The Abdus Salam International Centre for Theoretical Physics

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1957-25

Miniworkshop on Strong Correlations in Materials and Atom Traps

4 - 15 August 2008

Spin/charge frustration of correlated electrons on triangular lattice.

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"Miniworkshop on Strong Correlations in Materials and atom Traps", Trieste, August 4-15, 2008

Spin/charge frustration of correlated electrons in triangular-lattice organics

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Collaborators

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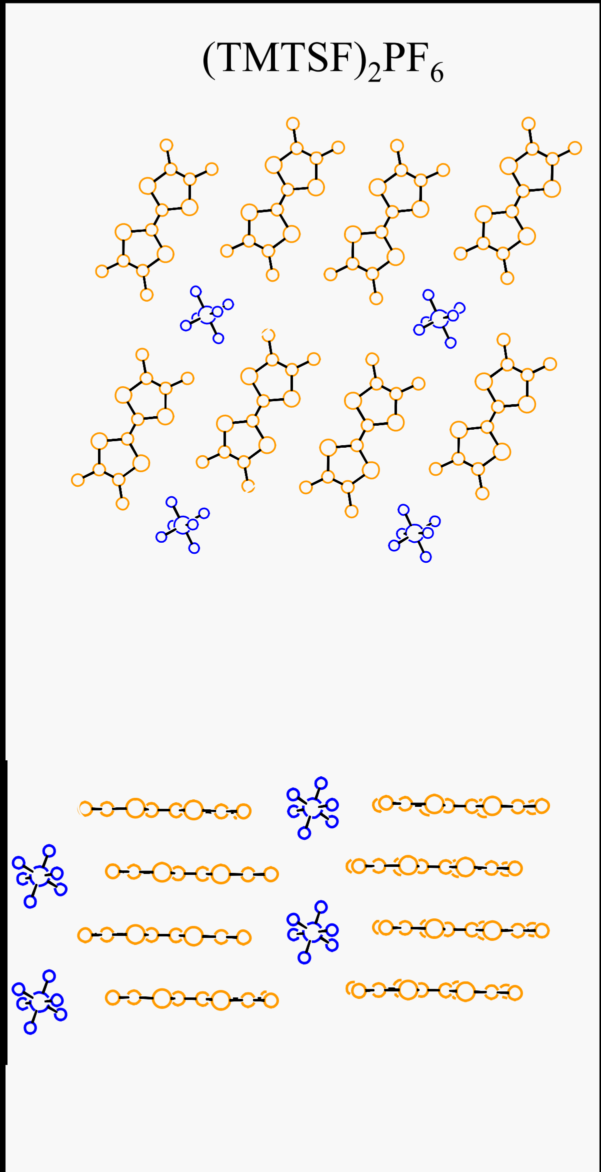
Y. Kurosaki, H. Kasahara, H. Hashiba, H. Ohike, K. Miyagawa

Kyoto Univ. **M. Maesato, G. Saito**

Outline

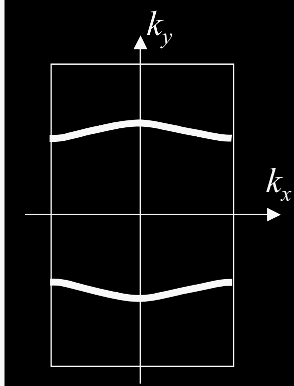
1. Three points for enjoying physics of organics
2. Spin ½ on triangular lattice ----- spin liquid
   - when pressurized; band width-varied
   - when doped; band filling-varied

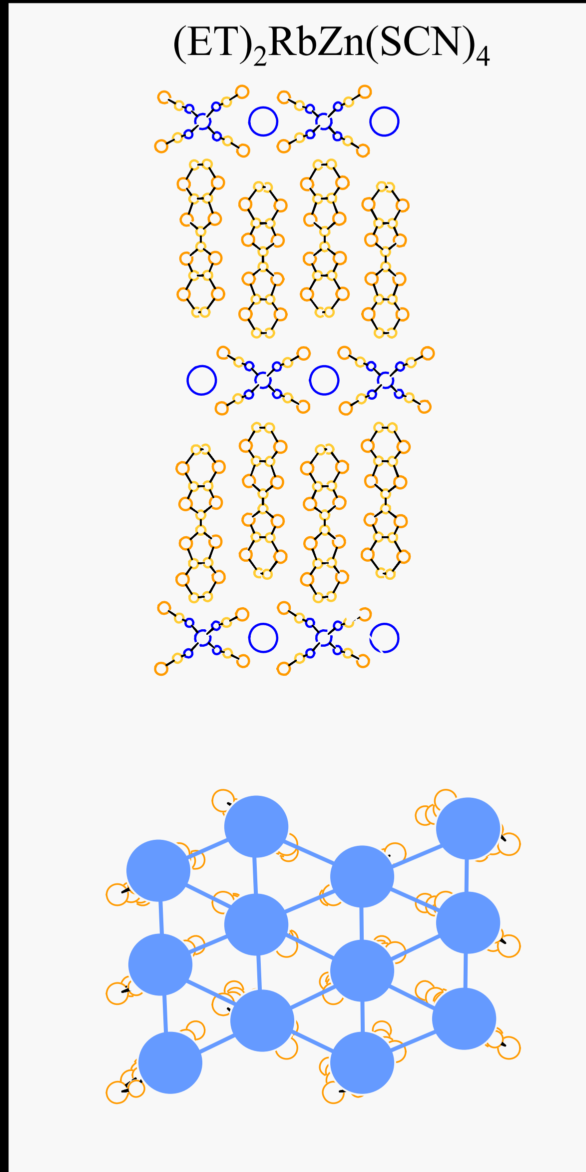
Structure: complicated in real space, but simple in k-space



Q1D

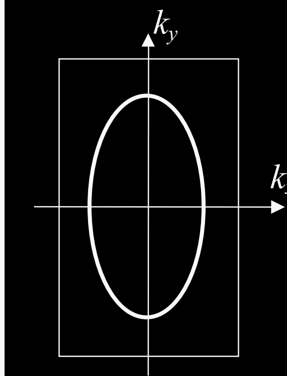
Fermi surface



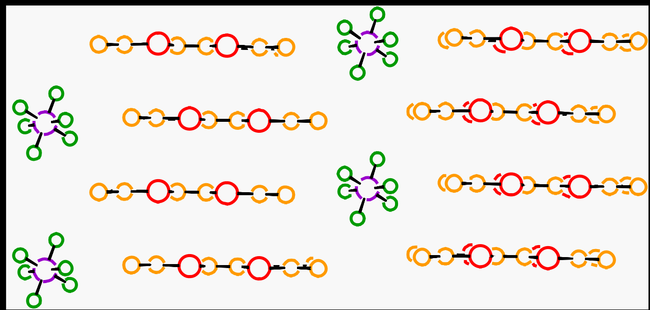


Q2D

Fermi surface

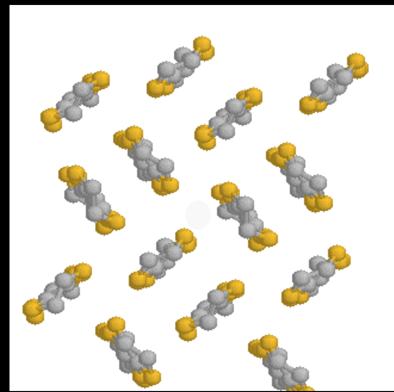


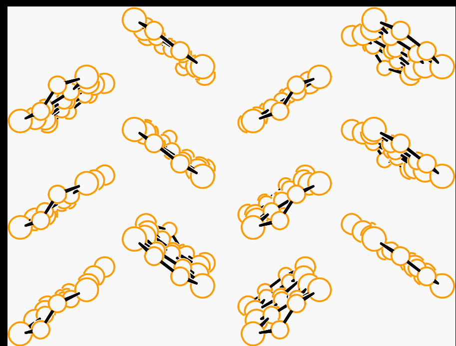
Variation in lattice geometry ➡ *Rich electronic phases*



SDW
Spin-Peierls transition
Wigner crystallization
Tomonaga-Luttinger liquid
Fermi liquid
Triplet SC ?

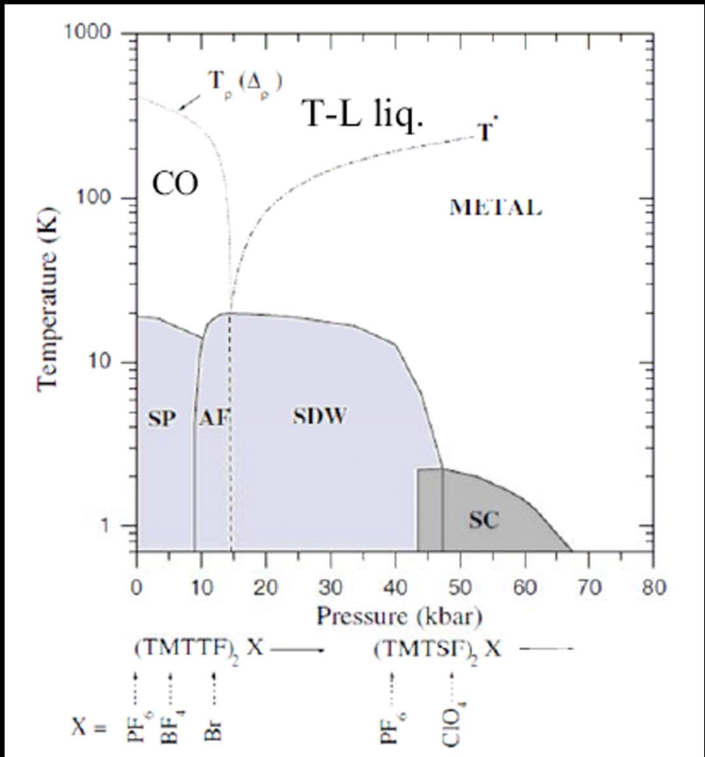
Mott transition
AF/ Spin liquid
d-wave SC

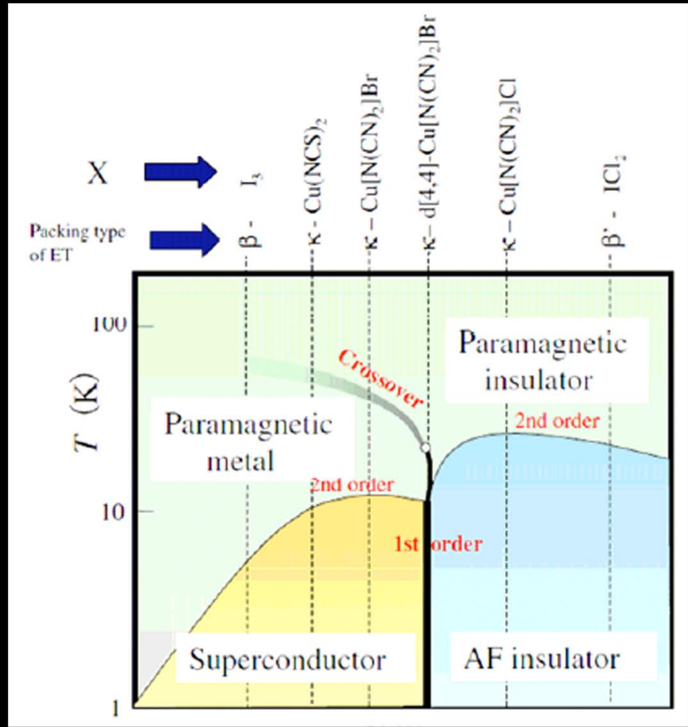


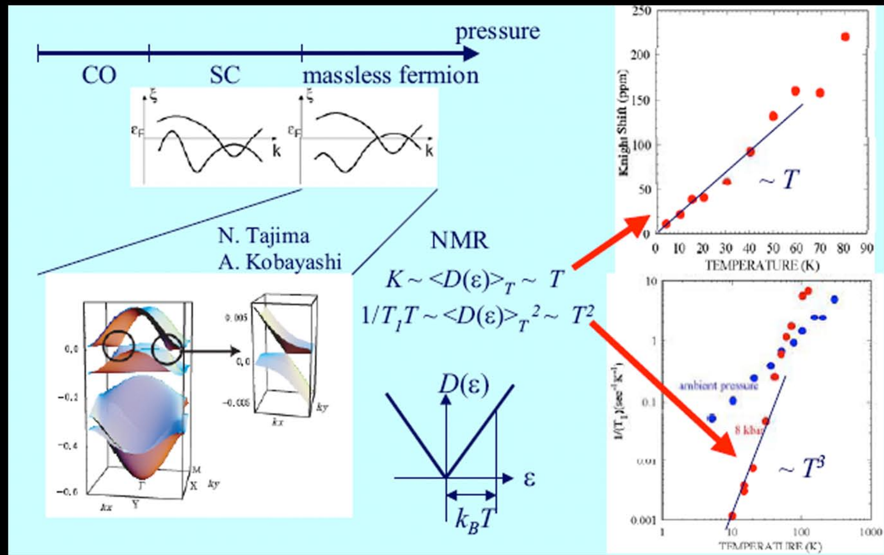


Wigner crystal/glass/liquid
Valence bond solid/glass
Massless Fermion
SC

Special Issue "Organic conductors" in J. Phys. Soc. Jpn. No.5, 75 (2006)

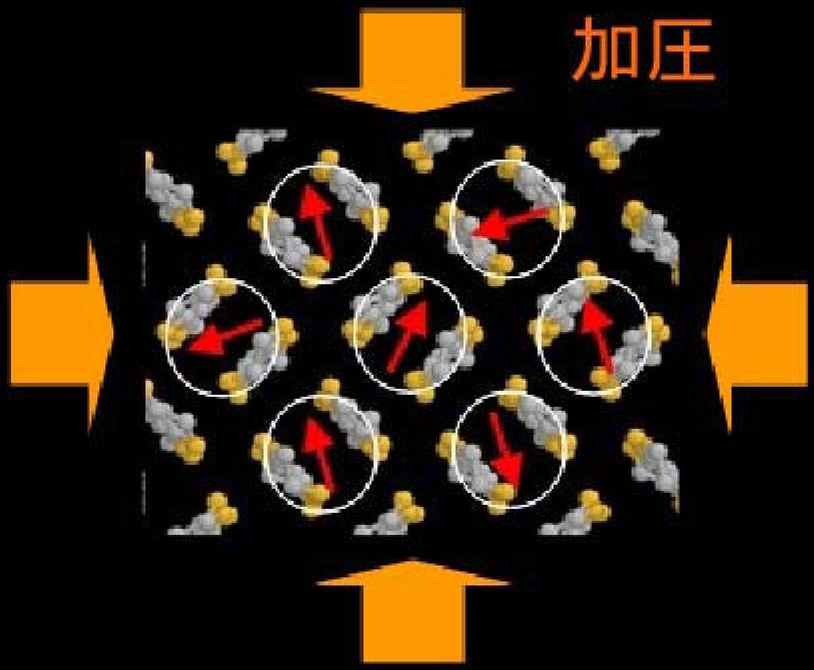

T-L liq.
CO
METAL
SP
AF
SDW
SC
Temperature (K)
Pressure (kbar)
(TMTTF)$_2$ X
(TMTSF)$_2$ X
X =
PF$_6$
BF$_4$
Br
PF$_6$
ClO$_4$


X
Packing type of ET
β - I$_3$
κ - Cu(NCS)$_2$
κ - Cu[N(CN)$_2$]Br
κ - d[4,4]-Cu[N(CN)$_2$]Br
κ - Cu[N(CN)$_2$]Cl
β' - ICl$_2$
T (K)
Crossover
Paramagnetic metal
Paramagnetic insulator
2nd order
2nd order
1st order
Superconductor
AF insulator


pressure
CO
SC
massless fermion
N. Tajima
A. Kobayashi
NMR
$K \sim \langle D(\varepsilon) \rangle_T \sim T$
$1/T_1T \sim \langle D(\varepsilon) \rangle_T^2 \sim T^2$
$D(\varepsilon)$
ε
k_BT
Knight Shift (ppm)
TEMPERATURE (K)
$\sim T$
ambient pressure
8 kbar
$\sim T^3$
TEMPERATURE (K)

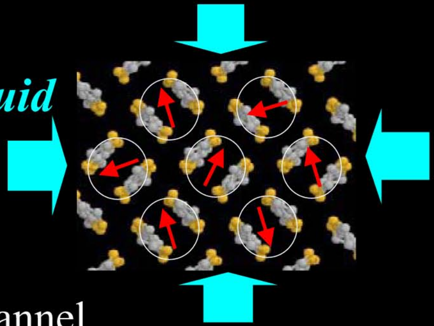
Highly compressible

➡ ***Pressure is quite effective to control organics***

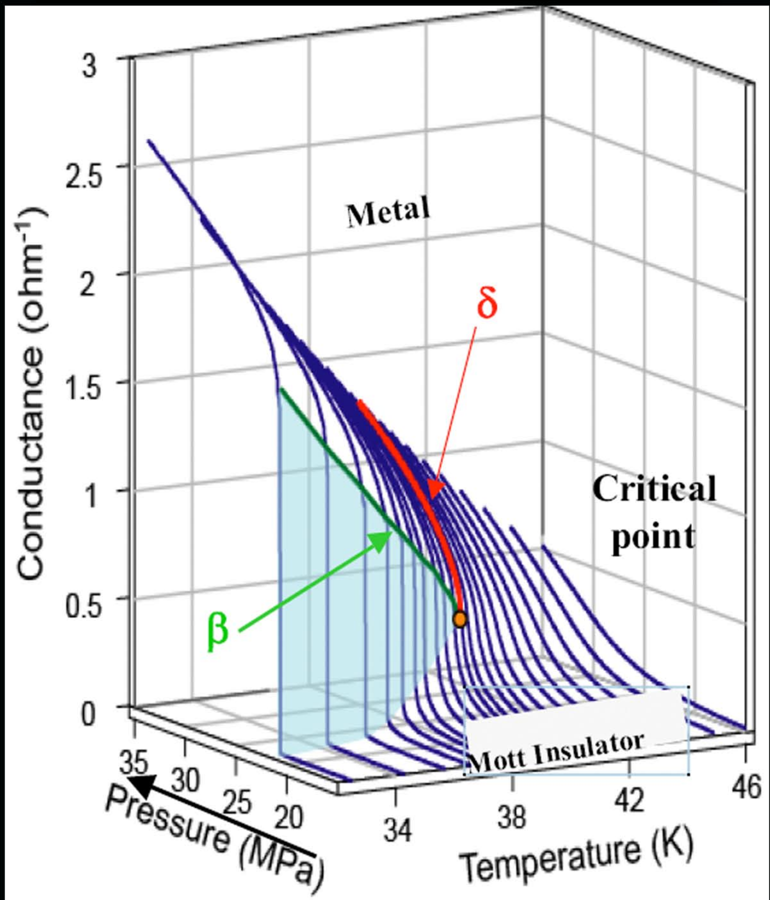


Mott transition in κ-(ET)$_2$Cu[N(CN)$_2$]Cl by pressure

Helium fluid pressure



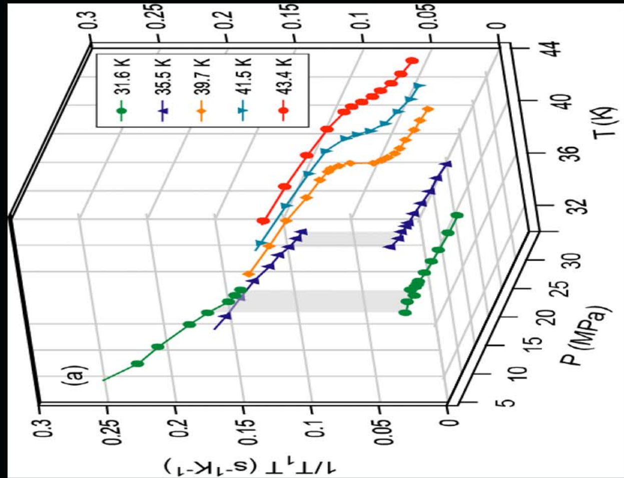
Charge channel

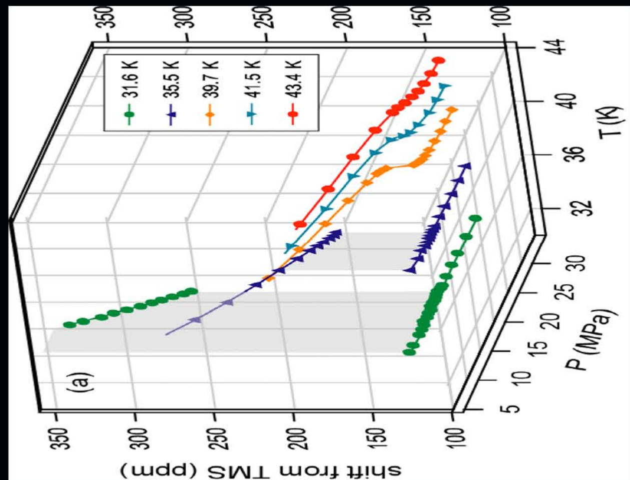


Kagawa *et al*., Nature 436 (2005) 534

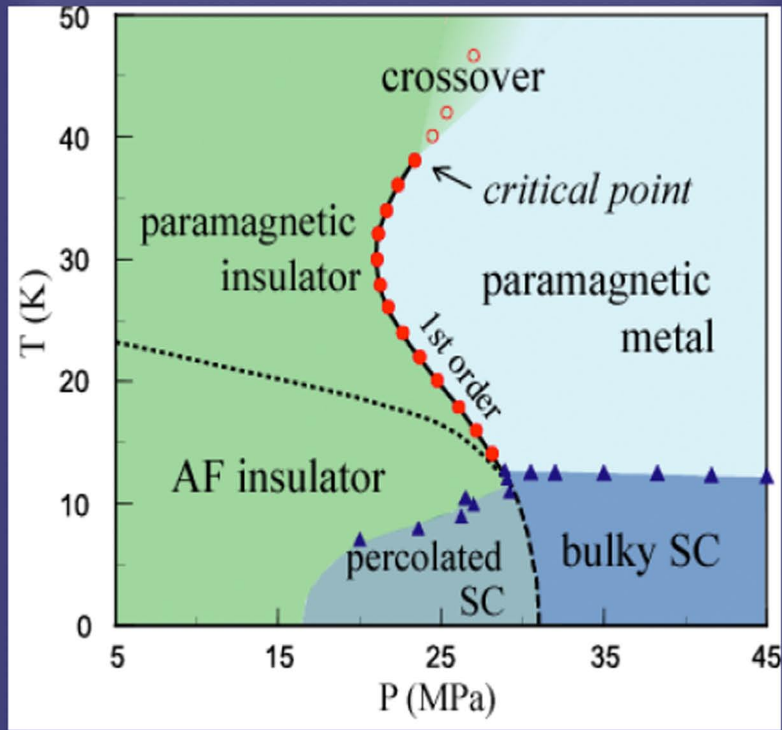
spin channel

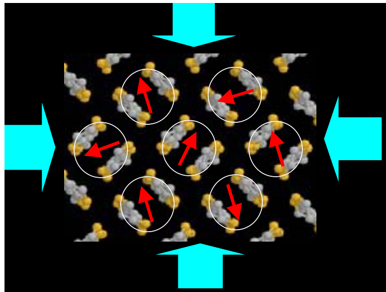
Kagawa *et al*. (2008)





Phase diagram

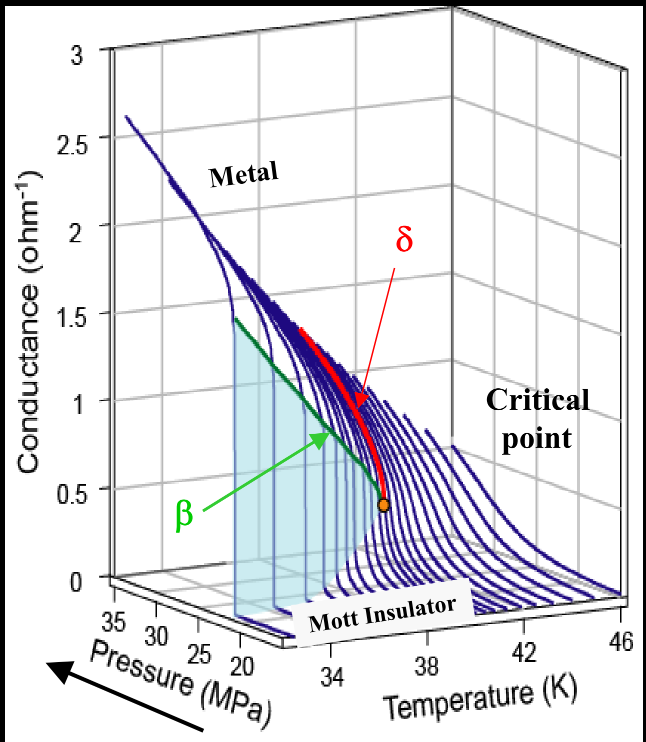




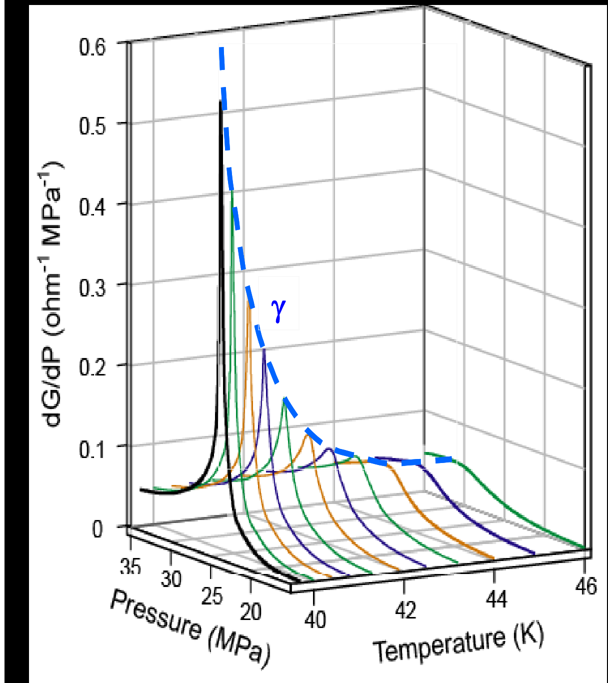
Mott Criticality and Mott scaling

Kagawa *et al*., Nature 436 (2005) 534

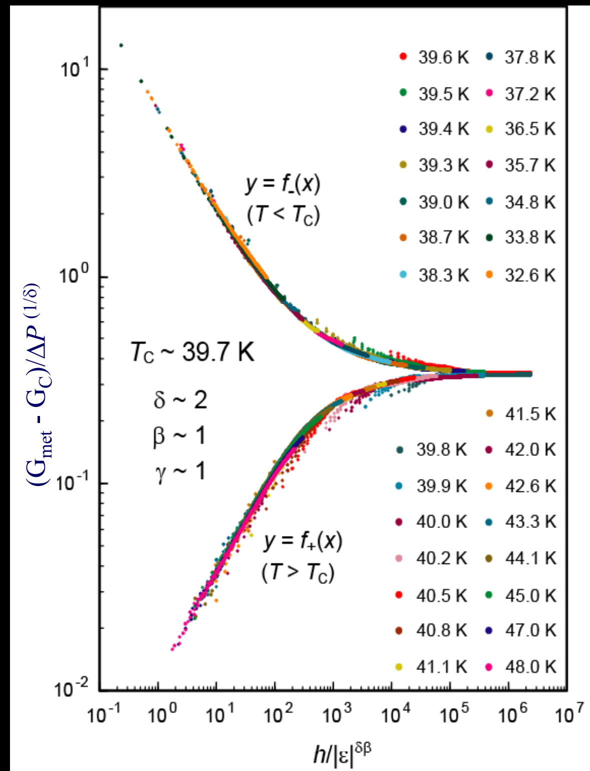
Conductance, *G*



Pressure derivative of Conductance, *dG*/*dP*



Scaling plot



Unconventional critical exponents

(δ, β, γ) ~ (2, 1, 1)

Scaling relation is fulfilled

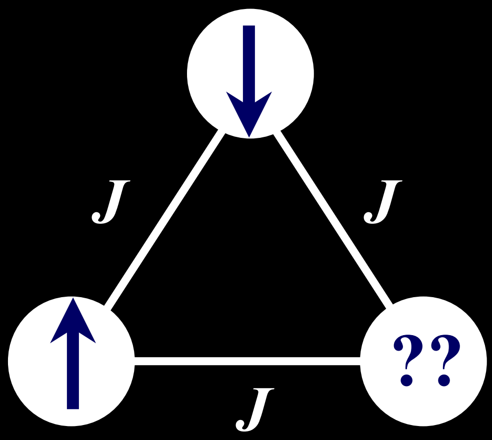
$$\delta = 1 + (\gamma / \beta)$$

Scaling function

$$G_{met}(P, T) - G_C = (\Delta P)^{1/\delta} f_{\pm}\left(\frac{\Delta P}{|\Delta T|^{\beta\gamma}}\right)$$

Interacting electrons on **Triangular lattice** are annoyed with frustration.

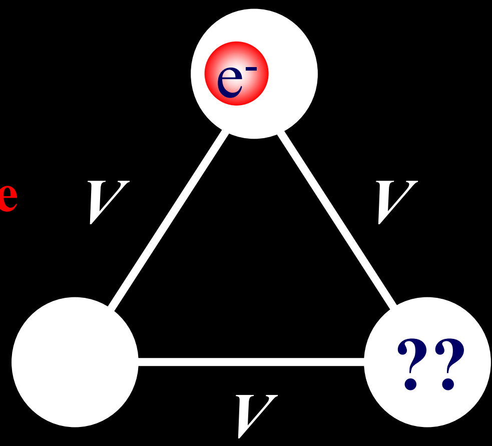
Spin-1/2 on the lattice
(½-filled band)



Antiferromagnetic interaction

frustration
in
spin and **charge**

Charge-1/2 on the lattice
(¼-filled band)



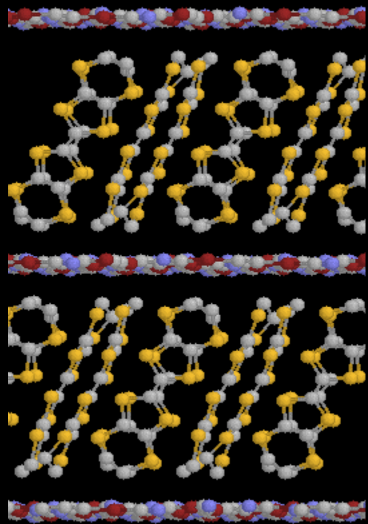
Inter-site Coulomb interaction

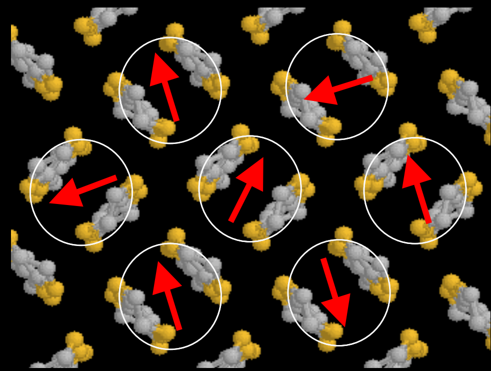
$$\mathcal{H} = \sum_{i,j,\sigma} t_{ij} C^{\dagger}_{i\sigma} C_{j\sigma} + \sum_{i} U n_{i\uparrow} n_{i\downarrow} + \sum_{<ij>} V_{ij} n_i n_j$$

Q2D organics κ-(ET)$_2$X; spin-1/2 on triangular lattice

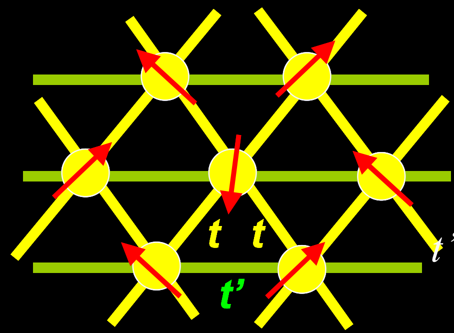
$ET^{+1/2}$

X^{-1}





Kino & Fukuyama dimer model



$t'/t = 0.5 \sim 1.1$

Triangular lattice
Half-filled band

Mott Trans. ⇨ (between $Cu[N(CN)_2]Cl$ and $Cu[N(CN)_2]Br$)

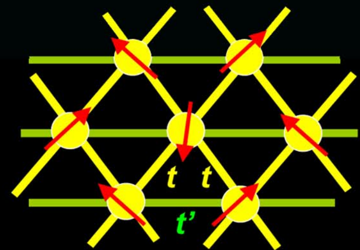
X^-	Ground state at ambient pressure	U/t	t'/t
$Cu_2(CN)_3$	Mott ins.	8.2	1.06
$Cu[N(CN)_2]Cl$	Mott ins.	7.5	0.75
$Cu[N(CN)_2]Br$	Metal (SC)	7.2	0.68
$Cu(NCS)_2$	Metal (SC)	6.8	0.84
$Cu(CN)[N(CN)_2]$	Metal (SC)	6.8	0.68
$Ag(CN)_2\ H_2O$	Metal (SC)	6.6	0.60
I_3	Metal (SC)	6.5	0.58

Ordering or quantum liquid

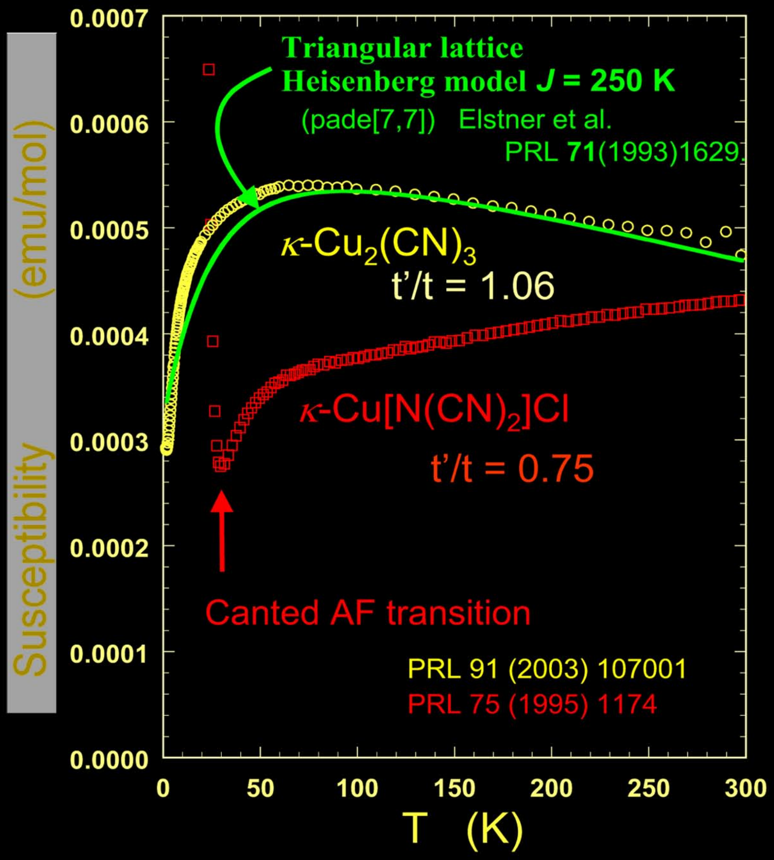
κ-$(ET)_2X$

Mott insulators

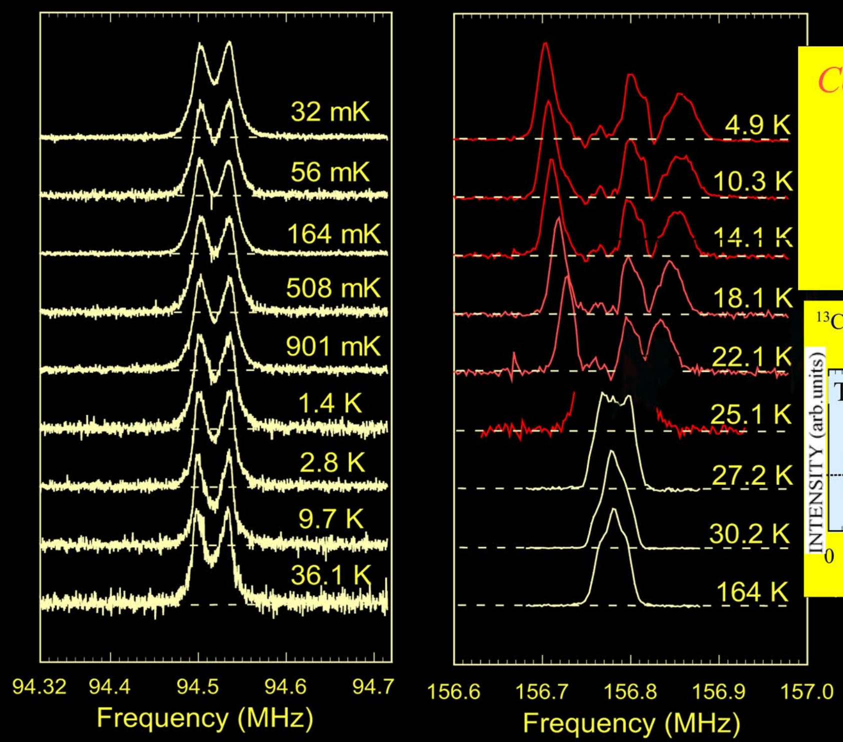
X	t'/t
$Cu_2(CN)_3$	1.06
$Cu[N(CN)_2]Cl$	0.75

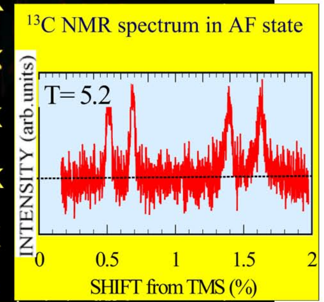


Uniform spin susceptibility



[1]H NMR spectrum probing internal field





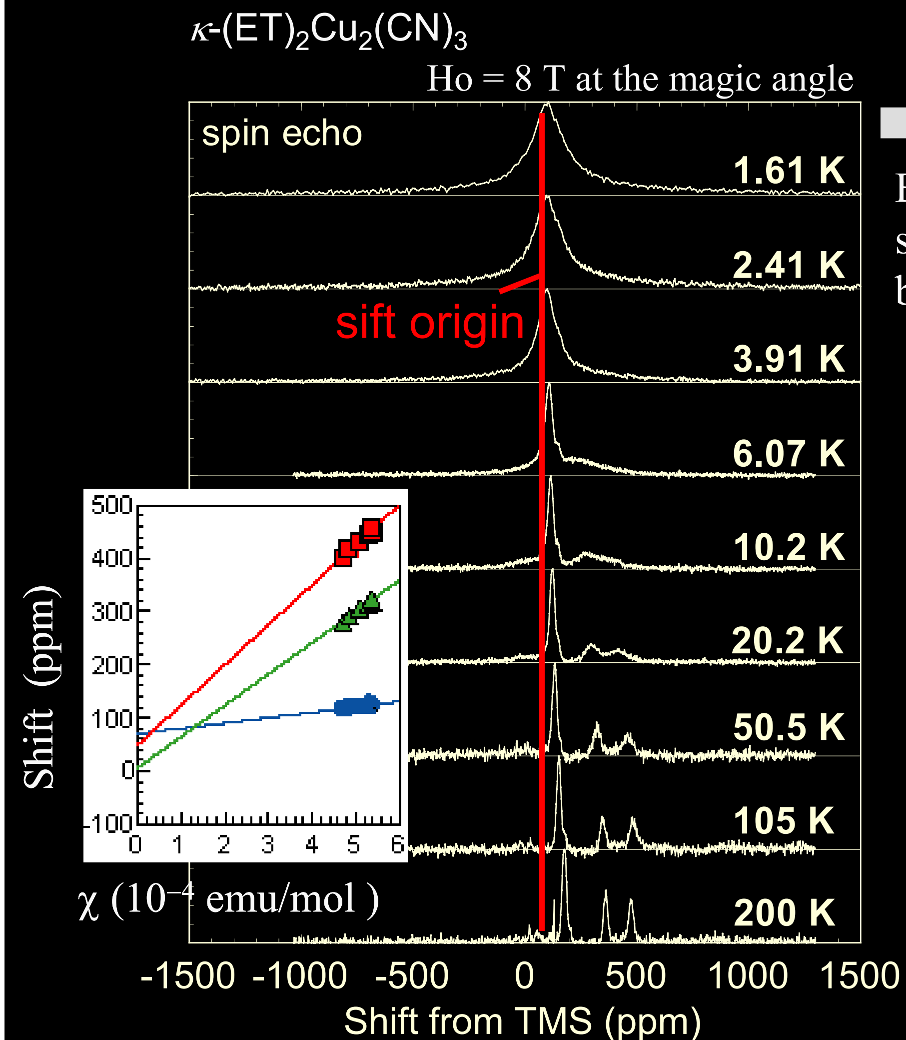
κ-$(ET)_2Cu_2(CN)_3$ κ-$(ET)_2Cu[N(CN)_2]Cl$

$t'/t = 1.06$ $t'/t = 0.75$

No ordering ⟺ AF ordered ($0.45\mu_B$)

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

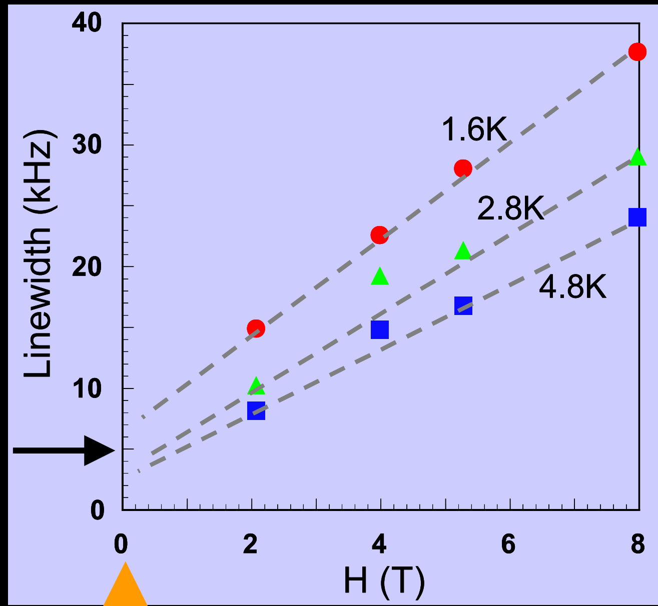
^{13}C NMR line broadening is induced by magnetic field at low-T



Bipolar & structureless broadening

Inhomogeneous staggered moments are field-induced.

Nuclear dipolar field

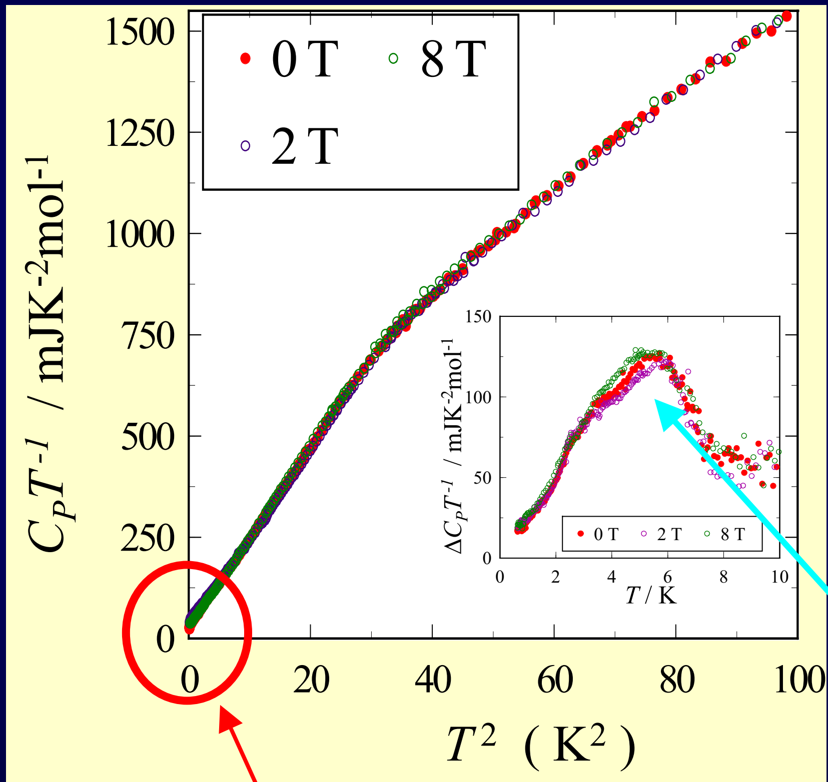


No internal field at H=0 by μSR (Ohira et al)

Spin anomaly around 5K and low-lying spin excitation in κ-$(ET)_2Cu_2(CN)_3$

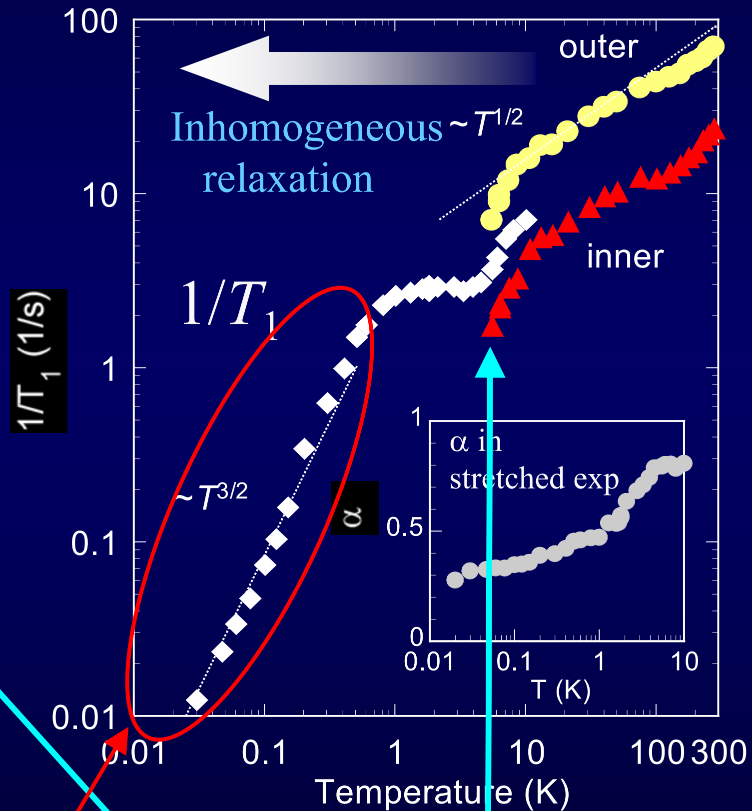
Specific heat

Yamashita *et al*., Nature Phys. (2008)



^{13}C NMR relaxation rate

Shimizu *et al*., PRB 70 (2006) 060510



Anomaly at 5-6 K

Low-lying spin excitation at low-T

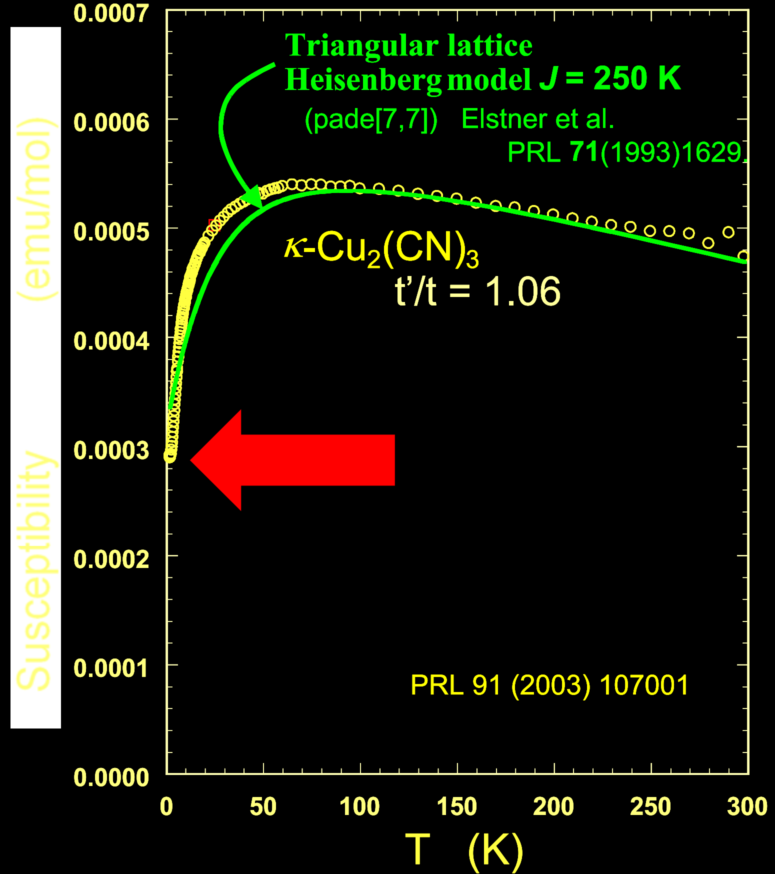
Wilson ratio ~ 1-2 !!!

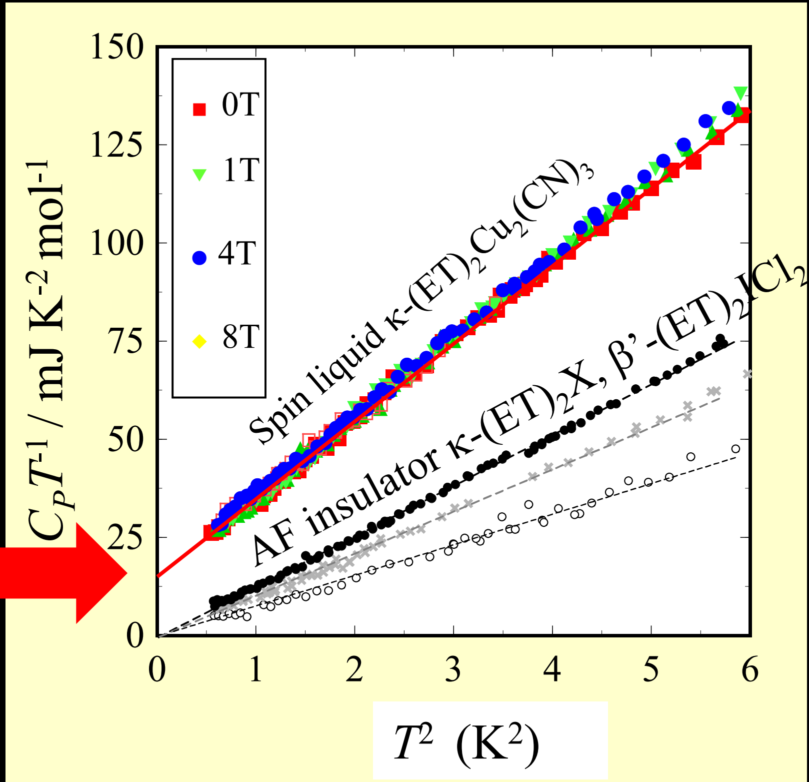
Degenerate Fermionic objects in Mott insulator

Spinon Fermi liq. ? P. A. Lee
Its instability at 5K ? P.A.Lee & T.Senthil

$\chi_{spin} = 3 \times 10^{-4}$ emu/mol

$\gamma = 15$ mJ/K^{2}mol



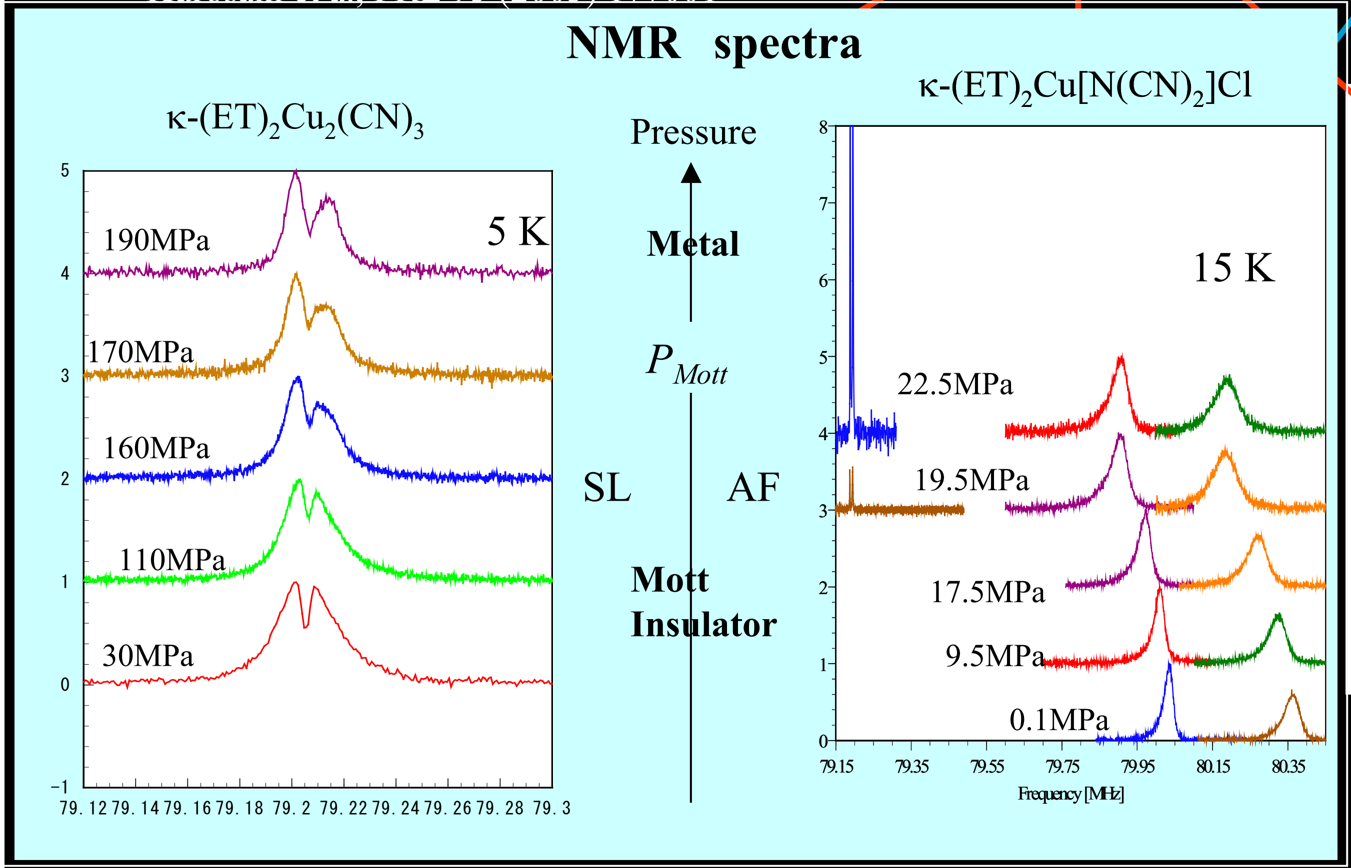


Yamashita *et al.*, Nature Phys. (2008)

Mott phase diagram of Antiferromagnet and spin liquid

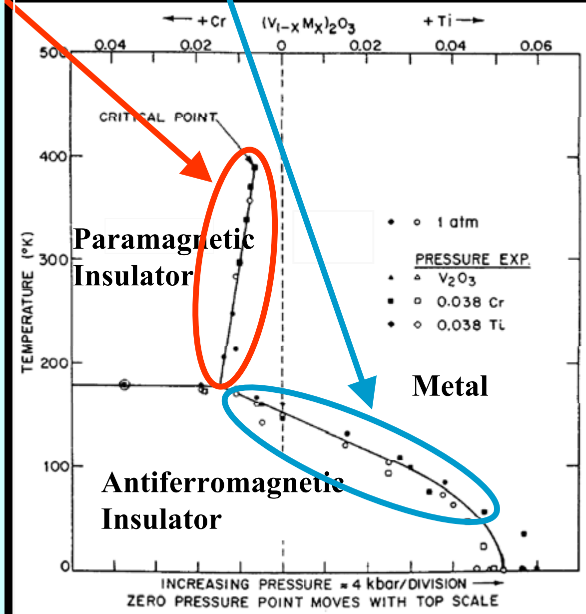
Clausius Clapeyron $dT/dP = \Delta V/\Delta S$ > 0 < 0

Kurosaki et a., PRL 95 (2005) 177001



Q2D spin liquid
κ-$Cu_2(CN)_3$

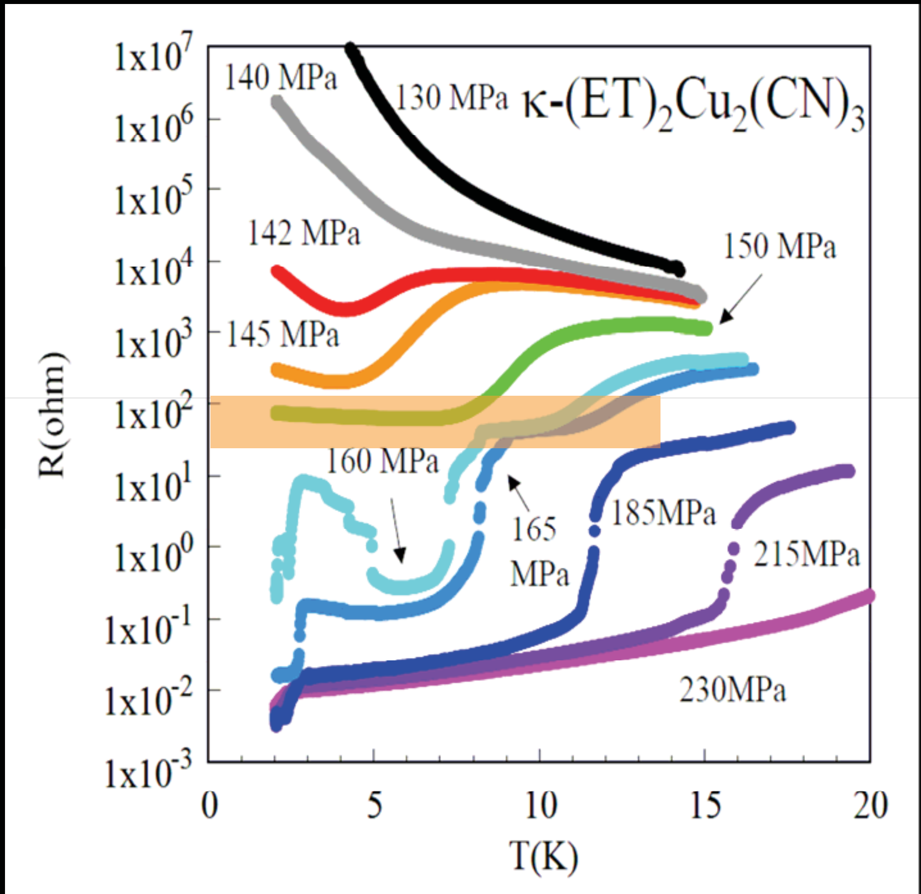
Q2D antiferromagnet
κ-$Cu[N(CN)_2]Cl$

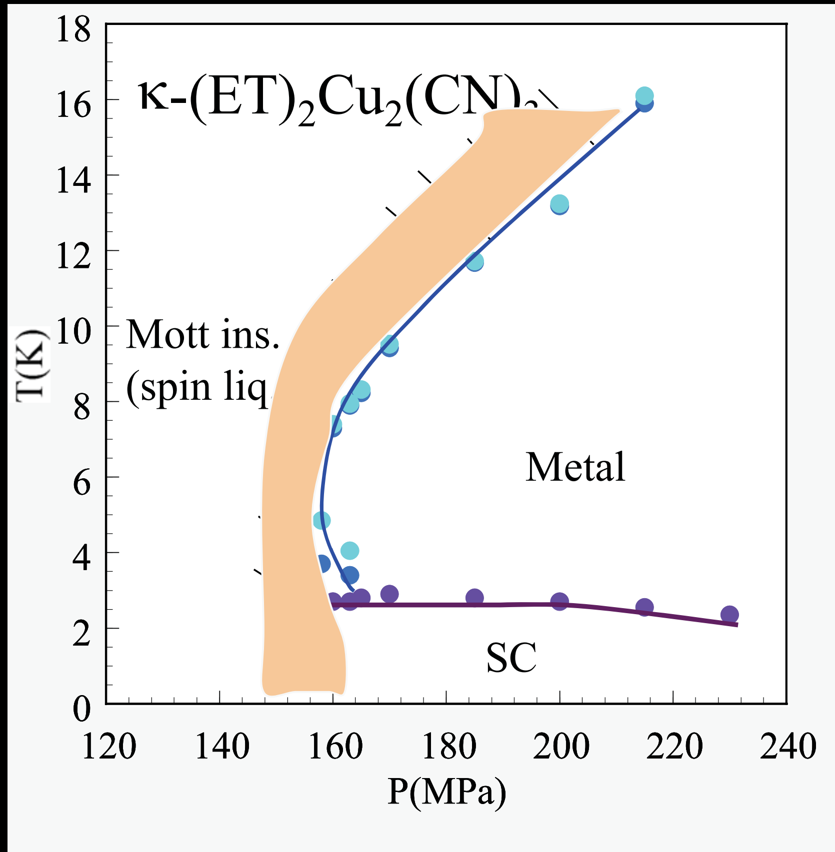


3D antiferromagnet
V_2O_3

Entropy of the spin liquid
is larger than that of Fermi liquid down to 1.5 K !

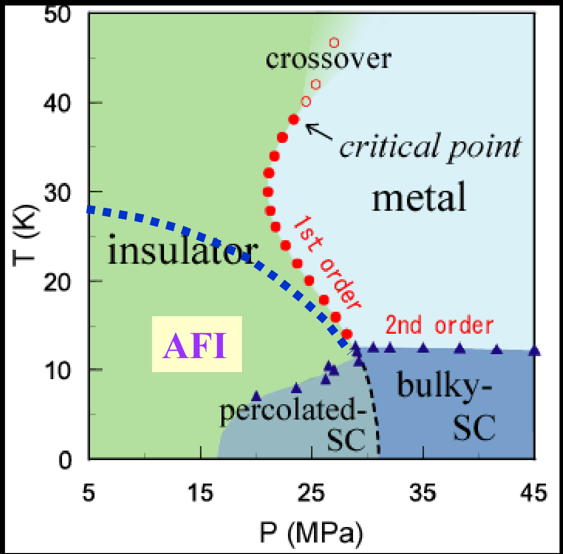
Charge-gapless non-metal just before Mott transition ?



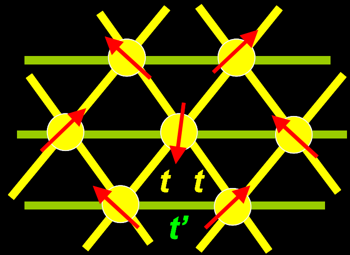


Charge gap in antiferromagnet and spin liquid

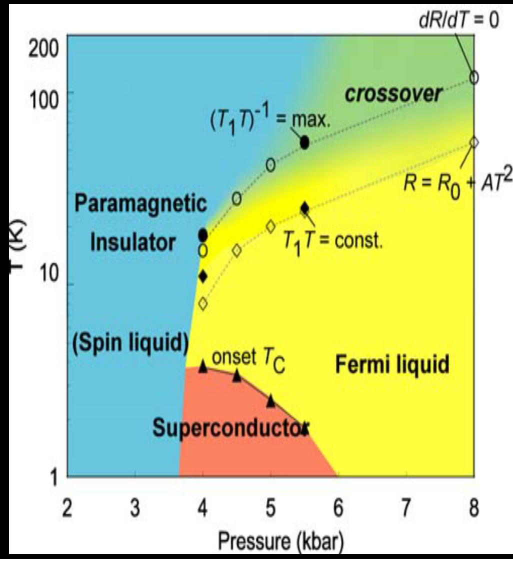
Charge gap is clearly opened on AF ordering, but remains undeveloped in spin liquid.

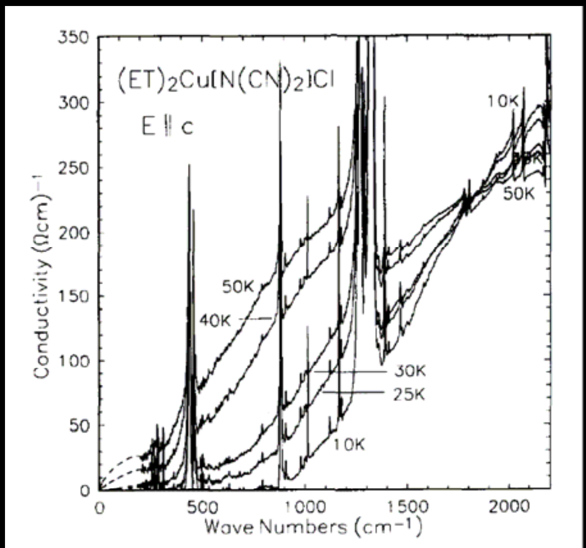


***t'/t* = 0.75**



***t'/t* = 1.06**

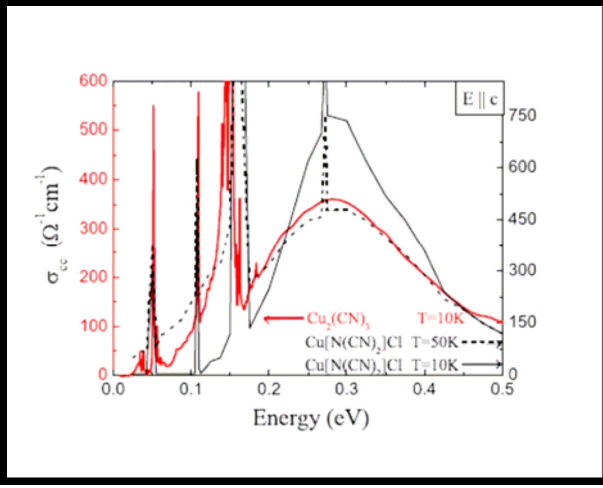


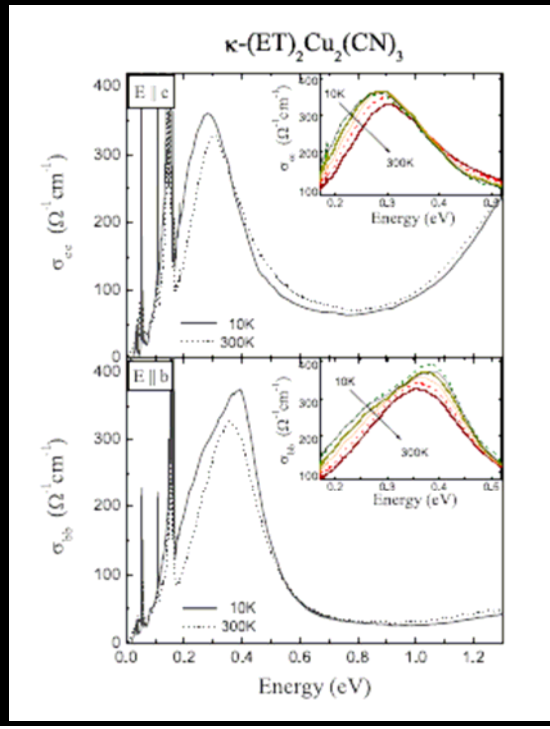


Kornelsen et al., SSC 81 (1992)343

Optical conductivity

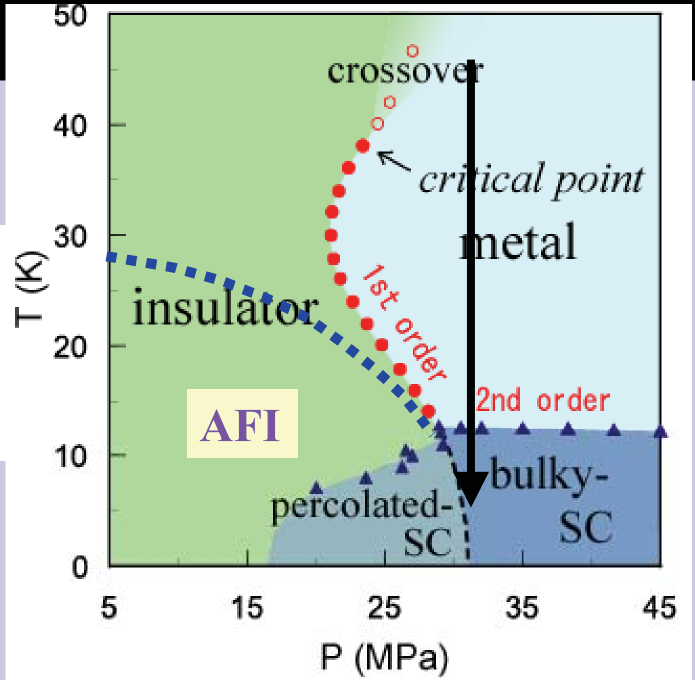
Kezsmarki et al.
PRB 74(2006)201101



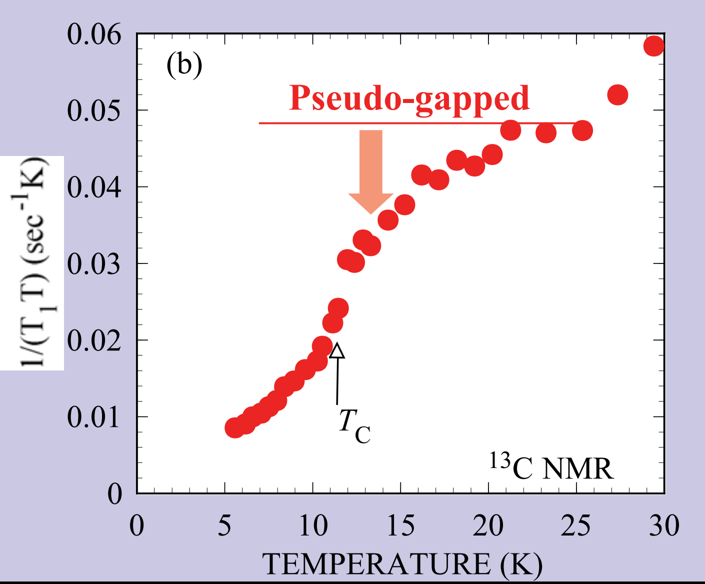


Pseudo-gapped nearby AFM

Not pseudo-gapped nearby spin liq.

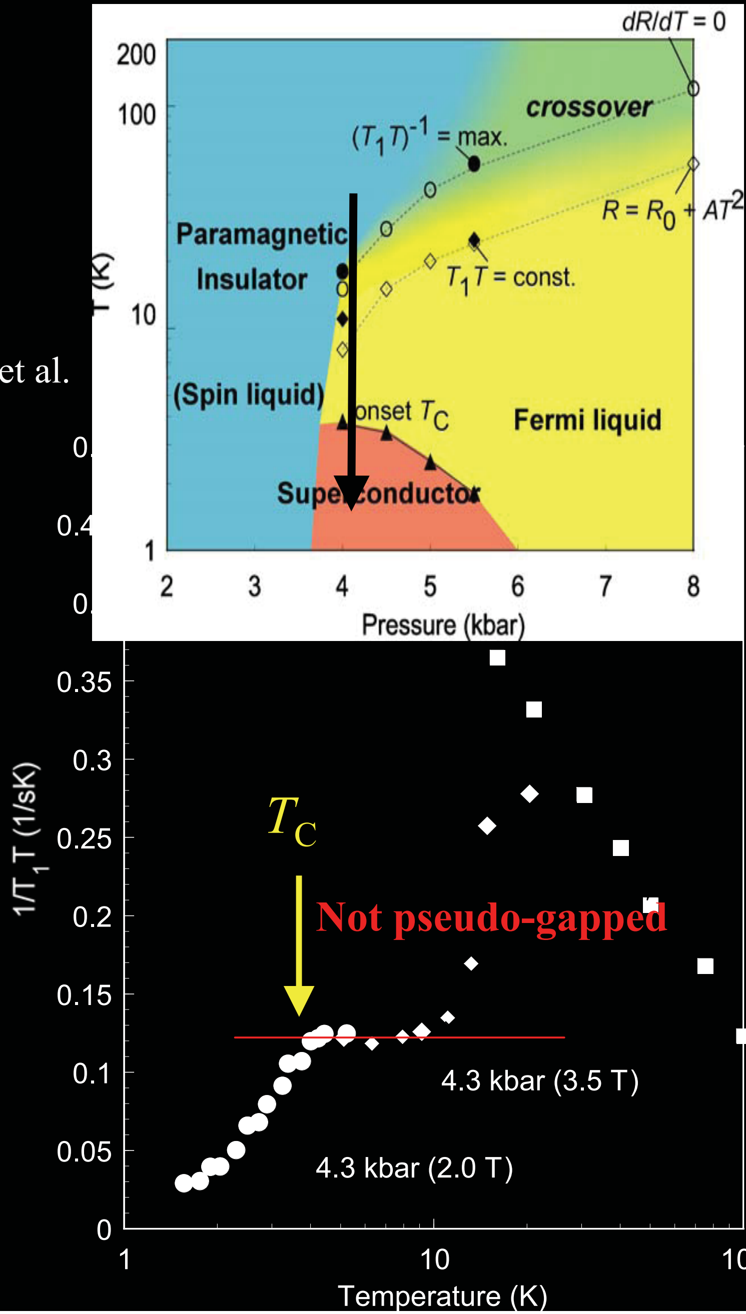


Miyagawa *et al.*, PRL89 (2002) 017003



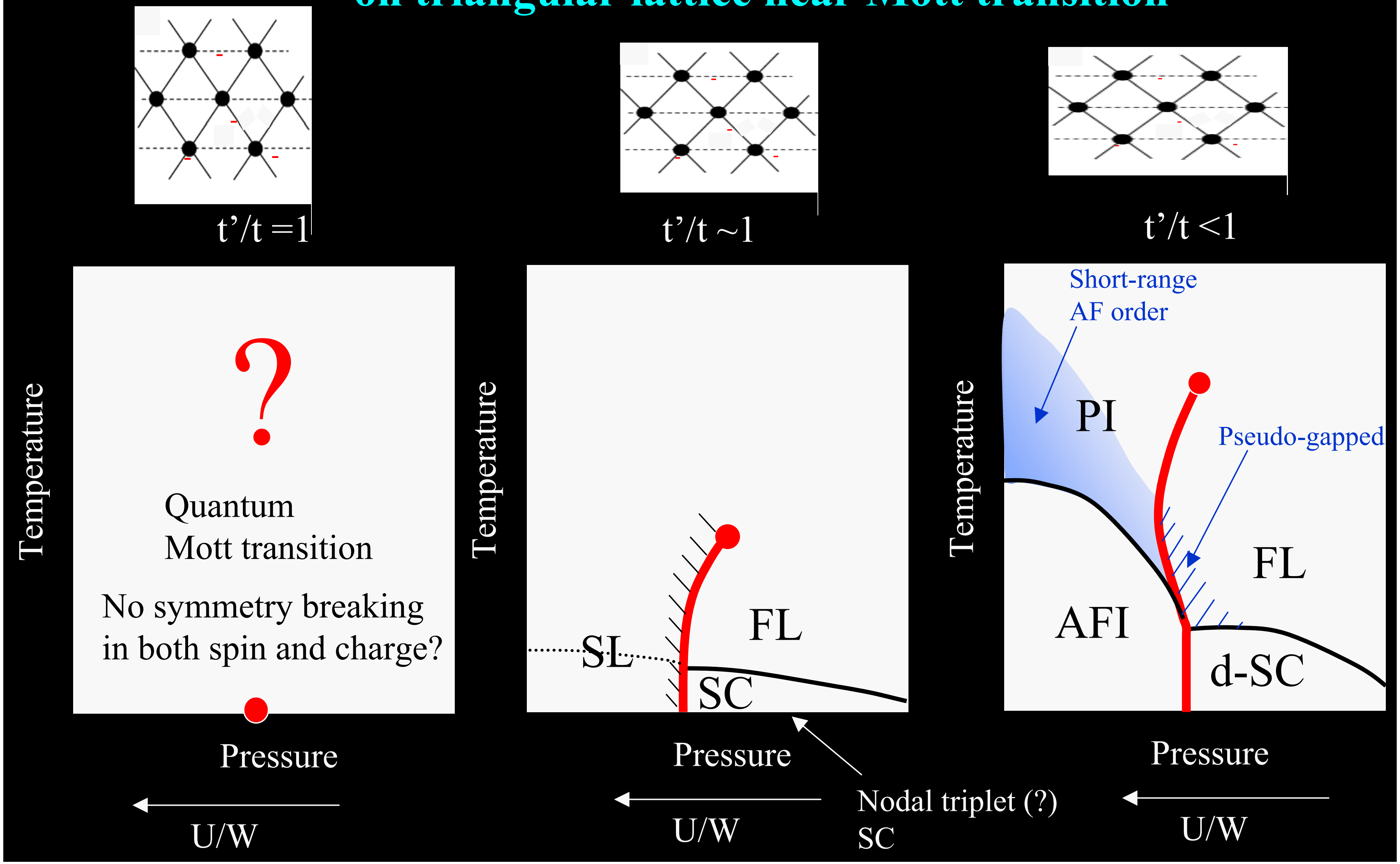
Shimizu et al. (2008)

^{13}C NMR $1/T_1T$



Guess on

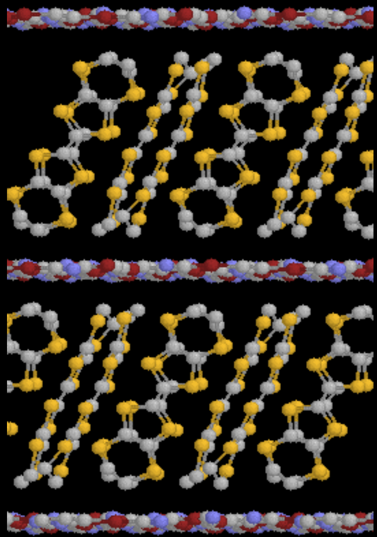
½-filled band correlated electrons on triangular lattice near Mott transition



Doped triangular lattice

ET layer

X layer



ET_4 ⟶ $(ET)_2^{+1+\delta}$

$Hg_{3-\delta}X_8$ (X=Br, Cl)

↑ Hole doping

κ-$(ET)_4Hg_{2.89}Br_8$ ----- 11% hole doped / dimer

κ-$(ET)_4Hg_{2.78}Cl_8$ ----- 22 % hole doped / dimer

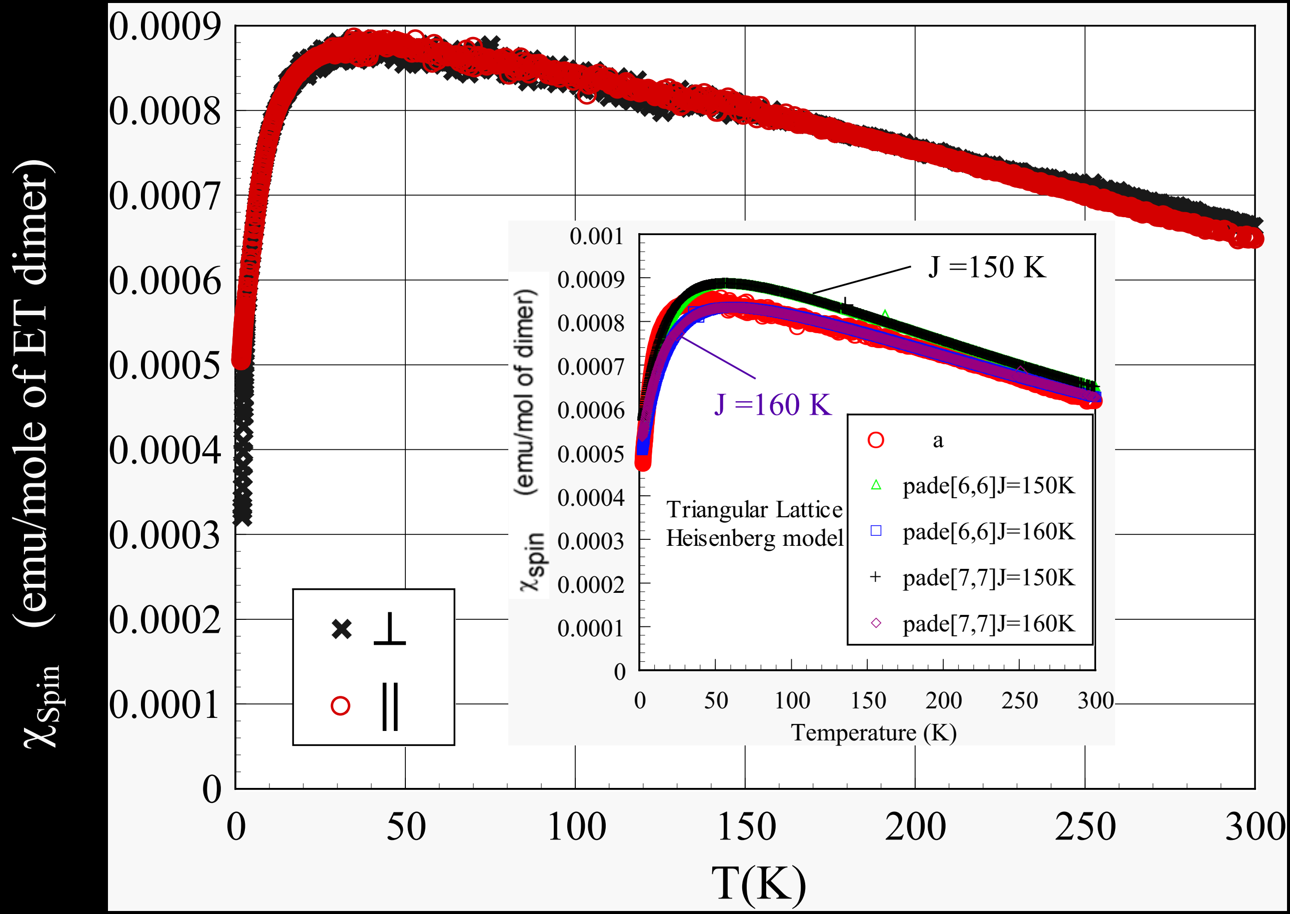
	U/t	*t'/t*	
κ-$(ET)_2I_3$	6.48	0.58	Metal/SC
κ-$(ET)_2Cu(NCS)_2$	6.98	0.86	
κ-$(ET)_2Cu[N(CN)_2]Br$	7.20	0.68	
κ-$(ET)_2Cu[N(CN)_2]Cl$	7.58	0.74	Mott insulator
κ-$(ET)_2Cu_2(CN)_3$	8.20	1.06	
κ-$(ET)_4Hg_{2.89}Br_8$	10.01	1.02	superconductor
κ-$(ET)_4Hg_{2.78}Cl_8$	10.32	1.11	metal

← **$(U/t)_{critical}$** (boundary between κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu[N(CN)_2]Cl$)

Magnetic susceptibility of doped triangular lattice

κ-$(ET)_4Hg_{2.89}Br_8$ (t’/t= 1.02)

Experiment by Taniguchi
Analysis by Shimizu

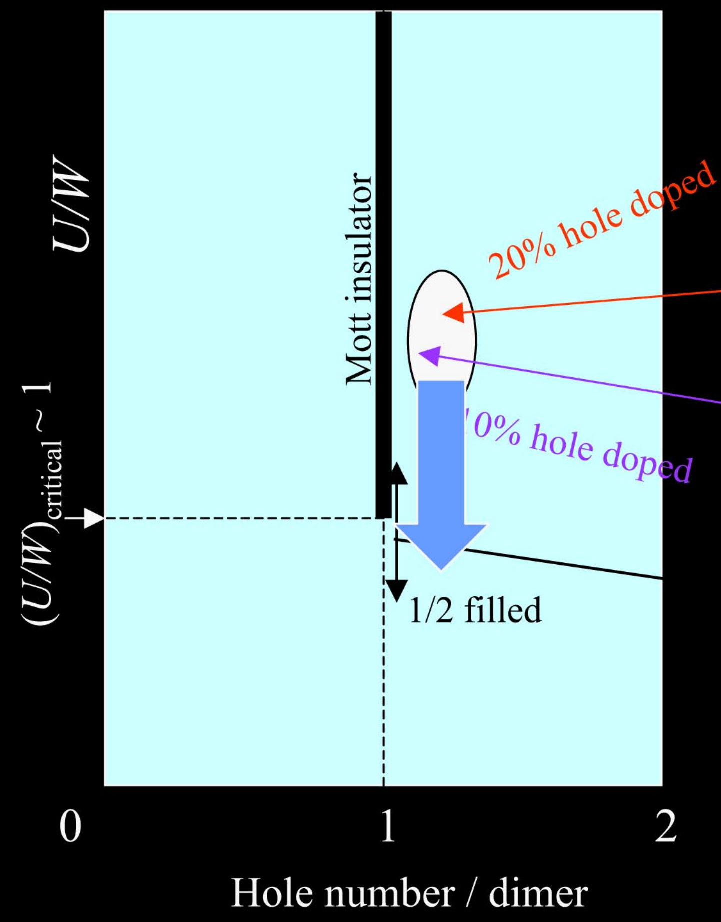


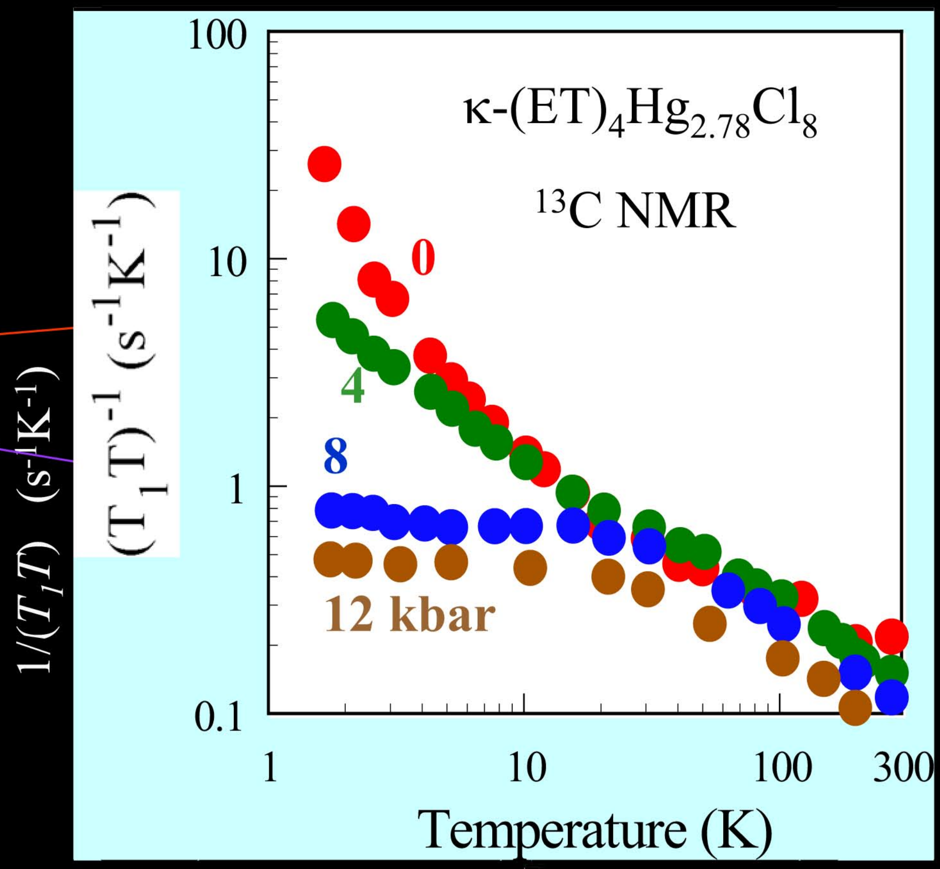
Doped triangular-lattice Mott insulator

κ-$(ET)_4Hg_{2.89}Br_8$ & κ-$(ET)_2Hg_{2.78}Cl_8$

Enhanced AF fluctuations at low-T without LRO ! → quantum critical fluctuations

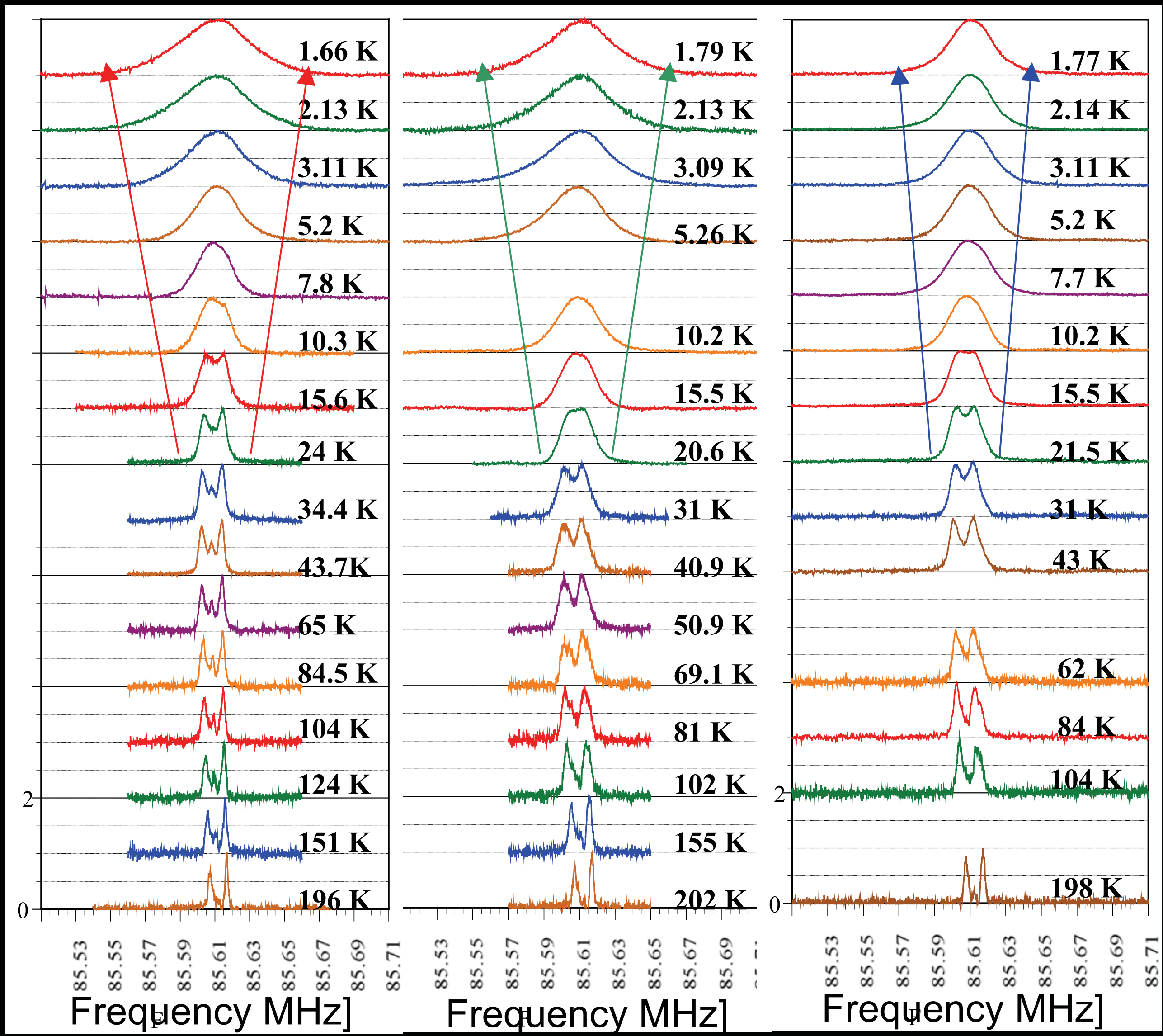
Enhanced inhomogeneity at low-T

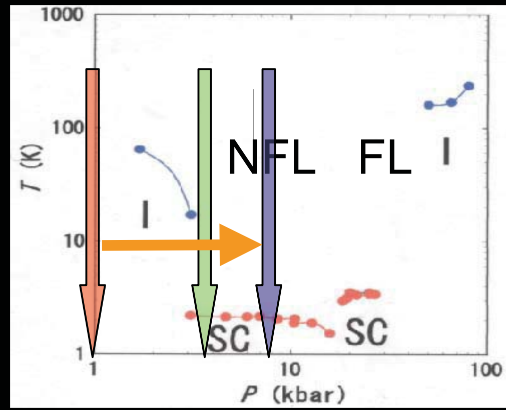


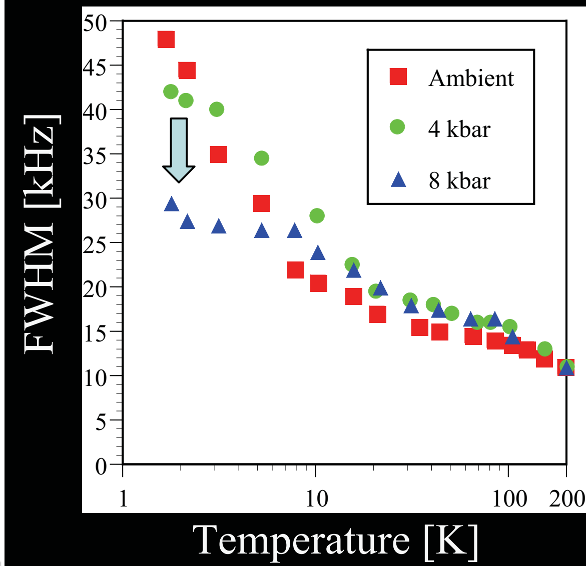


NMR spectra under pressure

0 kbar　4 kbar　8 kbar

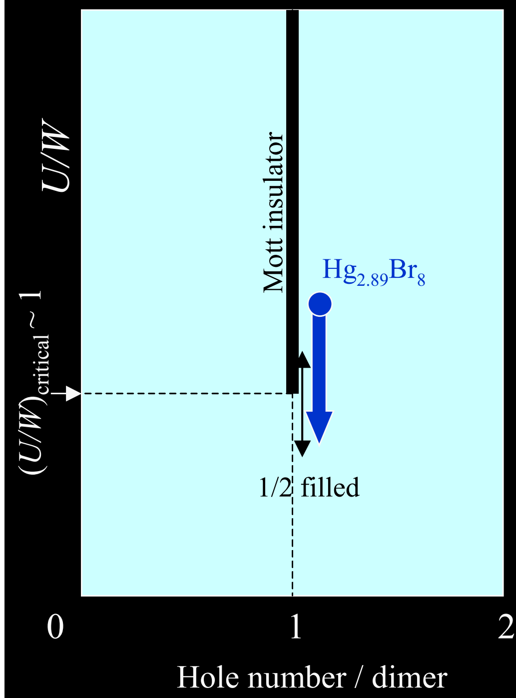




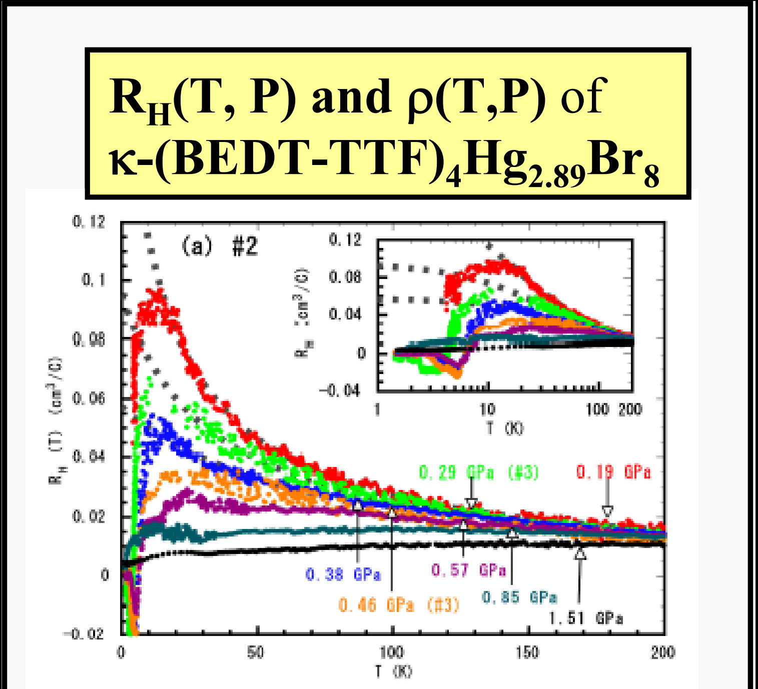


Pressure drives non-Fermi liquid to Fermi liquid

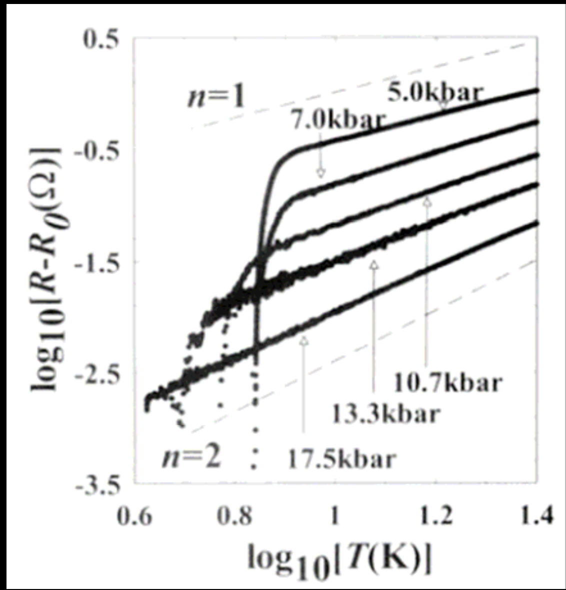
κ-$(ET)_4Hg_{2.89}Br_8$

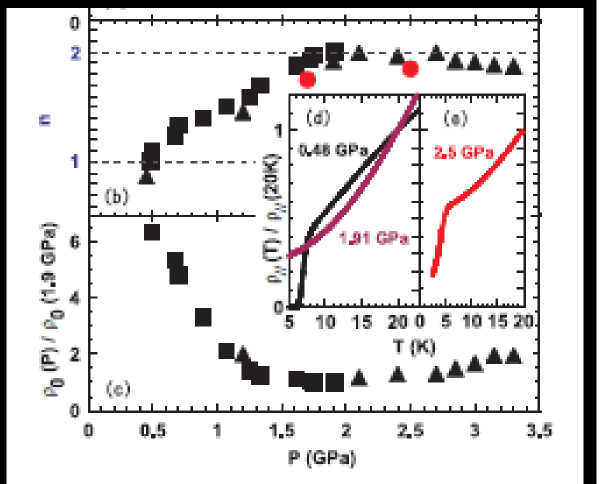


Hall coefficient



Resistivity





H. Taniguchi et. al., J.PSJ. 76 (2007) 113709

Doping without disorder → Mott insulator !! ??

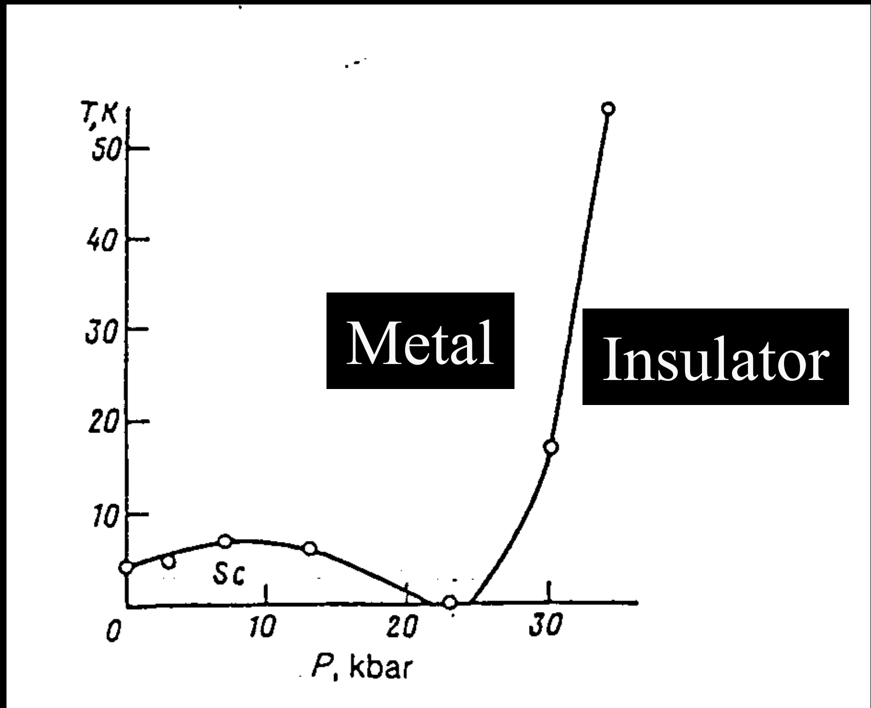
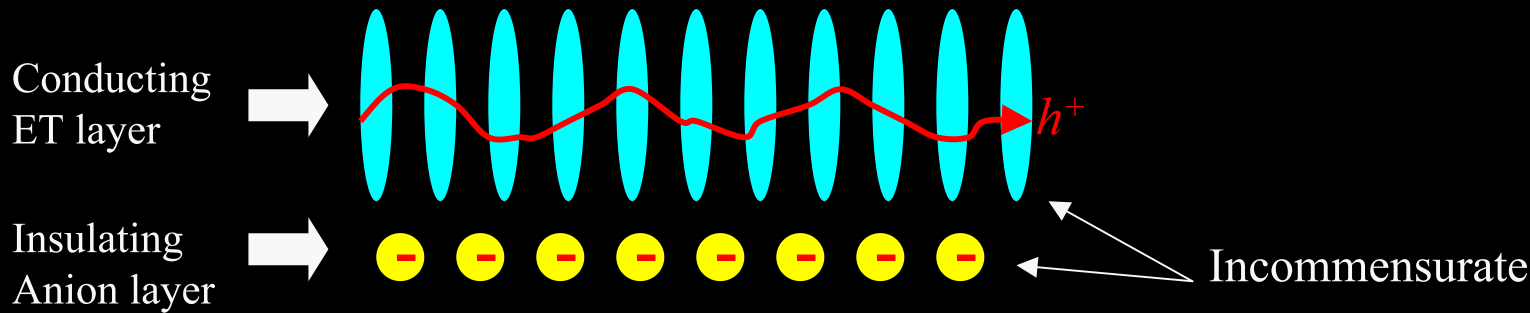


FIG. 5. T–p phase diagram for $(ET)_4Hg_{2.89}Br_8$.

Bud'ko et al.,
Sov. Phys. JETP 74 (1992) 983



Concluding remarks

Spin liquid or AF order depending on *t'/t*

Spin liquid appears near Mott transition

•Nature of Spin liquid

Low-lying excitations in NMR 1/T and finite γ

5K anomaly in NMR, C and κ -----hidden order or some crossover ?

•Spin frustration affects the charge near Mott transition

Charge-gapless non-metal just before Mott transition of spin liq.

----------- Novel interplay of spin and charge?

•Doped triangular lattice

χ well reproduced by triangular lattice Heisenberg model

spin fluctuations persistent down to low-T

experiments from *t-J* to Hubbard by pressure

Question

Inhomogenous spin states in nondoped and doped triangular lattice is intrinsic or extrinsic?