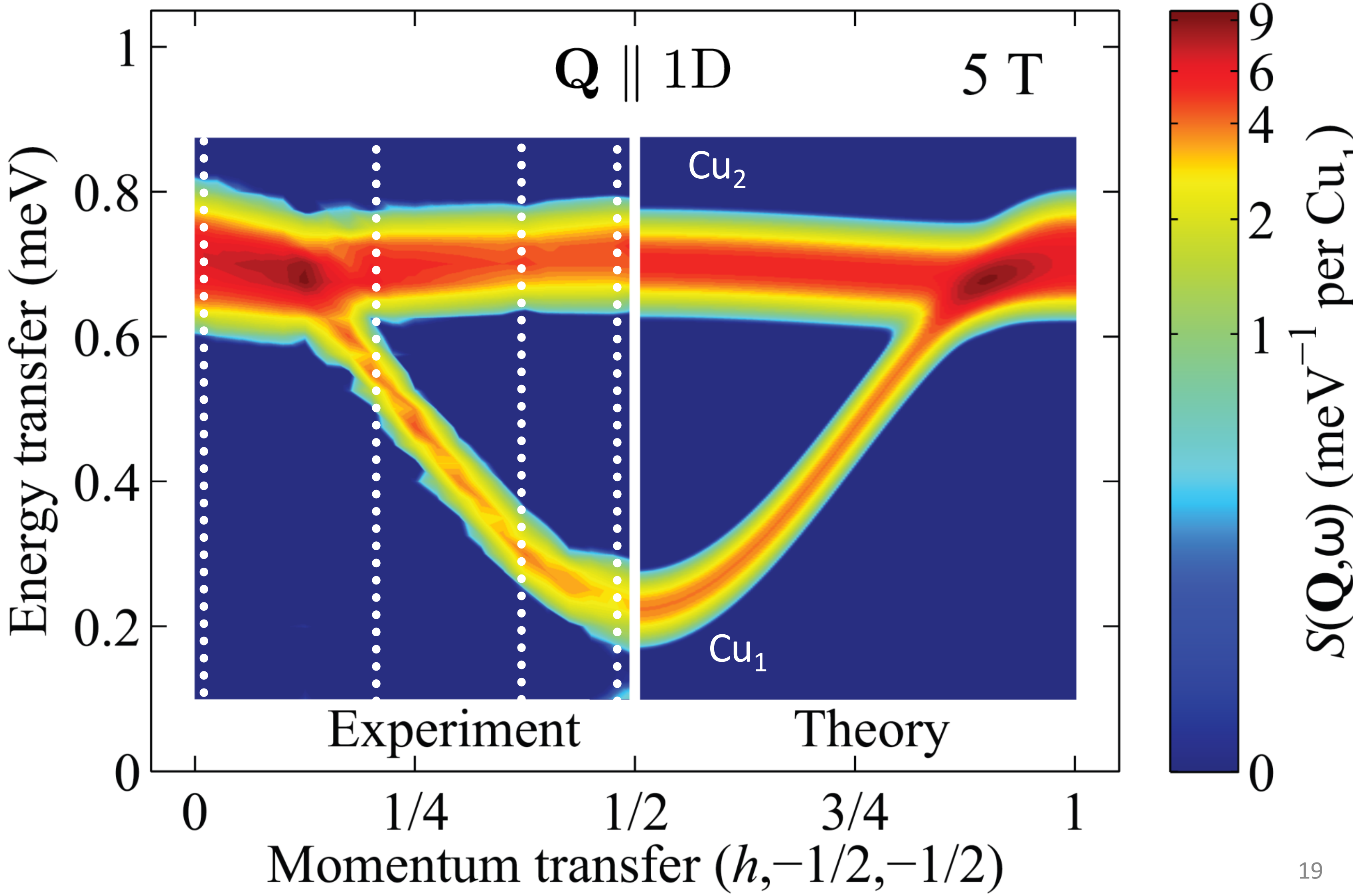


Spin ordering $T_N = 0.092$ K

Spin saturation $H_s = 3.62$ T

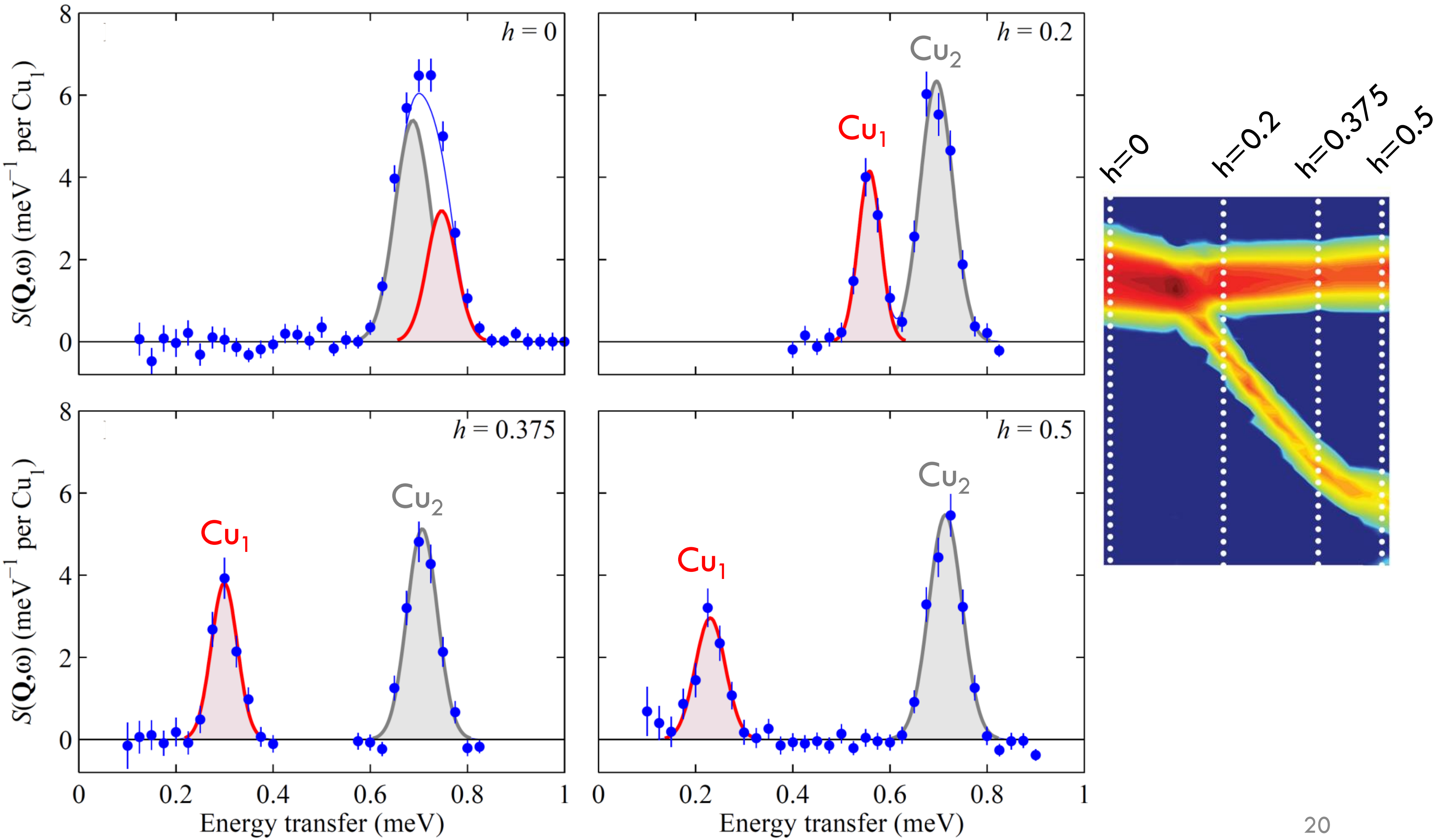
Magnon dispersion in the field polarized state

- Triple-axis experiment



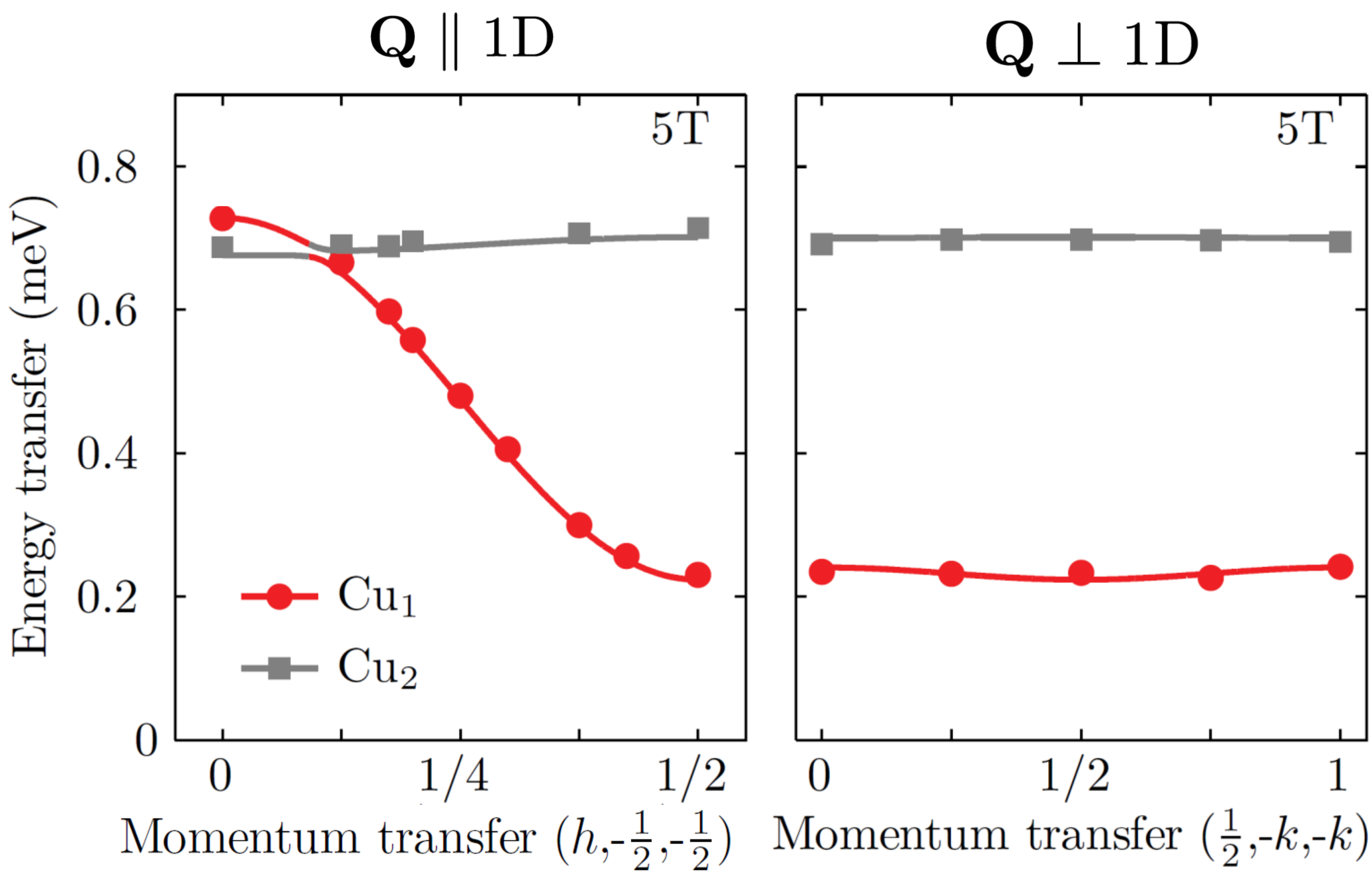
Quantitative analysis of scattering intensity

- Resolution limited magnon peaks



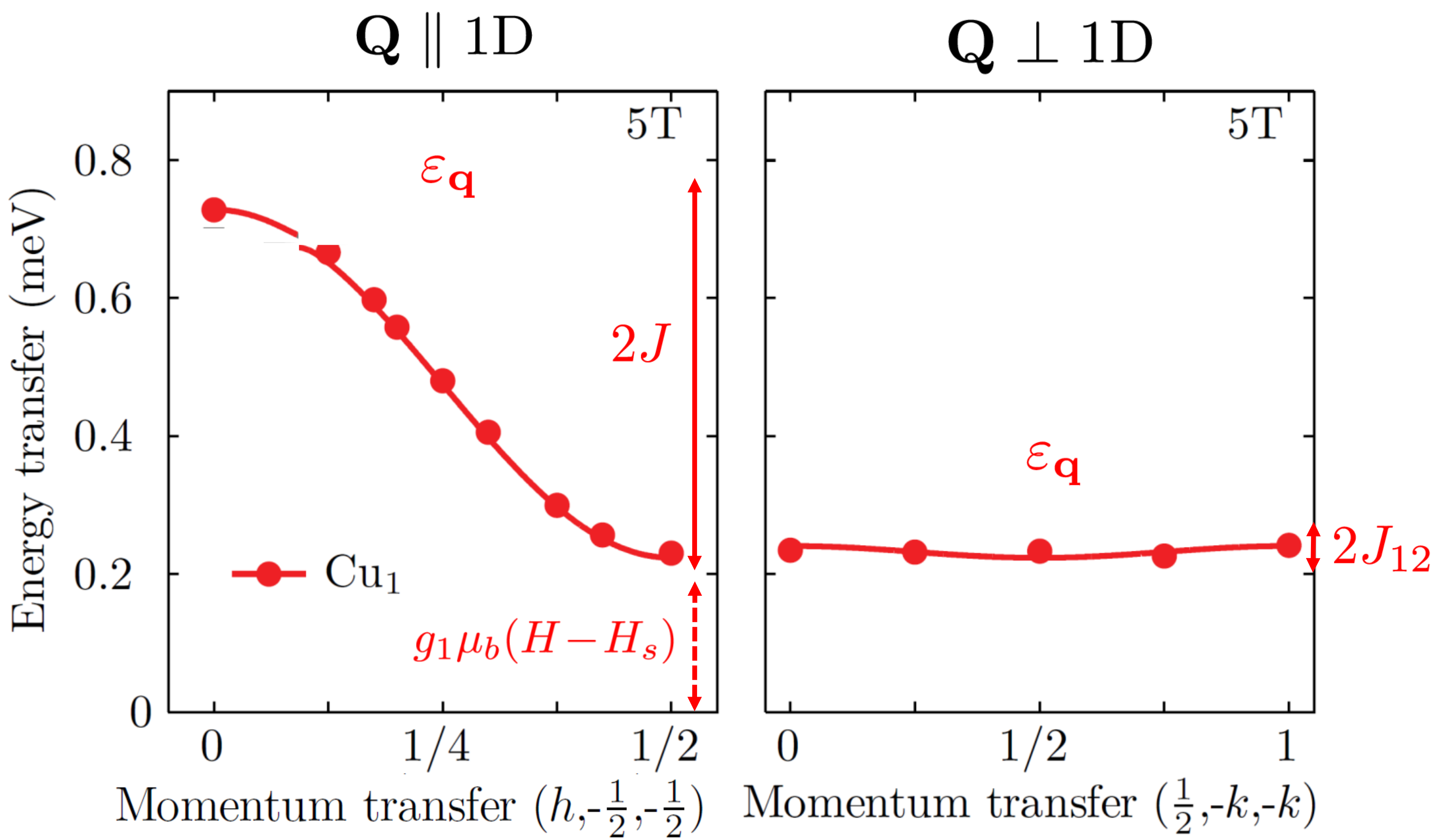
Quantitative analysis above saturation

- Eigenvalues from dispersion relation



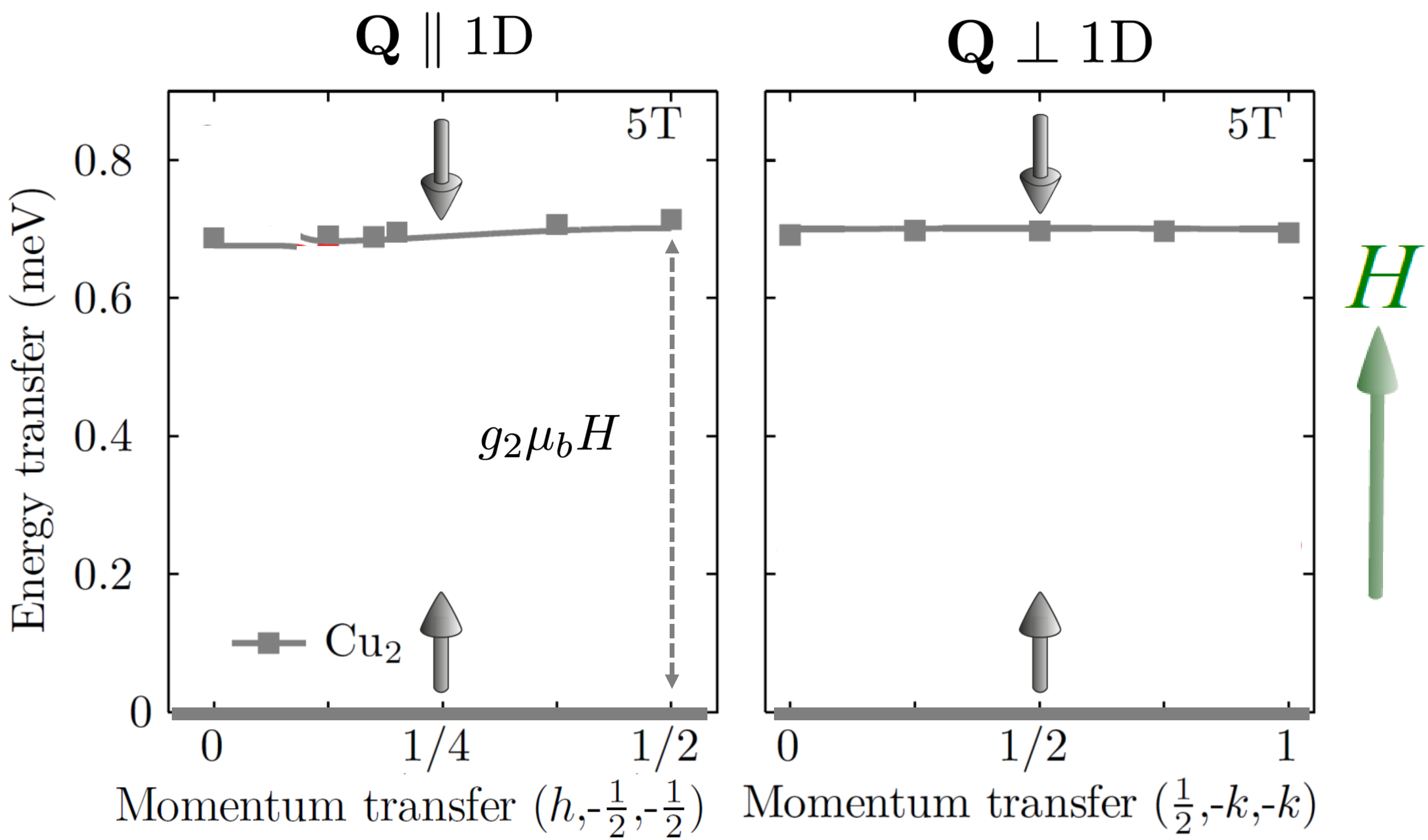
Quantitative analysis above saturation

- Quasi-one-dimensionality of Cu_1 chains



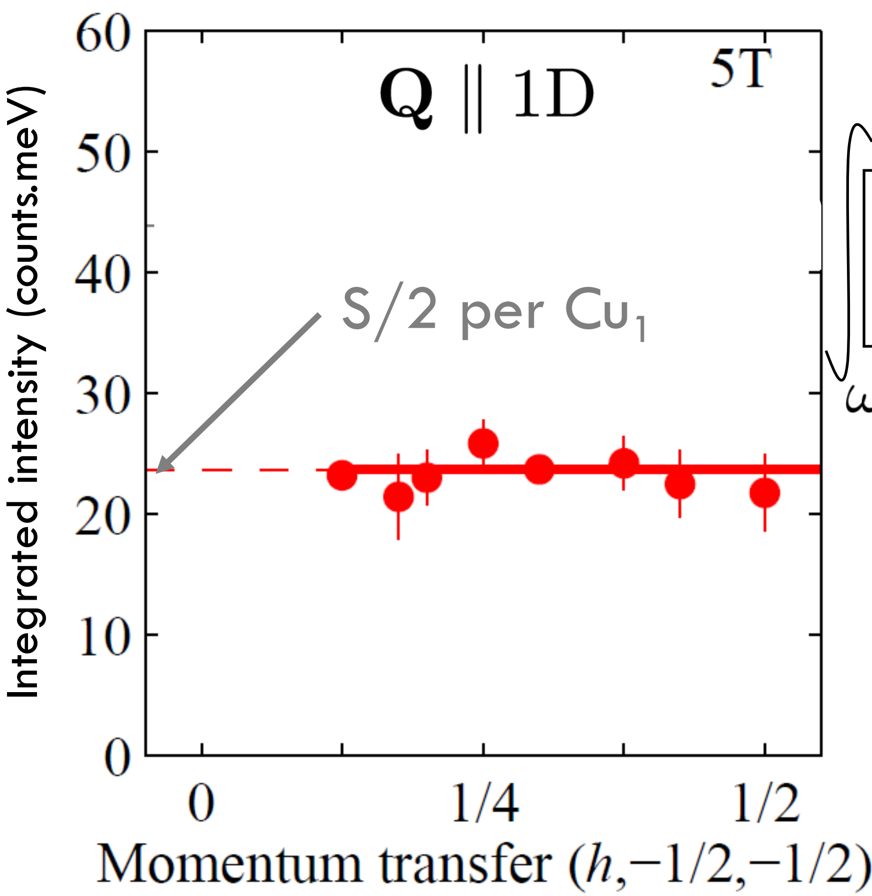
Quantitative analysis above saturation

- Quasi-isolated Cu_2 sites



Quantitative analysis above saturation

- Momentum-independent integrated intensity
- Self absolute normalization of measured intensities



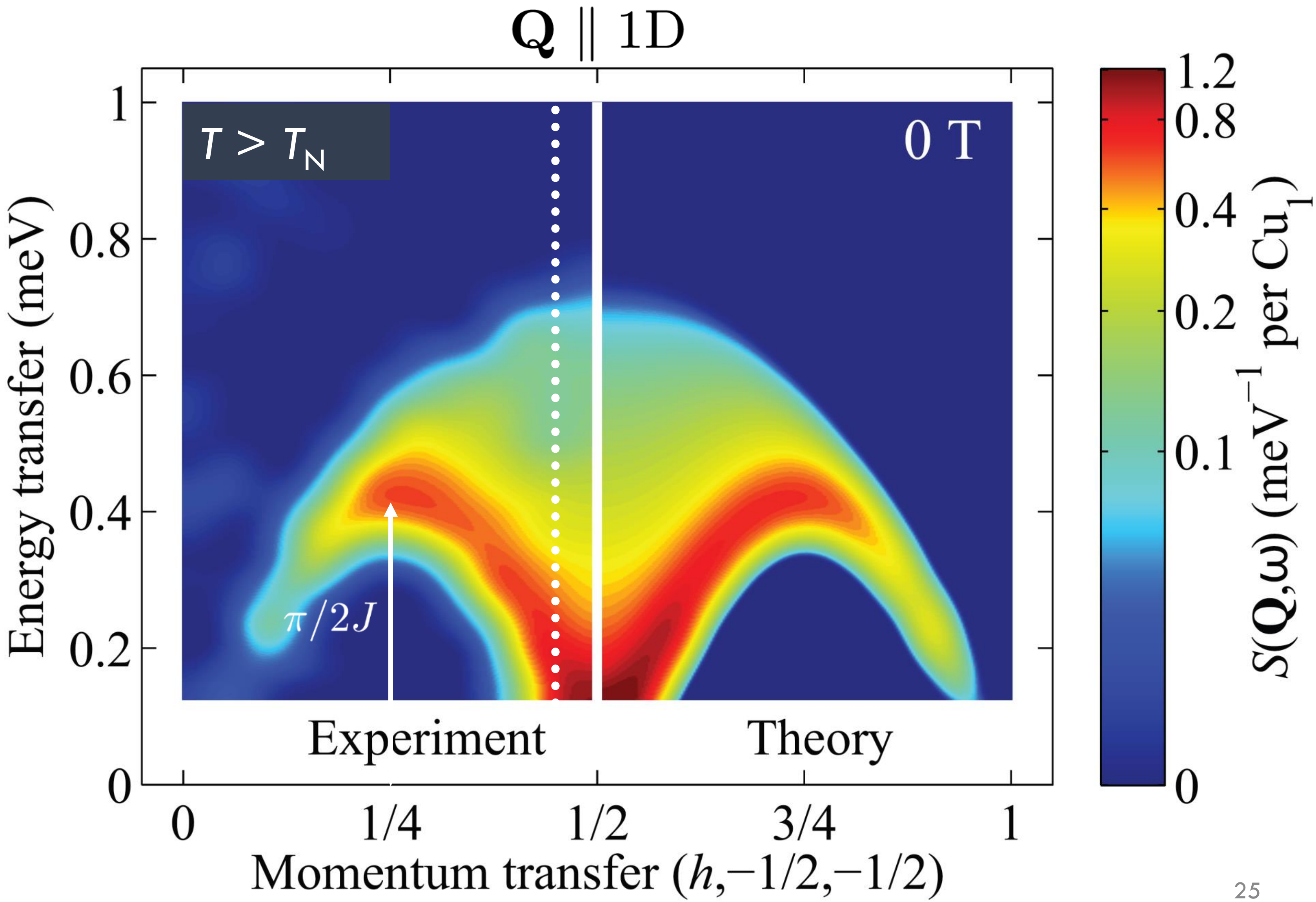
Measured intensity | Sample and instrument | Known exactly

$$\int_\omega \boxed{\frac{k_i}{k_f}\frac{\mathrm{d}^2\sigma}{\mathrm{d}E_f\,\mathrm{d}\Omega}} = I_0 r_0^2 |g f(\mathbf{Q})|^2 \int_\omega \boxed{\mathcal{S}^{yy}(\mathbf{Q},\omega)}$$

$$\mathcal{S}^{yy}(\mathbf{Q},\omega) = \boxed{\frac{S}{2}\delta(\hbar\omega - \varepsilon_{\mathbf{q}})}$$

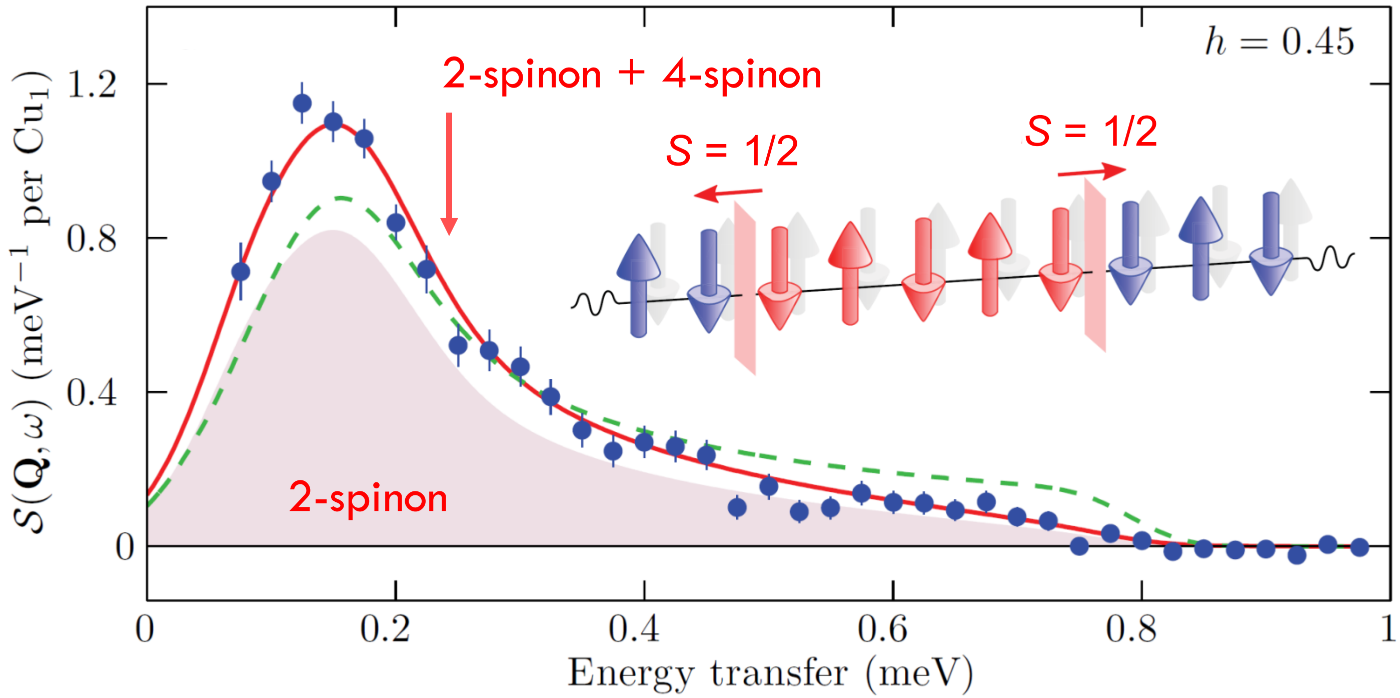
Spinon continuum in the zero-field state

- Triple-axis experiment



Quantitative analysis in zero field

- Absolute self normalization

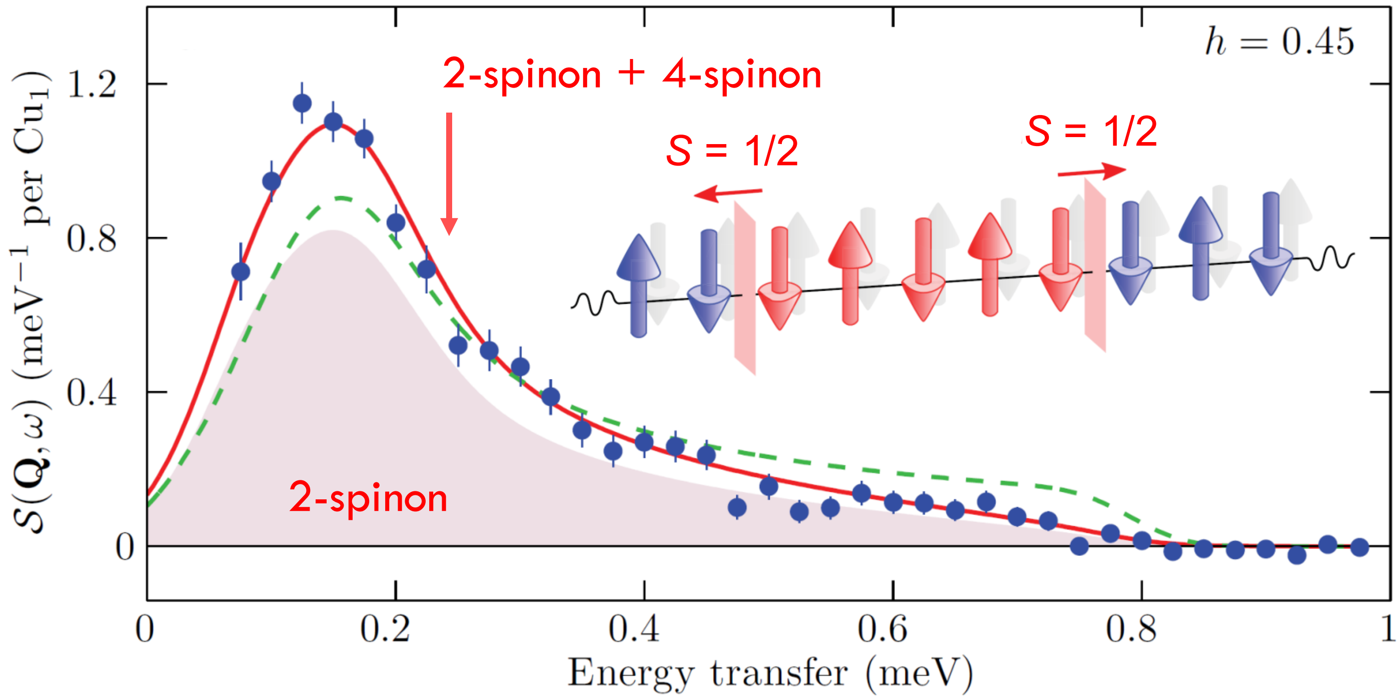


Known from 5T

Measured intensity $\boxed{\frac{k_i}{k_f}\frac{\mathrm{d}^2\sigma}{\mathrm{d}E_f\mathrm{d}\Omega}} = I_0 r_0^2 |g f(\mathbf{Q})|^2 \boxed{2\mathcal{S}^{yy}_{2+4}}$ Known exactly

Quantitative analysis in zero field

- Absolute self normalization



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Measured intensity $\boxed{\frac{k_i}{k_f}\frac{\mathrm{d}^2\sigma}{\mathrm{d}E_f\mathrm{d}\Omega}} = I_0 r_0^2 |g f(\mathbf{Q})|^2 \boxed{2S^{yy}_{2+4}}$ Known exactly

Model and experimental intensity in agreement
with a scaling factor 0.99(8)

Outline

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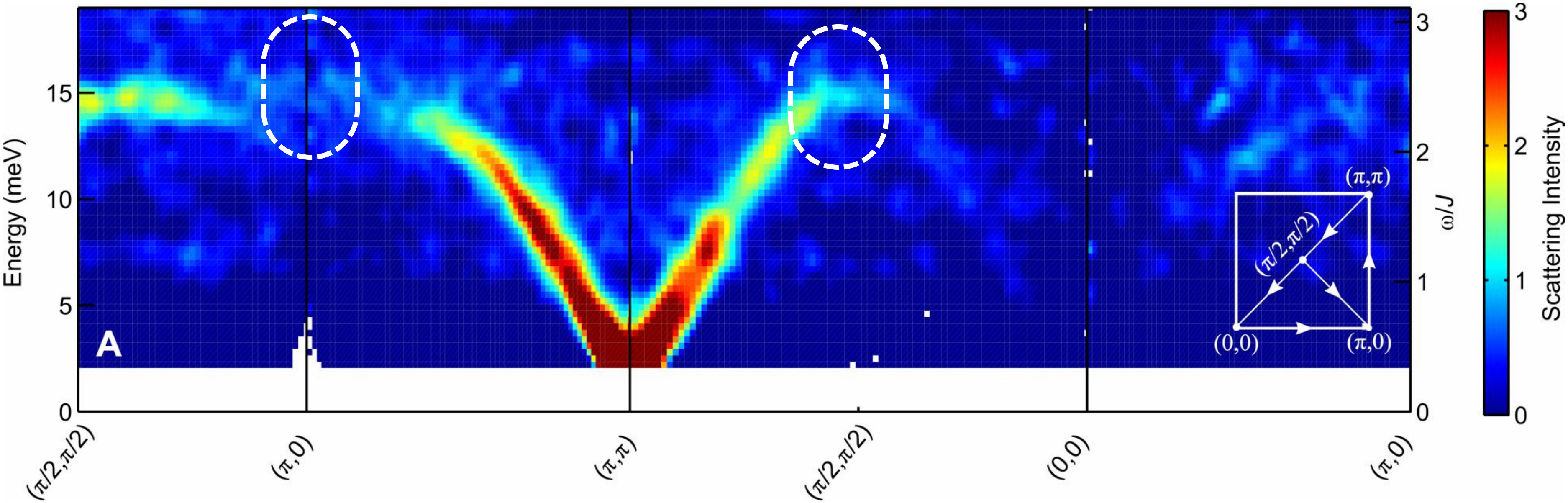
❑ Examples: Greatest Hits 2012-2025

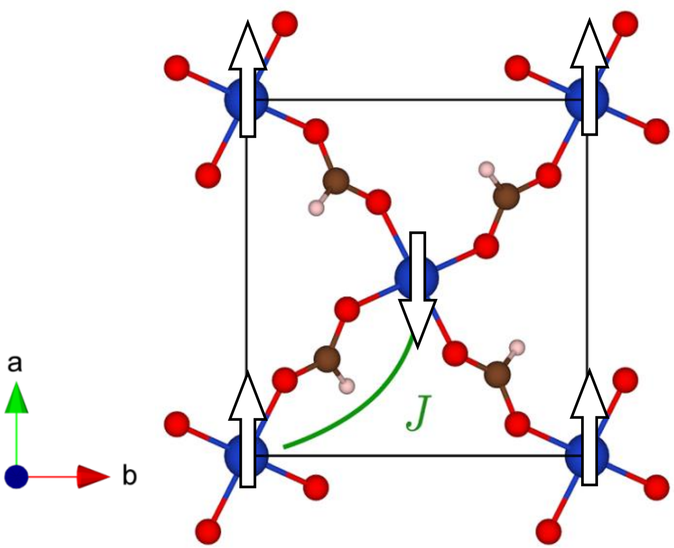
- Magnons: Square-lattice problems

Dalla Piazza, Mourigal, Enderle, Rønnow, Nature Physics **11**, 62-68 (2015)

A spin-1/2 square-lattice Heisenberg AFM

- ❑ Known for zone-boundary spin-wave anomaly at (π,0)

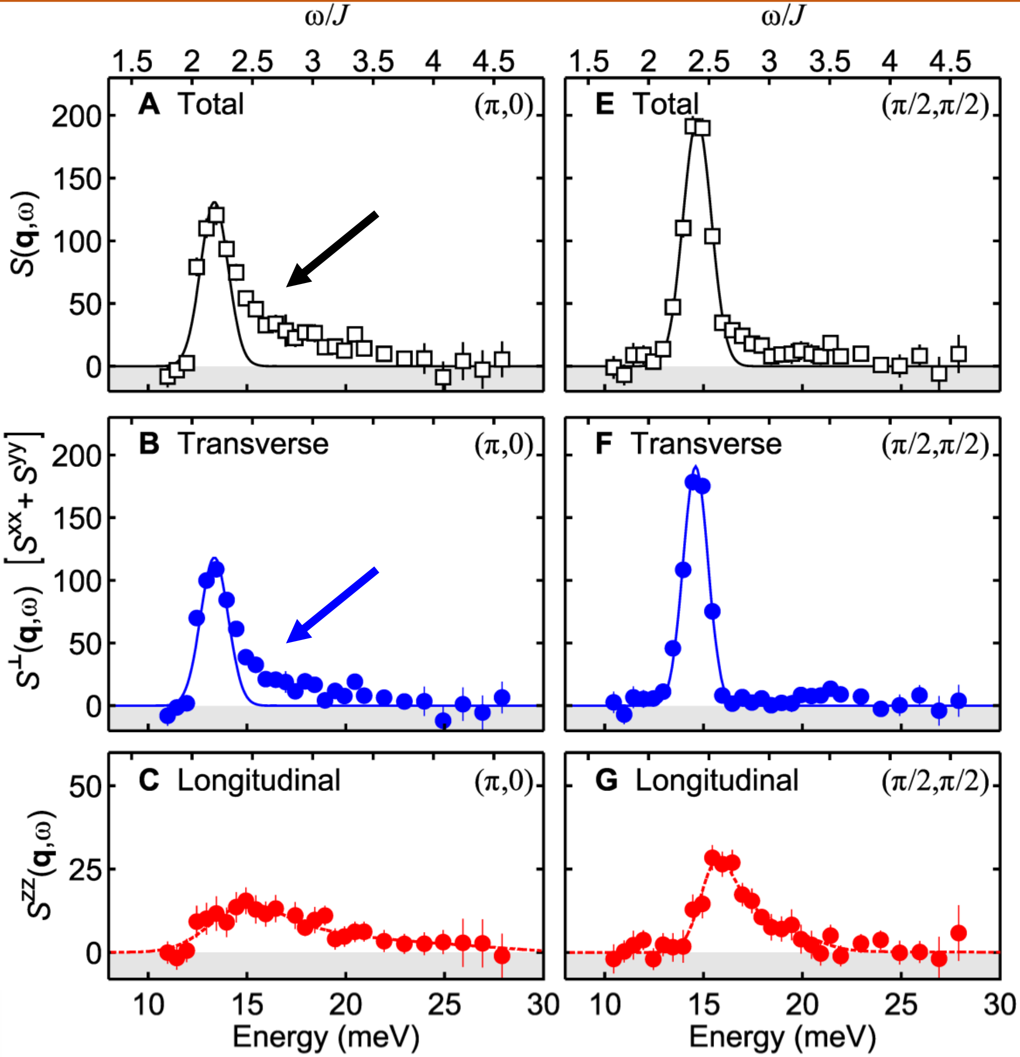




$Cu(DCOO)_2.4D_2O$ "CFTD"

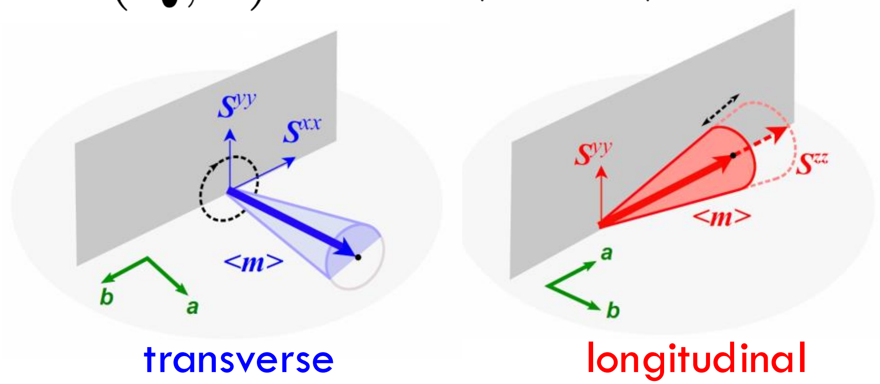
Néel order T_N = 16.5 K with 0.48 μ_B along ***a***

Insights from neutron polarization analysis



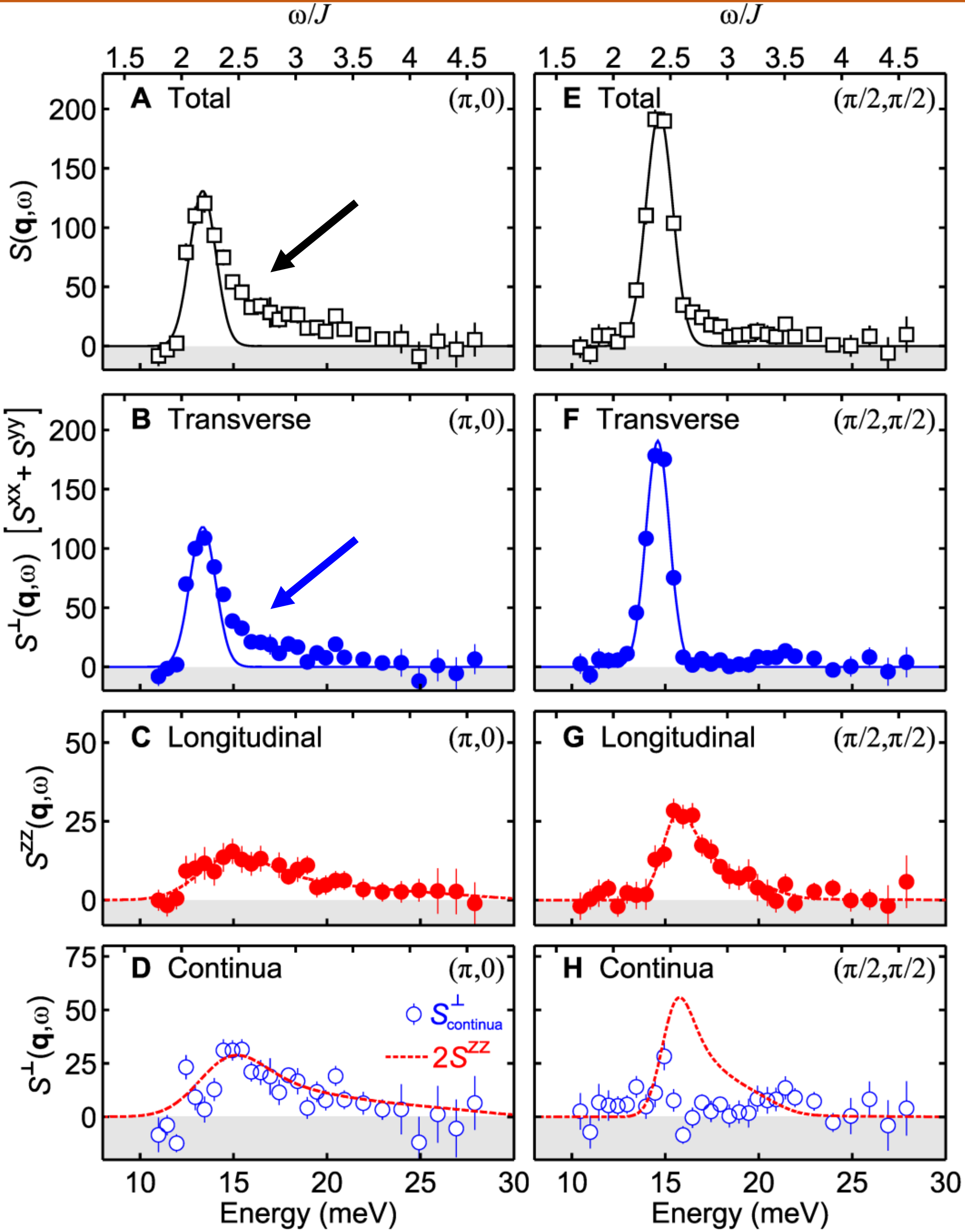
- Spin-space components

$$\mathcal{S}(\mathbf{Q},\omega) = \mathcal{S}^{xx} + \mathcal{S}^{yy} + \mathcal{S}^{zz}$$



can be separated

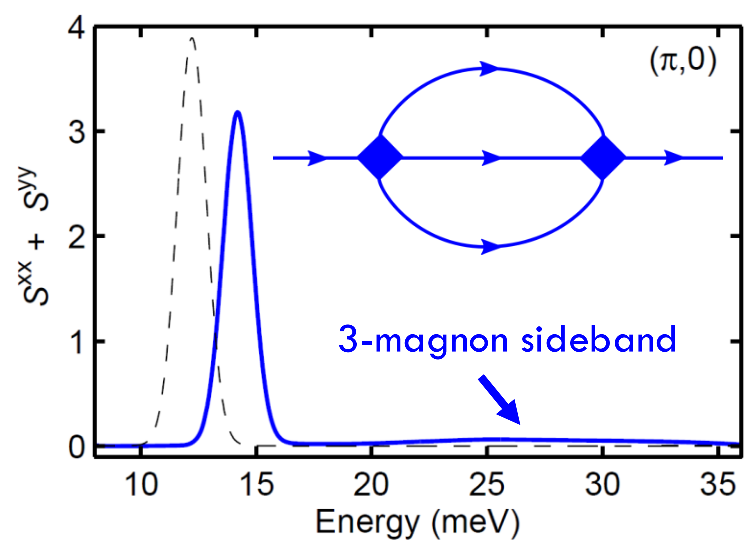
Insights from neutron polarization analysis



- ❑ Spin-space components

$$\mathcal{S}(\mathbf{Q},\omega) = \mathcal{S}^{xx} + \mathcal{S}^{yy} + \mathcal{S}^{zz}$$

- ❑ Transverse continuum

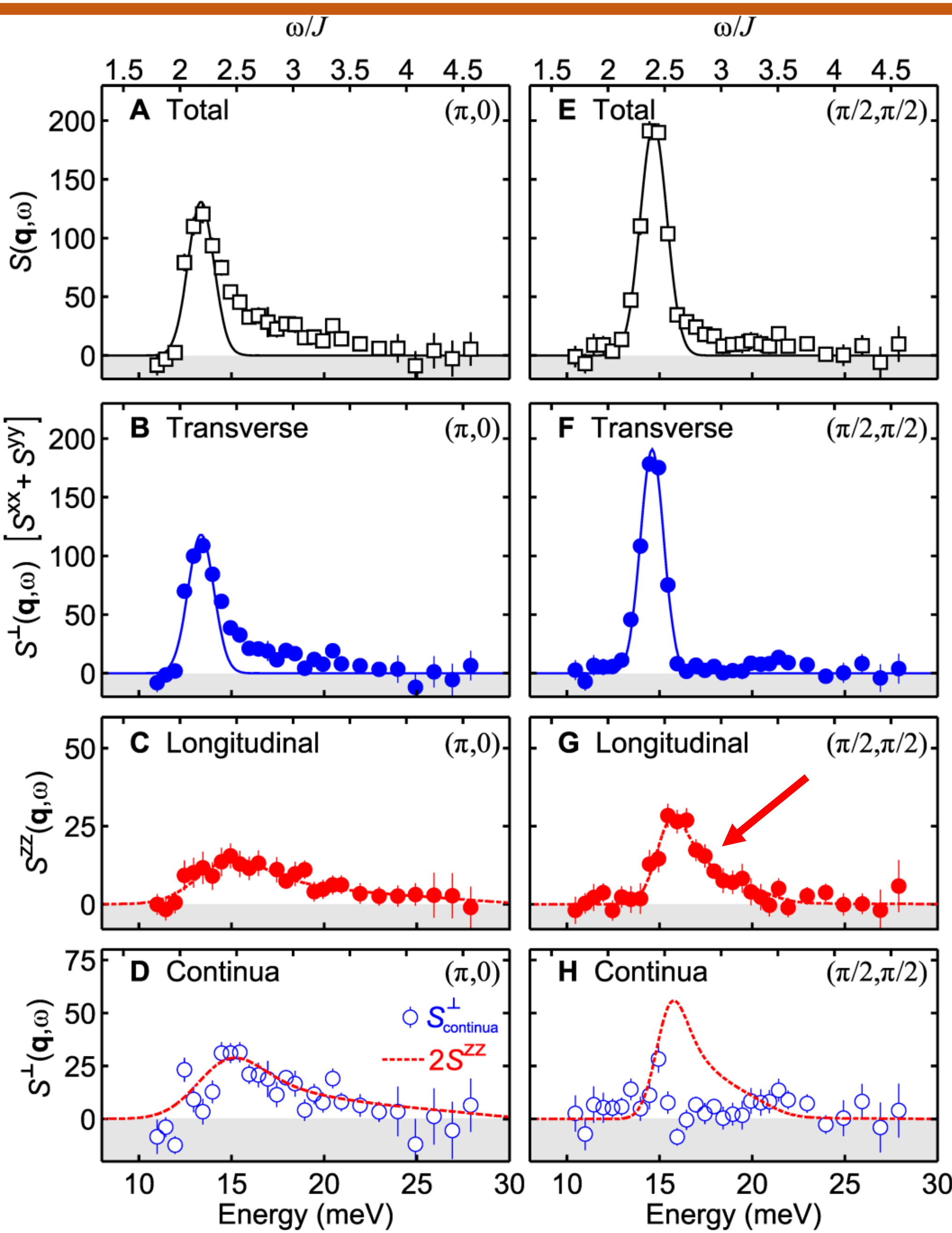


$1/S^2$ spin-wave theory (with MZ)

- ❑ Spin-isotropic continuum

$$\mathcal{S}_c^{xx} + \mathcal{S}_c^{yy} = 2\mathcal{S}^{zz}$$

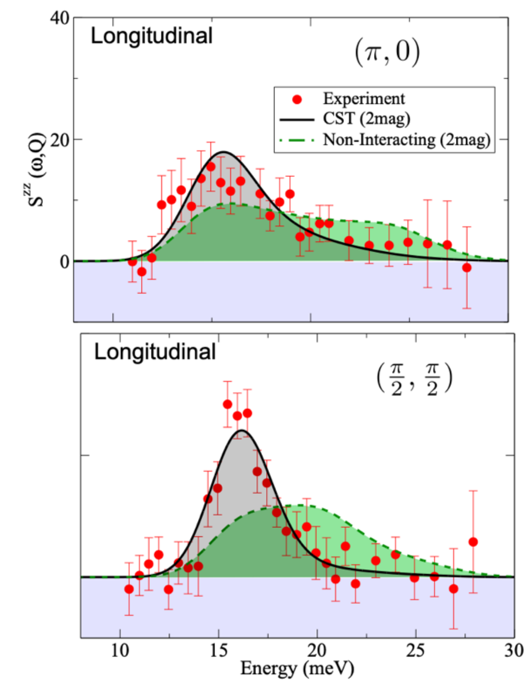
Insights from neutron polarization analysis



- Spin-space components

$$\mathcal{S}(\mathbf{Q},\omega) = \mathcal{S}^{xx} + \mathcal{S}^{yy} + \mathcal{S}^{zz}$$

- Longitudinal continuum



Mutually attracting magnons
(non-perturbative)

Powalski, Uhrig, Scipost (2018)

Field dependence and decays

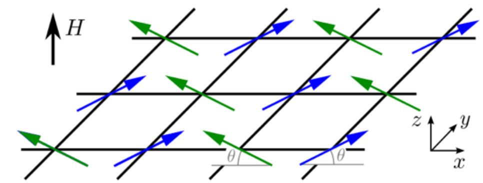
59. arXiv:1006.4838 [pdf, ps, other] cond-mat.str-el doi 10.1103/PhysRevB.82.144402

Field-induced decay dynamics in square-lattice antiferromagnet

Authors: M. Mourigal, M. E. Zhitomirsky, A. L. Chernyshev

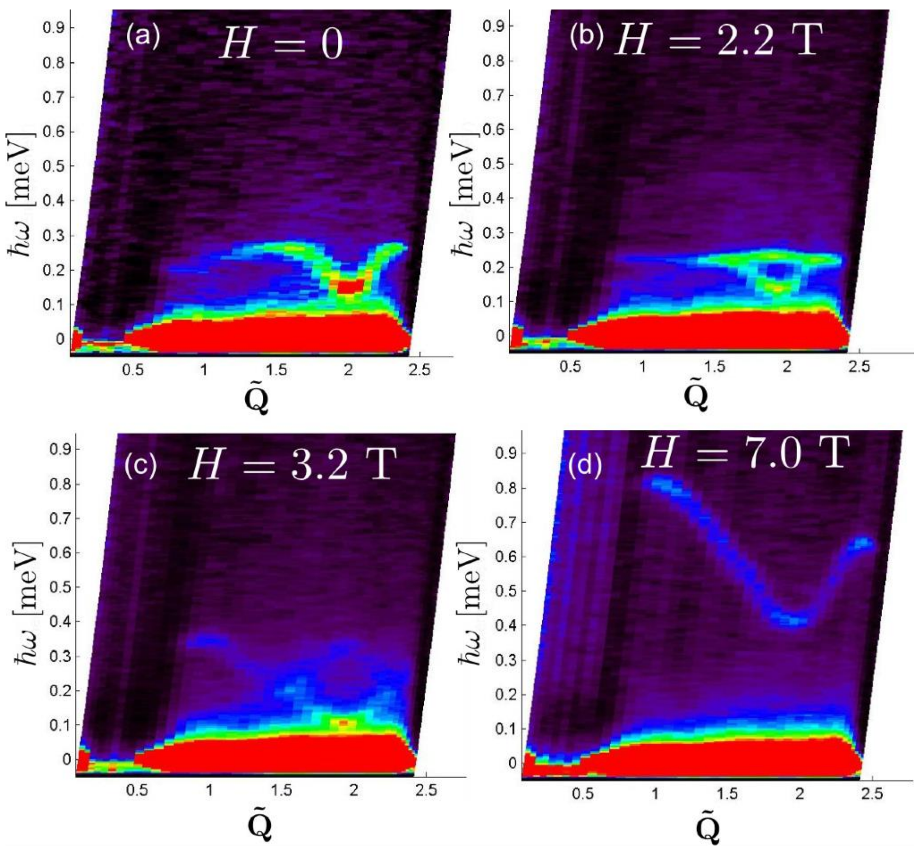
Abstract: Dynamical properties of the square-lattice Heisenberg antiferromagnet in applied magnetic field are studied for arbitrary value S of the spin. Above the threshold field for two-particle decays, the standard spin-wave theory yields singular corrections to the excitation spectrum with logarithmic divergences for certain momenta. We develop a self-consistent approximation applicable for S >= 1, which... ▽ More

Submitted 1 October, 2010; **v1** submitted 24 June, 2010; **originally announced** June 2010.
Comments: 12 pages, 11 figures, final version
Journal ref: Phys. Rev. B, 82, 144402 (2010)



15 years anniversary

- ❑ Need to switch compounds to track the field-evolution



$(5CAP)_2CuCl_4$ "CAPCC"

Spin ordering $T_N = 0.74$ K

Spin saturation $H_s = 3.62$ T

Christensen, Nielsen, Mourigal (in PhD Thesis 2011), McMorrow, Ronnow

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- Examples: Greatest Hits 2012-2025
  - Bound-states: Triangular uniaxial spin-1 system FeI_2

Bai, Zhang x2, Batista, Mourigal, Nature Physics **17**, 467–472 (2021)

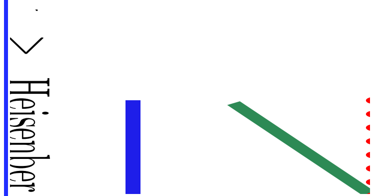
Legros, Zhang x2, Bai, Mourigal, Batista, Armitage PRL **127**, 267201 (2021)

Bai, Zhang x2, Mourigal, Batista, Nat. Comm. **14**, 4199 (2023)

Dahlbom, Barros, Mourigal, Bai, Batista, Phys. Rev. B **109** 014427 (2024)

Toy model for FeI_2

- Consider a spin-1 ferromagnetic easy-axis chain



su(3) coherent states

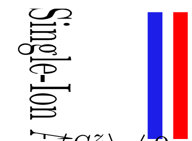
$$S^{\alpha} \quad \text{x3}$$

$$Q^{\alpha\beta} = \frac{(S^{\alpha}S^{\beta} + S^{\beta}S^{\alpha})}{2} - \frac{\delta^{\alpha\beta}}{3} \quad \text{x5}$$

Spin-1 degrees of freedom

$$\mathcal{H} = -D\sum_i (S_i^z)^2 + J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

ground-state



$\langle S_i^z \rangle \neq 0$ $\langle Q_i^{zz} \rangle \neq 0$

Anisotropy dominates exchange

Extended from: Petitgrand et al., JMMM 14, 275 (1979); Petitgrand et al., JMMM 15, 381 (1980); Katsumata et al., PRB 61, 11632 (2000)

Toy model for FeI_2

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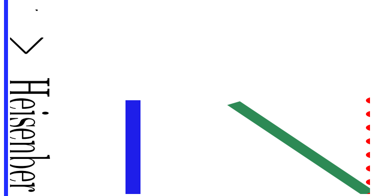

su(3) coherent states

S^{α} x3

$Q^{\alpha\beta} = \frac{(S^{\alpha}S^{\beta} + S^{\beta}S^{\alpha})}{2} - \frac{\delta^{\alpha\beta}}{3}$ x5

Spin-1 degrees of freedom

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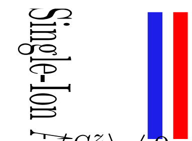
ground-state



$\langle S_i^z \rangle \neq 0$ $\langle Q_i^{zz} \rangle \neq 0$

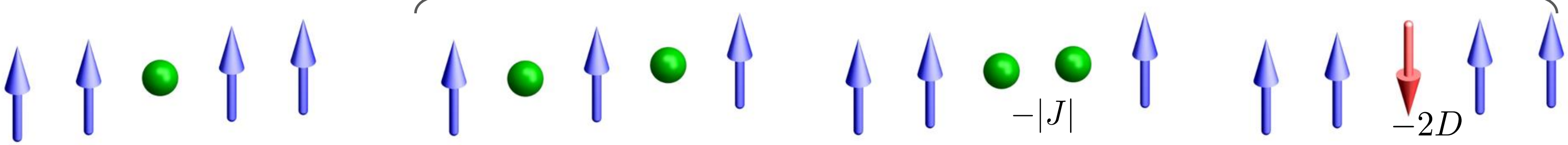
Anisotropy dominates exchange

- Excitations:

$|\Delta S^z| = 1$

$\overbrace{\qquad\qquad\qquad}^{|\Delta S^z| = 2}$

$[S_i^- S_i^-] \equiv Q_i^{xx} - Q_i^{yy} - 2iQ_i^{xy}$



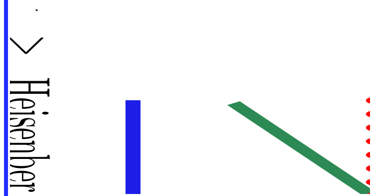
One-Magnon $+2|J| + D$ | Two-Magnon $+4|J|+2D$ | Two-Magnon Bound-State $+3|J|+2D$ | Single-Ion Bound-State $+4|J|$

low energy | high energy | lowest energy

Extended from: Petitgrand et al., JMMM 14, 275 (1979); Petitgrand et al., JMMM 15, 381 (1980); Katsumata et al., PRB 61, 11632 (2000)

Toy model for FeI_2

- Consider a spin-1 ferromagnetic easy-axis chain



su(3) coherent states

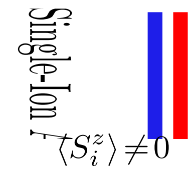
S^{α} x3

$$Q^{\alpha\beta} = \frac{(S^{\alpha}S^{\beta} + S^{\beta}S^{\alpha})}{2} - \frac{\delta^{\alpha\beta}}{3}$$ x5

Spin-1 degrees of freedom

$$\mathcal{H} = -D\sum_i (S_i^z)^2 + J\sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

ground-state



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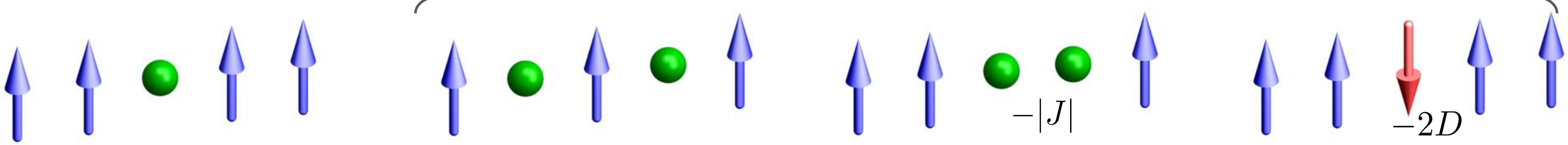


One-Magnon $+2|J| + D$

Two-Magnon $+4|J| + 2D$

Two-Magnon Bound-State $+3|J| + 2D$

Single-Ion Bound-State $+4|J|$

low energy

high energy

lowest energy

- Our new insight:

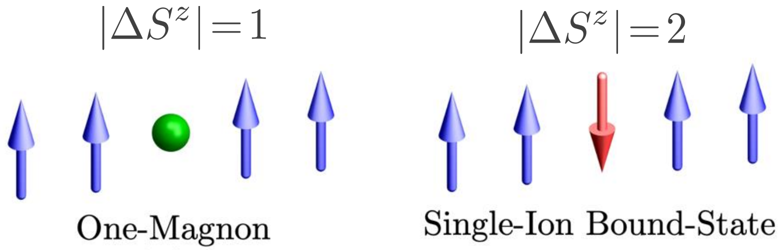
$$\begin{pmatrix} J_1^{xx} & 0 & 0 \\ 0 & J_1^{yy} & \boxed{J_1^{yz}} \\ 0 & \boxed{J_1^{yz}} & J_1^{zz} \end{pmatrix}$$

off-diagonal exchange

Extended from: Petitgrand et al., JMMM 14, 275 (1979); Petitgrand et al., JMMM 15, 381 (1980); Katsumata et al., PRB 61, 11632 (2000)

Low-energy excitations are hybridized in FeI_2

- Scale of exchange and anisotropy such that two flavors overlap in energy



spin-one Hilbert space

$$\mathcal{H} = -D\sum_i (S_i^z)^2$$

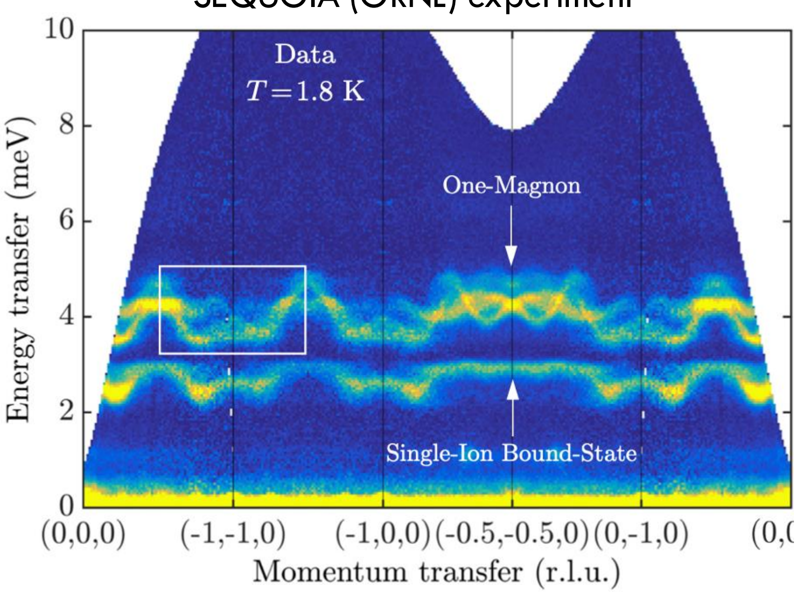
single-ion anisotropy

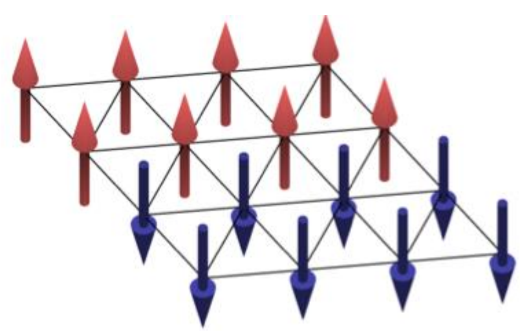
$$\begin{pmatrix} J_1^{xx} & 0 & 0 \\ 0 & J_1^{yy} & \boxed{J_1^{yz}} \\ 0 & \boxed{J_1^{yz}} & J_1^{zz} \end{pmatrix}$$

hybridization

- *Result:* Bound-state hybridized with magnons by anisotropic exchange

SEQUOIA (ORNL) experiment





Bai, Zhang, Batista, Mourigal, Nature Physics 17, 467–472 (2021)

Low-energy excitations are hybridized in FeI_2

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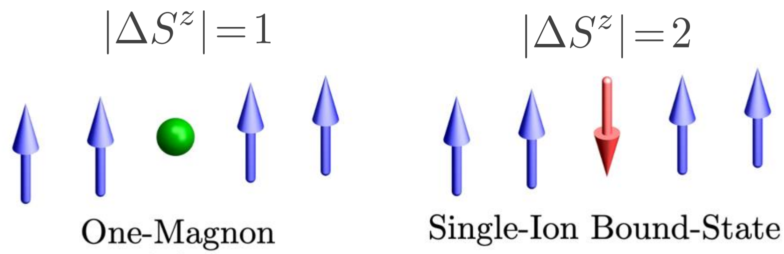


$$\mathcal{H} = -D \sum_i (S_i^z)^2$$

$$\begin{pmatrix} J_1^{xx} & 0 & 0 \\ 0 & J_1^{yy} & \boxed{J_1^{yz}} \\ 0 & \boxed{J_1^{yz}} & J_1^{zz} \end{pmatrix}$$

spin-one Hilbert space

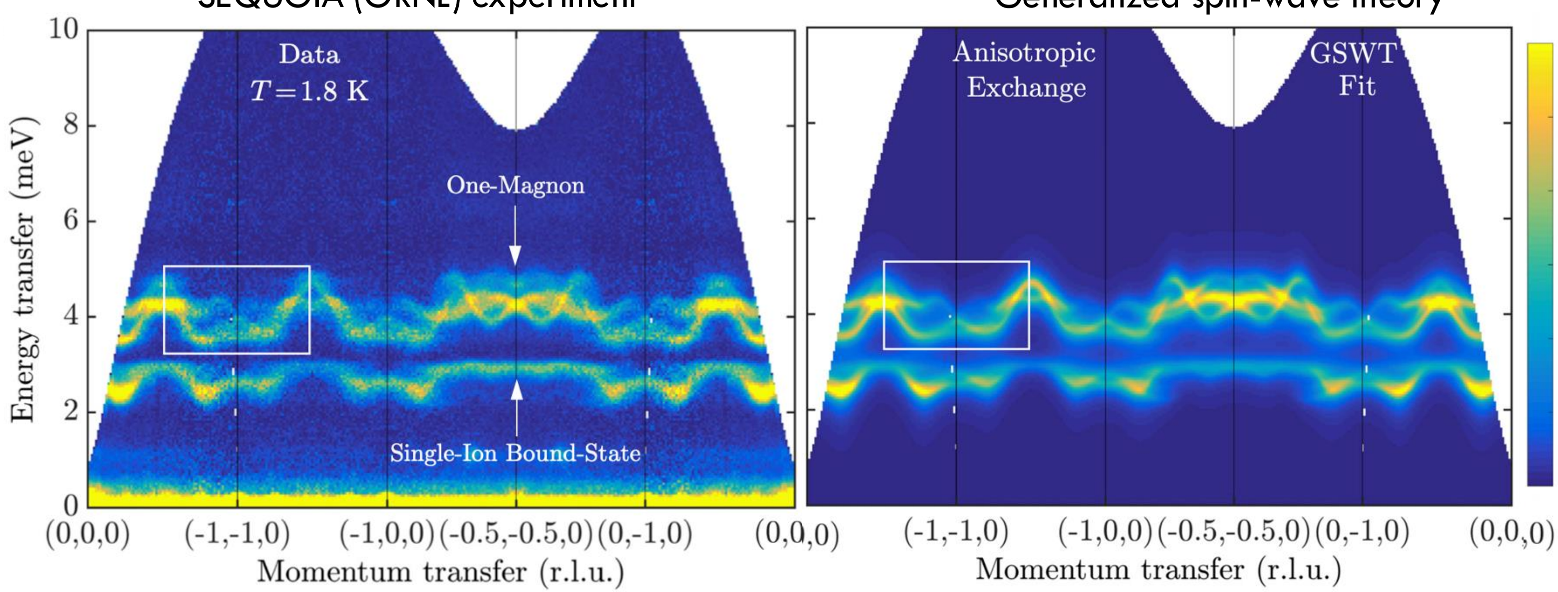
single-ion anisotropy

hybridization

- *Result:* Bound-state hybridized with magnons by anisotropic exchange

SEQUOIA (ORNL) experiment

Generalized spin-wave theory

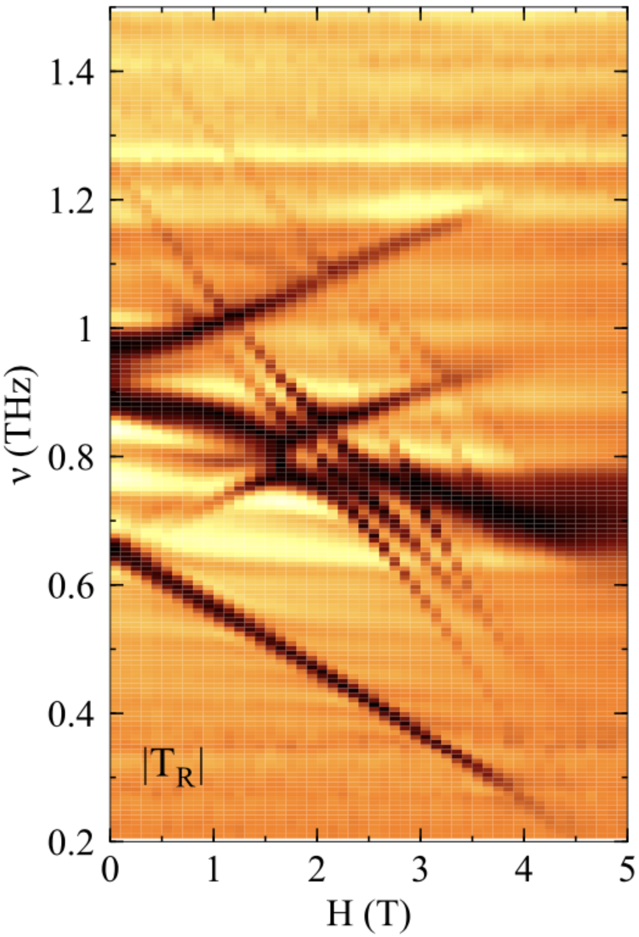


Bai, Zhang, Batista, Mourigal, Nature Physics 17, 467–472 (2021)

FeI_2 : higher order bound-states

- Time-domain Terahertz Spectroscopy

Right Polarized



A. Legros, S.-S. Zhang, X. Bai *et al.*, PRL 127, 267201 (2021)

FeI_2 : higher order bound-states

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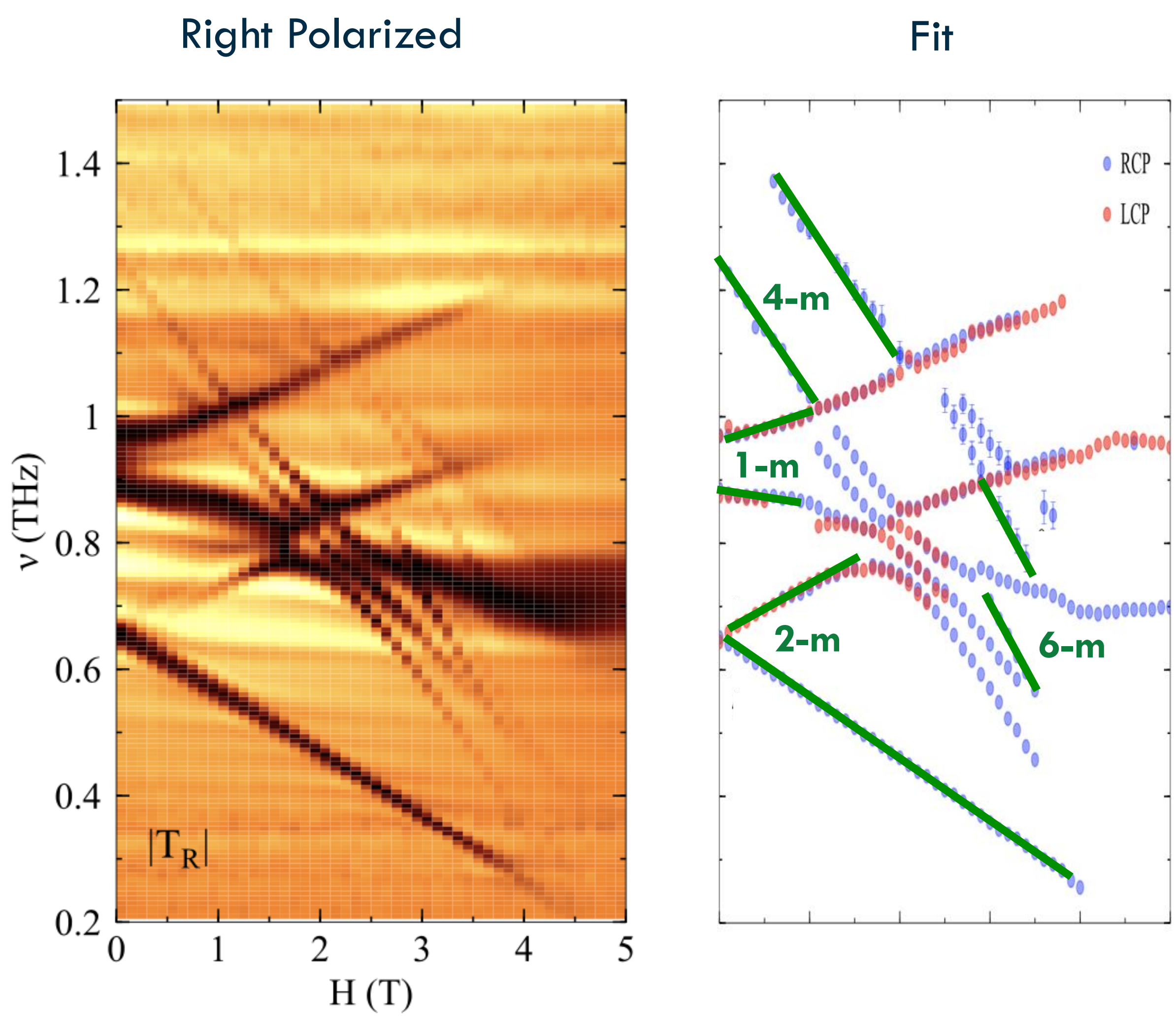

A. Legros, S.-S. Zhang, X. Bai *et al.*, PRL 127, 267201 (2021)

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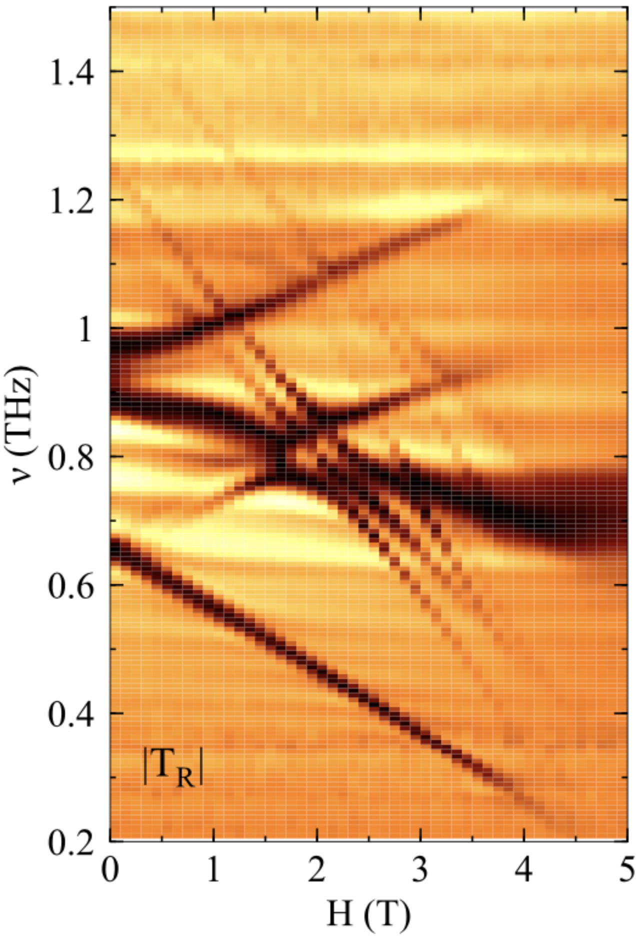
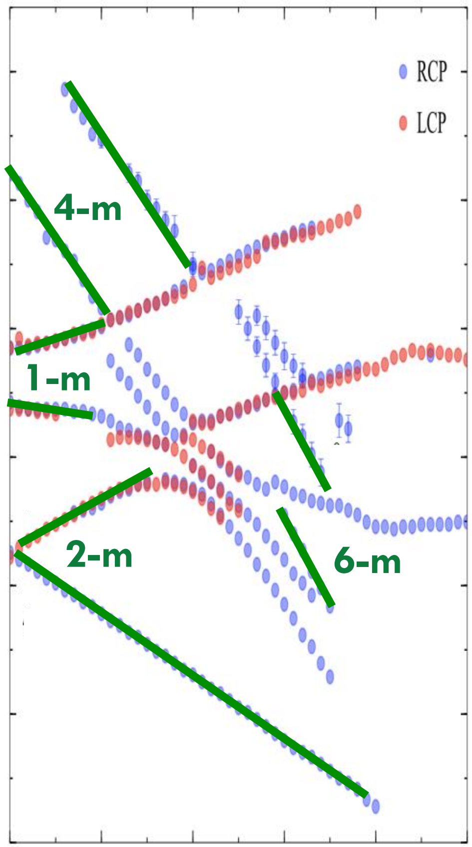
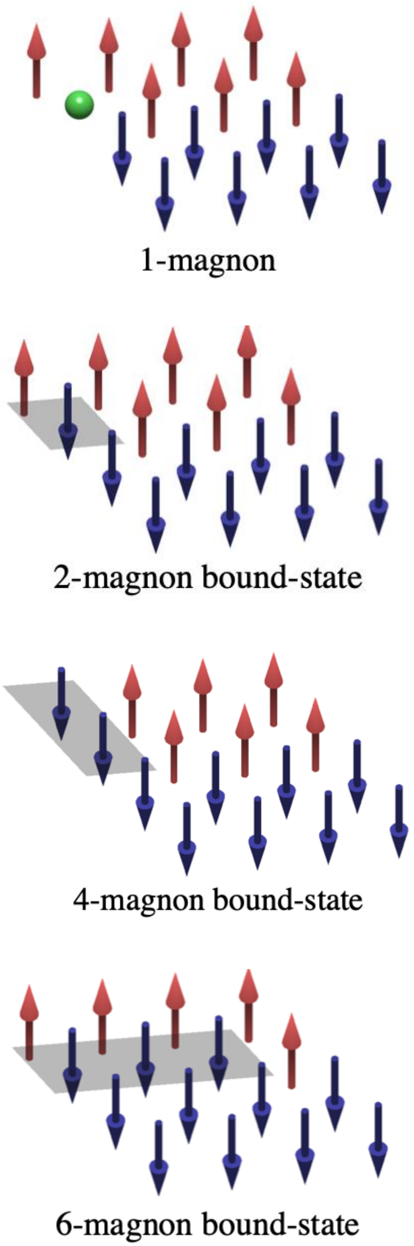


Fit



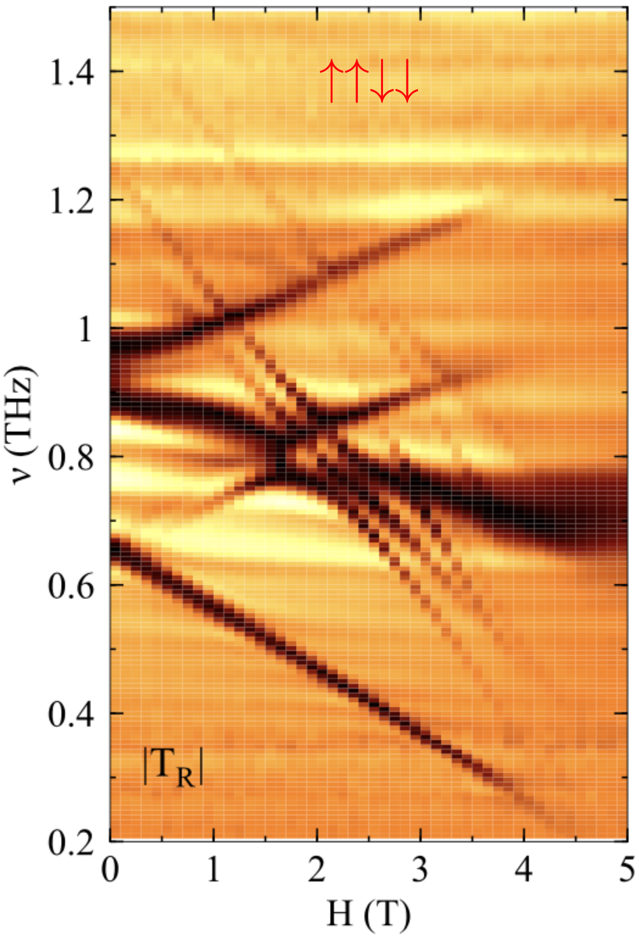


A. Legros, S.-S. Zhang, X. Bai *et al.*, PRL 127, 267201 (2021)

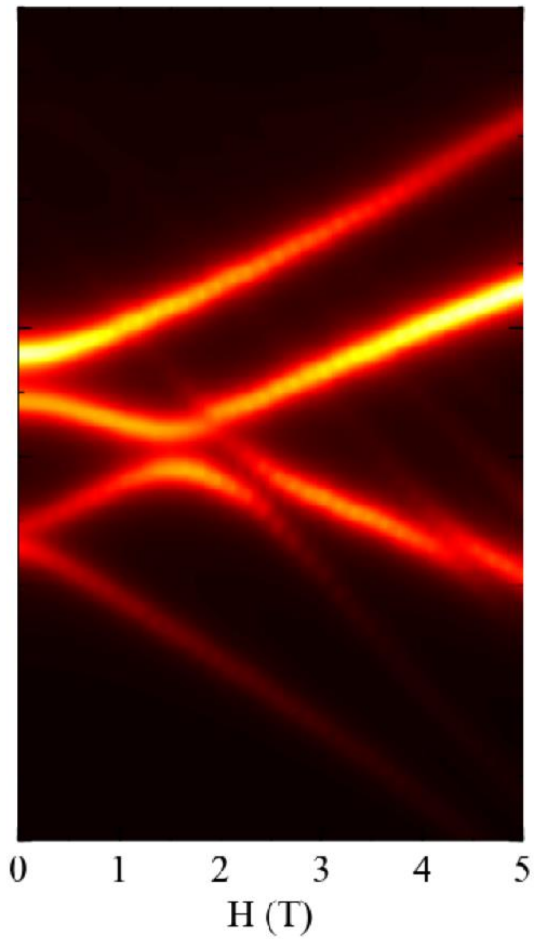
FeI_2 : higher order bound-states

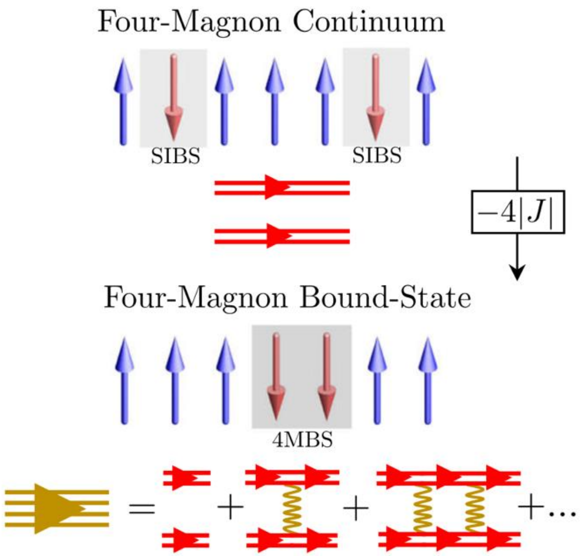
- Time-domain Terahertz Spectroscopy exquisite for field studies

Right Circ. Polarized



Truncated Hilbert Space
Diag. 5x5x5 with up to 4m



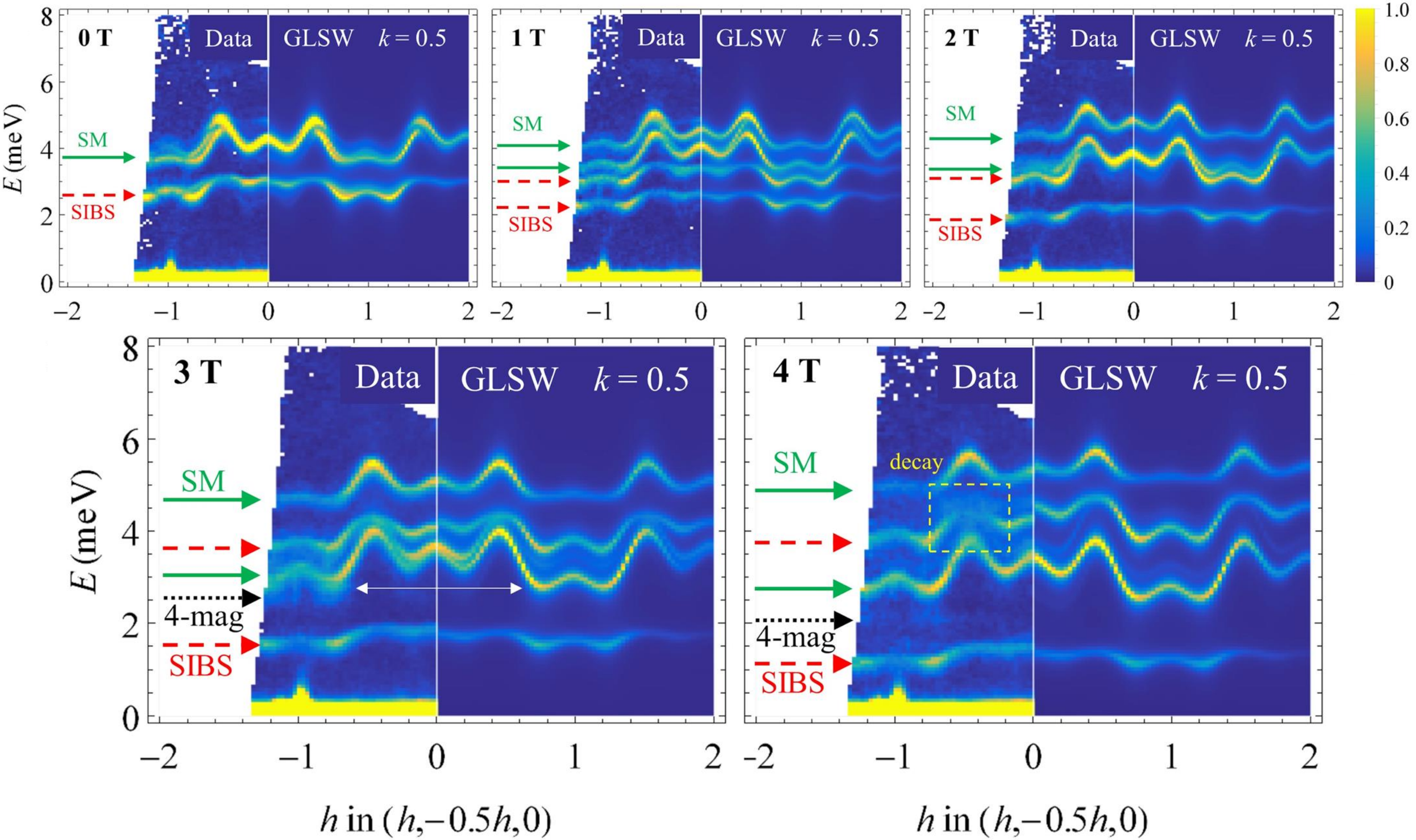


Hao Zhang

Shang-Shun Zhang

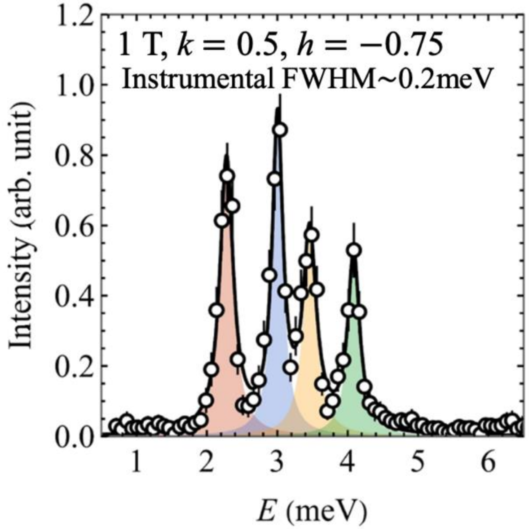
FeI_2 : magnetic field evolution of excitations

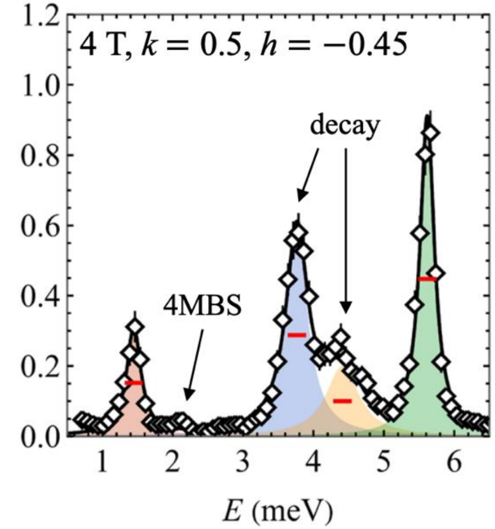
- Anisotropic exchange and field misalignment stabilizes single-domain

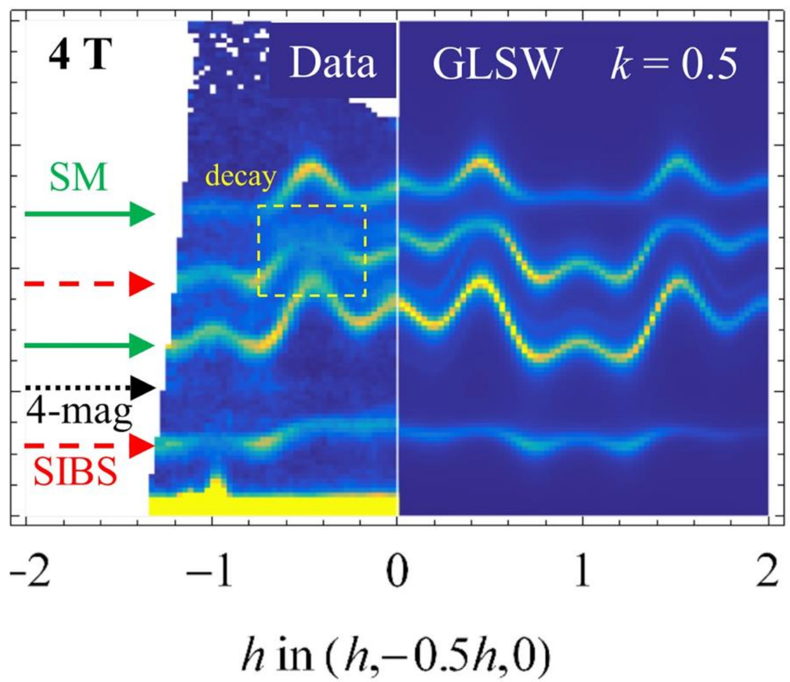


FeI_2 : magnetic field evolution of excitations

- Observe spontaneous decay





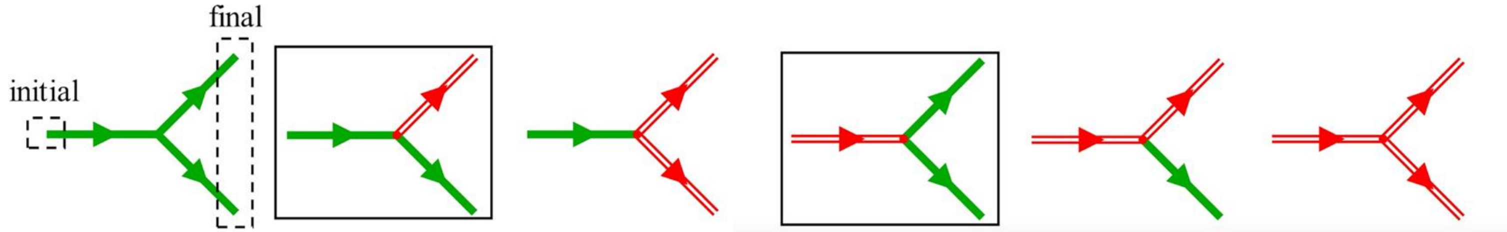


FeI_2 : magnetic field evolution of excitations

- Mechanism for spontaneous decay in FeI_2

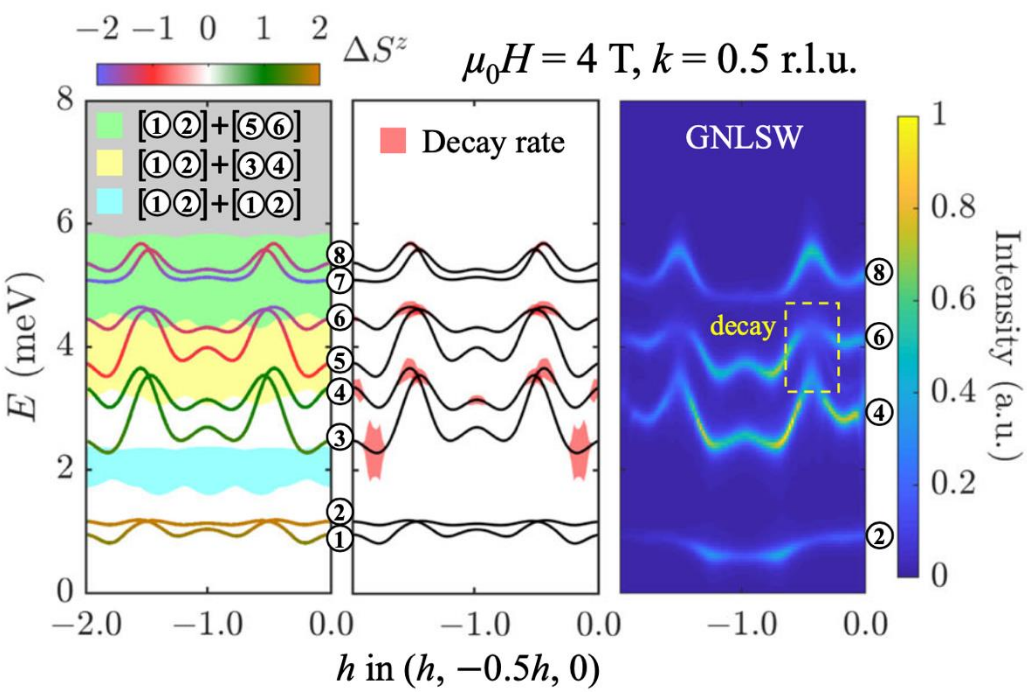
Anisotropic exchange opens decay channels between dipolar and quadrupolar modes

$$J^{\pm\pm} S_i^+ S_j^+$$



- Kinematic conditions for generalized spin-wave theory with one-loop corr.

4 T



Shang-Shun Zhang

FeI_2 : magnetic field evolution of excitations

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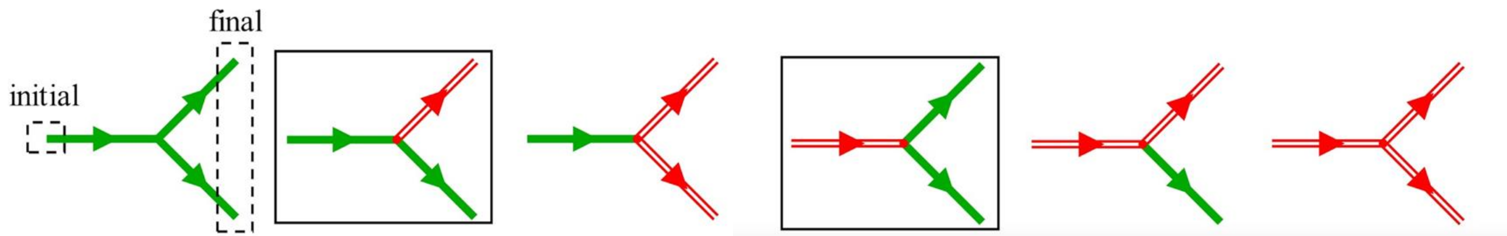
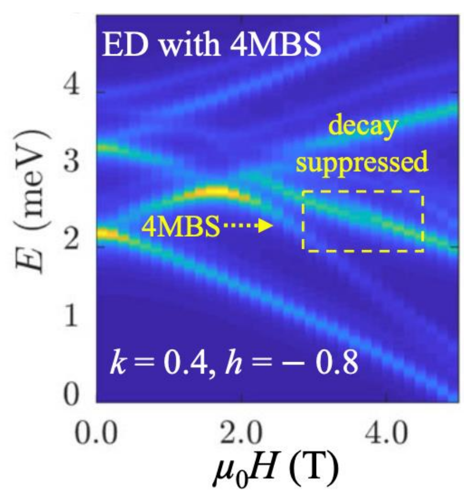
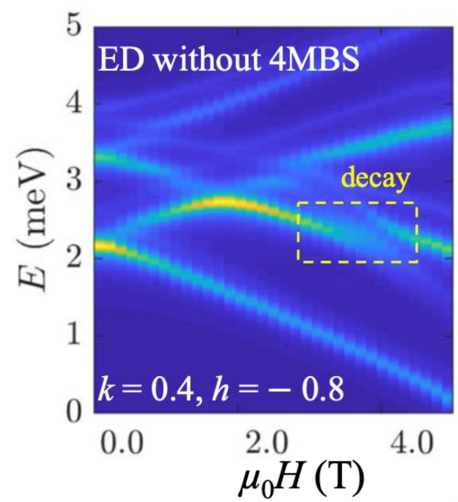
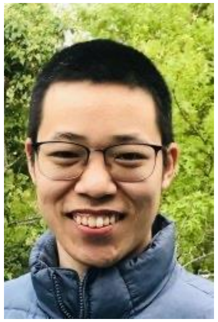


- Recombination of decay products into a heavy bound-state suppresses rate

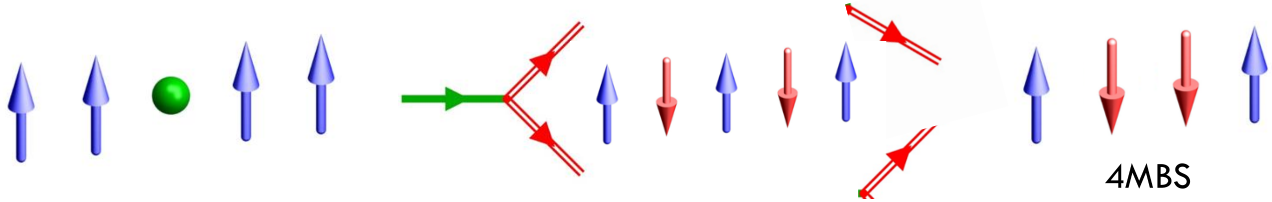
3 T







Shang-Shung Zhang



Outline

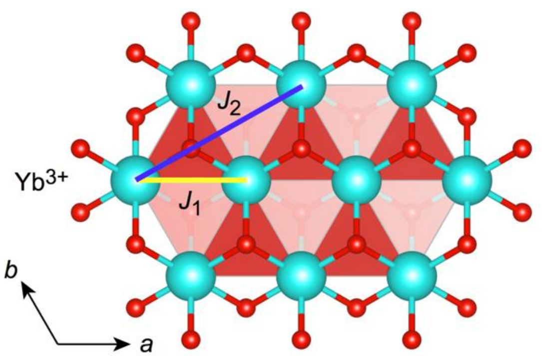
- Heavy Introduction
  - Quantum Magnetism Big Picture
  - Research Approach
  - Inelastic Neutron Scattering
  - Modeling

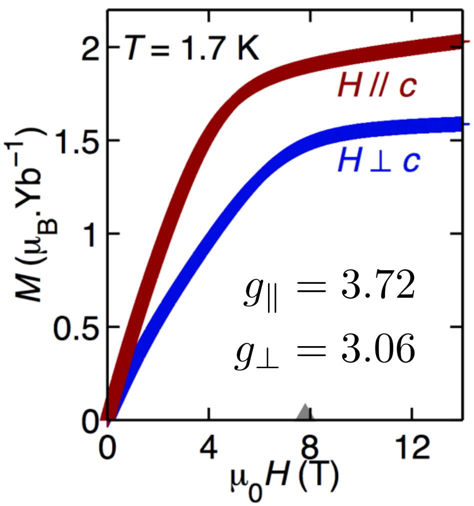
2

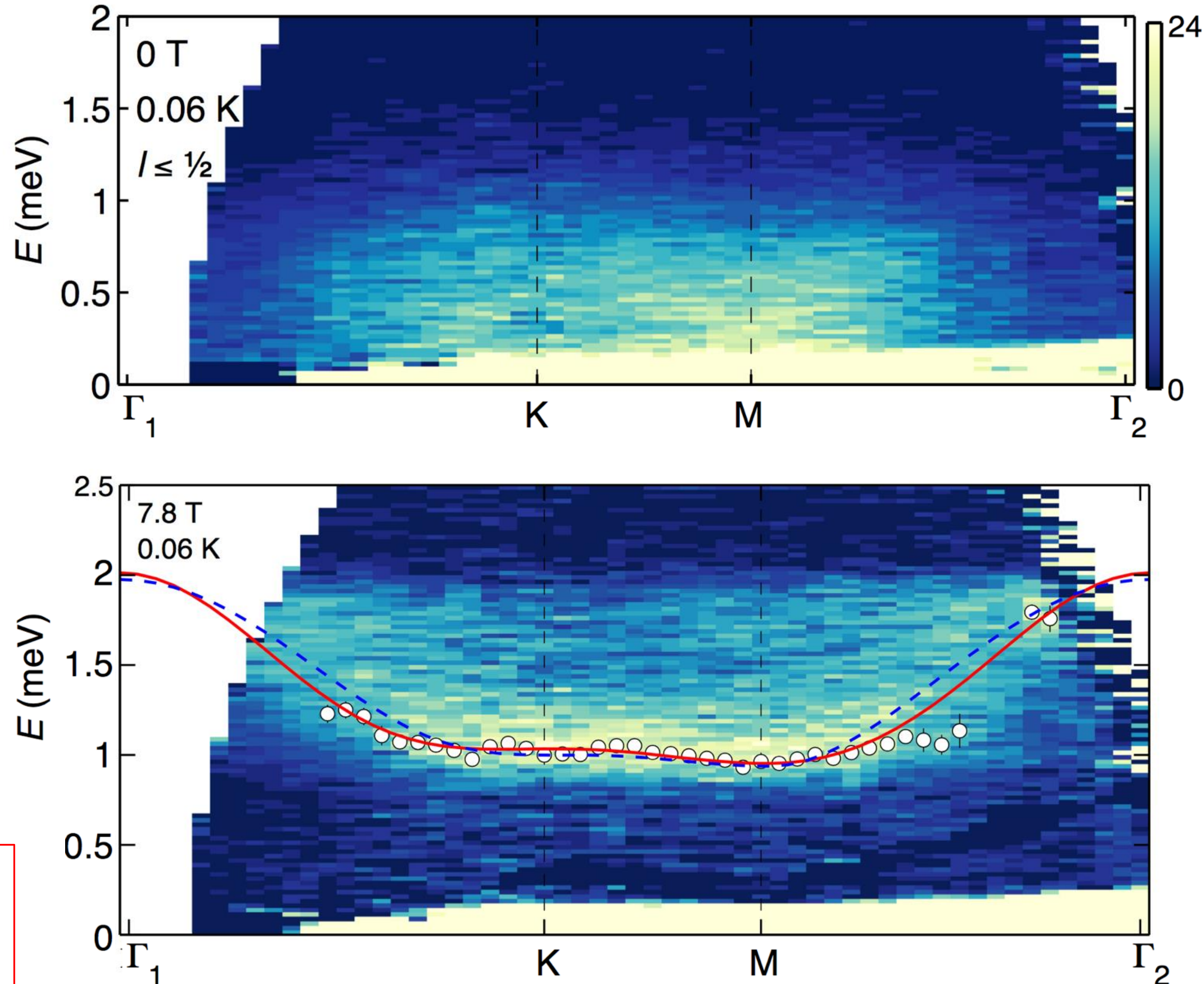
- Examples: Greatest Hits 2012-2025
  - Spinons: Heisenberg spin-1/2 chains in copper sulfate
  - Magnons: Square-lattice problems
  - Bound-states: Triangular uniaxial spin-1 system FeI_2
  - Continuous excitations does not always mean fractionalization/decay
- Conclusion (maybe)

Continuum does not always mean fractionalization/decay

- Rare-earth triangular antiferromagnet in $YbMgGaO_4$ (H_s~5.0T)





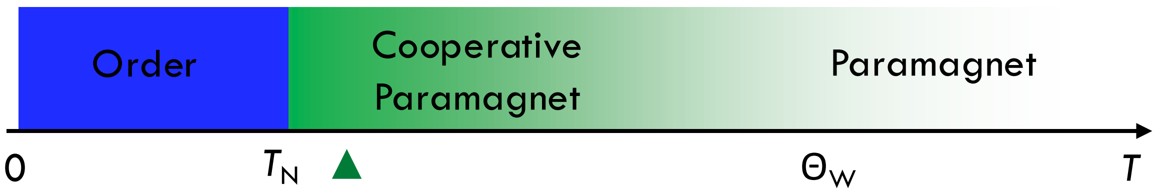


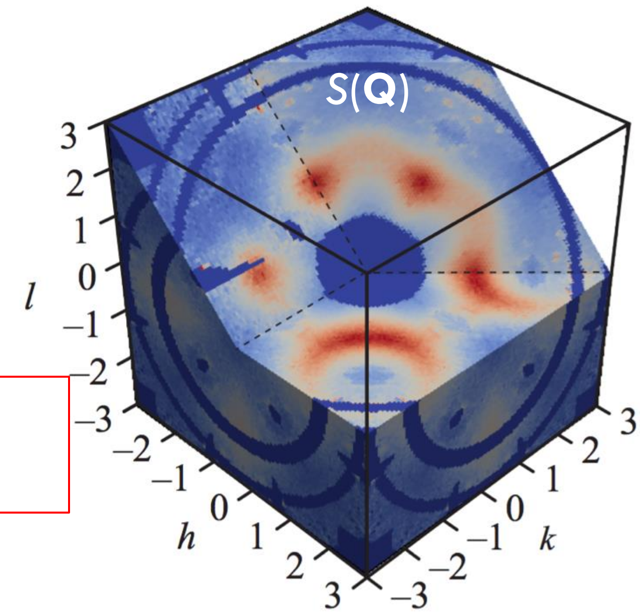
Magnetic excitations are broad because of exchange disorder

Paddison, Dun, Daum, Zhou, Mourigal, Nature Physics **13**, 117 (2017)

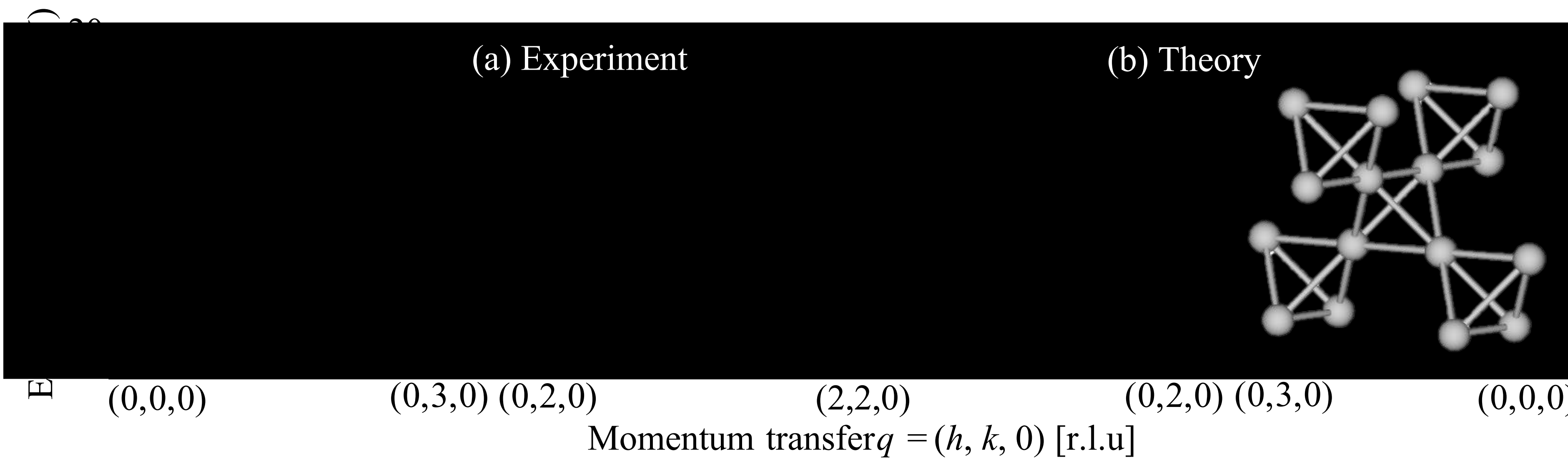
Continuum does not always mean fractionalization/decay

- The classical S=3/2 pyrochlore antiferromagnet $MgCr_2O_4$





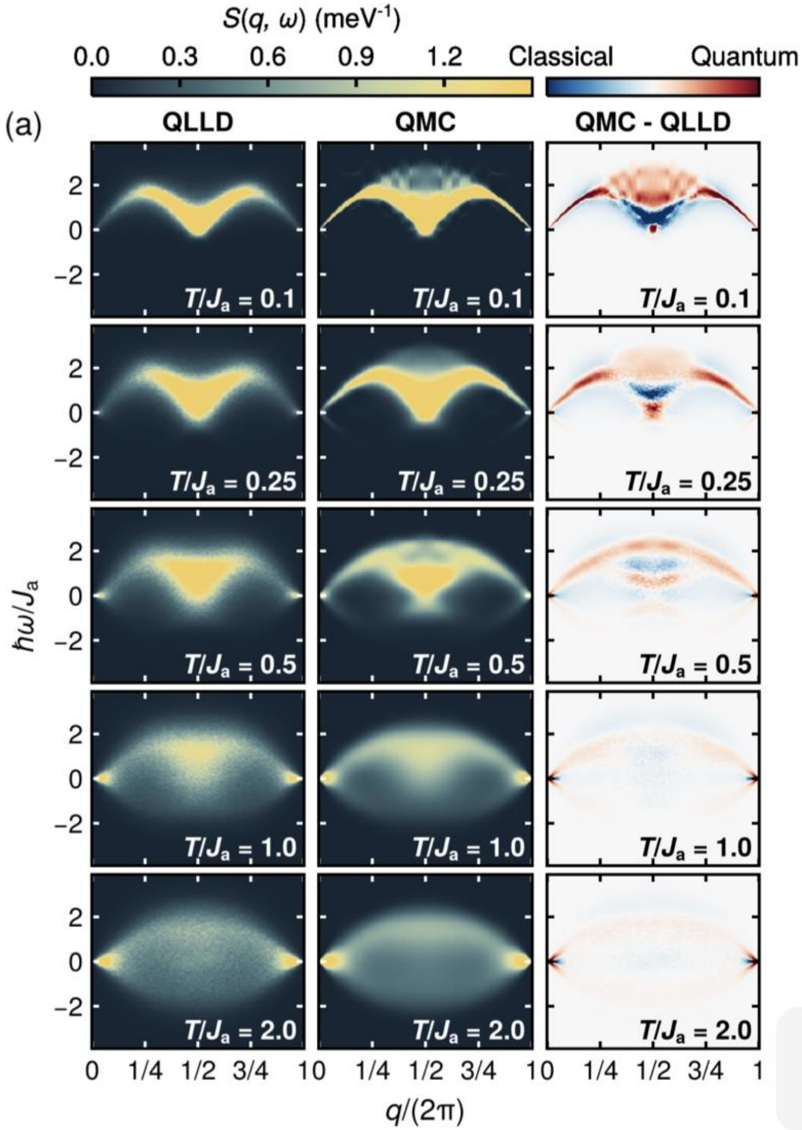
Magnetic excitations are broad because they ride slowly fluctuating correlated paramagnetic configurations



Bai, Paddison, Kapit, Broholm, Chalker, Mourigal, Phys. Rev. Lett. **122**, 097201 (2019)

Continuum does not always mean fractionalization/decay

- Finite T dynamics of the spin-1/2 chain emulated by classical dynamics



Classical non-linearities in the spin dynamics can mimic spinon continuum for temperatures as low as T/J ~ 0.5 for the Heisenberg chain

Kim and Mourigal, arXiv:2503.19975

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Conclusion

So magnons are stable, right?

Thank you for your attention!