Magnetic-Field and High-Pressure Studies of the Shastry-Sutherland Model Compounds

Sara Haravifard

Quantum Matter: Computation Meets Experiments

Frustrated Quantum Magnetism

Winter Conference

Aspen Center for Physics

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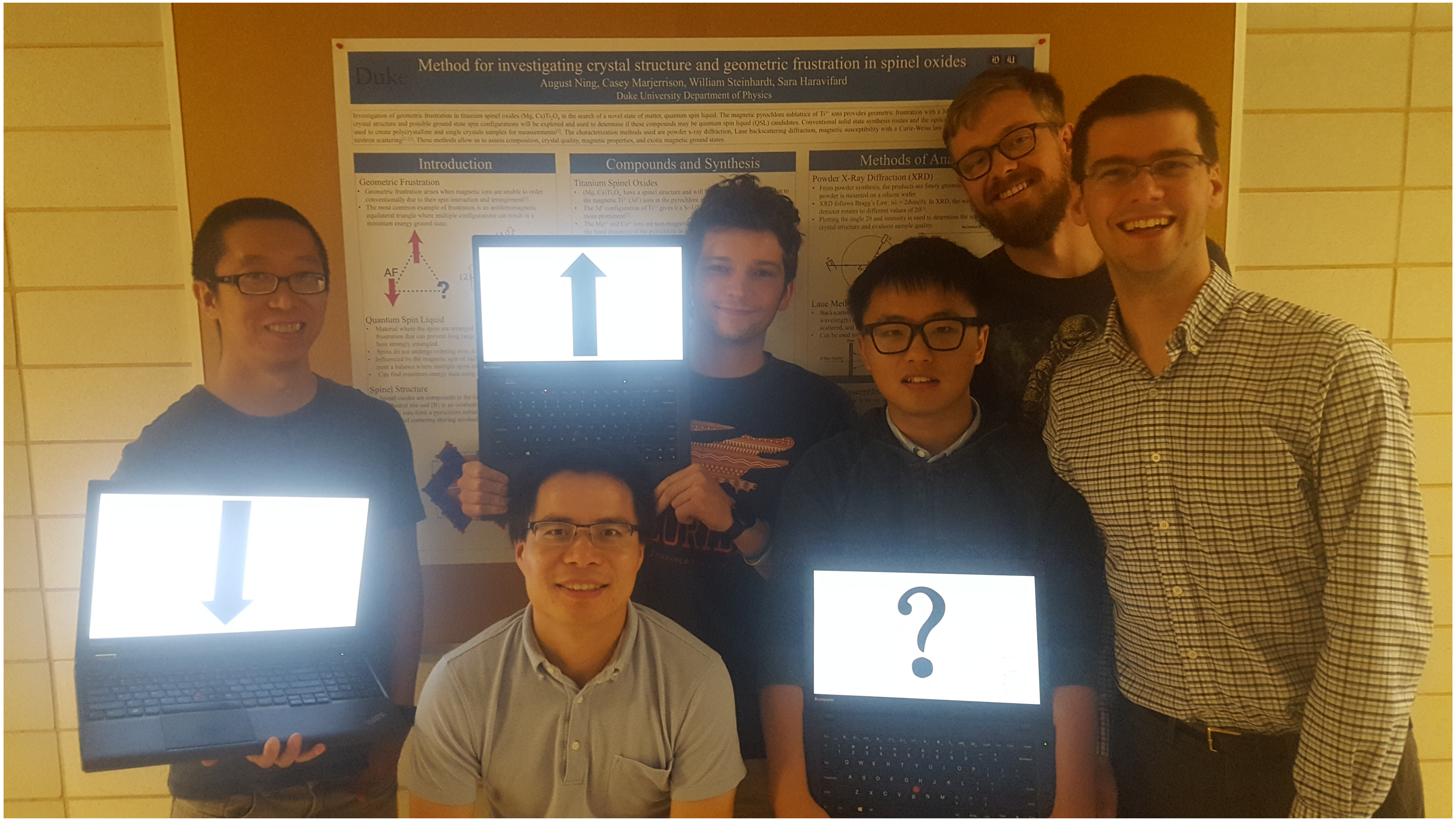
Cristian Batista	*University of Tennessee*
Adrian Feiguin	*Northeastern University*
Bruce Gaulin, Casey Marjerrison, Hanna Dabkowska	*McMaster University*

My Team @ Duke


Method for investigating crystal structure and geometric frustration in spinel oxides
August Ning, Casey Marjerrison, William Steinhardt, Sara Haravifard
Duke University Department of Physics
Introduction
Geometric Frustration
Quantum Spin Liquid
Spinel Structure
Compounds and Synthesis
Titanium Spinel Oxides
Powder X-Ray Diffraction (XRD)
AF
?

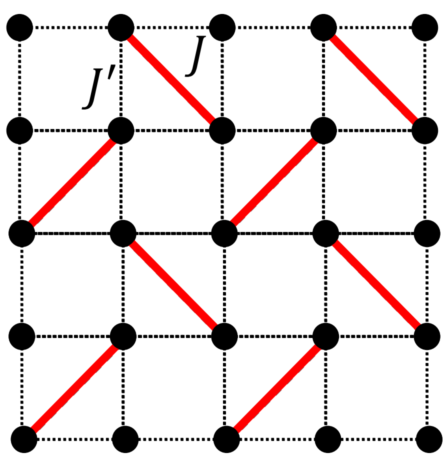
Shastry-Sutherland Model

$$\mathcal{H}_{ss} = J\sum \vec{S_i}\cdot\vec{S_j} + J'\sum \vec{S_i}\cdot\vec{S_j}$$

J , J' are Antiferromagnetic

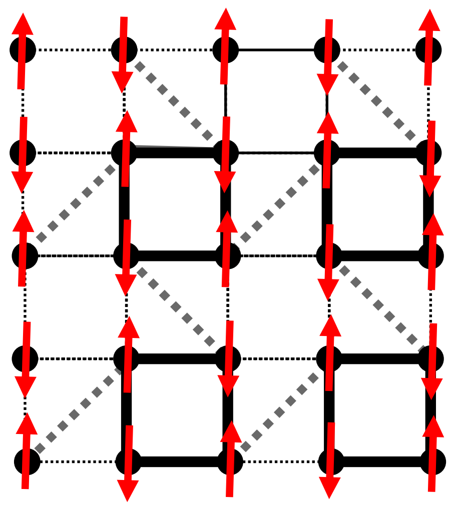
Case I – $J'/J < 0.69$

Isolated Singlet Dimer



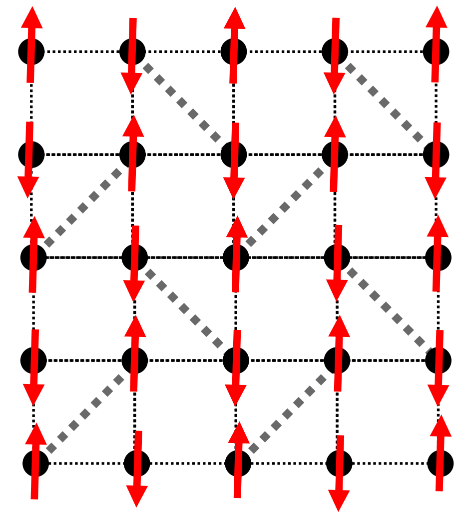
Case I → Case II

Plaquette State

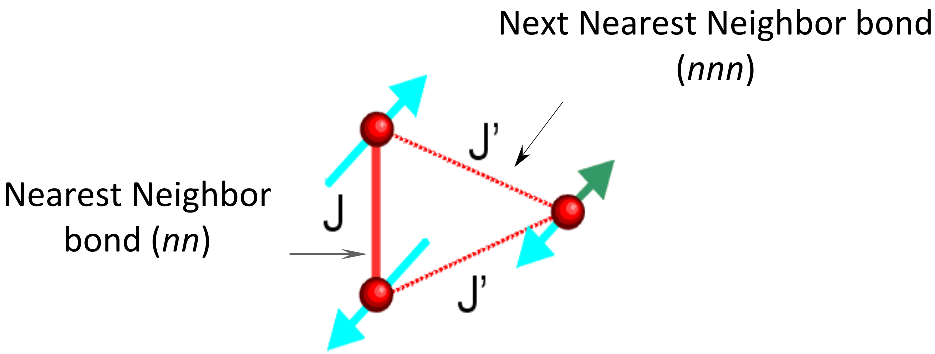


Case II – $J'/J > 0.85$

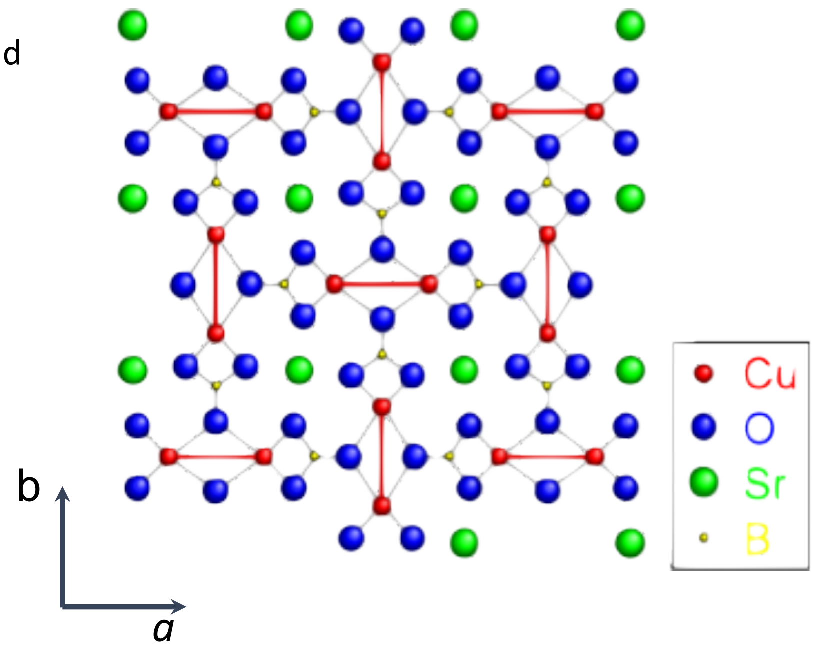
Ordered Neel AF



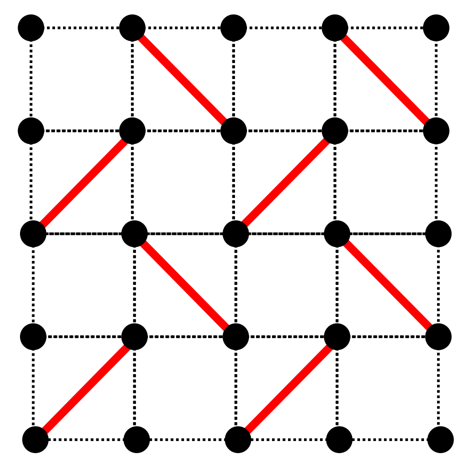
$SrCu_2(BO_3)_2$: Realization of Shastry-Sutherland Model



$$\mathcal{H}_{ss} = J \sum_{nn} \overrightarrow{S_i} \cdot \overrightarrow{S_j} + J' \sum_{nnn} \overrightarrow{S_i} \cdot \overrightarrow{S_j}$$

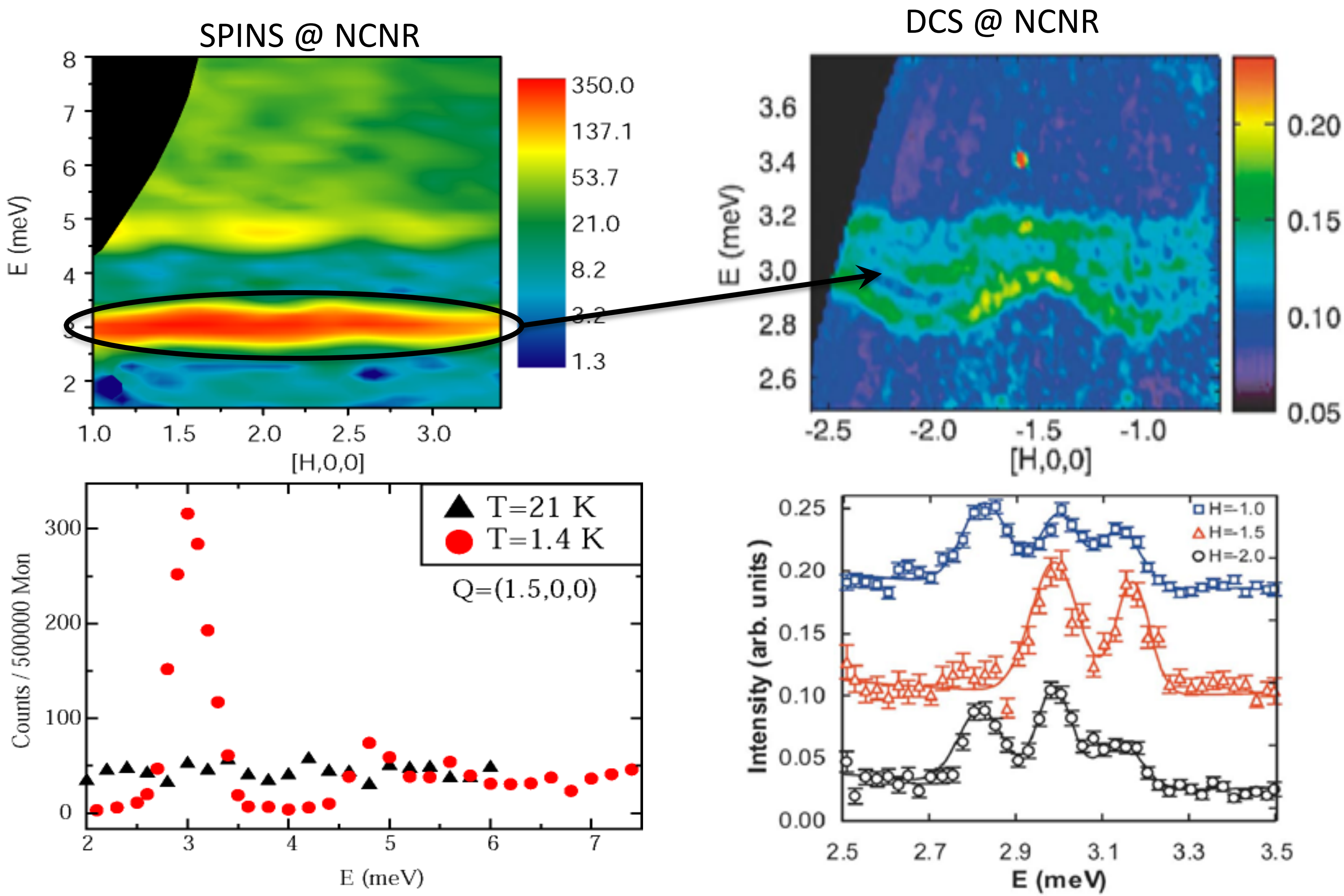


- $a - b$ plane topologically equivalent to Shastry-Sutherland lattice
- $x \equiv \frac{J'}{J} \sim 0.63$
- Weak inter-plane coupling
- Isolated singlet dimer ground state



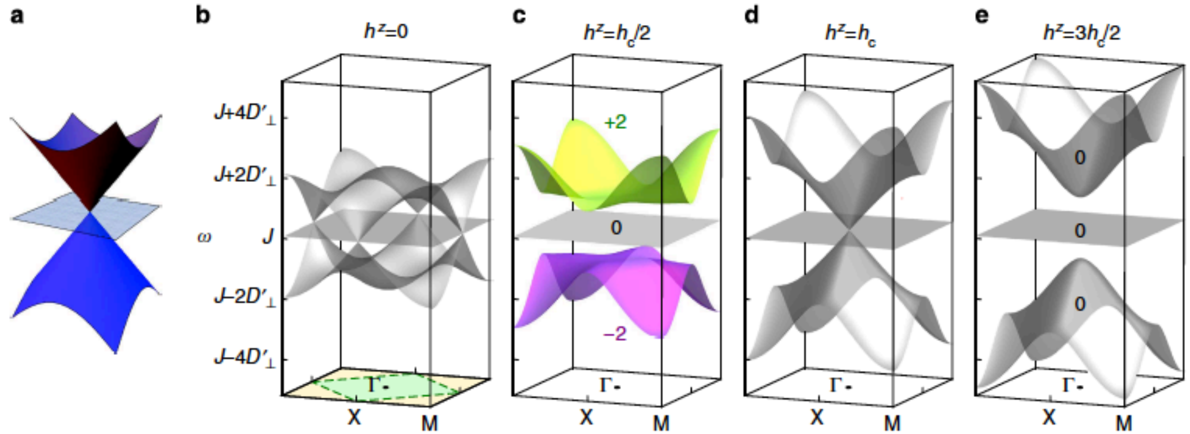
→Use pressure to change bond distances vary x, tune ground state

High Resolution Inelastic Neutron Scattering Results

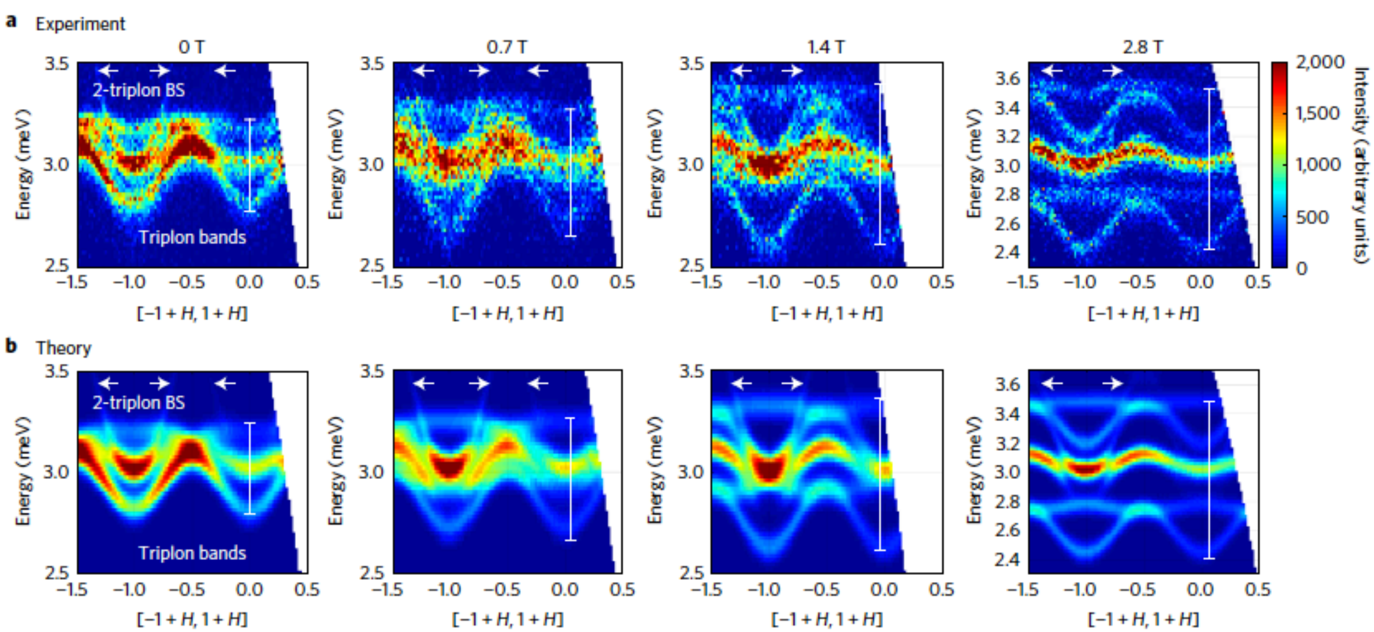


B.D. Gaulin *et al.* , *Phys. Rev. Lett.*, 93, 267202 (2004).

Field-Induced Topological States?



Romhányi, Judit, Karlo Penc, and Ramachandran Ganesh. "Hall effect of triplons in a dimerized quantum magnet." *Nature communications* 6 (2015): 6805.



McClarty, Paul A., et al. "Topological triplon modes and bound states in a Shastry–Sutherland magnet." *Nature Physics* 13.8 (2017): 736.

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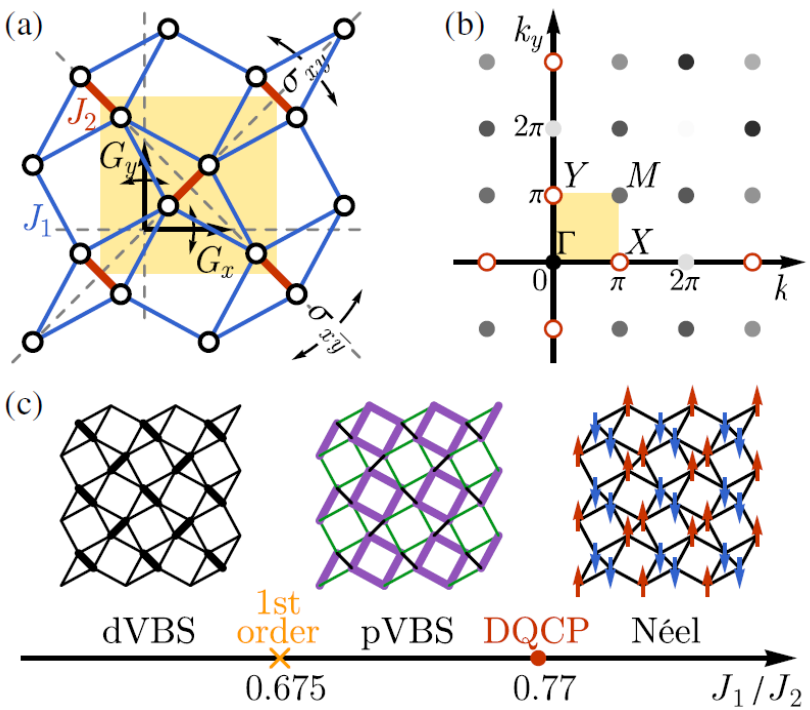
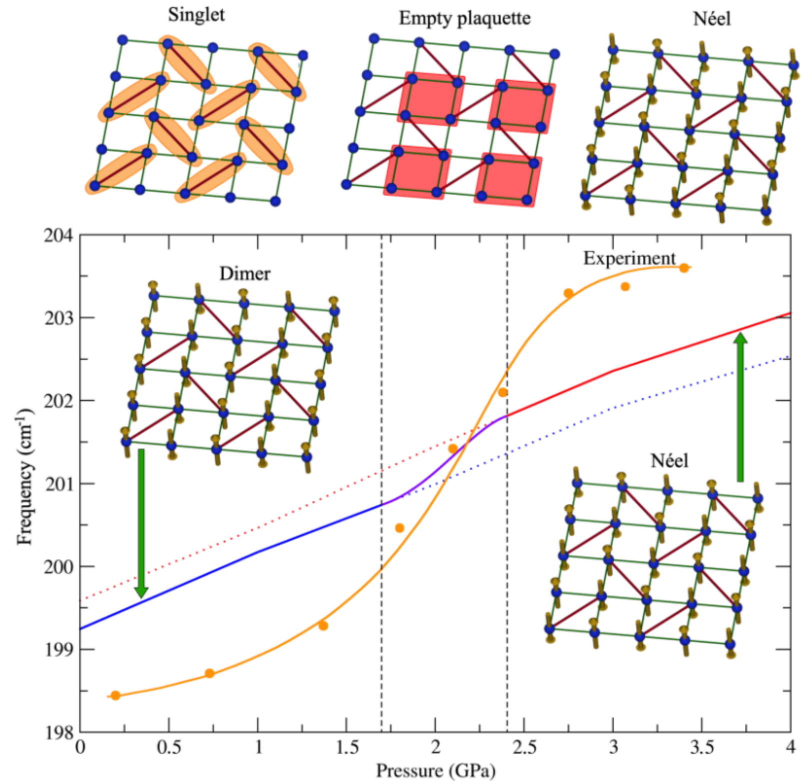
4-spin plaquette singlet



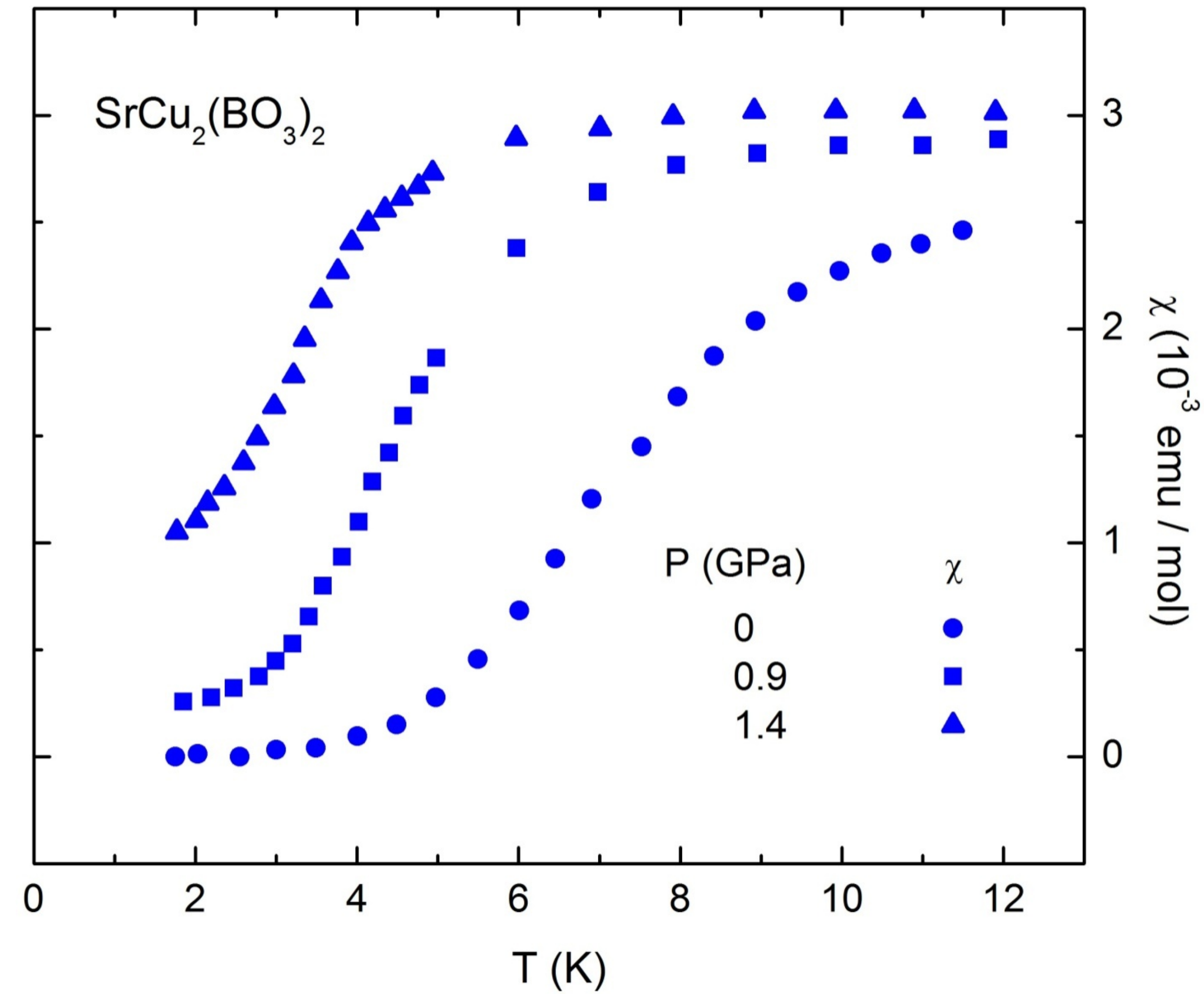
J. Y. Lee, *et al., PRX* ***9***, 041037 (2019)



Badrtdinov, Danis I., *et al. arXiv:2003.01212* (2020)

- **Nature of the 4-spin plaquette singlet?**
- **Deconfined quantum critical point**

Pressure Tuning of Energy Gap in $SrCu_2(BO_3)_2$



Waki *et al. , J. Phys. Soc. Jpn. ,* 76, 073710 (2007).

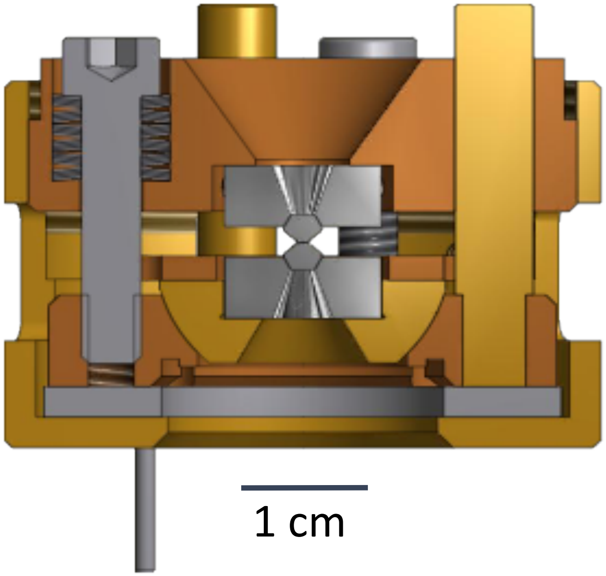
Pressure Tuning in Diamond Anvil Cell

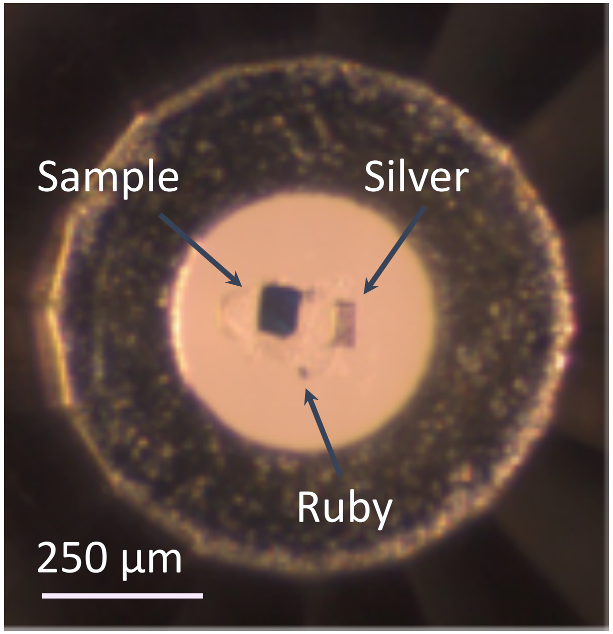
❖Vary coupling ratio by applying hydrostatic pressure to compress sample

❖Experimental range ~0.3-25 GPa: Diamond anvil cell

- Single-crystal sample confined between faces of two diamonds, metallic gasket
- Methanol/Ethanol pressure medium for hydrostaticity
- Ruby and silver manometers
- In situ pressure tuning via helium gas membrane

❖ X-ray measurements: Transmission geometry, synchrotron source





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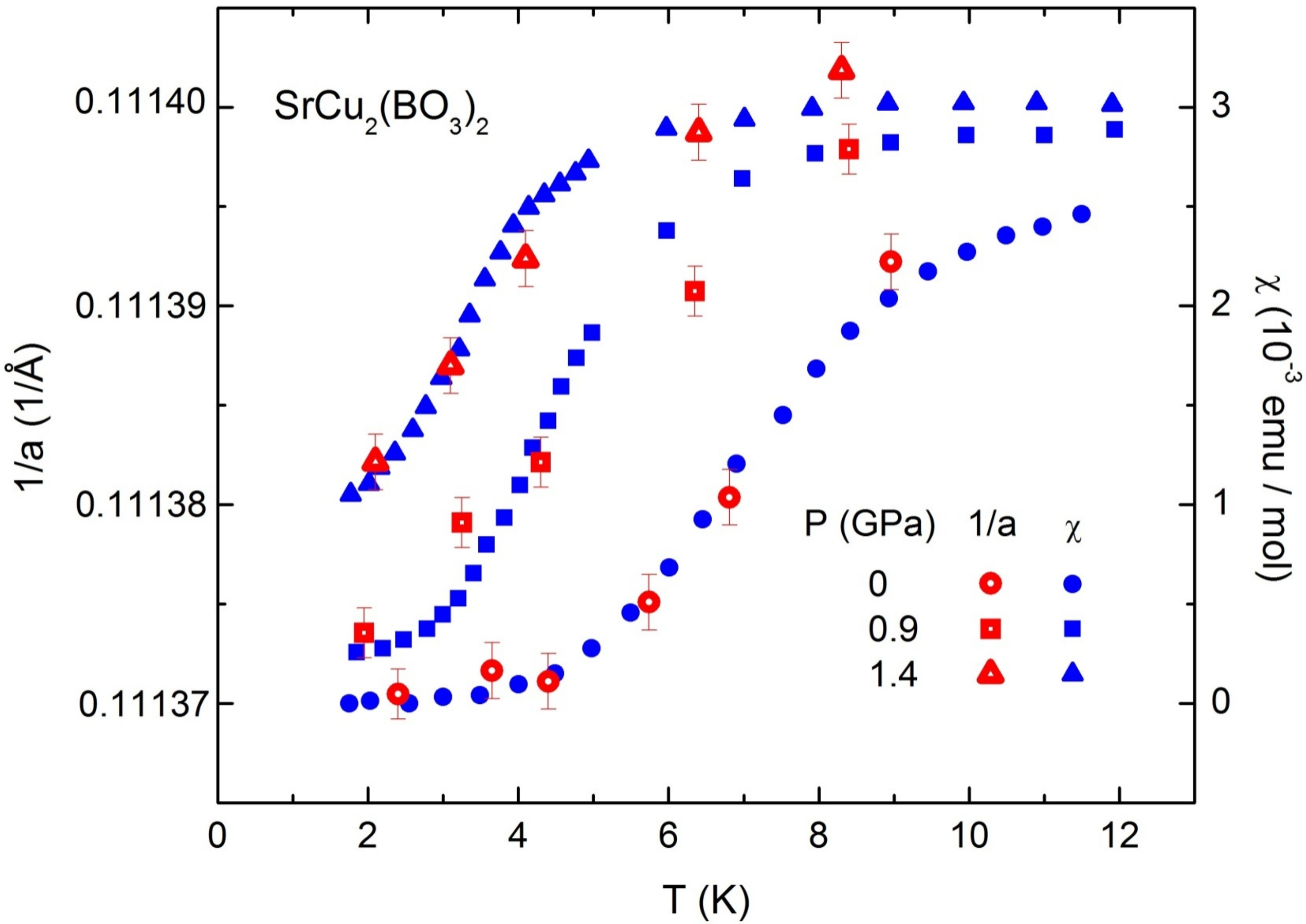
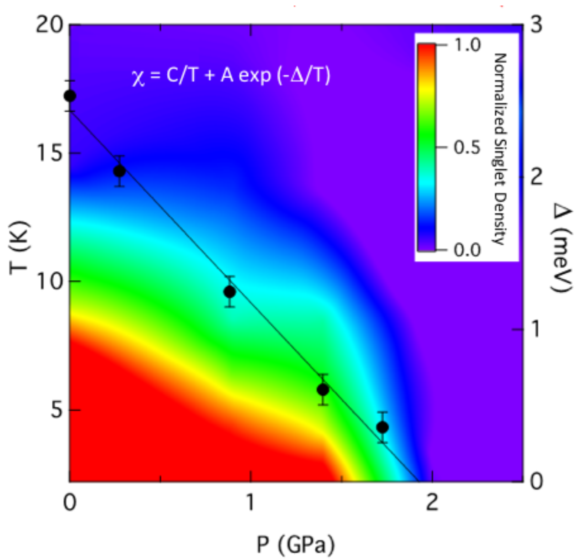
Strong Spin-Phonon Coupling in $SrCu_2(BO_3)_2$

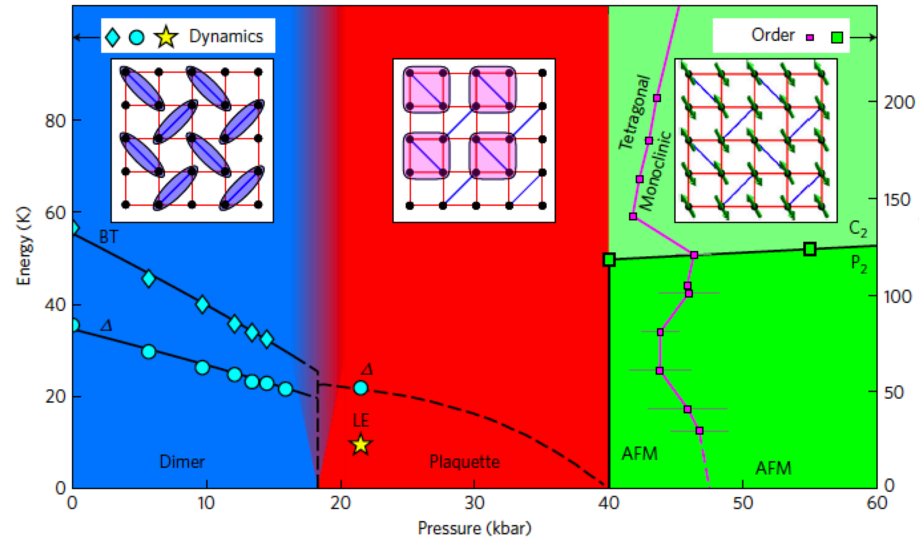


Waki *et al.* , *J. Phys. Soc. Jpn.* , 76, 073710 (2007).
Haravifard *et al.*, *Proc. Natl. Acad. Sci. USA* 109, 2286 (2012).

Mapping the Phase Diagram > Pressure

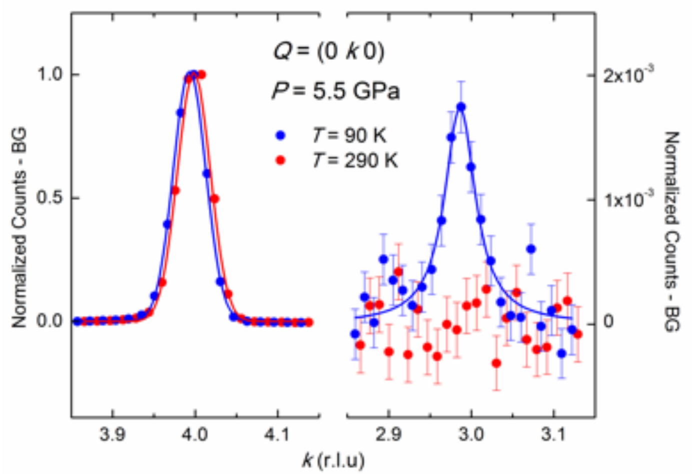


Haravifard *et al.*, *Proc. Natl. Acad. Sci. USA* 109, 2286 (2012).



Zayed, M. E., *et al. Nature Physics* 13.10 (2017).

SNAP @ SNS



Haravifard *et al.*, *Proc. Natl. Acad. Sci.* 111, 14372 (2014).

Boos, C., et al. "Theory of the intermediate phase of SrCu2(BO3)2 under pressure."
arXiv preprint arXiv:1903.07887 (2019).

Guo, Jing, et al. "Quantum phases of SrCu2(BO3)2 from high-pressure thermodynamics."
arXiv preprint arXiv:1904.09927 (2019).

Zhao, Bowen, Phillip Weinberg, and Anders W. Sandvik. "Symmetry-enhanced discontinuous phase transition in a two-dimensional quantum magnet." *Nature Physics* (2019): 1.

Badrtdinov, Danis I., *et al.* "SrCu2(BO3)2 under pressure: a first-principle study."
arXiv preprint arXiv:2003.01212 (2020).

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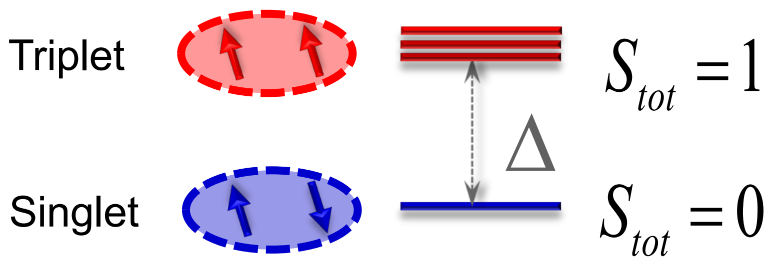
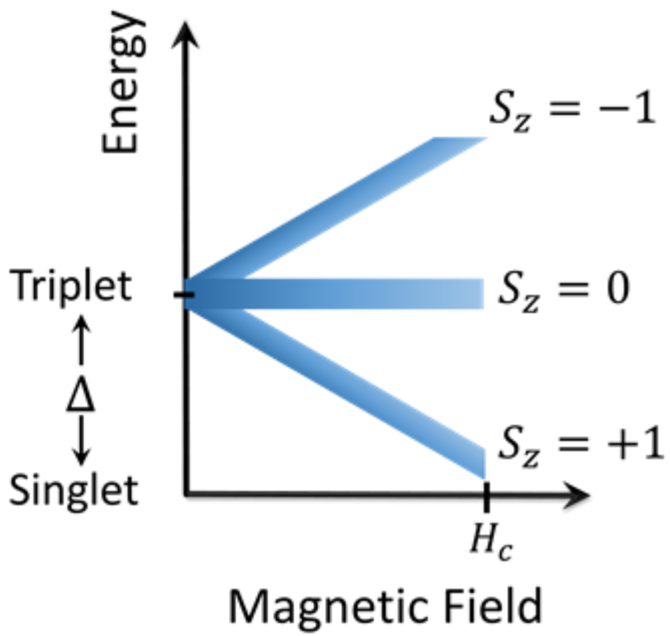
High Magnetic Field Induced Phenomena

Analogy can be portrayed between a spin ½ dimer system in an external magnetic field and a lattice gas of hard-core bosons.



$$\mathcal{H} = J\sum_{nn}\overrightarrow{S_i}\cdot\overrightarrow{S_j} + J'\sum_{nnn}\overrightarrow{S_i}\cdot\overrightarrow{S_j} + g\mu_B H\cdot\sum_i\overrightarrow{S_i}$$

Application of a magnetic field causes the *Zeeman splitting* of the three exited triplet states → The gap ultimately closes at the critical magnetic field $H_c = \Delta/(g\mu_B)$



Accordingly magnetic field exceeding H_c is required to create a finite number of triplets per unit cell of the crystal.

In this depiction:
singlet ↔ vacuum
triplets ↔ bosons with hard-core repulsion
magnetic field ↔ chemical potential

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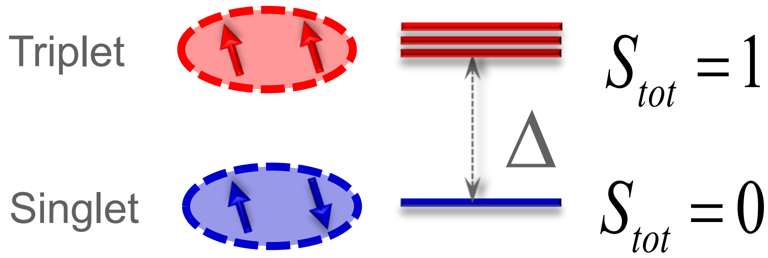
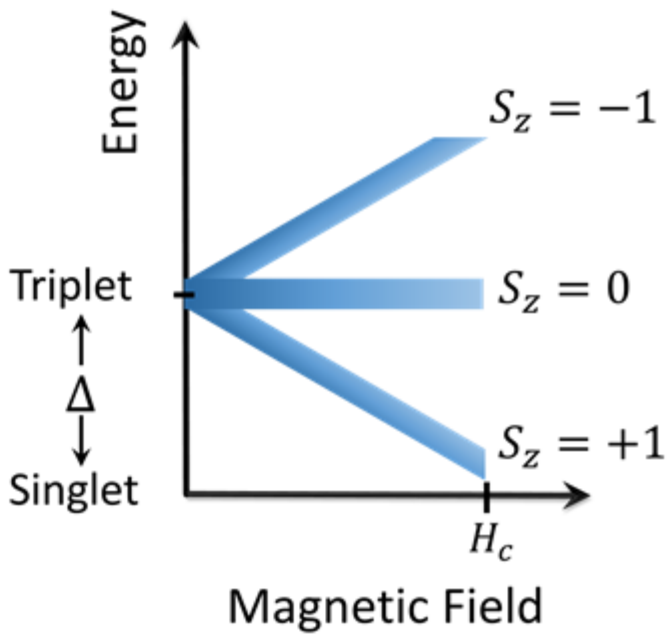
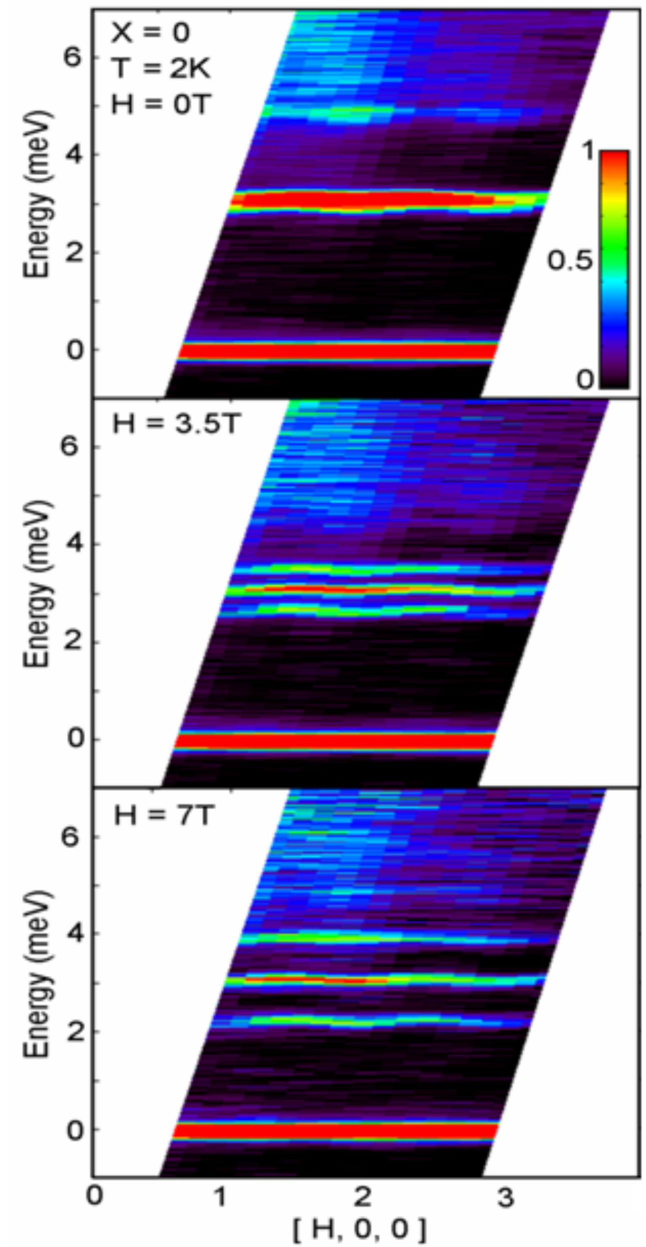
Zeeman Splitting of Triplet States in SCBO

$$\mathcal{H} = J \sum_{nn} \vec{S_i} \cdot \vec{S_j} + J' \sum_{nnn} \vec{S_i} \cdot \vec{S_j} + g\mu_B H \cdot \sum_{i} \vec{S_i}$$







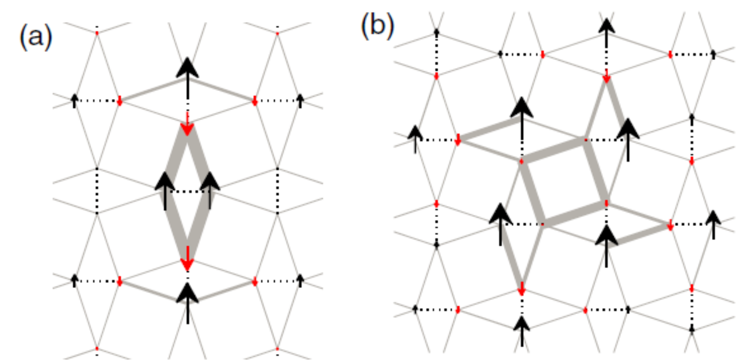
Haravifard *et al.* , *Phys. Rev. Lett.* 97, 247206 (2006).

Mapping the Phase Diagram > Magnetic Field

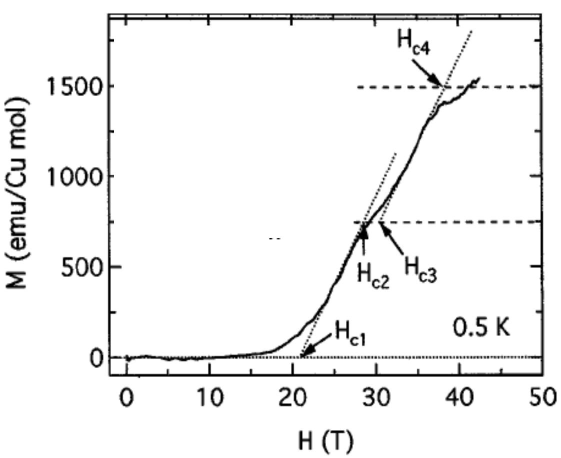
Origin of the <u>magnetization plateaus</u>?

G. Misguich, *et al., Phys. Rev. Lett.* **87,** 097203 (2001)
K. Kodama, *et al., Science* **298,** 395 (2002)
J. Dorier, *et al., Phys. Rev. Lett.* **101,** 250402 (2008)
M. Takigawa, *et al., Phys. Rev. Lett.* **110,** 067210 (2013)
and many more…

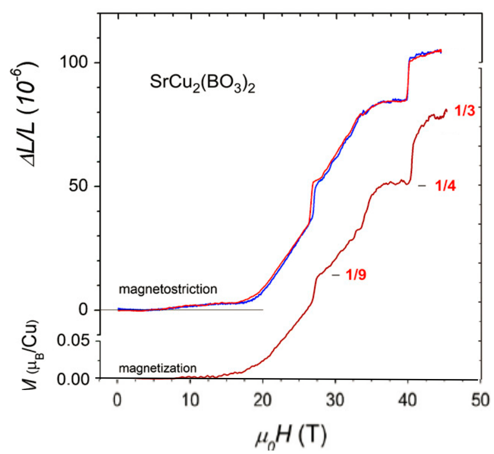
Bound states of triplons (pinwheels): formation of crystals



P. Corboz and F. Mila, *Phys. Rev. Lett.* **112,** 147203 (2014)



H. Kageyama, *et al., Phys. Rev. Lett.* **82,** 3168 (1999)



M. Jaime, *et al., Proc Natl Acad Sci USA* **109**, 12404 (2012)

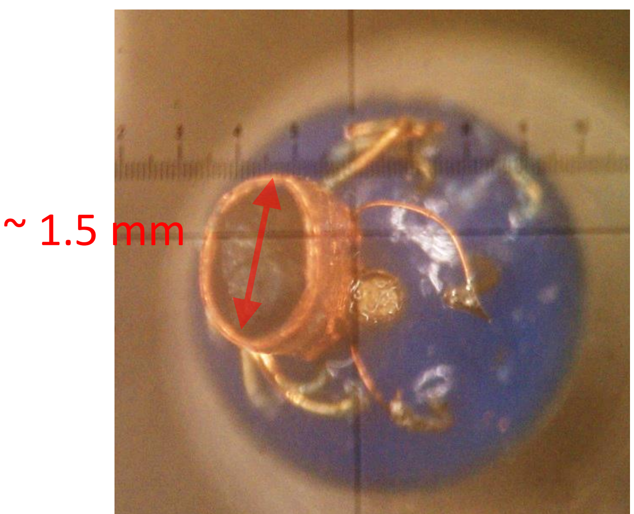
Tuning Magnetization Profiles with Pressure

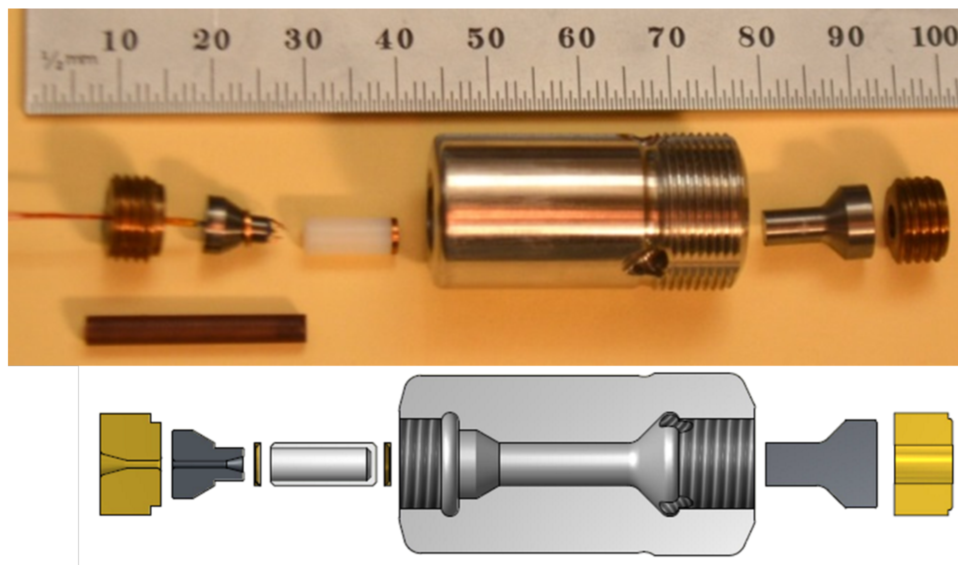
SCBO and spin ½ systems in general, intrinsic magnetic moments are weak, using large samples is crucial.

Applying hydrostatic pressures, can be achieved with the use of pressure cells which limit sample size to millimeter dimensions and smaller.

Alternatively, **Tunnel Diode Oscillators (TDO)** represent a new approach to this challenge. A TDO circuit is constructed of a self-resonating *LC* tank biased by a tunnel diode. Sample is located in a coil which acts as the inductor in the *LC* tank circuit

$\rightarrow \frac{\Delta f}{f} \propto \frac{dM}{dH}$ enabling us to precisely measure changes in the magnetic moment on the order of **10^{-12} emu**





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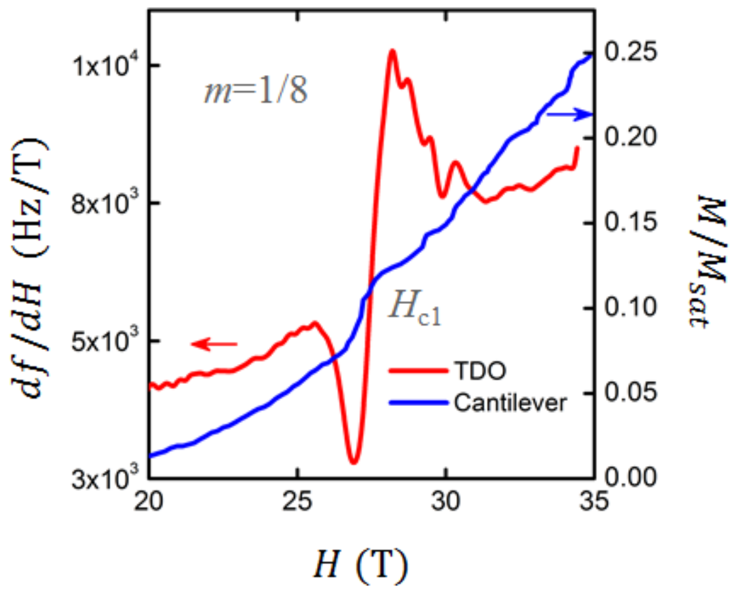
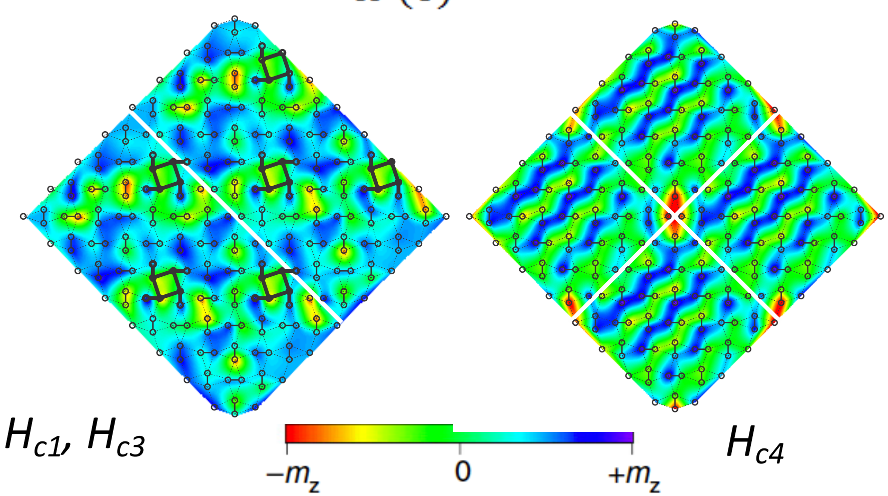
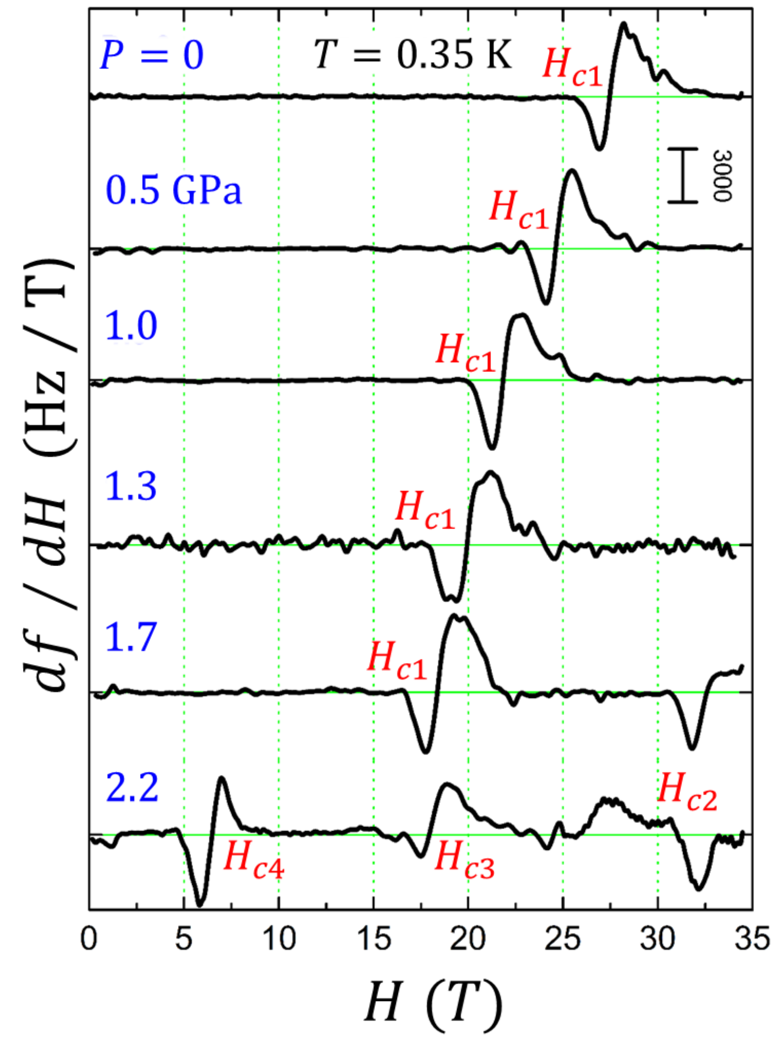
Mapping the Phase Diagram > Pressure & Magnetic Field





Computed at fixed magnetization S_z on lattices with different aspect ratios and open boundary conditions, keeping up to 3000 DMRG states.

$\Delta f/f \propto dM/dH$



Haravifard *et al., Nat. Commun. 7, 11956* (2016).

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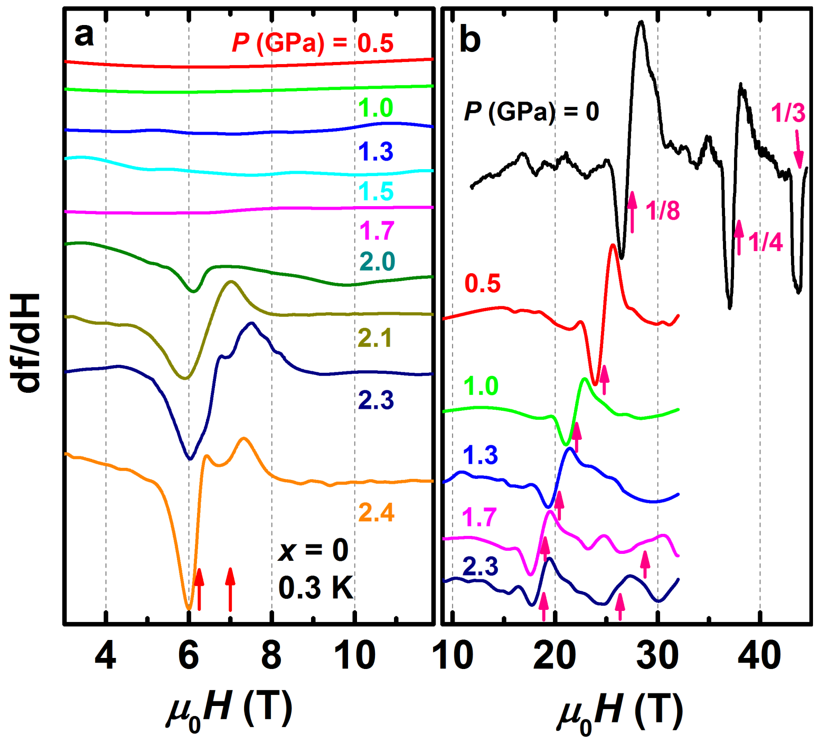
Energy Excitations in Plaquette State



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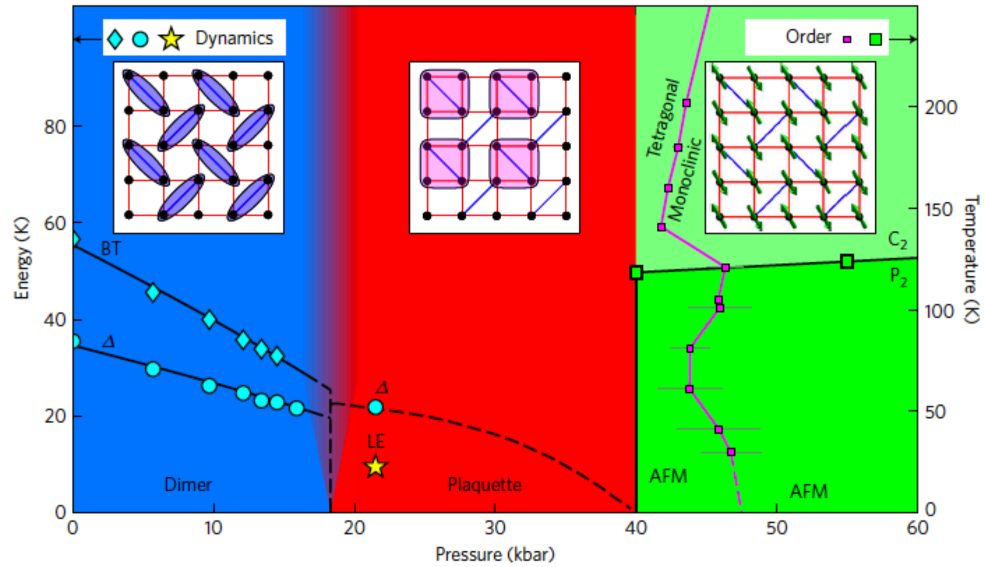

Zayed, M. E., *et al. Nature Physics* 13.10 (2017).

- Above 2.0 GPa, anomaly observed at ~ 7 T, corresponding to LE mode in neutron scattering data reported by Zayed *et al.* 2017.
- Above 2.2 GPa, LE mode splits
- While field of 1/8 plateau seems to saturate above 1.7 GPa, the field of 1/4 plateau seems to continue to decrease with pressure.

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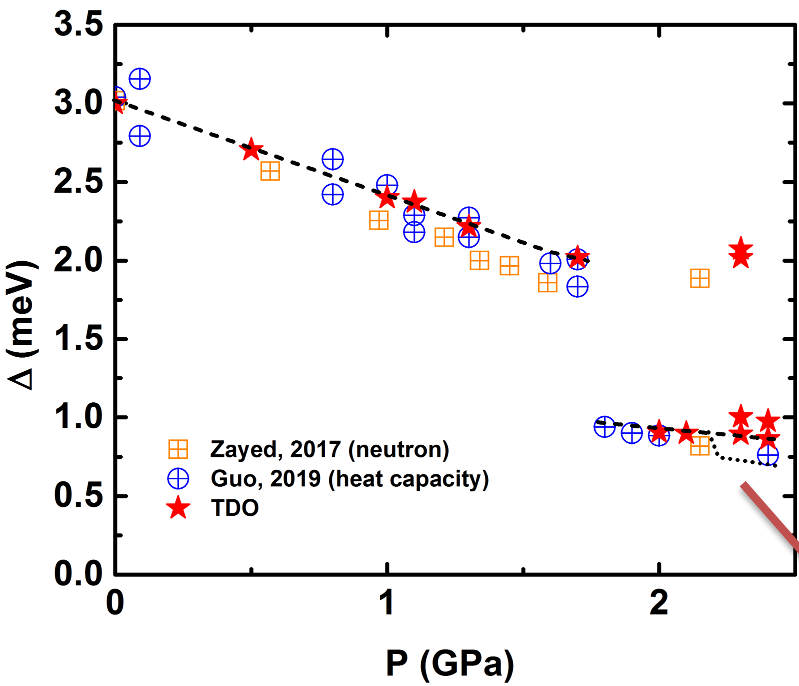
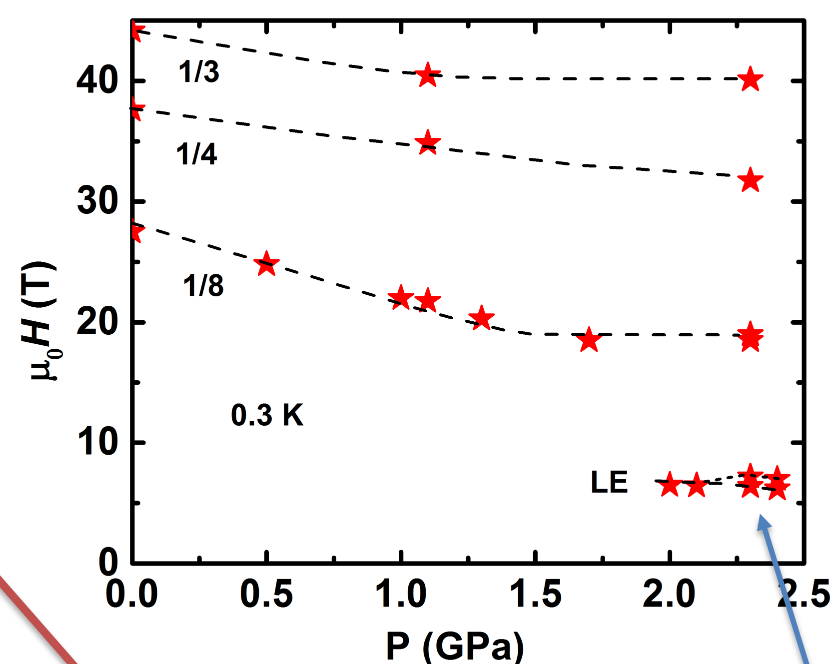
Δ-P Phase Diagram





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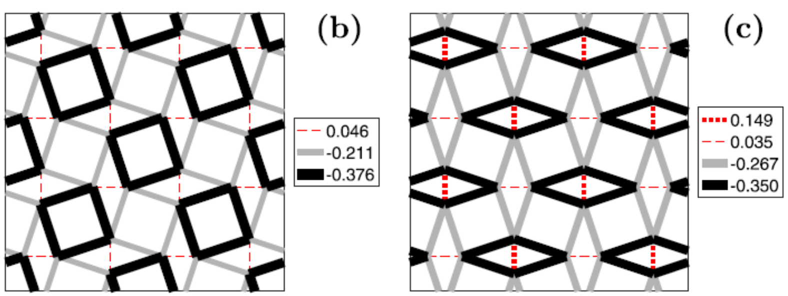

C. Boos, *et al., PRB* **100**, 140413(R) (2019)

2.2 GPa splitting

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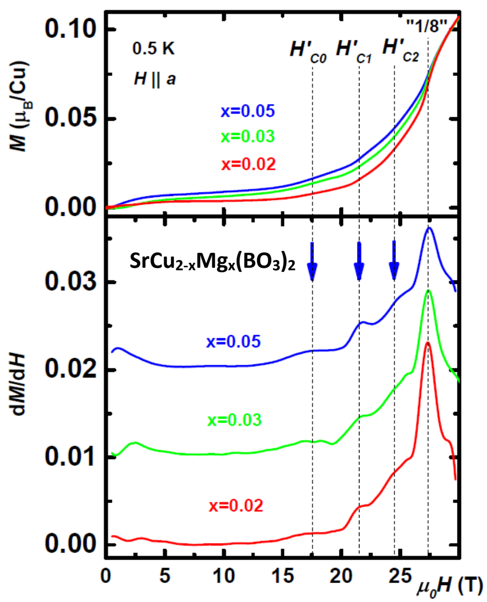
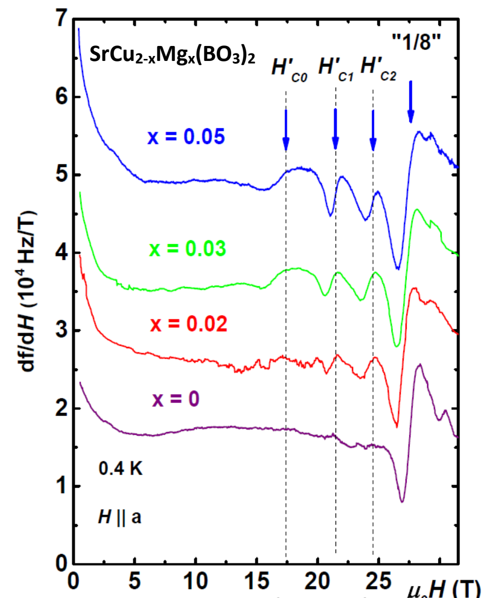
$SrCu_{2-x}Mg_x(BO_3)_2$: anomalies below 1/8 plateau

Magnetization measurement

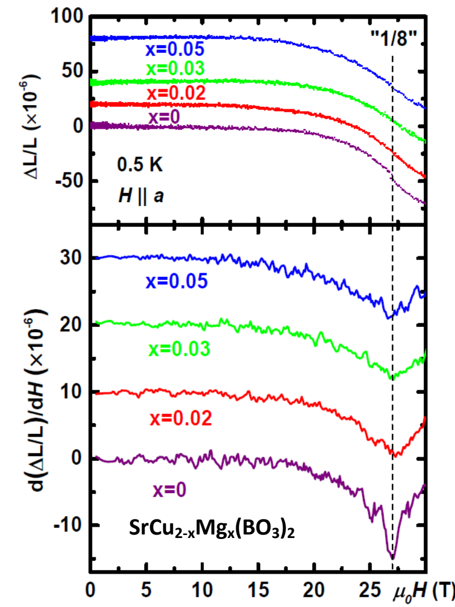


Tunnel diode oscillator (TDO)



df/dH ▯ d^2M/dH^2

Magnetostriction



- **Emergent magnetic states at H'_{c0}~17.5 T, H'_{c1} ~21.5 T and H'_{c2}~24.5 T**
- **Absence in magnetostriction: weak lattice coupling**
- **Superstructure of triplets with unit cell larger than that for 1/8? Not likely**

TDO technique:
R. Clover, *et al., Rev. Sci. Instrum.* **41**, 617 (1970)
Magnetostriction technique:
M. Jaime, *Sensor* **17**, 2572 (2017)

Shi, Zhenzhong, *et al*. "Emergent Bound States and Impurity Pairs in Chemically Doped Shastry-Sutherland System." Nature Commun. 10, 2439 (2019).

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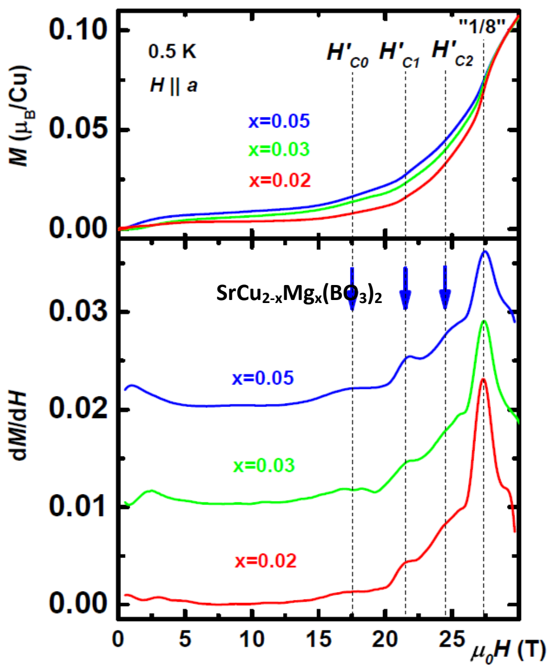
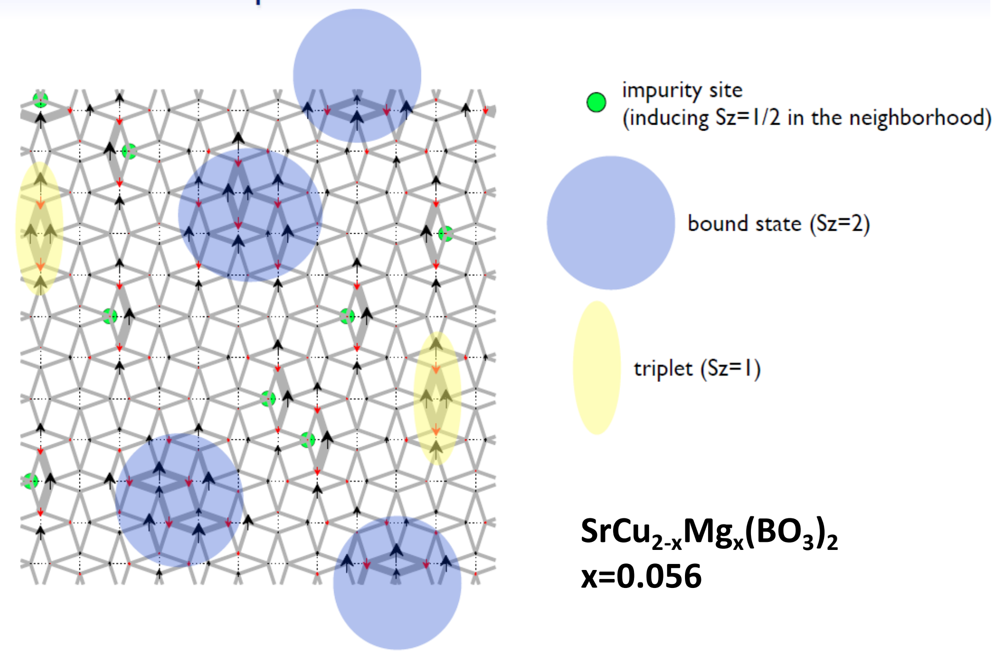
Infinite projected entangled-pair state (iPEPS): special configuration of impurities

Magnetization measurement





Better answer: Triplet, Bound states of triplets

Shi, Zhenzhong, *et al*. "Emergent Bound States and Impurity Pairs in Chemically Doped Shastry-Sutherland System." Nature Commun. 10, 2439 (2019).

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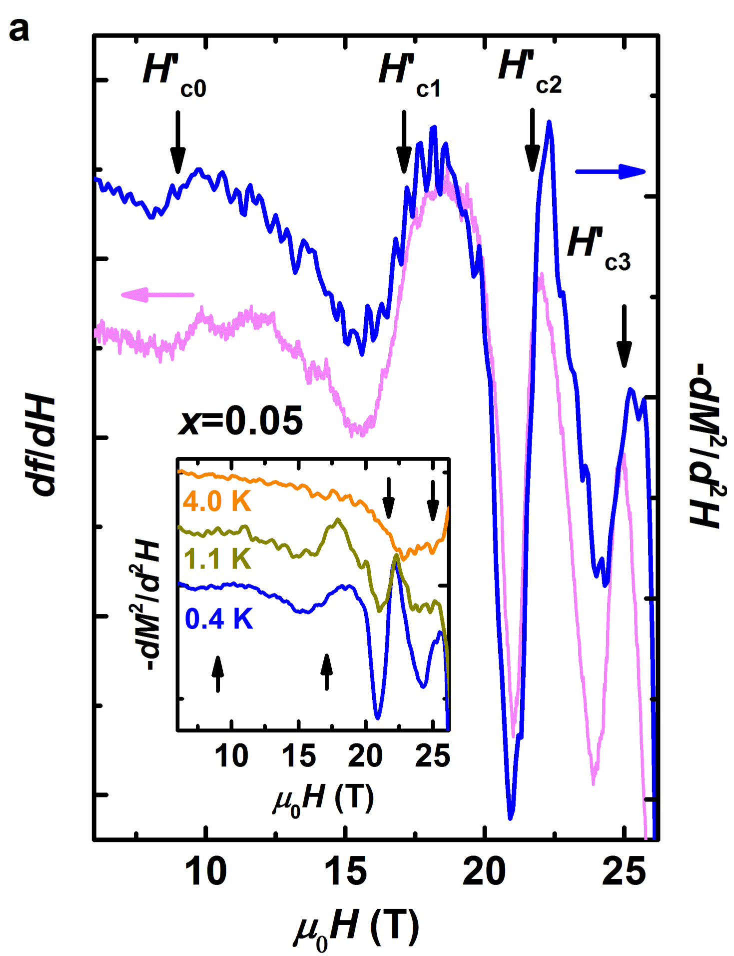
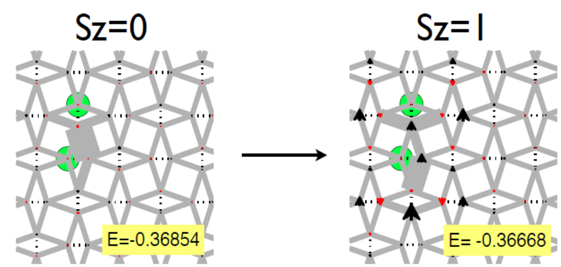
Special Configuration of impurities



Special 2-impurity configurations (8x8 unit cell, D=6)



Energy per excitation: **0.2381J**

Compared to free triplet excitation:

Energy per excitation: **0.430J**

Better answer: Bound states of impurities

Shi, Zhenzhong, *et al*. "Emergent Bound States and Impurity Pairs in Chemically Doped Shastry-Sutherland System." Nature Commun. 10, 2439 (2019).

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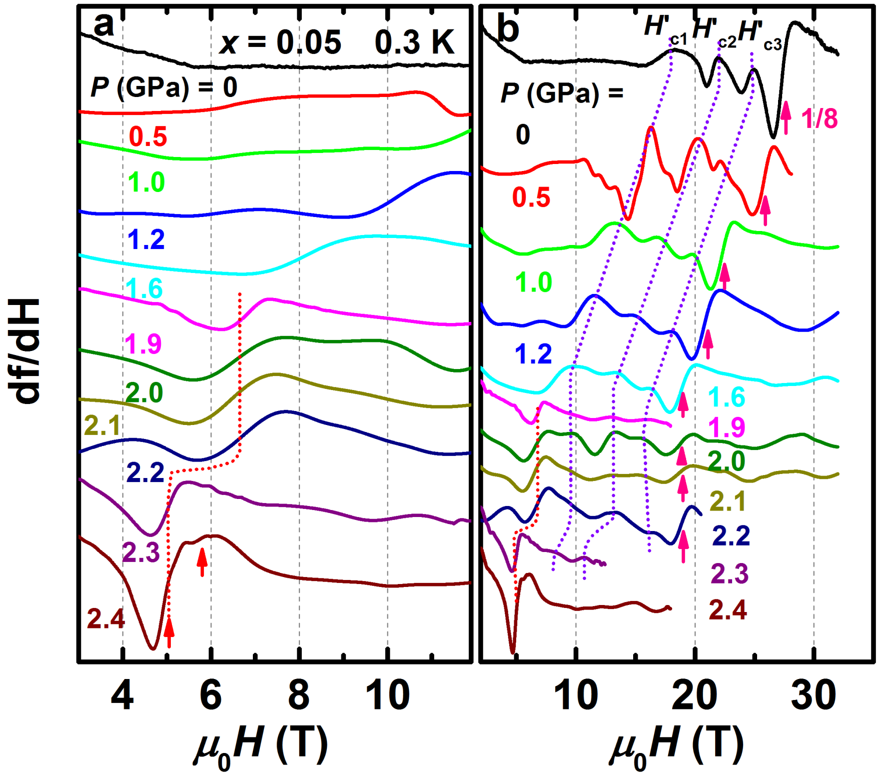
TDO x=0.05 @ Different Pressures



Unpublished

x=0.05 SCBO:

(1) 1/8 Plateaus vs pressure, see pink arrows.

(2) H'_{c1}, H'_{c2}, and H'_{c3} tracked as a function of pressure, all the way into the plaquette phase.

- **Above 1.9 GPa, anomaly observed at ~ 7 T, corresponding to LE mode in Zayed, 2017.**
- **Above 2.2 GPa, LE mode splits, as in the pure case.**
- **Above 2.2 GPa, unlike the pure case, LE mode suddenly softens.**

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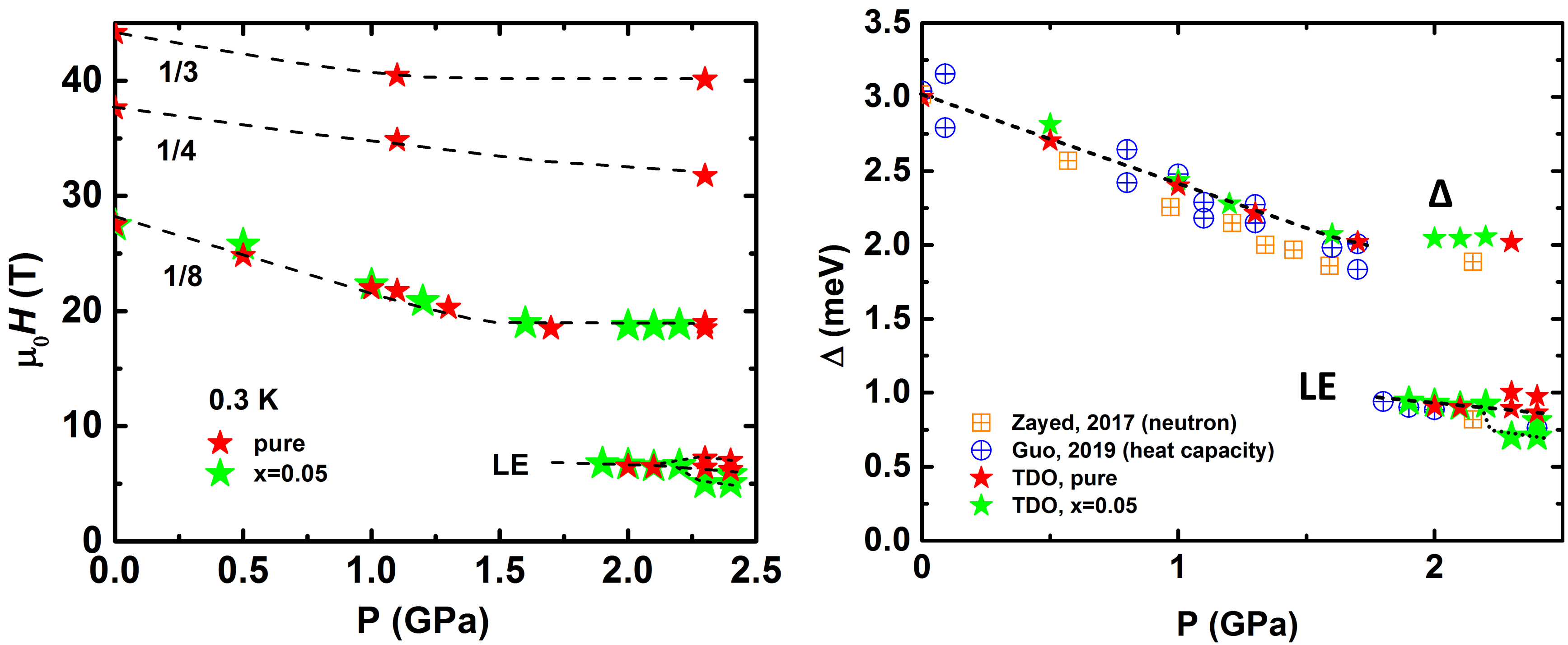
Δ-P Phase Diagram
x=0 & x=0.5



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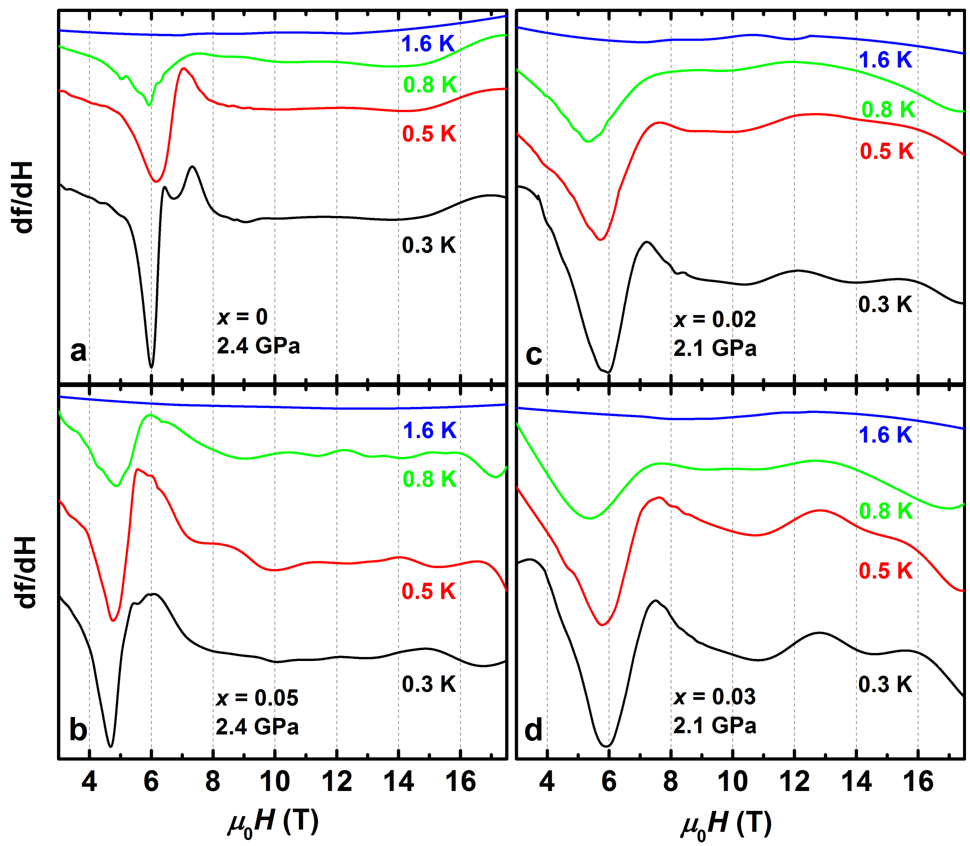
TDO all doping @ Different Temperature



For all dopings:

(1) The splitting of LE mode can only be seen below 0.5 K.

(2) LE mode anomaly disappears above 0.8 K.

(3) H'_{c1}, H'_{c2}, and H'_{c3} at high P can also be seen in x=0.02 and x=0.03 samples.

Unpublished

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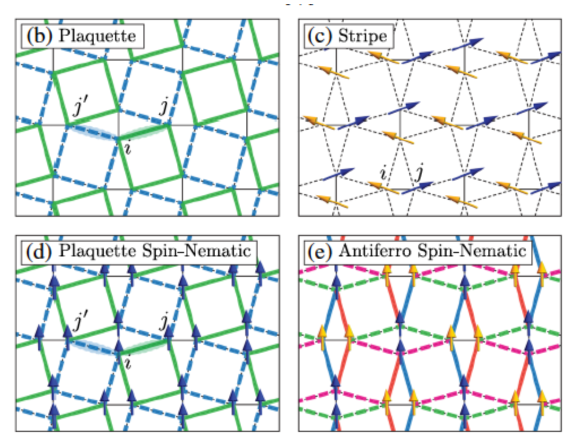
Field- & Pressure-Induced Emergent Phases > Nature of Plaquette State

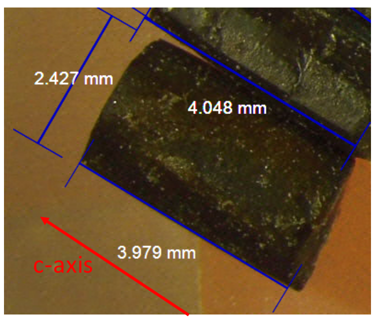
Wang, Zhentao, and Cristian D. Batista. "Dynamics and Instabilities of the Shastry-Sutherland Model." ***Physical review letters*** **120.24 (2018): 247201.**

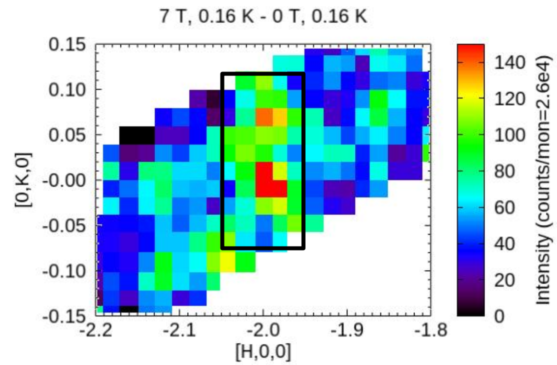
Badrtdinov, Danis I., *et al.* "SrCu2(BO3)2 under pressure: a first-principle study."
arXiv preprint arXiv:2003.01212 (2020).

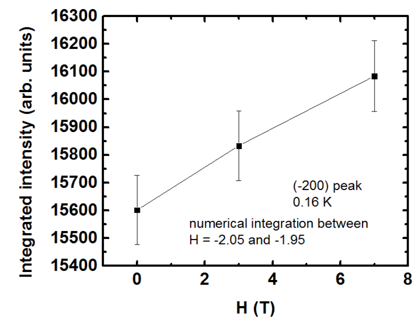
Boos, C., et al. "Theory of the intermediate phase of SrCu2(BO3)2 under pressure." *arXiv preprint arXiv:1903.07887* (2019).

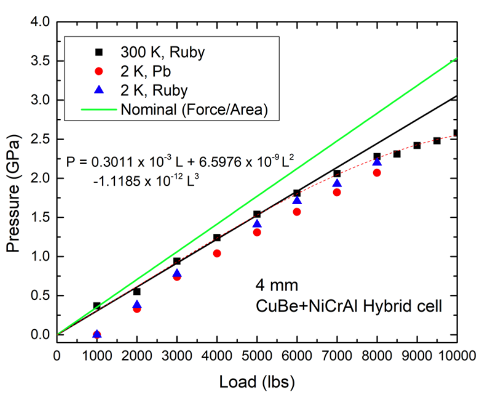
Guo, Jing, et al. "Quantum phases of SrCu2(BO3)2 from high-pressure thermodynamics." *arXiv preprint arXiv:1904.09927* (2019).

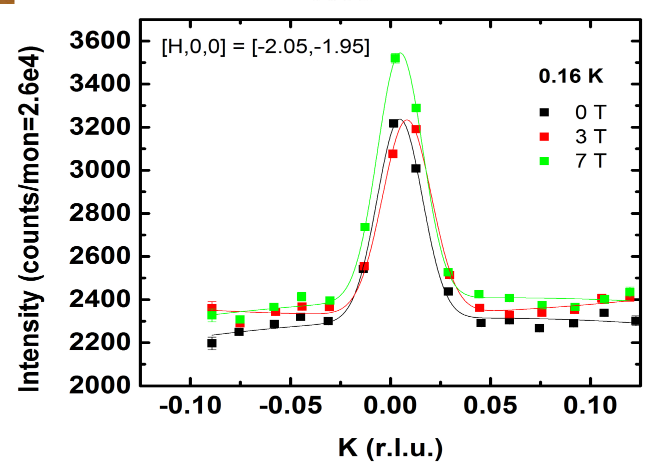


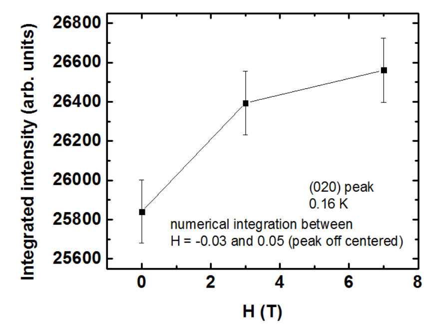












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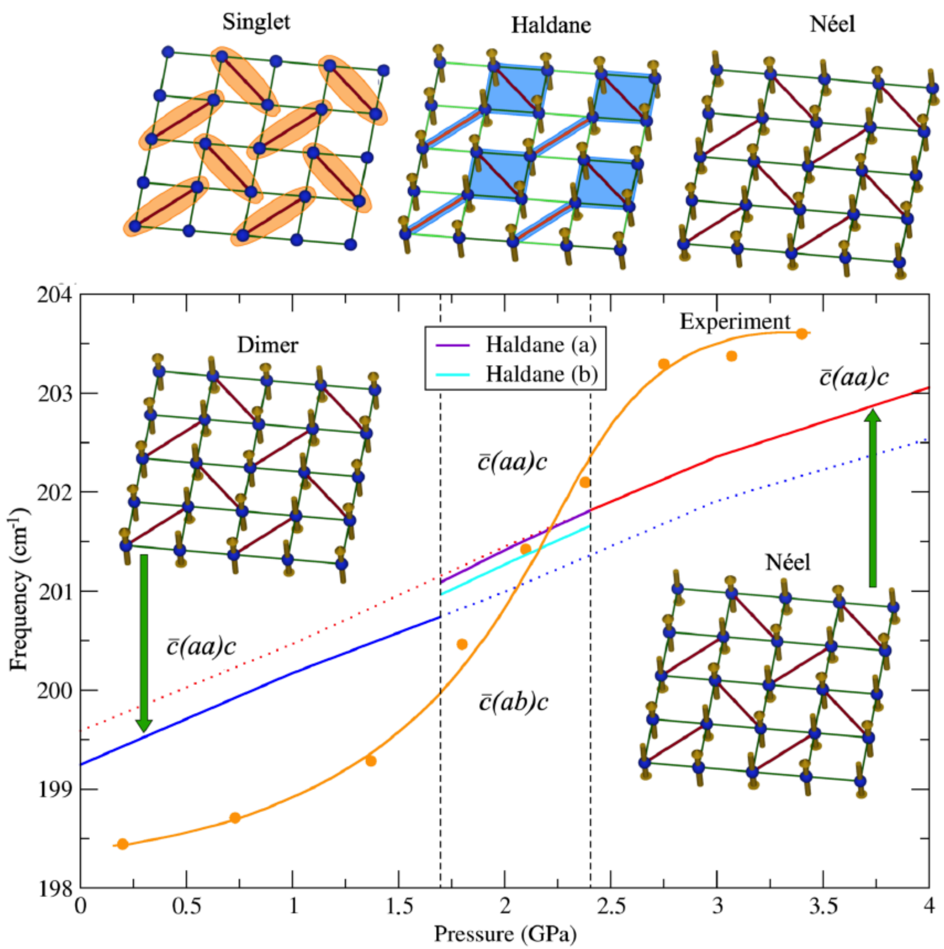
Pressure Evolution of Shastry-Sutherland Model



Magnetic configurations of Haldane the intermediate pressure phase:

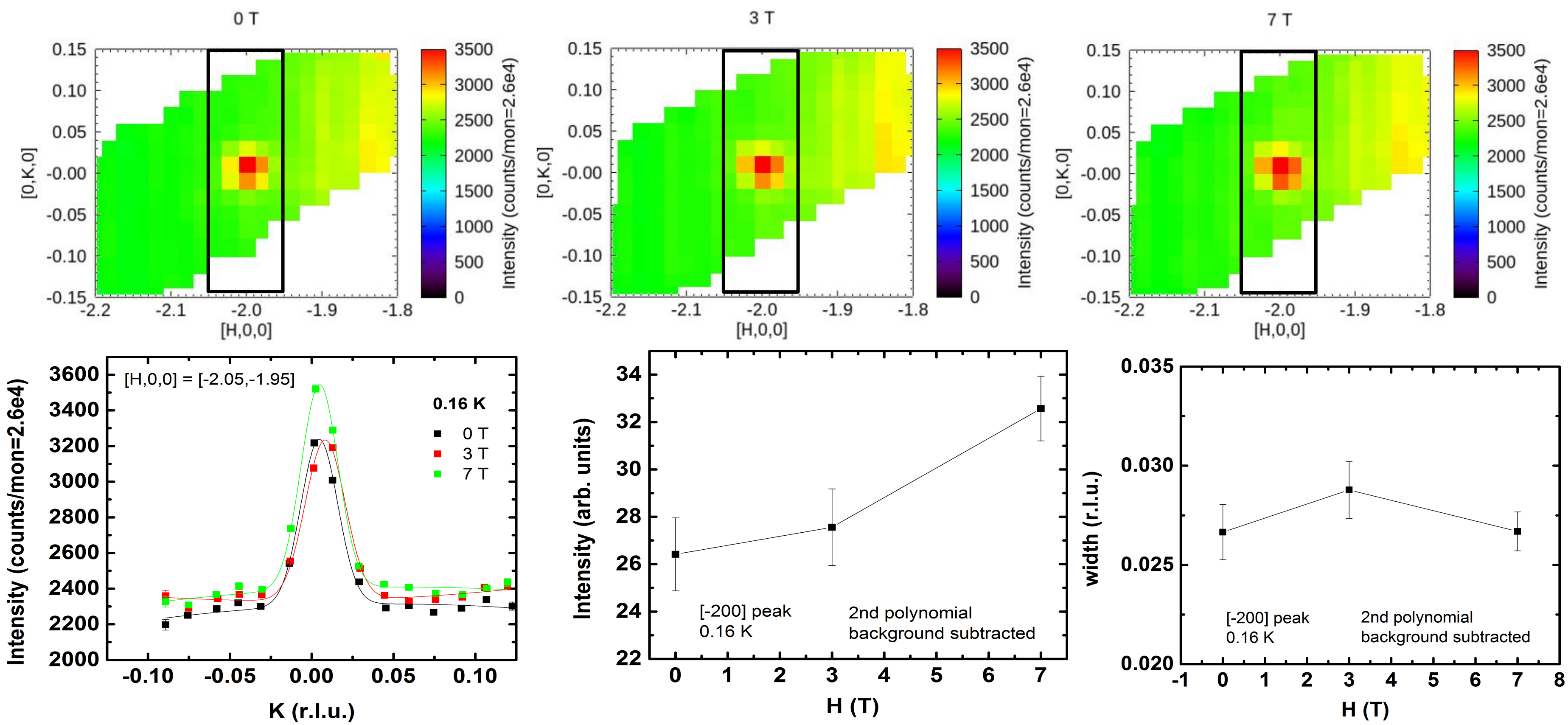
pantograph mode frequencies & DFT+U Calculations

Half of the nearest-neighbor dimers have antiparallel spins and the remaining half have parallel spins.

The choice of magnetic phase modifies the crystal structure.

Badrtdinov, Danis I., *et al. arXiv:2003.01212* (2020)

(-200) Bragg peak @ 0.16 K; 2.35 GPa



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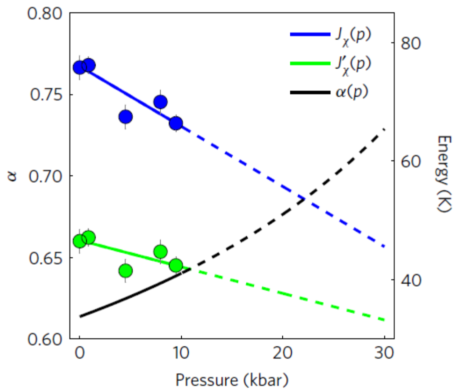
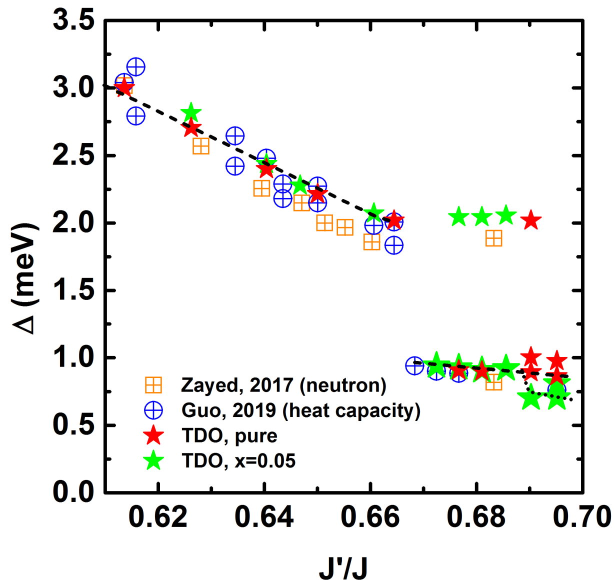
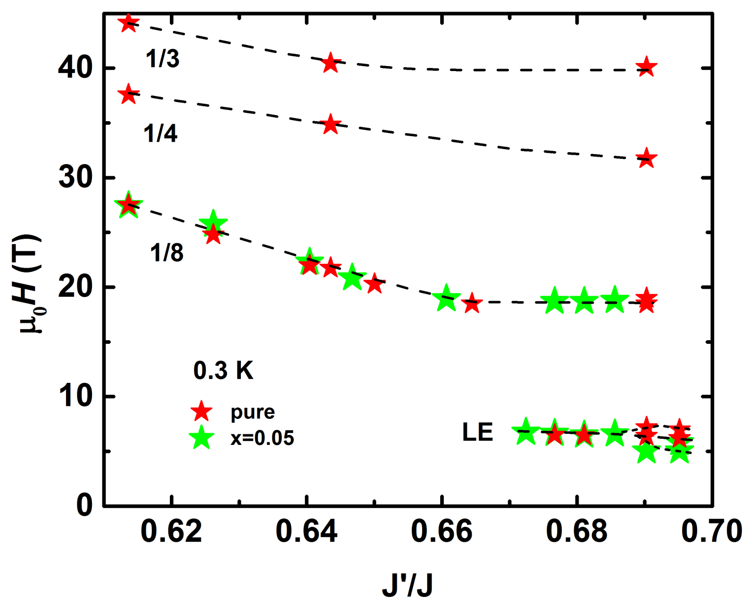
Pressure -> J'/J



M. E. Zayed et al. Nat. Phys. 2017





Pressure to J'/J relation from Zayed et al 2017 is used. Phase diagram in page 6 and 7 is replotted with J'/J.

Pressure (GPa)	J' (K)	J (K)	J'/J
0	46.5	75.7	0.614
1	42.2	65.7	0.640
2	37.8	55.6	0.677
2.4	36.0	51.7	0.695

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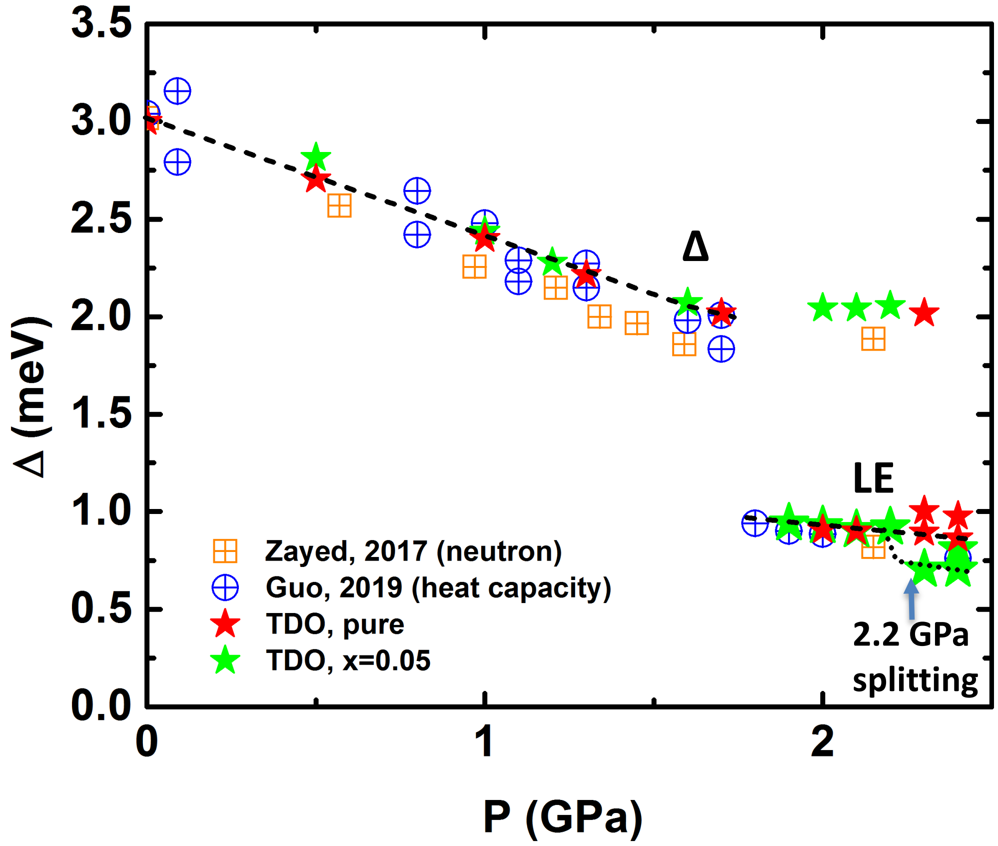
Questions



(1) What happens at P > ~2.2 Gpa? Additional symmetry breaking? Interlayer coupling? DM interaction?

(2) Why there is coexistence of H'_{c1}, H'_{c2}, and H'_{c3} and LE mode at high P? Spatial separation of different phases, or there are plaquette versions of H'_{c1}, H'_{c2}, and H'_{c3}?

(3) What is the nature of the plateaus in the plaquette phase? Plaquette two-triplon?

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Conclusion

❖Frustrated Magnets are effective candidates in exploring emergent quantum phenomena and topological states of matter.

❖Tuning parameters such as pressure, magnetic field, chemical doping can be used to induce QPTs and map phase diagrams.

❖Technical advancements – specially in sample environment - play key role.

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