

# Hidden Order in URu<sub>2</sub>Si<sub>2</sub> a DMFT Perspective

G. Kotliar

Work with Kristjan Haule and Jihoon Shim at Rutgers University.

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Thanks also to J. Mydosh, J. Denlinger, J. Allen, J.C.S Davis

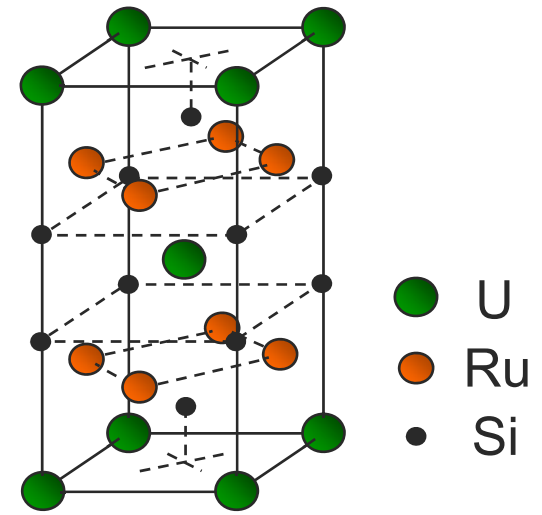
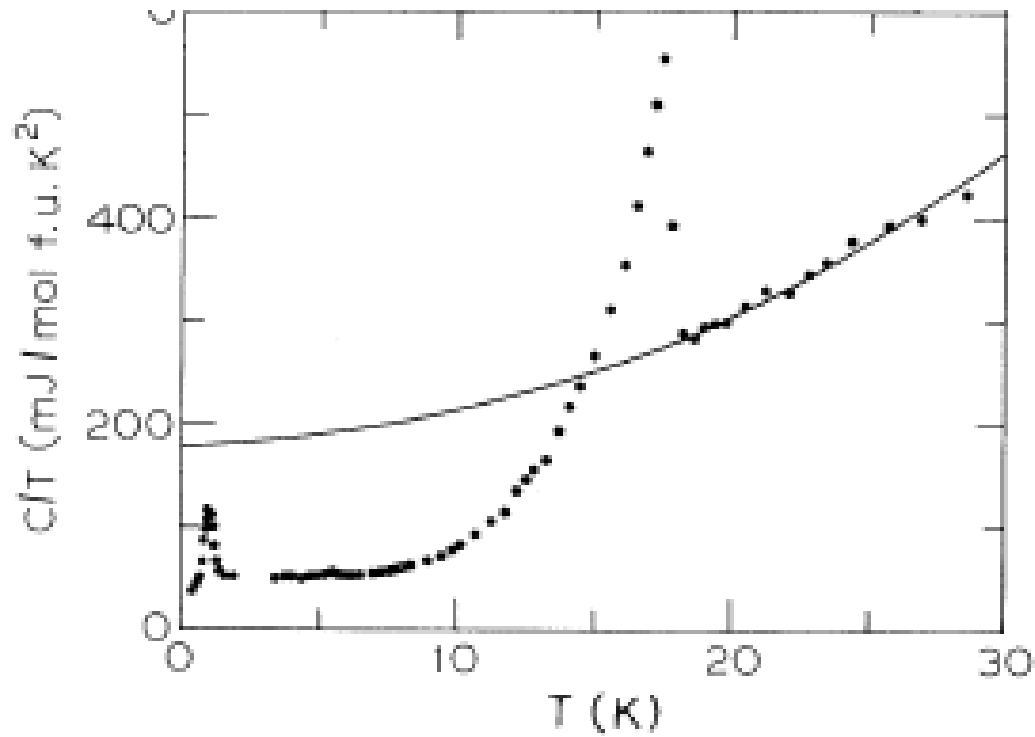


Presented at the Technion June 14<sup>th</sup> 2008  
Strongly Correlated Quantum Phases

# Outline

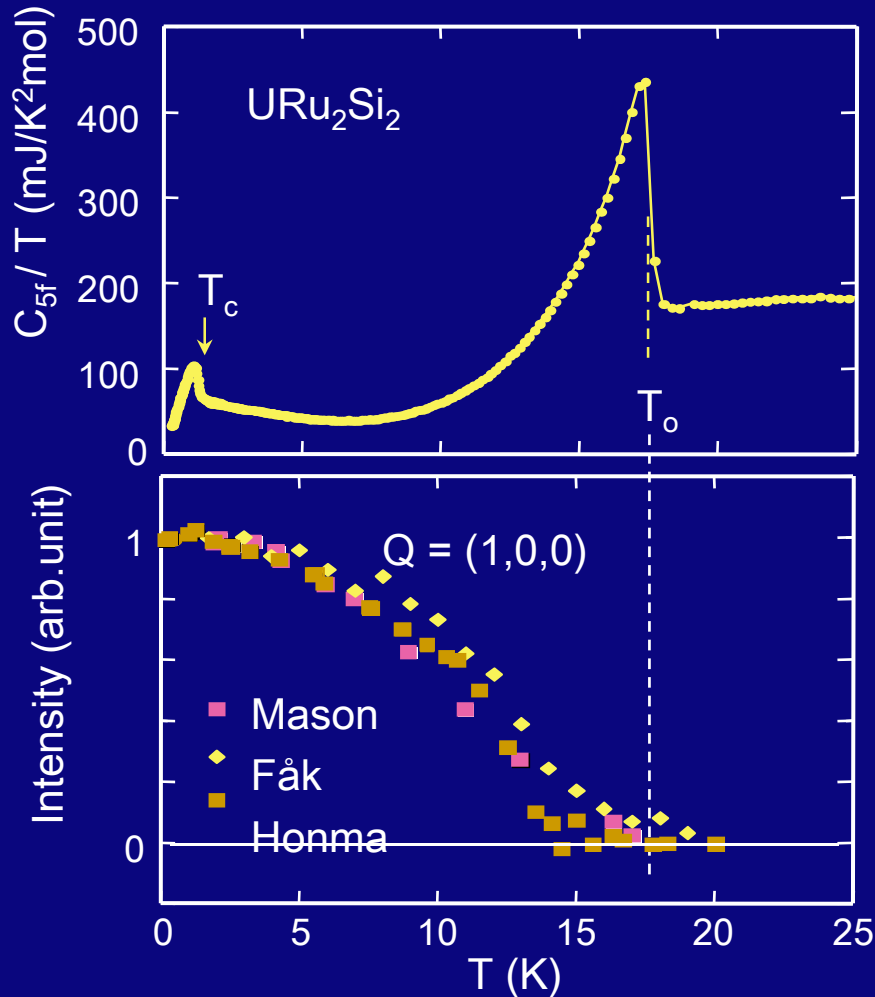
- **Motivating the material. URu<sub>2</sub>Si<sub>2</sub> , heavy fermions and Hidden Order Problem [ Definitely a Strongly Correlated Quantum Phase]**
- **DMFT.**
- **The “easy” case, Ce 115’s.**
  
- **URu<sub>2</sub>Si<sub>2</sub> Insights into the Hidden Order from DMFT.**
- **Conclusions: Motivating the study, a methodological**
- **perspective.**

# A Hidden Order The CMT dark matter problem. (A. Schofield)



$\text{URu}_2\text{Si}_2$ : T. T. M. Palstra, A. A. Menovsky, J. van den Berg, A. J. Dirkmaat, P. H. Kes, G. J. Nieuwenhuys and J. A. Mydosh *Physical Review Letters* **55**, 2727 (1985)

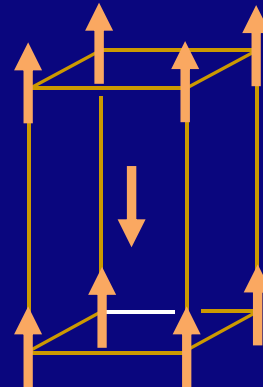
# Neutron Scattering. Specific heat vs. magnetic Bragg-peak intensity. $T_c$ 's.



$$S_{\text{mag}} \sim 0.2 R \ln 2$$



$$\mu_{\text{ord}} \sim 0.01 - 0.04 \mu_B$$

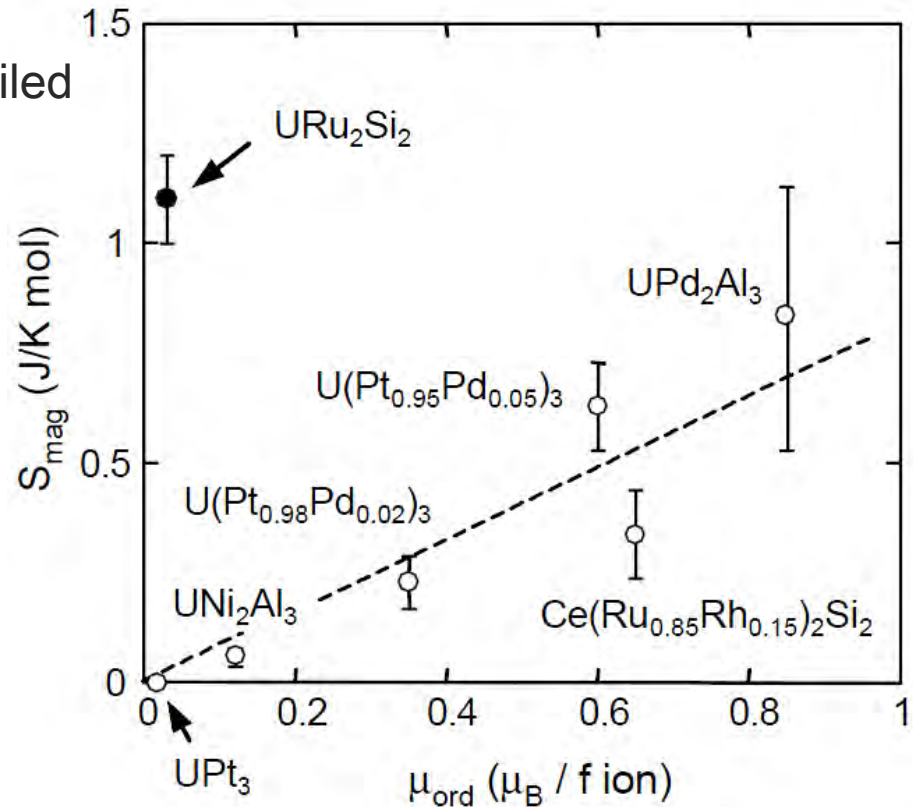


Type-I AF  $\xi_c \sim 100 \text{ \AA}$   
 $\xi_a \sim 300 \text{ \AA}$

# Hidden order

- Moment is tiny  
(likely small admixture of AFM phase)
- Large loss of entropy can not be reconciled with small moment
- Other primary symmetry breaking.

WHICH?



# Heavy Fermion Problem (more general).

- **Intermetallic compounds.**
- **Bare (high energy) degrees of freedom: open shell ions , i. e. Ce, U and conduction electrons.**
- **Low energy degrees of freedom. Quasiparticles composed of those degrees of freedom sometimes form a heavy Ferm liquid.  $M^*/M \sim 50-1000$ .**
- **Large variety of ground states, superconducting, magnetic , etc.**

# Uranium Heavy Fermions

## Relevance of the Kondo effect.

Dan Cox,  $5f^2$  configuration + crystal fields select a ground which is a non Kramers doublet.

Multichannel Quadrupolar Kondo effect in U . Non Fermi liquid.

Early work of D. Cox. U [Phys. Rev. Lett. 59, 1240 \(1987\)](#)

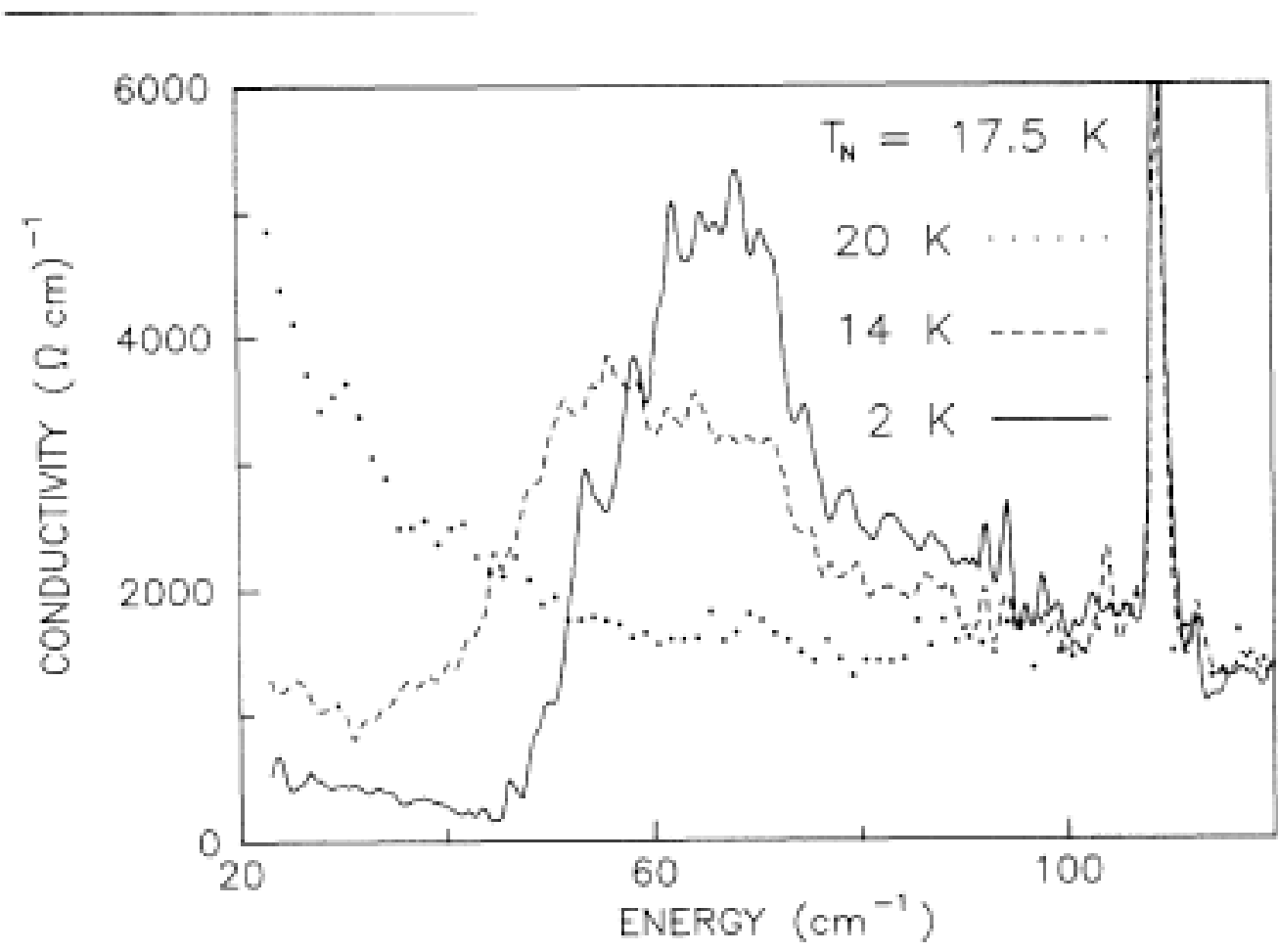
Other possibilities, magnetic Kondo effect when U is  $f^3$

When a heavy Fermi liquid is formed, what is the volume of the Fermi surface? Luttinger theorem: it contains  $n_f + n_{\text{cond}}$  electrons. Mod 2.

For  $f^1$  configuration ( Cerium ) the Fermi surface expands as the temperature is reduced. The  $T=0$  Fermi surface is well approximated by the LDA Fermi surface.

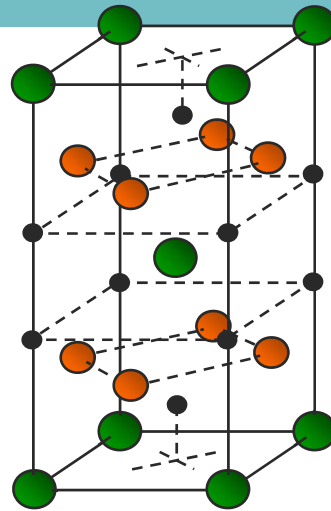
Is the true (experimental) Fermi surface of  $f^2$  compounds close to the LDA Fermi surface as well ? URu2Si2

Pseudo-gap opens at  $T_c$ .  $URu_2Si_2$  measured through optical conductivity, D. A. Bonn et al. PRL (1988).



# Small Effect at T<sub>0</sub>. Resistivity decreases as T decrease

URu<sub>2</sub>Si<sub>2</sub>



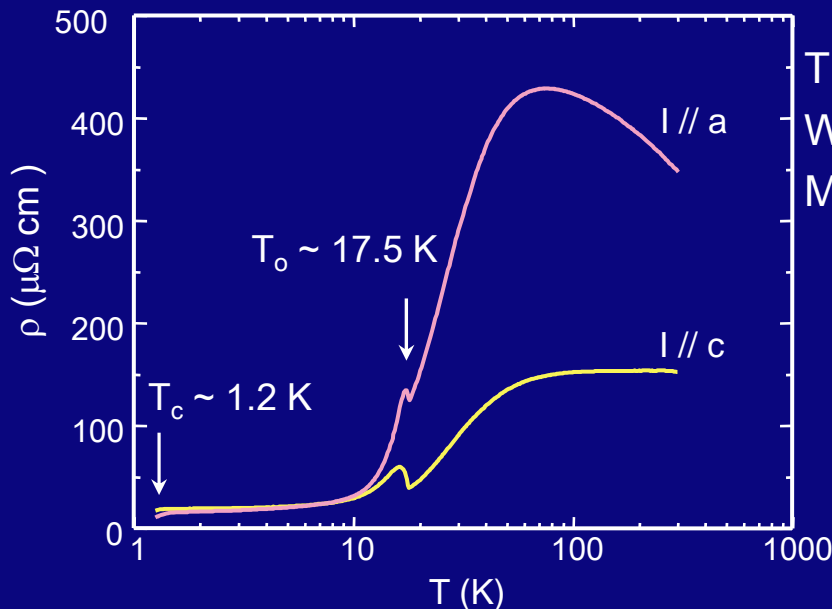
ThCr<sub>2</sub>Si<sub>2</sub> bct - type ( I4/mmm )

a = 4.127 (Å)

c = 9.570 (Å)



Heavy fermion at high T, low T HO + SC

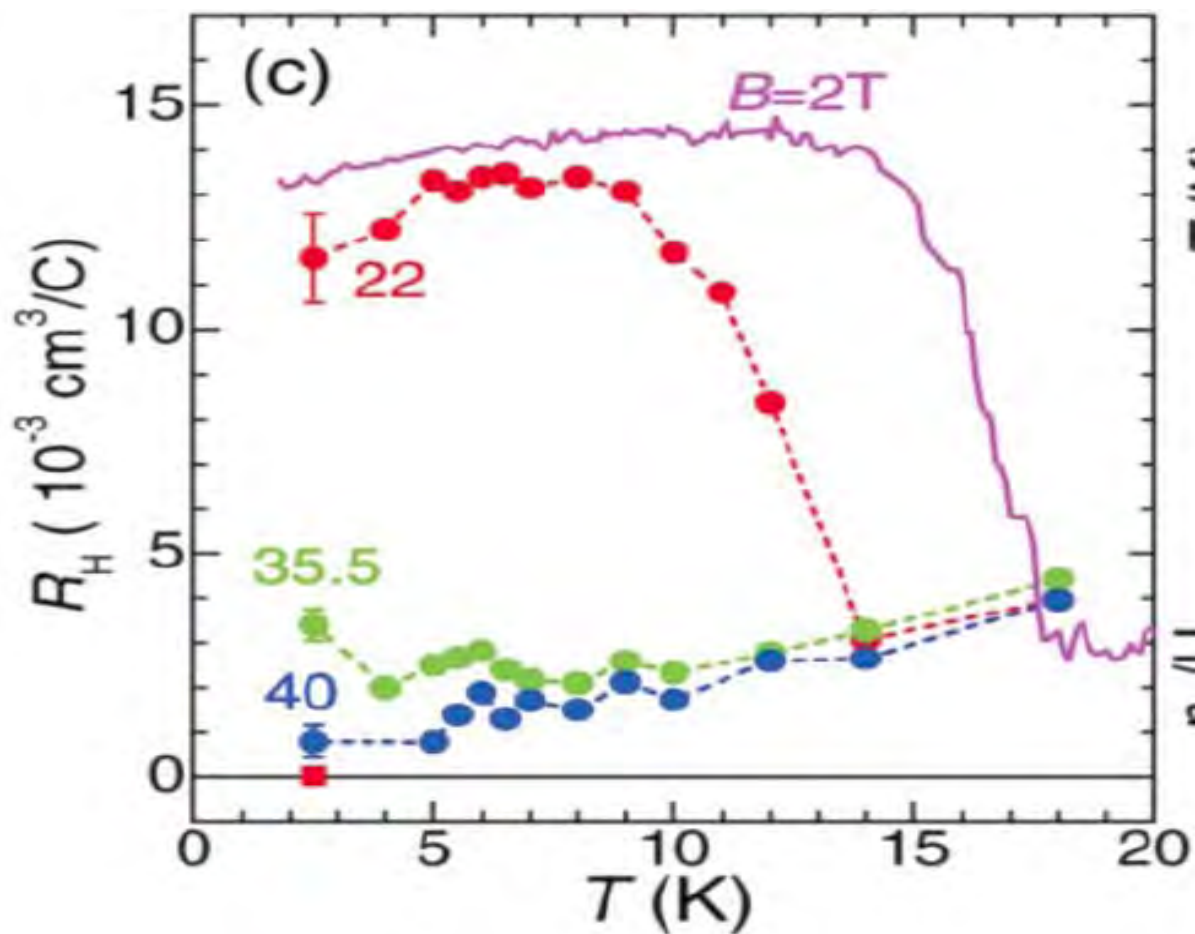


T.T.M. Palstra et al.(1985)

W. Schlabitz et al.(1986)

M.B. Maple et al.(1986)

Hall effect as function of temperature in different external fields, Y. S. Oh et al. PRL 98, 016401(2007). Fermi surface reconstruction in zero and small fields. Very large fields polarized Fermi liquid.

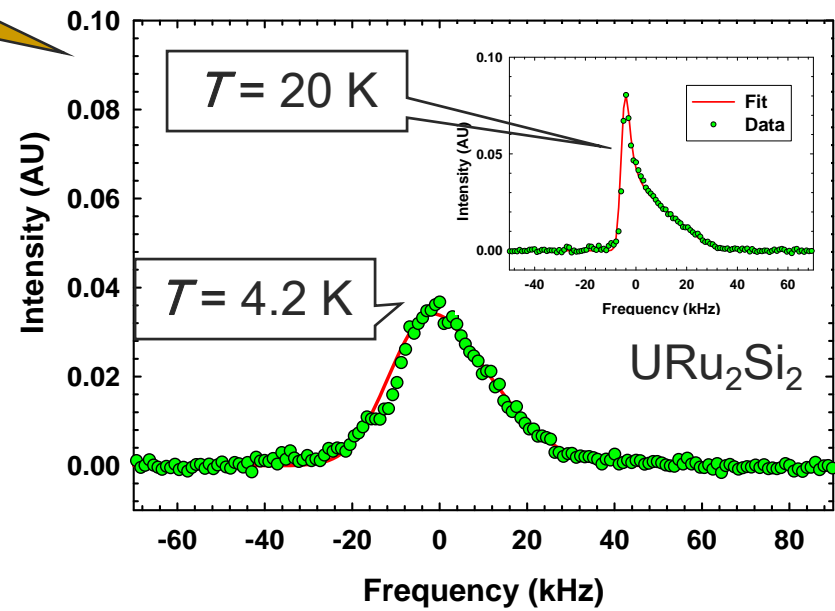




Bernal et. al. PRL,2002.

P. Chandra P. Coleman et. al. (orbital antiferro magnetism) time reversal symmetry breaking.

Si NMR Spectra

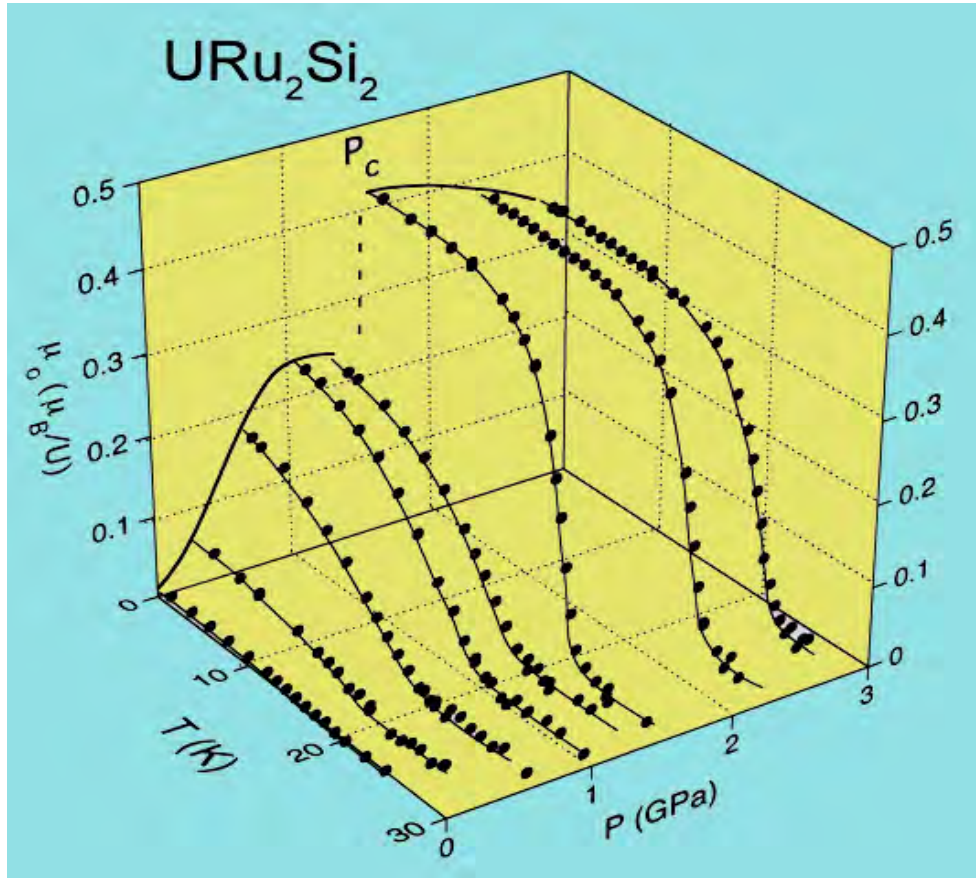


See however the later experiments in unstrained samples that do not show the effect.

## Some Proposals for hidden order in the literature

- Lev. P. Gorkov: 1996:
  - *Three spin correlators.*
- Chandra *et al.*, Nature'02
  - *Incommensurate Orbital Antiferromagnetism*
- Mineev & Zhitomirsky, PRB '05
  - *SDW with tiny moment.*
- Varma & Zhu, PRL'06
  - *Helical Order, Pomeranchuk instability of the Fermi surface ?*
- Elgazaar, & Oppeneer, Nature Materials'08
  - *DFT: with weak antiferromagnetic order parameter*
- *Santini and Amoretti PRL 04*
  - *Quadrupolar ordering.*
- *Fazekas and Kiss PRB 07*
  - *Octupolar ordering.*

# Neutron scattering under hydrostatic pressure

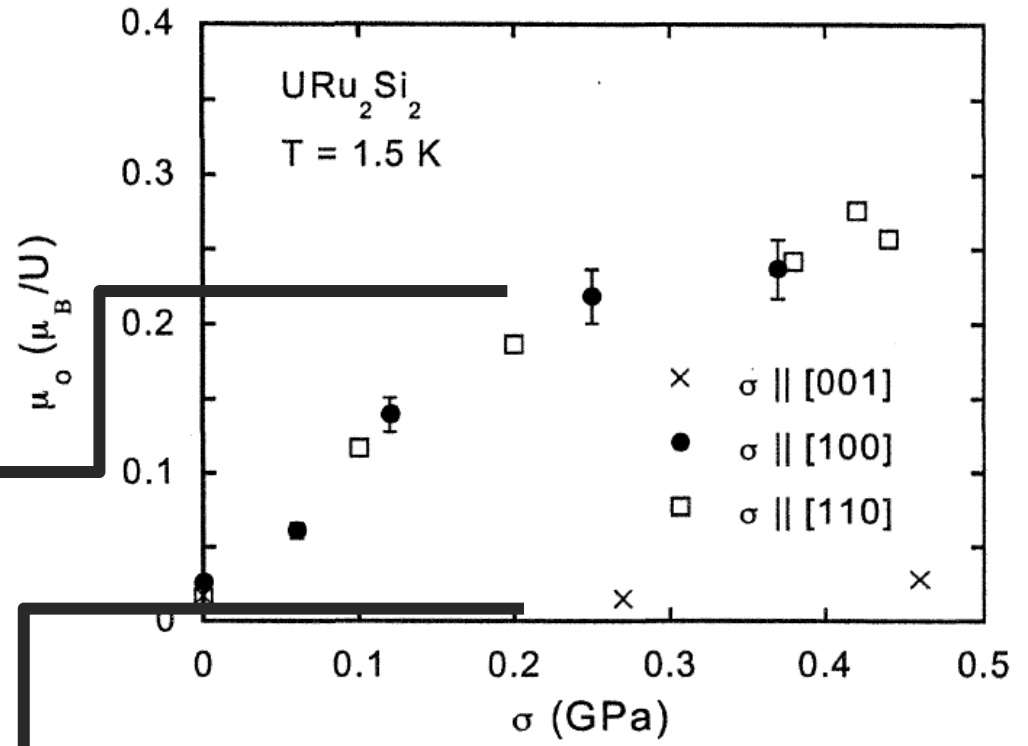
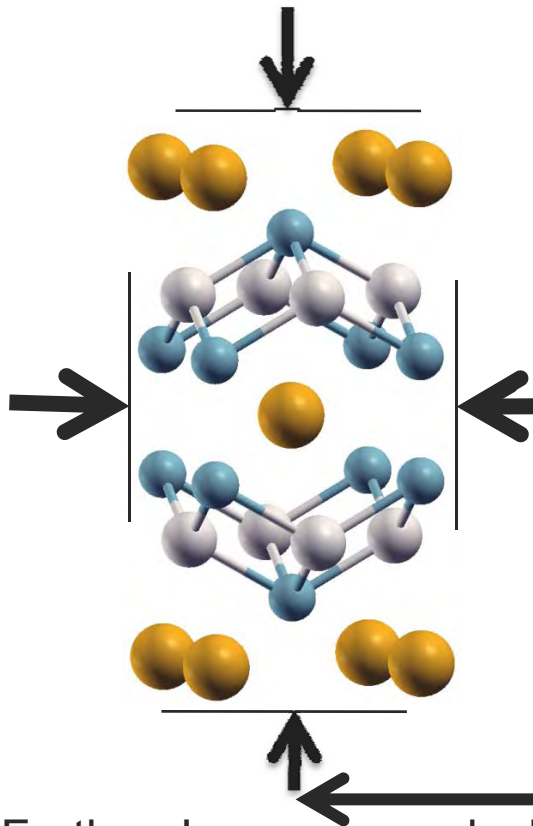


H. Amitsuka, M. Sato, N. Metoki, M. Yokoyama, K. Kuwahara, T. Sakakibara, H. Morimoto, S. Kawarazaki, Y. Miyako, and JAM  
PRL 83 (1999) 5114

# URu<sub>2</sub>Si<sub>2</sub> Stress in ab plane

Large moment when stress in ab plane

No moment when stress in c plane

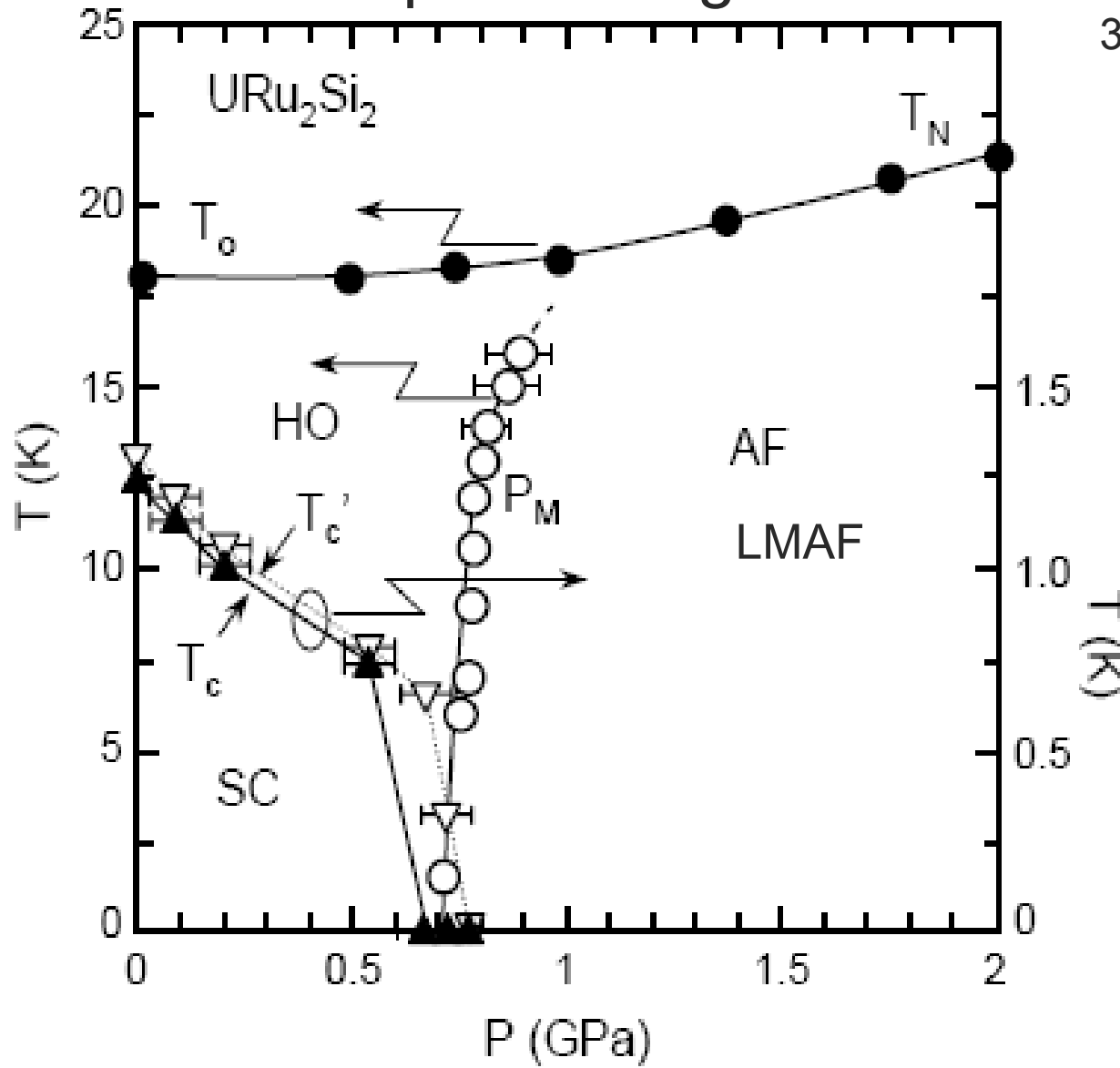


M Yokoyama, JPSJ 71, Supl 264 (2002).

Further Japanese work showed that NMR in unstrained samples did not broaden below T<sub>0</sub>

# P – T phase diagram

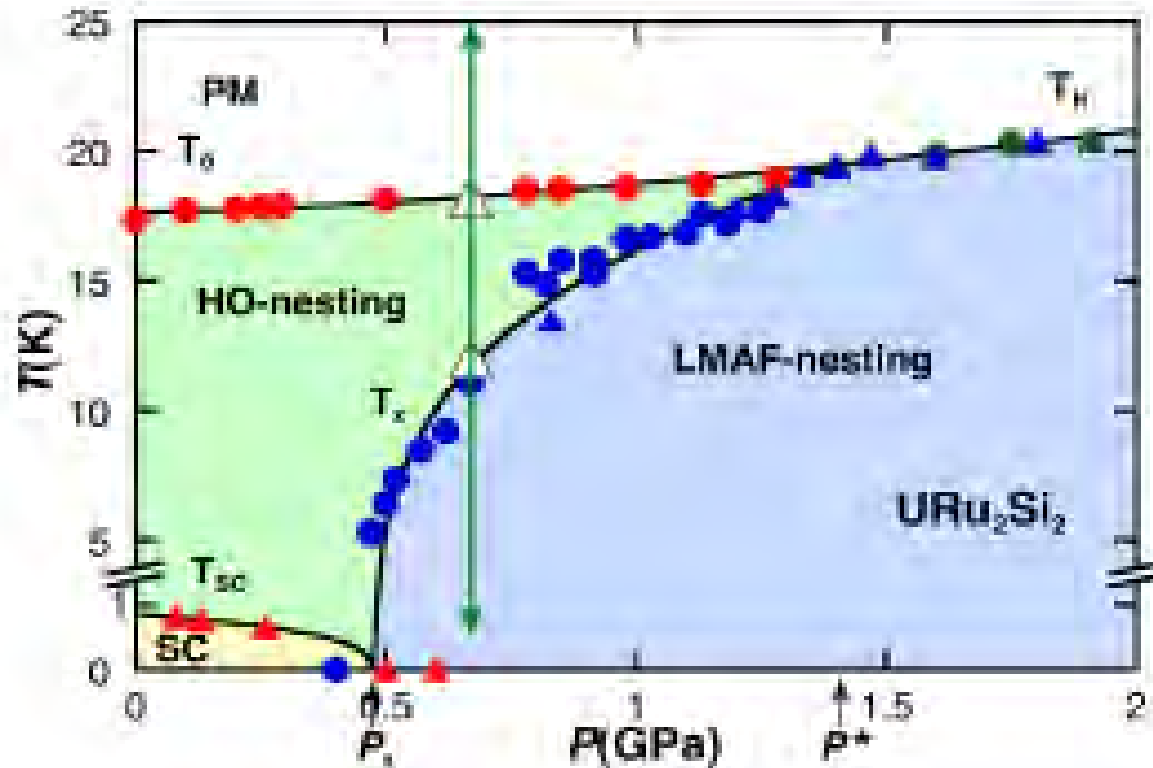
H. Amitsuka et al., JMMM  
310, 214(2007).



Little change in bulk properties with const. P when crossings into HO(T<sub>0</sub>) or LMAF(T<sub>N</sub>) phases, e.g. opening of similar gaps: *Adiabatic Continuity*.

Phase diagram T vs P based upon resistivity and calorimetric experiments under pressure.

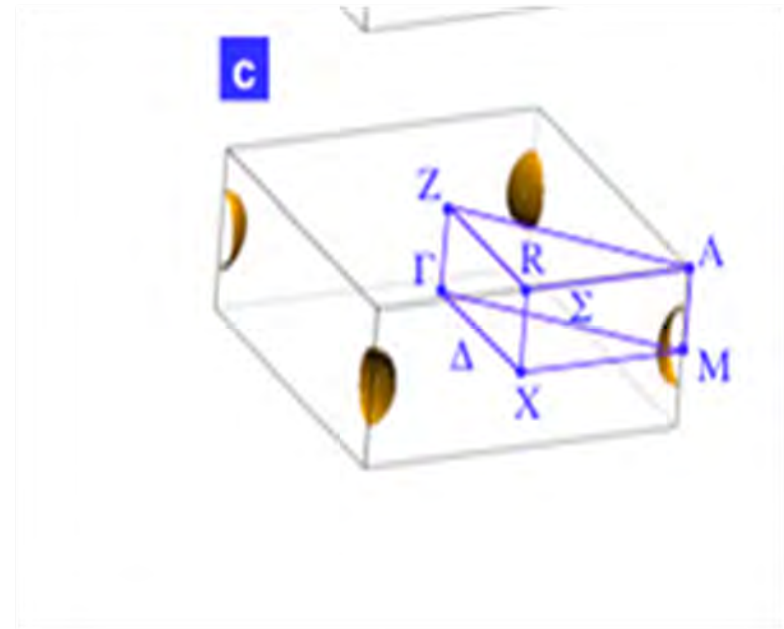
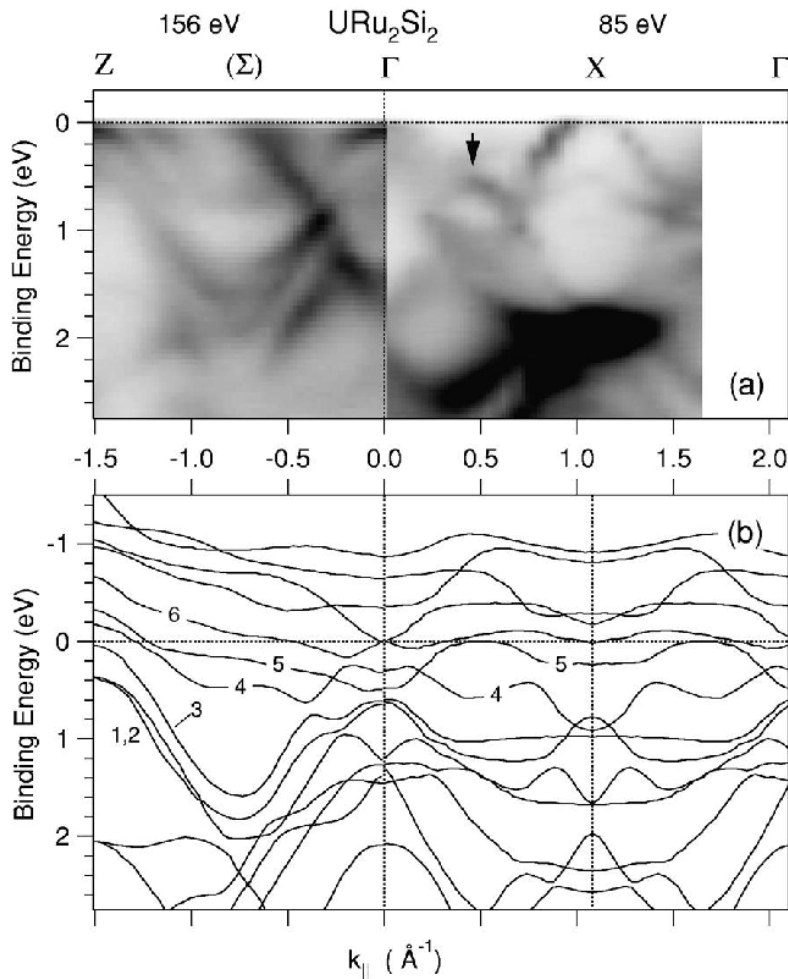
E. Hassinger et al. PRB 77, 115117(2008).



Similar to Amitsuka's T - P phase diagram

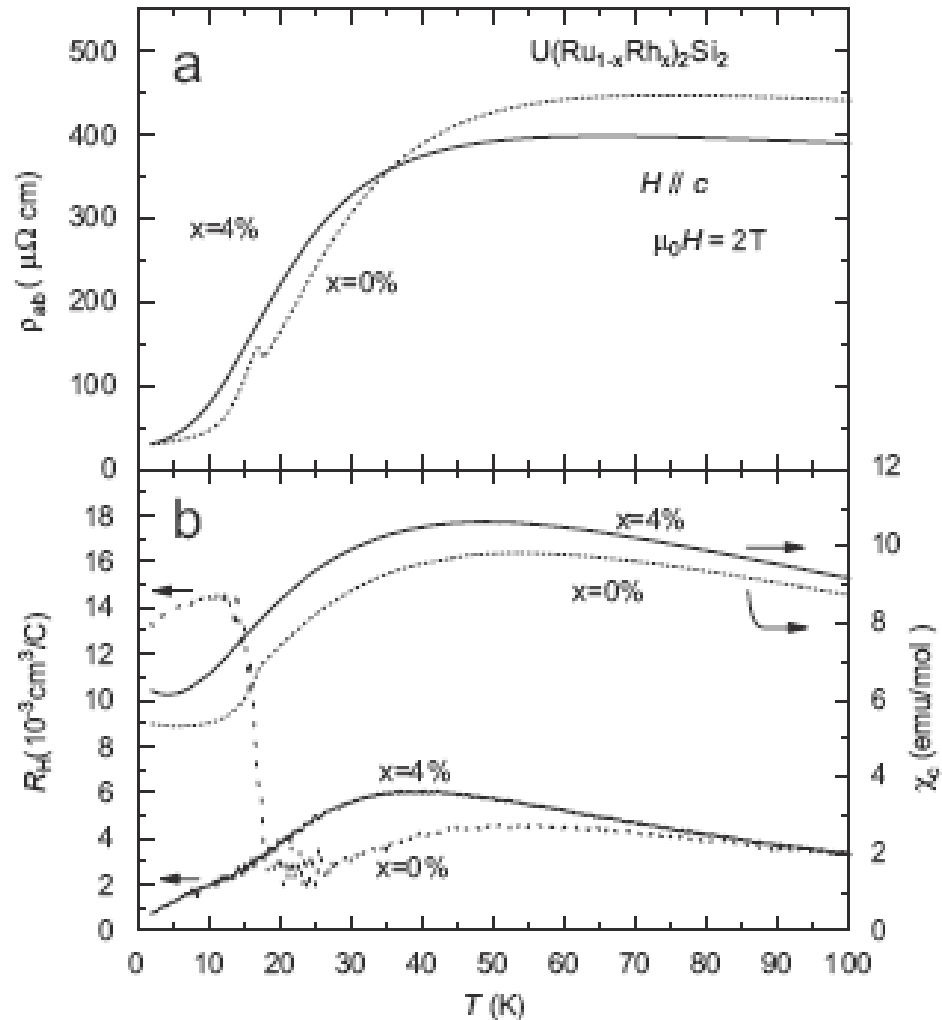


# ARPES does not agree with LDA



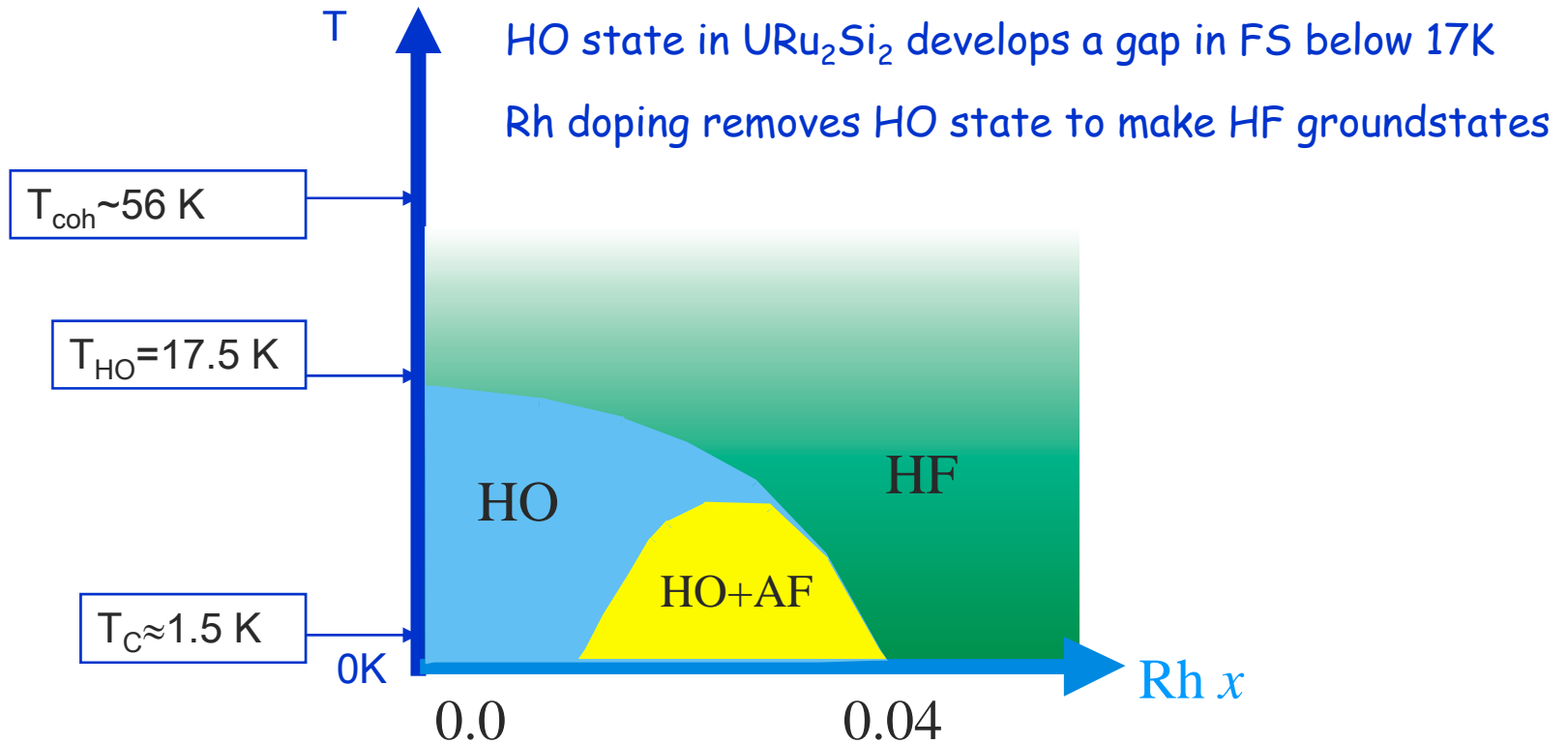
J.D. Denlinger et.al., 2001

# Comparison of low-field bulk properties - pure vs. 4%Rh



Y.S.Oh, K.H.Kim, N. Harrison, H.Amitsuka & JAM, JMMM 310,855(2007).

# Effects of Rh Doping. $U(\text{Ru}_{1-x}\text{Rh}_x)_2\text{Si}_2$



M. Jaime et al. PRL (2002)  
N. Harrison PRL (2003)  
K. H. Kim *et al.*, PRL (2003)  
K. H. Kim *et al.*, PRL (2004)

# Comments concerning Hidden Order

**URu<sub>2</sub>Si<sub>2</sub>, at high temperatures is not too different from a garden variety heavy fermion.**

**ARPES does not agree with LDA at 30 K.**

**HO can be totally destroyed by H and Rh-x**

**HO converts to LMAF by P thru a first order line.**

**HO and LMAF are remarkably similar (“Mydosh’s adiabatic continuity”)**

**HO opens some form of a gap in optics.**

**HO likely involves an electronic topological transition**

**[Hall Effect, also Nernst]**

**HO exhibits two INS modes:(100)@2meV and (1.400)@5meV of longitudinal fluctuations/excitations.**

**HO (but not LAMF) turns into superconductivity at 1.7 K.**

# Heavy Fermions Early Theoretical Work.

Early work : variational wave functions Varma, C. M. and Yafet, Y., Phys. Rev. B13, 295 (1975);

Slave Bosons MFT ,  $1/N$  , A. Auerbach and K. Levin. Phys. Rev. Lett. 57 (1986), p. 877

A. Millis and P.A. Lee. Phys. Rev. B 35, 3394 (1987)

Simple description of high and low T regimes.

Very simple Renormalized band theory at  $T=0$ .

G.Kotliar A. Ruckenstein extensions to finite U

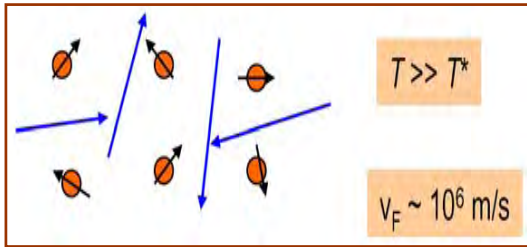
# Early work Generalized Anderson Lattice Model

$$\sum_{i,\sigma} \epsilon_f f_{i\sigma}^\dagger f_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} - \sum_{\langle i,j \rangle, \sigma} (t_{ij} + \mu \delta_{ij})(c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma})$$

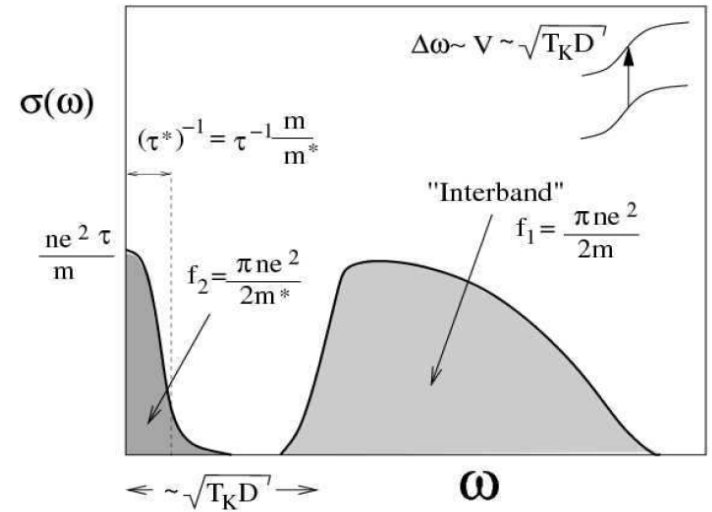
$$+ \sum_{\langle i,j \rangle, \sigma} V_{ij} f_{i\sigma}^\dagger c_{j\sigma} + c.c. = H_{ALM}$$

$$\sum_{i,\sigma} \epsilon_f f_{i\sigma}^\dagger f_{j\sigma} + \sum_i \lambda_i [b_i^\dagger i b_i + \sum_\sigma f_{i\sigma}^\dagger f_{i\sigma} - 1] - \sum_{\langle i,j \rangle, \sigma} (t_{ij} + \mu \delta_{ij})(c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma})$$

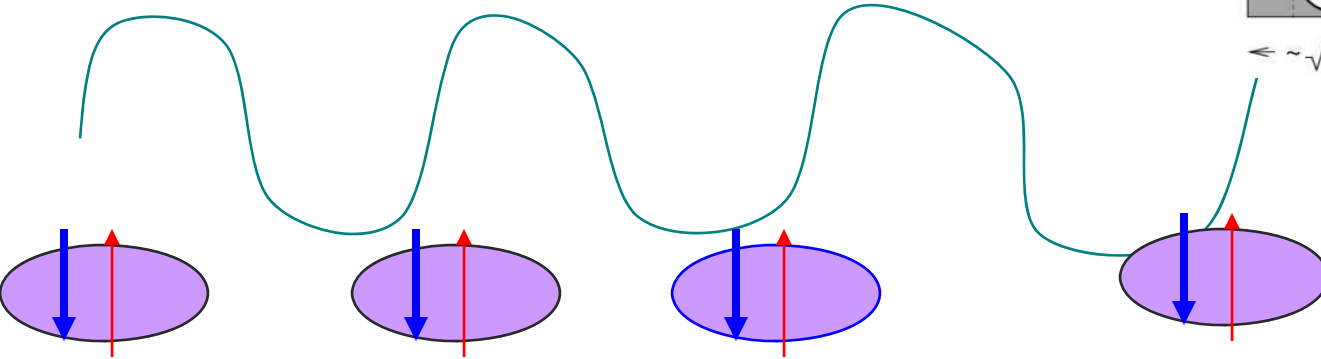
$$+ \sum_{\langle i,j \rangle, \sigma} V_{ij} f_{i\sigma}^\dagger b_i c_{j\sigma} + c.c. = H_{ALM}$$



• High temperature  
Ce-4f local moments



• Low temperature –  
Itinerant heavy bands



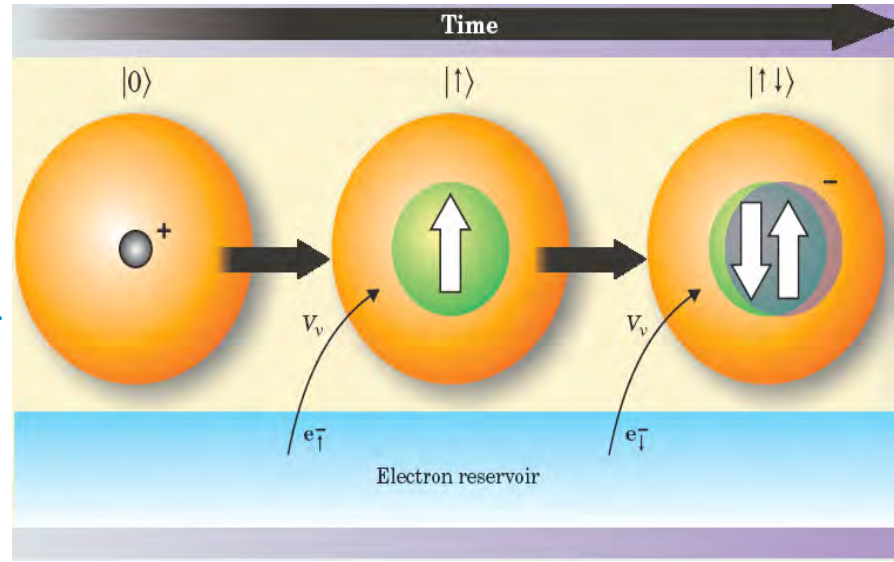
# Dynamical Mean Field Theory. Cavity Construction.

$$\begin{aligned}
 & - \sum_{i,j} J_{ij} S_i S_j - h \sum_i S_i \\
 & - \sum_{\langle i,j \rangle, \sigma} (t_{ij} + \mu \delta_{ij}) (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma} c_{i\sigma}) + \sum_{\alpha, \sigma} (V_\alpha c_{\alpha\sigma}^\dagger + c.c.) + \sum_{\alpha, \sigma} \epsilon_\alpha A_{\alpha\sigma}^\dagger A_{\alpha\sigma} + \sum_{\alpha, \sigma} \mu c_{0\sigma}^\dagger c_{0\sigma} + U c_{0\uparrow}^\dagger c_{0\uparrow} c_{0\downarrow}^\dagger c_{0\downarrow}
 \end{aligned}$$

$$H_{MF} = -h_{eff} S_o$$

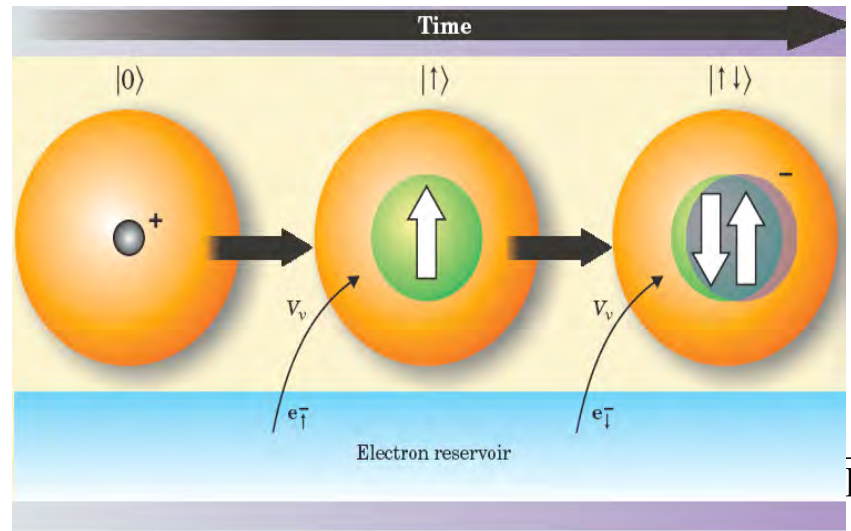
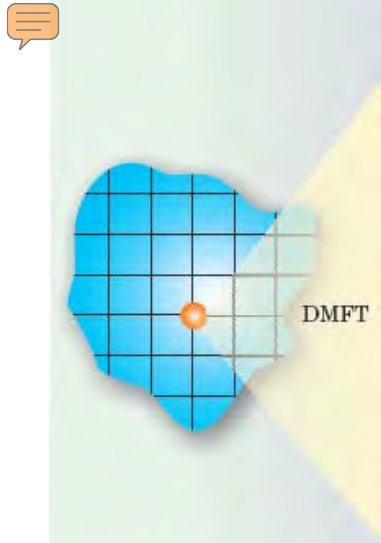


$A(\omega)$



$$\Delta(\omega) = \sum_{\alpha} \frac{V_{\alpha}^* V_{\alpha}}{\omega - \epsilon_{\alpha}}$$

$$\int_0^{\beta} \int_0^{\beta} c_{o\sigma}^\dagger(\tau) \left[ \frac{\partial}{\partial \tau} + \mu - \Delta(\tau - \tau') \right] c_{o\sigma}(\tau') + U \int_0^{\beta} n_{o\uparrow} n_{o\downarrow}$$



$$A(\omega) \quad \Delta(\omega)$$

$$m_i = th[\beta \sum_j J_{ij} m_j + h]$$

$$G_{imp}(i\omega_n) = \sum_k \frac{1}{[-\Delta(i\omega_n) + \frac{1}{G_{imp}(i\omega_n)[\Delta]} - t(k)]}$$

A. Georges, G. Kotliar (1992)

$$G_{imp}(i\omega_n)[\Delta] = \sum_k G_{latt}(i\omega_n, k)[\Delta]$$

# Dynamical Mean Field Theory

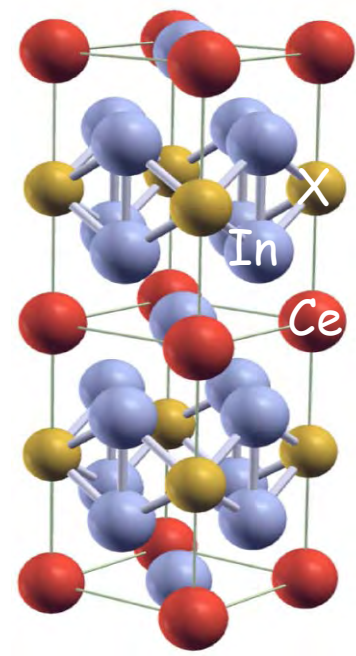
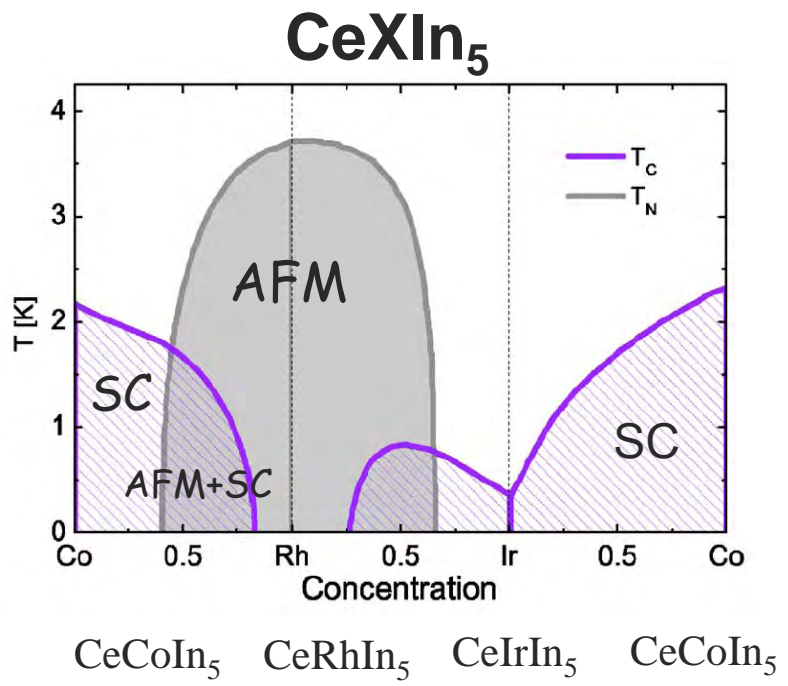
- Exact in the limit of large coordination (Metzner and Vollhardt 89) .
- Can treat arbitrary broken symmetry solutions  $\delta$  is site , spin, orbital, etc. dependent.
- Extension to real materials (Anisimov and Kotliar 1997, Kotliar et. al. RMP 2006).
- DMFT equations are still hard to analyze and solve.

$$|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |\uparrow\downarrow\rangle \longrightarrow |LSJM_J \gamma \dots\rangle$$

$$t(k) \longrightarrow \begin{pmatrix} H[k]_{spd, sps} & H[k]_{spd, f} \\ H[k]_{f, spd} & H[k]_{ff} \end{pmatrix}$$



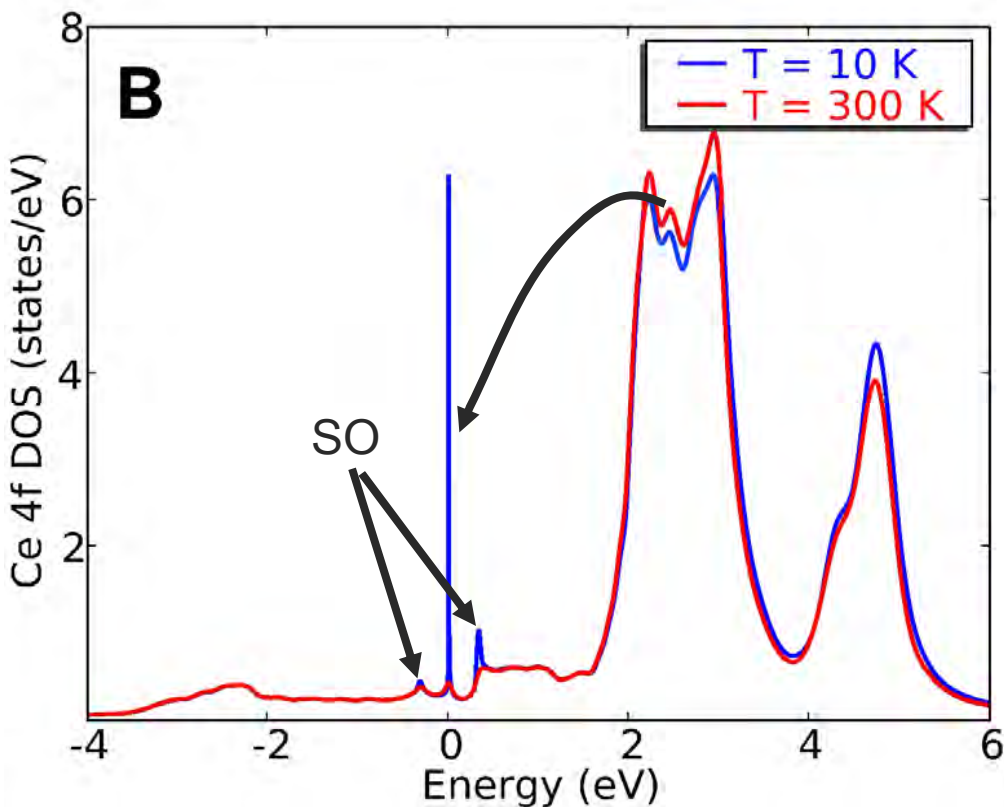
# LDA+DMFT exhibit A Ce (115)



	CeCoIn <sub>5</sub>	CeRhIn <sub>5</sub>	CeIrIn <sub>5</sub>	PuCoG <sub>5</sub>
Tc[K]	SC 2.3K	N 3.8 K	SC 0.4K	18.3K
C <sub>v</sub> /T[mJ/molK <sup>2</sup> ]	300	400	750	100

Control by TM substitution

# Temperature dependence of the local Ce-4f spectra



- At 300K, only Hubbard bands

- At low T, very narrow q.p. peak (width  $\sim 3$ meV)

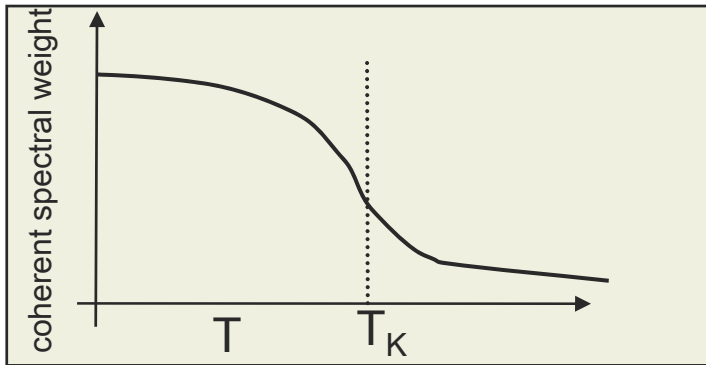
- SO coupling splits q.p.:  $\pm 0.28$ eV

- Redistribution of weight up to very high frequency

*J. H. Shim, KH, and G. Kotliar  
Science 318, 1618 (2007).*

# Buildup of coherence

Buildup of coherence in single impurity case

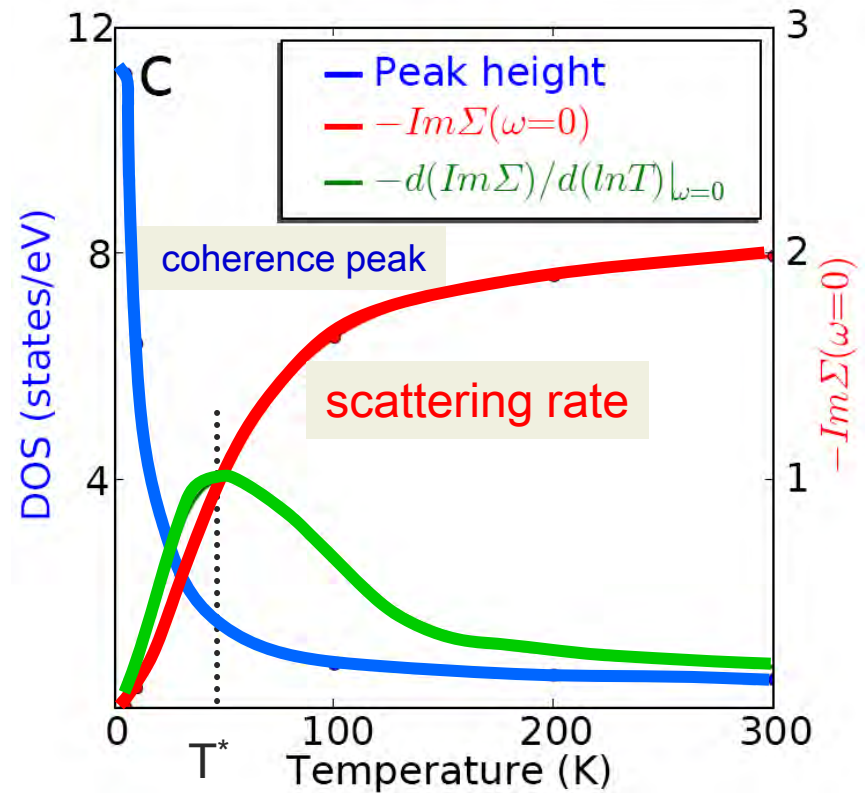


Slow crossover pointed out by NPF 2004

$$\rho_{KL} = \left(1 - \frac{T}{T^*}\right)^{3/2} \left(1 + \ln \frac{T^*}{T}\right) + C$$

Good agreement with Y. Yang & D. Pines  
Phys. Rev. Lett. 100, 096404 (2008).

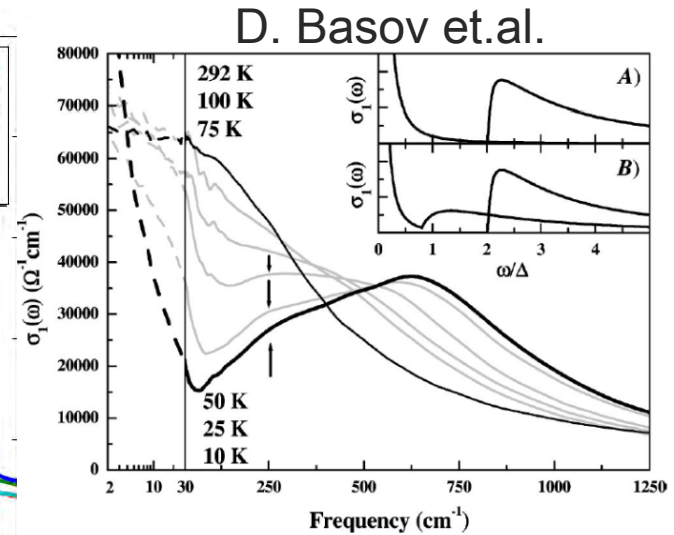
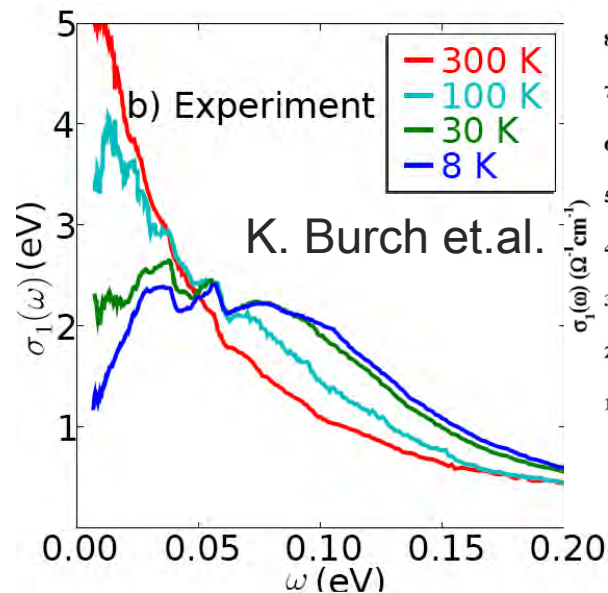
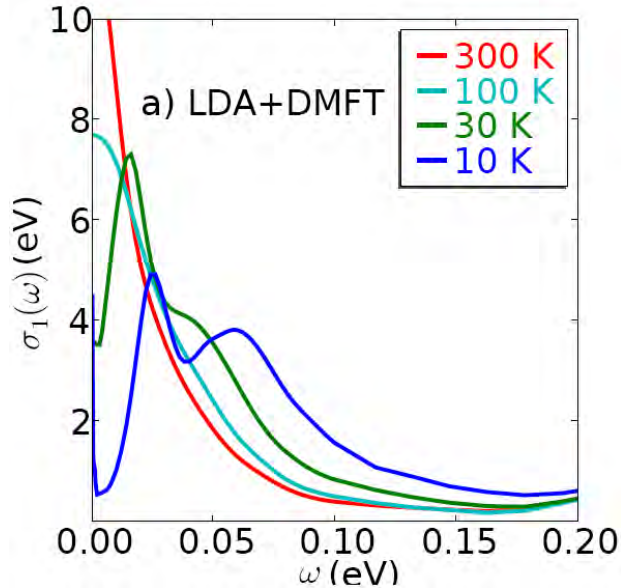
Very slow crossover!



Crossover around 50K

# Optical conductivity in LDA+DMFT

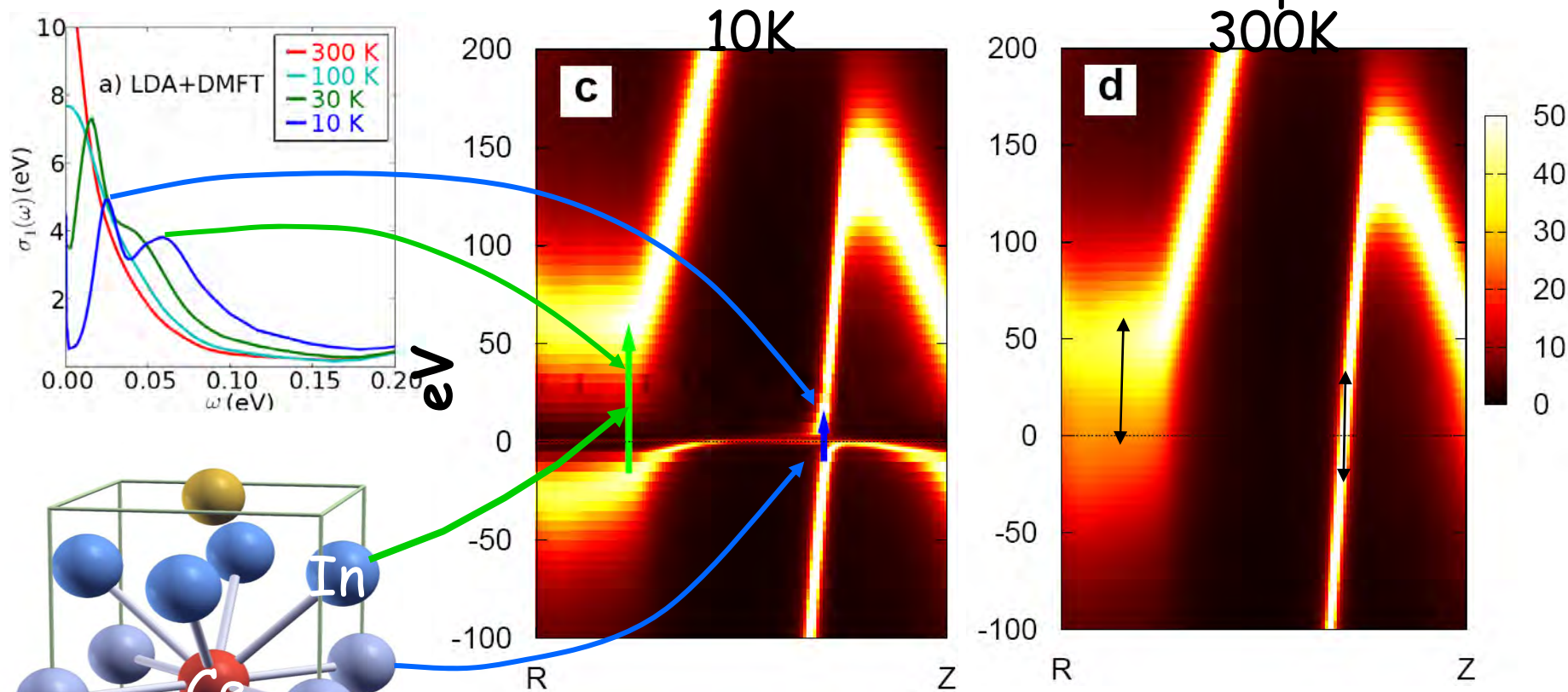
Shim, HK Gotliar Science (2007)



- At 300K very broad Drude peak (e-e scattering, spd lifetime  $\sim 0.1\text{eV}$ )
- At 10K:
  - very narrow Drude peak
  - First MI peak at  $0.03\text{eV} \sim 250\text{cm}^{-1}$
  - Second MI peak at  $0.07\text{eV} \sim 600\text{cm}^{-1}$

# Structure Property Relation: Ce115's Optics and Multiple hybridization gaps

J. Shim et al. Science

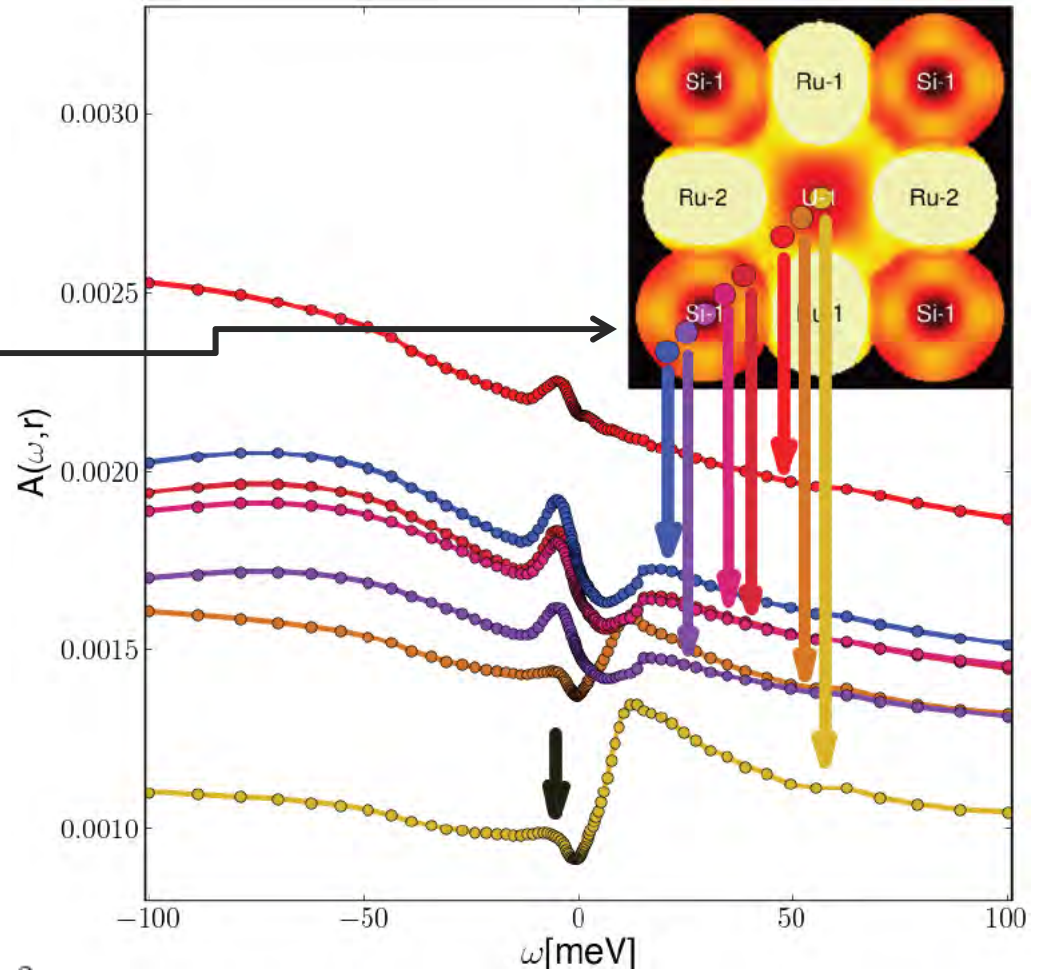
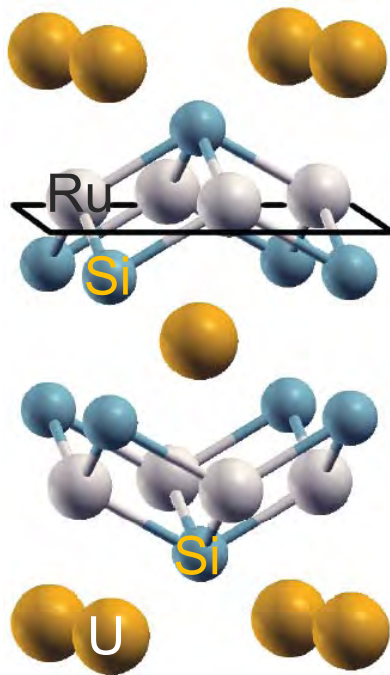


- Larger gap due to hybridization with out of plane In
- Smaller gap due to hybridization with in-plane In

# Conclusions Ce 115's

- Accounts for many of the observed features of Ce based heavy fermions.
- Contact with the phenomenology of NPF.
- Crossover is slower than in single impurity because of the self consistency condition feedback.
- Predictions for ARPES currently being tested.
- Validates renormalized band theory at  $T=0$  , explains why it is not a good guide to experiments at most temperatures.
- Accounts for Co 3d –Rh 4d -Ir 5d

# DMFT "STM" URu<sub>2</sub>Si<sub>2</sub> T=20 K



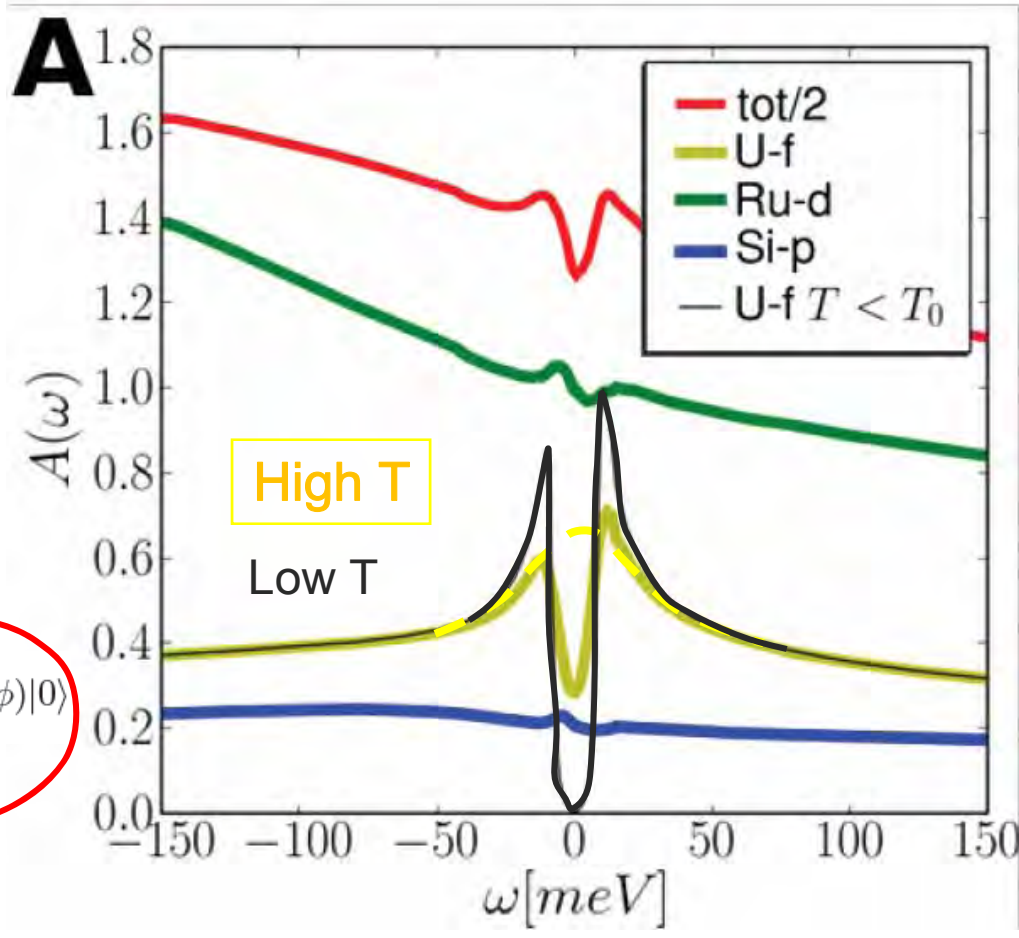
Fano lineshape:

$$A(\omega) \propto [(q^2 - 1) + 2q(\omega/\Gamma)] / [(\omega/\Gamma)^2 + 1]$$

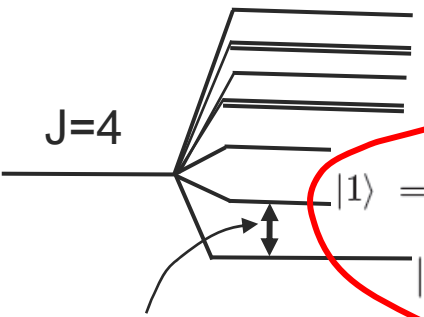
$q \sim 1.24$ ,  $\Gamma \sim 6.8$  meV, very similar to exp Davis

# At T=20 small effects on spd larger gapping of the f's. PES-DMFT

Partial DOS



Ground state atomic multiplet of f<sup>2</sup> configuration in tetragonal field



$$|1\rangle = \frac{\cos(\phi)}{\sqrt{2}}(|4\rangle + |-4\rangle) - \sin(\phi)|0\rangle$$

$$|0\rangle = \frac{i}{\sqrt{2}}(|4\rangle - |-4\rangle)$$

Only 35K!

|state> == |J=4, Jz>

DMFT allows two broken symmetry states at low T. Look for two sublattice structure.

Density matrix for U 5f state  
the J=5/2 subspace

Large moment phase:

$$\Delta = \begin{pmatrix} J=5/2 & -5/2 & -3/2 & -1/2 & 1/2 & 3/2 & 5/2 \\ -5/2 & \Delta_A & 0 & 0 & 0 & \Delta_\varepsilon & 0 \\ -3/2 & 0 & \Delta_B & 0 & 0 & 0 & \Delta_\varepsilon \\ -1/2 & 0 & 0 & \Delta_C & 0 & 0 & 0 \\ 1/2 & 0 & 0 & 0 & \Delta_{C'} & 0 & 0 \\ 3/2 & \Delta_\varepsilon & 0 & 0 & 0 & \Delta_{B'} & 0 \\ 5/2 & 0 & \Delta_\varepsilon & 0 & 0 & 0 & \Delta_{A'} \end{pmatrix}$$

Moment free phase:

$$\Delta = \begin{pmatrix} J=5/2 & -5/2 & -3/2 & -1/2 & 1/2 & 3/2 & 5/2 \\ -5/2 & \Delta_A & 0 & 0 & 0 & \Delta_\varepsilon + \Delta_\alpha & 0 \\ -3/2 & 0 & \Delta_B & 0 & 0 & 0 & \Delta_\varepsilon + \Delta_\beta \\ -1/2 & 0 & 0 & \Delta_C & 0 & 0 & 0 \\ 1/2 & 0 & 0 & 0 & \Delta_C & 0 & 0 \\ 3/2 & \Delta_\varepsilon - \Delta_\alpha & 0 & 0 & 0 & \Delta_B & 0 \\ 5/2 & 0 & \Delta_\varepsilon - \Delta_\beta & 0 & 0 & 0 & \Delta_A \end{pmatrix}$$

tetragonal symmetry broken->  
these terms nonzero

# Valence histogram point of view.

The DMFT density matrix has mostly weight in two singlet  $f^2$  configurations  
Definitely  $f^2$ , “Kondo “ limit,  $J=4$ , two low lying singlets  
Test via photoemission [Denlinger and Allen ]

Therefore there are two singlets relevant at low energies but they are not non  
Kramer doublets. Conspiracy between cubic crystal field splittings and tetragon  
al splittings bring these two states close.  
This is why URu<sub>2</sub>Si<sub>2</sub> is sort of unique.

$$|0\rangle = \frac{i}{\sqrt{2}}(|4\rangle - |-4\rangle)$$

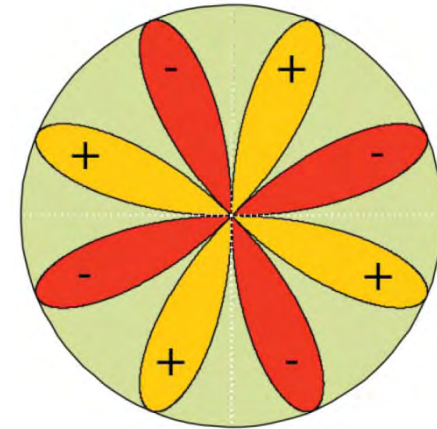
$$|1\rangle = \frac{\cos(\phi)}{\sqrt{2}}(|4\rangle + |-4\rangle) - \sin(\phi)|0\rangle$$

# DMFT excitonic order parameter

$$|0\rangle = \frac{i}{\sqrt{2}}(|4\rangle - |-4\rangle)$$

$$|1\rangle = \frac{\cos(\phi)}{\sqrt{2}}(|4\rangle + |-4\rangle) - \sin(\phi)|0\rangle$$

Hexadecapole



Order parameter:

$$\psi_i = \langle X_{01}(\mathbf{R}_i) \rangle \begin{cases} \text{Im}\psi \propto \langle J_z \rangle \\ \text{Re}\psi \propto \langle (J_x J_y + J_y J_x)(J_x^2 - J_y^2) \rangle \end{cases}$$

Different orientation gives different phases: “adiabatic continuity” explained.

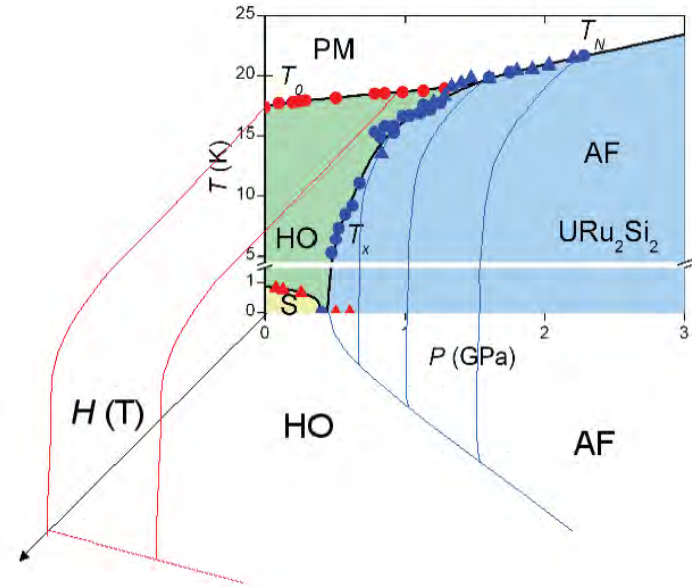
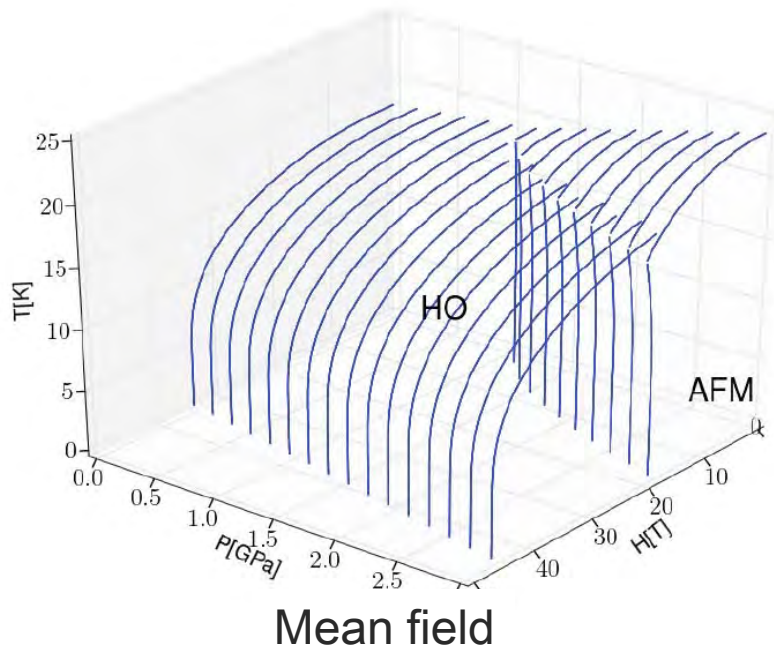
Hexadecapole order testable by resonant X-ray

In the atomic limit:

$$|gs\rangle = \cos(\theta)|0\rangle + \sin(\theta)e^{i\varphi}|1\rangle$$

$$\langle gs|\mathbf{J}|gs\rangle = 4 \cos(\phi) \sin(2\theta) \sin(\varphi) * (0, 0, 1)$$

# Simplified toy model phase diagram mean field theory

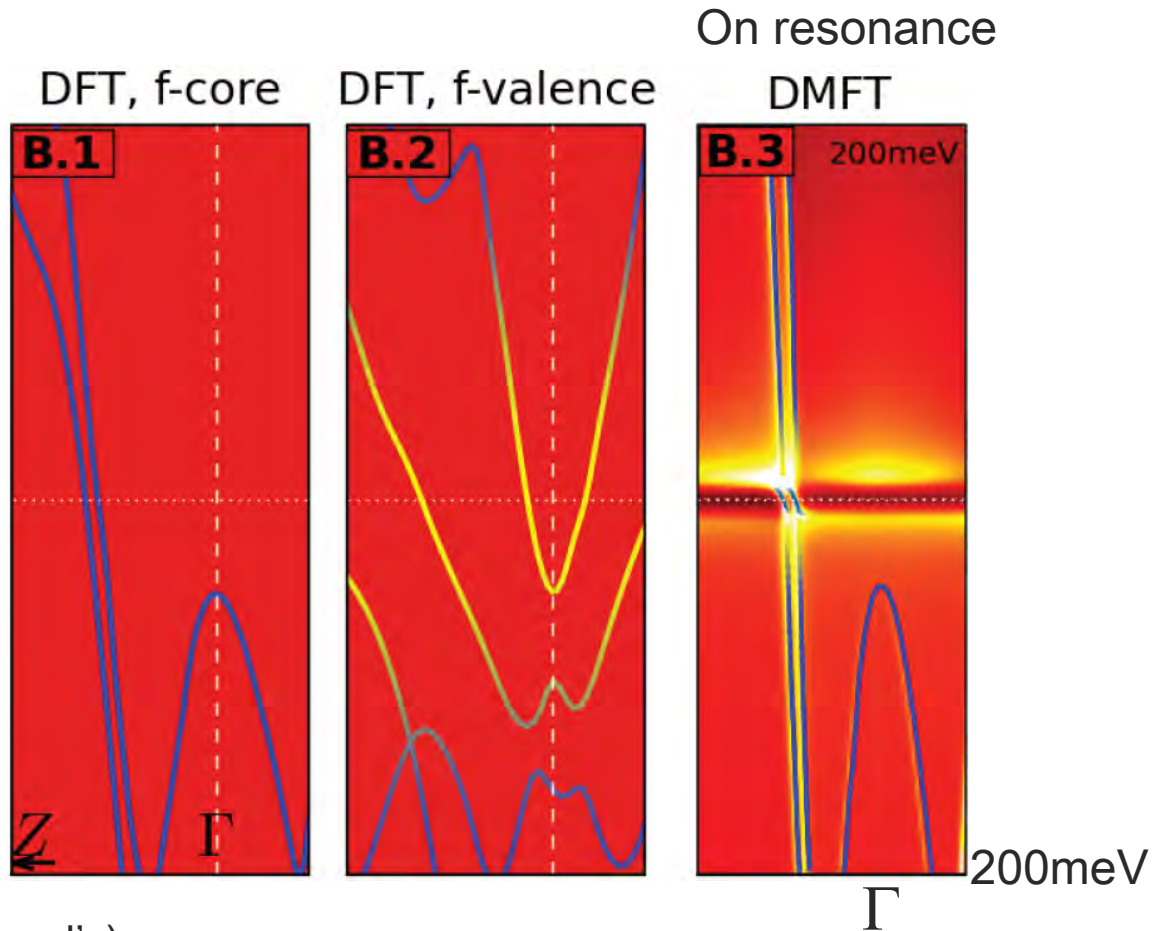


Exp. by E. Hassinger et.al. PRL 77, 115117 (2008)

# Arrested Kondo effect

- DFT f-core: goof description of bands 30meV away from EF
- DFT f-valence: many f-bands at EF, substantial disagreement with ARPES & DMFT

DMFT:  
 very narrow region of f-spectral weight  $\pm 10\text{meV}$  around EF appears below  $T^* \sim 70\text{K}$   
 Below 35K, partial gap starts to open -> singlet to singlet Kondo effect

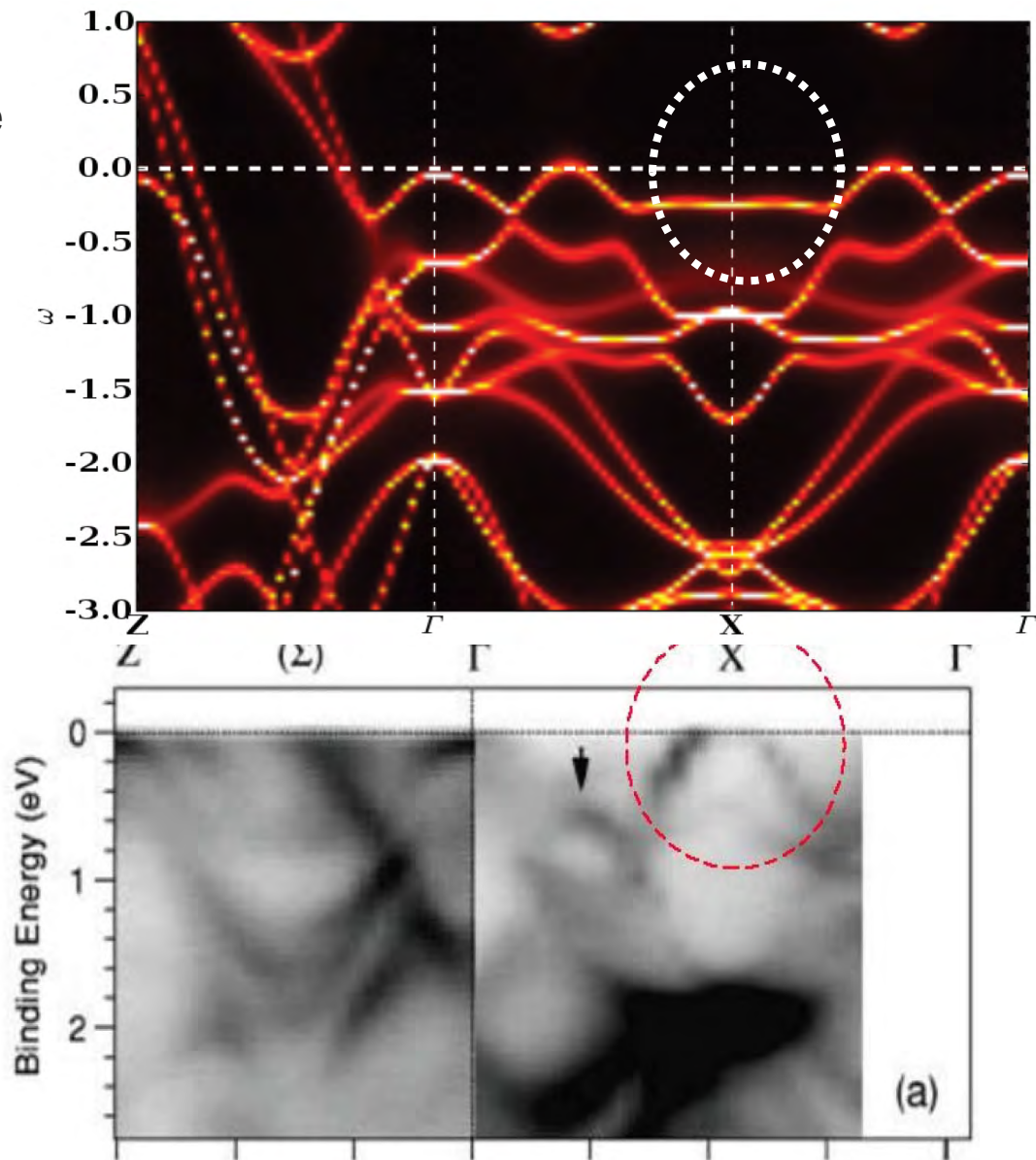


At low temperature, full gap in f's (not spd's).

# DMFT $A(k, \omega)$ vs ARPES

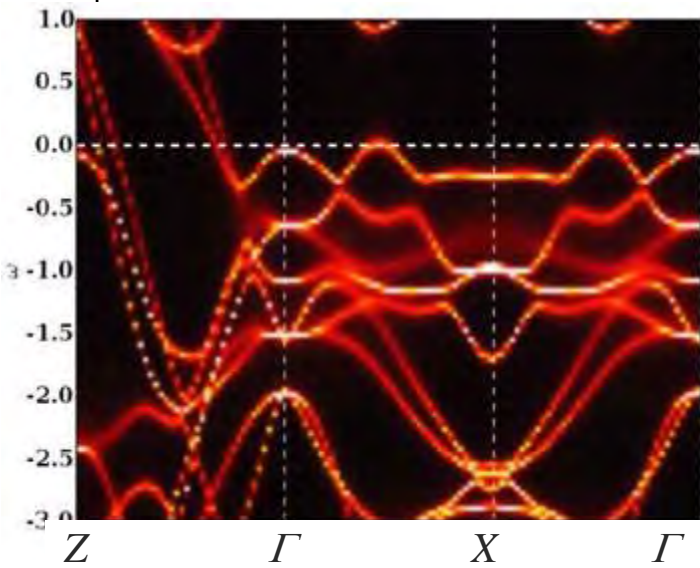
Off resonance

Very good agreement,  
except at X point



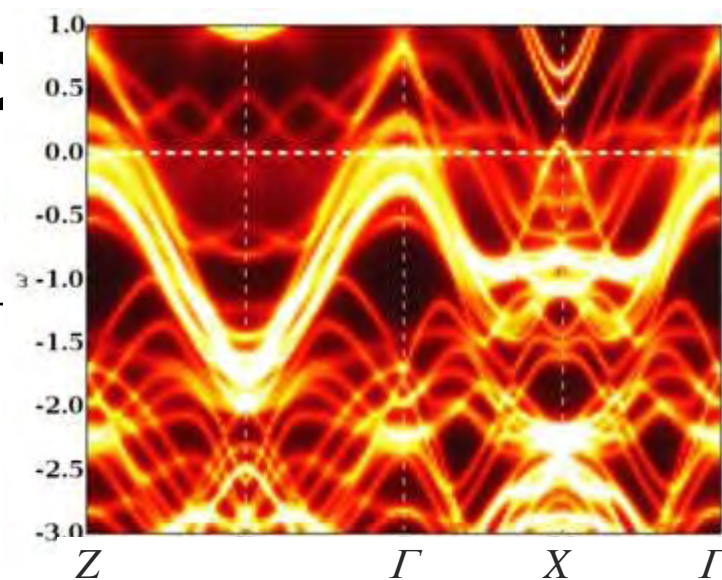
# Surface origin of pocket at X point

*LDA+DMFT - bulk*

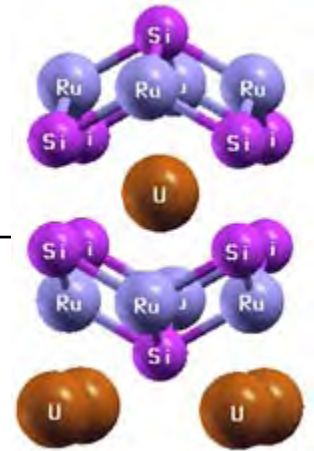


- *No hole-pocket at the X-point.*

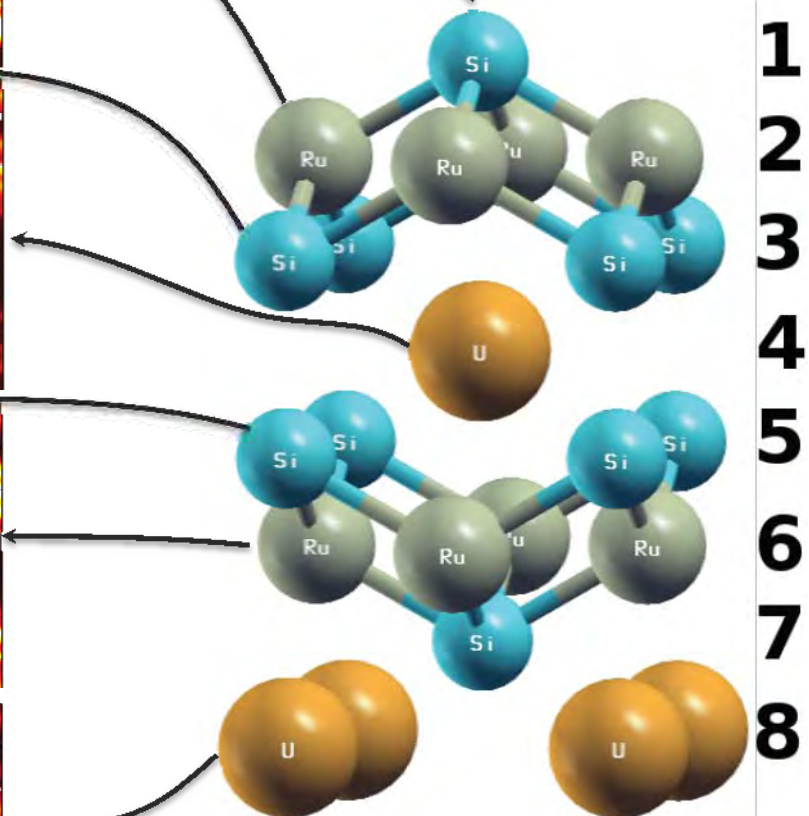
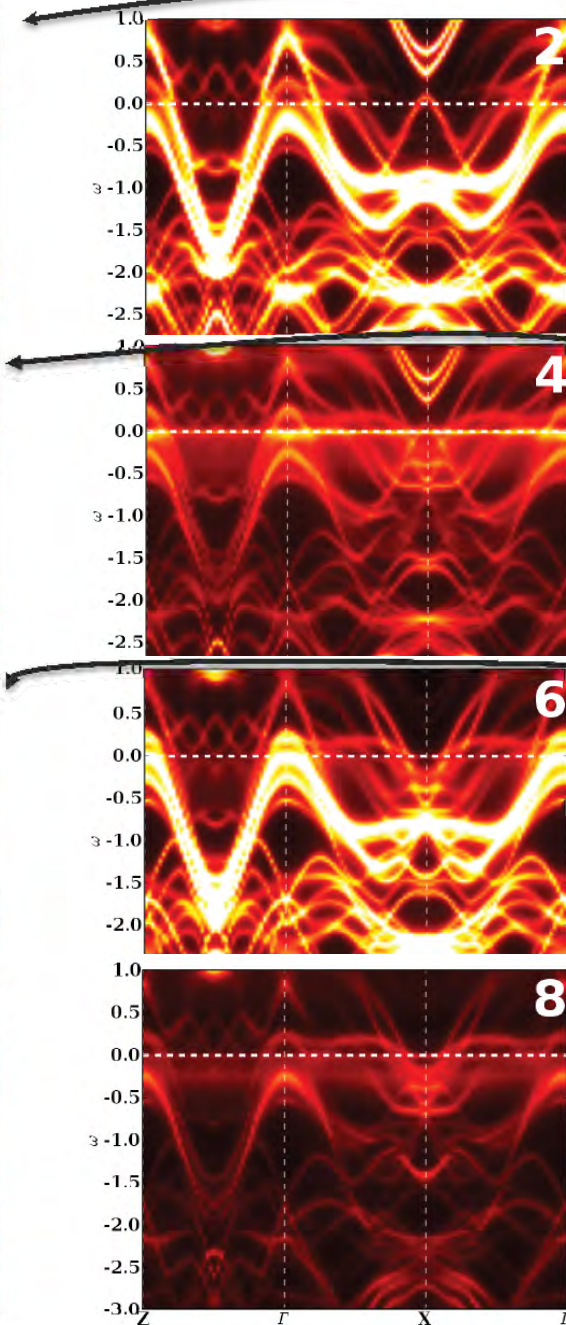
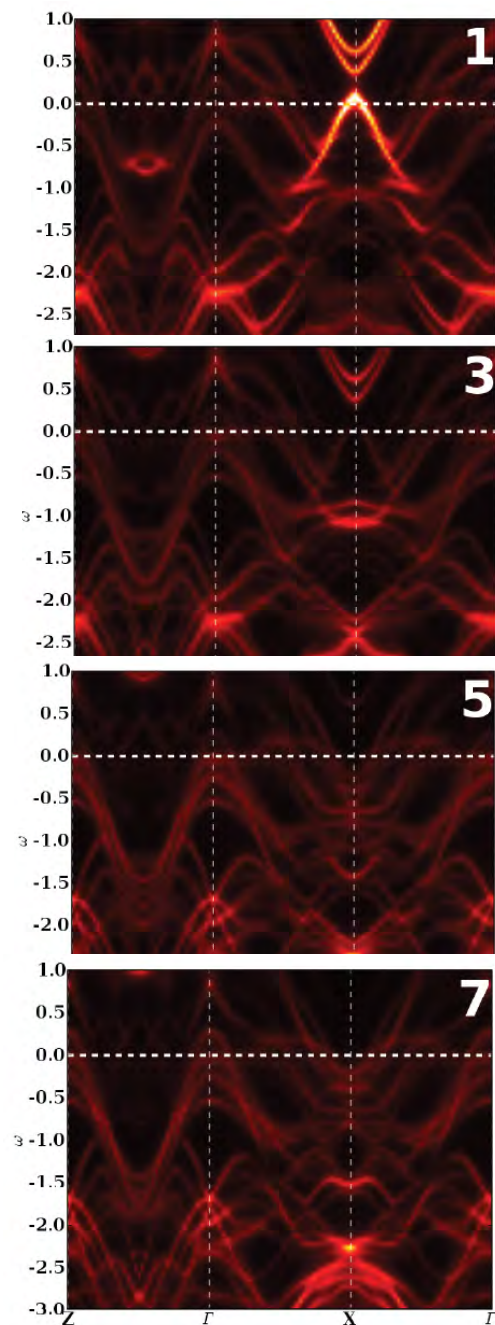
*LDA+DMFT - Si-terminated surface slab*



- *Hole pocket surface state appears at X-point!*



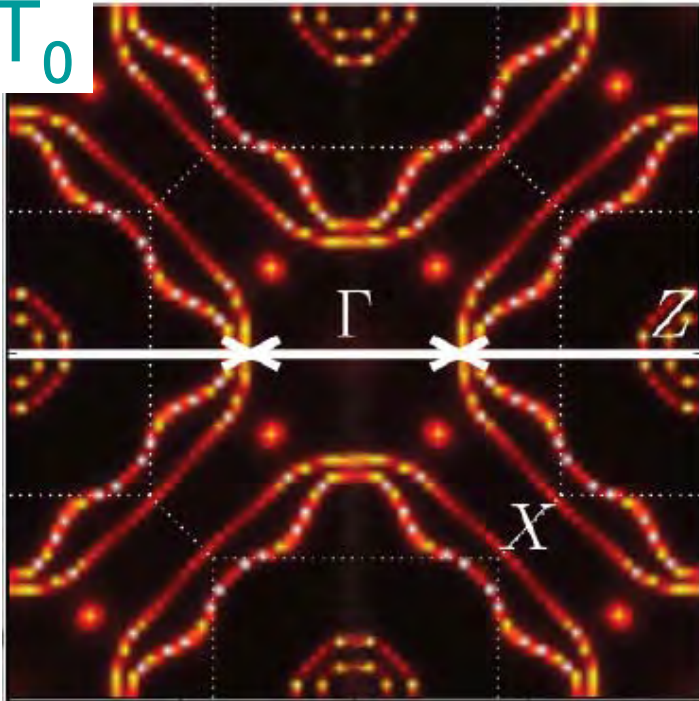
# Layer resolved spectra



1  
2  
3  
4  
5  
6  
7  
8

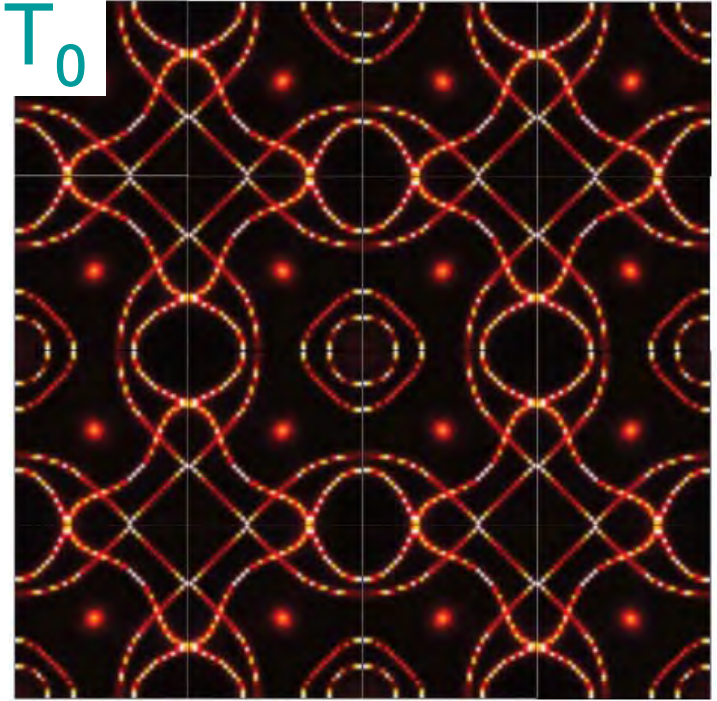
# Fermi surface nesting, reconstruction below $T_c$

$T > T_0$



Nesting  $0.6a^*$  and  $1.4a^*$

$T < T_0$



# Conclusions URu<sub>2</sub>Si<sub>2</sub>

- **5f<sup>2</sup> configuration in the Kondo limit**
- **Hidden order has hexadecapole character**
- **Simple connection between the LMAF and the HO state.**
- **Fermi surface reconstruction at low temperature**
- **Absorption of f degrees of freedom at very low energies is arrested by a (small) crystal field splitting**
- **D. Cox original guess for UBe<sub>13</sub> was (almost) right for URu<sub>2</sub>Si<sub>2</sub>.**
- **DMFT results should be confronted carefully with experiments.**

# Conclusion: some general comments.

- **DMFT approach. Can now start from the material.**
- **Can start from high energies, high temperatures, where the method (I believe ) is essentially exact, far from critical points, provided that one starts from the right “reference frame”.**
- **Still need better tools to analyze and solve the DMFT equations.**
- **Still need simpler approaches to rationalize simpler limits.**
- **Validates some aspects of slave boson mean field theories, modifies quantitatively and sometimes qualitatively the answers.**

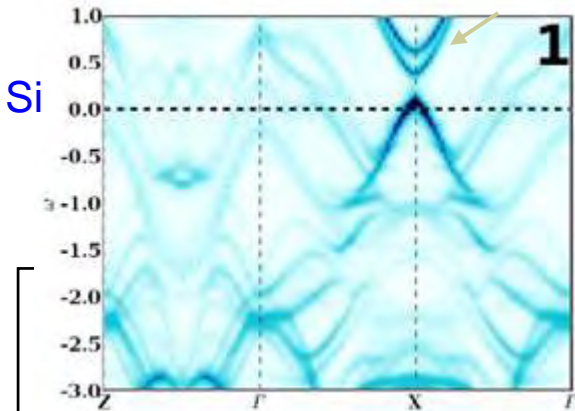
- **At lower temperatures, one has to study different broken symmetry states.**
- **At lower temperatures, one has to study different broken symmetry states.**
  
- **Compare free energies, draw phase diagram**
  
- **Beyond DMFT: Write effective low energy theories that**
- **match the different regions of the phase diagram.**
- **Close contact with experiments.**
- **Many materials are being tried, methods are being refined**
- **Contemplating material design using correlated electron systems.**

**תודה רבה**



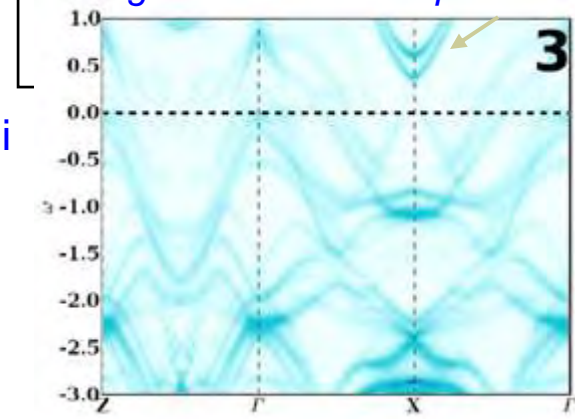
# Conclusions

- DMFT tools can be used to understand/predict properties of correlated materials
- Kondo effect in  $\text{URu}_2\text{Si}_2$  is arrested below crystal field splitting energy. Gives room to ordered states, either AFM state or orbital order.
- AFM state and hidden order state have the same order parameter: mixing between atomic singlet states.
- Orientation of the order parameter decided which state is stabilized.
- Mystery of  $\text{URu}_2\text{Si}_2$  hidden order solved.



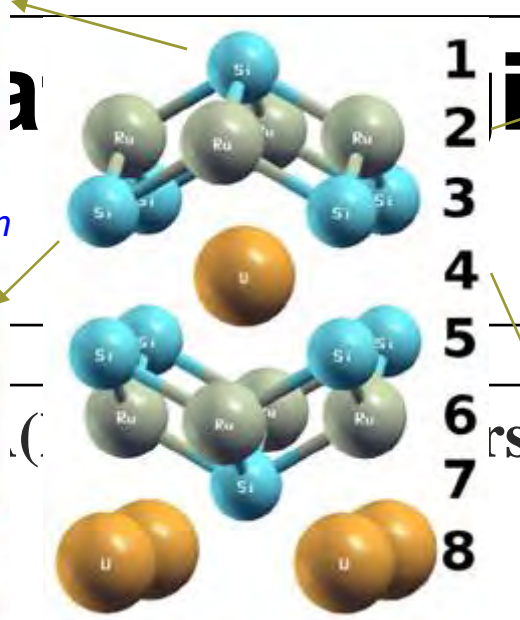
Si

• SS hole band distinctly originates from the top Si atom

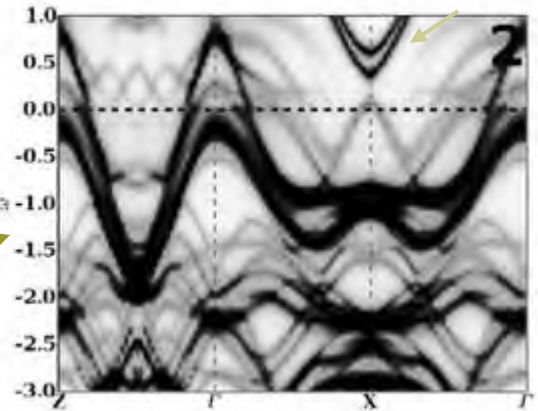


Si

• Very little 3rd layer Si contribution to the SS hole band

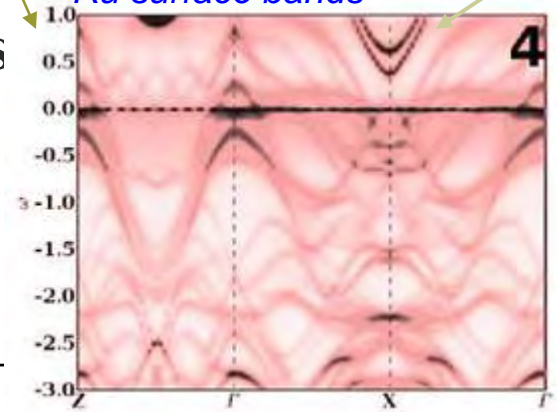


• Strong 2nd layer Ru contribution to  $G=Z$  equivalence



Ru

• Some U5f weight pulled into Ru surface bands



U