

Mottness-driven electronic BEC-BCS crossover in a doped spin liquid candidate

Kazushi Kanoda

Max Planck Institute for Solid State Research

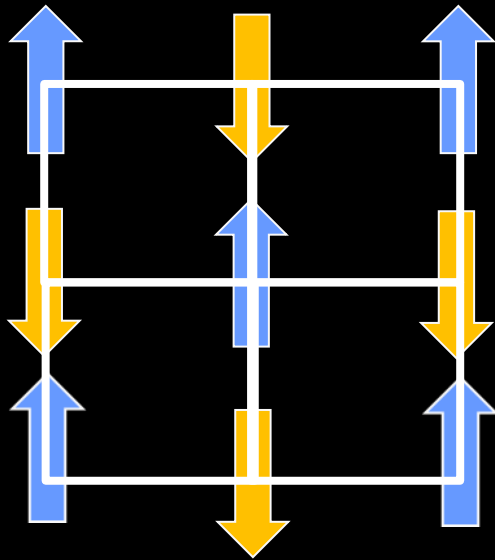
Physics Institute, University of Stuttgart

Department of Advanced Materials Science, University of Tokyo

- 1. Organic materials**
- 2. Quantum spin liquid and Mott transition**
- 3. Doped quantum spin liquid** - $\kappa\text{-(ET)}_4\text{Hg}_{2.89}\text{Br}_8$ -
 - non-FL to FL crossover
 - quantum critical phase
 - BEC-BCS crossover

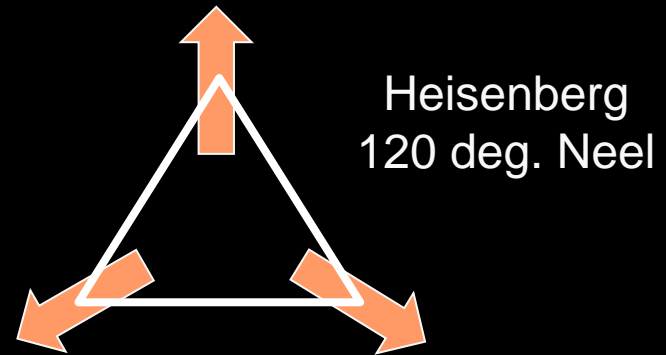
AF-interacting spins

happy on a square lattice



Neel state

unhappy on a triangular lattice



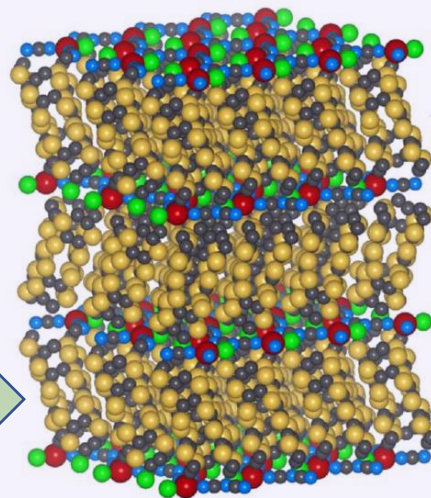
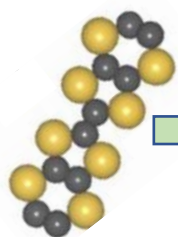
1973 P. W. Anderson
Quantum Spin liquid (QSL) ?

Organic materials

Electronic simplicity

Structural variety/compressibility

BEDT-TTF (ET)



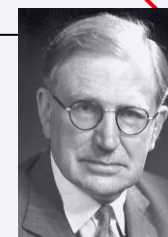
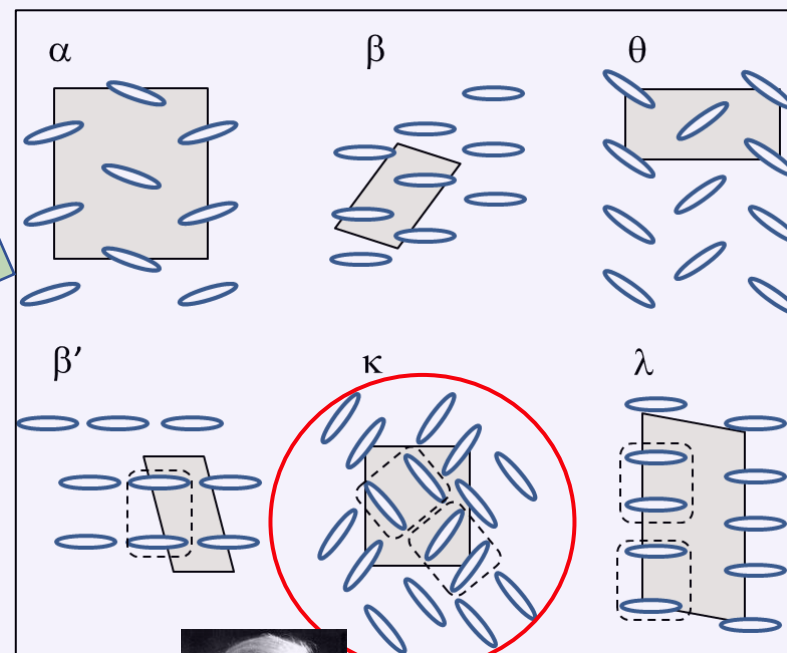
In-plane structure



Dirac electrons

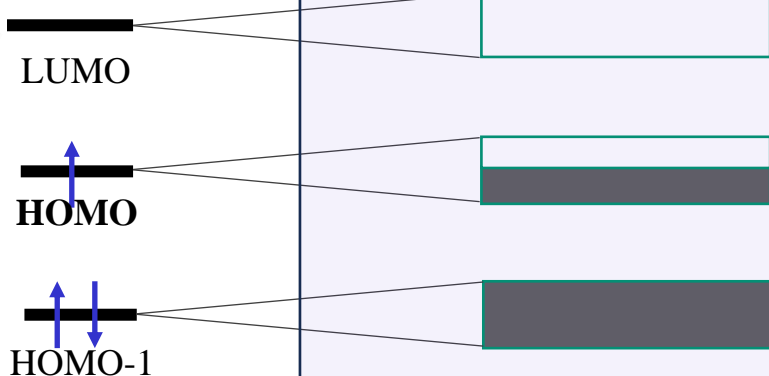


Wigner Xtal/glass



Mott physics

ϵ



LUMO

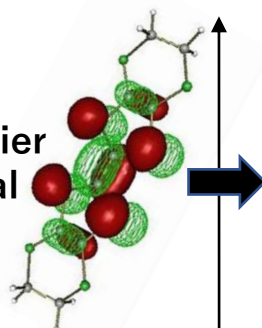
HOMO

HOMO-1

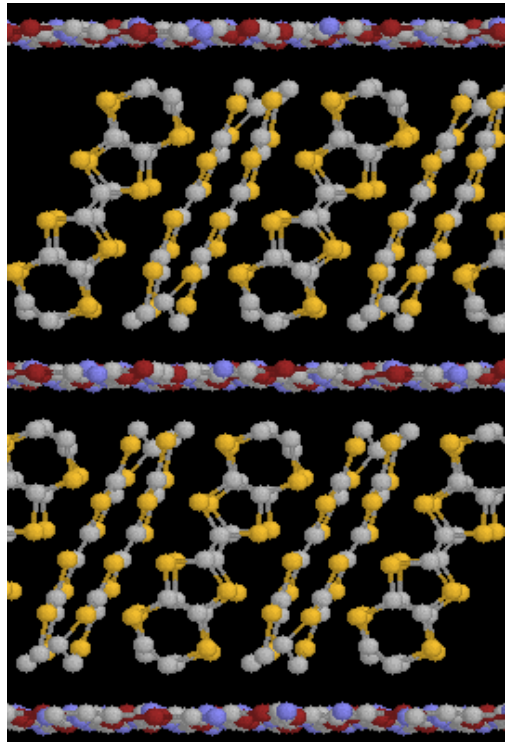
Molecular orbital

Electronic band

Frontier orbital

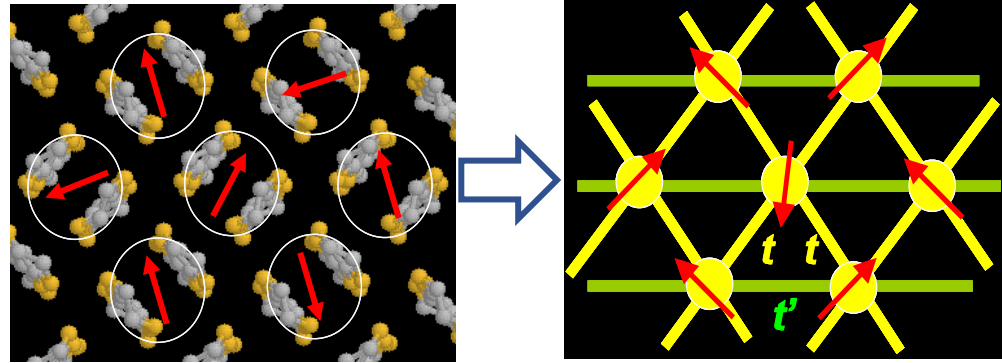


$\kappa\text{-ET}_2\text{X}$ Isosceles triangular lattice with a half-filled band

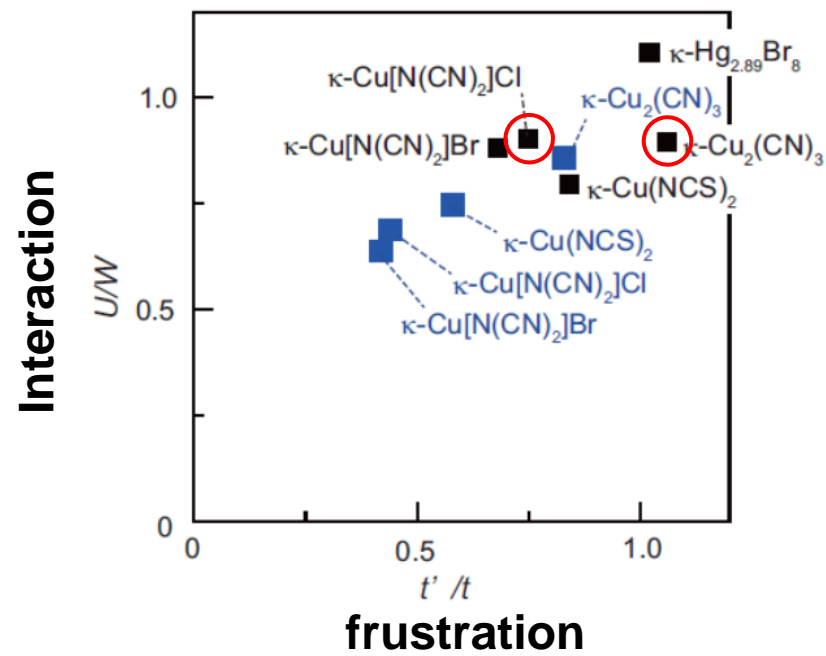


ET+0.5
X-1

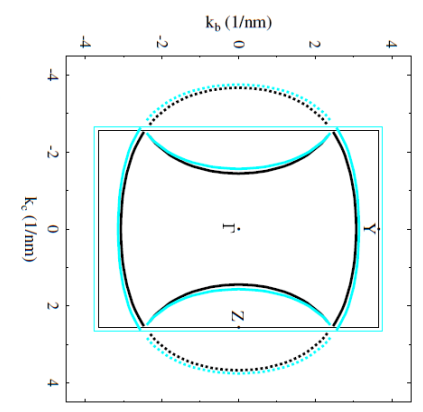
In-plane structure



■ Extended Hückel + tight binding
■ First principles calculation

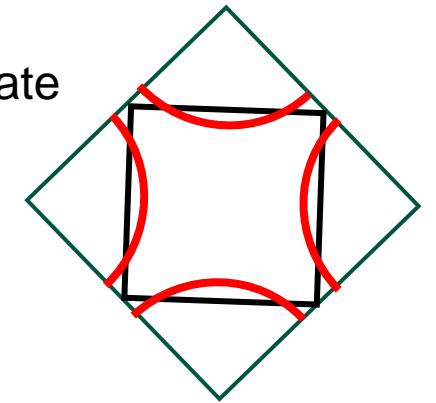


Fermi surface

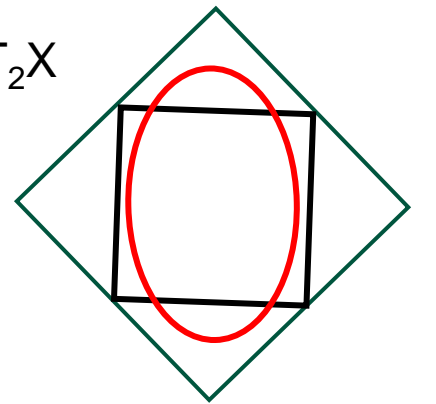


Kandpal *et al.*,
PRL 103, 067004 (2009)

cuprate



$\kappa\text{-ET}_2\text{X}$



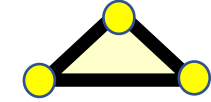
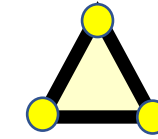
Quantum spin liquid (QSL) candidates with a triangular lattice

Shimizu et al.,
PRL (2003)

Phase diagram



^1H NMR spectra

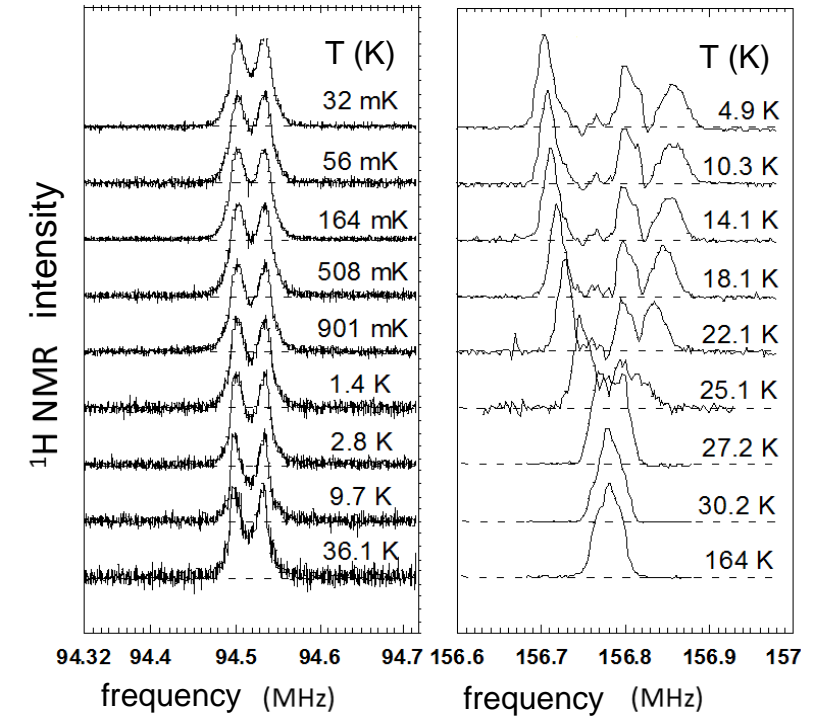


Triangular lattice

Deformed triangular lattice

$\kappa\text{-(BEDT-TTF)}_2\text{Cu}_2(\text{CN})_3$

$\kappa\text{-(BEDT-TTF)}_2\text{Cu}[\text{N}(\text{CN})_2]\text{Cl}$



No magnetic order

Magnetic order



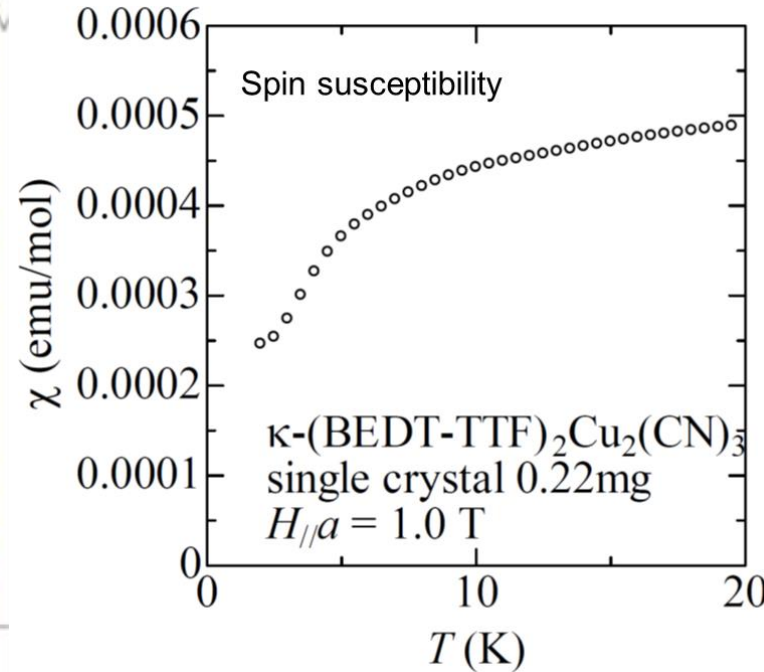
QSL !



AFM

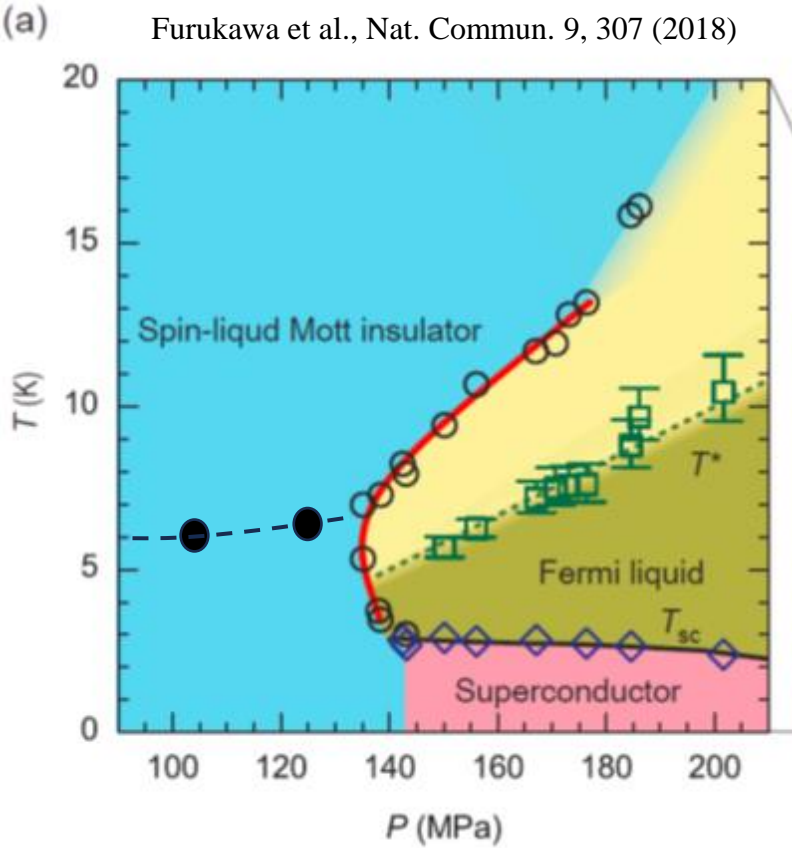
Furukawa et al., Nat. Commun. 9, 307 (2018)

Spin susceptibility



Negligible impurity spins

Vicinity of Mott transition

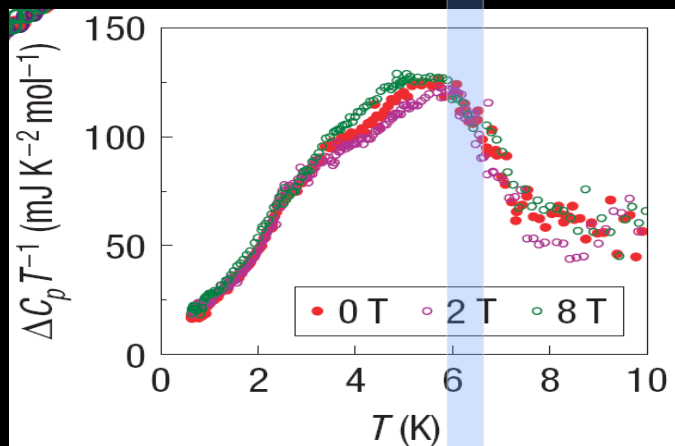


6K-anomaly in $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

$\Delta \sim 10 \text{ K}$

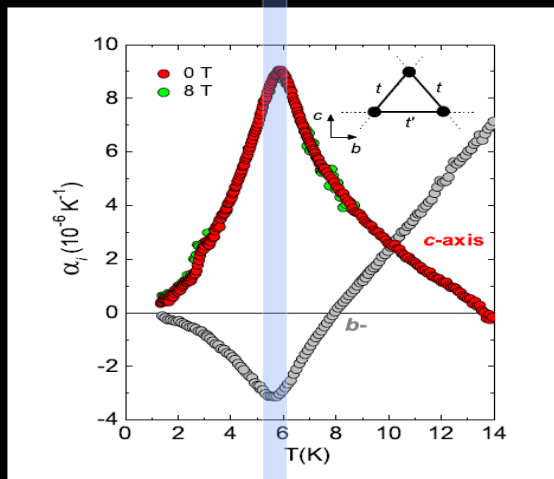
Specific heat

S. Yamashita *et al.*, *Nat. Phys.* **4**, 4559 (2008).



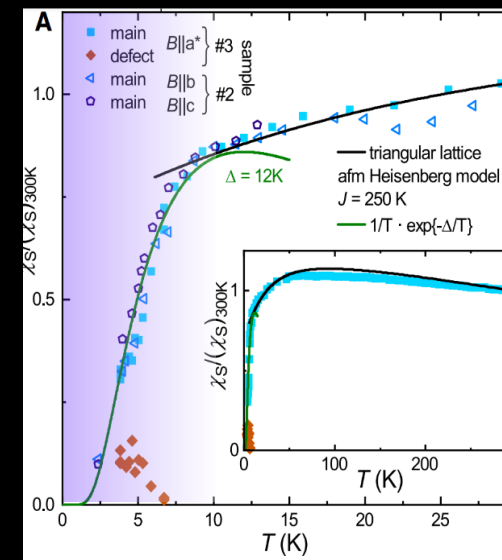
Thermal expansion coefficient

Manna *et al.*, *PRL* **104** (2010) 016403



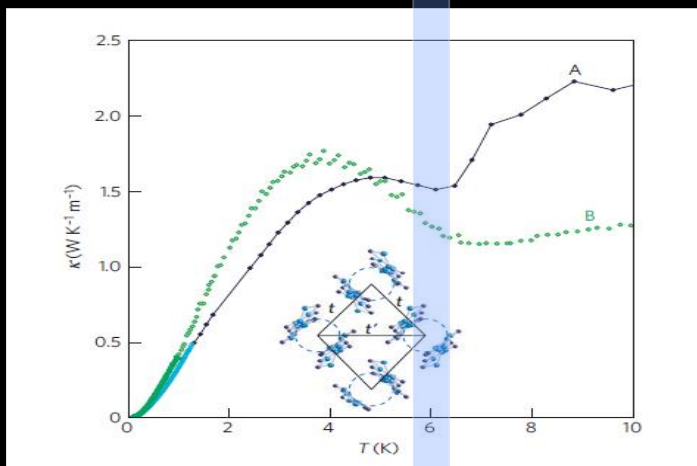
ESR intensity

Miksch *et al.* *Science* **372**, 276 (2021)



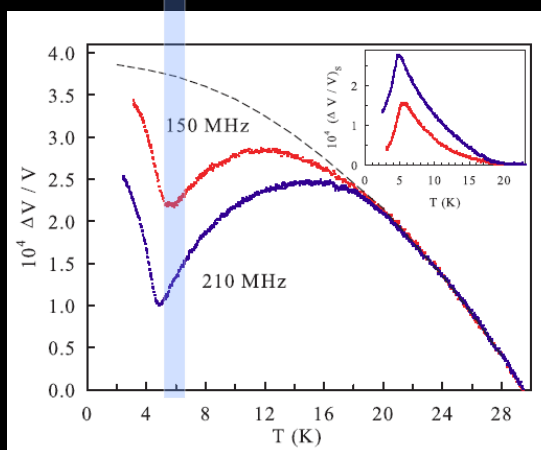
Thermal conductivity

M. Yamashita *et al.*, *Nat. Phys.* **5**, 44 (2009).

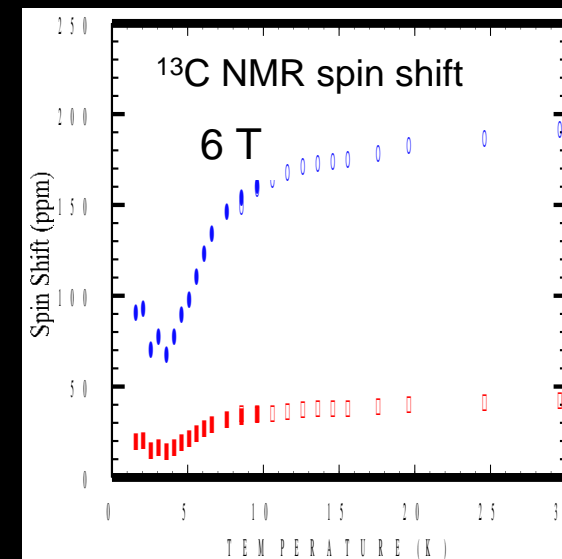


Ultrasound velocity

Poirier *et al.*,



^{13}C NMR spin shift

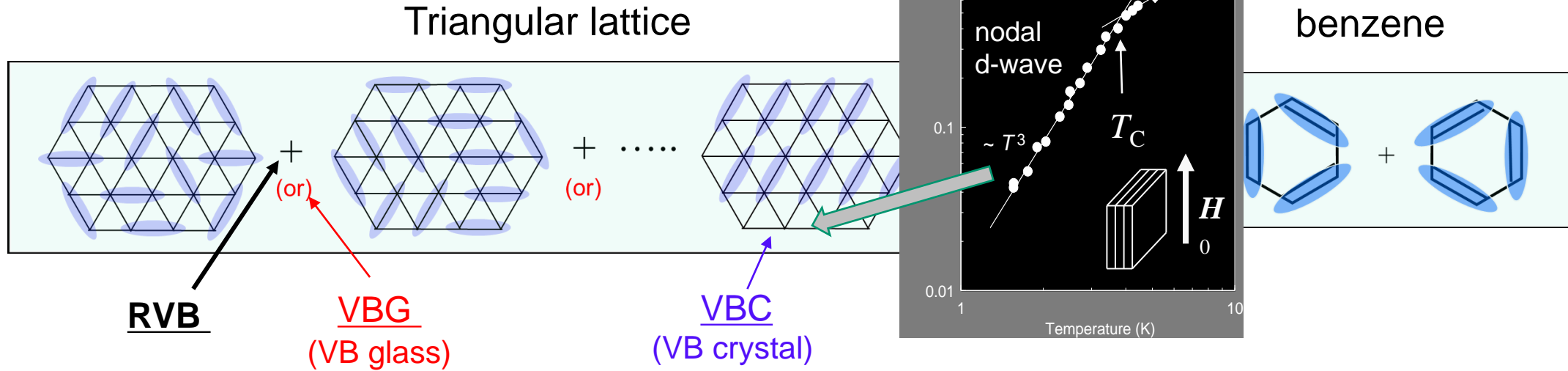


Quantum spin liquid (QSL) on triangular lattices



P. W. Anderson

Resonating valence bond (RVB)



Various types
of QSLs

- gapped $|RVB\rangle = P_G |BCS\rangle$
- gapless $|spinon FS\rangle = P_G |BCS; \Delta \rightarrow 0\rangle$
- gapless $|spinon Dirac cone\rangle = P_G |nodal BCS\rangle$
- gapped $|Chiral spin liquid\rangle = P_G |chiral SC\rangle$

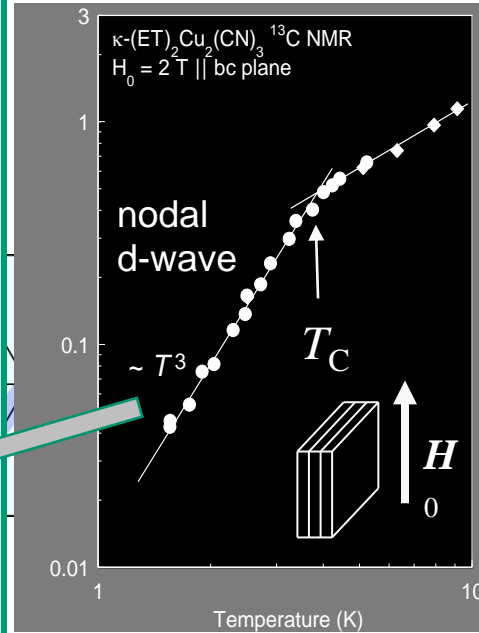
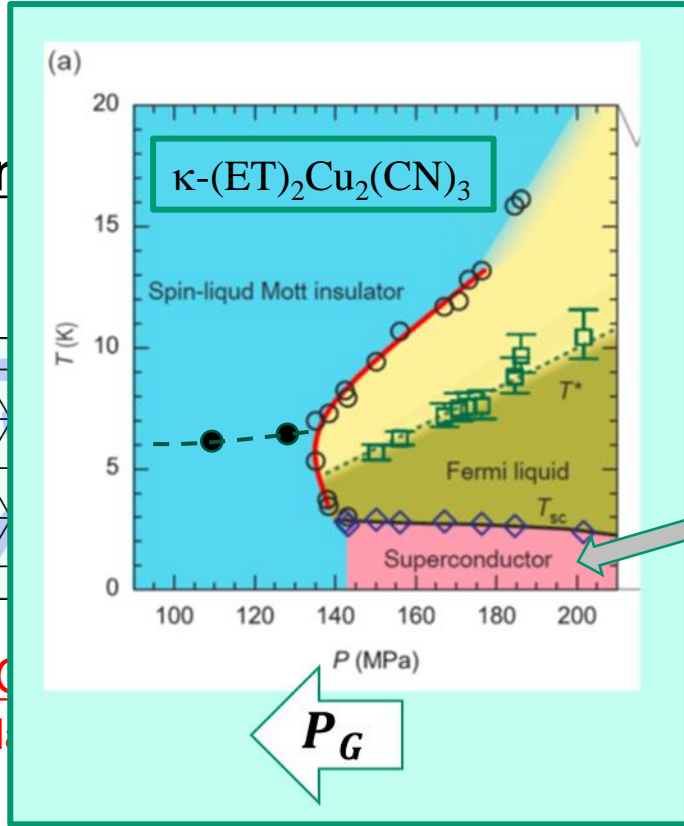
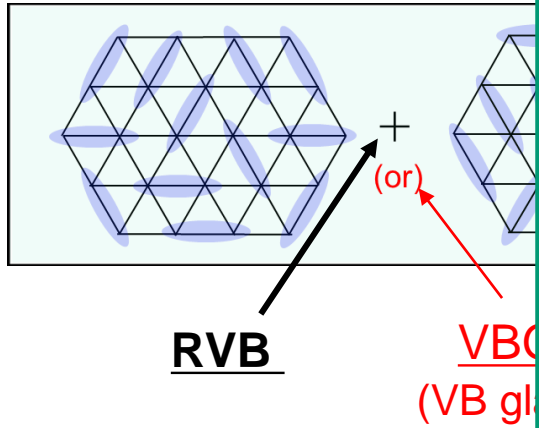
(P_G : projection operator
to remove double occupancies)
($P_G \sim$ Depressurize)

Quantum spin liquid (QSL) on triangular lattices

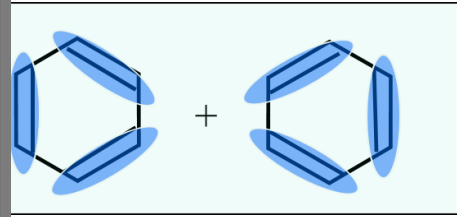


P. W. Anderson

Resonating



benzene

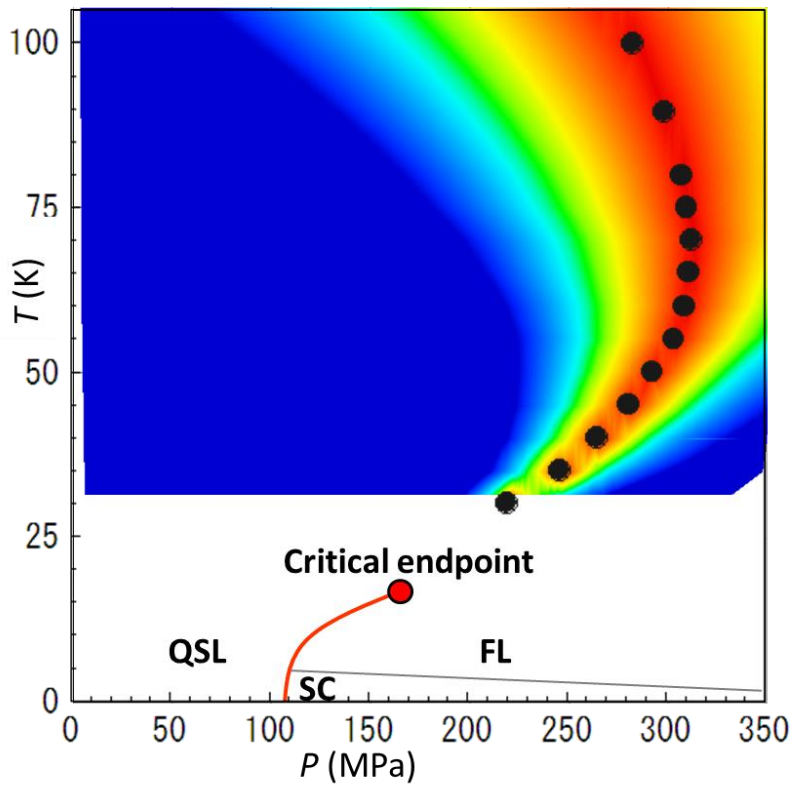


Various types of QSLs

- gapped $|RVB\rangle = P_G |BCS\rangle$
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- gapless $|spinon Dirac cone\rangle = P_G |nodal BCS\rangle$
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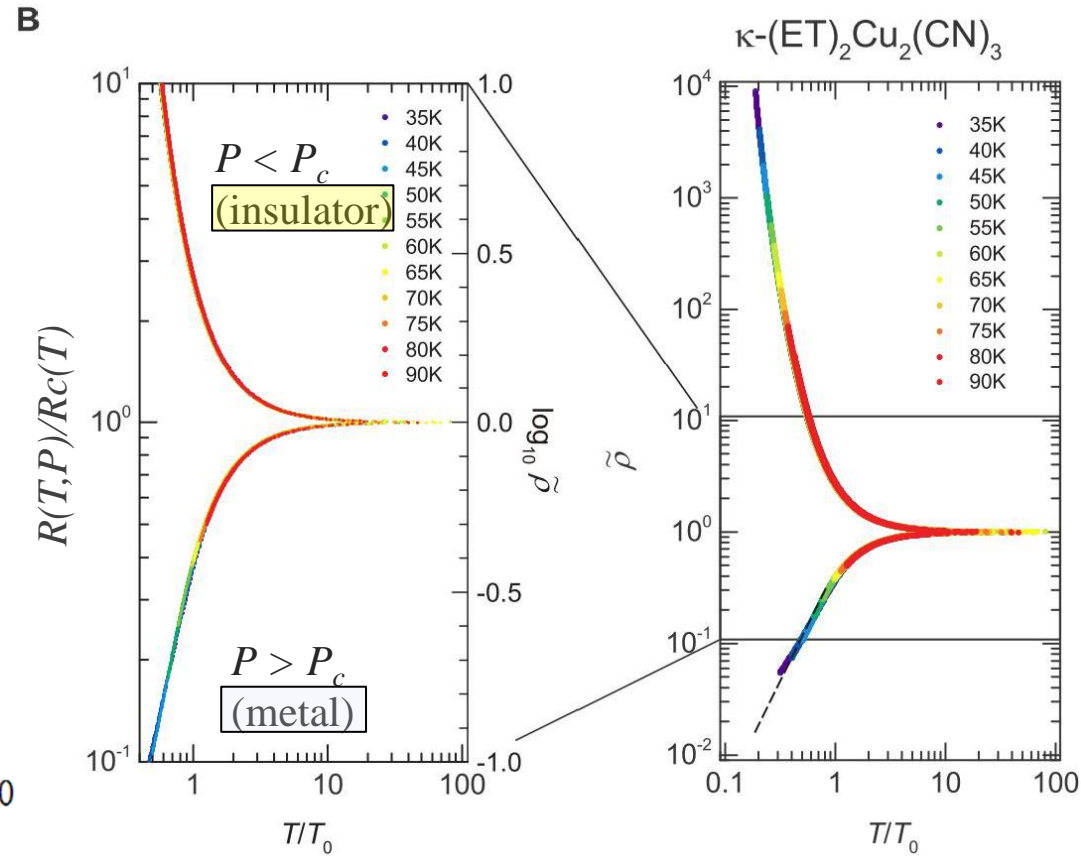
(P_G : projection operator to remove double occupancies)
 ($P_G \sim$ Depressurize)

Mott quantum critical scaling of resistivity



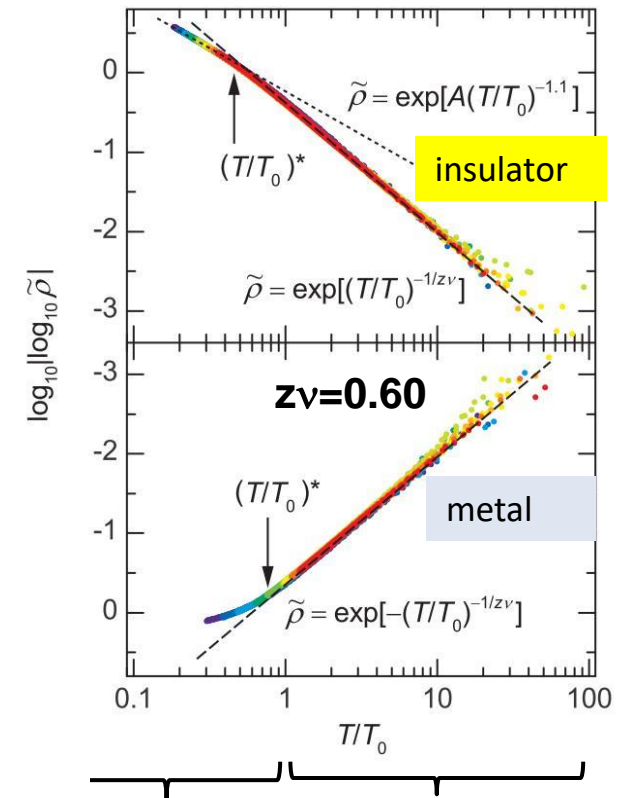
Furukawa et al., Nat. Phys **11**, 221 (2015)

Furukawa et al., Nat. Commun. **9**, 307 (2018)

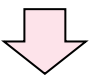


Dynamics determined by the ratio of thermal fluctuations to quantum fluctuations

$$T/T_0 = T / (c |P - P_c(T)|^{z\nu})$$

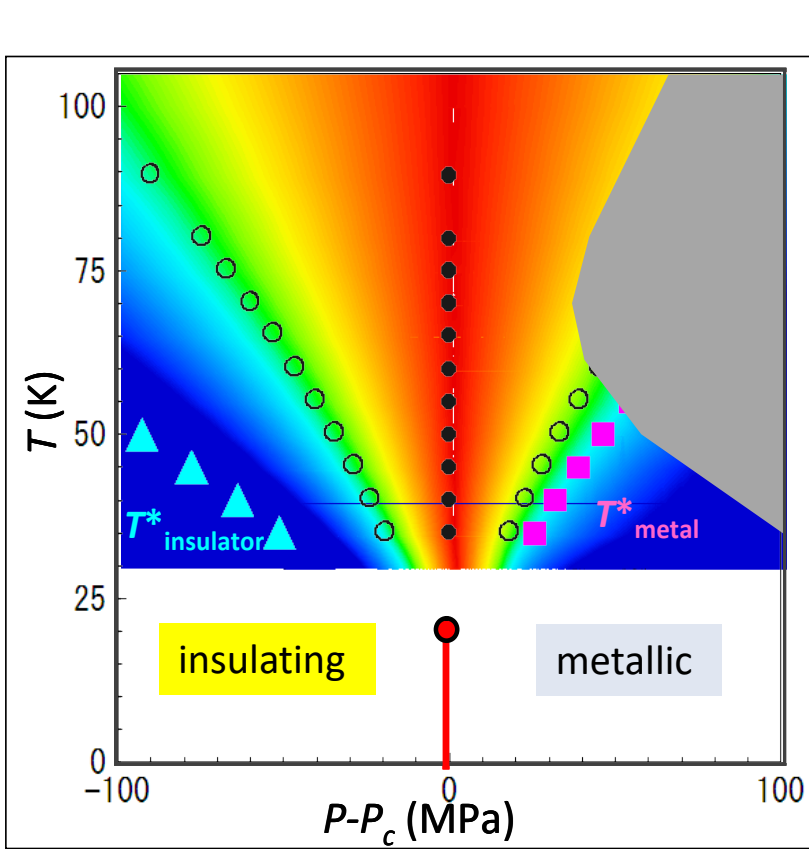


quasi-particles Quantum critical



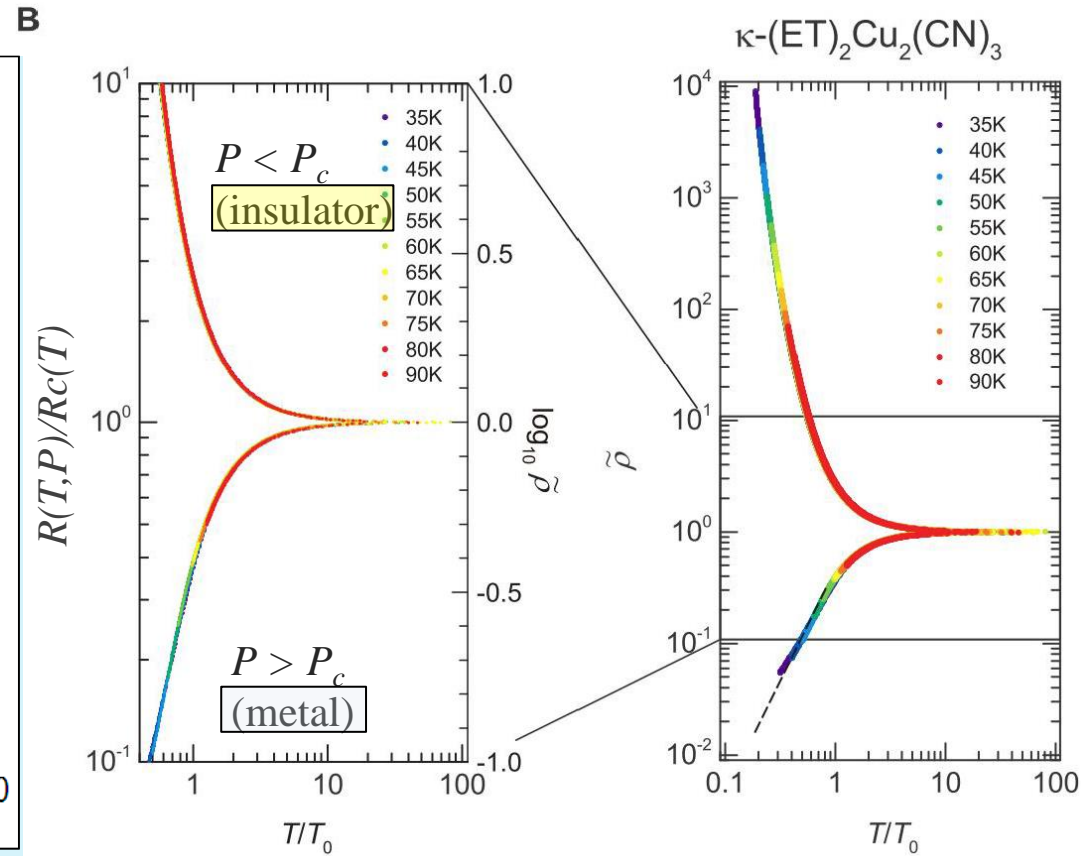
Holon/doublon density fluctuations

Mott quantum critical scaling of resistivity



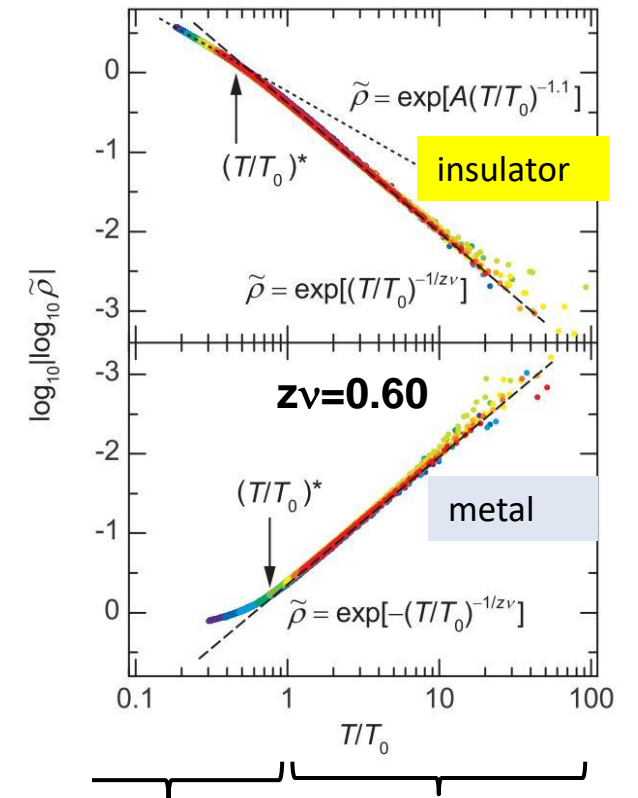
Furukawa et al., Nat. Phys **11**, 221 (2015)

Furukawa et al., Nat. Commun. **9**, 307 (2018)



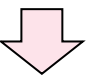
Dynamics determined by the ratio of thermal fluctuations to quantum fluctuations

$$T/T_0 = T / (c |P - P_c(T)|^{z\nu})$$



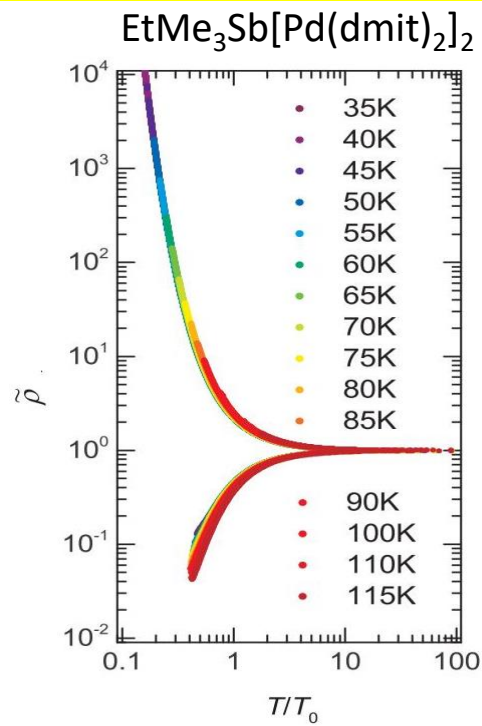
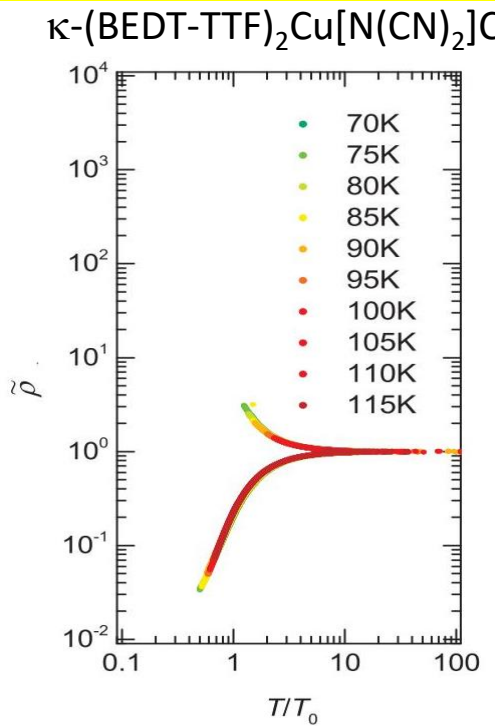
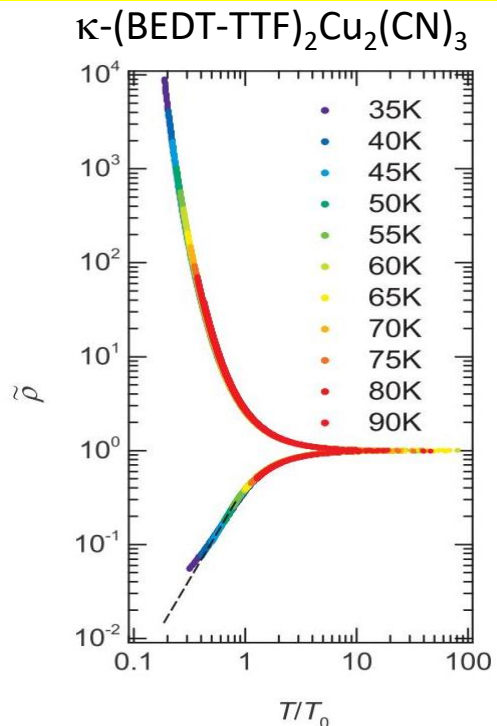
quasi-particles

Quantum critical



Holon/doublon density fluctuations

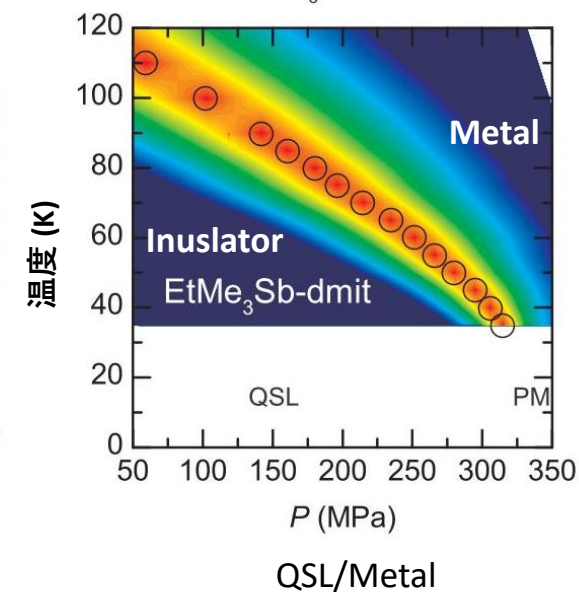
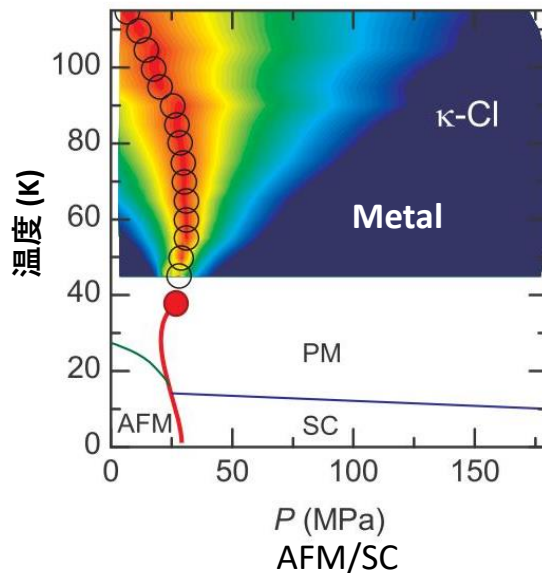
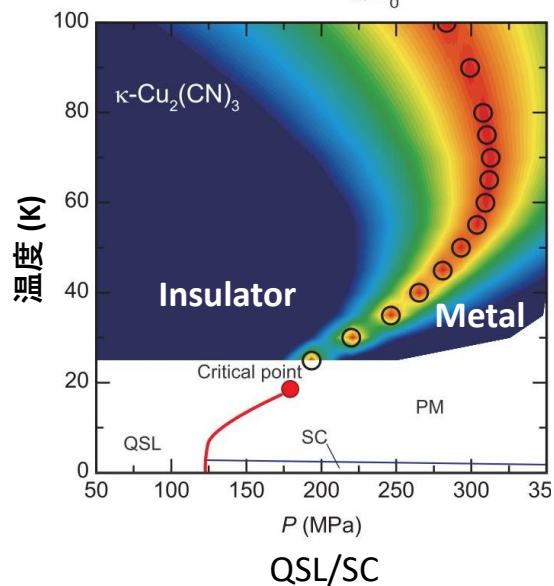
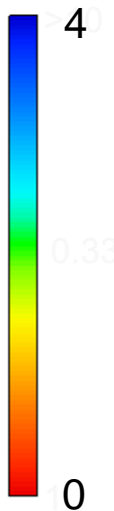
Material-independent universal quantum critical fluid ($z\nu=0.5-0.7$)



*Holographic duality
AdS/CFT*

*Furukawa et al.,
Nat. Phys 11 (2015) 221*

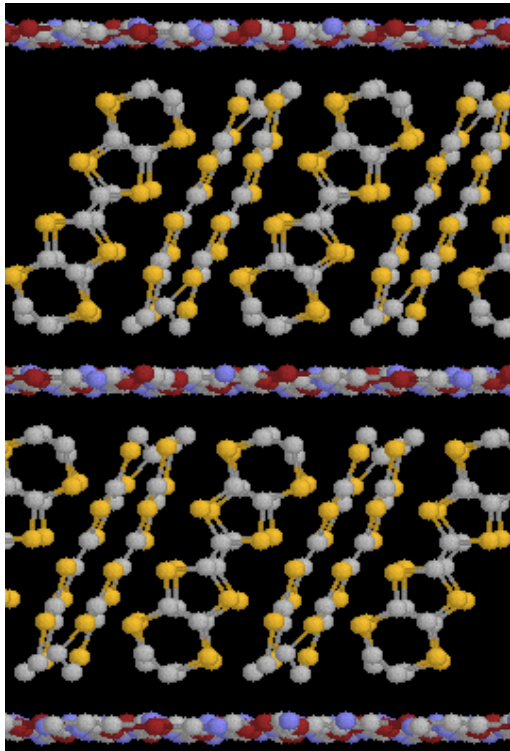
$|\text{Log}(R/R_c)|$



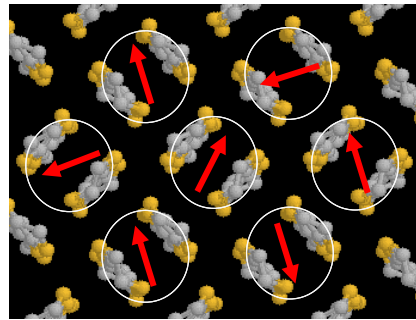
Doped spin liquid candidate $\kappa\text{-(ET)}_4\text{Hg}_{2.89}\text{Br}_8$ (11% hole doping)



Incommensurate sublattice of Hg against ET lattice



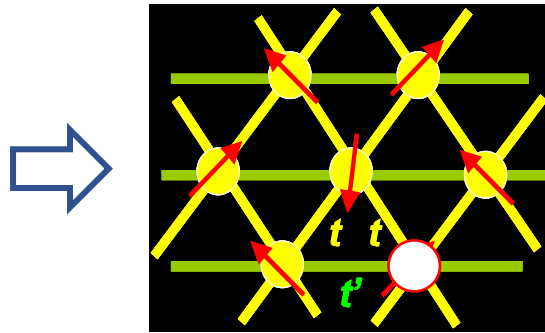
ET



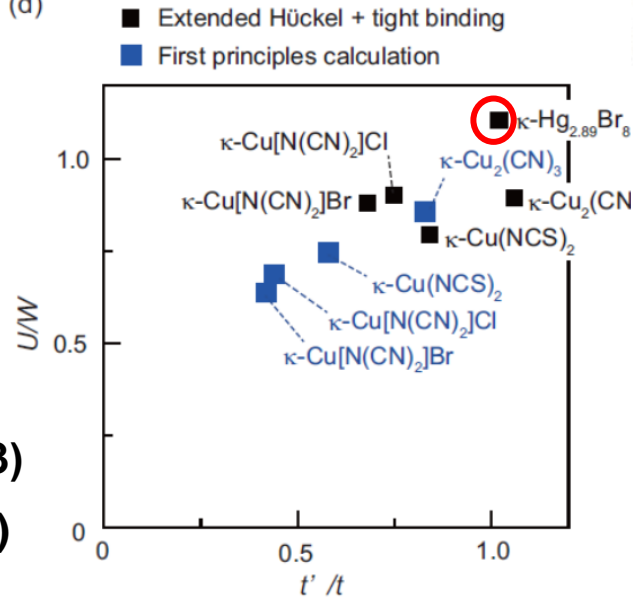
Hg_{2.89}Br₈

3.00 → half filling

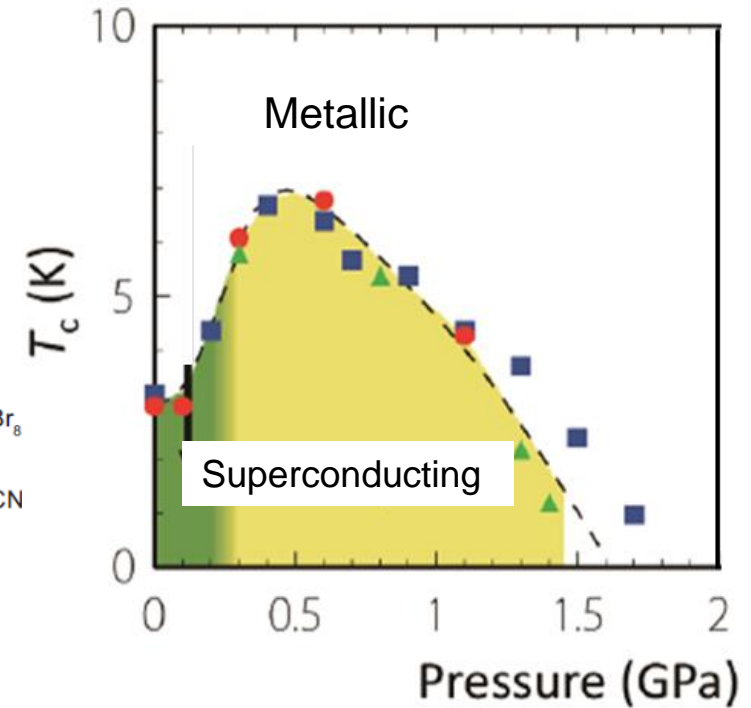
- 11% hole doping
- Triangular lattice ($t'/t=1.03$)
- Large U/W value ($U/W=1.1$)



(d)



Superconducting dome

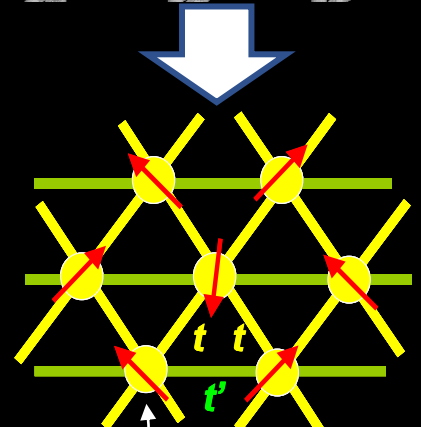
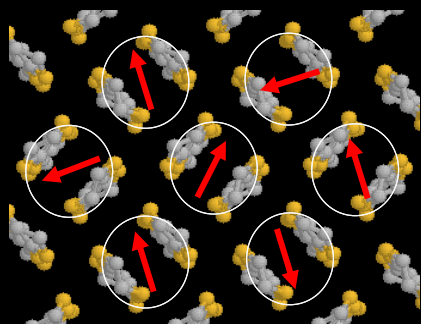


A doped spin liquid ?





triangular lattice

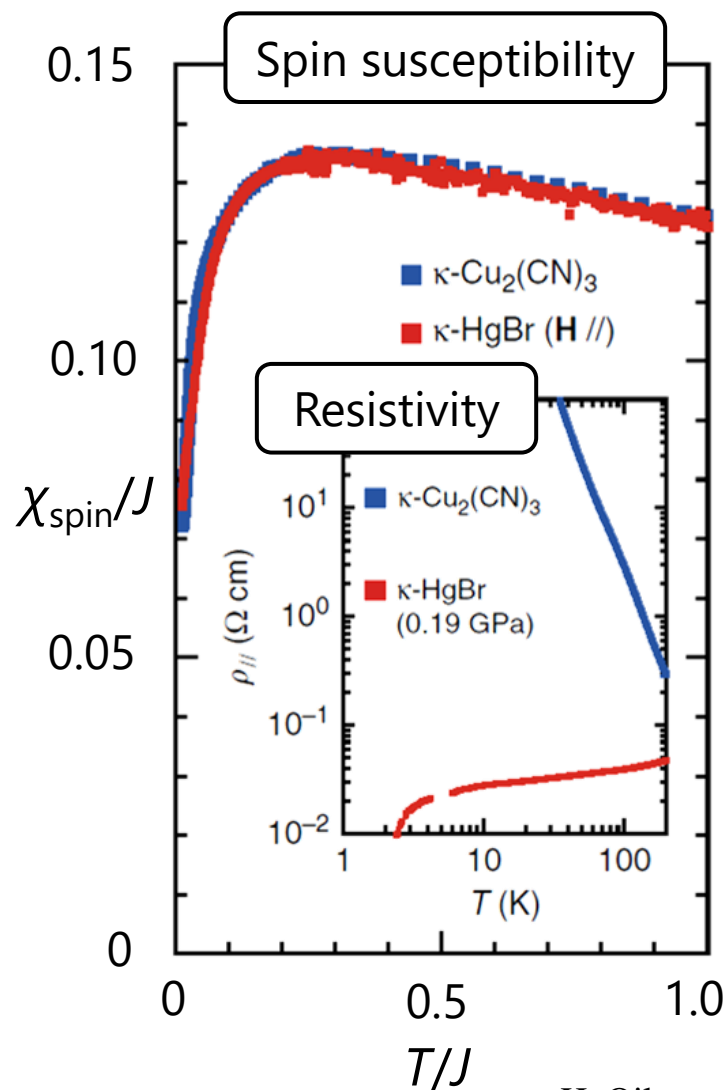


11% vacant !

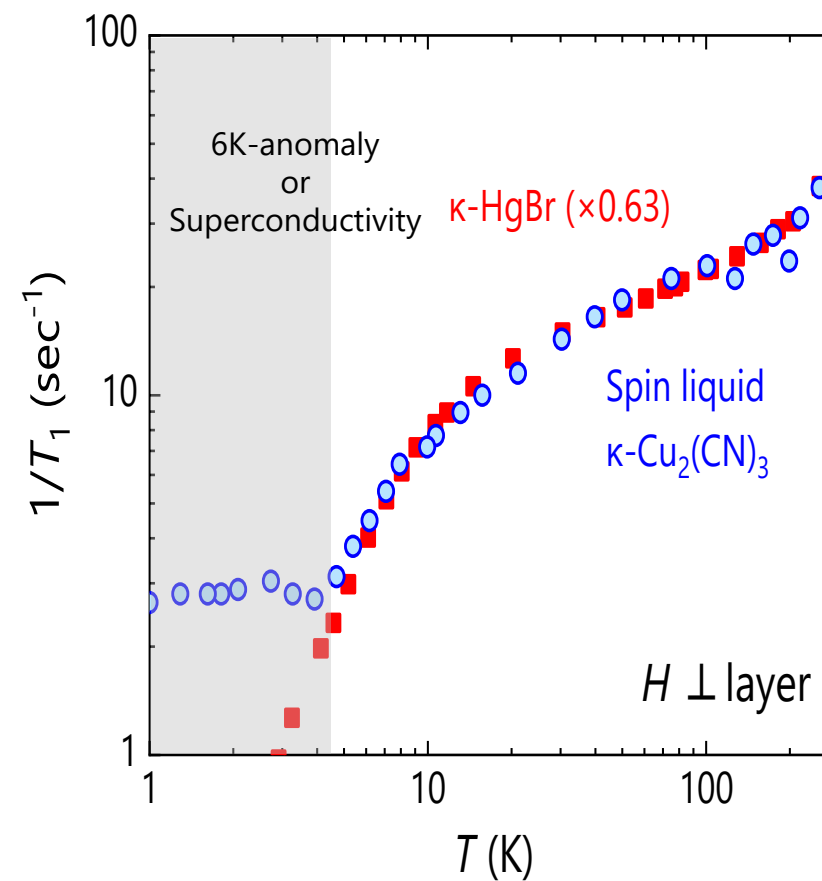
Magnetism remains QSL-like

Conductivity: insulator \rightarrow metal

\rightarrow Doped QSL



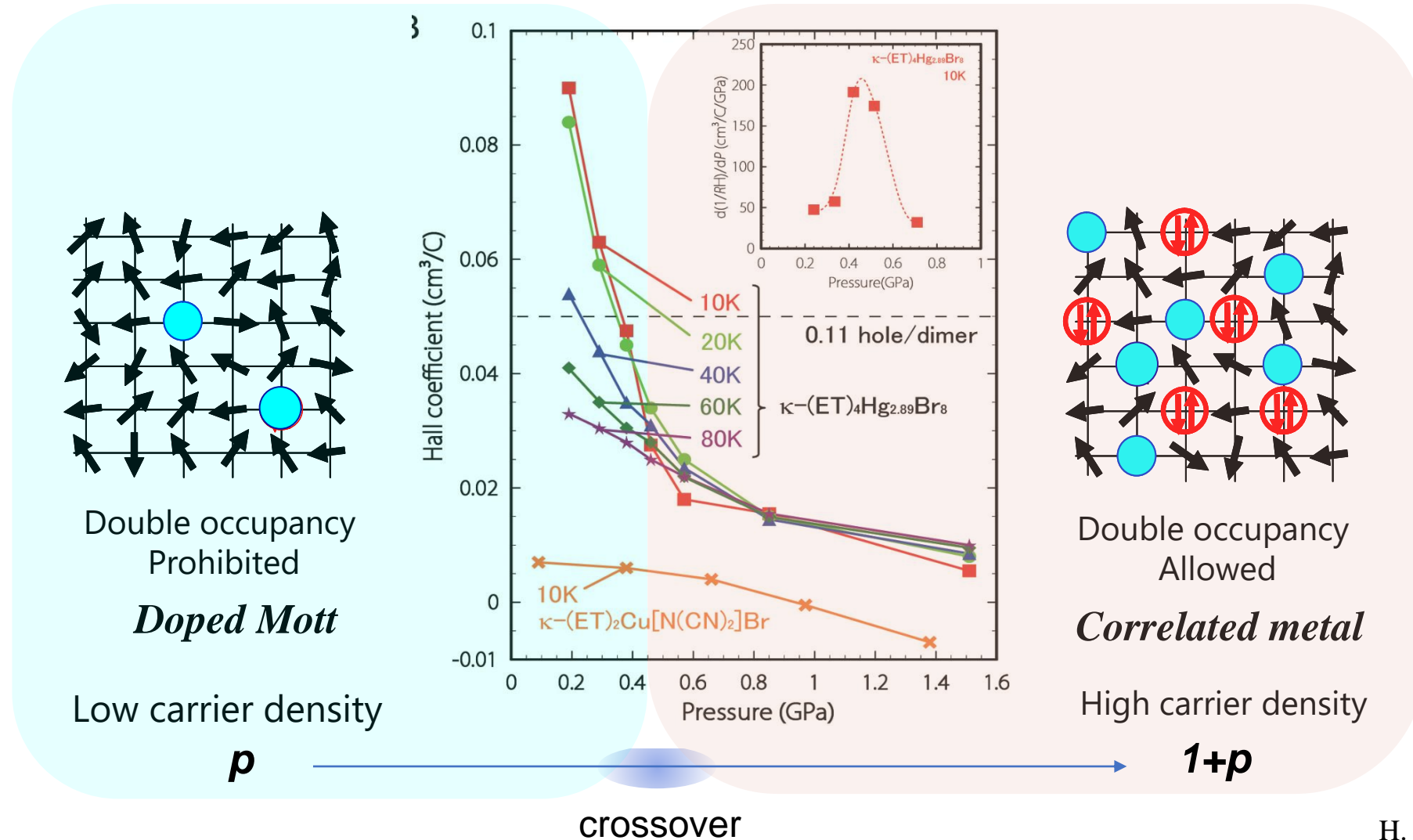
^{13}C NMR relaxation rate



Pressure dependence of Hall coefficient: from doped QSL to correlated metal

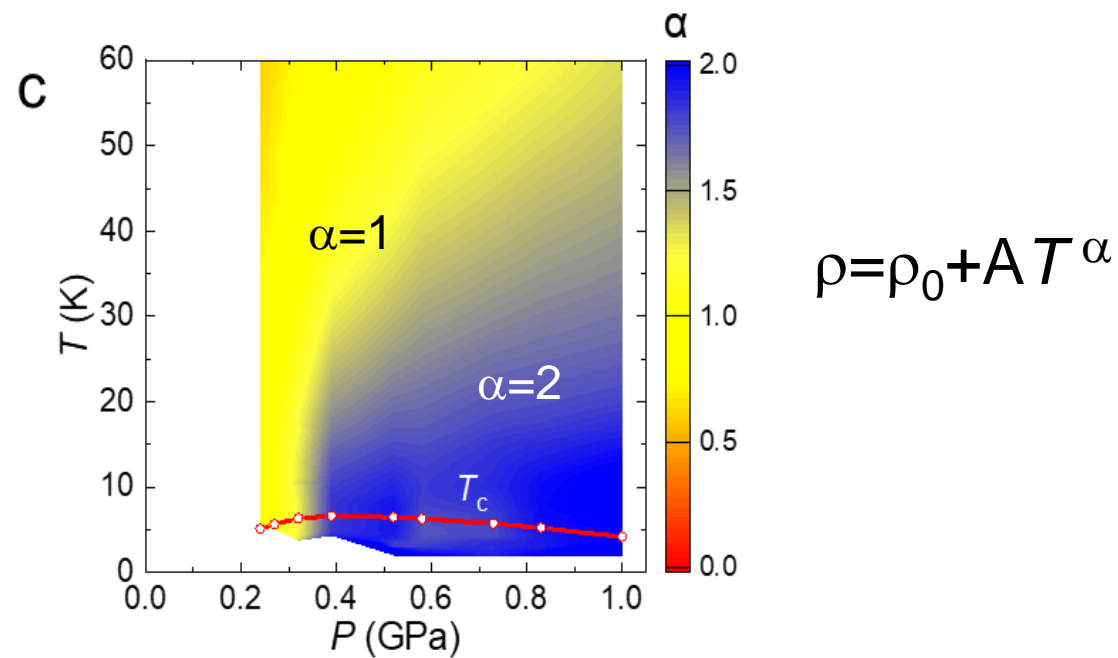
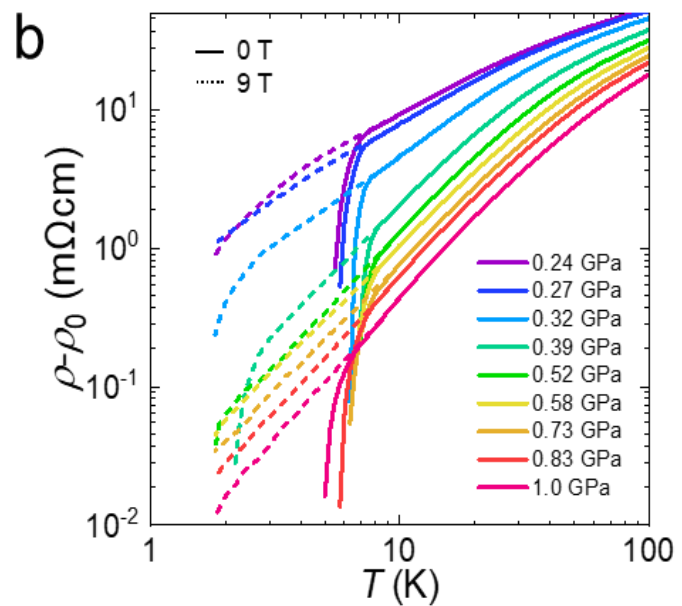
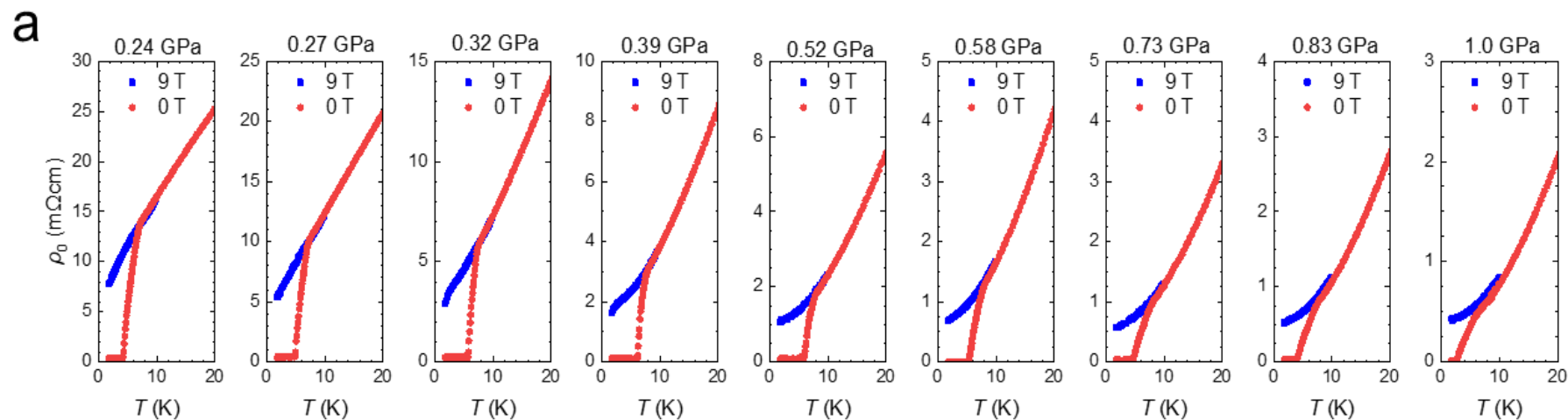


$$\text{Hall coefficient} \sim (\text{carrier number})^{-1}$$



Non-Fermi liquid to Fermi liquid crossover

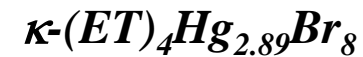
$\kappa\text{-(ET)}_4\text{Hg}_{2.89}\text{Br}_8$



Crossover from a conventional metal to a strange metal by pressure

Resistivity
non-Fermi liq. to fermi liq.

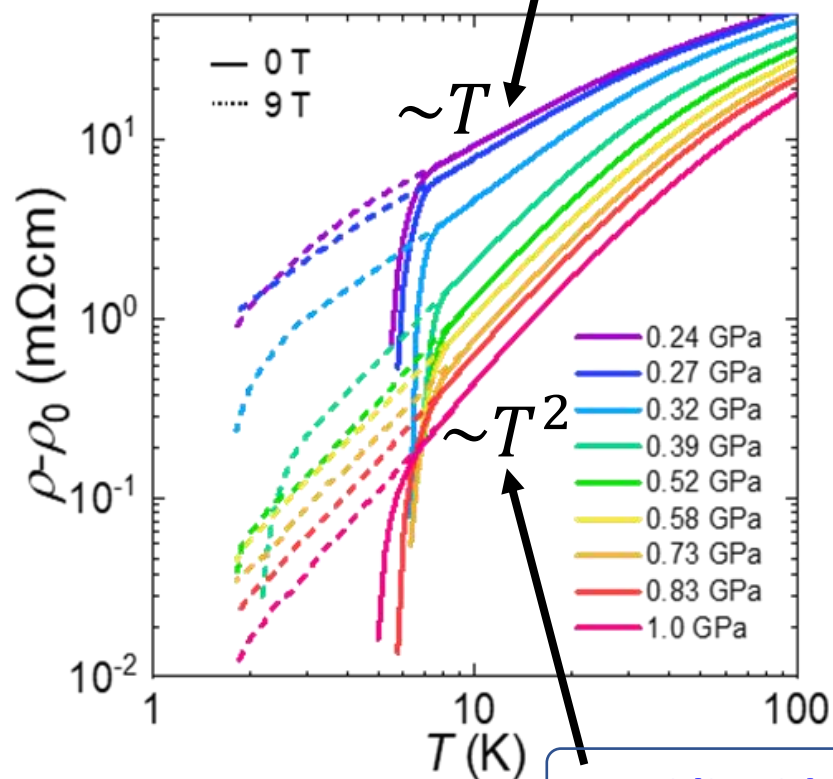
Seebeck coefficient
Quantum critical metal to normal metal



Non-Fermi liquid

Low P.

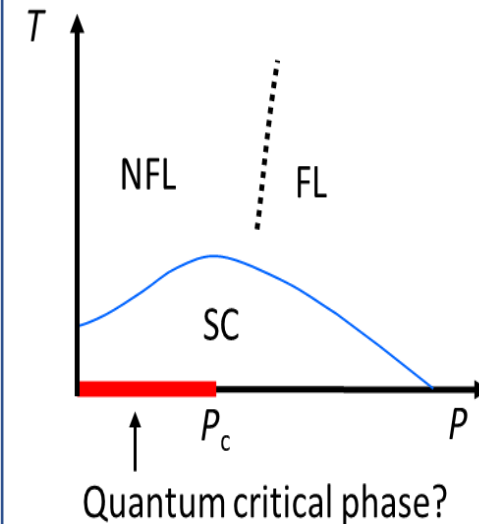
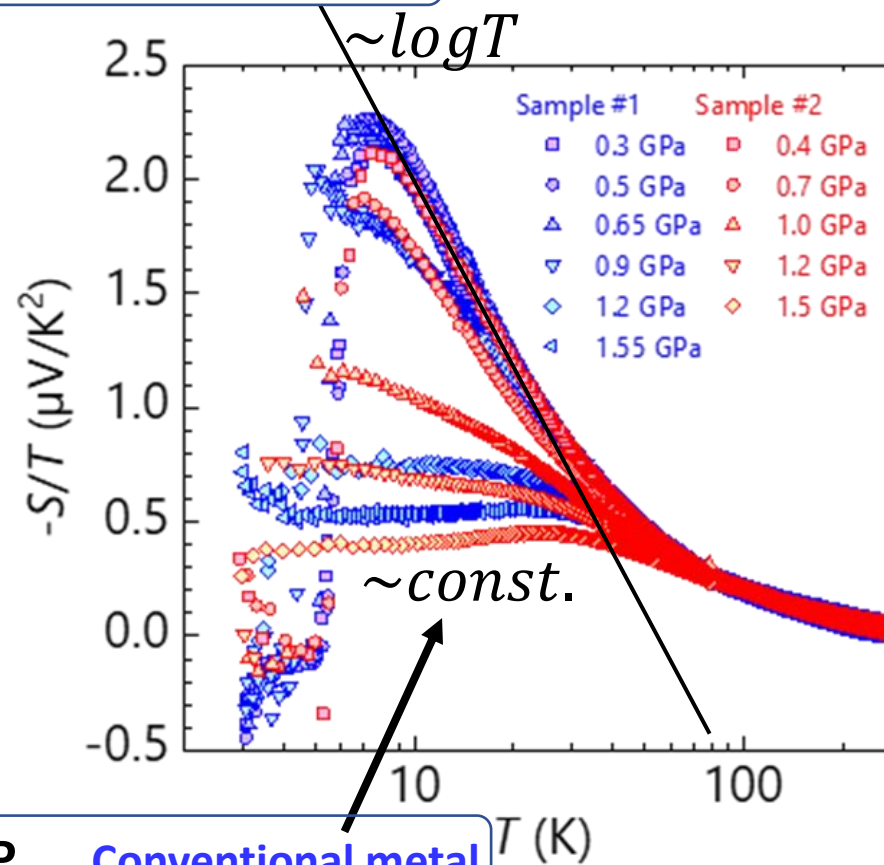
quantum critical

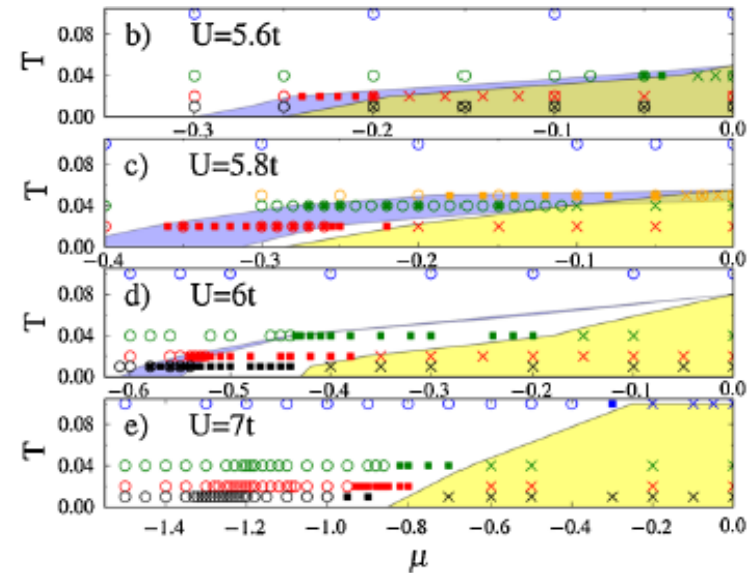
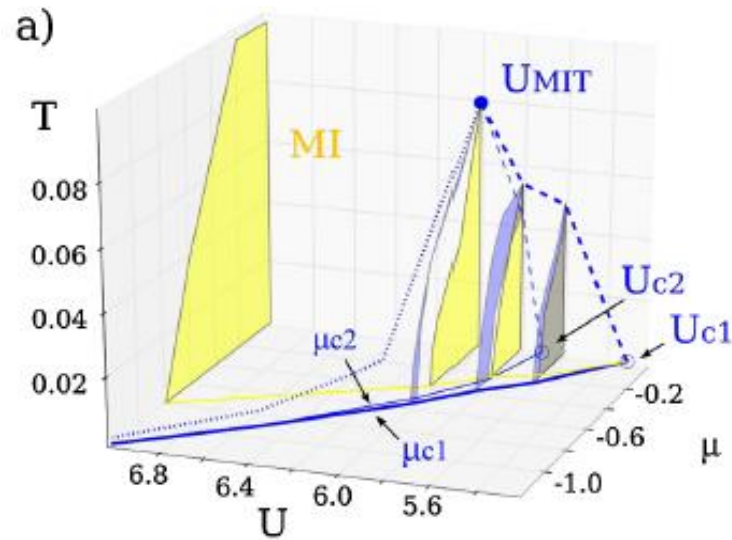


Fermi liquid

High P.

Conventional metal

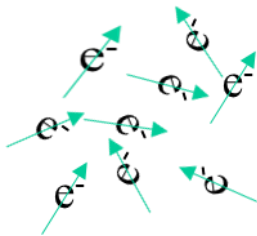


Finite Doping Signatures of the Mott Transition in the Two-Dimensional Hubbard ModelG. Sordi,¹ K. Haule,² and A.-M. S. Tremblay^{1,3}

Pressure-induced BEC to BCS

$\kappa\text{-(ET)}_4\text{Hg}_{2.89}\text{Br}_8$

Y. Suzuki *et al.*,
Phys. Rev. X **12**, 011016 (2022).



Strong interaction

Weak interaction

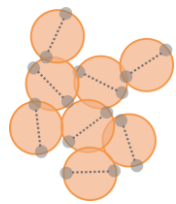
preformed pairs



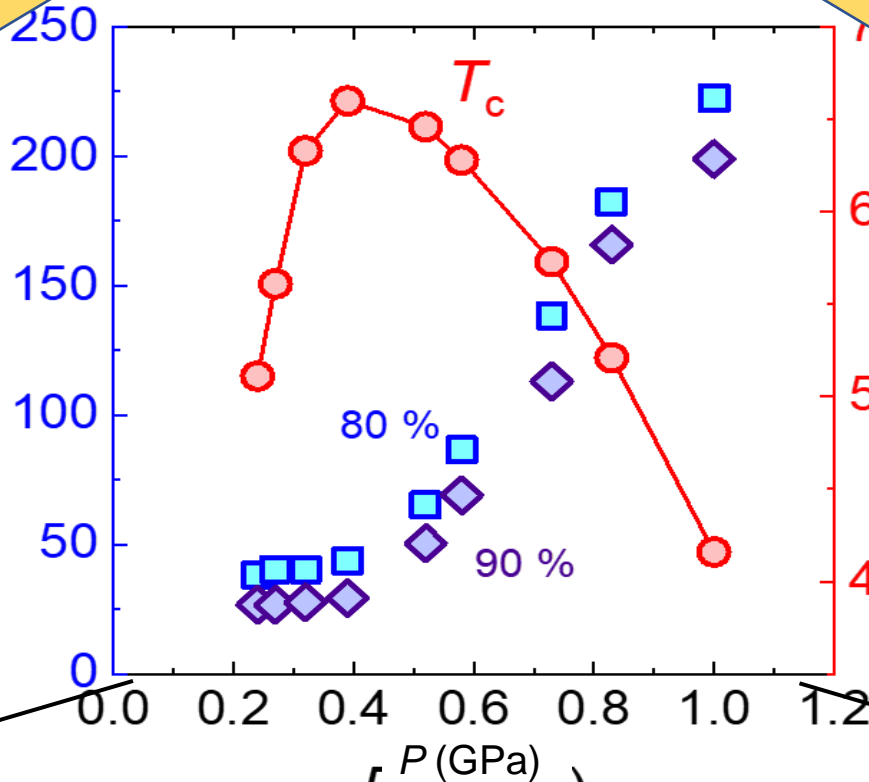
T_c

BEC

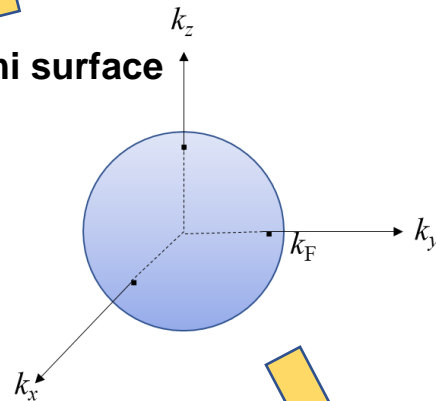
$k_F \xi \sim 2-3$
BEC-like



$\xi_{||}$ (Å)

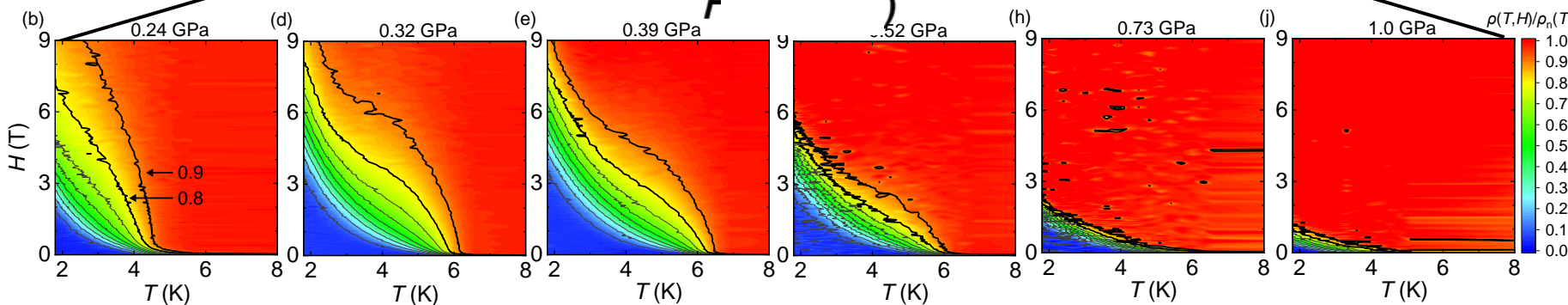
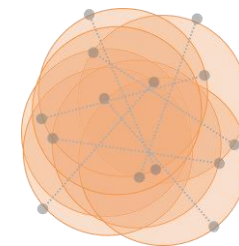


Fermi surface



BCS

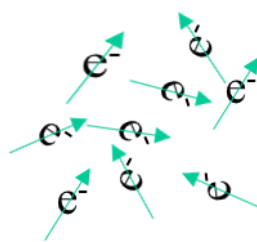
$k_F \xi \sim 50$
BCS-like



Pressure-induced BEC to BCS

$\kappa\text{-(ET)}_4\text{Hg}_{2.89}\text{Br}_8$

Y. Suzuki *et al.*,
Phys. Rev. X **12**, 011016 (2022).



Strong interaction

Weak interaction

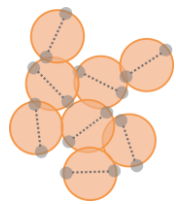
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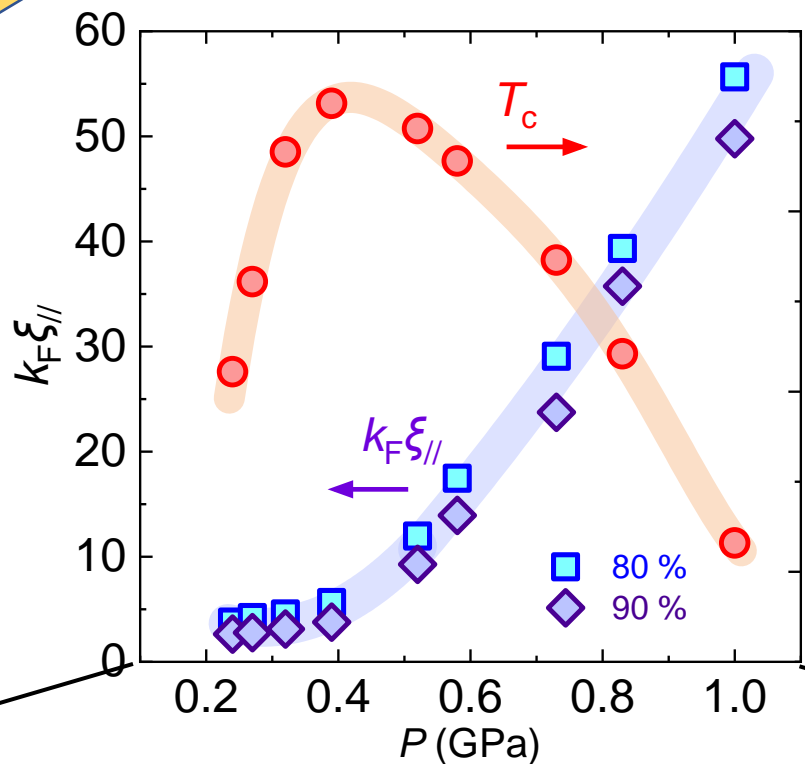
T_c

BEC

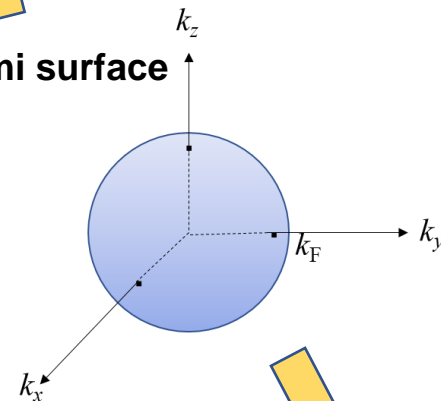
$k_F \xi \sim 2-3$
BEC-like



$\xi_{||}$ (Å)

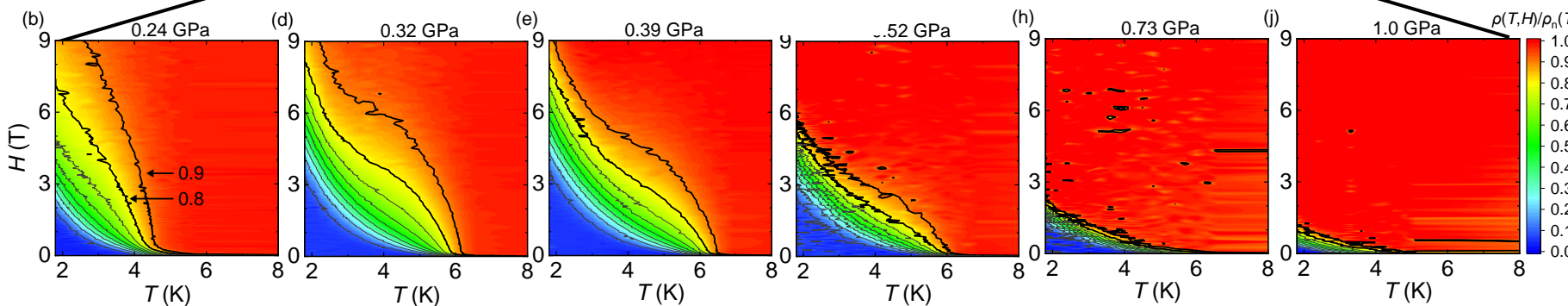
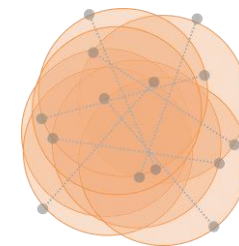


Fermi surface



BCS

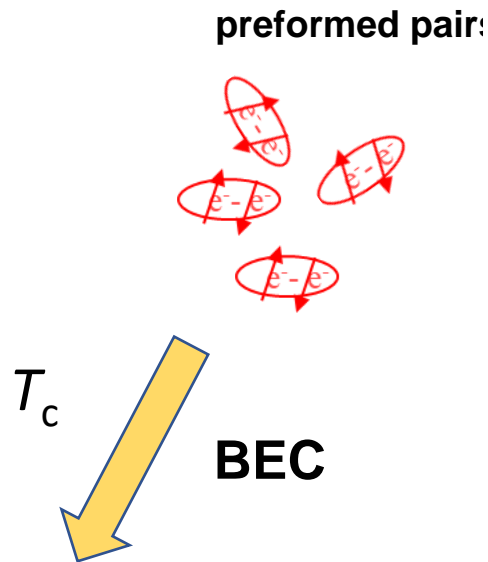
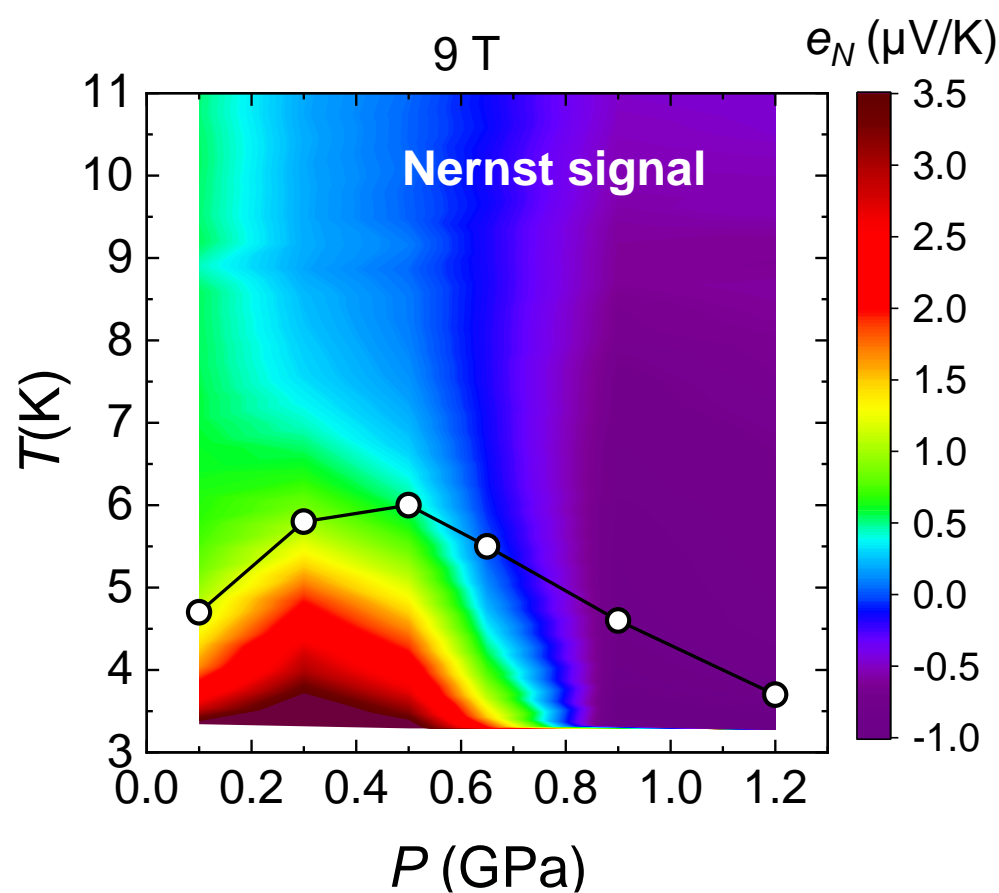
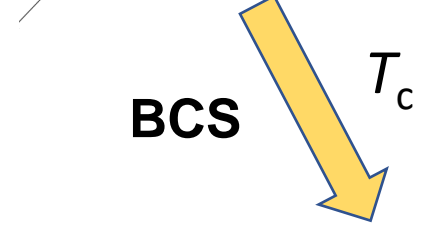
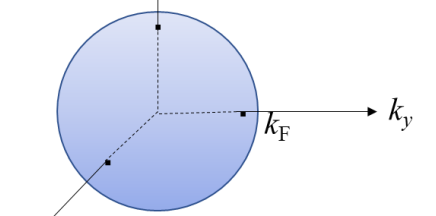
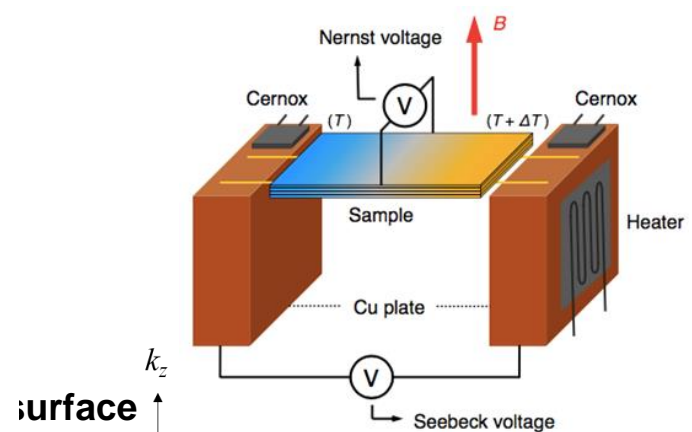
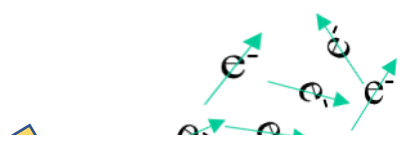
$k_F \xi \sim 50$
BCS-like



Pressure-induced BEC to BCS

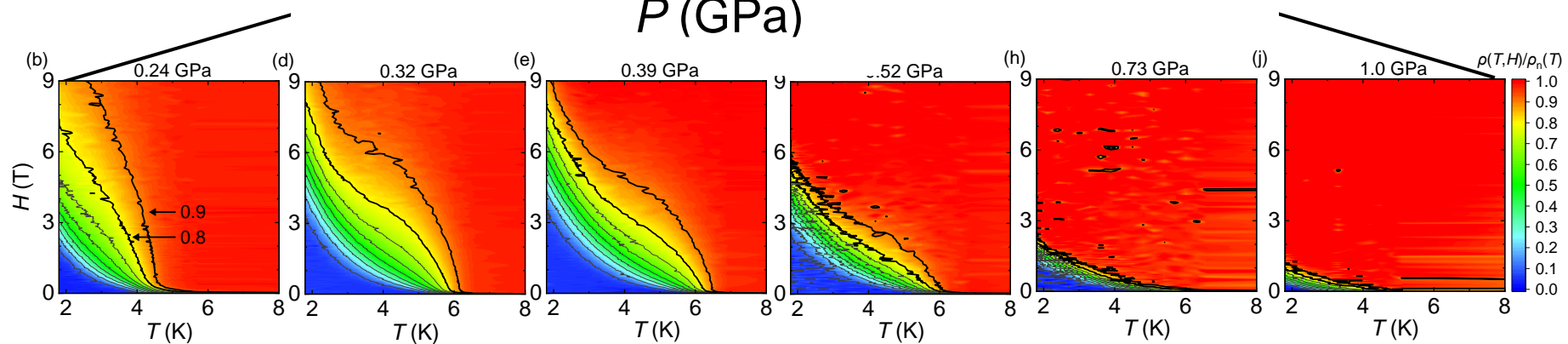
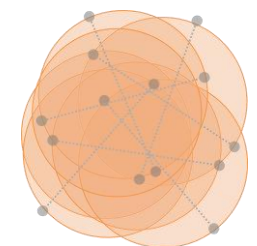
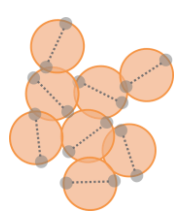


Y. Suzuki *et al.*,
Phys. Rev. X 12, 011016 (2022).

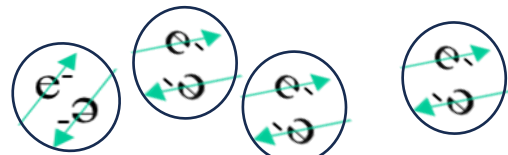


$k_F \xi \sim 2-3$
BEC-like

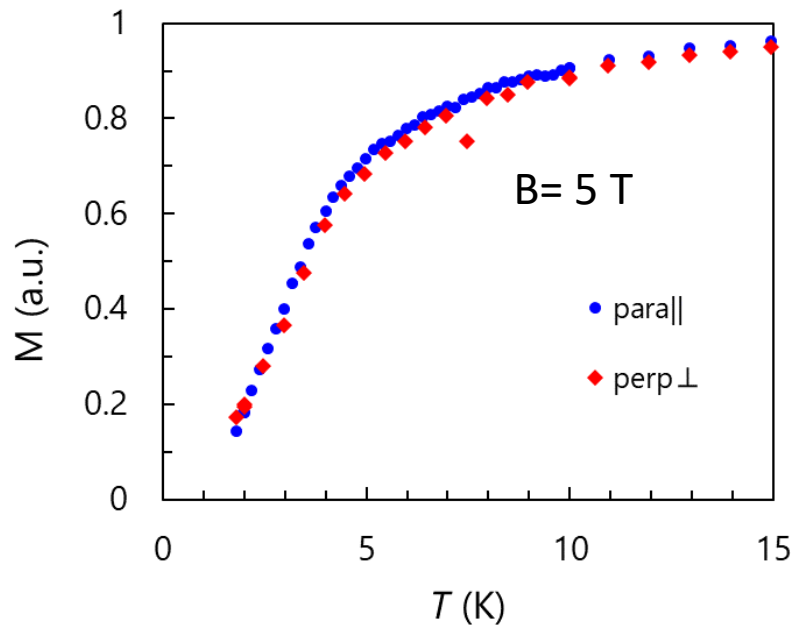
$k_F \xi \sim 50$
BCS-like



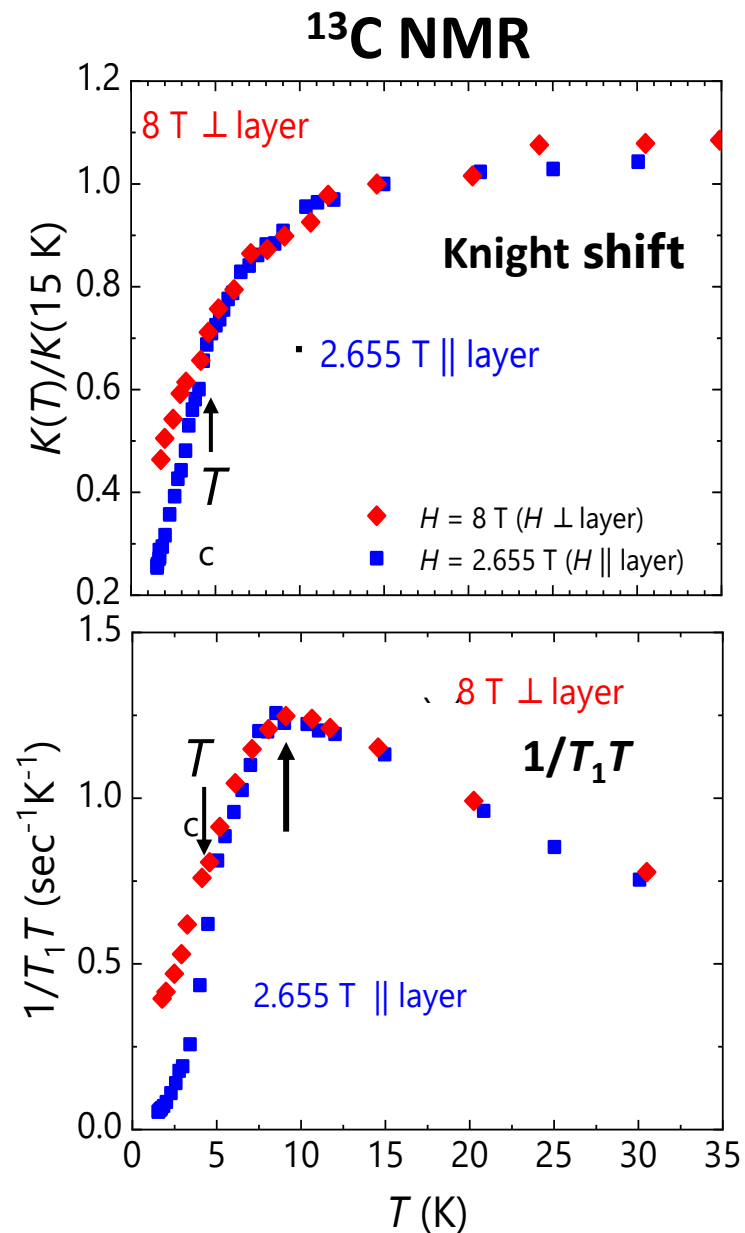
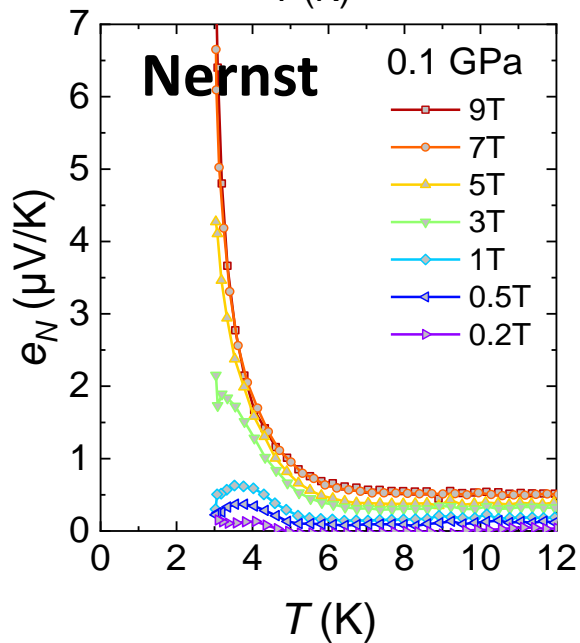
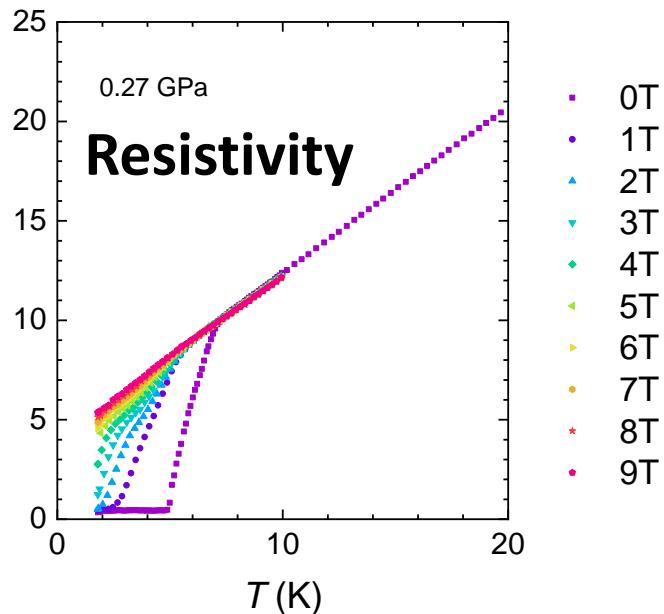
Bose metal in κ -(ET)₄Hg_{2.89}Br₈ at high magnetic fields ?



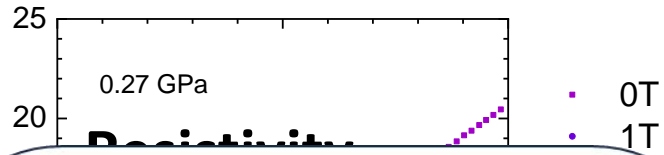
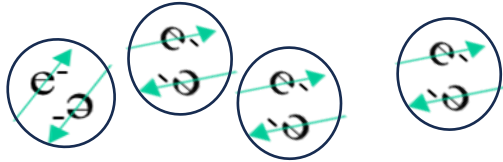
magnetic susceptibility



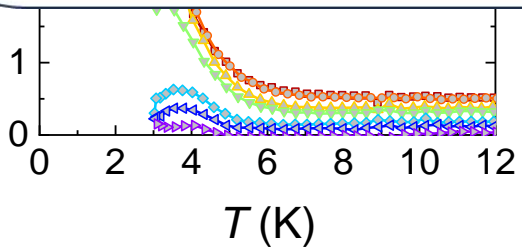
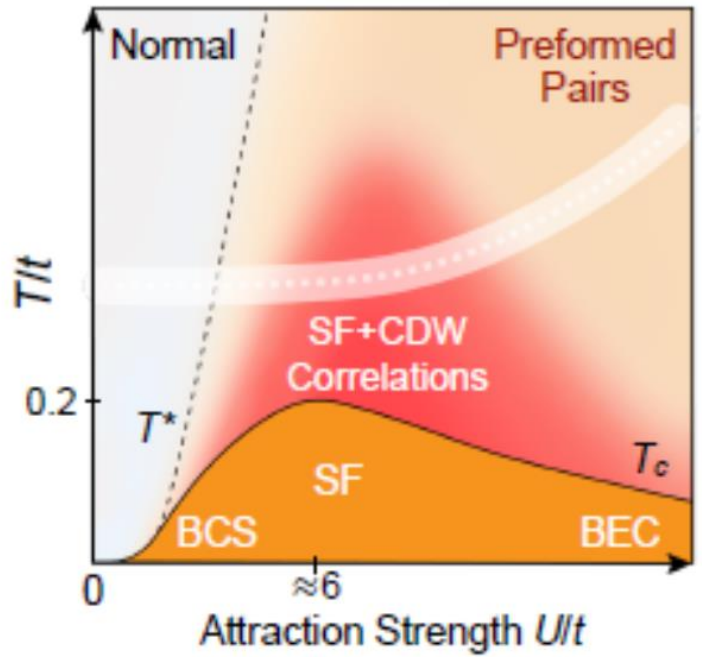
Not due to SC diamagnetism



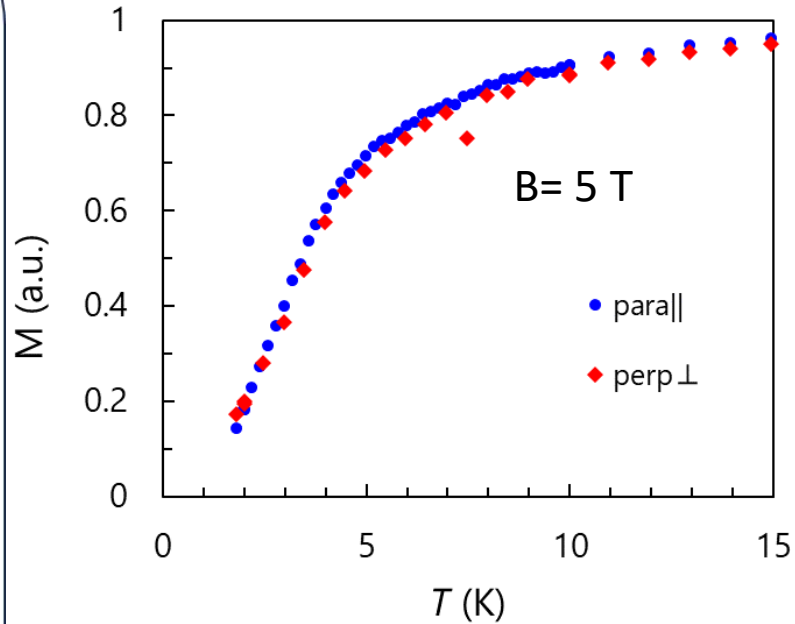
Bose metal in $\kappa\text{-(ET)}_4\text{Hg}_{2.89}\text{Br}_8$ at high magnetic fields ?



T. Hartke et al., arXiv: 2208.05948



magnetic susceptibility

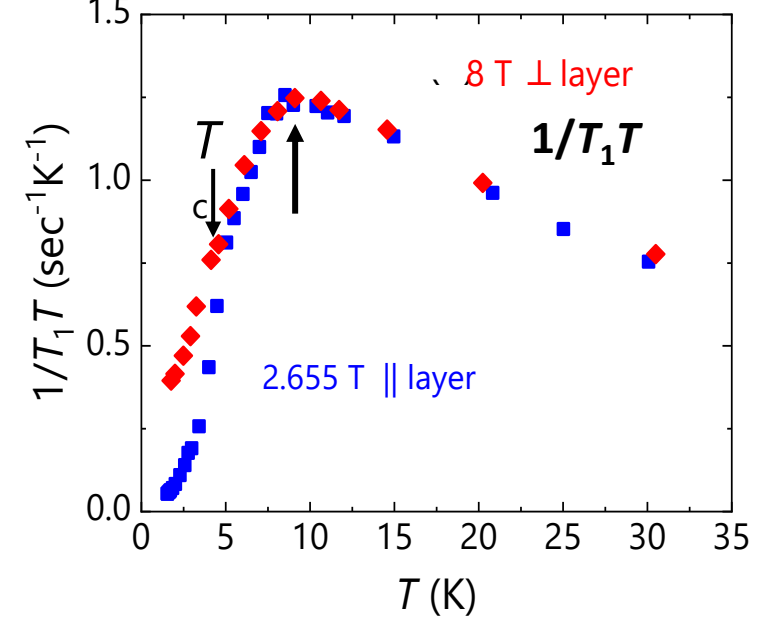
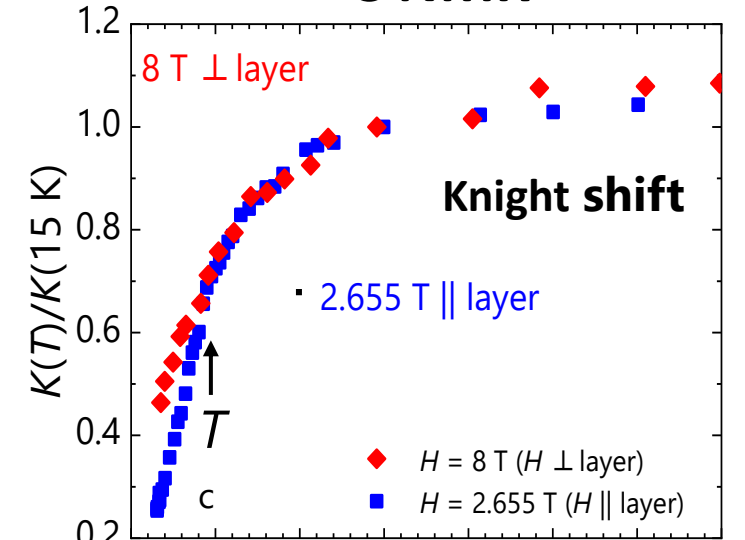


χ vanishes on cooling
without anisotropy.



Not due to SC diamagnetism

^{13}C NMR



Superfluid density

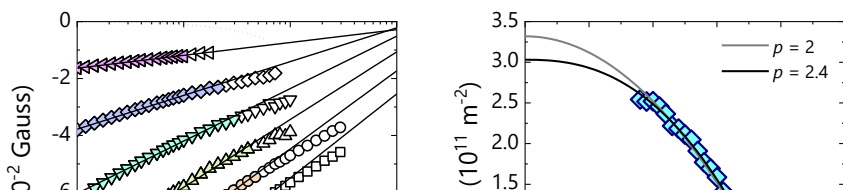


Wakamatsu et al., arXiv. 2205.03682

Uemura plot

magnetization \longrightarrow penetration depth

$$-4\pi M = \frac{\varphi_0}{8\pi\lambda^2} \ln\left(\frac{\beta H c_2}{H}\right) \quad (\text{Lang et al., PRB 46, 5822 (1992)})$$

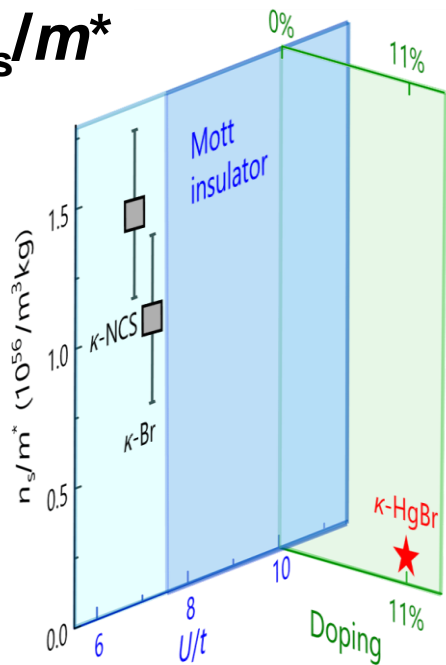


Reduced superfluid density

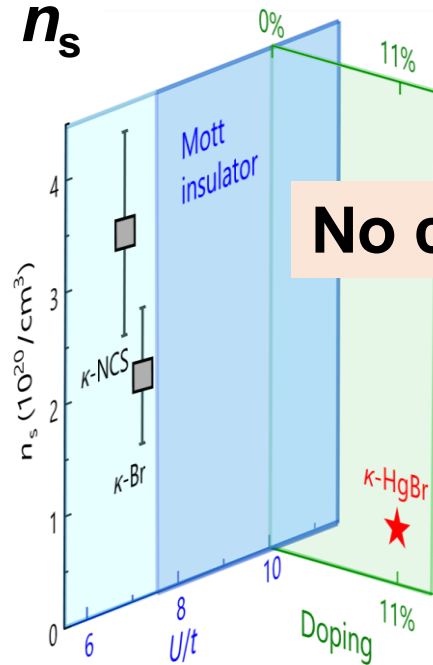
$n_s \sim 15\text{-}25\%$ of total carriers

$$\frac{n_s}{m^*} = \frac{c^2}{4\pi e^2} \frac{1}{\lambda_L^2}$$

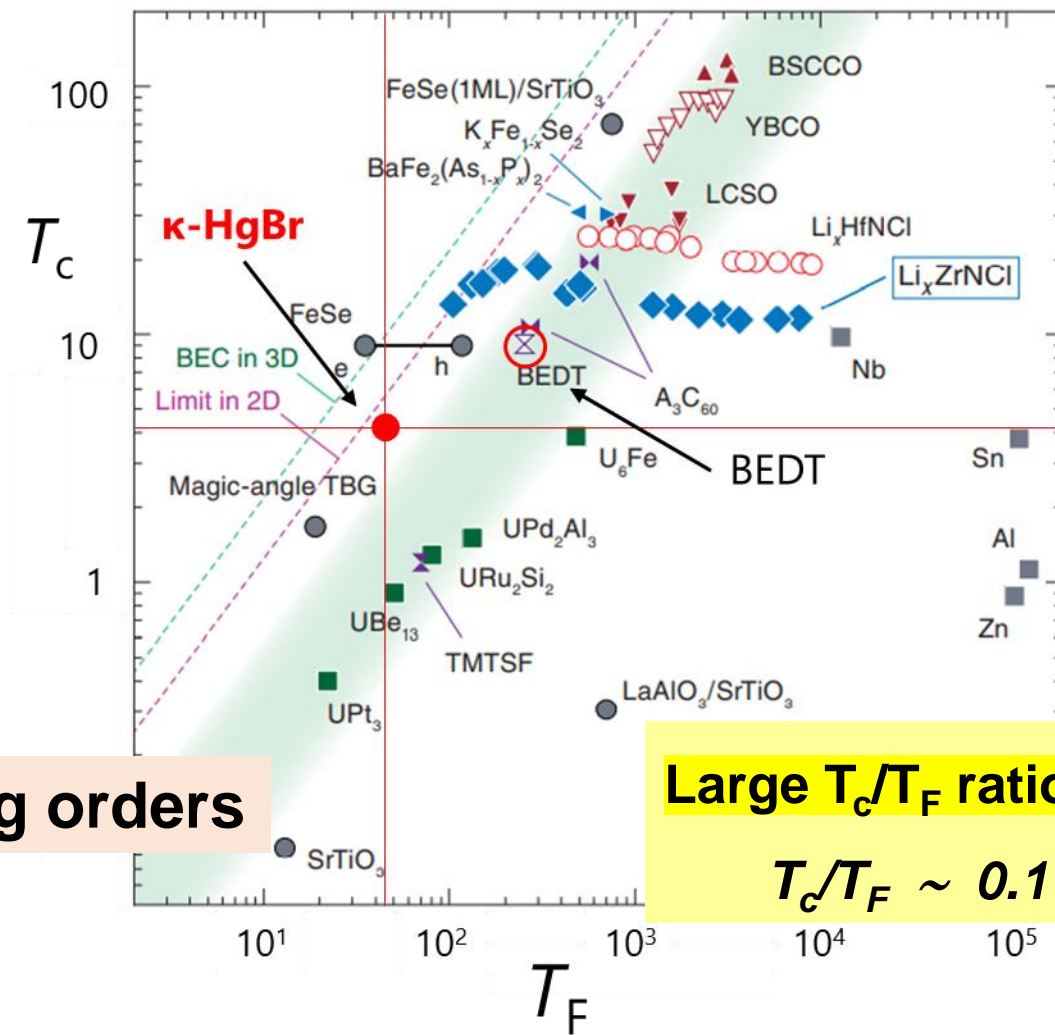
n_s/m^*



n_s



No competing orders



Large T_c/T_F ratio

$$T_c/T_F \sim 0.1$$

Y. J. Uemura, *et al.*, Phys. Rev. Lett. **66** 2665 (1991).

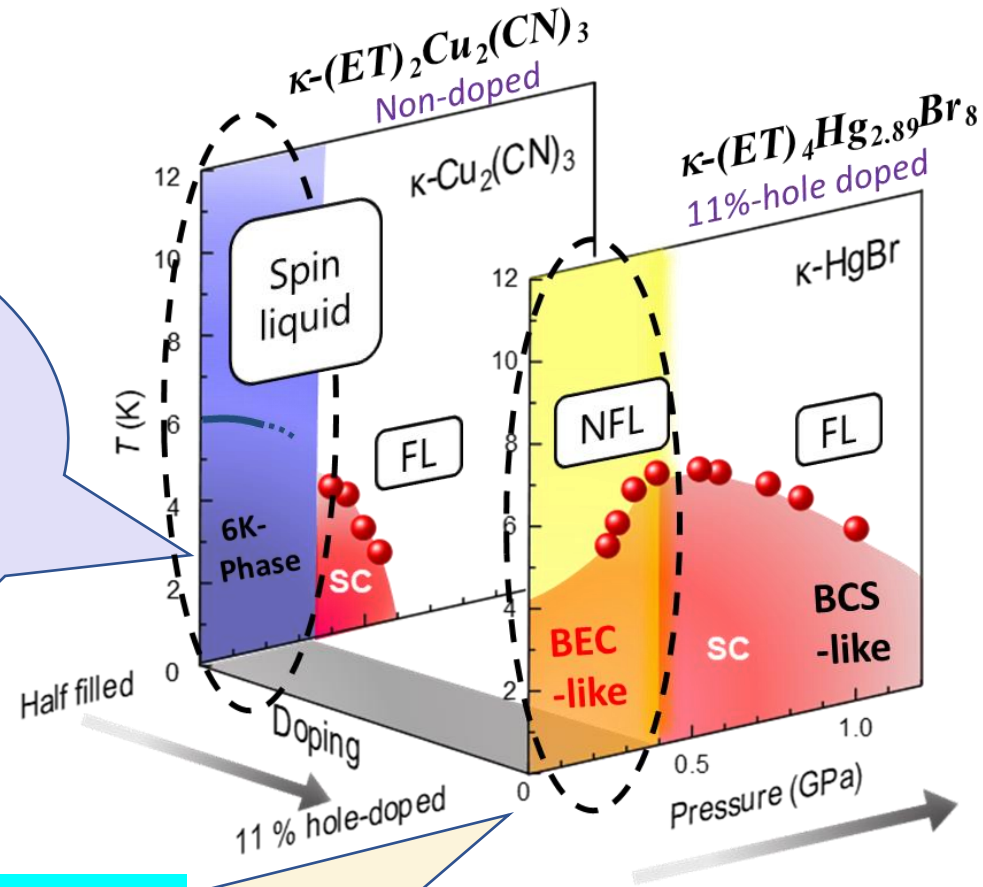
Y. Nakagawa, *et al.*, Science **372**, 190 (2021).

Summary

QSL $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

No magnetic order,
Spin excitations of ~ 10 K
Symmetry breaking not observed yet
Quantum oscillations not observed yet
6K anomaly
Spin-active state below 5K
Bosonic spinon vs fermionic spinon

Low-T state still elusive



Doped QSL $\kappa\text{-(ET)}_4\text{Hg}_{2.89}\text{Hg}_8$

Indications of NFL-FL crossover
NFL phase = quantum critical "phase"
Mottness-controlled BEC-BCS crossover
reduced superfluid density
preformed pairs

- H. Oike *et al.*, *PRL*114, 067002 (2015).
H. Oike, *et al.*, *Nat. Commun.* **8**, 756 (2017).
Y. Suzuki *et al.*, *PRX* **12**, 011016 (2022).
K. Wakamatsu *et al.*, *Nat. Commun.* in press
(arXiv:2201.10714)
H. Oike *et al.*, arXiv:2202.06032
K. Wakamatsu *et al.*, arXiv:2205.03682

Collaborators

**K. Miyagawa, Y. Shimizu, Y. Kurosaki, T. Furukawa, H. Oike,
Y. Suzuki, K. Wakamatsu, Y. Ueno, T. Fujii, J. Ibuka (U Tokyo)
G, Saito, M. Maesato (Kyoto U.)
T. Sasaki (Tohoku U., IMR)
H. Taniguchi, M. Itou (Saitama U.)
S. Yamashita, Y. Nakazawa (Osaka)
F. Pratt (Rutherford-Appleton), Blundell (Oxford)
Y. Kohama, A. Miyake, T. Nomura (ISSP)
B. Mikche, M. Dressel (Stuttgart U.)**