

Condensation of bosonic pairs in quantum magnets

F. Mila

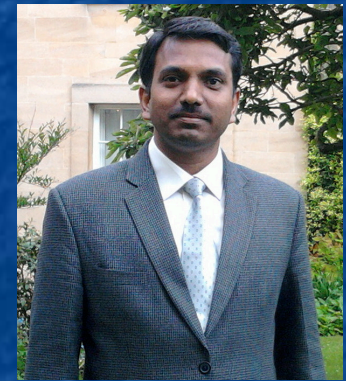
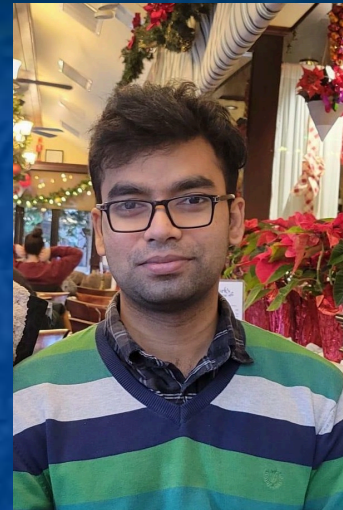
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Theorists

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Experimentalists

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Henrik Rønnow (Lausanne)

and many more...

Scope

- Spin nematics
- Pair condensation in spin-1/2 systems
 - Below saturation: bound states of magnons
 - From gapped phase: bound states of triplons
- Bosonic language: condensation of Cooper pairs
- Experimental evidence: triplet gap at transition
- Shastry-Sutherland compound $\text{SrCu}_2(\text{BO}_3)_2$
 - Inelastic neutron scattering up to 26 T
 - Dynamical structure factor from t-dependent DMRG
- Conclusions

Spin nematics

- Broken $SU(2)$ symmetry without magnetic order
- Spin-1 models with biquadratic interactions
 - Blume and Hsieh, 1969
- General classification
 - Andreev and Grishchuk, 1984
- Experimental realization in spin-1 systems?
 - $NiGa_2S_4$: spin-1 on triangular lattice Nakatsuji et al, 2005
 - Theory: Ferro- and AF- quadrupolar phases
 - Arikawa-Tsunetsugu, 2006; Läuchli, FM, Penc, 2006

Pair condensation in $S=1/2$ systems

- Spin nematic below saturation if FM interactions
- Bound states of magnons condense first

Shannon, Momoi, Sindzingre, 2006

Zhitomirsky and Tsunetsugu, 2010

- NMR signature in LiCuVO_4
 - No shift below saturation
 - No magnetic order

Orlova et al, 2017

Pair condensation in gapped AF

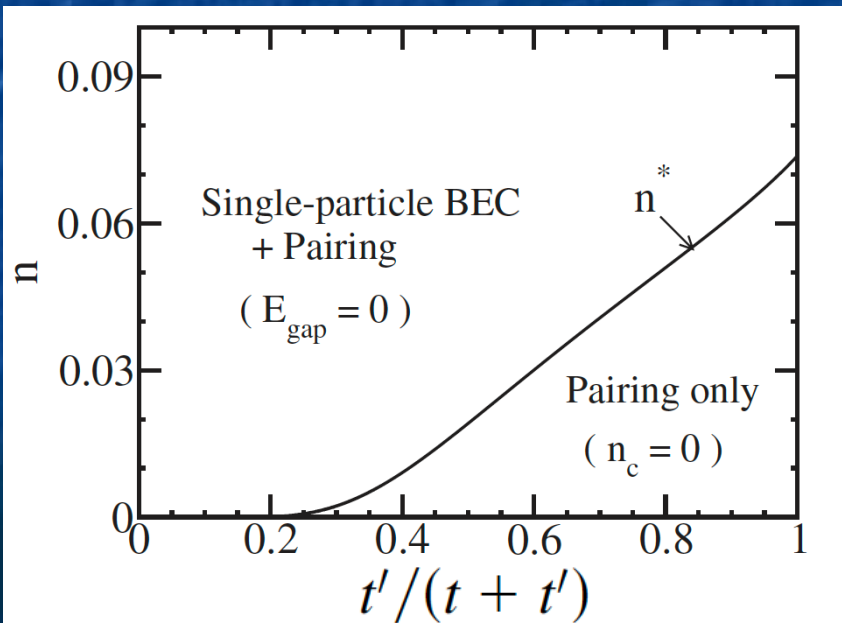
- Gapped antiferromagnets
 - Usually, **gap closing = BEC of triplets**
- **Always? No!**
 - If there is a **spin-2 bound state** below the two-triplet continuum, it will condense first
 - Non-zero magnetization, but **no BEC of triplets**

Absence of Single-Particle Bose-Einstein Condensation at Low Densities for Bosons with Correlated Hopping

Rachel Bendjama, Brijesh Kumar,* and Frédéric Mila†

Mean-field theory

$$H = -t \sum_{\mathbf{r}} \sum_{\delta=\pm x, \pm y} b_{\mathbf{r}+\delta}^\dagger b_{\mathbf{r}} - \mu \sum_{\mathbf{r}} n_{\mathbf{r}} \\ - t' \sum_{\mathbf{r}} \sum_{\delta=\pm x} \sum_{\delta'=\pm y} n_{\mathbf{r}} \{b_{\mathbf{r}+\delta}^\dagger b_{\mathbf{r}+\delta'} + \text{H.c.}\},$$



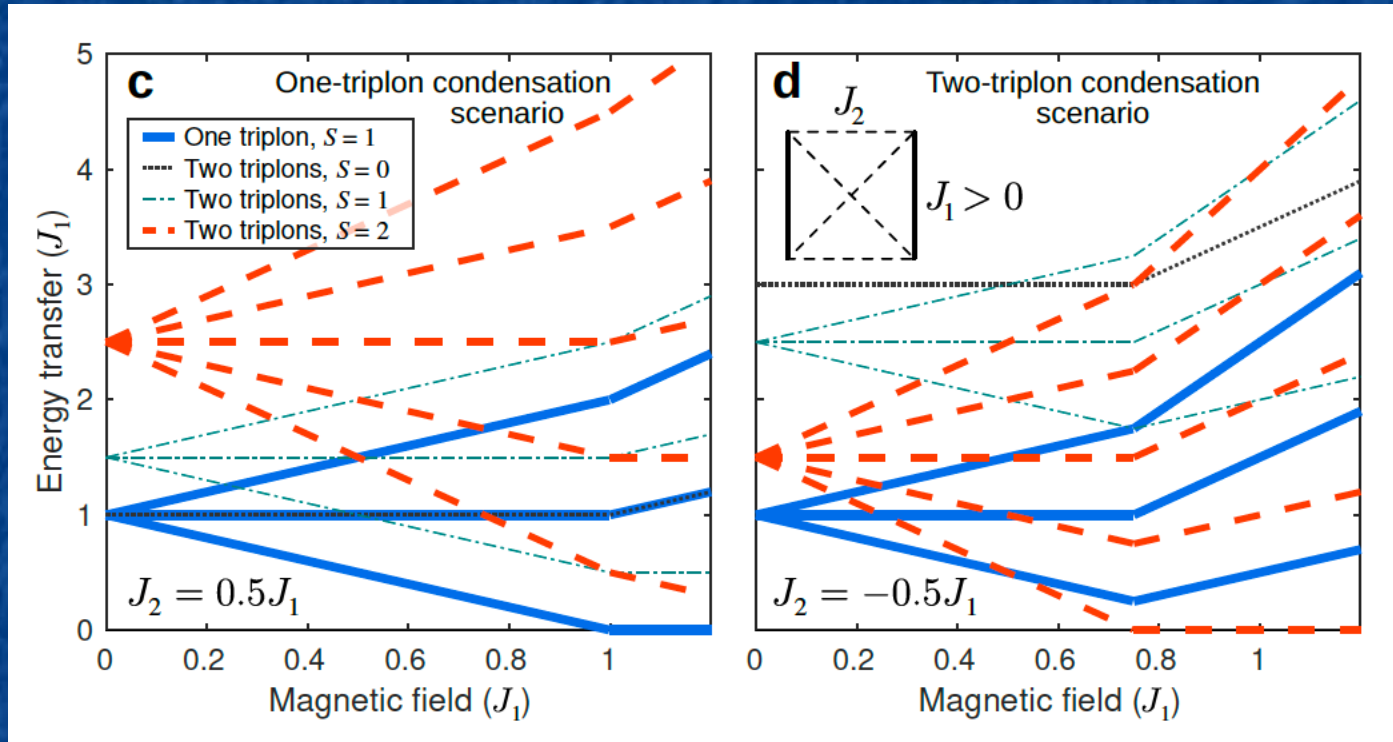
Pair condensation without
single particle condensation

(large enough correlated
hopping and small density)

Experimental signature?

- Follow spin-2 bound state up to condensation
→ tricky due to selection rules
- Follow the first-triplet excitation
→ gap does not close at the transition
(as the single particle gap in BCS theory)
→ kink at the transition

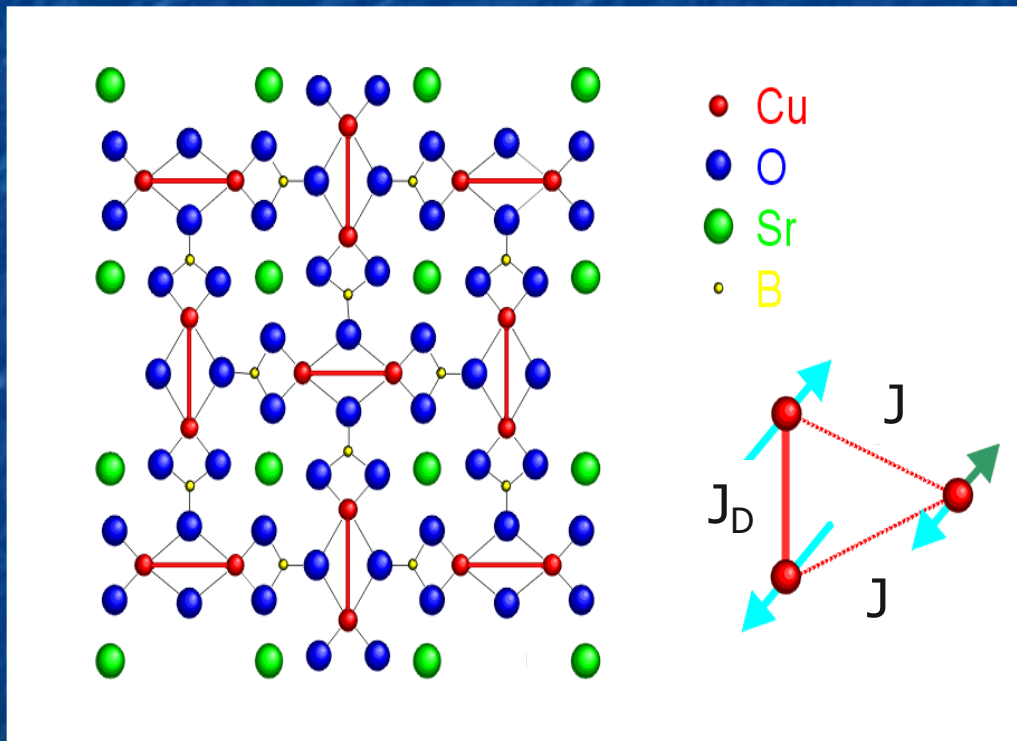
Toy model



Fogh, Nayak, ... FM, Rønnow, 2023

SrCu₂(BO₃)₂

Smith and Keszler, JSSC 1991



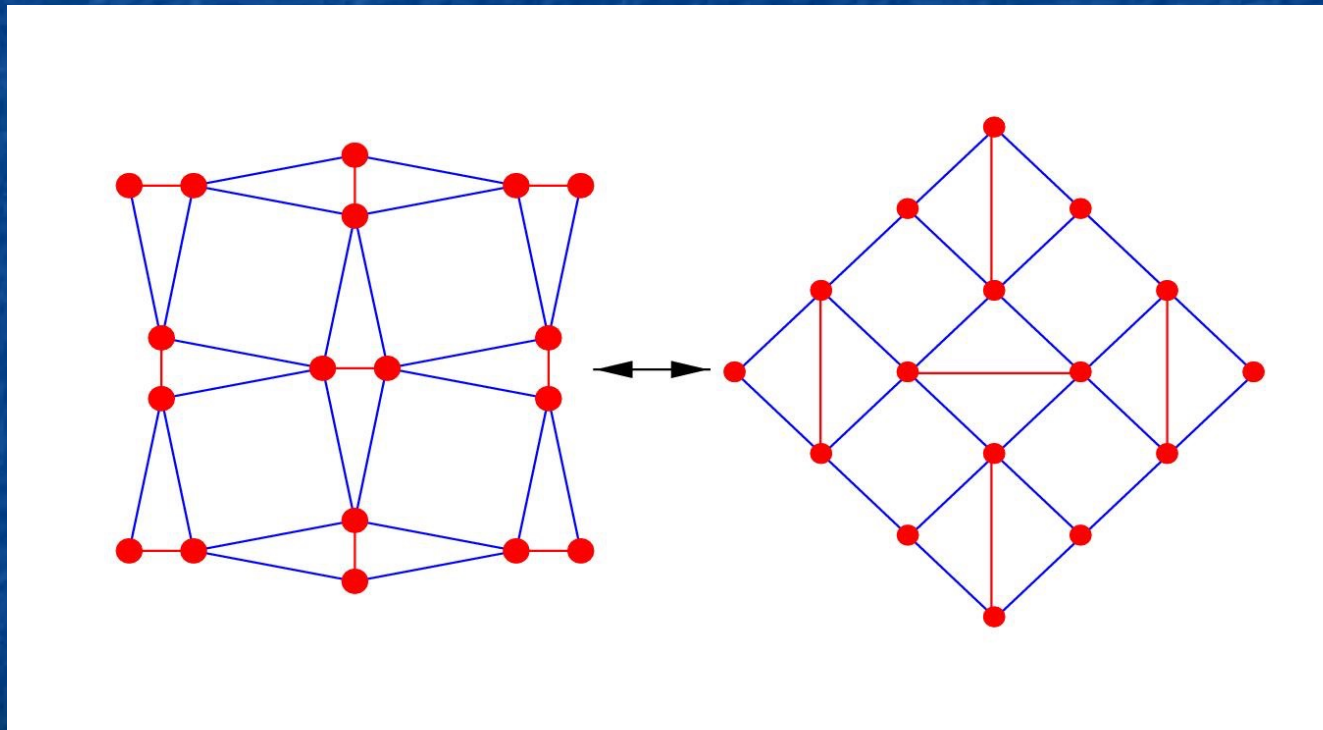
Cu²⁺ -> Spin 1/2

$J_D \approx 85 \text{ K}$

$J/J_D \approx 0.63$

Famous for its magnetization plateaus

From orthogonal dimer to Shastry-Sutherland model

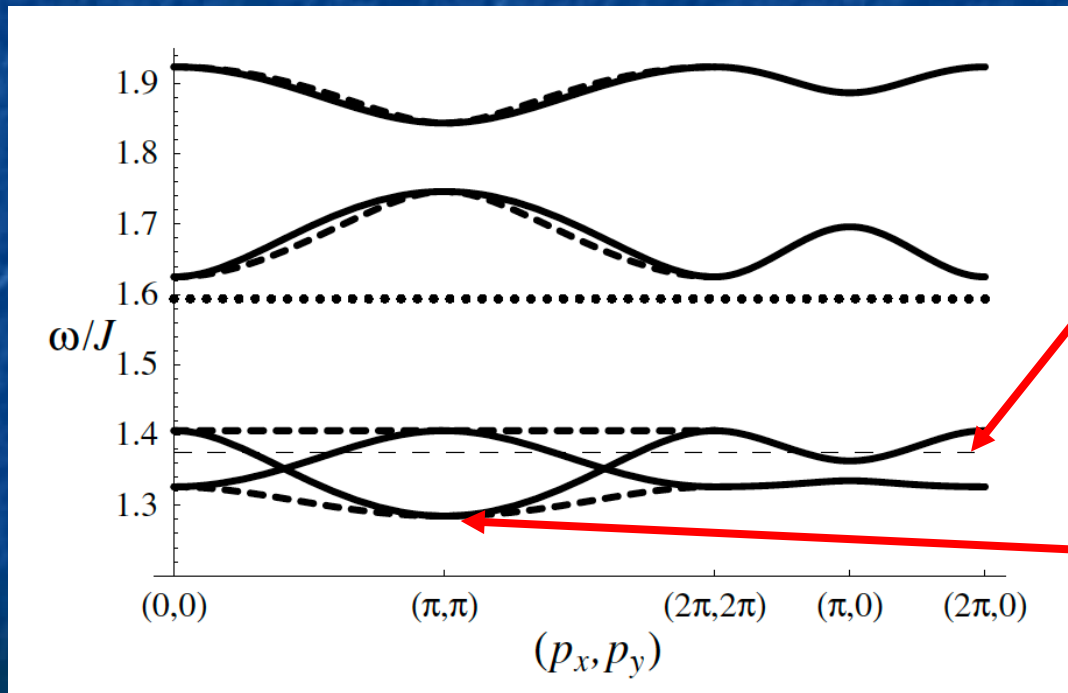


Shastry and Sutherland, 1981

Low-Lying Magnetic Excitation of the Shastry-Sutherland Model

K. Totsuka,^{1,*} S. Miyahara,² and K. Ueda²

3rd order perturbation theory in J/J_D



Two-triplet continuum

Spin-2 bound state

Pair condensation in $\text{SrCu}_2(\text{BO}_3)_2$

- Inelastic neutron scattering

- need data above 23 T

- only possible a few years ago in Berlin and for a very limited period of time

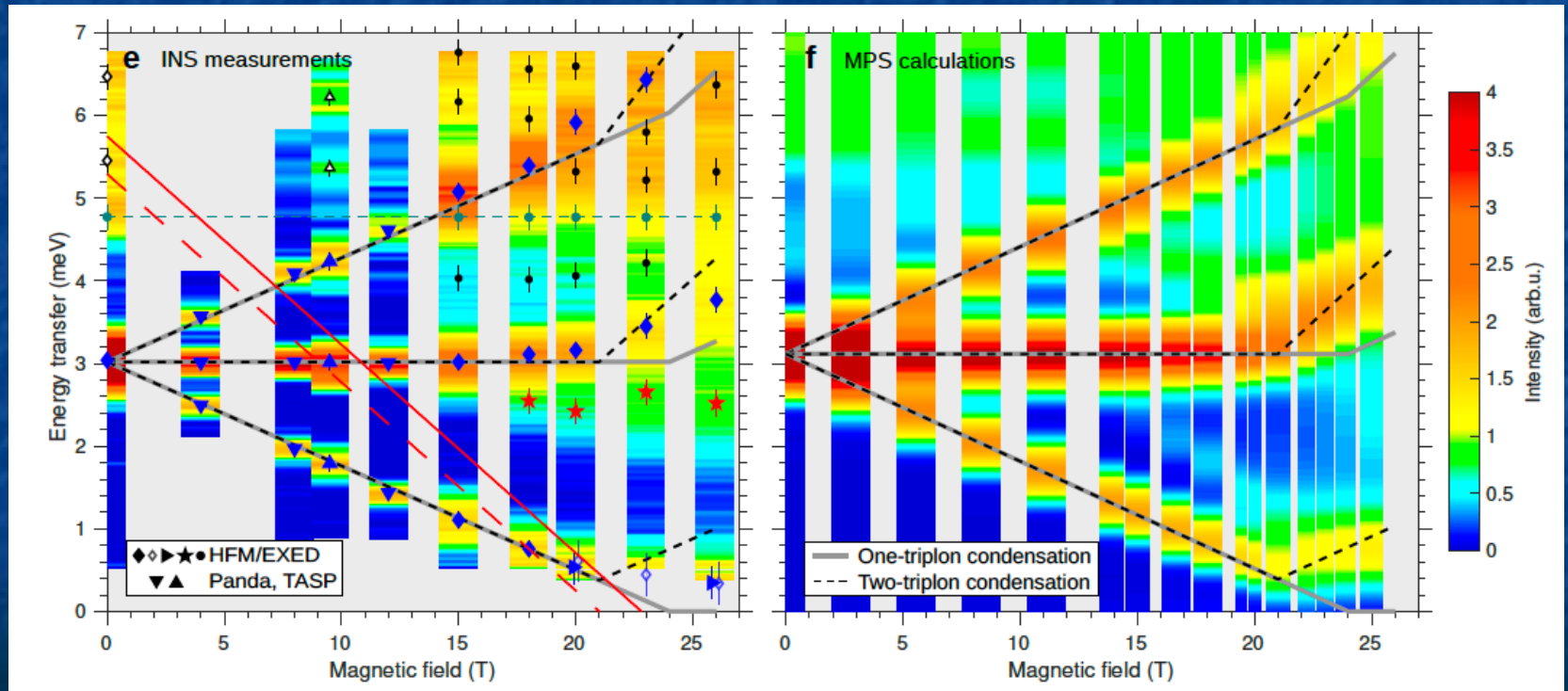
- Time-dependent DMRG on cylinders

- good approximation to dynamical structure factor of 2D systems

Field-induced bound-state condensation and spin-nematic phase in $\text{SrCu}_2(\text{BO}_3)_2$ revealed by neutron scattering up to 25.9 T

Ellen Fogh,^{1,*} Mithilesh Nayak,^{2,*} Oleksandr Prokhnenko,³ Maciej Bartkowiak,^{3,4} Koji Munakata,⁵ Jian-Rui Soh,¹ Alexandra A. Turrini,^{6,1} Mohamed E. Zayed,⁷ Ekaterina Pomjakushina,⁸ Hiroshi Kageyama,⁹ Hiroyuki Nojiri,¹⁰ Kazuhisa Kakurai,⁵ Bruce Normand,^{1,11} Frédéric Mila,² and Henrik M. Rønnow¹

Nature Communications, in press



Neutron scattering

DMRG

Discussion I

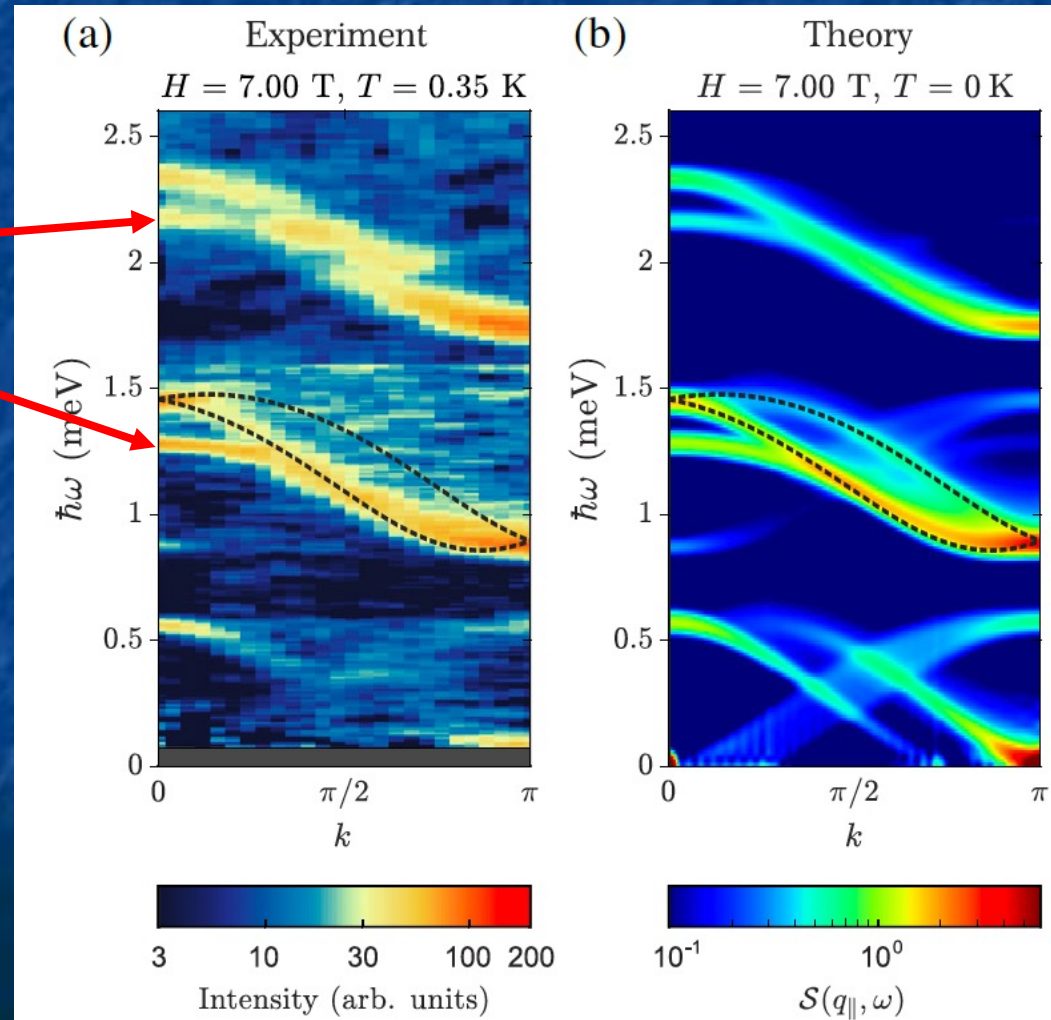
- **Momentum integrated results**
 - Better statistics
 - Small dispersion, so not a major problem
- **DMRG**
 - Width 4 for most results, some benchmarks at width 6
 - No adjustable parameters – all taken from previous experiments, including DM interactions
- **Minimum of triplet branch due to DM interactions?**
 - The minimum should occur at 24 T, where the triplet gap would close, not at 21 T

Discussion II

- Red line: spin-2 mode detected in ESR Nojiri et al , 2003
 - Would cross the singlet too late
 - Dashed-red line: our guess for the lowest spin-2 bound state
 - Not seen probably because of A_2 symmetry
- Wang and Batista, 2018
- Three-triplet excitations
 - Expected when exciting a triplet in a sea of 2-triplet bound states
 - Cannot be resolved with current accuracy

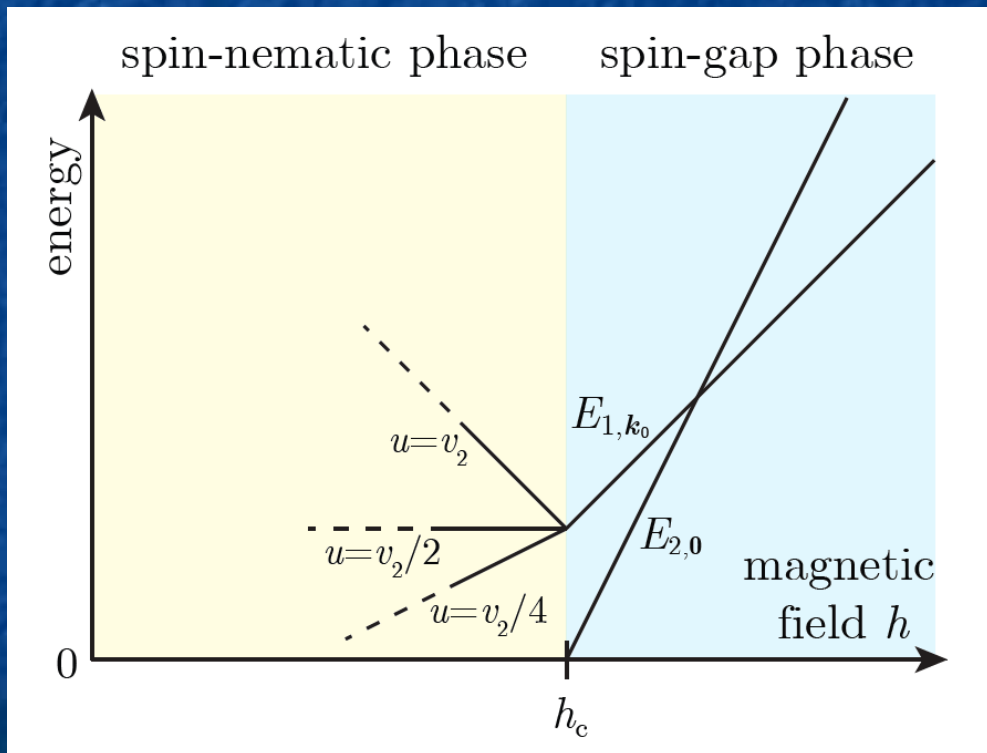
Magnetic-Field-Induced Bound States in Spin- $\frac{1}{2}$ LaddersMithilesh Nayak,^{1,*} Dominic Blosser^{2,†}, Andrey Zheludev,² and Frédéric Mila¹

2-triplet bound states
between a condensed triplet
and a triplet created by a
neutron



Kink of triplet branch

Condensation of bound state of magnons below saturation



u, v_2 : interaction parameters

Momoi, arXiv:2308.12569

Conclusions

- Spin nematics

- Growing evidence in various systems

- $\text{SrCu}_2(\text{BO}_3)_2$

- First example when closing the gap in a gapped AF

- First evidence of a persistent gap in the triplet spectrum and of a kink in its field dependence

- Other properties?

- $\Delta S=2$ steps in magnetization (hard to observe in macroscopic samples)

- Transitions between different phases, as in superfluid Helium 3?

More about DMRG results

