Where are we in the physics of 1D organic conductors (superconductors)?

D.Jérome
Université Paris-Sud, Orsay, France

1D 's: The first compounds to show superconductivity in organic matter (in 1979)

- -first low dimensional materials to reveal spin density waves
- -sliding of the SDW also shown
- -magnetic field induced SDW (FISDW)
- -bulk quantized Hall effect
- -spin-Peierls
- -angular dependent magneto resistance
- -charge ordering
- -exotic superconductivity
- -non-Fermi liquids expected, spin- charge separation

Most 1D materials are based on the fulvalene molecular skeleton

- -Fabre-Bechgaard family of organic cation salts $(TMTSF/TMTTF)_2X$
- -Isostructural series
- -1D structural features --> 1D electronic properties
- -Role of pressure (hydrostatic and chemical)
- -Need to study the parent compounds

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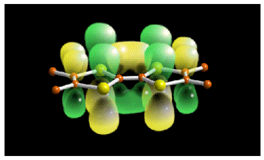
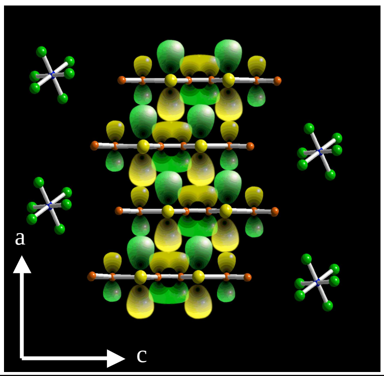
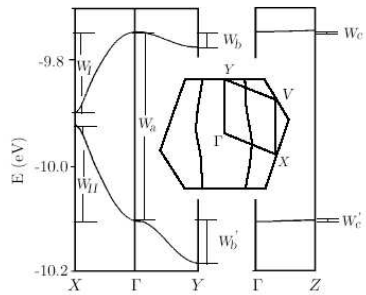
The Bechgaard molecule and salt

TMTSF



$(TMTSF)_2X$, $X=PF_6^-$,…. ClO_4^-,.





Wa ≈ 10...20 Wb ≈ 500Wc

Wa ≈ 0.4- 0.8 eV

1/2 hole per organic cation molecule

TM_2X should be 1D conductors « to a large extent »

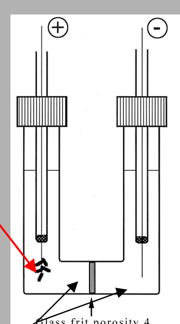
➡ « Luttinger physics » ?

Electrocrystallization

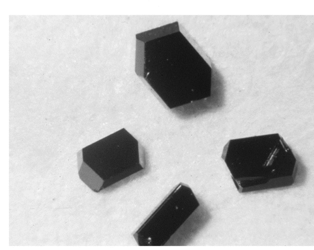
TMTSF $\underset{}{\overset{-e^-}{\rightleftarrows}}$ $TMTSF^{\bullet +}$

$2(TMTSF) + PF_6^-$ ---$\overset{-e}{}$-----> $[(TMTSF)_2PF_6]$ at anode

Non aquous solvent (pure):THF



electrolyte
$Bu_4N^+PF_6^-$

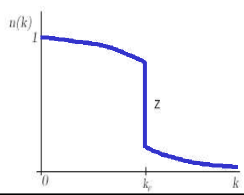


K.Bechgaard. JACS. 1981,103,2440
P.Batail, Chem Mater.1998,10, 3005-3015

Landau-Fermi theory

- Fermi statistics + fermion-fermion interactions
- Pauli principle restricts scattering near the Fermi sphere
- 'Dressed' particle and hole states near P_F with
  - lifetime $\tau \propto (\epsilon - E_F)^{-2} \to \infty$ at P_F $(T = 0\mathrm{K})$
  - correspondance $(\boldsymbol{P}, \sigma) \to (\boldsymbol{P}, \sigma)^*$

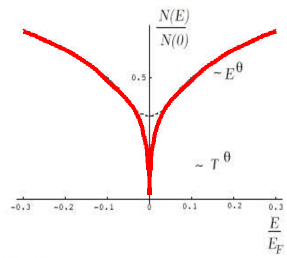
→ concept of **quasi-particles** at low energy
→ effective mass m^*
→ finite quasi-particle-weigth Z
→ collective modes: zero sound and paramagnons



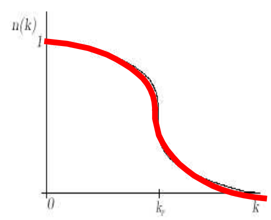
Luttinger liquid

QuasiParticles---> collective modes spin or charge

Density of states:



Fermi distribution:



$n(k) = n_{k_F} - C\,|k - k_F|^{\theta}$

Power laws

Single particle correlations

Bosonic excitations of collective modes, spin or charge

$$G(x,\tau) = -\langle T_\tau \psi(x,\tau)\psi^\dagger(0,0)\rangle$$

$$G(x) \sim \frac{e^{i \pm k_F x}}{\alpha^{-\theta}} \frac{1}{x^{1+\theta}}$$

$$\theta = \frac{1}{4}\left(K_\rho + \frac{1}{K_\rho} - 2\right)$$

Two particle correlations, SDW, SC

$$\chi_{SDW}(x) = \langle \boldsymbol{S}(x) \cdot \boldsymbol{S}^\dagger(0)\rangle$$

$$\sim \frac{e^{i2k_F x}}{x^{1+K_\rho}}$$

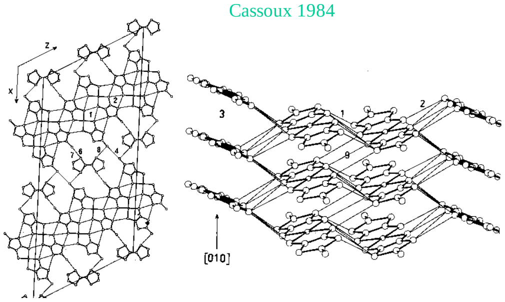
$$\chi_{SDW}(2k_F, T) \sim \left(\frac{T}{E_F}\right)^{-\gamma}$$

$$\gamma = 1 - K_\rho$$

No long range order at T=0 K

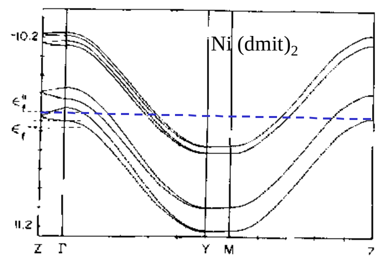
$TTF[Ni(dmit)_2]_2$ transport and band structure: A Luttinger liquid?

Cassoux 1984



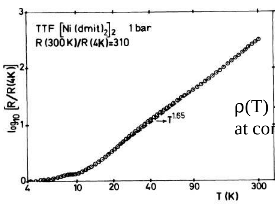
HOMO LUMO separation of acceptor < dispersion
----> A very 1D electronic compound
----> Overlap of bands
----> Non commensurate band filling, $TTF^{+0.76}$

CDW on Ni dmit chains (LUMO) at 40K
All other bands stay metallic at low temperature
Transport and NMR data
---> the case for a metallic Luttinger chain



LUMO

HOMO



$\rho(T) \sim T$
at constant volume

Canadell et-al J.Physique 50, 2967, 1989

Brossard et-al PRB 42, 3935, 1990

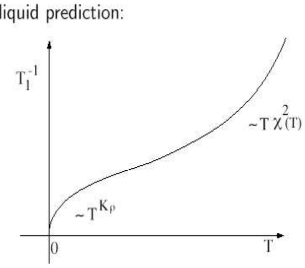
Luttinger liquid behaviour

Nuclear relaxation:

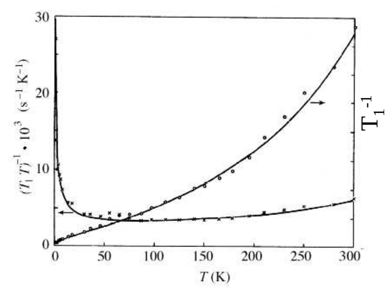
$$T_1^{-1} = C_0 T \chi^2(T) + C_1 T^{K_\rho}$$

Uniform + staggered (AF) contributions

Luttinger liquid prediction:



TTF[Ni(dmit)$_2$]: 1D metallic system (~ Luttinger liquid)



P.Wzietek et-al Eur.Phys.Lett 1998

$$(TT_1)^{-1} = C_0\chi^2(T) + C_1 T^{-\gamma}$$

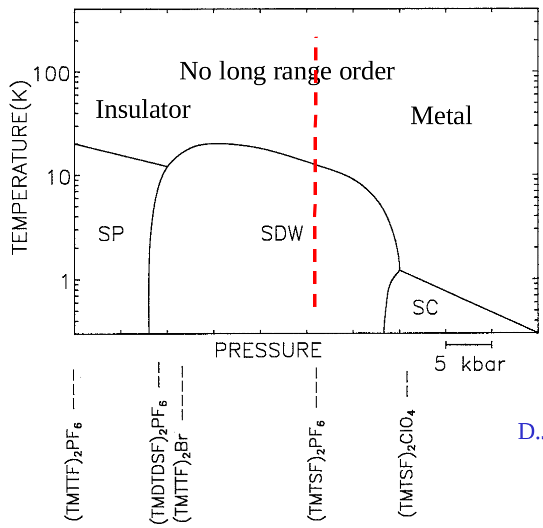
$\gamma = 1 - K_\rho = 0.6$

K_ρ => 0.4

TTF[Ni(dmit)$_2$]: 1D metallic system (~ Luttinger liquid)

Fabre and Bechgaard Salts with their Various Cross-Overs

The TM2X series



Isostructural compounds

The first compound in which organic superconductivity was discovered,1979

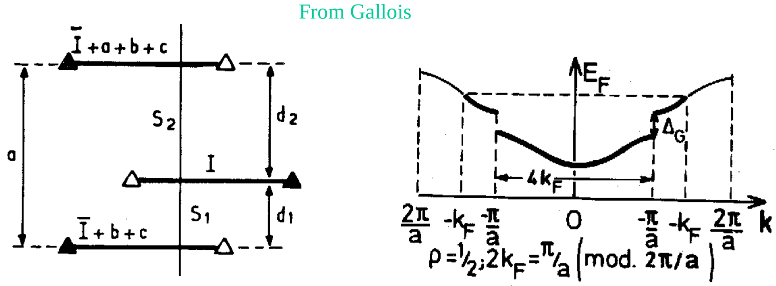
D.J Science 252, 1509, 1991

Combine chemistry and high pressure studies

Dimerization : is it the localizing agent?.

All compounds in the FB series have a centre of symmetry between the molecules
Stack dimerization is allowed
1/4 filled ---> 1/2 filled

From Gallois



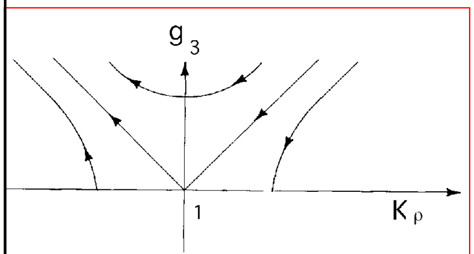
	TMTTF			TMTSF		
	PF6	ClO4	Br	PF6	ClO4	NO3
$\Delta=d_1-d_2$	0.1.	0.04	0.03	0.03	0.01	0.01

Smaller dimerization =stronger metallicity --> Umklapp scattering, Emery et-al 82

Commensurate conductor, half-filled band

Renormalization flow

g_3= Umklapp interaction => Mott localisation



$$K_\rho < K_{\rho\,critic} \Rightarrow \textit{Mott insulator}$$

Commensurability 2 $K_{\rho\,critic} = 1$

Weak repulsive interactions ----> Insulator

Quarter-filled 1D band

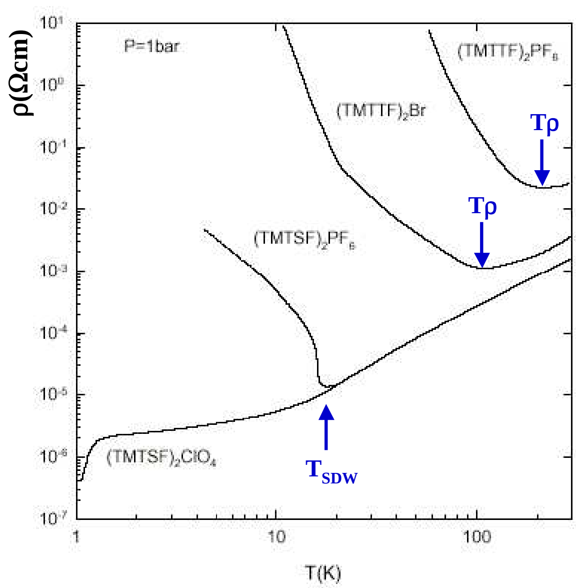
Commensurability 4 ⇒ $\boxed{K_{\rho\,critic} = 0.25}$

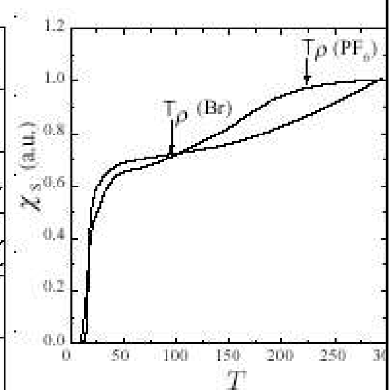
More repulsive interactions needed

(case of TM_2X) T.Giamarchi and H.J.Schulz

Spin-charge separation in Fabre-Bechgaard salts

Transport and susceptibility in TM_2X

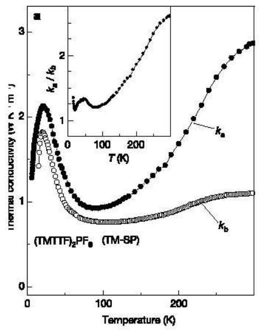


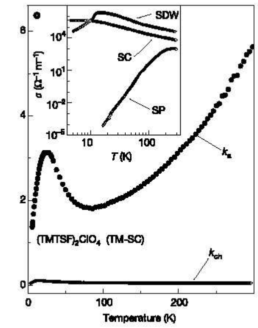


ρ degree of freedom decoupled from σ

Thermal Conductivity and spin-charge separation

Lorenz et-al, Nature 418, 614, 2002

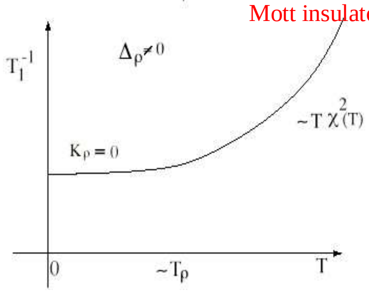




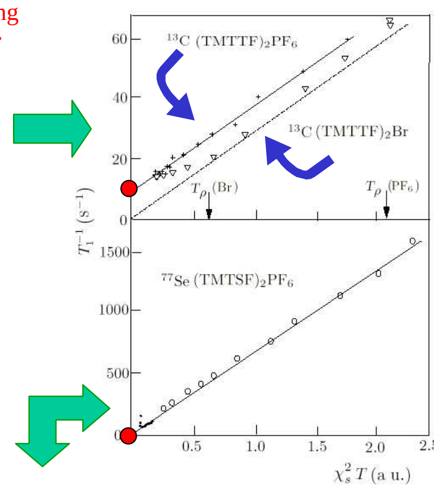
Conductivity from carriers via Wiedman-Franz law!!
Heat is carried by spin fluctuations
Spin-charge separation observed

Spin sector

In presence of a charge gap, $K_\rho = 0$: Strong coupling Mott insulator



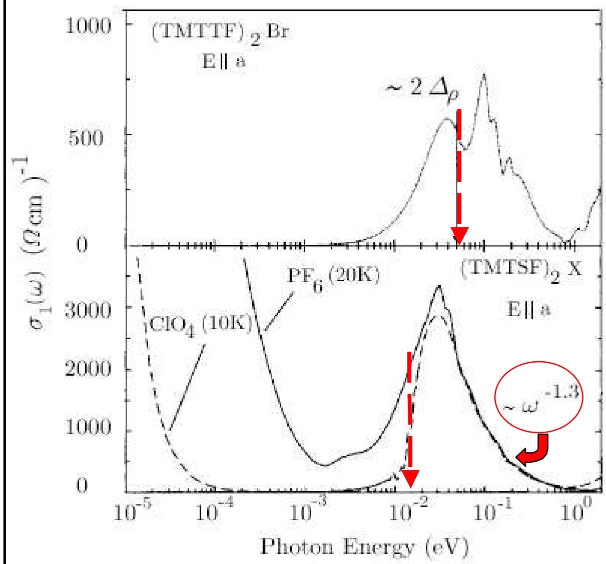
$$T^{-1} = C_0 T \chi_s^2(T) + C_1 \quad (K_\rho = 0)$$



$$T^{-1} = C_0 T \chi_s^2(T) + C_1 T^{K_\rho} \quad (K_\rho > 0)$$

No charge gap

Mott-Hubbard gap, $2\Delta_\rho$ and 1D features



S-compounds
Charge gap: 500-1000K

Se-compounds
Charge gap: 100-200K

$\sigma(\omega)\ \alpha\ \omega^{16K_\rho - 5}$

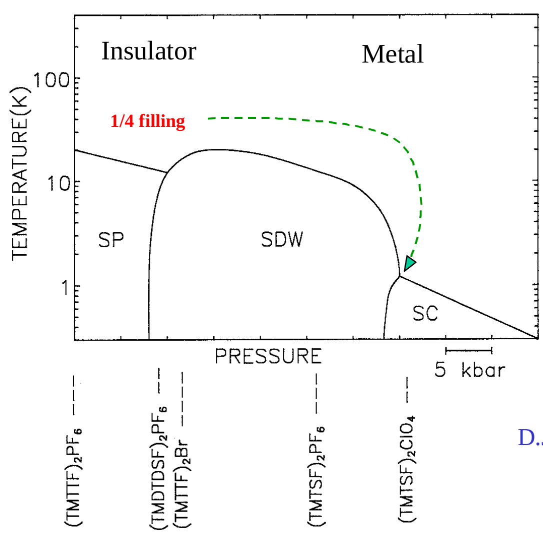
T.Giamarchi, PhysicaB,230,975,1997

in Se-compounds
1/4 filled Umklapp

$\rightarrow$ $K_\rho \approx 0.23$

Vescoli et-al, Science 1998

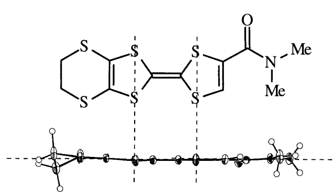
The TM2X series



D.J Science 252, 1509, 1991

Can we make a genuine quarter-filled 1D conductor with a strong localization?

Non-dimerized, strongly correlated salt: Quarter-filled band case



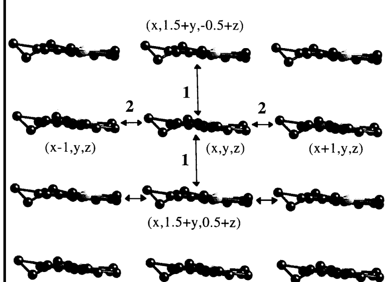
Molécule de EDT-TTF-CONHMe$_2$ vue de profil

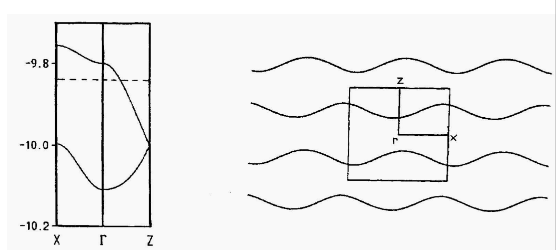
$+AsF_6$

K.Heuzé, P.Batail

-->Additional glide plane

NO dimerization





W = 0.3 eV, U/W>1

Large interchain coupling!

But strong renormalization of t_{perp}

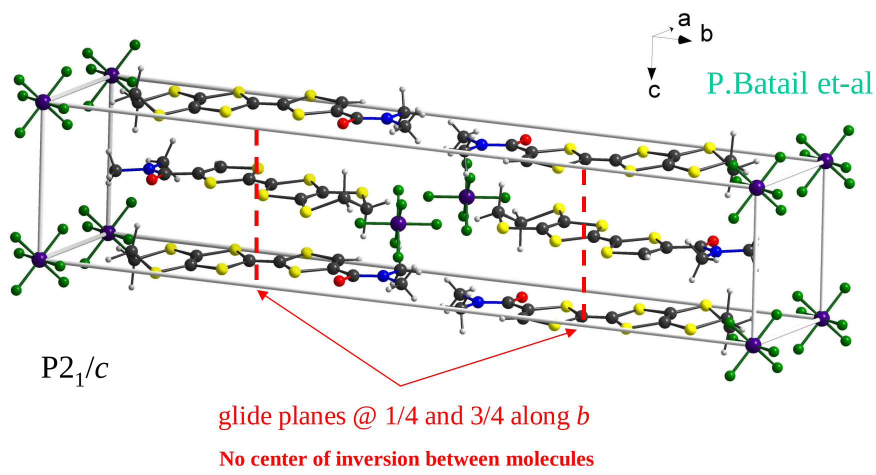


Figure 1. The monoclinic unit cell of $[EDT\text{-}TTF\text{-}CONMe_2]_2AsF_6$. The anions are at centers of symmetry.

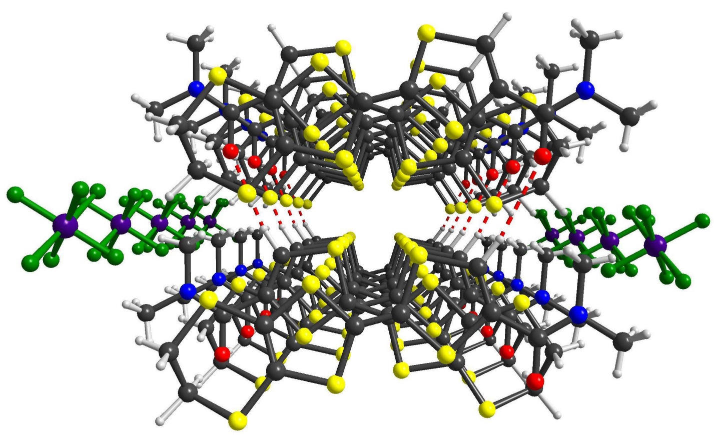
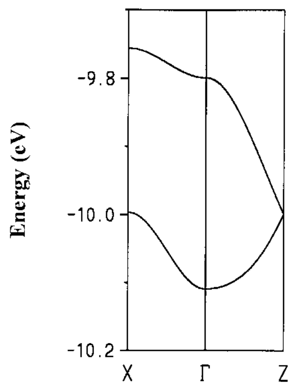
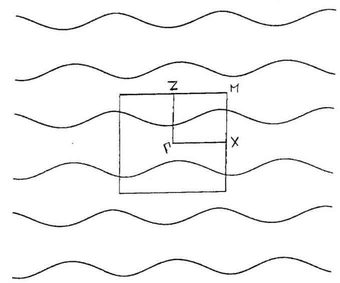


Figure 3. A perspective down the criss-cross stacks in $[EDT\text{-}TTF\text{-}CONMe_2]_2AsF_6$

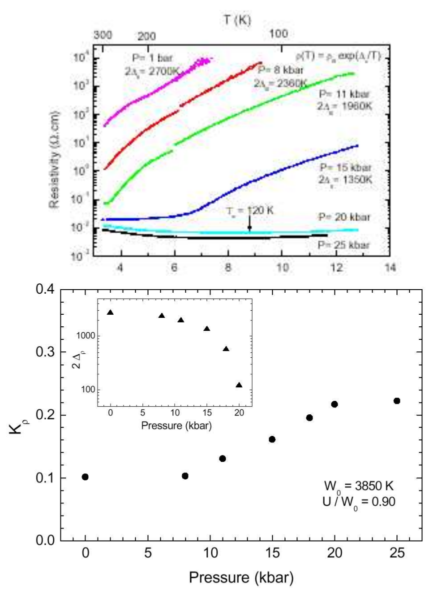
EDT Fermi surface





E.Canadell,Barcelona

Mott gap in EDT



Very large gap

$$2\Delta_\rho \approx W\left(\frac{g_{1/2n}}{W}\right)^{1/(2-2n^2K_\rho)}$$

$$g_{1/2} \approx U\frac{D}{W} \text{ and } g_{1/4} \approx U\left(\frac{U}{W}\right)^3.$$

$g_{1/2} < g_{1/4}$

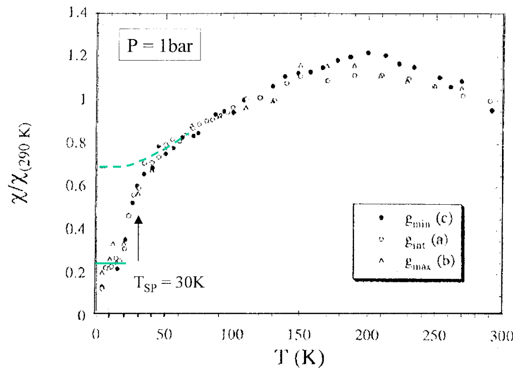
The case of 1/4 band filling

$$2\Delta_\rho = W\left(\frac{U}{W}\right)^{2/(1-4K_\rho)}$$

δlnW/δp=0.7%/kbar from optics under pressure in TTF-TCNQ, Welber et-al PRB, 1978

(EDT-TTF-CONMe$_2$)AsF$_6$

EPR susceptibility
spin-Peierls ground state
$\chi(0) \neq 0$ (10% χ(290K))



from R. Clérac and C. Coulon

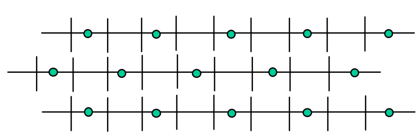
All intermolecular bounds are identical
Localized regime =optimization of coulombic forces

Walls between zig and zag configurations
--> free spins not involved in the formation of a Spin-Peierls singlet ----> free Heisenberg chains

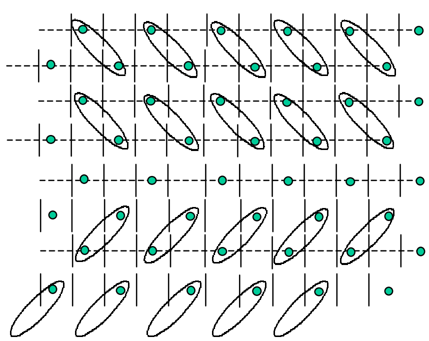
Density of free chains = 35%

Analoguous to the free radicals in undoped Cis-polyacetylene---> spin solitons

Special features of the spin-singlet ground state in EDT



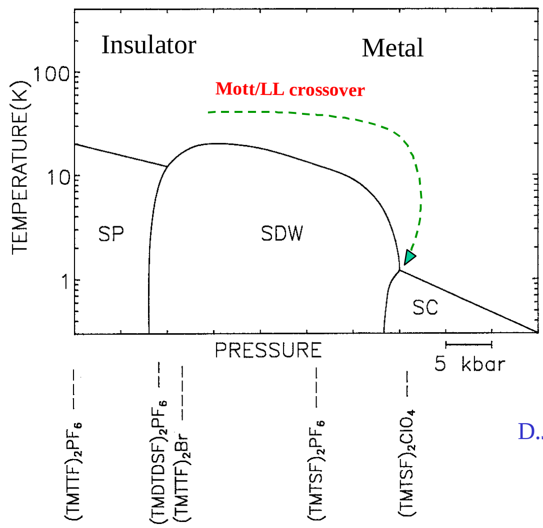
4k$_F$ Mott localization
with optimized Coulomb repulsion
at high temperature



Strong rigidity along the chains
Formation of transverse spin singlet +
lonely Heisenberg chains (domains)

Also the possibility of a singlet ground state
with RVB

The TM2X series

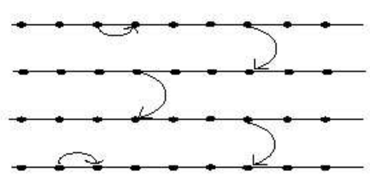


D.J Science 252, 1509, 1991

Chain-DMFT ...

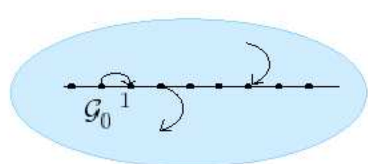
S.Biermann et-al, PRL,87, 276405-1,2001

... maps system of coupled chains



Coupled chains of interacting electrons

onto an effective chain problem

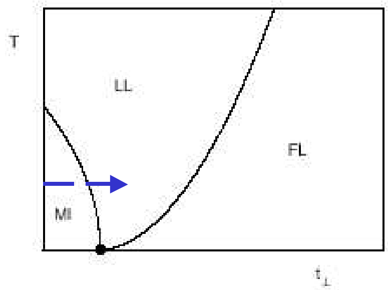


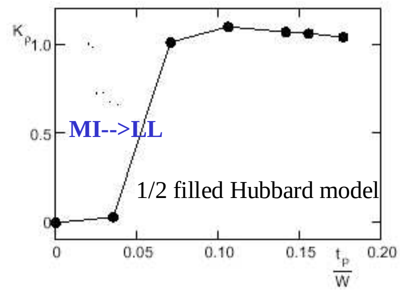
1D electrons hop to the self-consistent bath

with a self-consistency condition

The mean field is time dependent

Cross-overs:deconfinement and dimensional





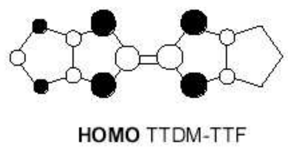
Deconfinement: 1D Mott--->LL

From S.Biermann et-al, PRL and cond-mat/0201542

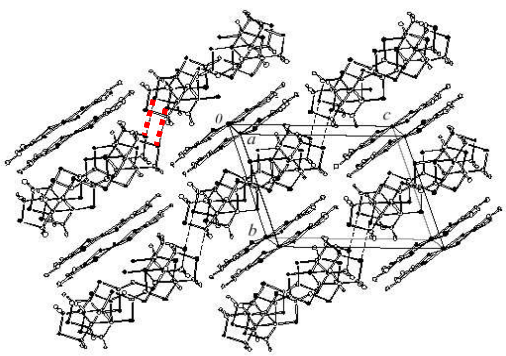
TTDM = a system in which the transverse coupling is minimized---> A 1D compound

$(TTDM\text{-}TTF)_2Au(mnt)_2$

E.B.Lopes et-al
EPJ-B, 2002

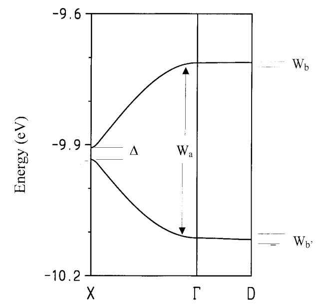


purely 1D compound from the structure



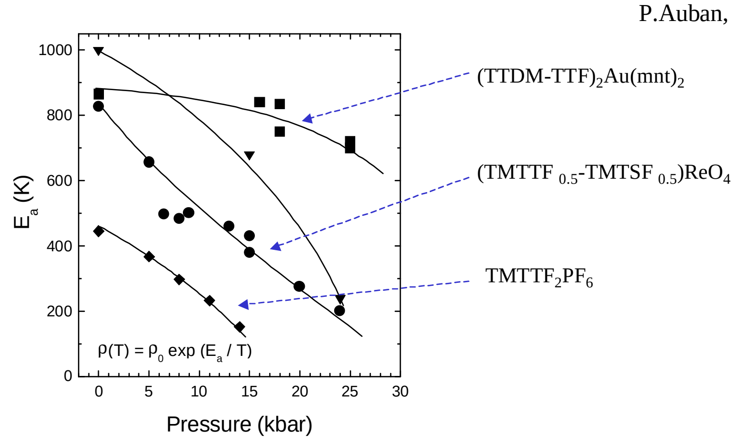
- **-Non-symetrical molecule**
- **-No interchain coupling between S-Sπ**
- **-Orthogonal $Au(mnt)_2$ anions**

- **-Strong intra chain dimerisation**
  On-site potential--> 1/2 filled



TTDM-TTF: a pressure resistant 1D compound

P.Auban, EPJ-B,2002

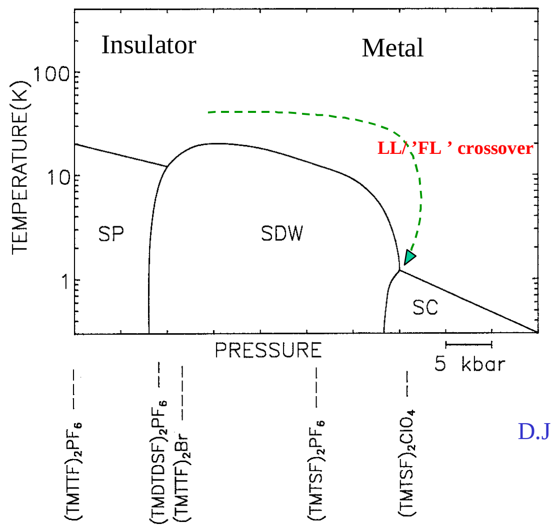


TTDM-TTF:

-A Mott insulator, gap= 1750K
1/2 filled **negligible pressure dependence in 25 kbar**

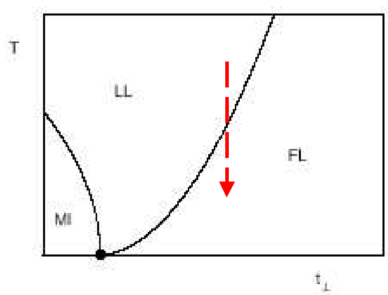
-Situation: **Mott gap >> interchain coupling, and stays under pressure**

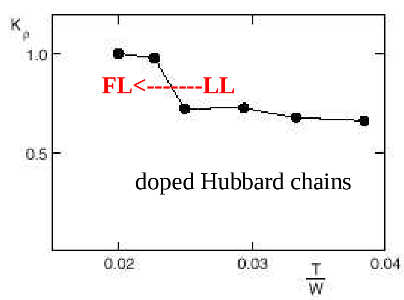
The TM2X series



D.J Science 252, 1509, 1991

Cross-overs: 1D/2D



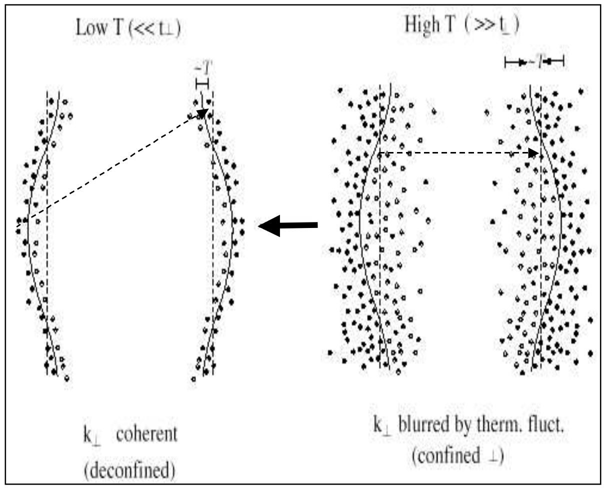


Dimensional cross-over: LL-->FL

From S.Biermann et-al, PRL and cond-mat/0201542

Transverse coupling and crossover

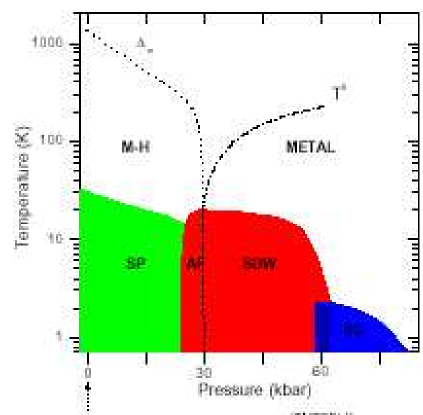
2D



1D

----> At high temperature: 1D physics is expected

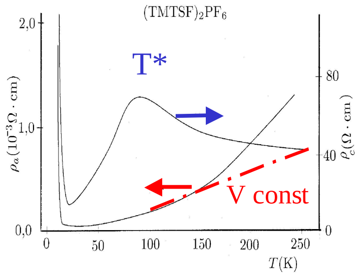
The extended phase diagram



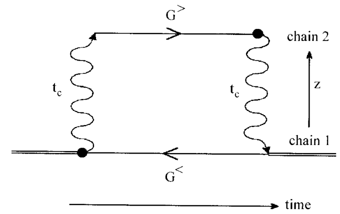
Two regimes in the FB phase diagram:
--The 1D regime $2\Delta_p > t_\perp$ Δ decreases –4%/kbar
--Faster decrease -> interplay between localization and transverse coupling :the cross over regime

Better knowledge of the interplay between Mott localization and crossover is needed
Use $TMTTF_2PF_6$ to minimize the compressibility effect. No need for conversion at constant volume

Longitudinal and transverse transport in $TMTSF_2PF_6$



holon-->electron-->holon



Transport along c is incoherent (optical data)

Conductivity probes the density of quasiparticles in the a-b plane

<u>High temperature regime</u>

T>T* (120K)

$\rho_a \propto T^{4n^2K\rho-3}$ ⟹ $K_\rho \approx 0.23$ ⟸ $\rho_c(T) \approx T^{1-2\alpha}$

Moser et-al
EPJ-B 1, 39, 1998

<u>Low T regime</u>

ρ_a **« Fermi like»--> T²**

ρ_c **incoherent**

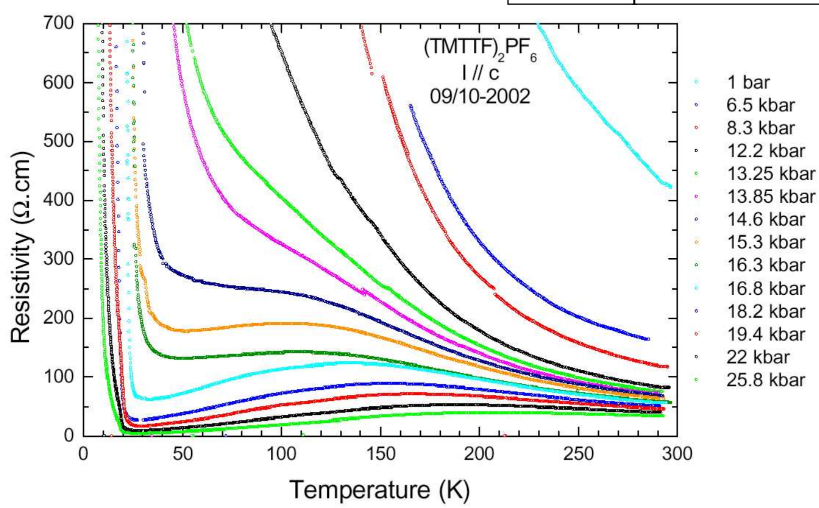
$1/\tau_c \sim t_c^2 \tau$ —> $\rho_c(T) = (\rho_a \rho_b)^{1/2}$

$$\sigma_c(\omega) = \frac{4e^2ct_c^2}{\hbar}\int dx' \int \frac{d\varepsilon}{2\pi}\frac{f(\varepsilon)-f(\varepsilon+\hbar\omega)}{\hbar\omega} \times A_{1D}(x',\varepsilon)A_{1D}(x',\varepsilon+\hbar\omega)$$

A.Georges et-al PRB 61, 16393, 2000

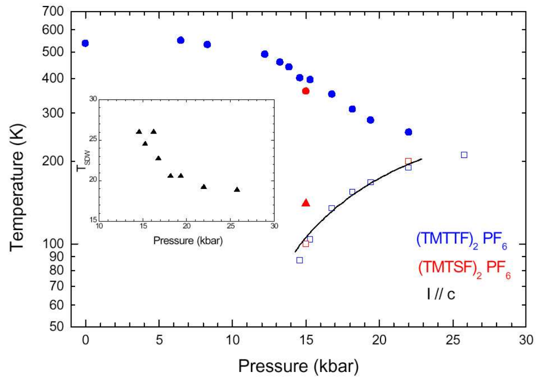
ρ_c in TMTTF2PF6

No constant volume conversion needed for this compound



P.Auban, recent data,unpublished

Mott gap and crossover from ρ_c TMTTF2PF6



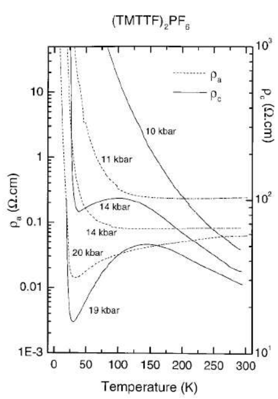
TMTSF$_2$PF$_6$
▲ Optical gap
□ Cross over

ρ_c is essentially dominated by the Mott gap at low pressure, ⟹ $\rho_c \approx \exp \Delta / kT$

At the highest pressure --> Luttinger metal, α= O.85, K_ρ= 0.20 ⟹ $\rho_c(T) \approx T^{1-2\alpha}$

ρ_a = exp Δ/kT, T<<Δ

or $T^{16K\rho-3}$,T>Δ



Moser, et-al 1998

-->Example 19-20 kbar:

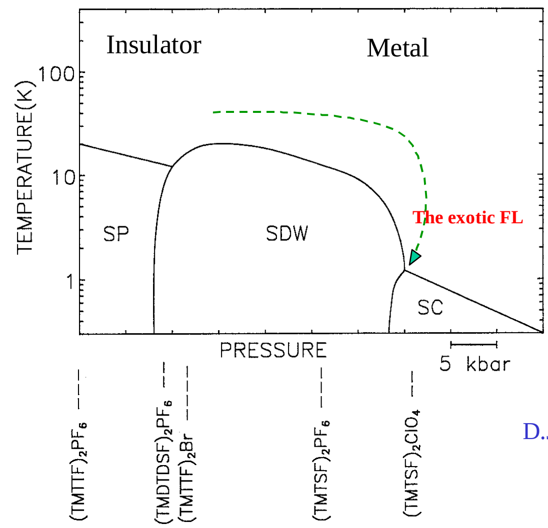
Mott activation energy=200K
ρ_c, --> cross over at 150-200K

$\rho_a = T^{16K\rho-3}$ (α T) above the Mott gap
and T^2 below cross over

-->14 kbar : crossover is below the Mott gap
ρ_a looks metallic even when the Mott gap is still fairly large
ρ_c gives a better measure of the Mott gap

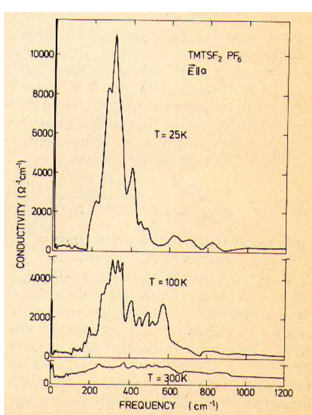
Fabre and Bechgaard Salts with their Various Cross-Overs

The TM2X series



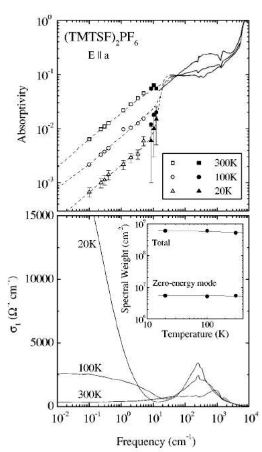
D.J Science 252, 1509, 1991

Narrow Drude peak in TMTSF2PF6



C.S. Jacobsen PRB 28, 7019, 1983

γ= 0.3 cm-1 at 25K



Schwartz et-al
PRB. 58,1261,1998

Constant spectral weight in the Drude mode

The zero frequency Drude Peak

Drude description of the conductivity

$$\sigma(\omega) = \frac{\omega_p^2 \tau}{1+\omega^2\tau^2}$$

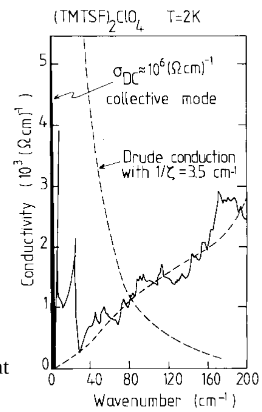
Near 12 K, $\sigma(0) = 3.10^{5}$ cm^{-1} and ω_p=9900cm^{-1}

---> $1/\tau$ = 4 cm^{-1} and

--> too much conductivity in the FIR regime

--> Large DC conductivity due to a zero frequency collective mode

T.Timusk, 1983-86

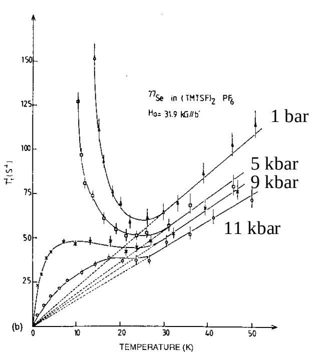


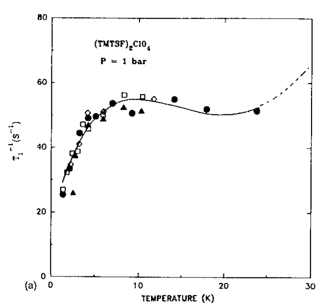
Plasma frequency Ωp=500cm^{-1} from the zero of the dielectric constant

Oscillator strenght up to ω $\int_0^{\omega} \sigma(\omega) d\omega = \frac{\pi}{2}\frac{n^{eff} e^2}{m^*} = 1\%$ *of total with* N_h

T(K)	2	10	25
γ_c cm^{-1}	0.005	0.04	0.09

The spin sector:NMR in the metallic state





T_1 anomaly attributed to the dimensional crossover T^*

In the pertubative RG approach $T^* = t_\perp (t_\perp / t)^{\alpha/(1-\alpha)}$

provides T^*~2-3K with K_ρ=0.23, $\alpha \sim 0.6$

--> T^* much too low compared to the crossover observed by transport or optics

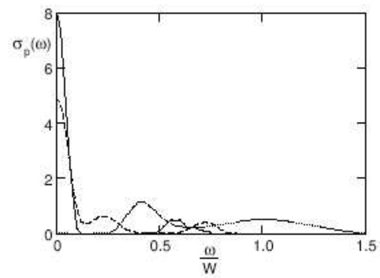
Absence of SDW ground state

Cannot be attributed to precursor effects?

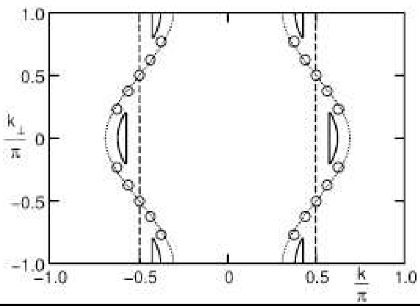
The low temperature Fermi liquid

1/2 filled Hubbard model

S.Biermann et-al



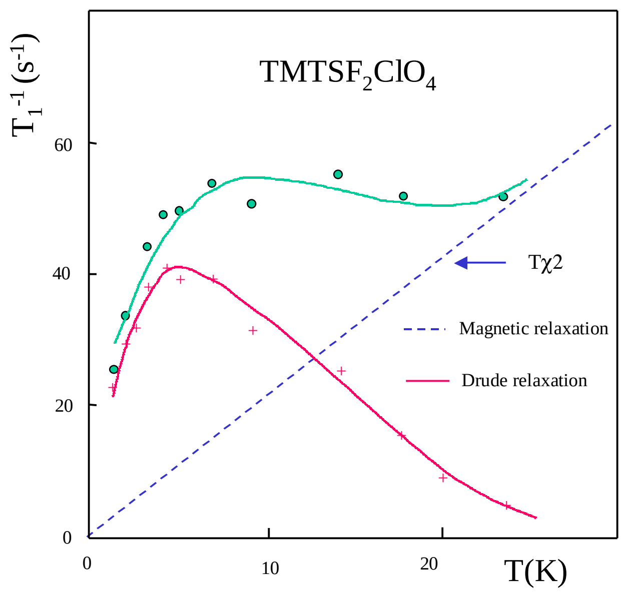
Inter-chain optical conductivity:
Mott gap +Drude peak, with tinyl weight



--Fermi surface analogous
to the non-interacting coupled chains
Explains the angular magnetoresistance giving
the bare coupling values
(Danner-Chaikin oscillations)

--Weak modulation of the QP weight on the FS

Magnetic relaxation and Drude relaxation

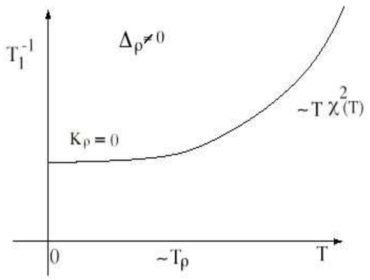


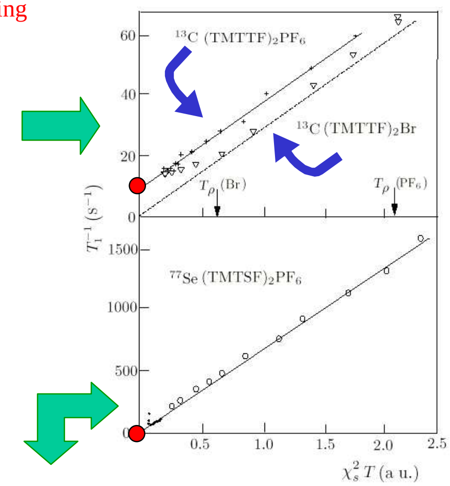
H//c or b
3-6Teslas

data Creuzet,
Wzietek, 1986

Spin sector

In presence of a charge gap, $K_\rho = 0$: Strong coupling





$$T^{-1} = C_0 T\chi_s^2(T) + C_1 \quad (K_\rho = 0)$$

$$T^{-1} = C_0 T\chi_s^2(T) + C_1 T^{K_\rho} \quad (K_\rho > 0)$$

No charge gap

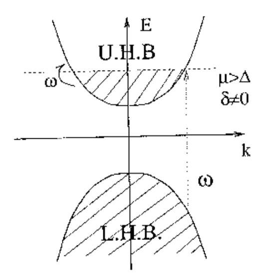
Nuclear relaxation close to commensurability

Cross over due to a Mott transition
with *in situ* doping T.Giamarchi

Existence of solitons close to commensurability
---> narrow Drude peak
with small spectral weight (doping/Δ)

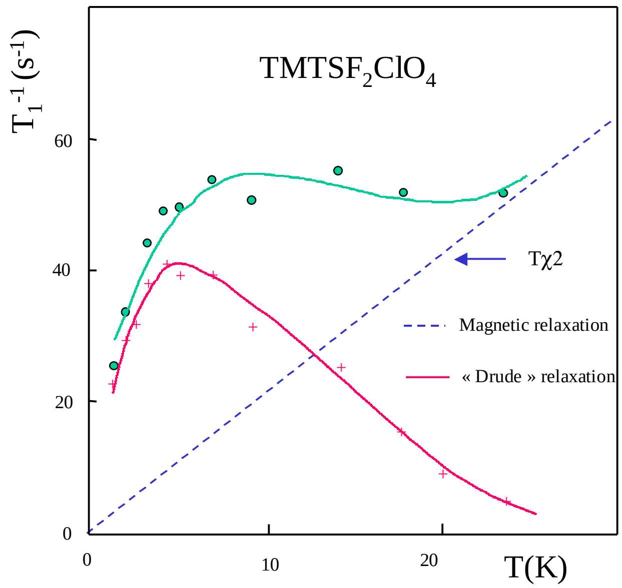
---Their motion is relaxing for the nuclear spins
via hyperfine coupling modulation
$1/T_1$--->τ_c A^2 x (Boltzmann factor)--> τ_c $T^{1/2}$

---Like relaxation in semiconductors
The effective number of carriers is increasing
at low temperature,
$1/T_1$--->N^{eff} $T^{1/2}$



Fabre and Bechgaard Salts with their Various Cross-Overs

Magnetic relaxation and Drude relaxation



H//c or b
3-6Teslas

Theory of the relaxation at low temperature must be revisited!!

Summary

-Fabre Bechgaard salts have commensurate band filling and 1D electronic properties------>1D Mott insulators

-1/4 filled Umklapp scattering is pertinent for the localization, U/W $\sim$ 1 probably not 1/2 filling

-Relevance of transverse coupling for the MI/LL crossover

-Repulsive Luttinger liquid properties visible at high temperature in the high T limit of the Mott insulator

-The 2D exotic Fermi liquid is stabilized by the transverse coupling and leads to a very narrow band Drude metal

Perspectives:

- Make an incommensurate conductors and check T_1 and transport vs T
- Reinterpretation of NMR at low temperature,
- Microscopic study of the superconducting state,
  role of non magnetic impurities,
  quasiparticle excitations in the SC phase, acoustic attenuation, NMR Knight shift and T_1
- High field studies of the FISDW, role of anions in TM2ClO4?

Epilogue

Bechgaard salts:

« The most interesting materials ever discovered »

Paul Chaikin

all organics are interesting !!

D.J

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