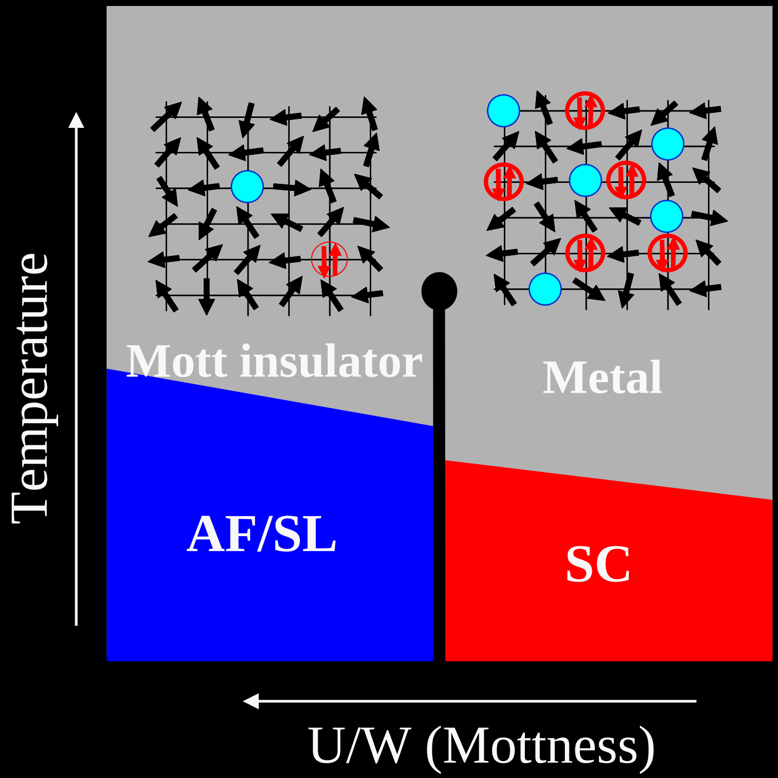
2. Mott transition and superconductivity

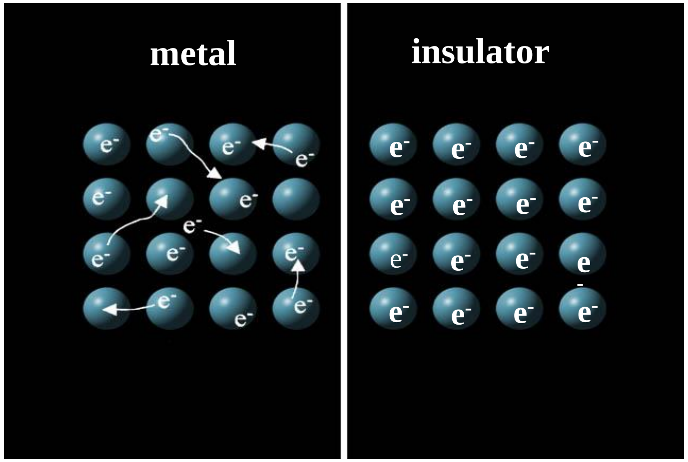
- high-energy universality vs low-energy diversity -



N. Mott (1949)

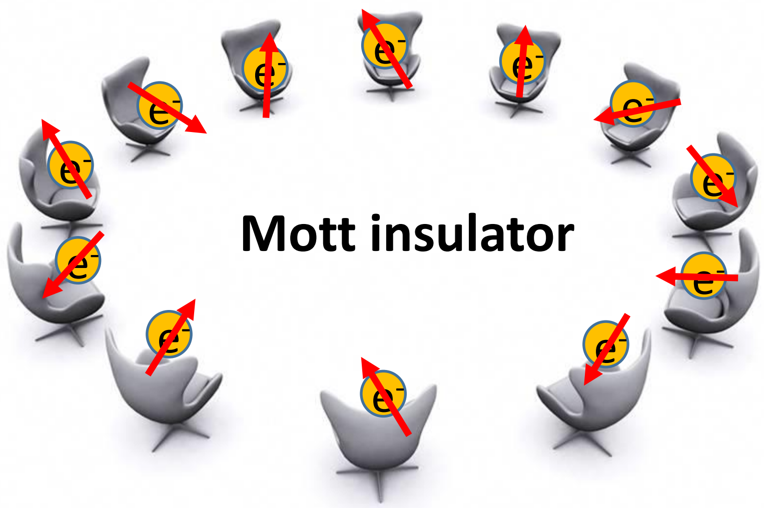
Mott metal-insulator transition

Competition of kinetic energy, ***W***, and Coulomb reputvive energy, ***U***



Wave-like Particle-like

Musical chairs



Double occupancies are allowed or not ?

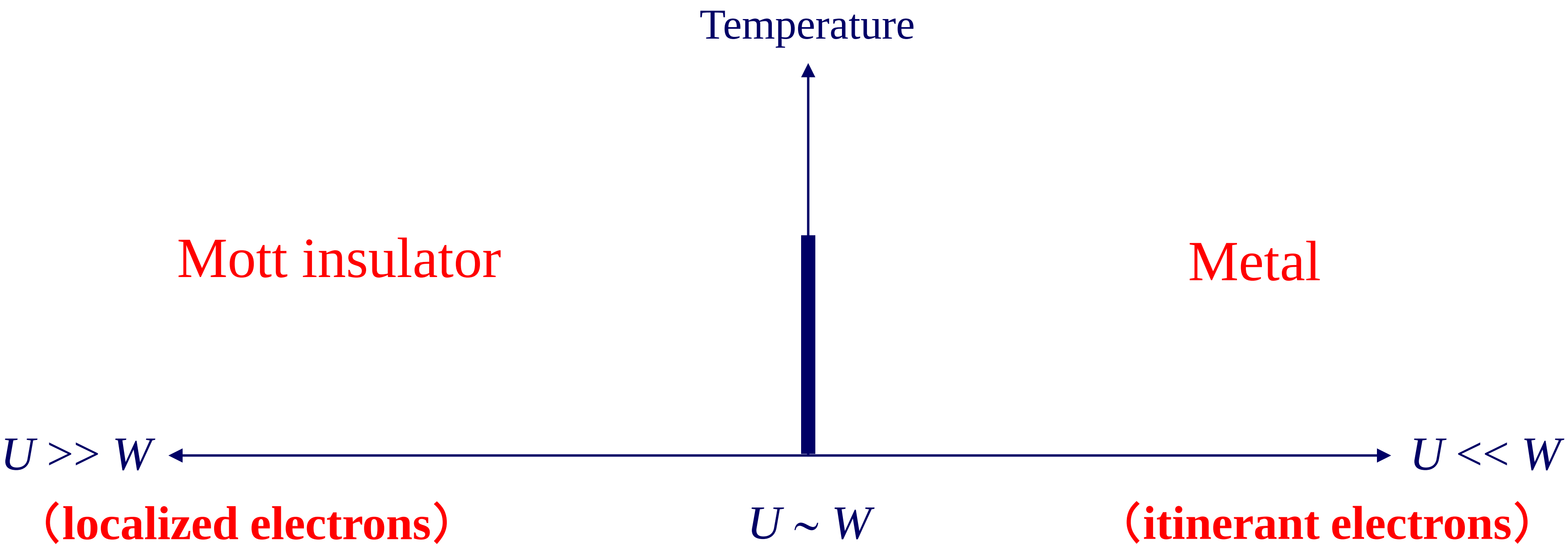
⬇

Metal or insulator

Mott transition

Competition between kinetic energy and Coulomb energy

(W : bandwidth) (U : Coulomb repulsion)



(localized electrons)

particlelike

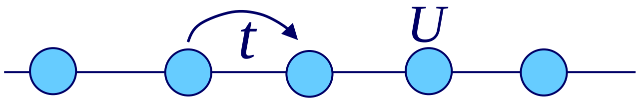
(itinerant electrons)

wavelike

Strongly interacting

Weakly interacting

Hubbard model



$n_{i,\uparrow} = c^*_{i,\uparrow} c_{i.\uparrow}$

$n_{i,\downarrow} = c^*_{i,\downarrow} c_{i.\downarrow}$

In the strong correlation limit, W~2zt << U

Heisenberg Hamiltonian $H = \sum_{(i,j)} J \mathbf{S}_i \mathbf{S}_j \quad (J = 4t^2/U)$

Antiferromagnet
(insulator)

Hubbard Hamiltonian

off-diagonal   diagonal

$$H = \boxed{\sum_{(i,j)\sigma} (t c^+_{i,\sigma} c_{j,\sigma} + h.c.)} + \boxed{U \sum_i n_{i,\uparrow} n_{i,\downarrow}}$$

Wannier   Bloch

$$c_{j,\sigma} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{R}_j} c_{\mathbf{k},\sigma}$$

$$H = \boxed{\sum_{\mathbf{k},\sigma} \varepsilon_{\mathbf{k}} c^+_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma}} + \boxed{\frac{U}{N} \sum_{k_1,k_2,k_3,k_4} c^+_{k_1,\uparrow} c^+_{k_2,\downarrow} c_{k_3,\uparrow} c_{k_4,\downarrow} \delta_{k_1+k_2,k_3+k_4}}$$

diagonal   off-diagonal

In the weak correlation limit, W~2zt >> U

scattering rate $\displaystyle \frac{1}{\tau(k_1)} = \frac{2\pi}{\hbar} \frac{1}{N^2} \sum_{k_2,k'_1,k'_2} U^2 \delta(\varepsilon_{k_1} + \varepsilon_{k_2} - \varepsilon_{k'_1} - \varepsilon_{k'_2}) \delta_{k_1+k_2,k'_1+k'_2} f_{k_2}(1 - f_{k'_1})(1 - f_{k'_2})$

$\propto T^2$ in 3D

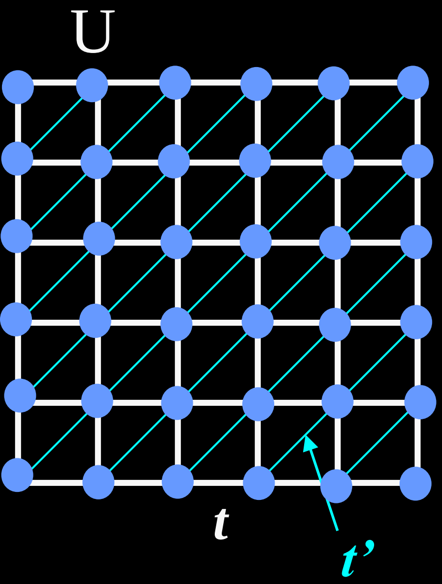
$\propto T^2 \log \frac{\varepsilon_F}{k_B T}$ in 2D

$\boldsymbol{\rho(T) \propto T^2}$
(metal)

Mott transition occurs at *W~U*,
but depends on dimension and lattice geometry

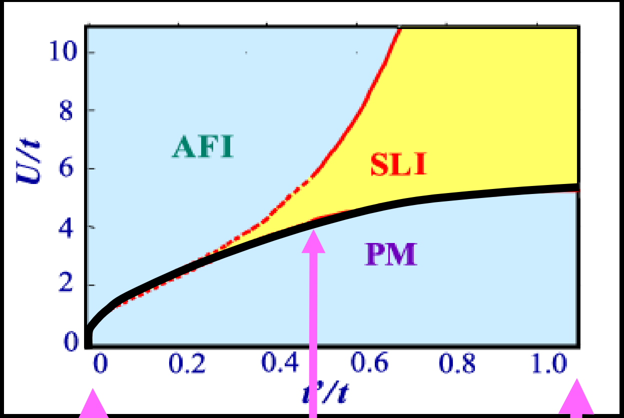
1D ½-filled Hubbard models are always Mott insulators.

2D ½-filled Hubbard model on anisotropic triangular lattice



PIRG

Imada et al. JPSJ (2003)



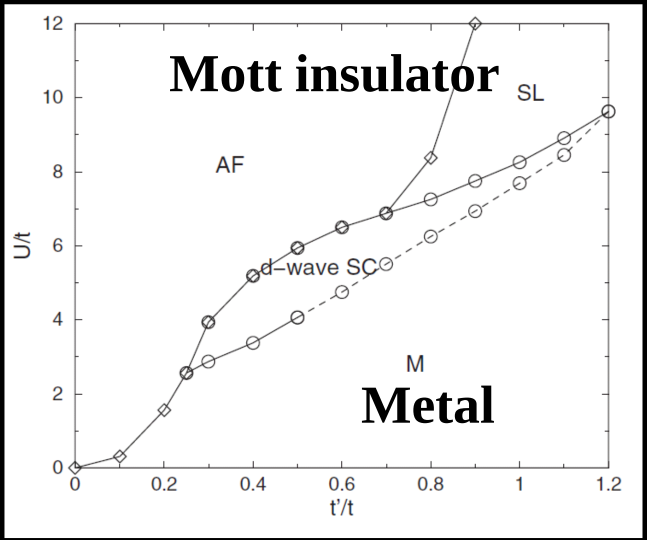
Mott transition line

Square lattice

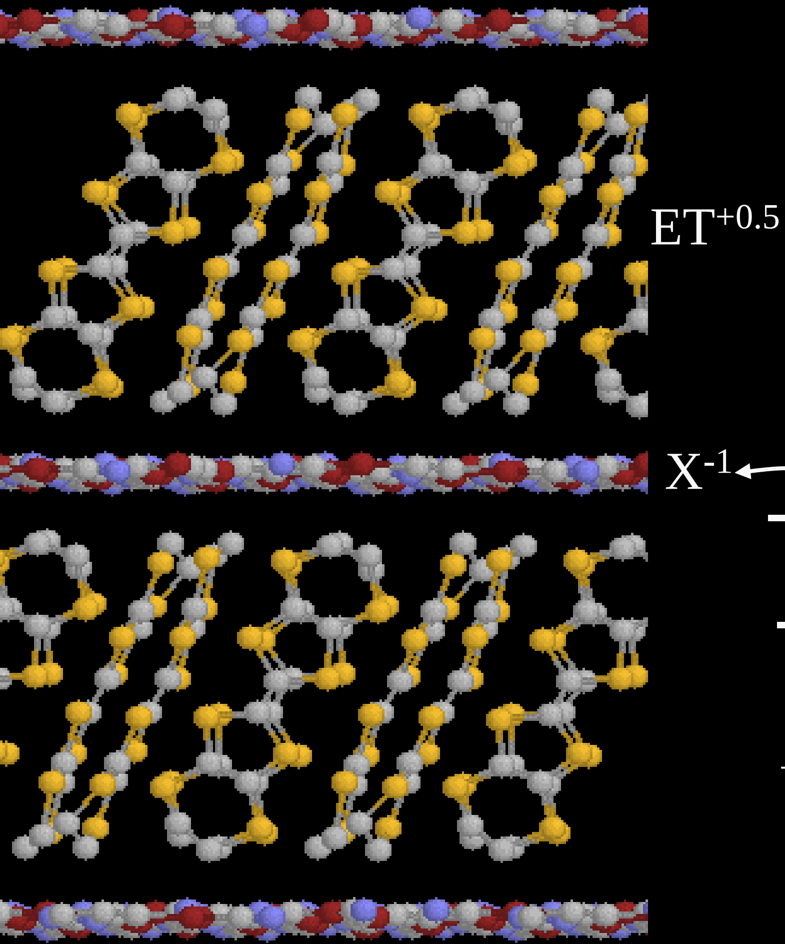
Triangular lattice

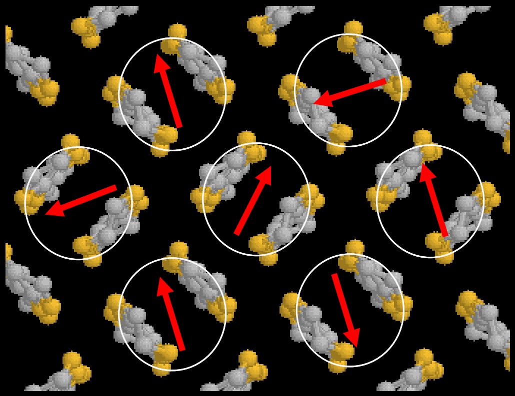
Cluster-DMFT

Tremblay et al. PRL (2006)

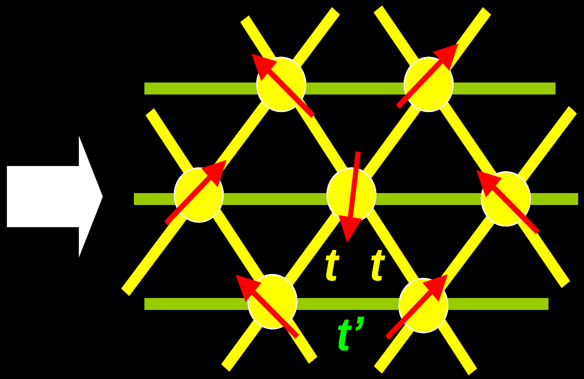


Organics κ-(ET)$_2$X; designable and cotrollable





Kino-Fukuyama (1996)

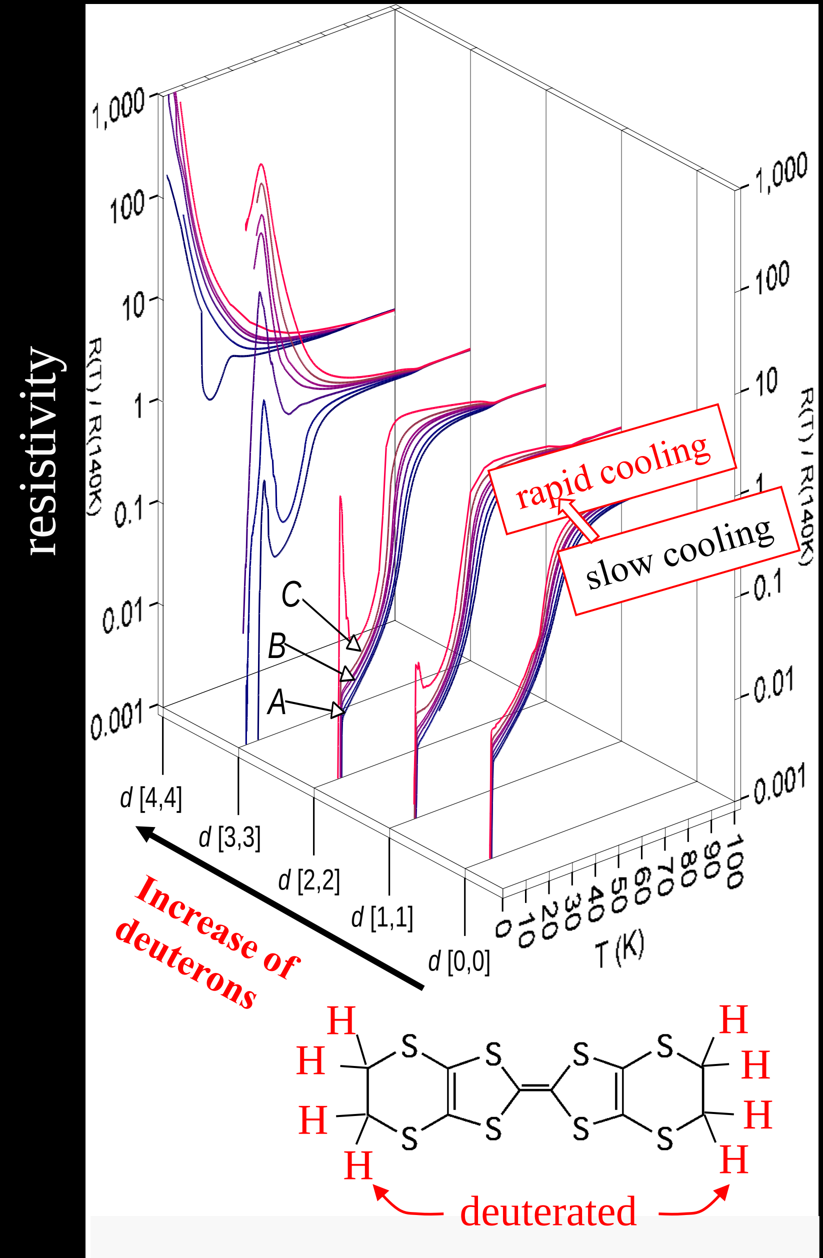


X$^-$	Ground state at ambient pressure	U/t	t'/t	
Cu$_2$(CN)$_3$	Mott ins.	8.2	1.06	0.80
Cu[N(CN)$_2$]Cl	Mott ins.	7.5	0.75	0.44
Cu[N(CN)$_2$]Br	Metal (SC)	7.2	0.68	
Cu(NCS)$_2$	Metal (SC)	6.8	0.84	
Cu(CN)[N(CN)$_2$]	Metal (SC)	6.8	0.68	
Ag(CN)$_2$ H$_2$O	Metal (SC)	6.6	0.60	
I$_3$	Metal (SC)	6.5	0.58	

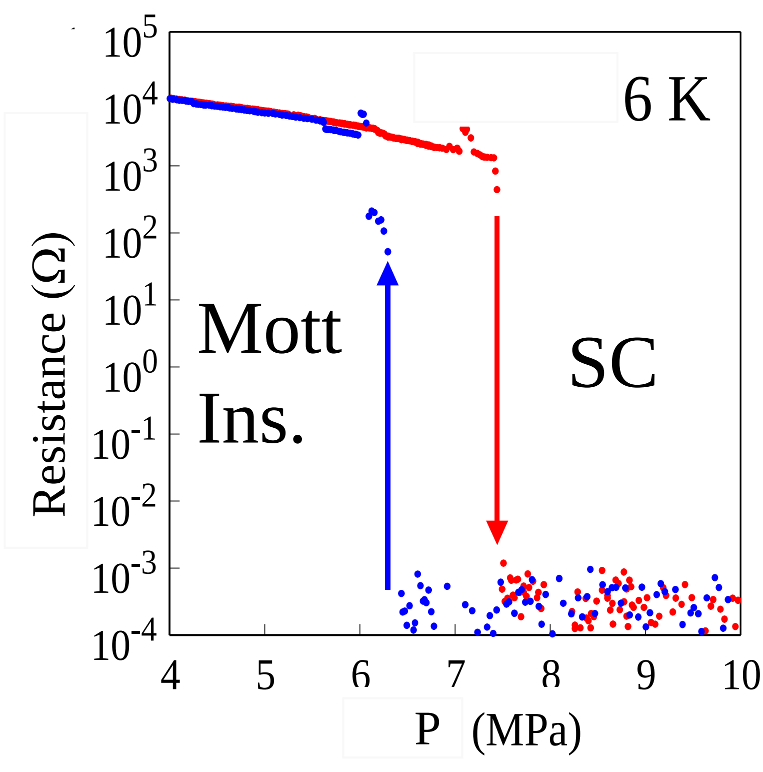
↑

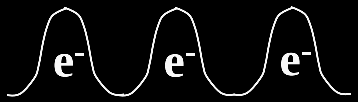
Ab initio
Kandpal *et al.*(2009)
Nakamura *et al.*(2009)

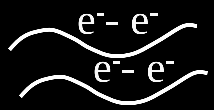
κ-$(ET)_2Cu[N(CN)_2]Br$ on the verge of superconductor-insulator transition



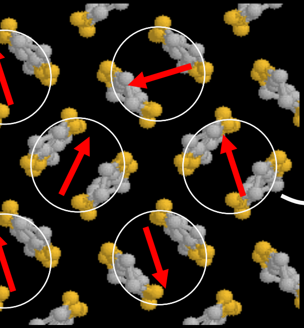
κ-(deuterated ET)$_2$Cu[N(CN)$_2$]Br

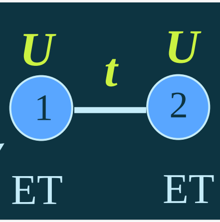




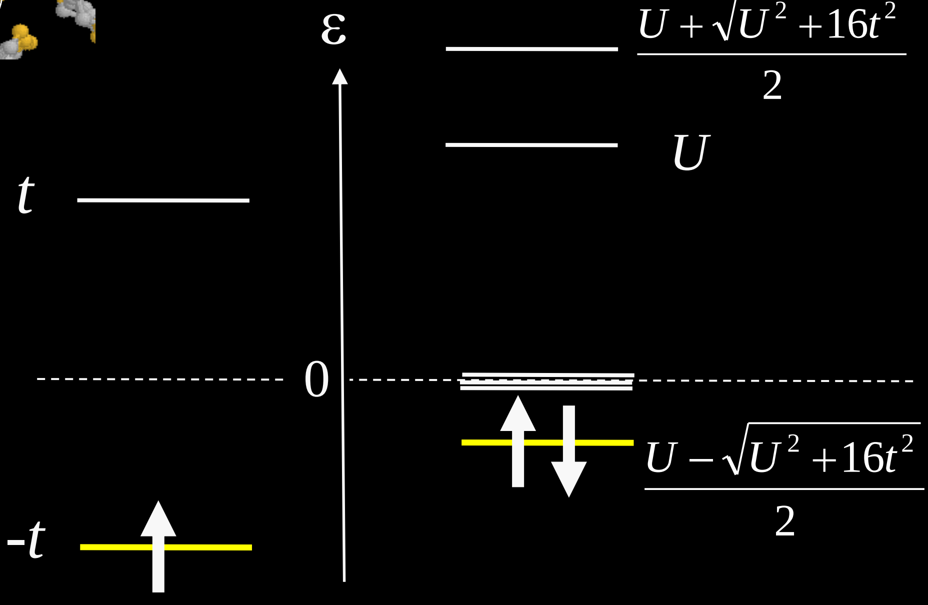


dimer with on-site Coulomb energy → Hubbard model of hydrogen molecule





$$H = -t\sum_{\sigma}(c^{+}_{1\sigma}c_{2\sigma} + c^{+}_{2\sigma}c_{1\sigma}) + U(n_{1\uparrow}n_{1\downarrow} + n_{2\uparrow}n_{2\downarrow})$$



$$\frac{U+\sqrt{U^2+16t^2}}{2} \qquad |\uparrow\downarrow,0\rangle + |0,\uparrow\downarrow\rangle + \frac{U-\sqrt{U^2+16t^2}}{4t}(|\uparrow,\downarrow\rangle - |\downarrow,\uparrow\rangle)$$

$$U \qquad |\uparrow\downarrow,0\rangle - |0,\uparrow\downarrow\rangle$$

$$0 \qquad |\uparrow,\uparrow\rangle, \quad |\downarrow,\downarrow\rangle, \quad |\uparrow,\downarrow\rangle + |\downarrow,\uparrow\rangle$$

$$\frac{U-\sqrt{U^2+16t^2}}{2} \qquad |\uparrow,\downarrow\rangle - |\downarrow,\uparrow\rangle + \frac{4t}{U+\sqrt{U^2+16t^2}}(|\uparrow\downarrow,0\rangle + |0,\uparrow\downarrow\rangle)$$

1-particle state

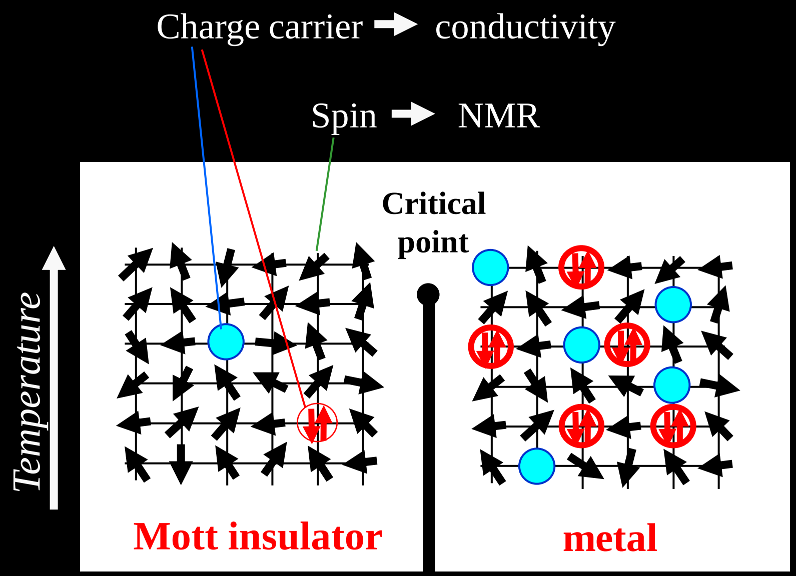
2-particle state

$$U_{eff} = \frac{U-\sqrt{U^2+16t^2}}{2} - 2(-t) = 2t + \frac{U-\sqrt{U^2+16t^2}}{2} \approx 2t$$

From band-structure calculations, $U_{eff} \sim 2t \sim 0.5$ eV and bandwidth $W \sim 0.4$ eV

comparable

Mott transition --- *Gas-liquid transition of doublon/holon fluid*



Charge carrier → conductivity

Spin → NMR

Temperature

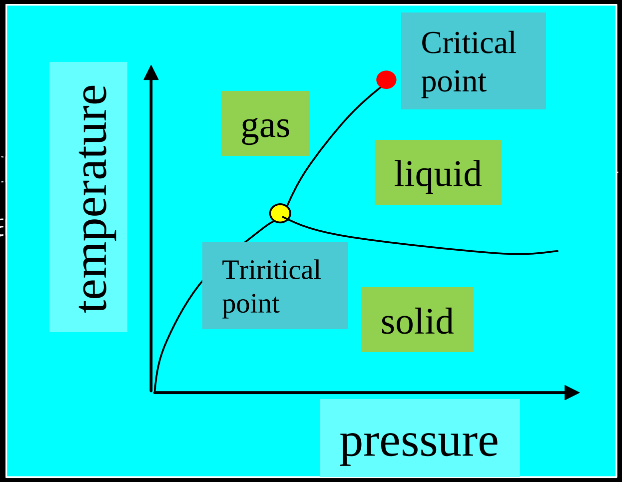
Critical point

Mott insulator

metal

Castel

Kotlia



temperature

Critical point

gas

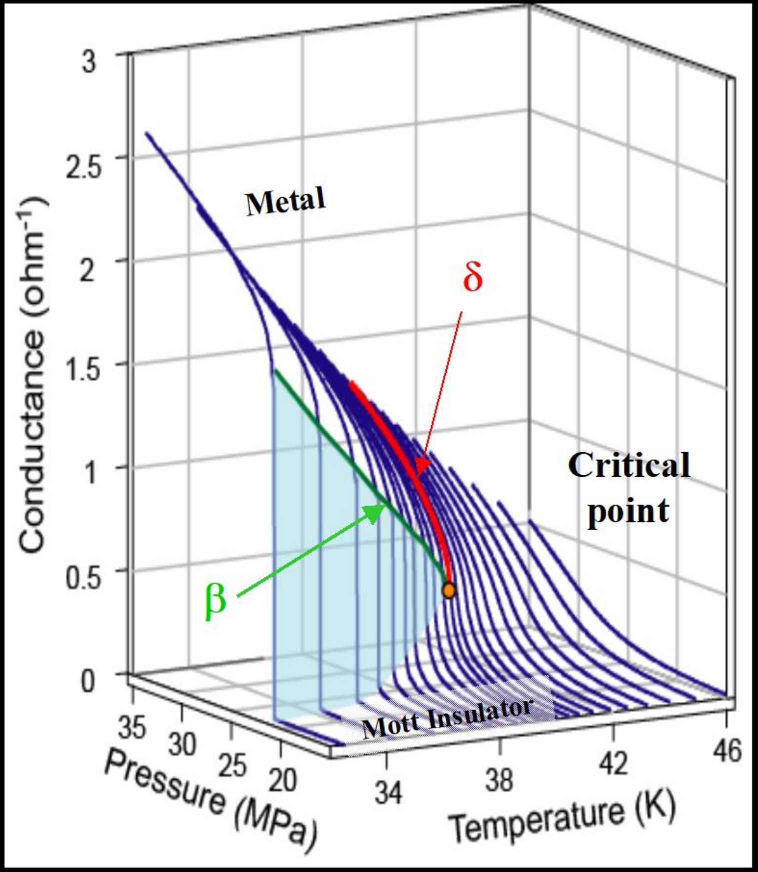
liquid

Tiritical point

solid

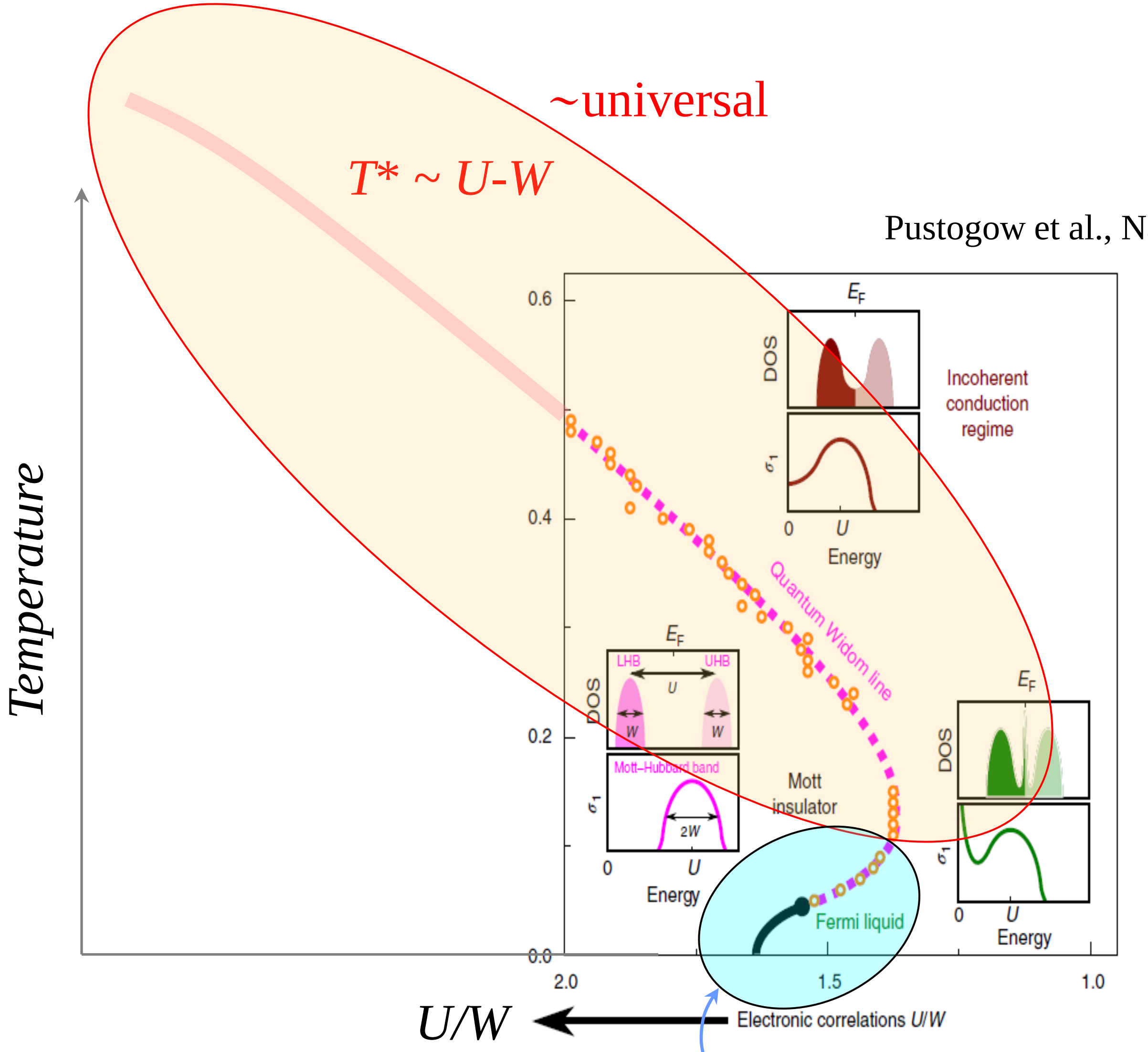
pressure

κ-(ET)$_2$Cu[N(CN)$_2$]Cl



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

Mott physics
high-energy universality and low-energy diversity



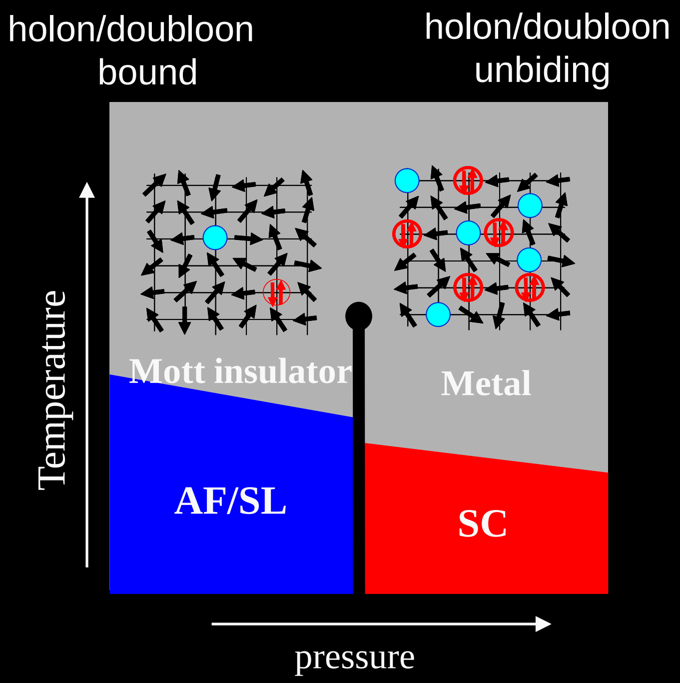
Pustogow et al., Nat. Mat. 17, 773 (2018)

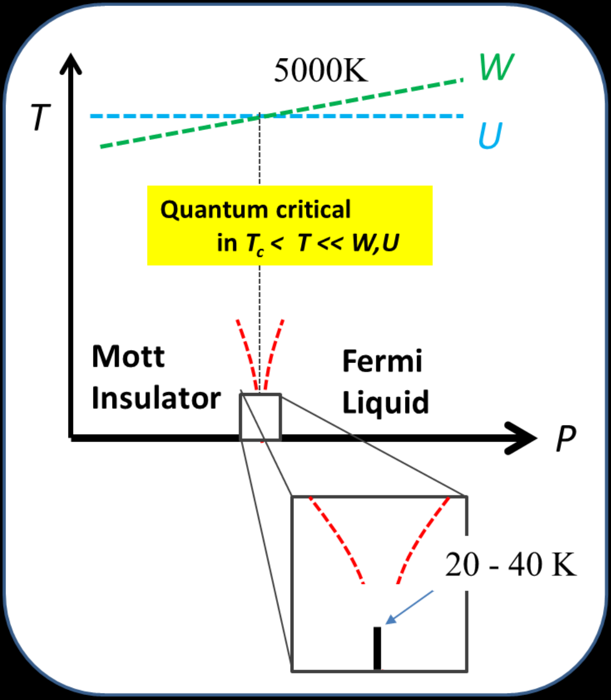
depends on the system's detail

Mott metal-insulator transition

- competition between U and W -

Mott phase diagram





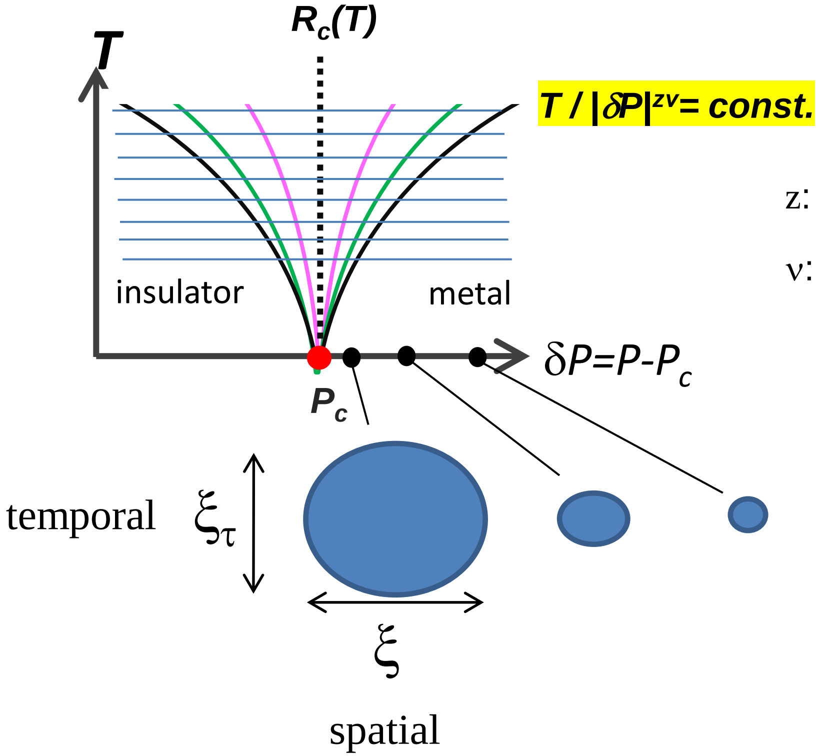
Quantum critical scaling

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

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

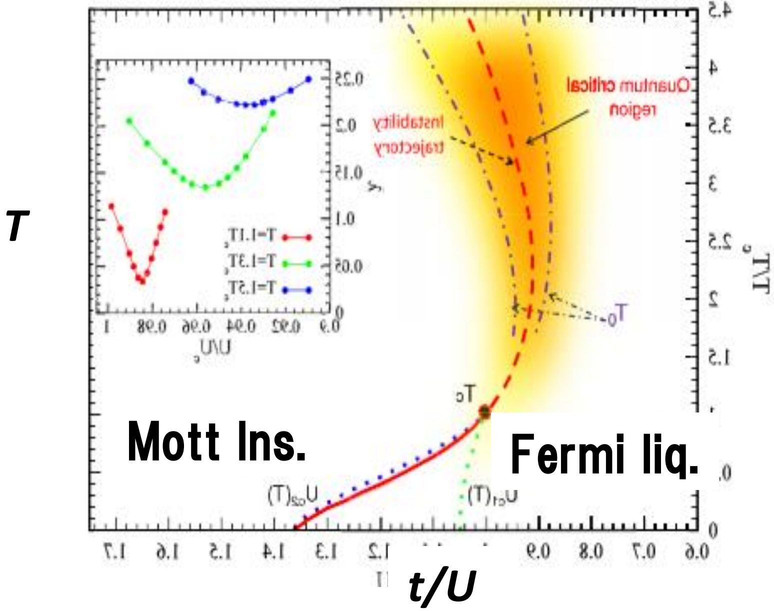
Quantum correlation time

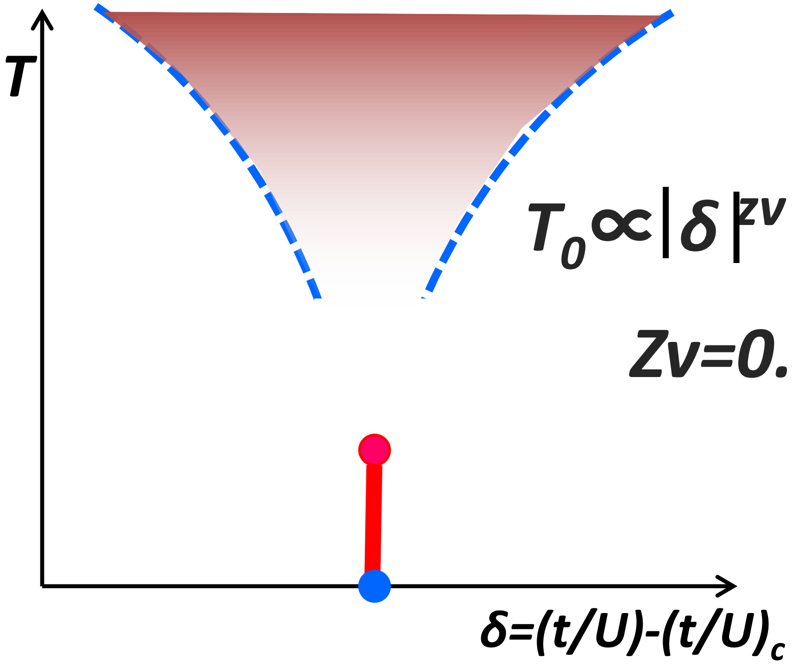
$\xi_\tau = \xi^z = |\delta P|^{-z\nu}$

DMFT of Hubbard model at high temperatures

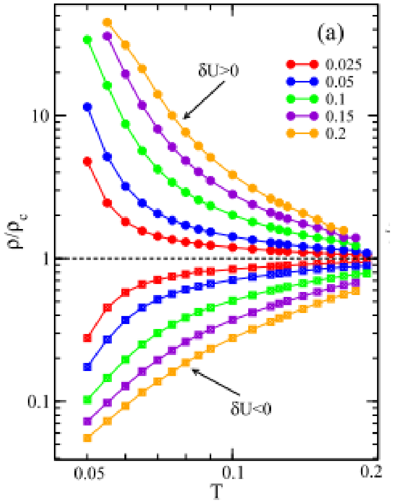
Quantum Critical Transport Near the Mott Transition **H. Terletska** *et al.*, PRL **107** (2011)

***T* - *t/U* phase diagram**

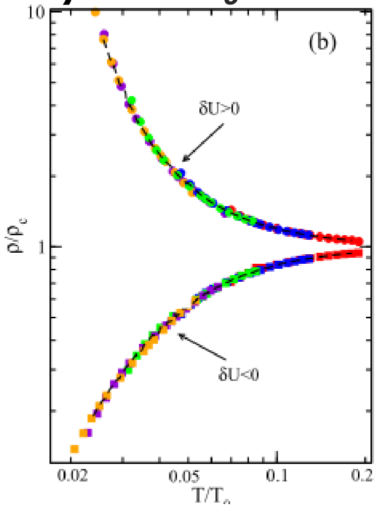




ρ* vs *T* *calc.



ρ* vs *T/T₀* *calc.

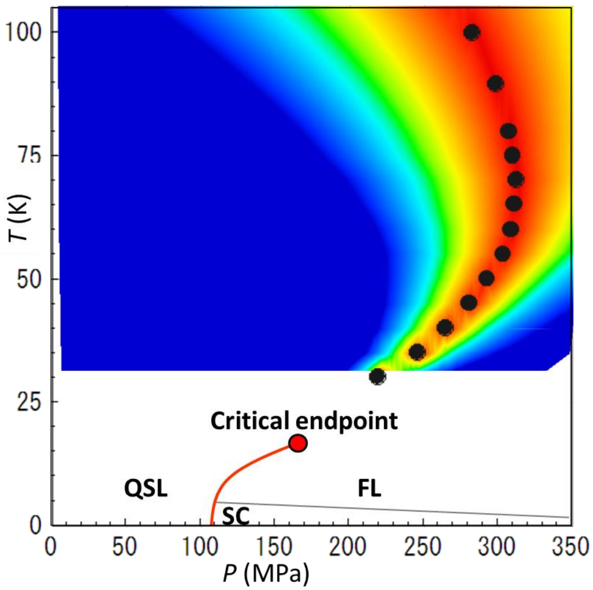


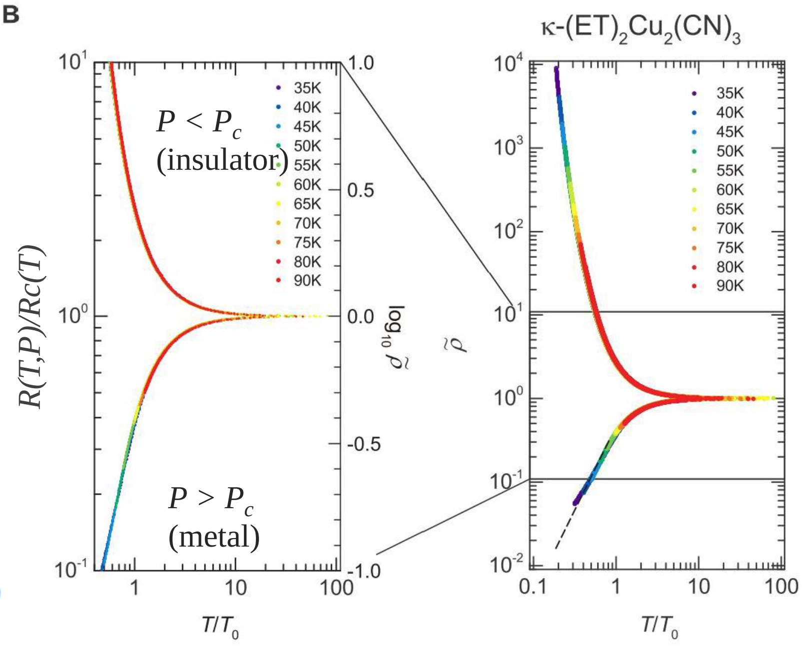
Resistivities ρ*(T,δ)* are scaled with the one parameter, *T/T₀*

Characteristic energy, $T_0 \propto \delta^{z\nu}$

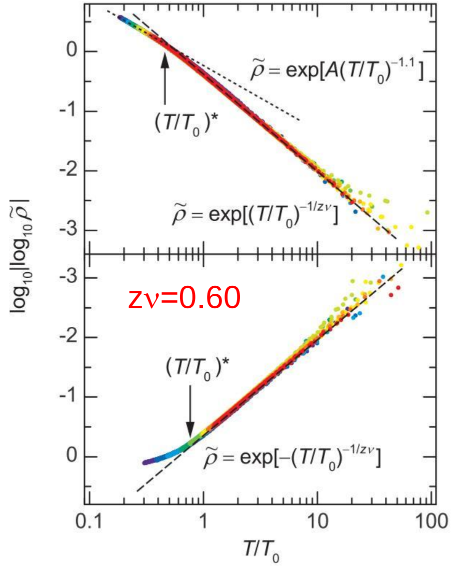
→ *quantum criticality*

Phase diagram & quantum critical scaling





Insulator branch



Metal branch

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

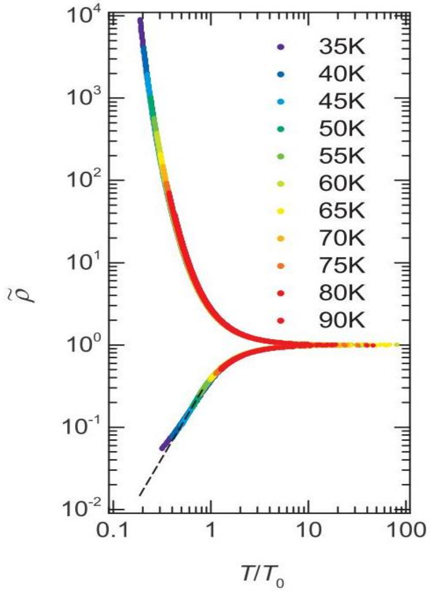
Furukawa et al., Nat. Commun. **9**, 307 (2018)

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

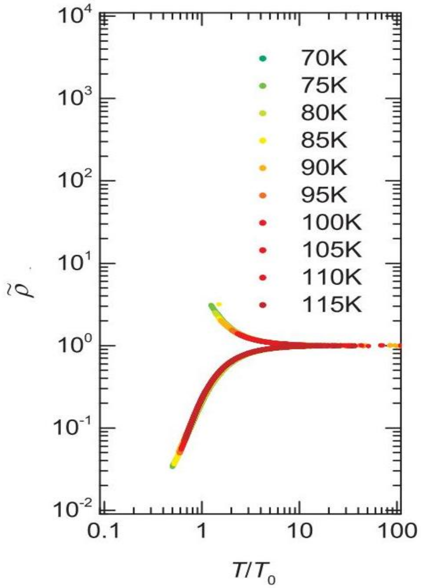
Near-universal quantum criticality (zν=0.5-0.7) at high energies

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

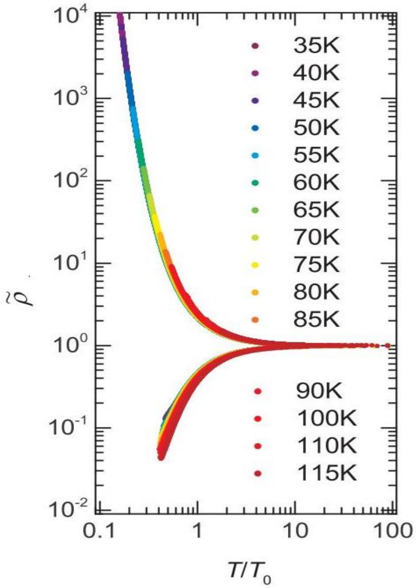
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$



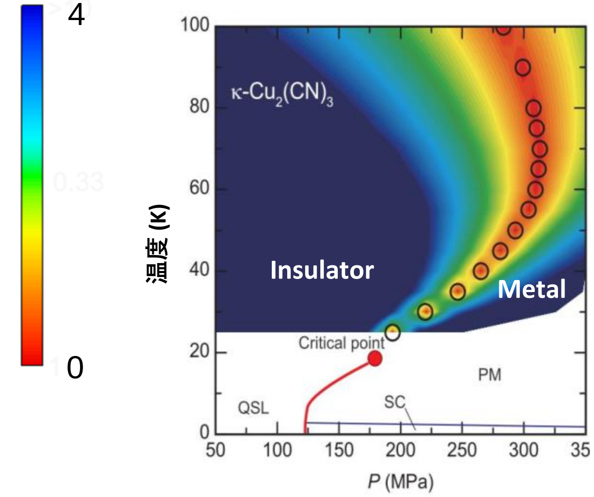
κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl



EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

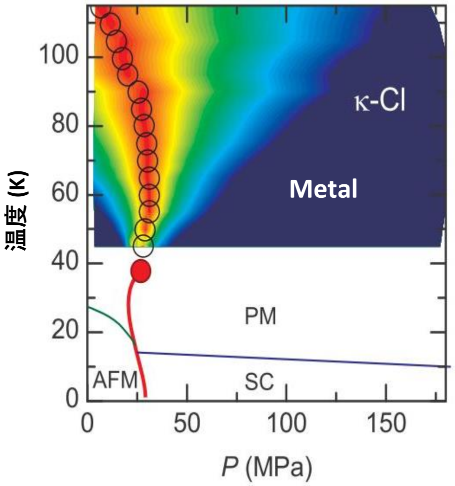


zν=0.62±0.02



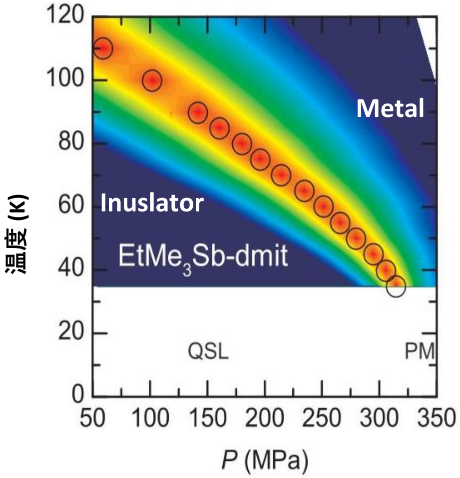
QSL/SC

zν=0.49±0.01



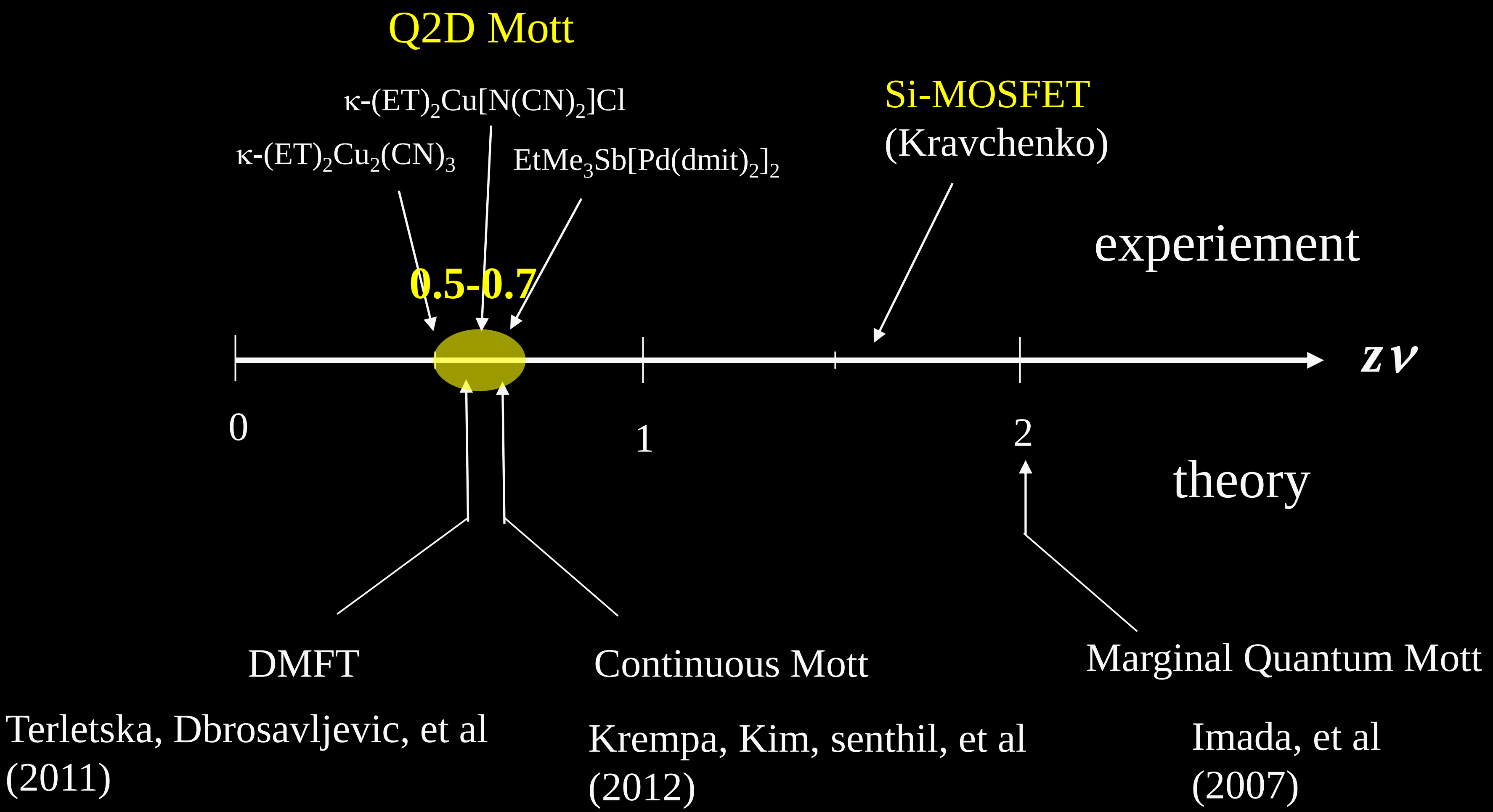
AFM/SC

zν=0.68±0.04



QSL/Metal

Critical exponents, $z\nu$, in metal-insulator transitions



Q2D Mott

κ-(ET)$_2$Cu[N(CN)$_2$]Cl

κ-(ET)$_2$Cu$_2$(CN)$_3$

EtMe$_3$Sb[Pd(dmit)$_2$]$_2$

Si-MOSFET
(Kravchenko)

experiement

theory

DMFT

Terletska, Dbrosavljevic, et al
(2011)

Continuous Mott

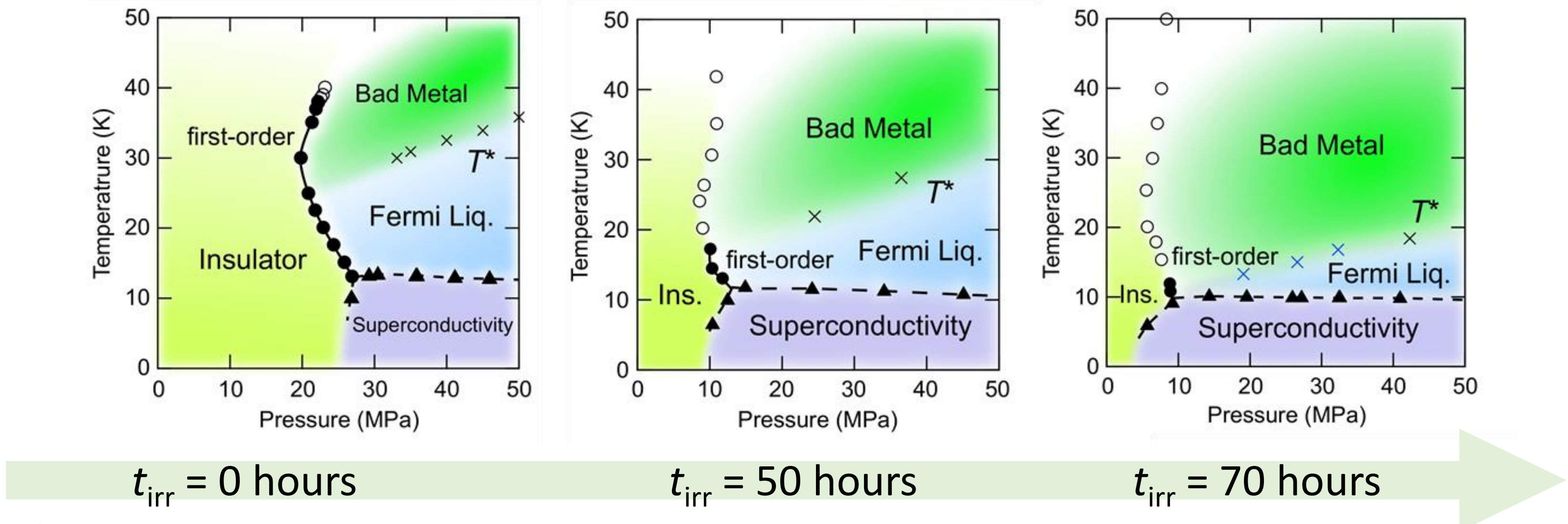
Krempa, Kim, senthil, et al
(2012)

Marginal Quantum Mott

Imada, et al
(2007)

1: Effect of disorder on Mott transition

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



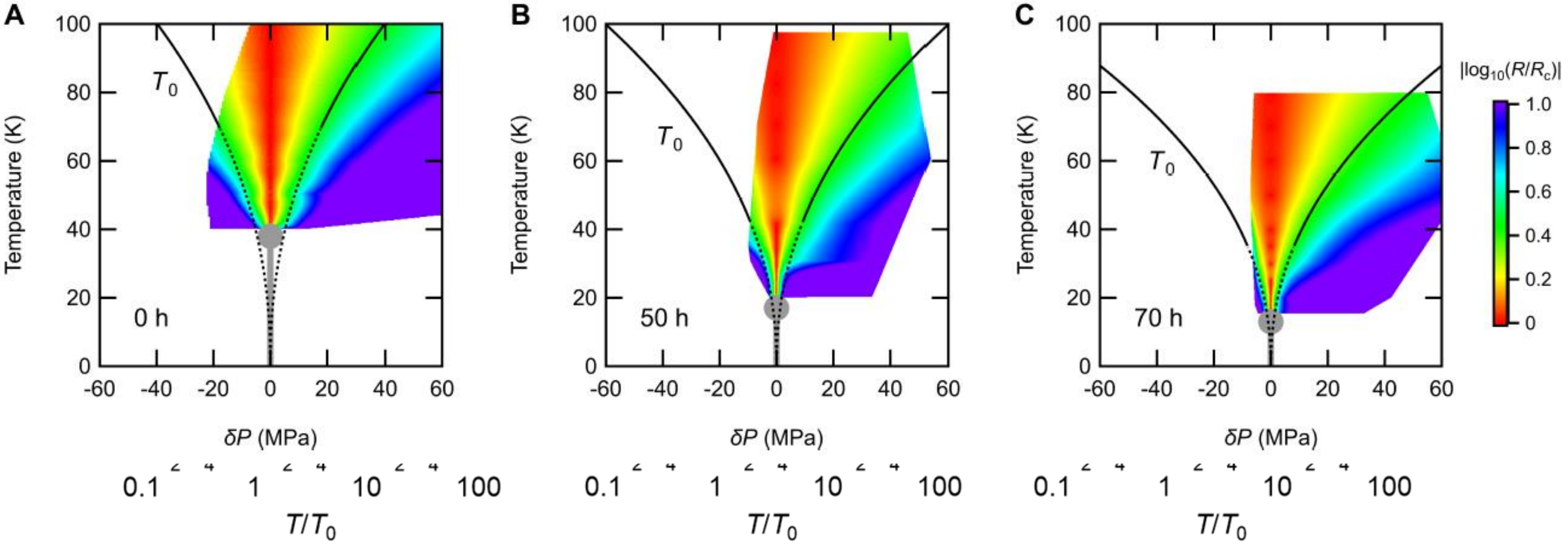
t_{irr} = 0 hours

t_{irr} = 50 hours

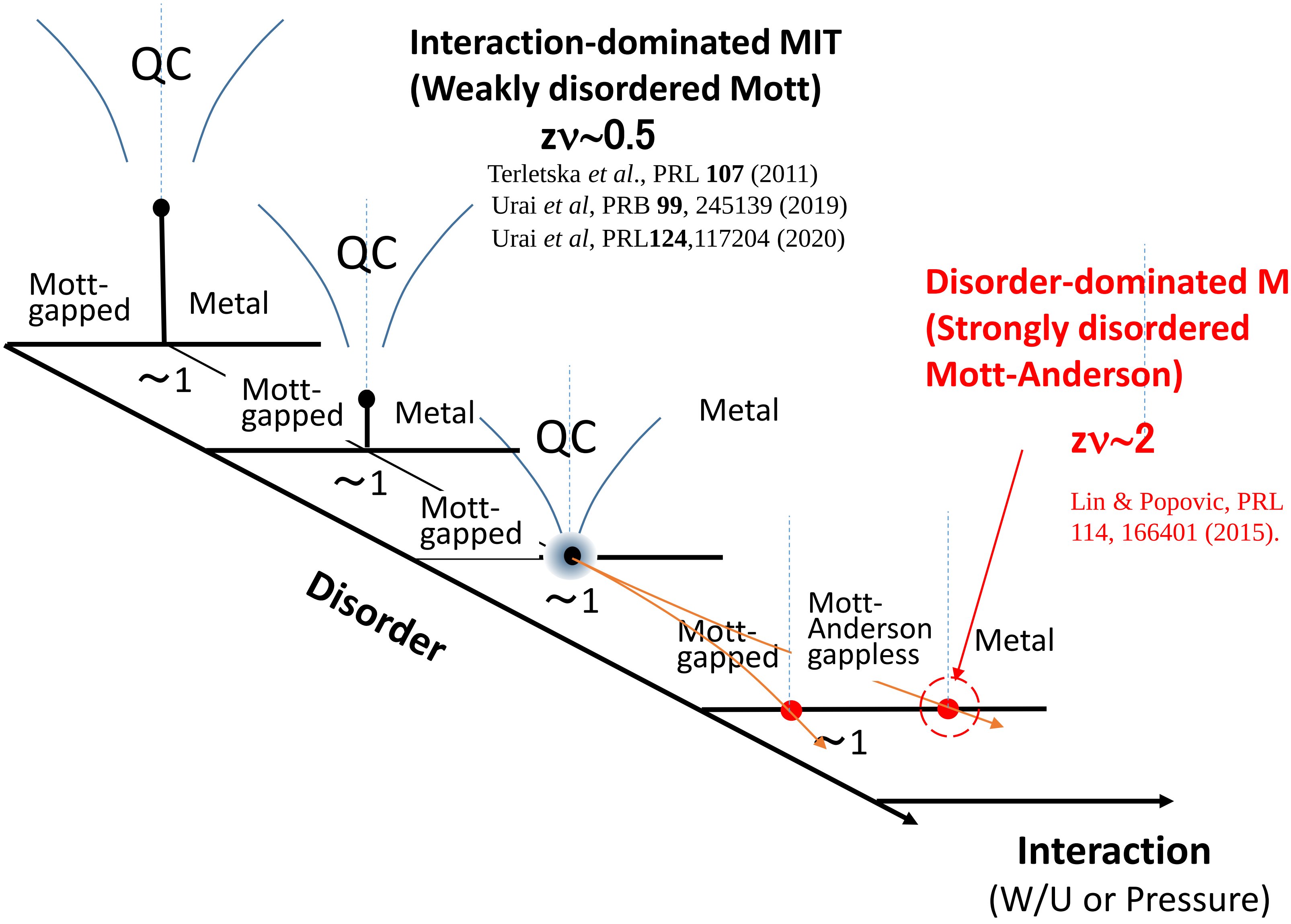
t_{irr} = 70 hours

✓ ***Enhancement of the quantum critical fluctuations***

Urai *et al*, PRB **99**, 245139 (2019)
Urai *et al*, PRL**124**,117204 (2020)



Perspective: from interaction-riven to disorder-driven quantum criticality



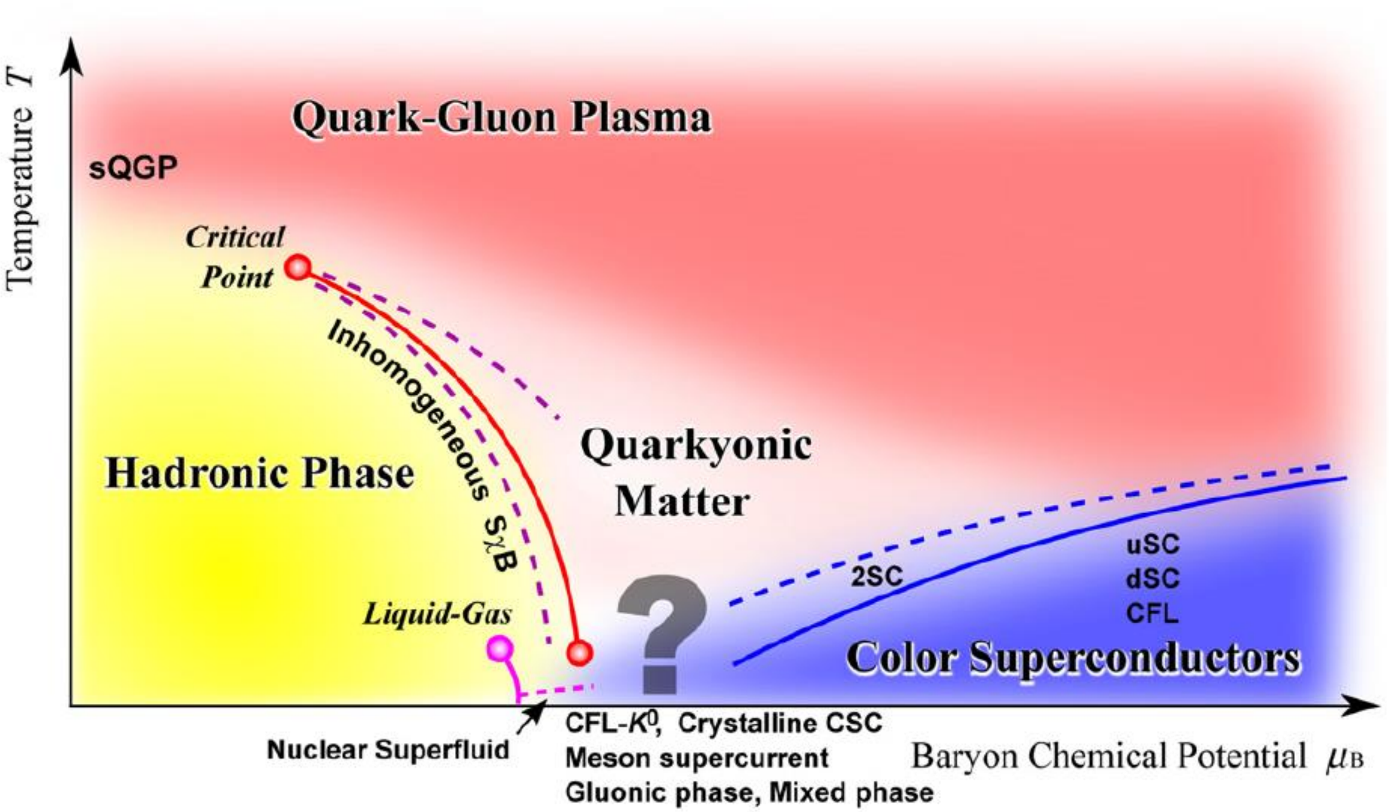
Interaction-dominated MIT
(Weakly disordered Mott)
$z\nu$~0.5

Terletska *et al.*, PRL **107** (2011)
Urai *et al*, PRB **99**, 245139 (2019)
Urai *et al*, PRL**124**,117204 (2020)

Disorder-dominated MIT
(Strongly disordered Mott-Anderson)
$z\nu$~2

Lin & Popovic, PRL 114, 166401 (2015).

Phase diagram of QCD

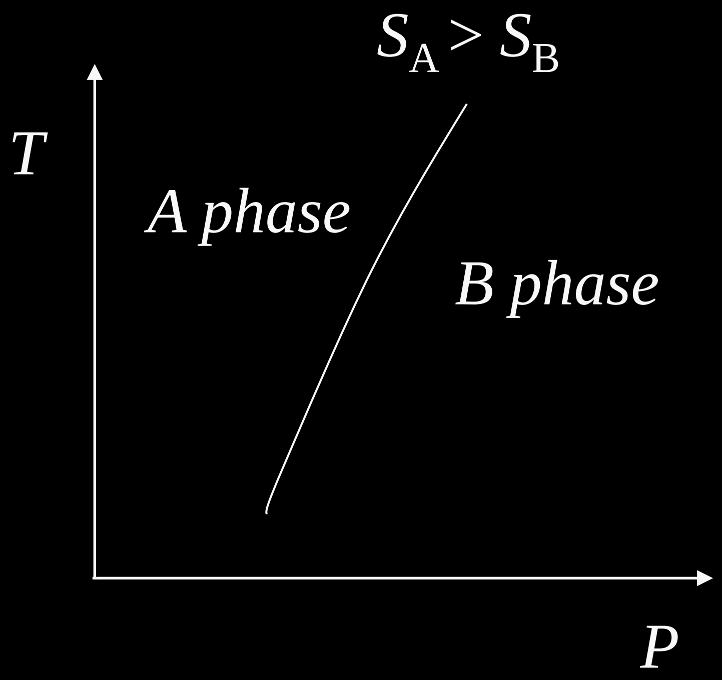


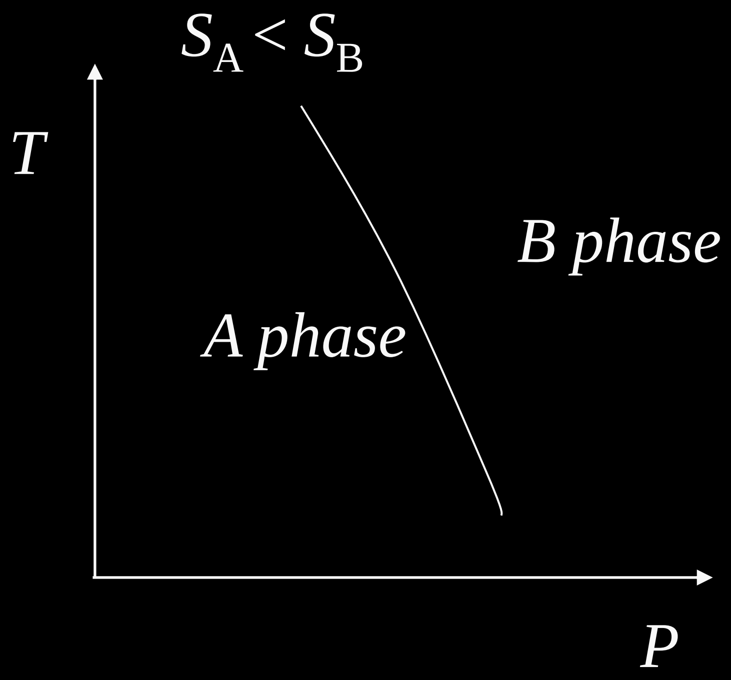
Fukushima and Hatsuda, Rep. Prog. Phys. 74 (2011) 014001

Thermodynamics of Mott transition

Slope of the phase boundary tells the nature of electronic phases

Clausius Clapeyron $$dT/dP = \underbrace{(V_A-V_B)}_{>0}/(S_A-S_B)$$



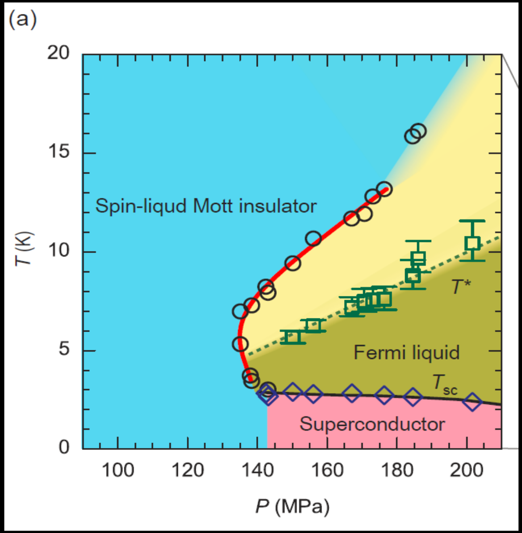


Thermodynamics of Mott transition

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

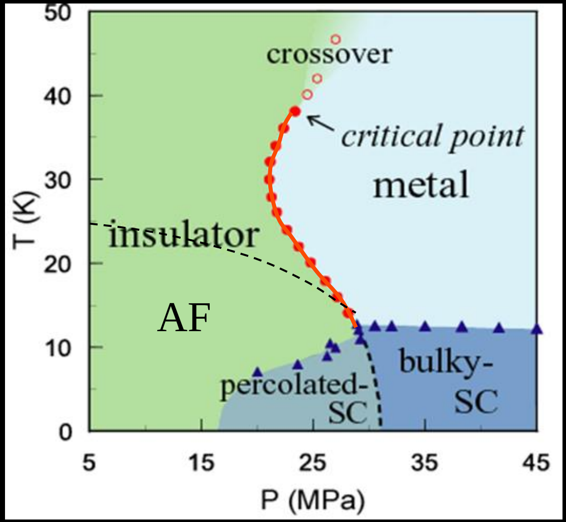
κ-(ET)$_2$Cu$_2$(CN)$_3$ t'/t =1.06

QSL



κ-(ET)$_2$Cu[N(CN)$_2$]Cl t'/t =0.75

AF

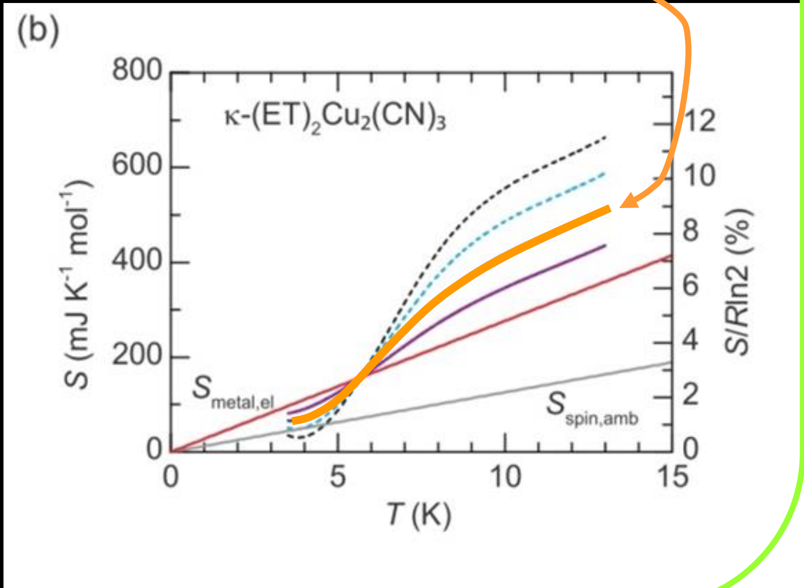


Clausius Clapeyron

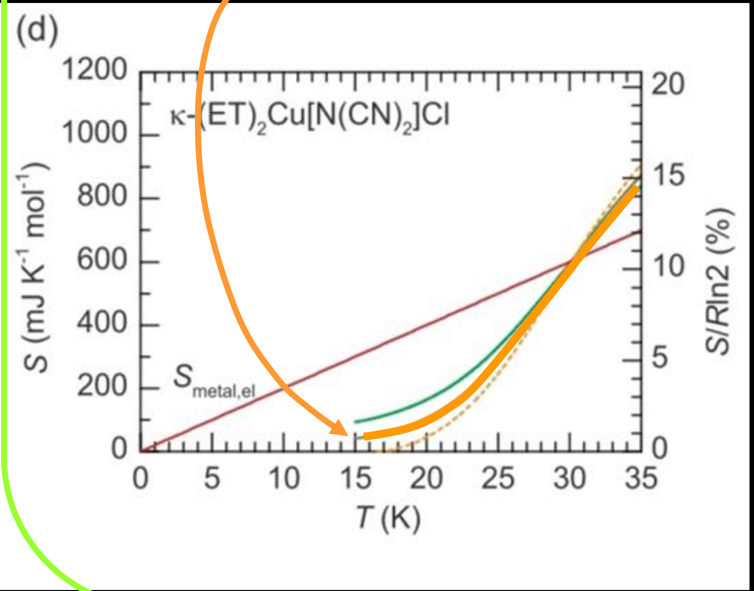
$$S_{Mott}-S_{metal}=(dP/dT)\Delta V$$

$$S_{Mott}=(dP/dT)\Delta V+\gamma T$$

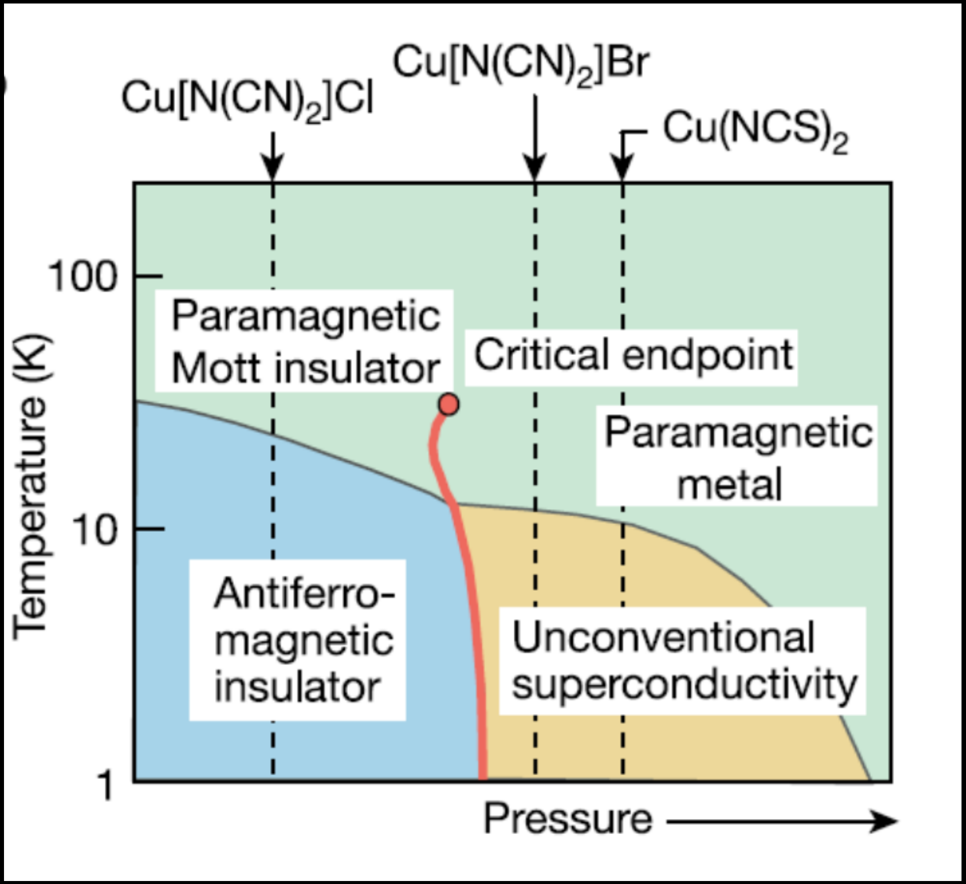
$\Delta V \leq 2\times10^{-8}$ cm/mol



$\Delta V \sim 5\times10^{-7}$ cm/mol (cf. $\Delta V_{exp} \leq 4.8\times10^{-7}$)



Superconductivity



T_c is enhanced

near Mott transiton → charge fluctuations

near AF order → Spin fluctuations

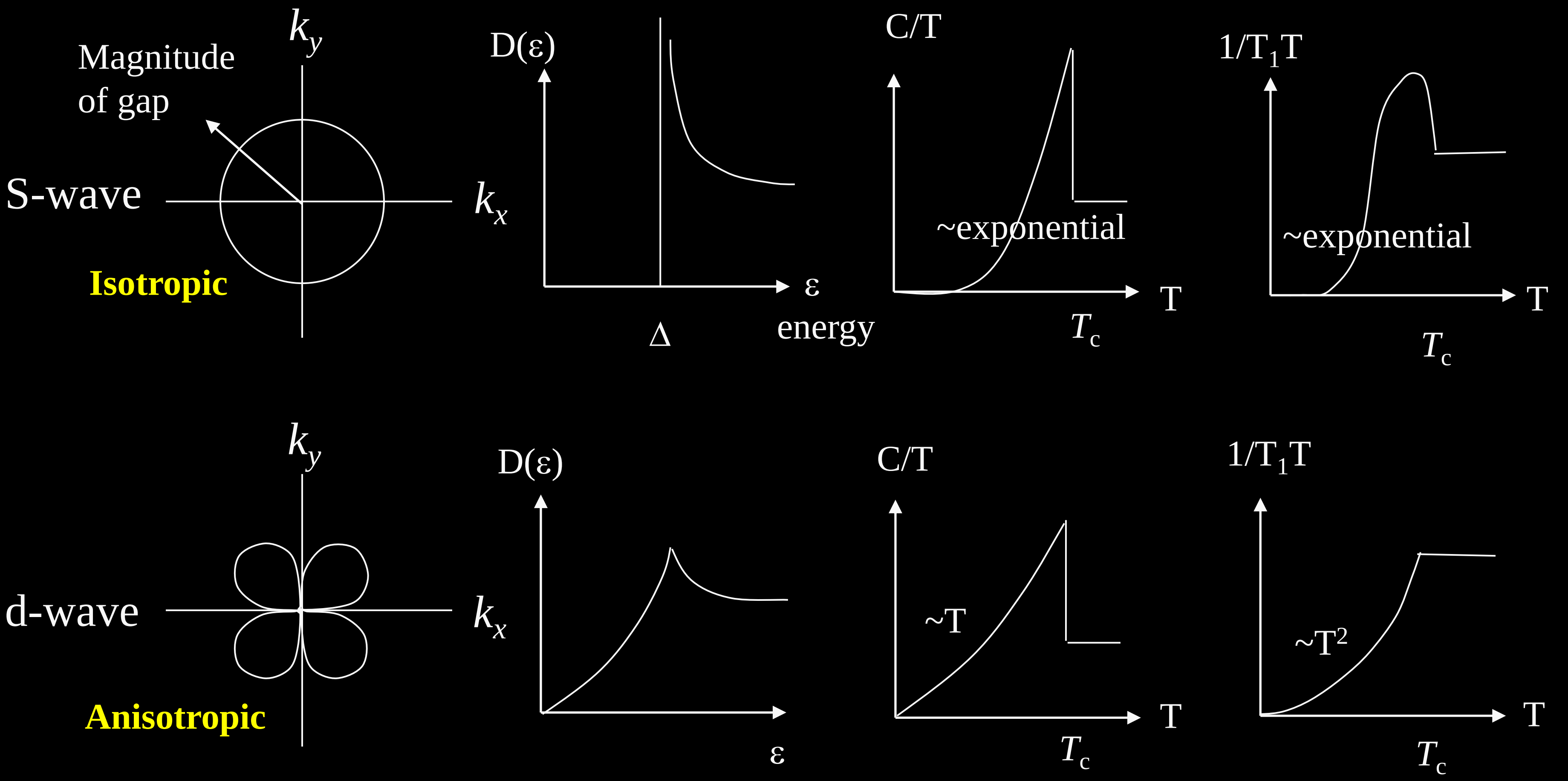
Probing pairing symmetry

Gap profile
In k-space

Quasi-particle
DOS

Specific heat
$C/T \sim \langle \rho(k_B T) \rangle$

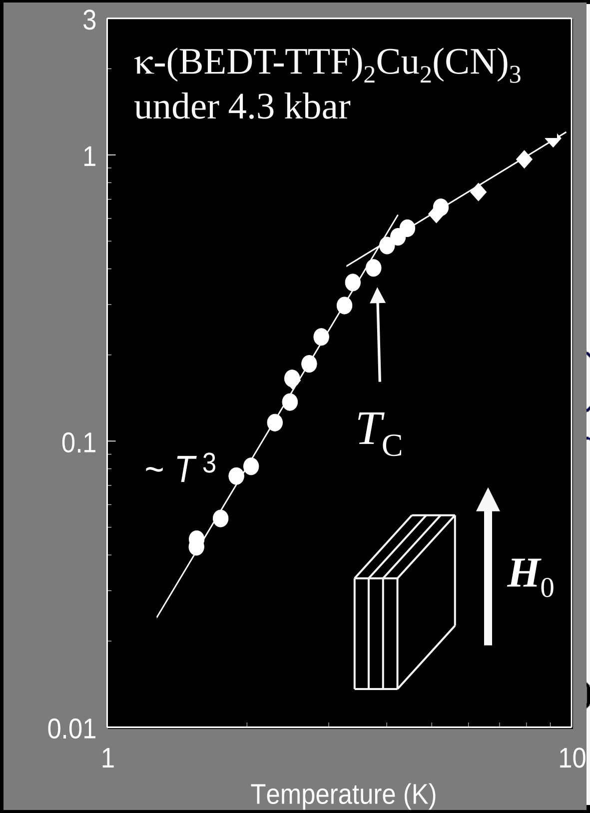
NMR relaxation rate
$1/T_1 T \sim \langle \rho(k_B T) \rangle^2$



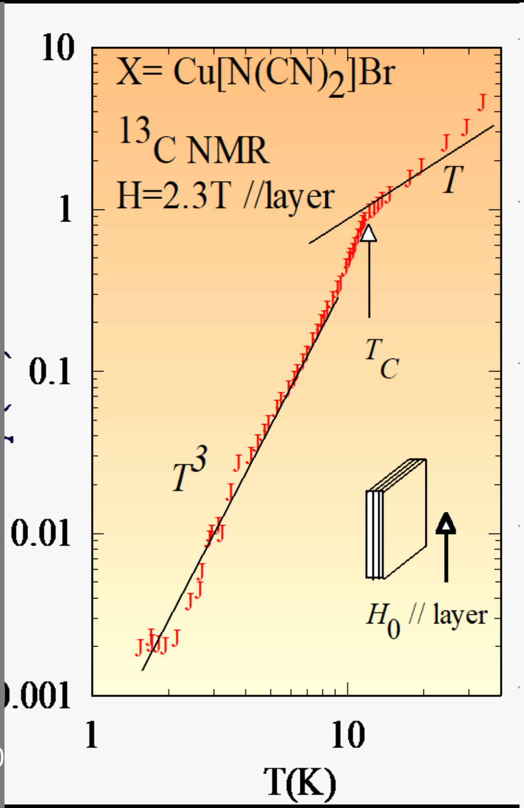
^{13}C NMR $1/T_1$ indicates nodal lines

$$1/T_1 \propto T^3$$

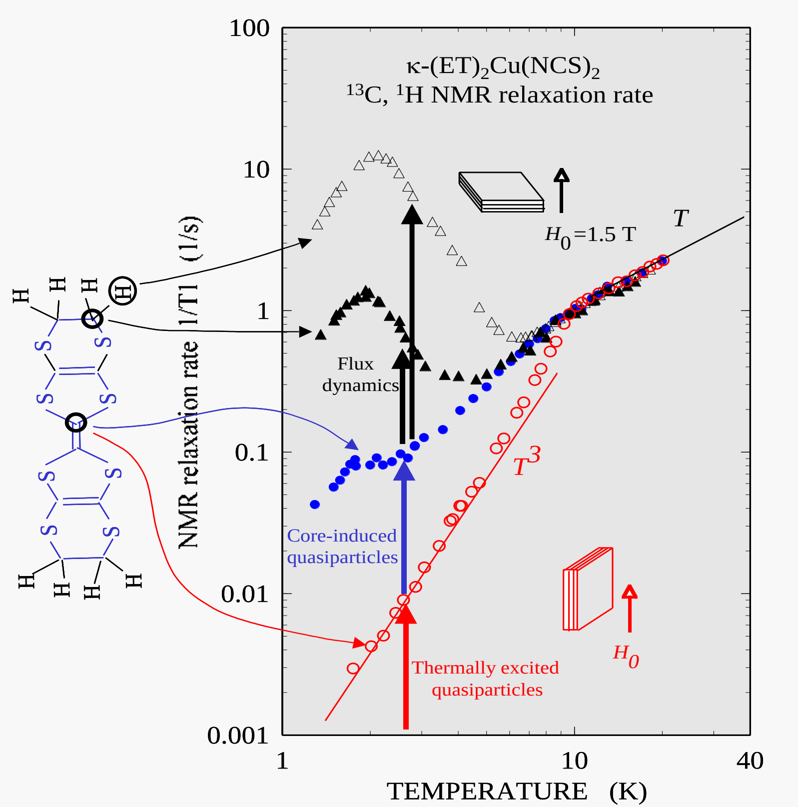
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$
Shimizu et al. (2006)



κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br
KK et al., (1996)



κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$
Miyagawa et al., (2004)

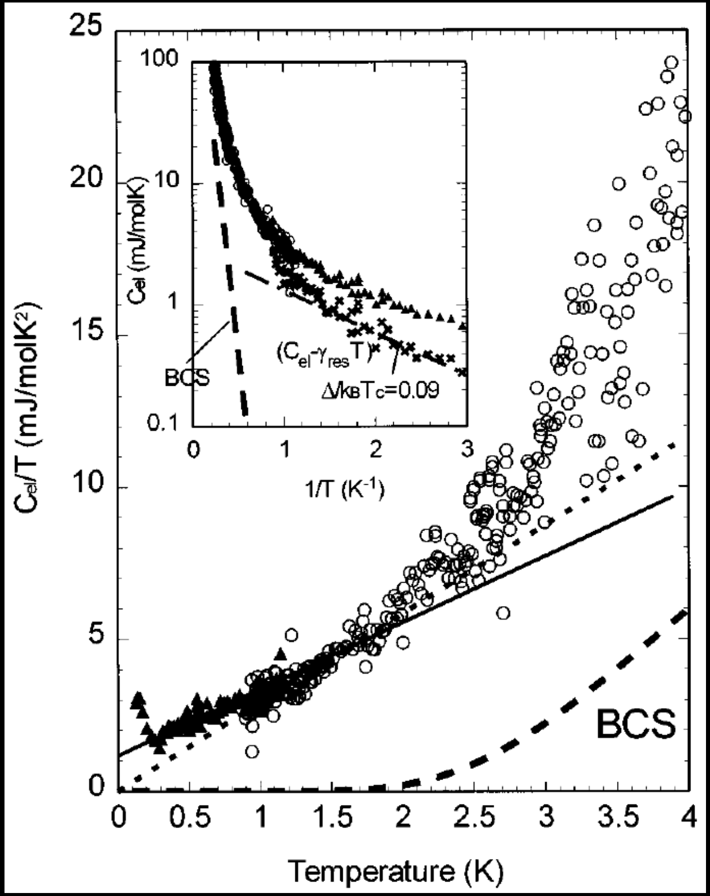


Specific heat indicates nodal lines

$C/T \propto T$

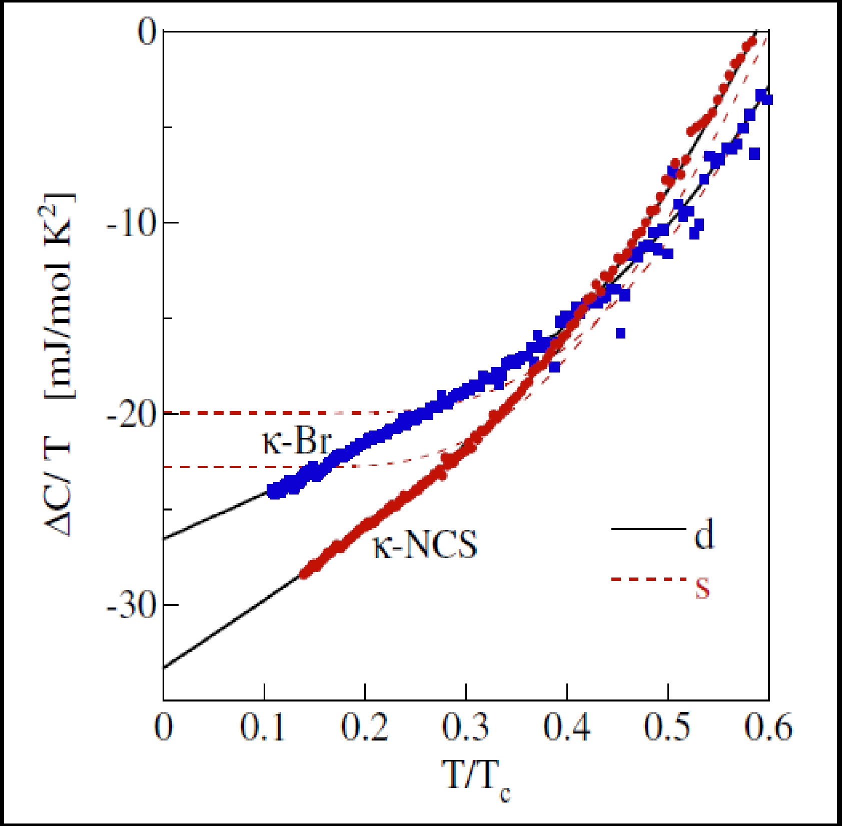
κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

Nakazawa et al., PRB(1997)



κ-(BEDT-TTF)$_2$Cu(NCS)$_2$
κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

Carrington et al., PRL(2007)



cf, fully gapped SC in

Elsinger et al., PRL. 84, 6098 (2000).

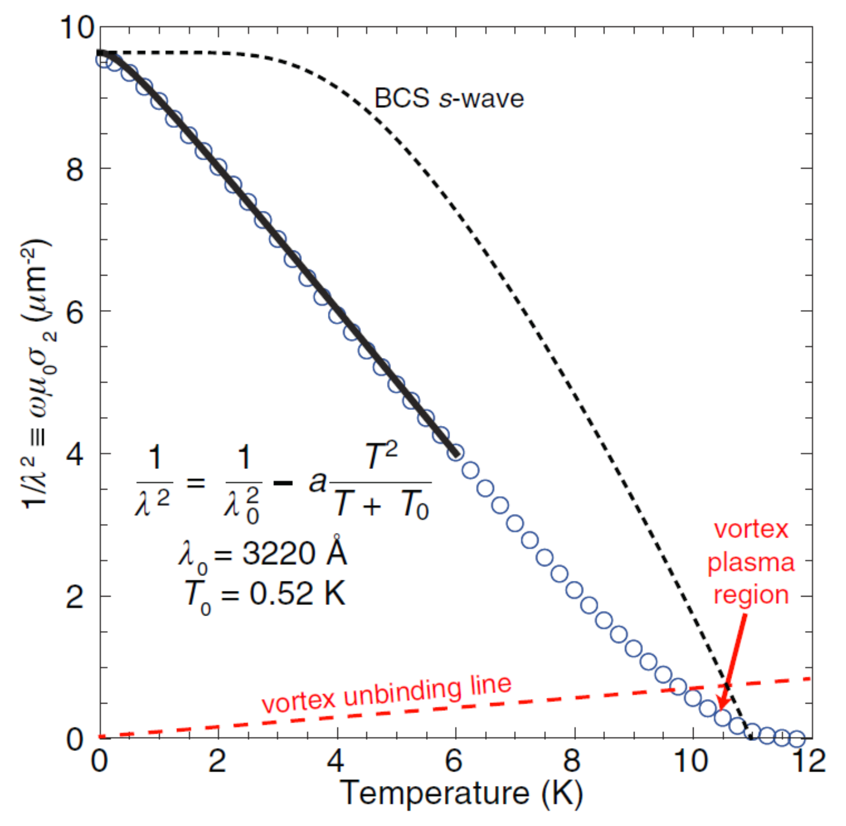
J. Muller *et al.*, PRB 65, 140509 (2002)

Superfluid density indicates nodal lines

$$\frac{n_s(T)}{n_s(0)} = 1 - \alpha T$$

κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

S. Milbradt et al., PRB.88.064501(2013)



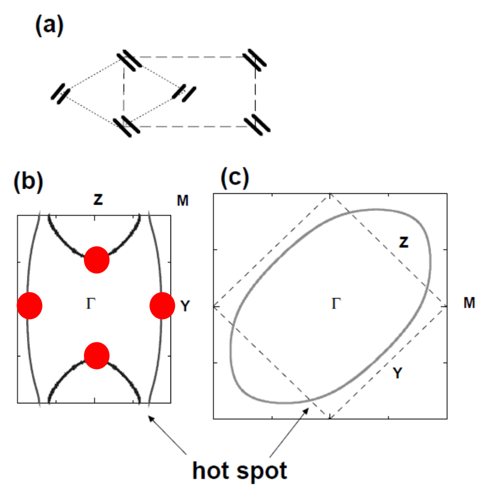
Superfluid density, n_s, deduced from penetration depth

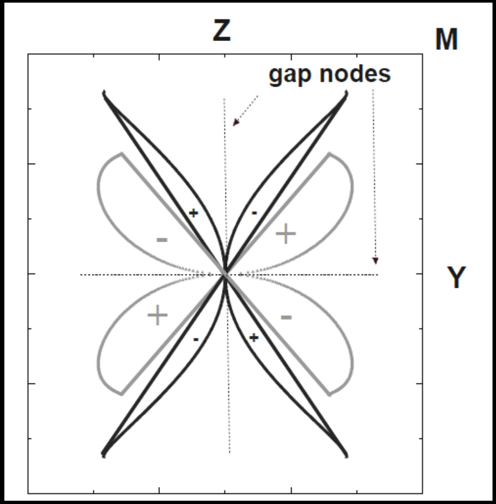
$$\frac{n_s}{m^*} = \frac{c^2}{4\pi e^2}\frac{1}{\lambda_L{}^2}$$

cf, fully gapped SC in Lang et al., PRL 1992, 69, 1443

Theoretical study of gap nodes

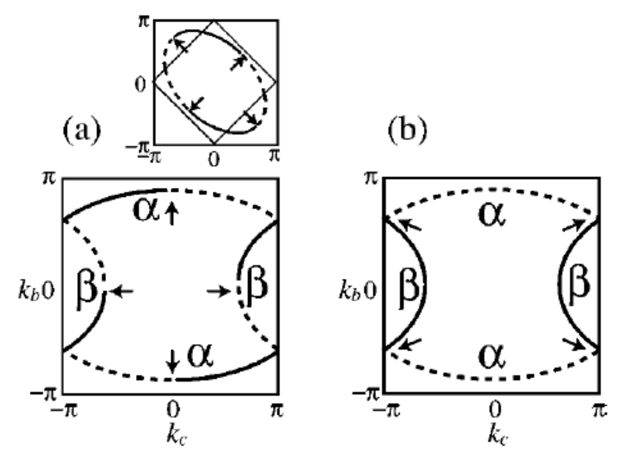
Schmalian PRL 81. 4232 (1998)



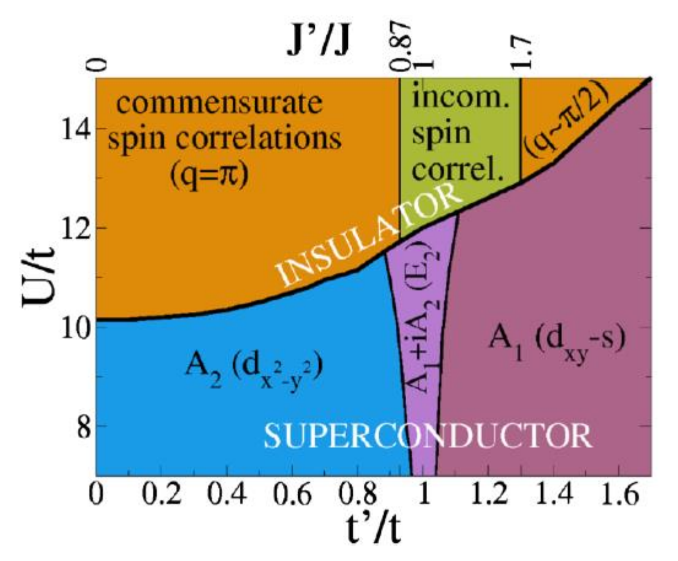


$d_{x^2-y^2}$ and d_{xy} are nearly degenarate

Kuroki *et al.*, PRB65, 100516(R) (2002)



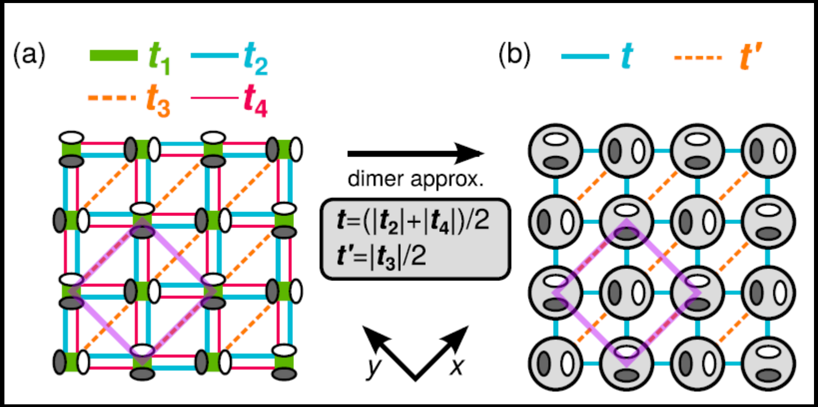
Powell & McKenzie, PRL 98, 027005 (2007)

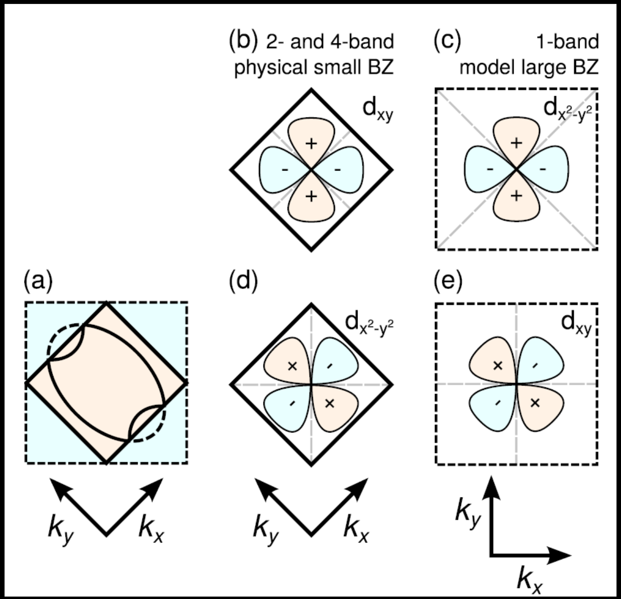


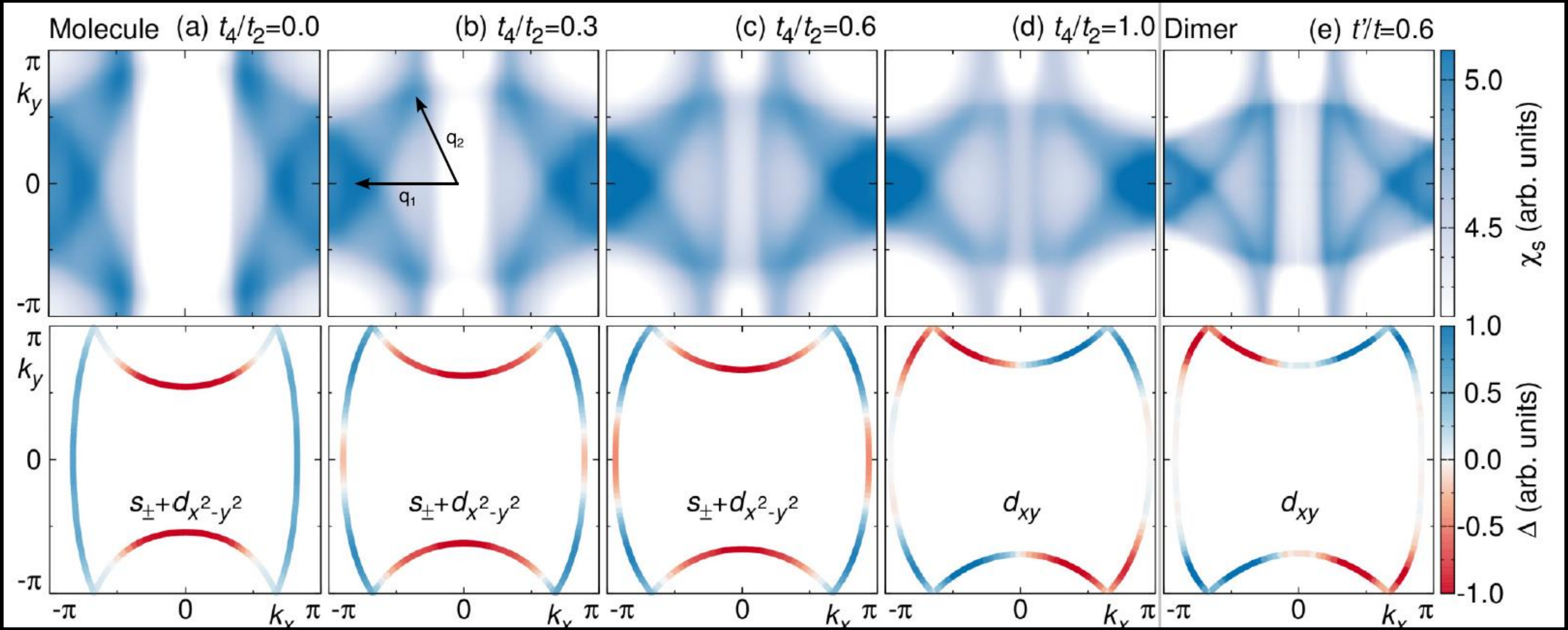
Theoretical study of gap nodes

$s + d_{x^2-y^2}$ and d_{xy} are nearly degenarate

Guterding et al., PRB 94, 024515 (2016)





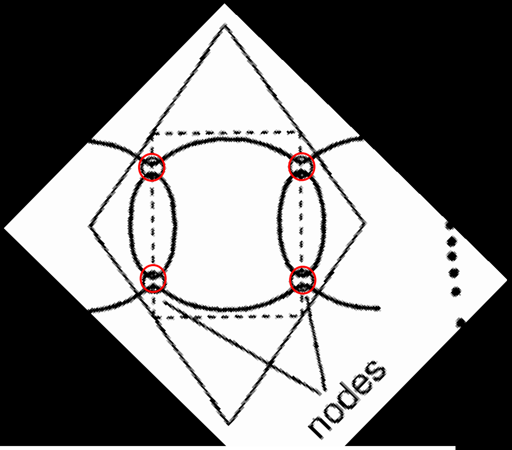


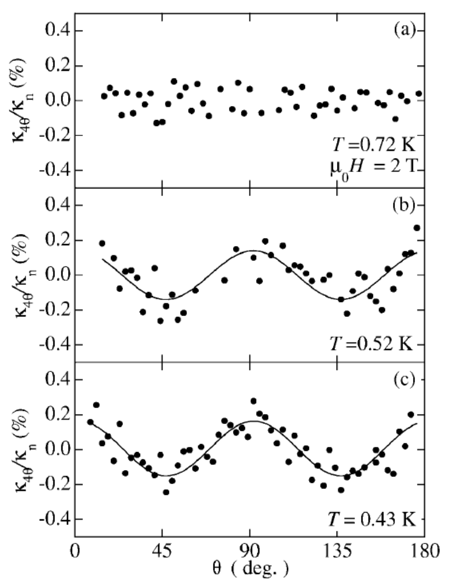
Field-angle dependent thermodynamic measurements

κ-(BEDT-TTF)$_2$X

Thermal conductivity

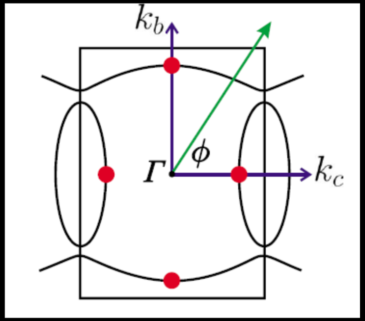
X=Cu(NCS)$_2$



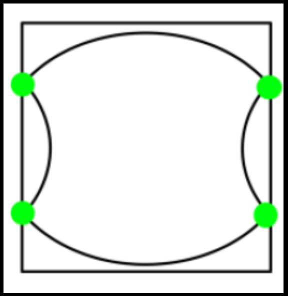


Specific heat

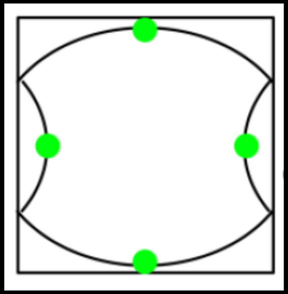
X=Cu(NCS)$_2$
X=Cu[N(CN)$_2$]Br

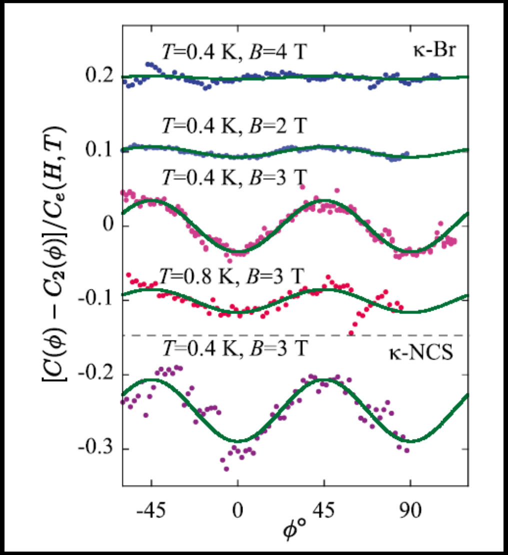


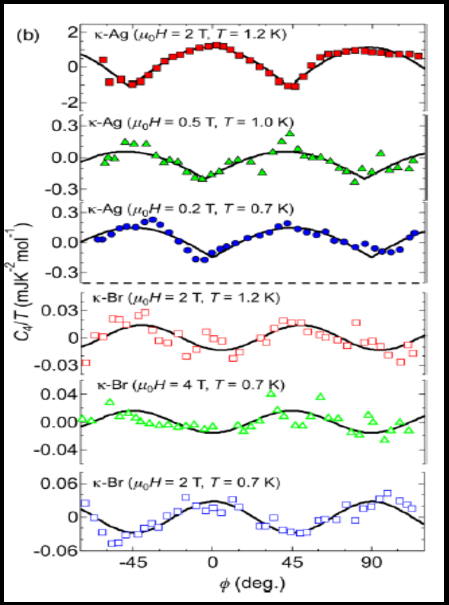
X=Cu[N(CN)$_2$]Br



X=Ag(CN)$_2$ H$_2$O







Izawa *et al*., PRL 88, 027002 (2002).

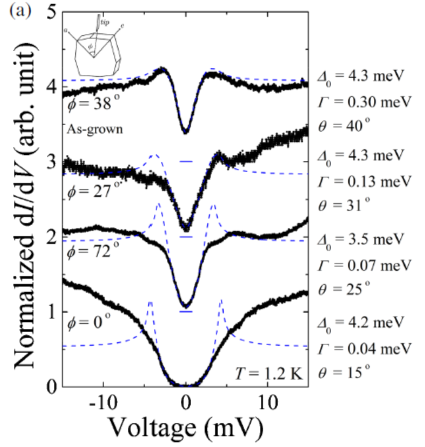
Malone *et al*., PRB 82, 014522 (2010).

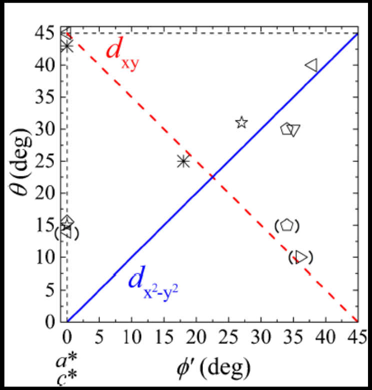
Imajo *et al*., PRB 103, L060508 (2021)

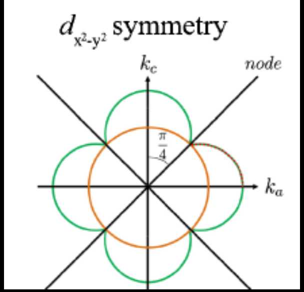
Superconducting gap: field-angle dependent measurements

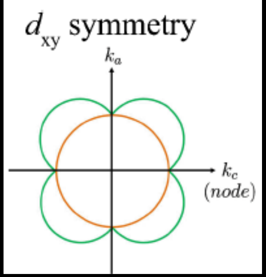
Phase separation of $d_{x^2-y^2}$ and d_{xy}

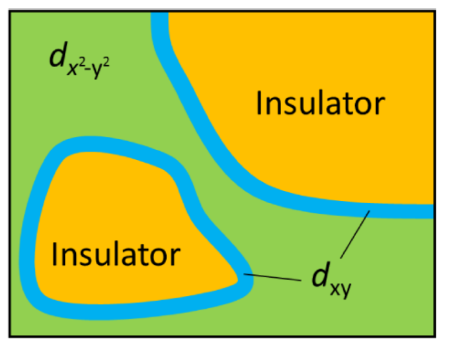
κ-(BEDT-TTF-d[3,3])$_2$Cu[N(CN)$_2$]Br







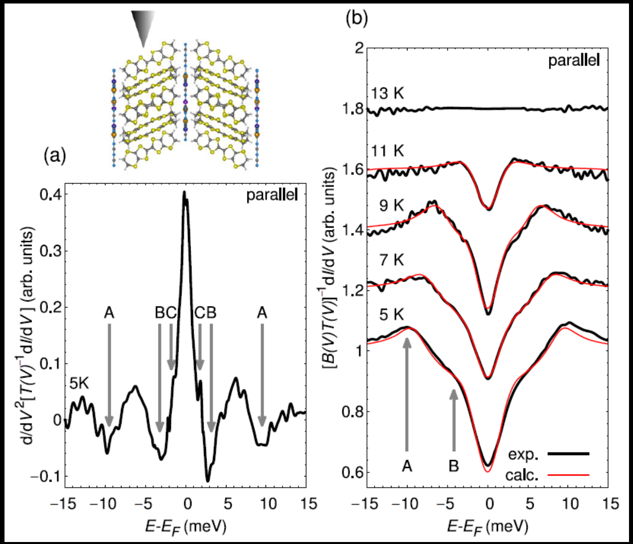


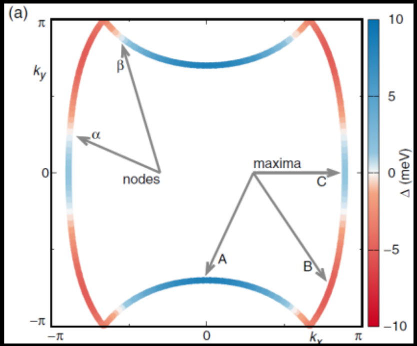


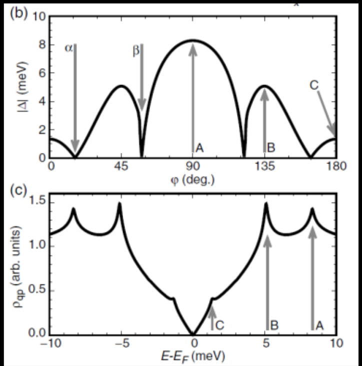
Oka et al., JPSJ 84, 064713 (2015)

Eight nodes

κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br





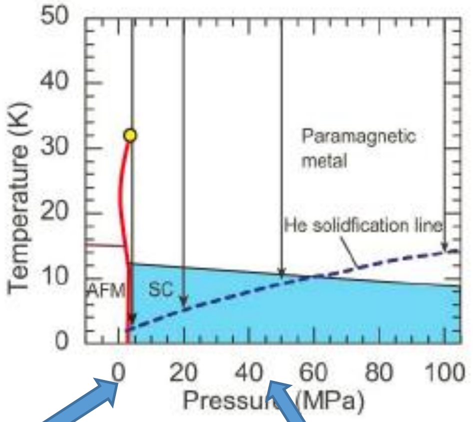


Guterding *et al.*, PRL 116, 237001 (2016)

Superconducting fluctuations persist up to twice as high as T_c

Preformed Cooper pairs

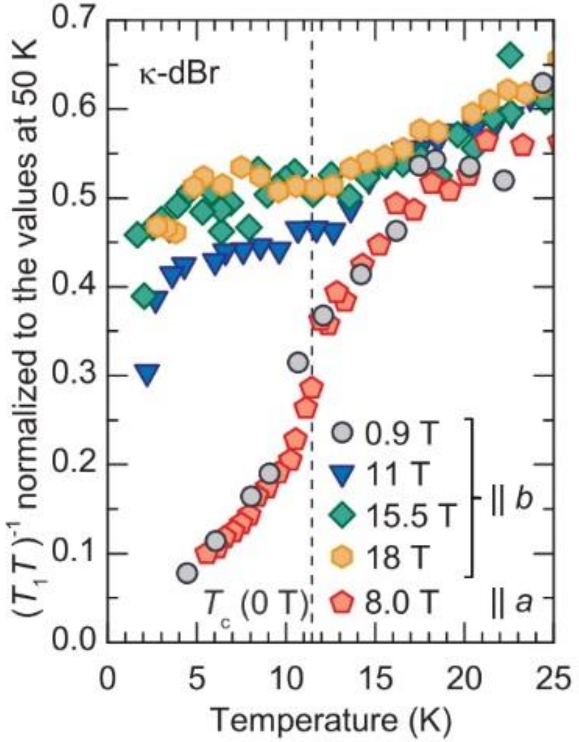
due to Correlation effect



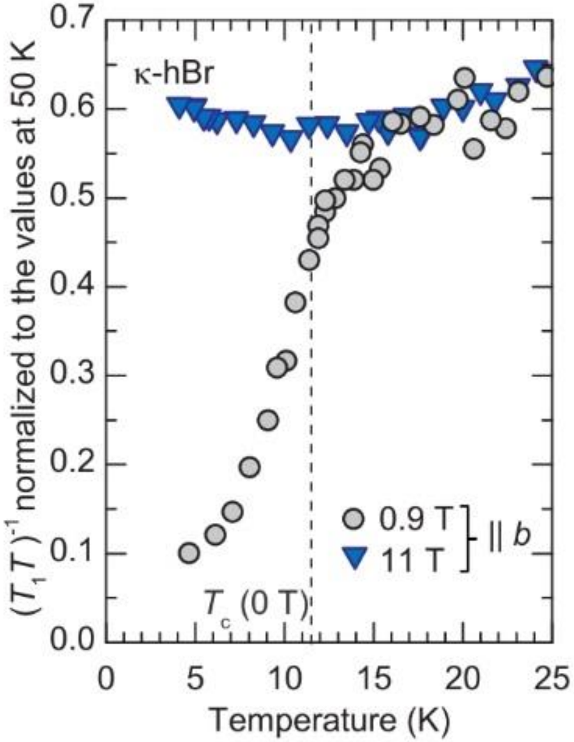
***NOT** due to low-D effect !*

More 2D-like

κ-(d[4,4], ET)$_2$Cu[N(CN)$_2$]Br



κ-(d[0,0], ET)$_2$Cu[N(CN)$_2$]Br



κ-(ET)$_2$Cu(NCS)$_2$

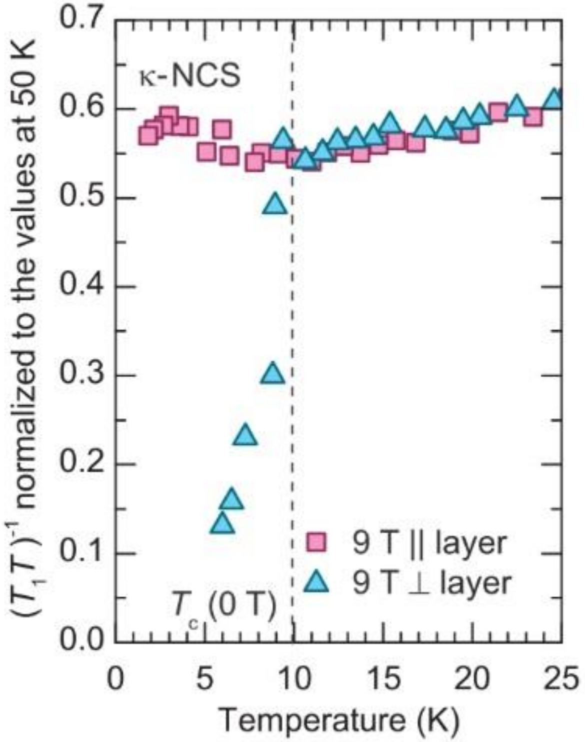
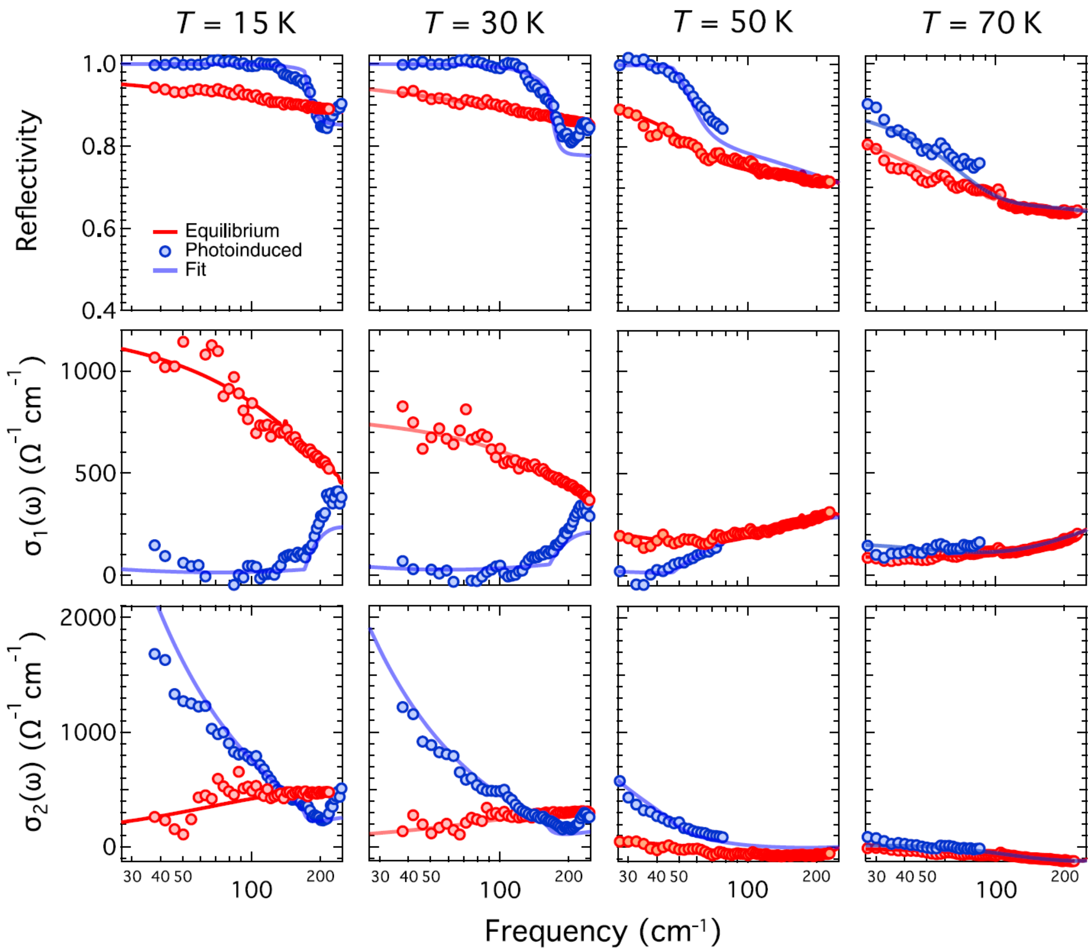
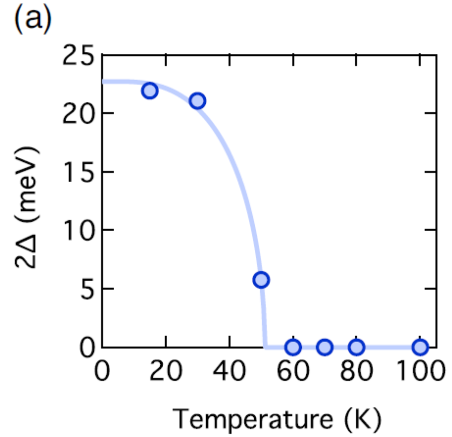


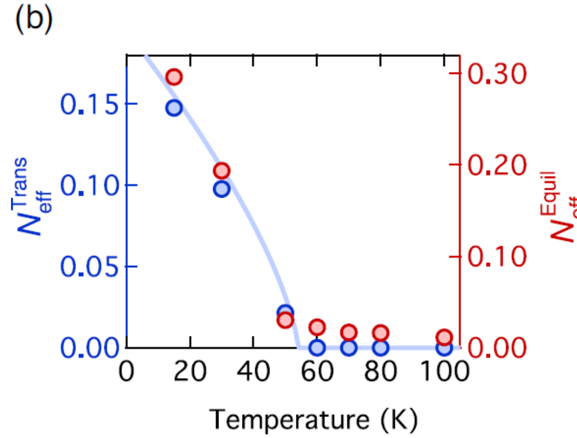
Photo-induced non-equilibrium superconductivity at 50 K

Midinfrared (($\nu_{\mathrm{pump}} \simeq 900\text{–}2000\ \mathrm{cm}^{-1}$) pumped teraheltz-probe optical conductivity

Blue spectra 1 picosec after photoexcitation

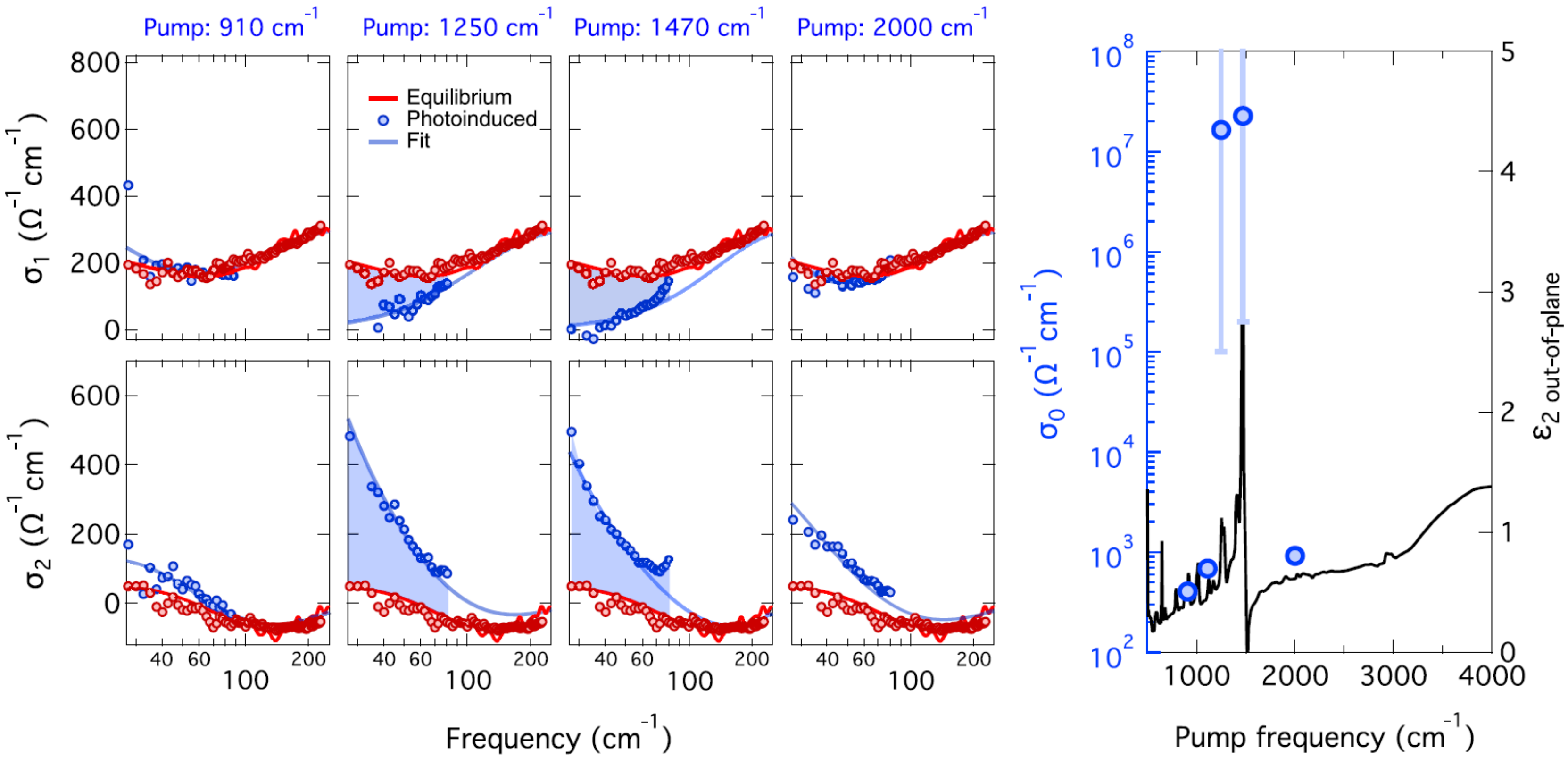






Buzzi *et al.*, PRX **10**, 031028 (2020)

Pump frequency dependence of σ_1 and σ_2 at 50 K



Buzzi *et al.*, PRX **10**, 031028 (2020)