YKIS2007 "Interaction and Nanostructural Effects in Low-Dimensional Systems", Kyoto, Nov. 14, 2007

Mott physics and frustration in 2D

--- spin liquid, Mott transition and superconductivity ---

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Collaborators

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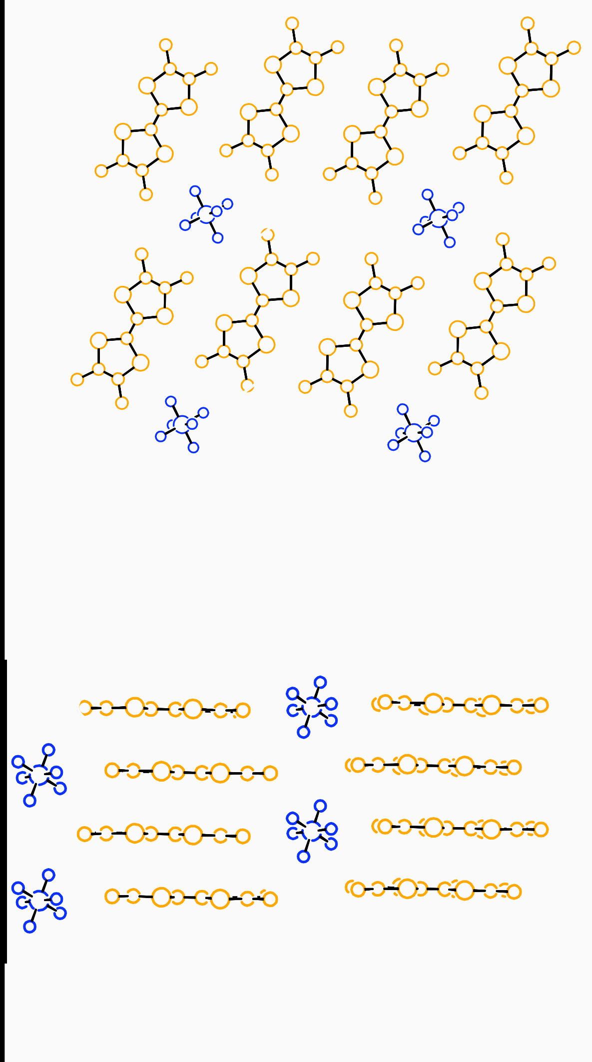
Kyoto Univ.: **M. Maesato, G. Saito**

Outline

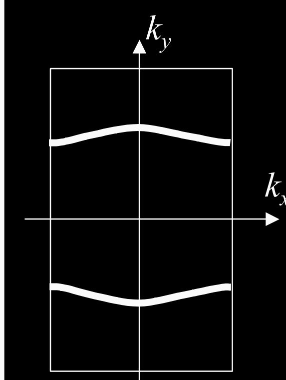
1. Introduction to organic conductors
2. Spin physics; Spin liquid vs AF ordering
3. Charge/spin interplay in Mott transition from AFI and spin liquid
4. Superconductivity and pseudogap

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

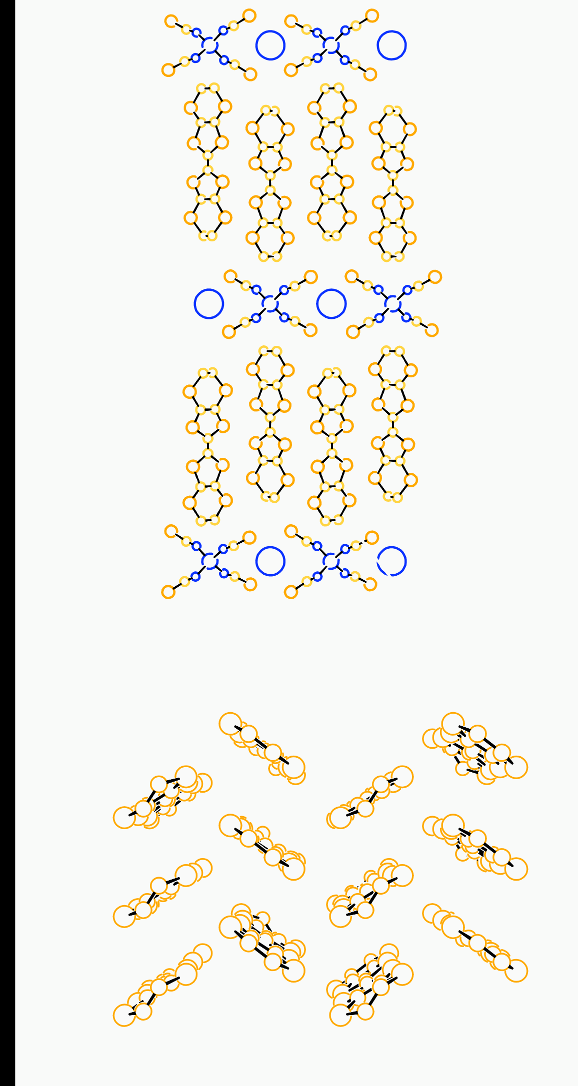
$(TMTSF)_2PF_6$



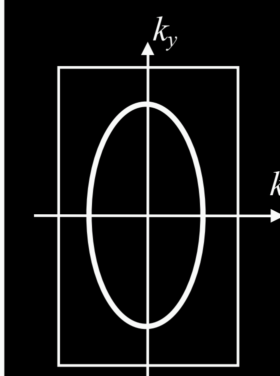
Q1D



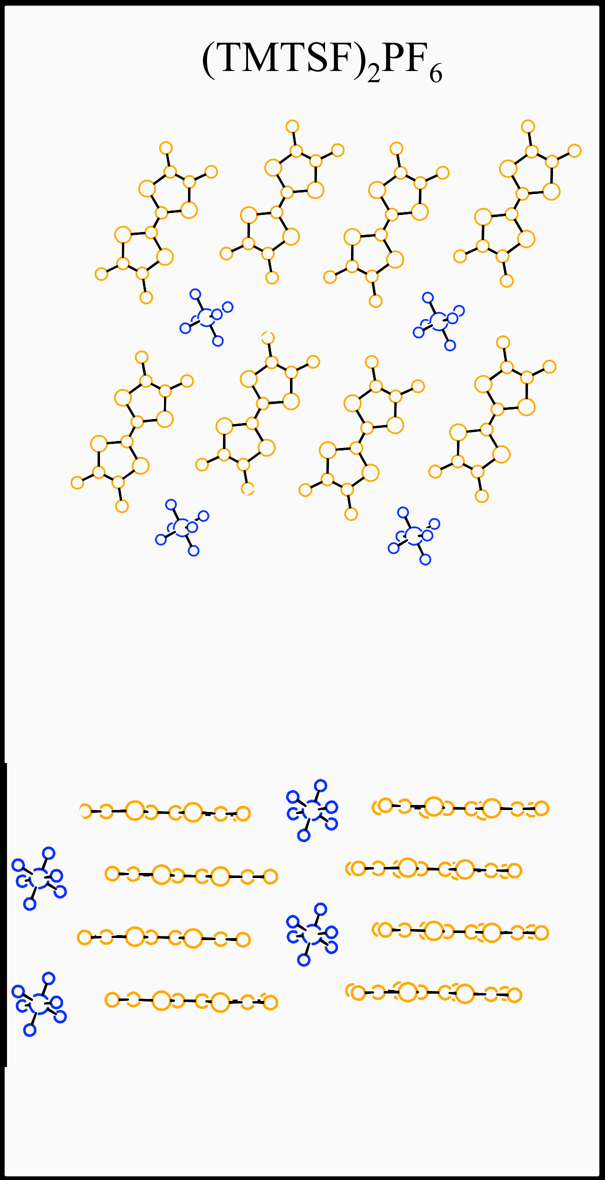
$(ET)_2RbZn(SCN)_4$

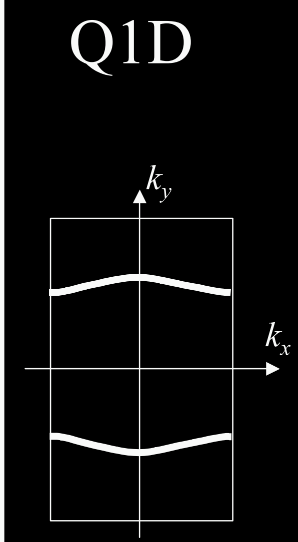


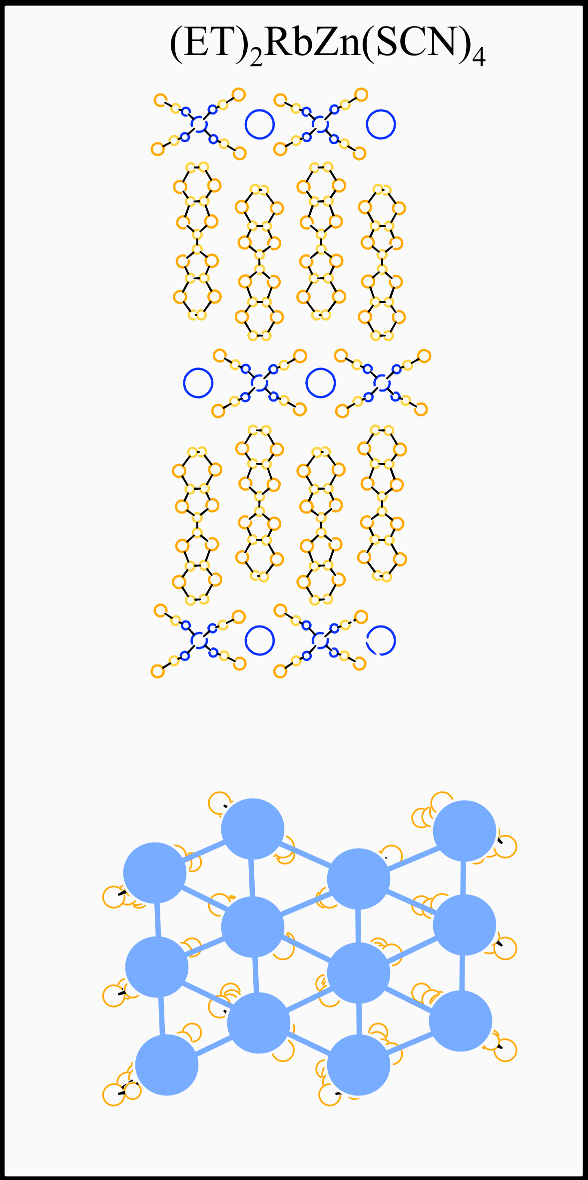
Q2D

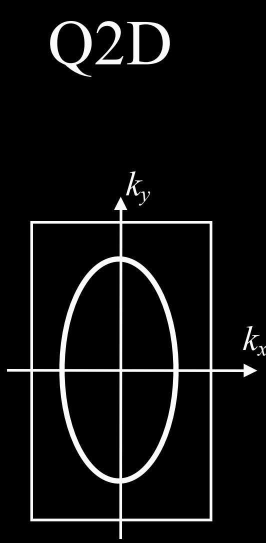


Structure: complicated in *real* space, but simple in *k*-space









Molecular conductors

Seemingly complicated structure in real space

but

Simple electronic structure in *k* space

(MO is a minimum electronic entity)

In many cases,

no orbital degeneracy

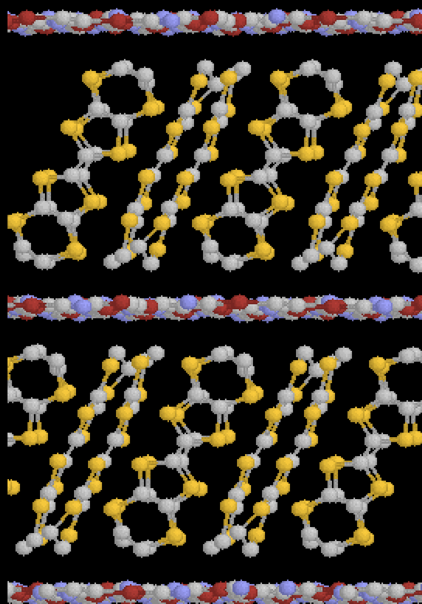
small spin-orbit interaction

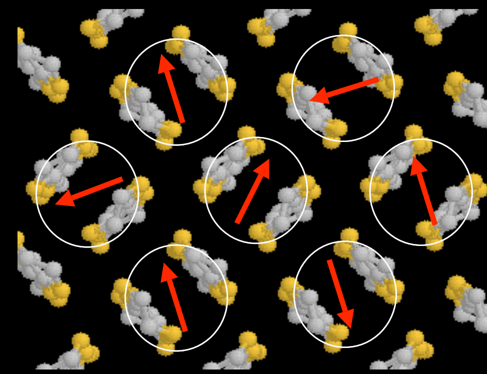
Highly compressible

Q2D organics κ-$(ET)_2X$; spin-1/2 on triangular lattice

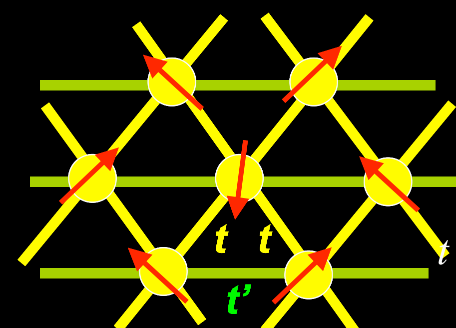
ET layer

X layer





Kino & Fukuyama dimer model



$t'/t = 0.5 \sim 1.1$

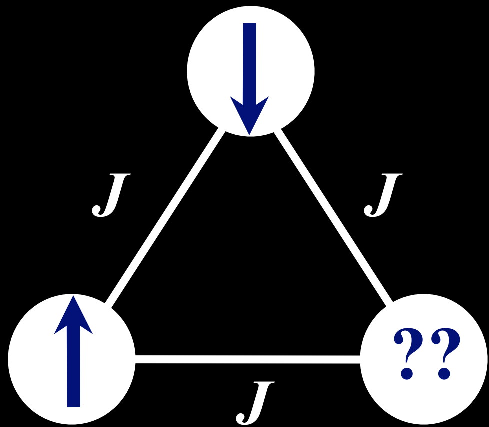
Triangular lattice
Half-filled band

X^-	Ground State	t'/t
$Cu_2(CN)_3$	Mott insulator	1.06
$Cu[N(CN)_2]Cl$	Mott insulator	0.75
$Cu[N(CN)_2]Br$	SC	0.68
$Cu(NCS)_2$	SC	0.84

Interacting electrons in κ-$(ET)_2X$ are

on triangular lattice and **near Mott transition**

Novel quantum matter



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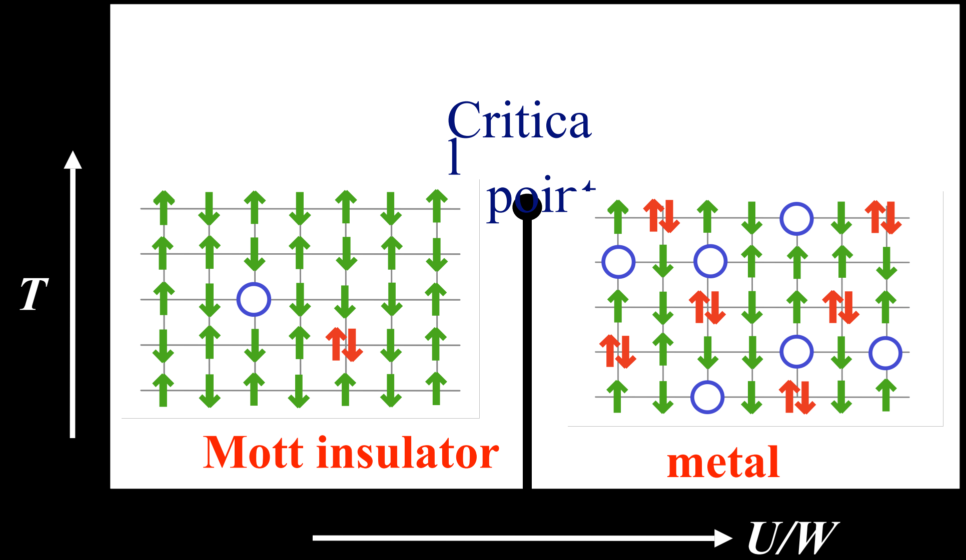
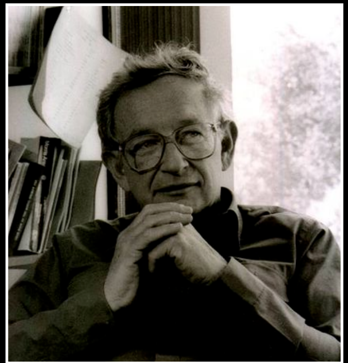


Geometrical frustration in spin degrees of freedom

Particle nature/ wave nature competition in charge degrees of freedom



P. W. Anderson

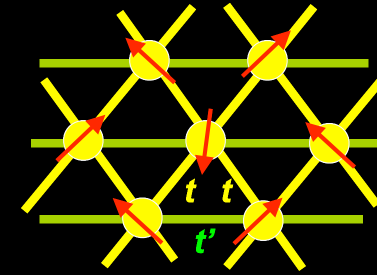
N. Mott

Ordering or quantum liquid

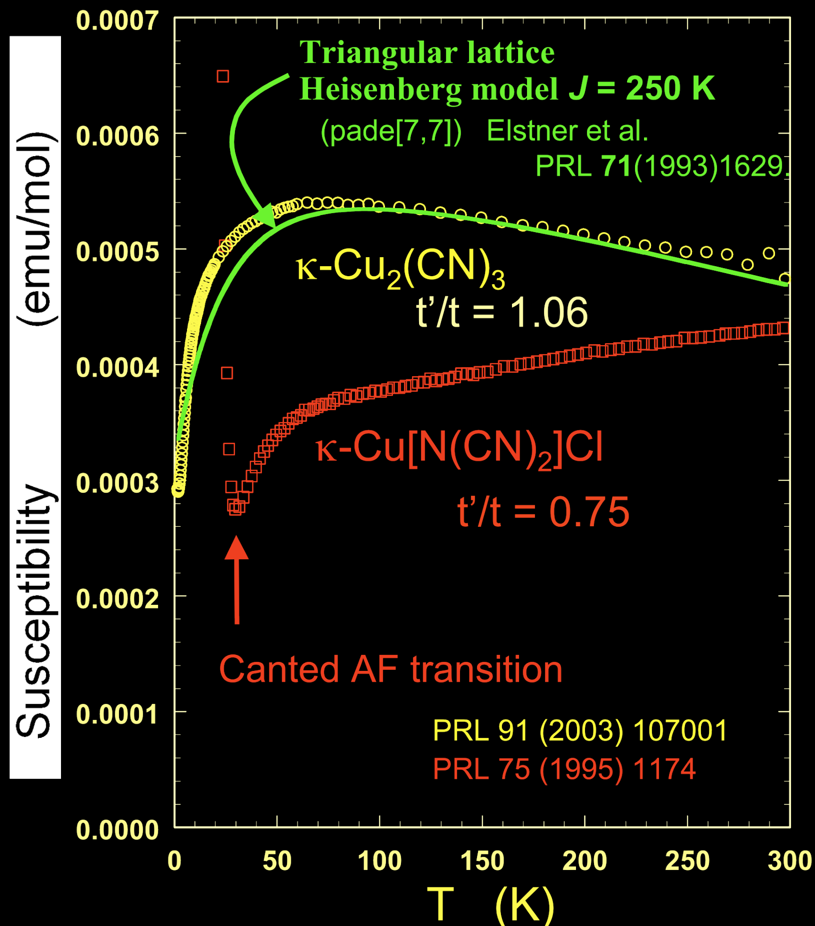
κ-(ET)$_2$X

Mott insulators

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

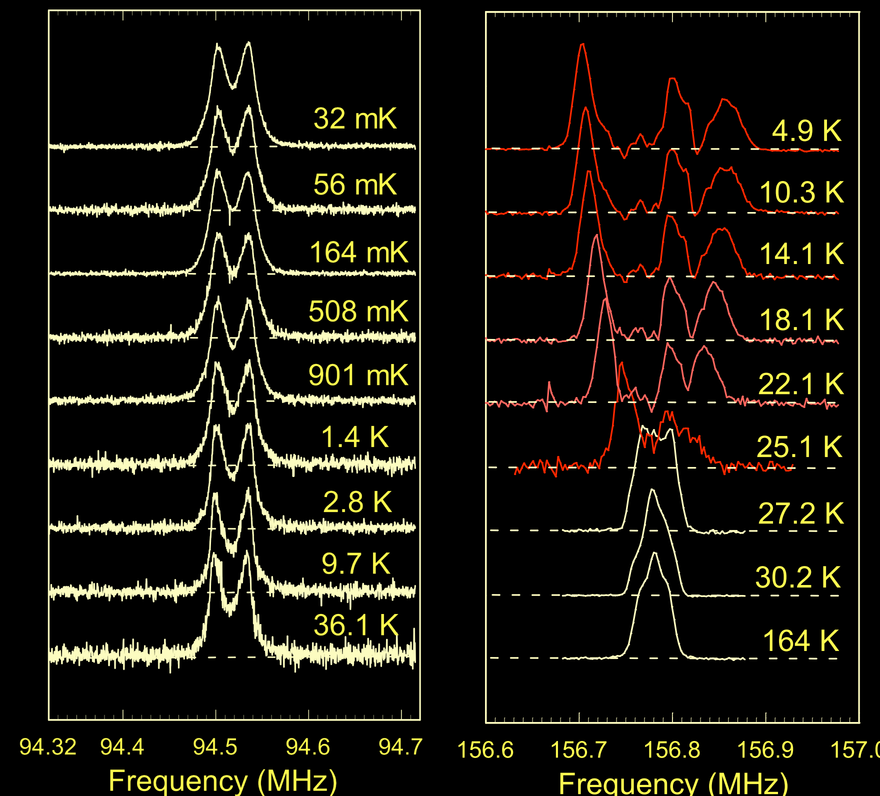


Magnetic susceptibility



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

[1]H NMR spectrum



κ-(ET)$_2$Cu$_2$(CN)$_3$ — κ-(ET)$_2$Cu[N(CN)$_2$]Cl

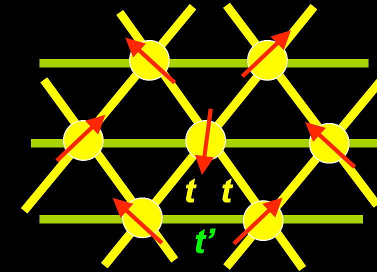
t'/t =1.06 — t'/t =0.75

No ordering ⟺ AF ordered (0.45μ_B)

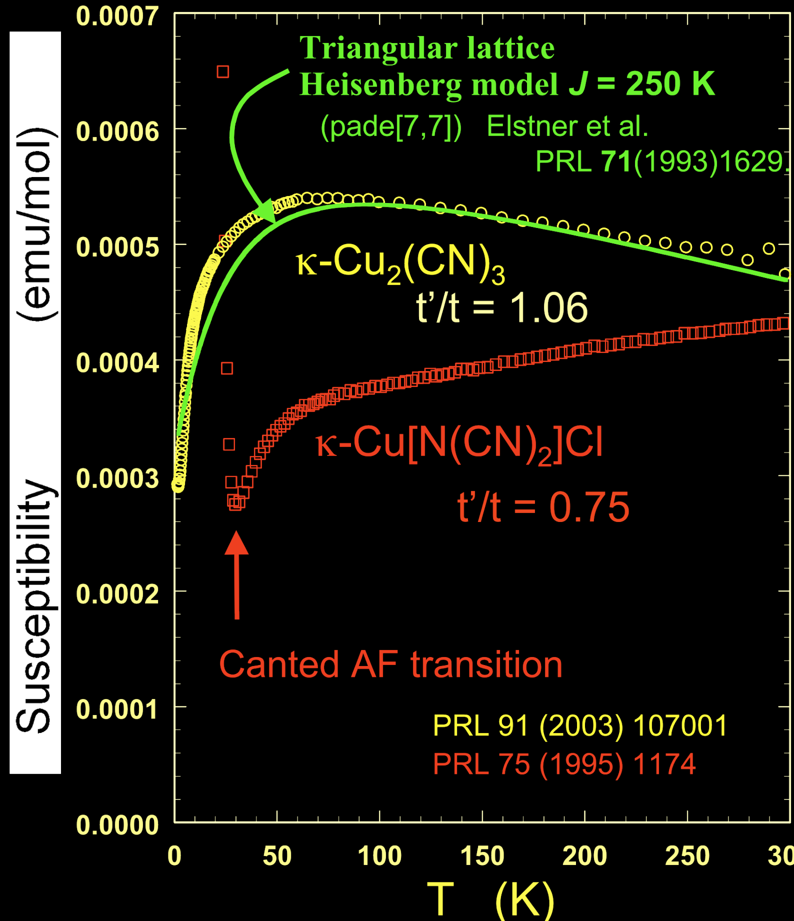
Ordering or quantum liquid

κ-$(ET)_2X$ Mott insulators

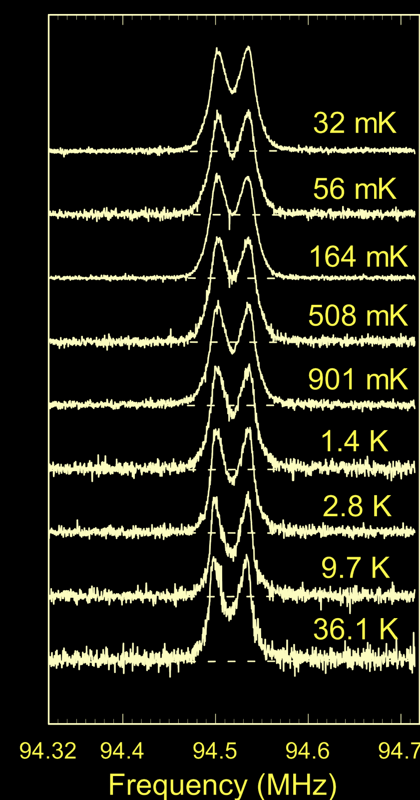
X	t'/t
$Cu_2(CN)_3$	1.06
$Cu[N(CN)_2]Cl$	0.75

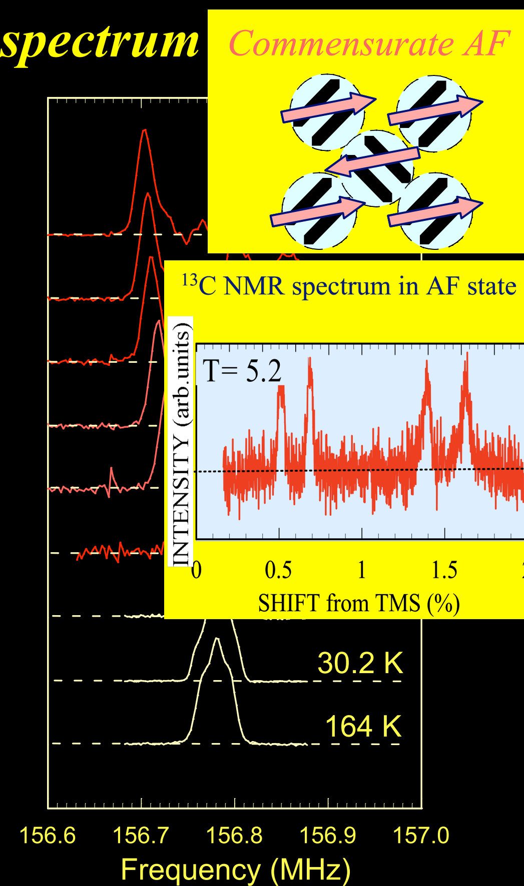


Magnetic susceptibility



^{1}H NMR spectrum





κ-$(ET)_2Cu_2(CN)_3$

$t'/t = 1.06$

No ordering ⟺

κ-$(ET)_2Cu[N(CN)_2]Cl$

$t'/t = 0.75$

AF ordered ($0.45\mu_B$)

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

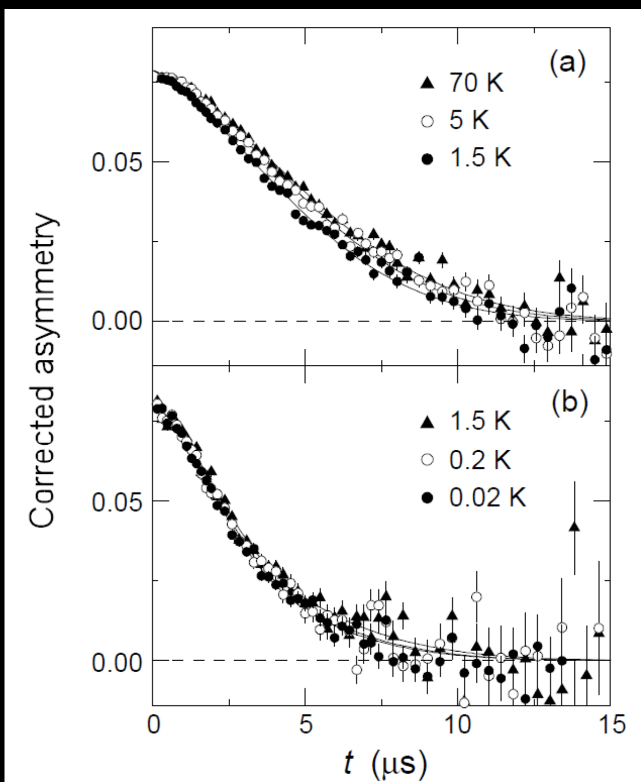
Zero-field μ*SR* of κ-$(ET)_2Cu_2(CN)_3$

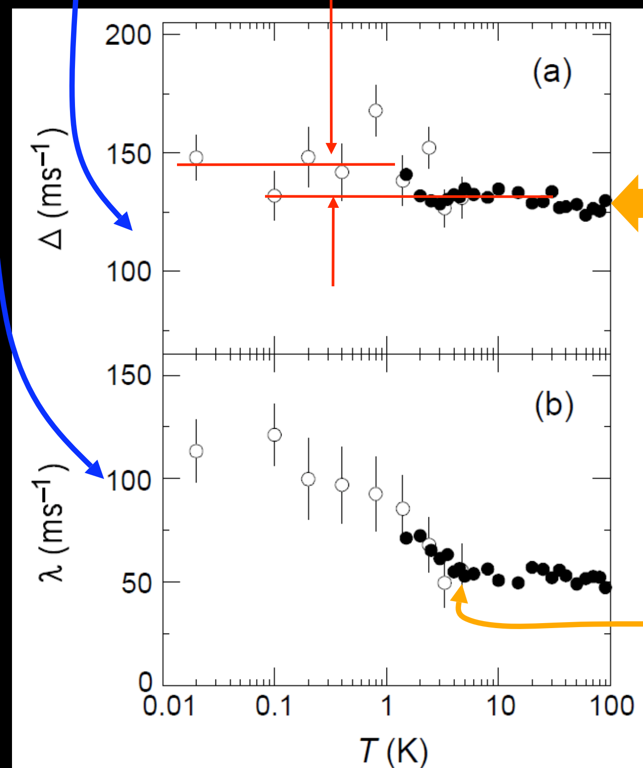
Ohira et al

Absence of long range order

$$A(t) = A\mathrm{e}^{-\lambda t}\mathrm{e}^{-(\Delta t)^2}$$

< 10^{-4} μ_B, if any





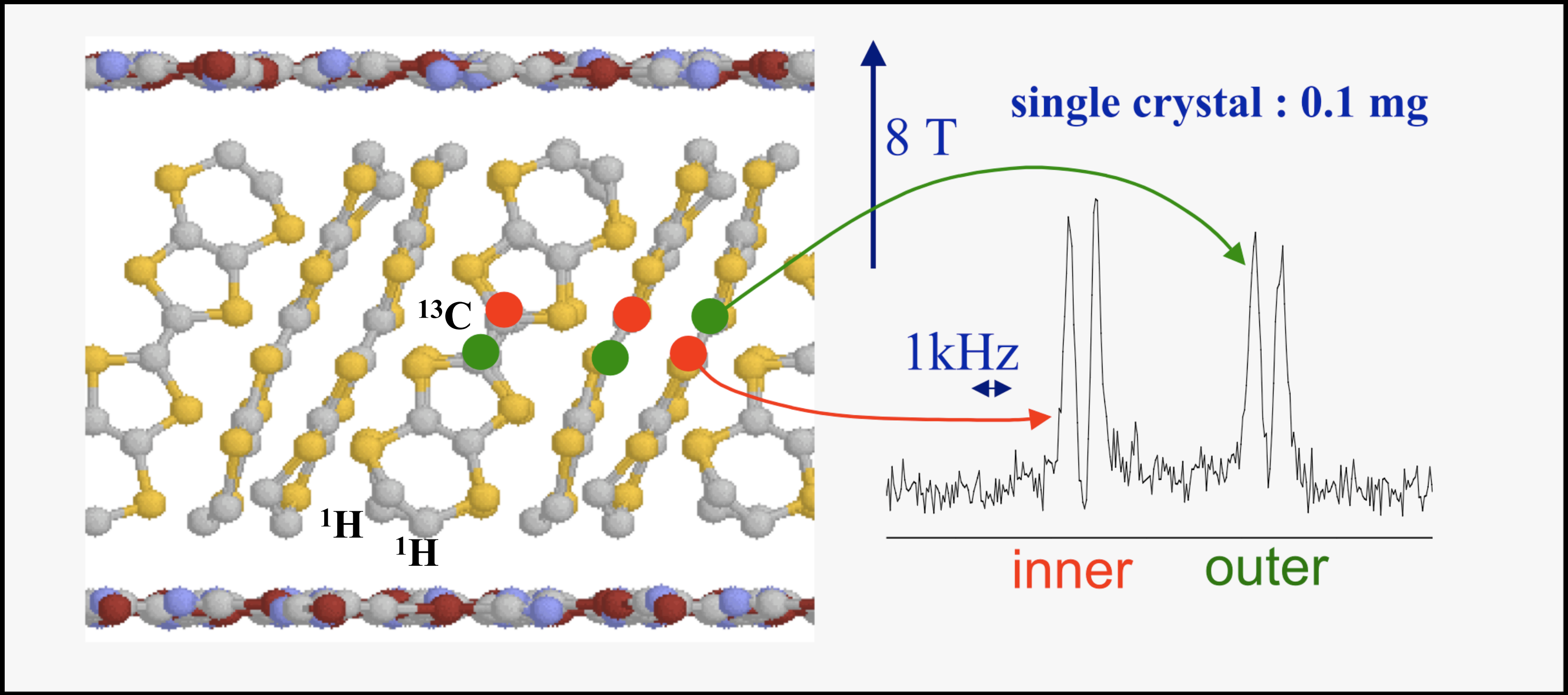
Distribution width of static field

Δ/γ_μ = 1.5 Gauss due to nuclear dipole

Relaxation rate due to dynamical fields

Anomaly around 4-5 K

^{13}C NMR ; sensitive probe to see electron spin



q-profile of spin fructuations

Kagawa (2007)

H_0 // z

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

Usually vanishing but nonzero in presence of D-M interaction

No *K*-χ proportionality

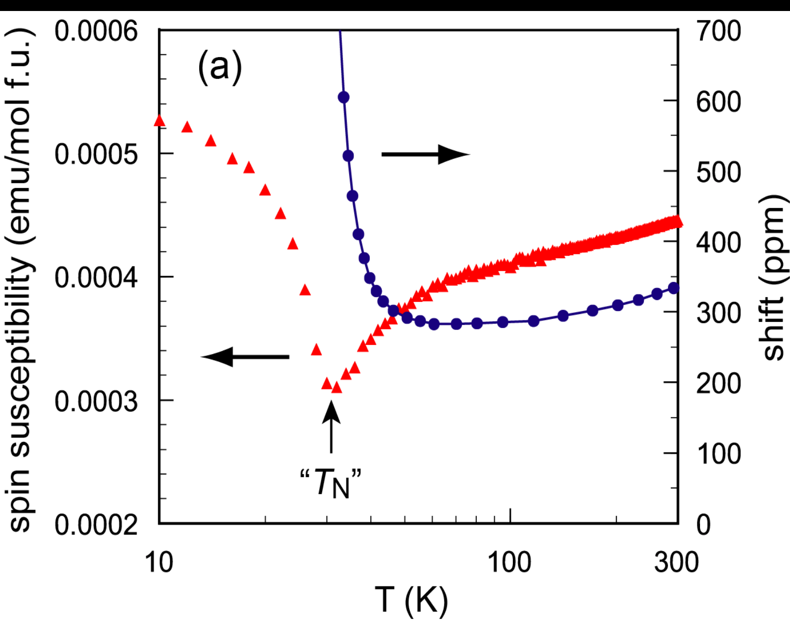
Enhanced short-range ordering at q=Q

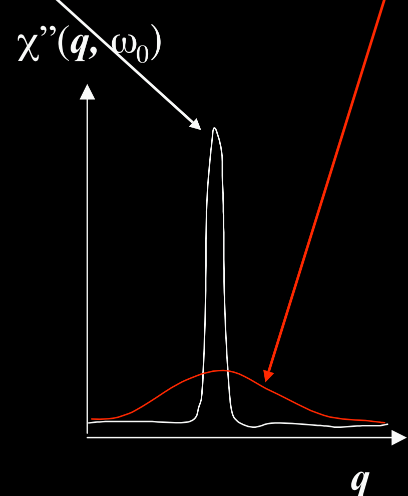
κ-$(ET)_2Cu[N(CN)_2]Cl$ (t'/t=0.75)

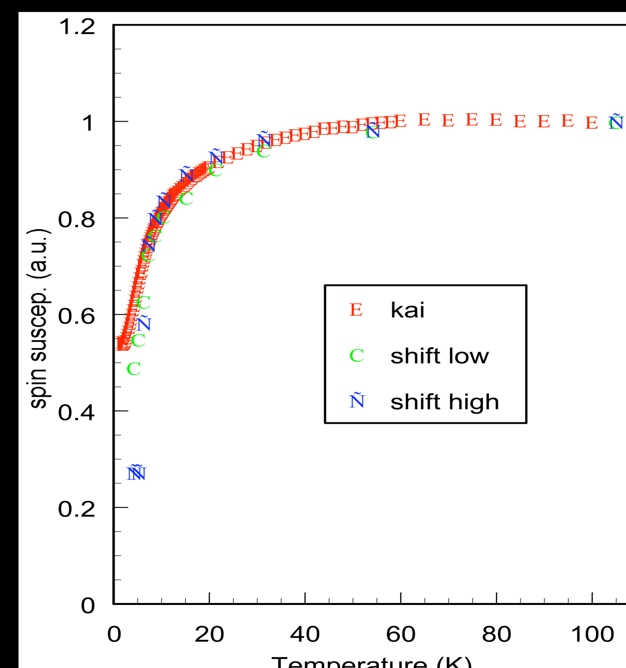
Good K-χ proportionality

Spin fluctuations in a wide **q** range

κ-$(ET)_2Cu_2(CN)_3$ (t'/t=1.06)

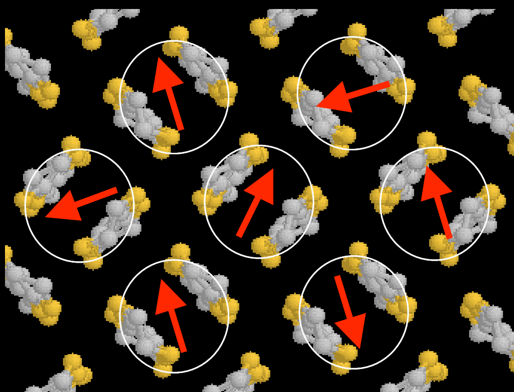






Dzyaloshinsky-Moriya interaction causes staggered magnetization under applied field

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

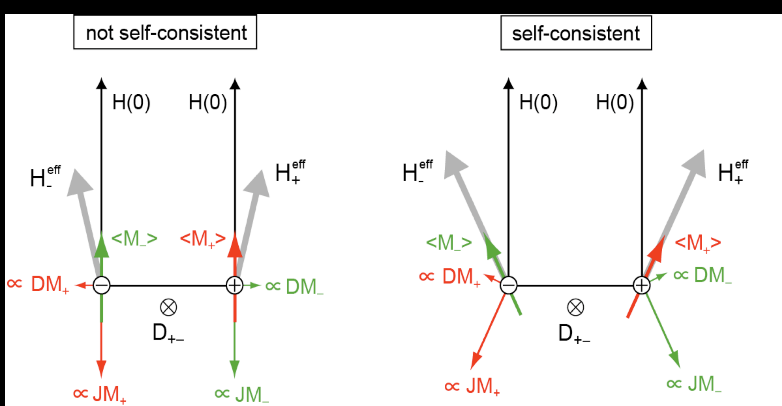


Molecular field at sublattice sites

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

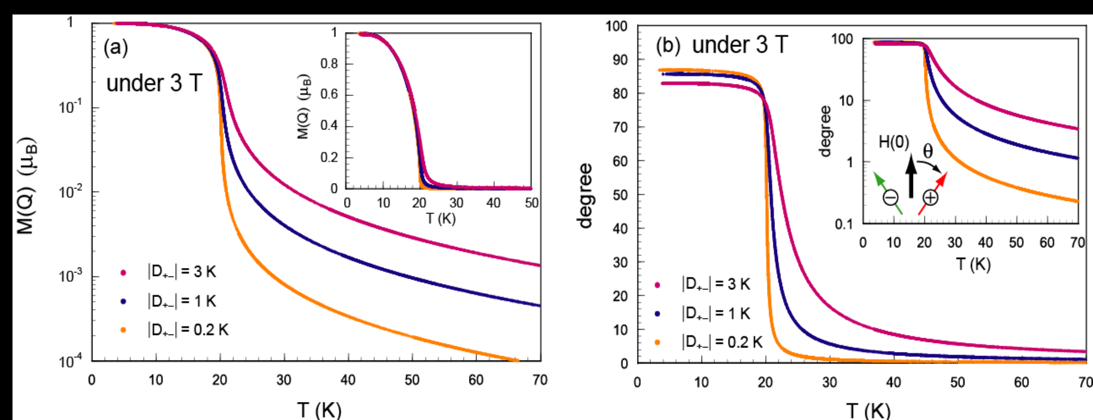
$$H_-^{eff} = -\frac{ZJ}{(g\mu_B)^2} < M_+ > - \frac{Z}{(g\mu_B)^2} D_{+-} \times < M_+ > + H_i$$

Spin configuration



Kagawa et al. (2007)

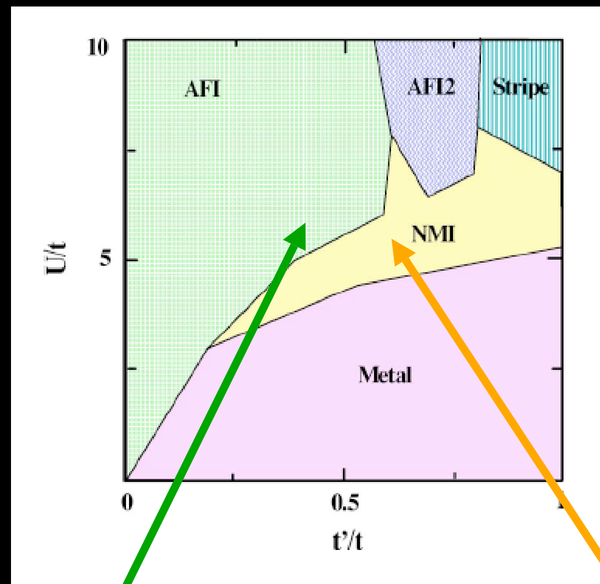
Molecular field calculation of staggered magnetization



Numerical on spin correlation

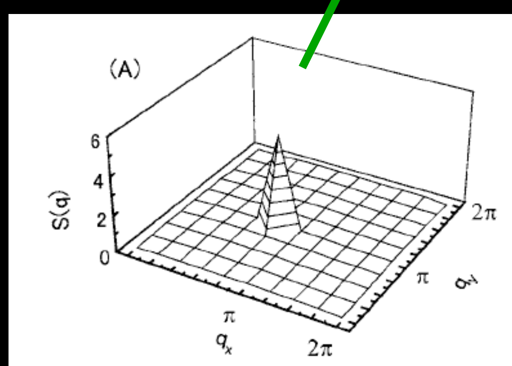
Mizusaki, Imada, PRB 74 (2006)014421

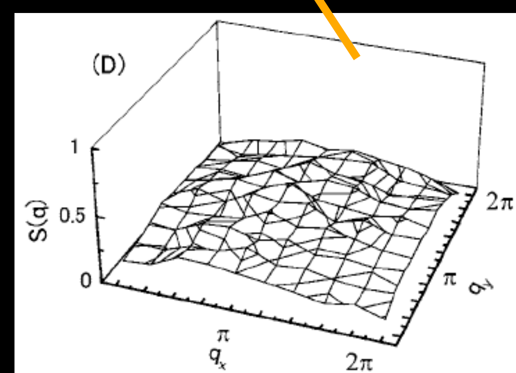
Phase diagram of Half-filled Hubbard model



AF region

Spin liquid region

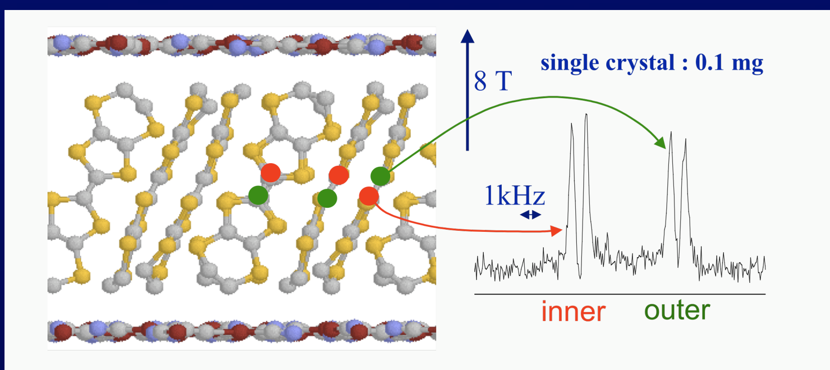


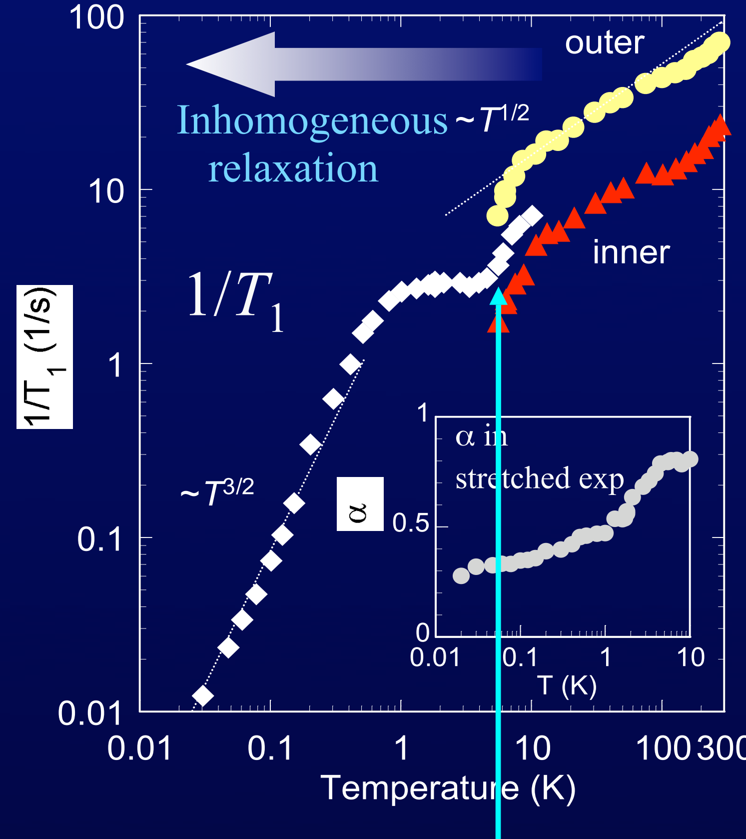


Spin excitation in κ-(ET)$_2$Cu$_2$(CN)$_3$

^{13}C NMR relaxation rate

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





$1/T_1$ ~ power law of T

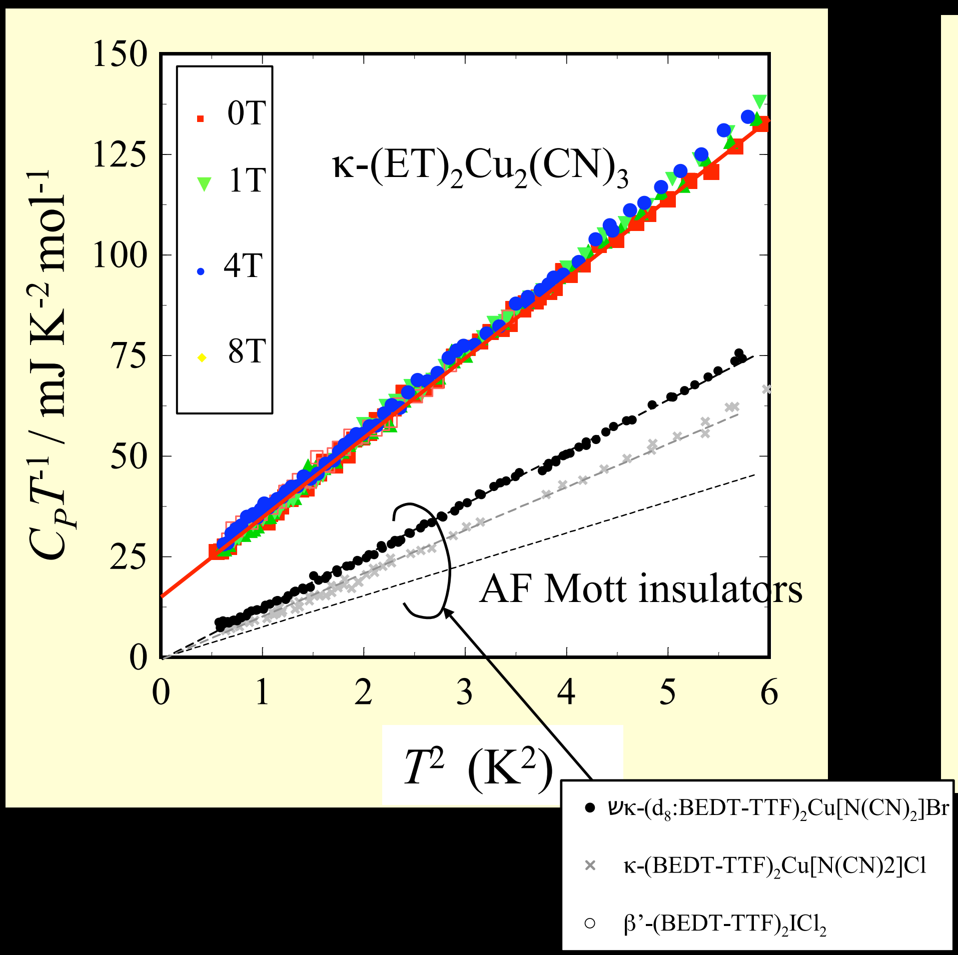
Low-lying spin excitation at low-T

Anomaly at 5-6 K

Specific heat by Yamashita and Nakazawa (Osaka Univ.)

At low temperatures

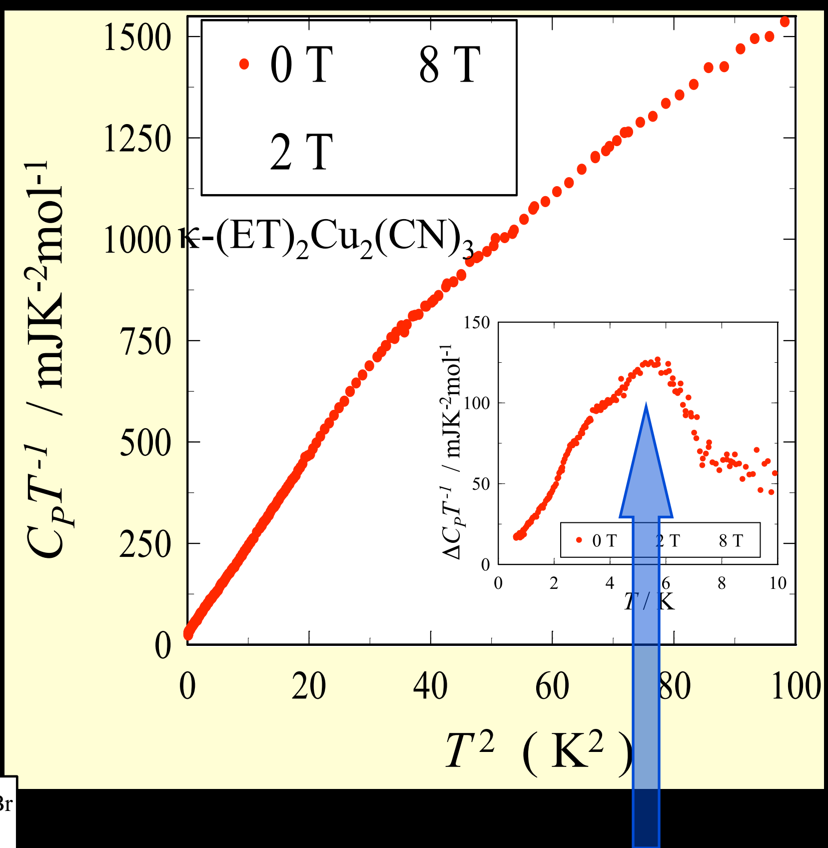
Finite γ ➡ Low-lying spin excitations



At higher temperatures;

Field-insensitive anomaly

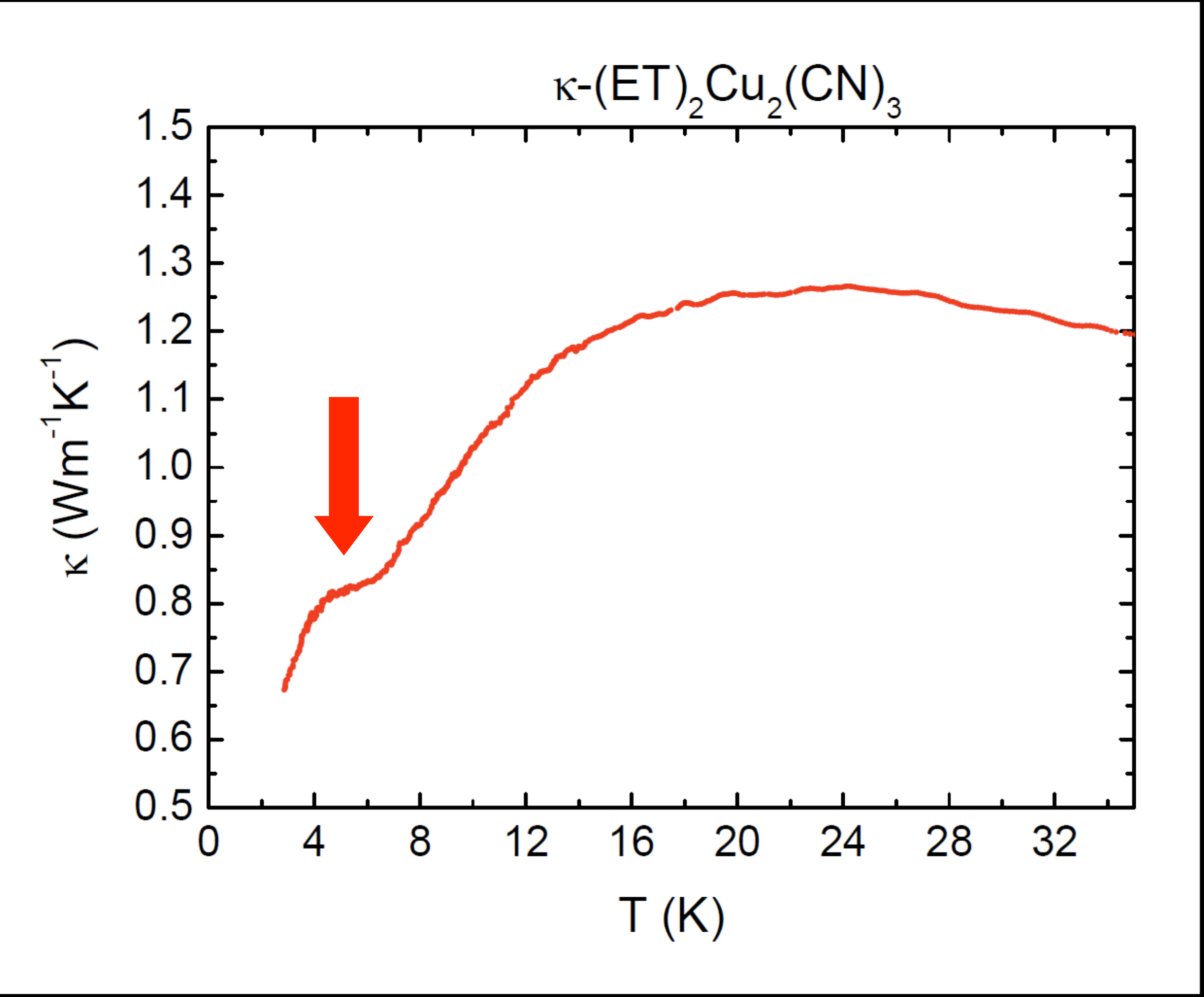
➡ Hidden order or some crossover ?

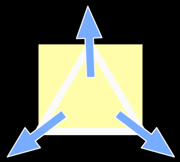


Anomaly at 5-6 K

Thermal conductivity also shows an anomaly around 5 K

Kezsmarki et al.

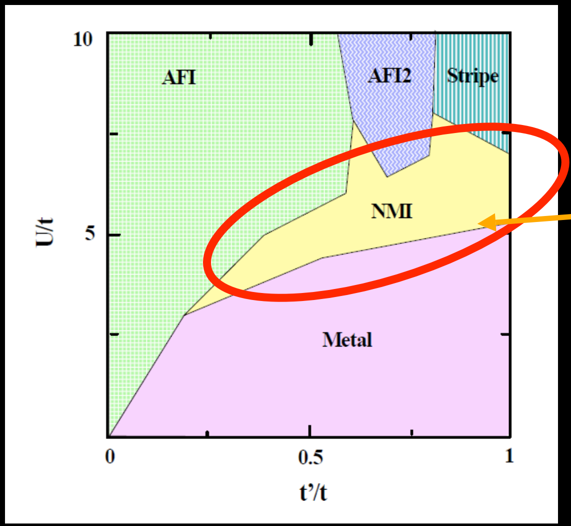


Why not  expected in Heisenberg model ?

→near the Mott transition

"Hubbard spin liquid"

Mizusaki & Imada (2006)

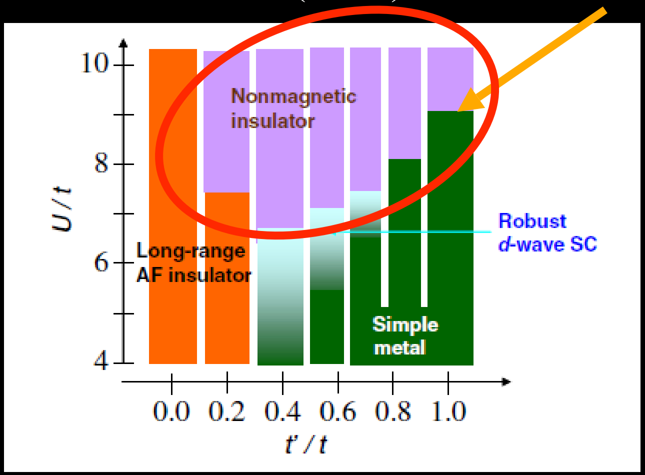


Mott transition

PIRG

Watanabe *et al*. (2006)

Mott transition



VMC

Nature of spin liquid ?

Baskaran, Mossner, Imada & Watanabe, Sorella, S.S. Lee, P.A. Lee, Senthil, Mismuich et al., Motrunich, M.P.A.Fischer, McKenzie, Schmalian, Watanabe………

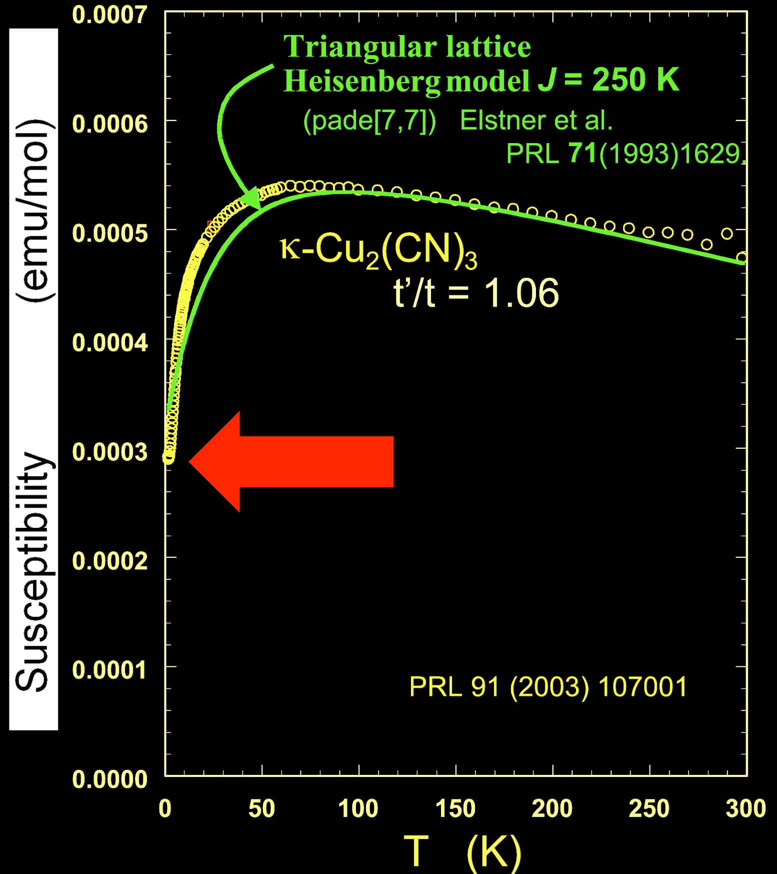
Chiral order
d-RVB
Spinon Fermi sea
Amperian pairing
…….

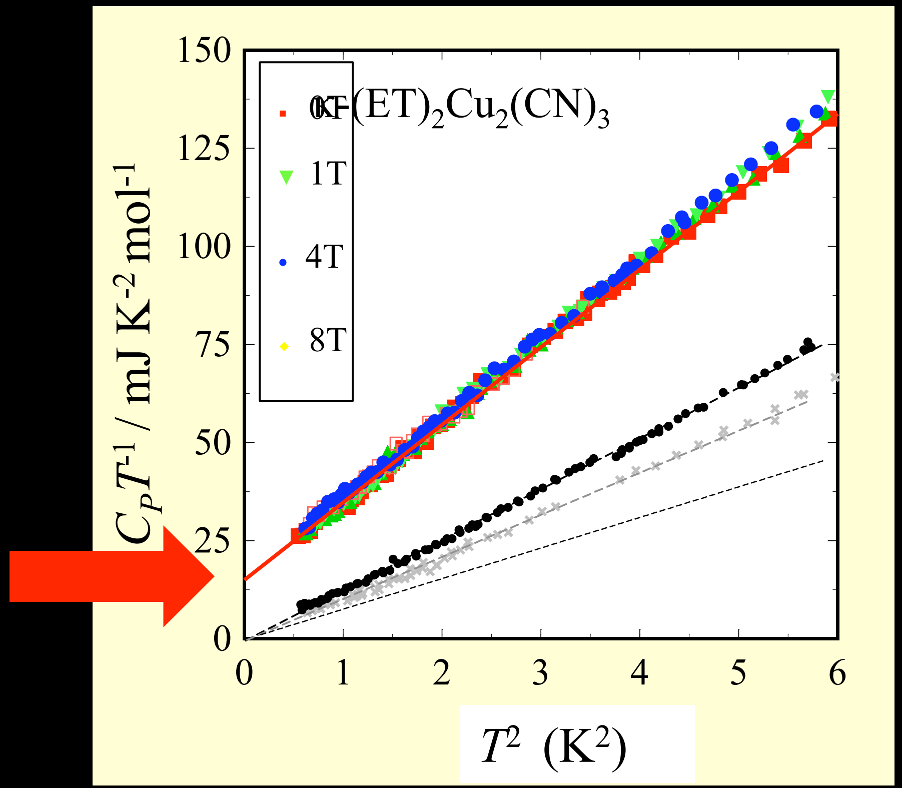
Wilson ratio ~ 1-2 !!!

⇨ Degenerate Fermionic objects in Mott insulator

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

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





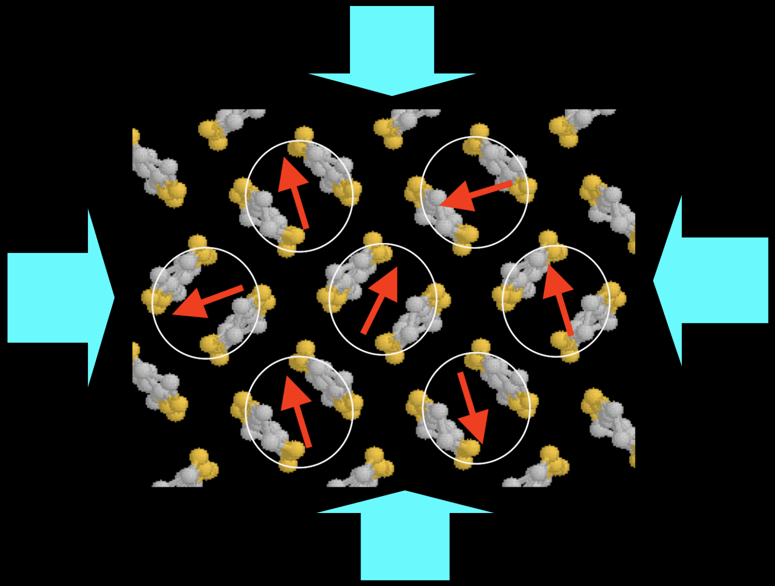
Charge degrees of freedom   MORE

Pressurize

SL insulator κ-$(ET)_2Cu_2(CN)_3$

and

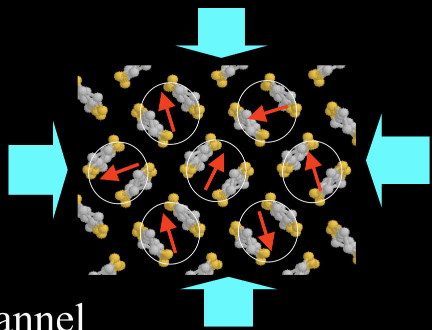
AF insulator κ-$(ET)_2Cu[N(CN)_2]Cl$



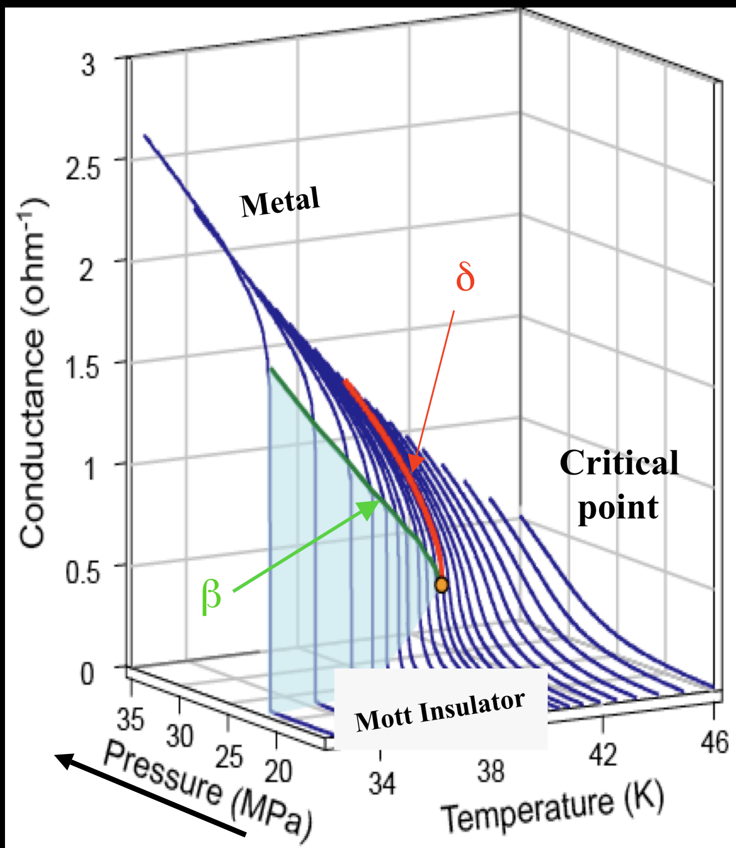
Mott transition in κ-$(ET)_2Cu[N(CN)_2]Cl$ by pressure

Kagawa *et al.*

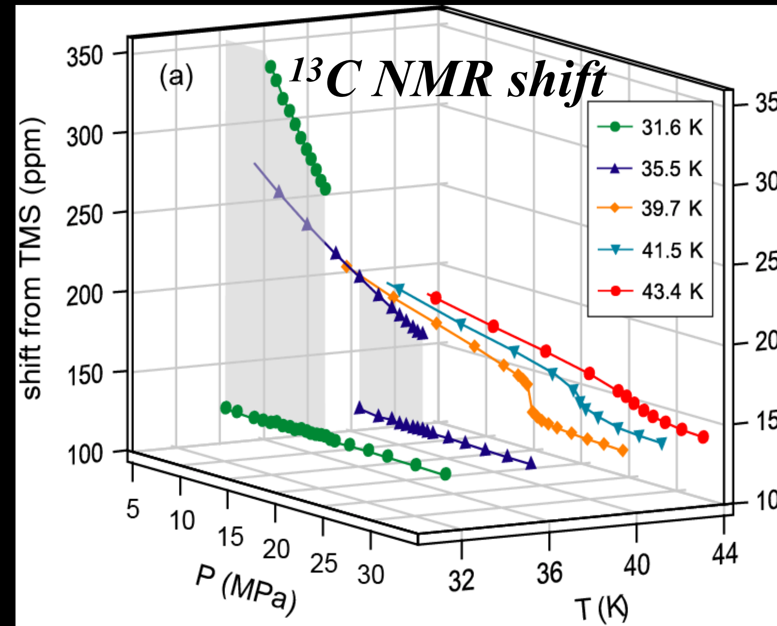
spin channel

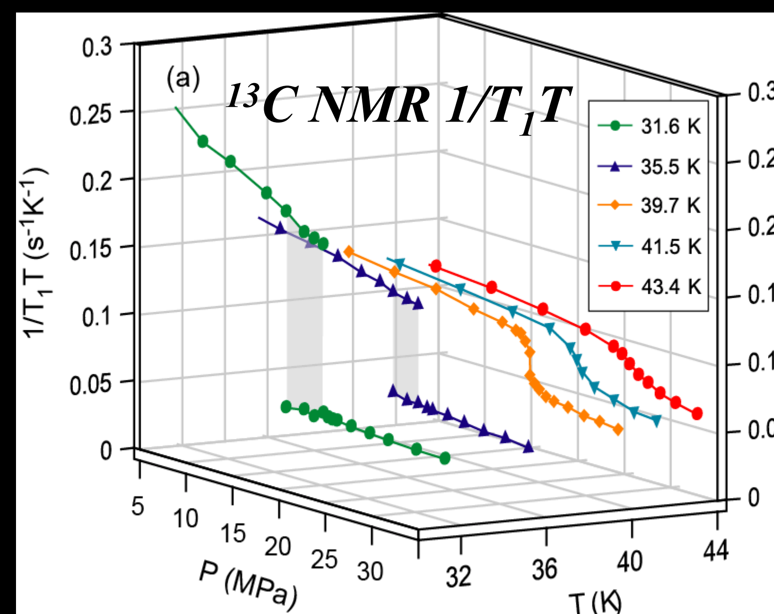


Charge channel



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

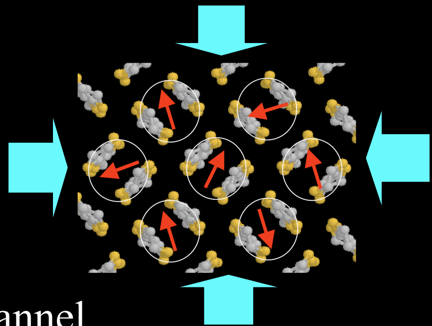




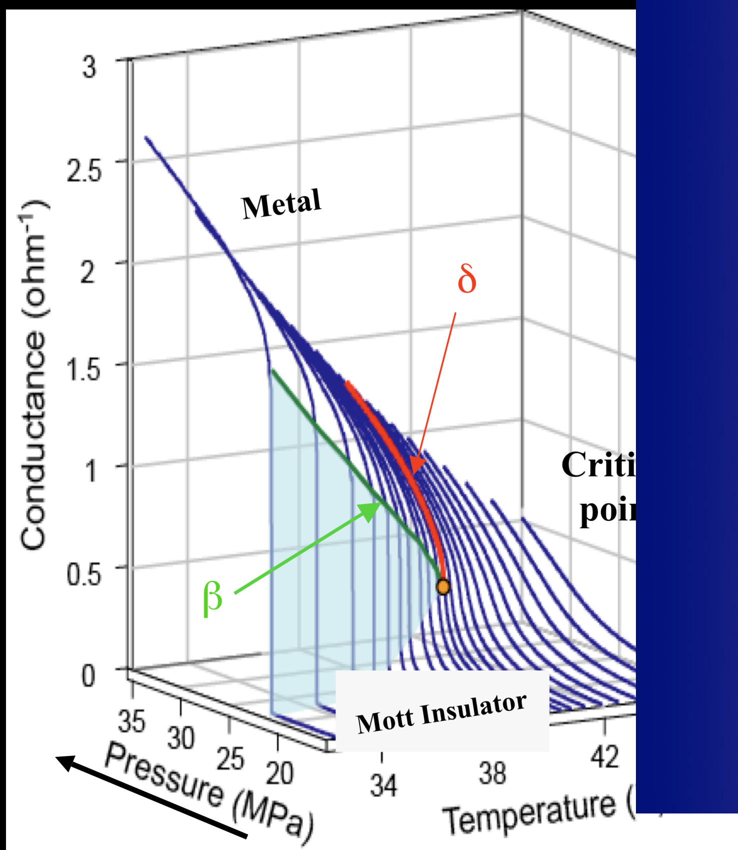
Mott transition in κ-(ET)$_2$Cu[N(CN)$_2$]Cl by pressure

Kagawa *et al.*

spin channel

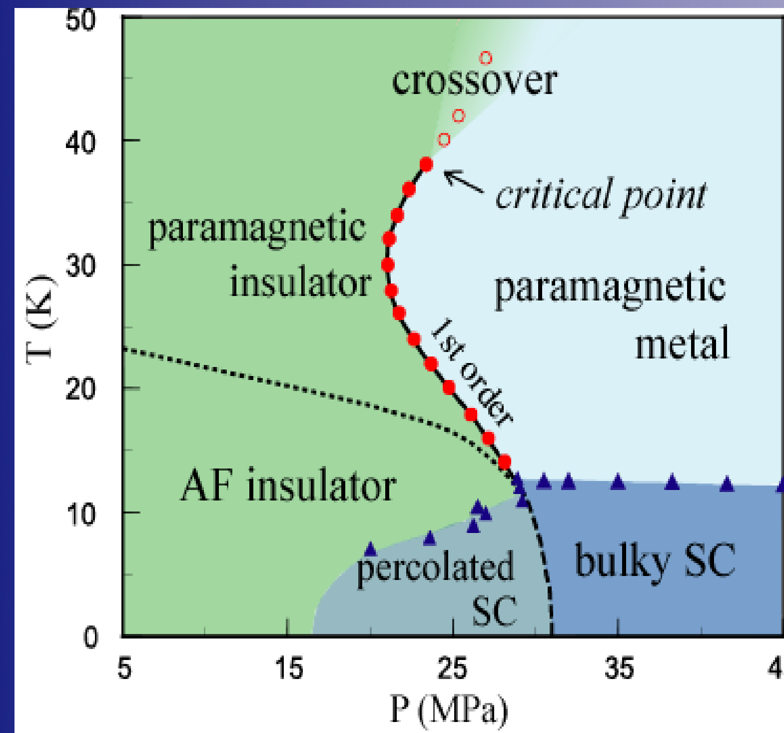


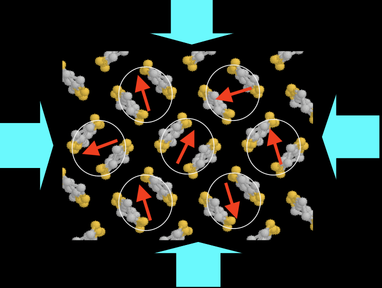
Charge channel



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

Phase diagram

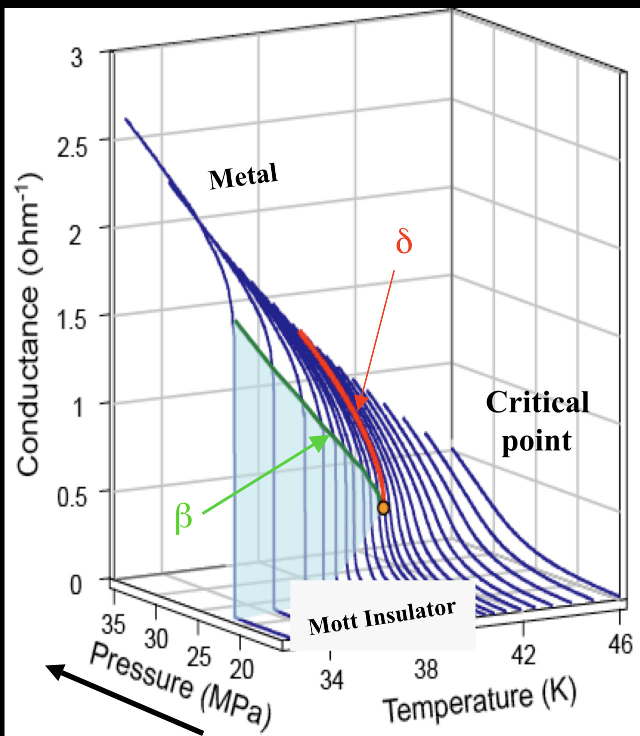




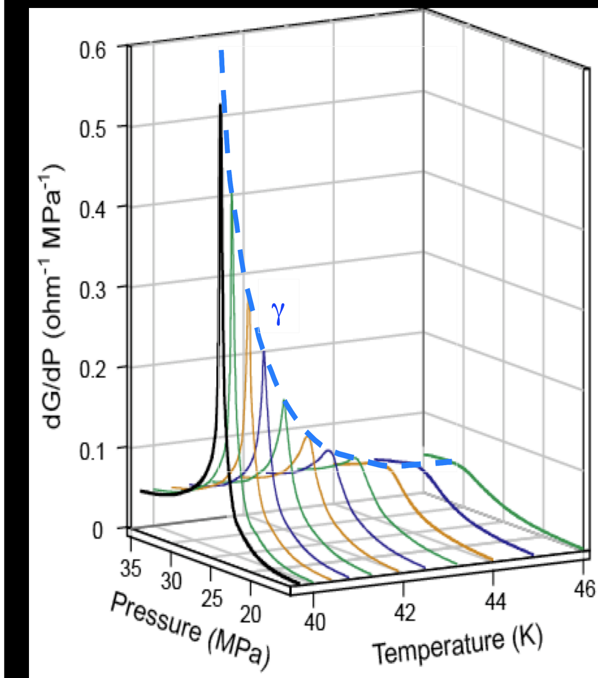
Mott Criticality and Mott scaling

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

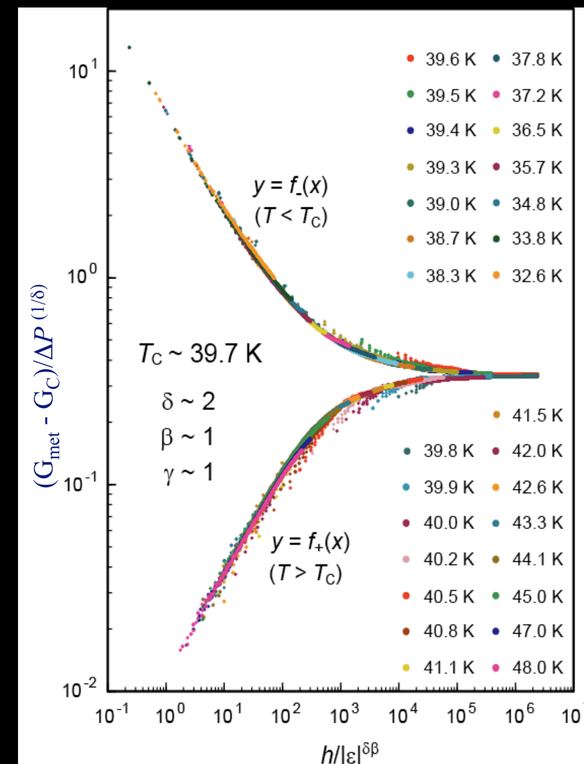
Conductance, *G*



Pressure derivative of Conductance, *dG/dP*



Scaling plot



Unconventional critical exponents

$(\delta, \beta, \gamma) \sim (2, 1, 1)$

Scaling relation is fulfilled

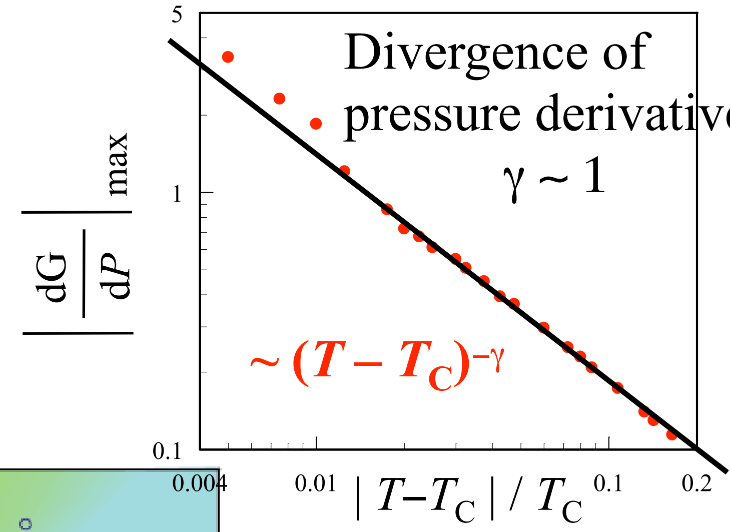
$$\delta = 1 + (\gamma / \beta)$$

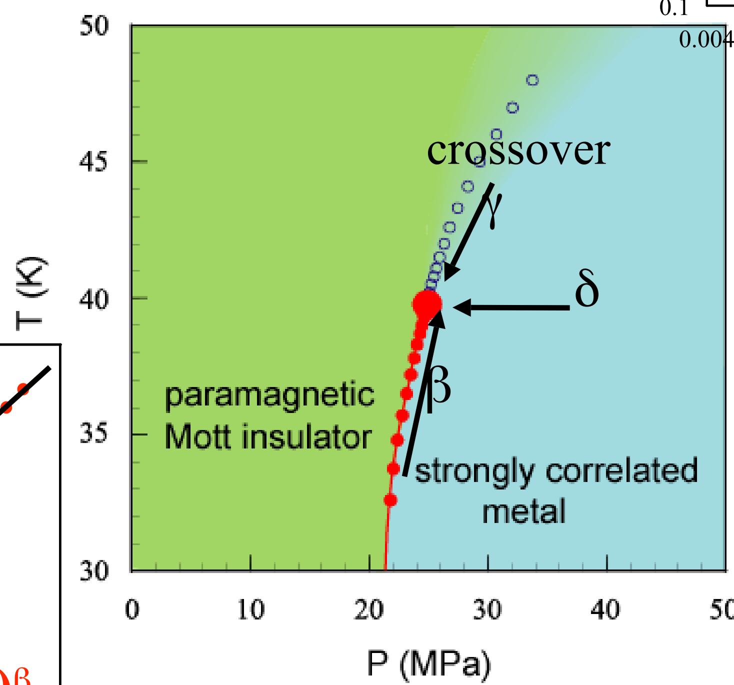
Scaling function

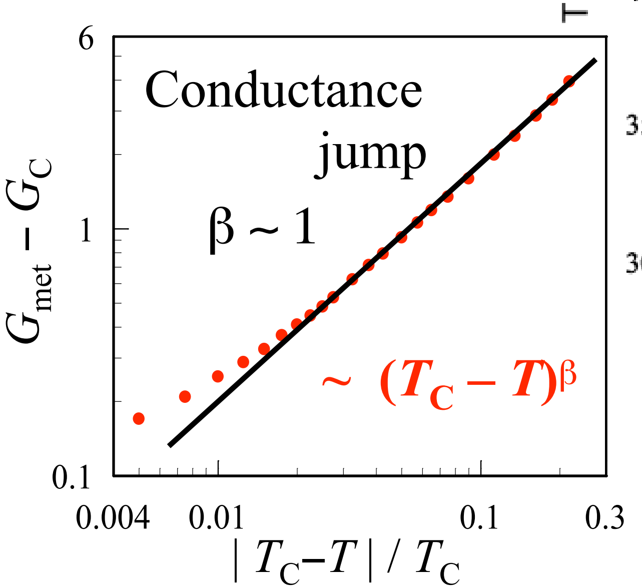
$$G_{met}(P, T) - G_C = (\Delta P)^{1/\delta} f_{\pm}\left(\frac{\Delta P}{|\Delta T|^{\beta\gamma}}\right)$$

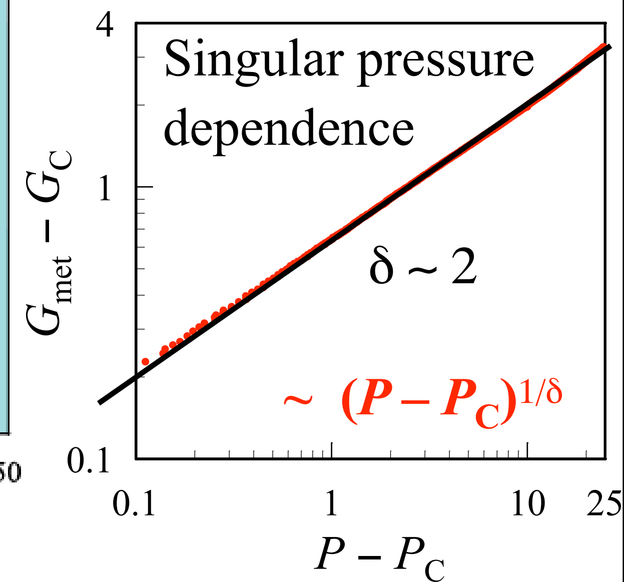
Unconventional critical exponents

(δ, β, γ) ~ (2, 1, 1)

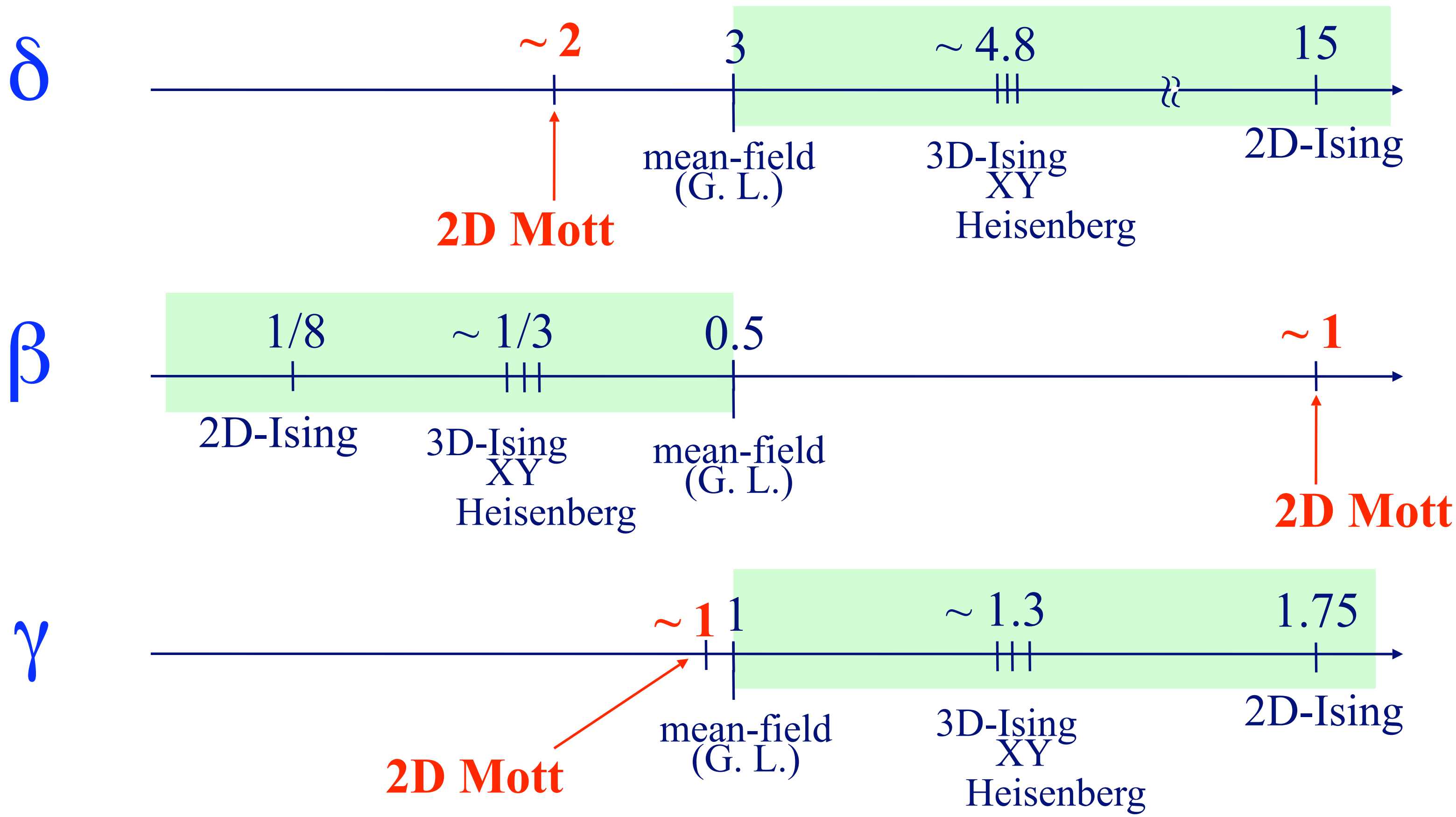








Anomalous exponents (δ, β, γ) ~ (2, 1, 1)



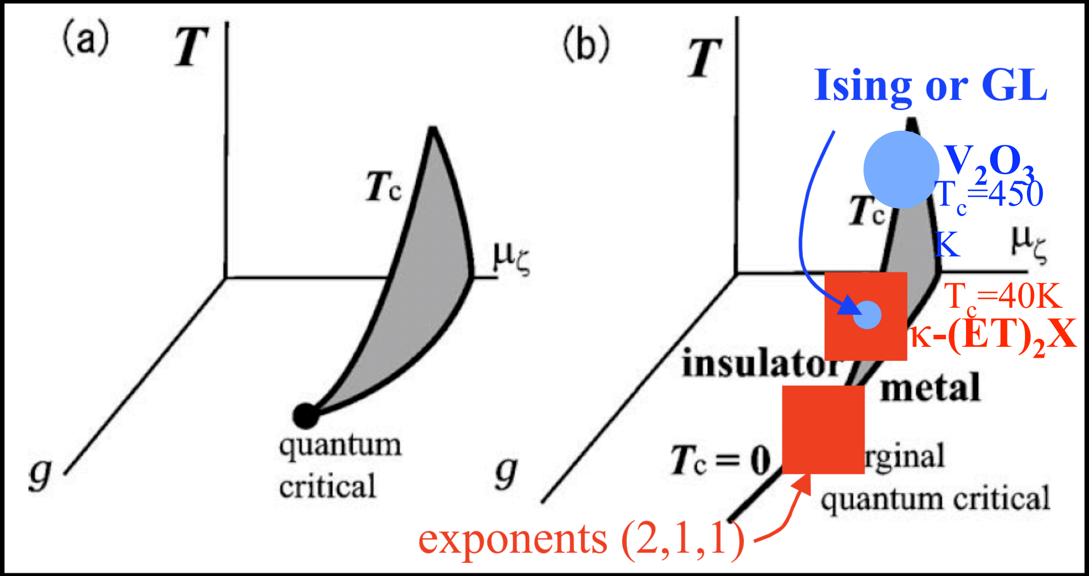
Quantum critical nature; Imada, Misawa

Universality class of quantum Mott transition; $(\delta, \beta, \gamma)=(2,1,1)$?

Theoretical

Filling-controlled Mott; Furukawa , Imada (1991, 1994)

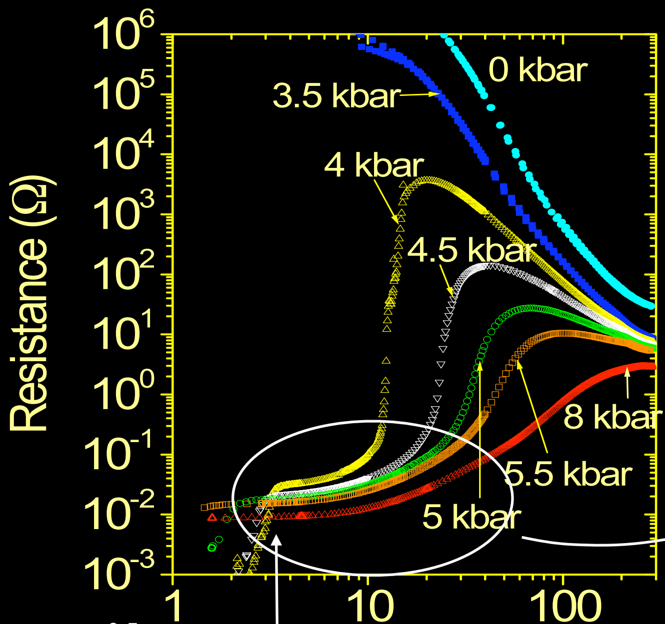
Band-width-controlled Mott; Imada, Misawa, Yamaji (2004,2006)

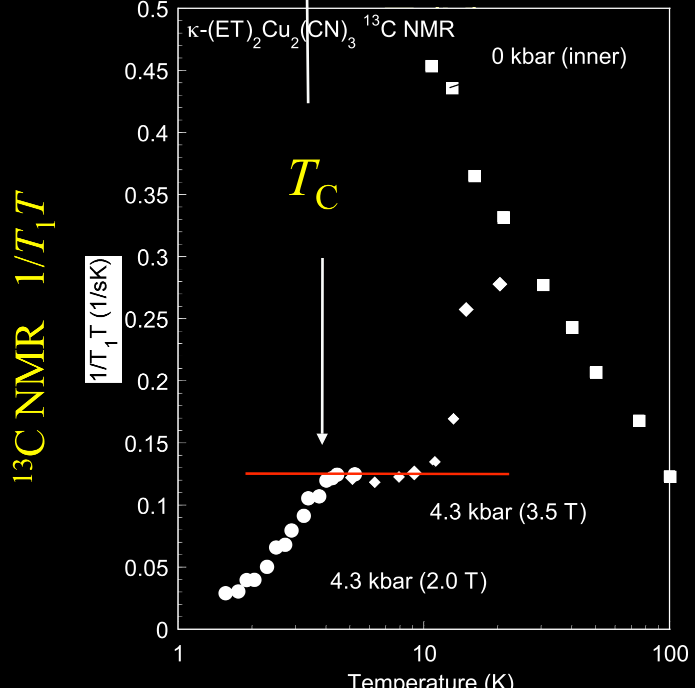


A possible involvement of quantum fluctuations at low but finite temperatures

Mott transition in κ-$(ET)_2Cu_2(CN)_3$ by pressure

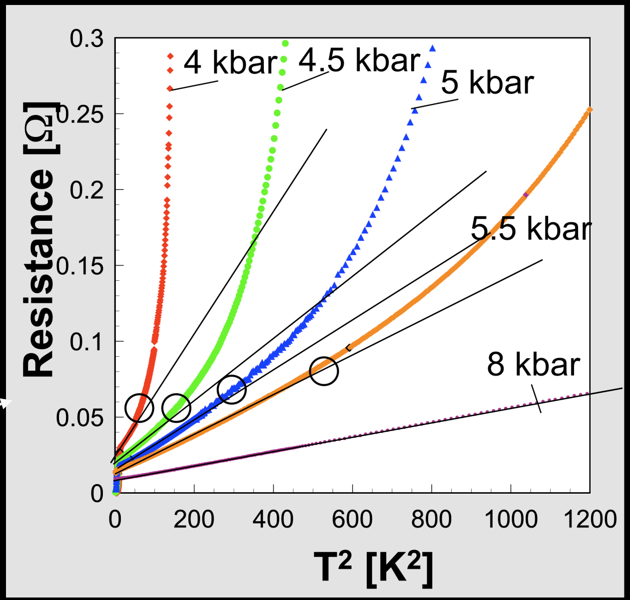
Kurosaki et al.; PRL 95 (2005) 177001; Komatsu et al. JPSJ (1996)





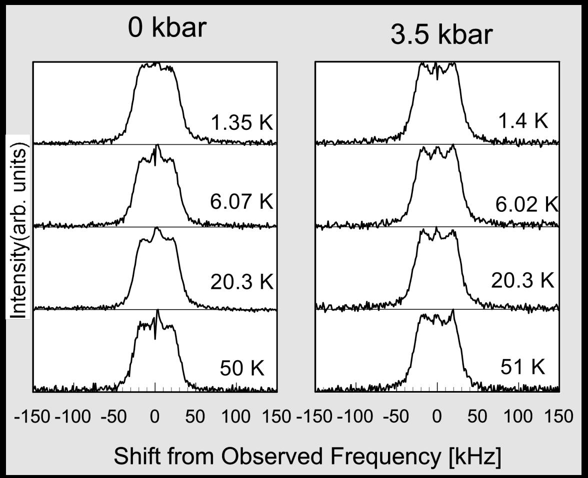
Fermi liquid

R vs T^2



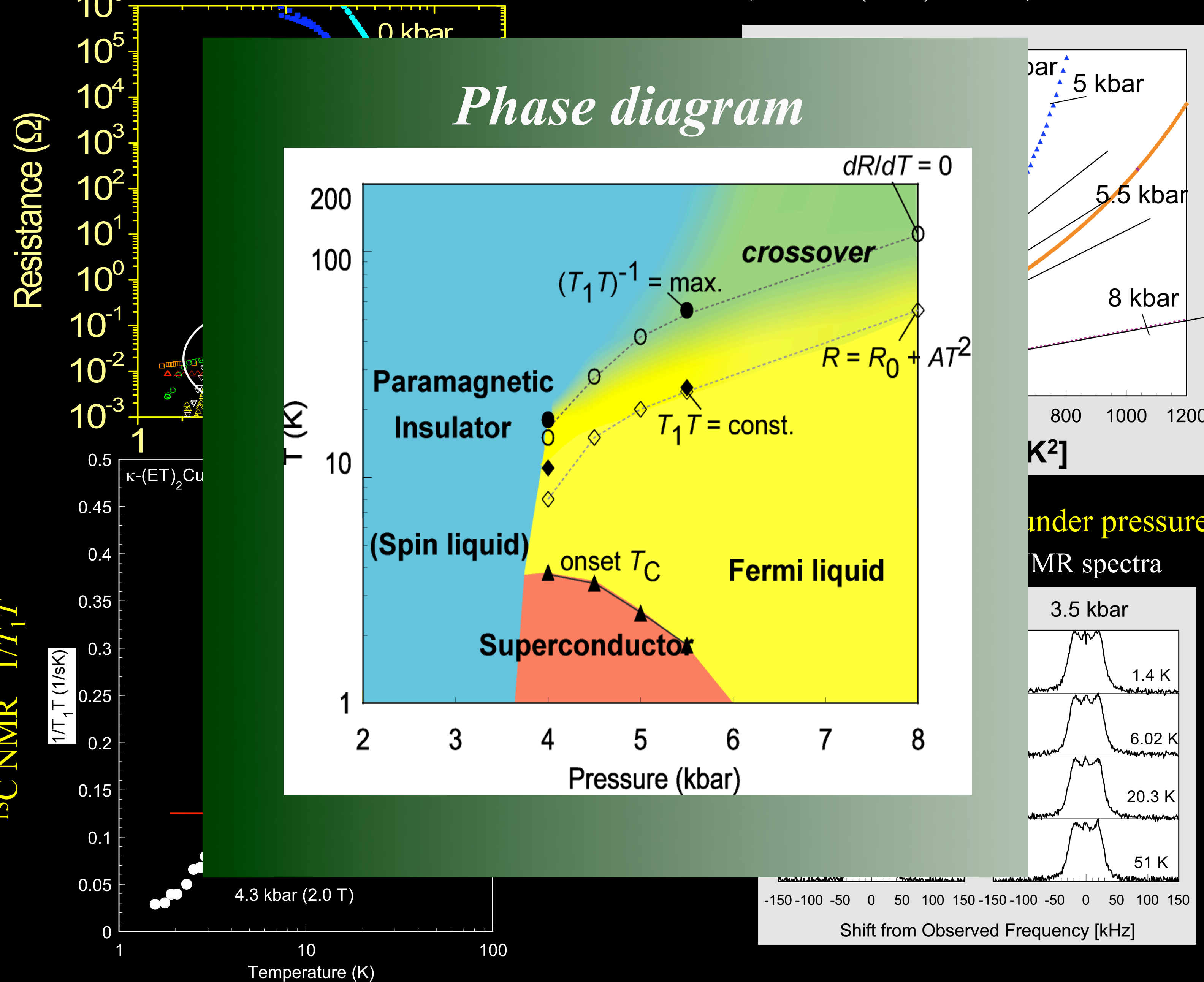
No magnetic ordering under pressure

No change in ^{1}H NMR spectra



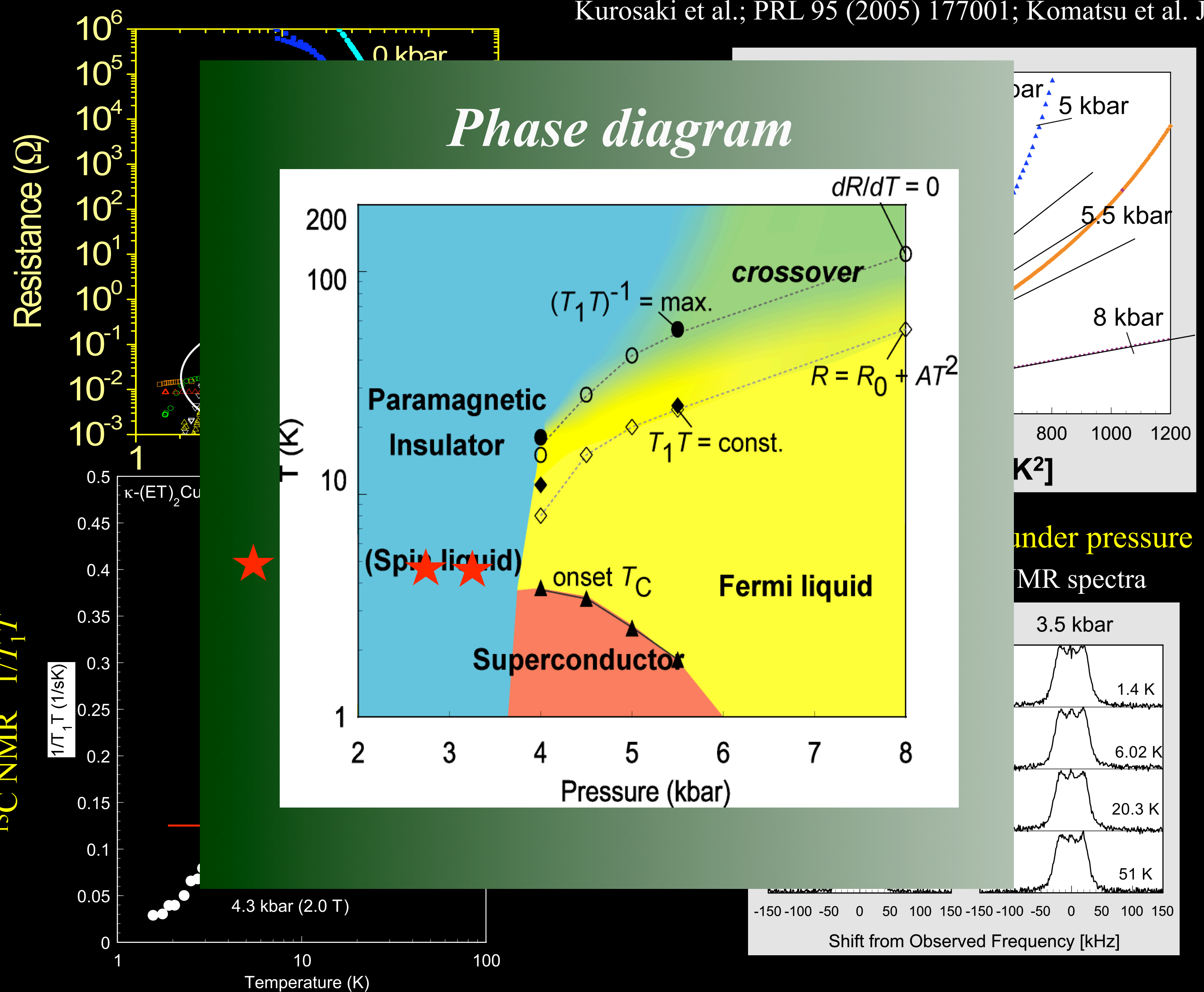
Mott transition in κ-$(ET)_2Cu_2(CN)_3$ by pressure

Kurosaki et al.; PRL 95 (2005) 177001; Komatsu et al. JPSJ (1996)



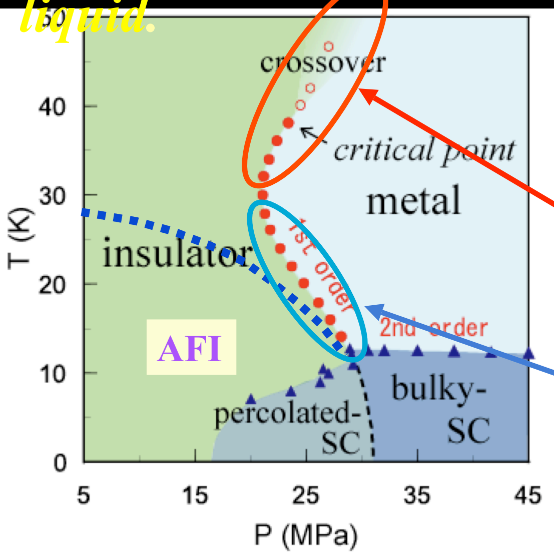
Mott transition in κ-$(ET)_2Cu_2(CN)_3$ by pressure

Kurosaki et al.; PRL 95 (2005) 177001; Komatsu et al. JPSJ (1996)

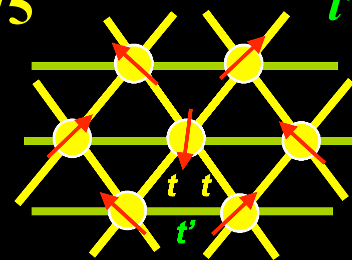


Antiferromagnet vs spin liquid ; charge gap

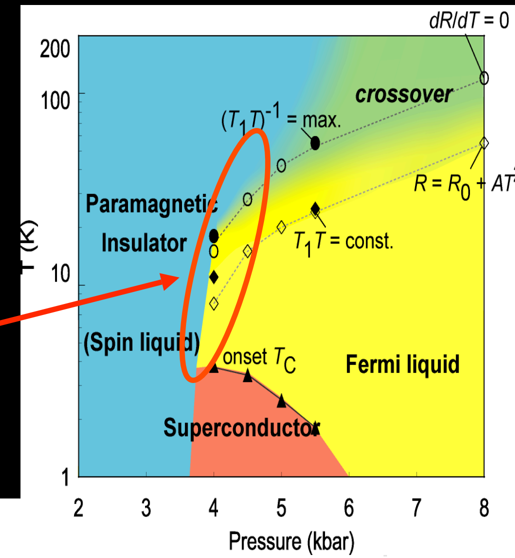
Charge gap is clearly opened on AF ordering, but remains undeveloped in spin liquid.



$t'/t = 0.75$



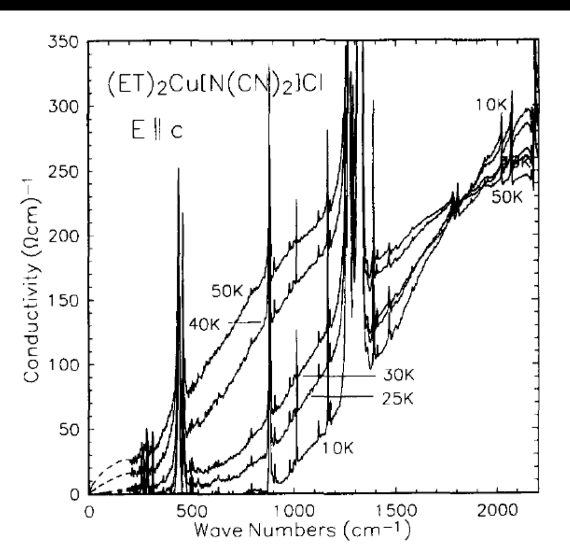
$t'/t = 1.06$



Clausius Clapeyron $dT/dP = \Delta V/\Delta S$

> 0

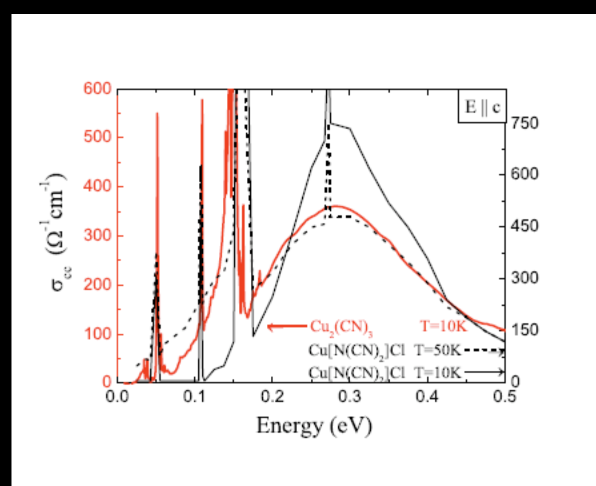
< 0

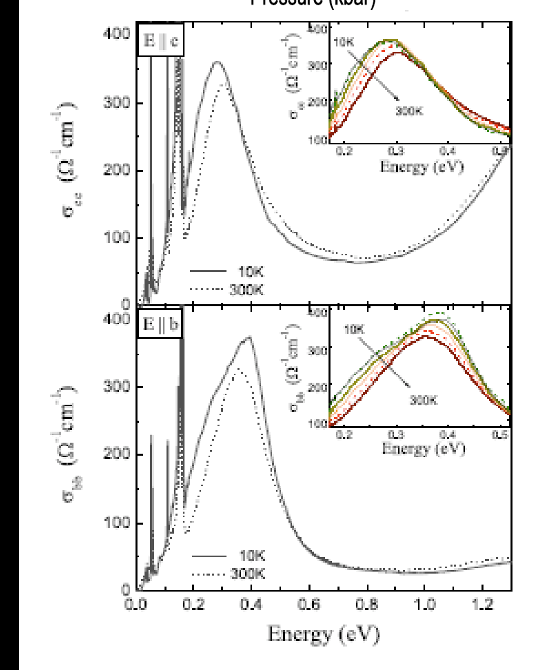


Kornelsen et al., SSC 81 (1992)343

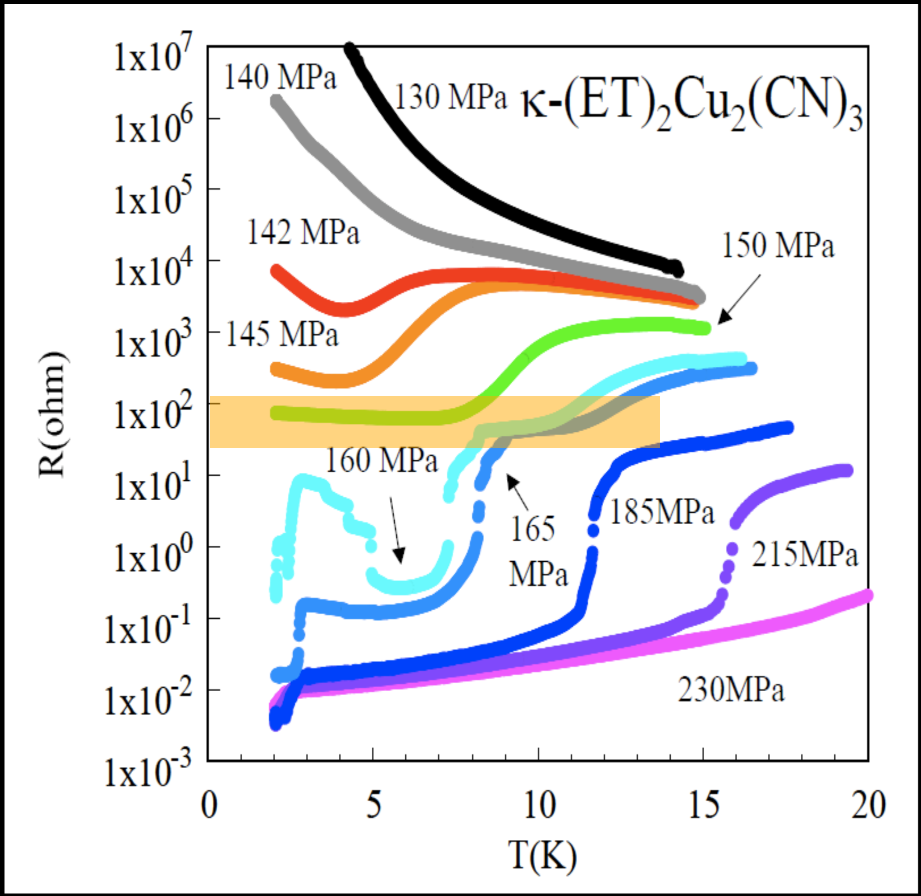
Optical conductivity

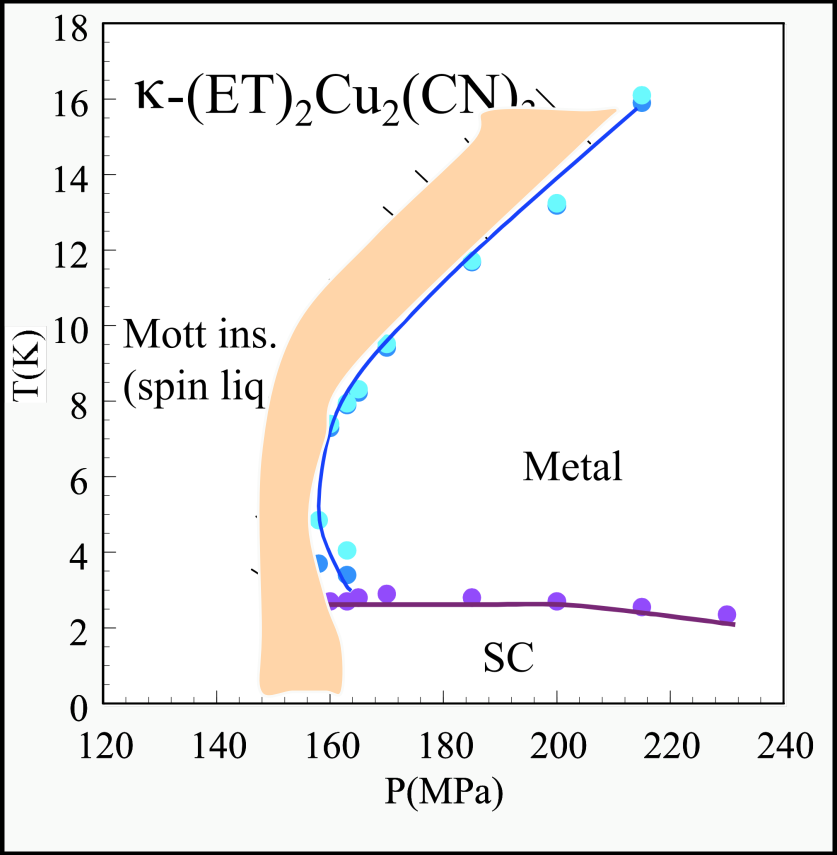
Kezsmarki, Tokura et al.
PRB 74(2006)201101(R)





Charge-gapless spin liquid just before Mott transition ?



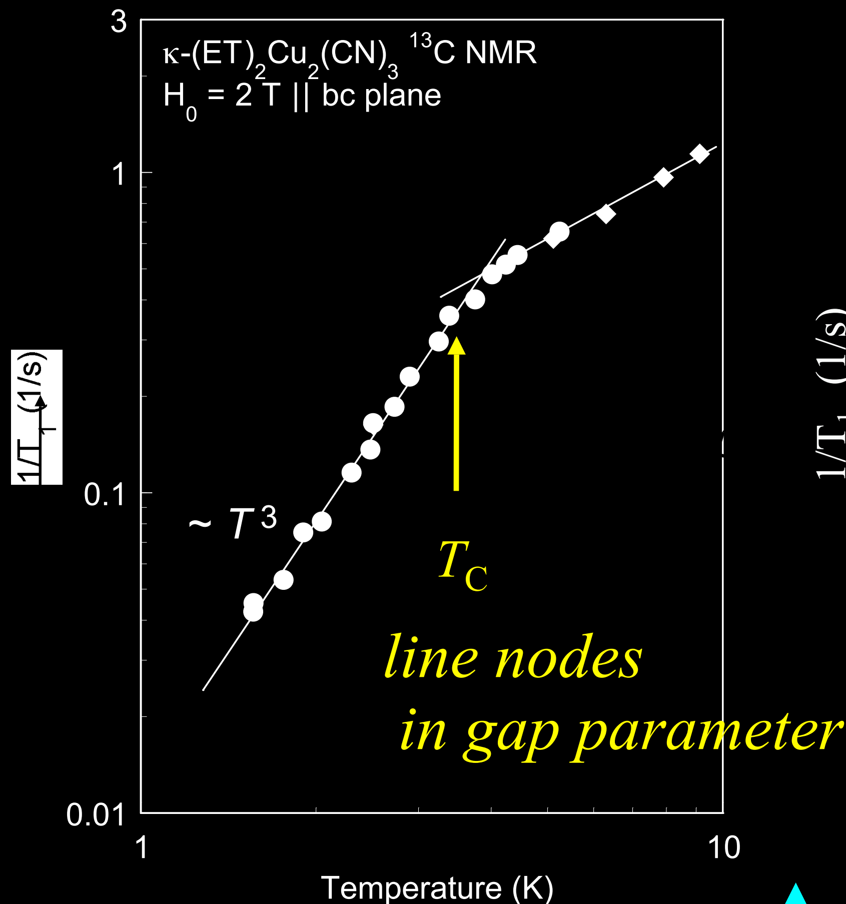


Issue 3; charge state of spin liquid around Mott transition

^{13}C NMR $1/T_1$ in the superconducting state

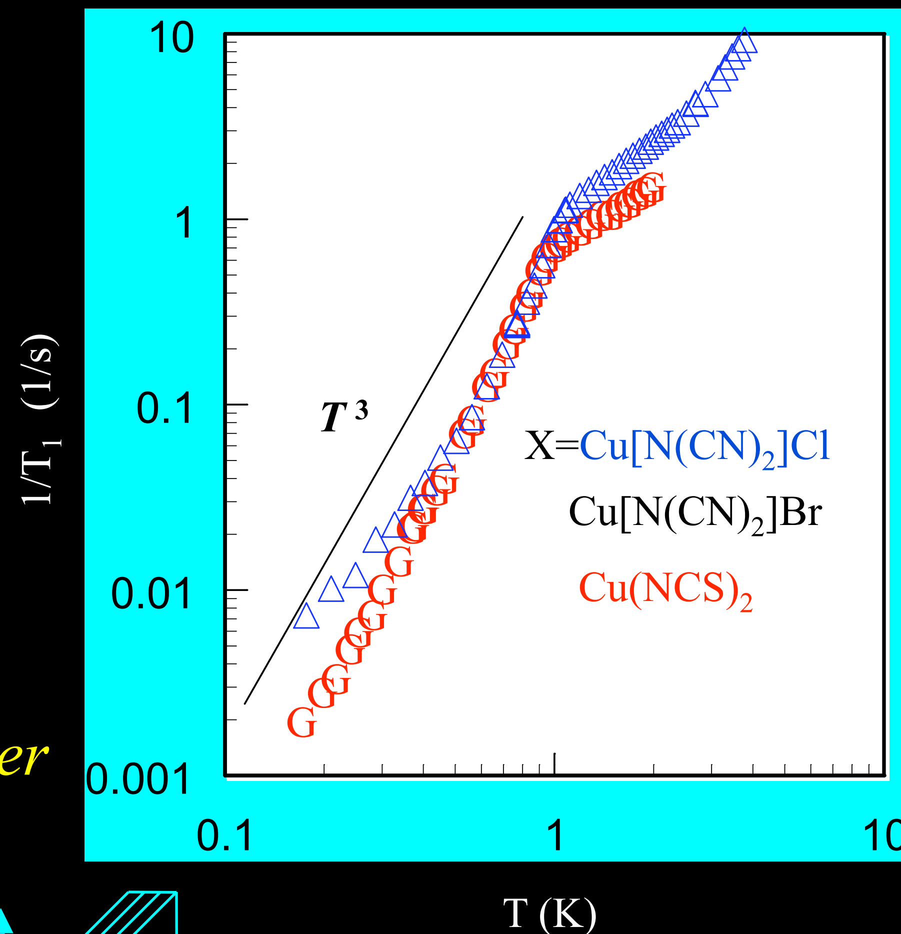
SC neighboring Spin liquid

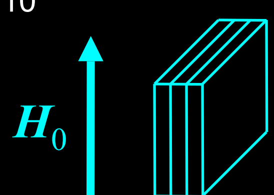
κ-$(ET)_2Cu_2(CN)_3$ under 4.3 kbar



SC neighboring AFI

κ-$(ET)_2X$

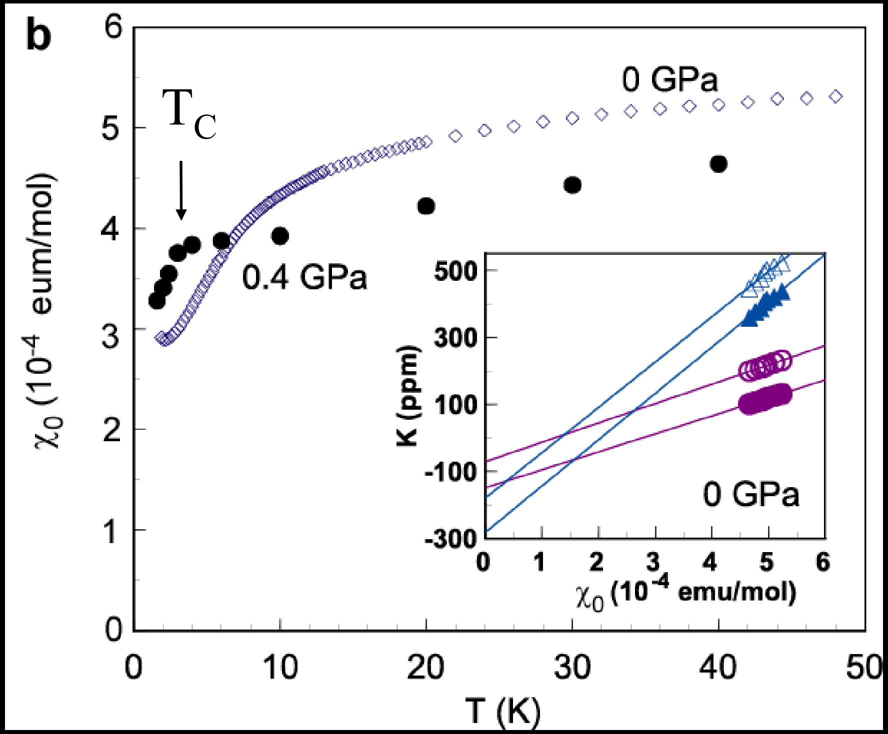


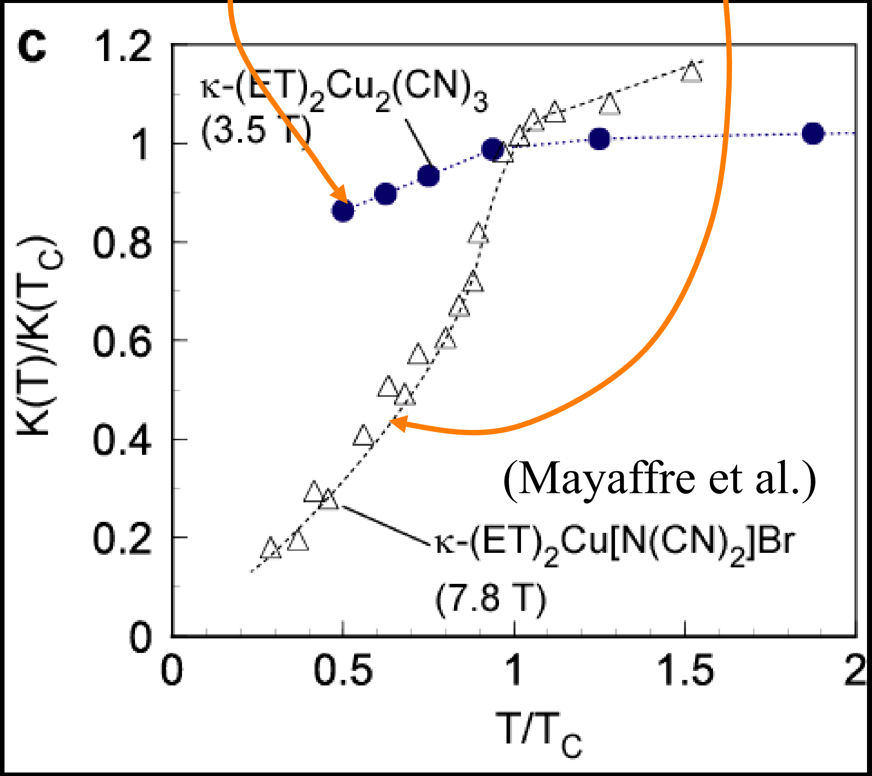


Shimizu et al. (2006)

^{13}C NMR Knight shift

SL-neighbored SC AF-neighbored SC



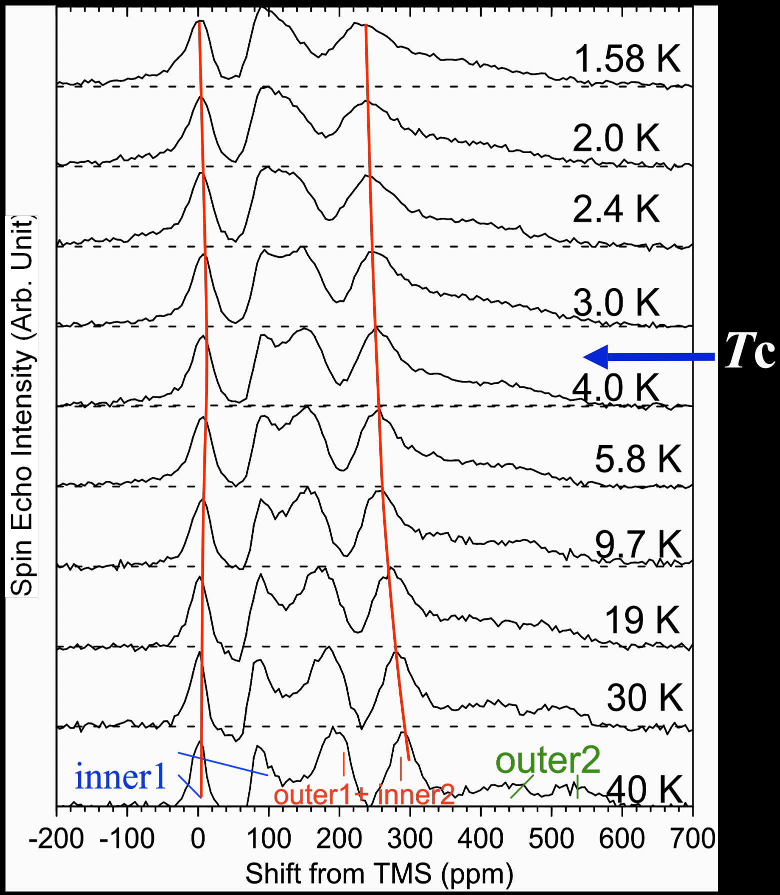


A quite small decrease in shift

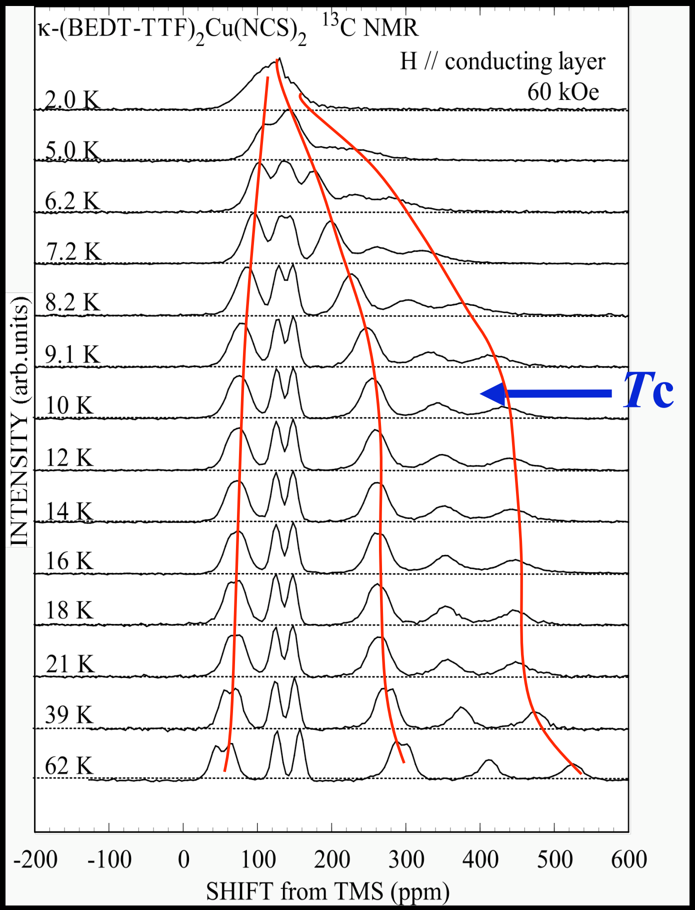
inconsistent with simple singlet pairing

^{13}C NMR spectra across Tc

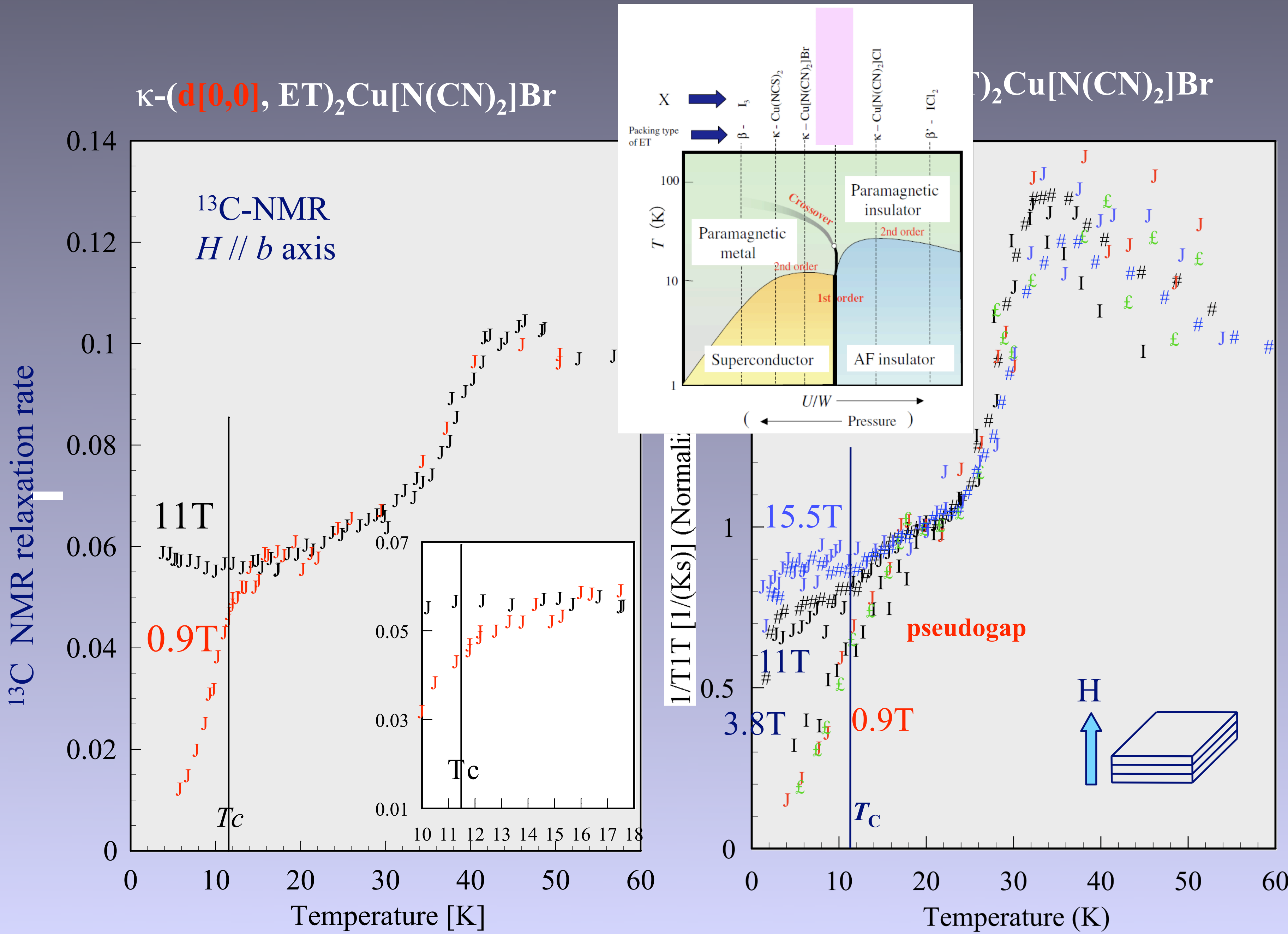
κ-$(ET)_2Cu_2(CN)_3$



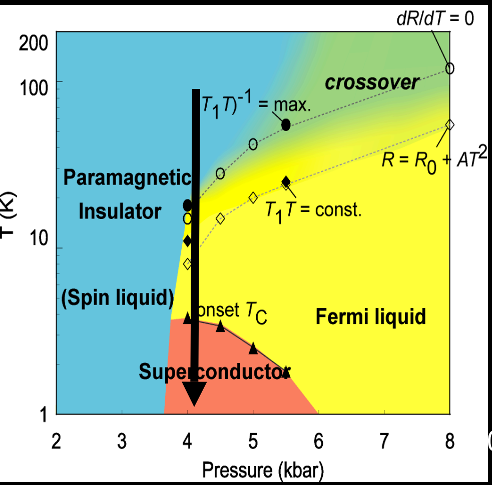
κ-$(ET)_2Cu(NCS)_2$



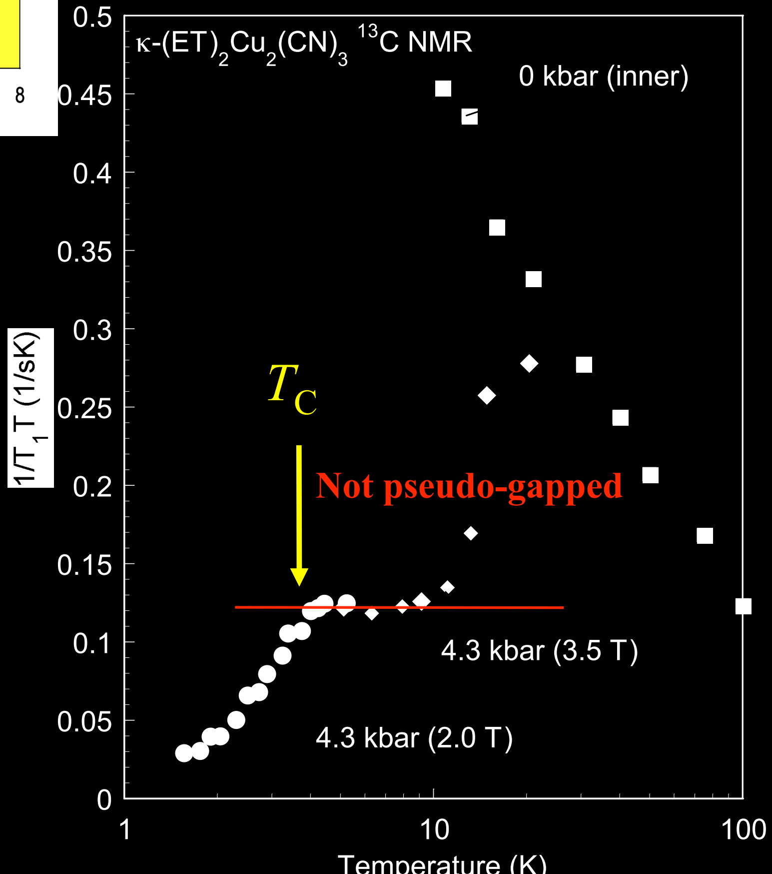
Pseudogap in $1/(T_1T)$ in anisotopic triangular lattice



Absence of pseudogap in k-$(ET)_2Cu_2(CN)_3$ under 4.3 kbar

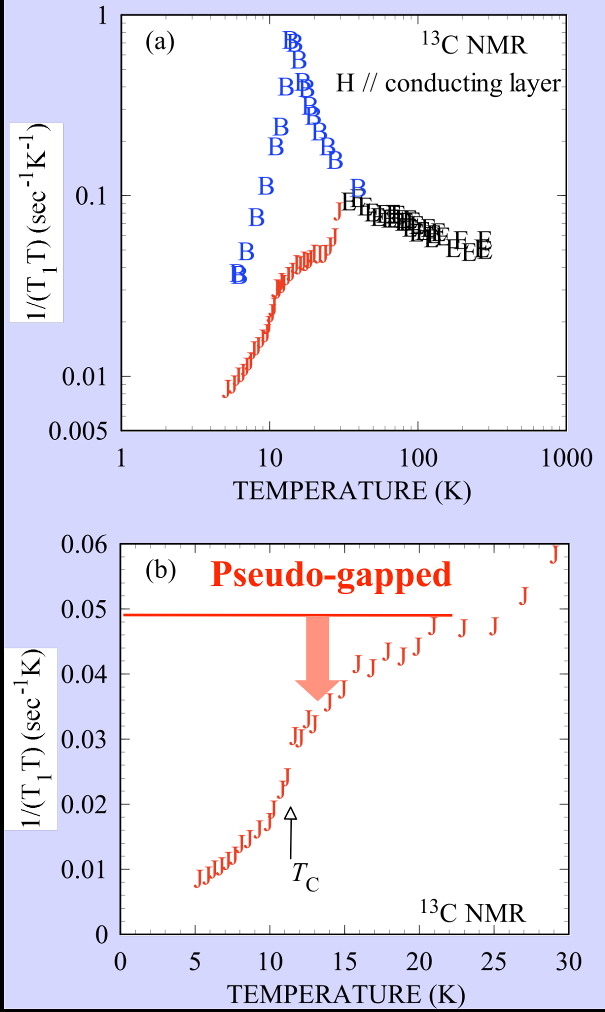


Spin liquid-neighbored SC
in κ-$(ET)_2Cu_2(CN)_3$



Shimizu et al.
(2006)

AF-neighbored SC
in deuterated κ-$(ET)_2Cu[N(CN)_2]Br$

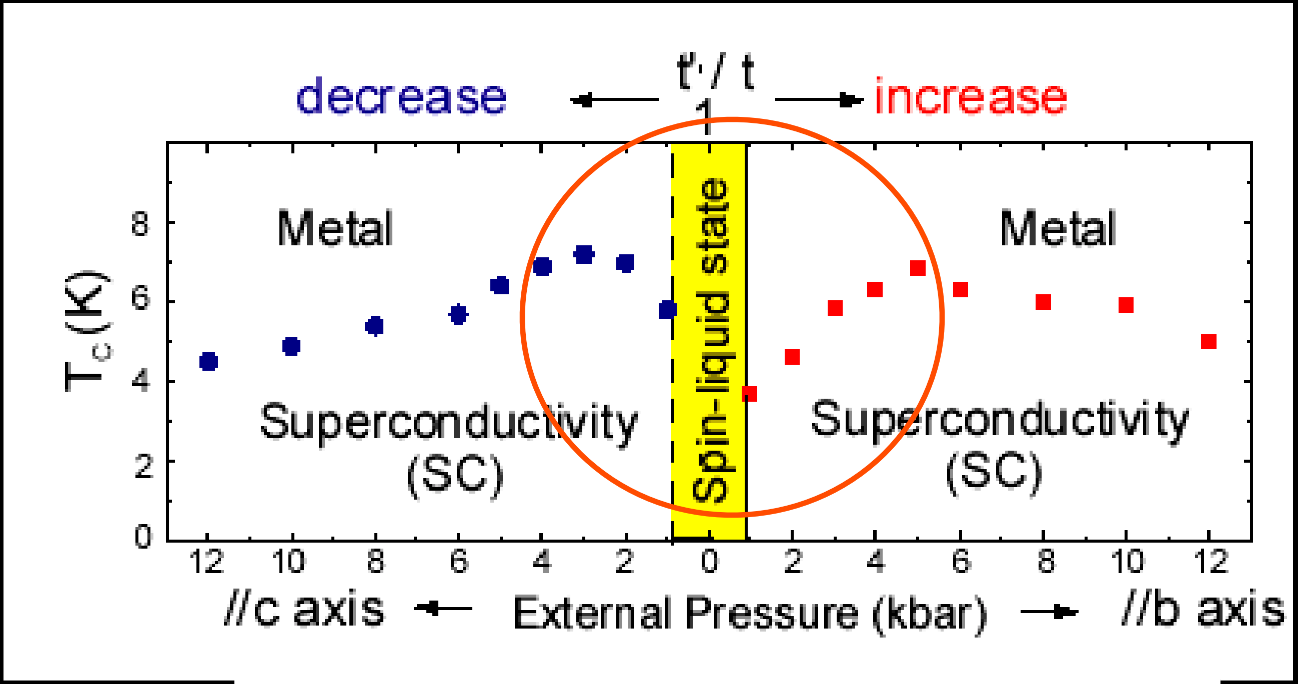


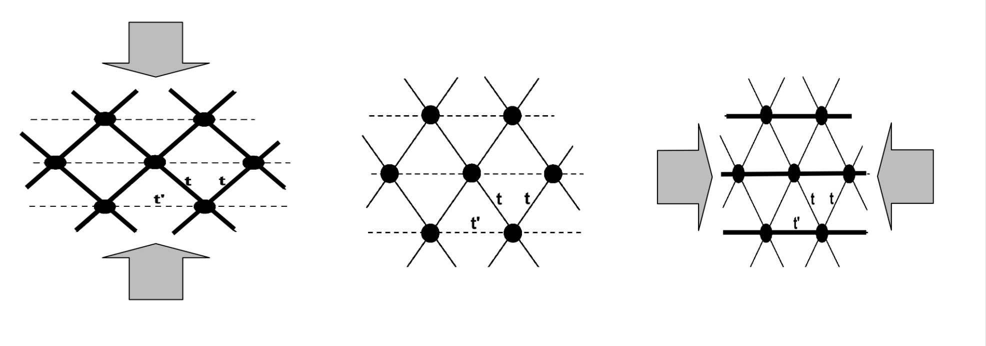
Miyagawa *et al.*, PRL89 (2002) 017003

T_C is initially enhanced when triangular lattice is deformed

Triangularity seems unfavorable to superconductivity

Uniaxial pressure study, Maesato et al. (2004)





Summary and guess on

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