Ruthenates as Hund's metals: overview of a success story, impurity insights & spectroscopical fingerprints

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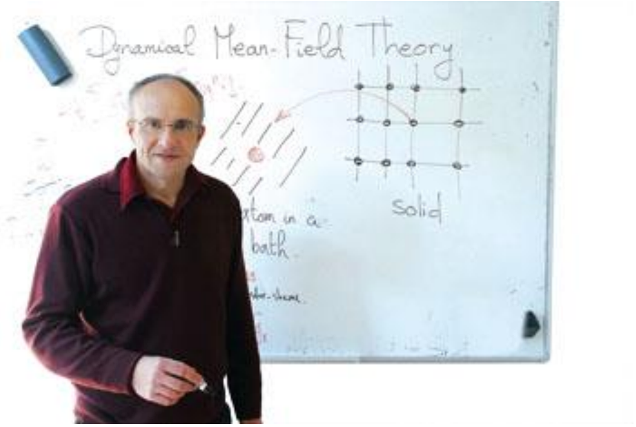
Aspen : “Quantum materials, Computations meet experiments, 12.3., 2020

IJS

People

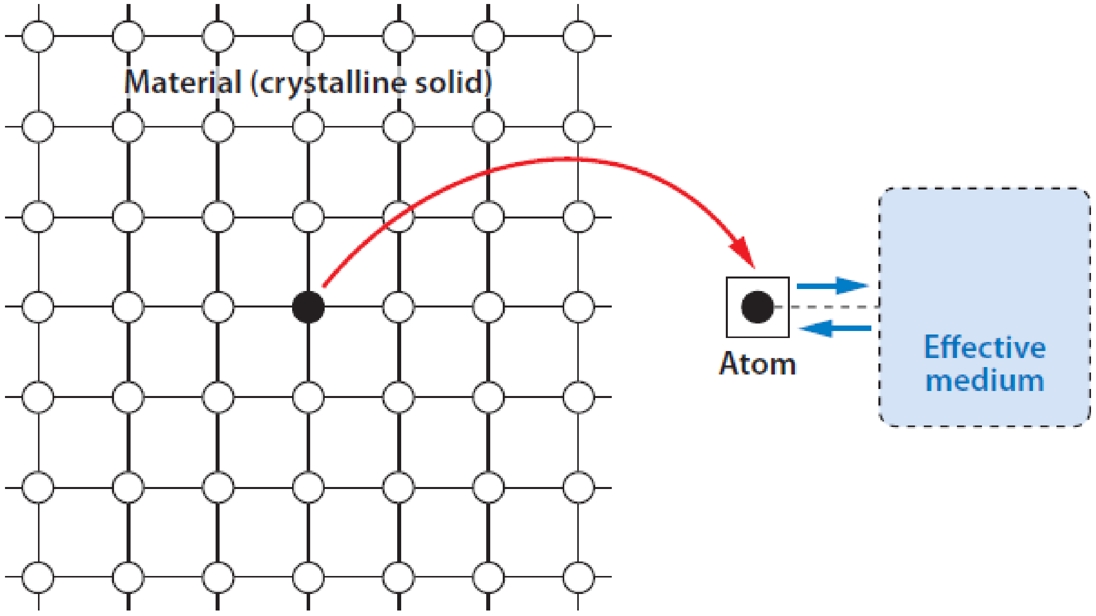
A. Georges, Flatiron,CdF,EP,UNIGE

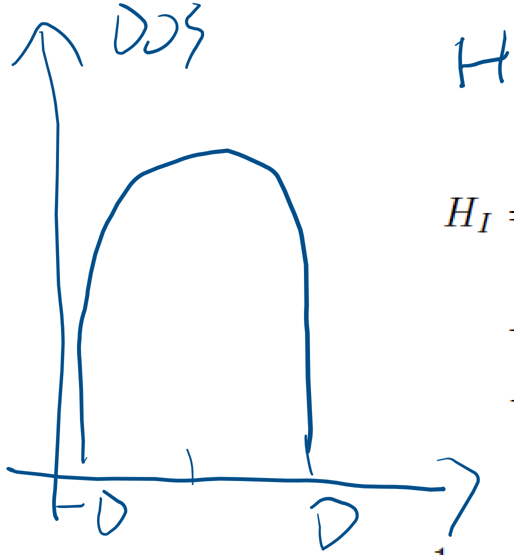


M. Aichhorn, Graz, Austria
M. Ferrero, EP, CdF, France
L. Pourovskii, EP, France
O. Parcollet, Flatiron
L. de'Medici, ESPCI
G. Kotliar, Rutgers
K. Haule, Rutgers
R. Zitko, Ljubljana
A. Horvat, Ljubljana
T. Miyake, Japan
H.T. Dang, Vietnam
A. Millis, Columbia & Flatiron

(LDA)+DMFT

- DMFT on a model (Bethe lattice
  Semicircular DOS)
- LDA+DMFT (using LDA bands)
- Solution of the impurity model
  (for featureless hybridization function)





$$H = H_{BAND} + \sum_i H_I$$

$$H_I = U \sum_m n_{m\uparrow} n_{m\downarrow} + \sum_{m<n,\sigma} [U' n_{m\sigma} n_{n\bar{\sigma}} + (U' - J) n_{m\sigma} n_{n\sigma} - J c^\dagger_{m\sigma} c_{m\bar{\sigma}} c^\dagger_{n\bar{\sigma}} c_{n\sigma}] - J \sum_{m<n} [c^\dagger_{m\uparrow} c^\dagger_{m\downarrow} c_{n\uparrow} c_{n\downarrow} + h.c.]$$

Hund

$$= \frac{1}{2}(U - 3J) N_d (N_d - 1) - 2J\mathbf{S}^2 - \frac{J}{2}\mathbf{L}^2$$

Outline

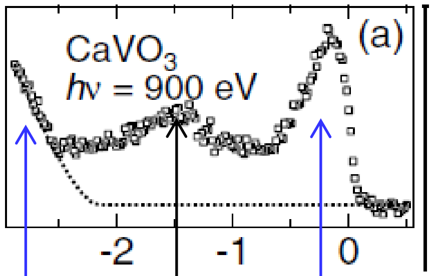
- Puzzle of correlations far from Mott (in wide band multi-orbital systems ruthenates, pnictides)
- DMFT : Hund’s metals : J & Janus behavior
- Impurity insights: separation of spin/orbital scales
- Spectral fingerprints
- NFL behavior in Kondo model

Problematics (>10 years ago)

3d -> 4d

-Orbitals become extended
-Interactions diminish
-Bands become broad

Vanadates : U ~ 4-5 eV
W ~ 2eV



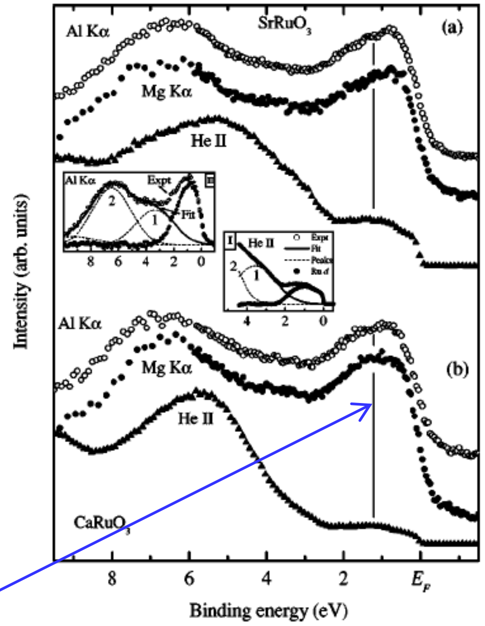
Sekiyama et al.
PRL'04

O

Hubbard band

QP

Ruthenates : U~2eV
W~2-3eV



“No” Hubbard band (in some cases)

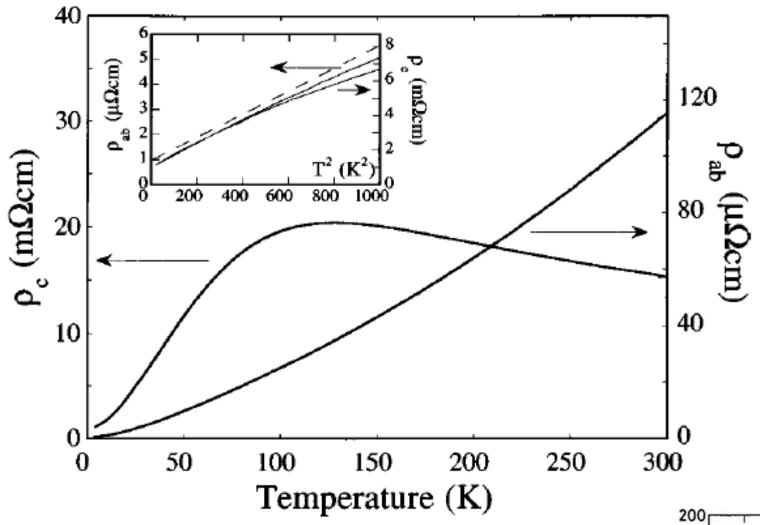
PHYSICAL REVIEW B **71**, 161102(R) (2005)

Evidence against strong correlation in 4*d* transition-metal oxides $CaRuO_3$ and $SrRuO_3$

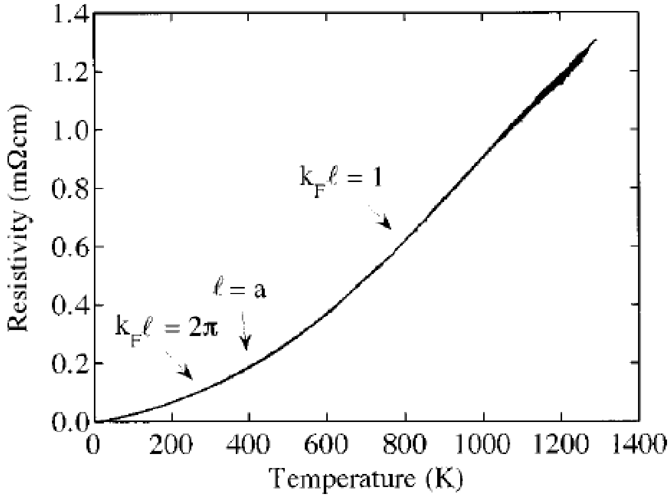
Kalobaran Maiti* and Ravi Shankar Singh

Low coherence scale in ruthenates [Sr2RuO4]

U<W, yet strong correlations : large mass, coherence-incoherence crossover at low T* & bad metal behavior

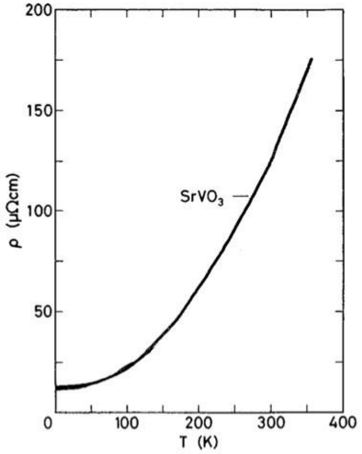


Hussey et al. PRB'98

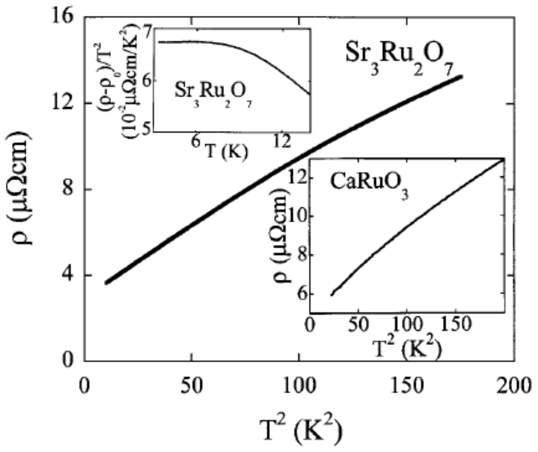


Tyler et al. PRB'98

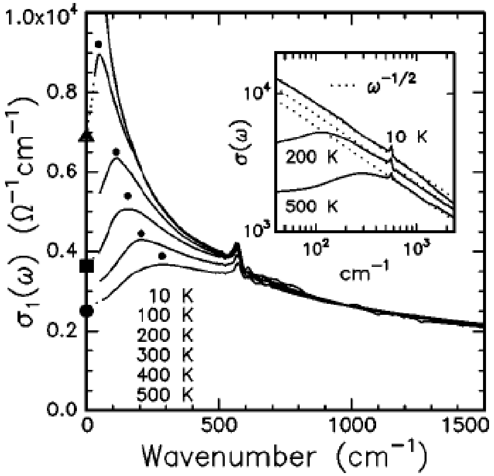
Vanadates : T^2 to a much larger T and smaller mass (at larger U/W)



Other ruthenates:also FL at a low T, and bad-metal/NFL behavior above



CaRuO$_3$



Capogna et al. PRL'02

Schneider et al.. PRL'14

Lee et al. PRB'02

Table 2 Ruthenates in a nutshell[a]

Compound	Magnetic order	γ/γ_{LDA}	$\rho \propto T^2$	Remarks
Sr_2RuO_4	PM	4	<25 K	Unconventional SC < 1.5 K
$SrRuO_3$	FM < 160 K	4	<15 K	$\sigma \propto \omega^{-0.5}$
$Sr_3Ru_2O_7$	PM	10	<10 K	Metamagnetic quantum-critical point and nematicity
$CaRuO_3$	PM	7	$T^{1.5}$ > 2 K	$\sigma \propto \omega^{-0.5}$, $\gamma = \gamma_{FL} + \log(T)$

Georges • de' Medici • Mravlje

Annu. Rev. Condens. Matter Phys. 2013. 4:137–78

Although Sr_2RuO_4 is a fully confirmed Fermi liquid at low temperatures, its properties at temperatures of approximately 30 K and above are more anomalous, raising the question of what should set such a low "crossover" scale in a material with a relatively high Fermi temperature of greater than 1000 K [12]. The behavior of $Sr_3Ru_2O_7$ was

in thin films of $CaRuO_3$ [15]. The situation in $SrRuO_3$ is more interesting still. High frequency measurements at relatively elevated temperatures suggested an anomalous $\sqrt{\omega}$ frequency dependence, leading to the proposal of a non-Fermi liquid metallic state [16]. Observation of a T^2

L. Capogna,[1] A. P. Mackenzie,[1,2] R. S. Perry,[1] S. A. Grigera,[1,2] L. M. Galvin,[1] P. Raychaudhuri,[1] and A. J. Schofield[1]
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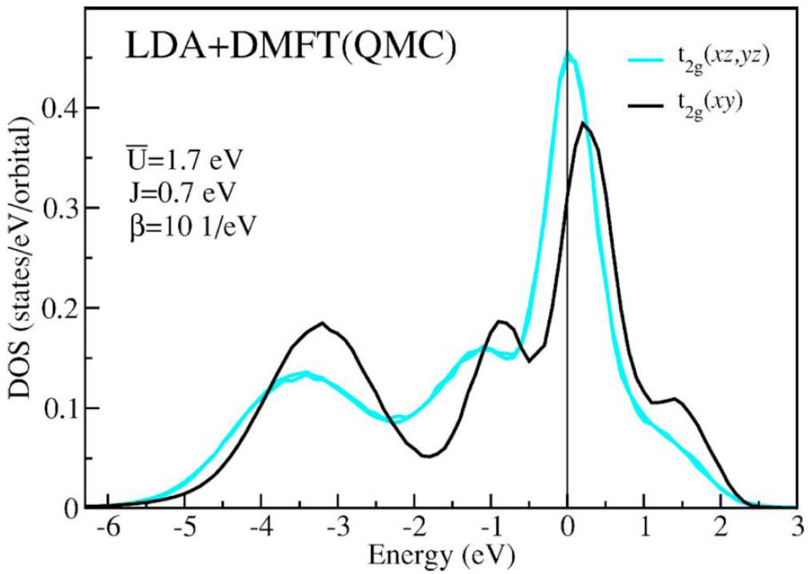
Department of Physics, Kyoto University, Kyoto 606-8502, Japan
(Received 23 April 2001; published 1 February 2002)

Presently we know how to explain this: the coherence scale is low due to Hund's rule coupling

I next quote some wrong statements in the theoretical literature at the time : illustrating that there was something qualitative to be learned…

1st LDA+DMFT on Sr_2RuO_4

PHYSICAL REVIEW B **75**, 035122 (2007)



The three-orbital, projected Hamiltonian together with the *ab initio* Coulomb interaction parameters were used as input for the QMC simulation of the effective quantum impurity problem arising in the DMFT. The simulations were performed for an inverse temperature $\beta = 10\ \mathrm{eV}^{-1}$ using 40 imaginary time slices ($\Delta\tau = 0.25$). Although the temperature chosen for the QMC calculations appears to be rather high, it is really sufficiently low, because it is much smaller than (i) the lowest atomic excitations (see the Appendix) and (ii) the characteristic low-energy scale ~ 0.5 eV obtained from the DMFT. The imaginary time QMC data were analytically continued by maximum entropy.[58] The results are shown in

[15] Z. V. Pchelkina, I. A. Nekrasov, Th. Pruschke, A. Sekiyama, S. Suga, V. I. Anisimov, and D. Vollhardt, Phys. Rev. B **75**, 035122 (2007).

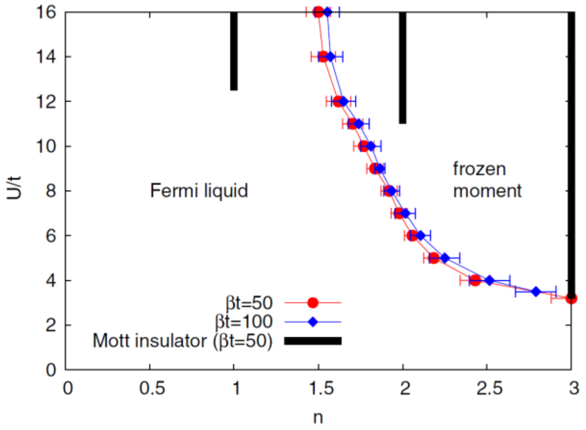
CTQMC era: one can reach low T

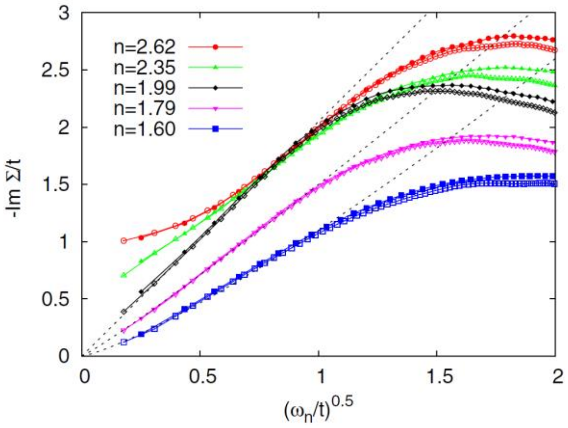
- Technical developments – (hybridization expansion continuous time Monte Carlo solvers, able to reach low T ~10K for multi-orbital)

Werner et al, PRL'06
Gull et al., RMP'11

Spin Freezing Transition and Non-Fermi-Liquid Self-Energy in a Three-Orbital Model

Philipp Werner,[1] Emanuel Gull,[2] Matthias Troyer,[2] and Andrew J. Millis[1]





In conclusion, we have shown that in a model, relevant to transition metal oxides with partly filled d-shells, with several electrons in a threefold degenerate level, an apparent spin-freezing transition occurs. While it is possible that the effects could be due to a rapid decrease of the spin coherence scale to values below the range accessible to us, the square-root self-energy and T-linear spin-spin correlation function are strong evidence for an actual $T = 0$ transition. The frozen-moment phase results from a calcu-

PRL **100**, 226402 (2008) PHYSICAL REVIEW LETTERS week ending 6 JUNE 2008

Correlated Electronic Structure of $LaO_{1-x}F_xFeAs$

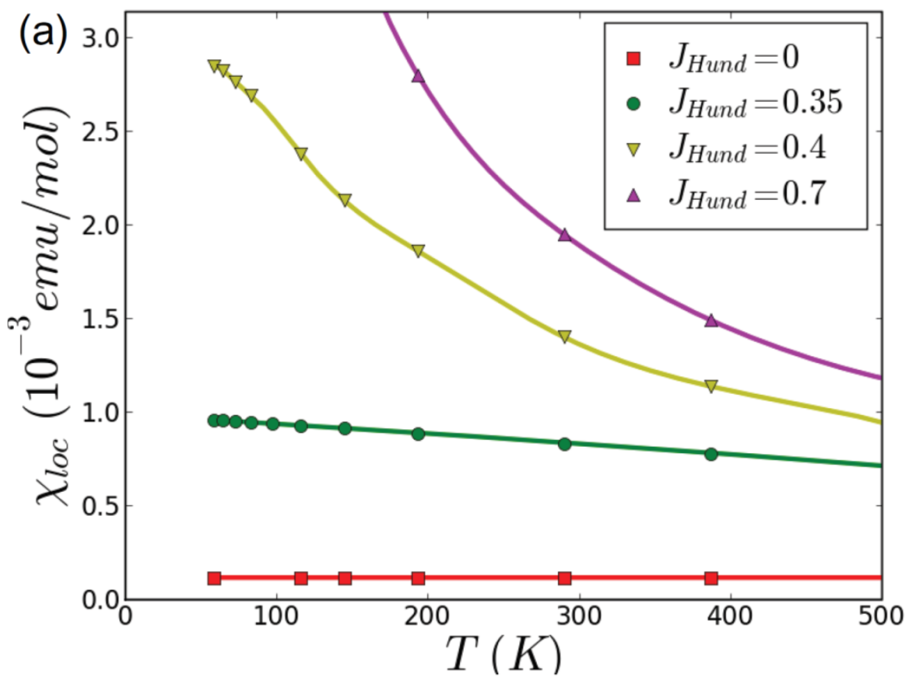
K. Haule, J. H. Shim, and G. Kotliar

In conclusion, we studied the band structure of the newly discovered superconductor $LaO_{1-x}F_xFeAs$, and we predict the orbital and momentum resolved spectral function and optical conductivity of the compound. Density functional theory predicts that a set of Fe $3d$ bands are crossing the Fermi level with no clear splitting into the e_g and t_{2g} manifold. The Coulomb correlations among the six electrons in the set of five Fe-$3d$ orbitals is strong enough to push the compound close to the metal insulator transition.

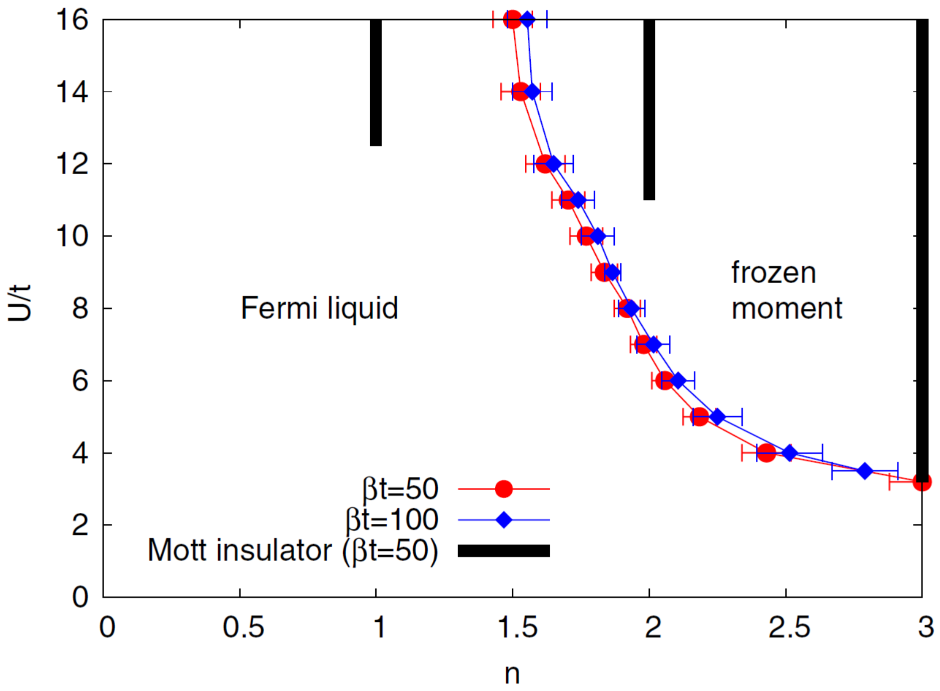
Different way of looking at ruthenates and pnictides was needed, the physics of J

Understanding the role of J

- Evolution with J first studied in pnictides (Haule&Kotliar NJP'09 (arxiv'08)) – see Kristjan's talk



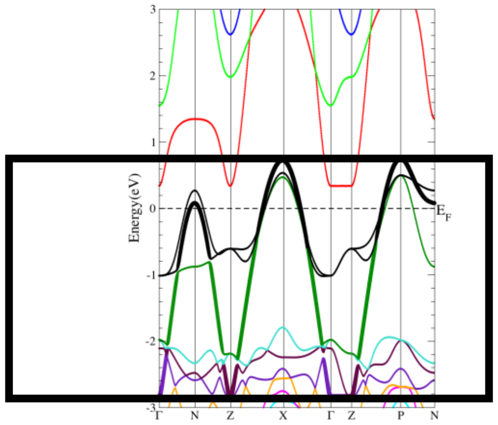
- Simultaneously – spin freezing regime (Werner,Gull,Troyer,Millis) PRL'08 (arxiv'08)) – see Philipp's talk



What about ruthenates?

Sr_2RuO_4 within LDA+DMFT

- Wannier function constructed out of t2g
- Full rotationaly invariant vertex is used
- Constrained RPA to calculate U(=2.3eV) & J
- Hybridization expansion CTQMC



$$
\begin{aligned}
H_I = U\sum_m n_{m\uparrow}n_{m\downarrow} &+ \sum_{m<n,\sigma}[U' n_{m\sigma}n_{n\bar{\sigma}} \\
&+ (U'-J)n_{m\sigma}n_{n\sigma} - J c^\dagger_{m\sigma}c_{m\bar{\sigma}}c^\dagger_{n\bar{\sigma}}c_{n\sigma}] \\
&- J\sum_{m<n}[c^\dagger_{m\uparrow}c^\dagger_{m\downarrow}c_{n\uparrow}c_{n\downarrow} + h.c.]
\end{aligned}
$$

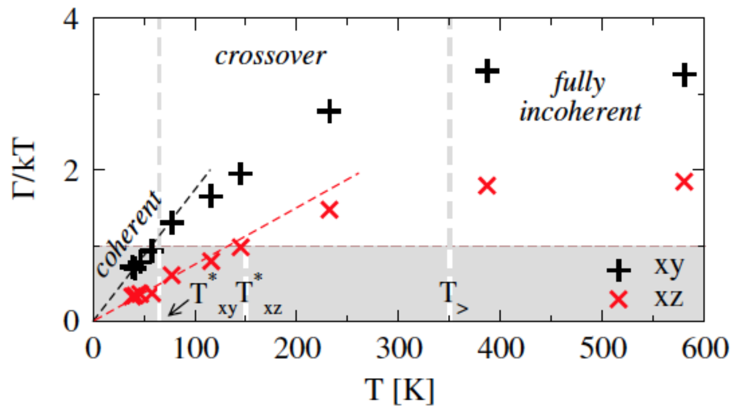
$$
\begin{aligned}
H &= (U - 3J)n(n-1)/2 - 2JS^2 - 1/2JL^2 \\
\vec{S} &= 1/2\sum\nolimits_{m\sigma\sigma'} c^\dagger_{m\sigma}\vec{\tau}c_{m\sigma'} \\
L_m &= i\sum\nolimits_{\sigma m'm''}\epsilon_{mm'm''}c^\dagger_{\sigma m'}c_{\sigma m''}
\end{aligned}
$$

Coherence scale drops due to Hund's rule coupling J

- T^* determined from T-dep of Γ=-Z ImΣ(0)
- T^* suppresed by J !

Hund's rule coupling

J [eV]	$m^*/m_{\mathrm{LDA}}\|_{xy}$	$m^*/m_{\mathrm{LDA}}\|_{xz}$	T^*_{xy} [K]	T^*_{xz} [K]	$T_>$ [K]
0.0, 0.1	1.7	1.7	>1000	>1000	>1000
0.2	2.3	2.0	300	800	>1000
0.3	3.2	2.4	100	300	500
0.4	4.5	3.3	60	150	350

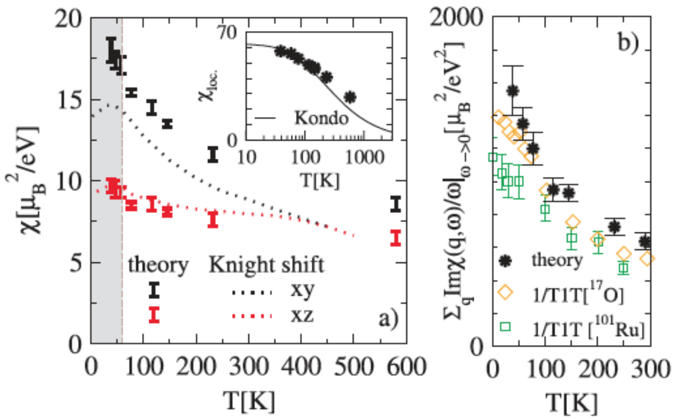


Mravlje et al. PRL'11

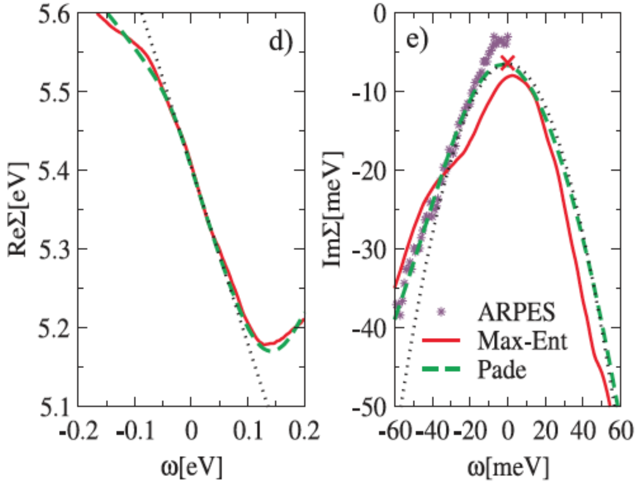
Masses in agreement with quantum oscillations & specific heat at physical value of J

Excellent agreement with exp.; Low T*,E* in experimental observables

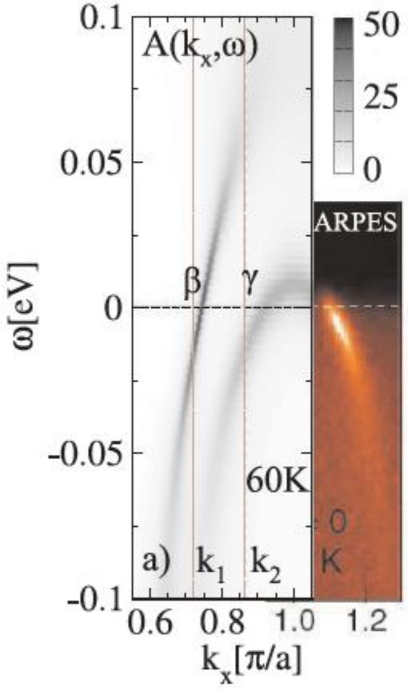
- Temperature dependences of NMR



- Low frequency kink in Σ



Excellent agreement.
Only if J is properly included.



Mravlje et al. PRL'11

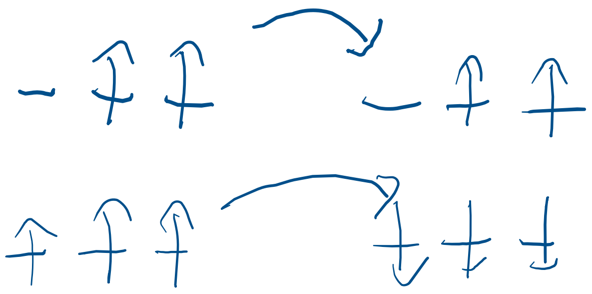
Janus behavior

Janus behavior due to Hund: away from half filling J reduces effective repulsion, yet it suppresses T*

- Effective interaction
- U_{eff}=E(N+1)+E(N-1) – 2 E(N)

- U-3J away from half-filling (eff. U **diminished** by J)

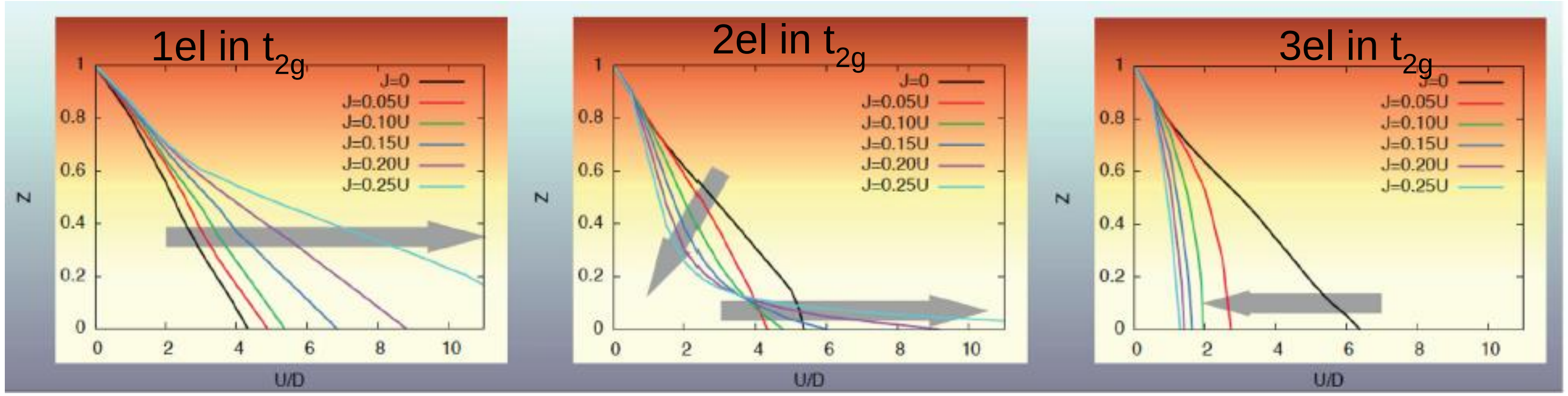
- U+2J at half filling (eff. U **increased** by J)

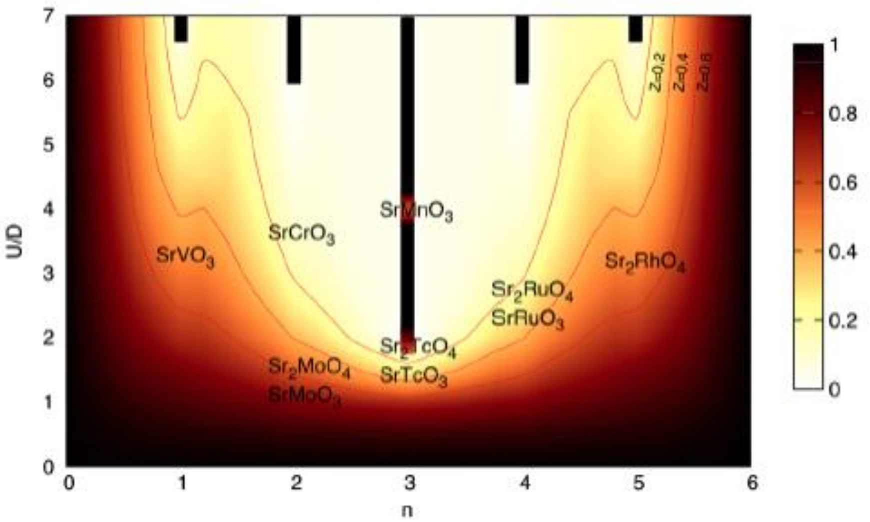


van der Marel, Sawatzky PRB’88
de’Medici PRB’11

DMFT study of three orbital Bethe lattice

- Quasiparticle weight Z

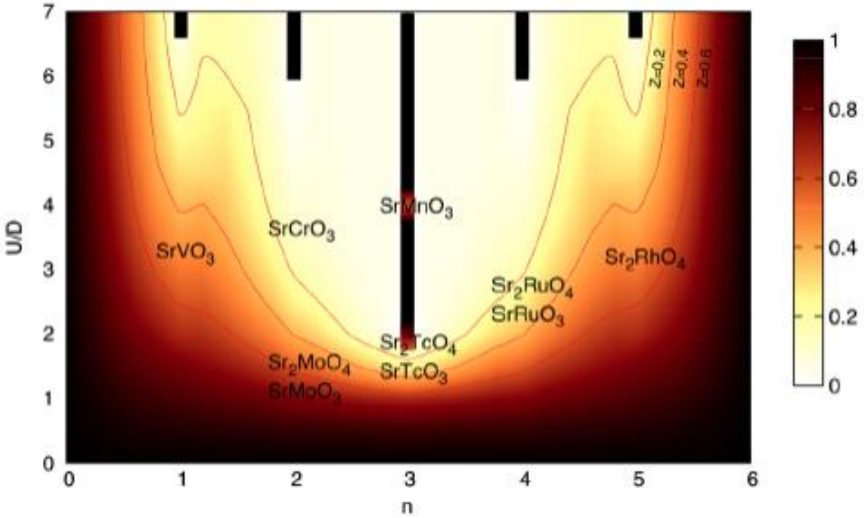




L. de'Medici, JM, A.Georges, PRL'11

Haule, Kotliar, NJP'09
Werner,Gull, Troyer,Millis PRL'08
Werner,Gull, Millis, PRB'09
Georges, de'Medici, Mravlje, Annu Rev CM'13
Yin, Haule, Kotliar,PRB'13
Aron, Kotliar PRB'15
Fanfarillo, Bascones PRB'15 ...

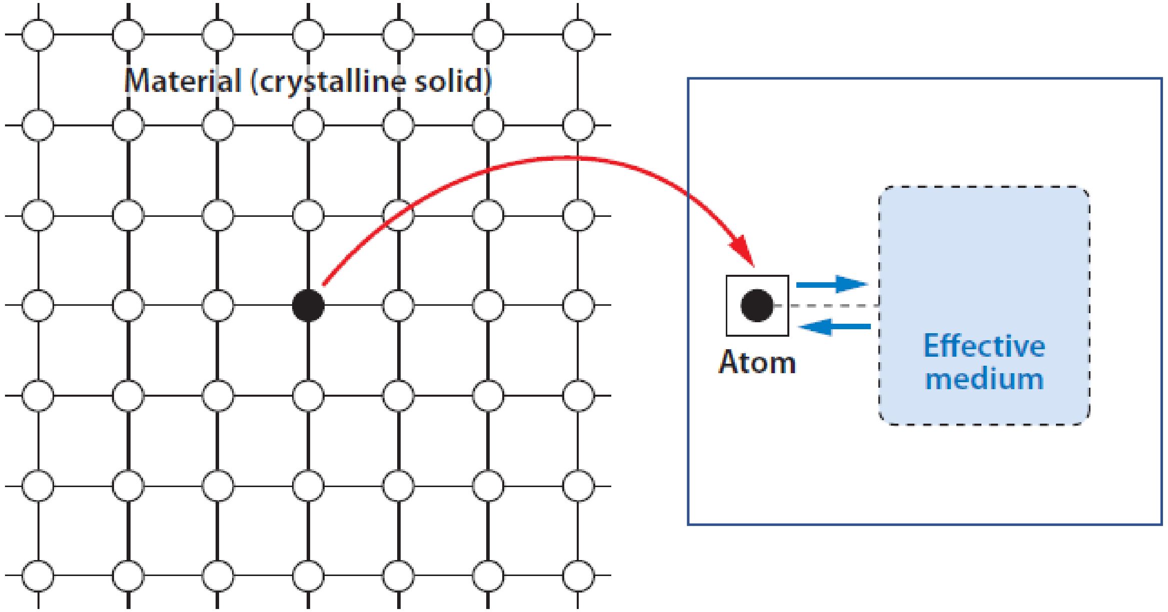
- Why such behavior?
- A fruitful line of thinking is to consider it as a doped half filled Mott ins.



de'Medici, Giovannetti, Capone, PRL'14
de'Medici, Hasan, Capone, Dai, PRL'09
Ishida, Liebsch, PRB'10
Misawa,Nakamura, Imada, PRL'12

- Here, I will be discussing insights from impurity models, instead

(LDA)+DMFT



Insights from impurity problem: Js small or even ferromagnetic

- Schrieffer-Wolff $H_{\rm K} = -P_n H_{\rm hyb}\left(\sum_a \frac{P^a_{n+1}}{\Delta E^a_{n+1}} + \sum_b \frac{P^b_{n-1}}{\Delta E^b_{n-1}}\right) H_{\rm hyb} P_n$

Kanamori: SU(2) angular momentum sym.

$$\boxed{H_{\rm imp} = \frac{1}{2}(U-3J)N_d(N_d-1) - 2J\mathbf{S}^2 - \frac{J}{2}\mathbf{L}^2}$$

For Nd=2 → S=1, L=1

$$H_K = J_p N_f + J_s \mathbf{S}\cdot\mathbf{s} + J_l \mathbf{L}\cdot\mathbf{l} + J_q \mathbf{Q}\cdot\mathbf{q} + J_{ls}(\mathbf{L}\otimes\mathbf{S})\cdot(\mathbf{l}\otimes\mathbf{s}) + J_{qs}(\mathbf{Q}\otimes\mathbf{S})\cdot(\mathbf{q}\otimes\mathbf{s})$$

$$Q^{bc}_{i,j} = \frac{1}{2}\left(L^b_{i,m}L^c_{m,j} + L^c_{i,m}L^b_{m,j}\right) - \frac{2}{3}\delta_{b,c}\delta_{i,j}$$
$$\mathrm{Tr}(Q^\alpha Q^\beta) = 2\delta_{\alpha,\beta}$$

Horvat, Žitko, Mravlje PRB’16

Dworin-Narath: SU(3) angular momentum sym.

$$\boxed{H_{\rm imp} = \frac{1}{2}(U-3J)N_d(N_d-1) - 2J\mathbf{S}^2}$$

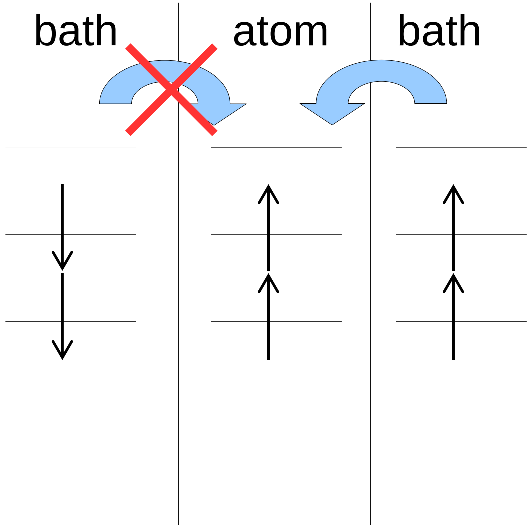
For Nd=2 → S=1, T fund.rep of SU(3)

$$H_K^{DN} = J_p N_f + J_s \mathbf{S}\cdot\mathbf{s} + J_t \mathbf{T}\cdot\mathbf{t} + J_{ts}(\mathbf{T}\otimes\mathbf{S})\cdot(\mathbf{t}\otimes\mathbf{s})$$

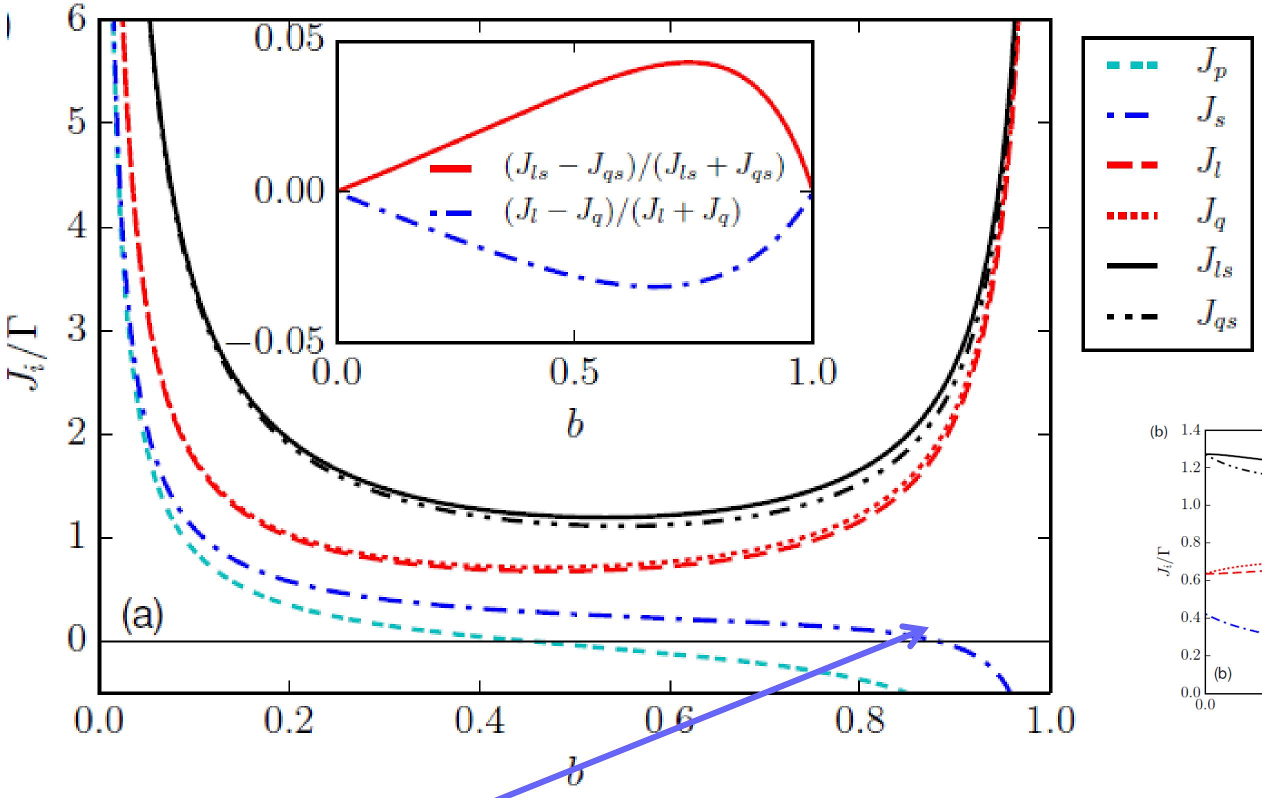
Yin, Haule, Kotliar PRB’12
Aron, Kotliar PRB’15
Stadler et al. PRL’15

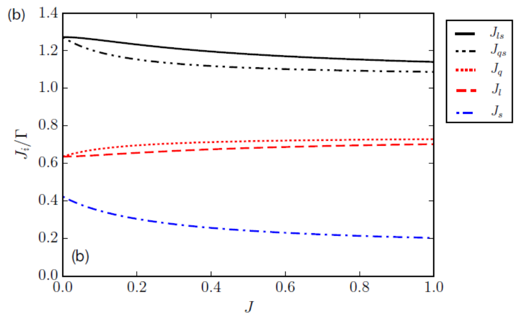
Js small or even ferromagnetic!
(ferromagnetic Kondo leads to unscreened moments)

- Why ferromagnetic? Fluctuations to N=3 (half-filled) states prefer ferromagnetic arrangement [in contrast to single-orbital!]



Kondo coupling constants

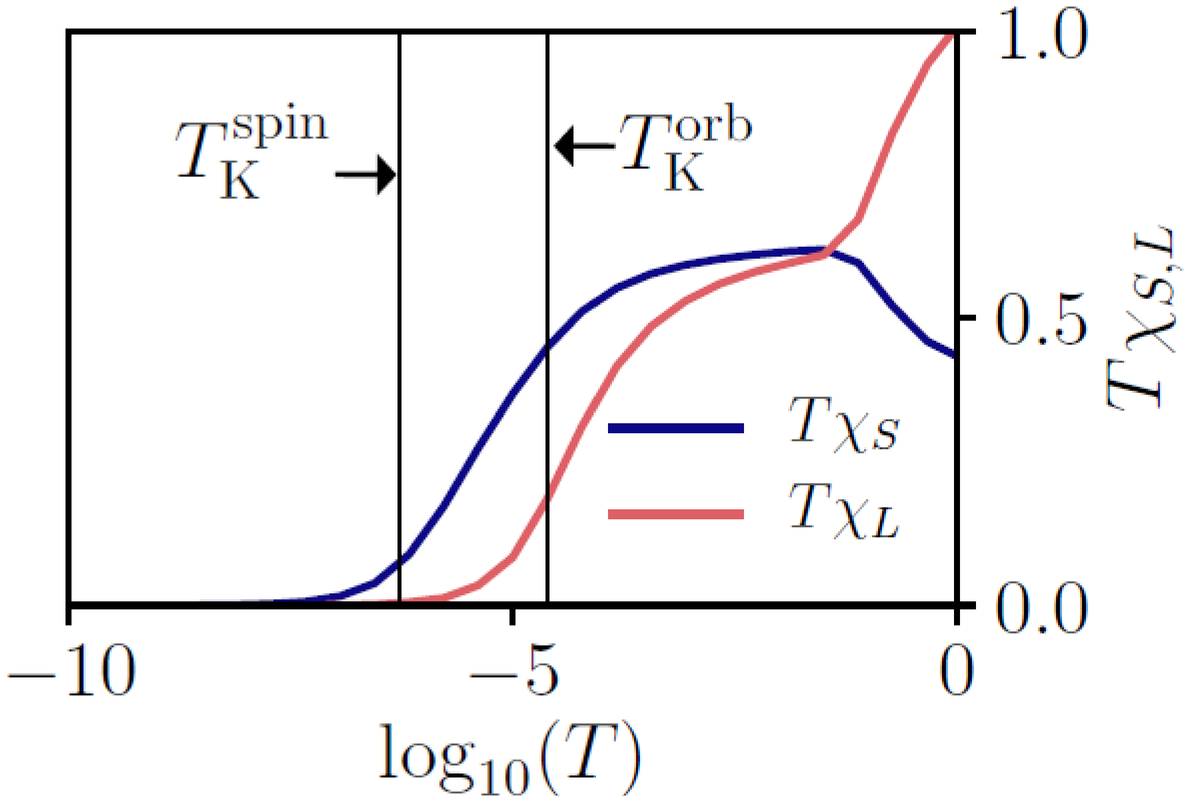




- Small or even ferromagnetic J_s
- Small splittings between quadrupole and orbital terms.
- Mixed terms are strongest.

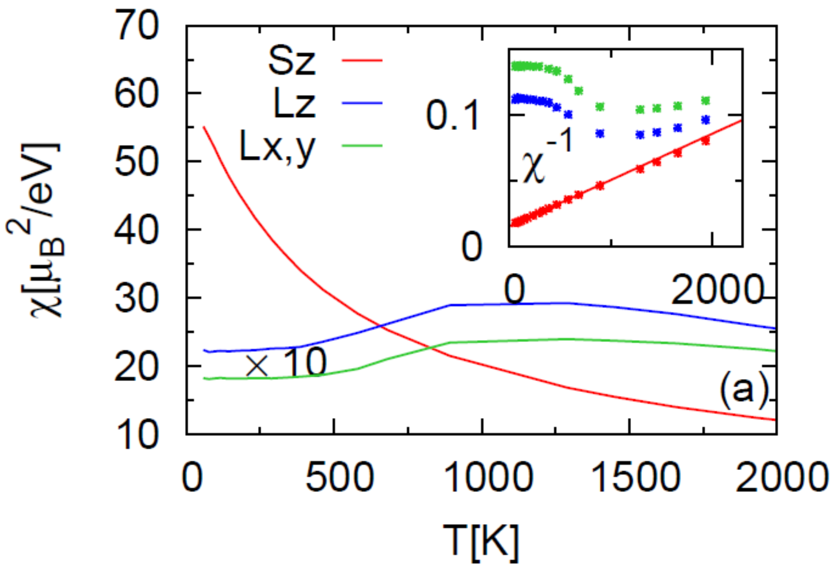
Yin, Haule, Kotliar PRB’12
Aron, Kotliar PRB’15
Stadler et al. PRL’15
Horvat, Mravlje, Zitko PRB’16

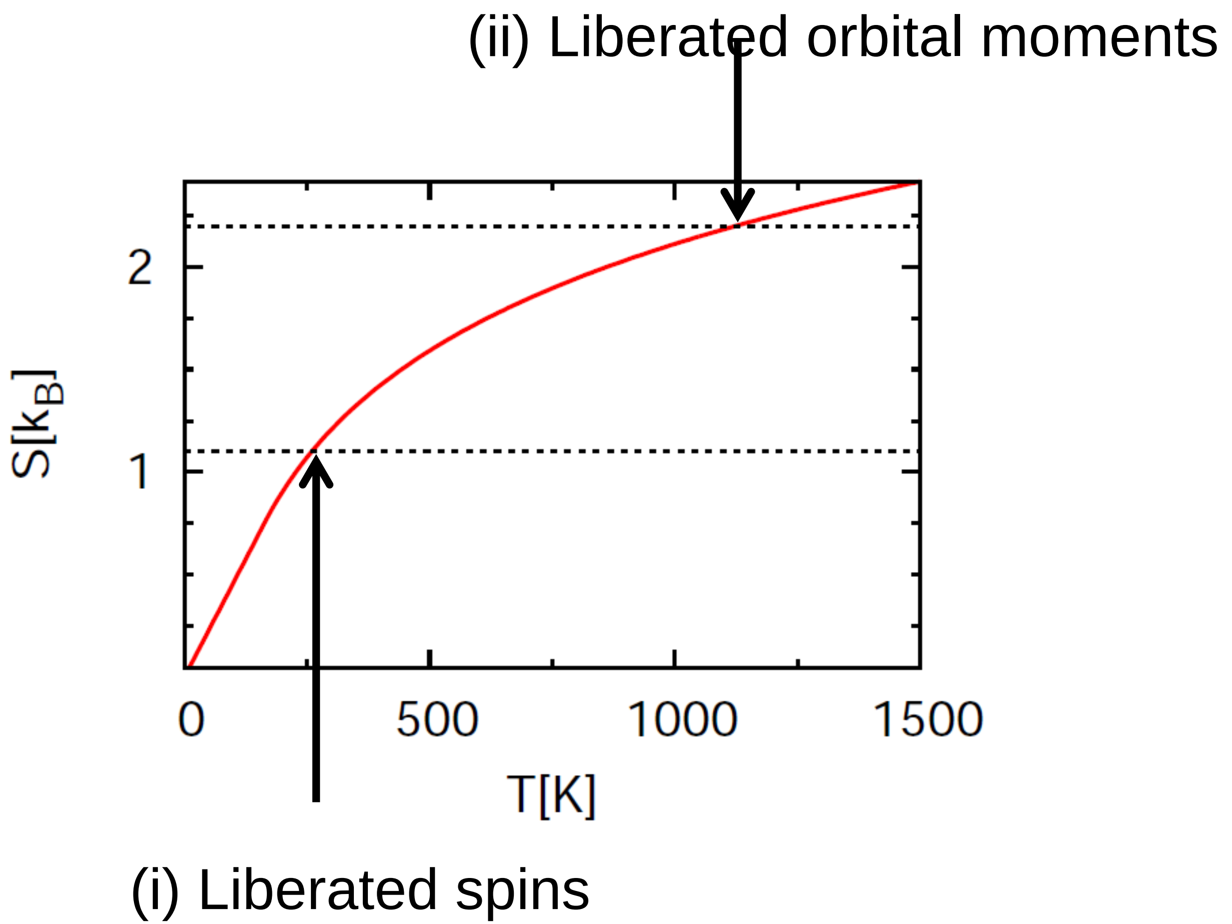
NRG calculation of 3 orbital Kanamori impurity: two scales



Horvat, Zitko, JM PRB'16
Aron, Kotliar PRB'15
Stadler et al. PRL'15

Returning to Sr_2RuO_4: Two-stage decoherence

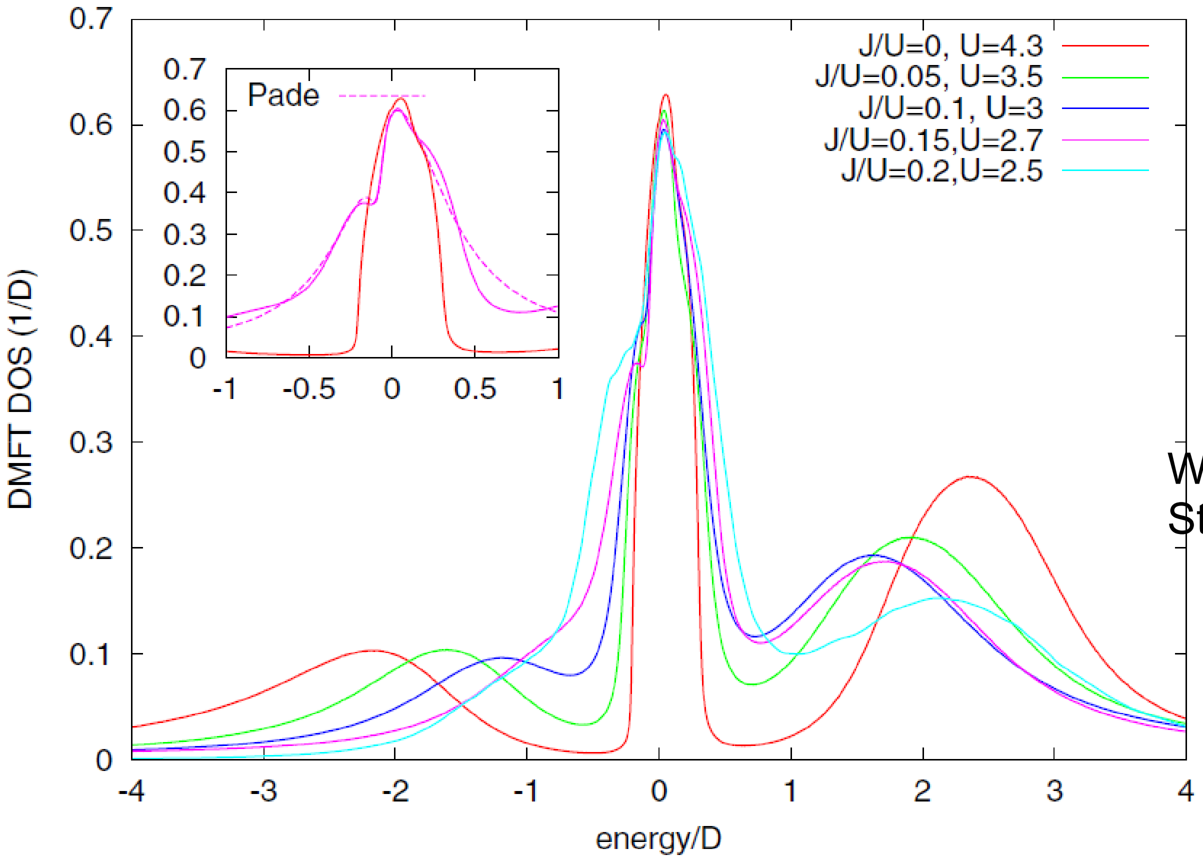




Mravlje, Georges, PRL’16

Spectroscopic fingerprints of Hund's metals (DMFT for a semicircular DOS)

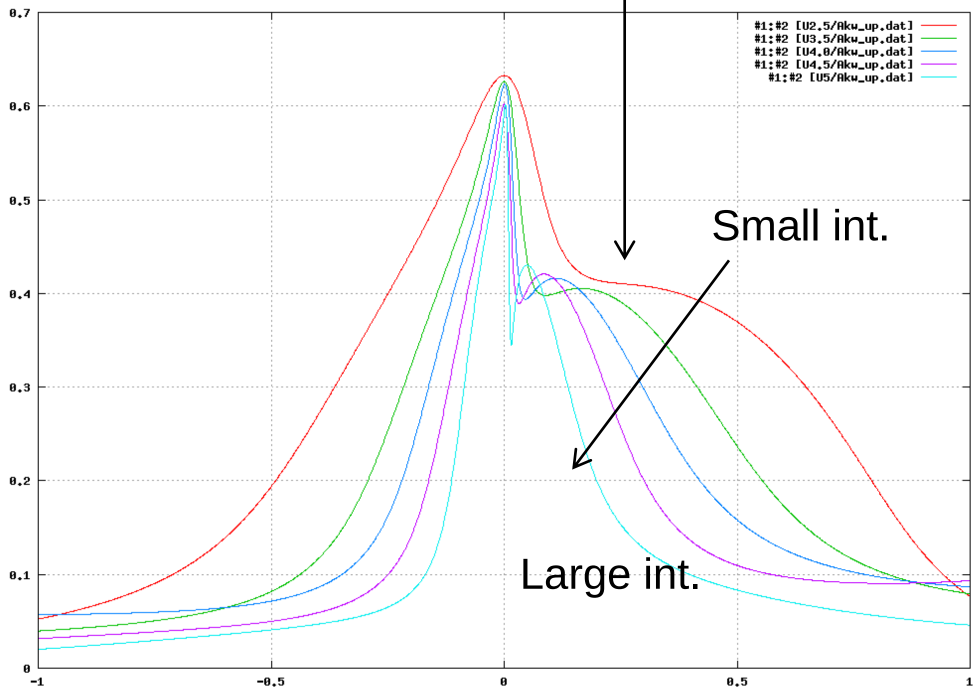
- Two-stage screening has consequence: Quasiparticle peak has structure

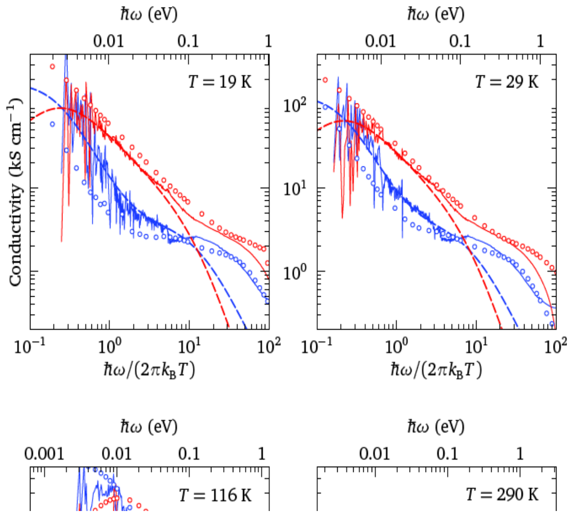


Wadati, JM et al. PRB'14.
Stadler et al.PRL'15, Ann. Phys.'19

Quasiparticle part of the spectra;DMFT semicirc. J/U=1/6 (n=4)

Frequency of the feature given by T_K^{orb}





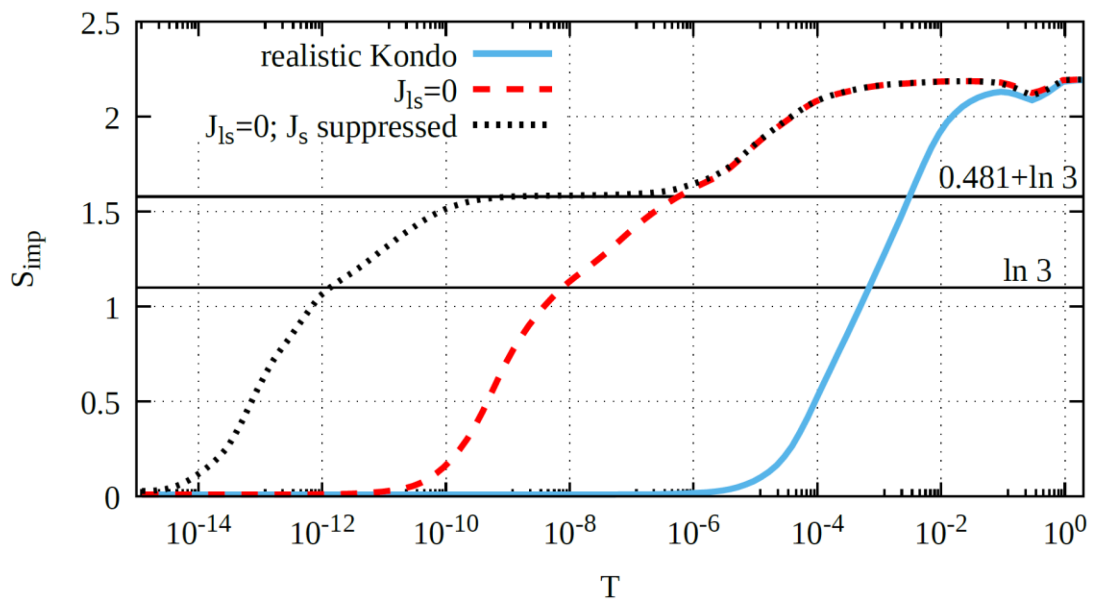
Observed in optical study of Sr_2RuO_4
Stricker,JM et al. PRL’14

Investigating incoherent regime by suppressing mixed terms : two-channel SU(3) overscreened Kondo

$$H_{\mathrm{imp}} = \frac{1}{2}\left(U - 3J\right) N_d (N_d - 1) - 2J\mathbf{S}^2$$

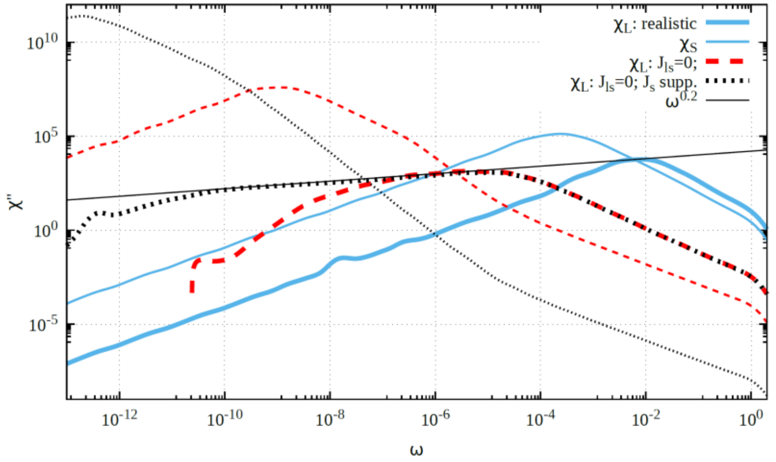
For Nd=2 → S=1, T fund.rep of SU(3)

$$H_K^{DN} = J_p N_f + J_s \mathbf{S}\cdot\mathbf{s} + \boxed{J_t \mathbf{T}\cdot\mathbf{t}} + J_{ts}(\mathbf{T}\otimes\mathbf{S})\cdot(\mathbf{t}\otimes\mathbf{s})$$

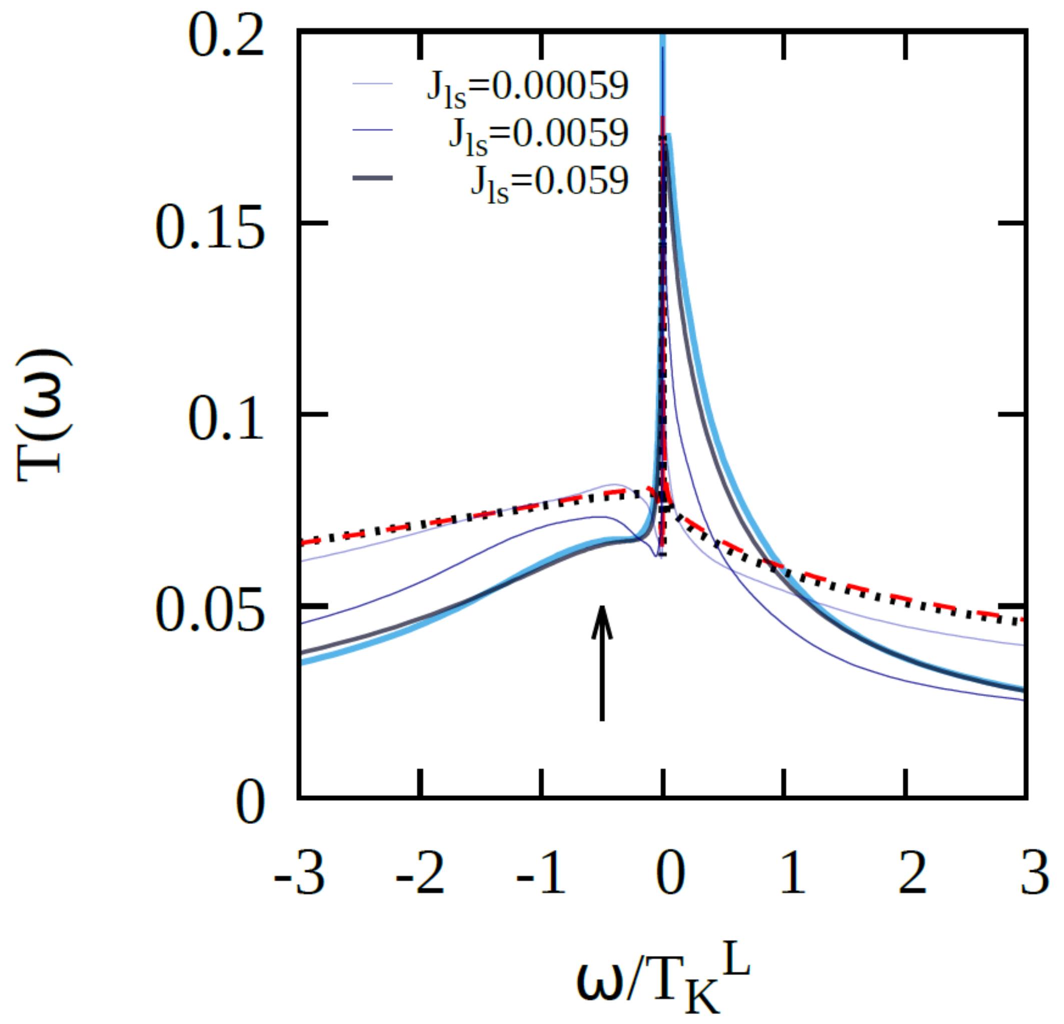


Retaining just **T.t** --- SU(3) object screned by two SU(3) channels (spin up & down)

Yin, Haule, Kotliar PRB’12
Aron, Kotliar PRB’15
Stadler et al. PRL’15
Horvat, Zitko, Mravlje, arxiv’19
More recent work von Delft, Kotliar → see talk by Jan



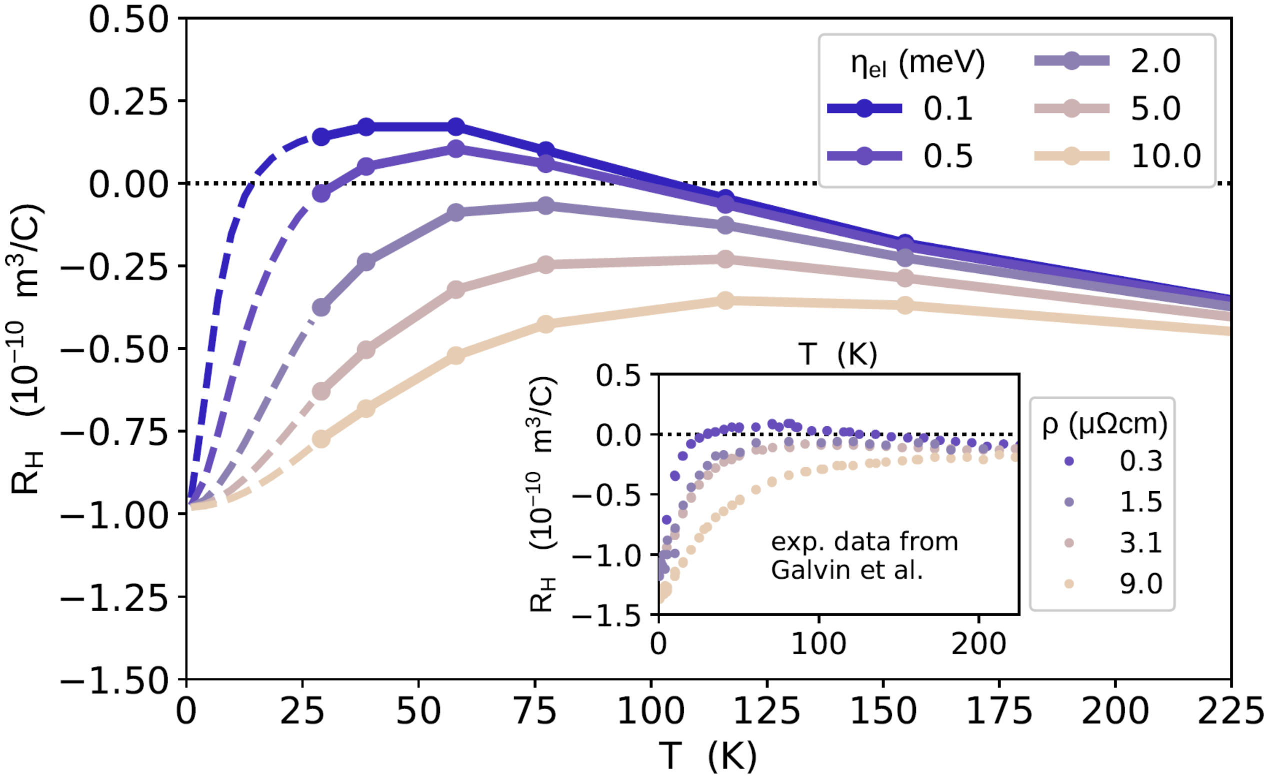
Spectral function: evolution with J_{ls}

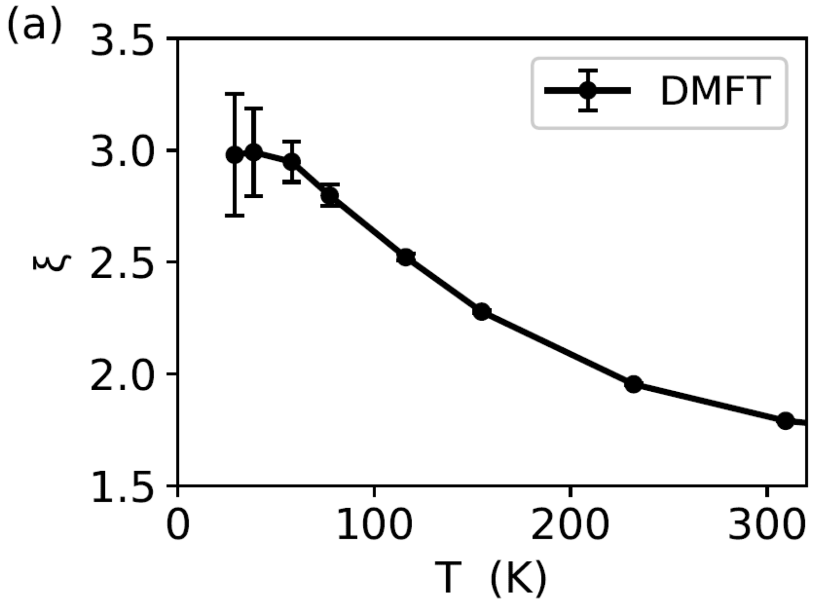


More recent works on Sr_2RuO_4: computations meet each other & further experiments

Strand, Zingl, …, Georges PRB’19 successful calculation of chi(k,omega)
Linden, Zingl, …, Schollwoeck PRB’20: DMRG calculation with included SOC
Kugler, Zingl, …, Georges, PRL’20: NRG LDA+DMFT calculation (confirming the picture + qp interactions)
Zingl, JM, Aichhorn, Parcollet, Georges, npj Quantum Materials ’19 (Hall effect)
Barber, Lechermann, …,Mazin, PRB’19 calculation of critical strain under uniaxial pressure
...

Hall (agrees with exp., but some deviations at higher T)





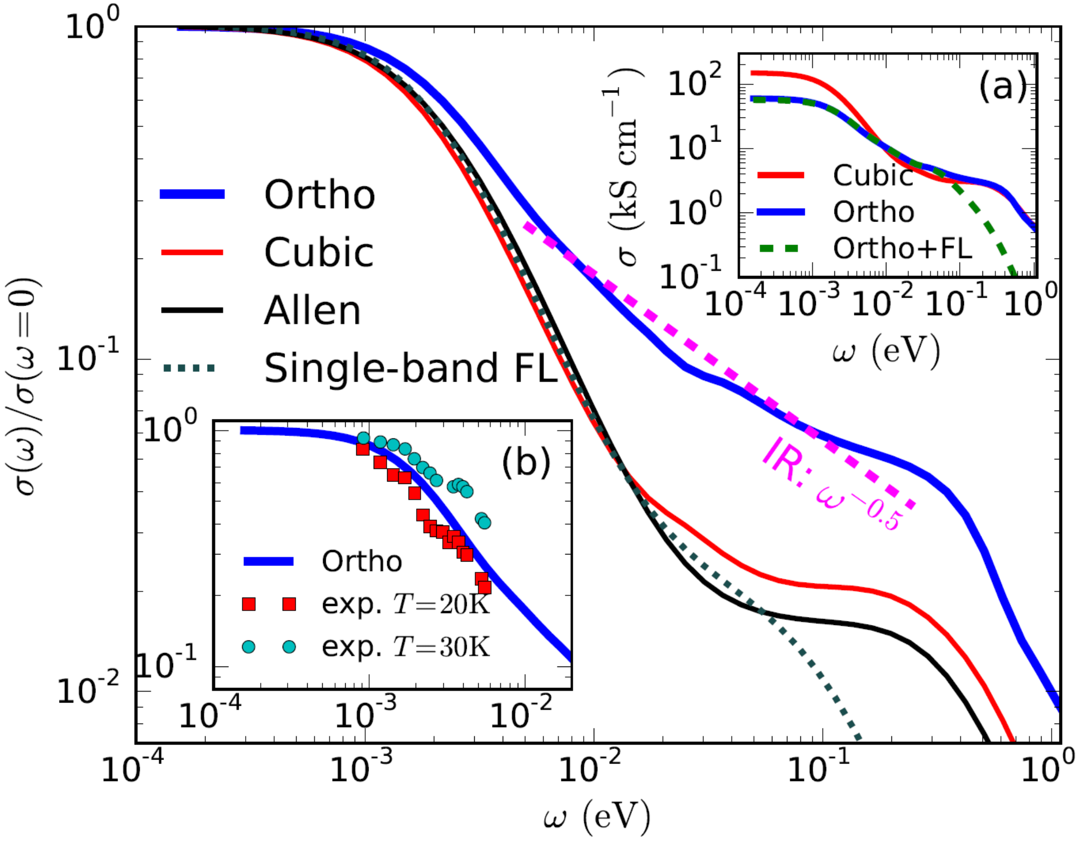
Zingl, JM, Aichhorn, Parcollet, Georges, npj Quantum Materials ’19,

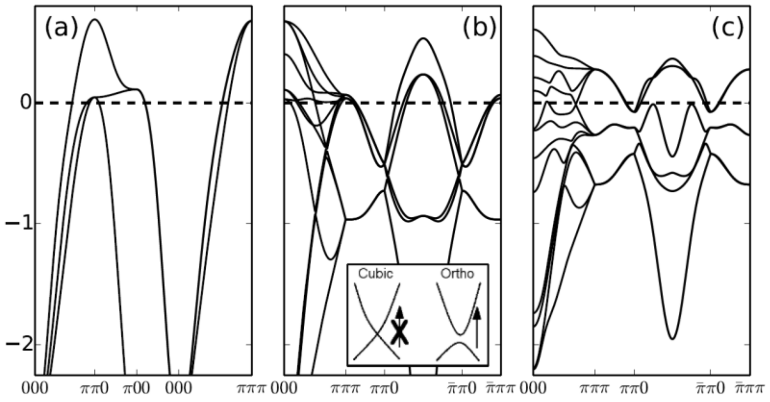
Summary

- Ruthenates are Hund's metals and are described by DMFT well
- Hund's metals have a low coherence scale due to J far from Mott
- There is intermediate T state with fluctuating spins and "quenched" orbitals
- Hund's metals have a spectroscopical fingerprint: a shoulder in the quasiparticle peak
- Idealized variant of this state: two-channel overscreened SU(3) (more : next talk)

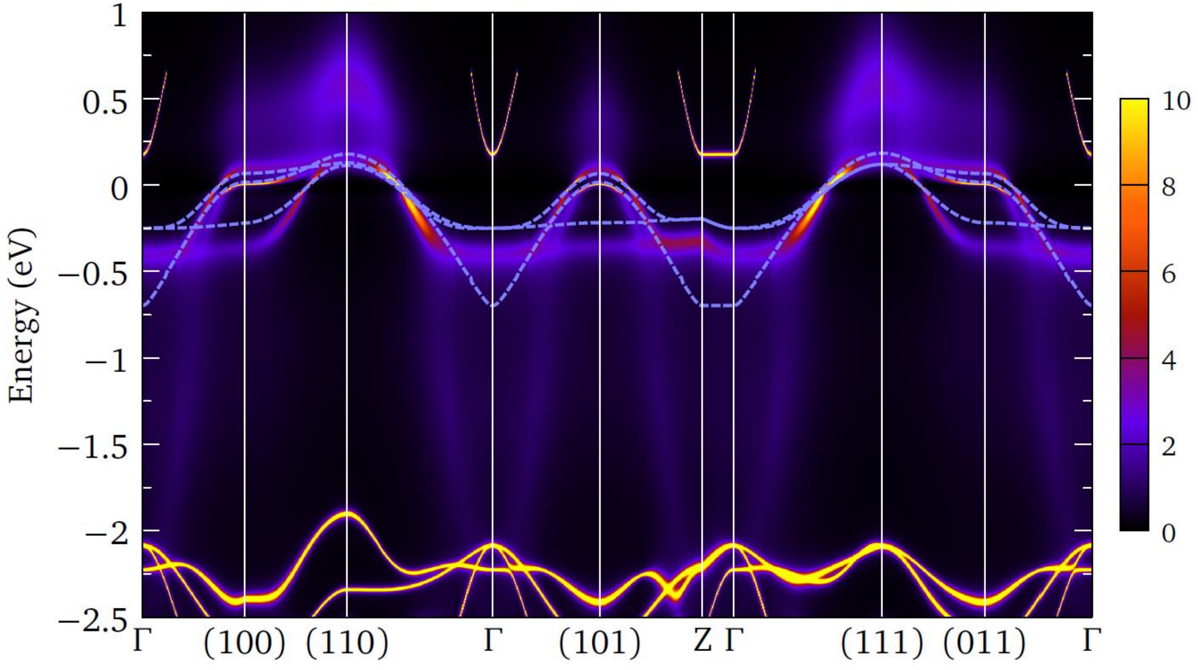
Thank you!

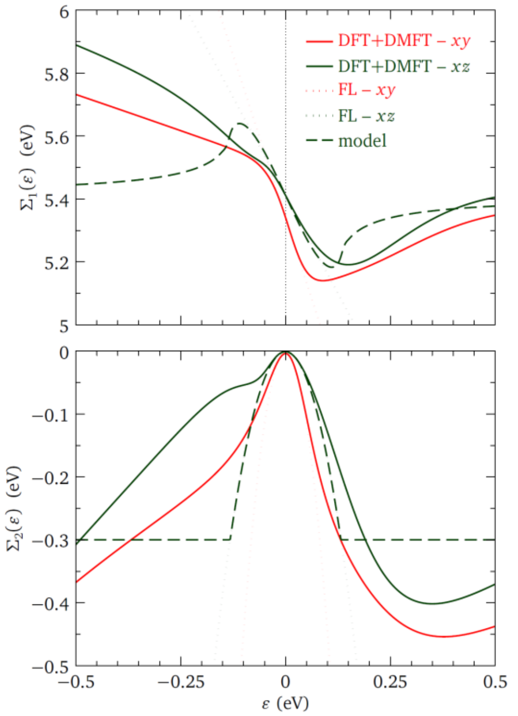
Optics on $CaRuO_3$: influence of low lying interband transitions

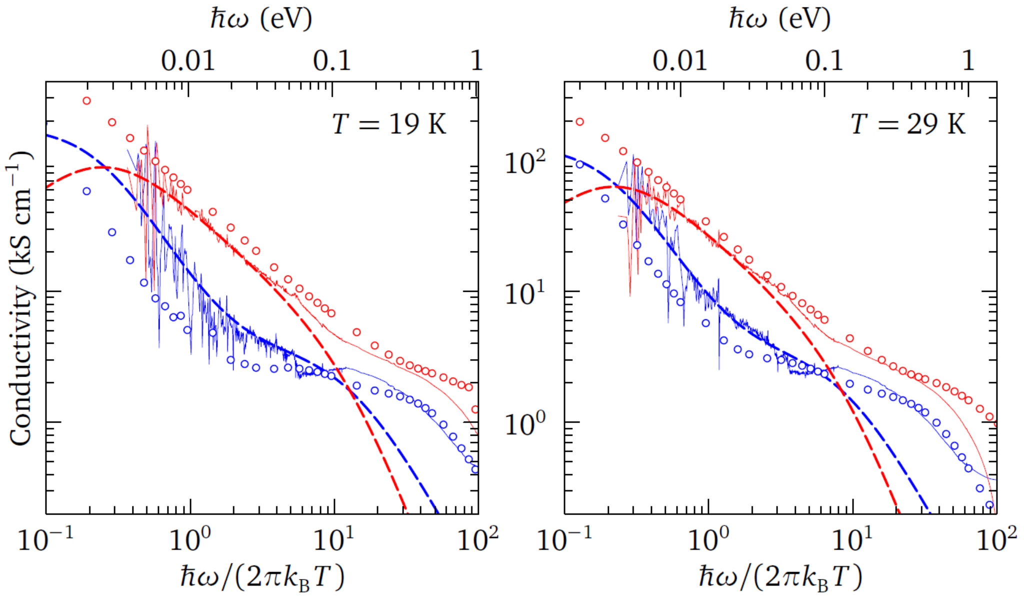


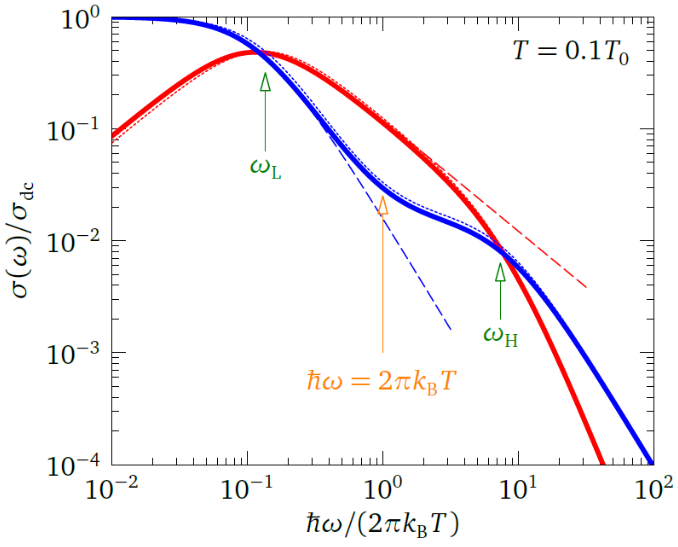


Dang, JM, Georges, Millis PRL'15









Observed in optical study
of Sr_2RuO_4
Stricker, JM,…, Georges, van der Marel, PRL’1
Berthod, JM, van der Marel, Georges PRB’14