The role of local structural disorder in non-Fermi liquid *f*-electron intermetallics

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Outline

- Introduction and Motivation
  - — Why worry or care about disorder?
  - — How can we include disorder in the theories?
  - — What can we do experimentally?

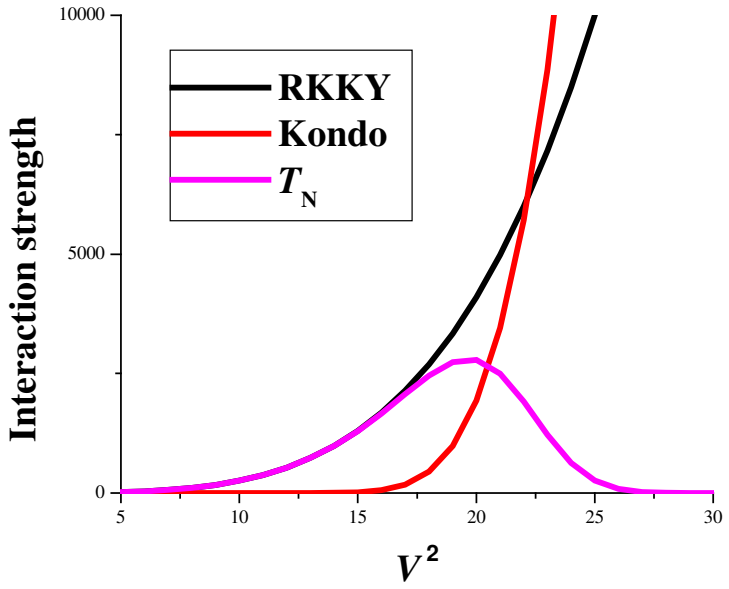
- $UCu_{5-x}Pd_x$ ($CeRhRuSi_2$)
  - — a disordered system
  - — disorder is partially tunable
  - — Main results are the same
- $U_3Ni_3Sn_4$ *C*/*T* magnetic field dependence

- Conclusions

Disorder and hybridization

binding energies: $k_B T_{RKKY} \sim N(0)\, V_{fd}^4/\varepsilon_f^2$

$k_B T_K \sim \exp[-\varepsilon_f / N(0)\, V_{fd}^2]$



Teaser: *Can molecular systems be included?*

What happens when interactions are equal and drive a zero temperature transition?

What if there is a distribution of interaction strengths?

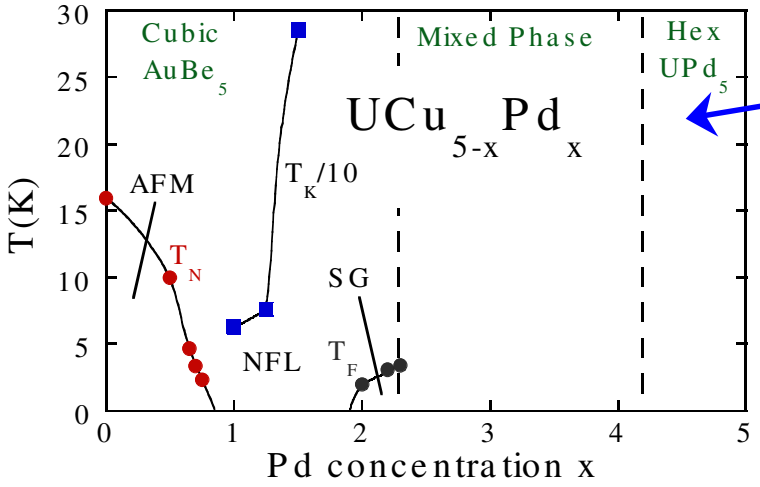
MAGNETIC GROUND STATES

	Interaction disorder →	
Hybridization →	(anti-) ferromagnets UCu_5	Spin glasses URh_2Ge_2
	Anderson lattices $Yb_{1-x}Lu_xAl_3$	non-Fermi liquids $UPdCu_4$

NFL?

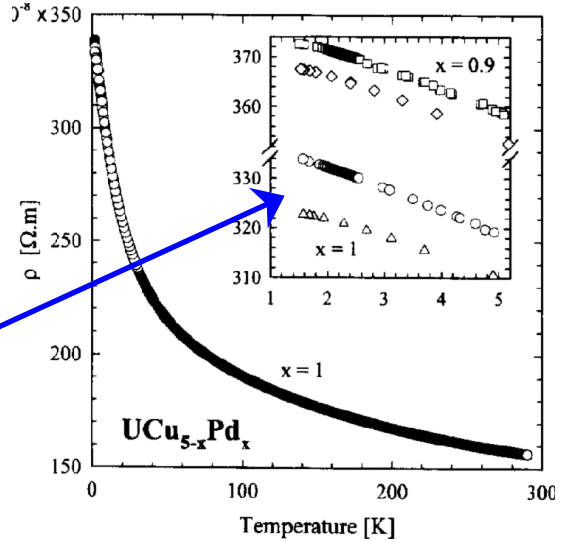
$U_3Ni_3Sn_4$

NFL behavior in $UCu_{5-x}Pd_x$

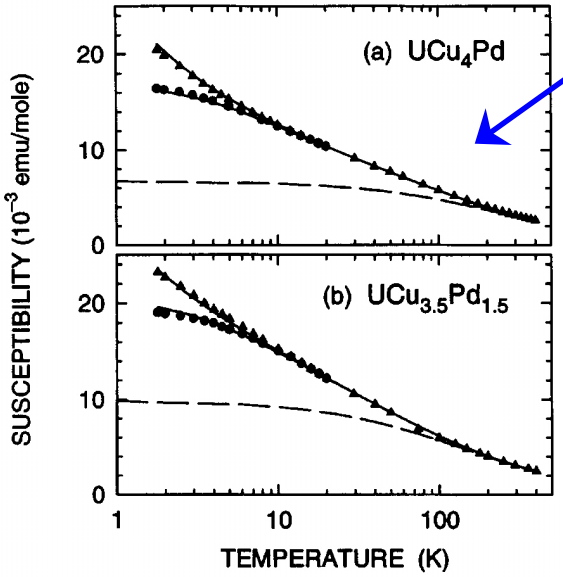


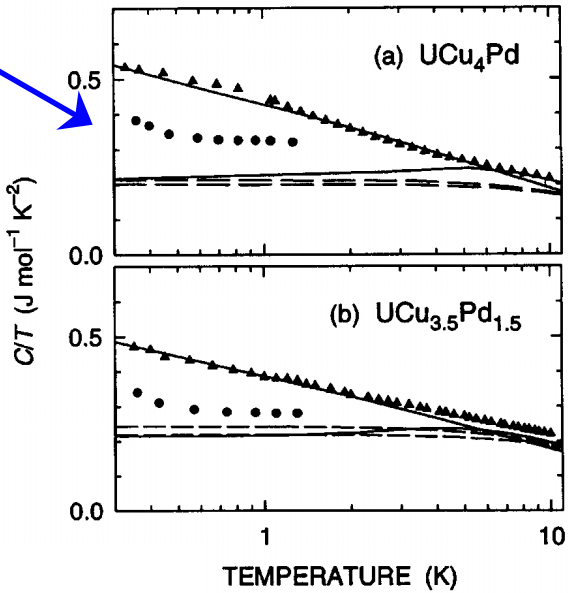
NFL behavior exists near phase boundaries

resistivity as T goes to 0 K has a weaker power law than T^2



Susceptibility and heat capacity have logarithmic divergences as T goes to 0 K.





Vollmer et al., PRB, 2000. Bernal et al. PRL, 1995. R. Chau, Ph.D. dissertation UCSD, 1998.

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Non-Fermi-liquid behavior in *d*- and *f*-electron metals

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Unanswered questions

- **Is disorder a necessary component?**
  - **— first NFL's were all substituted**
  - **— many new "ordered" ones coming online**
  - **— even "ordered" ones may have issues (eg. $CeCu_2Si_2$)**

- **Does disorder even matter?**
  - **— well, how much are we talking about?**

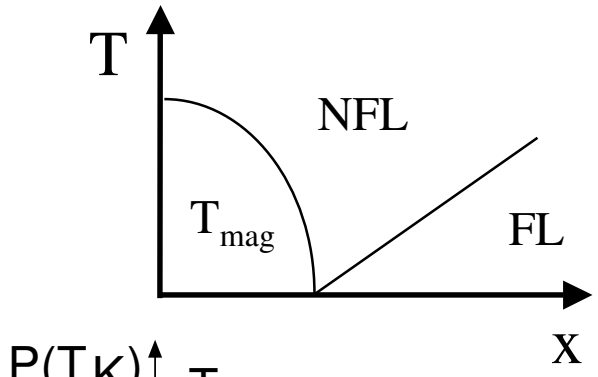
- **Is this a new state of matter?**

Three Possible Types of NFL Models

(There are others: multichannel Kondo, etc…)

Quantum Critical Point

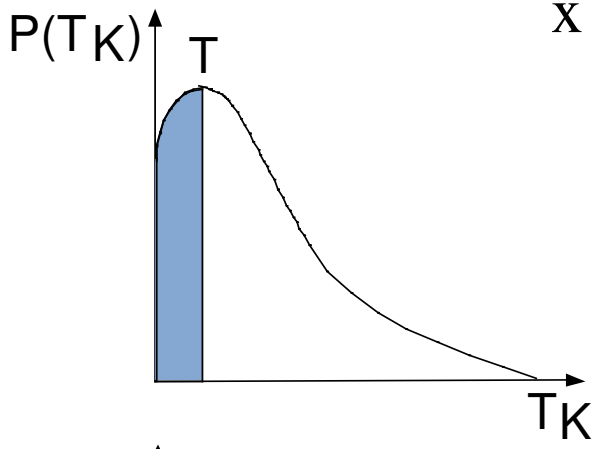
NFL is generated from critical fluctuations above a zero-temperature critical point (Millis *et al.* '93) (Rappoport *et al.*, 2001).



Kondo Disorder Model

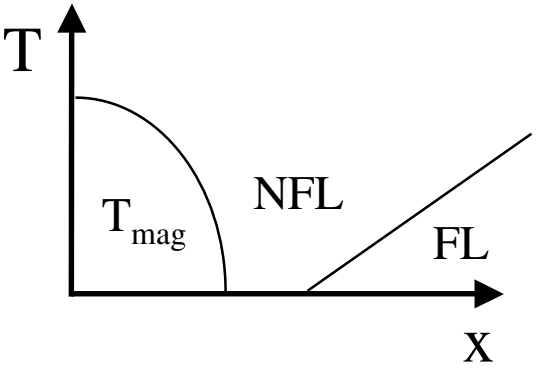
Bernal *et al.* '95

Disorder causes distribution of T_K's within a strict single-impurity model. Moments with $T_K < T$ are unquenched and give rise to NFL behavior.



Disorder+Competition (Griffiths)

NFL behavior due to proximity to a metal-insulator transition fixed point (Anderson localization,Miranda *et al.*) or to a magnetic/nonmagnetic fixed point (RKKY, Castro Neto *et al.*), each in presence of disorder and anisotropy.



Effects of lattice disorder (Kondo lattice disorder model, or KLDM)

Only Kondo: interactions

$$T_K = T_F \exp - \frac{\varepsilon_f}{N(0)V_{\text{Total}}^2}$$

tight-binding : model

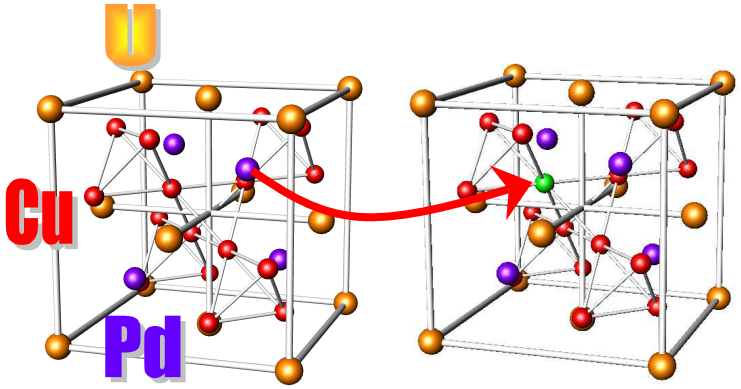
$$V_{fd} = \eta_{fd} \frac{(r_f^{\,5} r_d^{\,3})^{1/2}}{R_{f-d}^{\,6}}$$

Harrison and Straub

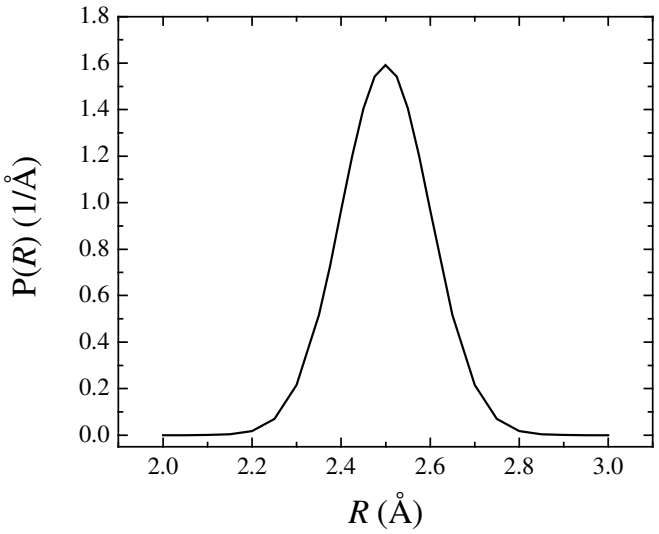
T_F: conduction band width
ε_f: *f*-level energy
V_{Total}: *f*/d hybridization energy

η_{fd}: constant
r_f: outer *f*-radius
r_d: outer *d*-radius
$R_{f\text{-}d}$: *f*-*d* bond length

- **Two types of lattice disorder: discrete and continuous**

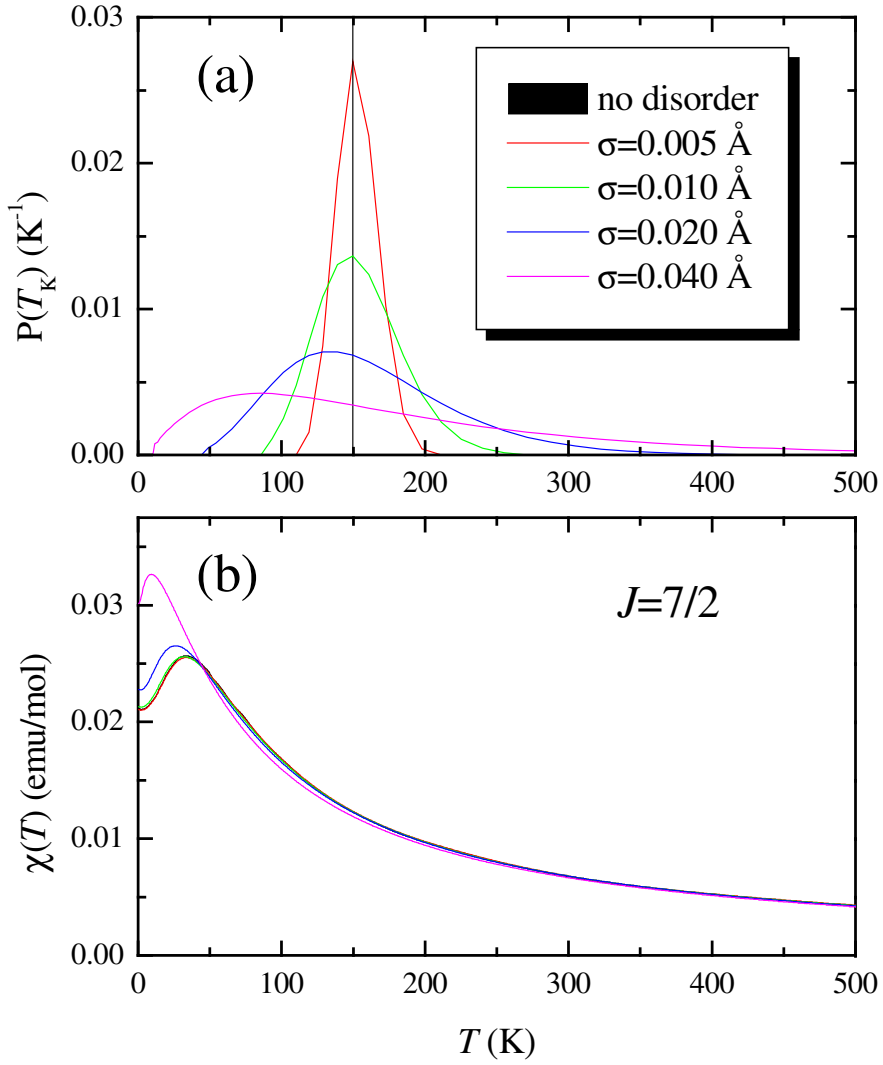


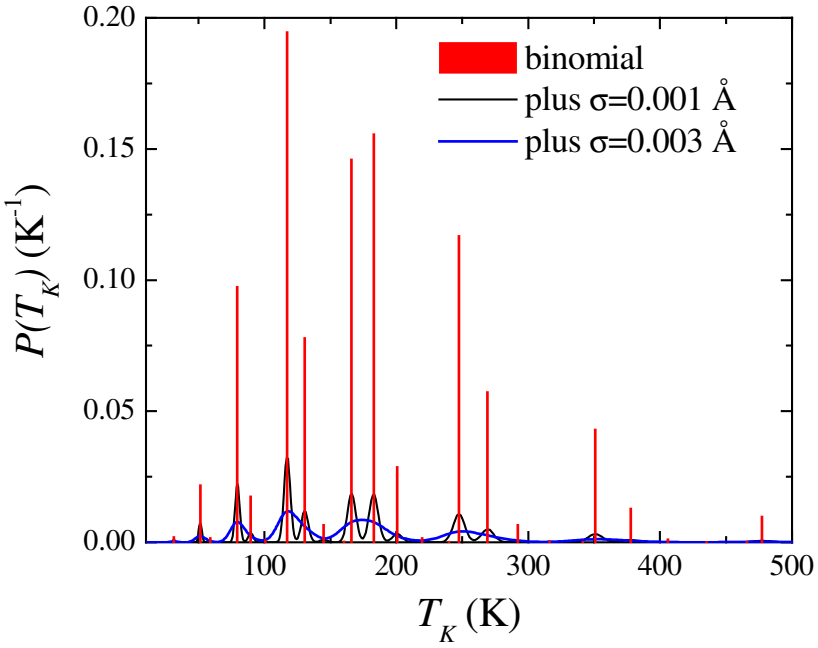
$$V_{Total} = \sum_{\text{bonds}} V_{fd}$$



- r_d varies as species change
- P(V) involves convolution with P(R)

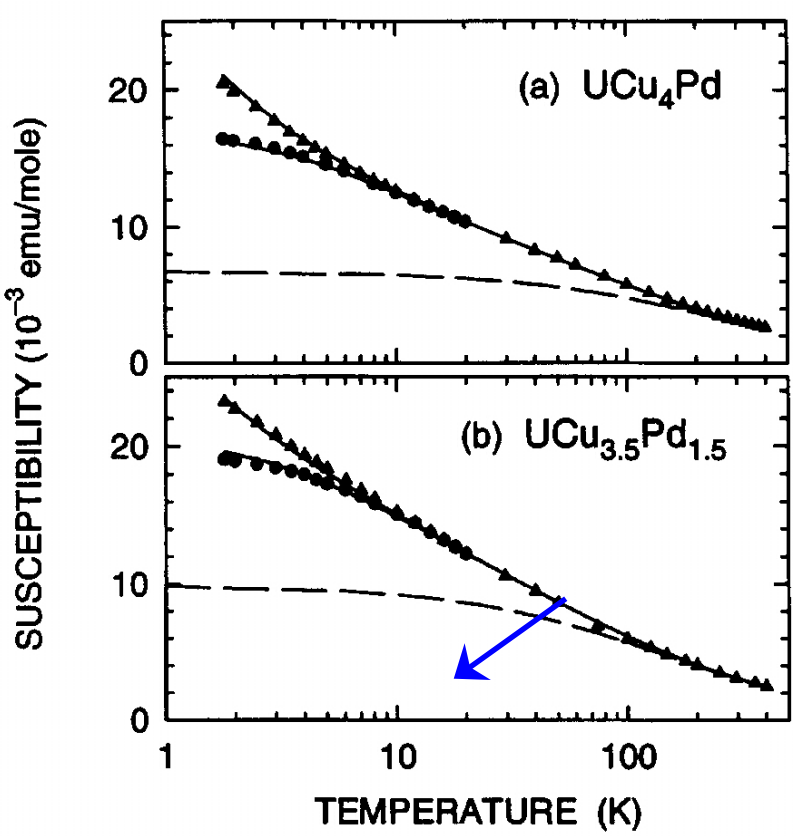
NFL *must* have continuous disorder in KLDM!

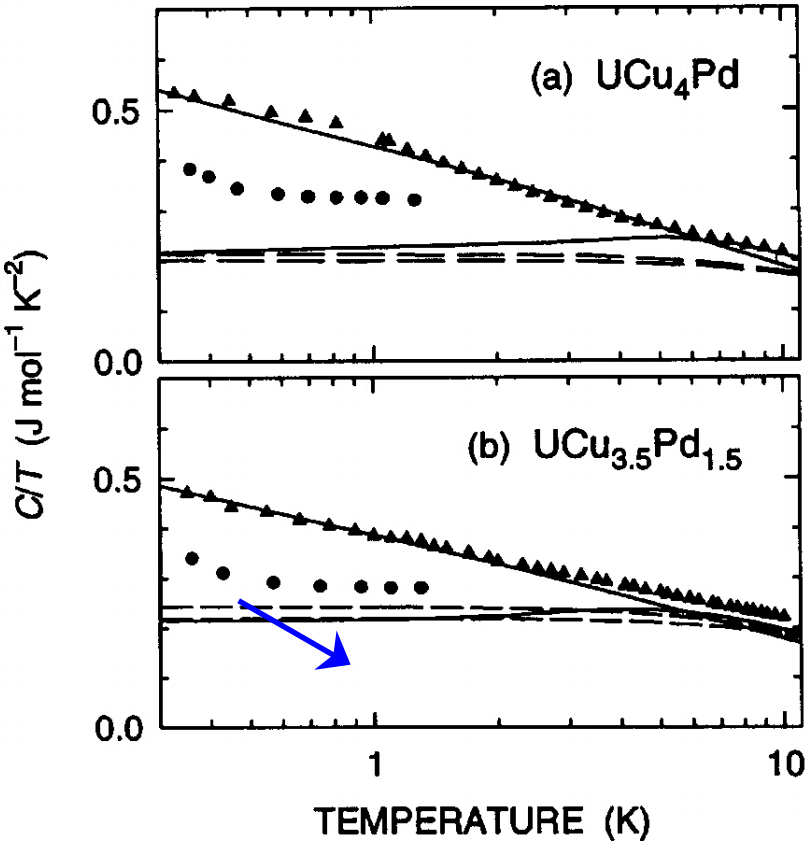




- **For KLDM to work, must have weight in P(T_K) at very low T_K.**
- **Site interchange will never provide this weight by itself!**

NFL behavior in $UCu_{5-x}Pd_x$

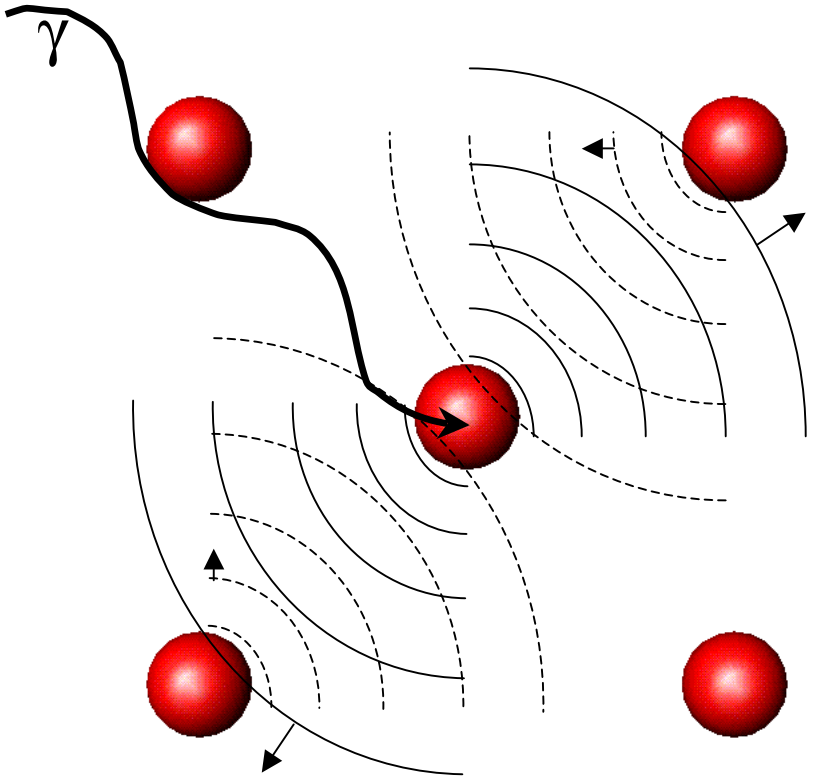




- **A distribution of T_K's can describe all these data!**
- Warning: this is pedantic: KDM has many problems!

Vollmer et al., PRB, 2000. Bernal et al. PRL, 1995. R. Chau, Ph.D. dissertation UCSD, 1998.

Interference of photoelectron waves



"I was brought up to look at the atom as a nice hard fellow, red or grey in colour according to taste."

- Lord Rutherford

- **Interference of outgoing and incoming part of photoelectron modulates absorption coefficient:**

$$\mu \propto \left| \langle f | \varepsilon \cdot r | i \rangle \right|^2$$

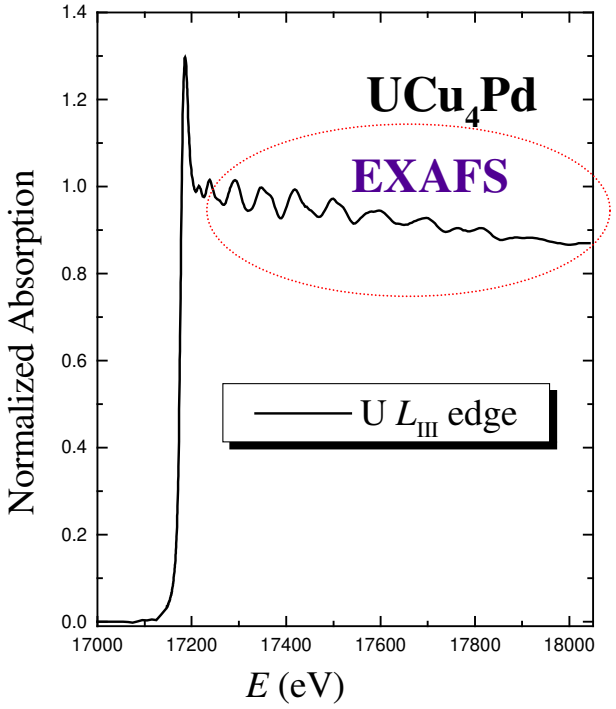
$$\mu = \mu_0 (1 + \chi(k))$$

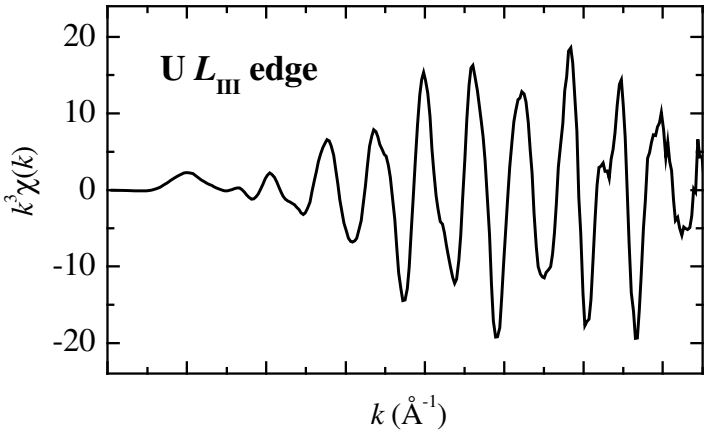
$$\chi(k) \propto \sum_i N_i \int g(r) \sin(2kr + \phi_{ci}) dr$$

g is a radial pair - distribution function

- **<u>Big advantage</u>: Atomic-species specific.**
- ***Disadvantages*: very short range (<~5-6 Å), sensitive to multiple scattering, overlapping edges...**

Extended x-ray absorption fine-structure (EXAFS)



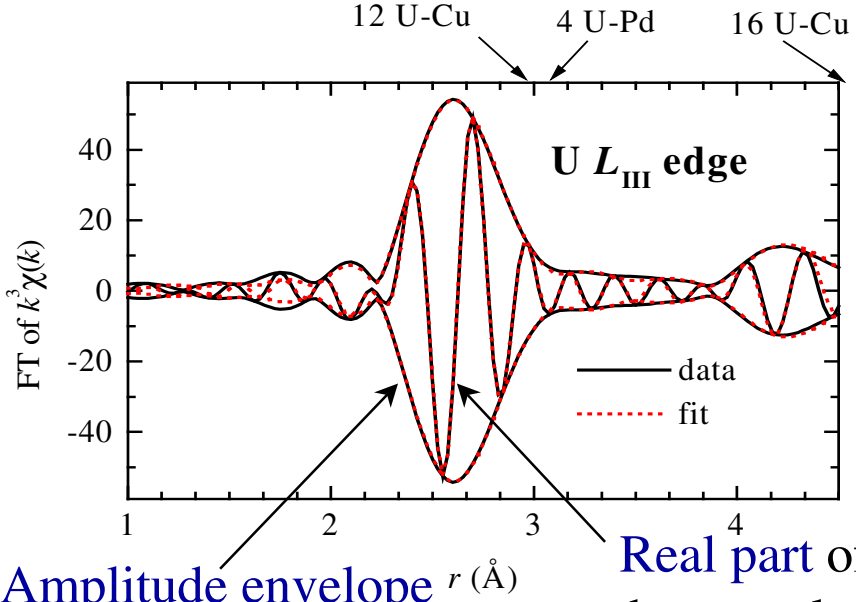


- sample absorption is given by

$\mu t = \log_e(I_1/I_0)$

- EXAFS $\chi(k)=[\mu(k)-\mu_0(k)]/\mu_0(k)$

$$\chi(k) \propto \sum_i N_i \int g(r) F(k,r) \sin(2kr + \phi_{ci}) dr$$

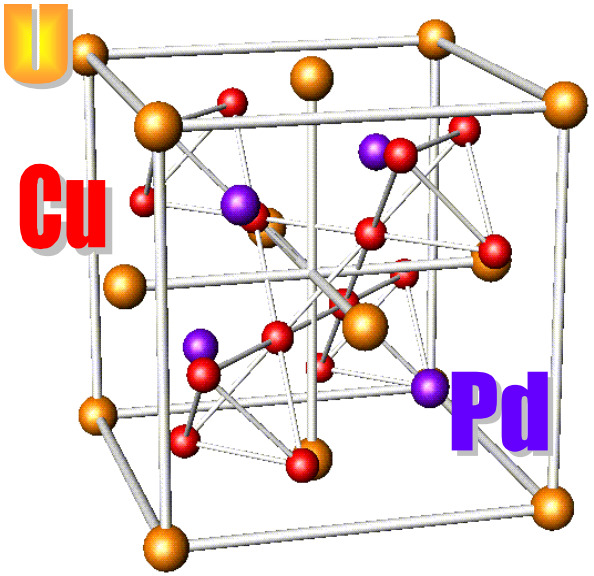


Amplitude envelope $[Re^2+Im^2]^{1/2}$

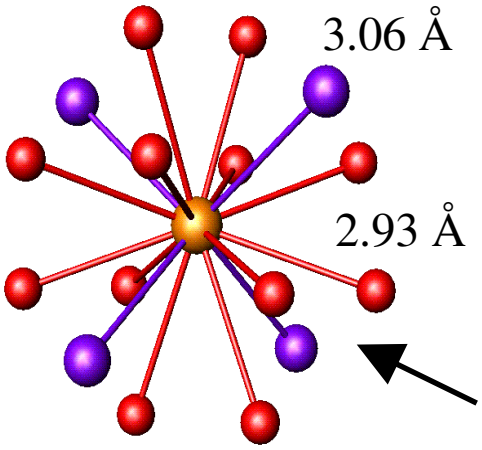
Real part of the complex transform

UCu_4Pd average and local structure

nominal

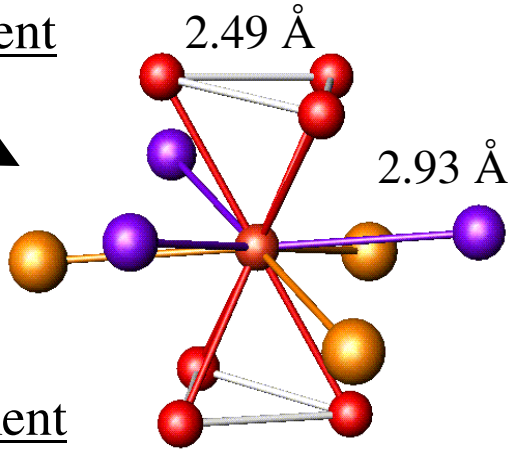


- **U (4*a*) environment is identical to Pd (4*c*) environment, except U/Pd are switched. Nearest-neighbors are Cu (16e) at ~2.93 Å**
- **Cu environment differs due to tetrahedrons. Nearest-neighbors are Cu at 2.49 Å**
- **Determine amount of site interchange by number of *Pd´*-Cu pairs at 2.49 Å**
- ***Definition:* Pd´ denotes a Pd on a 16e site, Cu´ denotes a Cu on a 4c site.**

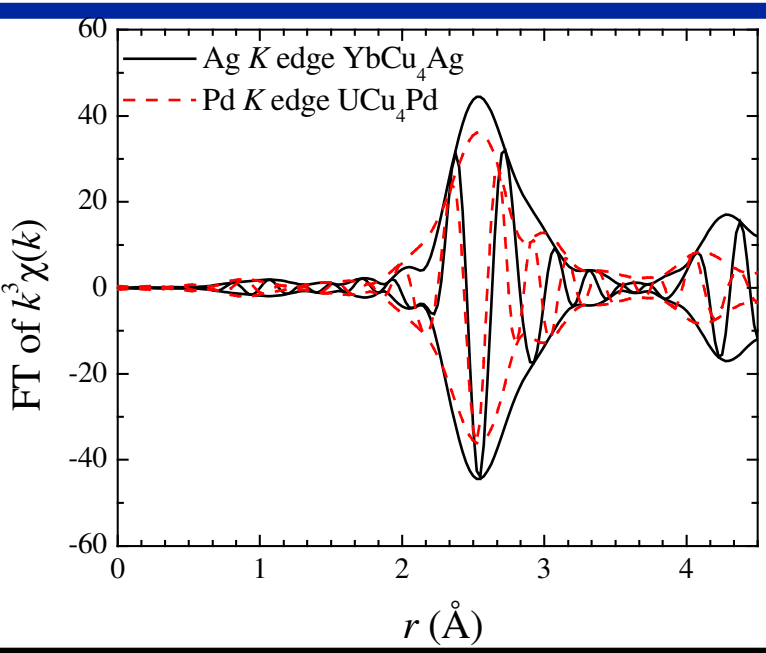


Local U and Pd environment

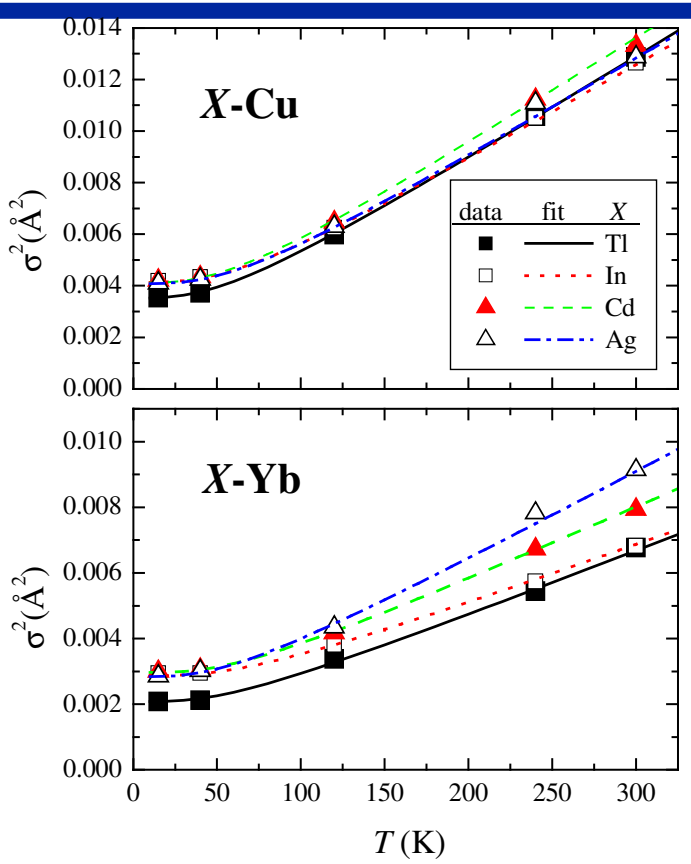
Local Copper environment



A "zero-disorder" example: $YbCu_4X$



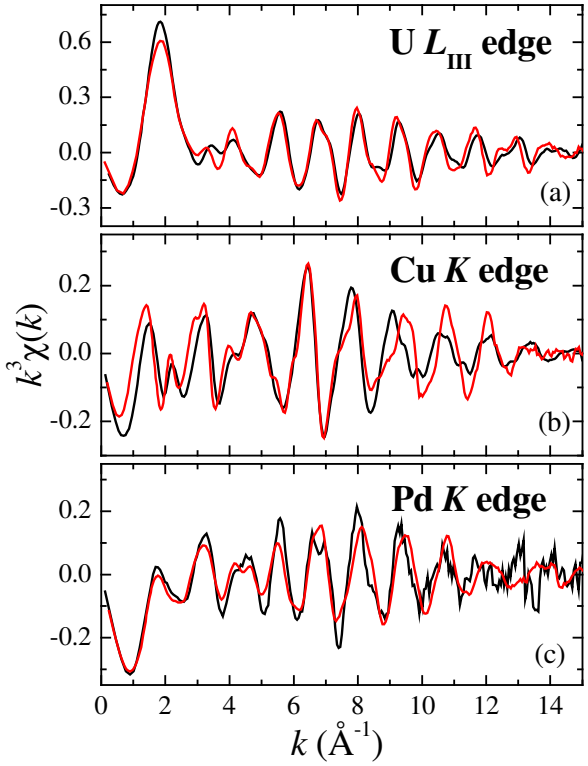
X	S02	Θ_{cD}(K)		σ_{static}^2(Å)		X/Cu inter-change
		Cu	Yb	Cu	Yb	
Tl	0.89(5)	230(5)	230(5)	**0.0004(4)**	**0.0005(5)**	**4(1)%**
In	1.04(5)	252(5)	280(5)	**0.0009(4)**	**0.0011(5)**	**2(3)%**
Cd	0.98(5)	240(5)	255(5)	**0.0007(4)**	**0.0010(5)**	**5(5)%**
Ag	0.91(5)	250(5)	235(5)	**0.0008(4)**	**0.0006(5)**	**2(2)%**

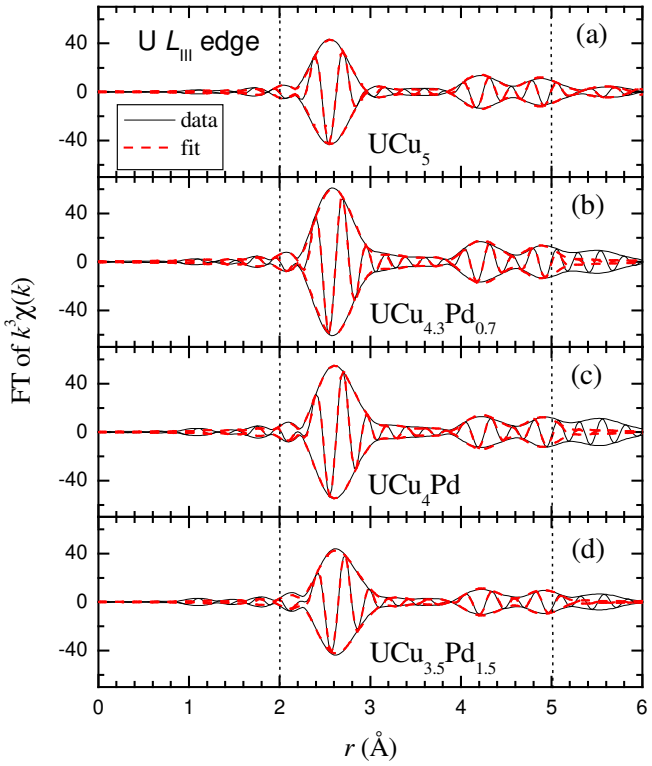


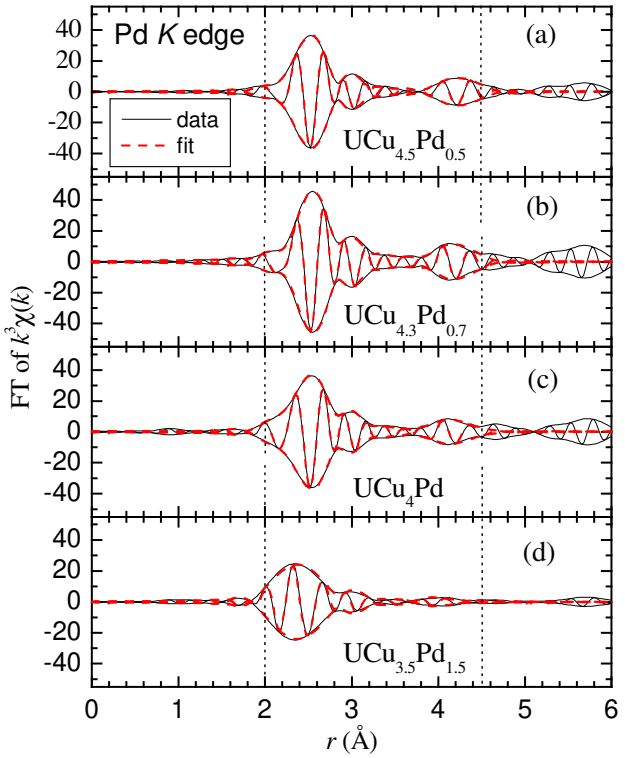
$$\sigma_{AB}^2(T)=\sigma_{static}^2+F(\mu_{AB},\Theta_{cD})$$

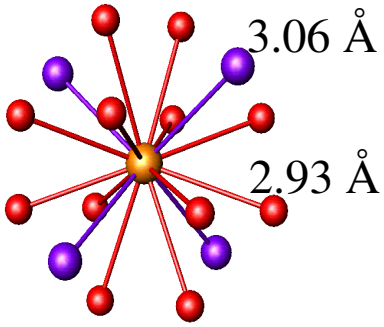
J. L. Lawrence et al., PRB 63, 054427 (2000).

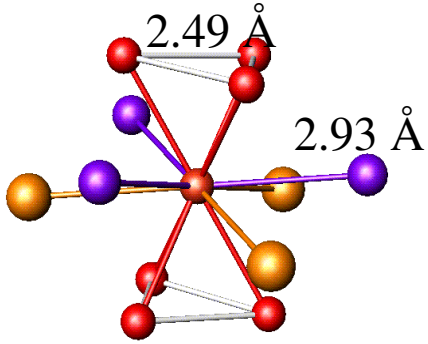
XAFS data on $UCu_{5-x}Pd_x$



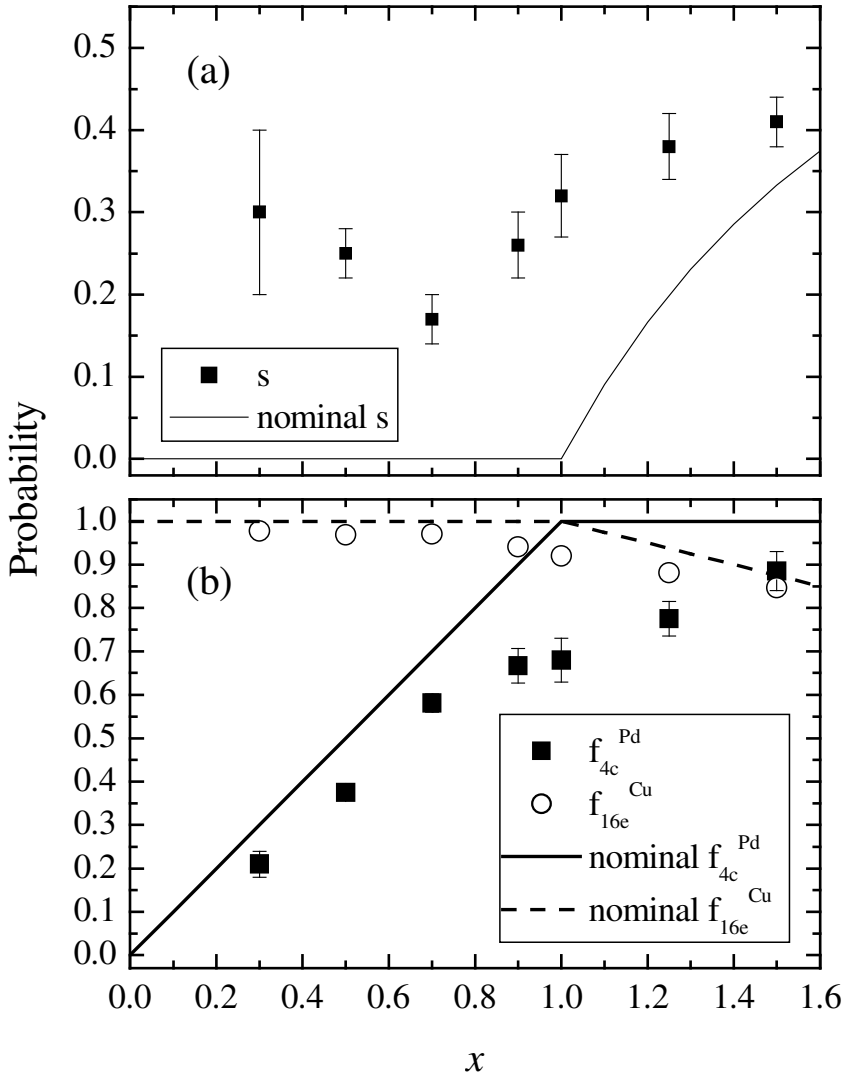








Pd *K*-edge fit results



- **Fit to all single-scattering paths out to the 16 Pd-Cu's at ~4.59 Å.**
- **Including all site interchange, fits use 15 paths.**
- **Like bond lengths constrained together.**
- **Like bond length Debye-Wallers constrained together ($\sigma_A^2=(\mu_B/\mu_A)\sigma_B^2$.**
- **Amplitude ratio's constrained.**

Two possible descriptions:

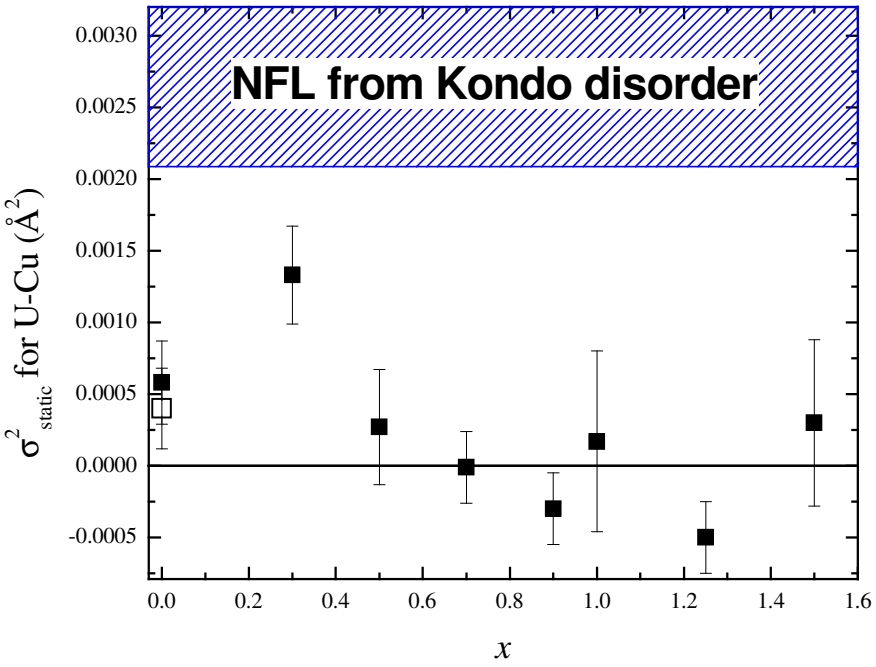
***s*, *x*: $s = N_{Pd}(16e)/N_{Pd}(Total)$**

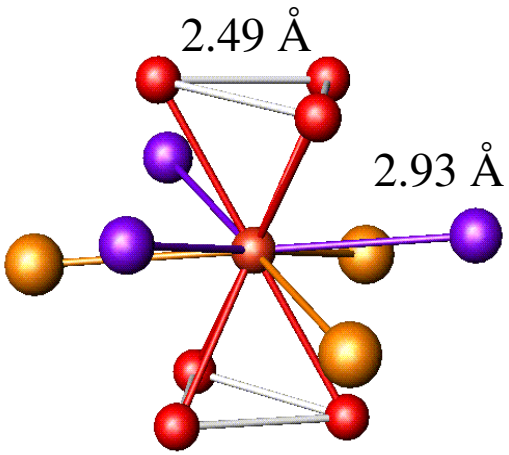
f_{4c}^{Pd}, f_{16e}^{Cu}: f_{4c}^{Pd} is fraction of 4*c* sites with Pd, etc.

e.g. Pd´-Cu @ 2.5 Å has $6S_0^2 s\, f_{16e}^{Cu}$ neighbors

C. H. Booth et al., PRL 81, 3960 (1998); E. D. Bauer et al., PRB 65, 245114 (2002).

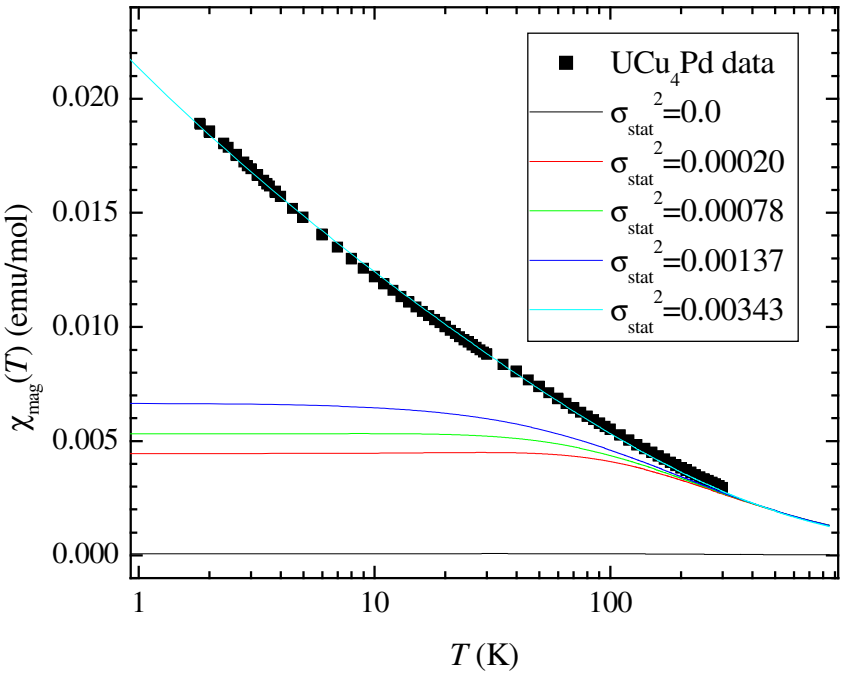
No measurable continuous U-Cu disorder!

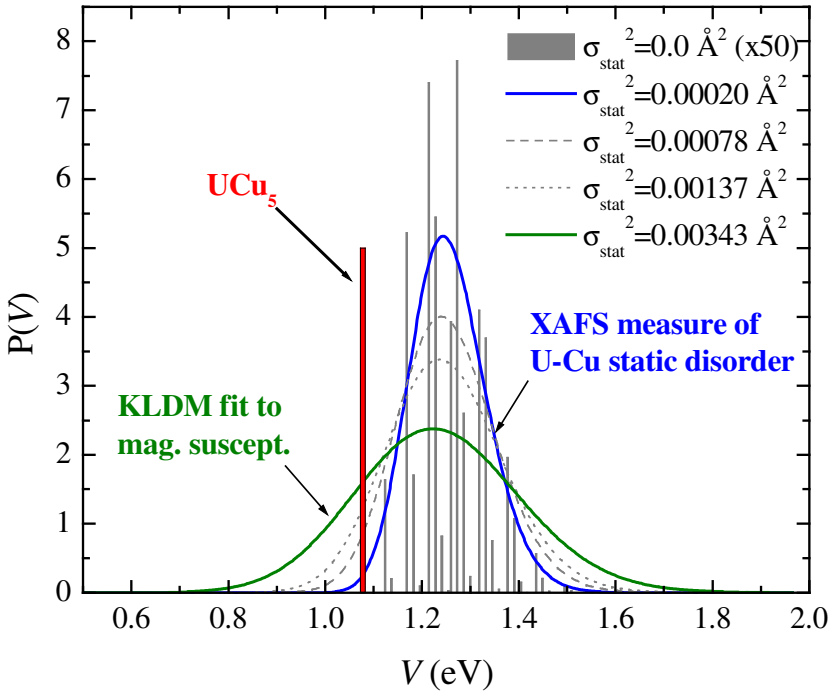




- NFL "limit" for KLDM is generous: we estimate the best fit with 0.0034 Å^2.
- Only the x=0.3 sample is anomalous… oxidation?
- Cu *K* edge fits indicate a nearest neighbor Cu-Cu distance of ~2.48 Å
- Pd *K* edge fits indicate a Pd´-Cu distance of ~2.55 Å
- Together no σ_{static}^2 for U-Cu, the Cu displacements near a Pd´ must be nearly perpendicular to the U-Cu pairs.

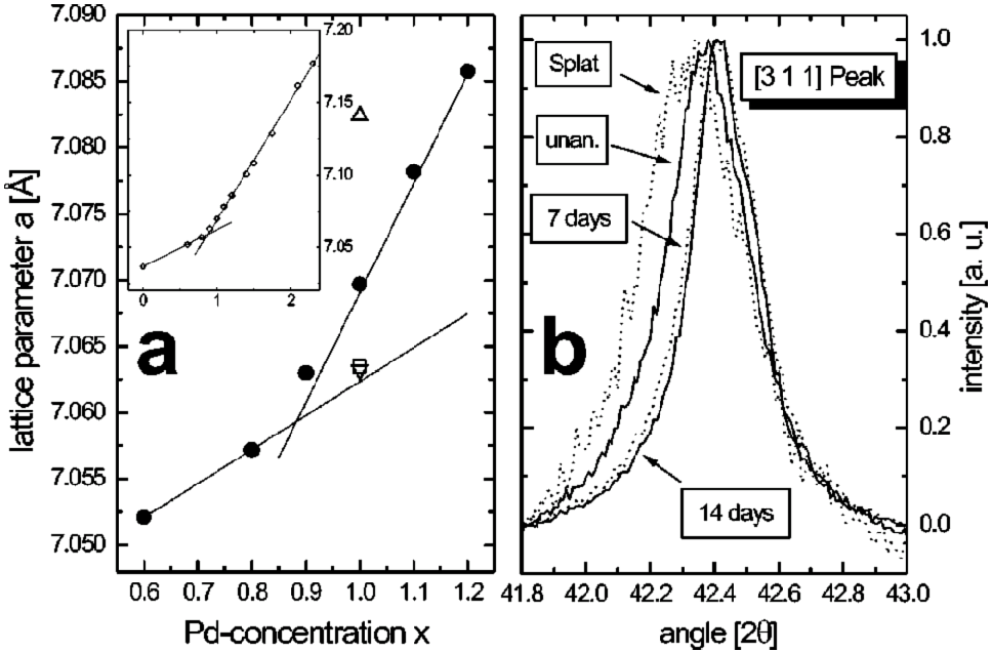
Disorder: Is it enough?

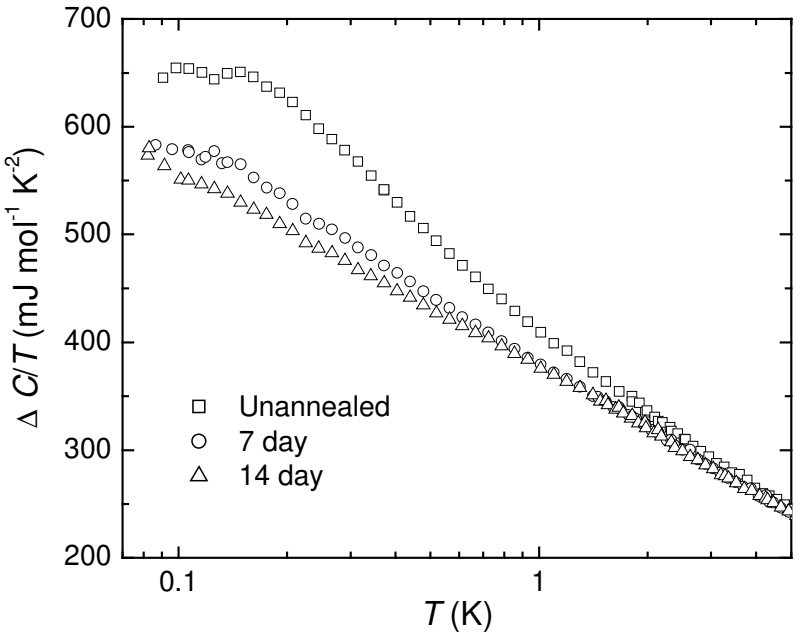




- ***KDM*: NFL is not from disorder in V_{fd}. This probably can't generate enough disorder in N(0) either (Miranda).**
- ***RKKY clusters?* ~0.5% of uranium environments have a V_{fd} that is equal to or less than that in UCu_5.**
- ***Anderson localization?* only 0.0025% of UCu_5-like uraniums have a similar neighbor.**

Effects of annealing

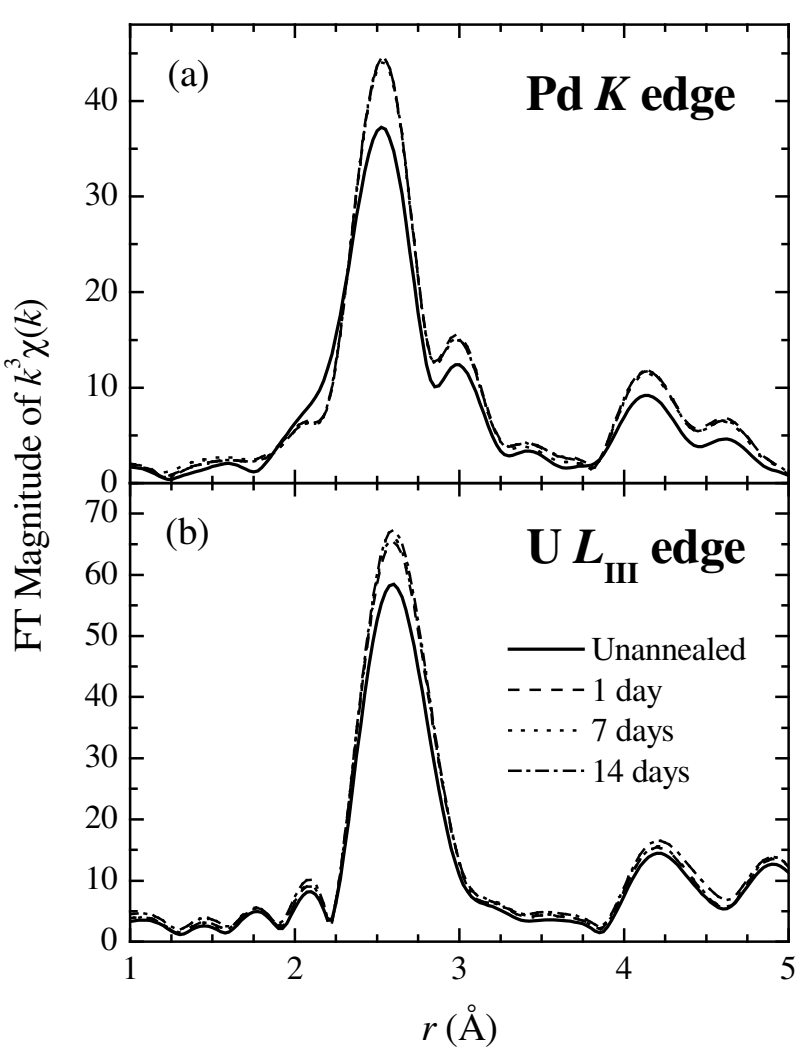




A. Weber et al., PRB 63, 205116 (2001).

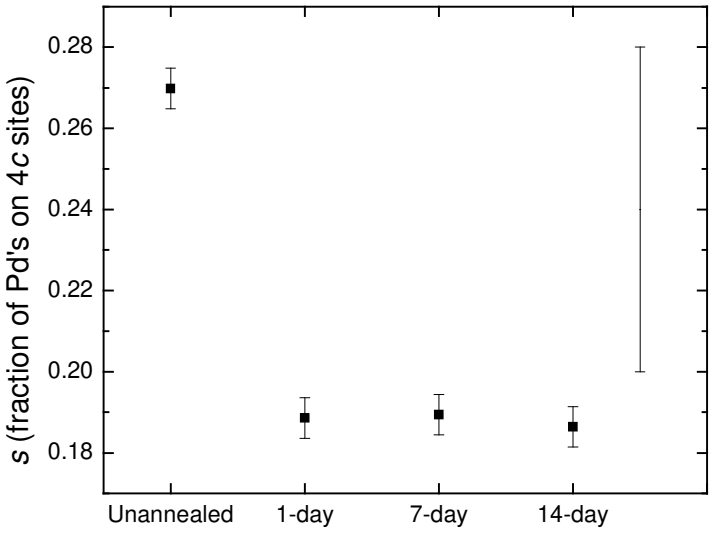
- **Annealing suppresses spin glass transition, removes linear resistivity but logarithmic C/T remains ?!?!?**
- **Quick point: Entropy under this logarithmic divergence is close to R log 2**

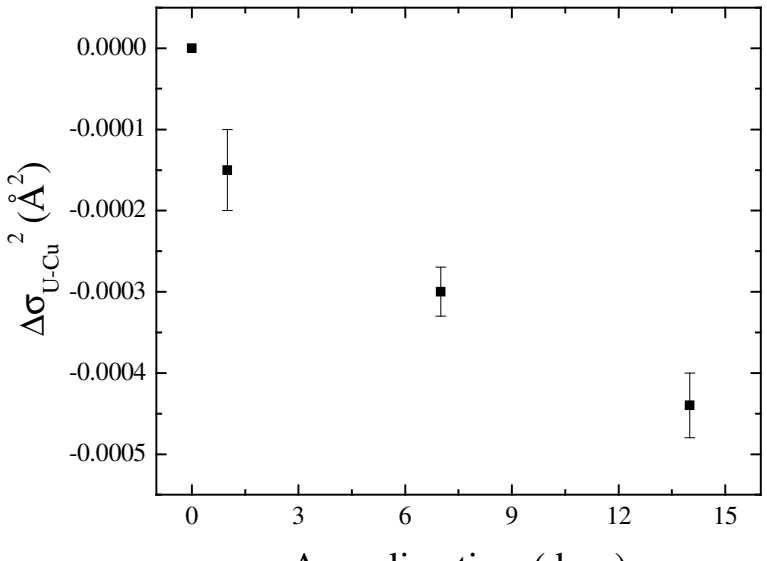
Both site interchange and bond length distributions affected by annealing



- Measure (*discrete*) site interchange with Pd *K* edge XAFS
- Measure (*continuous*) U-*X* bond length disorder with temperature dependence of distribution widths (Debye-Waller factors).
- *Complication*: U-Cu and U-Pd pairs strongly overlap, so need to be able to include degree of site interchange as a constraint to the U L_{III}-edge fits.
- *Solution:* Fit to a site interchange model.

Effects of annealing

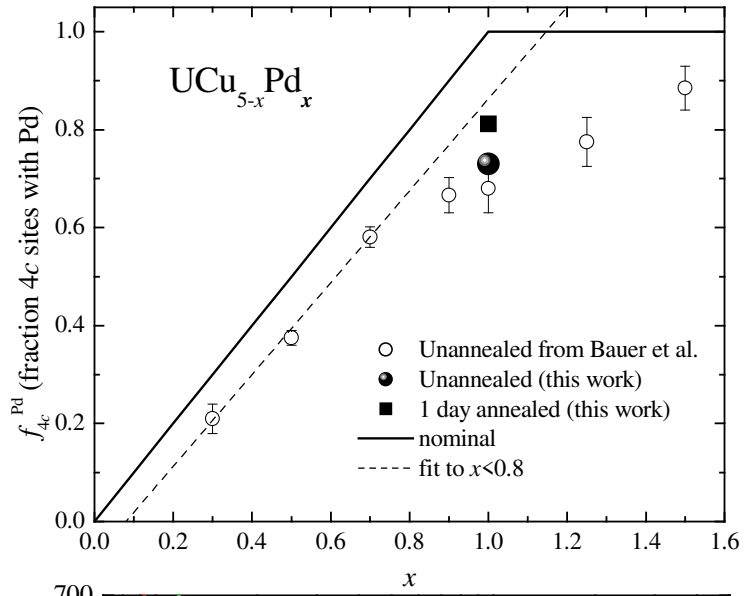


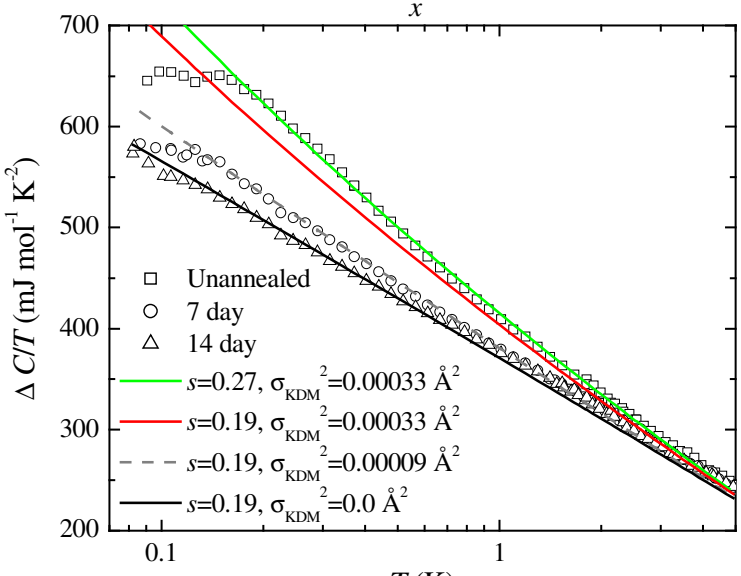


- **Structurally, two things happen:**
  - **site interchange is reduced, but not after more than 1 day of annealing**
  - **U-Cu bond length distribution width decreases, even after 14 days of annealing**

- **Main points:**
  - ***s* decreases, but is still fairly large**
  - **U-Cu orders, but it is already very close to fully ordered ($\Delta\sigma$ ~ -0.02 Å)**

Effects of annealing





- f_{4c}^{Pd} of unannealed samples very consistent with changes in lattice parameter: x vs. f_{4c}^{Pd} is linear, except with a change in slope at x ~ 0.85
- Annealing increases f_{4c}^{Pd}, similarly to change in d
- It is possible to parameterize *changes* in heat capacity as arising only from *changes* in s and $\sigma_{U\text{-}Cu}$

$$W_V^2 = W_0^2 + W_{KDM}^2$$

"Dark" width

$$W_{KDM}^2 \Rightarrow \sigma_{KDM}^2$$

- **ANSWER(?): NFL state is somehow "pre-loaded", possibly as a consequence of disorder.**

What the heck is W_0?

$$T_{\mathrm{K}} = T_F \exp - \frac{\varepsilon_f}{N(0)V_{\mathrm{Total}}^2}$$

- **KDM by itself does not work!**
  - **linear resistivity goes away on annealing (Weber et al., PRB 63, 205116 (2001)**
  - **μSR indicates glassy spin dynamics (MacLaughlin et al., PRL 245114)**
  - **Short range (< unit cell) magnetic correlations exist (Aronson et al., PRL 87, 197205 (2001)**
  - **Distribution of moments at high fields (>51 kOe) inconsistent with KDM (Buttgen et al., 62, 11545 (2000)**
- **Disorder can generate width in N(0) (not enough says Miranda, but could be says Cox)**
- **Is W_0 due to a QCP? Idea is similar to proposed by Grempel and Rozenberg PRB 60, 4702 (1999), and to Rappoport et al., PRB 64, 140402 (2001).**
- **Is clustering important?**

$CeRhRuSi_2$

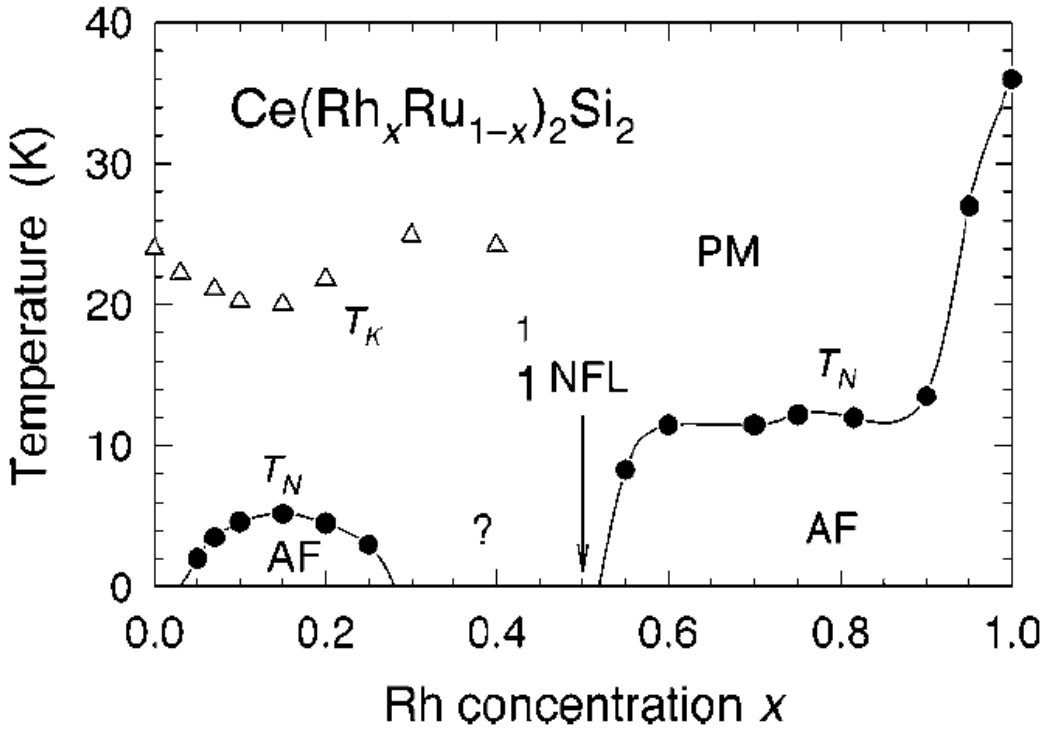
$U_3Ni_3Sn_4$

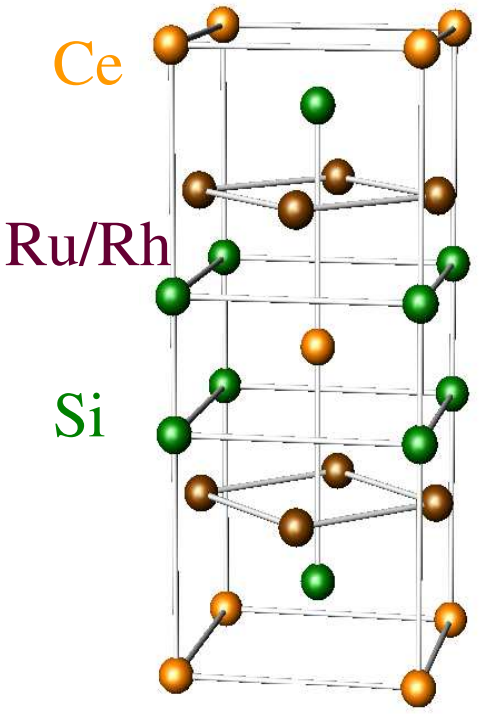
Data summary

Non-Fermi liquid (NFL) and $Ce(Ru_{1-x}Rh_x)_2Si_2$

		FL	NFL
Susceptibility $\chi(T)$	$\propto$	*constant*	-log(T)
Specific heat C(T)/T	$\propto$	γ	-log(T)
Electrical resistivity $\rho(T)$	$\propto$	T^2	T

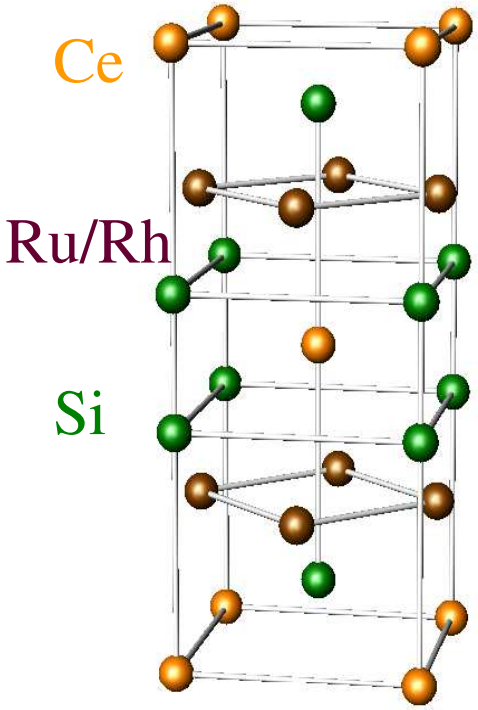
G. R. Stewart, RMP, (2001)

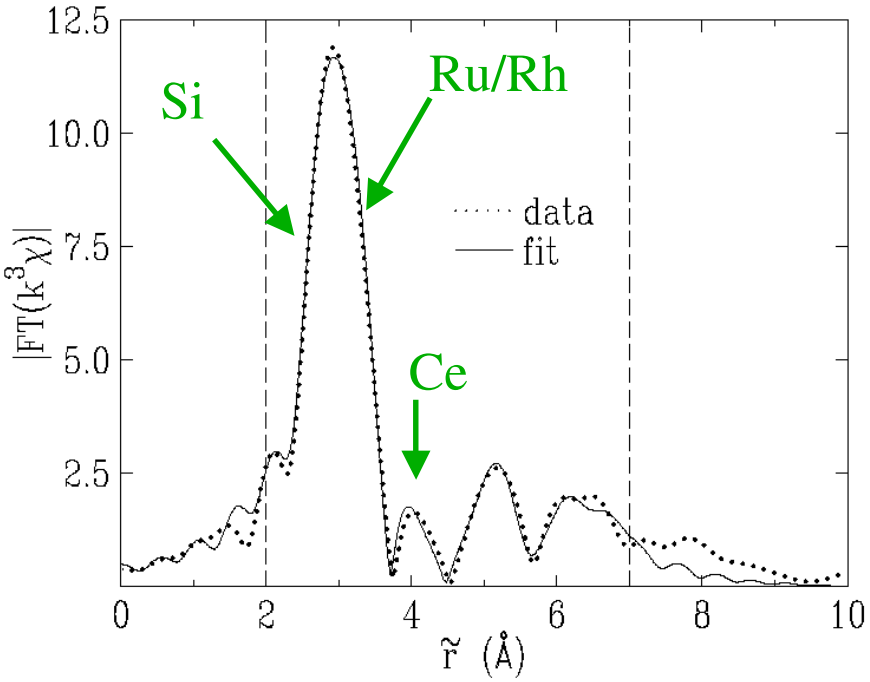




C. Y. Liu *et al.*, PRB **61**, 432 (2000)

XAFS Study near Ce atom in $CeRuRhSi_2$





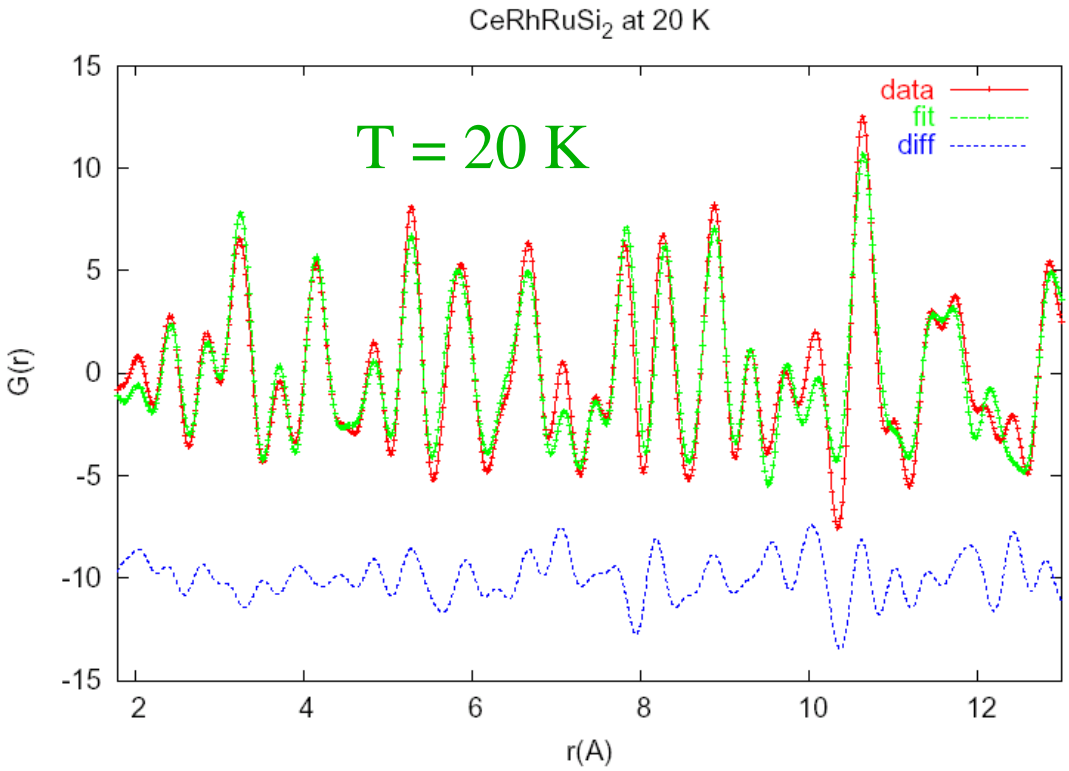
Ce L_3-edge
T = 20K

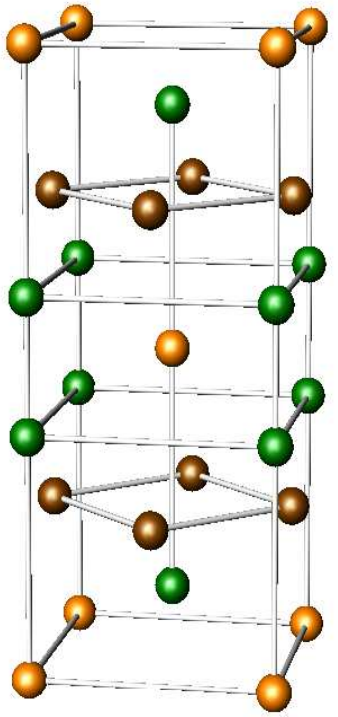
Bonding length (Å):
Ce-Si: 3.218(6)
Ce-Ru/Rh: 3.254(3)
Ce-Ce: 4.150(10)

Fitting parameters:

S_0^2	total data point	variables	fitting freedom	r-factor	reduced-χ^2
0.85(10)	23	14	9	0.0054	3.26

PDF Analysis of $CeRuRhSi_2$





Total disorder factors:

u11 (Ce)	u11(Ru/Rh)	u11 (Si)
0.000359(7)	0.0030(11)	0.0029(3)

Small total disorder factors suggest that static disorder is negligible!!!

Summary of structural disorder in $CeRuRhSi_2$

	σ^2 (Å^2)	σ^2(static) (Å^2)	bonding length (Å)
Ce-Si	0.0033(6)	0.0003(4)	3.215(6), 3.187
Ce-Ru/Rh	0.0014(3)	-0.0004(2)	3.254(3), 3.257
Ce-Ce	0.0011(9)	0.0003(14)	4.15(1), 4.161
Ru-Si	0.0011(3)	-0.0003	2.373(3), 2.399
Ru-Ru/Rh	0.0011(2)	-0.0001	2.934(3), 2.888
Ru-Ce	0.0022(2)	-0.0003	3.231(4), 3.257
Rh-Si	0.0015(3)	0.0004(3)	2.390(4), 2.399
Rh-Ru/Rh	0.0012(1)	-0.0003(2)	2.911(3), 2.888
Rh-Ce	0.0025(3)	-0.0002(2)	3.236(4), 3.257

T = 20 K

XAFS:
Short-range

	u(11) (Å^2)
Ce	0.00036(1)
Ru/Rh	0.0030(11)
Si	0.0029(3)

PDF:
Intermediate-range

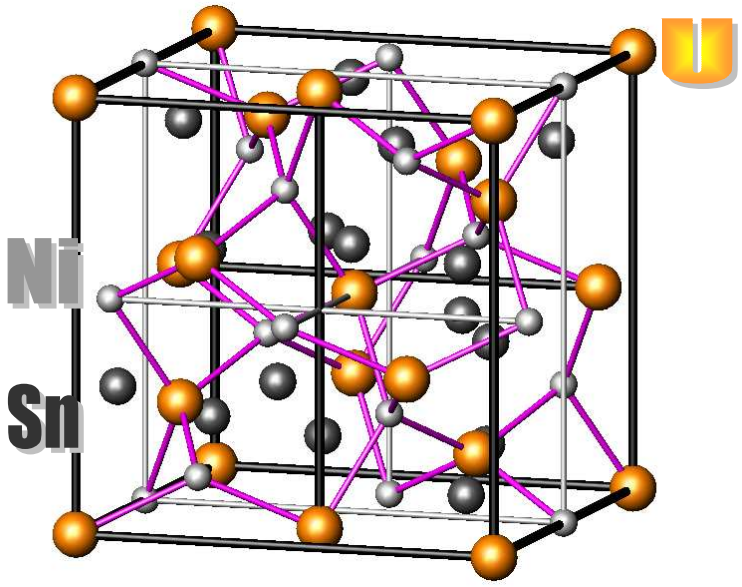
$U_3Ni_3Sn_4$

Last words

Is $U_3Ni_3Sn_4$ best described as near an AF QCP?

- $U_3Ni_3Sn_4$ is an undoped, ambient pressure non-Fermi liquid.
- Evidence of an AF critical point at –0.04 GPa (Estrela *et al.*, (2001)).
- A "Hertz and Millis" Quantum Critical Point?

$C/T \approx \gamma - A\,T^{0.5}$ (✓) $\quad \chi \propto T^{-0.3}$ (?, 0.5) $\quad \Delta\rho \propto T^{1.8}$ (?, 0.5)



- Cubic, bcc, I –43d, a_0=9.3524 Å
- residual resistivity 7 μΩ cm
- single crystal XRD good
- No temperature-dependent structural studies exist
- Disorder models have been shown to be capable of providing NFL behavior

No static offsets necessary

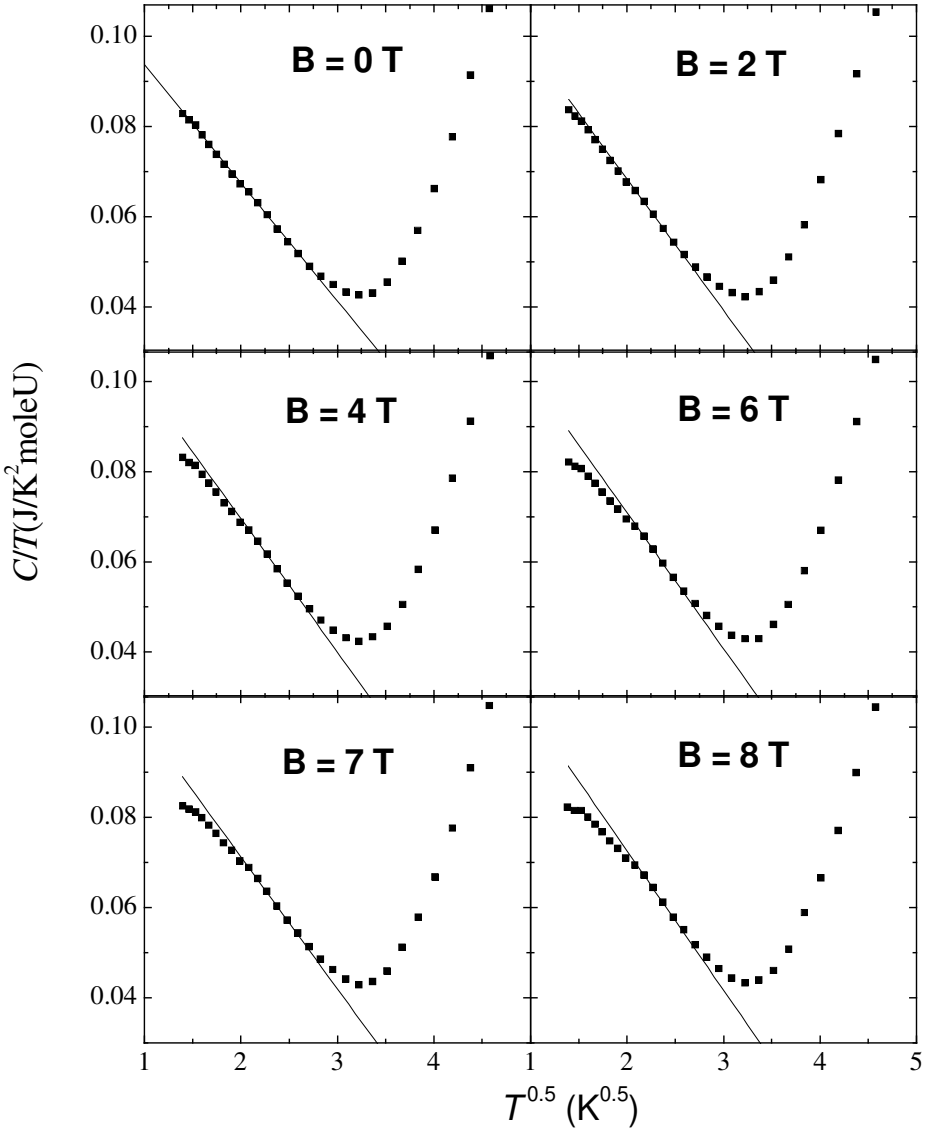
TABLE I: Final fit parameters to the U L_{III} and Sn K edge data at 20 K on three powder samples of $U_3Ni_3Sn_4$. U L_{III} edge fits have $S_0^2 = 0.73 \pm 0.06$ and $\Delta E_0 = -10.3 \pm 0.4$. Sn K edge fits have $S_0^2 = 0.95 \pm 0.06$ and $\Delta E_0 = -8.3 \pm 0.1$. Diffraction data was collected at room temperature.

			U2.9Ni3.0Sn3.9				U3.0Ni3.1Sn3.9				U3Ni3Sn4			
pair	N	R_{diff}	R	σ^2	σ^2_{static}	Θ_{cD}	R	σ^2	σ^2_{static}	Θ_{cD}	R	σ^2	σ^2_{static}	Θ_{cD}
U-Ni	4	2.864	2.848(4)	0.0019(4)	-0.0004(5)	282(2)	2.848(3)	0.0019(2)	-0.0005(5)	259(4)	2.848(3)	0.0018(2)	-0.0009(4)	252(5)
U-Sn	8	3.237	3.226(4)	0.0009(2)	-0.0009(3)	231(1)	3.226(3)	0.0009(2)	-0.0007(3)	233(1)	3.228(2)	0.0011(2)	-0.0006(3)	241(1)
U-U	8	4.374	4.355(5)	0.0014(2)	-0.0005(3)	173(2)	4.355(3)	0.0014(2)	-0.0000(3)	169(3)	4.36(1)	0.0016(3)	-0.0007(3)	159(4)
U-Ni	2	4.676	4.67(1)	0.0022(6)			4.67(1)	0.0022(4)			4.67(1)	0.0015(3)		
Sn-Ni	3	2.609	2.597(3)	0.0027(2)	-0.0003(2)	349(4)	2.599(3)	0.0027(2)	0.0008(3)	359(3)	2.604(3)	0.003(1)	0.001(1)	420(15)
Sn-U	6	3.237	3.232(7)	0.0016(2)	-0.0004(2)	246(2)	3.228(3)	0.0012(2)	-0.0004(2)	273(5)	3.223(5)	0.0006(4)	-0.0017(5)	202(12)
Sn-Sn	3	3.497	3.500(4)	0.004(1)	-0.000(1)	245(6)	3.496(3)	0.0017(5)	-0.0001(6)	250(20)	3.50(3)	0.003(3)	-0.003(3)	172(17)
Sn-Sn	2	4.050	4.03(1)	0.01(1)			4.02(1)	0.0024(7)			3.98(3)	0.003(3)		
Sn-Ni	3	4.232	4.16(5)	0.01(1)			4.22(1)	0.005(2)			4.25(3)	0.002(1)		
Sn-Sn	6	4.594	4.598(4)	0.0034(3)			4.596(3)	0.0023(2)			4.60(3)	0.002(1)		

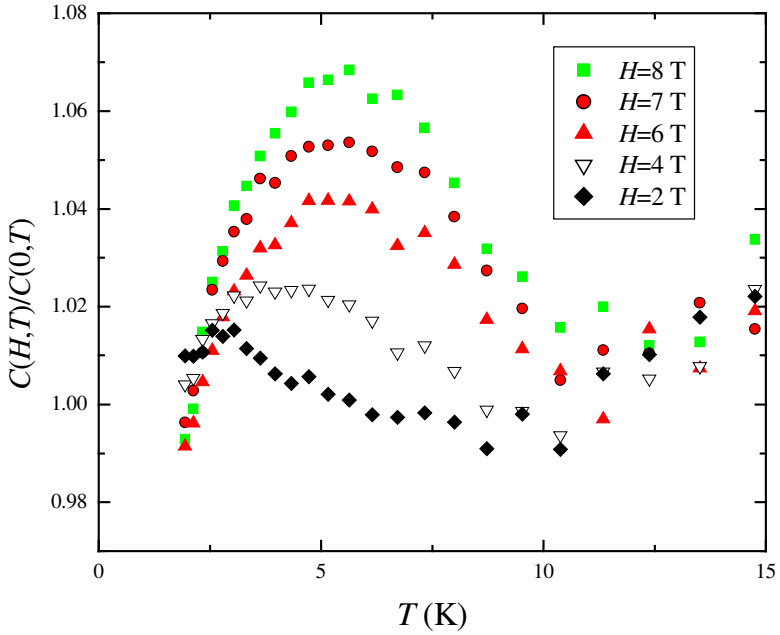
Atom pair	σ_{offset}^2(Å^2)	Θ_{cD}(K)
U-Ni	-0.0009(4)	252(5)
U-Sn	-0.0006(3)	241(1)
U-U	-0.0007(3)	159(4)

No evidence of site interchange either…

Field-dependence of heat capacity



- **Zero applied field, $C/T \sim T^{0.5}$, indicative of NFL behavior**
- **Fermi liquid behavior appears to be recovered in relatively small applied fields**
- **($U_3Ni_3Sn_4$ behaves similarly to $CeCoIn_5$, another system with a "negative pressure" critical point…)**

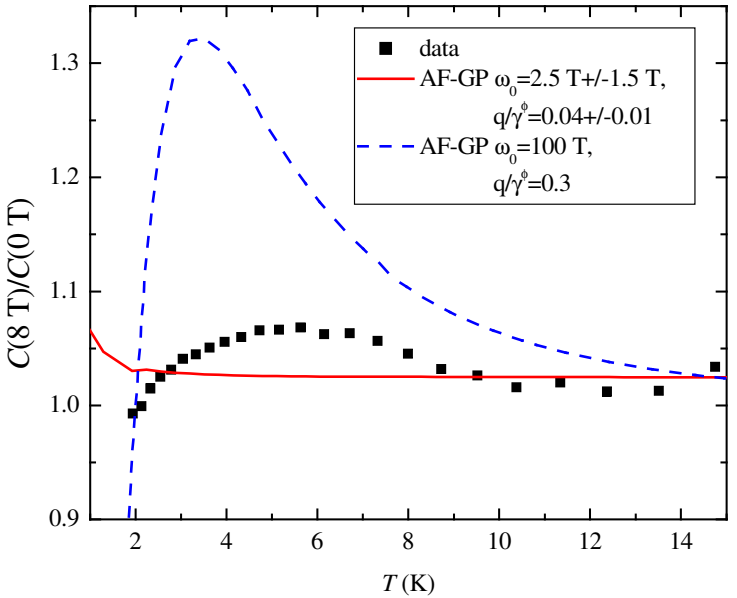


Comparison to Grifftihs-McCoy…
a Schottky anomaly?

- **High-field limit:**

$$C_{\mathrm{el}}/T \propto \frac{H^{2+\lambda/2}}{T^{3-\lambda/2}} \mathrm{e}^{-\mu_{\mathrm{eff}} H/T}$$

μ_{eff} is average effective moment of AF clusters… successfully applied to $La_{0.95}Ce_{0.05}RhIn_5$ (Kim et al, 2002)



- **More generally:**

$$C_{\mathrm{el}}(H,T) \propto \beta^2 \int_0^{\omega_0} d\Delta \Delta^{1-\lambda} (E_H^2 + \Delta^2) \operatorname{sech}^2(\beta\sqrt{E_H^2+\Delta^2}) \left[\ln\frac{\omega_0}{\Delta}\right]^{1-\theta}$$

$$E_H(\Delta) = q\mu_B \left[\frac{1}{\gamma}\ln(\frac{\omega_0}{\Delta})\right]^{\phi} H$$

Δ is the cluster tunneling energy

ω_0 is the tunneling energy for a single atom (cutoff)

q average moment within a cluster

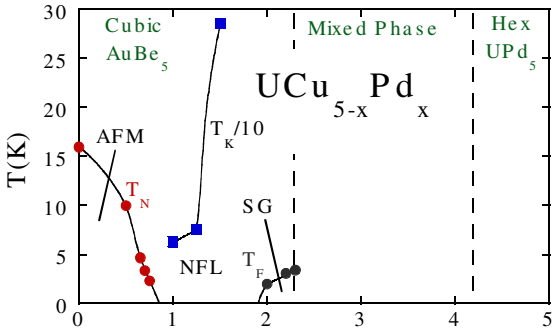
γ is an anisotropy parameter

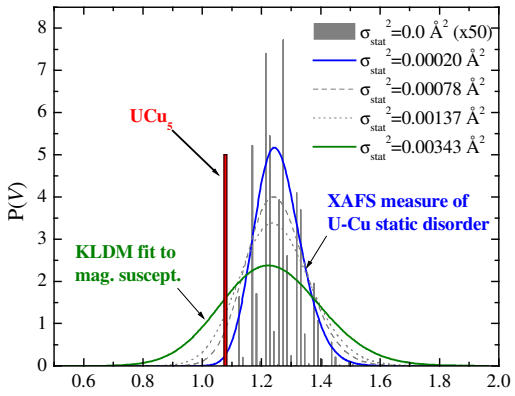
Data summary

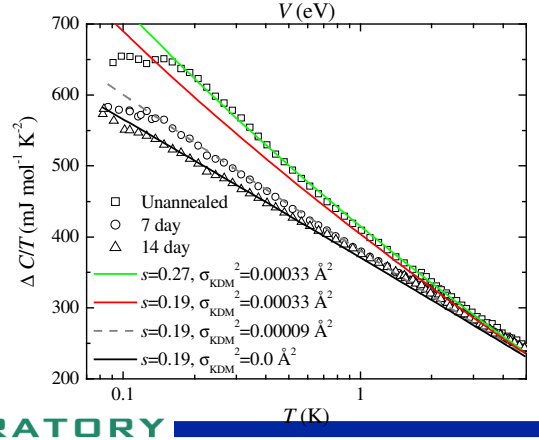
- UCu_4Pd (Disordered NFL)
  - — Pd/Cu site interchange, tunable by annealing
  - — Very little bond-length disorder
    - Not enough for KLDM (that's all in V_{fd})
    - Changes in annealing indicate there is at least a little, and it does affect the magnetic properties
- $CeRhRuSi_2$ (Disordered NFL)
  - — Very little, if any, bond length disorder
    - annealing has not, thus far, produced any change in any properties
- $U_3Ni_3Sn_4$ (Ordered NFL)
  - — Very little, if any, structural disorder
- $CeRhIn_5$, $CeIrIn_5$, Ce_2RhIn_8, Ce_2IrIn_8 (Ordered NFL's)
  - — Very little, if any structural disorder

Last words

- **Nature is sneaky: lattice disorder can hide!**
- **For UCu_4Pd, KLDM (Kondo *lattice* disorder model), with no disorder in $N(0)$ is not enough.**
- **Role of disorder still very much unclear!**
  - **Does disorder even matter? *Yes, but it can't explain everything!***
  - **definitely not conventional: either extremely sensitive or it is a minor player**
- **Clustering? Magnetic droplets? Griffiths-McCoy?**
  - **Probably not *exactly* Griffiths-McCoy**
  - **Could be… tough to see structurally**
- **Should doped and undoped systems be treated in the same way?**
  - **I'm leaning toward *yes*…**

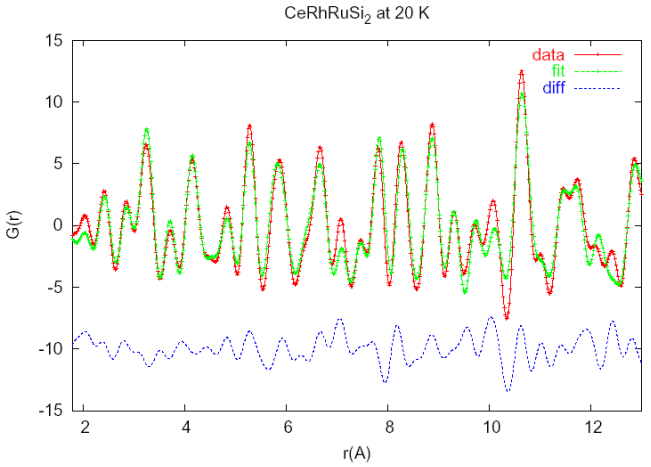


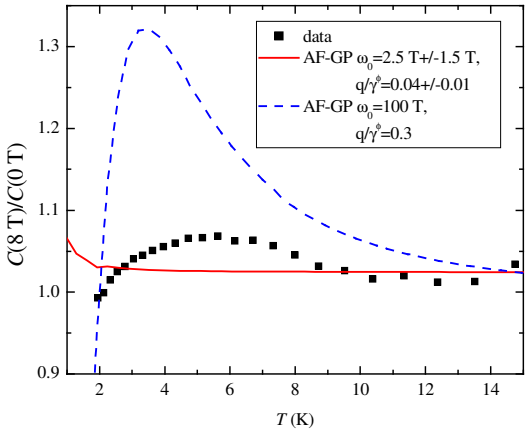




Last words (*continued…*)

- **Competing interaction descriptions seem most appropriate. Is RKKY or Anderson localization the important competing interaction?**
  Structurally, system seems to cross to RKKY, but is close to the boundary!
- **KDM could still work, if something else amplifies the effect of the disorder ("pre-loading").**
- **Even the "canonical" disordered NFL CeRhRuSi$_2$ is remarkably well ordered**
- **Should the disordered and the "well ordered" NFL's be considered as a whole?**
- **$U_3Ni_3Sn_4$ passes all the tests of a well ordered NFL**
- **In addition, FL/NFL development in field appears to not be the product of a Griffiths-McCoy singularity**
- **($U_3Ni_3Sn_4$ behaves similarly to $CeCoIn_5$, another system with a "negative pressure" critical point…)**





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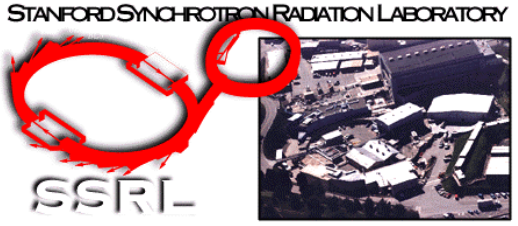
G. H. Kwei (*Lawrence Livermore National Laboratory*)



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