Understanding Transition Temperature Differences in Heavy Fermion Superconductors

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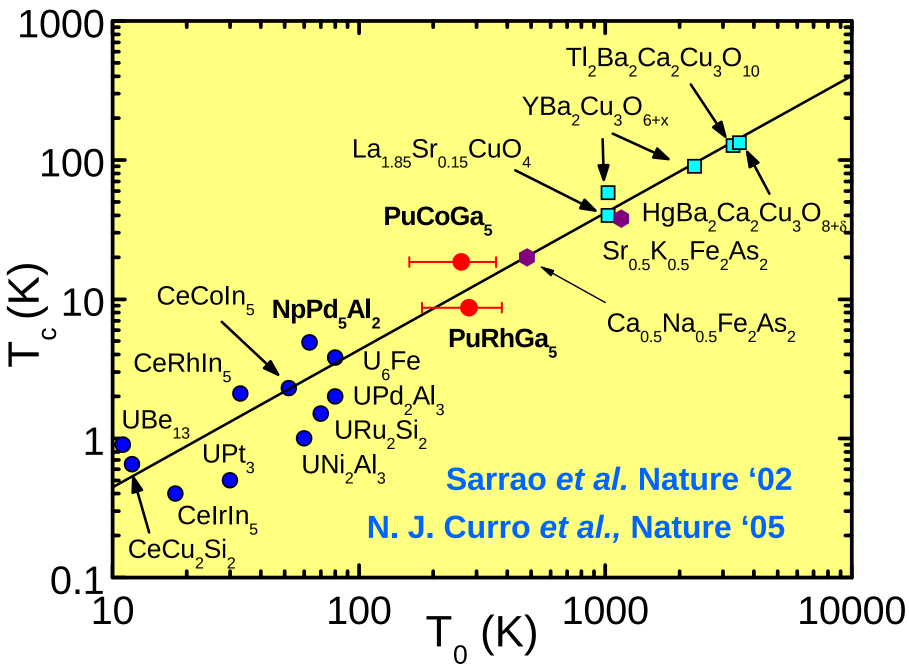
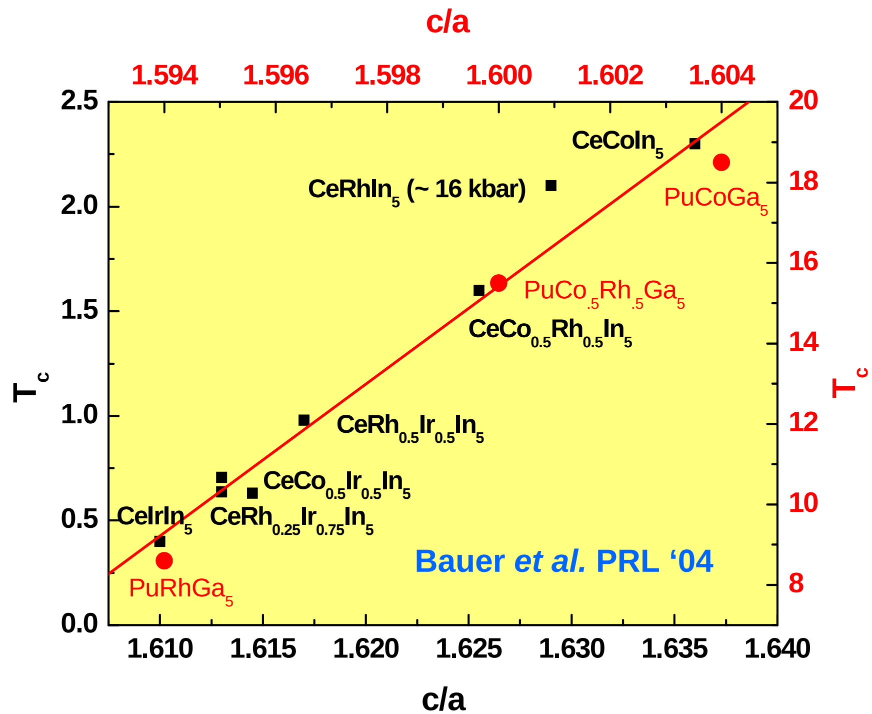
Objective: to develop, understand, and exploit new Matthias' Rules for unconventional superconductivity

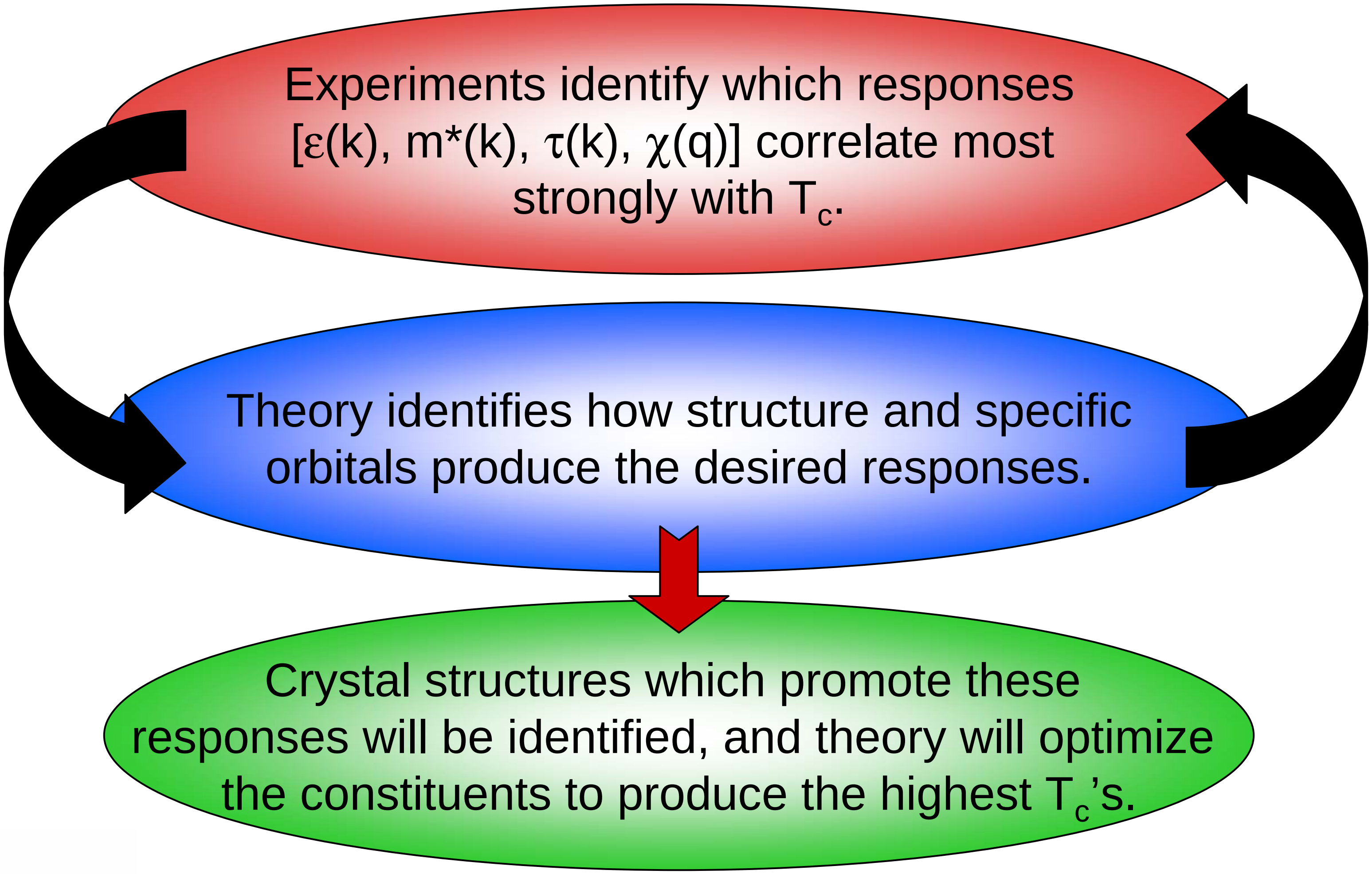




- "larger bandwidth => higher T_c's"
- "layered structures are good for SC"

But can we move beyond serendipitous discovery?

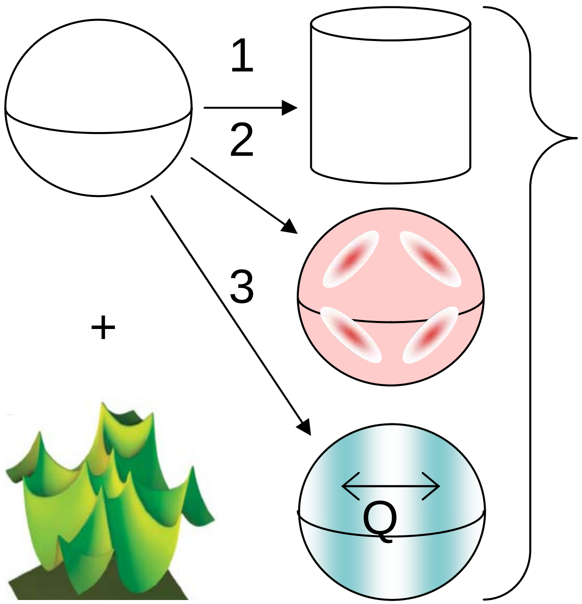
Design Approach

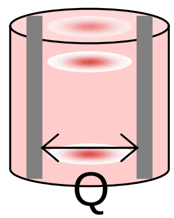


Experimental Goal: Identifying which properties correlate most with superconductivity.

Unconventional Superconductivity ⇨ Momentum space anisotropies are important.

Types of anisotropies:





ε(k): Fermi surface

m*(k): Effective mass

τ(k): Scattering rate

+

χ(q): Spin susceptibility

+

Δ(k): Superconducting gap

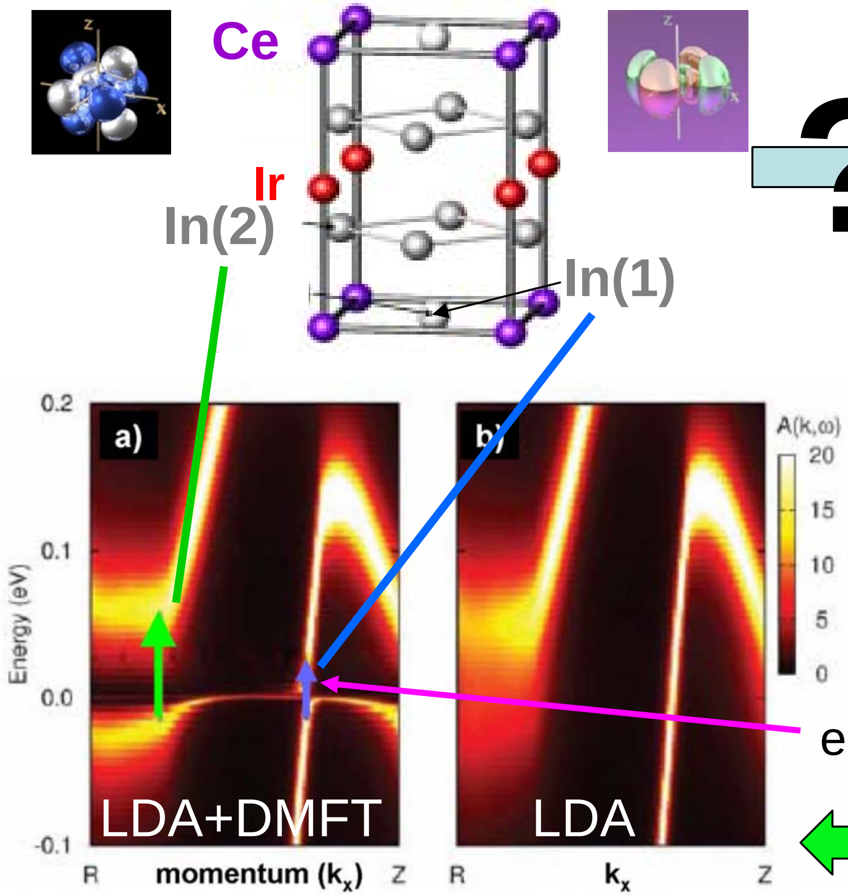
- Determining which anisotropies positively correlate with the superconducting energy scale will be achieved with the following
- Tuning Parameters:
  - Pressure
  - Magnetic field
  - Element substitution

Measurements we propose [anisotropies probed]:

ARPES [ε(k), m*(k), τ(k), Δ(k)]
NMR [χ(q) Δ(k)]
Point contact tunneling [Δ(k)]
Field-angle transport and thermodynamics [Δ(k)]

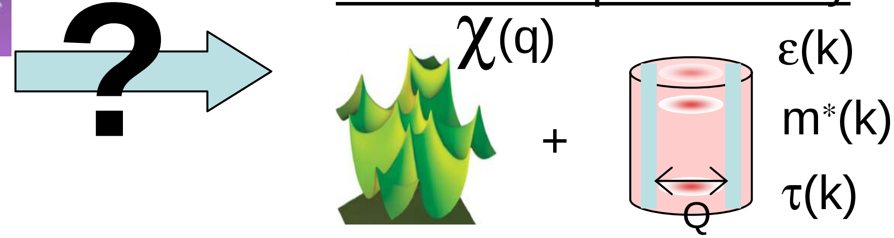
dHvA (SdH) (ε(k), m*(k), τ(k)]
Neutrons (selected) [χ(q), Δ(k)]

Theoretical Goal: Identify how crystal chemistry leads to the beneficial elements for superconductivity



Shim *et al.* Science '07

Important anisotropies identified experimentally:



How to identify the origin of anisotropies:

a) LDA

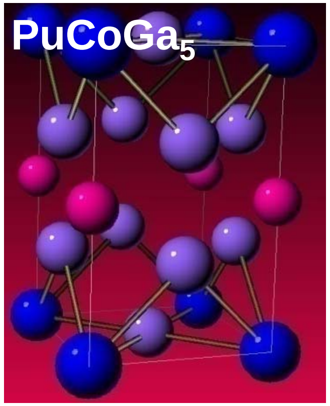
b) **DMFT** (strong on-site f correlations)

c) **Cluster DMFT** (include inter-site f-f correlations; Can also predict T_c)

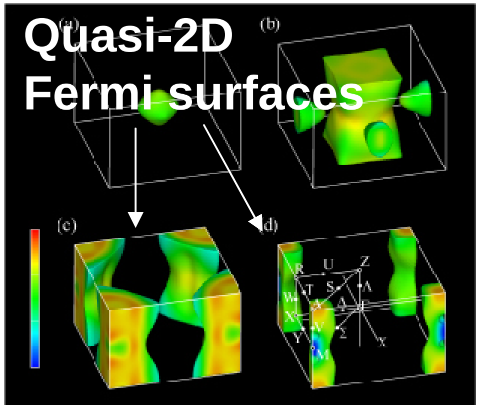
e.g. E scale is related to T_{sf}

DMFT example shows how In(1) and In(2) orbitals produce different gap magnitudes

Example of Possible Design Considerations

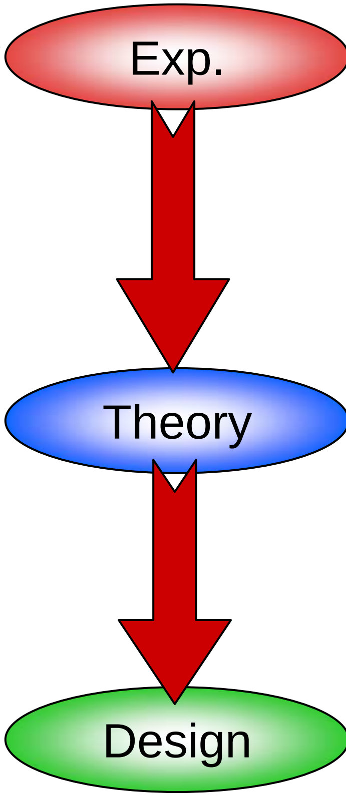


ARPES, NMR, Neutrons, Point contact tunneling, and field-angle transport and thermodynamics
[$\varepsilon(k)$, $m^*(k)$, $\tau(k)$, $\chi(q)$, $\Delta(k)$]

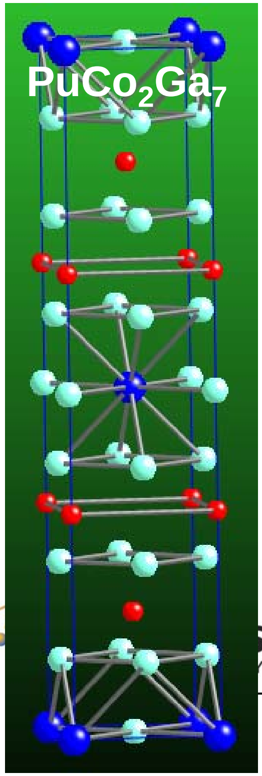


+ DMFT + CDMFT

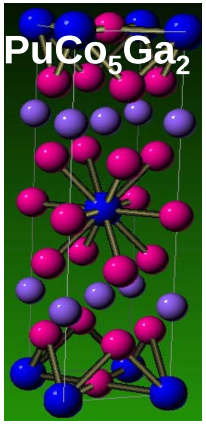
Maehira et al., PRL '03
Opahle and P. M. Oppeneer, PRB '03



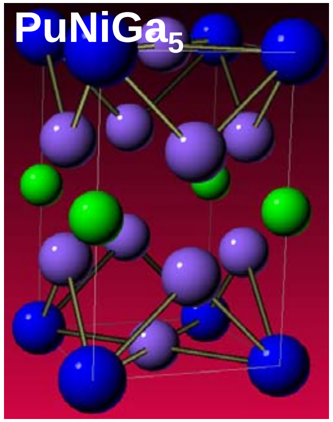
❶ More 2-D



❷ More spin anisotropy



❸ Larger Bandwidth



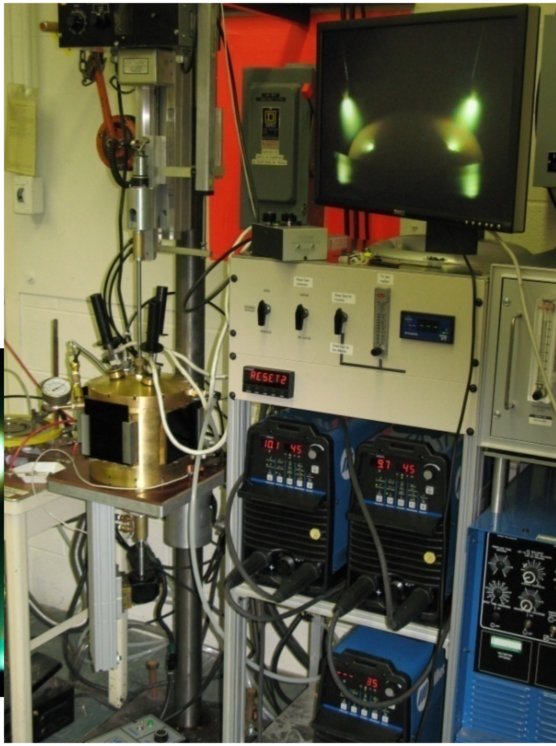
Many Possibilities!

A few highlights

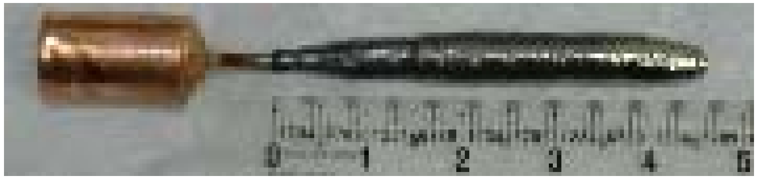
- High Purity Intermetallics
- Superconductivity in $CePt_2In_7$
- Preliminary results on various $PuCoGa_5$ measurements
- 2 impurity NRG-DMFT study
- Synthesis strategy and results

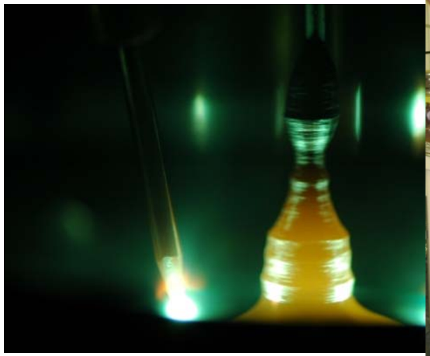
U-based Superconductor Crystal Growth

1) Electro-refined uranium (parts per BILLION impurity levels) [from AMES]

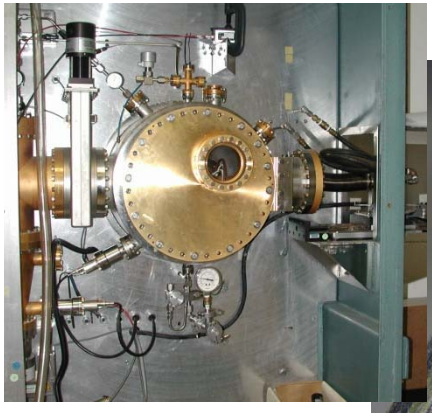


2) Czrochralski growth in new tri-arc furnace
   - Water-cooled tungsten seed rod and counter-rotating Cu hearth
   - Continuous Zr-gettered UHP Ar atmosphere (10^{-15} ppm O_2)





3) Zone-refinement of as-grown single crystals



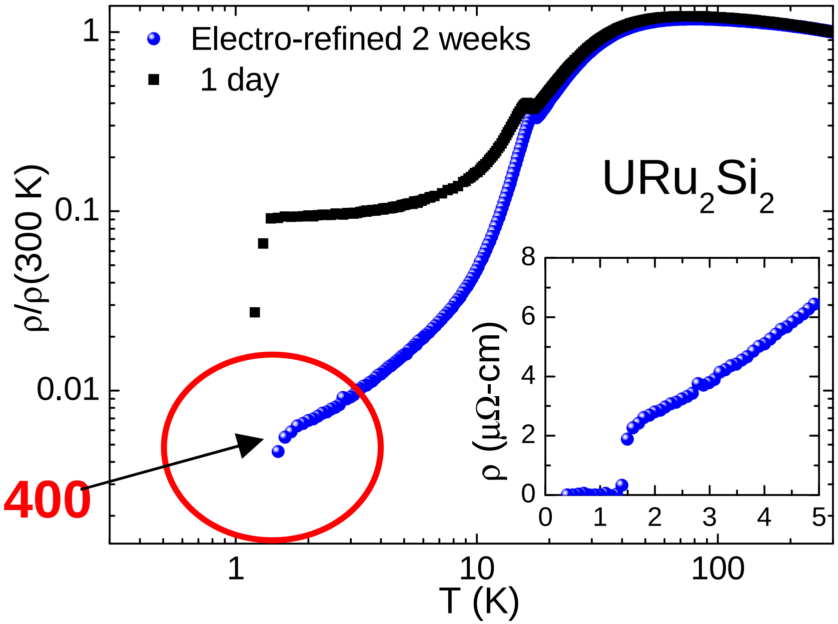
4) Electro-refinement of as-grown (or zone-refined) single crystals

Los Alamos
NATIONAL LABORATORY
EST. 1943
Operated by Los Alamos National Security, LLC for NNSA
NNSA

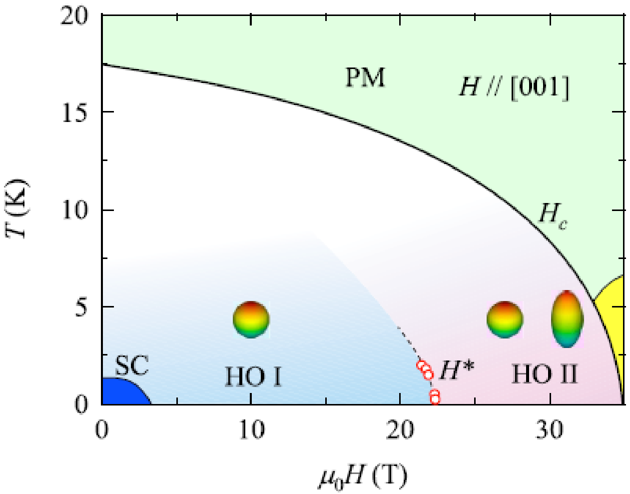
High Purity Crystals

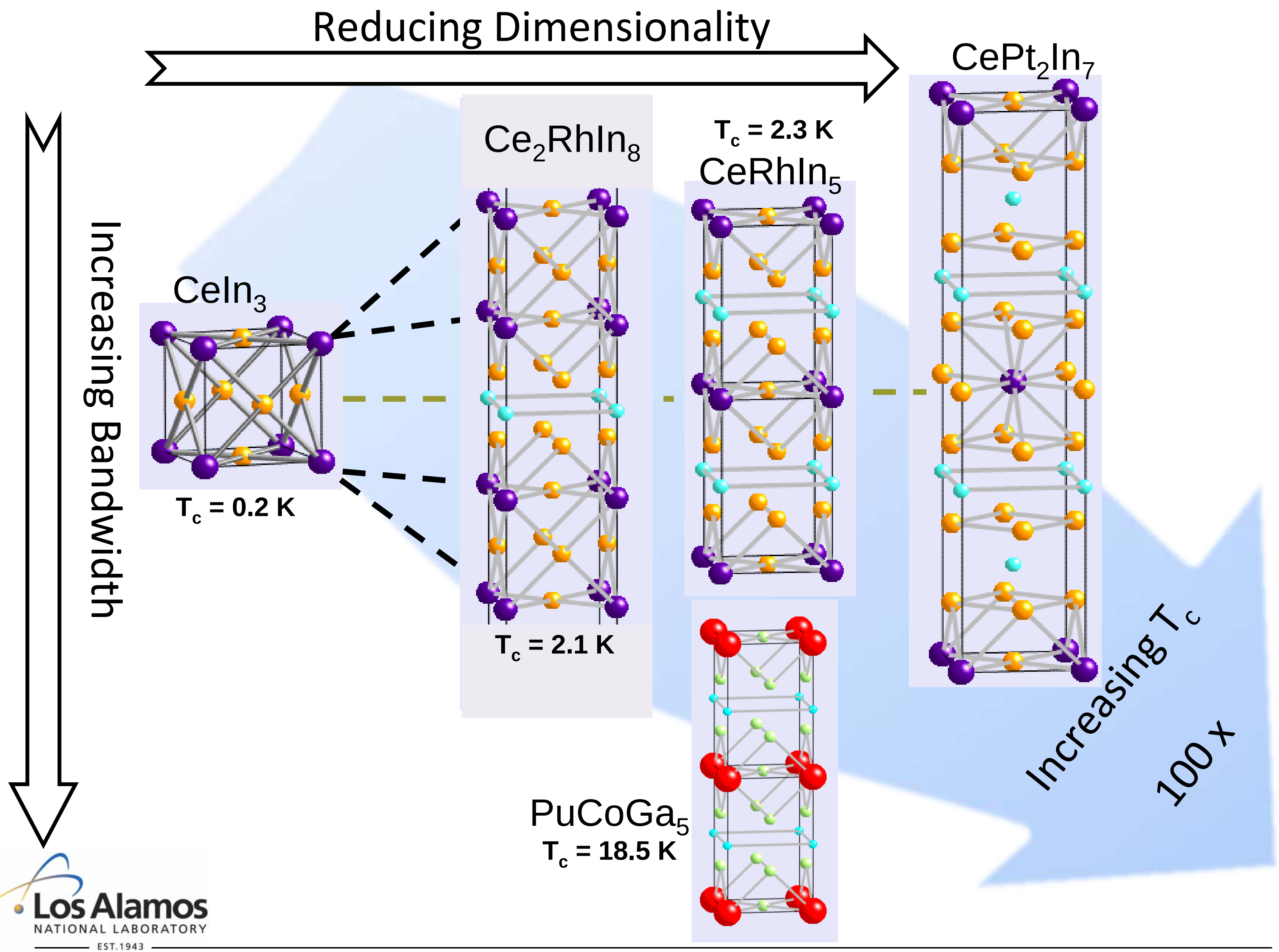
- Successful electro-refining at LANL of URu_2Si_2 with RRR = 400

- URu_2Si_2
- 1200 C
- 120 A!
- RRR = 400



- New high-field phase revealed in ultra-pure URu_2Si_2 single crystals (Shishido et al, PRL, 2009)




Reducing Dimensionality
Increasing Bandwidth
$CeIn_3$
T_c = 0.2 K
Ce_2RhIn_8
T_c = 2.1 K
T_c = 2.3 K
$CeRhIn_5$
$CePt_2In_7$
$PuCoGa_5$
T_c = 18.5 K
Increasing T_c
100 x

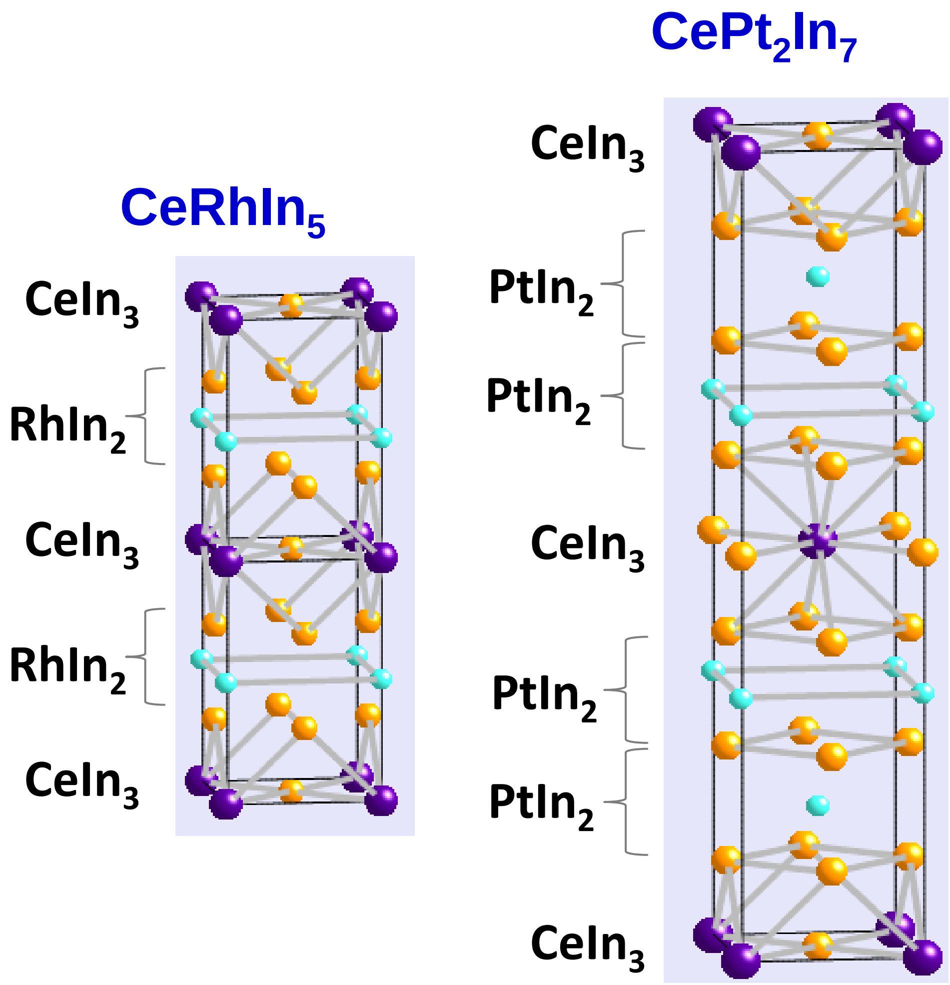
$CePt_2In_7$ Synthesis and Structure

- Arc-melted in ratio 1:2:7.5 on water-cooled Cu hearth w UHP Ar and Zr getter

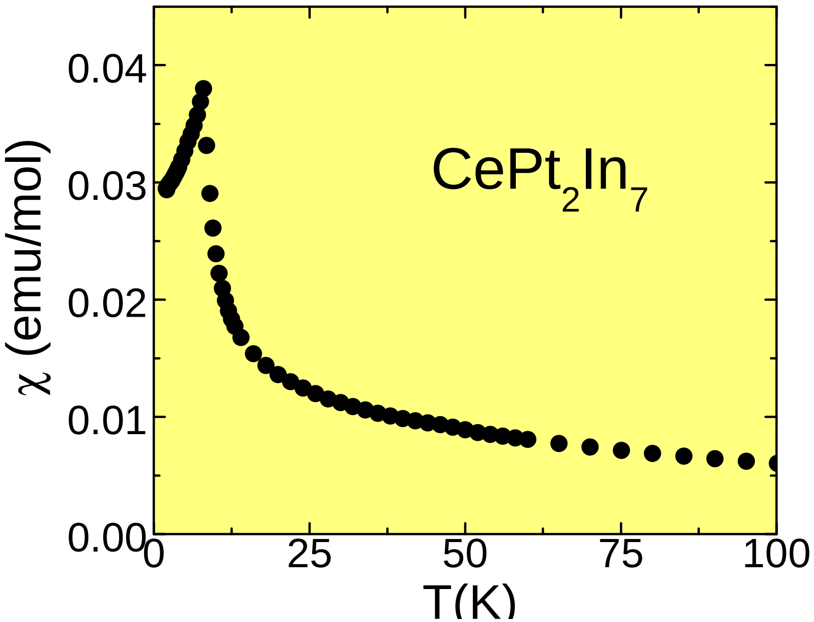
- Annealed 500 C for 2 wks

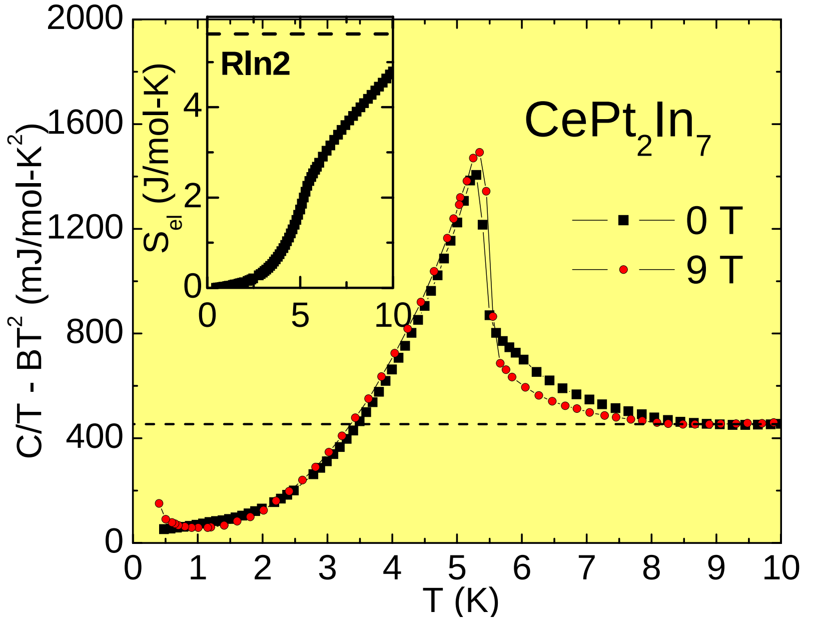
- Powder XRD reveals $CePt_2In_7$ crystallizes in I4/mmm (different from Ce115 with P4/mmm) with lattice parameters a = 4.611 Å and c = 21.647 Å (Kurenbaeva et al. Intermet. ’08)

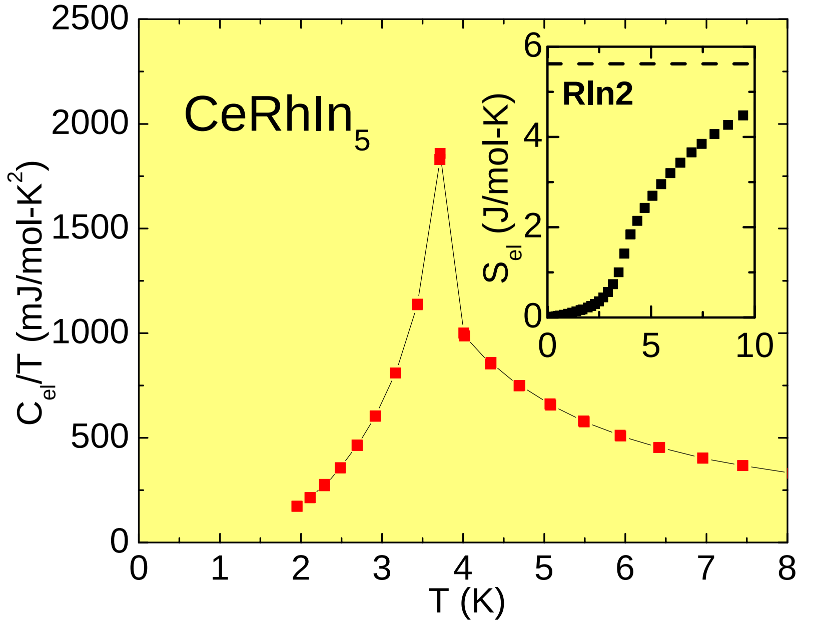
- Impurity phases (< 5%) $Ce_3Pt_4In_{13}$ and ?



$CePt_2In_7$ Properties

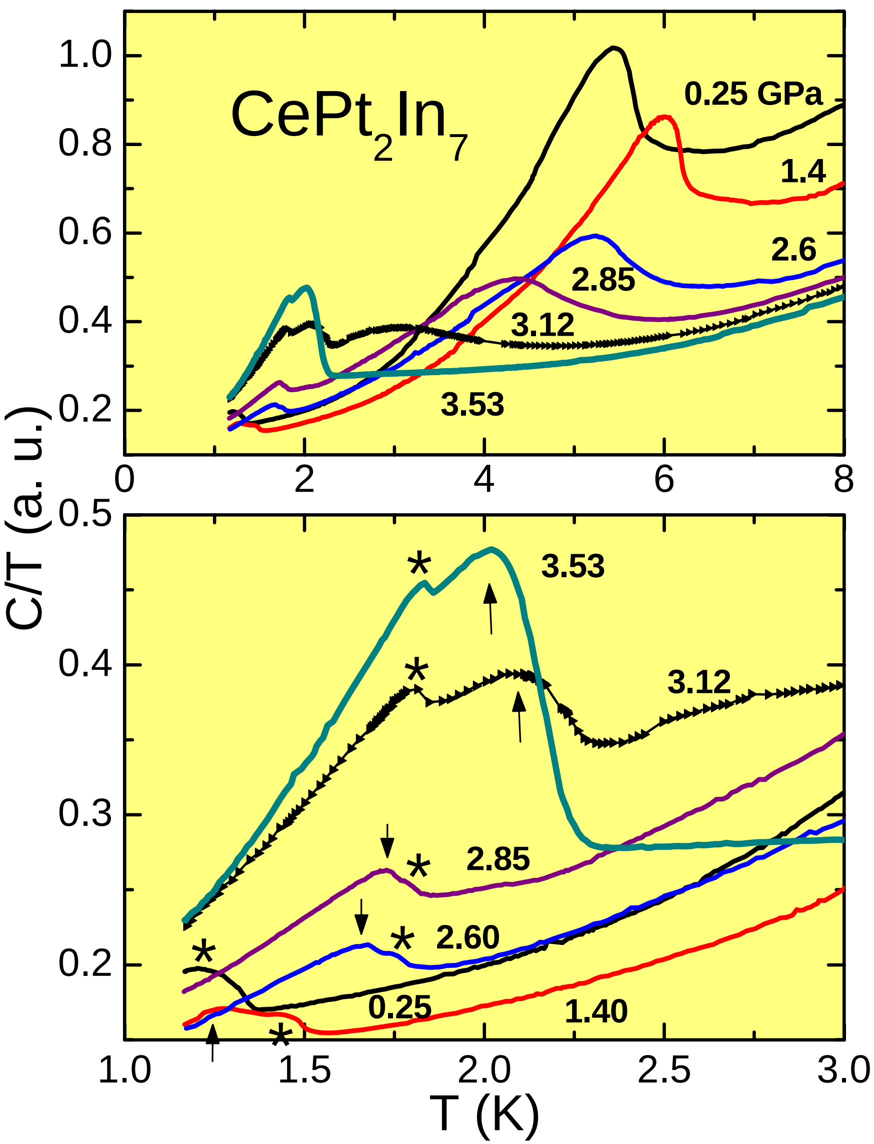




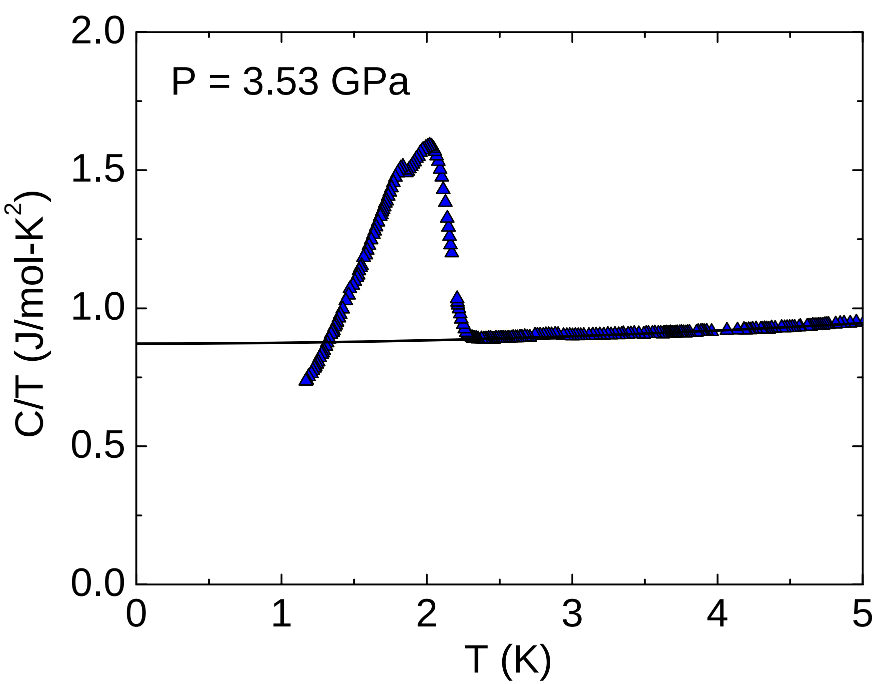


- **Anomaly at T_N = 5.5 K**
- **Moderately enhanced electronic specific heat coefficient γ ~ 50- 450 mJ/mol K²**
- **Similarity to $CeRhIn_5$:**
  - **Shape of transition in Cp**
  - **Reduced entropy at transition [1/3*Rln(2)]**
  - **Minimal magnetic field dependence**

$CePt_2In_7$: AC Calorimetry Under Pressure

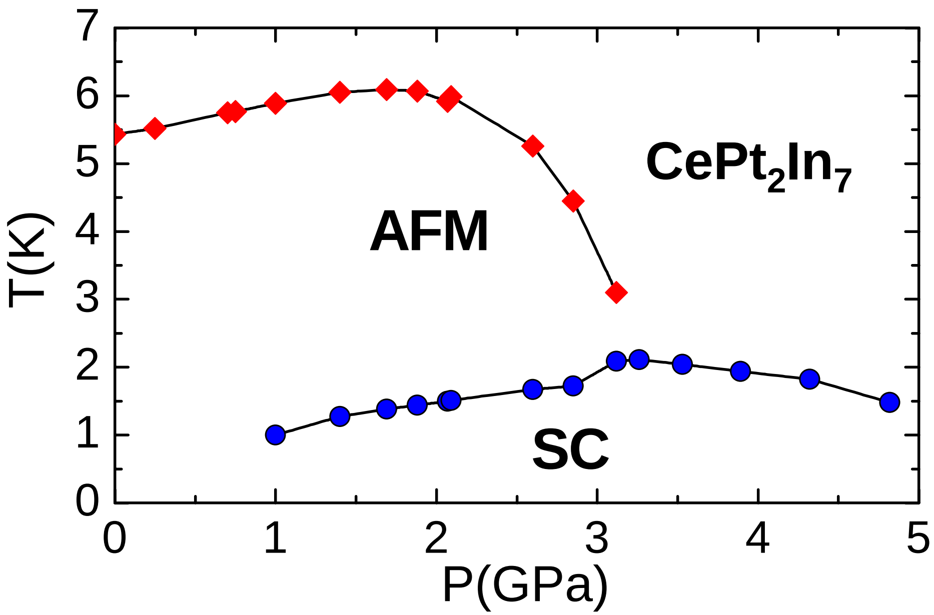


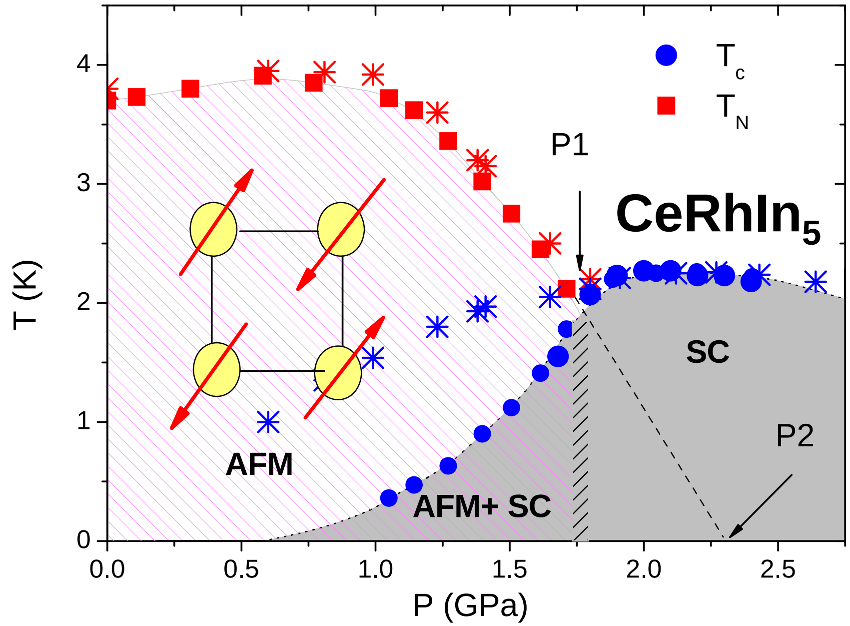
- AC calorimetry sample has < 2%
- $Ce_3Pt_4In_{13}$ (*) and an unknown impurity
- Large C/T value of SC transition at P > 2.85 GPa



$CePt_2In_7$ Phase Diagram

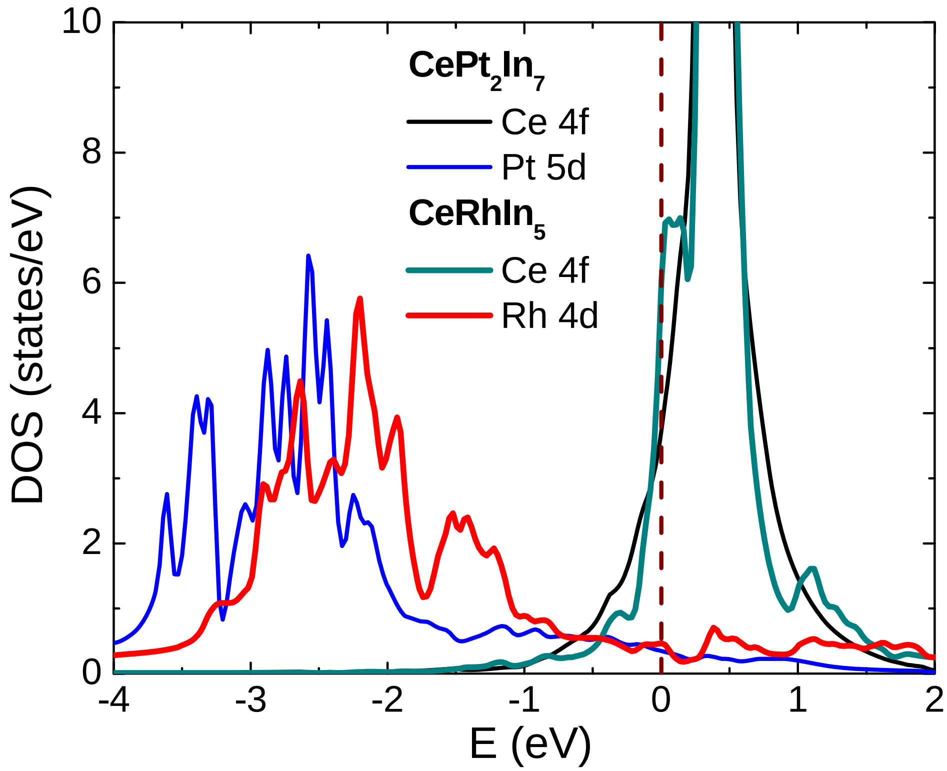
- **Similarity to $CeRhIn_5$:**
  - **Large coexistence region**
  - **C_p jump small in coexistence region, becomes large in pure SC state**





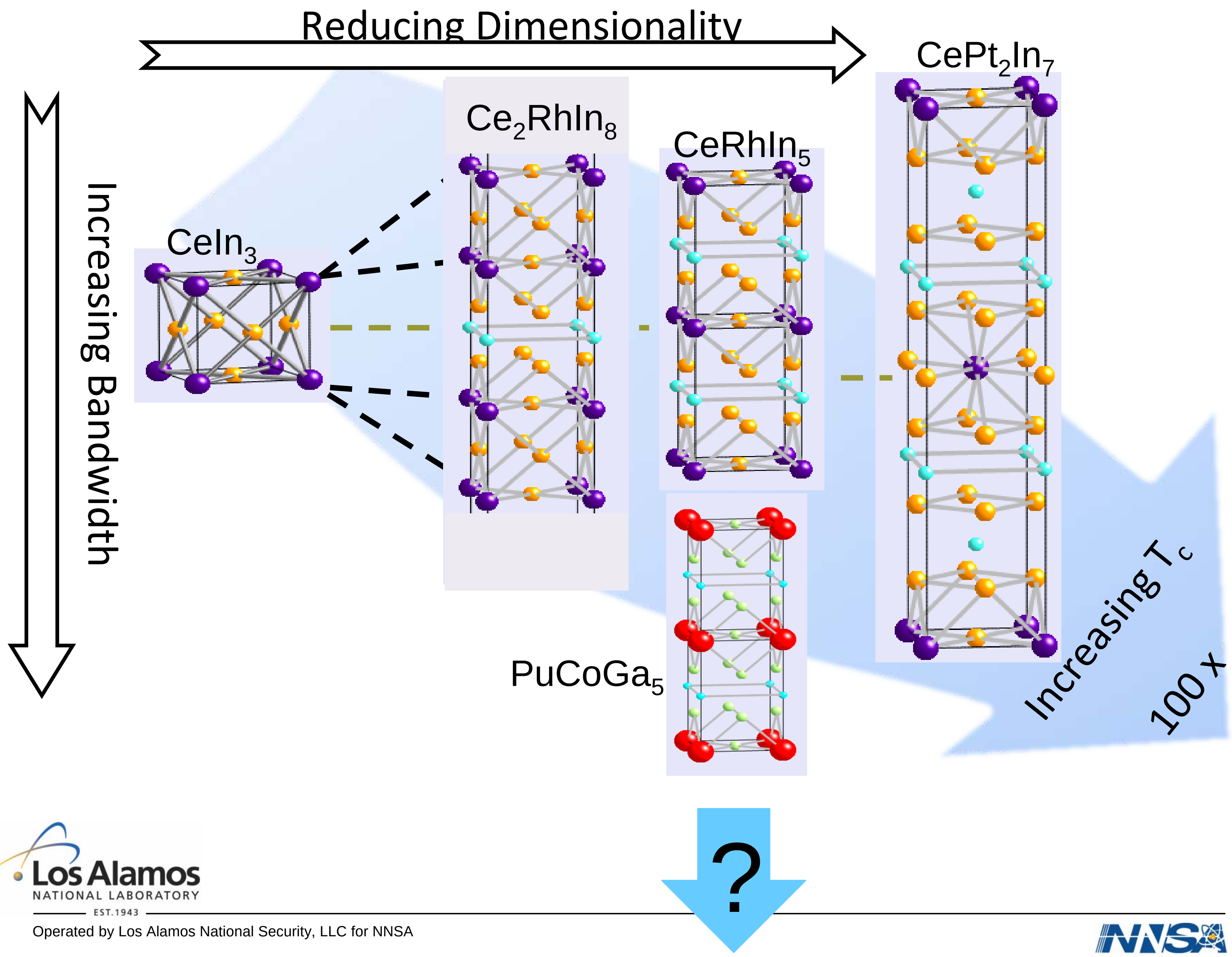
T_c^{max} ($CePt_2In_7$) = 2.1 K < T_c^{max} ($CeRhIn_5$)

$CePt_2In_7$

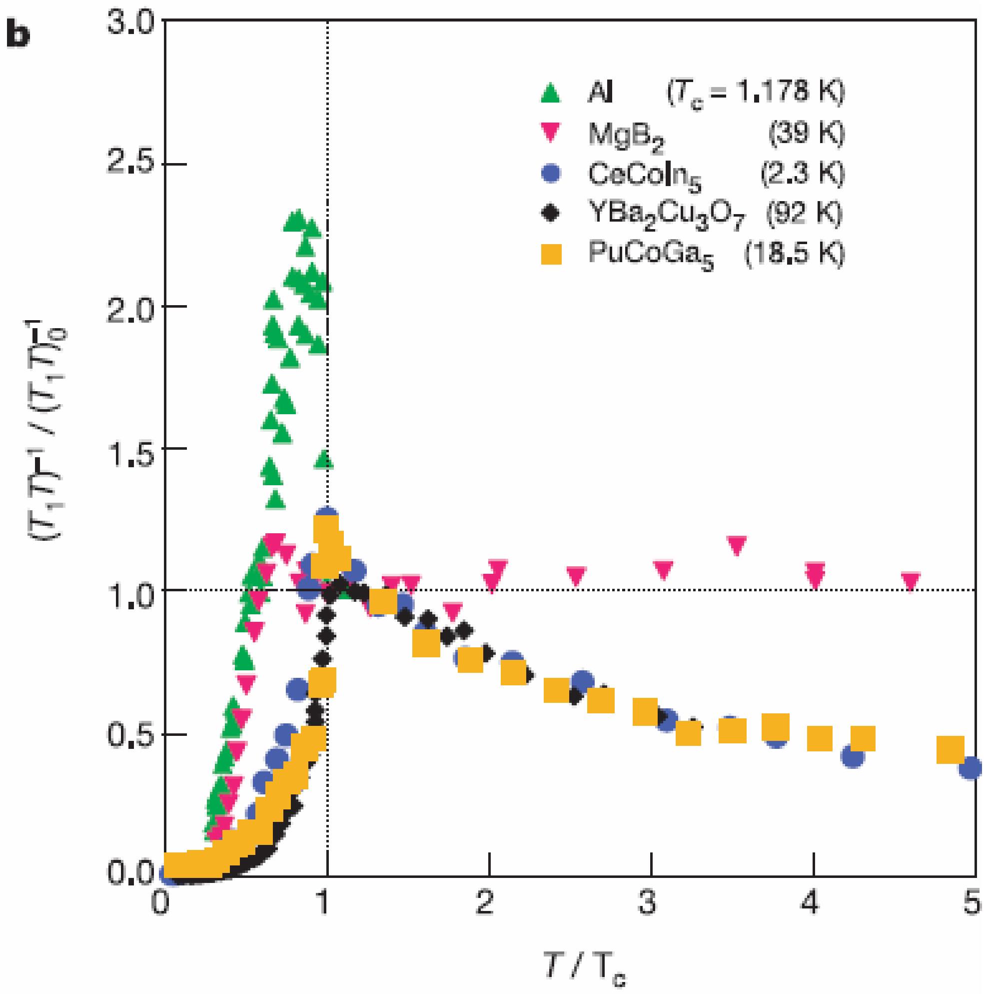


T_c (Ce_2PdIn_8) = 0.7 K versus T_c(Ce_2RhIn_8) = 2.1 K

Reduced density of states in Ni column compared to the Co column?


Reducing Dimensionality
Increasing Bandwidth
CeIn3
Ce2RhIn8
CeRhIn5
CePt2In7
PuCoGa5
Increasing Tc
100 x
?

Are $PuCoGa_5$ and $CeCoIn_5$ related?



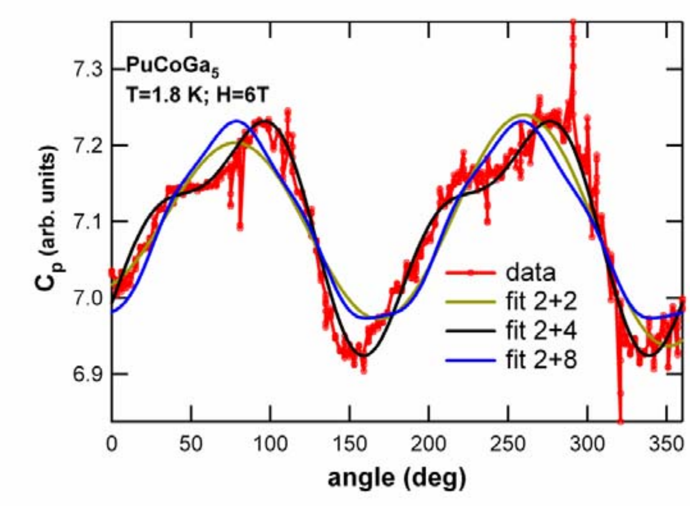
Is the Gap symmetry of Ce115's and Pu115's the same?

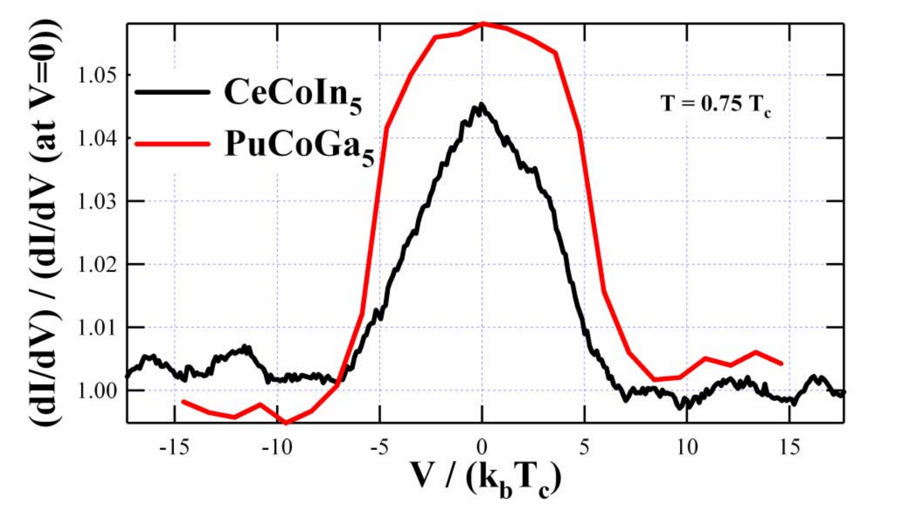
$1/T_1$ measurements indicate that $CeCoIn_5$ and $PuCoGa_5$ have nodes.

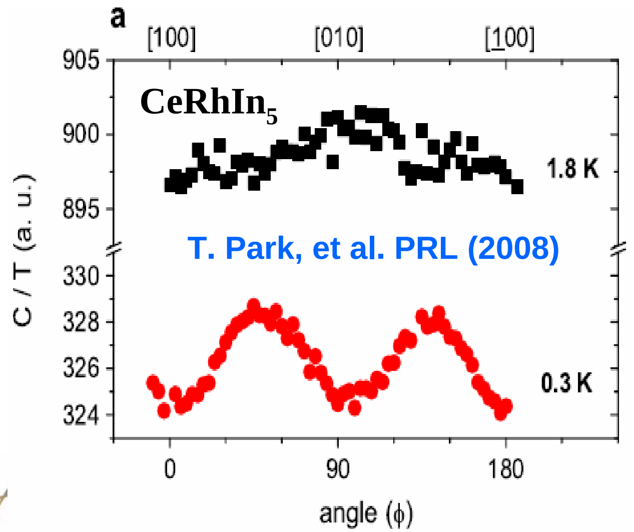
N.J. Curro, *et al*. Nature (2005)

Superconducting Gap Symmetry

Indication of 4-fold symmetry

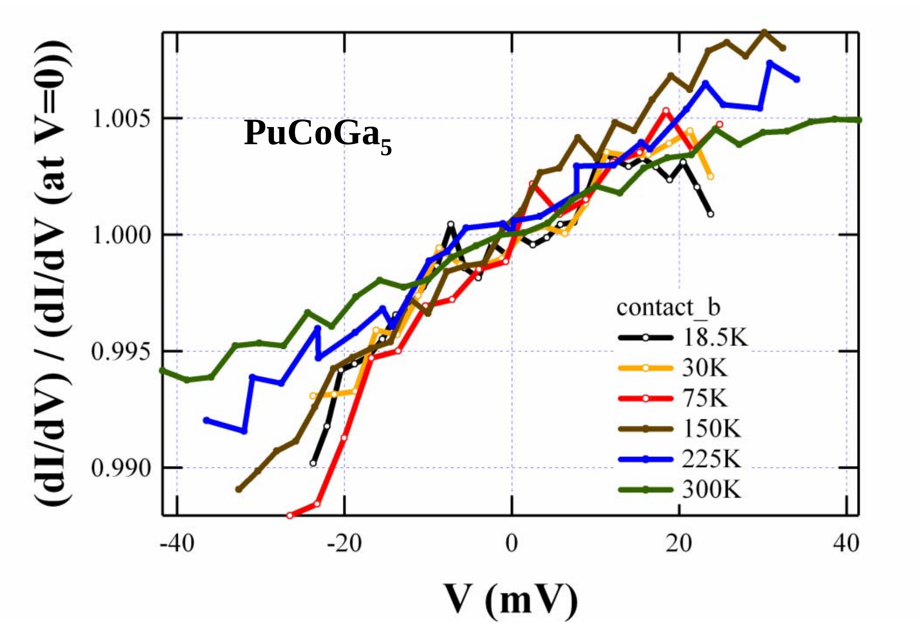


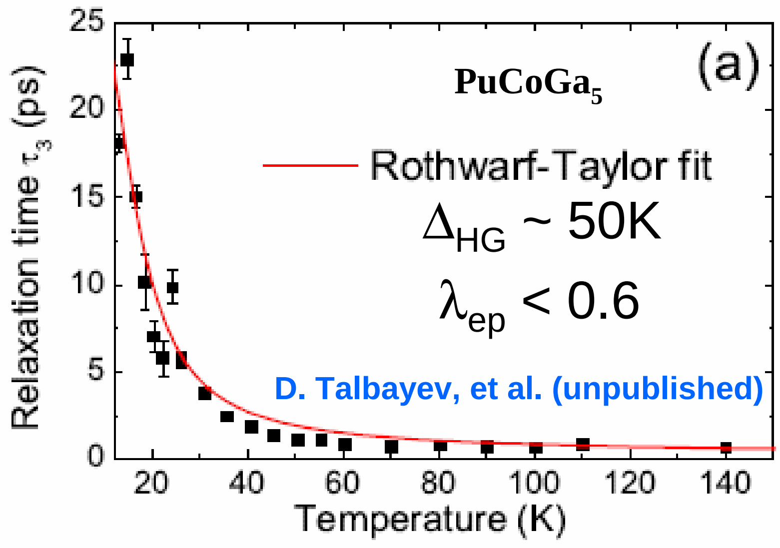




T. Park, et al. PRL (2008)

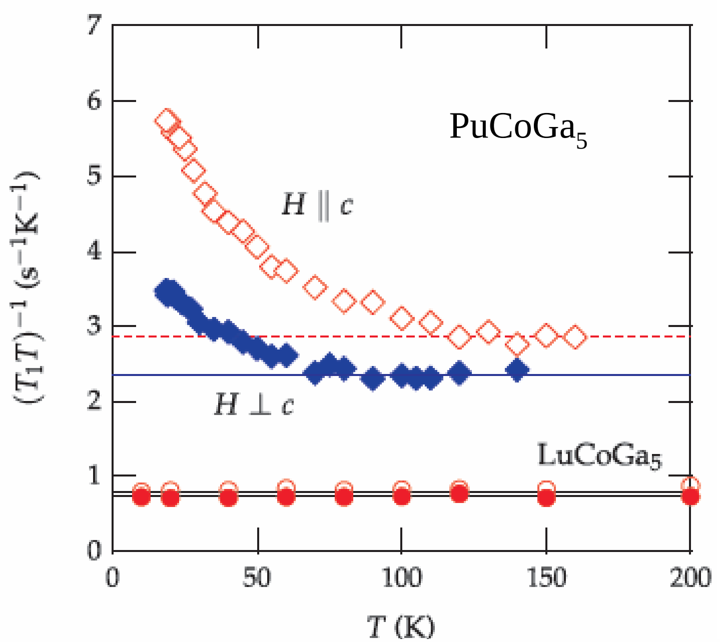
Electronic Structure





+ ARPES of Pu115 coming soon

Anisotropy of spin fluctuations



$$\left(\frac{1}{T_1}\right)_{\parallel} = \frac{2\gamma_n^2 k_B T}{\mu_B^2} \sum_{\boldsymbol{q}} A_{\perp}^2(\boldsymbol{q}) \frac{\mathrm{Im}\chi(\boldsymbol{q},\omega_0)}{\omega_0}$$

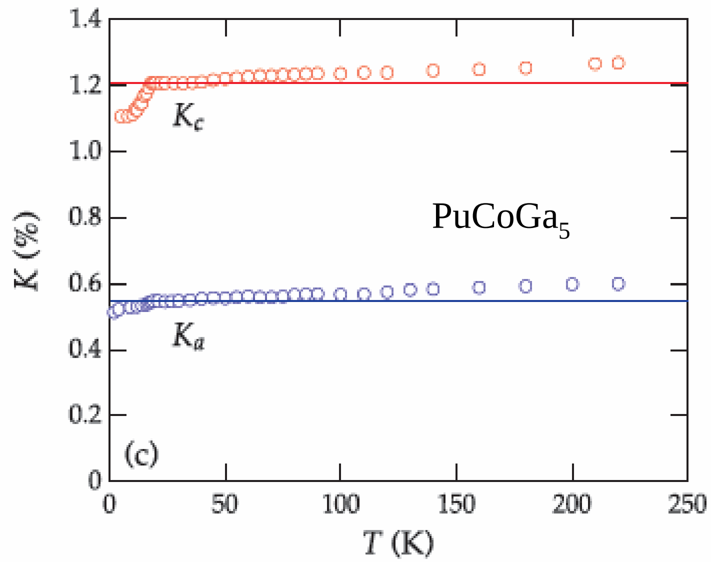
$$\simeq \frac{2\gamma_n^2 k_B T}{\mu_B^2} \sum_{\boldsymbol{q}} f^2(\boldsymbol{q}) A_{\perp}^2(0) \frac{\mathrm{Im}\chi(\boldsymbol{q},\omega_0)}{\omega_0}$$

$$\simeq \frac{\gamma_n^2 A_{\perp}^2(0)}{8\pi\Gamma_{\perp}^2}$$

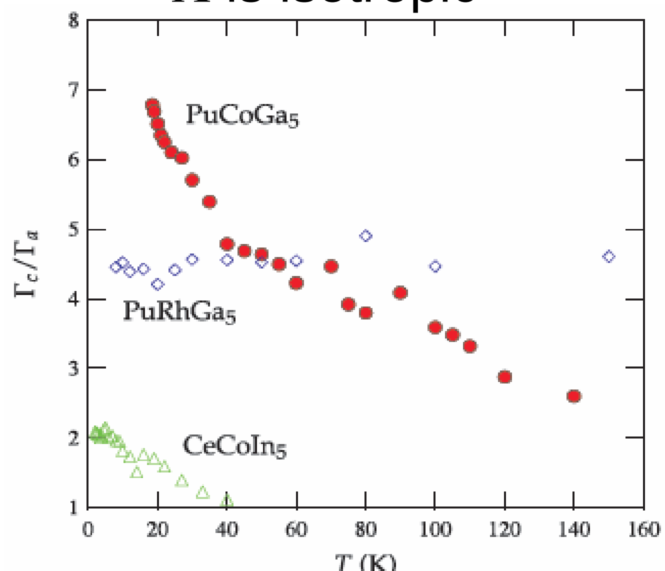
← From *K*-*X* plot

— S.F. energy scale

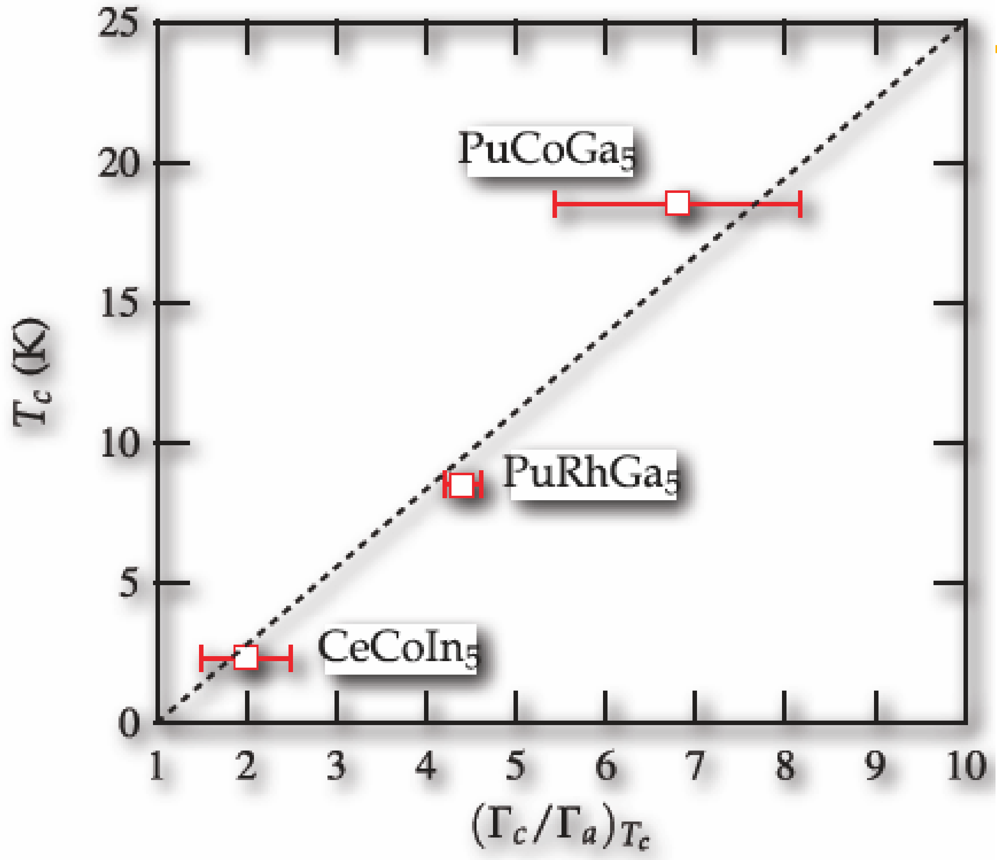
S. Kambe, *et al.* PRB (2007)



X is isotropic



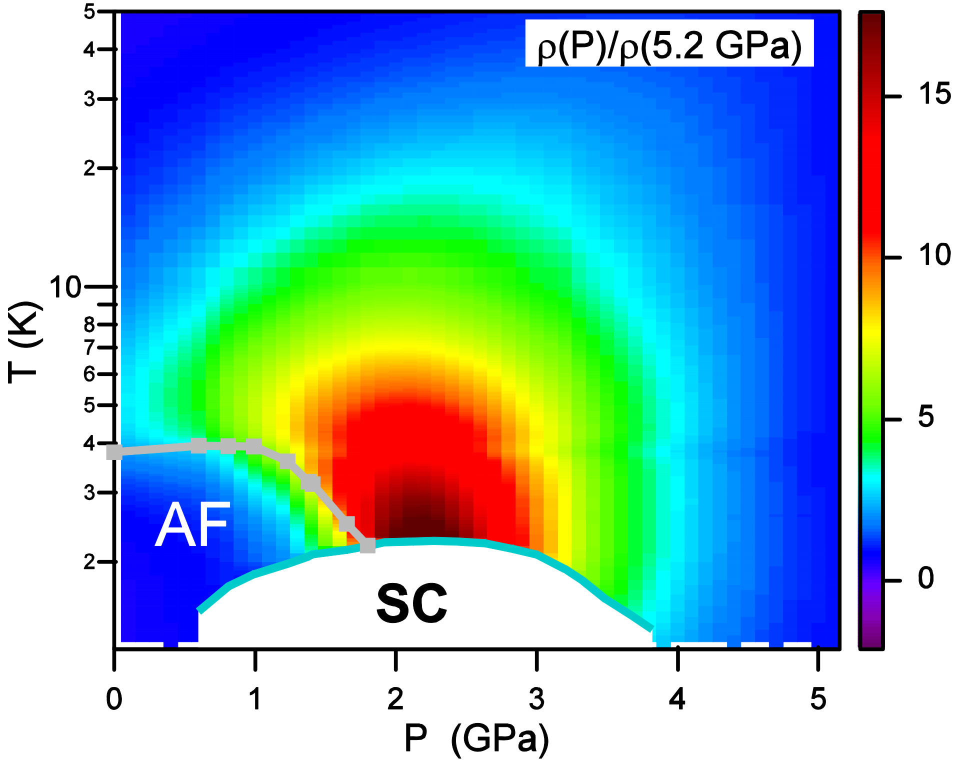
H. Sakai and S-H. Baek (unpublished)



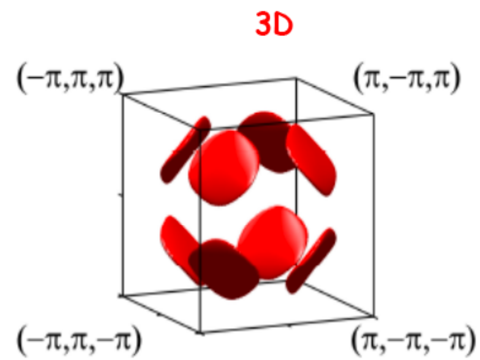
Implication is that stronger XY-type fluctuations are better for superconductivity in the 115 family

H. Sakai and S-H. Baek (unpublished)

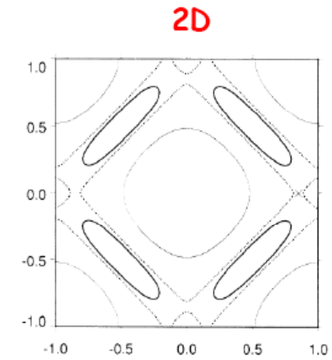
Influences of f-electron delocalization



$CeIn_3$



$$q = \left(\frac{\pi}{2}, \frac{\pi}{2}, \frac{\pi}{2}\right)$$



$$q = \left(\frac{\pi}{2}, \frac{\pi}{2}\right)$$

- “local” fluctuations generate superconductivity.

T. Park, *et al.* Nature (2008)

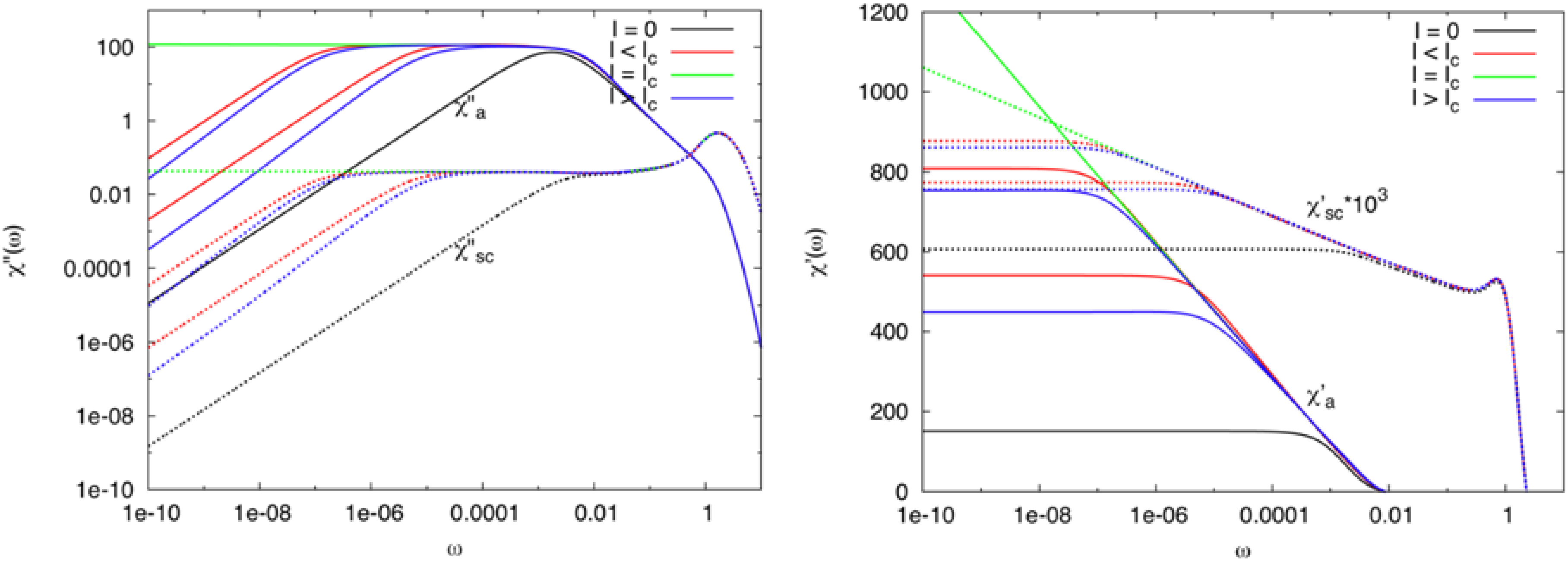
- partial f-electron delocalization creates heavy hole pockets reminiscent of the cuprates.

S. Sebastian, *et al*. PNAS (2009)

Role of intersite correlations?

- The **k**-dependence of Σ(**k**,ω) is important for unconventional superconductivity;
- Relevant energy scales in *f*-electron systems are smaller than in *d*-electron counterparts and low-energy approach should be explored.

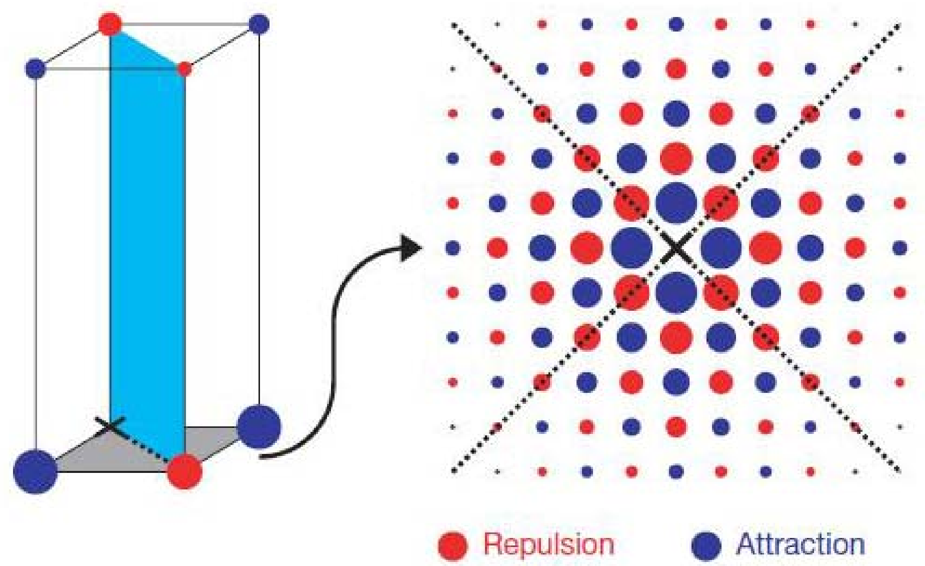
NRG results on minimal 2-impurity Anderson-Heisenberg model



- **Superconductivity is tied to the antiferromagnetic spin fluctuation and most favorable near the quantum criticality.**
- Cluster DMFT calculations in progress.

Lijun Zhu and Jian-Xin Zhu (unpublished)

Synthesis Strategies



Pines, *et al*. Nature (2008)

Important Points

(1) Antiferromagnetism

(2) Low ordered moments (itineracy)

(3) Tetragonal systems

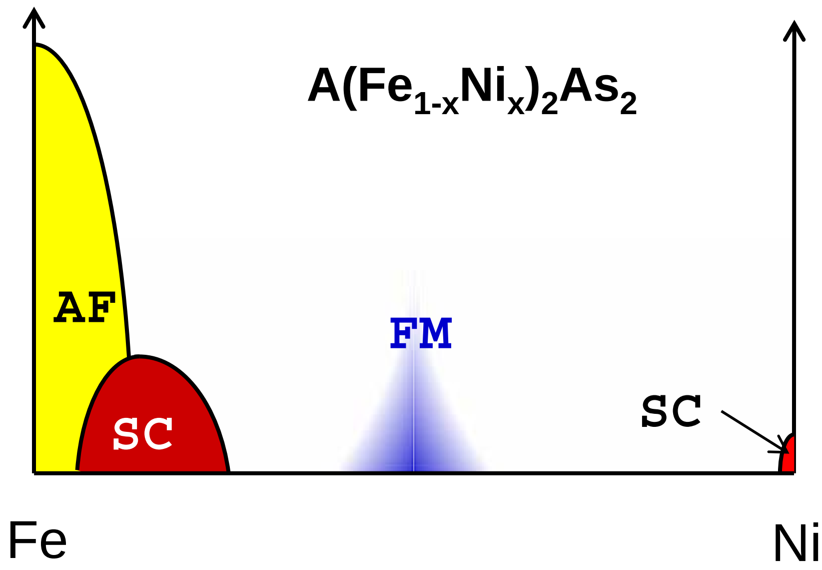
(4) Layered systems

(5) Large breadth of isostructural analogues (doping studies)

(6) Exploit common building blocks

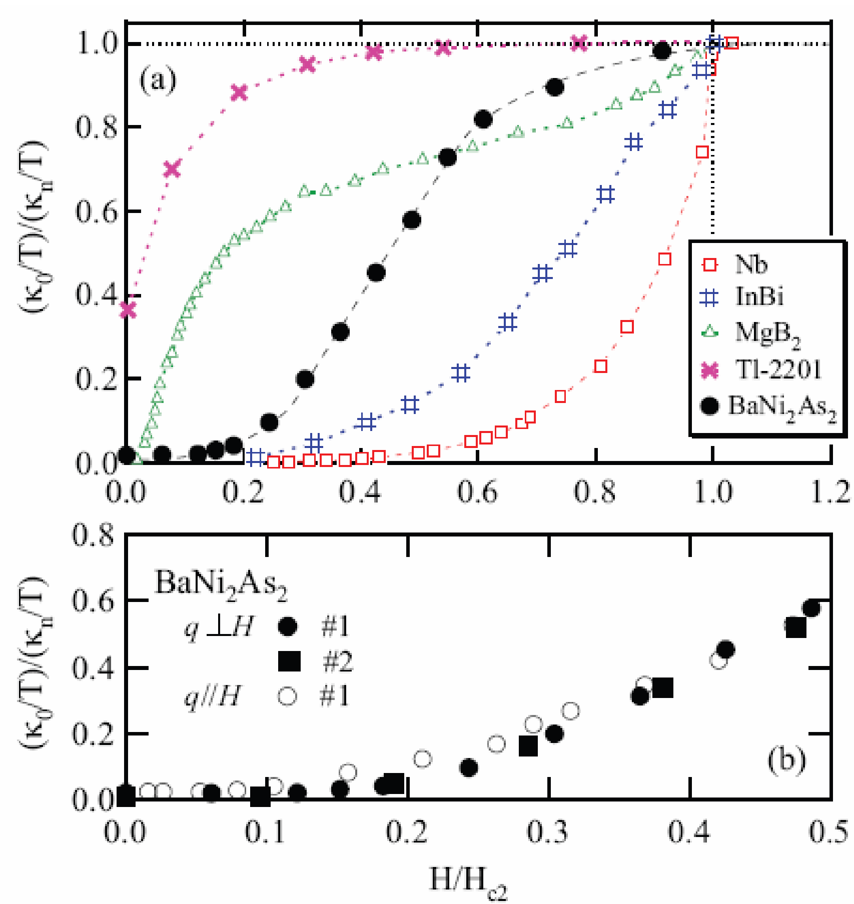
(7) Use Phase Stability Analysis

Ni-based pnictides



No evidence for magnetism or strong correlations in the Ni-based pnictides

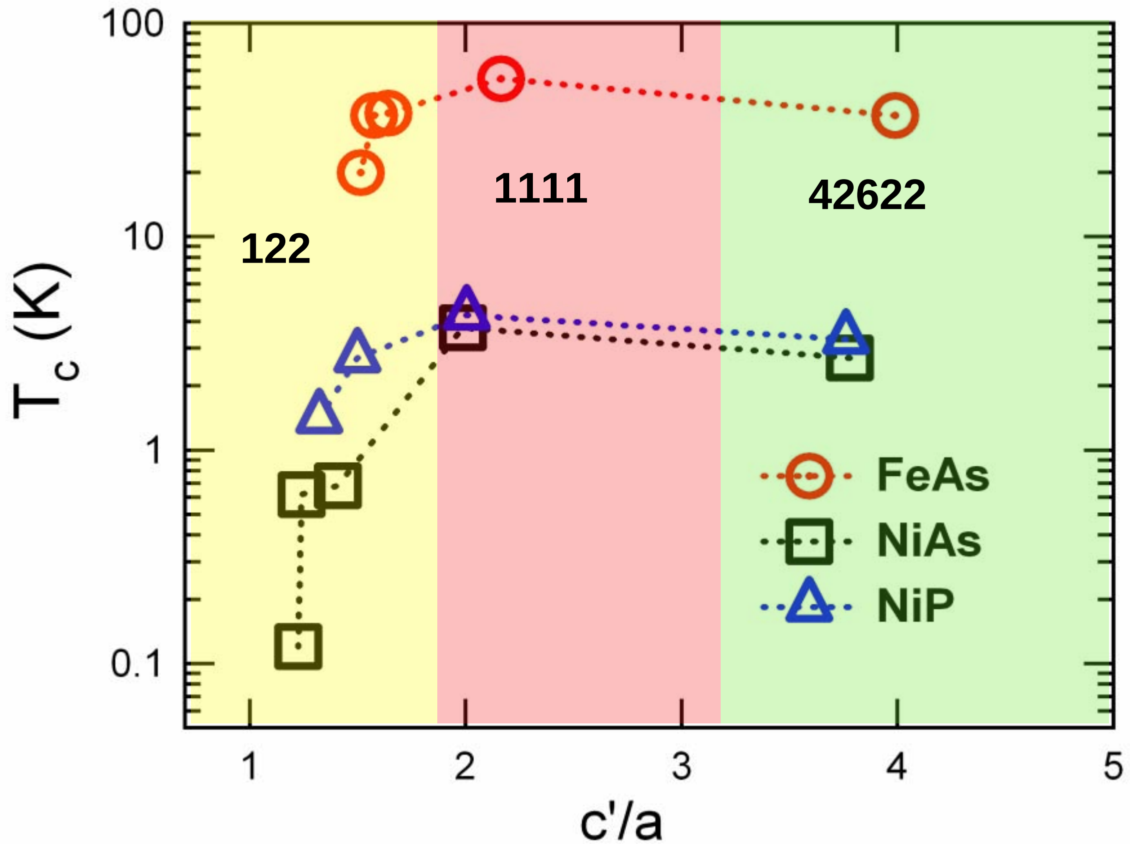
F. Ronning, *et al*. Physica C (2009)



Fully gapped superconductivity in Ni-based pnictides

N. Kurita, *et al*. PRL (2009)

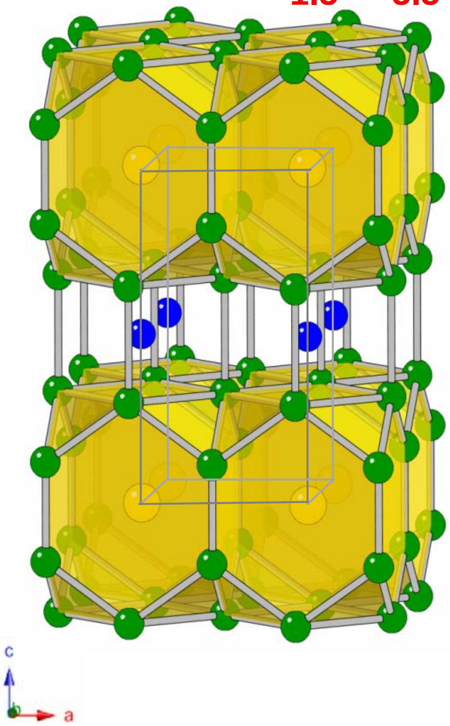
Implication for Fe-based pnictides?

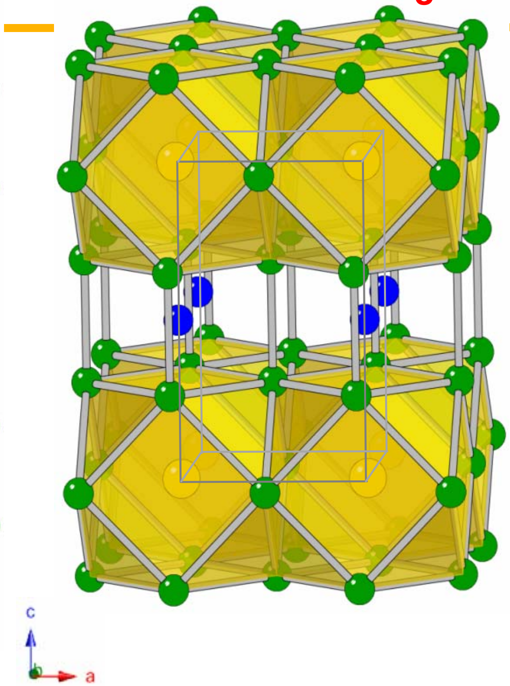


Evidence for electron-phonon pairing boosted by magnetism/strong electronic correlations?

Common building blocks

$CePd_{1.5}Al_{5.5}$ $CeRhIn_5$

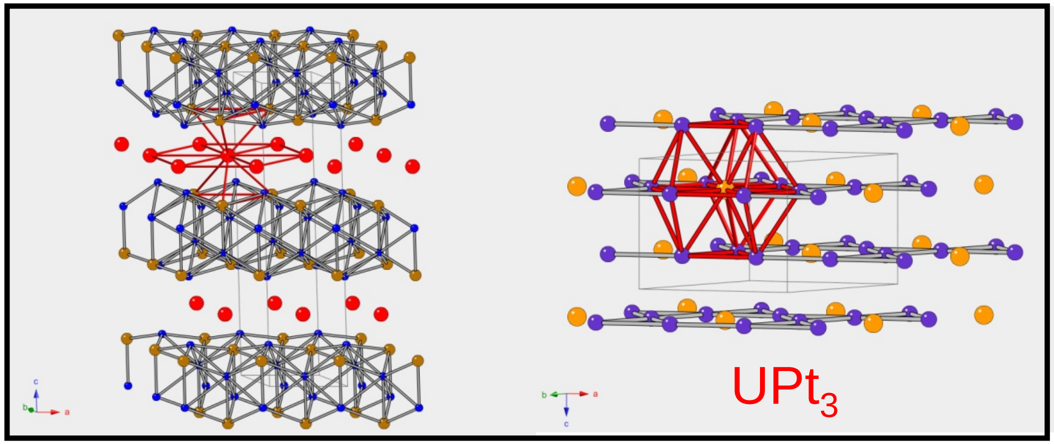




$CePd_{1.5}Al_{5.5}$: layered tetragonal system. Local moment ferromagnet

P.H. Tobash, *et al.* J. Solid State Chem. (in press)

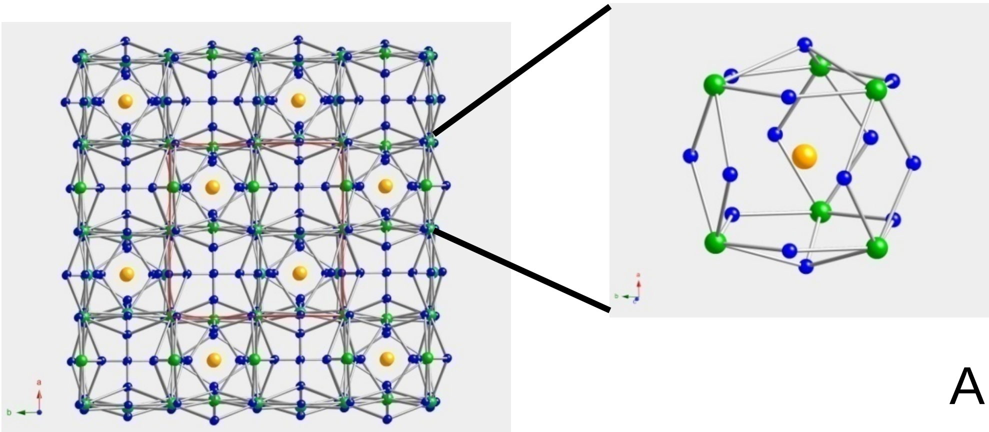
Similar U-coordination environments



$U_2Pt_6Al_{15}$: ~2/3 occupancy on U site. No SC down to 0.4K

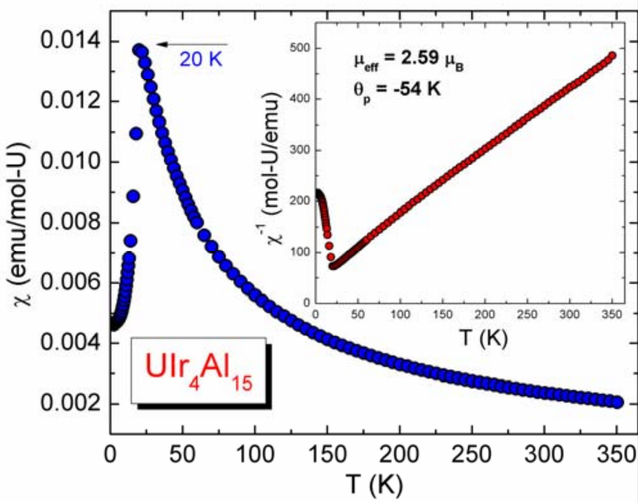
P.H. Tobash, *et al.* unpublished

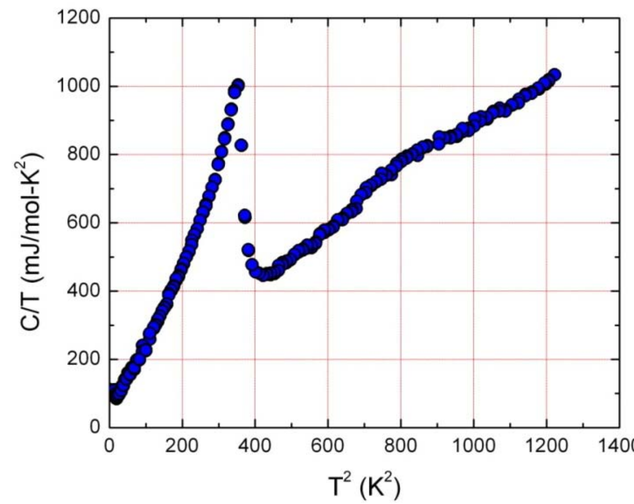
UIr_4Al_{15}



- Tetragonal – $P4_2/nmc$ (137)
- a = 9.0239(6) Å
- c = 15.513(2) Å
- V = 1263.2(2) $Å^3$

AF with T_N = 20K





16 RT_4Al_{15} analogs with (R=Ce, Pr, Sm, Yb, U; T=Fe, Co, Ru, Rh, Os, Ir) also synthesized

P.H. Tobash, *et al.* (in preparation)

Summary/Outlook

- **Developing a phenomenological approach to discover unconventional superconductors**

- $CePt_2In_7$ superconducts at 2.1K (more layered/smaller DOS?)

- Focus attention on 5f superconductors
    - Pu115's appear related to Ce115's
    - Can we exploit the larger bandwidth of 5f's relative to 4f's