

Heavy fermions: Interplay between Kondo entanglement, quantum criticality and unconventional superconductivity

F. Steglich

MPI for Chemical Physics of Solids, 01187 Dresden, Germany

Collaboration

J. Arndt, O. Stockert, S. Wirth (MPI CPS)
G. M. Pang, M. Smidman, H. Q. Yuan (CCM, ZJU)
E. Schuberth, M. Tippmann, L. Steinke (WMI)
E. M. Nica, R. Yu, Q. Si (RCQM, Rice U.)

Outline

Kondo effect

Heavy-fermion (HF) superconductivity (SC)

Quantum critical point (QCP) in HF metals

SC near an itinerant AF (SDW) QCP in CeCu_2Si_2

SC due to nuclear AF order in YbRh_2Si_2

INTUITED

SPEED

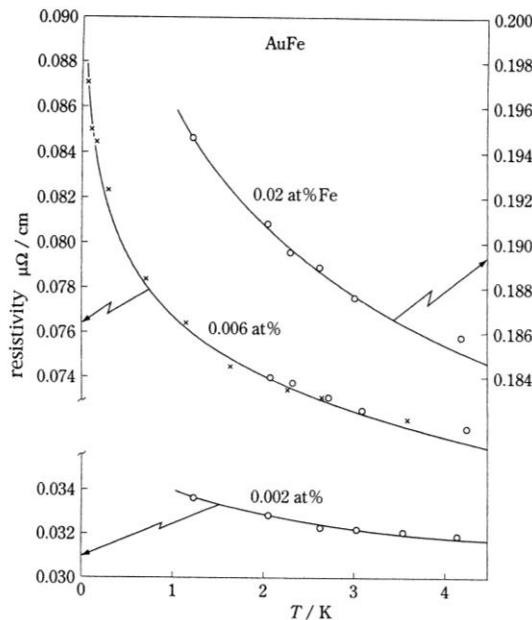


~ 1930: $\rho(T)$ anomaly in pure Cu [Meissner & Voigt (1930),
van den Berg et al. (1934)]

~ 1950: $\rho(T)$ anomaly due to transition-metal impurities

AuFe

Mac Donald et al. (1962)



J. Kondo (1964)



$$H_{\text{int}} = 2 J_K \mathbf{S} \cdot \mathbf{s}$$

$$J_K > 0$$

$$T_K \sim T_F \exp(-1/N_F J_K)$$

K. Wilson (1975)

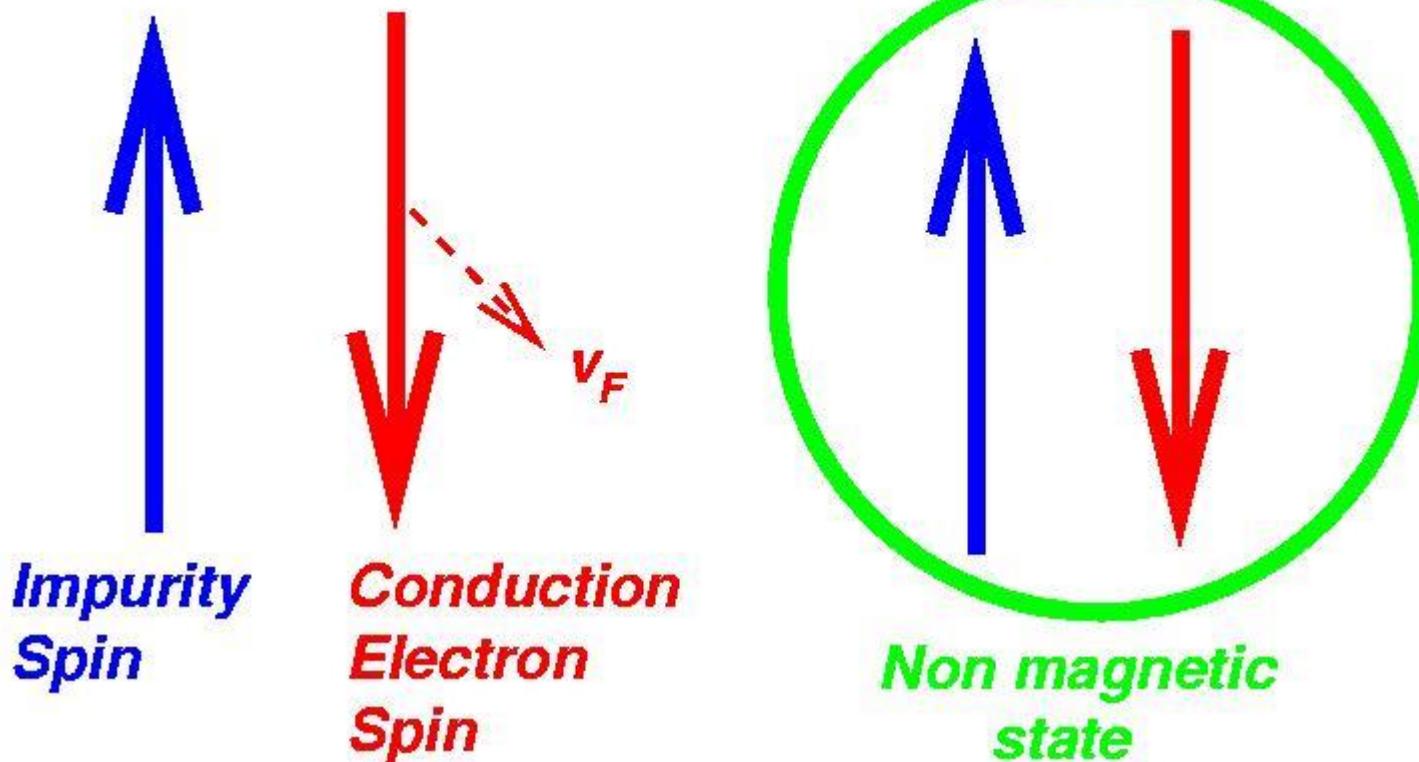
$$- dJ_K/dl = \beta(J_K) = J_K^2$$

$\rho(T)$ minimum

asymptotic freedom

Kondo effect

High T – weak coupling Low T – strong coupling



magnetic susceptibility

Curie Weiss law: $\chi \sim (T + \theta)^{-1}$, $\theta = f(T_K) > 0$

effective moment $\mu_{\text{eff}}(T)$: $= \chi \cdot T \rightarrow 0 (T \rightarrow 0)$

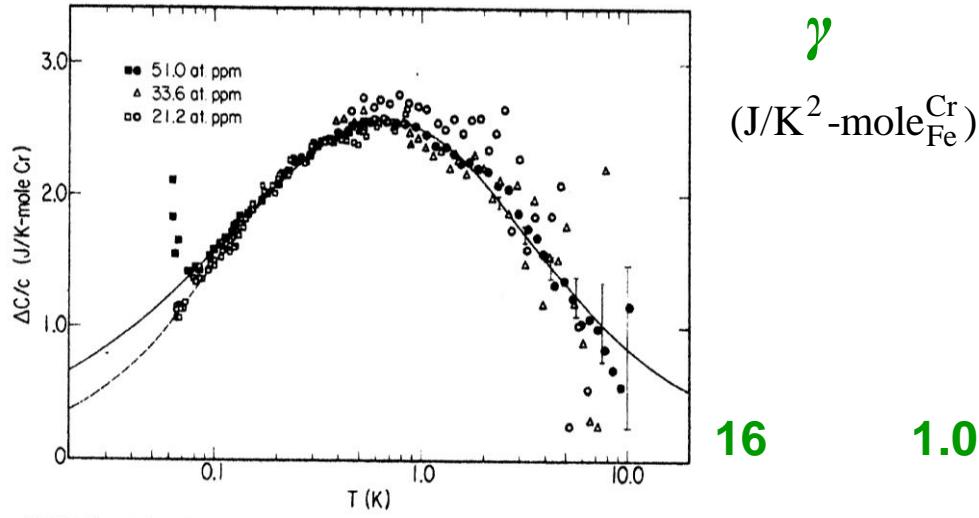
specific heat

Triplet & Philipps (1971): $\text{Cu}_{1-x}\text{M}_x$ (M : Cr, Fe)

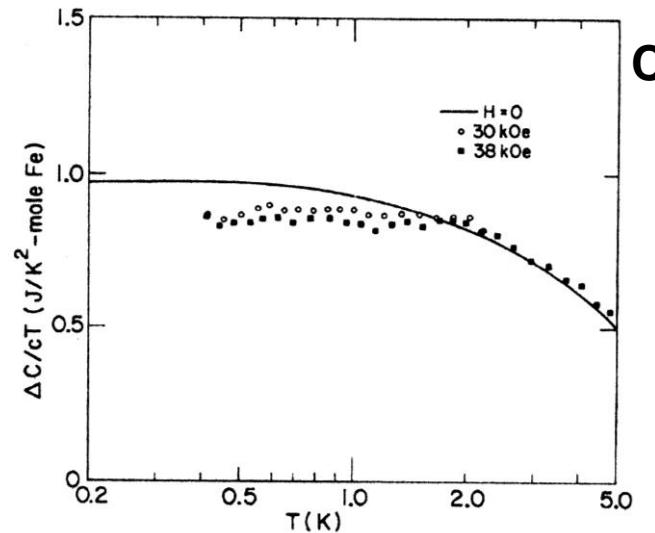
incremental specific heat: $\Delta C = C - C_{\text{Cu}}$

per mole M : $\Delta C/x = \gamma T$ ($T \ll T_K$): „Kondo resonance“

CuCr



CuFe



P. Nozières (1974) $T \ll T_K$: local Fermi liquid

outline

Kondo effect

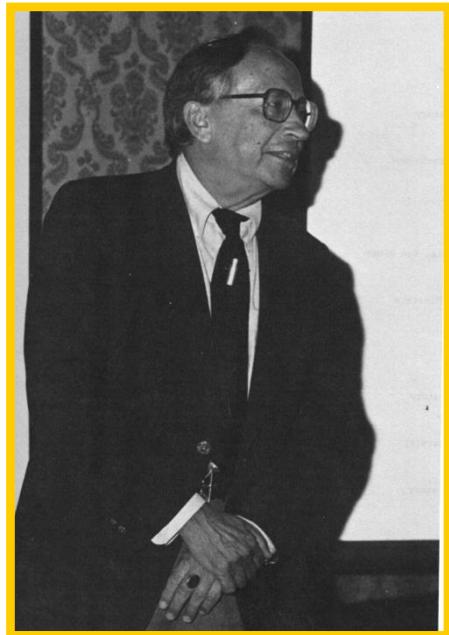
Heavy-fermion (HF) superconductivity (SC)

Quantum critical point (QCP) in HF metals

SC near an itinerant AF (SDW) QCP in CeCu_2Si_2

SC due to nuclear AF order in YbRh_2Si_2

T_c of $\text{La}_{0.99}\text{RE}_{0.01}$



VOLUME 1, NUMBER 3

PHYSICAL REVIEW LETTERS

AUGUST 1, 1958

SPIN EXCHANGE IN SUPERCONDUCTORS

B. T. Matthias, H. Suhl, and E. Corenzwit

Bell Telephone Laboratories,

Murray Hill, New Jersey

(Received July 15, 1958)

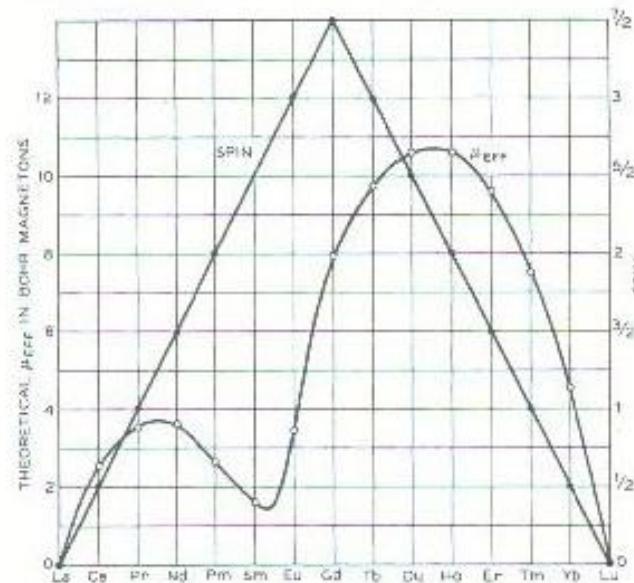


FIG. 1. Effective magnetic moments and spins of the rare earth elements (see reference 2).

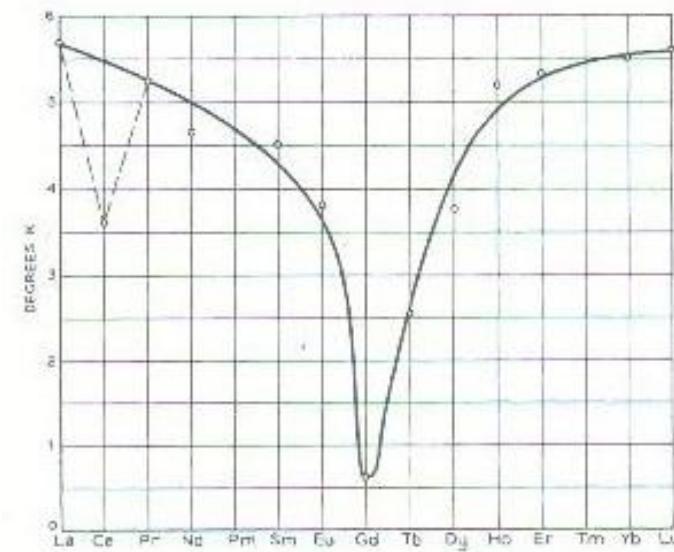


FIG. 2. Superconducting transition temperatures of 1 at % rare earth solid solutions in lanthanum.

4f-Virtual-Bound-State Formation in CeAl₃ at Low Temperatures

K. Andres and J. E. Graebner

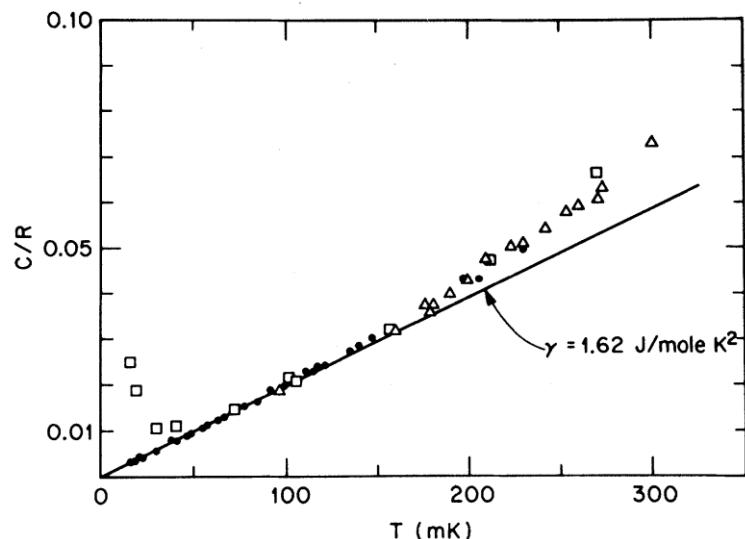
Bell Laboratories, Murray Hill, New Jersey 07974

and

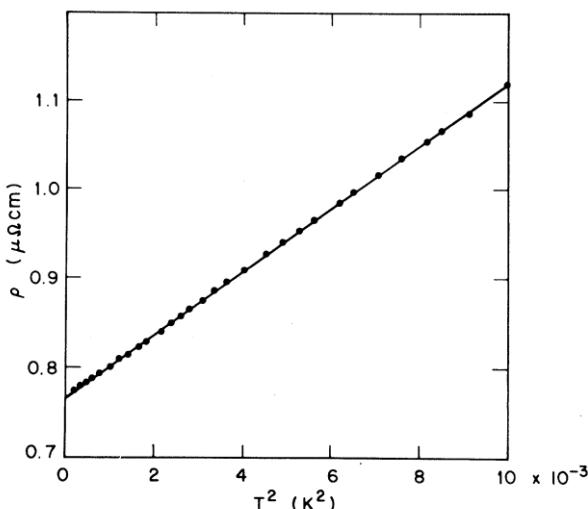
H. R. Ott

*Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule,**Hönggerberg, Zürich, Switzerland*

(Received 25 August 1975)

FIG. 1. Specific heat of CeAl₃ at very low temperatures in zero field (●, △) and in 10 kOe (□).

$$\gamma = 1.62 \text{ J/K}^2\text{mole}$$

FIG. 3. Electrical resistivity of CeAl₃ below 100 mK, plotted against T^2 .

$$A = 35 \mu\Omega\text{cm}/\text{K}^2$$

Superconductivity in CeCu_2Si_2

VOLUME 43, NUMBER 25

PHYSICAL REVIEW LETTERS

17 DECEMBER 1979

Superconductivity in the Presence of Strong Pauli Paramagnetism: CeCu_2Si_2

F. Steglich

Institut für Festkörperphysik, Technische Hochschule Darmstadt, D-6100 Darmstadt, West Germany

and

J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, and W. Franz

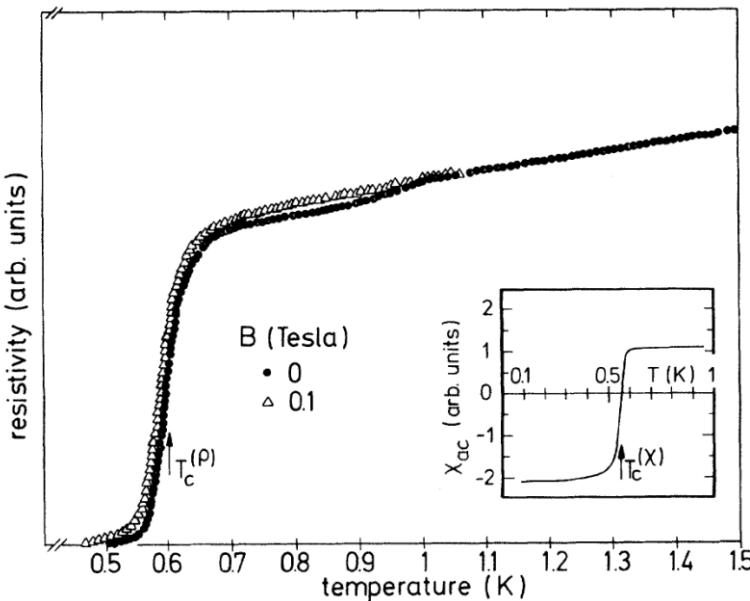
II. Physikalisches Institut, Universität zu Köln, D-5000 Köln 41, West Germany

and

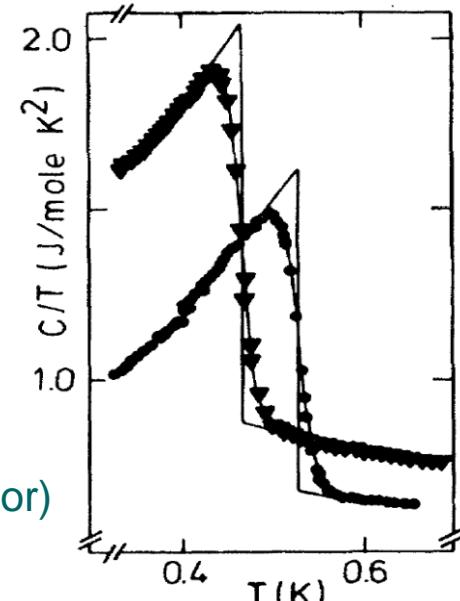
H. Schäfer

Eduard-Zintl-Institut, Technische Hochschule Darmstadt, D-6100 Darmstadt, West Germany

(Received 10 August 1979; revised manuscript received 7 November 1979)



100 at% Ce^{3+} ions necessary
for superconductivity
(LaCu_2Si_2 is not a superconductor)



Superconductivity in CeCu₂Si₂

VOLUME 43, NUMBER 25

PHYSICAL REVIEW LETTERS

17 DECEMBER 1979

Superconductivity in the Presence of Strong Pauli Paramagnetism: CeCu₂Si₂

F. Steglich

Institut für Festkörperphysik, Technische Hochschule Darmstadt, D-6100 Darmstadt, West Germany

and

J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, and W. Franz

II. Physikalisches Institut, Universität zu Köln, D-5000 Köln 41, West Germany

and

H. Schäfer

Eduard-Zintl-Institut, Technische Hochschule Darmstadt, D-6100 Darmstadt, West Germany

(Received 10 August 1979; revised manuscript received 7 November 1979)

... Since the Debye temperature Θ is of order of 200 K, we find $T_c < T_F^* < \Theta$ with $T_c/T_F^* \approx T_F^*/\Theta \approx 0.05$. This suggests that CeCu₂Si₂

- (i) behaves as a „high – T_c superconductor“ and
- (ii) cannot be described by conventional theory of superconductivity which assumes a typical phonon frequency $k_B\Theta/h \ll k_B T_F^*/h$, the characteristic frequency of the fermions.

Heavy-fermion superconductivity in CeCu₂Si₂

VOLUME 52, NUMBER 6

PHYSICAL REVIEW LETTERS

6 FEBRUARY 1984

Superconductivity in CeCu₂Si₂ Single Crystals

W. Assmus and M. Herrmann

Physikalisches Institut, Universität Frankfurt, D-6000 Frankfurt am Main, West Germany

and

U. Rauchschwalbe, S. Riegel, W. Lieke, H. Spille, S. Horn, G. Weber, and F. Steglich

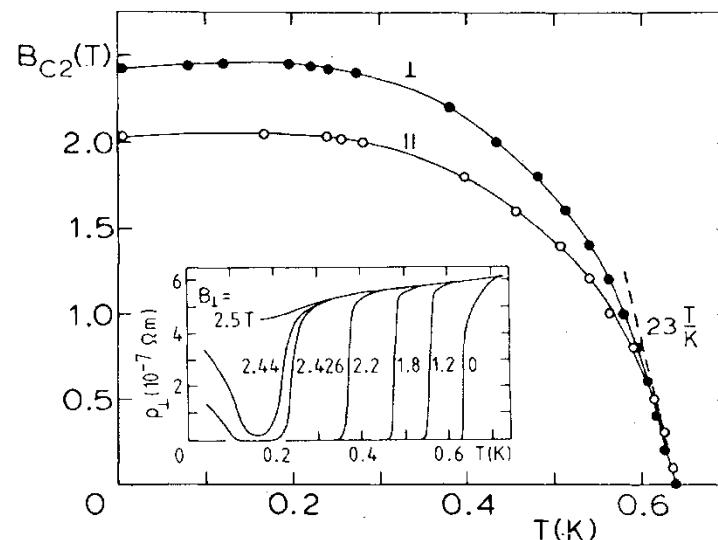
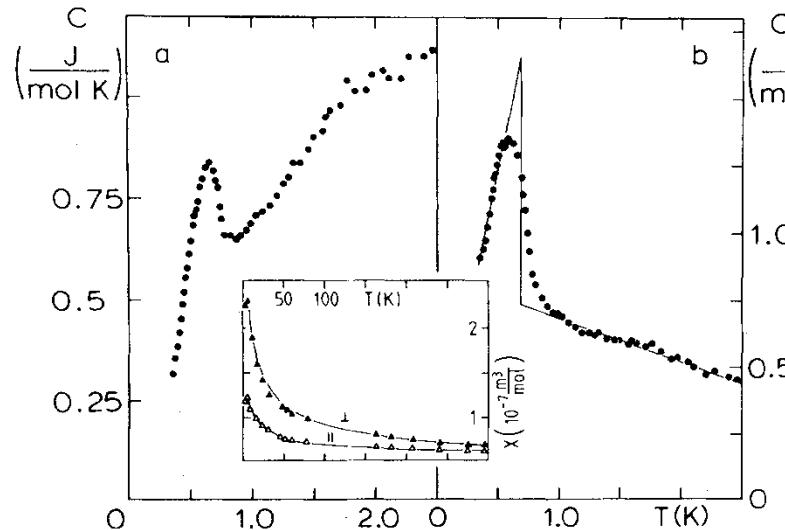
Institut für Festkörperphysik, Technische Hochschule Darmstadt, D-6100 Darmstadt, West Germany

and

G. Cordier

E. Zintl Institut, Technische Hochschule Darmstadt, D-6100 Darmstadt, West Germany

(Received 19 August 1983)



Heavy-fermion superconductivity in UBe₁₃

VOLUME 50, NUMBER 20

PHYSICAL REVIEW LETTERS

16 MAY 1983

UBe₁₃: An Unconventional Actinide Superconductor

H. R. Ott and H. Rudigier

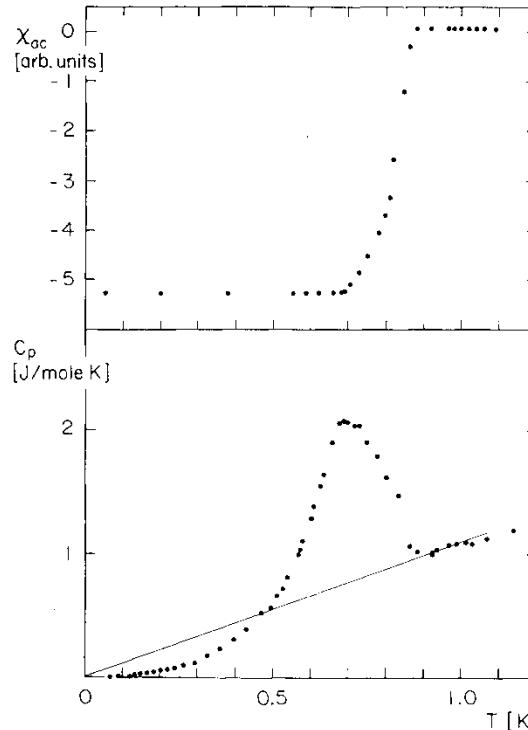
*Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule-Hönggerberg,
CH-8093 Zürich, Switzerland*

and

Z. Fisk and J. L. Smith

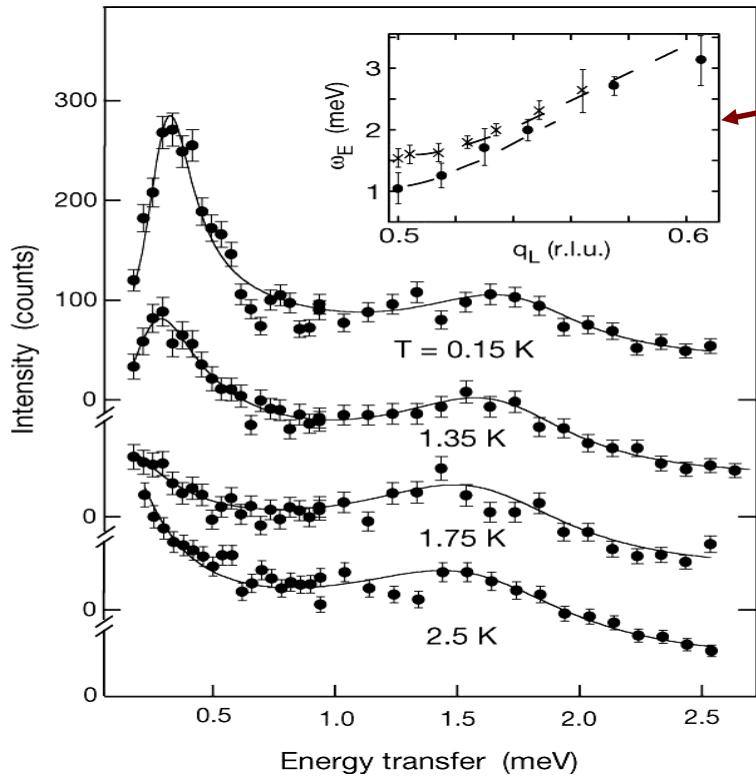
Los Alamos National Laboratory, Los Alamos, New Mexico 87545

(Received 14 March 1983)



UPd₂Al₃: Inelastic neutron scattering

[N. K. Sato et al., Nature 410, 340 (2001)]



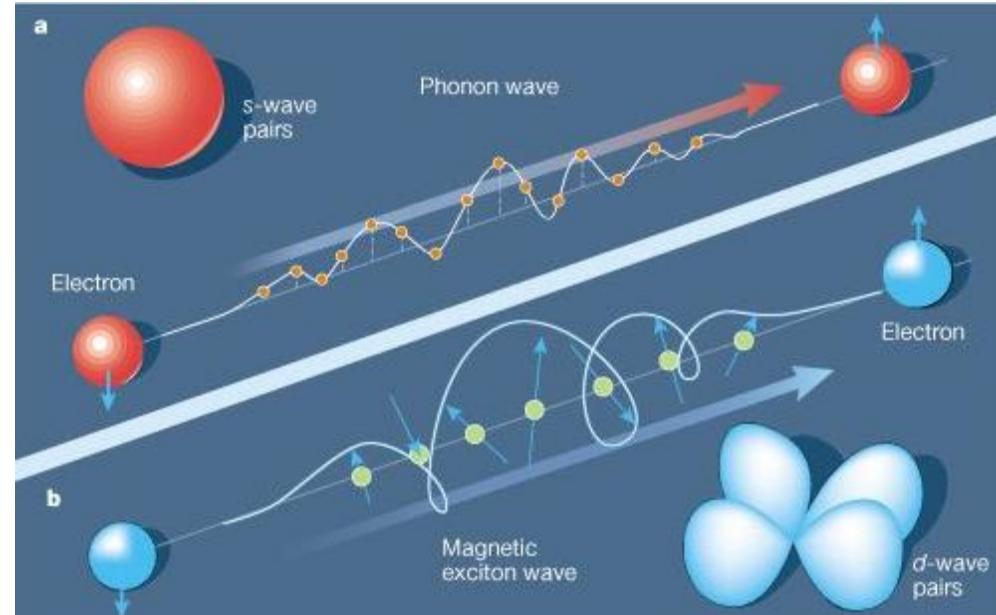
acoustic magnon
("magnetic exciton")

cf. M. Jourdan et al. '99:
anomaly in dI/dV vs V
at $V \approx 1$ mV ($T \ll T_c$)

$$\mathbf{Q} = \mathbf{Q}_{AF} = (0, 0, 1/2)$$

$$T_c = 1.8 \text{ K}$$

Magnetically driven SC



Cooper pairs formed by heavy electrons ("itinerant" 5f electrons)

superconducting glue provided by magnetic excitons in the system of "localized" 5f electrons

Heavy-Fermion Superconductors

	T_c (K)		T_c (K)
CeCu ₂ Si ₂	0.6 ('79 K)	Ce ₃ PdIn ₁₁	0.42 ('15 PR)
[$p = 2.9$ GPa:]	2.3 ('84 GE/GR)]	Ce ₃ PtIn ₁₁	0.32 ('15 PR)
CeNi ₂ Ge ₂	0.2 ('97 DA, '98 CA/GR)	PrOs ₄ Sb ₁₂	1.85 ('01 UCSD)
CeIrIn ₅	0.4 ('01 LANL)	PrIr ₂ Zn ₂₀	0.05 ('10 HI)
CeCoIn ₅	2.3 ('01 LANL)	PrTi ₂ Al ₂₀	0.2 ('12 TO)
Ce ₂ CoIn ₈	0.4 ('02 NA)	β-YbAlB₄	0.08 ('08 TO/IR)
Ce ₂ PdIn ₈	0.7 ('09 WR)	YbRh ₂ Si ₂	0.002 ('16 M/DD)
CePt ₃ Si	0.7 ('04 VI)	Eu metal	$p > 0$ 1.8-2.8 ('09 SL/OS)
CeCu ₂ Ge ₂ $p > 0$	0.6 ('92 GE)	UBe₁₃	0.9 ('83 Z/LANL)
CePd ₂ Si ₂	" 0.4 ('98 CA)	UPt ₃	0.5 ('84 LANL)
CeRh ₂ Si ₂	" 0.35 ('96 LANL)	URu ₂ Si ₂	1.5 ('84 K/DA)
CeCu ₂	" 0.15 ('97 GE)	U ₂ PtC ₂	1.5 ('84 LANL)
CeIn ₃	" 0.2 ('98 CA)	UNi ₂ Al ₃	1.2 ('91 DA)
CeRhIn ₅	" 2.1 ('00 LANL)	UPd ₂ Al ₃	2.0 ('91 DA)
Ce ₂ RhIn ₈	" 2.0 ('03 LANL)	URhGe	0.3 ('01 GR)
CeRhSi ₃	" 1.0 ('05 SE)	UCoGe	3.0 ('07 AM/KA)
CeIrSi ₃	" 1.6 ('06 OS)	UGe ₂	$p > 0$ 0.7 ('00 CA/GR)
CeCoGe ₃	" 0.7 ('07 OS)	UIr	" 0.14 ('04 OS)
Ce ₂ Ni ₃ Ge ₅	" 0.26 ('06 OS)	NpPd₅Al₂	5.0 ('07 OS)
CeNiGe ₃	" 0.4 ('06 OS)	PuCoGa₅	18.5 ('02 LANL)
CePd ₅ Al ₂	" 0.57 ('08 OS)	PuRhGa₅	8.7 ('03 KA)
CeRhGe ₂	" 0.45 ('09 OS)	PuCoIn₅	2.5 ('12 LANL)
CePt ₂ In ₇	" 2.1 ('10 LANL)	PuRhIn₅	1.7 ('12 LANL)
CeIrGe ₃	" 1.5 ('10 OS)	Am metal	$p > 0$ 2.4;1.7 ('05 KA)
CeAu ₂ Si ₂	" 2.5 ('14 GE)	YFe₂Ge₂	1.8 ('14 CA)
		CrAs	$p > 0$ 1.7 ('14 BEI/TO)

outline

Kondo effect

Heavy-fermion (HF) superconductivity (SC)

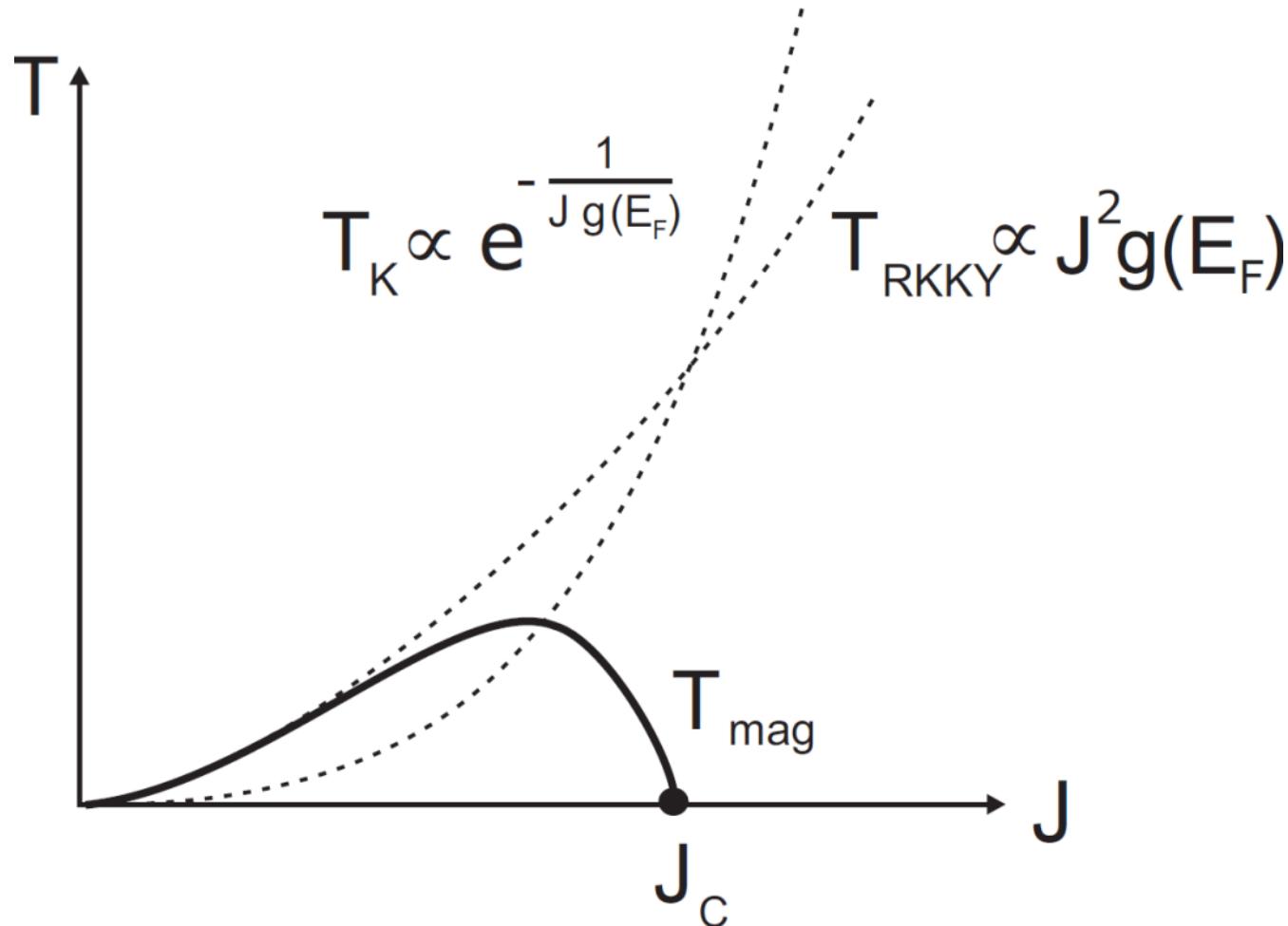
Quantum critical point (QCP) in HF metals

SC near an itinerant AF (SDW) QCP in CeCu_2Si_2

SC due to nuclear AF order in YbRh_2Si_2

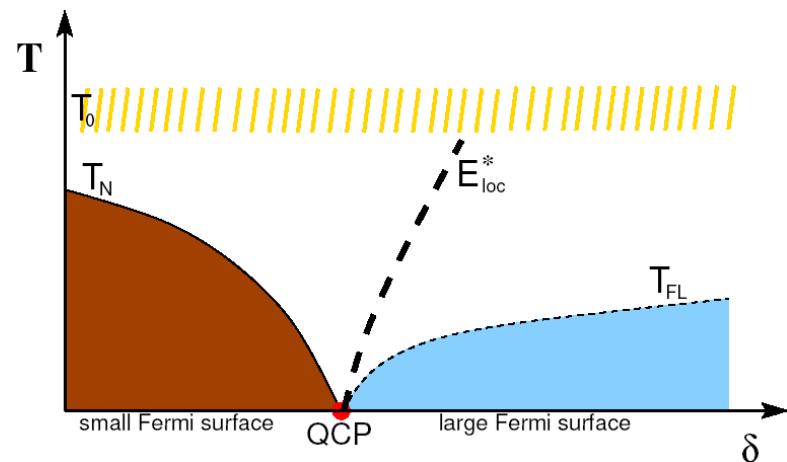
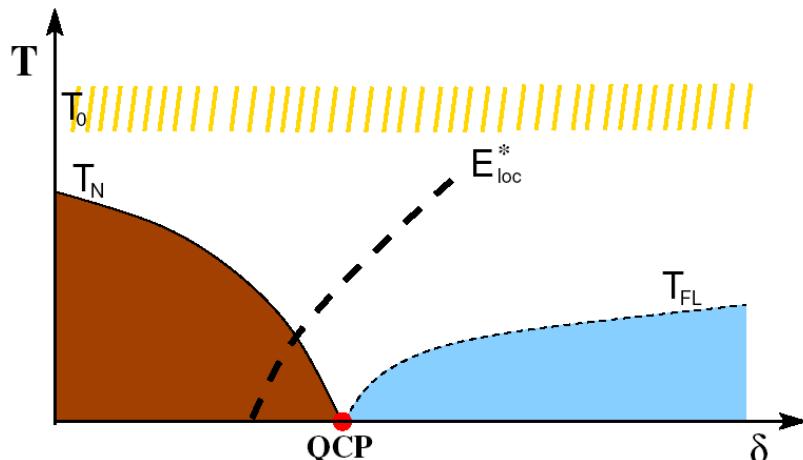
Doniach phase diagram

[S. Doniach, Physica B+C 91, 231 (1977)]



Two types of QCPs

[P. Gegenwart, Q. Si & F.S., Nature Phys. 4, 186 (2008)]

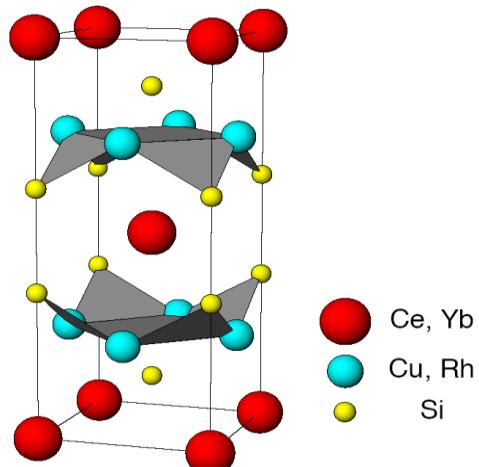


- SDW QCP
(Hertz, Moriya, Millis...)
HF's intact
FS large

exemplary material:



$$T_K \approx 15 \text{ K}$$



- Kondo breakdown QCP
(Si et al., Coleman et al., 2001)
HF's disintegrate
abrupt FS reconstruction



$$T_K \approx 30 \text{ K}$$

Quantum critical paradigm: AF QCP in pure HF metal \sim unconventional SC!

BCS – type SC?

No SC? $T_c < 10 \text{ mK}$

outline

Kondo effect

Heavy-fermion (HF) superconductivity (SC)

Quantum critical point (QCP) in HF metals

SC near an itinerant AF (SDW) QCP in CeCu_2Si_2

SC due to nuclear AF order in YbRh_2Si_2

CeCu₂Si₂

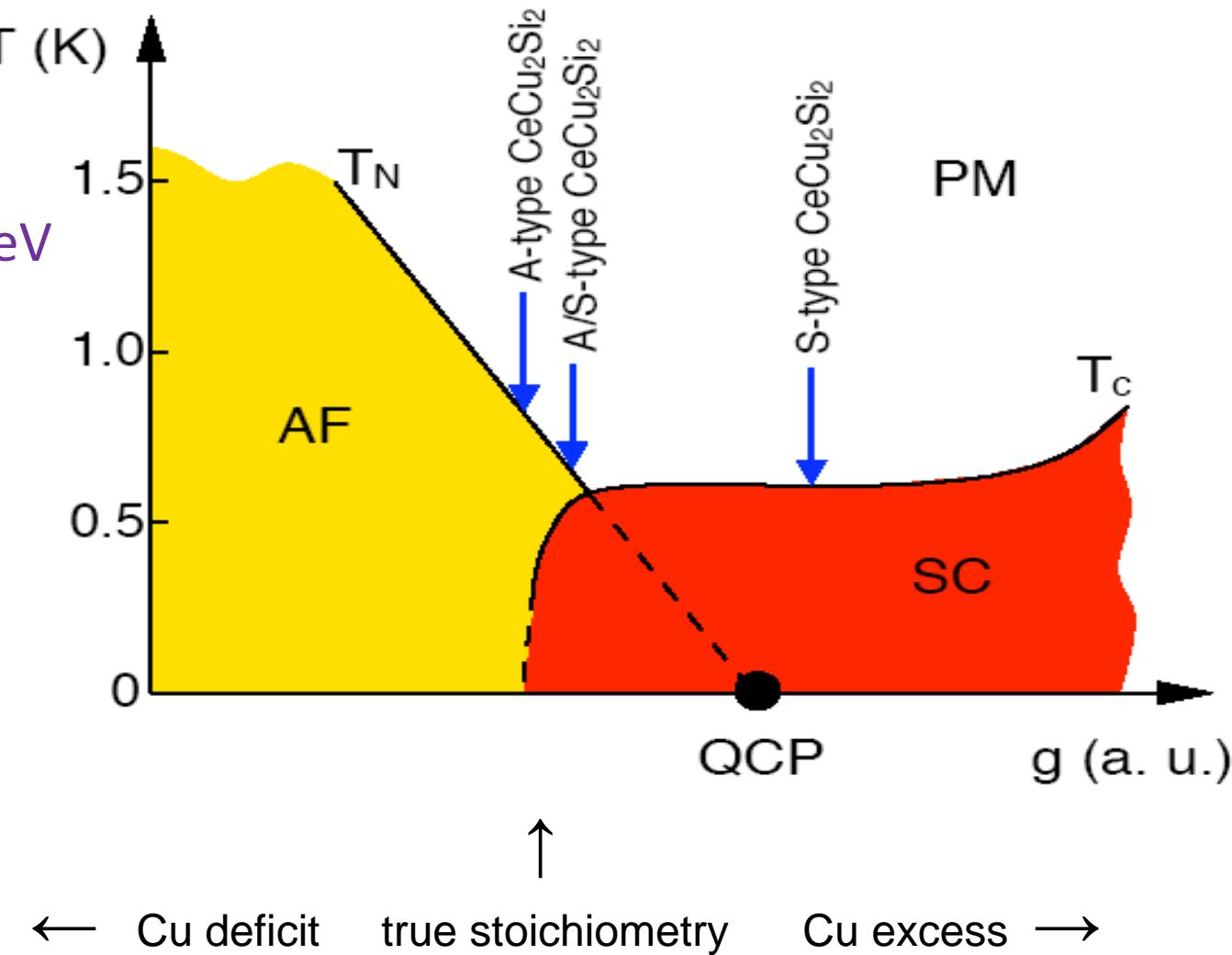
Homogeneity range: ~ 1% Cu/Si site exchange

$$\Delta_{\text{CF}} \gg k_B T_K \sim J_K \approx 2 \text{ meV}$$

$$S_{\text{eff}} = 1/2$$

Pairing interactions

$$\leq I_{\text{RKKY}} \approx J_K$$

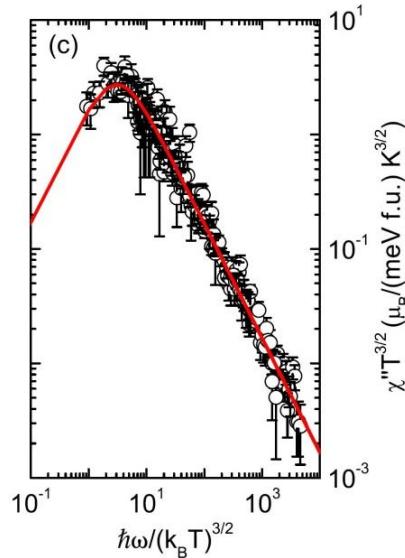


Quantum criticality in CeCu₂Si₂

$\Delta\rho \sim T^{3/2}$, $\gamma = \gamma_0 - bT^{1/2}$ P. Gegenwart et al. , Phys. Rev. Lett. **81**, 1501 ('98)

$$\chi'' T^{3/2} = f(\hbar\omega/(k_B T)^{3/2})$$

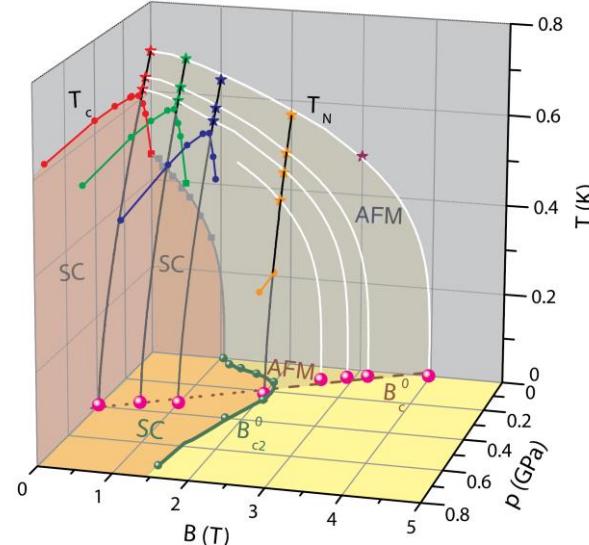
J. Arndt et al., Phys. Rev. Lett.
106, 246401 ('11)



3D-SDW QCP

B - p phase diagram

E. Lengyel et al.,
Phys. Rev. Lett. **107**, 057001 ('11)



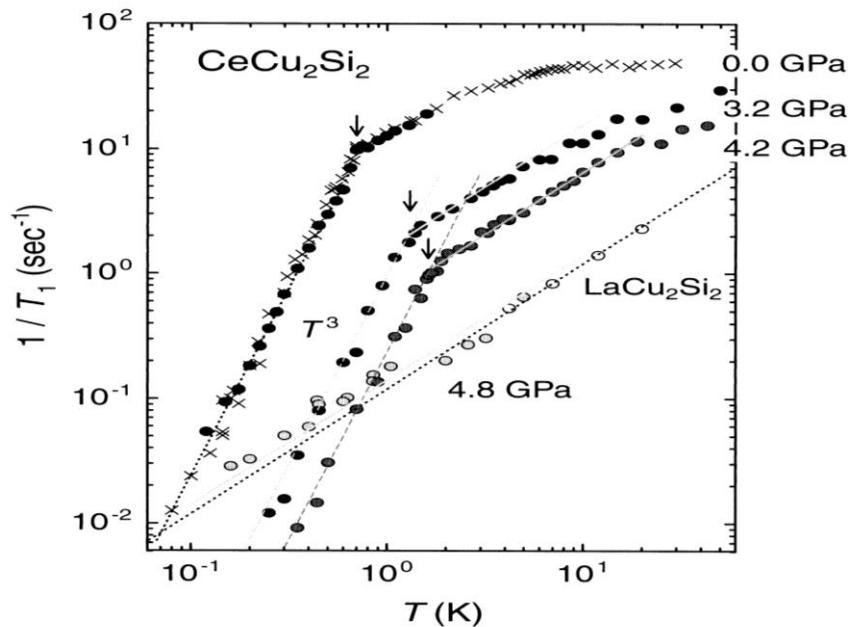
(1-band) d -wave superconductivity in CeCu_2Si_2

Cu NQR

K. Fujiwara et al., *JPSJ* **77**, 123711 ('08)

cf. also

K. Ishida et al., *PRL* **82**, 5353 ('99)



$T = T_c$: no Hebel-Slichter peak

$T < T_c$: $1/T_1 \sim T^3$

↷ d -wave SC, nodes of $\Delta(k)$

strong coupling

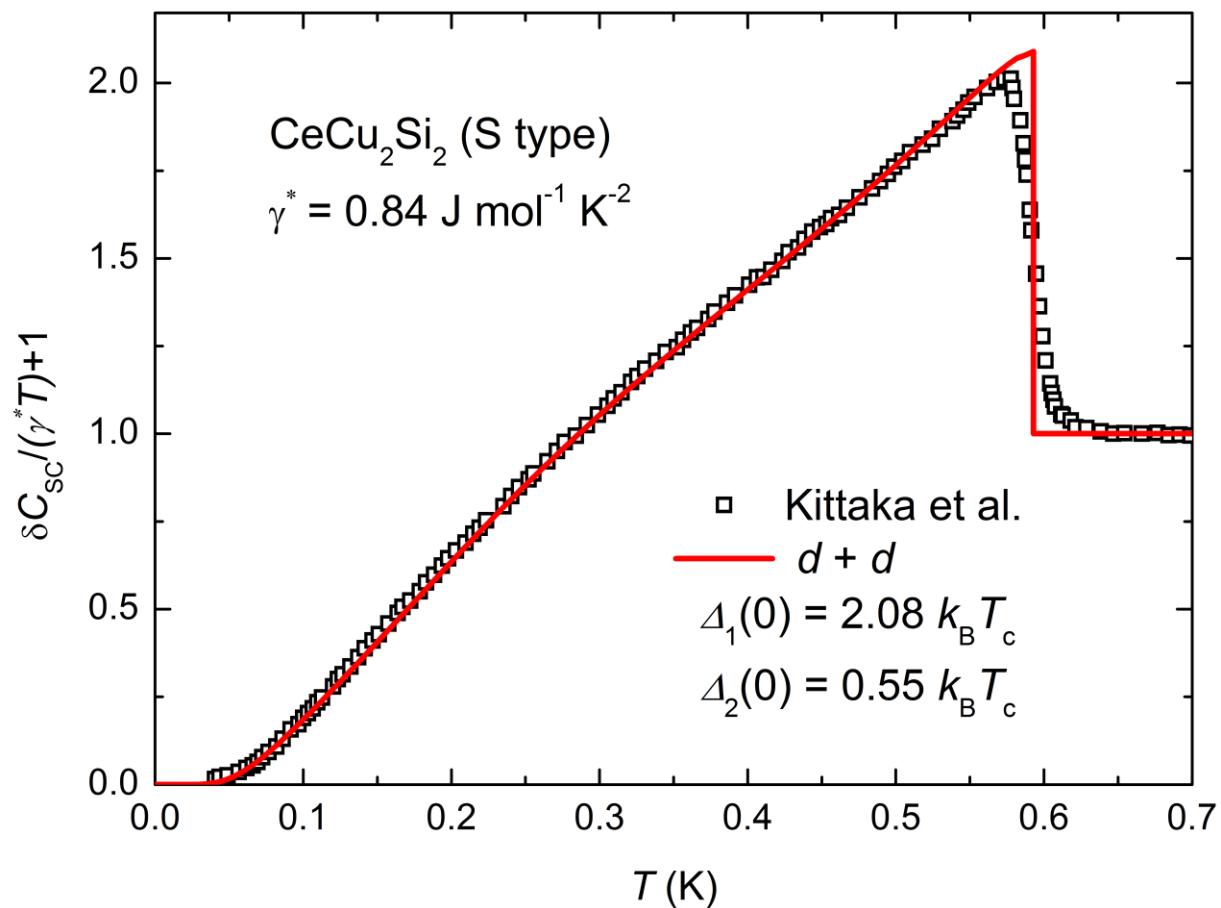
d -wave SC: $2\Delta_0/k_B T_c = 5$

[weak coupling

d -wave SC: $2\Delta_0/k_B T_c = 4.3$

T - dependence of specific heat for CeCu_2Si_2

[S. Kittaka et al., *PRL* **112**, 067002 (2014)]



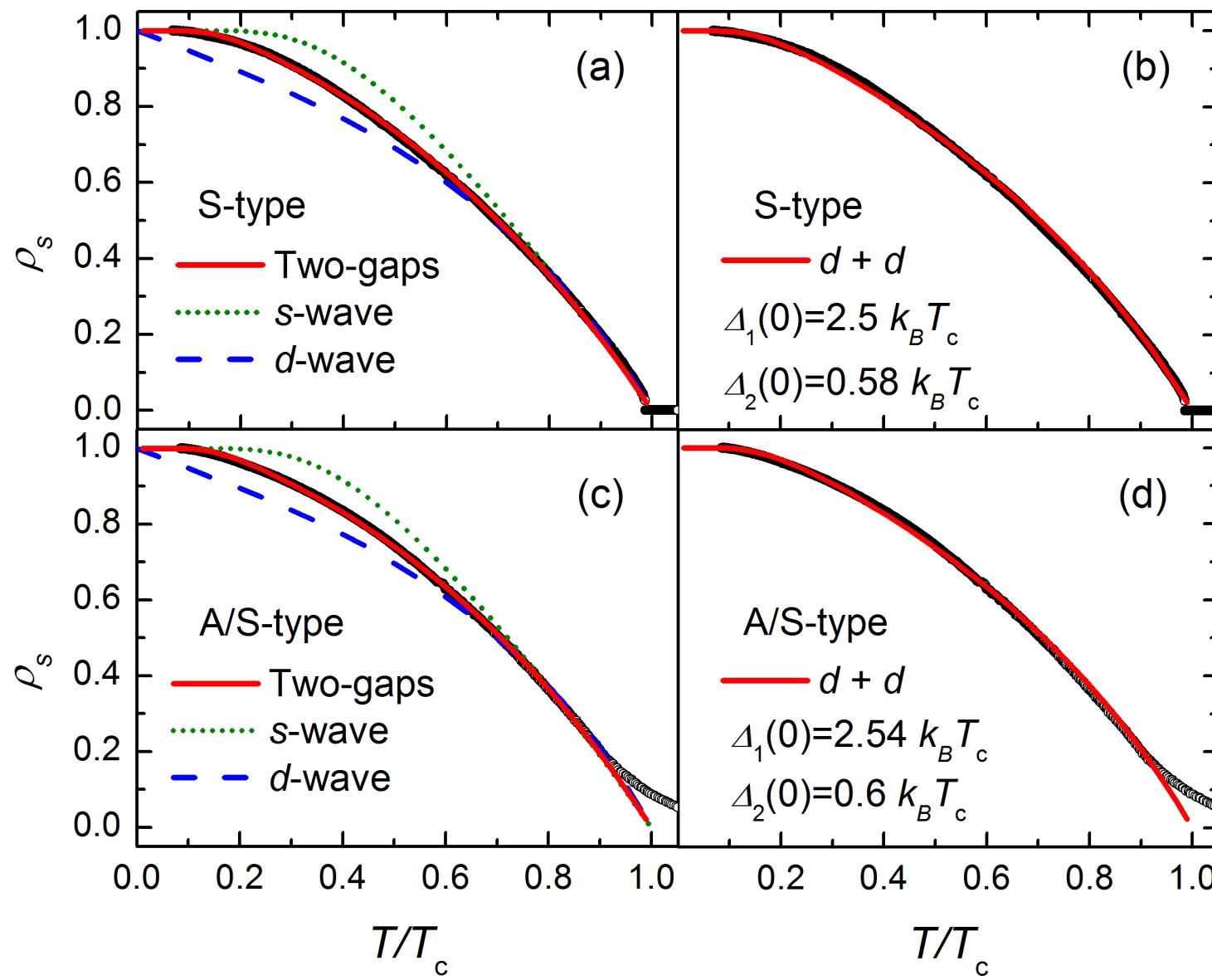
T. Takenaka et al., *PRL* **119**, 077001 (2017): T_c insensitive against el-irradiation

~

CeCu_2Si_2 : Two-band **s-wave superconductor** without sign-changing $\Delta(\mathbf{k})$ [BCS SC]

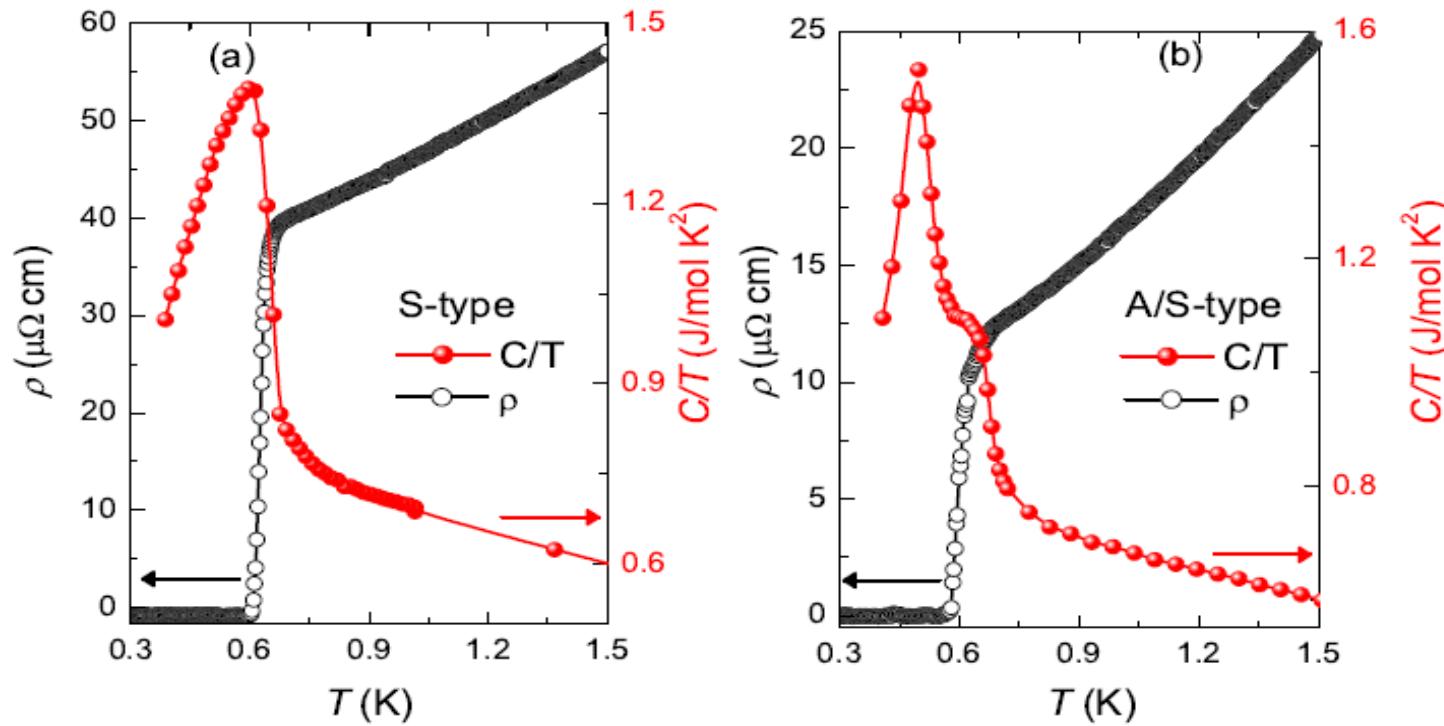
T - dependence of superfluid density $\varrho_s(T)$ in CeCu_2Si_2

[G. M. Pang et al., PNAS 115, 5343 (2018)]



Harmless disorder in CeCu_2Si_2

G. M. Pang et al., *PNAS* **115**, 5343 (2018)



$T_c \simeq 0.6 \text{ K}$ insensitive against variations of ϱ_0 : $\varrho_0(\text{"S"}) \simeq 4 \varrho_0(\text{"A/S"})$

- Cu/Si interchange < 1 % (change of ϱ_0)
- shift from lattice sites into interstitials (el. irradiation)

harmless

Atomic substitution in CeCu_2Si_2

[H. Spille, U. Rauchschwalbe, FS, *Helv. Phys. Acta* **56**, 165 (1983);

H. Q. Yuan, F. M. Grosche, M. Deppe et al., *Science* **302**, 2104 (2003)]

site dependence

Si-site:

x_c : (15 – 20) at% for Ge

$x = 0.1$: $l_{\text{mfp}} > \xi$; 0.25 : $l_{\text{mfp}} < \xi$

Cu-site:

$x_c \approx 1$ at%

for Mn, Pd, Rh ($\Delta T_K \approx +7$ mK)

Ce-site

(size-dependence):

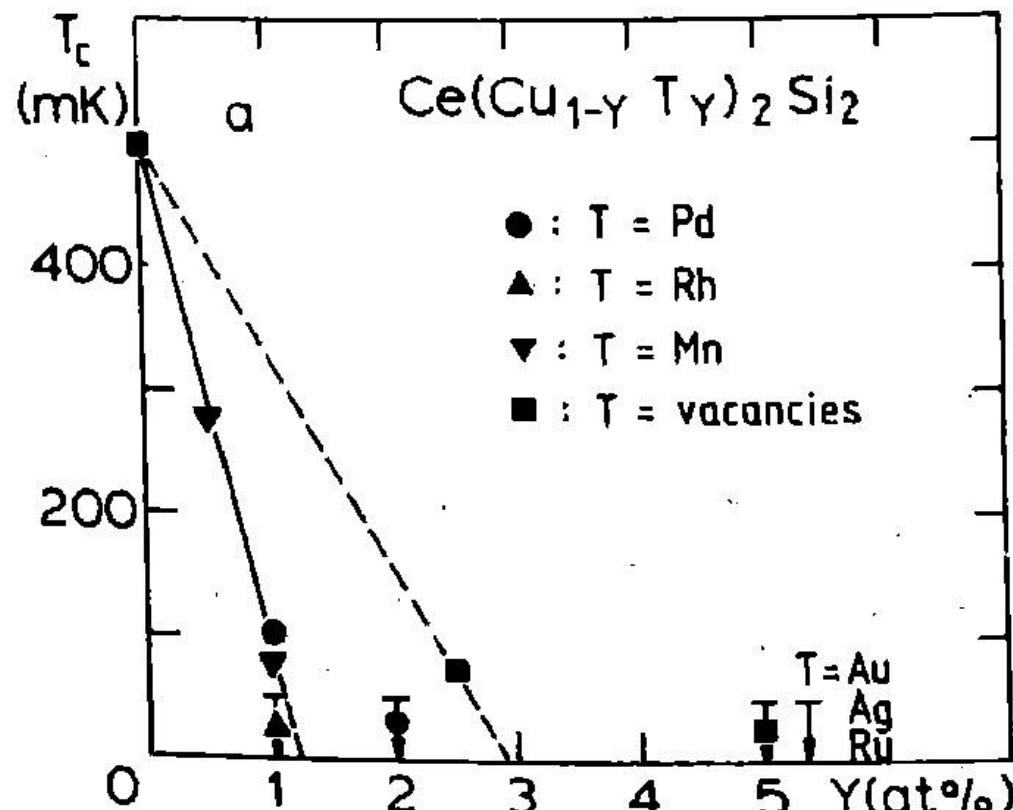
$\Delta r_{\text{Ce-M}}$ [Å] x_c [at%]

Sc + 0.28 1

Y + 0.13 6

Th + 0.06 20

La - 0.03 10

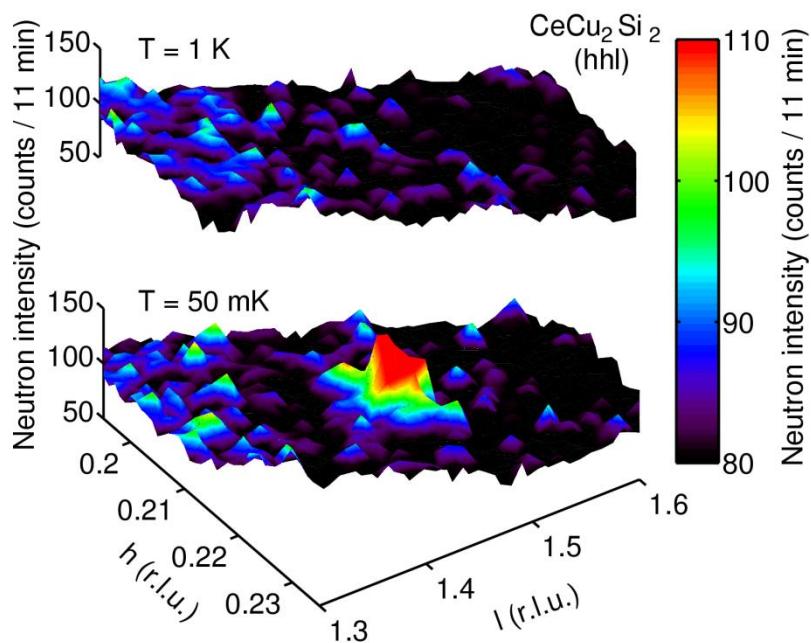


Incompatible with s_{++} pairing

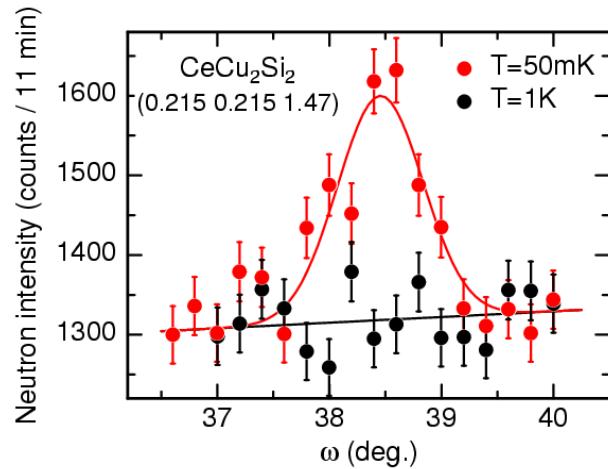
[P. W. Anderson, *Phys. Rev. Lett.* **3**, 325

Nature of the AFM (A) phase in CeCu₂Si₂

[O. Stockert, G. Zwicknagl et al., Phys. Rev. Lett. **92**, 136401 (2004)]



Observation of AF satellite peaks
in $(hh\bar{l})$ scattering plane

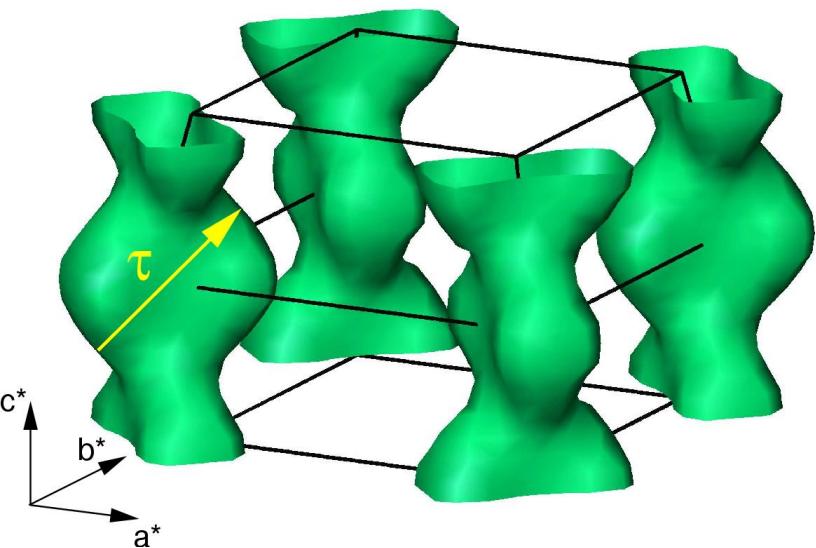


Long-range AF order with
propagation vector
 $\tau = (0.215 \ 0.215 \ 0.530)$ at $T = 50 \text{ mK}$

$$T_N \approx 0.8 \text{ K}$$
$$m_0 \sim 0.1 \ \mu_B$$

Nesting properties of Fermi surface in CeCu_2Si_2

[O. Stockert, G. Zwicknagl et al., PRL 92, 136401 (2004)]

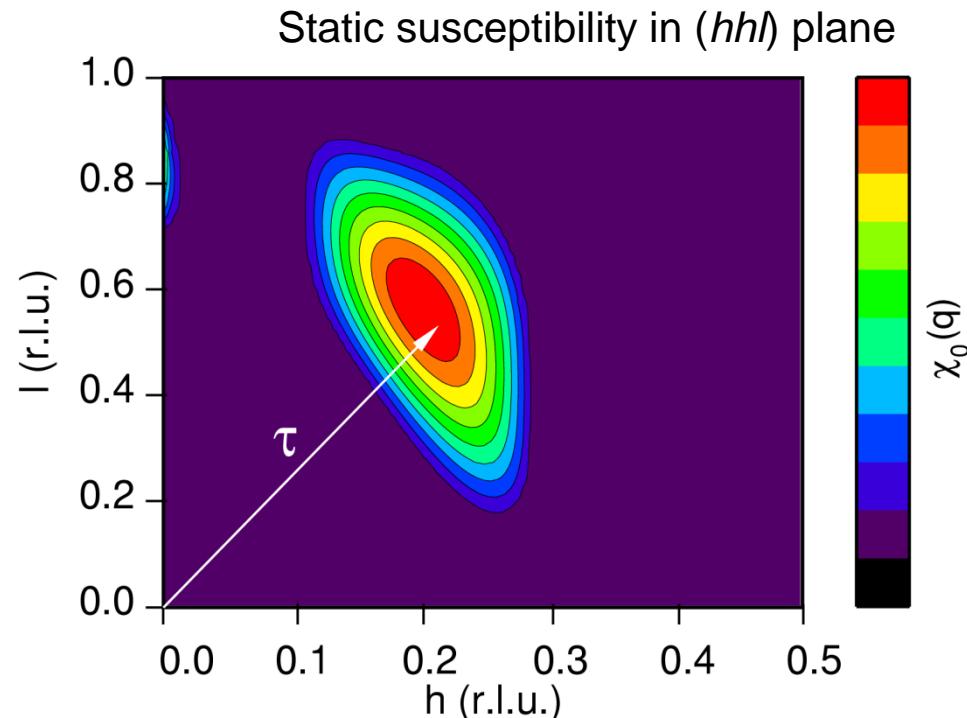


Nesting for incommensurate wave vector
 $\tau \approx (0.21 \ 0.21 \ 0.55)$

Fermi surface unstable with respect to formation of spin-density wave

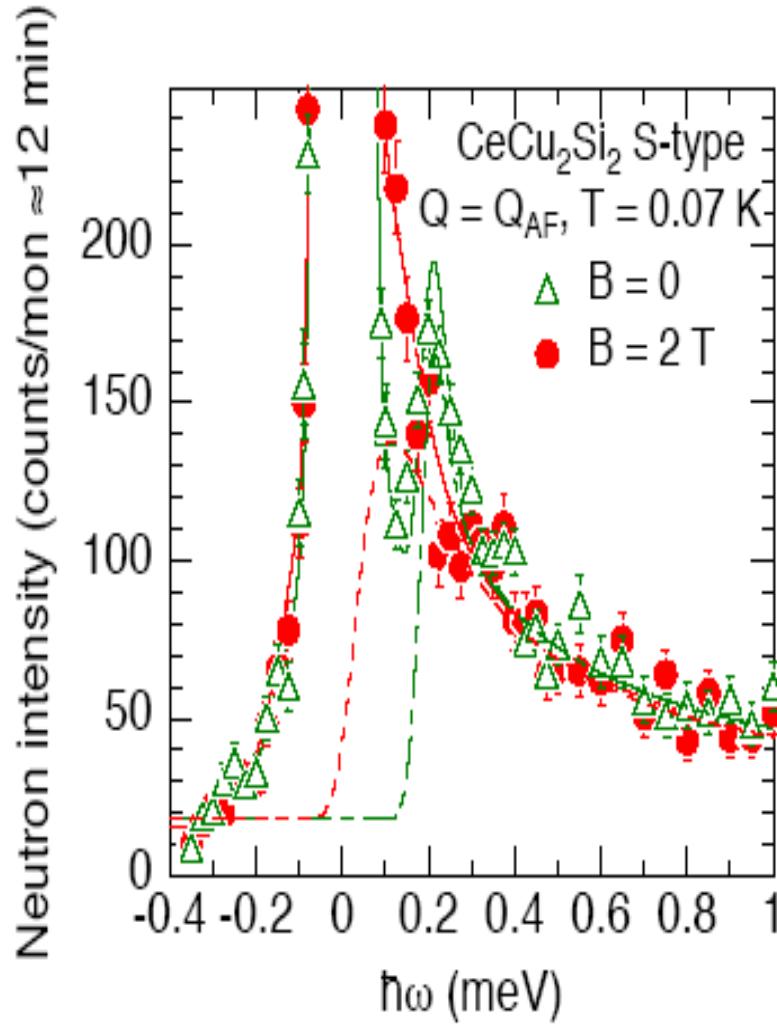
Fermi surface of heavy quasiparticles calculated with renormalized band method,
 $m^* \approx 500 m_e$

warped columns along tetragonal axis



Inelastic n-scattering reveals sign change of $\Delta(\mathbf{k})$

[O. Stockert et al., *Nature Phys.* 7, 119 (2011)]



Large INS intensity in sc state at $\mathbf{k} = \mathbf{Q}_{\text{AF}}$ and low $\hbar\omega$, i.e.,
coherence factor $\{1-\cos[\Phi(\mathbf{k})]\} \simeq 2$,
where $\Phi(\mathbf{k})$ is the
phase difference in $\Delta(\mathbf{k})$ between \mathbf{k} & $\mathbf{k} + \mathbf{Q}_{\text{AF}}$,
 $\simeq \Phi(\mathbf{k}) \simeq \pi$
 \simeq sign change of $\Delta(\mathbf{k})$ along \mathbf{Q}_{AF}
inside dominating HF band

→
No s - wave superconductor

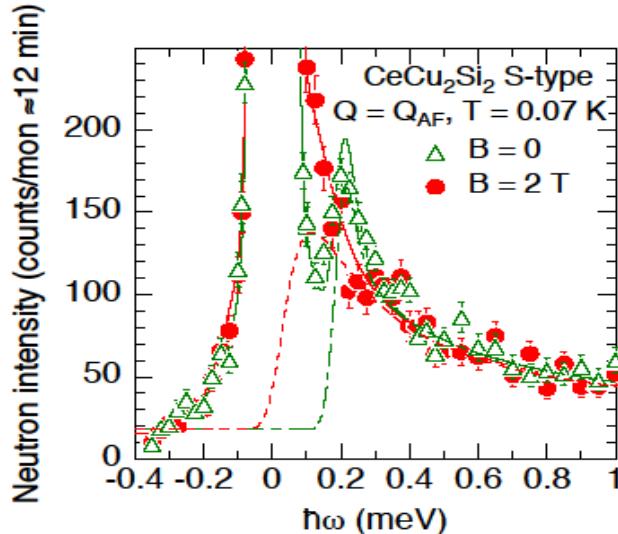
s_{++} : doesn't show sign change in $\Delta(\mathbf{k})$!
no onsite pairing of HFs: $U_{\text{eff}} \simeq k_B T_K$!
 s_+ : nesting wavevector *different* from \mathbf{Q}_{AF}
can't explain spin resonance!

→
“d+d band - mixing” Cooper pairing
[E. Nica et al. '16]

2 - band *d* - wave SC without nodes,
cf. ${}^3\text{He}$ - B phase (*p* - wave pairing)

Superconductivity in CeCu_2Si_2 near a (3D) SDW QCP

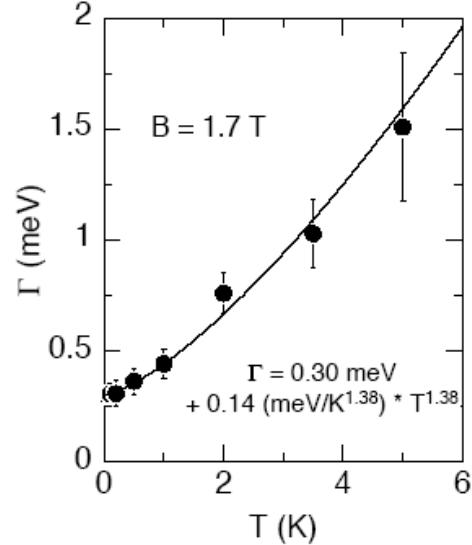
[O. Stockert et al., *Nature Phys.* **7**, 119 (2011)]



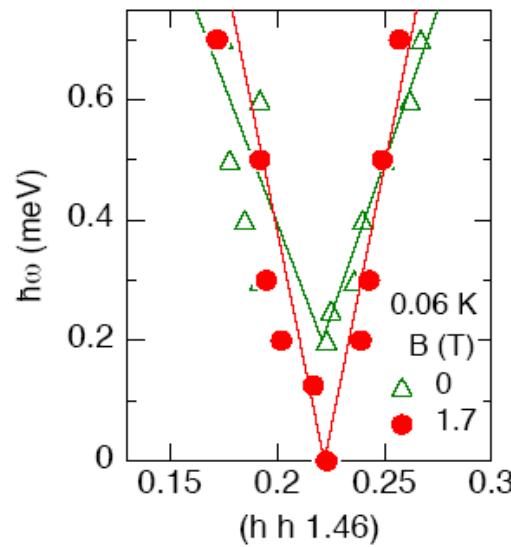
$B = 2 \text{ T}$: quasielastic line, HWHM $\sim T_K$
 $B = 0$: spin gap below peak at 0.2 meV

Propagating ‘paramagnon’ mode (not a ‘spin resonance’) at $3.9 k_B T_c$ [$< 2\Delta_1(T=0) \simeq 5 k_B T_c$]

[J. Arndt et al., *PRL* **106**, 246401 (2011)]



“slowing down”
 $\Delta\Gamma \sim T^\alpha$
 $\alpha = 1.38 \pm 0.16$
(3D SDW) $\alpha = 1.5$



outline

Kondo effect

Heavy-fermion (HF) superconductivity (SC)

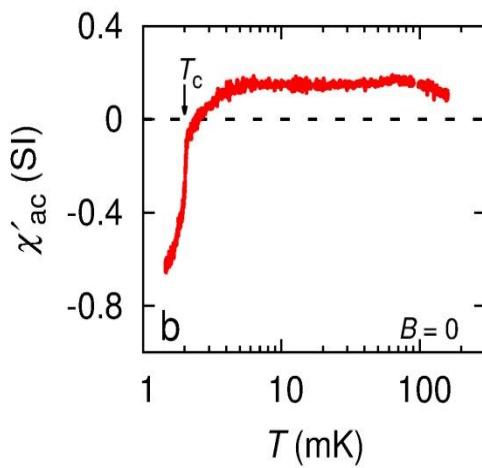
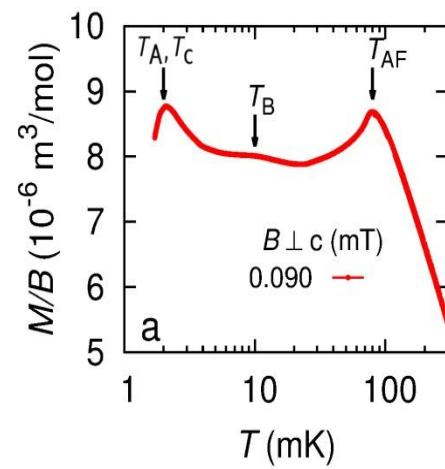
Quantum critical point (QCP) in HF metals

SC near an itinerant AF (SDW) QCP in CeCu_2Si_2

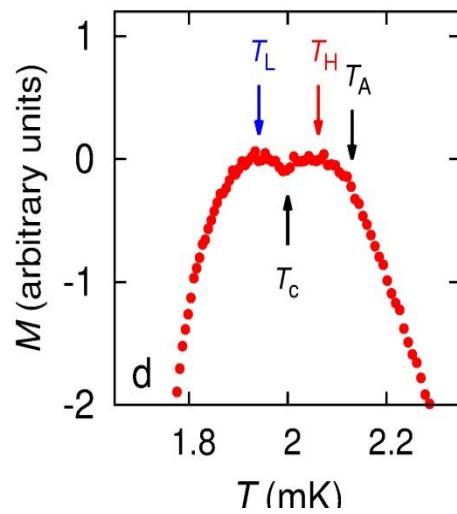
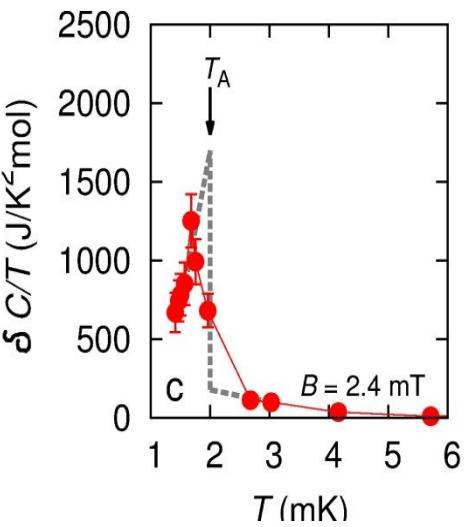
SC due to nuclear AF order in YbRh_2Si_2

YbRh_2Si_2 : Emergence of SC by nuclear AF order

[E. Schuberth et al., *Science* **351**, 485 (2016)]



a. $T_N \approx 70 \text{ mK}$: 4f - electr. AF order
 $T_B \approx 10 \text{ mK}$: increase of $M(T)$
 $T_A \approx 2 \text{ mK}$: new phase transition



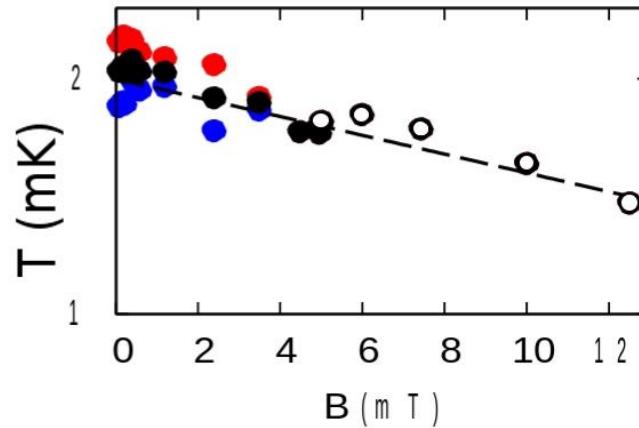
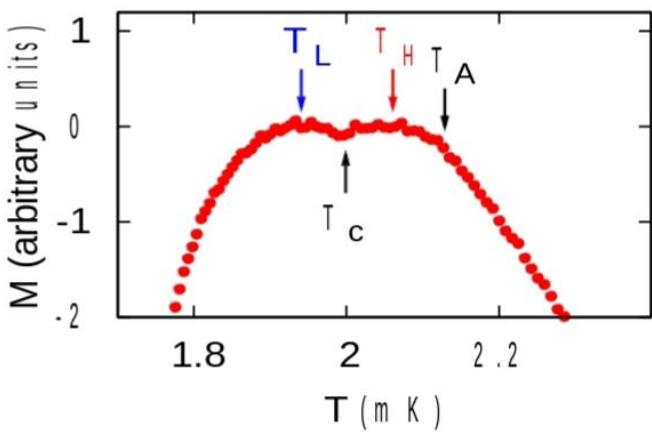
b. $T_c \approx 2 \text{ mK}$: SC [$\chi'(T)$: 1st order!]

c. $T_A \approx 2 \text{ mK}$: “A - phase”

d. $B < 4 \text{ mT}$: $T_A > T_c$

Summary YbRh_2Si_2

E. Schubert et al. (2016)



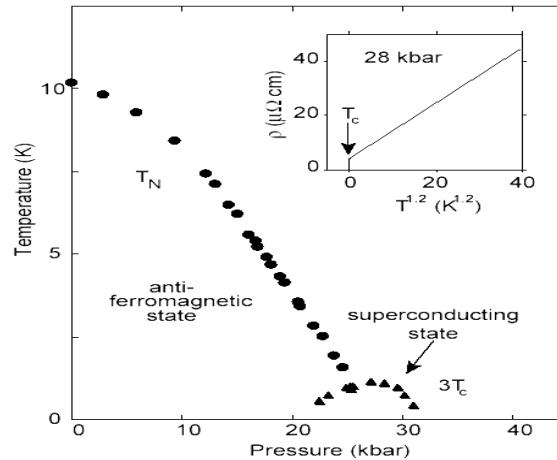
$B_{c2}' \simeq 25$ T/K
from Meissner measurements
(same from shielding measurements)

- $M_{\text{DC}}(T)$, $\chi_{\text{AC}}(T)$ prove : (bulk) heavy-fermion SC at $B < 4$ mT
- YbRh_2Si_2 :
 - SC near (4f – “Mott – type”) transition ($T = 0$), like CeRhIn_5 at $p > 0$
 - both systems form link between ($\simeq 50$) HFSCs and cuprates, organics, ... near true Mott transition

Quantum Critical Paradigm: Unconventional SC at HF AF QCPs

CePd₂Si₂

N. D. Mathur et al.,
Nature **394**, 39 (1998)



- YbRh₂Si₂:
HF SC, $T_c = 2$ mK
- CeCu₂Si₂:
fully gapped 2 - band d - wave superconductor
- Unconventional SC near AF QCPs: robust phenomenon

Further reading:
M. Smidman et al., *Phil. Mag.* **98**, 2930 (2018)