Entropy near the magnetization plateau of $SrCu_2(BO_3)_2$

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THE UNIVERSITY OF TOKYO

QMEC, March 23rd, 2021

The University of Tokyo
The Institute for Solid State Physics

IMGSL at ISSP

Pulsed high magnetic field laboratory
(International MegaGauss Science Laboratory)

Kindo group

1200 T generation & many condensed matter physics.

Kohama group

Matsuda group

Tokunaga group

YK is dedicated to technical developments.

1. Field generation (RSI 2015, 2021 & 2021)
2. Magnetocaloric effect (RSI 2010 & 2013)
3. Specific heat(RSI 2010, 2021 & MST2013)
4. Thermal conductivity (in preparation)
5. Pulsed high-field NMR (in preparation)
6. Neutron diffraction (ongoing project)

Outline

- Introduction

Past experimental and theoretical researches in $SrCu_2(BO_3)_2$

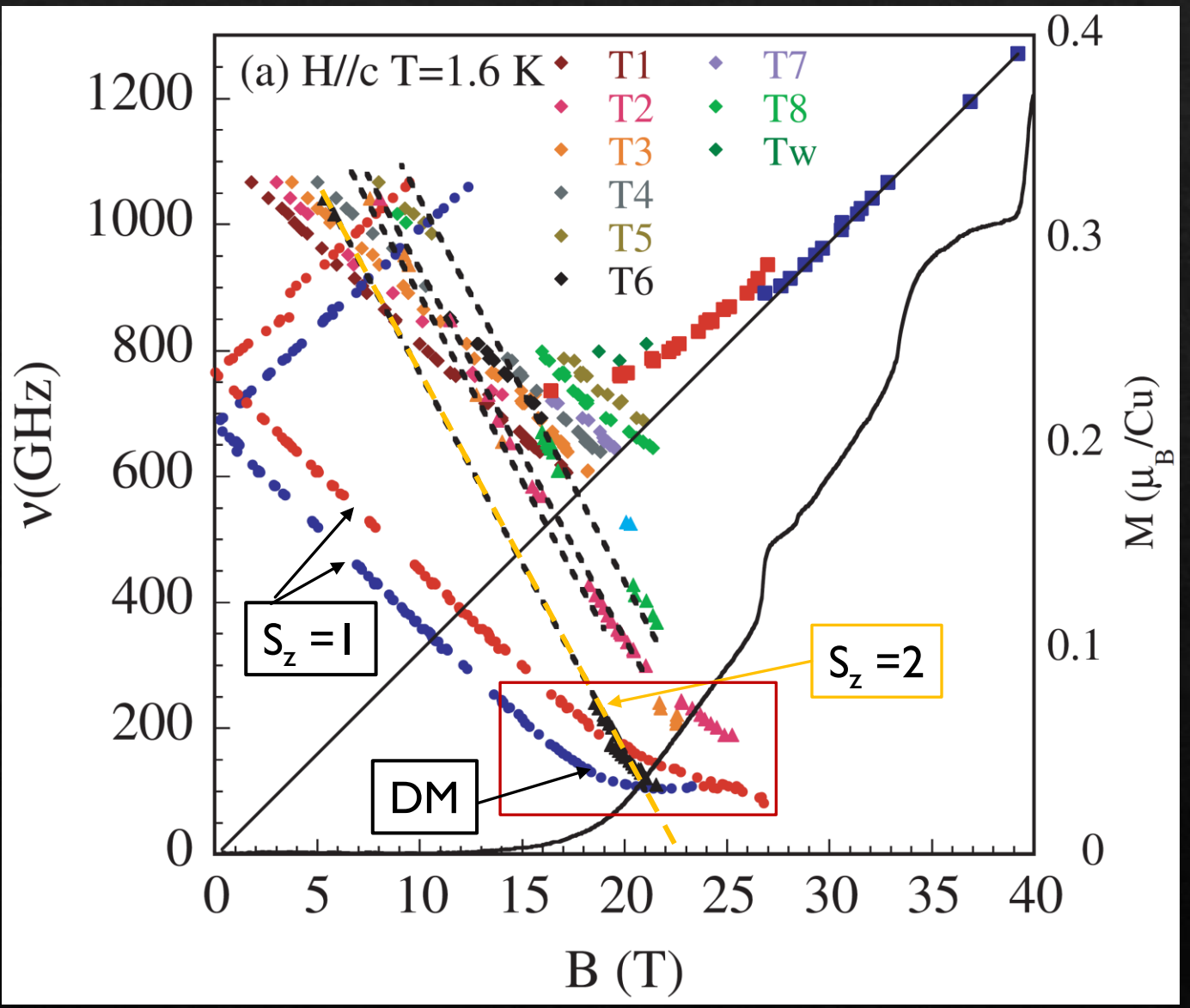
- Experiments

Magnetocaloric effect (MCE) in adiabatic condition

Specific heat in pulsed and static high-magnetic fields

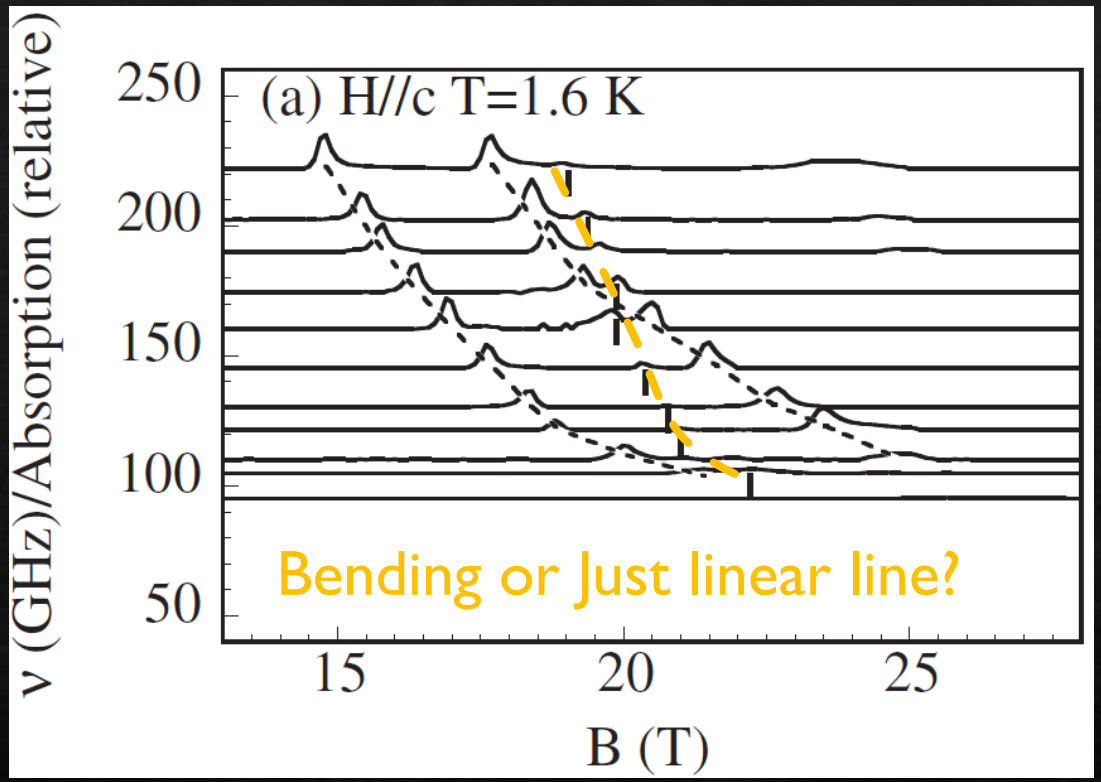
- Result & Discussion

ESR data for H//c



H. Nojiri, et al., *JPSJ* **72**, 3243 (2003)

$S_z = 2$ (spin-2 bound state) was found



No useful data below 100 GHz (~4.8 K)

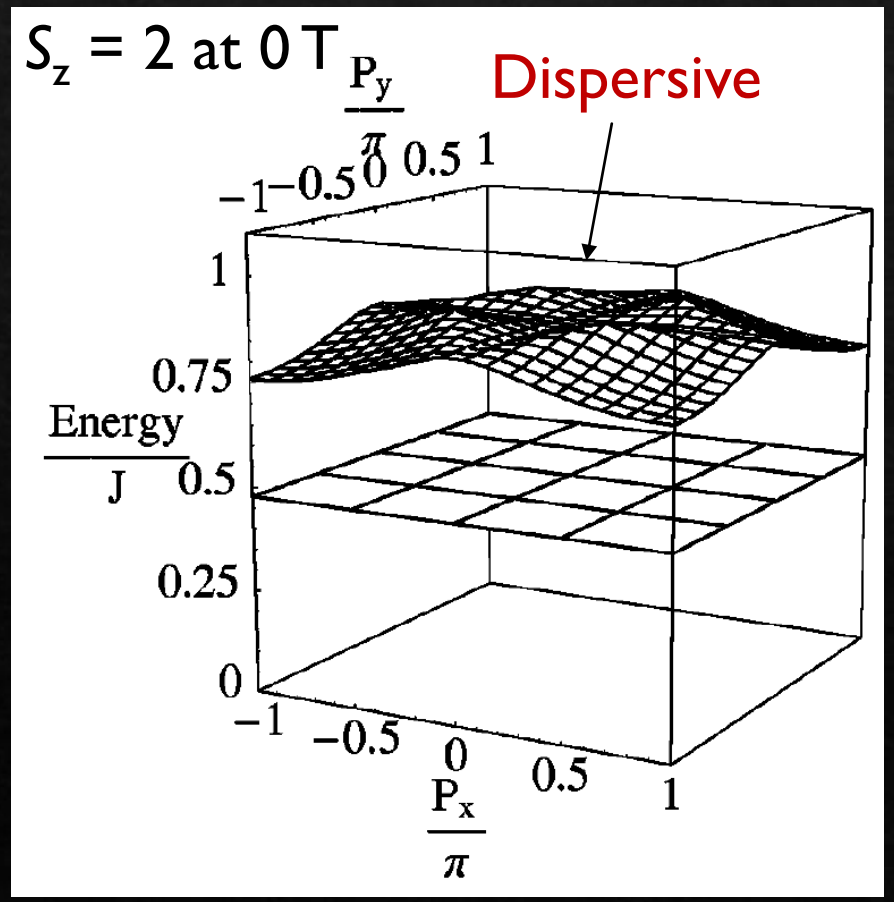
Does $S_z = 2$ become the ground state above ~23T? (linear approximation)

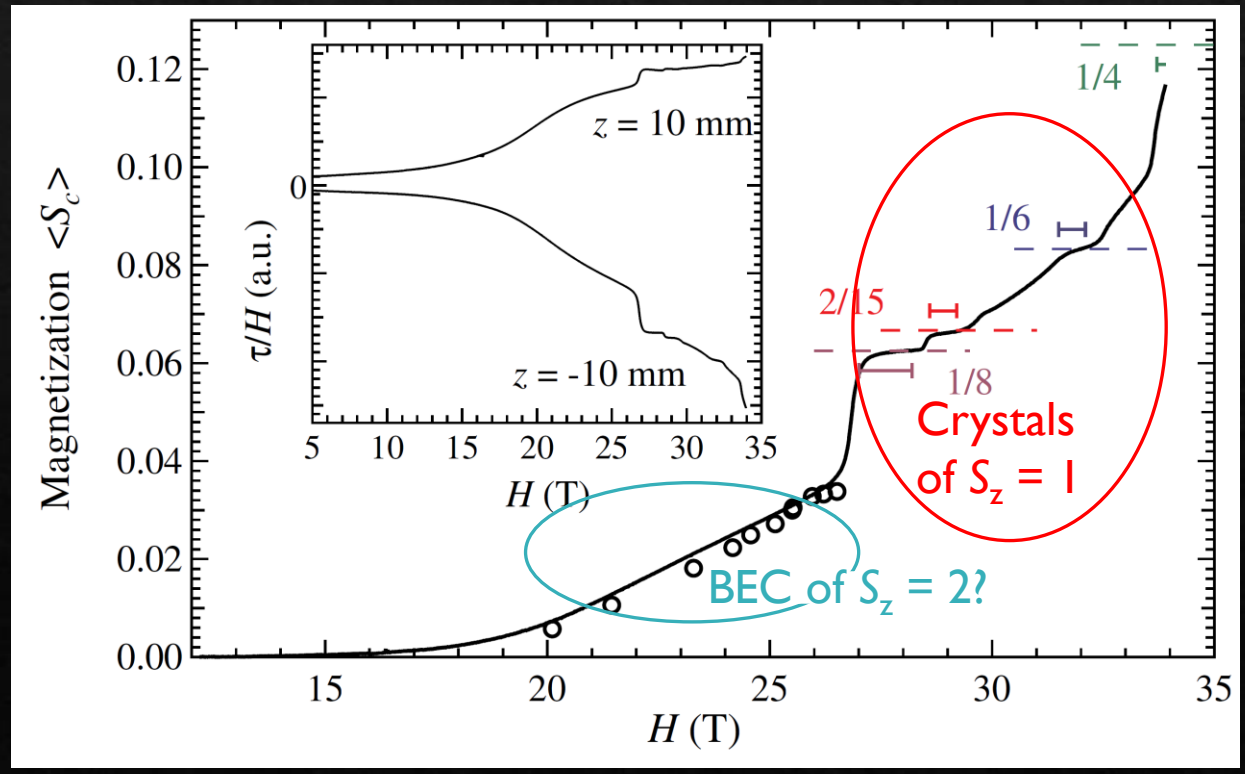
$SrCu_2(BO_3)_2$; The possible BEC of $S_z = 2$

The $S_z = 2$ might condense below 27 T, instead of triplon ($S_z = 1$).

field. By increasing the magnetic field more than the critical field, a macroscopic number of bound states condense instead of the single dimer triplets. Thus the non-plateau state at very low magnetization may be a superfluid of bound states and is different from the almost localized dimer triplet state discussed by Miyahara and Ueda.[16] The critical mag-

T. Momoi and K. Totsuka *PRB* **62**, 15067 (2000)

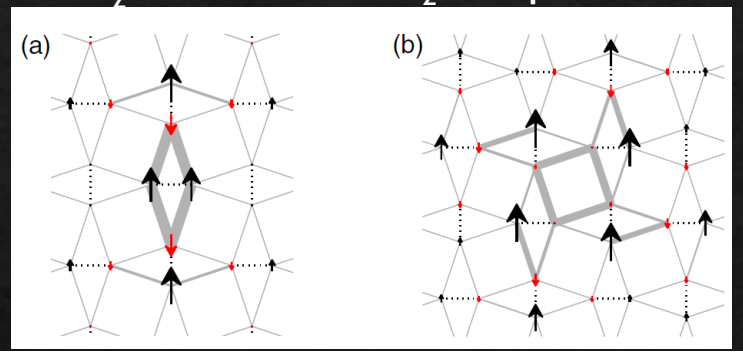


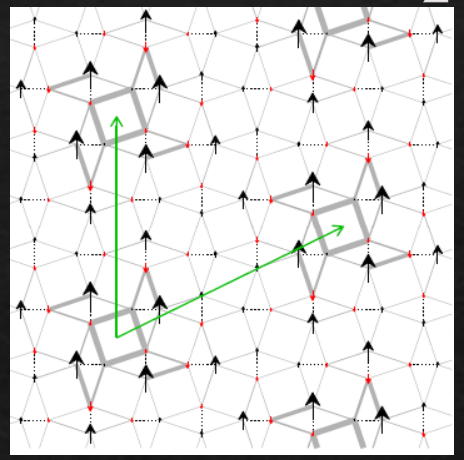


M. Takigawa, et al., PRL 110, 067210 (2013)

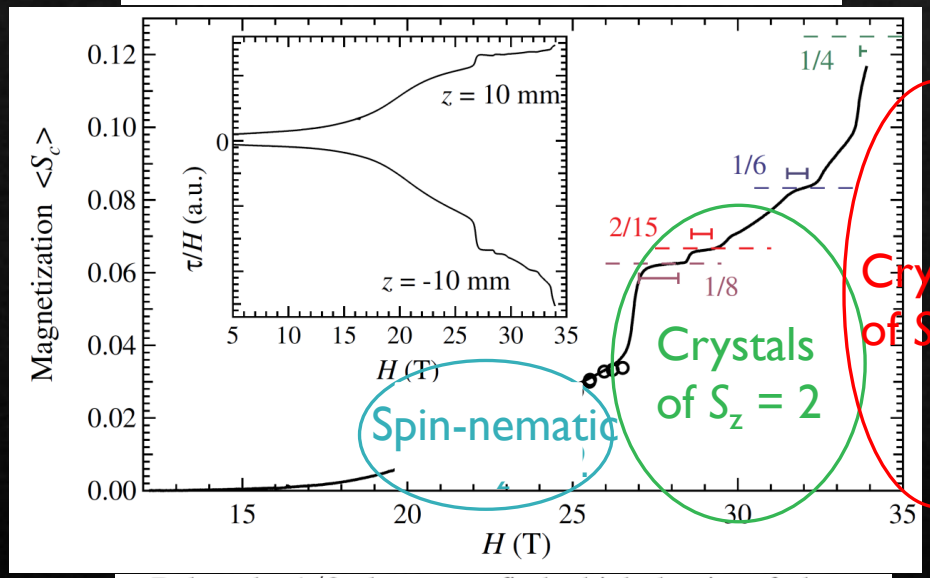
Condensation/crystallization of $S_z = 2$ in $SrCu_2(BO_3)_2$

$S_z = 1$ $S_z = 2$ pinwheel





1/8 plateau state
Quintuplet crystal

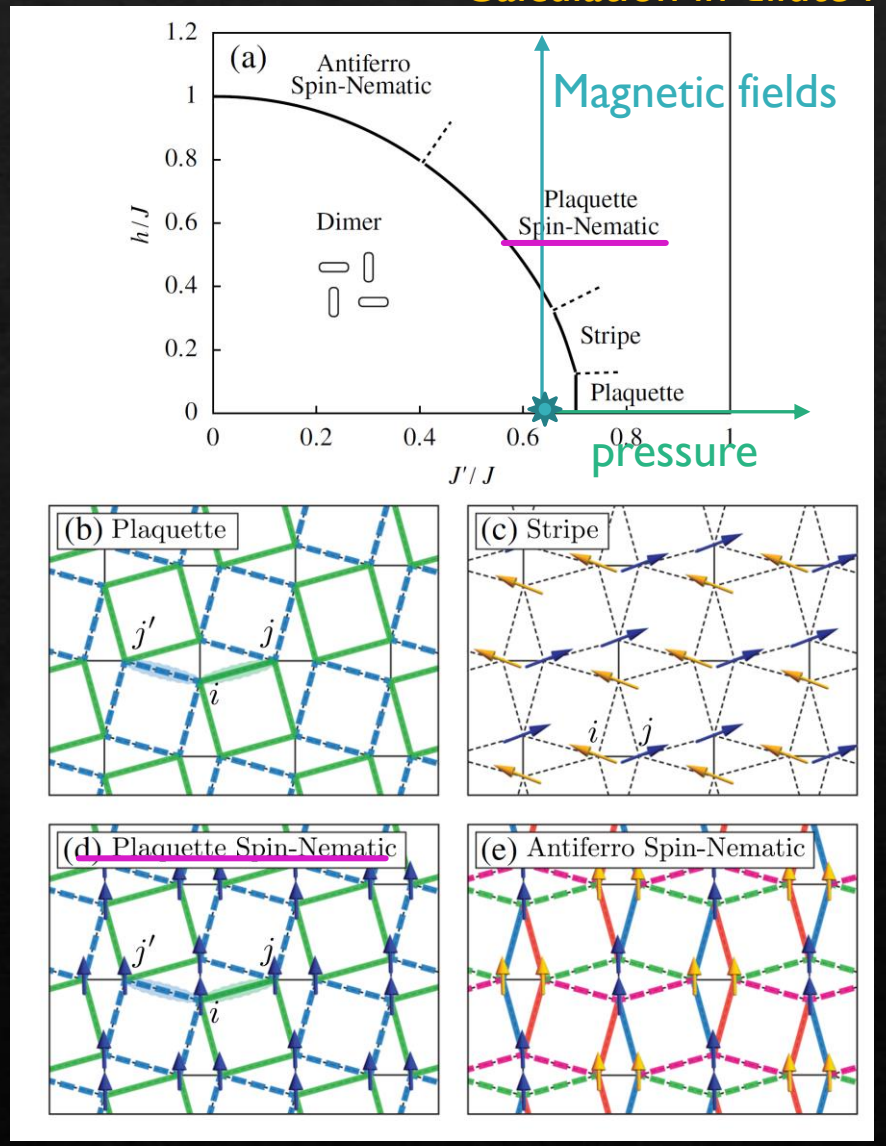


Below the 1/8 plateau we find a high density of plateaus which lie energetically very close, including plateaus at 1/12, 1/11, 1/10, 2/19, 1/9, see Fig. 4 and [44] (the 2/17 plateau lies slightly higher in energy). At even lower fields we enter the dilute region of bound states, where they start to delocalize (and eventually Bose condense). We did not study this region [marked by a dashed line in Fig. 4(b)] in detail.

Philippe Corboz and Frederic Mila, *PRL* **112** 147203 (2014)

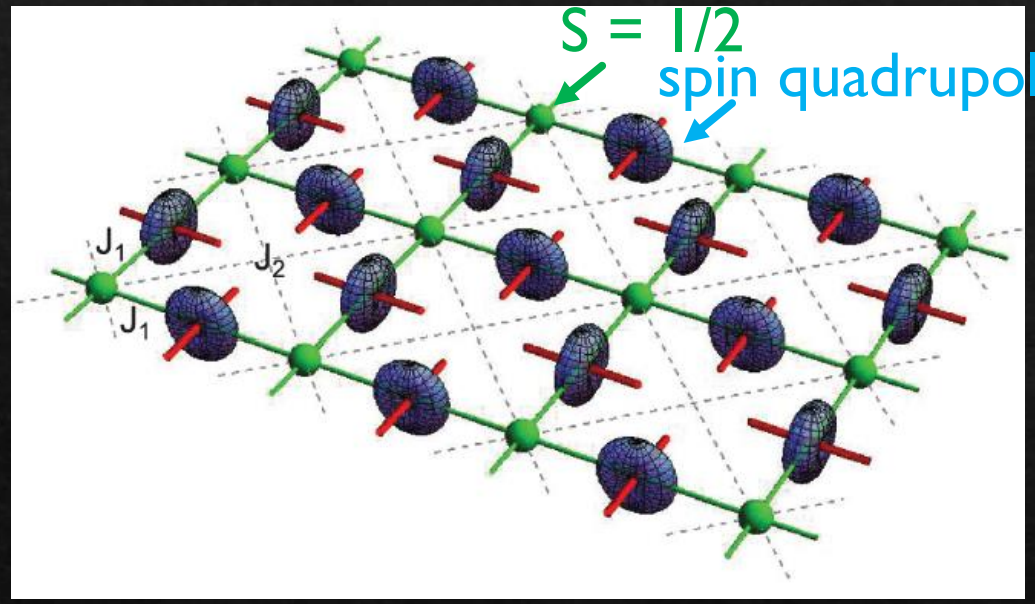
* Super solid (SS) and domain wall (DW) state were also predicted.

Calculation in dilute region



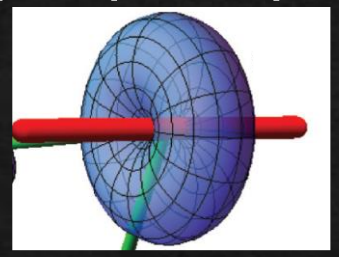
Zhentao Wang and Cristian D. Batista, *PRL* **120** 247201 (2018)

"Orthodox" Spin nematic (SN) states



Spin nematic (spin quadrupole order) in square lattice

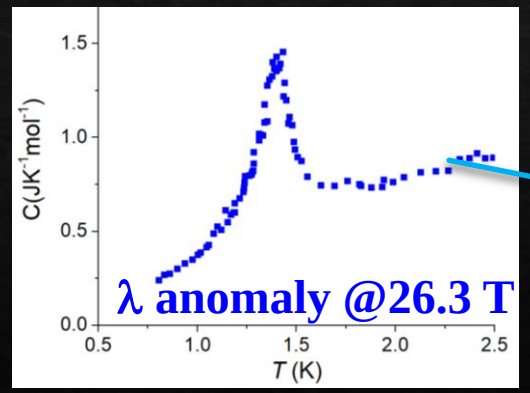
Order parameter
; spin quadrupole



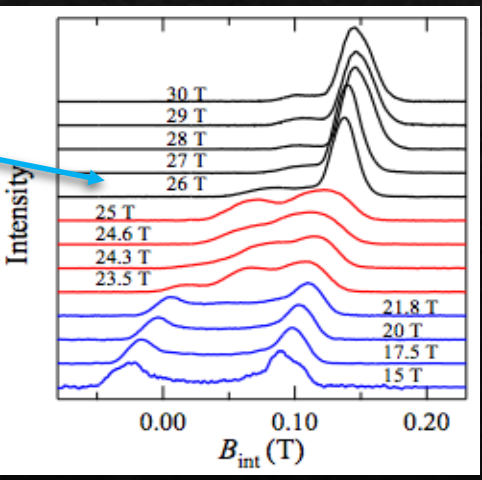
No internal field
⇒Hidden order

NMR, μSR, neutron scattering, and magnetization **cannot** detect the SN phase.

Volborthite



YK et al., *PNAS* **99**, 024413 (2019)



H. Ishikawa et al., *PRL* **114**, 227202 (2015)

Entropy is sensitive to any type of ordering, including SN. (e.g., Ferroelectric transition & Hidden orders ;URu_2Si_2 and volborthite)

Purpose

❖ To observe the possible SN phase below the 1/8 plateau.

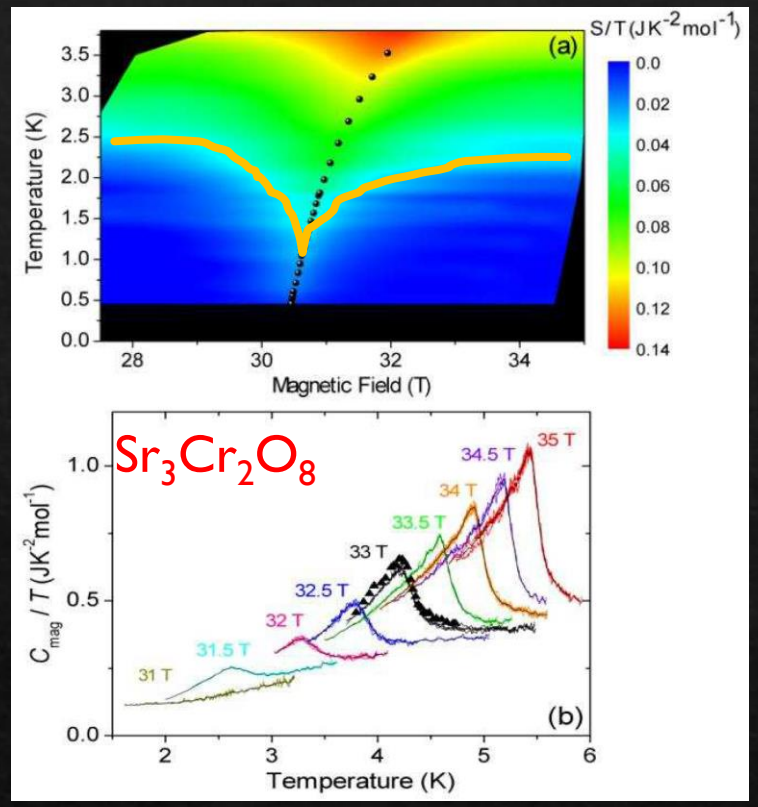
*We need to check whether $S_z = 2$ is a ground state above ~23T.

❖ To map out the high-field (*H*,*T*) phase diagram.

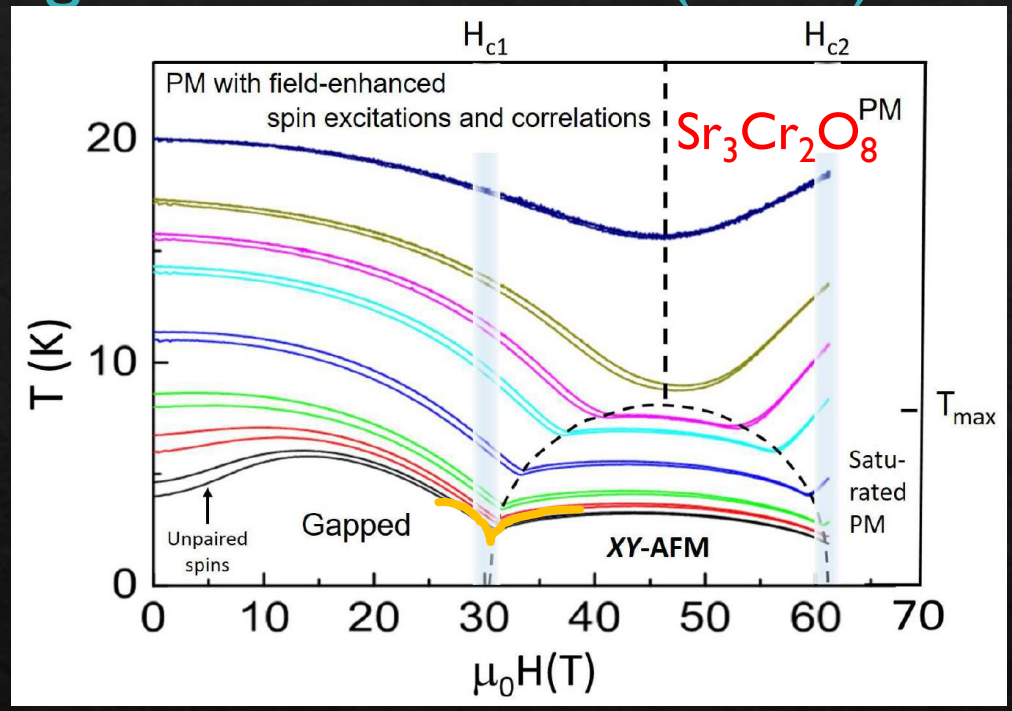
*Experimental (*H*,*T*) phase diagram has not reported above ~33T.

Experiment

Magnetocaloric effect (MCE) in adiabatic condition



A. Aczel et al., *PRL* **103**, 207203 (2009)



T. Nomura et al., *PRB* **102**, 165144 (2020)

T. Kihara *et al.*, *RSI* **84**, 074901 (2013).

T(*H*) curve
in adiabatic condition.
⇕
Isentropic *T*(*H*).

Yasuhiro Matsuda

ISSP

Subaru Akimoto

Measurement
& analysis

Hiroshi Kageyama

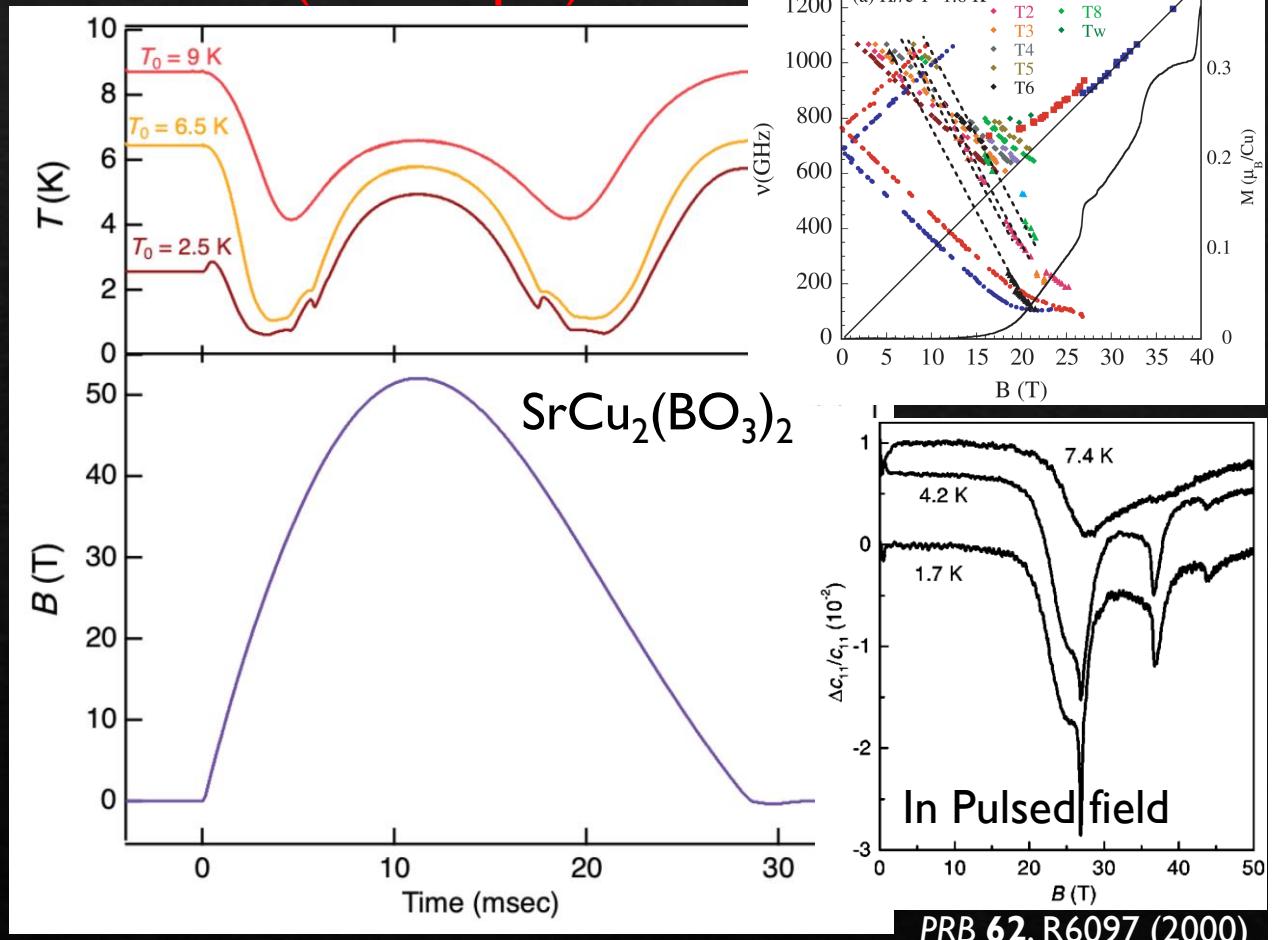
Kyoto University

$SrCu_2(BO_3)_2$ single crystal

MCE in adiabatic condition (*H* // *c*)

Short time *T*(*H*) measurement in vacuum

⇒ Adiabatic (isentropic) MCE



PRB **62**, R6097 (2000)

Sorry I have masked this part, since we are still preparing the draft.

We will submit the manuscript in near future and will show the data.

- Small entropy in 1/3 and 1/4 plateaus indicate the existence of gap.
- SS state has larger entropy than the 1/4 plateau (due to magnon?).

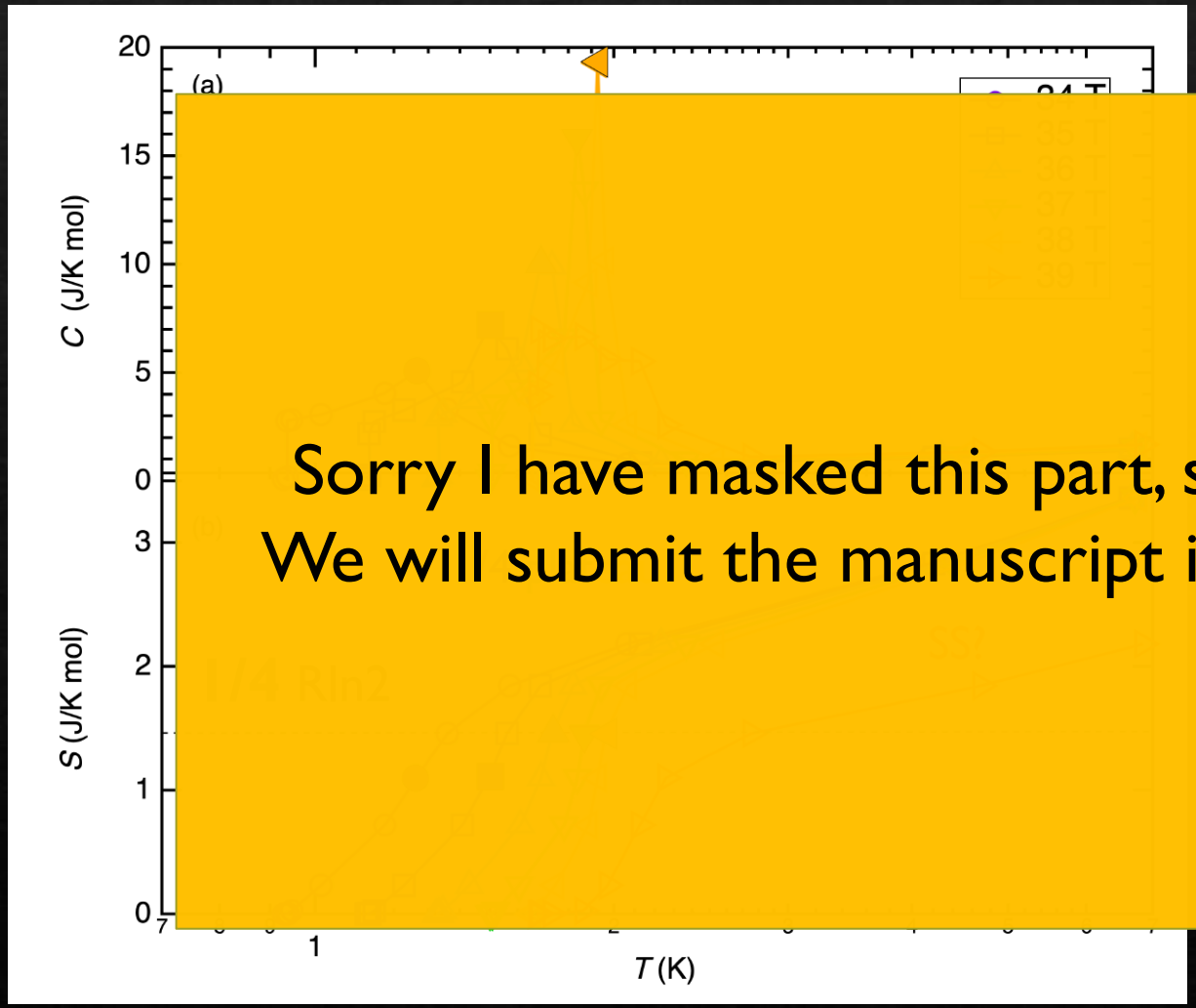
1/8, DW?,

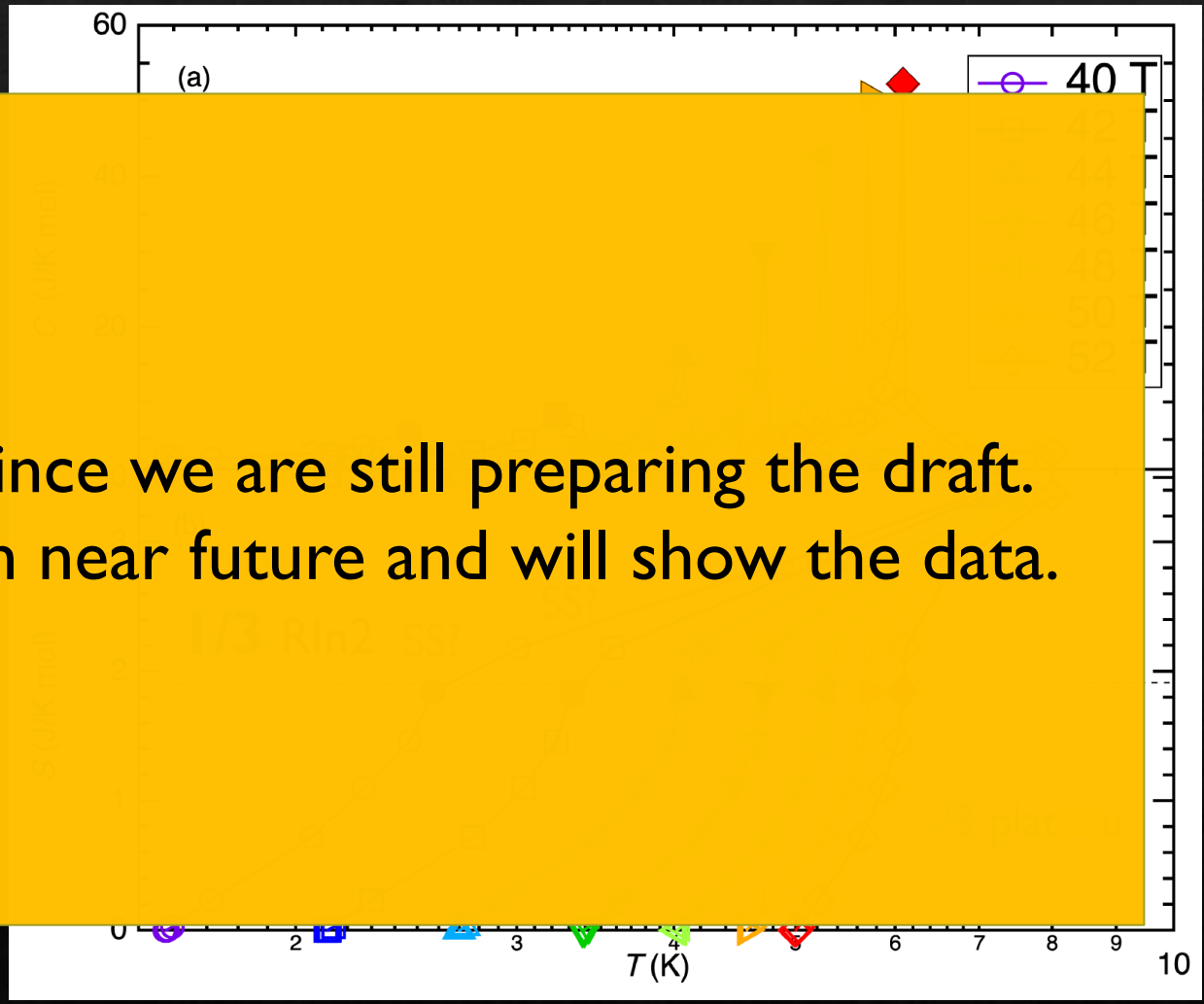
SS?

Cp from MCE data

$$C_M/T = \partial S_M/\partial T$$

Constant transition entropy for the same plateau !!!





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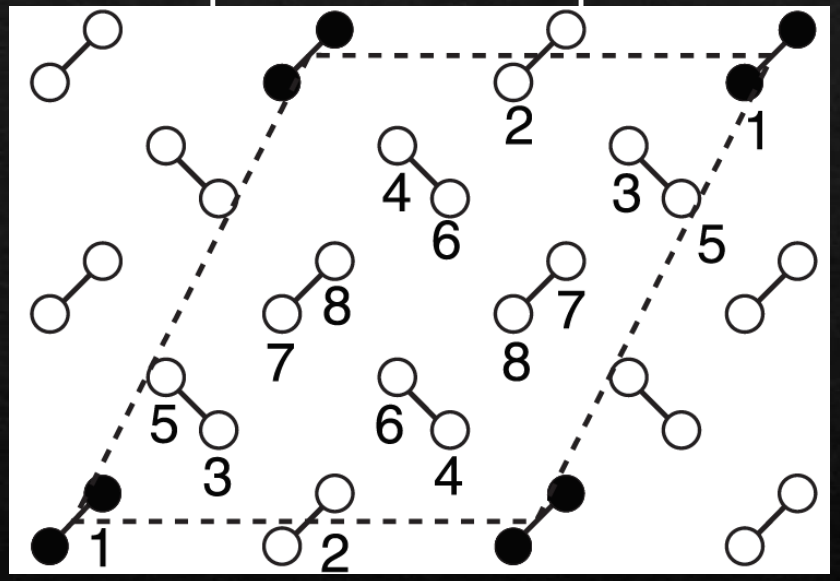
The extremely sharp singularity in Cp implies the 1st order character of the phase transition, a reminiscence of the gas to solid phase transition. The Cp anomaly for SS might be 2nd order ?

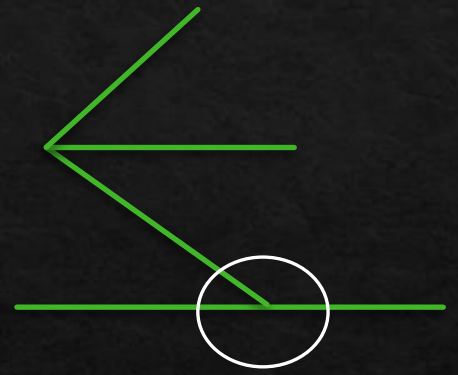
Absolute value of entropy (rough estimation)

Two degree of freedoms for a dimer; Triplet or singlet

$\Rightarrow S = k_B \ln(2)$ per dimer ($S_{sat} = k_B N_A \ln(2)$ per mole of $SrCu_2(BO_3)_2$)

Simple model of 1/8 plateau





*We only consider the lowest 2 levels

Number of triplon at $1/a$ plateaus is N_A/a per mol.

$N_A k_B/a \ln(2)$ could be the saturation entropy for the $1/a$ plateau phases.

(1/3 plateau $\Rightarrow R\ln(2)/3 = 1/3\ S_{sat}$, 1/4 plateau $\Rightarrow R\ln(2)/4 = 1/4\ S_{sat}$.)

Next step…

- Now we have rough idea about the entropy and (H,T) phase diagram in $SrCu_2(BO_3)_2$.
- To investigate low temperature entropy (below 1K), we couldn't use millisecond pulsed fields due to strong MCE.

- C_p measurements in long pulse & DC fields.

ISSP
Shusaku Imajo

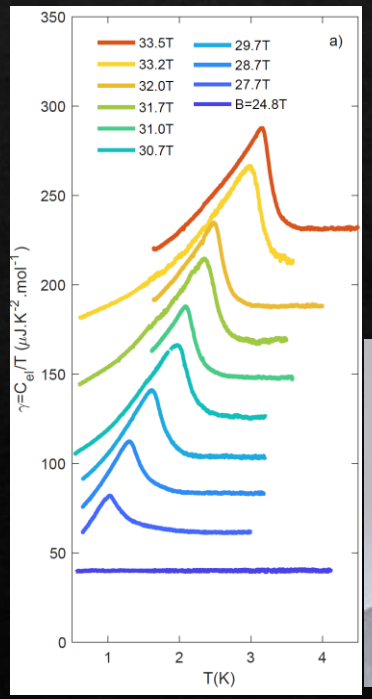
Up to ~44 T
Down to 0.6 K

Accepted in RSI
arXiv: 2012.02411

Grenoble
Christophe Marcenat

Up to 38 T,
Down to 0.5 K

C. Marcenat et al., *PRL* 126, 106801 (2021)



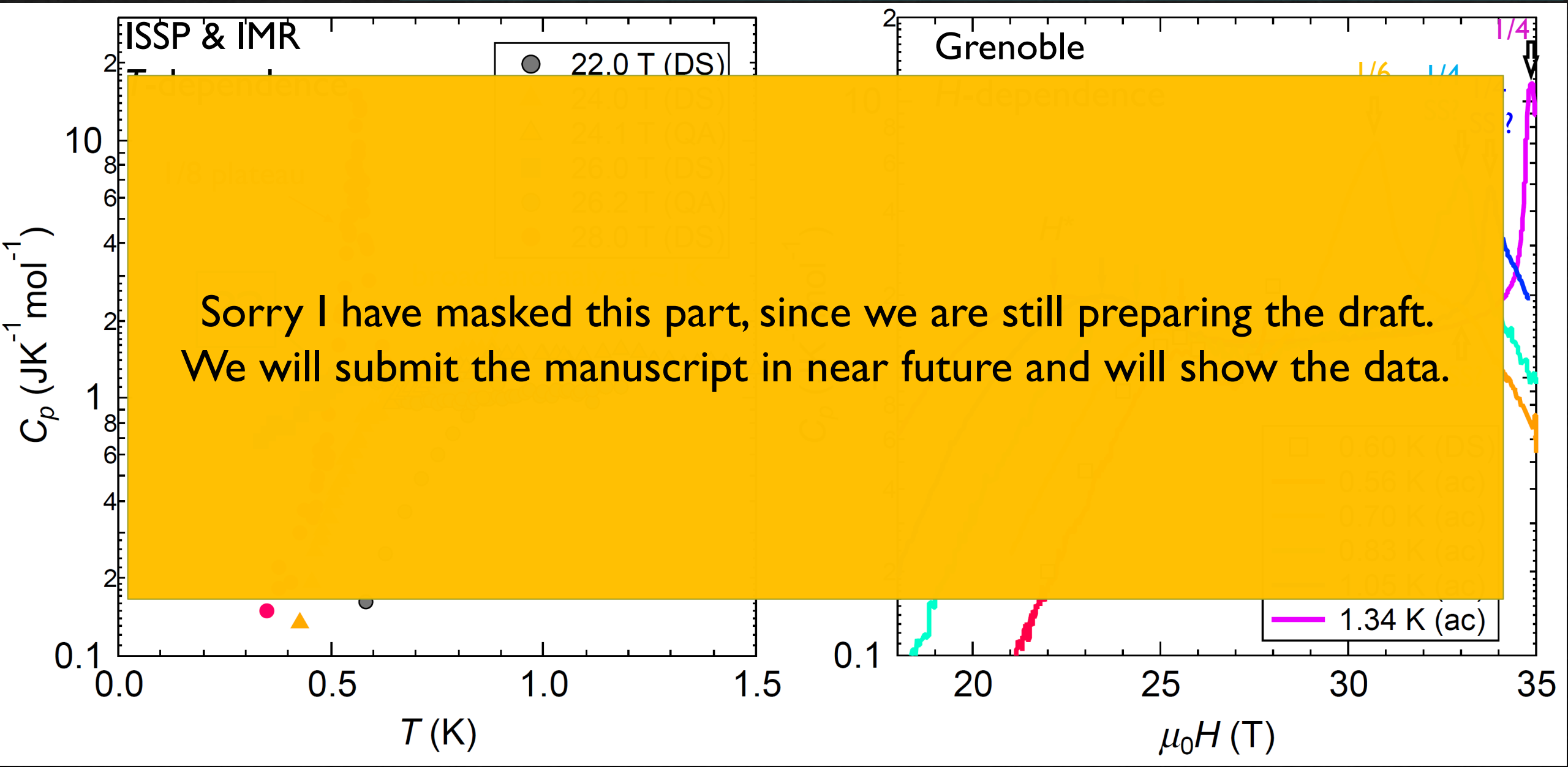
Tohoku University (IMR)
Takumi Kihara

Up to 30 T,
Down to 0.3 K

In preparation

Specific Heat (*H*//*c*) (ISSP, IMR, & LNCMI)

Sharp peaks were observed in plateau phases, while broad anomalies were seen below 24 T. We found a broad peak at ~0.5 K at 26 T.



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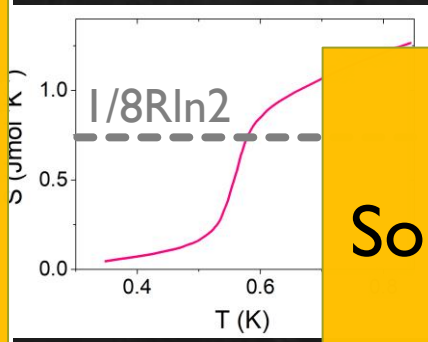
Specific Heat $C_p(T)$



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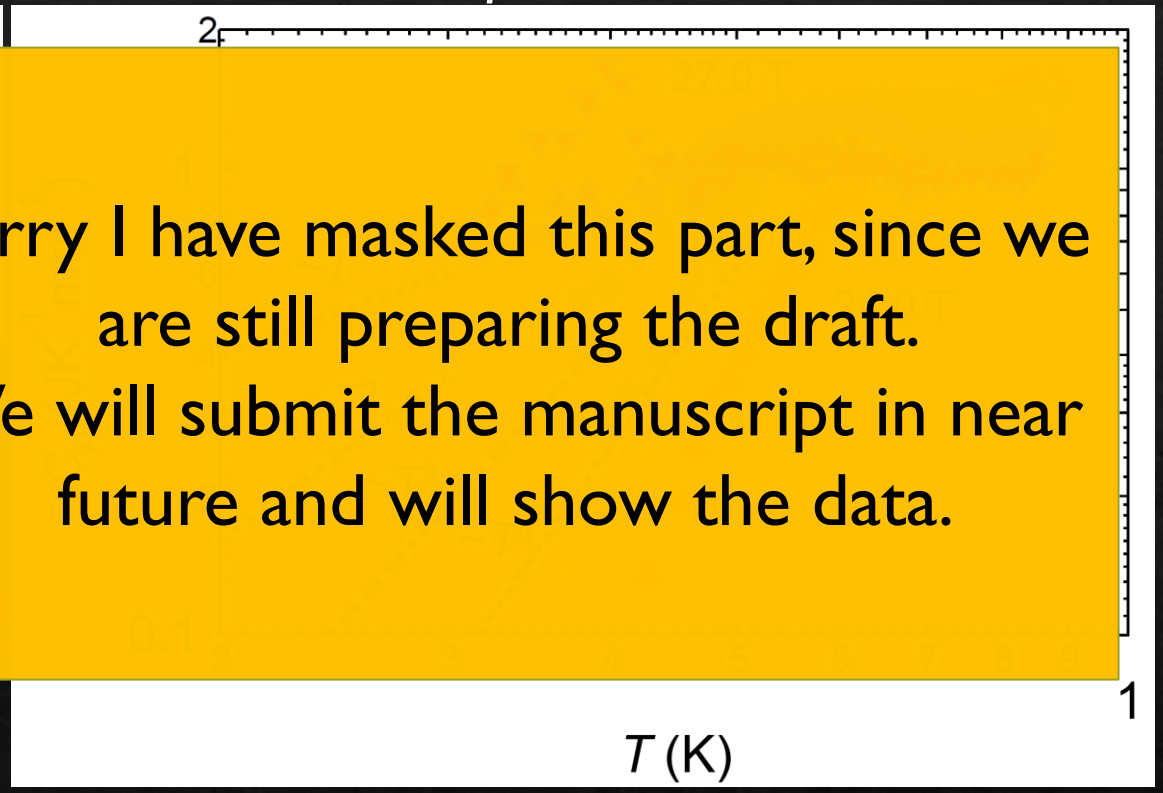

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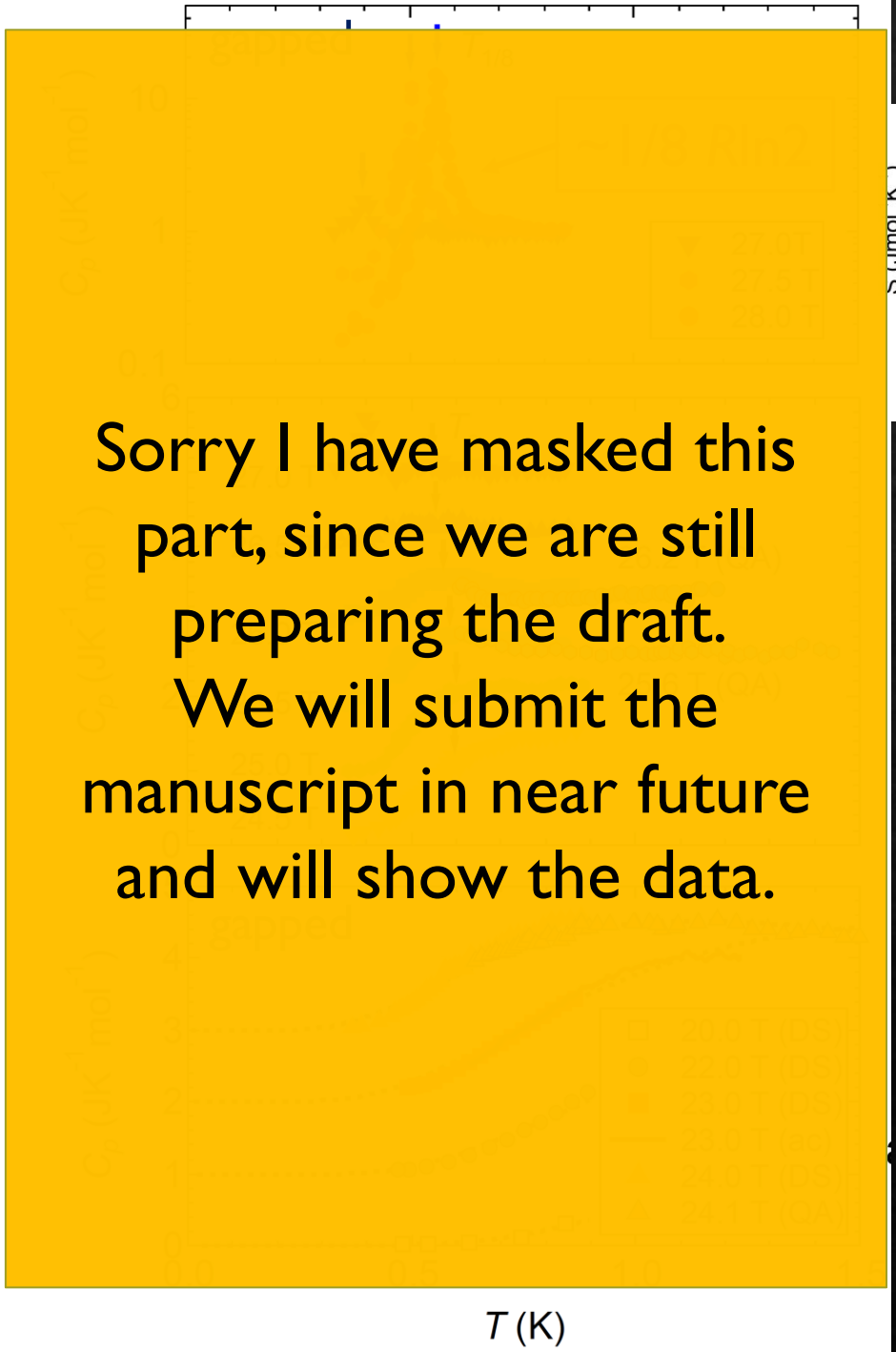
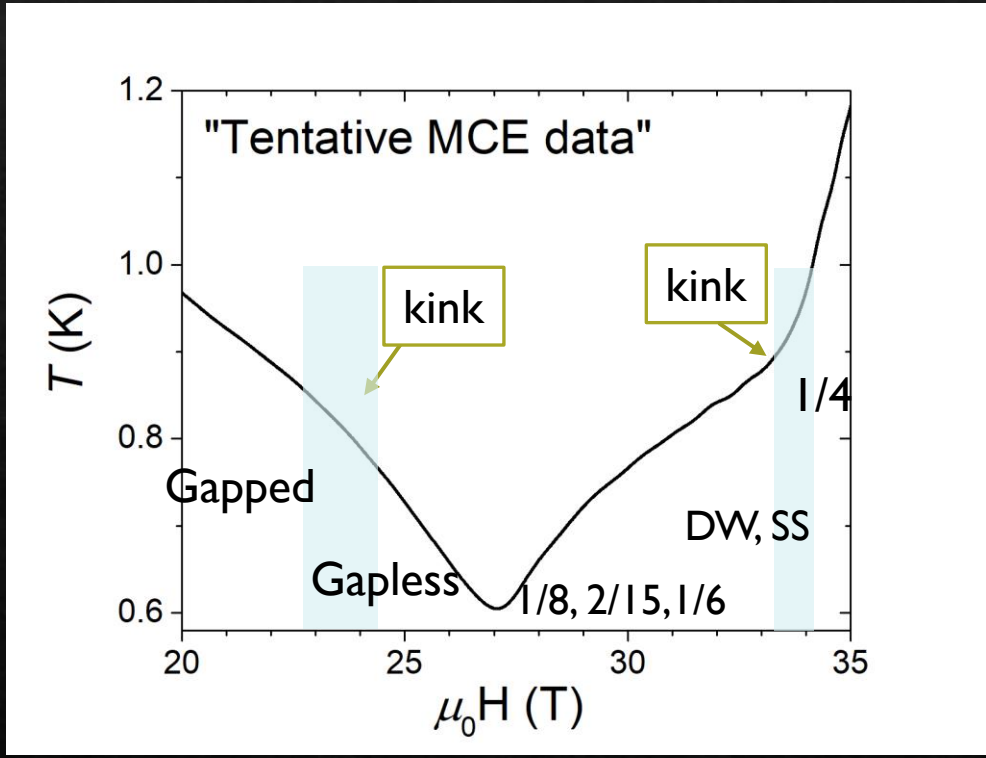


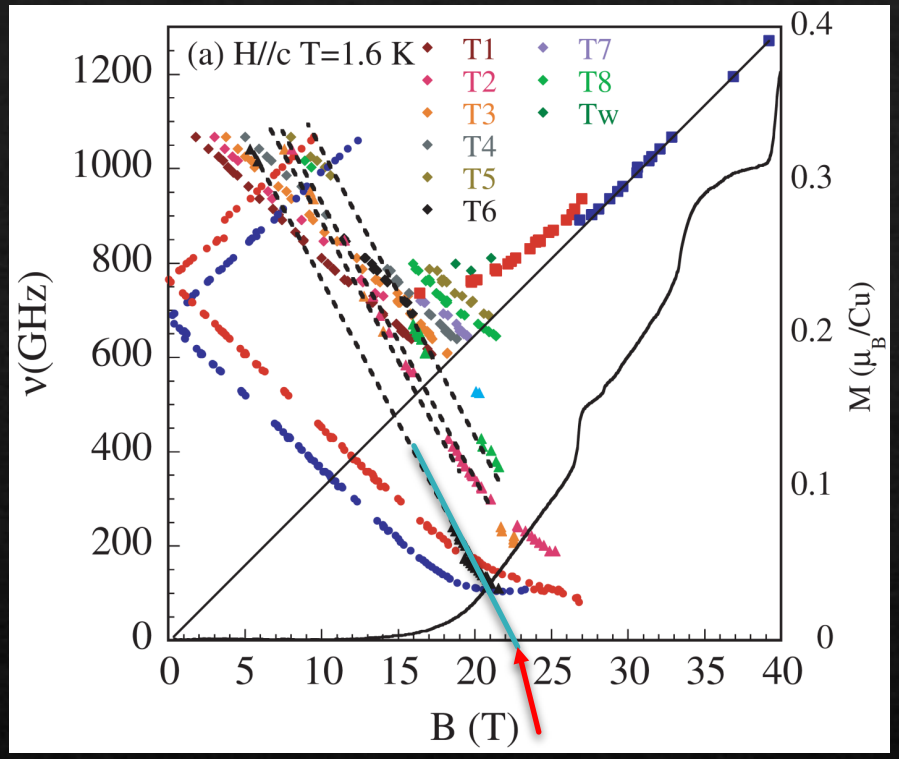
The $C_p(T)$ clearly deviate from exponential law and follows a power law at low T.

⇒ "Gapless" in the intermediate field region

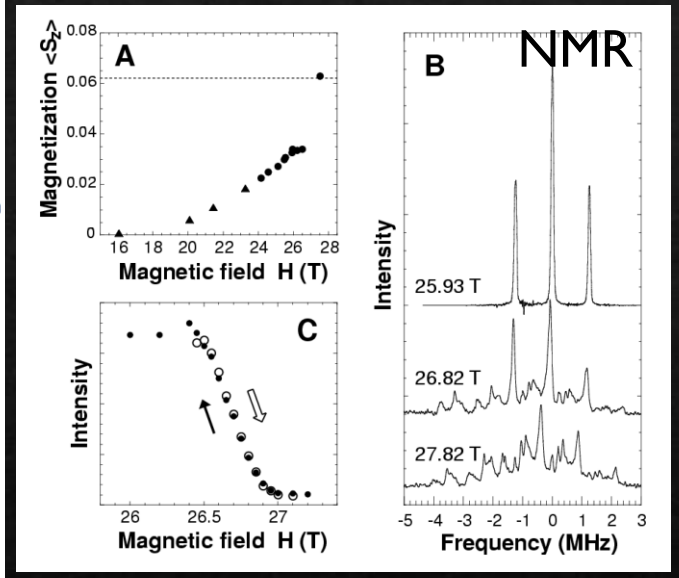
H. Tsujii, *JPSJ* **80**, 043707 (2011)
M. Hofmann et al., *PRL* **87** 047202 (2001).

MCE in long pulse (tentative data)





~23 T



K. Kodama et al., *Science* **298**, 395 (2002).

At ~24 and 33 T, there are two kinks in *T*(*H*) curve.
The high field one is due to the entrance to the SS or 1/4 plateau phase.
The low field one could be attributed to the band crossing between S_z = 0 and 2 states.
*Between 24 and 26.5 T, there might be a gapless phase, but no internal field is observed.

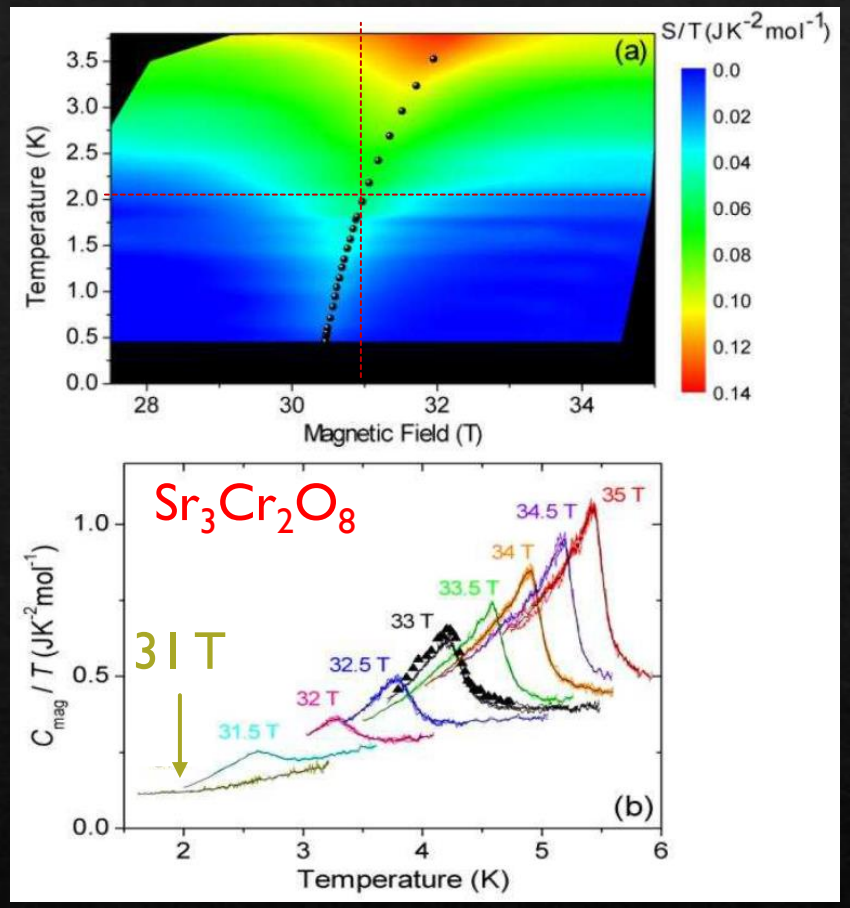
Discussion

- Large entropy should be released at low T ⇒ condensate of $S_z = 2$. But… why the peak in *C* isn't sharp??

2D-XY universality class? However, $SrCu_2(BO_3)_2$ has relatively strong interlayer coupling and the DM interaction.

- As seen in $Sr_3Cr_2O_8$ and other magnon-BEC systems, the peak in *C* is broaden at the edge of dome (dilute limit).

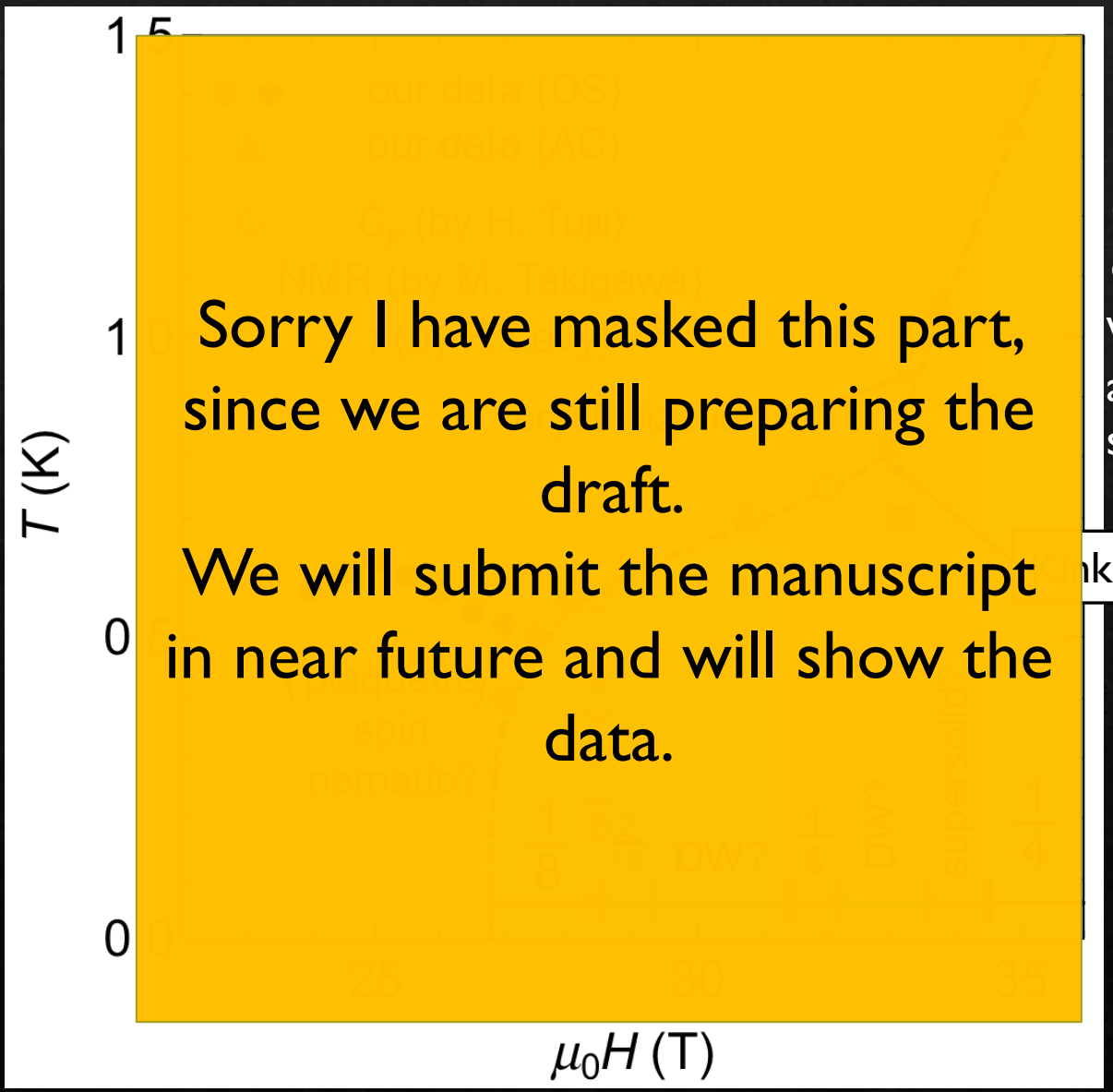
Frustration induced low dimensionality as seen in Han purple? or impurity? With impurity, a Bose glass is observed in Br-doped DTN.



A. Aczel et al., *PRL* **103**, 207203 (2009)

The application of pressure might help to observe the $S_z = 2$ condensate.

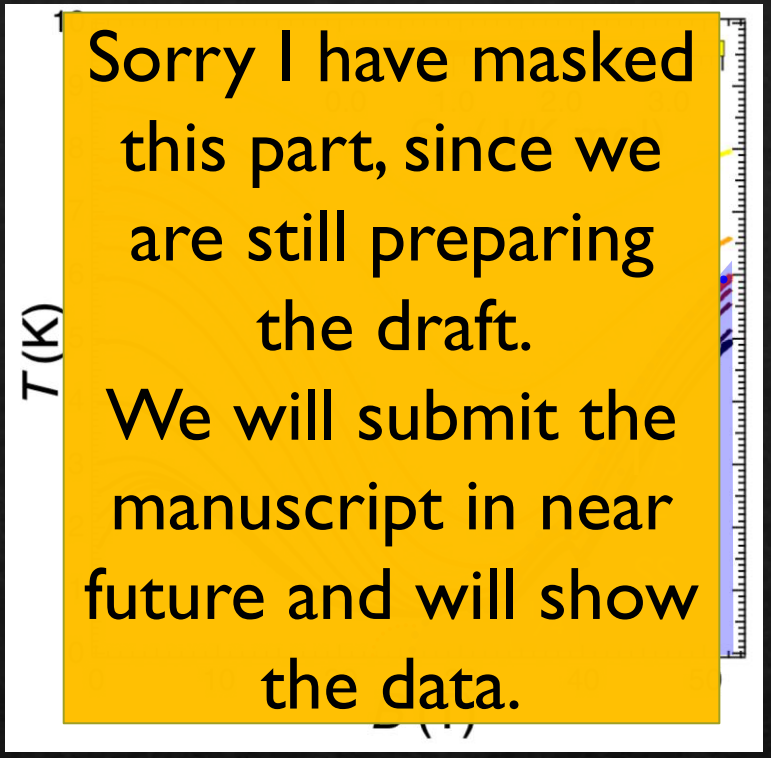
(*H*, *T*) phase diagram (*H*//*c*)



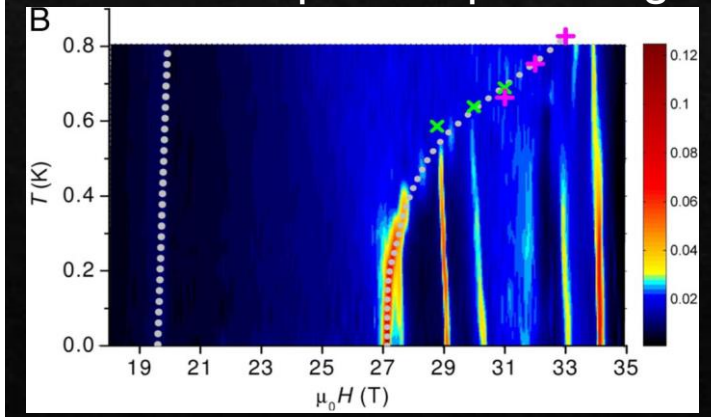
Clausius-Clapeyron eq.
$dT_c/dH_c = -\Delta M/\Delta S$

When ΔS is constant,
a plateau phase has dome-shaped phase boundary.

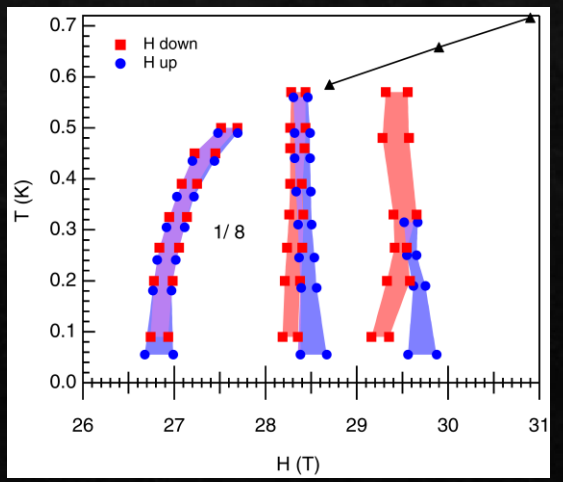
k in the phase diagram



Previous reports of phase diagram



S. E. Sebastian et al., *PNAS*, **105** 20157 (2008)



F. Levy et al., *EPL*, **81** 67004 (2008)

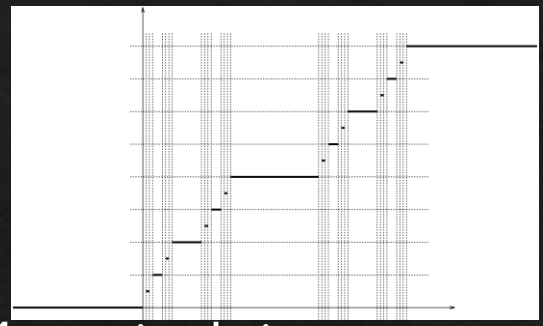
DW & SS is deduced from *PRL* **112** 147203 (2014)
(Plaquette) spin nematic is deduced from *PRB* **62**, 15067 (2000) & *PRL* **120** 247201 (2018)
Field region for each phases is taken from magnetization data; *PRL* 110, 067210 (2013)

Thank you for your attention!

1. Empirical rule; the entropy of the 1/a plateau is ~R/aln(2).
2. Broad anomaly at ~26 T; possible observation of the SN state.
3. "Mountain chain" like (H,T) phase diagram.

Two separate manuscripts are in preparation by S. Akimoto & S. Imajo.

Devil's staircase in M



Mountain chain
in (H,T) phase diagram

Mountain chain; High Tatras in Slovakia
[https://www.goodfreephotos.com]