Defects as a path to a new class of magnetic materials

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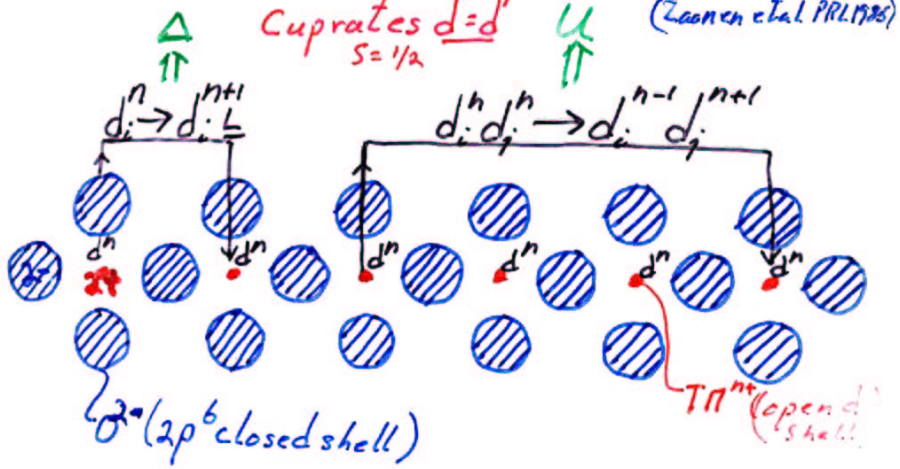
Why insulators M-H vs C T

ionic localized Ansatz

Cuprates $d^n = d^9$ $S = 1/2$ (Zaanen et al PRL 1985)



$$U = E_I^{TM^{n+}} - E_A^{TM^{n+}} - E_{pol}$$

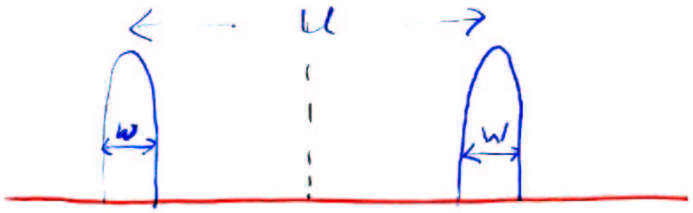
$$\Delta = E_I^{O^{2-}} - E_A^{TM^{n+}} - E_{pol}' + [V_M^{O} - V_M^{TM^{n+}}]$$

$$\sim 2\,E(\text{Madelung})$$

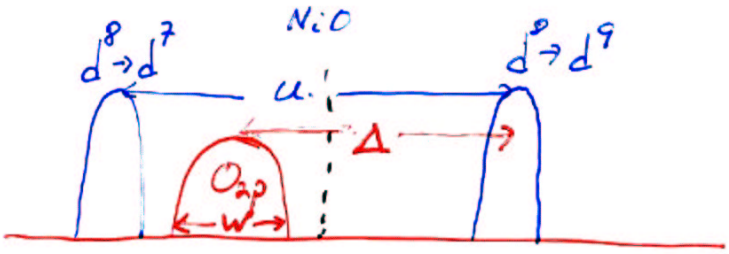
$$E_{pol} = \frac{1}{2}\sum_i \alpha_i F_i^2$$

[F_i = field on ion i]
[α_i = polarizability]

½ filled s band [H]

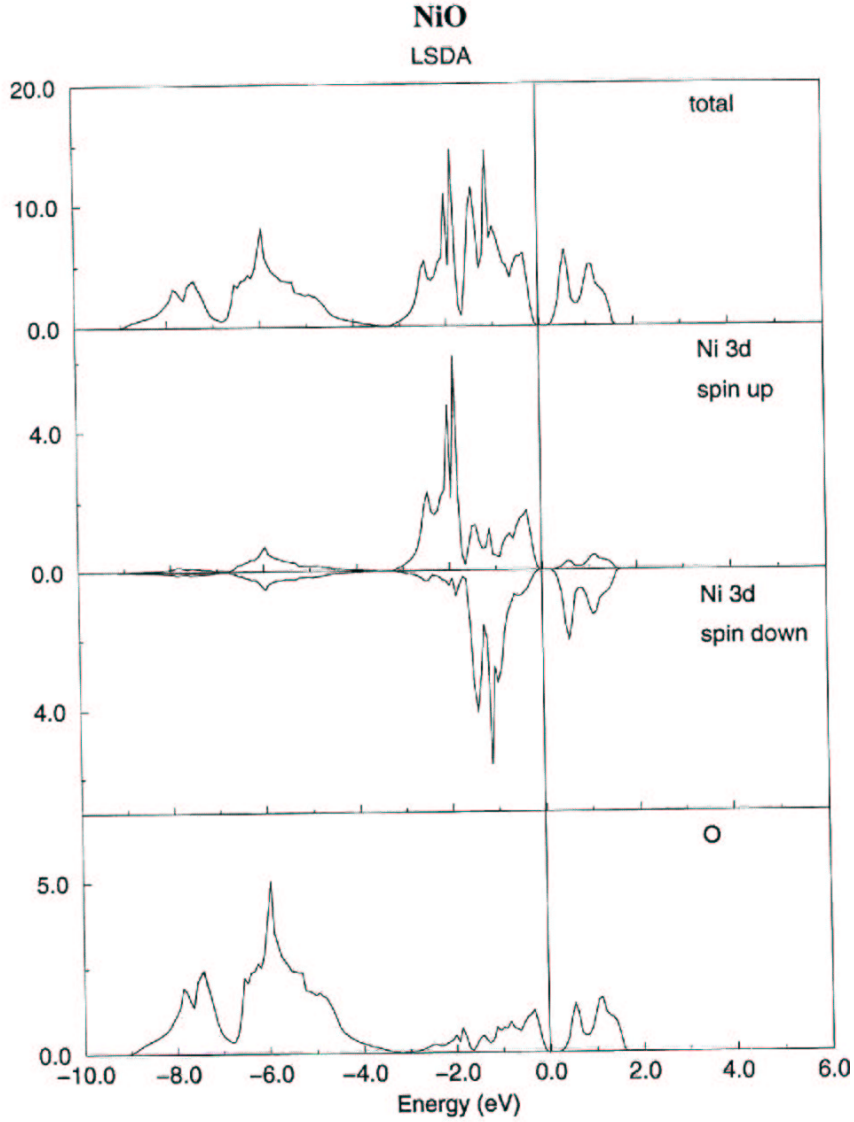


U > W magnetic Insulator

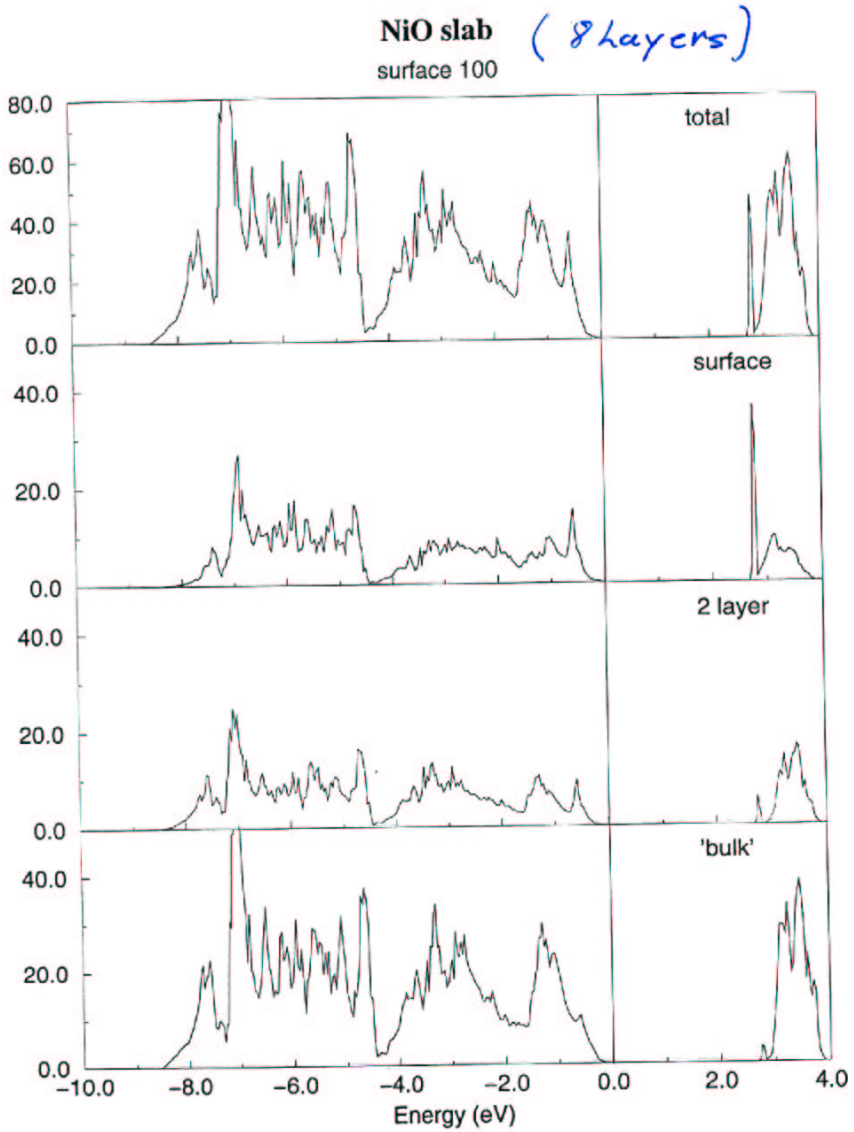


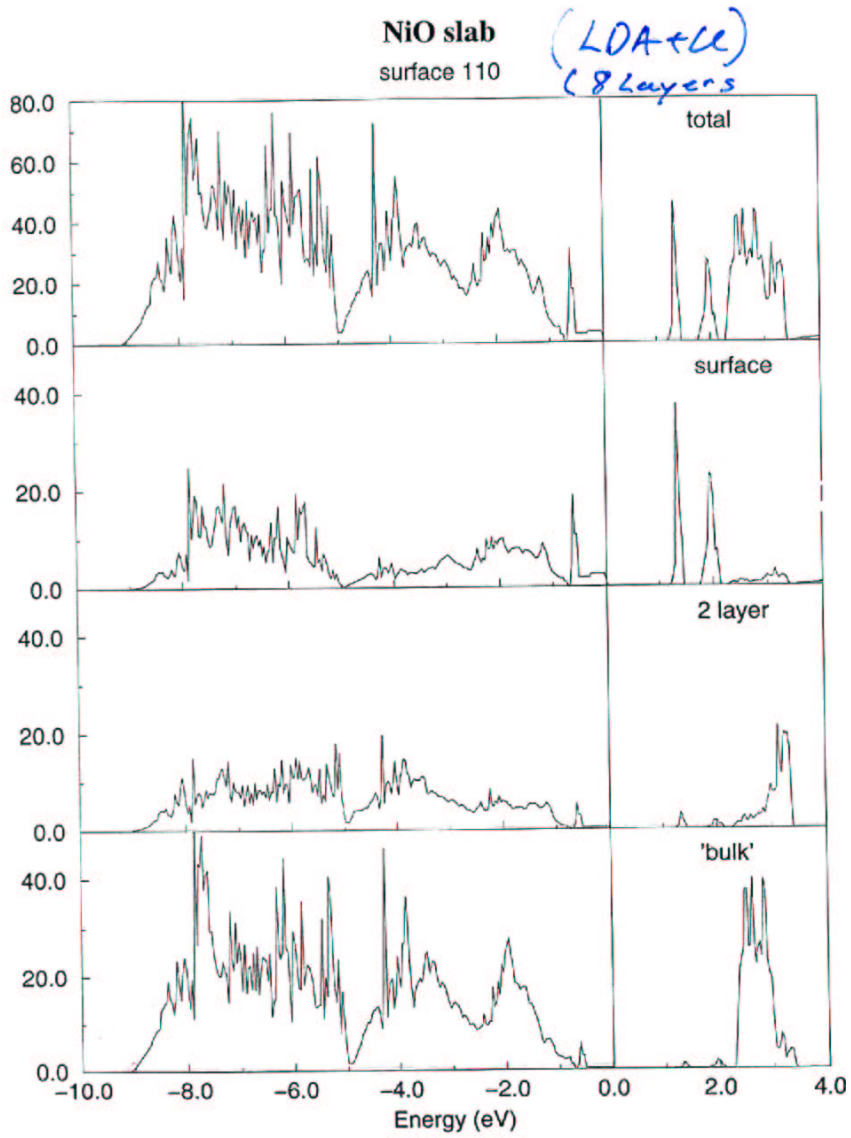
$\Delta < \frac{W}{2}$ → Metal - Self doped

ZSA Theory (1984)



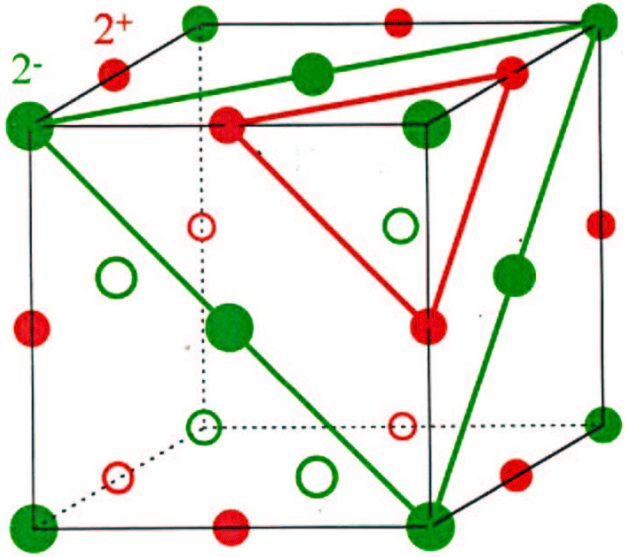
LDA+U Elfimov



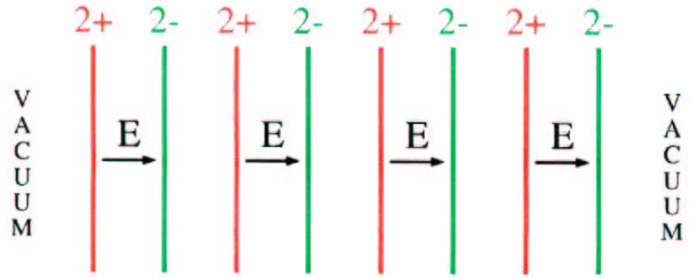


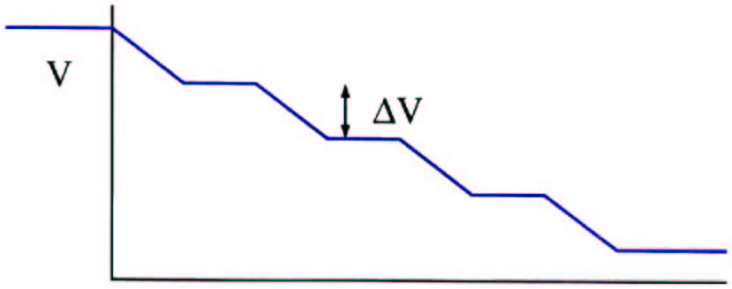
Polar Surfaces

- Existence of non-neutral or charged planes in crystal structures
- Rocksalt (111) surfaces: MgO, NiO



Finite slab of charged planes

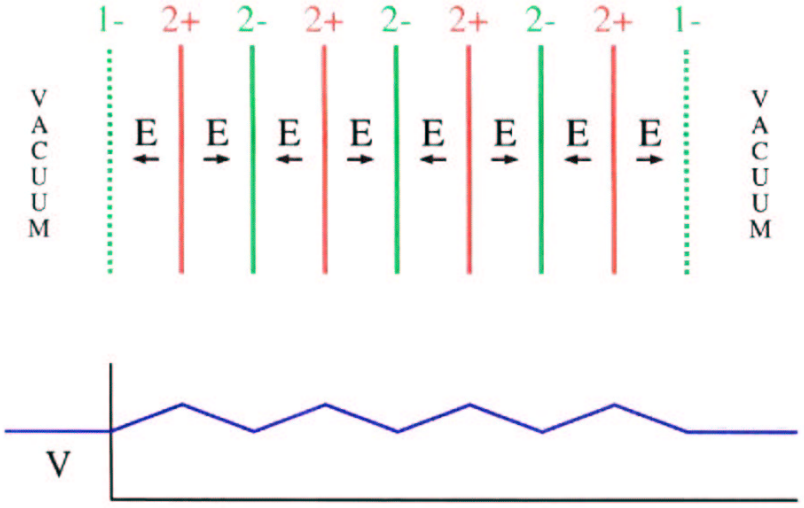




ΔV = 58 Volt per MgO or NiO double layer

IMPOSSIBLE !!

Finite slab of charged planes
Half-charge terminated



Potential is determined by the boundary conditions!!

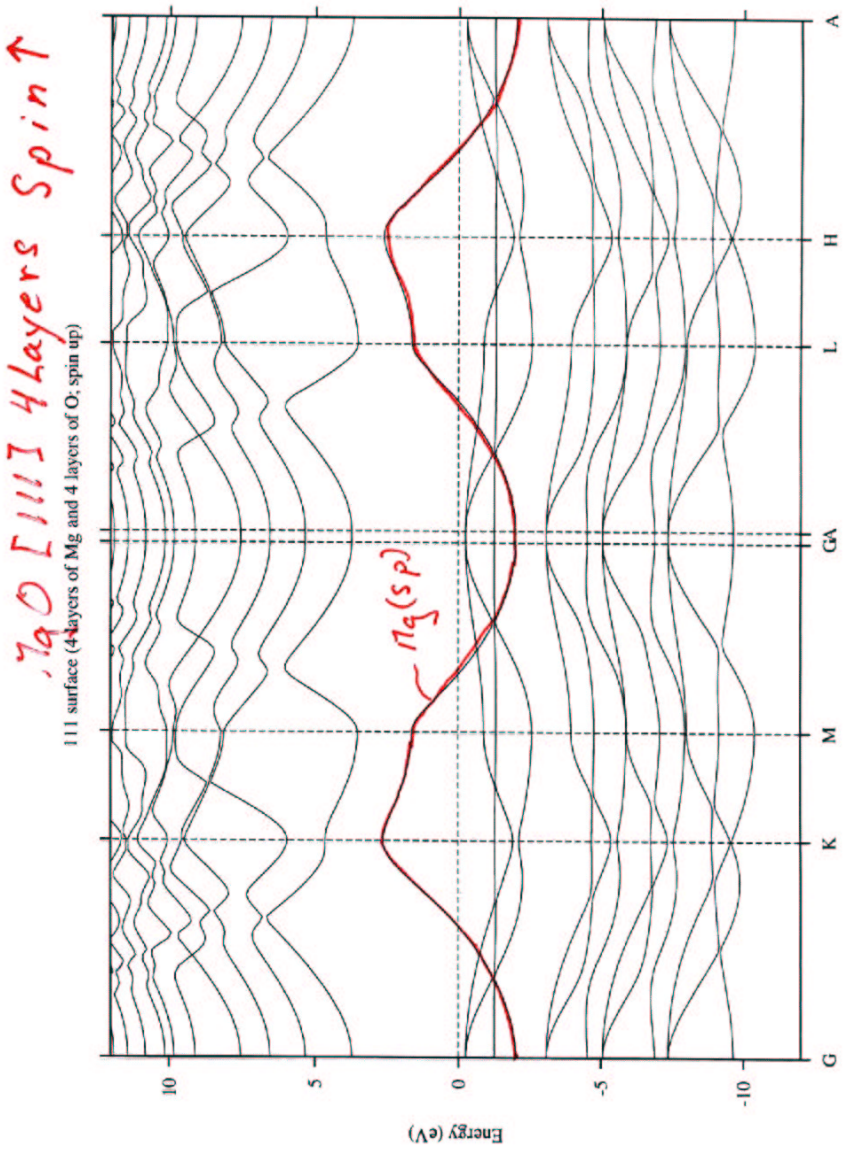
Surface

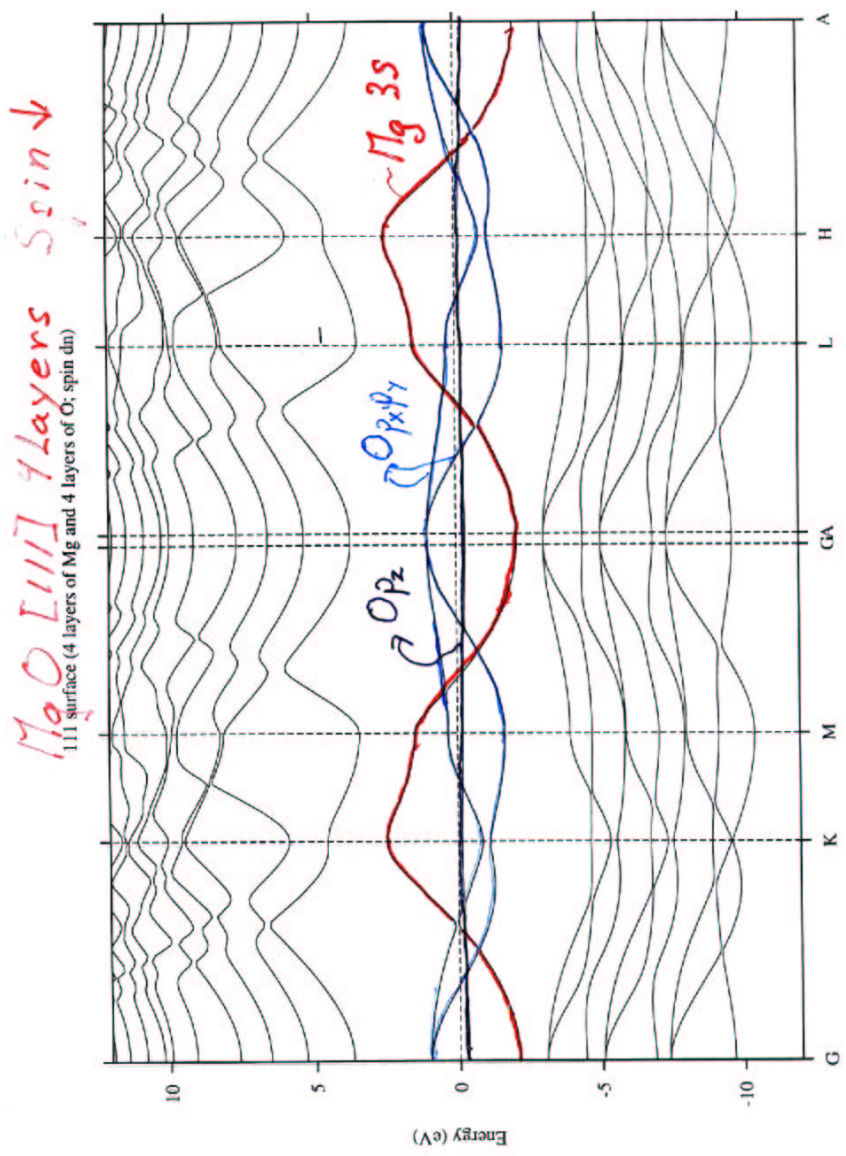
-facets: pyramids with neutral, e.g. (100) surfaces

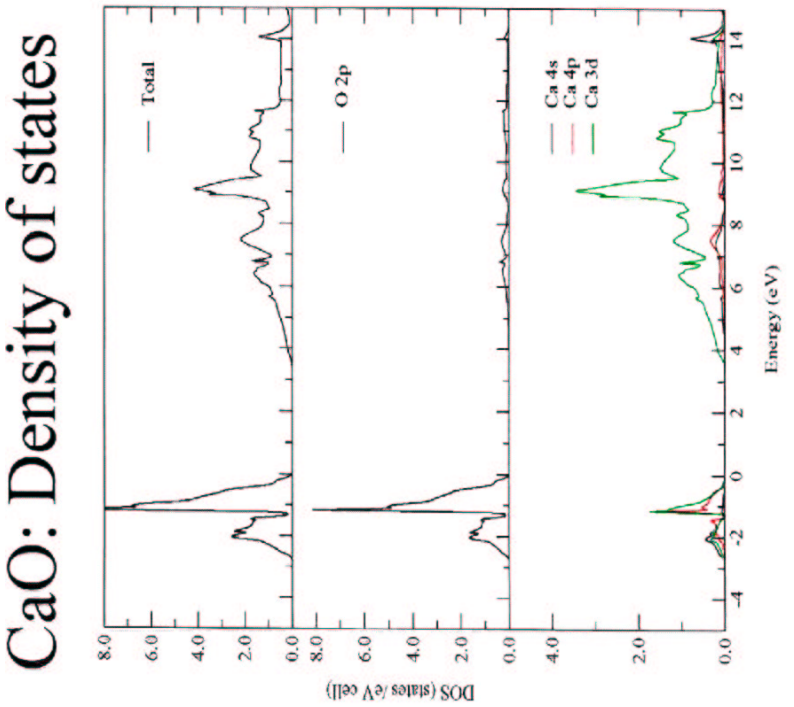
-reconstructs: e.g. octopolar at NiO (111)

-attracts charged contaminants: e.g. OH^-, I^-

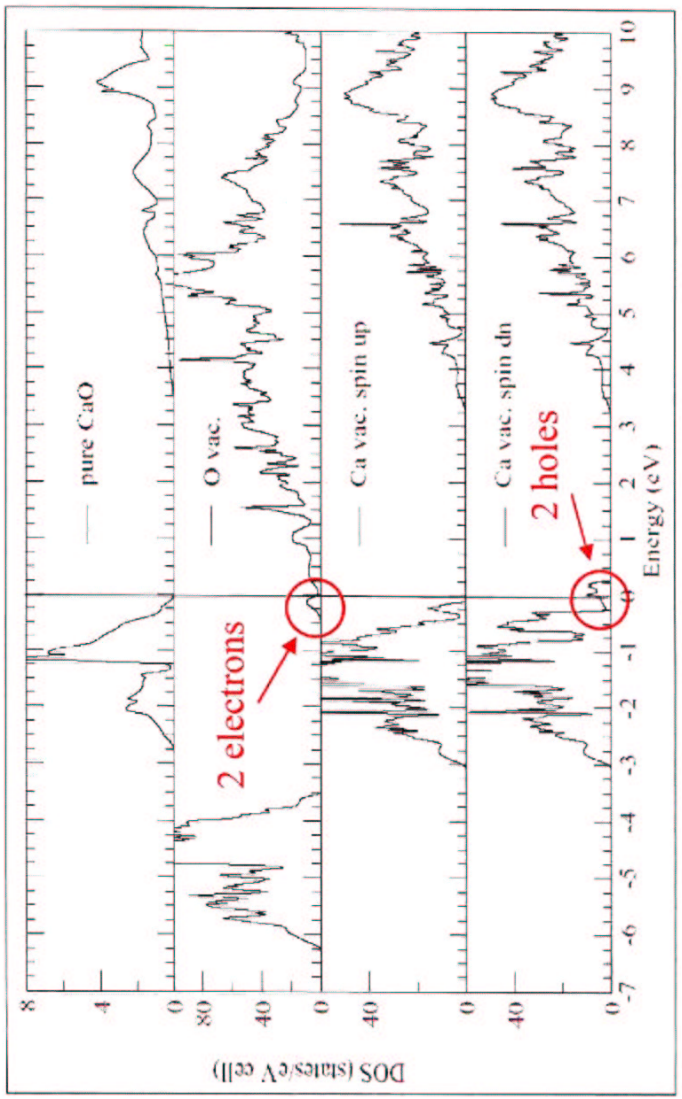
-charge redistribution: ionic charge at surface ≠ in bulk

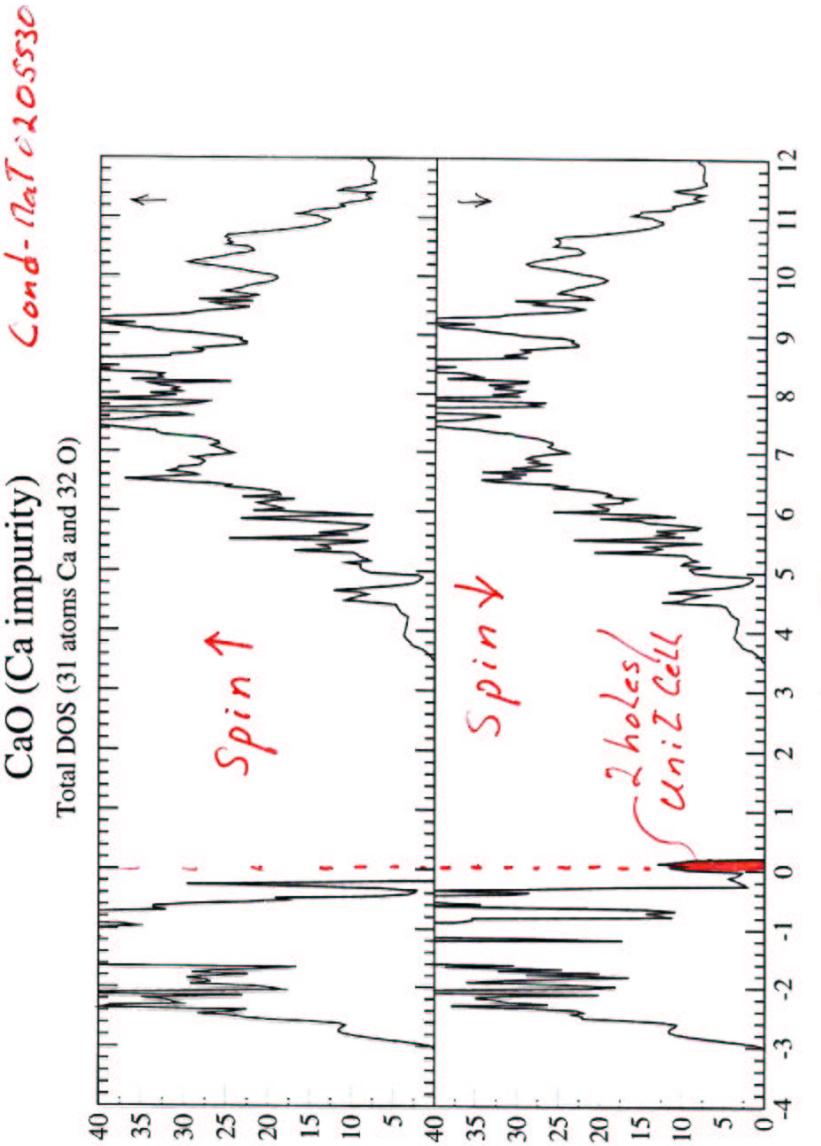



MgO [111] 4 layers Spin ↓
111 surface (4 layers of Mg and 4 layers of O; spin dn)
Mg 3s
O2p
O2p
Energy (eV)
10
5
0
-5
-10
G
K
M
GA
L
H
A


CaO: Density of states
Total
O 2p
Ca 4s
Ca 4p
Ca 3d
DOS (states /eV cell)
Energy (eV)

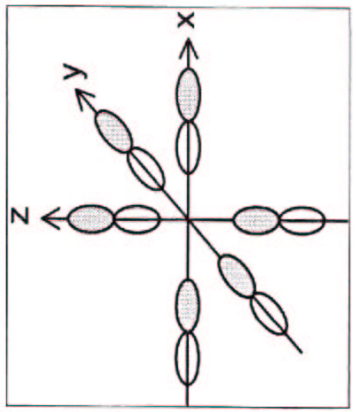
LSDA results: Total DOS



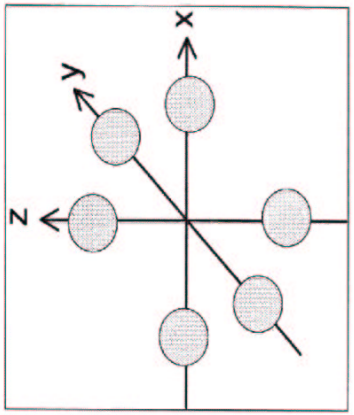


Model approach

"O_6" cluster: 2 holes in p-orbitals



"Ca_6" cluster: 2 electrons in s-orbitals



Efficiency Yunoki Cond. Mat. PRL

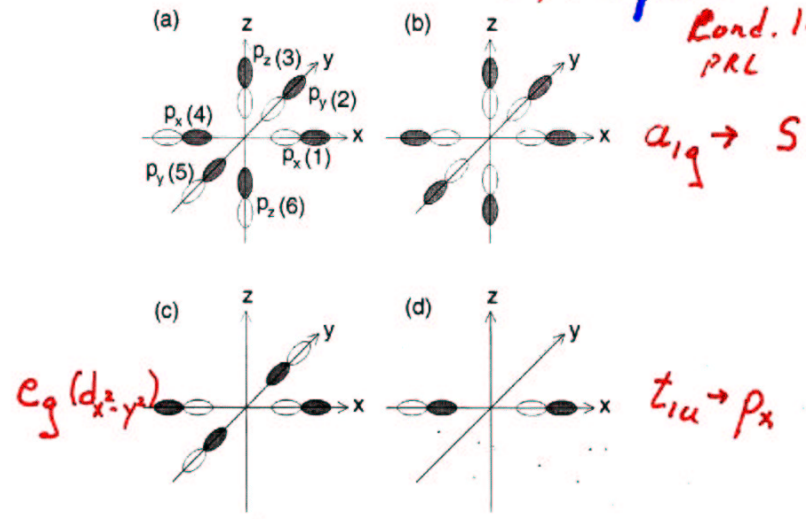


FIG. 3: (a) Definition of σ-orbitals. (b) molecular orbital with a_{1g} symmetry. (c) one of doubly degenerate molecular orbitals with e_g symmetry. (d) one of triply degenerate molecular orbitals with t_{1u} symmetry.

Ca vacancy in CaO

Definition of parameters

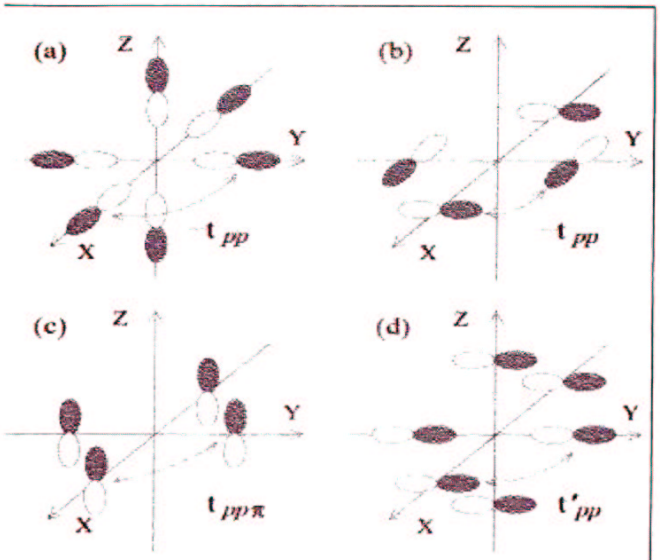


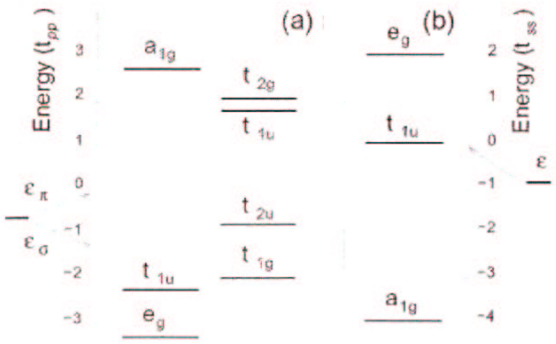
FIG. 1: An artists concept of the oxygen σ (a) and π (b,c) bonding orbitals relative to the O-vacancy bond direction surrounding a Ca vacancy. Also shown are the definitions of the hopping integrals (b) t_{pp}, (c) $t_{pp\pi}$, and (d) t'_{pp} given in terms of electrons.

$$t_{pp} = 1/2(t_{pp\sigma} - t_{pp\pi})$$

$$t'_{pp} = 1/2(t_{pp\sigma} + t_{pp\pi})$$

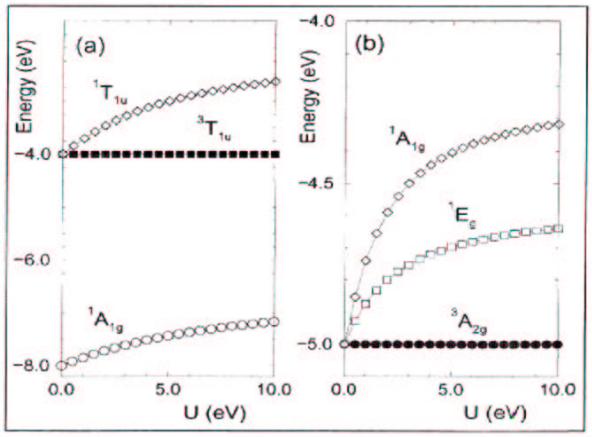
Results: Energy diagrams

Single-particle picture



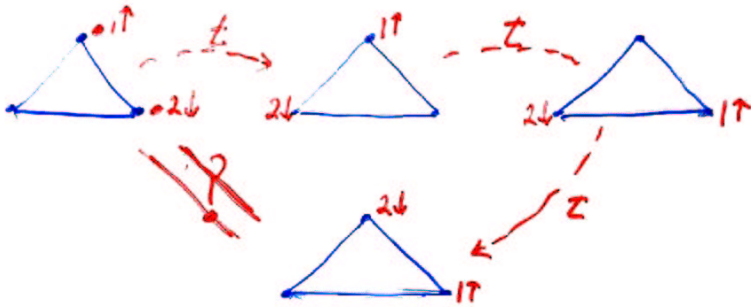
(a) HOLES in anion orbitals and
(b) ELECTRONS in cation orbitals.

Tree lowest states for two particles



(a) ELECTRONS in cation orbitals and
(b) HOLES in anion orbitals.

Magnetic Clusters



For Triplet −

For Singlet +

$$\begin{pmatrix} 0 & \Sigma & \pm\Sigma \\ \Sigma & 0 & \Sigma \\ \pm\Sigma & \Sigma & 0 \end{pmatrix}$$