Transition métal-non métal dans le composé $(BEDT\text{-}TTF)_8Hg_4Br_{12}(C_6H_5Br)_2$

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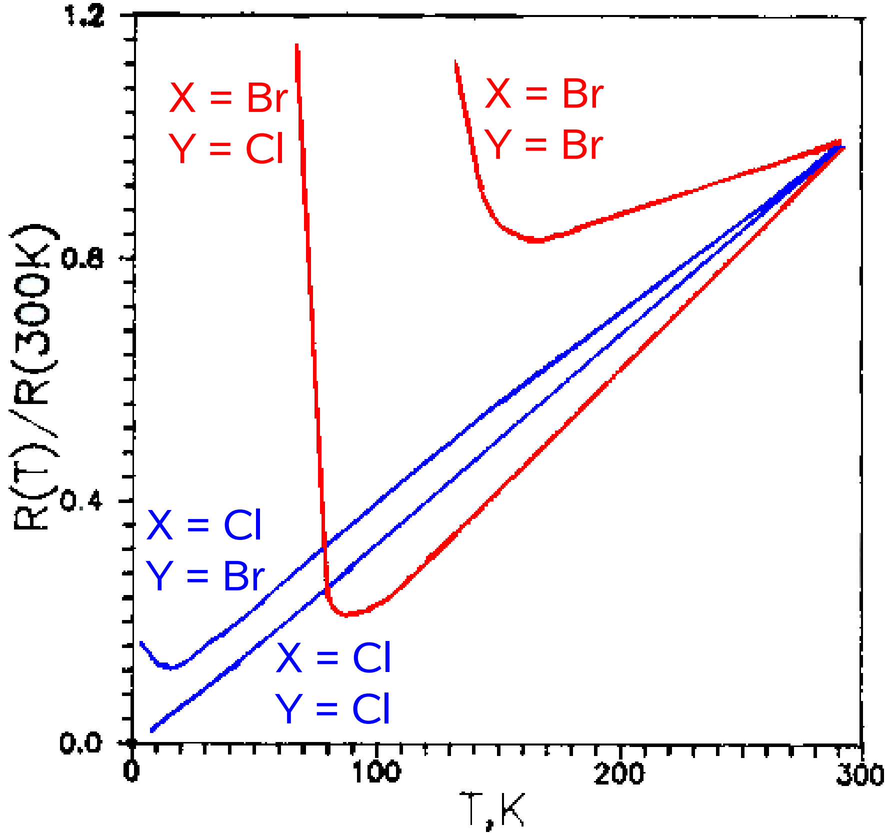
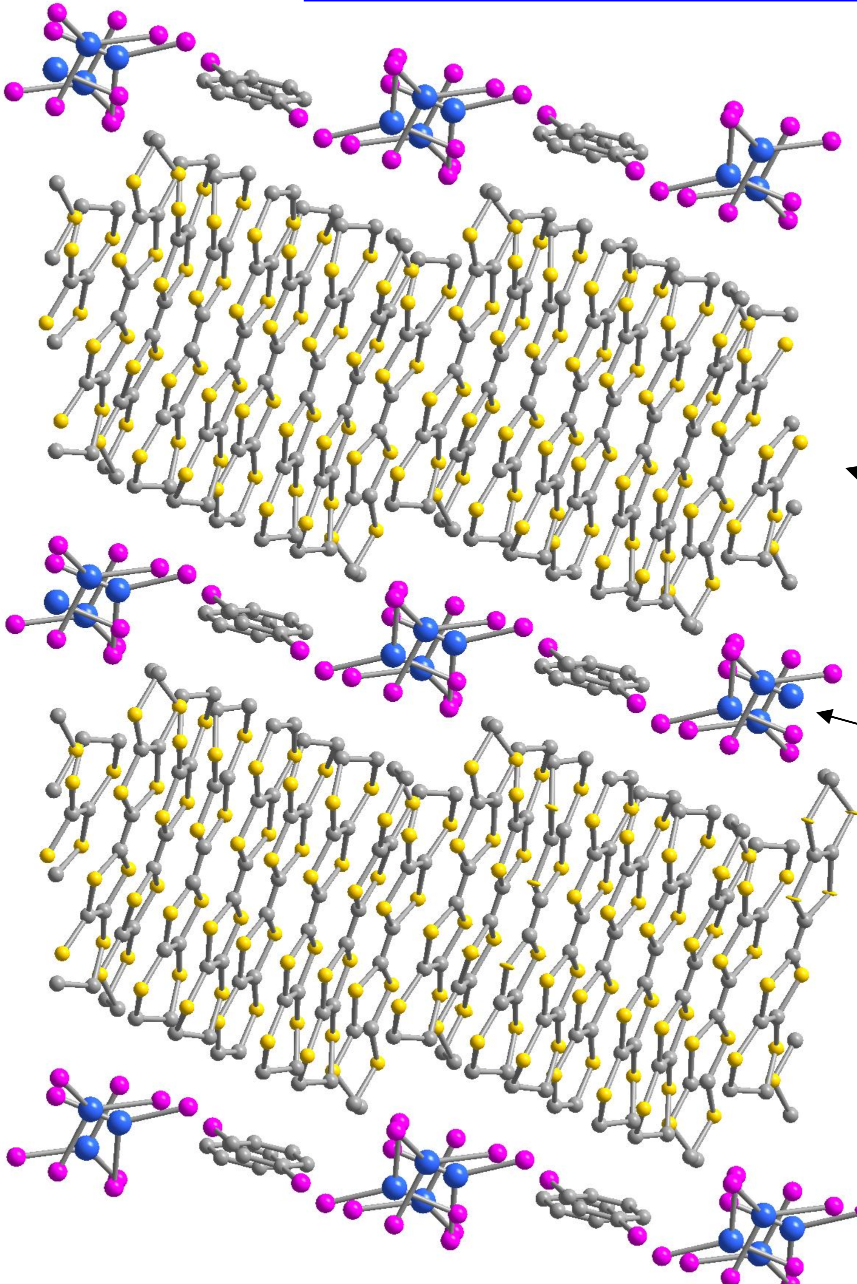
Motivation:
$(BEDT\text{-}TTF)_8Hg_4\mathbf{X}_{12}(C_6H_5\mathbf{Y})_2$ family, where (X, Y) = (Cl, Br)



X = Cl: metallic ground-state
X = Br: metal-non metal transition
even though the four members of this family are isostructural at room temperature

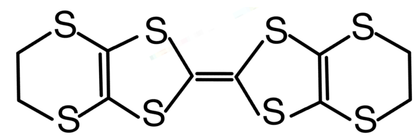
R. Lyubovskaya et al. Synth. Met. ***55-57*** *2899 (1993)*

$(BEDT\text{-}TTF)_8Hg_4X_{12}(C_6H_5Y)_2$: crystalline structure



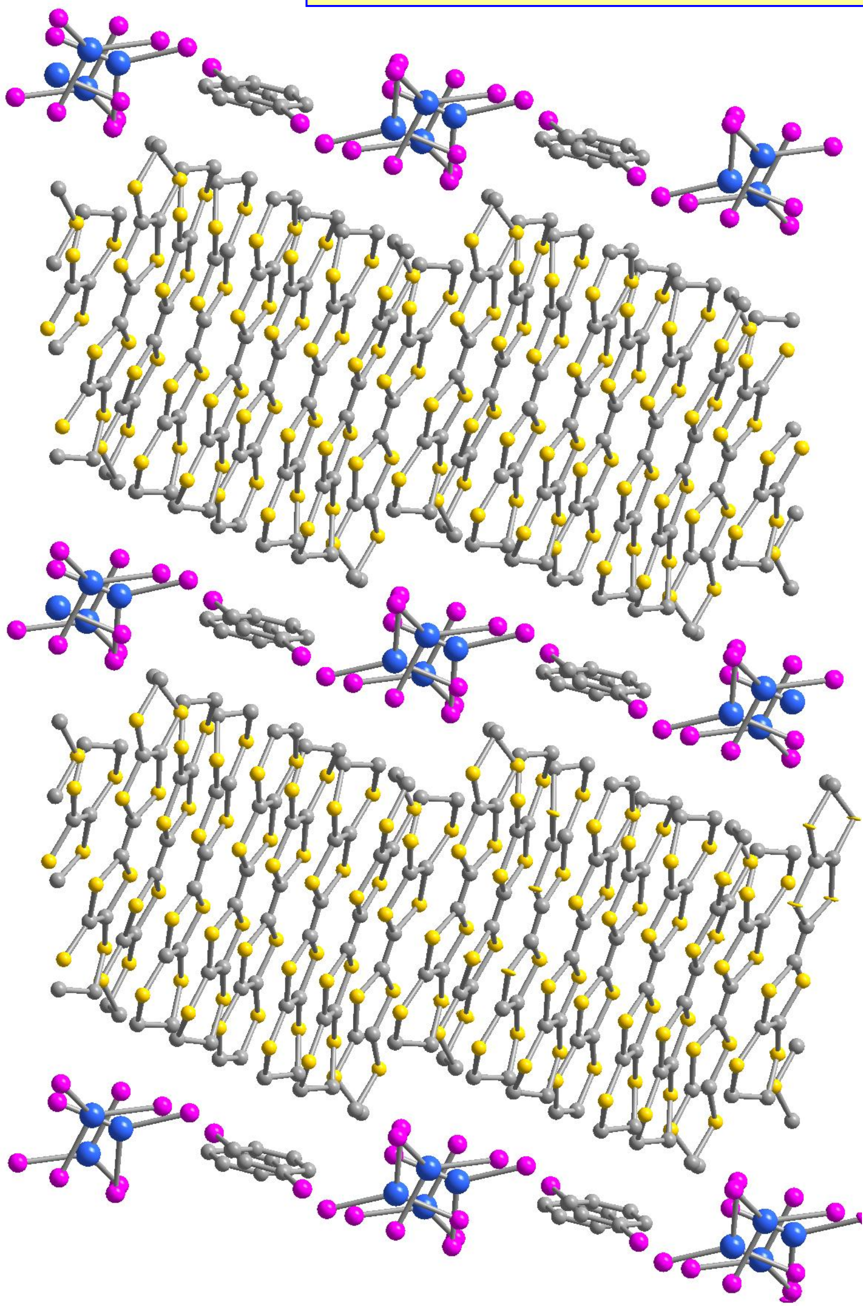
X = Y = Br (150 K)

conducting planes:
$(BEDT\text{-}TTF)^{0.5+}$ cation layers

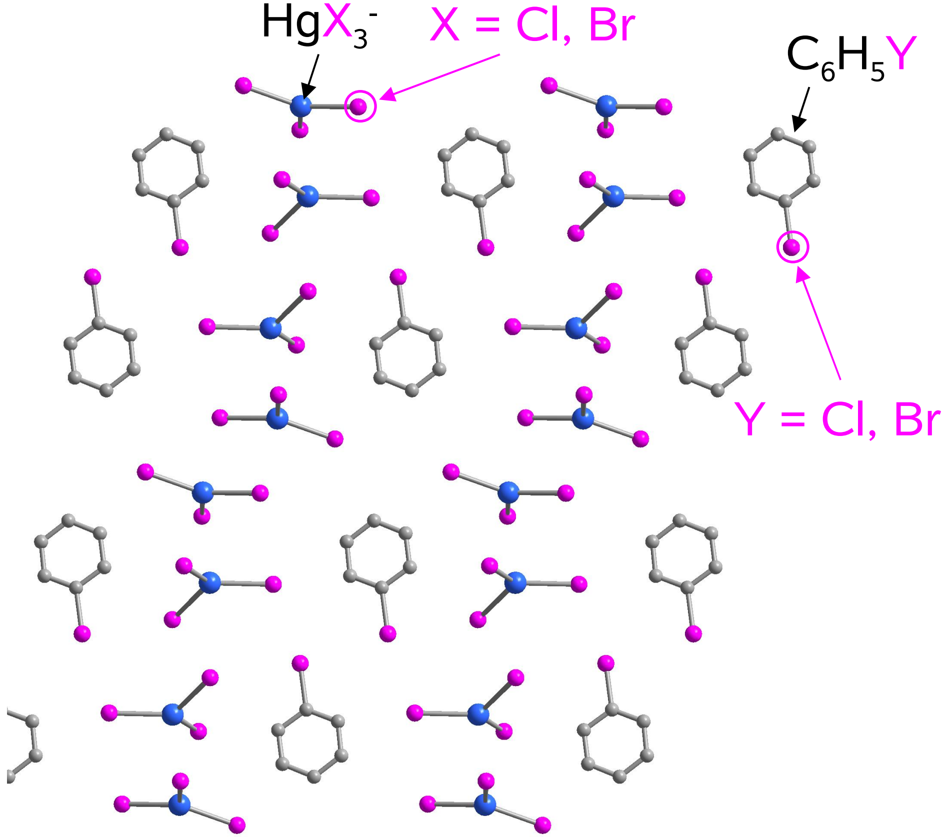


Insulating planes:
HgX_3^- anion + C_6H_5Y (solvent)

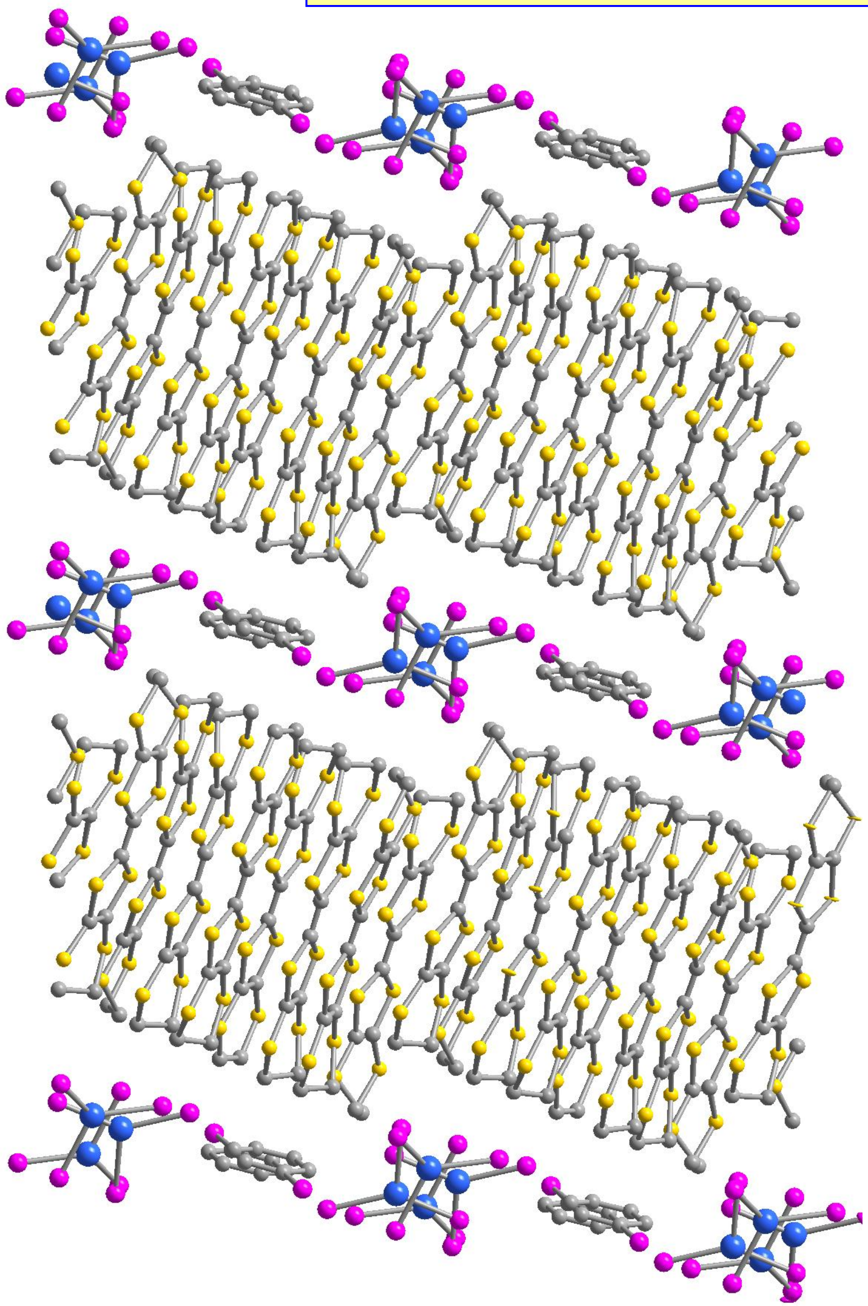
$(BEDT\text{-}TTF)_8Hg_4X_{12}(C_6H_5Y)_2$: crystalline structure

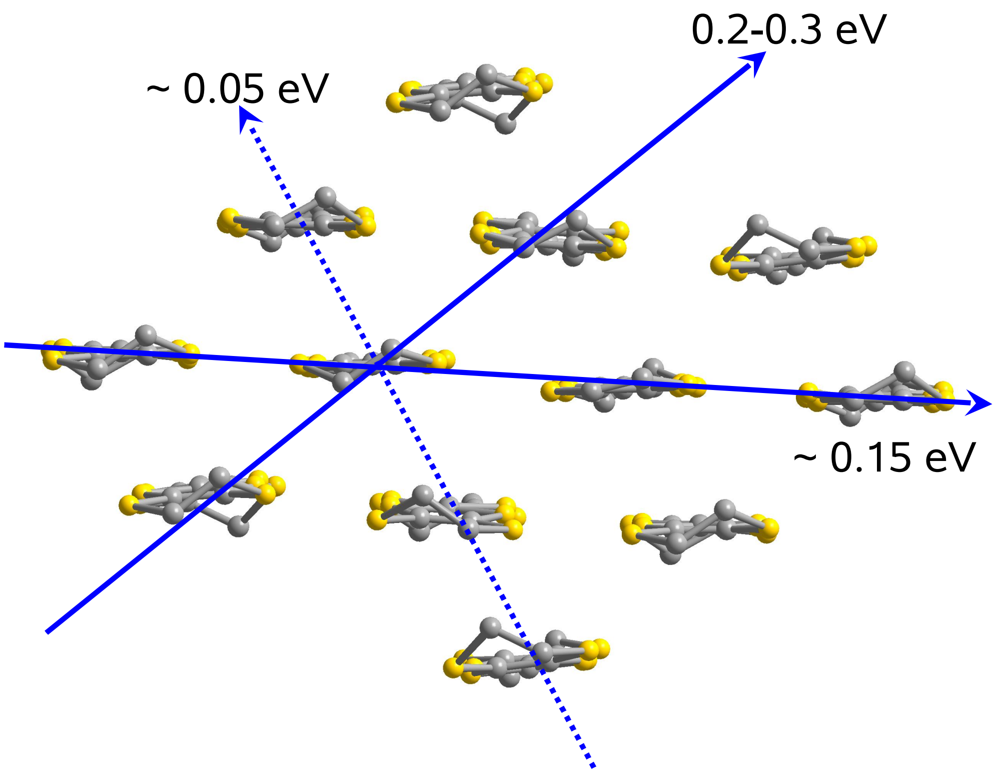


insulating layer

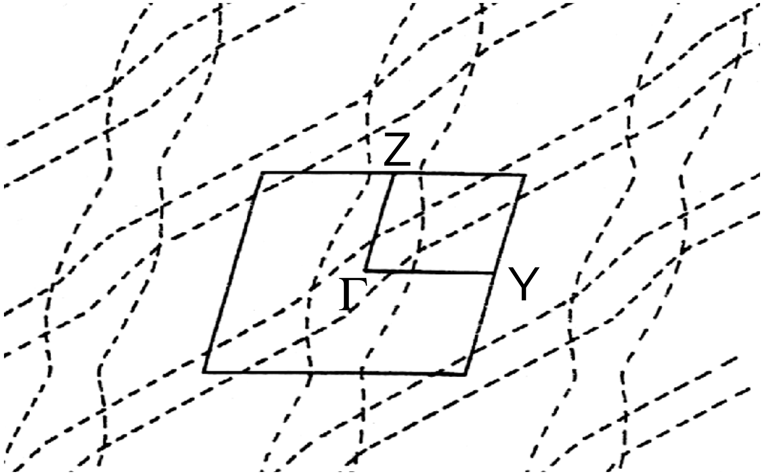


$(BEDT\text{-}TTF)_8Hg_4X_{12}(C_6H_5Y)_2$: crystalline structure





Electronic structure (Extended Hückel)

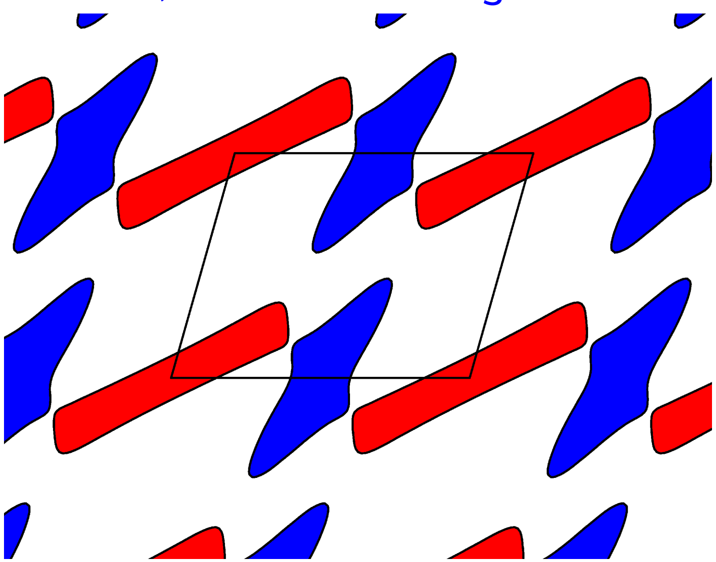


Two pairs of crossing q-1D sheets

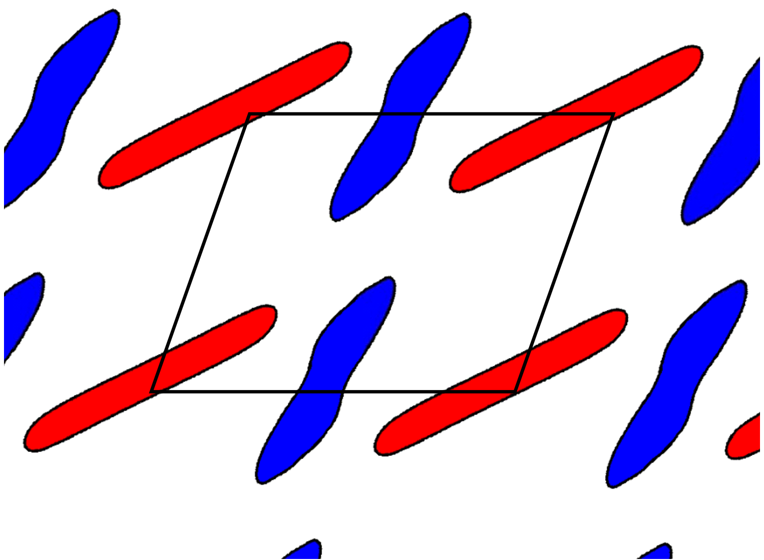
Degeneracy removals yields compensated electron and hole orbits

→ 2D Fermi surface

X = Cl, Y = Br: metallic ground-state



X = Y = Br:
metal non-metal transition at ~150 K.
Hidden nesting → density wave ?



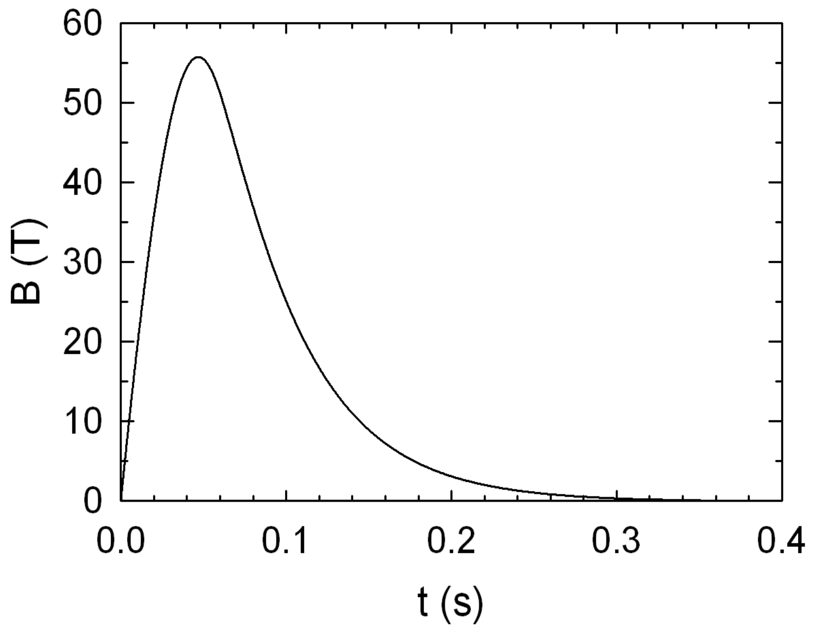
Experiments

Quantum oscillations

closed orbits → test for Fermi surface

- Periodic in 1/B
- Frequency proportional to orbit area (in k-space)
- Temperature and field-dependence yield physical parameters

Interlayer magnetoresistance in pulsed magnetic field (55 T)
→ Shubnikov-de Haas oscillations

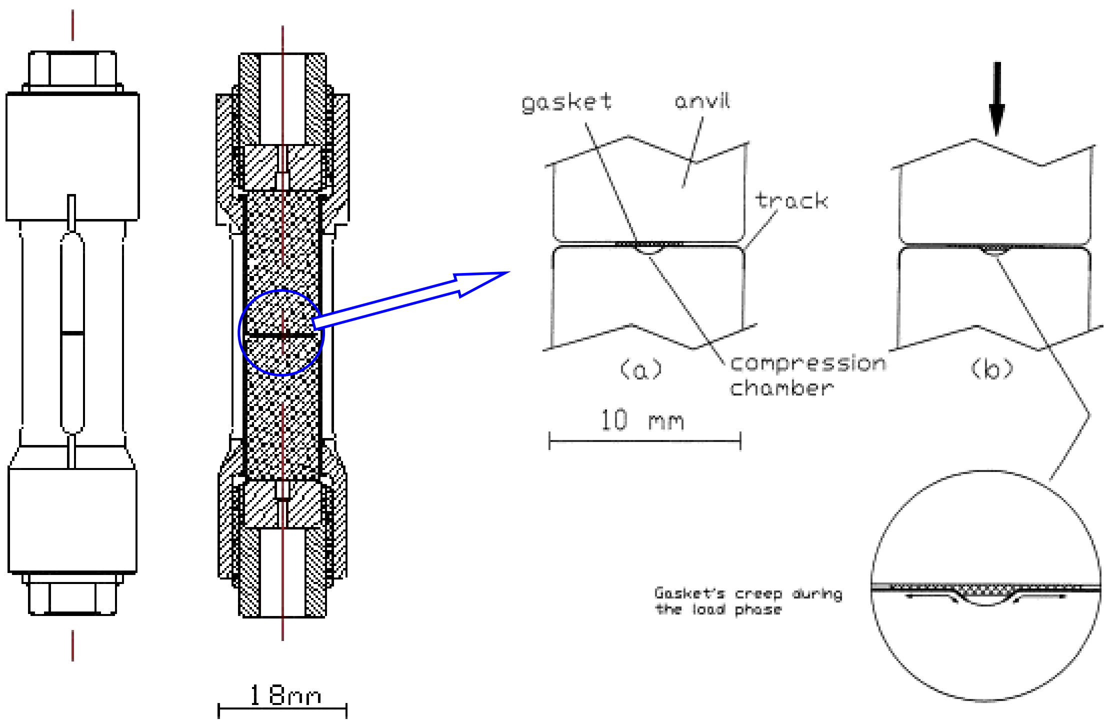


Experiments

Applied pressure powerful tool for tuning physical properties

Need for isothermal transport measurements at liquid helium temperatures in pulsed fields

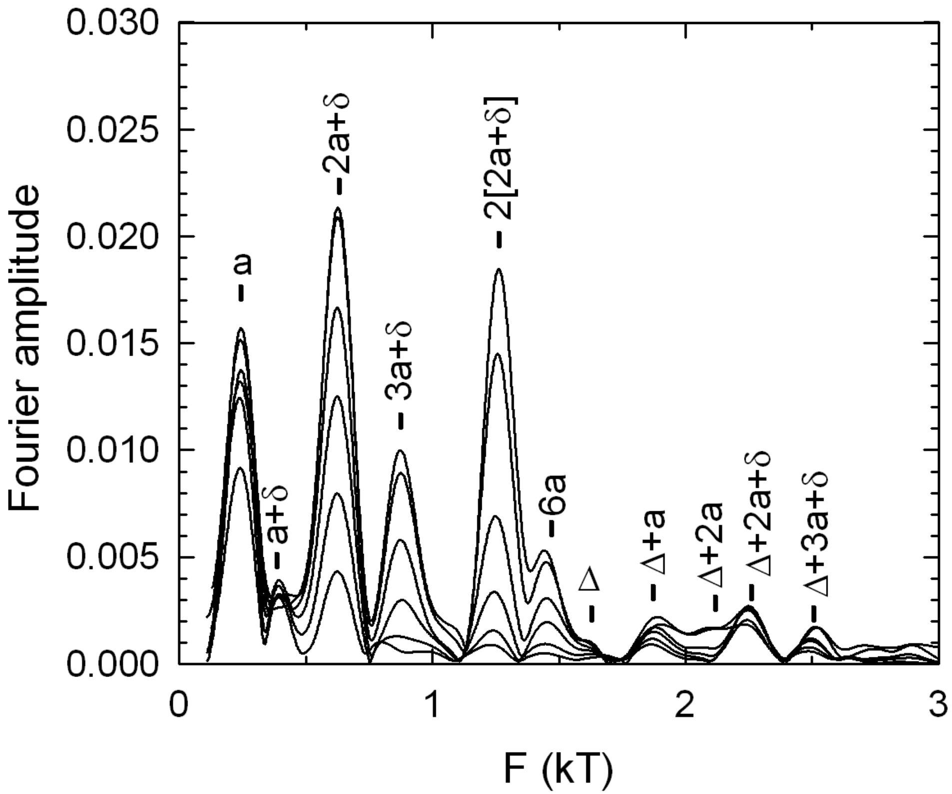
Pressure cell: zirconia anvils – Pmax = 1.2 GPa



*M. Nardone et al. Cryogenics **41** 175 (2001)*

Ambient pressure data for X = Cl: metallic ground state

Fourier analysis

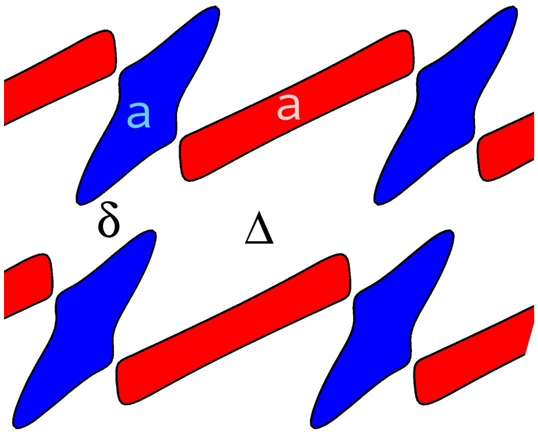


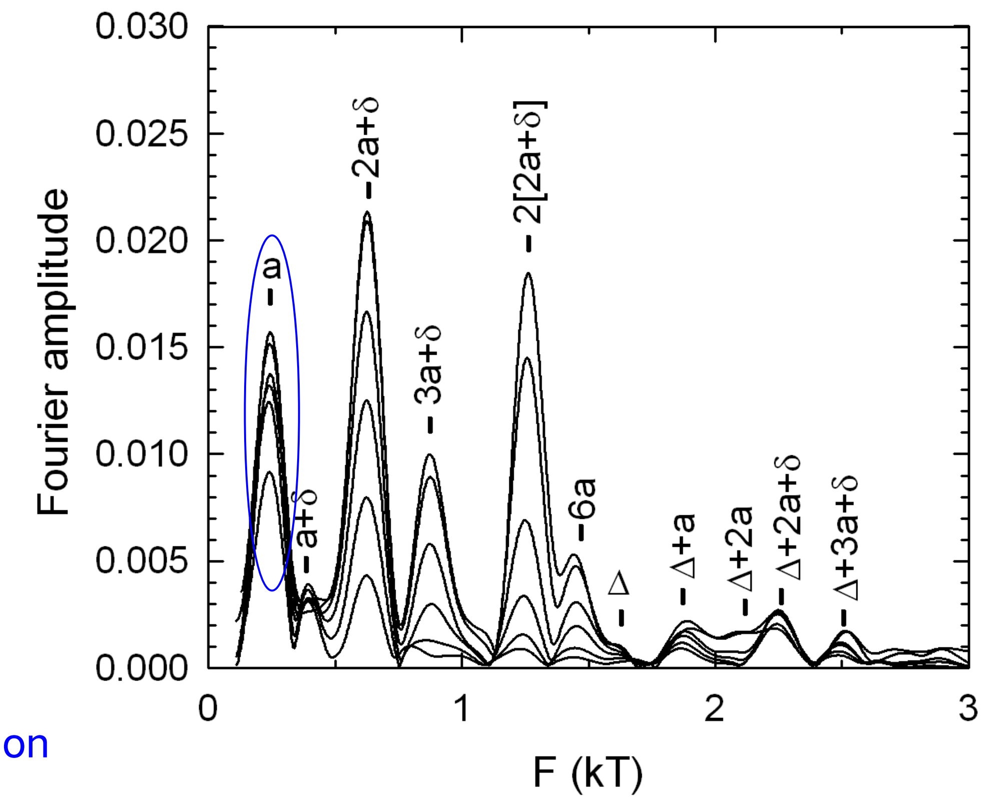
a lot of frequencies are observed

*Vignolles et al. Eur. Phys. J. B **31** 53 (2003)*

Ambient pressure data for X=Cl, Y=Br (metallic ground state)

Linear combinations of the frequencies linked to the basic electron and hole orbits (a) and of the δ and Δ pieces .





Closed magnetic breakdown orbits
Quantum interference paths

Forbidden orbits
- Landau bands
- Chemical potential oscillation

Model system for the study of quantum oscillations in networks of coupled orbits

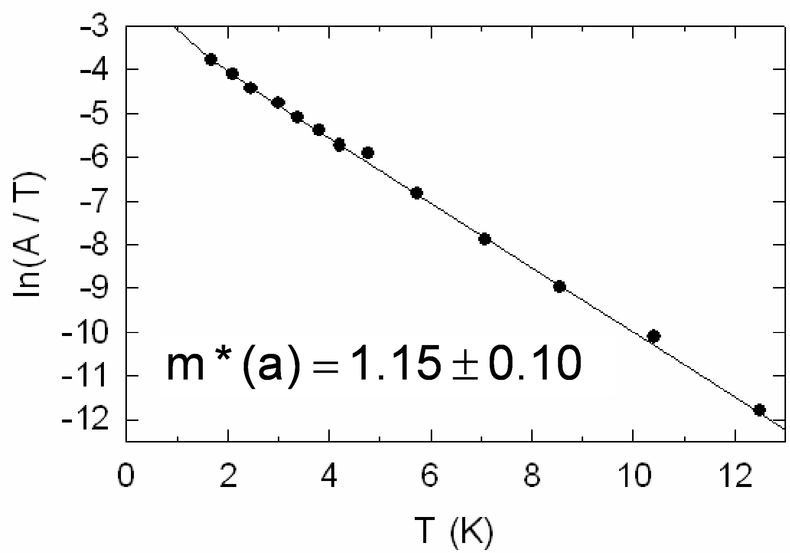
*Vignolles et al. Eur. Phys. J. B **31** 53 (2003)*

Derivation of physical parameters: Lifshits-Kosevich formalism

Fourier amplitude: $A \propto R_T R_D R_{MB}$

at a given field value: Fermi-Dirac smearing $A \propto R_T = \frac{Tm^*}{B} \Big/ \sinh\left(\frac{u_0 Tm^*}{B}\right)$

temperature dependence yields effective mass (m*)



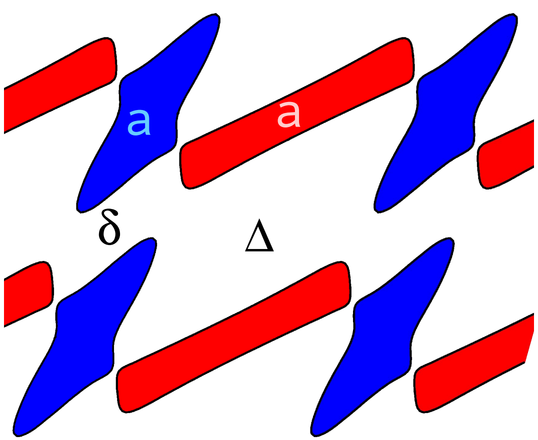
Derivation of physical parameters: Lifshits-Kosevich formalism

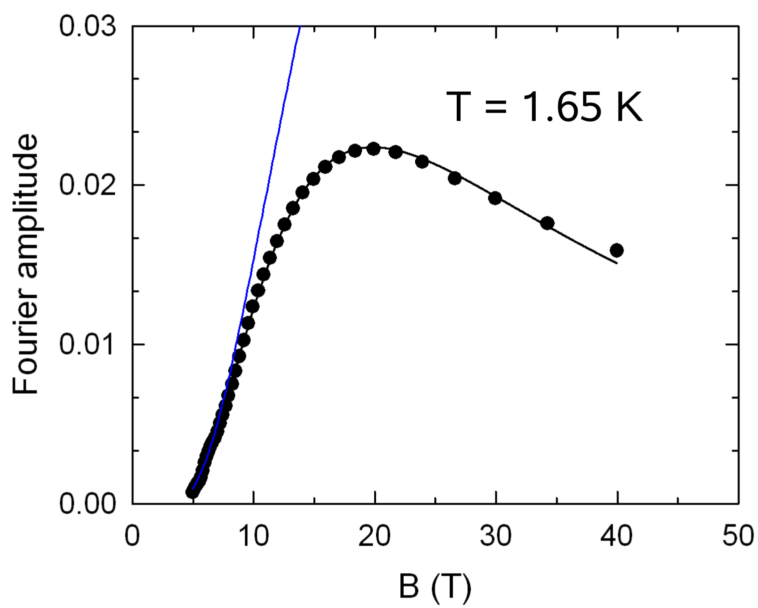
Fourier amplitude: $A \propto R_T R_D R_{MB}$

At a given temperature:

Dingle damping factor: $R_D = \exp\left(-\frac{u_0 T_D m^*}{B}\right)$ $T_D = \frac{\hbar}{2\pi k_B \tau}$ τ: scattering time

Magnetic breakdown damping factor: $R_{MB} = \left[1 - \exp(-B_0/B)\right]^2$ B_0: MB field





$B_0 = (35 \pm 10)$ T

$T_D = (0.2 \pm 0.2)$ K

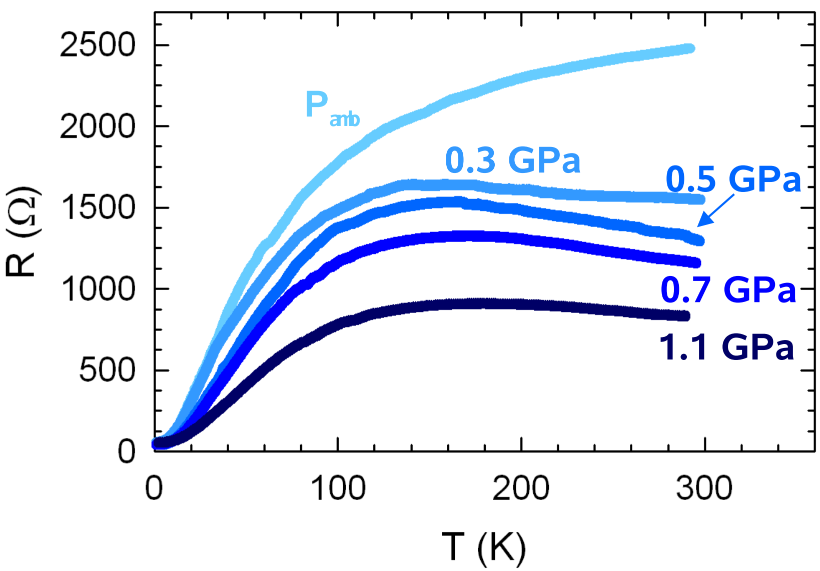
↓

$\tau^{-1} = (1.7 \pm 1.7) \times 10^{11} s^{-1}$

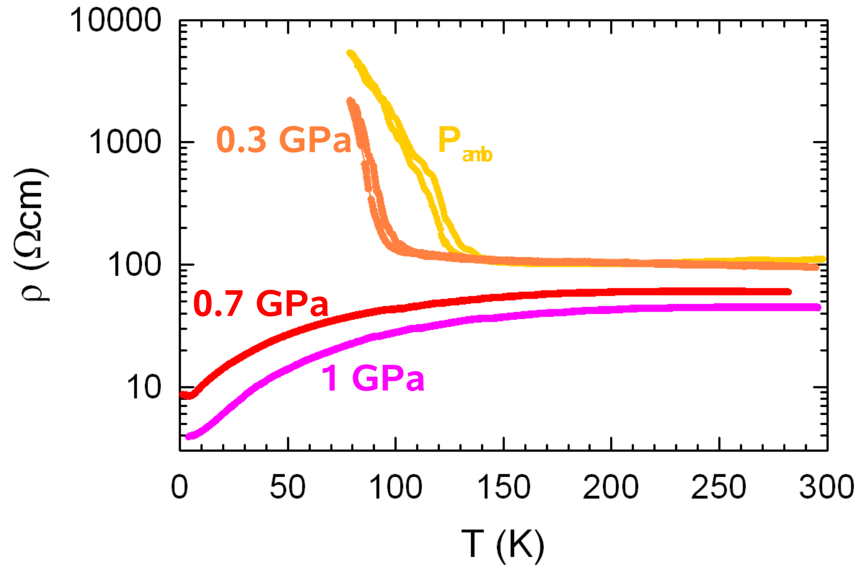
very good crystals!

Effect of applied pressure: zero-field resistance

X = Cl (metallic ground state)



X = Br: pressure-induced metallic state

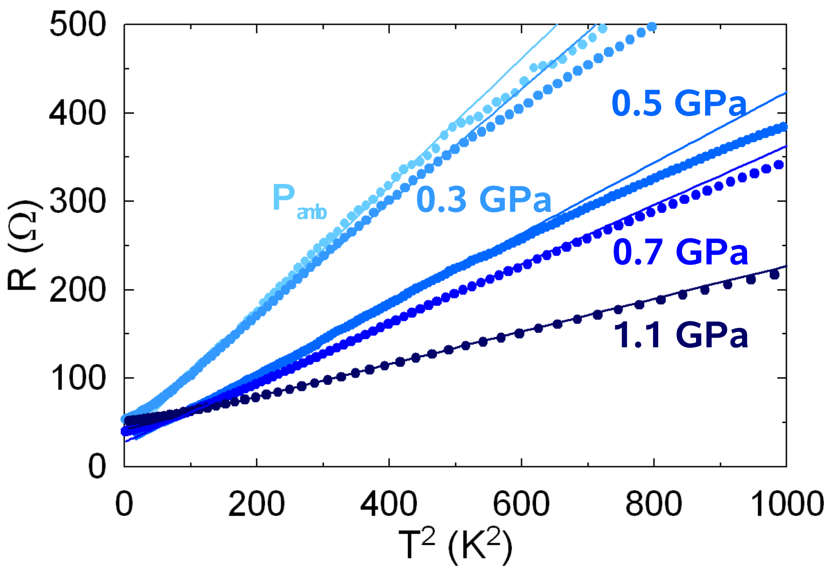


X = Cl: bump under pressure
Defects and (or) correlations

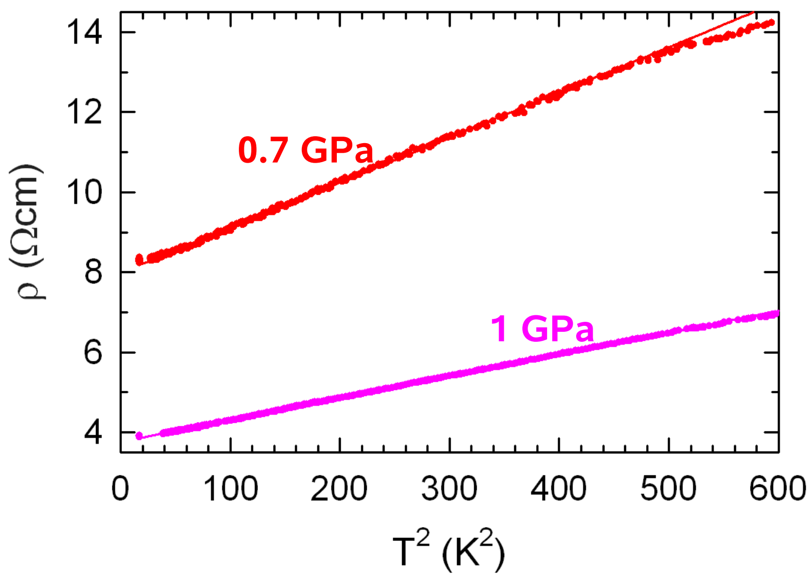
X = Br at low pressure: hysteresis
at high pressure: metallic state

Effect of applied pressure: zero-field resistance at low temperature

X = Cl (metallic ground state)



X = Br: pressure-induced metallic state



X = Cl and X = Br at high pressure:

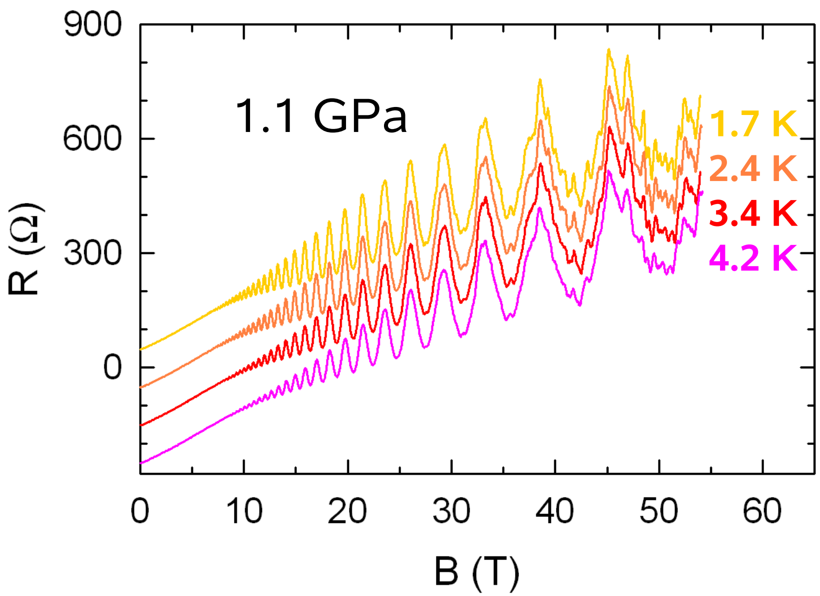
$$R = R_0 + AT^2$$

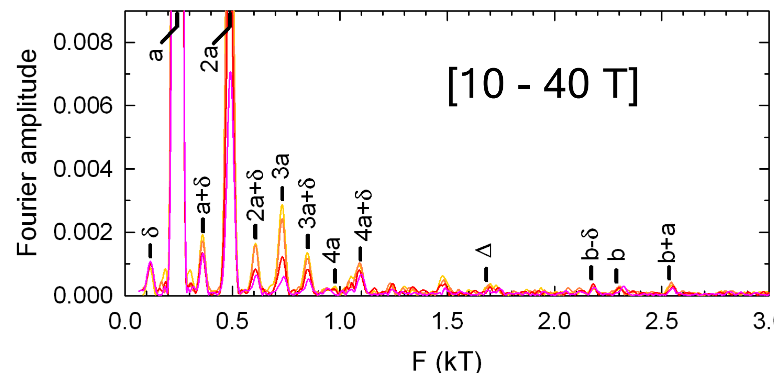
→ correlated Fermi liquid

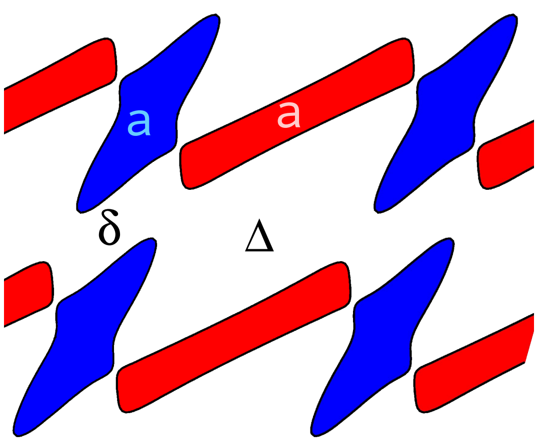
A decreases as applied pressure increases

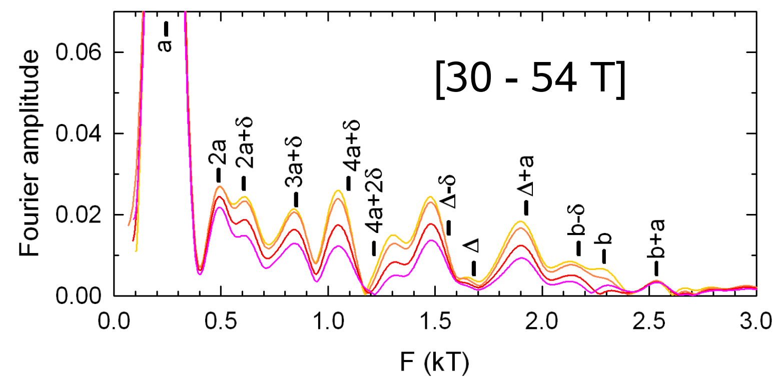
Effect of applied pressure: SdH oscillations

X = Cl (metallic ground state)





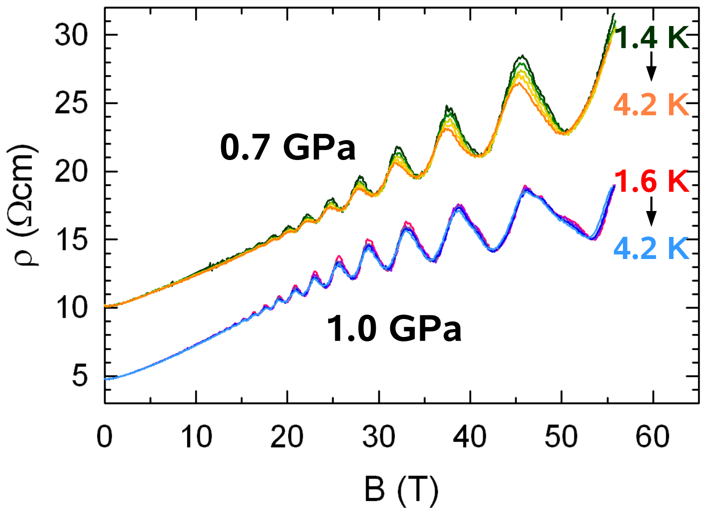


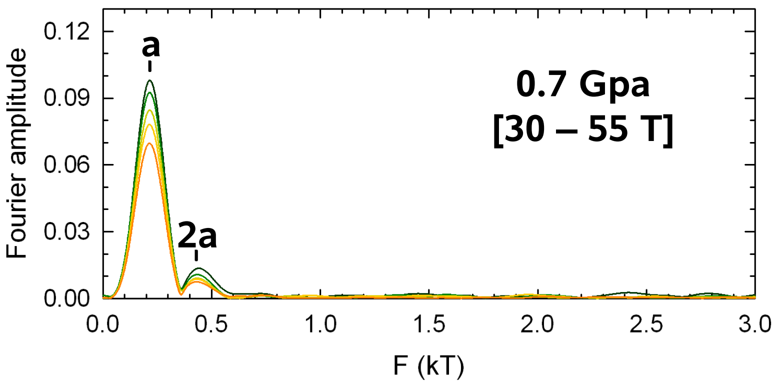


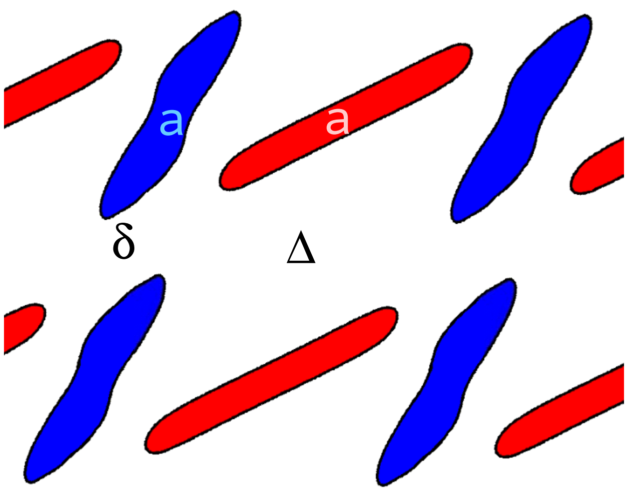
Still many frequency combinations
(magnetic breakdown)
Same features as at ambient pressure

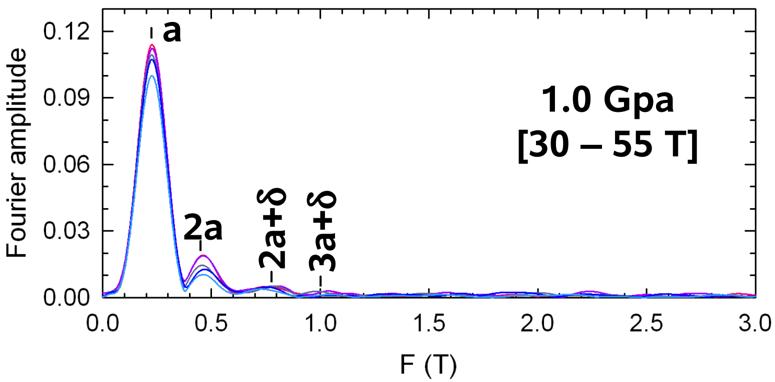
Effect of applied pressure: SdH oscillations spectra

X = Br (pressure-induced metallic ground state)



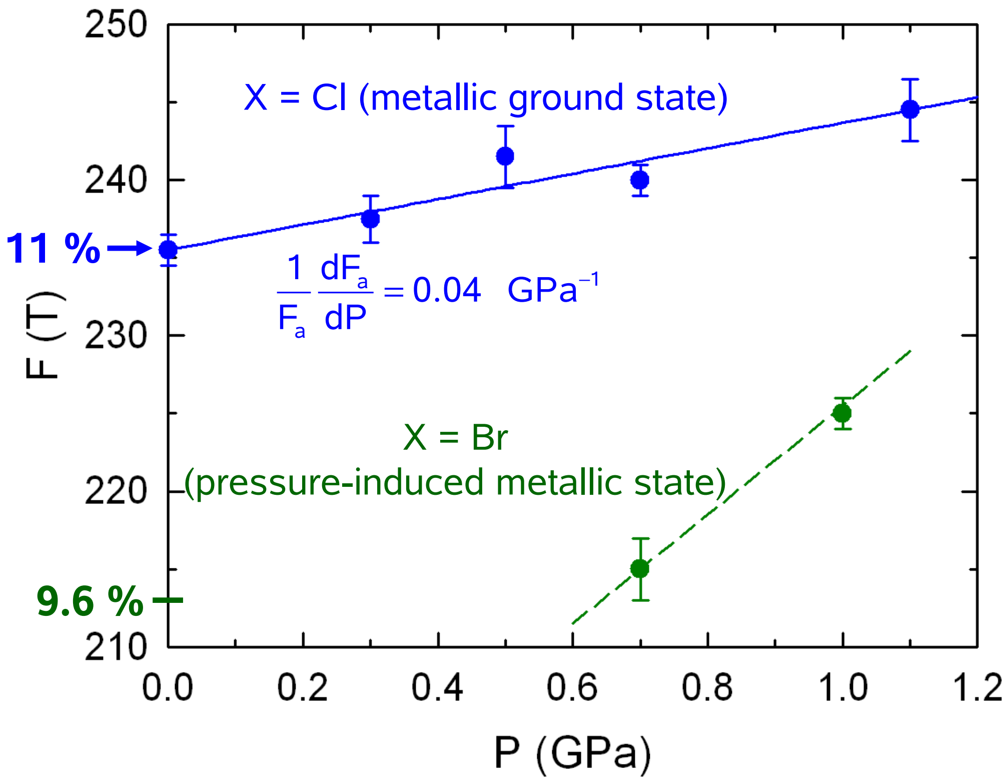


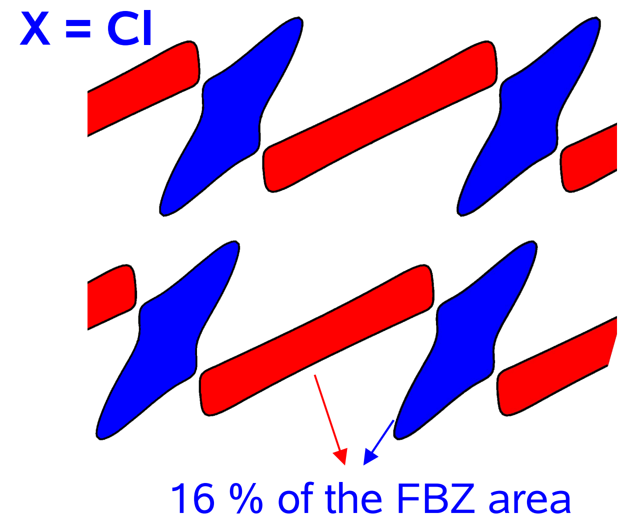


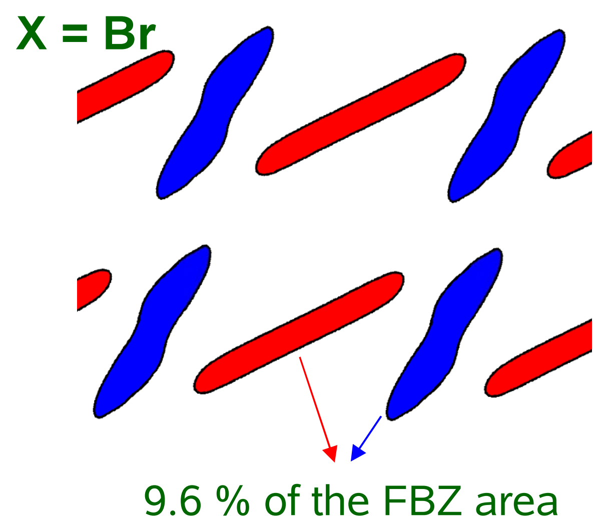


Almost no frequency combinations: suggests magnetic breakdown gap larger than for X = Cl

Effect of applied pressure:
Oscillations frequency linked to basic orbits



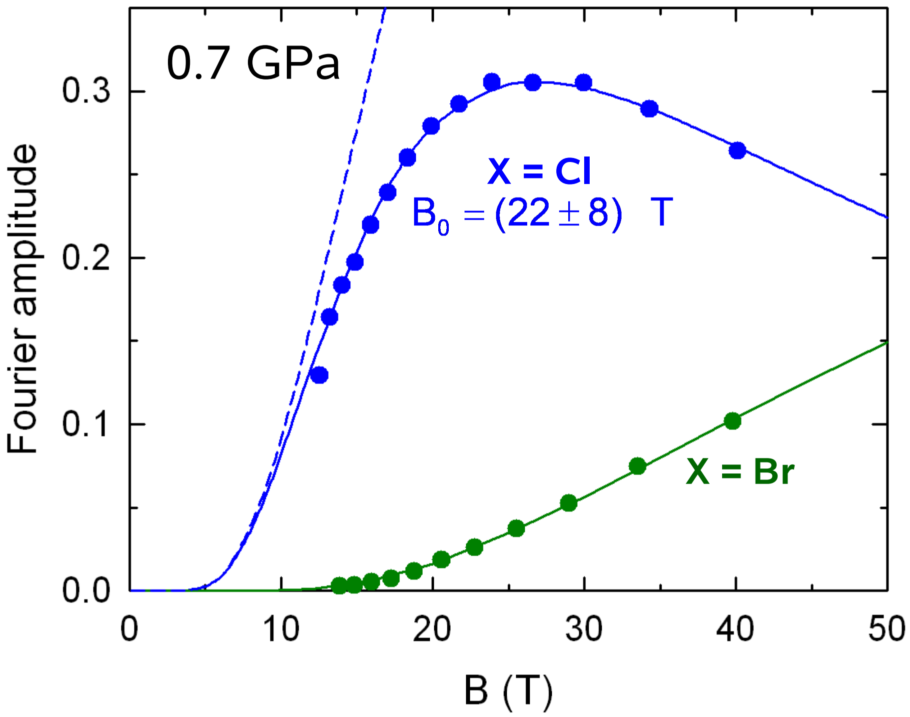


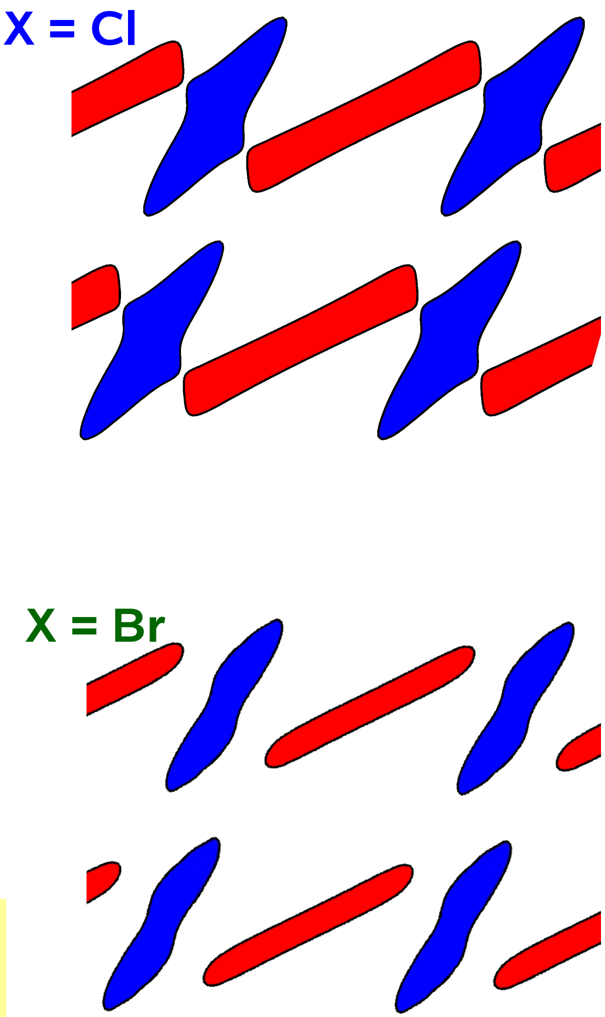


Pressure sensivity (For X = Cl) similar to most of other organic metals

Orbits area in good agreement with band structure calculations

Effect of applied pressure: Magnetic breakdown field

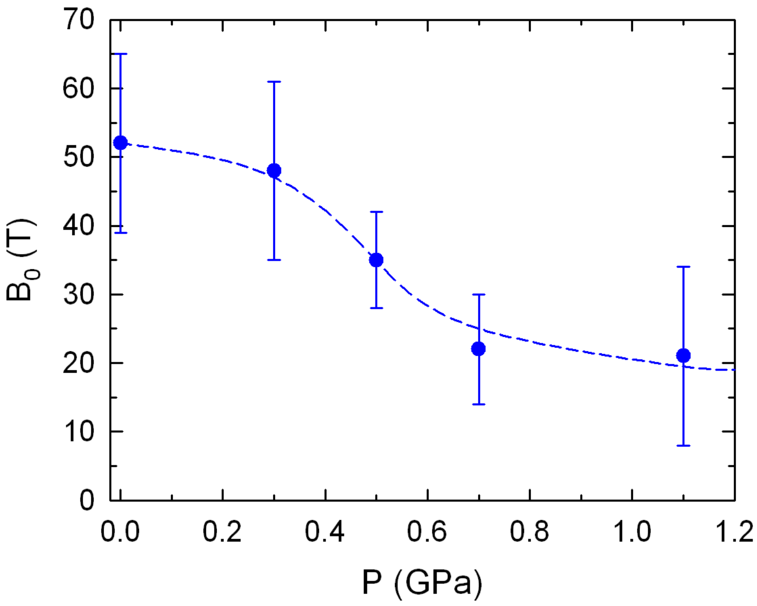




Magnetic breakdown not observed for X = Br
Accounts for the absence of frequency combinations.
Consistent with band structure calculations

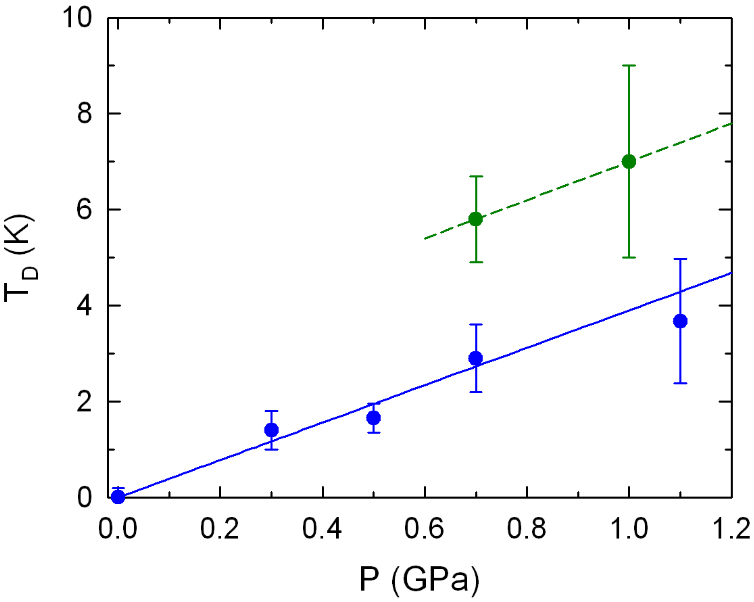
Effect of applied pressure:

Magnetic breakdown field



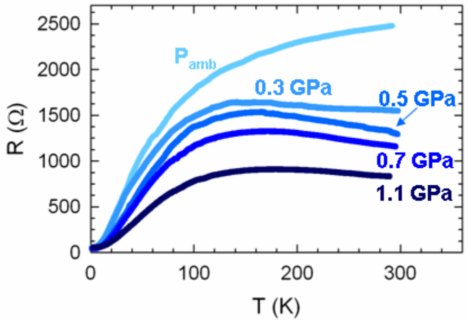
Strong decrease of the MB field

Dingle temperature (∝ scattering rate)

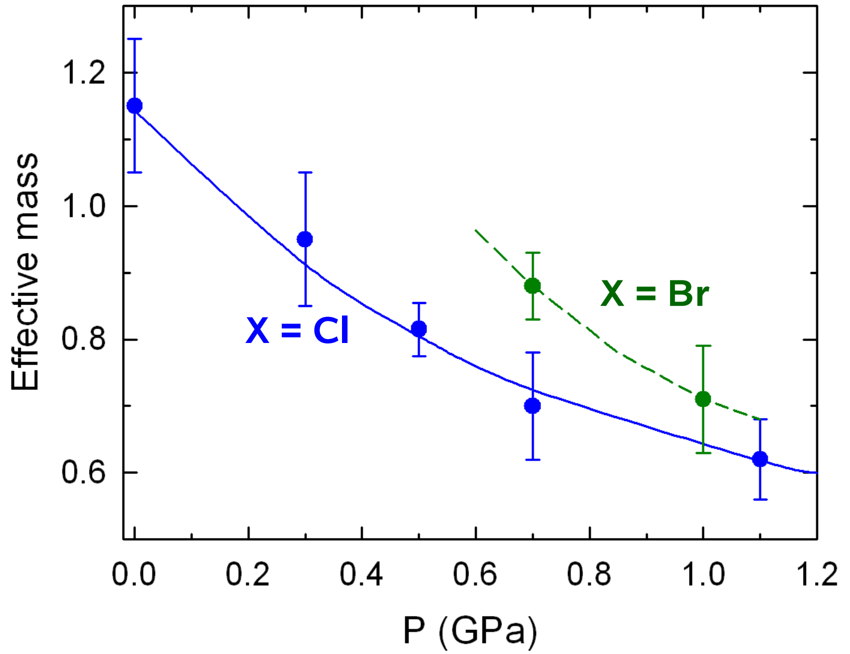


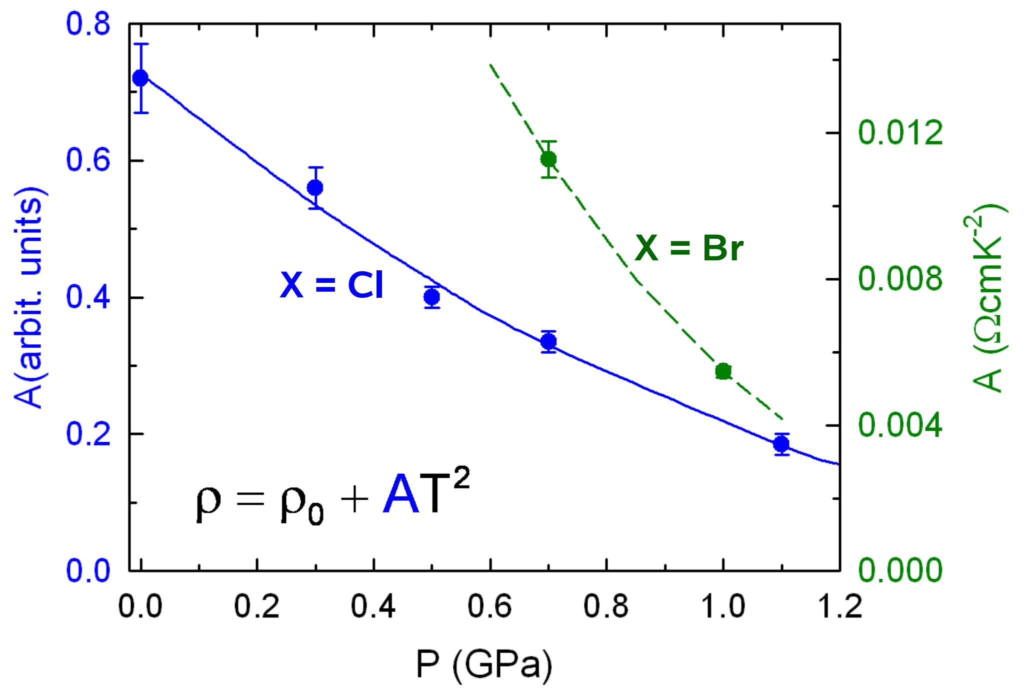
Strong (and reversible) increase of the scattering rate

may explain
zero-field resistance:



Effect of applied pressure:
Effective mass and zero-field resistance

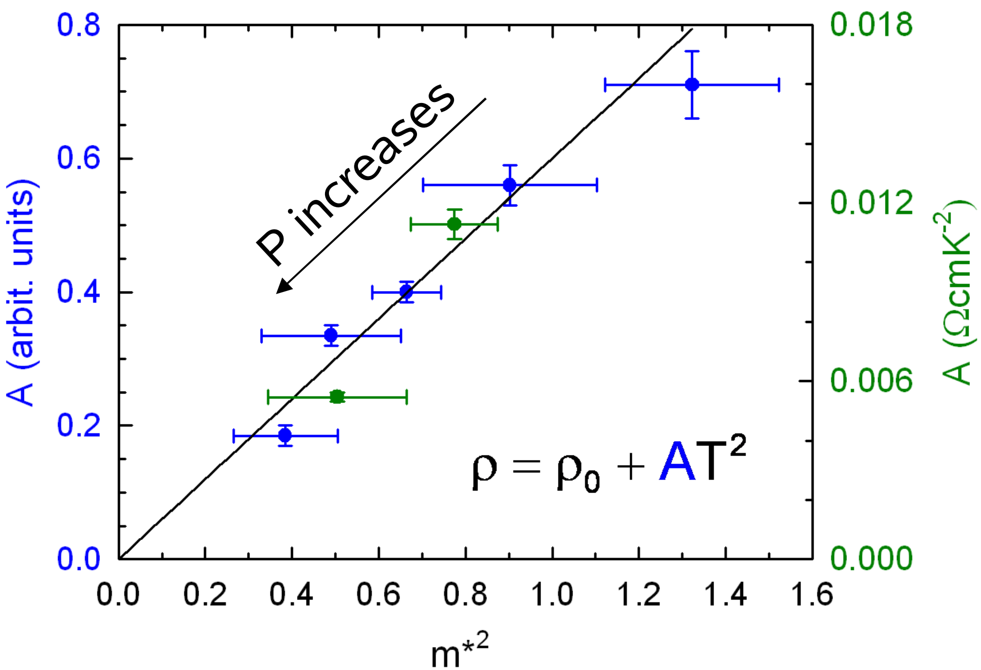




Strong decrease of both the effective mass and the coefficient (A) of the T^2 law of the zero-field resistance.

Effect of applied pressure:
Effective mass and zero-field resistance

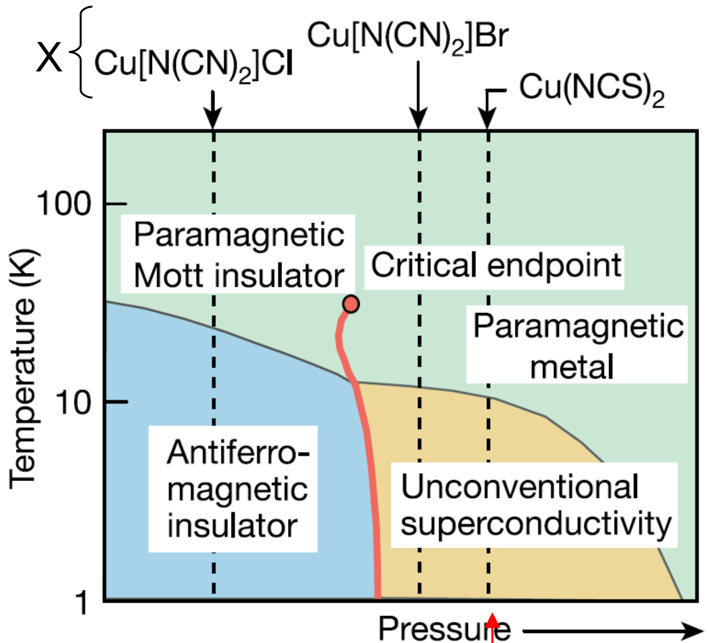
Low temperature resistivity driven by electron correlations: $A \propto m^{*2}$

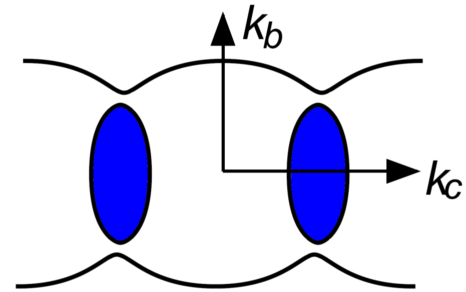


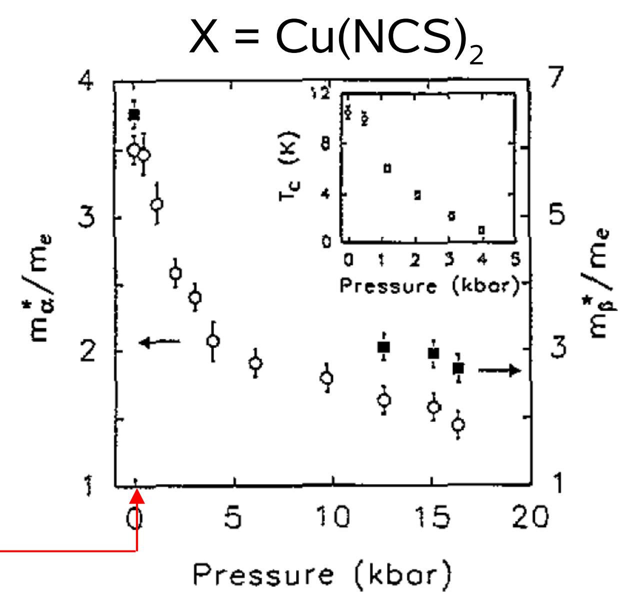
A roughly scales with m^{*2}

In the framework of the Brinkman-Rice scenario a divergence of the effective mass as approaching a Mott transition should be observed

Mott insulator state already reported for phase diagrams of organic conductors, although mainly in κ-(BEDT-TTF)$_2$X salts, with different Fermi surface







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

Caulfield et al. J.P.Cond.Mat ***6*** *2911 (1994)*

Summary

Good agreement with band structure calculations in the metallic state (Extended Hückel)
Zero-field interlayer resistivity:
- typical of strongly correlated Fermi liquid at low temperature (T^2 law)
- contribution of disorder under applied pressure at higher temperatures

Effective mass strongly decreases as pressure increases:
could suggest a Brinkman-Rice scenario (divergence of m*) for X =Br

Next steps

For X = Br:
X-ray data at low temperature (charge density wave, charge ordering?)
Magnetoresistance data in the pressure range 0.3 - 0.7 GPa (i.e. within the metal-non metal transition) to check the behaviour of the effective mass