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MINIWORKSHOP ON
STRONGLY CORRELATED ELECTRON SYSTEMS

15 JUNE - 10 JULY 1992

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# "Heavy-fermion magnets, super and semiconductors"

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# Heavy-fermion magnets, super- and semiconductors

## collaboration

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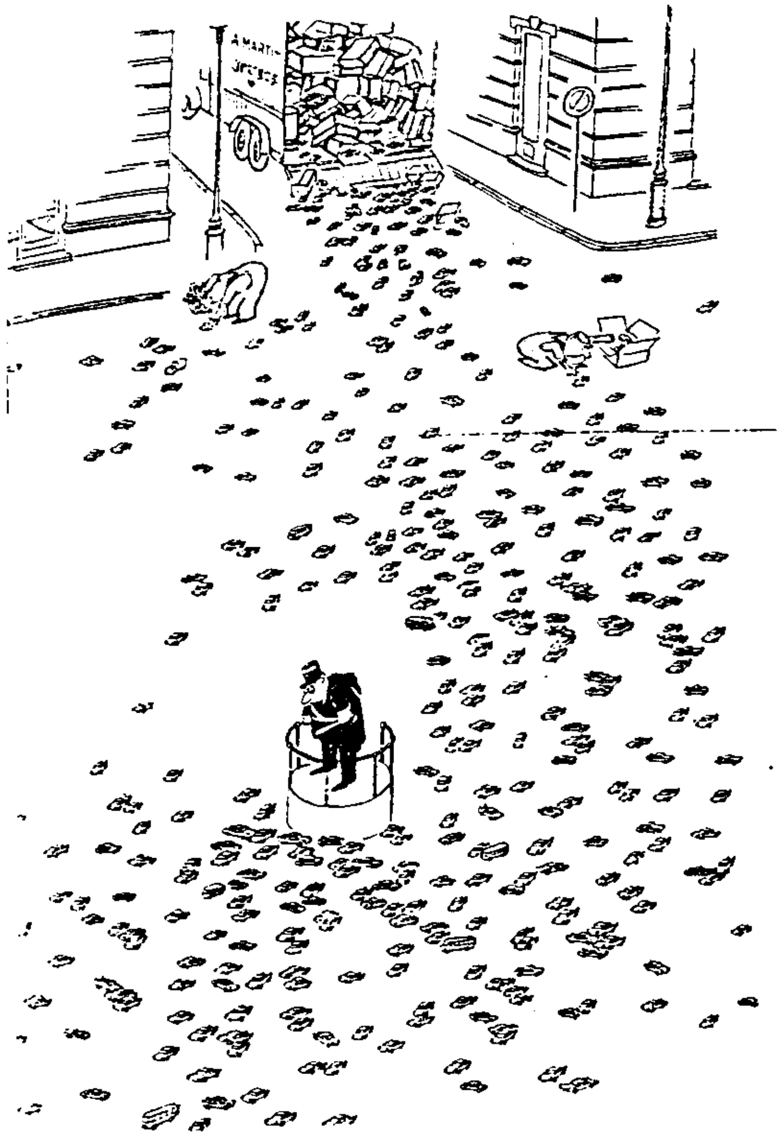
## outline

1. Magnetism & S/C in Ce-"122"
2. S/C and lattice instability in $CeCu_2Si_2$ (and cuprates)
3. New antiferromagnetically ordered heavy-fermion S/C: U-"123"
4. Heavy fermions in materials with extremely low (if not vanishing) carrier concentration: $Sm_3X_4$, $Yb_4As_3$

Intraatomic (f-shell) energies: U, J, ..

↓ correlated motion

counter-example:



Mott transition

cooperative magnetism

Superconductivity : novel types

electron-lattice coupling: lattice instabilities

# History

P. W. Anderson : Epilog, ICVF
Rochester '76

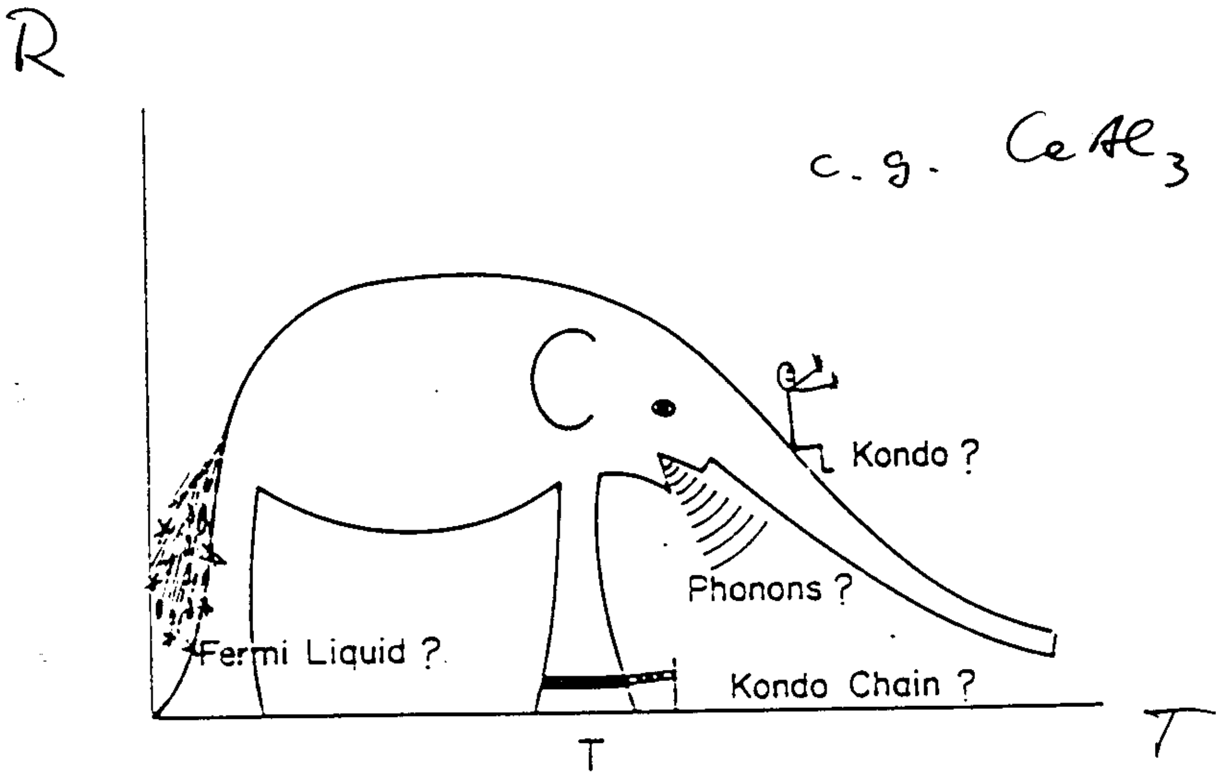



Fig. 2. Elephantine version of Fig. 1 (art work by PWA)

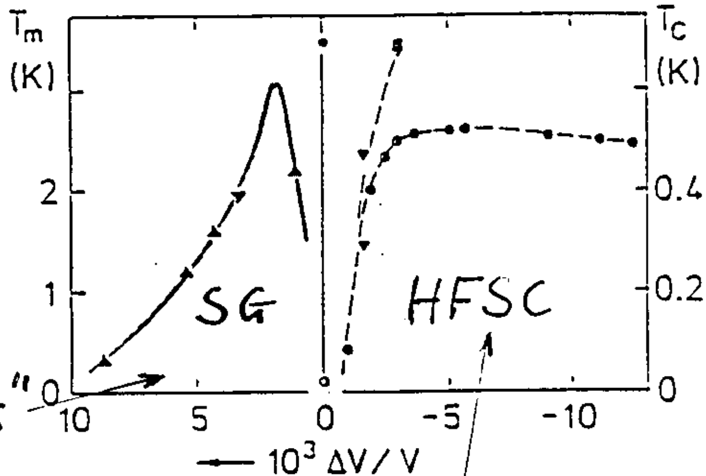
# 1.) Magnetism and superconductivity in Ce "122"

(ThCr$_2$Si$_2$ structure)

## magnetic – nonmagnetic transition
(Doniach '77)

local moments    heavy Fermi liquid

$CeCu_2Si_2$



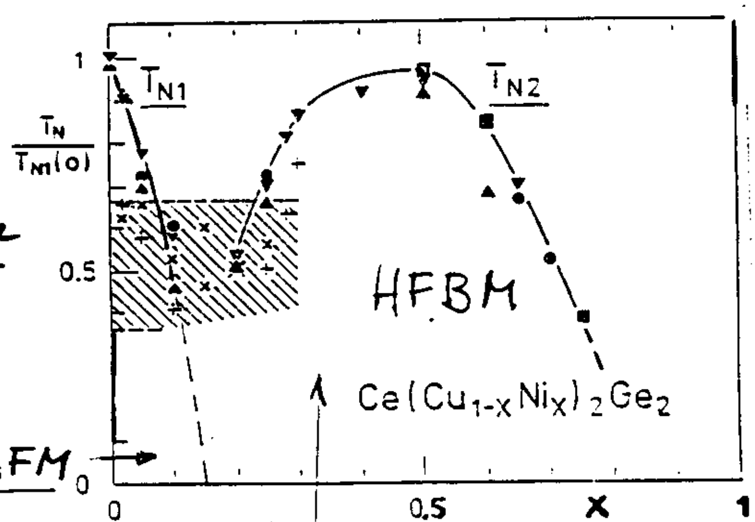

"Spin glass"

heavy-fermion superconductivity

Moshchalkov et al. '84

Rauchschwalbe et al. '85

$Ce(Cu_{1-x}Ni_x)Ge_2$




local-moment AFM

heavy-fermion band magnetism

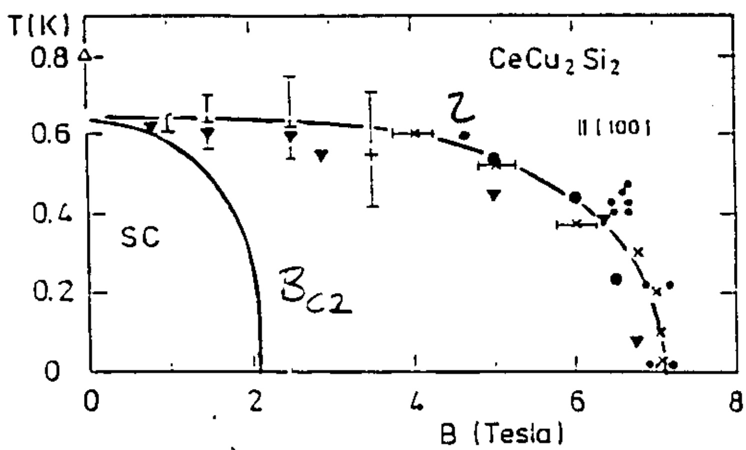
Sparn et al. '88

Loidl et al. '90

(theory: Grewe, Welslau '88)

$CeCu_2Si_2$

$T_c = 0.63$ K




Rauchschwalbe et al. '87

Nakamura et al. '88

Uemura et al. '89

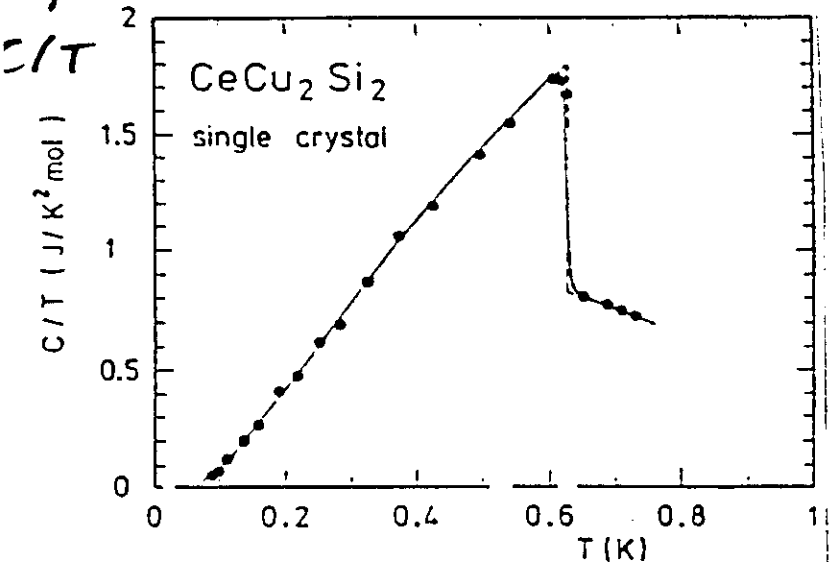
Hunt et al. '90

Bruls et al. '90

Lang et al. '91

## 1a) $CeCu_2Si_2$ single crystal ($m = 15\,mg$)

**Spec. heat** $T^* = 15\,K$ $T_c = 628\,mK$




$\underline{T \to 0:}$ $\gamma \simeq 1 \frac{J}{K^2 mol}$

($Cu$: $< 10^{-3}$ ″ )

$m^* \simeq 300\, m_{el}$

$v_F^* \simeq v_{sound}$

$\underline{T = T_c:}$ $\Delta C \gtrsim C_{normal}$

↳ heavy-fermion s/c

**thermal expansion** ($\Delta \ell \gtrsim 10^{-3}\,Å$)

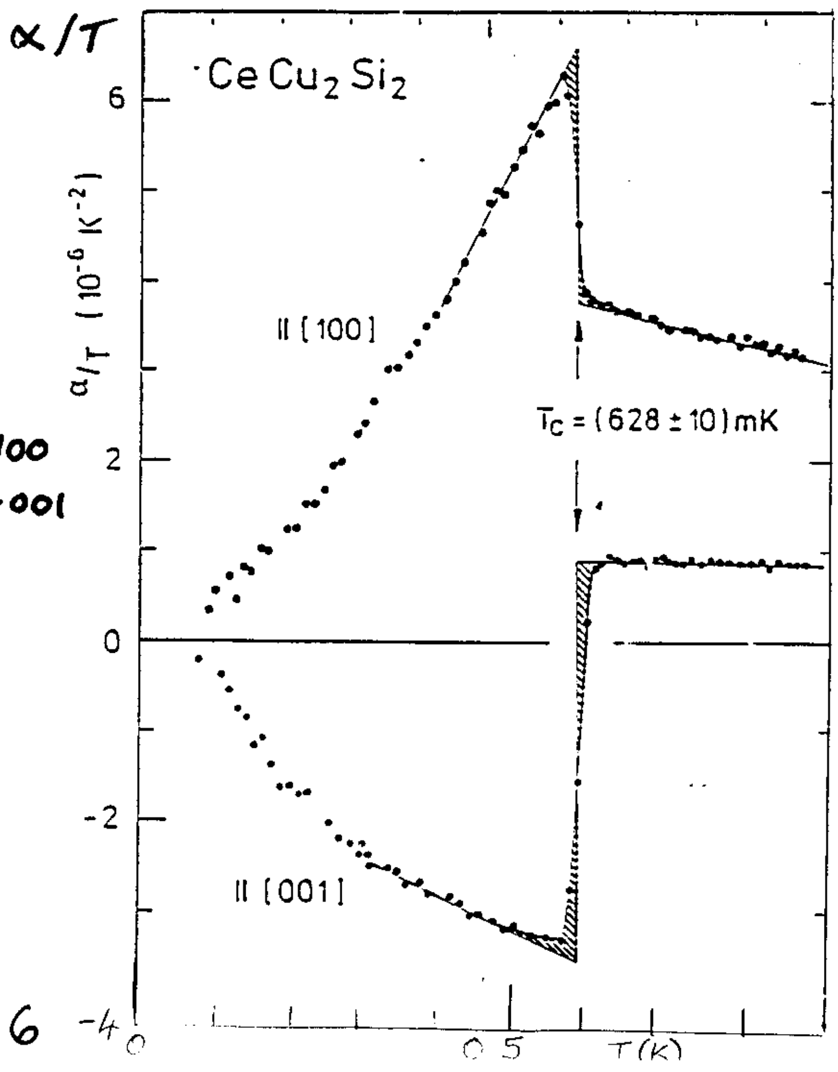
$\underline{T > T_c:}$

Grüneisen parameter

$$\Gamma = \frac{V_{mol}\ \beta}{\kappa_T\ C_V} \quad \Big| \quad \beta = 2\alpha_{100} + \alpha_{001}$$

$$= -\frac{\partial \ln T^*}{\partial \ln V}$$

$$= 63$$



# S/C phase transition

Ehrenfest : $\left(\frac{\partial T_c}{\partial p_i}\right)_{p_i \to 0} = V_{mol}\, T_c \,\frac{\Delta \alpha_i}{\Delta C_v}$

exp: $\frac{\partial T_c}{\partial p_a} = +(9 \pm 1)\,\frac{mK}{kbar}$ ; $\frac{\partial T_c}{\partial p_c} = -(14.5 \pm 1)\,\frac{mK}{kbar}$

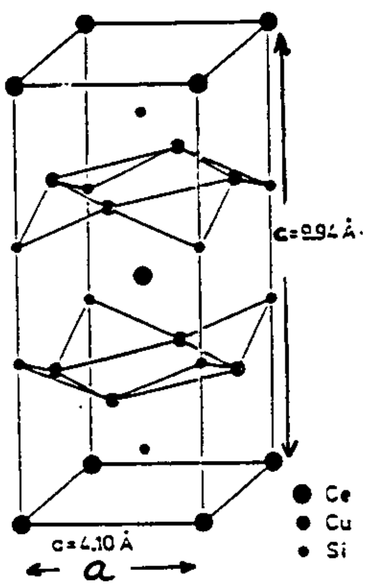
$\to \left(\frac{\partial T_c}{\partial p_{hydrost.}}\right)_{p_h \to 0} = 2\,\frac{\partial T_c}{\partial p_a} + \frac{\partial T_c}{\partial p_c} = 3.6\,\frac{mK}{kbar}$

(Eichler '85 : $4.0\,\frac{mK}{kbar}$ )

$$\frac{\partial T_c}{\partial \varepsilon_a} = -(c_{11} + c_{12})\,\frac{\partial T_c}{\partial p_a} - c_{13}\,\frac{\partial T_c}{\partial p_c} = -(15 \pm 9)\,K$$

$$\frac{\partial T_c}{\partial \varepsilon_c} = -2c_{13}\,\frac{\partial T_c}{\partial p_a} - c_{33}\,\frac{\partial T_c}{\partial p_c} = +(5 \pm 9)\,K$$

$CeCu_2Si_2$




conclusion:

HFSC requires sufficient compensation of 4f-moment

## $CeCu_2Ge_2$ ($ThCr_2Si_2$)

$T^* = 8K$

$T_{RKKY} \simeq 7K$

$T_N = 4.1K$

$V_c = 177\ \text{Å}^3$

## $CeCu_2Si_2$

$T^* = 15K$

$V_c = 167\ \text{Å}^3$

$C_B = 1200\ \text{kbar}$

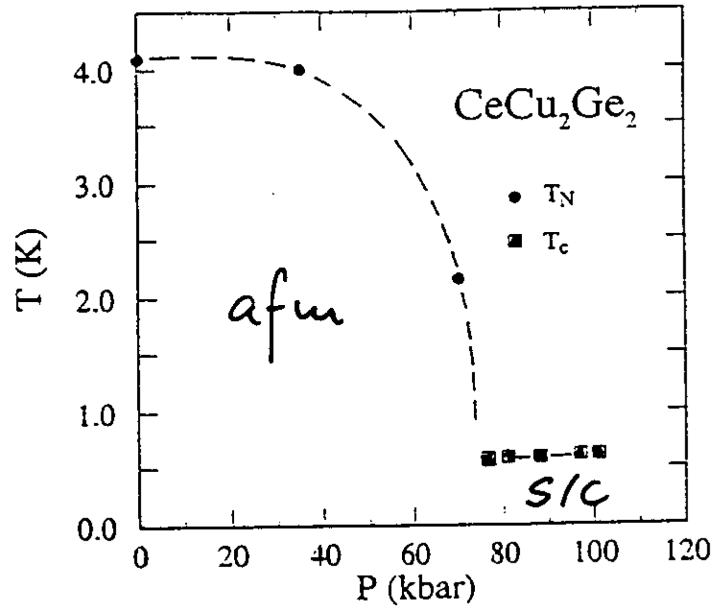
$\downarrow$

$CeCu_2Ge_2$ assumes $V_c = 167\ \text{Å}^3$

under $p_{hydrost} \approx 70\ \text{kbar}$

D. Jaccard, K. Behnia, J. Sierro

(Phys. Lett. A 163, 475 '92)



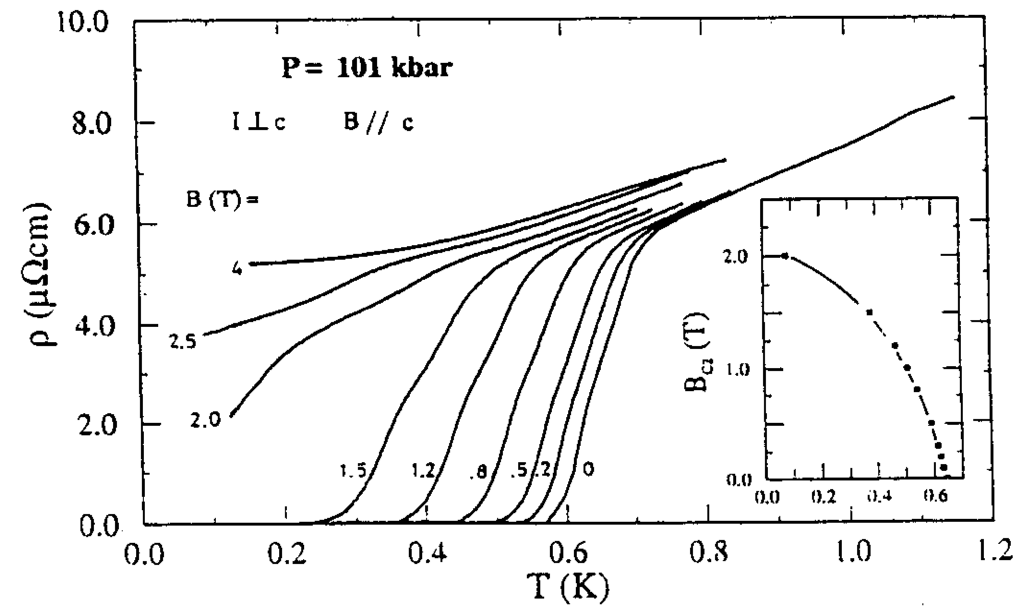

$\frac{dT_c}{dp} > 0$, small

$CeCu_2Ge_2$ ($p > 70$ Kbar) : $T_c = 0.64$ K

$B'_{c2} = -11$ T/K

$B_{c2}(0) = 2$ T

$\rho \uparrow$




16
~~20~~ min

Very similar to $CeCu_2Si_2$

1b) Alloying-induced transition from local-moment AFM to heavy-fermion "band magnetism" in

$Ce(Cu_{1-x}Ni_x)_2Ge_2$

Magnetic phase diagram

$CeCu_2Ge_2$

$T^* = 8K$

$T_{RKKY} = 7K$

$T_{N1} = 4.1K$

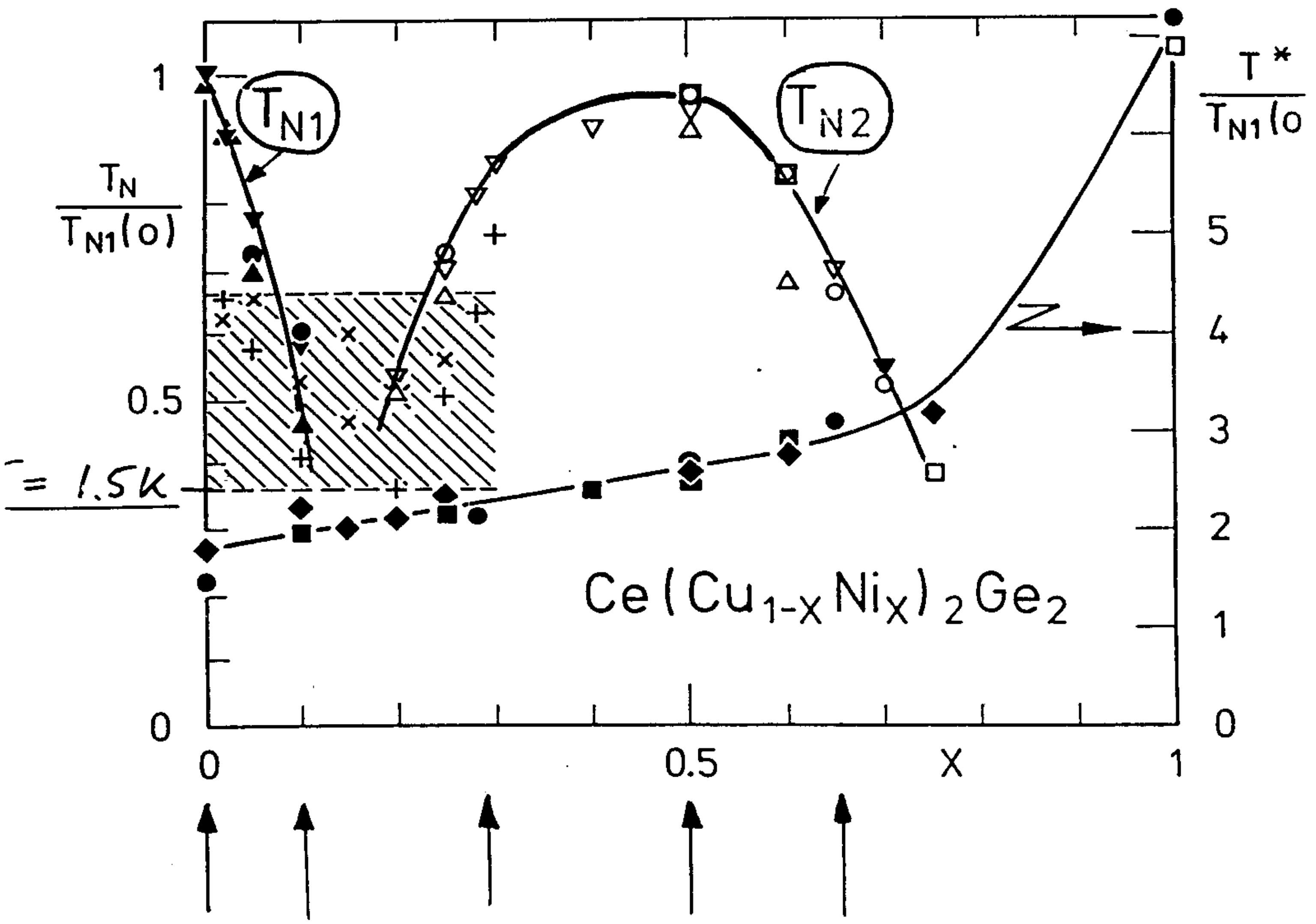
$\mu_s = 0.74\mu_B$

$CeNi_2Ge_2$

$T^* = 30K$

$\gamma = 0.4 J/K^2 mol$

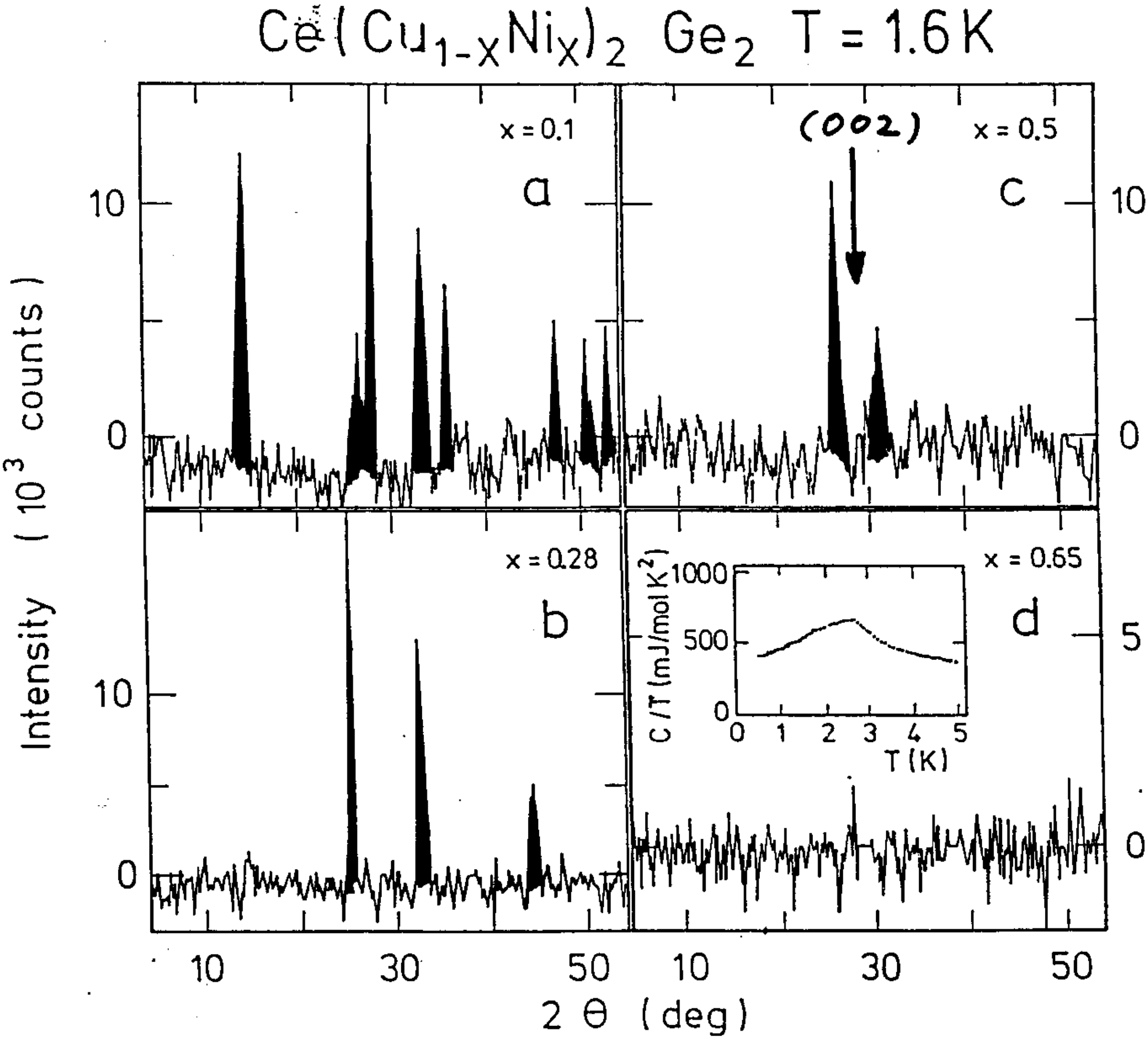
Fermi liquid



## Neutron diffraction (D16, ILL)

$I\,(1.5\,K) - I\,(6\,K)$

→ incommensurate magnetic structures



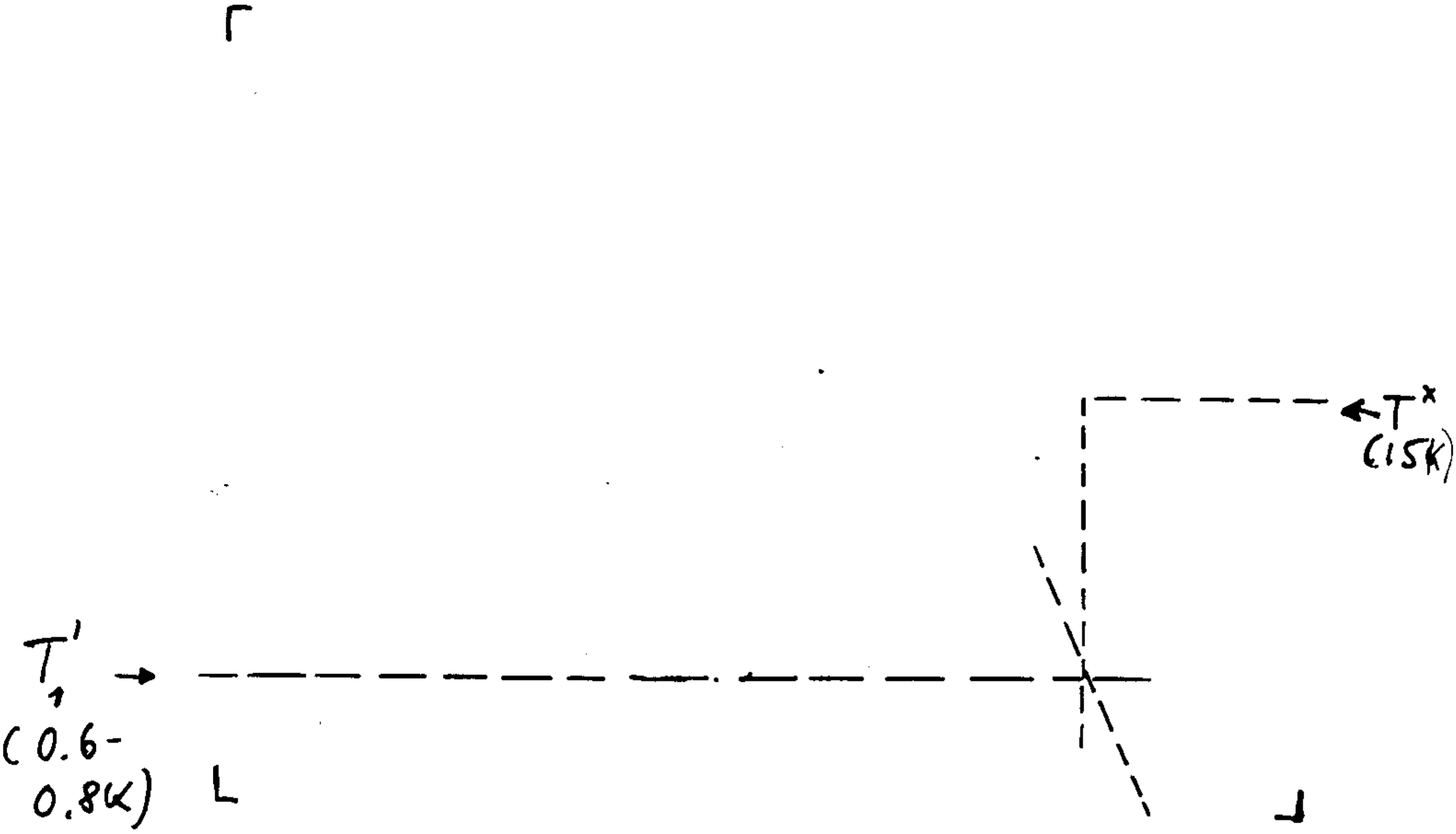

$x \leq 0.1$ : $q_0 \simeq 0.5\, K_{Bz}$ $\qquad \mu_S : 0.5 - 0.7\, \mu_B$

$x = 0.5$ : $q_0 \simeq 0.1\, K_{Bz}$ $\qquad \mu_S : 0.3\, \mu_B$

$x = 0.65$ : $\qquad \mu_S < 0.2\, \mu_B$

→ local-moment afm → heavy-fermion bandmagn

(in accord with theoretical predictions: Grewe et al. '88)

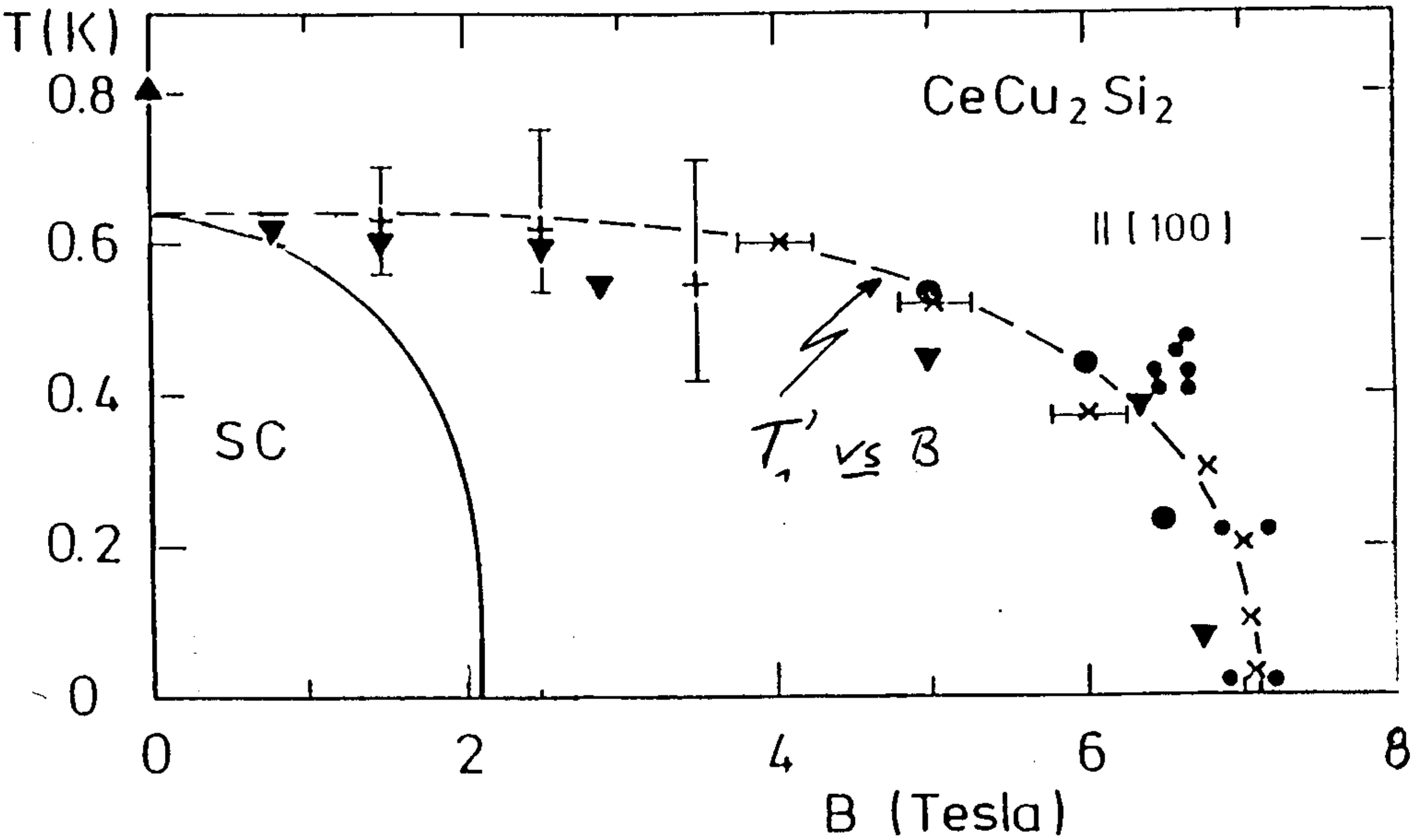



$CeCu_2Si_2$

## 2.) Superconductivity and lattice instability in $CeCu_2Si_2$ (and cuprates)

# 1c) $T - B$ phase diagram of bulk - s/c $CeCu_2Si_2$

▲ : μSR (Uemura et al. '89)
▼ : $c_{11}$ (Bruls et al. '90)
+ : NMR (Nakamura et al. '88)
× : $\Delta\rho(B)$ (Rauchschwalbe et al. '87)
• : dHvA (Hunt et al. '90)
● : $\alpha(T)$ (Lang et al. '91)




<u>$6T \lesssim B \leq 7T$</u> : <u>LRO</u> !

1st order ?
structural ?

<u>$B \leq 4T$</u> : <u>no</u> LRO?

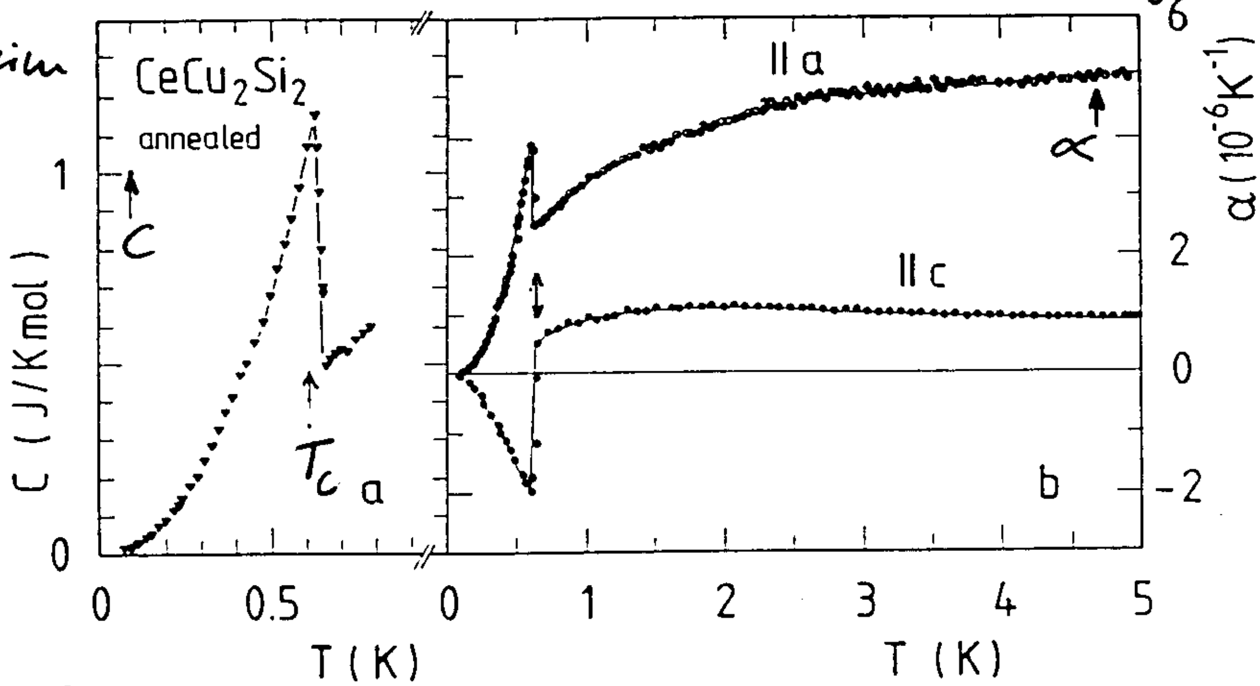
<u>$B \to 0$</u> : $T_1' \longrightarrow T_c = 0.63K$

2a) $CeCu_2Si_2$ single crystal (W. Assmus)

annealed bulk sc, $T_c = 0.63\,K$

M. Lang

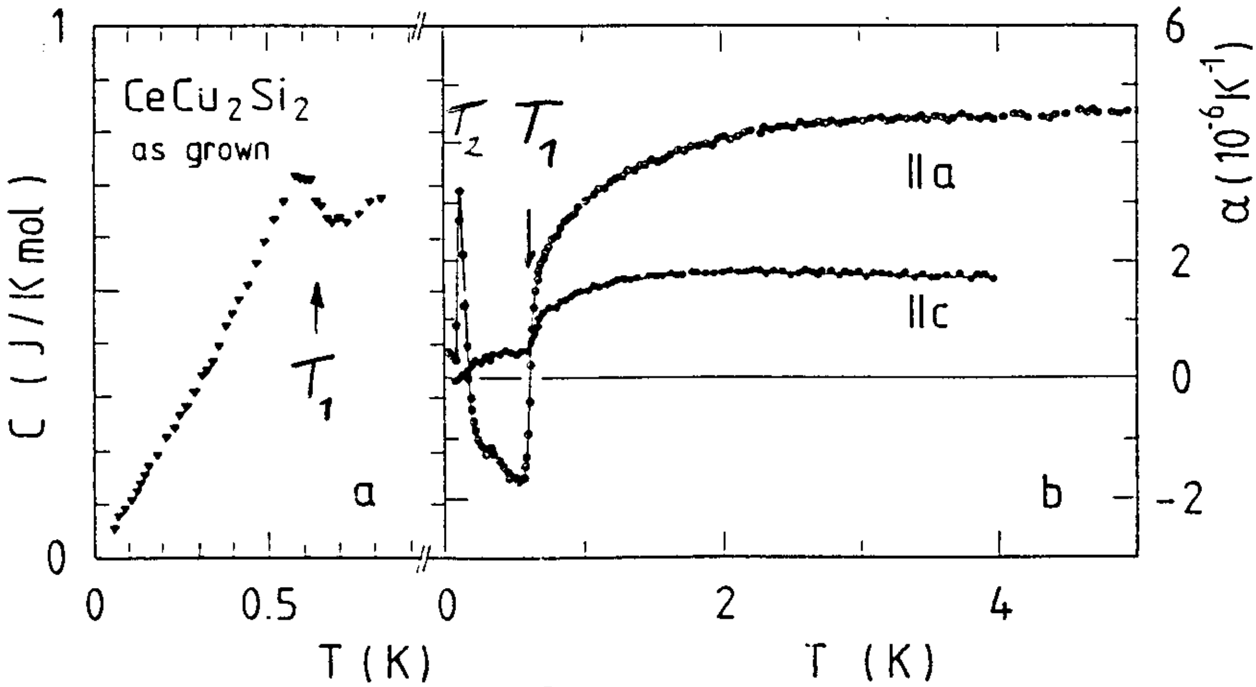
u. Ahlheim




$\frac{\partial T_c}{\partial p_a} > 0 \quad \frac{\partial T_c}{\partial p_c} < 0 : \left(\frac{\partial T_c}{\partial p_h}\right)_{p_h \to 0} \simeq +4\ \frac{mK}{kbar}$

(cf. Eichler '85)

„as grown" non-bulk sc




$T_1 = 0.63\,K \; ; \left(\frac{\partial T_1}{\partial p_h}\right)_{p_h \to 0} = -200\ \frac{mK}{kbar}$

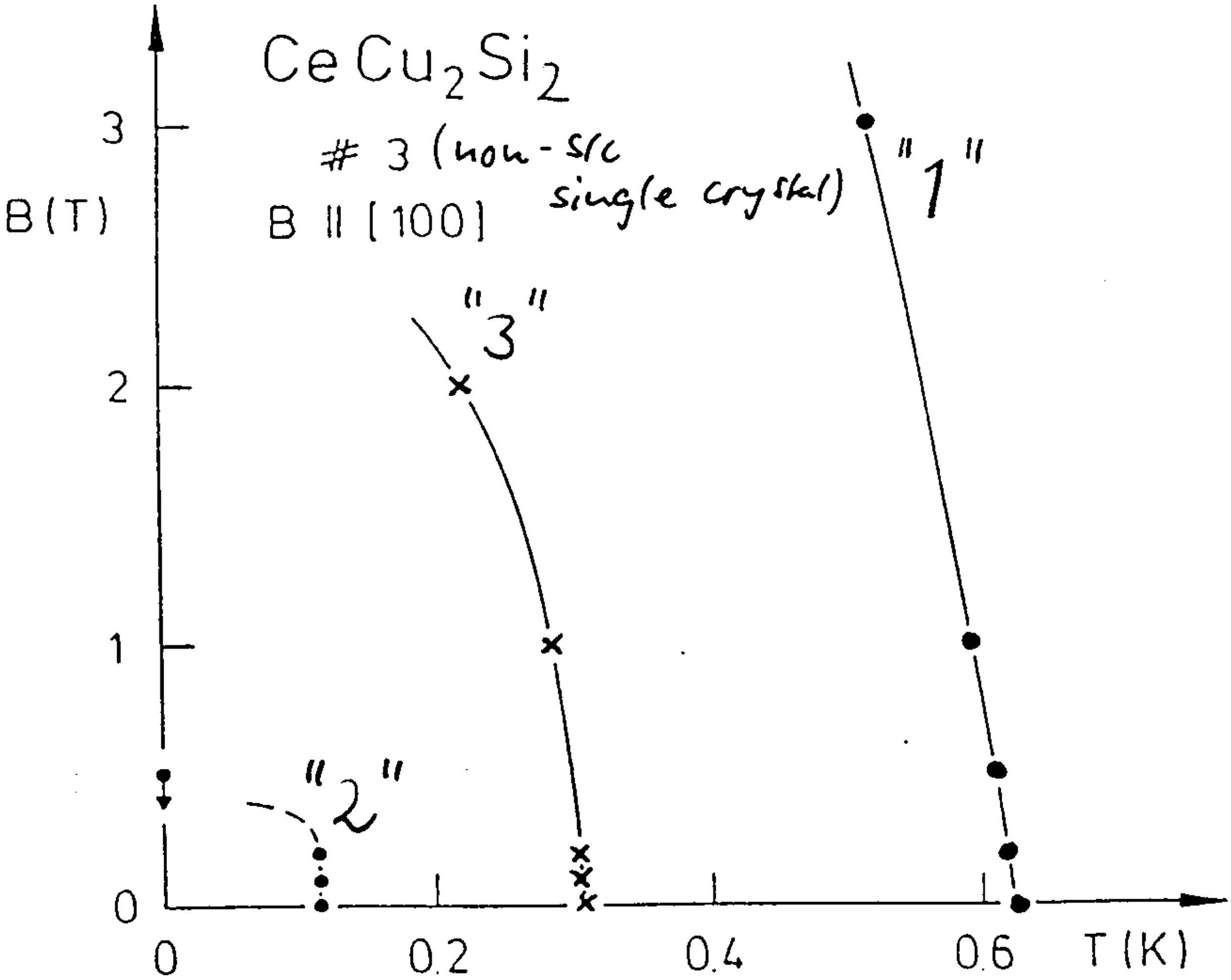
$T_2$ : afm?

lattice instability

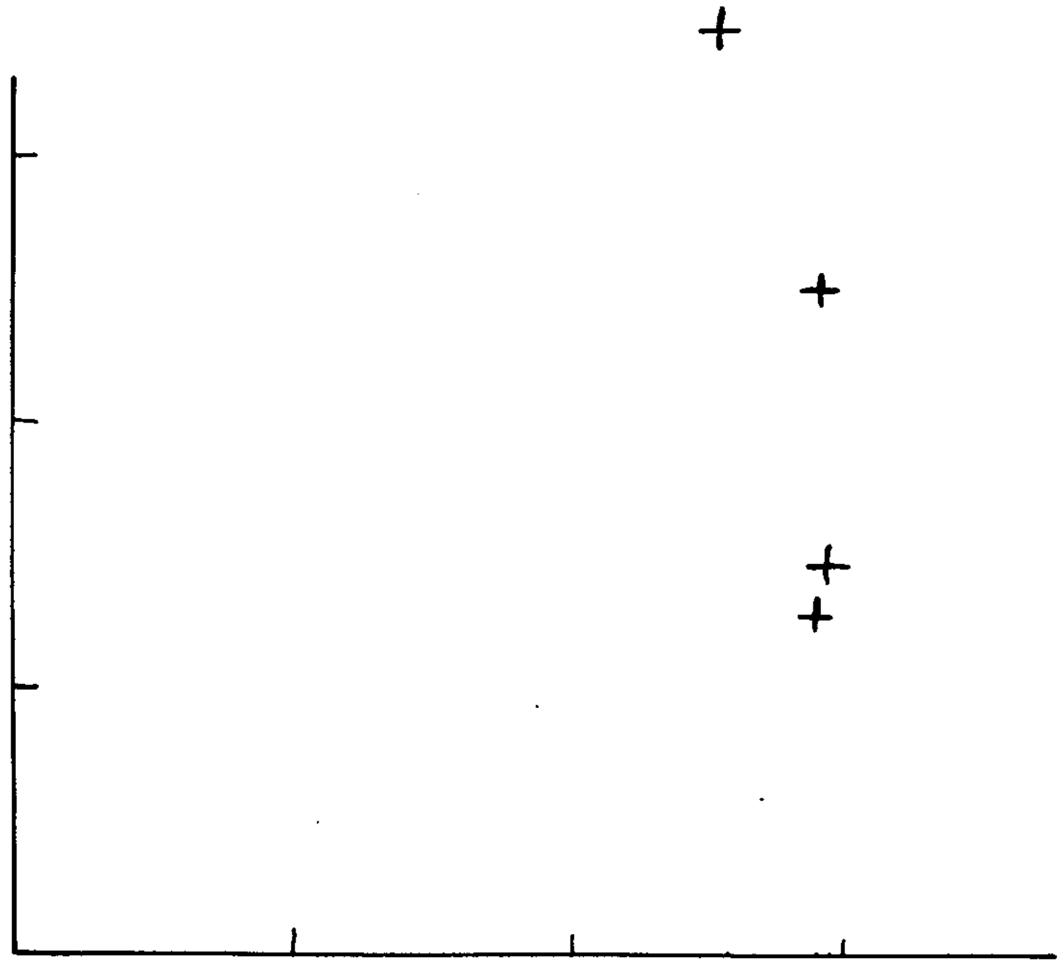
"1" : $\left(\frac{\partial T_1}{\partial p}\right)_{hydrost.} \simeq -200 \frac{mK}{kbar}$

$\Rightarrow$ structural transition?!

"2" : afm ?!



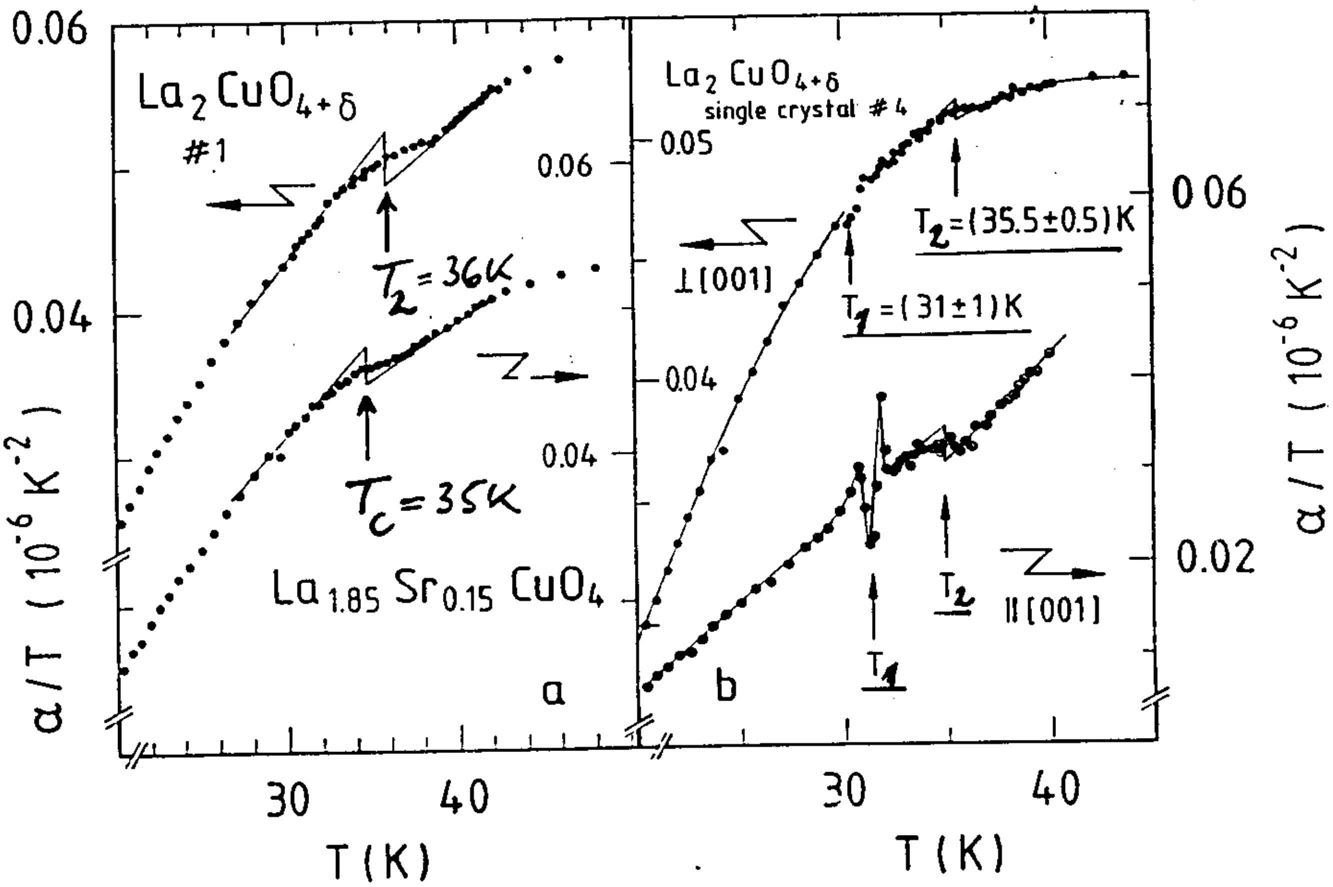

"3" : minority phase ?!



+ NMR : s/c
polycrystal

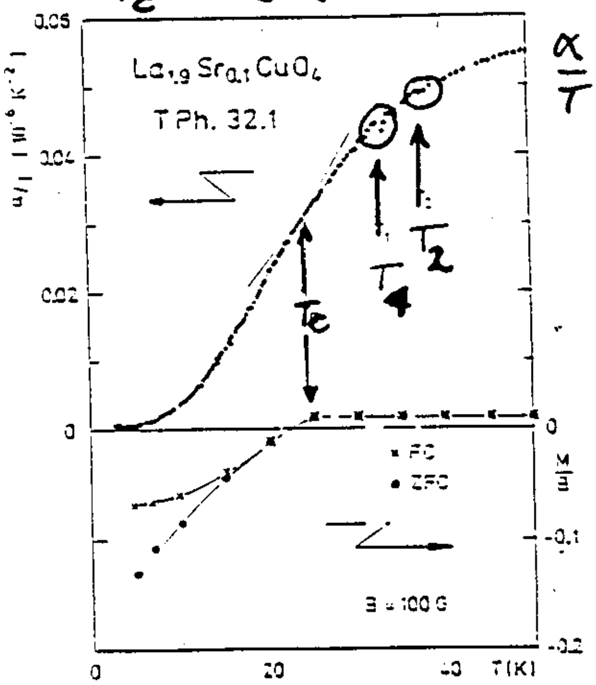
## 2b) Lattice instability and cuprate superconductivity (M. Lang et al. '91)

(Y. Hidaka)




$La_{1.9}Sr_{0.1}CuO_4$ (LTO structure)

$T_c = 25K$




$T_2 = (37 \pm 1)K$
$T_1 = (32 \pm 1)K$

↓

lattice instabilities

- property of stoichiometric $La_2CuO_4$
- independent of hole concentration

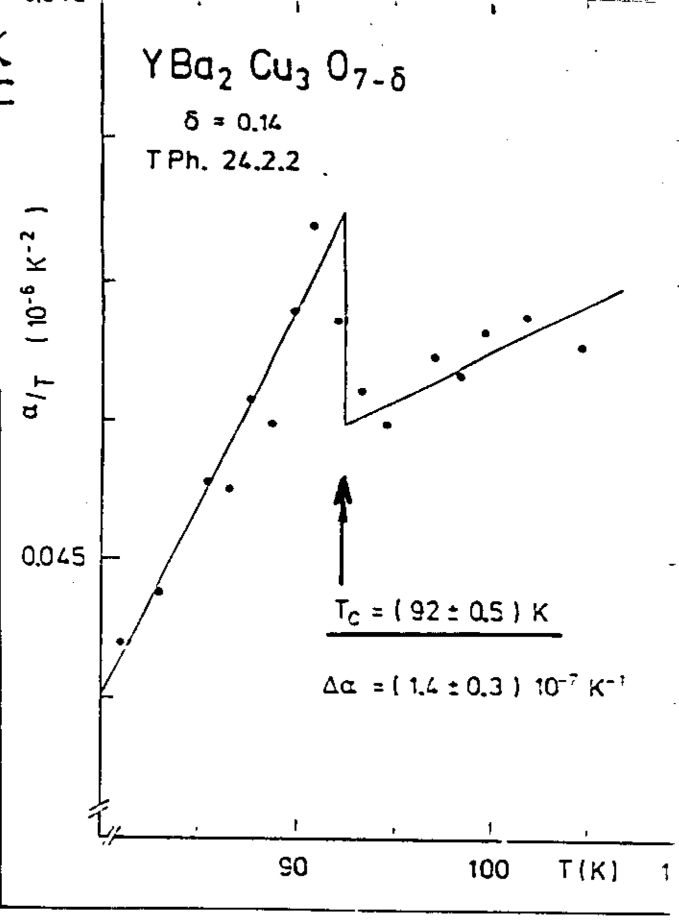
$YBa_2Cu_3O_{6.86}$

(T. Wolf)

$T_c = 92\,K$

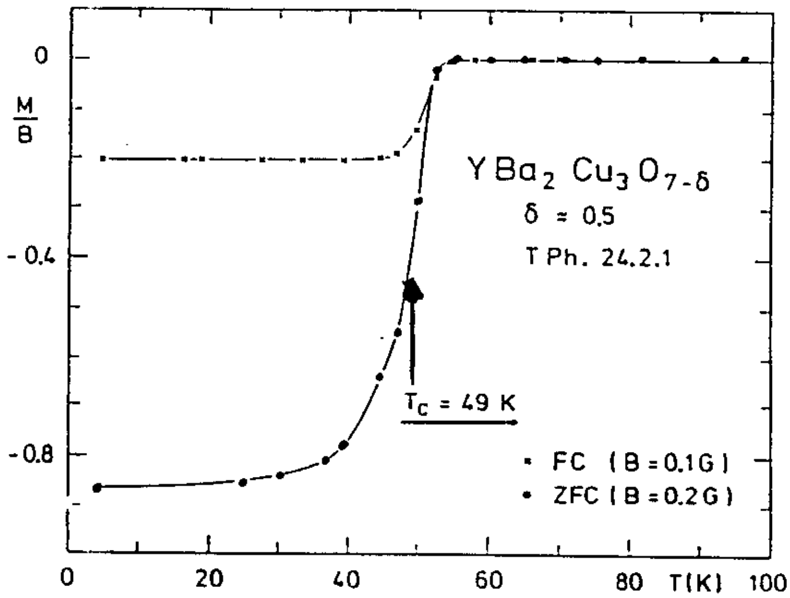
(bulk s/c)

$\frac{\alpha}{T}$

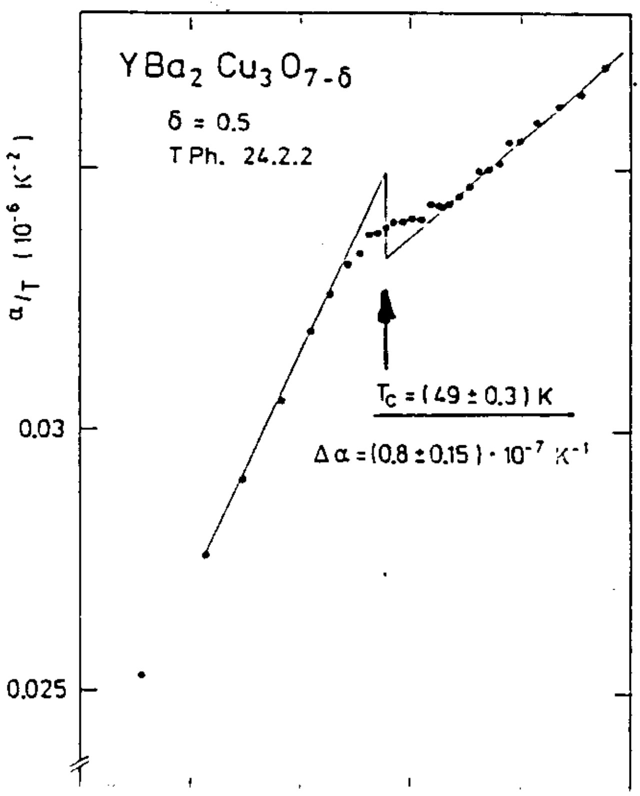


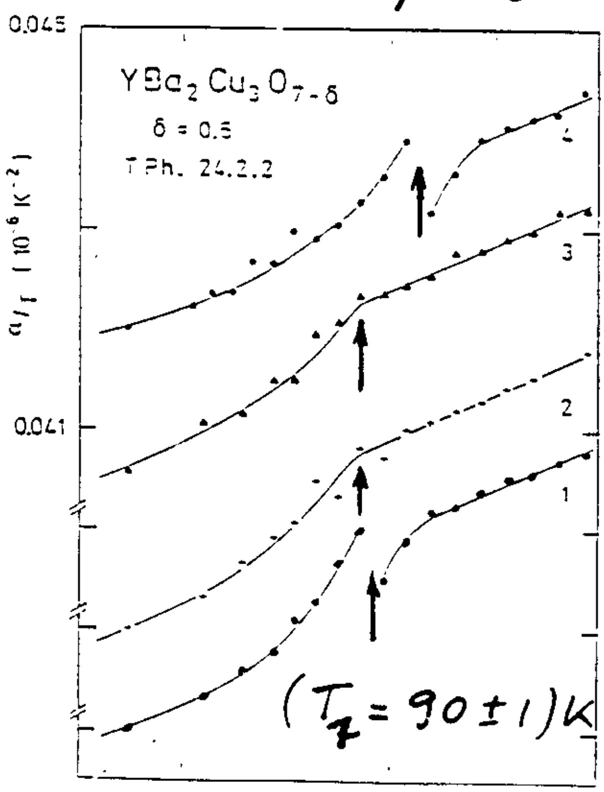

$YBa_2Cu_3O_{6.5}$

$T_c = 49K$




cf. Shaltiel et al. '89
disappearance
of $Cu^{2+}$-ESR at
$T_f = 90\,K$




$(T_f = 90 \pm 1)K$

## 3.) Coexistence of superconductivity and antiferromagnetism in U - compounds

$URu_2Si_2$ $T_c = 1.2\,K$ $T_N = 17\,K$

easy c-axis

$\mu_S \sim 3 \cdot 10^{-2} \mu_B$

(C. Broholm et al. '87)

$UPt_3$ $T_c = 0.5\,K$ $T_N = 6\,K$

easy (a,b) plane

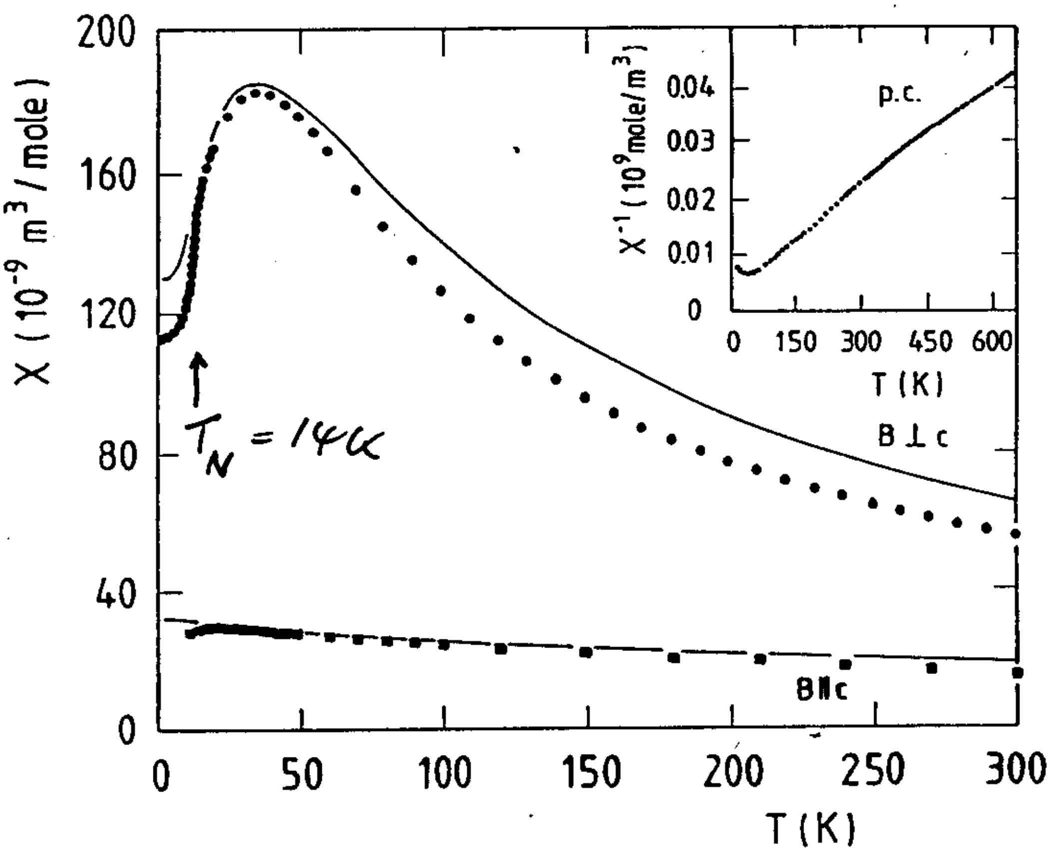
$\mu_S \sim 2 \cdot 10^{-2} \mu_B$

(G. Aeppli et al. '88)

$$T_c \sim 0.1\, T_N \;?$$

$\Rightarrow$ $UM_2Al_3$ (M: Ni, Pd) (C. Geibel et al. '91)

## • magnetic susceptibility




fit: $\chi^{-1} = \chi_{CF}^{-1} + a^{MF}$

$U^{4+}$ ($5f^2$)

CF levels

| | |
|---|---|
| 1006 K | $\vert\Gamma_5\rangle' = 0.2597\ \vert\mp 2\rangle + 0.9657\ \vert\pm 4\rangle$ |
| 562 K | $\vert\Gamma_3\rangle = 1/\sqrt{2}\ (\vert+3\rangle + \vert-3\rangle)$ |
| 152 K | $\vert\Gamma_5\rangle = 0.9657\ \vert\mp 2\rangle - 0.2597\ \vert\pm 4\rangle$ |
| 102 K | $\vert\Gamma_6\rangle = \vert\pm 1\rangle$ |
| 33 K | $\vert\Gamma_1\rangle = \vert 0\rangle$ |
| 0 K | $\vert\Gamma_4\rangle = 1/\sqrt{2}\ (\vert+3\rangle - \vert-3\rangle)$ |

inter-site correlations through

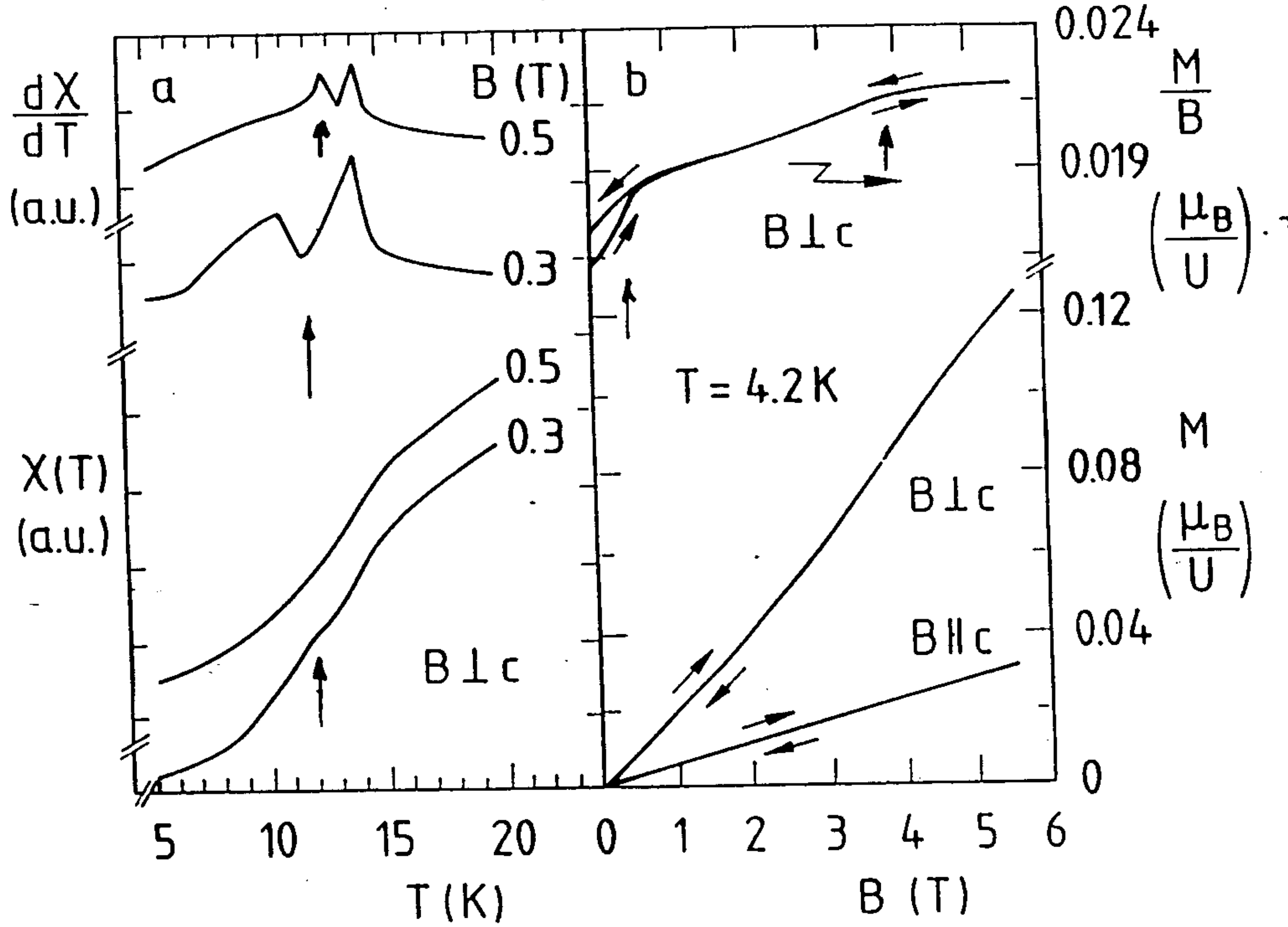
$a_{\parallel}^{MF} = 2 \cdot 10^7$ mole/m$^3$ ; $a_{\perp}^{MF} = 2.5 \cdot 10^6$ mole/m$^3$

B - field dependence of $\chi(T)$

magnetization $M(B)$

$UPd_2Al_3$ single crystal ($B \perp c$)

(N. Sato, T. Komatsubara)

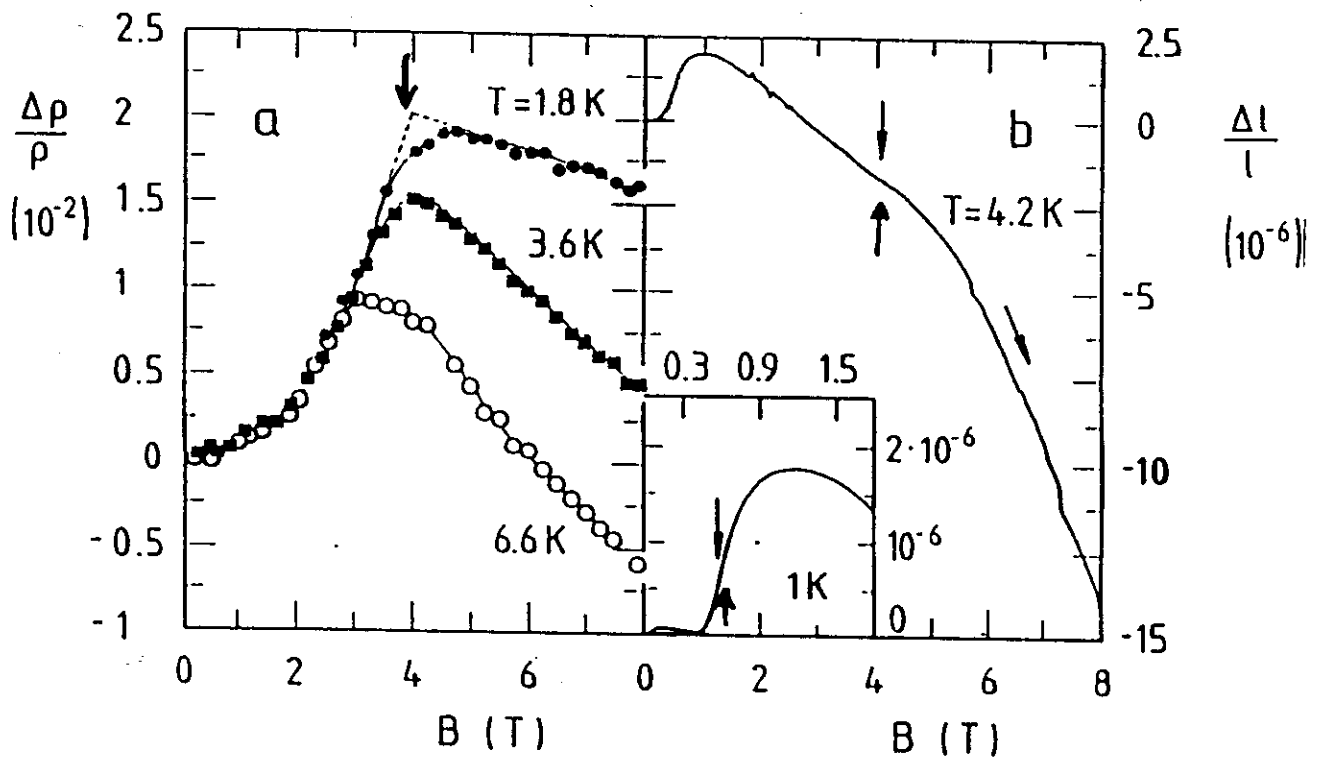



↑ : I ↔ II

**↑** : II ↔ III

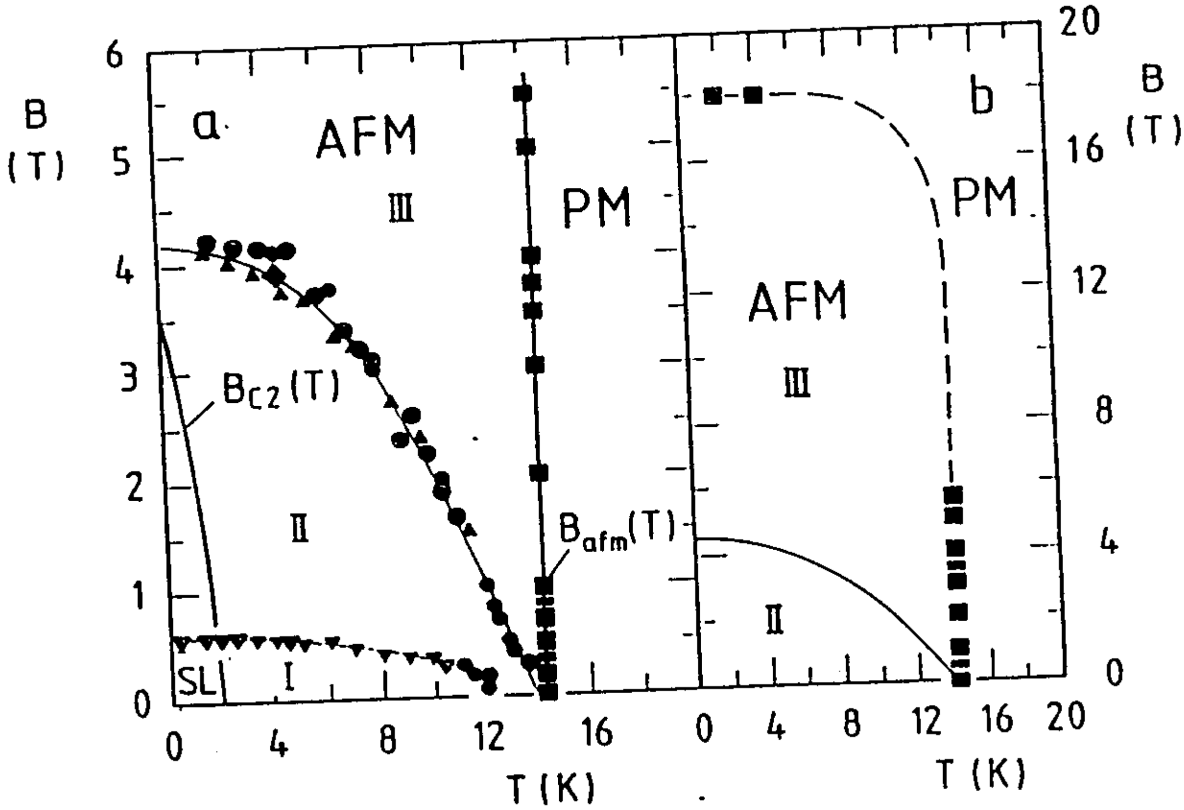
# $UPd_2Al_3$ single crystal ($B \perp c$)

transverse magnetoresistivity | magnetostriction



# Magnetic phase diagram of $UPd_2Al_3$ single crystal ($B \perp c$)

■ : M(B) de Visser et al. 92




## in-plane afm structures (hypothesis)

I : fm planes, + - + - along c : $(0, 0, \frac{1}{2})$
(A. Krimmel et al. 92 : n-diffr.)

II : collinear : $\vec{q} = (\frac{1}{\sqrt{3}}, 0, \frac{1}{2})$ (L-point)

III : canted : $\vec{q} = (\frac{1}{\sqrt{3}}, \frac{1}{3}, \frac{1}{2})$ (H-point)

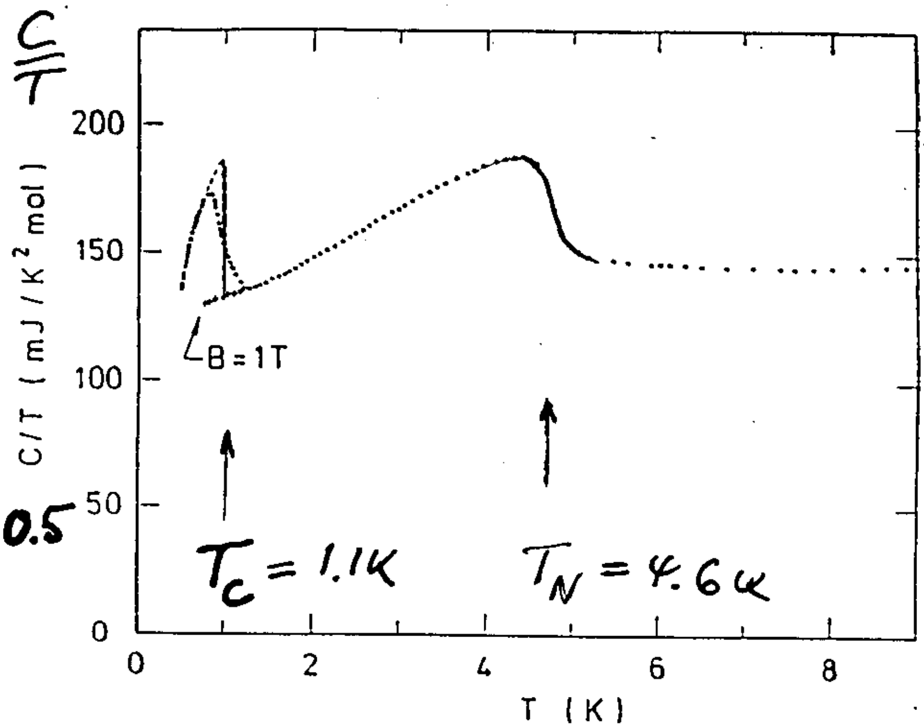
I, II : proposed by Sticht and Kübler through LDA calculation ('92)

## $UNi_2Al_3$

$\gamma_p \simeq 150 \ \frac{mJ}{K^2 mol}$

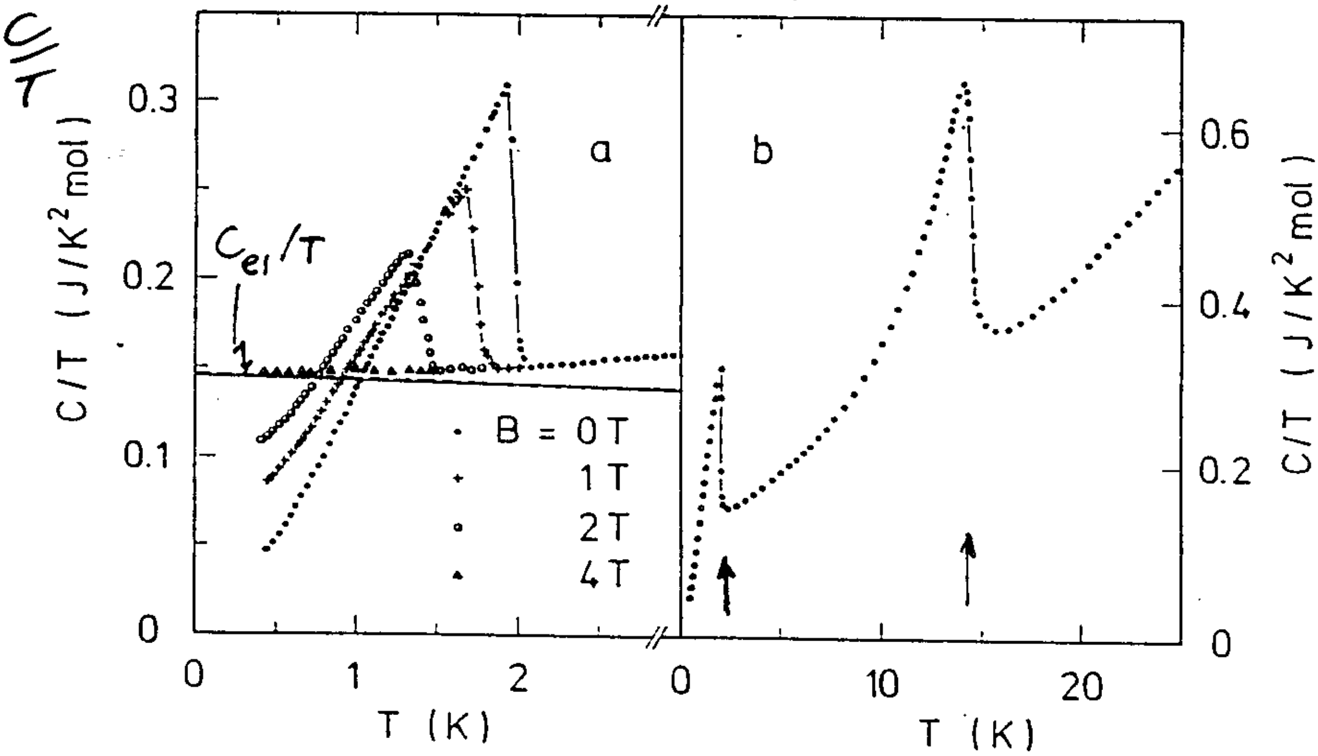
$\gamma_0 \leq 120$ "

$\frac{\Delta C}{C_{el}(T_c)} \geq \frac{\Delta C}{C_n(T_c)} = 0.5$




## $UPd_2Al_3$

$\gamma_0 = 150 \ \frac{mJ}{K^2 mol}$ $\qquad$ $\gamma_p = 210 \ \frac{mJ}{K^2 mol}$

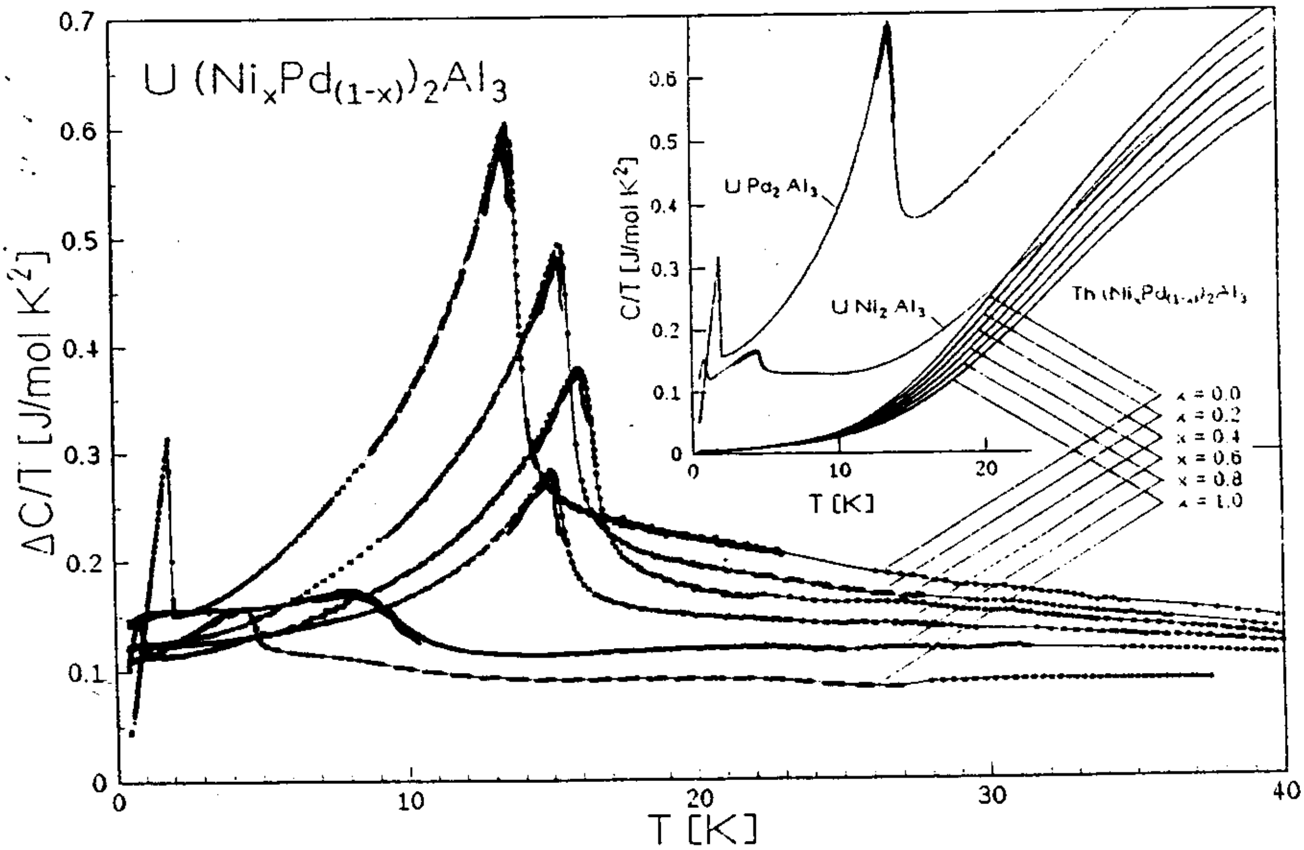



$\frac{\Delta C}{C_{el}(T_c)} = 1.4$ $\qquad$ $T_c = 2K \quad T_N = 14K$

# Specific heat of $U(Pd_{1-x}Ni_x)_2Al_3$

A. Böhm, C.D. Bredl

entropy at $T_N$ : $\left.\begin{matrix} 0.67 \\ 0.13 \end{matrix}\right\} R\ln 2$, M: $\begin{matrix} Pd \\ Ni \end{matrix}$



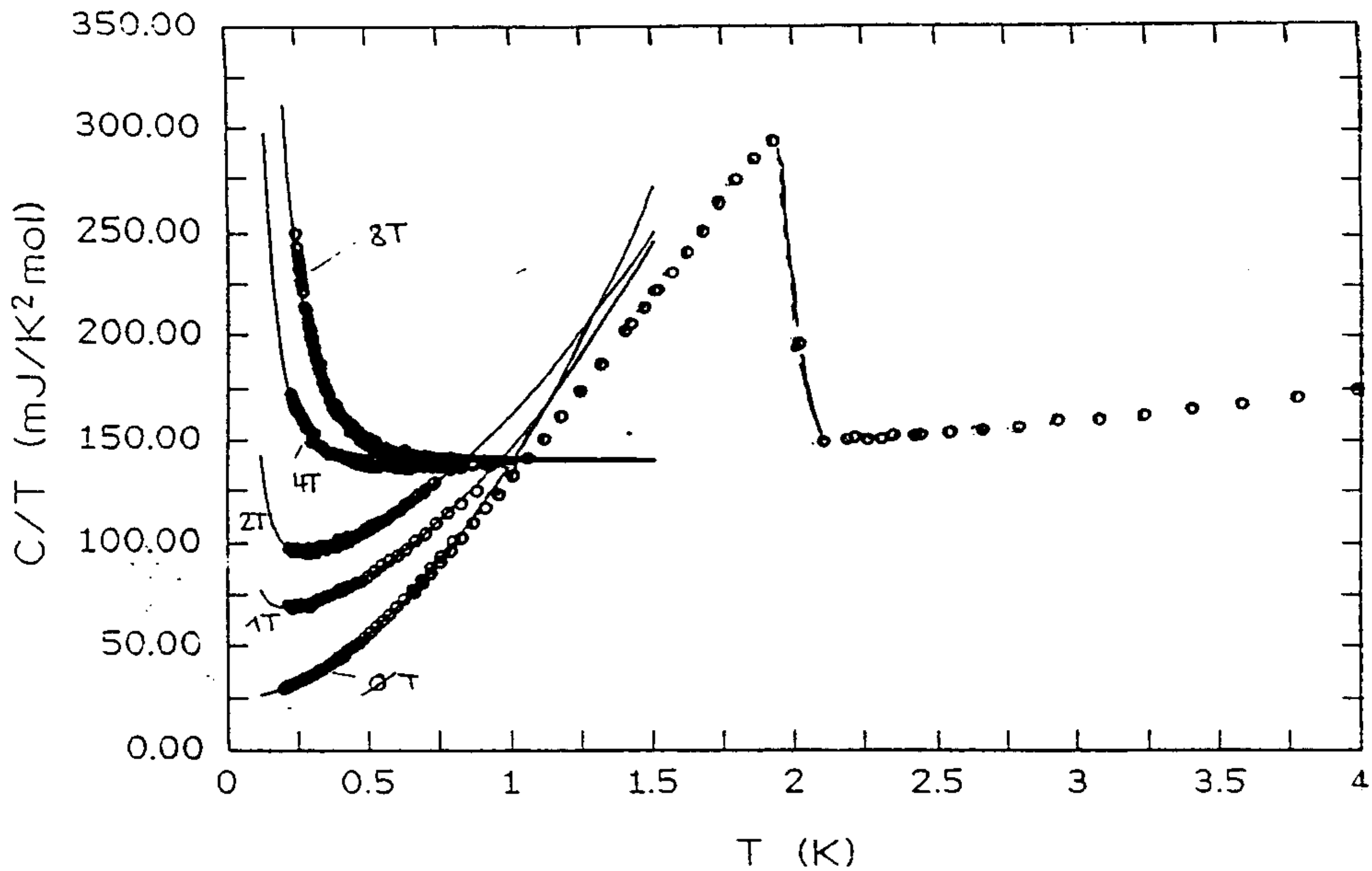

M = Ni : $\mu_s = 0.1\,\mu_B$ ($\parallel c$)

μSR (A. Amato et al. '92)

M = Pd : $\mu_s = 0.85\,\mu_B$ ($\perp c$)

n - scattering (A. Krimmel et al. '92)

## Low-T specific heat (R. Caspary '92)



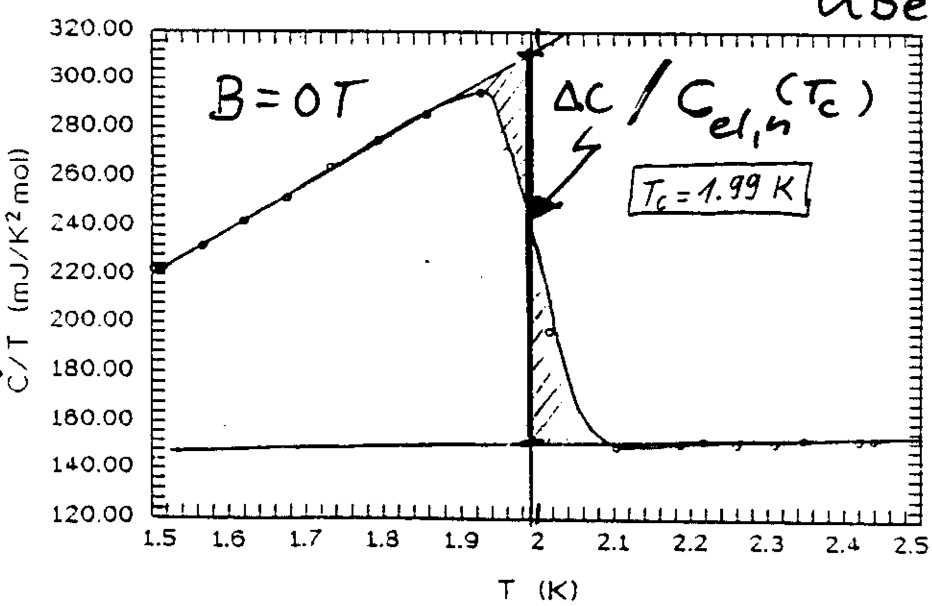

fit: $C = \underbrace{\frac{\alpha(B)}{T^2}} + \underbrace{\gamma_s(B)\cdot T} + \underbrace{\delta(B)\cdot T^{\varepsilon(B)}}$

$\alpha(0) \equiv 0 \quad \gamma_s(0) = 25\,\frac{mJ}{K^2 mol} \quad \varepsilon = 2.88 \pm 0.02$

similar: $CeCu_2Si_2$, $UBe_{13}$

$C^* = C - \gamma_s T$

$C_{el,n} = C^*_n - \underbrace{\beta T^3}_{\text{magnons}}$




$\Downarrow$

$\frac{\Delta C}{C_{el,n}(T_c)} = 1.40$

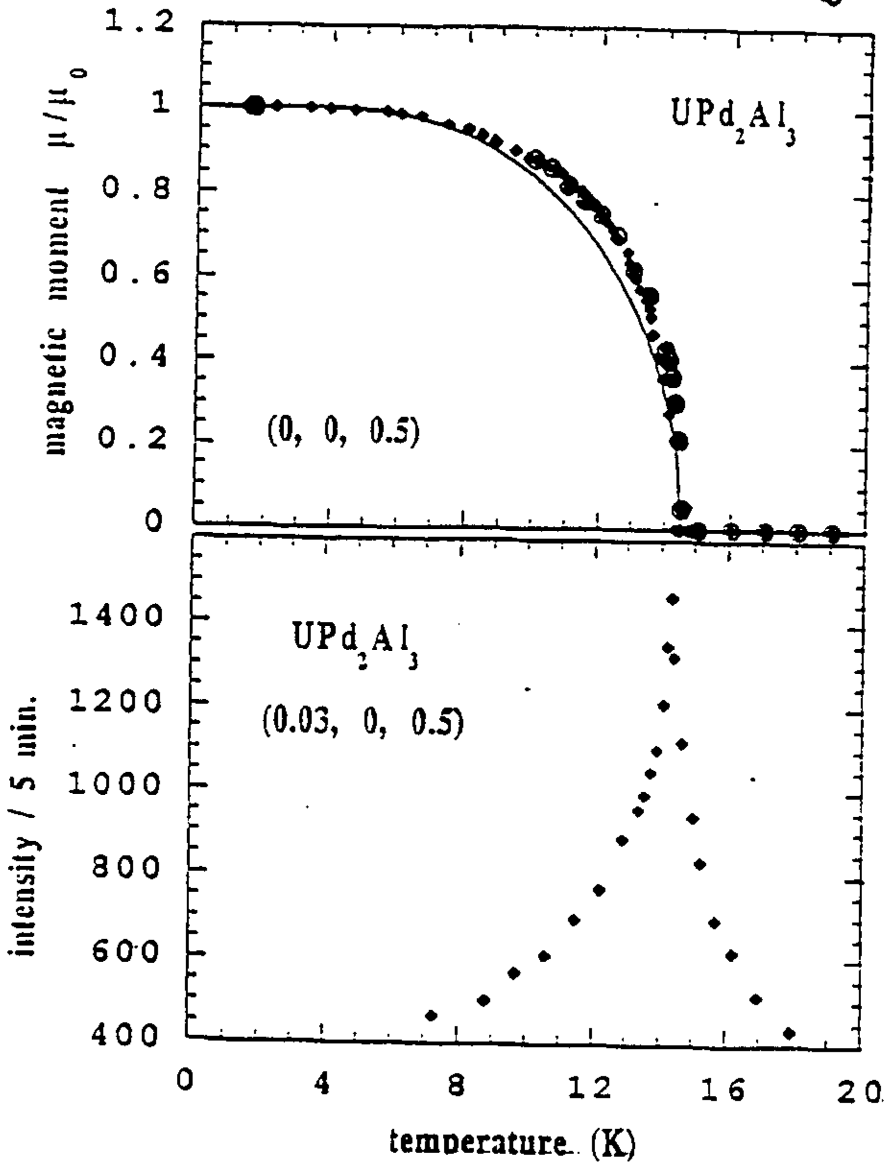
## Neutron diffraction on $UPd_2Al_3$ single crystal

◆ : TOPAN, JRR3M (Tokai, Jpn.), $\lambda_i = 2.44$ Å   A. Dönni et al. '92

○ : P2AX, Saphir (PSI, Switzerl.) $\lambda_i = 1.03$ Å   A. Krimmel et al. 92

magn. Bragg intensity

diffuse critical scattering




$\underline{T > T_N}$ : $\quad t := \frac{T - T_N}{T_N} \qquad \log \frac{\mu}{\mu_0}$ vs $\log t$

(MF) $\qquad t < 0.06$ : $(\mu/\mu_0) \sim t^{0.5}$

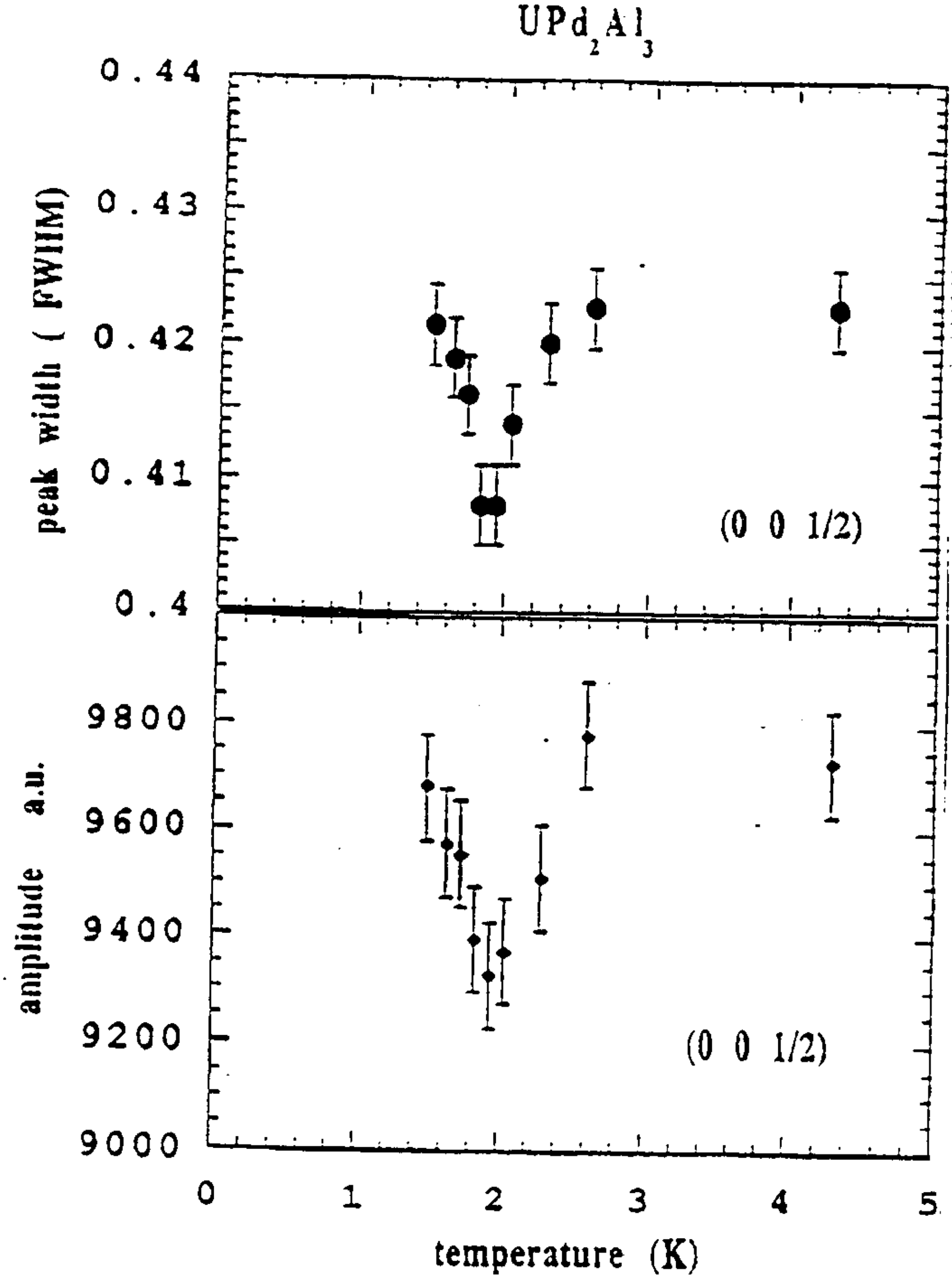
(3D Heisenberg) $\quad 0.06 < t < 0.1$ : $(\mu/\mu_0) \sim t^{0.36}$

"response" of afm to onset of s/c

(0 0 ½) Bragg peak

width

amplitude



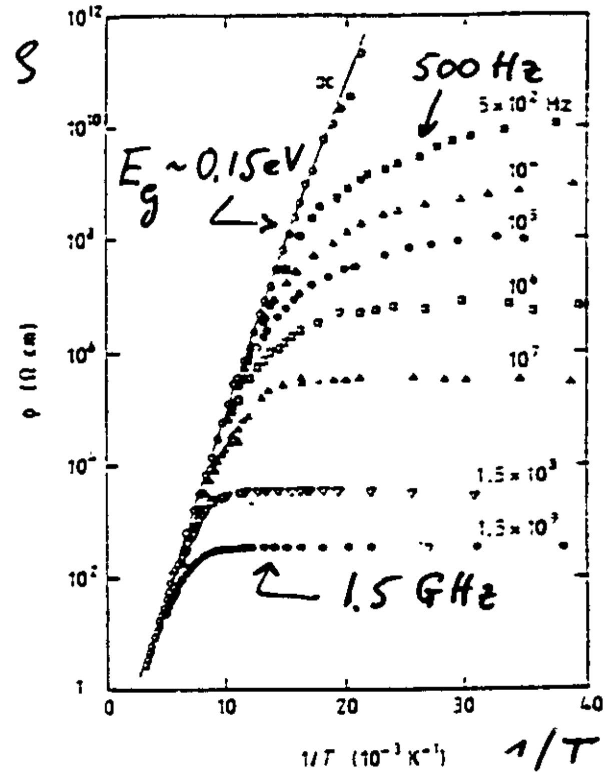
# 4.) Heavy Fermions in Compounds with extremely low carrier concentration

T. Suzuki, T. Kasuya et al.

- $Sm_3X_4$ , X: Se, Te (Th$_3$P$_4$ structure)

$\underbrace{Sm_2^{3+}}Sm^{2+}X_4^{2-}$

$J = 5/2$




Semiconductors!

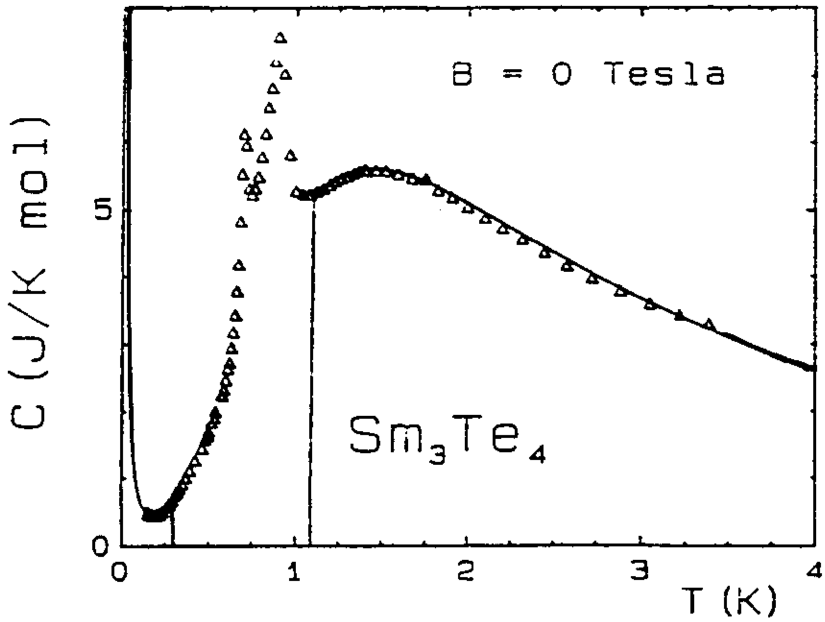
- $Yb_4As_3$ (anti-Th$_3$P$_4$ structure)

$\underbrace{Yb^{3+}}Yb_3^{2+}As_3^{3-}$

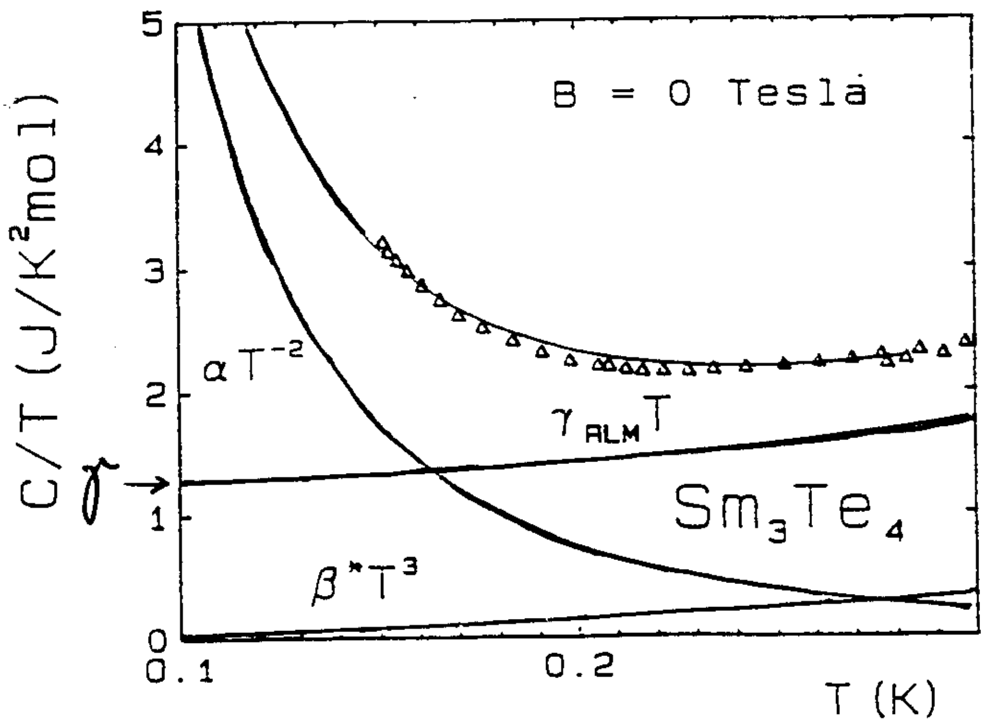
$J = 7/2$

carrier concentration$_{21}$ ($T \lesssim 100K$): $n \simeq \frac{10^{-3}}{Yb}$

## $Sm_3Te_4$ (U. Ahlheim et al. '91)

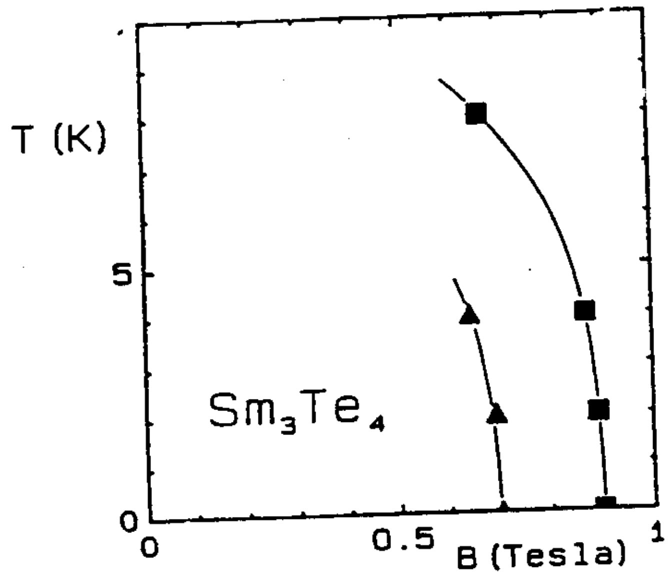



$$C(T) = \frac{\alpha}{T^2} + \beta T^3 + C_{Kondo} \quad \left( \underline{RLM}: \; T_K = 0.8K, \; \frac{E_z}{k_B} = 3.2K \right)$$

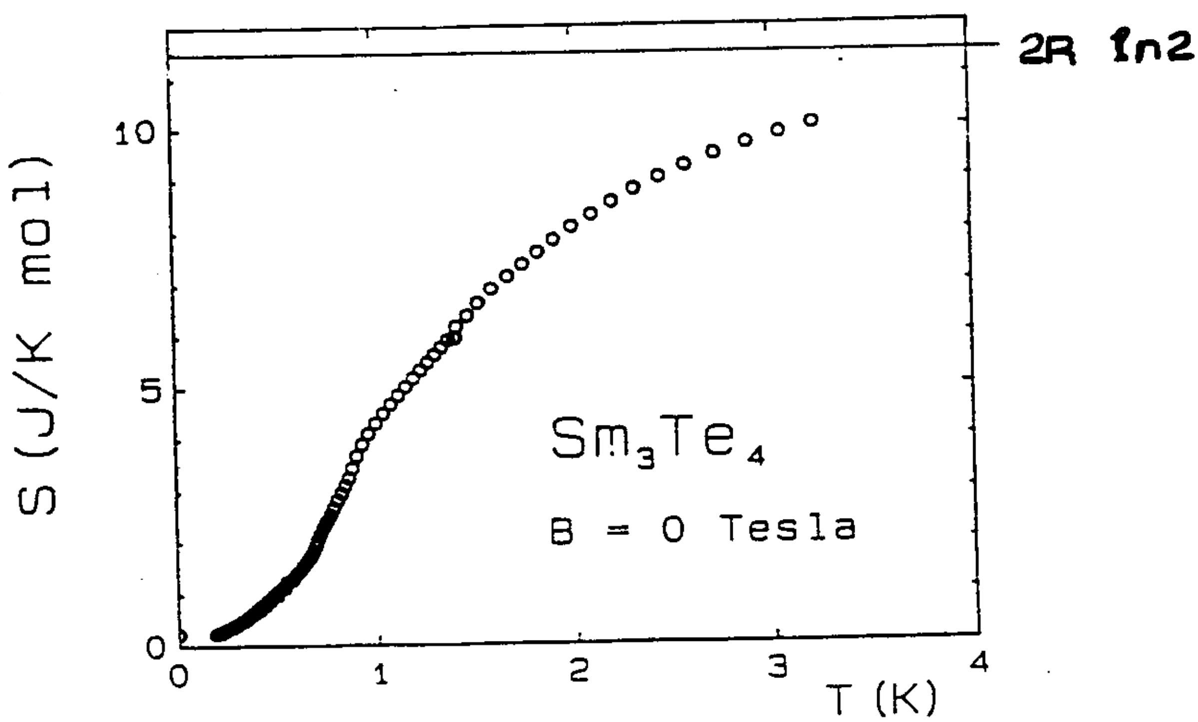



$$\gamma = 0.6 \frac{J}{K^2 \, mol\text{-}Sm^{3+}} \quad \left( Sm_3Se_4 : \gamma = 0.8 \frac{J}{K^2 \, mol\text{-}Sm^{3+}} \right)$$

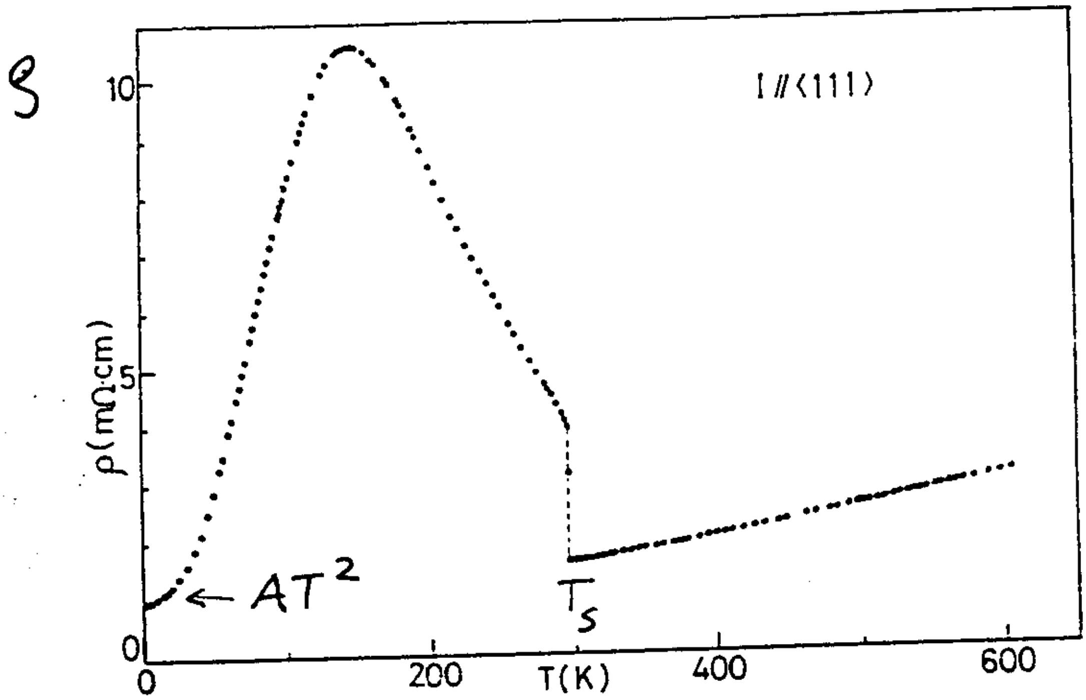
magnetic phase transitions

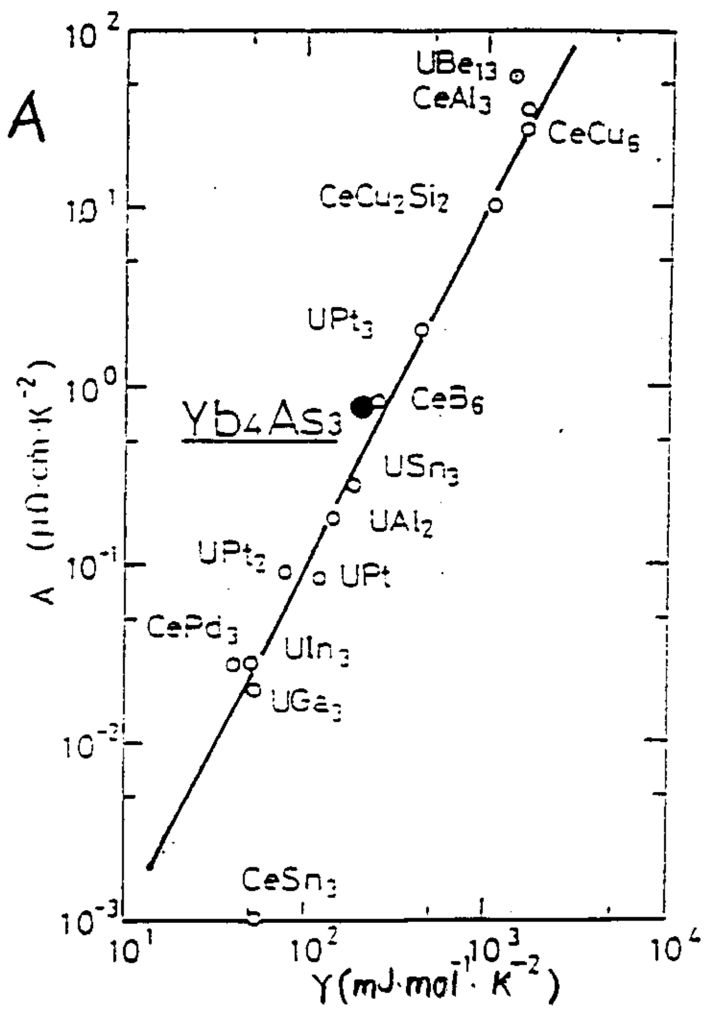



entropy per mole

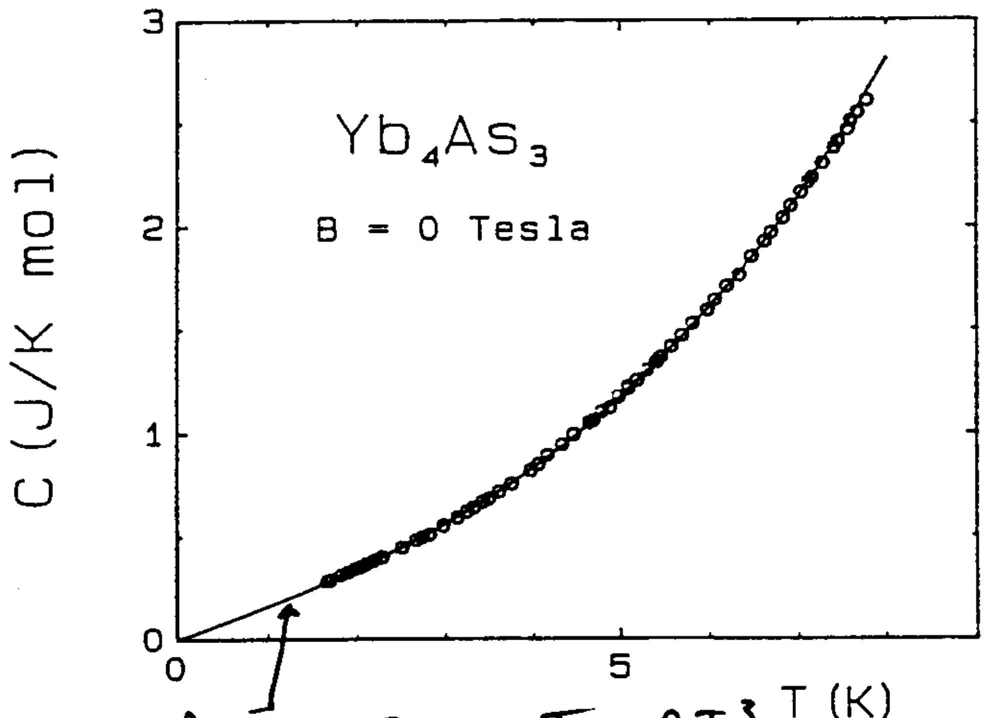


# $Yb_4As_3$ (A. Ochiai et al. '90)

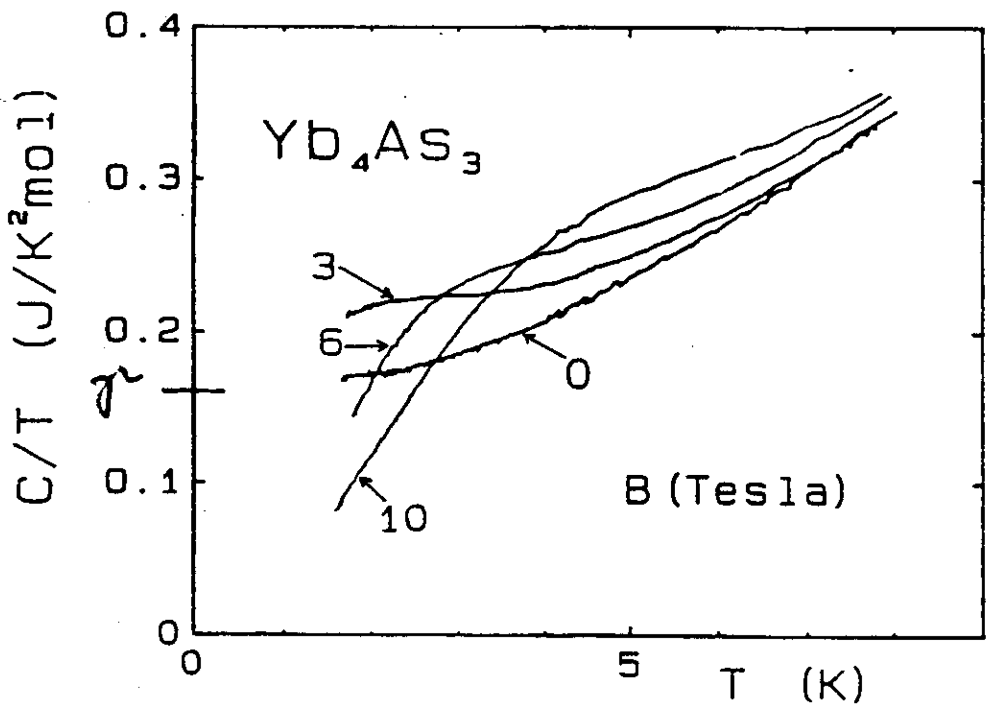


Specific heat $(T \geq 1.5K)$ : T. Suzuki et al. '91

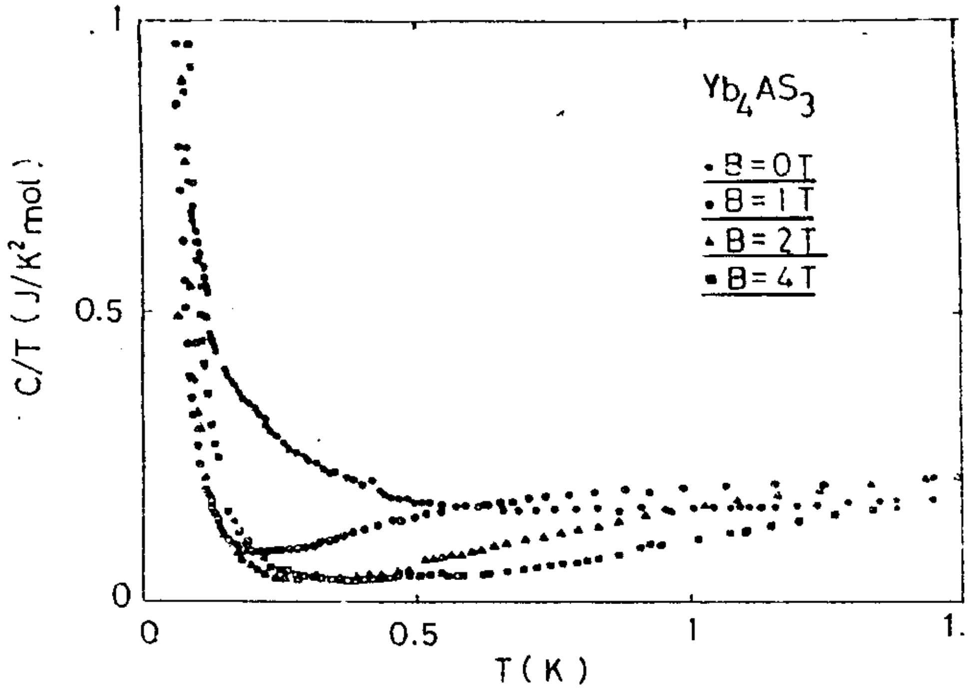



fit : $C = \gamma T + \beta T^3$



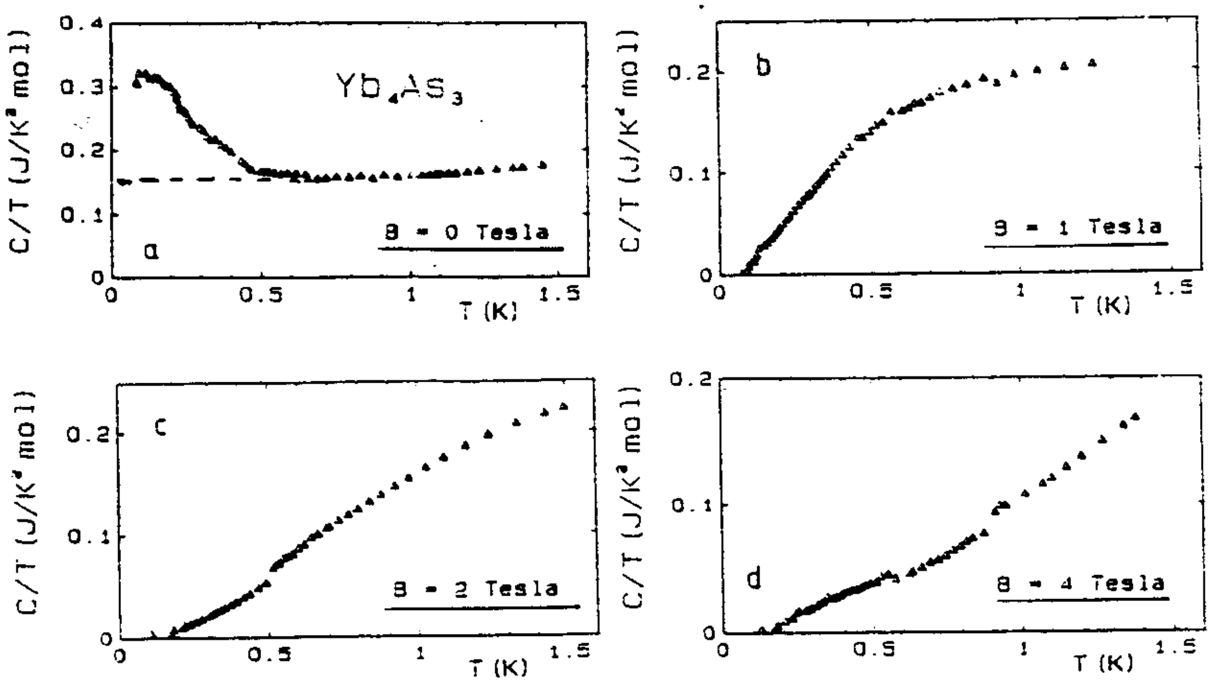

$\gamma \approx 160$ mJ/$K^2$ mole

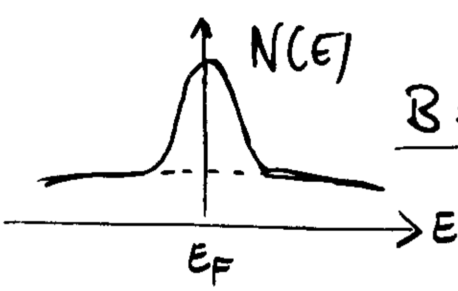
# Low-T specific heat (U. Ahlheim '91)



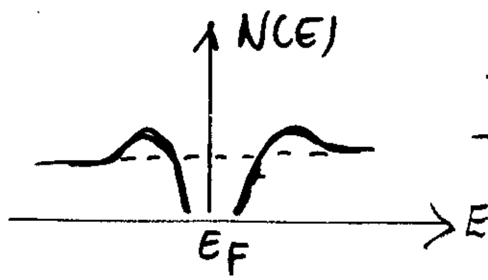

$$C^* = C - \frac{\alpha(B)}{T^2} \qquad T > 0.5\,K:\ \gamma^* = 0.16\,\frac{J}{K^2\,mol}$$

$\frac{C^*}{T}$




B = 0




B > 0

## Summary

**$Sm_3X_4$:** ($n = 0$)

- localized states at $E_F$ with extreme $N(E)$

(nature ?)

- specific heat of $S = 1/2$ Kondo impurity

(intra-atomic Kondo eff.?)

($\xi_{Kondo} \overset{?!}{<} \xi_{loc.}$)

**$Yb_4As_3$:** ($n = 10^{-3}/Yb$)

- $A$ vs $\gamma$ like in metallic HF compoun[ds]
- fine structure in $N(E)$ at $E = E_F$
- B-induced gap

(Kondo - lattice ?)

# perspective

good time for the elephant!


„Irgendwohin, wo es noch viele heile Porzellanläden gibt!“ Horst Haitzinger