Wigner-Mott transition, charge frustration and Hund's coupling in layered triangular lattices

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Outlook

- Coulomb induced charge ordering: Wigner/Mott
  electron correlations away from integer fillings

- Frustration of CO on triangular lattices: pinball liquid
  non FL, bad metal, heavy fermion behavior…

- Experimental realizations?
  $AgNiO_2$, multiband system
  pinball order stabilized by Coulomb + Hund's coupling

Wigner crystallization

DECEMBER 1, 1934 PHYSICAL REVIEW VOLUME 46

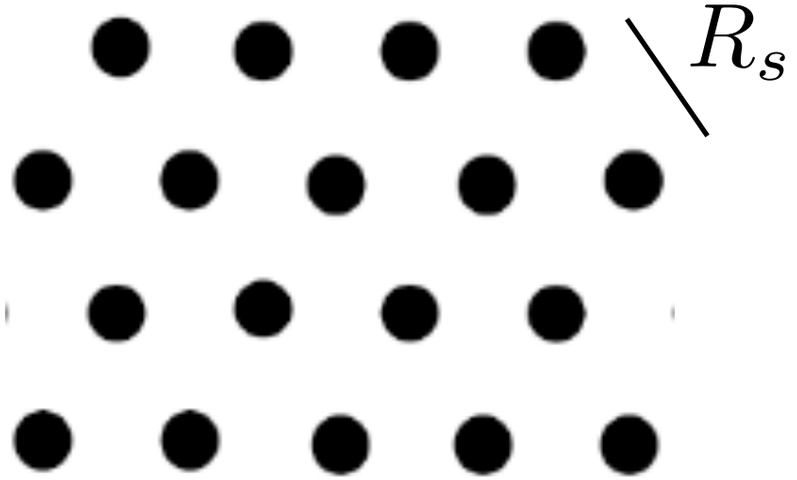
On the Interaction of Electrons in Metals

E. WIGNER, *Princeton University*

(Received October 15, 1934)

If the electrons had no kinetic energy, they would settle in configurations which correspond to the absolute minima of the potential energy. These are closed-packed lattice configurations, with energies very near to that of the body-centered lattice.

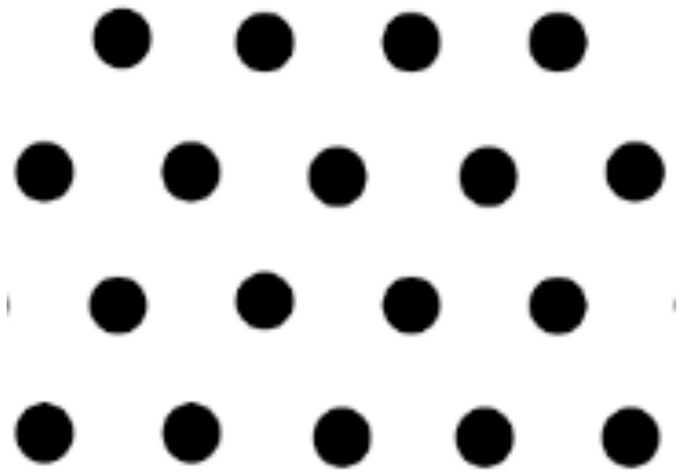
$$V(r) = \frac{e^2}{r}$$



$$\frac{e^2}{R_s} \gg \frac{\hbar^2}{2mR_s^2}$$

Coulomb interactions: Wigner vs Mott insulator

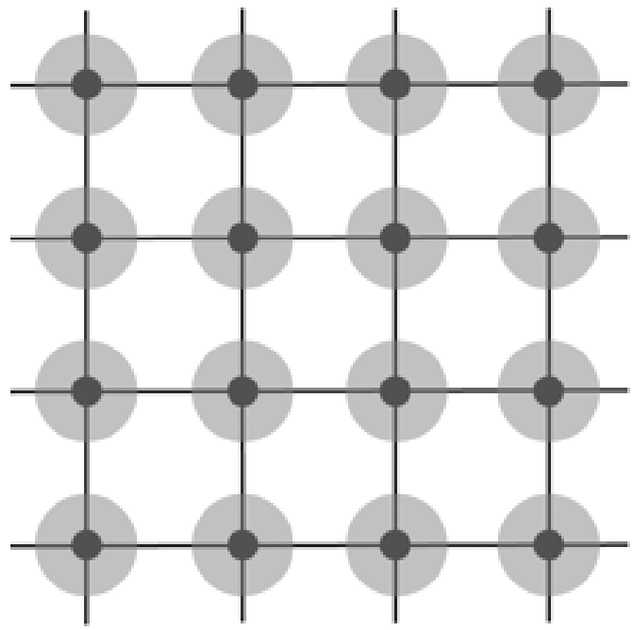
$$V(r) = \frac{e^2}{r}$$



$$\frac{e^2}{R_s} \gg \frac{\hbar^2}{2mR_s^2}$$

Wigner crystal

- no underlying lattice
- long range interactions
- low electron density

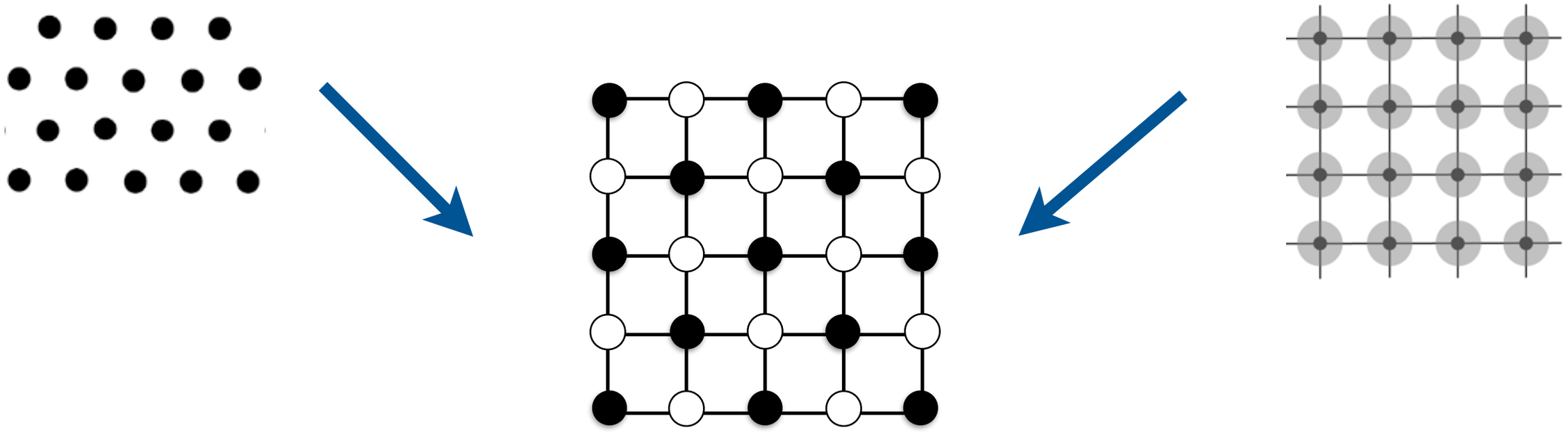


$$U \gg W$$

Mott insulator

- commensurate underlying lattice
- on-site interactions, Hubbard model
- high electron density, n=1

Coulomb induced CO in solids: Wigner AND Mott physics



order when V>>t

Wigner-Mott CO insulator

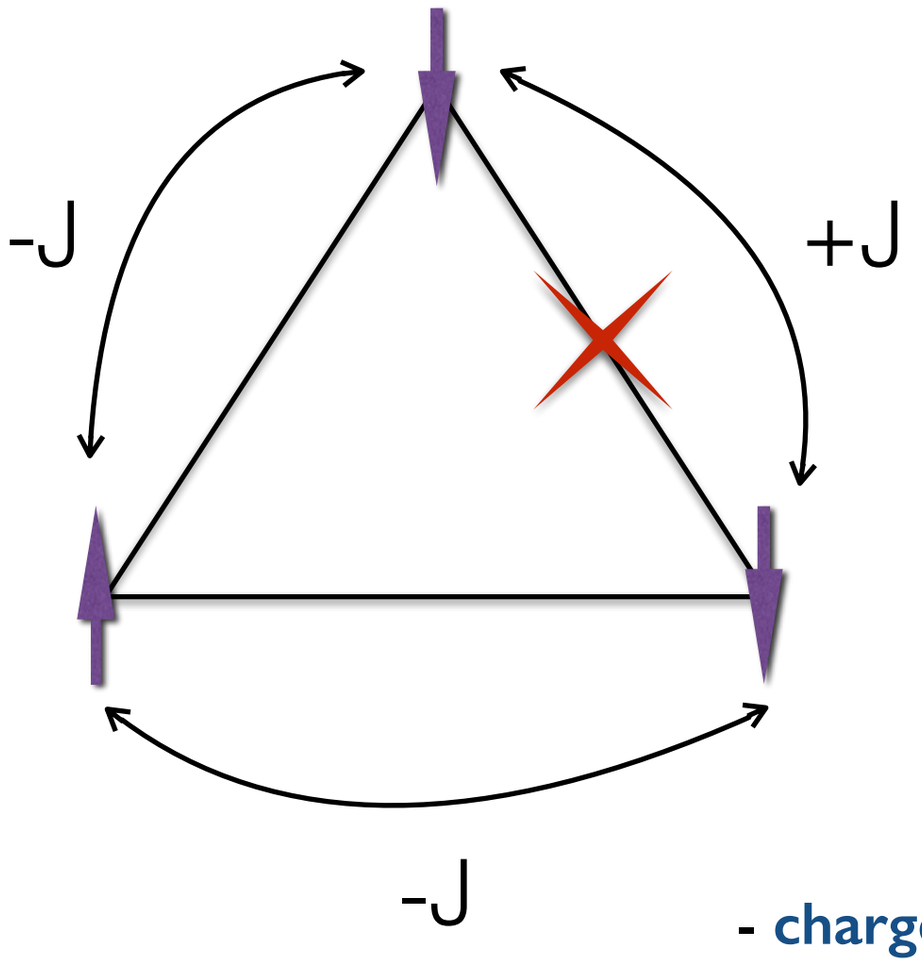
- intermediate electron density, n=1/2: quarter filling (common in 2D organics, some oxides)
- interplay of charge order and electronic correlations
- Extended Hubbard Model: on-site + long range interactions

$$H = -t\sum_{\langle ij\rangle\sigma}(c^{\dagger}_{i\sigma}c_{j\sigma} + \text{H.c.}) + U\sum_{i} n_{i\uparrow}n_{i\downarrow}$$

Triangle = Frustration

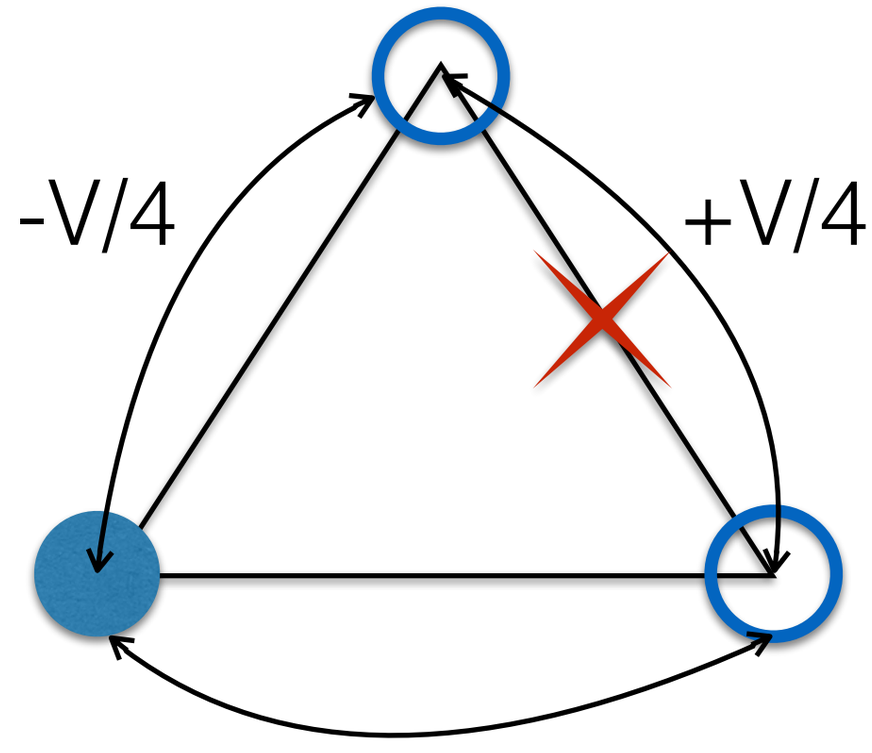
Magnetic

$$J \sum_{\langle ij \rangle} S_i S_j$$



Charge (n=1/2)

$$V \sum_{\langle ij \rangle} (n_i - 1/2)\,(n_j - 1/2)$$



- **charge frustration** is far less studied than its spin analogue
- contrary to spins, **charges are itinerant**
- to be **explored**!

Square vs.Triangular lattice

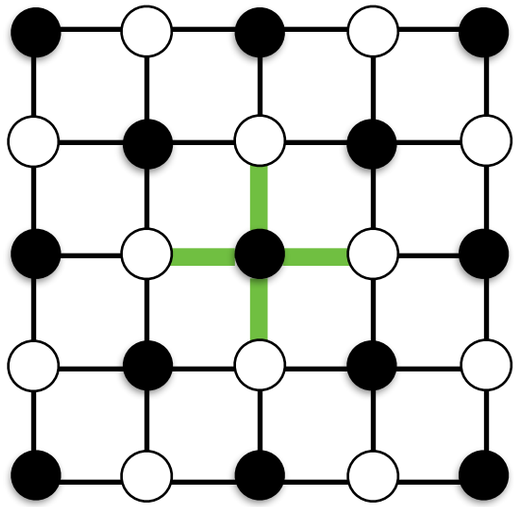
[Kaneko et al. PRB'06, Hotta et al. PRB'06, Cano-Cortés et al. PRL'10, PRB'11, Merino et al. PRL'13]

square lattice, one electron per two sites (n=1/2)

nearest neighbor
Coulomb repulsion

$$Vn_in_j$$

Extended Hubbard Model



all bonds are happy

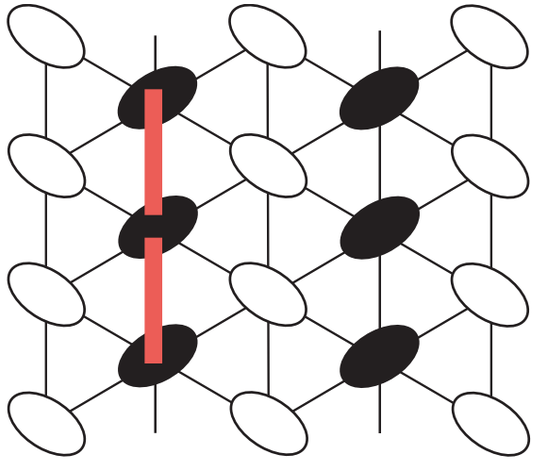
- "checkerboard" order
- CO insulator (gapped)
- 2x degenerate

Square vs.Triangular lattice

[Kaneko et al. PRB'06, Hotta et al. PRB'06, Cano-Cortés et al. PRL'10, PRB'11, Merino et al. PRL'13]

triangular lattice, one electron per two sites (n=1/2) θ-(BEDT-TTF)$_2$X

"stripes"



E/N=V/2

Classical ground state infinitely degenerate.

frustrated neighbors!

+ Thermal fluctuations
-> L. Rademaker tomorrow

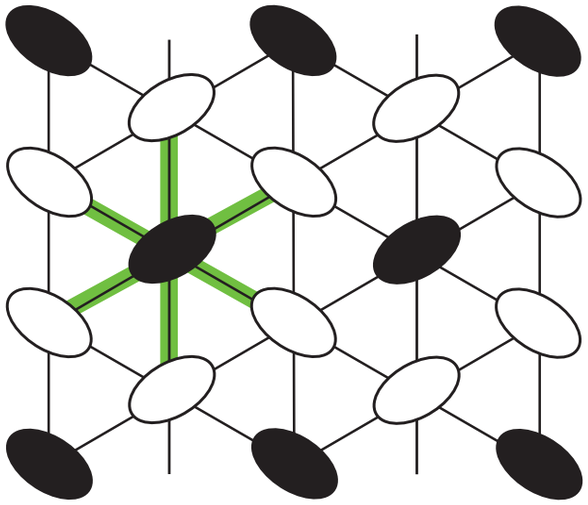
+ Quantum fluctuations
lead to exotic phases

the pinball liquid

[Kaneko et al. PRB'06, Hotta et al. PRB'06, Cano-Cortés et al. PRL'10, PRB'11, Merino et al. PRL'13]

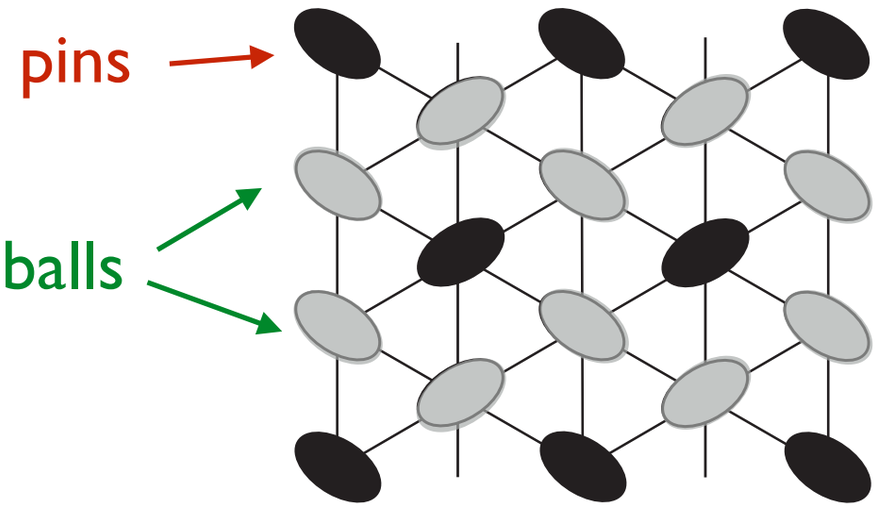
on the triangular lattice we can't make n=1/2 electrons happy,
but at least can we make them less unhappy ?

fill as many sites as possible
avoiding frustration (n=1/3)



E/N=0

all remaining charges to reach n=1/2
move to interstitials (honeycomb lattice)



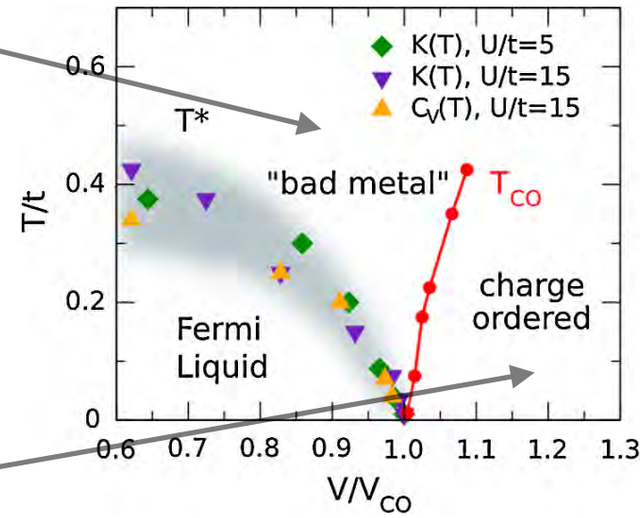
E/N=V/2

- same electrostatic energy as stripes, but gains kinetic energy
- frustration is released via quantum fluctuations
- coexisting localized charges (pins) and itinerant carriers (balls) on the honeycomb lattice
- pinball liquid: CO metal, with quite interesting properties

2 routes to non Fermi liquid behavior

1) quantum criticality: approaching the CO transition, scattering by critical charge fluctuations up to T~t (bad metallic properties, large mass enhancement, destruction of qp…) consistent with exp. in θ-$(BEDT\text{-}TTF)_2I_3$

[Cano-Cortés et al. PRL'10, PRB'11]
single band EHM on triangular lattice



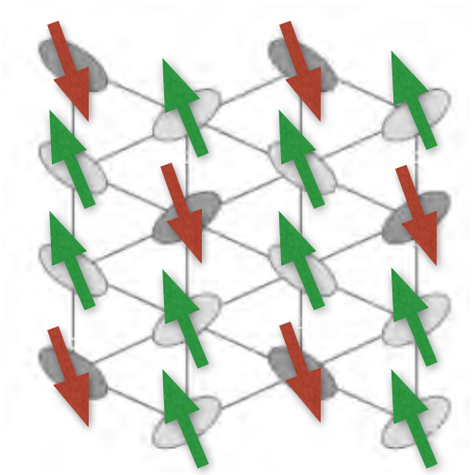
[Merino et al. PRL'13]

2) pinball liquid: within the CO phase, spontaneous separation into localized and itinerant carriers

analogy with **heavy fermions**

pins

balls



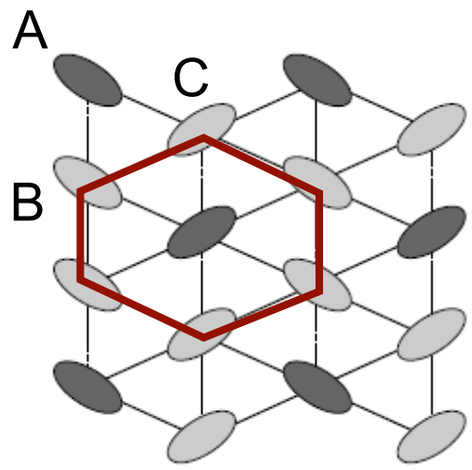
f-electrons

d-electrons

conduction electrons are scattered by the fluctuations of the localized moments

Pinball liquid: exciting properties

[Merino et al. PRL'13]



Can be mapped to the Periodic Anderson Model

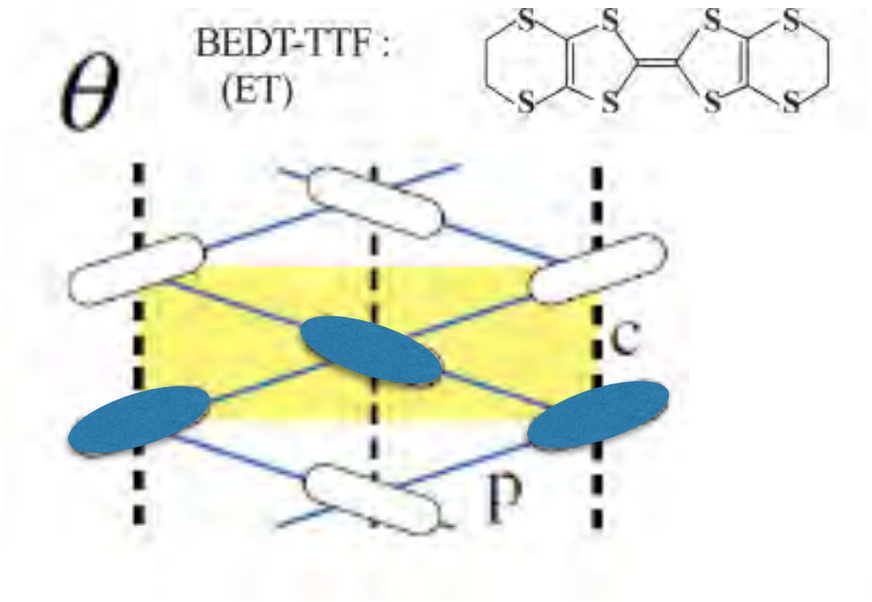
$$H_{\mathrm{PAM}} = \sum_{i\sigma} \epsilon_A a^\dagger_{i\sigma} a_{i\sigma} + \sum_{k\sigma\alpha} \epsilon_{k,\alpha} \gamma^+_{k\alpha\sigma} \gamma_{k\alpha\sigma}$$
$$+ \sum_{ik,\alpha} (V_{k,\alpha} e^{ikR_i} a^\dagger_{i\sigma} \gamma_{k\alpha\sigma} + \mathrm{H.c.})$$
$$+ U \sum_i a^\dagger_{i\uparrow} a_{i\uparrow} a^\dagger_{i\downarrow} a_{i\downarrow},$$

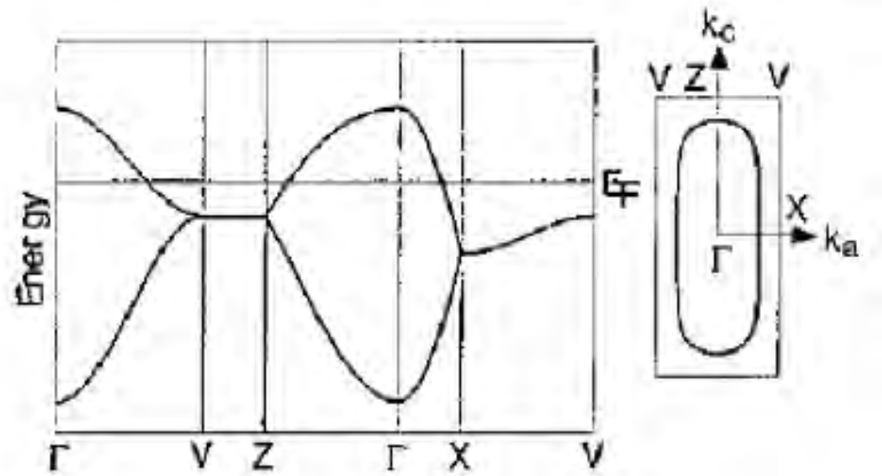
	heavy fermions	pinball liquid
localized moments	f-electrons	pins
conduction electrons	d-electrons	balls

Emergent heavy fermion behavior expected
(Non FL, bad metal, quantum criticality etc.)

Experimental realizations?

Layered ET_2X organic conductors





$$\varepsilon_{\boldsymbol{k}} = 2t_p[\cos(k_x) + \cos(k_y)] + 2t_c \cos(k_x + k_y)$$

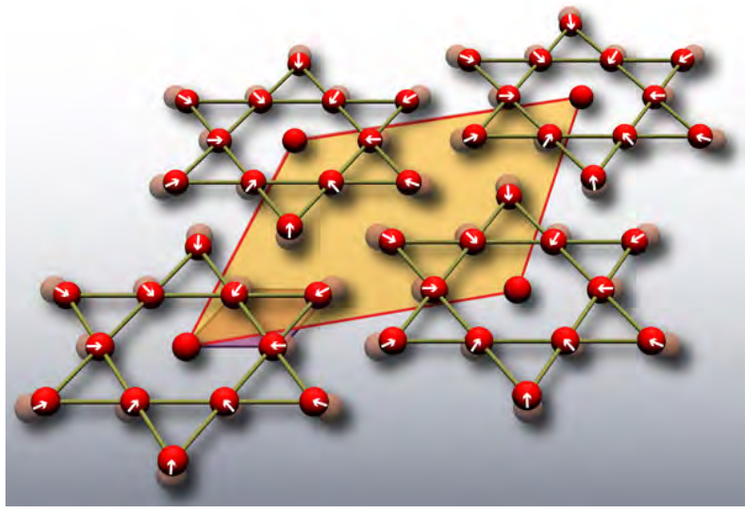
- charge transfer: 1 hole/2 molecules -> quarter filled bands
- single band tight binding description, W~0.5eV
- narrow bands, effects of interactions should be strong
- U,V>W lead to charge ordered phases
- BUT triangular lattice is anisotropic:

striped phases commonly observed, but no PL

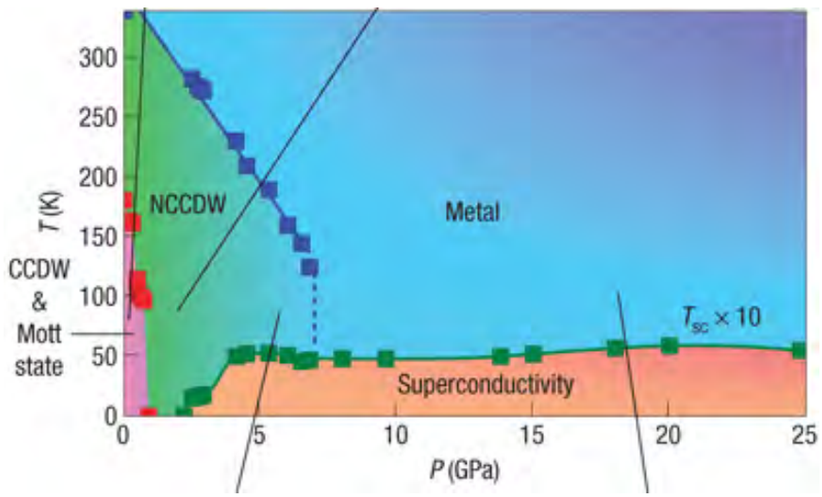
dichalcogenides, oxides

2D transition metal dichalcogenides

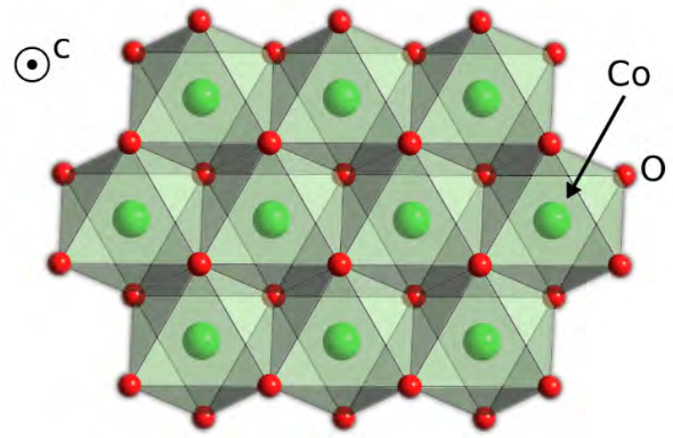
TaS_2,$NbSe_2$,…

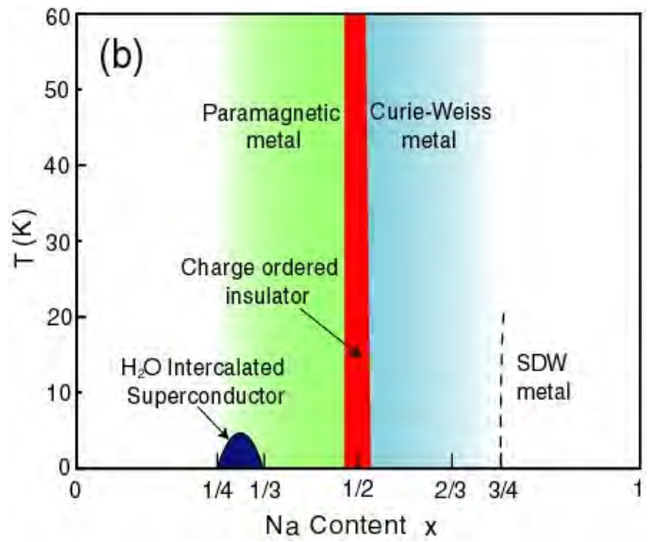


Sipos et al. Nat Mater (2008)



1T-TaS_2:
Mott physics driven by CO
David-star √13x√13 unit cell



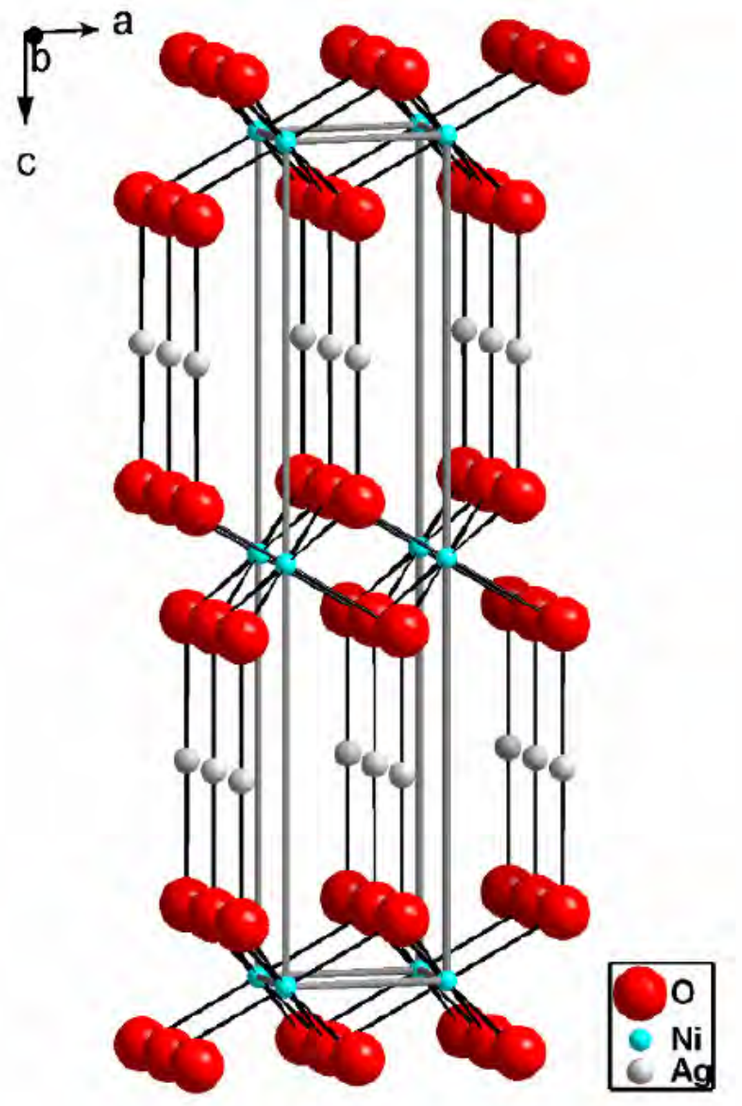


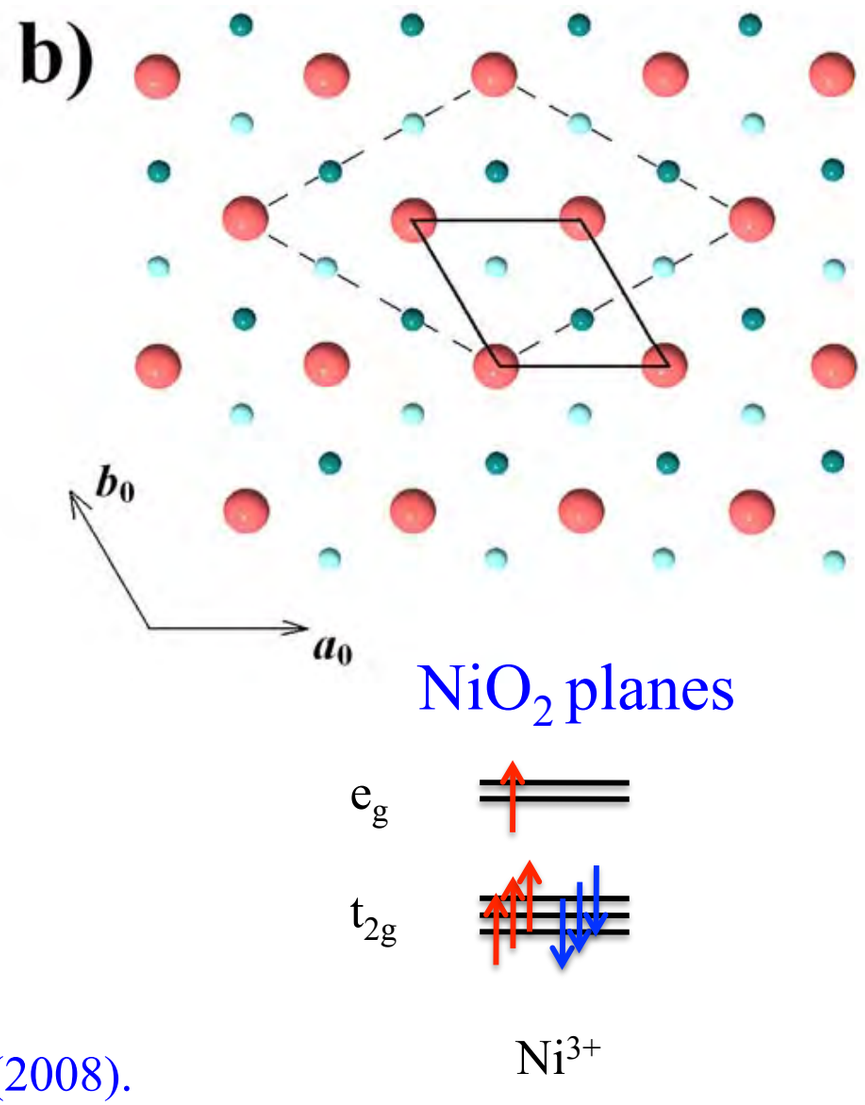
2D transition metal oxides

ex. cobaltates Na_xCoO_2

but also: dichalcogenide monolayers,
ionic liquid doped organic semiconductors,
adsorbate lattices, trapped cold ions,…

The case of AgNiO2

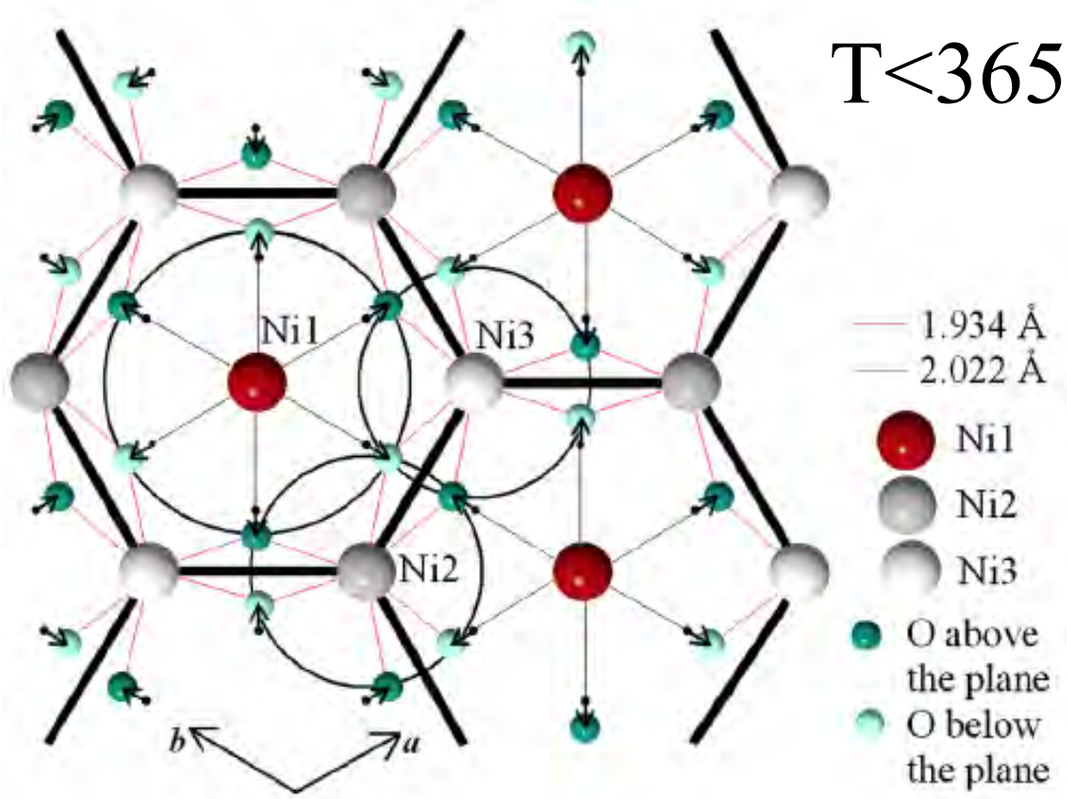


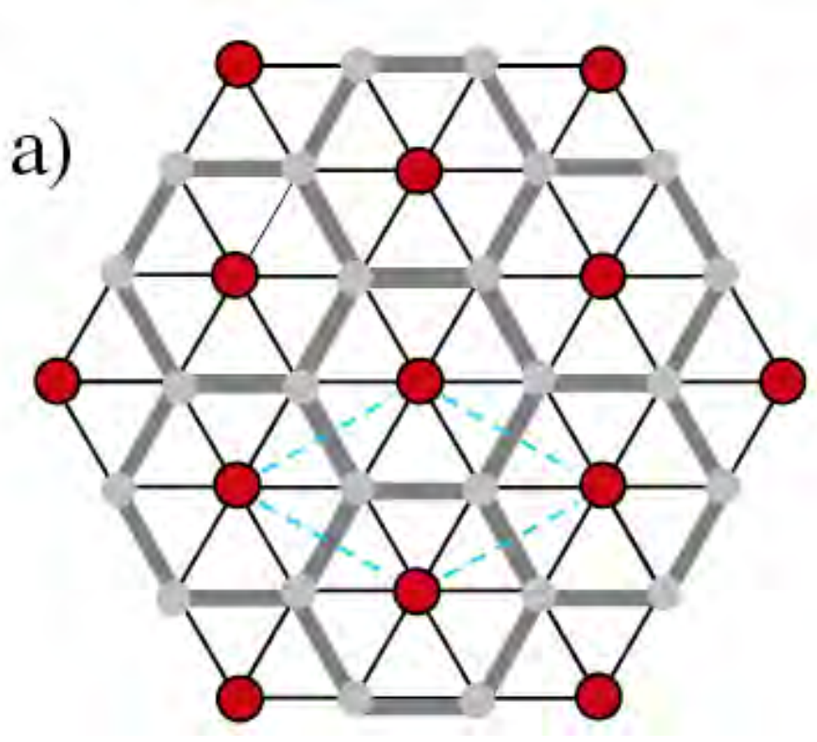


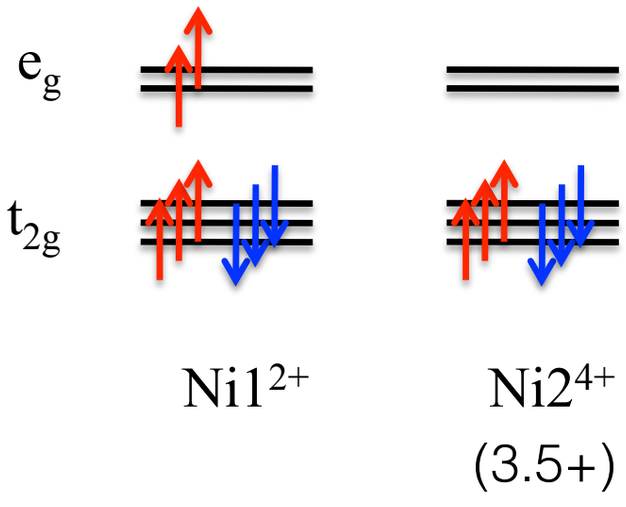
E. Wawrzynska, et. al. PRB 77, 094439 (2008).

- triangular lattice of Ni ions
- 1 electron per site per 2 orbitals -> quarter filling
- Pinball liquid physics expected!

X-ray / neutron scattering

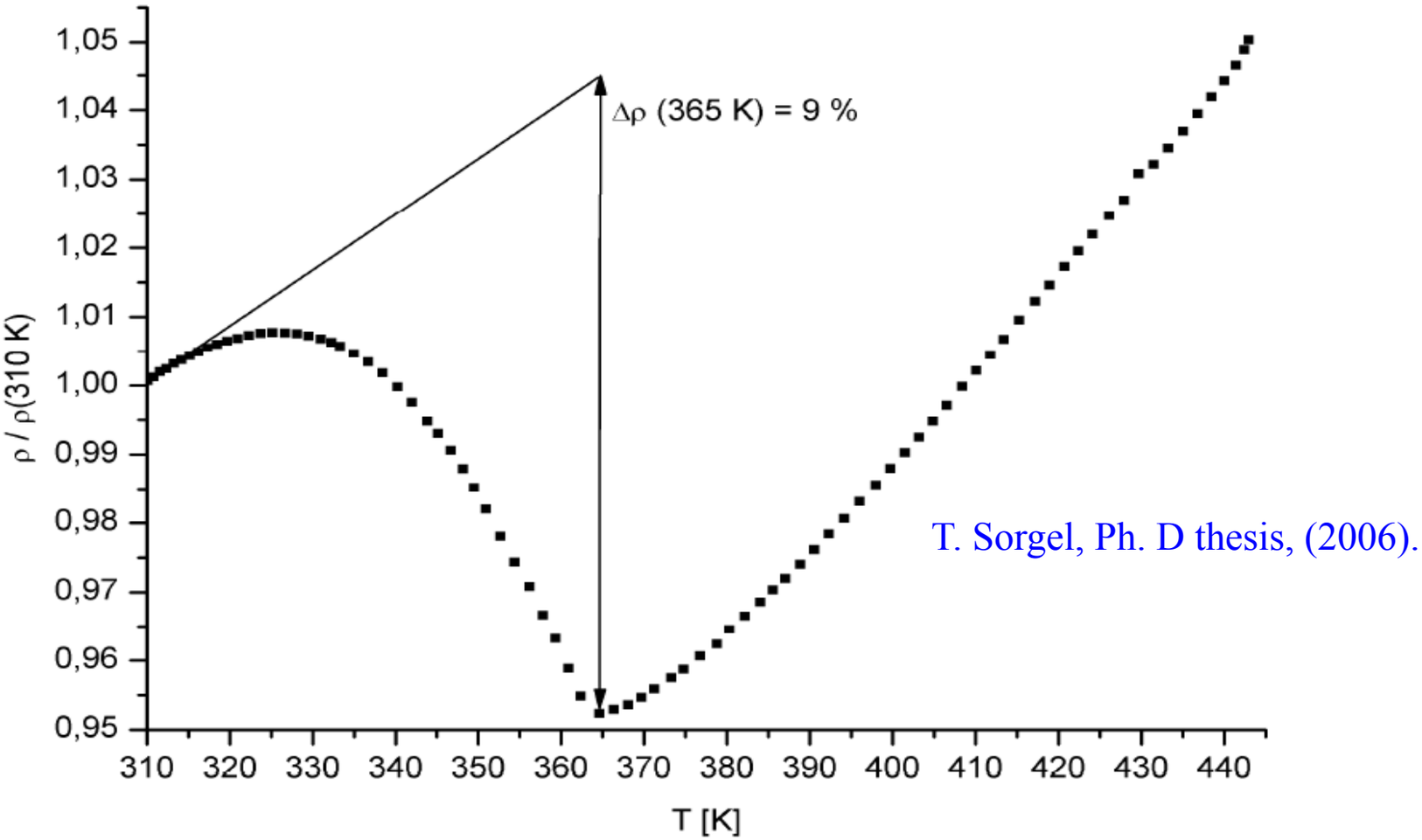






E. Wawrzynska, et. al. PRL 99, 157204 (2007);
PRB 77, 094439, (2008).

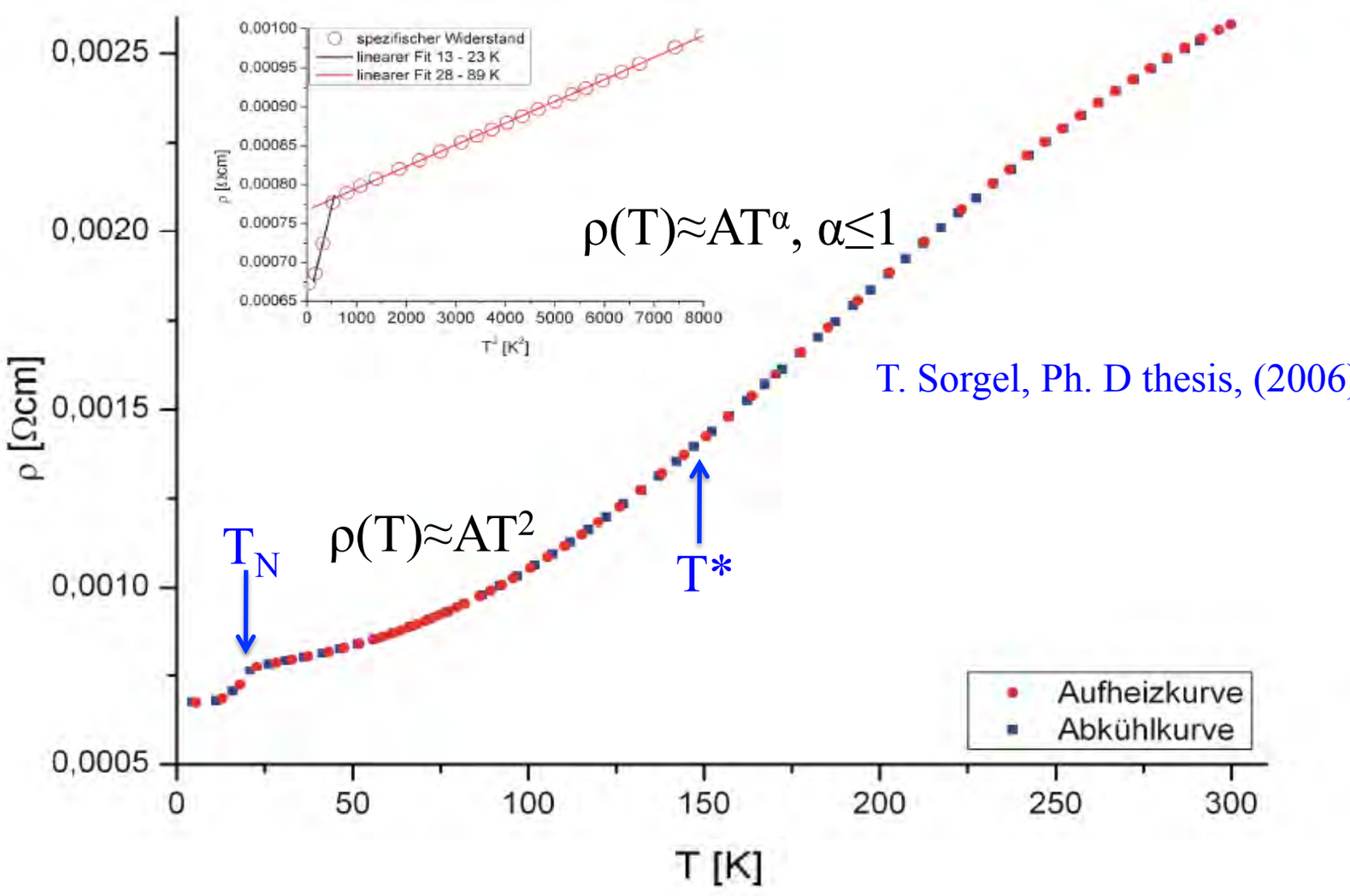
Resistivity (1): CO transition



T. Sorgel, Ph. D thesis, (2006).

- CO is a metal-metal transition
- weak anomaly in resistivity

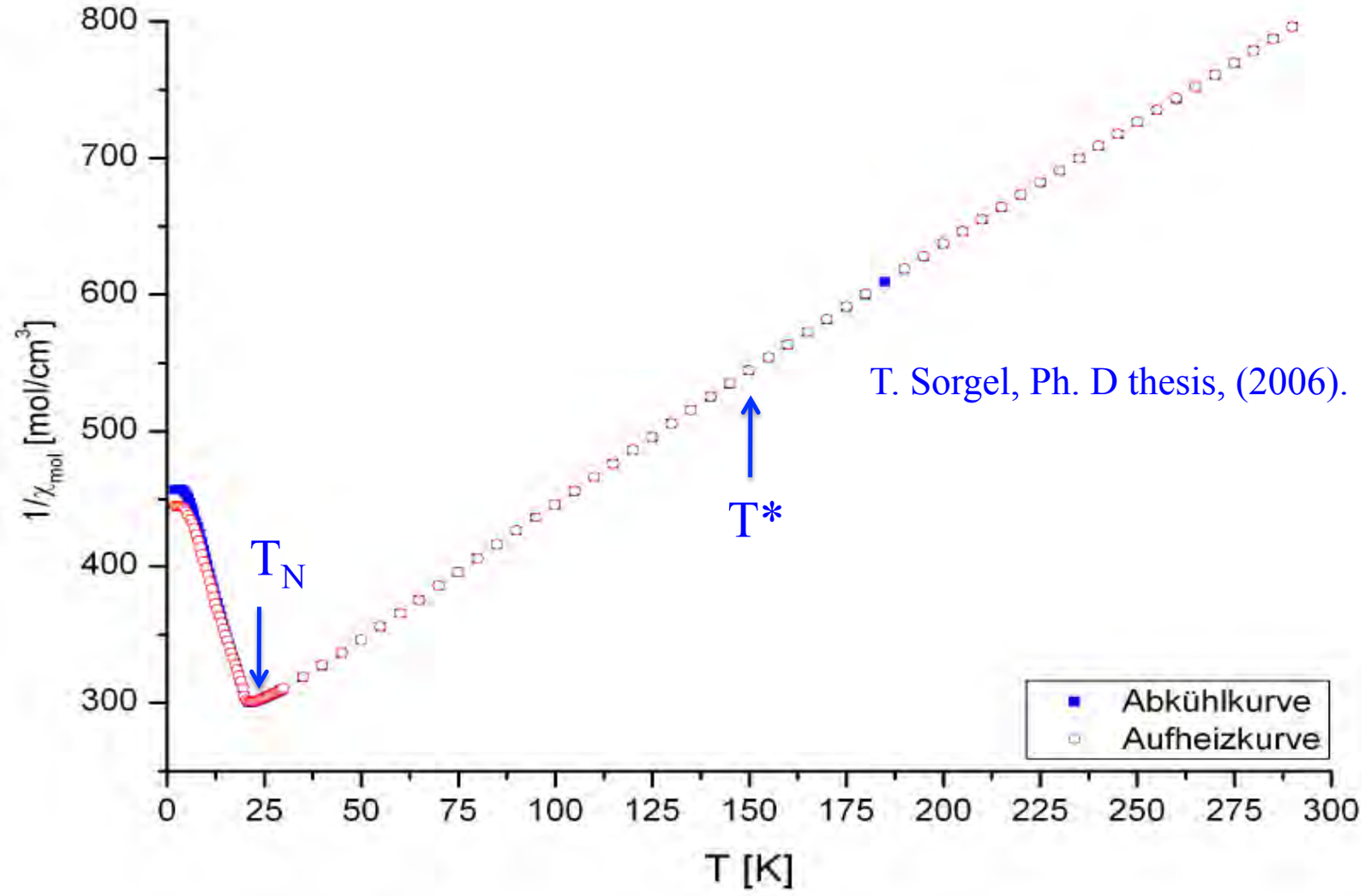
Resistivity (2): bad metal



T. Sorgel, Ph. D thesis, (2006).

- resistivity larger than Mott-Ioffe-Regel limit ($\sim m\Omega cm$)-> bad metal
- deviations from T^2 behavior above $T^*\sim 150K$

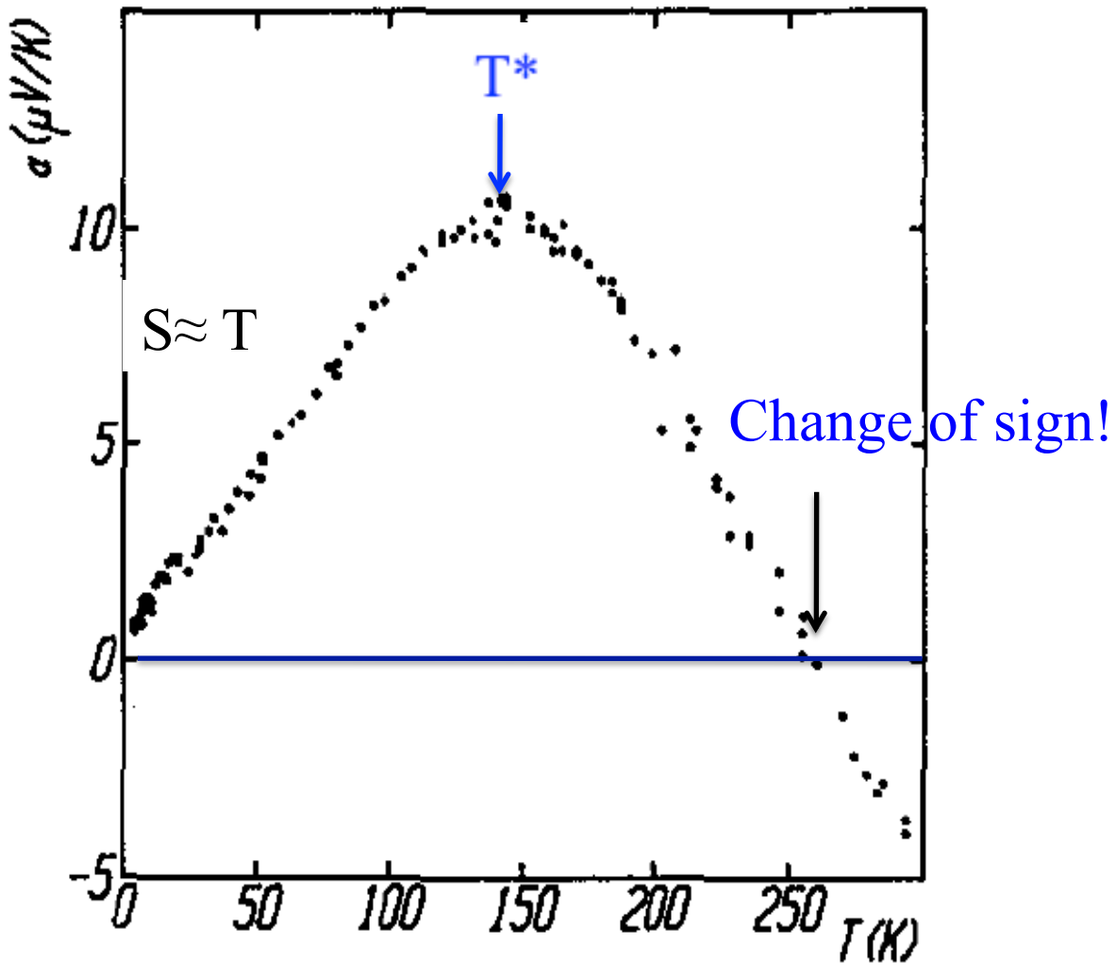
Curie-Weiss susceptibility



T. Sorgel, Ph. D thesis, (2006).

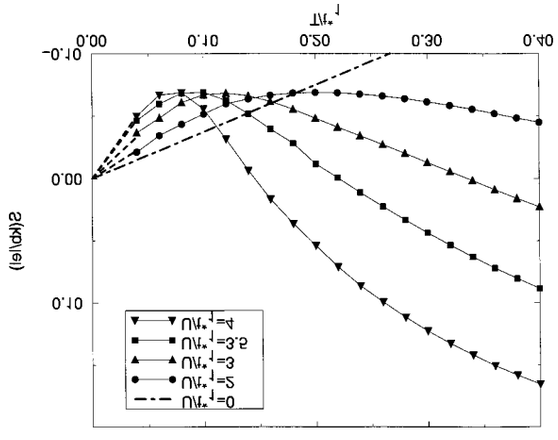
- Large local moments $\mu_{\text{eff}} = 1.81\text{–}1.96$ (from DFT $\mu_{\text{eff}} = 1.3\text{–}1.5$)
- change of slope at T*

Thermopower



A. Wichainchai, Jour. Sol Stat. Chem., 74 126 (1988).

[Merino, Mc Kenzie PRB(00)]



Optical conductivity

?

Electronic properties of $AgNiO_2$

[Ralko , Merino, Fratini, PRB'15;
Février, Fratini, Ralko, PRB'15]

Unlike conventional metals:

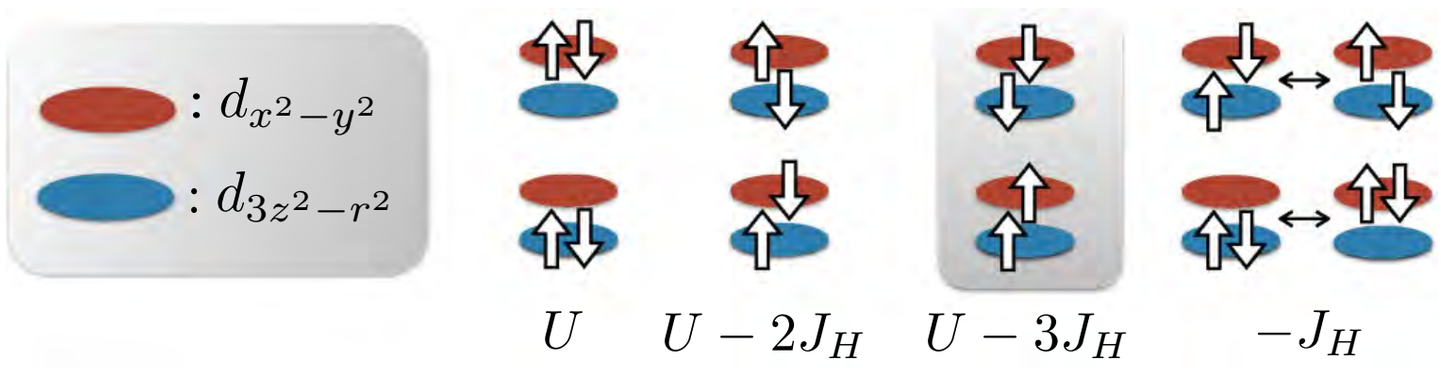
- Charge ordered and metallic, CO is threefold
- Large local moments, order at $T_N << T_{CO}$
- Large resistivity, deviates from T^2
- Maximum in thermopower at T* signals crossover from coherent to incoherent excitations. Change of sign at 260 K.
- Large specific heat (m*/m~2.6)
- No Fermi surface at RT (but ARPES only on polycrystals).
- electronic correlations, destruction of qp above T*

PINBALL LIQUID?

2 orbitals: Extended Hubbard Model + Hund

[Ralko , Merino, Fratini, PRB'15;
Février, Fratini, Ralko, PRB'15]

$$H = -t \sum_{\langle ij \rangle} \sum_{\tau,\sigma} (d^{\dagger}_{i,\tau\sigma} d_{j,\tau\sigma} + \text{H.c.}) + H_{\text{V}} + H_{\text{Hund}}$$



$$H_{\text{Hund}} = U \sum_{i,\tau} n_{i\tau\uparrow} n_{i\tau\downarrow} + (U - 2J_H) \sum_{i,\tau \neq \tau'} n_{i\tau\uparrow} n_{i\tau'\downarrow}$$

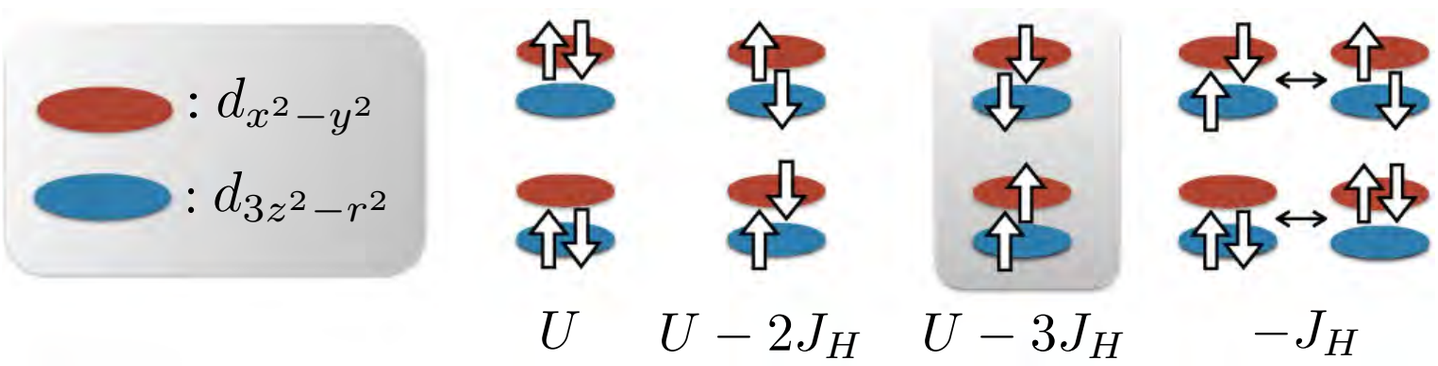
$$+ (U - 3J_H) \sum_{i,\tau<\tau',\sigma} n_{i\tau\sigma} n_{i\tau'\sigma}$$

$$- J_H \sum_{i,\tau \neq \tau'} (d^{+}_{i\tau\uparrow} d_{i\tau\downarrow}\, d^{+}_{i\tau'\downarrow} d_{i\tau'\uparrow} - d^{+}_{i\tau\uparrow} d^{+}_{i\tau\downarrow}\, d_{i\tau'\downarrow} d_{i\tau'\uparrow})$$

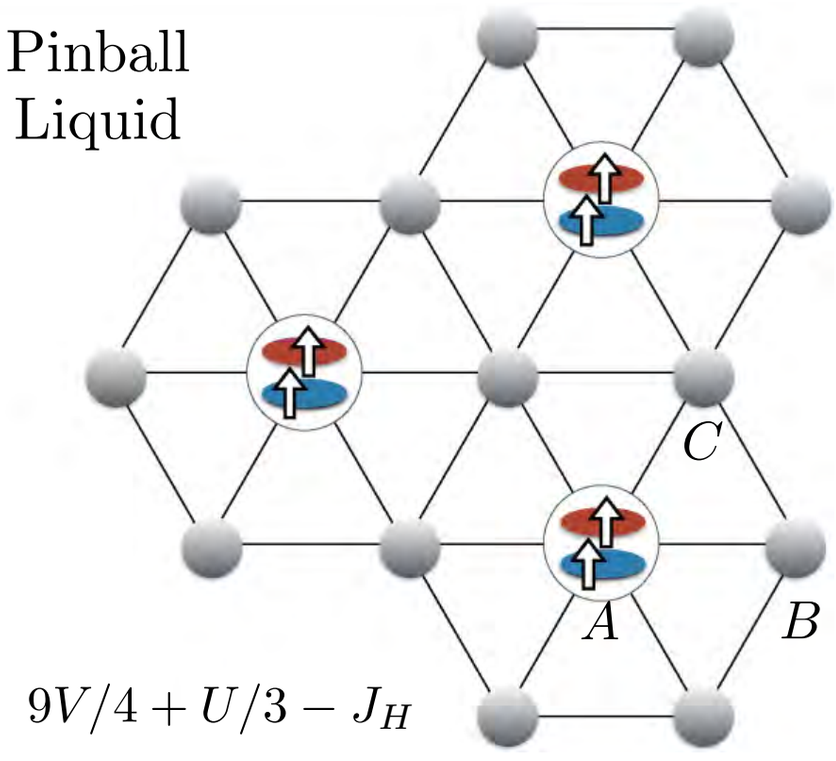
2 orbitals: Extended Hubbard Model + Hund

[Ralko , Merino, Fratini, PRB'15;
Février, Fratini, Ralko, PRB'15]

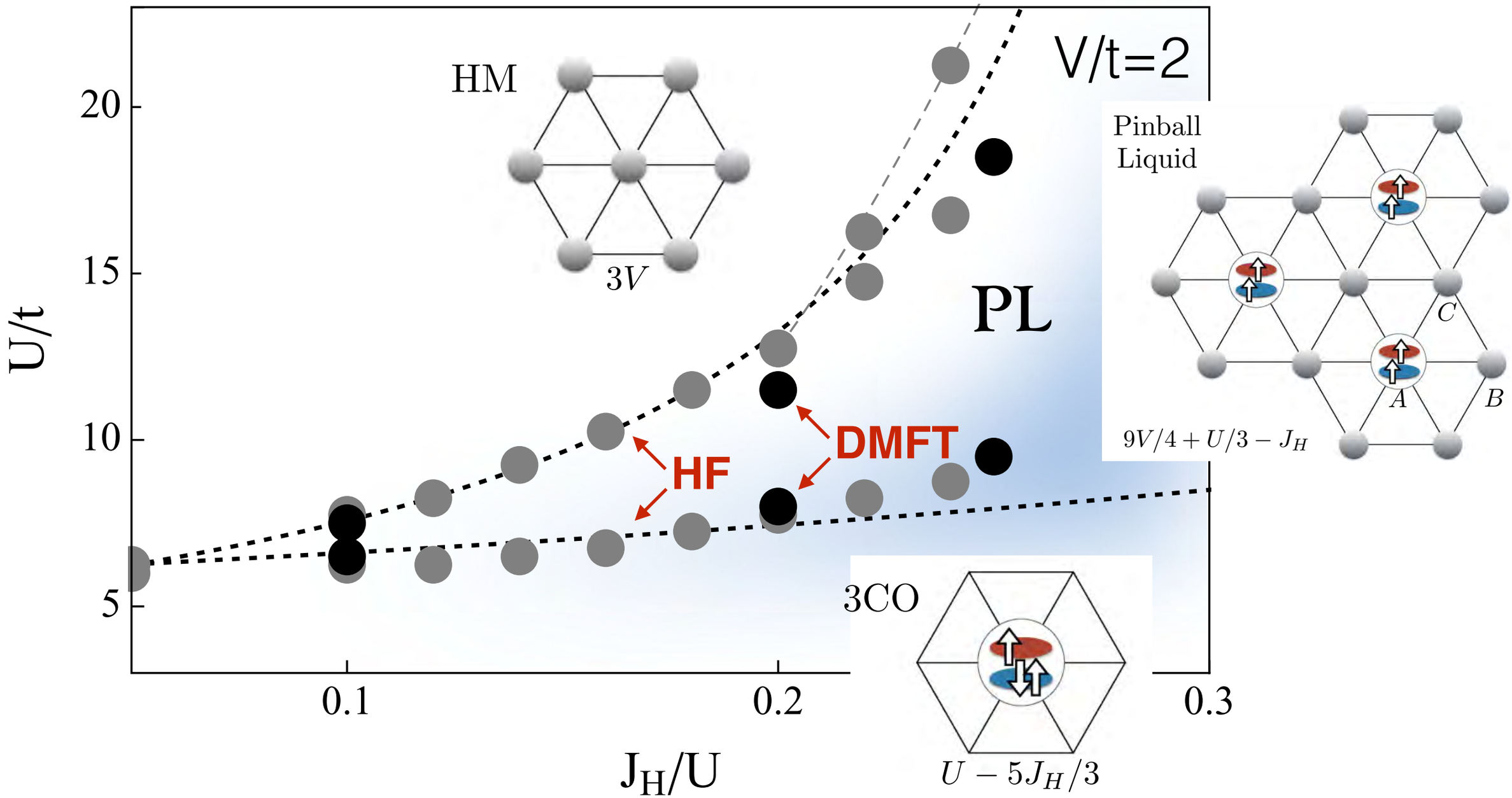
$$H = -t \sum_{\langle ij \rangle} \sum_{\tau,\sigma} (d^{\dagger}_{i,\tau\sigma} d_{j,\tau\sigma} + \text{H.c.}) + H_{\text{V}} + H_{\text{Hund}}$$



- Hund exchange is maximum on doubly occupied sites with aligned spins
- adds to Coulomb V and favors pinball liquid with large (spin 1) moments

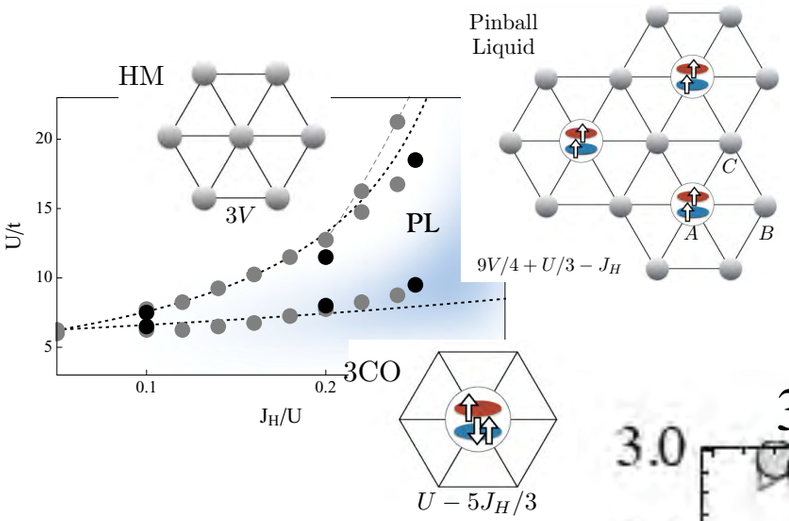


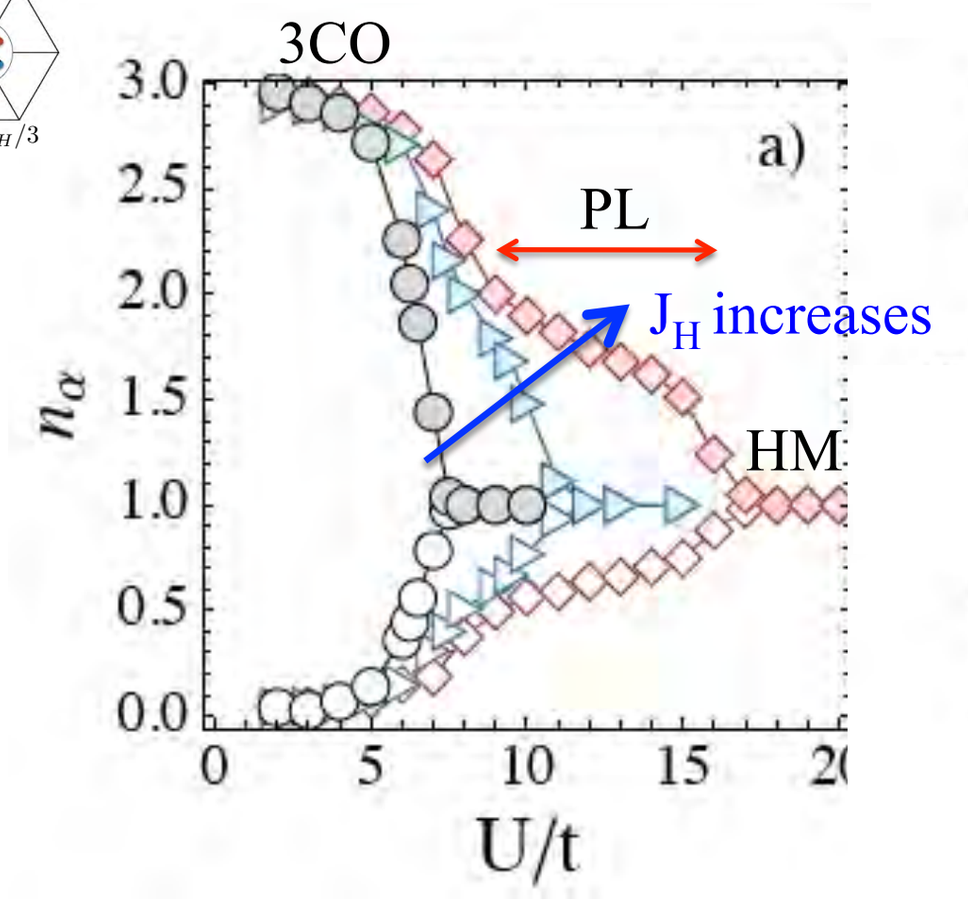
Phase diagram (HF + DMFT)



- pinball liquid stabilized by Hund coupling
- realistic range of microscopic parameters

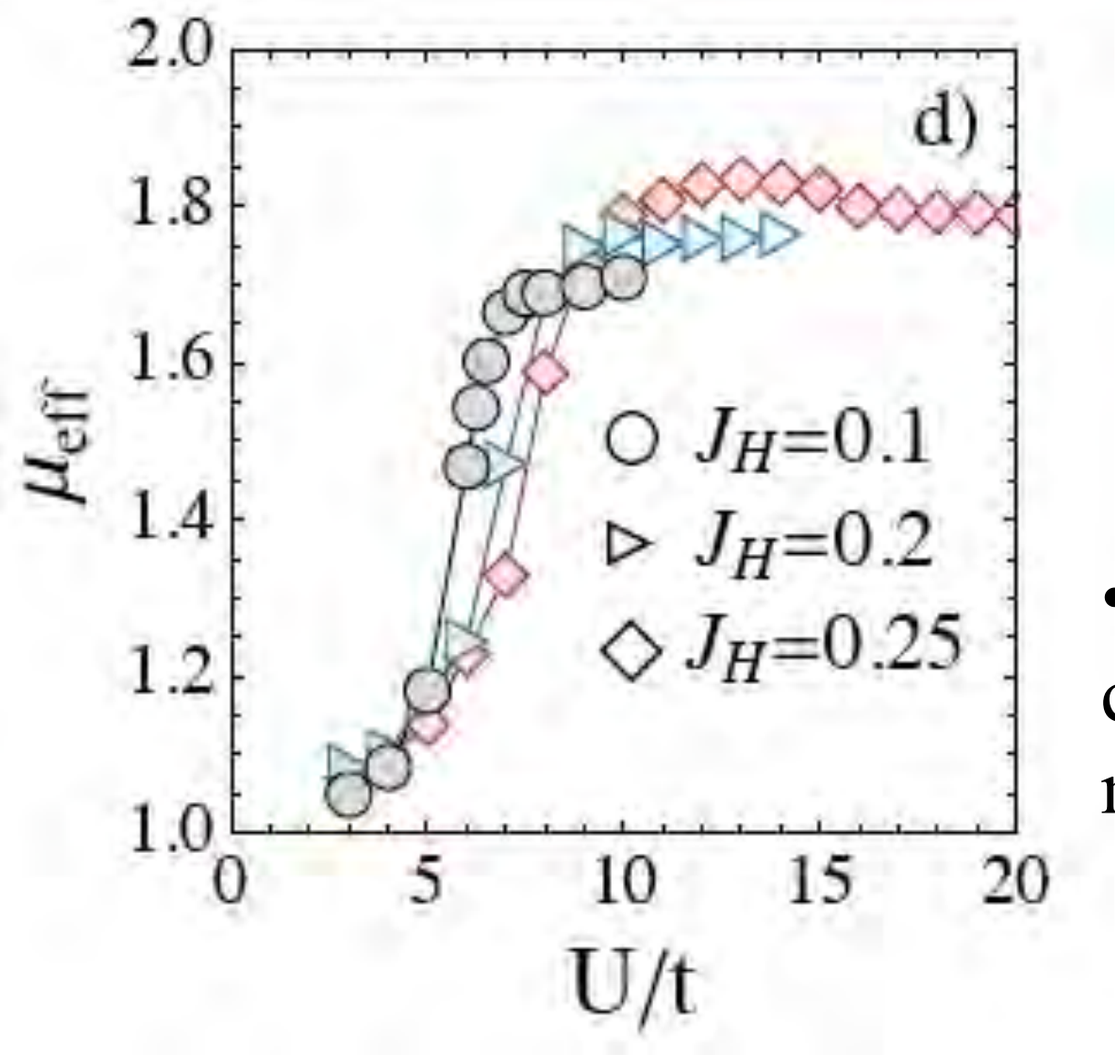
quasiparticle renormalization (DMFT)

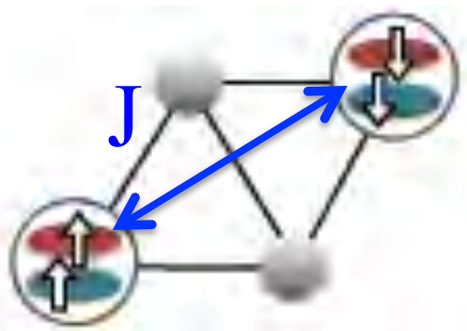




- Hund exchange stabilizes the pinball liquid and induces "Mottness" on the charge rich sublattice
- pinball liquid is strongly correlated, consistent with experiments

magnetic moments and ordering (DMFT)





• Antiferromagnetic J>0 consistent with experimental magnetic order.

• Experimental local magnetic moments: 1.8-1.95μ_B , consistent with DMFT for U/t>10.

Conclusions

- The interplay of **Coulomb induced charge ordering** and **geometric frustration** on the triangular lattice leads to many interesting phenomena

- $AgNiO_2$ contains rich and poorly understood physics: charge ordering, large magnetic moments, renormalization effects, “bad” metallic behavior. It is an experimental realization of a multiband **pinball liquid.**

- further confirmations?
  theory (T*) / experiment (optical conductivity?)