Metallic ground state near the Mott transition in organic conductors probed by magnetic quantum oscillations

Sebastian Oberbauer, Shamil Erkenov, Werner Biberacher, Mark V. Kartsovnik

Walther-Meissner-Institute, Bavarian Academy of Sciences and Humanities, Garching, Germany

and

Natalia D. Kushch

Institute of Problems of Chemical Physics, Russian Academy of Sciences, Chernogolovka, Russia

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Outline

- Introduction to the κ-$(BEDT\text{-}TTF)_2X$ salts:
  - structure;
  - phase diagram: bandwidth-controlled metal-insulator transition (MIT)

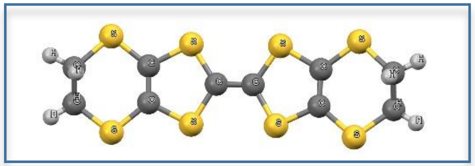
- Shubnikov-de Haas (SdH) oscillations in κ-Cl *near* & *at* the 1st-order MIT

- Effective cyclotron mass:
  - experiment vs. theory;
  - other materials

- Quasiparticle lifetime and possible pseudogap

- Summary

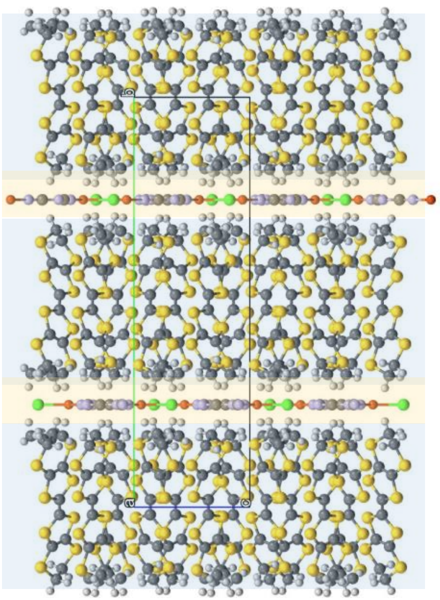
κ-(BEDT-TTF)$_2$X: basic features



BEDT-TTF (or **ET**): bis(ethylenedithio)tetrathiafulvalene

X : monovalent anion

Crystal structure

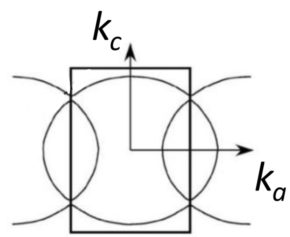


$\rho_b/\rho_{ac} \sim 10^3 - 10^4$

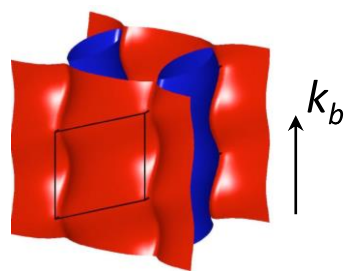
$\rho_{ac}(300\text{K}) \sim 10 - 20\ \text{m}\Omega\cdot\text{cm}$

$\rho(300\text{K})/\rho(4.2\text{K}) \sim 10^2 - 10^3$

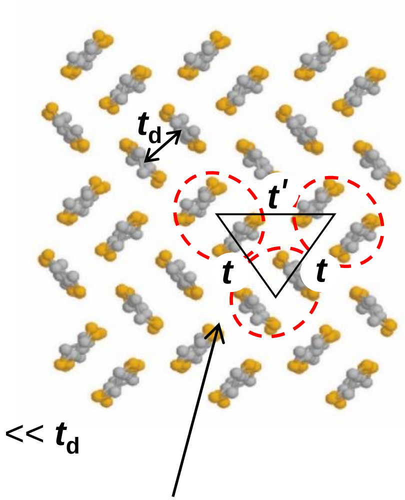
2D Fermi surface



3D Fermi surface



Conducting layer



t, $t' << t_d$

Anisotropic triangular lattice of dimers with one hole per dimer

⇒ narrow, ½-filled band

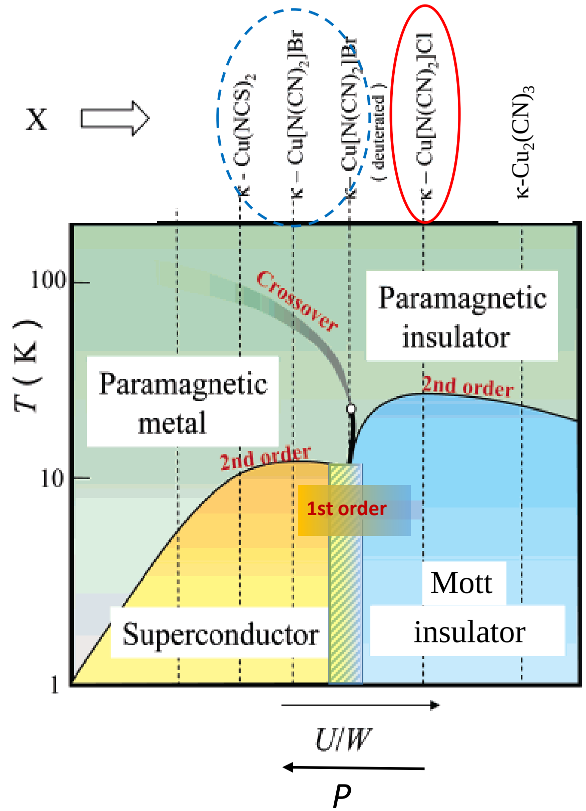
⇒ Mott instability

κ-(BEDT-TTF)$_2$X: basic features

Bandwidth-controlled
Mott-insulating instability

X = Cu[N(CN)$_2$]Cl [**κ-Cl**]

Cu[N(CN)$_2$]Br [κ-Br]
Cu(NCS)$_2$ [κ-NCS]
Cu$_2$(CN)$_3$ [κ-CN]



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

??

Evolution of metallic ground state in the very vicinity of the insulating state

Open issues

- <u>Does the Fermi surface keeps intact near the MIT?</u>

Theory predictions:

a strongly incoherent metal, $\tau^{-1} \sim 0.2t$ in the M/I coexistence region even at low *T*

*Park, Haule, & Kotliar, PRL **101**, 186403 (2008)*

or pseudogap & Fermi arcs

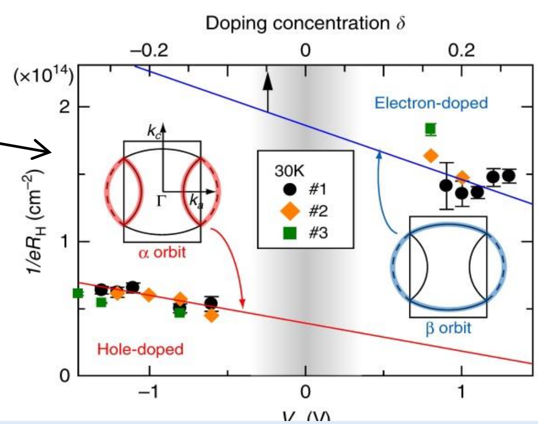
*Kang et al., PRB **84**, 064520 (2011); Merino et al., PRB **89**, 245130 (2014) , Tanaka, PRB **99**, 205133 (2019)…*

Possible experimental signs:

Hall coefficient in hole-doped (E-field effect) <u>κ-Cl</u> – only a part of the FS is restored

*Kawasugi et al., Nat. Comm. **7**, 12356 (2016)*

Band-*filling* control

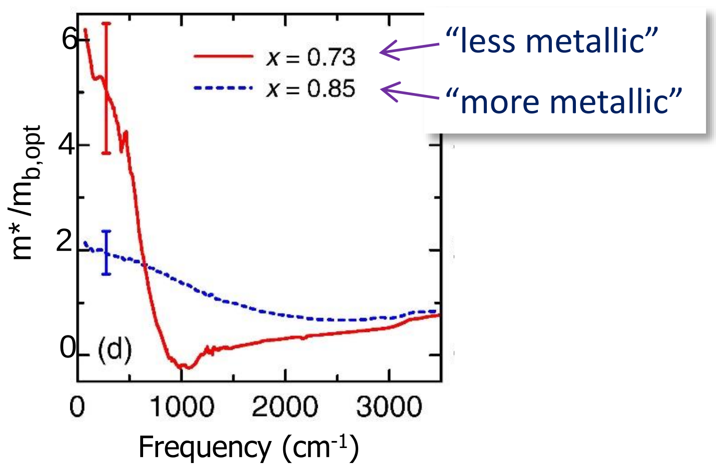


but what happens under <u>band-*width* control</u> (pressure)??

- <u>Effective mass renormalization</u>: a measure of electron interactions strength

κ-Br_xCl_{1-x}

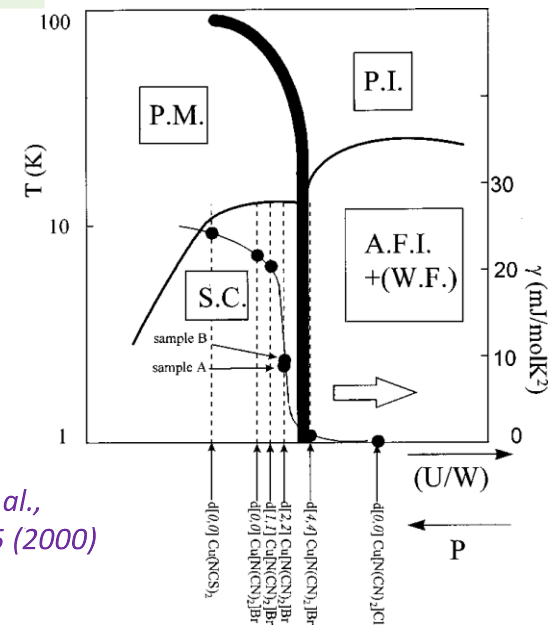
IR-optics



*Merino, Dressel, et al., PRL **100**, 086404 (2008)*

κ-Br with deuterated ET

Heat Capacity



*Y. Nakazawa et al., PRB **61**, R16295 (2000)*

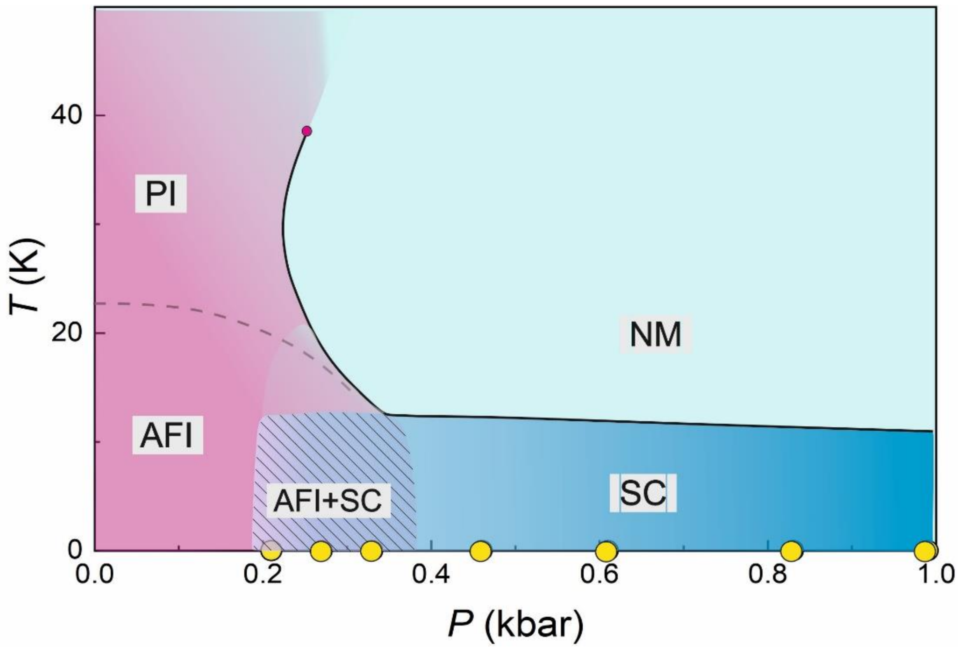
m^* strongly <u>increases</u> at approaching the MIT

but only 2 points…

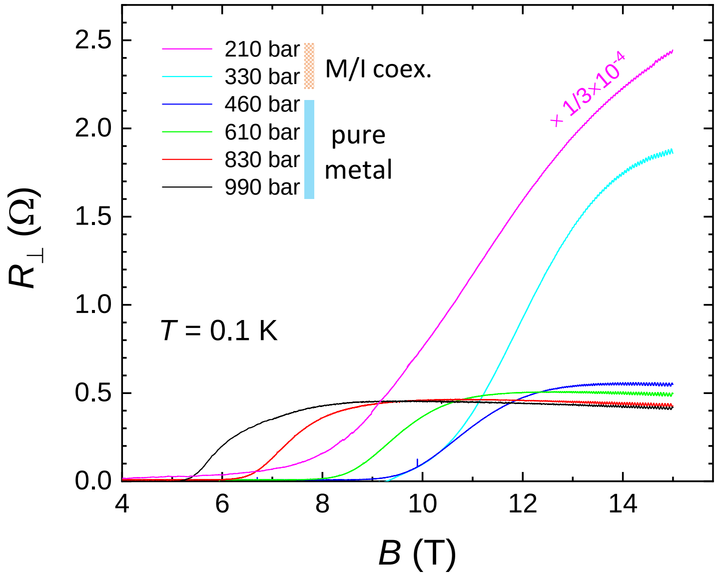
$\gamma \propto m^*$ <u>decreases</u> near the MIT

κ-Cl: SdH oscillations at different pressures

Phase diagram of κ-Cl

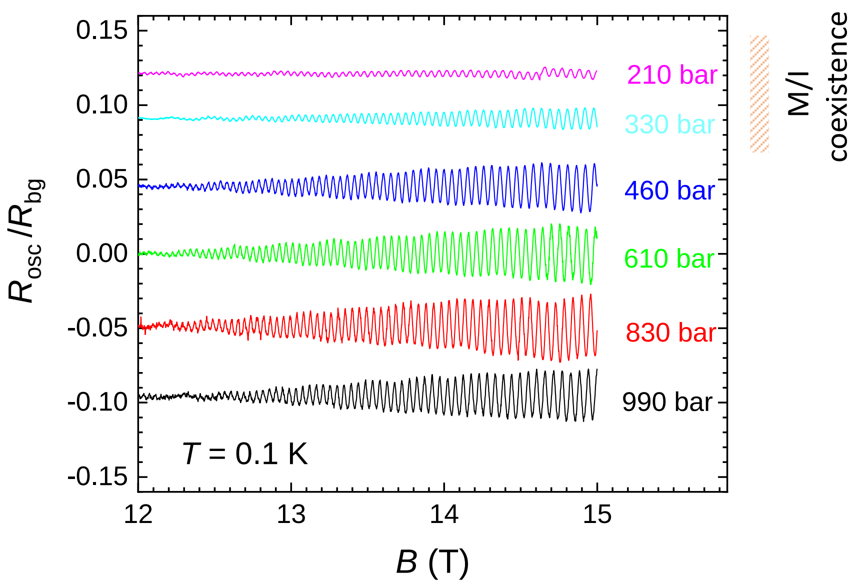


Field-dependent resistance

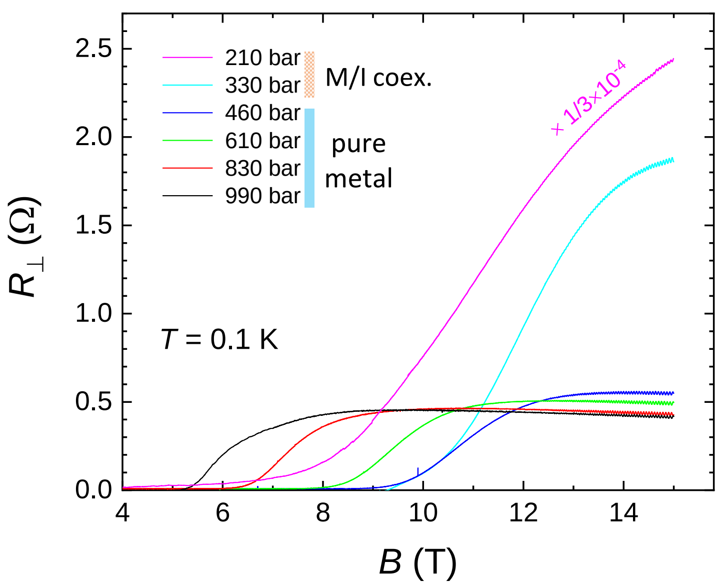


κ-Cl: SdH oscillations at different pressures

Shubnikov-de Haas (SdH) oscillations



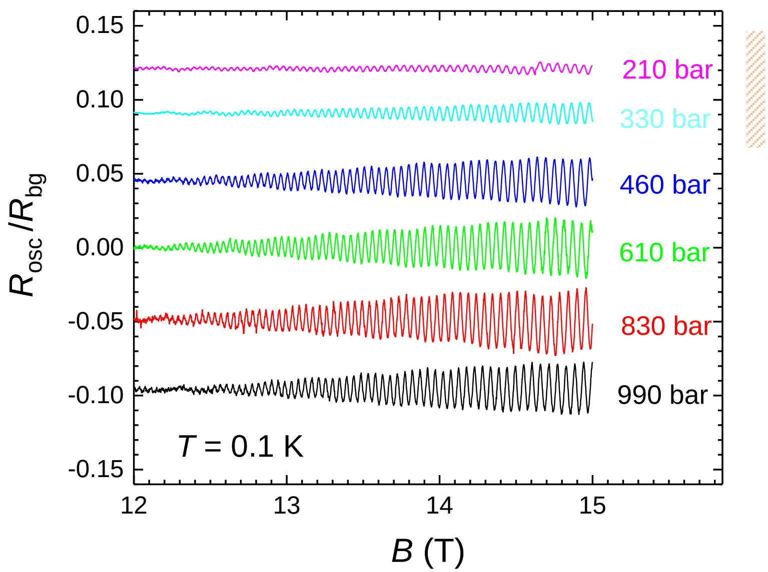
Field-dependent resistance

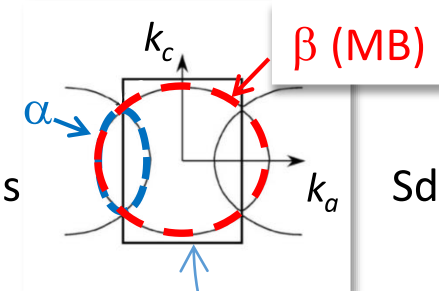


No dramatic change in the oscillation behavior

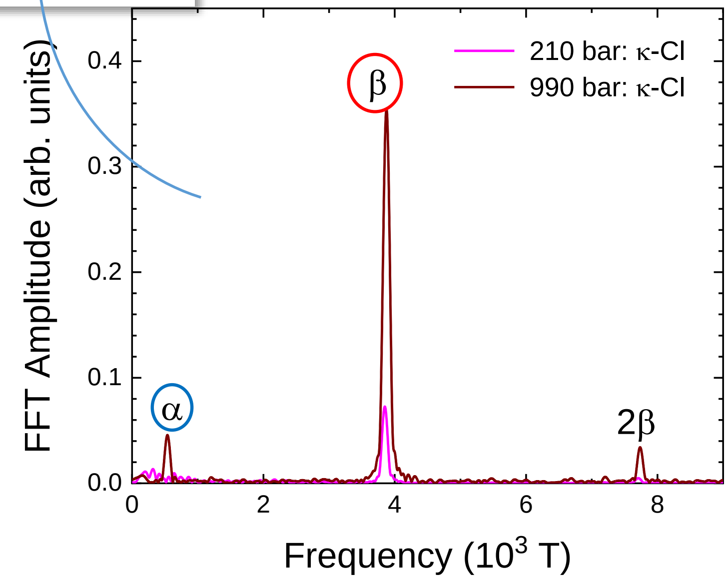
κ-Cl: SdH oscillations at different pressures

Shubnikov-de Haas (SdH) oscillations





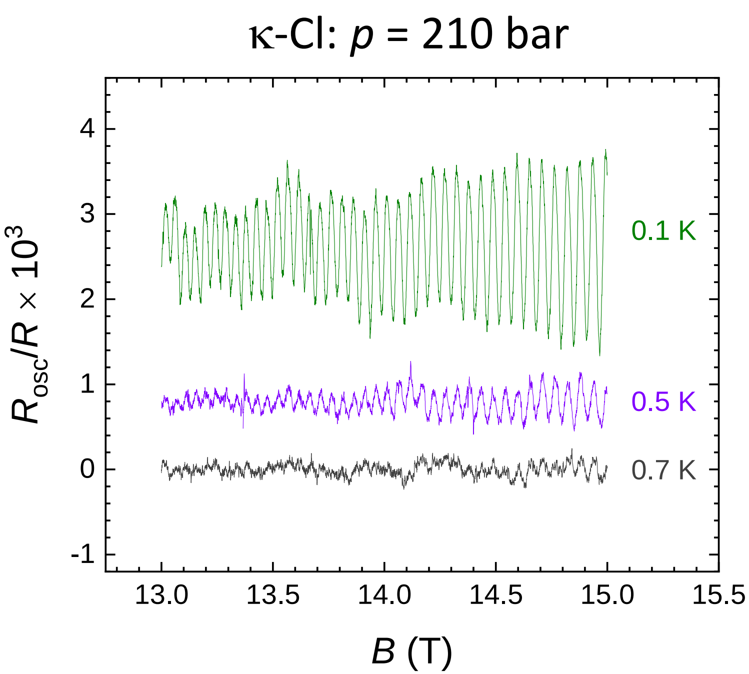
SdH frequencies

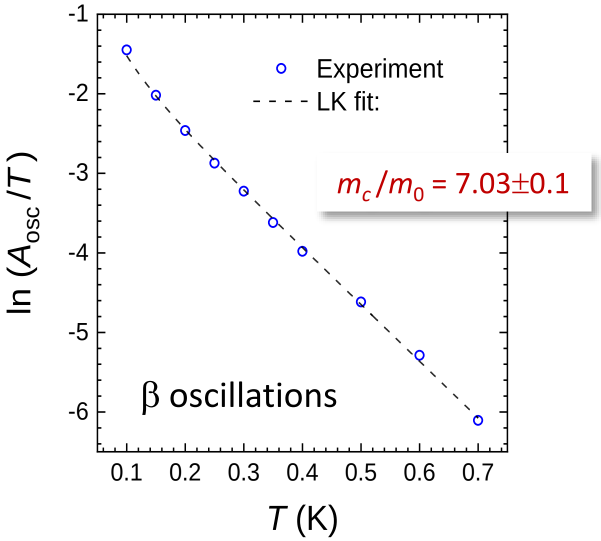


No dramatic change in the oscillation behavior

The large coherent Fermi surface is preserved

Effective cyclotron mass



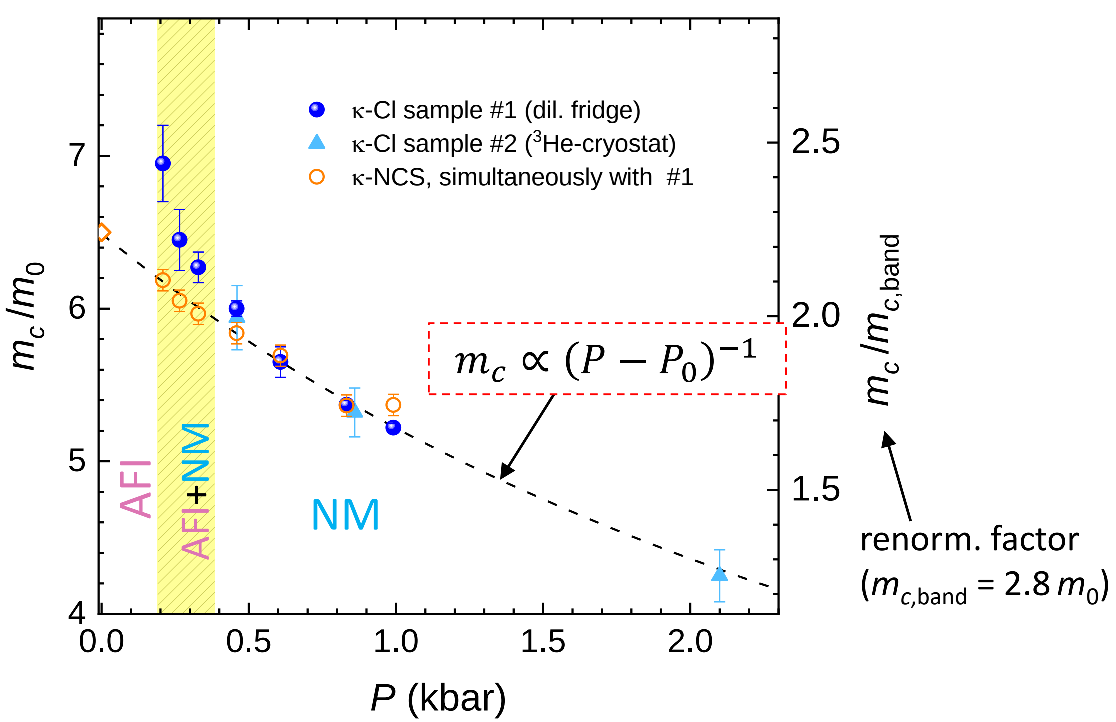


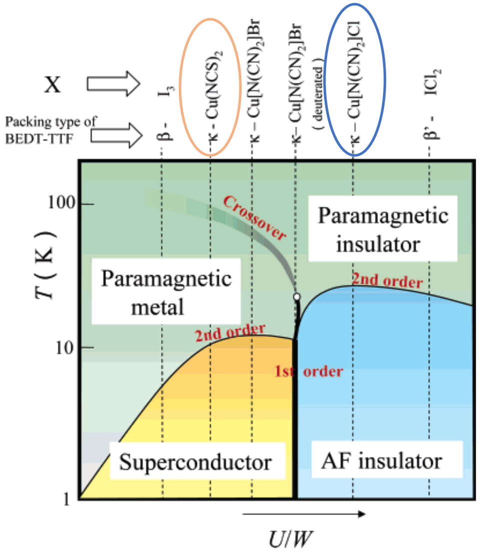
Lifshitz-Kosevich (LK) plot: $\frac{A_{\mathrm{osc}}}{T} \propto \frac{1}{\sinh(2\pi^2 k_{\mathrm{B}}T/\hbar\omega_c)} \approx \exp\left(-2\pi^2 \frac{k_{\mathrm{B}}T}{\hbar\omega_c}\right)$; $\hbar\omega_c = \frac{\hbar e}{m_c} B$

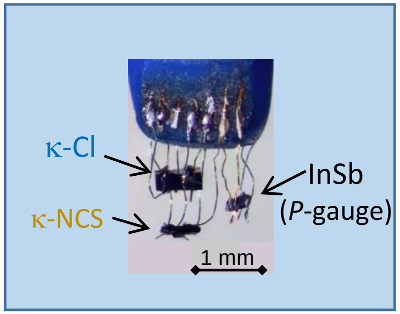
Effective cyclotron mass

Effective cyclotron mass

κ-Cl & κ-NCS







- The effective mass $m_c(p)$ steadily goes up at approaching the MIT, according to BR
- for κ-Cl the $m_c(p)$ dependence markedly curves up in the coexistence region

Effective cyclotron mass: experiment vs. theory

Theoretical predictions:

- Brinkman & Rice [*PRB* ***2****, 4302 (1970)*]: continuous Mott transition, no magnetic interactions,

  the mass diverges as: $\frac{m^*}{m_{\text{band}}} = Z^{-1} = \left[1 - \left(\frac{u}{u_c}\right)^2\right]^{-1} \approx \frac{1}{2}\left(1 - \frac{u}{u_c}\right)^{-1},$

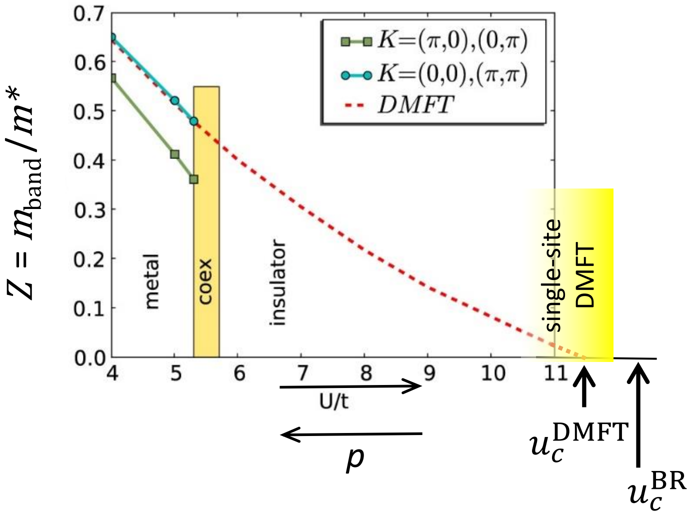
  Z – quasiparticle residue, $u \equiv U/t$ – interaction strength ratio

- present theories: - the divergence is cut-off at the 1st-order transition;
  - magnetic interactions shift the MIT to smaller U/t;
  - no dramatic difference in $Z(u)$.

e.g. CDMFT, with short-range magnetic correlations [*Park, Haule, & Kotliar, PRL* ***101****, 186403 (2008)*]:

At the transition:

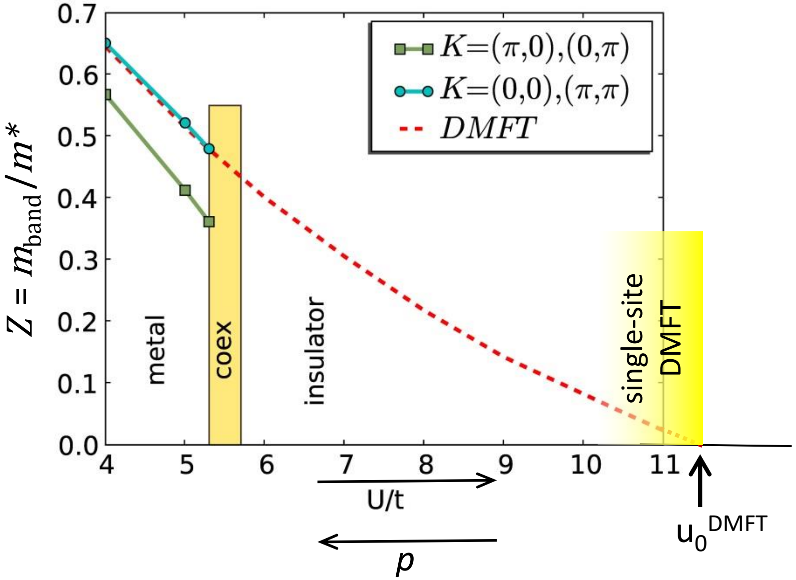
$\frac{m^*}{m_{\text{band}}} \sim 2-3$



$Z \cong 1 - \frac{u}{u_c}$

Effective cyclotron mass: experiment vs. theory

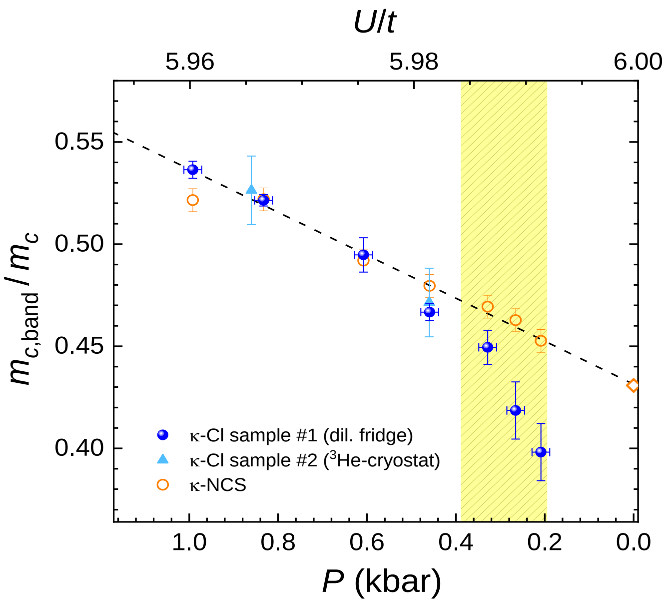
Theory:



$Z \cong C_Z(1 - u/u_0);$

$$C_Z = \begin{cases} = 2 & \text{(BR)} \\ \cong 1 & \text{(DMFT)} \end{cases}$$

Experiment on κ-Cl:



$$Z = \frac{m_{c,\text{band}}}{m_c} = \alpha(P - P_0) \cong C_Z(1 - u/u_0);$$

$P_0 = (-4.1 \pm 0.2)$ kbar;

$\frac{du}{dP} \cong 0.04\ \text{kbar}^{-1}$ from DFT calculations

[Kandpal et al., *PRB* ***2****, 4302 (1970)*]

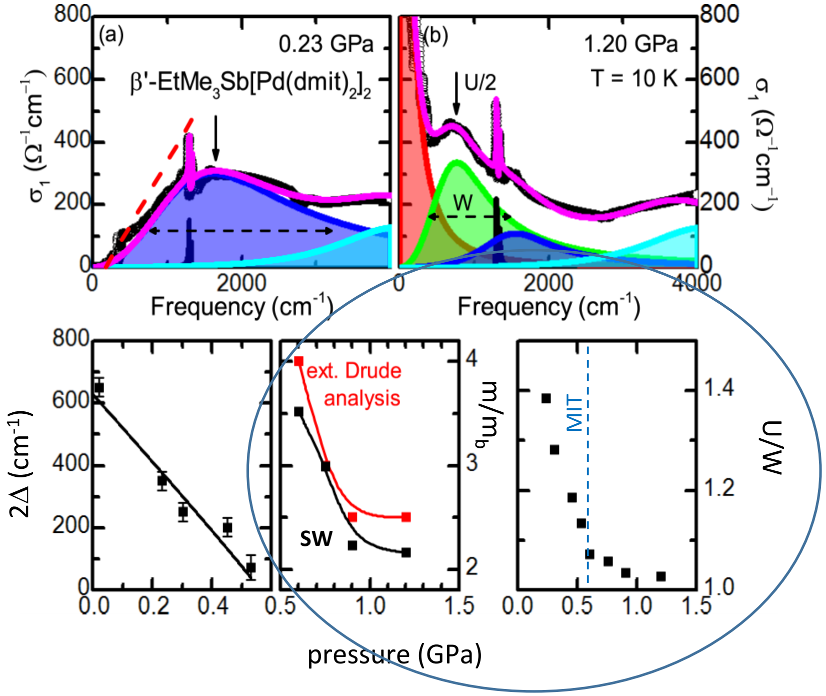
$\Rightarrow C_Z = 16.0 \pm 0.4$

Drastic discrepancy for the mass renormalization rate!

Effective cyclotron mass: other materials

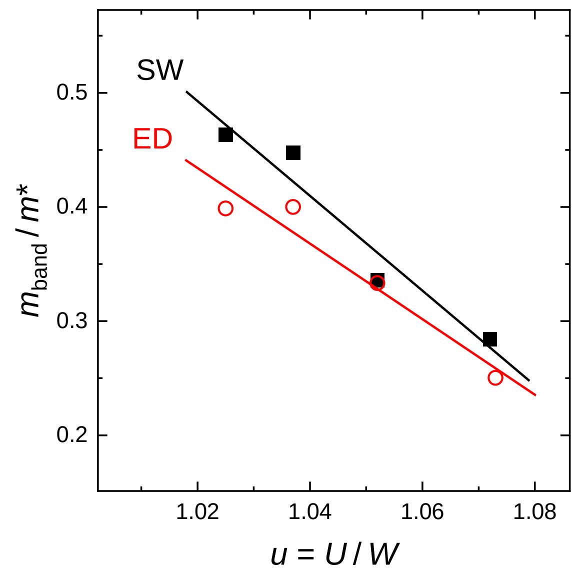
β'-EtMe$_3$Sb[Pd(*dmit*)$_2$]$_2$ – spin-liquid candidate

IR spectroscopy



Li et al., PRB ***99****, 115137 (2019)*

re-plotted



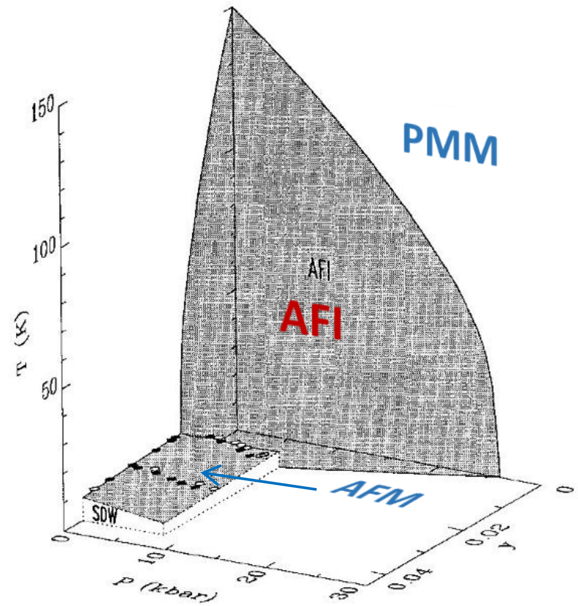
Fits: $\frac{m_{\text{band}}}{m^*} = C_Z(1 - u/u_0)$

$$C_Z = \begin{cases} 4.7 \pm 0.8; & \leftarrow \text{SW} \\ 3.8 \pm 0.9; & \leftarrow \text{ED} \end{cases}$$

$u_0 = 1.14 \pm 0.03$

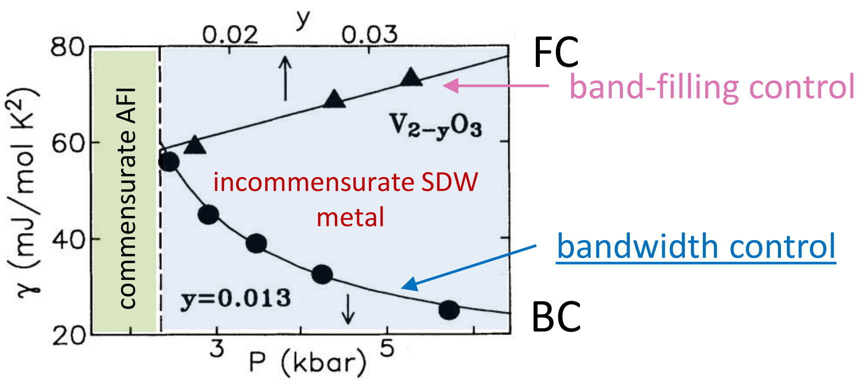
$C_Z \sim 4$ times bigger than DMFT

$V_{2-y}O_3$ – AF Mott insulator



*Bao et al., PRL **71**, 766 (1993)*

Heat capacity



*Carter et al., PRB **48**, 16841 (1993)*

- very rapid increase of the mass: 2 times over 3 kbar and
- and follows the simple BR-like relation:

$$m^* \propto \gamma = \gamma_s + \frac{\alpha\gamma_0}{(P-P_0)}$$

However,

– one-particle band mass unknown

– both the metallic and insulating states are AF-ordered

Summary

[S. Oberbauer et al., arXiv:2208.03230]

In the homogeneous metallic part of the phase diagram:

- the linear *P*-dependence of the inverse effective mass is apparently consistent with the theoretical predictions in close proximity to the Mott transition;
- however, the slope of this dependence is much steeper than expected from theory.

In the metal-insulator coexistence region:

- the inplane charge transport remains coherent; the Fermi surface is fully preserved; the scattering rate does not noticeably increase;
- the effective mass enhancement is significantly accelerated in comparison to the purely metallic region – still to understand!

Thanks for your attention!

Coming next…

Similar experiments on

- κ salts with different degrees of geometrical frustration, e.g. spin-liquid compounds;
- magnetic interactions, e.g., with localized paramagnetic moments: $(BETS)_2FeX_4$, κ-$(BETS)_2Mn[N(CN)_2]_3$