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Workshop on
Novel States and Phase Transitions in Highly Correlated Matter

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Recent developments in Ce115 and Pu115 Systems

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Recent Developments in Ce115 and Pu115 Systems

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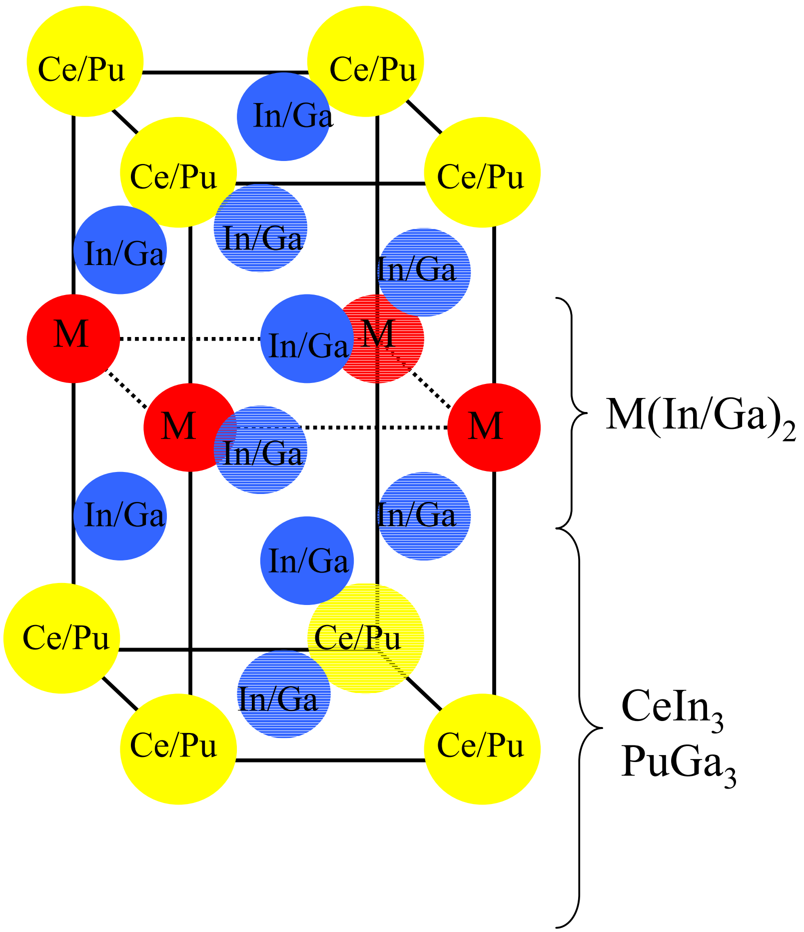
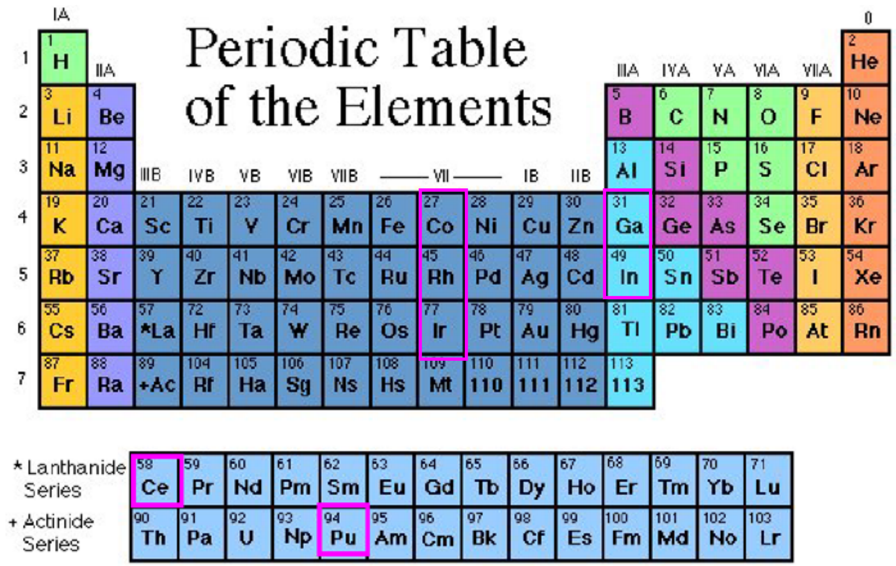
Outline:

- Crystal and electronic structures of $CeMIn_5$ and $PuMGa_5$ (M=Co, Rh, Ir)
- Overview of unconventional superconducting and normal states of $CeMIn_5$
- Problems posed by recent work on Ce115s
  - $\cot(\theta_H) \propto T^2$
  - possible Fulde-Ferrell-Larkin Ovchinnikov state in $CeCoIn_5$
  - mechanism of superconductivity in $CeIrIn_5$
- $PuMGa_5$ and their structure-property relationships to $CeMIn_5$
- Summary

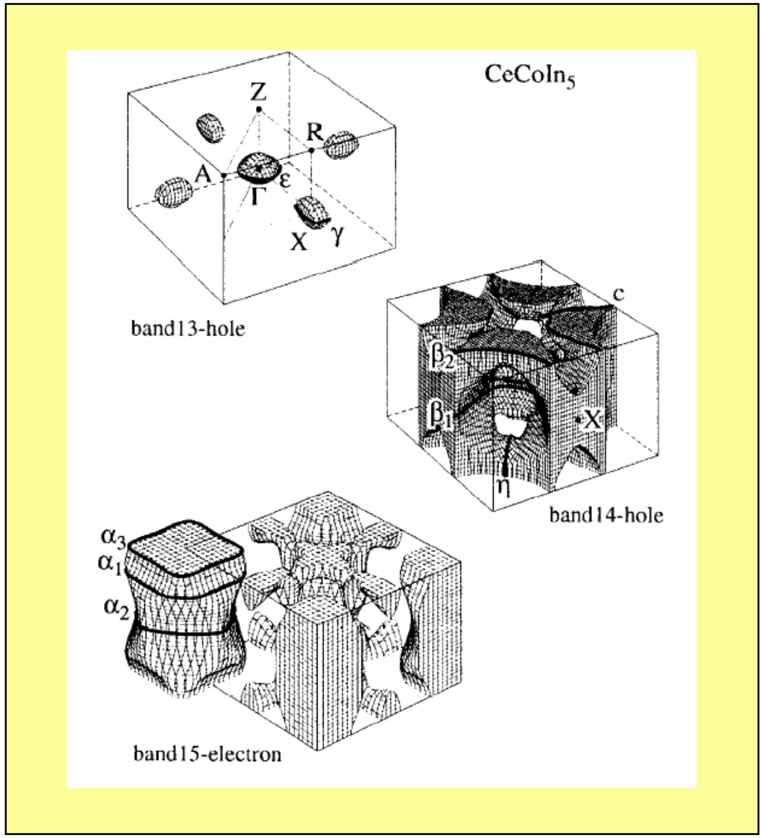
$HoCoGa_5$-structure type of $CeMIn_5$ and $PuMGa_5$



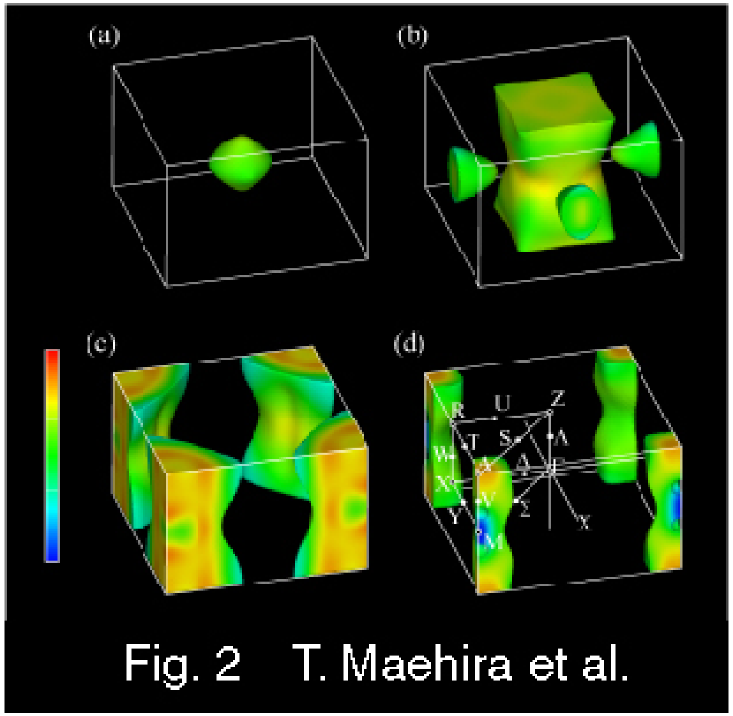


- $CeMIn_5$ and $PuMGa_5$ (M= Co, Rh, Ir)
- isoelectronic: Ce^{3+} ($4f^1$) and Pu^{3+} ($5f^5$)
- built from alternating layers of distorted fcc $CeIn_3$ ($PuGa_3$) and parallelepipeds ‘$M(In/Ga)_2$’
  - $CeIn_3$ unit: heavy-fermion antiferromagnet
  - $PuGa_3$ unit: similar to fcc δ-Pu

Electronic anisotropy: quasi-2D Fermi surface



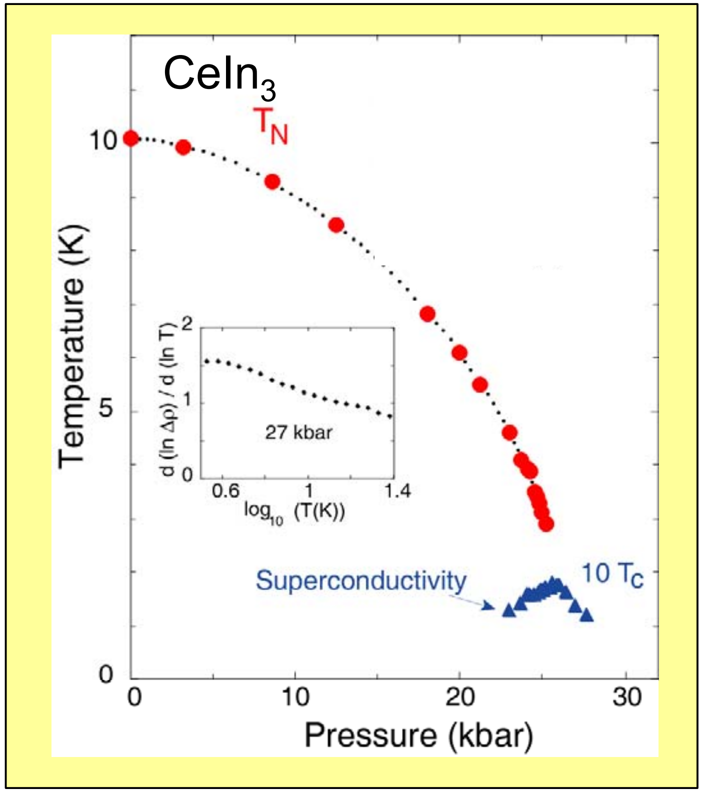
R. Settai et al., JPCM **13**, L627 (2001)



T. Maehira et al., PRL **90**, 207007 (2003); I. Opahle and P. M. Oppeneer, PRL **90**, 157001 (2003).

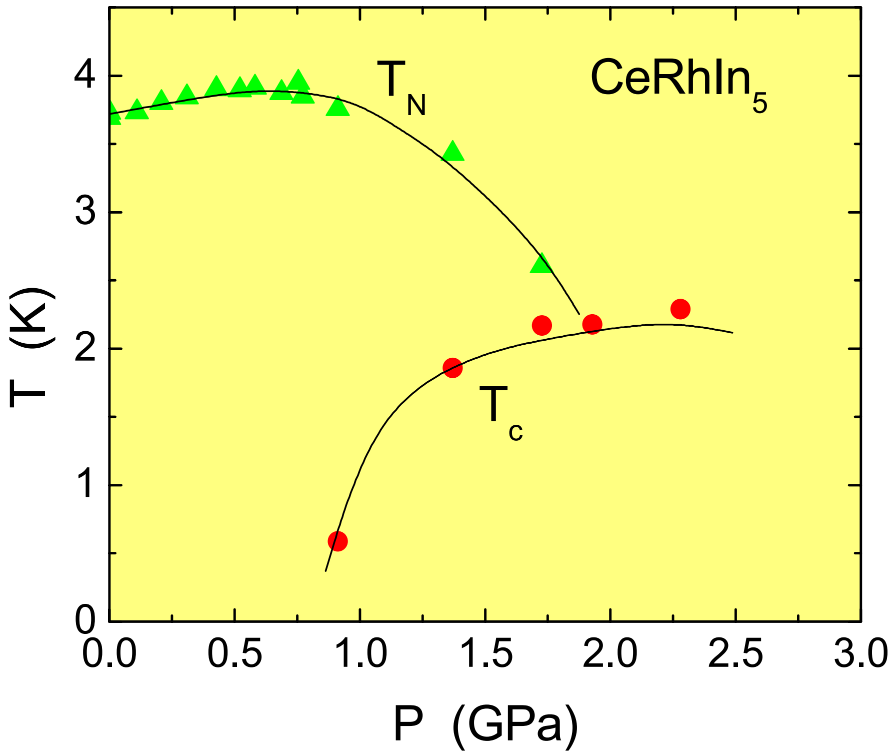
- dominant nearly cylindrical sheet ⇒ quasi-2D electronic structure
- Fermi-surface topology and large effective electron masses ($m^*/m_0 \geq 80$) confirmed by dHvA in $CeMIn_5$ (H. Shishido et al., JPSJ **71**, 162 (2002)); consistent with large electronic specific heat >400 mJ/mole K^2 of these heavy-fermion systems
- similar quasi-2D Fermi surfaces in $CeMIn_5$ and $PuMGa_5$
- neglects strong correlation effects

Superconductivity in proximity to antiferromagnetism



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

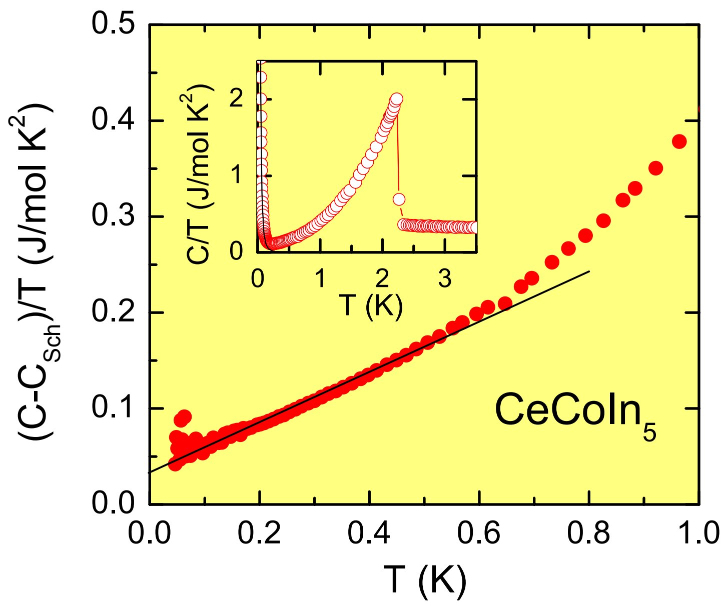
- T_c~ 200 mK at $P_c \approx$ 25 kbar
- suggests magnetic fluctuations mediate SC
- $T_c^{max}/T_N(P=0) \approx 0.02$



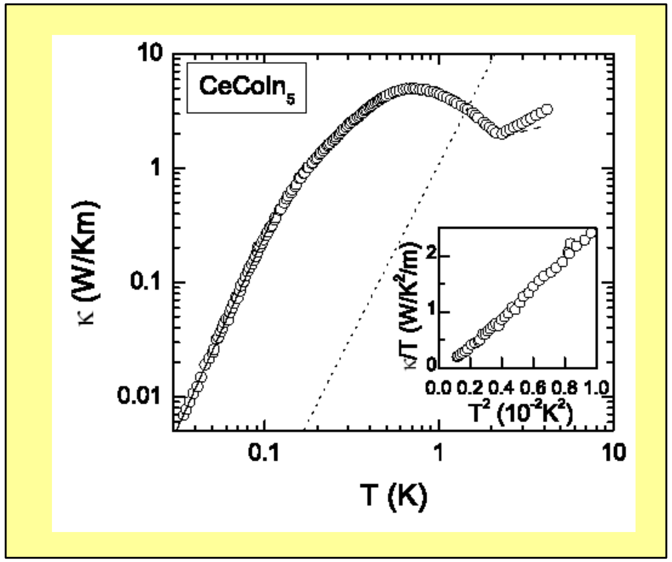
M. Nicklas et al., cond-mat/0405636

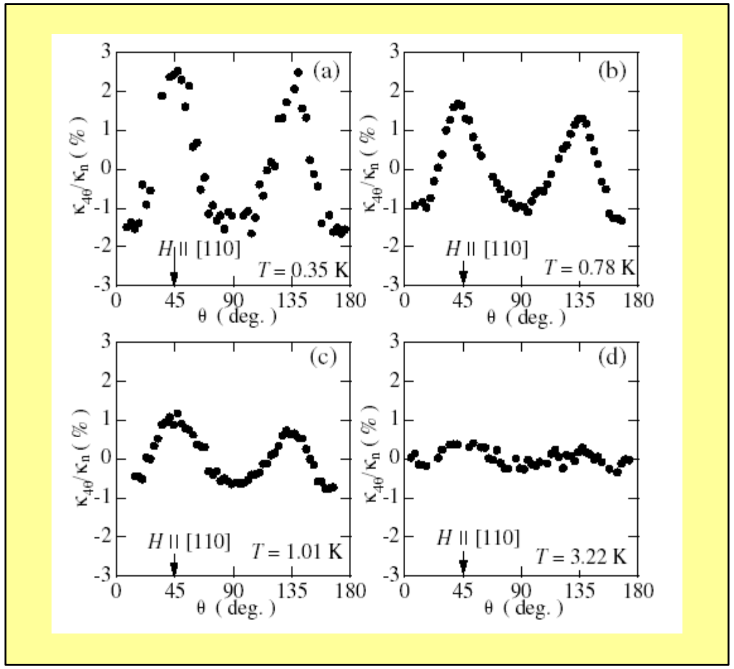
- T_c ~ 2.4 K at 2.5 GPa (25 kbar)
- broad P-range of coexisting AFM and SC; confirmed by NQR (Mito et al., PRL **90**, 077004 (2003)) and neutron diffraction (A. Llobet et al., PRB **69**, 024403 (2004))
- $T_c^{max}/T_N(P=0) \approx 0.6$

Unconventional superconductivity in $CeMIn_5$, e.g. $CeCoIn_5$



R. Movshovich et al., PRL 86, 5152 (2001)



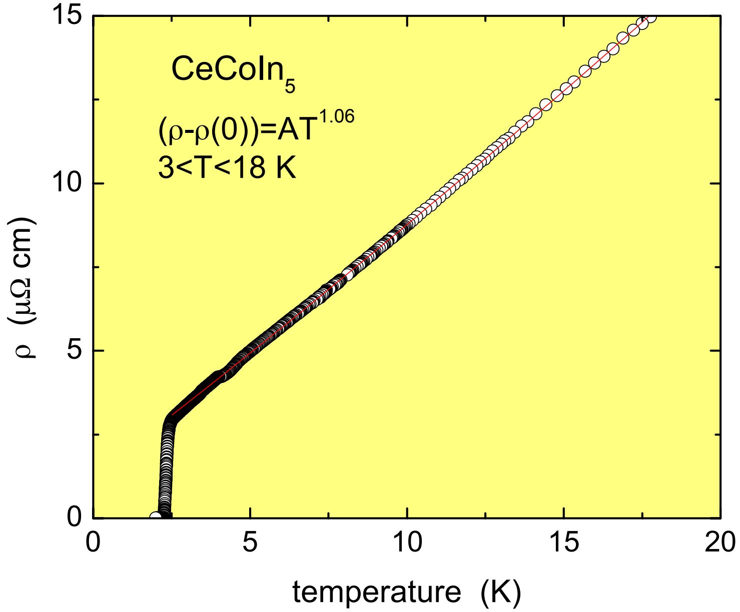


♦ power laws in C/T, κ and $1/T_1$ below T_c in each $CeMIn_5$ member (for brief review: JDT et al, Physica B **329**, 446 (2003))

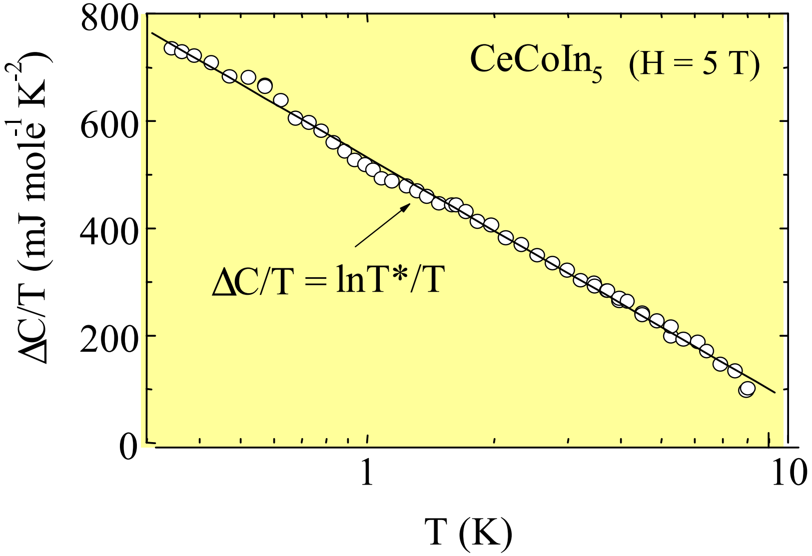
♦ strong 4-fold modulation of κ and C/T for H in the a-b plane of $CeCoIn_5$ (K. Izawa et al. PRL **85**, 057002 (2001)

♦ consistent with $d_{x^2-y^2}$ symmetry from κ(H) and with d_{xy} symmetry from C(H) (H. Aoki et al., J. Phys.: Condens. Matter **16**, L13 (2004))

Unconventional normal state in $CeCoIn_5$



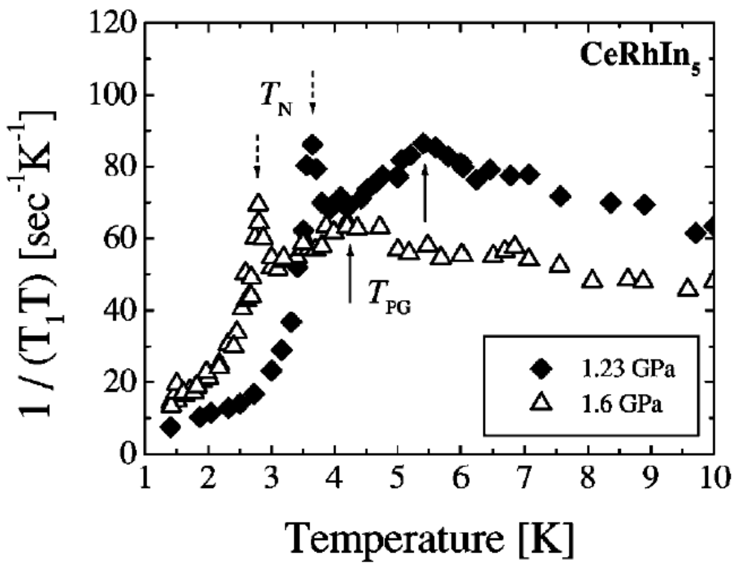
V. A. Sidorov et al., PRL **89**, 157004 (2002)



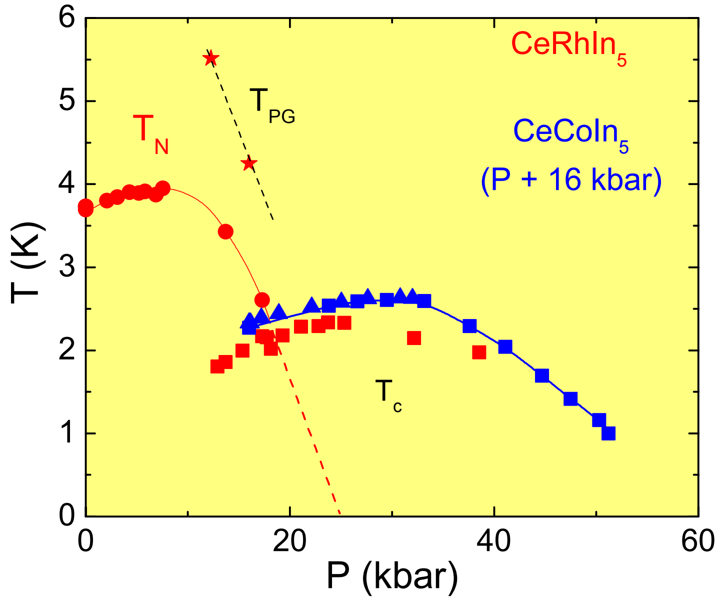
e.g., A. Bianchi et al., PRL 91, 257001 (2003)

- from $\sim T_c < T <$ 10-15 K, $(\rho-\rho_0) = AT^n$, where n=1.0±.05; not T^2 expected of a Landau Fermi liquid
- for $H > H_{c2}(0)$, $C/T \propto -\log T$ from at least 0.3 to 9 K
- T-linear resistivity and $C/T \propto -\log T$ characteristic of proximity to a quantum-critical point
- non-Fermi liquid temperature dependences as well in $CeIrIn_5$ and $CeRhIn_5$

T–P phase diagram for $CeRhIn_5$ and $CeCoIn_5$



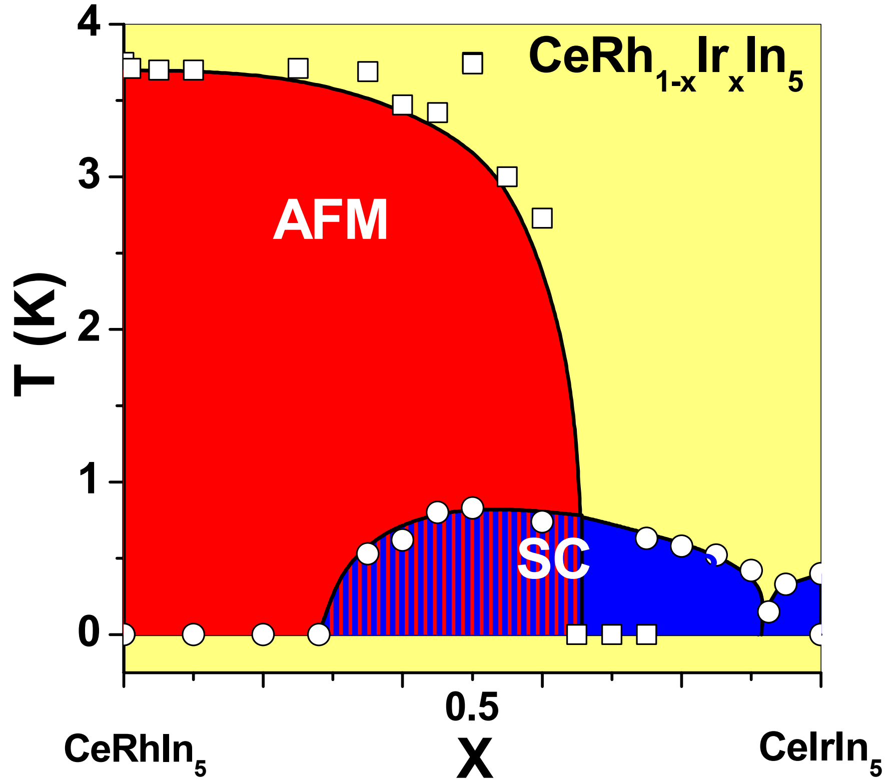
S. Kawasaki et al, PRB **65**, 020504 (2001)

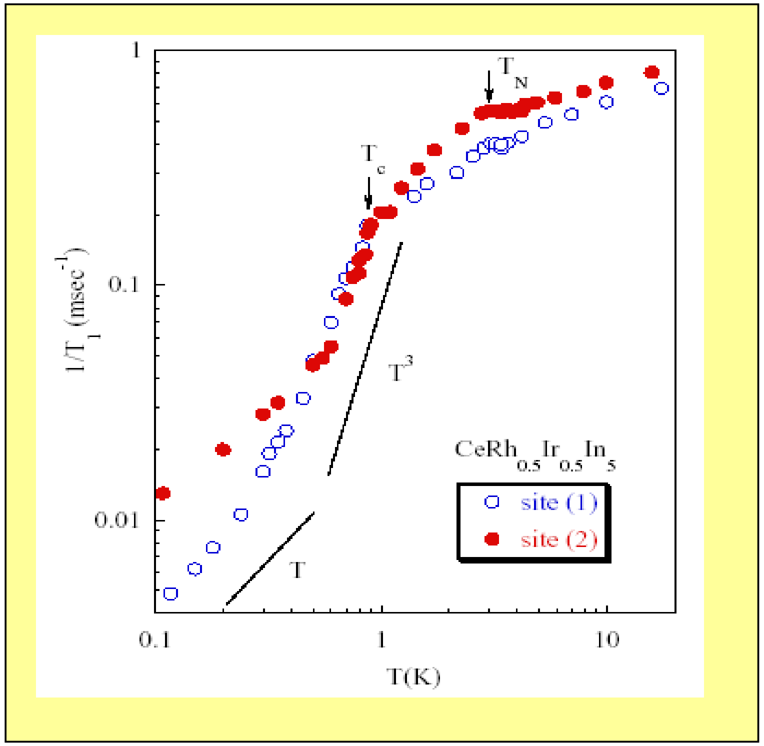


V. A. Sidorov et al., PRL **89**, 157004 (2002)

- ♦ possible evidence for pseudogap-like response in $1/T_1$ of $CeRhIn_5$ (approx. coincident with departure from T-linear ρ(T))
- ♦ smaller cell volume of $CeCoIn_5$ ⇒ +16 kbar chemical pressure relative to $CeRhIn_5$
- ♦ similar T-P diagram for $CeRhIn_5$ and T-(P+16 kbar) diagram for $CeCoIn_5$
- ♦ stars are signatures from NMR for a spin pseudogap in $CeRhIn_5$
- ♦ T-P for $CeCoIn_5$ and $CeRhIn_5$ similar to T-x phase diagram for cuprates ⇒ physics common to both?

Magnetism and superconductivity in $CeRh_{1-x}Ir_xIn_5$





P. G. Pagliuso et al., Phys. Rev. B **64**, 100503 (2001);
A. Bianchi et al., Phys. Rev. B **64**, 220504 (2001)

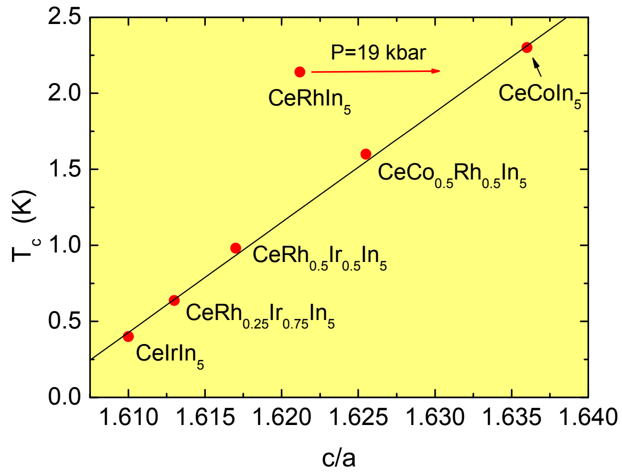
G.-q. Zheng et al., PRB (in press)

- coexistence of AFM and SC from thermodynamic and transport
- $1/T_1$ measured on same NQR line for all T $\Rightarrow$ superconductivity and magnetism from same electrons
- only a single T_1 below T_N $\Rightarrow$ spatially homogeneous coexistence of AFM and SC

Summary of $CeMIn_5$

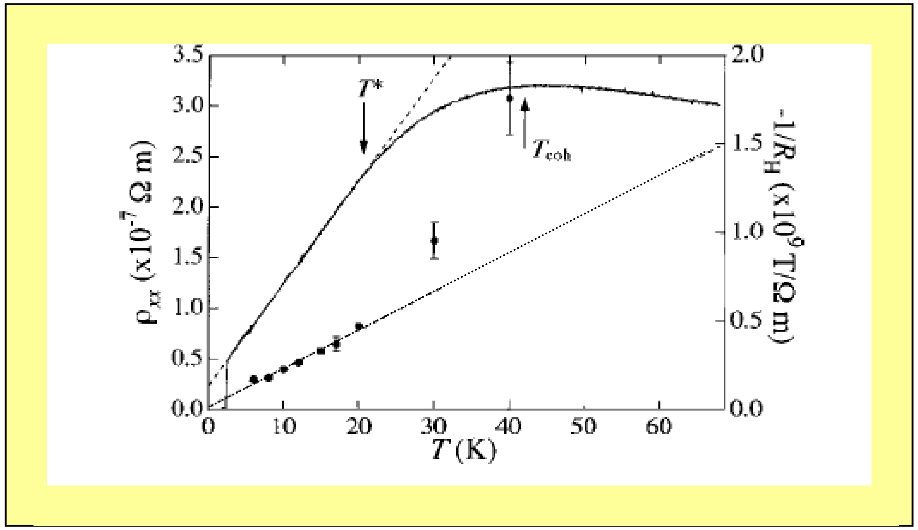
- layered crystal structure and corresponding quasi-2D electronic structure
- power laws below T_c ⇒ unconventional, probably d-wave, superconductivity
- broad ranges of P and x in which AFM and SC coexist at a microscopic level in $CeRhIn_5$ and $CeRh_{1-x}Ir_xIn_5$
- unconventional (NFL) normal states in each family member
- reasonable speculation that superconductivity is magnetically mediated
- possible evidence for a pseudogap in $CeRhIn_5$ under pressure

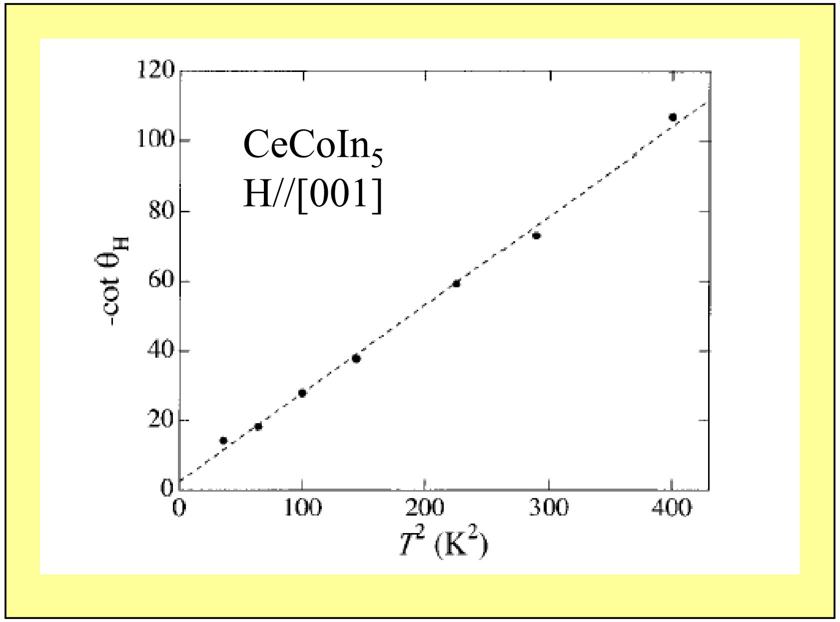
- in spite of richness in properties, T_c's apparently correlated with ratio of tetragonal lattice parameters *c/a* and not cell volume or M



- next--more recent work on Ce115s
  - $\cot(\theta_H) \propto T^2$
  - possible Fulde-Ferrell-Larkin Ovchinnikov state in $CeCoIn_5$
  - mechanism of superconductivity in $CeIrIn_5$

Hall effect in $CeCoIn_5$

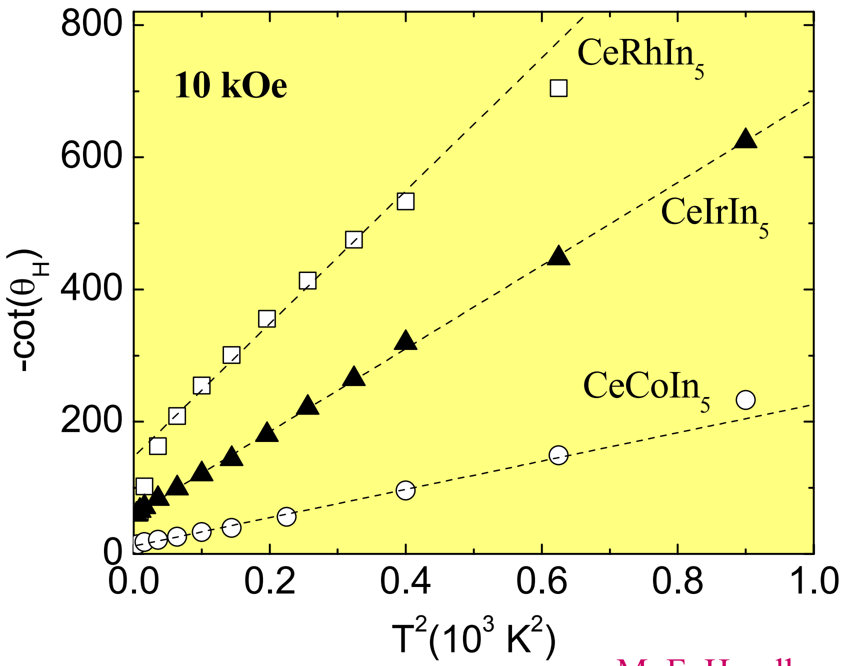


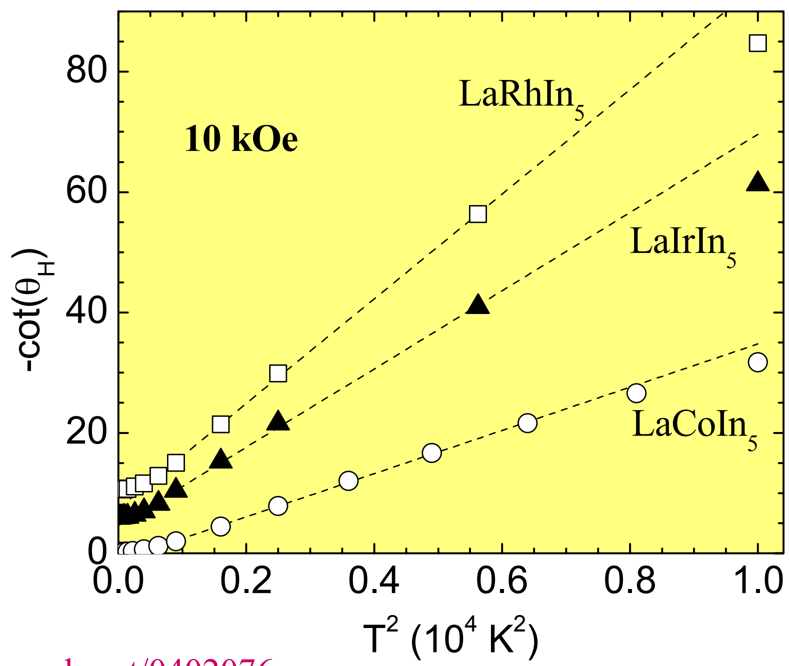


Y. Nakajima et al., J. Phys. Soc. Jpn. **73**, 5 (2004)

- $-1/R_H$ linear in T over same temperature range that $\rho \propto T$
- $-\cot(\theta_H) = \alpha + \beta T^2$, with $\alpha \approx 0$
- both as in cuprates (2 distinct relaxation rates associated with spinon and holon excitations in 2D CuO_2 planes) and in $YbRh_2Si_2$
- alternative: AFM fluctuations near a QCP
  - $\rho_{xx} \sim \xi_{AF} T^2 \sim \chi_Q T^2$
  - $R_H \sim \pm\xi_{AF} \sim \chi_Q$, where χ_Q = staggered susceptibility and ξ_{AF} = AF correlation length
  - $\chi_Q \sim 1/(T+\theta)$
  - therefore: $\rho_{xx} \propto T$, $1/R_H \propto T$ and $-\cot(\theta_H) \propto T^2$

Hall effect in 115s





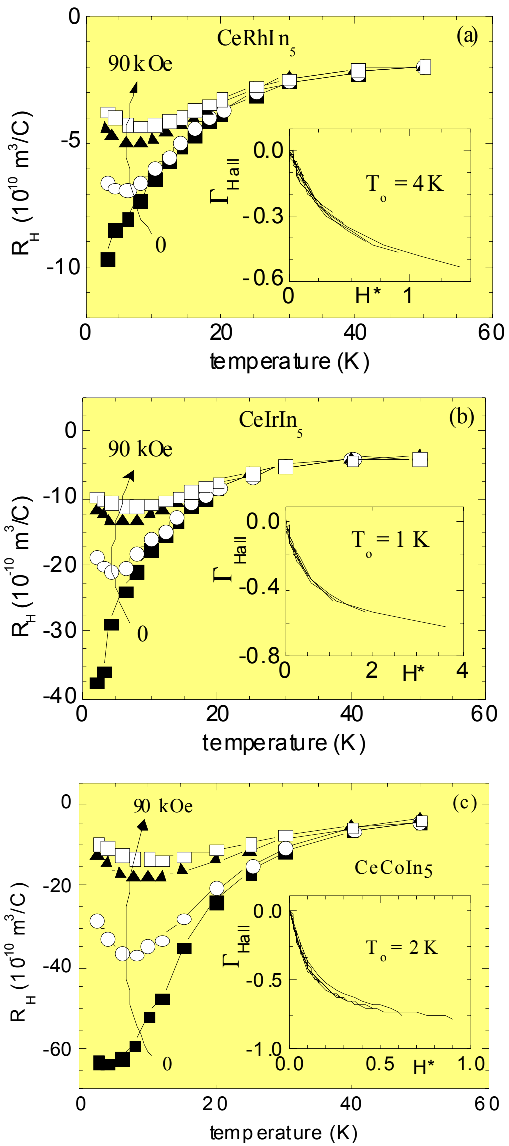
M. F. Hundley et al., cond-mat/0402076

- $-\cot(\theta_H) = \alpha + \beta T^2$, not only for $CeCoIn_5$ but also for $CeRhIn_5$ and $CeIrIn_5$

- perhaps due to AFM fluctuations near a QCP, but La analogues also show a similar dependence (at higher temperatures and in same sequence with roughly an order of magnitude smaller values)

- suggests interplay of AFM fluctuations and underlying electronic structure

Hall effect in 115s (cont.)



M. F. Hundley et al., cond-mat/0402076

- below about 35 K, where skew scattering is negligible and where $-\cot(\theta_H) \propto T^2$, H and T dependence of R_H, given by

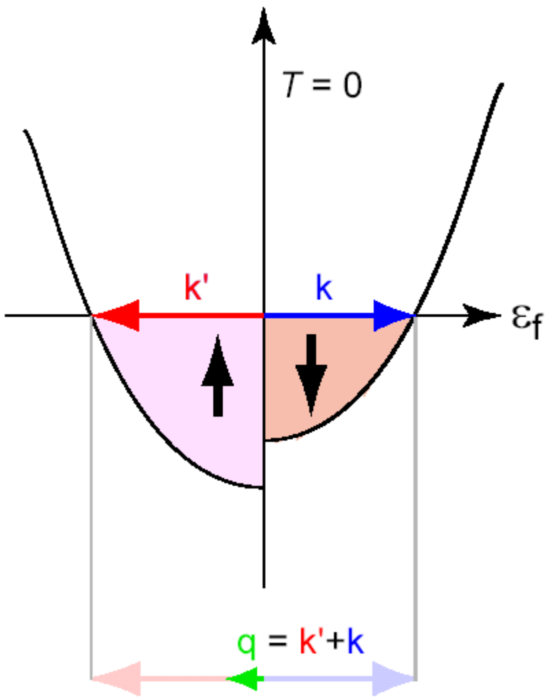
$$\Gamma_{Hall} = (R_H(H) - R_H(H\to 0))/R_H(H\to 0),$$

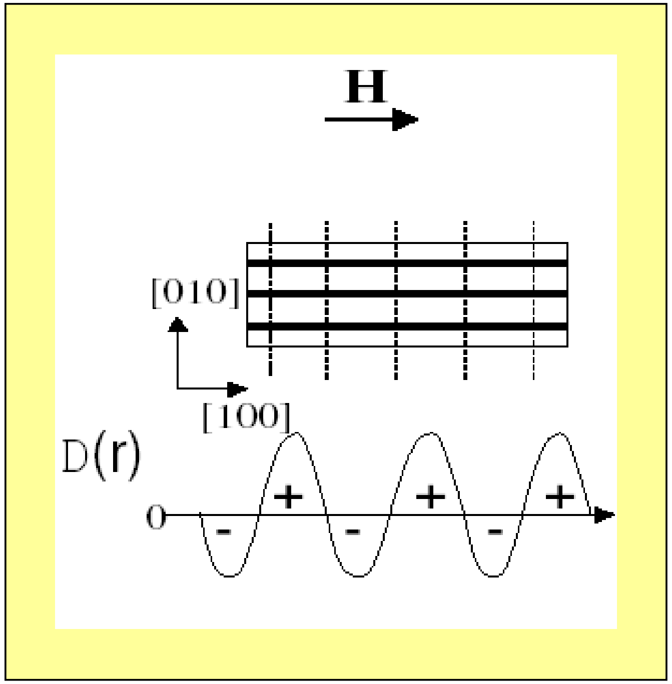
scales as $H^* = H/(T + T_0)^{\beta}$, where $\beta = 2.0 \pm 0.1$ and $T_0 = 4$, 1 and 2 K for $CeRhIn_5$, $CeIrIn_5$ and $CeCoIn_5$, respectively

- T_0 for $CeCoIn_5$ same as deduced from 2-fluid model of a Kondo lattice (S. Nakatsuji et al., PRL **92**, 016401 (2004)) in which T_0 reflects the scale on which the redistribution of f-spectral weight from high to low energies saturates

- scaling reflects the development of the heavy-mass state?

Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state





♦ magnetic field splits spin-up and spin-down bands, lowering the energy of the magnetic spin along the field

♦ if pairing electrons with opposite spins, momentum of the up-spin electron > momentum of the down-spin electron, resulting in a finite total momentum of the Cooper pair of q.

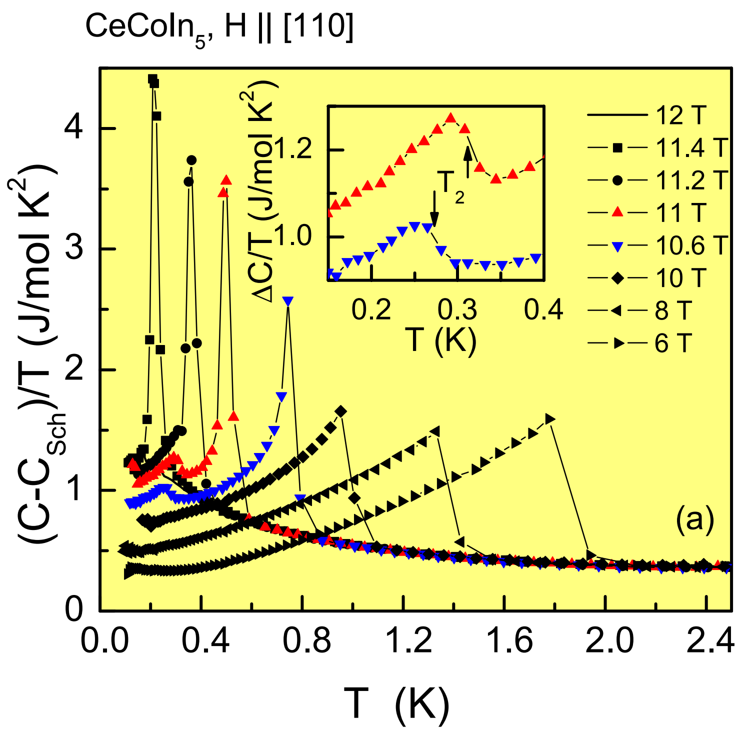
♦ SC gap modulated in real space:

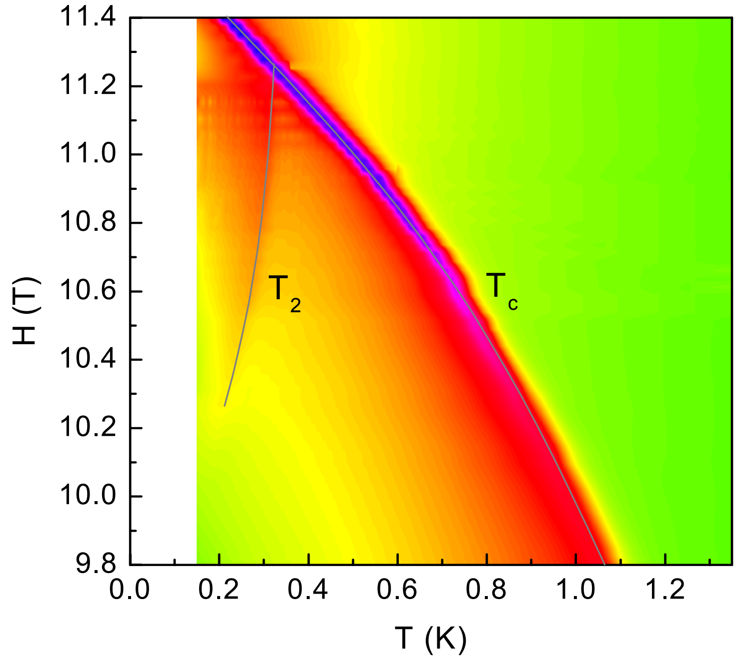
$\Delta(r) = |\Delta_q| e^{(iqr)}$ (Fulde-Ferrell)

$\Delta(r) = |\Delta_q| \cos(qr)$ (Larkin-Ovchinnikov)

♦ lines of gap zeros ⊥ H

FFLO in $CeCoIn_5$

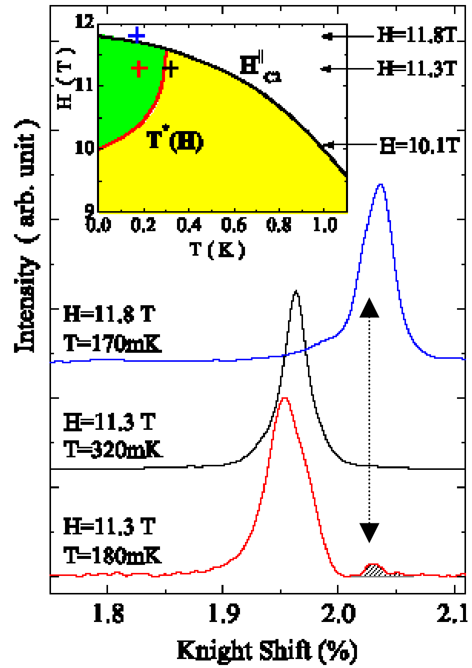




A. Bianchi et al., Phys. Rev. Lett. **91**, 257001 (2003)

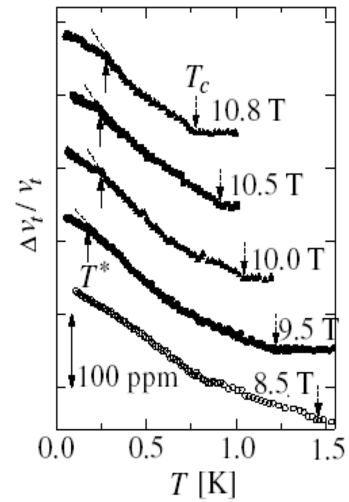
- conditions required to observe FFLO: clean limit and Pauli limited (Maki parameter $\alpha = \sqrt{2}H_{c2}^{orbital}/H_P > 1.8$)
- in $CeCoIn_5$, $l_{mfp} >> \xi_{GL}$ and $H_{c2}(T)$ strongly Pauli limited ($\alpha = 4.5$)
- as expected for Pauli limiting, crossover in $H(T_c)$ from second to first order below ≈1 K
- in the superconducting state below $H(T_c)$, find a second order transition at T_2, consistent with expectations of FFLO; similar results for H//[001]
- also evidence for 1st order transition at $H(T_c)$ and for feature at $T_2(H)$ in thermal conductivity (C. Capan et al., cond-mat/0401199)

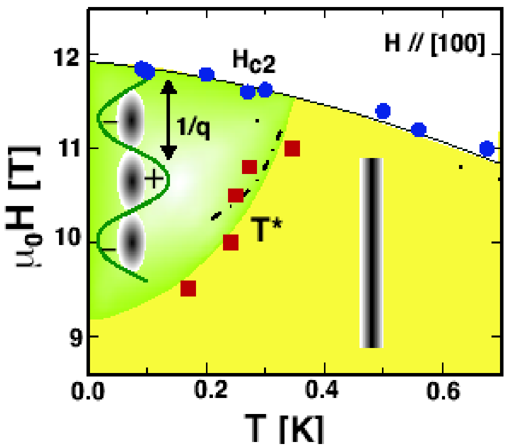
Further evidence for FFLO state



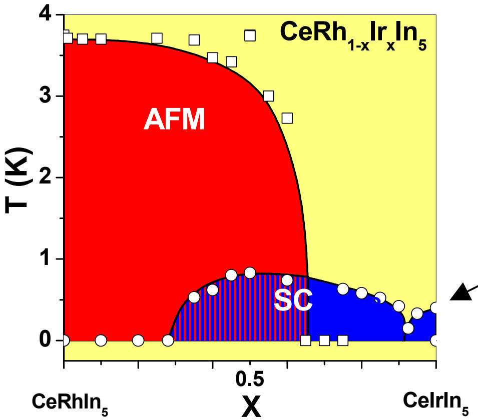
- ♦ ^{115}In-NMR finds a second resonance in FFLO regime (red curve) at approximately the same shift as in normal state (blue curve) and absent in SC but non-FFLO regime (black curve) (K. Kakuyanagi et al., cond-mat/0405661)
- ♦ expected response for FFLO normal quasiparticles in planes ⊥ H
- ♦ structure of vortex cores/lattice?

- ♦ change in slope at $T^*=T_2$ of relative shift of transverse sound velocity of the flux line lattice (T. Watanabe et al., cond-mat/0312062)
- ♦ possibly driven by a change in vortex core structure in FFLO state

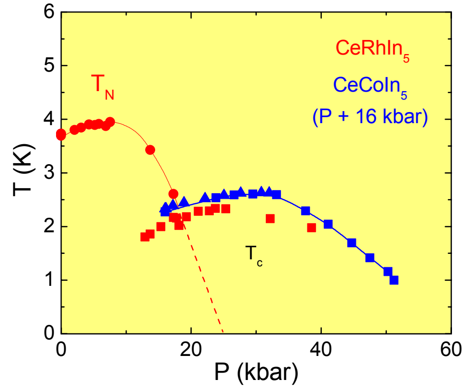


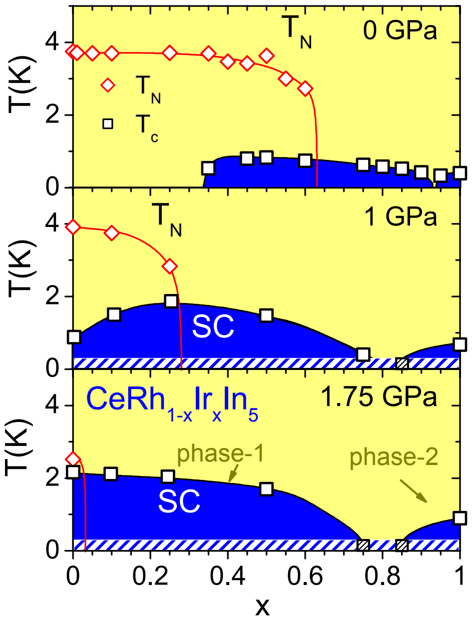


$CeIrIn_5$: apparently distant from antiferromagnetism

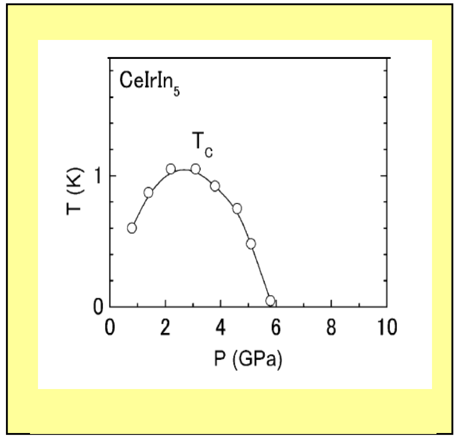


- SC in $CeRhIn_5$ and $CeCoIn_5$ in proximity to antiferromagnetism, but not obviously the case for $CeIrIn_5$





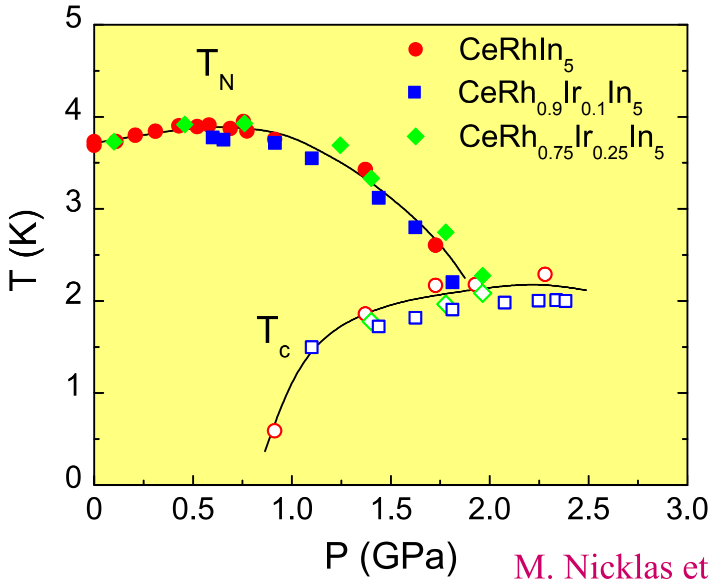
- under pressure, $CeIrIn_5$ even more ‘distant’ from AFM and isolated by a range of compositions where T_c =0
- $CeIrIn_5$ $T_c(P)$ domed-shaped, like $CeRhIn_5$ and $CeCoIn_5$
- mechanism of superconductivity?



T. Muramatsu et al., Physica C **388-389,** 539 (2003)

M. Nicklas et al., cond-mat/0405636

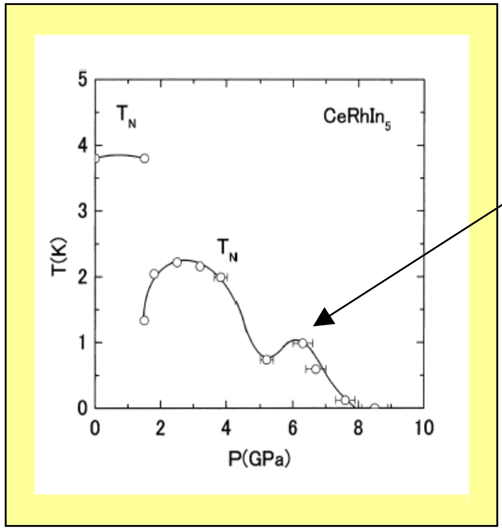
High pressure studies



M. Nicklas et al., cond-mat/0405636

♦ scaling of $T_c(P)$ and $T_N(P)$ by constant pressure shifts of 0, 0.1 and 0.6 GPa for x=0, 0.1 and 0.25, respectively $\Rightarrow$ Ir acts principally as as a positive chemical pressure given by $P_{Ir} \approx 10x^2$

♦ scaling breaks down for larger x, but qualitatively implies $CeRhIn_5$ under ≈ 10 GPa pressure relative to $CeIrIn_5$

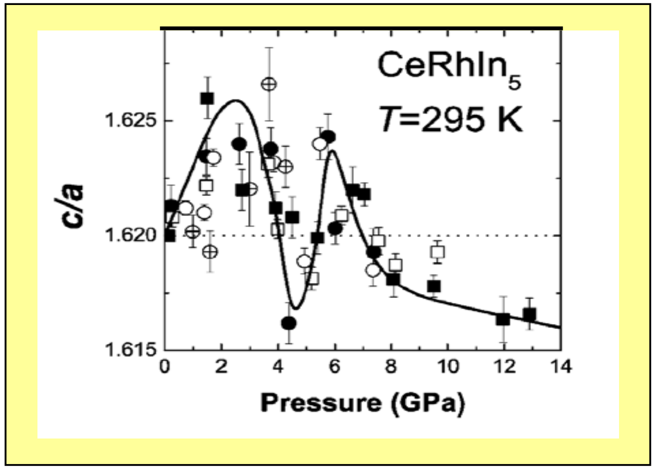


T. Muramatsu et al., J. Phys. Soc. Jpn. **70**, 3362 (2001)

♦ crudely, $P_{Ir}(x=1) \approx 6$ GPa, where second dome of SC emerges in $CeRhIn_5$

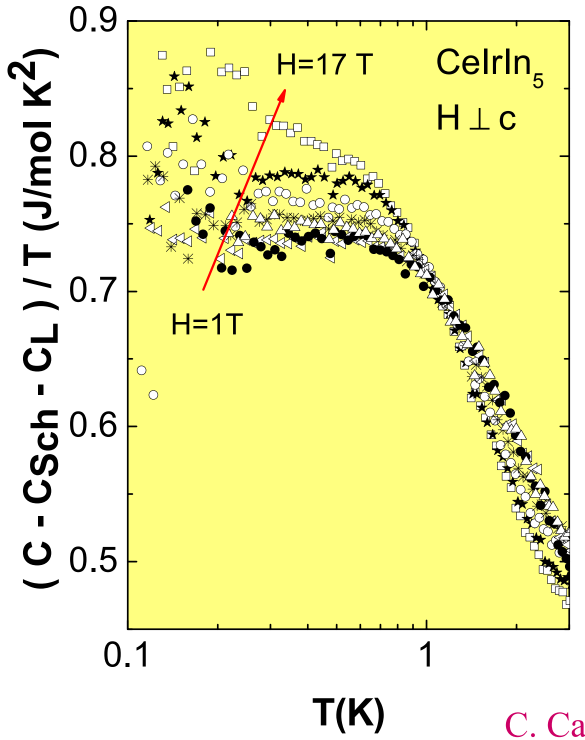
♦ max. T_c of 2nd dome ≈ max. T_c of $CeIrIn_5$

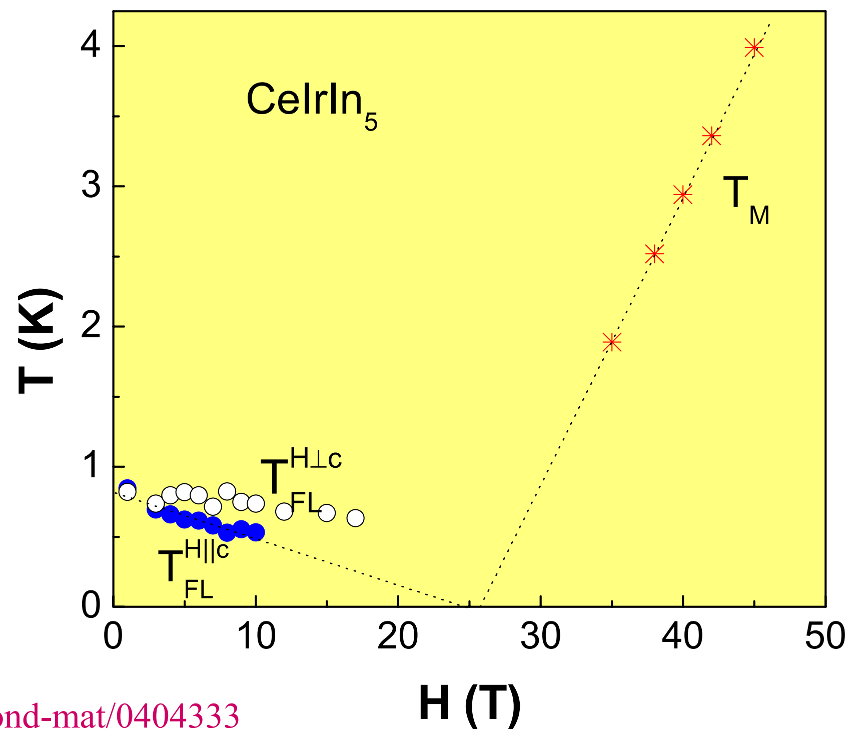
♦ consistent with smaller *c/a* ratio that tracks $T_c(P)$ $\Rightarrow$ SC more nearly 3D



R. S. Kumar et al., Phys. Rev. B **69**, 014515 (2004)

High field studies of $CeIrIn_5$





C. Capan et al., cond-mat/0404333

- with increasing field, low-T range below which C/T=const. narrows and C/T increasingly divergent ⇒ approach to a field-induced QCP near 25 T
- associated with metamagnetic transition $T_M(H)$ (J. S. Kim et al., Phys. Rev. B **65**, 174520 (2002))
- also, metamagnetic transition in $CeRhIn_5$ near 50 T (T. Takeuchi et al., J. Phys. Soc. Jpn. 70, 877 (2001))
- origin of the mean-field, field-induced transitions in $CeIrIn_5$ and $CeRhIn_5$ remain to be clarified, but speculate that these transitions and their fluctuations play a role in producing superconductivity in $CeIrIn_5$ at P=0 and, by inference, in $CeRhIn_5$ near 6 GPa.

Problems posed by new results

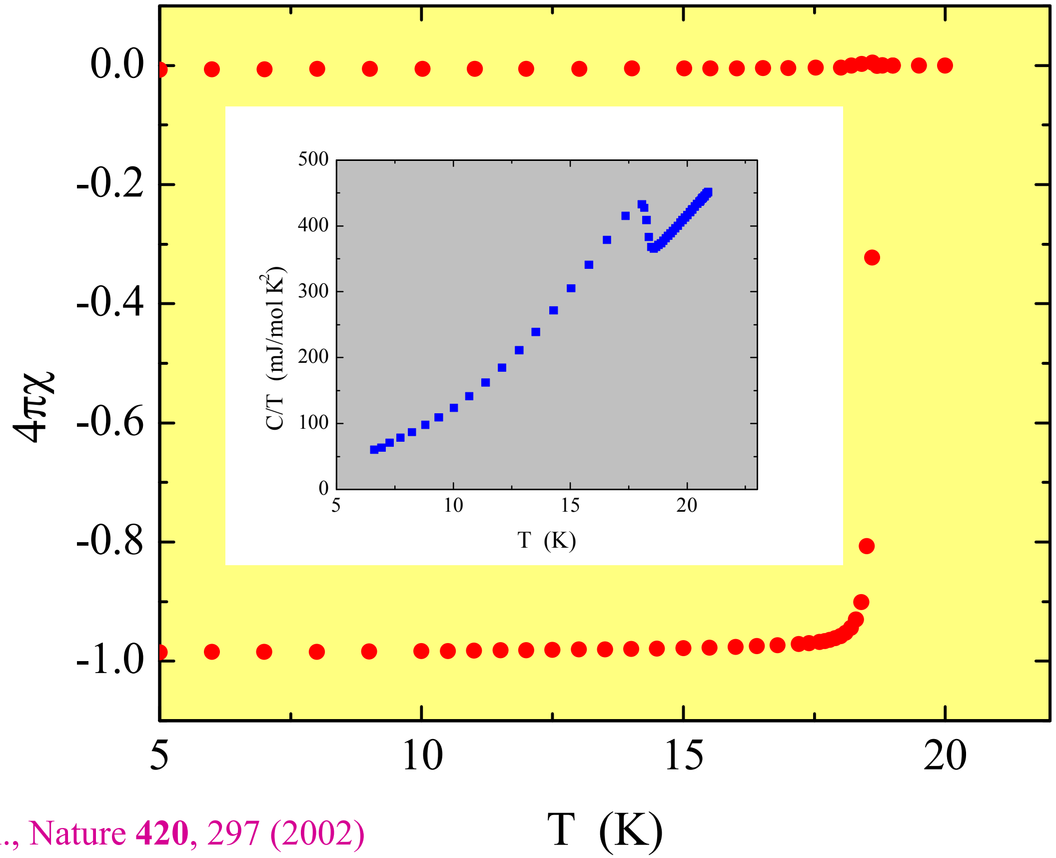
♦ What is the origin of $\cot(\theta_H) \propto T^2$? Is it related to and/or tell us anything about the cuprates? More generally, is it characteristic of proximity to a QCP?

♦ Is scaling of $R_H(H,T)$ revealing how the low-T heavy-mass state evolves out of high-energy energy excitations?

♦ If, as as may be the case, the FFLO state finally has been found after 40 years of searching, how do we understand the interplay between d-wave pairing (s=0, l=2) and the real-space modulation of a SC gap in which Cooper pairs have a finite momentum $|q| \sim 2\mu_B H/\hbar \nu_F$?

♦ Are excitations mediating SC in $CeIrIn_5$ qualitatively different than those responsible for SC in $CeRhIn_5$ and $CeCoIn_5$? Is there a field-induced QCP in $CeIrIn_5$ and $CeRhIn_5$? If so, what is the nature of this QCP and its excitations?

$PuCoGa_5$ superconductivity

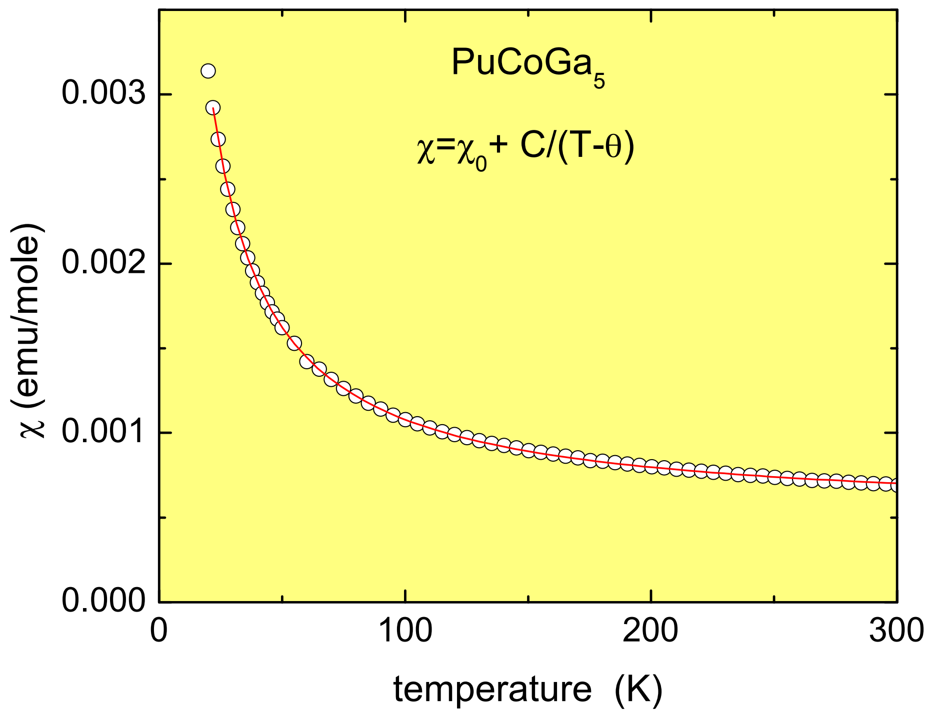


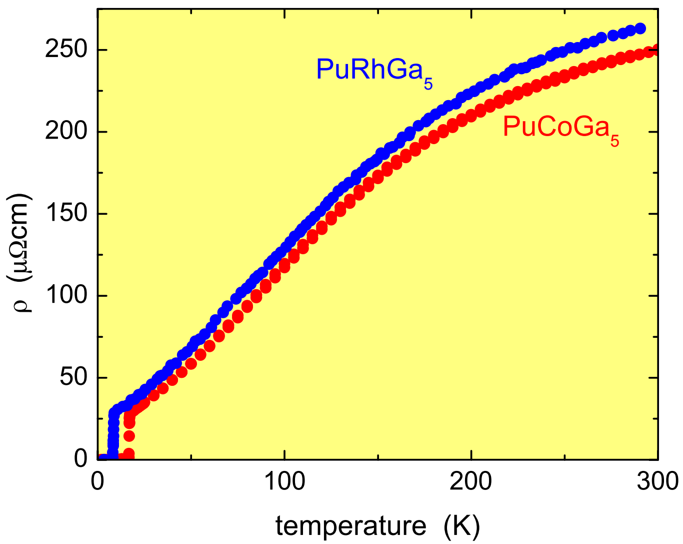
J. L. Sarrao et al., Nature **420**, 297 (2002)

- perfect diamagnetism (and strong flux pinning), zero resistivity and jump in C/T $\Rightarrow$ bulk superconductivity below T_c=18.5K
- assuming BCS weak coupling, $\Delta C/\gamma T_c$=1.43 $\Rightarrow$ γ =80 mJ/molK^2 ($\gamma \propto m^*/m_0$)
- $\Theta_D \approx$ 240 K; McMillan, μ^* =0.1 and λ =0.5 or 1.0 $\Rightarrow$ T_c=2.4-13.8 K
- possibly conventional superconductivity but maybe not

Normal state of $PuMGa_5$

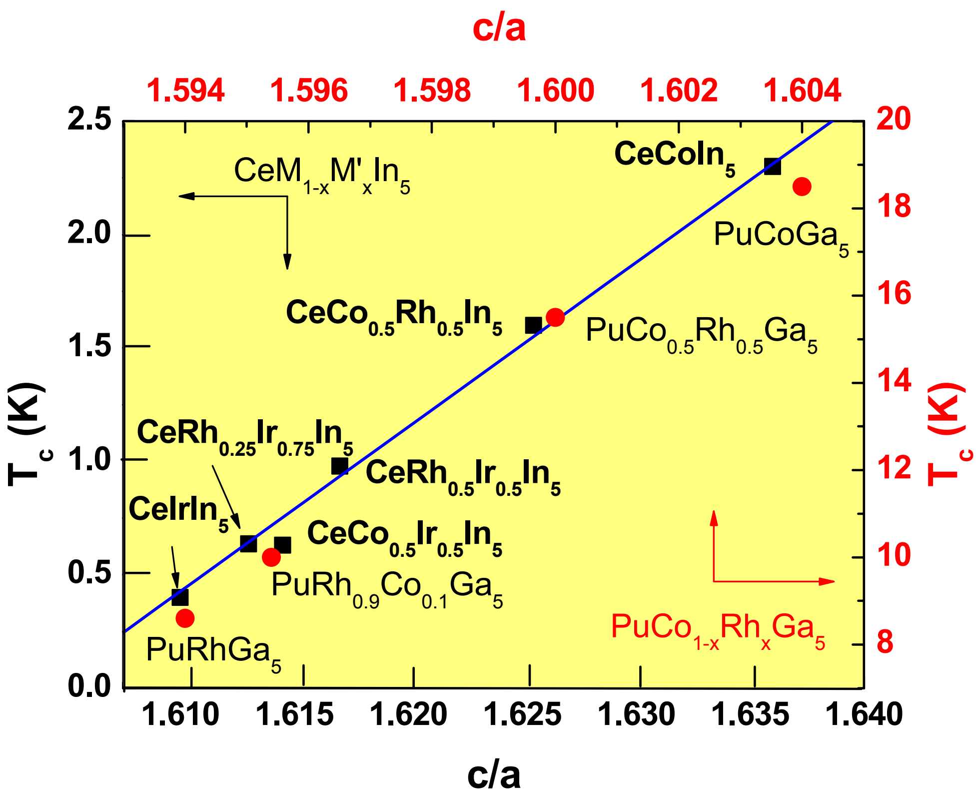
- normal state $\chi(T)$: $\chi = \chi_0 + C/(T-\theta)$, where $\chi_0 = 5.1 \times 10^{-4}$ emu/mole, $\theta = -2$K and effective moment $\mu_{eff} = 0.68\mu_B$
- μ_{eff} for $Pu^{3+} = 0.84\ \mu_B$ ($5f^5 6d^1 7s^2$)
- isostructural $PuRhGa_5$: $T_c = 8.7$ K and $\chi = \chi_0 + C/(T-\theta)$, with $\mu_{eff} = 0.60\mu_B$
- difficult to reconcile local-moment-like $\chi(T)$ with conventional superconductivity





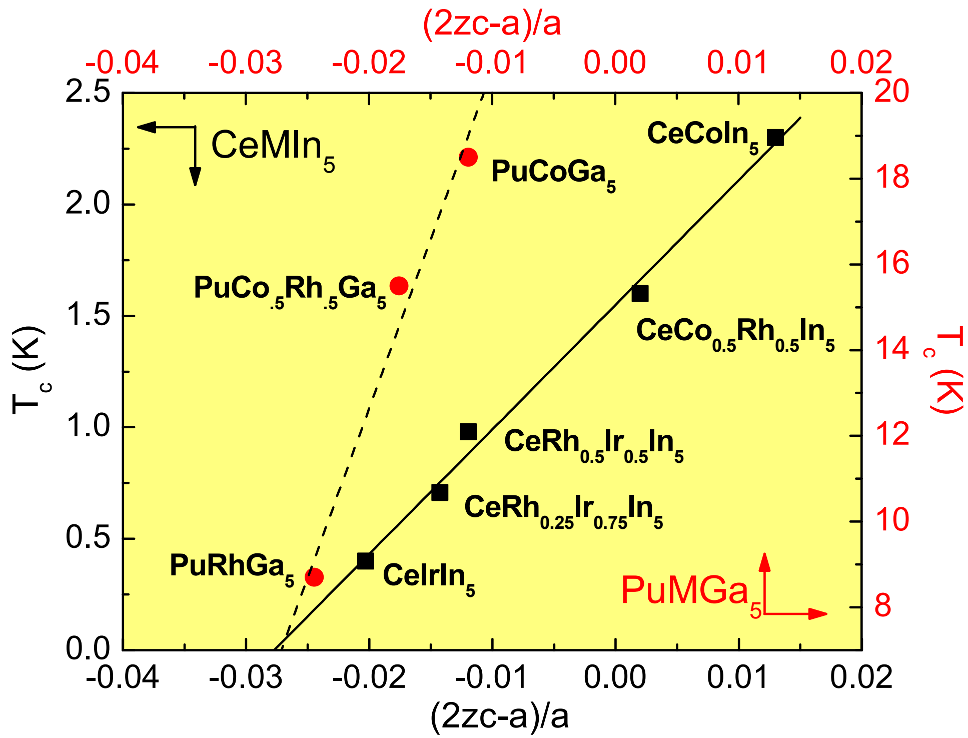
- 'S'-shaped $\rho(T)$ consistent with electron scattering by spin fluctuations having a characteristic energy scale $T_{sf} \approx 150$ K (Y. Bang et al., cond-mat/0402685))
  - $T_{sf}(PuCoGa_5) \approx 10 T_{sf}(CeMIn_5)$; similar factor of 10 ratio in T_c's

Correlation of T_c with c/a in $CeM_{1-x}M'_xIn_5$ and $PuCo_{1-x}Rh_xGa_5$



- nearly identical $\partial \ln T_c / \partial (c/a) \approx 80$ in both $CeMIn_5$ and $PuMGa_5$ systems
- suggests a common underlying mechanism of superconductivity and its dependence on anisotropy
- substantial evidence from power laws in C/T, κ and $1/T_1$ below T_c as well as proximity to AFM that superconductivity in $CeMIn_5$ is unconventional
- similar electronic structures in both systems and low density of states in their $M(In/Ga)_2$ structural units ⇒ origin of correlation may be in distorted fcc structural units $CeIn_3/PuGa_3$

Local structure–superconductivity relationships



♦ $T_c \propto (2zc\text{-}a)/a \propto c/a$ for both $CeMIn_5$ and $PuMGa_5$ with nearly identical relative slopes

$\Rightarrow$ distortion of fcc units underlies $T_c \propto c/a$

♦ local structural distortion reflects, as in isostructural $UMGa_5$ and $NpMGa_5$, sensitivity of magnetic interactions on hybridization of f-electrons with M-atom electrons (K. Kaneko et al., PRB **68**, 214419 (2003); K. Kaneko et al, unpublished)

$\Rightarrow$ further support for common magnetic mechanism of superconductivity in Pu115 and Ce115 systems

Summary

♦ Much remains to be learned about the superconducting and normal states of these fascinating materials

• $CeMIn_5$: almost certainly unconventional superconductors; $PuMGa_5$: maybe unconventional

♦ For spin-mediated (unconventional) superconductivity, qualitatively $T_c \propto T_{sf}$

• T_{sf} ($PuCoGa_5$) ≈ 10T_{sf} ($CeCoIn_5$) and T_c ($PuCoGa_5$) ≈ 10T_c ($CeCoIn_5$) –consistent with expectations for magnetically mediated superconductivity in both as possibly is local structure-superconductivity relationships

♦ Plausible relation among $CeIn_3$ → $CeCoIn_5$ → $PuCoGa_5$

• $CeIn_3$ + layering = $CeCoIn_5$, T_c: 0.2 K → 2.3 K

• $CeCoIn_5$ + T_{sf} tuning = $PuCoGa_5$, T_c: 2.3 K → 18.5 K

♦ If unconventional superconductivity in $PuMGa_5$, as preliminary evidence suggests, T_c's are high compared to other examples, except cuprates; possible relationship?

♦ Fate of magnetic moments below T_c? (no magnetic order detected above 1 K ≈ 0.05T_c)

♦ Precise configuration of Pu's five 5f electrons? not obviously all localized or itinerant

♦ A new window for understanding the most complex element Pu and particularly its fcc phase δ-Pu, which is similar to the essential structural unit $PuGa_3$ in $PuMGa_5$

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