Defects as a path to a new class of magnetic materials

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Towards a model Hamiltonian for V2O3

Why insulators N-H vs CT

Ionic localized Ansatz

Cuprates $d^9$ U

$S = \frac{1}{2}$

$\Delta \rightarrow d^2 \rightarrow d^1 \rightarrow d^0$

$U = E_I^{m=+} - E_A^{m=+} - E_{Pol}$

$\Delta = E_I^{\epsilon=2} - E_A^{m=+} - E_{Pol} + [V_{\eta}^{0} - V_{\eta}^{m=+}]$

$E_{Pol} = \frac{1}{2} \sum_i \alpha_i F_i^2$ [F = field on ion i; $\alpha_i$ = polarizability]
$\frac{1}{2}$ filled $s$ band $[H]

u \rightarrow \text{magnetic Insulator}

\Delta < \frac{W}{2} \rightarrow \text{Metal - Self doped}

ZSA Theory (1984)
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\textsuperscript{−}, I\textsuperscript{−}
- charge redistribution: ionic charge at surface ≠ in bulk
Towards a model Hamiltonian for V2O3

CaO: Density of states

Energy (eV)

[[Graph of CaO density of states]]

[[Graph of band structure]]

1.0 surface (4 layers of Mg and 4 layers of O)
LSDA results: Total DOS

CaO (Ca impurity)

Total DOS (CaO and 32 O)

Spin↑
Spin↓

2 holes/Uni Cell
Model approach

"O_6" cluster: 2 holes in p-orbitals

"Ca_6" cluster: 2 electrons in s-orbitals

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

\[
t_{pp} = \frac{1}{2}(t_{pp\sigma} - t_{pp\pi})
\]

\[
t_{pp\pi} = \frac{1}{2}(t_{pp\sigma} + t_{pp\pi})
\]

Results: Energy diagrams

Tree lowest states for two particles

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

Single-particle picture

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

FIG. 1: An artist's concept of the oxygen \( \sigma \) and \( \pi \) (loc) bonding orbitals relative to the O-vacancy bond direction, which contains a Ca vacancy. Also shown are the definitions of the bonding and antibonding orbitals and the \( t \) parameters given in terms of the electron affinities.
Magnetic Clusters

For Triplet $I^-$

For Singlet $I^+$

\[
\begin{pmatrix}
0 & 1 & \pm I \\
1 & 0 & \pm I \\
\pm I & \pm I & 0
\end{pmatrix}
\]