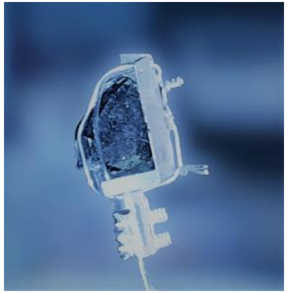
FRACTIONAL EXCITATIONS IN LOW DIMENSIONAL QUANTUM MAGNETS

Stephen E. Nagler

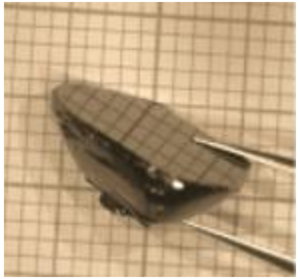
ORNL / UTK / PRB

Outline

- Quasi-1D example: KCuF$_3$



- Quasi-2D example: α-RuCl$_3$

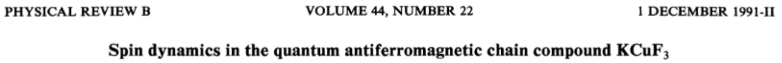


Collaborators on $KCuF_3$ experiments (over 30 years (!)) and α-$RuCl_3$ neutron scattering experiments (almost 10 years (!)):

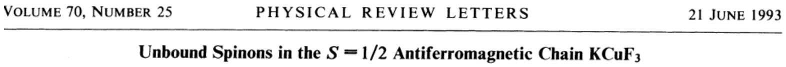
D. Abernathy, A. Aczel, G. Alvarez, A. Banerjee, C. Balz, T. Barthel, C. Batista, T. Berlijn, S. Bhattacharjee, C. Bridges, H. Cao, J.-S. Caux, B. Chakoumakos, Y. Cheng, M. Cothrine, R. A. Cowley, K. Dixit, M. Dupont, G. Ehlers, C. D. Frost, O. Garlea, G. E. Granroth, G. Halasz, X. Hu, L. Jansson, Y. Kamiya, J. Knolle, D. Kovrizhin, G. Khundzakishvili, B. Lake, P. Lampen-Kelley, P. Laurell, L. Li, L. Liang, Y. Liu, Z. Lu, M. Lumsden, D. Mandrus, R. Moessner, J. E. Moore, S. Mu, S. Okamoto, D. Pajerowski, T. G. Perring, C. Polanko, A. M. Samarakoon, C. Sarkis, S. K. Satija, A. Scheie, U. Schollwöck, N. E. Sherman, G. Shirane, M. B. Stone, Y. Takano, D. A. Tennant, A. M. Tsvelik, M. Vojta, X. Wang, D. Welz, , B. Winn, S. M. Yadav, K. Yamada, J.-Q. Yan, Y. Yiu, S. Zhang.

Selected (and parochial) $KCuF_3$ bibliography

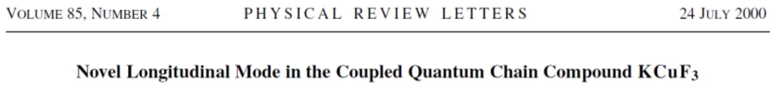
Older


PHYSICAL REVIEW B VOLUME 44, NUMBER 22 1 DECEMBER 1991-II

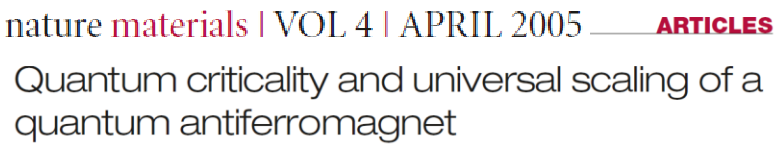
Spin dynamics in the quantum antiferromagnetic chain compound $KCuF_3$


VOLUME 70, NUMBER 25 PHYSICAL REVIEW LETTERS 21 JUNE 1993

Unbound Spinons in the $S = 1/2$ Antiferromagnetic Chain $KCuF_3$


VOLUME 85, NUMBER 4 PHYSICAL REVIEW LETTERS 24 JULY 2000

Novel Longitudinal Mode in the Coupled Quantum Chain Compound $KCuF_3$

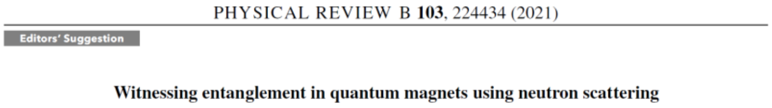

nature materials | VOL 4 | APRIL 2005 ARTICLES

Quantum criticality and universal scaling of a quantum antiferromagnet


PRL **111,** 137205 (2013) PHYSICAL REVIEW LETTERS week ending 27 SEPTEMBER 2013

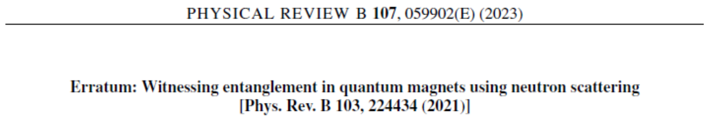
Multispinon Continua at Zero and Finite Temperature in a Near-Ideal Heisenberg Chain

Newer


PHYSICAL REVIEW B **103**, 224434 (2021)

Editors' Suggestion

Witnessing entanglement in quantum magnets using neutron scattering


PHYSICAL REVIEW B **107**, 059902(E) (2023)

Erratum: Witnessing entanglement in quantum magnets using neutron scattering [Phys. Rev. B 103, 224434 (2021)]


LETTERS

https://doi.org/10.1038/s41567-021-01191-6

nature physics

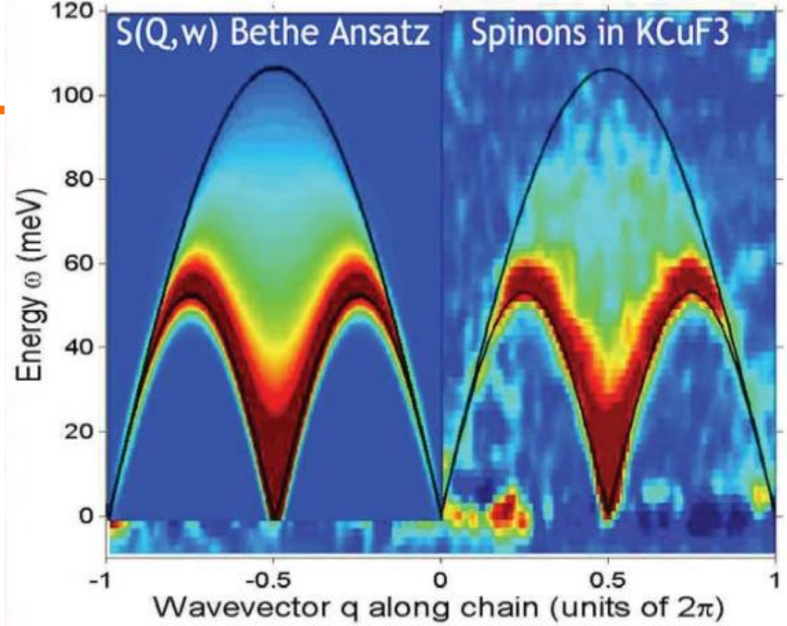
NATURE PHYSICS | VOL 17 | JUNE 2021 | 726-730 |

Check for updates

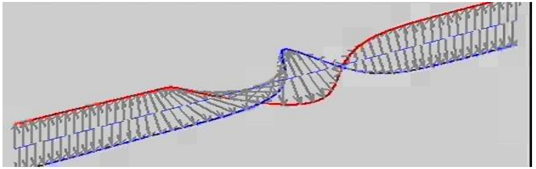
Detection of Kardar–Parisi–Zhang hydrodynamics in a quantum Heisenberg spin-1/2 chain

90+ years of AF Heisenberg chains …

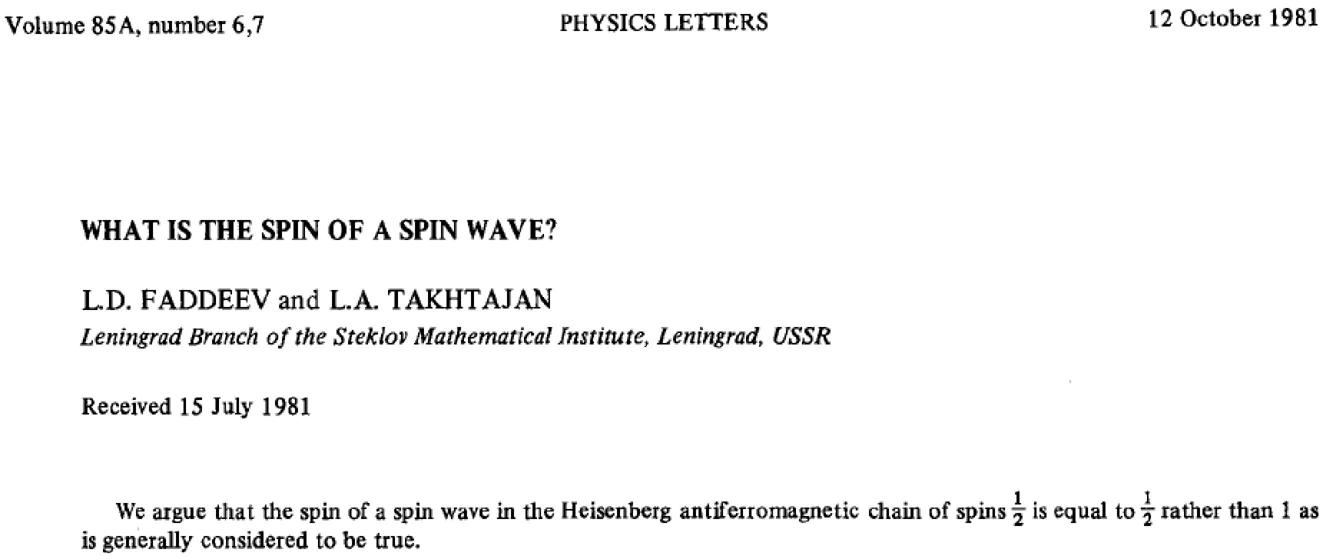
- Heisenberg (1928)
- Bethe (1931)
- des Cloizeaux and Pearson (1961)
- Müller et al. (1981)
- Faddeev & Takhtajan (1981)
- Haldane (1983)
- Prosen, Moore et al. (2019)



$$\hat{H} = 2J\sum_{r} \vec{S}_r \cdot \vec{S}_{r+1}$$



Fractionalized magnetic Excitations



Volume 85A, number 6,7 PHYSICS LETTERS 12 October 1981

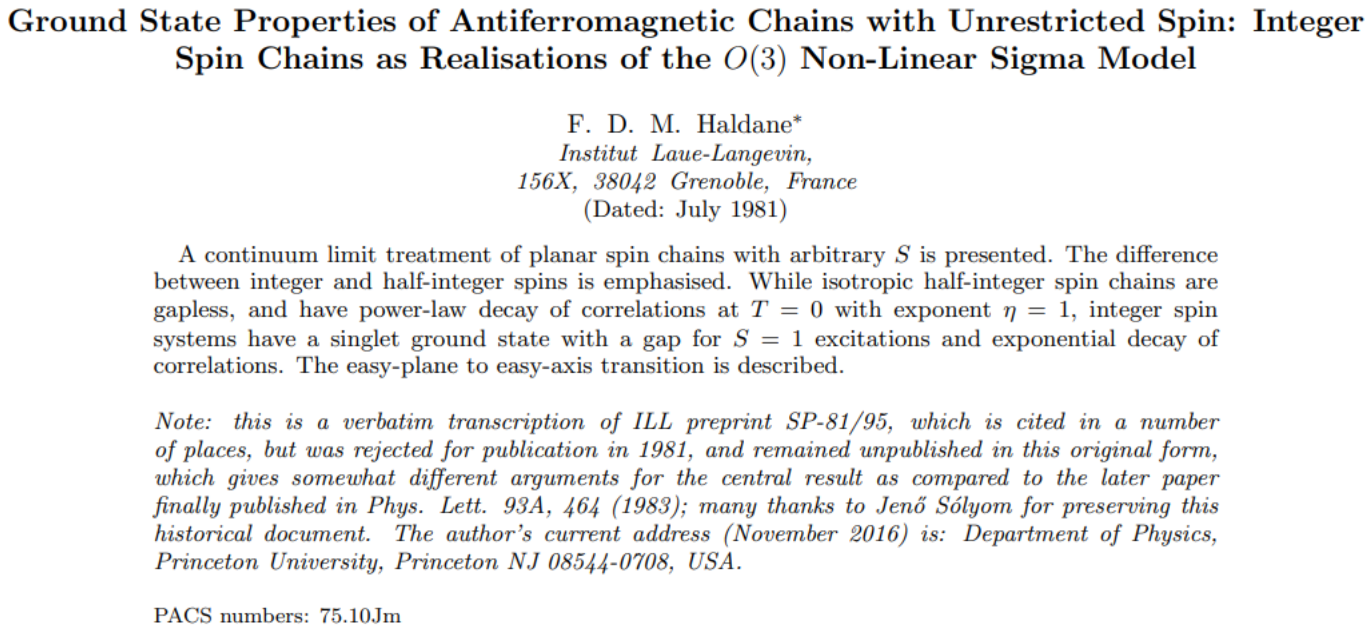
WHAT IS THE SPIN OF A SPIN WAVE?

L.D. FADDEEV and L.A. TAKHTAJAN
Leningrad Branch of the Steklov Mathematical Institute, Leningrad, USSR

Received 15 July 1981

We argue that the spin of a spin wave in the Heisenberg antiferromagnetic chain of spins $\frac{1}{2}$ is equal to $\frac{1}{2}$ rather than 1 as is generally considered to be true.

Fractionalized magnetic Excitations



Ground State Properties of Antiferromagnetic Chains with Unrestricted Spin: Integer Spin Chains as Realisations of the $O(3)$ Non-Linear Sigma Model

F. D. M. Haldane*
Institut Laue-Langevin,
156X, 38042 Grenoble, France

(Dated: July 1981)

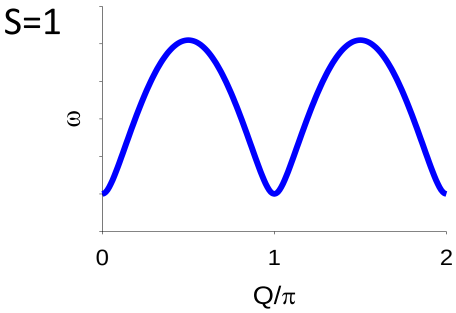
A continuum limit treatment of planar spin chains with arbitrary S is presented. The difference between integer and half-integer spins is emphasised. While isotropic half-integer spin chains are gapless, and have power-law decay of correlations at $T = 0$ with exponent $\eta = 1$, integer spin systems have a singlet ground state with a gap for $S = 1$ excitations and exponential decay of correlations. The easy-plane to easy-axis transition is described.

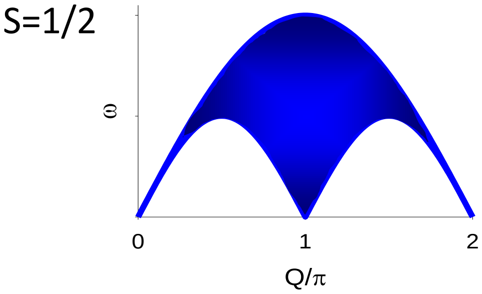
Note: this is a verbatim transcription of ILL preprint SP-81/95, which is cited in a number of places, but was rejected for publication in 1981, and remained unpublished in this original form, which gives somewhat different arguments for the central result as compared to the later paper finally published in Phys. Lett. 93A, 464 (1983); many thanks to Jenő Sólyom for preserving this historical document. The author's current address (November 2016) is: Department of Physics, Princeton University, Princeton NJ 08544-0708, USA.

PACS numbers: 75.10Jm

Quick facts about excitations the Heisenberg AF chain:

- Ground state is a singlet
- The natural excitations are “spinons” and carry spin 1/2 relative to the ground state unlike “magnons” or “spin-waves” which have spin 1 relative to the ground state
- spinons are created only in pairs (or even numbers)
- physically observed states have S_T=1
- for S=1,2,3, ... spinons are bound - result is an energy gap (Haldane gap)
- for S=1/2, 3/2, 5/2, … spinons are “free” - S(Q,ω) has a continuum



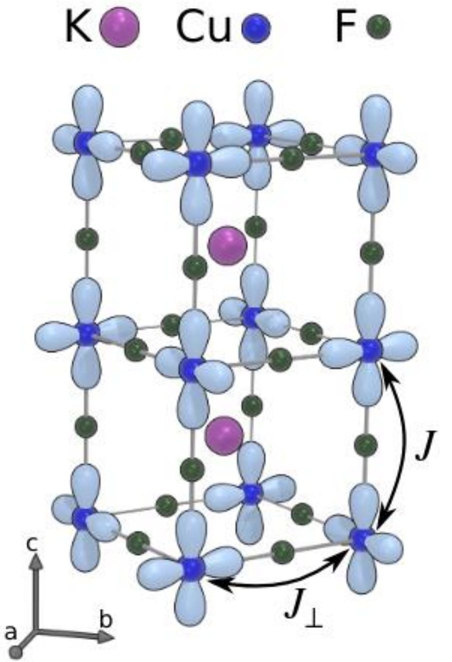


KCuF$_3$: a quasi-1D Heisenberg antiferromagnet

1D chains of $S=1/2$ Cu^{2+} ions extend along the c axis.

Hutchings, Phys. Rev. 1969

Long-range magnetic order near 40 K, but the system retains the fractional $S=1/2$ excitations of the Heisenberg spin chain.



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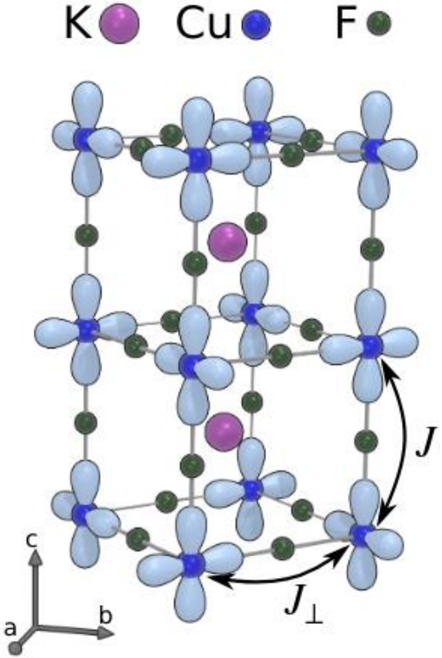
Hutchings, Phys. Rev. 1969



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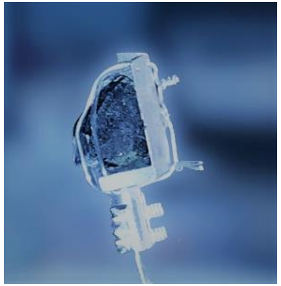
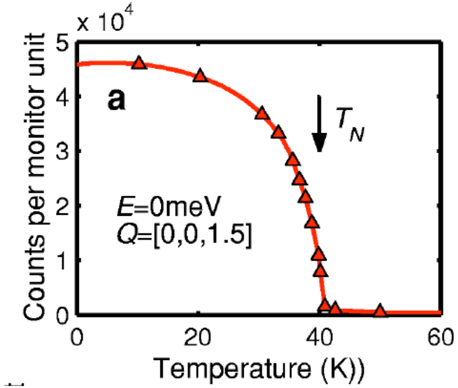
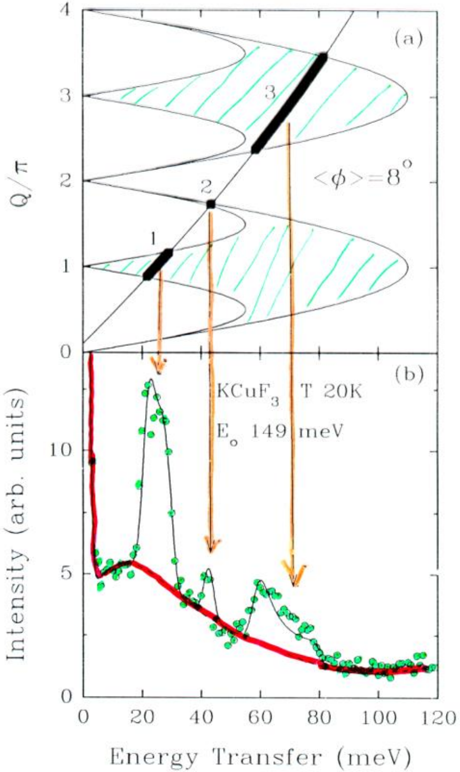
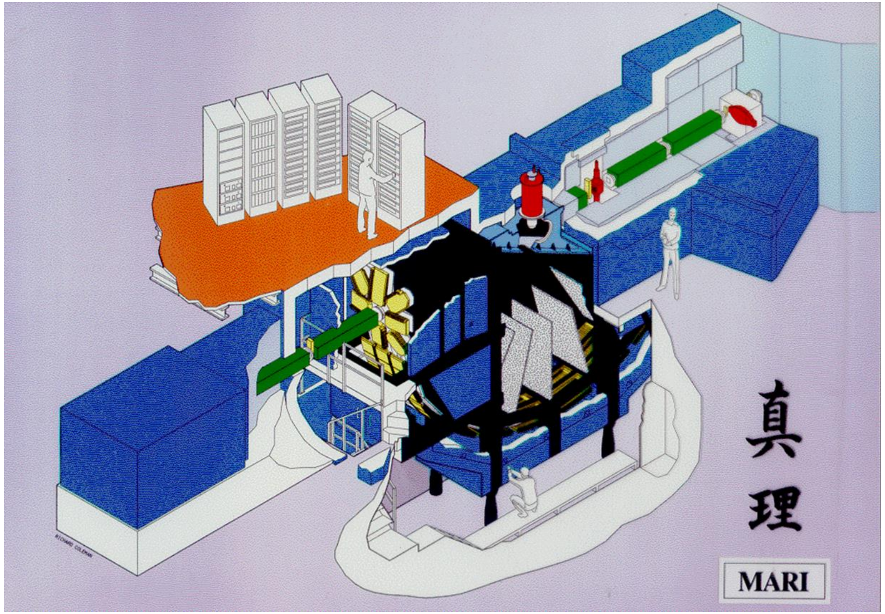




Early 1990s ToF neutron scattering results on $KCuF_3$





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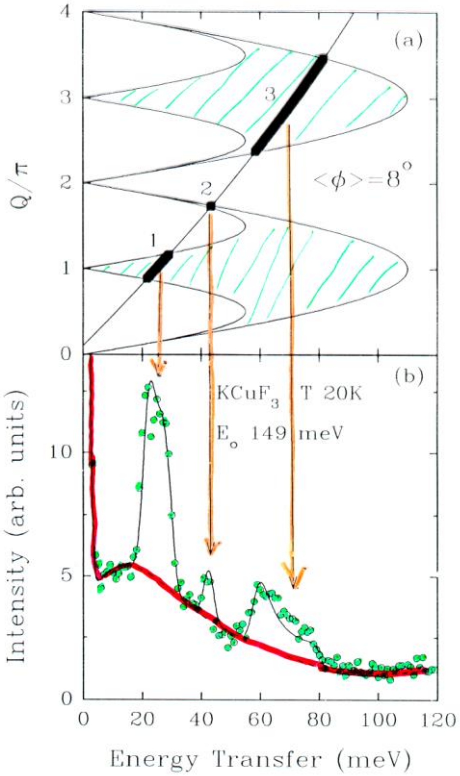
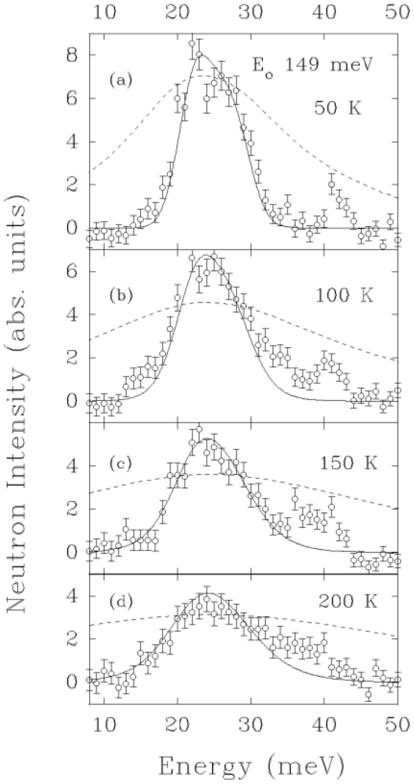
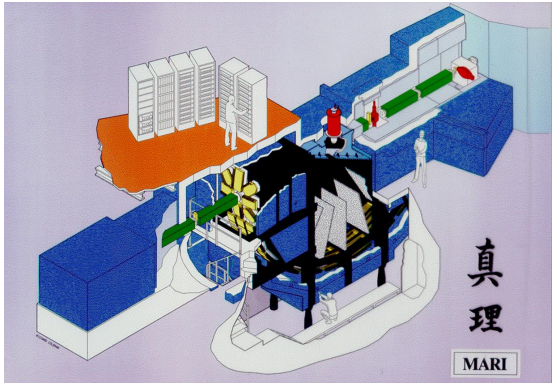
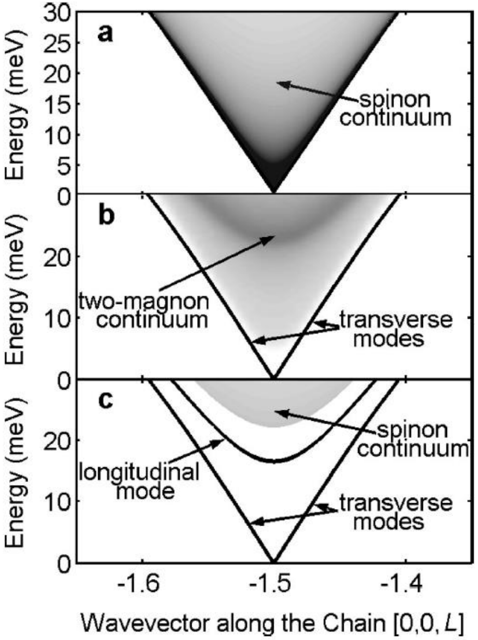






- The temperature dependence of the scattering is in excellent agreement with predictions of field theory (H.J. Schulz, PRB 34, 6372, (1986)).
- Classical theory (H.H. Kretzen *et al.*, Z. Phys. 271, 269 (1974)) underestimates the linewidth at T=0 and overestimates the thermal broadening.

Low energy excitations in the ordered state: magnons plus longitudinal mode or "Higgs mode"

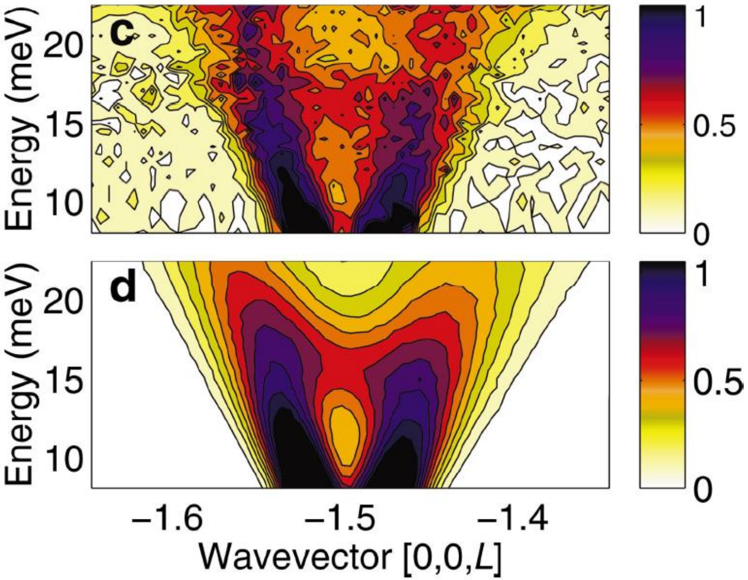


Pure 1D system

SWT to 2nd order

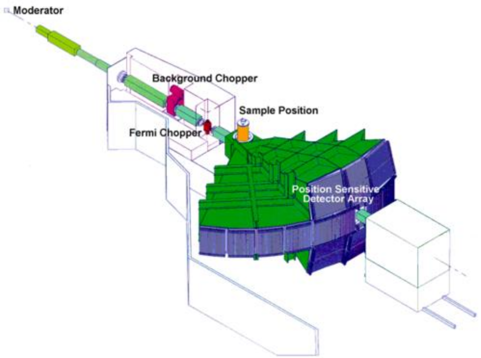
*Interchain coupling treated as perturbation

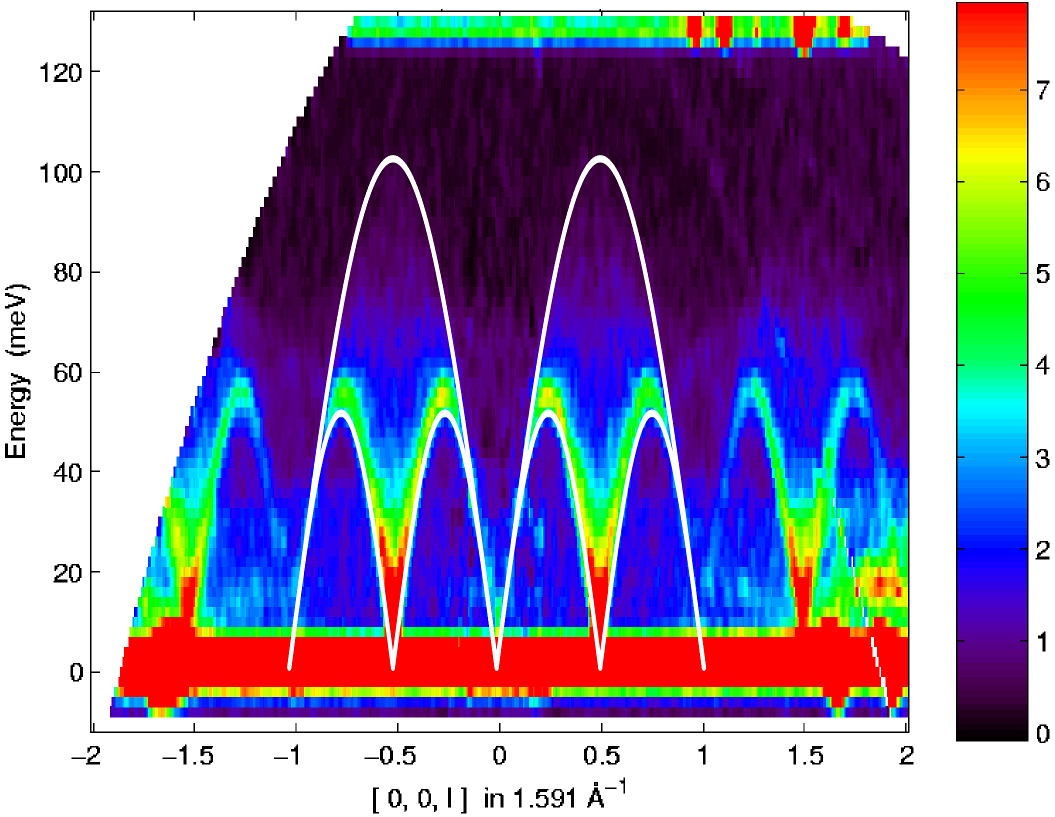
see PRL 85, 832 (2000), PRB 71, 134412 (2005)



*e.g. H.J. Schultz, PRL 77, 2790 (1996) F.H.L. Essler at al., PRB 56, 11001 (1997)

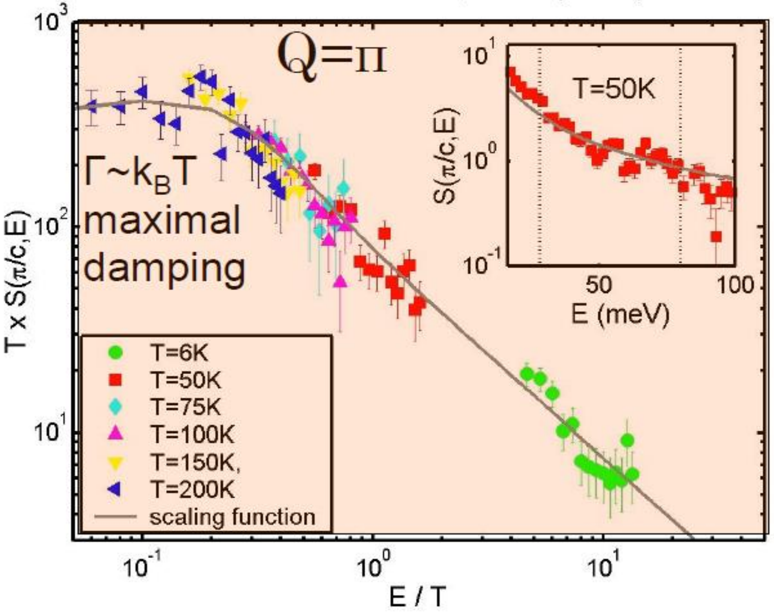
Early 2000's $KCuF_3$ on MAPS E_o = 150 meV   T = 6K

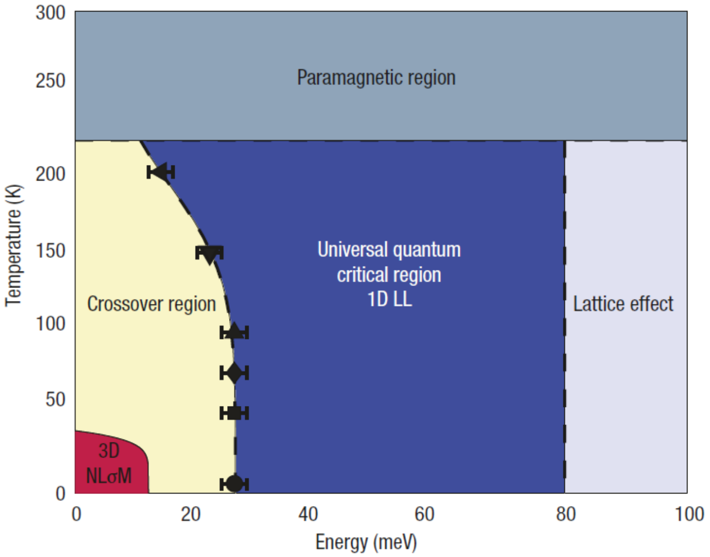




Universal theories for Luttinger liquid and Energy/Temperature scaling confirmed

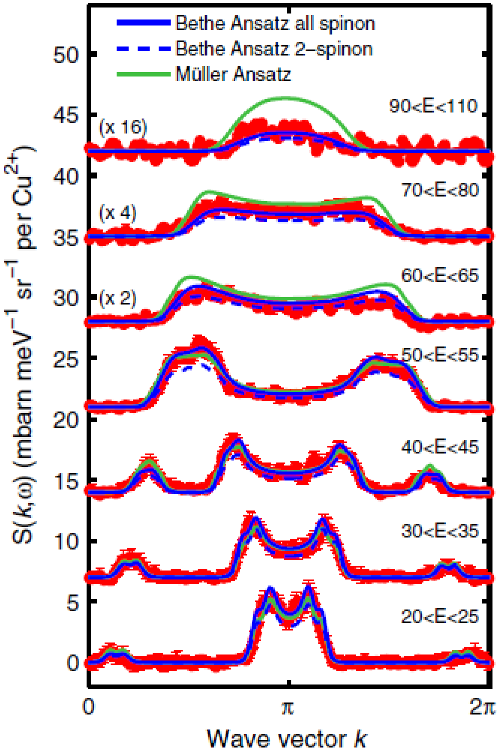
Nature Materials. 4, 329 (2005)

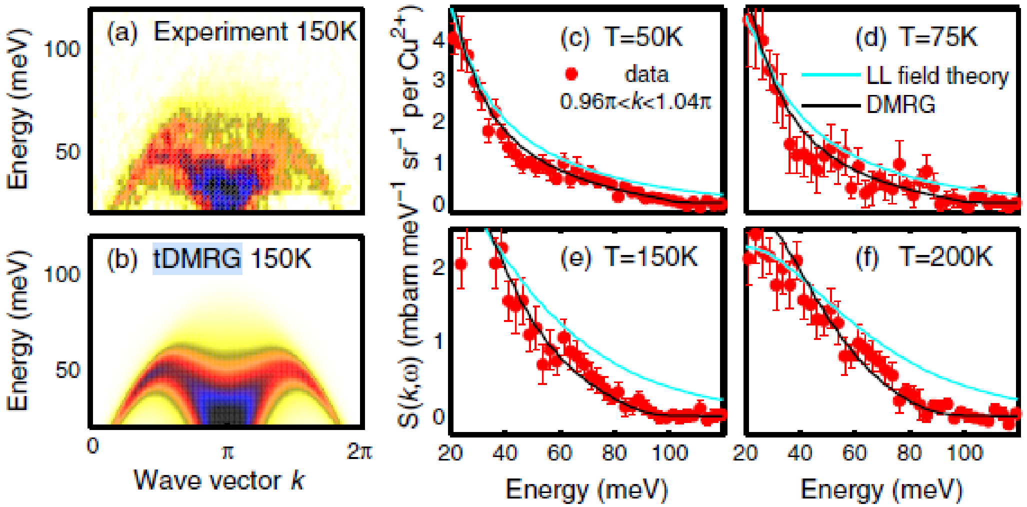




Quantitative agreement of Bethe Ansatz and tDMRG with experiment

PRL 2013





More recently:

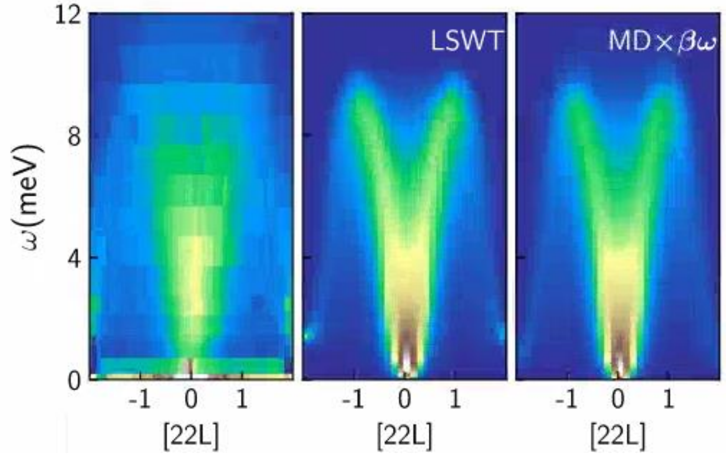
Neutron Scattering and Quantum Fisher Information

How do we measure quantum entanglement in a solid state system?

- Entanglement is significant for quantum matter:
  - Superconductivity
  - Fractional quantum hall
  - Quantum spin liquids

...but we don't have a great way to measure it in extended systems.

The traditional approach: compare data to a theoretical model.



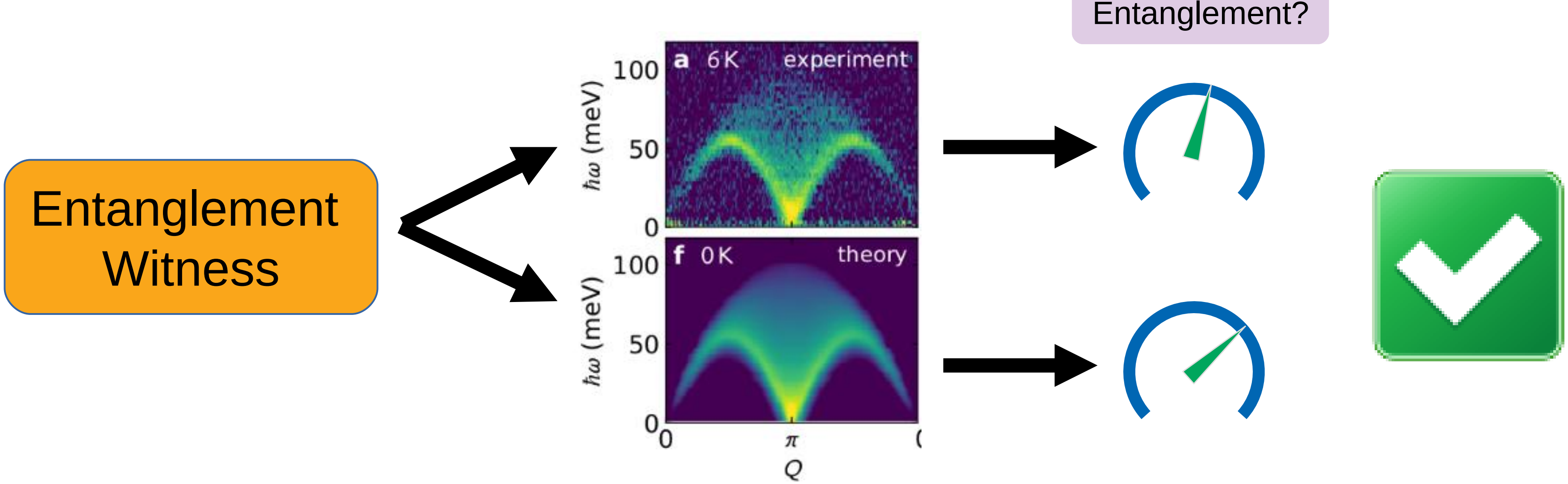
Example: NaCaNi$_2$F$_7$
Zhang at al., PRL122, 167203 (2019)

Problem: **only rarely** do we have good models for highly-entangled states.

We need a model-independent approach.

High-level overview:

We take entanglement witnesses, apply them to neutron scattering data, and measure quantum entanglement.



Entanglement Witness

Entanglement?

Then, we do the same thing for simulated data, to cross-check and verify that the entanglement witnesses work.

Quantum Fisher Information (QFI):

- QFI is related to the variance of an observable: how much the measurement tells you about the state of the system.
- It can be shown that for separable states of N particles, QFI $\leq N$. Conversely, for N entangled particles, QFI$\leq N^2$. (See, e.g., Hyllus et. al. (PRA 85, 022321 (2012) and references therein)
- One can flip this around: by measuring the QFI, you get a bound on the number of entangled particles.

Credit: Virgo

Neutron scattering measures the spin-spin correlation, and signatures of entanglement will in theory be encoded in the magnetic structure factor.

$$S(\vec{q},\omega) \propto \int e^{i\vec{q}\cdot\vec{r}} \langle \vec{S}_{i,\alpha} \vec{S}_{j,\beta} \rangle$$

The problem:

only *sometimes* do we know what to look for.

Quantum Fisher Information (QFI):

ARTICLES

PUBLISHED ONLINE: 21 MARCH 2016 | DOI: 10.1038/NPHYS3700

nature physics

Measuring multipartite entanglement through dynamic susceptibilities

Philipp Hauke[1,2]*, Markus Heyl[1,2,3], Luca Tagliacozzo[4,5] and Peter Zoller[1,2]

- Hauke et al [Nat. Phys, 2016] realized that QFI of magnetic spins can be related to an integral over dynamic susceptibility, and can be experimentally measured.

$$f_Q(T) = \frac{4\hbar}{\pi}\int_0^\infty \mathrm{d}\,(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_BT}\right)\chi''(\hbar\omega, T)$$

Credit: Virgo

C) **Quantum Fisher Information**: average multipartite entanglement of a spin with its neighbors.

$$f_Q(T) = \frac{4\hbar}{\pi}\int_0^\infty \mathrm{d}(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_BT}\right)\chi''(\hbar\omega, T)$$

Gives a lower bound on entanglement:

$$\mathrm{nQFI} = \frac{f_Q}{12S^2} > m$$

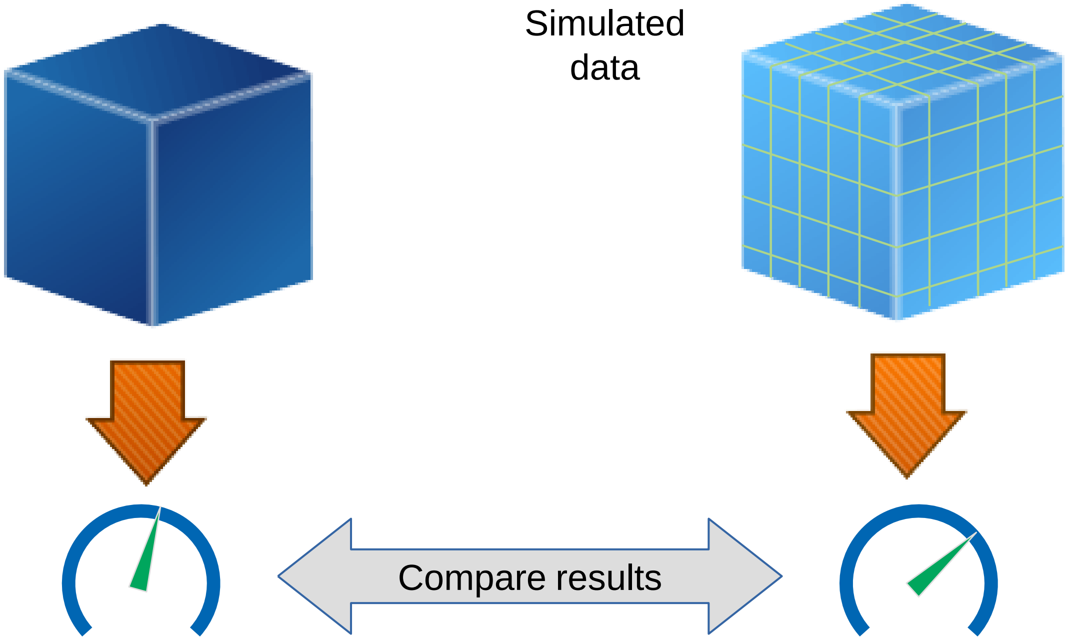
Conceptual definition: If a system has nQFI > m, it is *at least* m+1 partite entangled.

References:

- Hyllus et al, Phys. Rev. A **85**, 022321 (2012)
- Hauke et al, Nat. Phys. **12**, 778–782 (2016)

Strategy:

Benchmark **experimental** entanglement witnesses against **simulated** entanglement witnesses



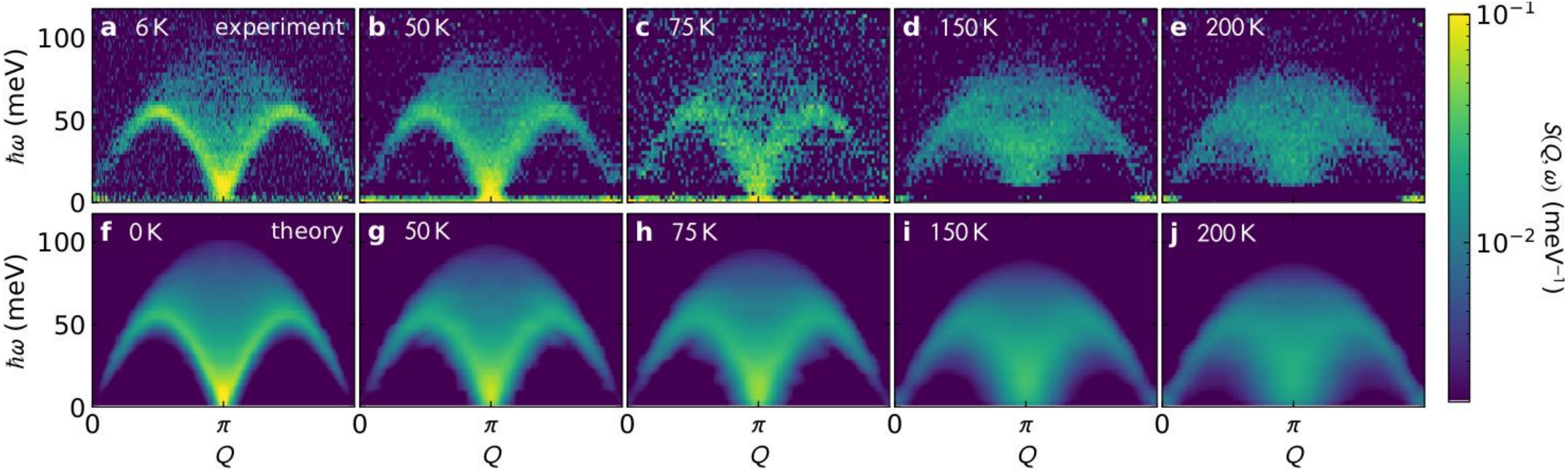
Experimental data

Simulated data

DMRG: finite T
Bethe Ansatz: zero T

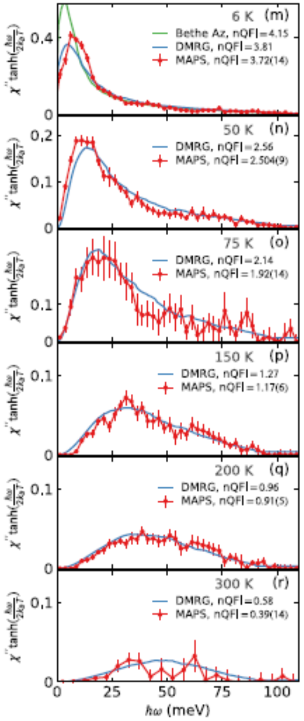
Compare results

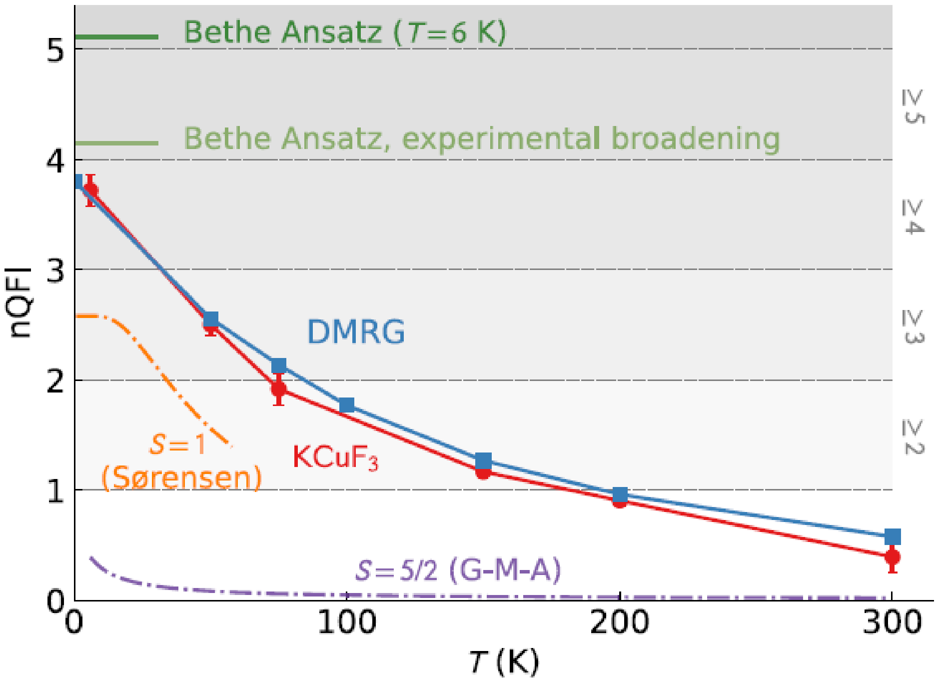
For 1D Heisenberg chain, there's excellent correspondence between DMRG theory and experiment



To simulate an idealized experiment, we apply resolution effects to DMRG

Quantum Fisher Information indicates > 4 partite entanglement at low T, in excellent agreement with theory



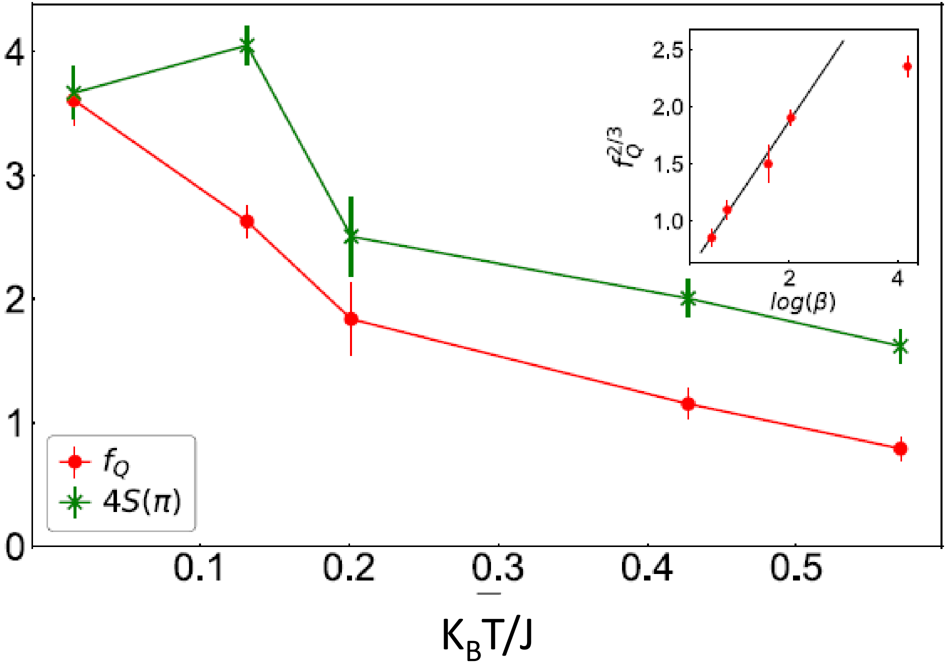


QFI also measured in Cs_2CoCl_4; see

P. Laurell et al., Phys. Rev. Lett. 127, 037201 (2021)

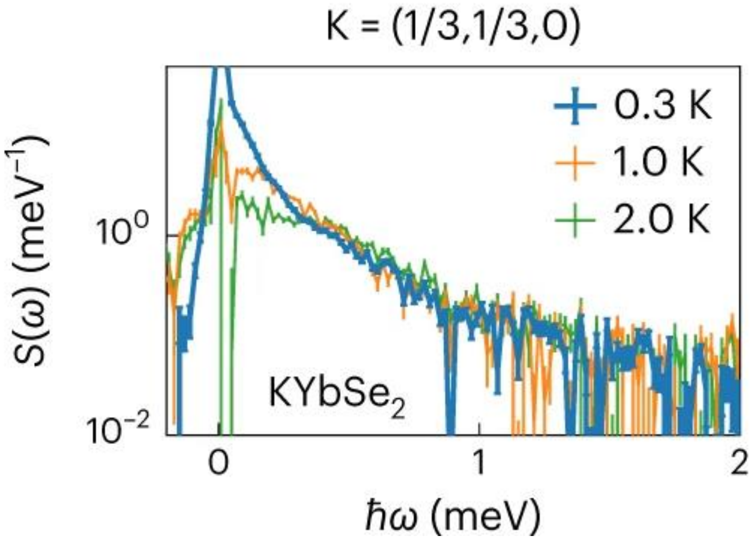
Menon *et al* (PRB **107**, 054422 (2023)) subsequently showed that the normalized QFI, or QFI density should scale as:

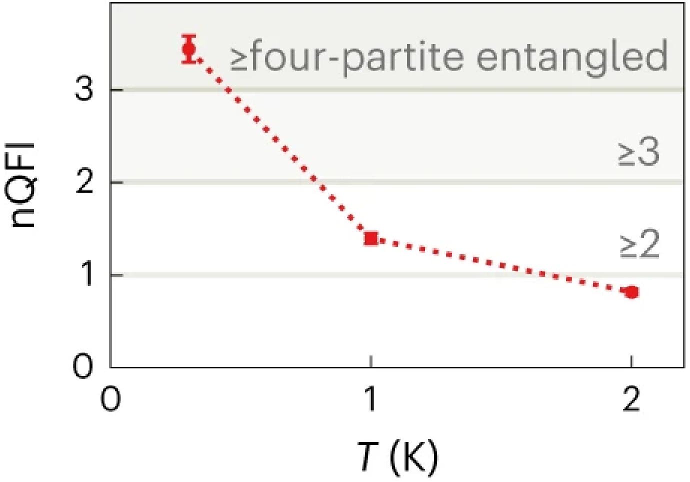
$$f_Q \sim \ln\left(\frac{1}{T}\right)^{3/2}$$



This agrees with $KCuF_3$ data except at very low T

Scheie *et al* (Nat. Phys. (2023)) https://doi.org/10.1038/s41567-023-02259-1 applied QFI to the triangular antiferromagnetic system KYbSe$_2$, showing > 4-partite entanglement at the lowest temperatures.





Working on a Ph.D.

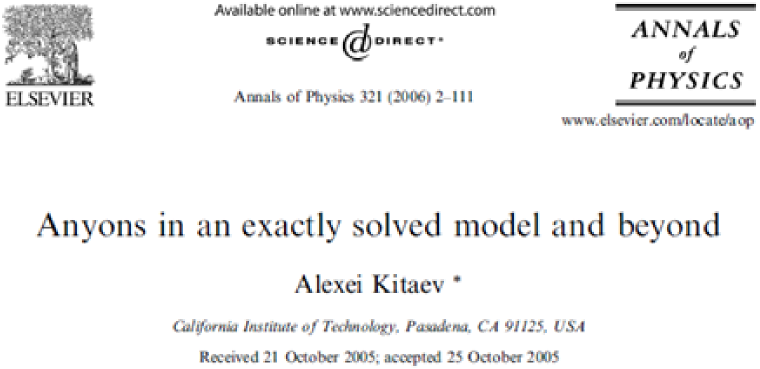
Ph.D. :
One learns more and more about less and less until they know everything about nothing!

Working on $RuCl_3$:

$RuCl_3$:
One learns less and less about more and more until they know nothing about anything!

Kitaev's model on honeycomb lattice – a special QSL

$$H_{\text{Kitaev}} = -\sum_{\gamma-\text{bonds}} K_\gamma \; S_i^\gamma S_j^\gamma$$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Annals of Physics 321 (2006) 2–111

ANNALS of PHYSICS

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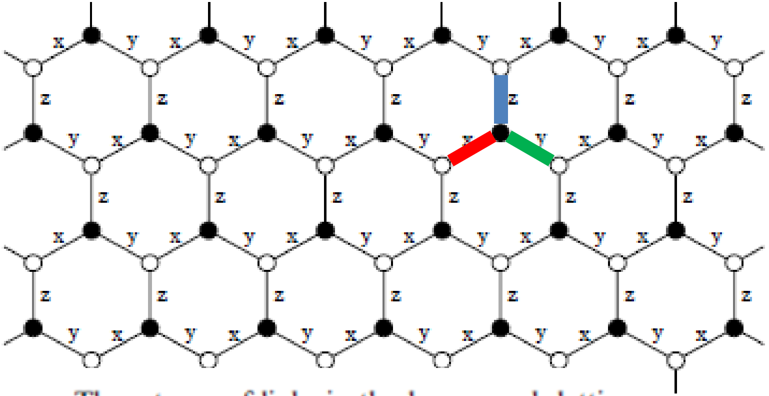
Anyons in an exactly solved model and beyond

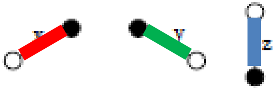
Alexei Kitaev *

California Institute of Technology, Pasadena, CA 91125, USA

Received 21 October 2005; accepted 25 October 2005



Three types of links in the honeycomb lattice.



- Kitaev interaction: Bond-directional dependent Ising coupling
- Exactly solvable Hamiltonian

→ ***quantum spin liquid*** ground state

Some features of the Kitaev QSL

- In real space, the only non-vanishing spin correlations are on-site and nearest neighbor

- Magnetic excitations are fractional and consist of "static" fluxes (visons) and mobile Majorana fermions

- The fractionalization of spins results in a specific heat C(T) with two separated peaks in temperature: a high T peak corresponding to itinerant MFs, and a low T peak corresponding to localized fluxes. Each peak carries entropy of 1/2*R*ln2

- Half-quantized thermal Hall conductivity $\kappa_{xy}/T = z(\pi/6)(k_B{}^2/\hbar)$

- Some response functions can be calculated exactly

Kitaev interactions in materials

PRL **102,** 017205 (2009) PHYSICAL REVIEW LETTERS week ending 9 JANUARY 2009

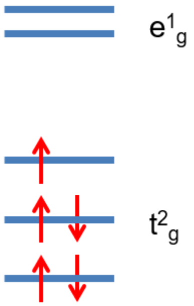
Mott Insulators in the Strong Spin-Orbit Coupling Limit: From Heisenberg to a Quantum Compass and Kitaev Models

G. Jackeli[1,*] and G. Khaliullin[1]

See also:
H. Takagi *et al.*, Nature Reviews Physics 1, (2019)

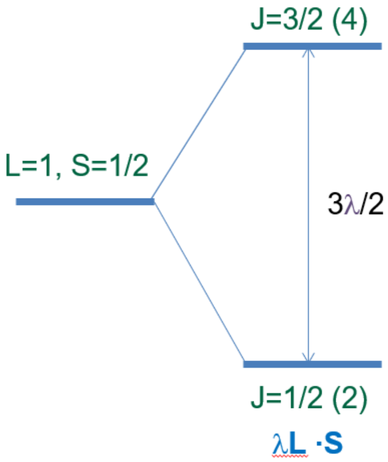
$$\mathcal{H}_{ij}^{(\gamma)} = 2KS_i^\gamma S_j^\gamma + J\boldsymbol{S}_i \cdot \boldsymbol{S}_j.$$

d^5 in low spin octahedral configuration

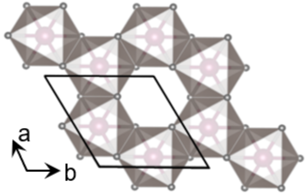


strong field limit
$S=1/2$, $L_{eff}=1$
e.g. ($5d^5$) Ir^{4+}
($4d^5$) Ru^{3+}

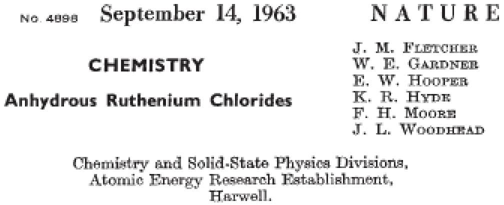
strong spin-orbit coupling



edge-sharing octahedra



α-$RuCl_3$: studied > 60 years ago



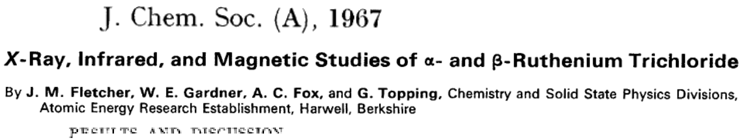
No. 4898 September 14, 1963 NATURE

CHEMISTRY

Anhydrous Ruthenium Chlorides

J. M. Fletcher
W. E. Gardner
E. W. Hooper
K. R. Hyde
F. H. Moore
J. L. Woodhead

Chemistry and Solid-State Physics Divisions,
Atomic Energy Research Establishment,
Harwell.



J. Chem. Soc. (A), 1967

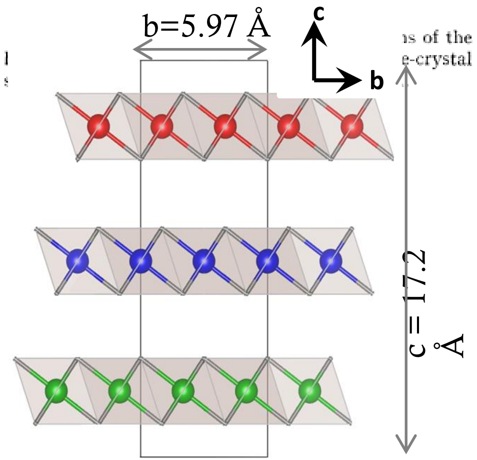
***X*-Ray, Infrared, and Magnetic Studies of α- and β-Ruthenium Trichloride**

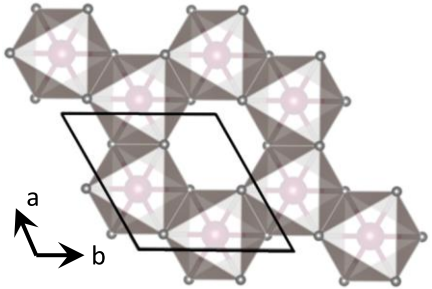
By **J. M. Fletcher, W. E. Gardner, A. C. Fox,** and **G. Topping,** Chemistry and Solid State Physics Divisions, Atomic Energy Research Establishment, Harwell, Berkshire

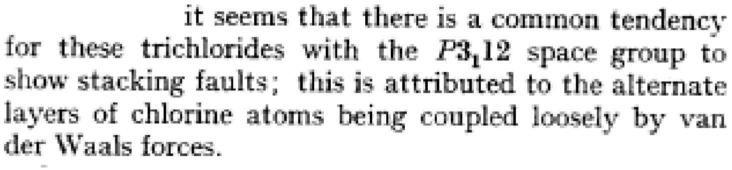
RESULTS AND DISCUSSION

α-$RuCl_3$:

- Honeycomb lattice
- Ru^{3+} (d^5) in octahedral low spin





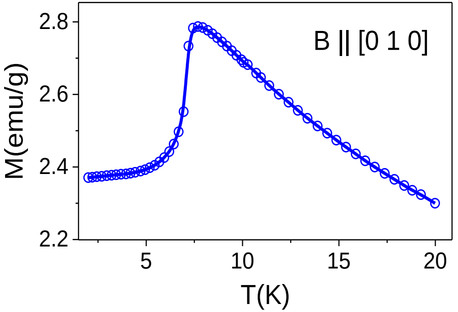


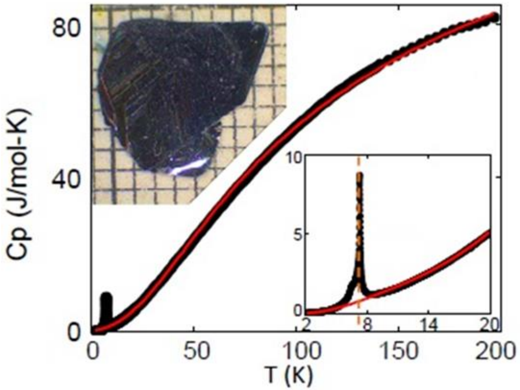
it seems that there is a common tendency for these trichlorides with the $P3_112$ space group to show stacking faults; this is attributed to the alternate layers of chlorine atoms being coupled loosely by van der Waals forces.

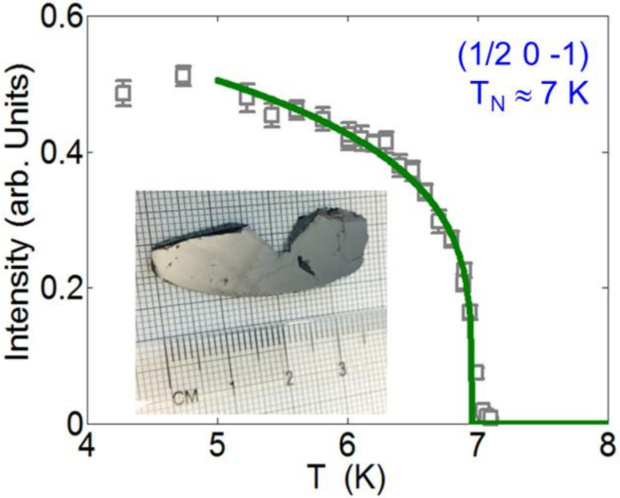
Magnetic order in α-RuCl$_3$

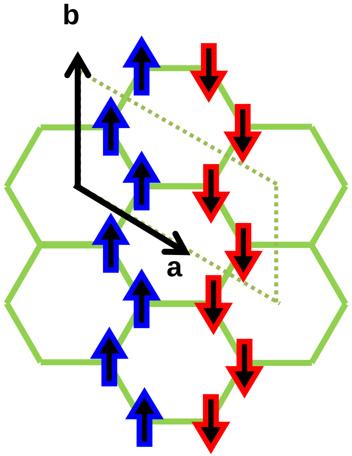
A. Banerjee, et al. Nature Mat. 15, 733 (2016)

Zigzag AFM order at low temperatures:







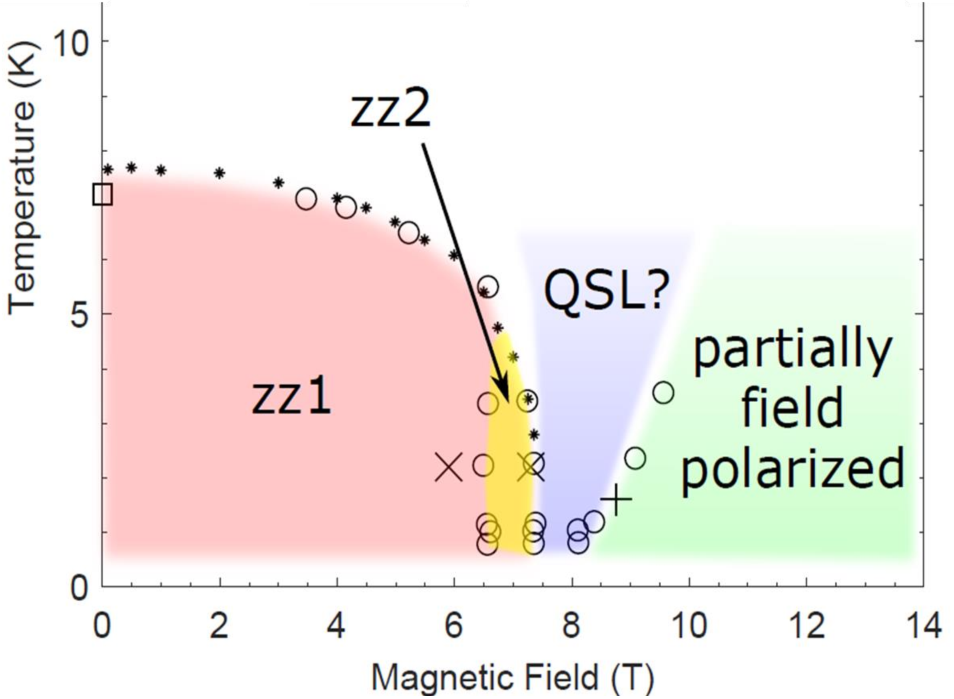


q=(1/2, 0, L)

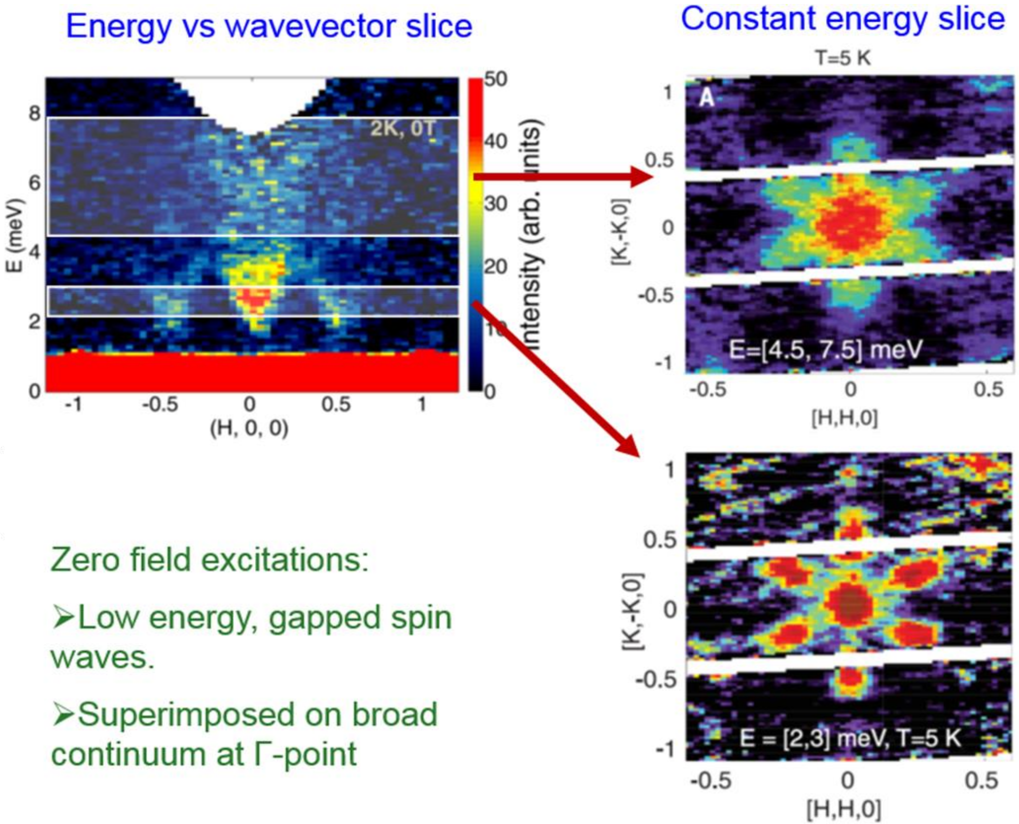
Temperature Field phase diagram

C. Balz et al., PRB **100**, 060405(R) (2019)

Field applied in the honeycomb plane, perpendicular to a Ru-Ru bond. Typically labeled as the “a” direction



α-RuCl$_3$ Inelastic Neutron Scattering


Energy vs wavevector slice

Constant energy slice

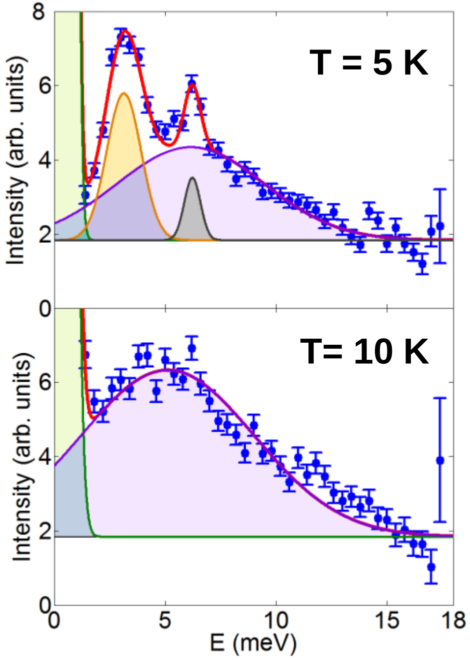
Zero field excitations:

- Low energy, gapped spin waves.
- Superimposed on broad continuum at Γ-point

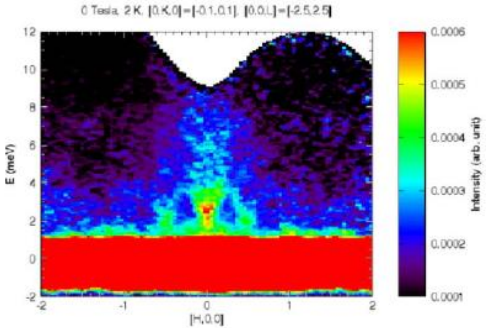
Banerjee et al., Science (2017)

T dependence of inelastic neutron scattering

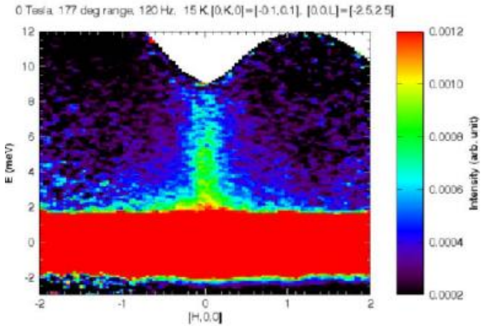
- At $T_N \approx 7$ K the spin waves disappear throughout the Brillouin zone
- Above T_N the continuum near the Γ point persists



T = 5 K



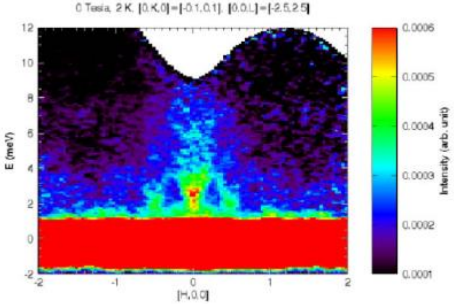
T = 15 K



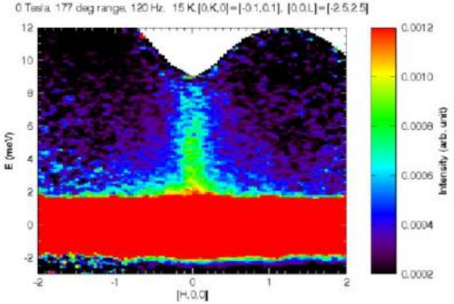
Banerjee et al., Science (2017),
Npj Quantum Materials (2018)

How does field affect the magnetic excitations?

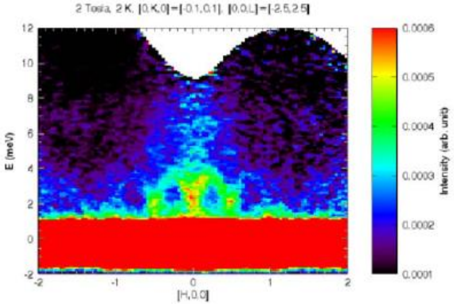
2K, 0T



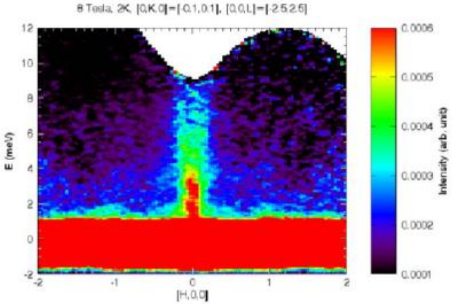
15K, 0T



2K, 2T



2K, 8T



Npj Quantum Materials **3**, 8 (2018).

Field dependence of scattering at specific Γ points

CHRISTIAN BALZ *et al.* PHYSICAL REVIEW B **100**, 060405(R) (2019)

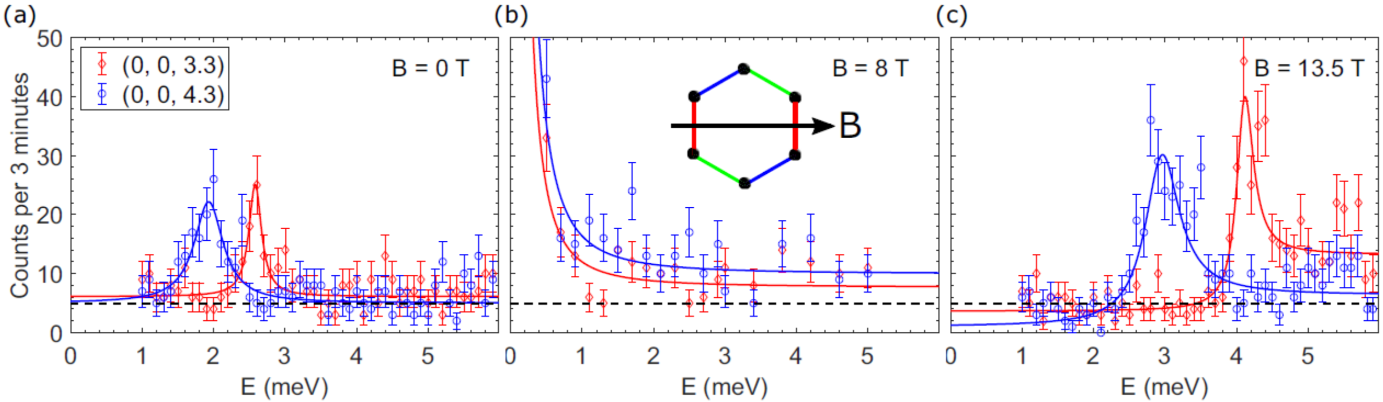
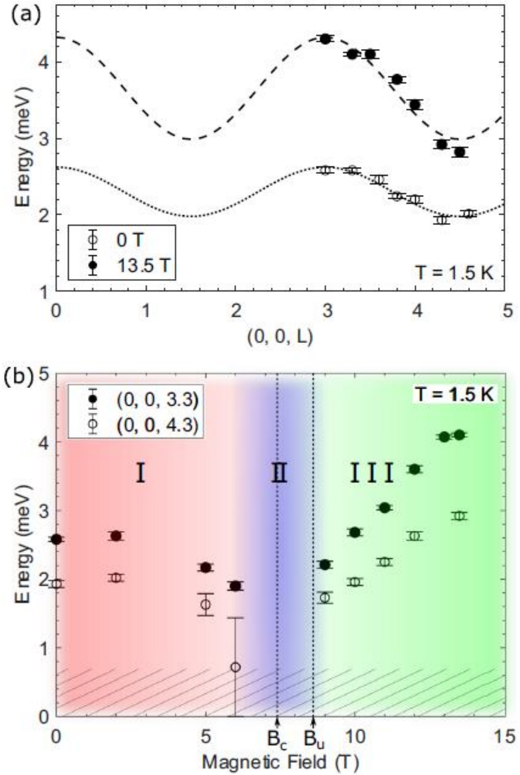


FIG. 1. Field dependence of the inelastic neutron scattering at the 2D Γ point for two values of the out-of-plane wave-vector transfer. Data obtained at 1.5 K on a 2 g single crystal of α-RuCl$_3$ using the FLEXX triple-axis spectrometer. (a) Zero-field data. A field of (b) 8 T and (c) 13.5 T was applied in the honeycomb plane perpendicular to a Ru-Ru bond [see inset of (b)]. The solid lines are fits and the dashed lines show the model free background for (0,0,3.3) as described in the text. Error bars represent one standard deviation assuming Poisson statistics.

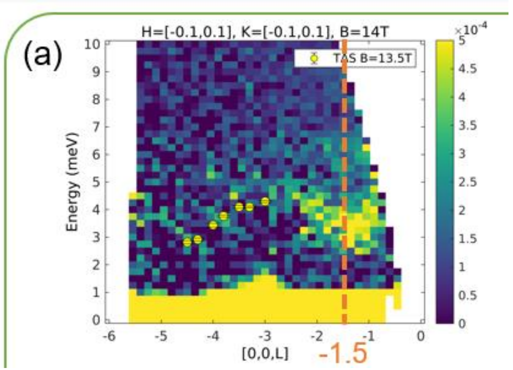
L dispersion and band width

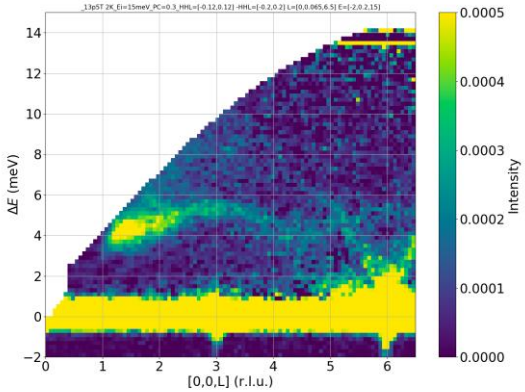


- The dispersion in L is a measure of the magnetic interactions perpendicular to the plane

- The reduction of the bandwidth near the region where magnons are not detected is a signature of enhanced two-dimensionality

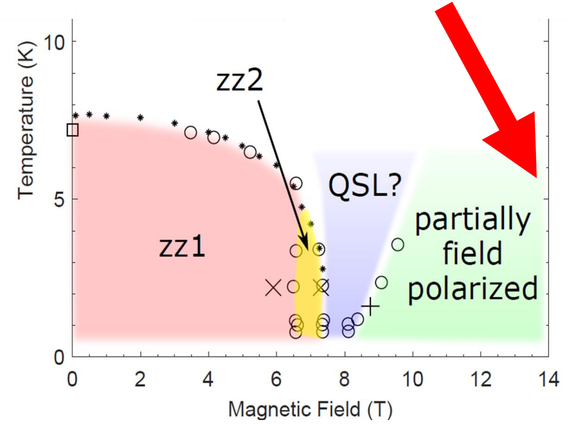
More recent data from ToF Measurements



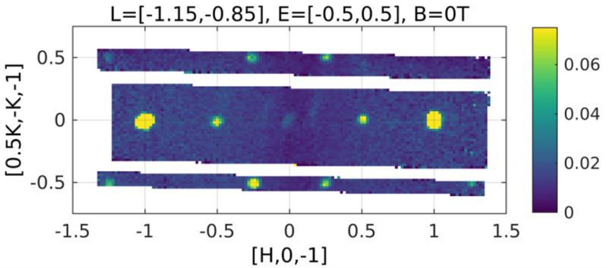


Measurements deep in partially polarized phase

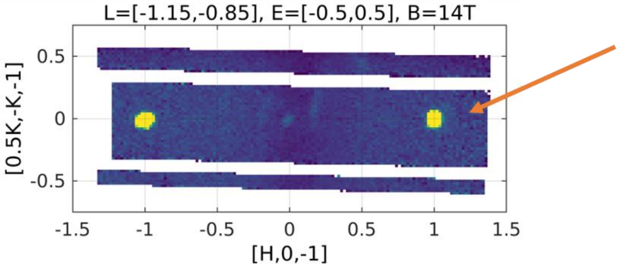
At 14 T the induced ferromagnetic moment is a few tenths μ_B per Ru

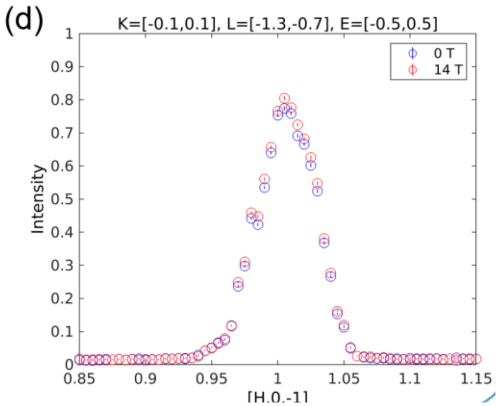


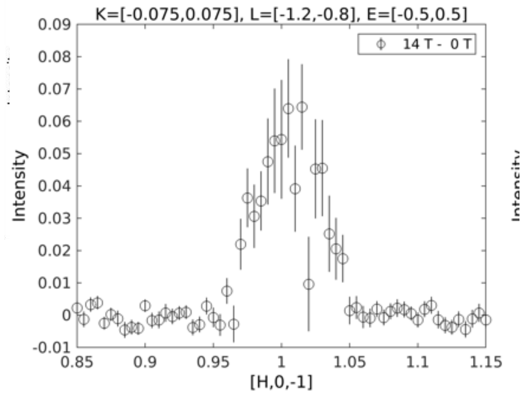
C. Balz et al., PRB **100**, 060405(R) (2019)



B = 14 T



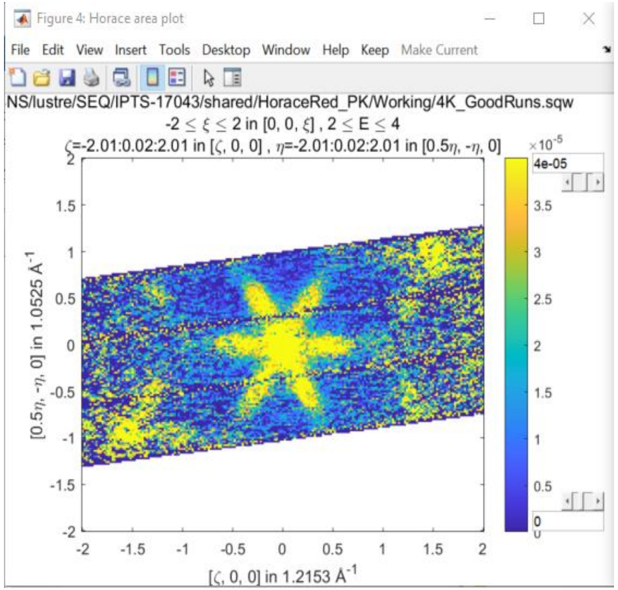




Measurements deep in partially polarized phase

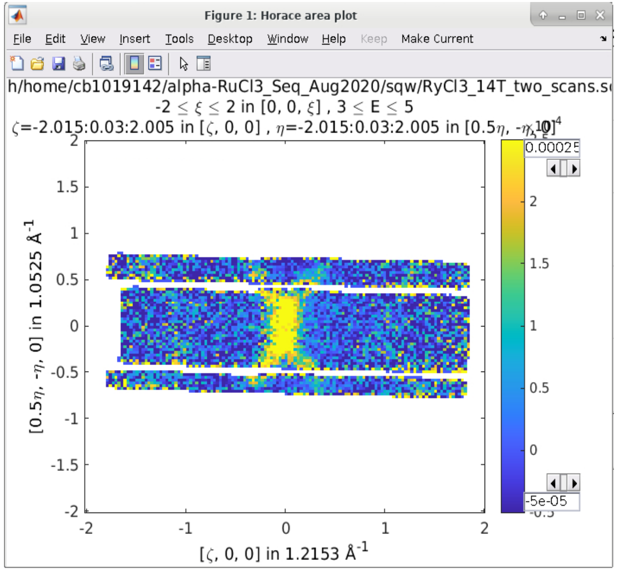
C. Balz et al., to be published

0 T

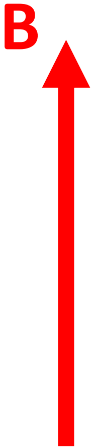


NS/lustre/SEQ/IPTS-17043/shared/HoraceRed_PK/Working/4K_GoodRuns.sqw
$-2 \leq \xi \leq 2$ in $[0, 0, \xi]$, $2 \leq E \leq 4$
ζ=-2.01:0.02:2.01 in $[\zeta, 0, 0]$, η=-2.01:0.02:2.01 in $[0.5\eta, -\eta, 0]$

14 T



h/home/cb1019142/alpha-RuCl3_Seq_Aug2020/sqw/RyCl3_14T_two_scans.s
$-2 \leq \xi \leq 2$ in $[0, 0, \xi]$, $3 \leq E \leq 5$
ζ=-2.015:0.03:2.005 in $[\zeta, 0, 0]$, η=-2.015:0.03:2.005 in $[0.5\eta, -\eta, 0]$



Summaries:

- QFI can be measured in experimental systems (maybe one should consider calculating it)
- We still don’t know the appropriate spin Hamiltonian describing α-$RuCl_3$ (but we’re working on it)

Thank you for your attention!

It’s time to get coffee