Evolution of the superconducting critical temperature in Yb-substituted $CeCoIn_5$

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IOP, Beijing

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General properties, synthesis

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Thermal conductivity

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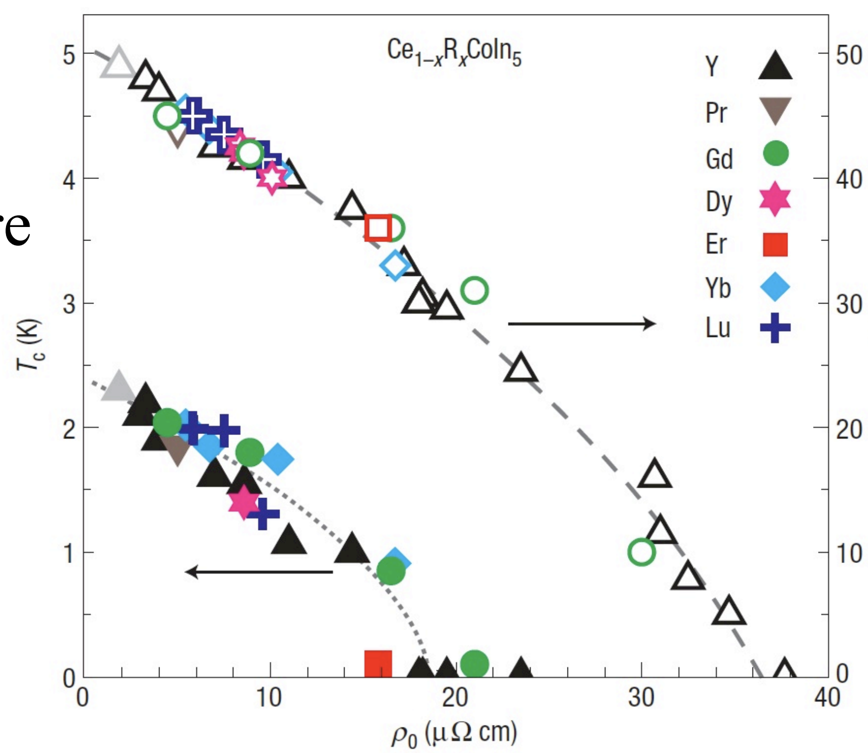
$Ce_{1-x}Yb_xCoIn_5$ – motivation

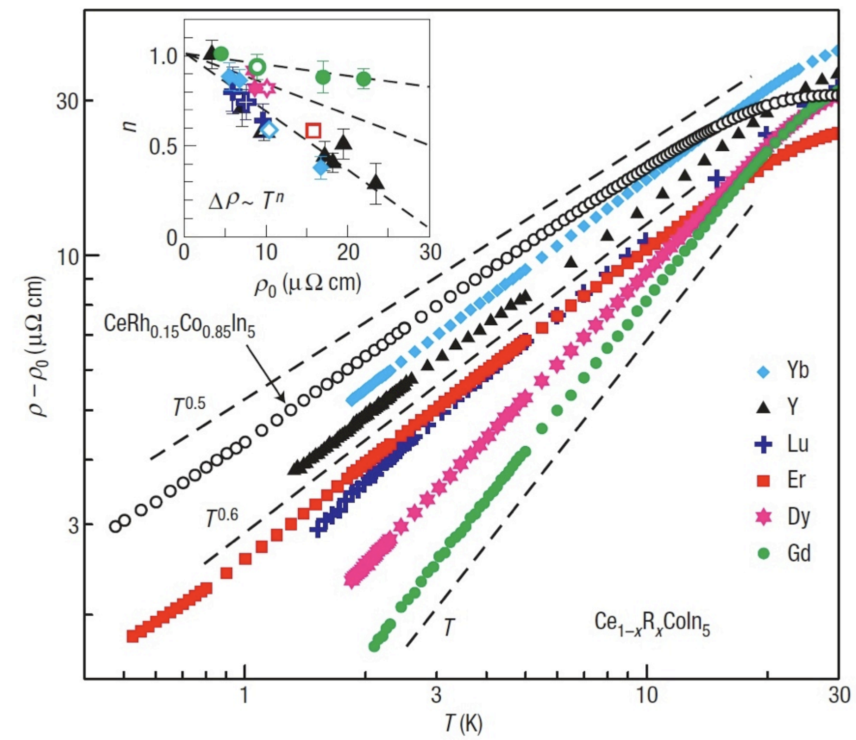
- $CeCoIn_5$:
  1. unconventional HF superconductor (T_c=2.3 K at ambient pressure),
  2. NFL behavior,
  3. magnetic field-induced QCP.

- $YbCoIn_5$: conventional nonmagnetic metal (1 K - 300 K).

- $Ce_{1-x}R_xCoIn_5$ system:
  - Both Cooper pair breaking and Kondo-lattice coherence are uniformly influenced by *R*.
  - NFL behavior is strongly dependent on *R*.

  (J. Paglione *et al*. 2007)





- $Ce_{1-x}Yb_xCoIn_5$ system:
  1. Electron-hole analogy between the $Ce^{3+}(4f^1)$ and $Yb^{3+}(4f^{13})$.
  2. Unstable valence of Ce($3+ \leq v_{Ce} \leq 4+$) and Yb($2+ \leq v_{Yb} \leq 3+$).
  3. Yb is expected to become more magnetic under pressure.

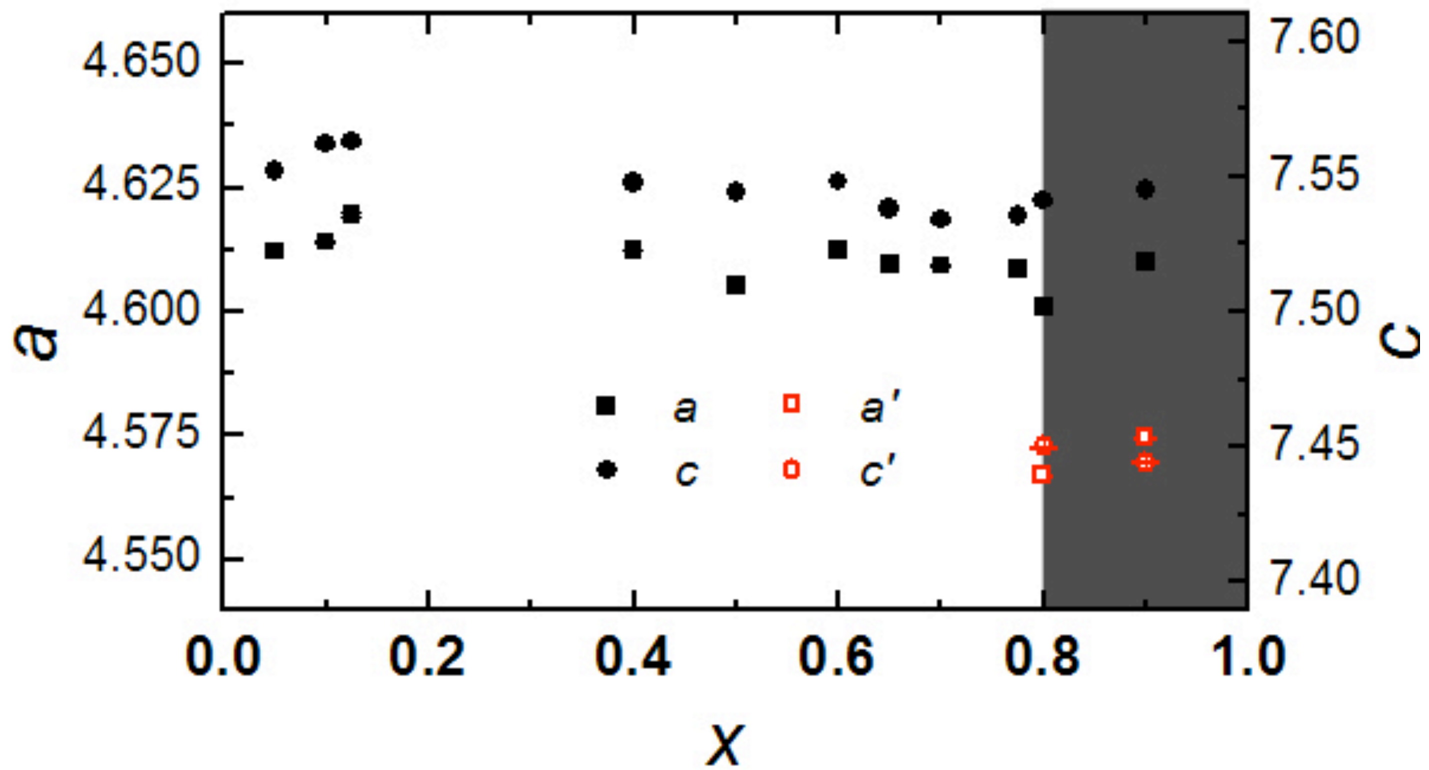
$Ce_{1-x}Yb_xCoIn_5$ – Organization of talk

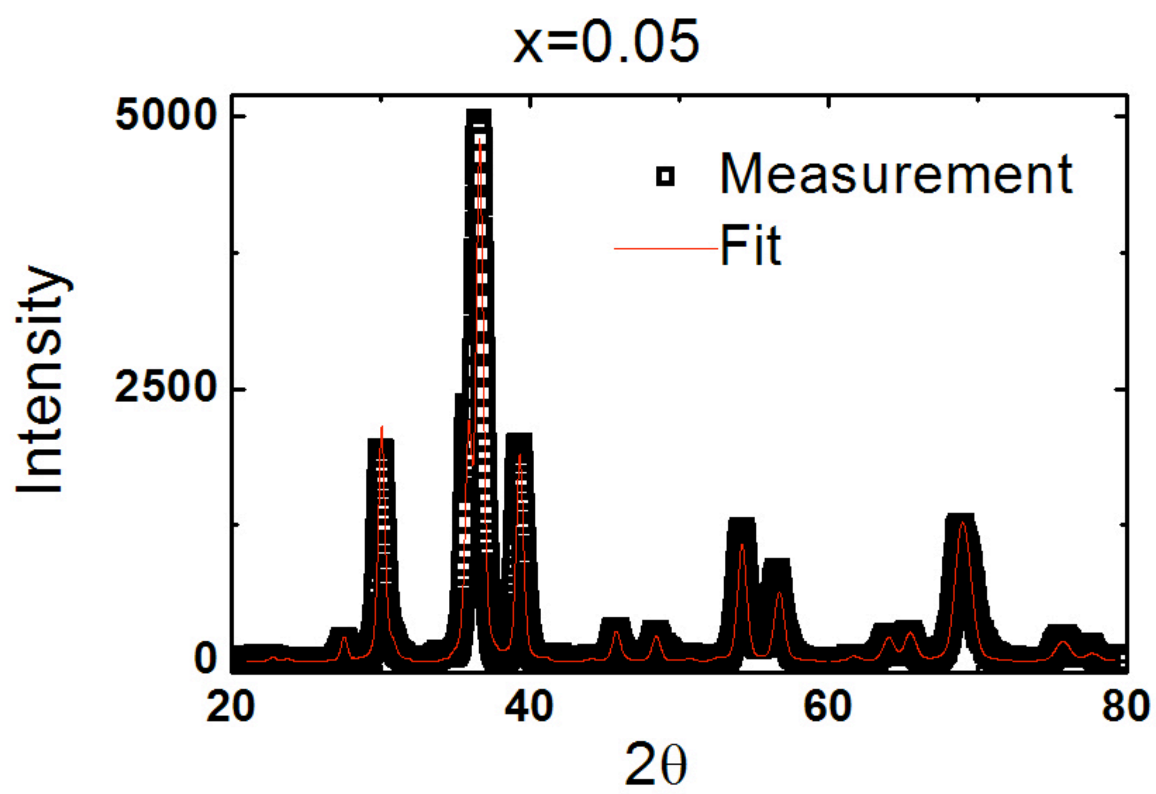
- **Review characteristics of the Yb-stabilized correlated electron state in $Ce_{1-x}Yb_xCoIn_5$**
- **Efforts to determine Ce and Yb valences as a function of x**
- **Evidence for changes in valence and physical properties near x = 0.2**
- **Unusual T_c vs x phase boundary ($T_c \propto$ Ce composition!)**
- **High pressure experiments to probe the normal and SC'ing states**

Publications reporting research of other groups on $Ce_{1-x}Yb_xCoIn_5$:

– C. Capan et al., EPL 92, 46004 (2010)
– C. H. Booth et al., PRB 83, 235117 (2011)
– A. Polyakov et al., PRB 85, 245119 (2012)
– M. Shimozawa et al., PRB 86, 144526 (2012)

Lattice parameters *a* and *c*





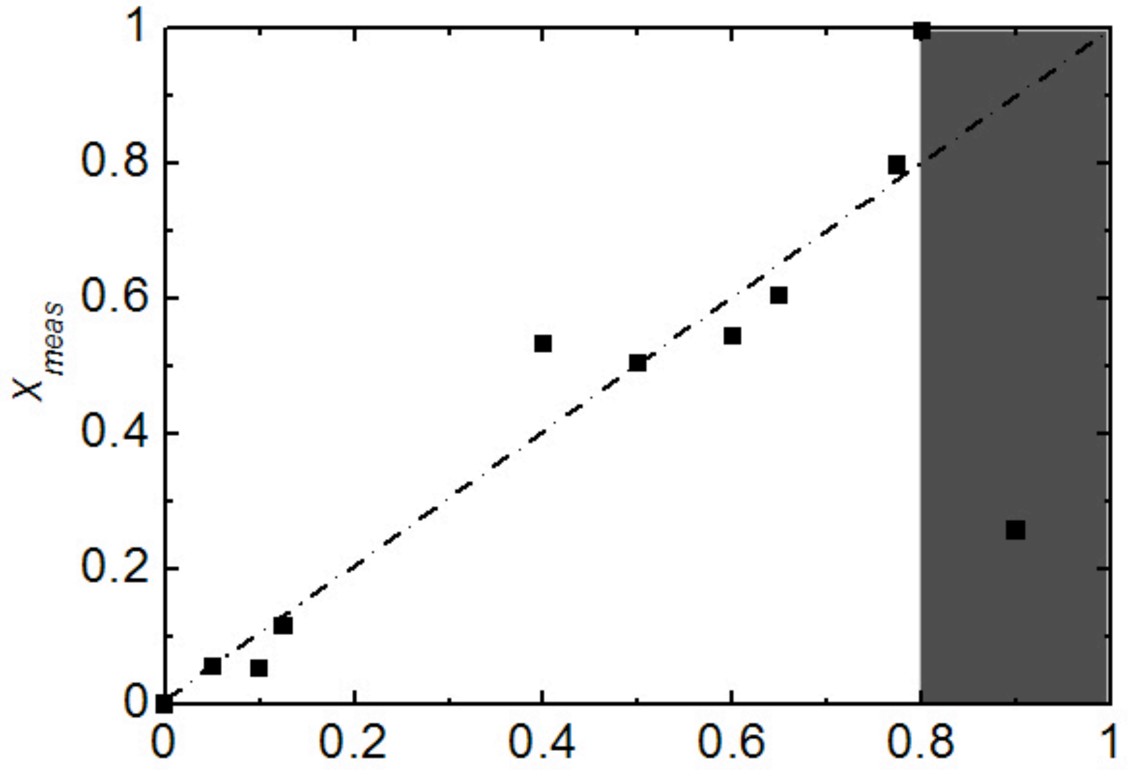
Vegard's Law:
a and *c* should decrease linearly with *x*, **if** there are no changes in

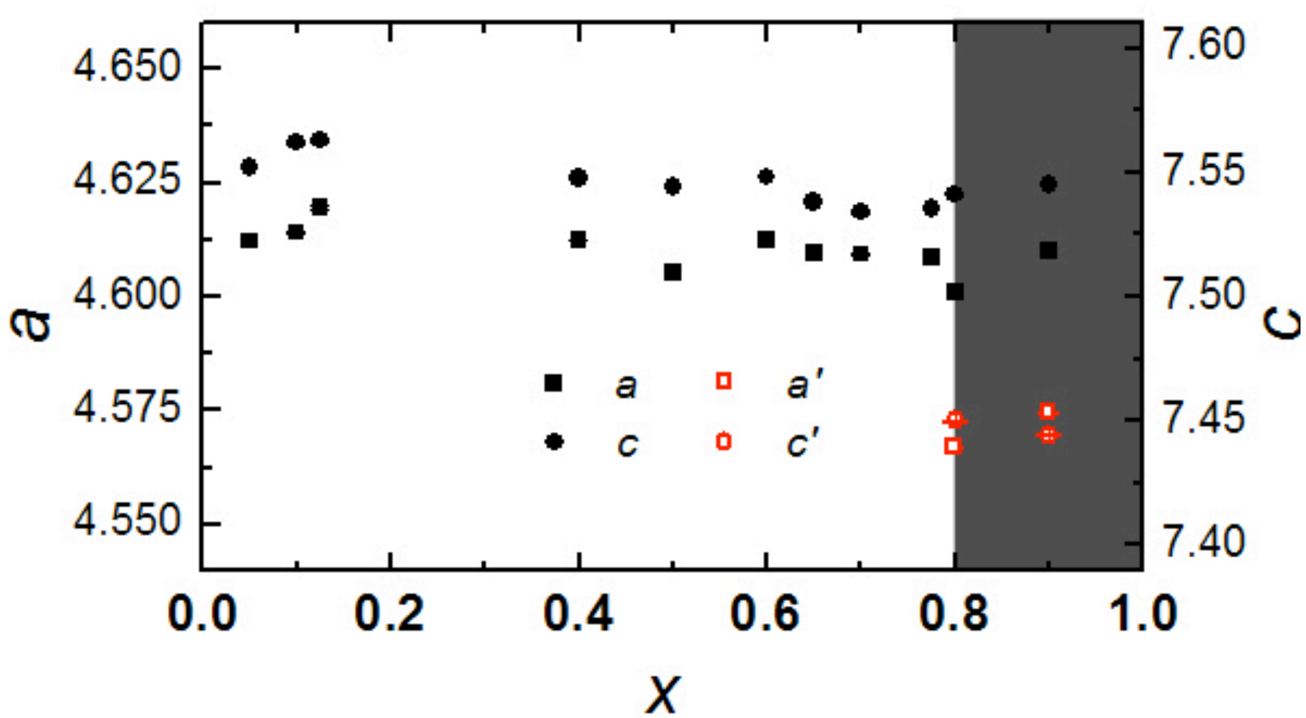
- the valence of the Ce and Yb ions,
- or bonding due to variation in the electron concentration

Ce and Yb ions do not retain
$\nu_{Ce} = 3+$ for x=0
$\nu_{Yb} = 2+$ for x=1

Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)

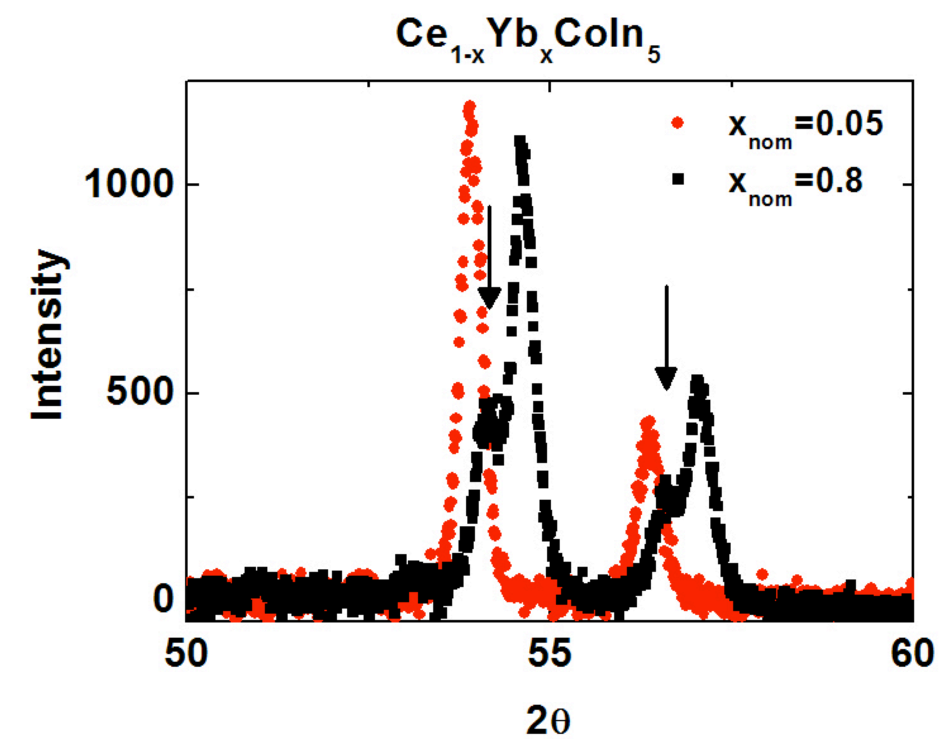
Measured Yb concentration x_{meas} from EDX vs. nominal x





• The EDX data reveal crystals with the expected Yb concentration form for $x < 0.8$,

• For $x \geq 0.8$, each peak in XRD profile splits into two peaks.



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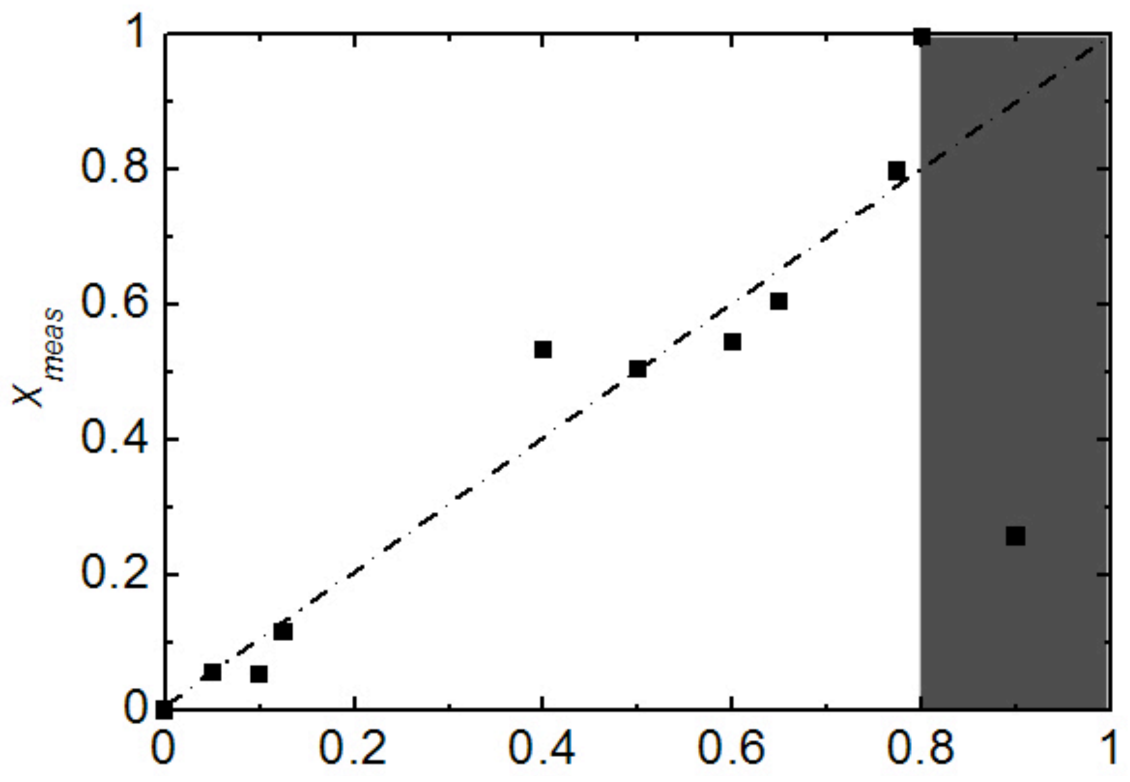

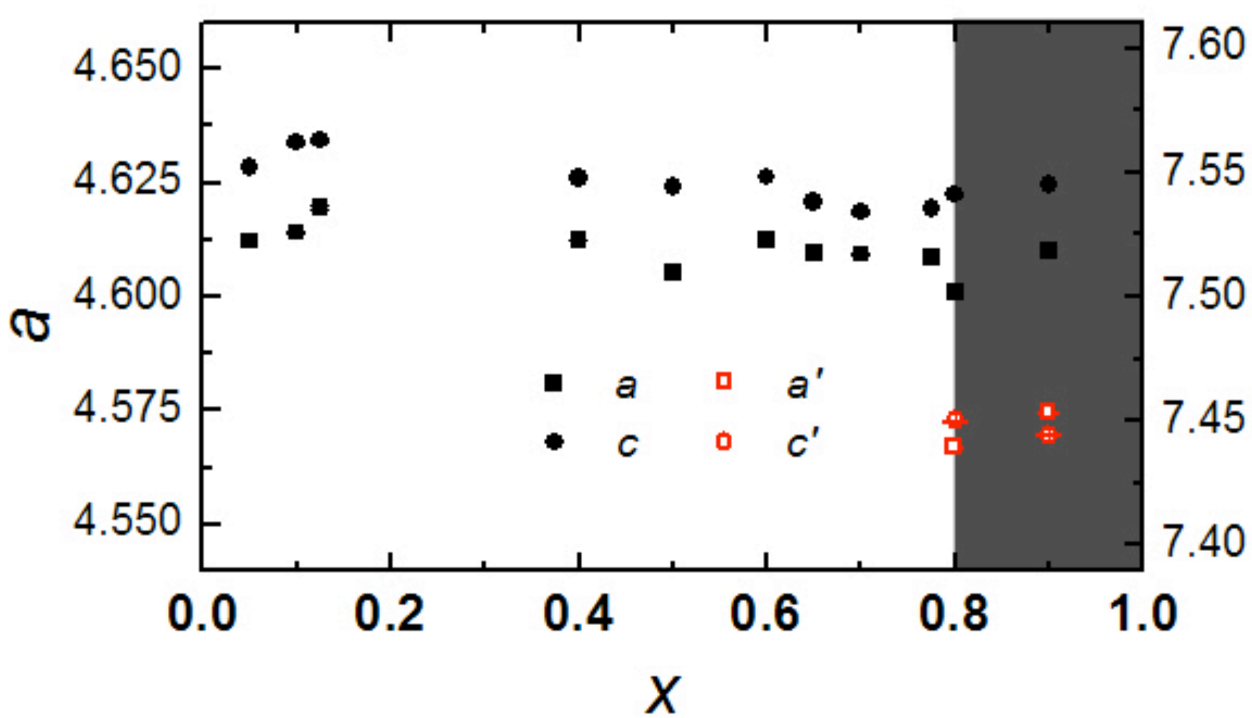

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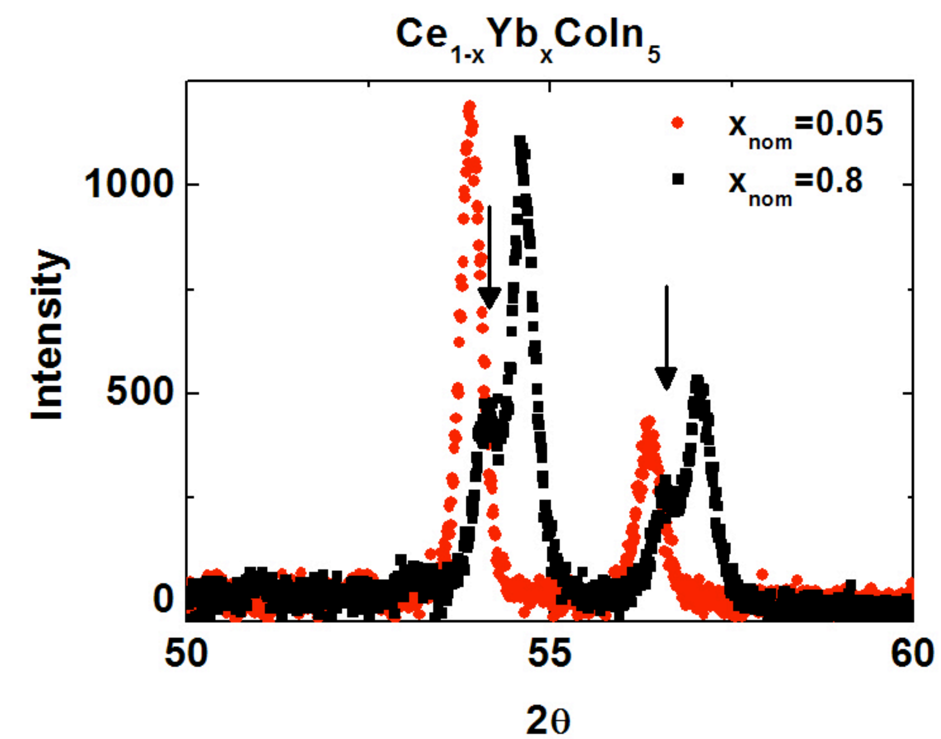
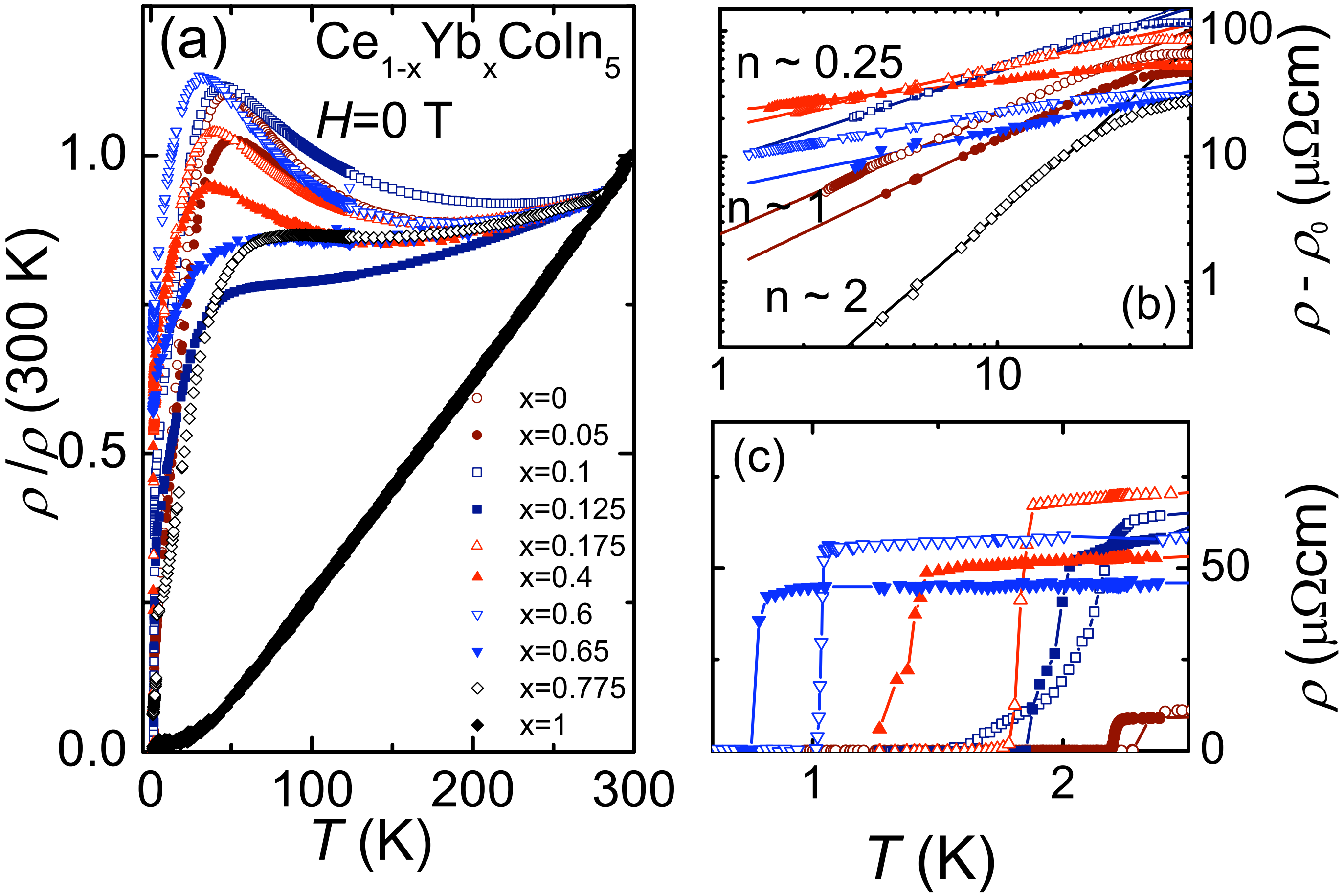


Phase separation occurs for $x \geq 0.8$

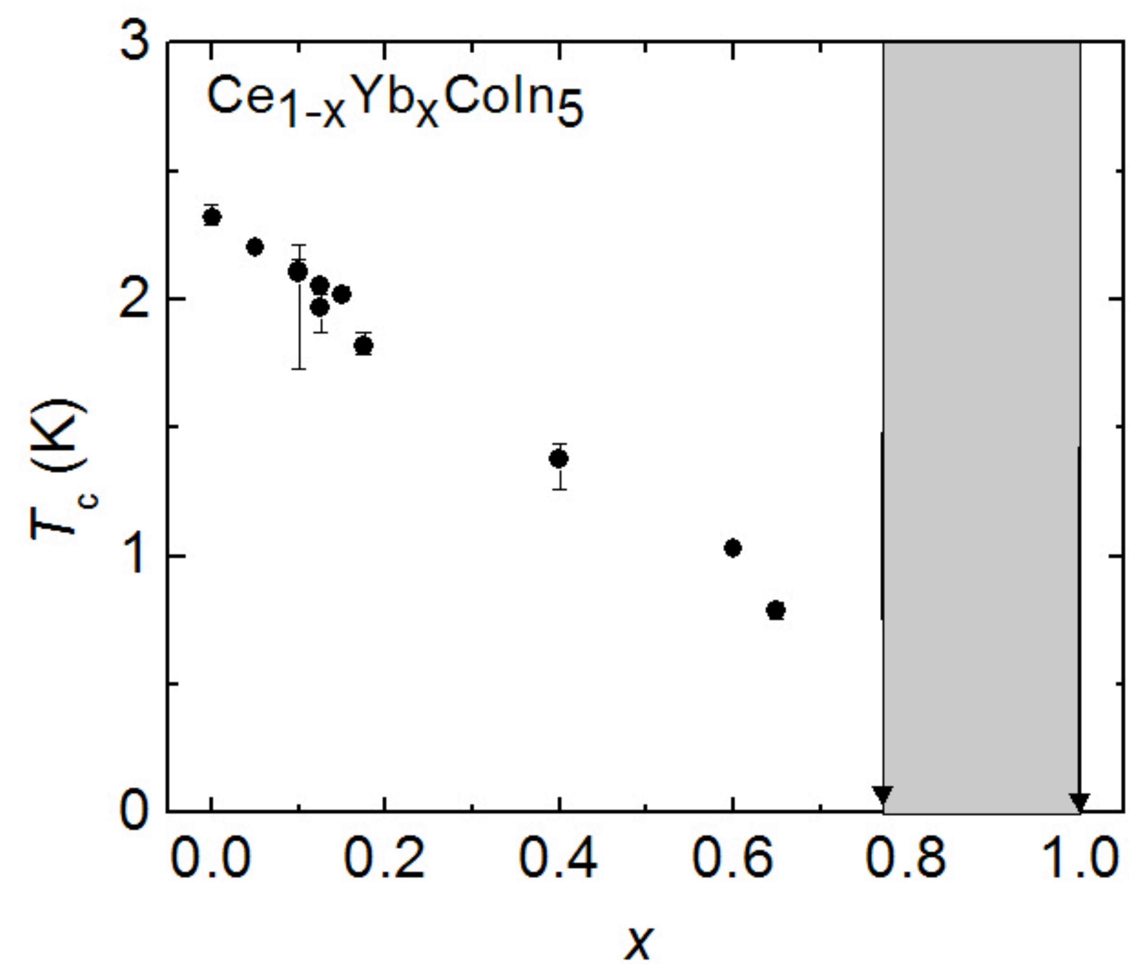
Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)

Electrical Resistivity ρ



For $x \leq 0.775$:
- $\rho(T) = \rho_o + AT^{n_\rho}$ ($T_c < T < 25$K)
- Sub-T-linear transport scattering rate (**NFL behavior**).

SC transitions are clearly observed in $\rho(T)$ for $0 \leq x \leq 0.65$.



Normalized ρ curves for $x \leq 0.775$ are typical of many HF materials:

- weak T dependence at high T;
- a maximum or broad hump at T^*;
- followed by a decrease in ρ with decreasing T.

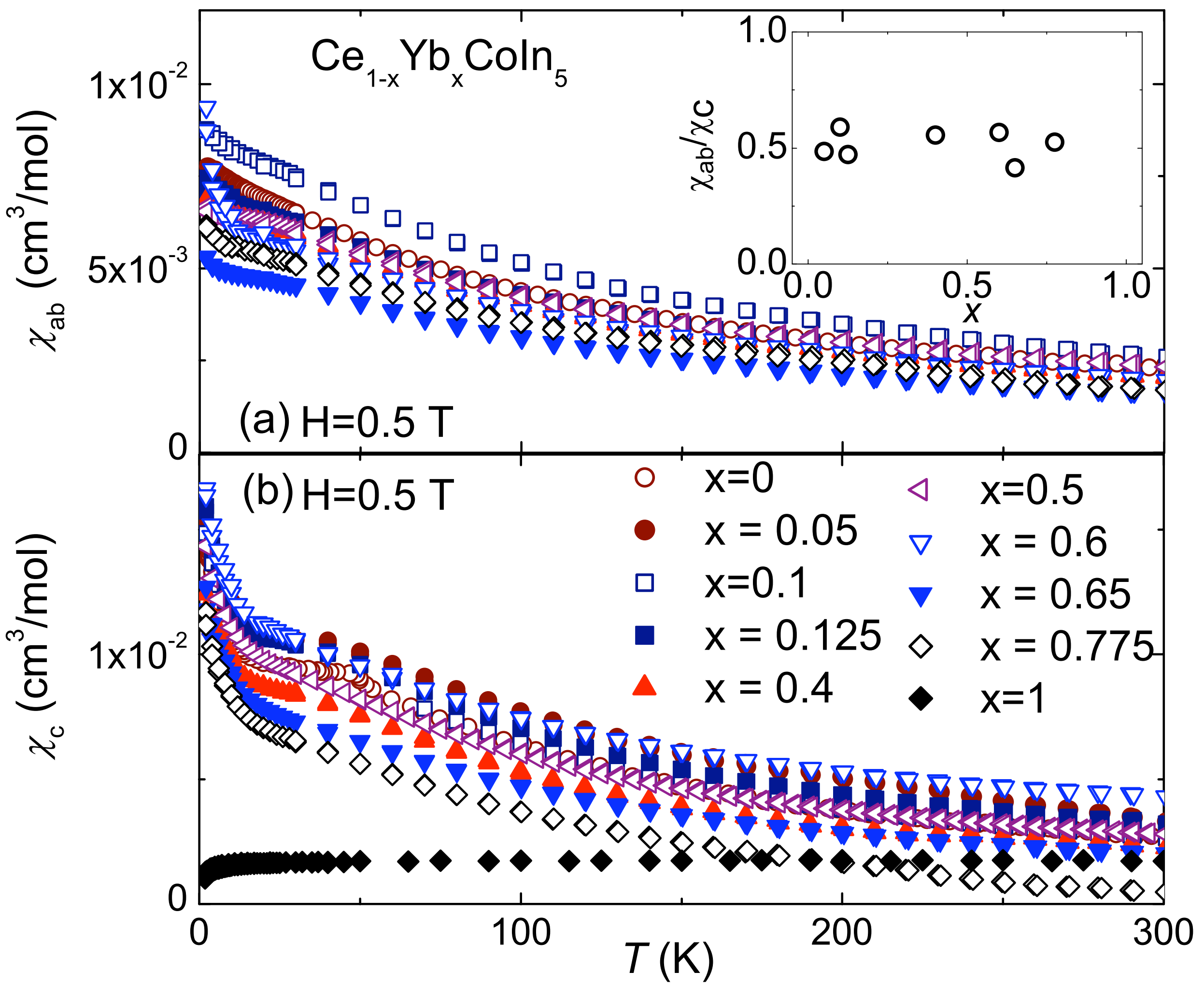
T^* remains roughly constant.

$YbCoIn_5$ does not exhibit correlated electron effects.

For other $Ce_{1-x}R_xCoIn_5$, SC is suppressed at $x \approx 0.25$.

Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)

Magnetic susceptibility χ



For $x \leq 0.775$, $\chi(T)$ is nearly identical to that of $x = 0$:

1) Curie-Weiss behavior at high T.
2) $\chi(T)$ saturates below 50 K (consistent with the coherent behavior in $\rho(T)$).
3) $\chi(T)$ increases upon cooling below 20 K (intrinsic effect), contrary to the behavior of ideal HF compounds. $\chi_c = \chi_c(0) + a/T^{n_\chi}$ (1.8 K < T < 20 K) **NFL behavior**

Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)

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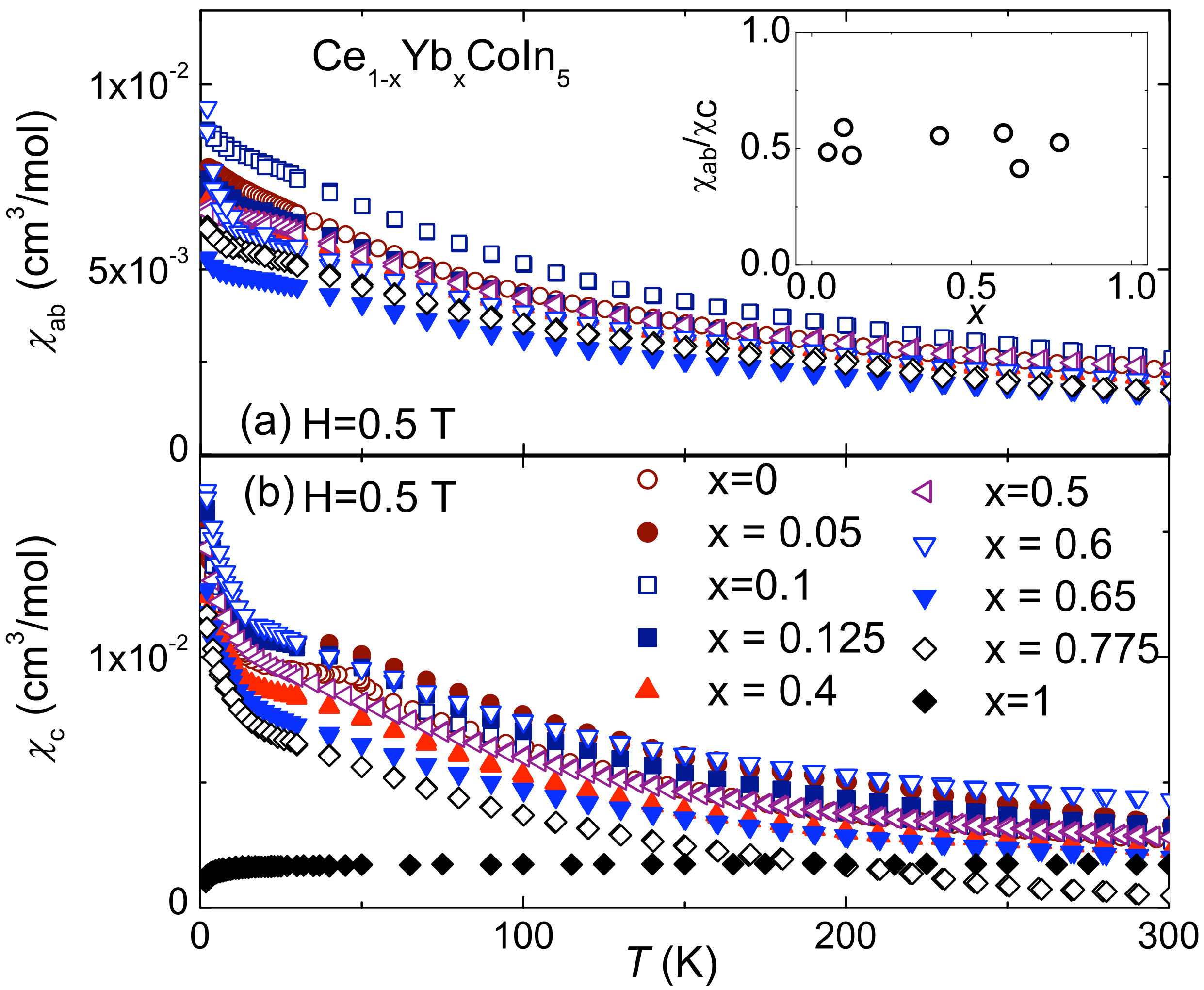

Yb ions does not enter the lattice in the nonmagnetic divalent state, in which case χ(*T*) should scale with (1-*x*).

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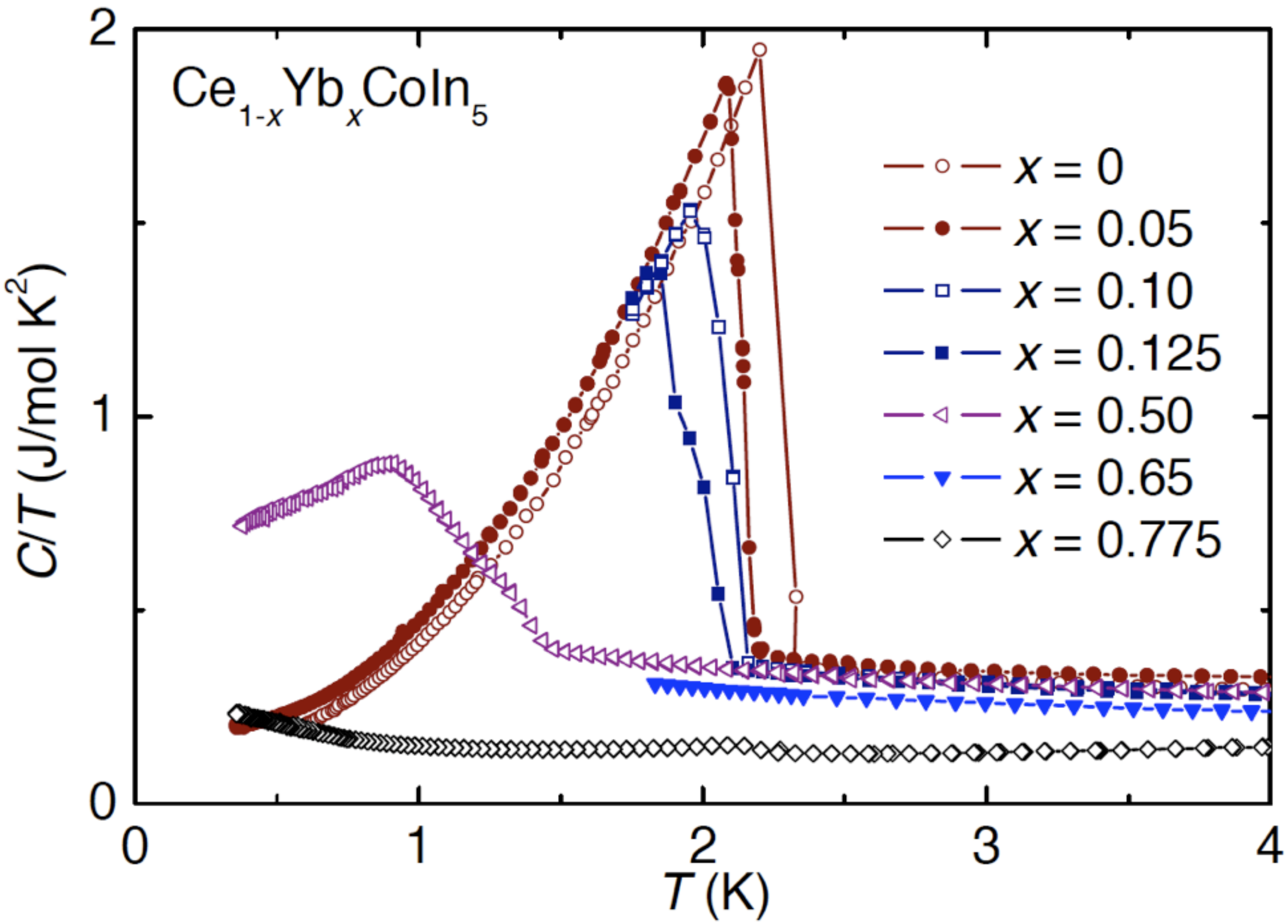
Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)

Specific heat



For x ≤ 0.775, C/T tends to increase with decreasing T down to the SC transition (**NFL)**

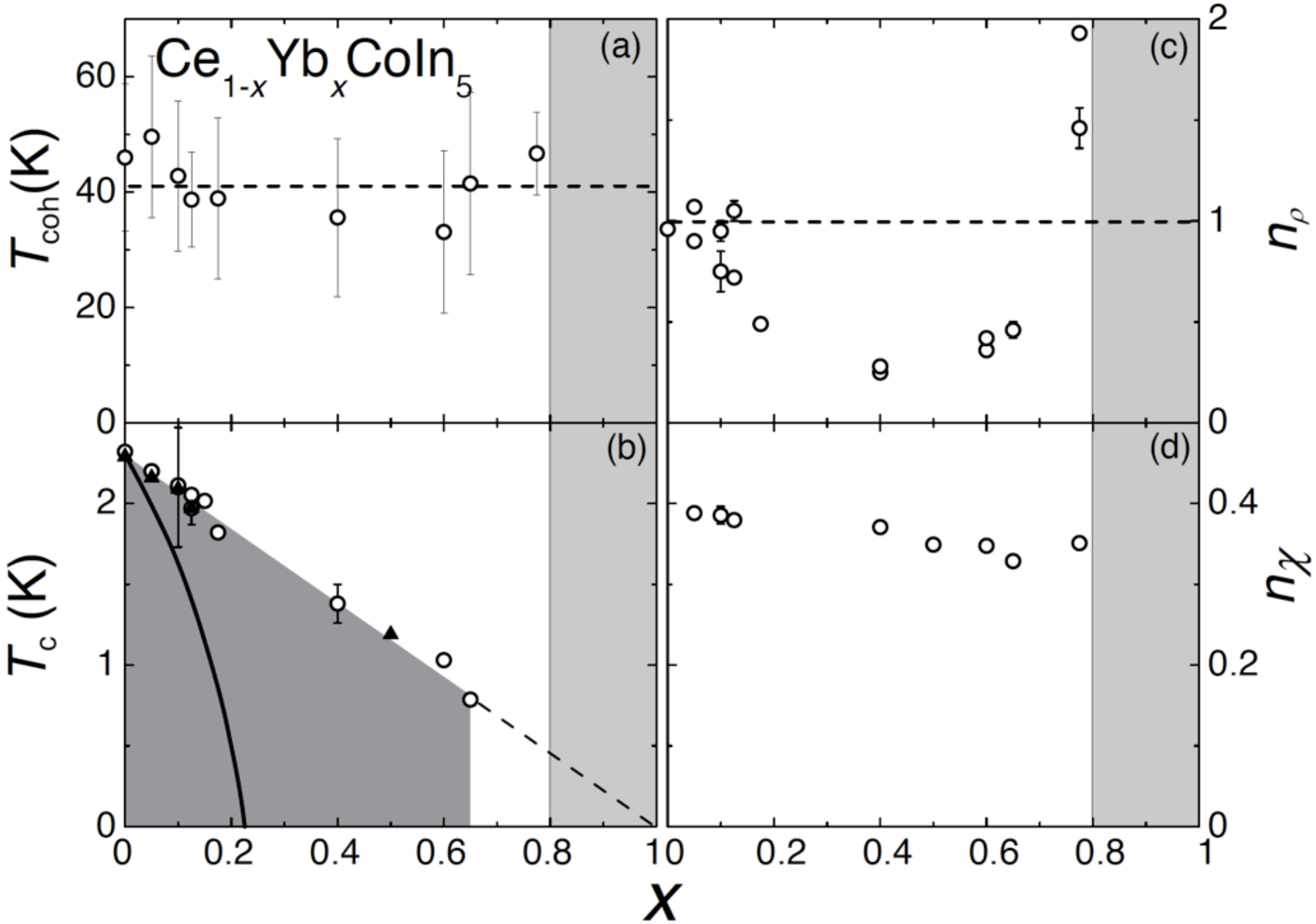
For x = 0.775,

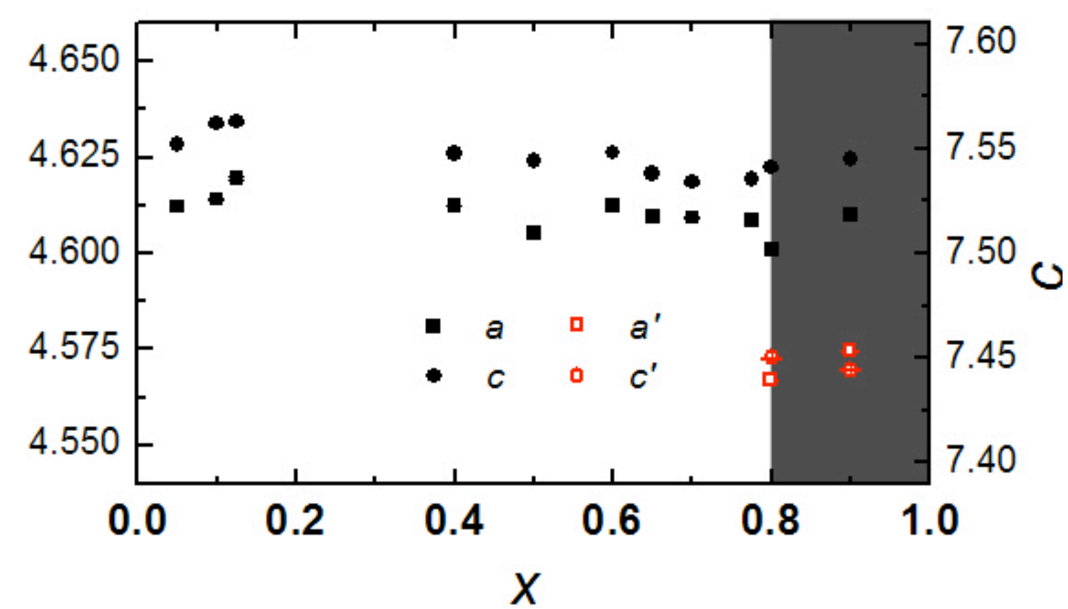
- $A \approx 0.036\ \mu\Omega cm/K^2$ (ground state is a heavy Fermi liquid), $\rho = \rho_o + AT^2$ ($T_c \leq T$ ~25K).
- Kadowaki-Woods ratio $R_{KW} = A/\Upsilon^2 = 1.86 \times 10^{-6}\ \mu\Omega cm(mol\text{-}K/mJ)^2$, intermediate between what is expected for Ce- and Yb- based heavy fermion compounds (Kadowaki and Woods 1986, Tsujii *et al*. 2005), emphasizing that **strong electronic correlations persist up to $x \approx 0.775$.**

Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)

Our study reveals that:

1) a and c remain nearly constant for $x \leq 0.775$, phase separation occurs when $x > 0.775$.
2) T_c is weakly suppressed with x. SC would disappear near $x = 1$ in the absence of phase separation.
3) T^* remains roughly constant up to x=0.775. T_c does not scale with T^*.
4) Strong electronic correlation persists up to x=0.775.
5) The NFL behavior is strongly influenced by x, a recovery of FL-like behavior is observed with increasing x. No apparent QCP.





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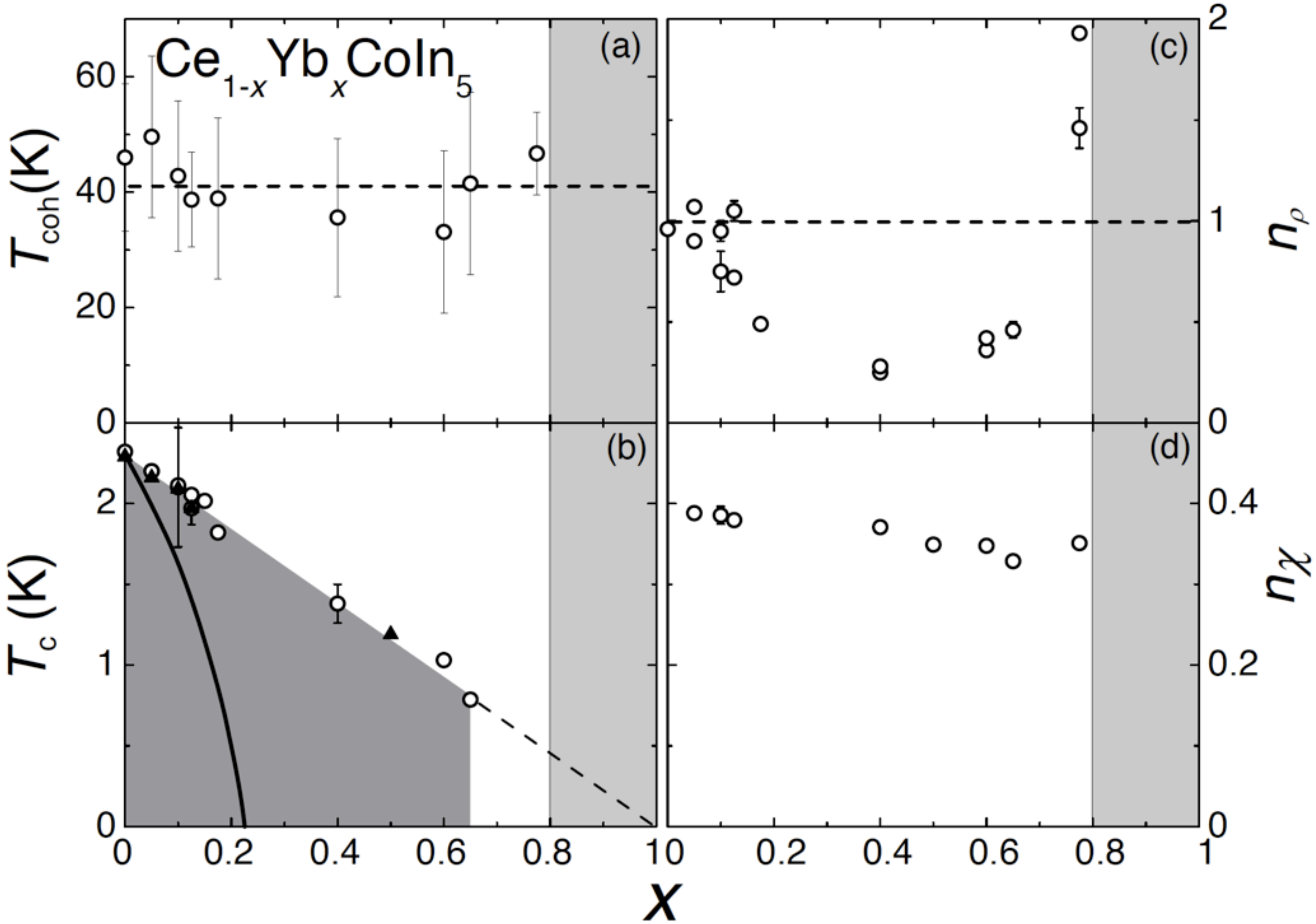
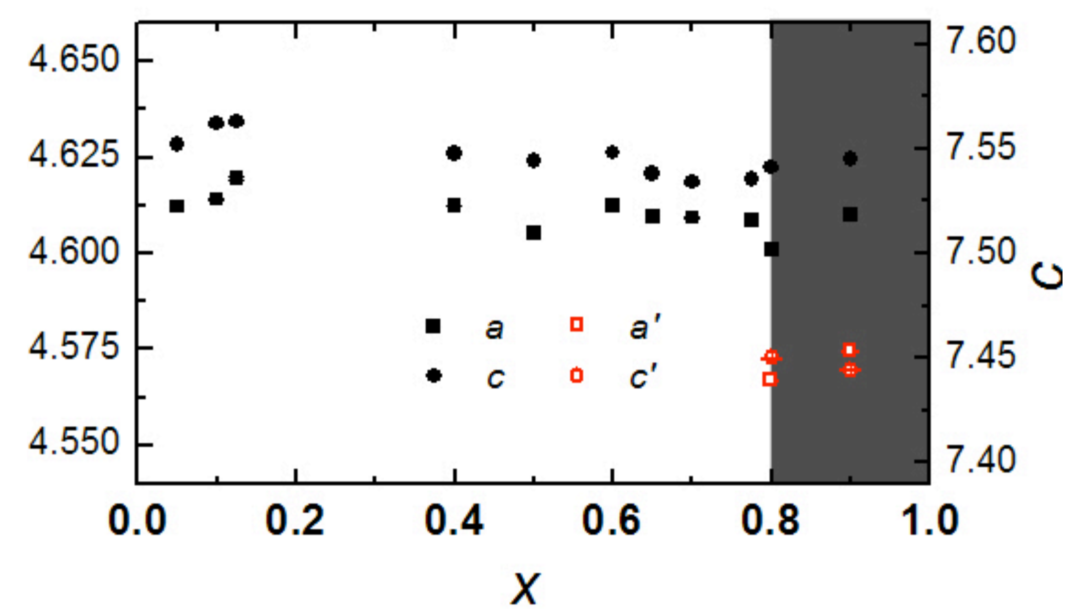




The correlated electron effects in $CeCoIn_5$ are only weakly affected by Yb substitution!

Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)

Ce and Yb cooperatively change their electronic states to preserve the Kondo-like lattice behavior and SC of $CeCoIn_5$. NFL state is strongly susceptible to the introduction of Yb ions.

A possible explanation: valence fluctuations arising from a cooperative IV state formed by the Ce and Yb ions, which stabilizes the electronic properties of $Ce_{1-x}YbCoIn_5$. The cooperative IV state provides a mechanism that may drive the observed NFL physics.

Note: Quantum valence criticality yields NFL-like anomalies

Watanabe and Miyake. 2010
Okada and Miyake. 2011

β-$YbAlB_4$ and $YbRh_2(Si_{0.95}Ge_{0.05})_2$: $C/T \sim -lnT$ (low T)
$\chi \sim T^{-n_\chi}$, n_χ = 0.5-0.6
$\Delta\rho \sim T$

Custers *et al*. 2003
Nakatsuji *et al*. 2008

$CeCu_2(Si_{1-x}Ge_x)_2$ Yuan *et al*. 2006

Ce and Yb Valence vs x

Valences of Ce and Yb in $Ce_{1-x}Yb_xCoIn_5$

L_{III} edge XANES

Ce: $v_{Ce} \approx +3$ for all x

Yb: $v_{Yb} \approx +2.3$ for all $0.2 \leq x \leq 1$

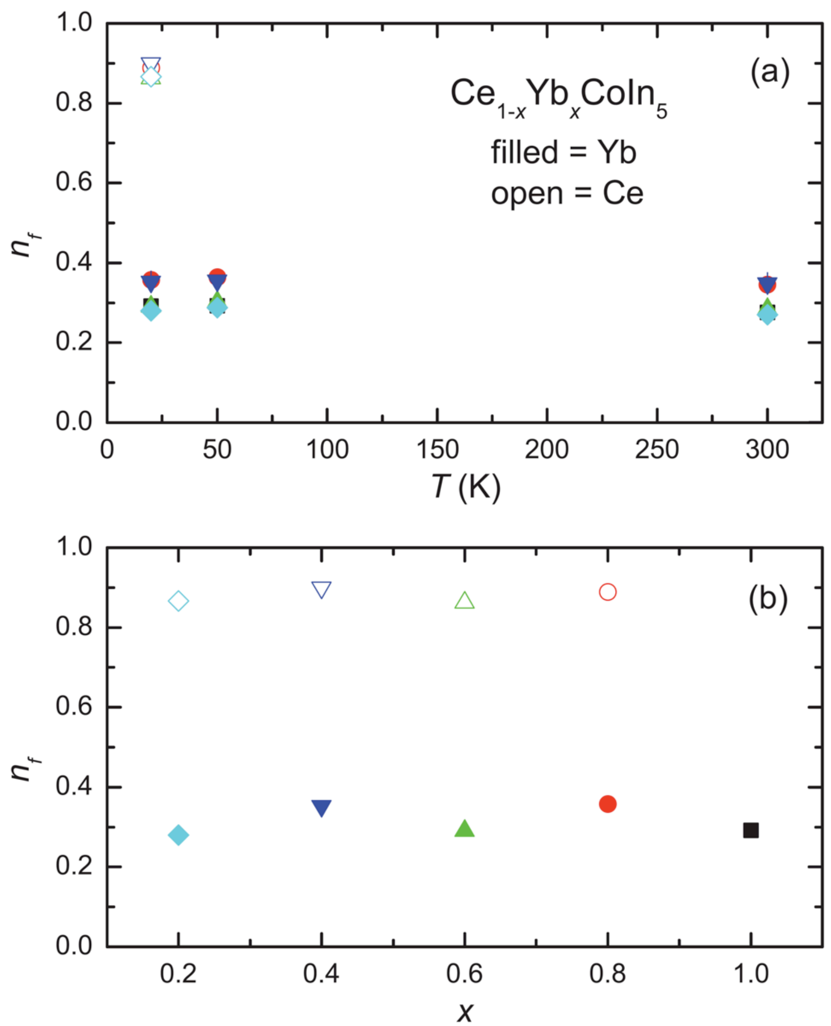
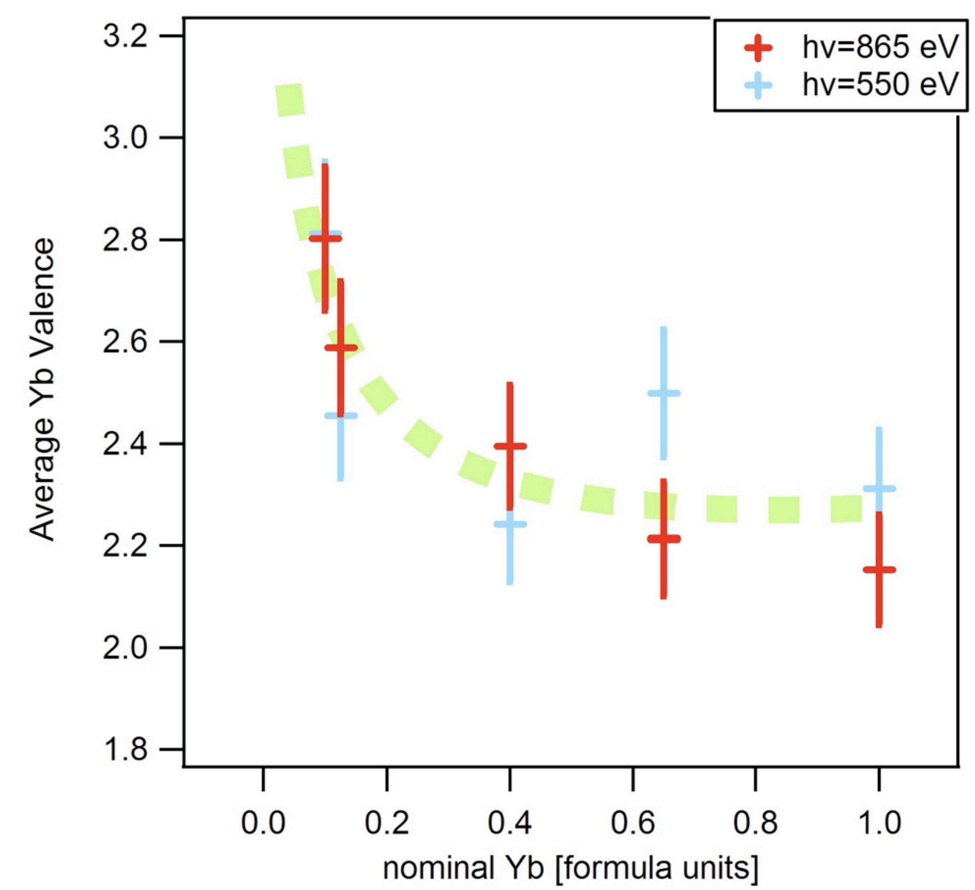


FIG. 8. (Color online) Fit results for n_f as a function of (a) temperature and (b) x, for both Ce and Yb orbitals. Note that n_f refers to the f-electron orbital occupancy for Ce and the f-hole orbital occupancy for Yb. The rare-earth valence is then $v = n_f + 3$ for Ce and $v = n_f + 2$ for Yb.

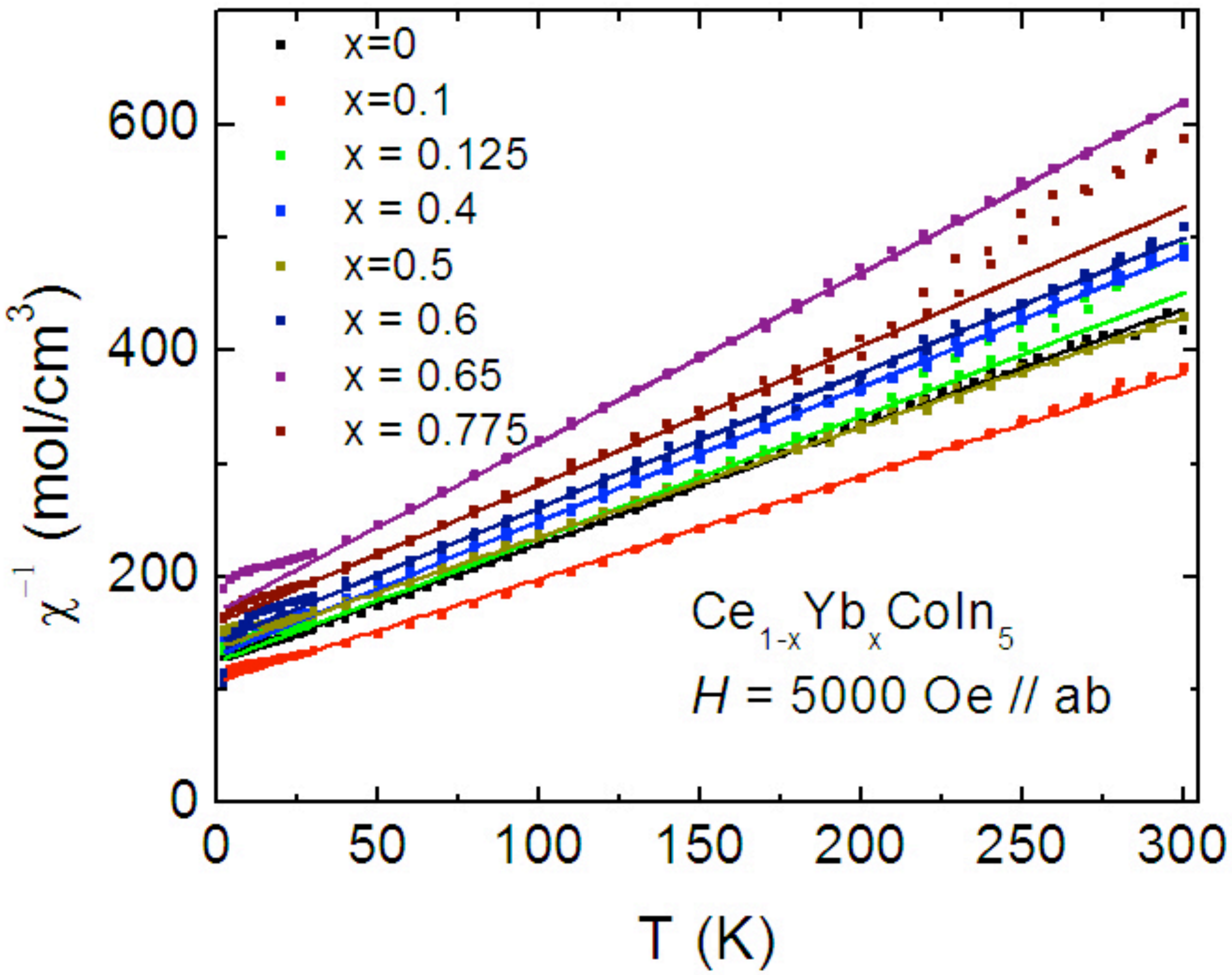
Booth *et. al*. PRB **83**, 235117 (2011)

- XAS near Ce M_4 and M_5 edges
  Ce: $v_{Ce} \approx +3$ for all x
- 4f XPS
  Yb: v_{Yb} drops from ~+3 at x=0 to ~+2.3 at x=0.2, then remains ~constant to x=1
- Yb valence transition below x ≈ 0.2



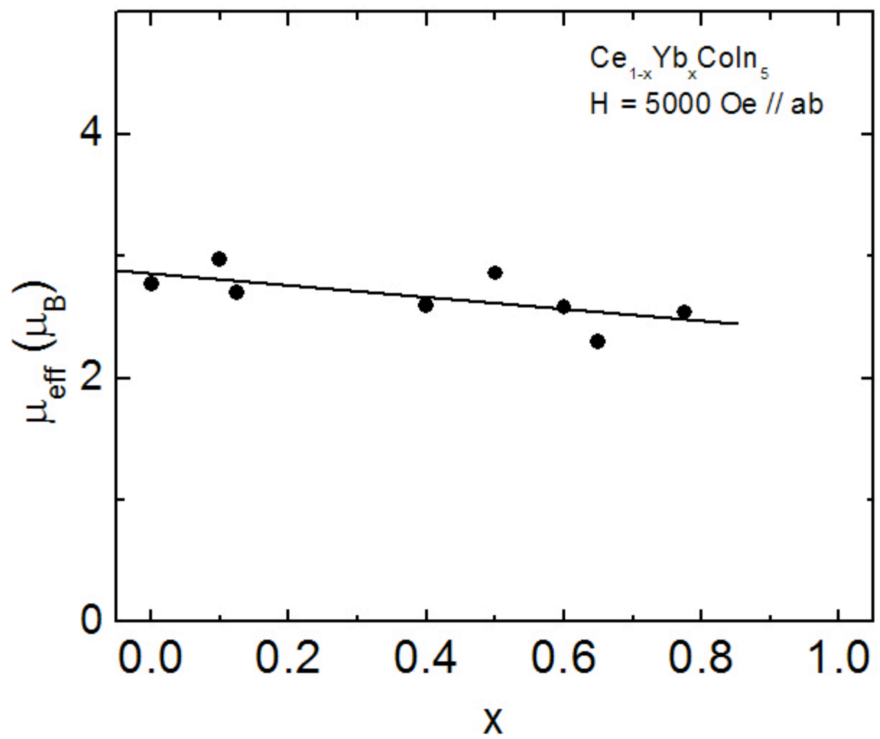
Dudy et al., manuscript in preparation (2012)

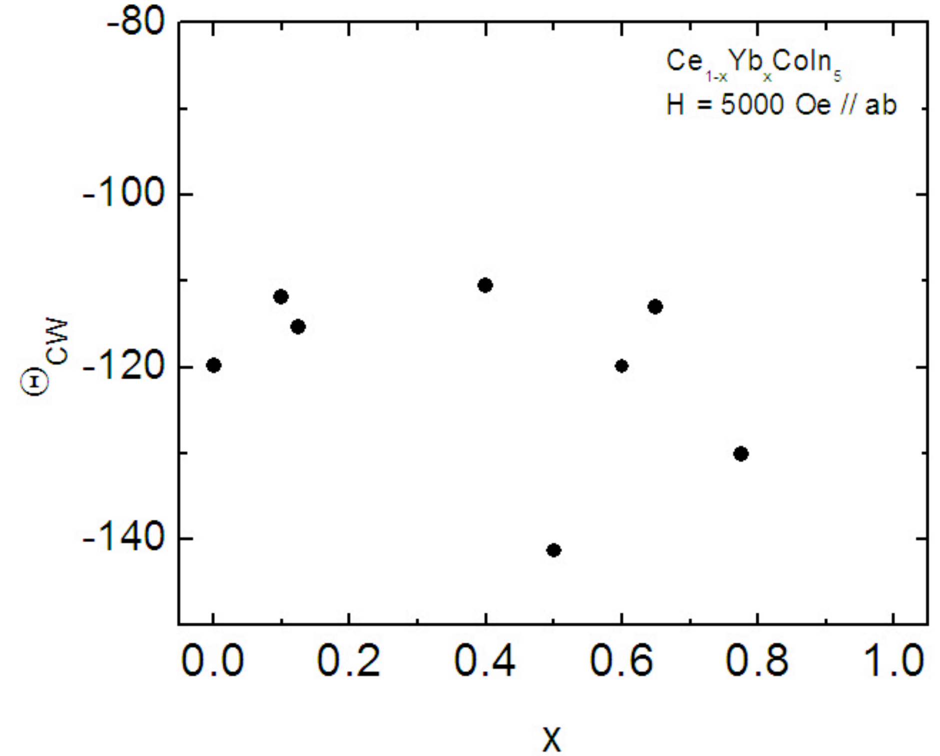
Yb valence from χ(x,T) measurements on $Ce_{1-x}Yb_xCoIn_5$



Straight lines above are the Curie-Weiss fits. Fitting parameters μ_{eff} and Θ are indicated in the figs on right side.

$$\chi = \frac{N_A \mu_{eff}^2}{3k_B(T - \Theta_{CW})}$$





Yb valence from $\chi(x,T)$ measurements on $Ce_{1-x}Yb_xCoIn_5$

- If Θ_{CW} is constant, then

$$\mu_{eff}^2(x) = \mu_{Ce}^2(1-x) + \mu_{Yb}^2(x) \quad (1)$$

- XPS, XANES measurements indicate

$v_{Ce} \approx +3$ for all x $\Rightarrow$ $\mu_{Ce} \approx 2.54\ \mu B$

- Solve Eq. (1) for $\mu_{Yb}(x)$ from $\mu_{eff}(x)$

$$V_{\mathrm{Yb}}(x) = \mu_{\mathrm{Yb}}(x)^2/\mu_{\mathrm{Yb3+}}^2 + 2 \quad (2)$$

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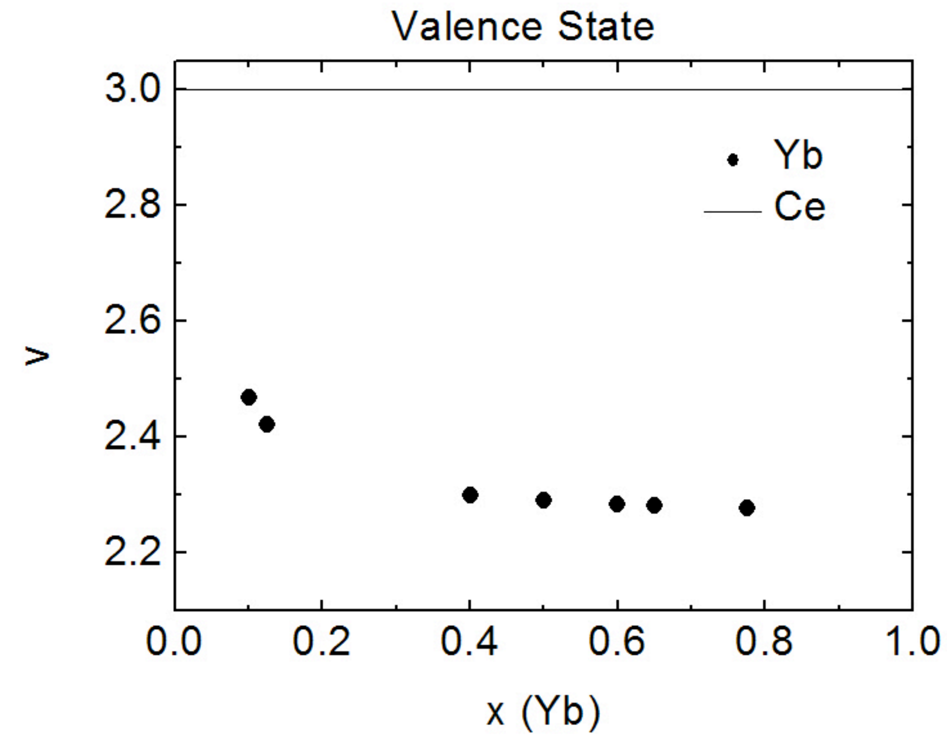

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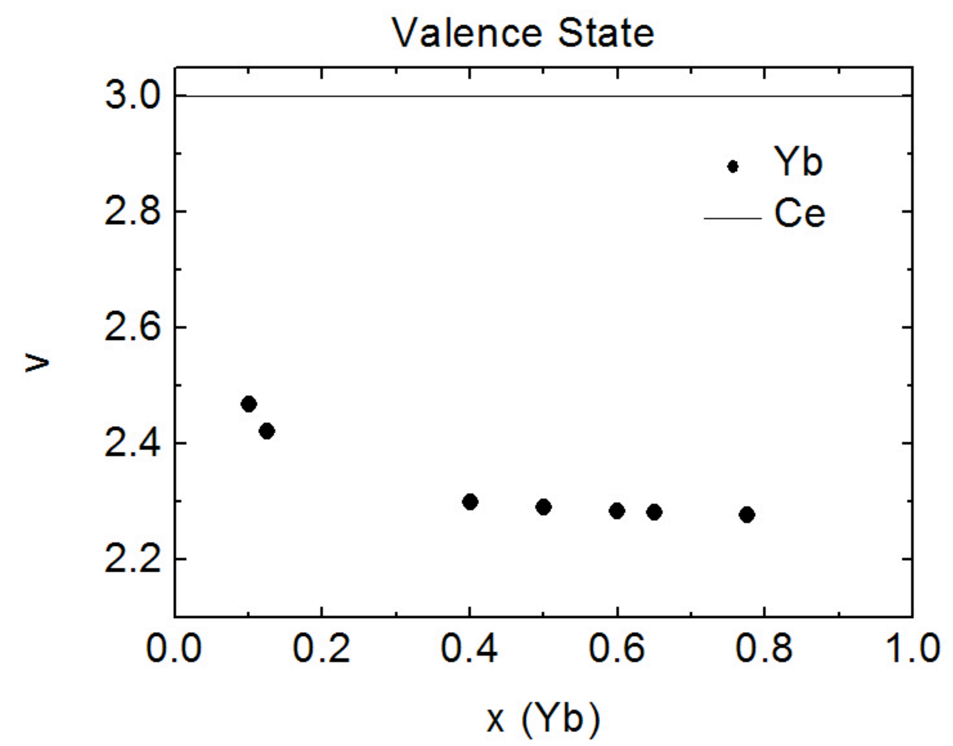

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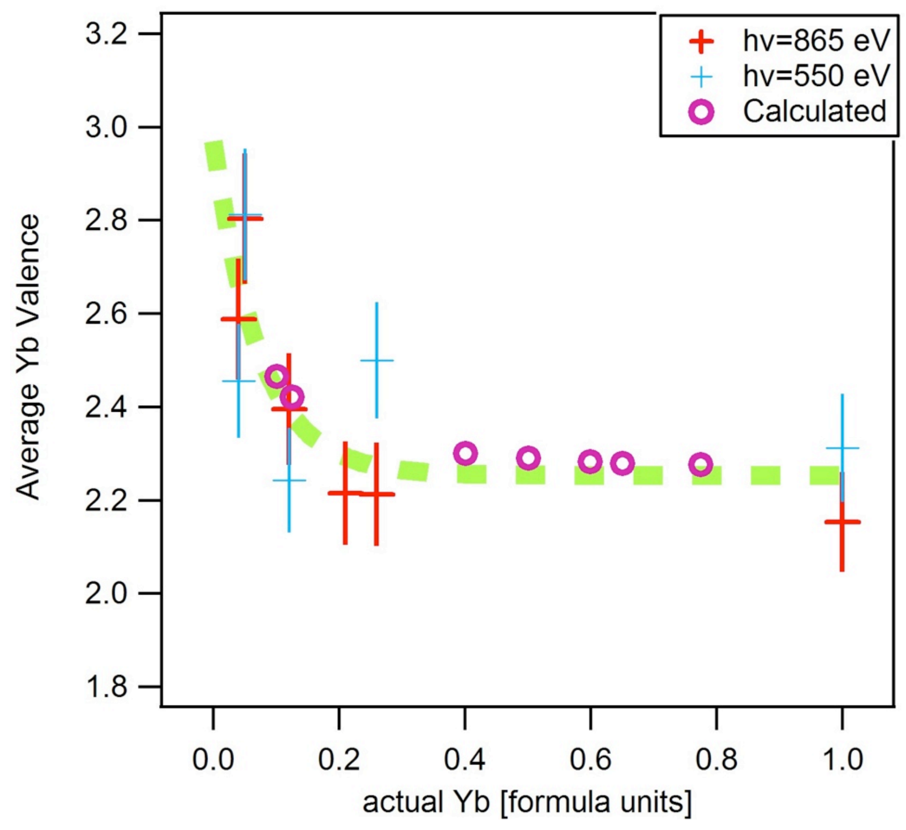
Comparison with XPS data of Dudy *et al*

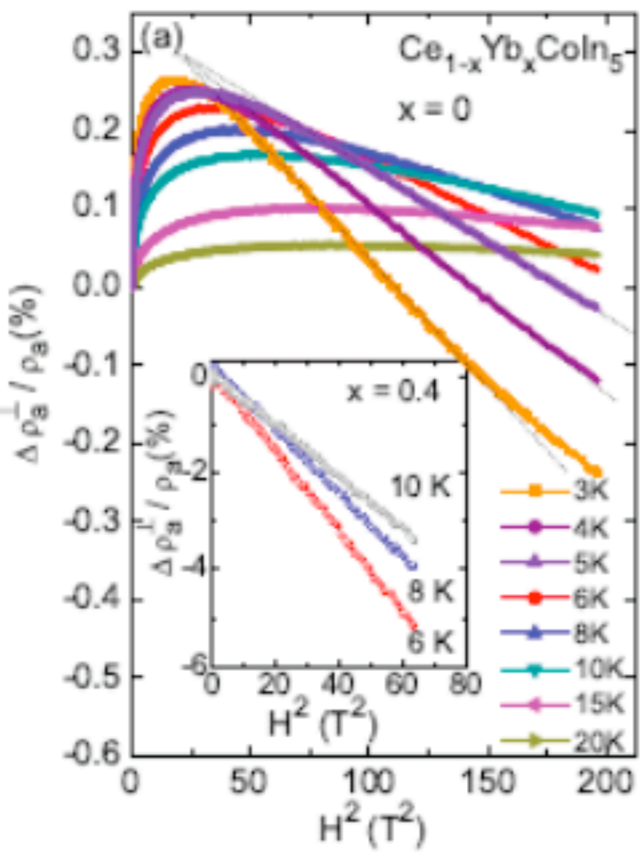
Note: the analysis does not account for CEF and valence fluctuation or Kondo effect modifications of chi(*T*) explicitly. It only assumes we can analyze χ(*T*) at high *T* in terms of a Curie-Weiss law and use effective moments to infer the valence. Could also be large errors (Curie-Weiss *T* are large in magnitude and some CEF splittings may be appreciable). Nonetheless, the analysis seems to be, at least, qualitatively consistent with XPS data.

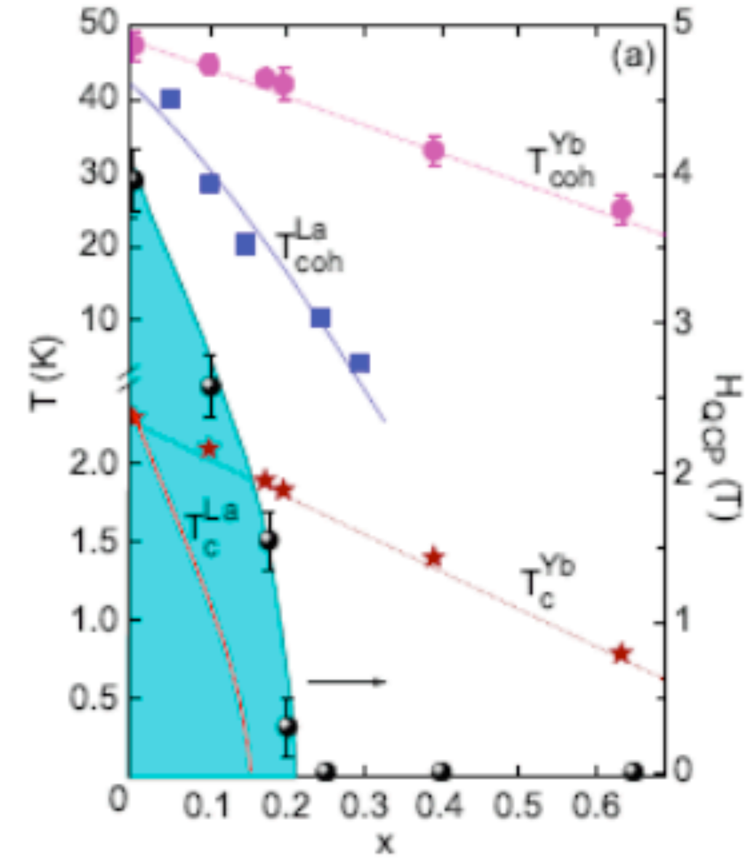


Change in electronic properties at $x \approx 0.2$

$Ce_{1-x}Yb_xCoIn_5$: Quantum criticality

- Quantum critical field $H_{QCP}(x)$ determined from analysis of magnetoresistance
- $H_{QCP} \rightarrow 0$ at $x \approx 0.2 \Rightarrow$ NFL behavior and unconventional SC associated with (a) quantum criticality for $x < \sim 0.2$; (b) another mechanism for $x > \sim 0.2$ (VF's?)
- Character of NFL behavior in $\rho(T)$ also changes near $x \approx 0.2$



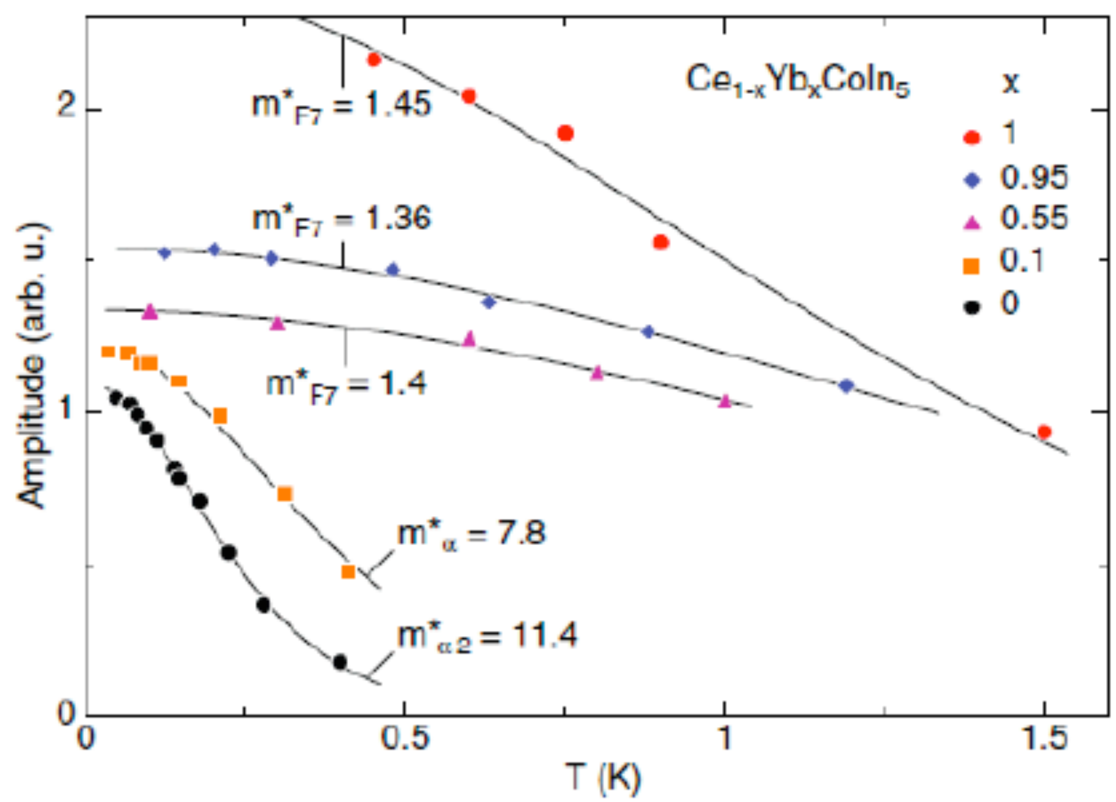


T. Hu, Y. P. Singh, L. Shu, M. Janoschek, M. Dzero, M. B. Maple, C. C. Almasan, archive:1208.4308.

$Ce_{1-x}Yb_xCoIn_5$: Evolution of Fermi surface

dHvA studies of $Ce_{1-x}Yb_xCoIn_5$

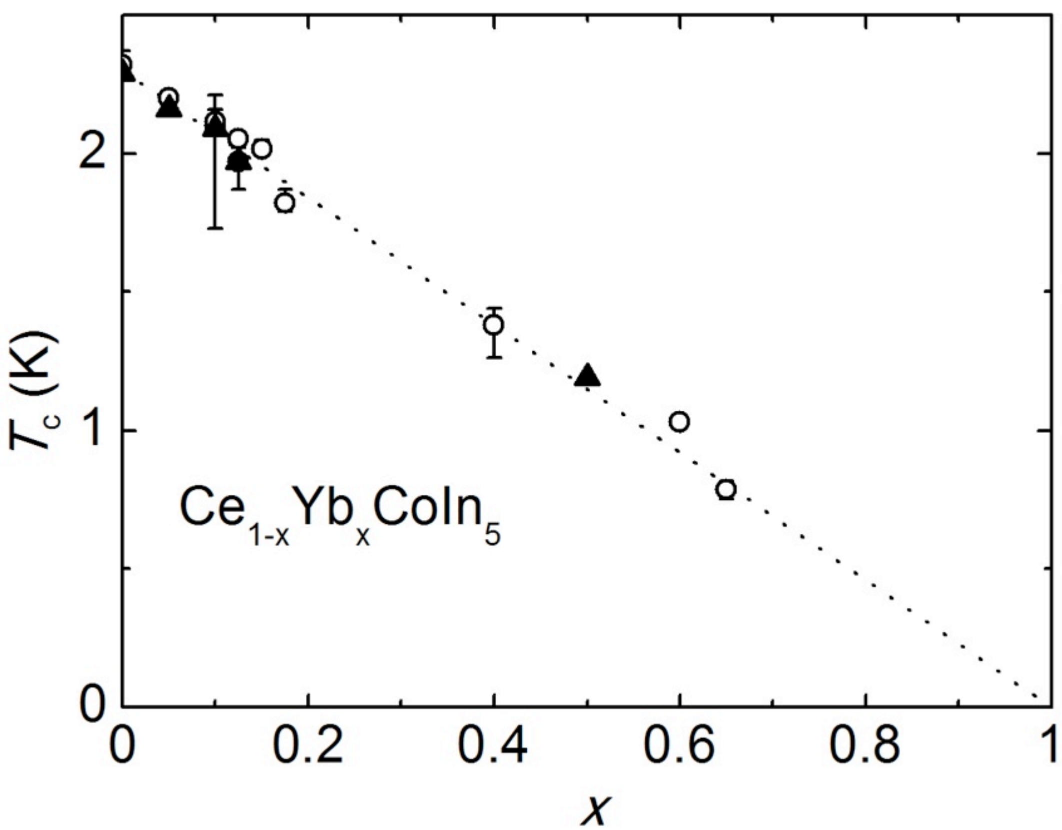
- x = 0.1: Slight change in heavy effective mass m* of $CeCoIn_5$ (x = 0)
- x = 0.2: Changes in FS topology
- x = 0.55 and above: Drastic reconstruction in FS and small effective masses



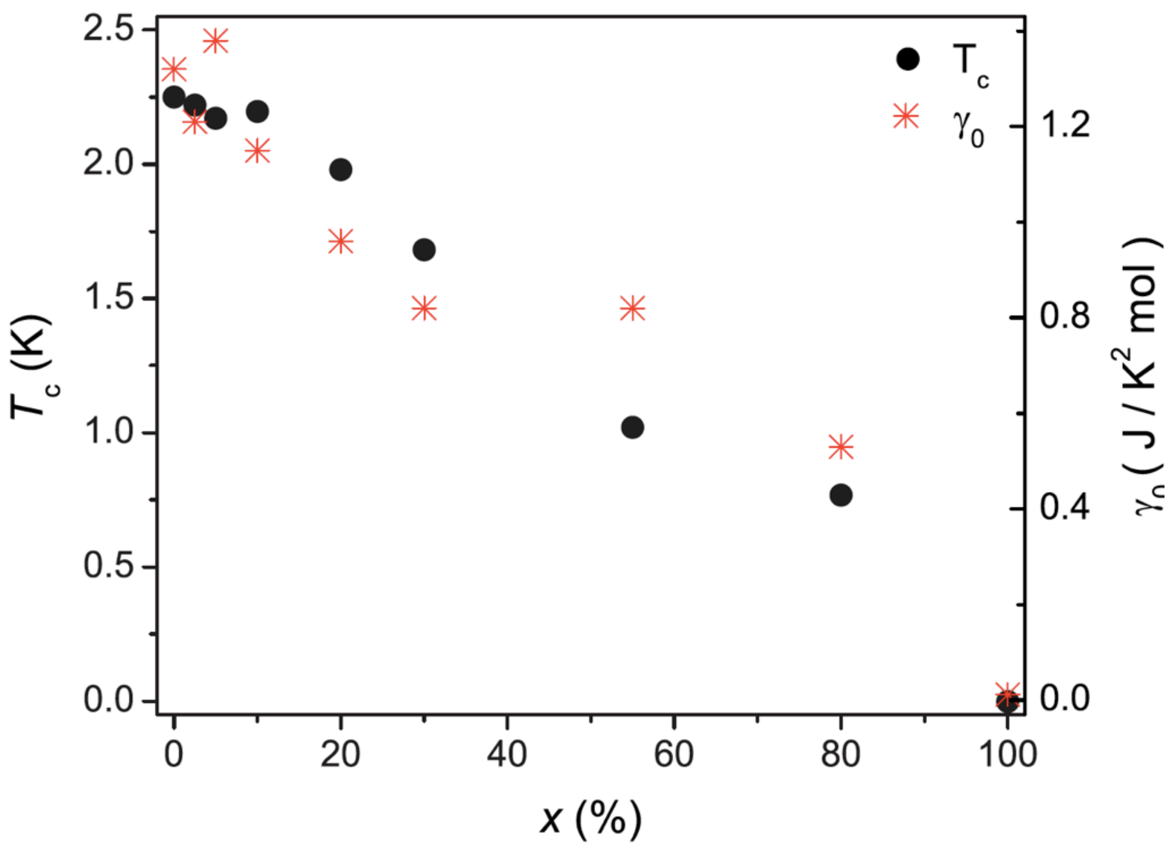
*A. Polyakov, O. Ignatchik, B. Bergk, K. Götze, A. D. Bianchi, S. Blackburn, B. Prévost, G. Seyfarth, M. Coté, D. Hurt, C. Capan, Z. Fisk, R. G. Goodrich, I. Sheikin, M. Richter, J. Woznitza, PRB **85**, 245119 (2012)*

$T_c \propto$ Ce concentration in $Ce_{1-x}Yb_xCoIn_5$

$T_c \propto$ Ce concentration



Shu, Baumbach, Janoschek et. al. PRL **106** 156403 (2011)



Booth et. al. PRB **83** 235117 (2011)

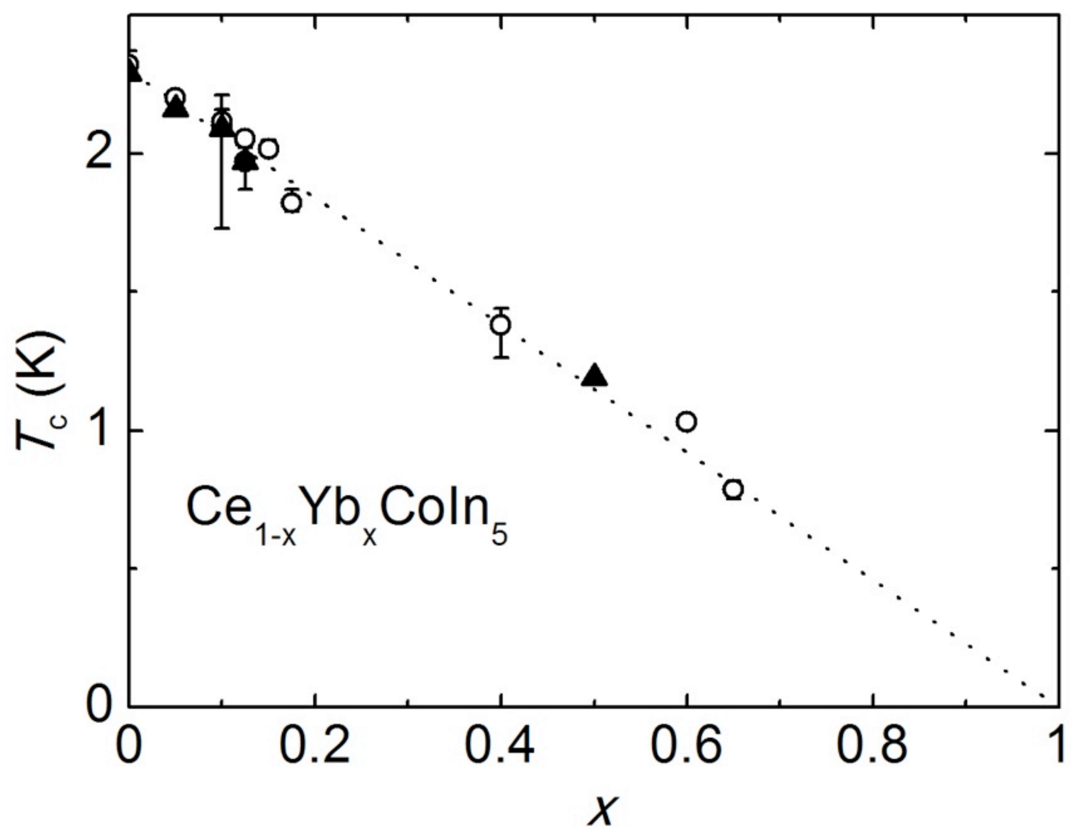
- Tc $\propto$ Ce concentration
- As if SC generated locally by Ce
- No obvious feature in *Tc*(x) near x ≈ 0.2
- Could provide clues to origin of unconventional SC of $CeCoIn_5$

Each Ce atom provides a composite pair

T_c and ρ_s increase linearly with Ce concentration

Communication with P. Coleman

$T_c \propto$ Ce concentration



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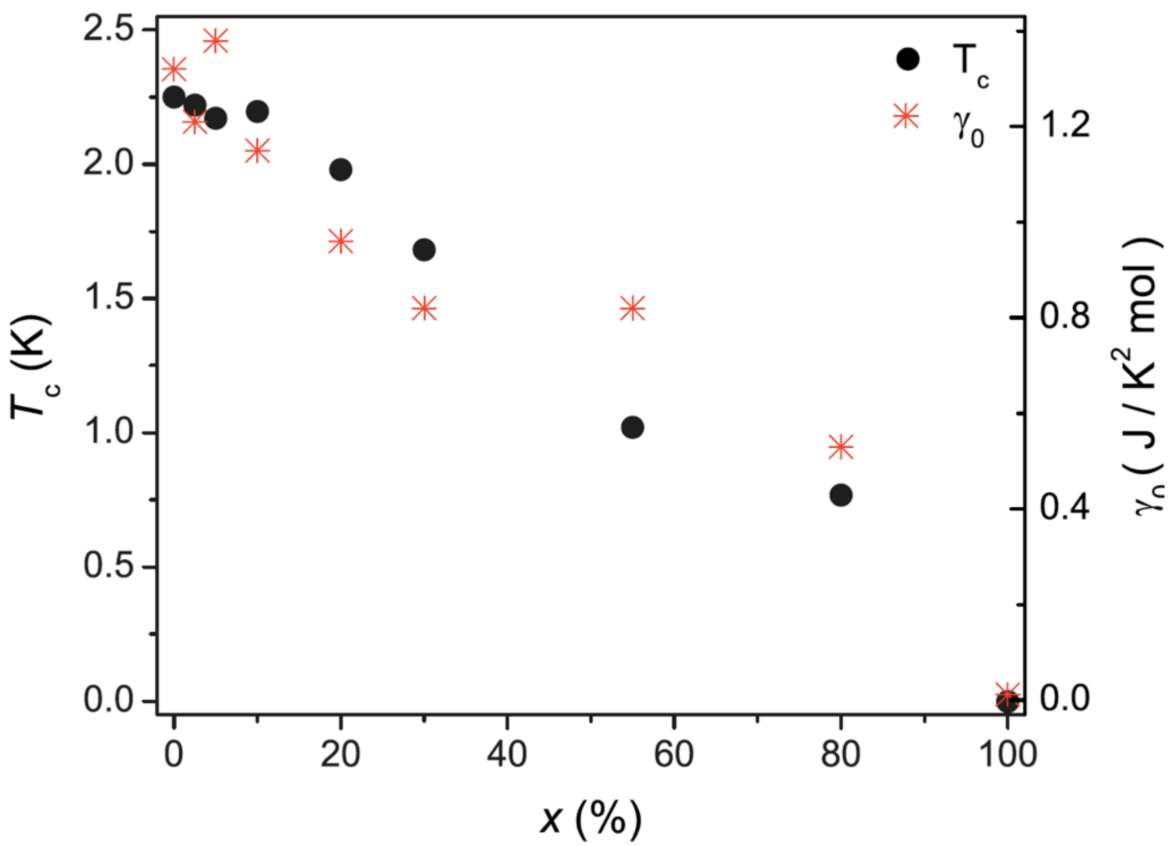

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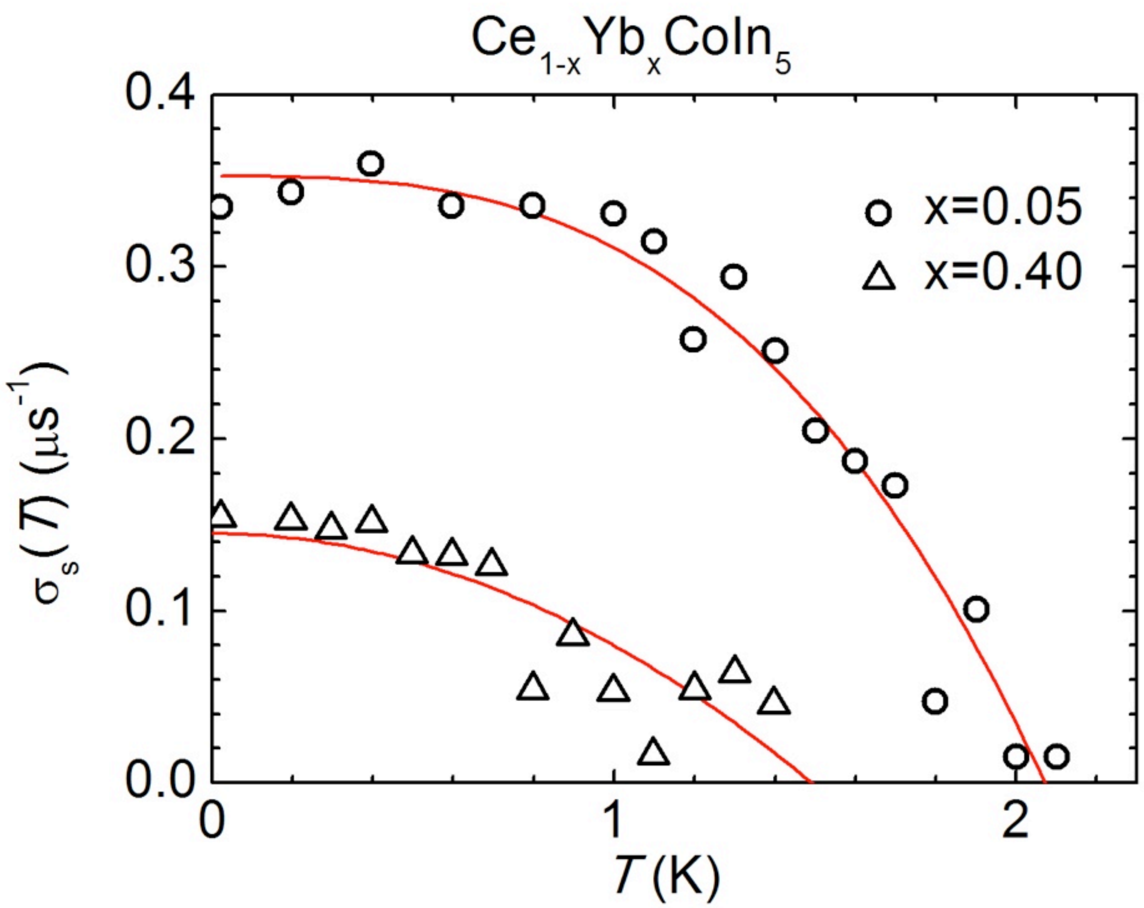
Communication with P. Coleman

TF-μSR

Measure absolute value of magnetic penetration depth $\propto \rho_s^{-1/2}$, to see if *T*c is controlled by ρ_s or whether ρ_s is roughly constant.

Muon's relaxation rate in the vortex state of $Ce_{1-x}Yb_xCoIn_5$



$$\sigma_s(T) = \sigma_s(0)(1-(T/T_c)^n)$$

x = 0.05

$$\sigma_s(0) = 0.35(1)$$
$$T_c = 2.07(4)$$
$$n = 2.9(4)$$

x = 0.40

$$\sigma_s(0) = 0.14(1)$$
$$T_c = 1.49(1)$$
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$$\lambda(0) = 5536 \text{Å}$$

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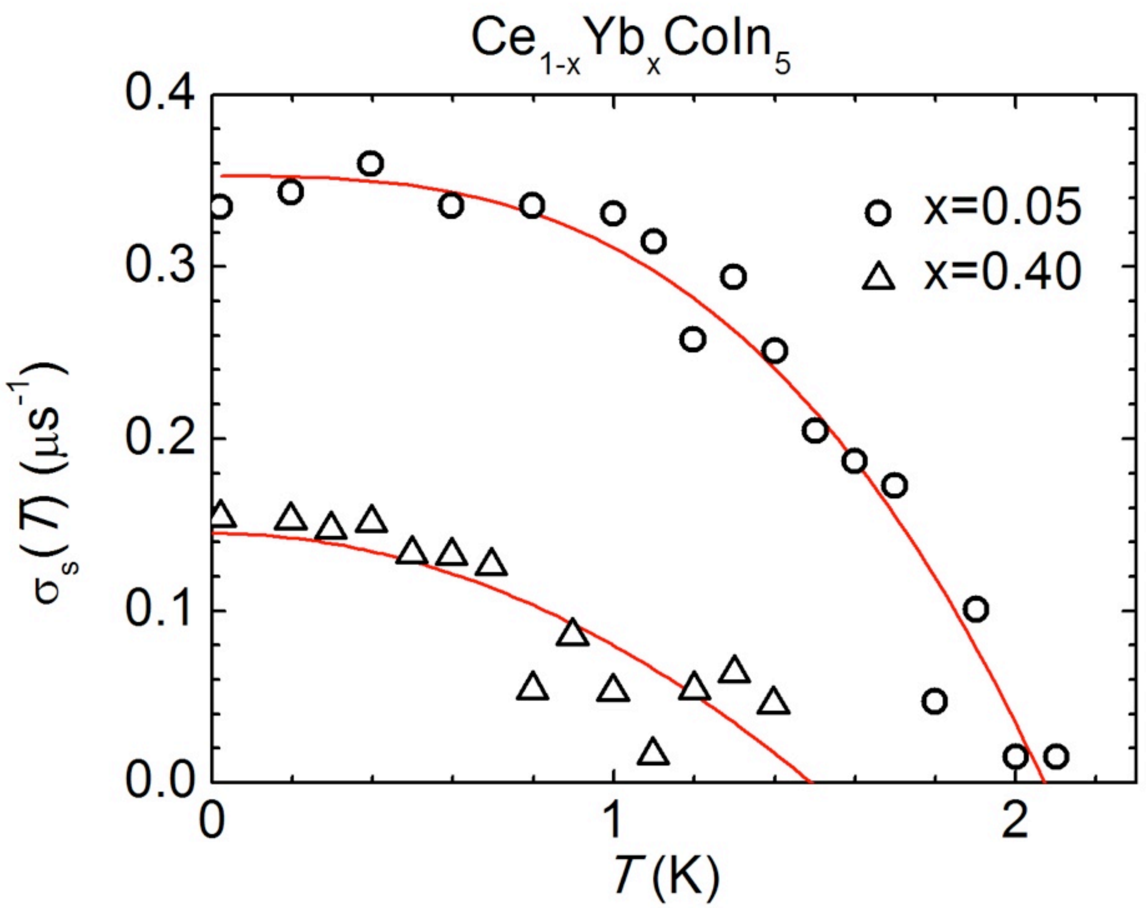

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W. Higemoto et al
JPSJ 71 1023

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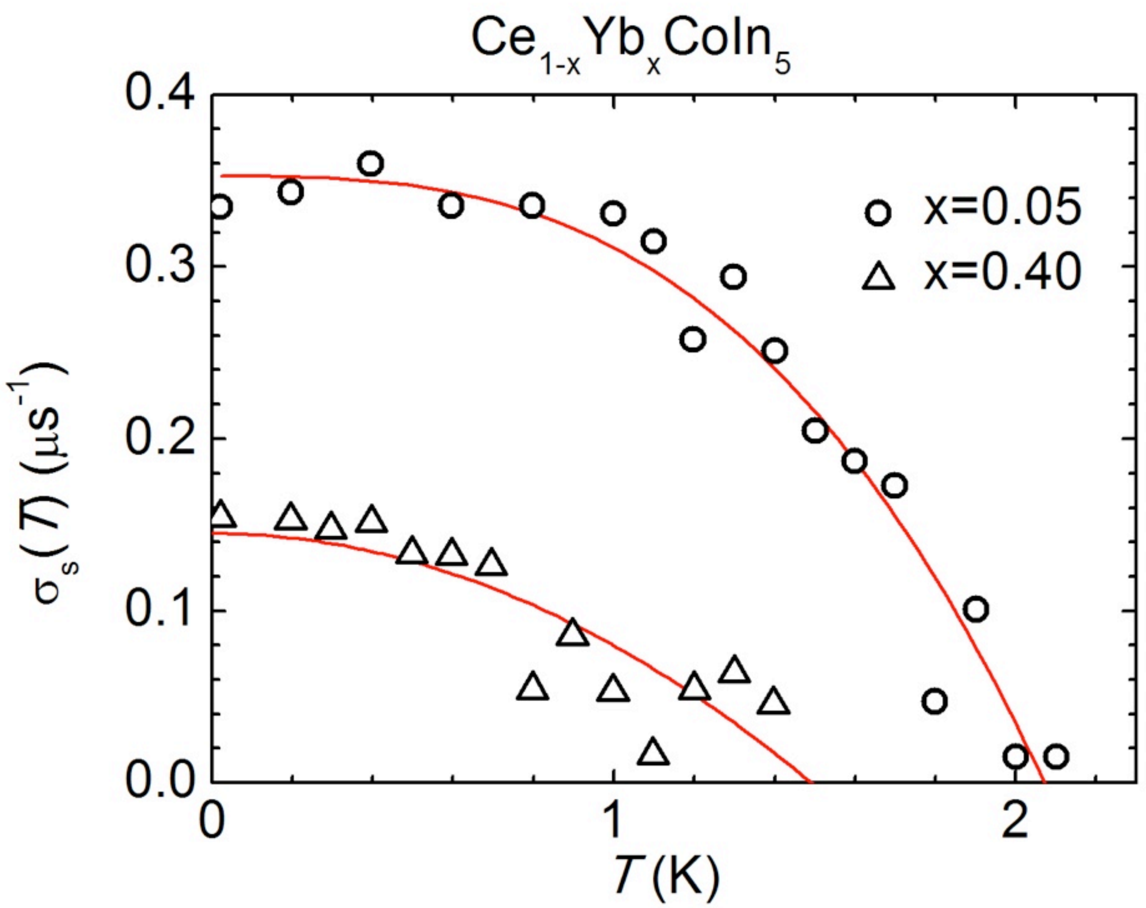

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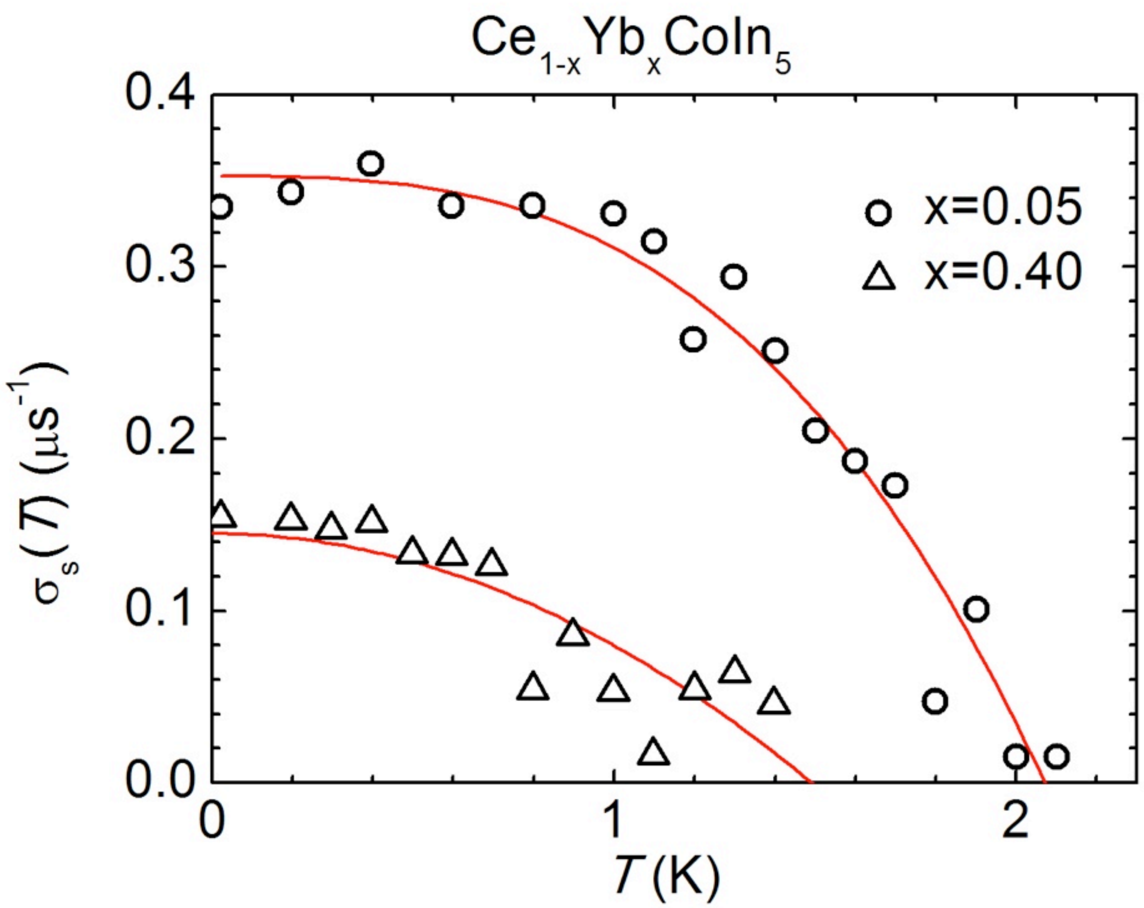

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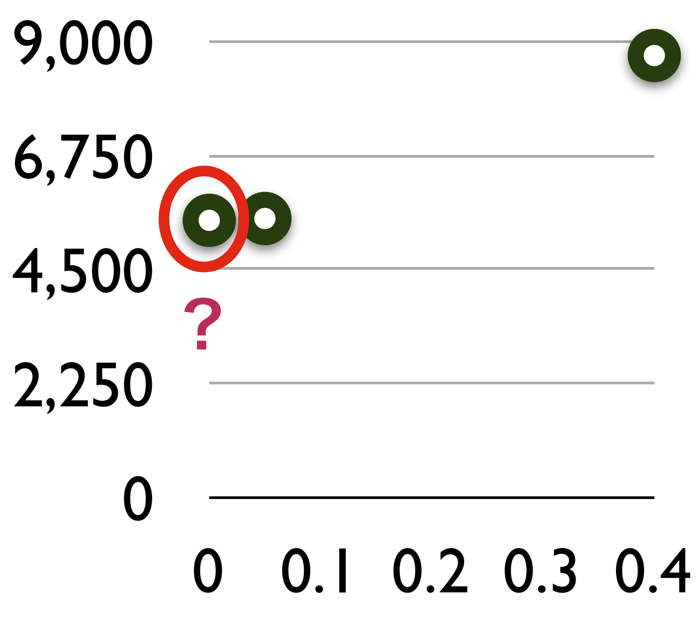

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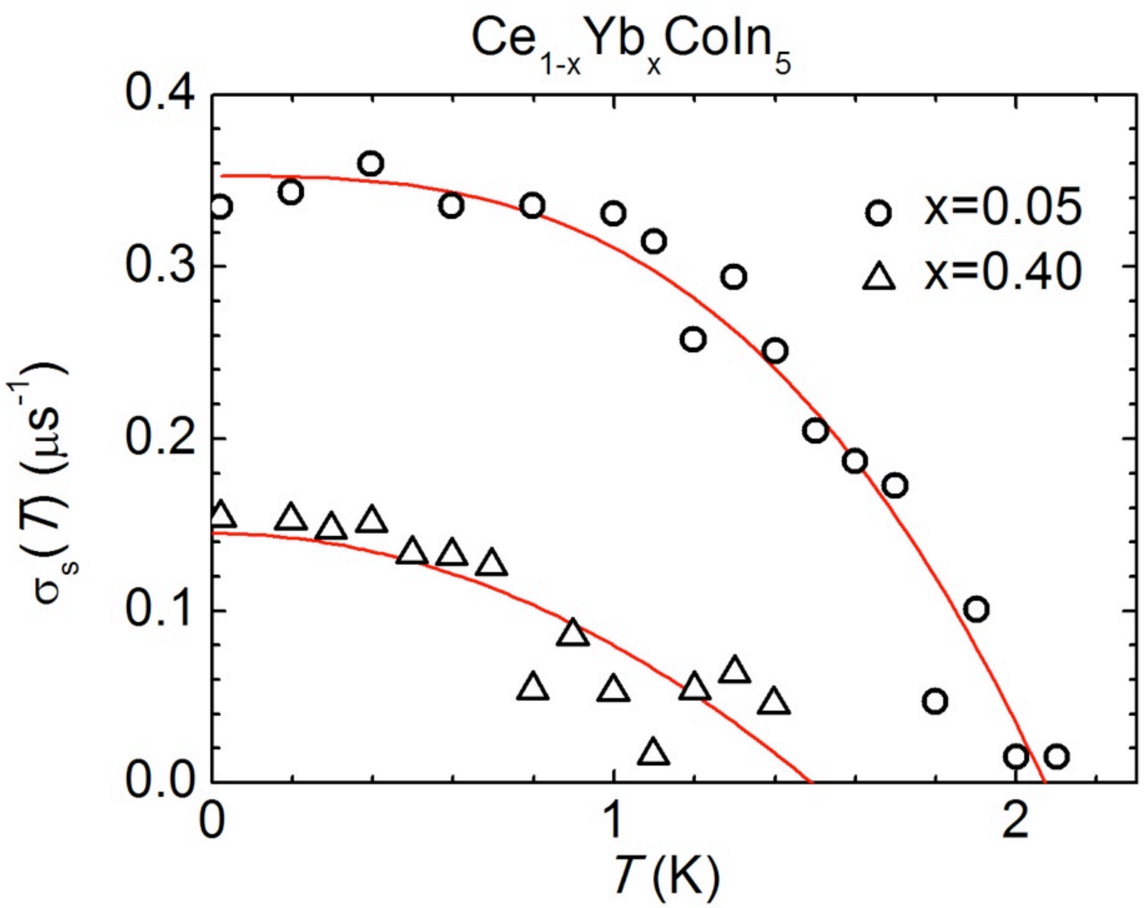
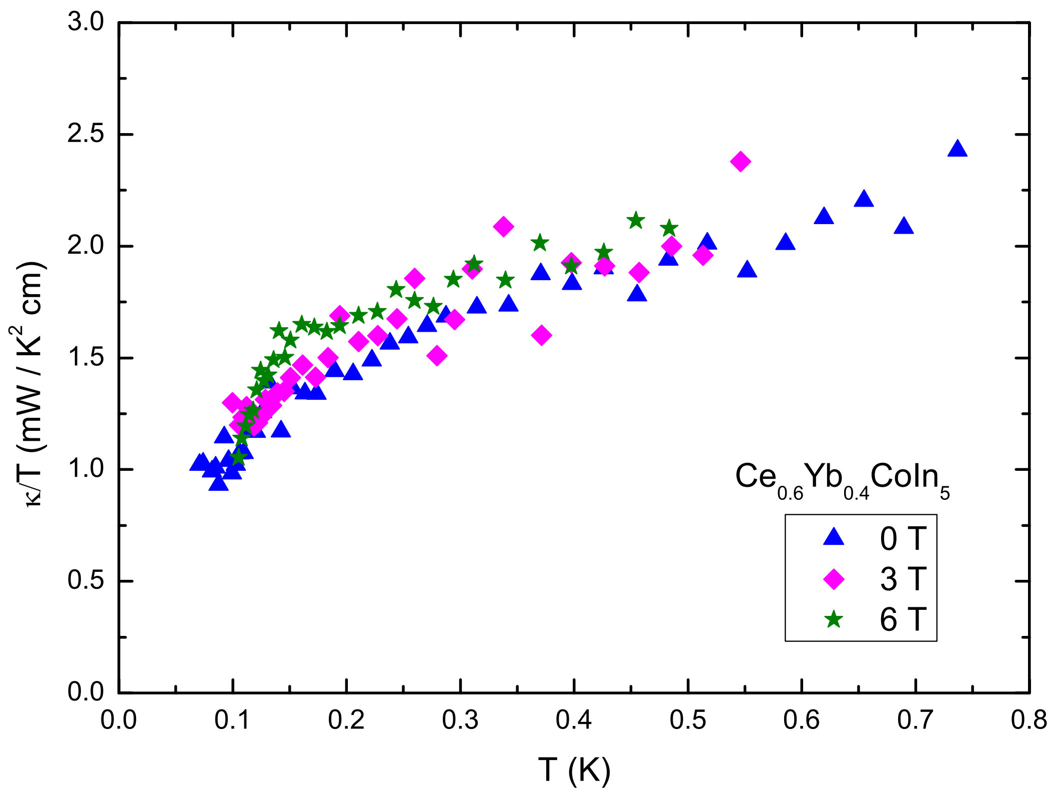


Thermal conductivity



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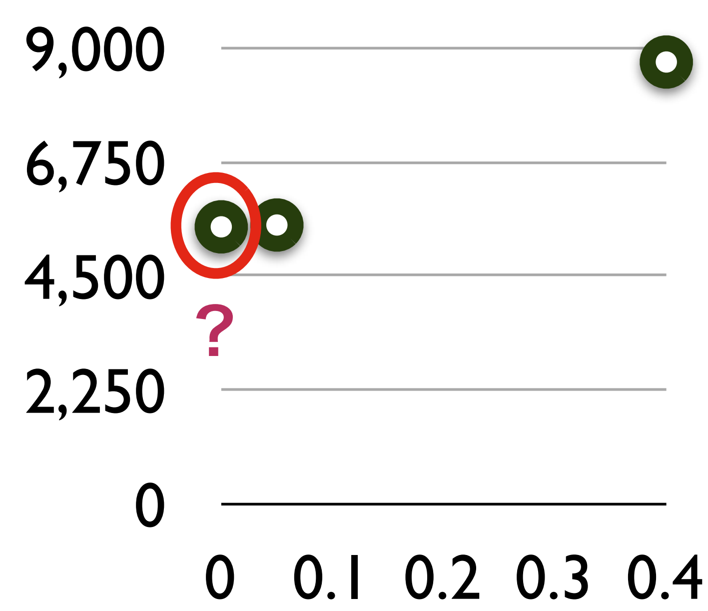
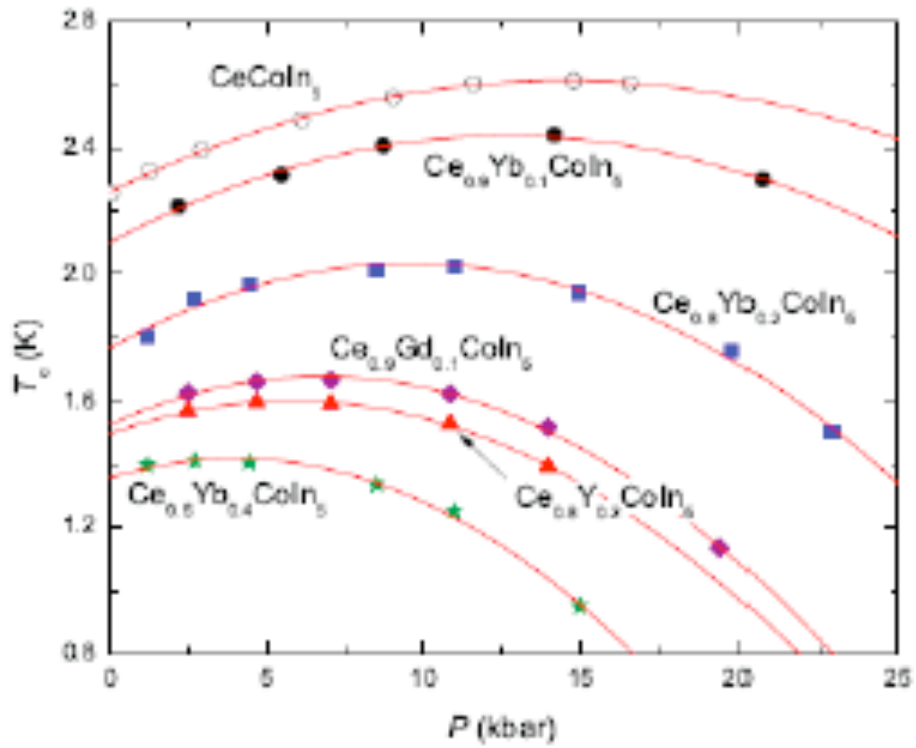
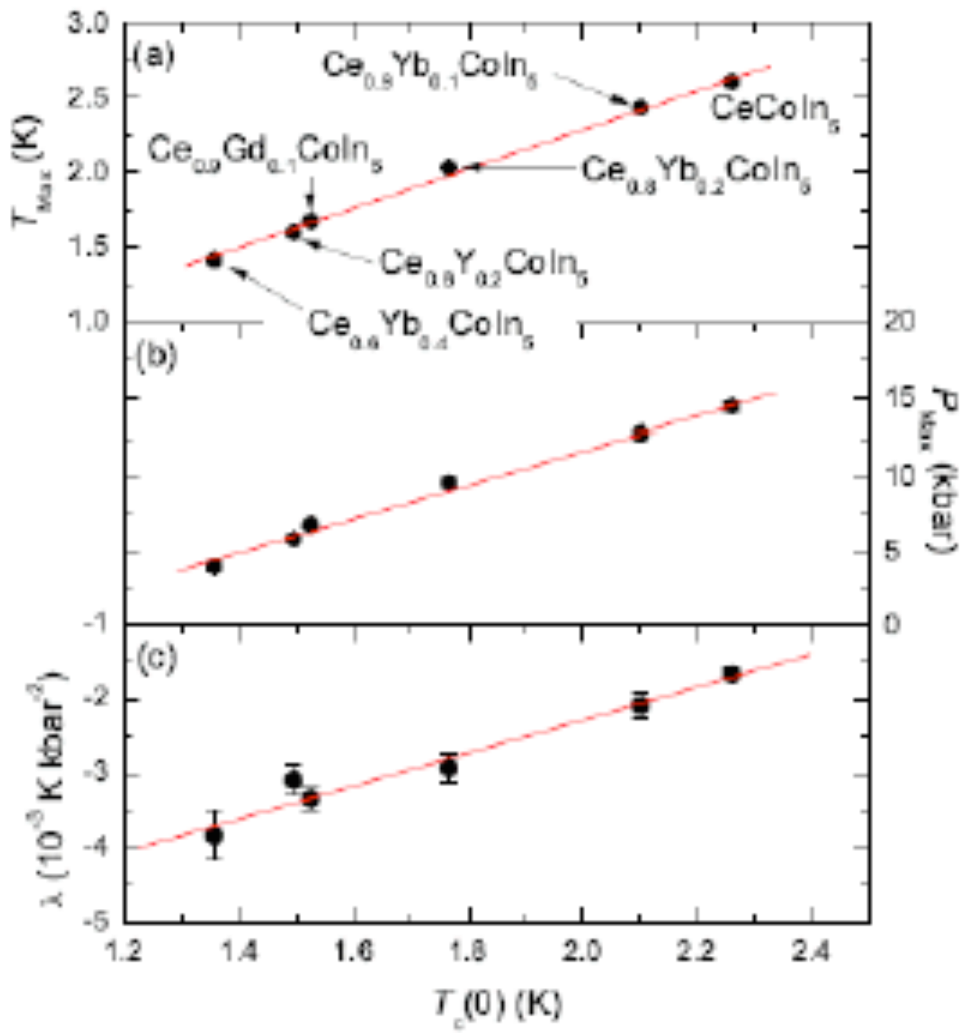


$Ce_{1-x}Yb_xCoIn_5$: Pressure dependence of T^* and T_c

Effect of pressure on superconductivity and Kondo-lattice coherence temperature in $Ce_{1-x}Yb_xCoIn_5$





- $T_c(P)$ data described by expression

$$T_c = T_{\rm Max} + \lambda (P - P_{\rm Max})^2$$

T_{Max}, P_{Max}, λ are linear functions of $T_c(0)$

- $T^*(P)$ data described by expression

$$T^*(P) = T^*(0) + \xi P$$

- Effect of pressure on $Ce_{1-x}R_xCoIn_5$ is independent of R and x in range 0 to 2.5 GPa
- Apparently, pressure does not change v_{Yb} and primarily affects electronic state of Ce ion

B. D. White, J. J. Hamlin, K. Huang, L. Shu et al, Phys. Rev. B 86 100502(R) 2012

Conclusions

- In the Ce1-xYbxCoIn5 system, Kondo coherence and SC are weakly dependent on x, while the NFL characteristics exhibit strong variations with x

- This may be due to cooperative behavior involving the unstable valences of Ce and Yb

- XPS, XANES, and magnetic susceptibility measurements indicate:
  - − Ce valence is close to +3 for all x
  - − Yb valence decreases from ~+3 at x ≈ 0 to ~+2.3 at x ≈ 0.2 and then remains constant at ~ +2.3 to x ≈ 1 ⇒Yb VALENCE TRANSITION

- Tc is proportional to Ce concentration! T_c does not scale with T^*

- The pressure dependences of T^* and T_c in $Ce_{1-x}R_xCoIn_5$ are independent of R ion (Yb, Y, and Gd) in the pressure range 0 to 2.5 GPa ⇒no change in v_{Yb} in this P range, need higher pressure!

x : Yb concentration

n_{Ce}^{3+} : number of Ce^{3+}

n_{Ce}^{4+} : number of Ce^{4+}

n_{Yb}^{3+} : number of Yb^{3+}

n_{Yb}^{2+} : number of Yb^{2+}

δV_{Ce} : the volume difference between each Ce^{3+} and Ce^{4+}, which should > 0

δV_{Yb} : the volume difference between each Yb^{3+} and Yb^{2+}, which should < 0

Ce and Yb ions adopt cooperative intermediate valence states, which means n_{Ce}^{3+}, n_{Ce}^{4+}, n_{Yb}^{3+}, n_{Yb}^{2+} adjust cooperatively so that

$$(1-x)(n_{\mathrm{Ce}^{3+}} - n_{\mathrm{Ce}^{4+}})\delta V_{Ce} + x(n_{\mathrm{Yb}^{3+}} - n_{\mathrm{Yb}^{2+}})\delta V_{Yb} = 0$$

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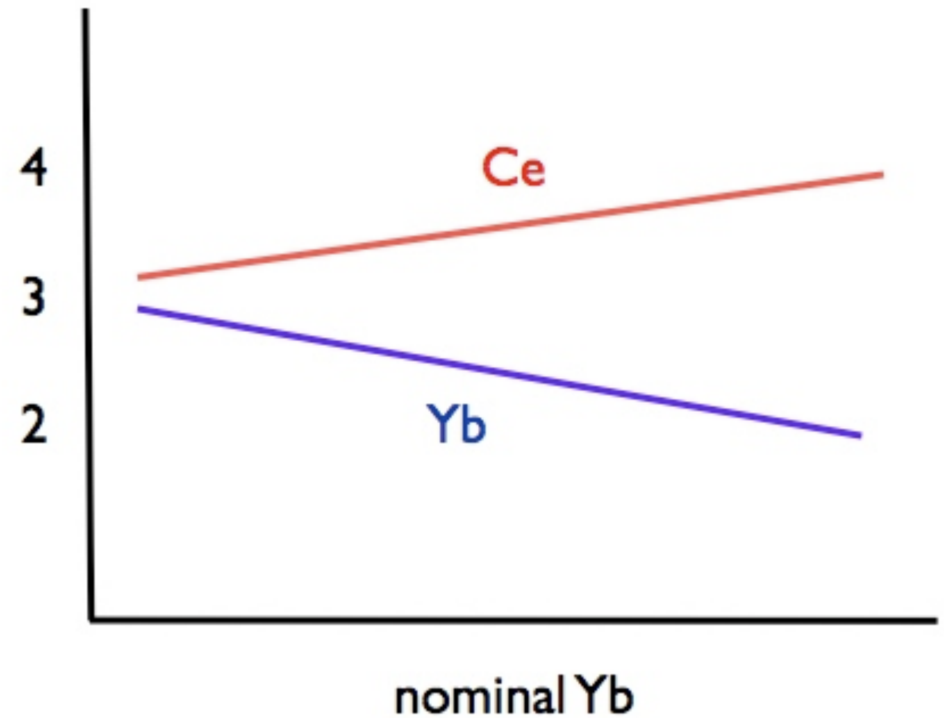


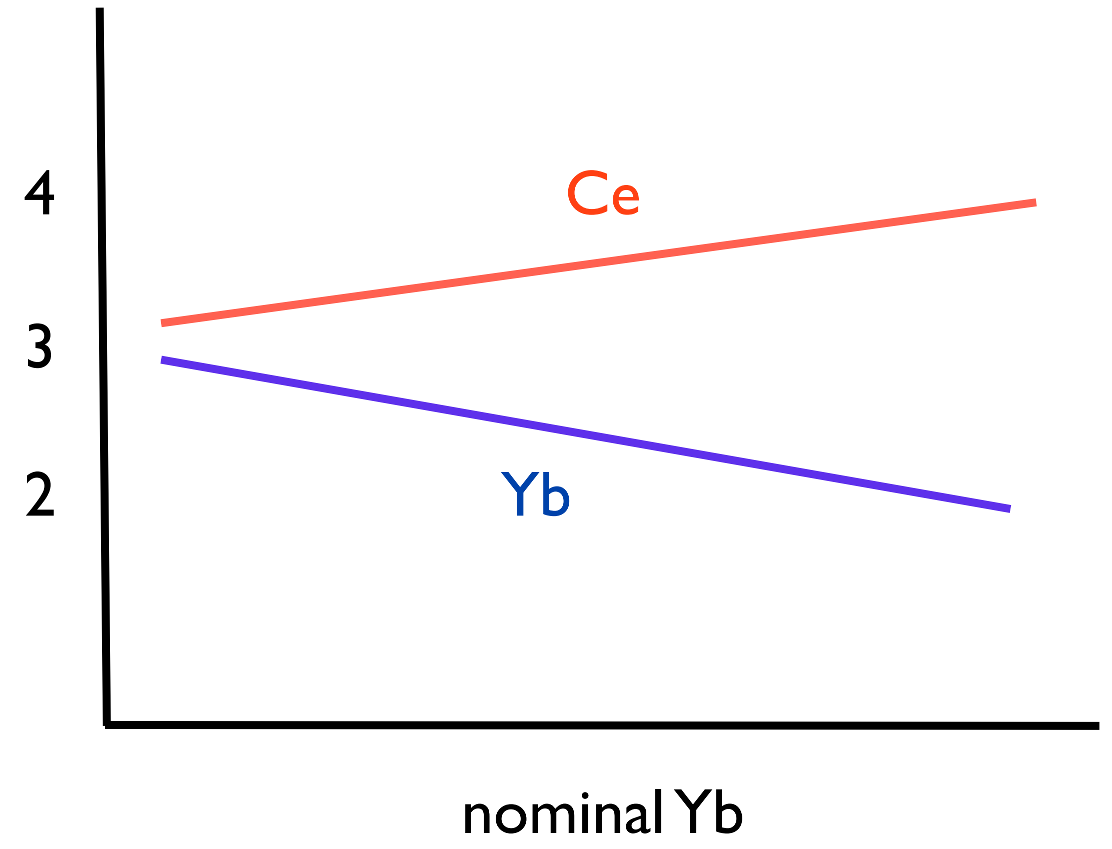
The pair-breaking effect in unconventional superconductors arises via

- potential (non-magnetic)
  Abrikosov-Gor'kov model ($T_c \to 0$, when $l_{mfp} \to \xi$)
- spin-flip scattering

  $\Delta T_c \propto \mathcal{J}^2 D_J$

  $\mathcal{J}$ is small (even-parity)

  D_J : 0.18 Ce^{3+} ($4f^1$)
  0.32 Yb^{3+} ($4f^{13}$)



For each x of $Ce_{1-x}Yb_xCoIn_5$,
if we call

- f-electron occupancy for Ce: n^{Ce}_f
- f-hole occupancy for Yb: n^{Yb}_f
- valence of Ce: ν_{Ce}
- valence of Yb: ν_{Yb}
- effective moment of Ce: μ_{Ce}
- effective moment of Yb: μ_{Yb},

then we have the relation:

$$\nu_{Ce} = 3n_f^{Ce} + 4(1 - n_f^{Ce})$$

$$n_f^{Ce} = 4 - \nu_{Ce}$$

$$n_f^{Ce} = \frac{\mu_{Ce}^2}{\mu_{Ce3+}^2}$$

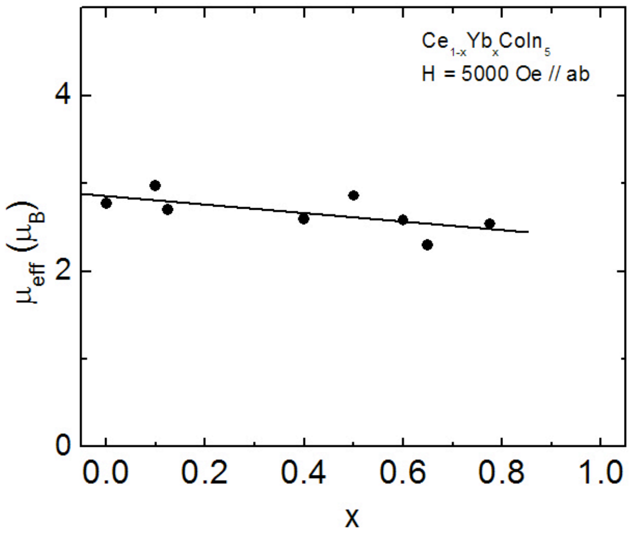
$$\nu_{Yb} = 3n_f^{Yb} + 2(1 - n_f^{Yb})$$

$$n_f^{Yb} = \nu_{Yb} - 2$$

$$n_f^{Yb} = \frac{\mu_{Yb}^2}{\mu_{Yb3+}^2}$$

where $\mu_{Ce3+} = 2.54\ \mu_B$
$\mu_{Yb3+} = 4.54\ \mu B$

$$\mu_{eff}^2(x) = \mu_{Ce}^2(1-x) + \mu_{Yb}^2(x)$$



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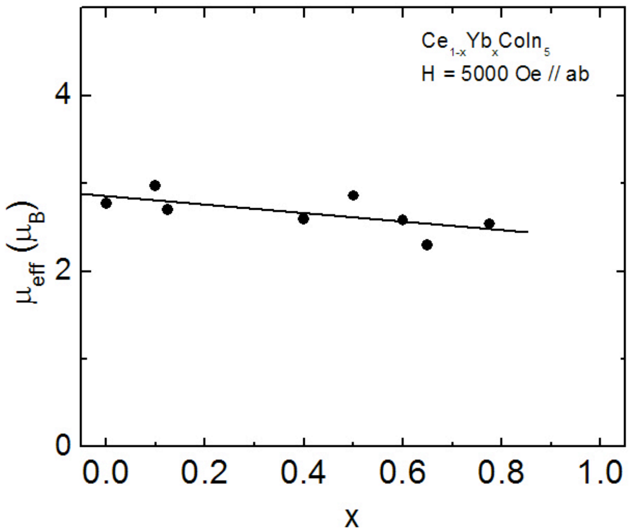

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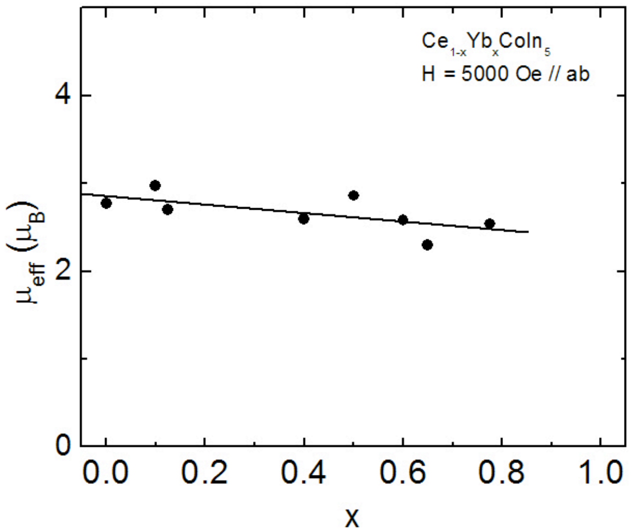

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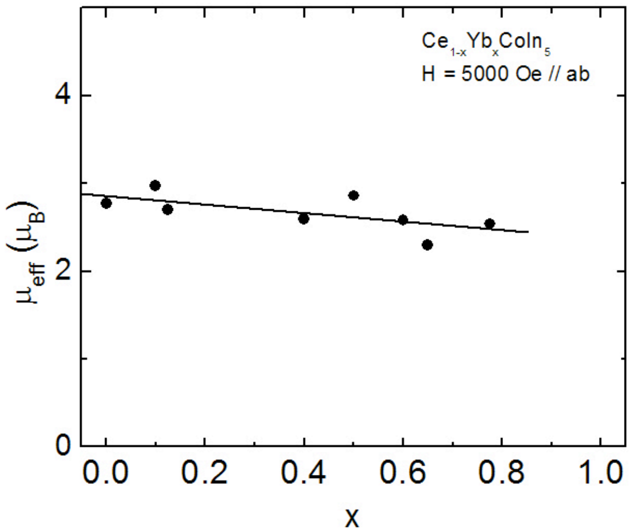

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