Discontinuous change of Hall coefficient across the field-induced phase boundaries in $U(Ru_{1-x}Rh_x)_2Si_2$

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Outline

- Backgrounds of URu_2Si_2 (Γ_5 model)
- Metamagnetic quantum criticality in URu_2Si_2
- Multiple phase formation in $U(Ru,Rh)_2Si_2$
- Hall effect studies: discontinuous FS change in $U(Ru,Rh)_2Si_2$
- Implications to HO

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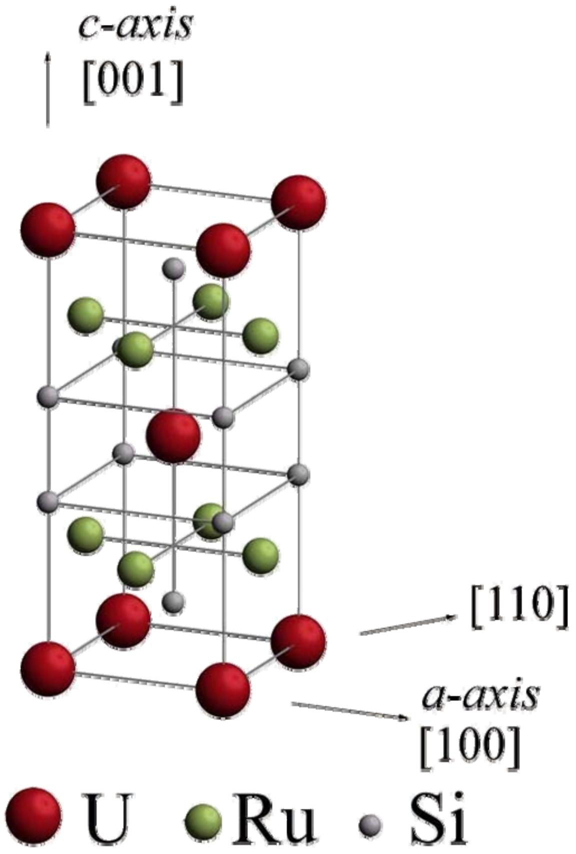
URu_2Si_2, Structure and Heavy Fermion States

Body Centered Tetragonal

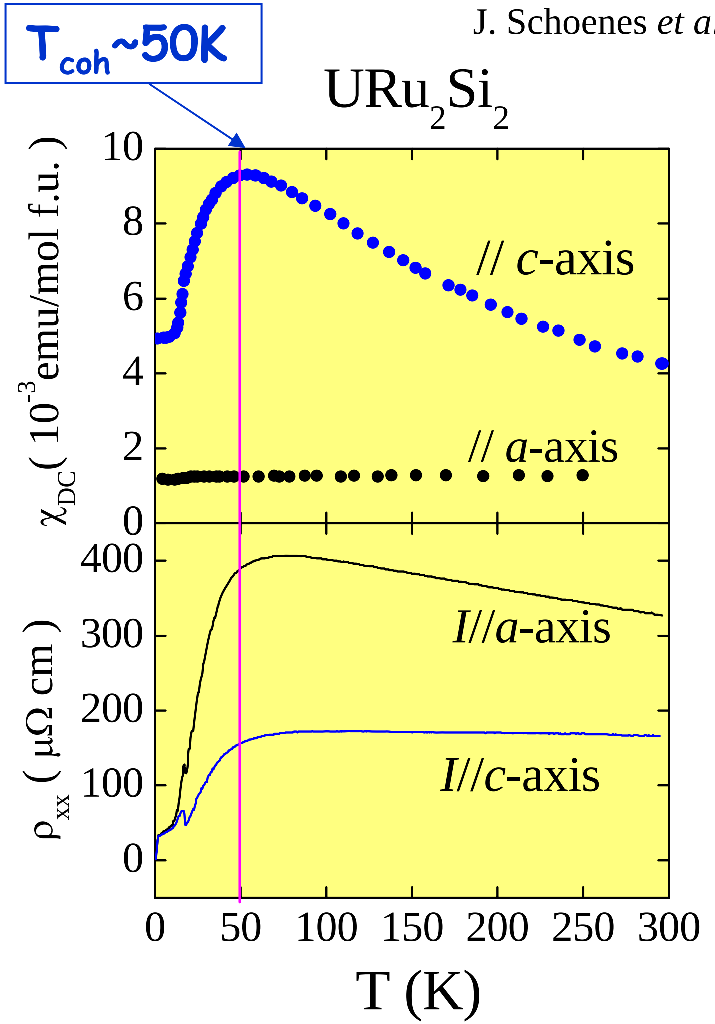
a= 4.124 Å , c=9.582 Å at 4.2 K



W. Schlabitz et al.,
4th Int. Conf. on Val. Fluc.
Cologne, **1984**. (polycrystals)

A.A. Menovsky, single crystals grown by Czochralski "tri-arc" method, **1985**.

J. Schoenes *et al*. PRB (1987)

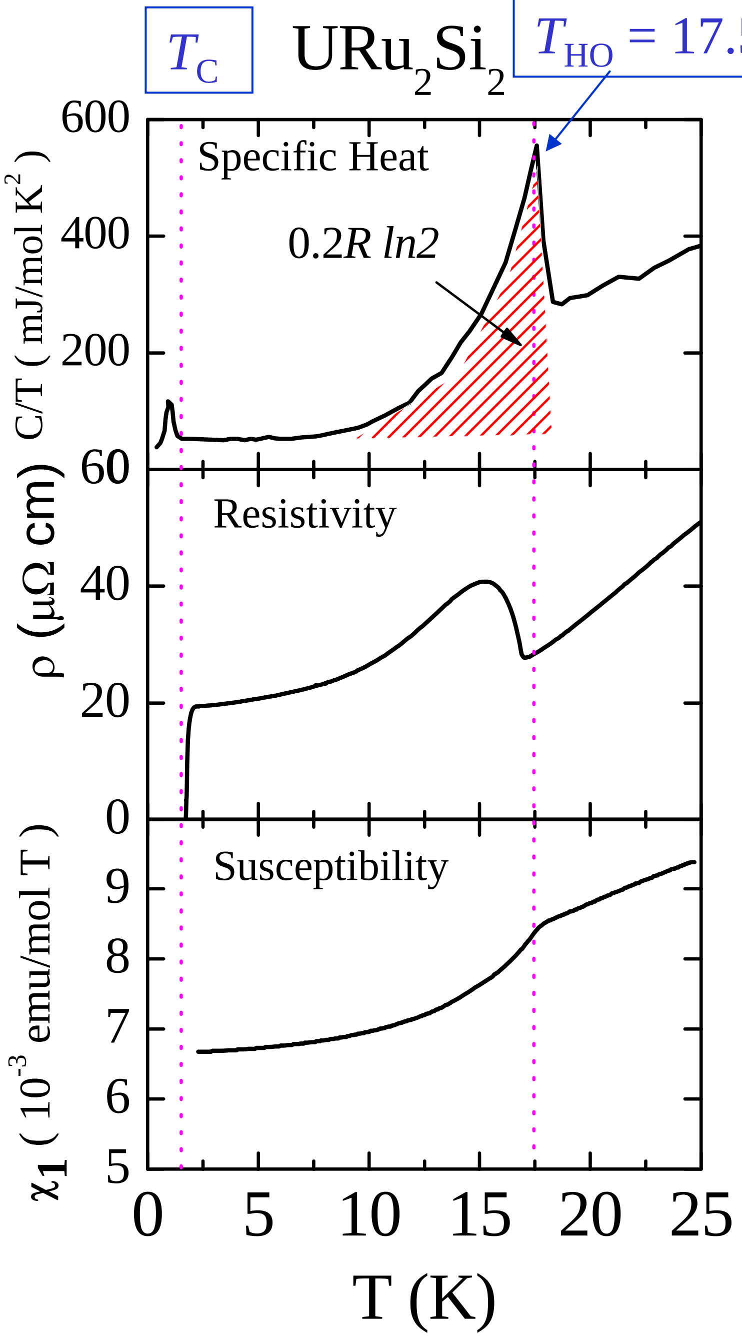


T. T. M. Palstra *et al*., PRL 55, 2727 (1985); PRB 33, 6527 (1986)

$U(Ru,Rh)_2Si_2$: Kondo lattice system, forming 5f quasiparticle bands with Ising like spin degrees of freedom (very anisotropic *g*-factor; g_z=2.6, g_x=g_y=0)

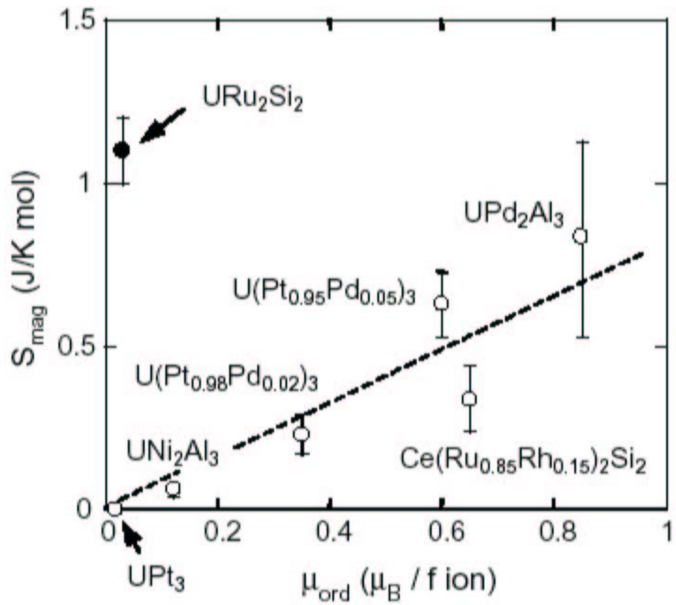
[N. Harrison *et al*., cond-mat/0506384]

URu_2Si_2, Hidden Order (HO) Phase



Palstra *et al.*,PRL55, 2787 (1985); Ramirez et al., PRL 68,2680 (1992);K. H. Kim (unpublished).

[Amitsuka *et al*. Physica B 312,390(2002)].



Small AFM moment 0.03 μ_B can't account for large entropy change [Broholm *et al*. PRB 43, 12809(1991)]

Numerous proposals for the HO

<u>Dipolar</u>

<u>Nondipolar</u>

Orbital antiferromagnetism

Unconventional density wave

Helical order

Electric quadrupole order

…

URu_2Si_2, Microscopic Magnetism: key facts

^{29}Si NMR under P

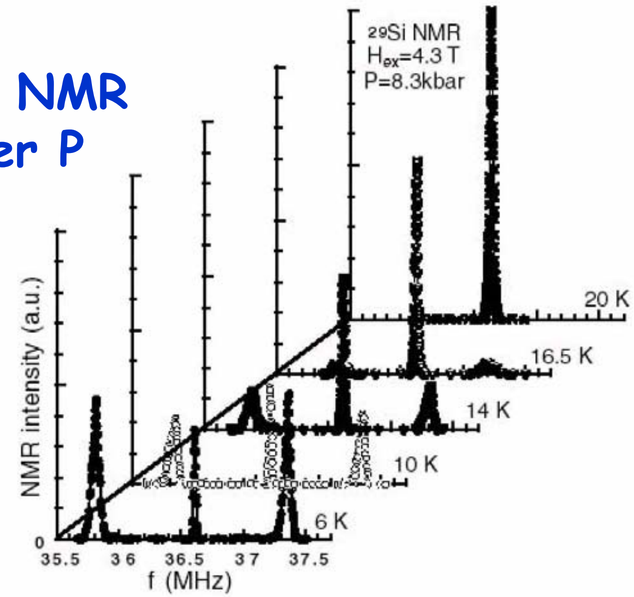


FIG. 1. T evolution of ^{29}Si NMR spectrum for $H_{ex} \parallel c$ axis at 8.3 kbar.

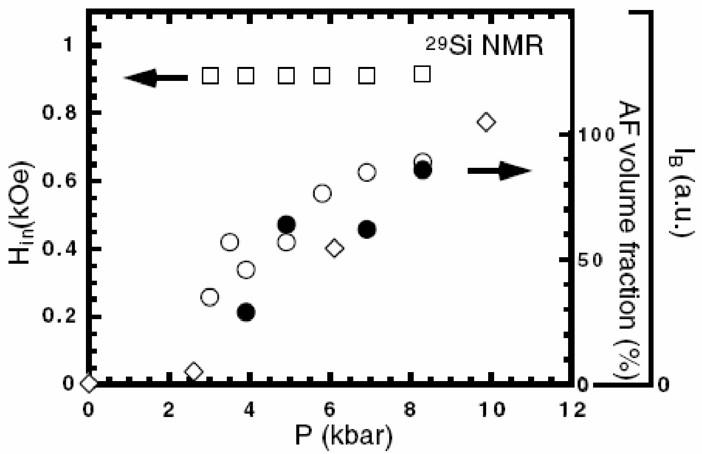


FIG. 3. Pressure variations of H_{in} (□) and the AF volume fraction (•, ∘) obtained by ^{29}Si NMR (see text). For comparison, the (100) magnetic Bragg intensity, I_B, data (◇) deduced from Ref. [15] are also plotted against pressure in the figure.

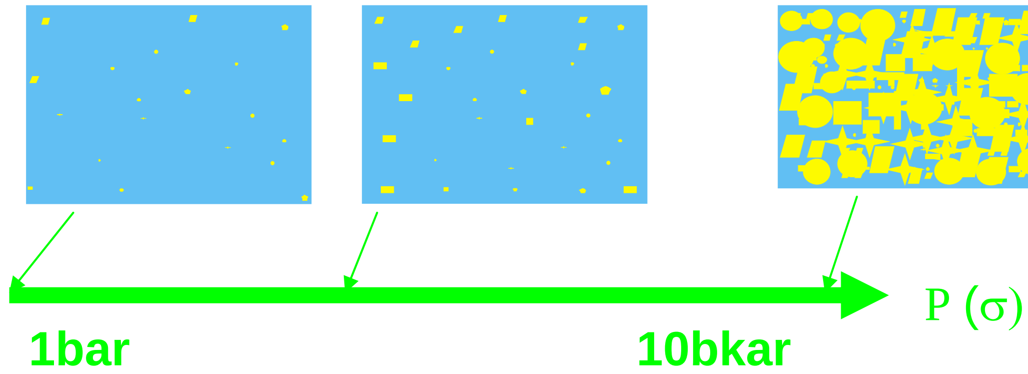
[Matsuda *et al*. PRL 87, 087203(2001)]

Antiferromagnetic Phase is MINORITY PHASE !!

NMR intensity dependent on pressure while internal field (splitting) is independent; <u>AFM Bragg peaks seen by neutrons is from minority phase</u>

<u>*One dominant picture (still under debate):*</u>

At *P*=1 atm, 1 % AF phase coexists with the paramagnetic HO phase (very strain sensitive)

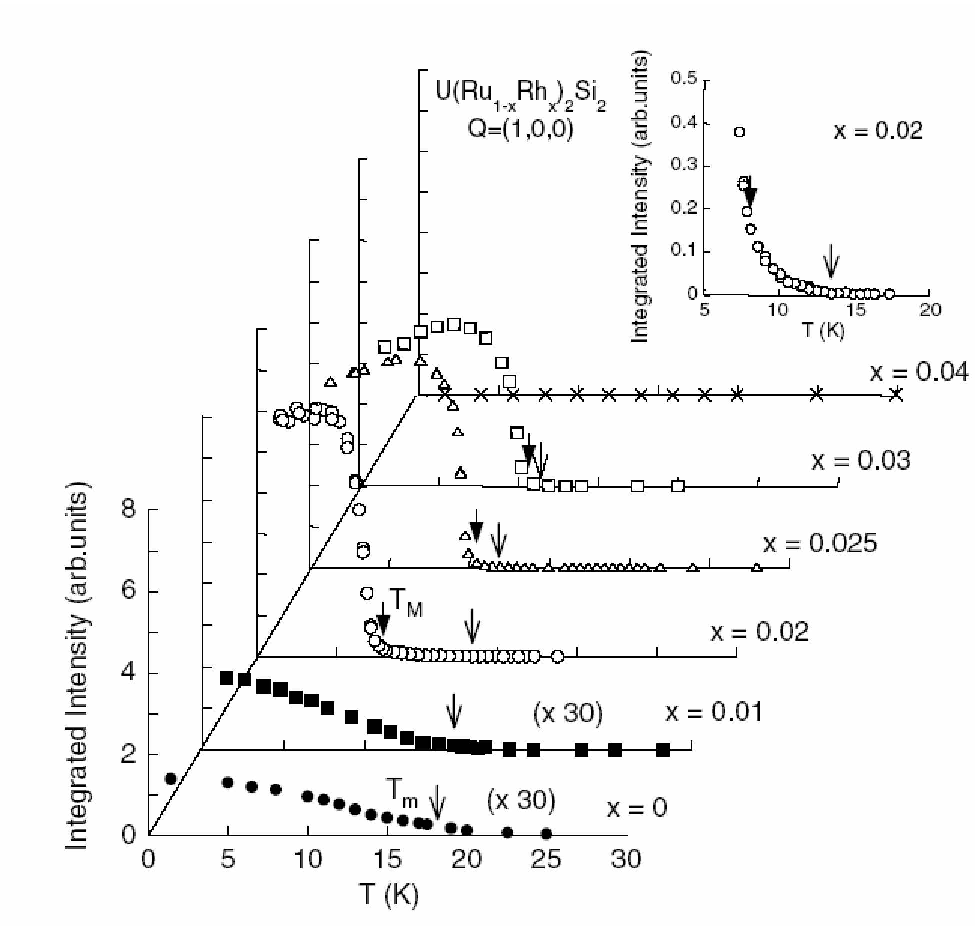


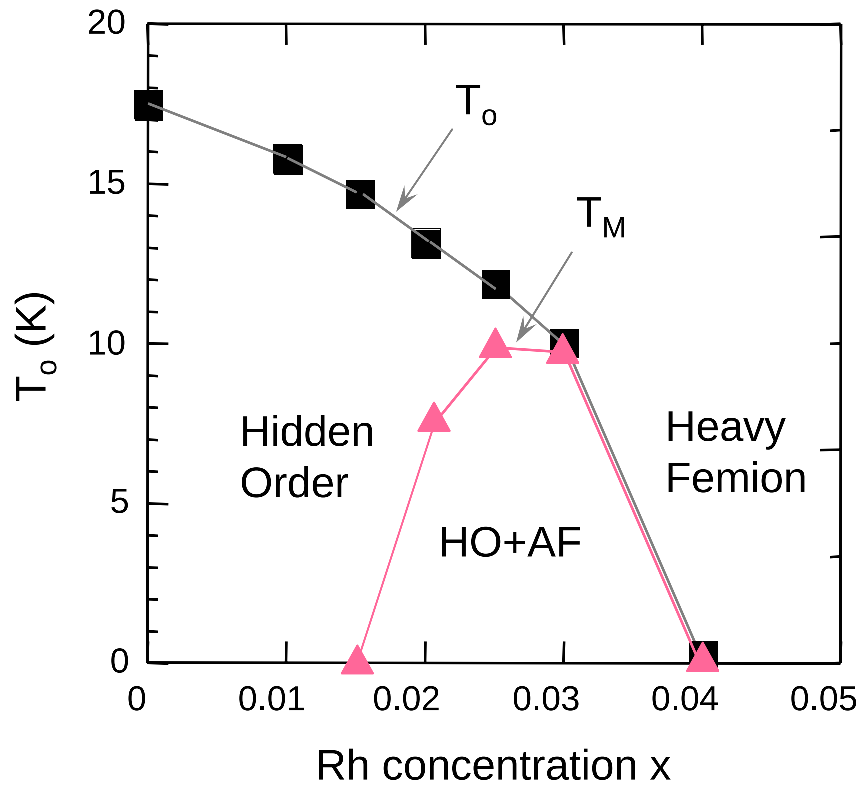
P: hydrostatic pressure

σ: Uniaxial pressure (*c/a* ratio)

HO competes with AFM with P or σ

$U(Ru_{1-x}Rh_x)_2Si_2$: Effects of Rh doping (x < 0.05)





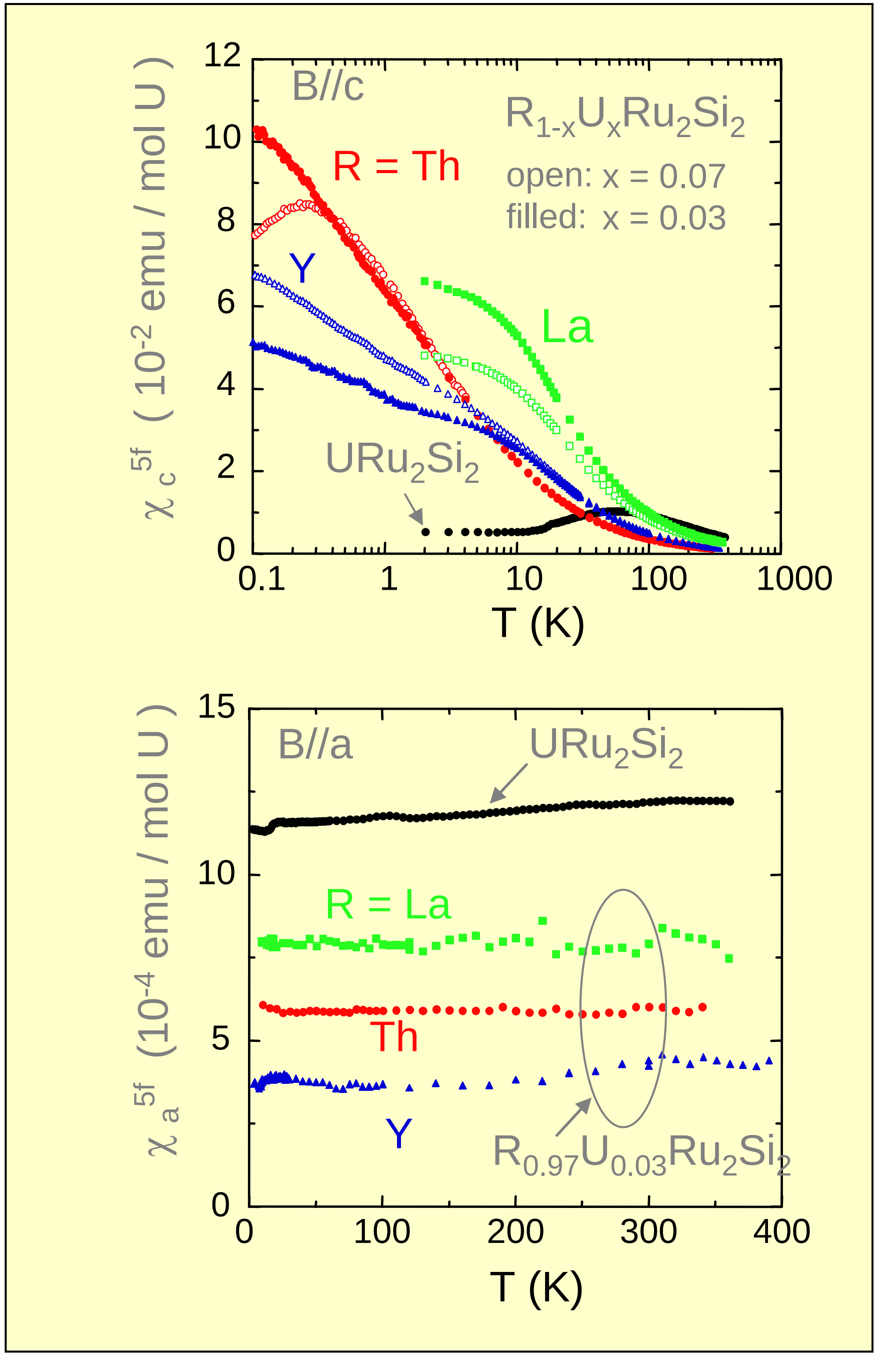
M. Yokoyama *et al*., J. Phys. Soc. Jpn 73, 545 (2004)

Rh doping: increase of *c/a* ratio + disorder + carrier density

Sensitive chemical tuning parameter of electronic structure;

important for present studies

Dilute-uranium-limit properties of URu_2Si_2



Amitsuka and Sakakibara (1994)

➔ Strong uniaxial magnetic anisotropy

➔ $\chi_c \rightarrow \infty$ ($T \rightarrow 0$) in the dilute U limit

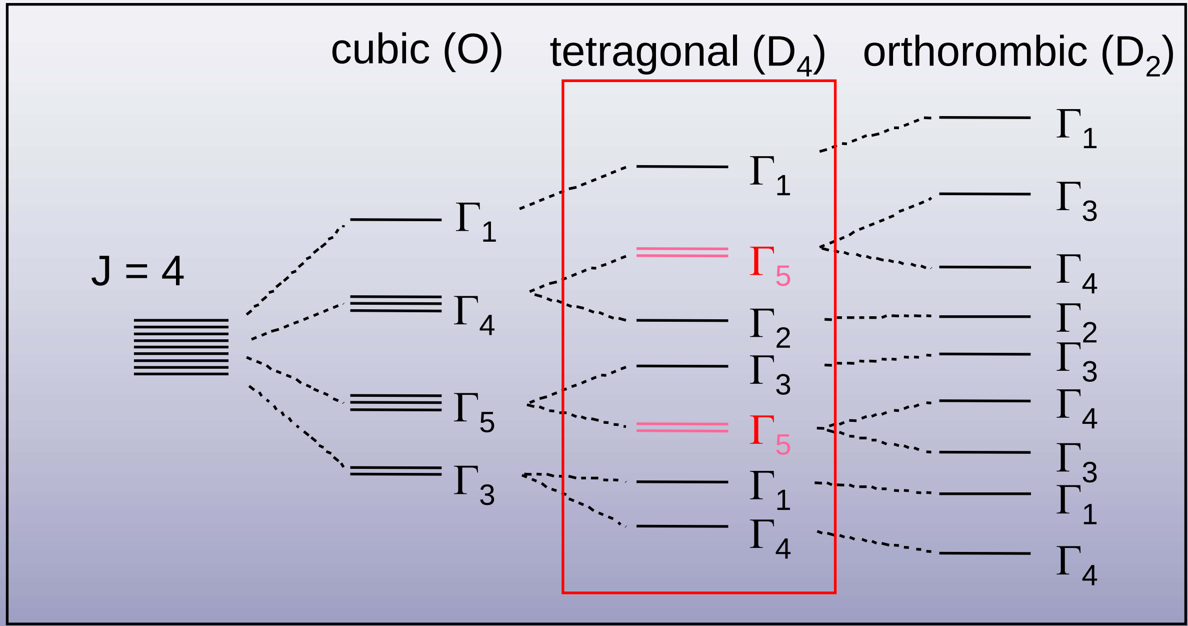
➔ Heavy electron state at low T and NFL behavior in χ_c

(interpreted as the two-channel Kondo effect)

Quite consistent with Γ_5 CEF ground states (non-Kramers doublets)

Γ_5 Crystal Electric Field Doublets of U^{4+} ($5f^2$)

$5f^2$ $^3H_4$ (S=1, L=5, and J=4)



Several experiments suggest that $U(Ru,Rh)_2Si_2$ has Γ_5 CEF ground states of U^{4+} ($5f^2$)

$$\left|\Gamma_5 \pm\right\rangle = \cos\alpha\left|\pm 3\right\rangle + \sin\alpha\left|\mp 1\right\rangle = \begin{cases} \left|\uparrow\right\rangle \\ \left|\downarrow\right\rangle \end{cases}$$

Γ_5 non-Kramers doublets

Γ_5 explains Ising anisotropy since g-factors vanish in basal plane

$$|\Gamma_5 \pm\rangle = \cos\alpha\,|\pm 3\rangle + \sin\alpha\,|\mp 1\rangle = \begin{cases} |\uparrow\rangle \\ |\downarrow\rangle \end{cases}$$

Γ_5 possesses spin and quadrupole moments for which quadrupole moments modeled using $S_{x,y}$ pseudospin moment [Ohkawa 1999, Amitsuka, Sakakibara (1994)]

$$J_z \longrightarrow \begin{array}{cc} |\uparrow\rangle & |\downarrow\rangle \end{array} \begin{pmatrix} +g_z & \\ & -g_z \end{pmatrix}$$

$$\boxed{H_{Zeeman} = -\frac{1}{2}\sum_i \sum_{\lambda=x,y,z} g_\lambda \mu_B H_\lambda S_{i\lambda}}$$

$g_z = g_J\,(3\cos^2\alpha - \sin^2\alpha) = 2.6$

$$J_{x,y} \longrightarrow 0$$

$g_x = g_y = 0$

U^{4+} : $5f^{\,2}$

$$|\Gamma_5 \pm\rangle = \cos\alpha\,|\pm 3\rangle + \sin\alpha\,|\mp 1\rangle = \begin{cases} |\uparrow\rangle \\ |\downarrow\rangle \end{cases}$$

$$O_z = J_z \longrightarrow \begin{array}{cc} |\uparrow\rangle & |\downarrow\rangle \end{array} \begin{pmatrix} m_z & \\ & -m_z \end{pmatrix} \propto S_z$$

$$O_{x2\text{-}y2} = \frac{1}{2}(J_+^2 + J_-^2) \longrightarrow \begin{pmatrix} & q_1 \\ q_1 & \end{pmatrix} \propto S_x$$

$$O_{xy} = \frac{-i}{2}(J_+^2 - J_-^2) \longrightarrow \begin{pmatrix} & -iq_2 \\ iq_2 & \end{pmatrix} \propto S_y$$

$S = \frac{1}{2}$ pseudo-spin

$g_x = g_y = 0$

$g_z \neq 0$

jm_j multipole expansion of H_{CEF}

$$H_{multi} = -\frac{1}{2}\Sigma\,\Sigma\,A_{\Gamma;\,ij}\,O_{\Gamma;\,i}O_{\Gamma;\,j}$$

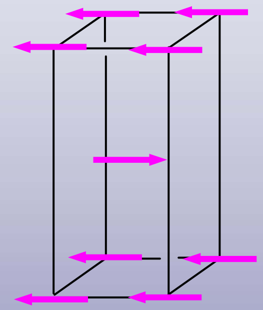
$$\Downarrow$$

$$H_{spin} = -\frac{1}{2}\sum_{\lambda=x,y,z}\sum_{i,j} J_{\lambda;\,ij}\,S_{\lambda;\,i}S_{\lambda;\,j}$$

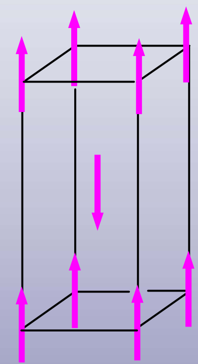
Γ_5 antiferro-quadrupolar ordering model for HO

$$H_{spin} = -\frac{1}{2}\sum_{\lambda=x,y,z}\sum_{i,j} J_{\lambda;\,ij}\, S_{\lambda;\,i} S_{\lambda;\,j}$$

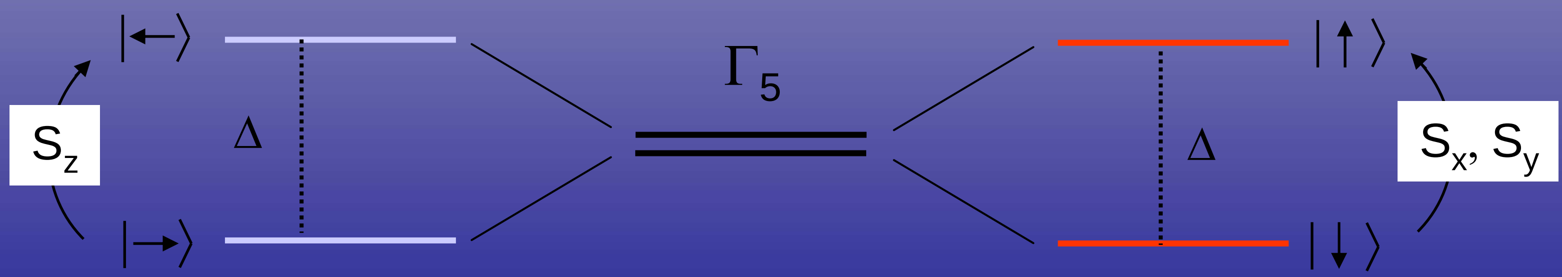
XY	Ising
$(\lvert J_x \rvert = \lvert J_y \rvert > \lvert J_z \rvert)$	$(\lvert J_x \rvert = \lvert J_y \rvert < \lvert J_z \rvert)$
$\sqrt{\langle S_x \rangle^2 + \langle S_y \rangle^2} \neq 0$	$\langle S_z \rangle \neq 0$
$\langle S_z \rangle = 0$	



quadrupole order



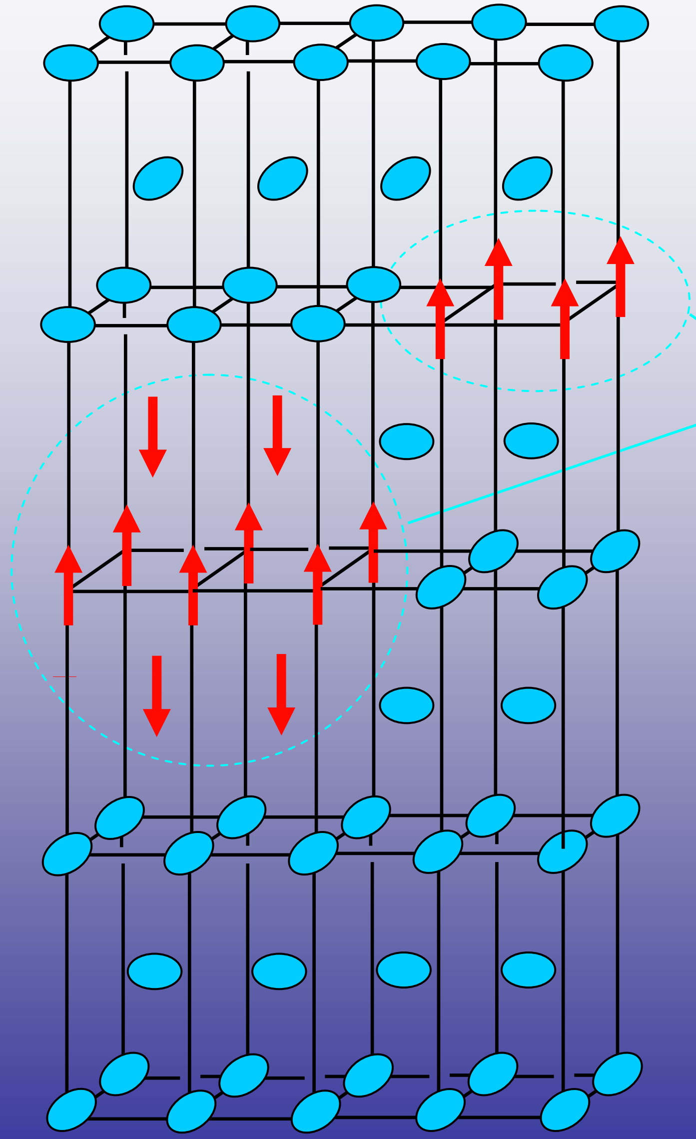
magnetic order



detectable by NS

Tunable through pressure, magnetic fields etc

Γ_5 quadrupolar ordering model for HO



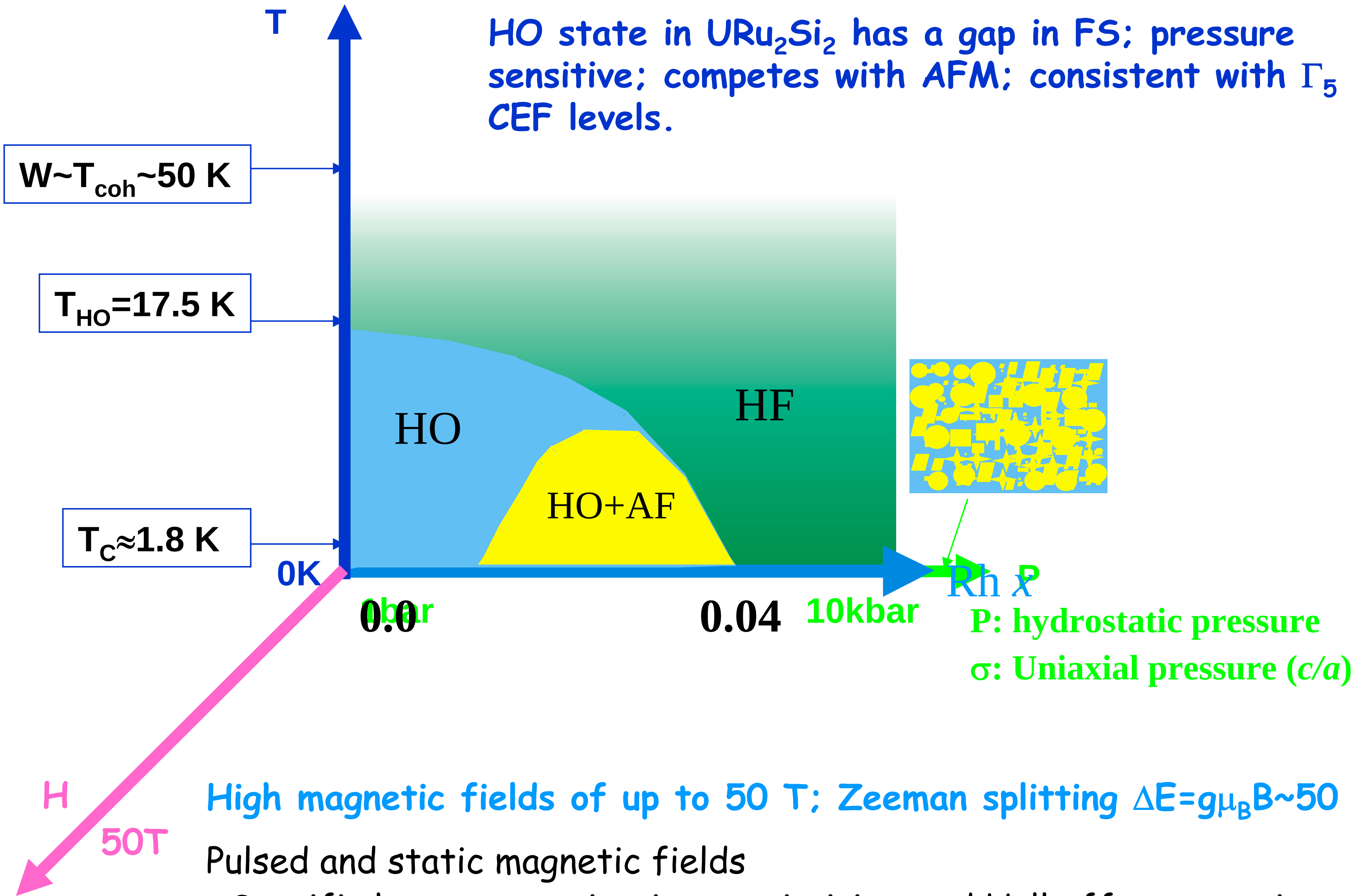
AF and ferro. clusters
serving as domain walls
of quadrupole order

weak dipolar internal fields

AF Quadrupole scenario for HO is consistent with the fact that HO is sensitive to pressure and strain

URu_2Si_2, Key Energy Scales

Key energy scales at H=0 T, P=1 atm



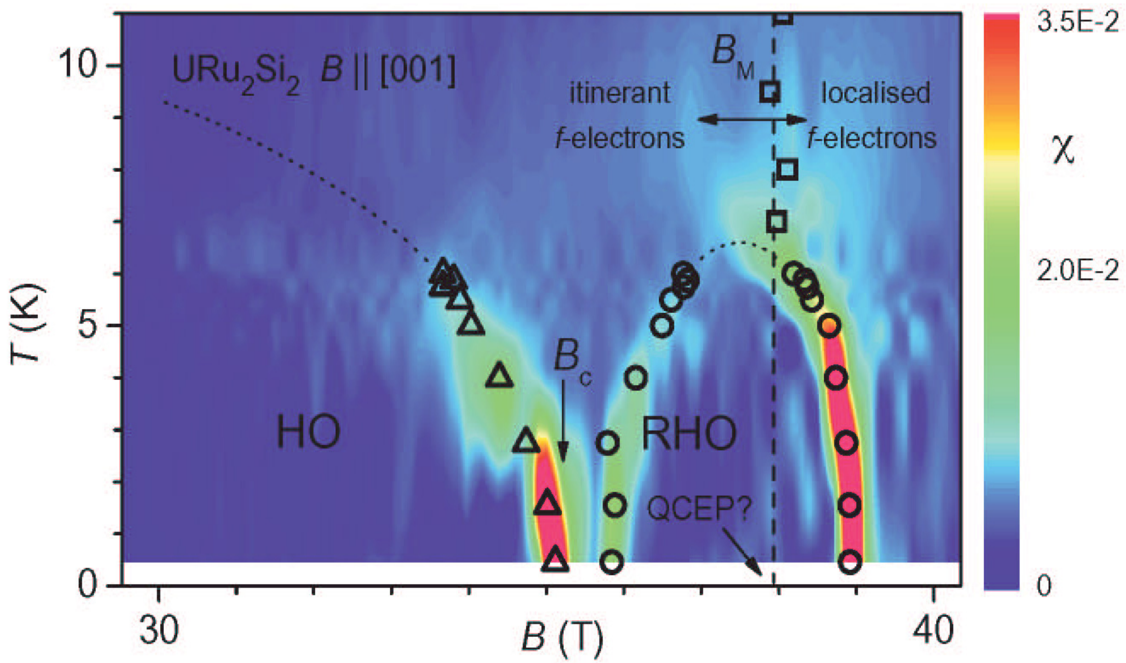
High magnetic fields of up to 50 T; Zeeman splitting $\Delta E = g\mu_B B \sim 50$ K

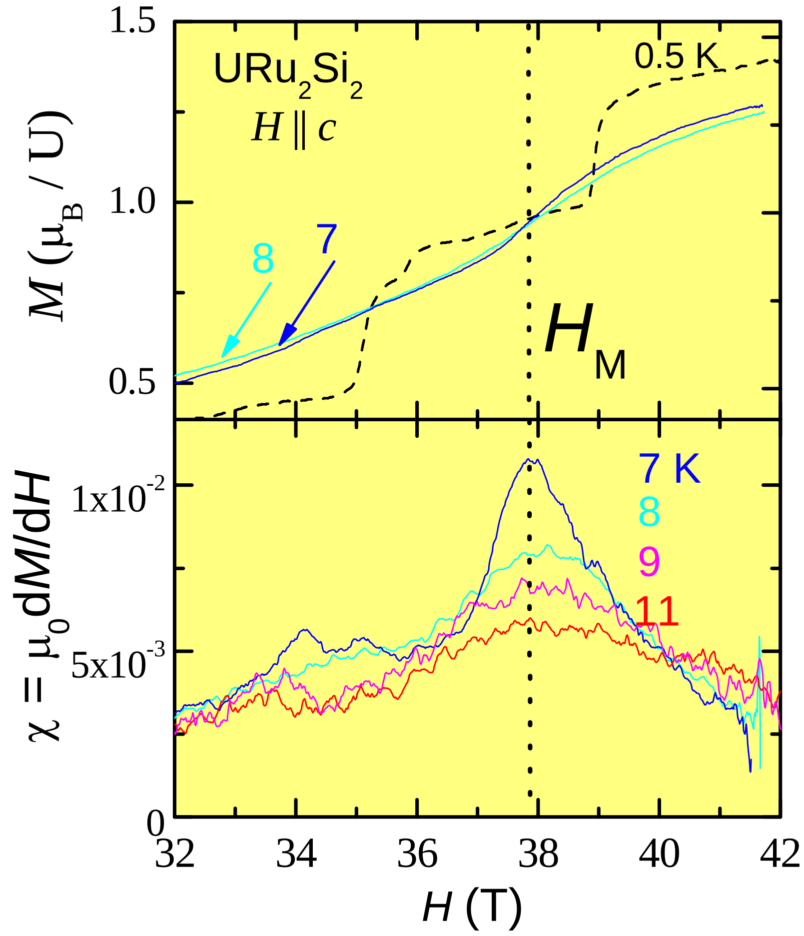
Pulsed and static magnetic fields

: Specific heat, magnetization, resistivity, and Hall effect experiments.

Metamagnetic Transition above Phase III in URu_2Si_2

N. Harrison *et al*, PRL90, 06402 (2003)





χ=dM/dB confirm the new phase (phase III) boundary observed through heat capacity study.

M. Jaime, K. H. Kim *et al*., PRL89, 287201 (2002);
J. S. Kim *et al*., PRB 67, 014404 (2003)

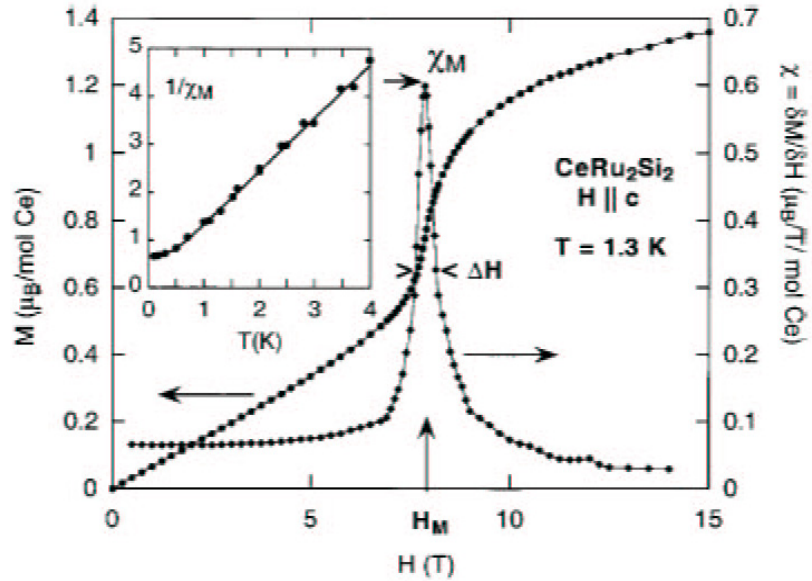
New surprise:

Metamagnetism above phase III, similar to those of $CeRu_2Si_2$ and $Sr_3Ru_2O_7$ → Possibility of metamagnetic quantum criticality?

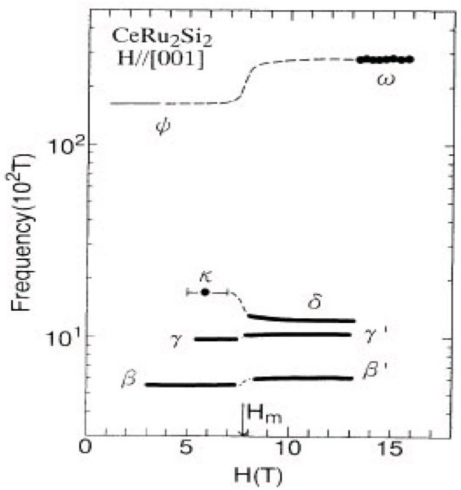
Heavy Fermion Metamagnetism : e.g. $CeRu_2Si_2$

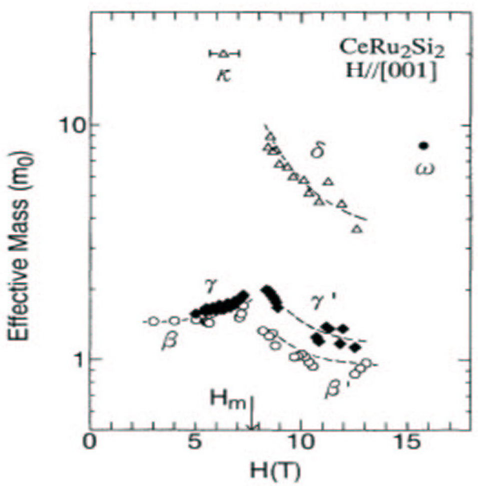
Magnetization, susceptibility and transport resemble that seen in $Sr_3Ru_2O_7$,[S. Grigera et al. Science 294, 329(2001)]

Maximum in γ (Sommerfeld coefficient) was also observed.



[J. Flouquet et al. Physica B 319, 251(2002)]

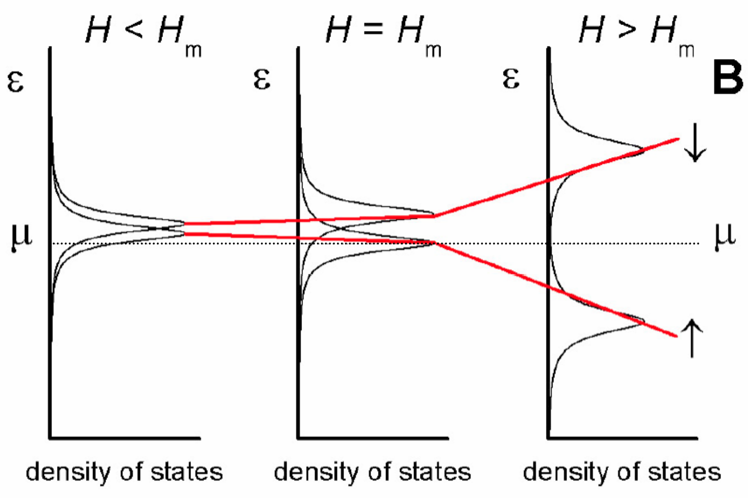




Effective mass undergoes divergent behaviour
[H. Aoki et al, PRL 1993]

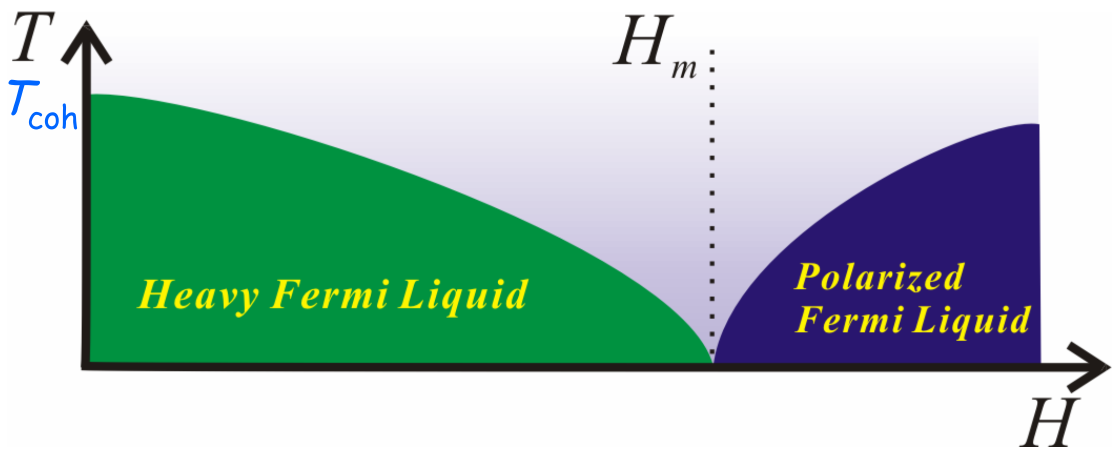
Generic Picture of Metamagnetic QC in Heavy Fermions

[Edwards and Green, Z.Phys.B103,243(1997)]



Hybridized band model

[A. V. Alehandro et al., PRL 2005]



itinerant *f* localized *f* ?

Fermi surface change

Zeeman splitting $\Delta\varepsilon > W$ (*f*-electron bandwidth): entire spin component of *f*-band can be depopulated, causing virtual *f*-electron localization

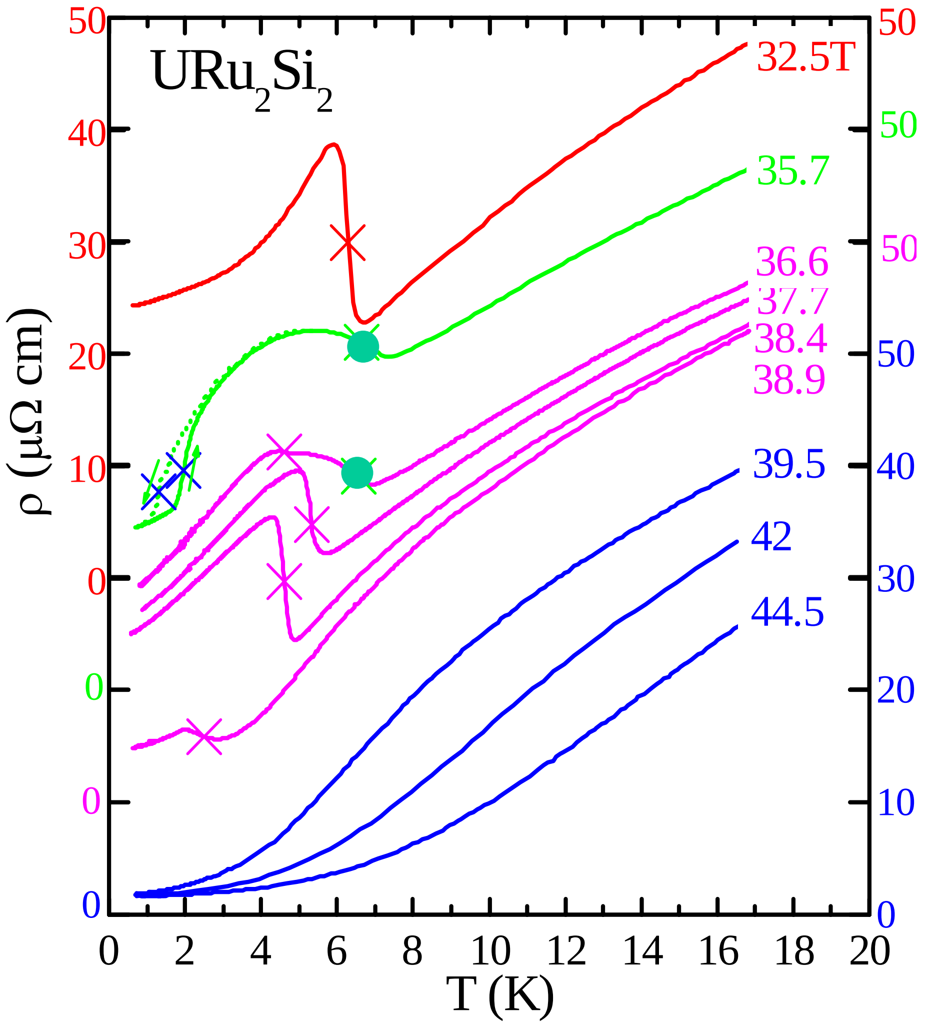
(1) System changes from HFL to PFL states forming a QCP?: continuous or discontinuous? A symmetry-breaking order parameter ?

(2) Fermi liquid scale vanishes as $H \rightarrow H_m$ on both sides of H_m

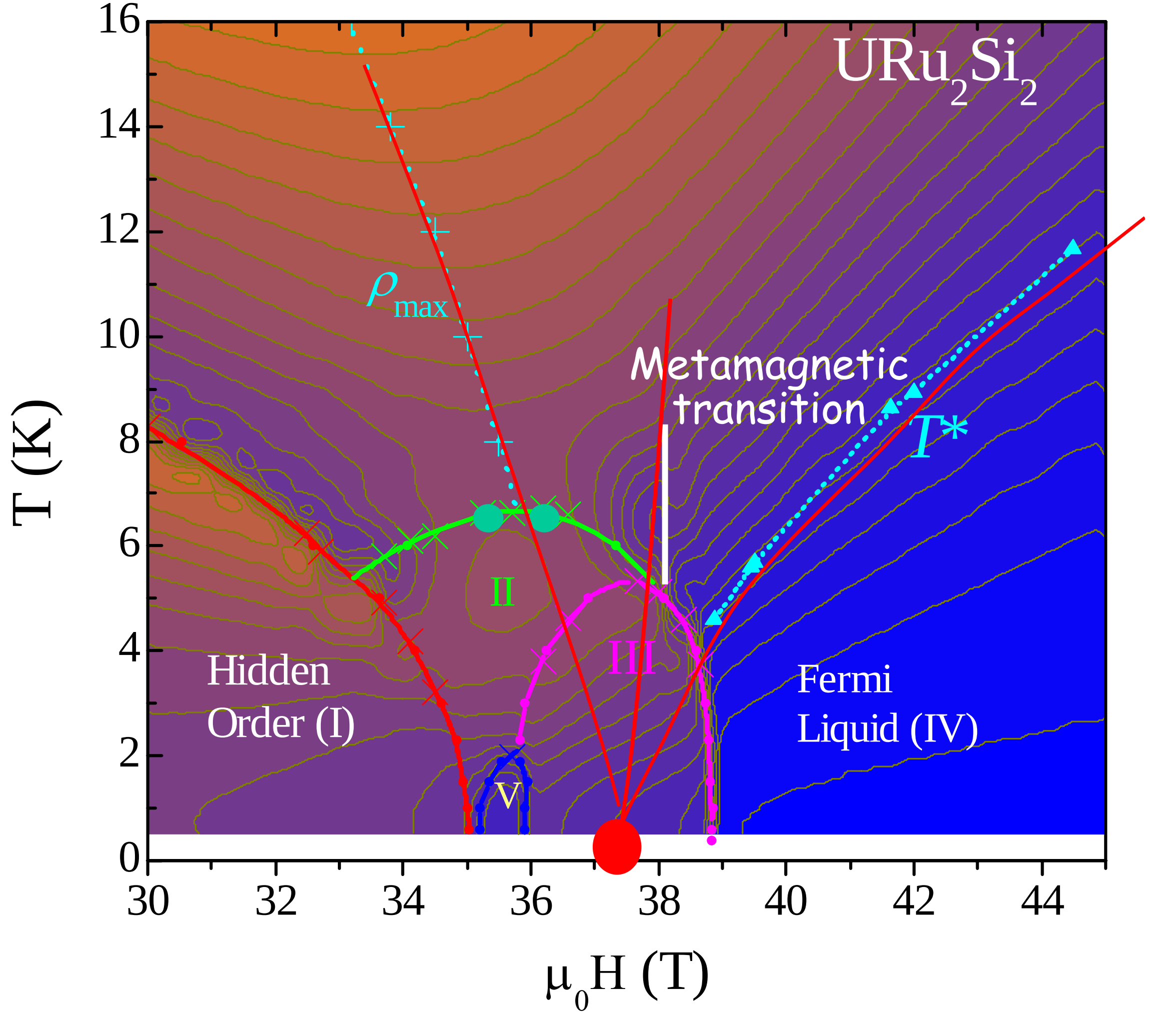
Metamagnetic quantum criticality in URu_2Si_2?

→Motivated us to perform careful transport studies!!

Multiple phases formed near the putative QCP in URu_2Si_2



Resistivity intensity plot



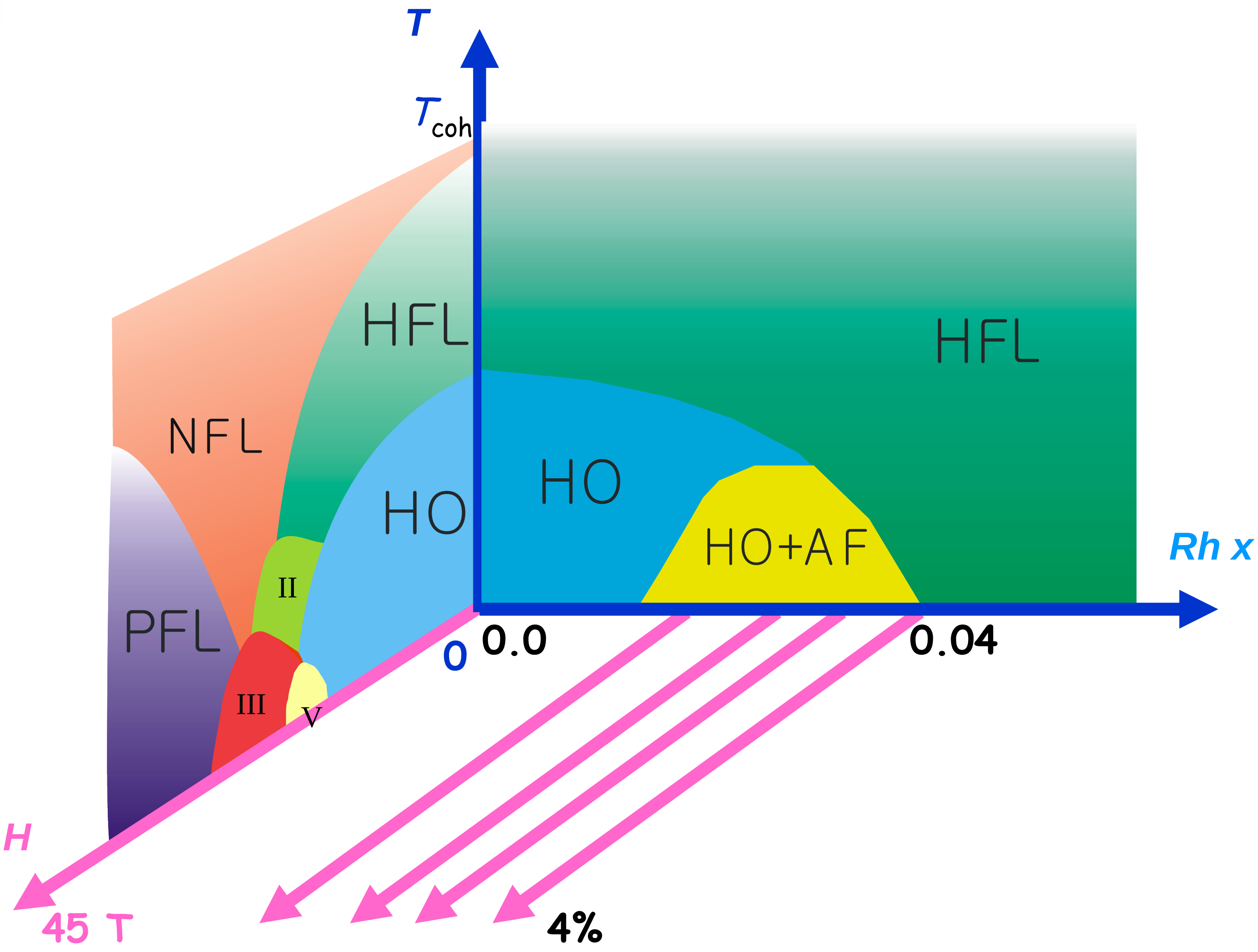
[K. H. Kim et al. PRL 2002]

(1) Multiple new phases (phase II & phase V), were found.

(2) Highly hysteretic 1st order phase boundaries in (HO, II, III, V)

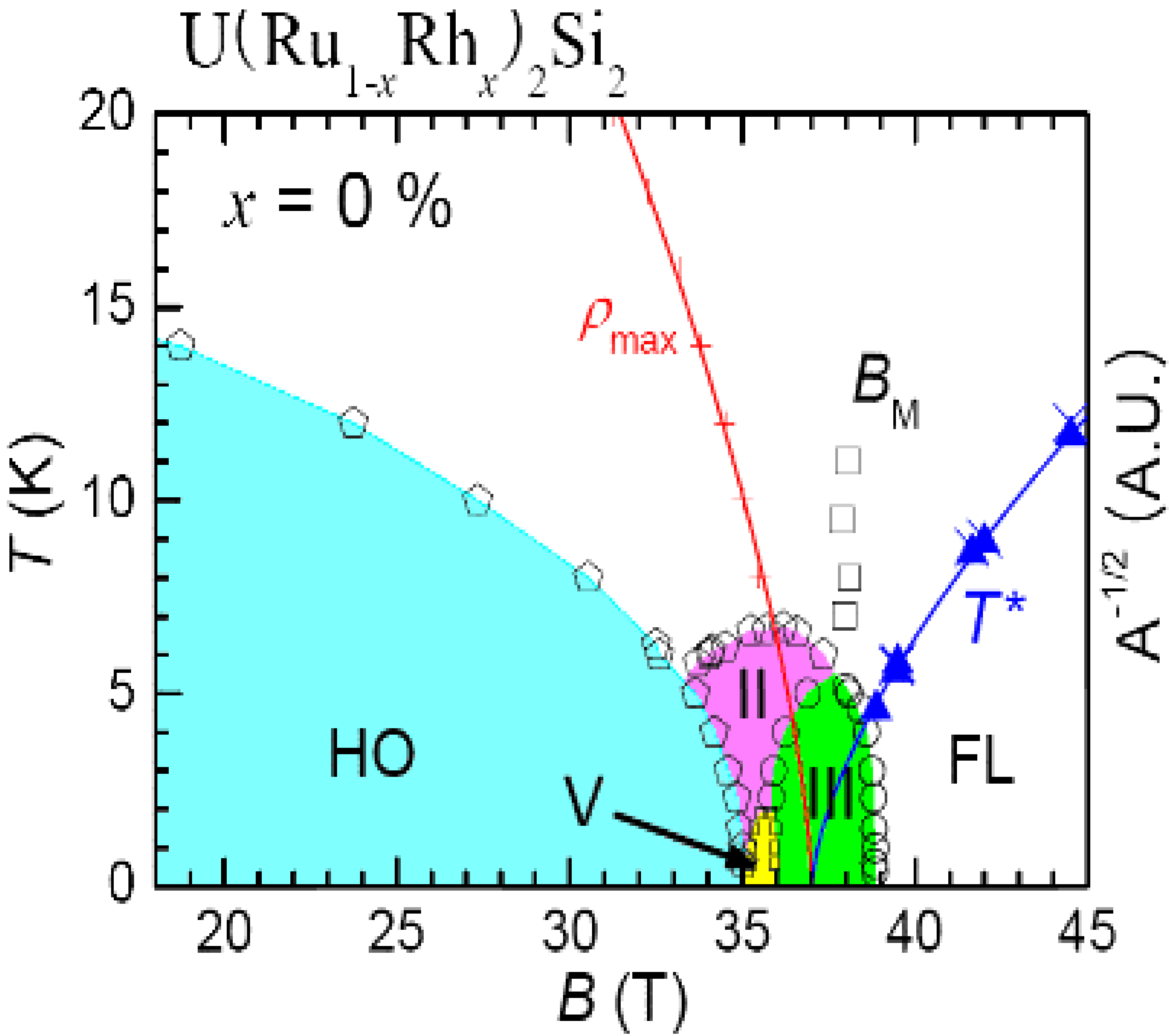
(3) Suggests the hidden metamagnetic QCP; but identification is not easy.

Motivation to $U(Ru_{1-x}Rh_x)_2Si_2$



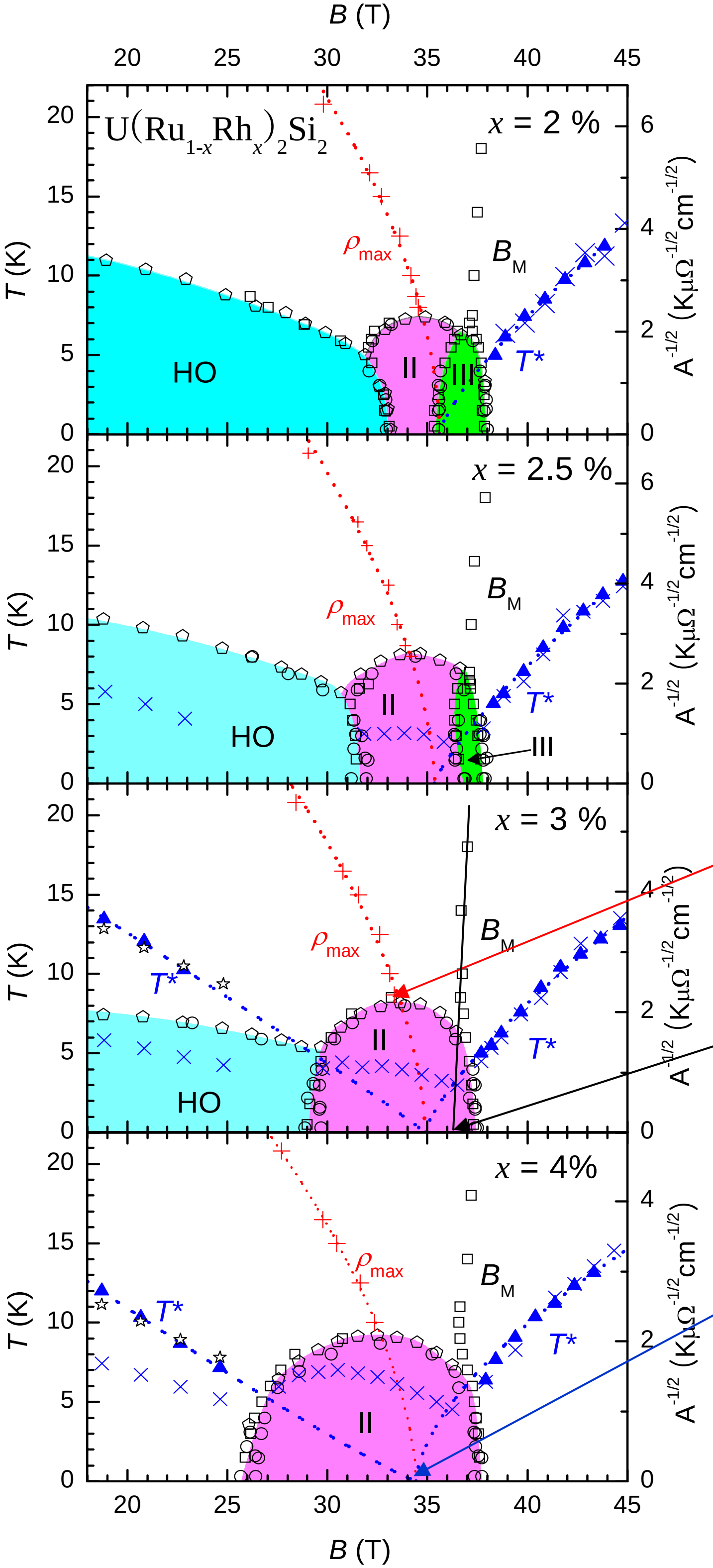
CAN THE COMPLEX PHASE FORMATION CAN BE SIMPLIFIED?

Is the creation of new phases linked to the metamagnetic QCP?



U(Ru1-xRhx)2Si2
x = 0 %
ρmax
BM
T*
HO
II
III
V
FL
T (K)
B (T)
A-1/2 (A.U.)
0
5
10
15
20
25
30
35
40
45

Phase Diagram of $U(Ru,Rh)_2Si_2$



B_{II} Center of phase II

B_M Metamagnetic transition field

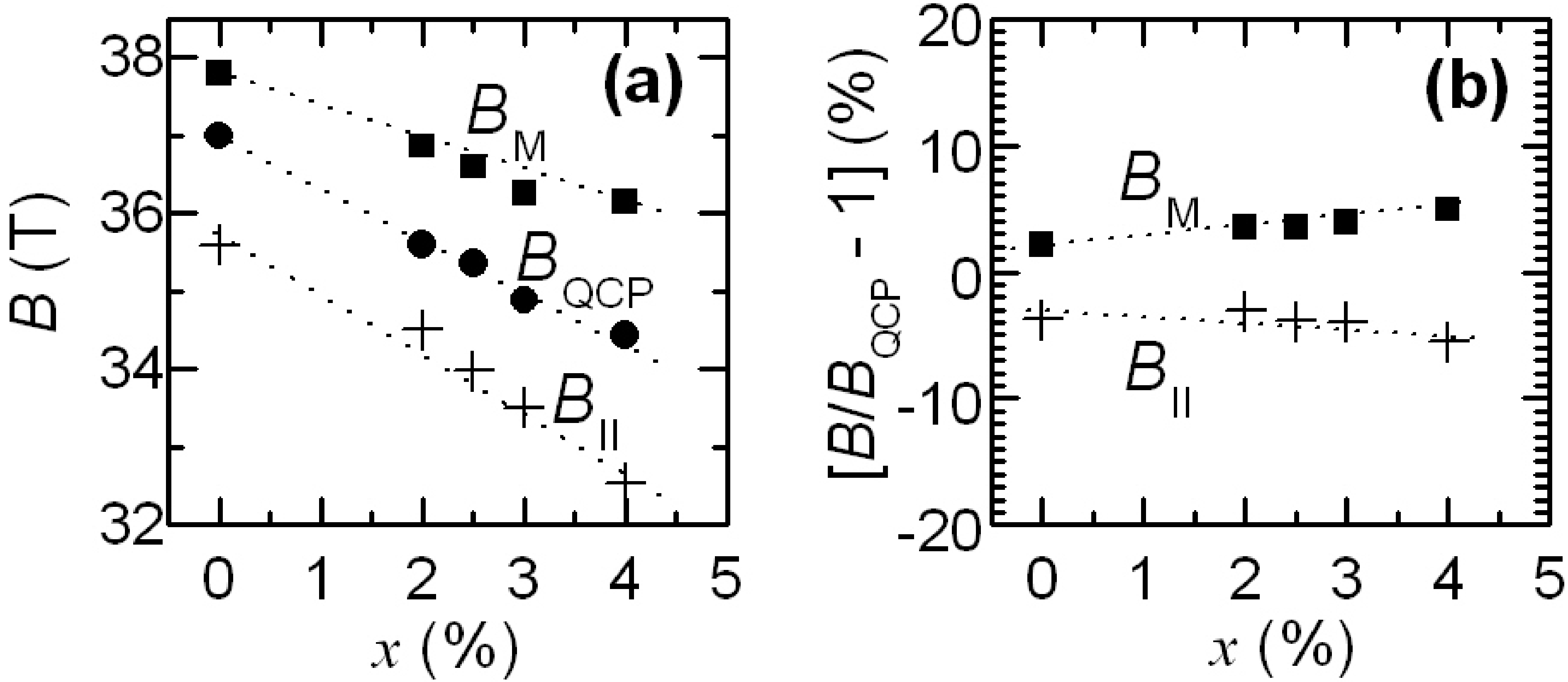
B_{QCP} Putative QCP from ρ_{max}, T^*, $A^{-1/2}$

K. H. Kim et al. PRL 93, 126404 (2004)

Nexus among B_M , B_{II} , and B_{QCP}

$U(Ru_{1-x}Rh_x)_2Si_2$

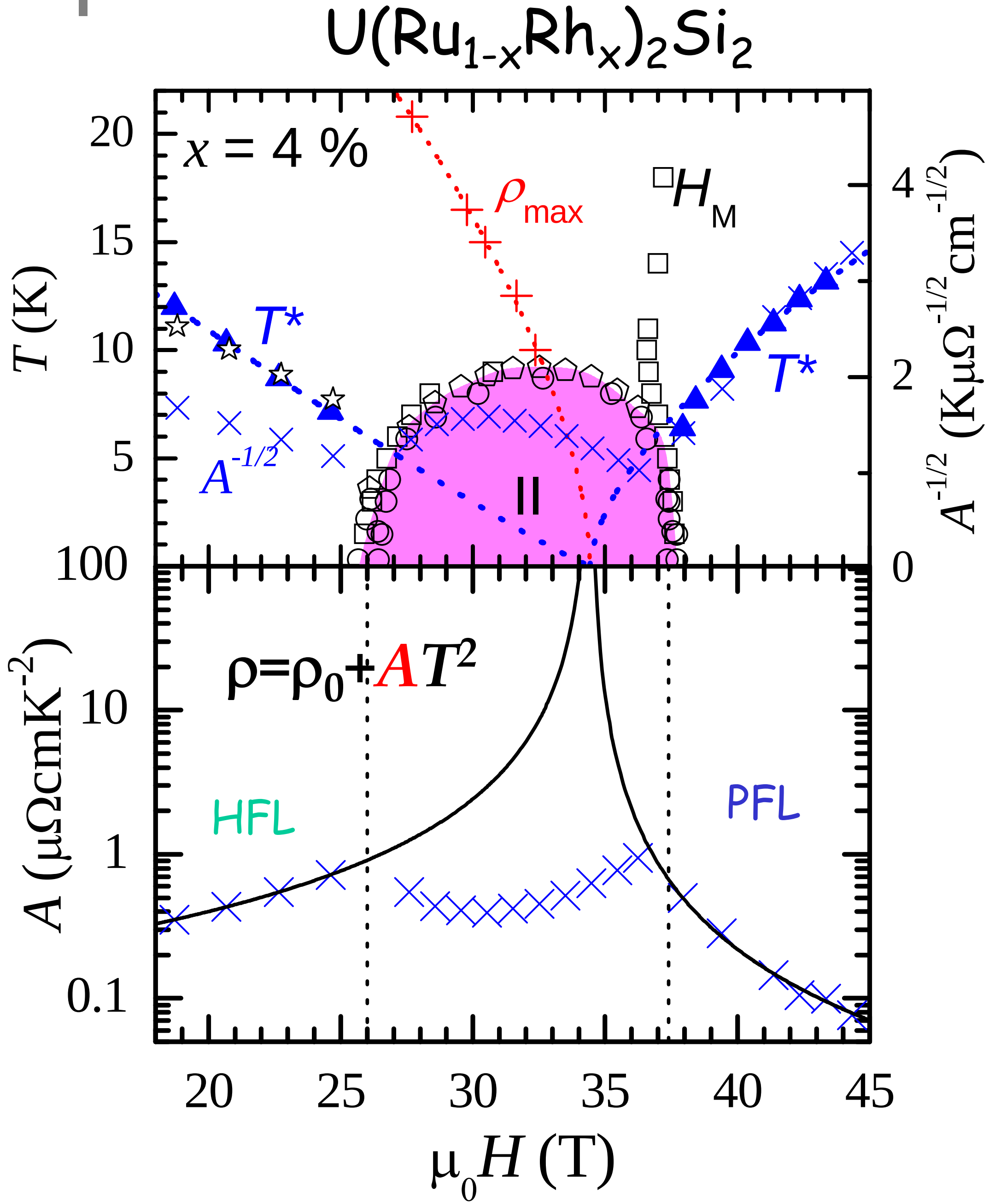
K. H. Kim *et al*. PRL 93, 126404 (2004)



A clear example showing a link between B_{II} (effective center of phases) and B_{QCP} !!

Avoid effective mass divergence by forming a phase

K. H. Kim *et al*. PRL 93, 126404 (2004)



T^* crossover temperature from T^2 (FL) to NFL (dρ/dT|$_{max}$)

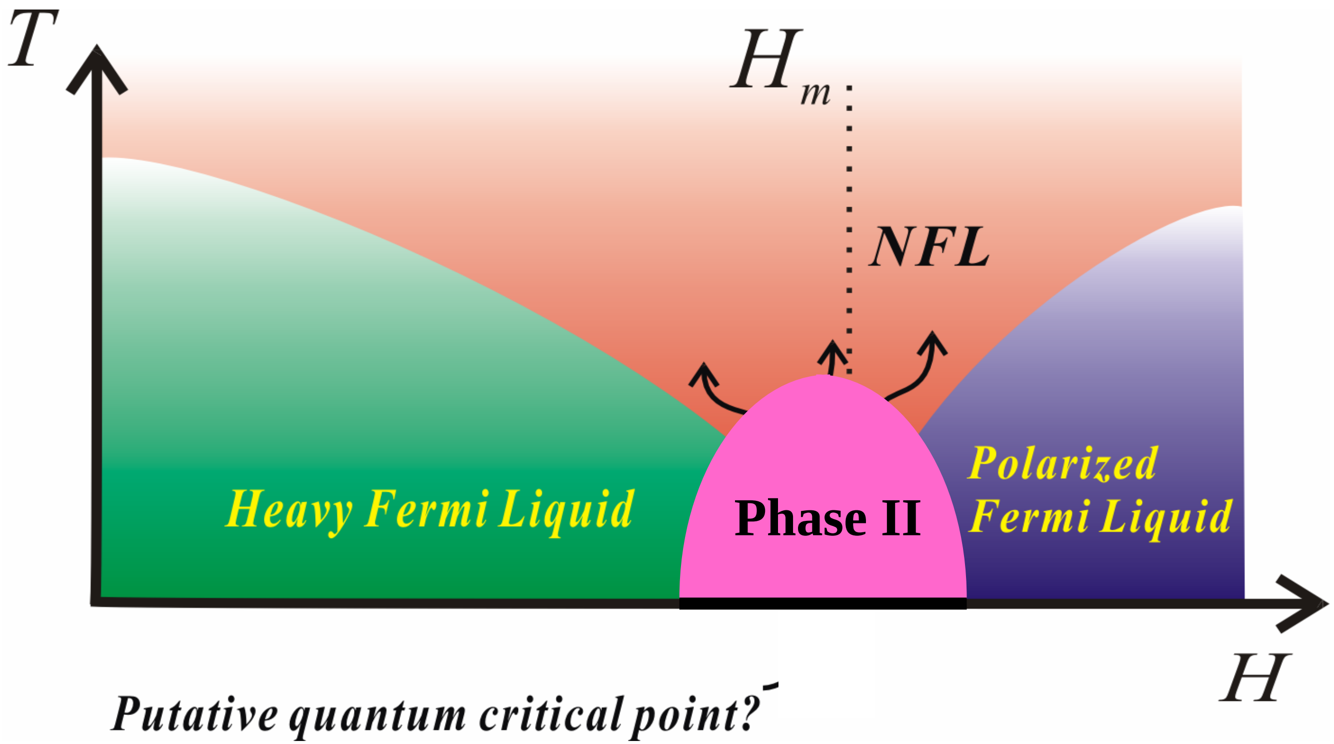
Collapse of T^*, $A^{-1/2}$ with the same scaling $|H\text{-}H_{QCP}|^{\alpha 1}$

ρ_{max} scales with $|H\text{-}H_{QCP}|^{\alpha 2}$

$T^* \propto A^{-1/2}$ ($\propto 1/D(\varepsilon_F) \propto \varepsilon_F \propto 1/m^*$)

Quasiparticle instability (i.e. large density of states, heavy electron mass) is avoided inside phase II.

$U(Ru_{0.96}Rh_{0.04})_2Si_2$: A new example of a single phase formation at the metamagnetic QCP



Two Fermi liquids: one paramagnetic and one with polarized *f*-electrons

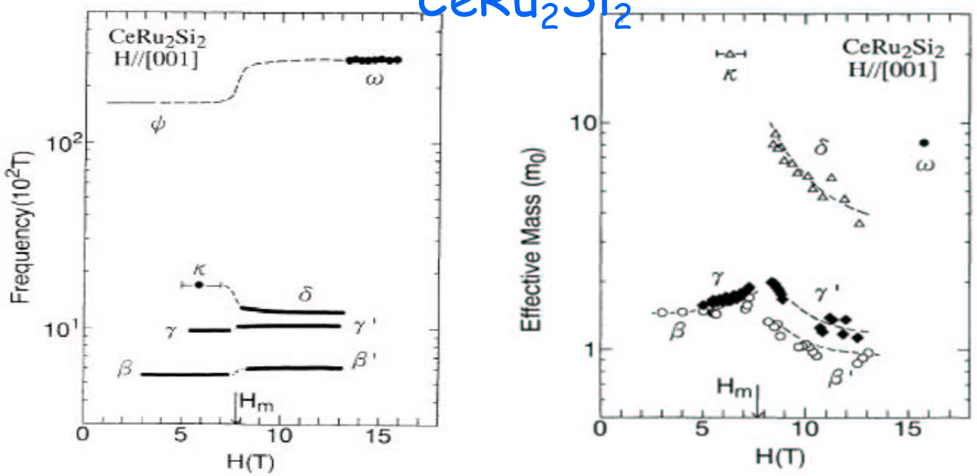
New phase forms so as to avoid quasiparticle instability (i.e. large density of states, mass divergence, fluctuations)

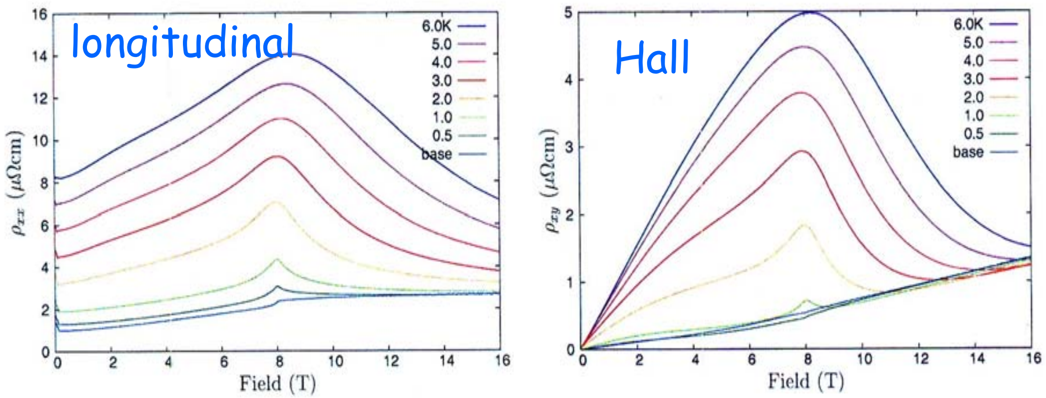
Extreme complexity results when other phases (such as HO) come within close proximity

Analagous to phase formation (i.e. superconductivity) as a function of pressure or doping, where new phase conceals QCP.

Motivation to Hall Effect Studies

$CeRu_2Si_2$



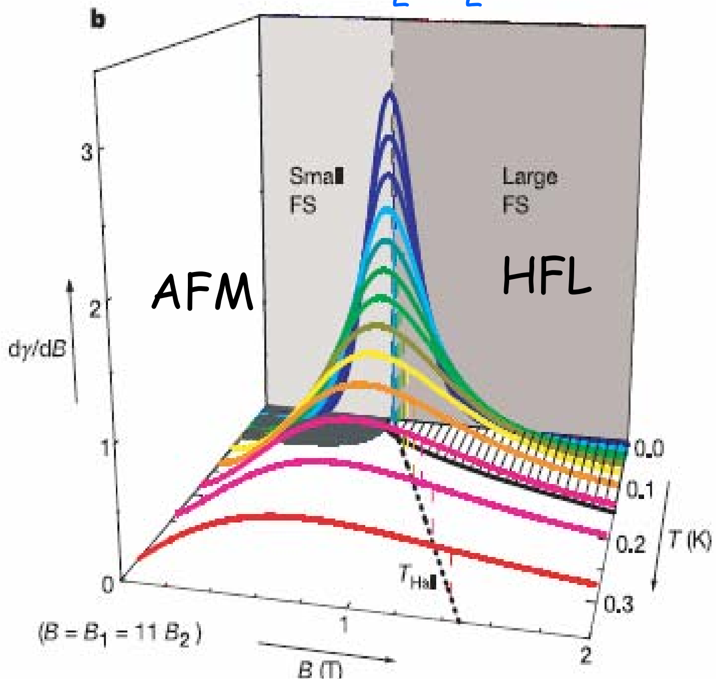


FS change discontinuously
[H. Aoki et al, PRL 1993]

vs.

FS change continuously
[R. Daou et al., PRL 2006]

$YbRh_2Si_2$



A jump in $\dfrac{d\rho_{xy}}{dB}$ at the QCP

from AFM to heavy Fermi liquid ground state.

Consistent with the local criticality picture.

[S. Paschen et al, Nature 432, 881 (2006)]

Hall Resistivity: $U(Ru_{1-x}Rh_x)_2Si_2$, Rh 0 %

[Y. S. Oh et al. to be published]

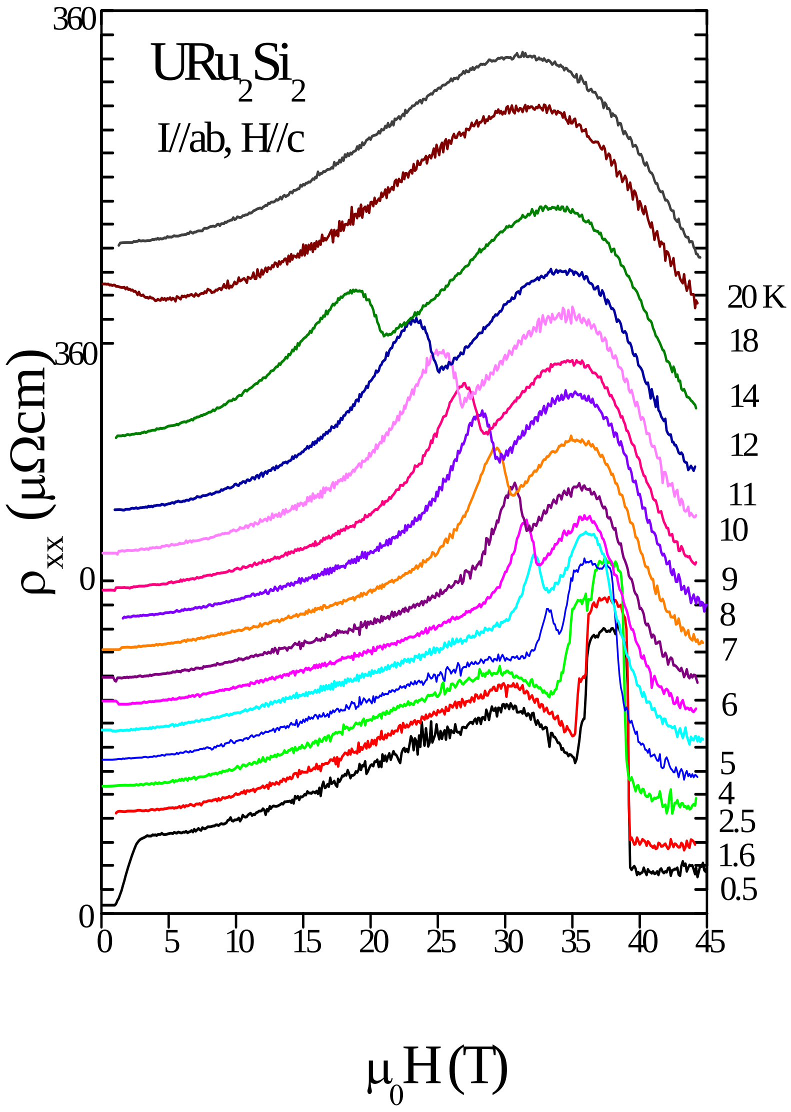
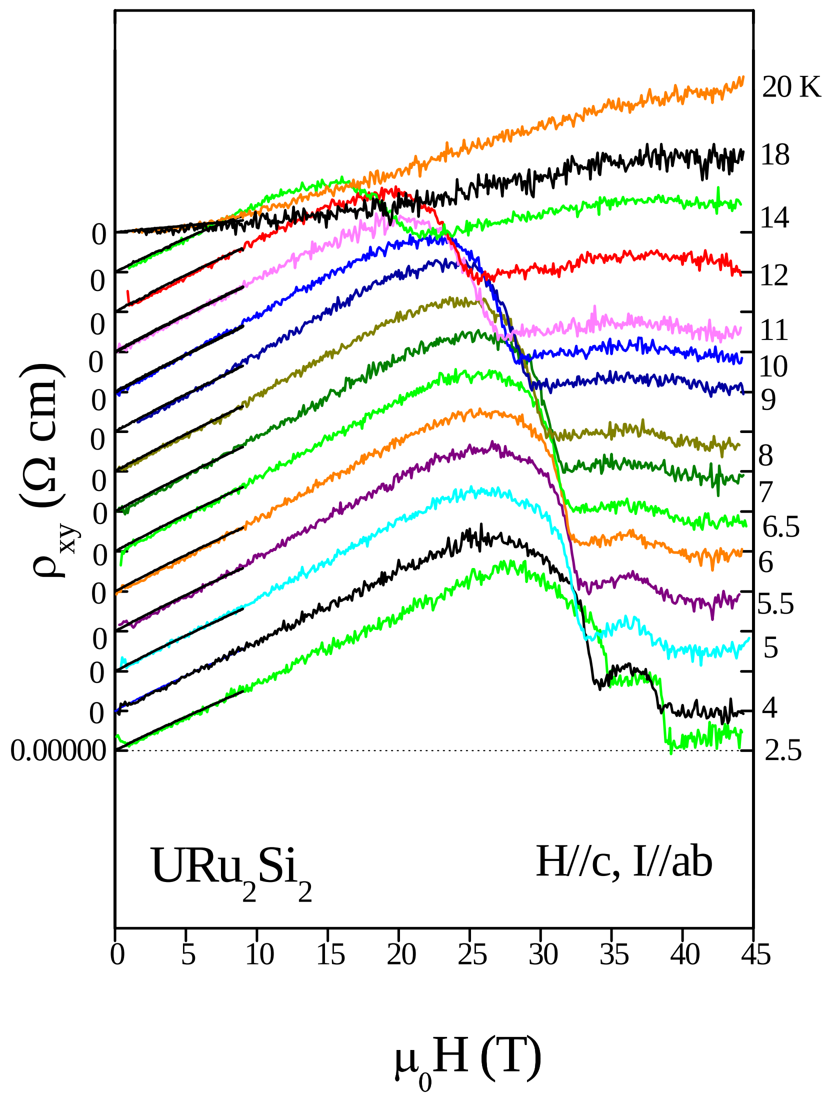


Fig. 1. Kim, Harrison, and Mydosh

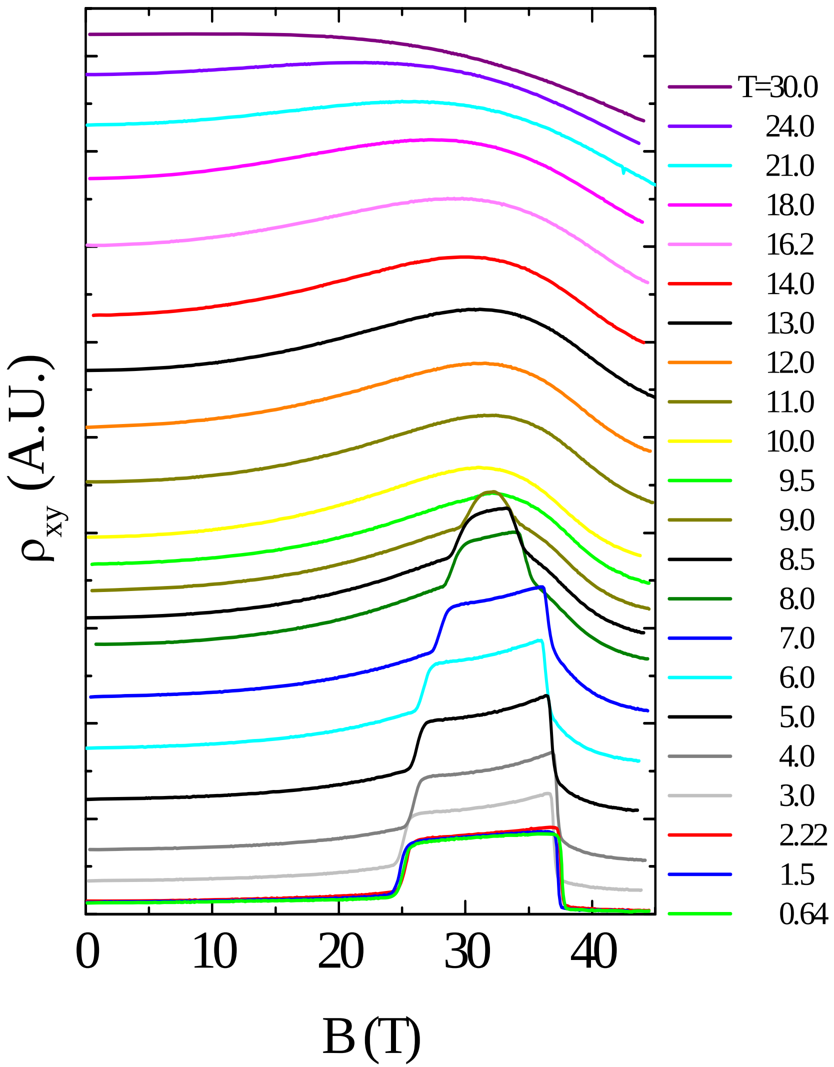


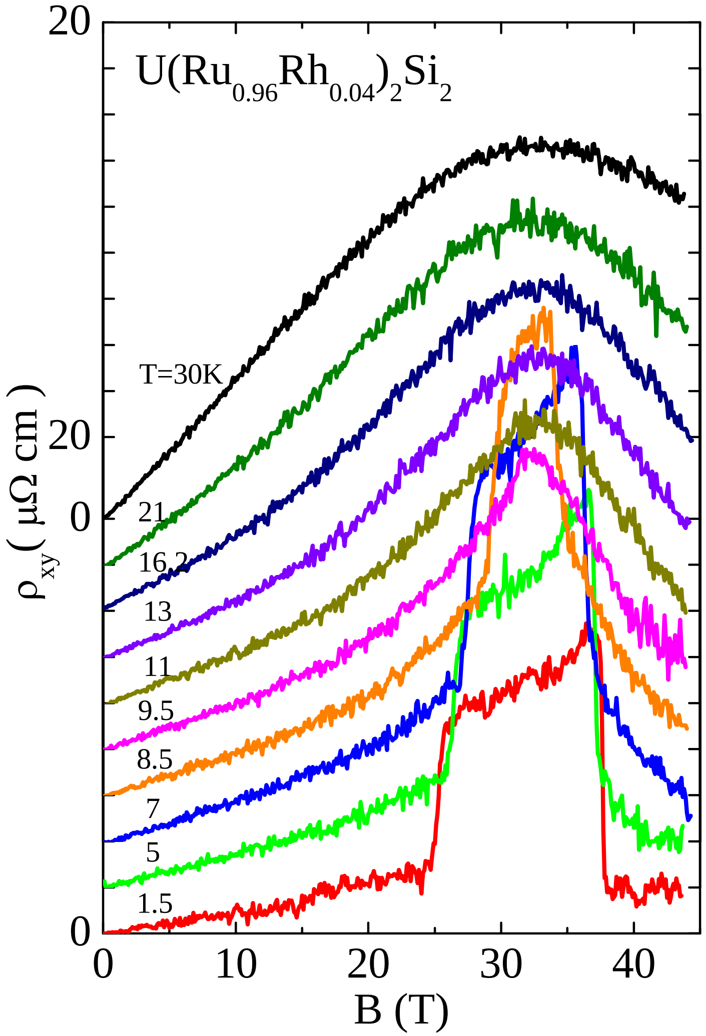
Shape of ρ_{xy} at high temperatures is similar to ρ_{xx}

ρ_{xy} suggestive of large gap in the Fermi surface inside HO

Hall Resistivity: $U(Ru_{1-x}Rh_x)_2Si_2$, 4 %

[Y. S. Oh et al. submitted]



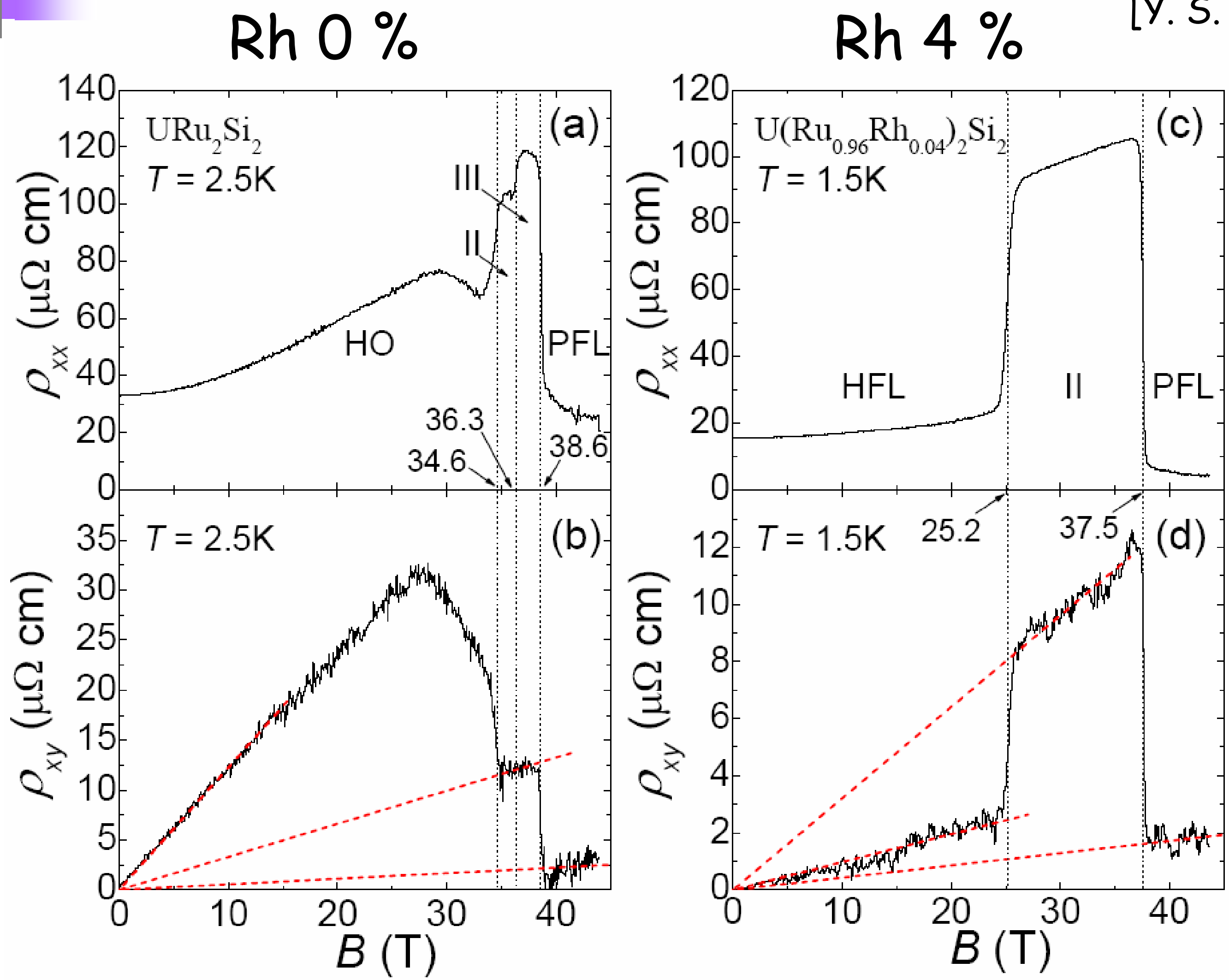


Shape of ρ_{xy} at high temperatures is similar to ρ_{xx}.

ρ_{xy} **suggestive of large gap in the Fermi surface inside phase II**

Key Features of Hall Resistivity: Rh 0 & 4 %

[Y. S. Oh et al. to be published]



$$R_H = \rho_{xy} / B \propto \frac{1}{ne}$$

Large FS gap inside HO & phase II

Three different linear slopes of ρ_{xy} strongly suggest the Fermi surface with different volumes in each phase region.

Information on Fermi Surface for Rh 0 %

H. Ohkuni et al., Phil. Mag. B 79, 1045 (1999)

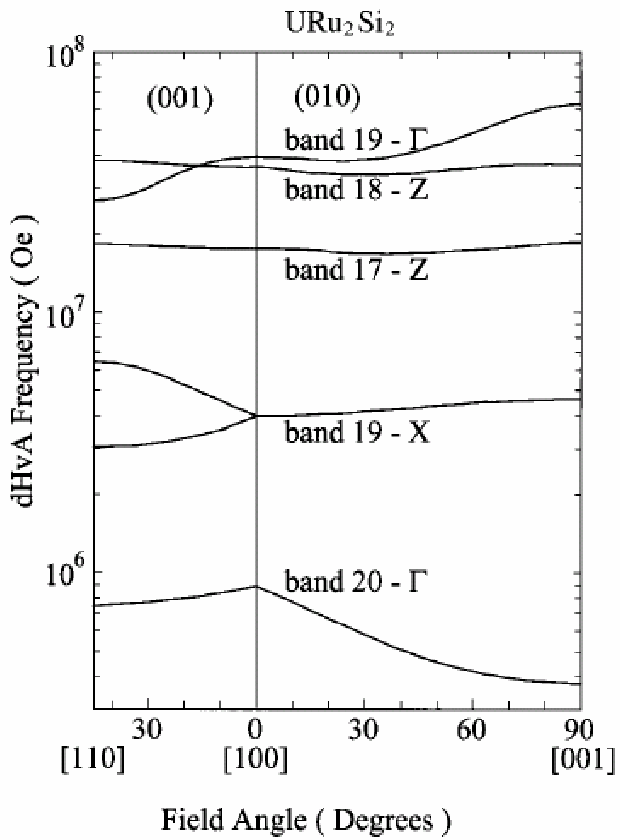


Figure 14. Angular dependence of the theoretical dHvA frequency in the paramagnetic state of URu_2Si_2.

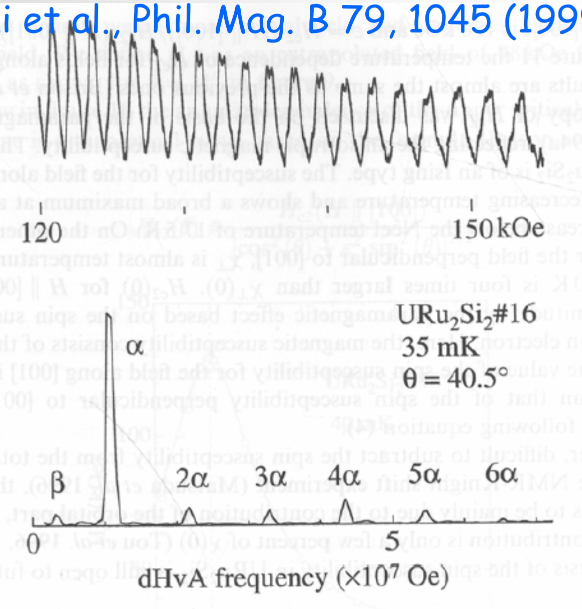


Figure 12. (Continued)

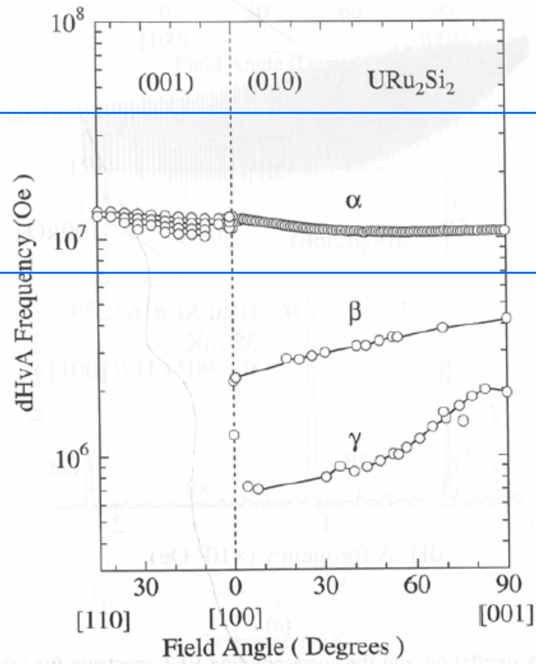


Figure 13. Angular dependence of the dHvA frequency in URu_2Si_2.

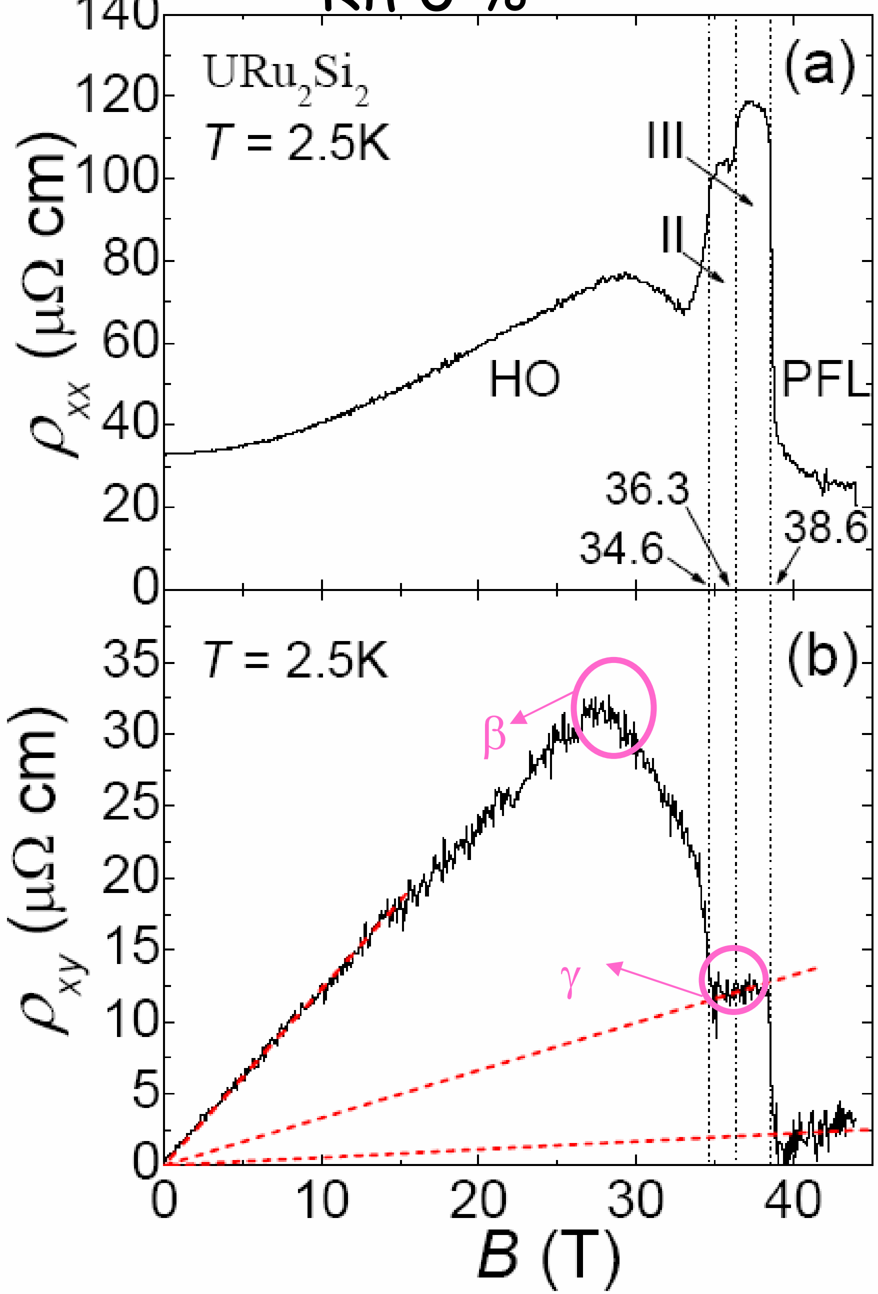
Table 2. De Haas–van Alphen frequency F, the cyclotron effective mass m_c^*, the band mass m_b, the Dingle temperature T_D and the mean free path l in URu_2Si_2.

	Experimental				Theoretical		
$H \parallel [001]$	$F(\times 10^6$ Oe)	$m_c^*(m_0)$	$T_D(K)$	l(Å)		$F(\times 10^6$ Oe)	$m_b(m_0)$
α	10.5	13	0.035	5500	band 17 (Z)	18.5	1.62
					band 18 (Z)	36.8	2.72
					band 19 (Γ)	62.9	8.02
β	4.2	25	0.045	1400	band 19 (X)	4.6	3.20
γ	1.9	8.2	0.11	1200	band 20 (Γ)	0.3	0.25

Experiments observe mostly light hole bands with m*=13 (α), 25 (β), 8.2m$_0$ (γ)

Key Features of Hall Resistivity: Rh 0 & 4 %

Rh 0 %



[Y. S. Oh et al. to be published]

Hall effect is dominated by the most mobile α band.

$$\varepsilon_F = \frac{heF}{m^*} = g_z \sigma \mu_B B_P$$

F is the dHvA frequency of each α, β, and γ band.

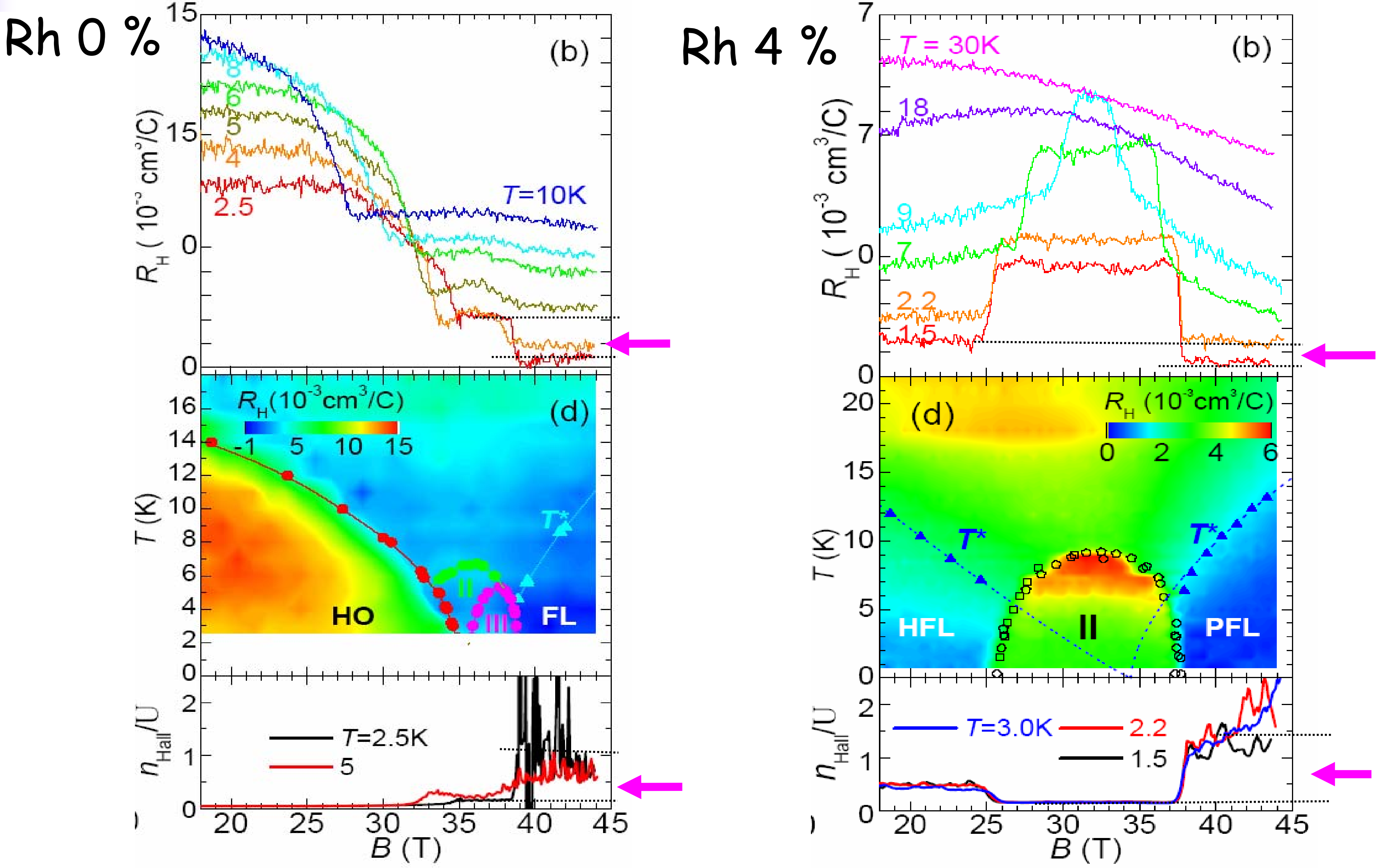
$B_p \approx 120, 25$, and 35 T for the α, β, and γ.

($g_z \approx 2.6$, experimentally determined)

A sudden depopulation of a carrier pocket, will cause the chemical potential to re-equilibriate itself amongst the remaining pockets, leading to changes in their sizes and net observable changes in R_H.

Rapid increase in carriers in the highest mobility α pocket is the primary cause in the destruction of the HO state.

Hall Coefficients: $U(Ru_{1-x}Rh_x)_2Si_2$, Rh 0 and 4 %

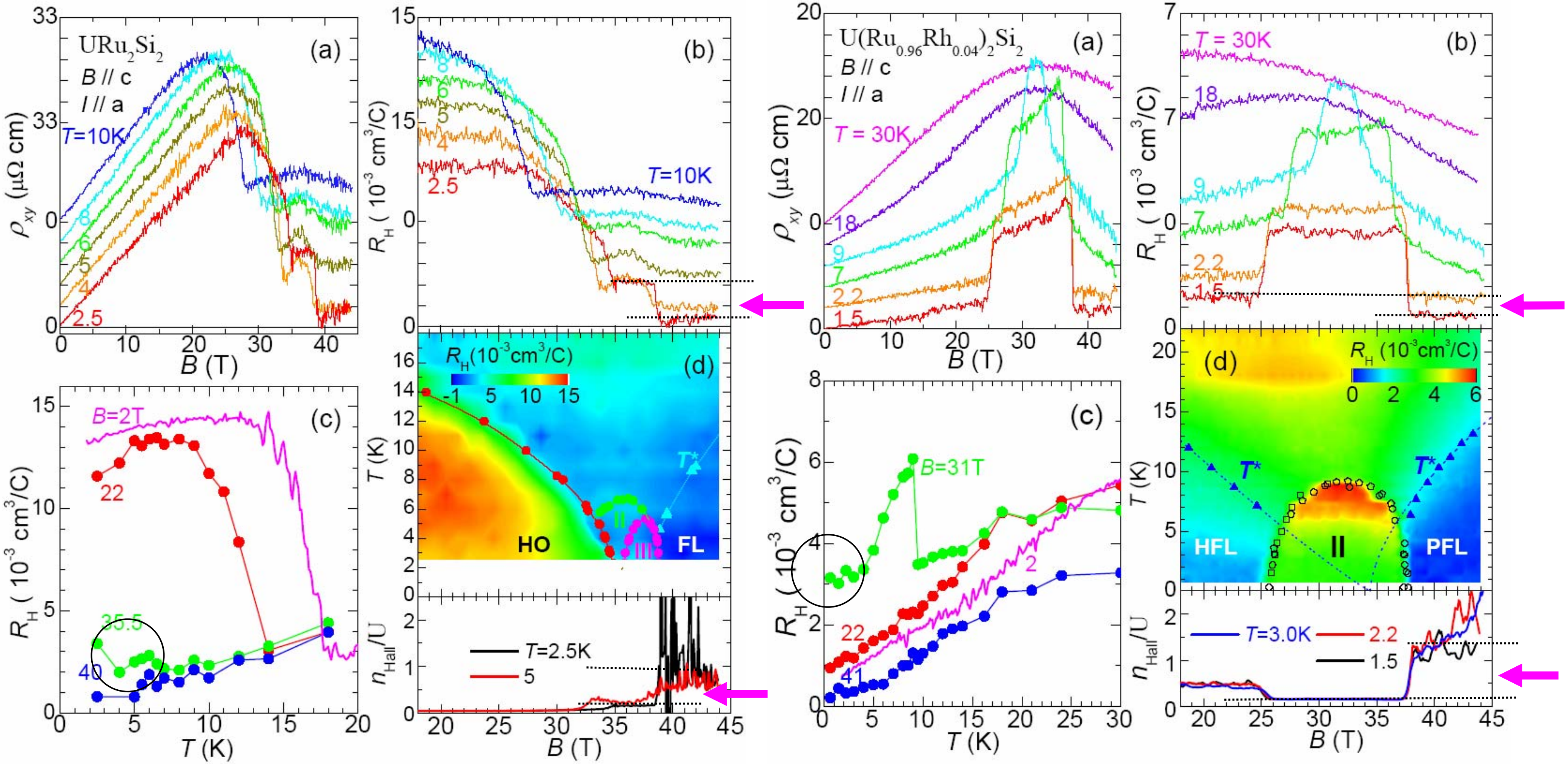


Abrupt R_H change suggests a discontinuous Fermi volume change.

A jump in carrier number (Δn ~1 el/U) across phase II or metamagnetism supports the possibility of FS reconstruction.

Hall Coefficients: $U(Ru_{1-x}Rh_x)_2Si_2$, Rh 0 and 4 %

[Y. S. Oh et al. to be submitted]



n_{Hall}~0.15/U in Phase II in both compounds

n_{Hall}~1.0 el/U in PFL of both compounds

In URu_2Si_2, n_H increases with polarization

Implications from the Hall Effect Studies

1. URu_2Si_2 is a compensate semi-metal ; n_{Hall}~0.20/U at $T>T_{HO}$.
   It became more extreme by formation of HO;, n_{Hall}~0.03/U at $T<T_{HO}$

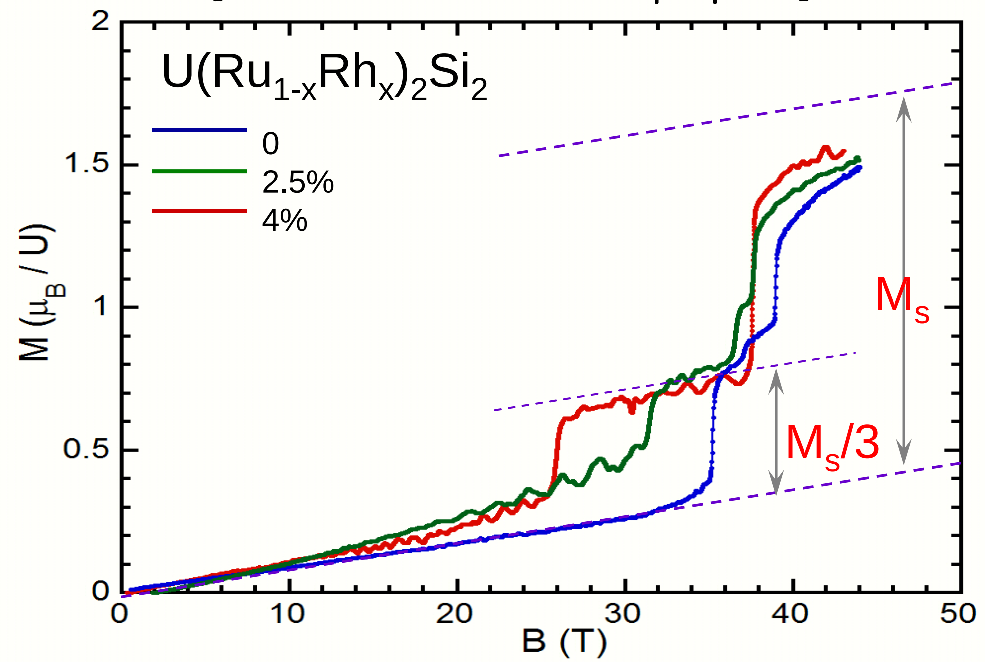
2. R_H has well defined plaeaux in each phase region, and sharply jumps, establishing a link among the carrier density, degree of polarization and order parameters.

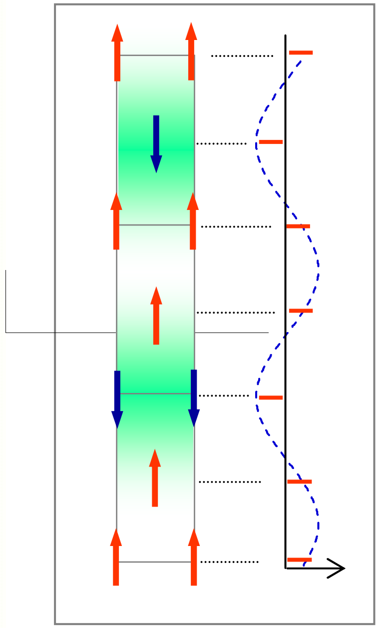
3. In URu_2Si_2, the order parameter becomes increasingly less efficient at gapping FS as it becomes more polarized at each transition, finally yielding ~1 hole per U beyond 39 T at ~1.5μ_B per U.

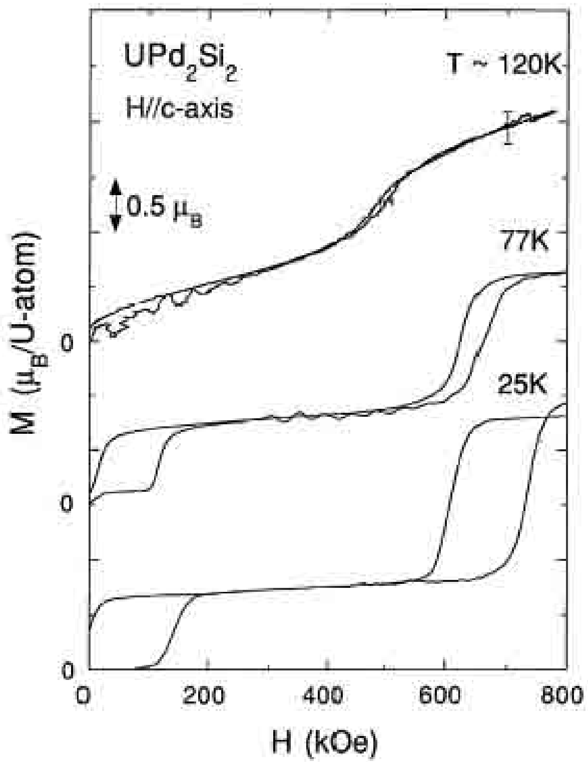
HO forms a gap in the α pocket, possibly through hybridization with the local XY ordering of Γ_5 CEF levels (density wave like?).

Clues to Phase II & Γ_5 CEF Doublets

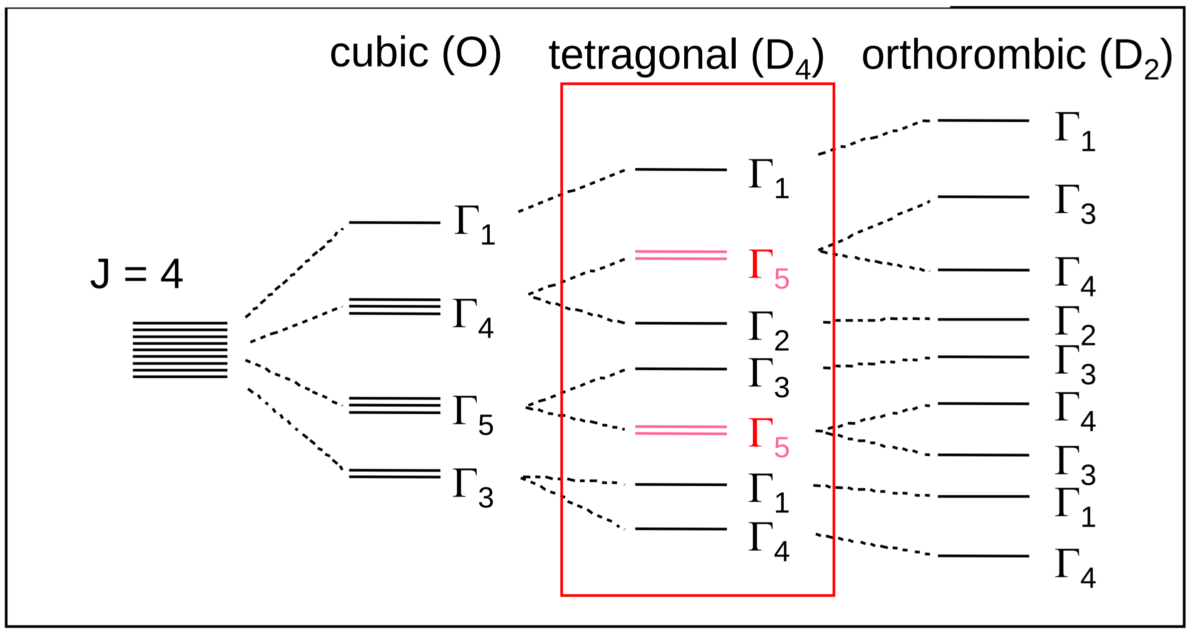
[K. H. Kim et al. SCES05, preprint]







[Honma et al.,J.Phys.Soc.Japan 67,1017(1998)]

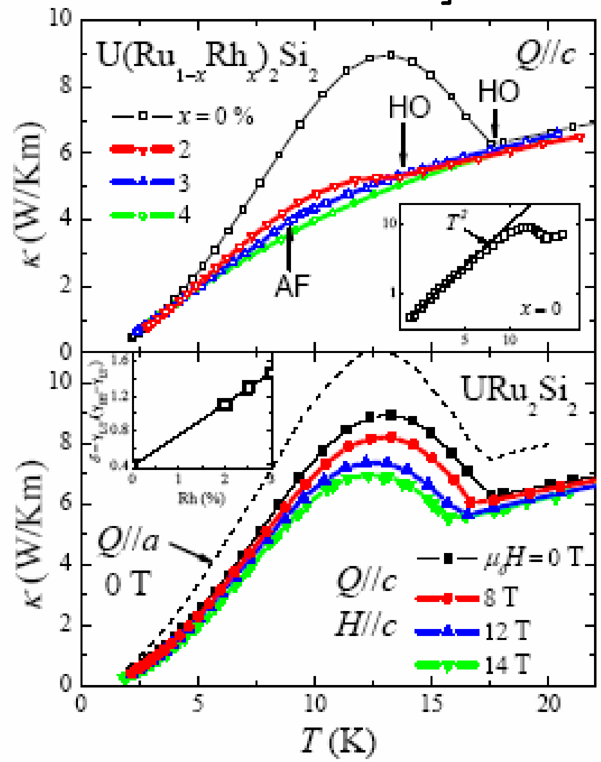


Groundstate of UPd_2Si_2

very well described in terms of Γ_5 CEF doublets, albeit without Kondo effect

Opening of FS partial gap results in the increase phonon thermal conductivity in the Rh 0 % and $PrFe_4P_{12}$

[Sharma et al. PRL 2006]



Very similar to the case of URu_2Si_2:

1. Semimetal due to heavy el channel and light hole channel

2. Large Nernst effect

3. Itinerant carriers (FS) are reduced with the AFQ order (no magnetic order).

4. Increased phonon thermal conductivity

Drastic Change in Transport of Entropy with Quadrupolar Ordering in $PrFe_4P_{12}$

[Pourret et al. PRL 2006]

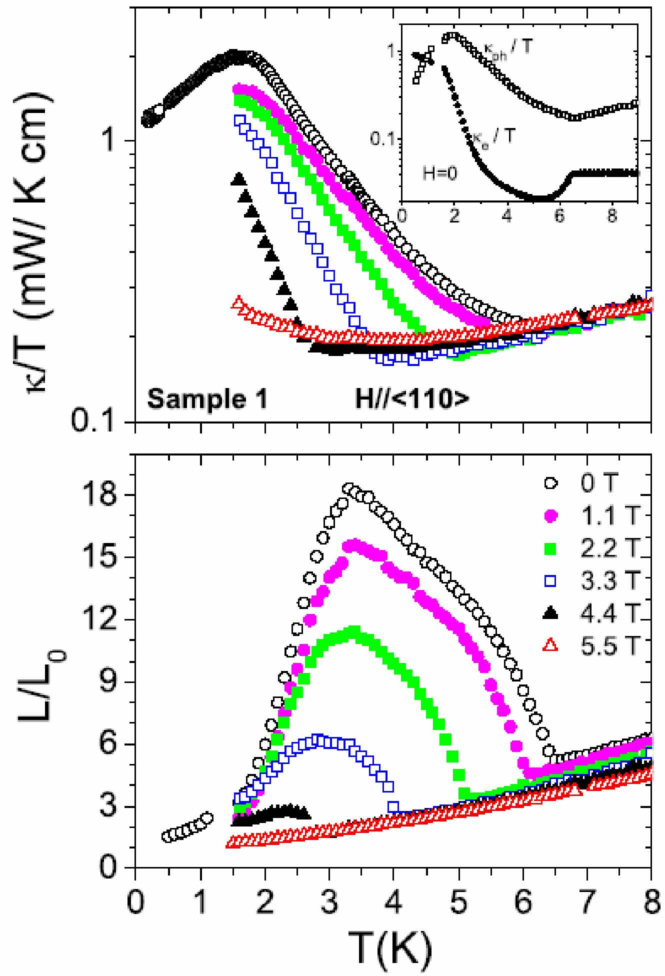


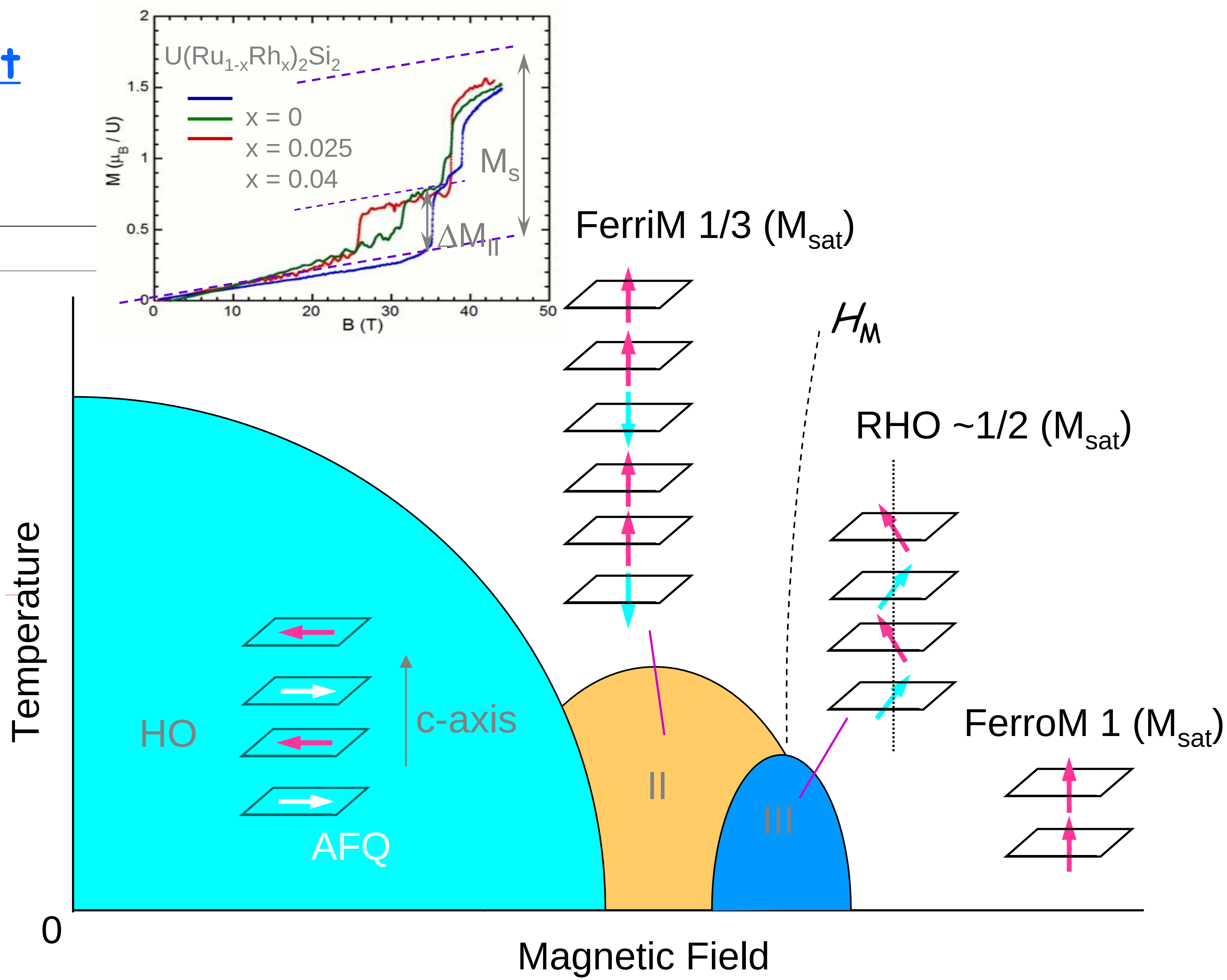
FIG. 1 (color online). Temperature dependence of the thermal conductivity divided by temperature (upper panel) and the normalized Lorenz number (lower panel) for different magnetic fields in $PrFe_4P_{12}$. The inset in the upper panel shows the temperature dependence of the phononic and electronic contributions to thermal conductivity at zero field assuming the validity of the WF law in the whole temperature range.

Proposal for Multiple Phases

Purely local moment picture assumed

Itinerant picture requires one to consider quasiparticles with e, J_z and $J_{x,y}$ quantum numbers

1/3 state perhaps like that seen in UPd_2Si_2 with neutrons [Honma et al. JPSJ 67 (1998) 1017]



Summary

1) $U(Ru,Rh)_2Si_2$ shows the nexus between phase formation and the field-induced quantum criticality:

   i) Quantum criticality similar to other itinerant metamagnets
   ii) Unambiguous evidence for phase formation
   iii) Nexus between phase formation and QCP, via avoiding quasiparticle instability
   iv) Discontinuous Hall constant jump comparable to Δn ~1 electron/U. strongly supports the significant FS reconstruction across phase boundaries .

2) Further thoughts:

   i) Our data support the Γ_5 doublet ground states ($5f^2$) in $U(Ru,Rh)_2Si_2$

   [i.e. 1/3rd *M*sat in phase II, large hysteresis in phase II, highly anisotropic *g* value]

   → HO can be related to the itinerant version of antiferro-quadrupolar ordering of Γ_5 doublets.

   ii) Quantum criticality with FS reconstruction?.
   iii) Theory of field-induced transition from heavy FL to spin-polarized FL; A new order parameter?