# Recent insights on the normal state of $Sr_2RuO_4$

## High-resolution photoemission and Hall coefficient

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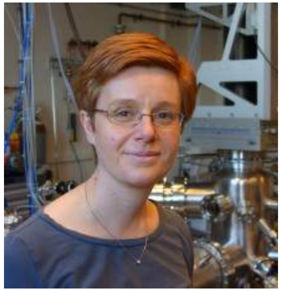
Workshop – Collège de France - 11.06.2019

# Acknowledgments – Experiment / Theory collaboration

Felix Baumberger



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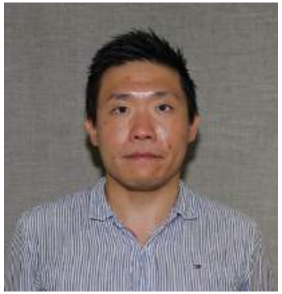

Minjae Kim
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**High-resolution photoemission on $Sr_2RuO_4$ reveals correlation-enhanced effective spin-orbit coupling and dominantly local self-energies**
A. Tamai, **MZ**, E. Rozbicki, E. Cappelli, S. Ricco, A. de la Torre, S. McKeown Walker, F. Y. Bruno, P.D.C. King, W. Meevasana, M. Shi, M. Radovic, N.C. Plumb, A.S. Gibbs, A.P. Mackenzie, C. Berthod, H. Strand, M. Kim, A. Georges, F. Baumberger
**PRX 9, 021048 (2019)**

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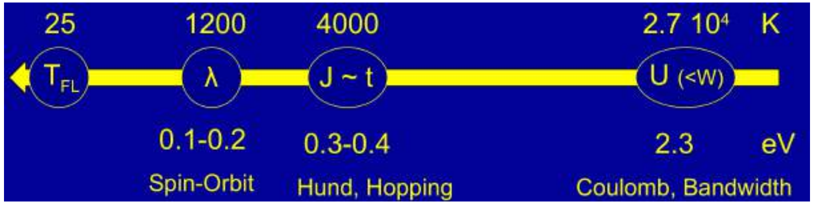
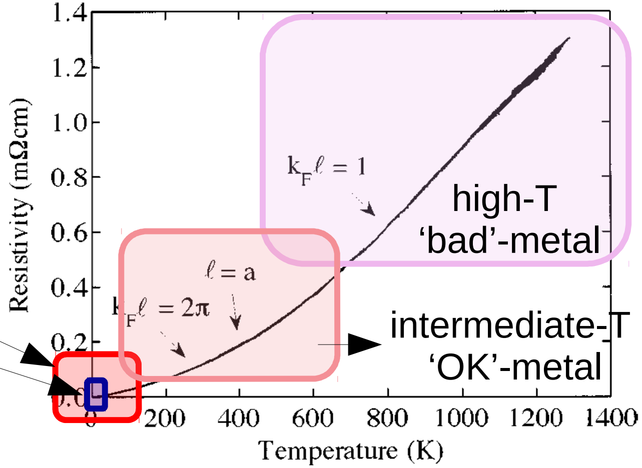
# $Sr_2RuO_4$ – one of the best studied TMO

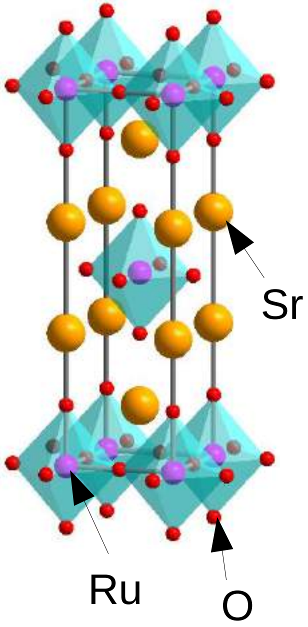
- huge high-quality crystals
- investigated with many experimental techniques




Rich physics:

- Hund's physics, spin-orbit coupling
- van-Hove singularity close to $E_F$
- Fermi liquid ($T_{FL}$ ~ 25 K)
- superconductor ($T_c$ ~ 1.5 K)

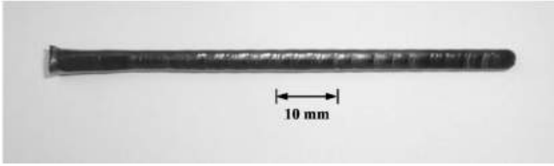



"Looks" simple:

- quasi-2D tetragonal structure
- well defined low-energy subspace
- no structural or magnetic transitions

**BUT still challenging to understand in all details.**

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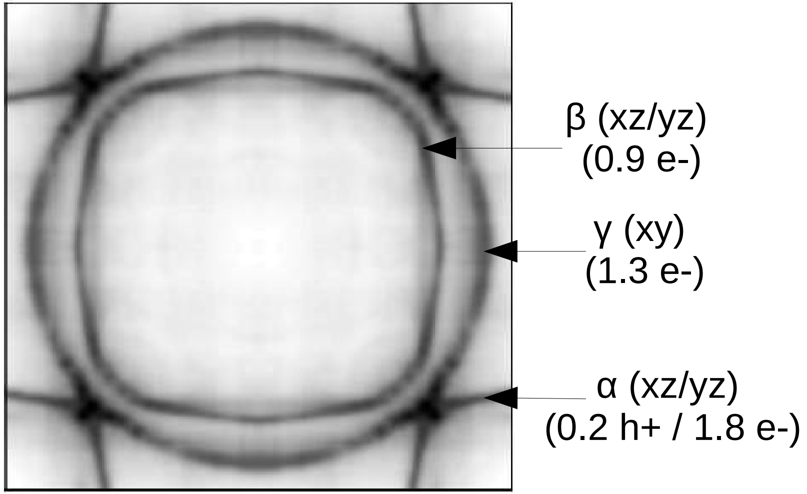
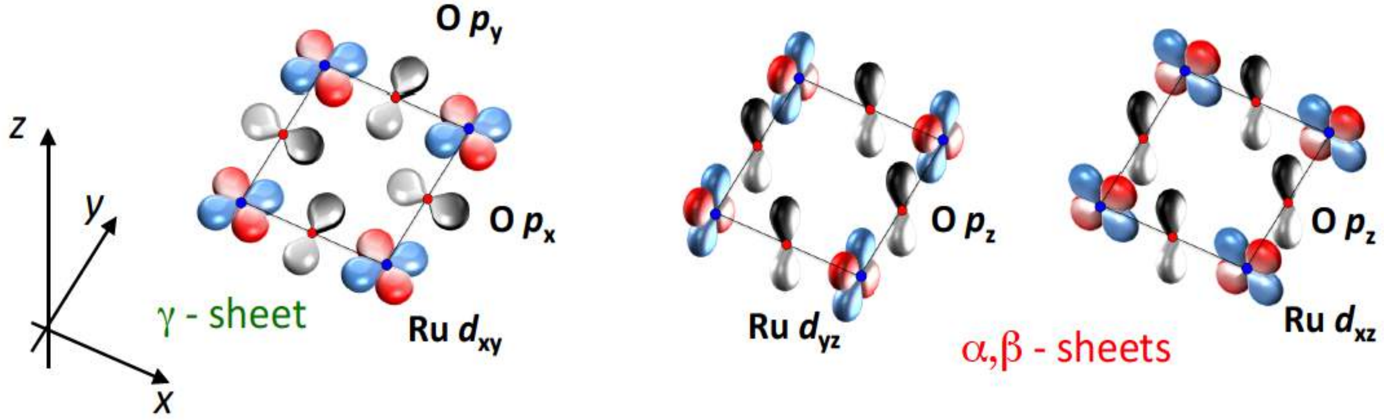
Review Articles:
A. Mackenzie and Y. Maeno, RMP 75, 657 (2003), C. Bergemann, Adv. Phys. 52, 639 (2003)

# Fermi surface basics

- 4 electrons in Ru-$t_{2g}$ shell
- $d_{xy}$ yield a quasi 2D γ-sheet
- $d_{xz/yz}$ have directional hopping along x/y yielding the α/β sheets




A. Damascelli, PRL 85, 5194 (2000)

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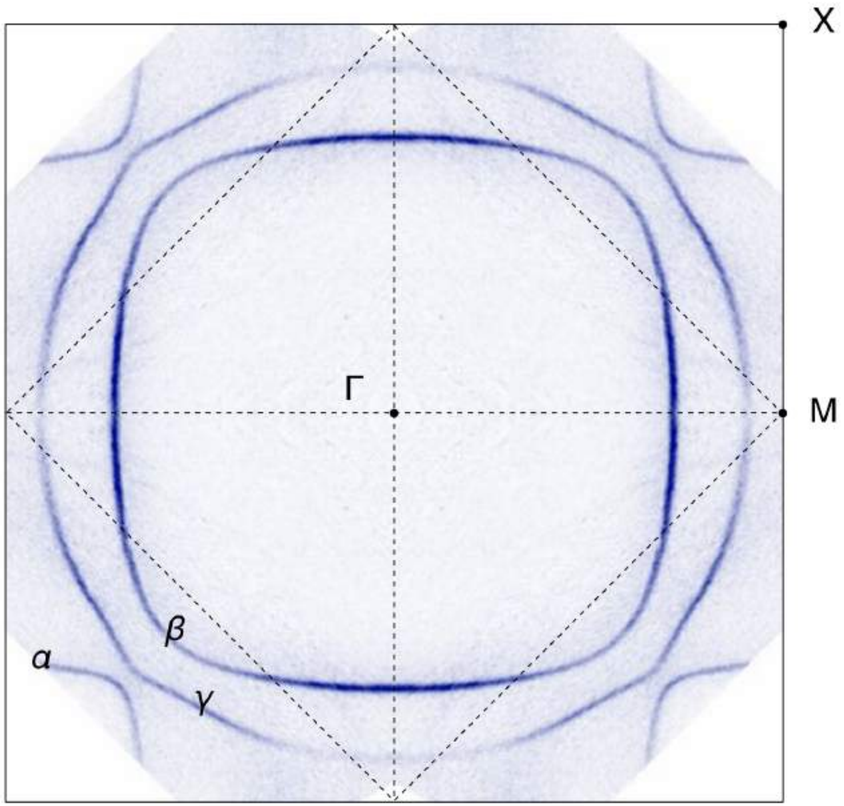
# High-resolution laser-ARPES Fermi surface

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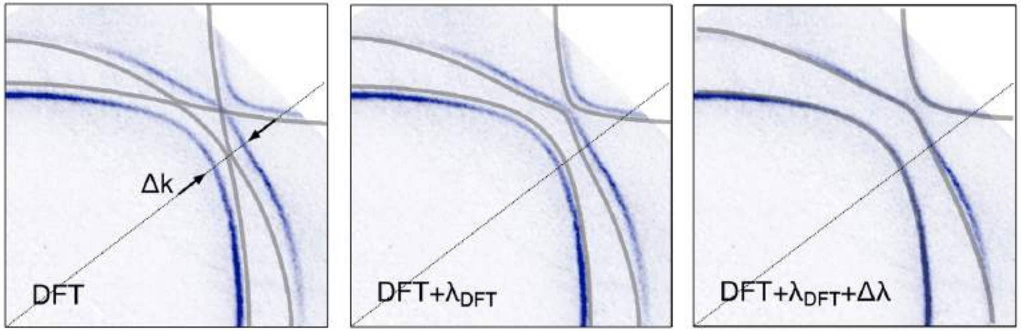
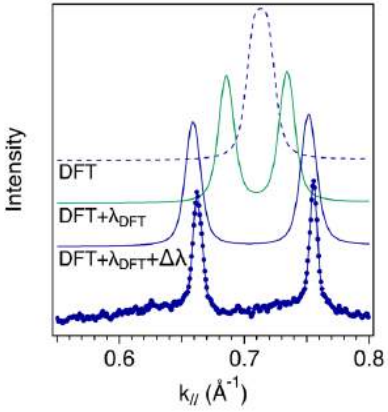
# Fermi surface reveals enhancement of spin-orbit coupling (SOC)

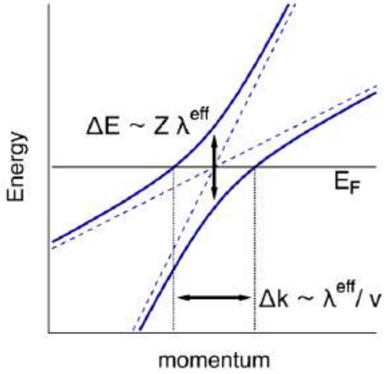



Cannot be explained by tuning crystal-field splitting

Enhanced-SOC theoretically predicted

Liu et al. PRL 101, 026408 (2008),
Zhang et al. PRL 116, 106402 (2016),
Kim et al. PRL 120, 126401 (2018)




Extract SOC from Fermi surface splitting:

$$\Delta k = \lambda^{\rm eff}/v$$

$$\lambda^{\rm eff} = \lambda_{\rm DFT}\Delta k^{\rm QP}/\Delta k^{{\rm DFT}+\lambda_{\rm DFT}} = 205(20)\ \ {\rm meV}$$

$$\lambda_{\rm DFT} = 100\,{\rm meV}$$

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Using energy splitting at Γ: C. N. Veenstra, et al., PRL 112, 127002 (2014)

# Non-interacting reference Hamiltonian

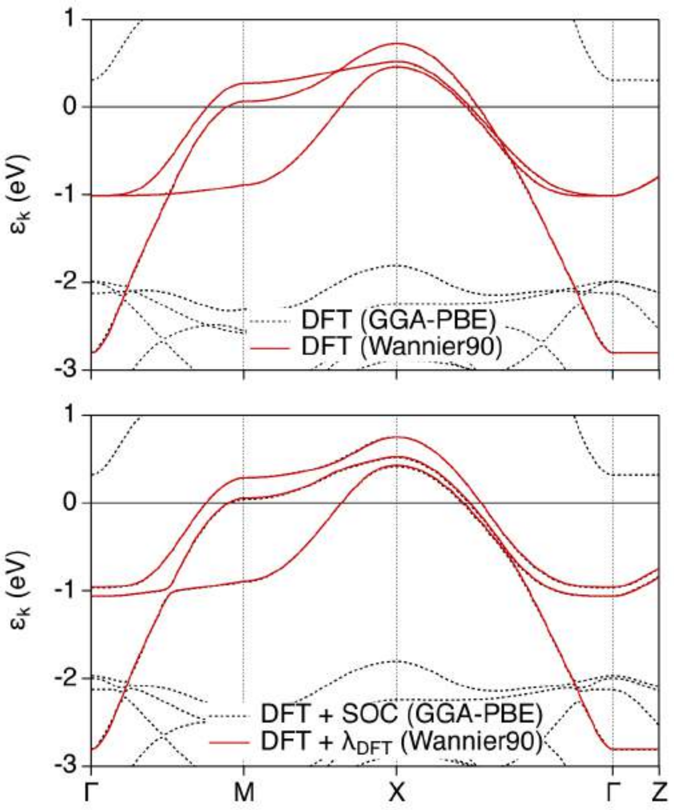
1) DFT without SOC (Wien2k, GGA-PBE)

2) Construct Hamiltonian $\hat{H}^{\mathrm{DFT}}$ from maximally-localized Wannier functions for the three-orbital $t_{2g}$ subspace

3) Add static SOC:

$$\hat{H}^{\mathrm{SOC}}_{\lambda} = \begin{pmatrix} \varepsilon_{xy} & 0 & 0 & 0 & \frac{\lambda_{xy}}{2} & \frac{i\lambda_{xy}}{2} \\ 0 & \varepsilon_{yz} & -\frac{i\lambda_z}{2} & -\frac{\lambda_{xy}}{2} & 0 & 0 \\ 0 & \frac{i\lambda_z}{2} & \varepsilon_{xz} & -\frac{i\lambda_{xy}}{2} & 0 & 0 \\ 0 & -\frac{\lambda_{xy}}{2} & \frac{i\lambda_{xy}}{2} & \varepsilon_{xy} & 0 & 0 \\ \frac{\lambda_{xy}}{2} & 0 & 0 & 0 & \varepsilon_{yz} & \frac{i\lambda_z}{2} \\ -\frac{i\lambda_{xy}}{2} & 0 & 0 & 0 & -\frac{i\lambda_z}{2} & \varepsilon_{xz} \end{pmatrix}$$

4) Use $\lambda_{\mathrm{DFT}}$ = 100 meV and $\lambda_{\mathrm{DFT}}+\Delta\lambda$ = 200 meV




Software packages:

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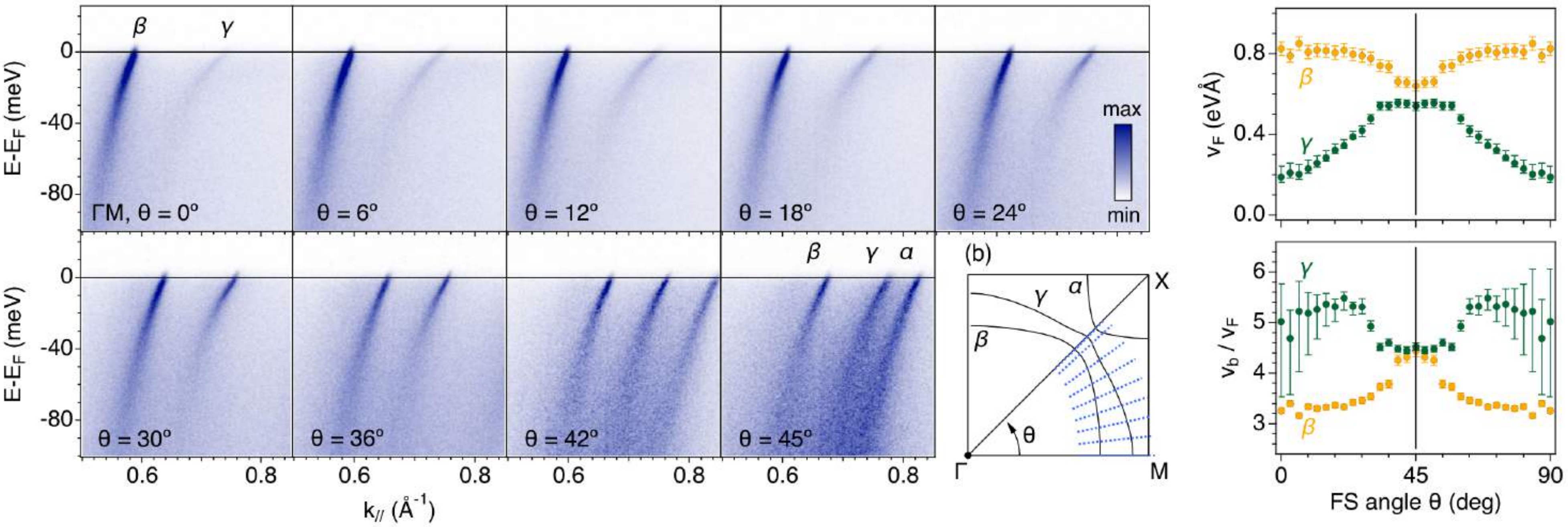
# Quasiparticle dispersion along several angular cuts

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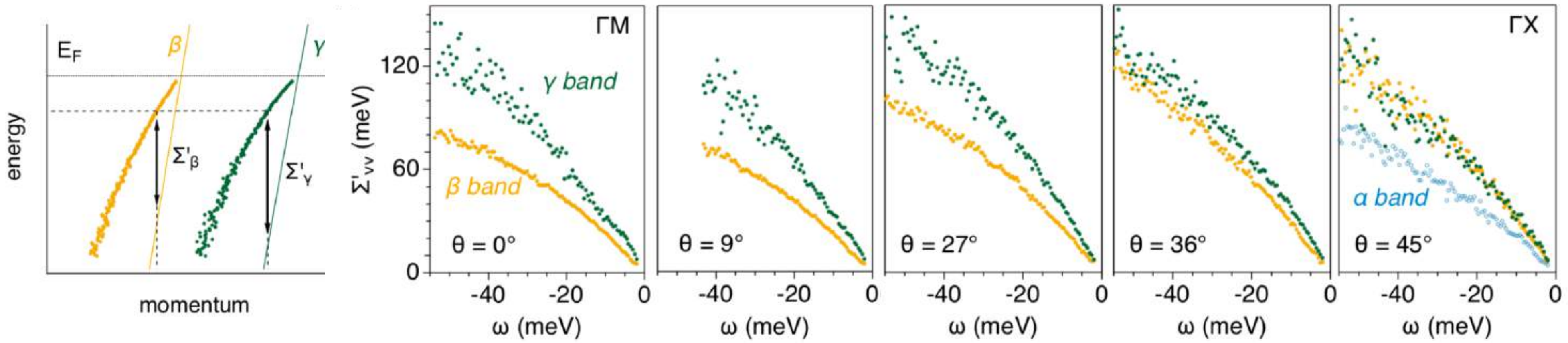
## Self-energy in band basis ( $\nu = \{\alpha, \beta, \gamma\}$ )

$$G^{-1}_{\nu\nu'}(\omega, \boldsymbol{k}) = [\omega + \mu - \varepsilon_\nu(\boldsymbol{k})]\,\delta_{\nu\nu'} - \Sigma_{\nu\nu'}(\omega, \boldsymbol{k})$$

Quasiparticle dispersion: $\det\left[(\omega - \varepsilon_\nu(\boldsymbol{k}))\,\delta_{\nu\nu'} - \Sigma'_{\nu\nu'}(\omega, \boldsymbol{k})\right] = 0$

Assume diagonal self-energy: $\omega - \varepsilon_\nu(\boldsymbol{k}^\nu_{\max}(\omega)) = \Sigma'_\nu(\omega, \theta)$ $\boldsymbol{k}^\nu_{\max}(\omega)$ … maximum of dispersion




Self-energy strongly depends on angle Θ (i.e. momentum) in band basis.

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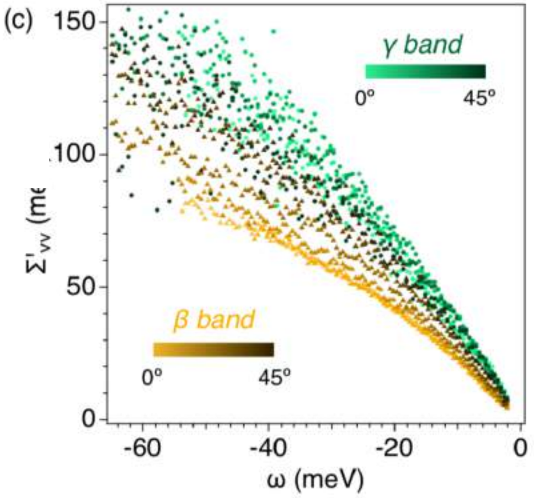
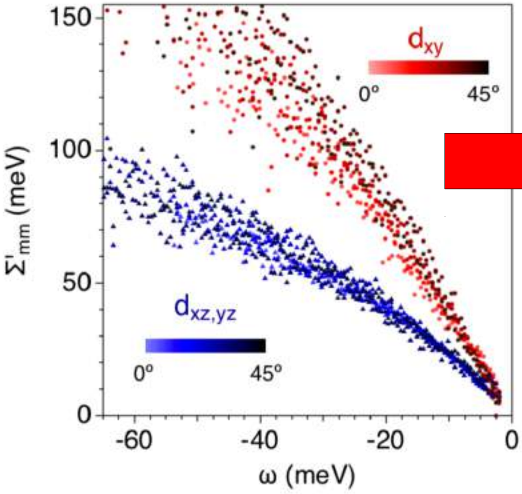
# Self-energy in orbital basis (m = {xy, xz, yz})

Work in orbital basis $|\chi_m(\boldsymbol{k})\rangle$ ( i.e. Wannier functions )

Extract self-energy with: $$\det\left[(\omega - \Sigma'_m(\omega, \theta_{\boldsymbol{k}}))\delta_{mm'} - \hat{H}^0_{mm'}(\boldsymbol{k})\right] = 0$$




Collapse of self-energies

**Direct experimental justification of "locality ansatz" à la DMFT**

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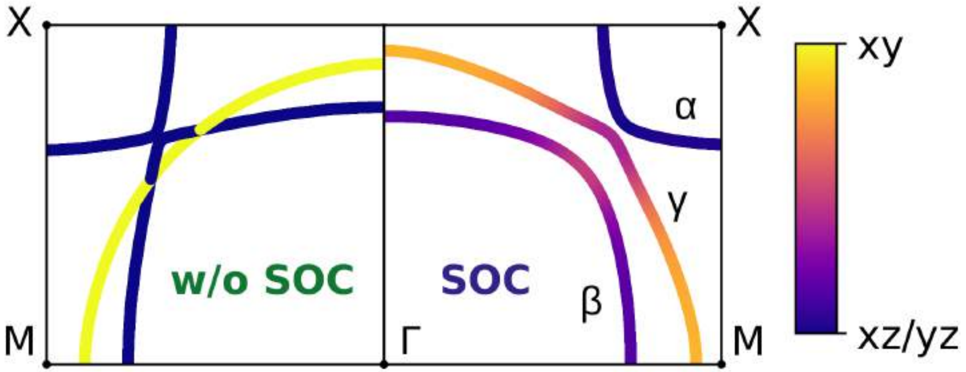
# Angular dependence of orbital content of quasiparticles

$$\Sigma_{\nu\nu'}(\omega,\boldsymbol{k}) = \sum_{mm'} U^*_{m\nu}(\boldsymbol{k})\, \Sigma_{mm'}(\omega,\boldsymbol{k})\, U_{m'\nu'}(\boldsymbol{k}) \cong \sum_{m} U^*_{m\nu}(\boldsymbol{k})\, \Sigma_{m}(\omega)\, U_{m\nu'}(\boldsymbol{k})$$

with matrix elements $U_{m\nu}(\boldsymbol{k}) = \langle \chi_m(\boldsymbol{k}) | \psi_\nu(\boldsymbol{k}) \rangle$

reflecting the orbital-content $|U_{m\nu}(\boldsymbol{k})|^2$.

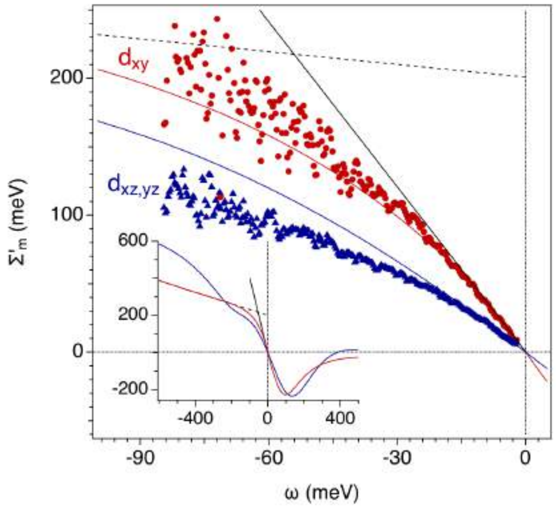



Ansatz made by DMFT

$\Sigma_{\nu\nu'}(\omega,\boldsymbol{k})$ gains ***k***-dependence due to ***k***-dependence of $U_{m\nu}(\boldsymbol{k})$

Orbital-content of quasiparticle states is strongly angular dependent (SOC)

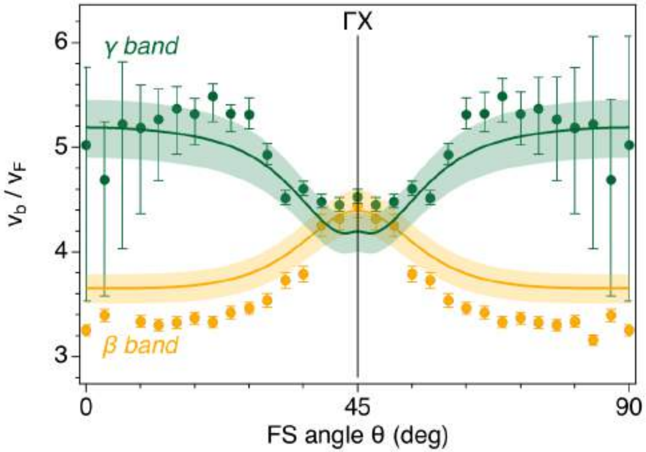
# Comparison to DMFT



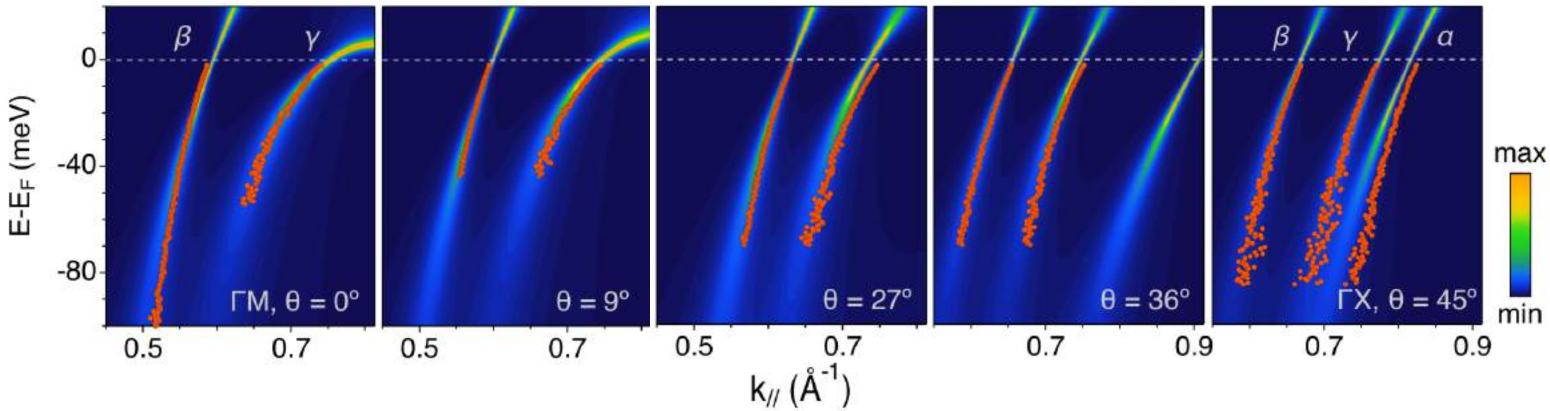

Electronic origin of self-energy “kinks”

Excellent agreement up to 30 meV

Discrepancies at higher energies?




angular-dependent Z




@ 29 K ($T_{FL}$ ~ 25 K):

$1 / Z_{xy} = 5.5$

$1 / Z_{xz/yz} = 3.3$

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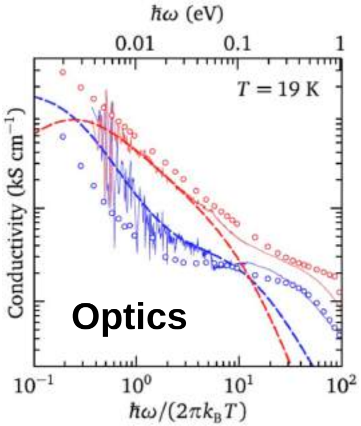
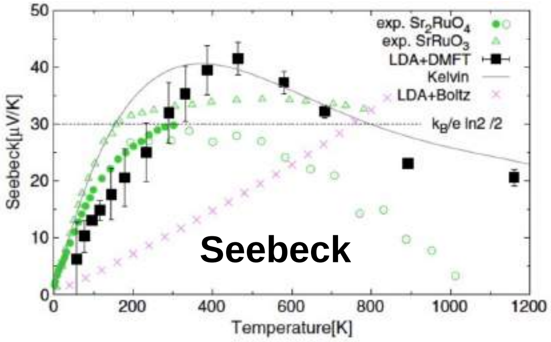
# Other successes of DMFT for $Sr_2RuO_4$

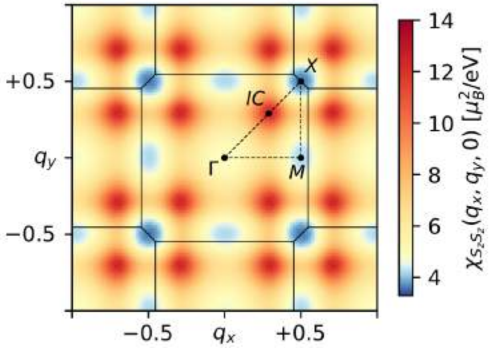



D. Stricker, et al.,
PRL 113, 087404 (2014)

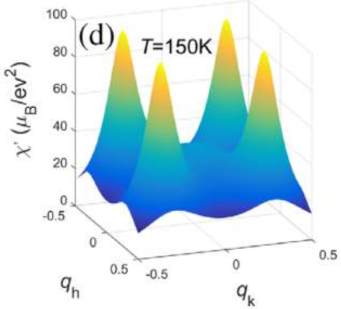



J. Mravlje, et al.,
PRL 117, 036401 (2016)

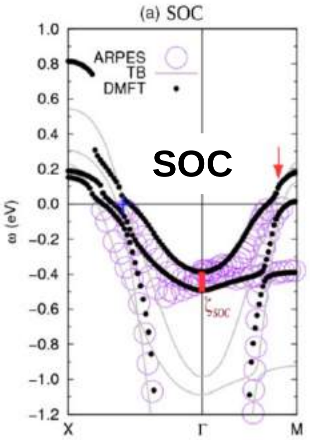
Susceptibility



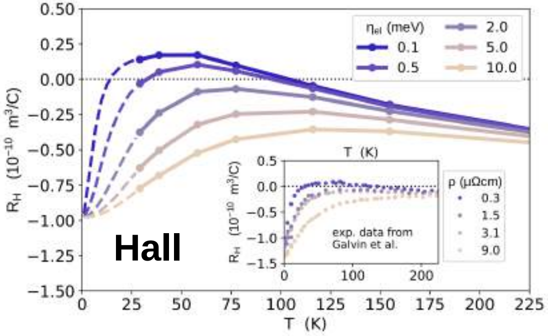

H. Strand, et al.,
arXiv:1904.07324 (2019)



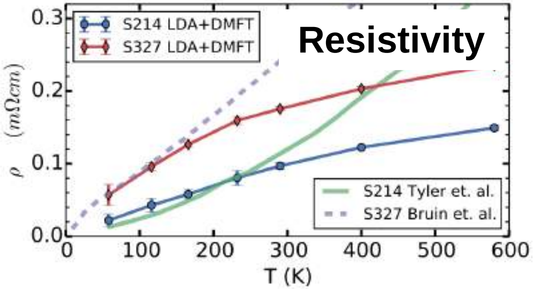

INS: P. Steffens, et al.,
PRL 122, 047004 (2019)




M. Kim, et al.,
PRL 120, 126401 (2018)




**MZ**, et al., arXiv:1902.05503 (2019)




X. Deng et al.,
PRL 116, 256401 (2016)

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# Overview

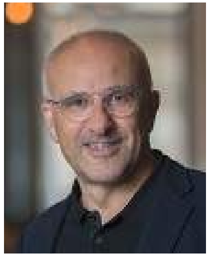
- High-resolution laser-ARPES reveals quasiparticle physics of $Sr_2RuO_4$ with unprecedented accuracy
- Enhancement (by factor of ~ 2) of spin-orbit coupling by electronic correlations confirmed
- Momentum independent ansatz (DMFT) for self-energy of each orbital works well
- Angular dependence of quasiparticle properties largely explained by angular dependence of orbital content (controlled by spin-orbit)

**Hall coefficient signals orbital differentiation in the Hund's metal $Sr_2RuO_4$**

**MZ**, J. Mravlje, M. Aichhorn, O. Parcollet, A. Georges

**arXiv:1902.05503**



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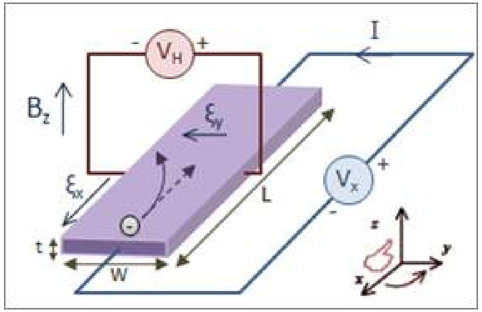
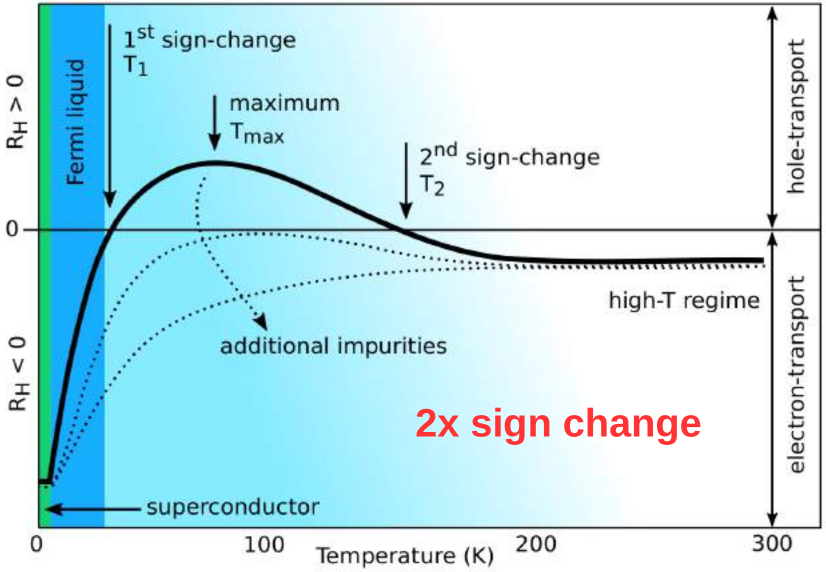
# Hall coefficient $R_H$



Wikipedia

simple metal,
one type of carrier:

$$R_H = -\frac{1}{ne}$$




→ signature of multi band/orbital nature
→ interplay of electron and hole-like Fermi surface sheets
→ scattering rates strongly T and Fermi surface sheet dependent

Experiments: N. Shirakawa et al., JPSJ 64, 1072-1075 (1995),
A. P. Mackenzie et al., PRB 54, 7425, (1996), L. M. Galvin et al., PRB 63, 161102 (2001)
Earlier theoretical works: C. Noce, et al. PRB 59, 2659 (1999),
I. Mazin, et al., PRB 61, 5223 (2000), C. Noce, et al. PRB 62, 9884 (2000)

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# Scattering rate ratios

Boltzmann transport theory:

- constant isotropic scattering time approx.
  $\sigma_{xx} = \sigma_{yy} \sim \tau = 1/\eta$ and $\sigma_{xy} \sim \tau^2$

  $\rightarrow R_H = \dfrac{\sigma_{xy}}{\sigma_{xx}\sigma_{yy}}$ does not depend on η

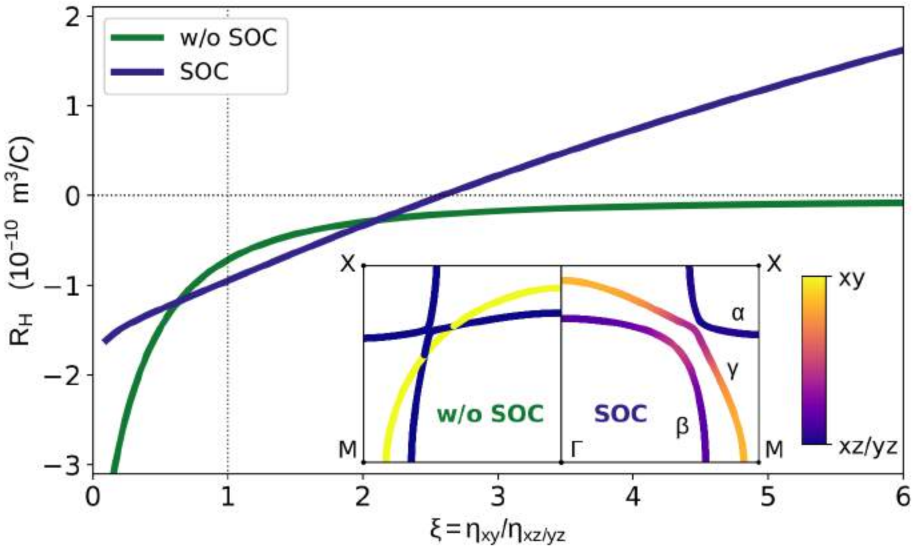
- orbital-dependent scattering rates $\eta_{xy}$ and $\eta_{xz} = \eta_{yz}$
  $\rightarrow R_H$ depends on ratio $\xi = \eta_{xy} / \eta_{xz/yz}$

- $R_H$ monotonically increases ratios

- SOC essential for $R_H > 0$

- T-dependence of scattering rate ratios
  to explain the experiments?

$$\eta_\nu(\mathbf{k}) = \sum_m \left| \langle \chi_m(\mathbf{k}) \,|\psi_\nu(\mathbf{k})\rangle \right|^2 \eta_m$$




BoltzTraP2 software package:
G. K. Madsen et al., CPC 175, 67 (2006)
G. K. Madsen et al., CPC 231, 140 (2018)

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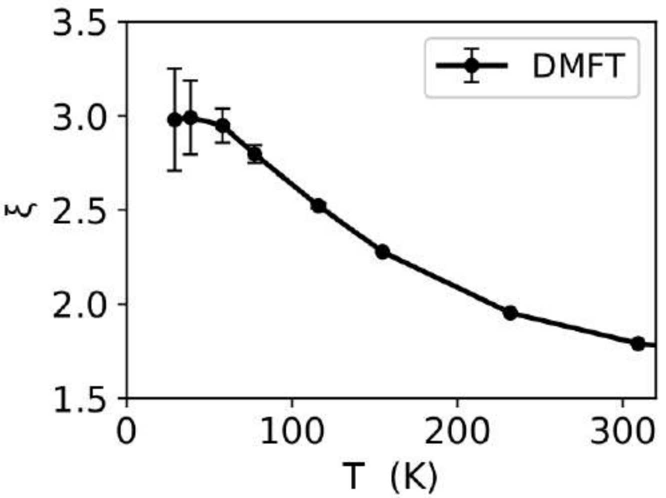
# Inelastic electron-electron scattering

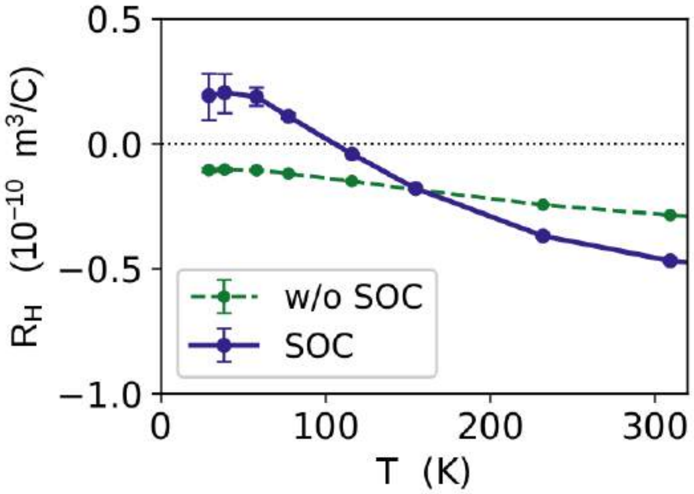



DFT+DMFT:

- $\eta_{xy} > \eta_{xz/yz}$
- orbital-differentiated coherence-incoherence crossover
- ξ strongly T-dependent

Crossover from incoherent electrons to coherent Fermi liquid

J. Mravlje, et al.,PRL 106, 096401 (2011),
X. Deng, et al., PRL, 116, 256401 (2016)




→ turns $R_H$ positive with zero crossing at 120 K

In FL ratio ξ is independent of T:

**Why does $R_H$ turn negative again at low T?**

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# The full picture

Inelastic scattering from DMFT @ 29 K:

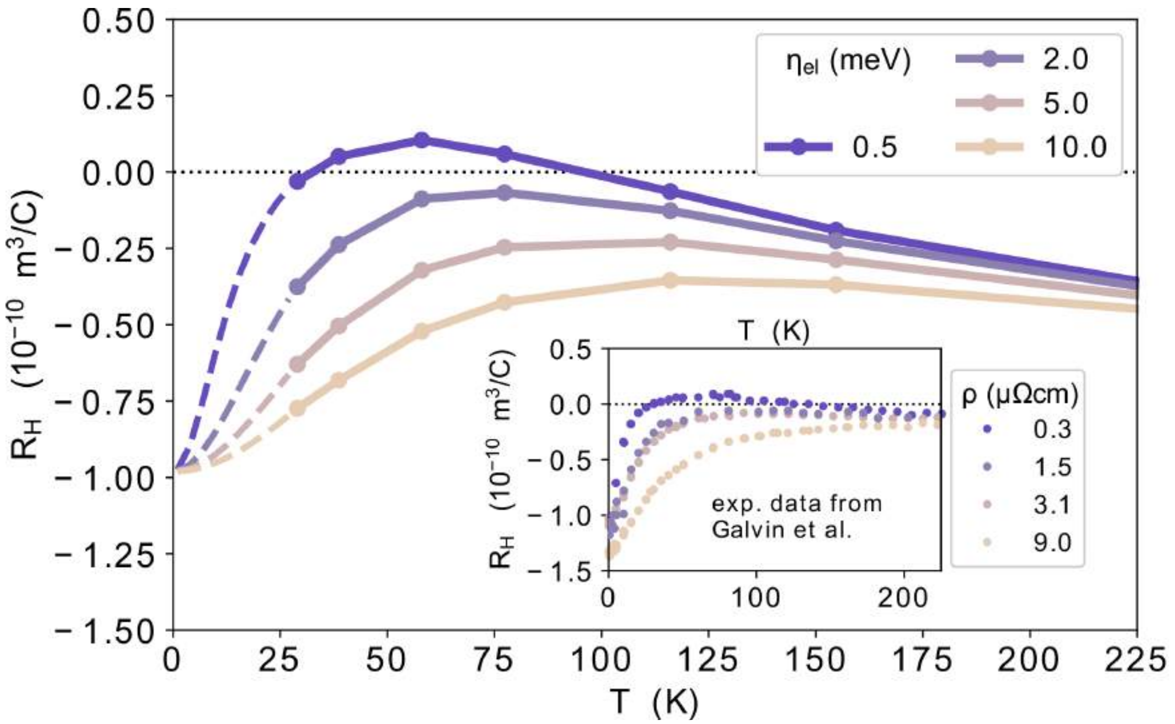
$\eta_{xy}$ = 5.5 meV

$\eta_{xz/yz}$ = 1.9 meV

Add isotropic impurity scattering:

$$\eta_m = \eta^{inel}_m + \eta^{el}$$

Dominates $R_H$ at: high-T ($\eta^{inel}_m$), low-T ($\eta^{el}$)




Experiment:
L.M. Galvin et al., PRB 63, 161102 (2001)

Sign-changes are signature of 2 cross-overs:

- coherent-to-incoherent regime
- inelastic-to-elastic scattering

Well separated temperature scales in clean samples.

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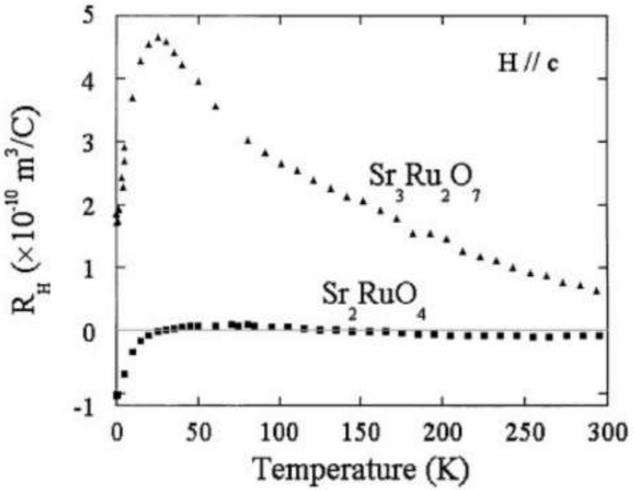
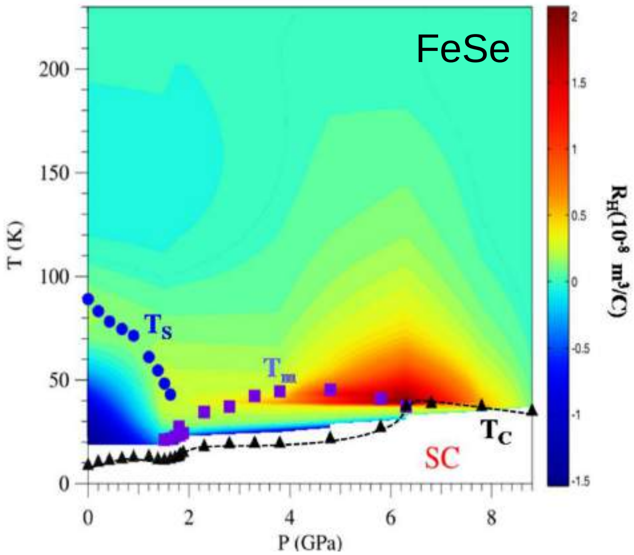
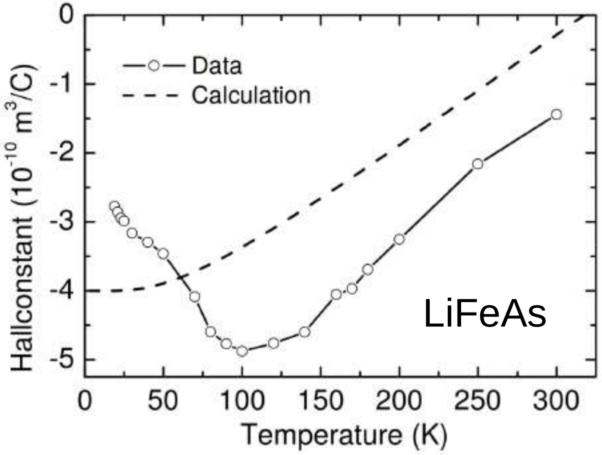
# Ingredients for T-dependence of $R_H$

- Fermi surface sheets of different or mixed orbital character
- T and orbital dependent scattering rates
- Balanced electron/hole contributions → sign-changes can emerge

Other Hund's metals:




R. Perry, et al., Phys. B 284, 1469 (2000); J. P. Sun, et al., PRL 118, 147004 (2017); O. Heyer, et al., PRB 84, 064512 (2011)

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