Ab initio many-body approaches to half-metallic CrO_2

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1. Introduction

Half-metallic ferromagnets are characterized by one electron theories to be complete spin-polarized at zero temperature. Among these CrO_2 shows a measured polarisation of up to *96%* (at low temperatures).
To describe the spin-polarization in CrO_2, we use finite temperature Dynamical Mean-Field Theory (DMFT) and the zero temperature Variational Cluster Approach which incorporate either local or non-local many-body interactions, respectively [1].

2. The material

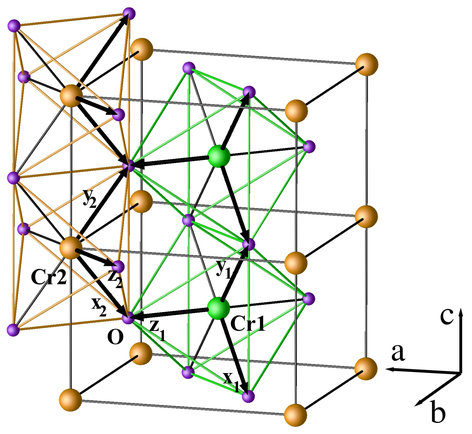

Figure 1: *CrO_2-lattice, Cr-atoms are octahedrally coordinated by oxygen atoms (purple).*

Why this compound?

CrO_2 has interesting properties including:

- being classified as an half-metallic ferromagnet, experiments show a spin-polarization of the conduction electrons larger than 90%; this is also supported/predicted by band-theory
- therefore it is an ideal candidate as a source for spin-polarized electrons for spin-injection (at low temperatures)
- several experiments (including photoemission, soft x-ray absorption, resistivity, optics) indicate the importance of electron correlations

Electronic structure and lattice

- CrO_2 forms a rutile structure with Cr-ions forming a tetragonal body-centered lattice
- the Cr-ions are in the center of the CrO_6 octahedra
- Cr^{4+} has a closed shell Ar-core and two additional $3d$-electrons
- thus, the $3d$-orbitals split into a t_{2g}-triplet and an excited e_g-doublet (which is irrelevant since there are only two $3d$-electrons)
- due to a distortion of the octahedra, the cubic symmetry is reduced to tetragonal and this fact partially lifts the degeneracy of the t_{2g}-orbitals into a d_{xy}-ground-state and d_{yz+zx}/d_{yz-zx} excited states

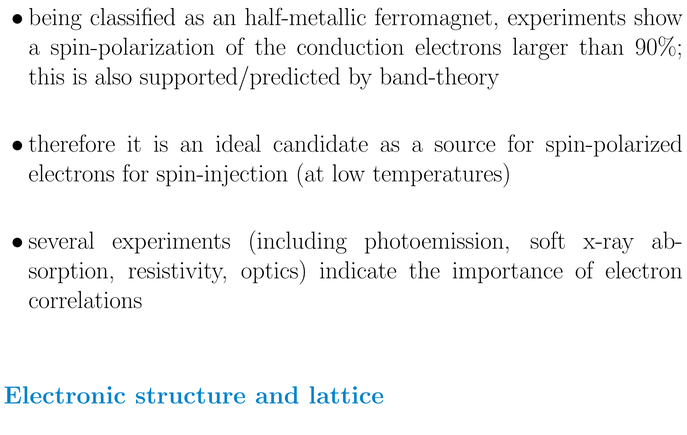

Figure 2: *Cr1 is orange, Cr2 is green, and O is purple. Red/blue indicates a positive/negative sign. Upper Panel: Cr1 t_{2g} orbitals. Lower Panel: Cr2 t_{2g} orbitals. d_{xy} (left). This orbital is singly degenerated. d_{yz-zx} (middle) and d_{yz+zx} (right). These two orbitals are nearly degenerated [2].*

3. The model

A multi-band interacting Hubbard-model

We describe multi-orbital correlation effects by:

$$H = H_0 + \frac{1}{2}\sum_{imm',\sigma} U_{mm'} n_{im\sigma} n_{im'-\sigma} + \frac{1}{2}\sum_{imm'(\neq m),\sigma} (U_{mm'} - J_{mm'}) n_{im\sigma} n_{im'\sigma} \quad (1)$$

- σ is the spin index
- m, m' are local orbitals at sites i or j
- The on-site Coulomb interactions are expressed in terms of two parameters: Coulomb interaction U and Hund's exchange J; a constrained LSDA-method was used to calculate their values [2]: $U_{mm} = U \approx 3\text{eV}$, $U_{mm'(\neq m)} = U - 2J \approx 1.2\text{eV}$, $J_{mm'} = J \approx 0.9\text{eV}$

H_0 being

$$H_0 = \sum_{m,m'}\sum_{i,j,\sigma} t^{R_i - R_j}_{m,m'} c^\dagger_{im\sigma} c_{jm'\sigma}, \quad (2)$$

where the parameters were calculated using the NMTO-method [2]; there are the on-site energy terms

$$t^{000}_{m',m} = \begin{pmatrix} -502 & 0 & 0 \\ 0 & -283 & 0 \\ 0 & 0 & -251 \end{pmatrix}, \quad (3)$$

the first nearest neighbor hoppings

$$t^{001(\text{Cr1}\to\text{Cr1})}_{m',m} = \begin{pmatrix} -119 & 0 & 0 \\ 0 & -177 & 0 \\ 0 & 0 & 196 \end{pmatrix}, \quad (4)$$

and the second nearest neighbor hoppings

$$t^{\frac{111}{222}(\text{Cr1}\to\text{Cr2})}_{m',m} = \begin{pmatrix} -4 & 0 & 0 \\ 32 & 0 & 0 \\ 0 & 142 & -204 \end{pmatrix}. \quad (5)$$

The unit is meV and *a local coordinate system* is used for each atom in the unit cell, *Cr1* and *Cr2*, as seen in Fig.1.

4. The methods

Variational Cluster Approach (VCA)

It is based on the *Self-energy-functional approach (SFA)* and the *Cluster Perturbation Theory (CPT)*.

CPT involves 3 steps:

- Divide the lattice into identical N-site clusters
- Evaluate one particle Green's function → Exact Diagonalization
- Treat intercluster hopping in perturbation theory

→ Short distance exact, long distance RPA-like

However there is no self-consistency involved → study of symmetry broken phases is not possible

Within the VCA one can

- add a single particle operator $\mathbf{h}$ to the cluster Hamiltonian and remove it perturbatively (e.g. superconducting pairing, staggered magnetic field,...)
- determine then the optimum, self-consistent $\mathbf{h}$ by the SFA grand potential Ω (M. Potthoff, Eur. Phys. J. **B** 32, 429 (2003))

Dynamical Mean-field Theory (DMFT)

To compare results we also carried out a multi-orbital realistic LSDA+DMFT calculation, which includes the full Cr d-manifold, as well as a complete *spd*-basis set for all the atoms in the unit cell.

In addition, we include both self-energy and charge self-consistency, which means that many-body effects are taken into account in the evaluation of the LSDA potentials.

5. Results

Zero temperature DOS (VCA)

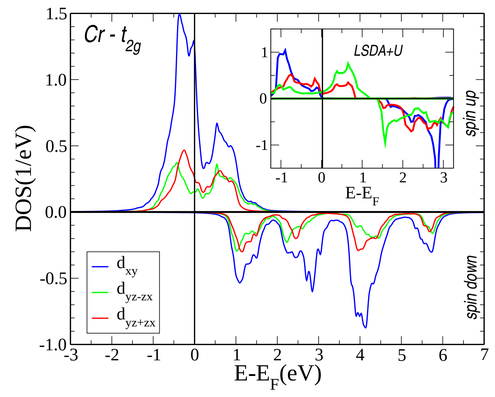

Figure 3: *Density of states obtained from VCA ($T = 0$), the inset shows the LSDA+U density of states for the same parameters*

- LSDA+U (see inset): shows a large pseudogap, with a minimum in the majority channel DOS at Fermi-energy E_F; → formed by the separation of the d_{yz+zx} and d_{yz-zx} orbitals.
- VCA results: correlation effects reduce the pseudogap feature (compared to LSDA+U) and shift it to higher energies.
- presence of a significant contribution of the d_{xy} DOS at Fermi-energy
- occupation of the d_{xy} orbital: $n^{xy} \approx 0.85$, while for the other two orbitals $n^{yz+zx} \approx 0.50$ and $n^{yz-zx} \approx 0.46$.
  → very good agreement with our LSDA+DMFT calculation

This suggests that d_{xy}-electrons should be considered as quite itinerant, though with a large effective mass, rather than as localized moments.

Finite temperature DOS (DMFT)

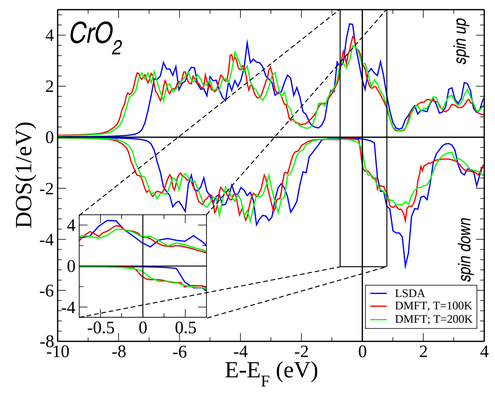

Figure 4: *Density of states obtained from DMFT, the inset shows the results for a smaller energy window around the Fermi-level.*

- the LSDA Fermi-level intersects the majority-spin bands near a local minimum and lies in the band gap of the minority spin.
- DMFT results: Finite temperatures and correlation effects close this minimum around the Fermi-level
- non-quasiparticle states at $\approx 0.25\text{eV}$, their origin are spin-polaron processes. Their tails reduce polarization at Fermi-energy! At $T = 0$ (see Fig.3) they are far away from Fermi-energy (at 1eV)

6. Conclusion

At $T = 0$ the VCA-results show that the non-quasiparticle contribution in depolarization vanishes → CrO_2 is fully polarized.
At finite temperatures DMFT-results show the presence of non-quasiparticle states, which reduce the spin-polarization in agreement with recent experiments.

We show that in contrast to previous results (LSDA+U [3] or DMFT [4]), the fully self-consistent LSDA+DMFT results yield an *itinerant* d_{xy}-orbital and a ferromagnetic phase in agreement with the non-local VCA approach.

References

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