Correlations in multi orbital metals: influence and fingerprints of the Hund's coupling

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Trieste SIF Congress, 25.sep`13
Outline

- Electronic correlations in 4d metals (ruthenates)
- Hund's coupling
  - Strong, filling dependent effect
  - isolated atom
  - multi orbital Kondo problem
  - Bulk – DMFT
- Photoemission on molybdates
Introduction

- Electronic correlations:
  - FL liquid behavior, if at all, below a very low $T_{FL}$
  - heavy (slow) quasiparticles (large specific heat enhancement, small quasiparticle residue)
  - Competing ground-states
- Famous examples: vanadates (3d), cuprates (3d) [heavy fermion materials (4f)]
- Interesting properties of those originate in narrow bands and large interactions ($W<<U$)
- What about 4d, where $U\sim W$? Are they boring good metals?
3d→4d; correlations don't diminish...

- SrVO$_3$ ↔ CaRuO$_3$. Paramagnetic metals, $W \sim 2.5$eV.

3d: SrVO$_3$, moderately correlated
$m^*/m_{\text{LDA}} \sim 2$ (U=4.5, J=0.6eV).
parabolic FL $< 100$K. 1 el in V $t_{2g}$

4d: CaRuO$_3$, strongly! Correlated
$m^*/m_{\text{LDA}} \sim 7$ (U=2.3, J=0.4eV)
No sign of a FL until $< 2$K. 4 el in Ru $t_{2g}$

All ruthenates have interesting props. →
Correlations are there.

![Graphs showing resistivity vs temperature for SrVO$_3$ and CaRuO$_3$.](image)

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**Table 2 Rhenenate in a nutshell**

<table>
<thead>
<tr>
<th>Compound</th>
<th>Magnetic order</th>
<th>$\gamma/\gamma_{\text{LDA}}$</th>
<th>$\rho \times T^\delta$</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sr$_2$RuO$_4$</td>
<td>PM</td>
<td>4</td>
<td>$&lt; 25$ K</td>
<td>Unconventional SC $&lt; 1.5$ K</td>
</tr>
<tr>
<td>SrRu$_3$O$_7$</td>
<td>FM $&lt; 160$ K</td>
<td>4</td>
<td>$&lt; 15$ K</td>
<td></td>
</tr>
<tr>
<td>Sr$_3$Ru$_2$O$_7$</td>
<td>PM</td>
<td>10</td>
<td>$&lt; 10$ K</td>
<td>Metamagnetic quantum-critical point and nematicity</td>
</tr>
<tr>
<td>CaRuO$_3$</td>
<td>PM</td>
<td>7</td>
<td>$T^{1.5} &gt; 2$ K</td>
<td>$\sigma = \omega^{0.5}$, $\gamma = \gamma_W + \log(T)$</td>
</tr>
<tr>
<td>Co$_2$RuO$_4$</td>
<td>AF $&lt; 110$ K</td>
<td>$x$</td>
<td>$x$</td>
<td>Insulator $&lt; 310$ K</td>
</tr>
</tbody>
</table>

*Abbreviations: AF, antiferromagnet; FM, ferromagnet; PM, paramagnet; SC, superconductor*
... but they are of different nature

- Unlike in 3d, LDA successful here (even quantitatively, ground state, Fermi surface)
- Little signatures of Hubbard bands in photoemission
• Correlations are present in ruthenates, with \( U \sim < W \), but only at small energy scales.

• Reconciled by DMFT work pointing to the **Hund's coupling** \( J \).

• Influence of \( J \) small at large energy scales, simply because \( J \ll U < W \).

• For same reason, \( J \) was not considered as important until past 5 or so years.
Multi-orbital problem

- Kinetic part + Hubbard on-site interaction + Hund's rule coupling
- Kanamori interaction Hamiltonian

\[ H_K = U \sum_m \hat{n}_{m\uparrow} \hat{n}_{m\downarrow} + U' \sum_{m \neq m'} \hat{n}_{m\uparrow} \hat{n}_{m'\downarrow} + (U' - J) \sum_{m < m', \sigma} \hat{n}_{m\sigma} \hat{n}_{m'\sigma} + \\
- J \sum_{m \neq m'} \hat{d}_{m\uparrow} \hat{d}_{m'\downarrow} \hat{d}_{m'\uparrow} \hat{d}_{m\downarrow} + J \sum_{m \neq m'} \hat{d}_{m\uparrow} \hat{d}_{m'\downarrow} \hat{d}_{m'\uparrow} \hat{d}_{m\downarrow}. \]

- \( T_{2g} \) case \( U' = U - 2J \).

\[ H_{t_{2g}} = (U - 3J) \frac{\hat{N}(\hat{N} - 1)}{2} - 2JS^2 - J\frac{L^2}{2}. \]

Energy lowered, for large spin and orbital momentum.

Achieved when els are in different orbitals and have parallel spins.
Two effects of $J$:

1. **1st modified atomic charge gap**
   - Effective interaction
     \[ U_{\text{eff}} = E(N+1) + E(N-1) - 2E(N) \]
   - $U-3J$ away from half-filling ($U_{\text{eff}}$ **diminished** by $J$)
   - $U+(M-1)J$ at half filling ($U_{\text{eff}}$ **increased** by $J$)

2. Slater all d-states Hamiltonian (# of orbs. $M=5$)

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D. Van der Mare and G. Sawatzky, PRB 37 (1988) 10674 [VdMS]
2\textsuperscript{nd} effect: J suppresses kinetic energy

- J lowers atomic degeneracy, hence prohibits some of the hopping
- Example: Create a charge excitation in half-filled two orbital problem. J allows it to move only in one of two possible ways.

- Filling dependence: → J affects deg. for n>1

<table>
<thead>
<tr>
<th>N</th>
<th>S</th>
<th>L</th>
<th>Degeneracy = (2S + 1)(2L + 1)</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>0, (6)</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>0, [15\epsilon]</td>
</tr>
<tr>
<td>1, (5)</td>
<td>1/2</td>
<td>1</td>
<td>6</td>
<td>-5J/2, [10\epsilon - 5J/2]</td>
</tr>
<tr>
<td>2, (4)</td>
<td>1</td>
<td>1</td>
<td>9</td>
<td>\epsilon - 5J, [6\epsilon - 5J]</td>
</tr>
<tr>
<td>2, (4)</td>
<td>0</td>
<td>2</td>
<td>5</td>
<td>\epsilon - 3J, [6\epsilon - 3J]</td>
</tr>
<tr>
<td>2, (4)</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>\epsilon, [6\epsilon]</td>
</tr>
<tr>
<td>3</td>
<td>3/2</td>
<td>0</td>
<td>4</td>
<td>3\epsilon - 15J/2</td>
</tr>
<tr>
<td>3</td>
<td>1/2</td>
<td>2</td>
<td>10</td>
<td>3\epsilon - 9J/2</td>
</tr>
<tr>
<td>3</td>
<td>1/2</td>
<td>1</td>
<td>6</td>
<td>3\epsilon - 5J/2</td>
</tr>
</tbody>
</table>

\(^a\text{The boxed numbers identify the ground-state multiplet and its degeneracy for J > 0.}\)
Known for a long time in impurity problems


\[
H_{DN} = \sum_{k} \sum_{m=1}^{M} \sum_{\sigma=\uparrow,\downarrow} \left( \varepsilon_k c_{km\sigma}^\dagger c_{k\sigma} + V_{km} c_{km\sigma}^\dagger d_{m\sigma} + V_{km}^* d_{m\sigma}^\dagger c_{km\sigma} \right) + \left( U - \frac{3}{2} J \right) \frac{\hat{N}_d (\hat{N}_d - 1)}{2} - J S_d^2
\]

Kondo temperature diminished by $J$


DMFT

- Both atomic, and kinetic aspect are incorporated in DMFT
- Moreover, DMFT is a self consistent impurity model, with correct atomic physics (and local itinerant physics)

These effects thus persist in bulk. Pioneering work in Hund's in DMFT:

- 2 and 3 band Hubbard: PhD thesis de'Leo'04 (SISSA)
- 2 band Hubbard: Pruschke, Bulla EPJB'05
- Iron pnictides: Haule,Kotliar, NJP'09
- 3 band Hubbard: Werner,Gull, Troyer, Millis,PRL'08
DMFT study of a 3-orbital problem (semicircular DOS)

Quasiparticle residue $Z \sim (m_{\text{LDA}}/m)$:

Consequences of changing eff. U and suppression of kinetic energy seen.

Competing effects → long tail with small $Z$ at “Janus filling” $n=2$.  

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*PRL 107, 256401 (2011)*  
PHYSICAL REVIEW LETTERS  
week ending 16 DECEMBER 2011

**Janus-Faced Influence of Hund’s Rule Coupling in Strongly Correlated Materials**

Luca de’ Medici, Jernej Mravlje, and Antoine Georges
- Bright colors = small $Z$ = strong correlations
- Bars indicate Mott insulator
- Materials placed according to specific heat enh. (if app.)

"Janus filling": ruthenates & pnictides molybdates
Absence of Hubbard bands in molybdates

At same Z, SrMoO$_3$, has smaller U~3 and wider bands, (for SrVO$_3$ U~4.5)

Electronic correlations and Hund’s coupling effects in SrMoO$_3$ revealed by photoemission spectroscopy

Photoemission of a Hund's metal

- Adjusting U in a t2g model so that $Z=0.25$ at different ratios of $J/U$. Hubbard bands are pulled in!

![Graph showing photoemission spectra for different $J/U$ ratios with $Z=0.25$.]

H. Wadati et al. arXiv:1308.4475
Kutepov et al. PRB'10
Yin et al. Nat. Materials'11
Hund's metals are correlated only at small energy scales.

Kramers-Kronig analysis
Consider quadratic $\text{Im}\Sigma = -A\omega^2$ up to $\omega_c$, 0 for $|\omega| > \omega_c$
Then $\text{Re}\Sigma = -2\omega_c A\omega + \ldots$

Hund's metals have a large curvature $A$, but that only extends to a small cutoff. Relatively small scattering at large frequencies. The slope in Re part is likewise limited only to a small cutoff.
Summary

- Hund's coupling can induce correlations in systems with broad bands and small U.

- Its effects strongly depend on filling, at “Janus” filling strongly correlated behavior with a low $T_{FL}$, large mass and weakly resolved Hubbard bands is found.

- Strong scattering at small energy scales, relatively weak scattering at strong energy scales.

Some aspects I did not cover...

- Orbital selectivity
- Optical signatures?

See:

Yin et al. Nat.Mater’11
J. Mravlje et al. PRL’11
N. Lanata et al., PRB’13
de’Medici et al. ArXiv 1212.3966

Werner et al. PRL’08 and in particular refs therein

Thank you! … and thanks to my collaborators

A. Georges, Ecole Polytechnique, College de France (Paris)

L. de' Medici (ESPCI, Paris)

and

\[ H_{CSK} = \sum_{k\sigma} \varepsilon_k c_{k\sigma}^\dagger c_{k\sigma} + J_K \sum_m \bar{S}_m \cdot \bar{\sigma}_m^c - J \left( \sum_m \bar{S}_m \right)^2 \]

- 2 S=1/2 spins coupled to 2 channels and ferromagnetic J

\[ T_K = T_{K,1} \ast \left( \frac{T_{K,1}}{J} \right) \] Jayaprakash et al, PRL, 1981, Nevidomskiy, Coleman, PRL 2009

NRG data L.de Medici et al, PRL'11
“Spin freezing”

- Strong correlations far from Mott insulator, metallic state with large mag. moment, power law freq. dependence of Matsubara self energy close to transition.

Fermi liquid recovered at very low T.

PhD thesis de'Leo '04
Unpublished data from de'Medici et al
PRL work.
Yin et al. PRB 2013
$\text{Sr}_2\text{RuO}_4$