Phase Diagram of Shastry-Sutherland Compound $SrCu_2(BO_3)_2$ at High-Pressure and High-Field

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INTERNATIONAL WORKSHOP ON QUANTUM MAGNETS IN EXTREME CONDITIONS

Please join via Zoom

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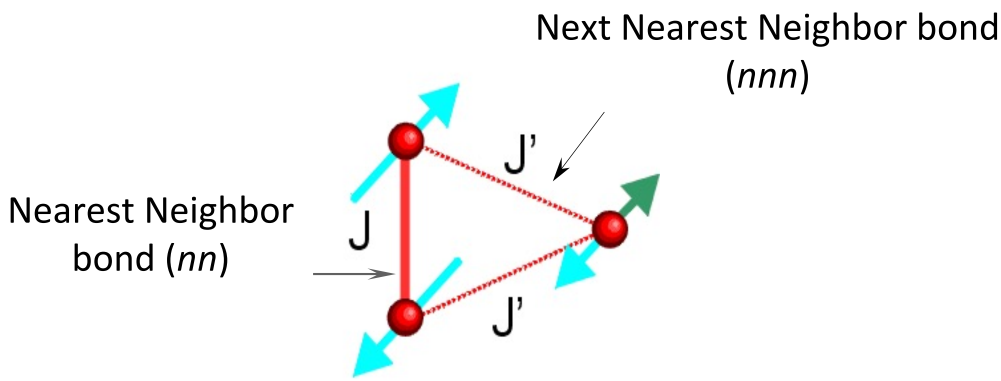
Caltech

Adrian Feiguin
Northeastern University

Duke University

My Team @ Duke (pre-COVID!)

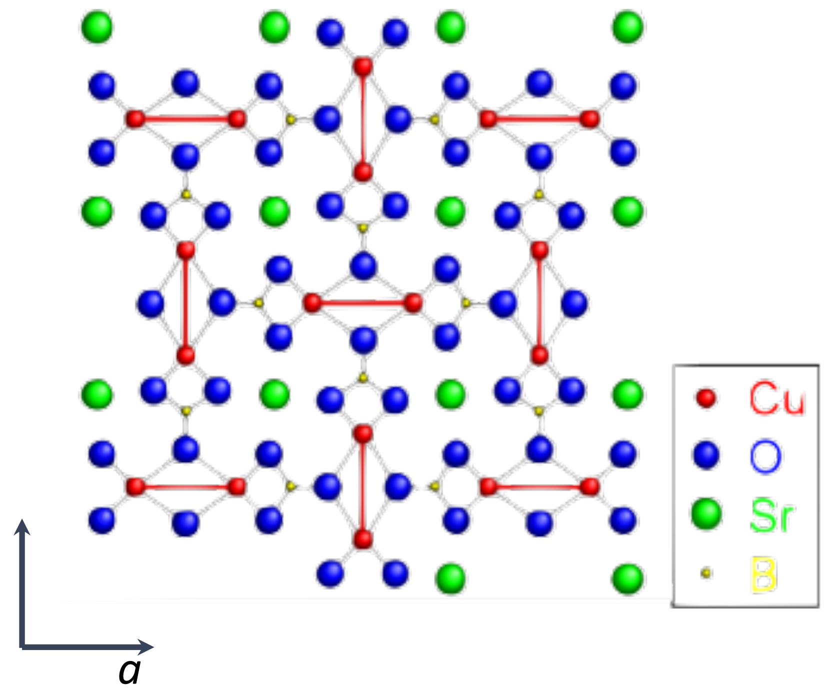
$SrCu_2(BO_3)_2$: Realization of Shastry-Sutherland Model

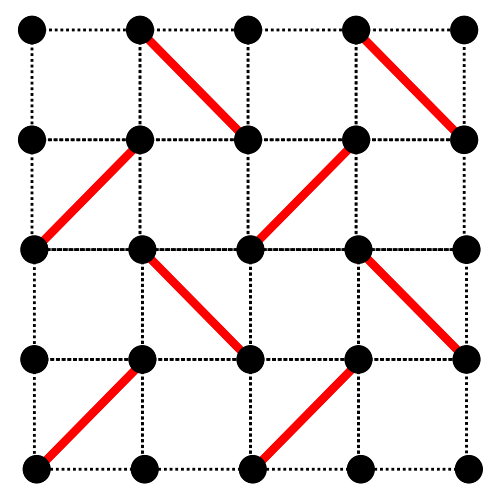


$$\mathcal{H}_{ss} = J \sum_{nn} \overrightarrow{S_i} \cdot \overrightarrow{S_j} + J' \sum_{nnn} \overrightarrow{S_i} \cdot \overrightarrow{S_j}$$

- $a - b$ 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





Tuning knob 1: Pressure
Ground states at high pressure

❖Vary coupling ratio by applying hydrostatic pressure to compress sample

❖Experimental range ~1-7 GPa: Diamond anvil cell

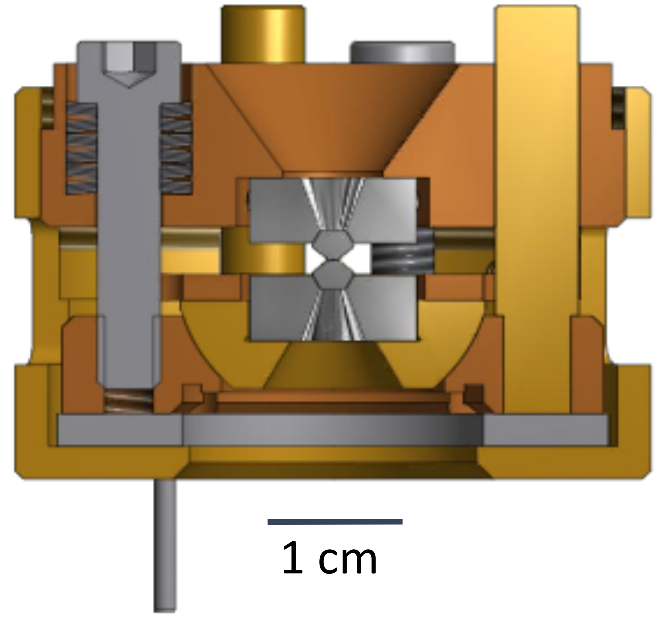
- Single-crystal sample confined between faces of two diamonds, metallic gasket

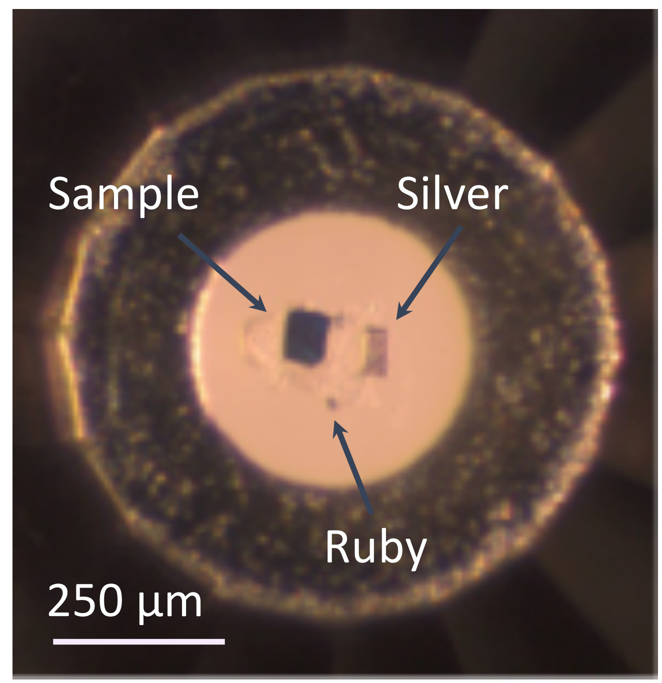
- Methanol/Ethanol pressure medium for hydrostaticity

- Ruby and silver manometers

- In situ pressure tuning via helium gas membrane

❖ X-ray measurements: Transmission geometry, synchrotron source





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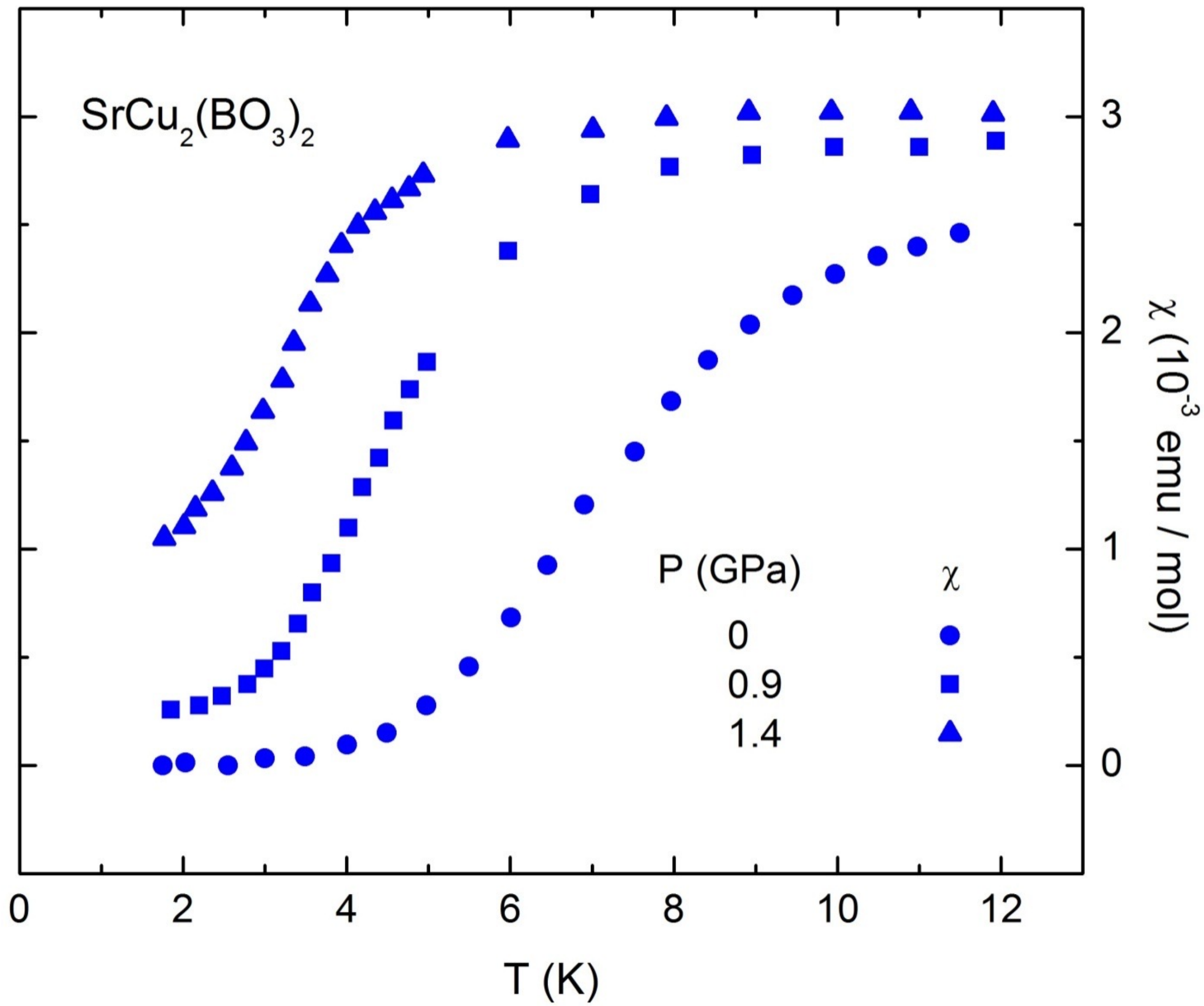
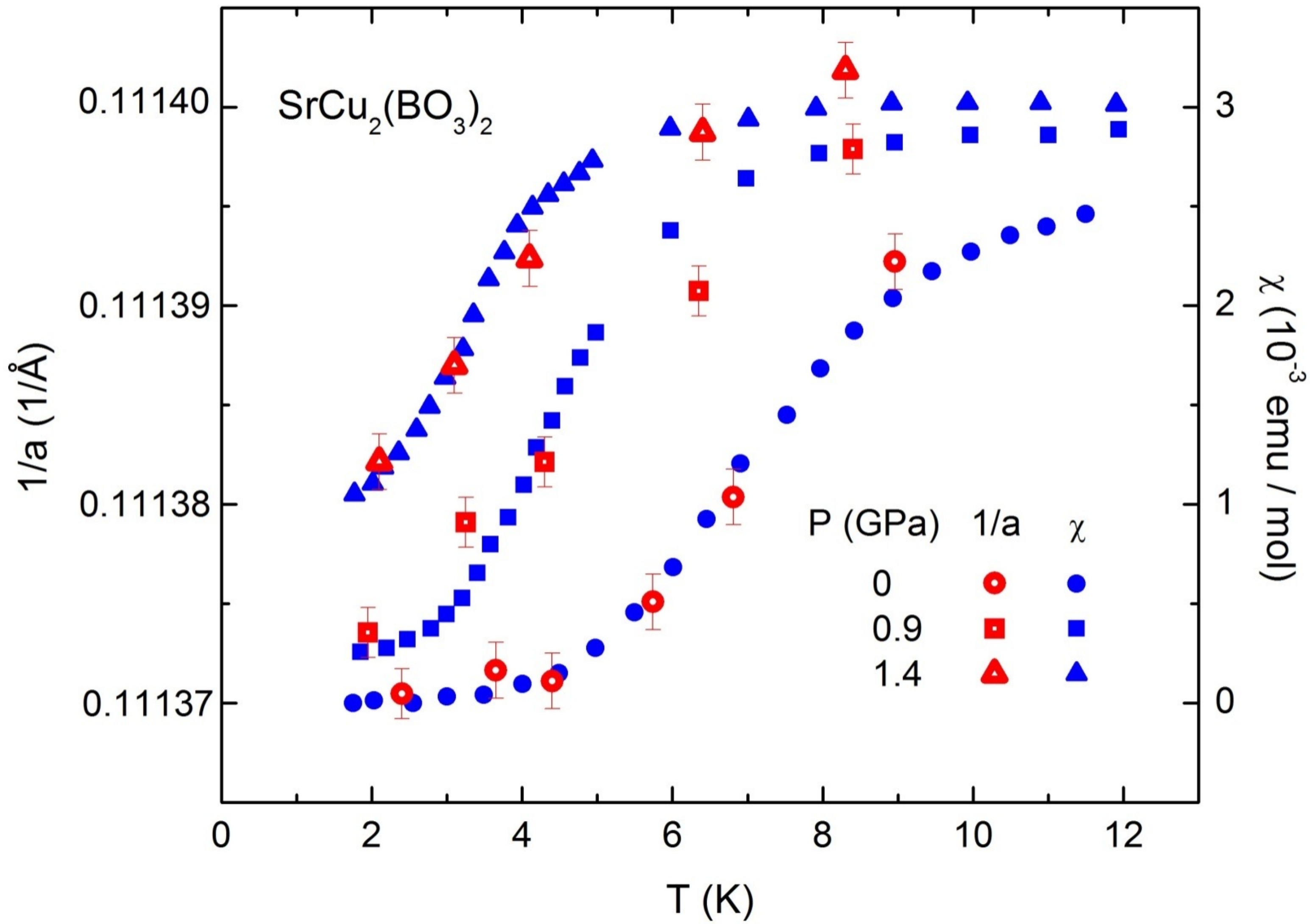
Pressure Tuning of Energy Gap in $SrCu_2(BO_3)_2$



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

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).

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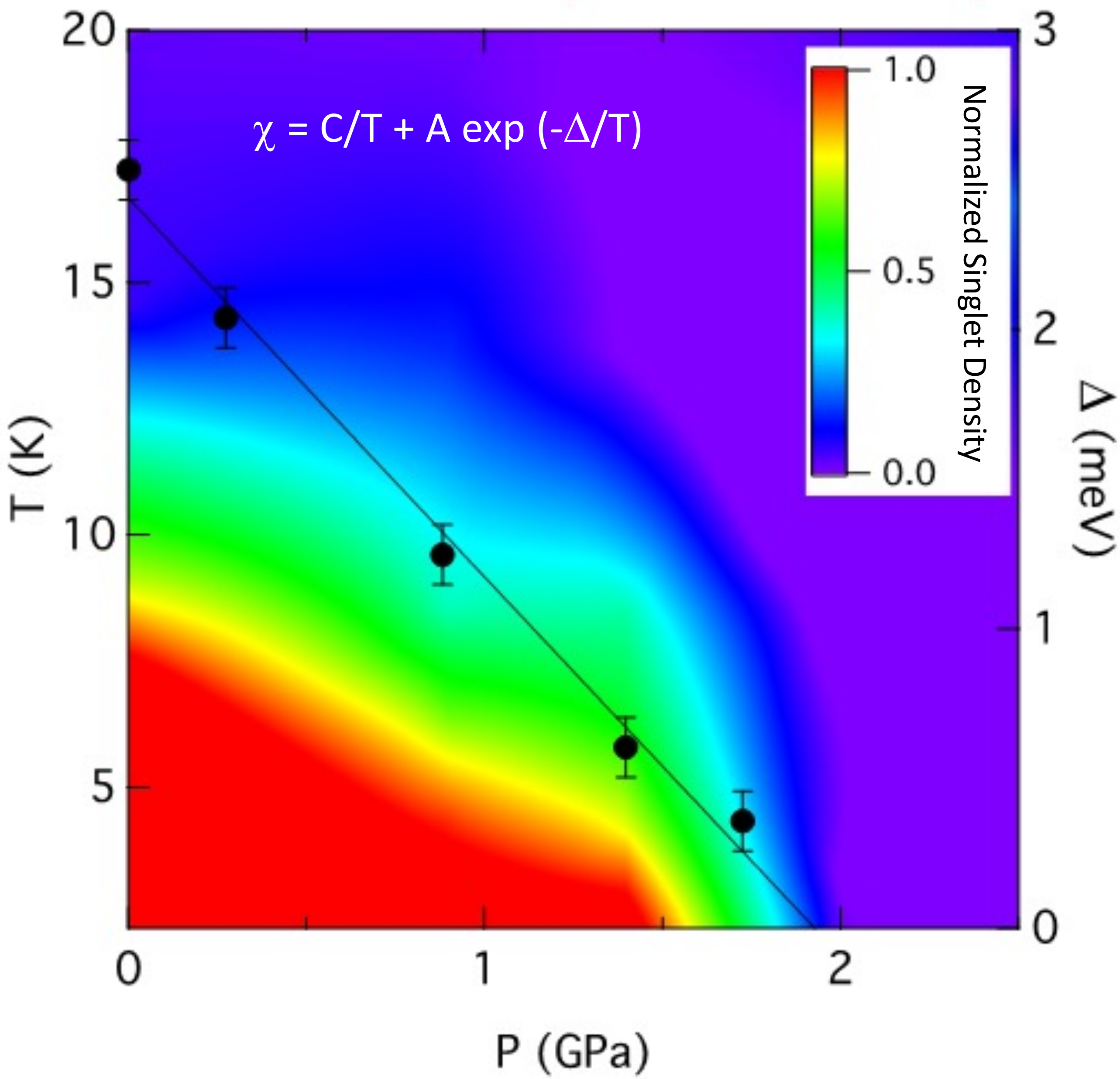
Singlet Density & First Phase Transition



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

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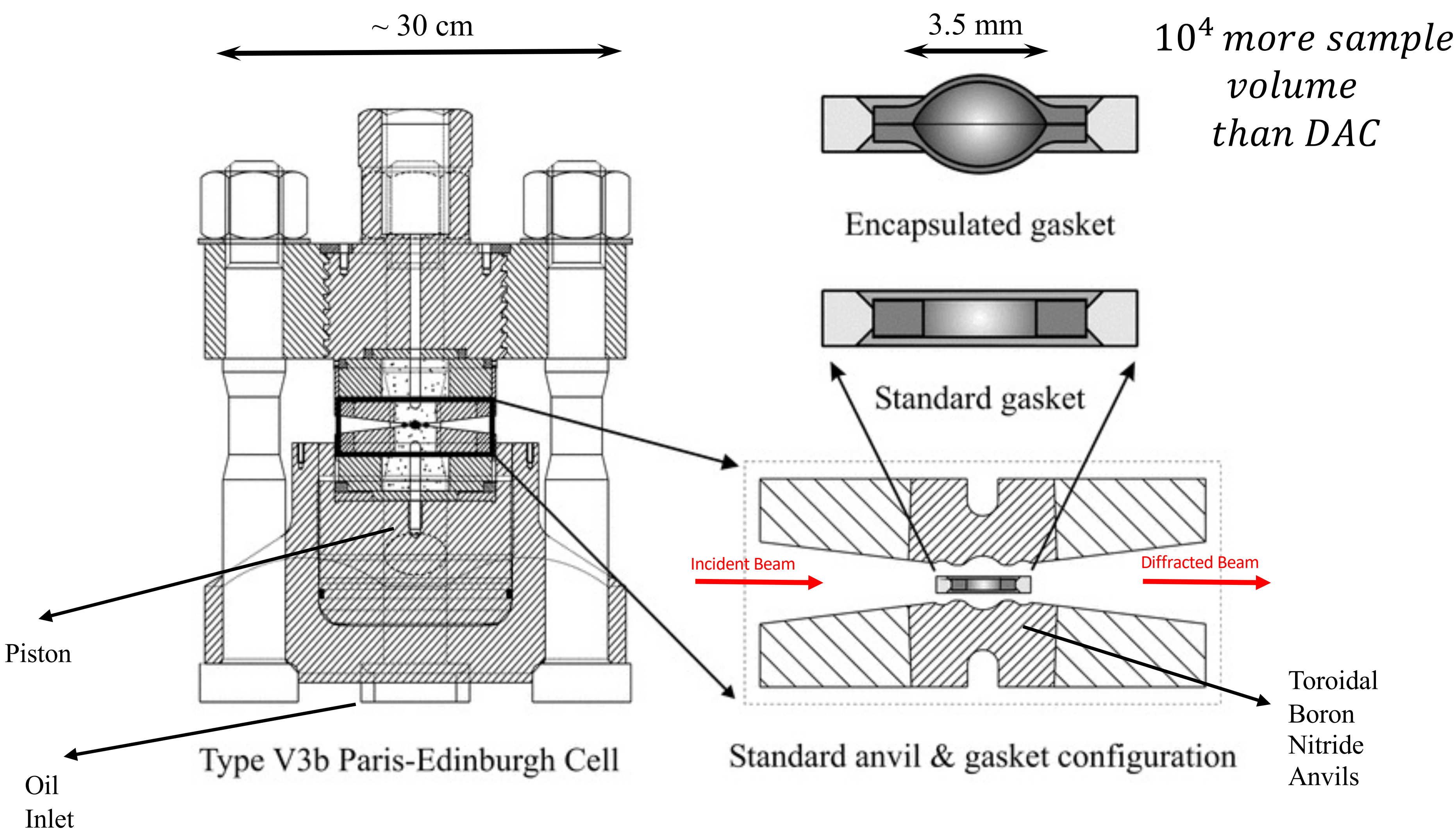
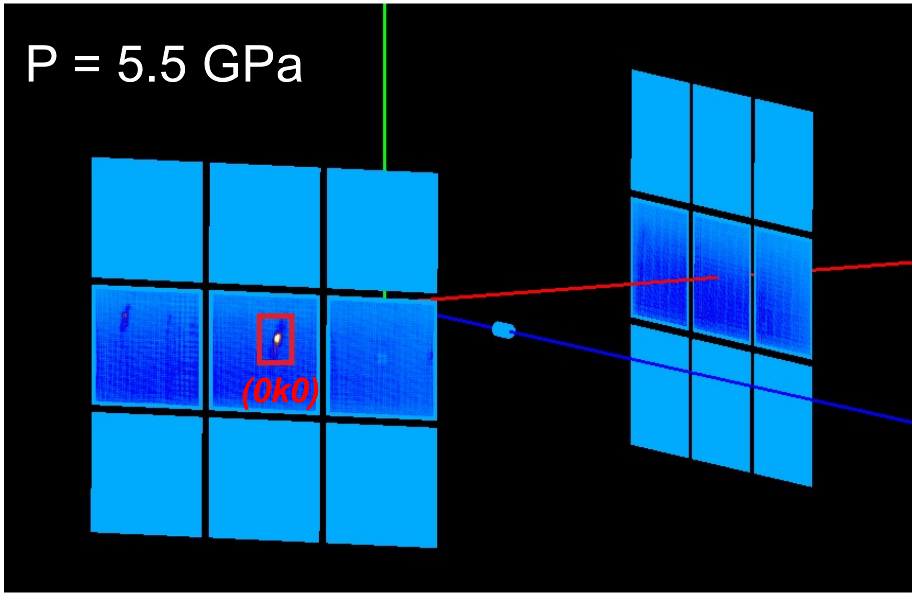
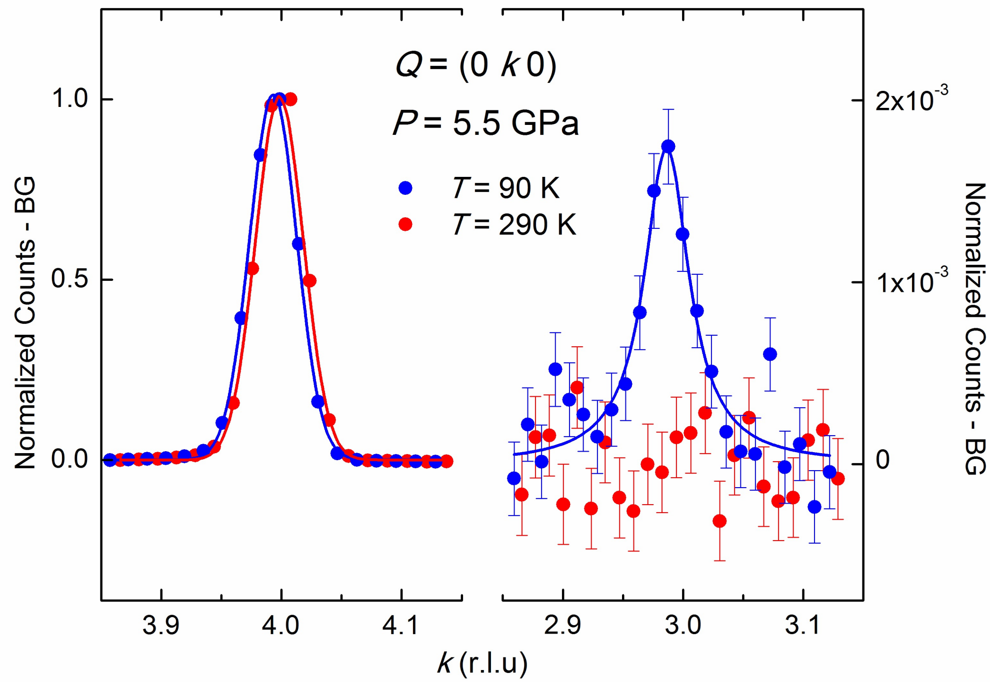
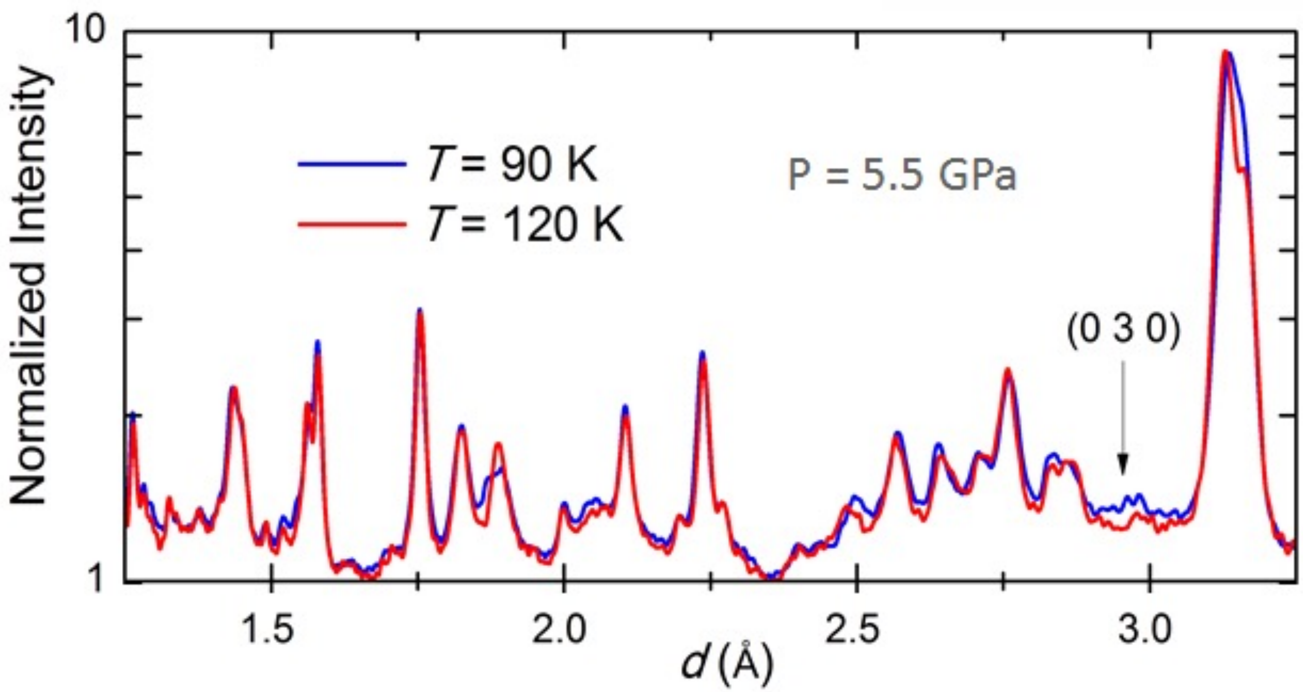
High Pressure Neutron Diffraction Measurements
Paris-Edinburgh Cell



High Pressure Neutron Diffraction Measurements Conducted at SNAP

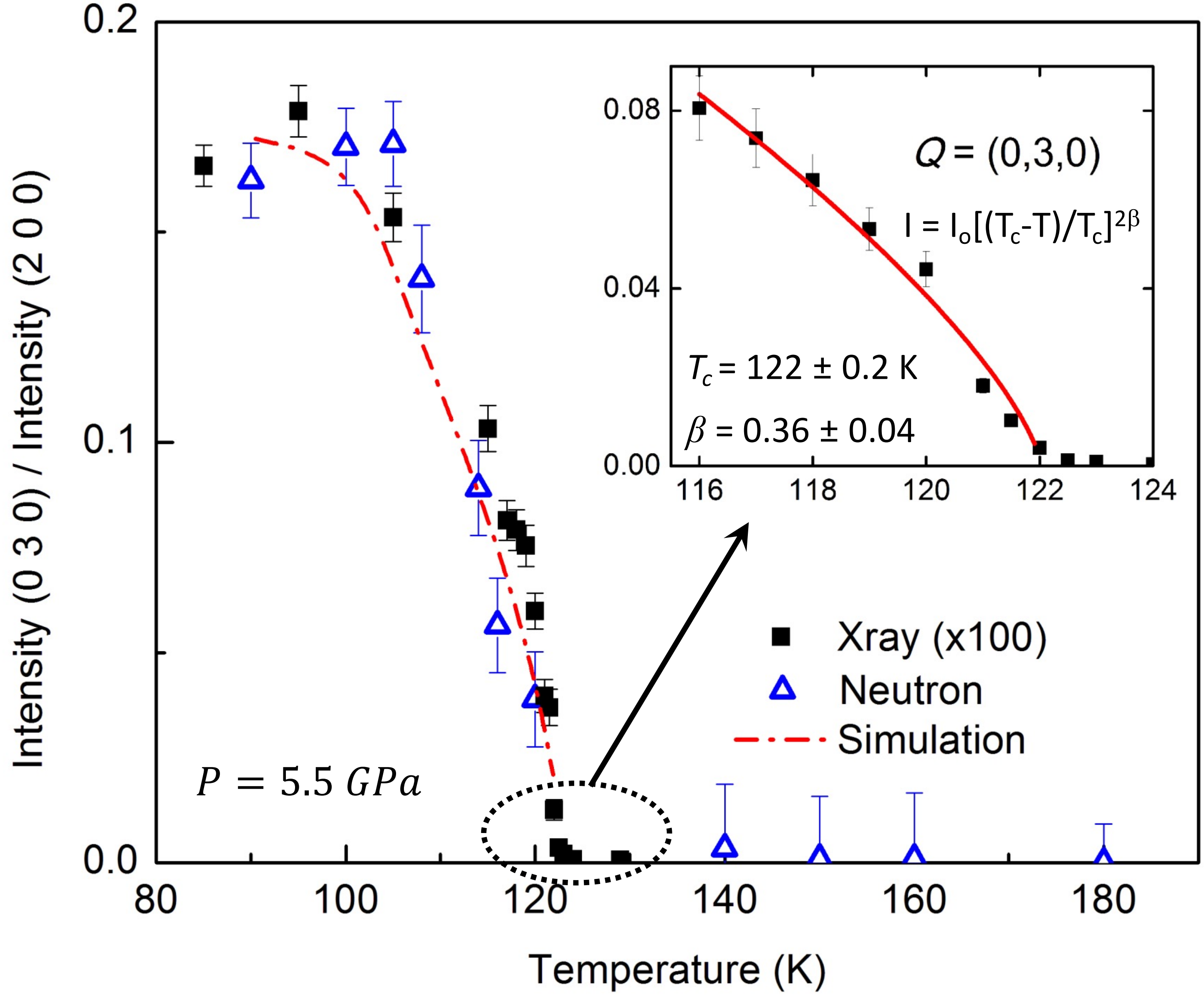






Haravifard *et al.*,
Proc. Natl. Acad. Sci.
111, 14372 (2014).

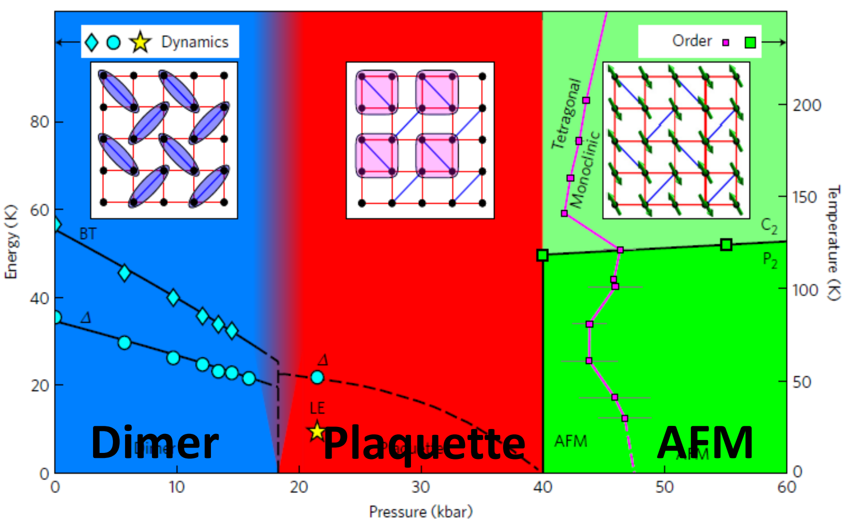
3-Dimensional Nature Shown by Forbidden Peaks and Critical Behavior



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

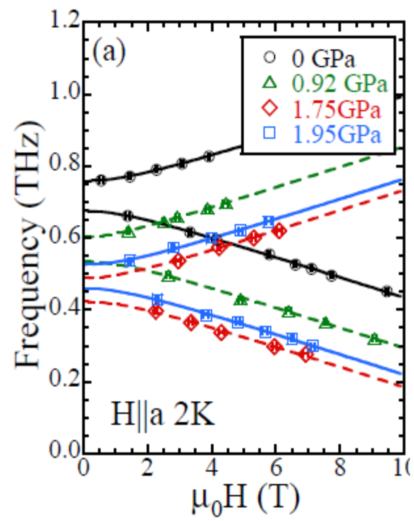
Tuning knob 1: Pressure
Ground states at high pressure

Inelastic neutron scattering



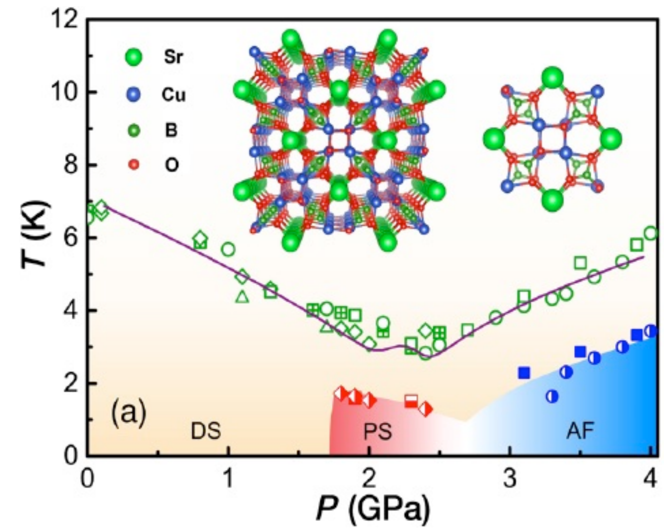
M. Zayed, *et al., Nat. Phys.* **13**, 962 (2017)

Terahertz ESR



T. Sakurai, *et al., J. Phys. Soc. Jpn.* **87**, 033701 (2018)

Specific heat



J. Guo, *et al., PRL.* **124**, 206602 (2020)

Extreme environments needed →

T < ~2 K
P ~ 1.8 GPa: dimer -> plaquette
P ~ 2.5 GPa: plaquette -> AFM

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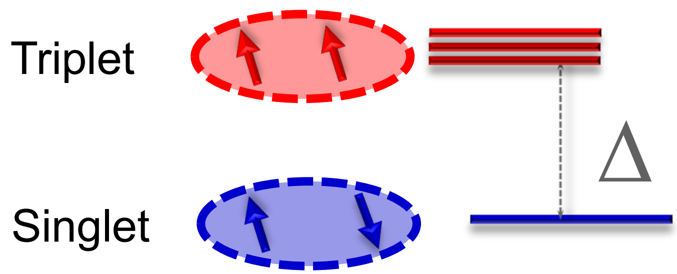
Tuning knob 2: Magnetic field
Magnetization plateaus

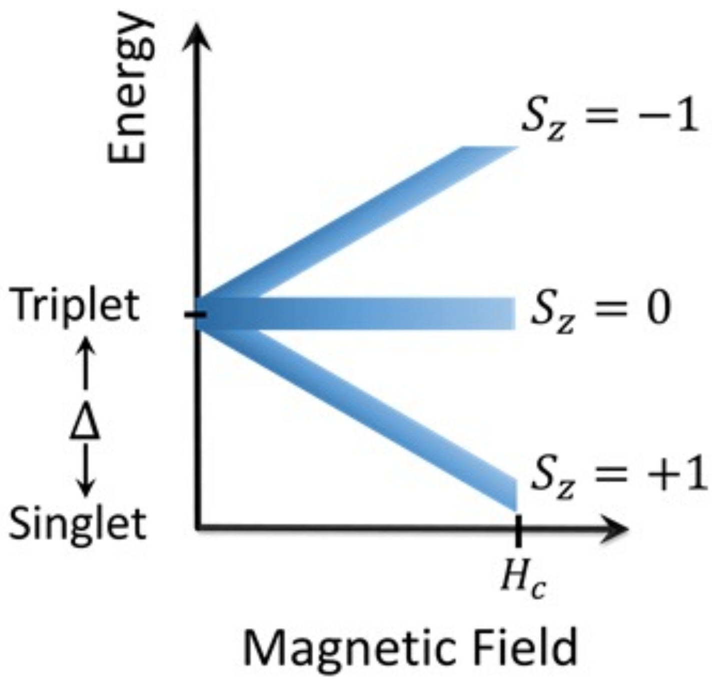
Analogy can be portrayed between a spin ½ dimer system in an external magnetic field and a lattice gas of hard-core bosons.



$S_{tot} = 1$

$S_{tot} = 0$

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



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

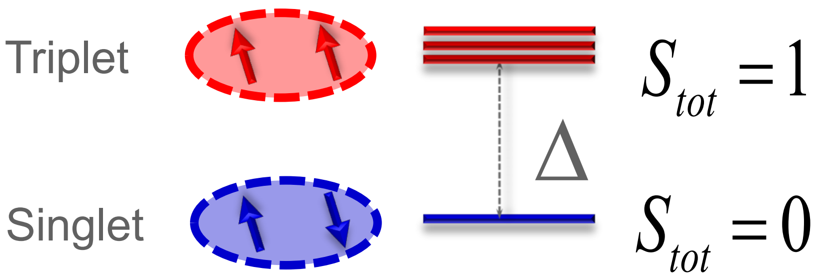
Accordingly magnetic field exceeding H_c is required to create a finite number of triplets per unit cell of the crystal.

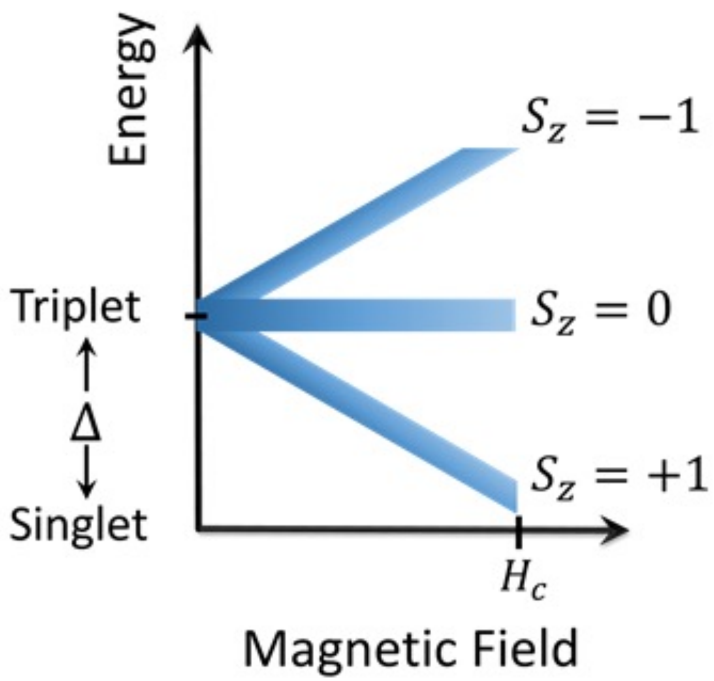
In this depiction:

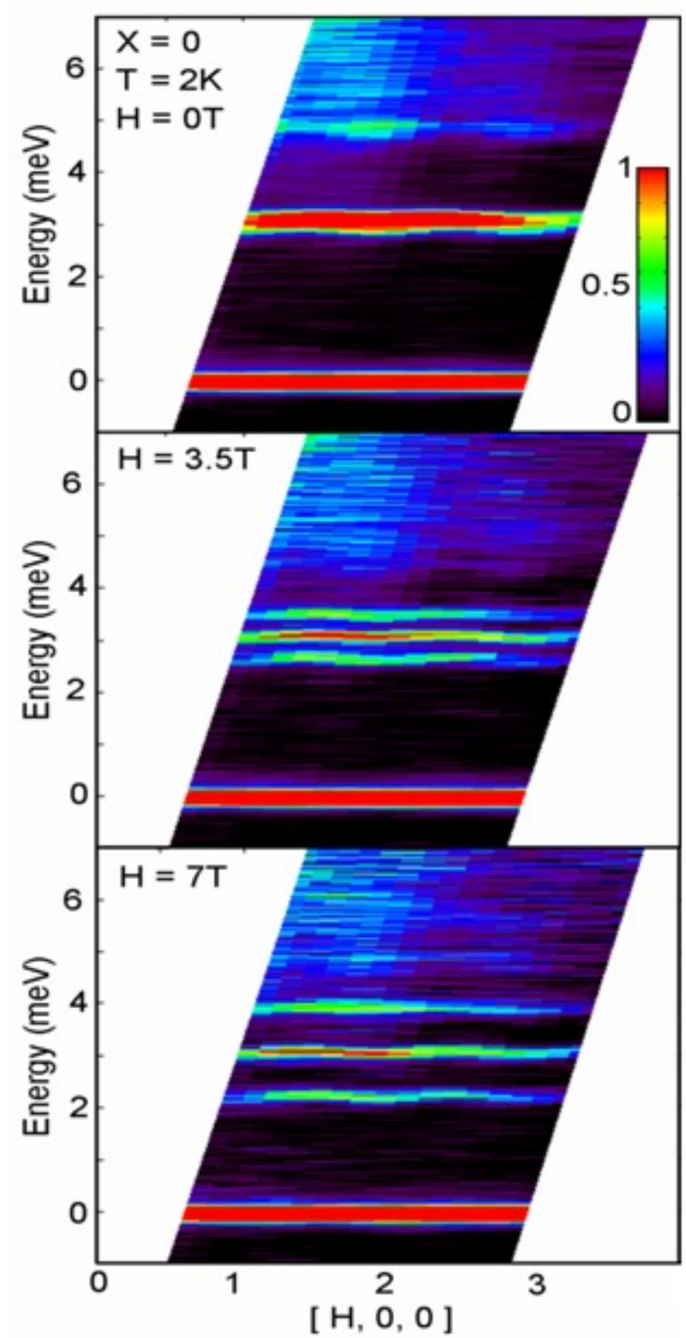
singlet ↔ vacuum
triplets ↔ bosons with hard-core repulsion
magnetic field ↔ chemical potential

Zeeman Splitting of Triplet States in SCBO

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







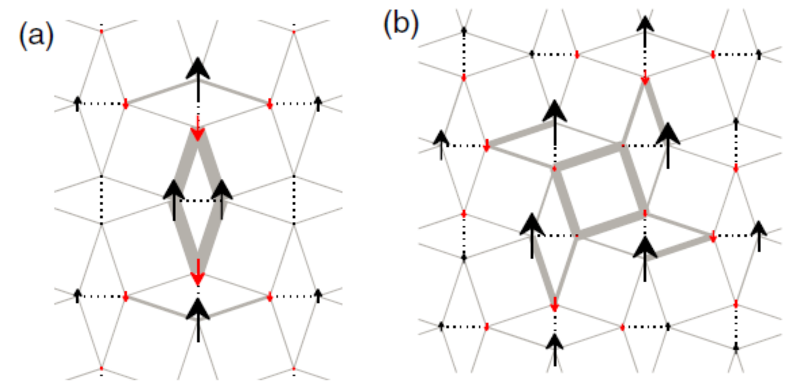
Haravifard *et al.* , *Phys. Rev. Lett.* 97, 247206 (2006).

Mapping the Phase Diagram > Bound States

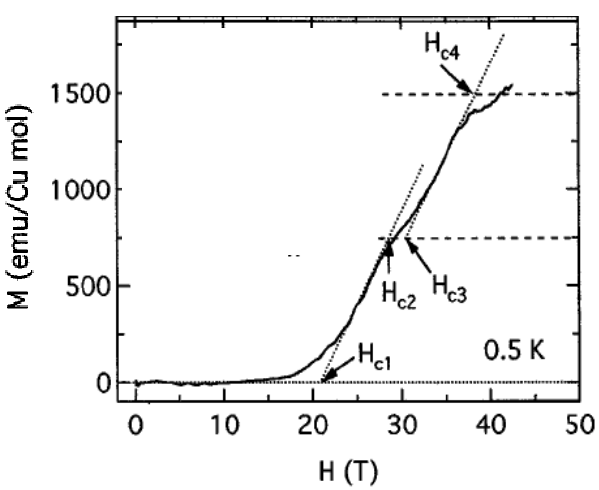
Origin of the <u>magnetization plateaus</u>?

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…

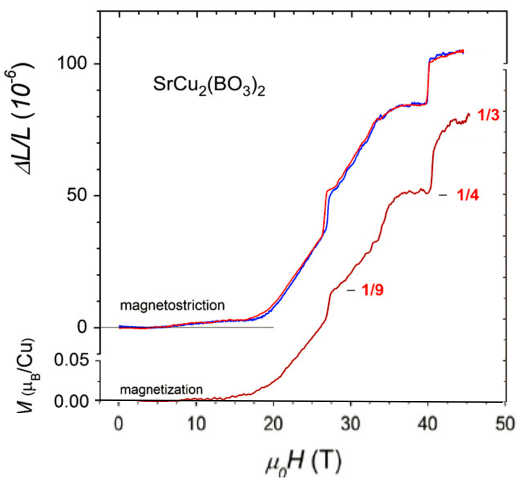
Bound states of triplons (pinwheels): formation of crystals



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)

Extreme environments needed

T < ~2K;
μ_0H >> ~18 T (closing of spin gap)

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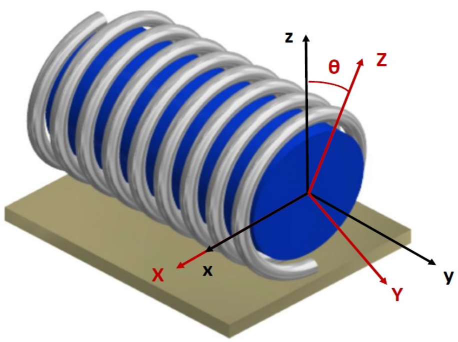
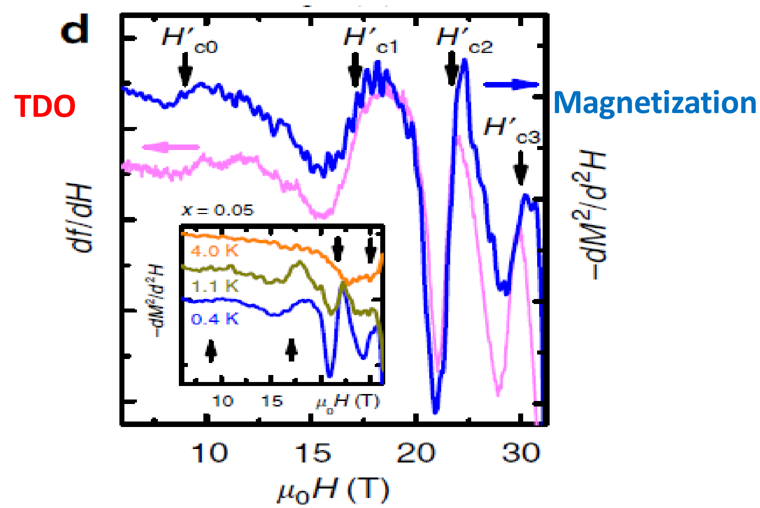
Tunnel Diode Oscillator Technique

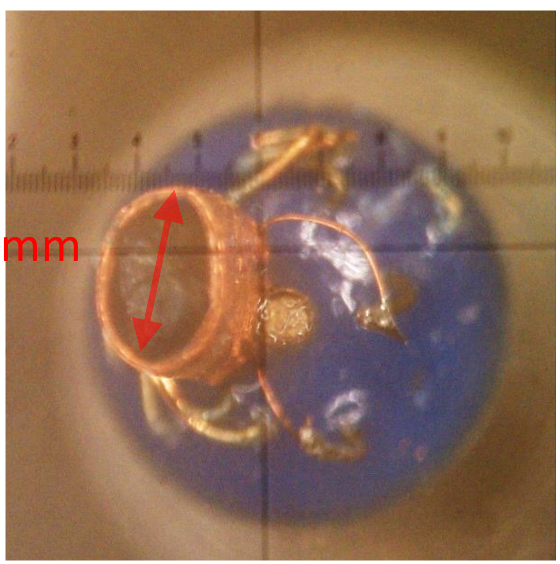
Tunnel Diode Oscillators (TDO) represent a new approach to this challenge. A TDO circuit is constructed of a self-resonating *LC* tank biased by a tunnel diode. Sample is located in a coil which acts as the inductor in the *LC* tank circuit

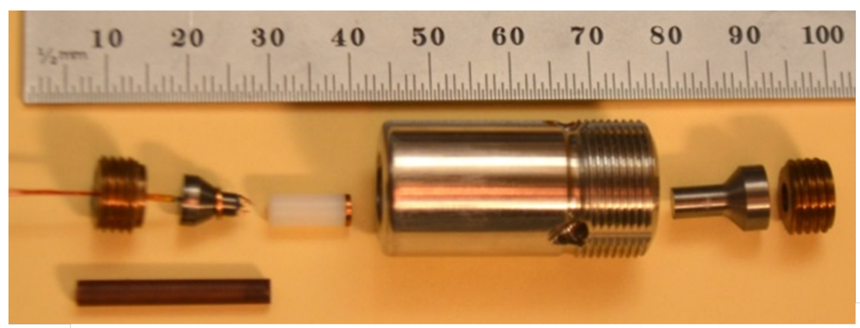
→ $\frac{\Delta f}{f} \propto \frac{dM}{dH}$ enabling us to precisely measure changes in the magnetic moment on the order of **10^{-12} emu**

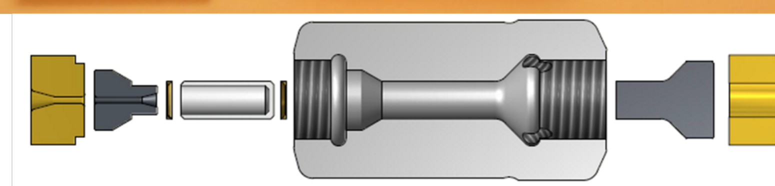




~ 1.5 mm

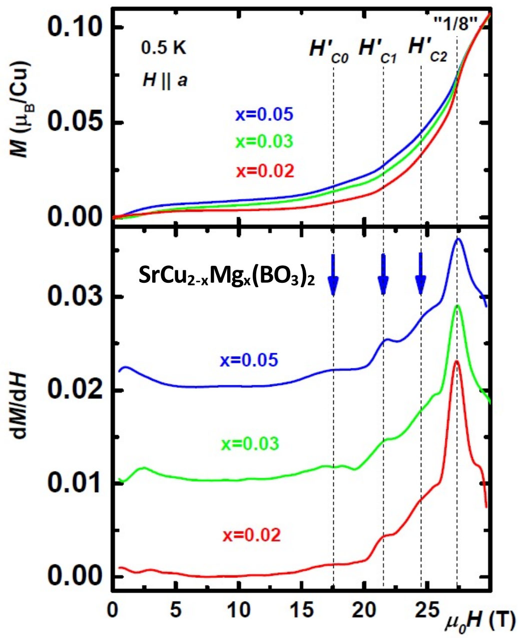




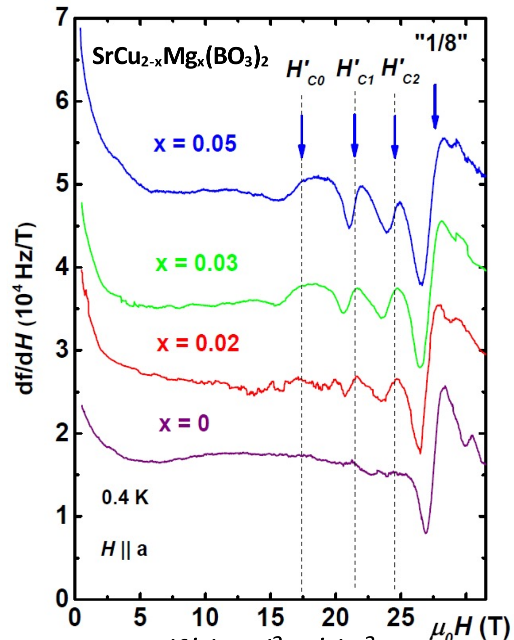


Tuning knob 3: Chemical Doping
$SrCu_{2-x}Mg_x(BO_3)_2$: Anomalies below 1/8 plateau

Magnetization measurement

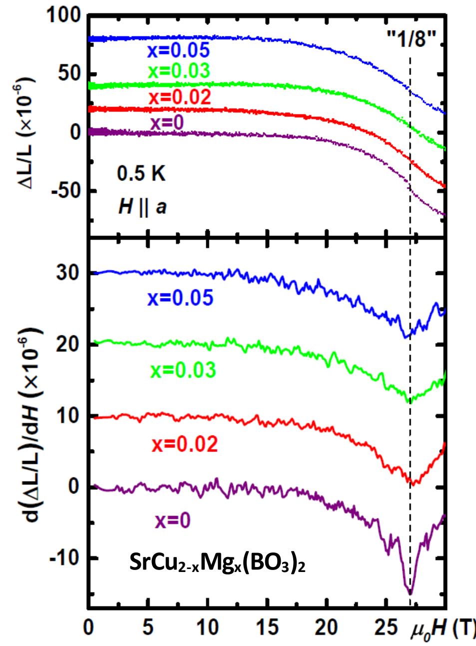


Tunnel diode oscillator (TDO)



$df/dH \sim d^2M/dH^2$

Magnetostriction



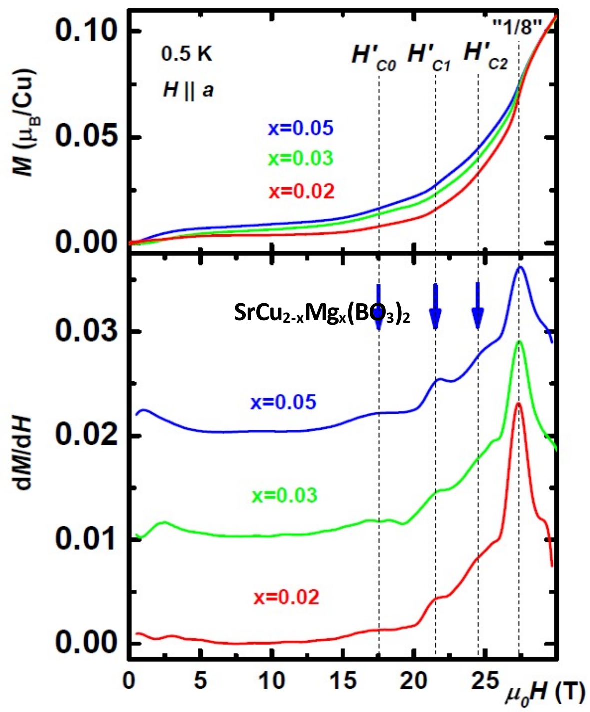
- **Emergent magnetic states at H'_{c0}~17.5 T, H'_{c1}~21.5 T and H'_{c2}~24.5 T**
- **Absence in magnetostriction: weak lattice coupling**
- **Superstructure of triplets with unit cell larger than that for 1/8? Not likely**

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

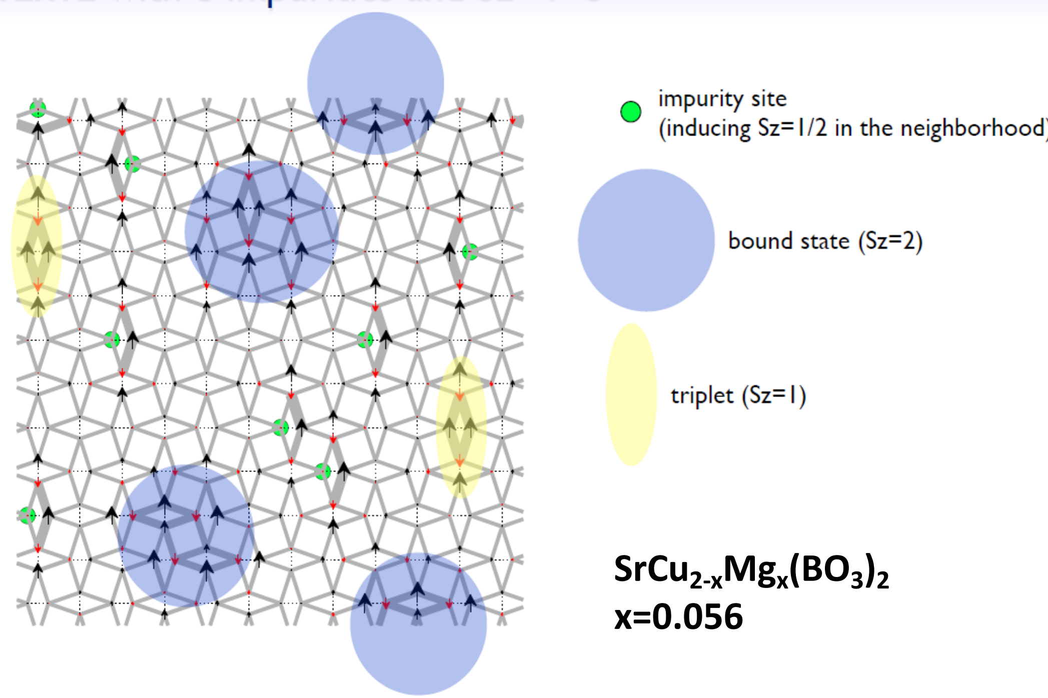
Shi, Zhenzhong, *et al*. "Emergent Bound States and Impurity Pairs in Chemically Doped Shastry-Sutherland System." Nature Commun. 10, 2439 (2019).

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

Magnetization measurement



12x12 with 8 impurities and Sz=4+8



$SrCu_{2-x}Mg_x(BO_3)_2$
x=0.056

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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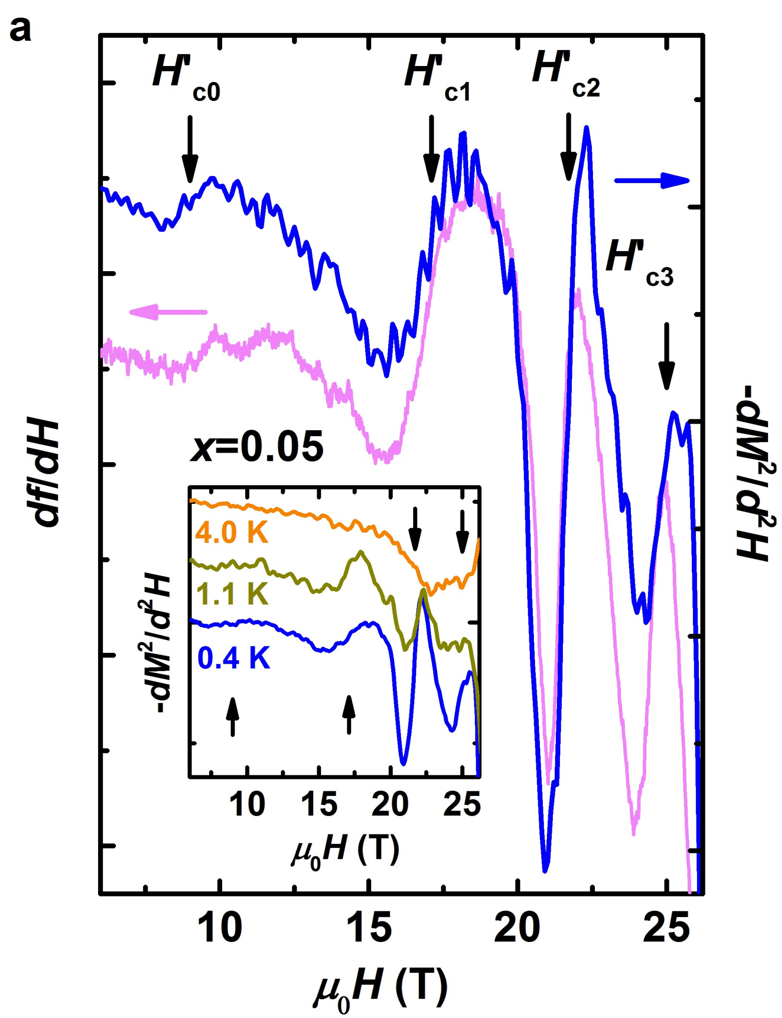
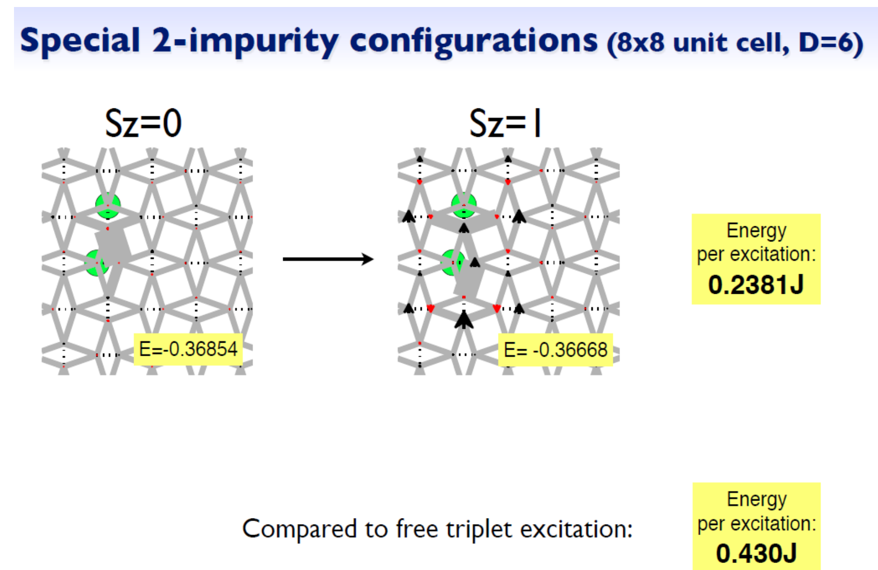
Special Configuration of impurities

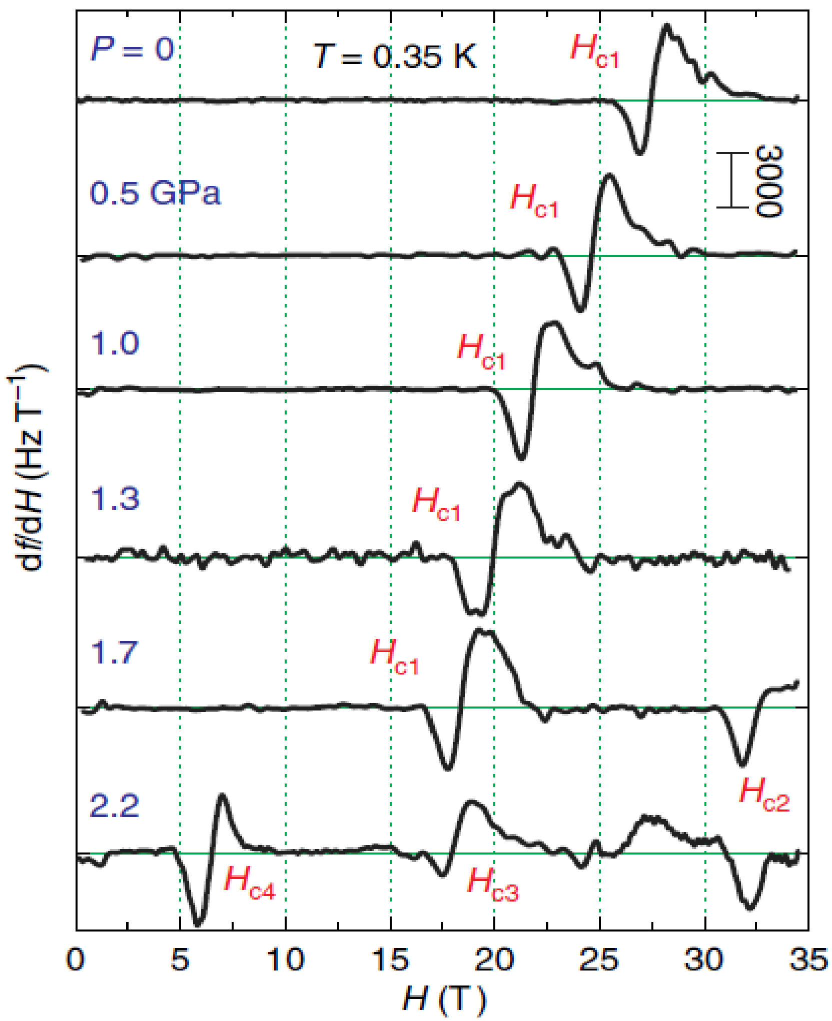




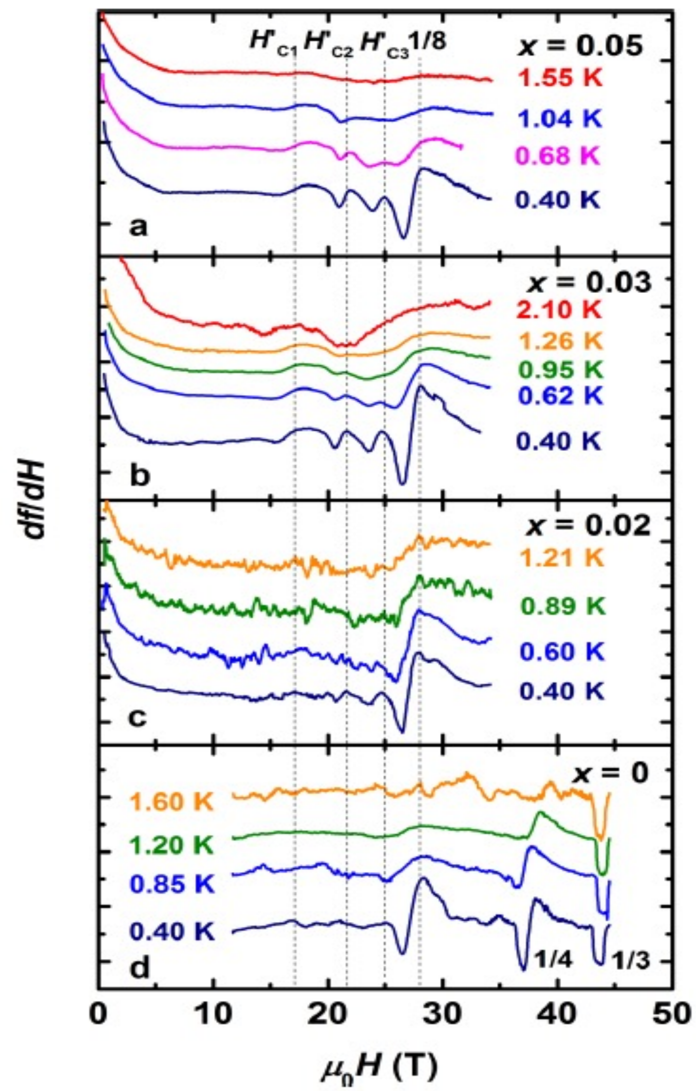
Better answer: Bound states of impurities

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

Utilizing all 3 Tunning Knobs



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



Shi, Zhenzhong, *et al*. Nature Commun. 10, 2439 (2019).

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Conclusions

❖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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