Magnetic Moment Collapse drives Mott transition in MnO

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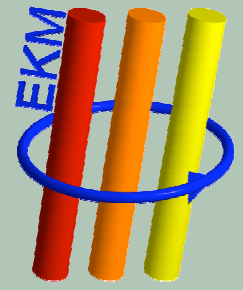
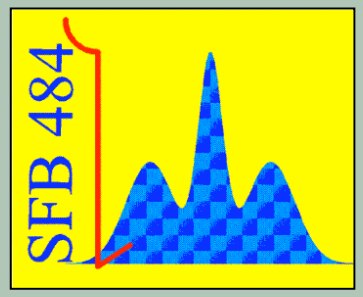
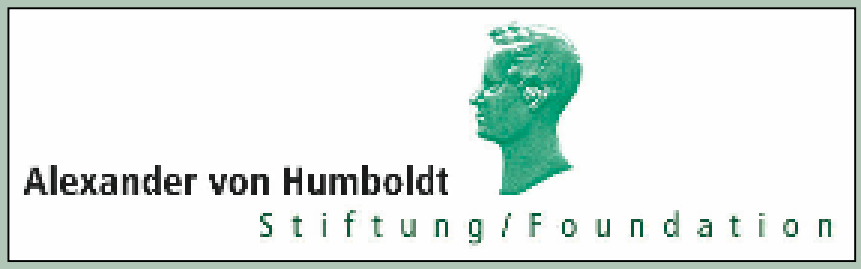






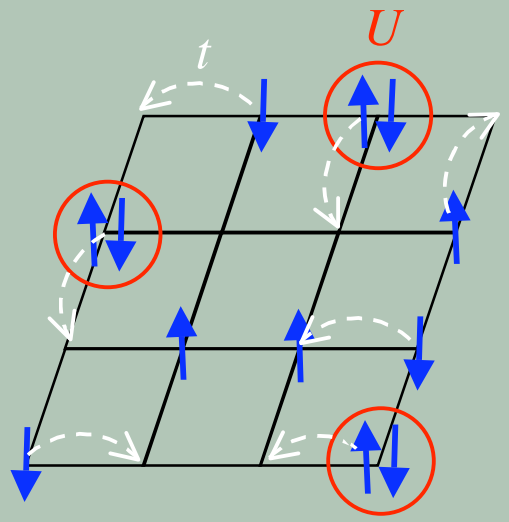
in collaboration with:

V. I. Anisimov, A. V. Lukoyanov, W. E. Pickett, R. T. Scalettar, D. Vollhardt, P. Werner

Outline

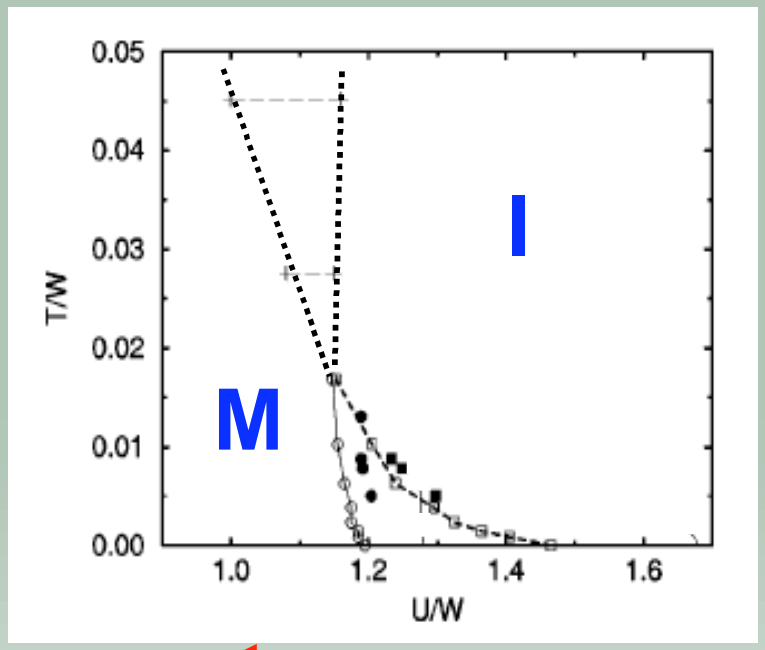
- Introduction
- LDA+DMFT
- NiO - charge transfer insulator
- MnO under pressure - DMFT results
  - moment collapse
  - metal-insulator transition
- Fe_2O_3 role of AFM order (last minute)

1-band Hubbard model

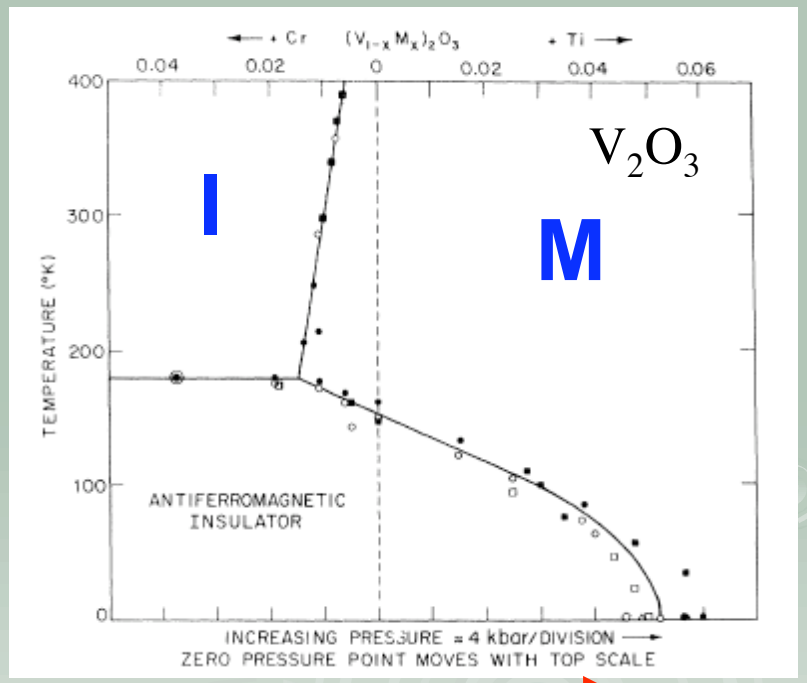


D=∞

control parameters T, W/U



Pressure



Pressure

*Bulla et al. PRB **64**, 045103 (2001)*

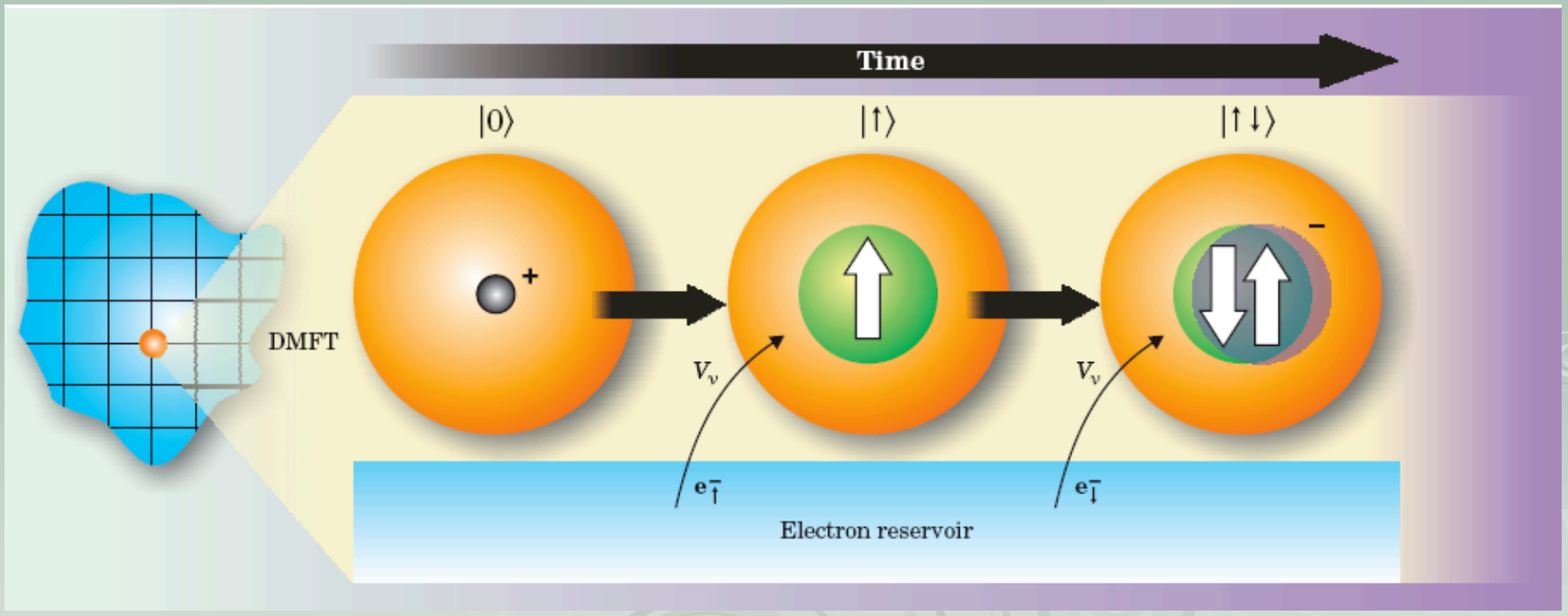
*for review A. Georges et al. RMP **68**, 13 (1996)*

*McWhan et al. PRL **27**, 941 (1971)*

Dynamical mean-field theory & computational tools

Dynamical Mean-Field Theory (DMFT)

- Single out an atom from the lattice
- Replace the rest of the lattice by an effective medium
- Time resolved treatment of local electronic interactions
- Reconstruct lattice quantities



A. Georges et al. RMP ***68****, 13 (1996)* ***Physics Today*** (March 2004) Kotliar, Vollhardt

DMFT vs Weiss molecular field

$$\mathrm{H} = t_{ij} c_{i\sigma}^{+} c_{j\sigma} + U n_{i\uparrow} n_{i\downarrow}$$

$$G_{ii}(\tau) = -\left\langle c_i(\tau) c_i^{+}(0) \right\rangle$$

$$\mathrm{H}_{loc} = (\varepsilon_c - \mu) c_\sigma^{+} c_\sigma + U n_\uparrow n_\downarrow + V_\alpha \left(c_\sigma^{+} b_{\alpha\sigma} + H.c. \right) + \varepsilon_\alpha b_{\alpha\sigma}^{+} b_{\alpha\sigma}$$

$$\Rightarrow \Delta(\omega)$$

$$\Delta(\omega): \quad \left(\omega + \mu - \varepsilon_c - \Delta(\omega) - \Sigma(\omega)\right)^{-1} = \sum_{\mathbf{k}} \left(\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma(\omega)\right)^{-1}$$

$$\mathrm{H} = -J_{ij} S_i S_j - h S_i$$

$$m_i = \left\langle S_i \right\rangle$$

$$\mathrm{H}_{loc} = -h_{eff} S$$

$$h_{eff} = zJm + h$$

DMFT implementation

LDA $\Rightarrow$ $H=\Sigma_{\mathbf{k}}H_0(\mathbf{k})+\Sigma_{\mathbf{R}}\mathbf{U}_{ij}n_{\mathbf{R}i}n_{\mathbf{R}j}$

Set of 8x8 matrices: Mn 3d, O 2p orbitals

FP-LMTO, paramagnetic solution

Compute local GF

$G(i\omega_n)$

Construct impurity problem

$\mathcal{G}(i\omega_n)$, $\mathbf{U}_{ij}$

Solve impurity problem

$\Sigma(i\omega_n)$

Analytic continuation (MaxEnt)

One- and two-particle quantities on real axis

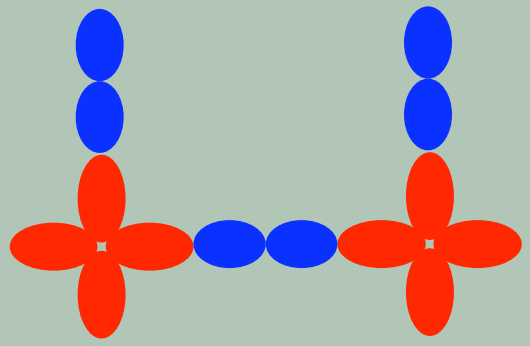
Hirsch-Fye QMC, 45 HS fields
T=1160 K, # of τ-slices 100

DMFT features

- many-body dynamics is local: $\Sigma(\omega)$, $\Gamma(\nu,\nu',\omega)$
- only single-particle quantities (self-energy $\Sigma(\omega)$) need for self-consistency
- DMFT is non-perturbative approach
  exact in $D=\infty$
- DMFT contains T-dependence beyond Fermi-Dirac due to local Kondo physics

NiO: charge-transfer insulator

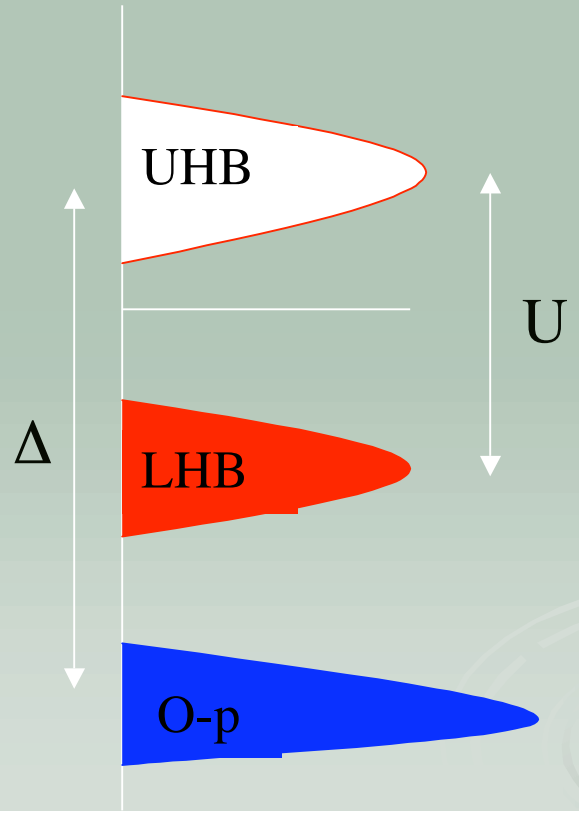
Transition metal oxides: ZSA scheme



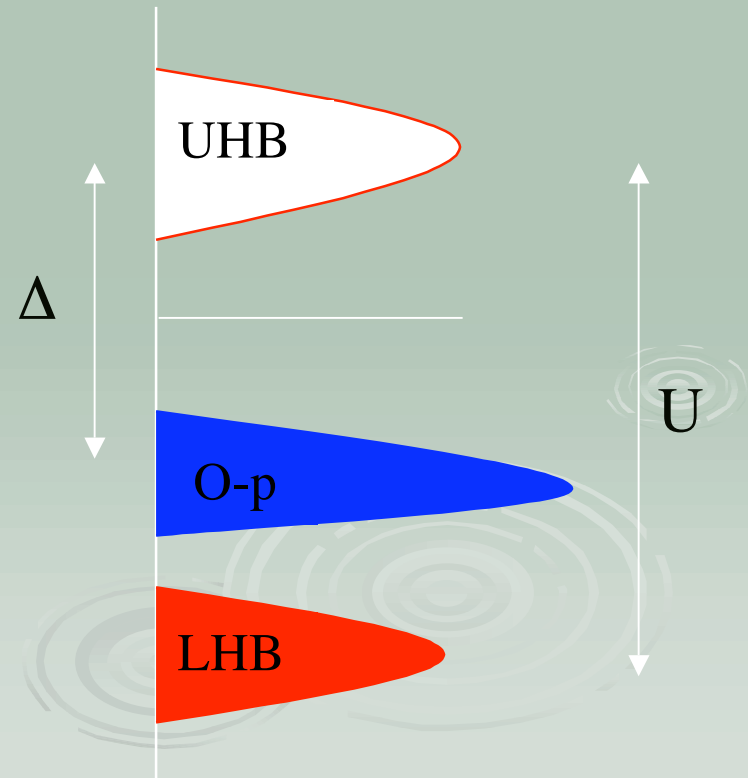
O - 2p

Transition metal - 3d

Mott-Hubbard type (Ti-O, V-O)

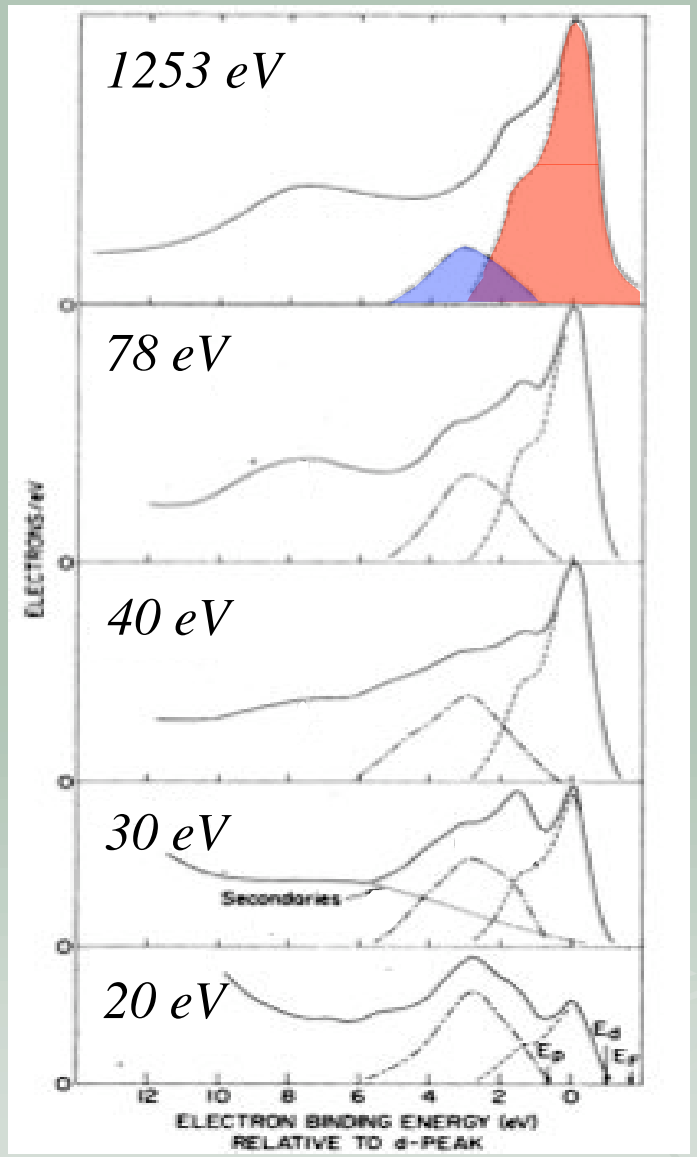


charge-transfer type (Ni-O,Cu-O)



NiO

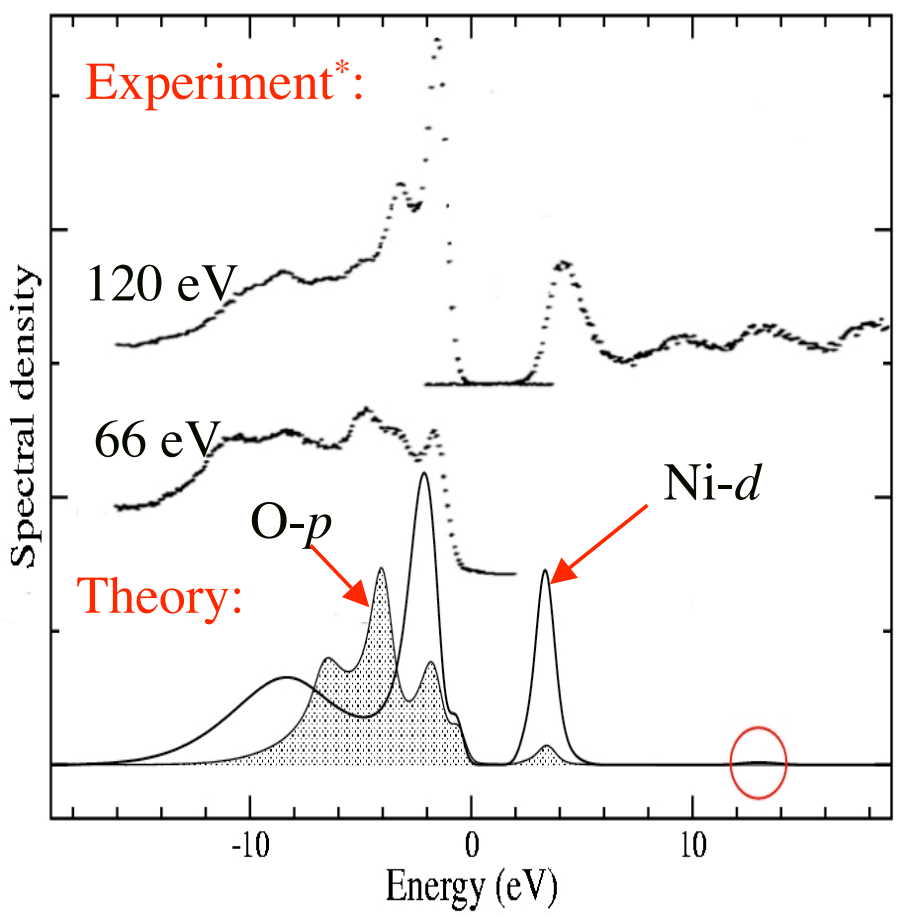
*Eastman & Freeouf, PRL **34**, 395 (1974)*:



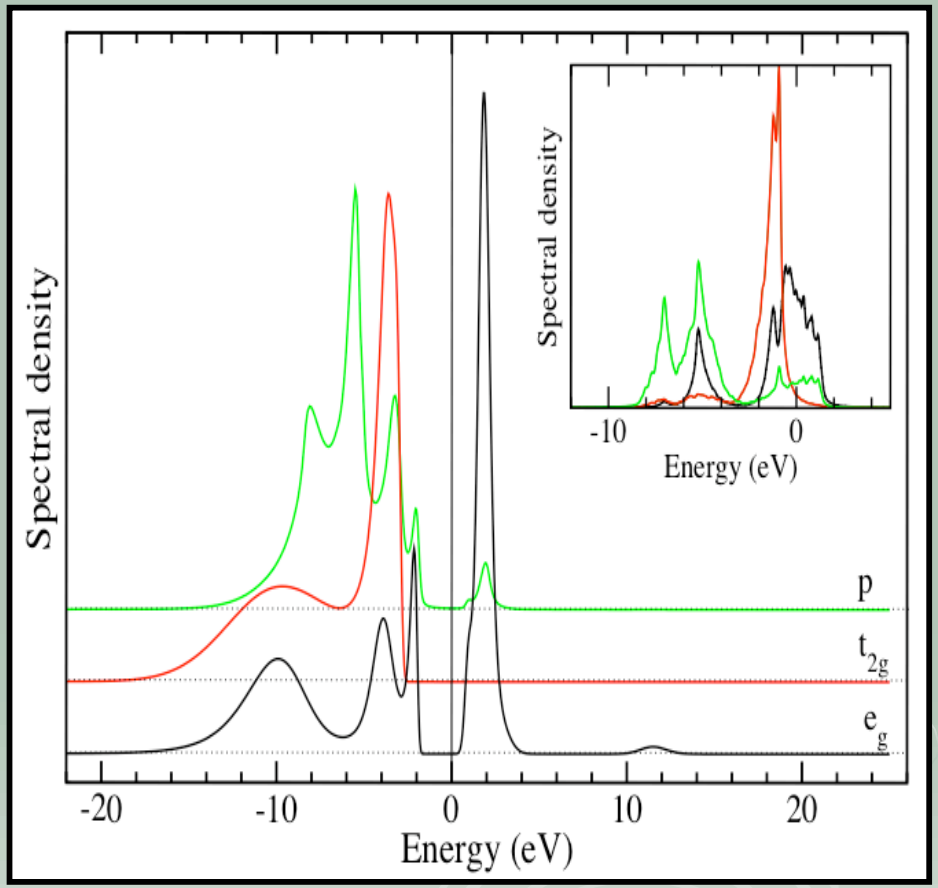
- Is NiO charge-transfer insulator?
- How does hole doping affects the spectrum?
- Role of AFM order?

NiO - DMFT (QMC)

Photoemission spectrum



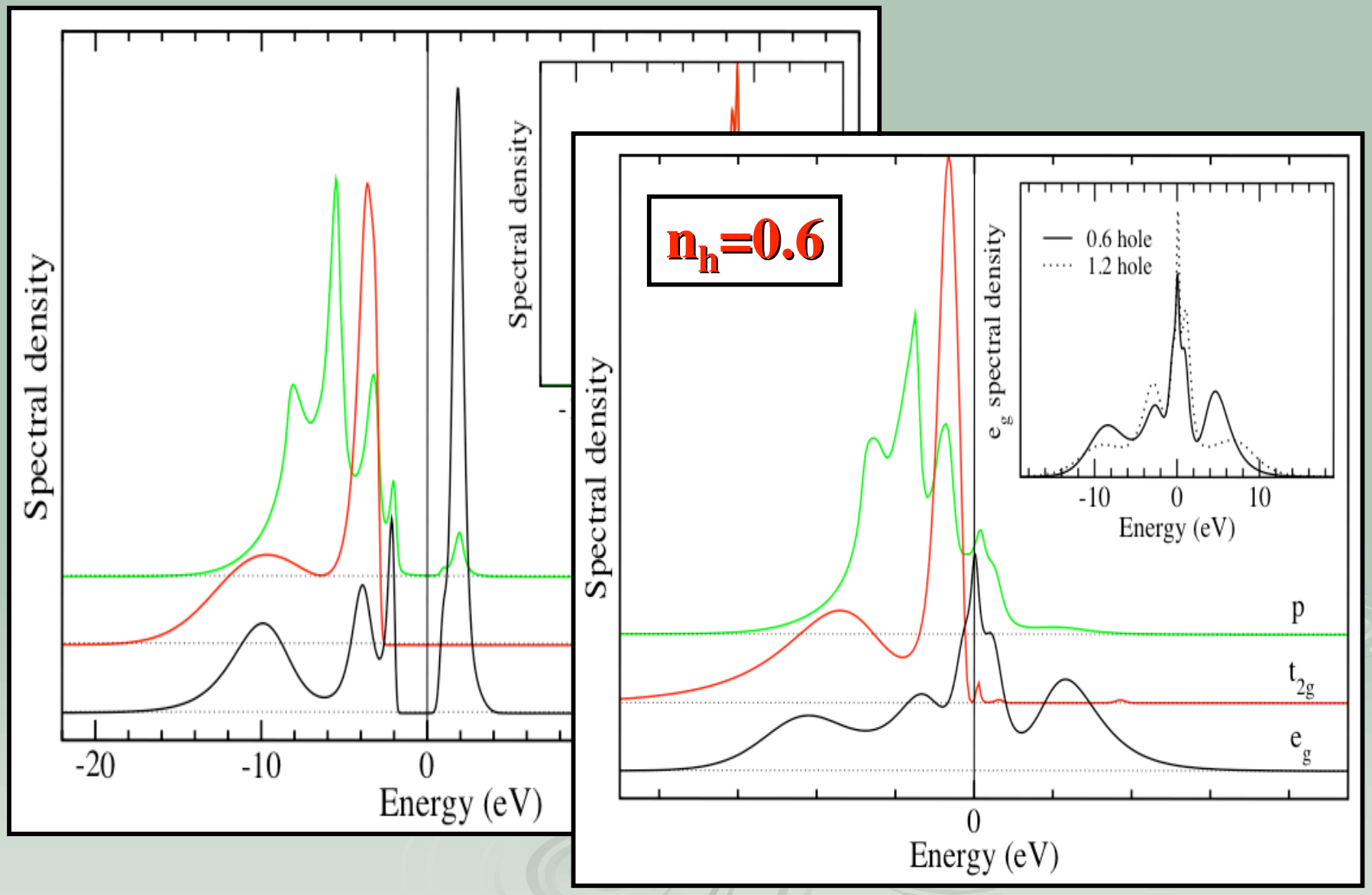
Orbital resolved spectal density



* *Sawatzky & Allen, PRL* ***53****, 2339 (1984)*

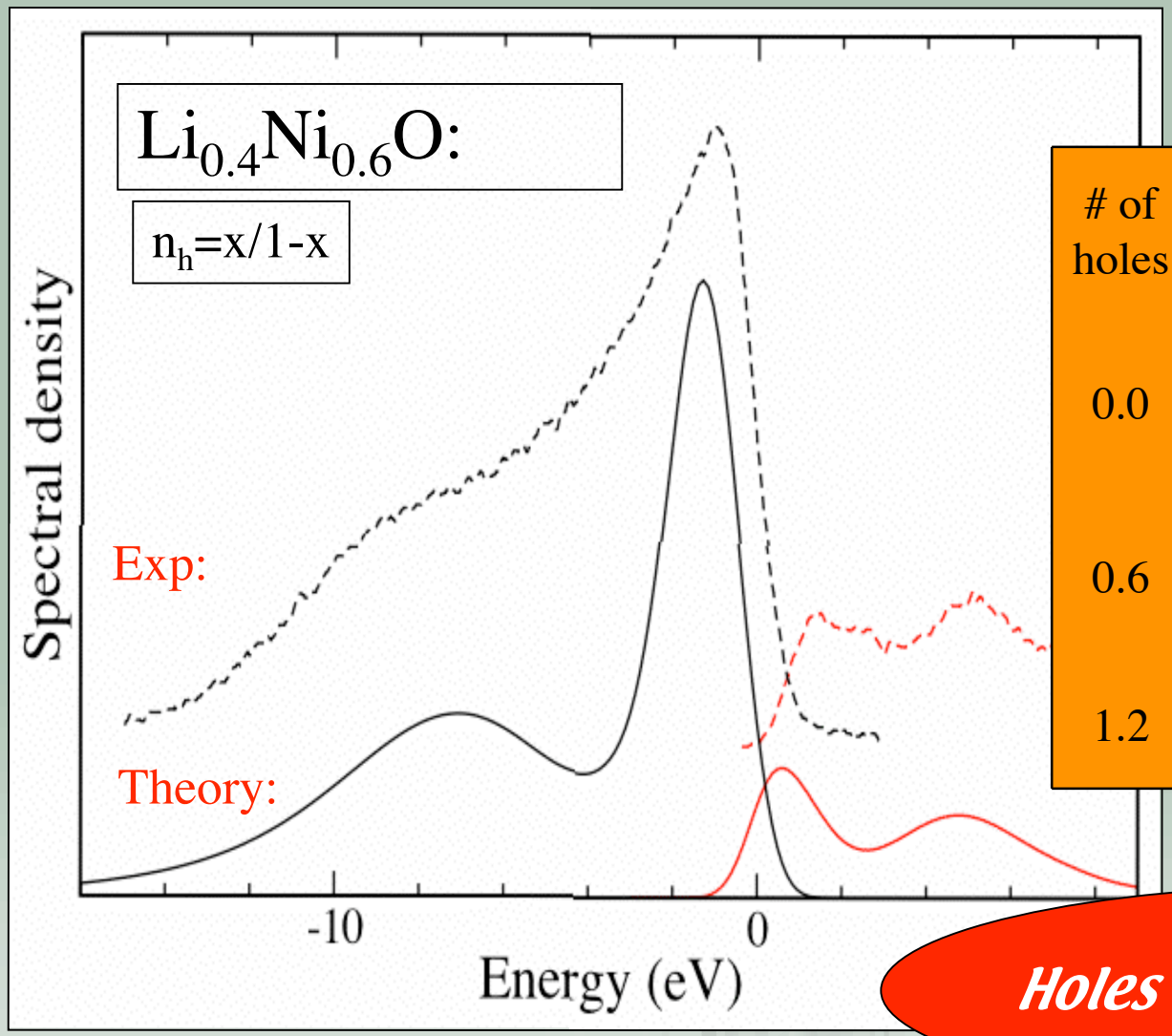
JK et al., PRB ***75****, 165115 (2007)*

Hole doping of NiO



Experimental realization $Li_xNi_{1-x}O$

*van Elp et al. PRB **45**, 1612 (1992)*



# of holes	e_g	t_{2g}	p	m_s
0.0	0.55	1.00	0.97	1.85
0.6	0.53	0.99	0.89	1.61
1.2	0.53	0.98	0.80	1.45

Holes reside on O - sites !

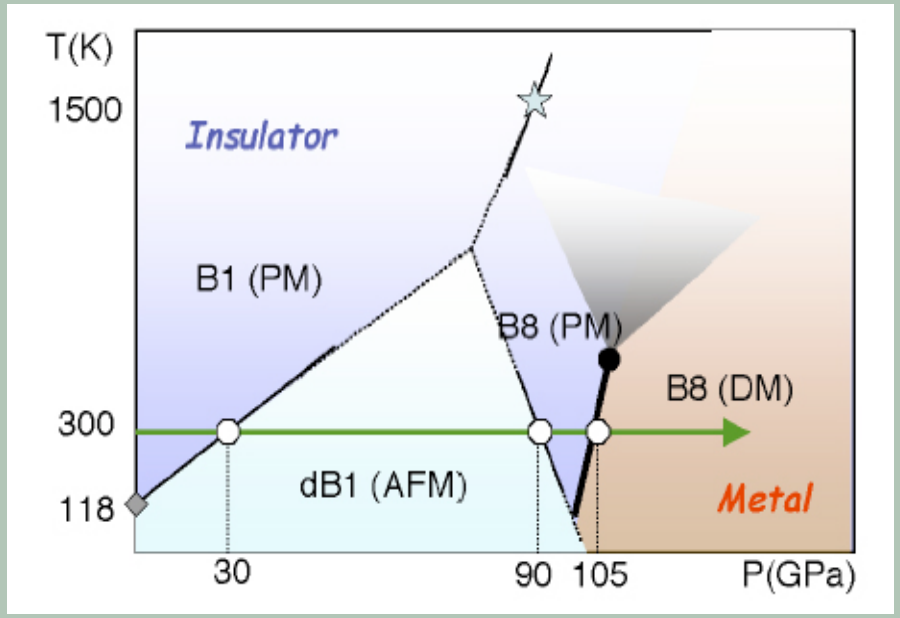
Conclusions I

- NiO fits the description of Mott insulator:
  strong local correlations open a charge gap
  gap in present in paramagnetic phase

- NiO is charge-transfer system:
  doped holes have O-p character
  doping behavior vs PES significantly different from uncorrelated materials

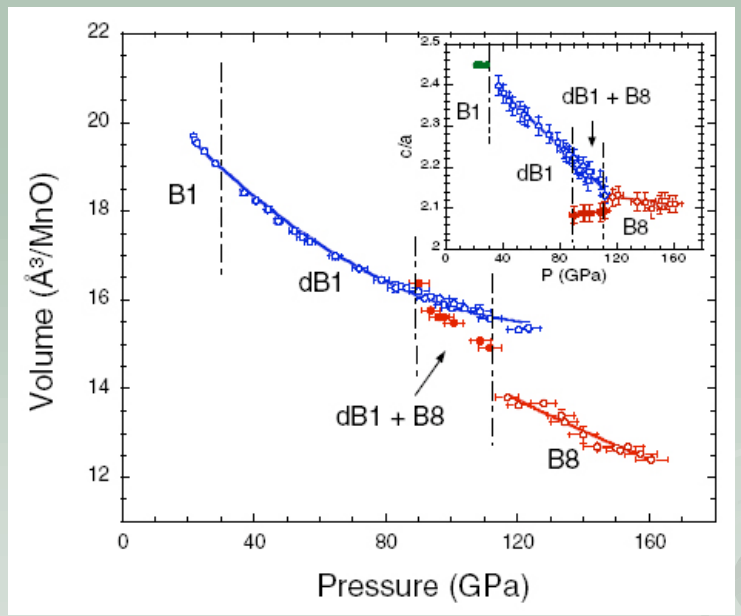
MnO under pressure

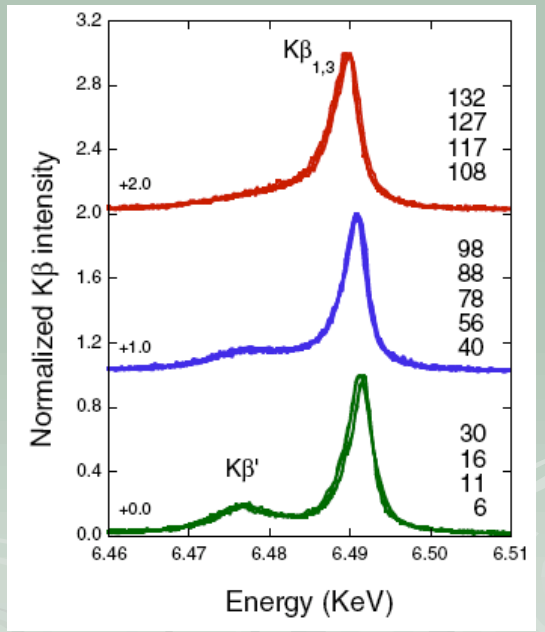
MnO - structure + local moment



Conceptual phase diagram for MnO

- moment collapse
- insulator -> metal transition
- volume collapse
- structural transition



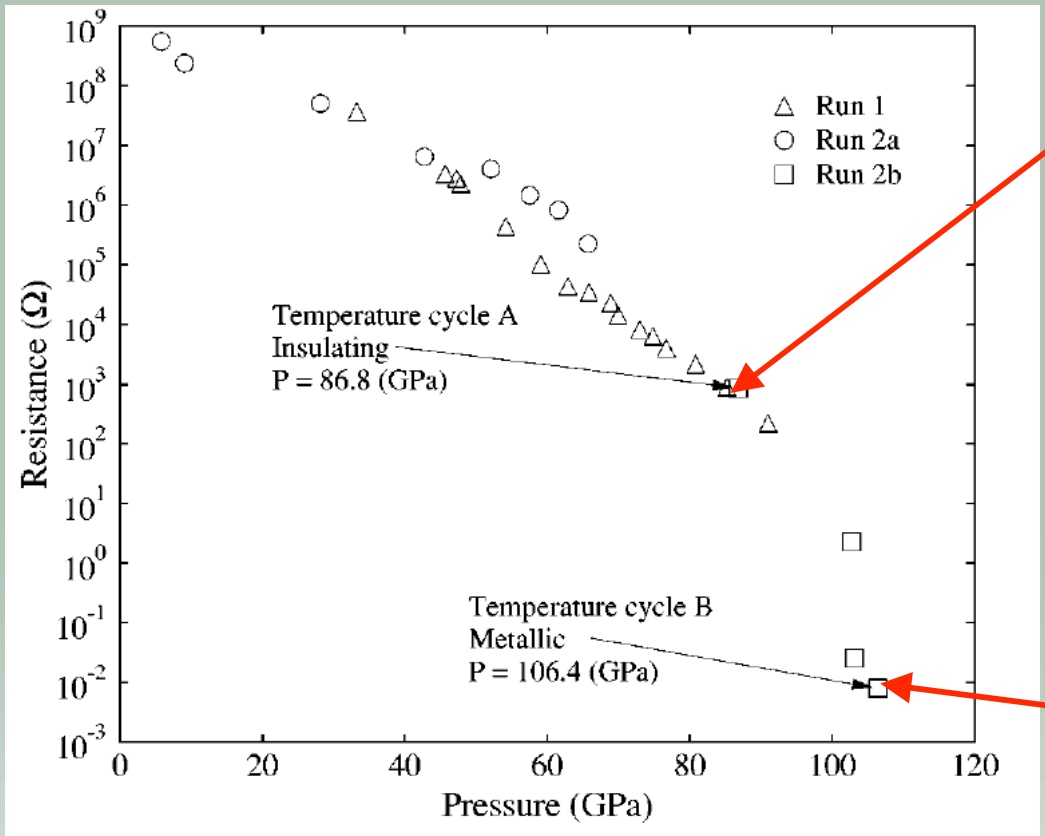


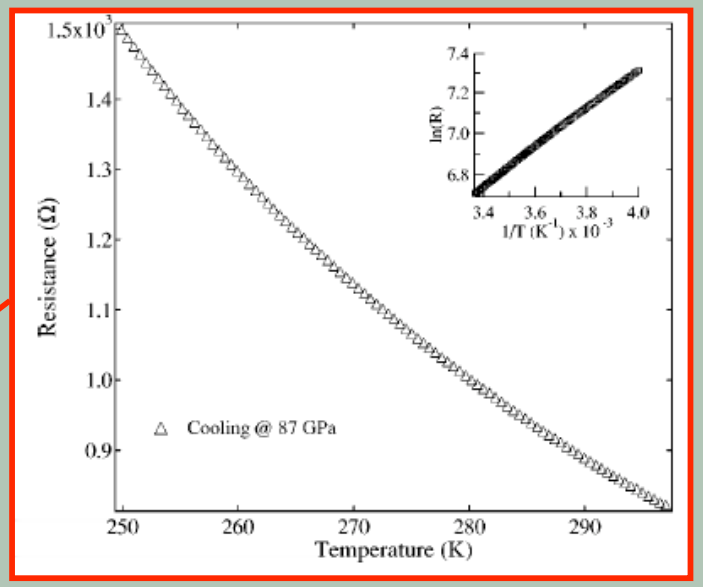
Kβ x-ray emission

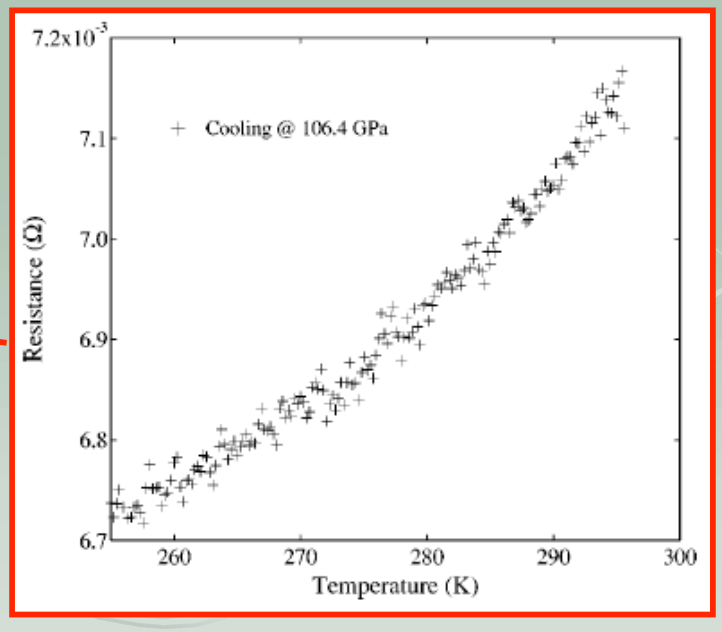
C. S. Yoo et al., Phys. Rev. Lett. ***94****, 115502 (2005)*

Insulator -> metal transition

Resistance:







*Patterson et al., Phys. Rev. B **69**, 220101R (2004)*

MnO - magnetic moment vs volume

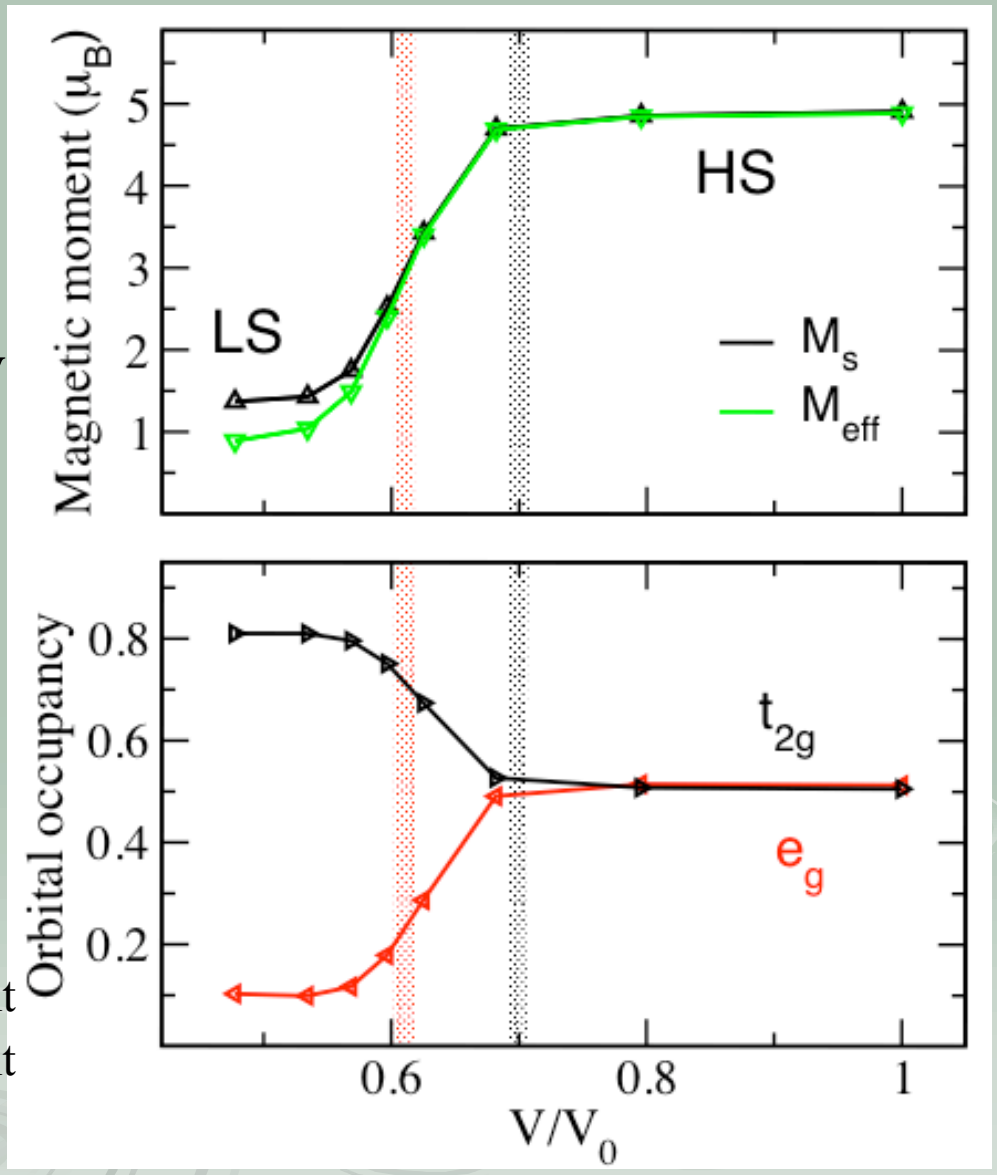
No change down to v_c=0.68

Fluctuations increase dramatically in metallic phase at/below p_c

Orbital occupations follow the atomic scenario of J vs Δ_{cf} competition

$M_s^2=\langle m_z^2\rangle$ instantaneous moment

$M_{eff}^2=T\int d\tau\langle m_z(\tau)m_z(0)\rangle$ screened moment



MnO –

No change d

Fluctuations in
in metallic phase a

Orbital occupatio ollow
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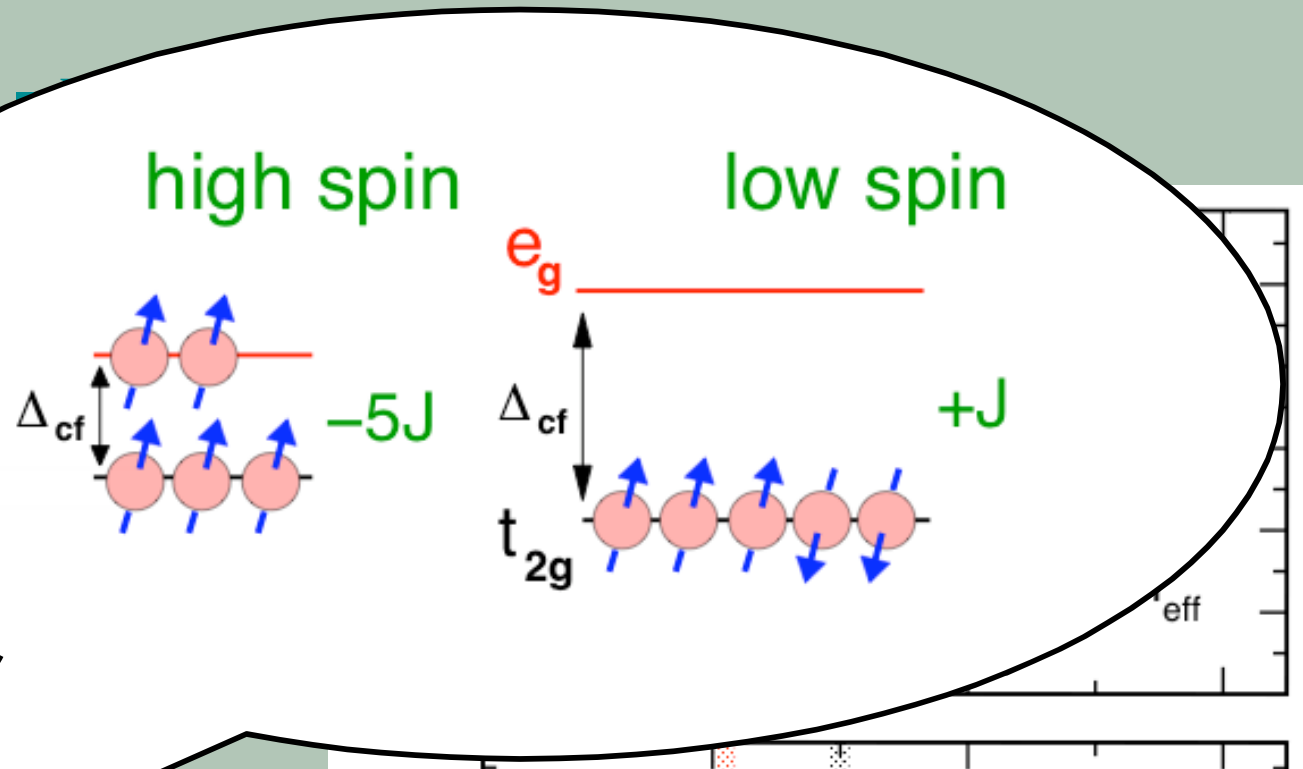
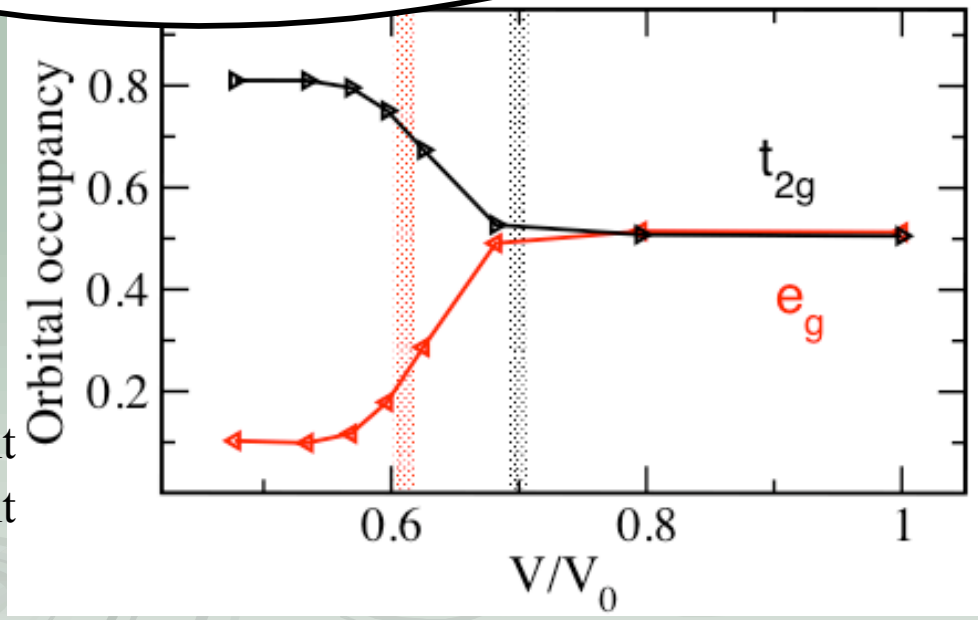
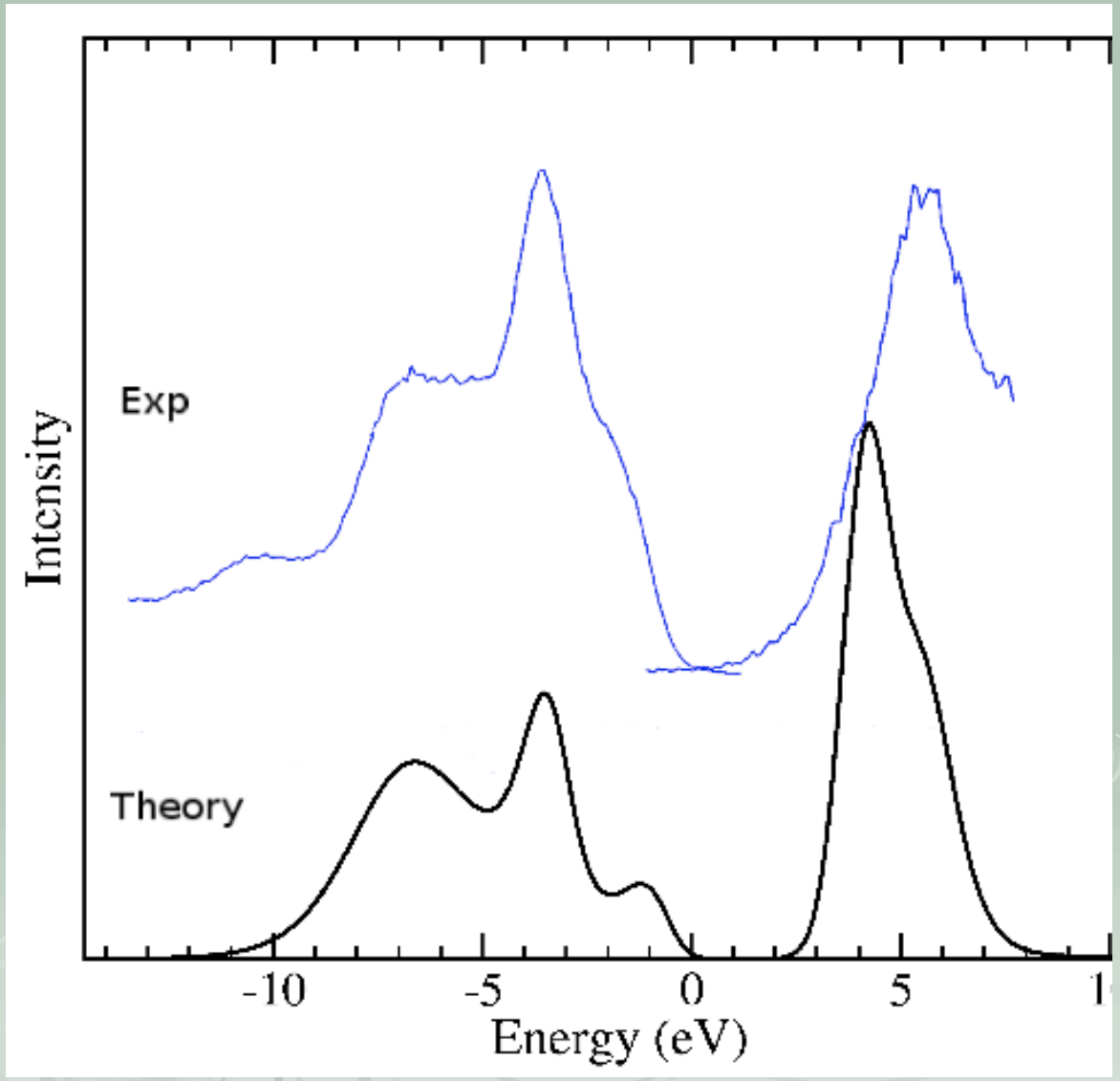




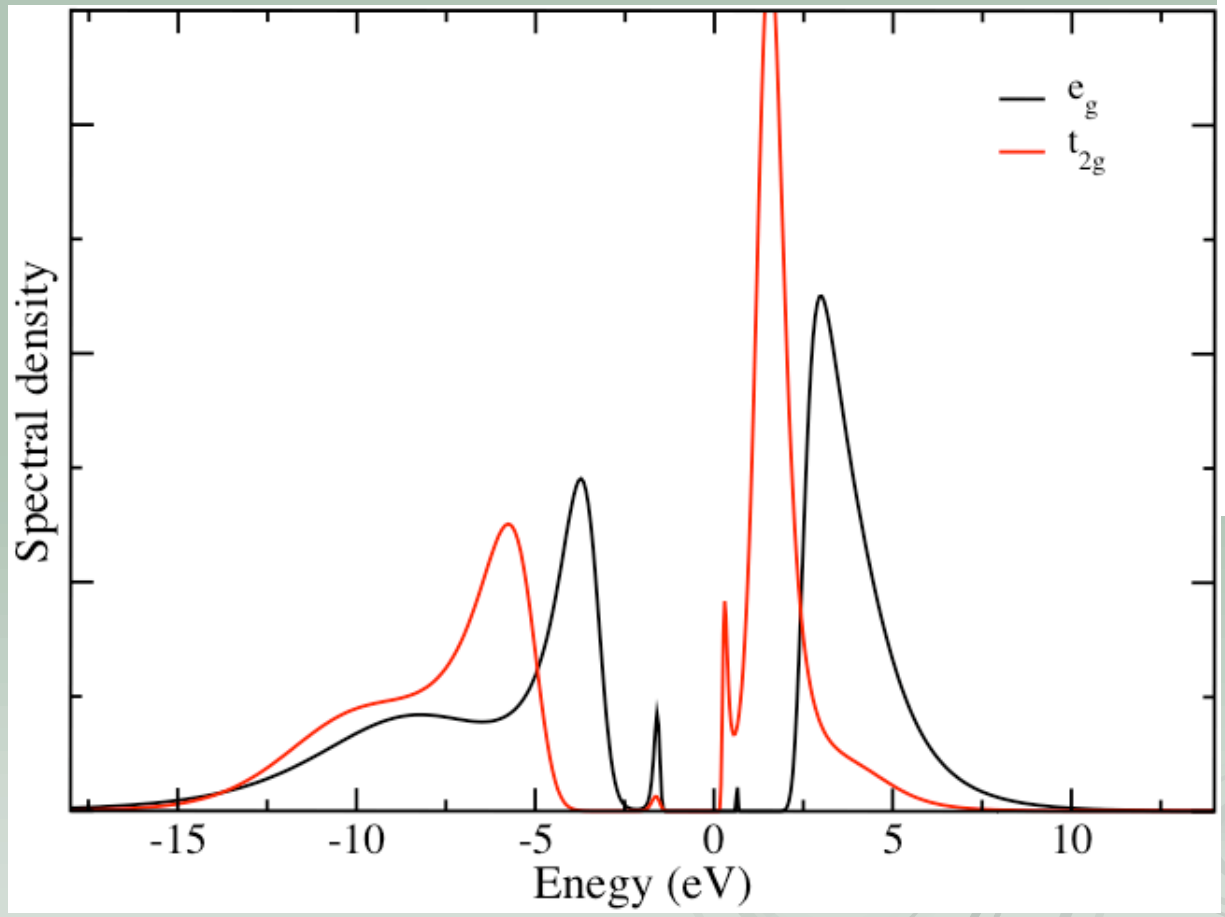
$M_s^2=\langle m_z^2\rangle$ instantaneous moment
$M_{eff}^2=T\int d\tau\langle m_z(\tau)m_z(0)\rangle$ screened moment

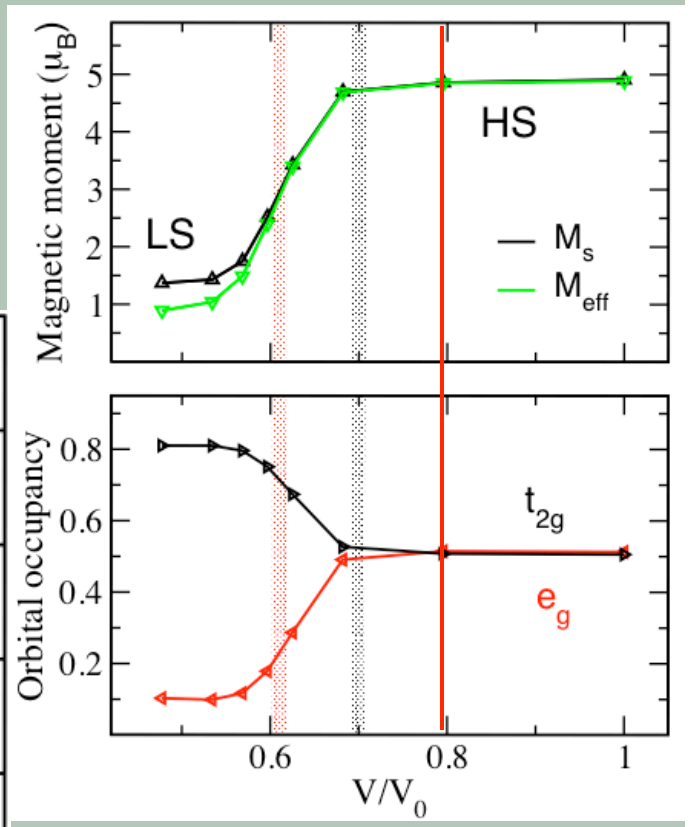
MnO - spectral density at ambient pressure

PES/BIS (*van Elp et al. PRB* ***44****, 1530 (1991)*)
vs
Mn-*3d* spectral density:
(*U*=6.9 eV, *J*=0.86 eV)

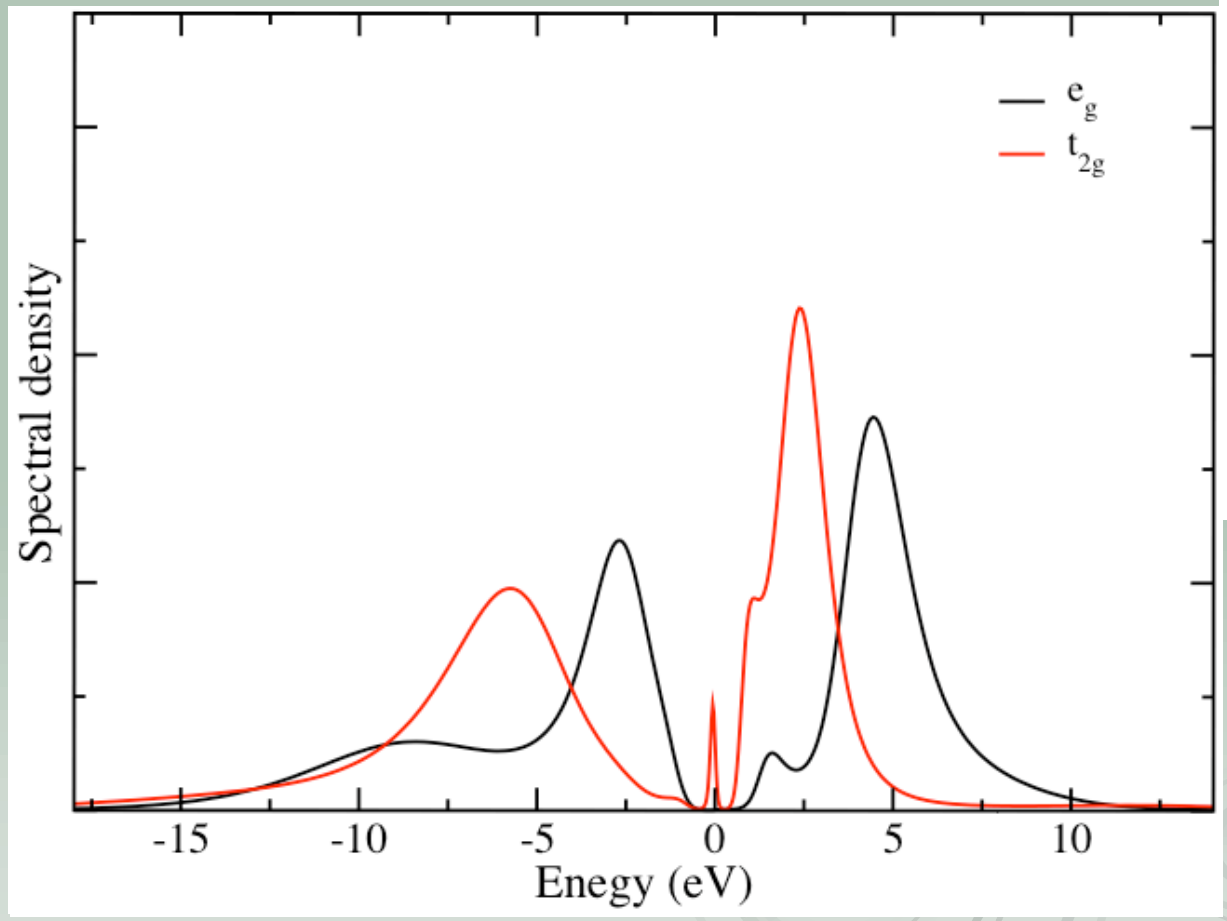


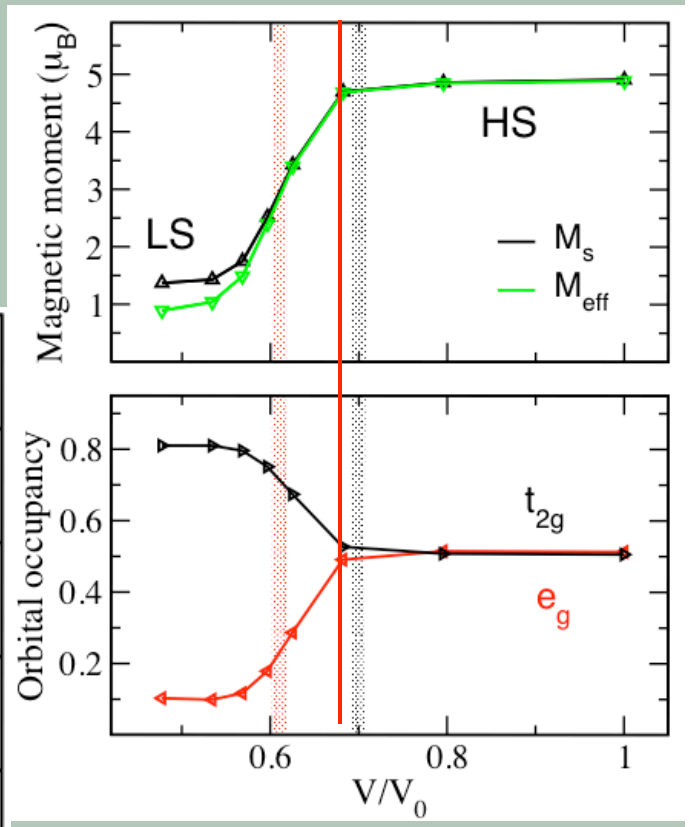
Pressure induced metallization



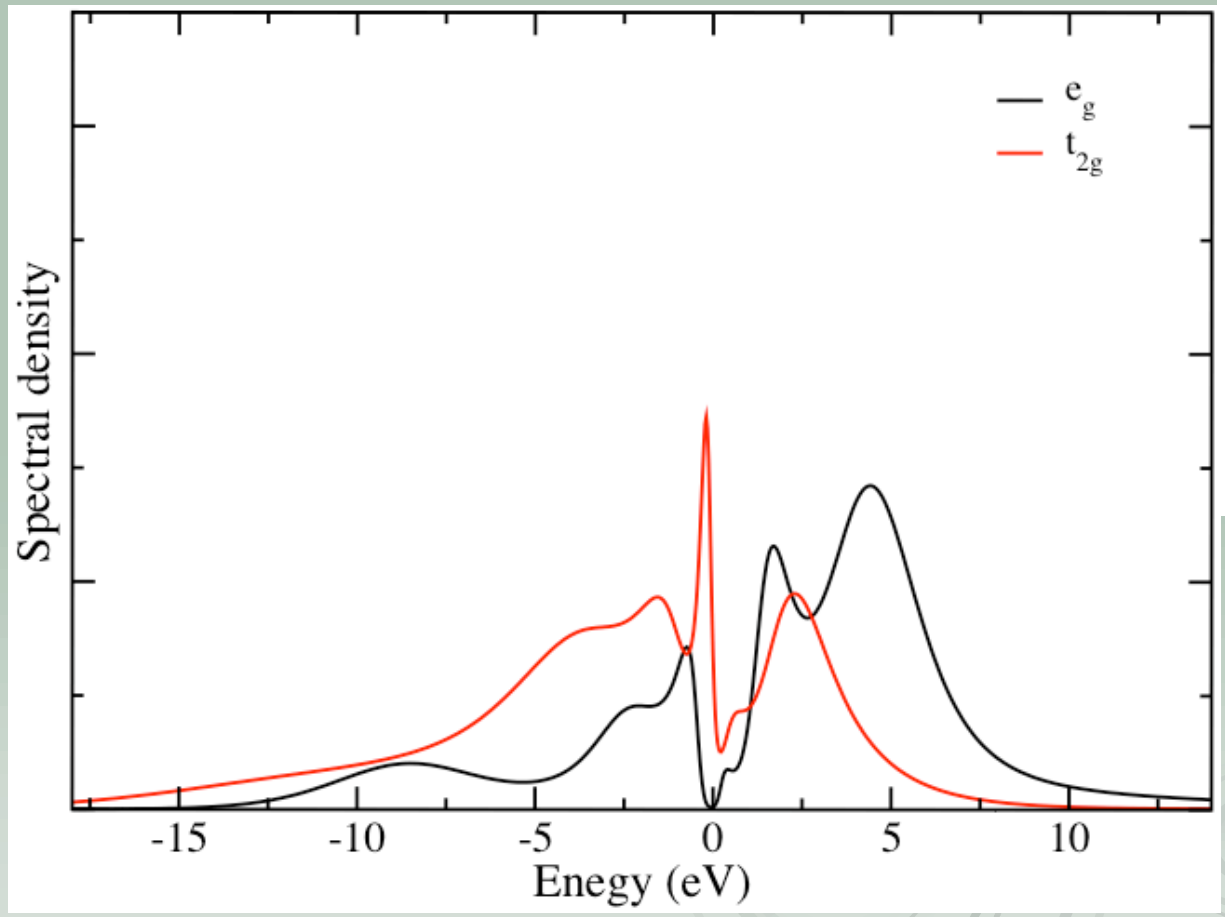


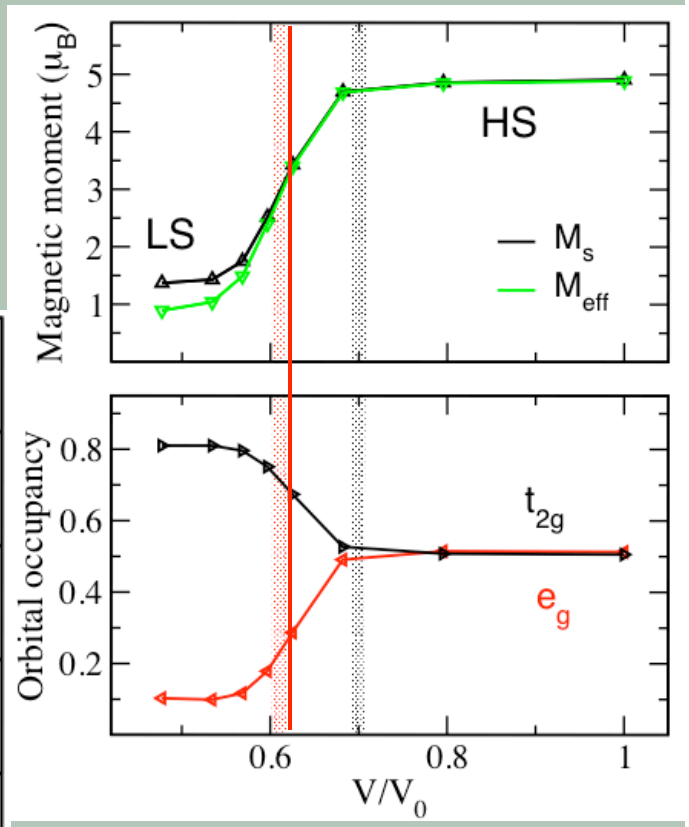
Pressure induced metallization



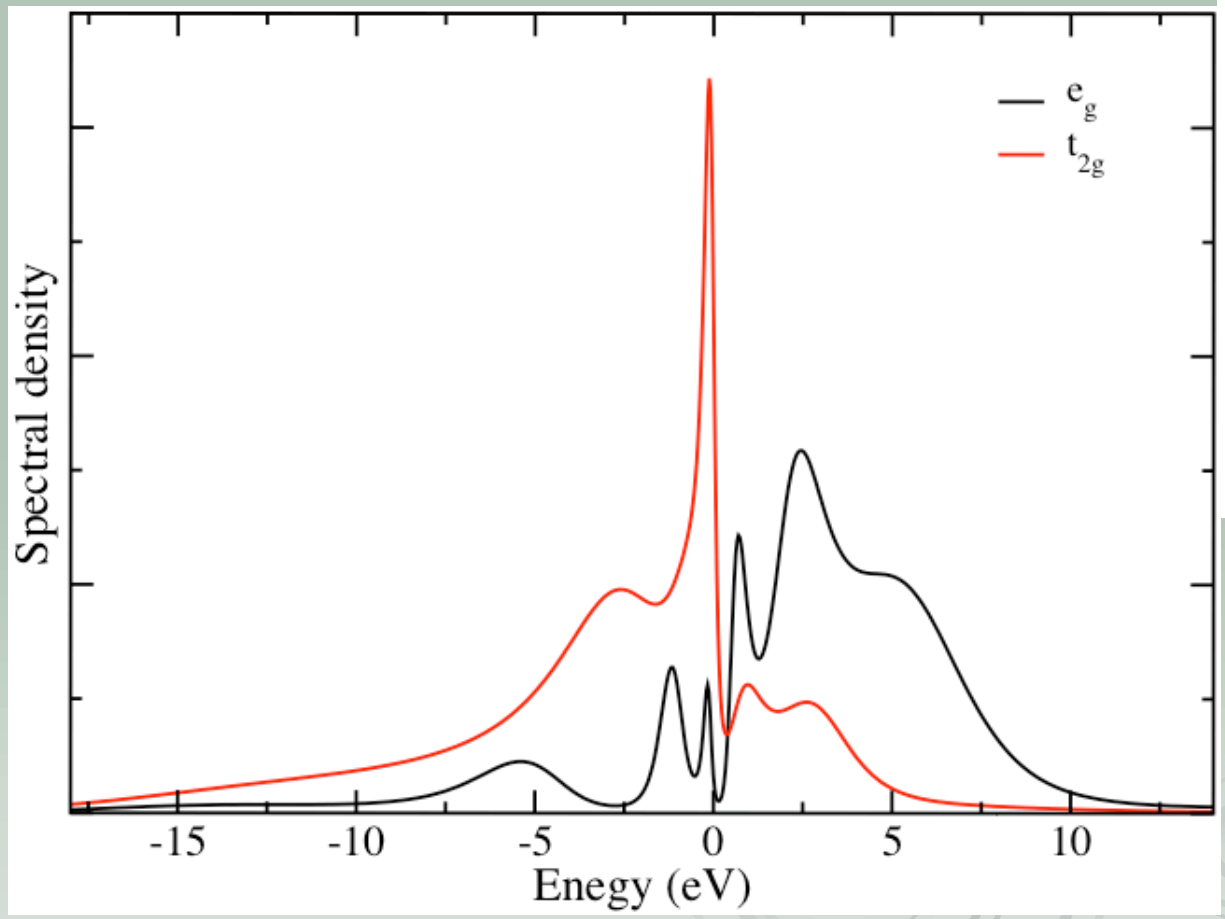


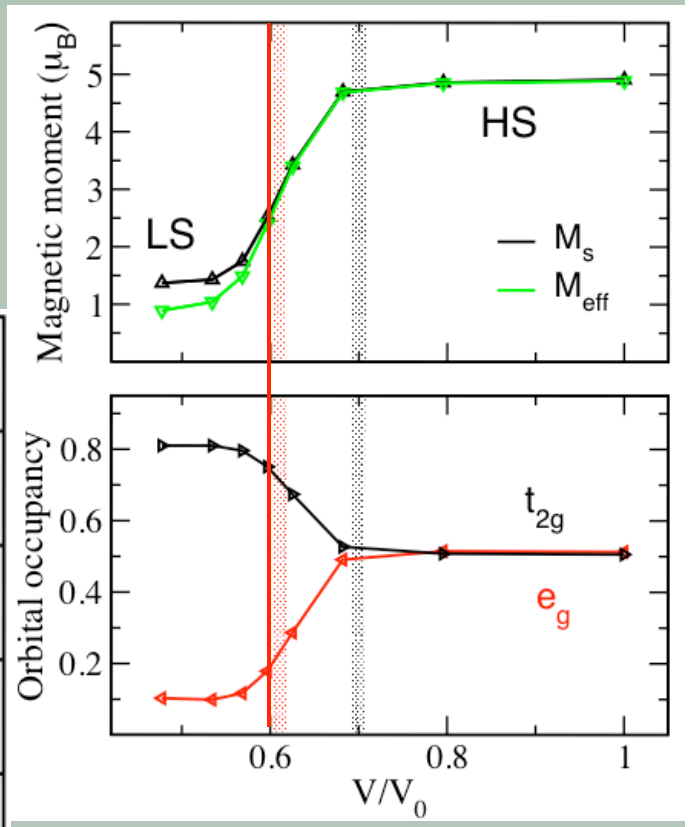
Pressure induced metallization



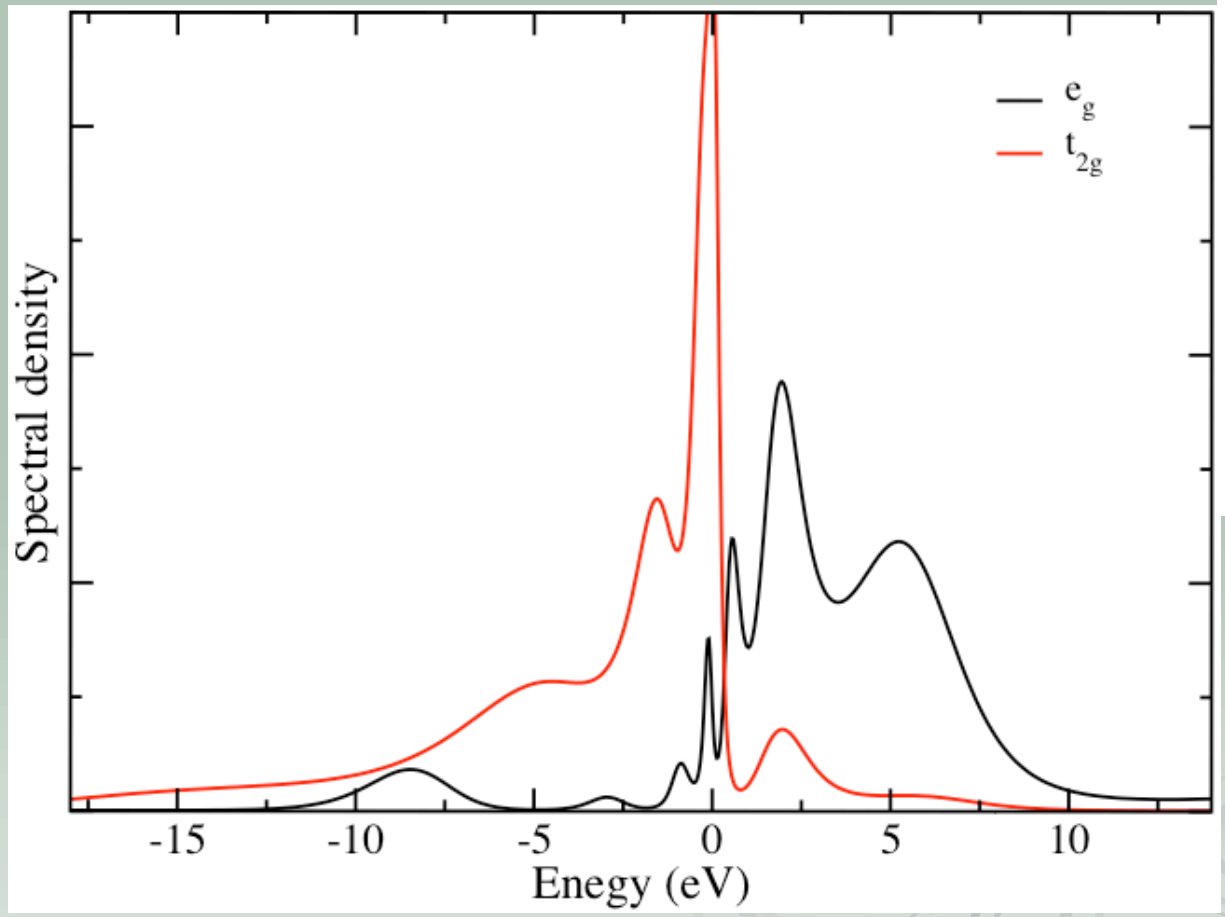


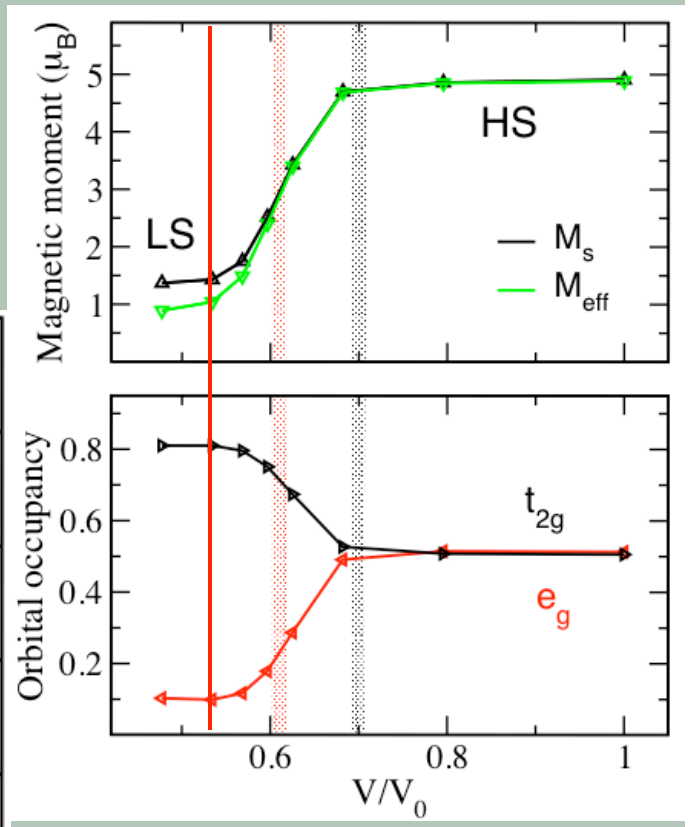
Pressure induced metallization





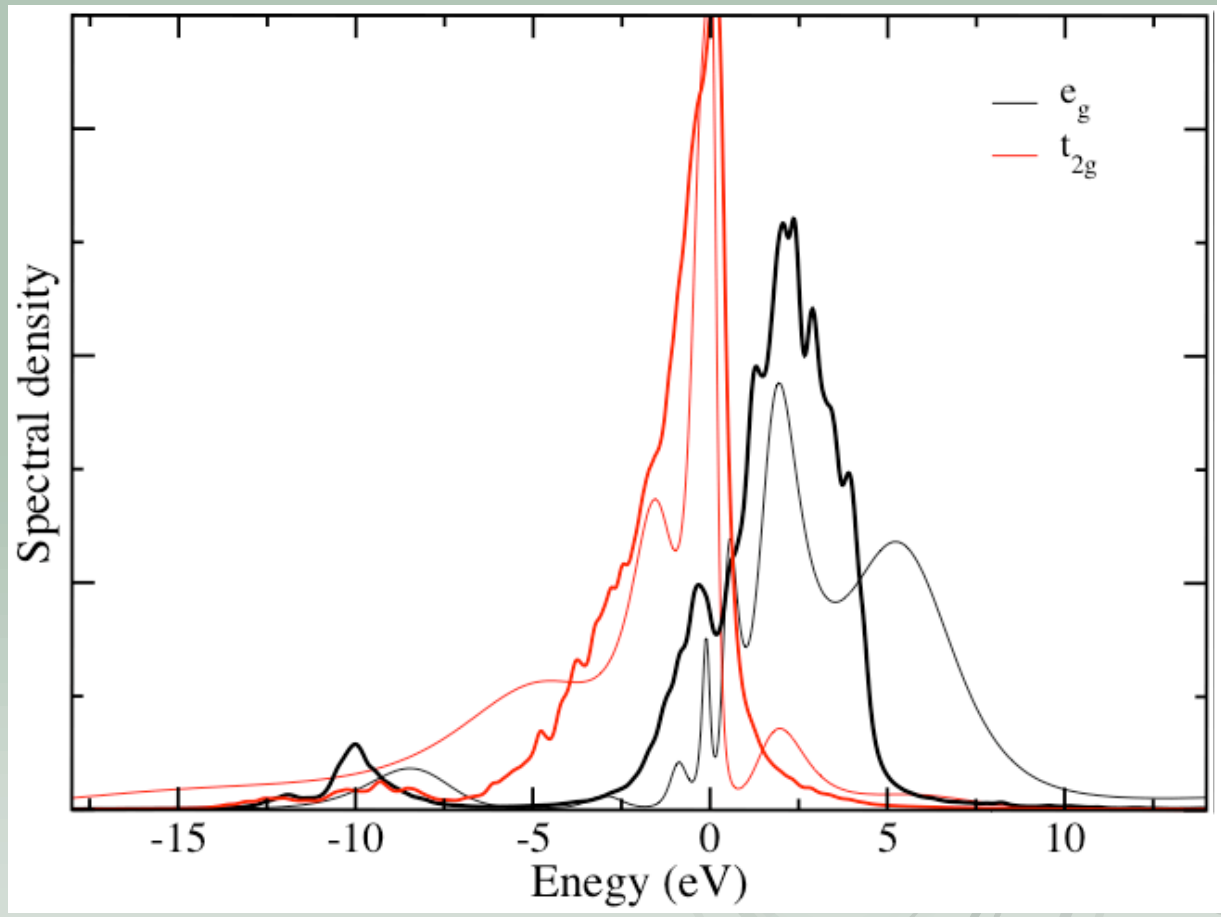
Pressure induced metallization

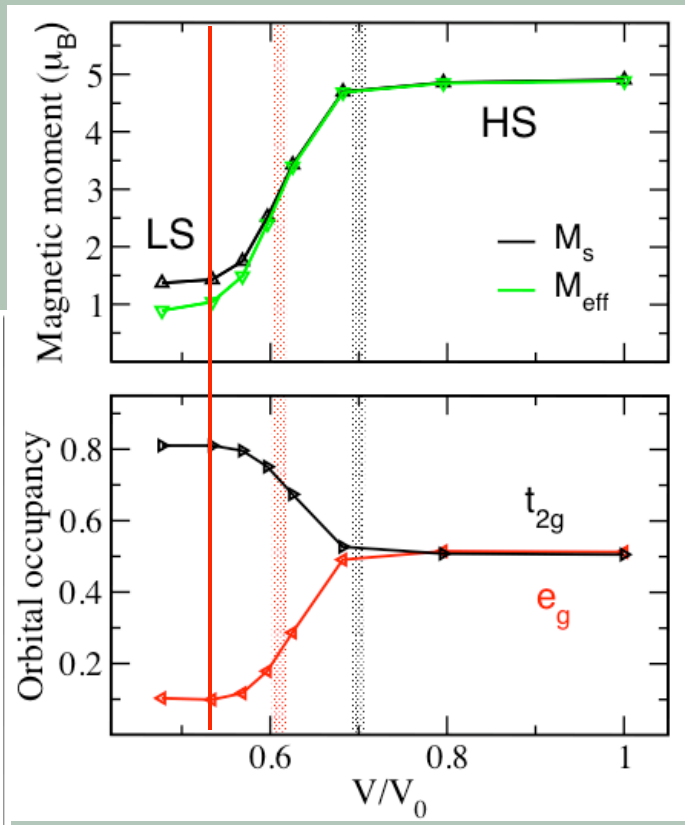




Pressure induced metallization

Correlation effects weaker in LS





Conclusions II

- LDA + DMFT captures well moment collapse, metal-insulator transition and volume collapse in MnO
- Moment collapse and MIT happen simultaneously.
- Mott transition is 'orbital selective'.
- Moment collapse mechanism? (bandwidth or crystal field)

  Crystal field
- Why is HS insulating and LS metallic? (bandwidth?)

  Effective $U_{eff}=E(d^{n+1})+E(d^{n-1})-2E(d^{n})$:

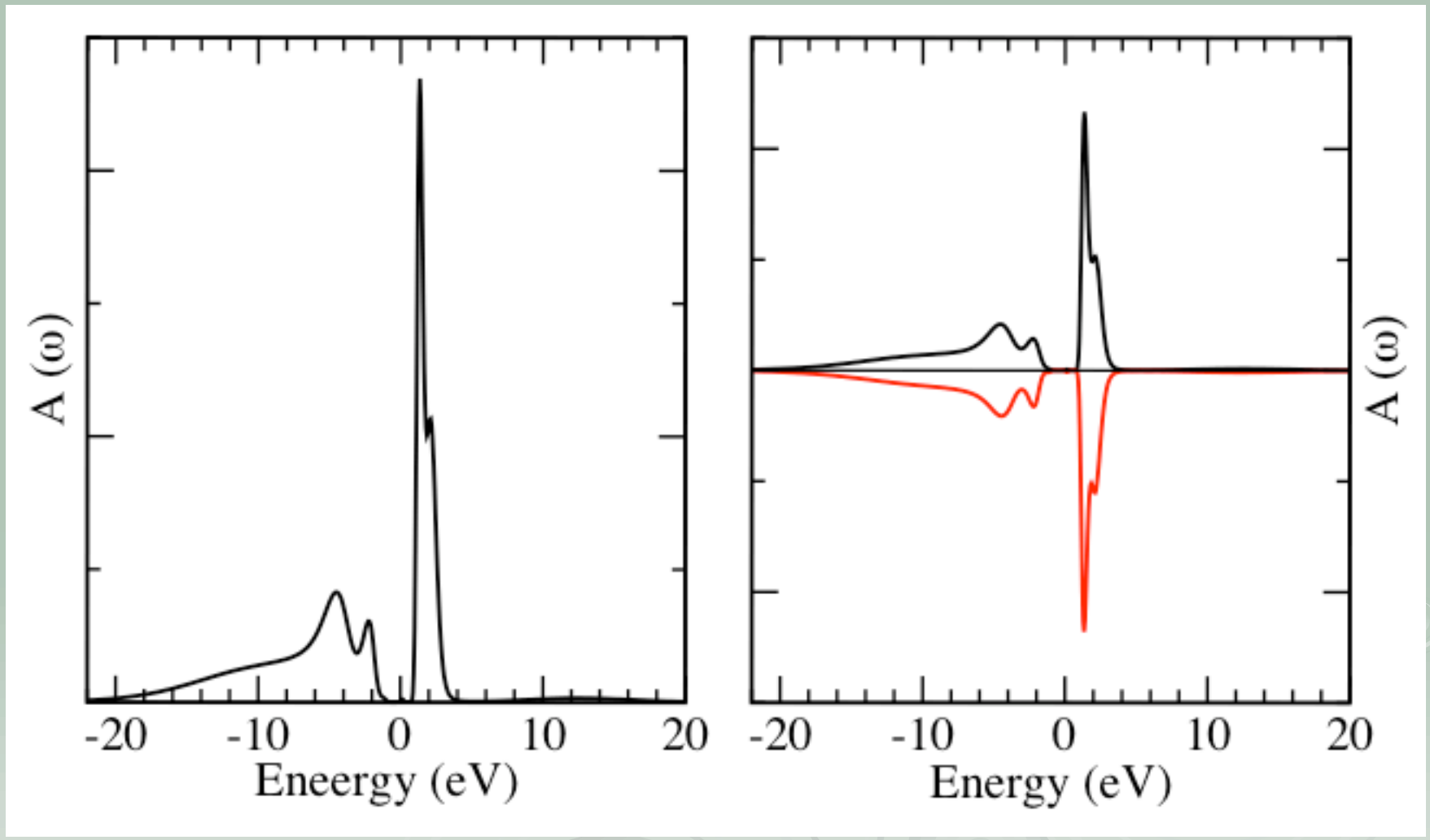
  U_{eff}(HS) ~ 10.3 eV >> U_{eff}(LS) ~ 5.9 eV

  Constraints imposed by the local 'ground state'
- Similarity to 2b-model of Werner & Millis, *PRL* **99**, 126405 (2007)

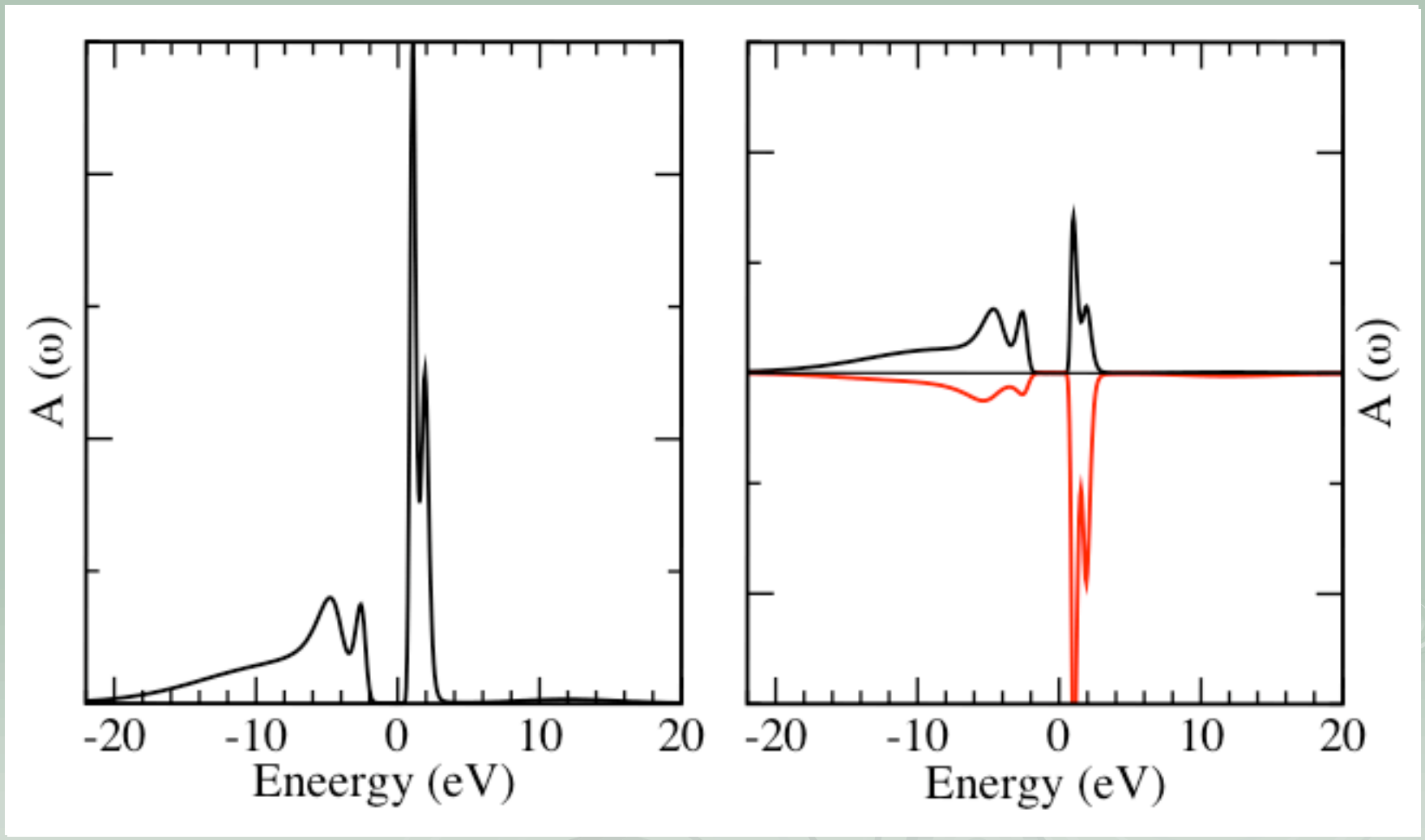
JK, A. L. Lukoyanov, V. I. Anisimov, R. T. Scalettar, and W. E. Pickett, Nature Materials 7, 198 (2008)

Fe_2O_3: role of AFM order

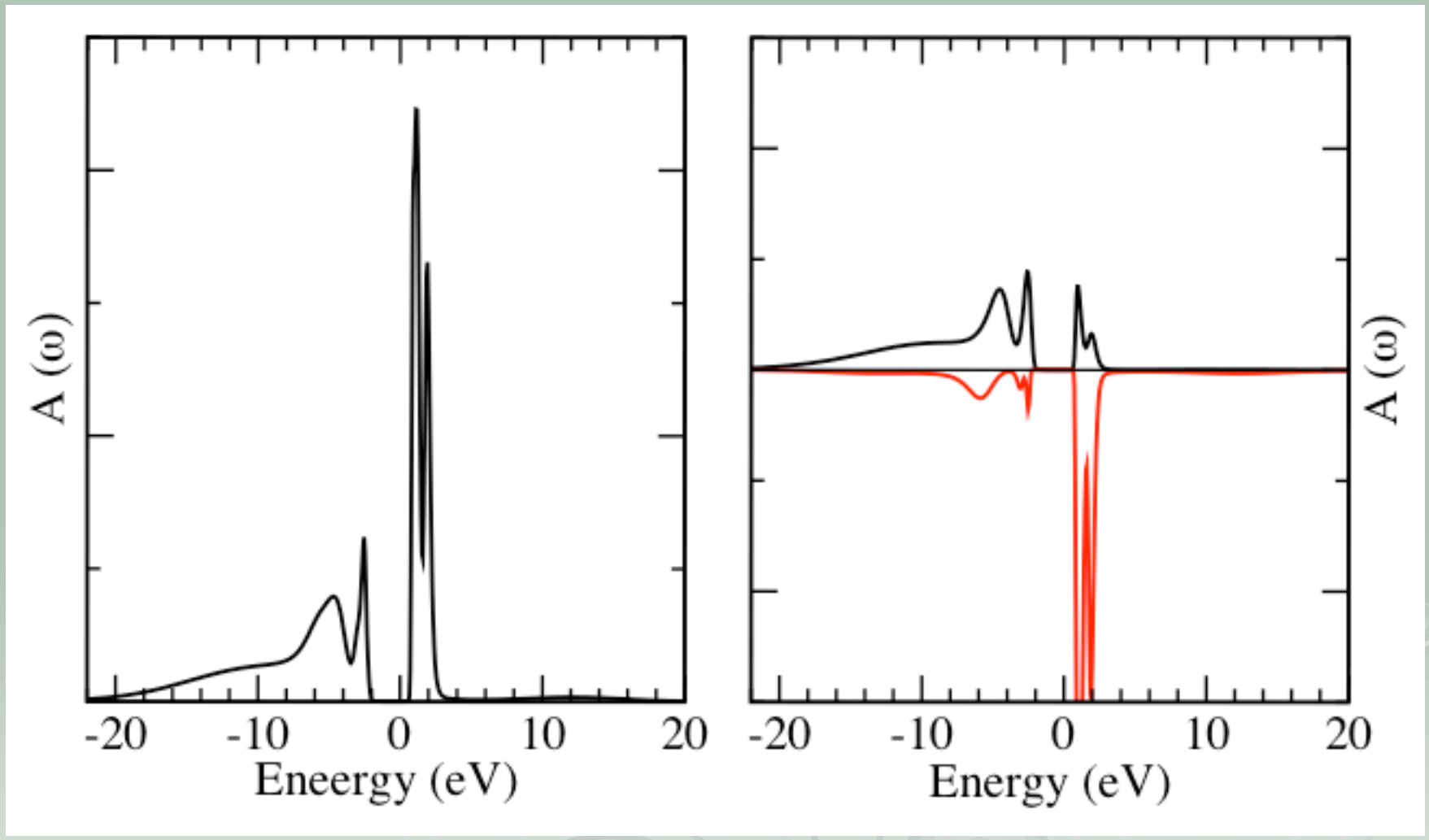
AFM order Fe_2O_3



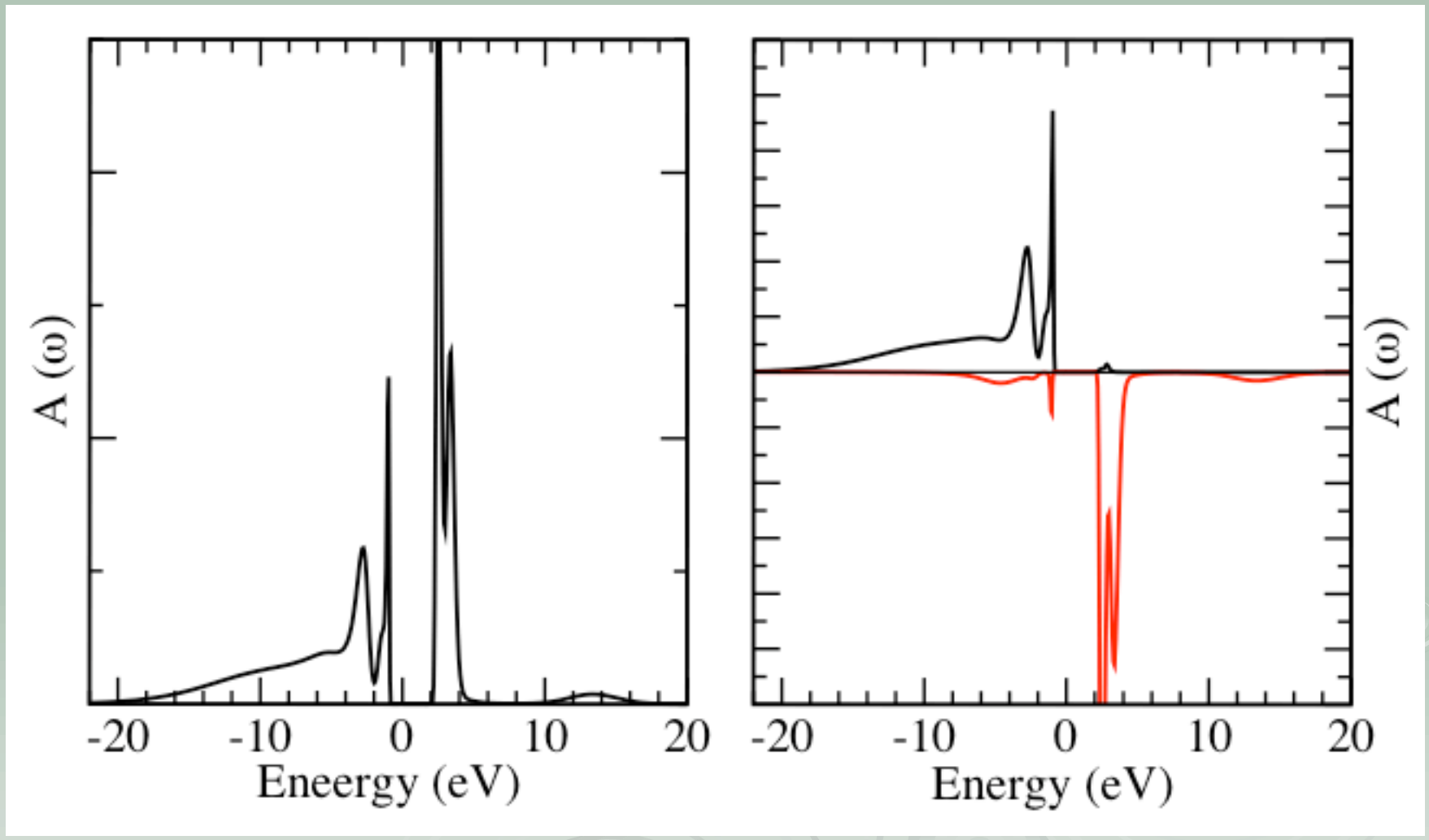
AFM order Fe_2O_3



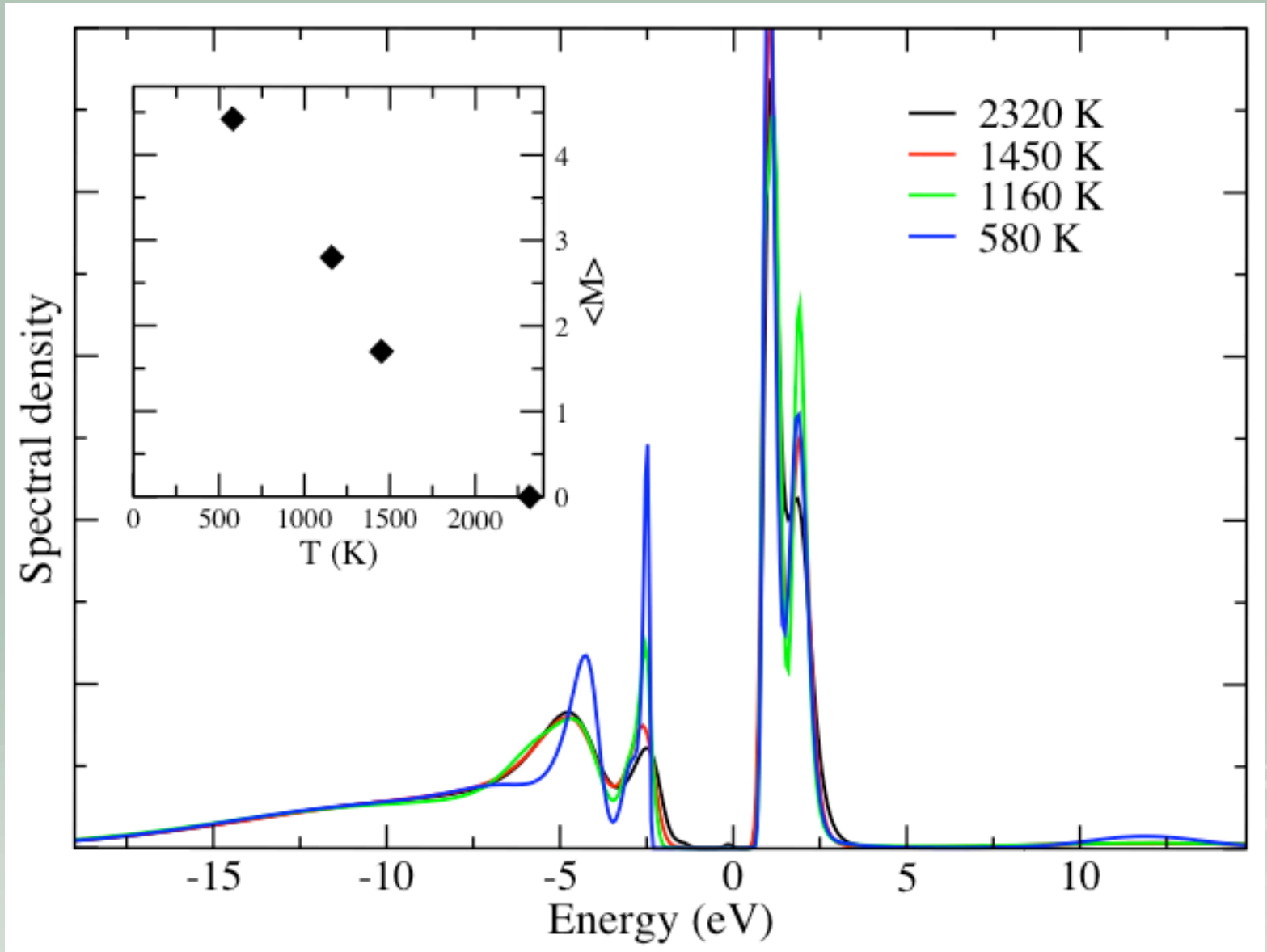
AFM order Fe_2O_3



AFM order Fe_2O_3



AFM order Fe_2O_3



Conclusions III

- Magnetic order have small effect on the single-particle spectrum of Mott insulator

JK, D. M. Korotin, and M. A. Korotin, unpublished

Outlook

- How general is the moment collapse mechanism of Mott transition and how does LDA+DMFT perform for other 'moment collapse materials' ? e.g. Fe_2O_3 (d^5), FeO (d^6)
- Role of covalency?
- Relationship to structural changes? Magnetic order?

- Construction of H_0 for multi-band systems:

double counting => charge-transfer energy, charge self-consistency?

- rotationally invariant interaction, general (off-diagonal) self-energy, spin-orbit coupling - can be done with continuous time QMC