NMR Studies of Heavy Fermion Materials

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Outline

a) Introduction to NMR

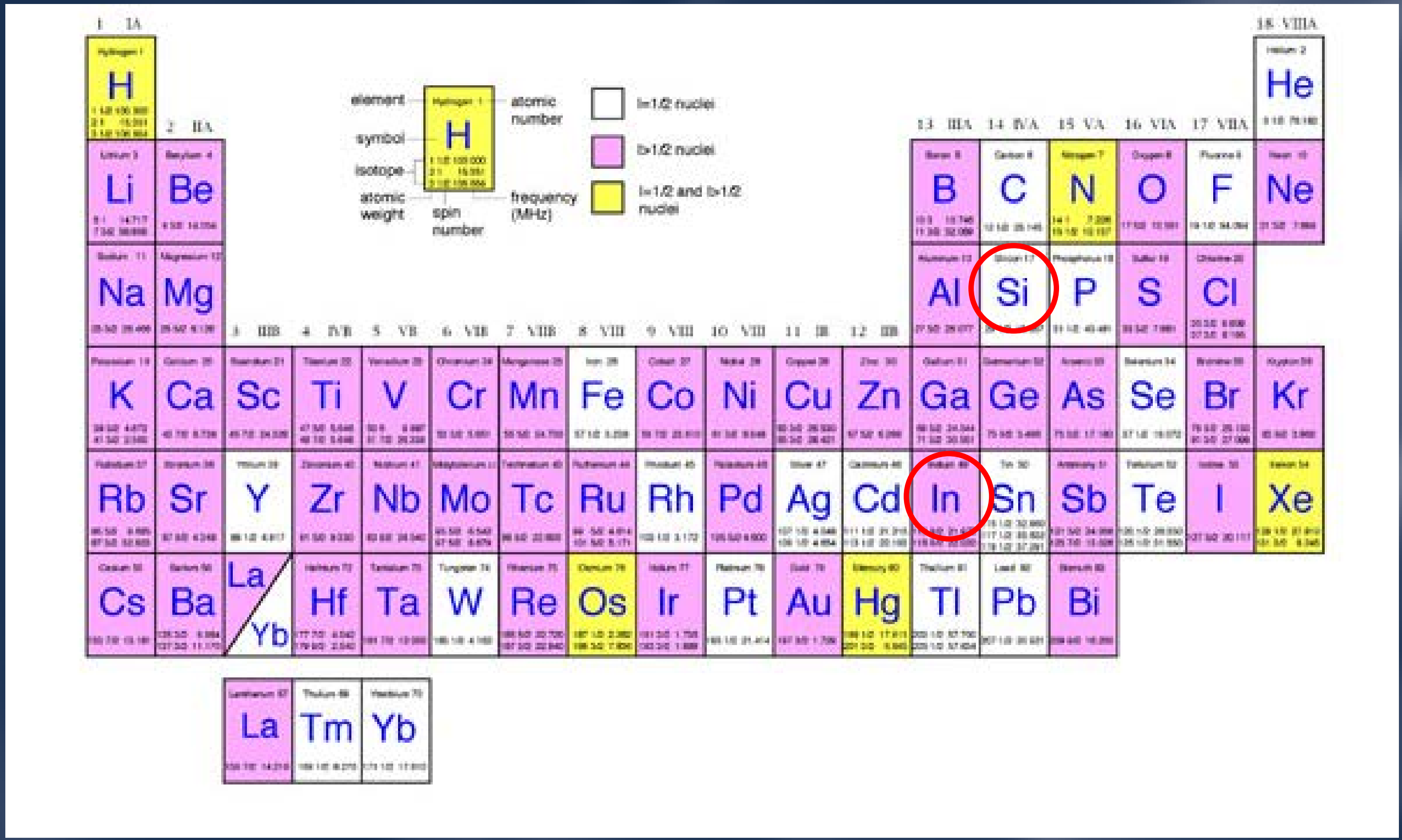
b) NMR in a Kondo Lattice

c) $CeMIn_5$ Compounds

d) URu_2Si_2

e) New Information from impurities

NMR and the Periodic Table



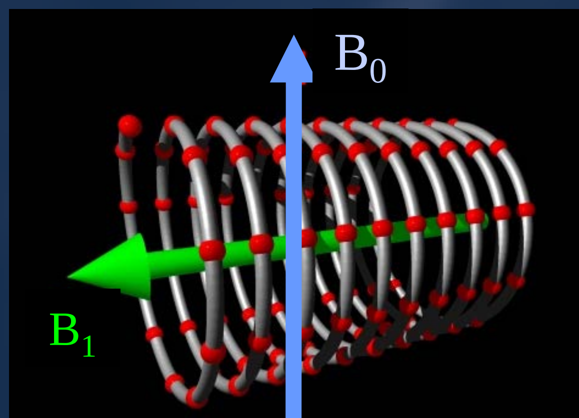
Practically every material has a nucleus with a spin

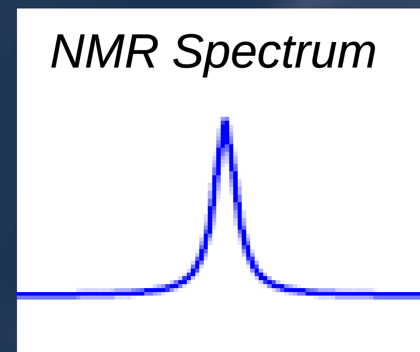
Probe spin and charge properties of the ***material*** *by observing the properties of the* ***nuclei***

Nuclear Spin Hamiltonian

$$\hat{\mathcal{H}}_{\rm nuc} = \hat{\mathcal{H}}_{\rm Z} + \hat{\mathcal{H}}_{\rm Q} + \hat{\mathcal{H}}_{\rm hf} + \hat{\mathcal{H}}_{\rm N-N}$$

Zeeman Interaction





frequency ~ MHz

$|I_z = +½\rangle$

$|I_z = -½\rangle$

B

$$\hat{\mathcal{H}}_{\rm Z} = g\mu_{\rm N}\hat{\mathbf{I}}\cdot\mathbf{B}$$

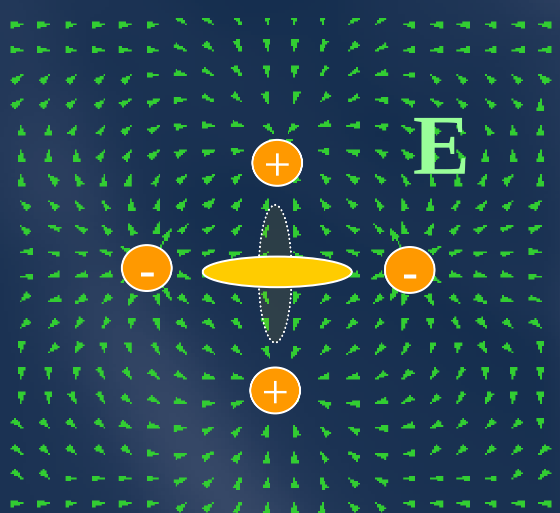
nuclear spin — magnetic field

$$\hbar\omega = g\mu_{\rm N}B$$

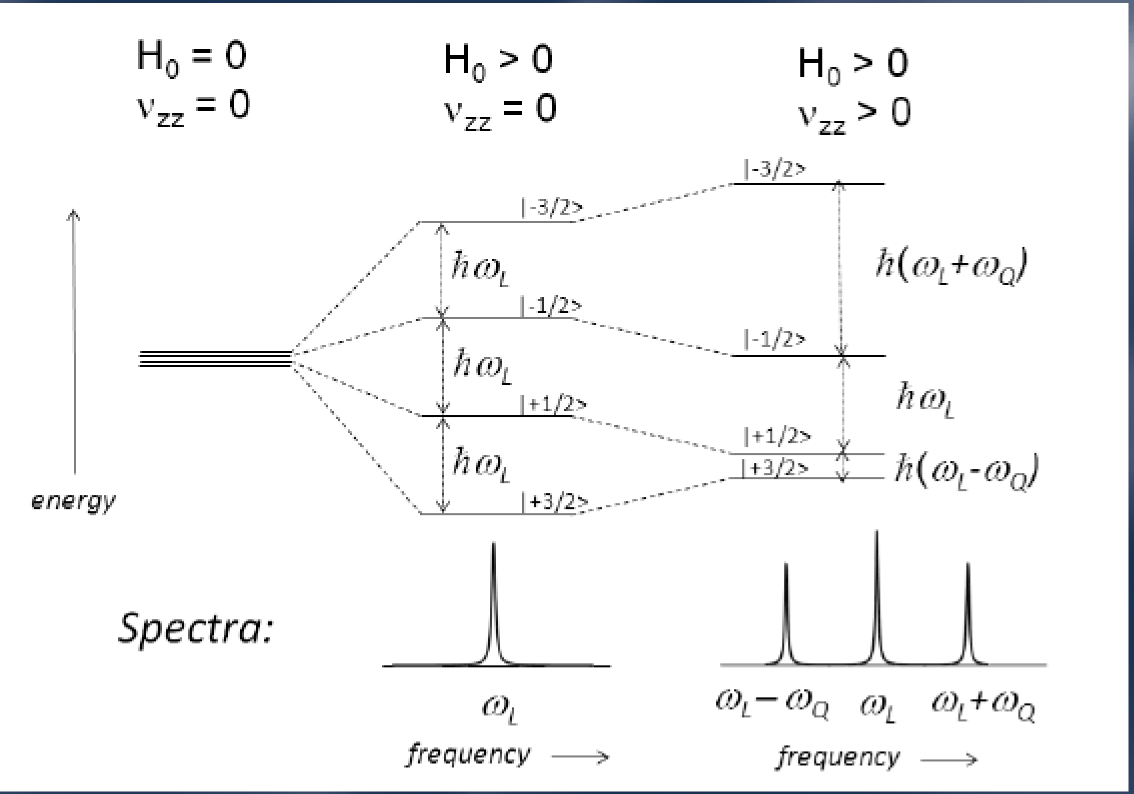
Quadrupolar Interaction

$$\mathcal{H}_Q = \frac{eQ}{6I(2I-1)} \sum_{\alpha\beta} V_{\alpha\beta} \left(\frac{3}{2} (\hat{I}_\alpha \hat{I}_\beta + \hat{I}_\beta \hat{I}_\alpha) - \delta_{\alpha\beta} \hat{I}^2 \right)$$

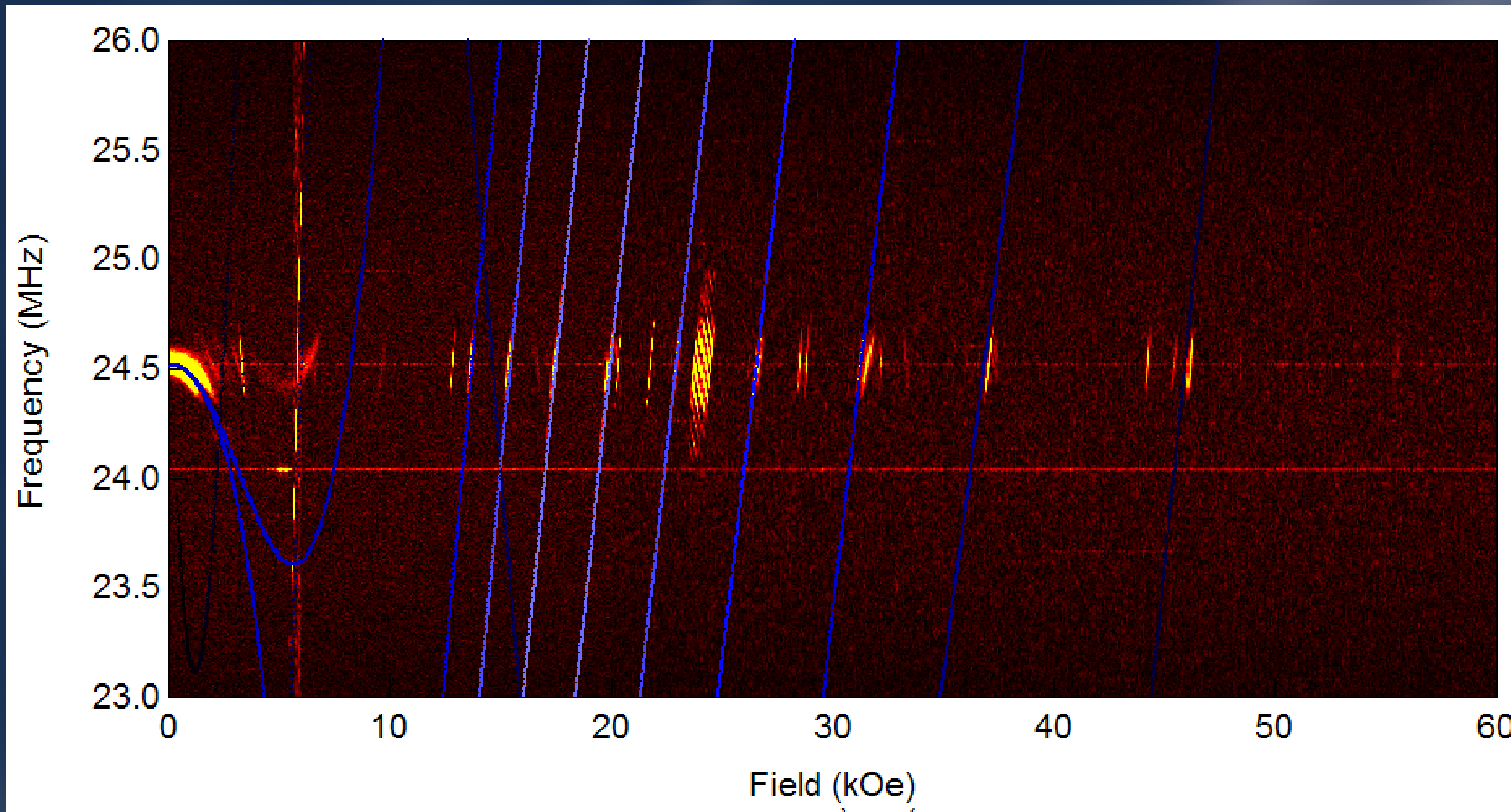
- Coupling between quadrupolar moment and **electric field gradient tensor**



- *Complex spectra for high spin nuclei*



Spectra: Quadrupolar Effects



^{115}In: $I=9/2$, large γ, large Q

$I=1/2$?

$f=\gamma_N H$

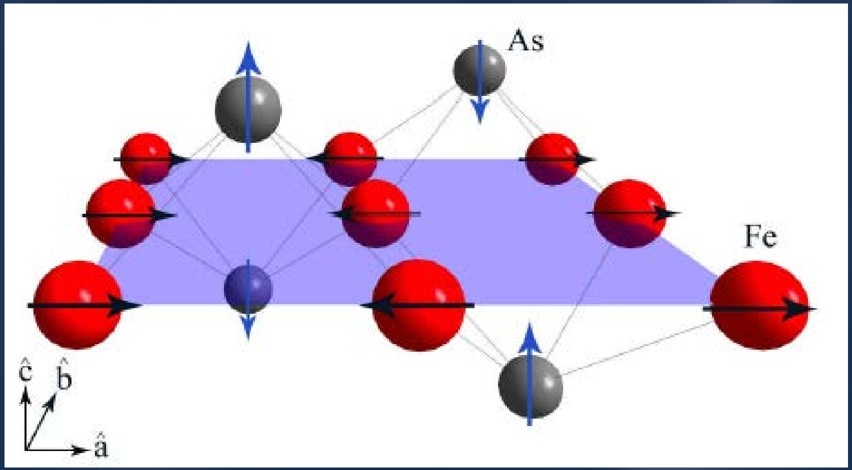
Extract EFG tensor in single crystals

Hyperfine Hamiltonian

•Most important interaction experienced by the nuclei

•Provides window onto behavior of electrons

$$\hat{\mathcal{H}}_{\mathrm{hf}} = \hat{\mathbf{I}} \cdot \mathbb{A} \cdot \hat{\mathbf{S}}$$



Transferred Hyperfine Coupling

• General case is a tensor coupling

Knight shift:

$$K = A\chi(\omega{=}0, \mathbf{q}{=}0)$$

Spin lattice relaxation:

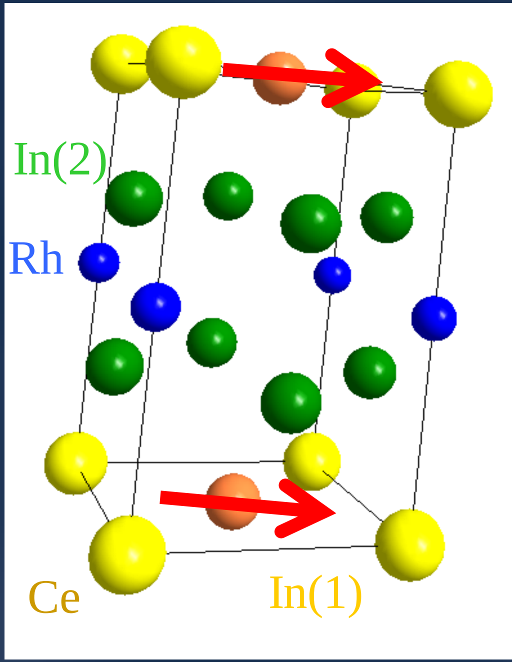
$$T_1^{-1} \sim A^2 \Sigma_{\mathbf{q}} \chi''(\omega, \mathbf{q})/\omega$$

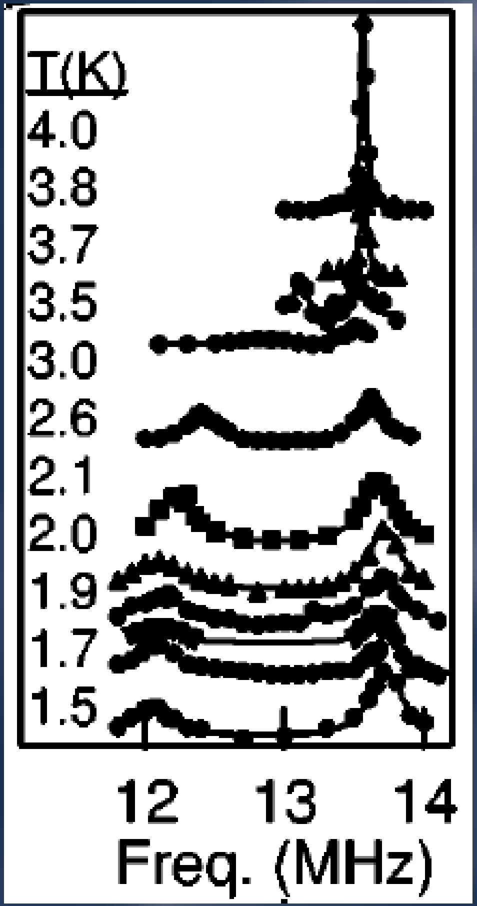
Static Internal Fields:

$$\mathrm{H}_{\mathrm{int}} = \Sigma_{\mathbf{i}}\, A S_{\mathrm{i}}$$

Internal Fields

$CeRhIn_5$



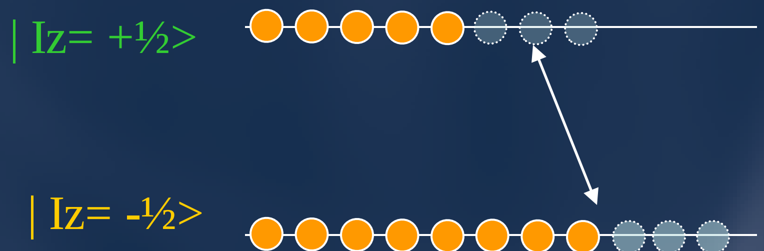


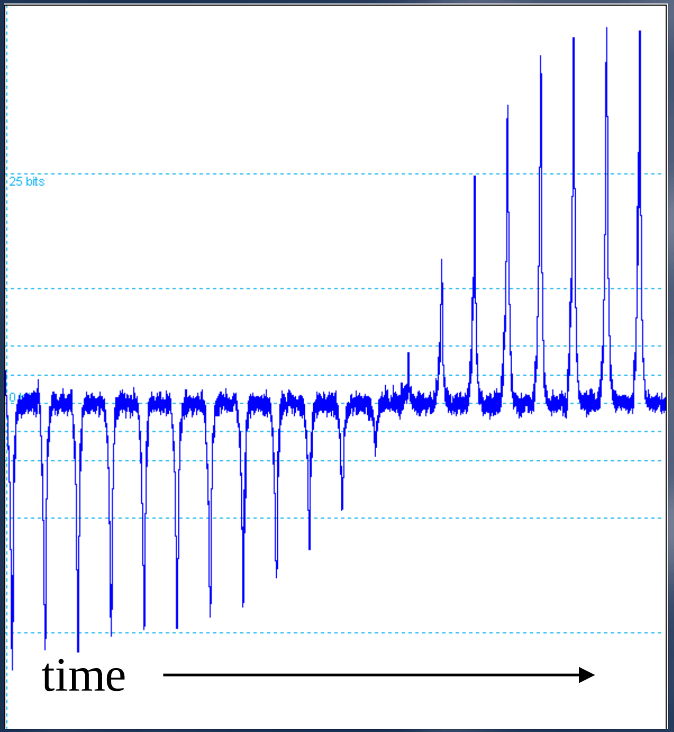
Curro et al., PRB, **2000**, 62 6100

Static internal magnetic fields from ordered electron spin moments

Antiferromagnetism, Ferromagnetism, SDW, etc.

Nuclear Spin Dynamics





By applying rf pulses, we can perturb the equilibrium Boltzmann distribution, and then watch as the system relaxes to a finite spin temperature

T_1 is the characteristic relaxation time, and it is determined by the dynamical electron spin susceptibility:

$$T_1^{-1} = \gamma^2 k_B T \lim_{\omega \to 0} \sum_{\boldsymbol{q}} A^2(\boldsymbol{q}) \frac{\chi''(\boldsymbol{q}, \gamma)}{\hbar\omega}$$

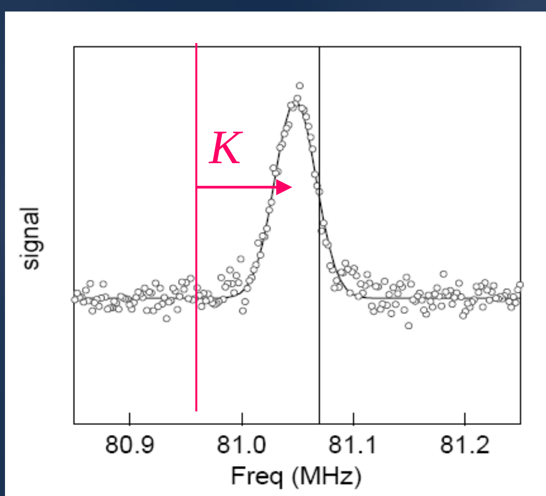
The Knight Shift

$$\mathcal{H} = \gamma\hbar\hat{\mathbf{I}}\cdot\mathbf{H}_0 + \hbar\hat{\mathbf{I}}\cdot\mathbf{A}\cdot\hat{\mathbf{S}} + g\mu_B\hat{\mathbf{S}}\cdot\mathbf{H}_0$$

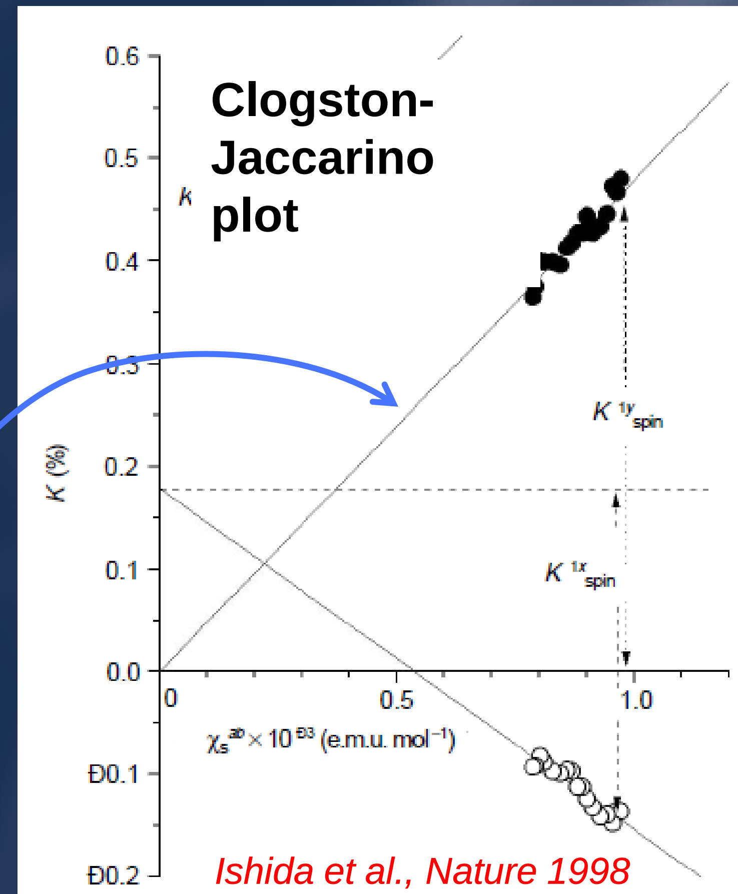
Nuclear spin — Hyperfine coupling — Electron spin

$$\hat{\mathbf{S}} \to \langle\hat{\mathbf{S}}\rangle = \chi\mathbf{H}_0/g\mu_B$$

$$\mathcal{H} \to \mathcal{H}_{\text{eff}} = \gamma\hbar\hat{\mathbf{I}}\cdot(\mathbf{1}+\mathbf{K})\cdot\mathbf{H}_0$$

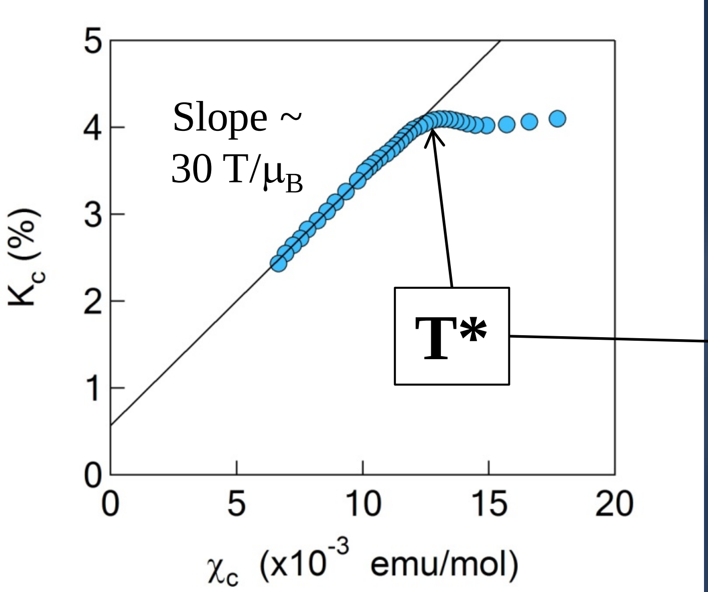


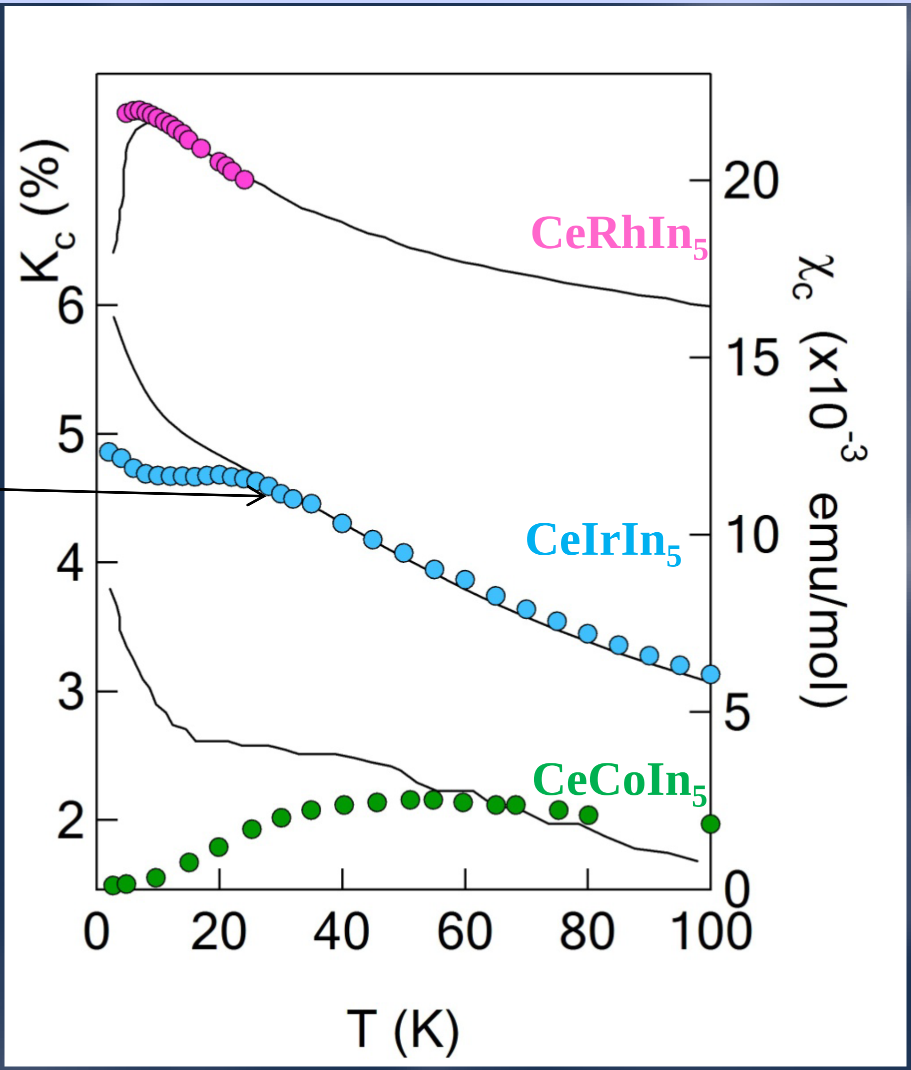
Slope = A



A usually measured in field/moment (~ T/μ_B) or energy units ($\gamma\hbar\mu_B A$ ~ $0.1\,\mu eV$)

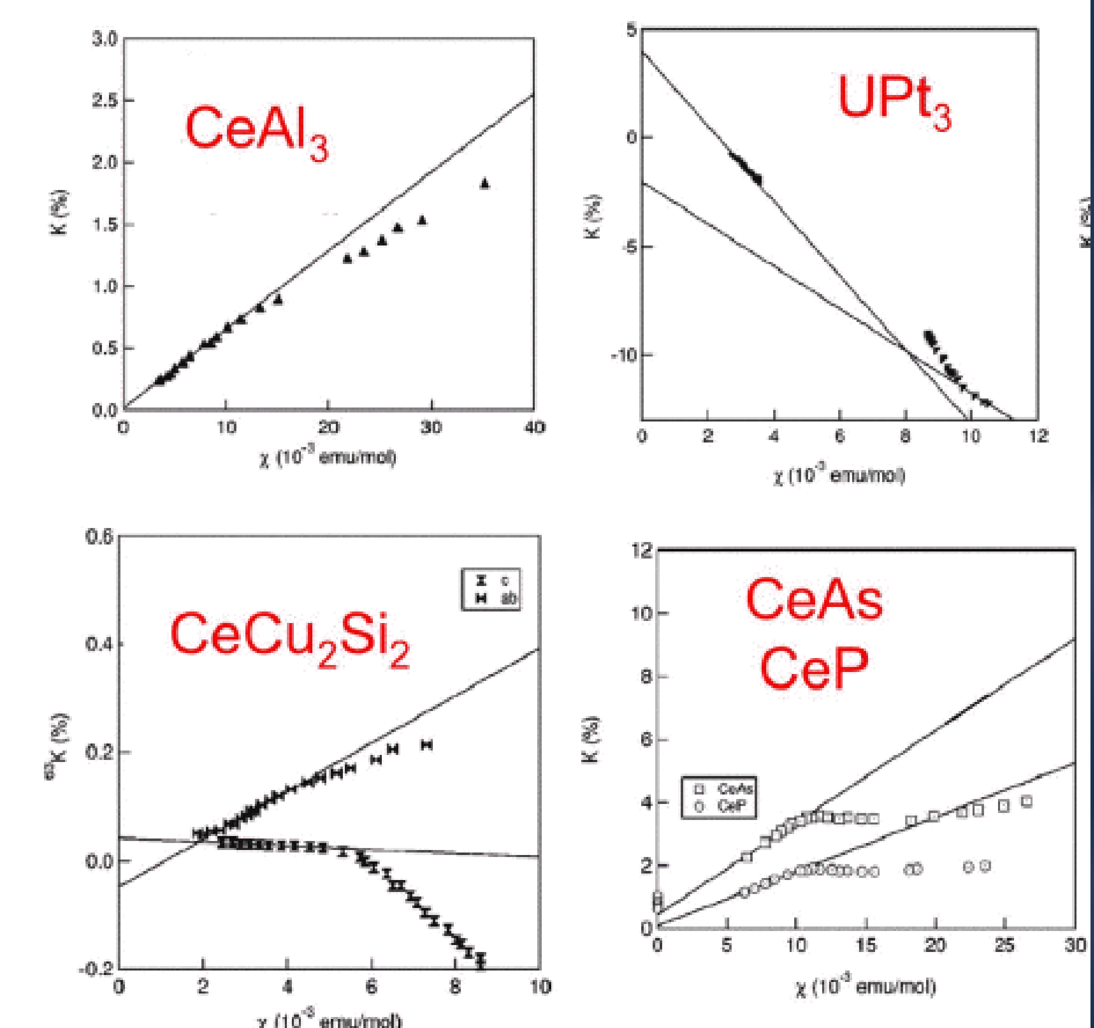
Knight Shift Anomalies





Breakdown of linear relationship below coherence temperature T*

Other Examples



Curro et al., PRB 70, 135117 (2004)

adapted from various sources – original data found in:

M. Lysak and D. MacLaughlin, Phys. Rev. B 31, 6963 (1985)

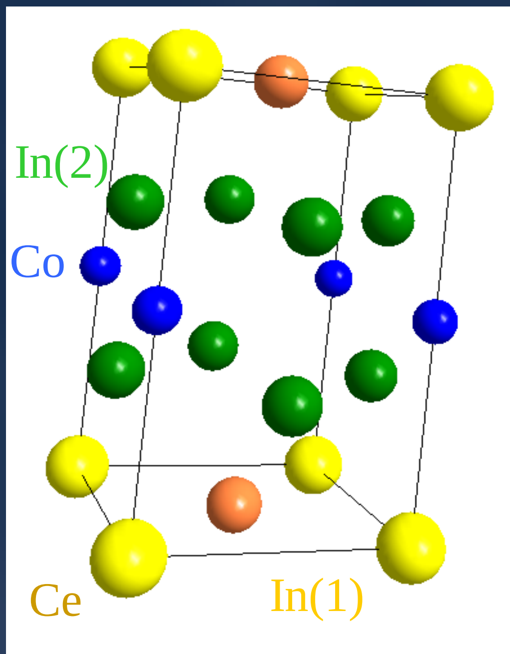
M. Lee, G. Moores, Y.-Q. Song, W. Halperin, W. Kim, and G. Stewart, Phys. Rev. B 48, 7392 (1993)

T. Ohama, H. Yasuoka, D. Mandrus, Z. Fisk, and J. L. Smith, J. Phys. Soc. Jpn. 64, 2628 (1995)

S. Myers and A. Narath, Solid State Commun. 12, 83 (1973)

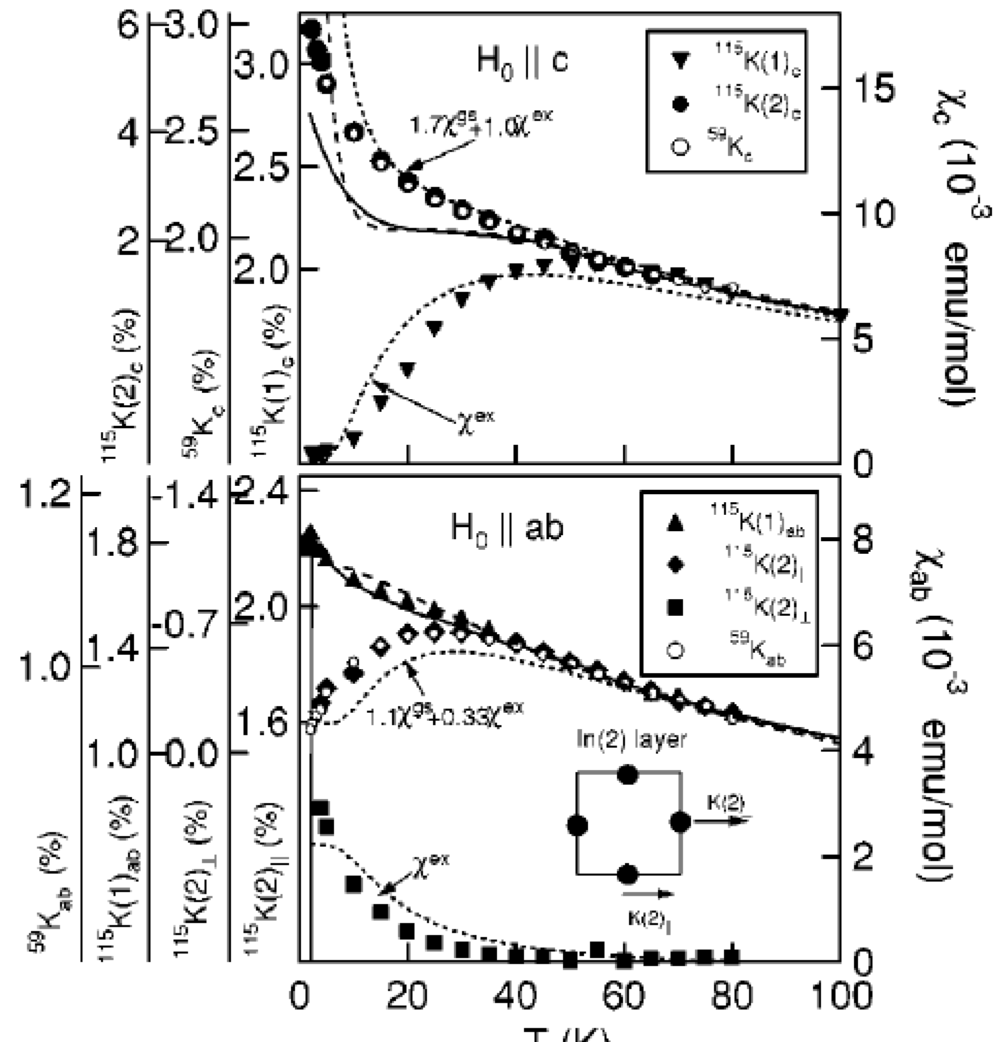
Anisotropy and Site Dependence

$CeCoIn_5$



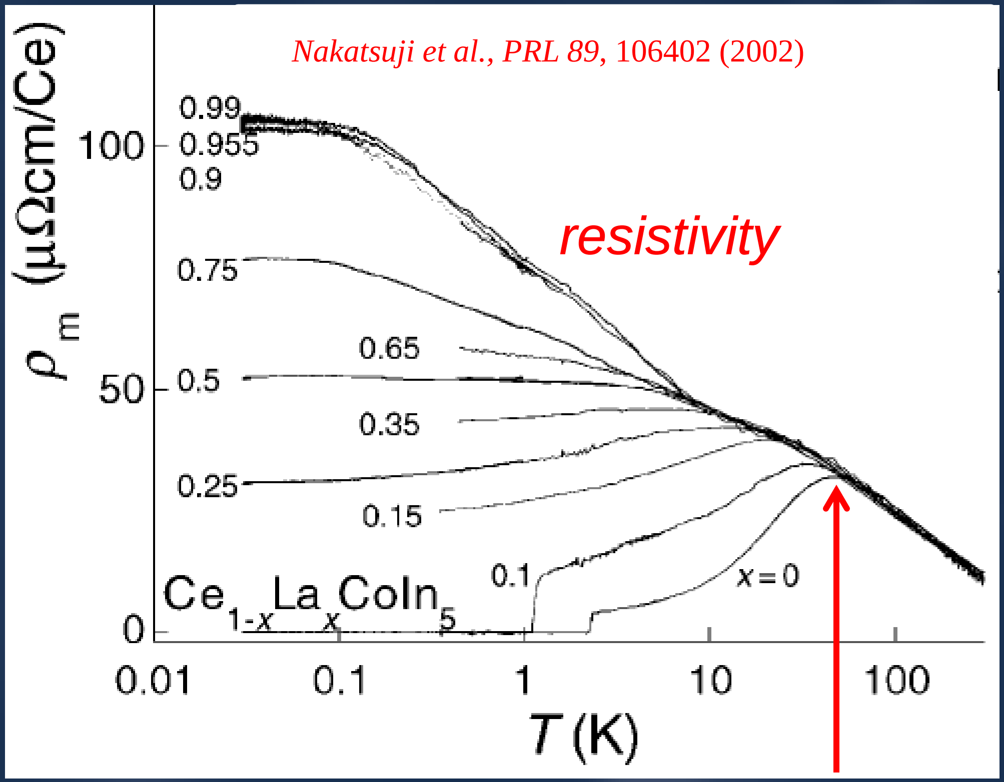
Different behaviors at different sites from relative sign of *A* vs *B*

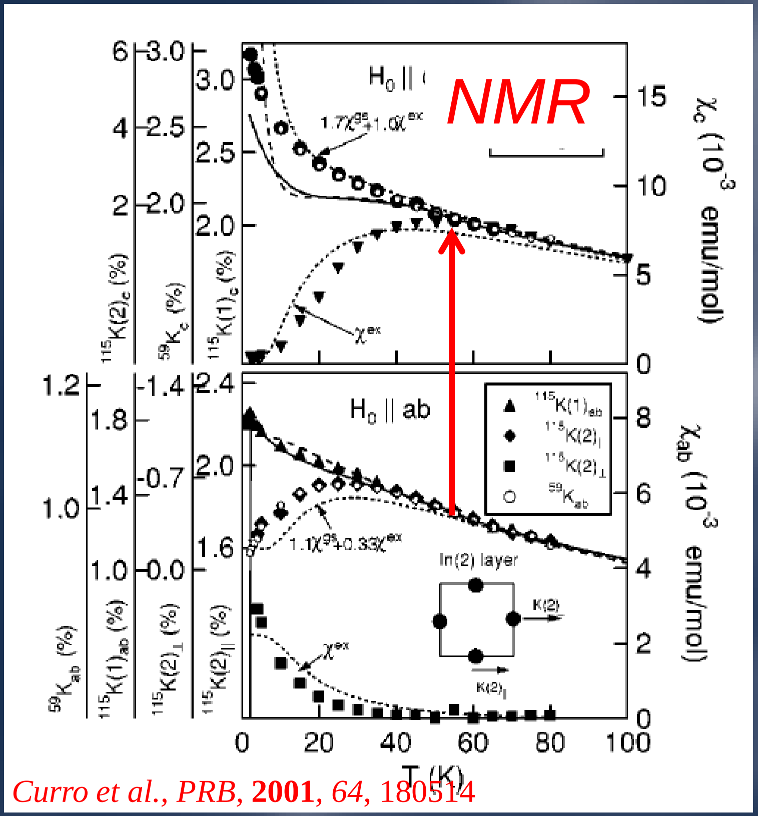
T is anisotropic*



Curro et al., PRB, **2001**, *64,* 180514

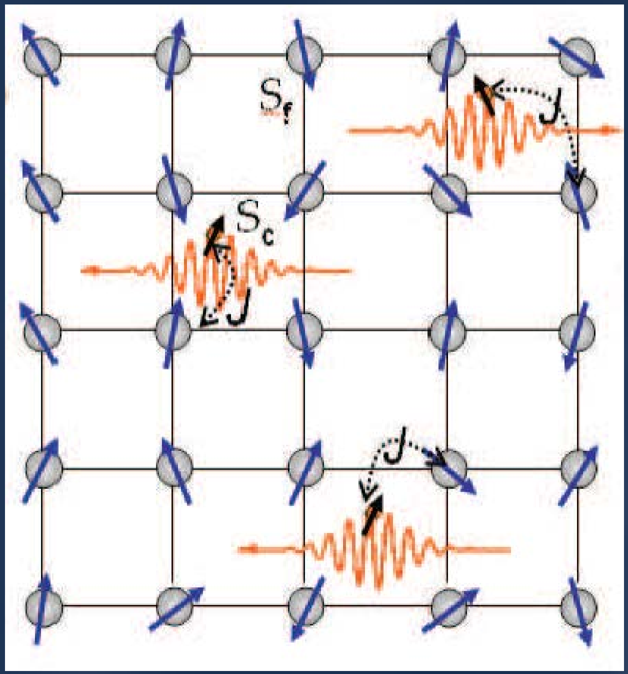
Spin vs Charge Signatures

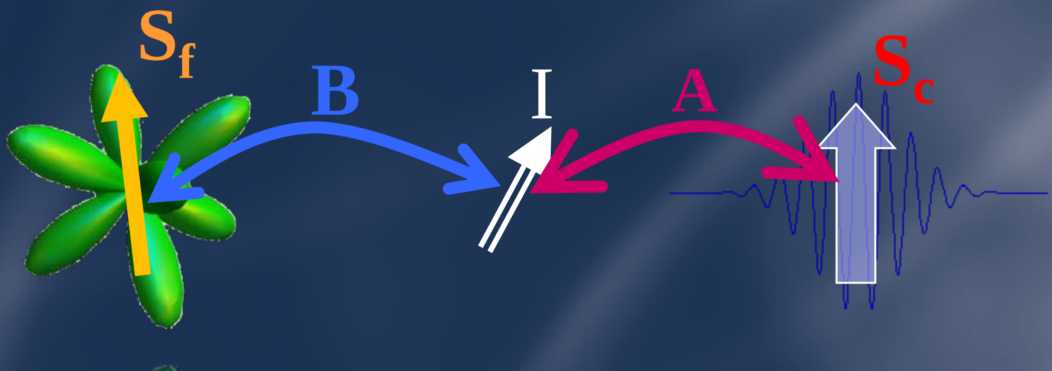




T matches the onset of coherence*
Seen also in several other experimental probes
(Hall effect, tunneling, etc.)

Kondo Lattice Hyperfine Couplings





$$\mathcal{H}_{\text{hyp}} = \hat{\mathbf{I}} \cdot \mathbb{A} \cdot \mathbf{S}_c + \hat{\mathbf{I}} \cdot \mathbb{B} \cdot \mathbf{S}_f$$

- Two types of transferred hyperfine couplings
- Breakdown of linear K-χ relationship
- New window onto correlation functions
- Measures the onset of heavy fermion coherence

Correlation Functions

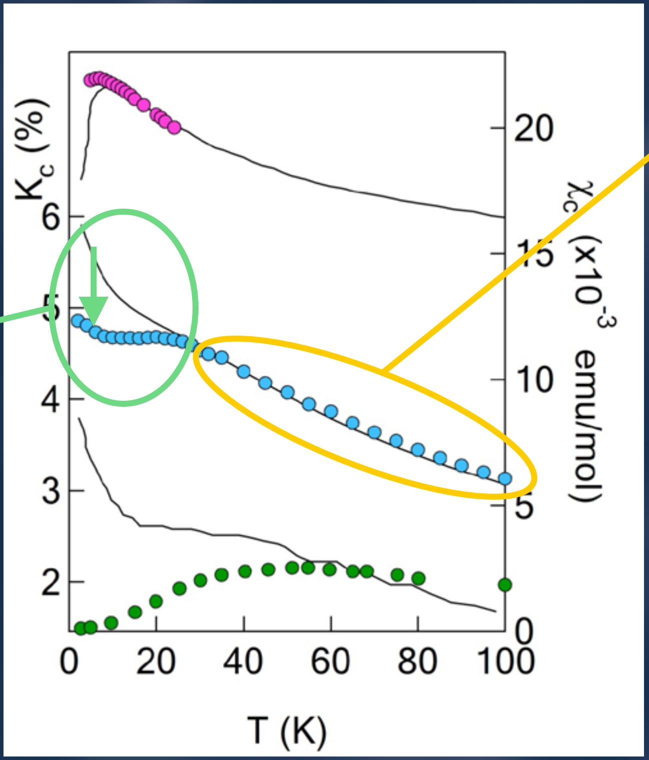
$$\chi_{cc} = \langle S_c S_c \rangle$$
$$\chi_{cf} = \langle S_c S_f \rangle$$
$$\chi_{ff} = \langle S_f S_f \rangle$$

$$K = \frac{A}{\gamma\hbar}\chi_{cc} + \frac{A+B}{\gamma\hbar}\chi_{cf} + \frac{B}{\gamma\hbar}\chi_{ff}$$

$$\chi = \chi_{cc} + 2\chi_{cf} + \chi_{ff}$$

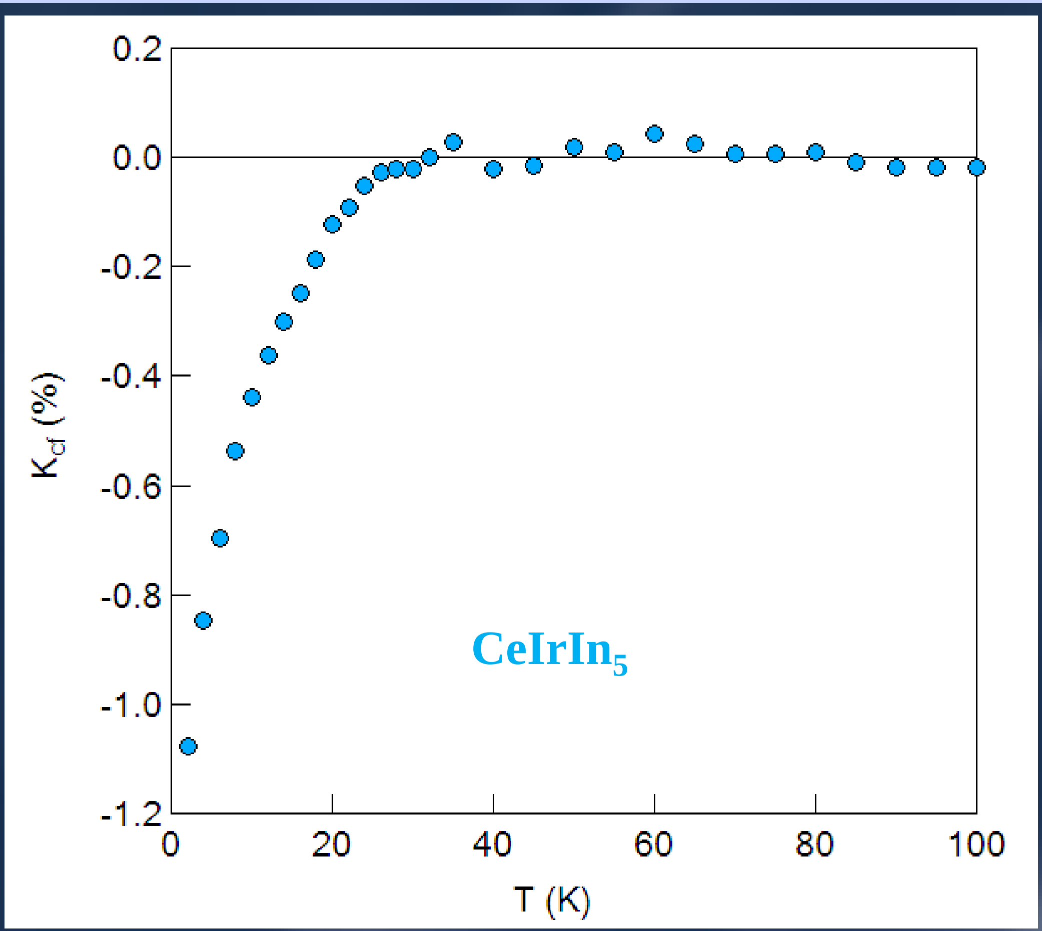
χ_{cf} grows below T^*

$K_{cf} = K - B\chi$



χ_{ff} dominates at high T, so $K \approx B\chi$

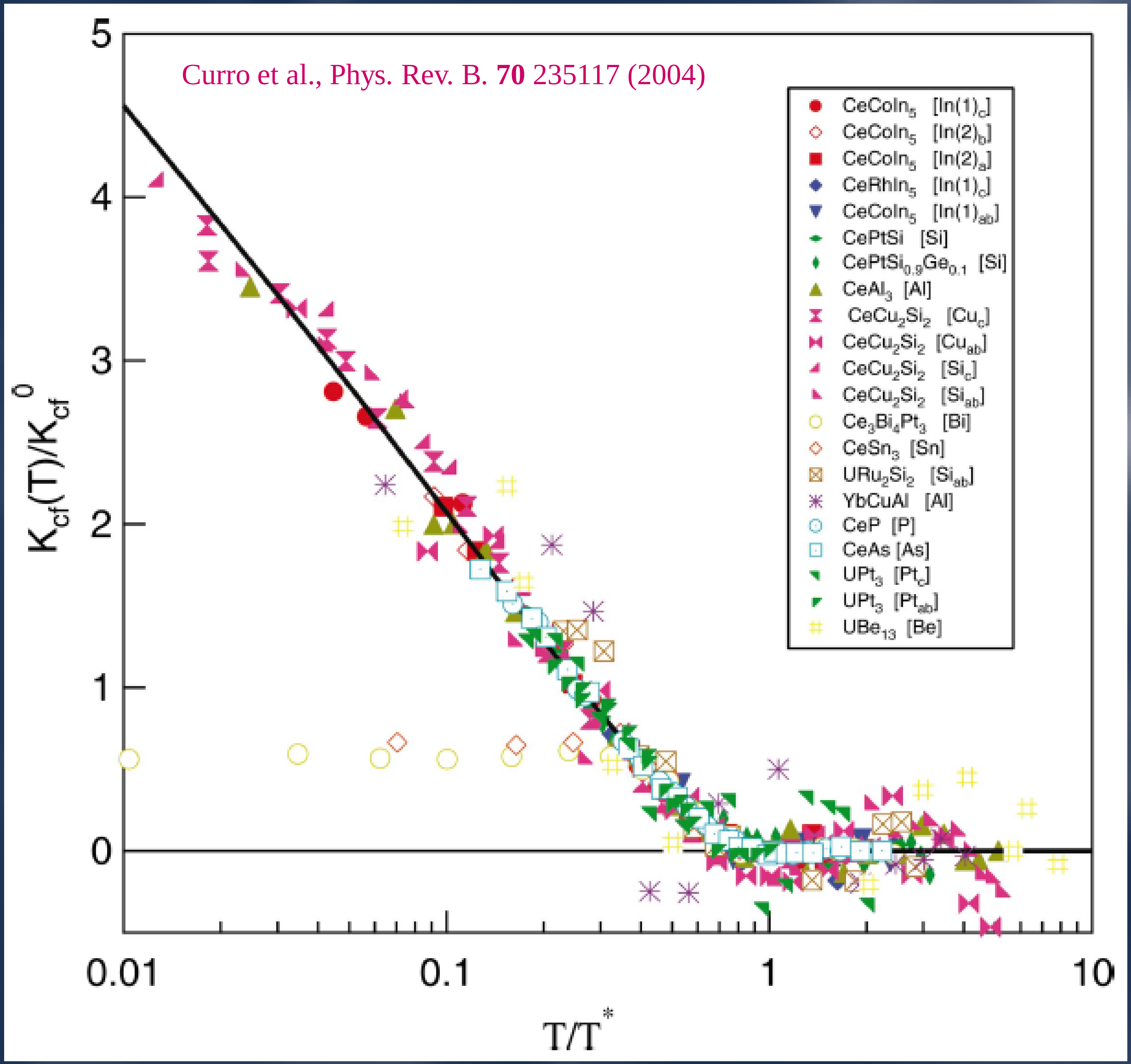
K_{cf} versus T



$$K_{cf} = K - B\chi$$
$$= (A - B)(\chi_{cf} + \chi_{cc})$$

Scaling Behavior

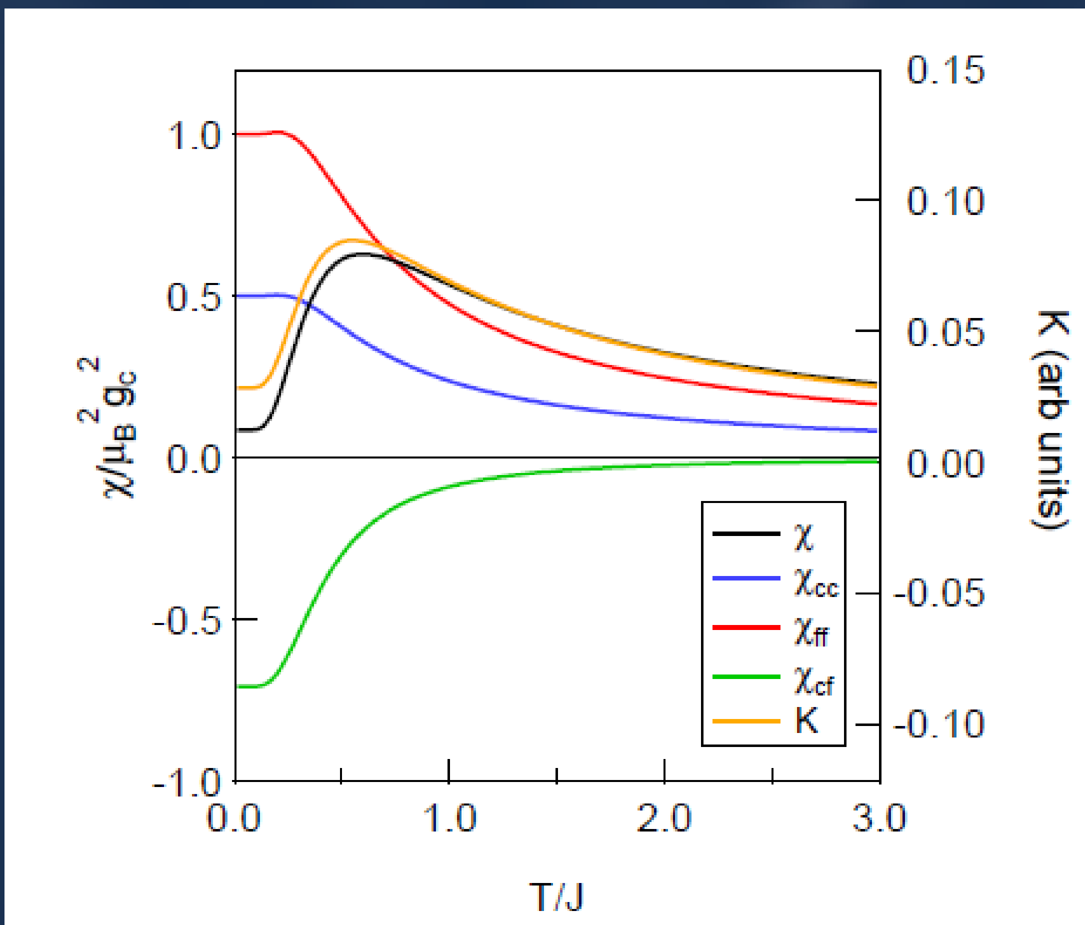
$K_{cf} = (A\text{-}B)\, \chi_{cf}$ scales with T/T^* universally among the heavy fermion compounds

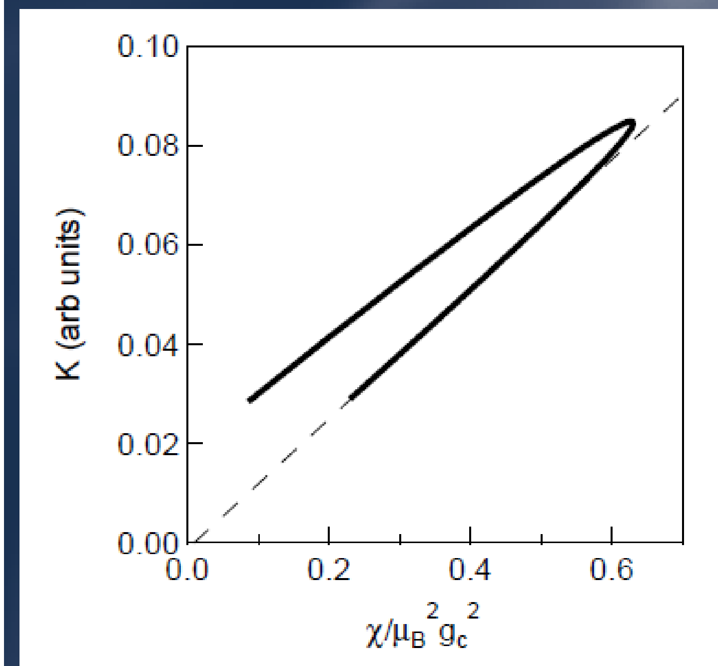


Toy Model

Assume two free spins, S_c and S_f that are coupled by J (exactly solvable)

$$\hat{\mathcal{H}} = g_c \mu_B \mathbf{S_c} \cdot \mathbf{H} + g_f \mu_B \mathbf{S_f} \cdot \mathbf{H} + J\mathbf{S}_c \cdot \mathbf{S}_f$$





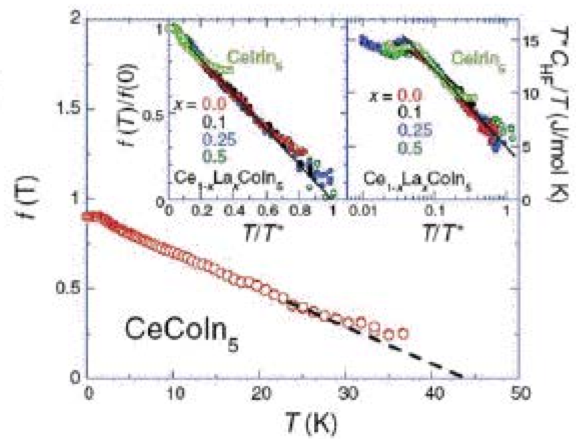
Two Fluid Interpretation

Nakatsuji, Pines & Fisk [PRL **92** 16401 (2004)] -Analysis of dilute $CeCoIn_5$ via two fluids

$$\chi(T) = [1 - f(T)]\chi_{\mathrm{KI}}(T) + f(T)\chi_{\mathrm{HF}}(T)$$

$$C_{\mathrm{MAG}}/T = [1 - f(T)](C_{\mathrm{KI}}/T) + f(T)(C_{\mathrm{HF}}/T),$$

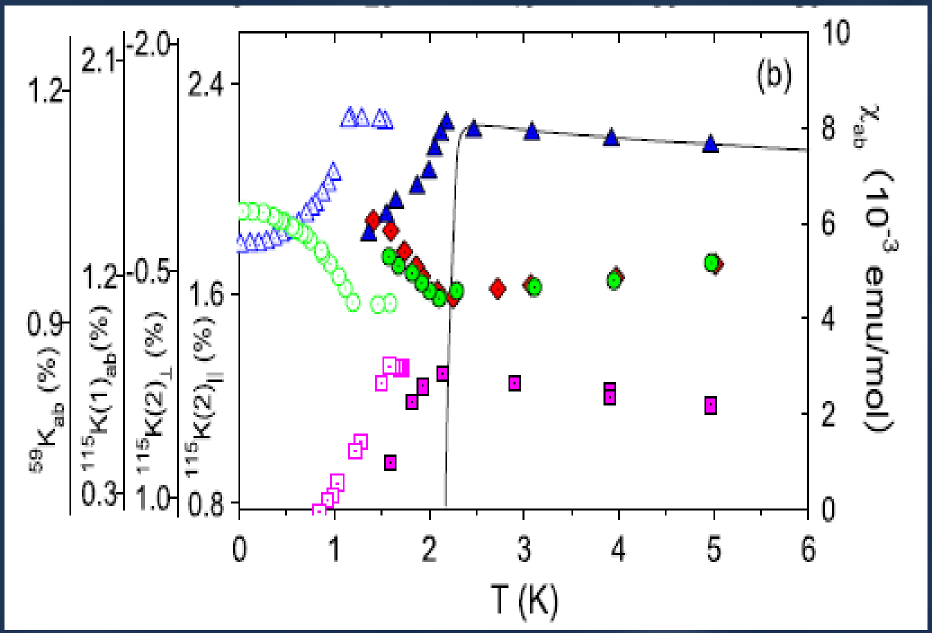
Second "heavy fermion" component emerges below T^*

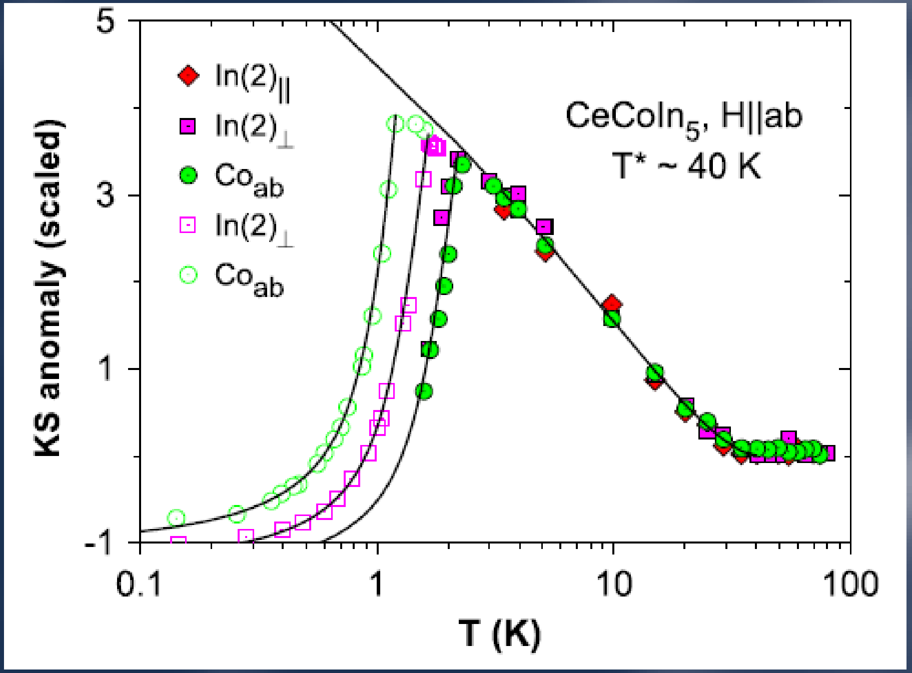


- Identify χ_{cf} with χ_{HF}
- Temperature dependence: $\chi_{cf} \sim (1-T/T^*)^{3/2}[1+\ln(T^*/T)]$
- Yang and Pines, Nature (2008),

  Phys. Rev. Lett. (2008)

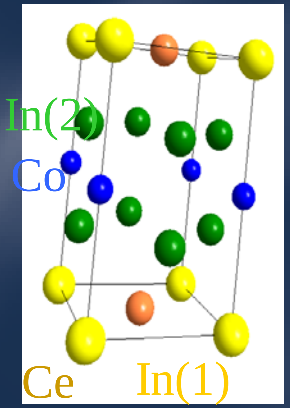
Behavior at Superconducting Transition





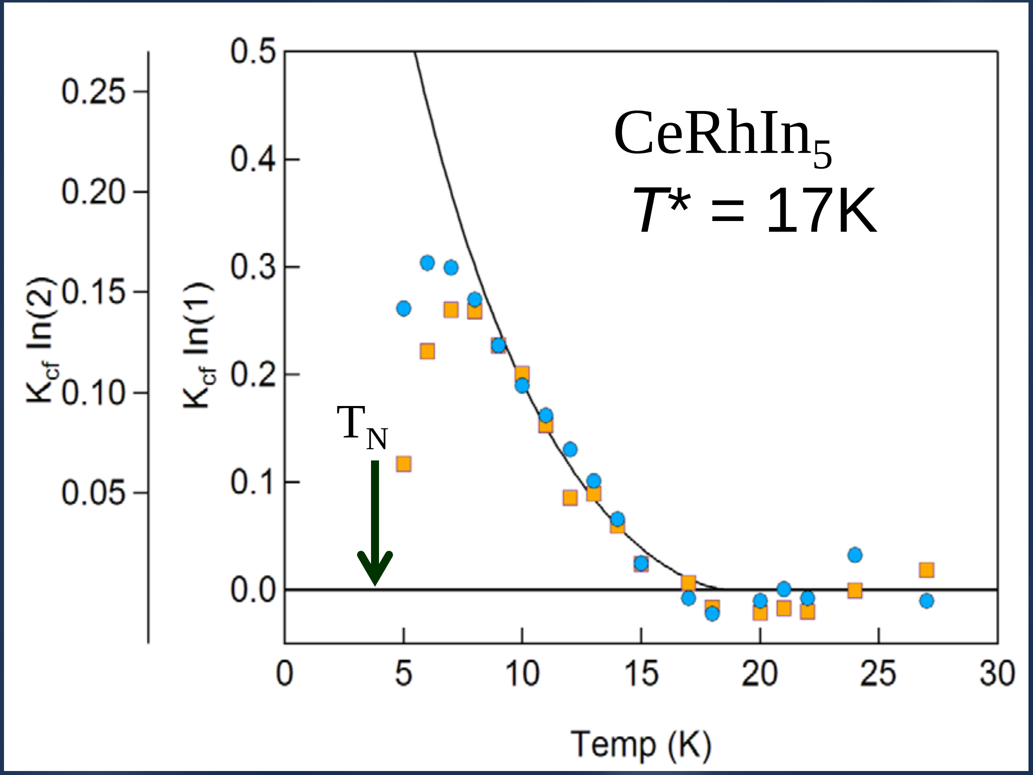
Yang et al., PRL, 103, 19700 (2009)

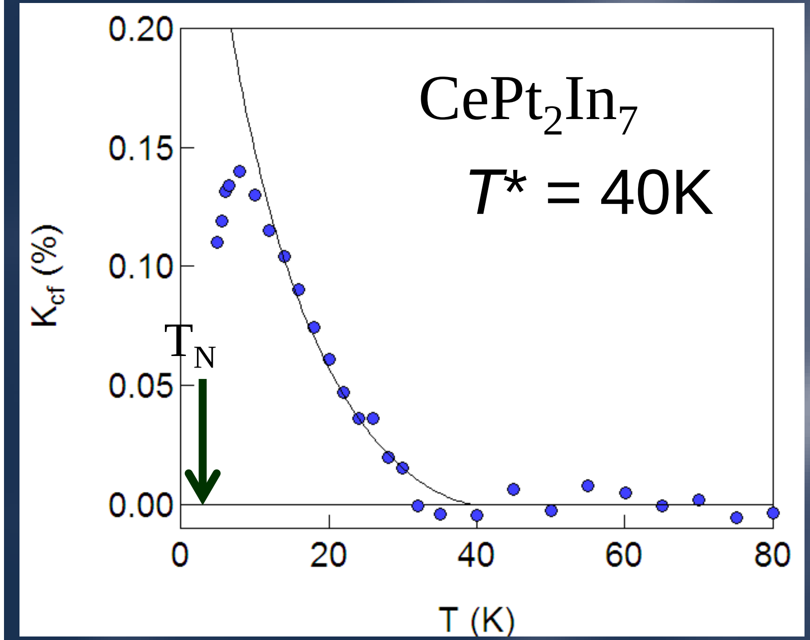
Superconductivity - $CeCoIn_5$



- Scaling persists down to T_c
- Spin singlet superconductivity emerges in heavy electron fluid

Behavior at AFM Transition

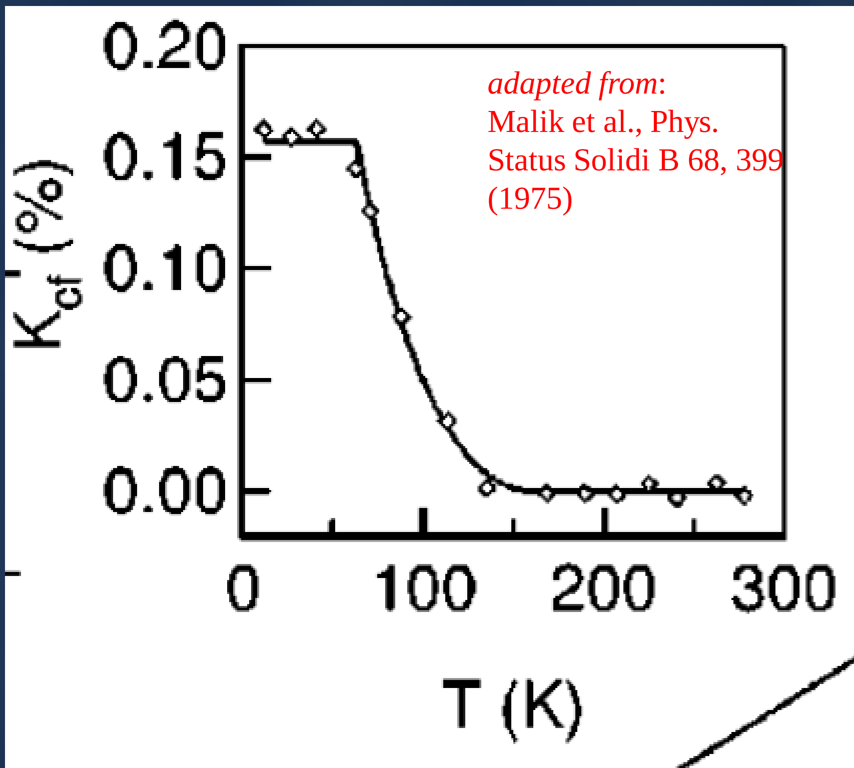




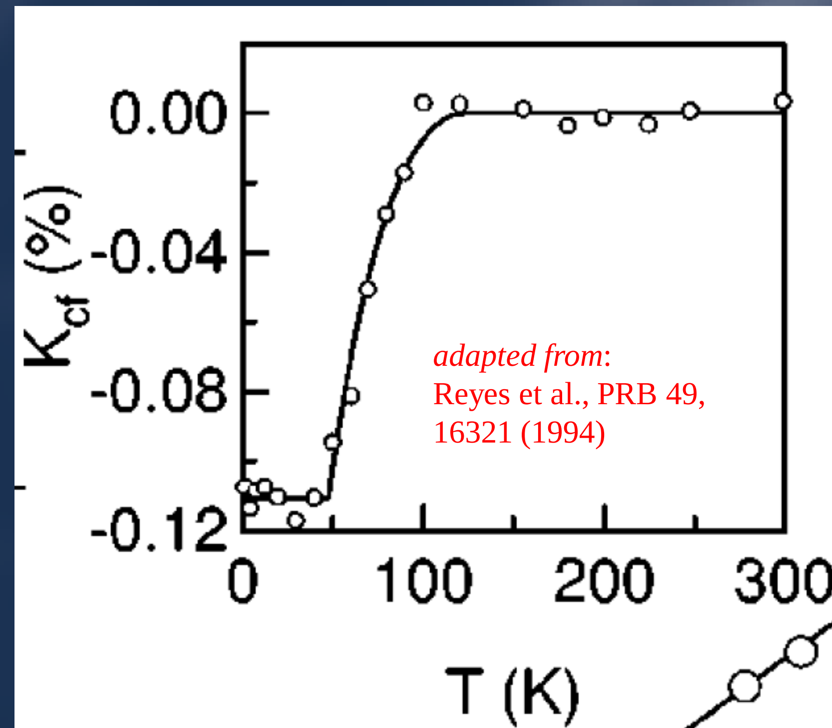
Antiferromagnetism - $CeRhIn_5$, $CePt_2In_7$

- Scaling breaks down prior to onset of long range magnetic order
- Local moment portion gains weight at expense of heavy electron component

Other Behaviors



Mixed valent - $CeSn_3$

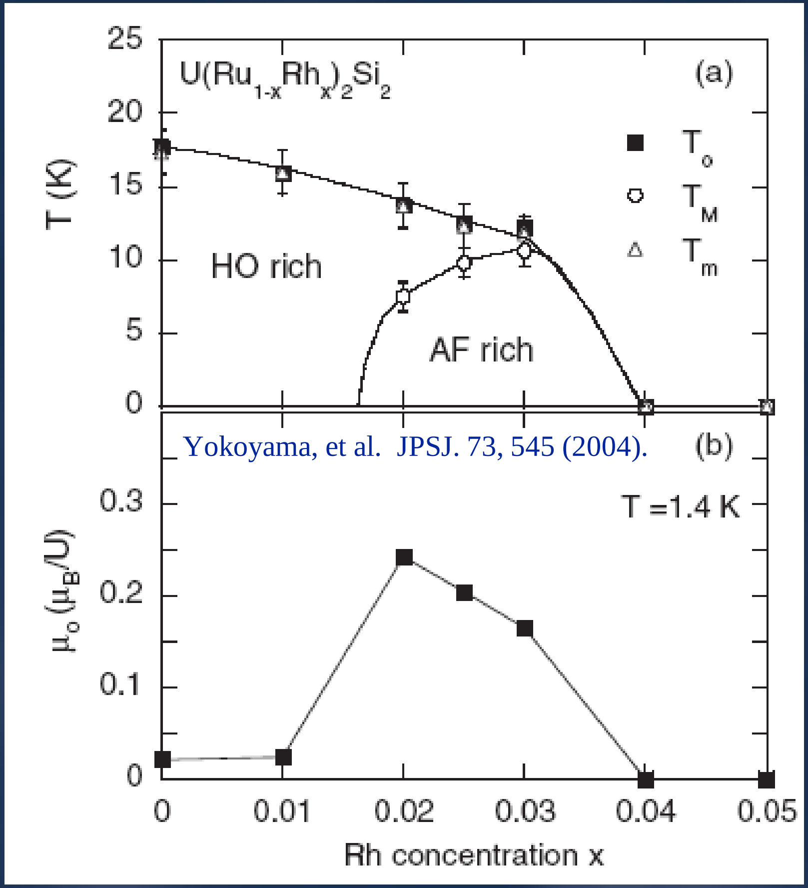


Kondo Insulator – $Ce_3Bi_4Pt_3$

In these cases χ_{cf} appears to saturate around 0.5T^*

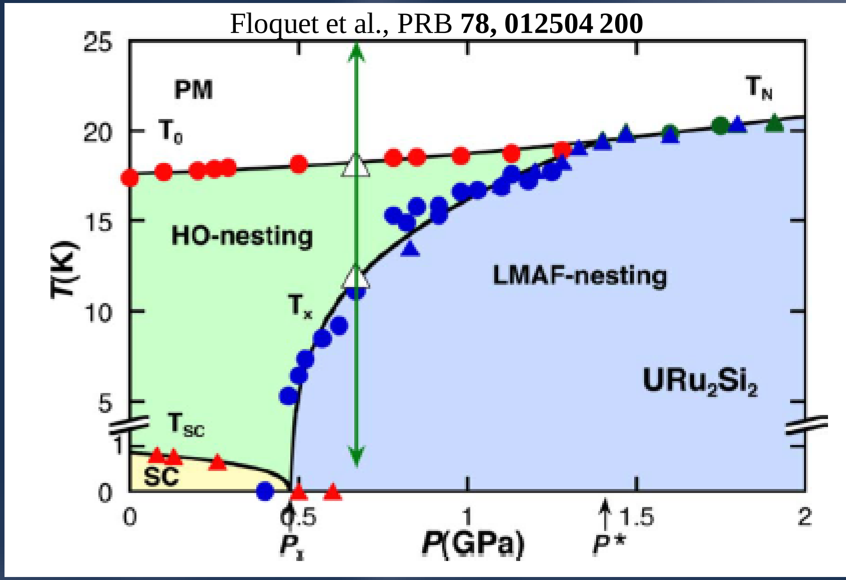
Curro et al., Phys. Rev. B. **70** 235117 (2004)

Rh doping



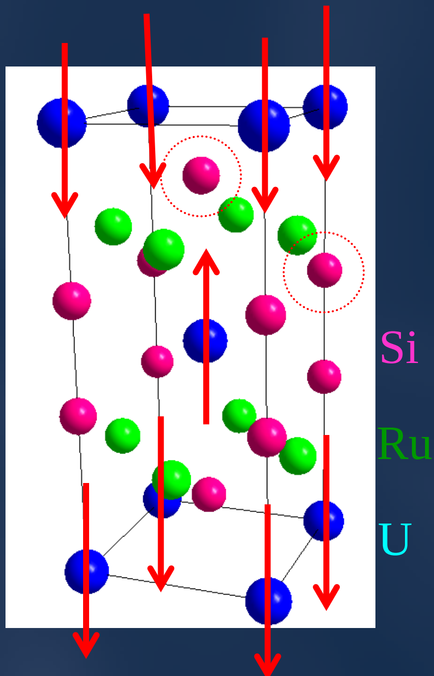
Rh doping gives rise to a phase of antiferromagnetism

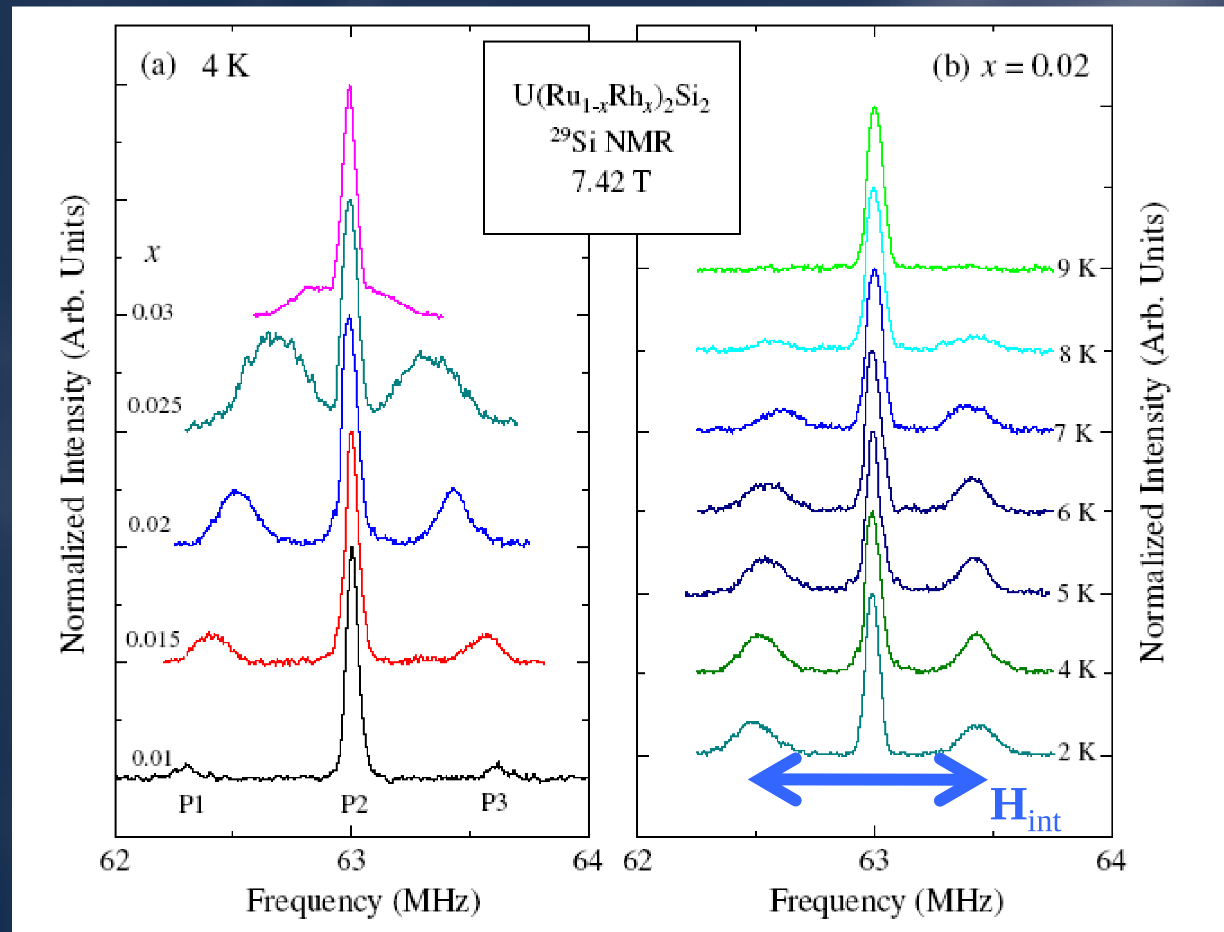
Coexistence/competition of two order parameters?



What can NMR tell us about this antiferromagnetism?

Si NMR Spectra

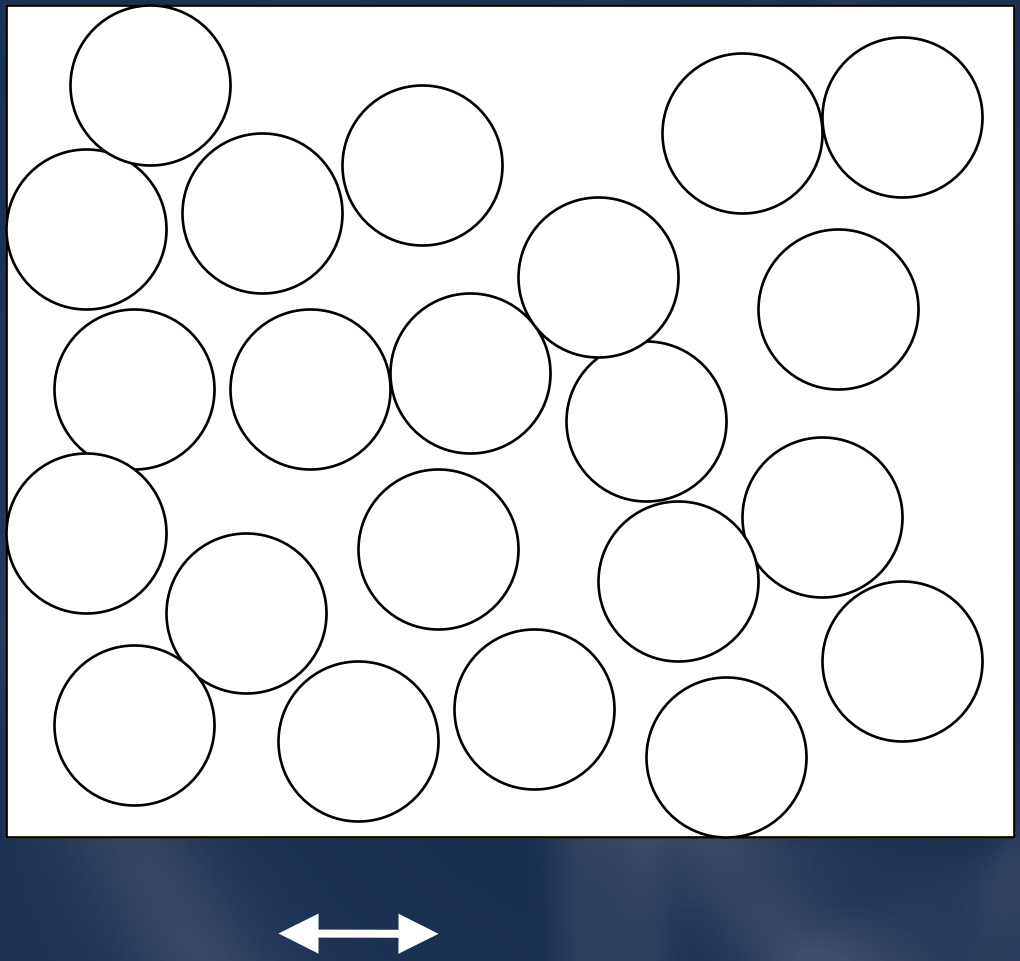




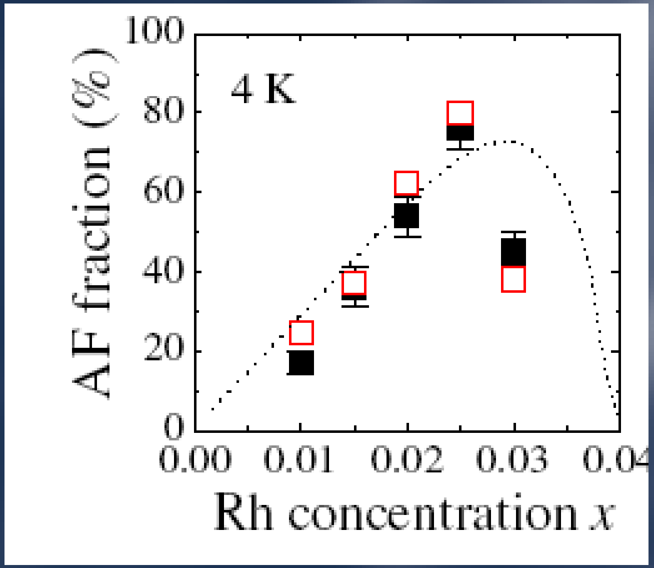
Baek et al., Phys. Rev. B 81, 132404 (2010)

Inhomogeneous coexistence of **two components**: antiferromagnetism and paramagnetic

Antiferromagnetic nanobubbles



~ 2nm

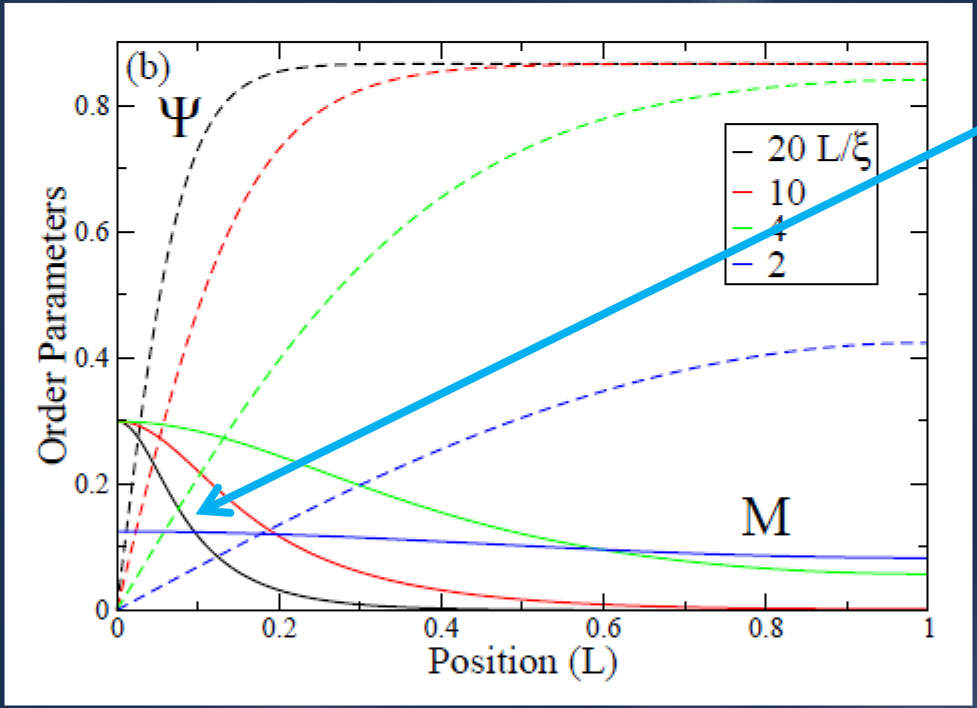


Ginsburg-Landau Analysis

$$F[\Psi, \mathbf{M}] = F_{HO} + F_{AF} + F_C$$

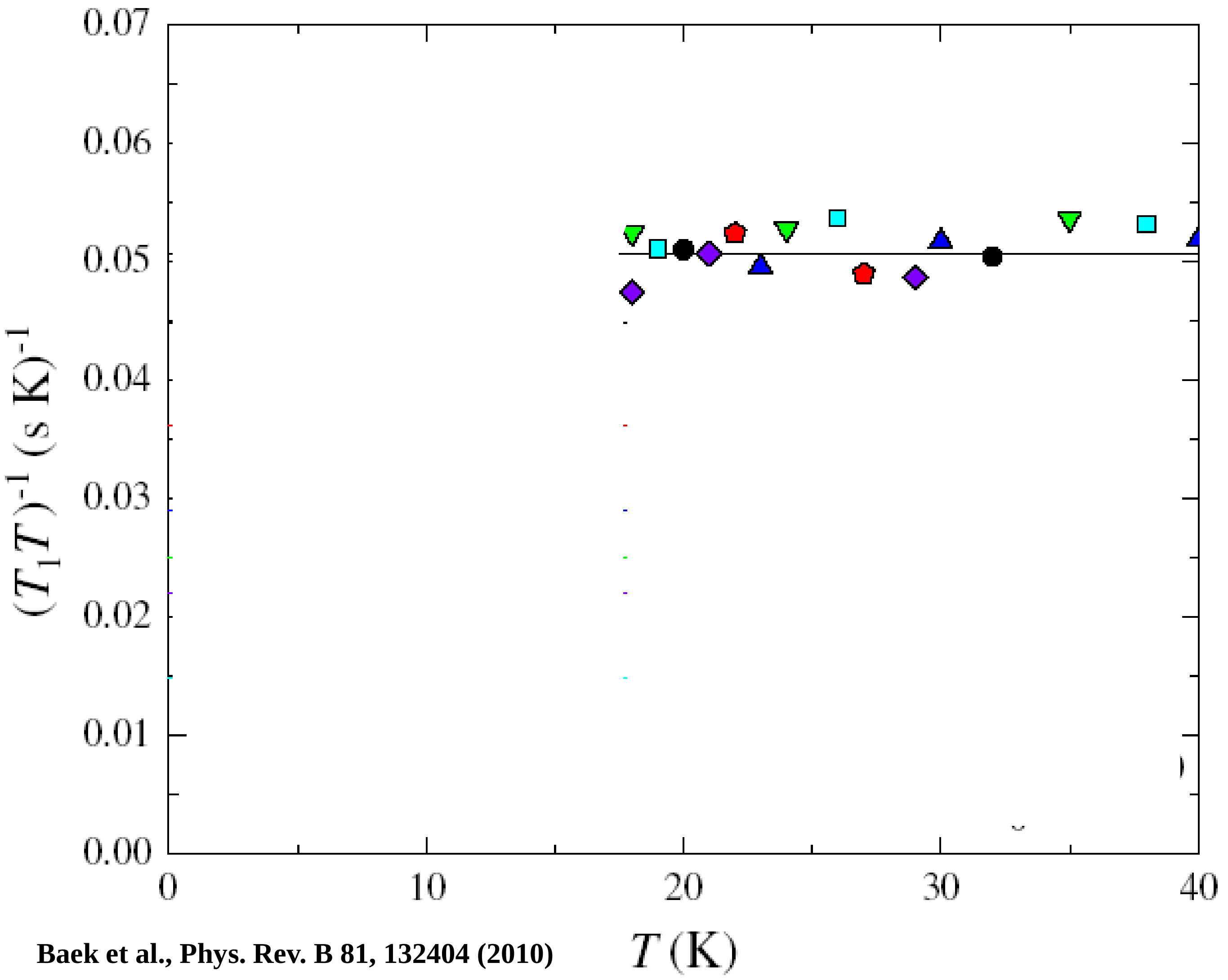
$$F_C[\Psi, \mathbf{M}] = g_1 \Psi^2 |\mathbf{M}|^2 + g_2 |\mathbf{M}|^2 |\nabla\Psi|^2 + g_3 |\mathbf{M} \cdot \nabla\Psi|^2$$

Gradient coupling terms lead to nucleation of inhomogeneous AF order around impurity site

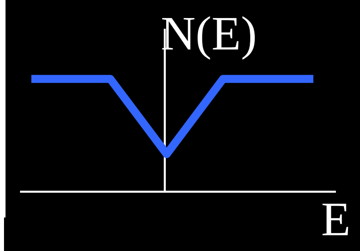


Baek et al., Phys. Rev. B 81, 132404 (2010)

Filling of the energy gap

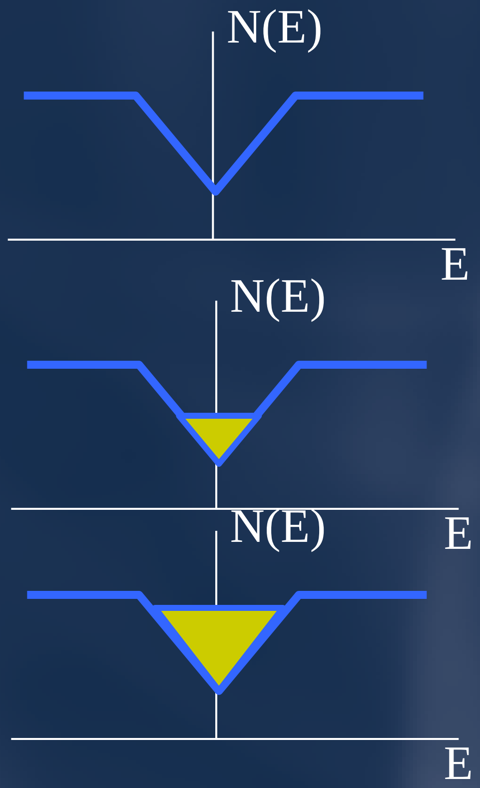


Baek et al., Phys. Rev. B 81, 132404 (2010)

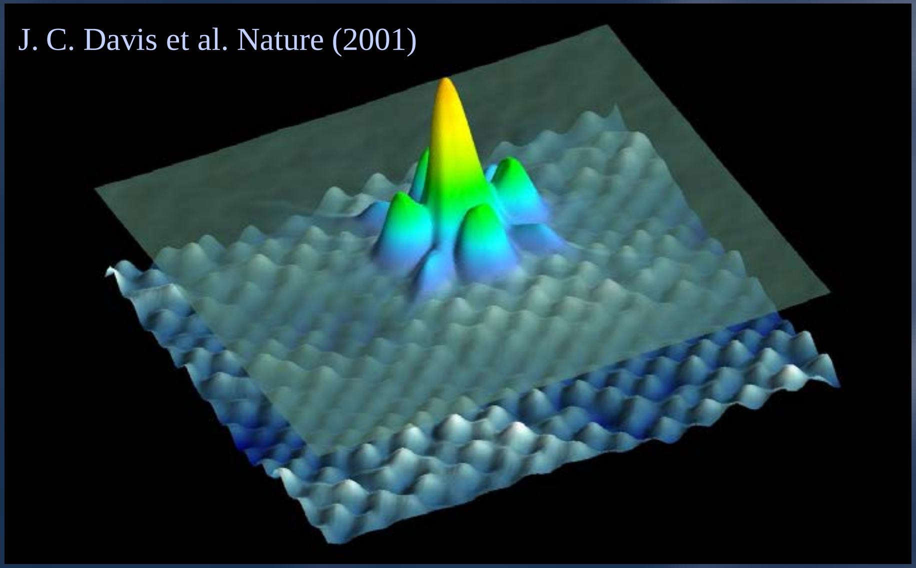


Localized Impurity states fill in the gap!

Impurity States in Gap



A Zn impurity state in the gap of a high temperature superconductor



Scanning Tunneling Microscopy

Localized states are a generic feature of any gap

Spectral weight should be proportional to doping

H. Kruis et al, PRB 64, 054501(2001);
Balatsky et al RMP, 78, 373, (2006)

Conclusions

(1) Universal scaling of χ_{cf}

Microscopic theory?

When causes scaling to stop?

-Onset of heavy fermion fermi liquid state (?)

-Antiferromagnetic order

-Superconductivity, Hidden order

(2) Gapping of spin fluctuations in URu_2Si_2 at 30K

-“Pseudogap”?

-Hidden order precursor effects?

-Low energy collective mode?

-Hybridization gap?

(3) Gradient coupling of HO and AFM constrains microscopic theory