Assessment of magnetic properties and electronic structure of SrRuO$_3$ using LDA, LDA+U and LDA+DMFT

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Outline

Experimental overview
• Ground state structural and magnetic properties
• Spectral properties
• Strain/defect sensitivity

Theoretical overview
• DFT
• DFT+X, X=U,SIC,DMFT

Why the conflicting results?

Results
• Magnetism vs. structure
• Spectral properties
• Influence of electron correlation
• Influence of spin-orbit coupling
• Orbital magnetism
Experimental Ground State Properties

Ground state structure

T<825K
   Orthorhombic
   Tilting angle (180-ϕ)/2 : 3.5°-10.5°
   Rotation angle (90-θ)/2 : 7.5°-8.5°
   Unit cell volume: 401-408 a.u.³

825K<T<945K
   Tetragonal

T>945K
   Cubic

Experimental Spectral Properties

   Okamoto, Unpublished (1999)

- PES of 100ML single crystal vs polycrystalline SrRuO$_3$


- Soft-XAS of polycrystalline SrRuO$_3$
Sensitivity to Perturbations

- Ca doping on the Sr sites - poor metal, non FL (or very low $T_{FL}$), non magn. - tilting angle increases by factor of 3
- Doping Cu on the Ru site - insulating spin-glass
- Double-perovskite, Sr$_2$RuYO$_6$ - AFM
- Universal MAE - Easy axis parallel to strain
Short notes on previous computational work

DFT
- Strong dependence on XC functional
- Strong dependence on implementation
- Tilting of orbital moments

DFT + SIC
- Unphysical localization of Ru 4d electrons
- Half metallicity
- Magnetic moments above experimental bounds
- Exchange constants in first shell overestimated

DFT + U
- Half metallicity for U>2eV
- Strong dependence on implementation - FLL DC localizes more
- Magnetic moments above experimental bounds
- Exchange constants in first shell overestimated

DFT + DMFT
- cDMFT on low-temperature structure shows high coherence temperature. Moment of 0.5\(\mu\)B for reasonable effective mass.
- Cubic phase - bad metal, non FL
Applied Computational Methods

DFT basis set FP-LMTO (RSPt)

DFT within the local density approximation, parametrizations by:
• von Barth and Hedin
• Perdew and Zunger
• Perdew and Wang

DFT within the generalized gradient approximation, parametrizations by:
• Armiento and Mattsson
• Perdew, Burke and Ernzerhof

LDA+U for U=2eV, 3eV and 4eV, J=0.6eV from cRPA calculations of Ru.
• Around mean field (AMF)
• Fully localized limit (FLL)

LDA+DMFT using spin-polarized T-matrix fluctuation exchange solver with perturbation expansion in $G_0$ and $G_{\text{Hartree-Fock}}$. Using U=2eV, 3eV and 4eV, J=0.6eV. $\Sigma_0$ double counting.
Magnetism vs. structure

Lattice constant known to be highly dependent on Ru stoichiometry

- Best performance: Functional by Armiento and Mattsson
- PBE functional severely under binding, too large moment.
Spectral properties

LDA+U(AMF) affects mainly t2g states far from $E_f$

LDA+U(FLL) affects mainly eg states far from $E_f$

Close to $E_f$
- LDA+U half metallic
- LDA+DMFT neg. spin-pol -> pos.

Influence of electron correlation
Experimental volume - von Barth Hedin LDA as ‘U=0’

- LDA+U gives too large moment
- LDA+FLEX, U=4eV too large quenching
Influence of electronic correlation

Experimental volume - electron-electron contribution to effective mass

- Experimental results ranging from $3.1 < m^* < 6.9$.
- ARPES indicates majority is from electron-boson coupling
Influence of spin-orbit coupling

Tilting of orbital moments with respect to spin-moments

Rotation of spin-moment from cartesian axes (Elk code)
Starting axis along cartesian z: 6.9° from z
Starting axis along cartesian x: 5.5° from x
Influence of spin-orbit coupling

Orbital moments and occupation

\[ w_{10} \]

\[ [eV] \]

\[ SPTF G0 \]

\[ SPTF Ghf \]

\[ LDA+U AMF \]

\[ LDA+U AMF \]
Conclusions

• Most properties of stoichiometric SrRuO₃ can be described using DFT, best functional is AM05
• Sensitivity to defects and strain indicates necessity to go beyond LDA/GGA when these phenomena are investigated
• Rotation of orbital-moment with respect to spin-moment indicates full-U is essential

• Experimental studies on spin-polarization close to Fermi-level needed to show if SrRuO₃ is negatively spin-polarized
• Experimental study of rotated orbital moments needed