$GaTa_4Se_8$: a new prototype of 3D Mott-Hubbard physics

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Mott transition

A correlation driven metal-insulator transition

- Competition between Coulomb potential and kinetic energies
- Proposed by Mott following a suggestion by Peierls (Proc. Phys. Soc. London, Ser. **A49**, 72, 1937):

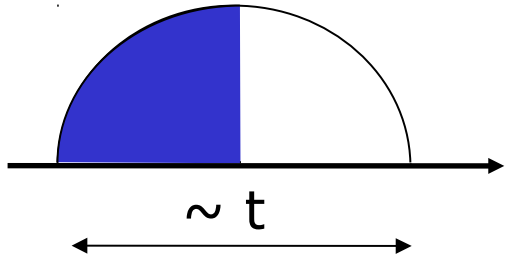
"it is quite possible that the electrostatic interaction between the electrons prevents them from moving at all. At low temperatures the majority of the electrons are in their proper places in the ions. The minority which have happened to cross the potential barrier find therefore all the other atoms occupied, and in order to get through the lattice have to spend a long time in ions already occupied by other electrons. This needs a considerable addition of energy and so is extremely improbable at low temperatures"

Hubbard model

The Hubbard model is a minimal model for the metal – insulator transition:

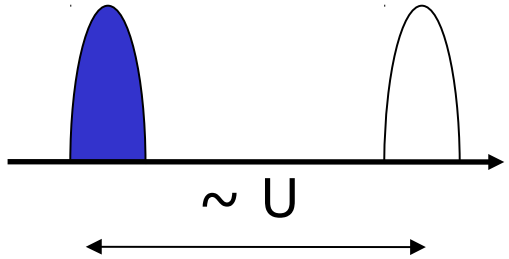
$$H = -\sum_{\langle ij \rangle, \sigma} t_{ij} \left(c^{\dagger}_{i\sigma} c_{j\sigma} + c^{\dagger}_{j\sigma} c_{i\sigma} \right) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

U = 0



metal

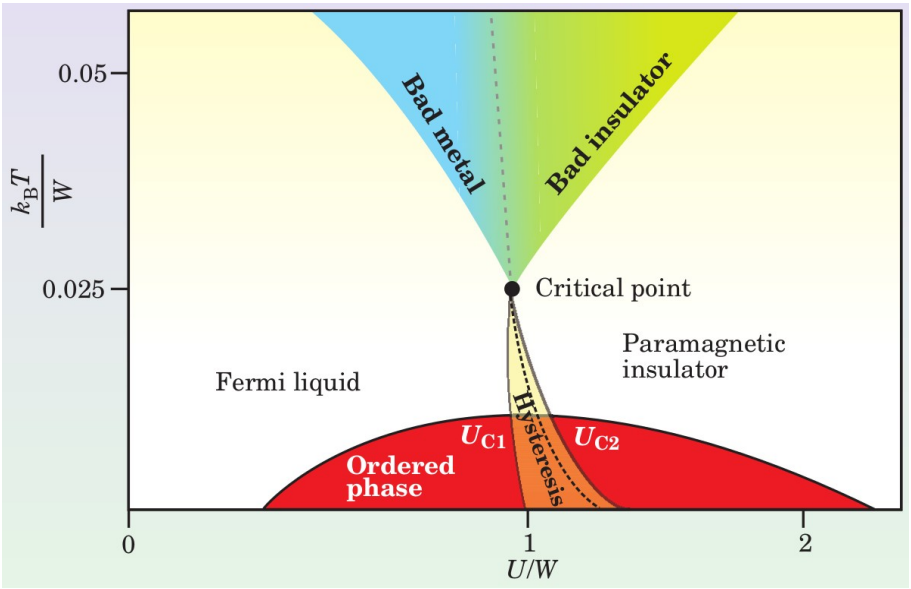
t = 0



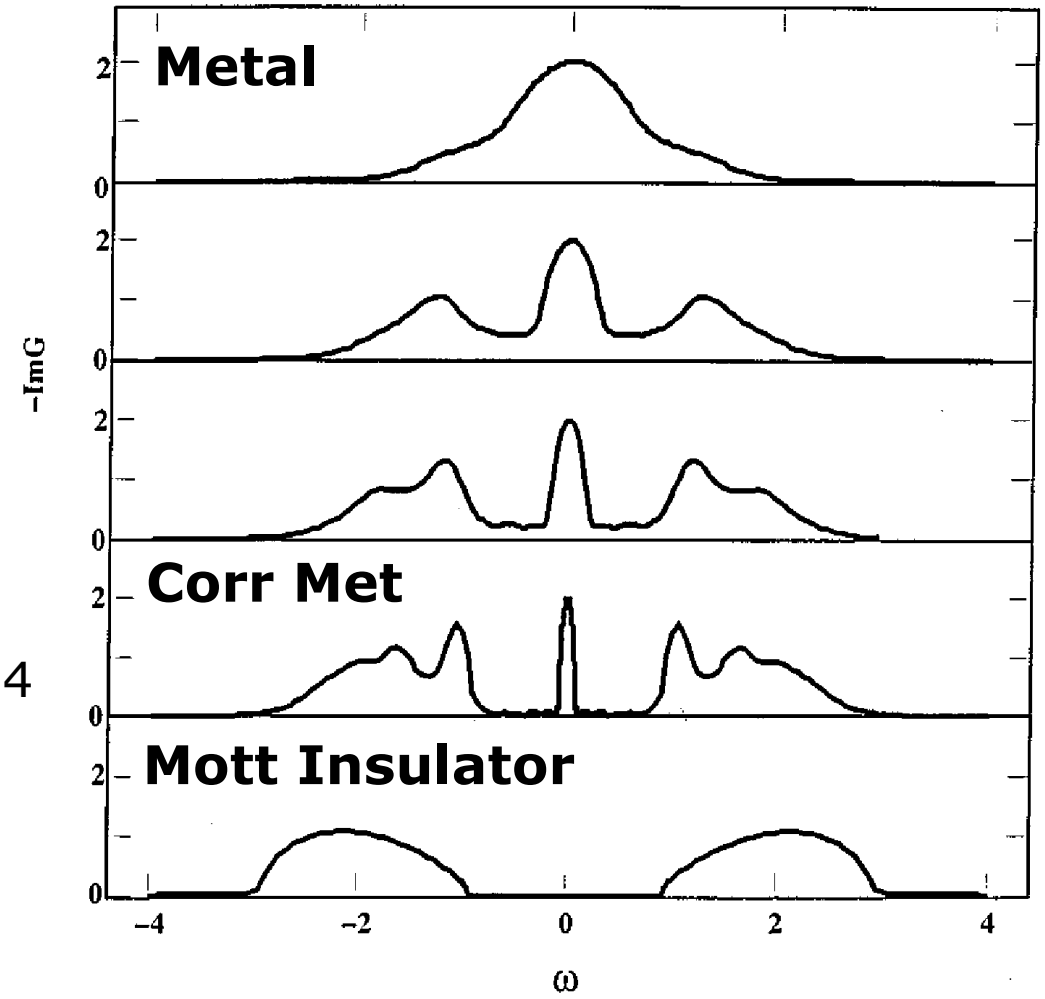
insulator

DMFT of the Mott - Hubbard transition

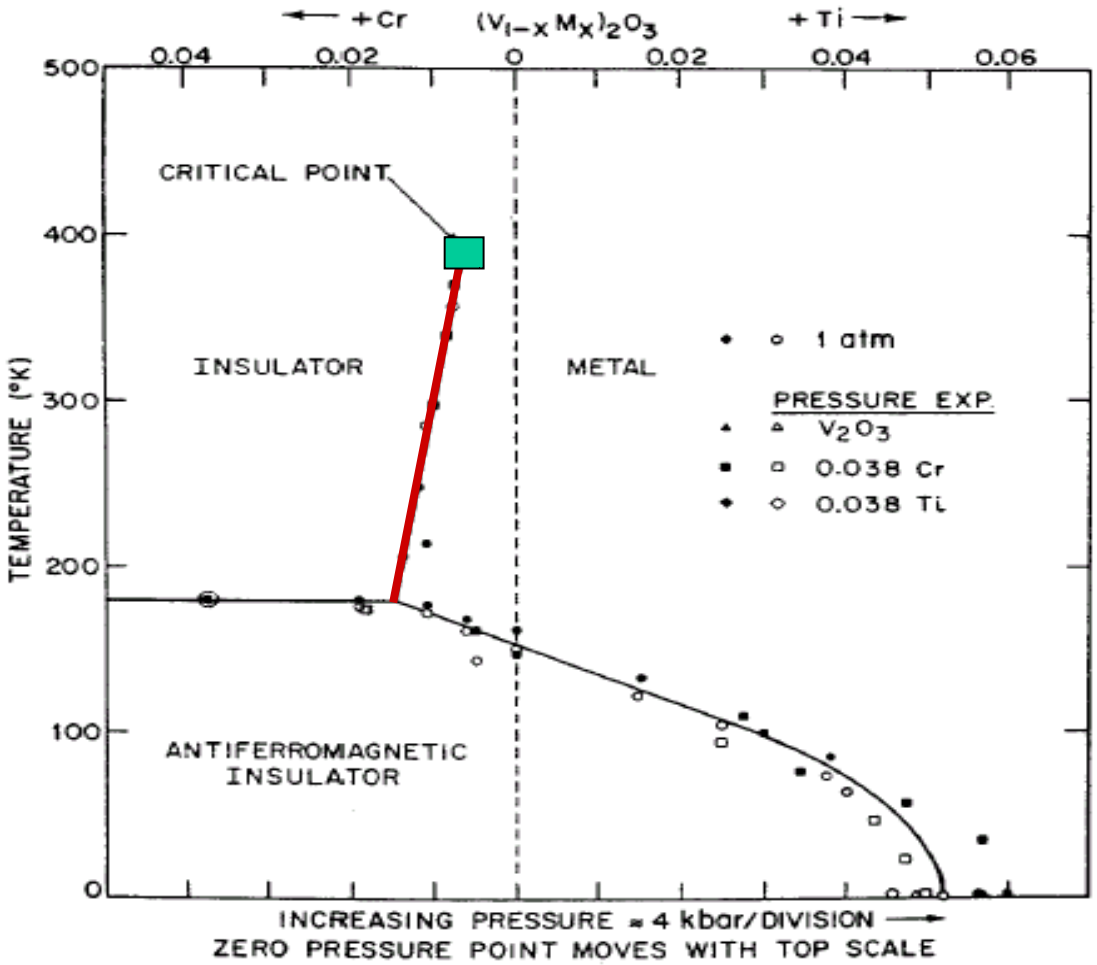
XY Zhang, M Rozenberg, G Kotliar, PRL '92



G Kotliar and D Vollhardt, Phys. Today '04



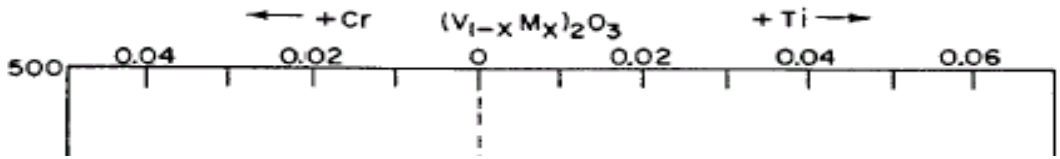
The classic example: Mott transition in V_2O_3



pressure or chemical substitution

McWhan et al., PRB '71 '73

The classic example: Mott transition in V_2O_3



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Mott–Hubbard transition in V_2O_3 revisited

Feature Article

P. Hansmann[1,2], A. Toschi[1], G. Sangiovanni[1,3], T. Saha-Dasgupta[4], S. Lupi[5], M. Marsi[6], and K. Held[*,1]

The isostructural metal-insulator transition in Cr-doped V_2O_3 is the textbook example of a Mott–Hubbard transition between a paramagnetic metal (PM) and a paramagnetic insulator. We review recent theoretical calculations as well as experimental findings which shed new light on this famous transition. In particular, the old paradigm of a doping-pressure equivalence does not hold, and there is a microscale phase separation for Cr-doped V_2O_3.

ZERO PRESSURE POINT MOVES WITH TOP SCALE

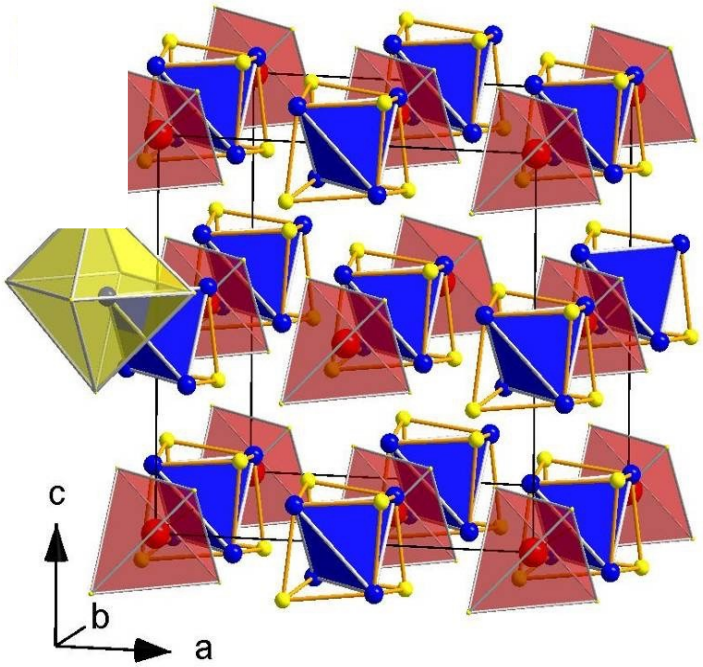
pressure or chemical substitution

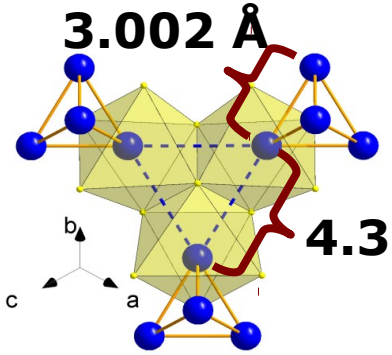
McWhan et al PRB '71 '73

AM_4X_8 family:tailor-made 3D Mott systems?

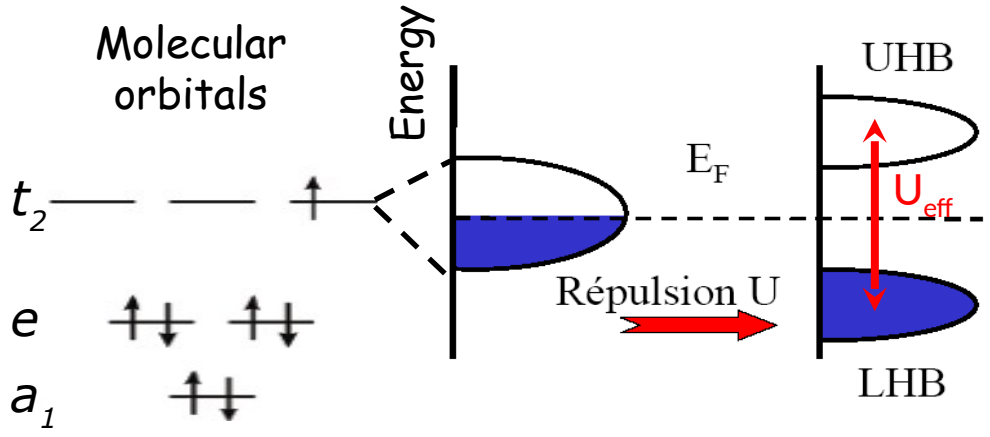
A = Ga, Ge
M = V, Nb, Ta
X = S, Se

$F\bar{4}3m$





Tetrahedral Clusters

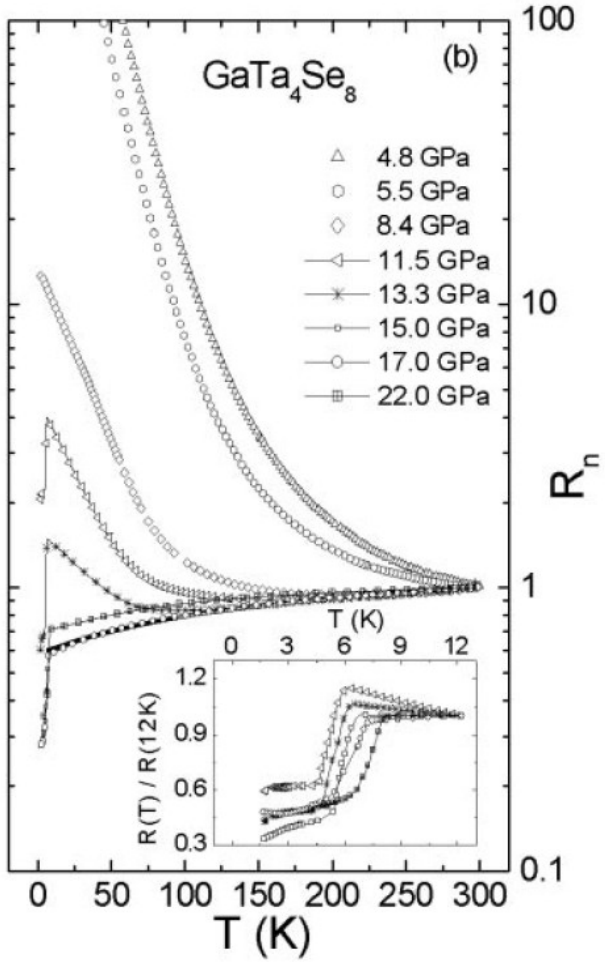


Gap ~ 0.70 eV

Pocha, R. et al., *J. Am. Chem. Soc.* 127, *8732* (2005)

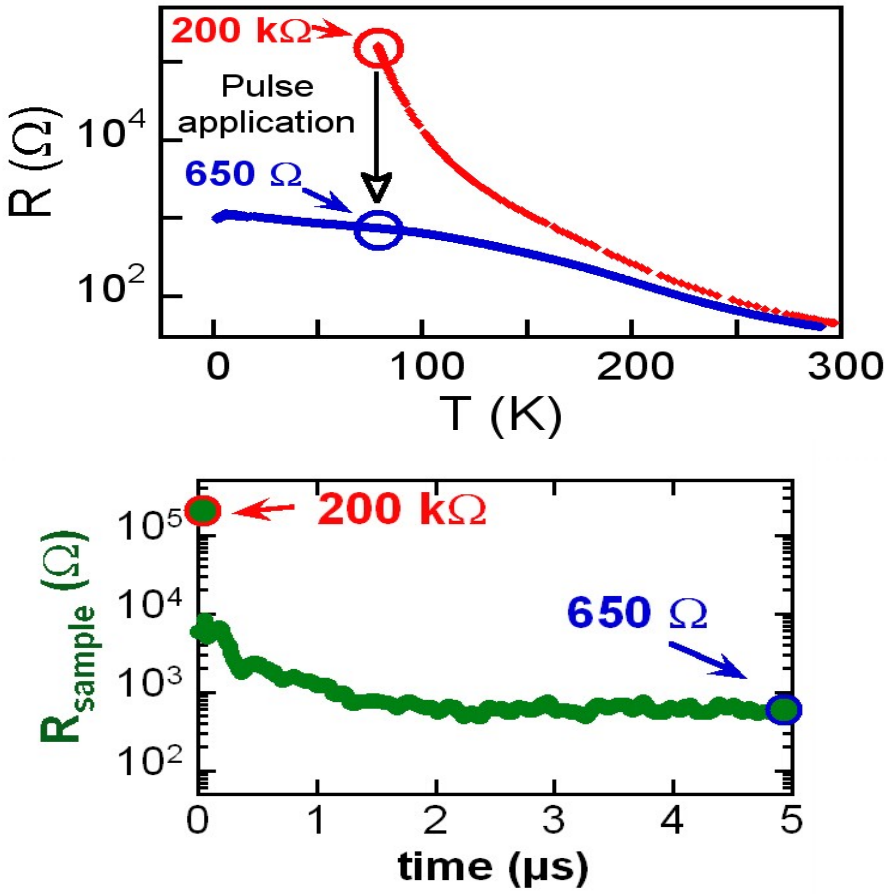
Metal – Insulator transition in $GaTa_4Se_8$

Pressure driven Mott transition



Abd-Elmeguid et al., PRL '04

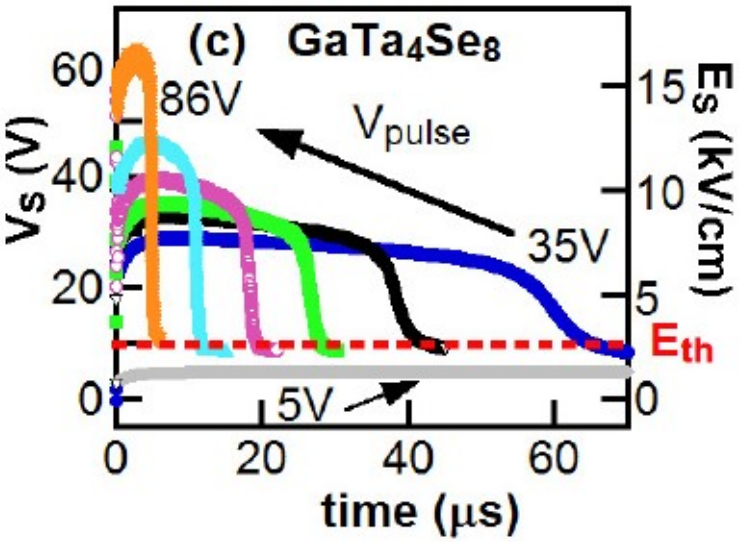
E-field driven Mott transition



Non Volatile Resistive Switching, t <100 ns

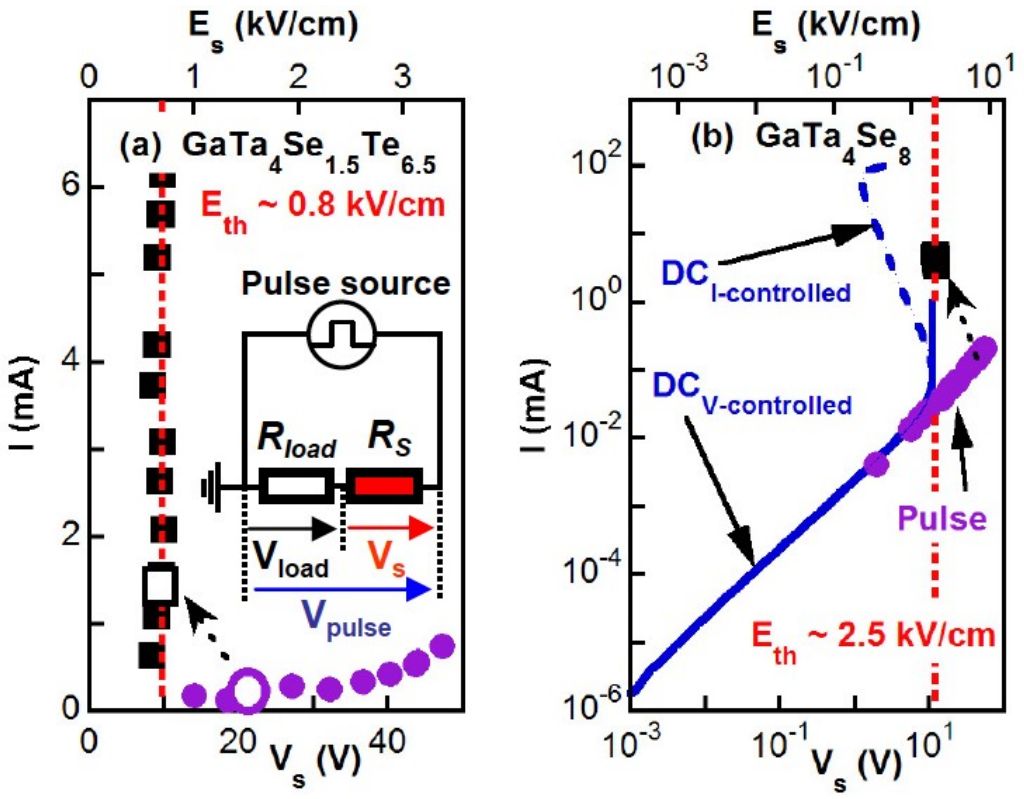
C. Vaju et al., Adv. Mater. '08

Metal – Insulator transition in $GaTa_4Se_8$



E-field threshold

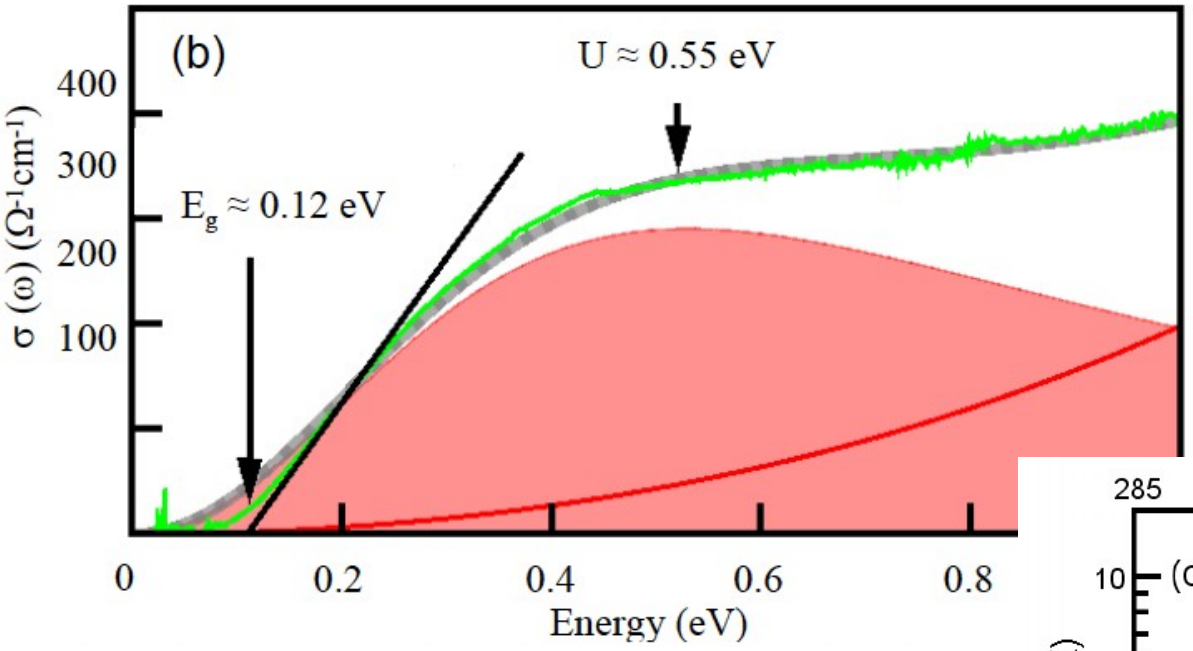
E-field driven Mott transition in GTS



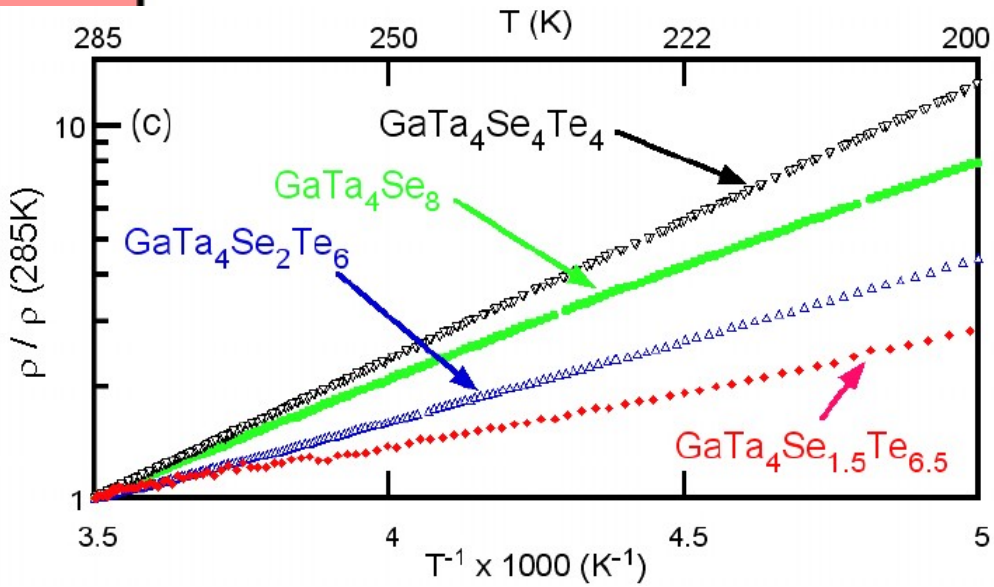
V. Guiot et al. Nat Comm (2013)

Metal – Insulator transition in $GaTa_4Se_8$

Bandwidth (or gap) control by Se – Te substitution



$GaTa_4Se_{8-x}Te_x$



V. Guiot et al., Nat. Comm. **4**, 1722 (2013)

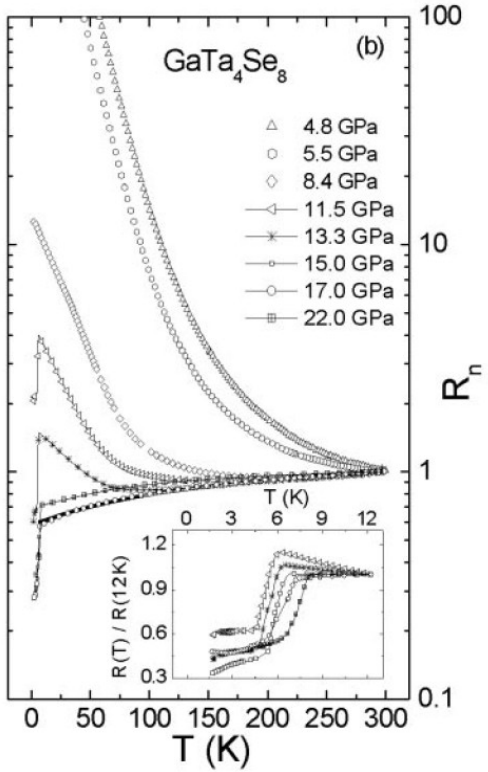
Is $GaTa_4Se_8$ really a Mott-Hubbard system?

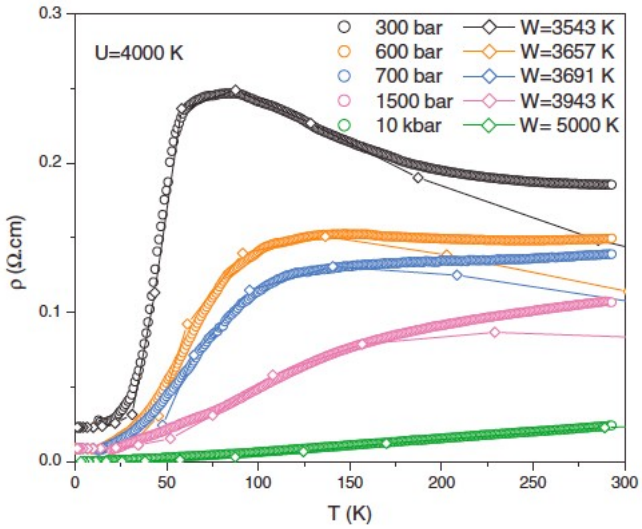
"Ideal system"

- 1 electron per Ta_4 cluster
- 3D fcc lattice
- Paramagnetic
- LDA predicts a metal

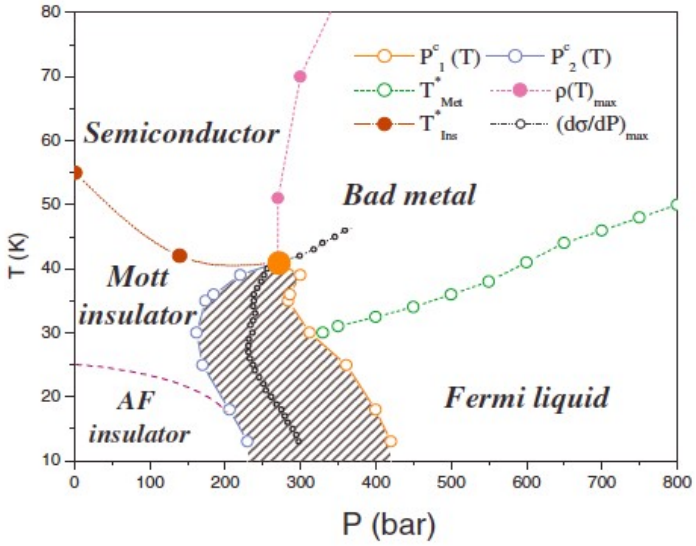
But does not match some DMFT key predictions

- No hysteresis
- ρ(T) does not have non-monotonic behavior





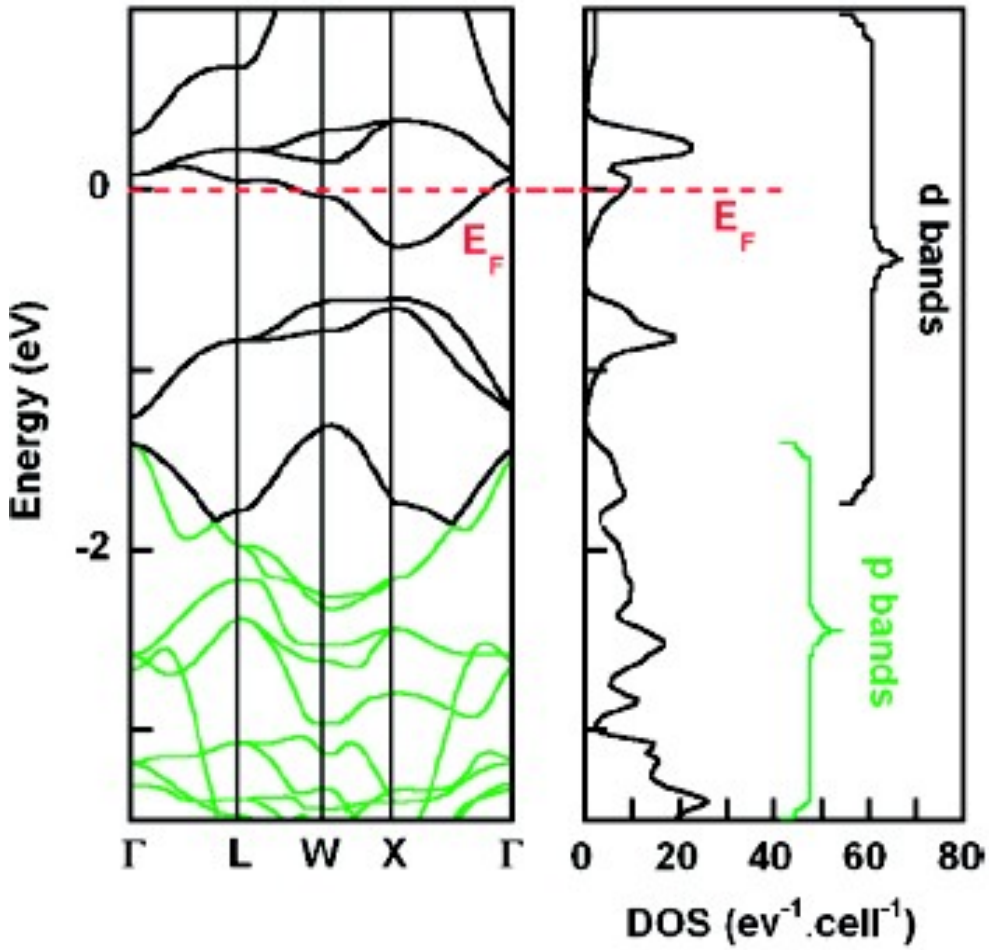
K-organics



Limelette PRL (2003)

Is $GaTa_4Se_8$ really a Mott-Hubbard system?

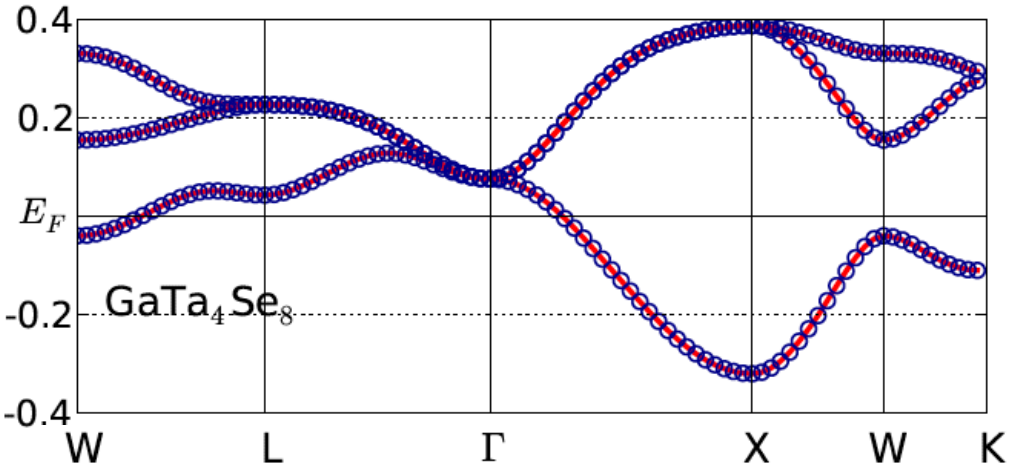
Conduction bands are isolated and have pure Ta character
Nice system for an LDA+DMFT study

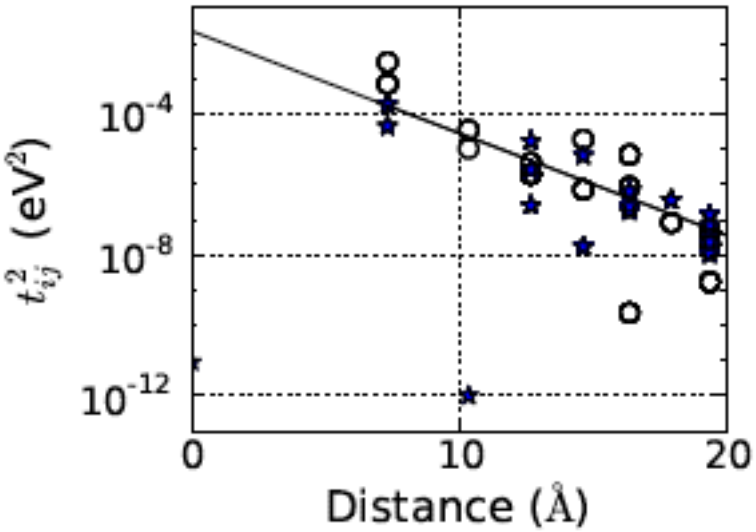


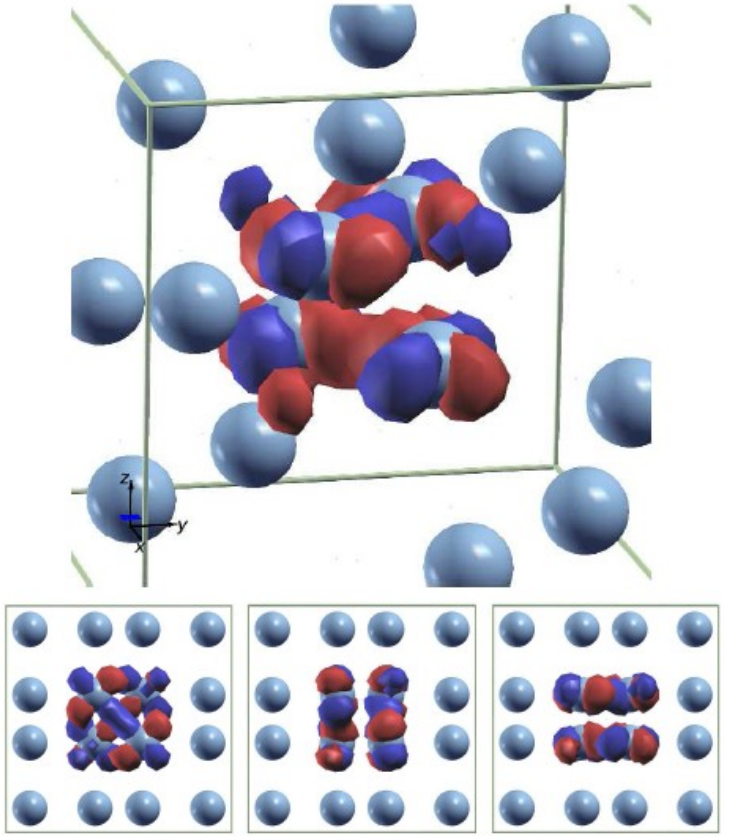
V. Guiot et al., Chem. Matter (2011)

Is $GaTa_4Se_8$ really a Mott-Hubbard system?

Wannier maximally localized molecular orbitals for Ta_4 tetrahedra



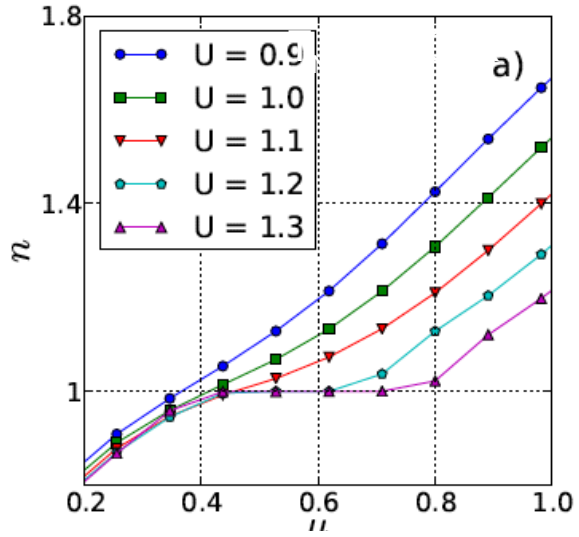




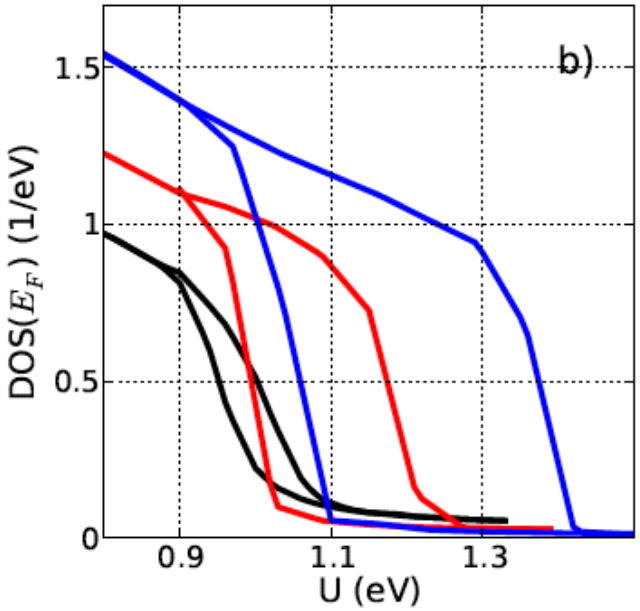
Cubic FCC structure, t_{2g} symmetry

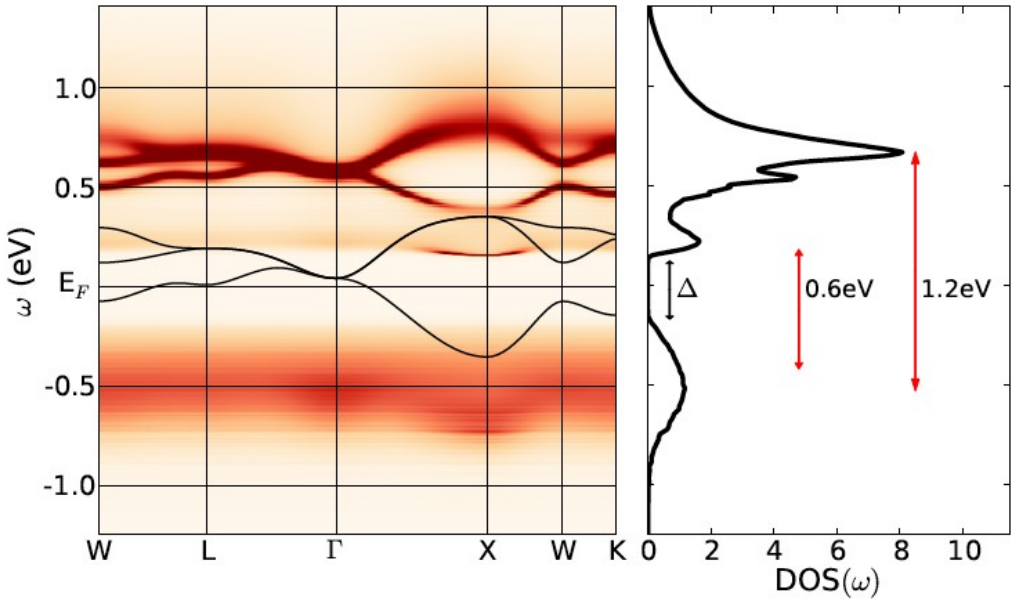
LDA + DMFT

Uc = 1.2 eV

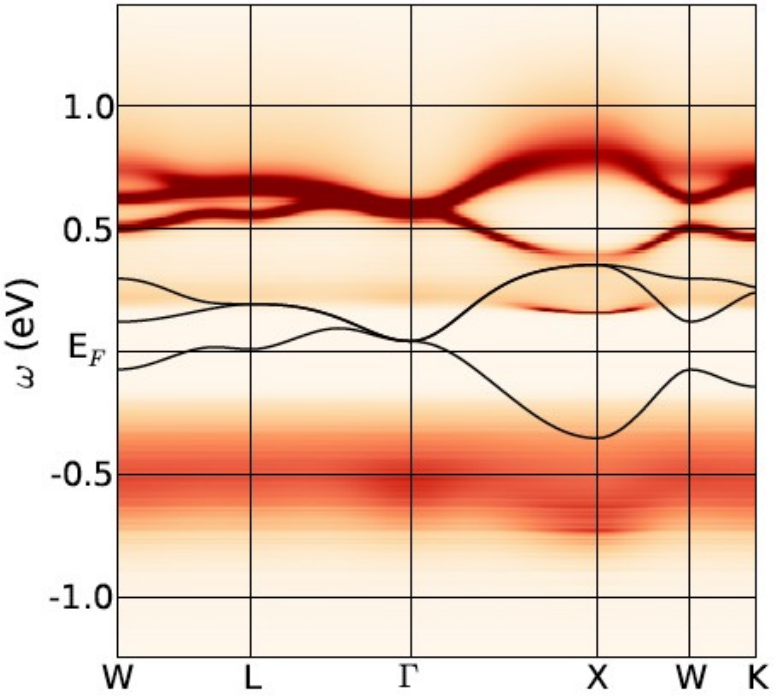


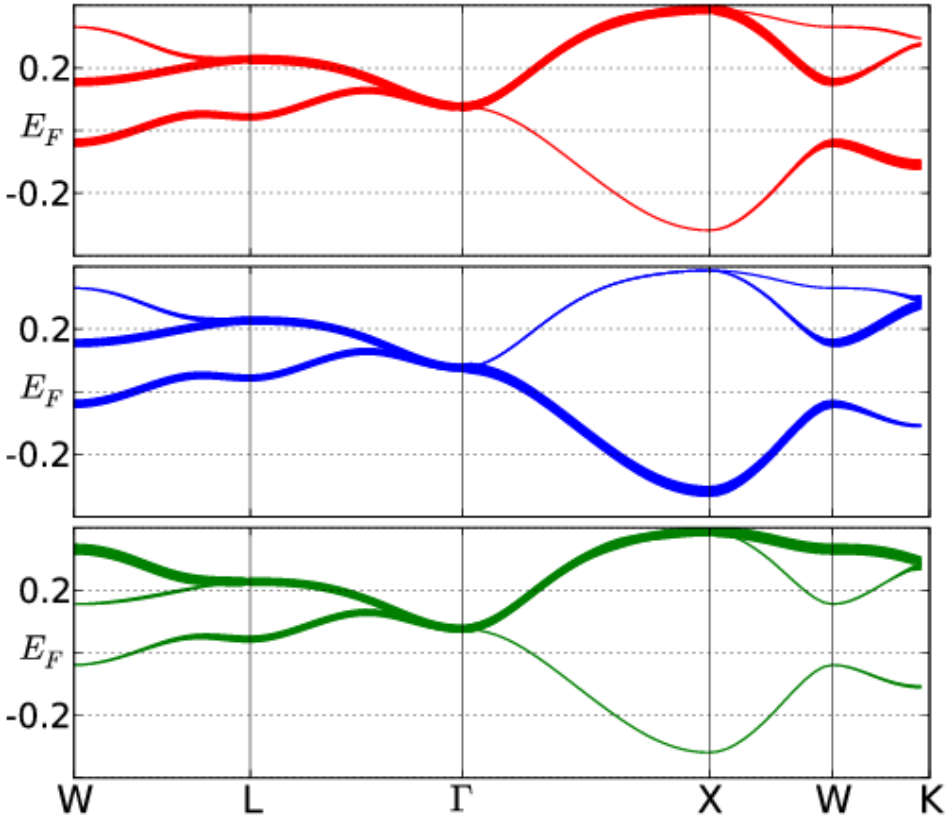
hysteresis





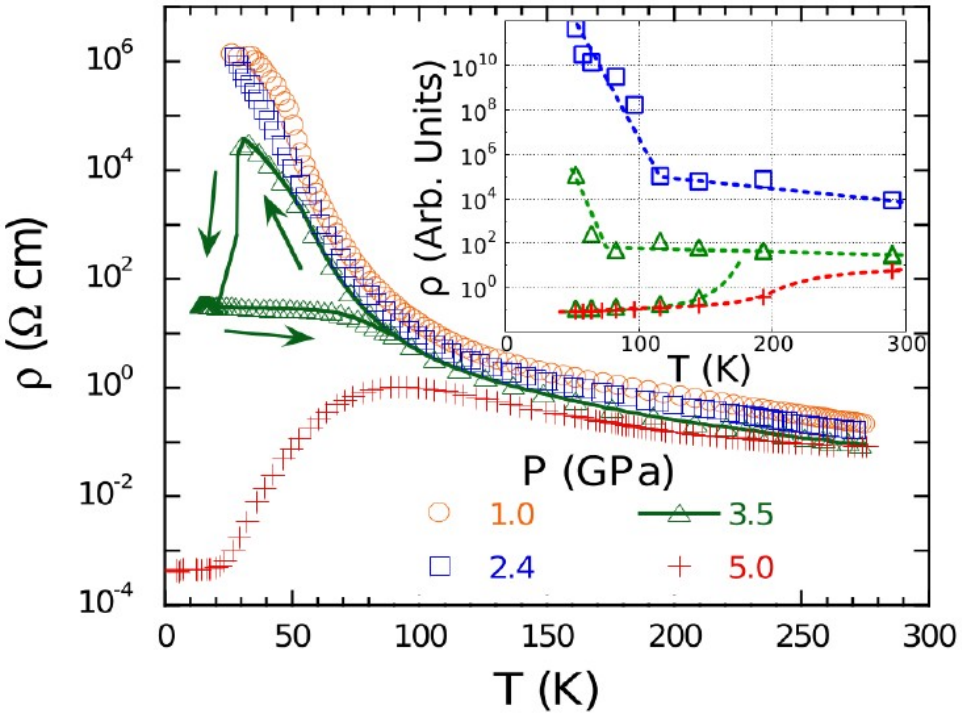
LDA + DMFT

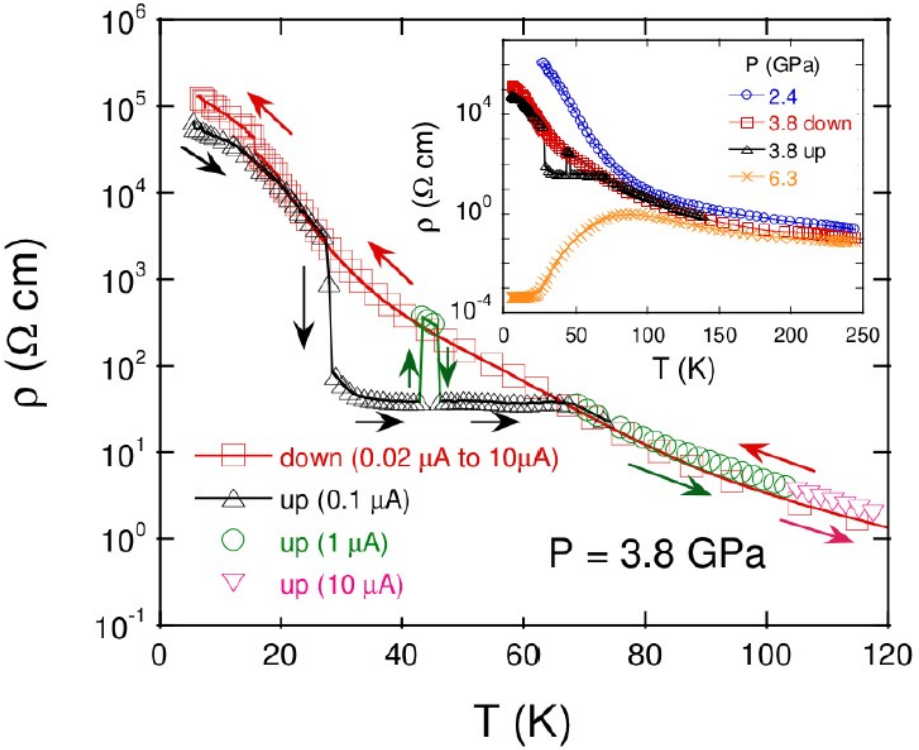




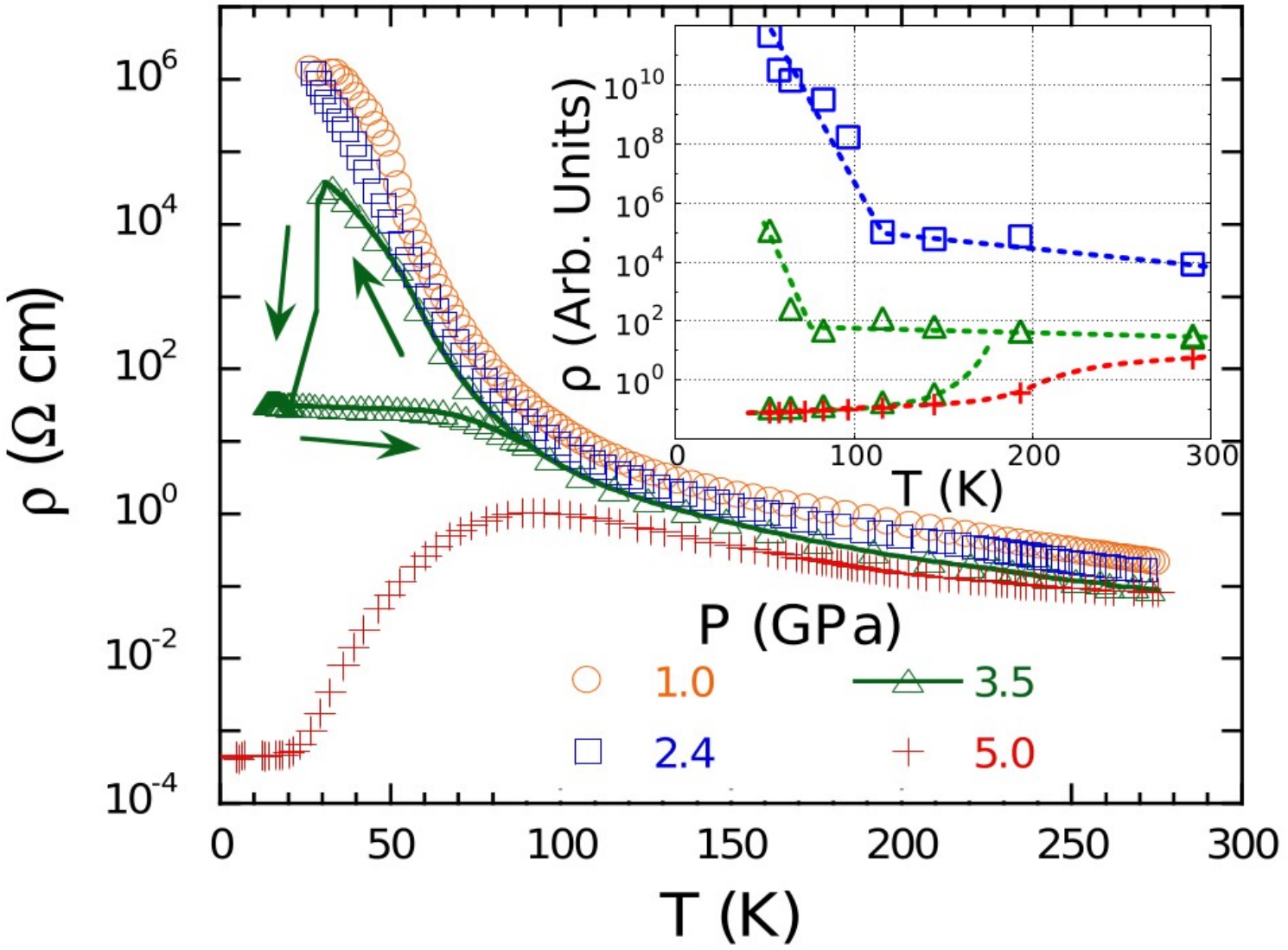
GTS under pressure

Resistivity is non-monotonic in the metal and has hysteresis at the IMT

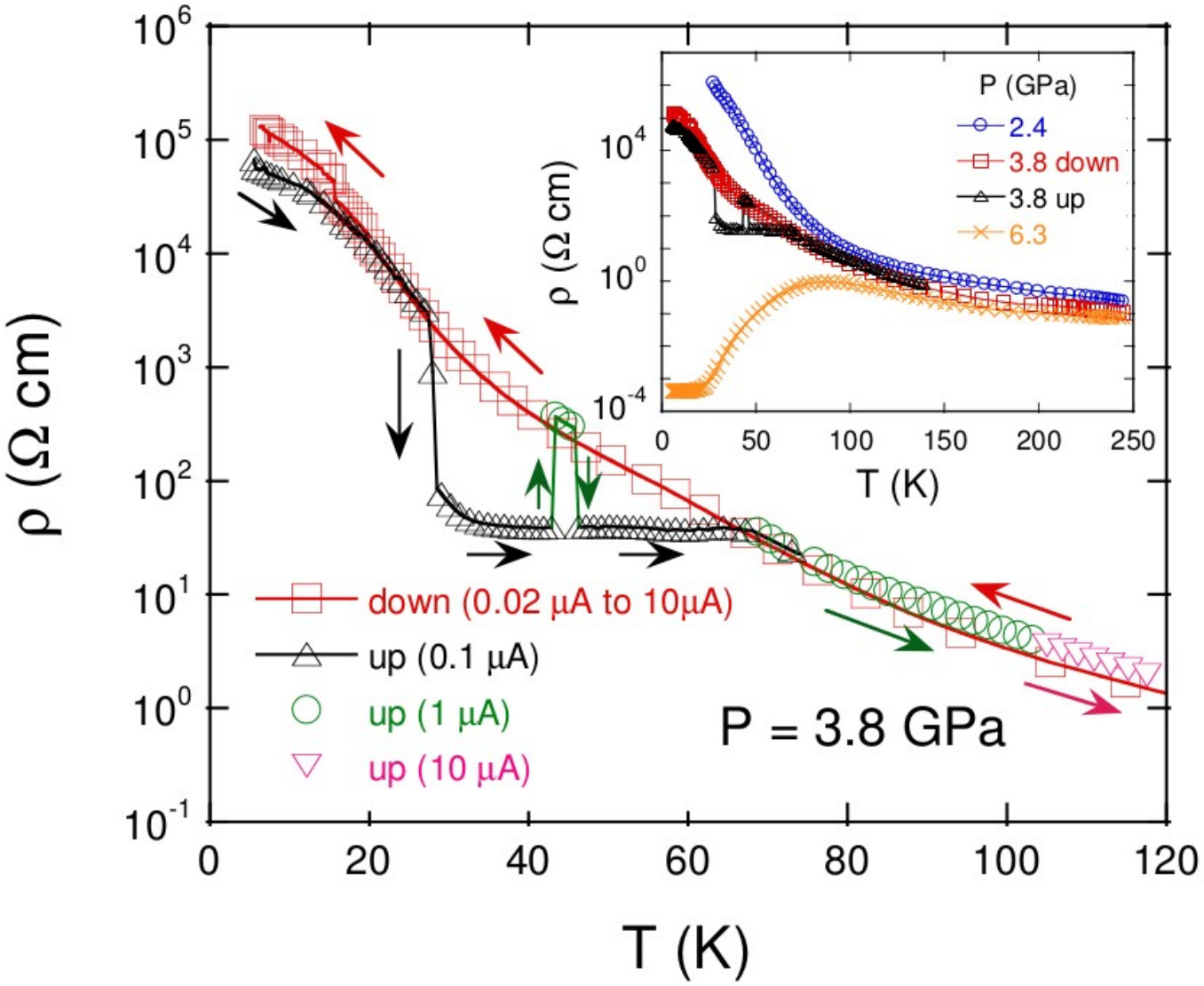




GTS under pressure



GTS under pressure



Summary

- Resistivity experiments under quasi-hydrostatic pressure on single crystals uncover the first order character of the transition
- Resistive bi-stability control at the transition validates the existence of two competing solutions

Thank you!

PRL **113**, 086404 (2014)
EPL **100**, 57004 (2012)