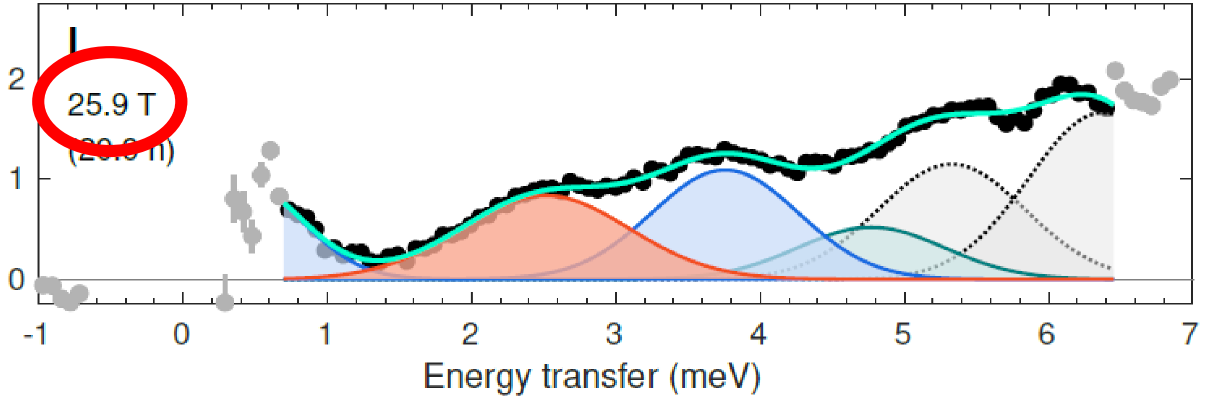
PAUL SCHERRER INSTITUT

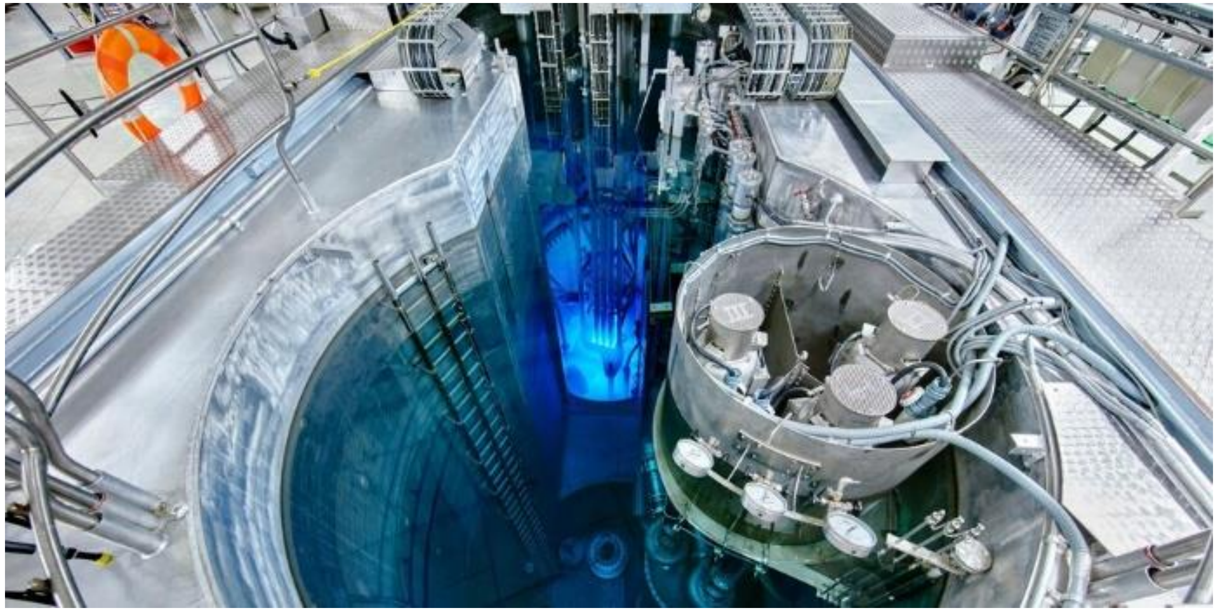
Quantum Magnetism at High Magnetic Fields

Bruce Normand, PSI and EPFL

Institute of Physics, CAS,
Beijing, 26.09.23



High Magnetic Fields and Neutron Scattering



Inelastic neutron scattering requires large* facilities.
The upper magnetic field limit of a standard cryostat is 16 T.

Generating higher magnetic fields also requires large facilities …**

At the **Helmholtz Zentrum Berlin**, the decision was made to build a high-field facility next to the nuclear reactor, at which the **HFM/EXED spectrometer** operated in diffraction, spectroscopy and low-*Q* modes at fields up to 25.9 T.

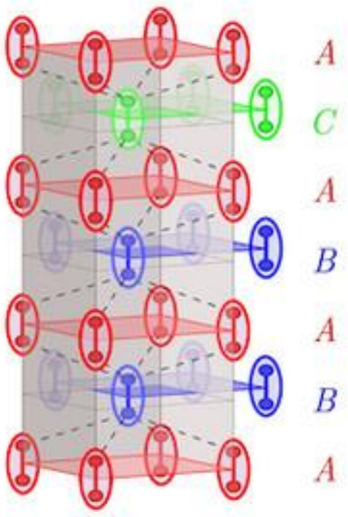
However, then domestic political considerations related to the policies of the Green Party led to the reactor being closed, limiting the period of operation to 2015-2019.

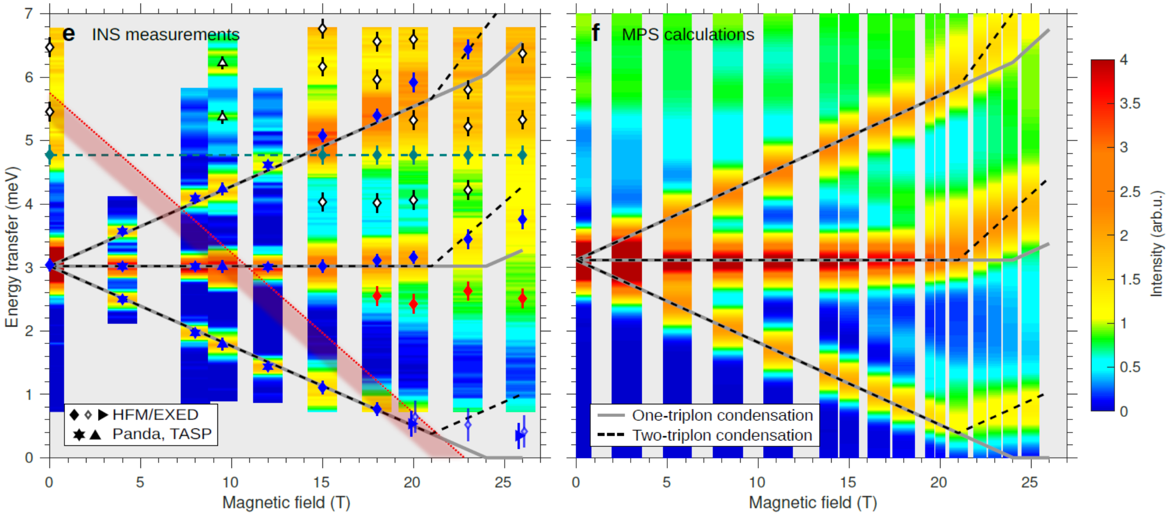
*price tag into 10 figures …

**price tag into 9 figures …

Roadmap

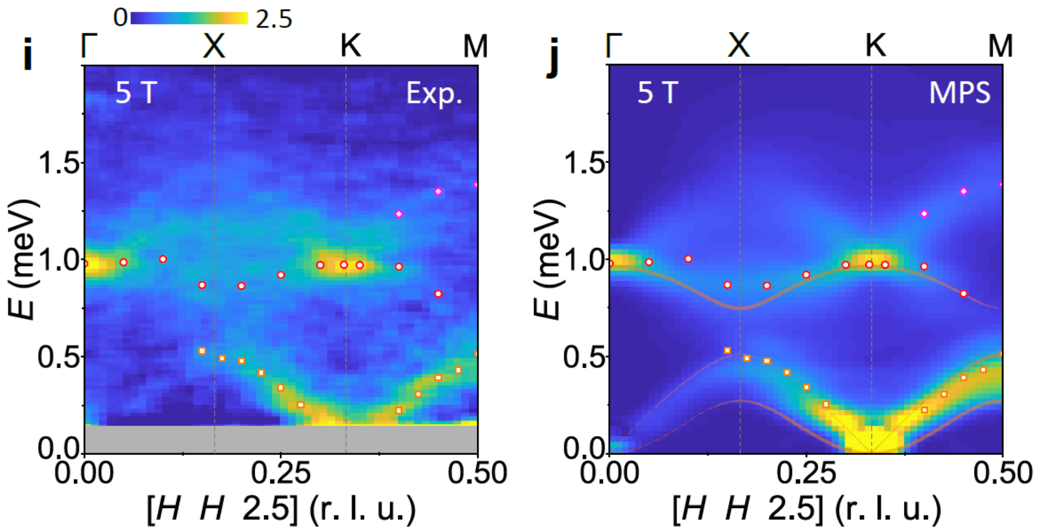
1) Novel quantum phases:
field-induced spin nematic





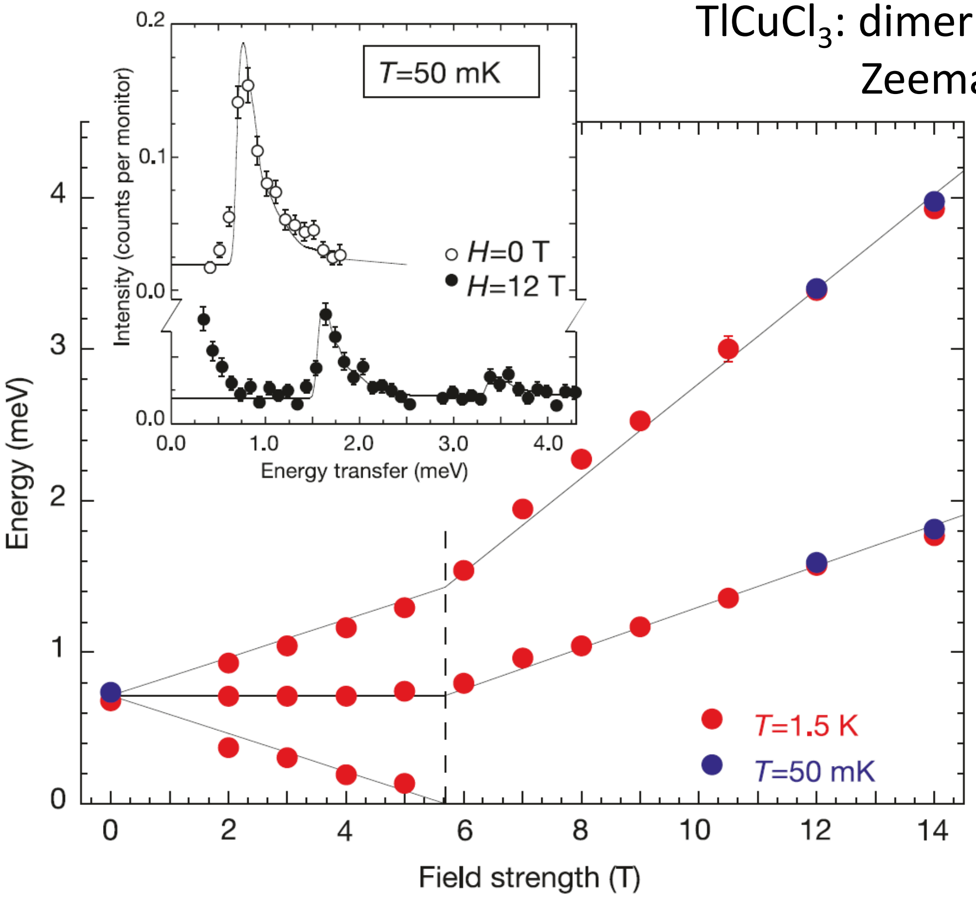
2) Novel quantum phase transitions:
quasi-2D Bose-Einstein universality

3) Novel excitation spectra:
field-controlled continua

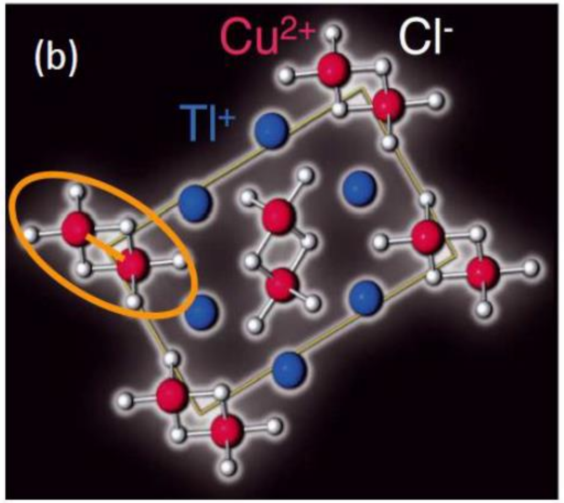


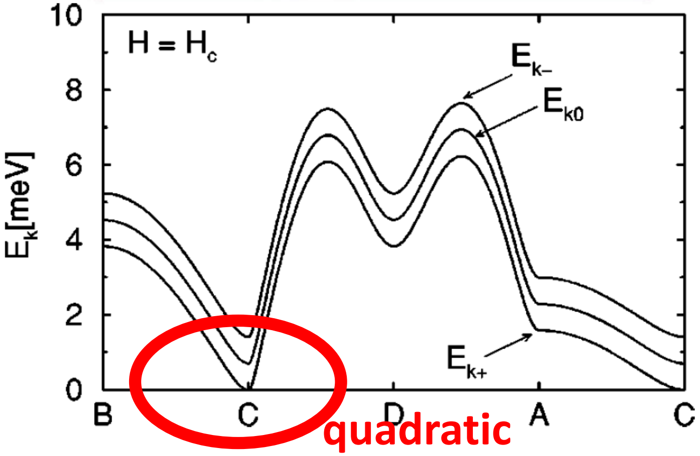
"Bose-Einstein Condensation of Magnons"

Heisenberg model $\hat{\mathcal{H}} = \sum_{[i,j]_m} J_m \boldsymbol{S}_i \cdot \boldsymbol{S}_j$ in an applied magnetic field $\hat{\mathcal{H}}_m = -g\mu_B \sum_i \boldsymbol{B} \cdot \boldsymbol{S}_i$

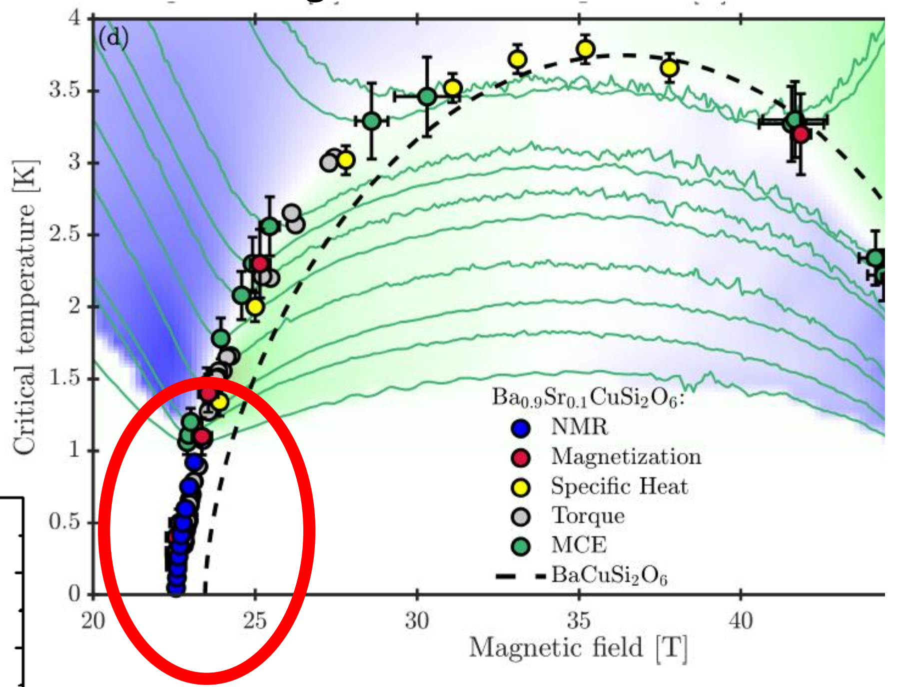


$TlCuCl_3$: dimerised quantum spin system,
Zeeman-split triplet excitation





Quantum phase transition: spontaneous breaking of U(1) symmetry, field-induced transverse magnetic order.



$$T_c(H) = \alpha(H - H_{c1})^{\phi}$$

$$\phi = z/d \qquad \omega \propto k^z$$

ϕ = 2/3 for free bosons in 3D:
Bose-Einstein universality

[1] Ch. Rüegg *et al.*, Nature **423**, 62 (2003).

[2] M. Matsumoto, B. Normand, T. M. Rice and M. Sigrist, Phys. Rev. B **69**, 054423 (2004).

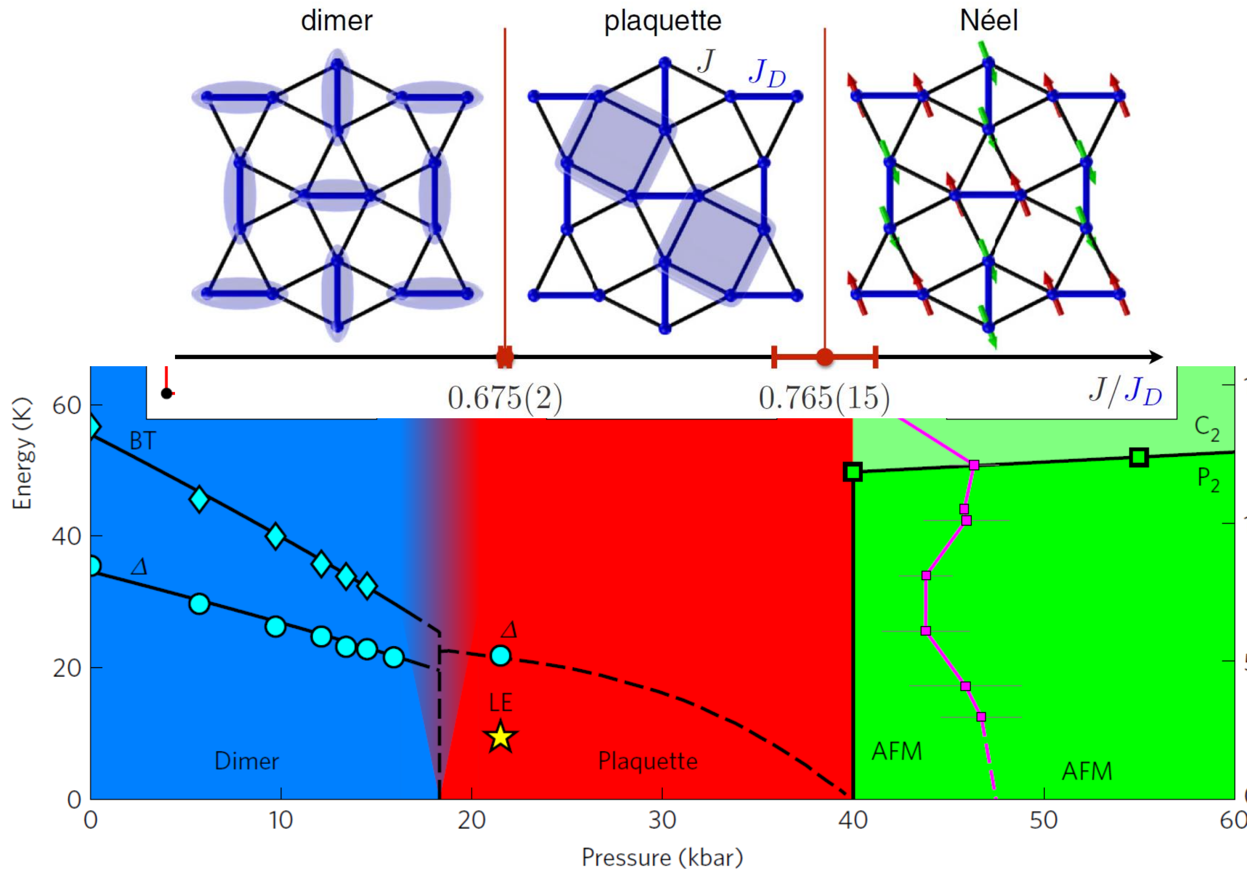
$SrCu_2(BO_3)_2$: the ultimate 2D frustrated quantum magnet

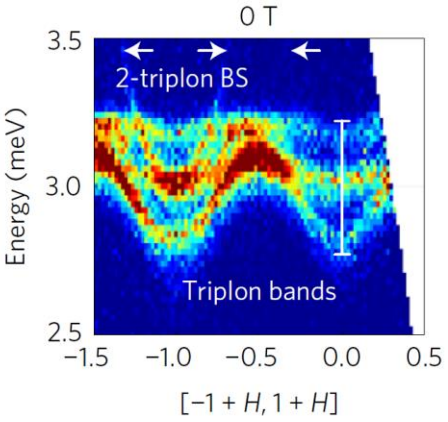
***S* = ½, ideal frustration: Shastry-Sutherland model**

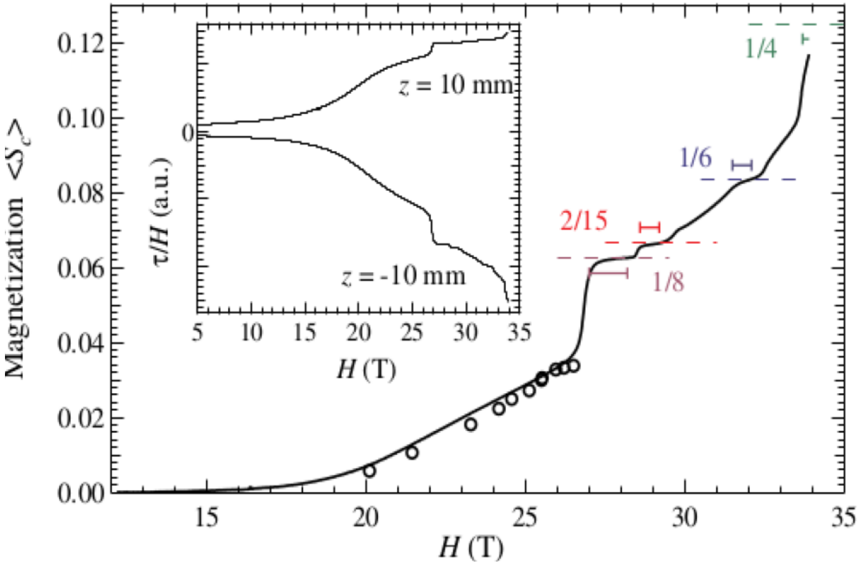
Exact dimer ground state **Strongly bound states**

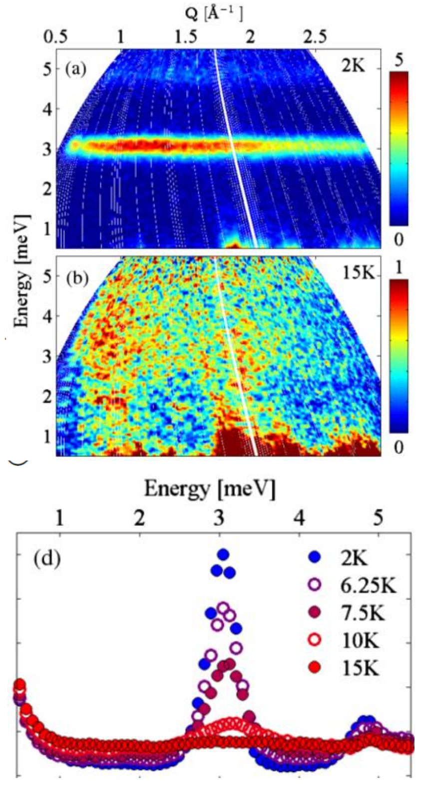
Magnetisation plateaux **Anomalous thermodynamics**

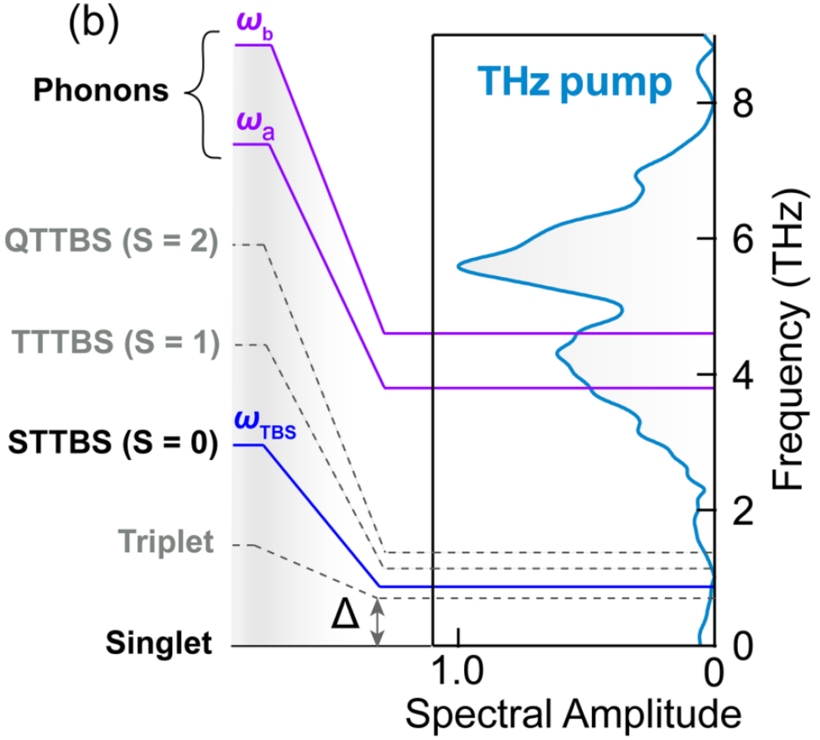
Pressure-induced QPTs (2, both first-order)







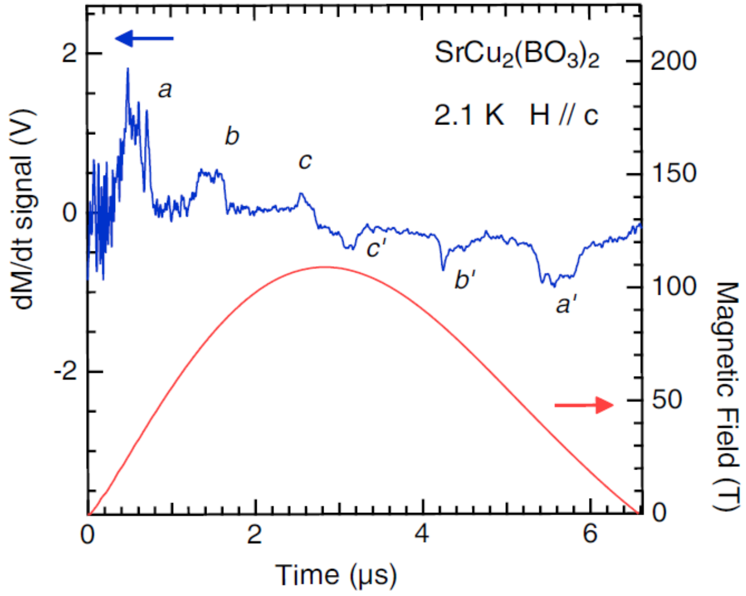


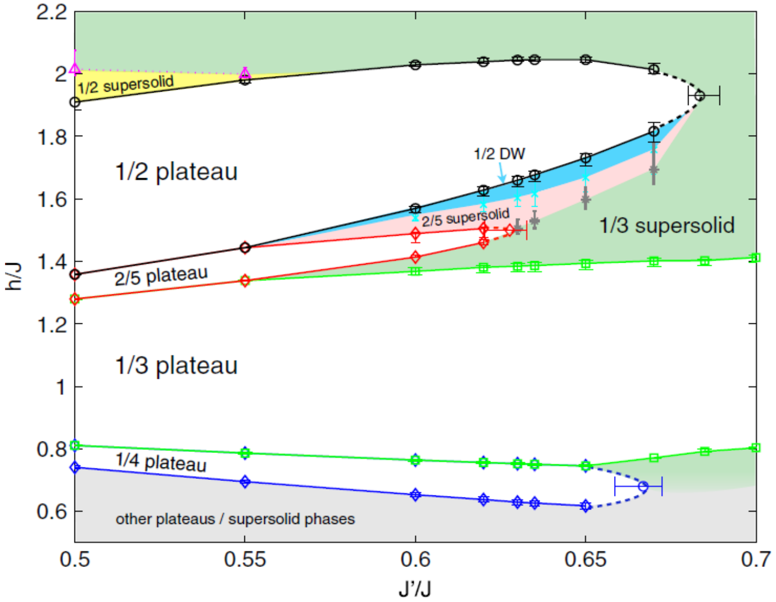


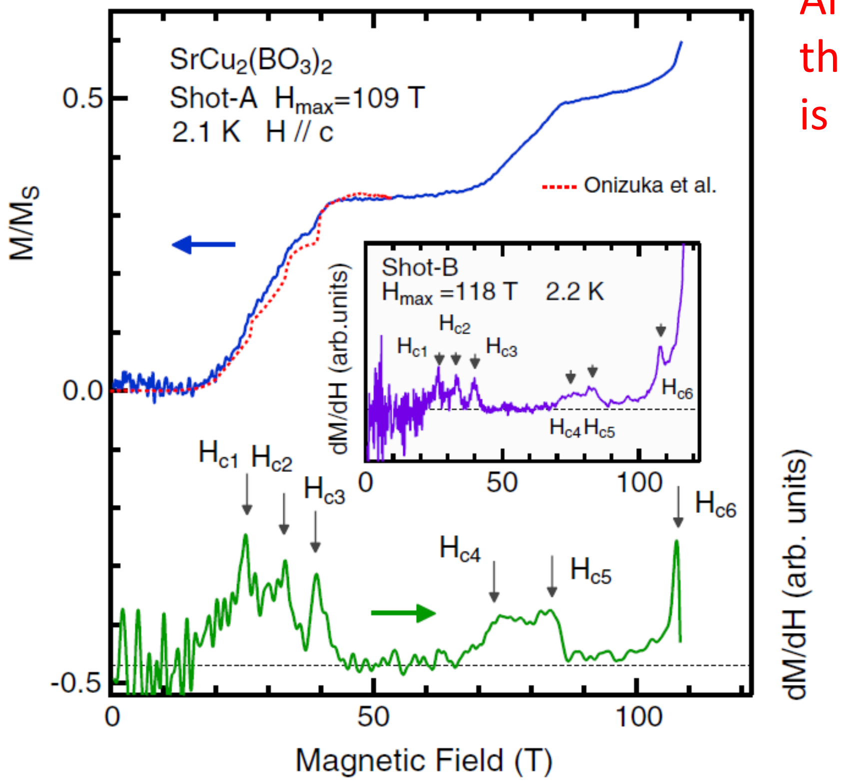
Y. H. Matsuda *et al.*, PRL **111**, 137204 (2011).
H. Nojiri *et al.*, JPSJ **72**, 3242 (2003).

Magnetisation in SCBO

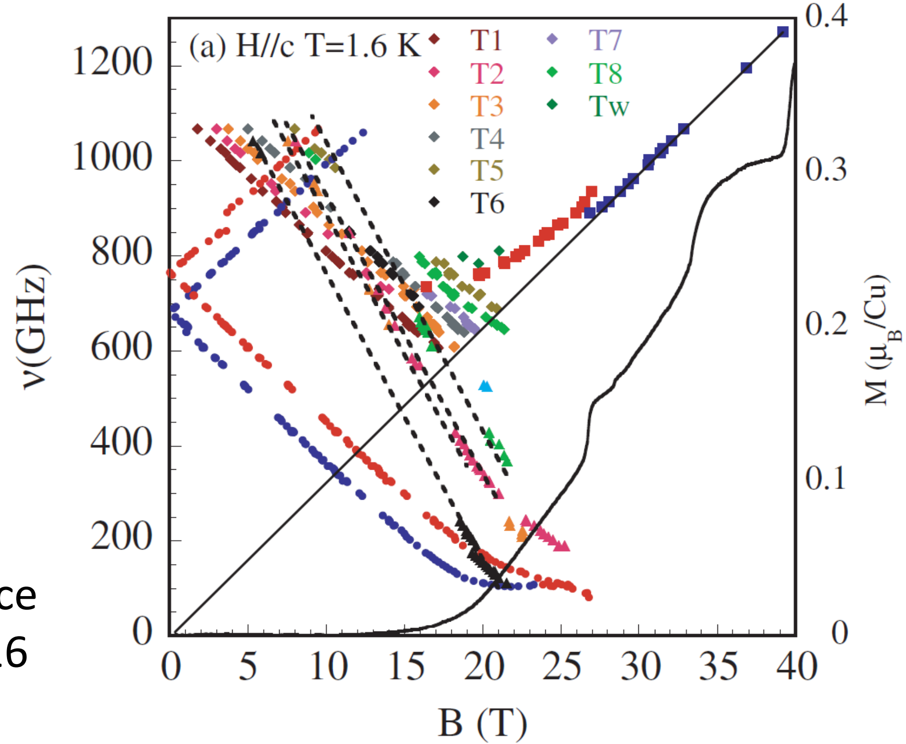
One observes a spectacular series of fractional magnetisation plateaux (1/8, 2/15, 1/6, 1/4, 2/5, 1/3, 1/2) up to saturation at 140 T. While far from fully understood, multiple supersolid phases form due to combined singlet formation and lattice distortion.







An open question since the outset has been the closure of the first gap to an excited state: is it the triplet or the S_z = 2 quintet branch ?

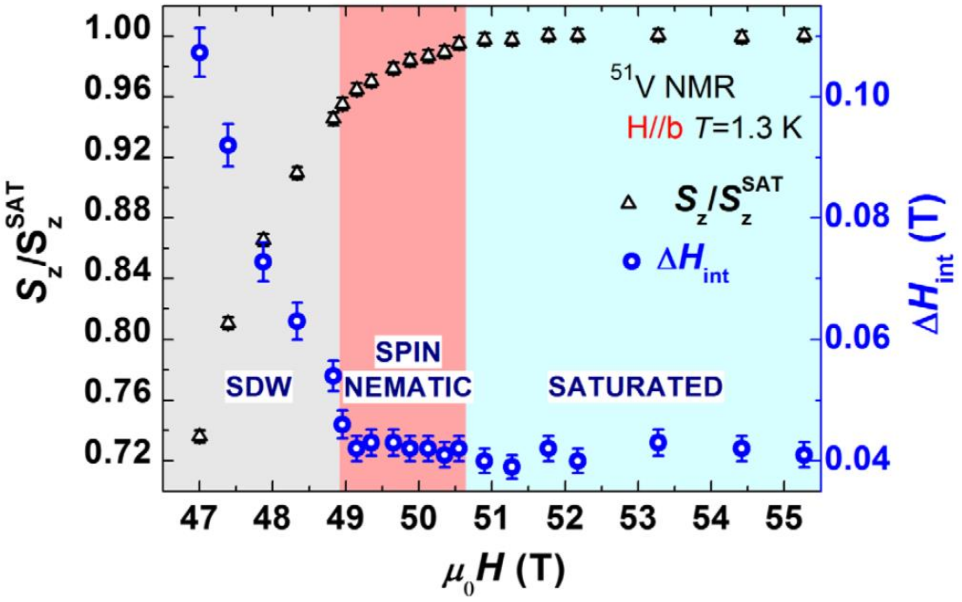


The closure of the zero-field gaps takes place precisely in the INS-dark regime between 16 and 26 T.

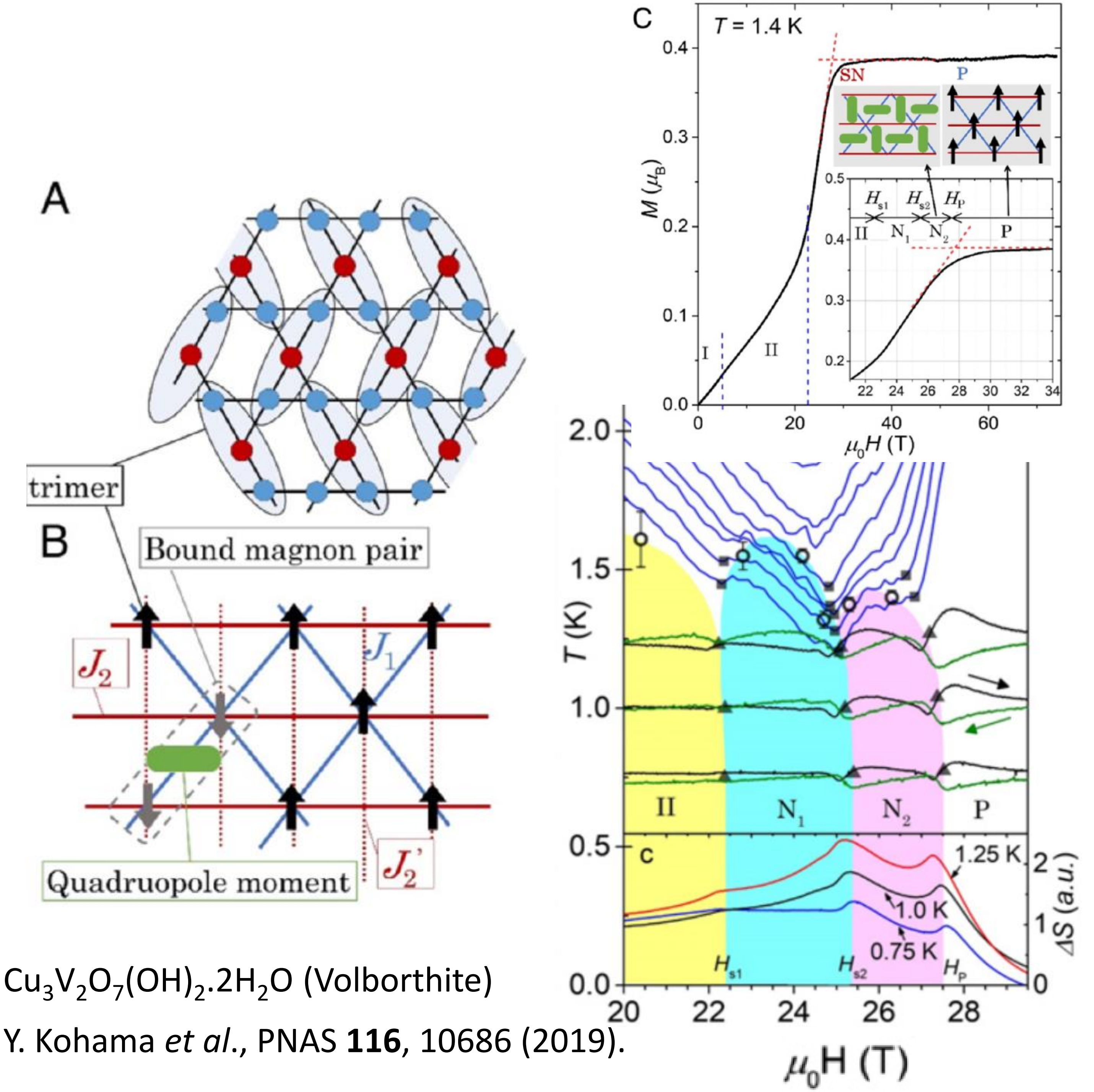
Spin-nematic state

What happens if the S = 2 state touches down first ?
This is called a "spin nematic," a state whose order parameter is not the regular magnetic (dipolar) order but the **spin quadrupole moment**.

This order parameter is difficult to measure by conventional probe techniques, particularly those available at high fields (where the spin nematic is expected under most scenarios).



LiCuVO$_4$: A. Orlova *et al.*, PRL **118**, 247201 (2017).



$Cu_3V_2O_7(OH)_2.2H_2O$ (Volborthite)

Y. Kohama *et al.*, PNAS **116**, 10686 (2019).

Spin-nematic states in SCBO ?

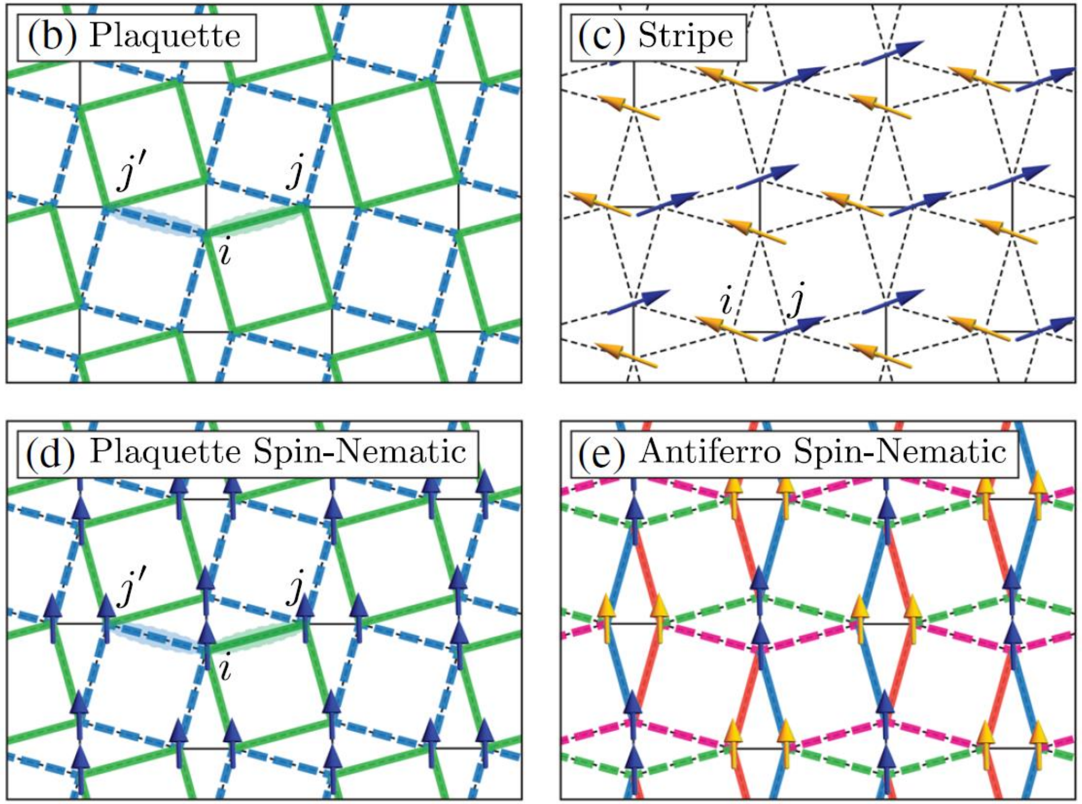
Why is this a difficult question to answer ?

(1) Standard momentum-resolved experimental probes do not observe S = 2 excitations, while light-scattering methods probe them only at momentum Q = (0,0) and for specific point-group symmetries (excluding both the E and A_2 states).

(2) The Dzyaloshinskii-Moriya coupling mixes S = 1 and S = 2 states, confusing the issue.

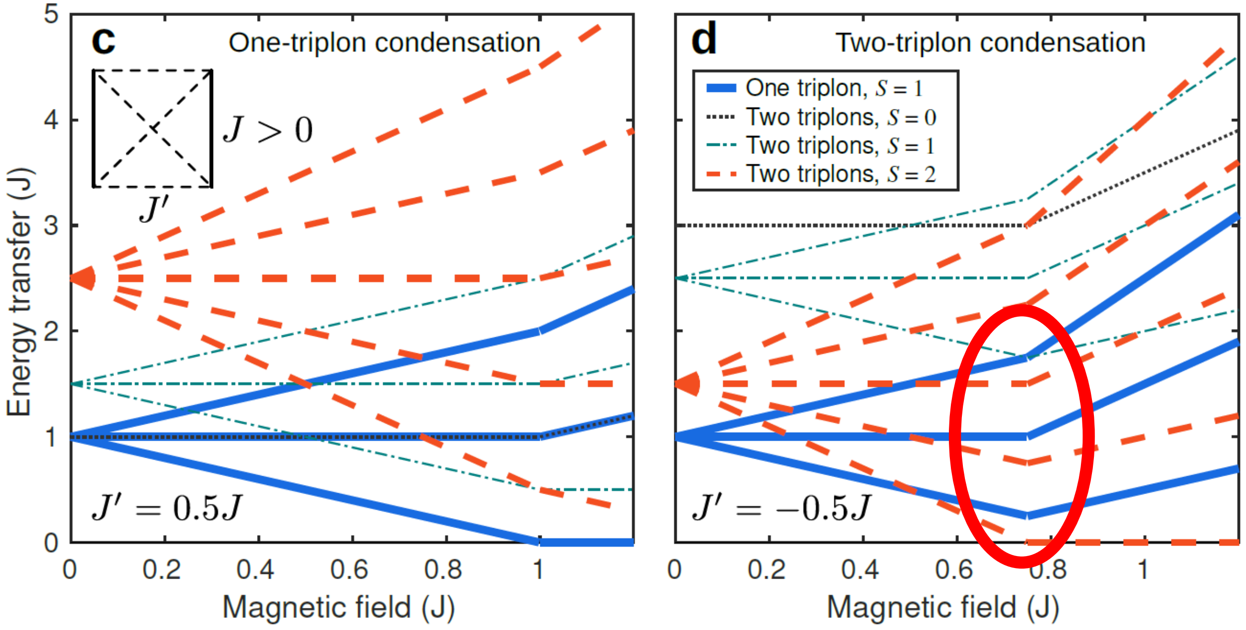
(3) The |2,2> states form 4 bands [1], offering different possible spin-nematic states depending on which lies lowest.

A_2



E

[1] Z. Wang and C. D. Batista, PRL **120**, 247201 (2020).



We will answer the question not by trying to probe the ground state but by examining the spectrum of excited states: both the field evolution and the type and number of states will differ significantly as a function of the nature of the ground state.

INS up to 25.9 T

In essence there is no $\boldsymbol{Q}$-dependence at all, so it is valid to integrate over all $\boldsymbol{Q}$. This results in some broadening of the observed energy levels.

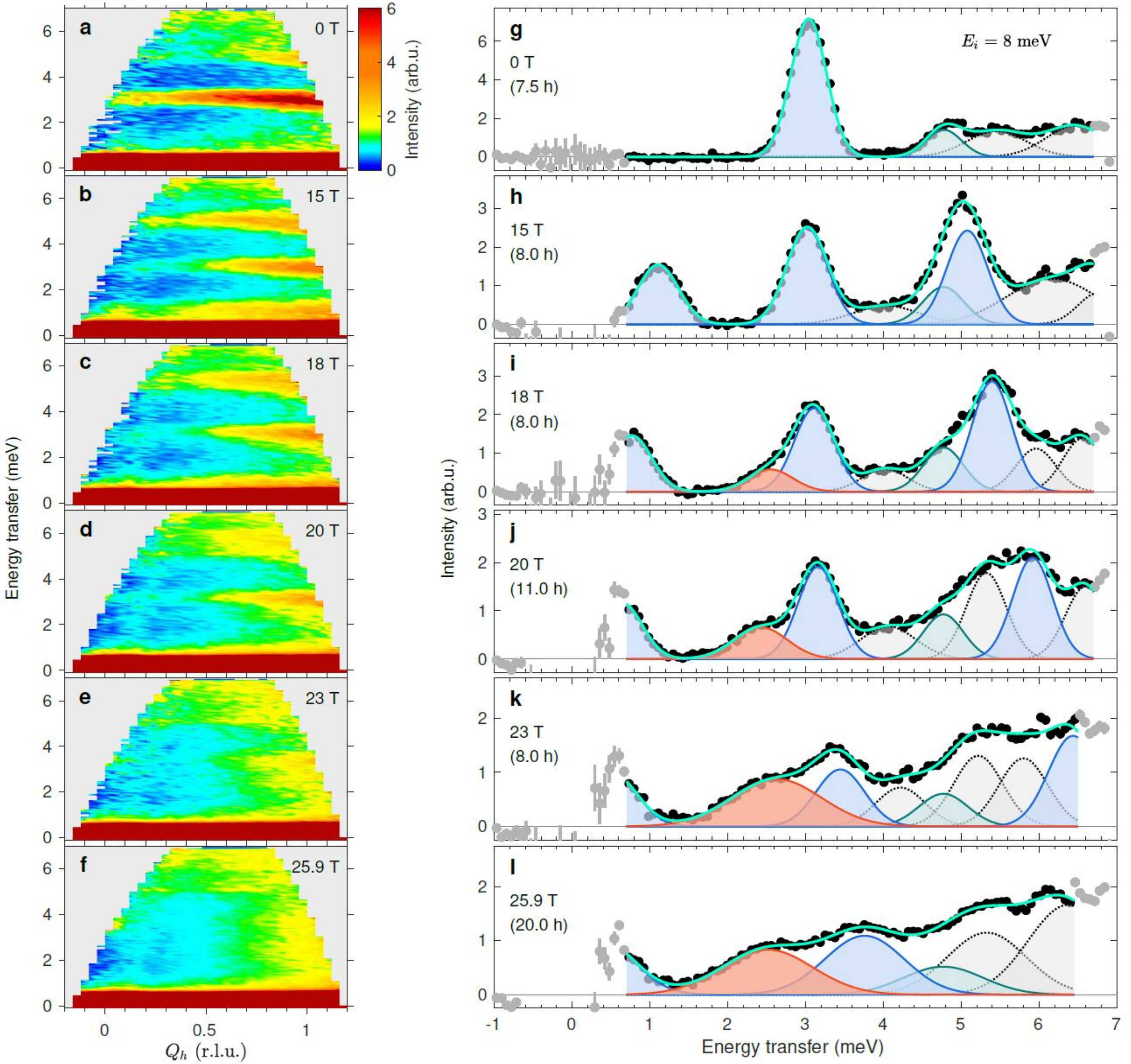
The Zeeman splitting of the three one-triplon branches forms a clear benchmark.

Careful fitting of constrained Gaussian peaks to a fixed number of the observed excitation features reveals a significant amount of excess scattered intensity emerging below the t_0 mode (red) and below the t_- mode (turquoise) at and above 18 T. At 26 T this intensity exceeds the weight in the one-triplon branches.

However, the resolution is not sufficient to determine either the number or the positions of the possible modes contributing to this intensity.

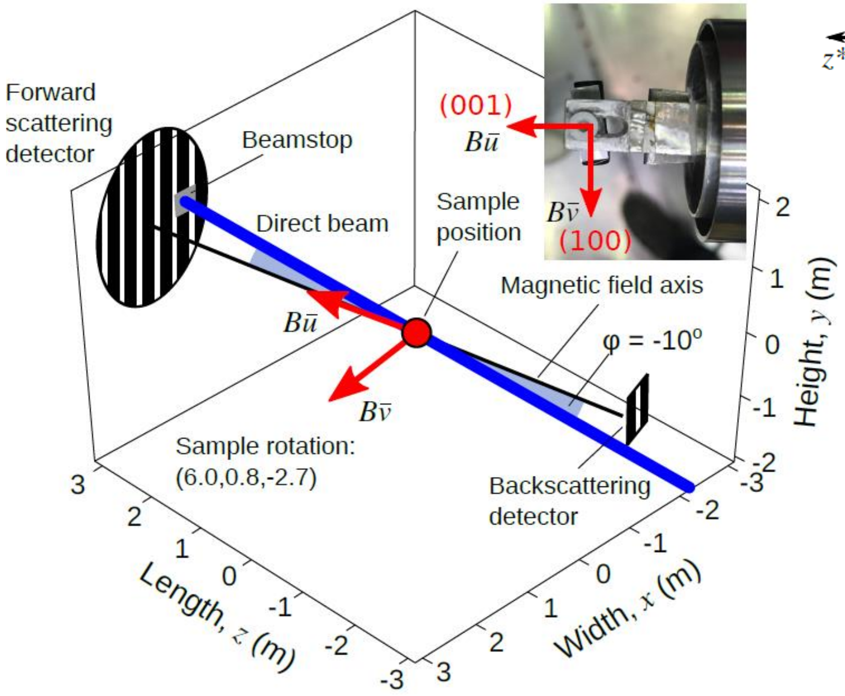
Some theoretical input required …

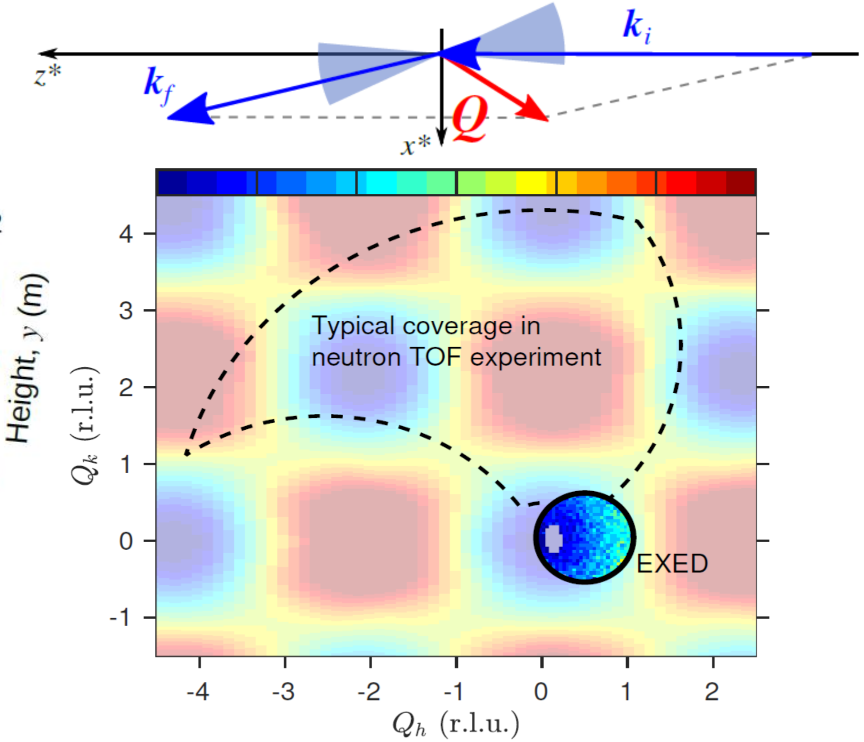
E. Fogh *et al.*, Field-induced bound-state condensation and spin-nematic phase in $SrCu_2(BO_3)_2$ revealed by neutron scattering up to 25.9 T, arXiv:2306.07389.

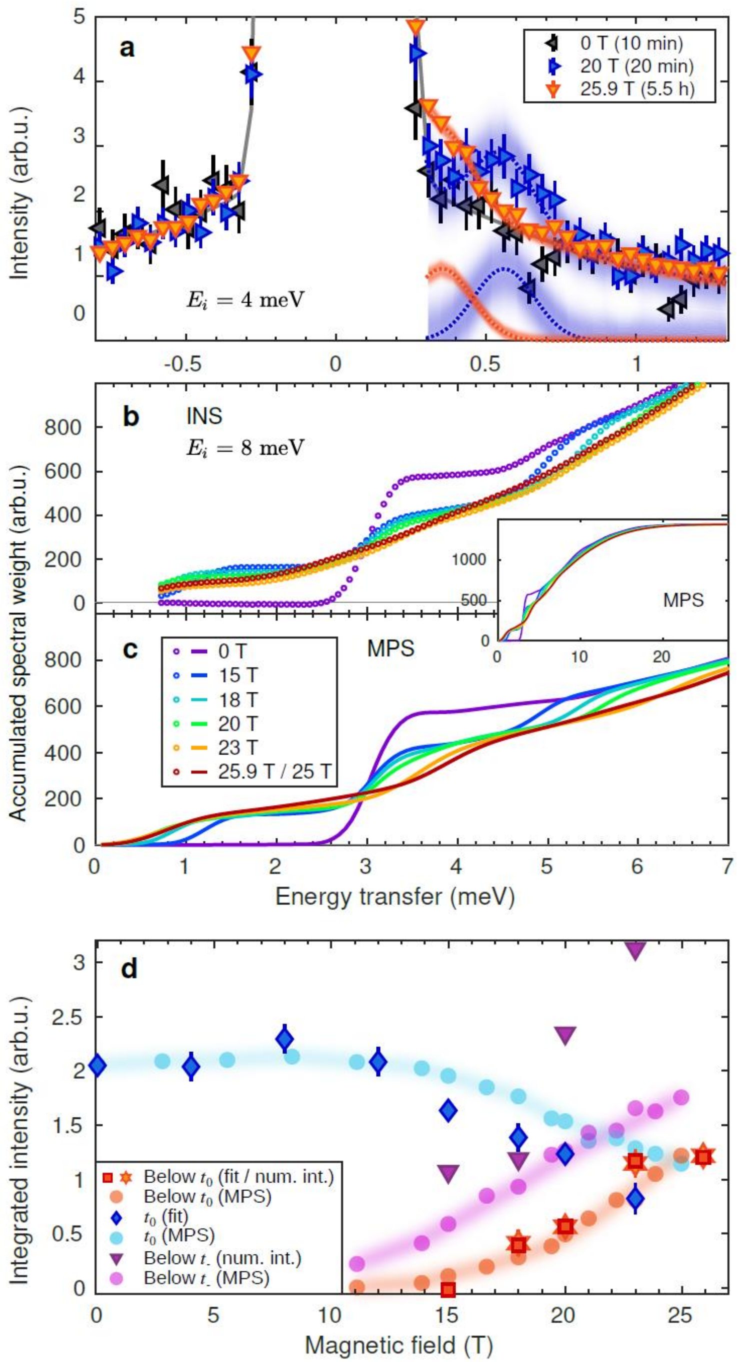


INS Details

Important note: this is a really hard experiment – the geometry is highly constrained by the presence of the high-field magnet, in real space and hence in reciprocal space.







Nevertheless, it is still possible to extract further valuable information:

(a) The lowest-lying modes can be followed down to approximately 0.3 meV.

(b,c) The integrated weight reveals the one-triplon branch crossings and can be compared with numerics to benchmark the total weight in all excitations.

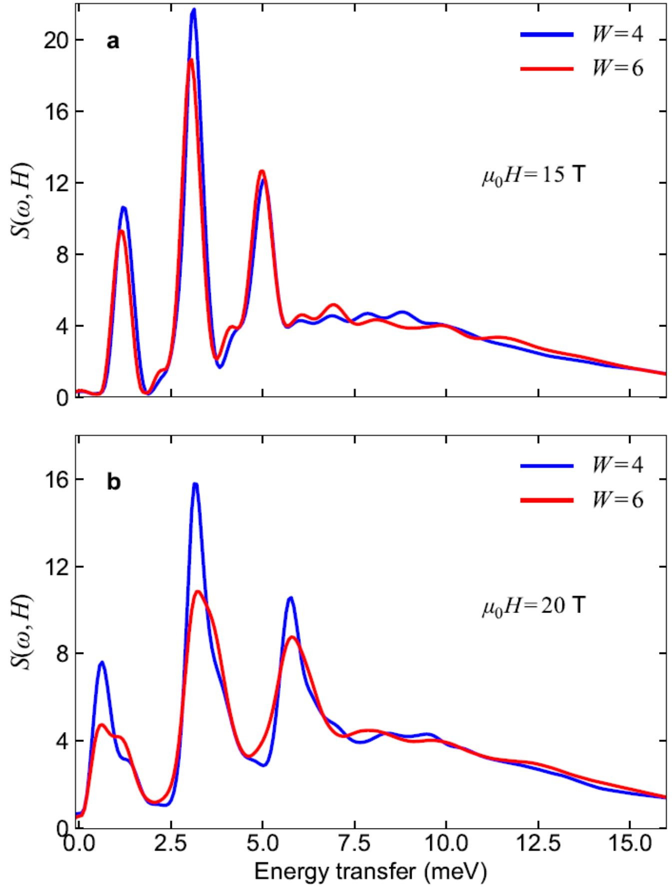
(d) It is possible to quantify the shifts in spectral weight from the one-triplon branches to the multi-triplon states: the effect is quite dramatic.

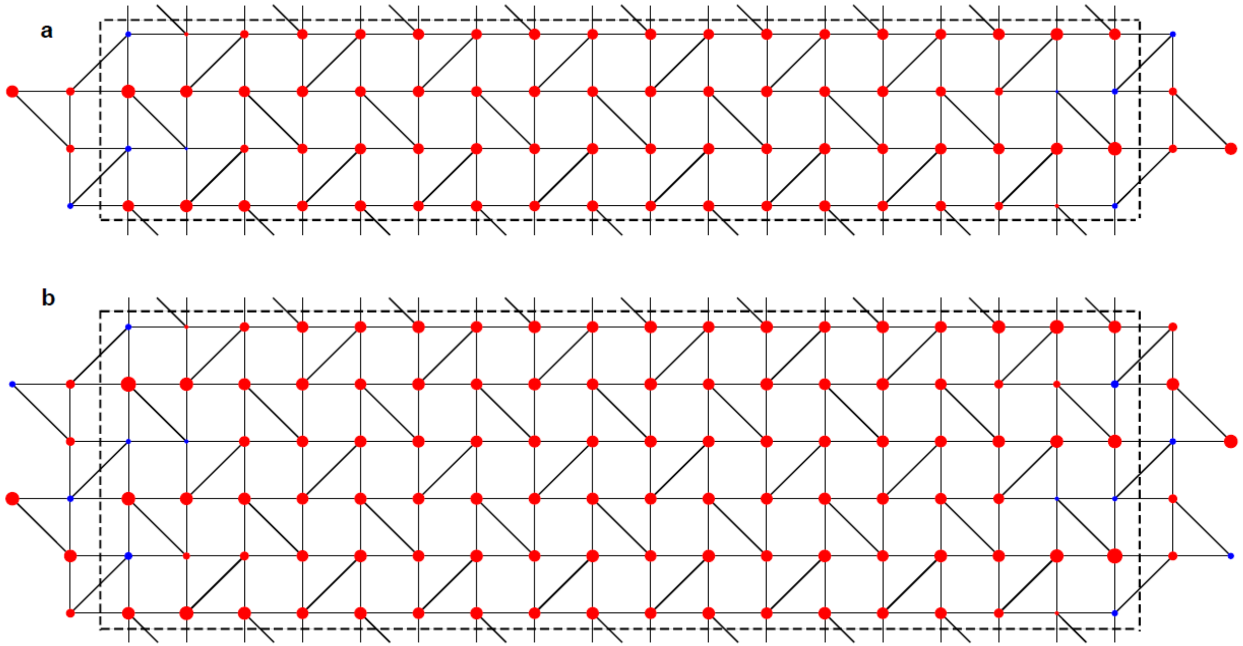
Cylinder MPS

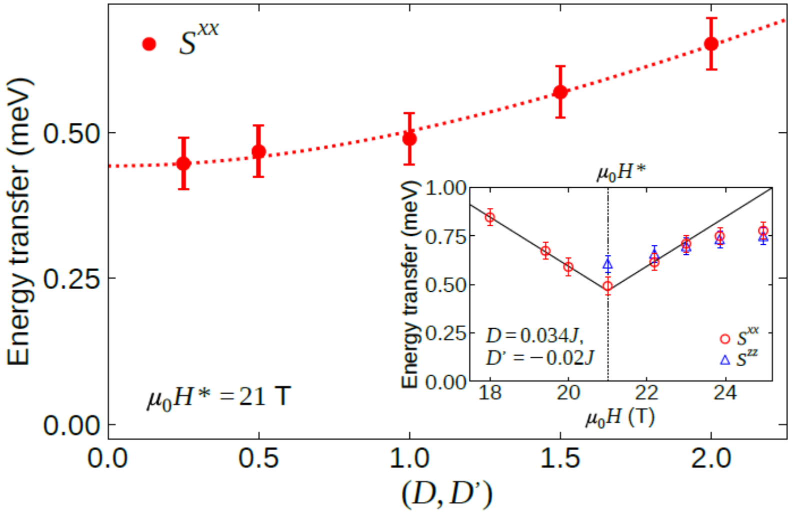
Cylinder MPS methods offer a powerful “new” means of computing the spectral functions of systems that can be formulated as a quasi-1D problem. Cylinder widths W = 4 and 6 are extremely small, and one would like to know that they are in some way representative of the 2D system. The calculation time scales as $W^2\exp(3W)$.

In the ideally frustrated geometry of the Shastry-Sutherland model we profit from the extreme localisation of the magnetic excitations, which makes such small W acceptable.

Technical note: it is not possible to stabilise a state of uniform magnetization in an applied field unless the DM interactions are included; we use those of SCBO.



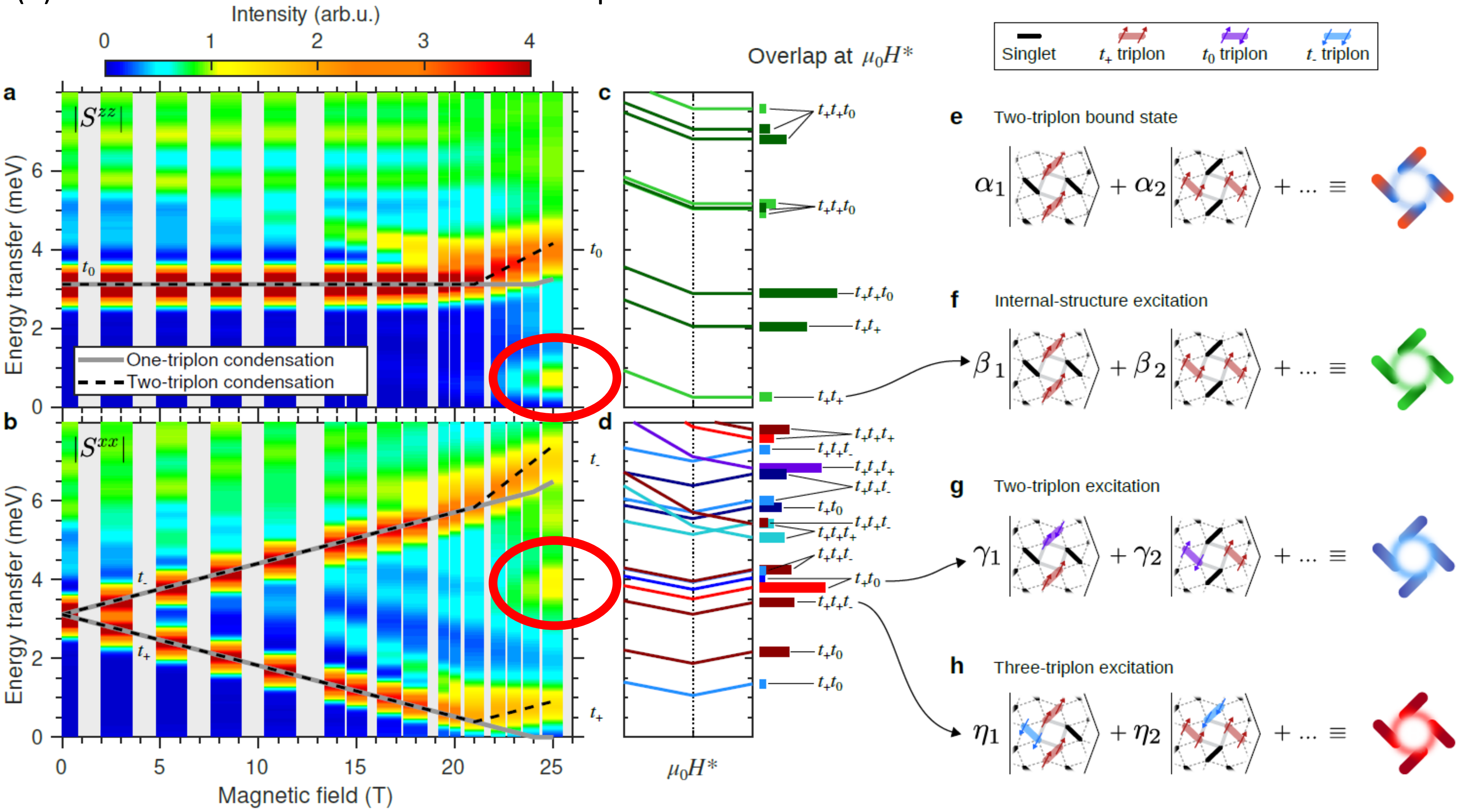




Important: could the DM interaction produce the gap observed in the INS experiment ? **NO**

MPS + ED

By MPS one may calculate the longitudinal and transverse response functions separately.
(1) Characteristic behaviour of the one-triplon branches: minimum at 0.6 meV at 21 T.



(2) Clear anomalous contributions in S_{zz} at low ω due to internal modes of the |2,2> manifold and in S_{xx} at intermediate ω due to $|t_+t_0>$ excitations.

(3) Large additional spectral weight due to three-triplon excitations, among which the $|t_+t_+t_->$ modes lie lowest.

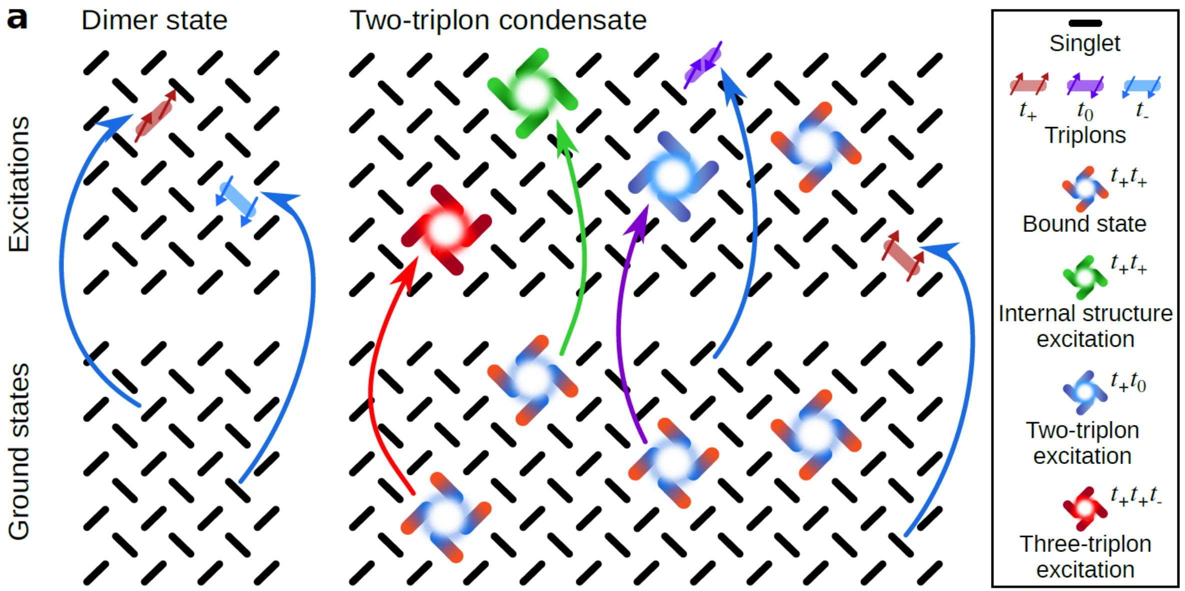
We perform ED calculations at $H = 0$ and with no DM terms to identify the state quantum numbers $|S,S_z>$. For this 4x4 is sufficient.

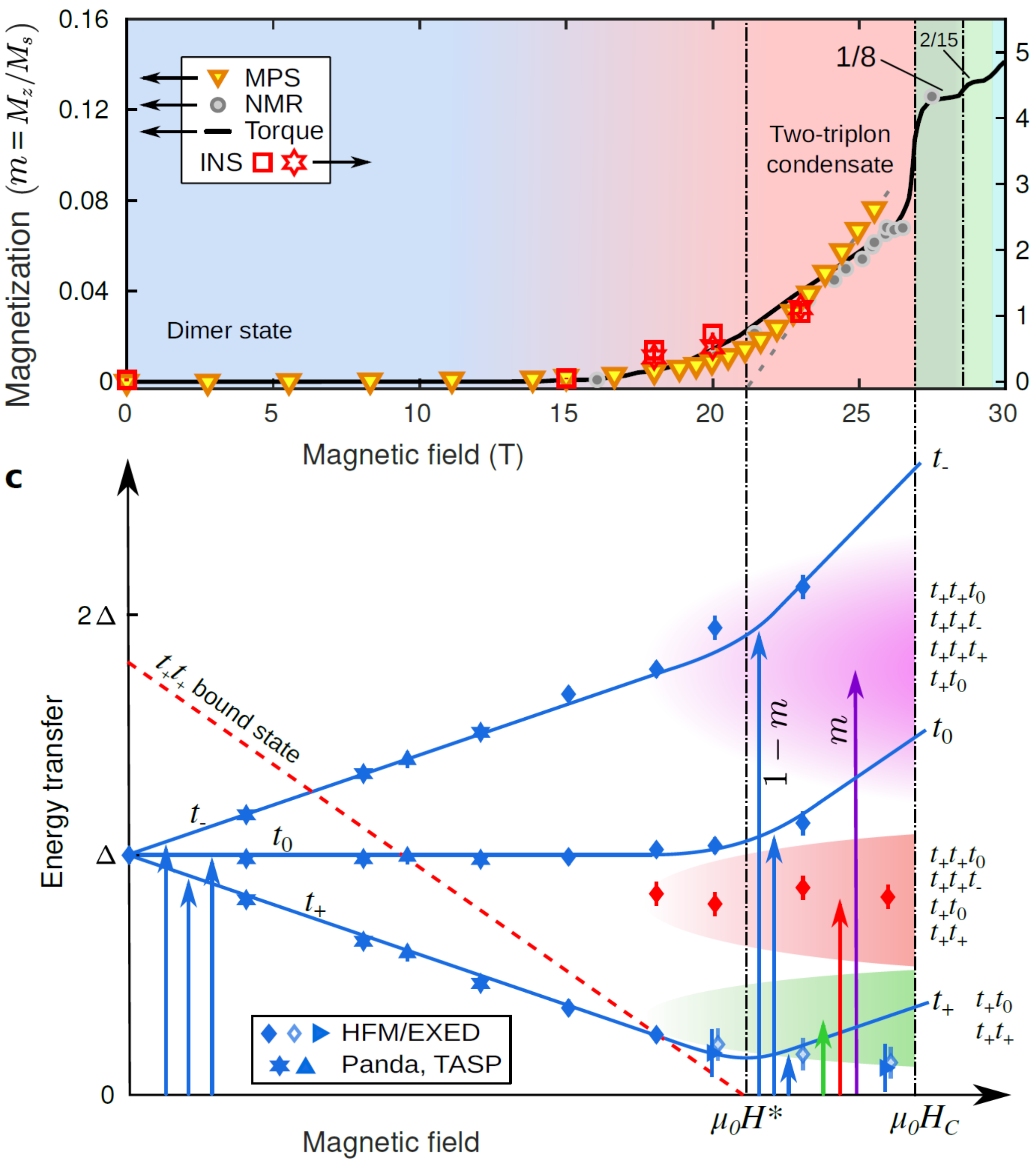
Summary I

The spin-nematic ground state is difficult to detect directly. However, its fingerprints are very clear in the spectrum:

- clear field-evolution of the one-triplon branches;
- number, type and weight of multi-triplon excitations.

The spin-nematic phase has a close analogy with super-conductivity: it is a BEC of Cooper pairs of bosons, with a finite single-particle (i.e. triplon) gap of half the binding energy. These are ultralocal Cooper pairs ("BEC" limit). *This phase also competes with other instabilities, such as the bosonic CDW order that prevails on the 1/8 plateau.*

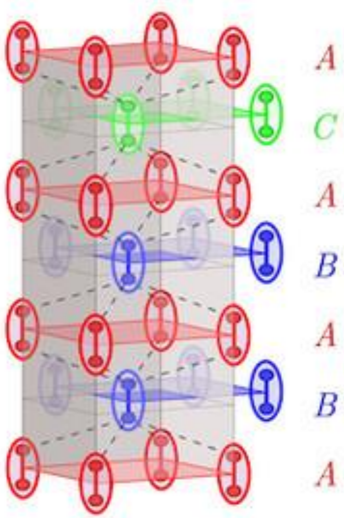


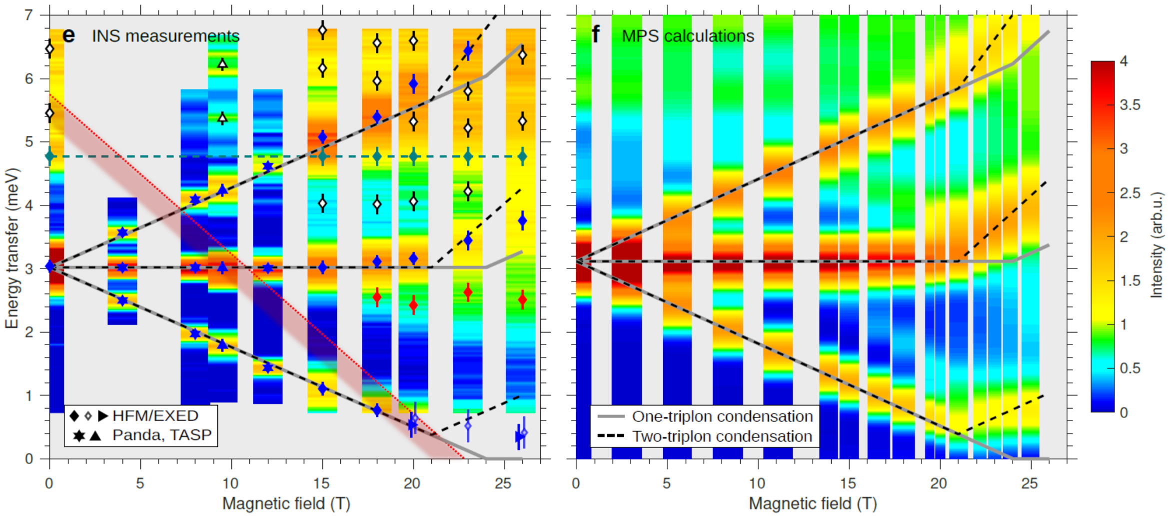


Roadmap

1) Novel quantum phases:

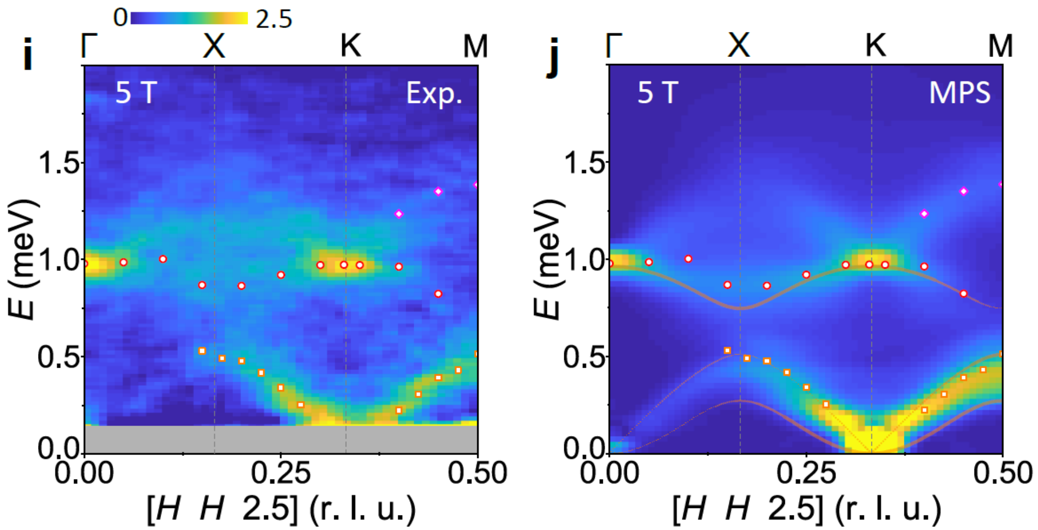
field-induced spin nematic





2) Novel quantum phase transitions:

quasi-2D Bose-Einstein universality



3) Novel excitation spectra:

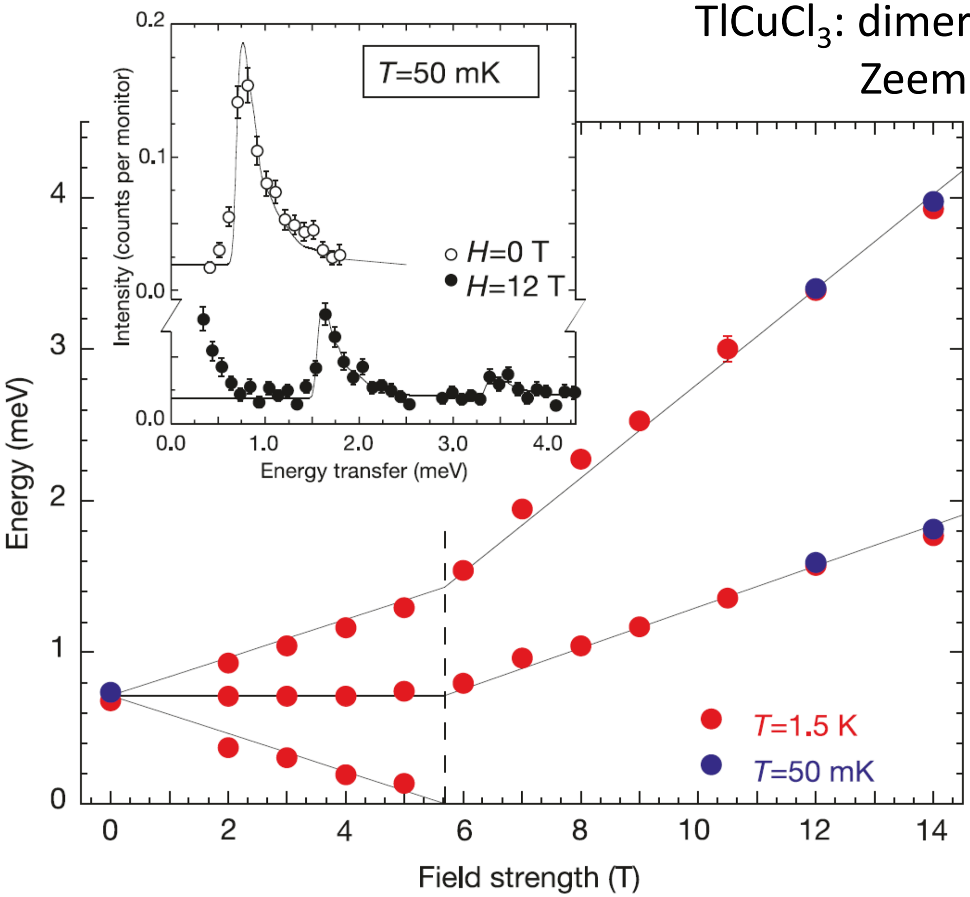
field-controlled continua

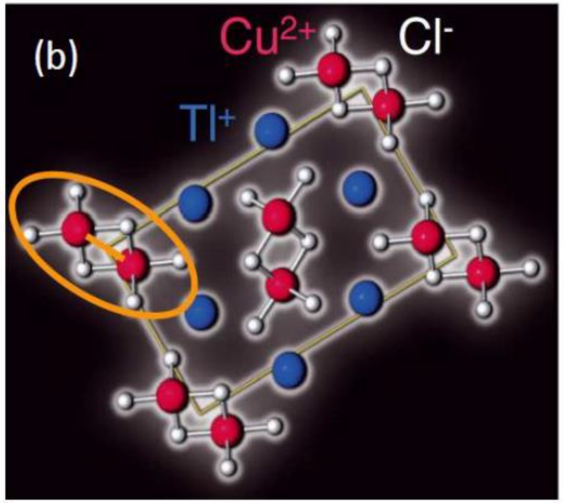
“Bose-Einstein Condensation of Magnons”

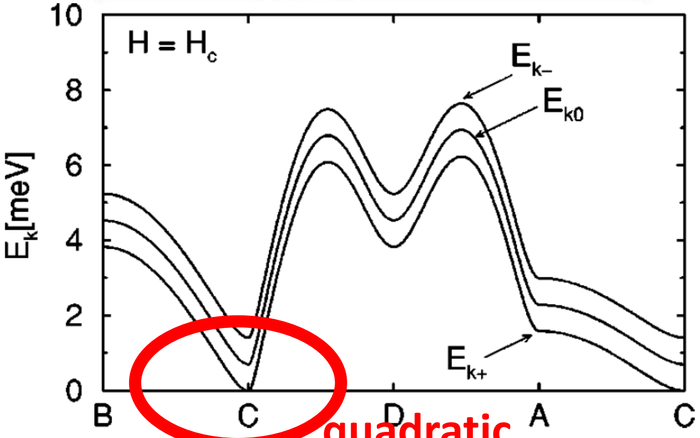
Heisenberg model $\hat{\mathcal{H}} = \sum_{[i,j]_m} J_m \boldsymbol{S}_i \cdot \boldsymbol{S}_j$ in an applied magnetic field $\hat{\mathcal{H}}_m = -g\mu_B \sum_i \boldsymbol{B} \cdot \boldsymbol{S}_i$

$TlCuCl_3$: dimerised quantum spin system,
Zeeman-split triplet excitation

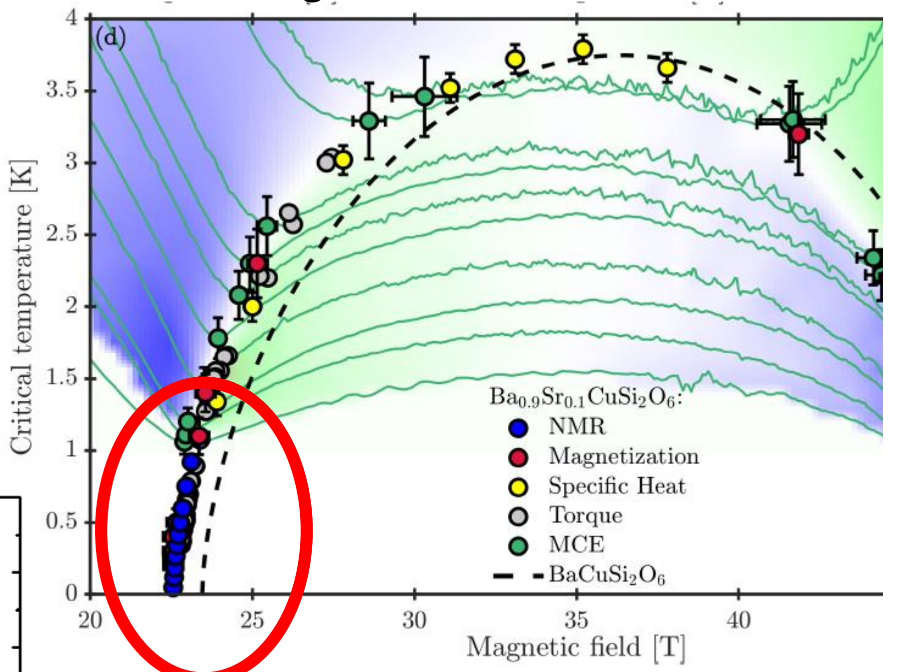
Quantum phase transition: spontaneous breaking of U(1) symmetry, field-induced transverse magnetic order.







quadratic



$$T_c(H) = \alpha(H - H_{c1})^{\phi}$$

$$\phi = z/d \qquad \omega \propto k^z$$

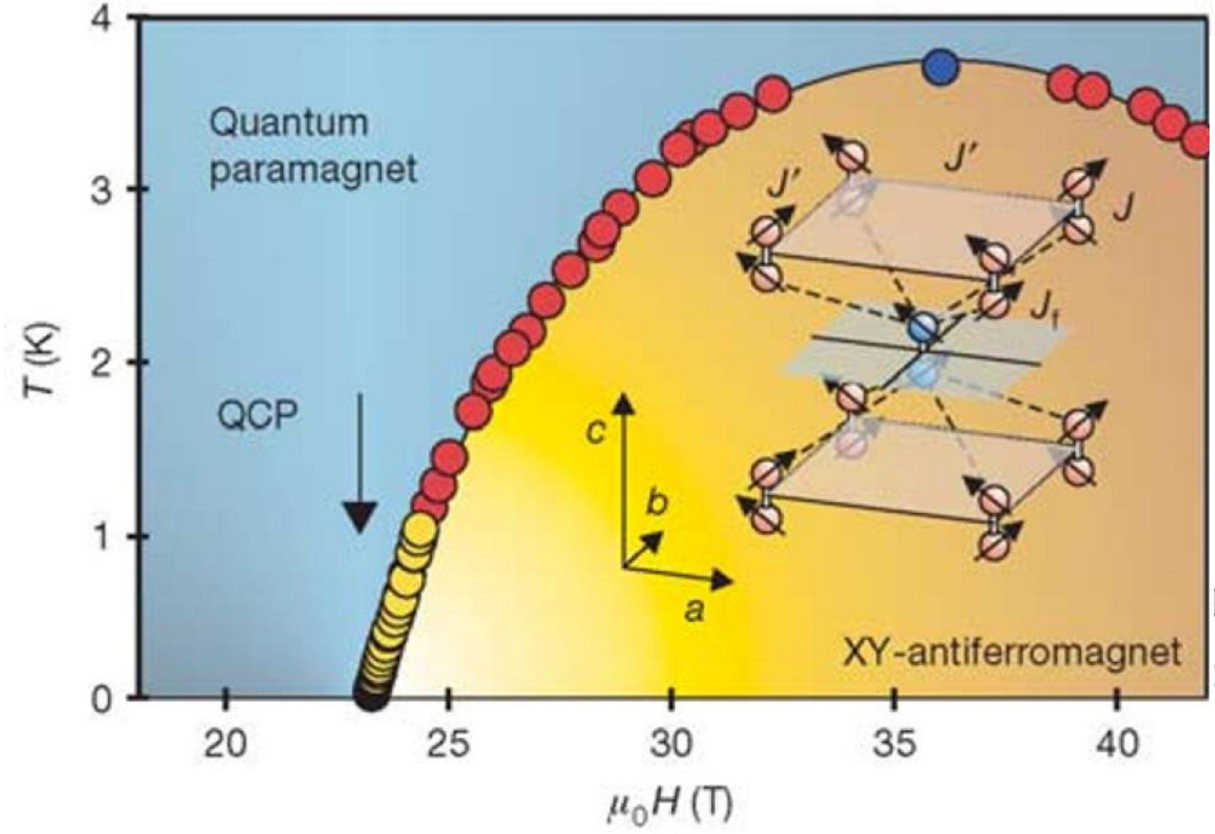
$\phi = 2/3$ for free bosons in 3D:
Bose-Einstein universality

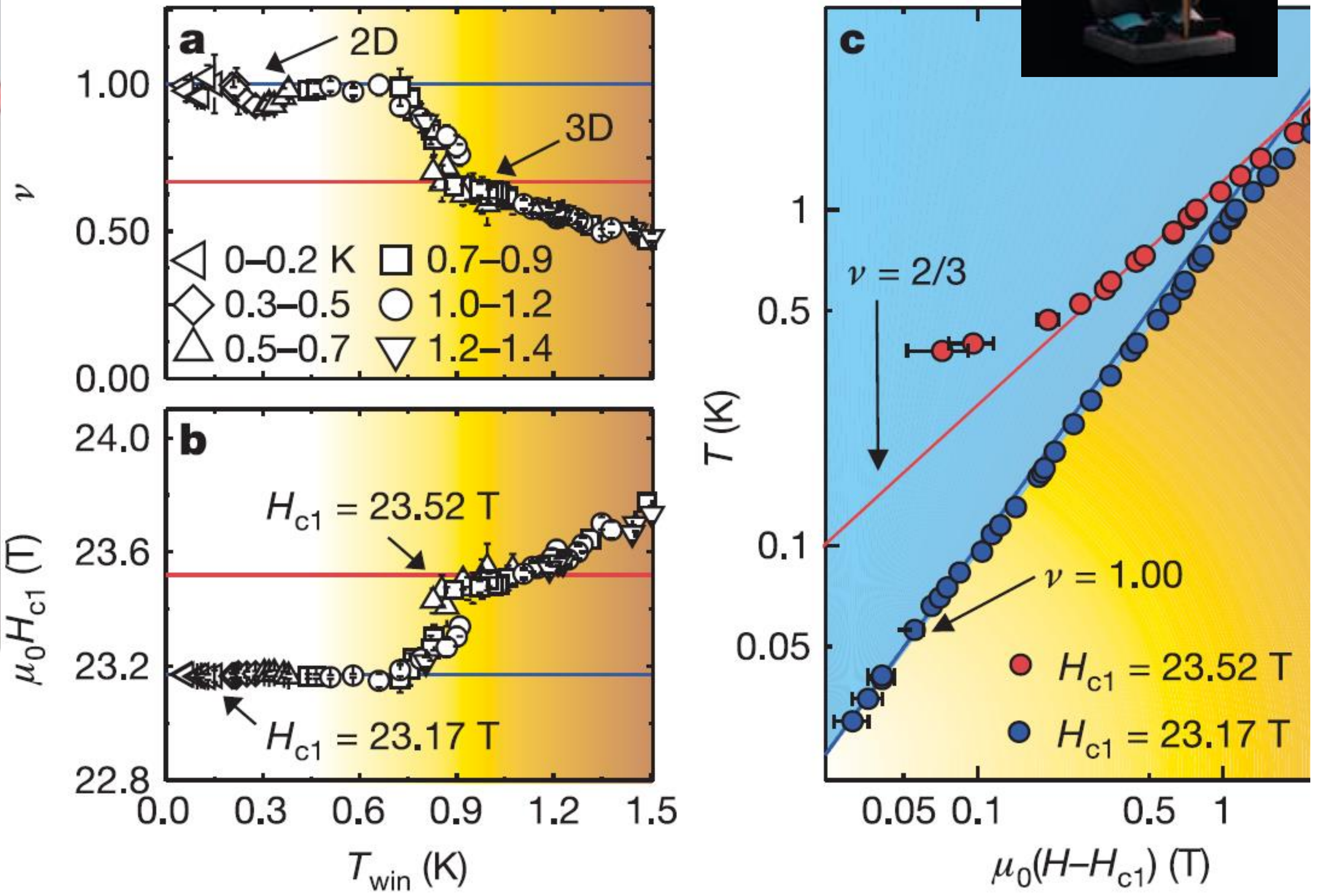
[1] Ch. Rüegg *et al.*, Nature **423**, 62 (2003).

[2] M. Matsumoto, B. Normand, T. M. Rice and M. Sigrist, Phys. Rev. B **69**, 054423 (2016).

Han Purple: Dimensional Reduction ?

$BaCuSi_2O_6$ is a purple pigment known in the Han Dynasty. It is also a bilayer square-lattice quantum magnet with S = 1/2 Cu^{2+} spin dimers arranged in an offset (bcc) configuration. Early measurements of the magnetic torque [1] suggested that the critical exponent was not ϕ = 2/3 but 1, characteristic of 2D, leading to a theory of frustration-induced “dimensional reduction” that was then hotly contested …

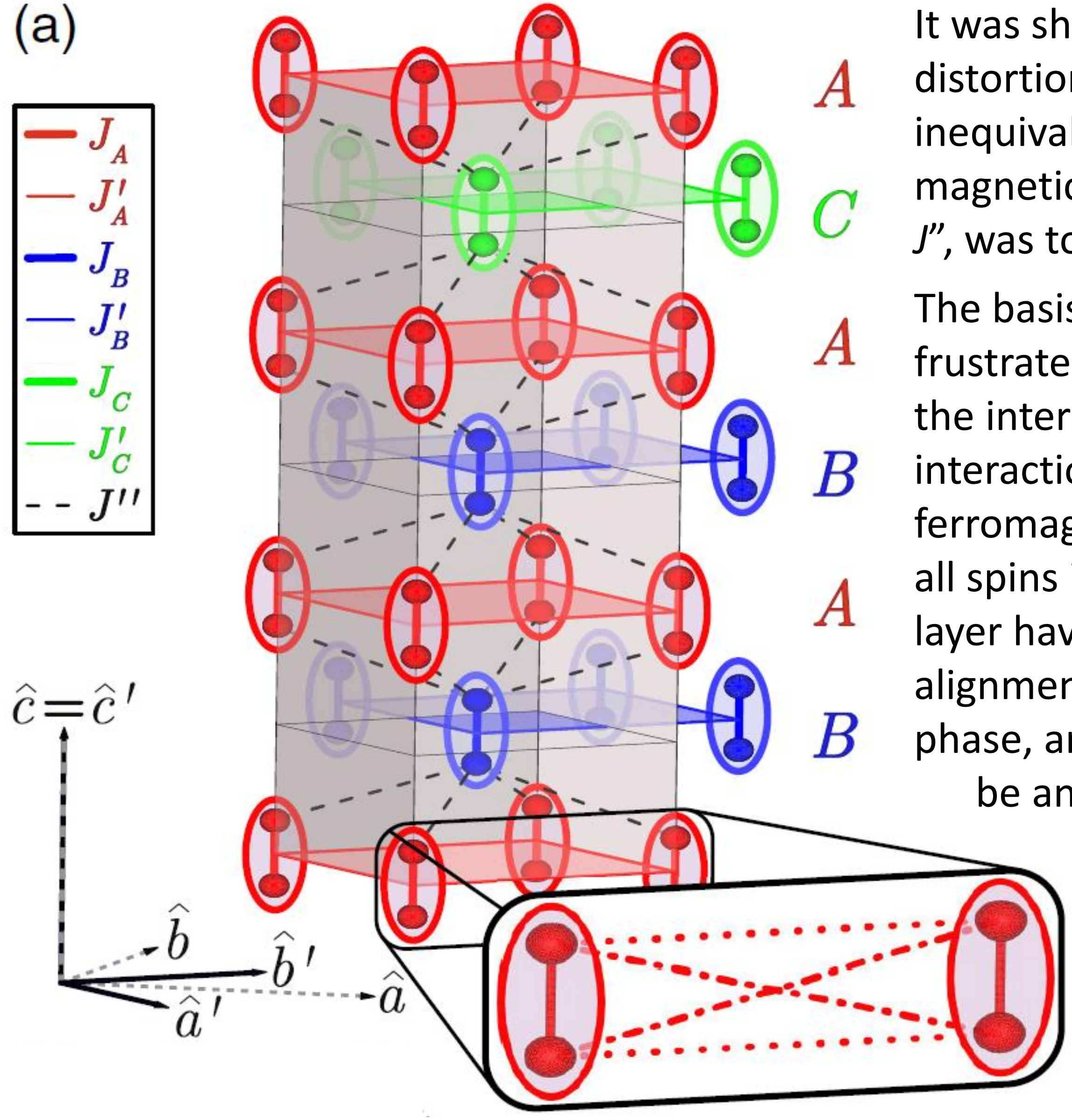




Later research [2]: it’s not frustrated …

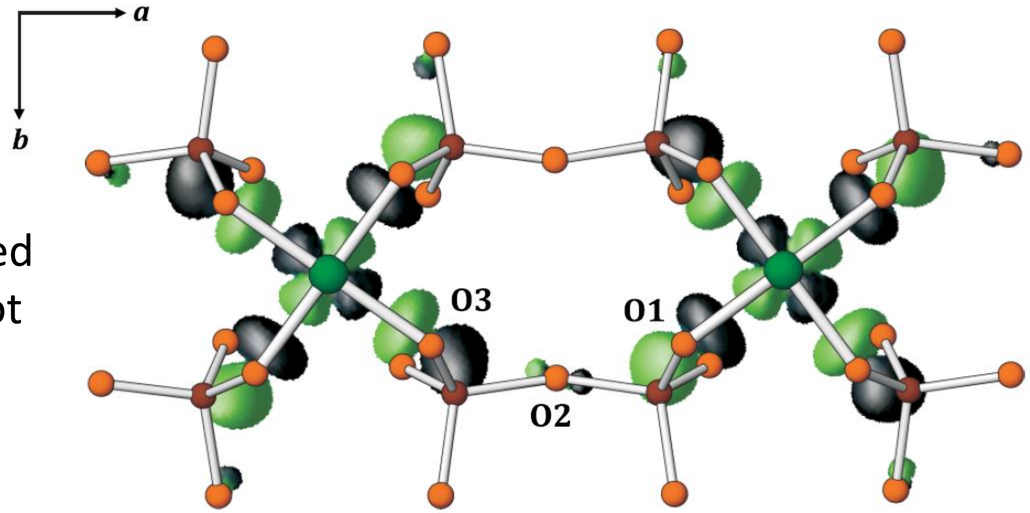
[1] S. E. Sebastian *et al.*, Nature **441**, 617 (2006).
[2] V. V. Mazurenko, M. V. Valentyuk, R. Stern and A. A. Tsirlin, PRL **112**, 107202 (2014).

$BaCuSi_2O_6$: layer-modulated structure



It was shown rather early that the tetragonal-to-orthorhombic structural distortion taking place at 90 K also causes (at least) 3 magnetically inequivalent bilayers [1]. At this time it was not possible to identify the full magnetic Hamiltonian, and in particular the direct interbilayer coupling, *J*", was too small to resolve (effective *J*" is much lower still … below).

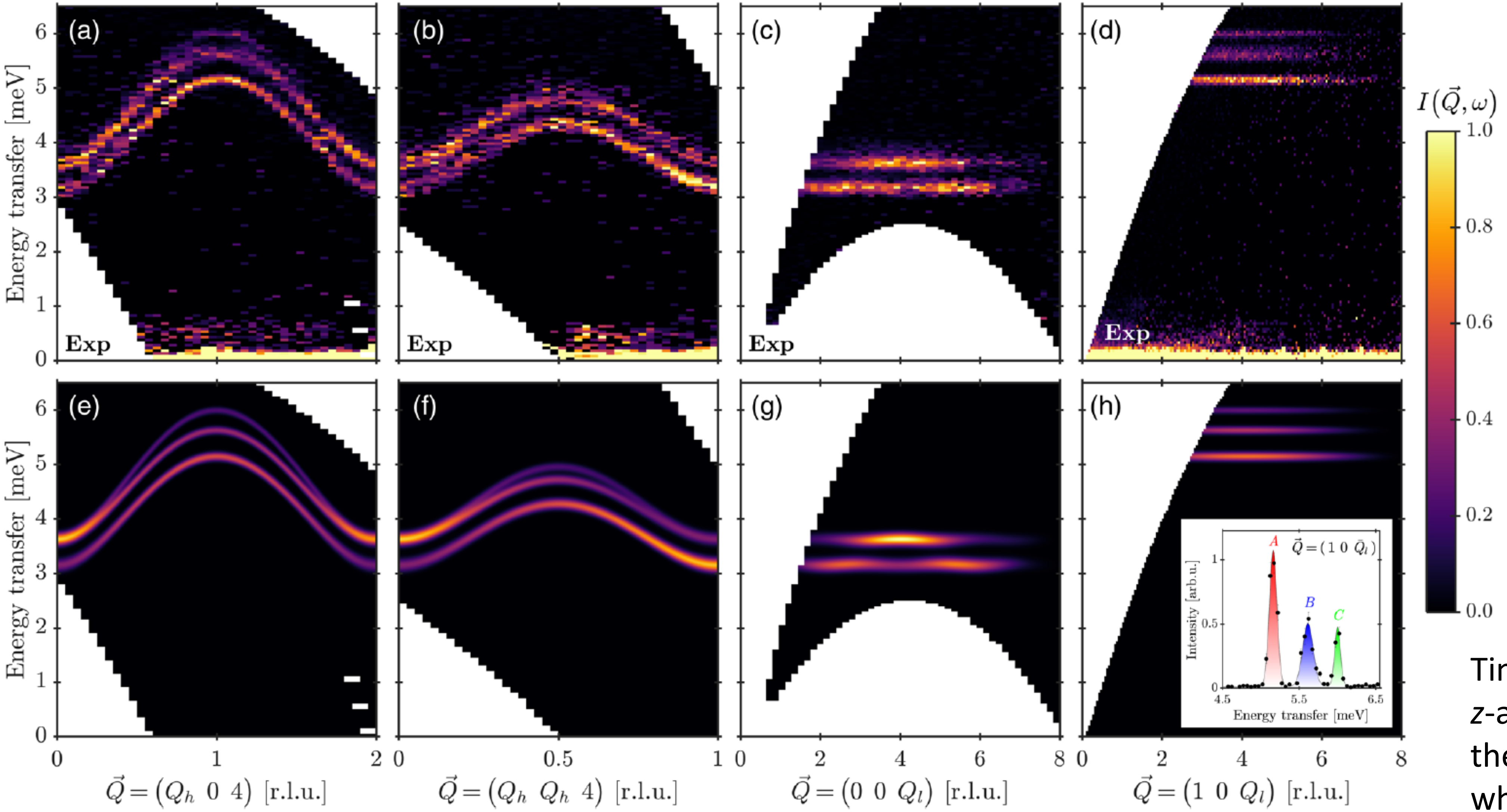
The basis of the claim that the interbilayer coupling should not be frustrated was the observation that intrabilayer orbital alignments favour the interlayer superexchange path, making the **effective** intralayer interaction ferromagnetic. Then all spins in the same layer have the same alignment in the ordered phase, and there cannot be any frustration.



[1] Ch. Rüegg *et al*., Phys. Rev. Lett. **98**, 017202 (2007).
Figure: S. Allenspach *et al*., Multiple Magnetic Bilayers and Unconventional Criticality without Frustration in $BaCuSi_2O_6$, Phys. Rev. Lett. **124**, 177205 (2020).

$BaCuSi_2O_6$: magnetic Hamiltonian

Back up to zero field: first show this is three-bilayer system and establish the magnetic interactions .



Interaction parameters in meV:

$$J_A = 4.275(5)$$
$$J_B = 4.72(1)$$
$$J_C = 4.95(2)$$
$$J'_A = -0.480(3)$$
$$J'_B = -0.497(8)$$
$$J'_C = -0.57(1)$$
$$J'' = -0.04(1)$$

Clear ratio of intensities A:B:C = 3:2:1

$$H_{c1} = 23.4\ \mathrm{T}$$

with

$$g_{\parallel} = 2.435$$

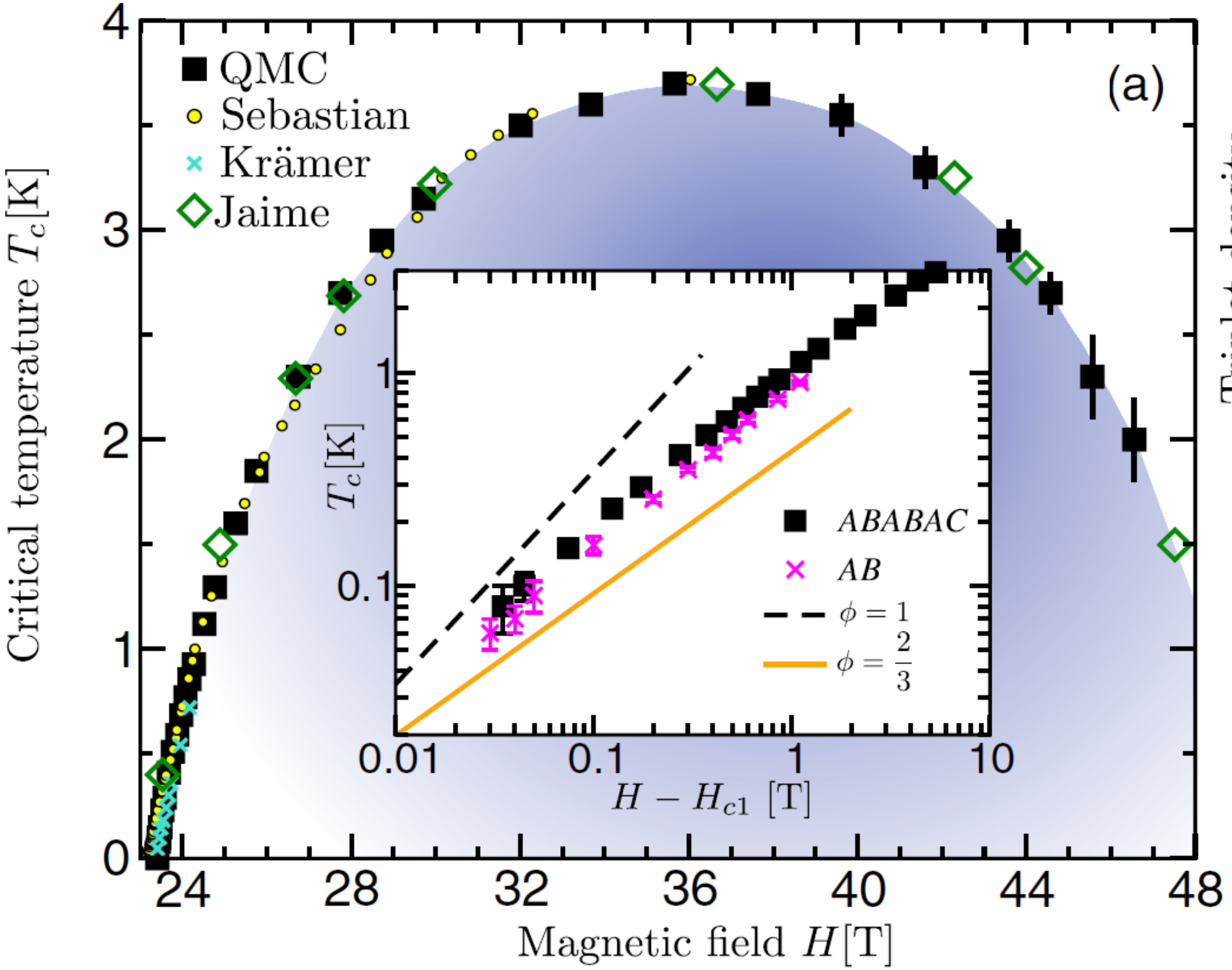
Tiny J'' fitted not from the z-axis dispersion but from the intensity distribution, which is quite sensitive.

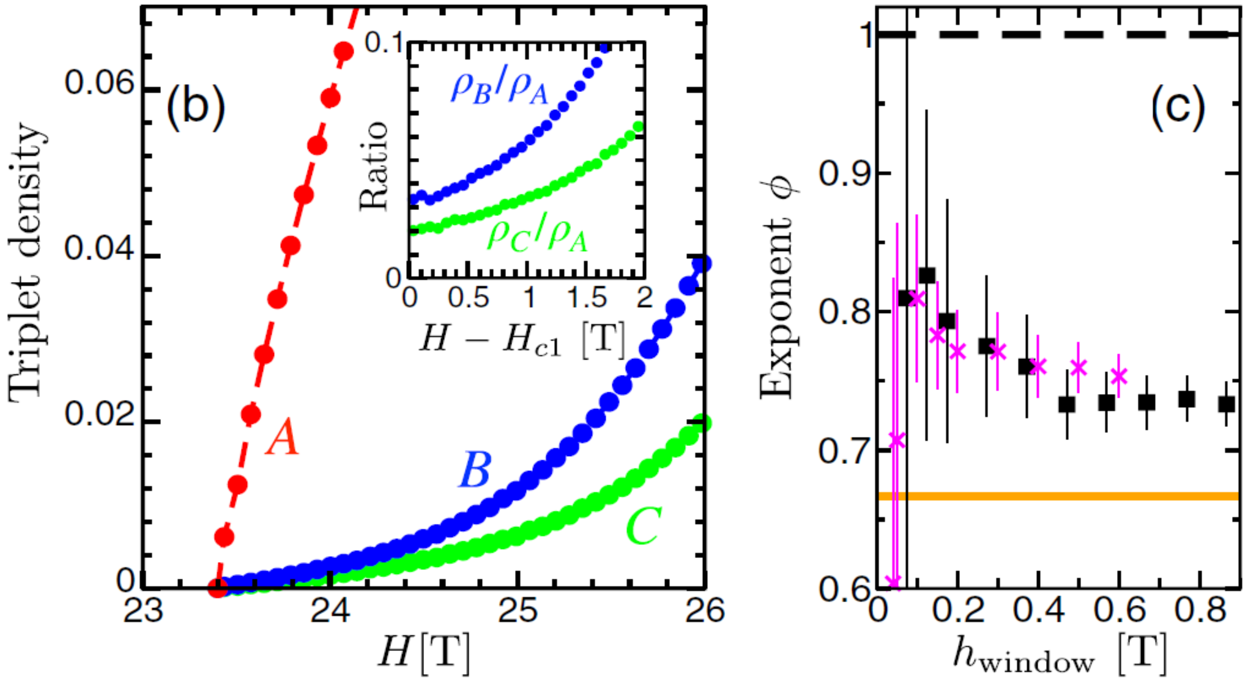
Field-Induced QCP: Quantum Critical Scaling

The presence of layer AB modulation introduces a new energy scale $J_B - J_A$ and hence an effective interlayer interaction:

$$\tilde{J}'' = (J'')^2/[J_B - J_A]$$

In $BaCuSi_2O_6$, these energies are respectively **5 K** and **0.04 K**. They introduce an extra field-temperature scaling regime near the QCP with nontrivial and nonuniversal scaling.



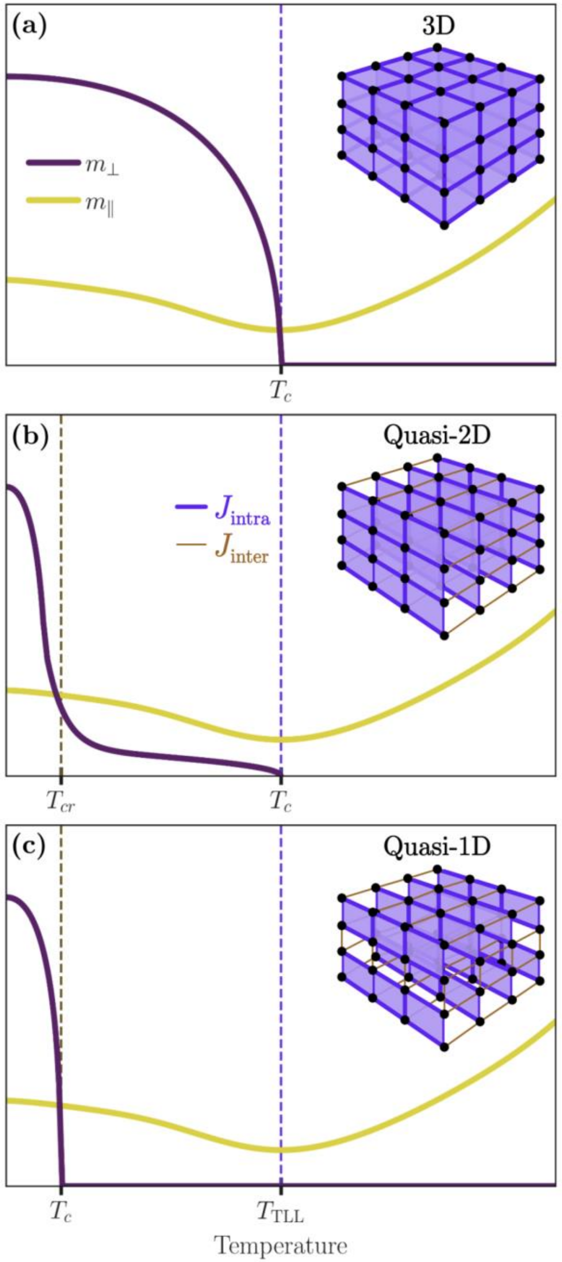


Define $h = H - H_c$, then the effective triplet tunneling between A bilayers is a smooth function of h, finite as $h \rightarrow 0$,

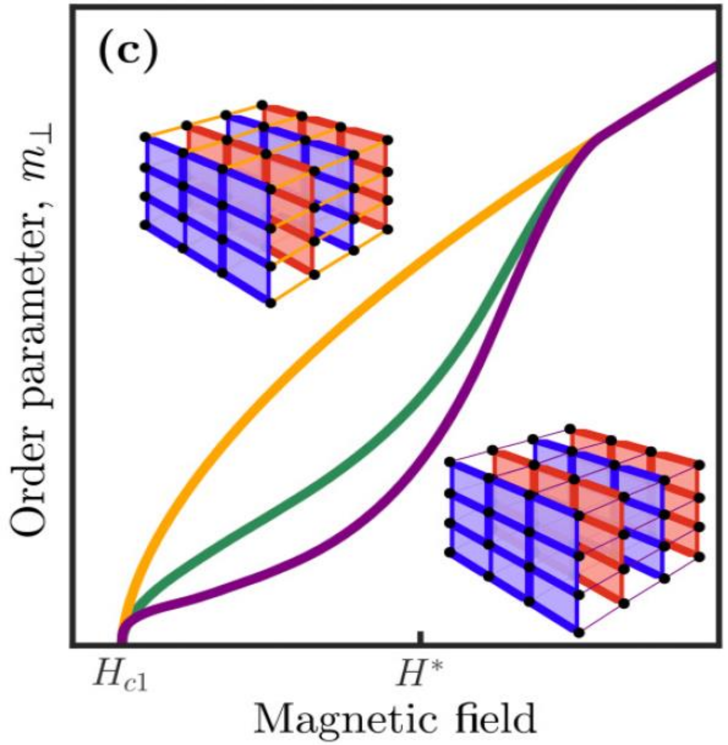
$$t_{3D}^{\text{eff}}(h) = t_c^{\text{eff}} + a_1 h + a_2 h^2 + O(h^3)$$

giving an anomalous effective exponent $2/3 < \phi(h) < 1$ when h falls in the scaling regime defined by the layer modulation.

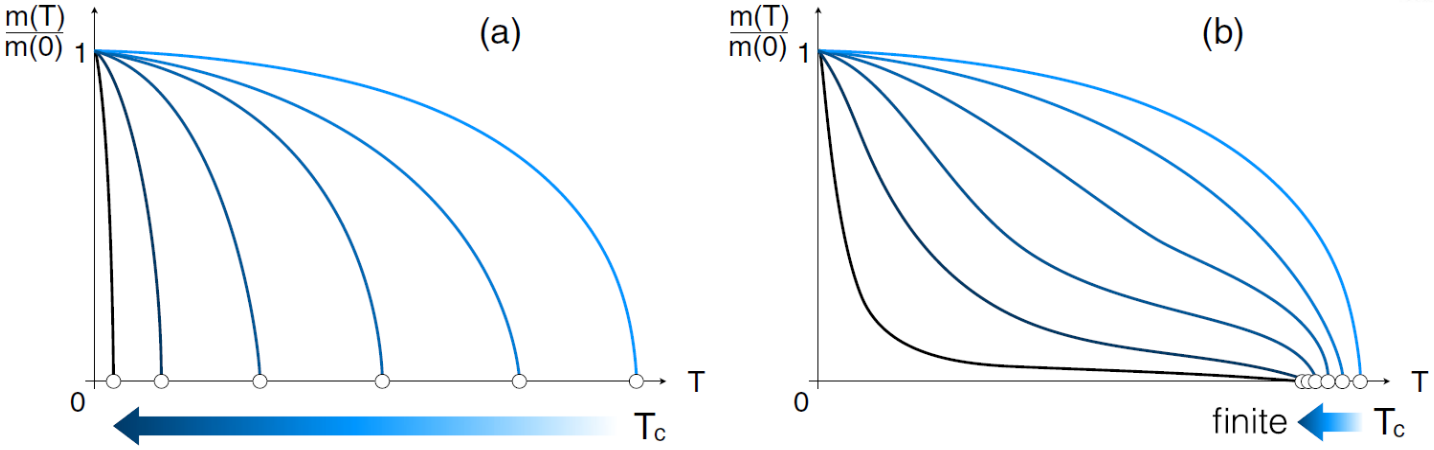
Prediction: Peculiarities in Quasi-2D



It has been predicted [1] on the basis of simple paradigm models that 2D systems could show some anomalous forms of behaviour in their magnetic ordering phenomena. The origin of this type of physics should lie in the proximity to the Berezinskii-Kosterlitz-Thouless (BKT) regime.



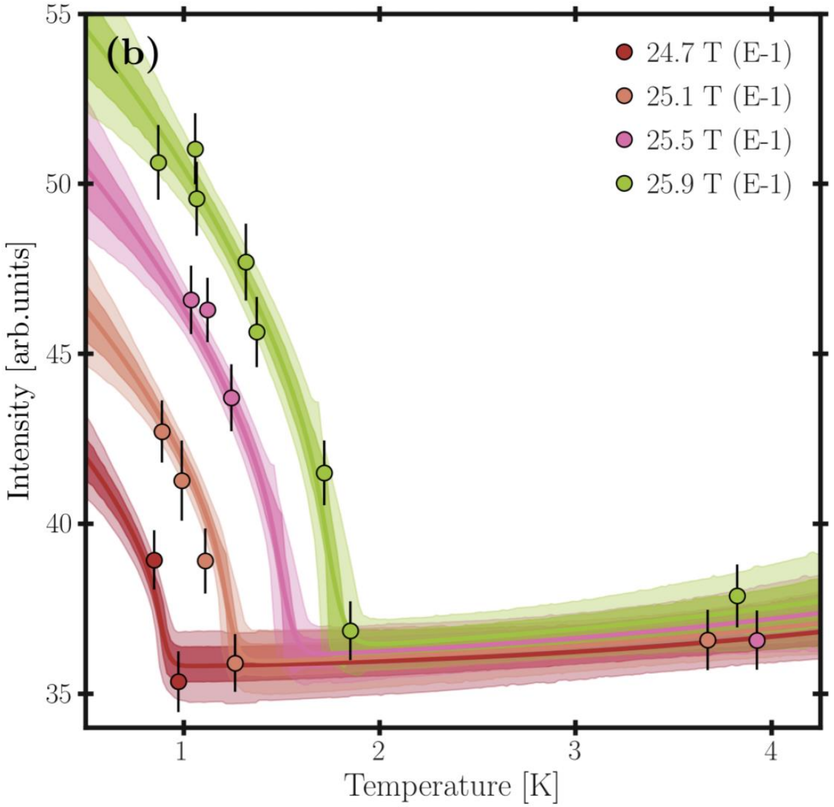
Having a strongly modulated 2D structure, with the new effective interlayer interactions these allow, presents a new set of opportunities to confirm or deny these predictions.

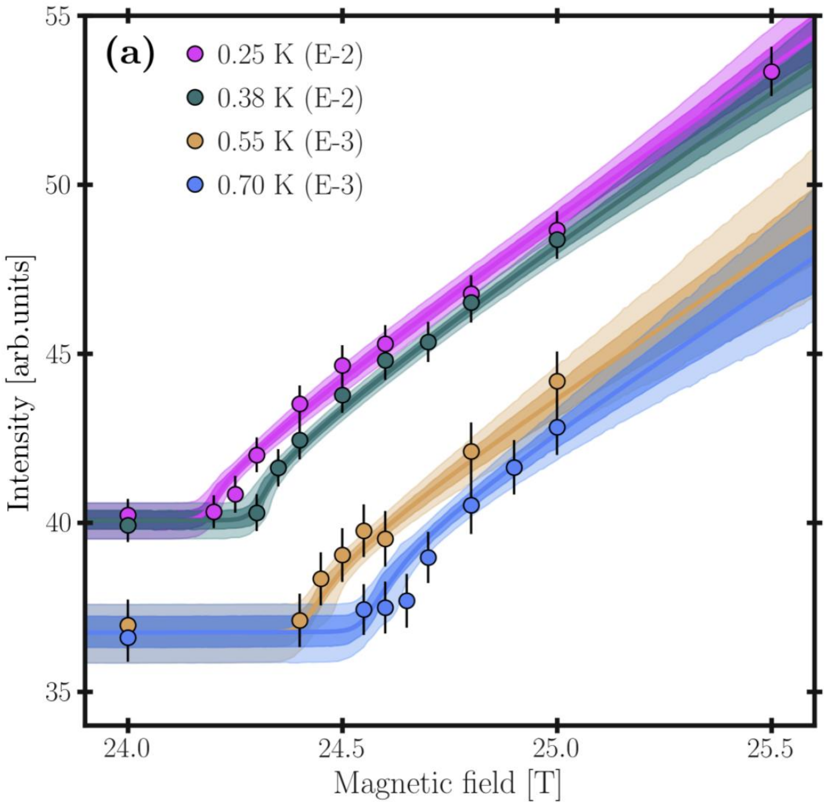


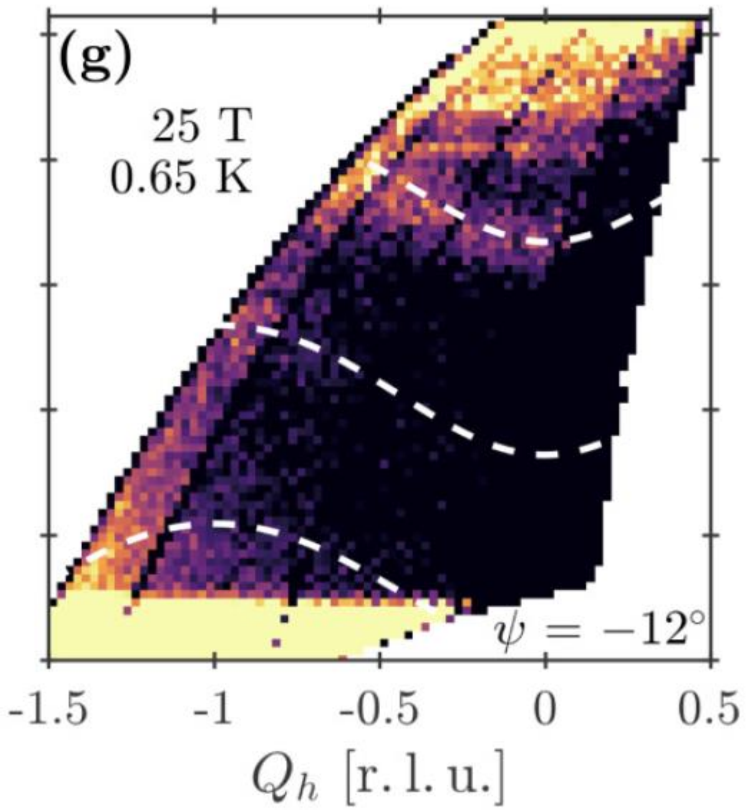
[1] S. C. Furuya, M. Dupont, S. Capponi, N. Laflorencie and T. Giamarchi, Phys. Rev. B **94**, 144403 (2016).

Experiment: High-Field Neutron Scattering

Back to HFM/EXED to measure (1) magnetic structure and (2) excitations. **Conclusion: looks very conventionally 3D.**







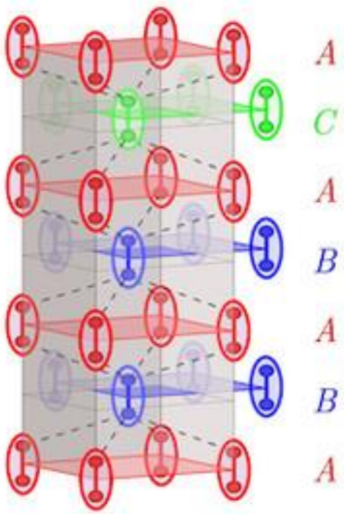
S. Allenspach *et al*., Investigating field-induced magnetic order in Han purple by neutron scattering up to 25.9 T, Phys. Rev. B **106**, 104412 (2022).

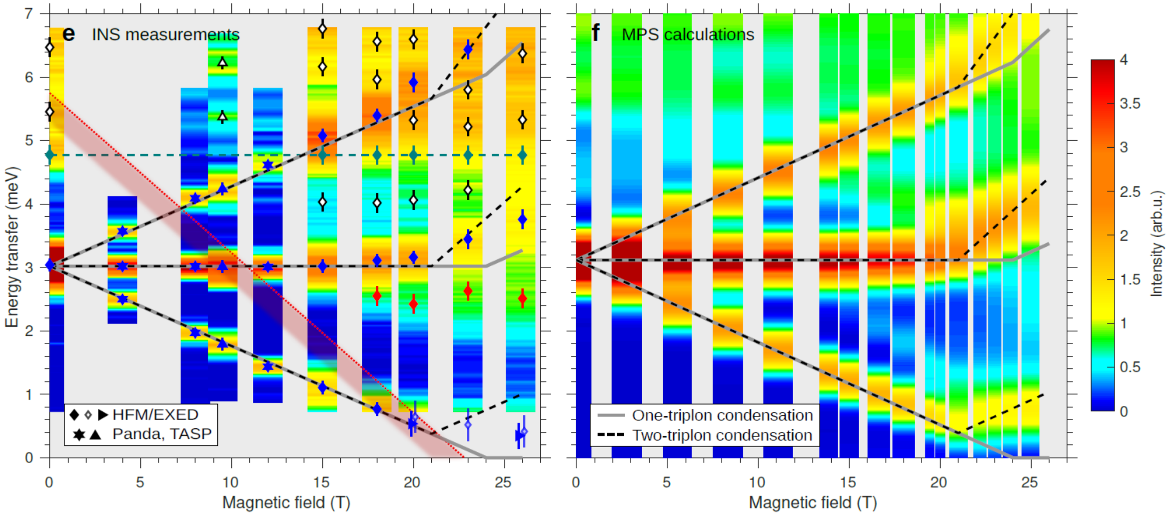
Summary II

$BaCuSi_2O_6$ is a fascinating example of a quasi-2D quantum magnet with layer modulation, which introduces completely new and potentially much lower interlayer energy scales; this is highly relevant in the new age of stacked atomically thin magnetic materials. However, $BaCuSi_2O_6$ itself is not nearly quasi-2D enough to exhibit dramatic new physics …

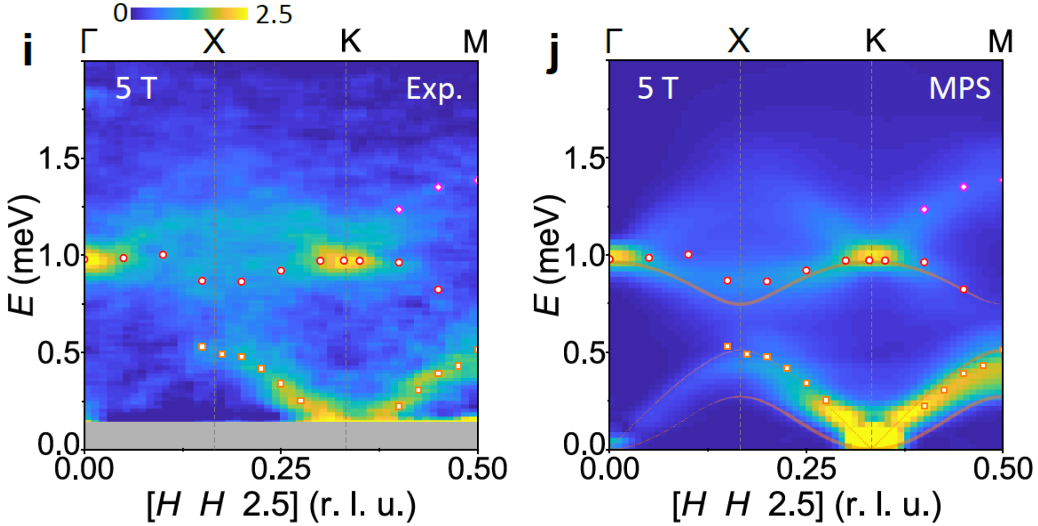
Roadmap

1) Novel quantum phases:
field-induced spin nematic





2) Novel quantum phase transitions:
quasi-2D Bose-Einstein universality



3) Novel excitation spectra:
field-controlled continua

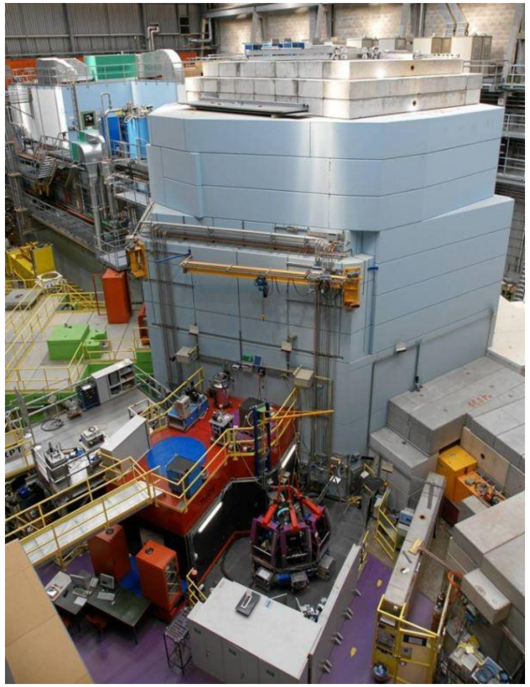
Neutron Scattering: Sources

Spallation Neutron Source (SNS), Oak Ridge, USA

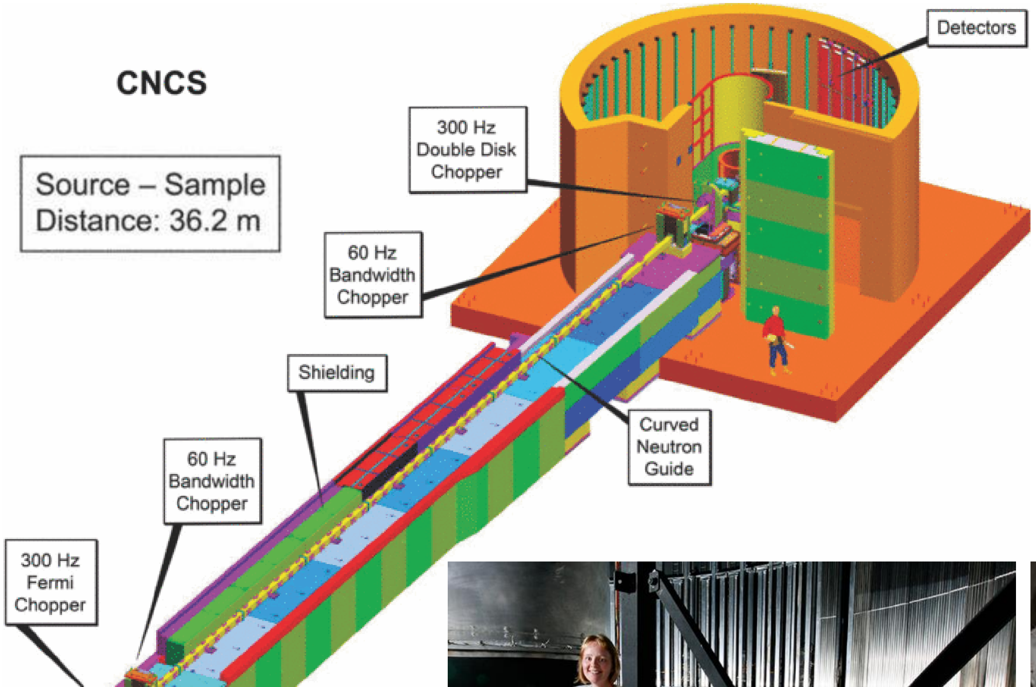
Spallation Neutron Source SINQ, PSI, Switzerland

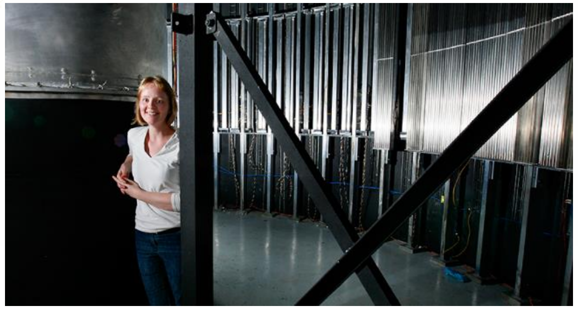
Proton accelerator

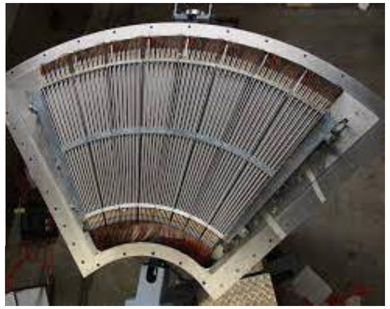
Target station



Neutron Scattering: Detectors







CNCS (Cold Neutron Chopper Spectrometer), ORNL

CAMEA (Continuous Angle Multiple Energy Analysis) spectrometer, PSI

SINQ beam hall

Now add complex sample environments: some or all of high-pressure cells, cryomagnets and dilution refrigerators …

S = ½ Triangular-Lattice Heisenberg Antiferromagnet (TLHAF)

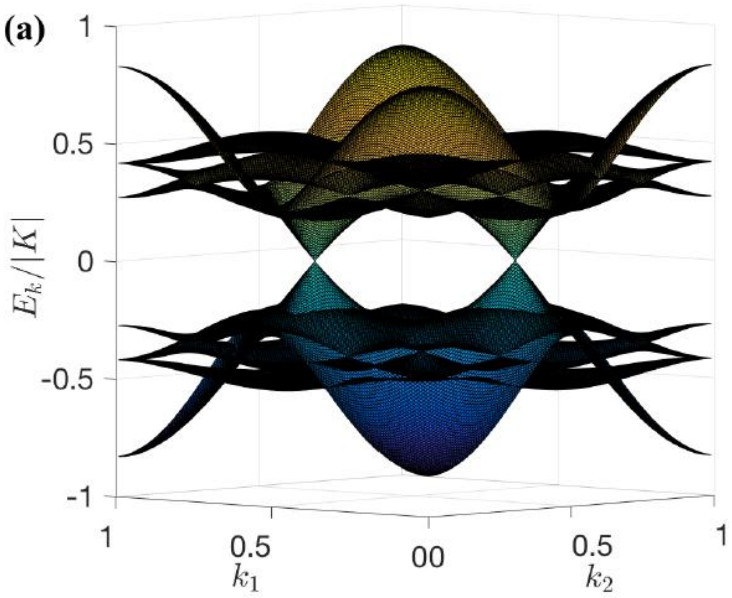
The TLHAF is the paradigm problem in frustrated quantum magnetism and spin liquids: in 2023 it is 50 years since Anderson proposed the short-range RVB state.

Now the nearest-neighbour TLHAF is known to be ordered, but the extent of order is not agreed at all. This makes the TLHAF a paradigm for weak magnetic order in the presence of strong quantum fluctuations.

Still the quantum spin liquid (QSL) phase is not far away: it is believed to be the ground state of the J_1-J_2 TLHAF with $0.06 < J_2/J_1 < 0.15$ (many references).

Method	E_b	M_0	Year	Max. size
This work	−0.18334(10)	0.161(5)	2020	∞
SB+1/N [30]		0.224	2018	432
DMRG [29]	−0.1837 (7)		2016	
CC [28]	−0.1838	0.21535	2016	10*
SB [26]		0.2739	2015	∞
SWT [26]		0.2386	2015	∞
SE [26]		0.198(34)	2015	∞
CC [27]	−0.18403(7)	0.198(5)	2015	10*
CC [24]	−0.1843	0.1865	2014	10*
VMC [25]	−0.18163(7)	0.2715(30)	2014	324
SWT [26]	−0.18228	0.24974	2009	∞
VMC [22]	−0.18233(3)	0.265	2009	576
DMRG [16]		0.205(15)	2007	~140
FN [17]	−0.17996(1)	0.1625(30)	2006	324
FNE [17]	−0.18062(2)	0.1765(35)	2006	324
SE [21]	−0.18340(13)	0.19(2)	2006	∞
VMC [19]	−0.1773(3)	0.36	2006	108
ED [13]	−0.1842	0.193	2004	36
DMRG [20]	−0.1814		2001	144
GFMC [15]	−0.18193(3)	0.205(10)	1999	144

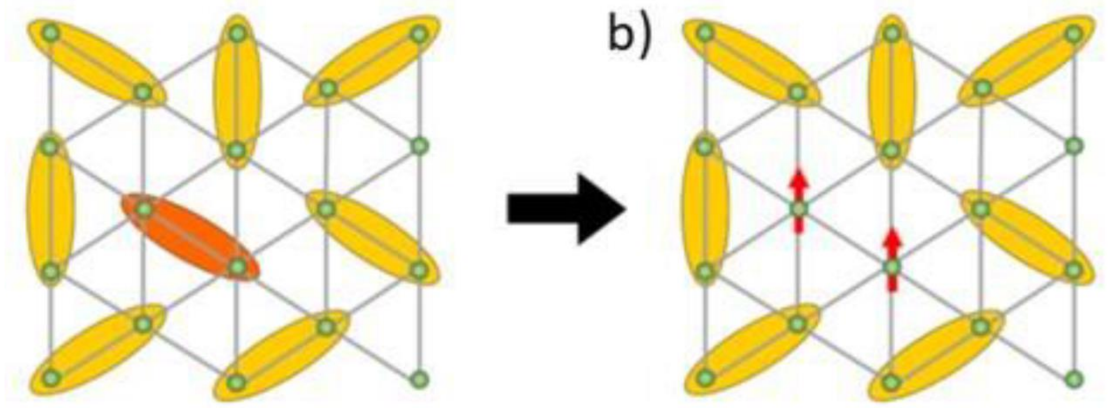
Q. Li *et al.*, PRB **105**, 184418 (2022).



$\Gamma/|K| = 0.1 \qquad J = 0$

From this standpoint the TLHAF remains one of the hottest topics in frustrated quantum magnetism …

Kitaev QSL: fractional spins maximising kinetic energy, i.e. in ***k***-space..

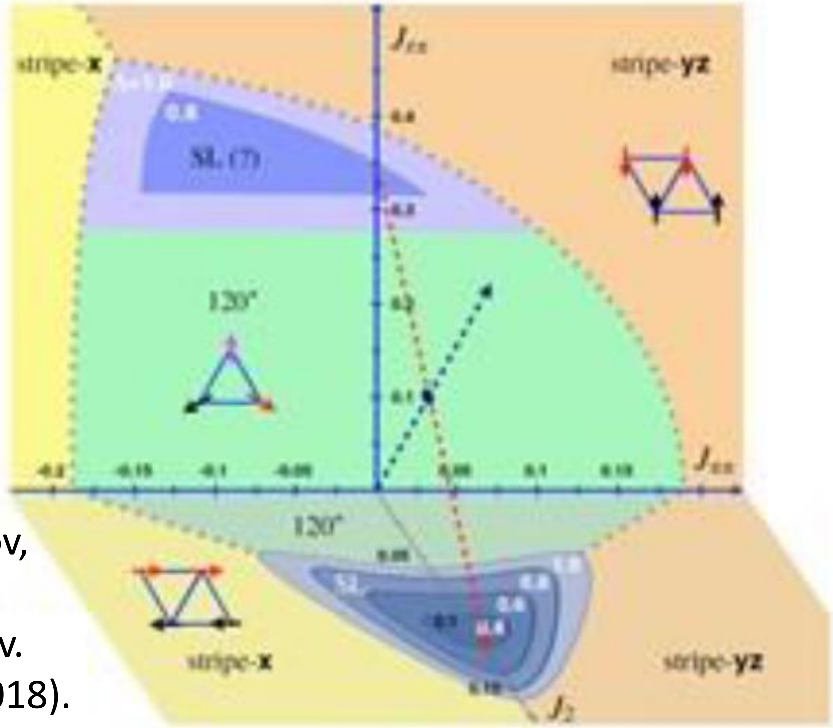


Anderson QSL: local singlets gaining resonance energy.

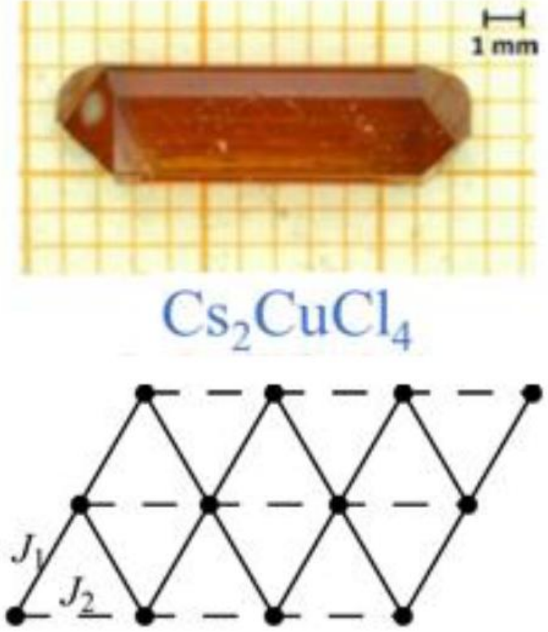
TLAF History

Despite its intrinsic theoretical interest, research on quantum TLAF models has been driven largely by new families of TL materials:

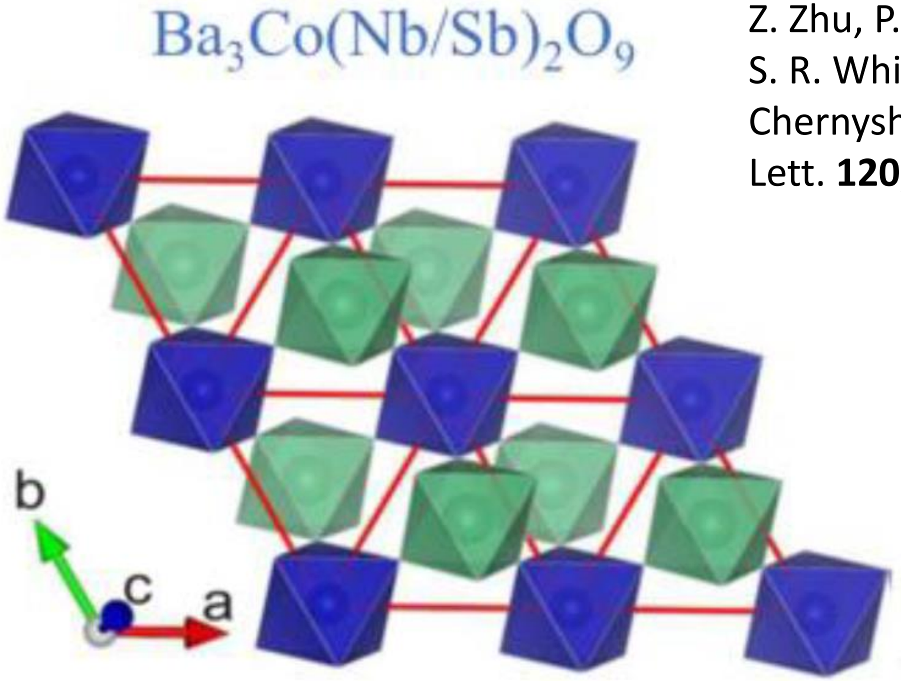
- Cs_2CuCl_4, Cs_2CuBr_4: spatially anisotropic TLHAF.
- $Ba_3CoNb_2O_9$ and $Ba_3CoSb_2O_9$: TLHAF with XXZ spin anisotropy.
- $YbMgGaO_4$, $AYbX_2$ (A = Na, K, Rb, Cs; X = O, S, Se): generalised spin anisotropy.



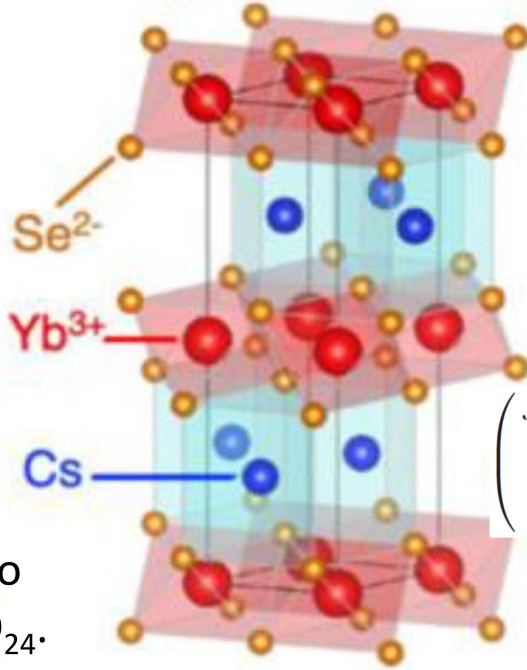
Z. Zhu, P. A. Maksimov, S. R. White and A. L. Chernyshev, Phys. Rev. Lett. **120**, 207203 (2018).



DM interactions in these materials give rise to complex field-induced magnetic phase diagrams.



The Co^{2+} ions in this geometry have only weak XXZ spin anisotropy, which seems to vanish in the ultra-2D limit of $Ba_8CoNb_6O_{24}$.



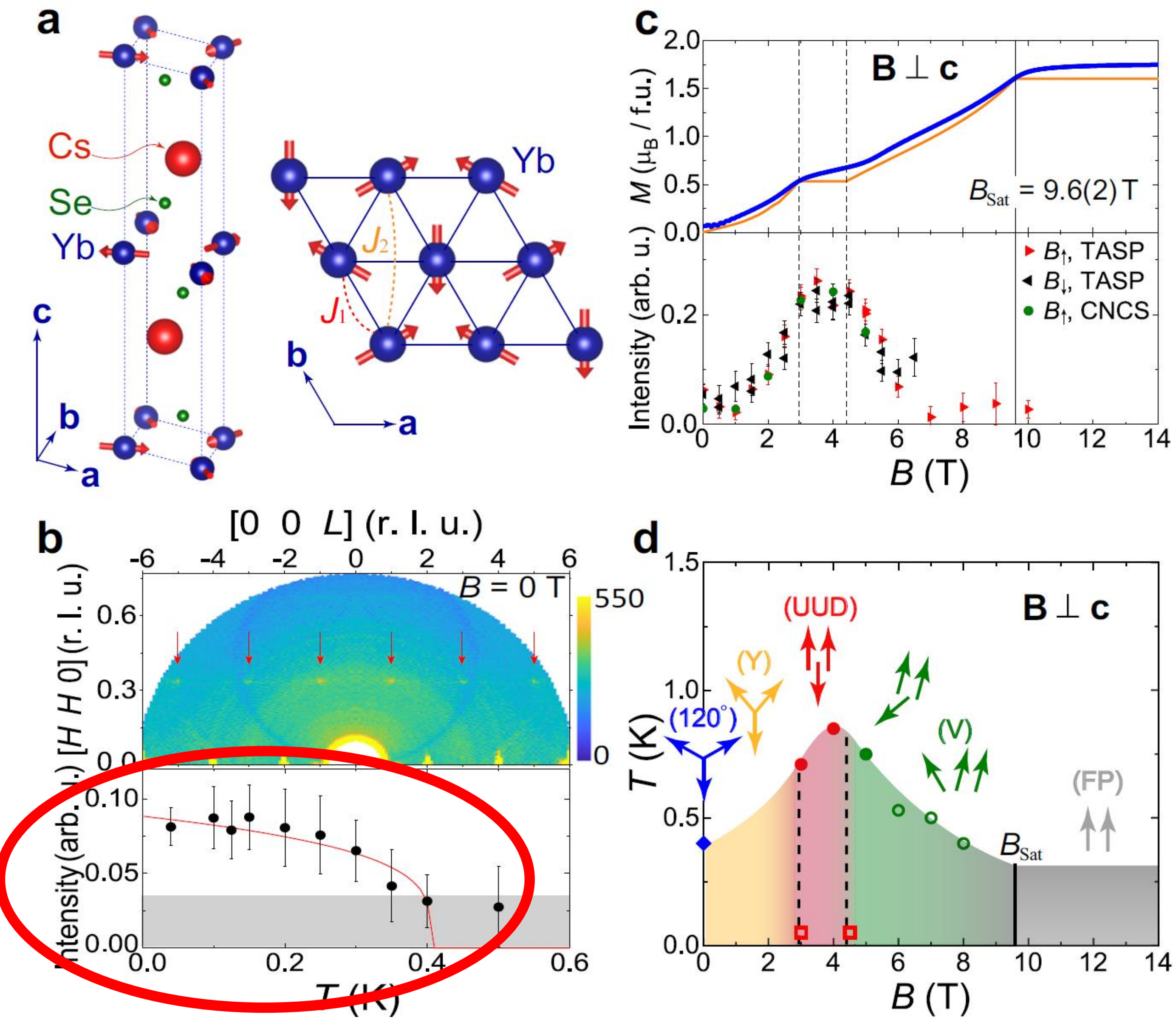
$$\hat{\mathcal{H}}_{ij} = \mathbf{S}_i^T \begin{pmatrix} J_{xx} & J_{xy} & J_{xz} \\ J_{yx} & J_{yy} & J_{yz} \\ J_{zx} & J_{zy} & J_{zz} \end{pmatrix} \mathbf{S}_j$$

$$\begin{pmatrix} J + 2J_{\pm\pm}\tilde{c}_\alpha & -2J_{\pm\pm}\tilde{s}_\alpha & -J_{z\pm}\tilde{s}_\alpha \\ -2J_{\pm\pm}\tilde{s}_\alpha & J - 2J_{\pm\pm}\tilde{c}_\alpha & J_{z\pm}\tilde{c}_\alpha \\ -J_{z\pm}\tilde{s}_\alpha & J_{z\pm}\tilde{c}_\alpha & \Delta J \end{pmatrix}$$

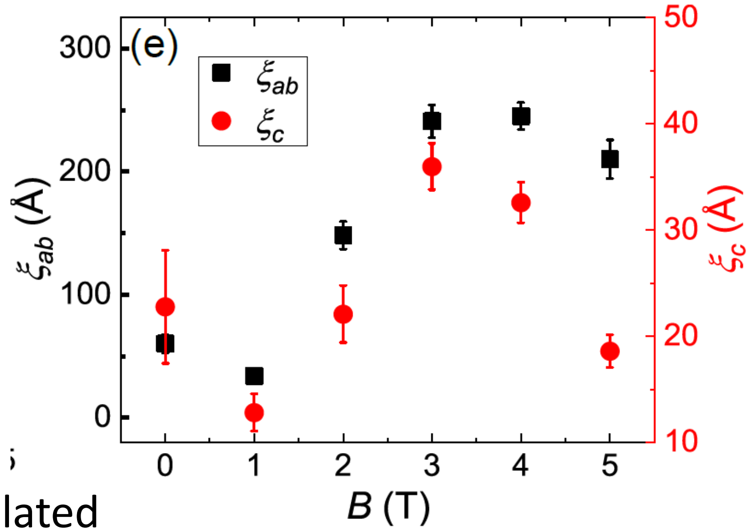
Yb^{3+}: complex pseudospin-1/2

CsYbSe$_2$: near-ideal TLHAF

T. Xie *et al.*, Complete spectral response of the spin-1/2 triangular-lattice antiferromagnet CsYbSe$_2$, npj Quantum Materials **8**, 48 (2023).

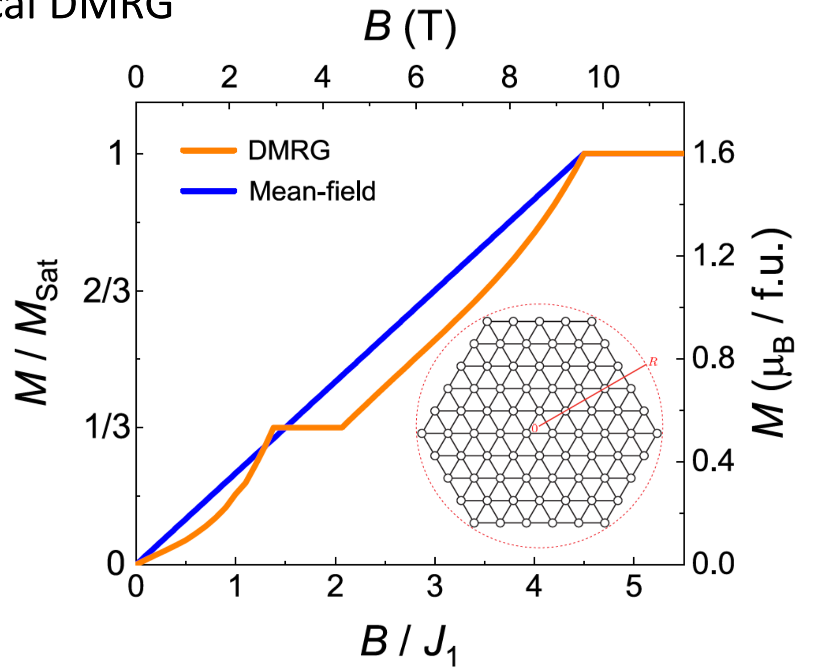


Real 120^o magnetic order at zero field, but not truly long-ranged.



Magnetisation calculated by grand canonical DMRG method, using J parameters extracted from the spin-wave dispersion in the saturated regime (next).

Ideal agreement with standard phase diagram.



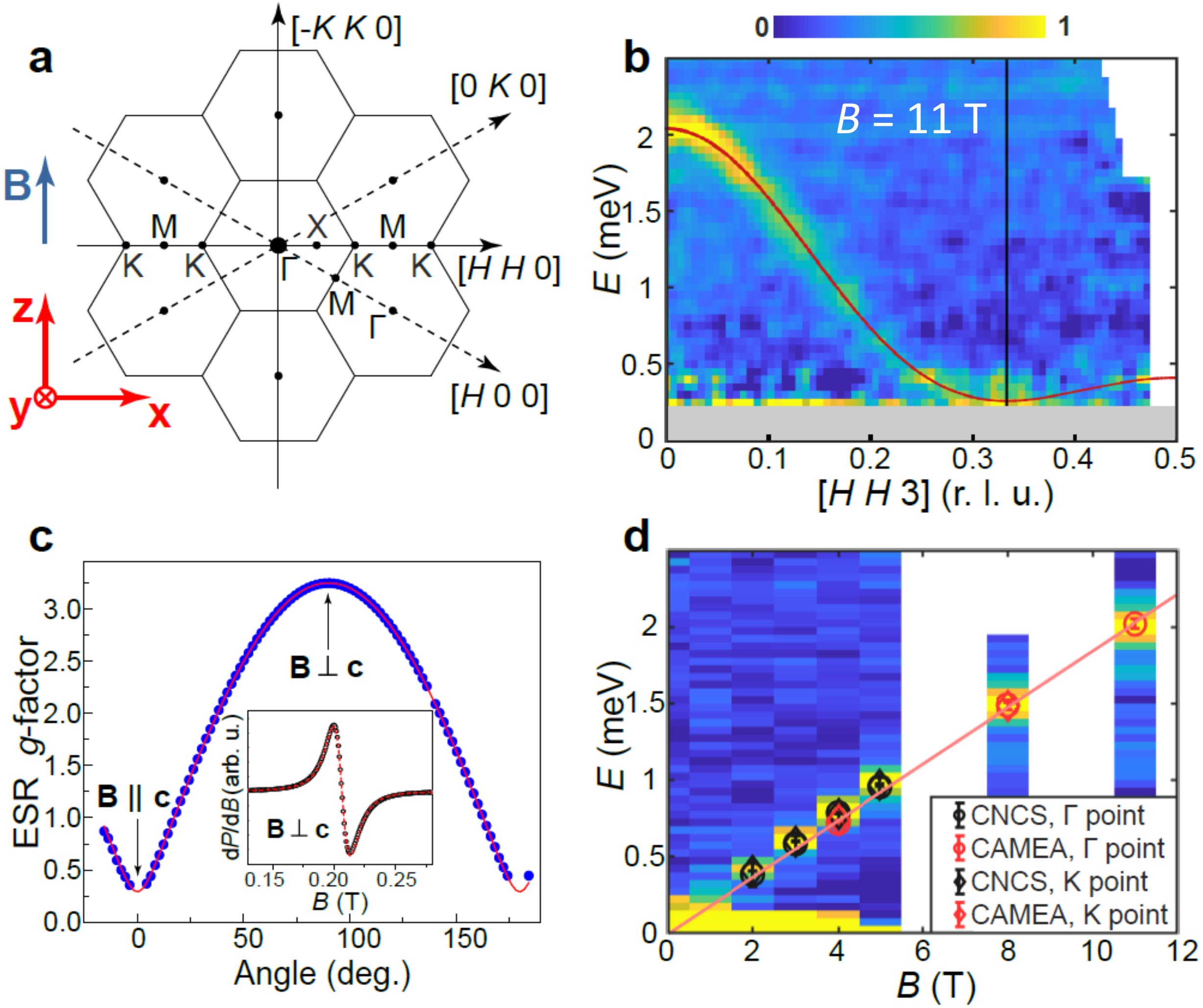
INS: Magnetic Hamiltonian

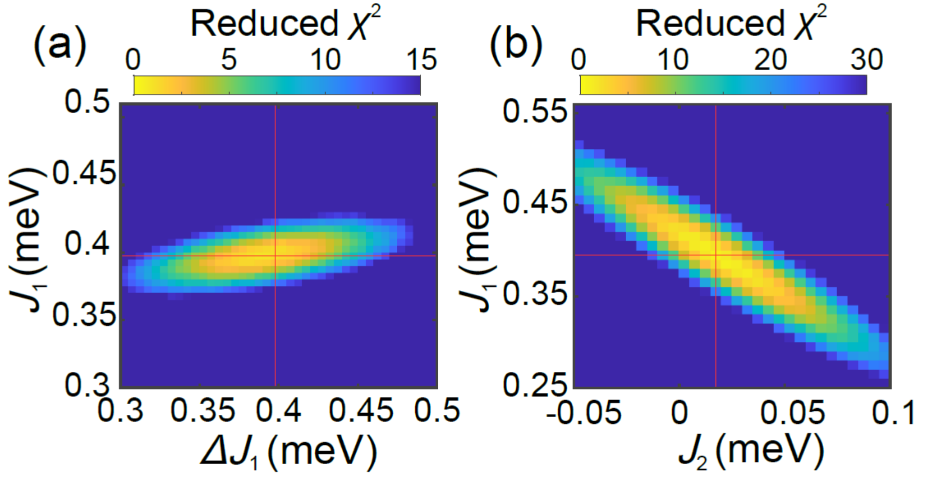
The sample: 200 coaligned crystallites, 1° mosaic, total mass 0.5 g.

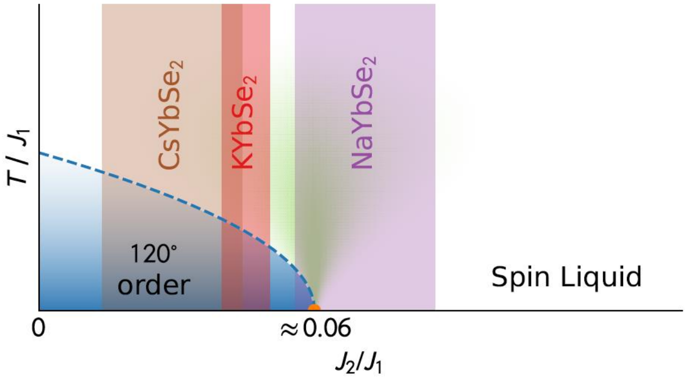
The breakthrough: apply a field above saturation, then the excitation is a conventional spin wave, allowing an accurate fit of the Hamiltonian.

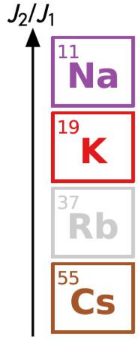
$$\mathcal{H} = J_1 \sum_{\langle i,j \rangle} (S_i^x S_j^x + S_i^z S_j^z + \Delta S_i^y S_j^y) + J_2 \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j - \mu_B g_{ab} B \sum_i S_i^z$$

- $\Delta = 1$: Heisenberg
- $J_2/J_1 = 0.03$



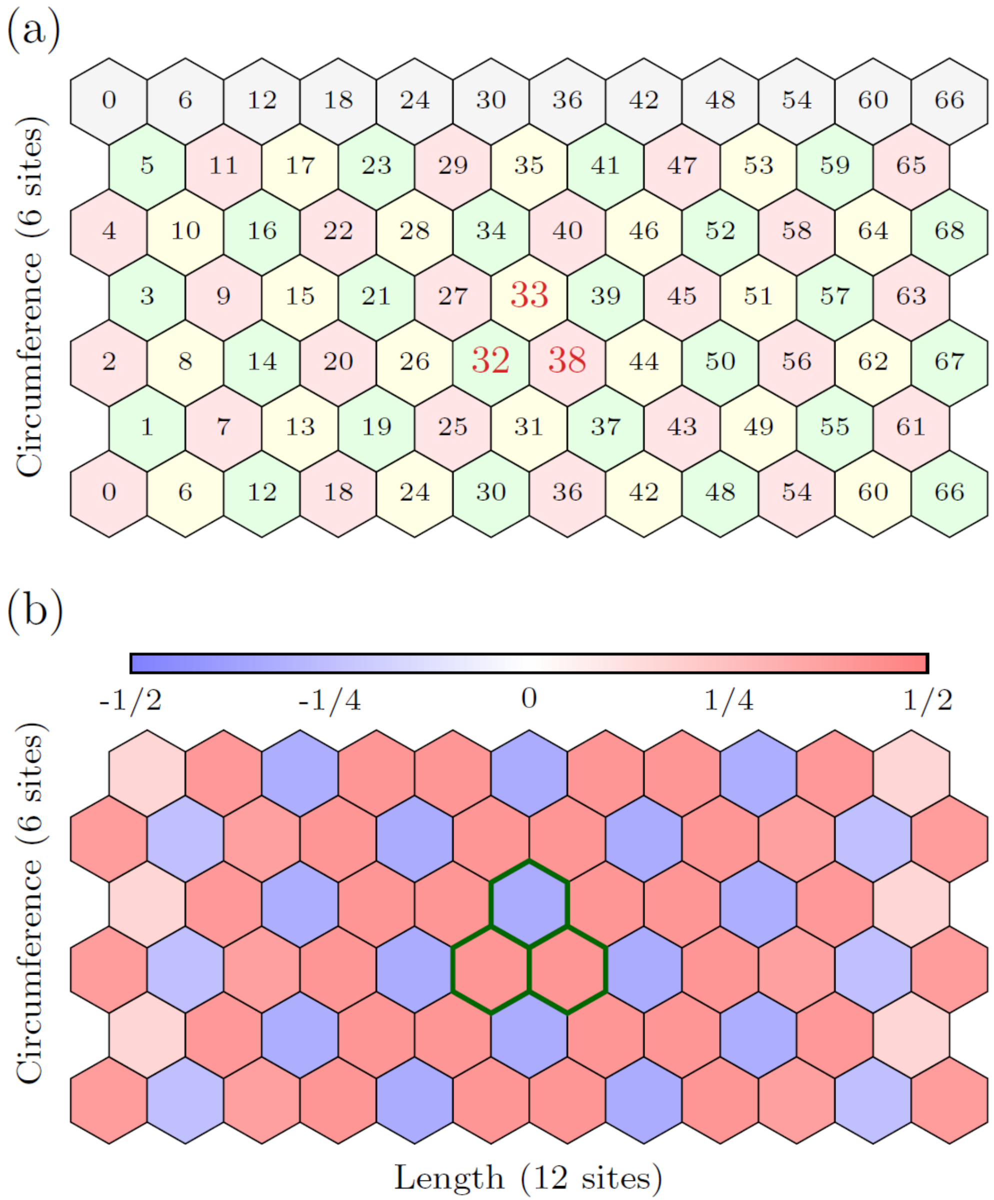






J_2/J_1 probably explains the differences between the $AYbSe_2$ materials.

Cylinder MPS



Cylinder MPS applied to the TLHAF with J_2/J_1 = 0.03: we compute

$$C_{\mathbf{r}}^{\alpha\beta}(\mathbf{x},t) = \langle \hat{\mathcal{S}}_{\mathbf{r}+\mathbf{x}}^{\alpha}(t)\hat{\mathcal{S}}_{\mathbf{r}}^{\beta}(0)\rangle$$
$$= \langle 0|\hat{S}_{\mathbf{r}+\mathbf{x}}^{\alpha}\, e^{-i(H-E_0)t}\, \hat{S}_{\mathbf{r}}^{\beta}|0\rangle - \langle 0|\hat{S}_{\mathbf{r}+\mathbf{x}}^{\alpha}|0\rangle\, \langle 0|\hat{S}_{\mathbf{r}}^{\beta}|0\rangle$$

Preserving U(1) symmetry means that $\alpha\beta \in \{zz, +-, -+\}$

To deal with the finite cylinder length and circumference it is conventional to apply a Gaussian filter to the correlation function, primarily in order to remove finite-size artifacts but also to mimic the experimental resolution.

$$C_{\mathbf{r}}^{\alpha\beta}(\mathbf{x},t) \to e^{-\sigma_t t^2} e^{-\sigma_x (\mathbf{e}_L\cdot\mathbf{x})^2}\, C_{\mathbf{r}}^{\alpha\beta}(\mathbf{x},t)$$

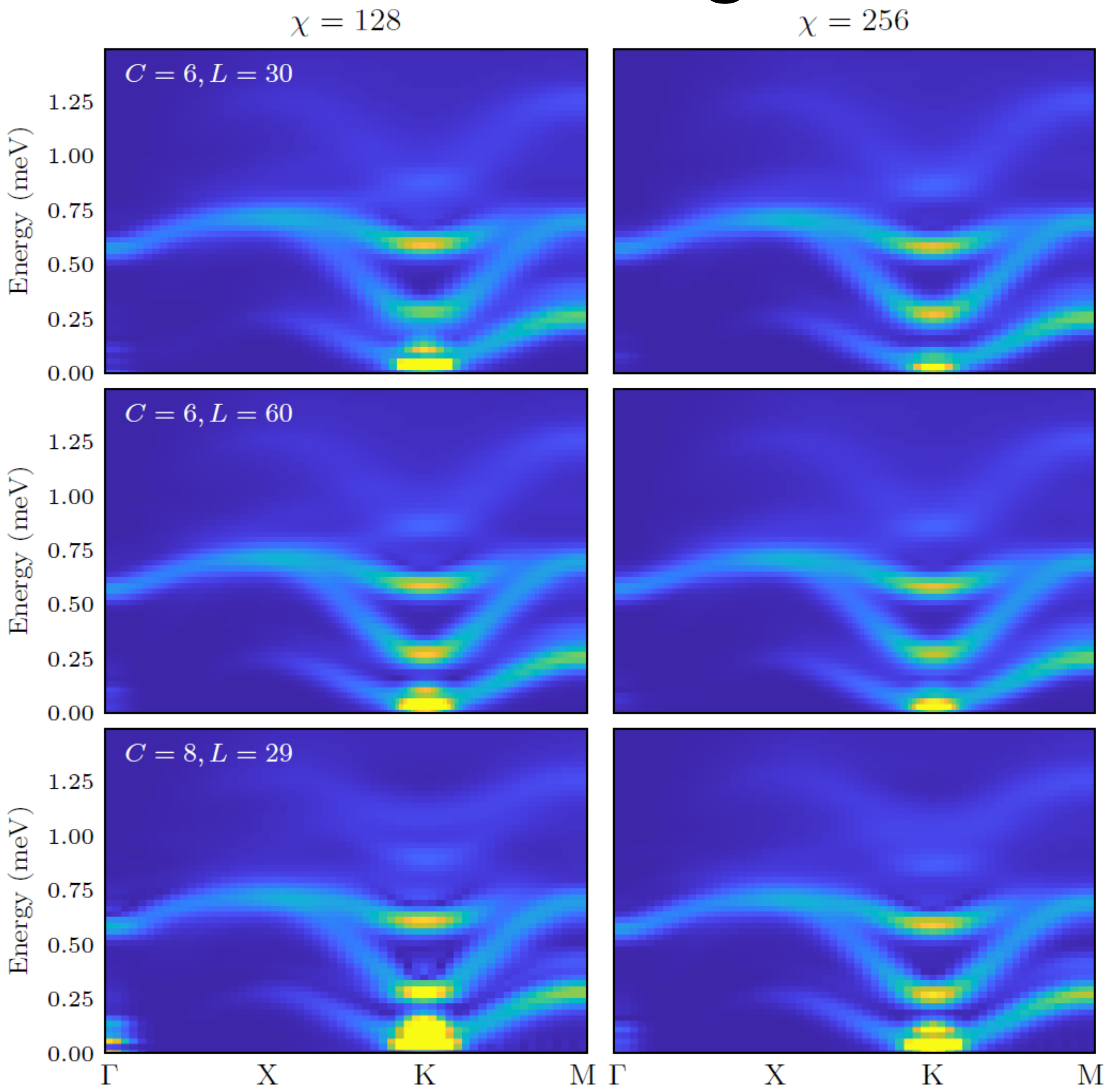
$$\sigma_t = 0.005 J_1^2 \qquad \sigma_x = 0.02/a^2$$

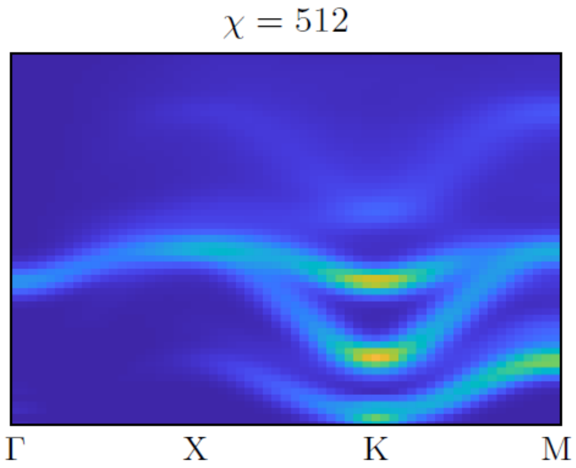
Because it's pure Heisenberg: $C_{\mathbf{r}}^{\alpha\beta}(\mathbf{x},-t) = \overline{C_{\mathbf{r}}^{\alpha\beta}(\mathbf{x},t)}$

Sum over the 3 centre sites to restore translational symmetry and the perform the double Fourier transformation:

$$S_{\alpha\beta}(\mathbf{Q},\omega) = \frac{2}{3}\sum_{\mathbf{r}}\int_0^{\infty} \mathrm{d}t \sum_{\mathbf{x}} e^{-i\,\mathbf{x}\cdot\mathbf{Q}} \left[\cos(\omega t)\,\mathrm{Re}\, C_{\mathbf{r}}^{\alpha\beta}(\mathbf{x},t) - \sin(\omega t)\,\mathrm{Im}\, C_{\mathbf{r}}^{\alpha\beta}(\mathbf{x},t)\right]$$

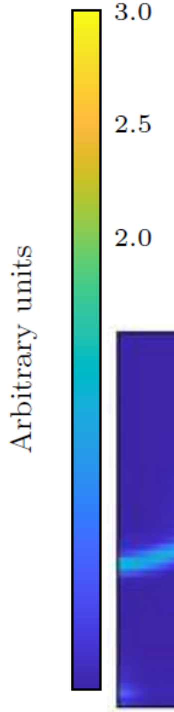
MPS Benchmarking and Gaussian Filters





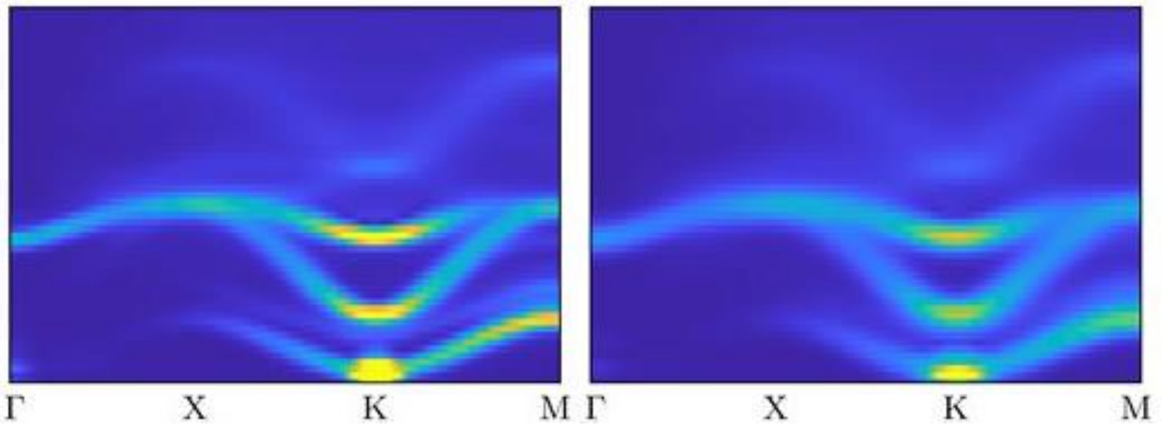
We computed with

- C up to 8
- L up to 60
- χ up to 1024



However, the results are already converged at much less demanding values because σ_t and σ_x are big …

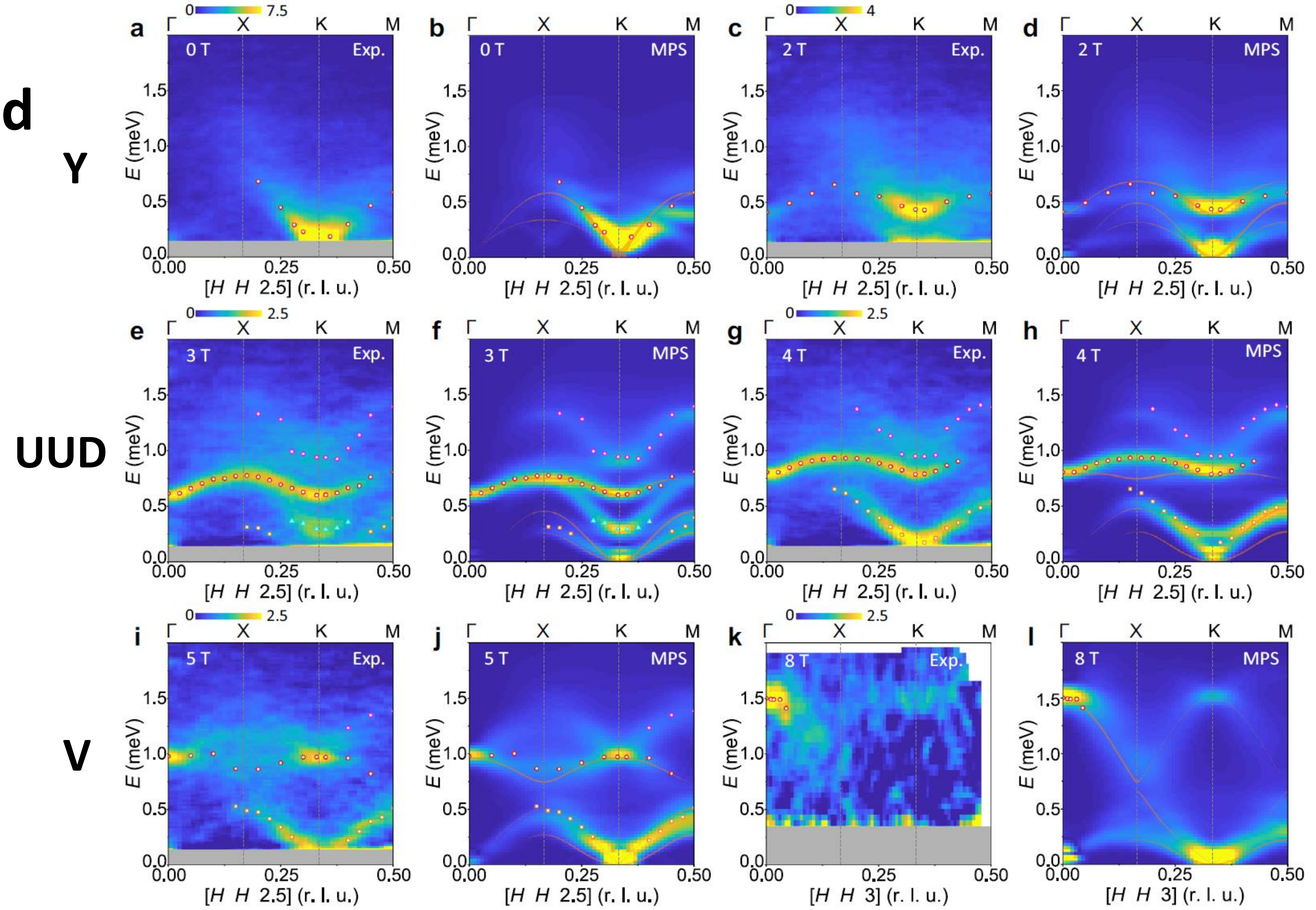
If we really tried to optimise the MPS calculation then both could be 2.5 times smaller (1.5 times better resolution) before artifacts begin to appear.



Complete Field-Induced Spectral Response

Good ΔS = 1 branches only in the UUD phase; otherwise the response is sharp only at the Γ and K points. Every other feature looks like a **continuum**, i.e. no well-defined spin-wave branches even at low energies.

Certainly linear spin-wave theory (shown by the orange lines) does not work well at any field (although this is not a surprise).



Quantitative Fit

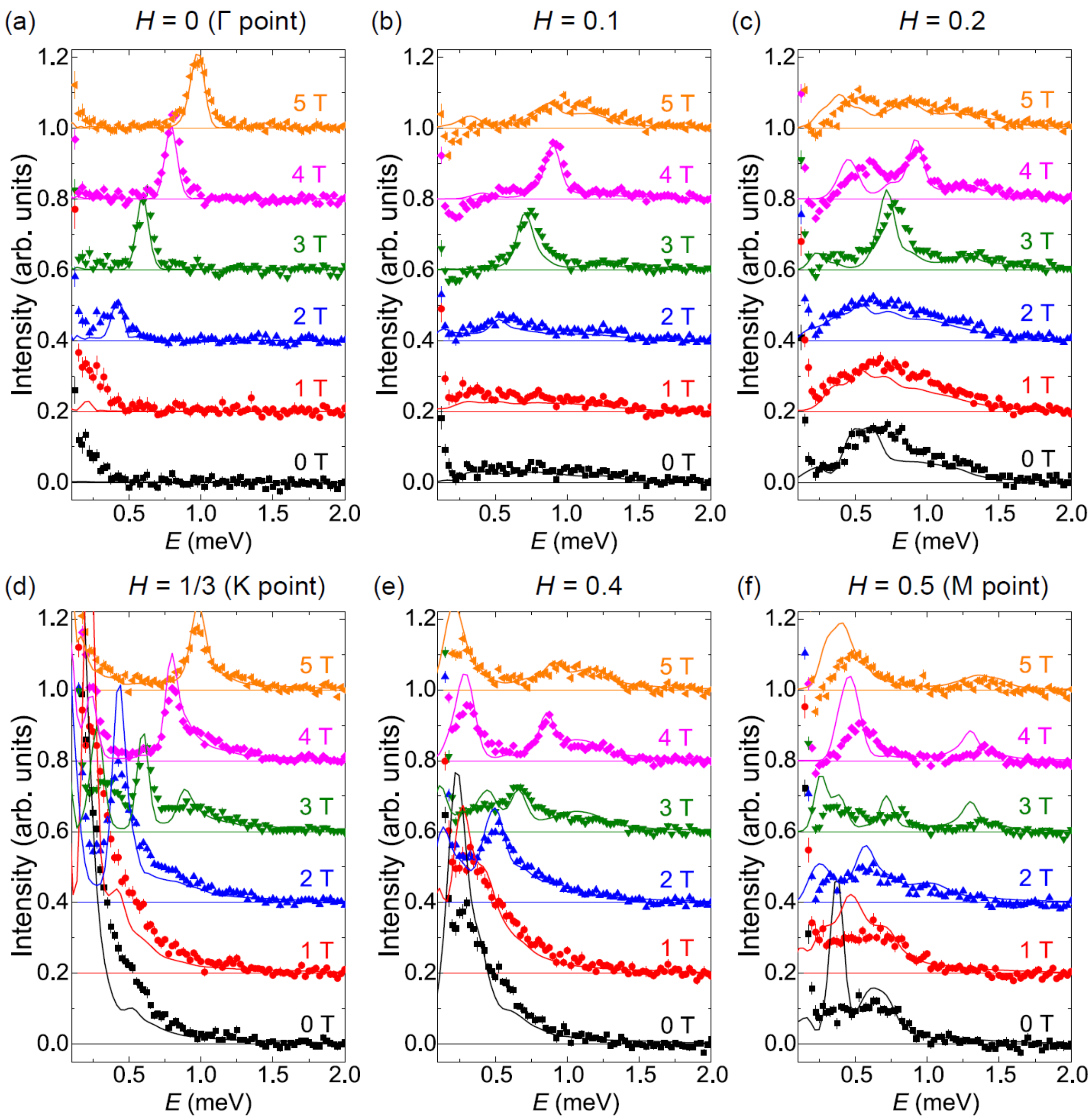
Beyond the pretty colours, the MPS calculations provide, with one scale factor, a **quantitatively accurate account** of the measured spectral functions at all fields and wave-vectors other than very low $|Q|$ and low H.

Thus we can claim to have provided a mostly **unbiased quantitative benchmark** by which all biased analytical approaches can be judged.

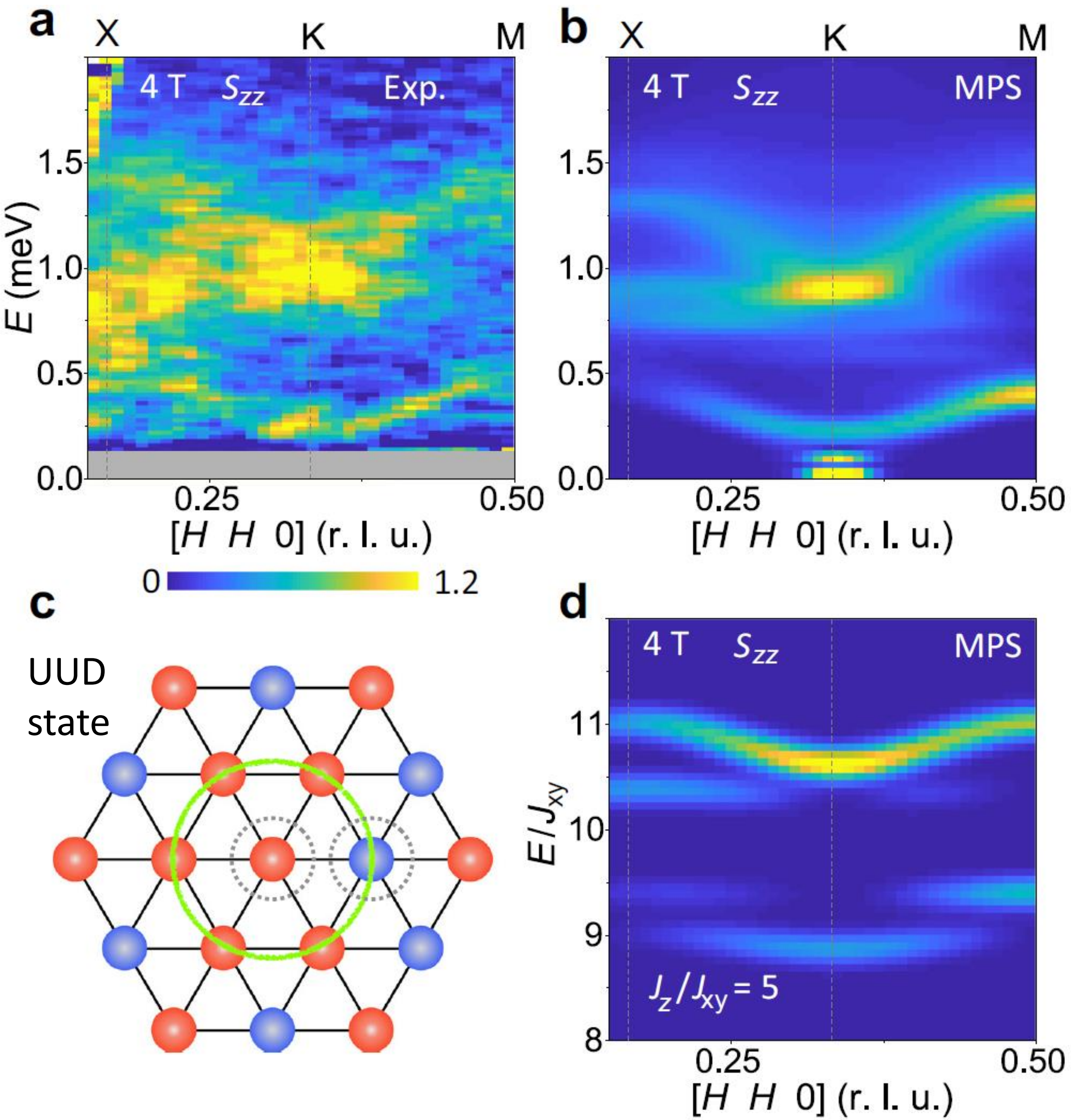
One analytical approach that is successful in the UUD phase is nonlinear spin-wave theory.

Some new terminology:
Expressibility: ability of a method to reproduce the physics of the system;
Interpretability: ability of a method to give insight into the underlying physics of the system.
An unbiased numerical method allows us to divide up the problem of understanding into these 2 steps, but by definition does not offer a lot of interpretability.

Still we can now test nonlinear and interacting spin waves, multimagnon bound states and parton methods (Schwinger bosons, slave fermions) much more accurately.

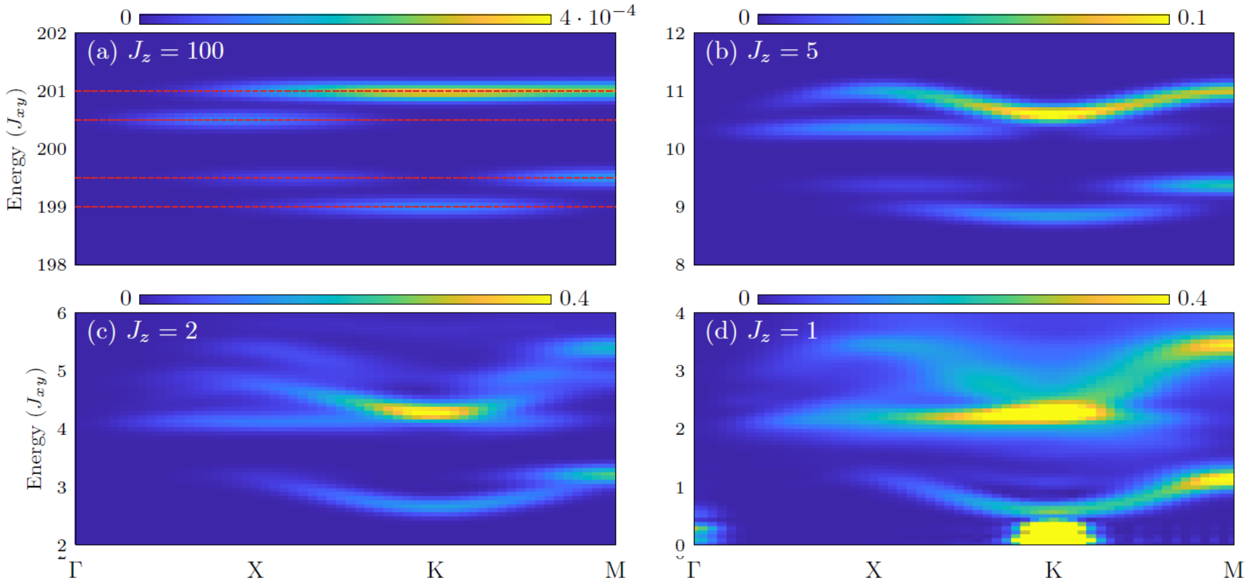


Longitudinal Response



In experiment, the highly anisotropic *g*-factor makes it possible to separate out the longitudinal response. S_{zz} is readily computed by MPS.

If we do attempt a biased analytical interpretation, we can build a model of single quantum spin-flips that is exact in the Ising limit: here two spin-flips cost 3*J* except on neighbouring sites, where they cost 2*J*. This binding effect is preserved all the way back to the Heisenberg limit, where it accounts for the sharp low-lying branch and the broad, dispersive continuum.



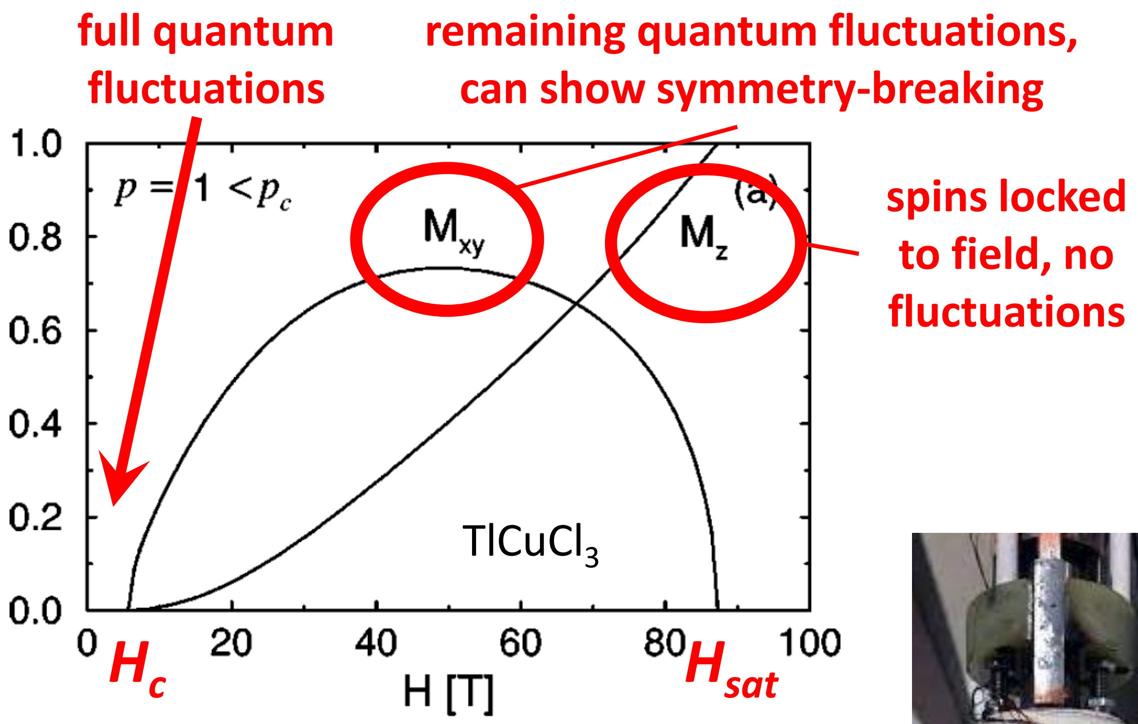
Summary III

I only talked about the Heisenberg model …

Here the field can be regarded as the ideal control knob to "switch off quantum fluctuations."

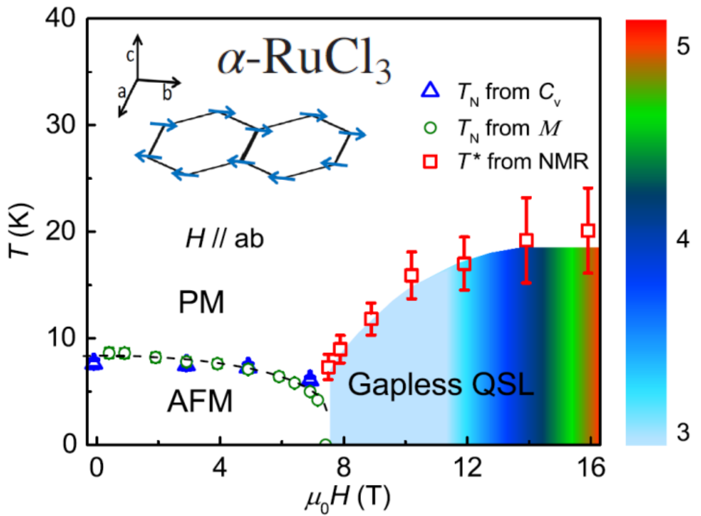
We have found rich physics even in this case

- **novel quantum spin nematic phase**
- **novel nonuniversal quantum critical scaling**
- **novel field-induced excitation continua**

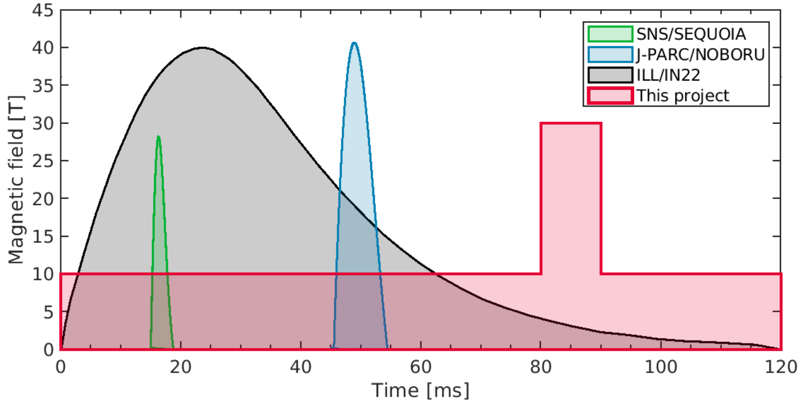


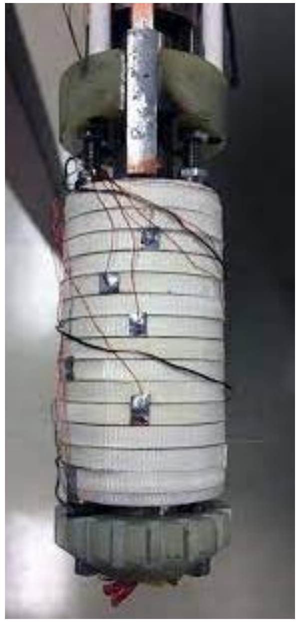
Where Next ?

Experiment is moving towards pulsed fields: ready access to 50 T with minicoils and "smart power electronics" in the LC circuit.



Theory: in many systems with more complex Hamiltonians, applying the field can destabilise magnetism and induce more complex phases – as in the proximate Kitaev materials.





Conclusion: there is still a big future for experimental quantum magnetism in high applied fields; for theorists, all the things we never thought we could do might be possible after all.