Field-dependent thermal, magnetic, and lattice properties of the QSL candidate α-$RuCL_3$

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Yan, JQ; Mandrus, DG; Takano, Y; Brosha, EL; Rosa, PFS;
Nagler, SE; Kindo, K**; Jaime, M**

PHYSICAL REVIEW B **102**, 214432 (2020)

24/03/2021

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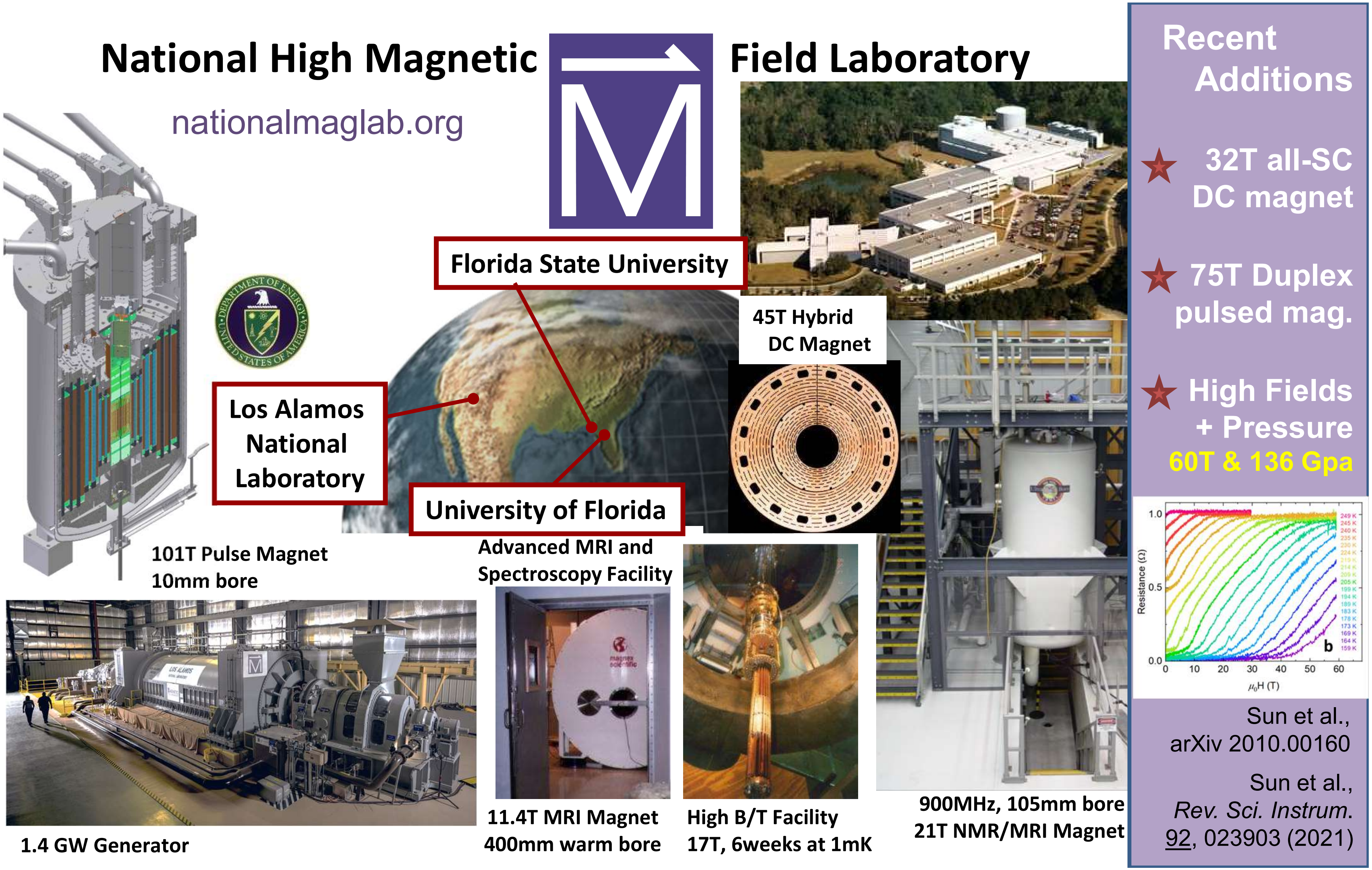
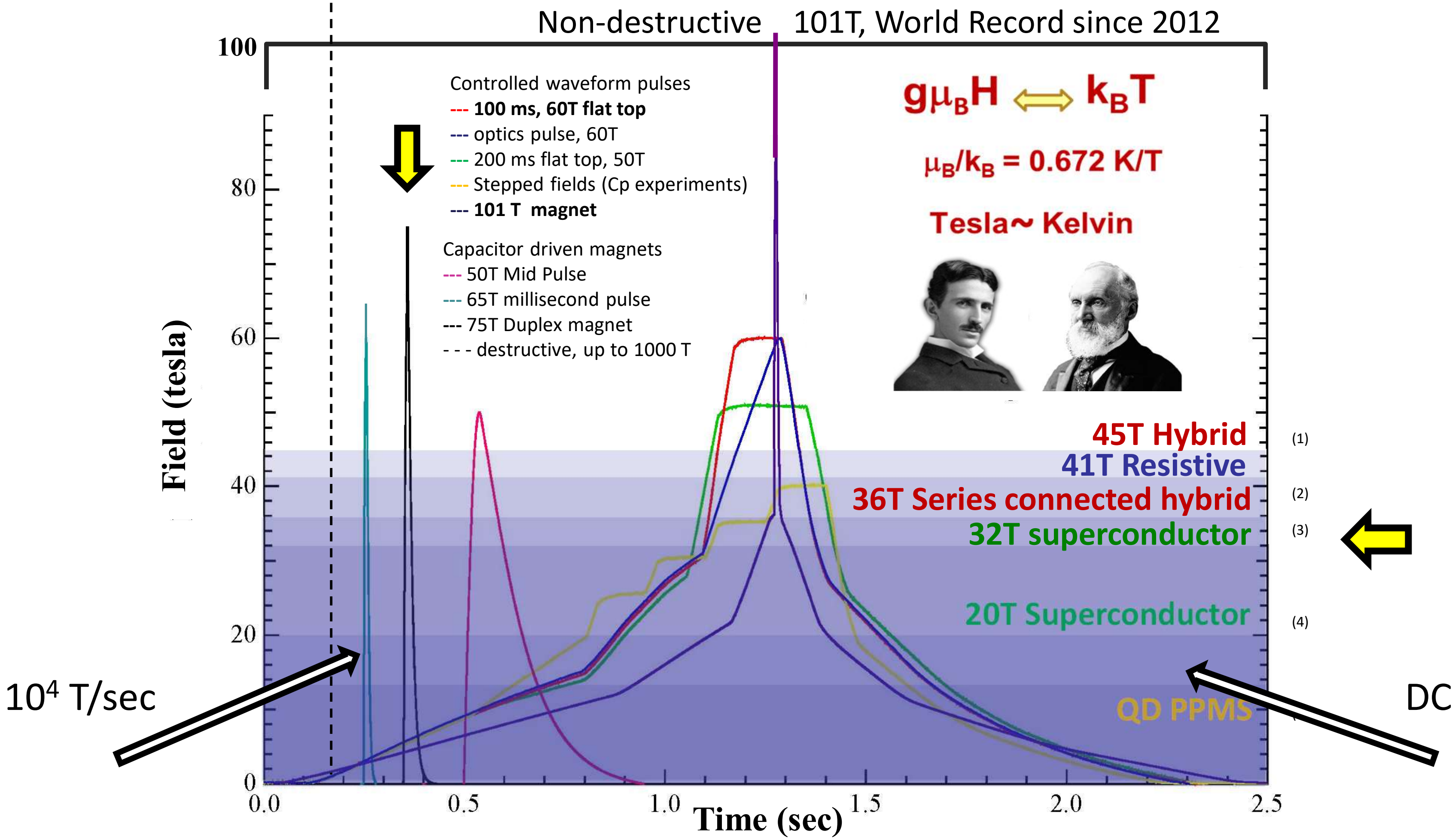
LA-UR


National High Magnetic Field Laboratory
nationalmaglab.org
Florida State University
Los Alamos National Laboratory
University of Florida
45T Hybrid DC Magnet
101T Pulse Magnet 10mm bore
Advanced MRI and Spectroscopy Facility
1.4 GW Generator
11.4T MRI Magnet 400mm warm bore
High B/T Facility 17T, 6weeks at 1mK
900MHz, 105mm bore 21T NMR/MRI Magnet
Recent Additions
★ 32T all-SC DC magnet
★ 75T Duplex pulsed mag.
★ High Fields + Pressure
60T & 136 Gpa
Resistance (Ω)
μ_0H (T)
b
Sun et al., arXiv 2010.00160
Sun et al., *Rev. Sci. Instrum.* 92, 023903 (2021)

What are Extreme Magnetic Fields?

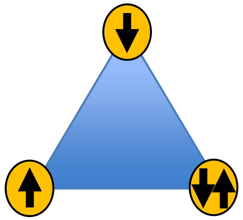


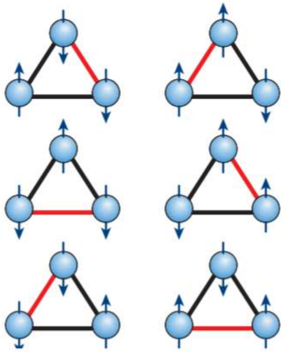
(1) (2) Nb3Sn SC cable-in-conduit conductor CICC (3) YBCO, Nb3Sn, and NbTi. (4) (5) Nb3Sn and NbTi

Slide 3

MJ2 Marcelo Jaime; 24.03.2021

Quantum Spin Liquid: Frustrated electrons on a triangular lattice



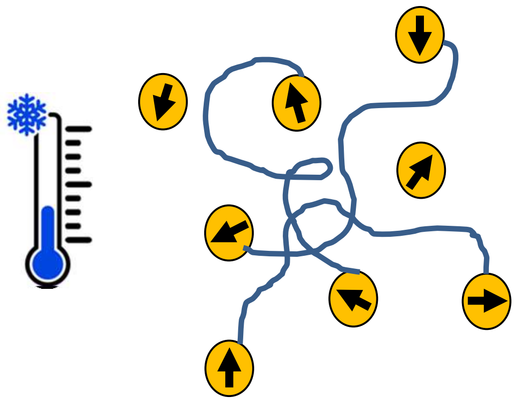


You can't satisfy all of the electrons all of the time.

The simplest example of frustration: A triangle of three anti-ferromagnetically interacting Ising spins, each of which must point up or down.

It is impossible for all three spins to be antiparallel, so instead of two ground states (up and down), there are six grounds states (see below)

Wannier (1950) and Anderson (1973) proposed such a spin-disordered quantum state**.** *Candidate materials have, however, remained elusive …* until recently.



→ Quantum Spin Liquid

Figure credits: F.L. Pratt

Quantum Spin Liquid: Mostly known for what is it not! What are its properties?

- A **Quantum Spin Liquid** is a system with magnetic moments where there are no broken symmetries.

- It is a state of matter never *seen* before yet expected as T→ 0.
  - -The moments do <u>not</u> break translational symmetries, so they are like a liquid.
  - -There is <u>no</u> magnetic order, which is suppressed by quantum fluctuations.
  - -It is <u>not</u> a paramagnet. Interactions lead to long-ranged entanglement of the wave functions.

- The *low energy properties* of a Quantum Spin Liquid are likely related to phenomena such as:
  - -Quantum fluctuations (small spins)
  - -Quantum entanglement (non-local effects)
  - -Quantum coherence (macroscopic wave function)
  - -The topology of the quantum wave function (strong SOC)

- The *signature properties* of a Quantum Spin Liquid are not fully known, but they include:
  - -Non-local and topological excitations.
  - -Fractionalized excitations.
  - -'Nothingness' (absence of observables, frustration parameter "*f*")

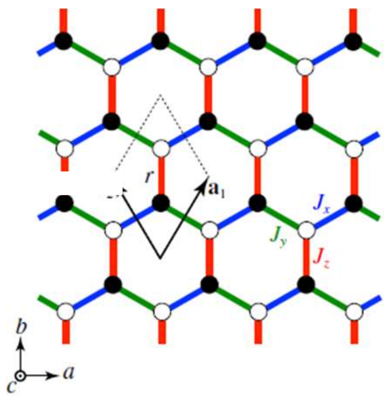
- QSL ground states may be topologically protected, suggesting possible role as qubits in quantum information applications.
- **Platform to study <u>Majorana fermions as quasiparticle excitations</u> in CM physics** $\gamma_i^\dagger = \gamma_i,$

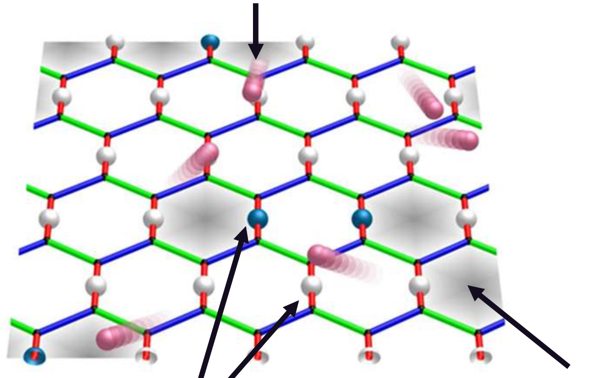
Motome and Nasu JPSJ 89, 012002 (2020).

QSL state in Kitaev magnets: *spin fractionalization*

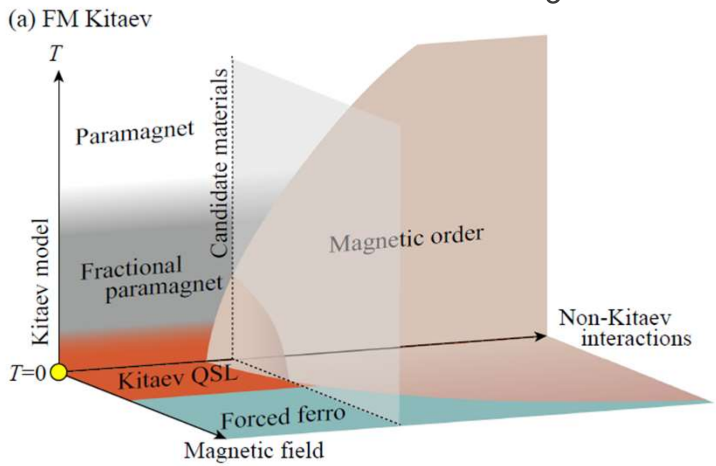
Kitaev model, for a spin 1/2 on a 2D honeycomb structure with bond-dependent interactions

$$\mathcal{H} = -\sum_{\mu=x,y,z} J_\mu \sum_{\langle i,j\rangle_\mu} S_i^\mu S_j^\mu,$$





α-$RuCl_3$

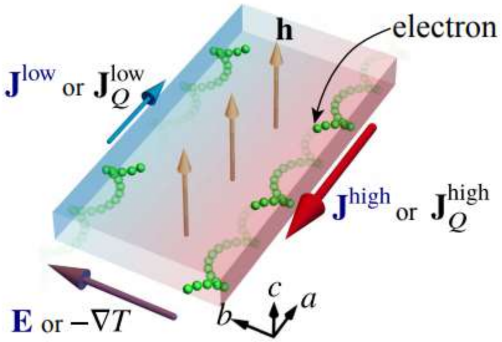


Alexei Kitaev A, Ann. Phys. 321, 2 (2006).
G. Jackeli and G. Khaliullin
Phys. Rev. Lett. 102, 017205 (2009)

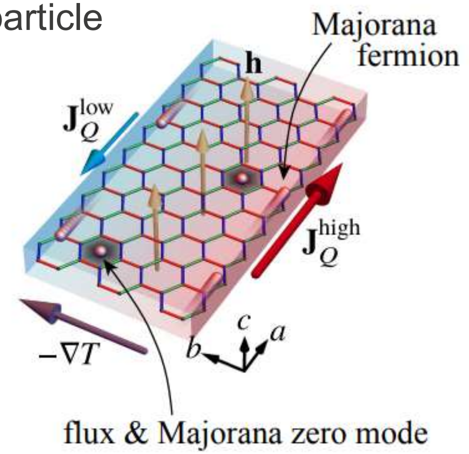
Motome and Nasu JPSJ 89, 012002 (2020) → Finite Temperature
Takagi, et al., Nat. Rev. 1, 264 (2019)
Knolle and Moessner, Ann. Rev. CMP. 10, 451 (2019)
Janssen and Vojta, J. Phys.: Cond. Matt. 31, 423002 (2019)
M. Hermanns, et al., Ann. Rev. CMP 9, 17 (2018)
Winter, et al., J. Phys. Cond. Matt. 29, 493002 (2017)
Nussinov and van den Brink, Rev. Mod. Phys. 87, 1 (2015)

QSL state in α-$RuCl_3$

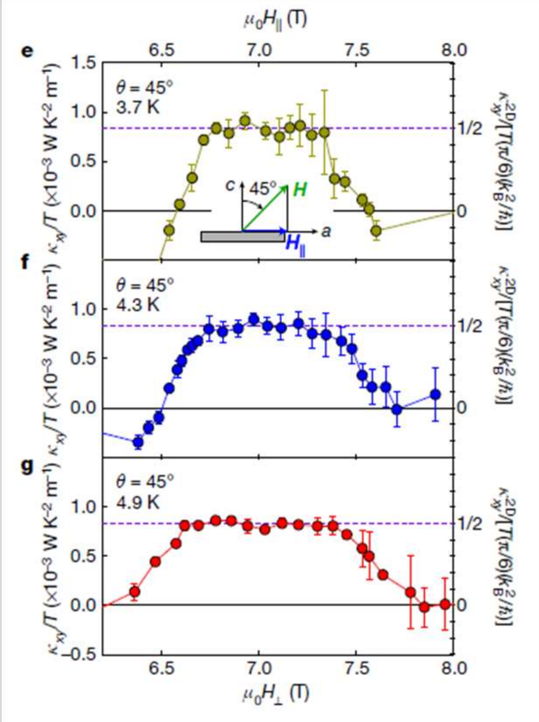
Charged particle QHE



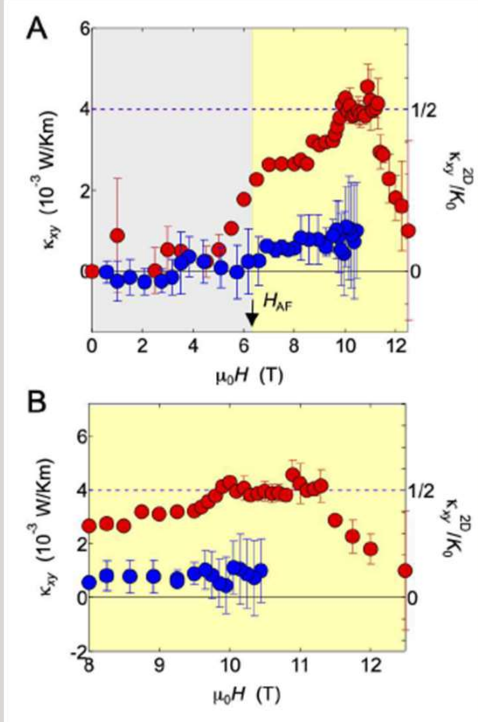
Neutral particle QHE



Half-integer quantized thermal Hall effect



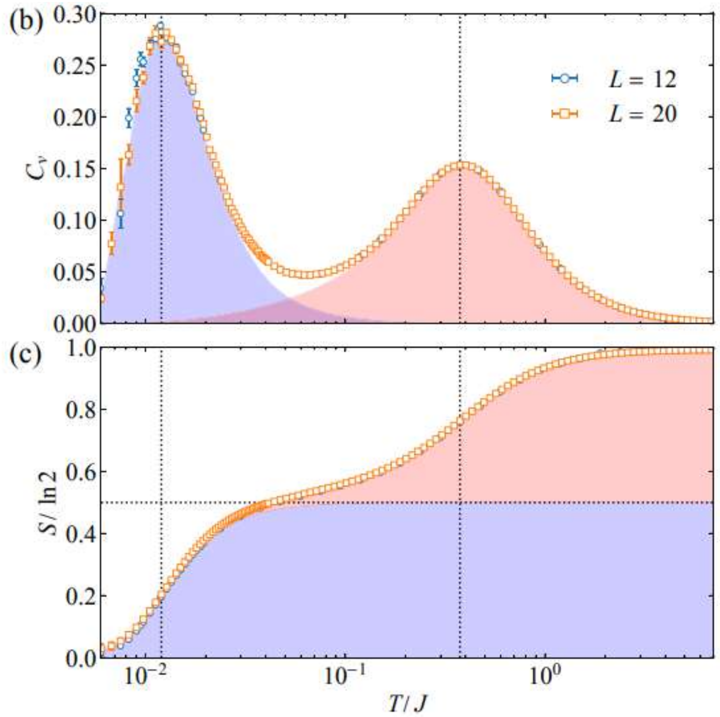
Kasahara *et al.* Nature 559, 227 (2018)



Yokoi *et al.* arXiv: 2001.01988 (2020)

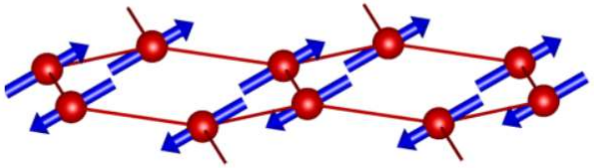
Observable fractionalized thermal properties?

Motome and Nasu JPSJ 89, 012002 (2020) → <u>Finite Temperature</u>

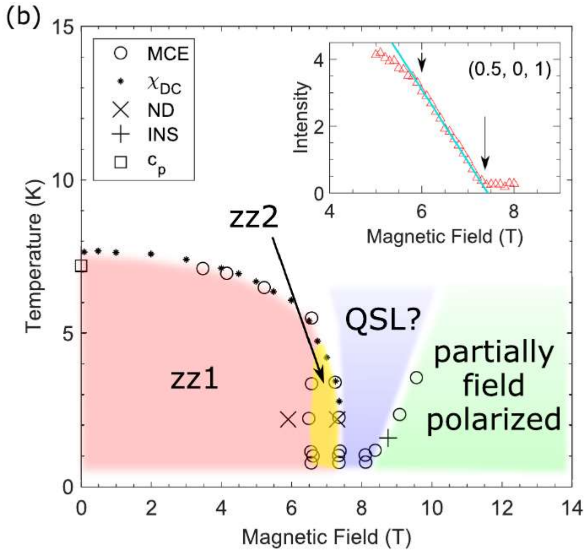


-T dependences of (b) the specific heat Cv, and (c) the entropy S per site normalized by ln 2 for the honeycomb Kitaev model with isotropic coupling Jx = Jy = Jz = J.
-The data are obtained by the Majorana-based QMC simulations for the clusters with N = $2L^2$ spins (L = 12 and 20).
-The reddish and bluish shades show the contributions from the itinerant Majorana fermions and the localized Z_2 fluxes, respectively.
-The horizontal dotted line in (c) represents 1/2 ln 2.

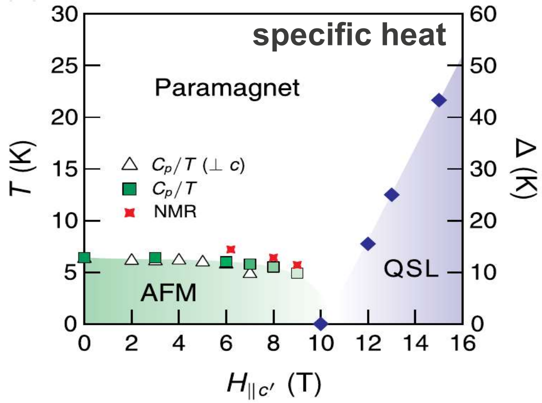
QSL state in α-$RuCl_3$



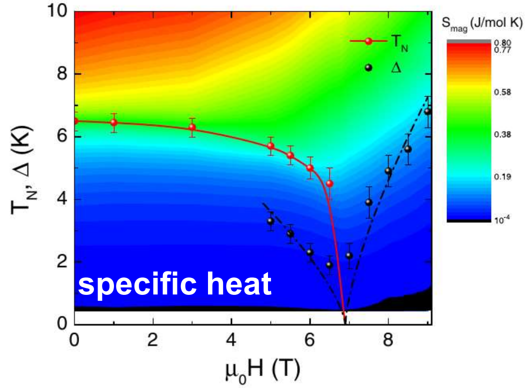
Cao et al. PRB 93, 134423 (2016)



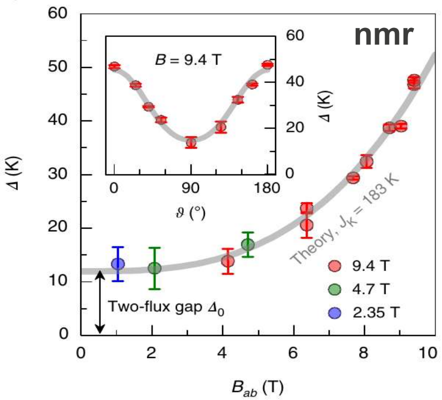
Balz *et al*. PRB 100, 060405 (2019)



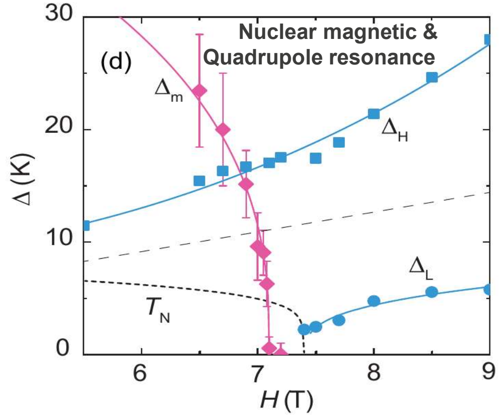
Baek *et al*. PRL 119, 037201 (2017)



Wolter *et al*. PRB 96, 041405 (2017)

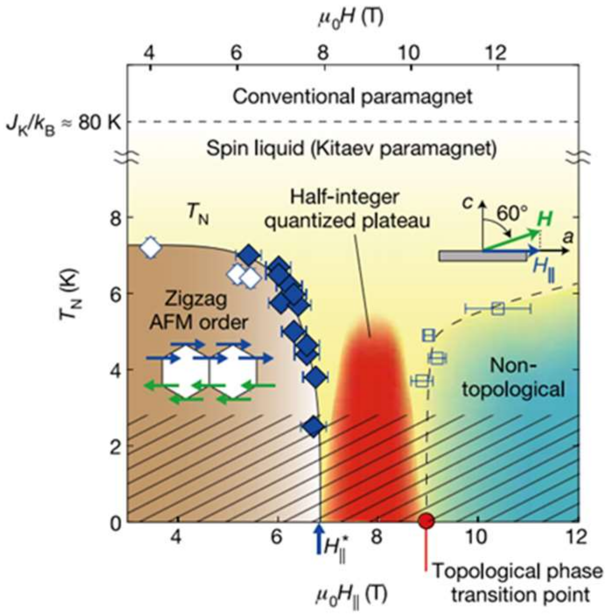


Janša et al. Nature 14, 786-790 (2018)

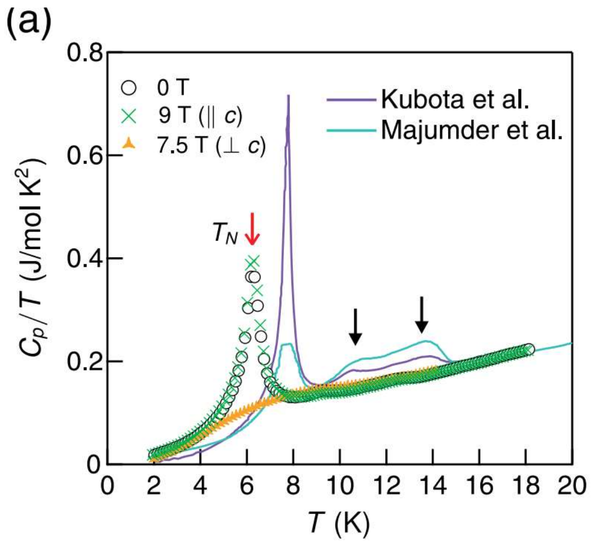


Nagai *et al*. PRB 101, 020414(R) (2020)

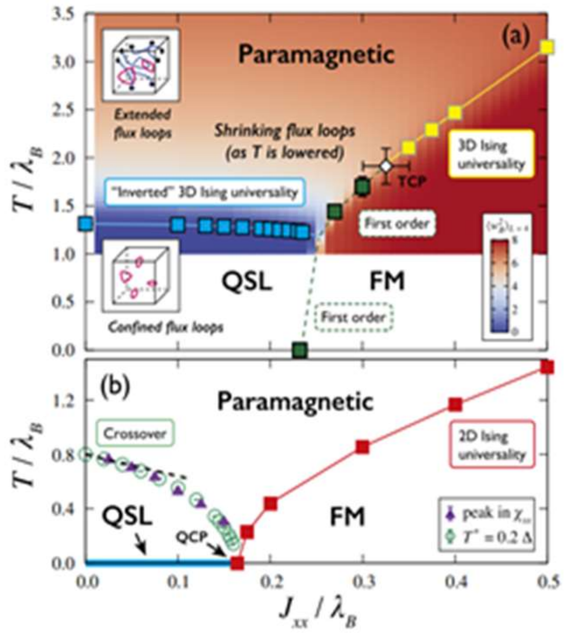
QSL state in α-$RuCl_3$: many questions – few answers



Takagi, et al., Nat. Rev. 1, 264 (2019)



Baek *et al*. PRL 119, 037201 (2017)



Kamiya, et al.; PRB 92, 100403(R) (2015)

Why thermal expansion, magnetocaloric effect?

- Fundamental thermodynamic quantities:

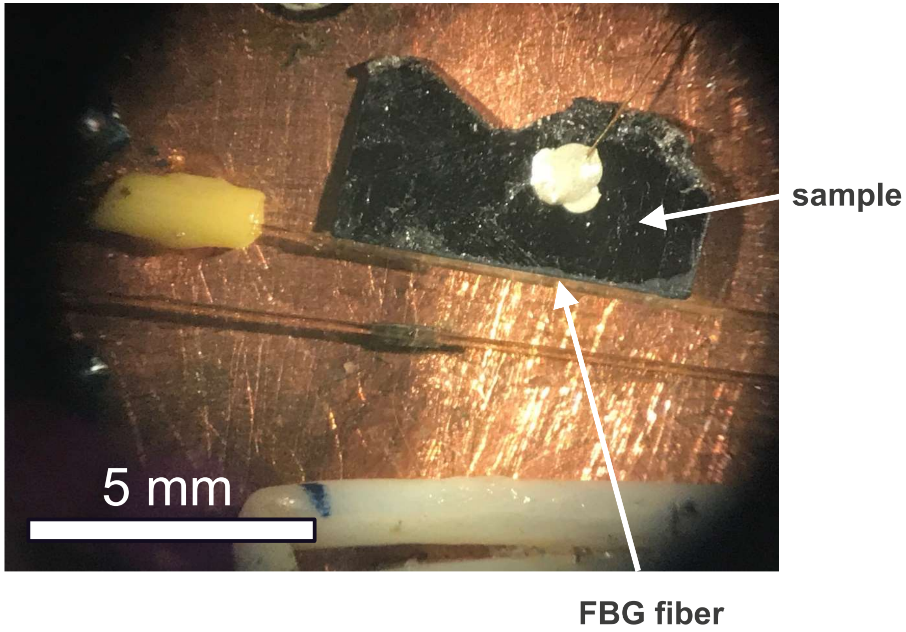
  thermal expansion: $\alpha = \partial^2 G / \partial p \partial T$

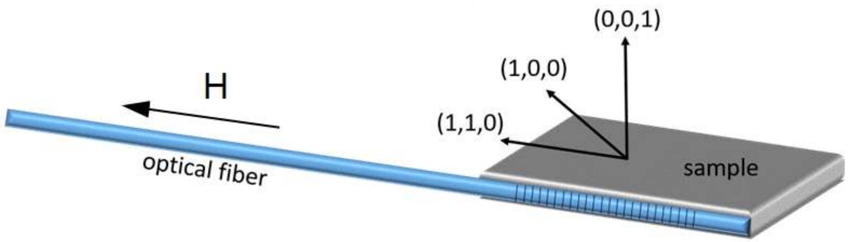
  magnetostriction: $\lambda = \partial^2 G / \partial p \partial H$

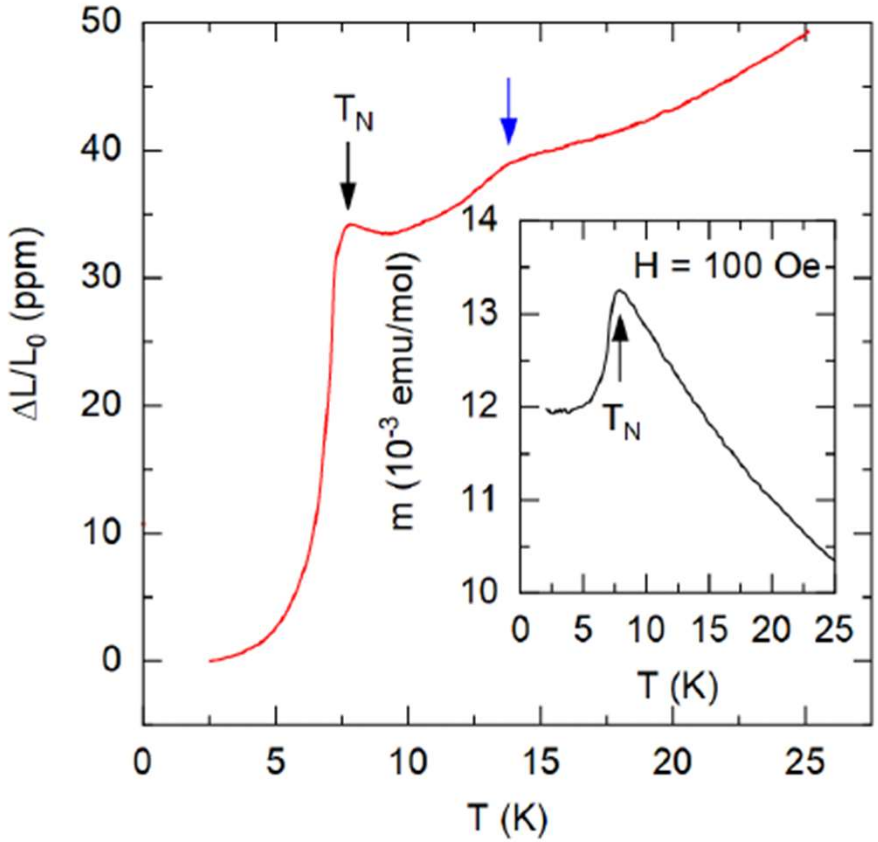
  ➡
  - Identify states of matter
  - Detect and understand phase transitions

➡
- Detect additional phase transition above $H_c \approx 7T$
- Study behavior at temperatures below 2K

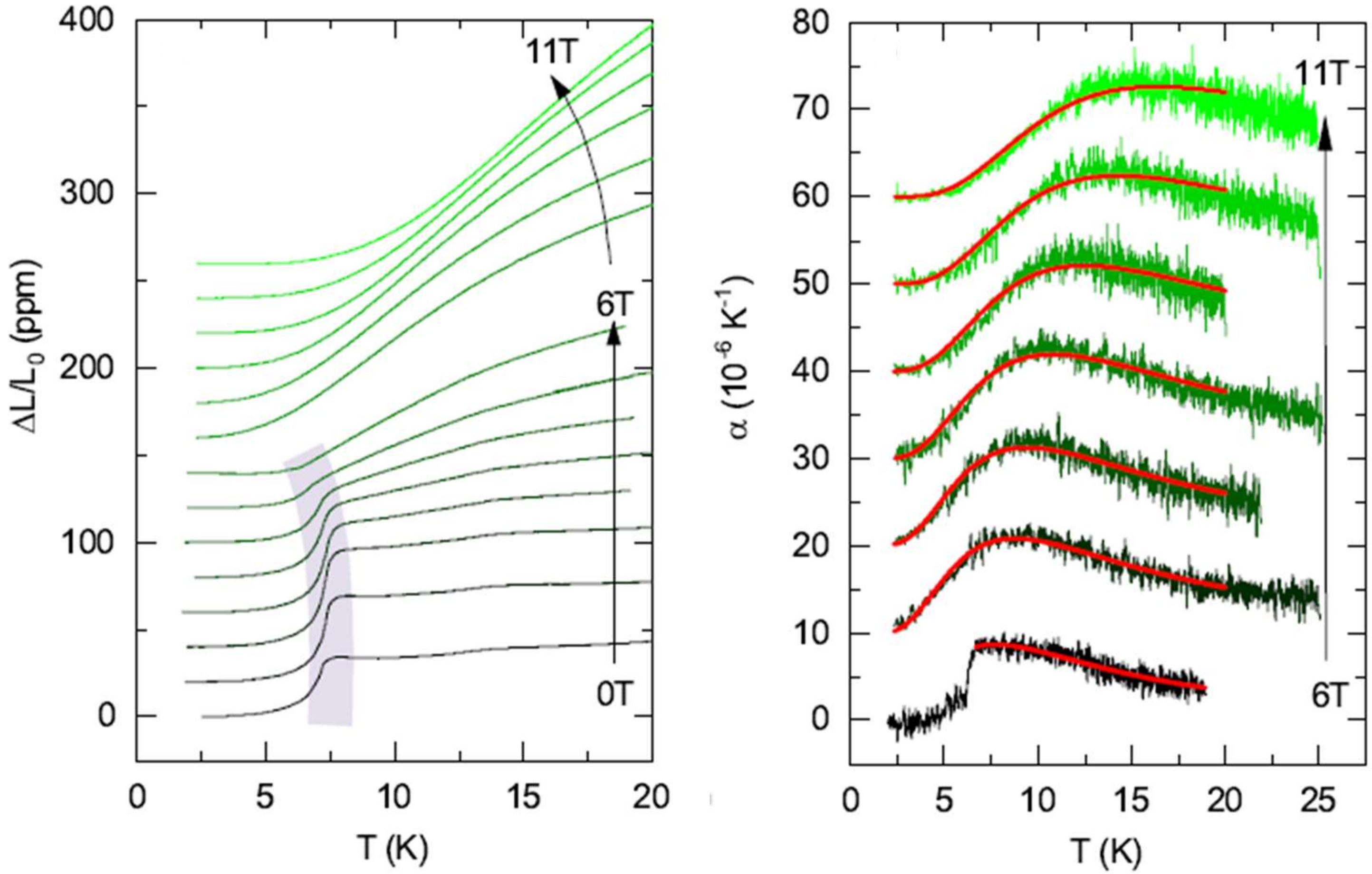
α-$RuCl_3$ thermal expansion





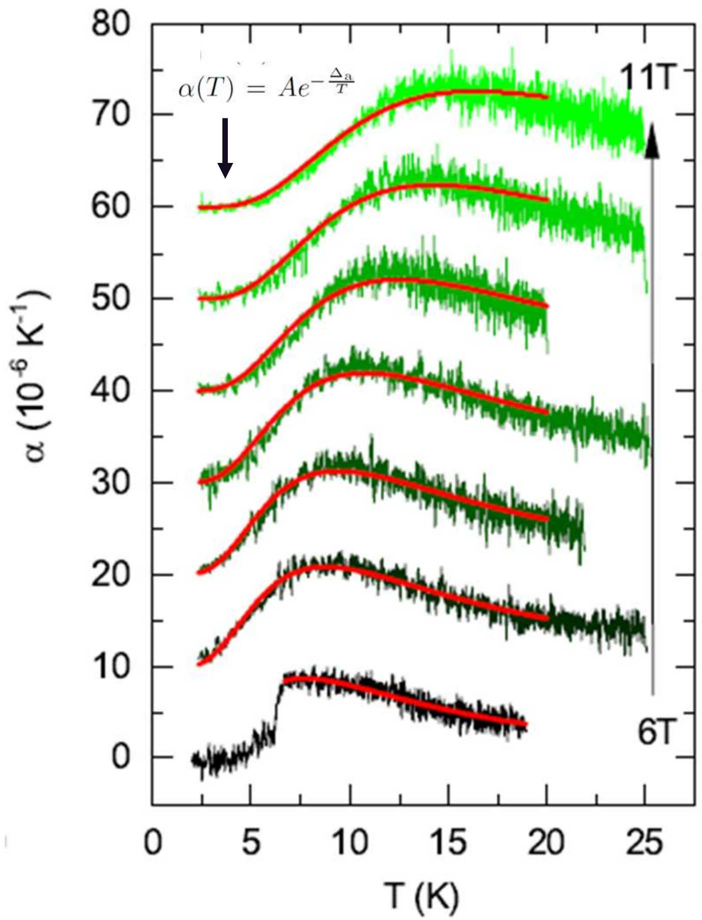


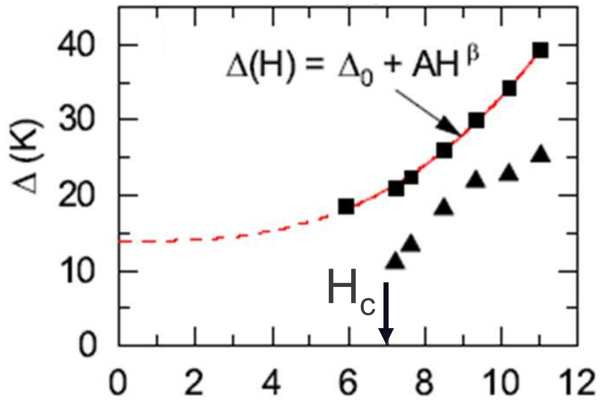
α-$RuCl_3$ thermal expansion



energy gap Δ(H) from Schottky fit of α(T)

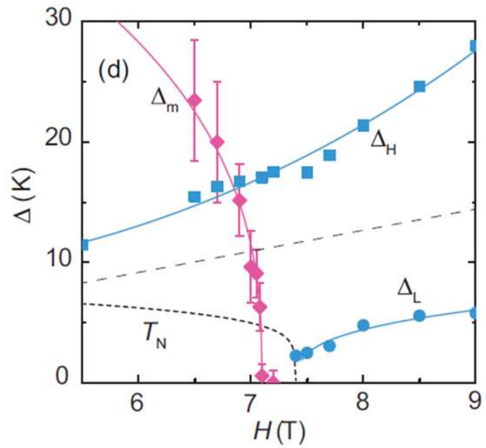
α-$RuCl_3$ thermal expansion





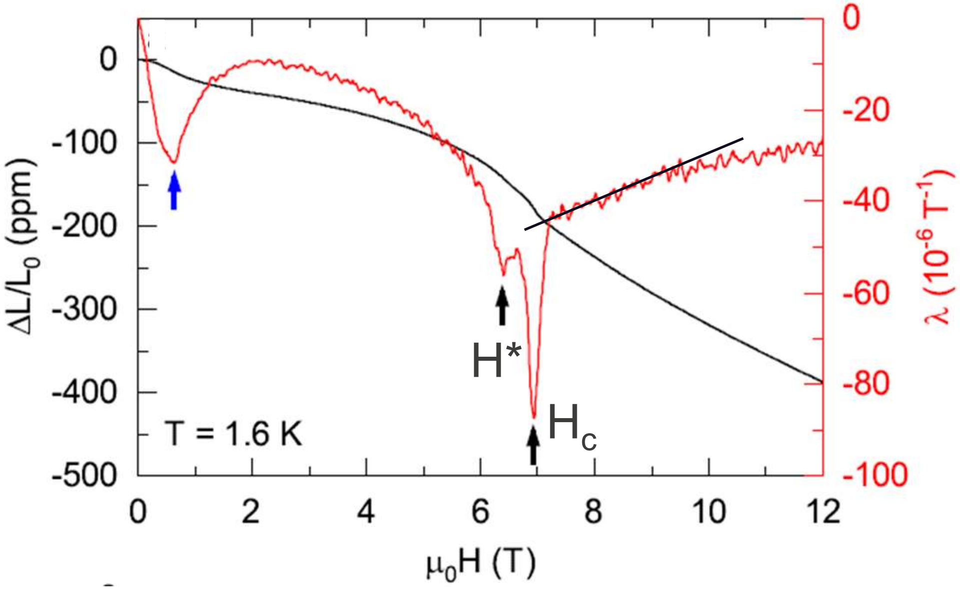
$\Delta_0 = 14\text{K}$

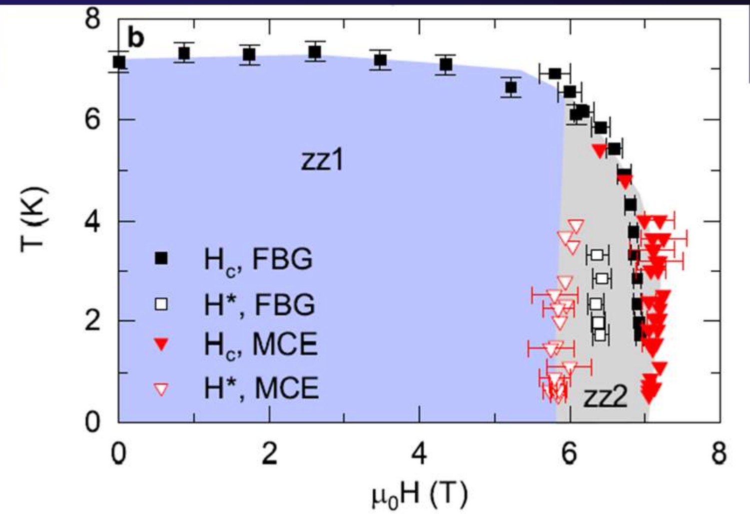
$\beta = 2.9$

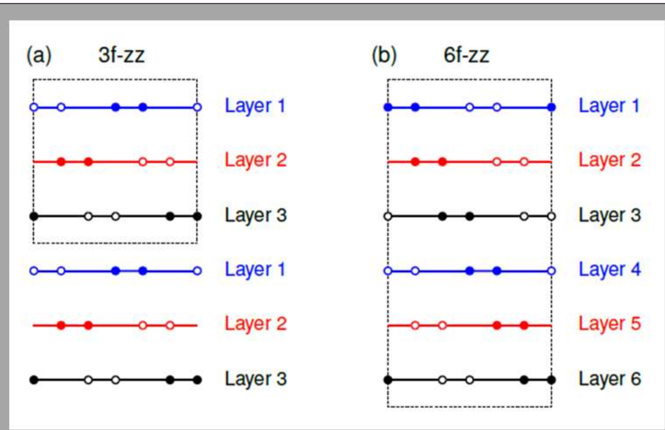


Nagai *et al*. PRB 101, 020414(R) (2020)

α-$RuCl_3$ magnetostriction



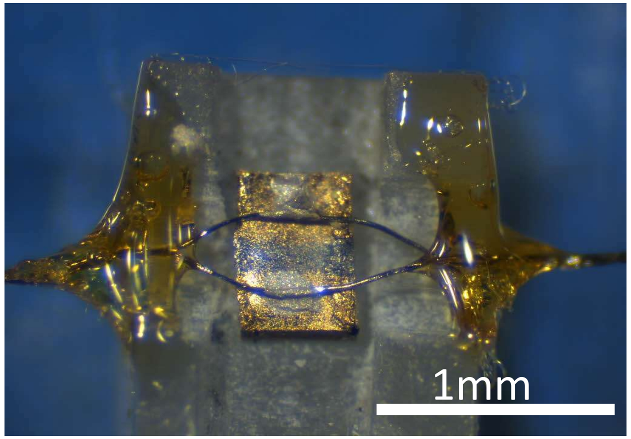


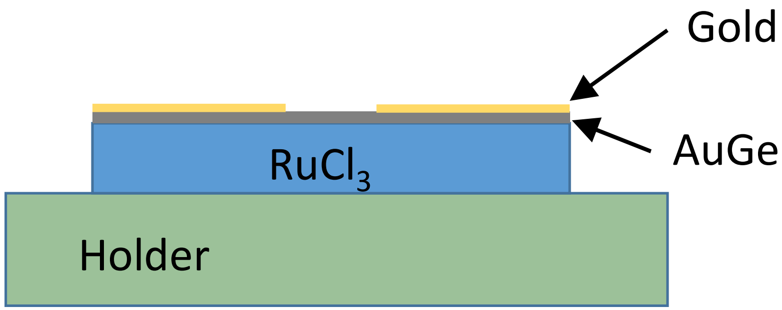


C. Balz et al., arXiv:2012.15258v2

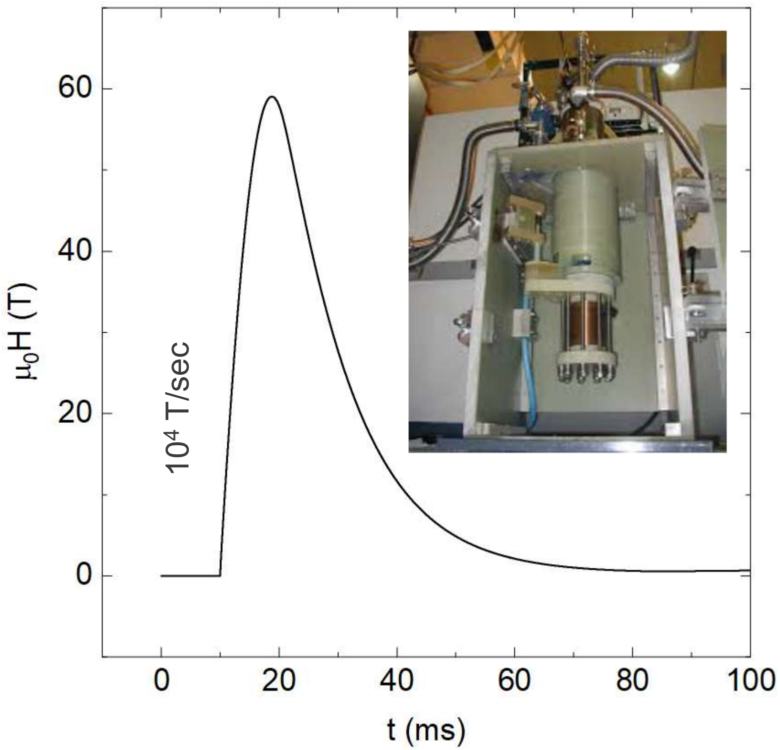
- Phase diagram in agreement with previous results
- No obvious phase transition above H_c
- Finite energy gap at H_c

Magnetocaloric measurements in pulsed fields



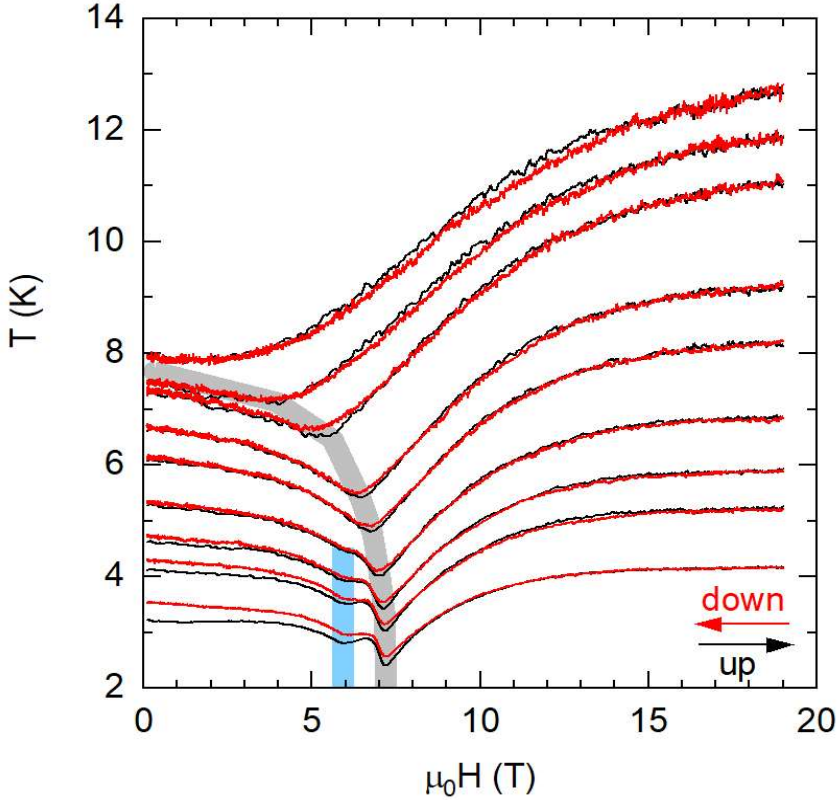


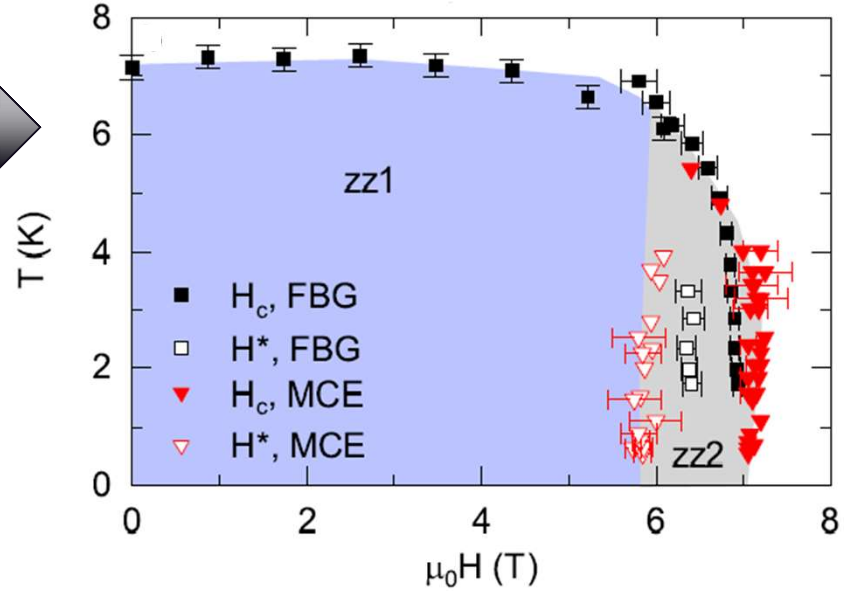
65 T short pulse magnet



α-$RuCl_3$ magnetocaloric effect

quasi-adiabatic conditions

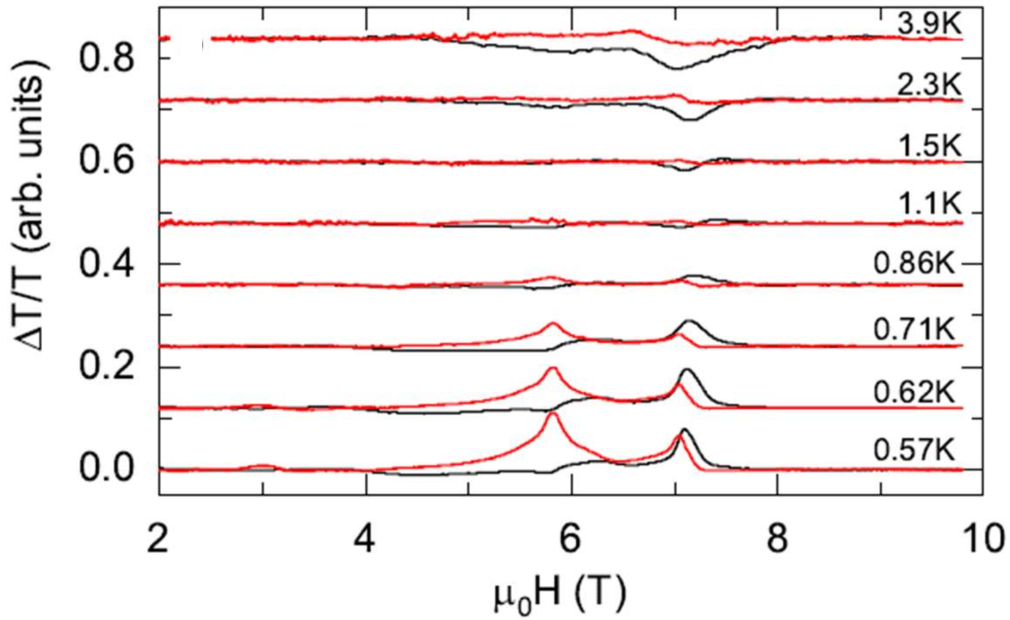




What happens at lower T?

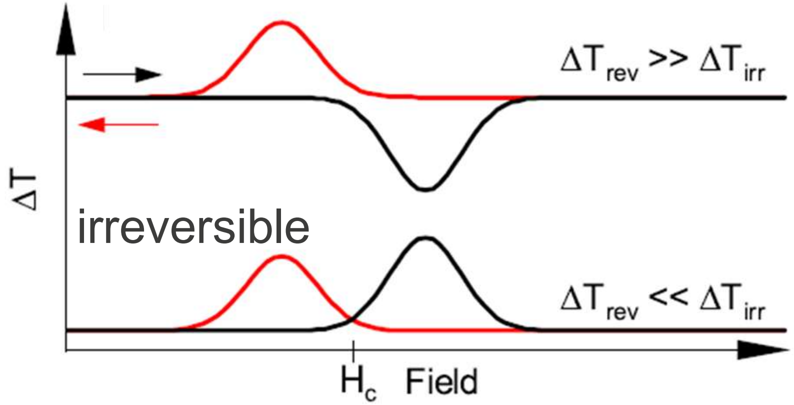
α-$RuCl_3$ magnetocaloric effect

quasi-isothermal conditions



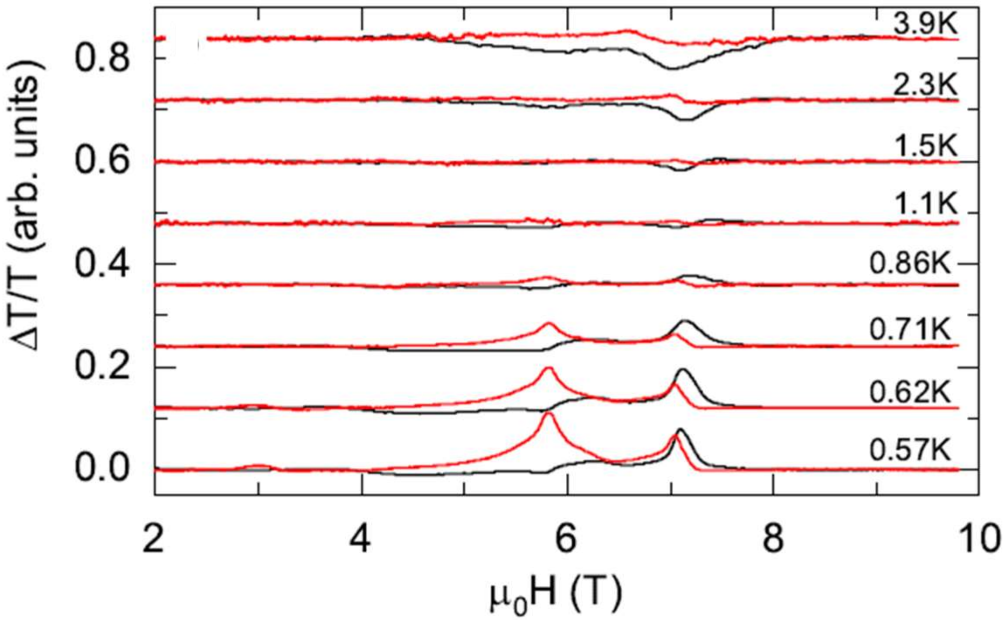
reversible

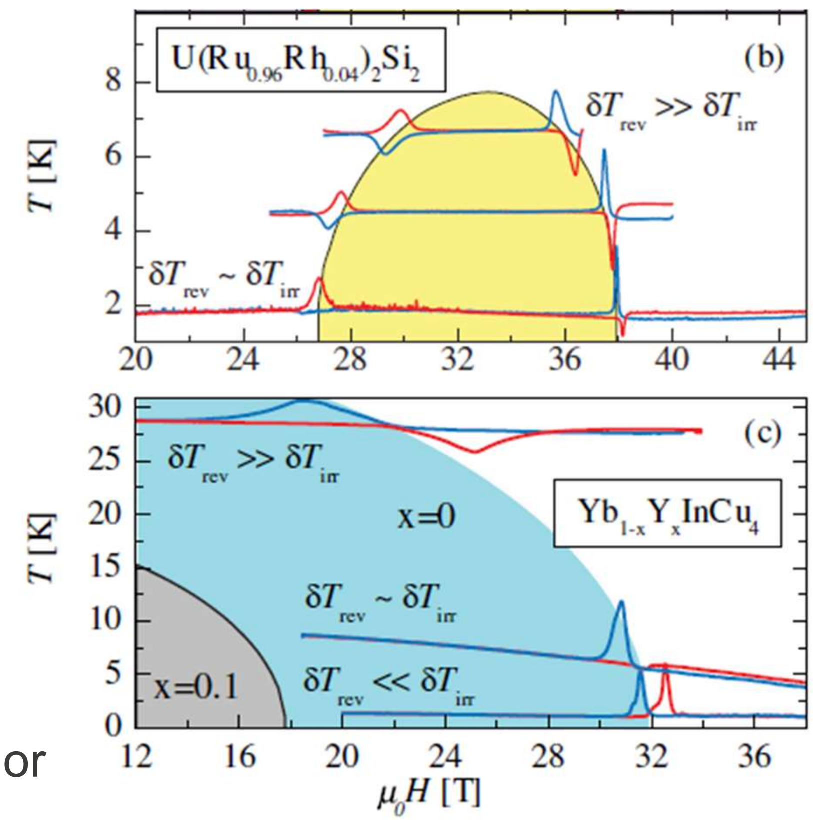
irreversible



α-$RuCl_3$ magnetocaloric effect

quasi-isothermal conditions





- Both transitions show irreversible behavior below 1K
- Phase transition above H_c ?

Silhanek *et al*. PRL 96, 136403 (2006)

Summary

- Confirmed phase diagram and strong magnetoelastic coupling in α-$RuCl_3$ with FBG and MCE measurements
- Thermal expansion coefficient displays a Schottky anomaly revealing an energy gap consistent with Majorana fermion + Z_2 flux excitations.
- Isothermal MCE reveals dissipative processes at low temperature indicating first order behavior. Sign of underlying FM interactions?
- We see very little, if any, entropy associated to the QSL-saturated PM boundary

Rico Schönemann, Shusaku Imajo, Franziska Weickert, Jiaqiang Yan, David G. Mandrus, Yasumasa Takano, Eric L. Brosha, Priscila F. S. Rosa, Stephen E. Nagler, Koichi Kindo, and Marcelo Jaime, *Phys. Rev. B* **102**, 214432 (2020)

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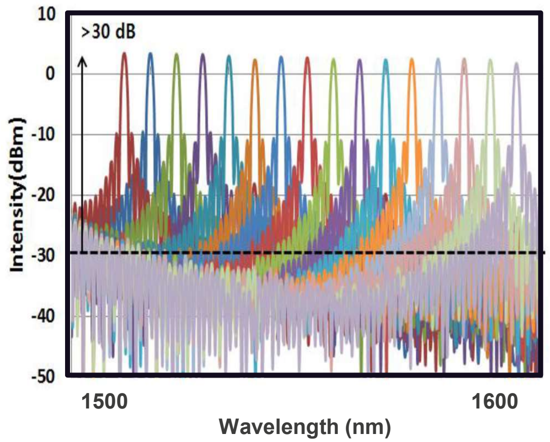
Priscilla Rosa
Eric Brosha
Marcelo Jaime

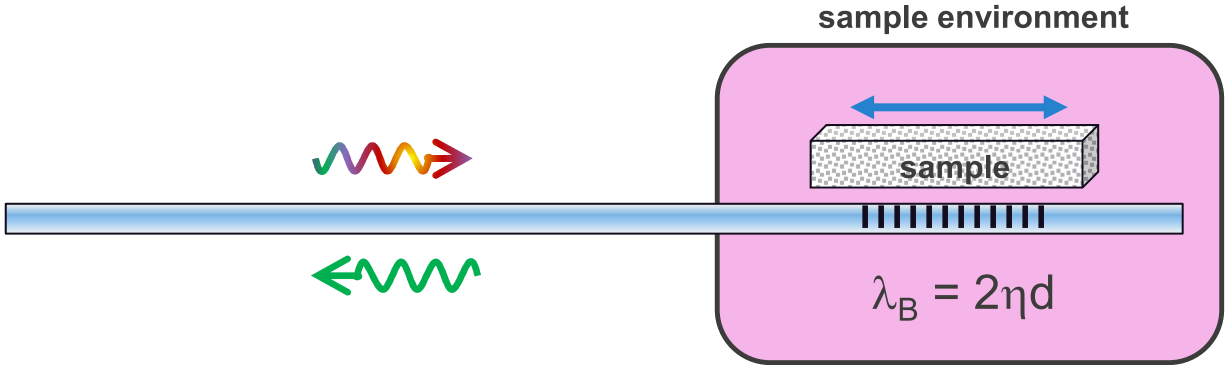
NATIONAL HIGH MAGNETIC FIELD LABORATORY

Los Alamos NATIONAL LABORATORY EST. 1943

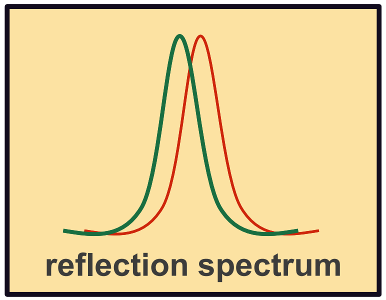
QUANTUM SCIENCE CENTER

FBG measurement technique





Micron Optics Hyperion
Swept wavelength light
source + detector



$\Delta L/L \propto \Delta\lambda_B/\lambda_B$

- Data acquisition rate: 5 kHz
- Resolution: $\Delta L/L \approx \mathbf{10^{-8}}$

10T anomaly: Phase transition vs crossover

PHYSICAL REVIEW LETTERS **125,** 097203 (2020)

Thermodynamic Perspective on Field-Induced Behavior of α-$RuCl_3$

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Measurements of the magnetic Grüneisen parameter (Γ_B) and specific heat on the Kitaev material candidate α-$RuCl_3$ are used to access in-plane field and temperature dependence of the entropy up to 12 T and down to 1 K. No signatures corresponding to phase transitions are detected beyond the boundary of the magnetically ordered region, but only a shoulderlike anomaly in Γ_B, involving an entropy increment as small as $10^{-5} R \log 2$. These observations put into question the presence of a phase transition between the purported quantum spin liquid and the field-polarized state of α-$RuCl_3$. We show theoretically that at low temperatures Γ_B is sensitive to crossings in the lowest excitations within gapped phases, and identify the measured shoulderlike anomaly as being of such origin. Exact diagonalization calculations demonstrate that the shoulderlike anomaly can be reproduced in extended Kitaev models that gain proximity to an additional phase at finite field without entering it. We discuss manifestations of this proximity in other measurements.

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