KEK IPNS-IMSS-QUP Joint workshop, 2022. 02. 09

Frustration of jostling electrons on triangular lattices: - quantum charge glass and quantum spin liquid -

三角格子上で犇めき合う電子の迷い：量子電荷ガラスと量子スピン液体

1. **Quantum charge glass**
2. **Quantum spin liquid**

2'. Doped quantum spin liquid

Kazushi Kanoda

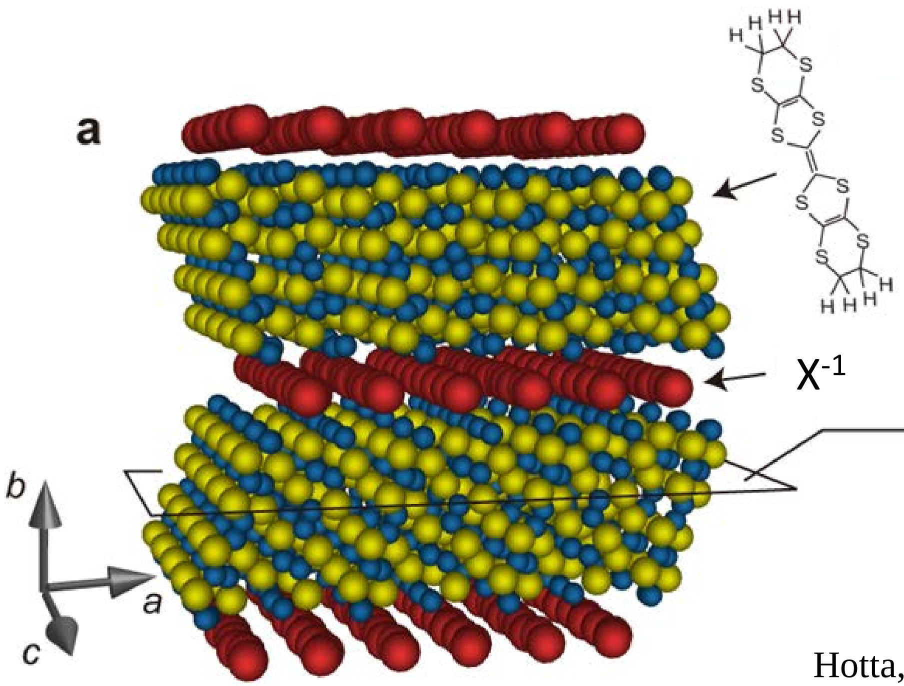
Applied Physics, University of Tokyo

Organic materials → Physics of interaction and geometry

Various in-plane structures

⇓

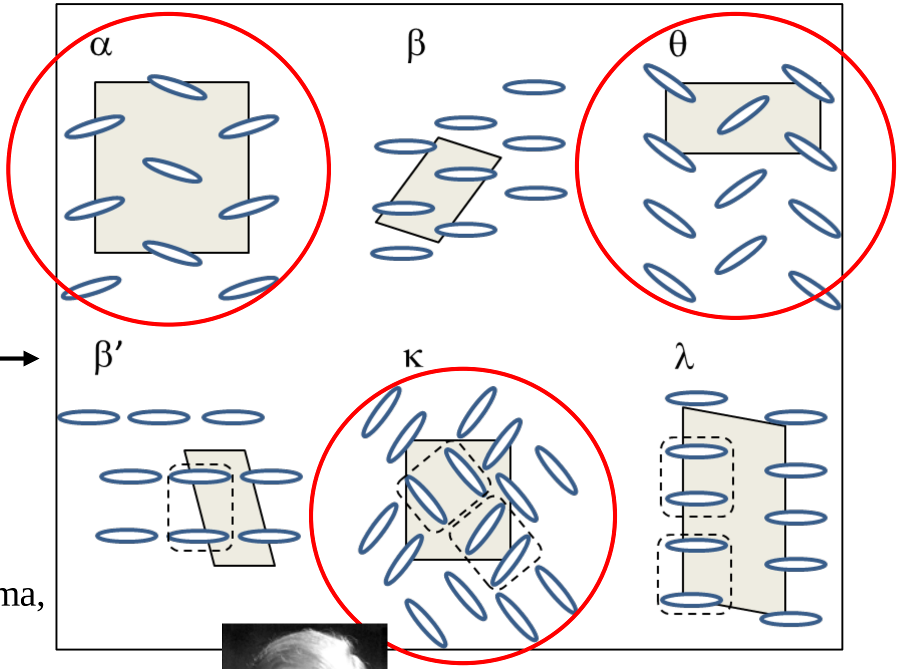
Diverse correlation phenomena



Massless Dirac ele.

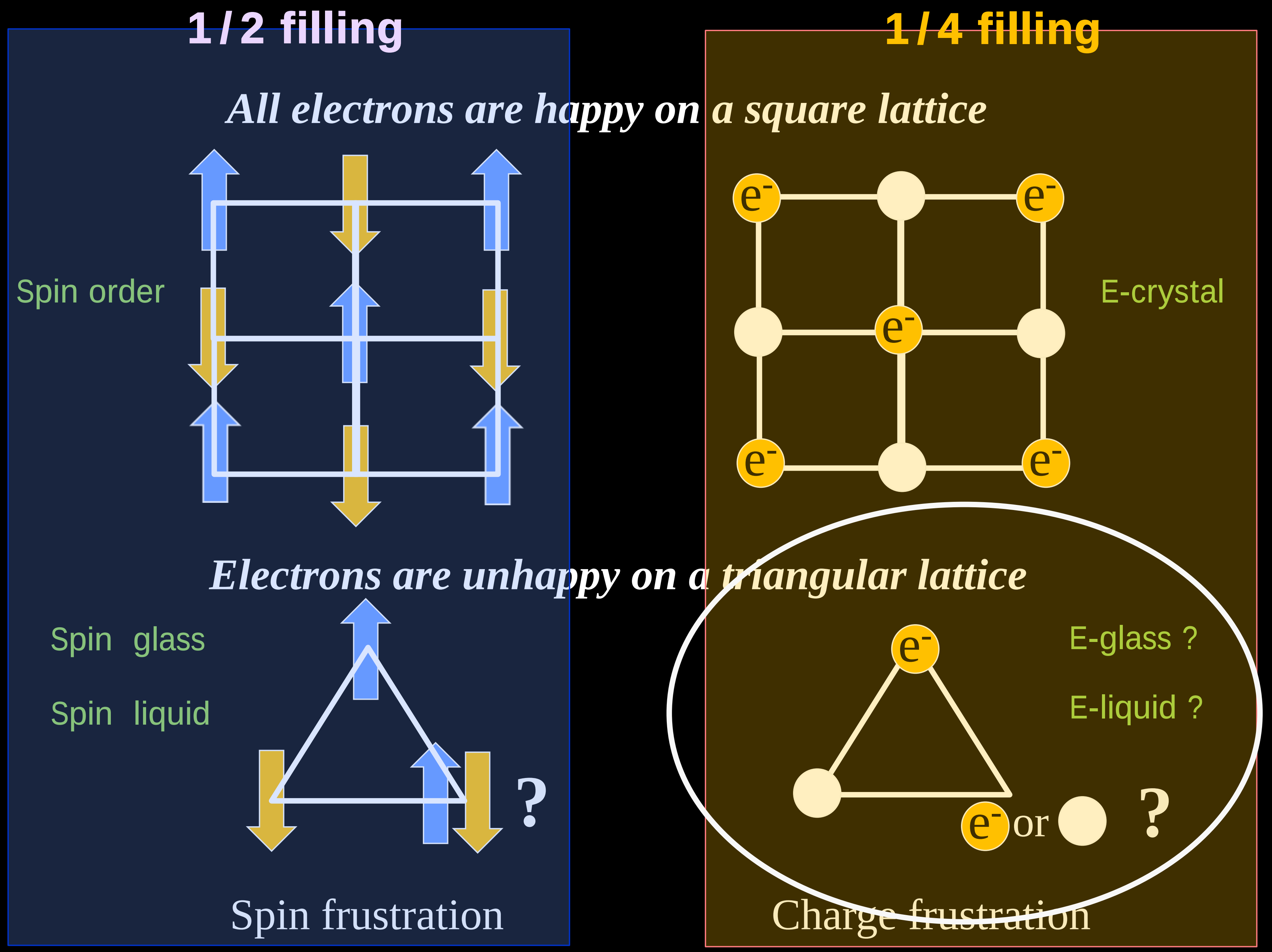
Wigner Xtal/glass

+1/2



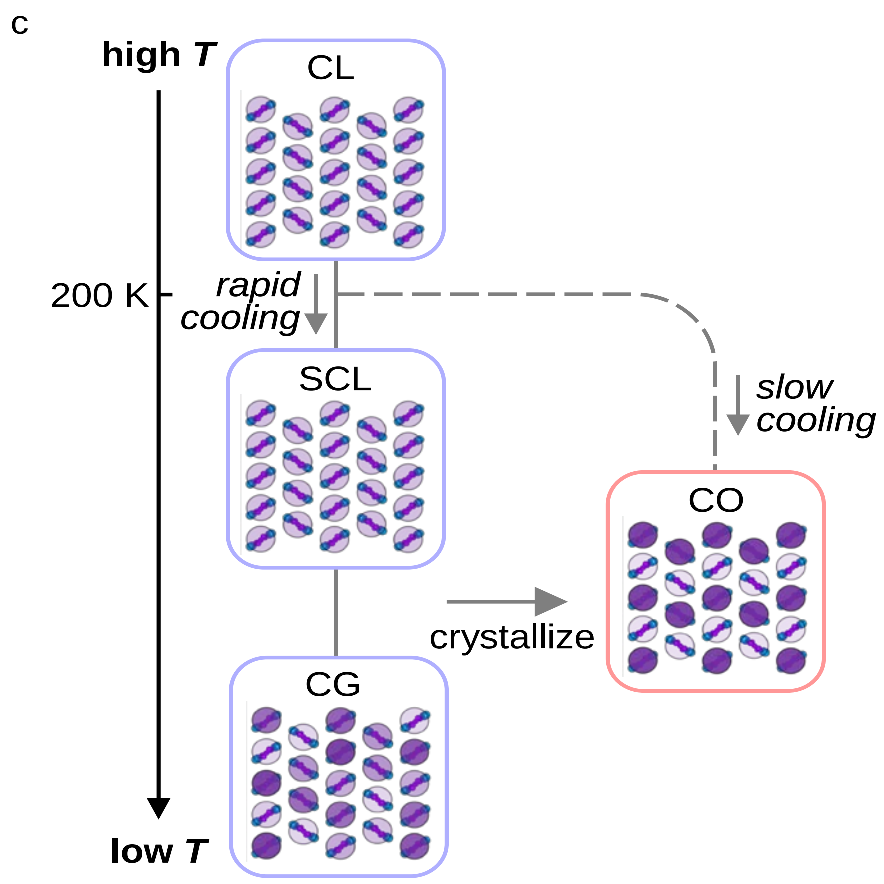
Hotta, Seo, Fukuyama, Chem. Rev., 2004

Mott physics

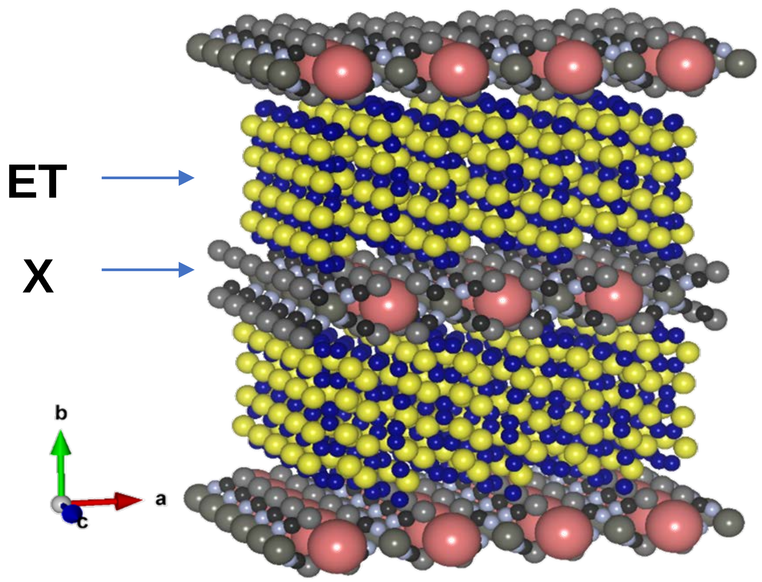

1/2 filling
1/4 filling
All electrons are happy on a square lattice
Spin order
E-crystal
e-
Electrons are unhappy on a triangular lattice
Spin glass
Spin liquid
E-glass ?
E-liquid ?
?
e- or ?
Spin frustration
Charge frustration

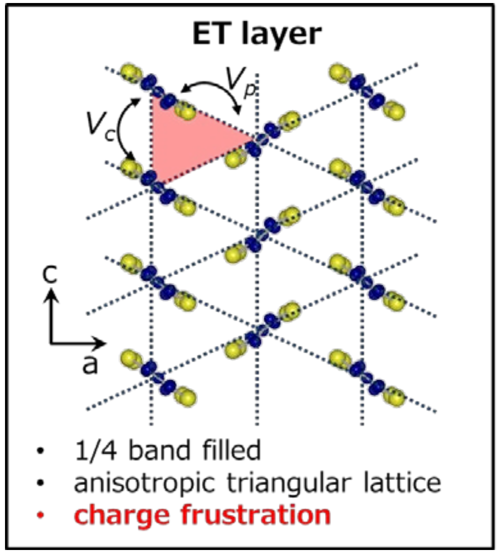
Electron glass

- ✓ **Quantum nature**
- ✓ **Controllable lattice geometry**



Charge frustrated materials, θ-$(ET)_2X$





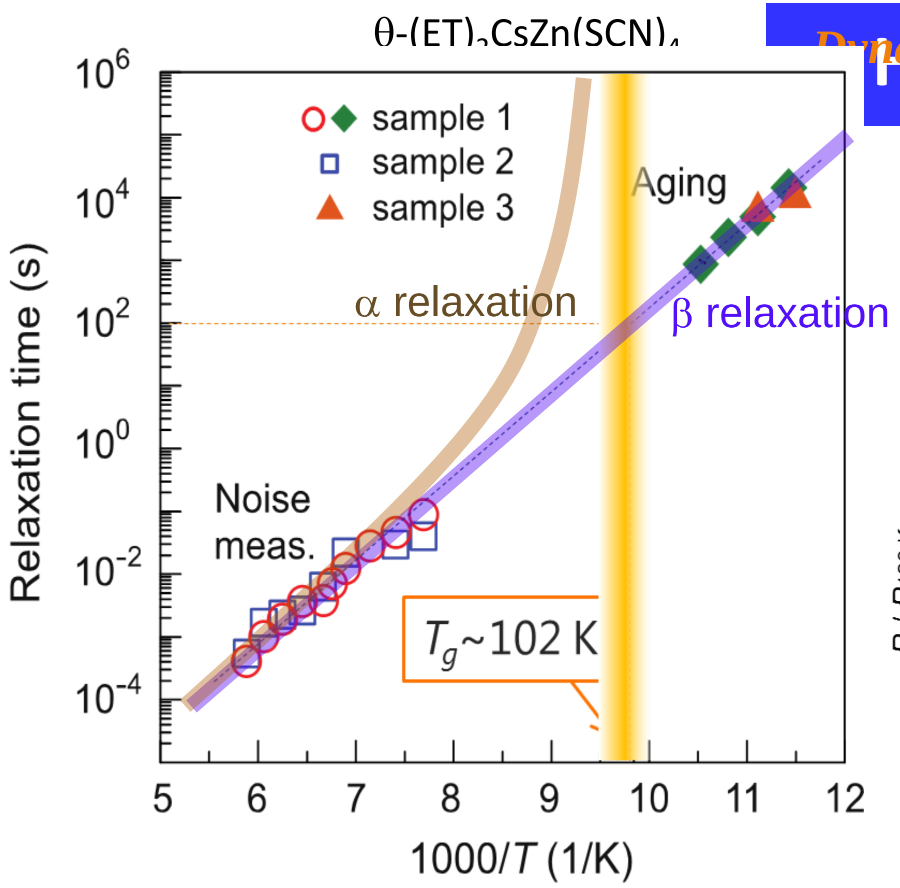
1. **Hallmarks of classical glass**
2. **E-crystallization**
3. **From classical to quantum charge glass**

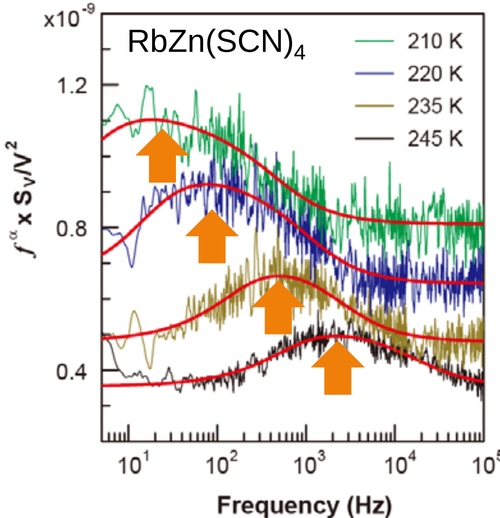
Hallmarks of Glass

- Slow dynamics
- Non-equilibrium
- Short or middle-range correlation

Dynamics
Hallmarks of Glass

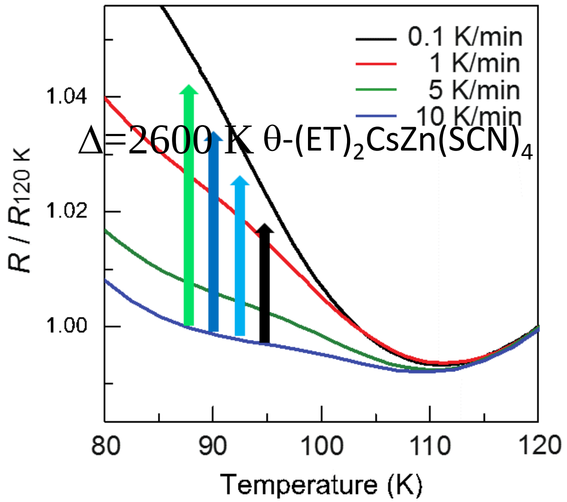
Kagawa *et al.*, *Nat. Phys.* **9**, 419 (2013).
Sato *et al.*, *PRB* **89**, 121102 (2014)
Sato *et al.*, *JPSJ* **83**, 083602 (2014)
Sato *et al.*, *JPSJ* **85**, 123702 (2016)



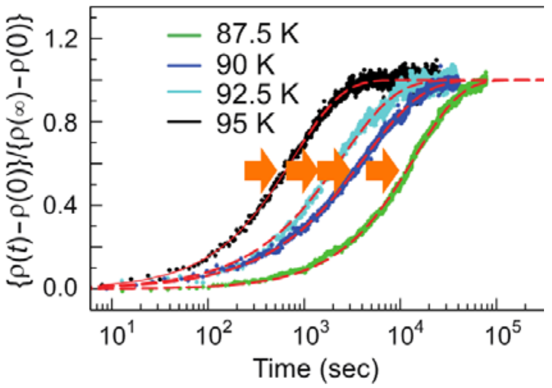


Non-equilibrium nature

ρ depends on cooling rate

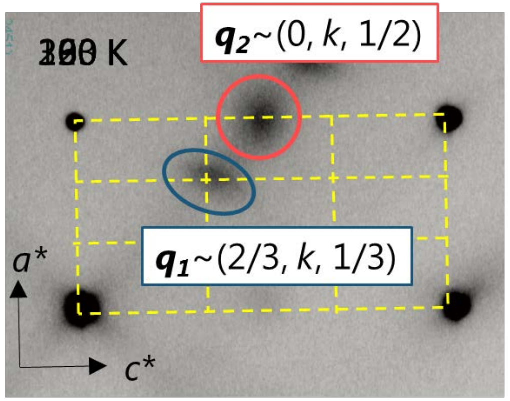


Aging

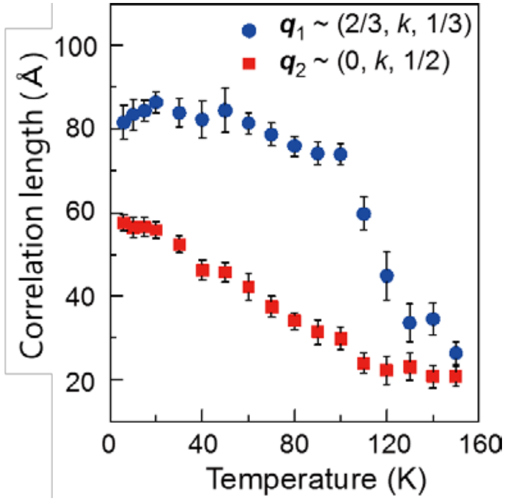


middle-range correlation

X-ray diffuse scattering



Correlation length levelling off

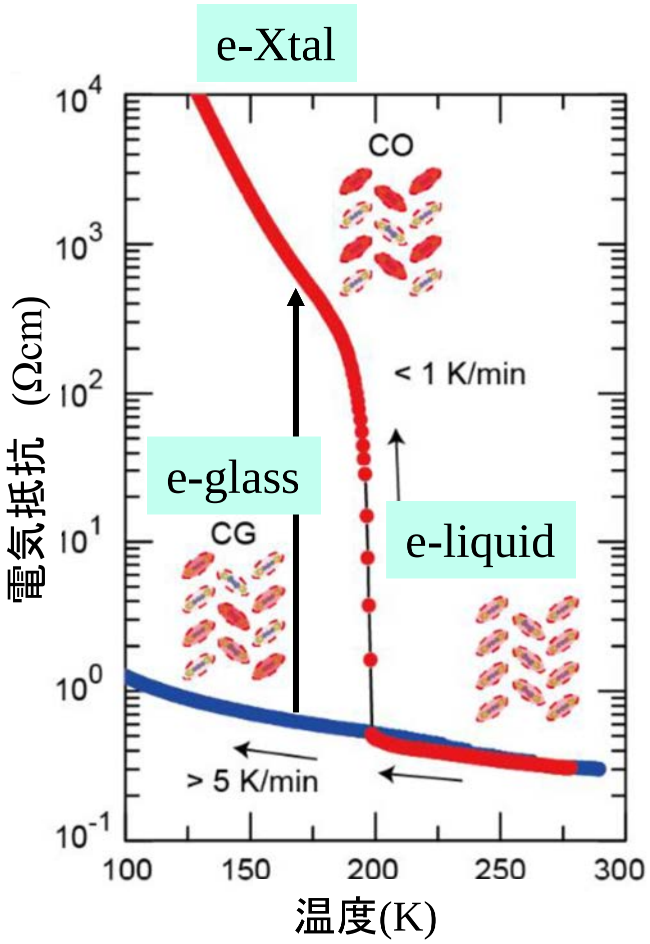


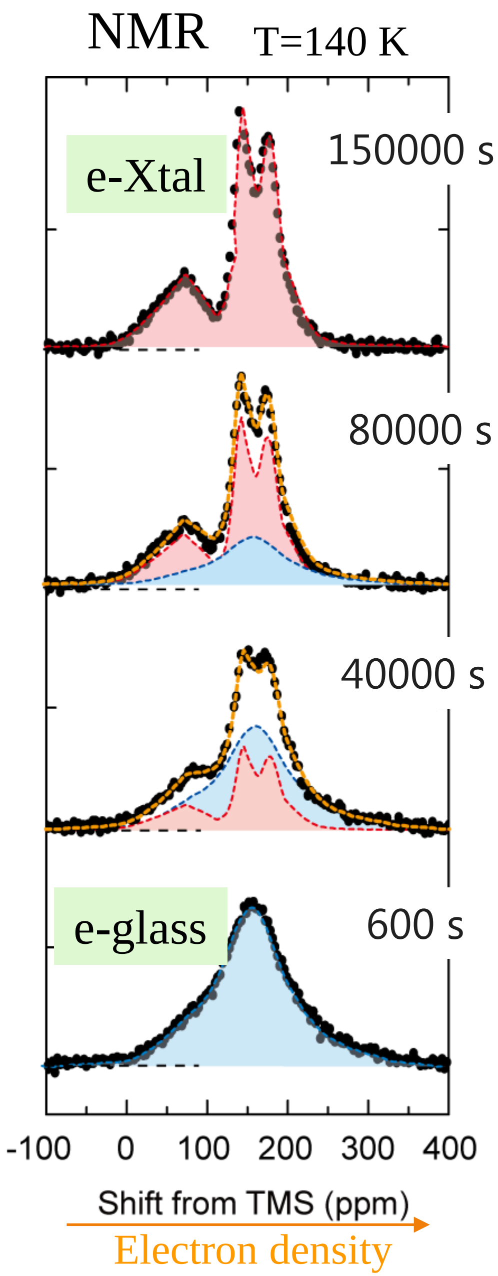
Electronic crystallization: ρ and NMR

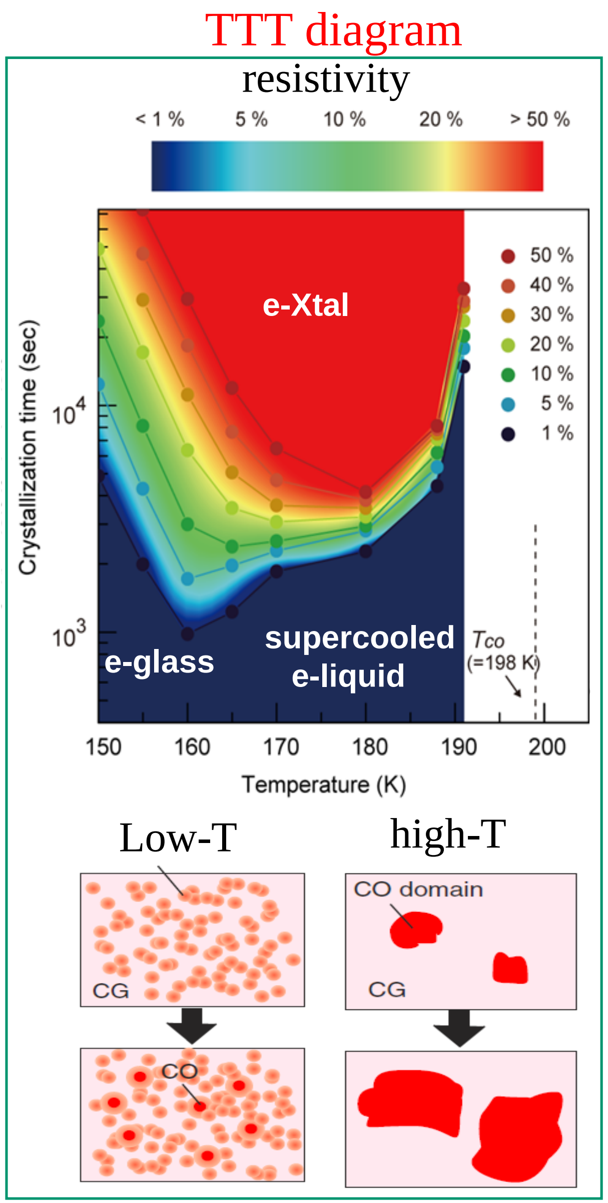
T. Sato et al., Science **357**, 1378 (2017)

(*cf. S. Sasaki et al., Science* **357**, 1381 (2017))

θ-$(ET)_2RbZn(SCN)_4$



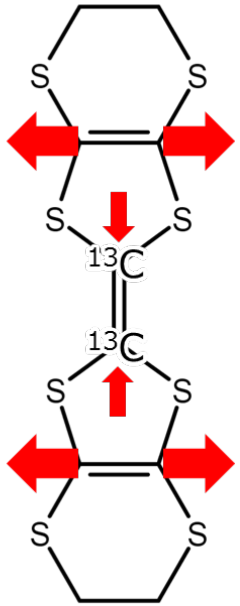




Electronic crystallization: Raman spectroscopy

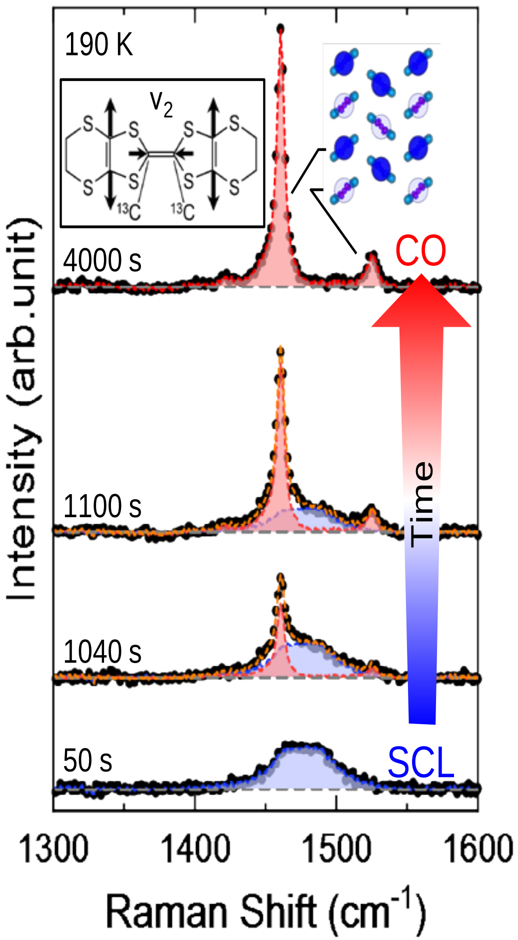
θ-$(ET)_2RbZn(SCN)_4$

Charge-sensitive ν_2 mode

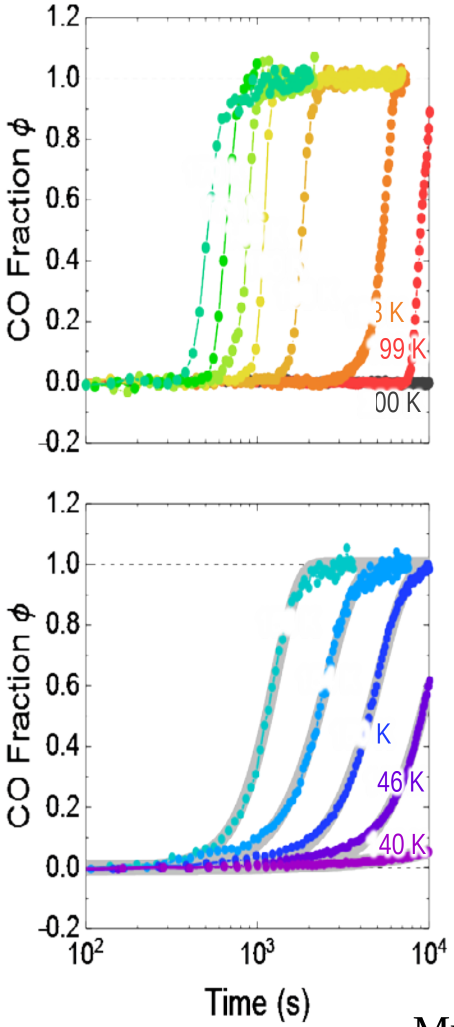


K.Yamamoto, *et al.*,
Phys. *Rev*. B **65**, 085110 (2002).

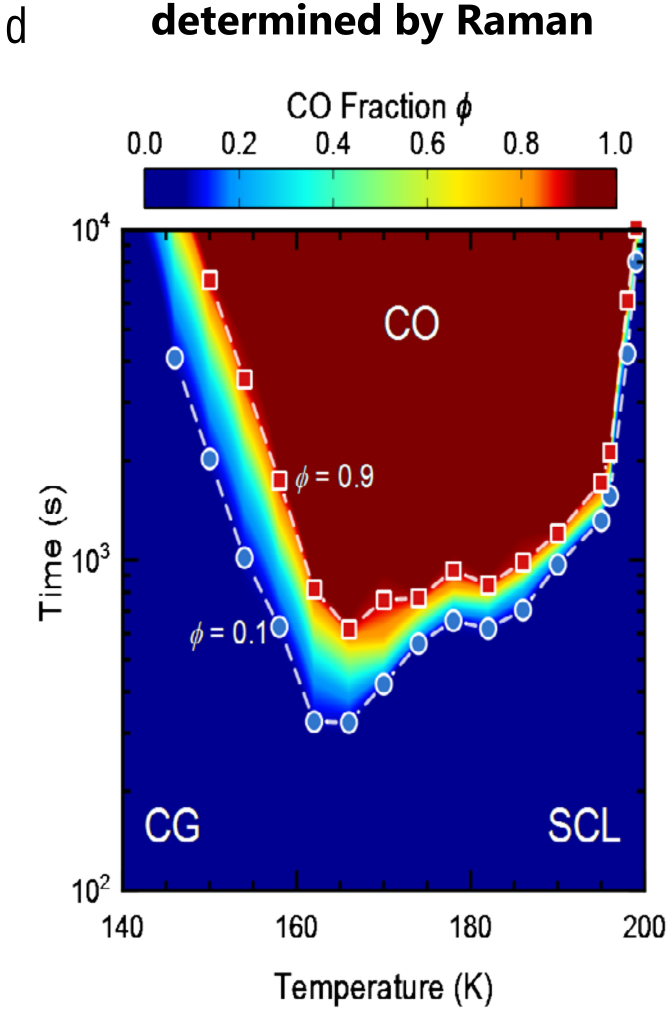
Evolution of Raman spectrum during E-crystallization



Time evolution of E-Xtal fraction



TTT diagram determined by Raman

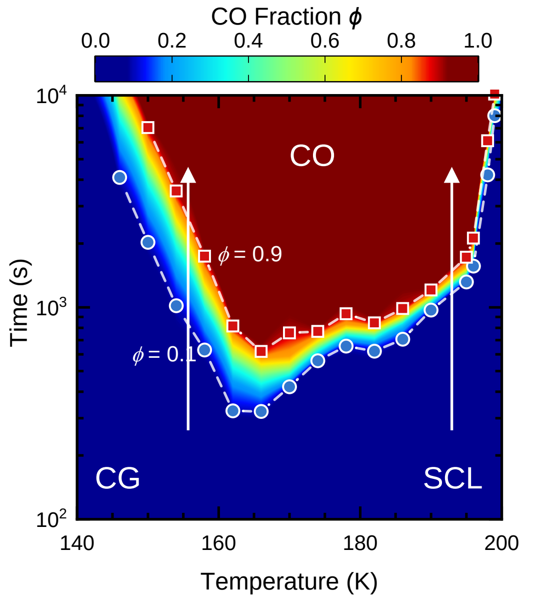


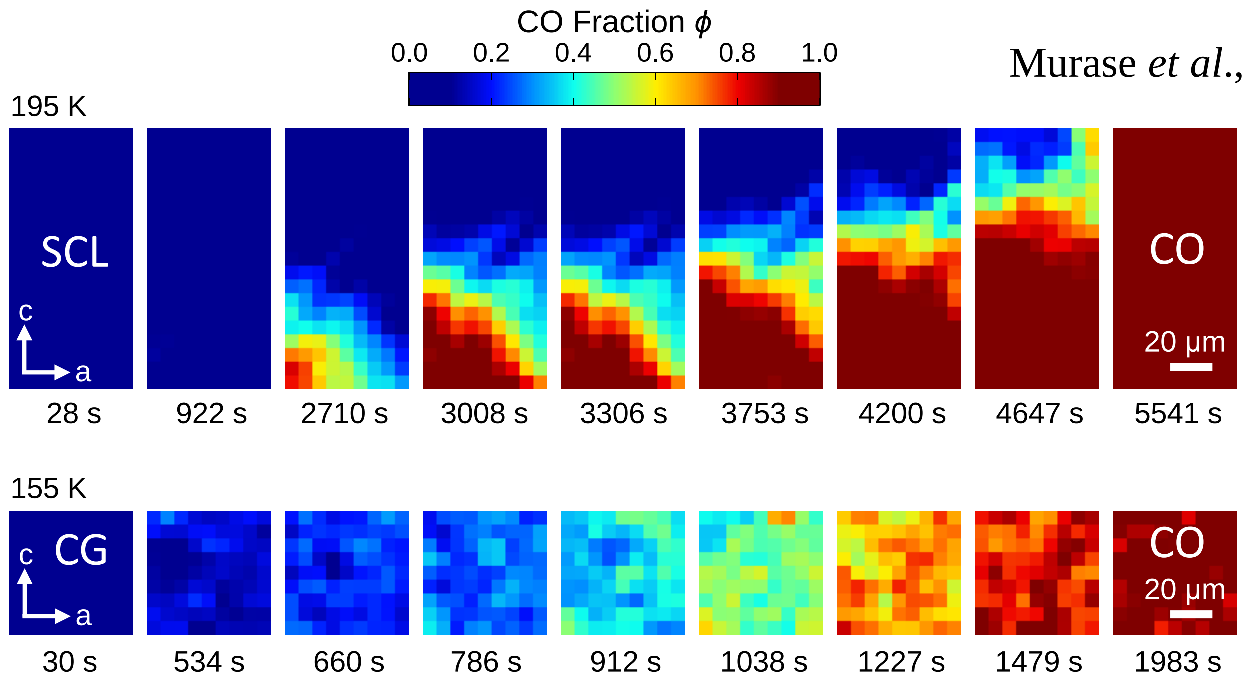
Murase *et al.*, arXix:2201.04855

Spaciotemporal observation of electronic crystallization: classical glass

Murase *et al.*, arXix:2201.04855

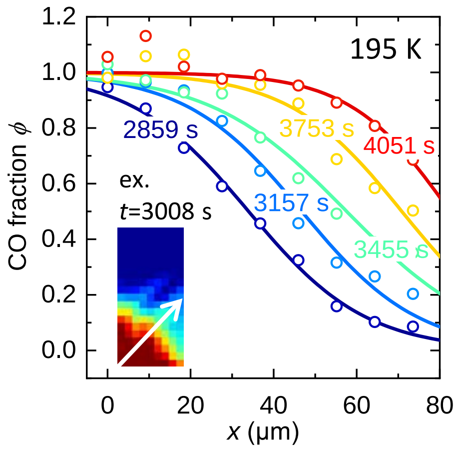
TTT diagram

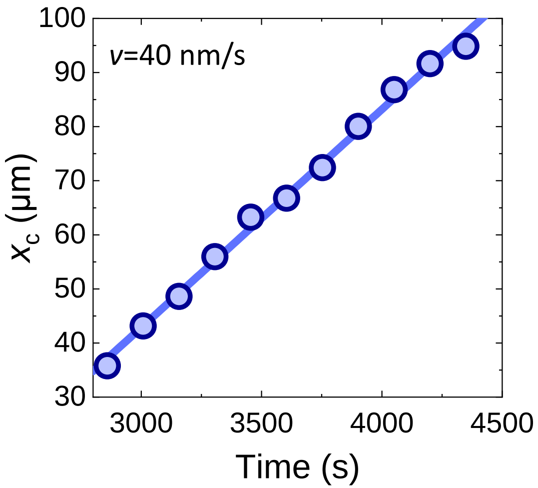


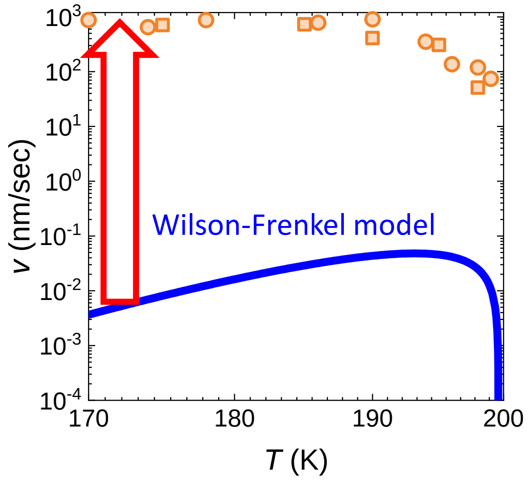


Ultrafast crystal growth: quantum effect ?

five orders of magnitude faster than expected in the classical model !





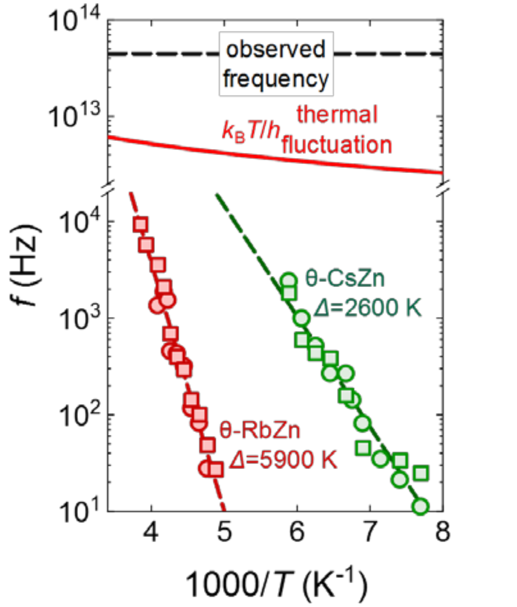


From classical to quantum charge glass

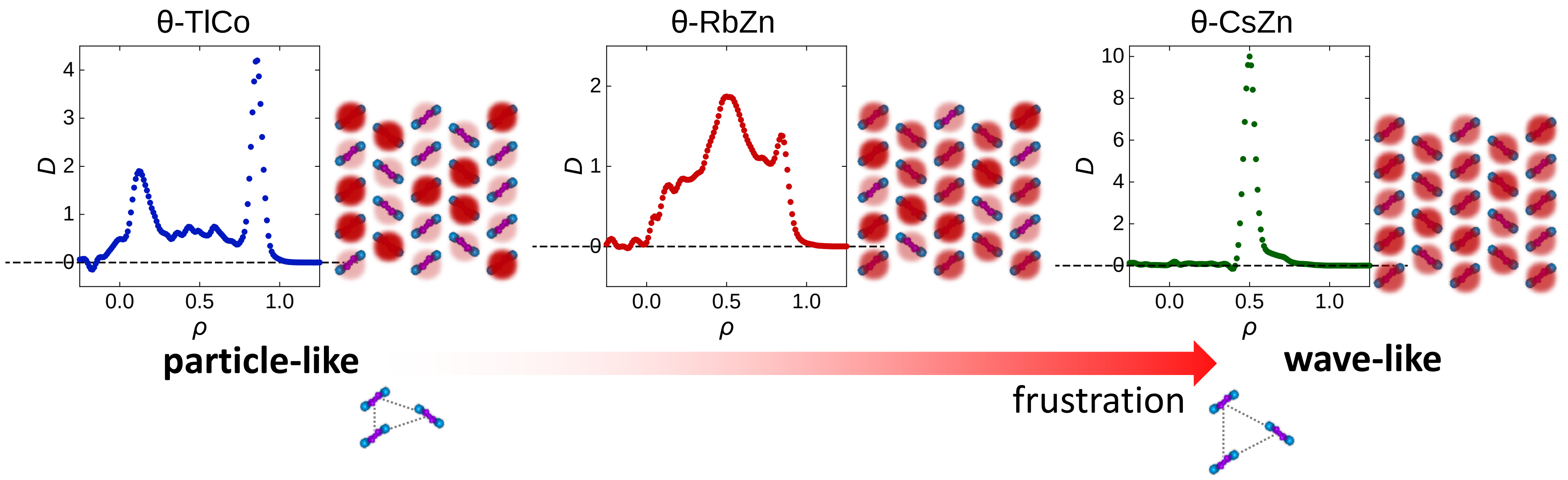
Murase *et al.*, unpublished

Observation time scale

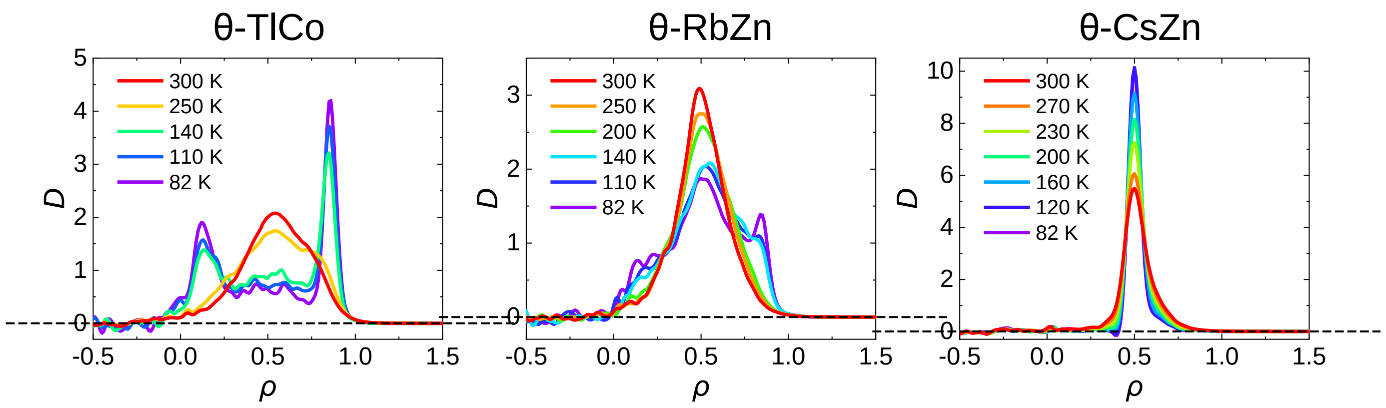
$f_{obs} >> f_{thermal} >>>> f_{glass}$

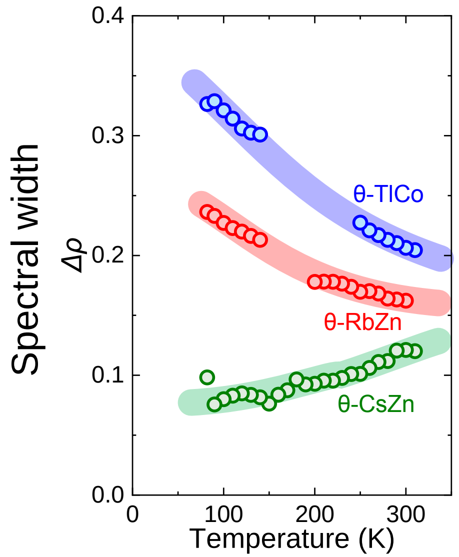


Frustration dependence of charge density in CG at 82 K

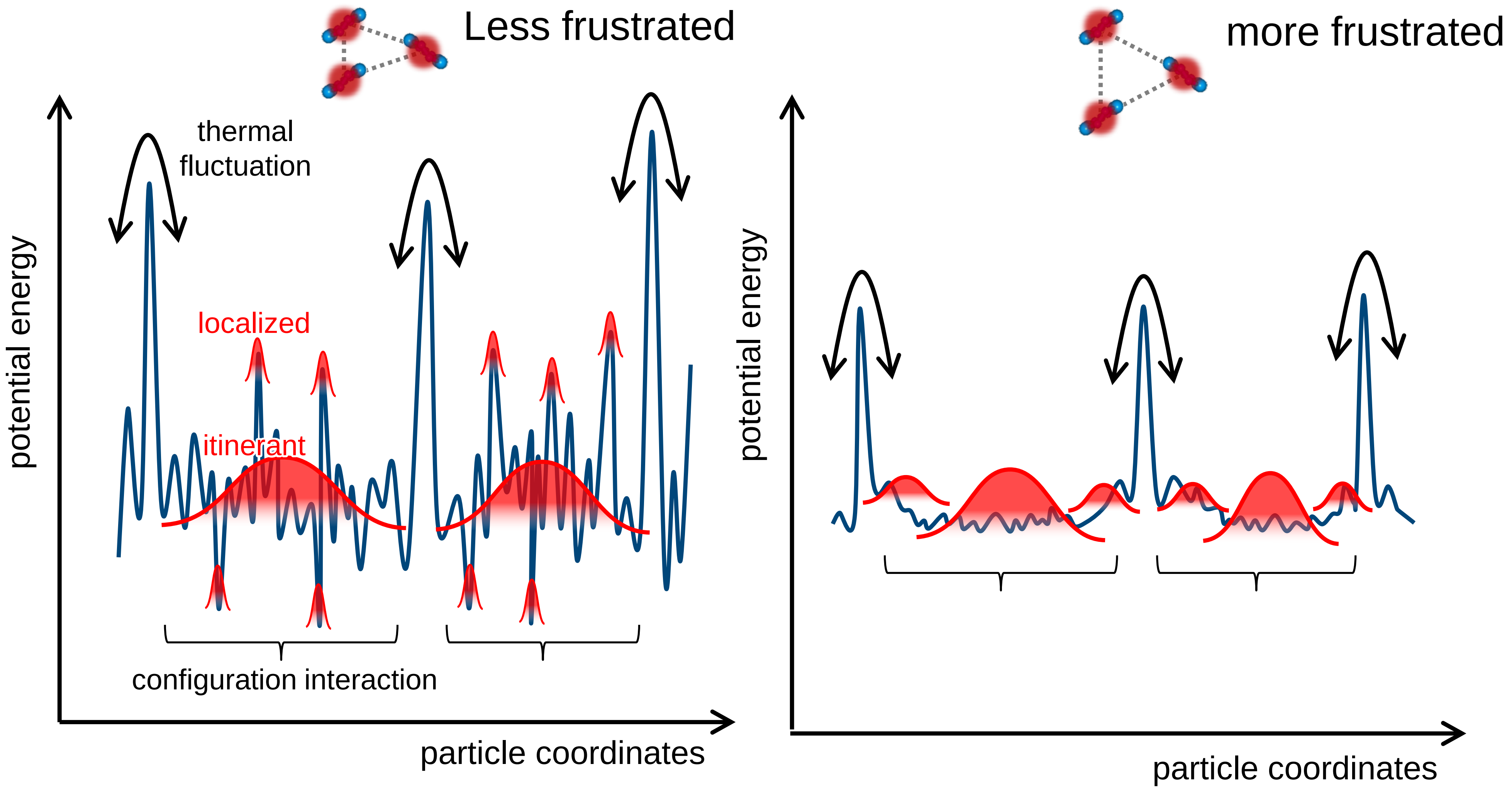


Temperature dependence of charge density



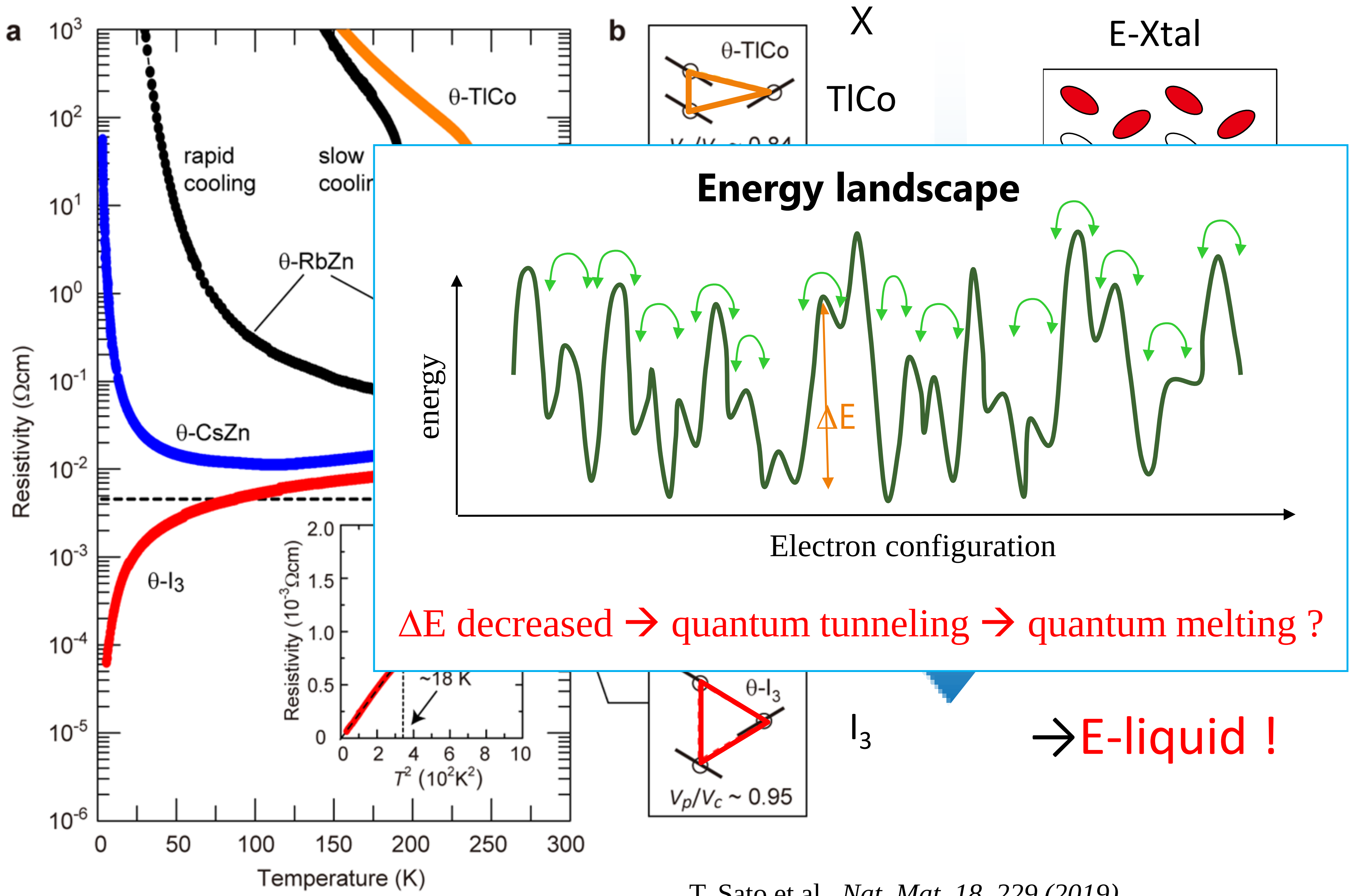


Discussion | Energy landscape



Classical glass **Quantum glass**

Possible quantum melting of E-glass

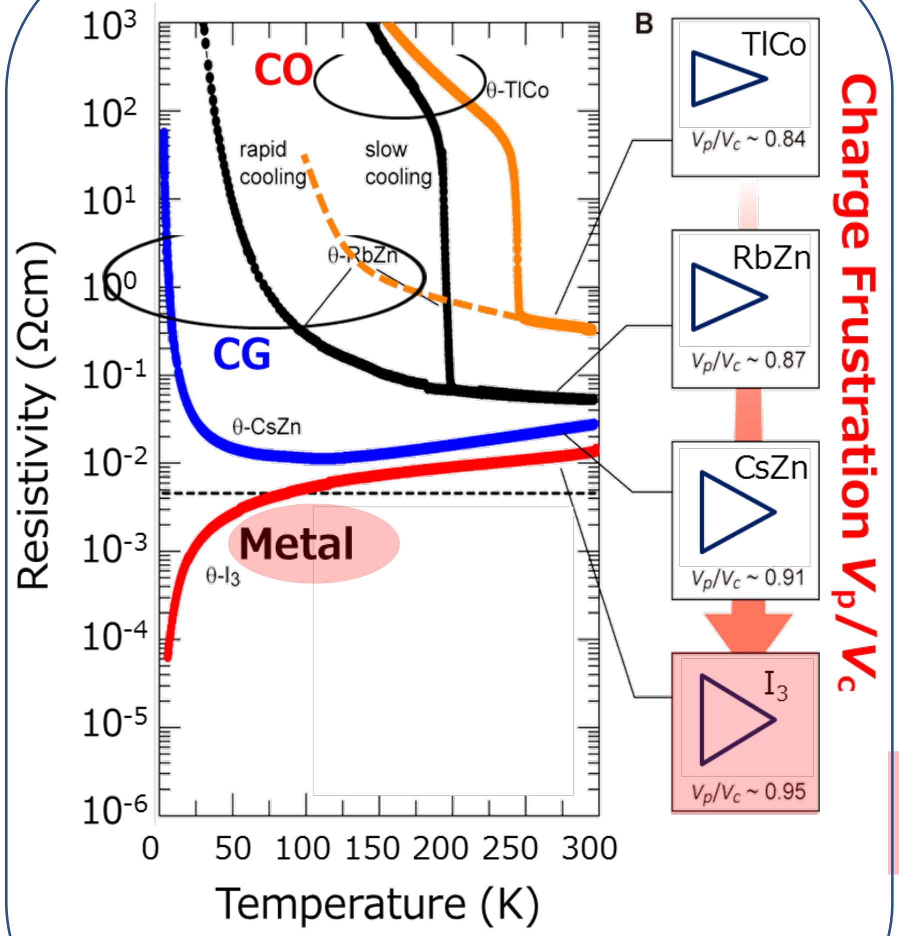


ΔE decreased → quantum tunneling → quantum melting ?

T. Sato et al., *Nat. Mat. 18, 229 (2019)*

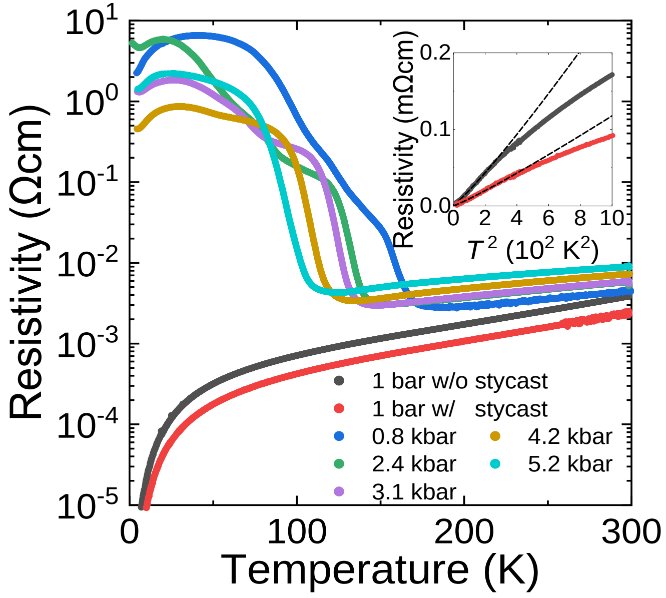
4. Frustration driven quantum melting of CO/CG

先行研究

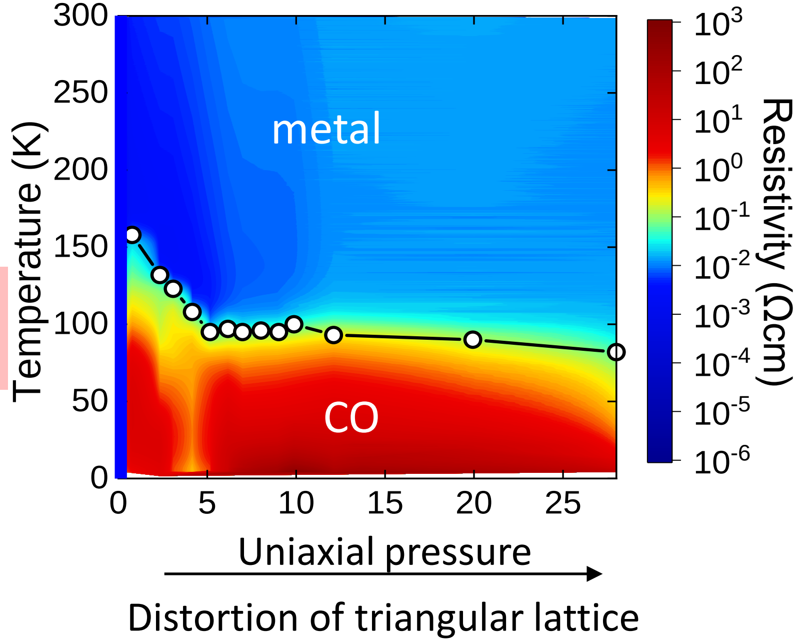


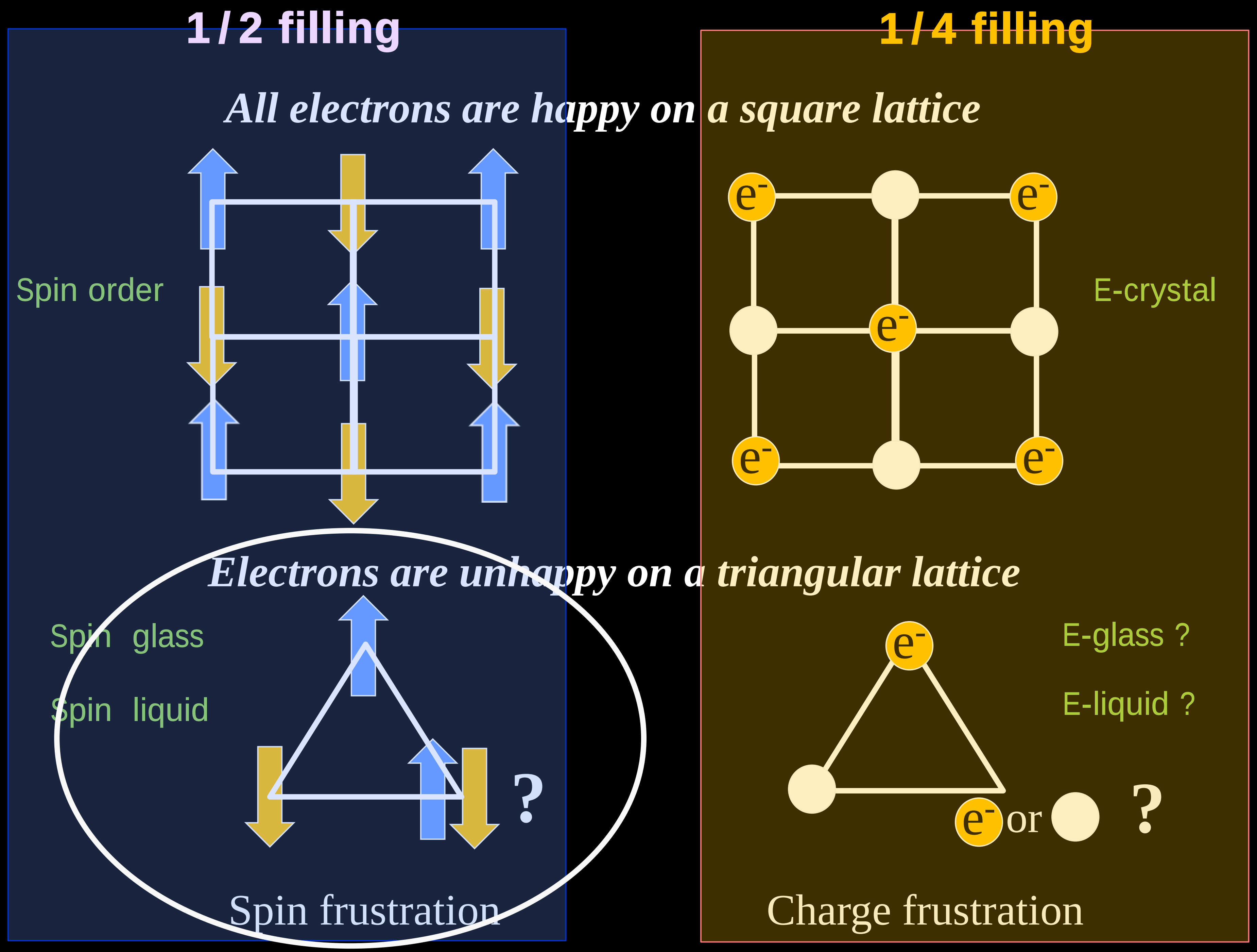
T. Sato, *et al.*, *Nat. Mater.* **11** (2019).

Uniaxial pressure



Murase et al., unpublished

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All electrons are happy on a square lattice
Spin order
E-crystal
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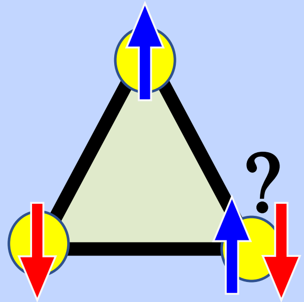
Electrons are unhappy on a triangular lattice
Spin glass
Spin liquid
?
E-glass ?
E-liquid ?
e- or ?
Spin frustration
Charge frustration

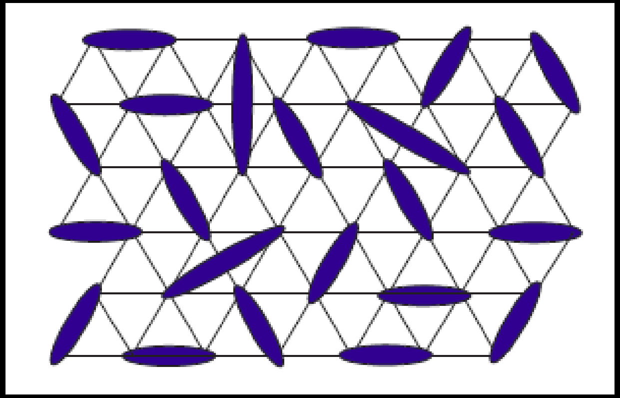
Quantum spin liquid



Spin frustration

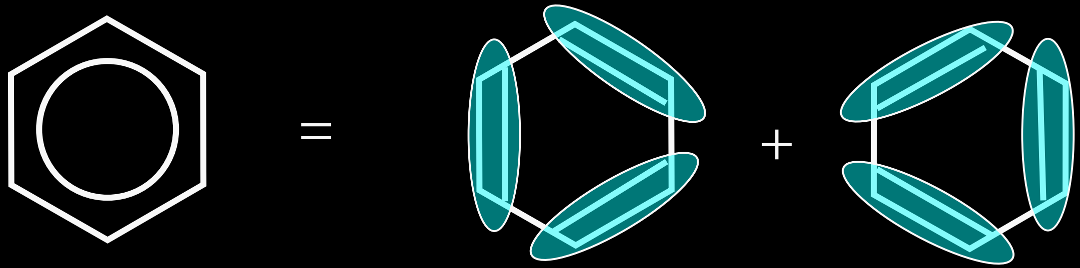
Highly degenerate spin configurations →QSL

Anderson' idea of spin liquids:
Resonating Valence Bond (RVB) state



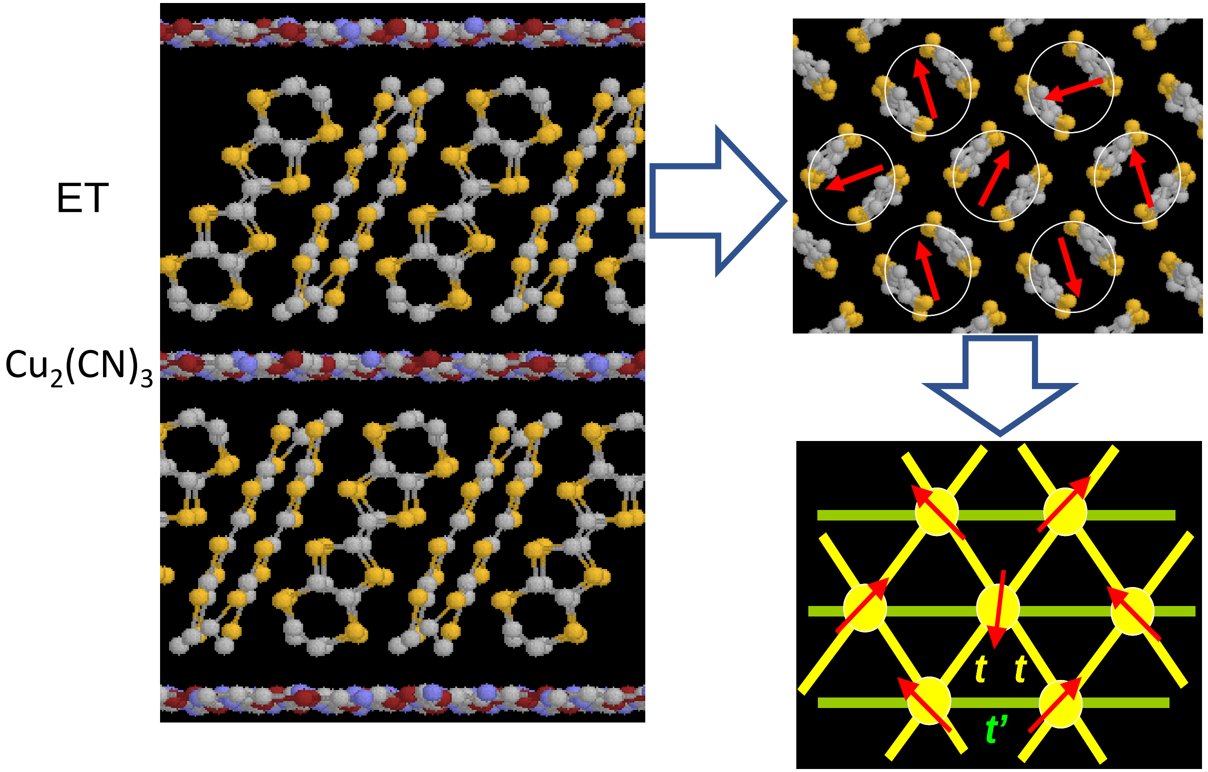
+ …… ………..

In analogy with benzene



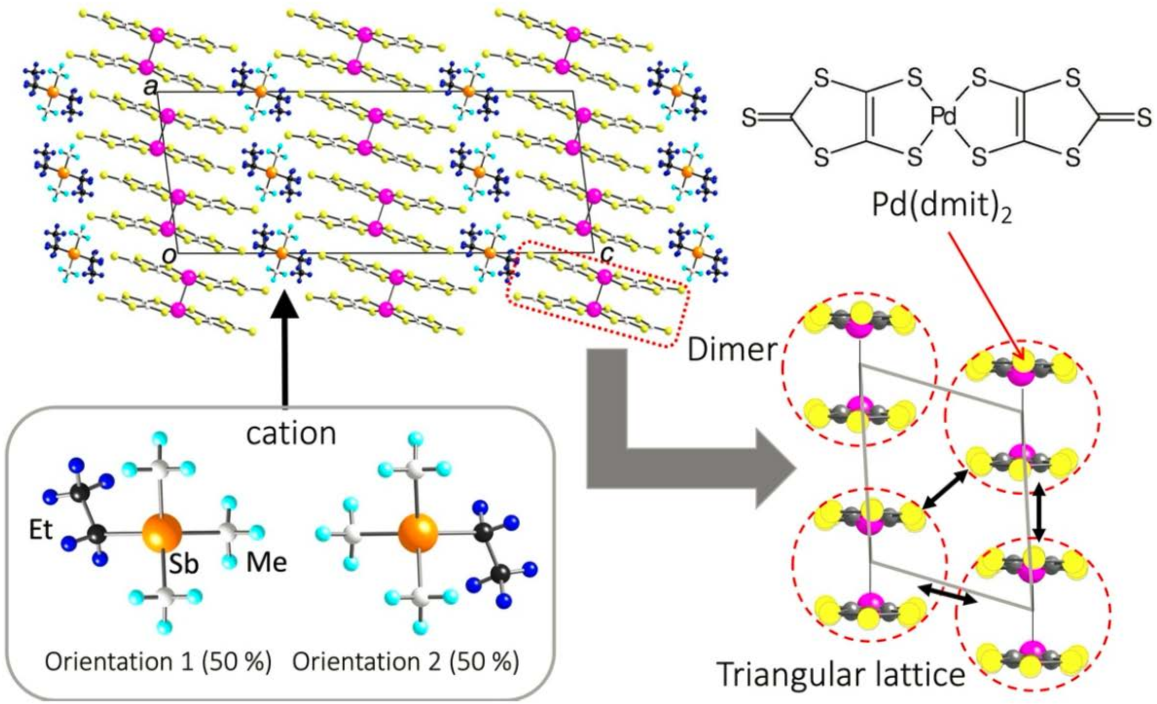
Quantum spin liquid (QSL) candidates with a triangular lattice

κ-$(ET)_2Cu_2(CN)_3$



β'-$EtMe_3Sb[Pd(dmit)_2]_2$

Kato, RIKEN

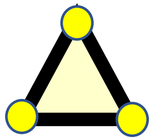


QSL manifestations

清水(現名大）
宮川

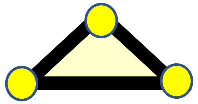
^{1}H NMR

Shimizu et al., Phys. Rev. Lett. (2003)



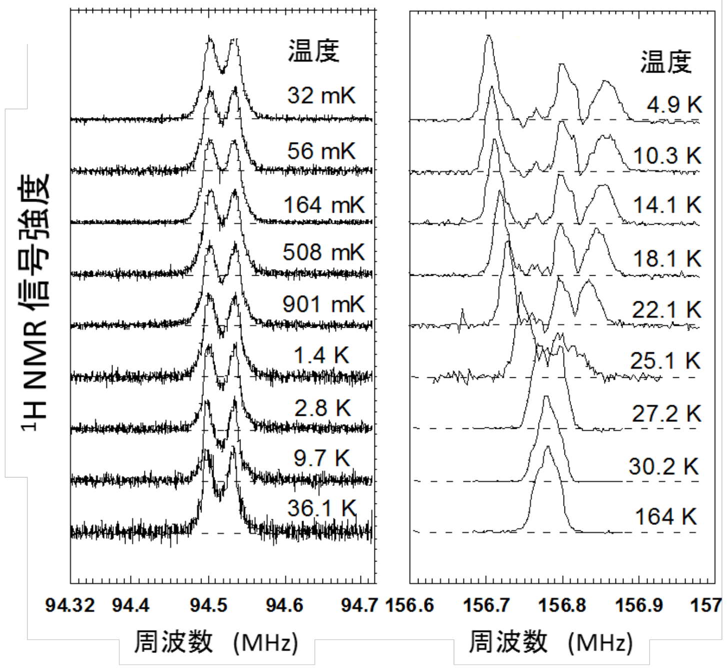
Triangular lattice

κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$



Deformed triangular lattice

κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Cl$



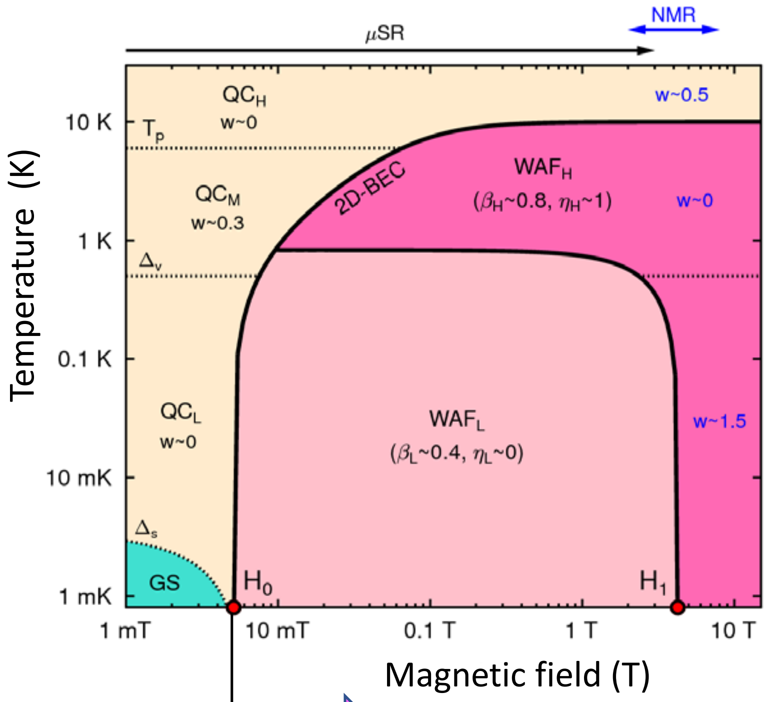
No magnetic order ⇩ **QSL！**

Magnetic order ⇩ AFM

Behavior of the QSL candidate is not simple.

B-T phase diagram

Pratt et al. Nature (2011)



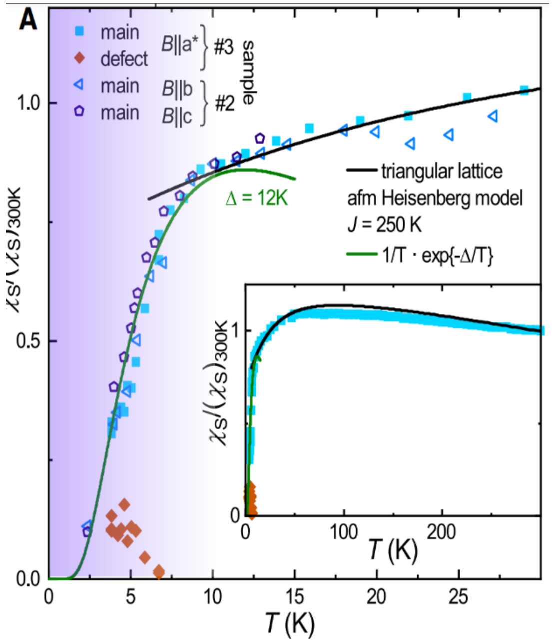
Field-induced inhomogeneous moments

~ % μ_B

6 K anomaly in spin degrees of freedom; ESR and NMR

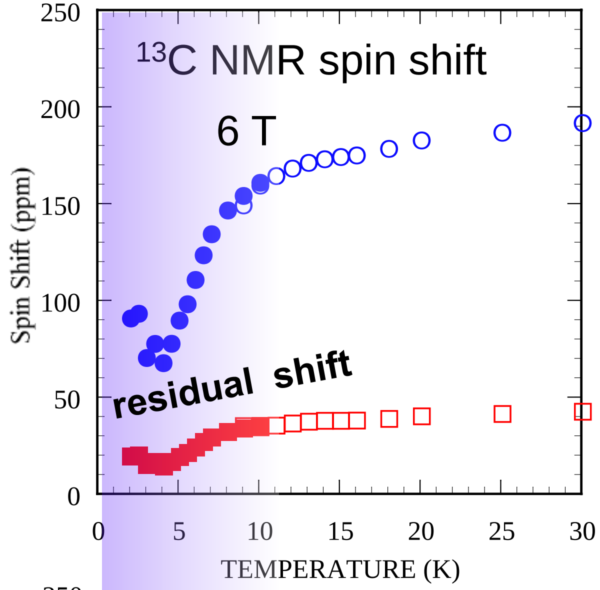
EPR

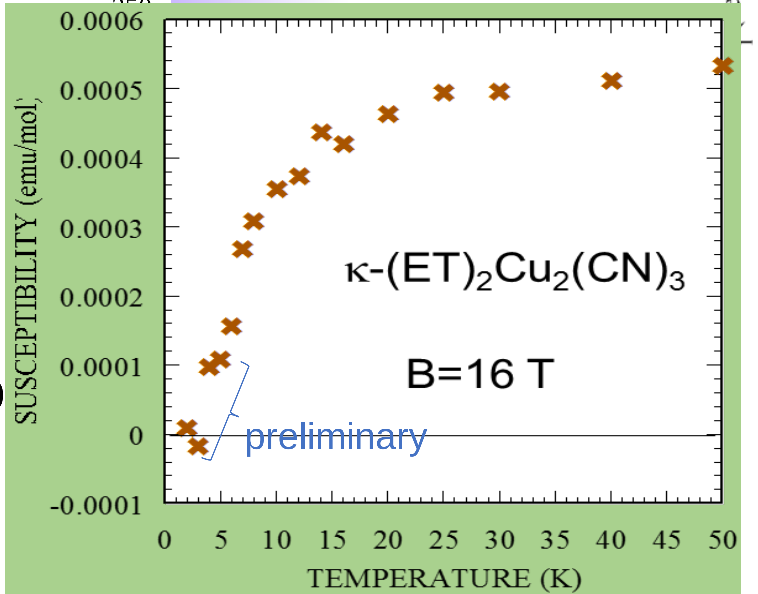
Paramagnetic spins die



Miksch et al. Science 372, 276 (2021)

NMR spectra

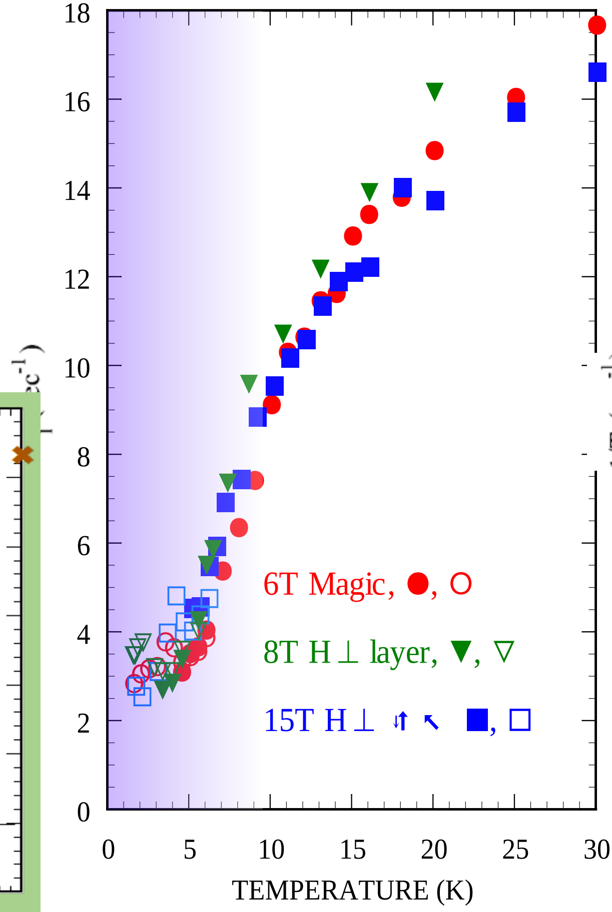


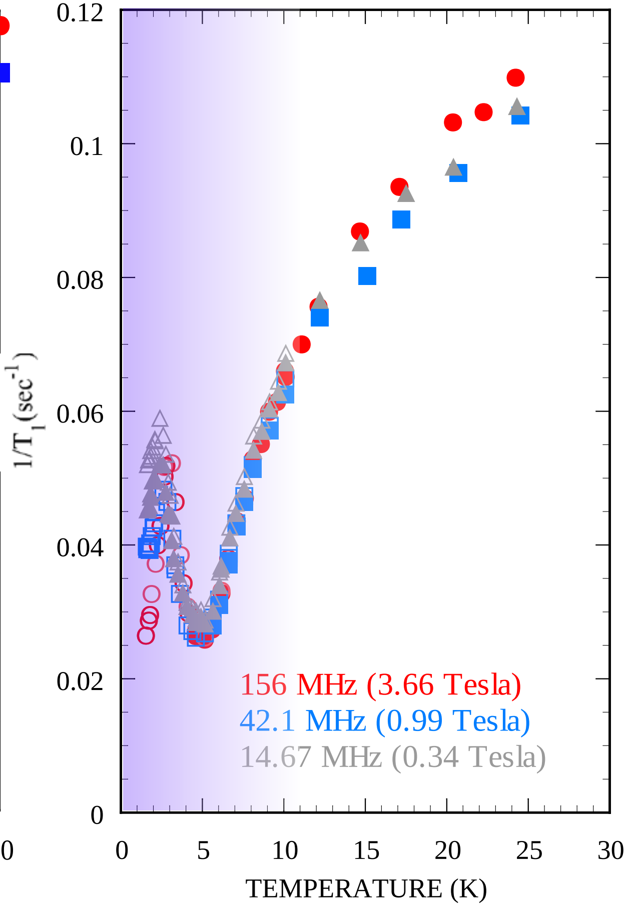


unpublished

NMR relaxation rate

Field-insensitive above 5 K

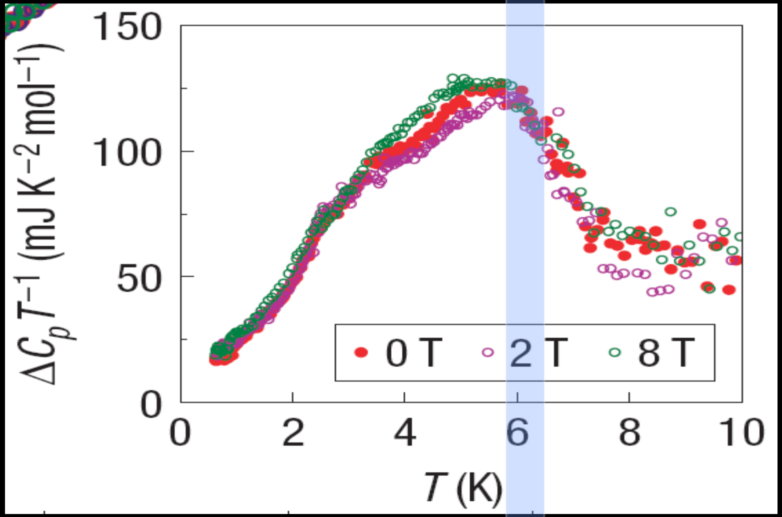




6K-anomaly in κ-(ET)$_2$Cu$_2$(CN)$_3$

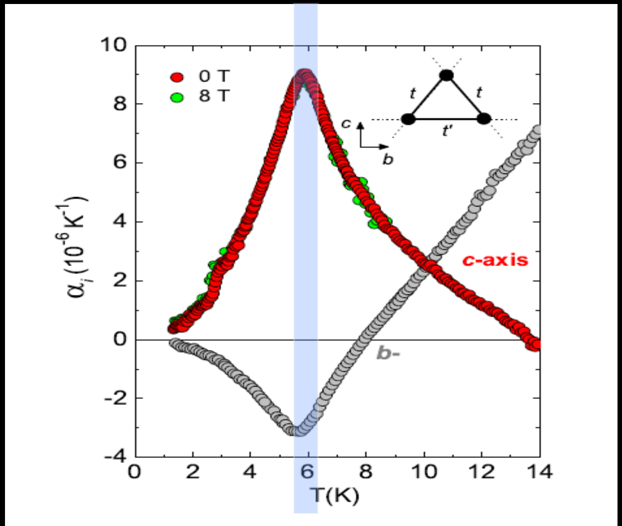
Specific heat

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



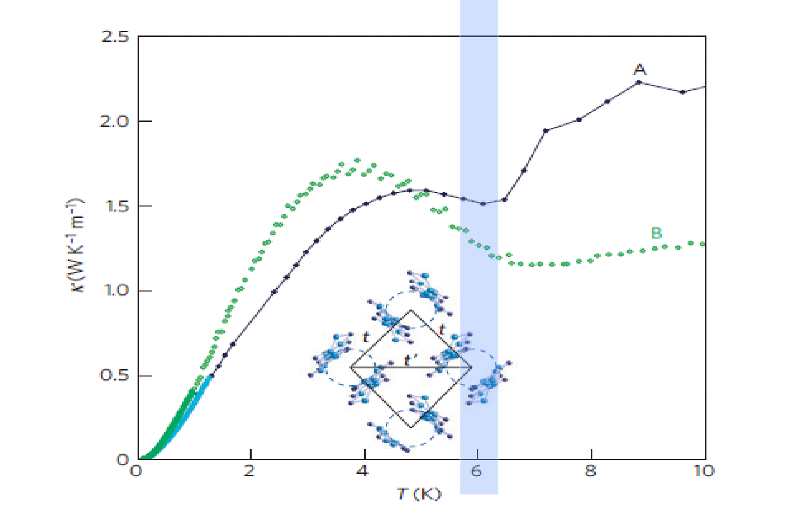
Thermal expansion coefficient

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



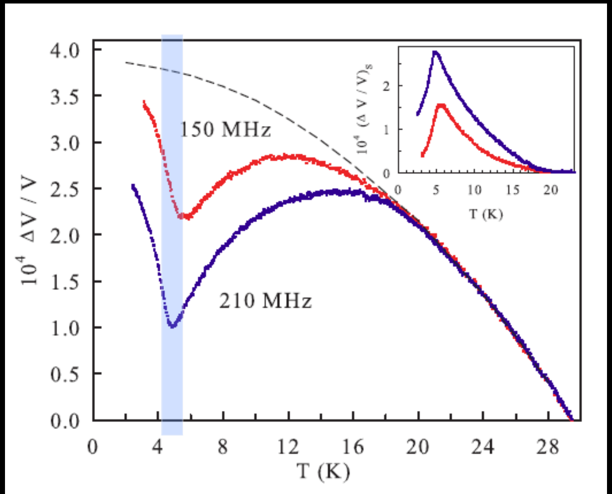
Thermal conductivity

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



Ultrasound velocity

Poirier et al.,



To make Mott-localized electrons mobile,

pressurize the system

allow double occupancies

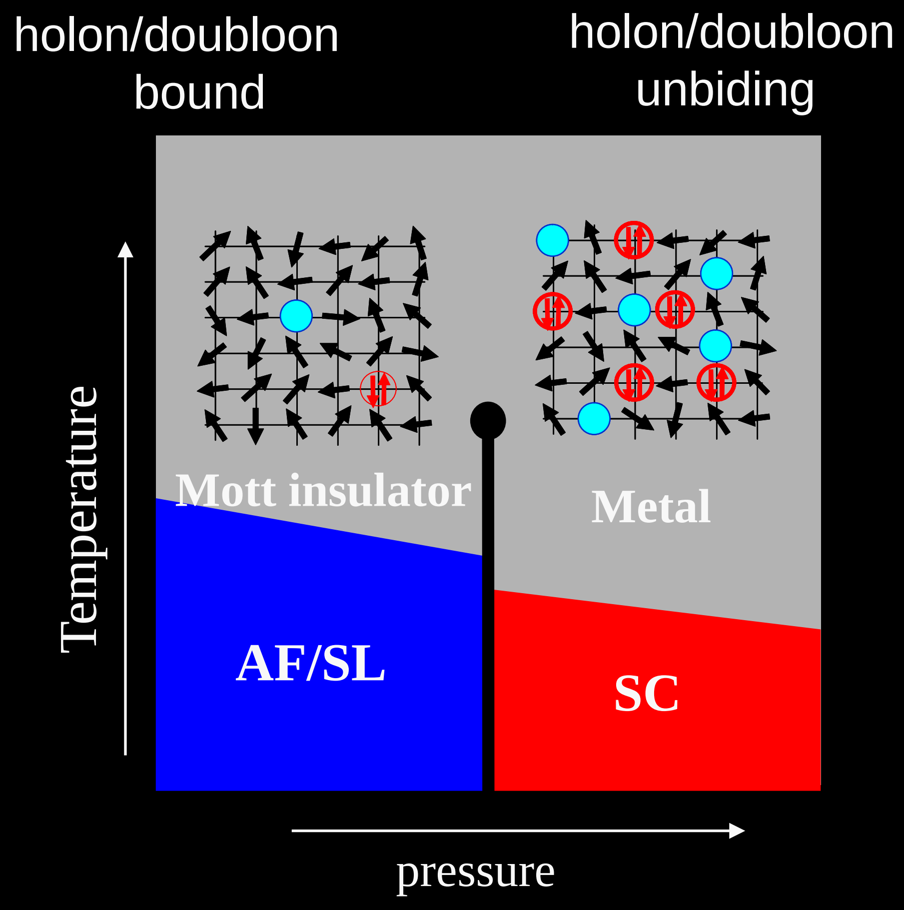
dope the system

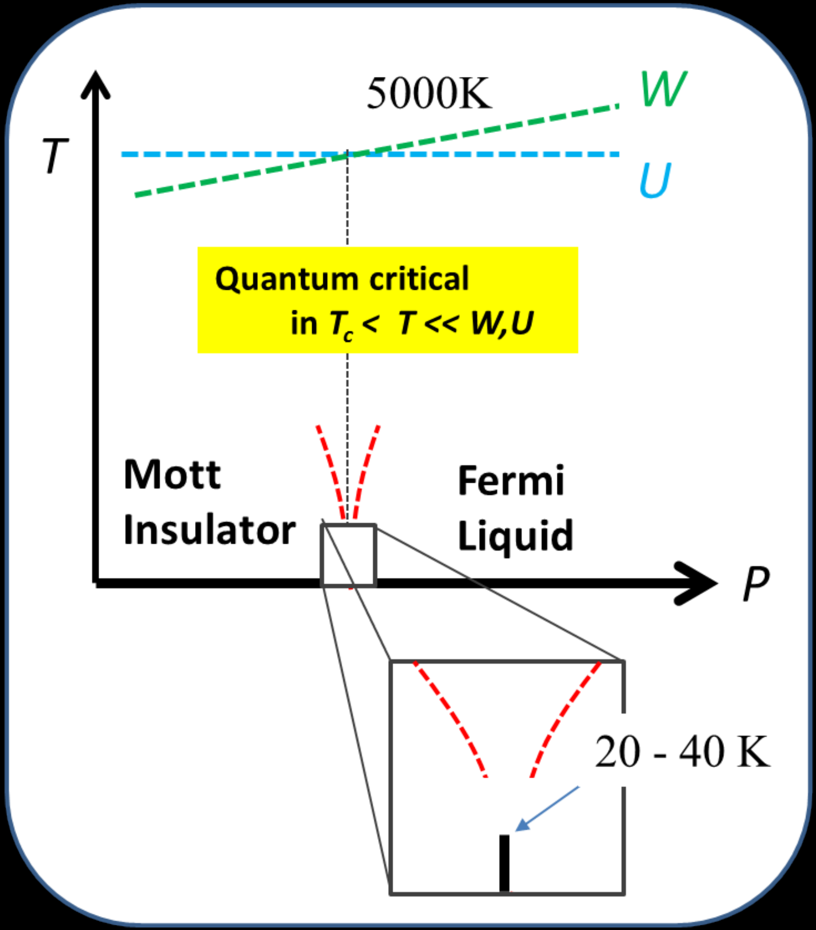
make vacancies

Mott metal-insulator transition

- competition between U and W -

Mott phase diagram





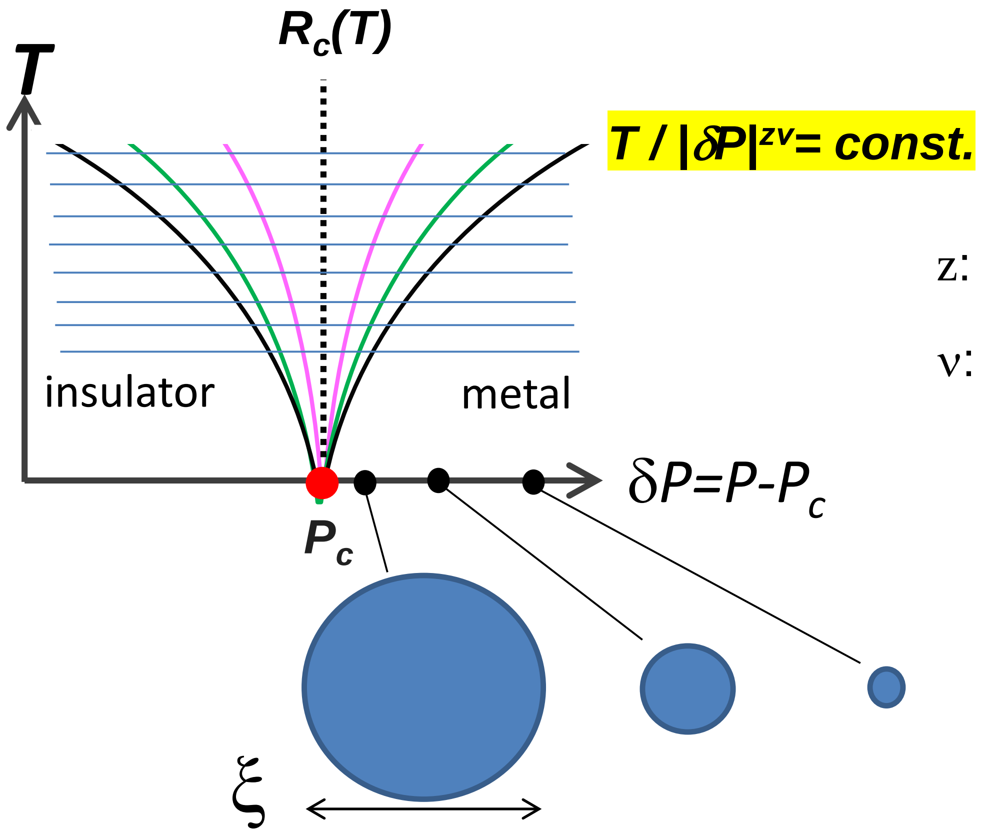
Quantum critical scaling

Time scale of fluctuations

- Quantum fluctuations $\xi_\tau = \xi^z = |\delta P|^{-z\nu}$
- Thermal fluctuations $L_\tau = \hbar / k_B T$

$$R(T,\delta P)/R_c(T) \rightarrow f(\xi_\tau / L_\tau) = f(T/|\delta P|^{z\nu})$$

One-parameter scaling



z: dynamical critical exponent

ν: critical exponent of correlation length

correlation length

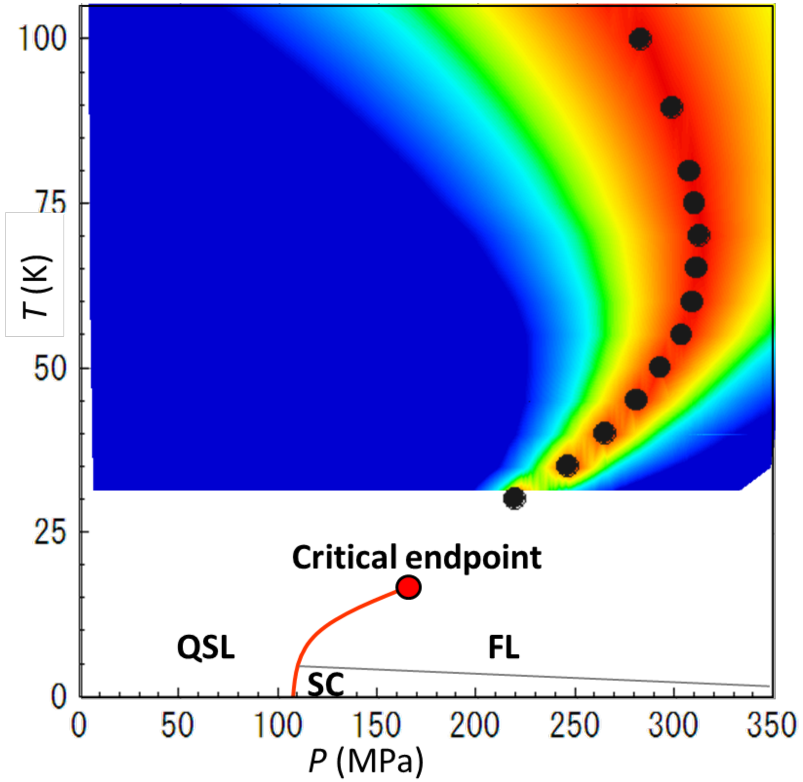
$\xi = |\delta P|^{-\nu}$

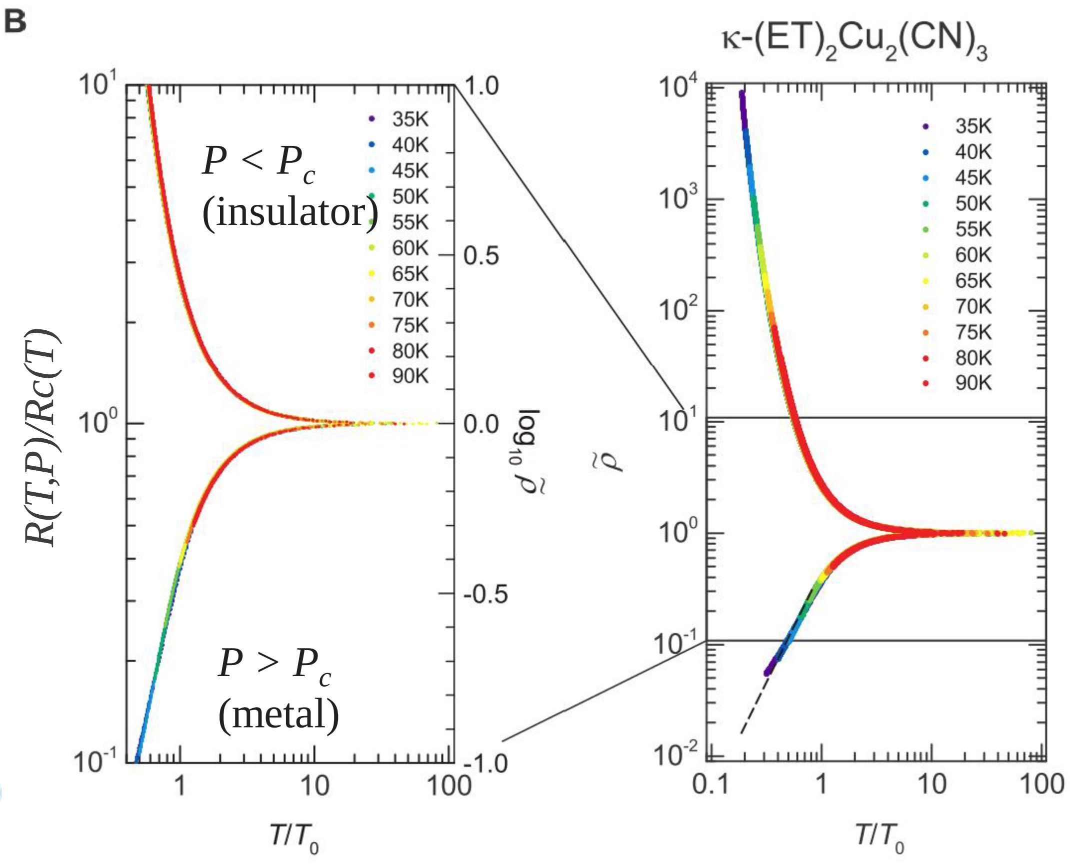
Quantum correlation time

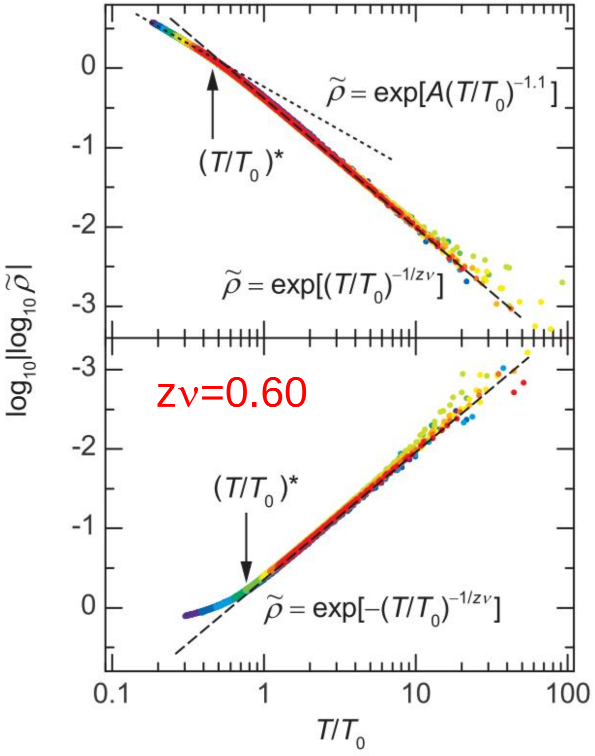
$\xi_\tau = \xi^z = |\delta P|^{-z\nu}$

Phase diagram & quantum critical scaling

Insulator branch







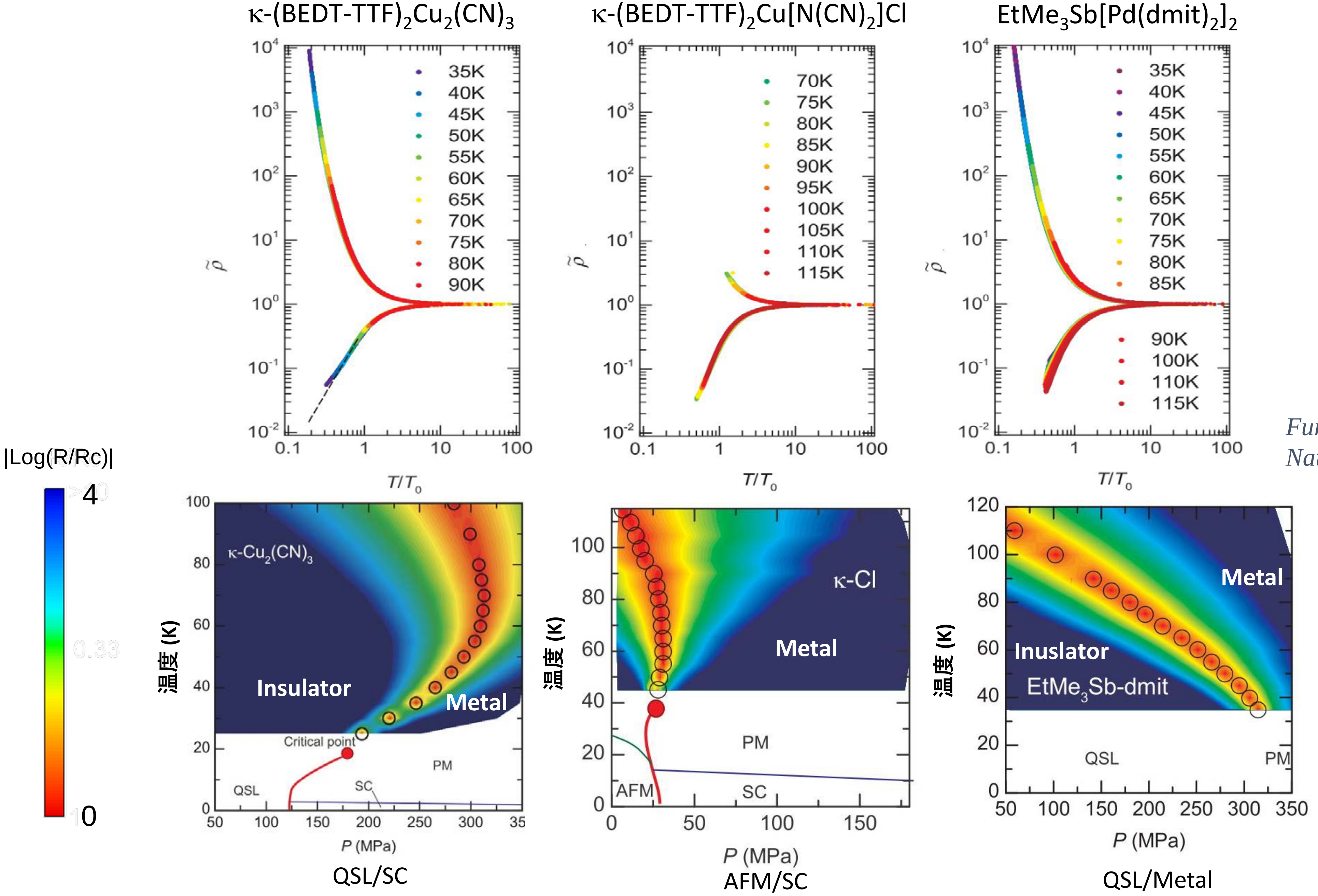
Metal branch

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

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

$$T/T_0 = T\,/\,(c\,|P\text{-}P_c(T)|^{z\nu=0.60}\,)$$

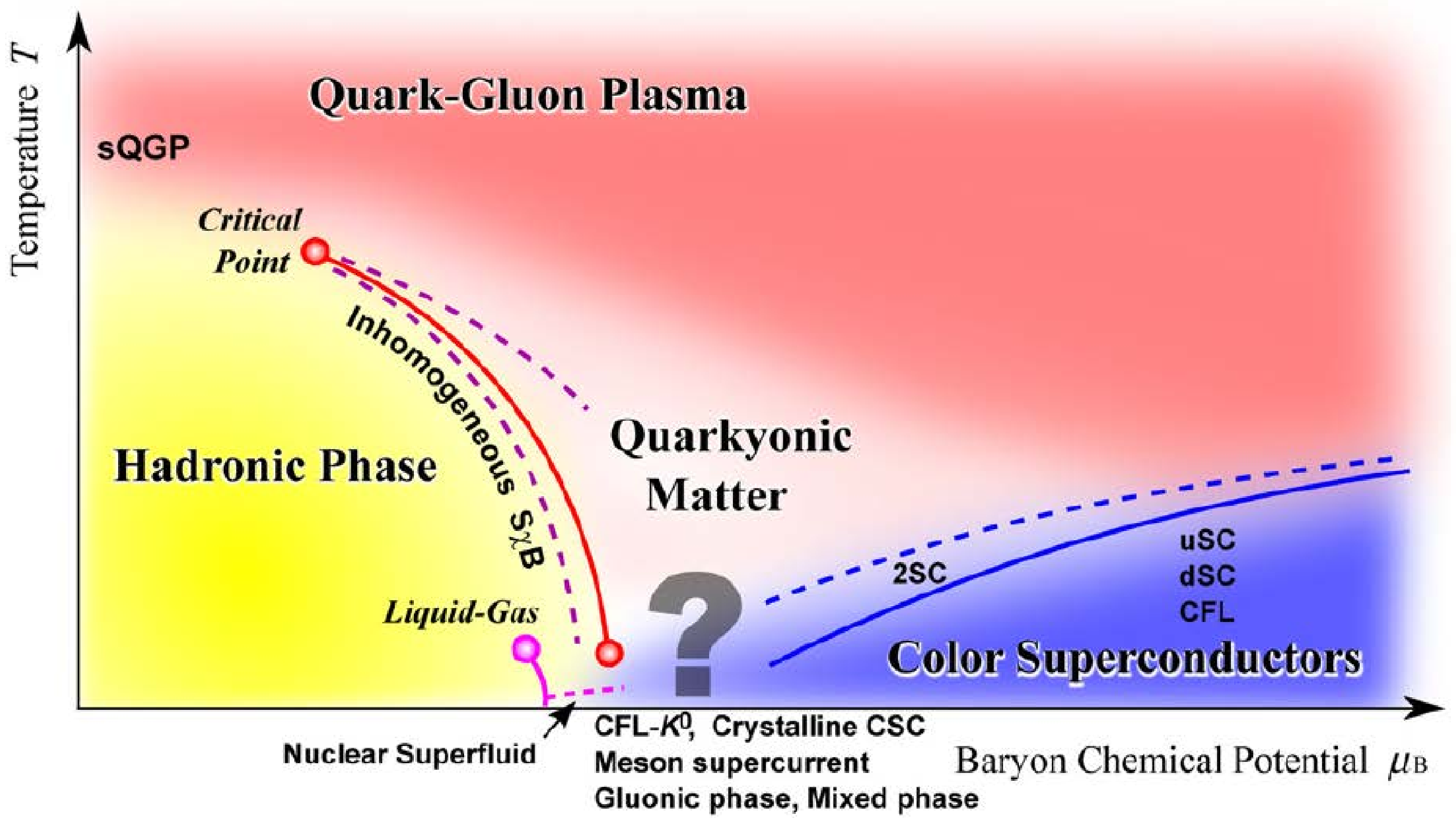
Material-independent universal quantum critical fluid (zν=0.5-0.7)



Holographic duality AdS/CFT

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

Phase diagram of QCD



Fukushima and Hatsuda, Rep. Prog. Phys. 74 (2011) 014001

Two ways to make Mott-localized electrons mobile,

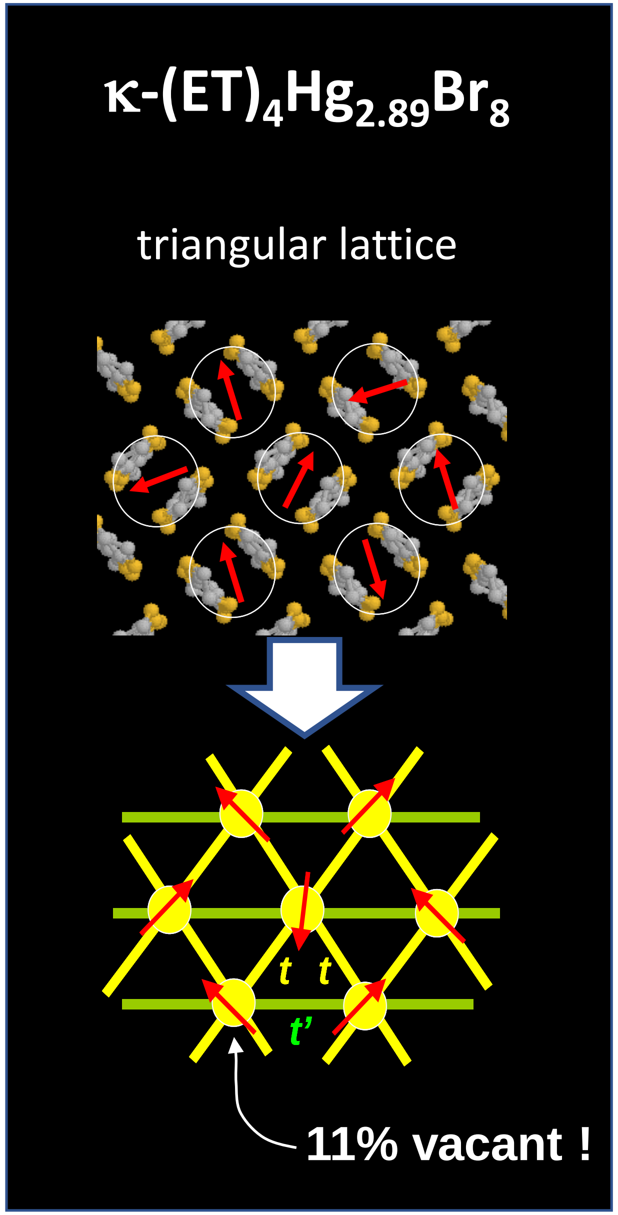
pressurize the system

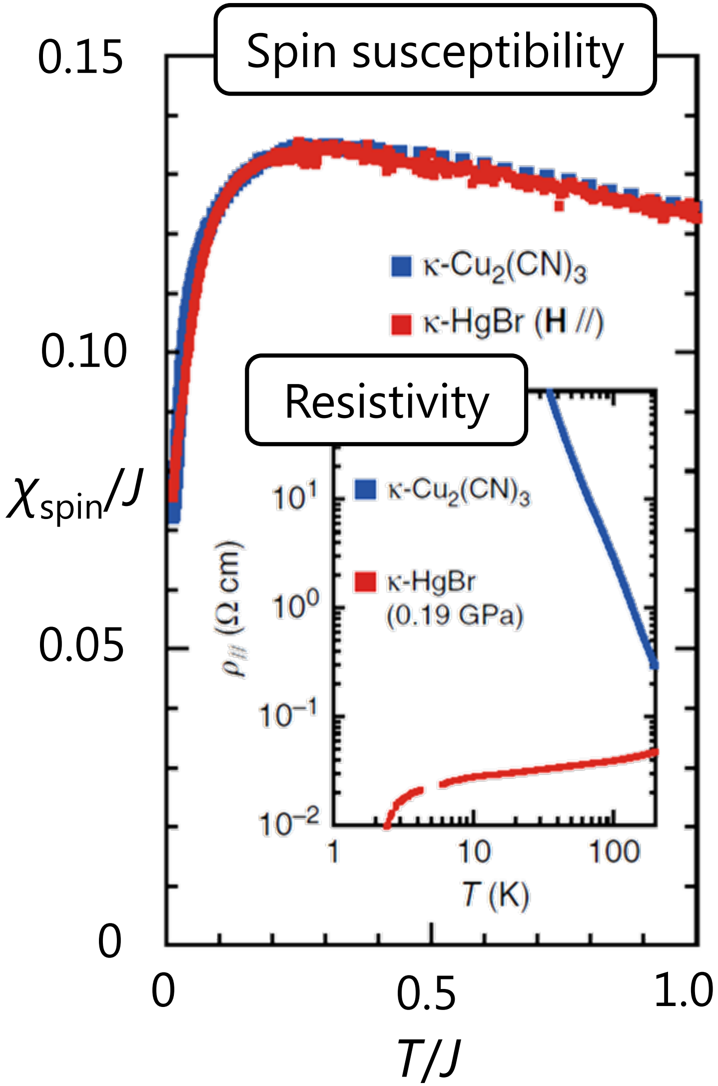
allow double occupancies

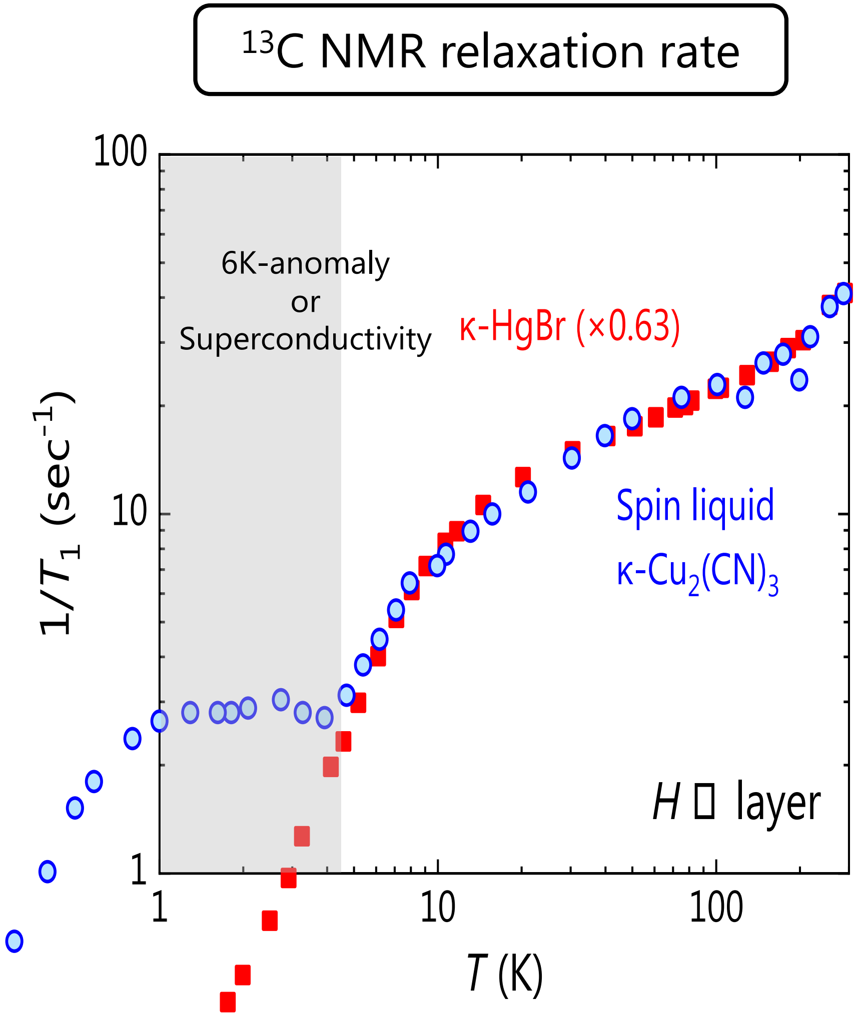
dope the system

make vacancies

Doped spin liquid candidate κ-$(ET)_4Hg_{2.89}Br_8$ *(11% hole doping)*

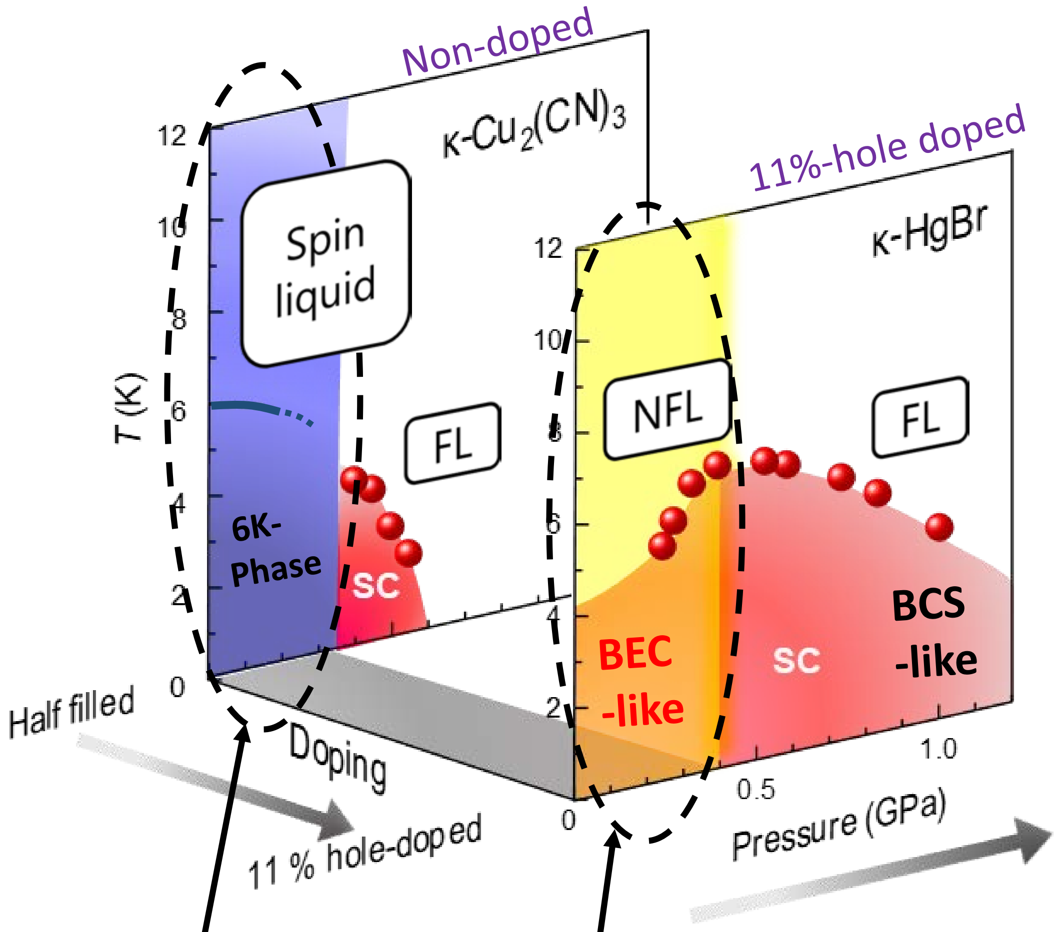






H. Oike, *et al*., Phys. Rev. Lett. 114, 067002 (2015); Nat. Commun. **8**, 756 (2017).

Phase diagram : non-doped and doped spin liquid materials



QSL phase	→	non-Fermi liquid
6-K phase	→	BEC-like SC

Suzuki et al, Phys RevX 12_011016 (2022)

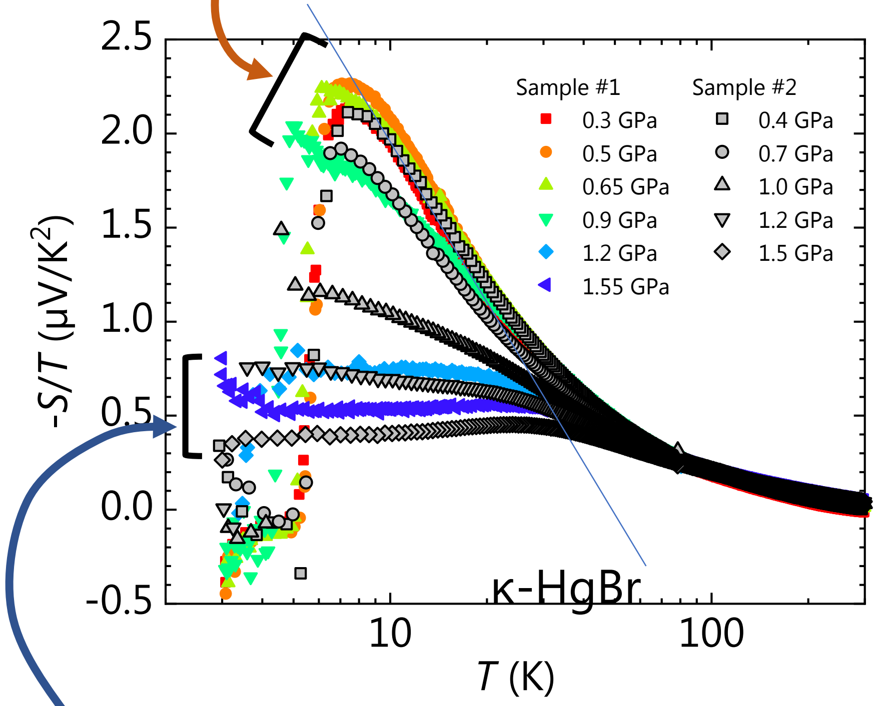
Quantum critical "*phase*"

Wakamatsu et al., arXiv:2201.10714

Seebeck coefficient S (∇T//c-axis)

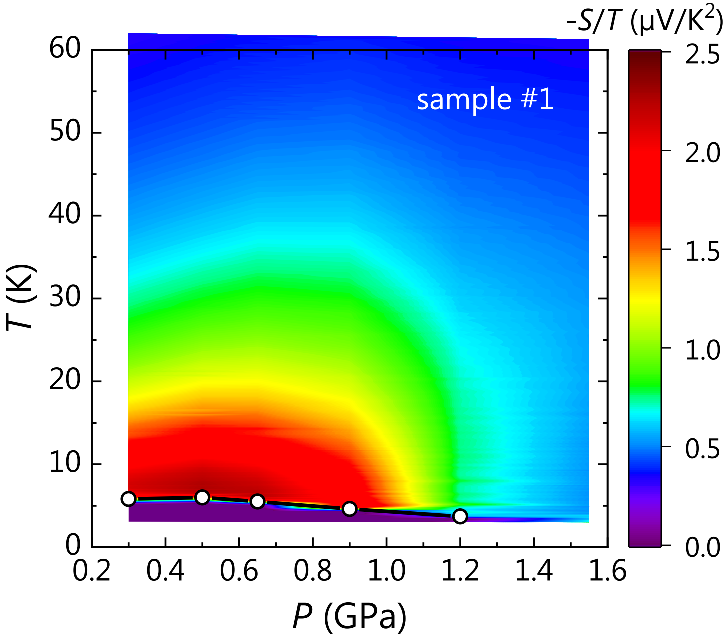
At lower pressures

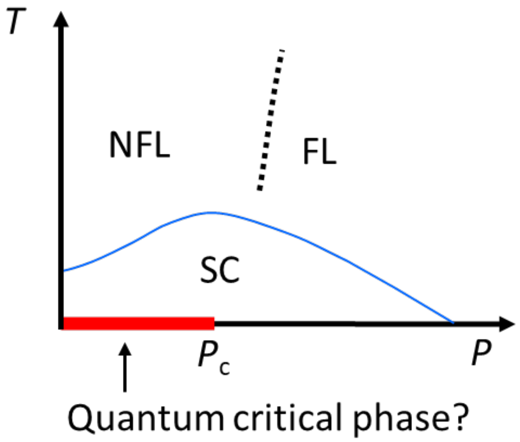
***S/T* ∝ ln *T* → quantum criticality**



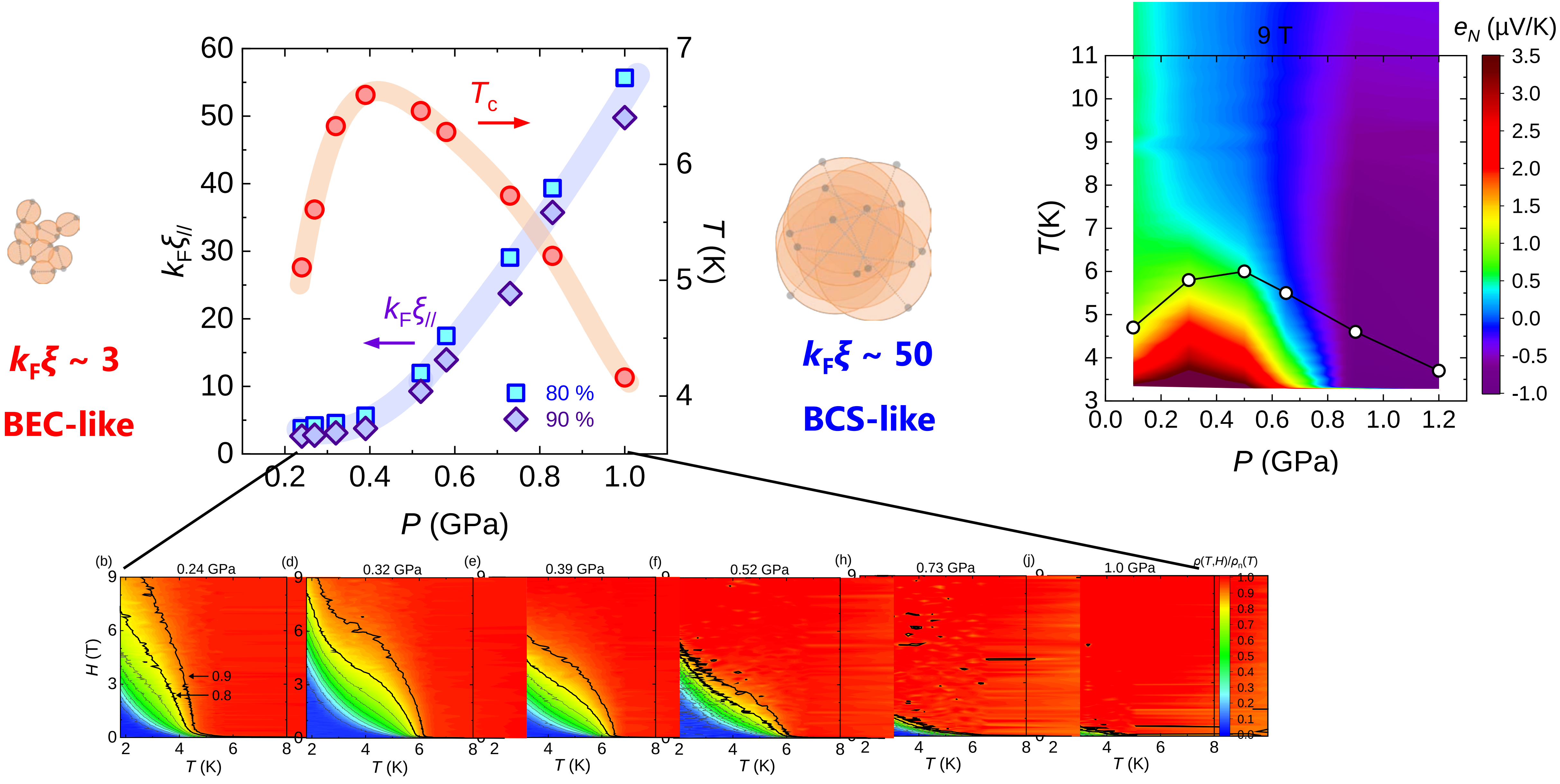
At higher pressures

***S/T* = const. → Fermi liquid**





BEC to BCS crossover in a doped QSL candidate κ-$(ET)_4Hg_{2.89}Br_8$



Suzuki et al, Phys RevX 12_011016 (2022)

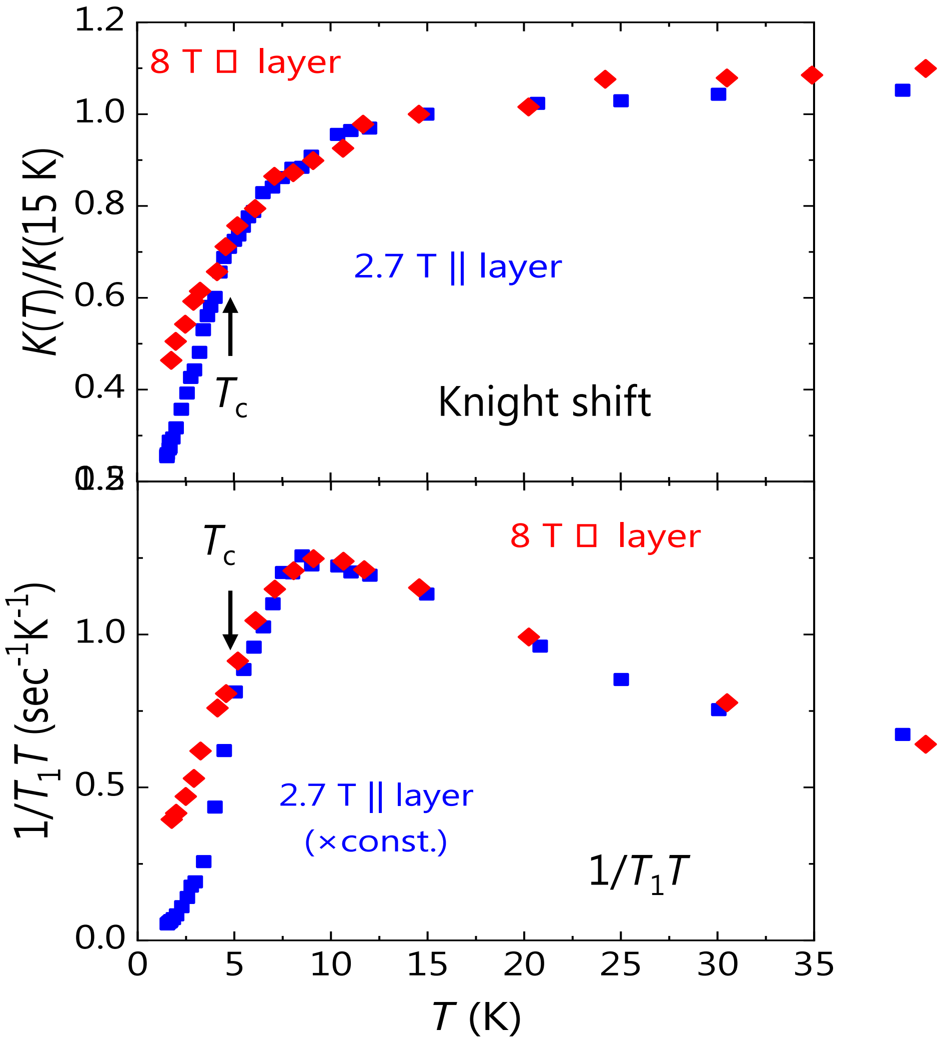
cf. field-robust SC: Imajo et al., Phys. Rev. Res. 3, 033026 (2021).

Preformed Cooper pairs in a doped QSL candidate, κ-$(ET)_4Hg_{2.89}Br_8$

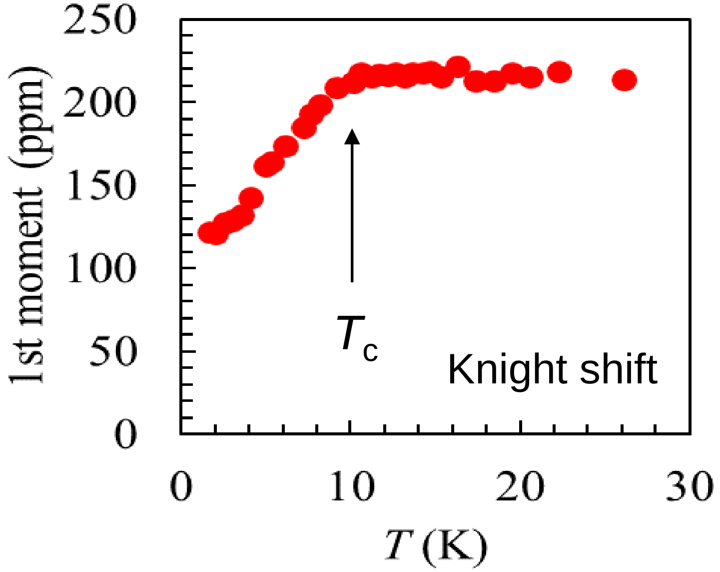
Superconductivity = (Cooper pairing)×(BEC)

unpublished

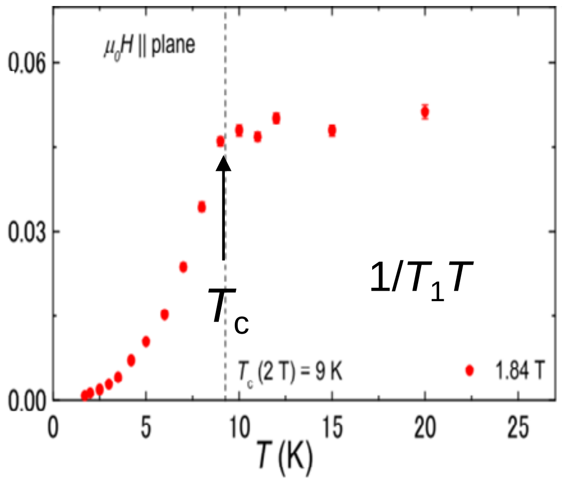
κ-$(ET)_4Hg_{2.89}Br_8$



κ-$(ET)_2Cu(NCS)_2$

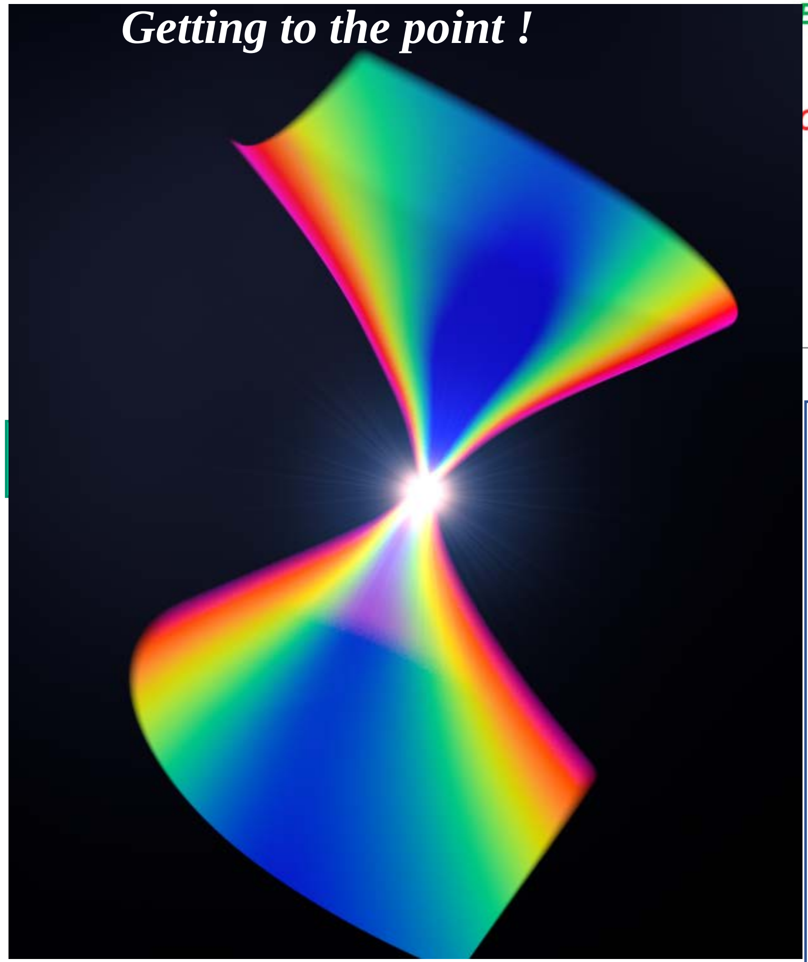


K. Miyagawa *et al.*, Chem. Rev. **104**, 5635 (2004).

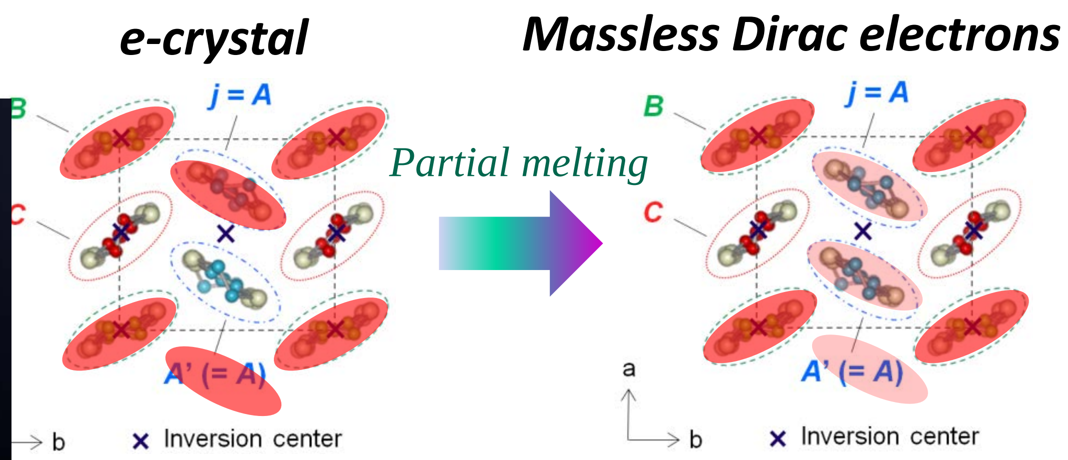


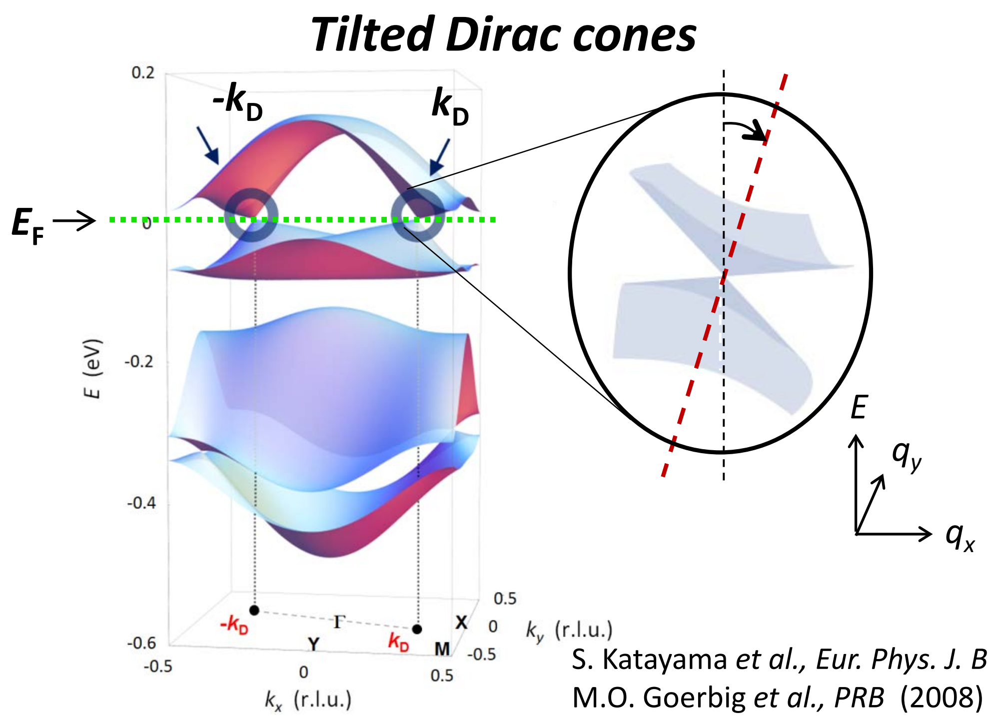
T. Kobayashi *et al.*, Phys. Rev. B. **89**, 165141 (2014).

Organic Conductor α-(BEDT-TTF)$_2$I$_3$



Transport exp.
Tajima et al.

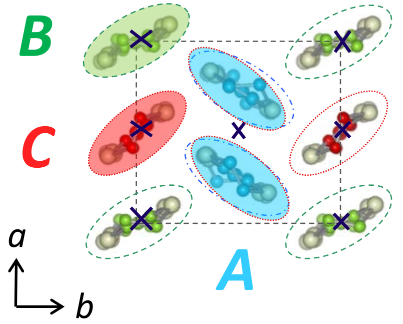


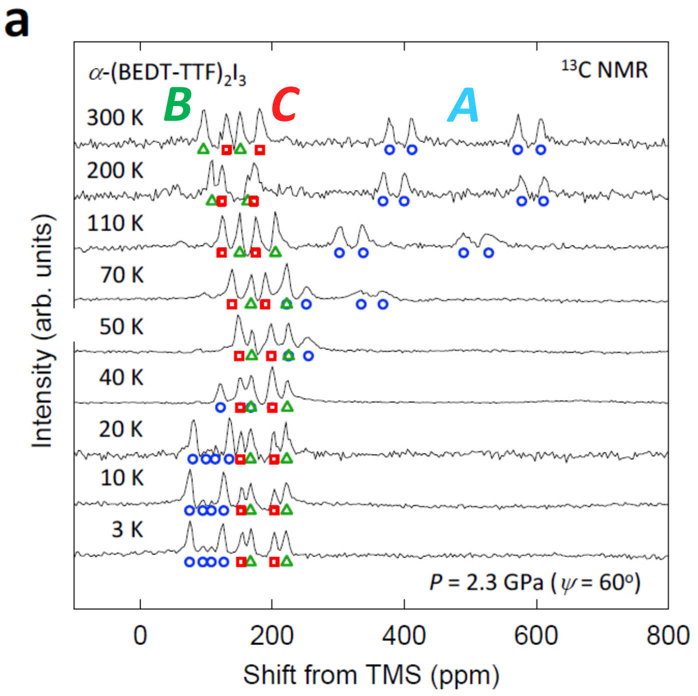


S. Katayama *et al.*, *Eur. Phys. J. B* (2009)
M.O. Goerbig *et al.*, *PRB* (2008)
K. Kajita *et al.*, *JPSJ* (2014)

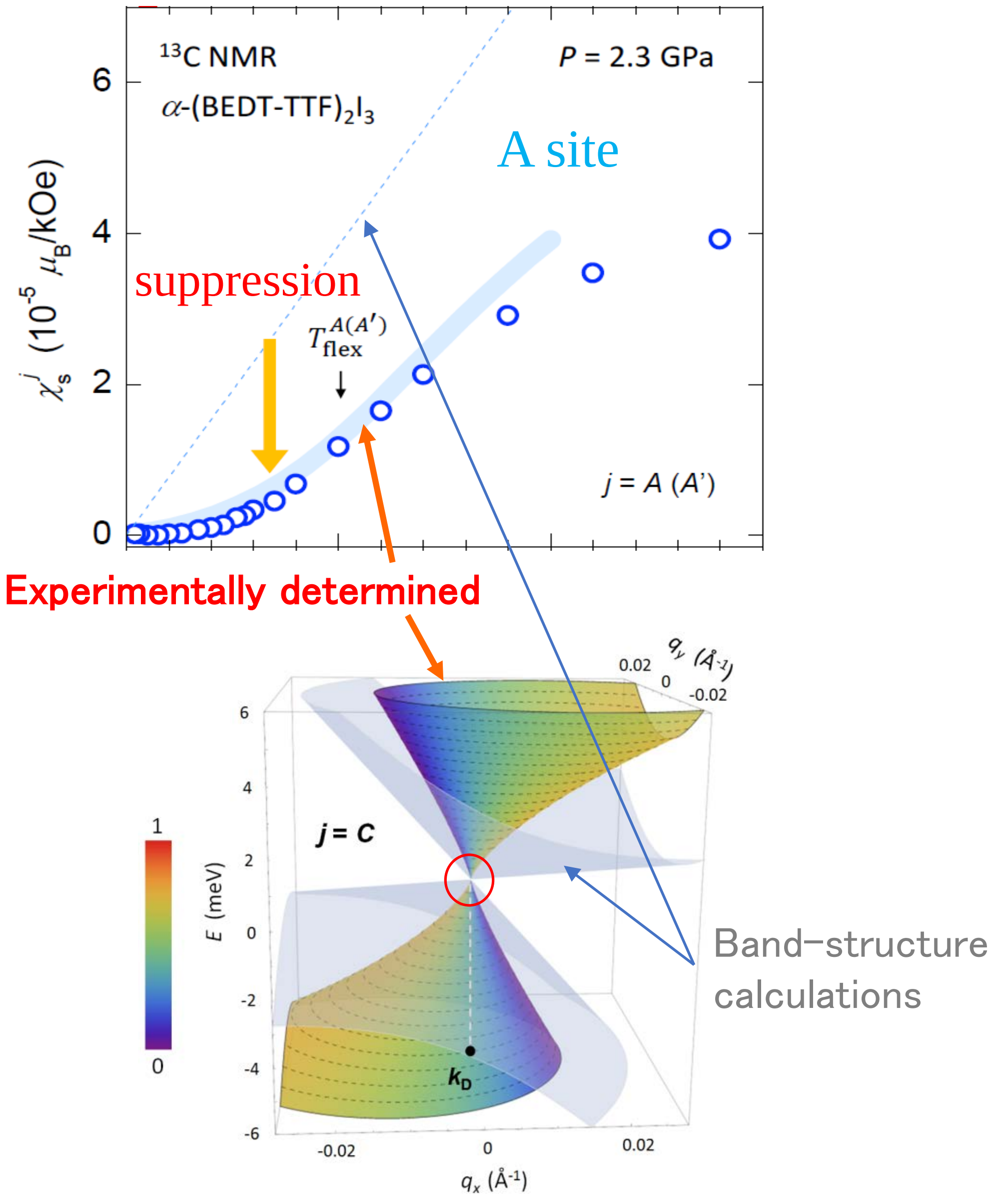
Site-selective ^{13}C-NMR probes reshaping of Dirac cone

Hirata et al., Nat. Commun. **7**, 12666 (2016)



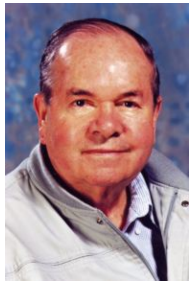


Spin susceptibility deduced from ^{13}C Knight shift



Unscreened Long-range Coulomb Interactions

***Logarithmic divergence* of v_F at low energies**



Isotropic tilted cone

$$v(k) = v\left(1 + \frac{\alpha}{4}\ln(\Lambda/k)\right), \quad \Lambda/k \gg 1$$

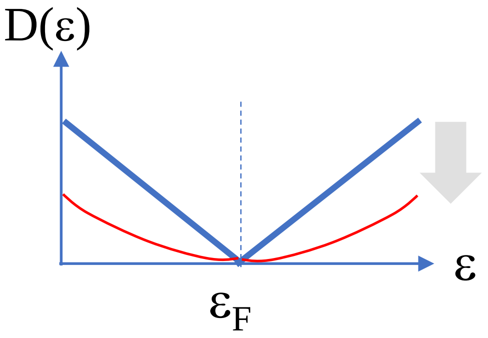
A. A. Abrikosov *et al.*, *JETP* **32** (1971)

Anisotropic tilted cone

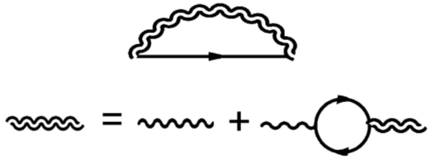
$$E_\zeta(\boldsymbol{q}) = \hbar\left(\boldsymbol{w}_0 \cdot \boldsymbol{q} + \zeta\sqrt{v_x{}^2 q_x{}^2 + v_y{}^2 q_y{}^2}\right)$$

S. Katayama *et al.*, *Eur. Phys. J. B* **67** (2009)

Massless Dirac electrons



RPA self energy



cf. Isobe & Nagaosa, *JPSJ* **81** (2012): using HF exchange term 

RG equations (leading order in 1/*N*; *N* >> 1)

$$\frac{1}{v_x}\frac{dv_x}{dl} = \frac{8}{\pi^2 N}\int_0^{2\pi}\frac{d\varphi}{2\pi} 2\cos^2\varphi\, F(g_\varphi)$$

$$\frac{1}{v_y}\frac{dv_y}{dl} = \frac{8}{\pi^2 N}\int_0^{2\pi}\frac{d\varphi}{2\pi} 2\sin^2\varphi\, F(g_\varphi)$$

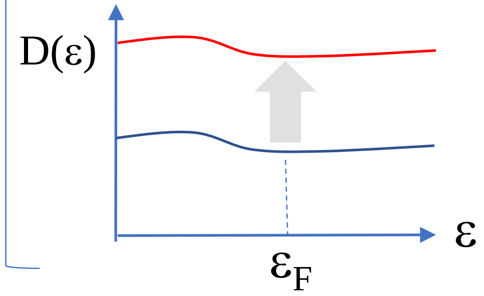
$N = 4$: fermion flavor, $\boldsymbol{q} = (\boldsymbol{k} - \boldsymbol{k}_D) = q(\cos\varphi, \sin\varphi)$

$l = \ln(\Lambda/q)$

$F(g_\varphi) = \left(-\pi/2 + g_\varphi + \arccos g_\varphi/\sqrt{1 - g_\varphi{}^2}\right)/g_\varphi$

$g_\varphi = 2\pi e^2 N/16\,\varepsilon\sqrt{v_x{}^2\sin^2\varphi + v_y{}^2\cos^2\varphi}$

Conventional metal



Notion of running coupling constant

mass generation by interactions

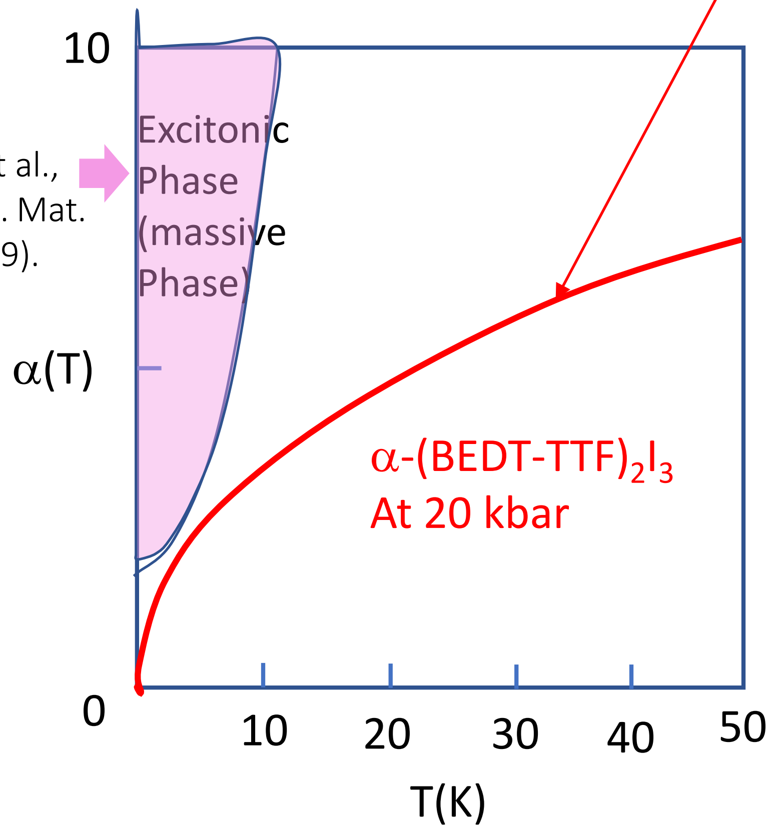
$$\alpha = \frac{E_C}{E_K} = \frac{e^2/\varepsilon r}{v_F \hbar k_F} \propto \frac{e^2/\varepsilon r}{v_F \hbar / r} = \frac{e^2}{\hbar v_F \varepsilon}$$

$$v_F(T) = v_F{}^0 \left[1 + \frac{\alpha^0}{4} \ln\left(\frac{\Lambda}{T}\right) \right]$$

$$\alpha(T) = \frac{e^2}{\hbar v_F(T)\varepsilon} = \frac{\alpha^0}{1 + (\alpha^0/4)\ln(\Lambda/T)}$$

Coulomb for Weyls

Khveshchenko et al., J. Phys. Condens. Mat. 21, 075303 (2009).

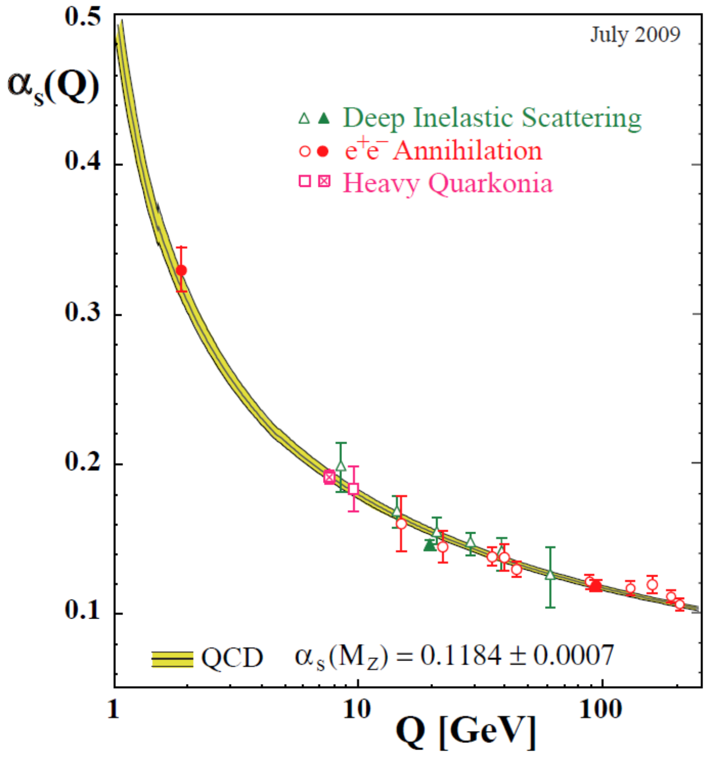


$D(E_F)V(\boldsymbol{q})$

$V(\boldsymbol{q}) \propto 1/|\boldsymbol{q}|$

$0 \times \infty \rightarrow ?$

QCD: strong interactions



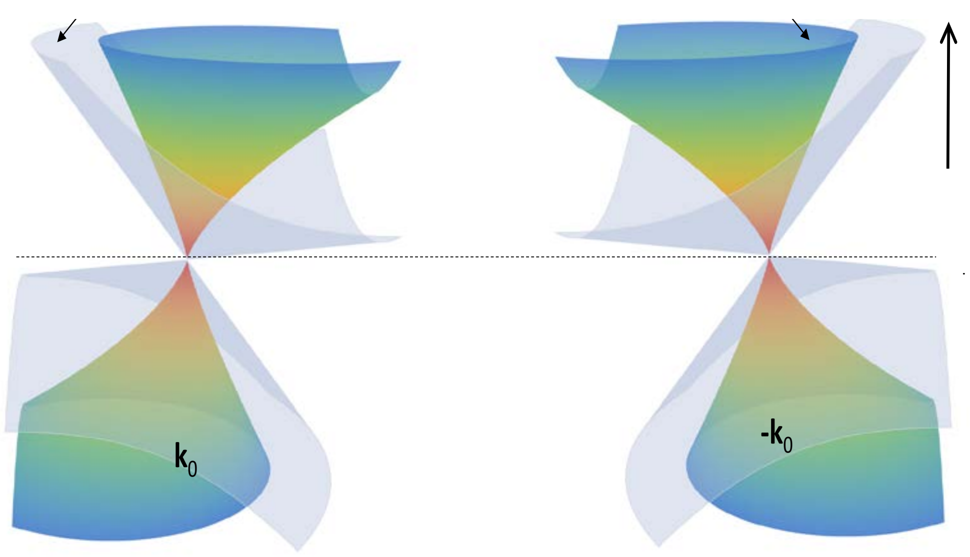
Eur.Phys.J.C64:689,2009

平田、小林（名大）

Strongly correlated Massless Dirac electrons

Dirac cone reshaping

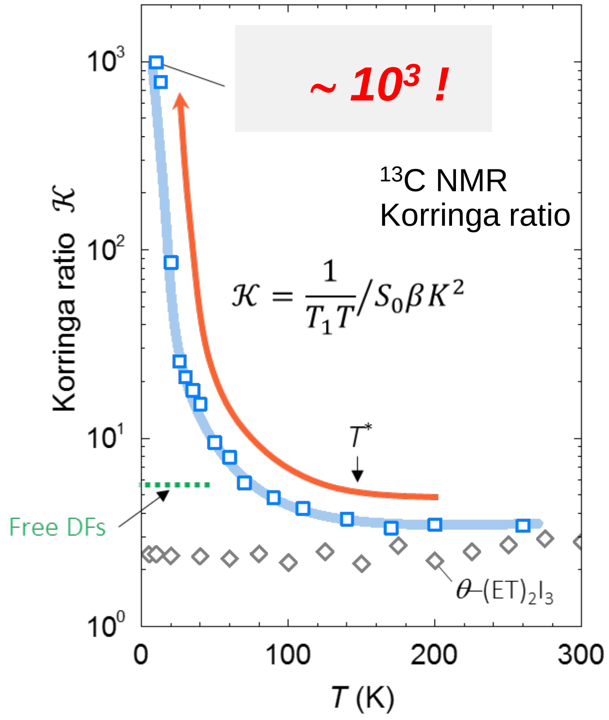
Logarithmic velocity renormalization



Hirata et al., Nat. Commun. **7**, 12666 (2016)

Diverging Korringa ratio

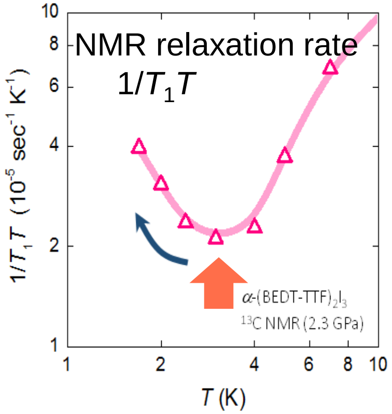
Unusual spin correlations

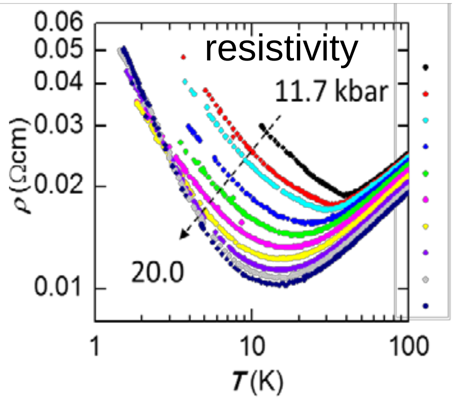


Hirata et al., Science **358**, 1403 (2017)

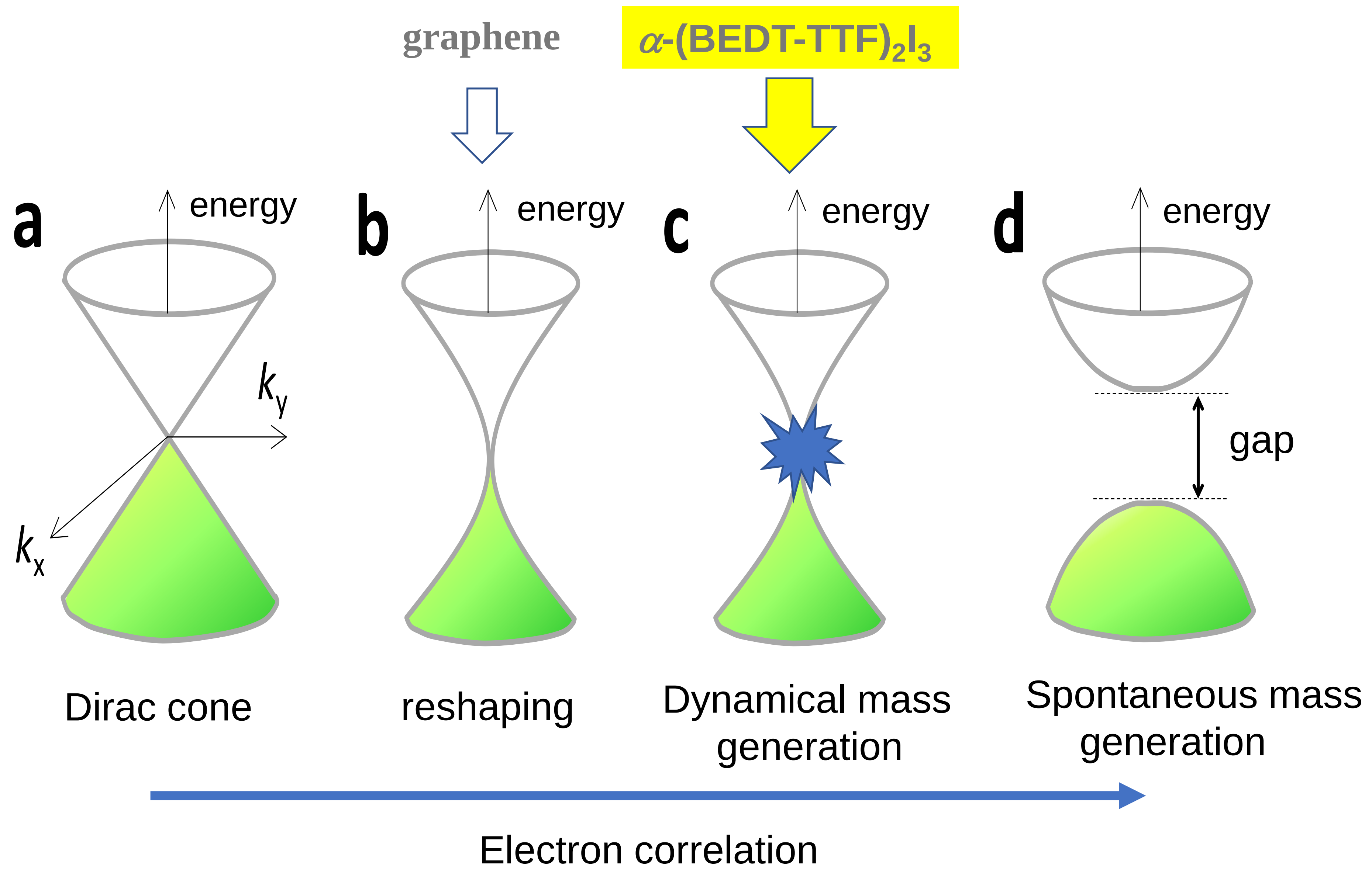
Low-T anomalies in spin and charge

Incipient excitonic instability





Liu et al., PRL. **116**, 226401 (2016)


graphene
α-(BEDT-TTF)2I3
a
energy
ky
kx
b
energy
c
energy
d
energy
gap
Dirac cone
reshaping
Dynamical mass generation
Spontaneous mass generation
Electron correlation

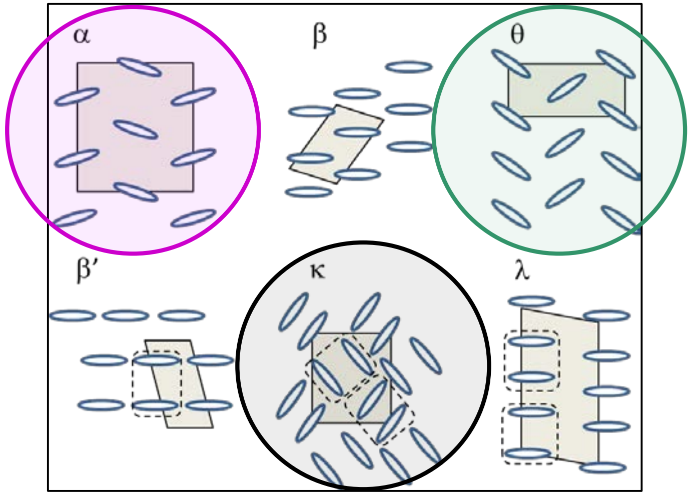
summary

Diverse manifestations of electron correlation In various molecular arrangements

Massless Dirac electrons

Dirac cone reshaping
Unusual spin correlation
Dynamic mass generation

Particle physics



Electronic Xtal/glass

Non-equilibrium
Slow dynamics
Crystal growth
Quantum glass

Soft matter physics

Mott physics

Quantum Mott criticality
Preformed Cooper pairs
Spin liquid
BEC-BCS crossover

これらすべての物理は一つの分子ETの配列の違いのみから生まれる

全ては繋がっている

Today’s talk is based on collaboration with

K. Miyagawa, Kagawa, Shimizu, Kurosaki, Furukawa, H. Oike, M. Urai, Y. Suzuki, K. Wakamatsu, Y. Ueno, T. Fujii, J. Ibuka, T. Sato, H. Murase (U Tokyo)

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