Jan 8, 2012 Beijing

Unconventional superconducting characteristics of heavy fermion actinide superconductors

Yoshinori Haga
Advanced Science Research Center
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Collaborators

Np samples :

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S. Ikeda, H. Sakai (JAEA-Tokai)

Pu samples :

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TRU measurements / characterization :

T.D. Matsuda, N. Tateiwa, E. Yamamoto, A. Nakamura
H. Chudo, Y. Tokunaga, S. Kambe, H. Yasuoka (JAEA-Tokai)

Rare earth samples :

F. Honda, R. Settai, Y. Onuki (Osaka Univ.)

Discussions :

K. Gofryk (LANL), J.-C. Griveau, E. Colineau, R. Caciuffo (ITU)
Z. Fisk

Jan 8, 2012 Beijing

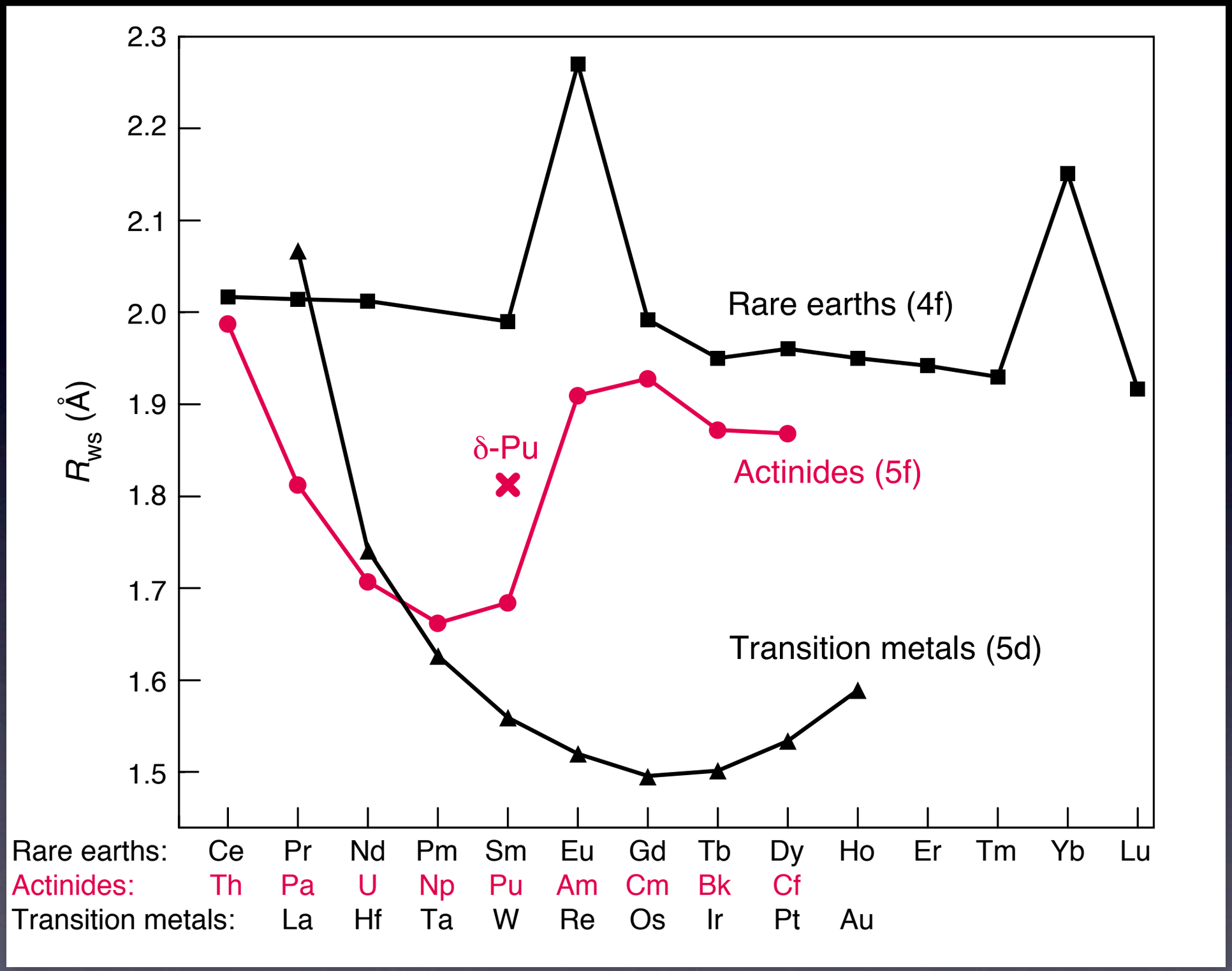
Outline

1. Introduction to actinide material

2. Crystal growth facility

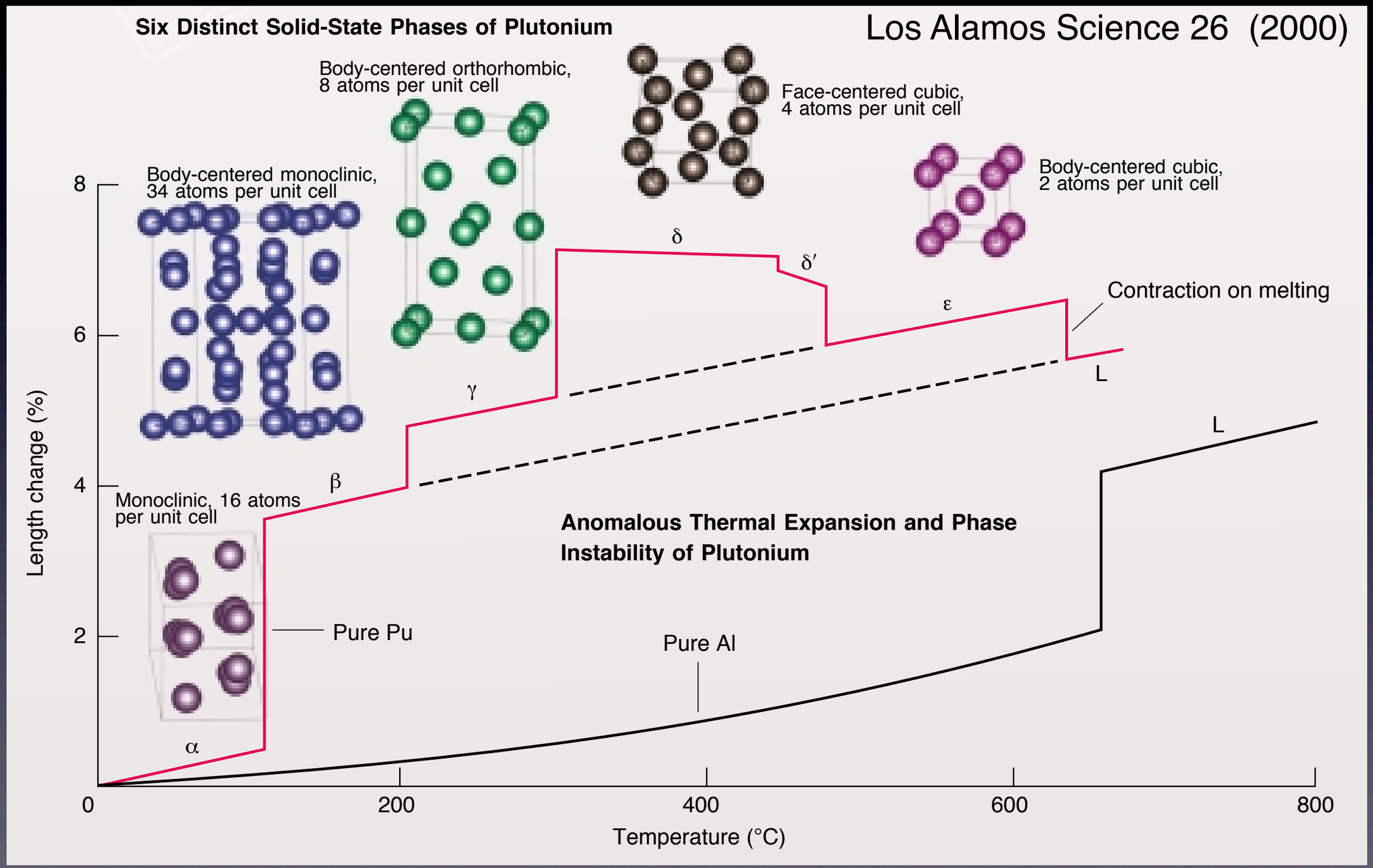
3. Bulk magnetization anomaly in the superconducting state

Wigner-Seitz radius



Los Alamos Science (2000)

4f : La Ce Pr Nd Pm Sm ...
5f : Ac Th Pa U Np Pu ...



Structural Transition in Pu metal

f-electron Heavy Fermion superconductors

	compound	T_c (K)	C_e / T (mJ/K²mol)	T_m (K)
1979	$CeCu_2Si_2$	0.6	~ 1000	
	UPt_3	0.45, 0.55	500	5 (fluctuating)
	UBe_{13}	0.9	1000	
	U_2PtC_2	1.5	75	
1990	URu_2Si_2	1.4	60	17.5
	UPd_2Al_3	2.0	145	14.3
	UNi_2Al_3	1.0	300	4
2001	$CeIn_3$, $CePd_2Si_2$, $CeRh_2Si_2$: pressure-induced			
	$CeCoIn_5$	2.3	500	
	$CeRhIn_5$: pressure-induced			
2002	$CeIrIn_5$	0.4	500	
	$PuCoGa_5$	18.5	70	
	$PuRhGa_5$	8.5	70	
	URhGe	0.15	150	10 (ferro)
	$CePt_3Si$	0.5	300	2.3
	$PrOs_4Sb_{12}$	1.5	~ 500	
2006	$CeTSi_3$ (T = Rh, Ir), $CeCoGe_3$, UIr : pressure-induced			
2007	$NpPd_5Al_2$	4.9	200	
	UCoGe	0.8	150	3 (ferro)

1911 Hg

1961 Nb_3Sn

1986 cuprate

2001 MgB_2

2004 diamond

2008 Fe-As

Sample Preparation

Starting material (available amount)

Th (kg), U (kg), NpO_2 (10 g), ^{239}Pu (10 g), $^{242}PuO_2$ (1 g)

Purification of starting material

Th, U : solid state electrotransport
Np : amalgamation process
Pu : coupled-reduction technique

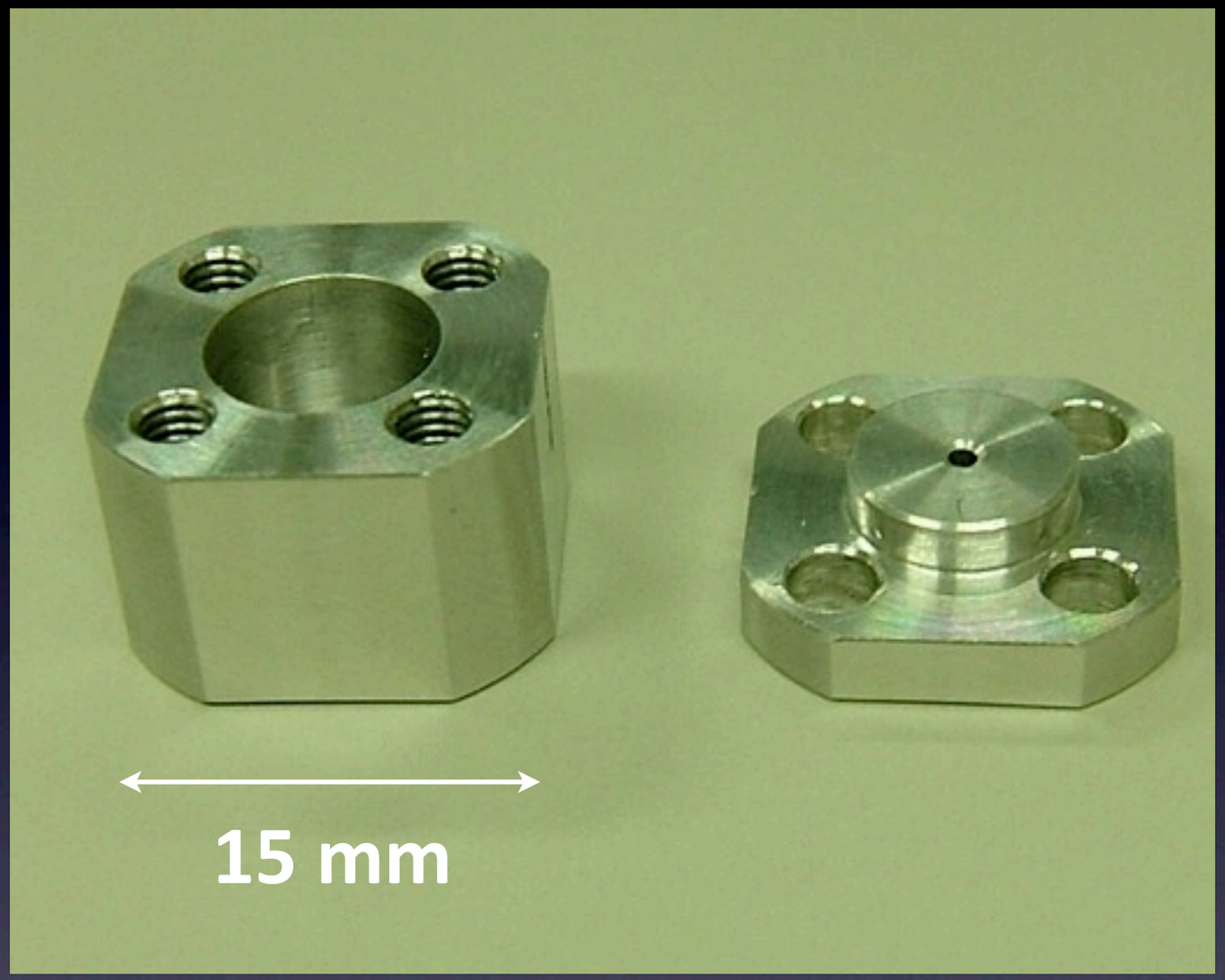
Crystal growth technique

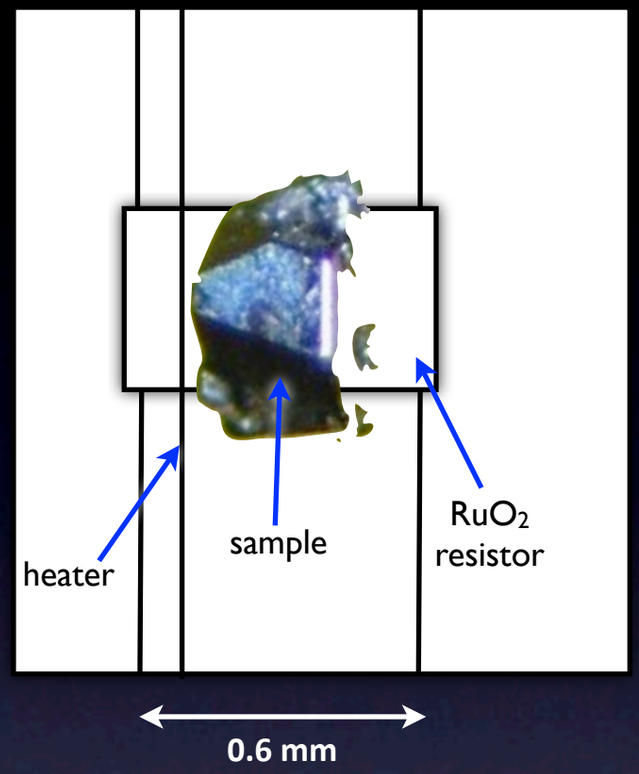
Czochralski pulling: Th & U
Flux growth : all elements
Bridgman growth : all elements
Vapor transport : Th, U, Np

Characterization

XRD (powder / single crystal), EPMA

mini vacuum chamber for Transuranium measurement





specific heat setup
semi quantitative ac-method

metallic container (Al or Bs) (15 x 15 x 15 mm^3)
indium-sealed (no superleak)
electrical feed through
fits in DR and 15 T magnet with rotator

JAEA Tokai

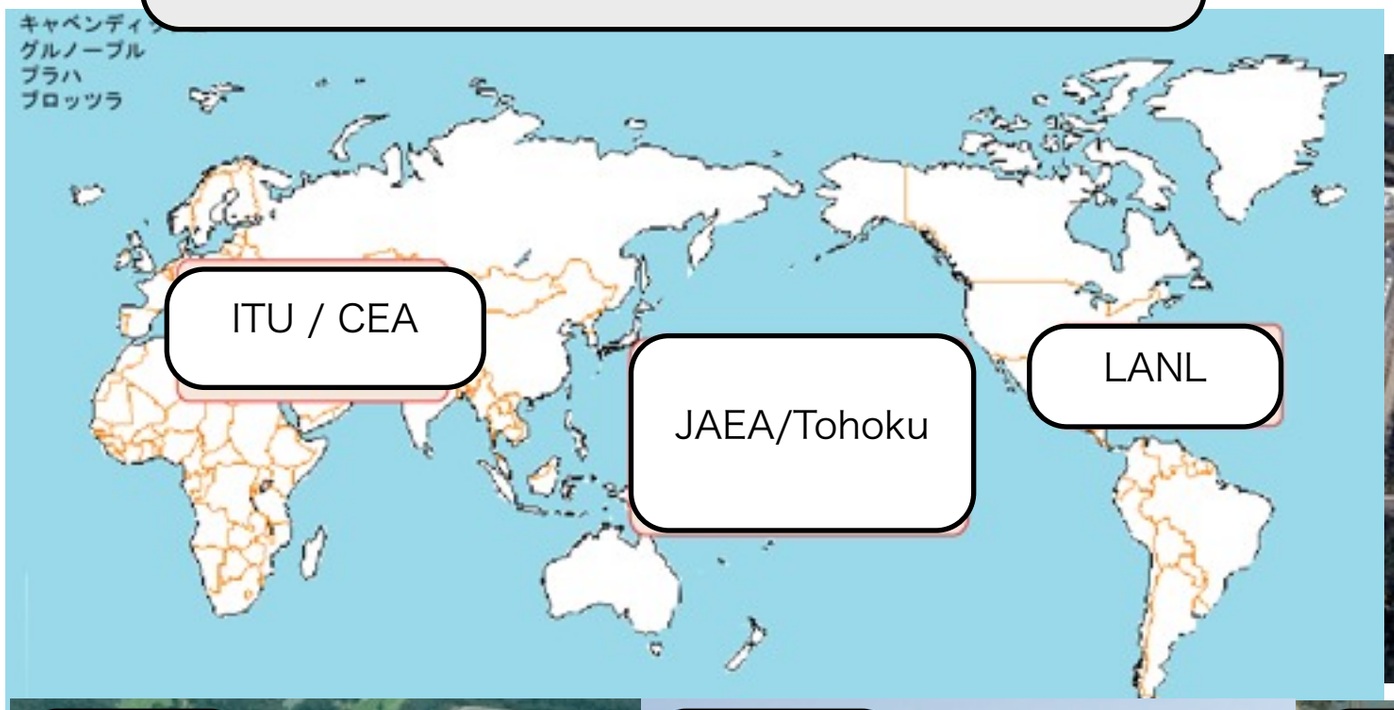
Th, U compounds

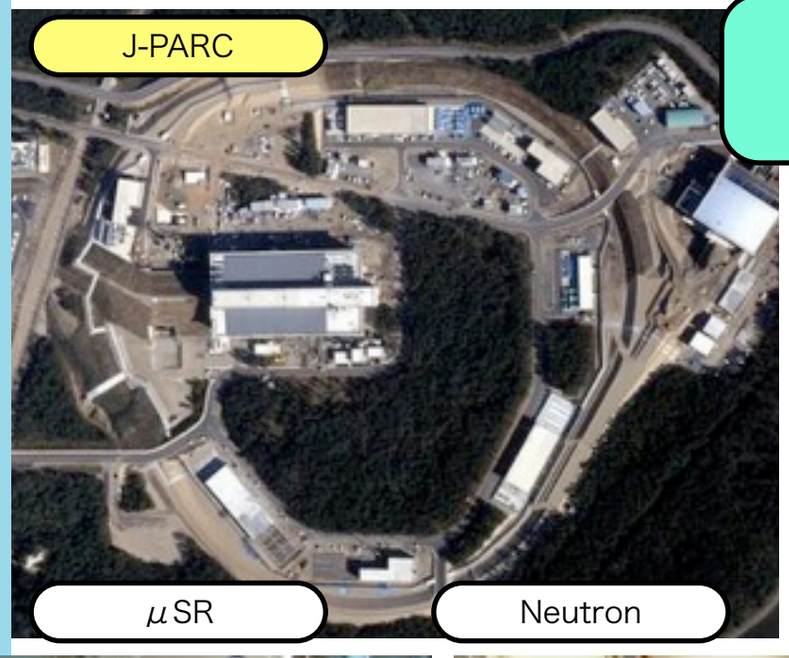
Bulk properties
Characterization
Neutron reactor : JRR-3
Neutron/Muon : J-PARC
High pressure

JAEA Oarai / Tohoku Univ.

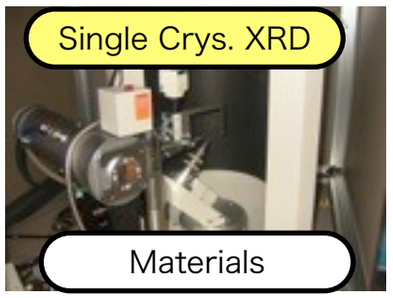
Np, Pu compound

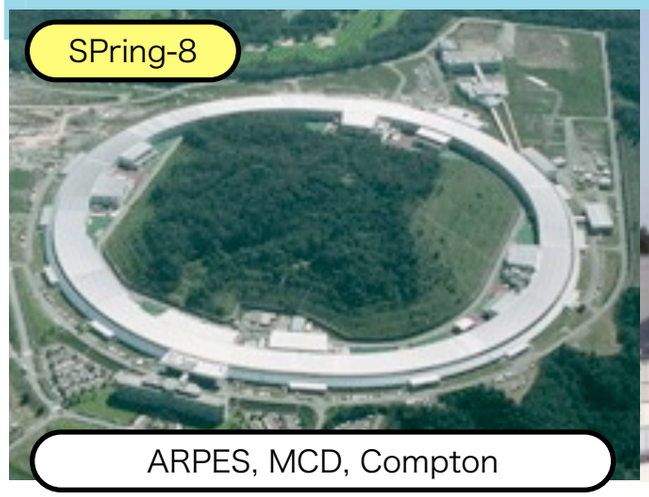
dHvA
NMR/NQR
Mossbauer

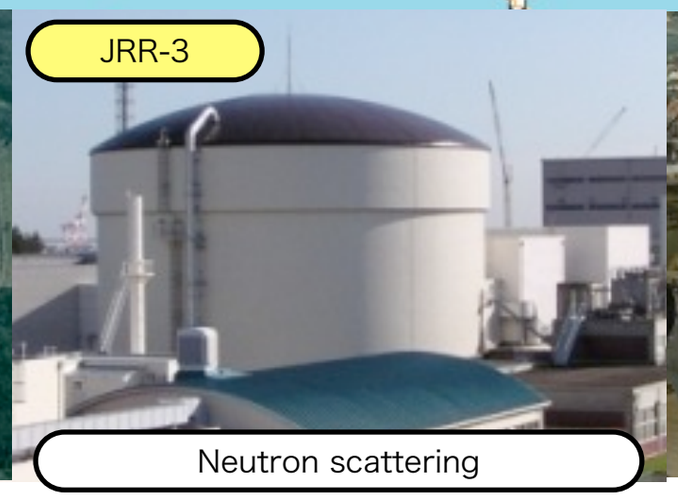


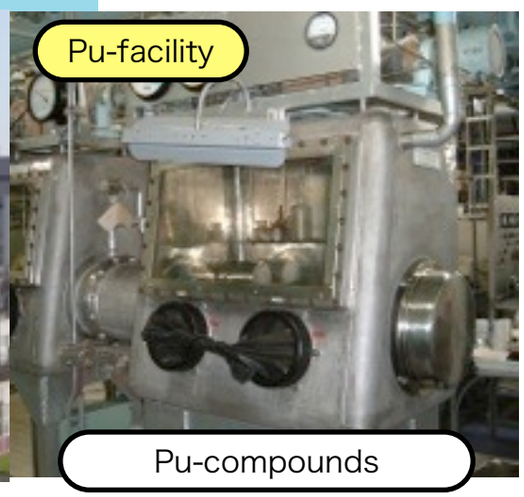


Osaka Univ.
Kyoto Univ.
ISSP Tokyo, etc



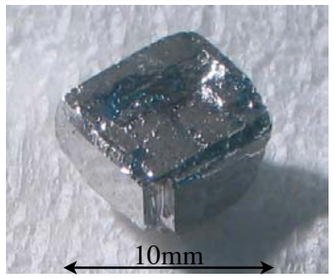




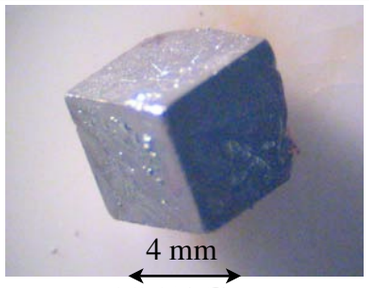


Actinide single crystals

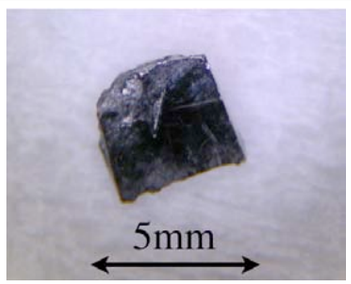
Aoki, Homma, Matsuda, Ikeda, Haga et al. 2003-2011



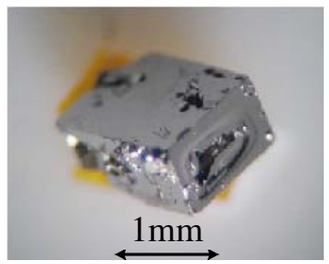
$NpCoGa_5$



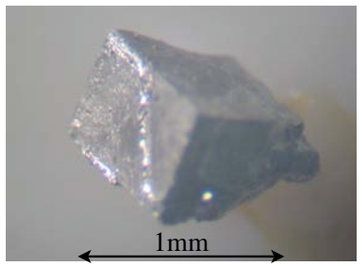
$NpRhGa_5$



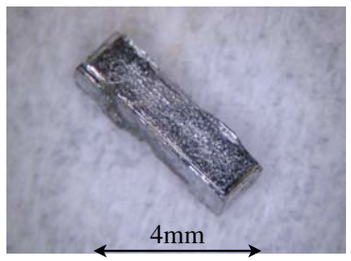
$NpNiGa_5$



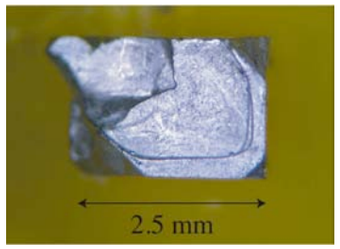
$NpGe_3$



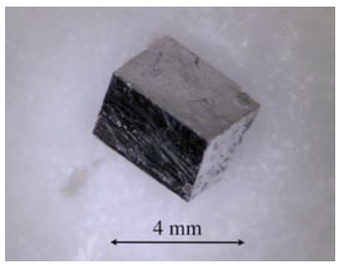
$NpFeGa_5$



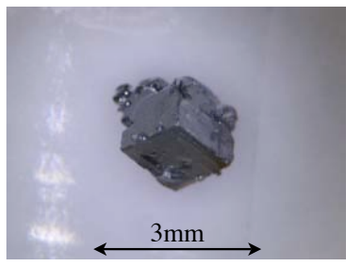
$NpPtGa_5$



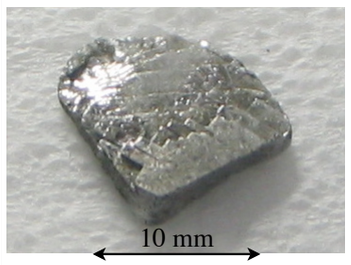
$NpIn_3$



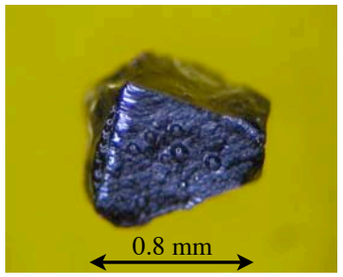
$NpSn_3$



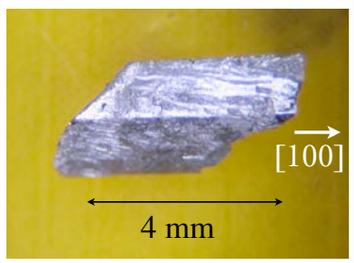
$NpPb_3$



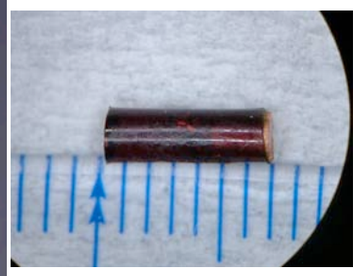
$NpSb_2$



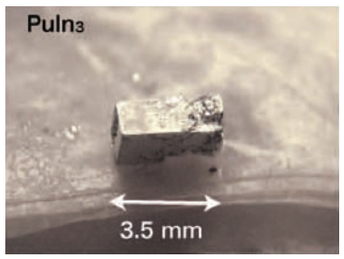
NpO_2



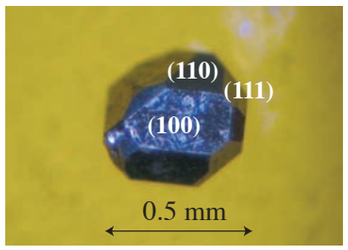
$NpAl_4$



密封された$PuRhGa_5$

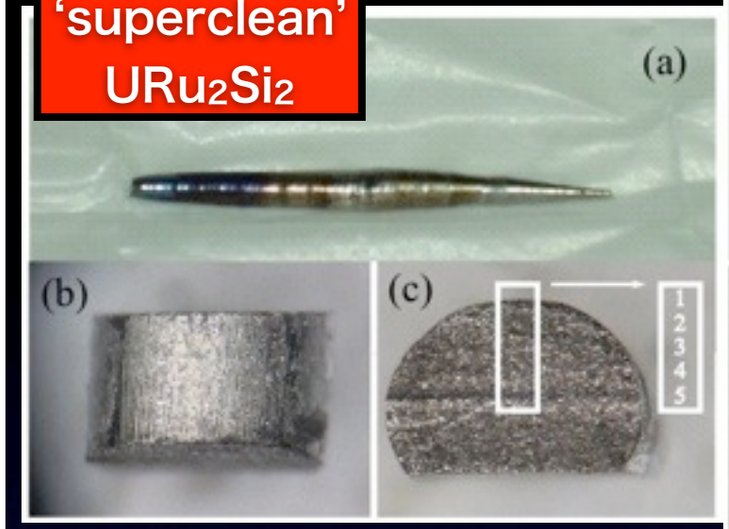


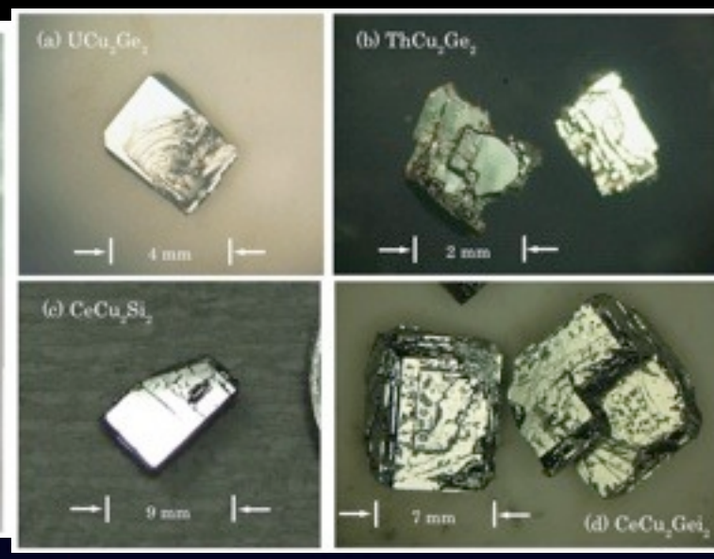
$PuIn_3$

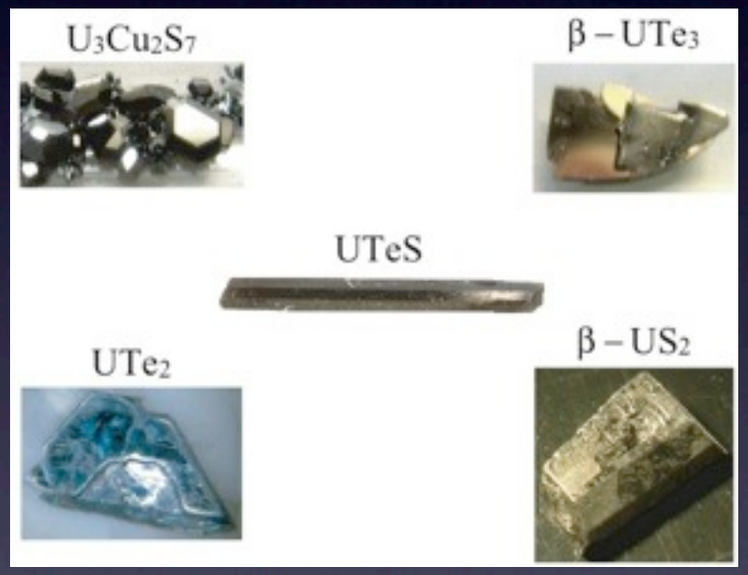


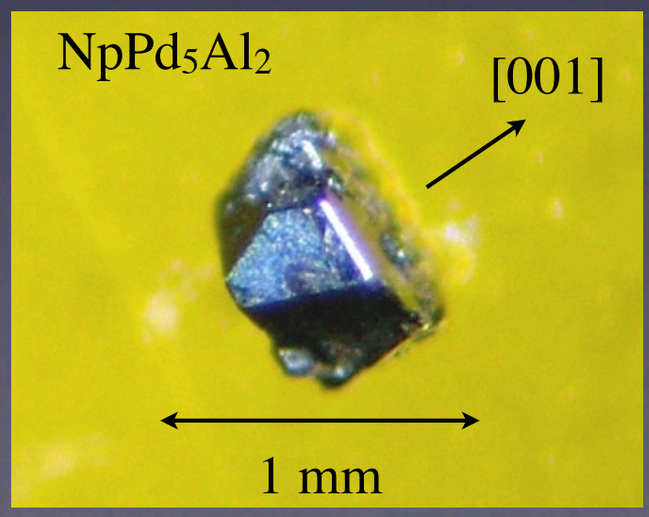
$NpFe_4P_{12}$

‘superclean’
URu_2Si_2





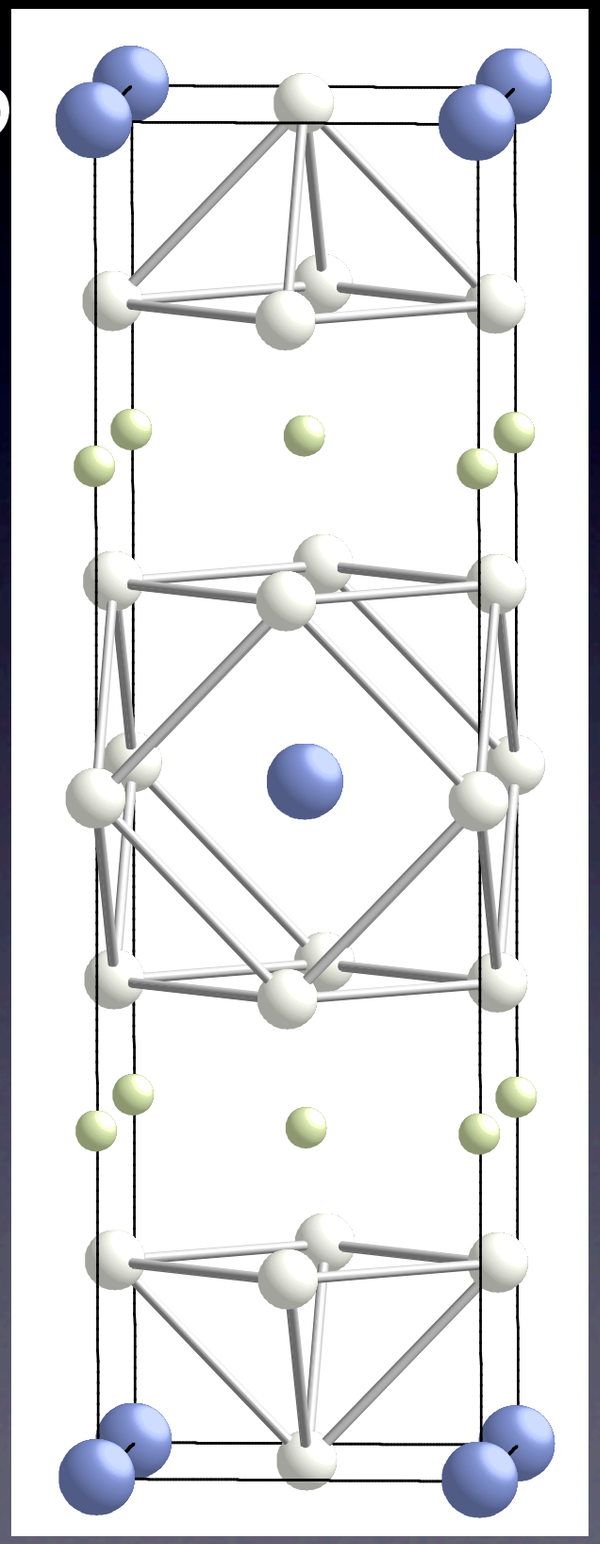


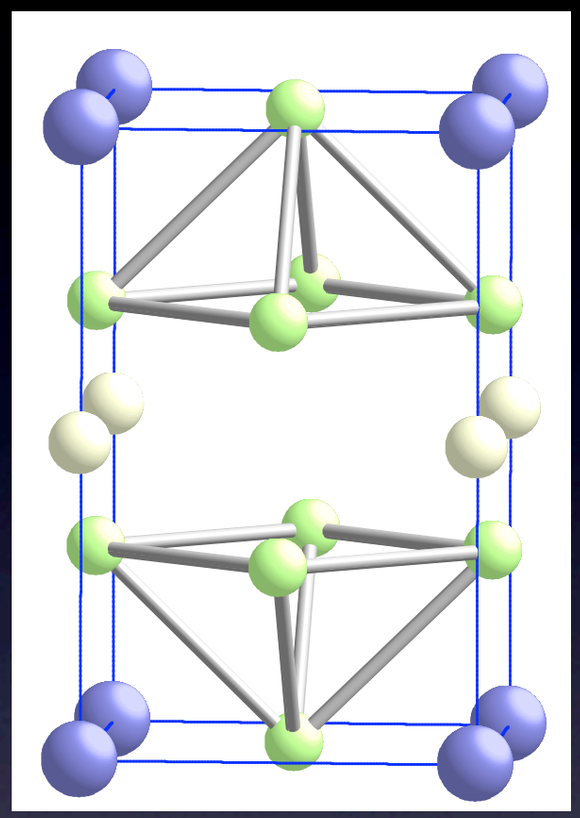


$NpPd_5Al_2$

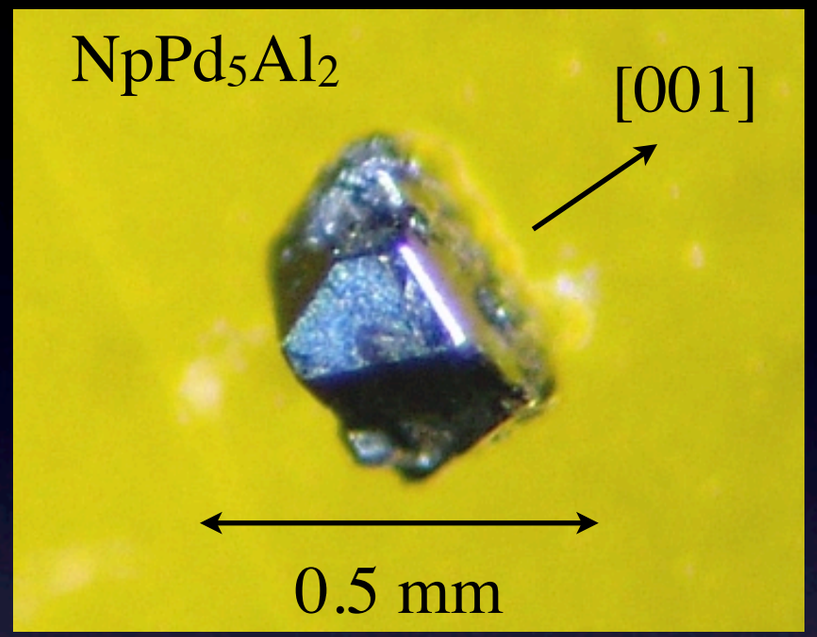
$NpPd_5Al_2$ $PuRhGa_5$

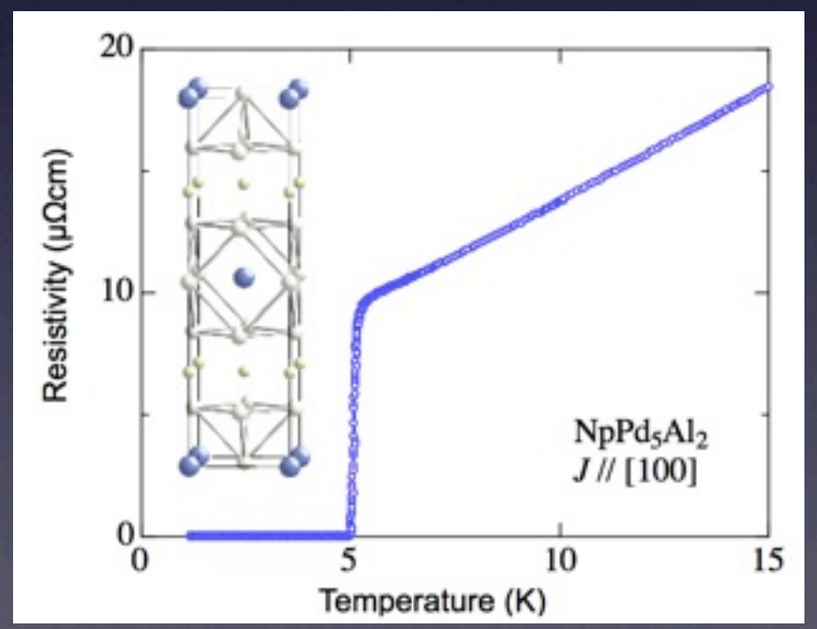
Np Pd Al





Pu Ga Rh

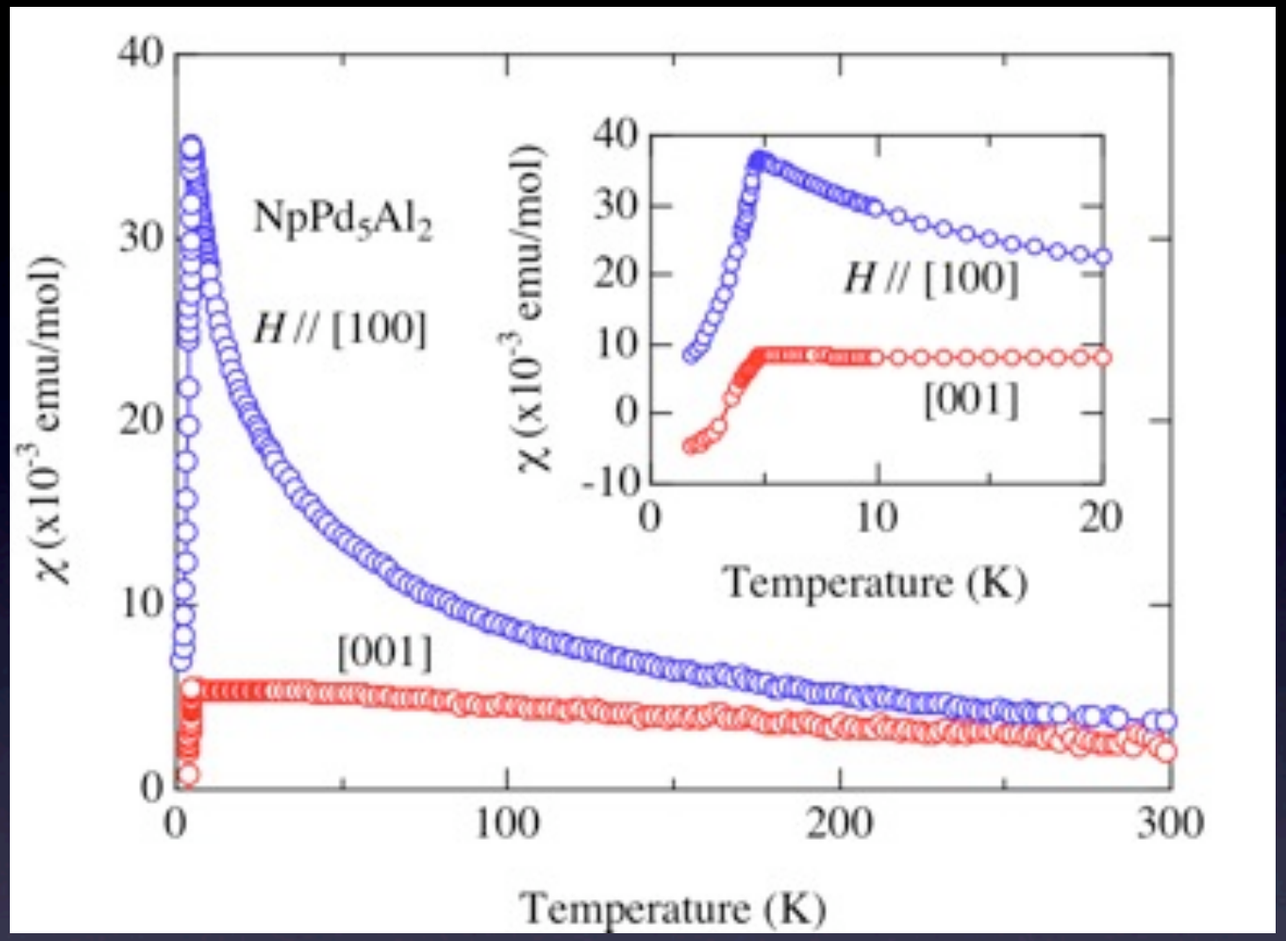




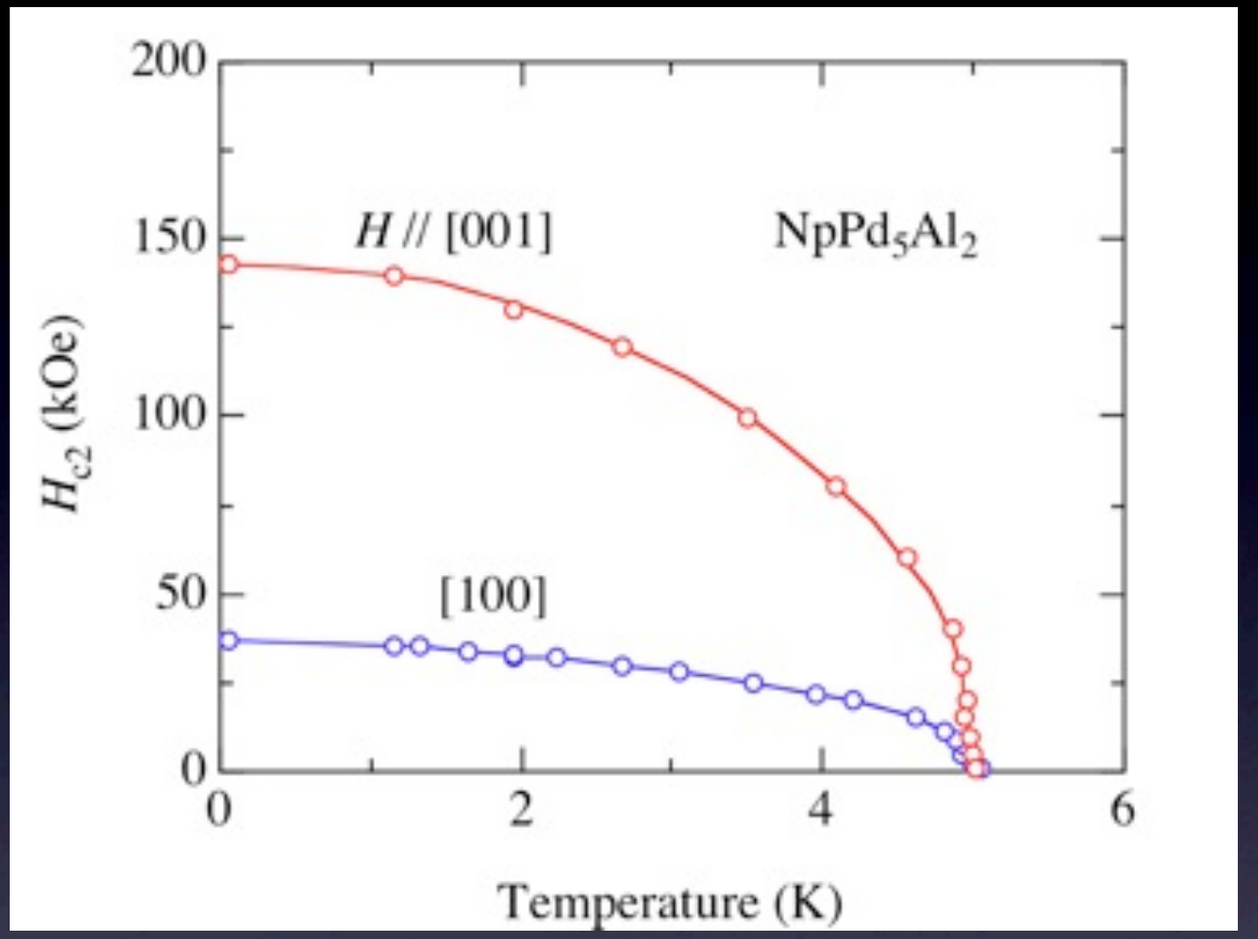
- first Np-HFSC
- structural similarity with Pu-115 structure

D. Aoki et al., J. Phys. Soc. Jpn. 76 (2007) 063701.

Anisotropy in Magnetism and Superconductivity



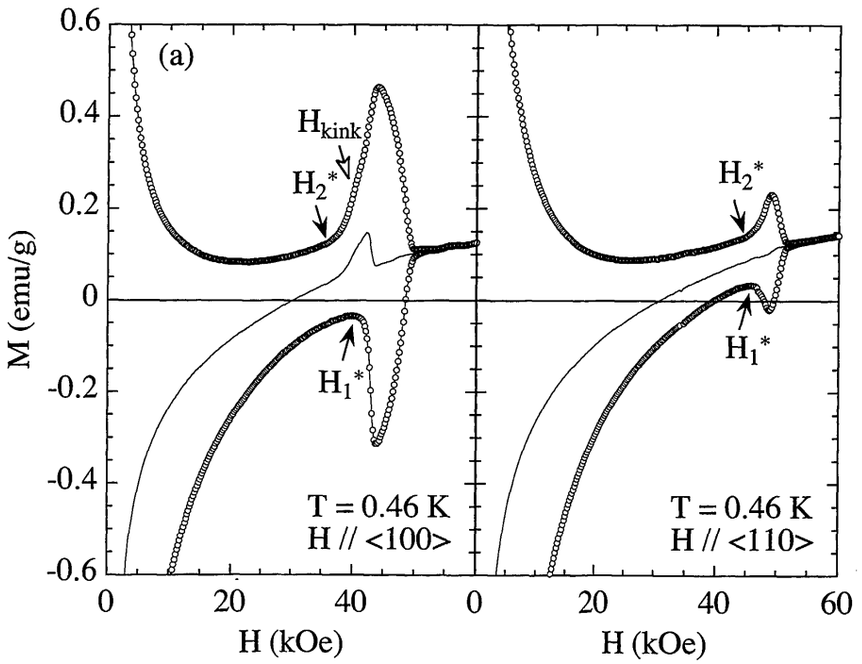
XY-type anisotropy
3.22 μ_B/Np for [100]
(3.62 μ_B/Np for Np^{4+}, 2.68 for Np^{3+})



H_{c2} suppressed for magnetic easy axis
Pauli-limiting --- singlet pairing suggested

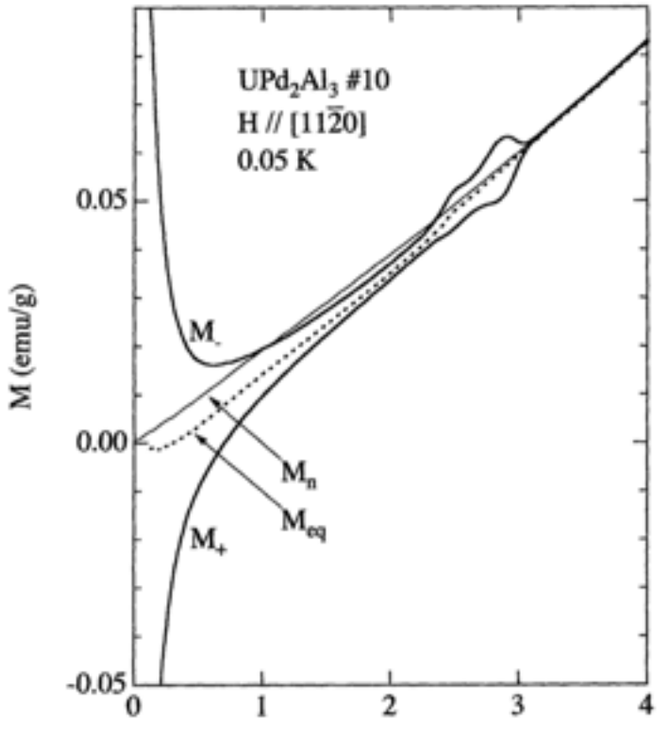
HFSC without 1st order transition at H_{c2} : $CeRu_2$, UPd_2Al_3, UPt_3, UBe_{13}

$CeRu_2$: s-wave



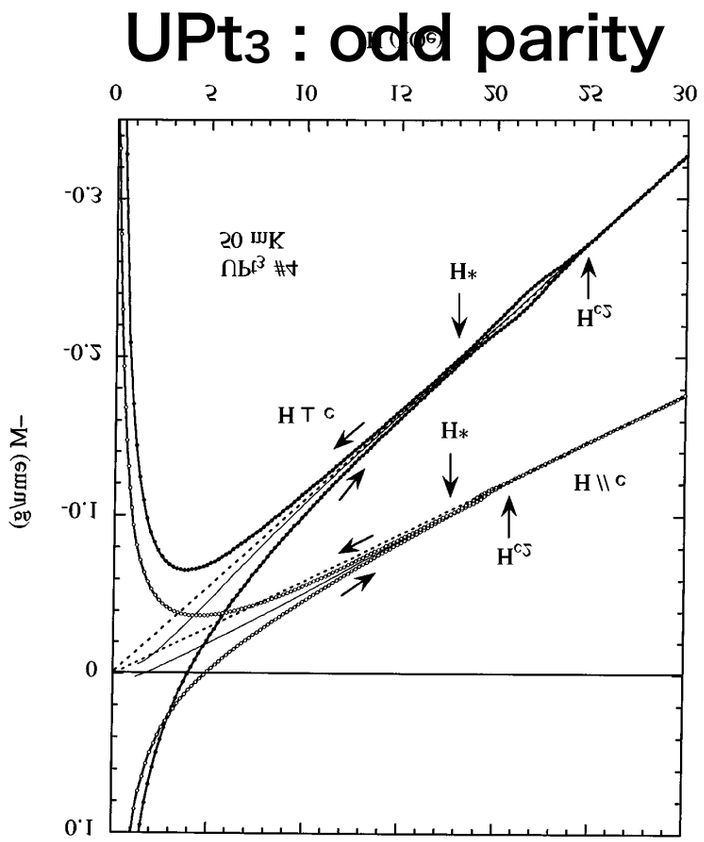
K. Tenya et al., J. Phys. Soc. Jpn. **68** (1999) 224.

UPd_2Al_3 : d-wave

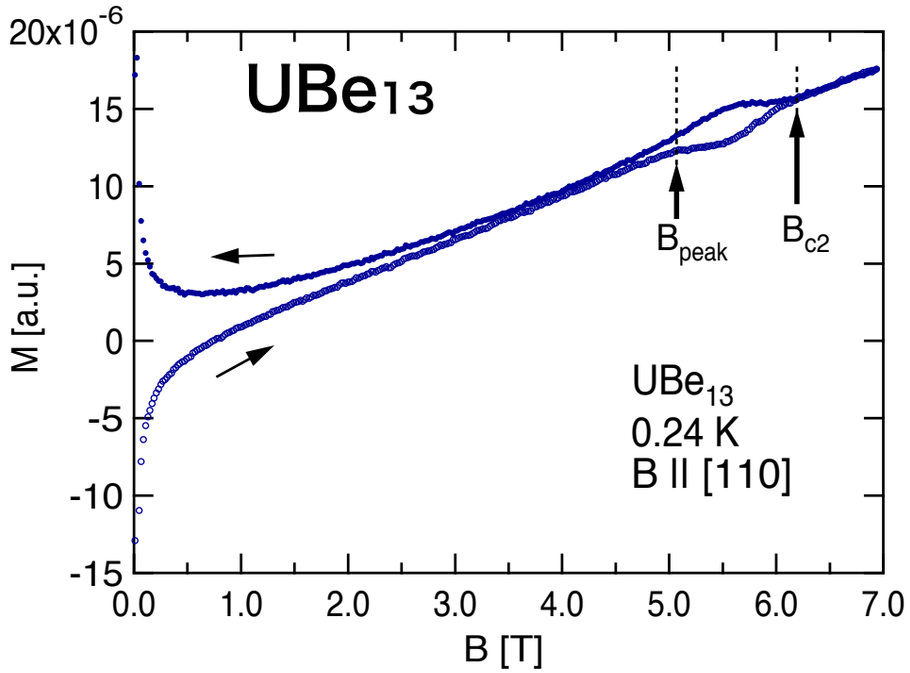


Y. Haga et al., J. Phys. Soc. Jpn. **65** (1996) 3646.

UPt_3 : odd parity

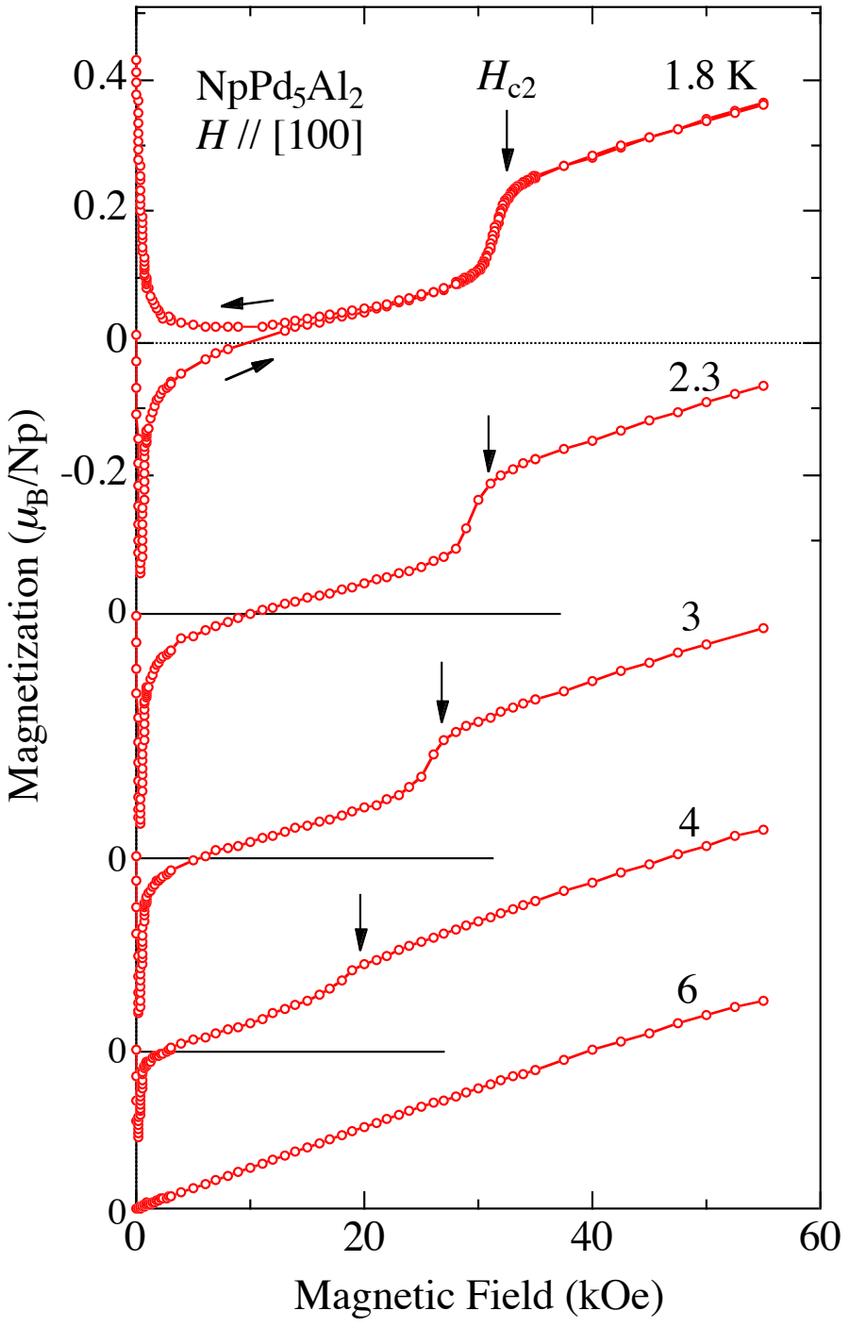


K. Tenya et al., **PRL** 77, 3193 (1996)

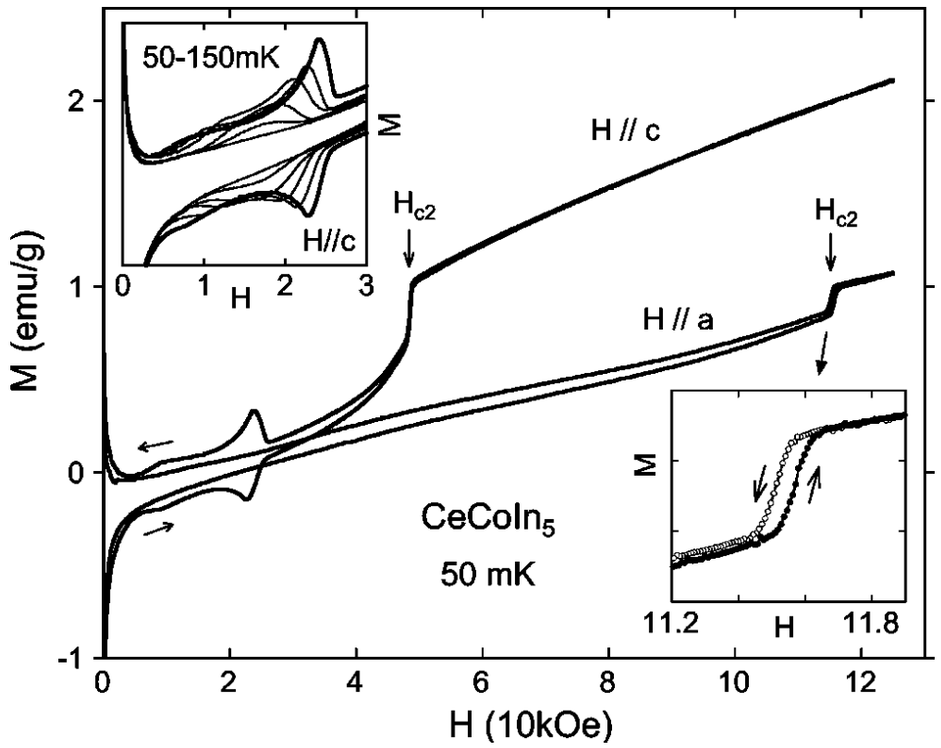


Y. Shimizu et al., J. Phys. Conf. Ser. **273** (2011) 012084

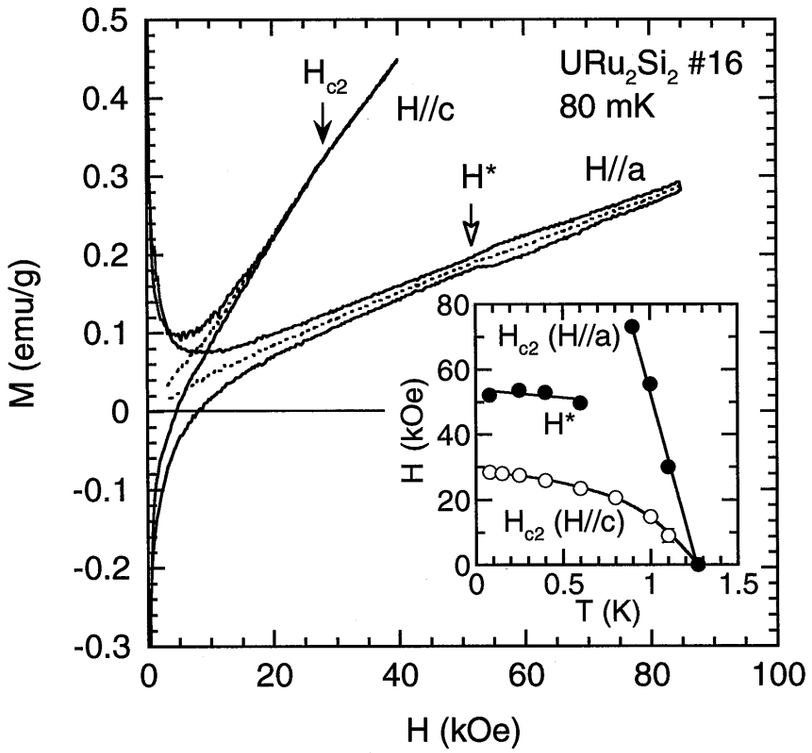
HFSC with 1st order transition at H_{c2} : $NpPd_5Al_2$, $CeCoIn_5$, URu_2Si_2



D. Aoki et al., J. Phys. Soc. Jpn. **76** (2007) 063701.

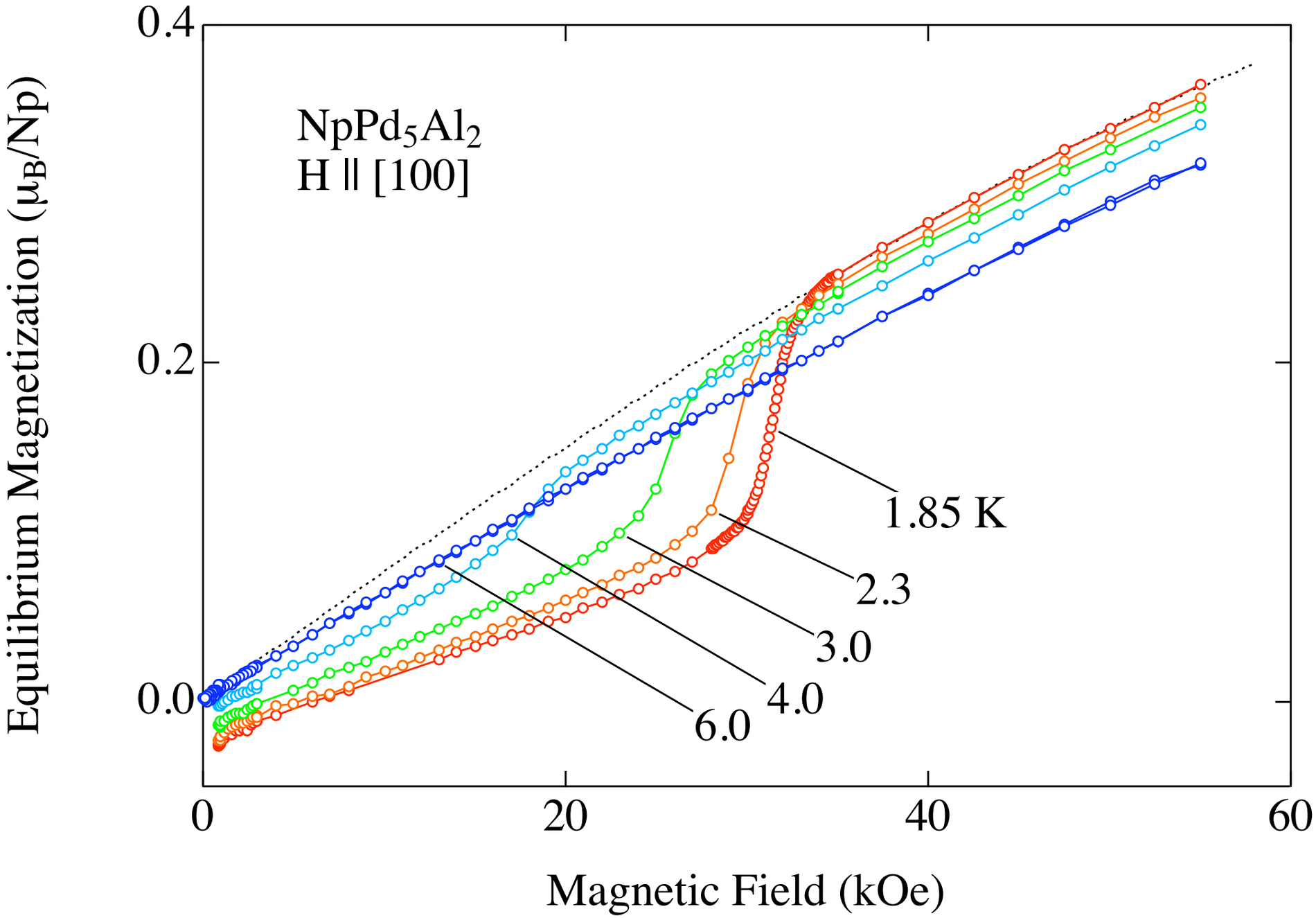


T. Tayama et al., RPB **65**, 180504R (2002)



K. Tenya et al., Physica B **281&282** (2000) 991

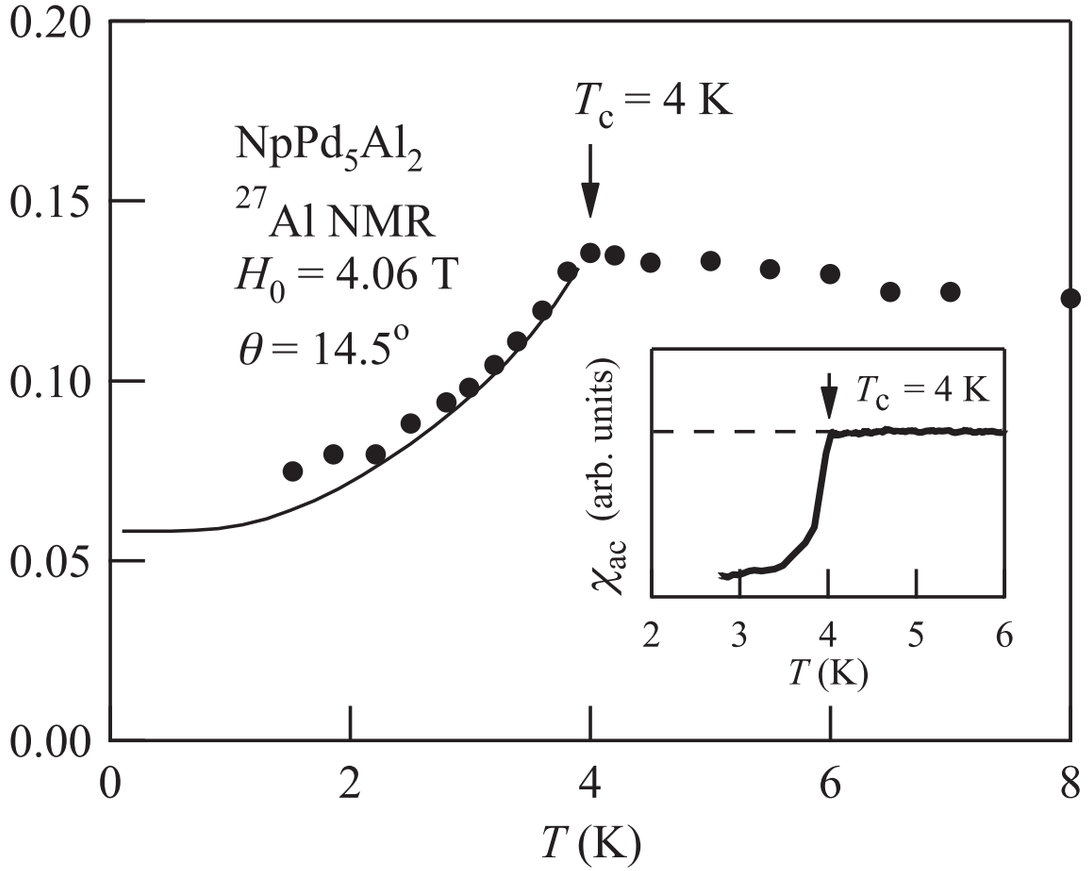
Equilibrium magnetization :1st order transition at H_{c2}



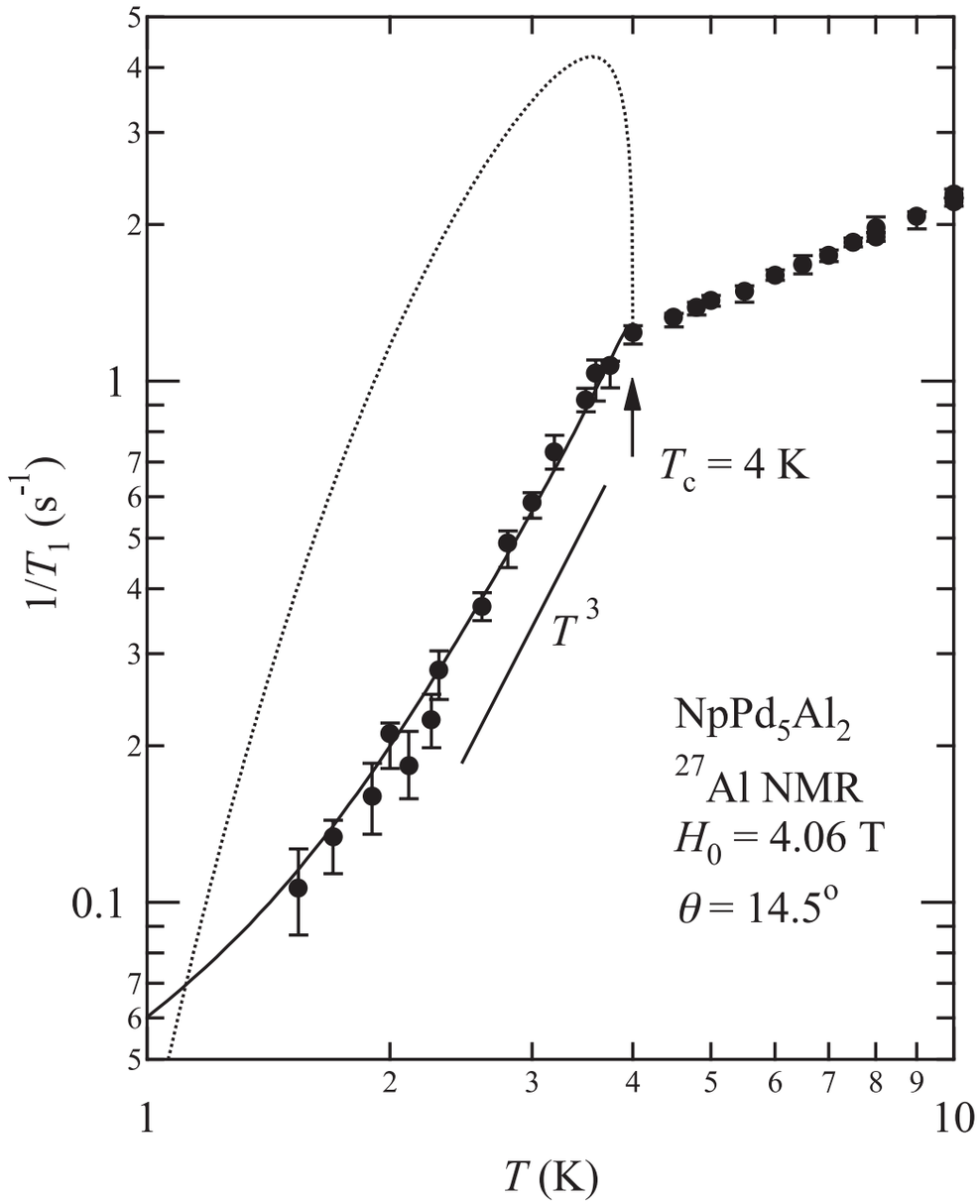
$M_{eq} = (M_+ + M_-) / 2$

$M_{norm} = \chi_1 H + \chi_3 H^3$

Al-NMR

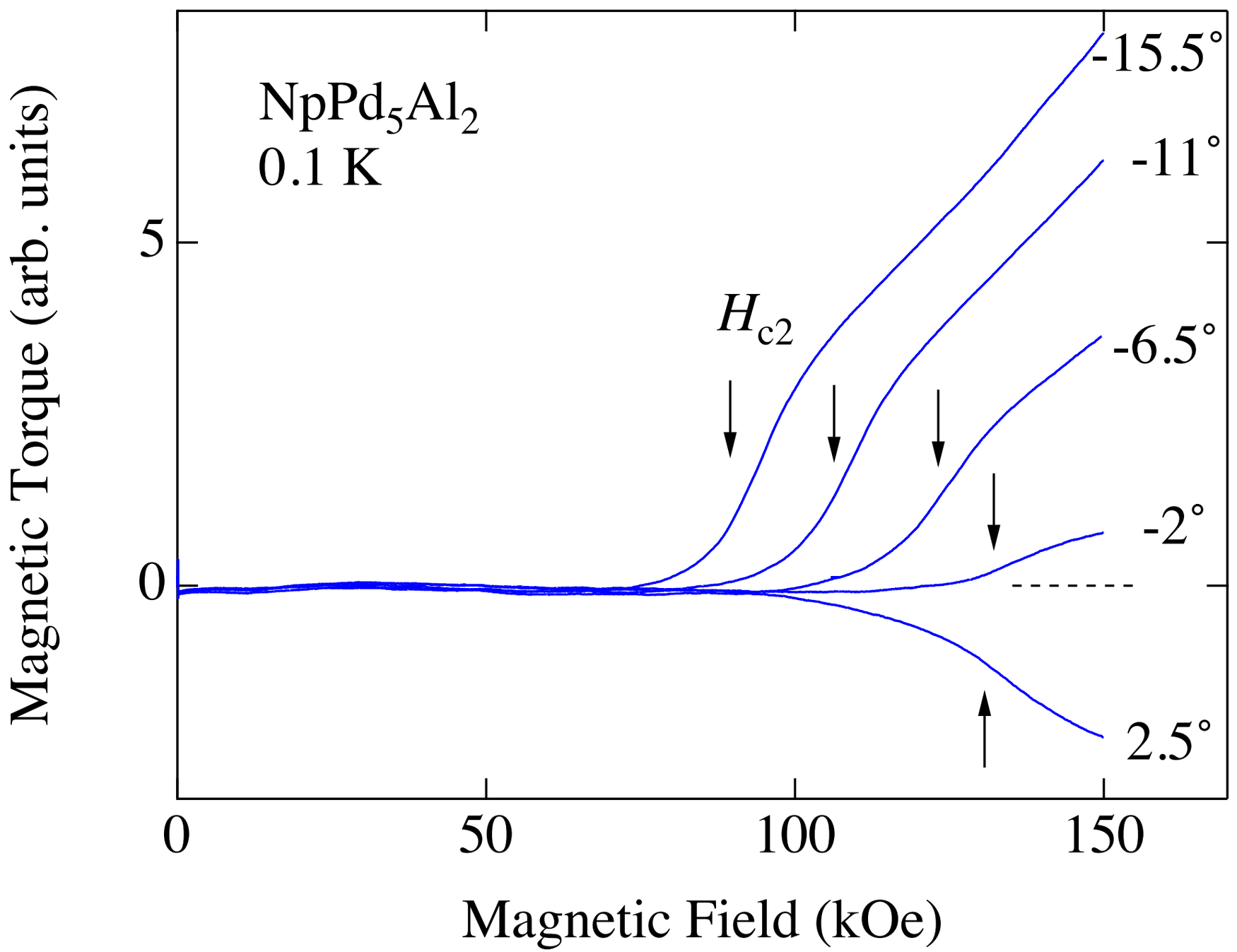


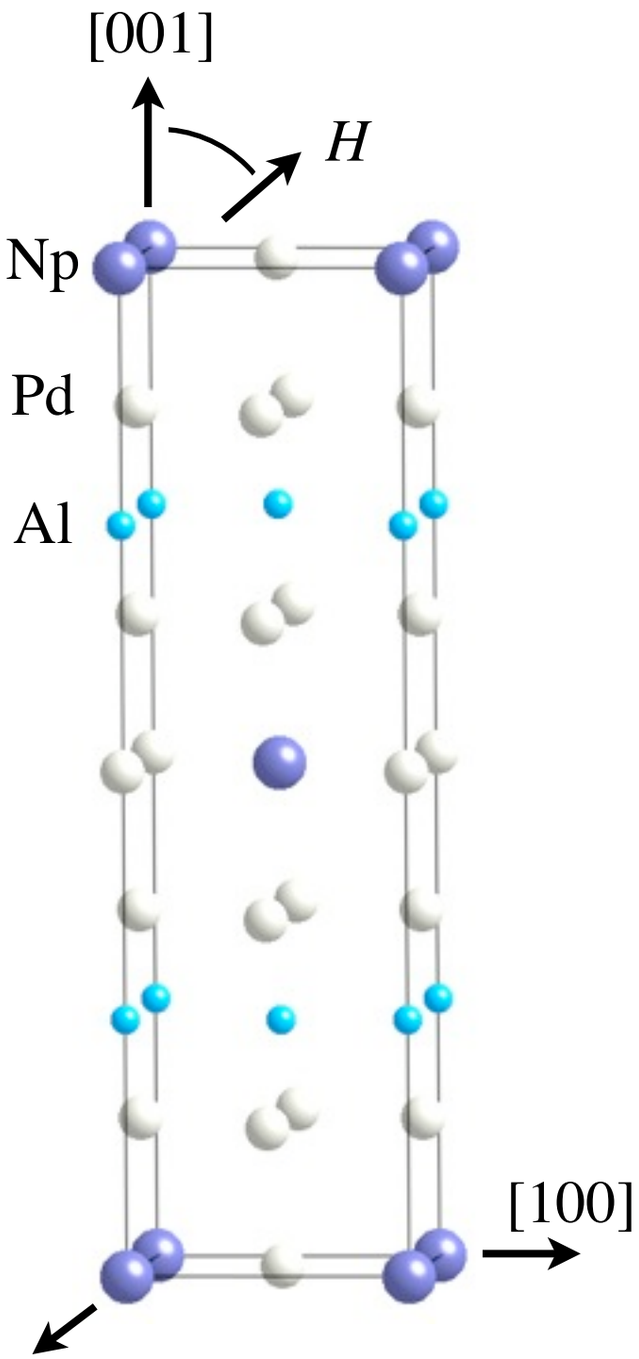
Singlet pairing & nodes on superconducting gap



H. Chudo et al., JPSJ (2008)

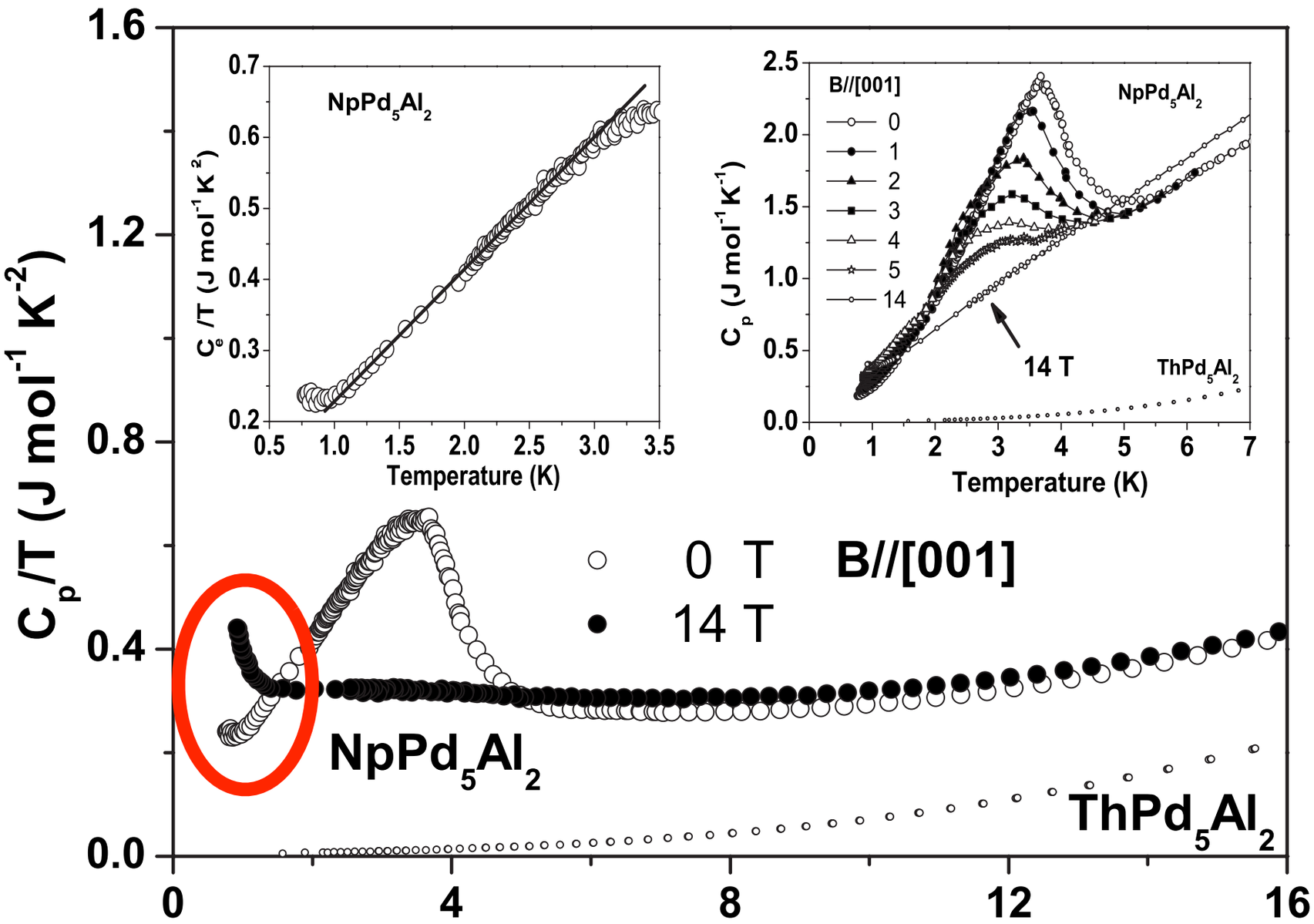
Magnetic Torque around [001] axis





$1/2\chi_a H_{c2}(a)^2 \approx 1/2\chi_c H_{c2}(c)^2$

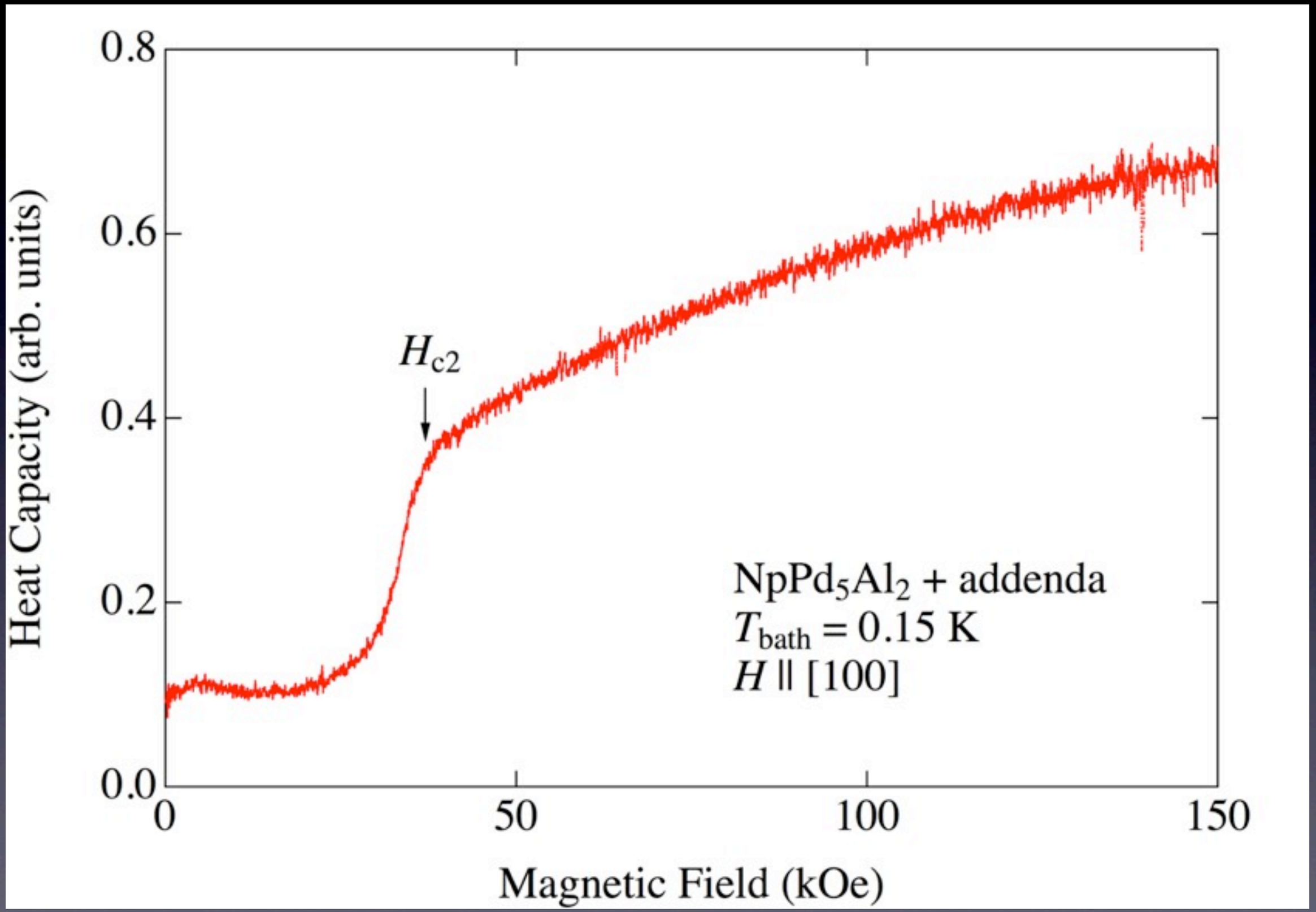
low-T $C_p(T)$ anomaly



^{237}Np nuclear Schottky ?
→200-300T internal field !

Griveau et al., PRB 77 (2008) 212502.

sudden recovery of Heavy Fermion above H_{c2}



$NpPd_5Al_2$: Heavy Fermion superconductor

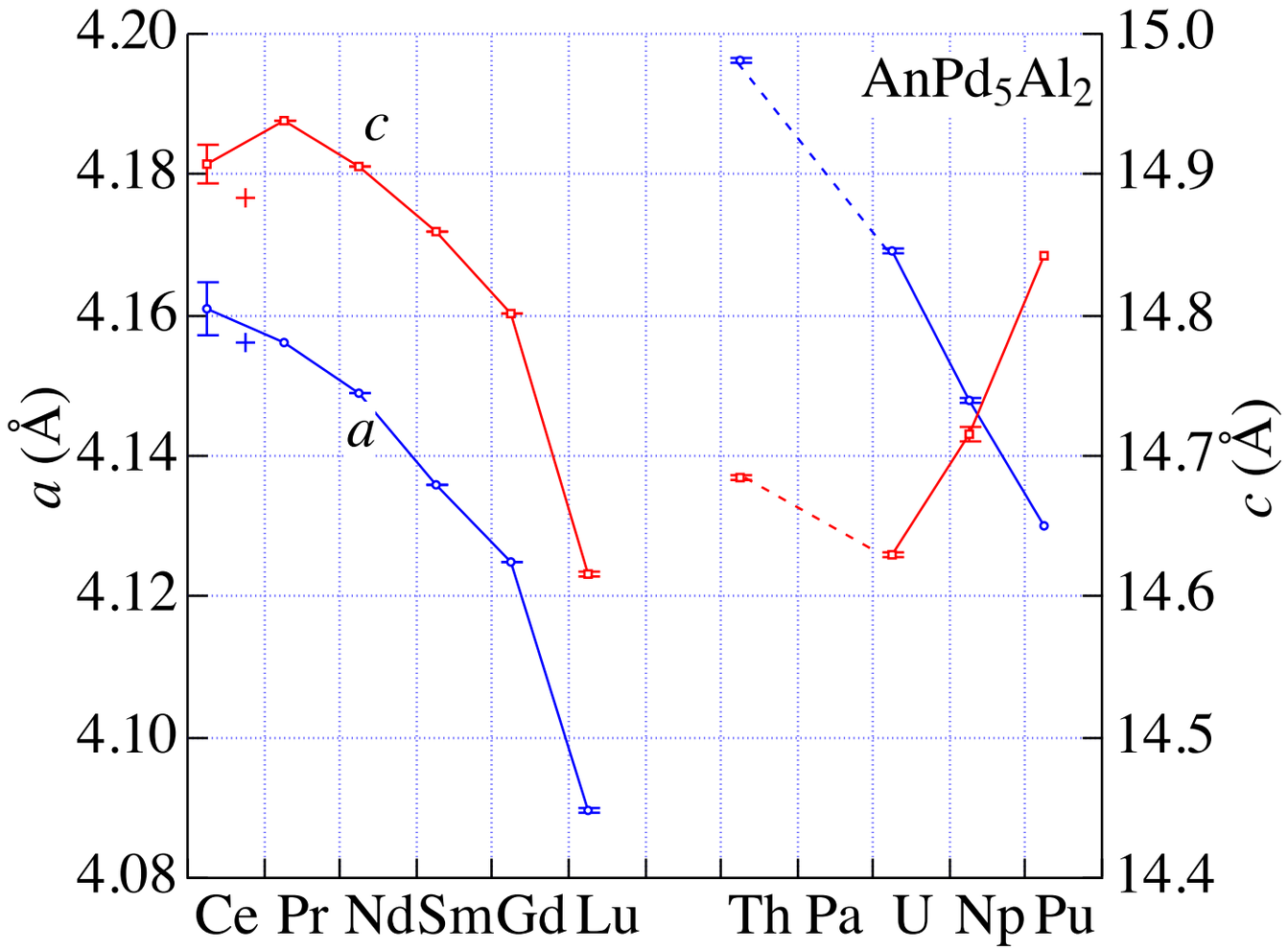
tetragonal $ZrNi_2Al_5$-type
T_c = 4.9 K
$C(T_c)/T$ = 200 mJ/K^{2}mol, $\Delta C/\gamma T_c$ = 2.23
Pauli-limited critical field
1st order transition at H_{c2}
Curie-Weiss law in normal state Np^{3+} or Np^{4+}
- vanishes in the superconducting state ...
- huge heavy fermion spin susceptibility (> 80 % of bulk χ)

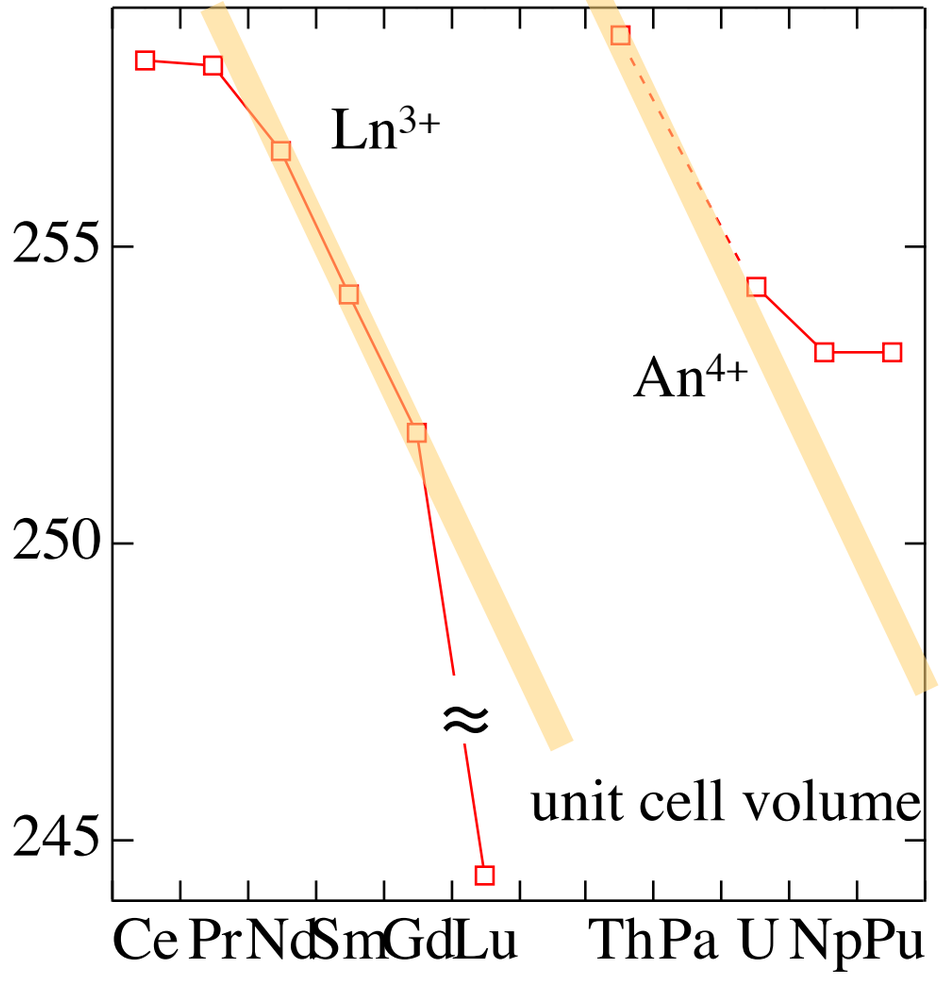
How about other heavy Fermion superconductors

- UPd_2Al_3 ～ 10 % of bulk χ contribute to SC : NMR, μSR
- $CeCoIn_5$ ～ 60 %
- UPt_3 small ? (odd parity)
- URu_2Si_2 small ?
- UBe_{13} small ?
- $CeCu_2Si_2$ ～ 100 % ? from NMR

- Pu superconductors ?
- High-T_c cuprates ?
- pnictides ?
- related to spin-resonance ?

unit cell volume : localized ? itinerant ?



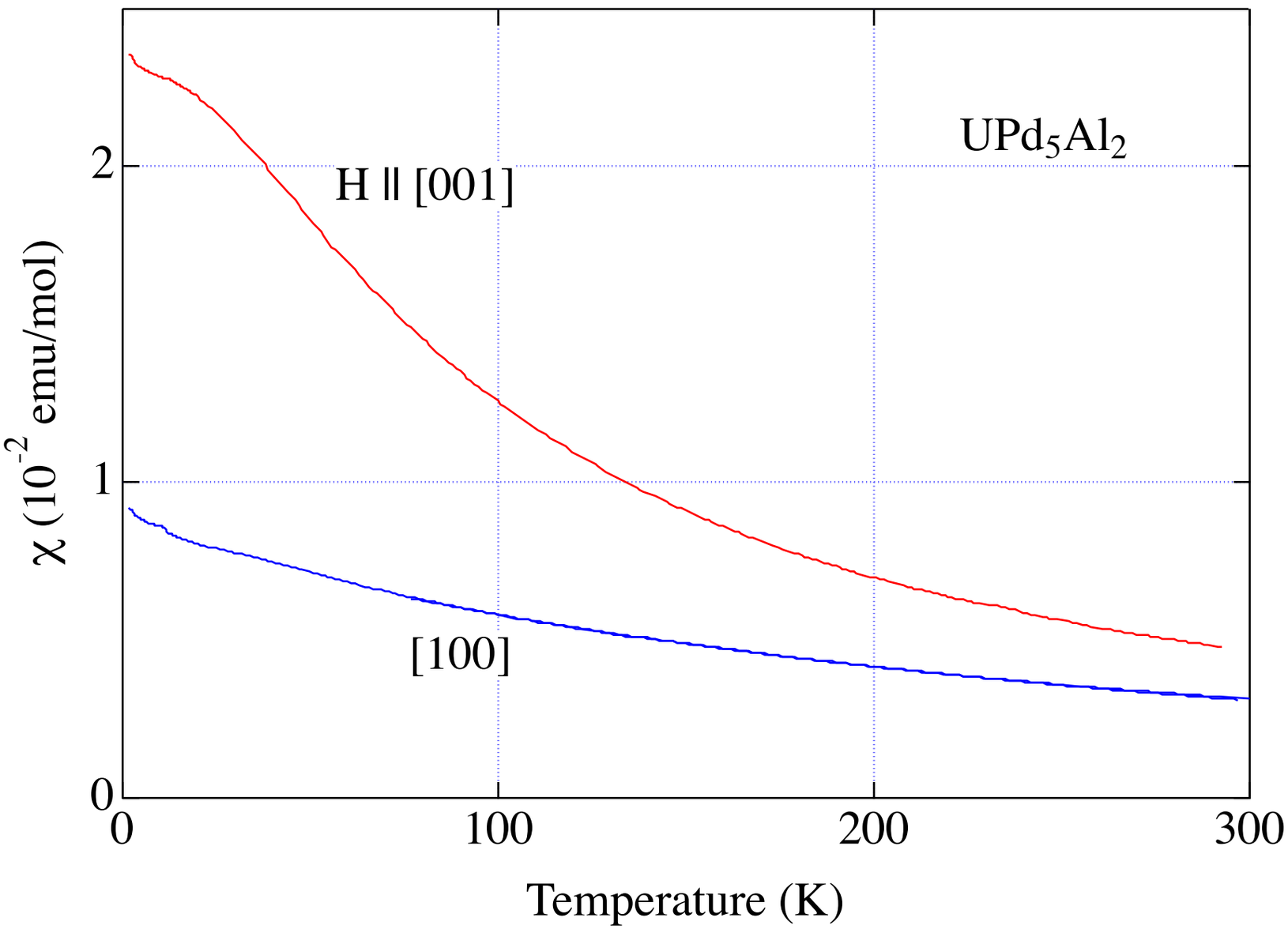


LnPd_5Al_2 follow the ***Ln*$^{3+}$** line except for Ce

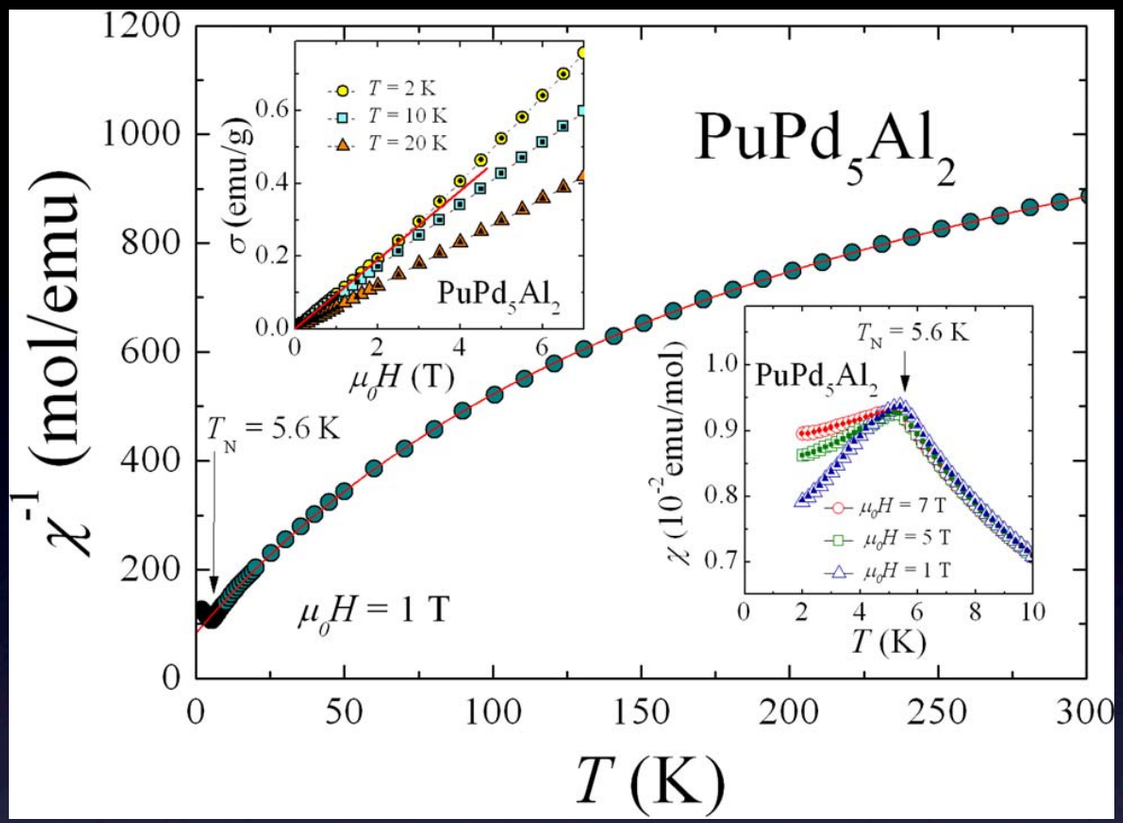
AnPd_5Al_2 : ***An*$^{4+}$** for Th, U. Np-Pu- approach to trivalent : consistent with $\chi(T)$

Y. Haga et al. (2008), J-C. Griveau et al., (2008), R.A. Ribeiro et al., (2009)

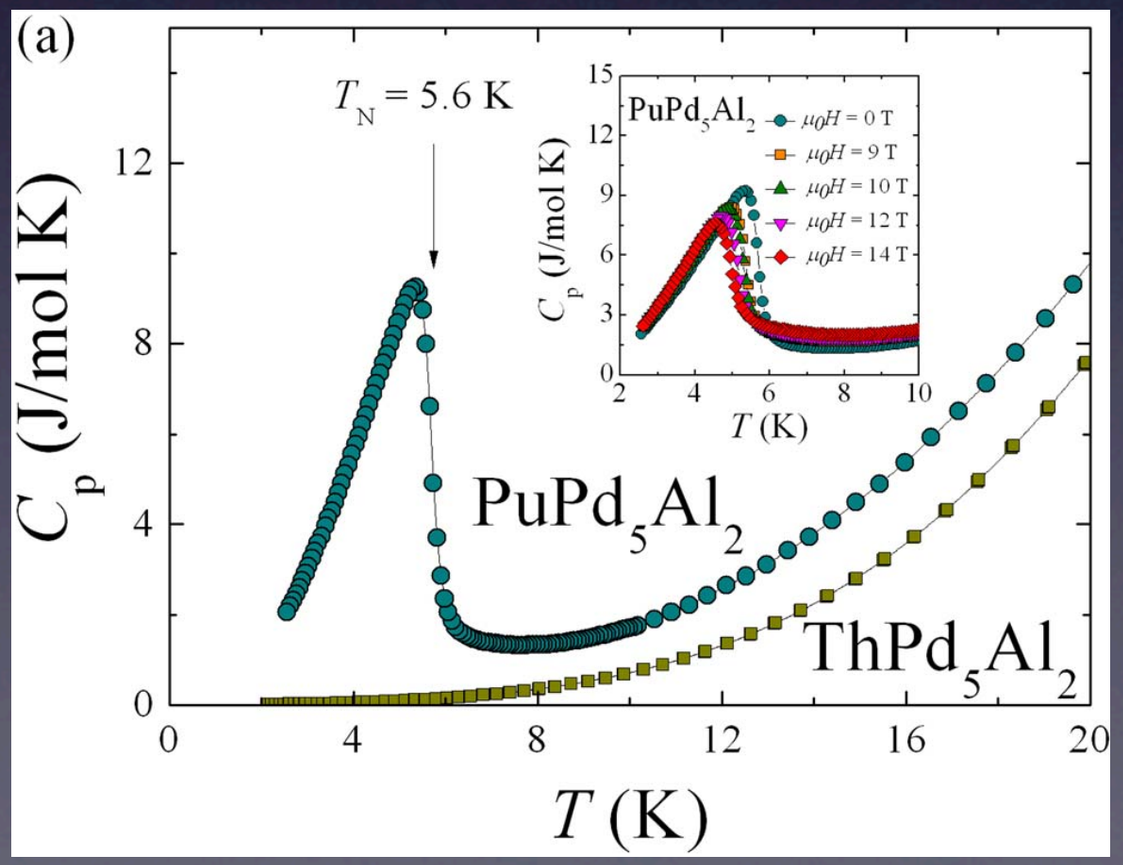
UPd_5Al_2 : Uniaxial magnetic anisotropy $NpPd_5Al_2$: XY-type C-W law above 50 K, CEF-singlet ? ($5f^2$) as in $PrPd_5Al_2$ ($4f^2$)

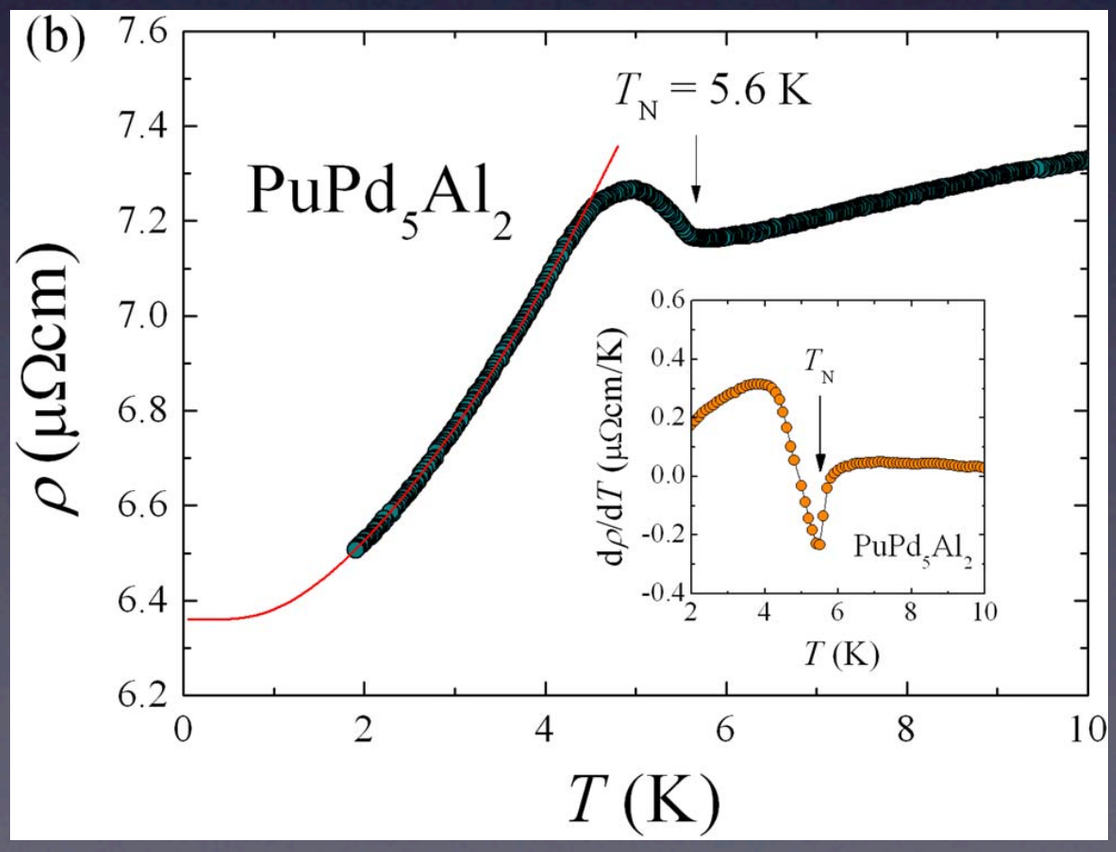


$PuPd_5Al_2$: Antiferromagnetic at 5 K Gofryk (2008)



consistent with Pu^{3+}





Summary

$NpPd_5Al_2$: first-order transition at H_{c2}

magnetic susceptibility and electronic specific heat suppressed below H_{c2}

large electronic entropy remains at low temperatures. ^{237}Np nuclear contribution ?

UPd_5Al_2 : CEF-singlet ground state : U^{4+} suggested

magnetic anisotropy in $AnPd_5Al_2$

Ce, Pr, U - uniaxial

Np - XY type

Ce under pressure ?? Pu ??