“Quantum Matter: Computation Meets Experiments”, Aspen, March 11, 2020

Quantum spin liquid
and Mott quantum criticality

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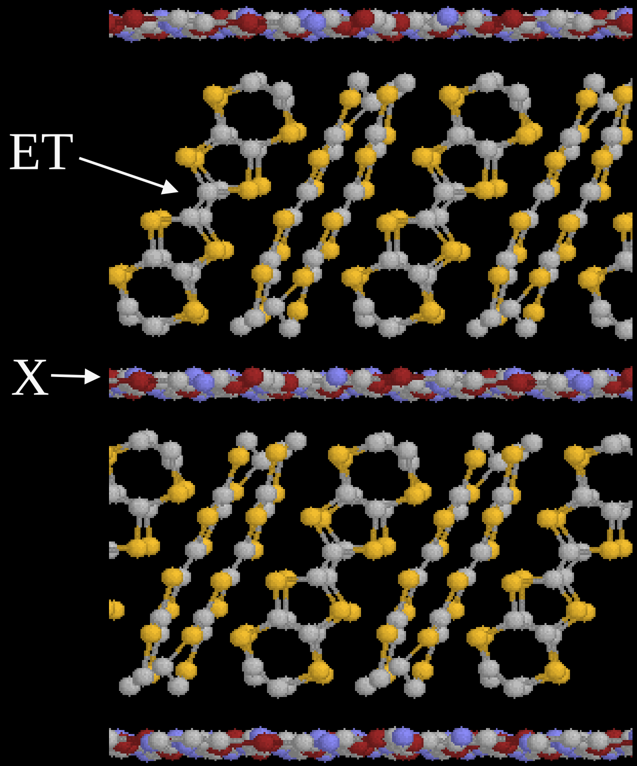
Applied Physics, UTokyo

Outline of talk

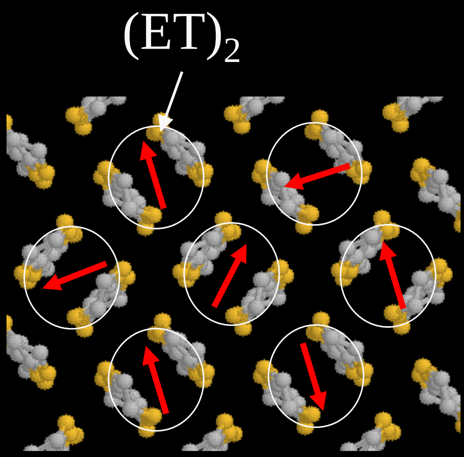
- Spin liquid
    - Spins --- localized or itinerant ?
    - Instability of spin liquid --- singlet formation & FFLO ?
- Doped spin liquid
    - non-Fermi liquid to Fermi liquid crossover
    - BEC to BCS crossover
- Mott quantum criticality
    - Disorder effect

κ-$(ET)_2X$ --- half-filled bands, quasi-triangular lattices

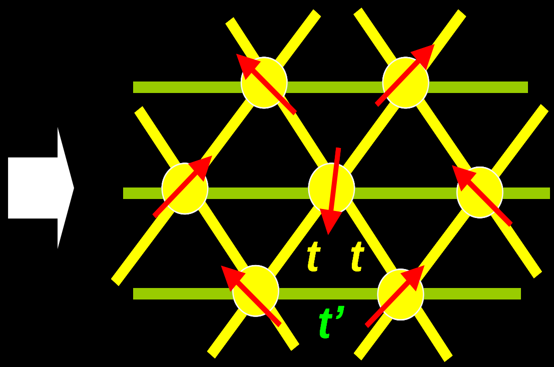
Layered structure



In-plane structure



Isoceles triangular lattice



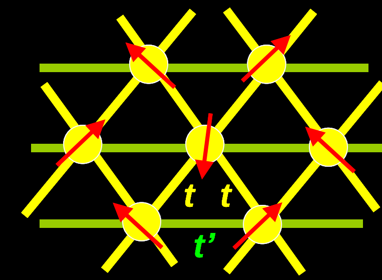
Triangularity, t'/t, is varied by X

X^-	Ground State	t'/t	
$Cu_2(CN)_3$	SL Mott insulator	1.06	triangle
$Cu[N(CN)_2]Cl$	AF Mott insulator	0.75	deformed triangle

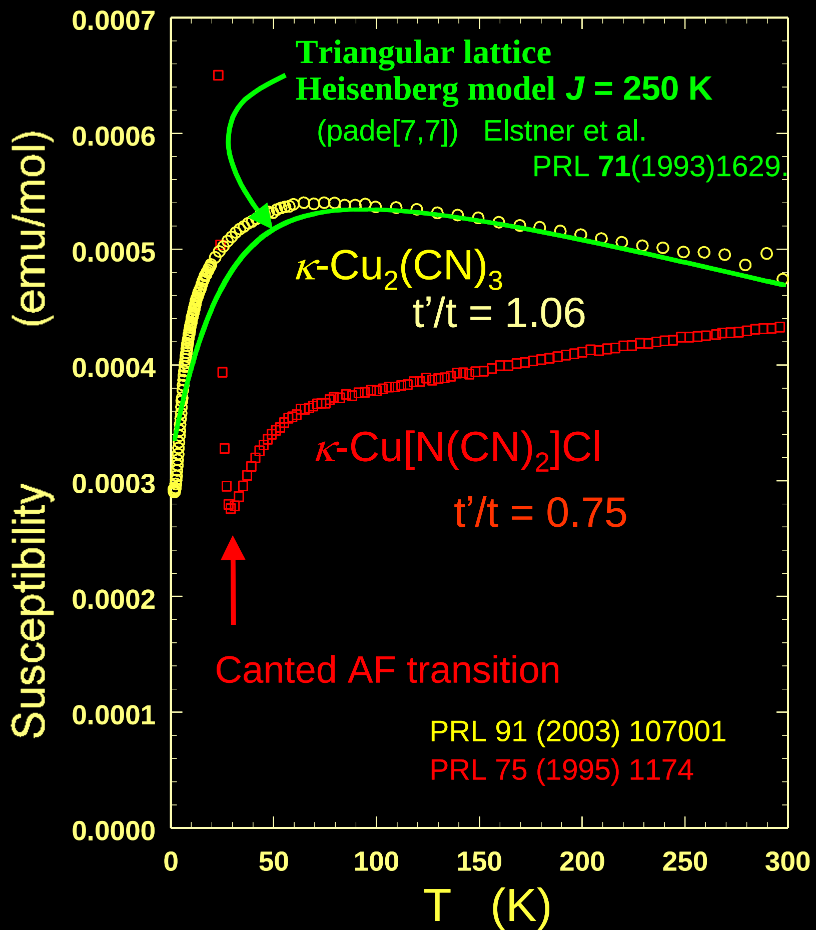
Mott insulators κ-$(ET)_2X$

Spin ordering or not ?

X	t'/t
$Cu_2(CN)_3$	0.80-1.06
$Cu[N(CN)_2]Cl$	0.44-0.75

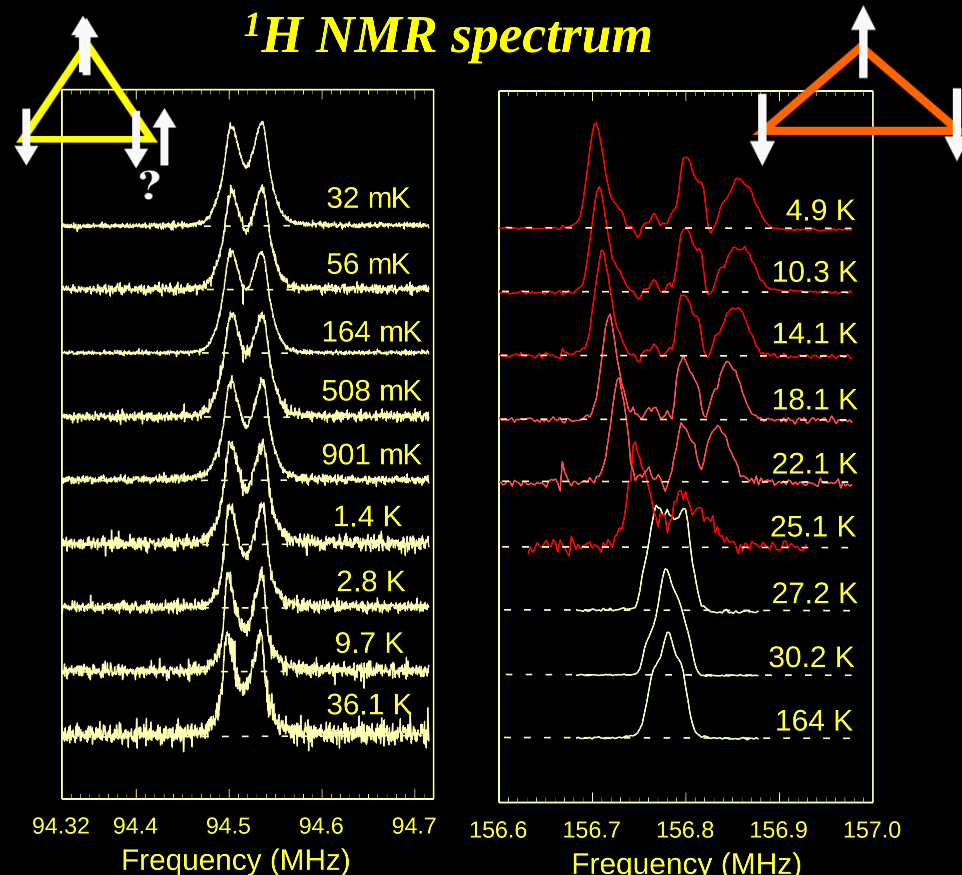


Magnetic susceptibility



Also see Zheng et al. PRB 71 (2005) 134422

^{1}H NMR spectrum



κ-$(ET)_2Cu_2(CN)_3$	κ-$(ET)_2Cu[N(CN)_2]Cl$
t'/t =1.06	t'/t =0.75
No ordering ⟺	AF ordered (0.45μ_B)

Knight shift (*K*) vs Magnetization (*M*)

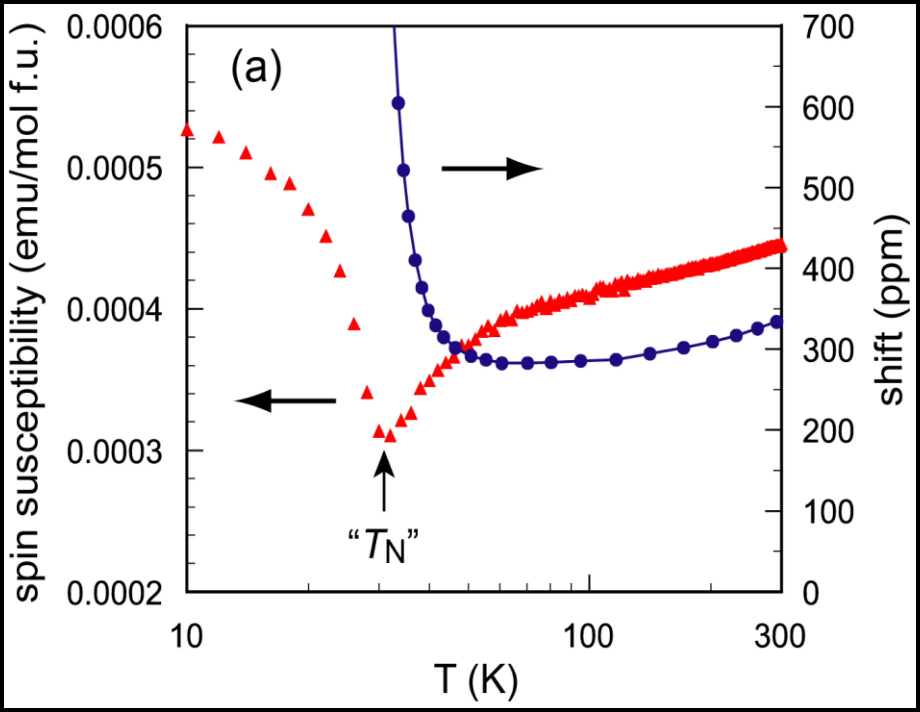
H_0 // z

$$\langle K\rangle = A_{zx}\cdot\langle M_x\rangle + A_{zy}\cdot\langle M_y\rangle + A_{zz}\cdot\langle M_z\rangle$$

Usually vanishing but nonzero in presence of D-M interaction

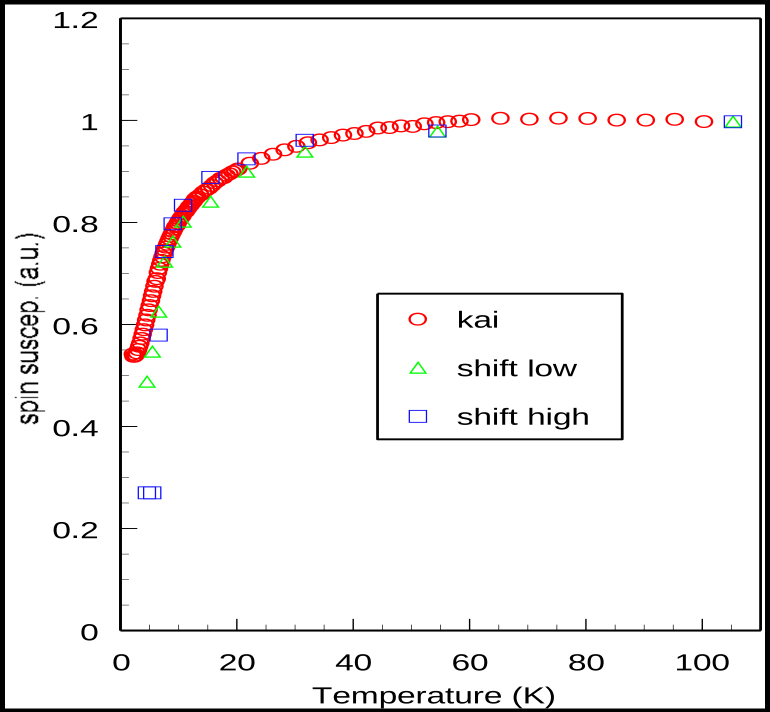
No *K*-χ proportionality

κ-$(ET)_2Cu[N(CN)_2]Cl$ (t'/t=0.75)



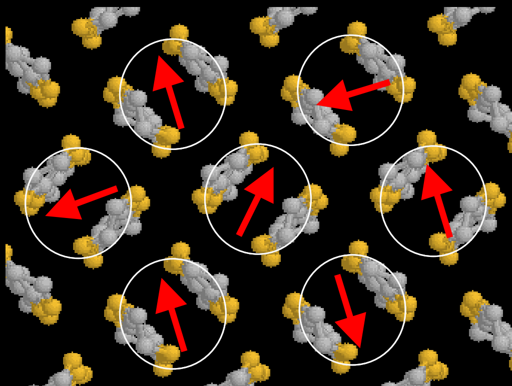
Good K-χ proportionality !?

κ-$(ET)_2Cu_2(CN)_3$ (t'/t=1.06)



Dzyaloshinsky-Moriya interaction causes staggered magnetization in a **localized spin system** under applied field

$$H = J\sum_{<i,j>}\hat{S}_i\cdot\hat{S}_j + g\mu_B\sum_i \hat{S}_i\cdot H_i + \sum_{<i,j>} D_{ij}\cdot(\hat{S}_i\times\hat{S}_j)$$

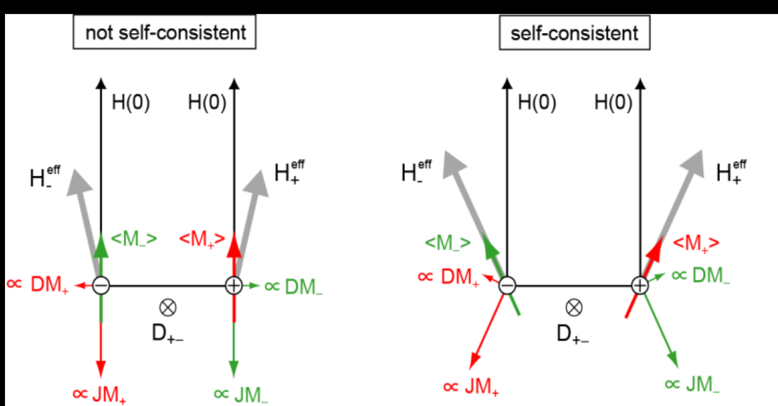


Molecular field at sublattice sites

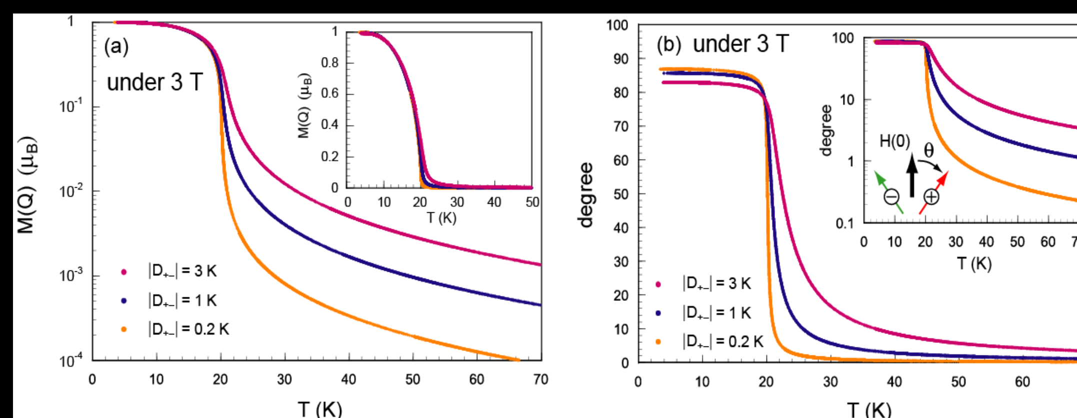
$$H_+^{eff} = -\frac{ZJ}{(g\mu_B)^2}<M_->-\frac{Z}{(g\mu_B)^2}<M_->\times D_{+-}+H_i$$

$$H_-^{eff} = -\frac{ZJ}{(g\mu_B)^2}<M_+>-\frac{Z}{(g\mu_B)^2}D_{+-}\times<M_+>+H_i$$

Spin configuration



Molecular field calculation of staggered magnetization

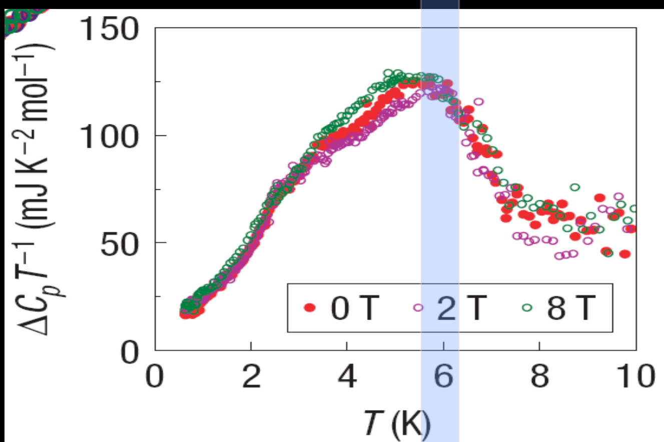


Kagawa et al. PRB 78 (2008) 184402

Thermodynamic anomaly at 6K in κ-$(ET)_2Cu_2(CN)_3$

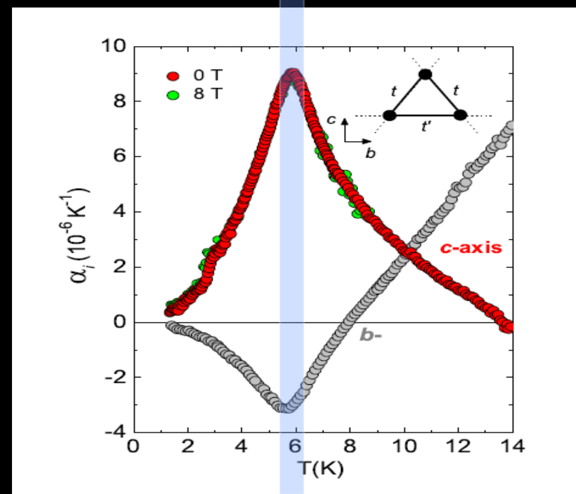
Specific heat

S. Yamashita *et al*., *Nat. Phys*. 4 (2008) 459



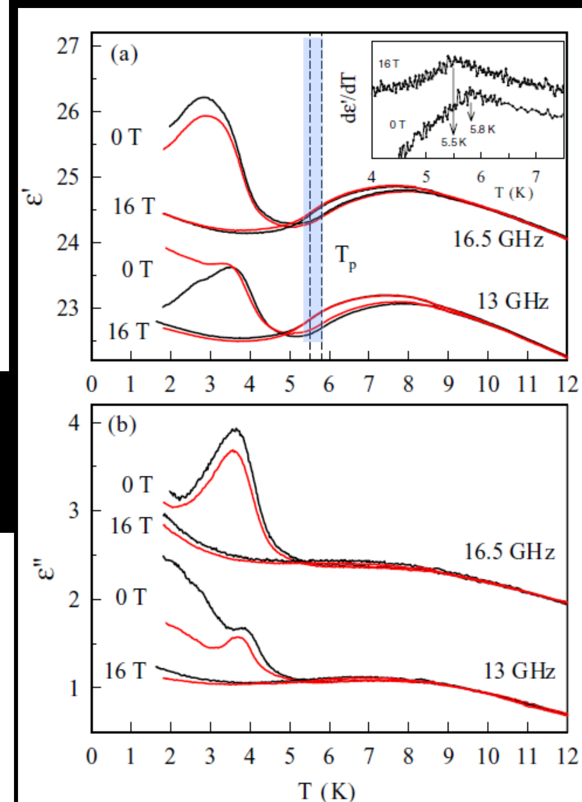
Thermal expansion coefficient

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



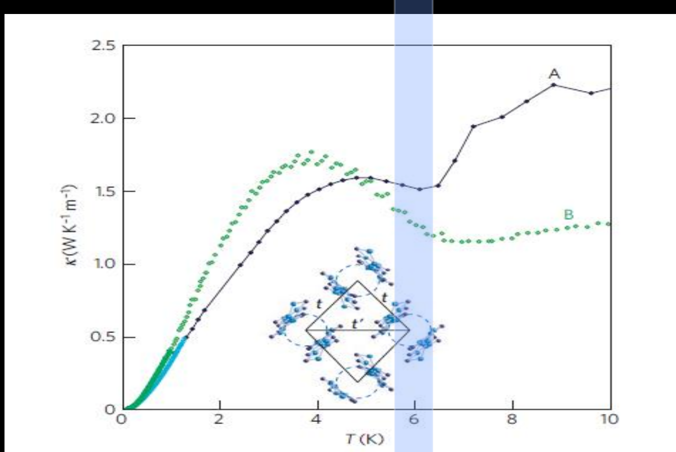
Dielectric function

Poirier et al., *PRB* 85, 134444 (2012)



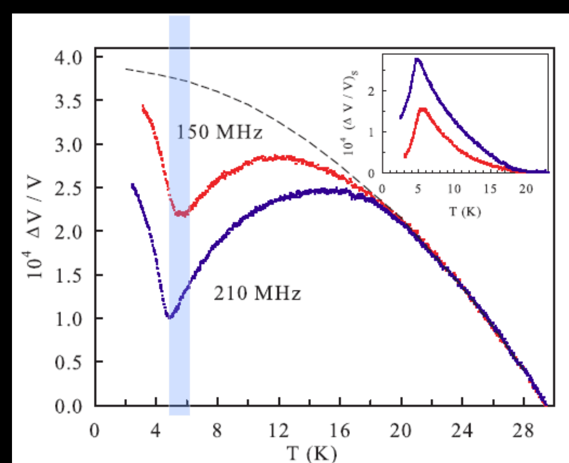
Thermal conductivity

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

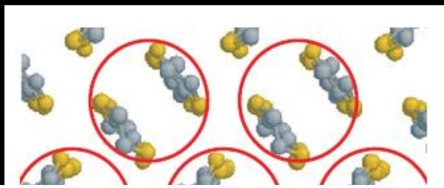


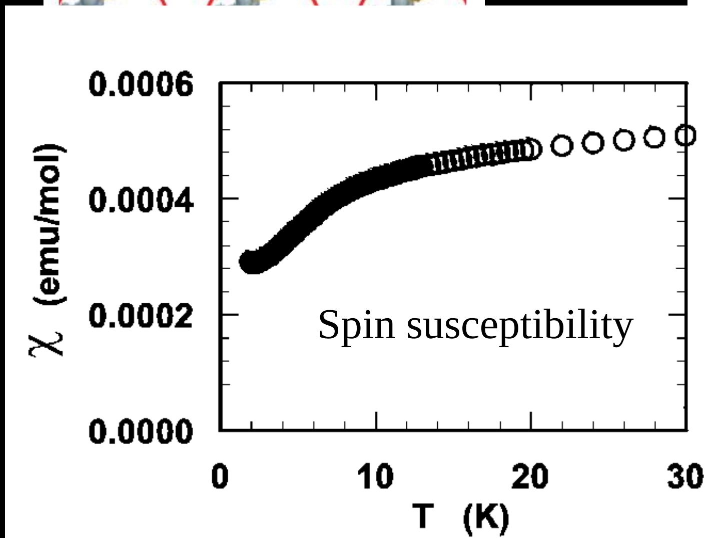
Ultrasound velocity

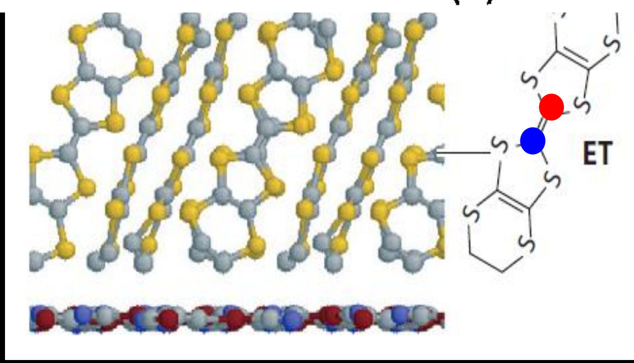
Poirier et al., PRB (2012)

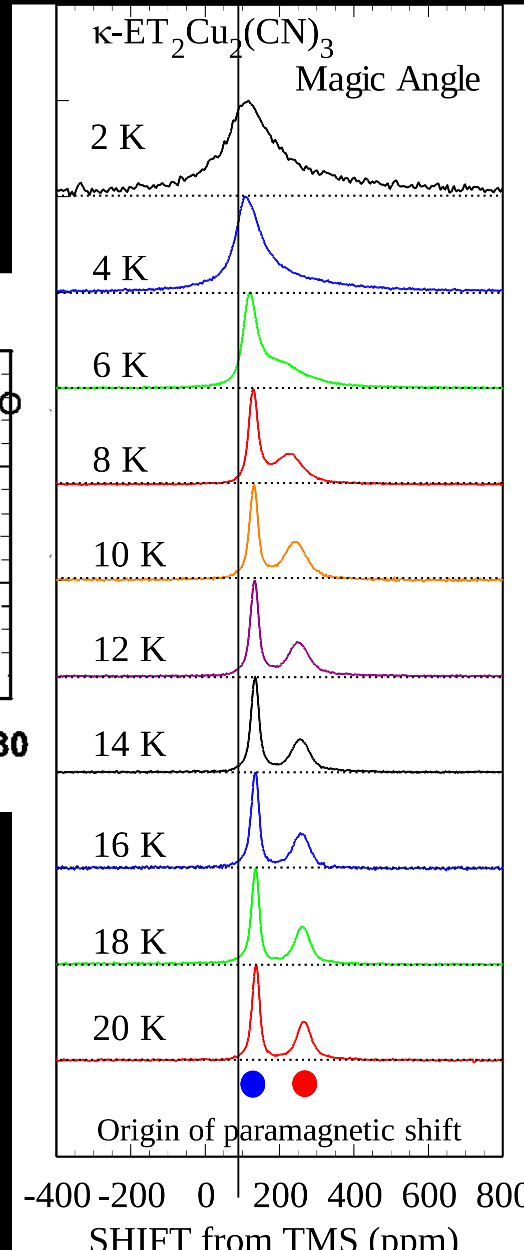


^{13}C NMR under a parallel field

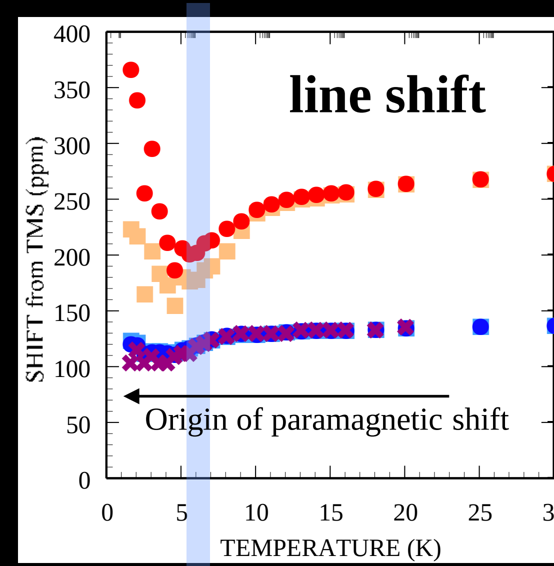






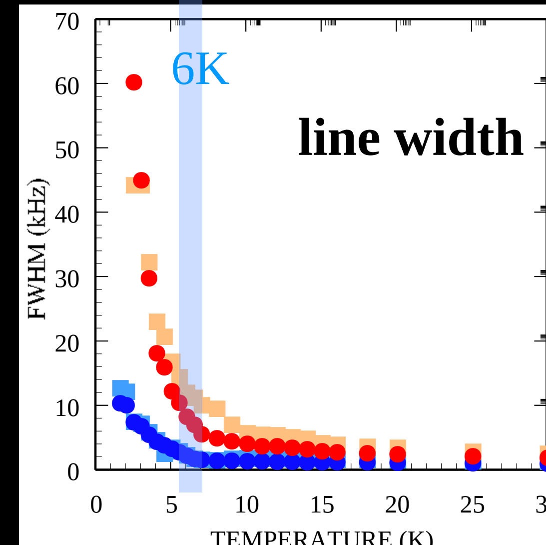


a decrease in local χ

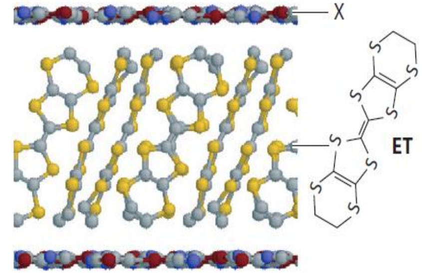


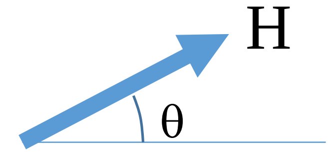
line broadening

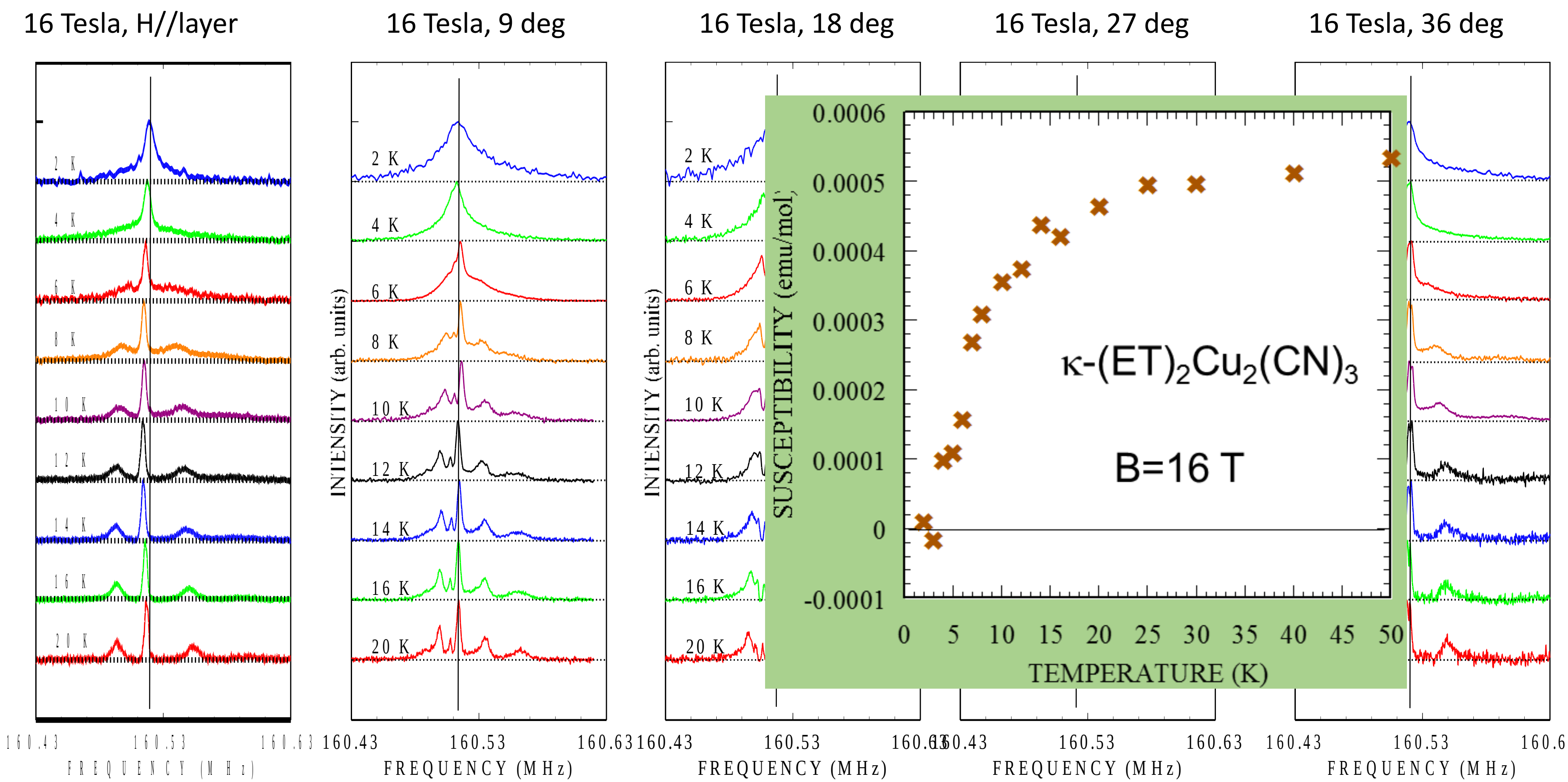
Inhomogeneous staggered moment



^{13}C NMR@16 Tesla







The anomalous NMR behavior below 6 K persists even under 16 T.

Puzzling experimental indications

Absence of manifestation of DM interactions

→ Itinerant spins (spinon FS ?)

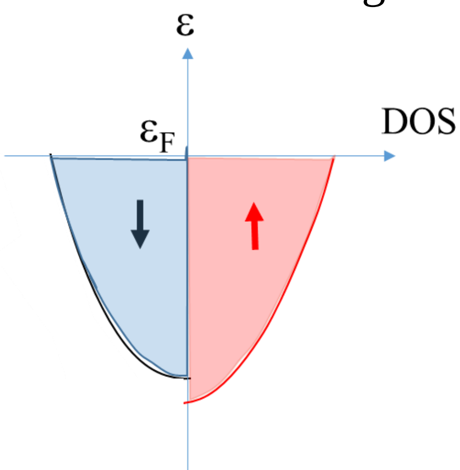
6K-anomaly --- Instability of spin liquid ?

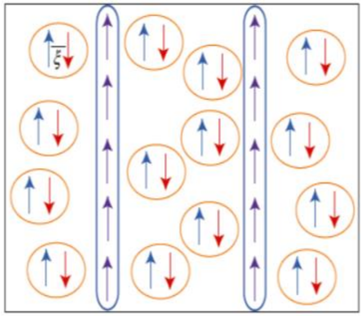
→ Transition to nonmagnetic state

→ Robust to applied field

→ Inhomogeneous local fields

Unpaired spins generate inhomogeneous local fields.





FFLO of singlet spinon pairing ???

κ-$(ET)_4Hg_{2.89}Br_8$ a doped triangular lattice (11% hole doping)

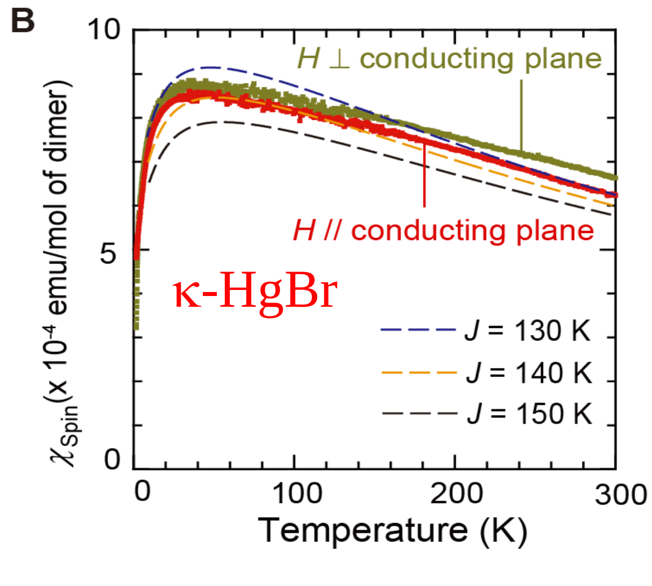
Lyubovskaya, 1987

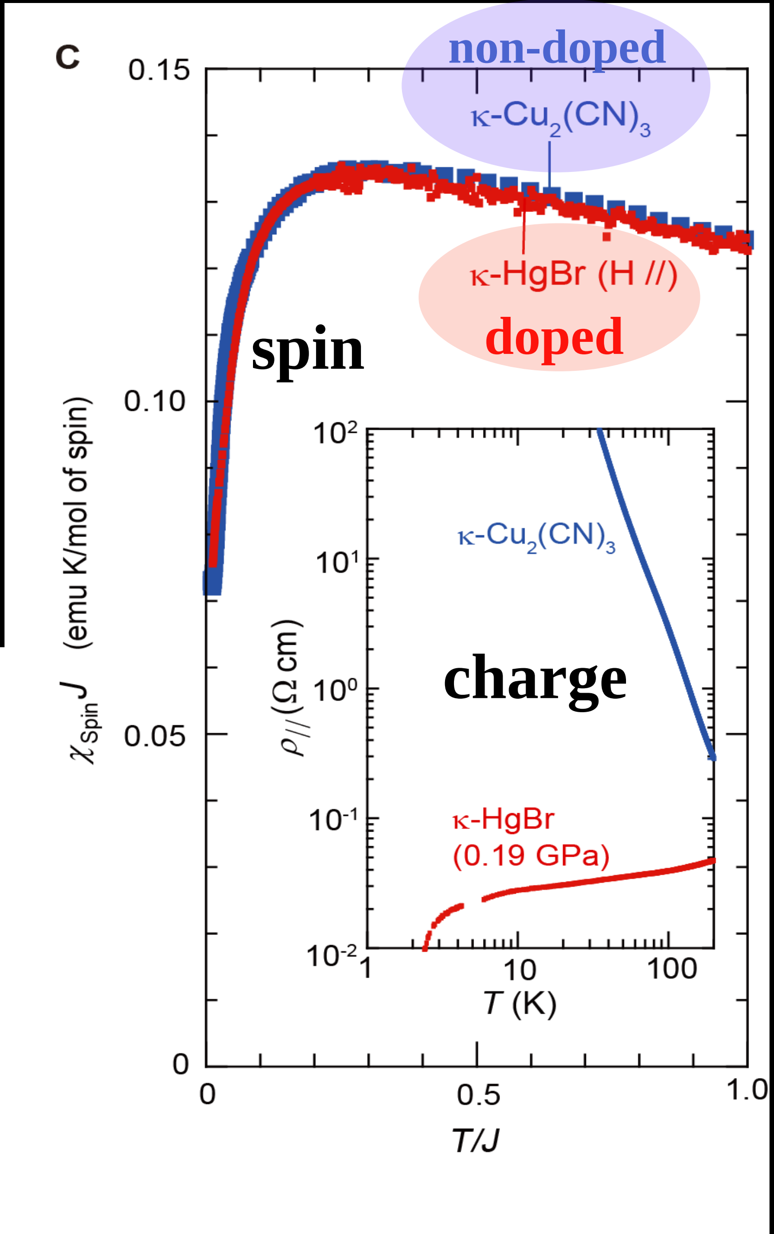
Spin liquid nature !

Spin-charge separation !

Oike et al., *Nature Commun. 8, 756 (2017)*

Triangular-lattice Heisenbeg model
J=140 K



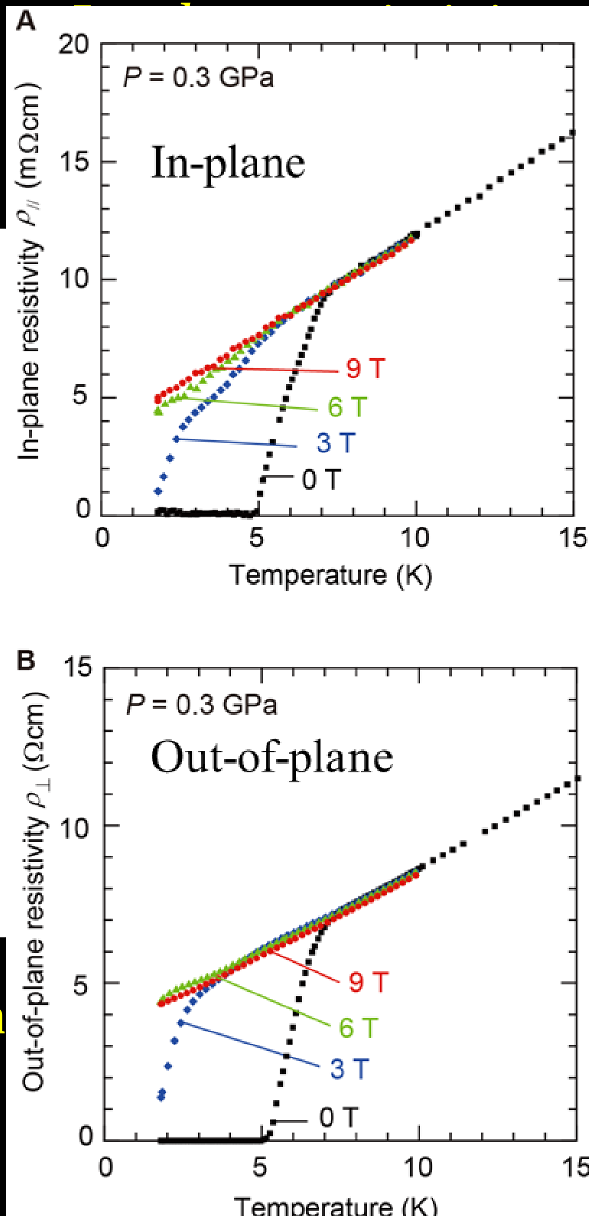


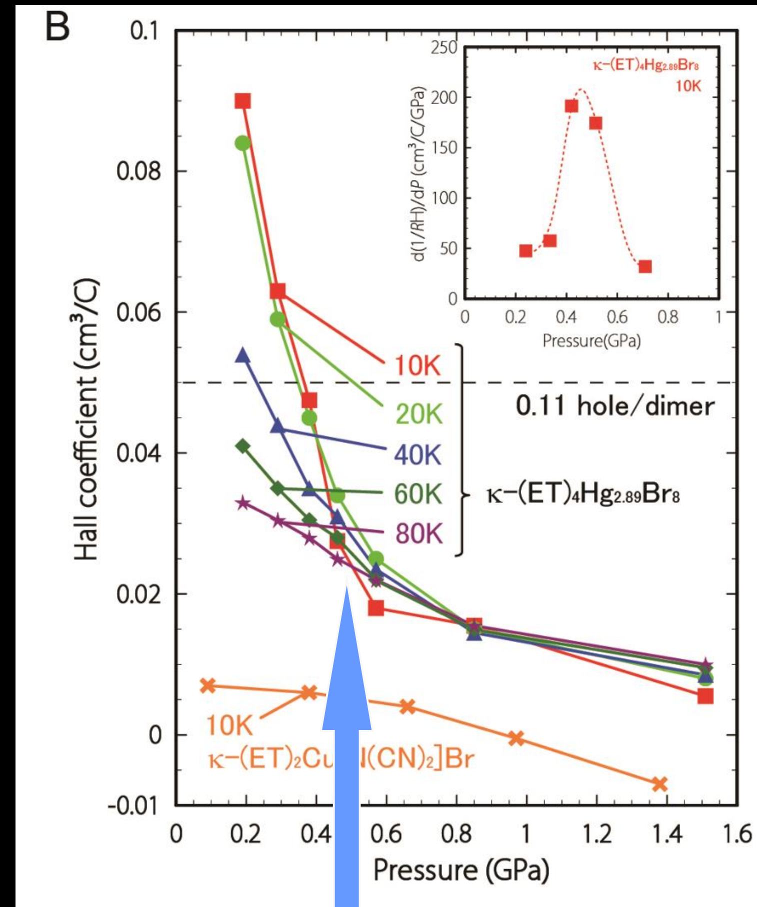
κ-$(ET)_4Hg_{2.89}Br_8$

Hall coefficient ~ **(carrier number)**$^{-1}$

Oike et al., *PRL*114 (2015) 067002

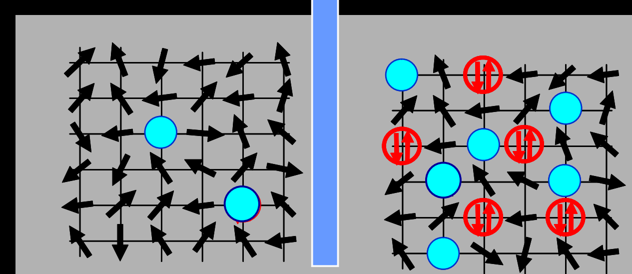
2017)





non

low carrier density



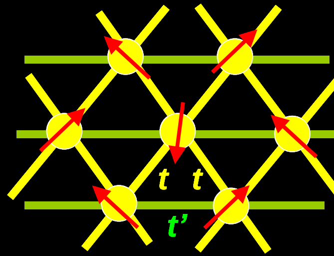
high carrier density

or sharp crossover

Doped Mott

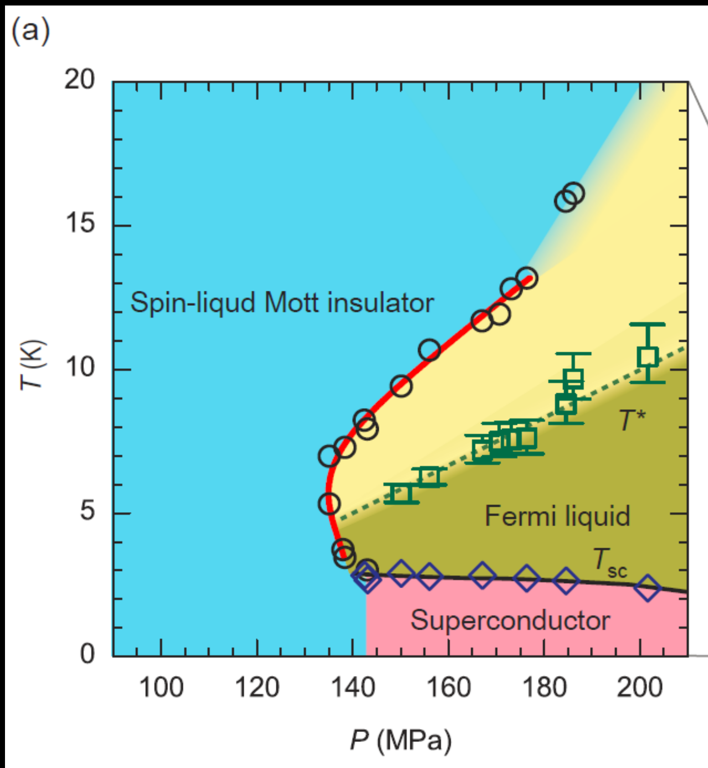
Correlated metal

Doping an triangular-lattice Mott insulator



Non-doped

κ-$(ET)_2Cu_2(CN)_3$ t'/t=0.80-1.0

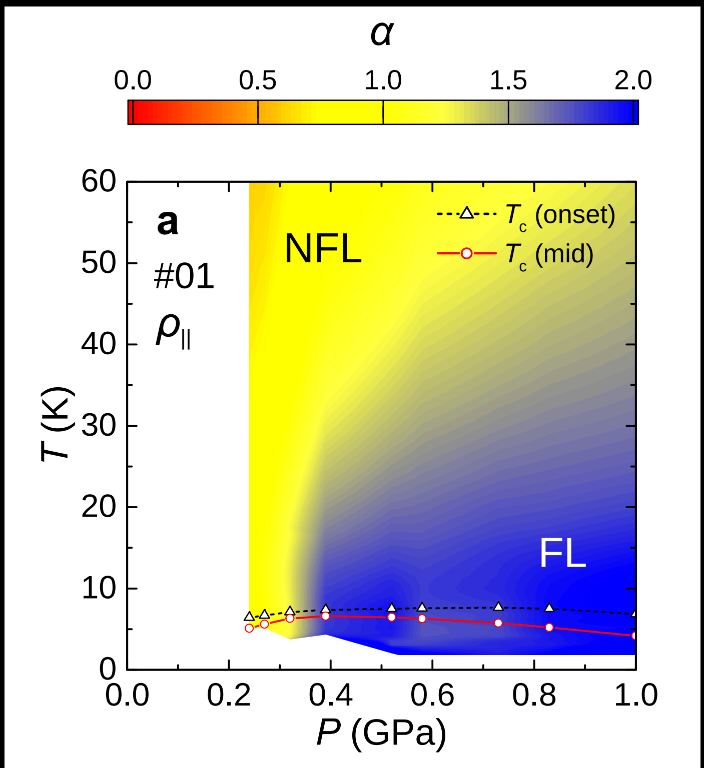


Mott insulator ⇔ metal (FL)

Furukawa etal. Nat. Phys 11 (2015) 221

11%-hole doped

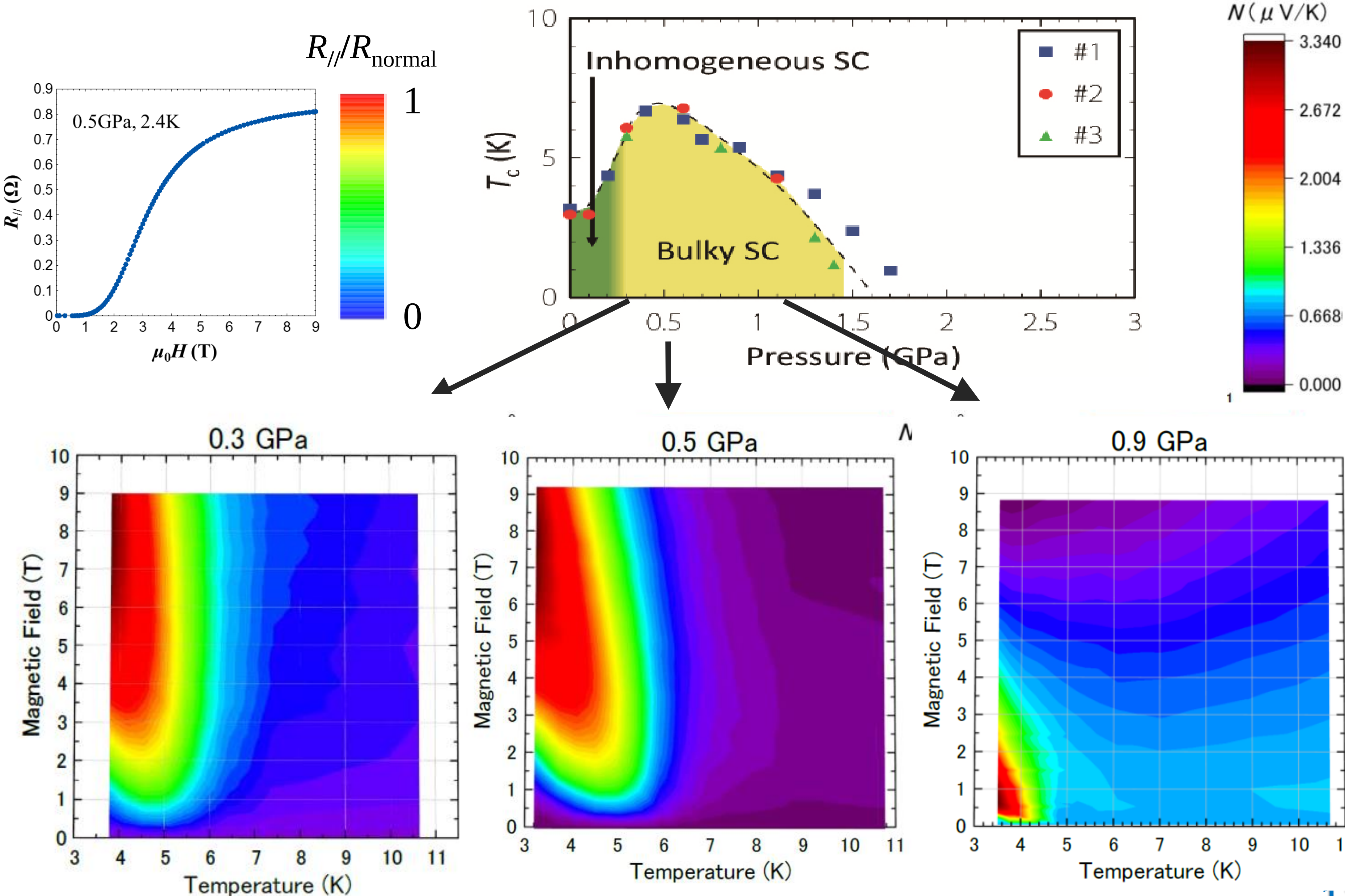
κ-$(ET)_4Hg_{2.89}Hg_8$ t'/t ~ 1.0



Metal (non-FL) ⇔ metal (FL)

*Oike et al., PRL.*114.(2015) 067002

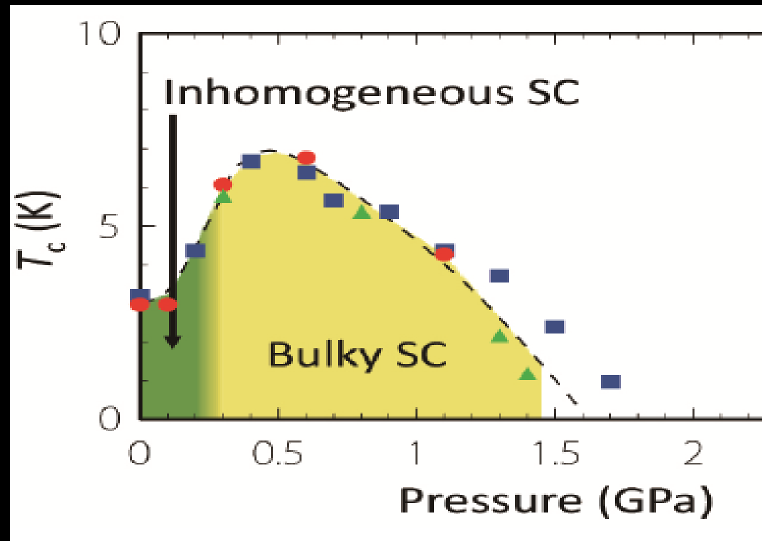
Upper critical field H_{C2}



BEC to BCS crossover in κ-(ET)$_4$Hg$_{2.89}$Br$_8$

Low carrier density

Non-Fermi liquid

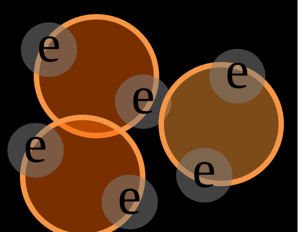


high carrier density

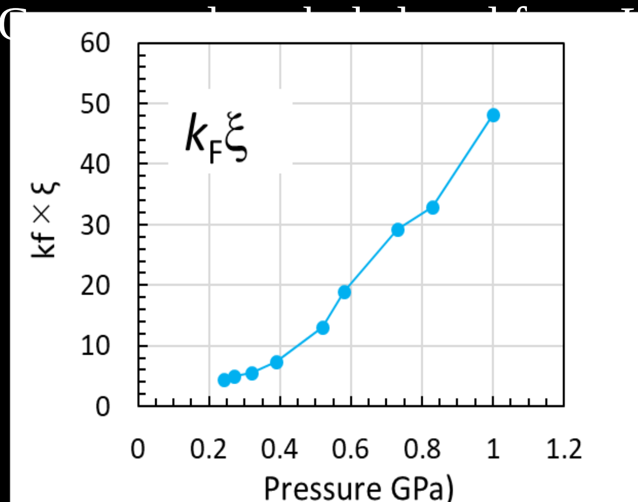
Fermi liquid

BEC like

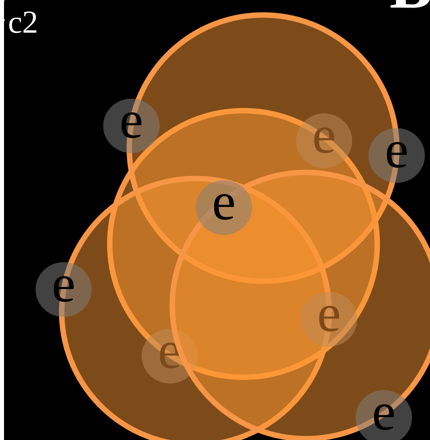
d **~** ***ξ***



$k_F\ \xi$ ~ 2-3



BCS like

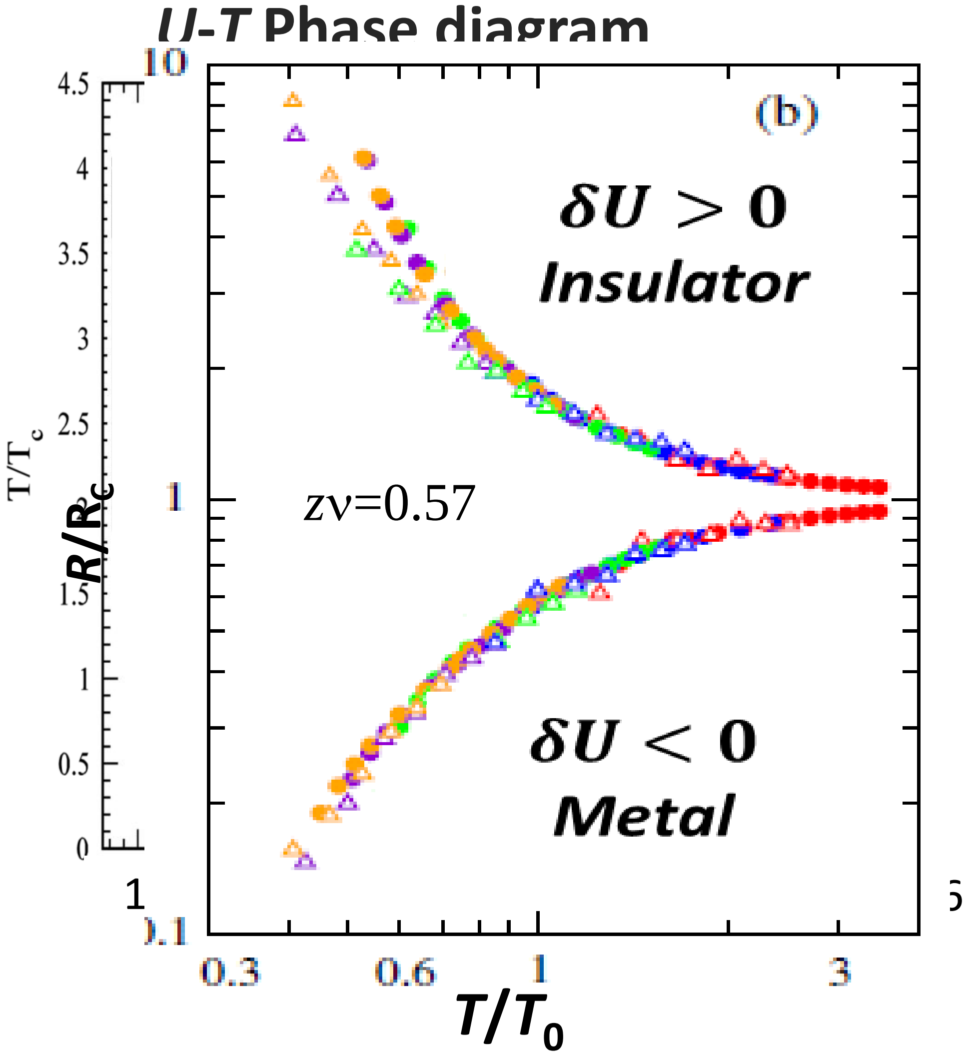


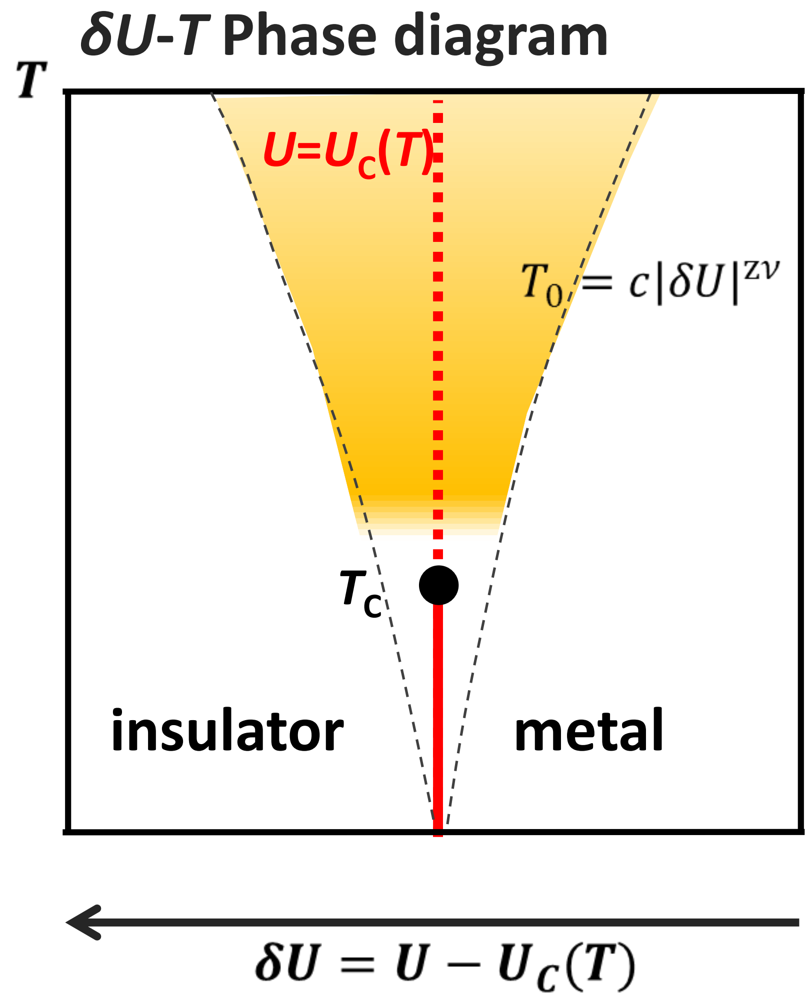
d **<<** ***ξ***

$k_F\ \xi$ ~30-50

Single-site DMFT of Hubbard model

H.Terletska , V.Dobrosavljevic *et al.,Phys. Rev. Lett* **107**, 026401(2011)

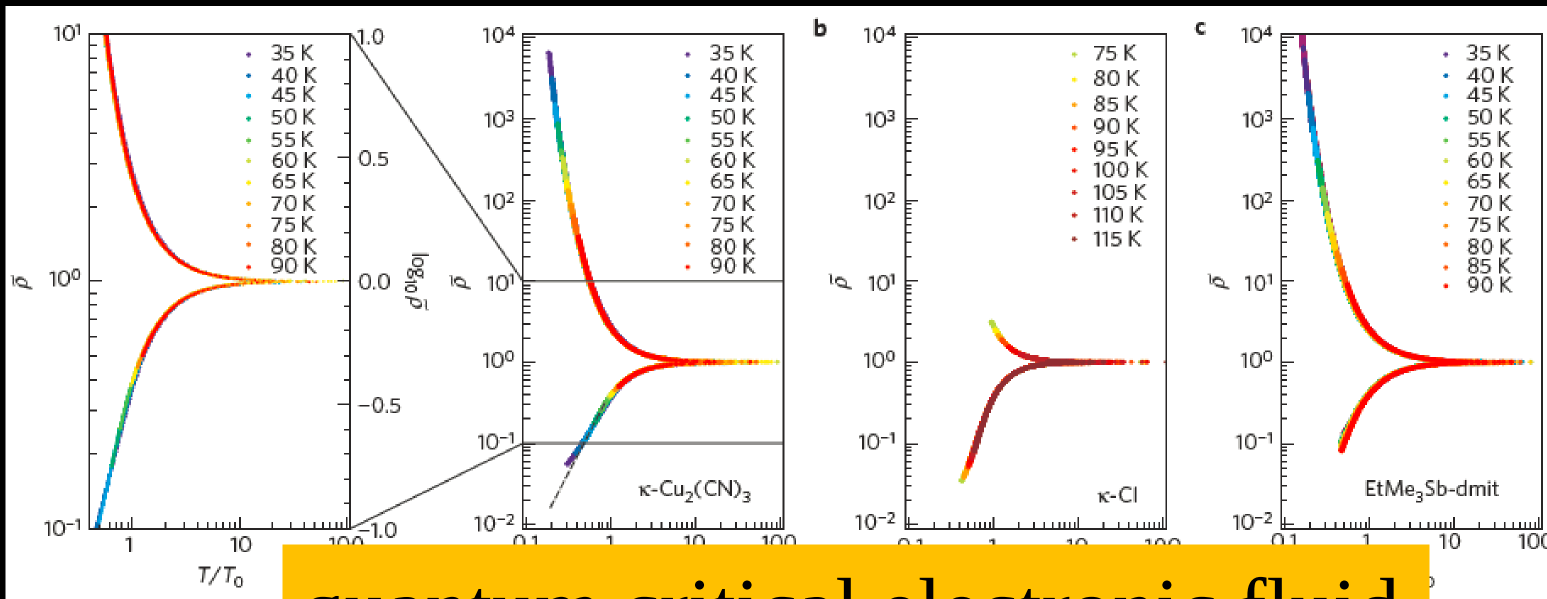




QC scaling --- independent of competing ground states

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

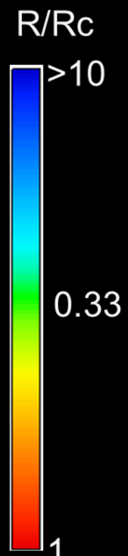
Theoretical: H.Terletska *et al.*,*PRL* **107** (2011) 026401

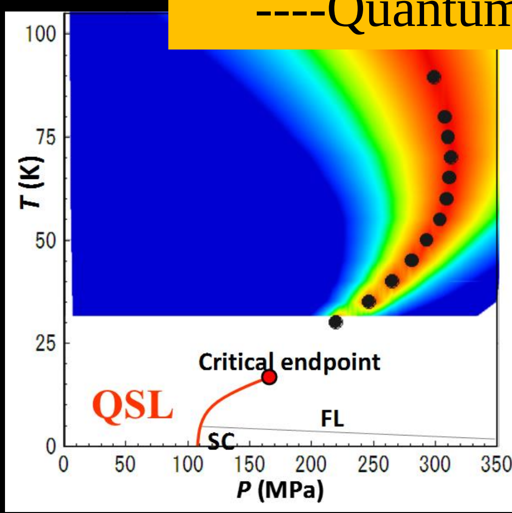


quantum critical electronic fluid

----Quantum analog of supercritical water----

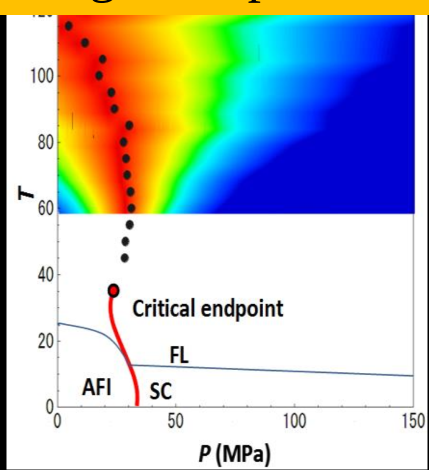
8±0.04





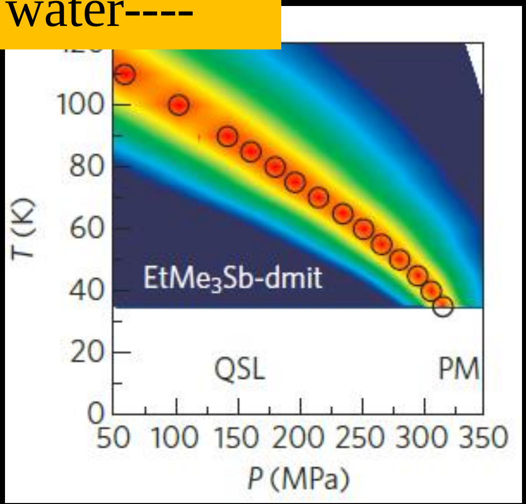
SL/SC

κ-$(ET)_2Cu_2(CN)_3$



AFI/SC

κ-$(ET)_2Cu[N(CN)_2]Cl$

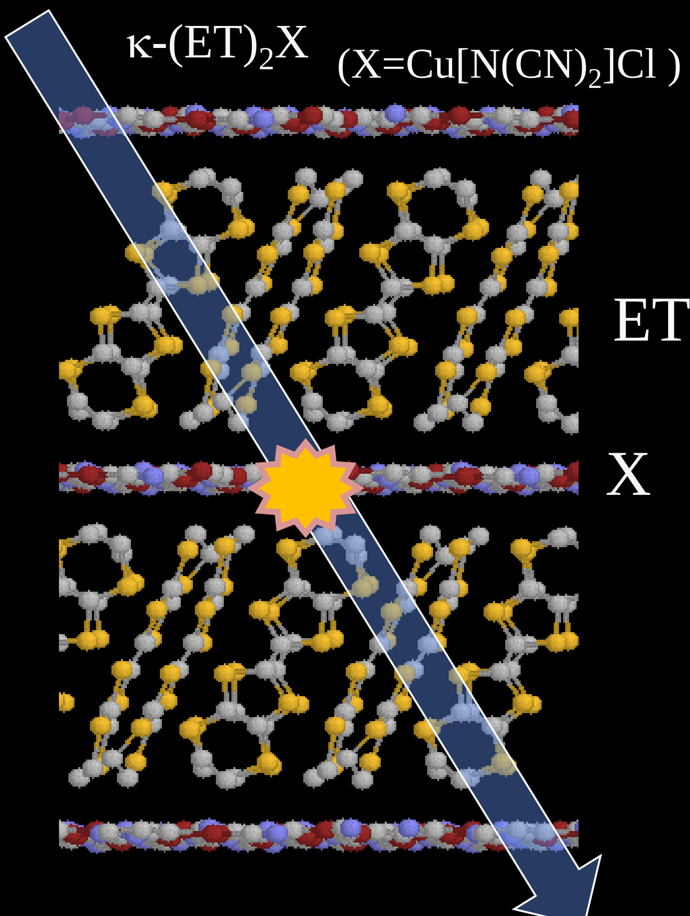


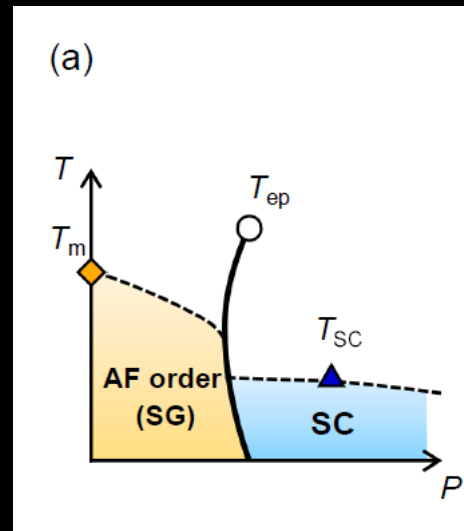
SL/Metal

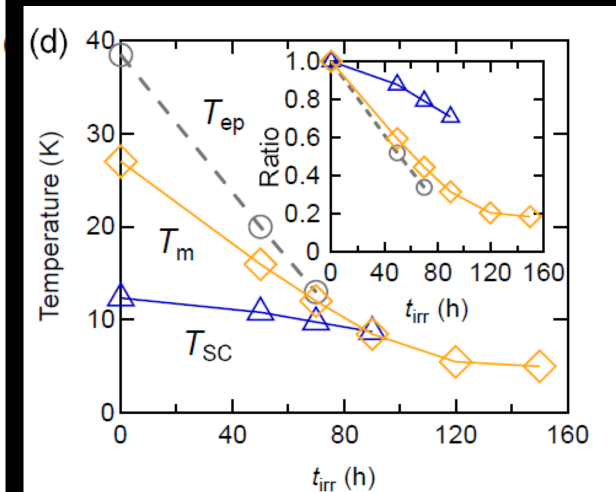
$EtMe_3Sb[Pd(dmit)_2]_2$

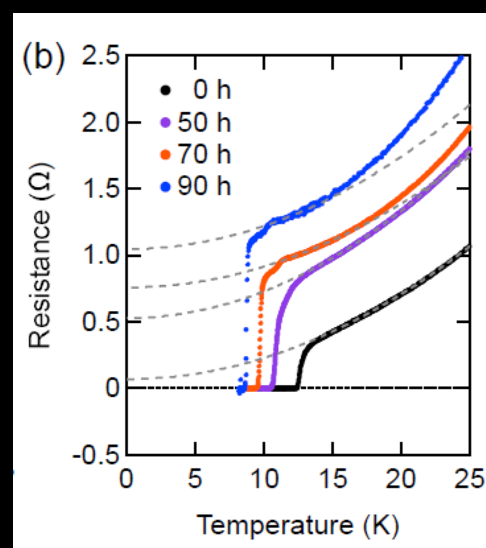
Disorder induced by X-ray irradiation

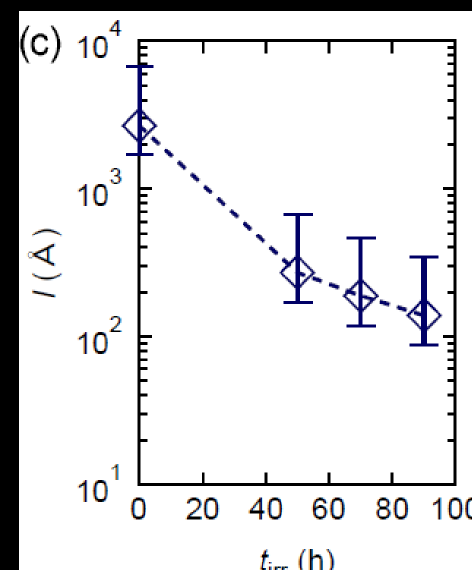
damaged in insulating layers







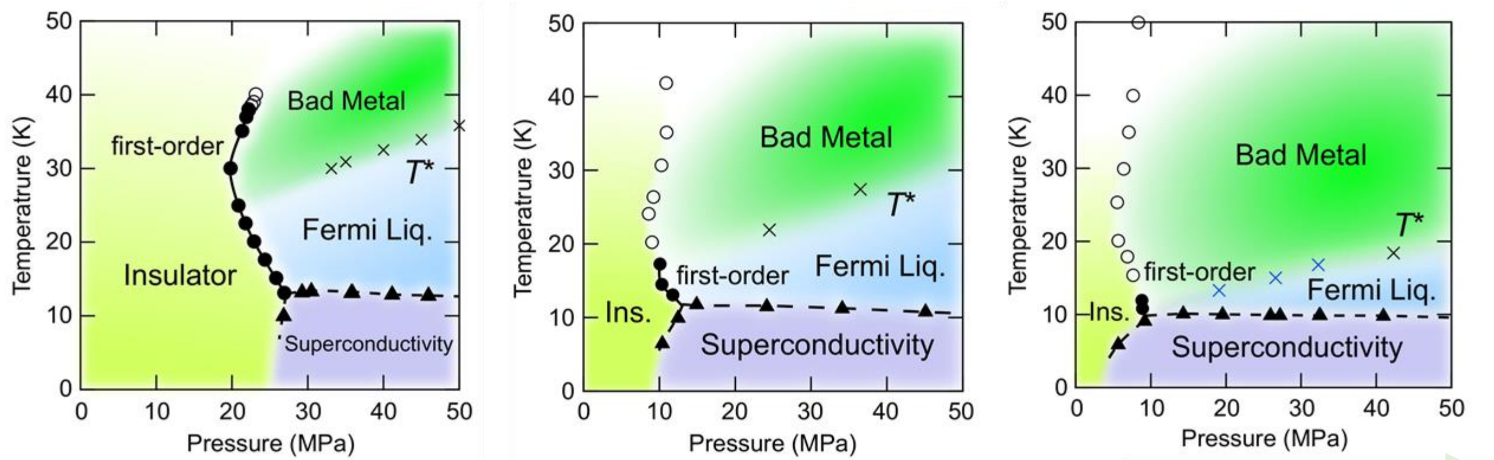




T. Sasaki, *Crystals* **2**, 374 (2012)

1: Effect of disorder on Mott transition

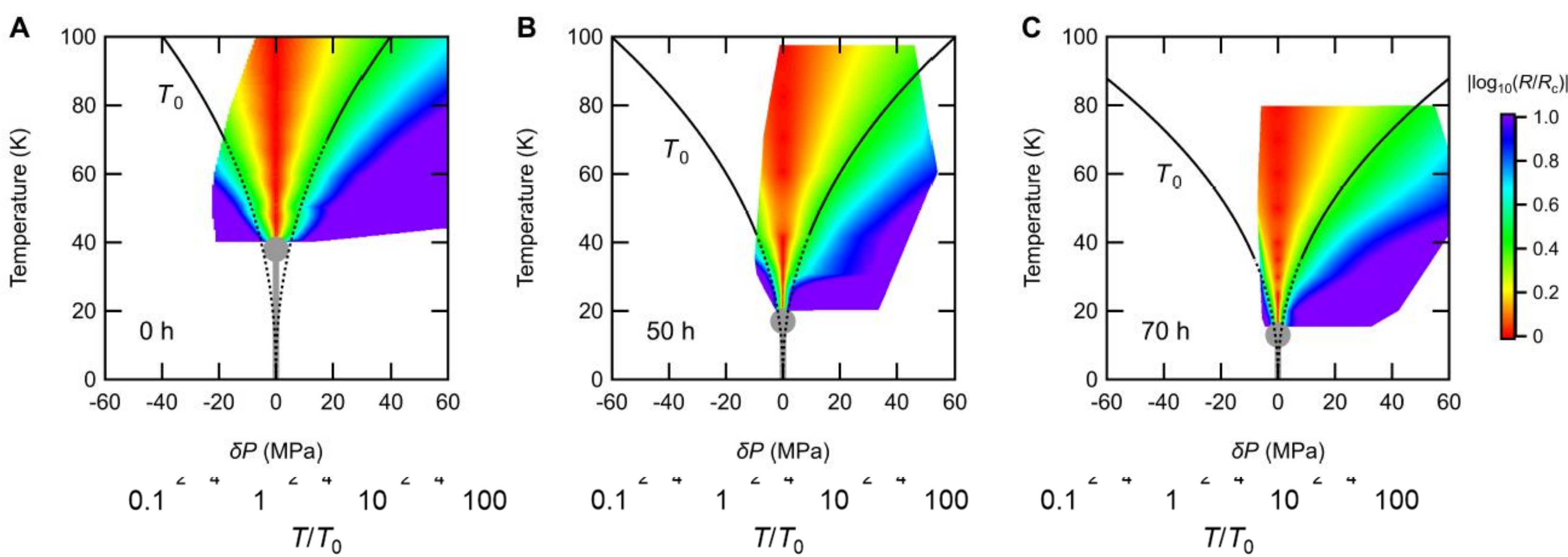
✓ *Drastic suppression of the critical endpoint = 2D Ising character*

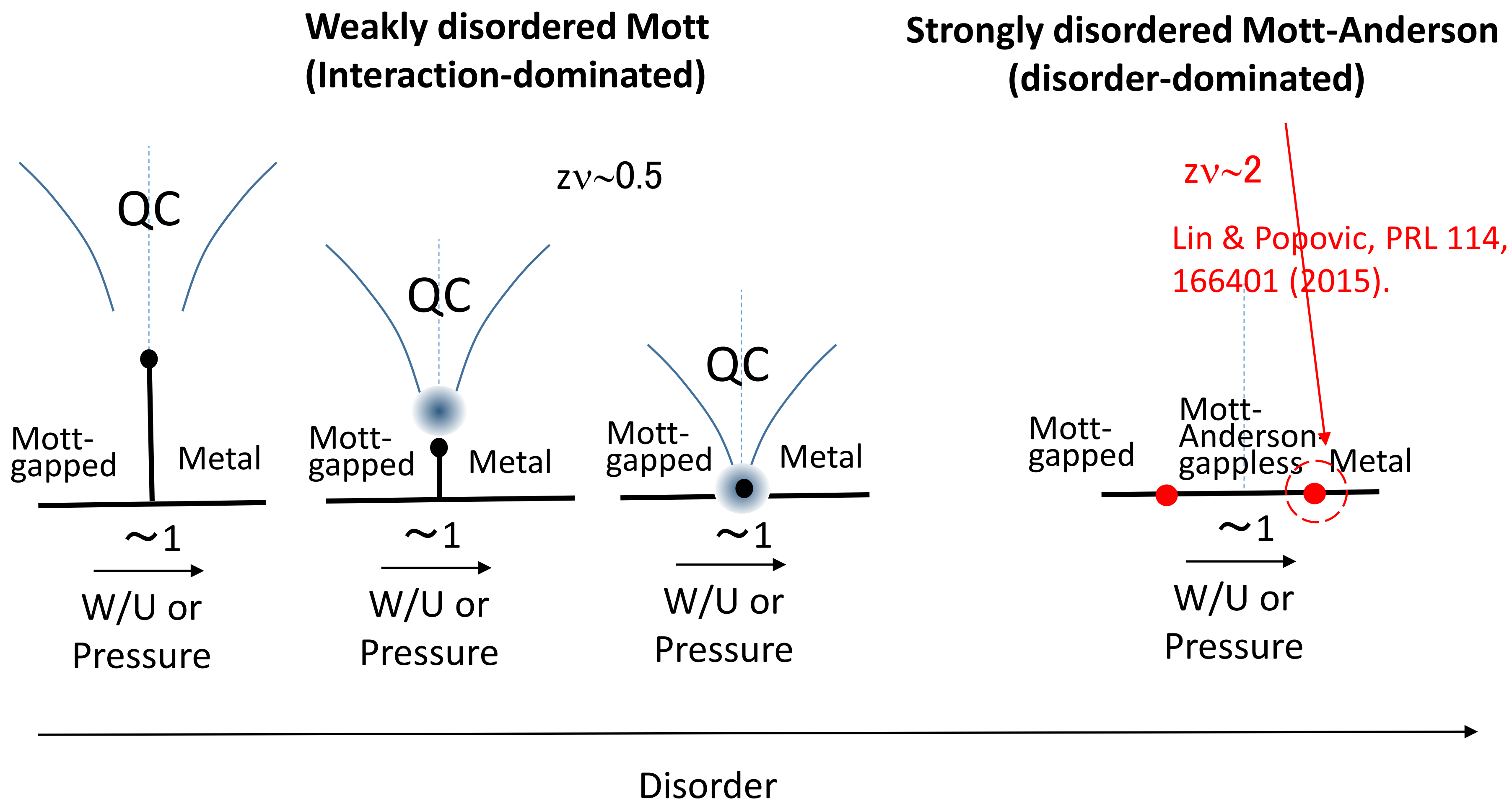


t_{irr} = 0 hours t_{irr} = 50 hours t_{irr} = 70 hours

✓ *Enhancement of the quantum critical fluctuations*

Urai et al, PRB (2019)





Weakly disordered Mott
(Interaction-dominated)
Strongly disordered Mott-Anderson
(disorder-dominated)
zν~0.5
zν~2
Lin & Popovic, PRL 114, 166401 (2015).
QC
QC
QC
Mott-gapped
Metal
Mott-gapped
Metal
Mott-gapped
Metal
Mott-gapped
Mott-Anderson gappless
Metal
~1
~1
~1
~1
W/U or Pressure
W/U or Pressure
W/U or Pressure
W/U or Pressure
Disorder

Today's talk

- Spin liquid
  - Absence of DM effect → itinerant spins ?
  - 6K anomaly → Instability of spinnon FS with singlet FFLO ?
- Doped spin liquid
  - non-Fermi liquid to Fermi liquid crossover
  - BEC to BCS crossover
- Mott quantum criticality
  - Disorder unveils Mott quantum criticality behind 1st-order transition

Collaborators

UTokyo (applied Phys.)	T. Furukawa (*Tokyo U. Sci.*), H. Oike, M. Urai, Y. Suzuki, K. Wakamatsu, J. Ibuka, Y. Shimizu (*Nagoya U*), H. Hashiba, Y. Kurosaki, K. Miyagawa
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Tohoku U.	M. Saito, S. Iguchi, T. Sasaki
RIKEN	R. Kato
NIMS	T. Isono, S. Uji
Inst. Phys. Zagrev	S. Tomic
UPMC	V. Ilakovac-Casess
Sherbrooke U.	M. Poirier