Bad metallic behavior, entropy & thermopower: DMFT results on ruthenates

Jernej Mravlje

SPICE Workshop,
Schloss
Waldthausen,
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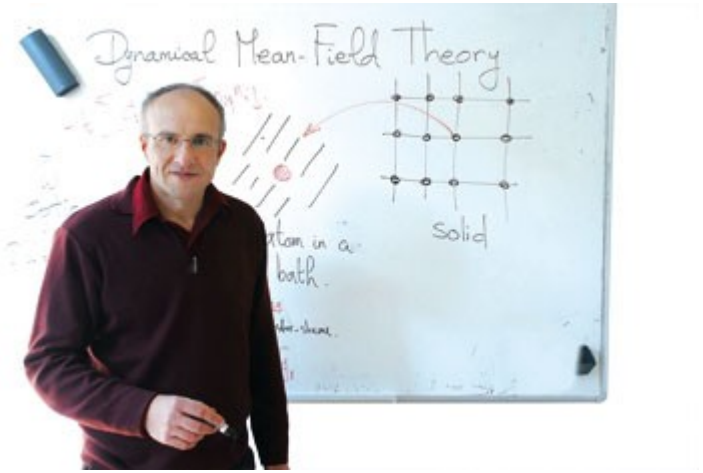
IJS

People



A. Georges, CdF,EP,UNIGE

&

X. Deng, Rutgers,NJ,
L. de'Medici, Grenoble
R. Zitko, Ljubljana
M. Aichhorn, Graz, Austria
M. Ferrero, EP, France
L. Pourovskii, EP, France
O. Parcollet, EP, France
G. Kotliar, Rutgers
K. Haule, Rutgers

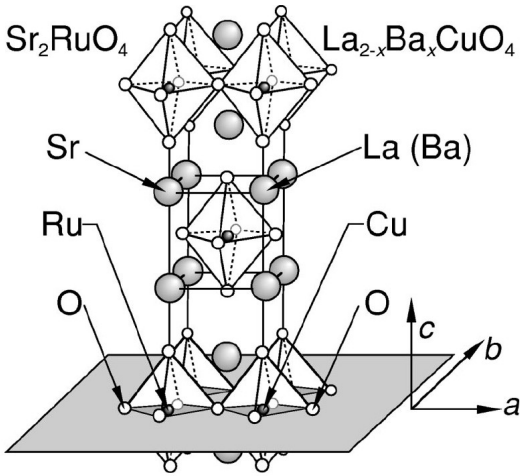
Task

- Warm correlated metals
- What precise things can be said about correlated metal at room T & above?
- What are ingredients of “incoherent soup”?

Outline

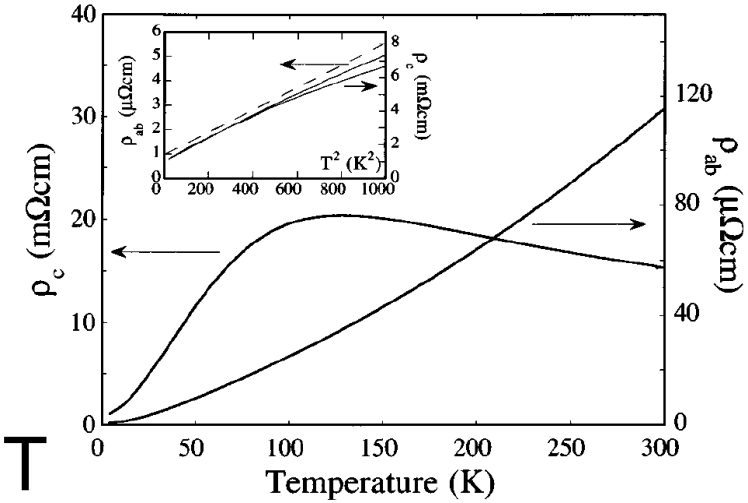
- Motivatation: bad metallic transport in ruthenates
- Results
  - Single band: Resillient qps
  - Multi band: Ruthenates as Hund's metals
  - Entropy and thermopower; revealing DOF by transport : ab-plane Seebeck in Sr_2RuO_4
- Conclusions

Cold ruthenates do interesting things...



FLs at low T

T_{FL}



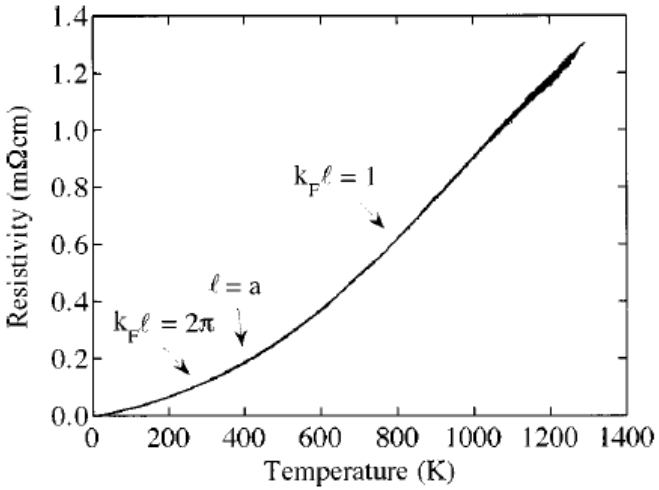
Hussey et al. PRB'98

Table 2 Ruthenates in a nutshell[a]

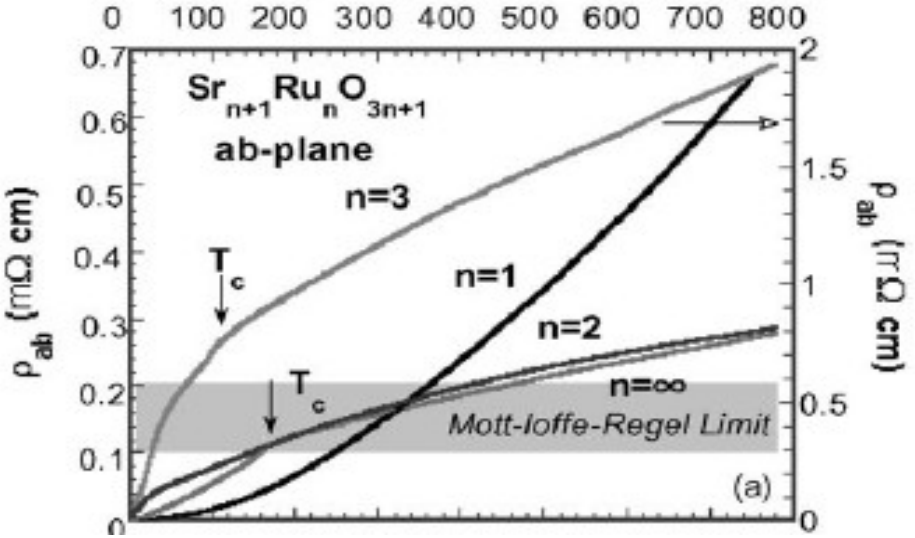
Compound	Magnetic order	γ/γ_{LDA}	$\rho \propto T^2$	Remarks
Sr_2RuO_4	PM	4	<25 K	Unconventional SC < 1.5 K
$SrRuO_3$	FM < 160 K	4	<15 K	$\sigma \propto \omega^{-0.5}$
$Sr_3Ru_2O_7$	PM	10	<10 K	Metamagnetic quantum-critical point and nematicity
$CaRuO_3$	PM	7	$T^{1.5} > 2$ K	$\sigma \propto \omega^{-0.5}$,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

...but here we prefer them warm. bad metals at high T

Sr_2RuO_4



Hussey et al. PRB'98
Tyler et al, PRB'98



Cao et al. Solid State Comm (2004)

Warm ruthenates are "bad metals" as $k_F l < 1$ at high T

WHAT IS A *BAD METAL*?

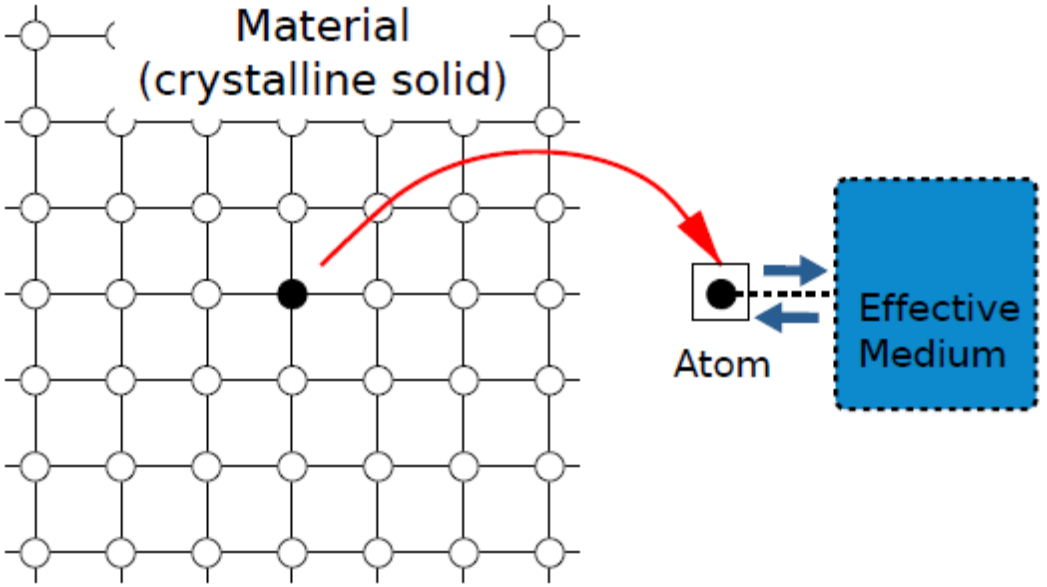
DOES MIR HAVE MEANING FOR SCES?

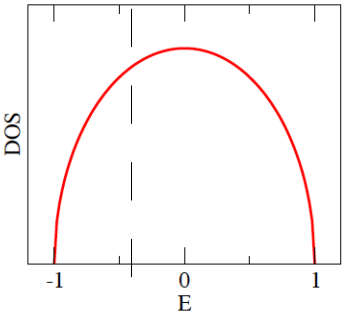
WHEN DO QP VANISH?

SHOULD ONE BE THINKING IN TERMS OF QPs AT ALL?

Gunnarsson, Calandra & Han, RMP (2003)
Hussey, Takenaka & Takagi, Phil. Mag. (2004)

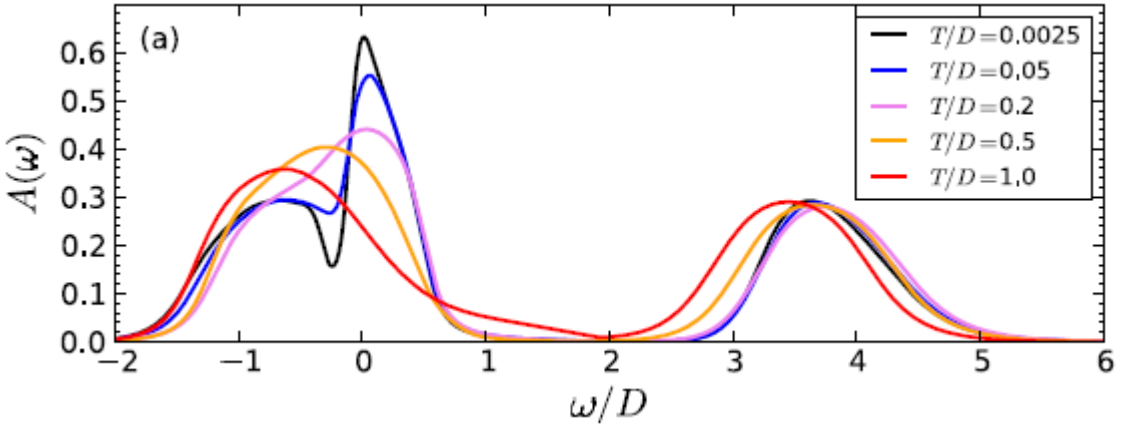
Dynamical mean-field theory



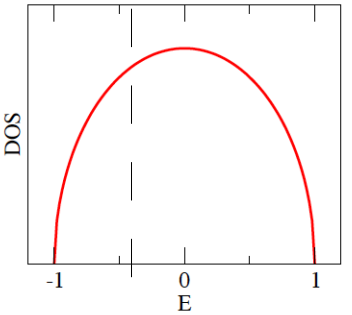


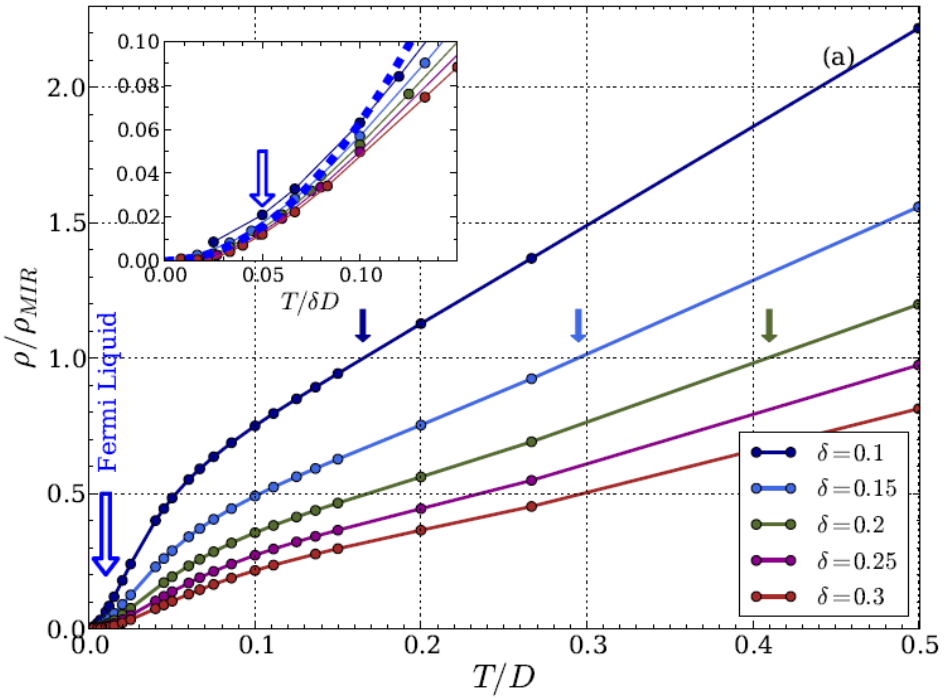
Georges et al. RMP'96
Continous time Monte Carlo solvers: Gull et al. RMP'11;
Parcollet et al.: TRIQS project
NRG solver : Zitko: NRGLJUBLJANA

DMFT model results : doped (single band) Mott insulator



U=4D, doping 0.2

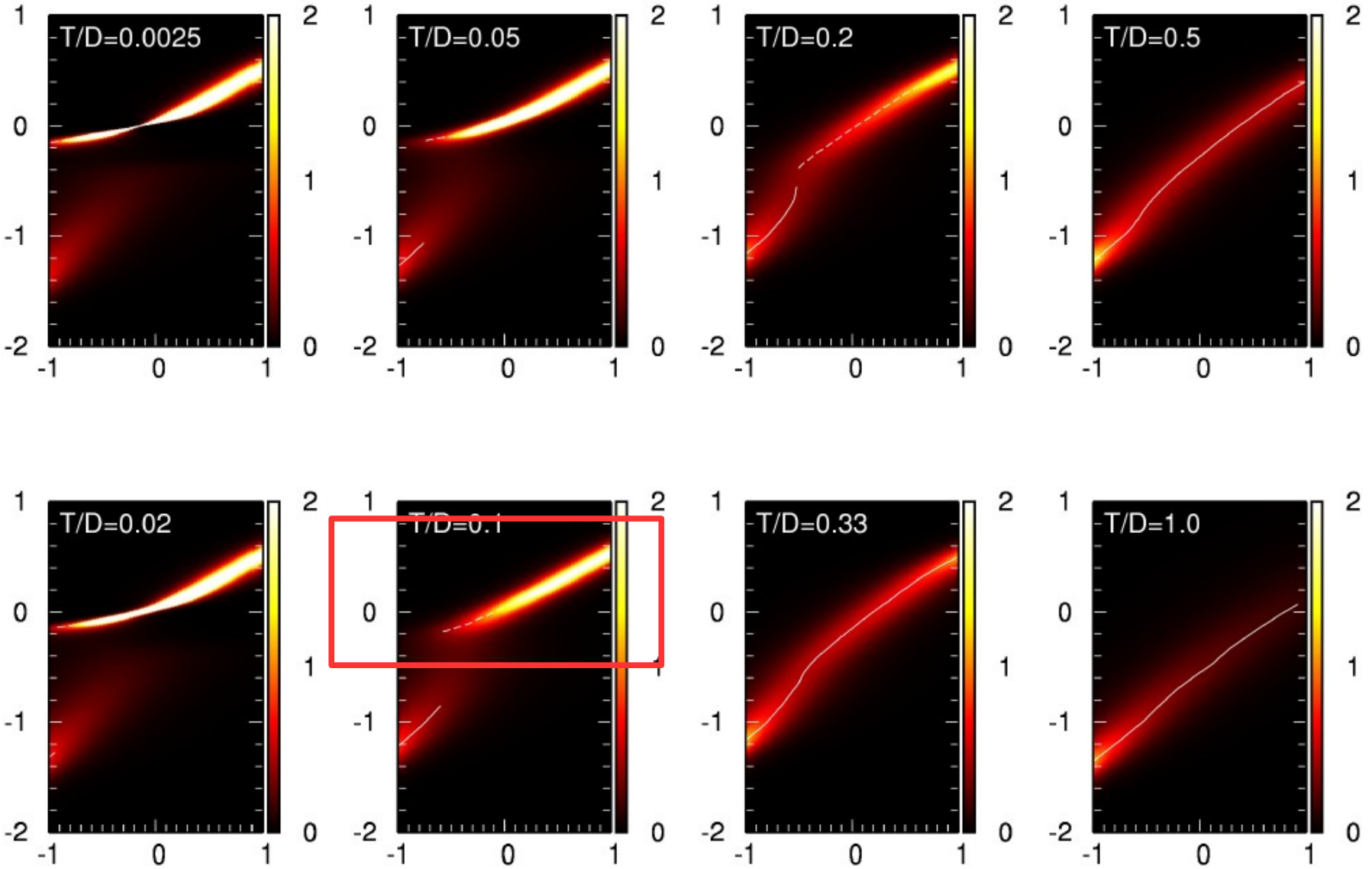




X.Deng, JM et al. PRL'13
See also J. Vucicevic et al., PRL'15, POSTER!

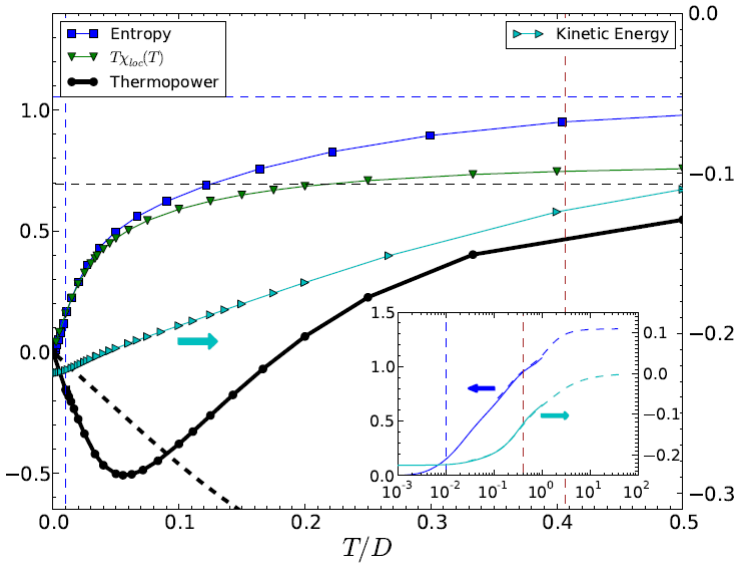
$k_F l = 1$

… and calculated A(k,ω) resillient quasiparticles



Comparison to thermodynamic qs.

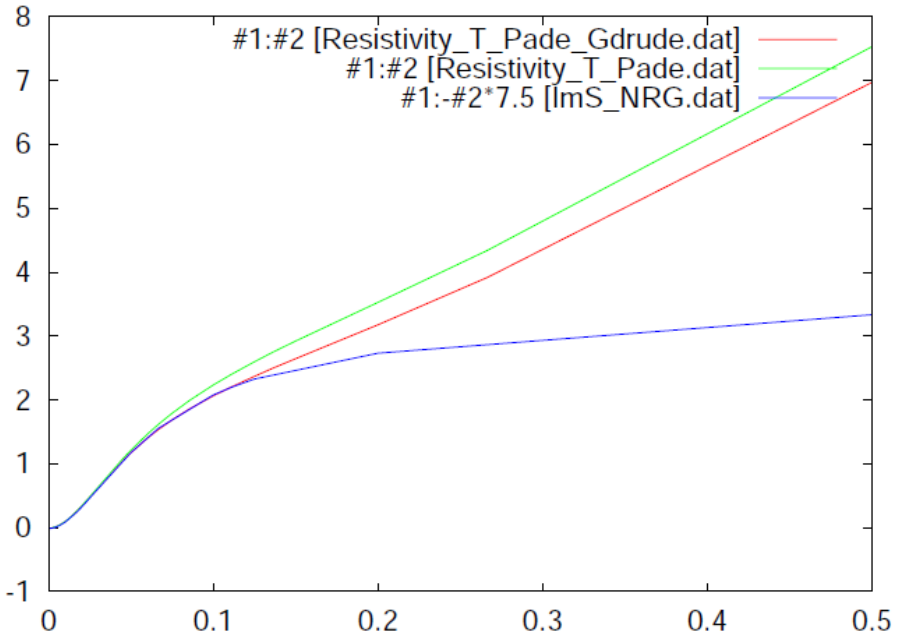
- Quasiparticles survive to regimes where entropy is large; two-fluid picture with coexisting quasiparticles & local moments

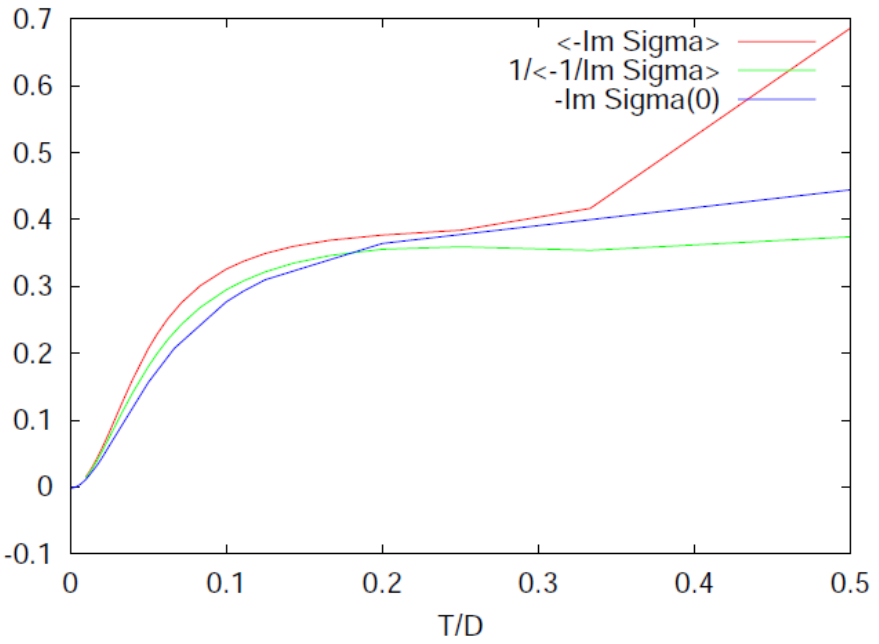


About resistivity saturation & generalized Drude

- Scattering rate saturates, resistivity not

$$\sigma = \int -\left(\frac{\partial f}{\partial \omega}\right) \tilde{\Phi}(\omega)\tau(\omega)d\omega \approx n_{\text{eff}} e^2/m \int d\omega \tau(\omega)$$



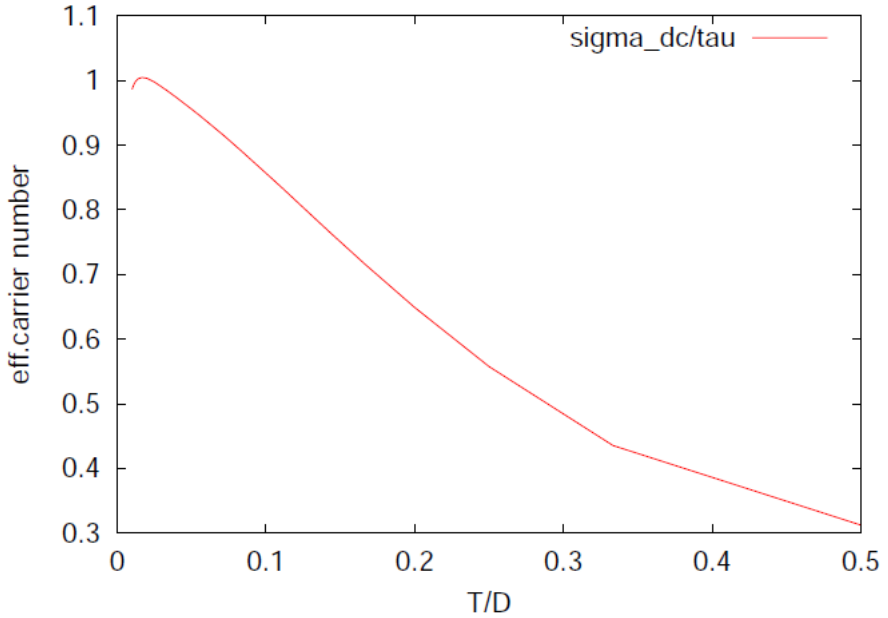


Carrier number

- Within Drude description, at high temperature τ saturates, plasma frequency (effective carrier number) keeps dropping

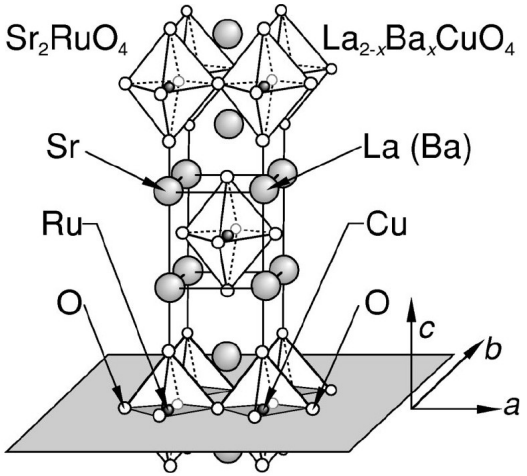
$$\sigma = \frac{ne^2}{m}\tau = \omega_p^2\tau$$

- Bad metal is like doped semiconductor, controlled by T-dependence of carrier # not their scattering

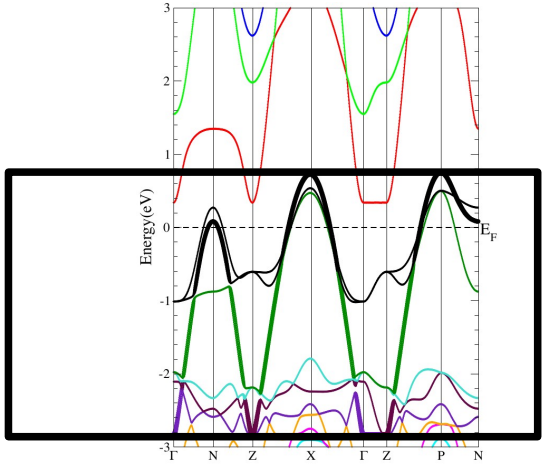


Correlations in multiorbital systems: Sr_2RuO_4

- Ruthenates are not Mott insulators
- bands broad W=3eV, U small U<W
- why correlated?
- 4 els in 3 t_{2g} orbitals

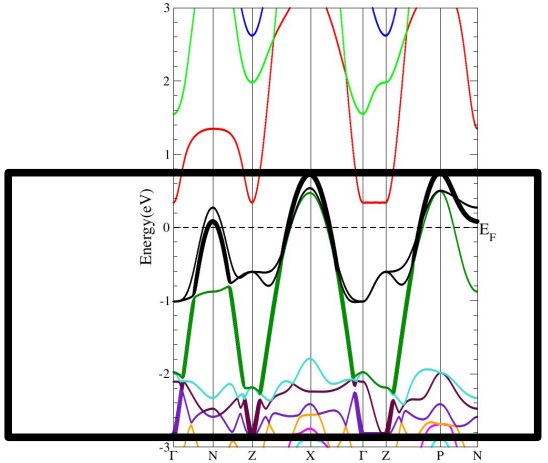


JM et al. PRL'11



Sr_2RuO_4 within LDA+DMFT

- Wannier function constructed out of t2g
- Interaction on t_{2g} atom



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

$$H = (U - 3J) n(n-1)/2 - 2JS^2 - 1/2 JL^2$$

$$\vec{S} = 1/2 \sum_{m\sigma\sigma'} c^\dagger_{m\sigma} \vec{\tau} c_{m\sigma'}$$

$$L_m = i \sum_{\sigma m' m''} \epsilon_{m m' m''} c^\dagger_{\sigma m'} c_{\sigma m''}$$

Hund's rule coupling

•LDA + DMFT reproduces exp. masses ; U=2.3 <W

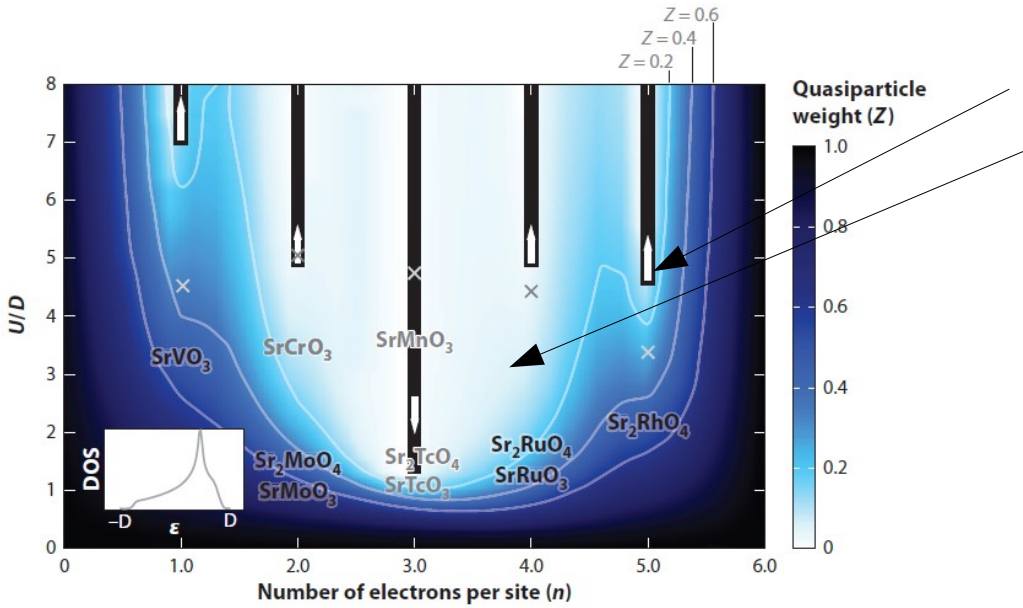
J [eV]	$m^*_{xy}/m_{\rm LDA}$	$m^*_{xz}/m_{\rm LDA}$	T^*_{xy}[K]	T^*_{xz}[K]	$T_>$[K]
0.0, 0.1	1.7	1.7	> 1000	> 1000	> 1000
0.2	2.3	2.0	300	800	> 1000
0.3	3.2	2.4	100	300	500
0.4	4.5	3.3	60	150	350

• J causes correlations, orbital differentiation and low coherence scale

JM et al. PRL'11

Hund's metals

$$H = (U - 3J)n(n-1)/2 - 2JS^2 - 1/2JL^2$$



2 effects of Hund's coupling:
Except at half filling it **increases** Uc
It suppresses Z when there is more than one el. de'Medici, JM, Georges, PRL'11.

Hund's metals: work by groups of Kotliar and Haule, and Millis and Werner,

Georges, de'Medici, JM, Annu Rev. CM'13

Effective Kondo problem has a suppressed spin-spin Kondo coupling (or even ferro) due to J. Suppressed spin-coherence scale.

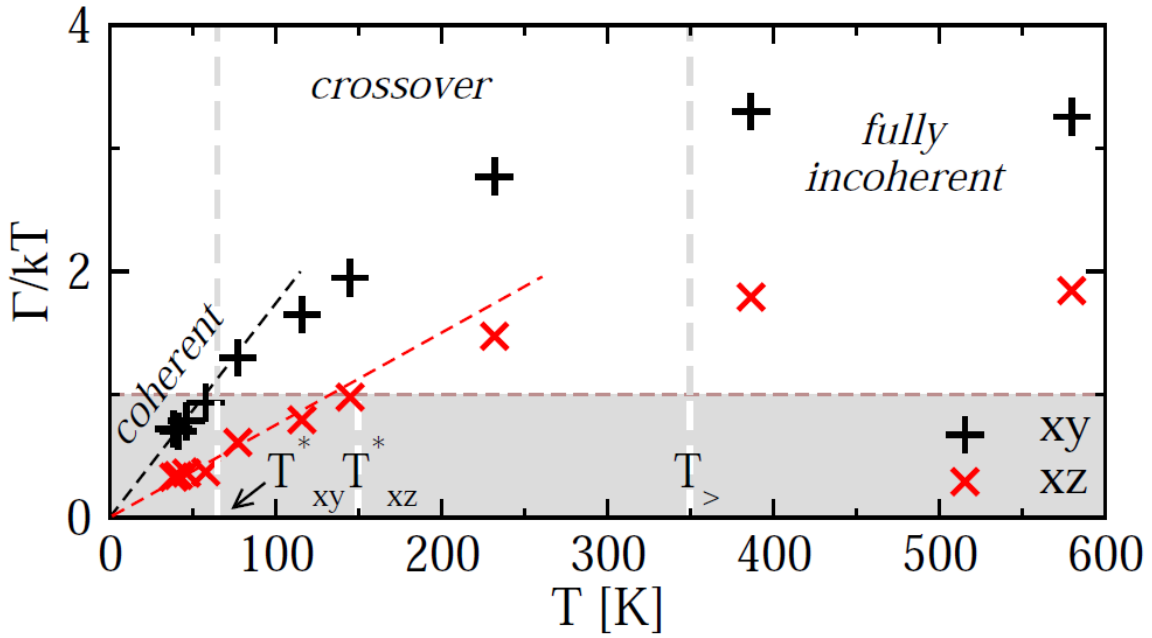
Yin, Haule, Kotliar PRB'12
Okada PTP'73; Aron Kotliar PRB'15

$$H_{\rm int} = J_{\rm p}\,\psi^\dagger_{a\sigma}\psi_{a\sigma} + J_0\,S^\alpha\left(\psi^\dagger_{m\sigma}\frac{\sigma^\alpha_{\sigma\sigma'}}{2}\psi_{m\sigma'}\right) + K_0 T^a\left(\psi^\dagger_{m\sigma}\frac{\tau^a_{mm'}}{2}\psi_{m'\sigma}\right) + I_0 S^\alpha T^a\left(\psi^\dagger_{m\sigma}\frac{\sigma^\alpha_{\sigma\sigma'}}{2}\frac{\tau^a_{mm'}}{2}\psi_{m'\sigma'}\right),$$

Sr_2RuO_4: Crossover to incoherent regime

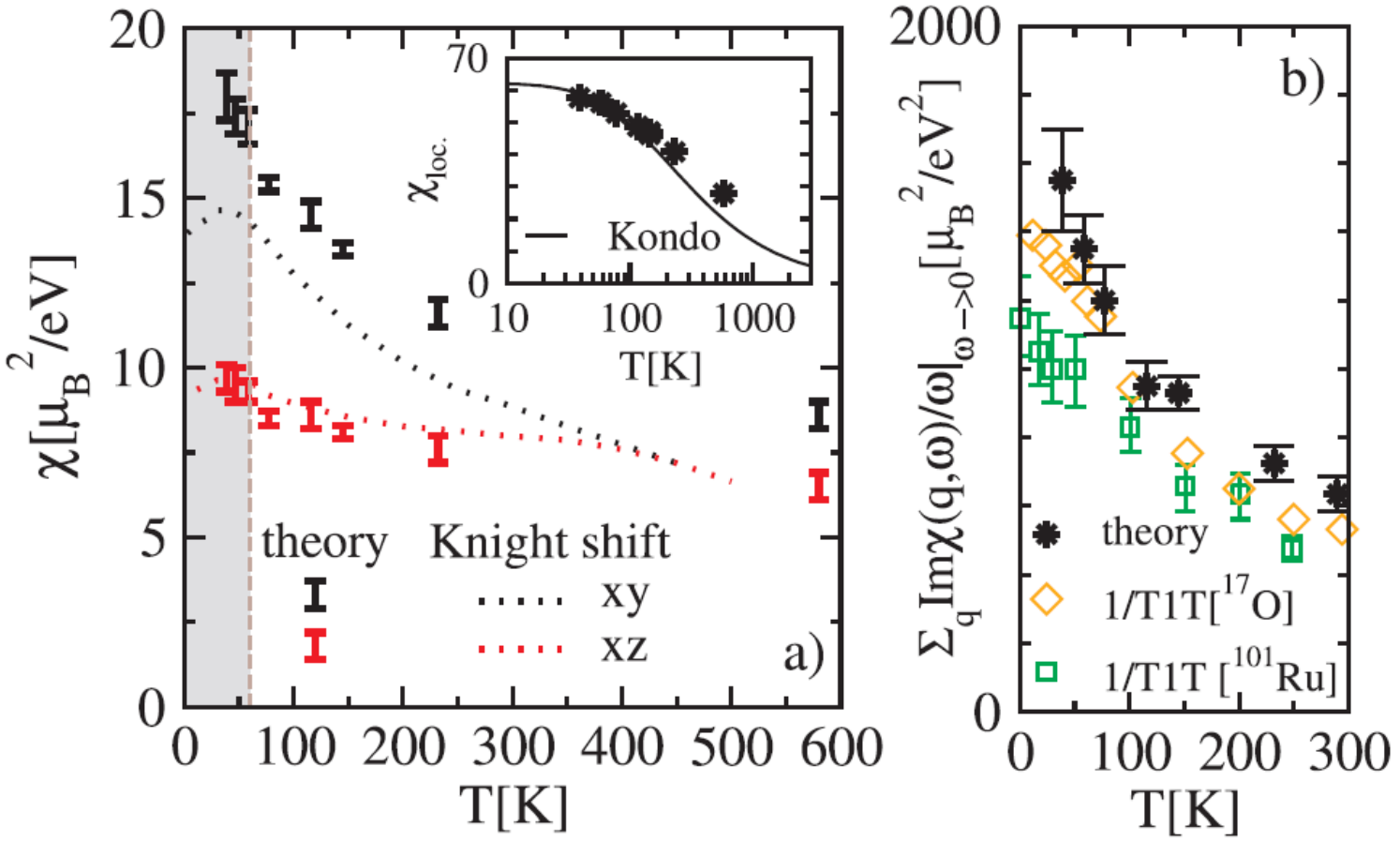
- Scattering rate:
  Fermi liquid $\Gamma = -Z\mathrm{Im}\Sigma(\mathrm{i}0^+)$ < kT

crosses over above T^* from ~T^2 to ~ T

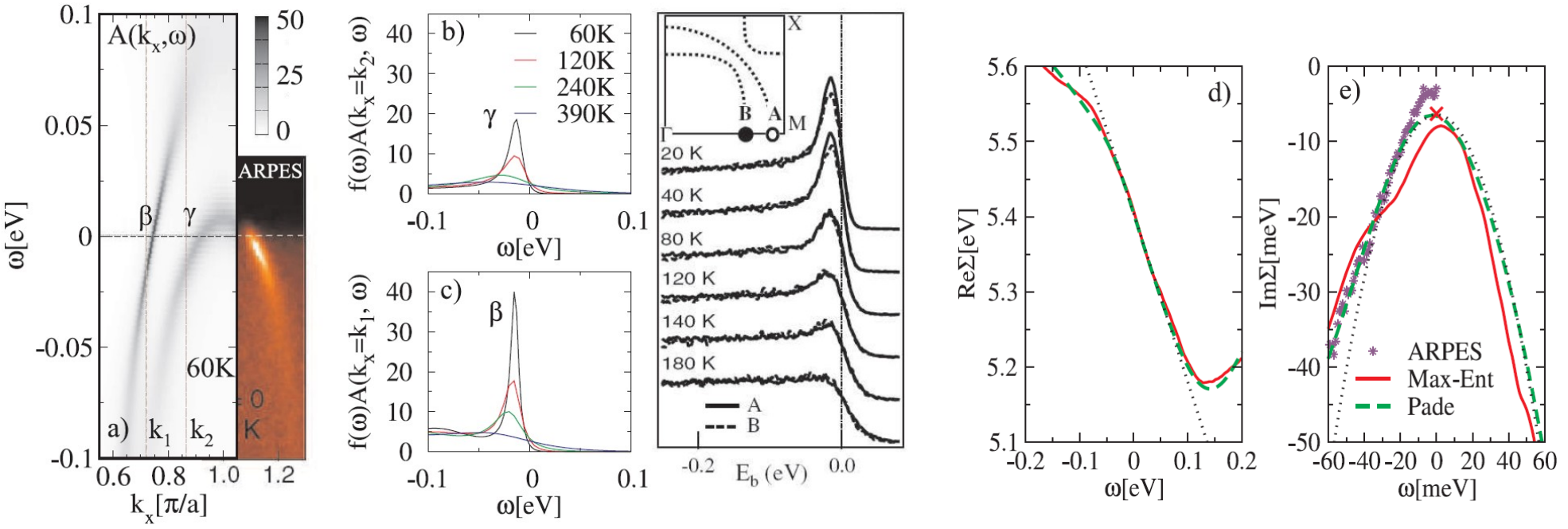


At T*: Gamma/kT =1

NMR


20
15
10
5
0
0
100
200
300
400
500
600
T[K]
$\chi[\mu_B{}^2/eV]$
70
0
$\chi_{loc.}$
10
100
1000
T[K]
Kondo
theory
Knight shift
xy
xz
a)
2000
0
0
100
200
300
T[K]
$\Sigma_q Im\chi(q,\omega)/\omega|_{\omega->0}[\mu_B{}^2/eV^2]$
b)
theory
1/T1T[^{17}O]
1/T1T [^{101}Ru]

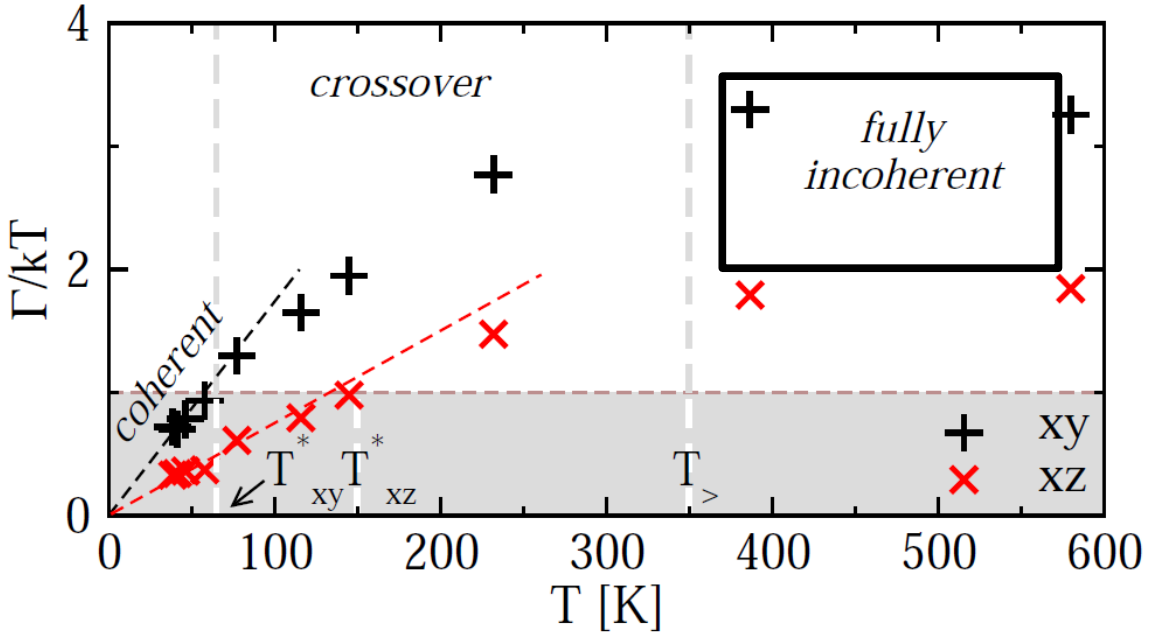
ARPES



Shen et al.,PRL'07 Wang et al.,PRL'04 Ingle et al.,PRB'05

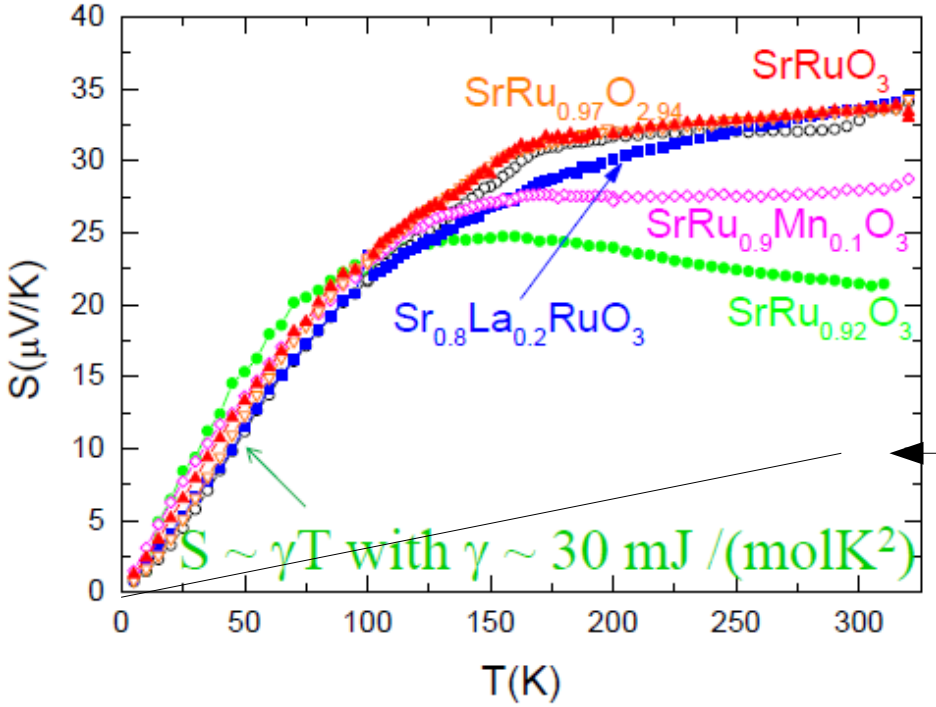
Disappearence of (resillient!) q.ps. at high T

Characterizing the incoherent regime better: thermopower



Seebeck in ruthenates

- Low T: linear in T (Mott)
- Higher T: saturation
- Why ~universal behavior ? Entropic origin?



VALUE ?

LDA slope

(plot from S. Hebert)
CRISMAT
Klein,Hebert et al PRB'06

Entropic considerations? Heikes' formula

- As chem. pot ~ T at large T

$$\alpha = \Delta V/\Delta T = -\frac{1}{eT}\frac{L^{j,j_Q}}{L^{j,j}} = -\frac{1}{eT}\frac{L^{j,j_E}}{L^{j,j}} + \frac{\mu}{eT}$$

$$j_Q = j_E - \mu j$$

$$\alpha_{\text{Heikes1}} = \mu/eT = (\partial S/\partial_n)_E$$

Region of applicability	Thermopower
$kT \gg U_0,\ U_j, t$	$-\frac{k}{\lvert e\rvert}\ln\frac{2-\rho}{\rho}$
$U_0 \gg kT \gg U_j, t$	$-\frac{k}{\lvert e\rvert}\ln\frac{2(1-\rho)}{\rho}$

Chaikin, Beni, PRB'76

- Two possibilities:
  - 1. “Heikes” :evaluate μ/T in atomic limit
  - 2. Take μ/T from calculation and evaluate Seebeck
- Shastry: Kelvin formula; (wrong) static limit of Kubo

$$\alpha_K = \partial_\mu/\partial_T = \partial S/\partial n_T$$

Silk, Terasaki, Schofield, PRB'09
Peterson, Shastry, PRB'10

- Heikes formula succesful in localized systems, e.g. $La_{1-x}Sr_xVO_3$ next to the MIT Uchida et al. PRB'11
- Here we have a metal (admitedly a bad one). Are we expecting entropic/atomic estimates to work here?

Seebeck in LDA+DMFT

$$S = -\frac{k_B}{e_0}\frac{\mathcal{A}_1}{\mathcal{A}_0}$$

$$\mathcal{A}_n = \frac{2\pi}{\beta\hbar}\int_{-\infty}^{\infty} d\omega \Phi(\omega)(-\partial f/\partial\omega)(\beta\omega)^n$$

$$\Phi(\omega) = \frac{1}{V}\sum_k \mathrm{Tr}\left[A_k v_k A_k v_k\right]$$

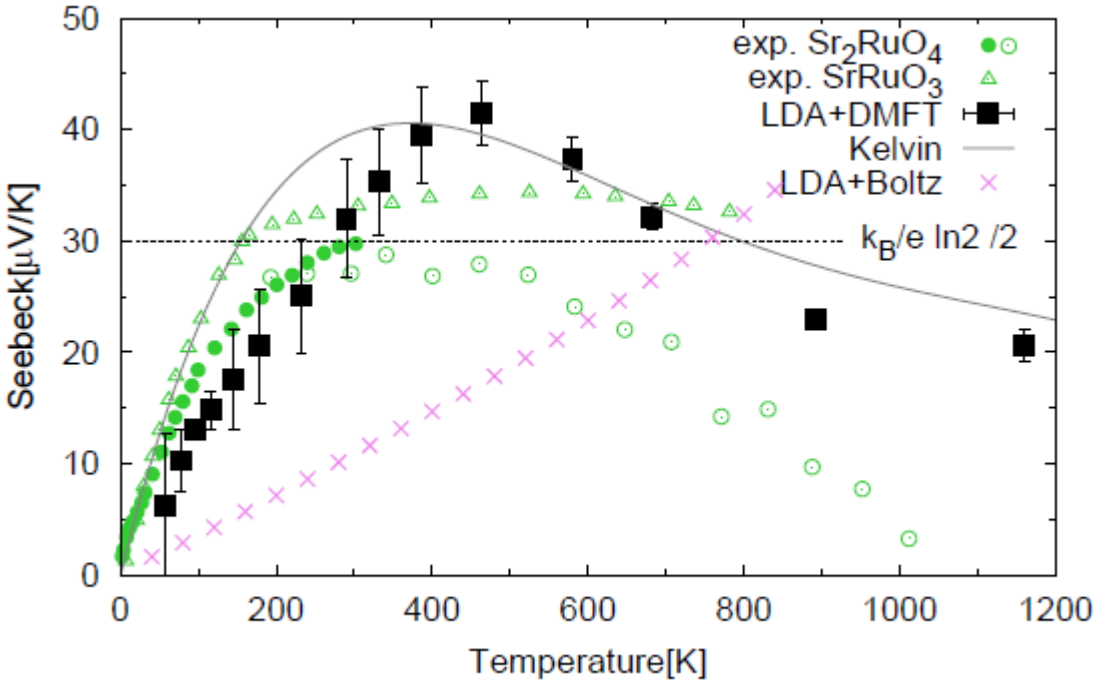
$$A(k)_{\nu\nu'} = -\frac{1}{\pi}\mathrm{Im}\left[\omega - \epsilon_k\delta_{\nu\nu'} - \Sigma(w,k)_{\nu\nu'}\right]^{-1}$$

Inplane thermopower

- Kelvin formula describes data well

$$\alpha_{\mathrm{Kelvin}} = (1/e_0)\partial\mu/\partial T.$$

$$(\partial\mu/\partial T)_n = -(\partial S/\partial n)_T$$

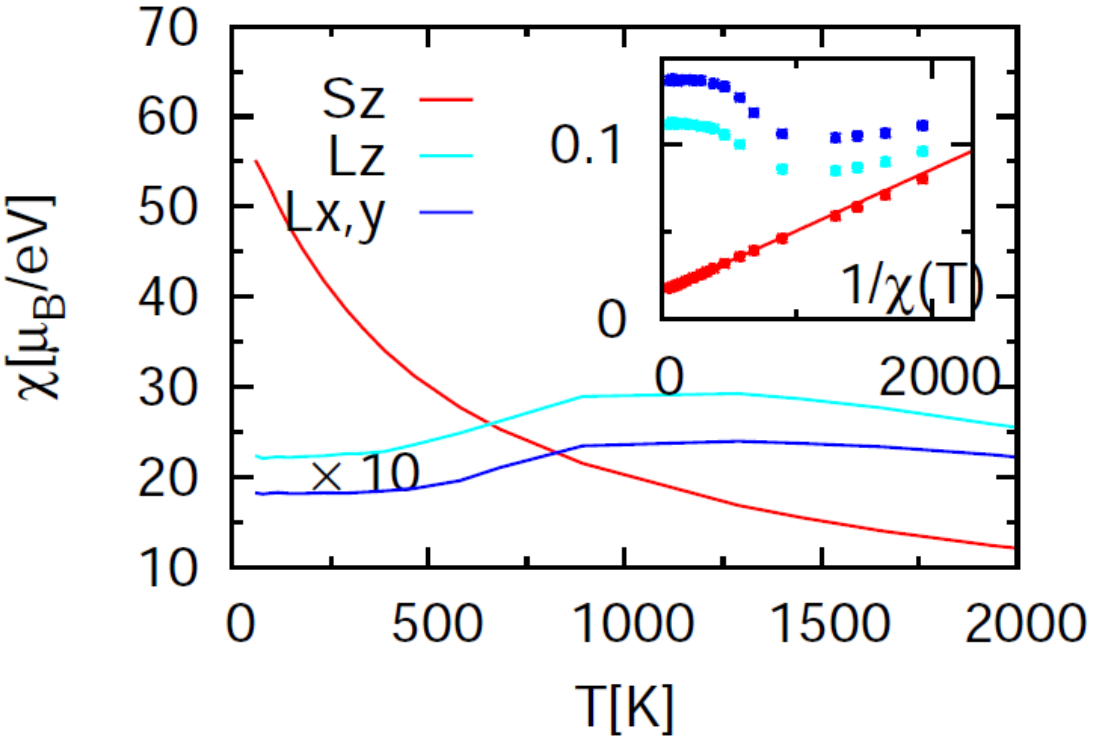


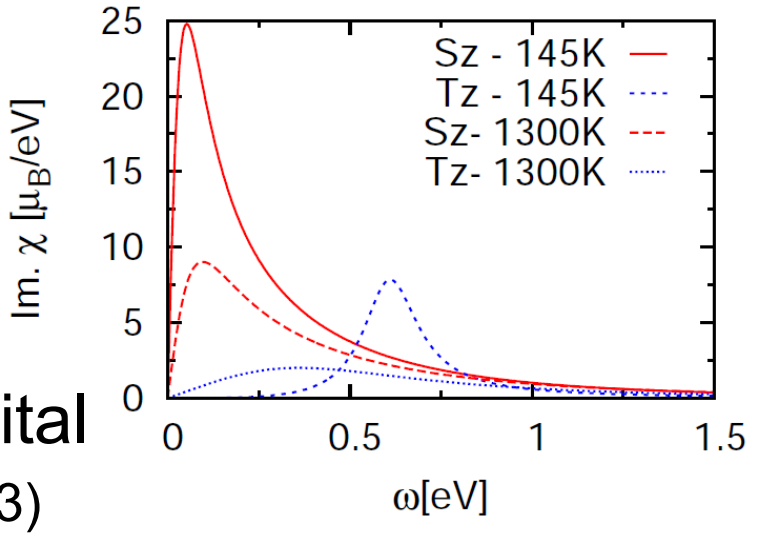
Xu, Xu et al PRL'08
Keawprak et al. Mat. Trans'08

- Seebeck coefficient in 214 follows ent. consid.
- What does this teach us?

DOF

- Spins fluctuate whereas orbitals are quenched!



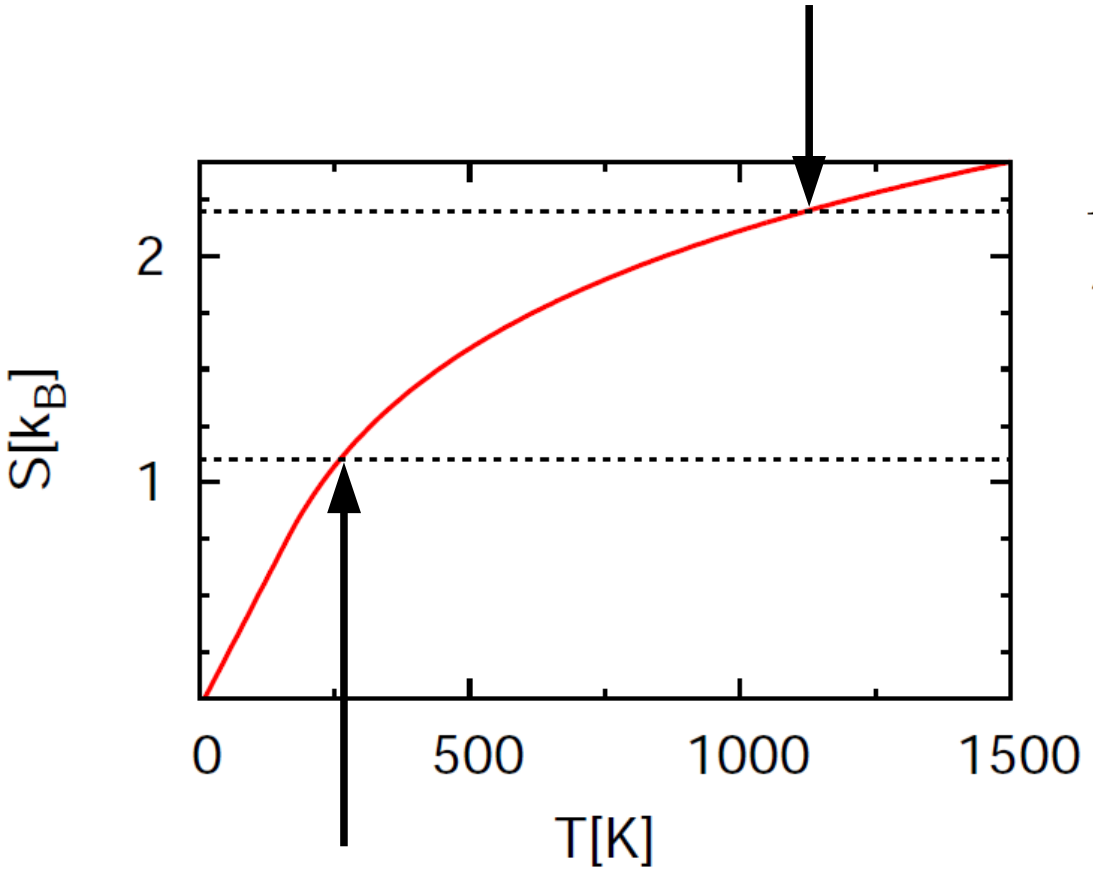


Impurity problem of Hund's metals has ferro spin and antiferromagn. orbital Kondo coupling. (Yin, Haule, Kotliar, PRB'13)

2 stage decoherence

- Entropy

(ii) Liberated orbital moments



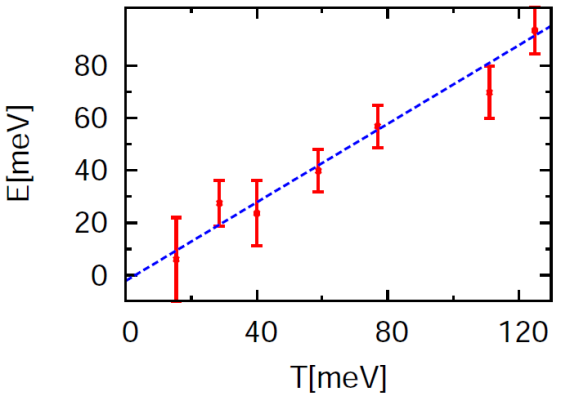
(i) Liberated spins

$$E = \gamma T^2/2$$

$$\gamma = 38\mathrm{mJ/molK}^2$$

$$S = \gamma T; T < T_0$$

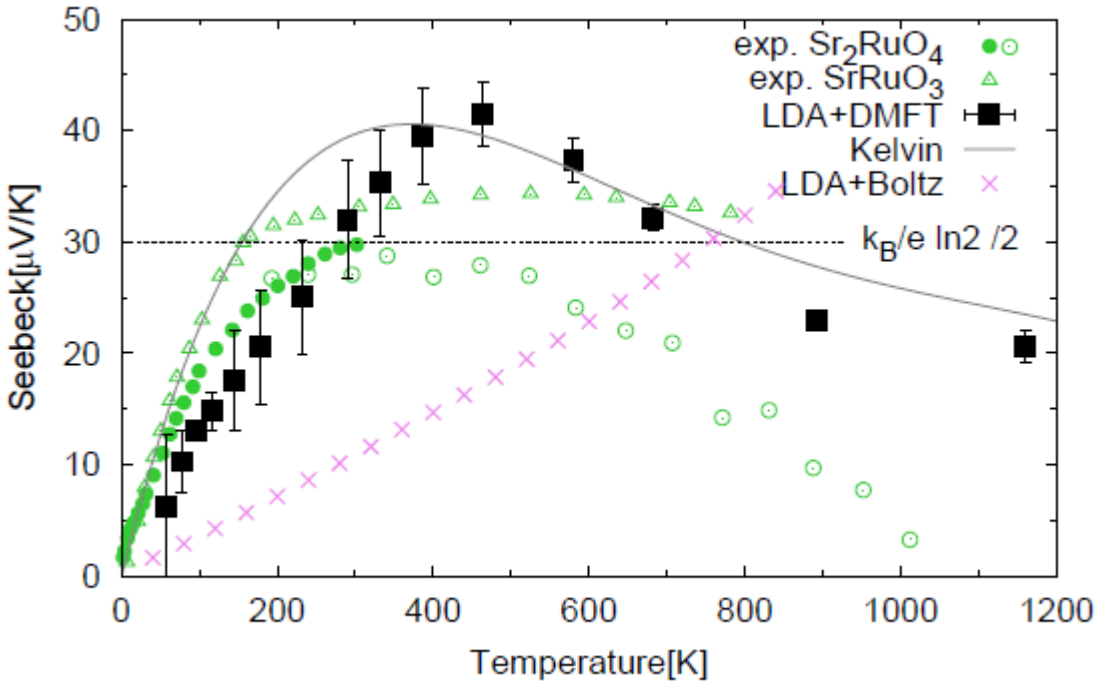
$$S = \gamma T_0 + c\log(T/T_0); T > T_0$$



$$T_0 = c/\gamma \qquad c = 0.75k_B$$

Consequences of this for Seebeck

- Knowing DOF one can attempt Heikes analysis



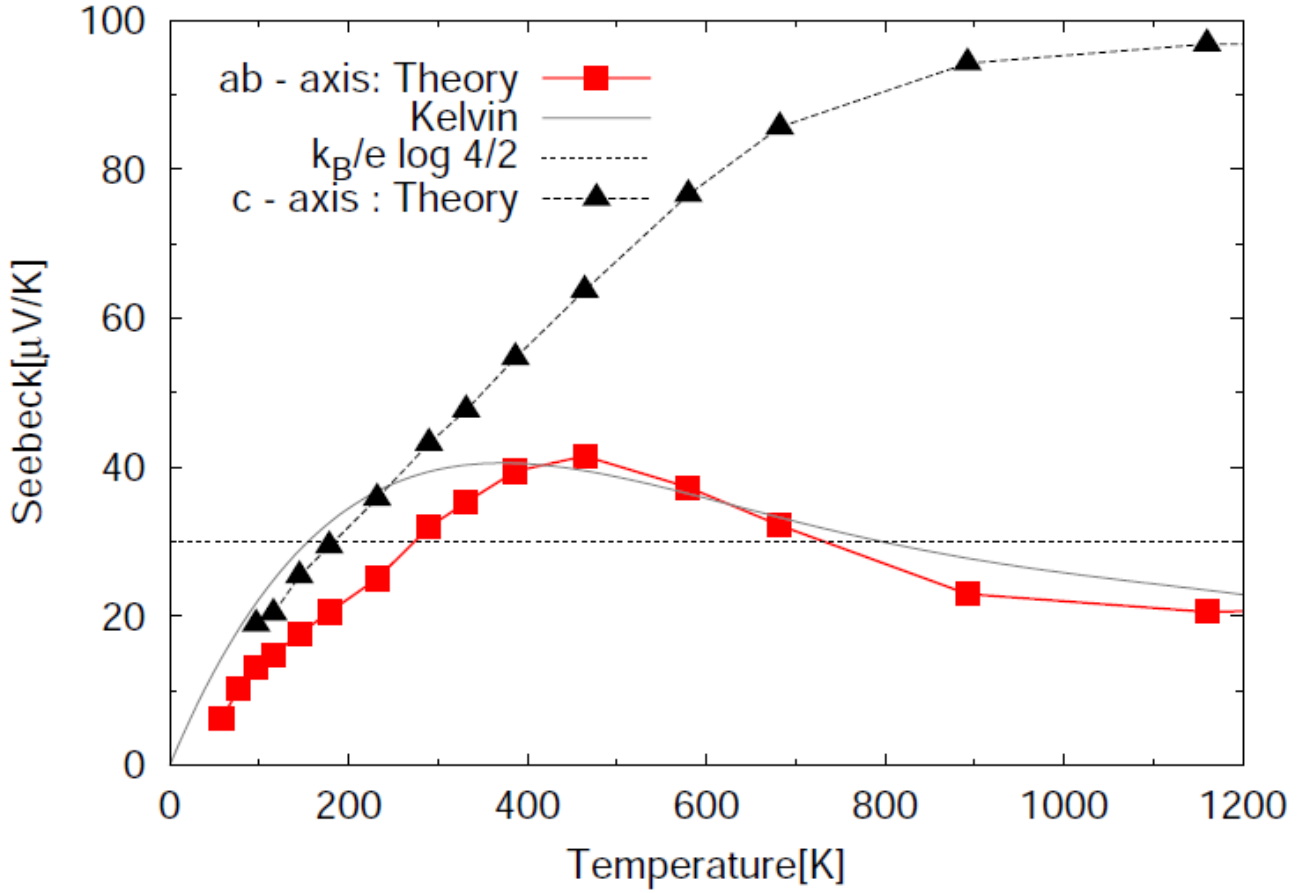
$$S_H = k_B/2e \log(d_{N-1}/d_{N+1})$$
$$= k_B/2e \log(4/2) \approx 30\mu\mathrm{V/K}$$

JM and A. Georges, arXiv'15
See also Klein et al. PRB'06

d_{N-1}=(2S+1)=4 ; N-1=3, S=3/2
d_{N+1}=(2S+1)=2 ; N+1=5, S=1/2

Can Seebeck be universally associated to entropy and hence be used as a probe to characterize degrees of freedom?

No! C-axis Seebeck (prediction)



Limitations of entropic interpretations ; noninteracting example

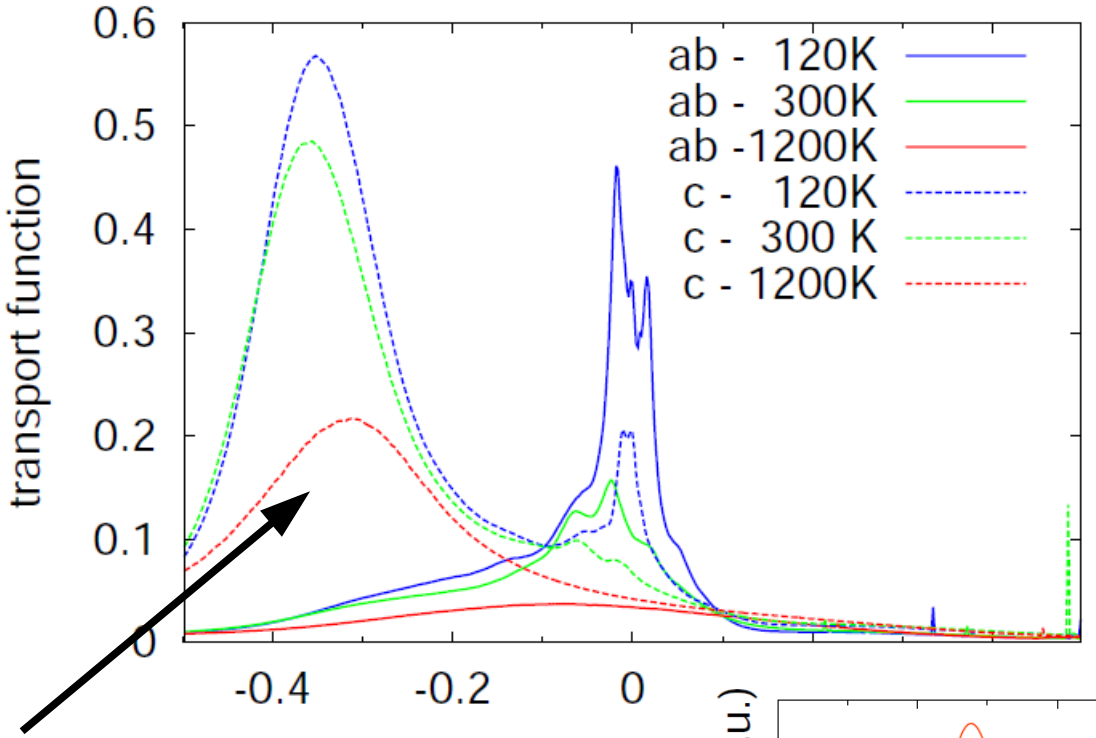
$$d\mu/dT = \int \rho(\epsilon)\epsilon(-\partial f/\partial\epsilon)d\epsilon / \int \rho(\epsilon)(-\partial f/\partial\epsilon)d\epsilon$$

$$\alpha = k_B/e \int T(\omega)\beta\omega(-\partial f/\partial\omega) / \int T(\omega)(-\partial f/\partial\omega)$$

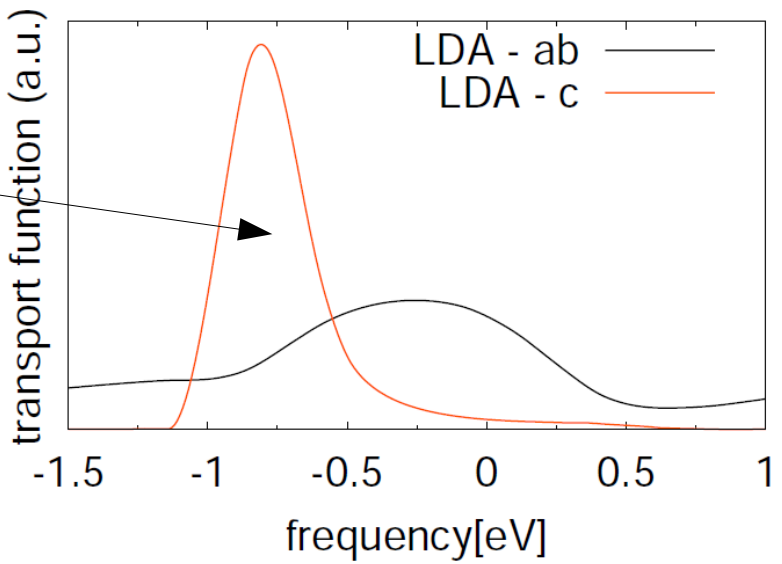
$$T(\omega) = \frac{2\pi e^2}{V} \sum_{\vec{k}} \mathrm{Tr}\, v_{\vec{k}} A_{\vec{k}}(\omega) v_{\vec{k}} A_{\vec{k}}(\omega).$$

Silk, Terasaki, Schofield, PRB'09

Transport functions

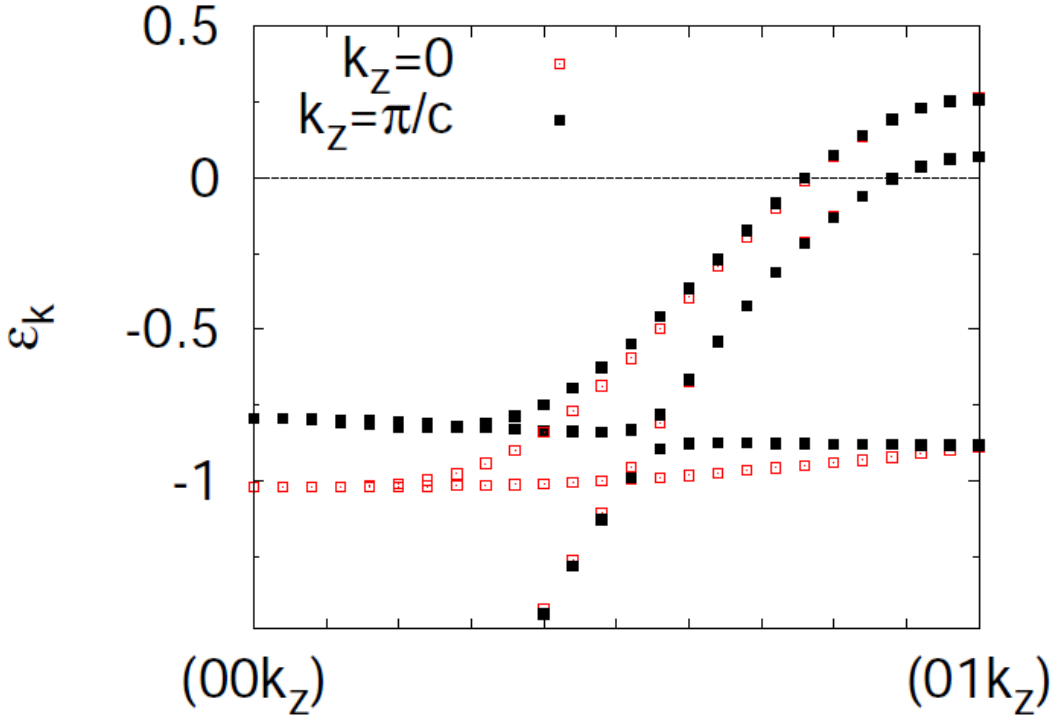


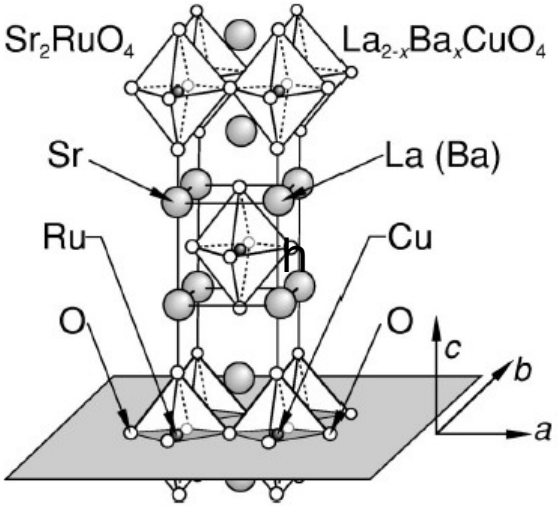
Strong peak at w<0
Present already in LDA

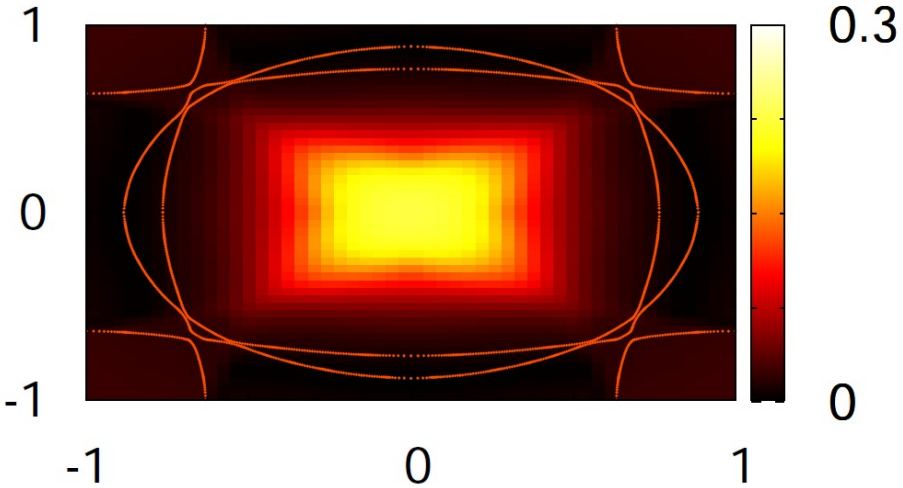


“Hole filtering mechanism”

$$\epsilon \sim \epsilon(k_x, k_y) - 2t_z \cos(k_k/2)\cos(k_y/2)\cos(k_z/2)$$

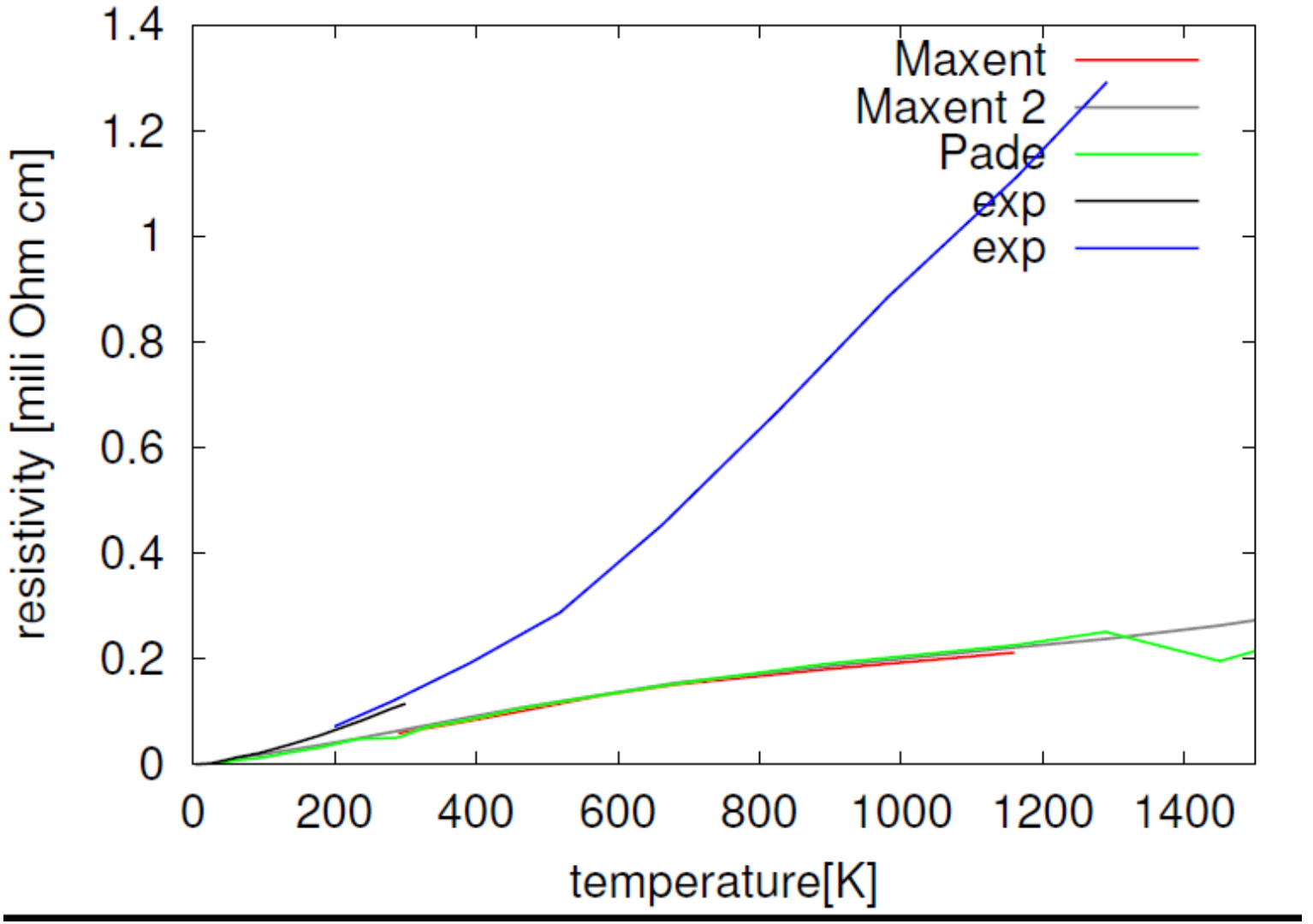




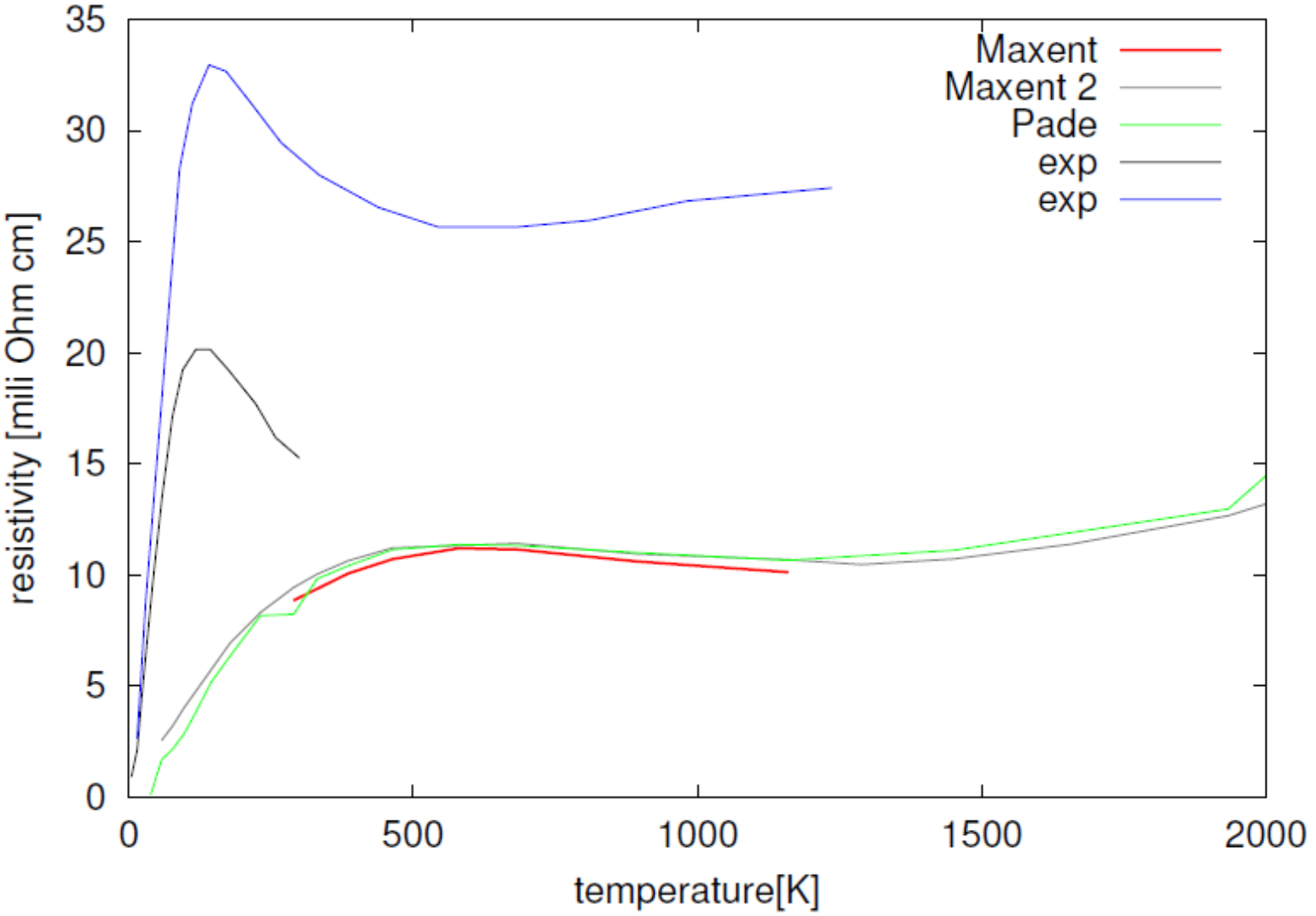


Conclusions

- Resillient quasiparticles vanishing at MIR
- Hund's coupling causes correlations
- Two stage crossover to incoherent state
- Seebeck at room T points to quenched orbitals and free spins

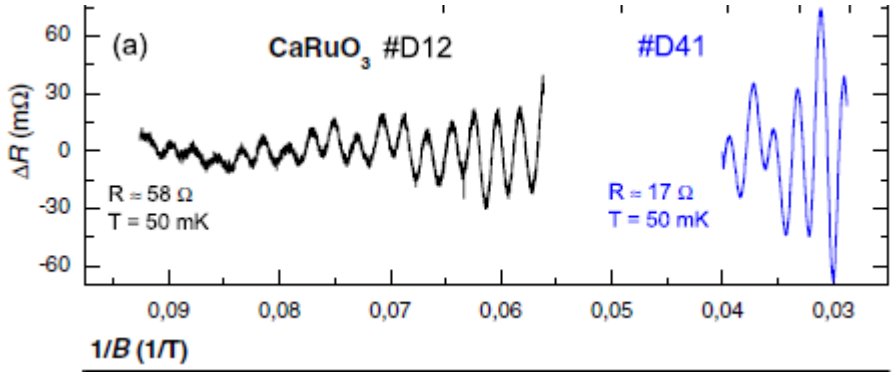

resistivity [mili Ohm cm]
temperature[K]
Maxent
Maxent 2
Pade
exp
exp
0
0.2
0.4
0.6
0.8
1
1.2
1.4
0
200
400
600
800
1000
1200
1400

Resistivity

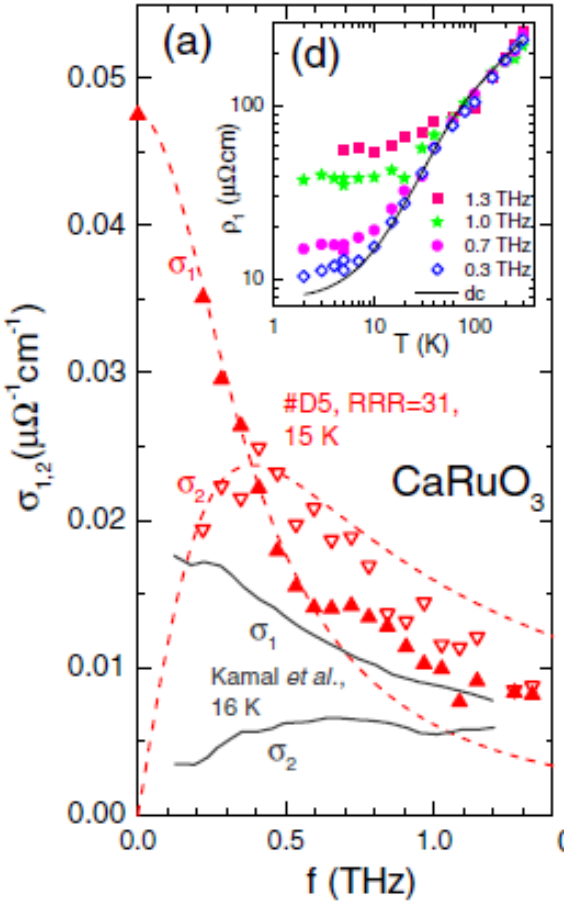


THANK YOU!

Quantum oscillations and non-Drude behavior in $CaRuO_3$

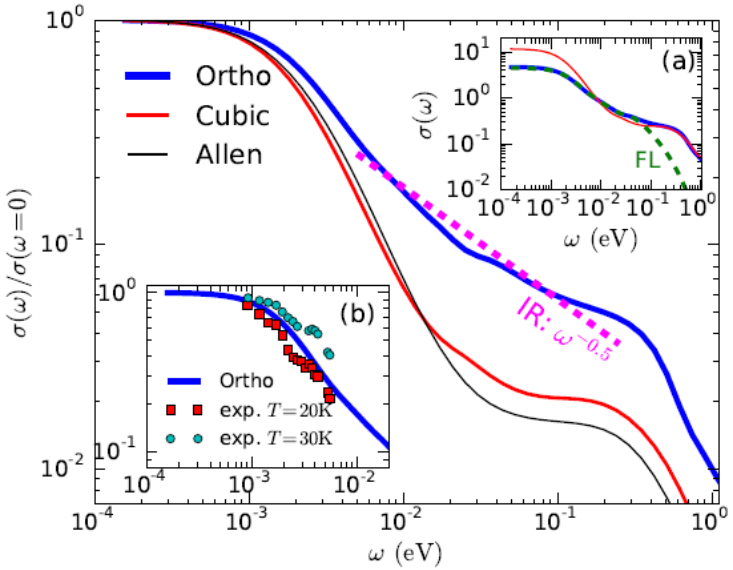


Schneider et al., PRL'14

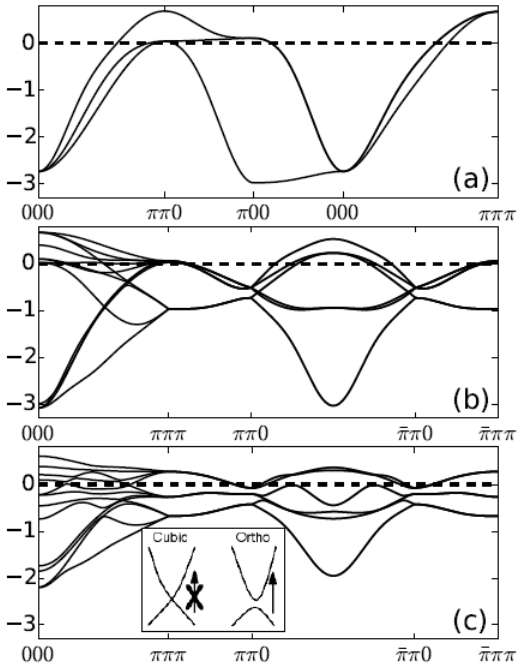


Non-Drude/power law optics in $CaRuO_3$ is a band structure effect

- Series of minigaps opened by orthorombic distortions in the meV range!



Dang, JM, Georges, Millis, arXiv'15

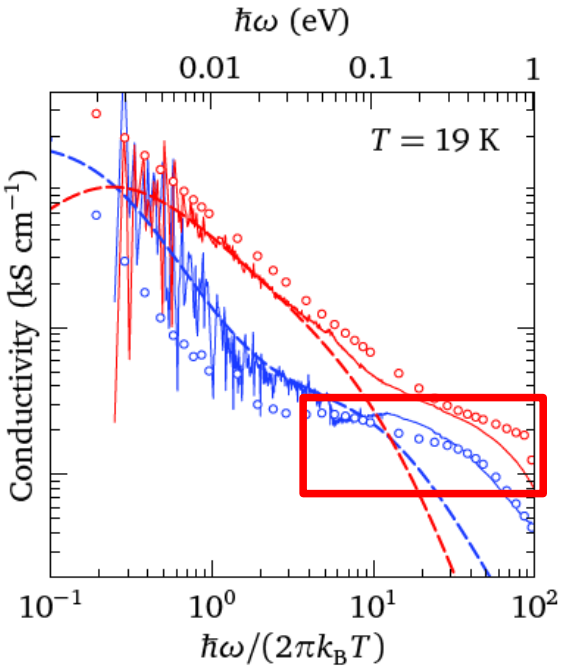


Consequences

- Existence of RQPs enables rewriting transport a la Boltzmann
- Shifts thinking from “what is going on with scattering” to “what is going on with dispersions”

Deng, JM et al, PRL'13
Wu, Kotliar, Haule,”Hidden FI PRL'13

- Success of such thinking: explaining fine structure of Drude peak in optical spectroscopy of Sr_2RuO_4



Stricker, JM et al. PRL'14

Transport in DMFT (i)

- Vertex correction vanish, from Kubo formula one has

$$\sigma = \frac{2\pi e^2}{\hbar} \int d\omega \left(-\frac{\partial f}{\partial \omega} \right) \frac{1}{V} \sum_k v_k A_{\mathbf{k}}(\omega) v_k A_{\mathbf{k}}(\omega)$$

$$v_k = \partial \varepsilon_{\mathbf{k}} / \partial \mathbf{k}_x \;\; ; \;\; A_{\mathbf{k}}(\omega) = -(1/\pi) \mathrm{Im}[\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma(\omega)]^{-1}$$

- Velocity distribution function (units $[x]^{2\text{-}d}$ $[\omega]$)

$$\Phi(\varepsilon) = (1/V) \sum_{\mathbf{k}} (\partial \varepsilon_{\mathbf{k}} / \partial \mathbf{k}_x)^2 \delta(\varepsilon - \varepsilon_{\mathbf{k}})$$

$$\sigma = \frac{2\pi e^2}{\hbar} \int d\omega \left(-\frac{\partial f}{\partial \omega} \right) \int d\epsilon \Phi(\epsilon) A_\epsilon(\omega) A_\epsilon(\omega)$$

- For semicircular DOS:

$$\Phi(\epsilon) \;=\; \Phi(0) \left[1 - (\epsilon/D)^2 \right]^{3/2}$$

Transport in DMFT (ii)

- Velocity distribution function (units $[x]^{2-d}$ $[\omega]$)

$$\Phi(\varepsilon) = (1/V)\sum_{\mathbf{k}} (\partial\varepsilon_{\mathbf{k}}/\partial\mathbf{k}_x)^2 \delta(\varepsilon - \varepsilon_{\mathbf{k}})$$

$$\sigma = \frac{2\pi e^2}{\hbar}\int d\omega \left(-\frac{\partial f}{\partial\omega}\right)\int d\epsilon \Phi(\epsilon) A_\epsilon(\omega) A_\epsilon(\omega)$$

- At low T, only $\omega \sim 0$, $k \sim k_F$ states contribute.

$1/\rho_{\mathrm{MIR}} \equiv e^2\Phi(0)/\hbar D$ a natural unit for conductivity, used henceforth

- If evaluated for a 2d electron gas, one gets

$$\sigma = (k_F l)\, e^2\Phi(\varepsilon_F)/\hbar\varepsilon_F = (k_F l)e^2/h$$

$k_F l$ times the conductivity quantum, thus this choice corresponds to ρ_{MIR} for criterion $k_F l = 1$

Real world (i.e.bandstructure) effects

- c-axis Seebeck

Ruthenates via Heikes (slide from S. Hebert)

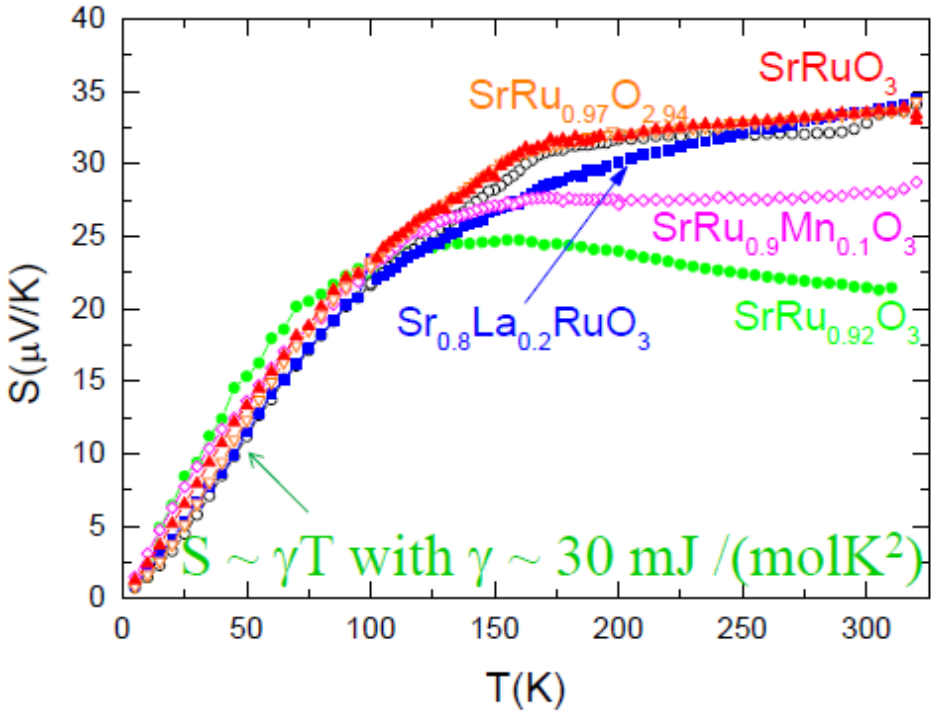
2 remarks
What happens to x?

Where are orbital degrees of freedom?

$SrRuO_3$

Spin entropy

$$S = -\frac{k_B}{|e|}\ln\left(\beta\frac{1-x}{x}\right) \quad \text{with} \quad \beta = \frac{2S_n+1}{2S_{n+1}+1}$$



Y. Klein et al., PRB73, 052412 (2006)

Ru^{3+}/Ru^{4+}

$$\beta = \frac{2\times(1/2)+1}{2\times 1+1}$$

$S_{spin} = 35\mu V/K$

$Sr_{0.8}La_{0.2}RuO_3 : v_{Ru} = 3.80$

Ru^{4+}/Ru^{5+}

$$\beta = \frac{2\times 1+1}{2\times(3/2)+1}$$

$S_{spin} = 25\mu V/K$

$SrRu_{0.9}Mn_{0.1}O_3 : v_{Ru} = 4.11$

(slide from S. Hebert)

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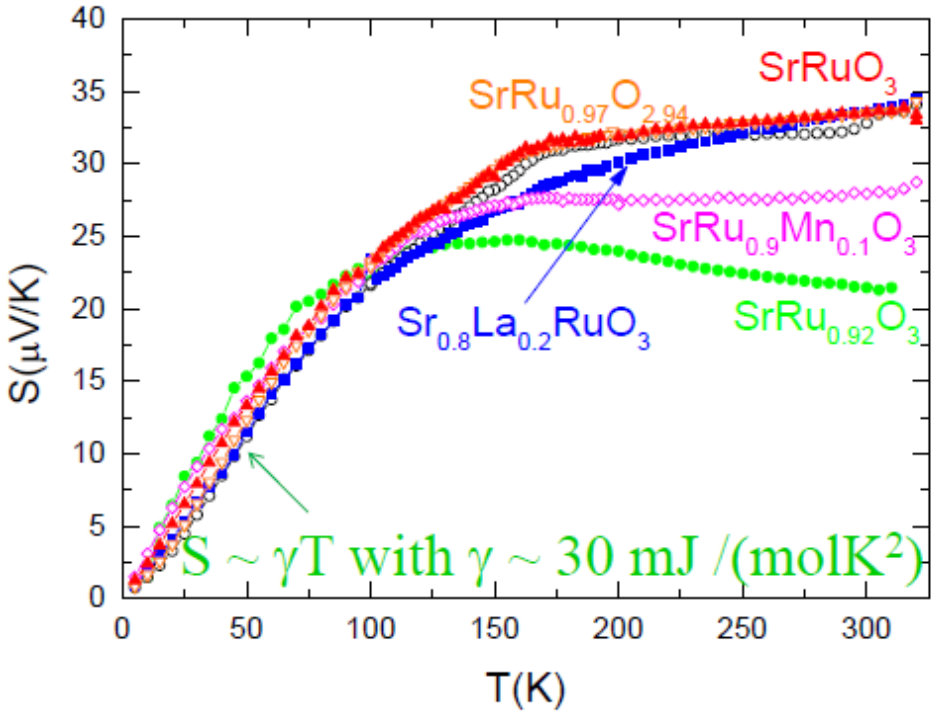

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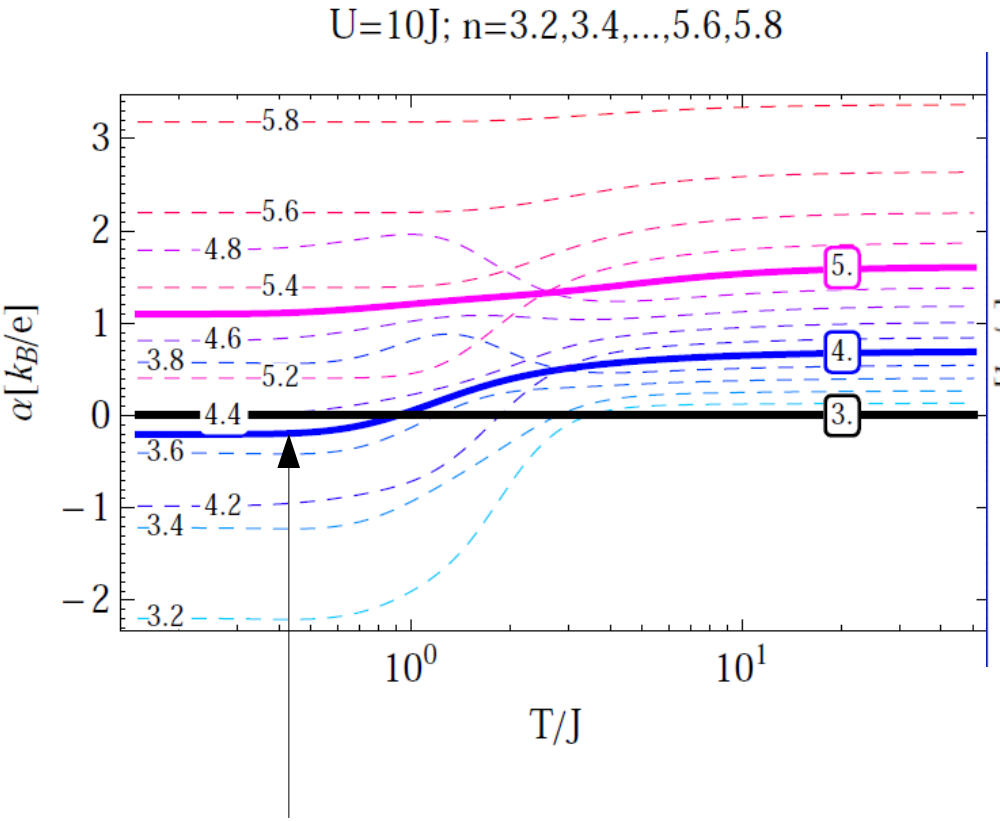
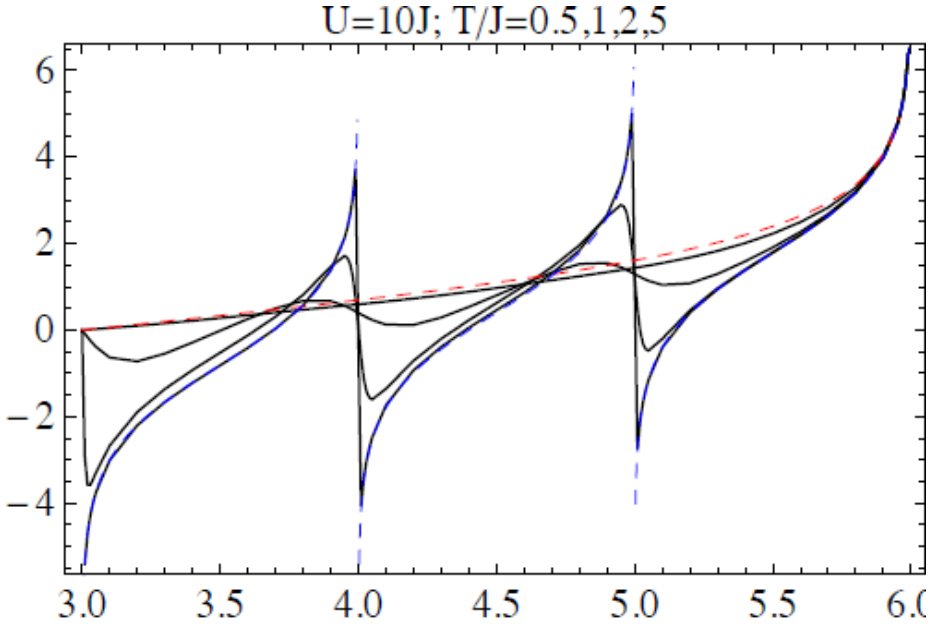
(slide from S. Hebert)

t_{2g} atom

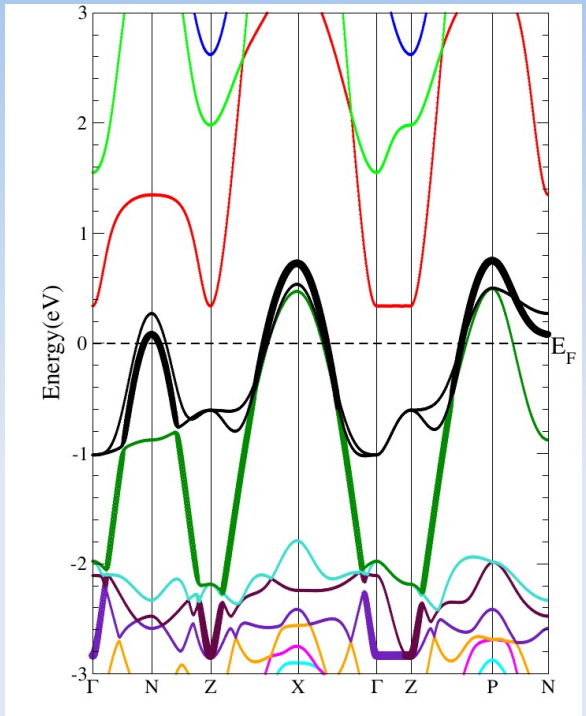
$$H_{\text{int}} = (U - 3J)\frac{\hat{N}(\hat{N}-1)}{2} - 2J\vec{S}^2 - \frac{1}{2}J\vec{T}^2$$

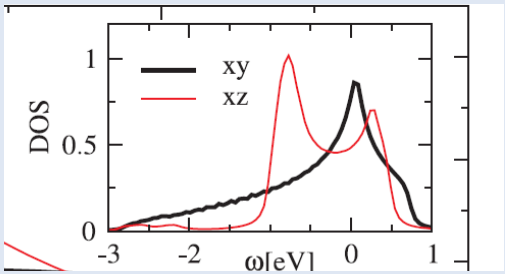




Negative result if orbital moments are kept!

Sr_2RuO_4: el. structure



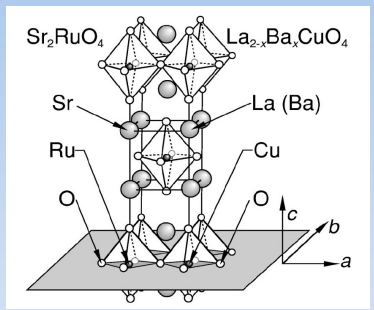


In ionic picture, 4 electrons on Ru; crystal field splitting → t_{2g} orbitals: xy and degenerate xz, yz

Wide xy band (2d like ϒ sheet); narrower xz, yz quasi 1d.

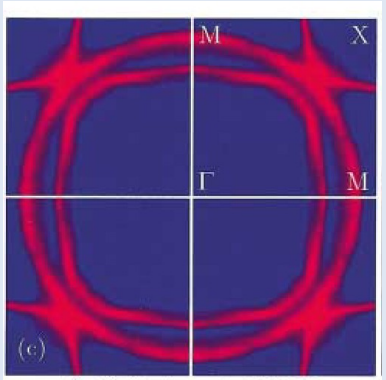
Fermi surfaces of DFT, quantum oscillations, ARPES agree quite well

Mackenzie et al, PRL'96



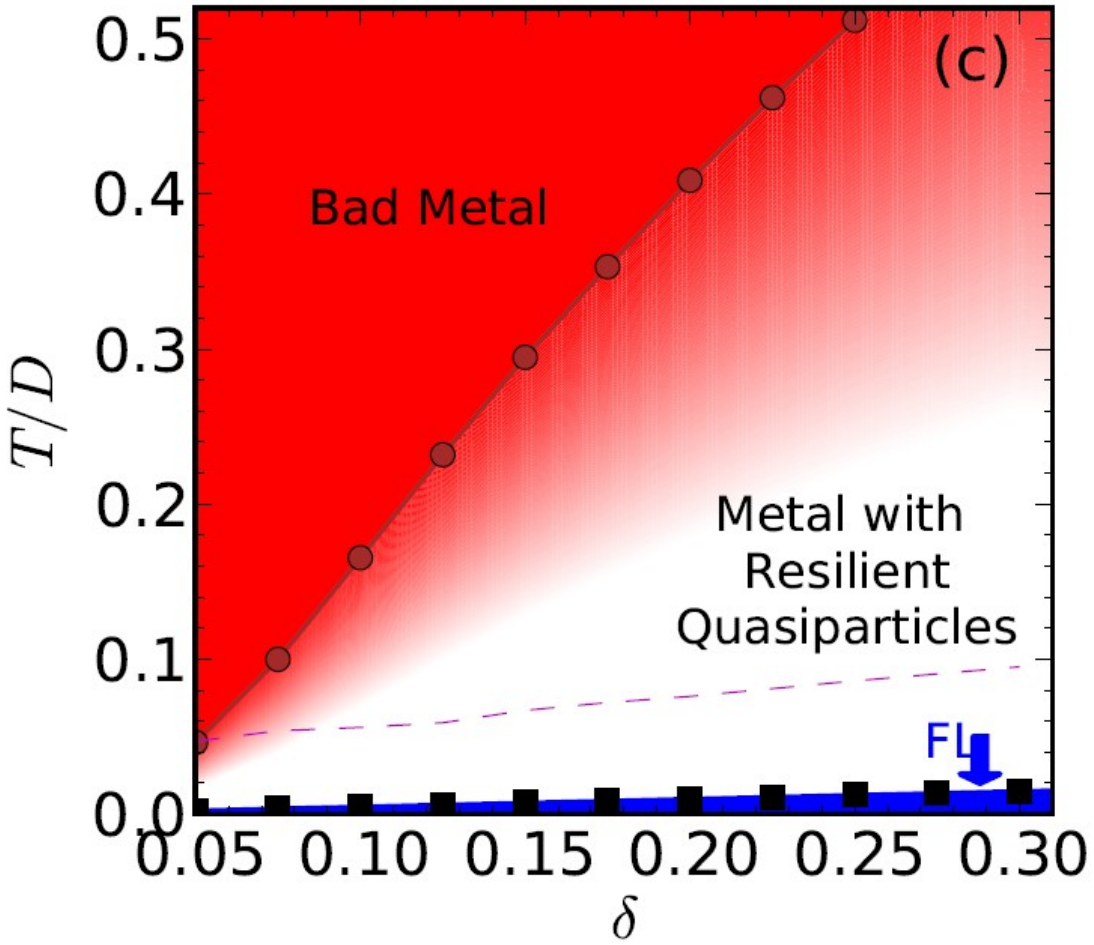
Oguchi, PRB'95
Singh, PRB'95

Damascelli, Shen et al., PRL'00



“Phase” diagram

- QPs persit until close to MIR (~ Brinkman-Rice scale δD)



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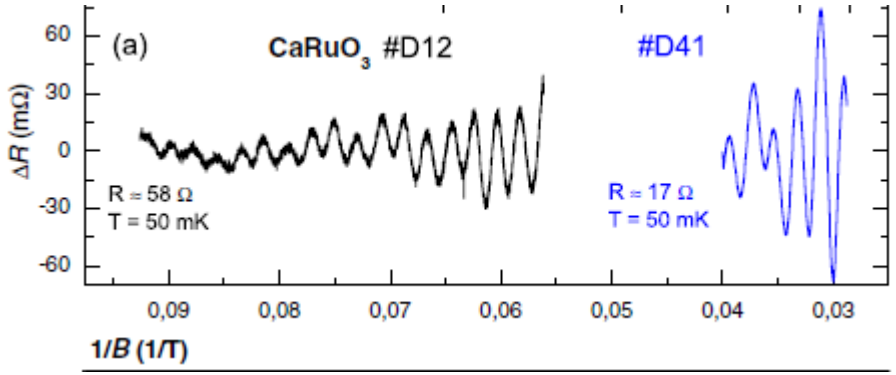
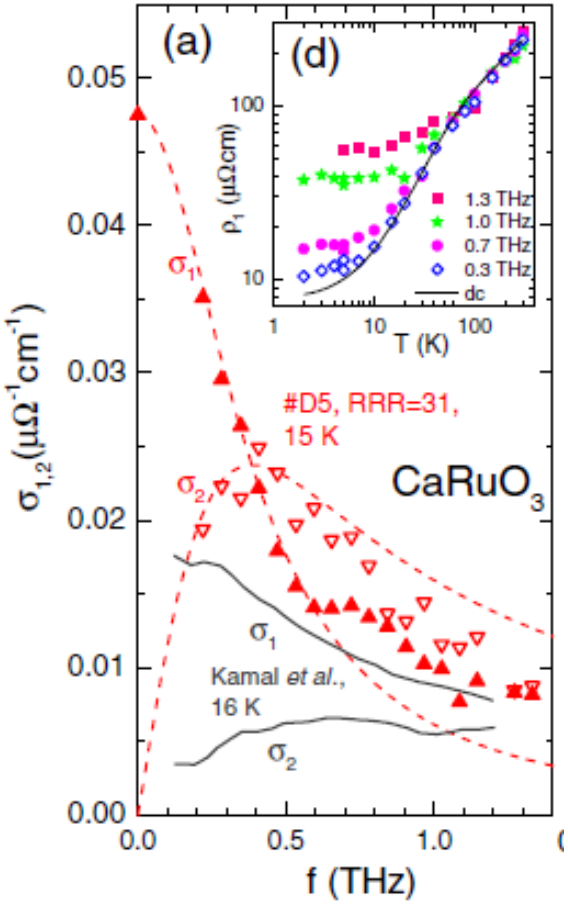


Schneider et al., PRL'14



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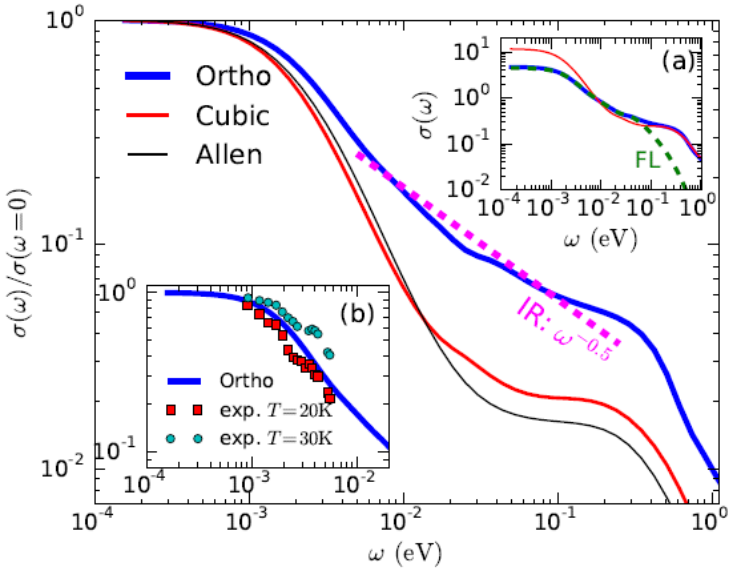

Dang, JM, Georges, Millis, arXiv'15