Unconventional metallic state in the 2D Hubbard-Wigner lattice

Simone Fratini

Institut Néel-CNRS, Grenoble

Instituto de Ciencia de Materiales Madrid

Jaime Merino

Universidad Autónoma de Madrid

Outline

- Theoretical problem: Mott vs. Wigner insulators.
- Experimental problem: the quarter-filled layered molecular crystals θ-ET_2X, anomalous metallic properties (low D, correlations, charge order, ...).
- Long-range Coulomb interaction on a lattice: the Hubbard-Wigner model.

  $\rightarrow$ intermediate anomalous metallic phase
- Future experiments?

SF & J. Merino, Phys. Rev. B 80, 165110 (15 oct 2009)

SF, G. Rastelli, PRB 75, 195103 (2007)
SF, B. Valenzuela and D. Baeriswyl, Synth. Met. 141, 193 (2004)
B. Valenzuela, SF, D. Baeriswyl, PRB 68, 045112 (2003)

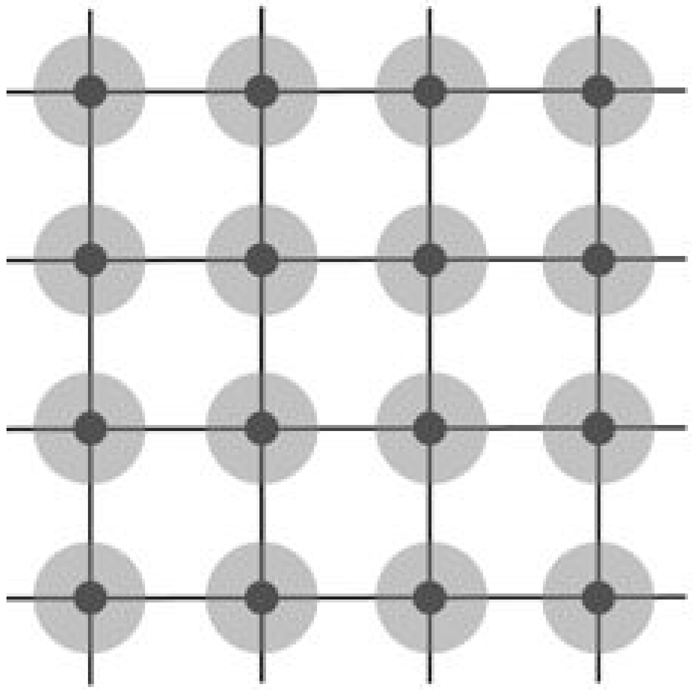
H.Seo, J. Merino, H. Yoshioka, M. Ogata, JPSJ 75, 051009 (2006)

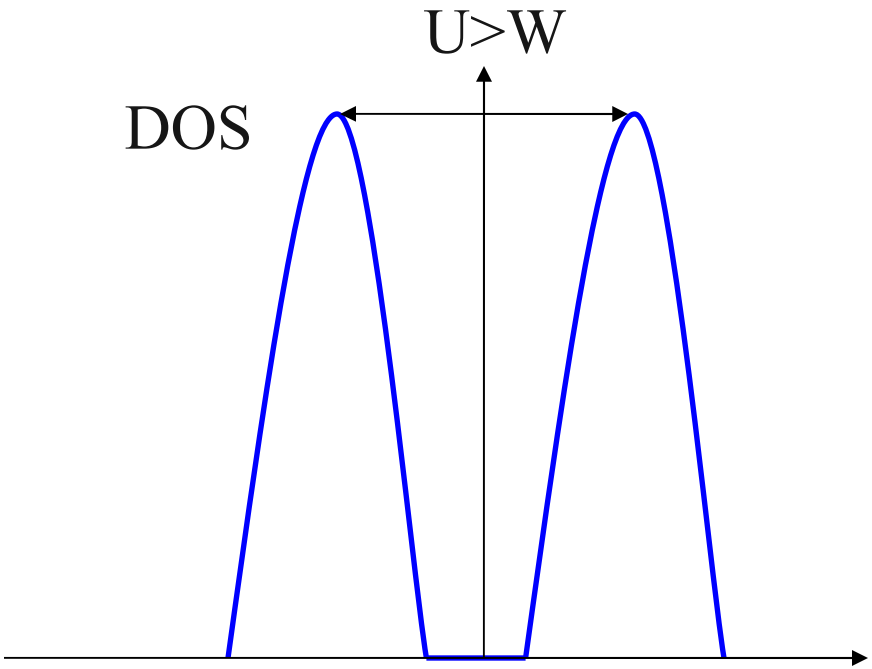
Mott vs. Wigner insulators

Kinetic energy of electrons is overcome by the Coulomb repulsion energy: cuprates, organics, fullerenes, manganites, ruthenates,…

→ insulating (localized) due to interactions contrary to predictions of band theory

Mott insulators

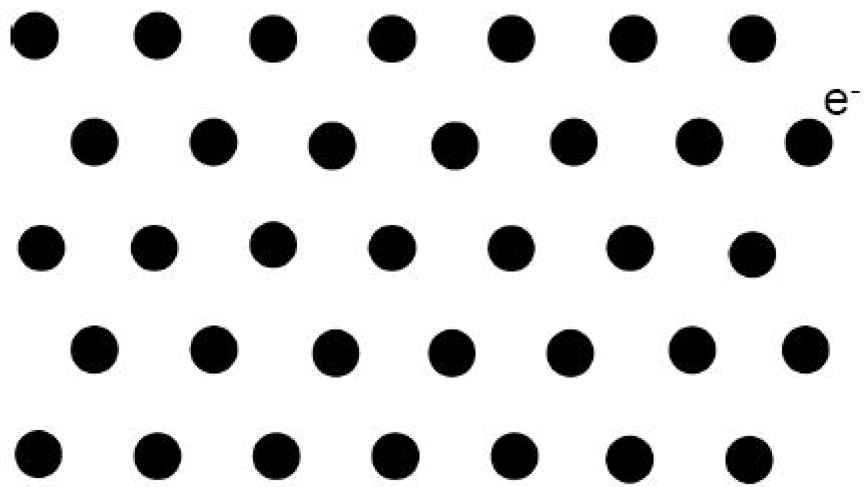




- Integer filled bands: cuprates, κ-organics, fullerenes.
- Large on-site Coulomb repulsion, U>W (bandwidth), on a lattice: d-orbitals, molecular orbitals in a crystal.

Wigner crystal in a 2D electron gas

Hexagonal Wigner crystal in 2D



$$V(r) = \frac{e^2}{r}$$

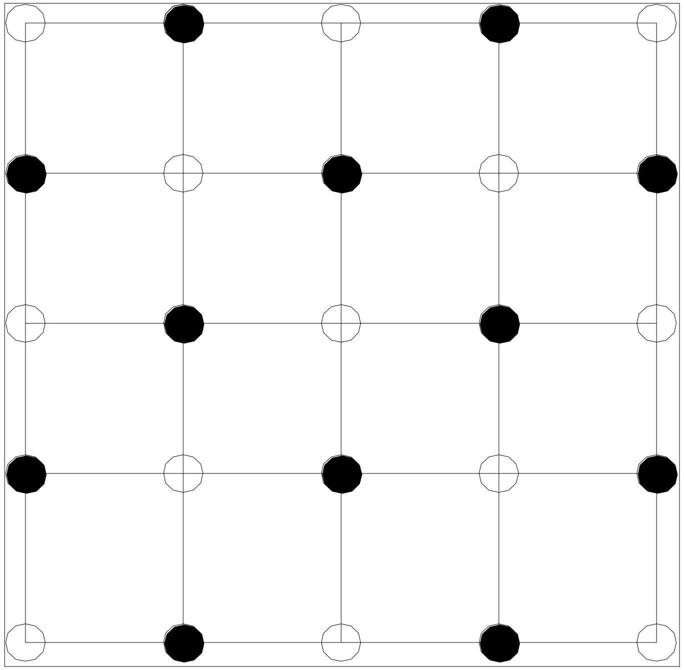
- 2DEG with no underlying crystal lattice.
- At low-density, long-range Coulomb repulsion dominates over the kinetic energy, electrons crystallize on a triangular lattice. [Wigner (1934)]

Wigner crystal on a 2D lattice

Simplest example: WC on a square lattice at 1/4-filling

Lattice commensurability localizes electrons on alternating sites instead of the usual triangular arrangement.

[Cocho, EPL, (1986), Baeriswyl, Fratini, J. Phys. IV, (2005)]

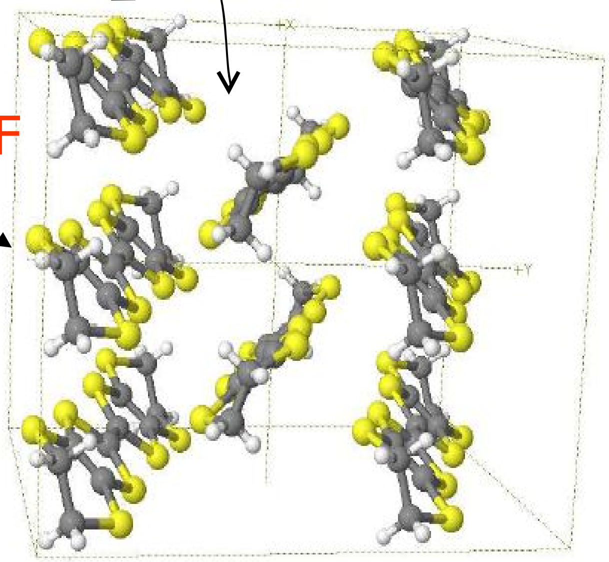


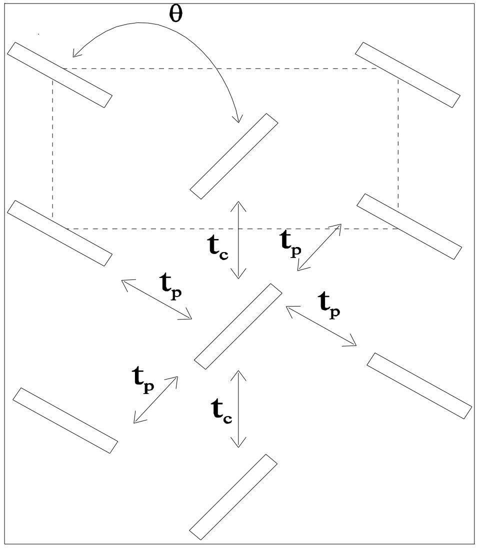
- Wigner crystal physics due to Long Range interactions
- Mott physics (electronic correlations) due to local U

Organic salts θ-(BEDT-TTF$^{0.5+}$)$_2$X$^-$

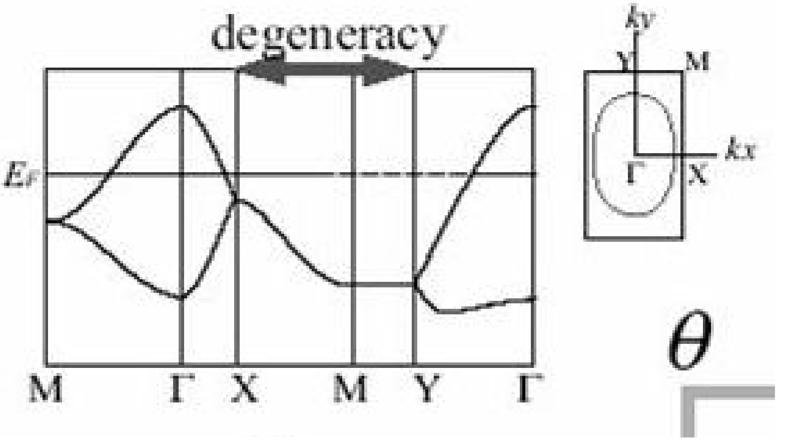
θ

0.5$^+$ per molecule BEDT-TTF

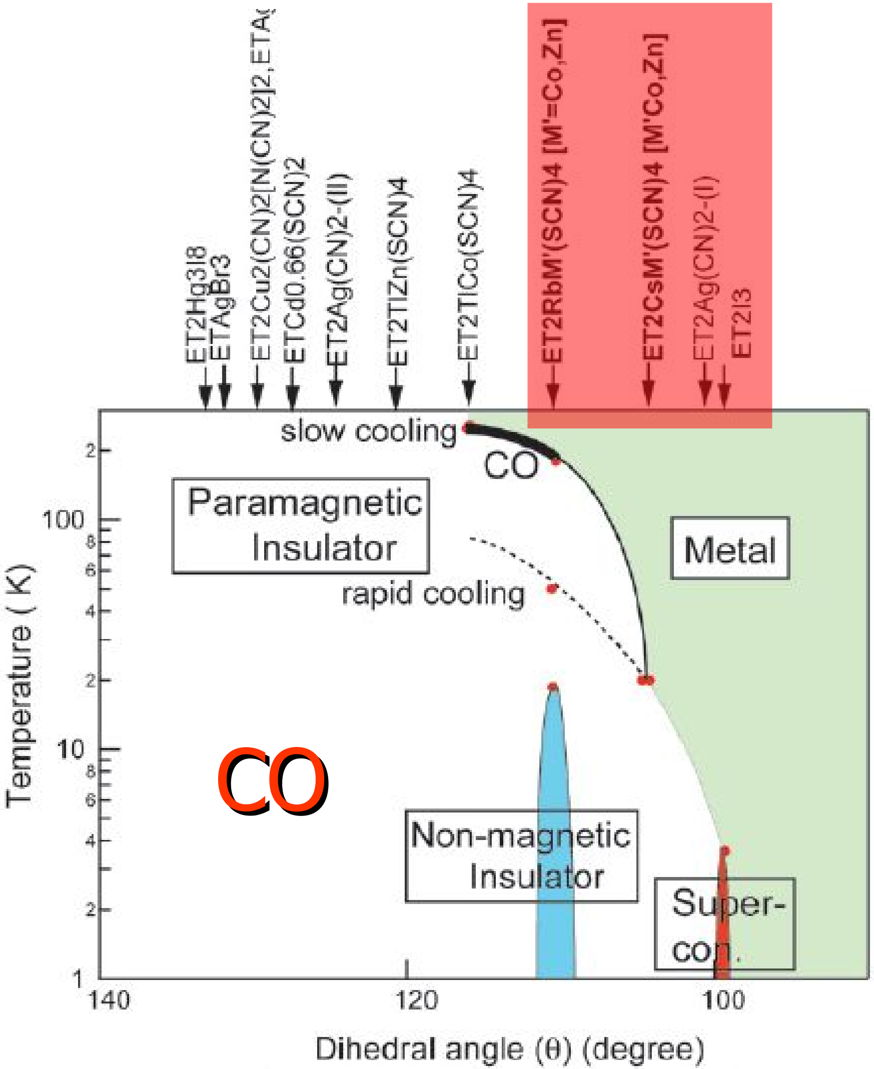




Tight-binding predicts quarter-filled HOMO bands: metallic state



Phase diagram

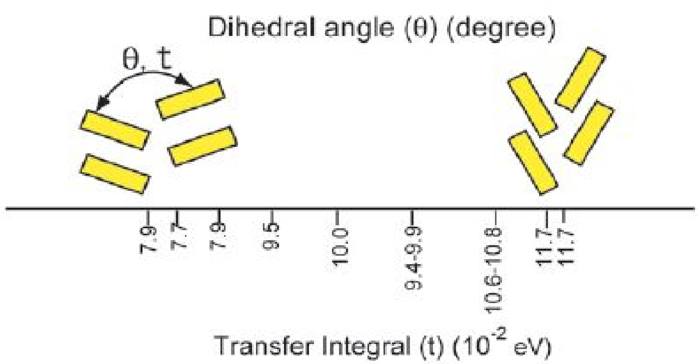


[Mori, JPSJ (2006)]

On the verge of metal/CO transition

- large U> W
- bad metal, V~W
- poorly screened interactions

→ long-ranged



Hubbard-Wigner model

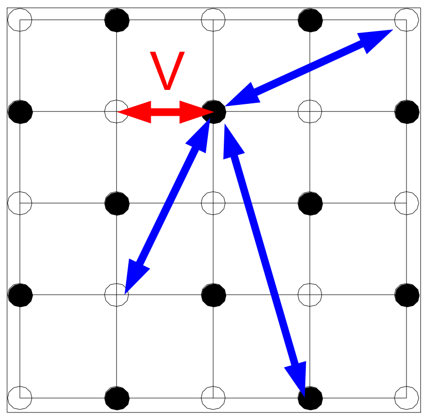
$$\hat{H} = \hat{T} + \hat{U} + \hat{V}$$

$$\hat{T} = -t \sum_{<ij>\sigma} c^{+}_{i\sigma} c_{j\sigma} + h.c.$$

$$\hat{U} = U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$ On-site repulsion

$$\hat{V} = \sum_{i \neq j} V_{ij}\, n_i n_j$$ Non-local repulsion

¼ filling, square lattice



- Extended Hubbard Model (EHM): $V_{ij} = V(n.n.)$
- Hubbard-Wigner Model (HWM): $V_{ij} = \dfrac{V}{|i-j|}$

both models give charge ordering at V>Vco, but...

V_{CO} - homogeneous metal to CO transition

Long range Coulomb interactions stabilize the metallic state ("softer" charge fluctuations).

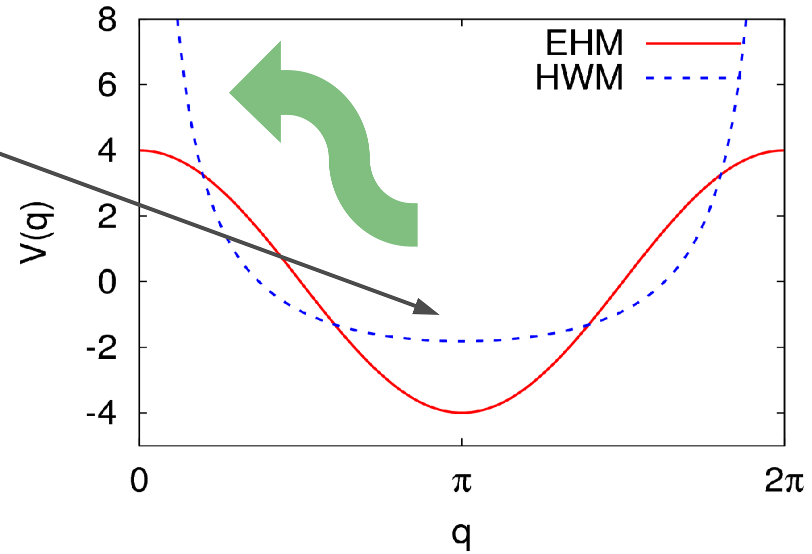
- RPA charge susceptibility $Q=(\pi,\pi)$:

  $$\chi_c(\mathbf{Q}) = \chi_0(\mathbf{Q}) / \left[1 + V(\mathbf{Q})\chi_0(\mathbf{Q})\right]$$

  $\rightarrow$ V_∞(HWM) =2.5 V_∞(EHM)

- ED: V_∞(HWM)=4t >> V_∞(EHM)=1.5t

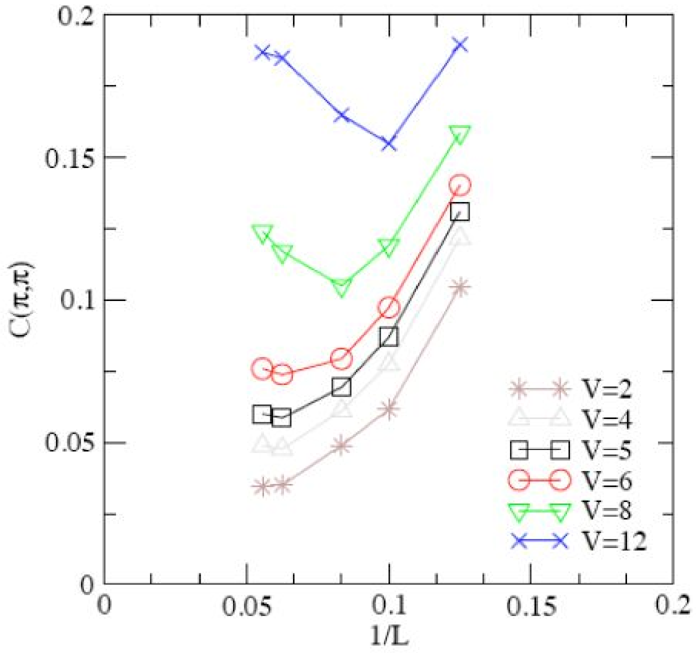
dominant interactions shifted to 1/q singularity



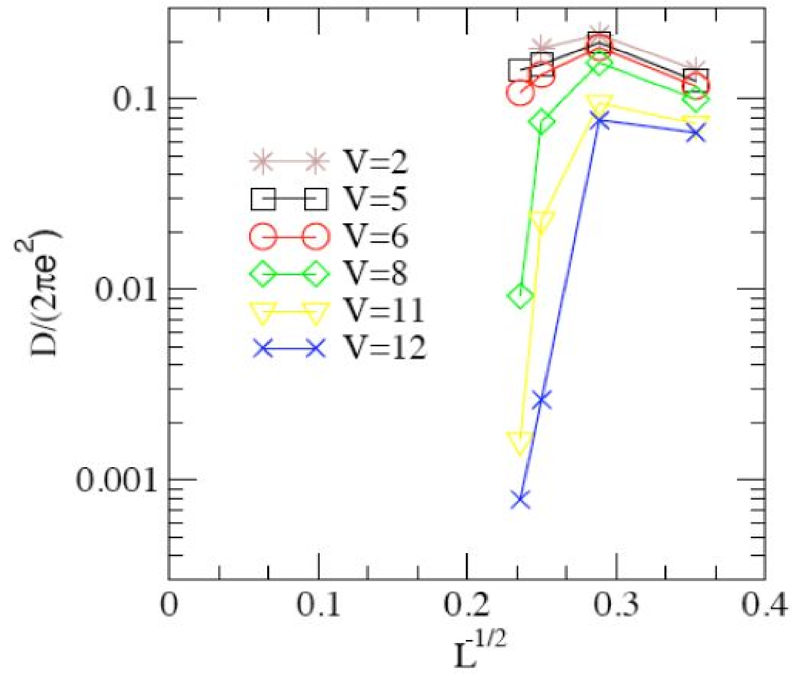
$\rightarrow$ Explains why compounds are metallic despite nominally large V and small bandwidths

$\rightarrow$ yet, the large energy scale V is still present locally in metallic phase: LR/SR separation, richer physics than in short range models

V_{MI} - Metal to Insulator transition > V_{CO}



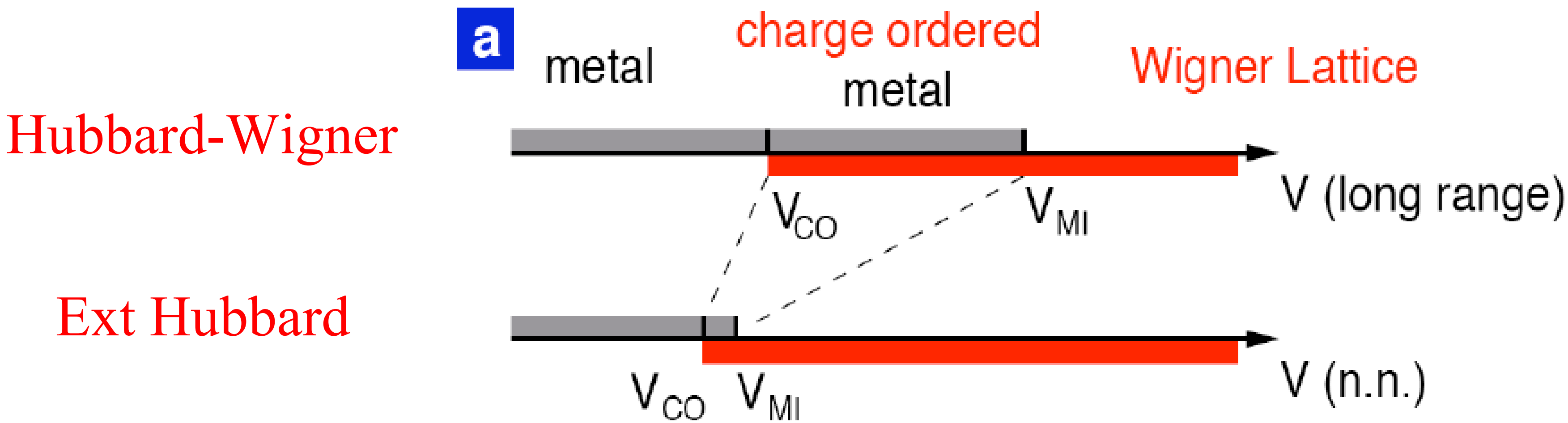
Charge order transition occurs at V_{co} ~4t. (1.5t if EHM)



Metal-insulator transition occurs at V_{MI} =8t

V_{MI} >V>V_{CO} : Intermediate phase separates Wigner crystal from homogeneous metal: charge ordered metallic phase !!!

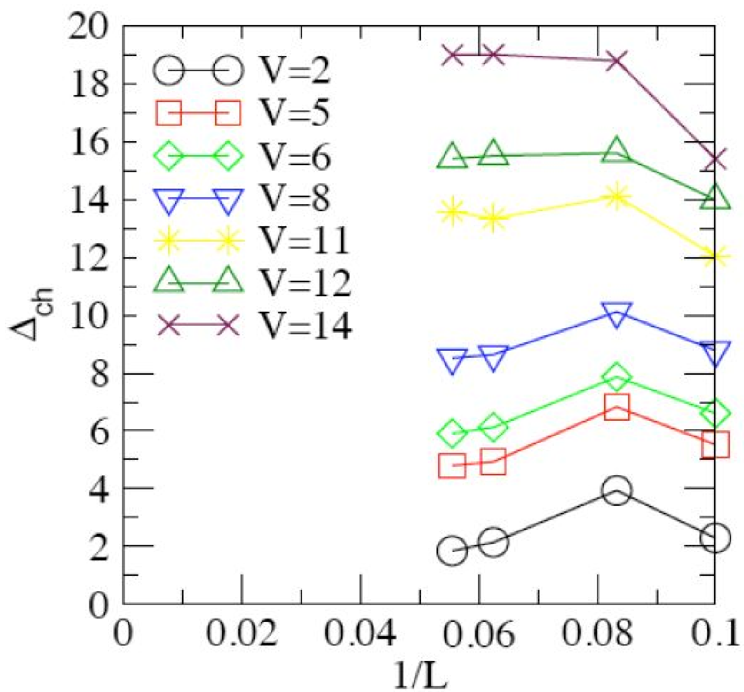
Phase diagram from ED



Long range repulsion:

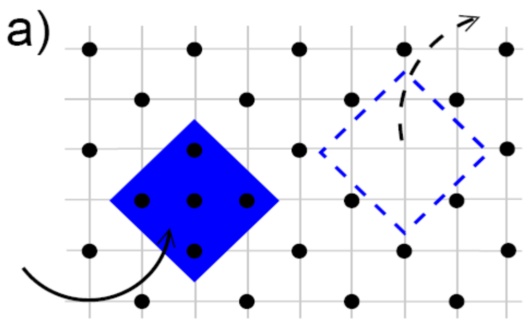
- V_{CO} increases compared to EHM
- Intermediate « strange » phase emerges: CO and metallic

Charge gap: even more anomalous!

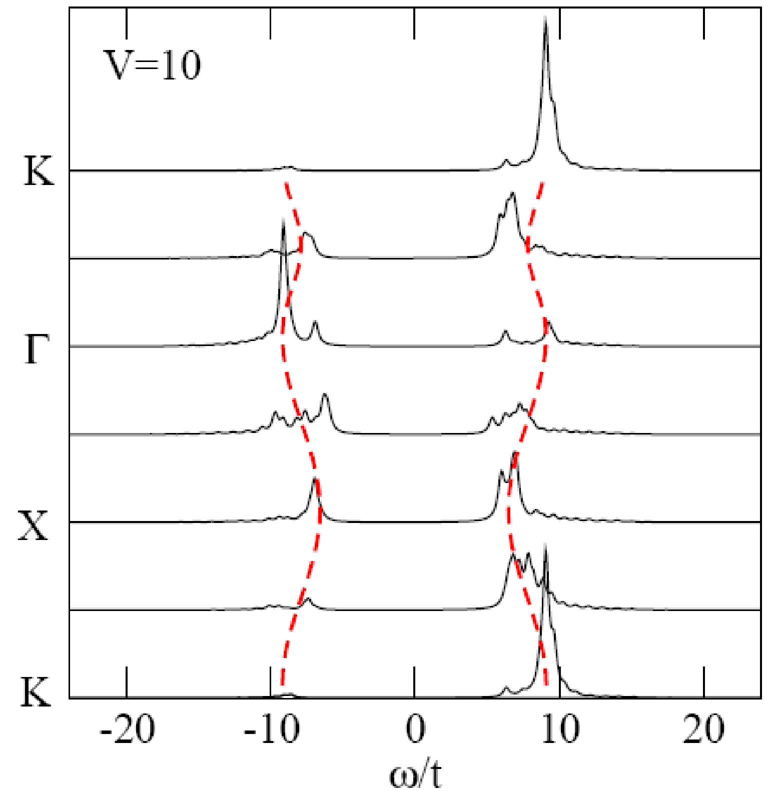


- One-particle (photoemission) charge gap opens up at the CO transition, V_{CO} =4-5t.
- Metallic phase without DOS at the Fermi energy for V_{CO} <V<V_{MI} !
  → who carries the current? collective excitations?

Collective excitations, large V/t limit: charged (photoemission)



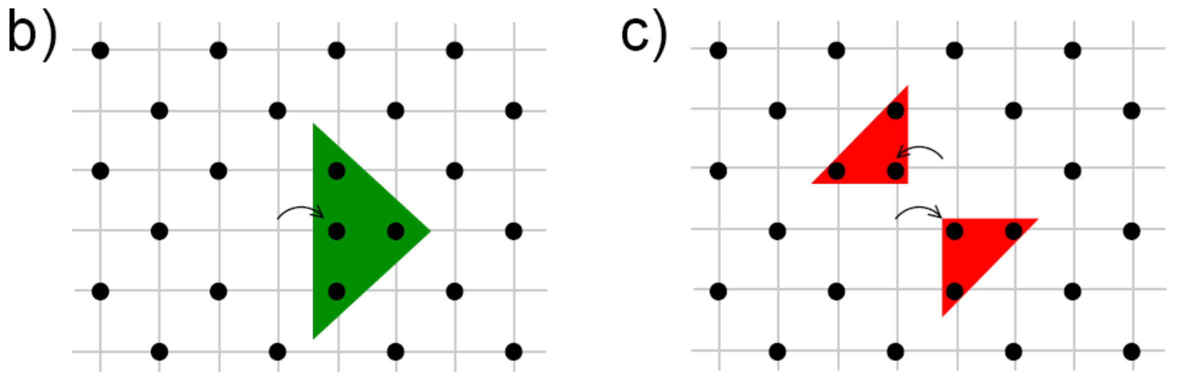
$$E_k = E_0 - 2t_1[\cos(2k_x) + \cos(2k_y)]$$
$$-2t_2[\cos(k_x + k_y) + \cos(k_x - k_y)].$$



"Pinball liquid" (cf. Hotta, Furukawa)
charges flowing on top of an underlying CO pattern, pin/ball separation

Collective excitations: charged (sails and butterflies) $\sigma(\omega)$, ε

dipolar, quadrupolar

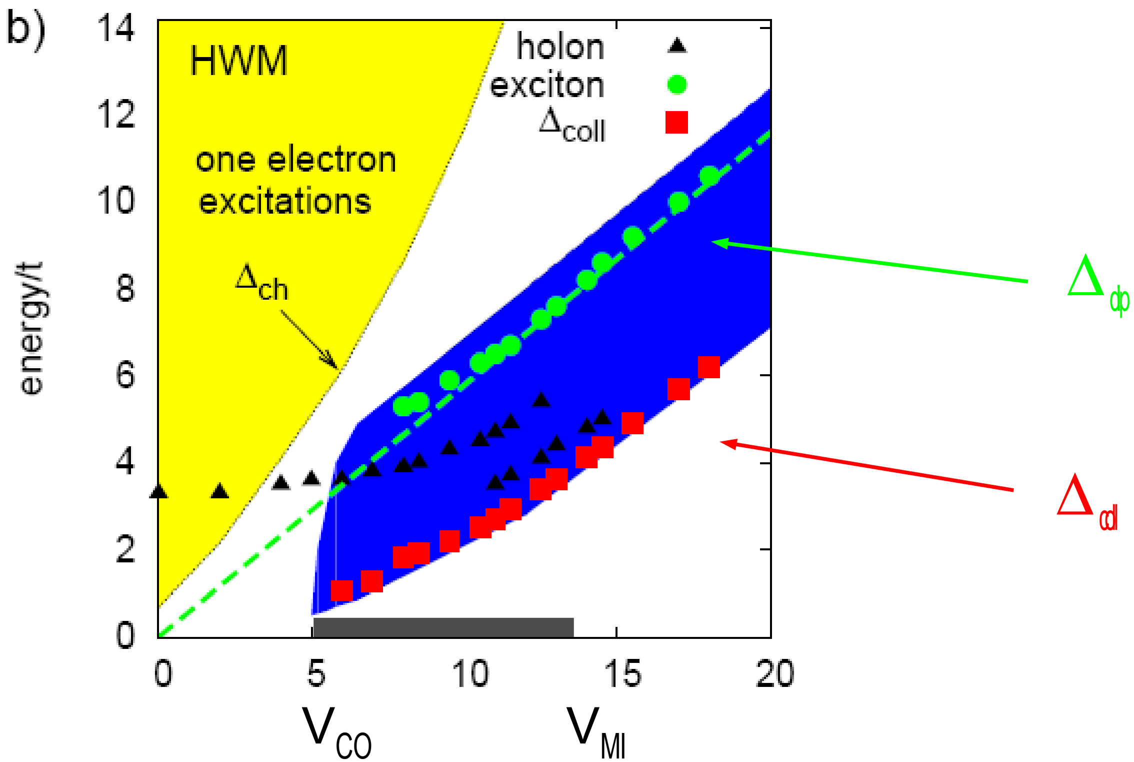


Δ_{dp} =3V $\leftarrow$ Extended Hubbard $\rightarrow \Delta_{quad}$ =4V

Δ_{dp} =0.62V $\leftarrow$ Hubbard-Wigner $\rightarrow \Delta_{quad}$ =0.65V

"Electrostatic frustration": large degeneracy due to long range Coulomb interaction leads to delocalization of defects!

Collective excitations: ED



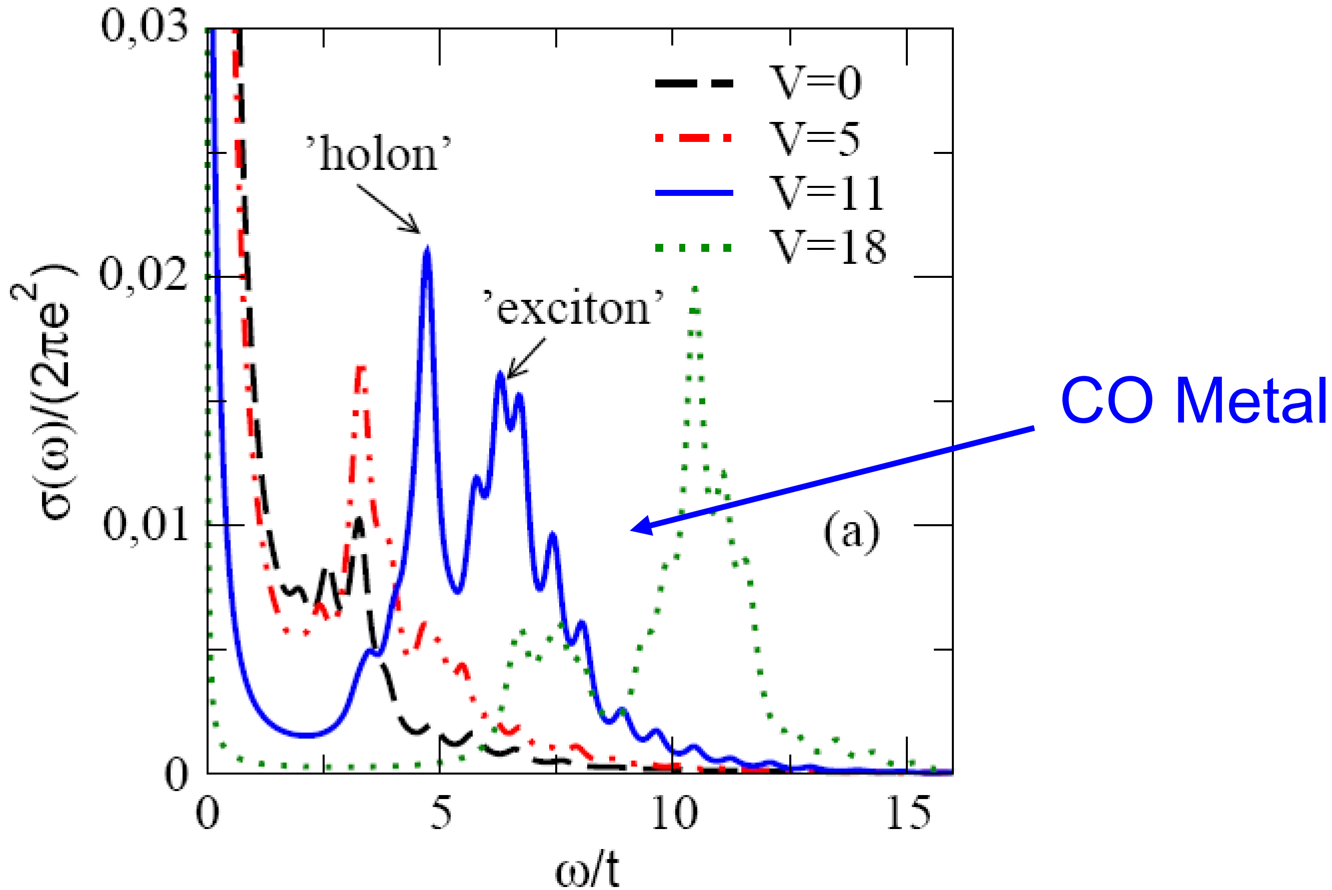
- Defects gain kinetic energy $\propto$t, by deconfinement.

$\Delta_{cd} = \Delta_{cp}$ -4.2t. [only ~t^2/V in EHM]

- Defects drive the transition to the anomalous phase, as they dominate the low energy behavior close to V_{MI} (cf. 1D case)
- Their precise nature is unknown (mixing with “Mott” collective excitations)

Optical conductivity in the anomalous phase

Both features related to Mott correlated metal and Wigner crystal



Conclusions

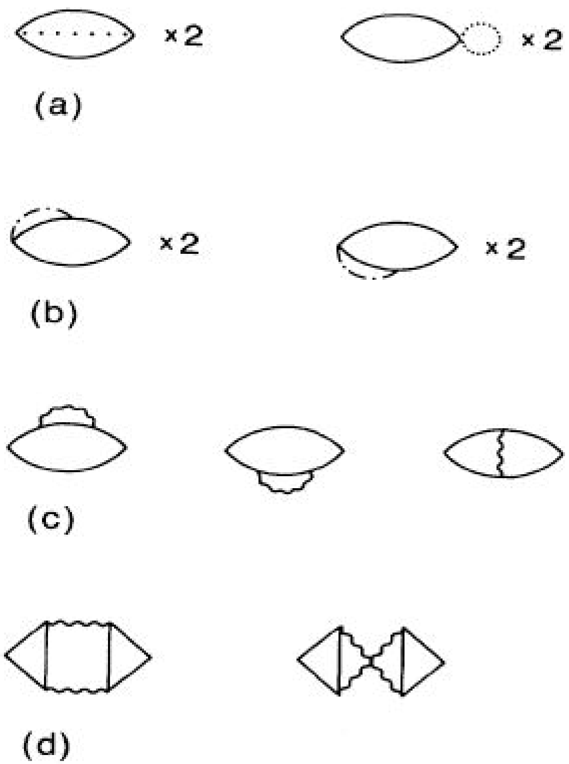
- Anomalous metallic state with Wigner crystal charge order AND Mott correlations occurs in phase diagram of 2D Hubbard-Wigner model:
  - Drude peak is non-zero signalling a metallic state.
  - One-electron excitations display a gap → collective modes
  - Delocalized defects due to long range Coulomb repulsion are responsible of current flow despite the charge gap.
- Key experiments:
  - Optical spectroscopy should observe a Drude peak and excitonic + “holon” band.
  - Photoemission experiments should observe a charge gap.
  - X-ray, NMR, should observe charge ordering

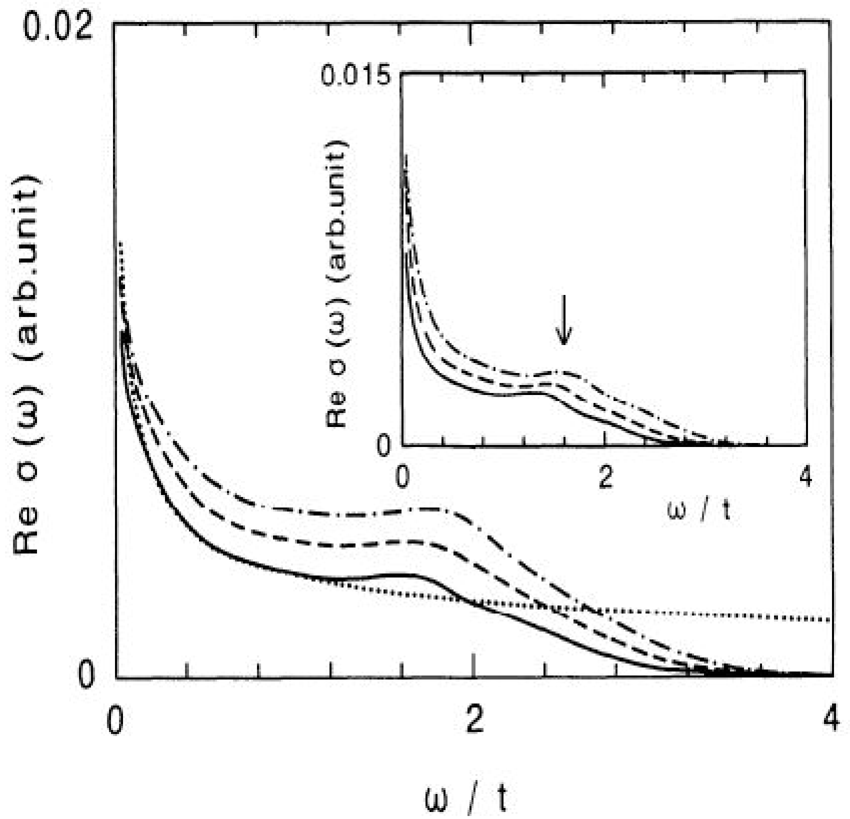
Relevance to organics: θ-(BEDT-TTF)$_2$X

Quarter-filled organics are ideal systems for exploring the quantum melting of a Wigner crystal on a lattice (tunable V/t)

- X=I_3 is a correlated metal, with no visible Drude peak and resistivity well beyond the Mott-Ioffe-Regel limit [Takenaka PRL (2005)]
- X=RbZn(SCN)$_4$ salt displays a CO transition at T_{CO} =195 K.
  - Resistivity has a metallic behavior for $T>T_{CO}$.
  - Short range ordering found by NMR for $T>T_{CO}$. [Chiba PRL (2004)]
  - Thermopower shows both localized and mobile carriers [Abdel-Jawad PRB (2009)]
  - STM/STS finds (?) a large gap of 0.6 eV for $T<T_{CO}$
    Even above T_{CO} a small gap of about 0.1 eV is observed. [Ichimura JLTP (2009)].
- X=CsZn(SCN)$_4$ salt displays nonlinear resistivity ascribed to collective excitations [Inagaki JPSJ (2004), Sawano Nature (2005)]

‘Holon’ in strongly correlated 2D metal

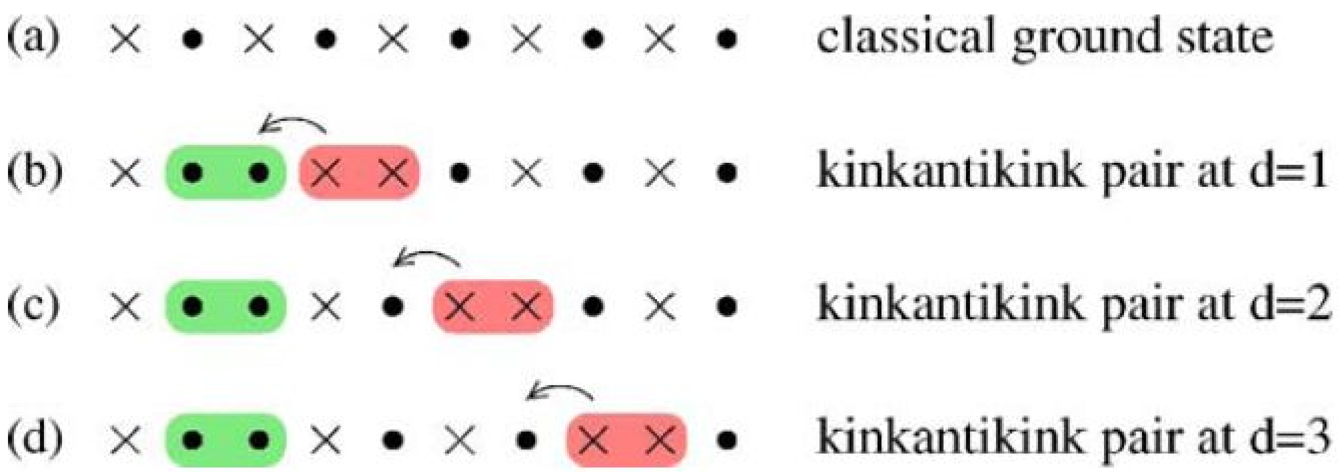




Large-N theory of ‘t-J’ model: Bang & Kotliar, PRB (1993).

Wigner crystal on a 1D chain: collective excitations and charge fractionalization

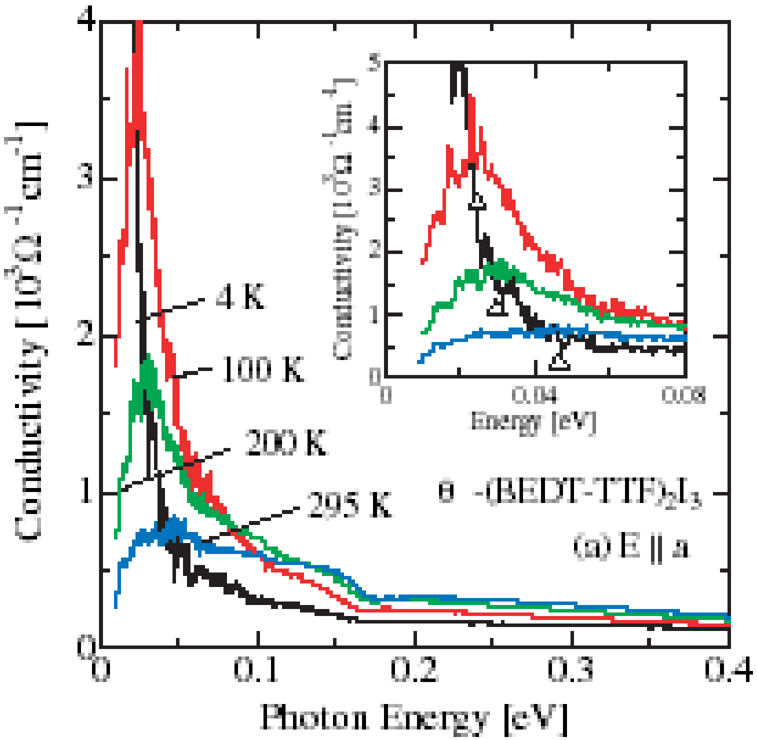
J. Hubbard, PRB (1978), Fratini et. al., Synth. Met. (2004), Mayr and Horsch, PRB (2006).

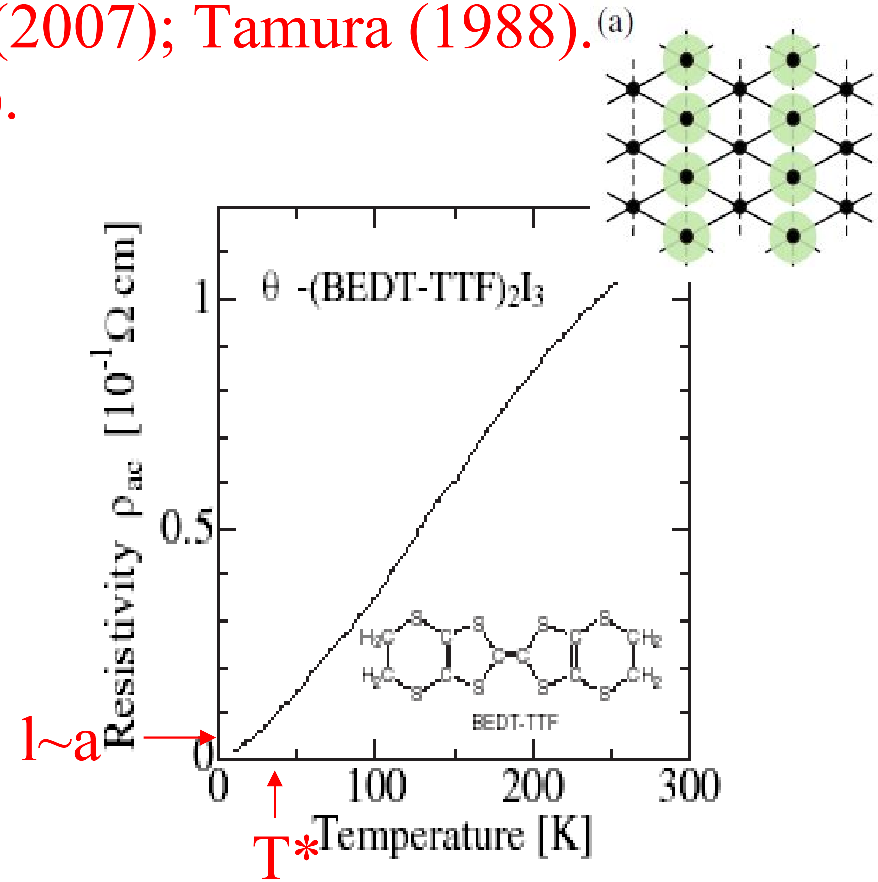


The degeneracy of the classical ground states
leads to the formation of domain walls of fractional charge.

Correlated metal: θ-$(ET)_2I_3$

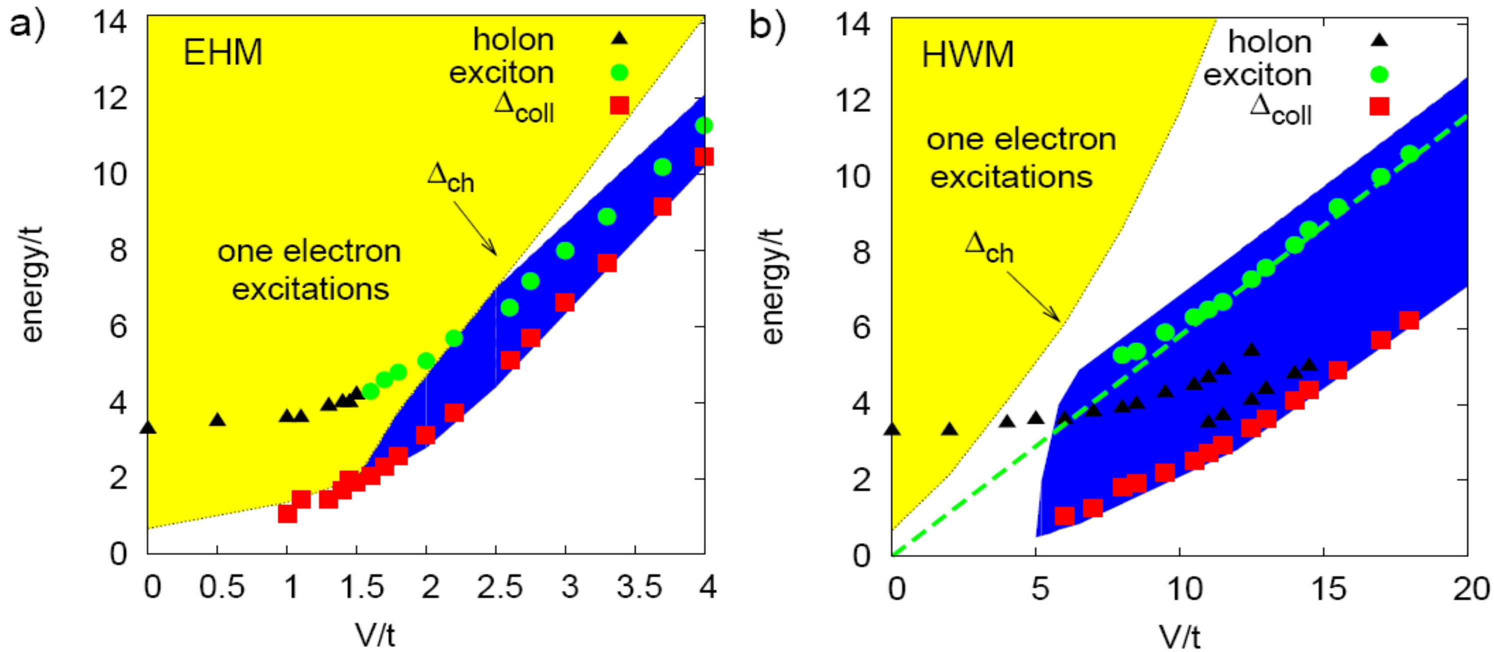
Takenaka, et. al., PRL (2005); PRL (2007); Tamura (1988).
Gunnarsson and Vafayi, PRL (2007).





•No visible Drude peak at T>4 K.
•Metallic resistivity increases above the Mott limit T>50 K.

Collective excitations: HWM vs EHM



- In the EHM, the transition occurs when Δ_{ch}~W → V/t~2
- In the HWM defects gain kinetic energy ∝t, by deconfinement and remain as the low energy sector down to the transition.

$\Delta_{coll}=\Delta_{dp}-4.2t$. [only ~t^2/V in EHM]