Understanding Correlated Electron Materials: A Dynamical Mean Field Perspective

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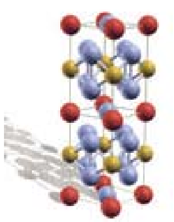
Work in collaboration with Gabi Kotliar



Support:

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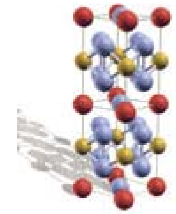
ACS Chemistry for Life™

KITP, Santa Barbara, 2010

Motivation



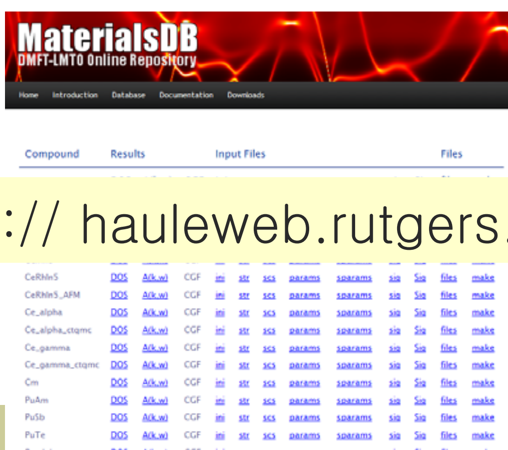
Motivation: Testing our ability to understand the physical properties of strongly correlated materials.

The driving force: Experimental probes Optics,APRES,STM, …

interplay

Computational tools (LDA,DMFT)

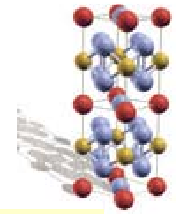
Developing DMFT into an electronic structure tool, understanding qualitatively universal and system specific aspects.



http:// hauleweb.rutgers.edu

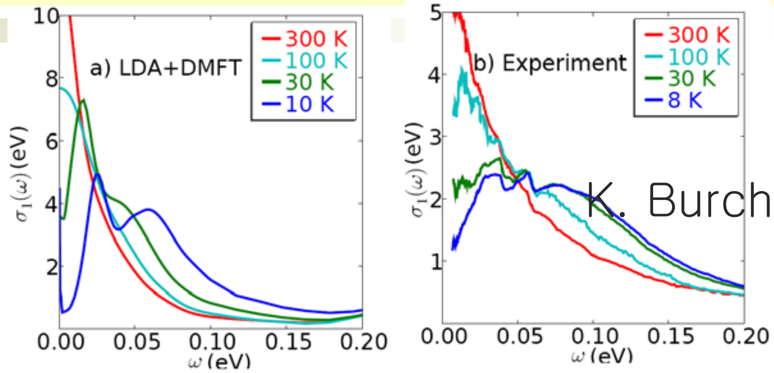
Wien2K+DMFT

multiorbital CTQMC, full potential basis, charge self-consistent



Protracted screening and multiple hybridization Gaps in Ce115's

J.H.Shim, KH, G.Kotliar, Science 318, 1615 (2007)

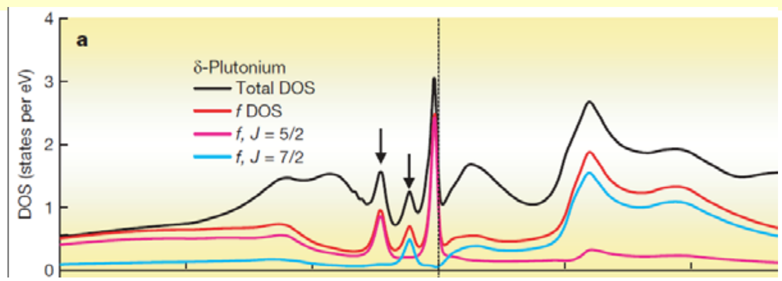


K. Burch et.al.

Tools allow to identify system specific fingerprints which gives us confidence in our understanding of correlated electron materials

Quasiparticle multiplets in Plutonium

J.H.Shim, KH, G.Kotliar, Nature 446, 513 (2007).



Hidden Order in URu2Si2, Kondo effect

KH, G. Kotliar, Nature Physics 5, 796 - 799 (2009).

URu_2Si_2 - heavy fermion with hidden

URu_2Si_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).

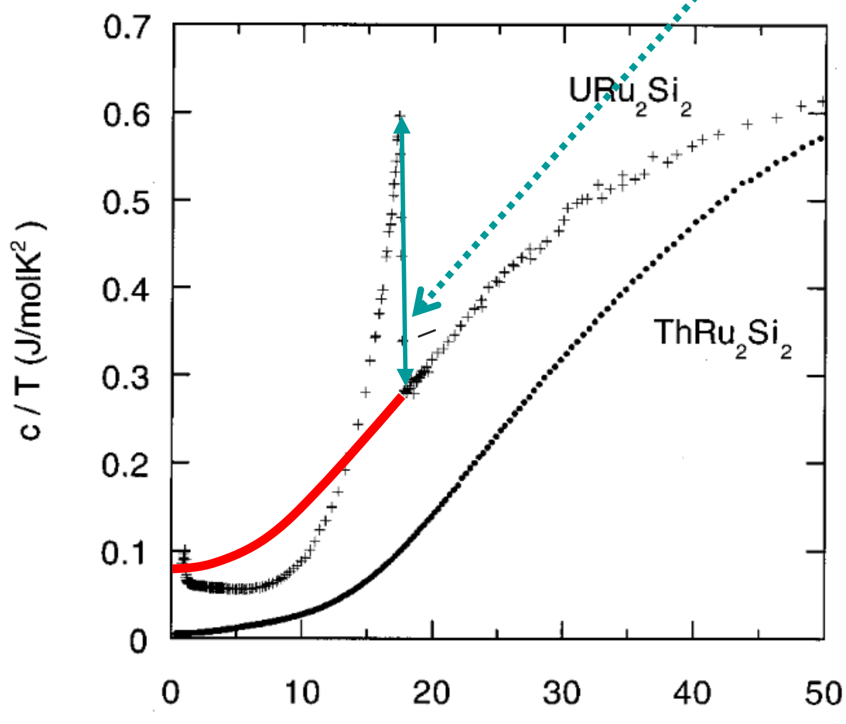
Curie−Weiss: $\mu_z^{eff} \sim 2.2\ \mu_B$

Second order phase transition

Elec. cv: $\gamma \sim 70$ mJ/mol K^2

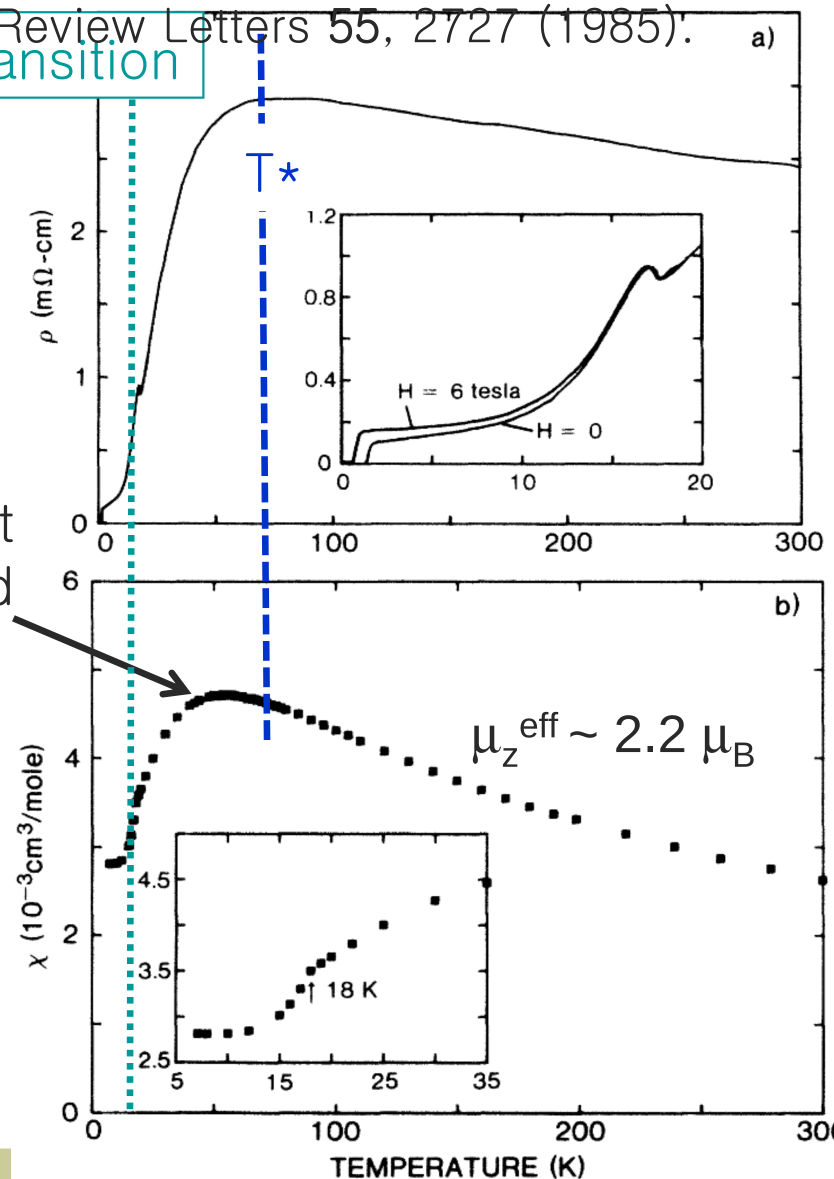
Coherence: T*~70K

SOPT: T_0~17.8K

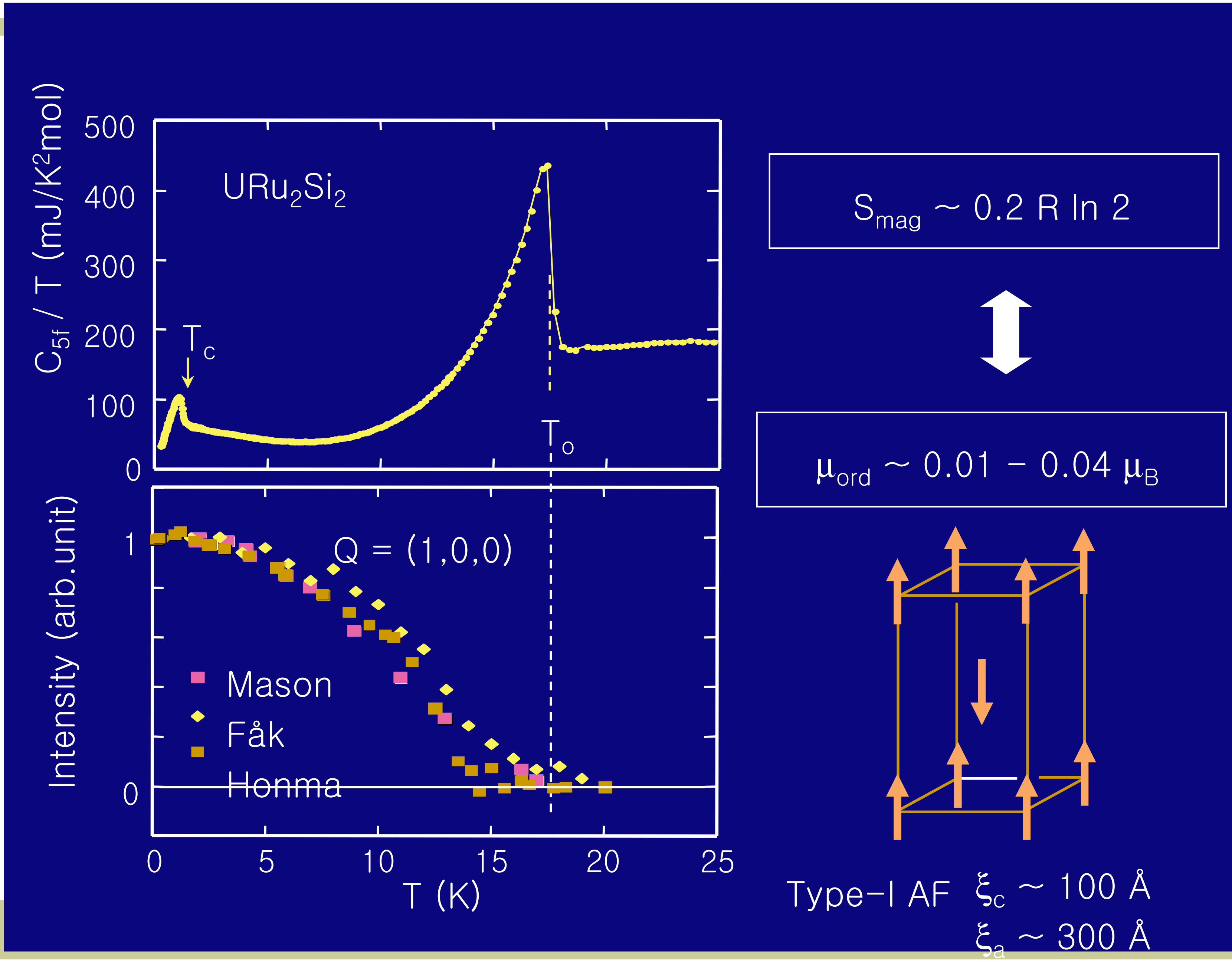


N. H. van Dijk, PRB 56, 14493 (1997).

Moment screened



Neutron Scattering, Specific heat vs. magnetic Bragg-peak intensity. Tc's.



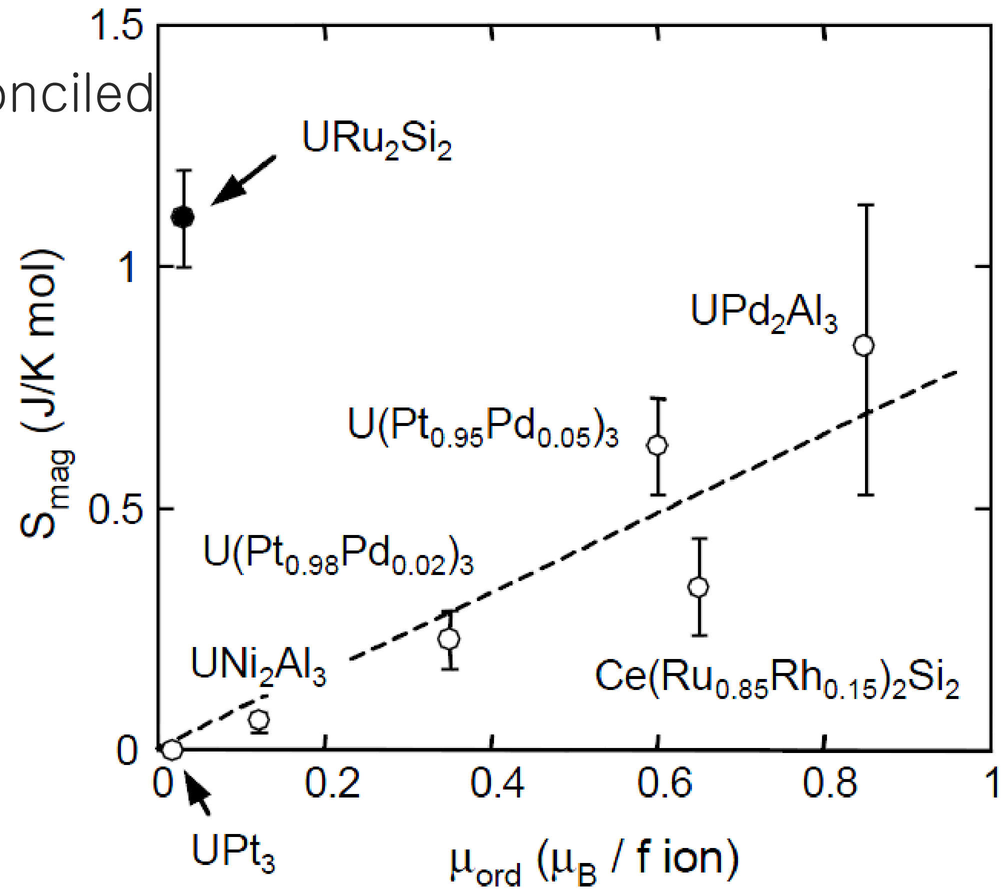
Hidden Order: The CMT dark matter problem.

- Moment is tiny
  (likely small admixture of AFM phase)

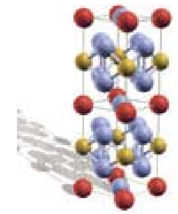
- Large loss of entropy can not be reconciled by small moment

- Some other symmetry breaks.
  Hidden order parameter

WHICH?

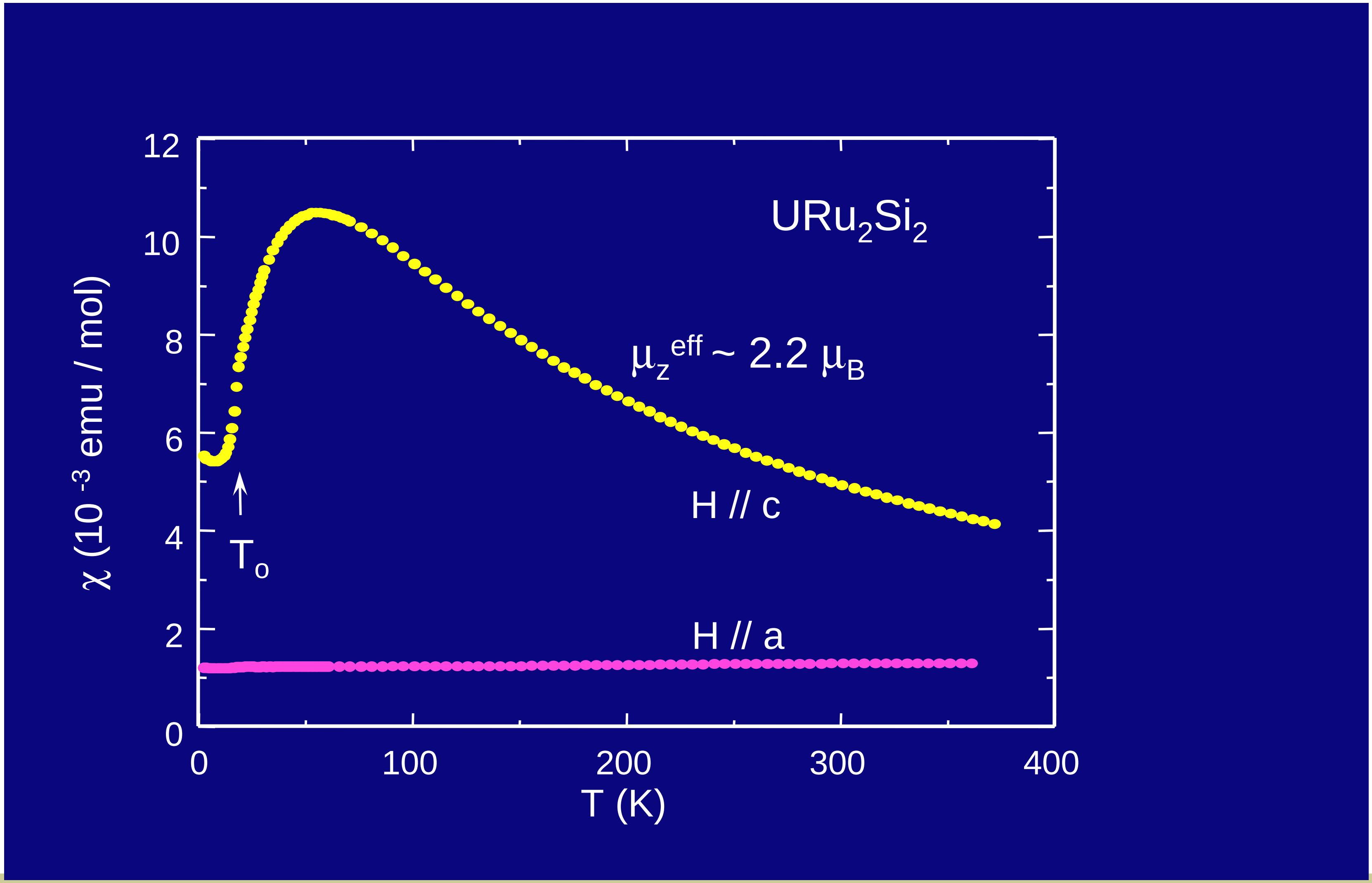


Magnetic susceptibility



Anisotropy. Moments in z−direction

Kondo? f^3 f^2 mixed ?



Optical conductivity

Pseudo-gap opens at Tc: D. A. Bonn et al. PRL (1988)

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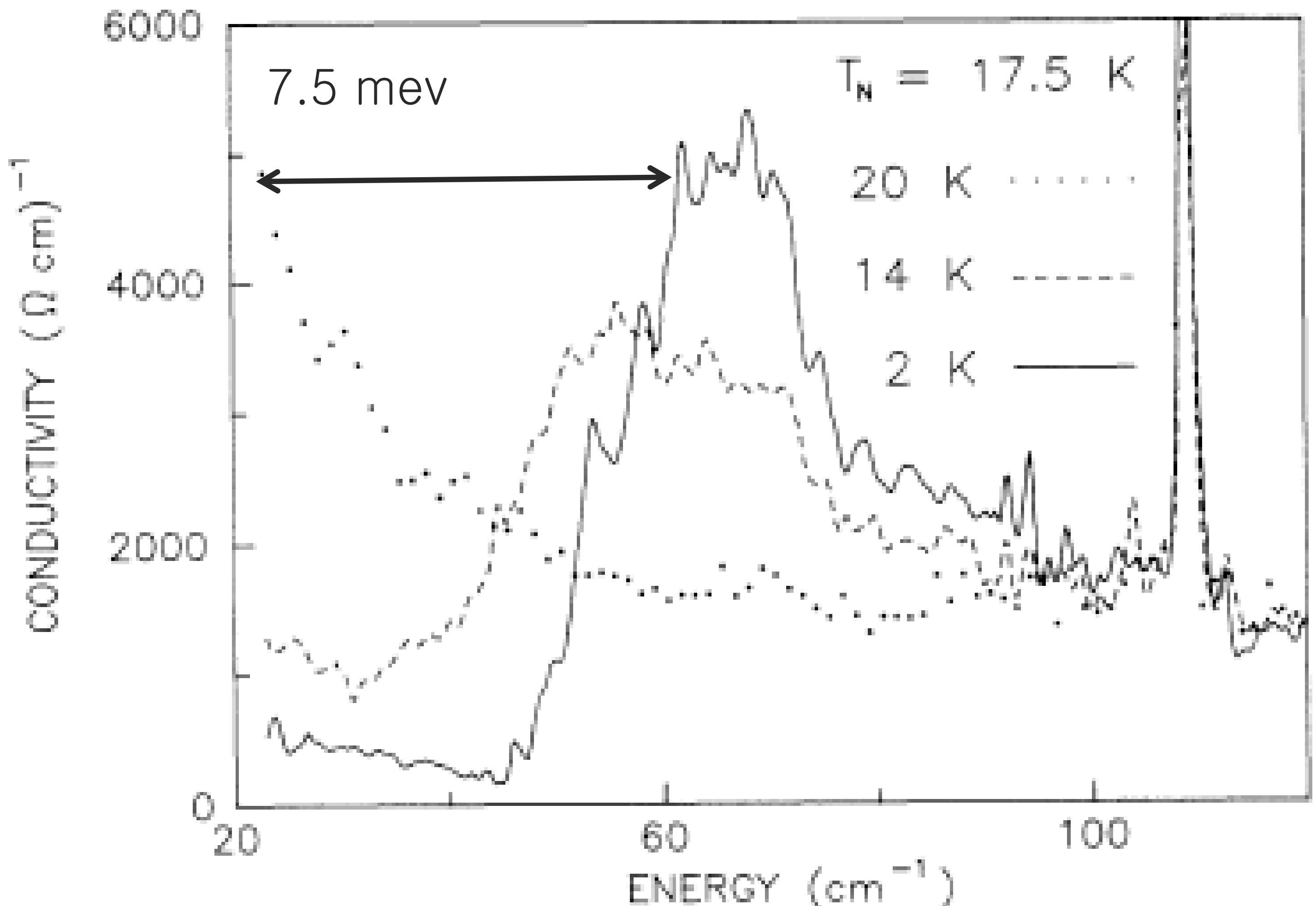
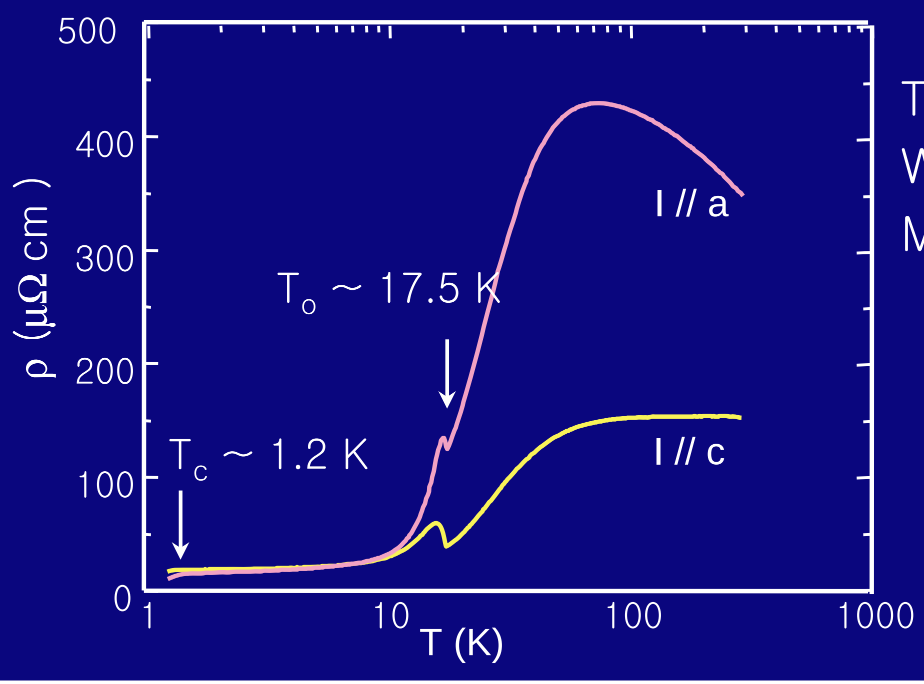


Resistivity

keeps decreasing with decreasing T



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

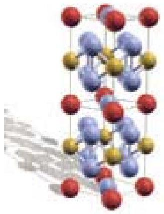
W. Schlabitz et al.(1986)

M.B. Maple et al.(1986)

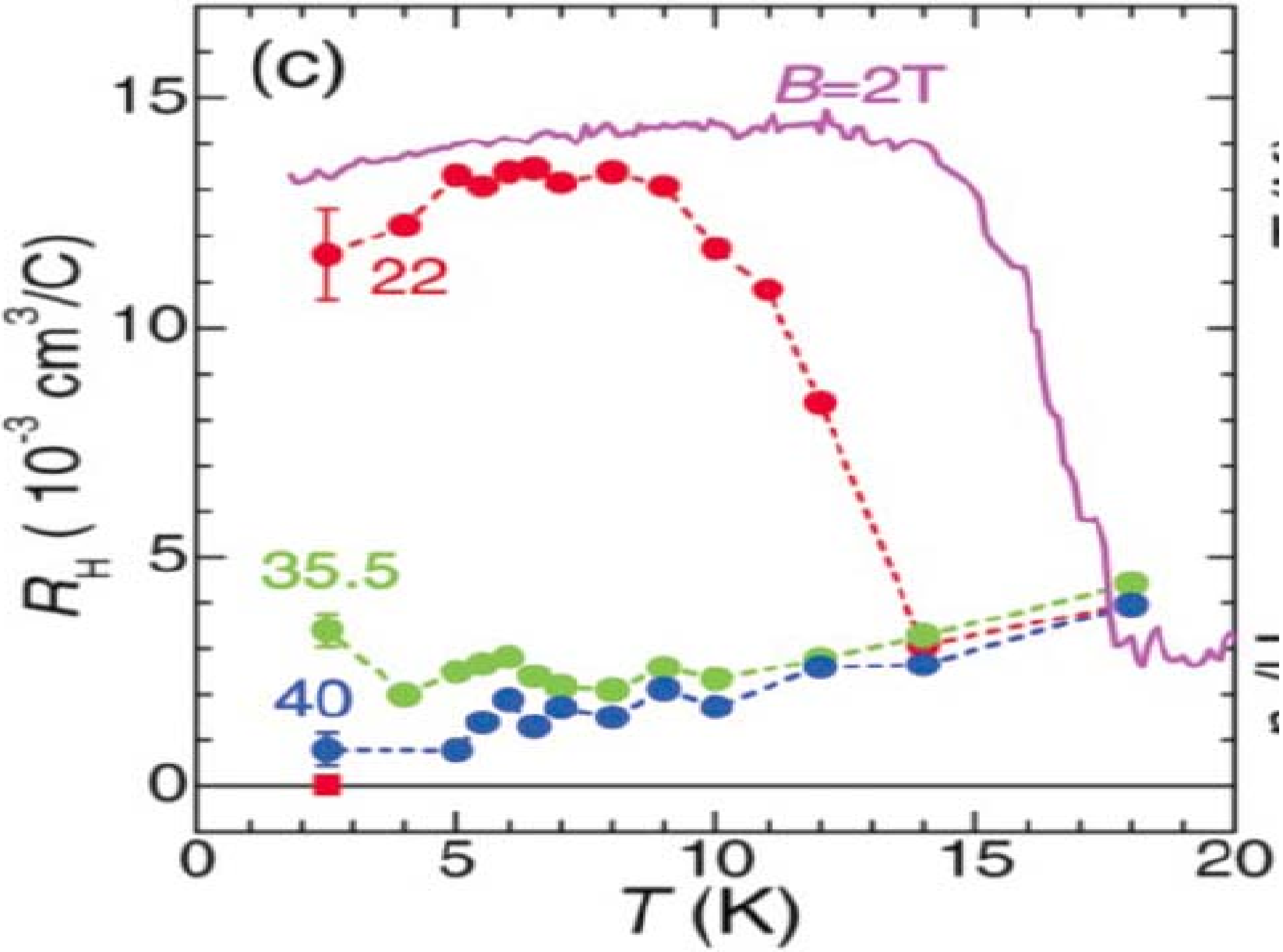
Heavy fermion at high T,low T HO + SC

Hall effect as function of temperature in different external fields,

Y.S. Oh et al. PRL 98, 016401(2007).



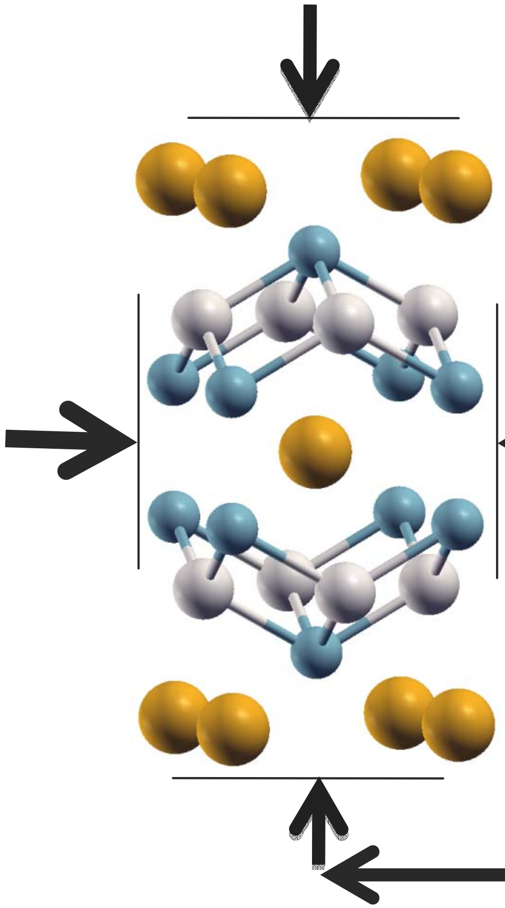
- •Fermi surface reconstruction in zero and small field
- •Very large fields polarized Fermi liquid.

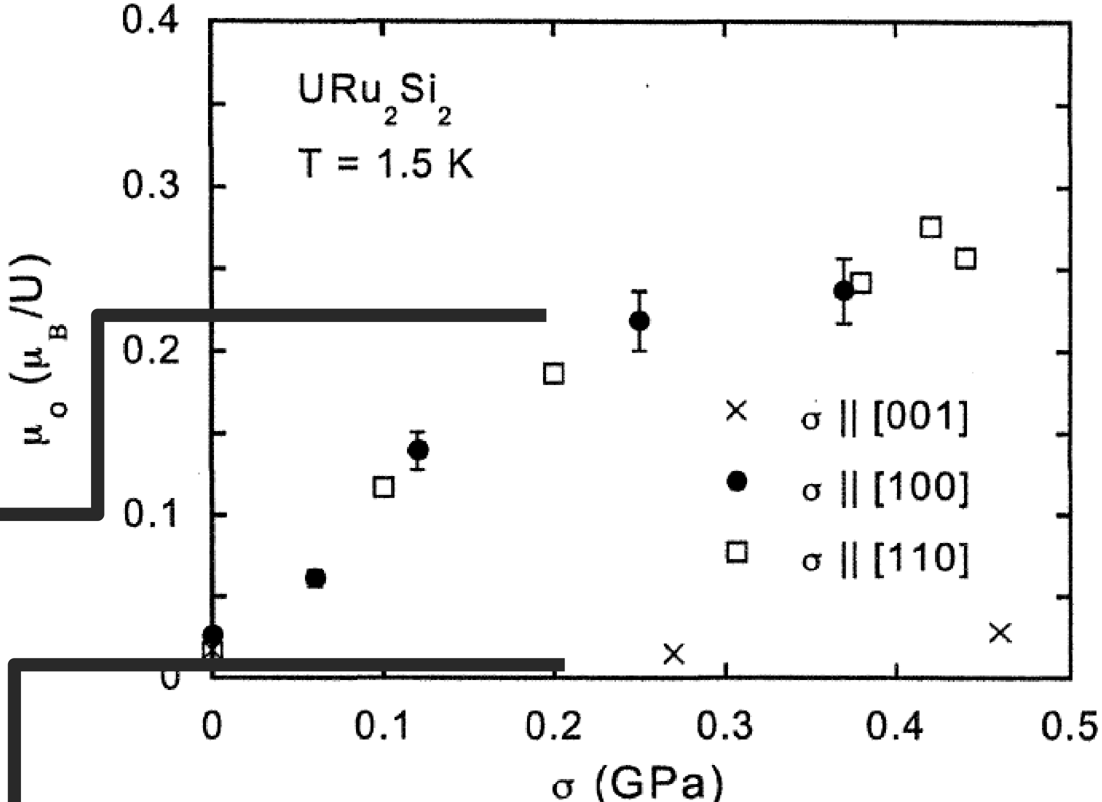


URu_2Si_2 Stress in ab plane

Large moment when stress in ab plane

No moment when stress in c plane





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

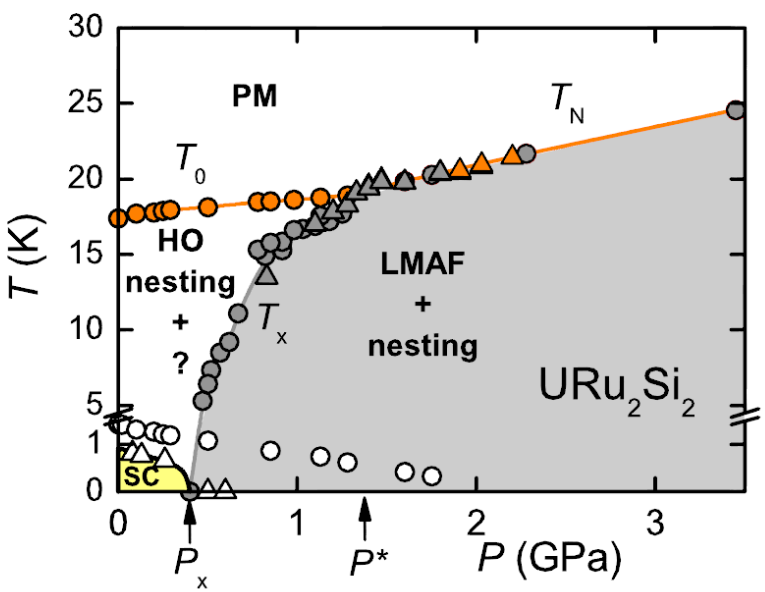
Adiabatic continuity between HO & AFM phase

PRL **98,** 166404 (2007) PHYSICAL REVIEW LETTERS week ending 20 APRIL 2007

Field-Induced Fermi Surface Reconstruction and Adiabatic Continuity between Antiferromagnetism and the Hidden-Order State in URu_2Si_2

Y. J. Jo,[1] L. Balicas,[1] C. Capan,[2] K. Behnia,[3] P. Lejay,[4] J. Flouquet,[5] J. A. Mydosh,[6] and P. Schlottmann[1]

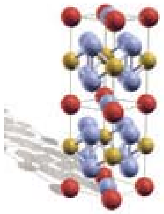
H − *T* phase diagram. Instead of phase separation between HO and antiferromagnetism our observations indicate adiabatic continuity between both orderings with field and pressure changing their relative weight.

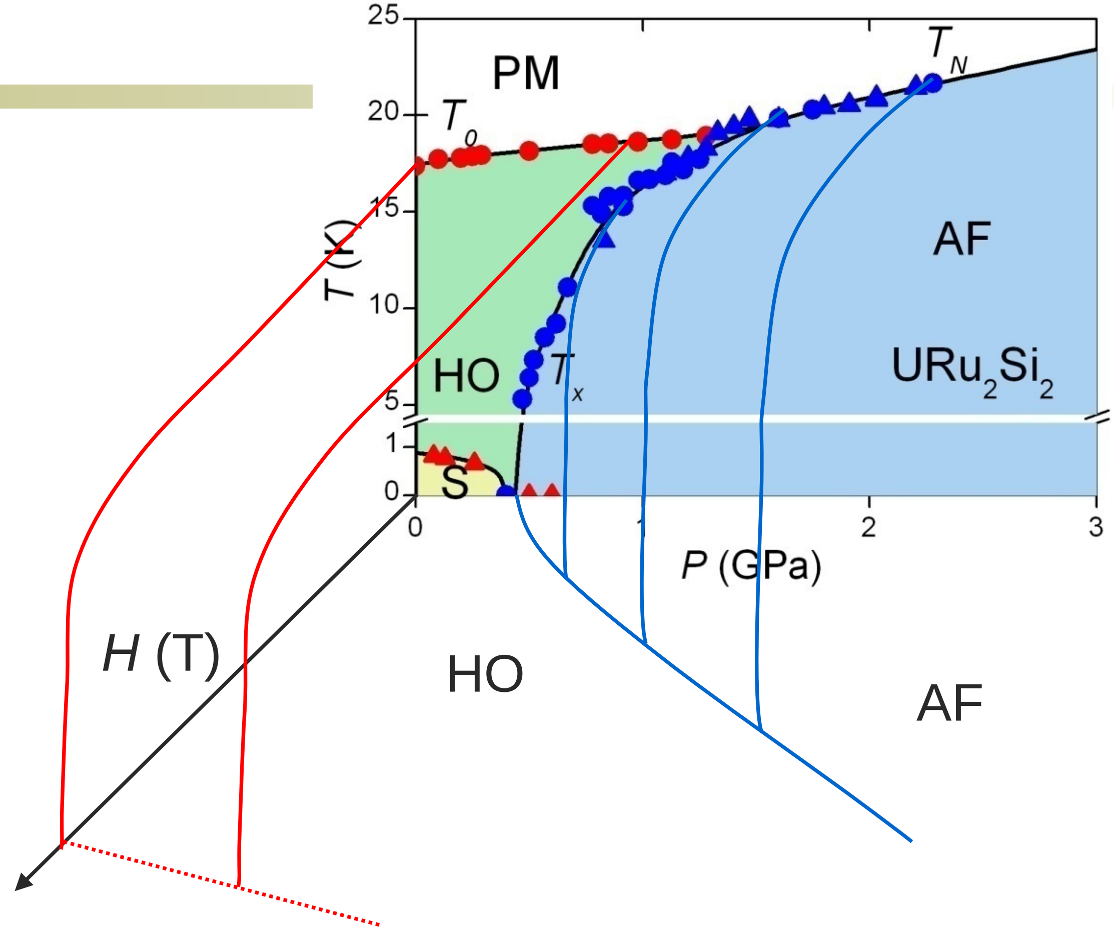


- HO under pressure converted to AFM phase through 1st order transition
- Similar T_0 and T_N
- Almost identical thermodynamic quantities (jump in Cv), quantum osc

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

Pressure magnetic field phase diagram

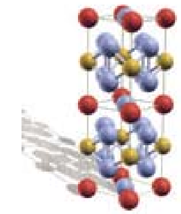


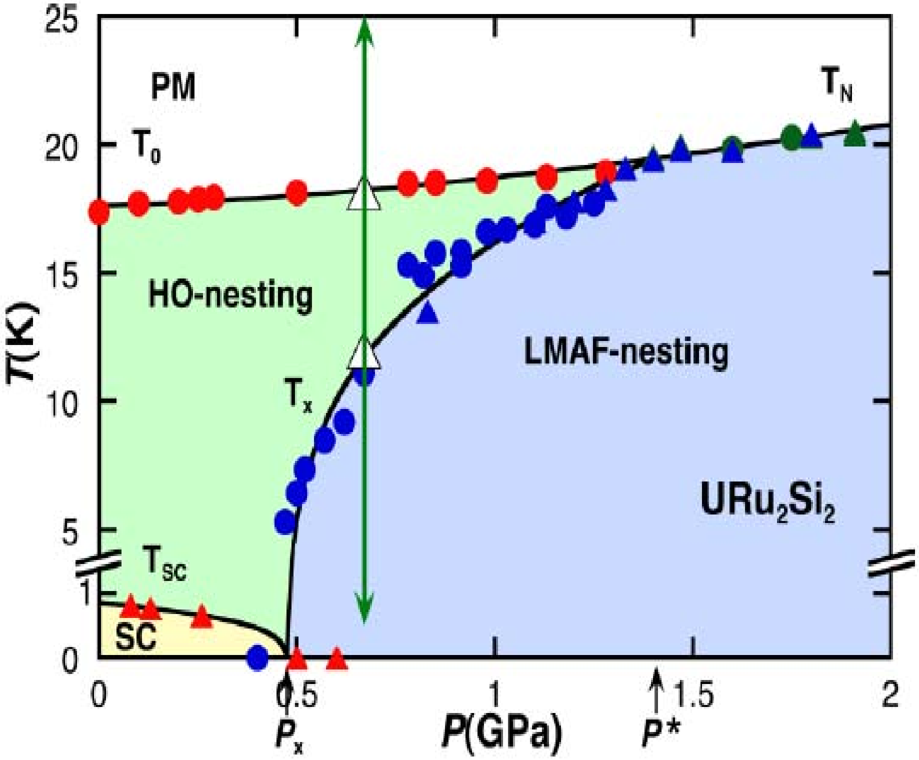


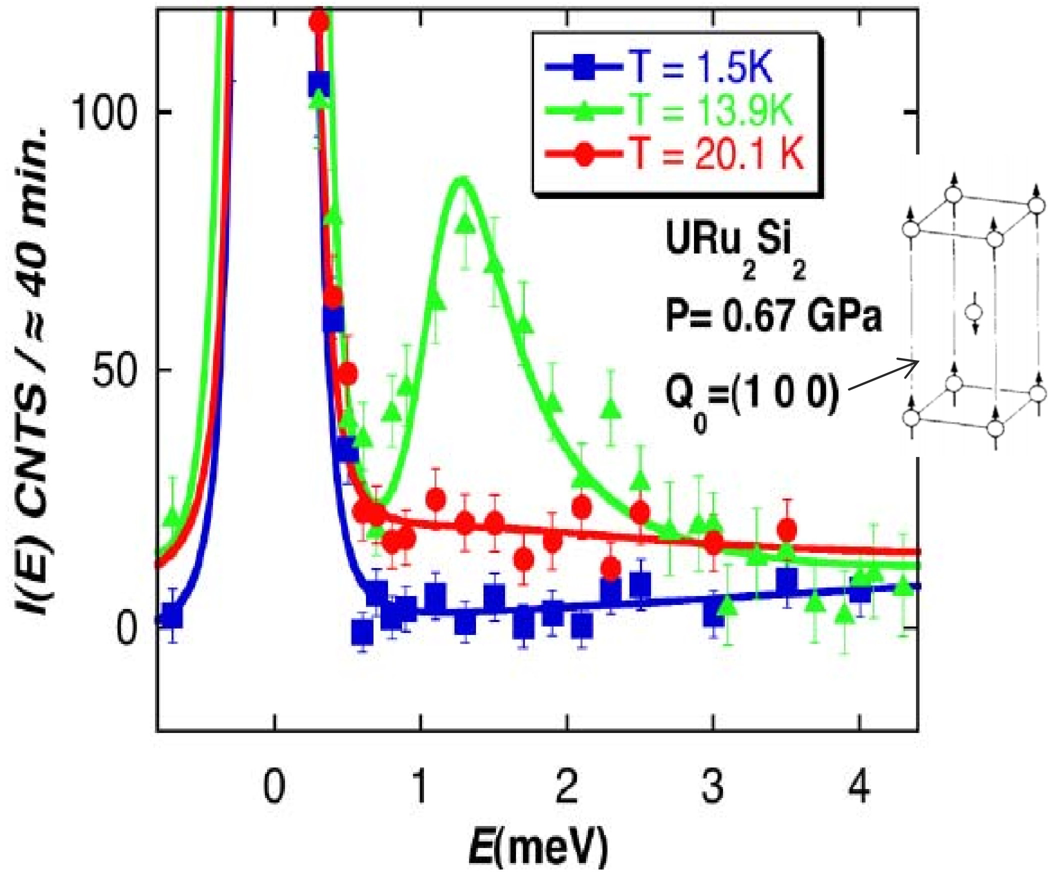
E. Hassinger et.al. PRL 77, 115117 (2008)
Aoki et.al., JPSJ 053701 (2009)

- Pressure induces AF phase, field has opposite effect

Key experiment: Neutron scattering

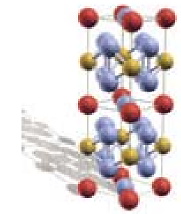




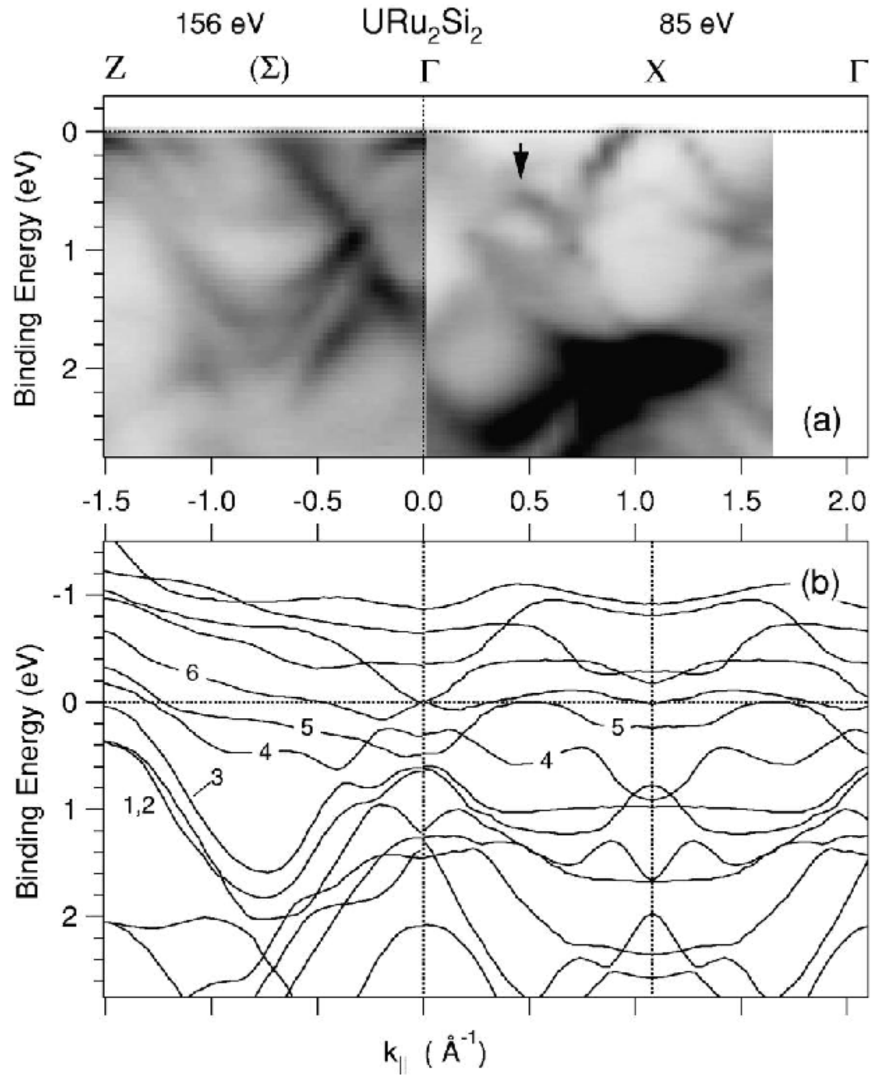


The low energy resonance

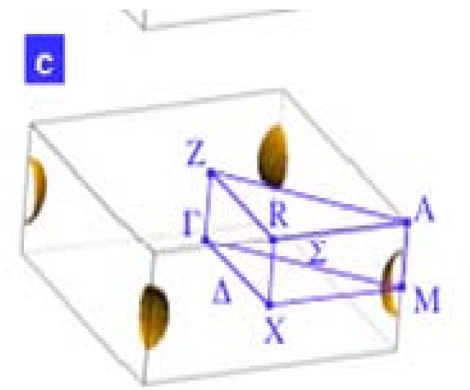
A.Villaume, F. Bourdarot, E. Hassinger, S. Raymond, V. Taufour, D. Aoki, and J. Flouquet, PRB 78, 012504 (2008)



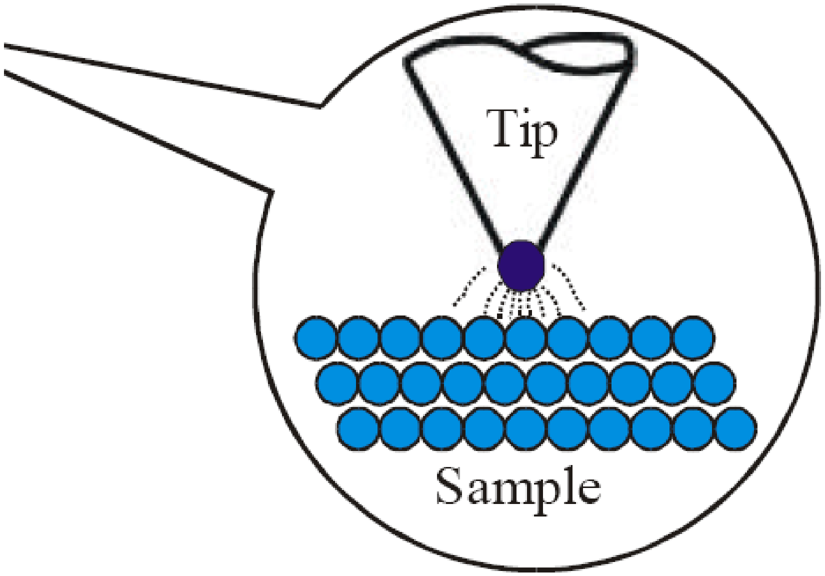
ARPES does not agree with LDA



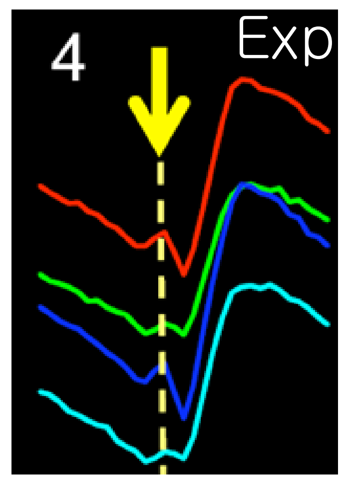
J.D. Denlinger et.al., 2001

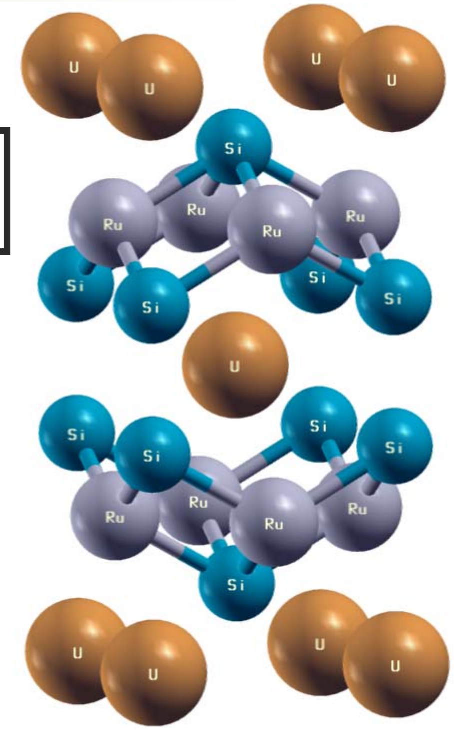


Scanning Tunneling Microscopy



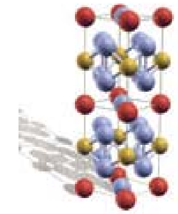
dI/dV → $\rho(r,\omega)$



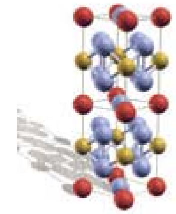


G. Luke, A. Schmidt, P. Wah
l,
M. Hamidian, J. C. Davis,
unpublished

Comments concerning Hidden Order



- URu2Si2, at high temperatures is not too different from a garden variety heavy fermion.
- ARPES does not agree with LDA at 30 K.
- HO converts to LMAF by pressure through a first order line.
- HO can be stabilized by B and destroyed by Rh-doping
- 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: (1,0,0)@1.5-2.0meV and
  (1.4,0,0)@5meV of longitudinal fluctuations/excitations.
- HO (but not LAMF) turns into superconductivity at 1.7 K.



Some proposals for the hidden order in the literature

•Lev. P. Gorkov: 1996:

-Mixed valency, coupling to lattice degrees of freedom.

• Chandra *et al.*, Nature'02

- Incommensurate Orbital Antiferromagnetism (based on "old" NMR)

• Mineev & Zhitomirsky, PRB '05

- SDW (with tiny moment… problem with entropy)

• Varma & Zhu, PRL'06

- Helical Order, Pomeranchuk instability of the Fermi surface ?

• Elgazaar, & Oppeneer, Nature Materials'08

- DFT: antiferromagnetic order parameter, but weak AFM moment (can not explain large entropy loss, stress, adiabatic continuity, moment in z dir.…)

• Santini and Amoretti PRL 04

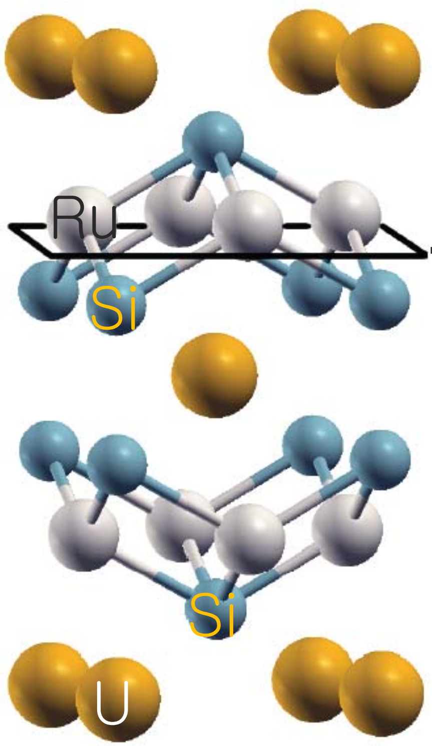
-Quadrupolar ordering.

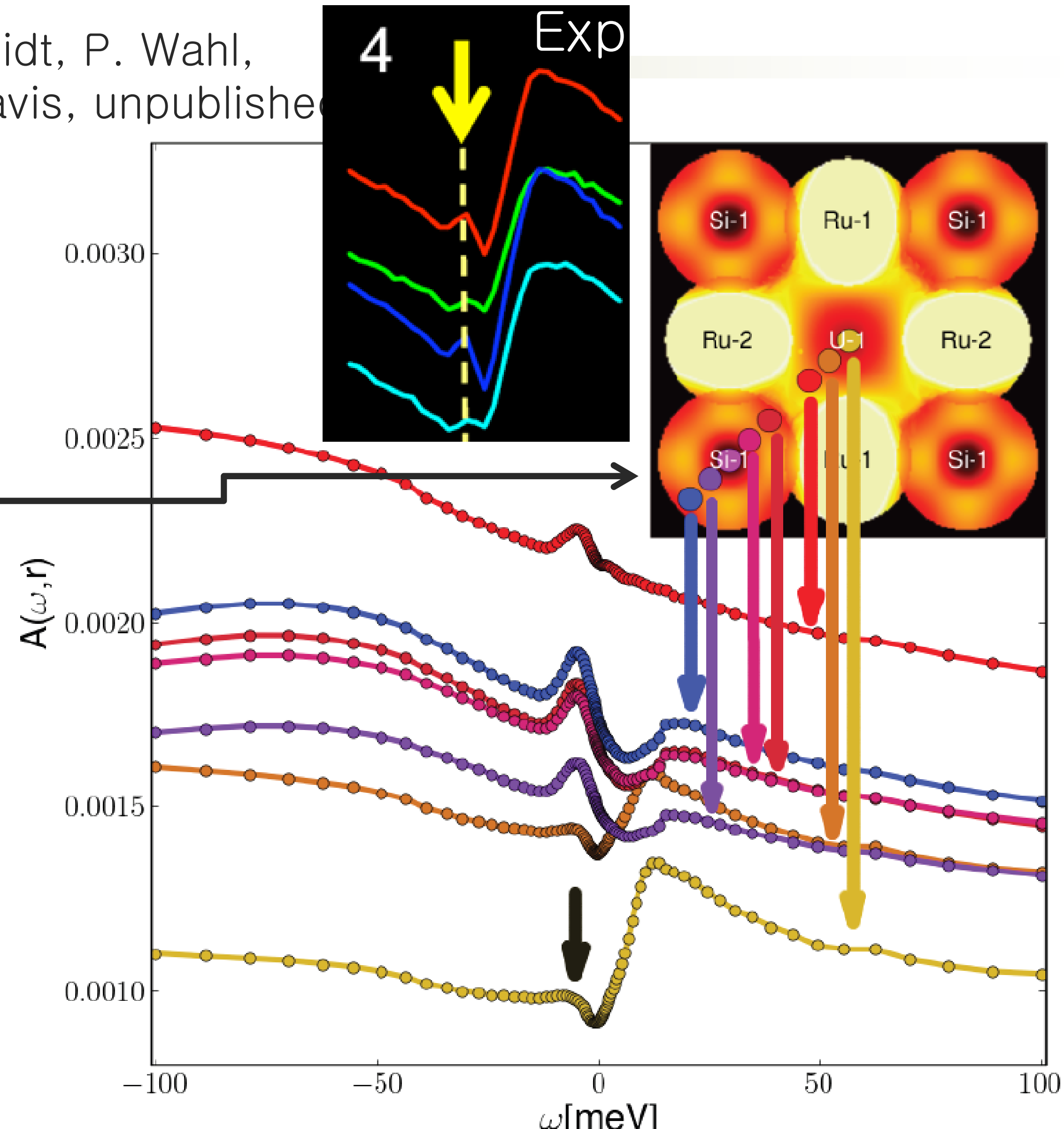
• Fazekas and Kiss PRB 07

-Octupolar ordering. *[Many Many more , even recently , including us ☺]*

Dynamical Mean Field calculation

G. Luke, A. Schmidt, P. Wahl,
M. Hamidian, J. C. Davis, unpublished



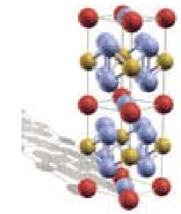


Fano lineshape:

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

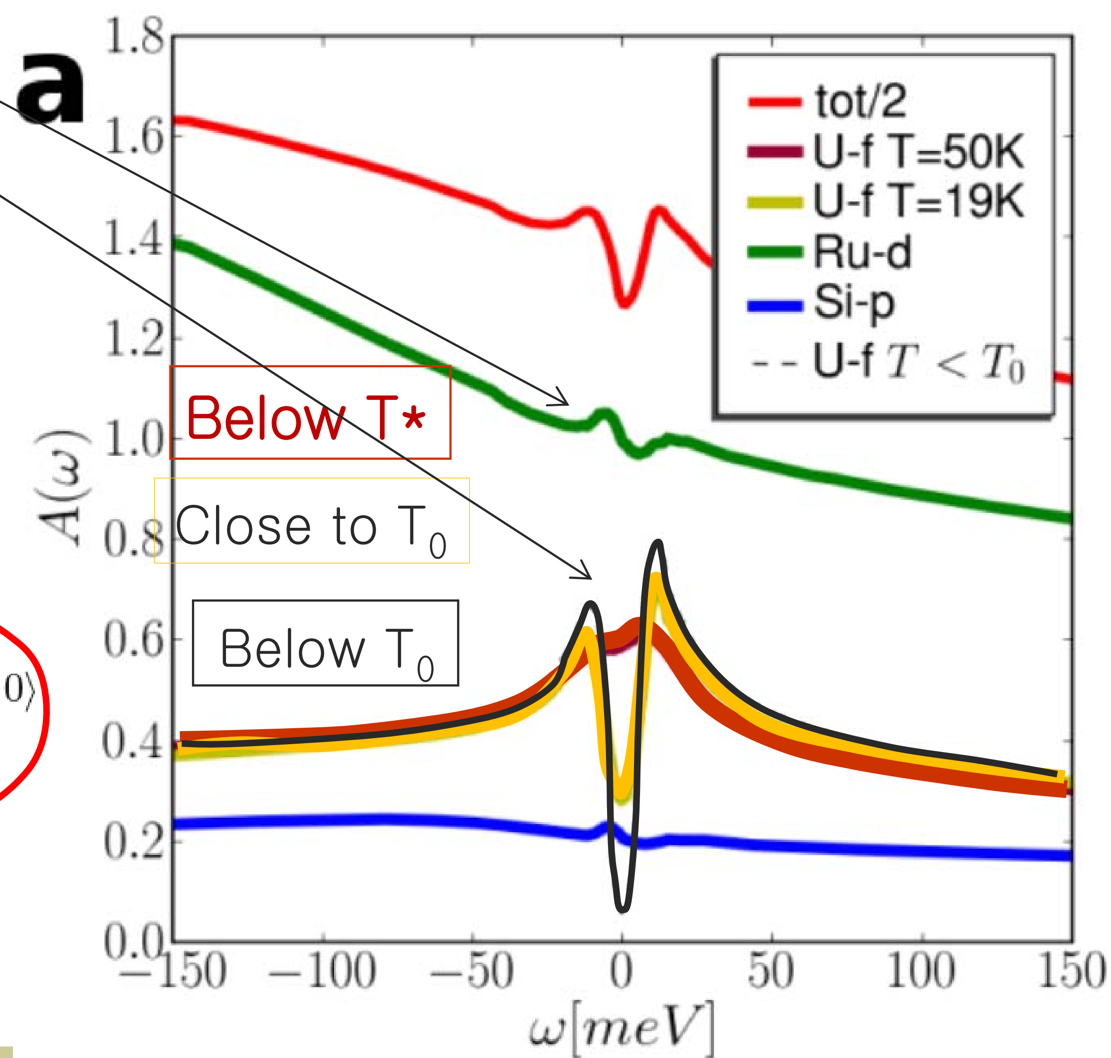
q~1.24, Γ~6.8meV, very similar to exp

Origin of gapping?

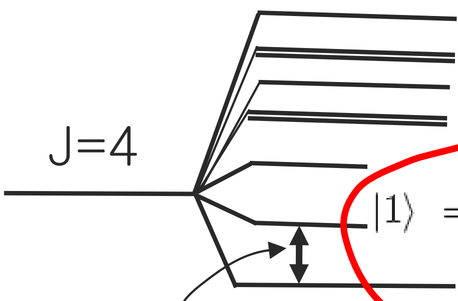


Partial DOS

Small effects on spd electrons
Large effect on U−f's



Ground state atomic multiplet of f^2 configuration in tetragonal field

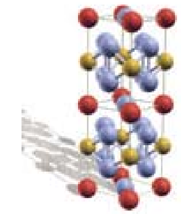


$$|1\rangle = \frac{\cos(\phi)}{\sqrt{2}}(|4\rangle + |-4\rangle) - \sin(\phi)|0\rangle$$

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

Only 35K!

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

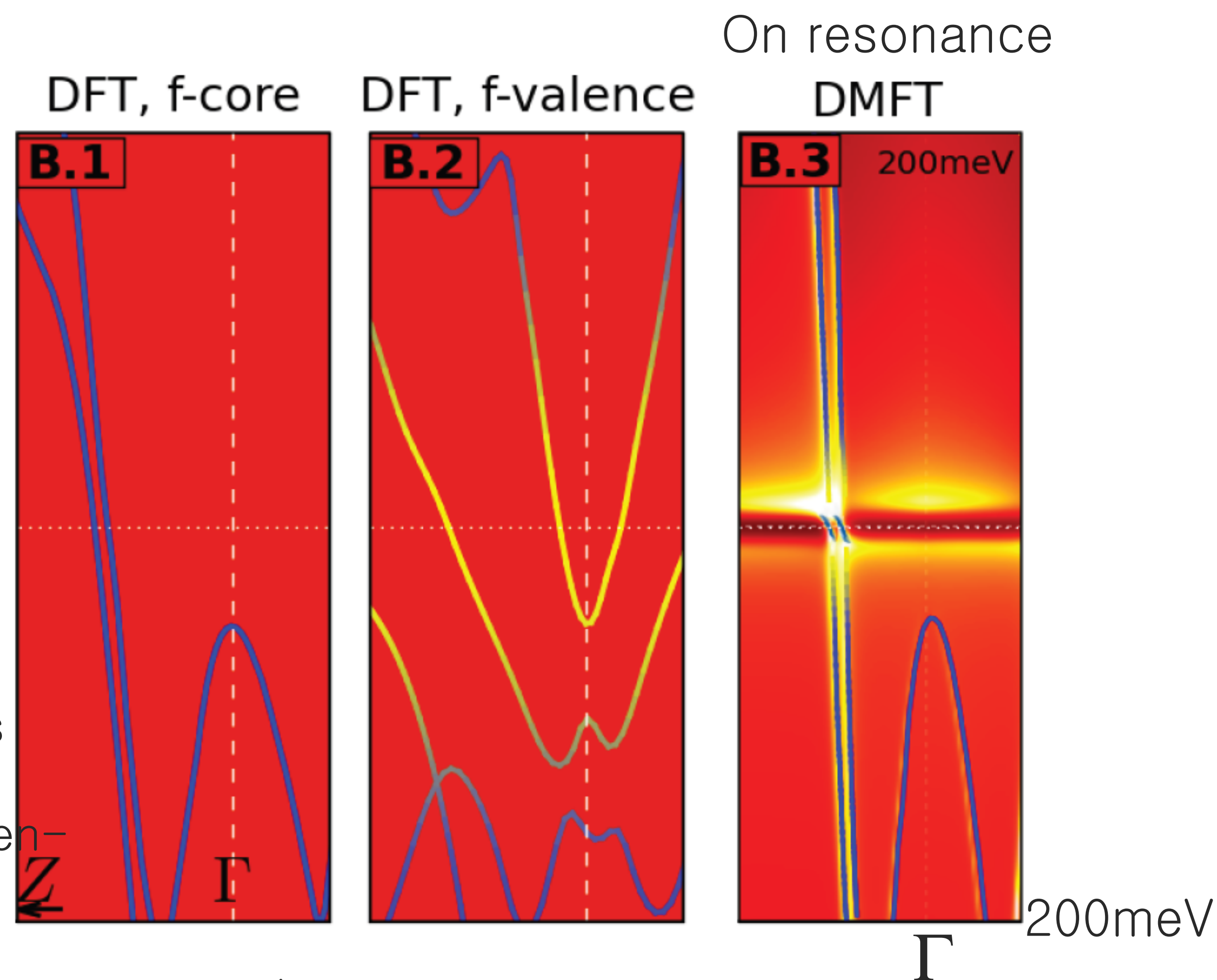


Partially Arrested Kondo effect

•DFT f–core: goof descriptio n of bands 30meV away from EF

•DFT f–valence: many f–bands at EF, substantial disagreement with ARPES & DMFT

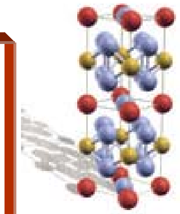
DMFT:
very narrow region of f–spectral weight ±10meV around EF appears below T*~70K
Below 35K, partial gap starts to open–
–>singlet to singlet Kondo effect



At low temperature, two broken symmetry states!

KH, G. Kotliar, Nature Physics 5, 796 - 799 (2009).

DMFT allows two broken symmetry states at low T



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

Large moment phase:

$$\Delta = \left(\begin{array}{c|cccccc} \text{J=5/2} & -5/2 & -3/2 & -1/2 & 1/2 & 3/2 & 5/2 \\ \hline -5/2 & \Delta_A & 0 & 0 & 0 & \Delta_\varepsilon & 0 \\ -3/2 & 0 & \Delta_B & 0 & 0 & 0 & \Delta'_\varepsilon \\ -1/2 & 0 & 0 & \Delta_C & 0 & 0 & 0 \\ 1/2 & 0 & 0 & 0 & \Delta'_C & 0 & 0 \\ 3/2 & \Delta_\varepsilon & 0 & 0 & 0 & \Delta'_B & 0 \\ 5/2 & 0 & \Delta'_\varepsilon & 0 & 0 & 0 & \Delta'_A \end{array}\right)$$

Moment free phase:

$$\Delta = \left(\begin{array}{c|cccccc} \text{J=5/2} & -5/2 & -3/2 & -1/2 & 1/2 & 3/2 & 5/2 \\ \hline -5/2 & \Delta_A & 0 & 0 & 0 & \Delta_\varepsilon + \Delta_\alpha & 0 \\ -3/2 & 0 & \Delta_B & 0 & 0 & 0 & \Delta_\varepsilon + \Delta_\beta \\ -1/2 & 0 & 0 & \Delta_C & 0 & 0 & 0 \\ 1/2 & 0 & 0 & 0 & \Delta_C & 0 & 0 \\ 3/2 & \Delta_\varepsilon - \Delta_\alpha & 0 & 0 & 0 & \Delta_B & 0 \\ 5/2 & 0 & \Delta_\varepsilon - \Delta_\beta & 0 & 0 & 0 & \Delta_A \end{array}\right)$$

tetragonal symmetry broken->
these terms nonzero

Valence histogram point of view

The DMFT density matrix has most weight in two singlet f^2 configuratio
ns
Close to f^2 “Kondo” limit, (n_f~2.2), J=4, two low lying singlets.

Therefore there are two singlets relevant at low energies but they are n
ot Kramer doublets: Conspiracy between cubic crystal field splittings
and tetragonal splittings bring these two states close.

This is why URu2Si2 is sort of unique.

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

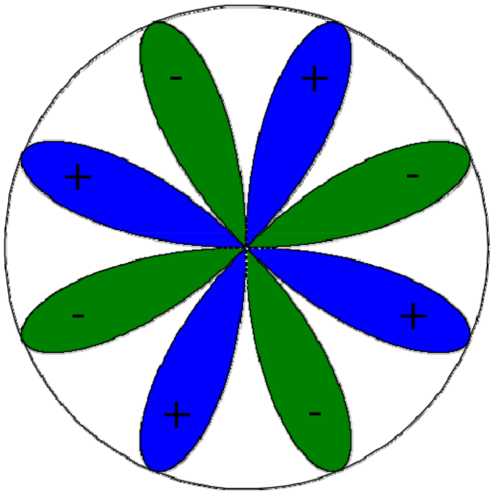
$$|1\rangle = \frac{\cos(\phi)}{\sqrt{2}}(|4\rangle + |-4\rangle) - \sin(\phi)|0\rangle$$

Both states are time-reversal invariant

DMFT order parameter

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

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



Order parameter:

$$\psi_i = \langle X_{01}(\mathbf{R}_i)\rangle$$

$$\mathrm{Im}\psi \propto \langle J_z\rangle$$

$$\mathrm{Re}\psi \propto \langle (J_xJ_y + J_yJ_x)(J_x^2 - J_y^2\rangle$$

Different orientation gives different phases: adiabatic continuity explained!

Does *not* break the *time reversal*, *nor C4* symmetry. It breaks *inversion symmetry*.

In the atomic limit:

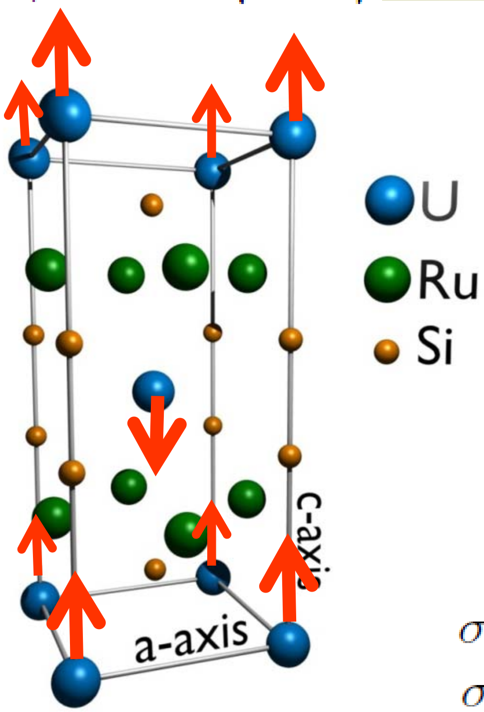
$$|gs>= \cos(\theta)|0\rangle + \sin(\theta)e^{i\varphi}|1\rangle$$

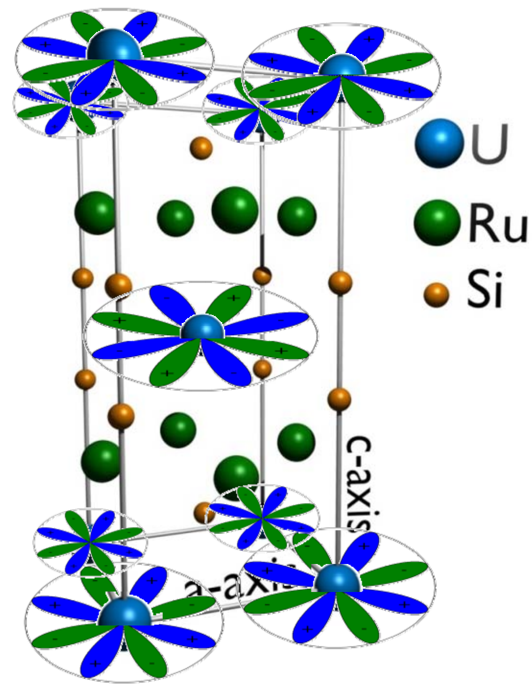
$$\langle gs|\mathbf{J}|gs\rangle = 4\cos(\phi)\sin(2\theta)\sin(\varphi)*(0,0,1)$$

Moment only in z-direction!

The two broken symmetry states

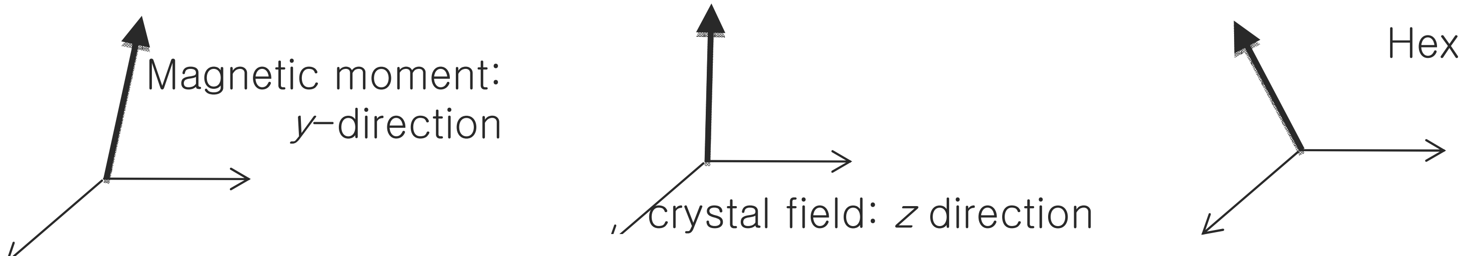
$$\mathrm{Im}\psi \propto \langle J_z \rangle \qquad \mathrm{Re}\psi \propto \langle (J_xJ_y + J_yJ_x)(J_x^2 - J_y^2) \rangle$$





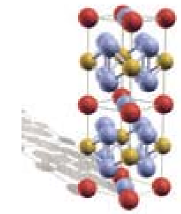
A toy model

$$\sigma_3 = |\emptyset\rangle\langle\emptyset| - |1\rangle\langle 1|$$
$$\sigma_1 = |\emptyset\rangle\langle 1| + |1\rangle\langle\emptyset|$$
$$\sigma_2 = i(|1\rangle\langle\emptyset| - |\emptyset\rangle\langle 1|)$$



XY−Ising

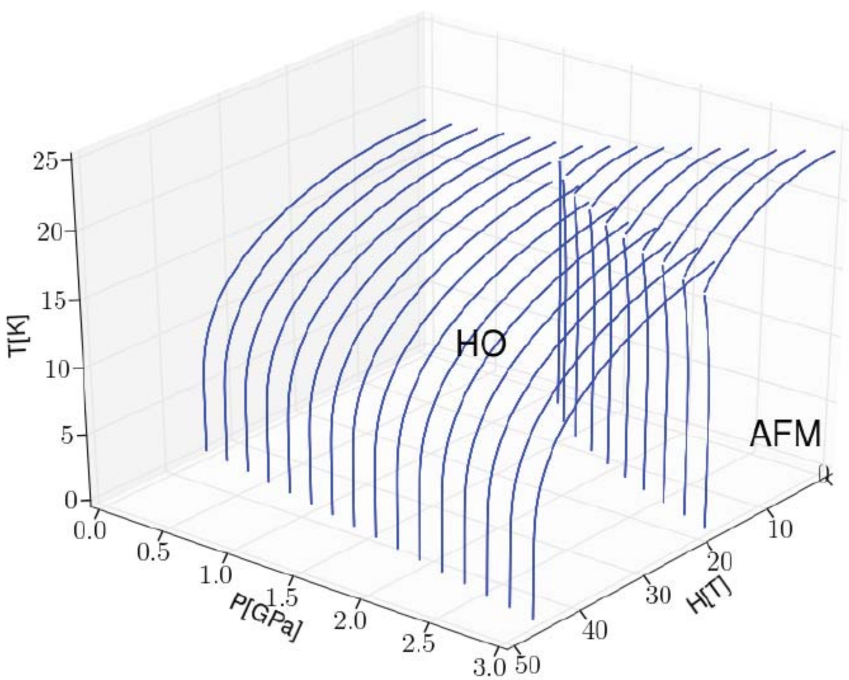
$$H = \sum_i -\frac{\Delta}{2}\sigma_3^i - \frac{1}{2} g\mu_B B |\langle 1|J_z|\emptyset\rangle| \sigma_2^i + \sum_{i,j} \frac{1}{2}(J_{ij}^1 \sigma_1^i \sigma_1^j + J_{ij}^2 \sigma_2^i \sigma_2^j)$$



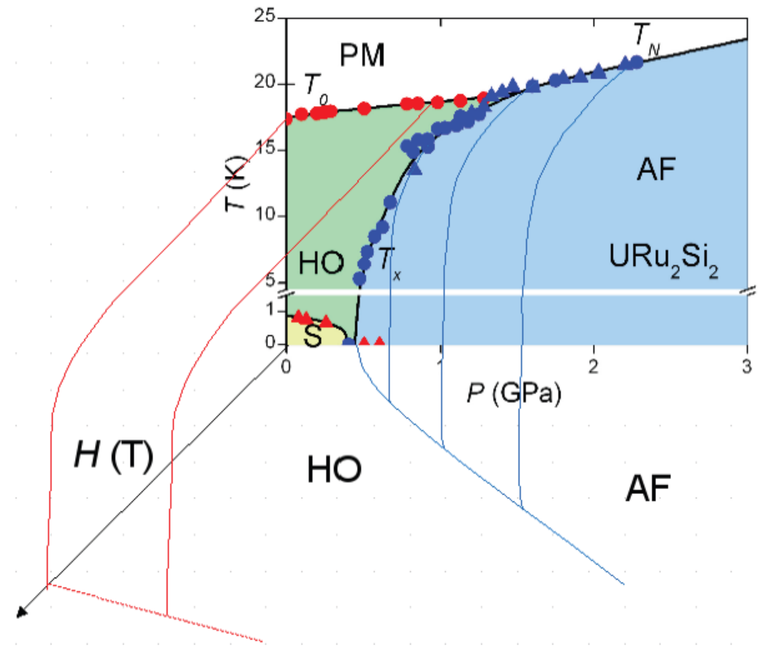
HO & AFM in magnetic field

KH & G.Kotliar, arxiv: arXiv:0907.3892

$$F[h,\psi] = \frac{1}{2}\sum_{ij,\alpha=(1,2)} J_{ij}^{\alpha}\,\psi_i^{\alpha}\psi_j^{\alpha} - \sum_{i,\alpha=(1,2)} (h_i^{\alpha}+b^{\alpha})\psi_i^{\alpha} - T\sum_i \log\left(\cosh\left(\beta\sqrt{(\Delta/2)^2+(h_i^1)^2/2+(h_i^2)^2/2}\right)\right)$$



Mean field



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

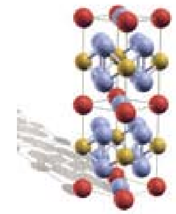
Only two fitting parameters: J_{eff}^1 , J_{eff}^2

$$J_{eff} = 4|J_1| + 8J_2$$

determined by exp. transition temperature :

$$J_{eff}^1 = \frac{\Delta}{\tanh(\Delta/(2T_0))}$$

$$J_{eff}^2 = \frac{\Delta}{\tanh(\Delta/(2T_N))}$$



HO & AFM under stress

KH & G.Kotliar, arxiv: arXiv:0907.3892

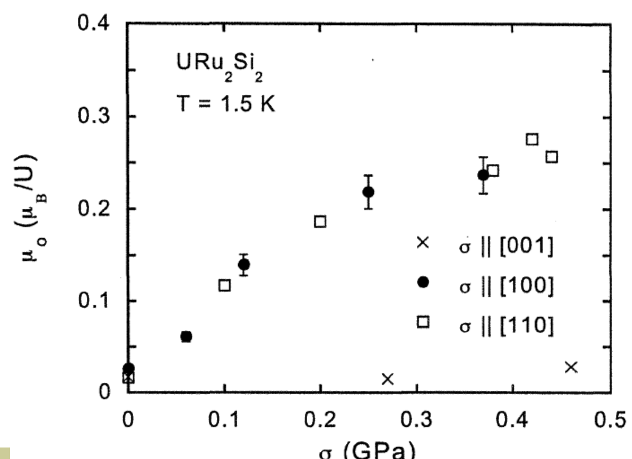
s sensitive to compression (strain), modeled by: $J_{ij}^{\alpha} \rightarrow J_{ij}^{\alpha}(1 + g_{\alpha}(\varepsilon_{xx} + \varepsilon_{yy}))$

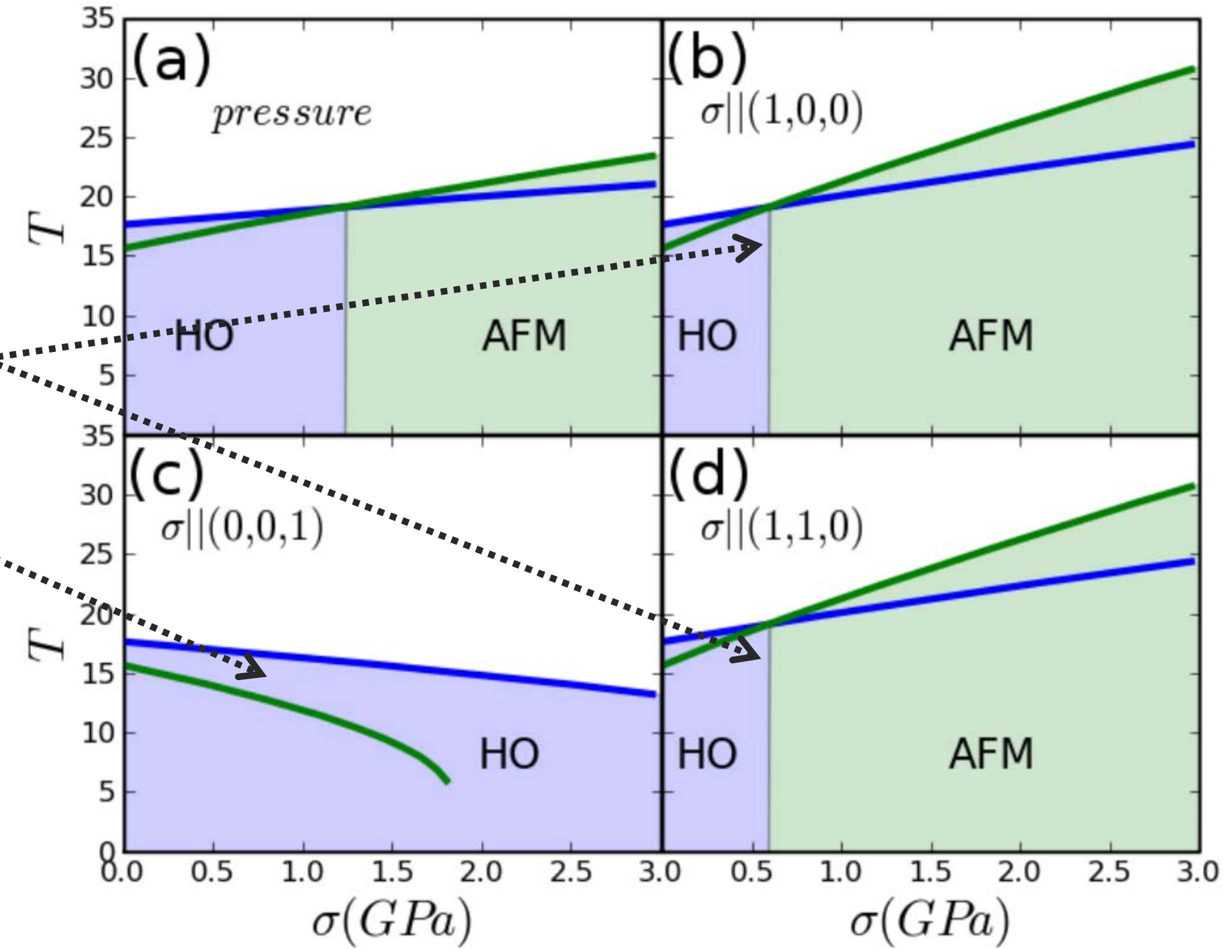
$$\sigma = C\varepsilon$$

Very different effect of
in plane stress
and uniaxial stress

In plane stress favors AFM state

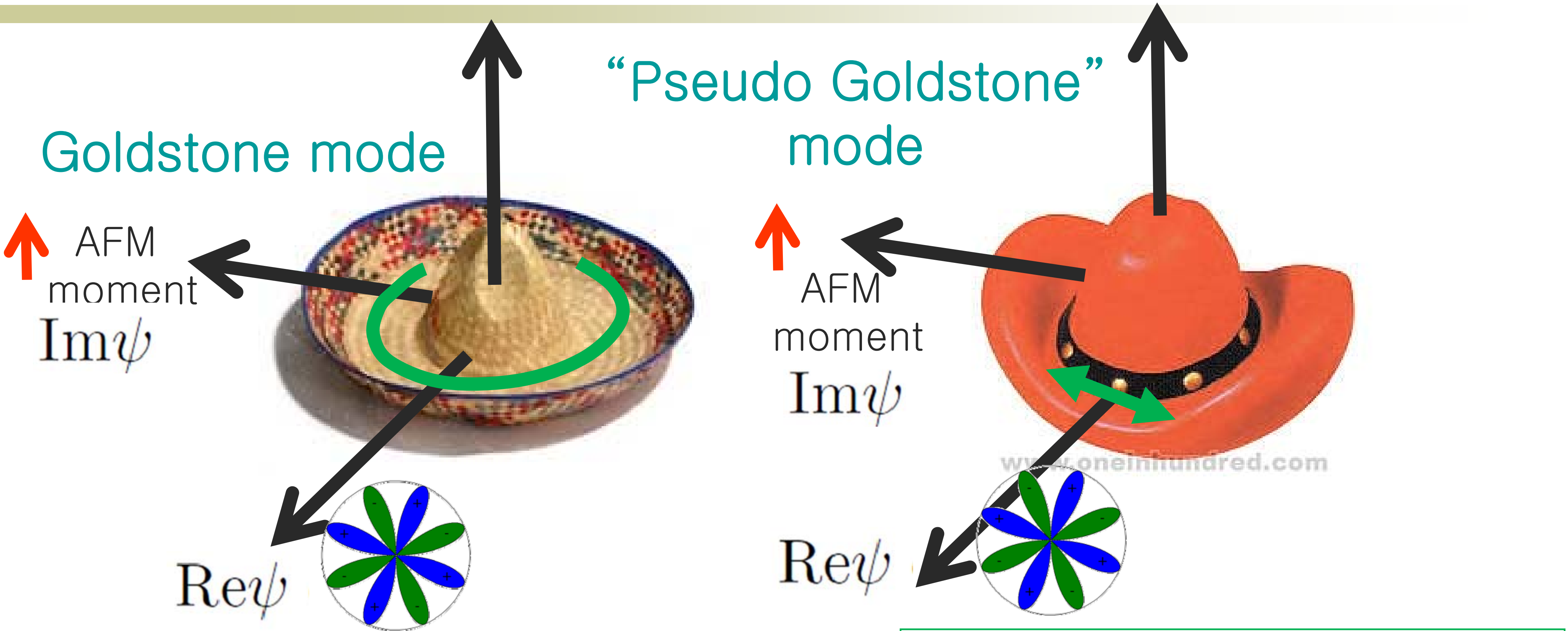
c−axis stress favors HO





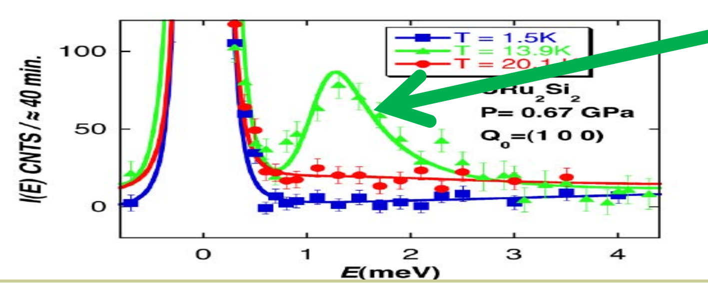
M Yokoyama, JPSJ 71, Supl 264 (2002).

Neutron scattering experiments



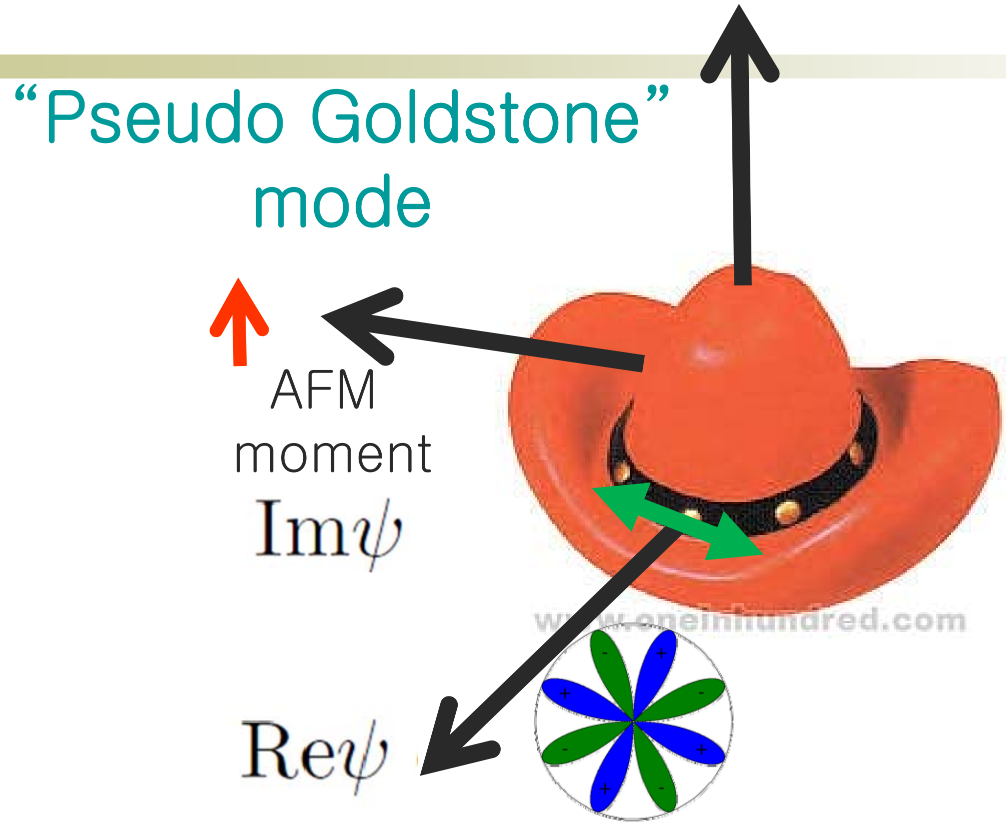
hexadecapole

Symmetry is approximate
"Pseudo–Goldstone" mode
Fluctuation of m – finite mass



The exchange constants J
are slightly different in the two phases (~6%)

Predictions for the mode

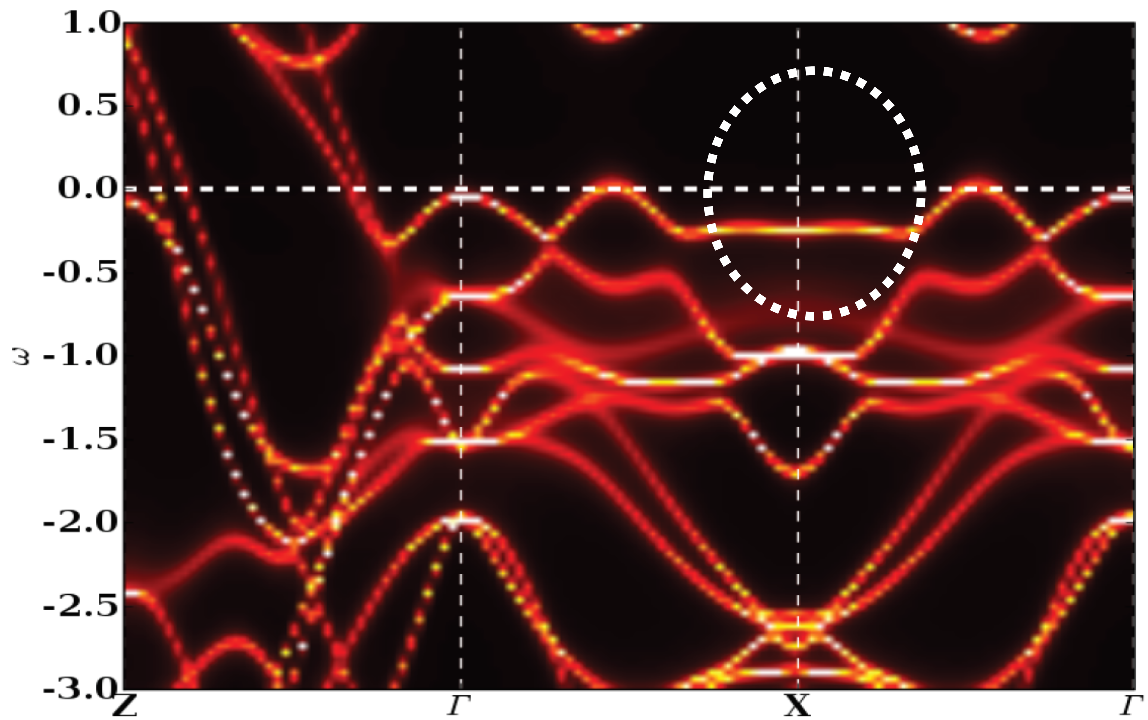


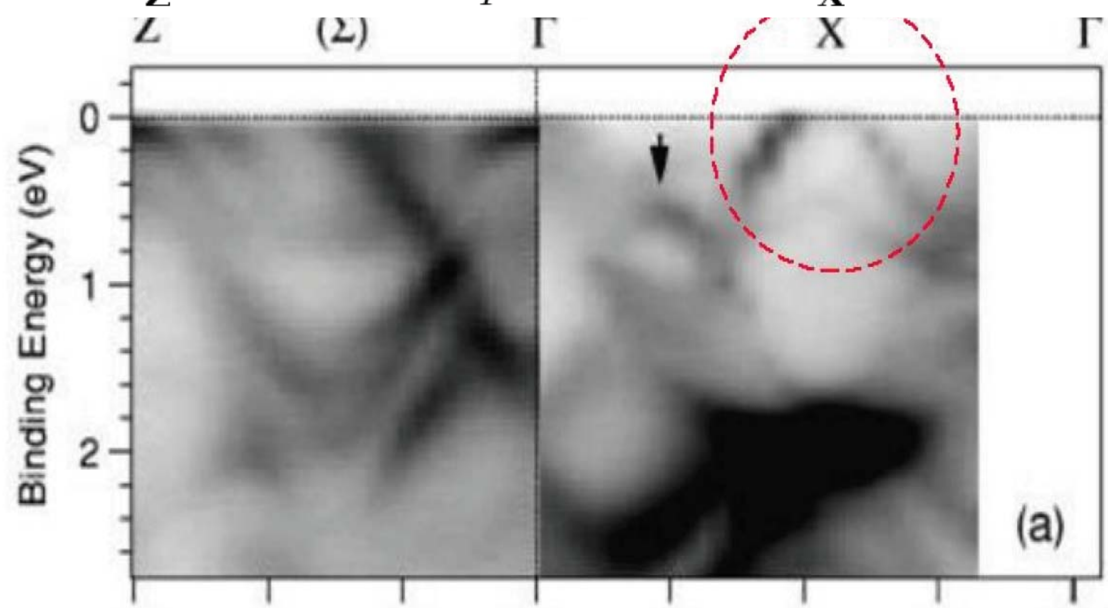
Predictions:
The mode energy should decrease with pressure
(since the difference in the exchange constants decreases with increasing pres sure)
The mode energy should increase with magnetic field
(since the magnetic field destabilizes the antiferromagnetic phase).

DMFT A(k,ω) vs ARPES

Off resonance

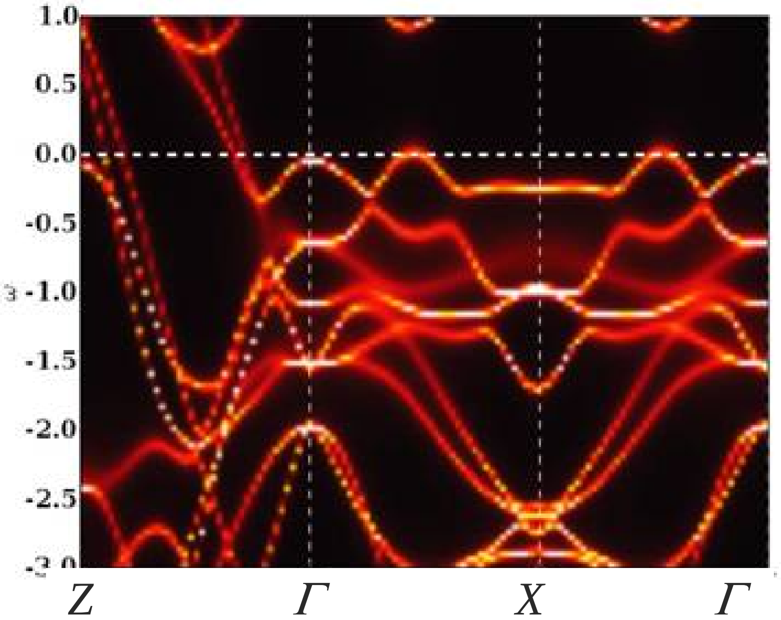
Very good agreement,
except at X point





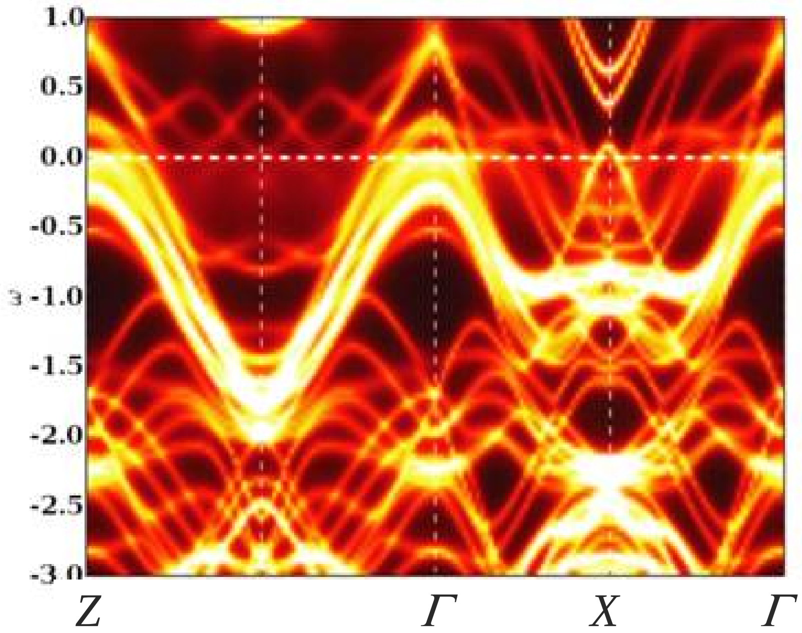
Surface origin of pocket at X point

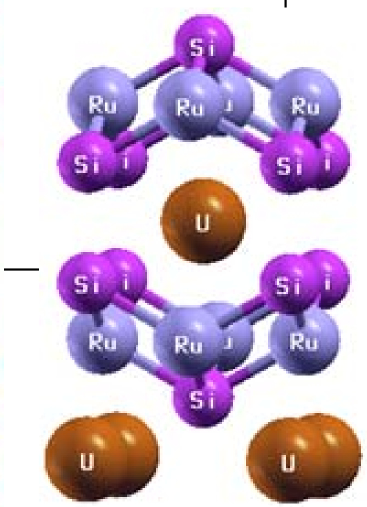
LDA+DMFT - bulk



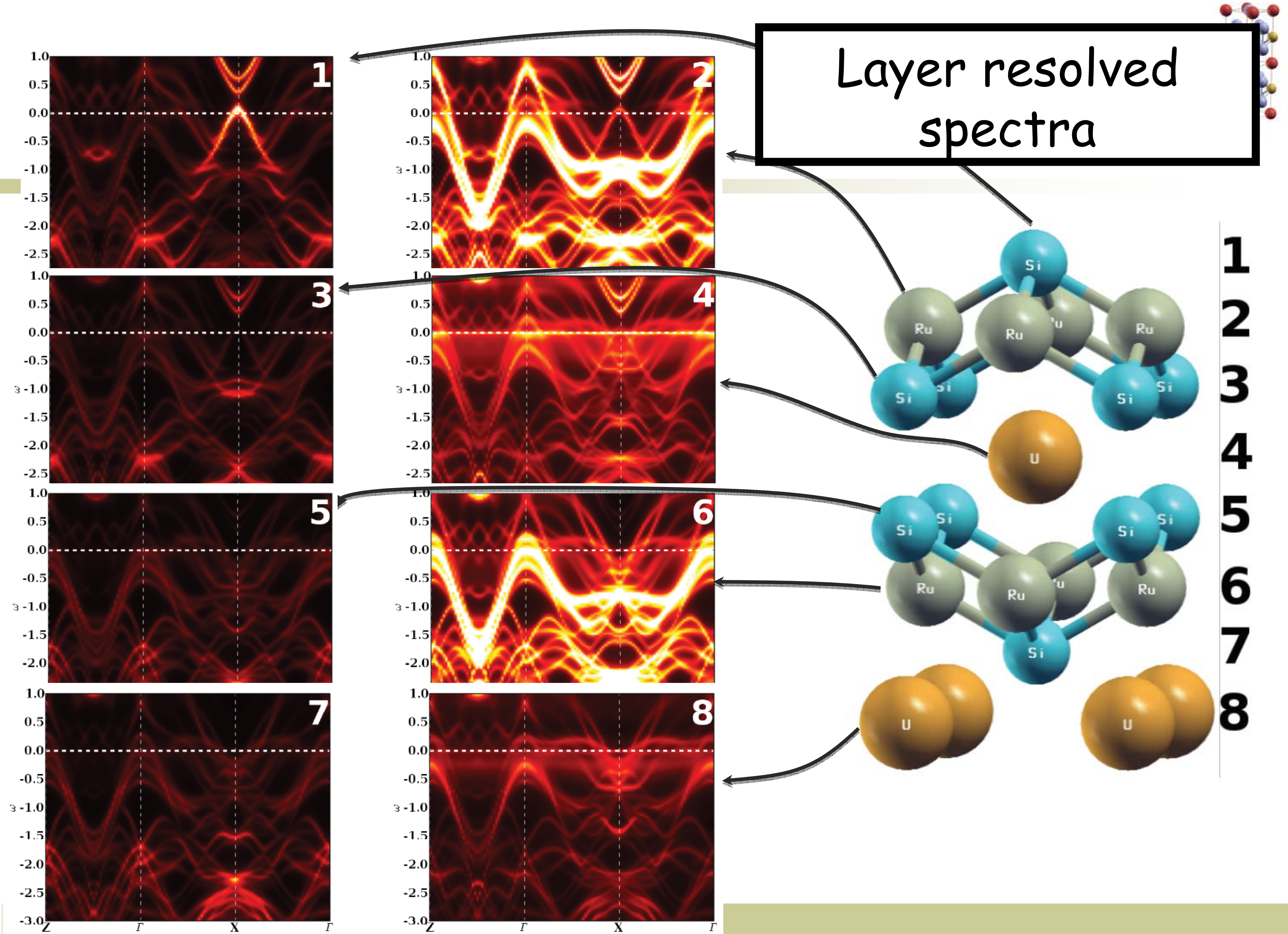
• No hole-pocket at the X-point.

LDA+DMFT - Si-terminated surface slab



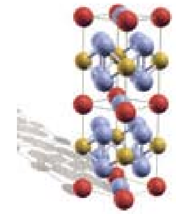


• Hole pocket surface state appears at X-point!

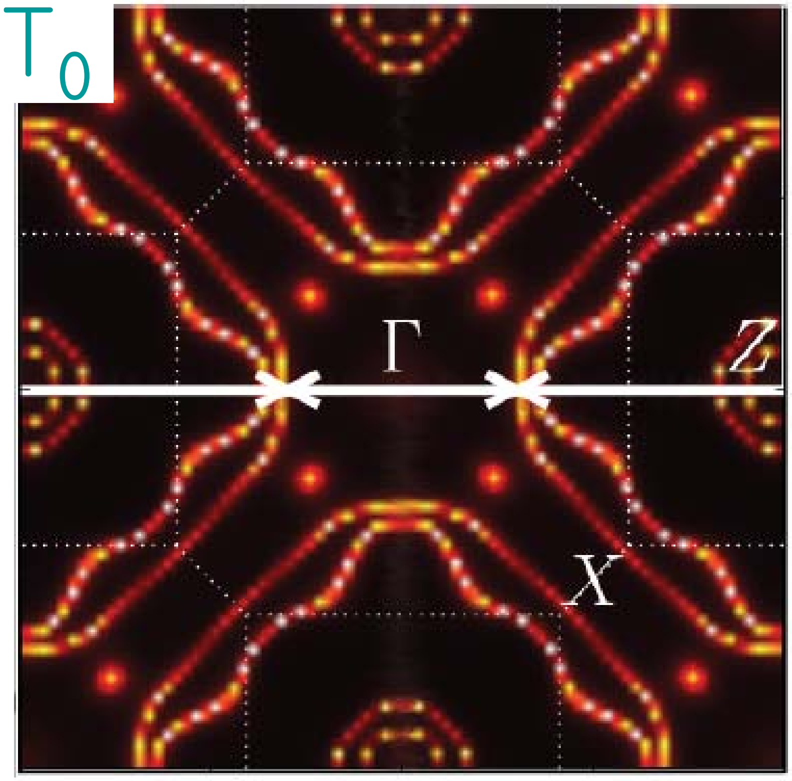


Layer resolved spectra
1
2
3
4
5
6
7
8
1.0
0.5
0.0
-0.5
-1.0
-1.5
-2.0
-2.5
-3.0
Z
Γ
X
Γ
Si
Ru
U

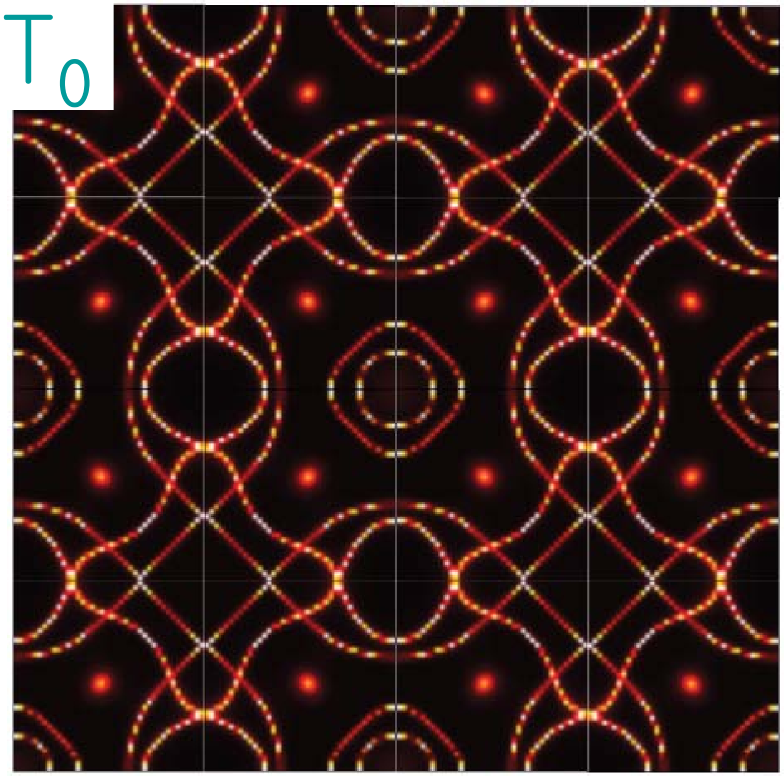
Fermi surface nesting, reconstruction below Tc



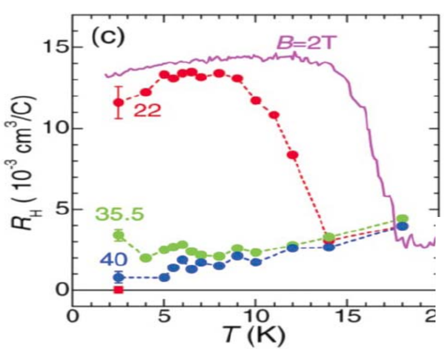
$T>T_0$



$T<T_0$



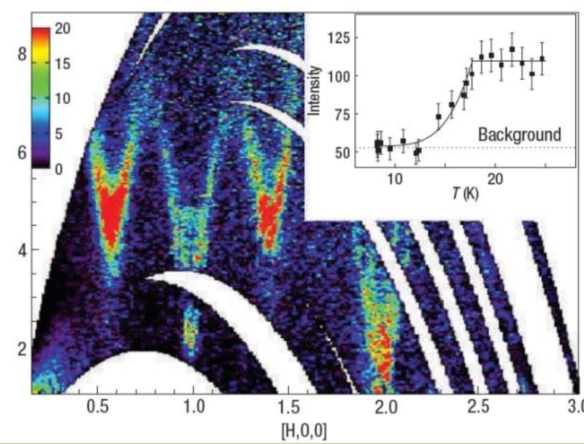
Nesting 0.6a* and 1.4a*



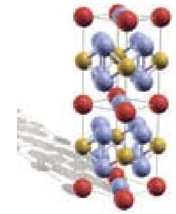
2 incommensurate peaks
(0.6,0,0), (1.4,0,0)

Fermi surface
reconstruction

Wiebe et.al. 2008



Conclusions



•DMFT tools can be used to understand (predict) properties of correlated mate rials

•Kondo effect in URu_2Si_2 is partially arrested bellow crystal field splitting energ y.

Gives room to ordered states, either AFM state or orbital order.

•AFM state and hidden order can be unified by a complex order parameter: “adiabatic continuity”

•Hidden order has hexadecapole character (does not break time reversal symmetry, nor C4 symmetr y)

•In the hidden order, fluctuations of the magnetic moment as a pseudo-Goldstone mode

•In AFM state there is a pseudo-Goldstone mode of hexa -decapole symmetry

Thank you!