
Prof. Christian Rüegg :: Paul Scherrer Institute & University of Geneva
Head of Research Division Neutrons and Muons

# New Phases and Dynamics in Low-dimensional Quantum Magnets - Computation and Experiments

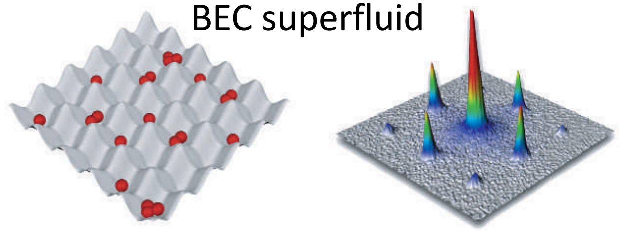
Quantum Matter: Computation Meets Experiments, Aspen-Villigen, 11th March 2020

# Quantum Materials as Solid-State Quantum Simulators

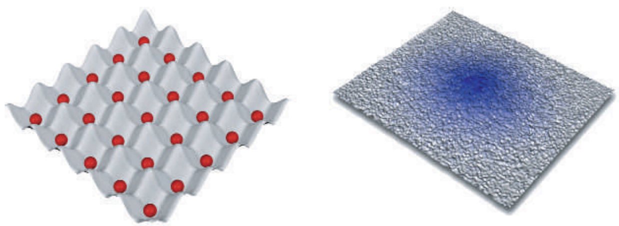
Nature may be described by a number of fundamental theoretical models. These are studied in collisions of high-energy particles, gases of ultra-cold atoms and in quantum materials e.g. by neutron scattering.

## Ultracold atoms

BEC superfluid



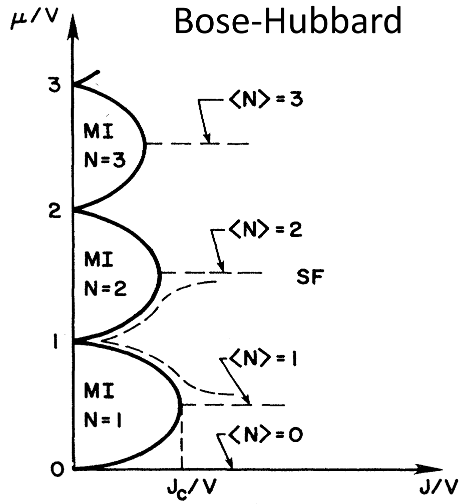
Mott insulating state



- M. Greiner *et al.*, Nature **415**, 39 (2002).
- I. Bloch, Nature Physics **1**, 23 (2005).

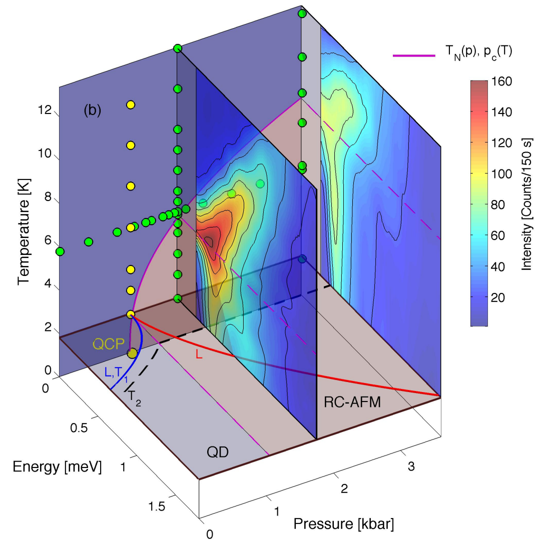
## Theory & Simulations

Bose-Hubbard




- M.P.A. Fisher *et al.*, Phys. Rev. B **40**, 546 (1989).

## Quantum magnets




- P. Merchant *et al.*, Nature Physics **10**, 373 (2014).

High-precision studies of new phases and dynamics by many established computational and experimental techniques:

- T. Giamarchi, Ch. Rüegg, O. Tchernyshyov, Nature Physics **4**, 198 (2008).
- C. Kollath, T. Giamarchi, and Ch. Rüegg, in *Universal Themes of BEC*, CUP (2017).

# Theory and Computation – QMC, ED, DMRG …

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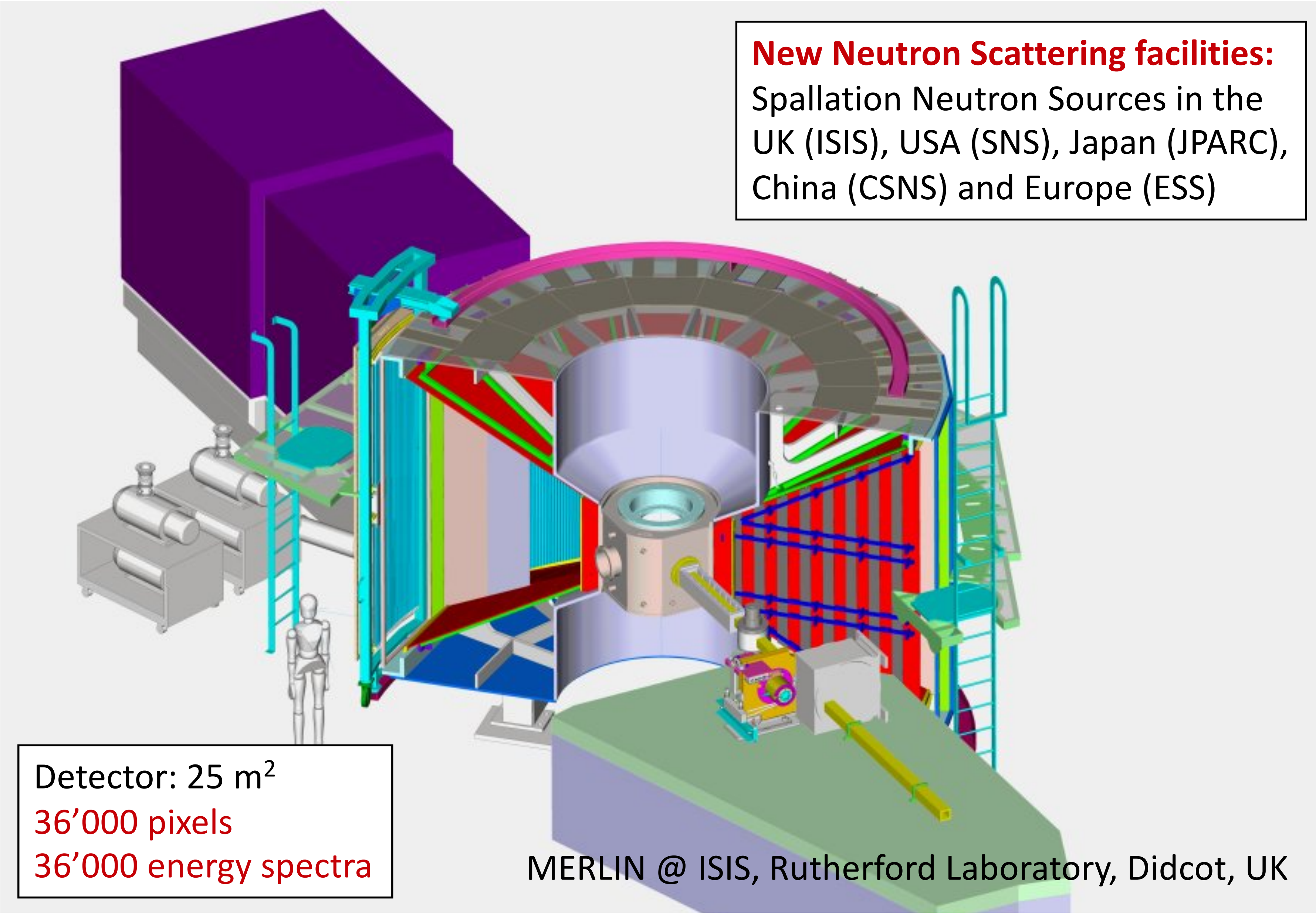
# Neutron Time-of-Flight Spectrometers

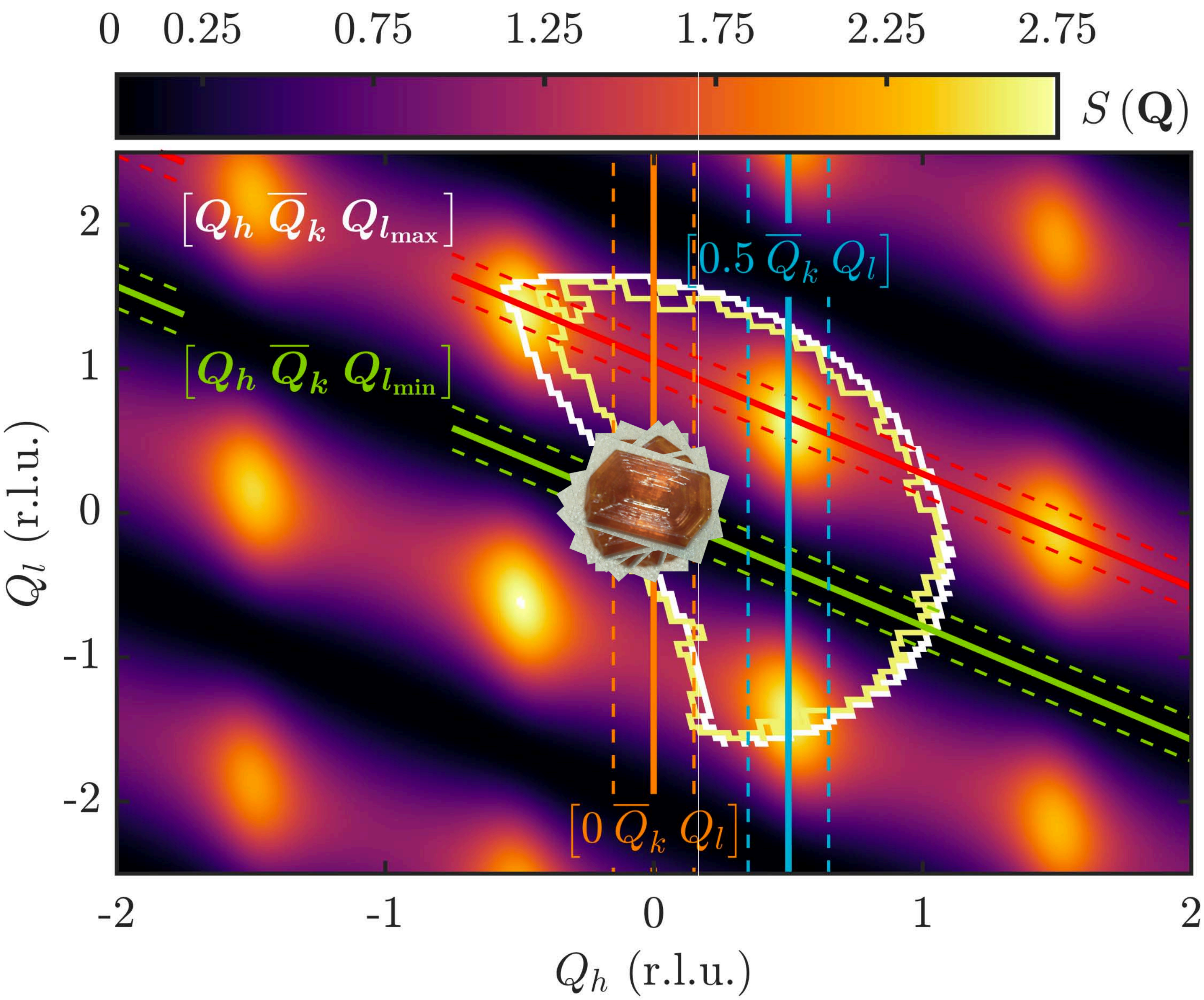


**New Neutron Scattering facilities:**
Spallation Neutron Sources in the UK (ISIS), USA (SNS), Japan (JPARC), China (CSNS) and Europe (ESS)

Detector: 25 $m^2$
36’000 pixels
36’000 energy spectra

MERLIN @ ISIS, Rutherford Laboratory, Didcot, UK

# Neutron Time-of-Flight Spectrometers



# Spallation Neutron Source SNS

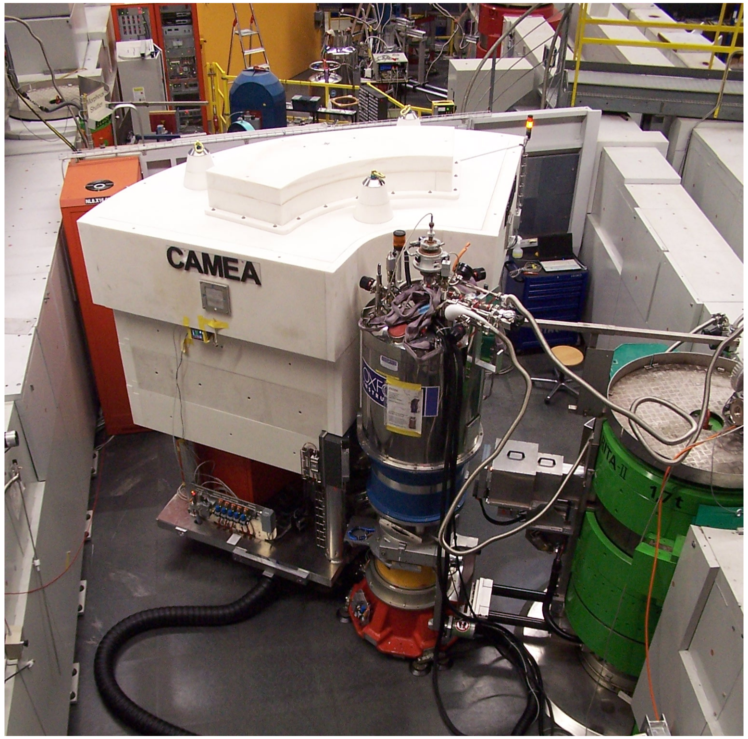
# European Spallation Source ESS

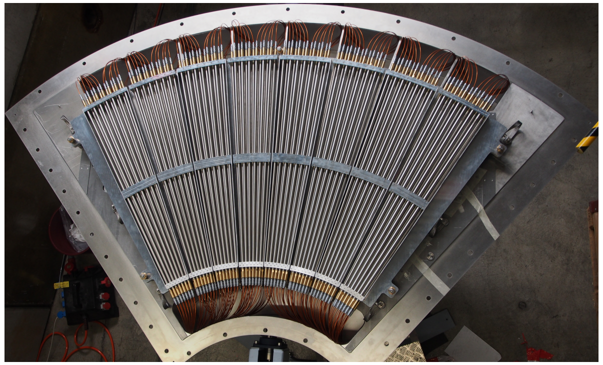
# Paul Scherrer Institute – ETH Organisation

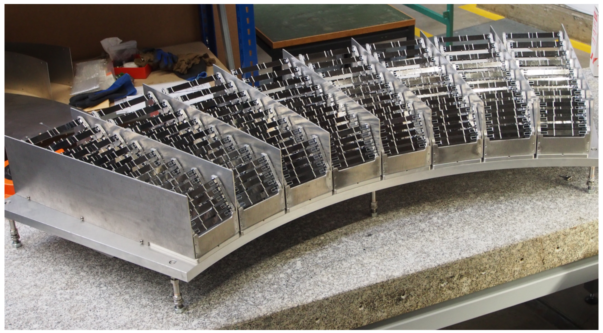
# New Spectrometer CAMEA at SINQ

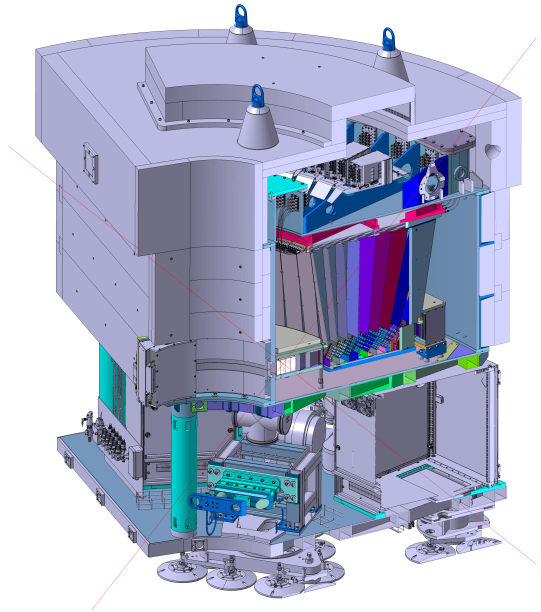
## Scientific Case

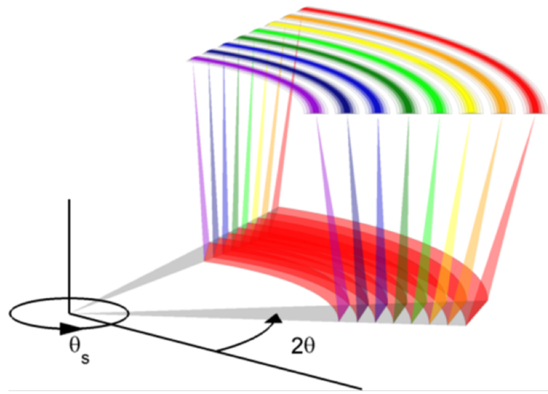
- **Small samples** of new emergent materials
- **Multi-extreme conditions** (temperature, **pressure**, **magnetic** and electric fields)









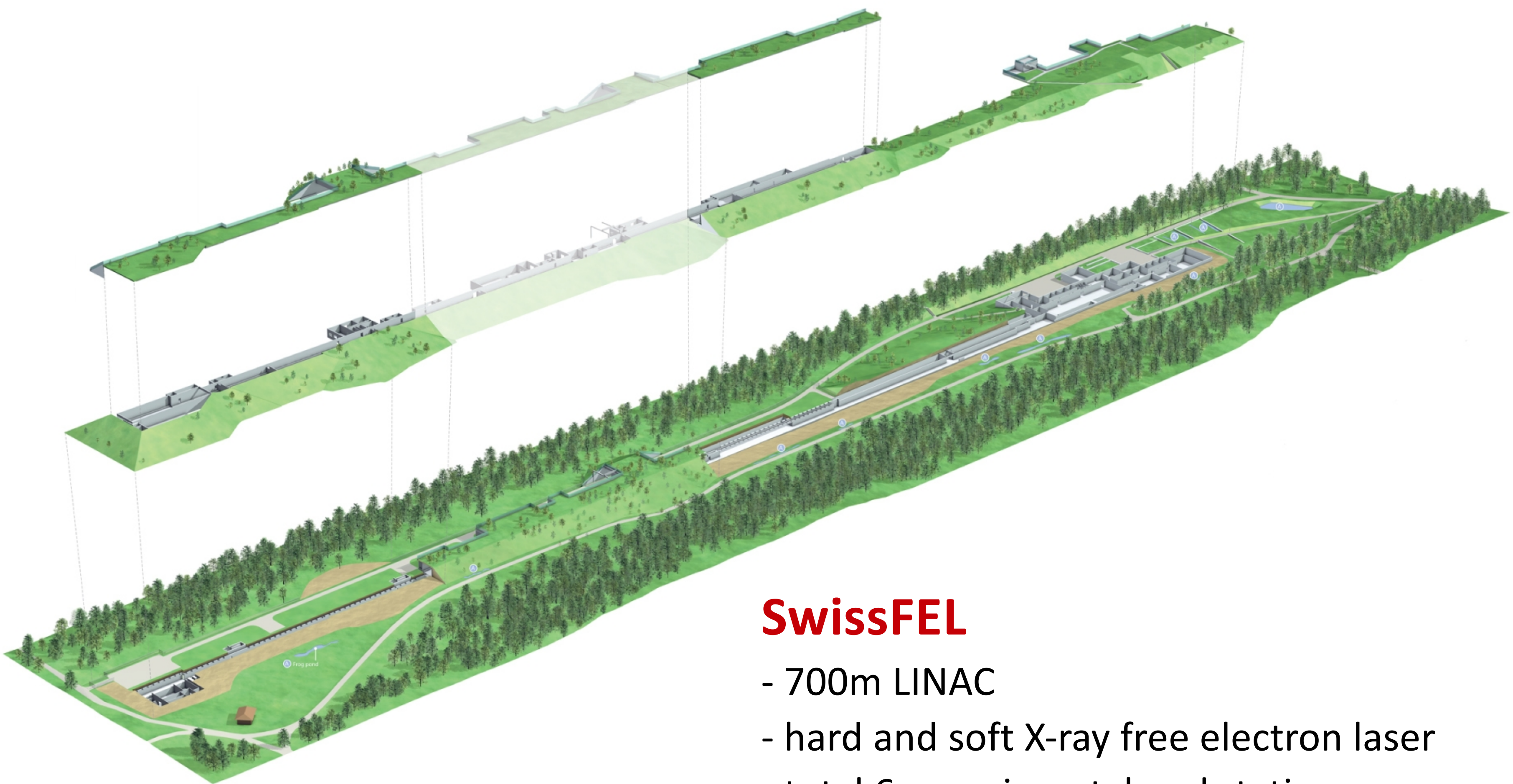

**Multiplex Neutron Spectrometer**

**Collaboration:** PSI-EPF Lausanne (H.M. Ronnow)

EPFL ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

# Swiss Free Electron Laser - SwissFEL

# Swiss Free Electron Laser - SwissFEL



**SwissFEL**

- 700m LINAC
- hard and soft X-ray free electron laser
- total 6 experimental end stations
- pilot experiments 2018 and users 2019

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# Outline

## New Phases and Dynamics in Low-dimensional Quantum Magnets

I: **Introduction:** New experimental opportunities

II: **Spin-Luttinger Liquid and Disorder in quasi-1D systems:**
Metal-organic spin-ladder $(C_5H_{12}N)_2CuBr_4$ and $(C_5H_{12}N)_2CuCl_4$
- S. Ward *et al.* PRL **118**, 177202 (2017).

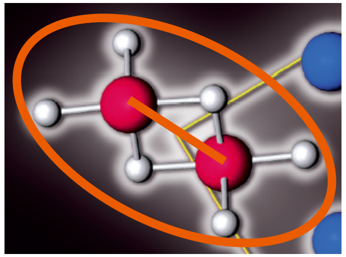
III: **BEC in quasi-2D systems:** Dimensional reduction in $BaCuSi_2O_6$
- S. Allenspach *et al.*, arXiv:1911.04161

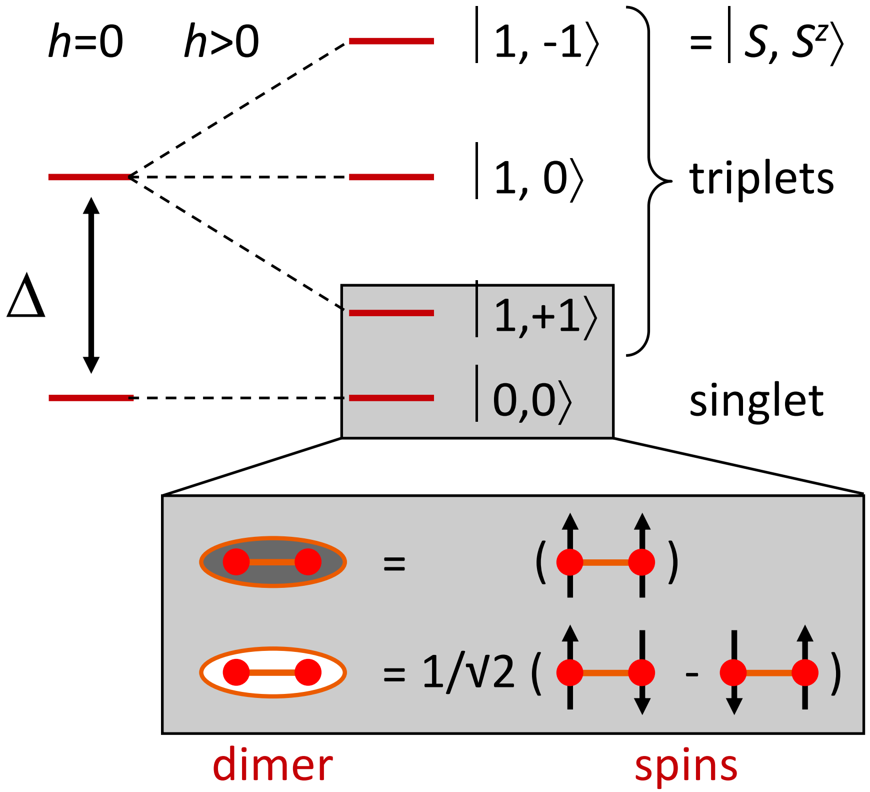
IV: **Out-of-equilibrium phenomena:** Pump-probe experiments using FELs

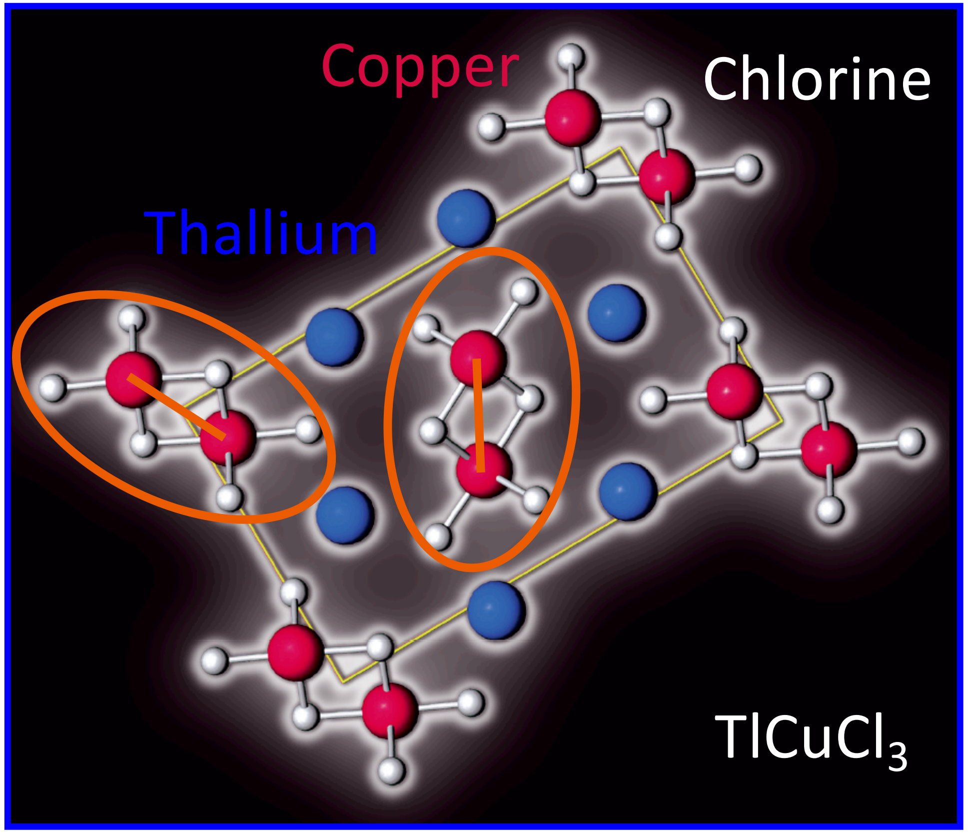
V: **Outlook and Conclusions**

# Quantum Spin Dimer Magnets

## Dimers in $TlCuCl_3$






- antiferromagnetic
- fluctuating moments
- no magnetic order
- “singlet” ground state

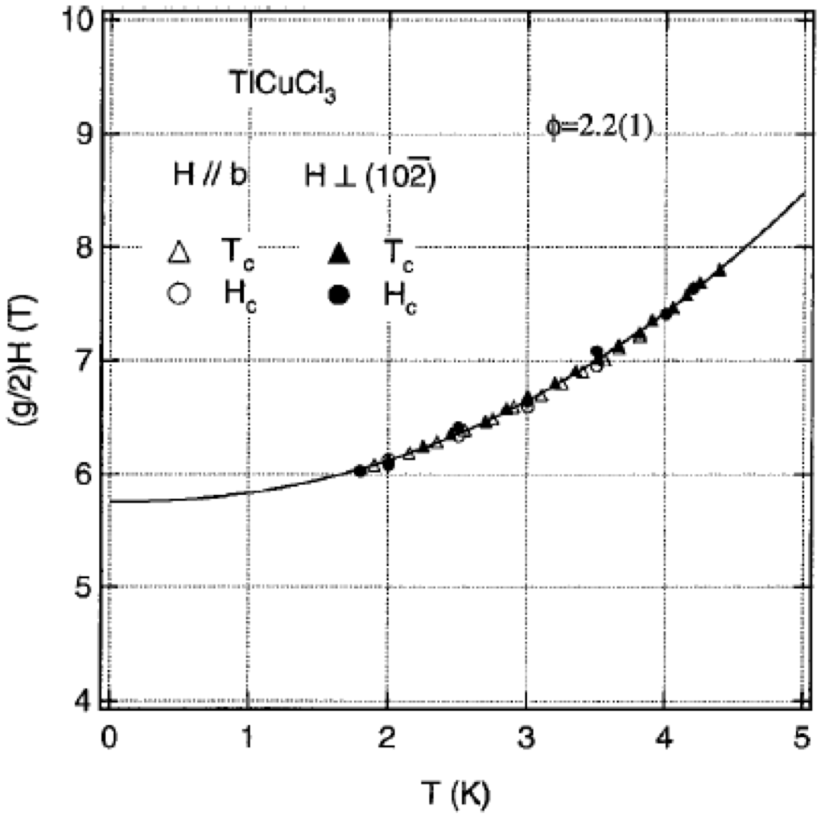
**SPIN SINGLETS**

# BEC in Condensed Matter Systems

VOLUME 84, NUMBER 25 PHYSICAL REVIEW LETTERS 19 JUNE 2000

## Bose-Einstein Condensation of Dilute Magnons in $TlCuCl_3$

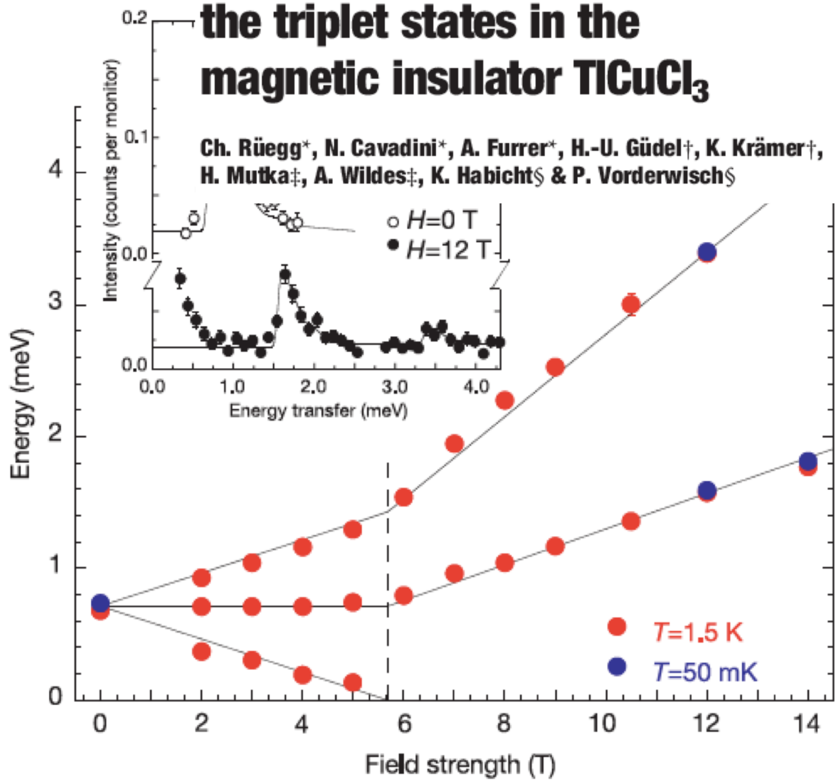
T. Nikuni,* M. Oshikawa, A. Oosawa, and H. Tanaka
*Department of Physics, Tokyo Institute of Technology, Oh-okayama, Meguro-ku, Tokyo 152-8551, Japan*
(Received 6 August 1999)




- T. Nikuni *et al.*, Phys. Rev. Lett. **84**, 5868 (2000).

## Bose–Einstein condensation of the triplet states in the magnetic insulator $TlCuCl_3$

Ch. Rüegg*, N. Cavadini*, A. Furrer*, H.-U. Güdel†, K. Krämer†, H. Mutka‡, A. Wildes‡, K. Habicht§ & P. Vorderwisch§




- Ch. Rüegg *et al.*, Nature **423**, 62 (2003).

# Spin-Singlet Quantum Magnets

**Spin-to-Boson Mapping:**

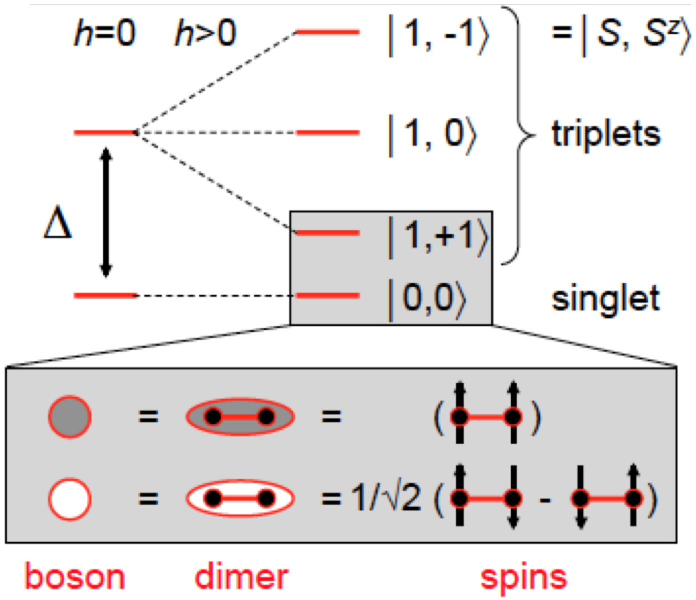
$$H_b = t \sum_{\langle i,j \rangle} \left[ b_i^\dagger b_j + \mathrm{h.c.} \right] + J_Z (n_i - 1/2)(n_j - 1/2) - \mu \sum_i (n_i - 1/2).$$

Direct control of boson Hamiltonian by magnetic field, pressure and chemistry:

$$H = \sum_{\langle i,j \rangle} \left( \frac{J_{XY}}{2} \left[ S_i^+ S_j^- + \mathrm{h.c.} \right] + J_Z S_i^z S_j^z \right) - h \sum_i S_i^z$$

$$S_{i,k}^+ \rightarrow \frac{1}{\sqrt{2}} (-1)^{i+k} b_i^\dagger$$

$$S_{i,k}^z \rightarrow \frac{1}{4} \left[ 1 + 2 \left( b_i^\dagger b_i - \frac{1}{2} \right) \right],$$




High-precision studies of BEC, Mott-insulating phases, Bose glass, supersolids in quantum magnets

- T. Giamarchi, Ch. Rüegg, O. Tchernyshyov, Nature Physics **4**, 198 (2008).
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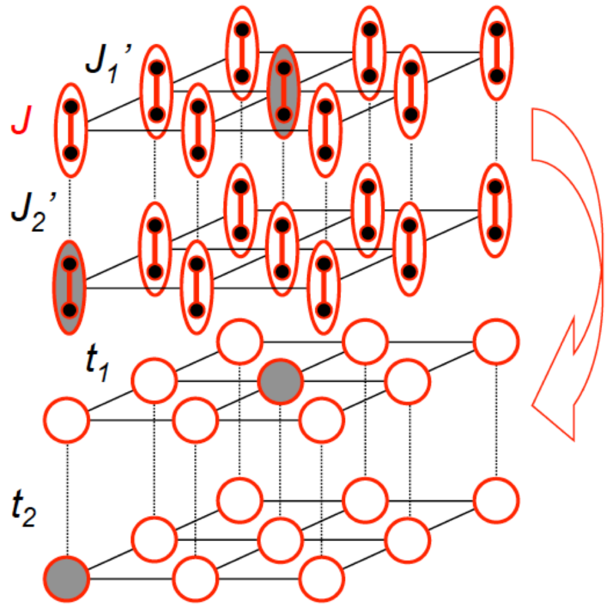


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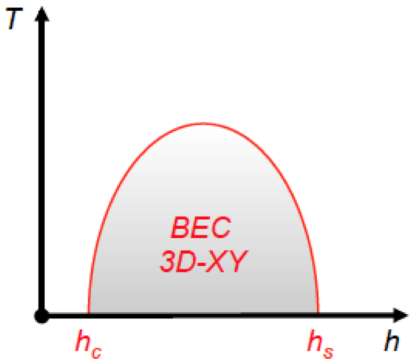
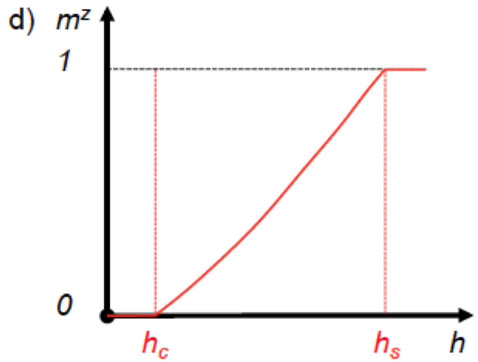
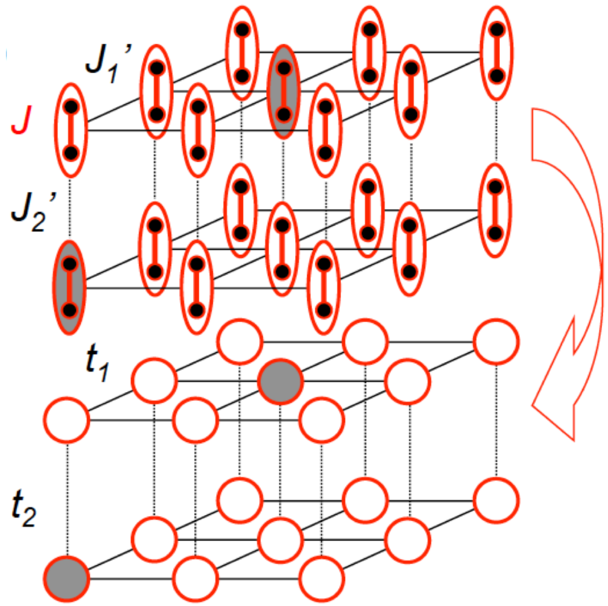
$$H_b = t \sum_{\langle i,j \rangle} \left[ b_i^\dagger b_j + \text{h.c.} \right] + J_Z (n_i - 1/2)(n_j - 1/2) - \mu \sum_i (n_i - 1/2).$$

Direct control of boson Hamiltonian by **magnetic field**, **pressure** and **chemistry**:

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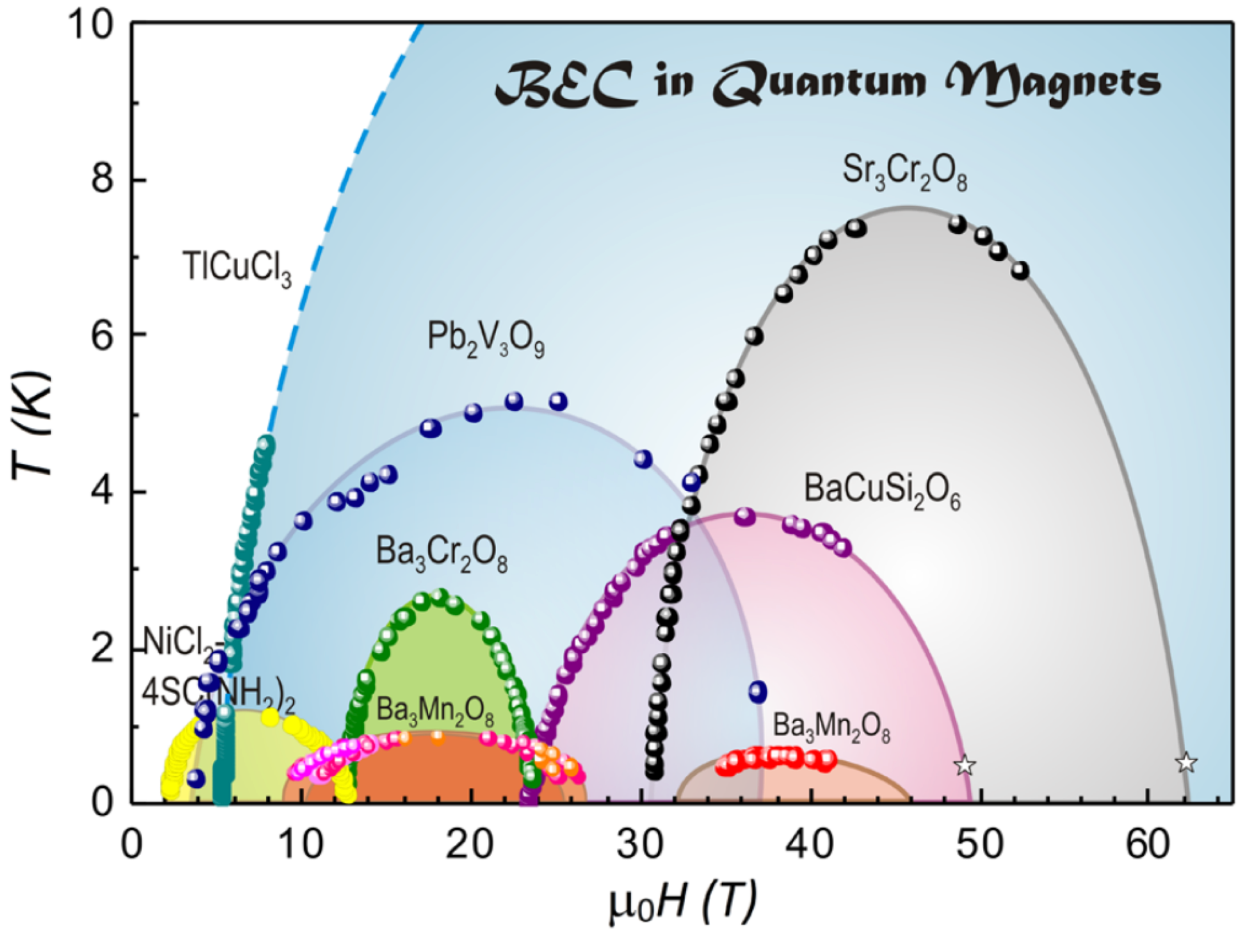
# BEC in Quantum Magnets



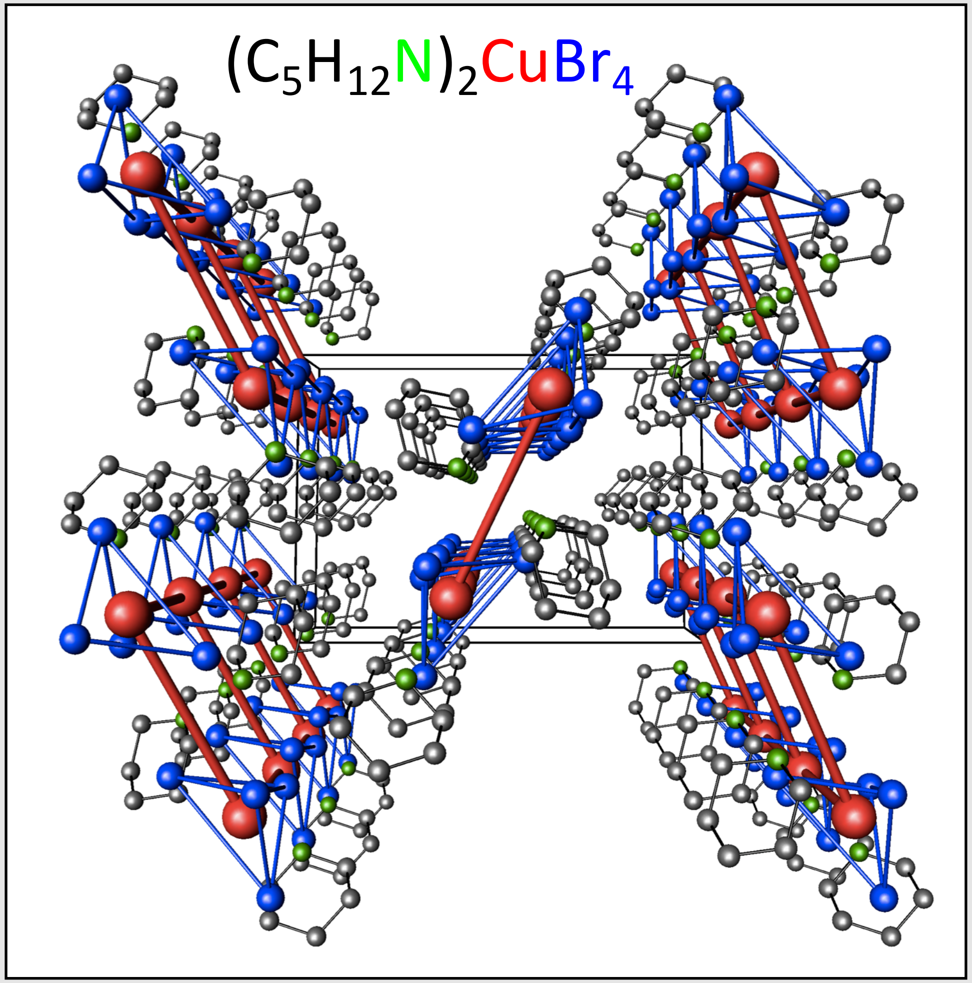

## BEC Magnets

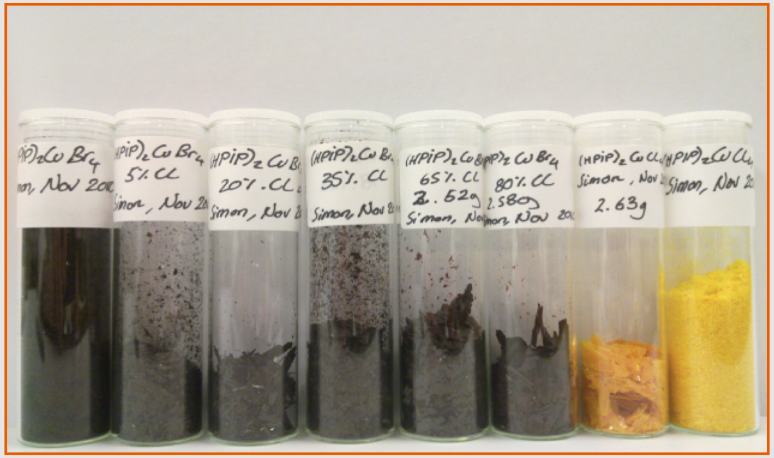
- $S$ = 1/2 dimer
- $S$ > 1/2 dimer
- $S$ = 1 Haldane
- $S$ = 1, large aniso. D

## Dimensionality ?
## Disorder ?
## Frustration ?
## Out of Equilibrium?
## Metals ?

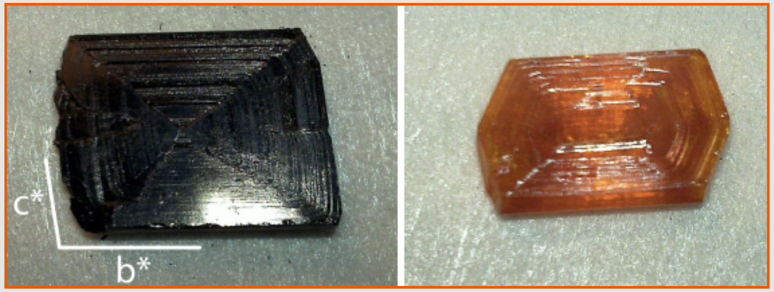
- V. Zapf, M. Jaime, C.D. Batista, Rev. Mod. Phys. **86**, 563 (2014).

# Dimers in Metal-Organic Quantum Materials



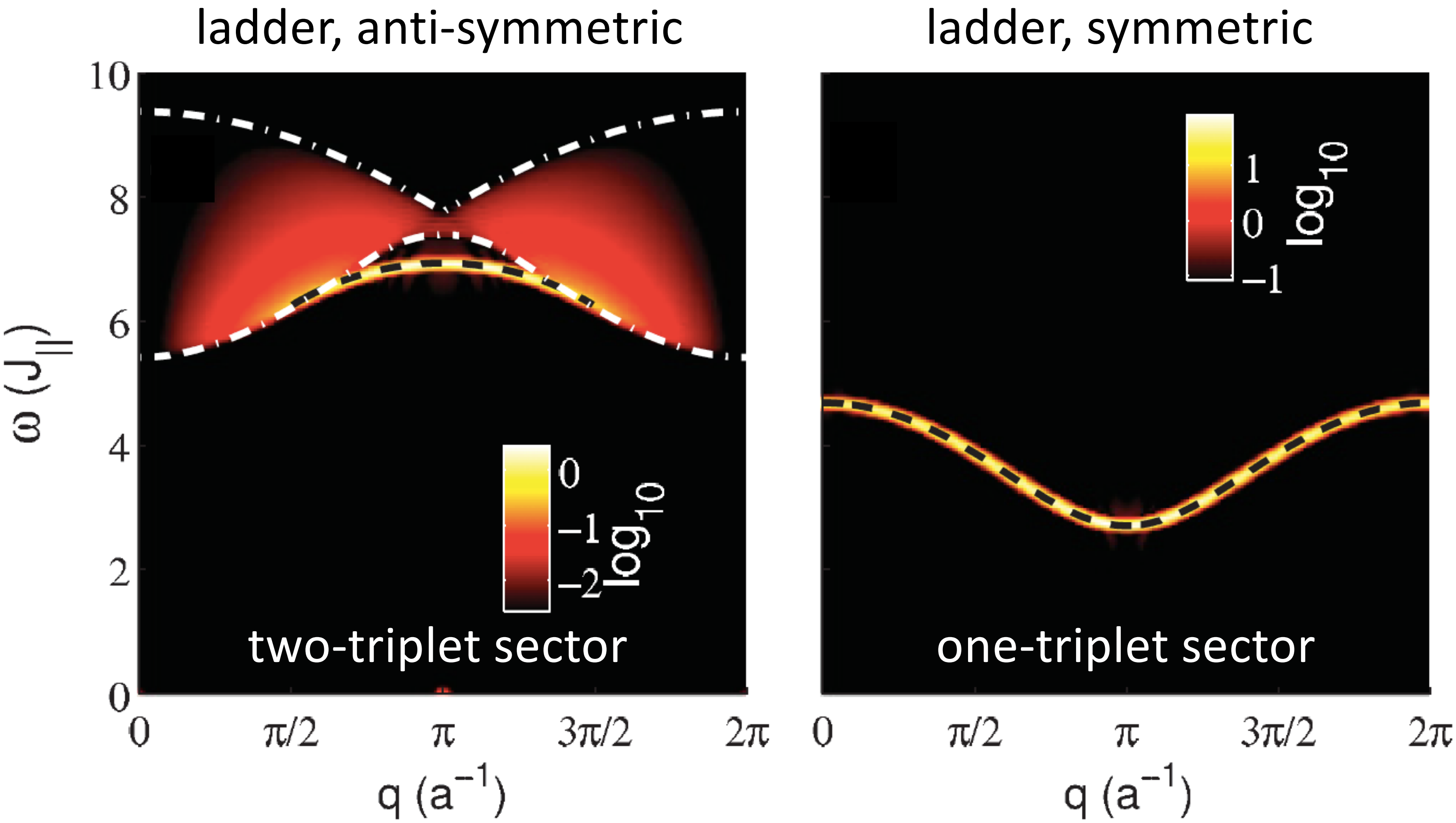
$(C_5H_{12}N)_2CuBr_4$ → $(C_5H_{12}N)_2CuCl_4$




K. Krämer, University of Bern

Dimers form one-dimensional spin-ladder structure

# DMRG: Complete Ladder Excitation Spectrum

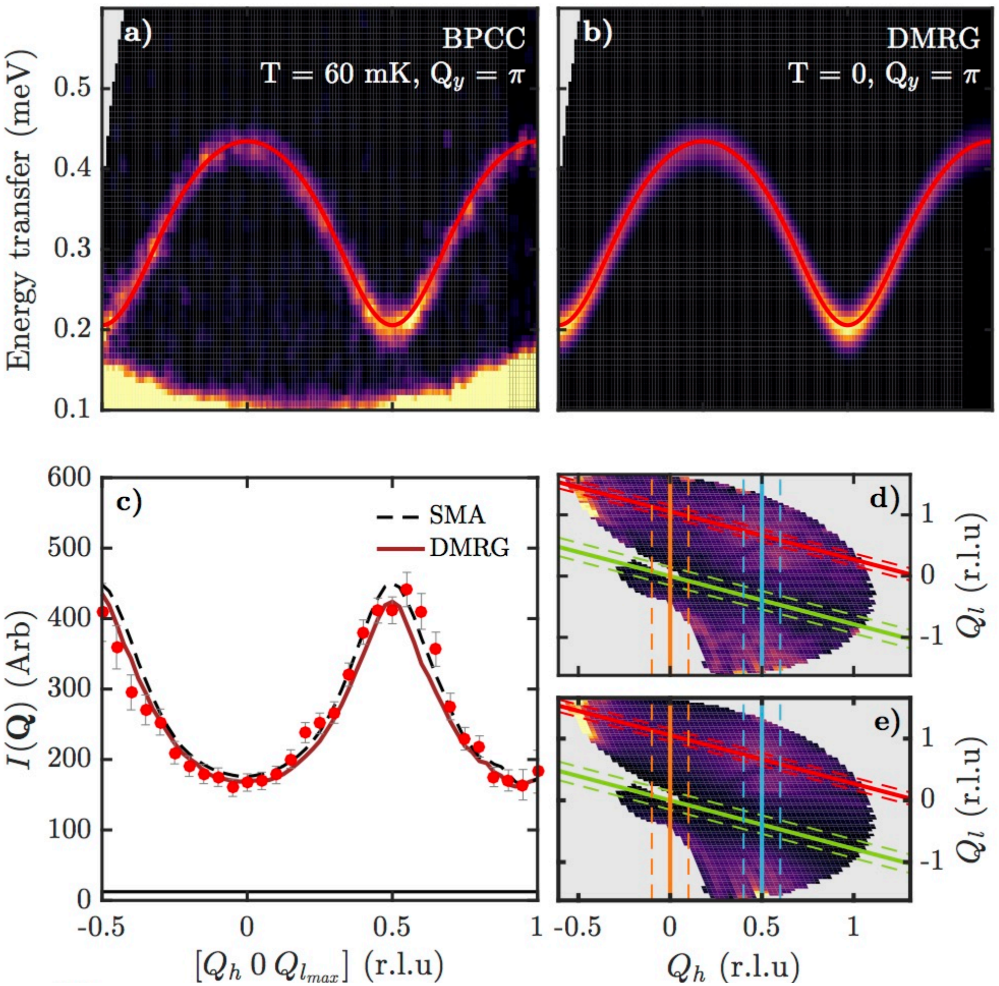


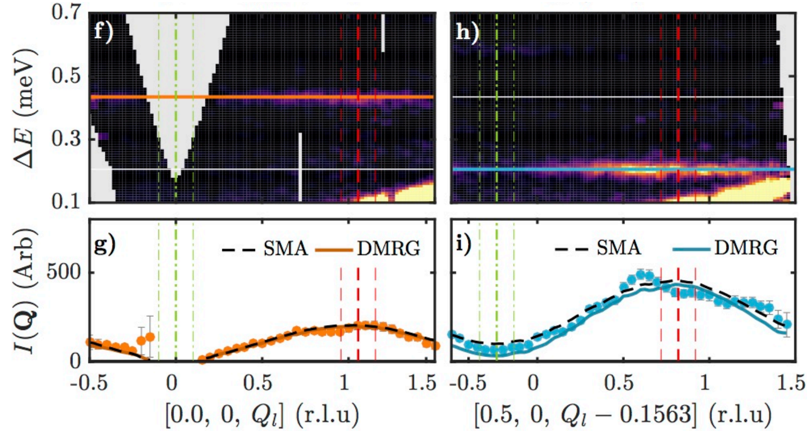

New calculations of dynamical structure factors of the full ladder:

**DMRG:** P. Bouillot, C. Kollath, A. Läuchli, T. Giamarchi *et al.*, Phys. Rev. B **83**, 054407 (**2011**).

# Quantum Spin Ladder – $(C_5H_{12}N)_2CuCl_4$

one-triplet band, anti-symmetric



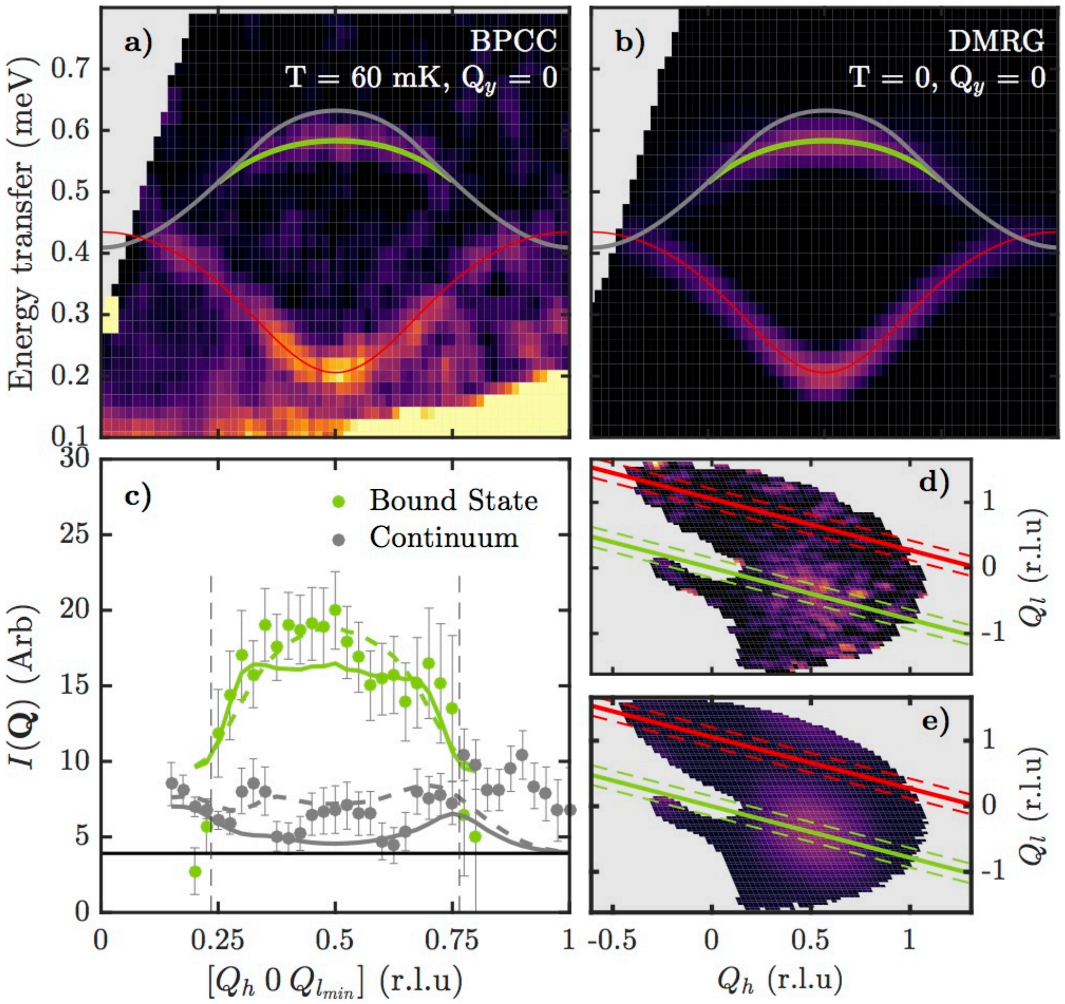




$(C_5H_{12}N)_2CuCl_4$
$J_r = 0.295(8)$ meV, $J_l = 0.116(1)$ meV
$J' < 0.006$ meV, $J_r/J_l \approx 2.5$

$(C_5H_{12}N)_2CuBr_4$
$J_r = 1.108(3)$ meV, $J_l = 0.288(3)$ meV
$J' = 0.007(3)$ meV, $J_r/J_l \approx 3.9$

- S. Ward, Ch. Rüegg, B. Normand, K. Schmidt, C. Kollath, Th. Giamarchi, *et al.* PRL **118**, 177202 (2017).

# Quantum Spin Ladder – $(C_5H_{12}N)_2CuCl_4$




two-triplet bound state, symmetric

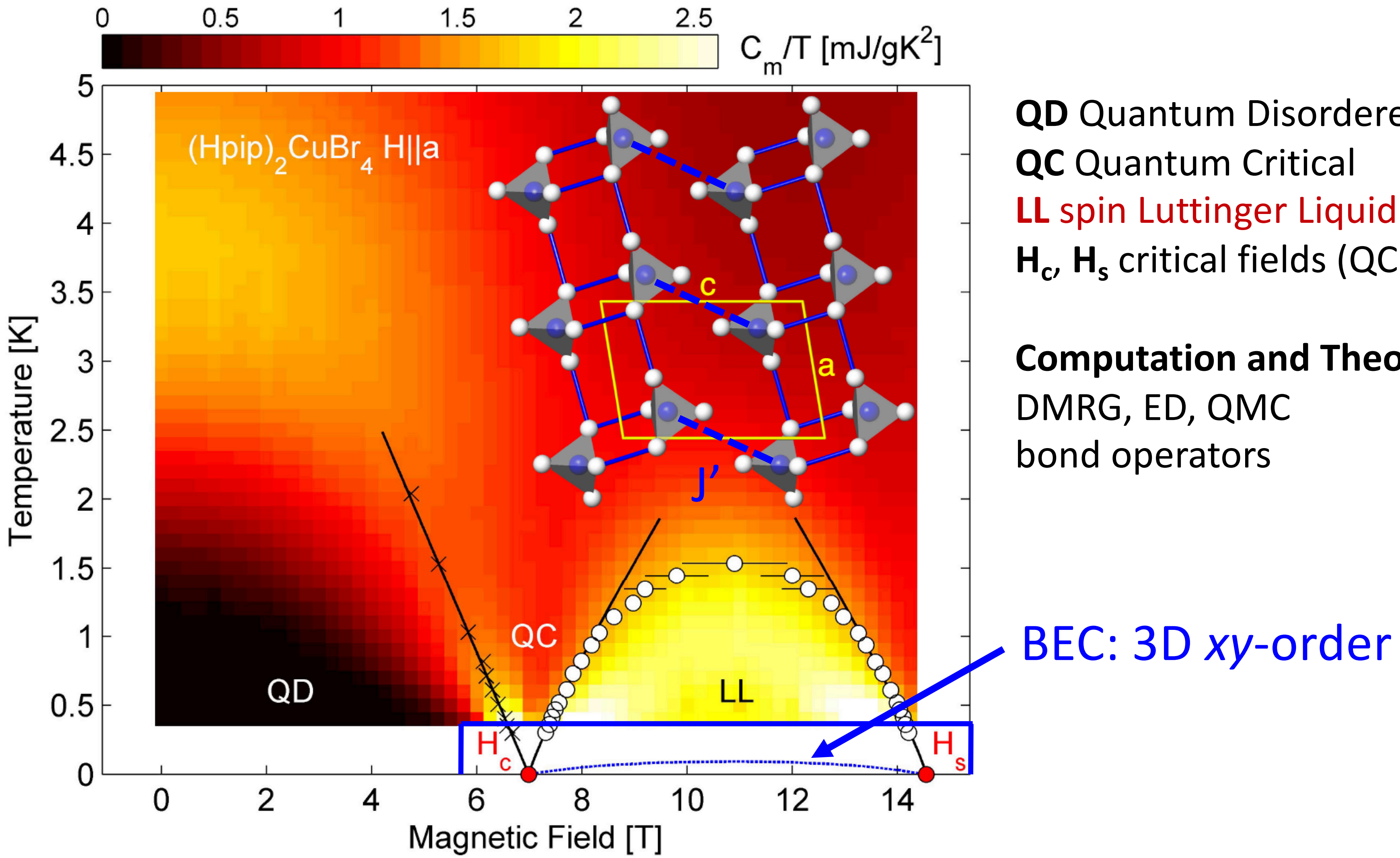
one-triplet band,
anti-symmetric
(finite integration width)

structure factor (exp)

structure factor (calc)

- S. Ward, Ch. Rüegg, B. Normand, K. Schmidt, C. Kollath, Th. Giamarchi, *et al.* PRL **118**, 177202 (2017).

# Spin Luttinger-liquid and BEC in 1D Magnets

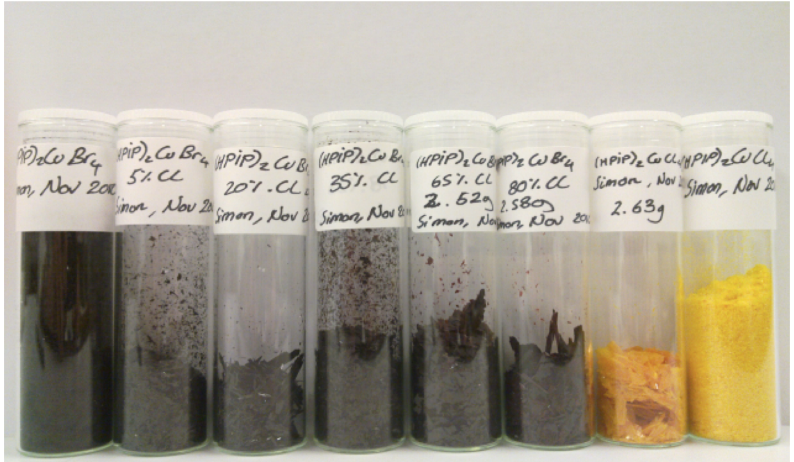



**QD** Quantum Disordered
**QC** Quantum Critical
**LL** spin Luttinger Liquid
**$H_c$, $H_s$** critical fields (QCPs)

**Computation and Theory:**
DMRG, ED, QMC
bond operators


- Ch. Rüegg *et al.,* Phys. Rev. Lett. **101**, 247202 (2008).
- B. Thielemann *et al.,* Phys. Rev. B **79**, 020408(R) (2009).
- H. Ryll *et al.* Phys. Rev. B **89**, 144416 (2014).
- S. Ward *et al.* Phys. Rev. Lett. **118**, 177202 (2017).

# Quenched Disorder – $(C_5H_{12}N)_2CuBr_{4-x}Cl_x$



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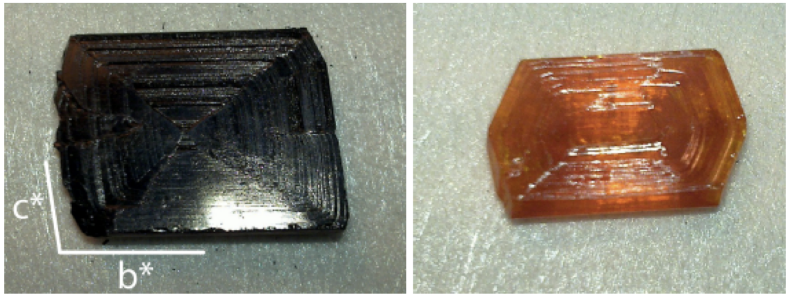
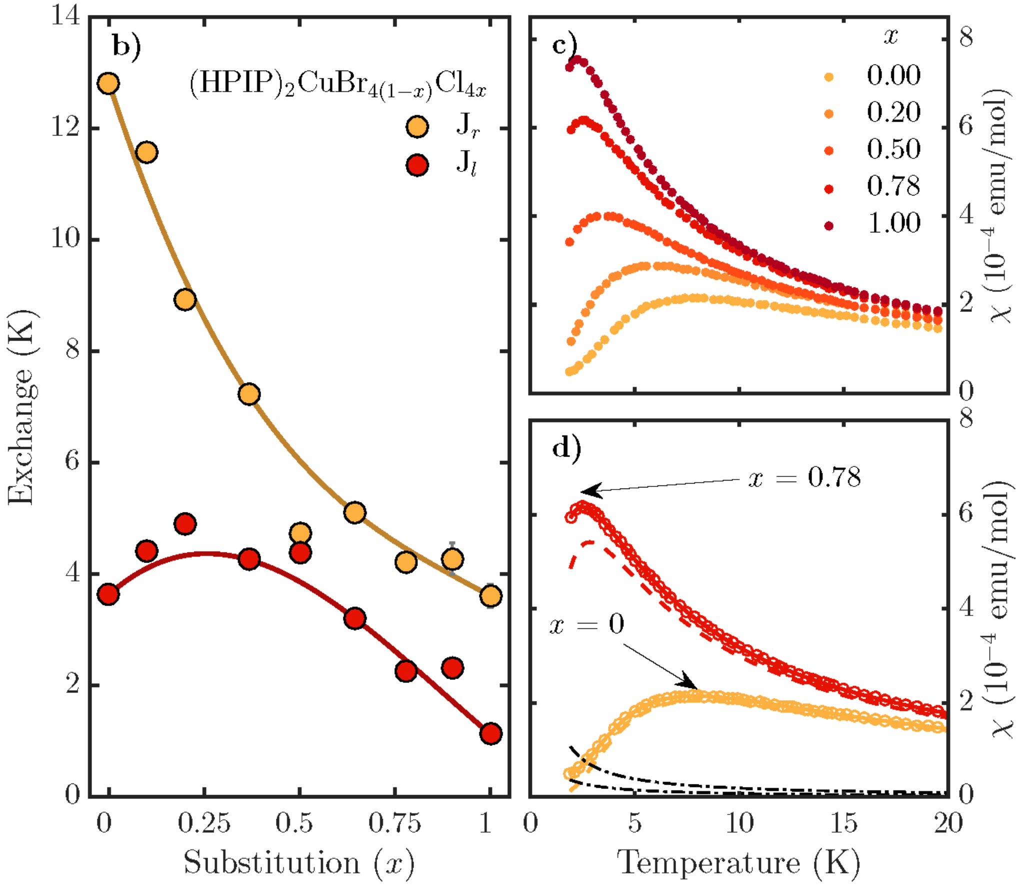
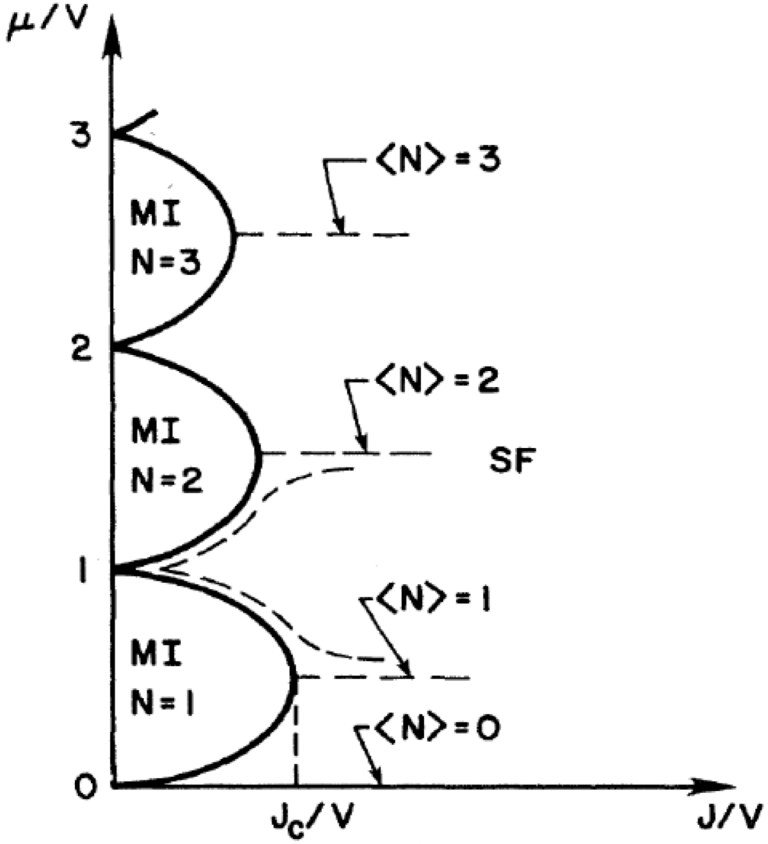


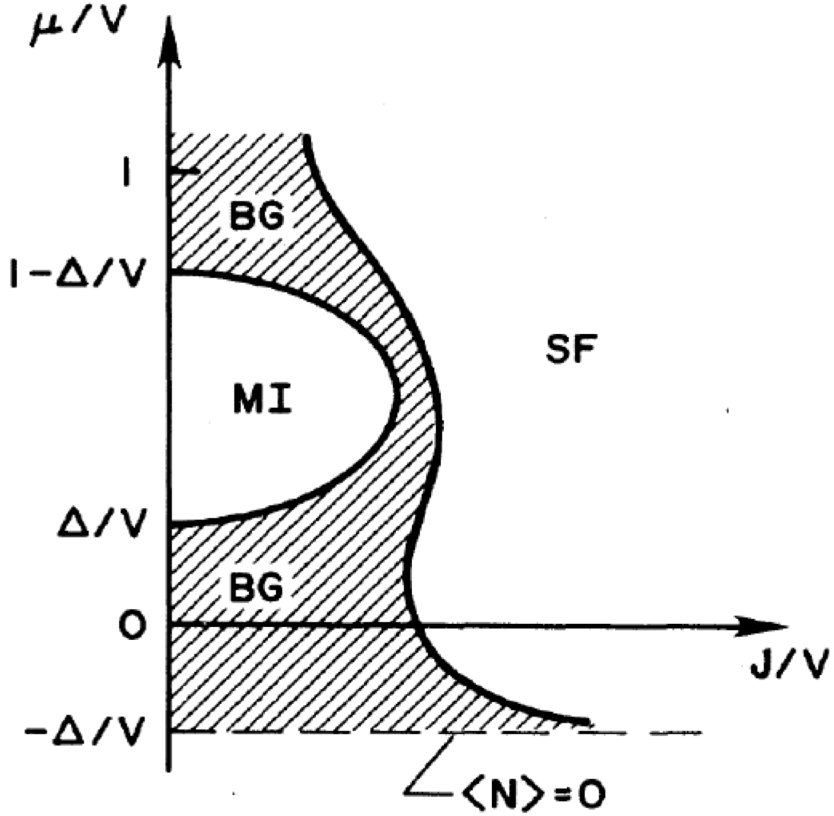

• S. Ward, Ch. Rüegg, S. Furuya, Th. Giamarchi, M. Höhrmann, K. Schmidt *et al.*

# Theory

Bose-Hubbard model

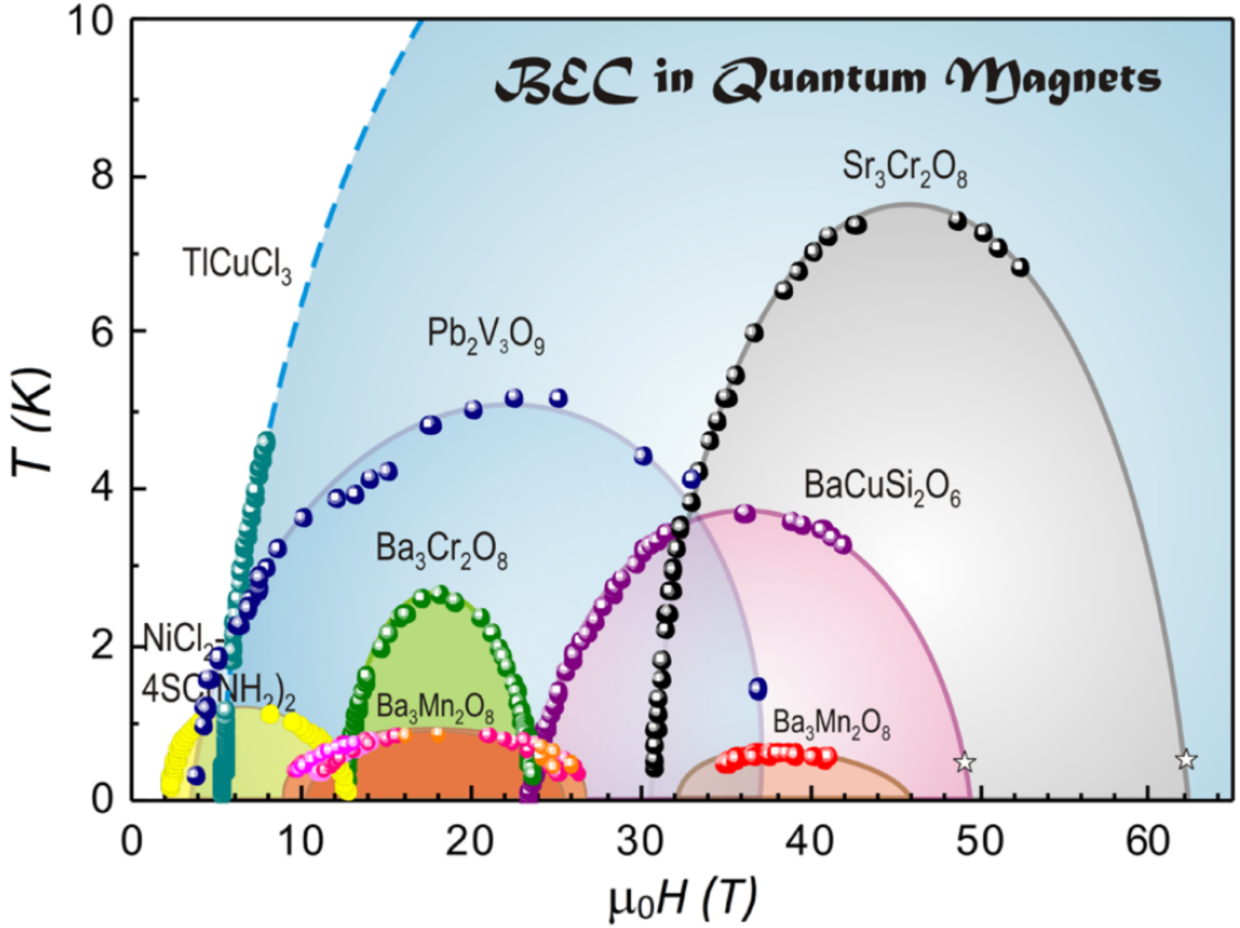



Bose-Hubbard model with distribution in chemical potential => Bose Glass




- T. Giamarchi and H.J. Schulz, Phys. Rev. B **37**, 325 (1988).
- M.P.A. Fisher *et al.*, Phys. Rev. B **40**, 546 (1989).

# BEC in 3D Quantum Magnets




## BEC Magnets

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- *S* > 1/2 dimer
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- *S* = 1, large aniso. D

**Dimensionality ?**

**Disorder ?**

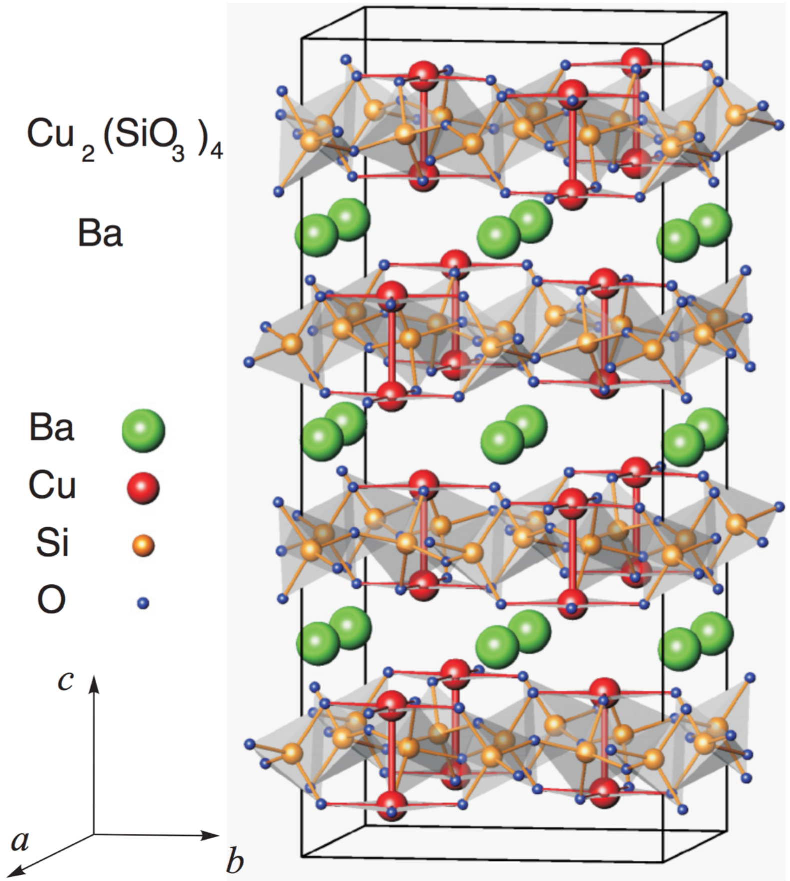
**Frustration ?**

**Out of Equilibrium?**

**Metals ?**

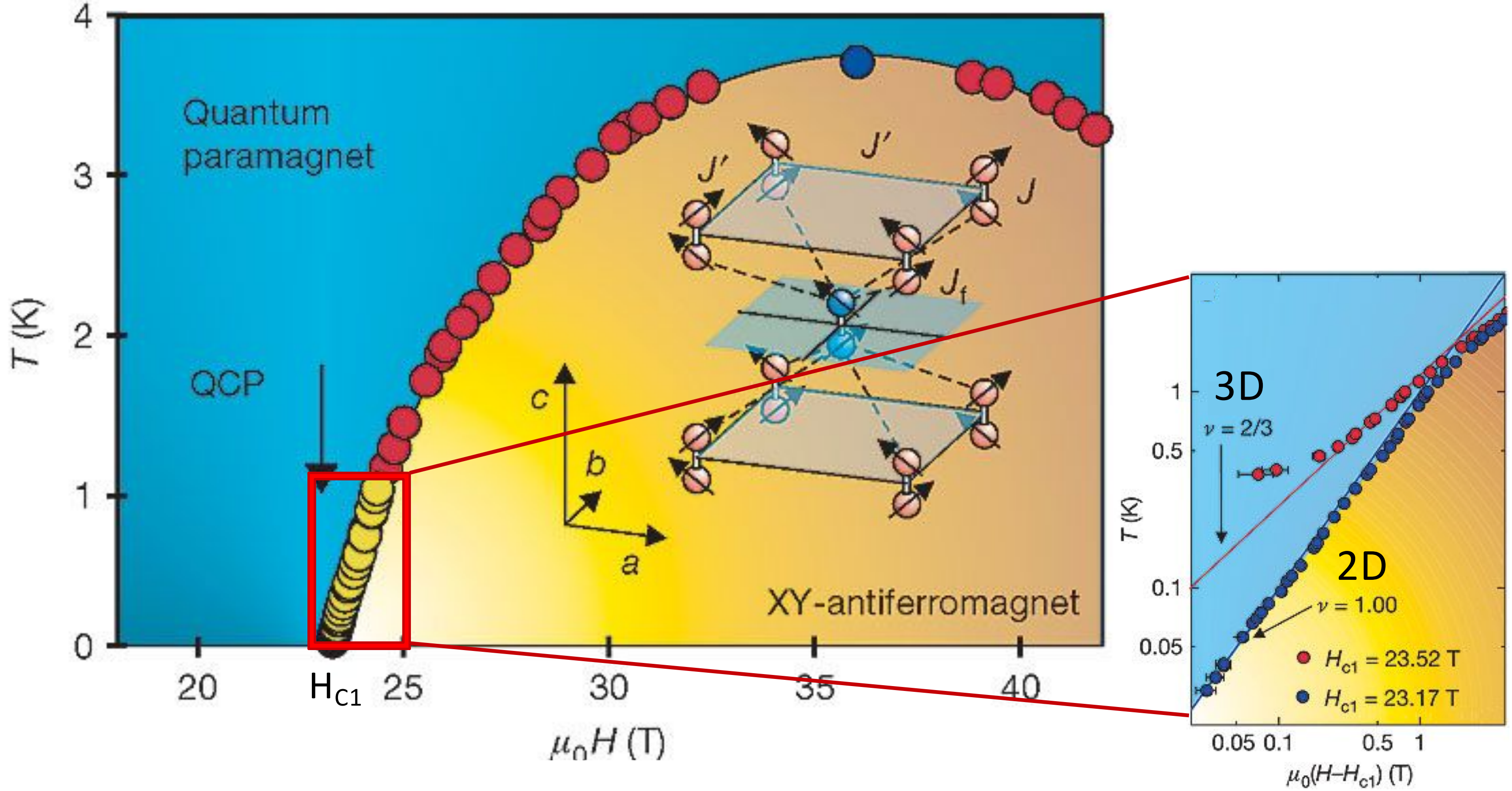
- V. Zapf, M. Jaime, C.D. Batista, Rev. Mod. Phys. **86**, 563 (2014).

# Han Purple - $BaCuSi_2O_6$




- D.V. Sheptyakov *et al.*, PRB **86**, 014433 (2012).

# Dimensional Reduction - $BaCuSi_2O_6$




- S.E. Sebastian *et al.*, Nature **411**, 617 (2006).

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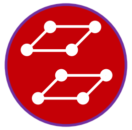
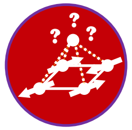
# Dimensional Reduction ?



Bilayers are **fully** decoupled

⇨ **Does not exist in nature!**

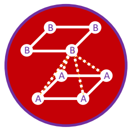



**Frustrated** inter-bilayer exchange

S.E. Sebastian *et al.*, Nature, **411**, 617 (2006).
C.D. Batista *et al.*, PRL **98**, 257201 (2007).
J. Schmalian and C.D. Batista, PRB **77**, 094406 (2008).

⇨ **Unlikely!** M. Maltseva and P. Coleman , PRB **72**, 174415 (2005).
O. Rösch and M. Vojta, PRB **76**, 224408 (2007). ⇨ **effective 3D**

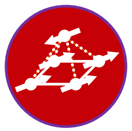

Frustration **and** different bilayers



O. Rösch and M. Vojta, PRB **76**, 224408 (2007).
Ch. Rüegg *et al.*, PRL **98**, 017202 (2007).
S. Krämer *et al.*, PRB **76**, 100406(R) (2007).
N. Laflorencie and F. Mila, PRL **102**, 060602 (2009).
D.V. Sheptyakov *et al.*, PRB **86**, 014433 (2012).
S. Krämer *et al.*, PRB **87**, 180405(R) (2013).

⇨ **Is bilayer-stacking essential?**

## Ab-initio calculations of exchange interactions by DFT:

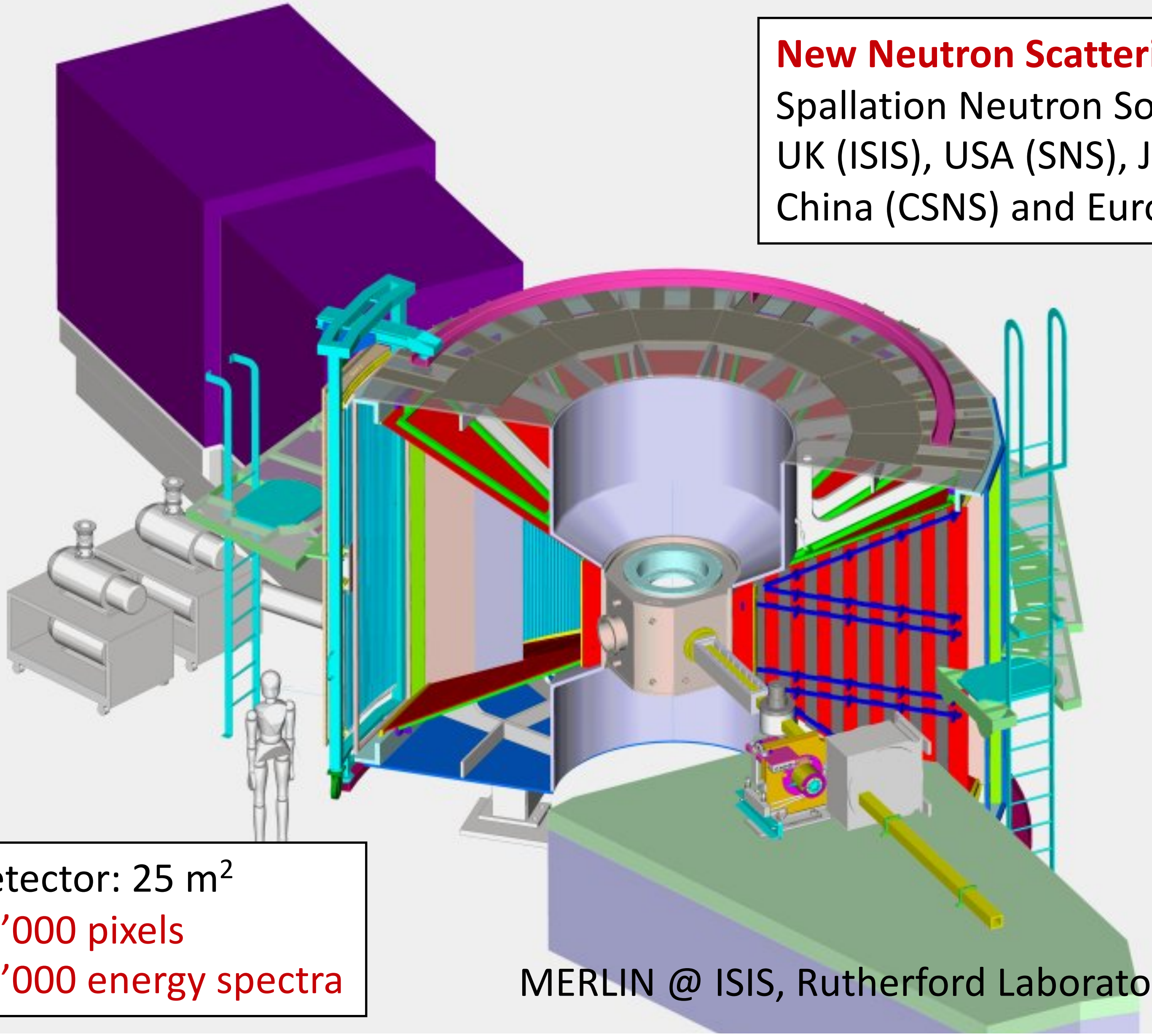


**FM** intra-bilayer exchange ⇨ **No** frustration

V.V. Mazurenko *et al.*, PRL **112**, 107202 (2014).

⇨ **What is the sign of the intra-bilayer exchange?**
⇨ **Why are there 2D exponents?**

# Neutron Time-of-Flight Spectrometers

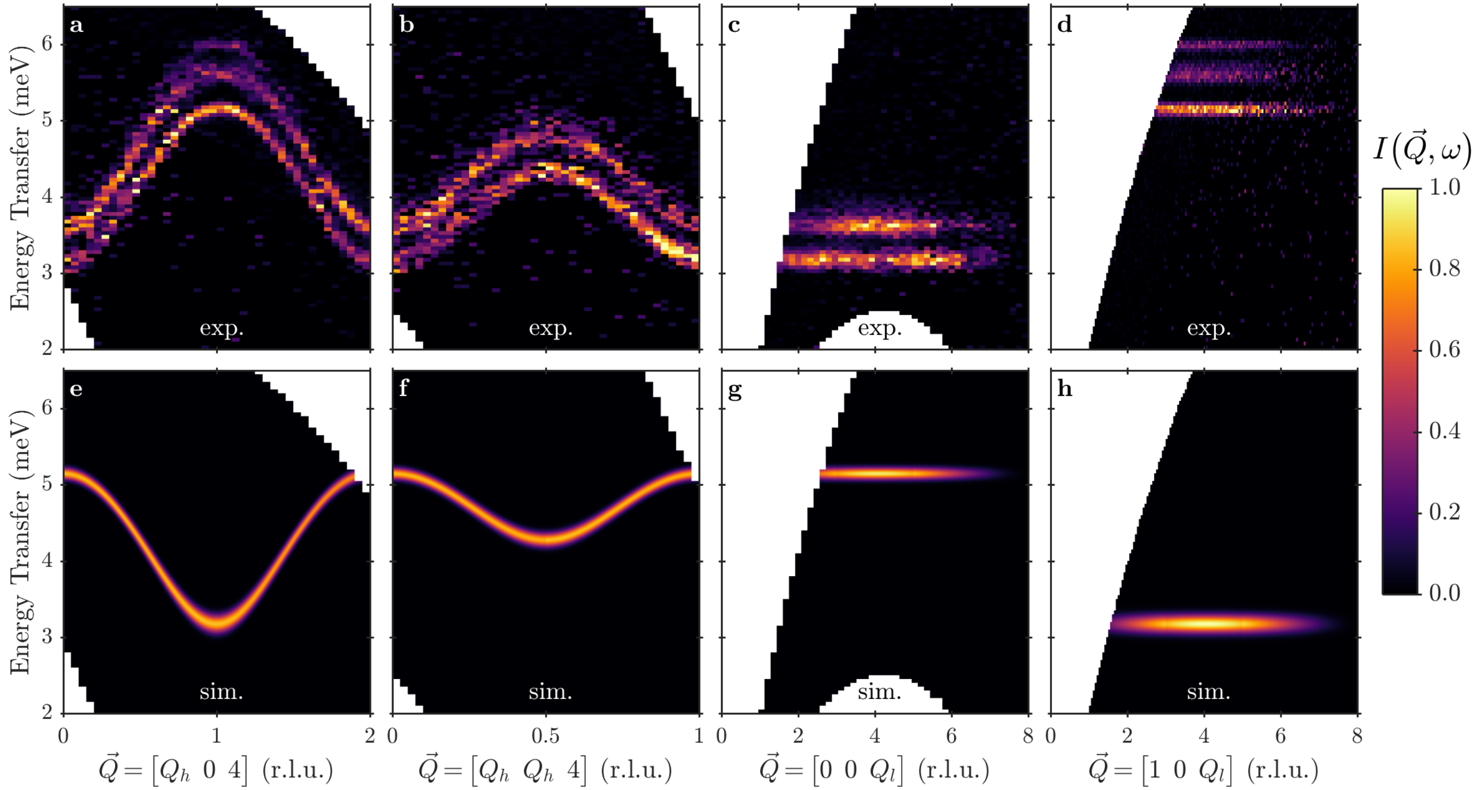


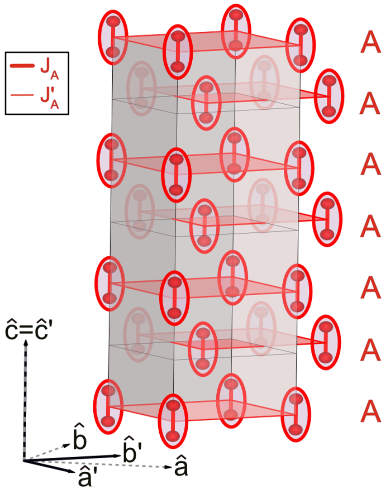
**New Neutron Scattering facilities:**
Spallation Neutron Sources in the UK (ISIS), USA (SNS), Japan (JPARC), China (CSNS) and Europe (ESS)

Detector: 25 $m^2$
36’000 pixels
36’000 energy spectra

MERLIN @ ISIS, Rutherford Laboratory, Didcot, UK

# Experimental vs. Modeled Spectrum

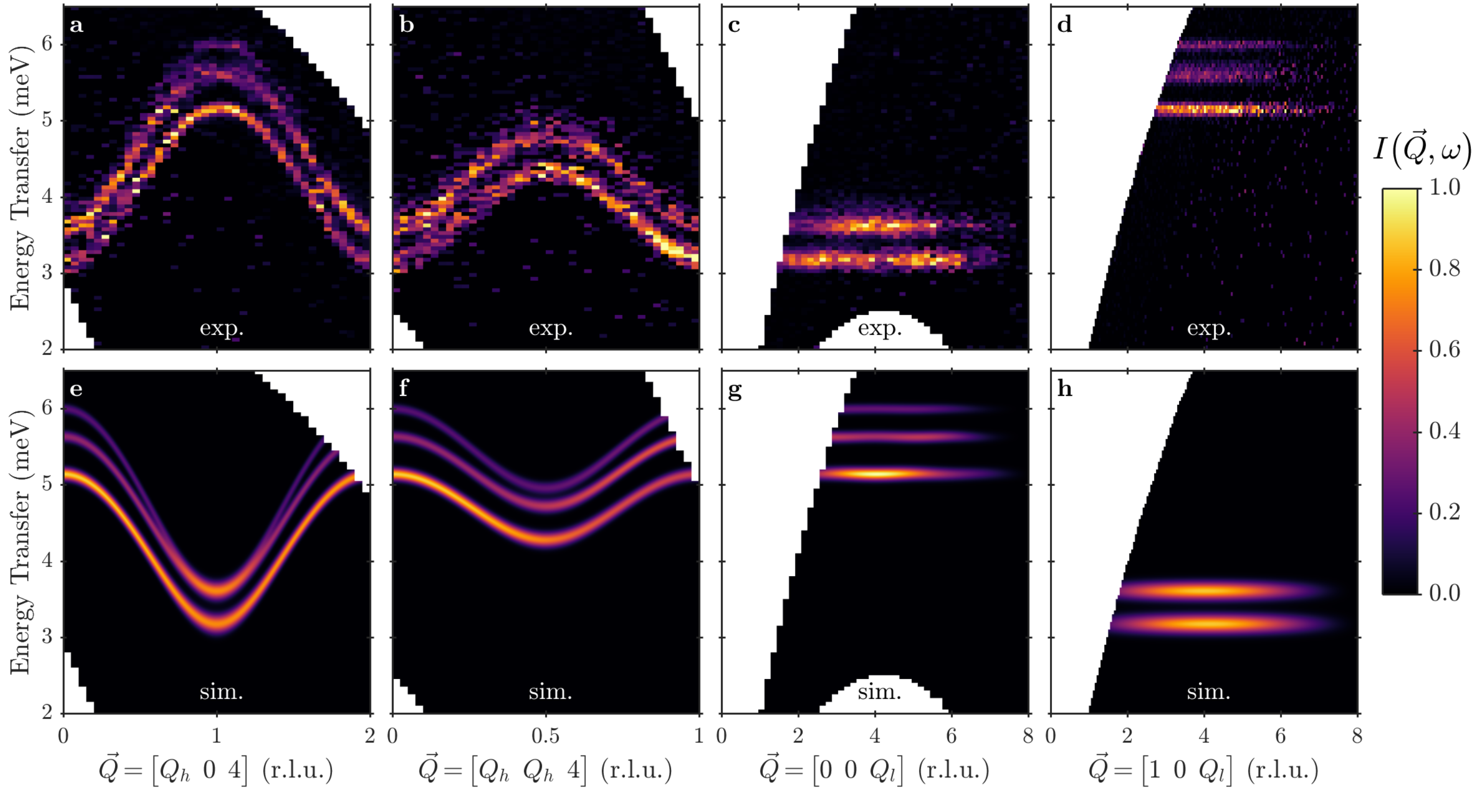


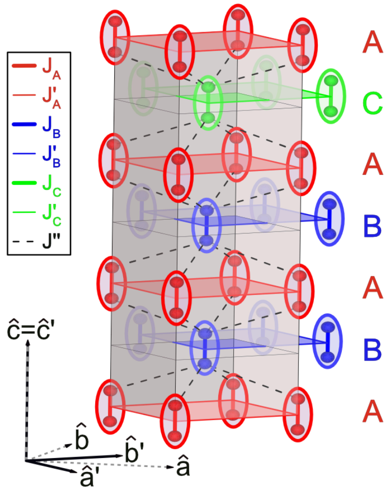




| | Intradimer (J) | Intra-bilayer (J') | Inter-bilayer (J'') |
|---|---|---|---|
| A | AF | AF | - |
| B | - | - | |
| C | - | - | |

- S. Allenspach *et al.*, arXiv:1911.04161

# Experimental vs. Modeled Spectrum

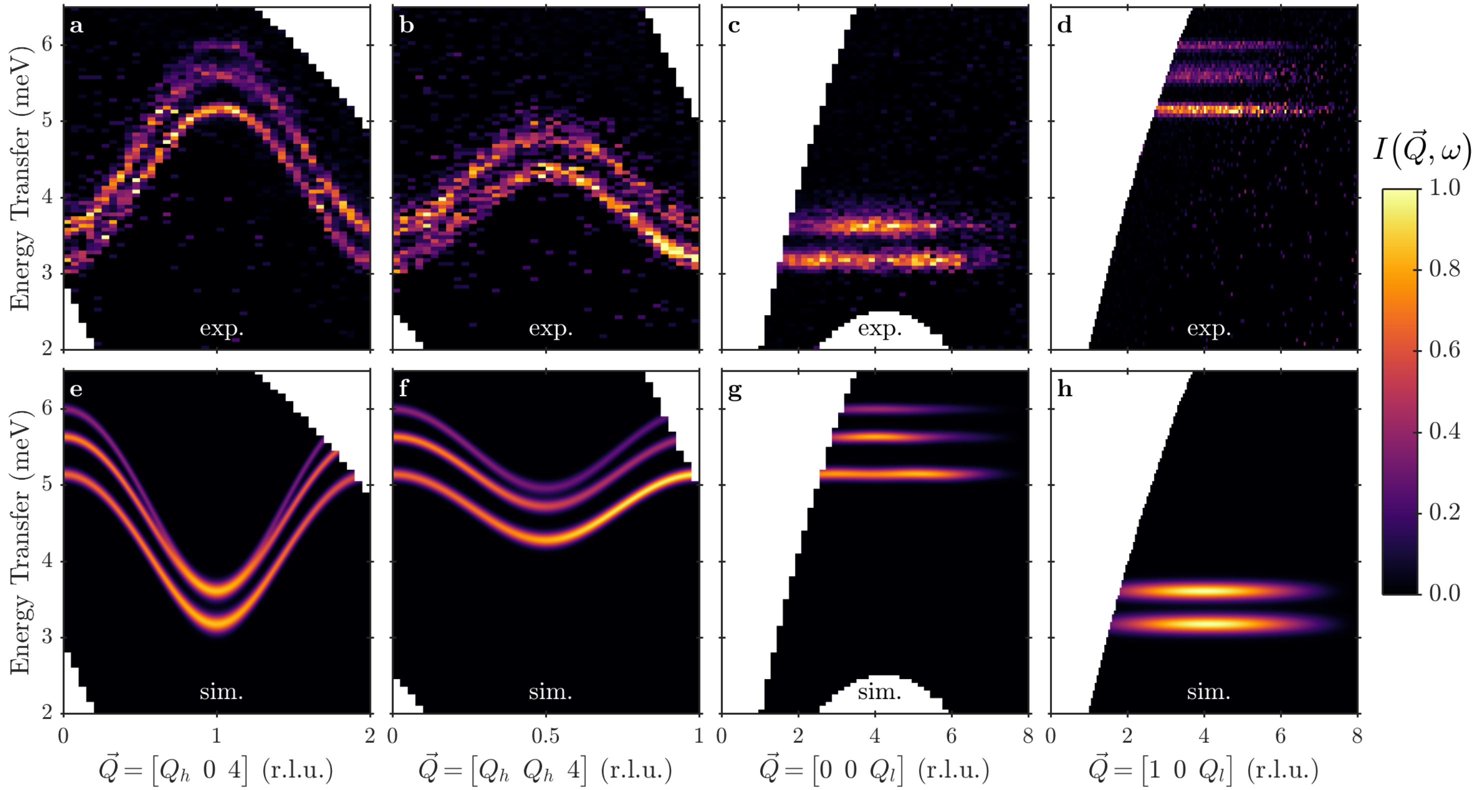


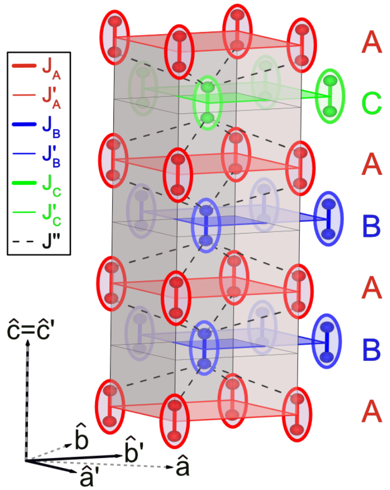




| | Intradimer (J) | Intra-bilayer (J') | Inter-bilayer (J'') |
|---|---|---|---|
| A | AF | AF | AF |
| B | AF | AF | |
| C | AF | AF | |

- S. Allenspach *et al.*, arXiv:1911.04161

# Experimental vs. Modeled Spectrum

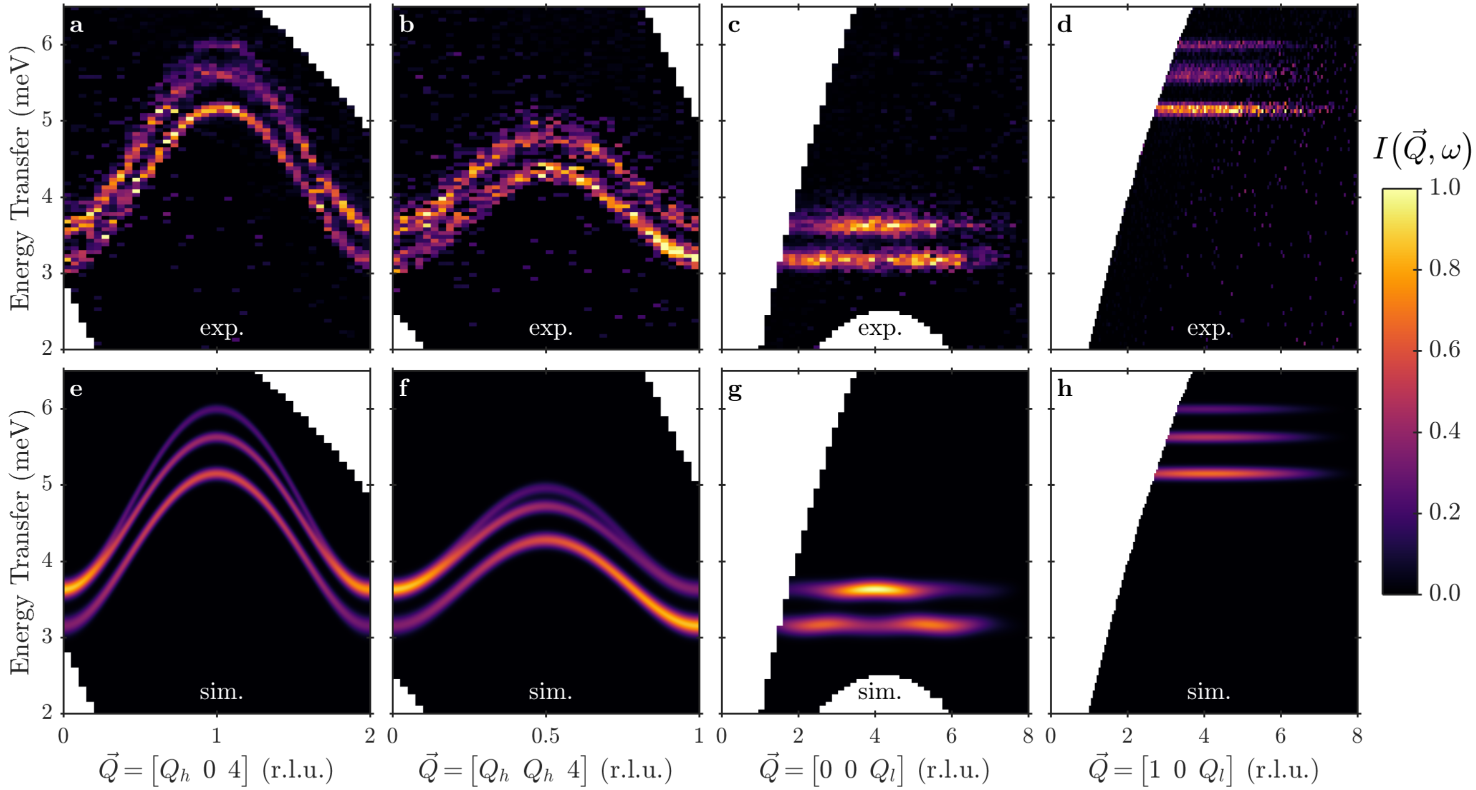


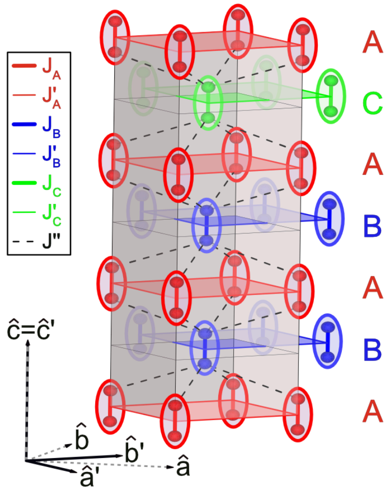




| | Intradimer (J) | Intra-bilayer (J') | Inter-bilayer (J'') |
|---|---|---|---|
| A | AF | AF | |
| B | AF | AF | FM |
| C | AF | AF | |

- S. Allenspach *et al.*, arXiv:1911.04161

# Experimental vs. Modeled Spectrum

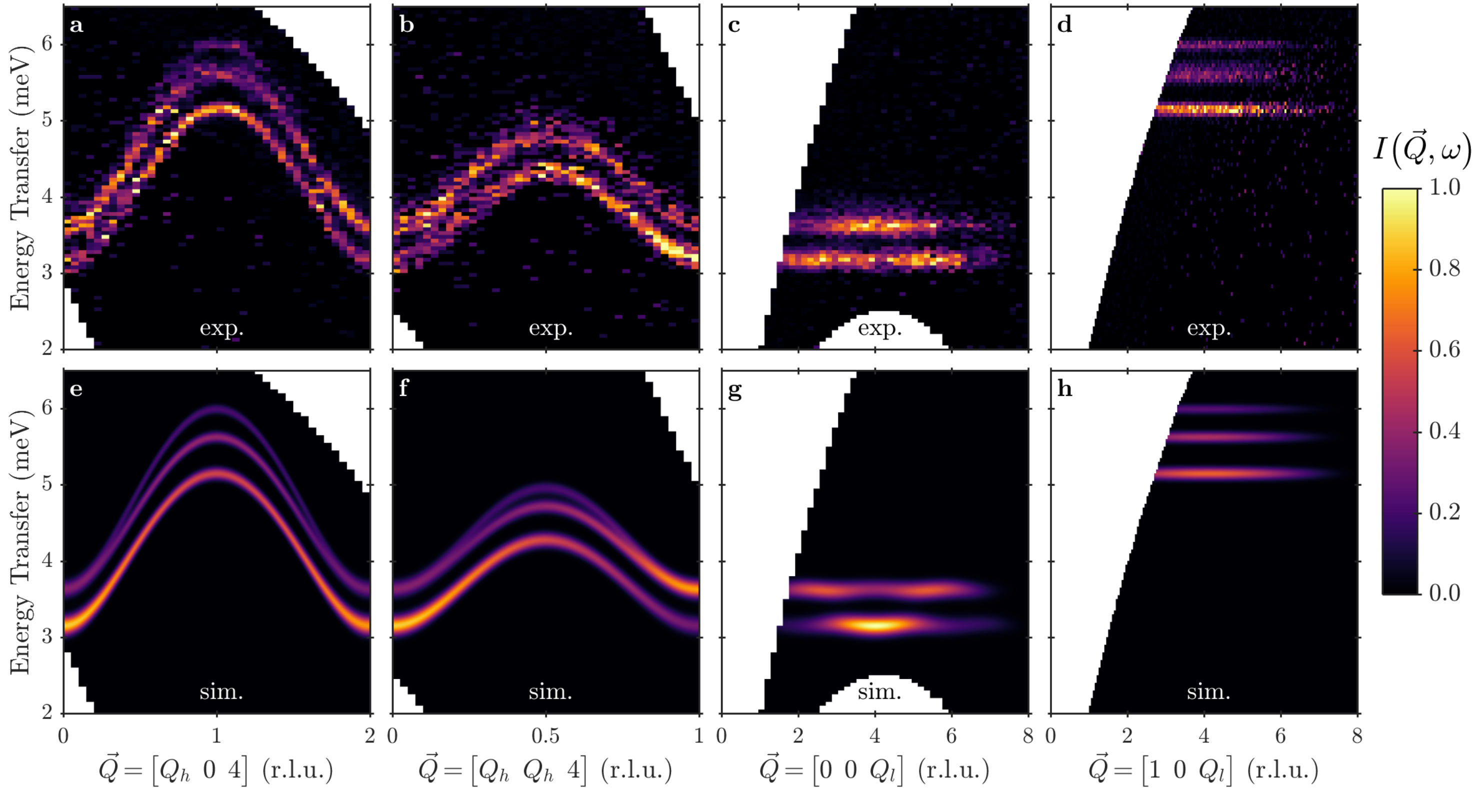


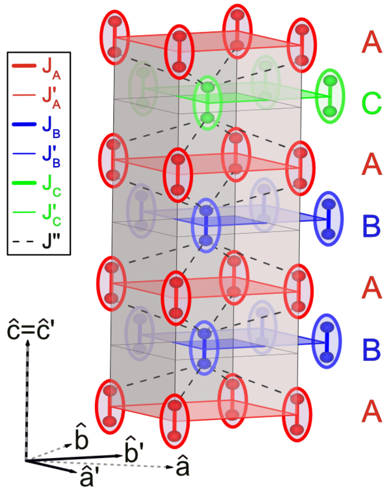




| | Intradimer (J) | Intra-bilayer (J') | Inter-bilayer (J'') |
|---|---|---|---|
| A | AF | FM | FM |
| B | AF | FM | |
| C | AF | FM | |

- S. Allenspach *et al.*, arXiv:1911.04161

# Experimental vs. Modeled Spectrum

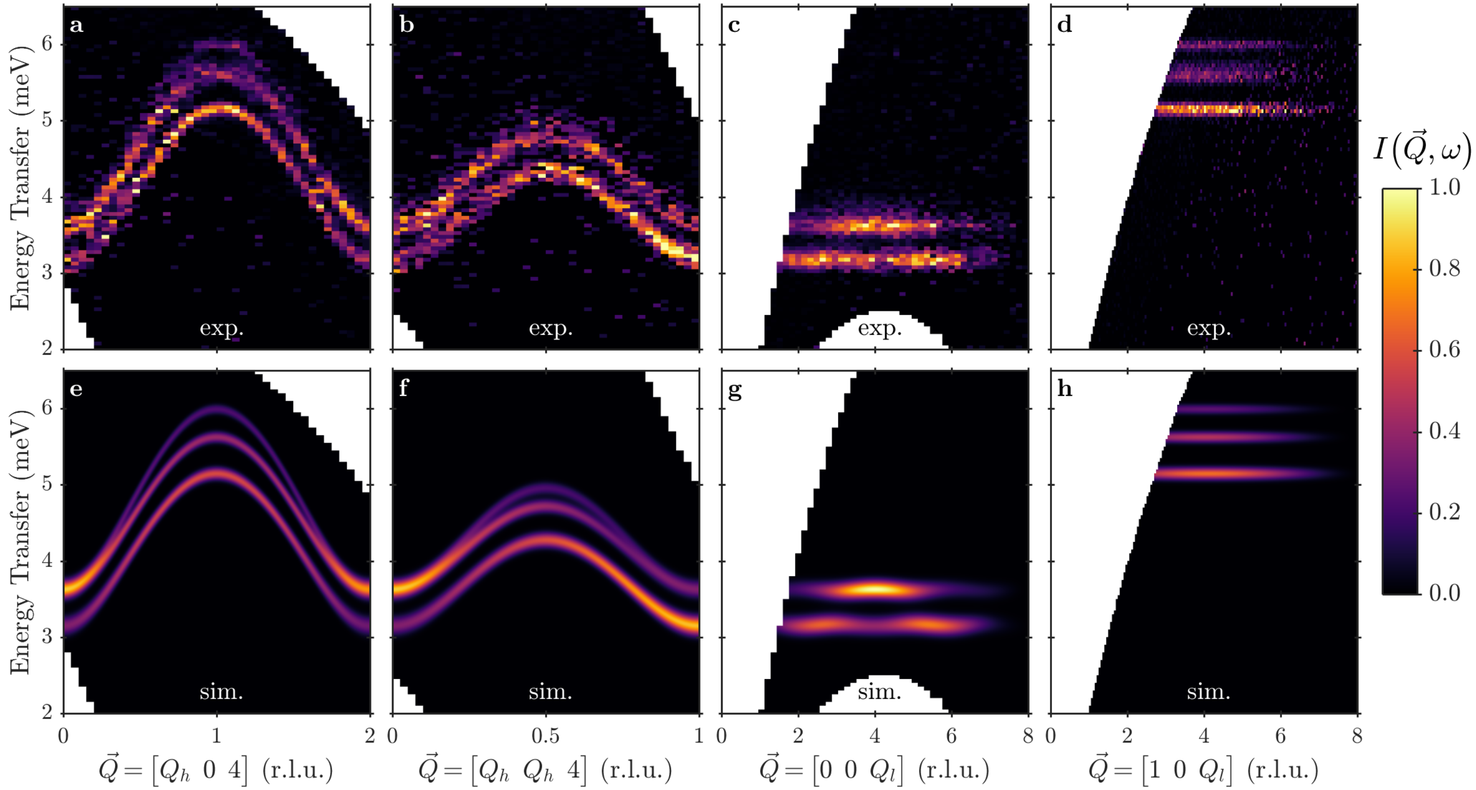


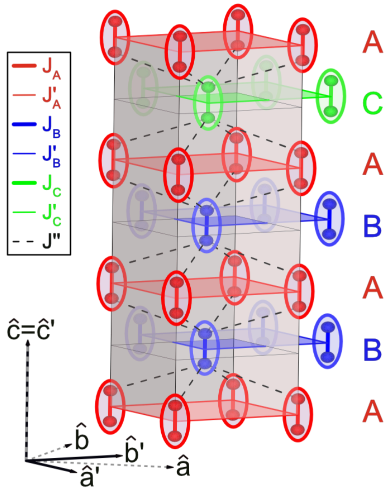




| | Intradimer (J) | Intra-bilayer (J’) | Inter-bilayer (J’’) |
|---|---|---|---|
| A | AF | FM | |
| B | AF | FM | AF |
| C | AF | FM | |

• S. Allenspach *et al.*, arXiv:1911.04161

# Experimental vs. Modeled Spectrum



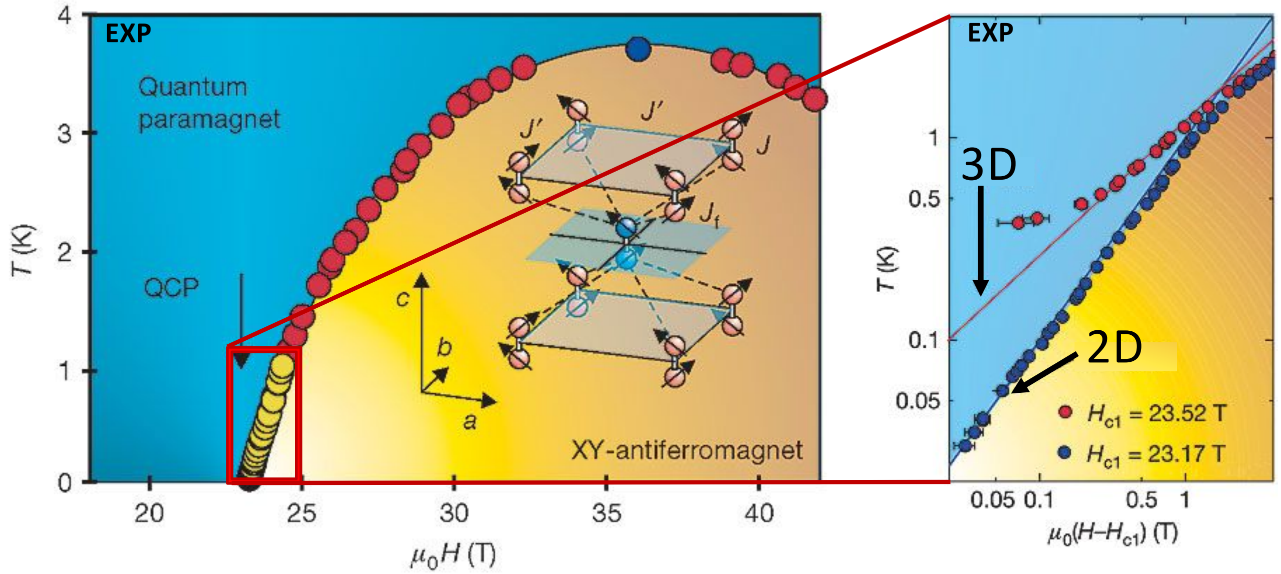




| | Intradimer (J) | Intra-bilayer (J') | Inter-bilayer (J'') | |
|---|---|---|---|---|
| A | 4.275(5) | -0.480(3) | | |
| B | 4.72(1) | -0.497(8) | -0.04(1) | meV |
| C | 4.95(2) | -0.57(1) | | |

• S. Allenspach *et al.*, arXiv:1911.04161

# Quantum Monte Carlo Phase Diagram

Experimental Phase Diagram




• S.E. Sebastian *et al.*, Nature, **411**, 617 (2006).

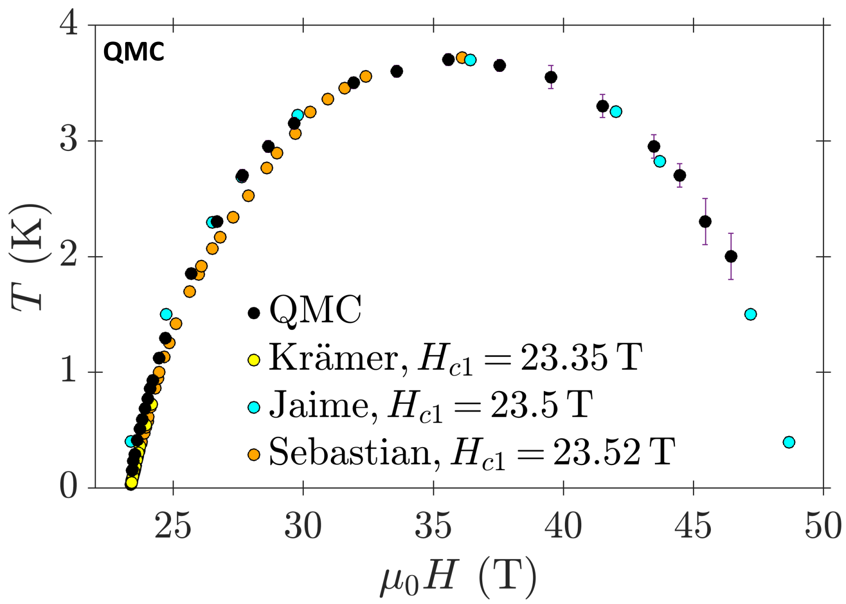
Quantum MC Phase Diagram

⇨ FM $J_{\text{intra-bilayer}}$

⇨ FM $J_{\text{inter-bilayer}}$

⇨ 3 dimer types with stacking

**N.Laflorencie** and **F.Mila**




⇨ Phase boundary ($H_c$,$T_c$) reproduced

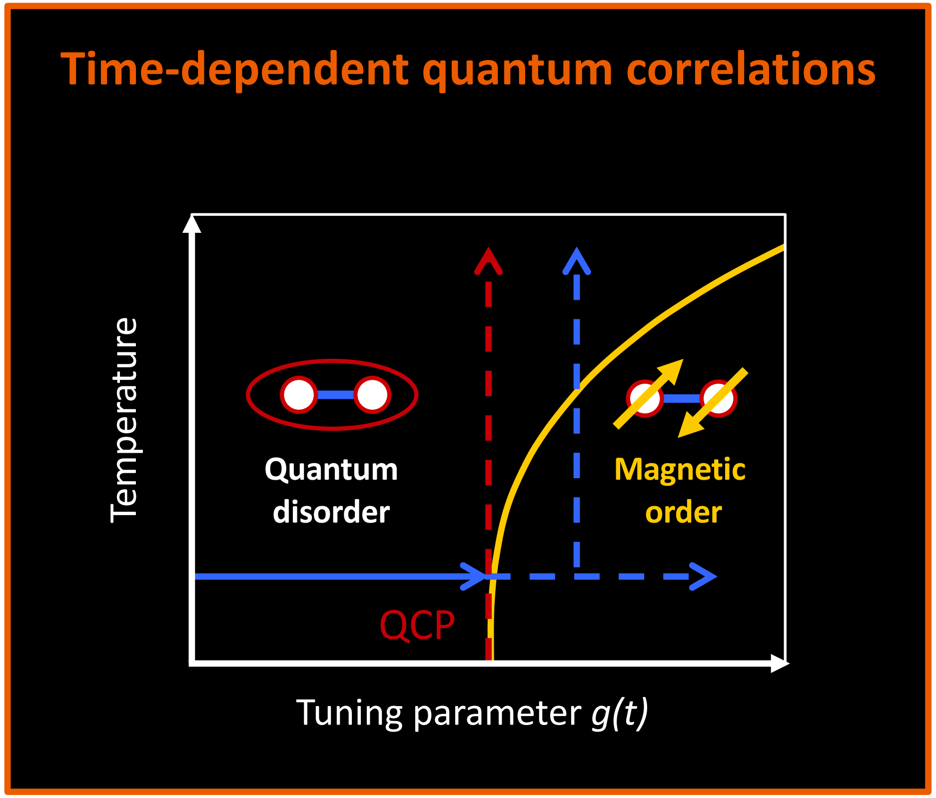
⇨ Bilayer populations can be simulated (NMR)

⇨ Ongoing QMC simulation in the critical region

• S. Allenspach *et al.*, arXiv:1911.04161

# Swiss Free Electron Laser - SwissFEL

# ERC - Hyper Quantum Criticality




## Experiments at SwissFEL and LCLS:

**1) Quantum spin chain**

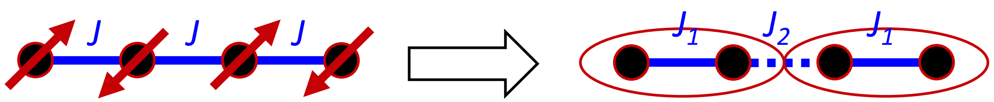
- Coupling to optical phonon
- Evolution of ground state (gapped/gapless) and of 3D order

**2) Quantum dimer materials**

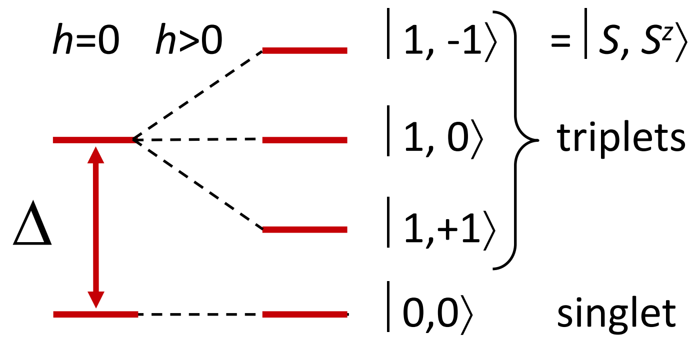
- Quantum quench in pulsed field
- THz pumping of phonons and magnetic quasi-particles
- Relaxation, time-scales

## Dynamic Control of Quantum Correlations:

- Phonon modes coupling to exchange interactions




- Direct pumping of quasi-particles
- Pulsed magnetic fields



# Collaborators

- **Paul Scherrer Institute**
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  <u>P. Puphal</u>
  <u>S. Ward</u>

- **Samples**
  K. Krämer (Bern)
  I. Fisher (Stanford)
- **Theory**
  T. Giamarchi (Geneva)
  C. Kollath (Bonn)
  F. Mila (EPFL)
  N. Laflorencie (CNRS)
  B. Normand (PSI)
  A. Läuchli (Innsb.)
  K. Schmidt (Nürnberg)
- **Experiments:** SINQ, ILL, ISIS, J-Parc

FNSNF Swiss National Science Foundation
erc
EPSRC Engineering and Physical Sciences Research Council
European Commission

## New Phases and Dynamics in Low-dimensional Quantum Magnets

New neutron spectrometers allow us to map the full static and dynamic correlation functions of quantum magnets.

Magnon BEC now observed in q-1D, q-2D and 3D lattices with exiting physics: spin-Luttinger liquid, "dimensional reduction", effects of disorder.

Free-electron lasers offer new opportunities for out-of-equilibrium studies (of systems with well-known Hamiltonians).