Workshop on ``Heavy Fermion and Quantum Phase Transitions”
11th Nov. @ IOP Beijing, China

Superconductivity Induced by Longitudinal Ferromagnetic Fluctuations in UCoGe

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Collaborators

Kyoto Univ.

UCoGe

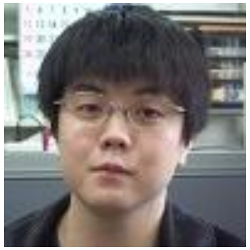
YCoGe

Theory

NQR
NMR

Y. Ihara (PD)

Hokkaido Univ.(AP)



T. Hattori (D2)

K. Karube (D1)

Y. Tada,

ISSP (AP)

Prof. S. Fujimoto

Nagoya Univ.

Tohoku Univ. (IMR)

Sample Preparation
Characterization,
Bulk measurements

Prof. N. K. Sato

AP. K. Deguchi

Prof. I. Satoh

Ferromagnetic Superconductor

Ferromagnetism & **Superconductivity** (with a spin-singlet pairing)

Mutually exclusive ?!

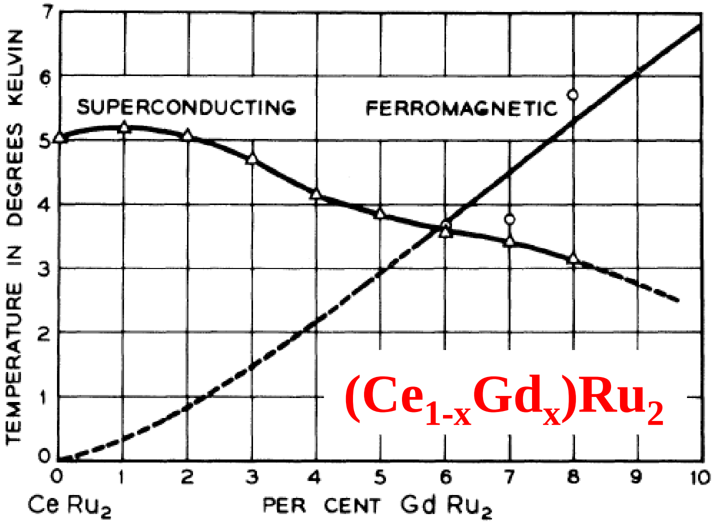
Coexistence of ferromagnetism and superconductivity

Examples

$(Ce_{1-x}Gd_x)Ru_2$ (’58)

x=0.12 **T_{SC} ~ 4 K**

T_{Curie} ~ 3 K



$(Ce_{1-x}Gd_x)Ru_2$

$RuSr_2GdCu_2O_8$ (’96)

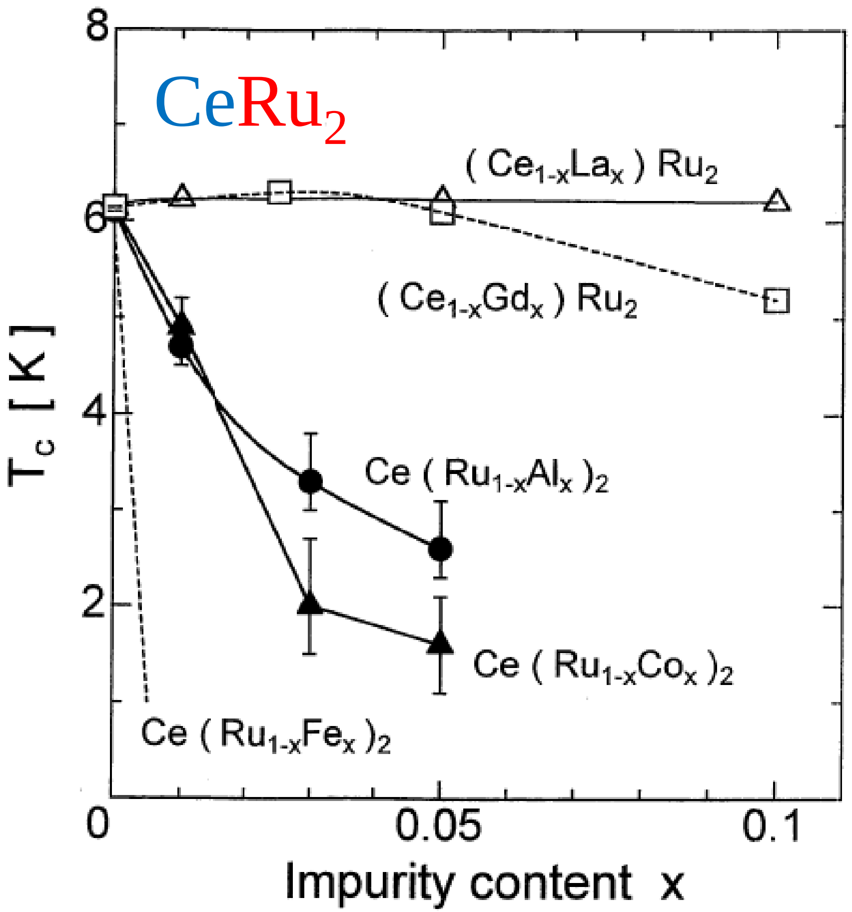
magnetic ordering **T_{Curie}(Ru) ~ 133 K,**

T_N(Gd) ~ 2.2 K

SC transition **T_{SC}(Cu) ~ 16 K,**

B. T. Matthias *et al.*
Phys. Rev Lett. 1, 449 (1958)

Impurity Effect in $CeRu_2$



Mukuda, Ishida, Kitaoka and Asayama,
JPSJ **67**, 2101 ('98)

$CeRu_2$: Cubic Laves structure
s-wave SC

Magnetic Impurity effect
Ru site: very weak
Ce site: vey robust.
⇒ Ru site plays an important role for SC,
FM is related with the Gd ion.

SC and FM are related with different electrons

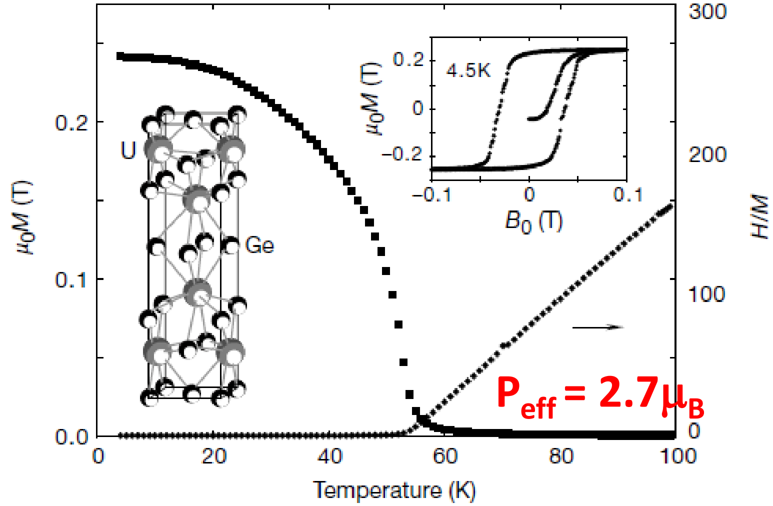
Break Through

S.S.Saxena *et al.*
Nature 406, 587 (00)

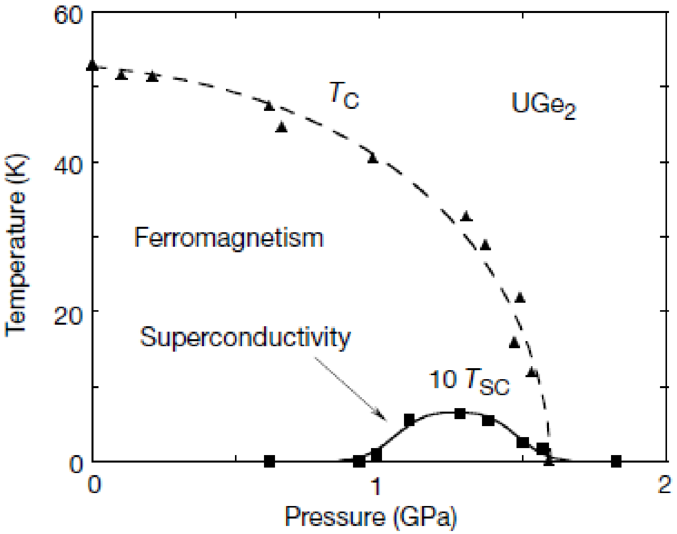
Coexistence of superconductivity and ferromagnetism in URhGe

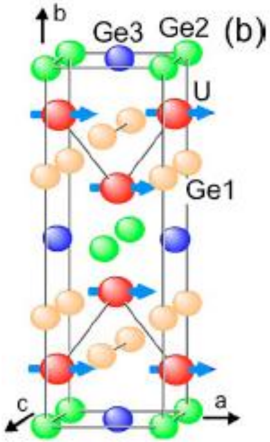
Dai Aoki*, Andrew Huxley*, Eric Ressouche*, Daniel Braithwaite*, Jacques Flouquet*, Jean-Pascal Brison†, Elsa Lhotel† & Carley Paulsen†

UGe$_2$



P$_{eff}$ = 2.7μ_B





Sheikin *et al.*

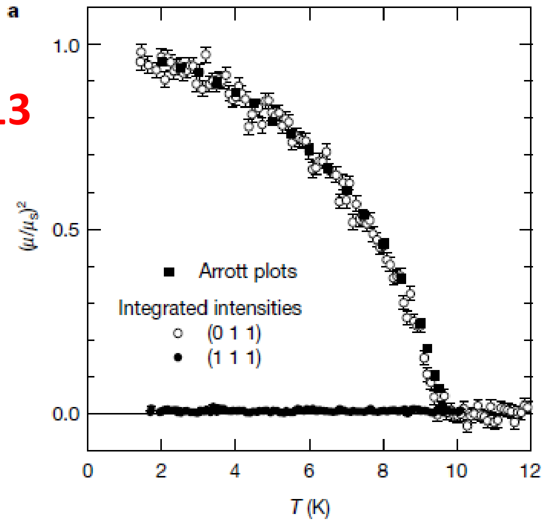
D. Aoki *et al.*
Nature 413, 613 (03)

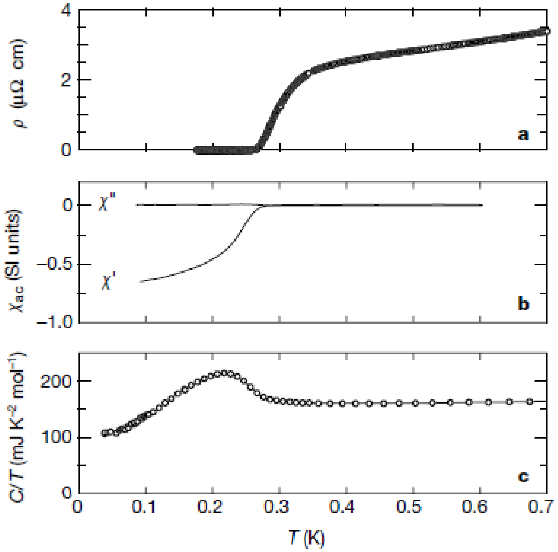
T_{Curie} = 9.5 K

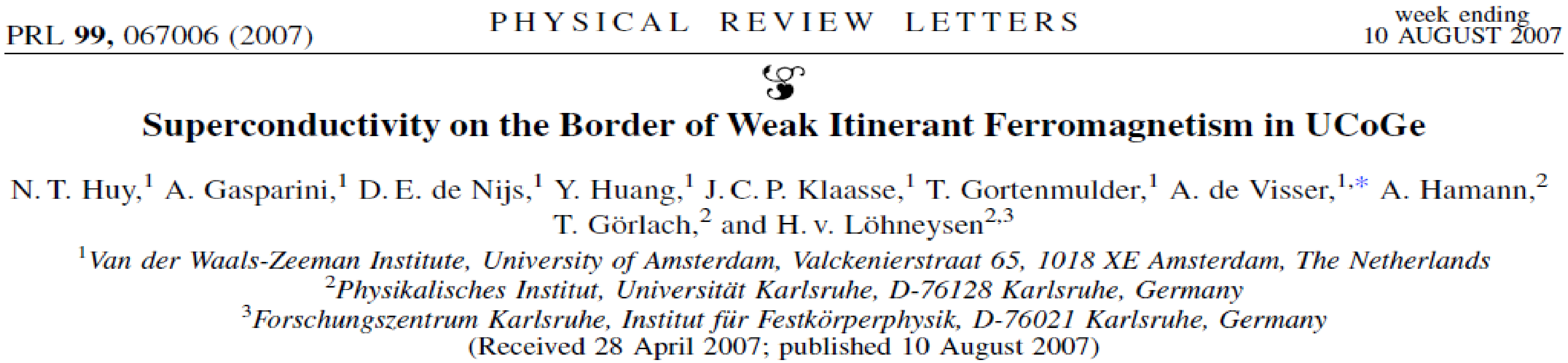
P_{eff} = 1.8 μ_B

P_s = 0.42 μ_B

T_{SC} = 0.3 K







PRL **99,** 067006 (2007) PHYSICAL REVIEW LETTERS week ending 10 AUGUST 2007

Superconductivity on the Border of Weak Itinerant Ferromagnetism in UCoGe

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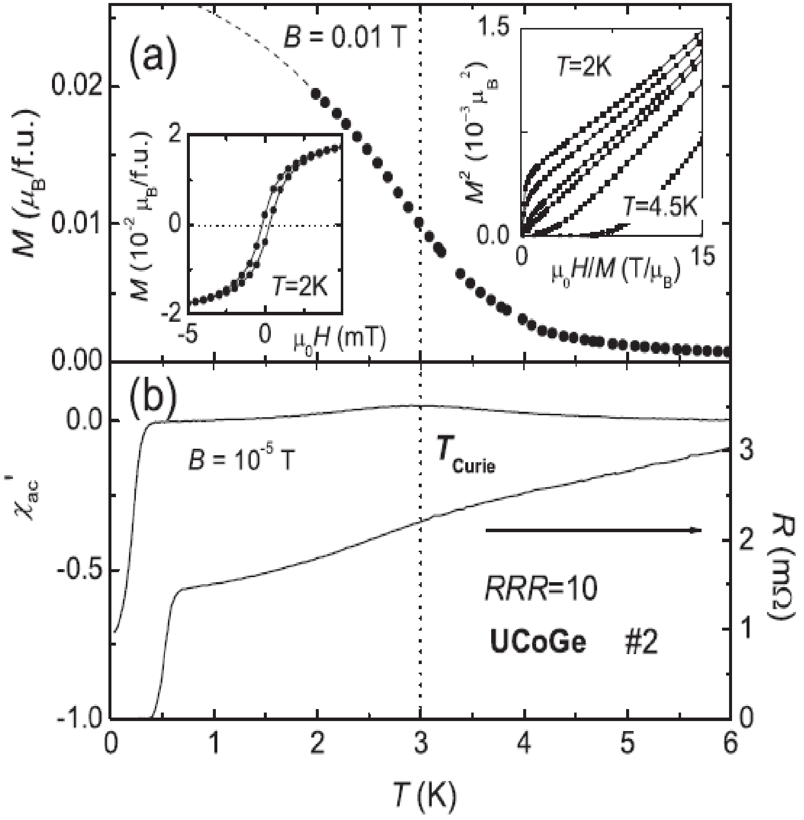
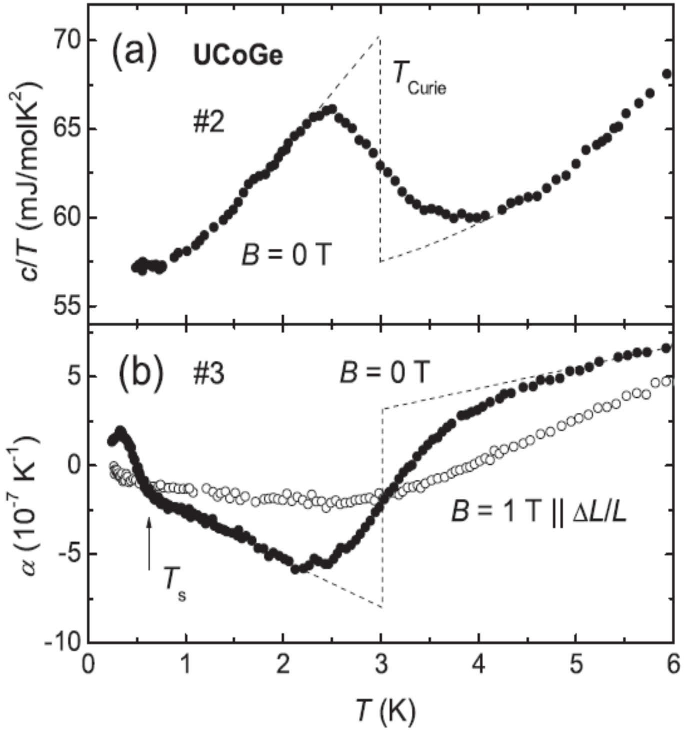


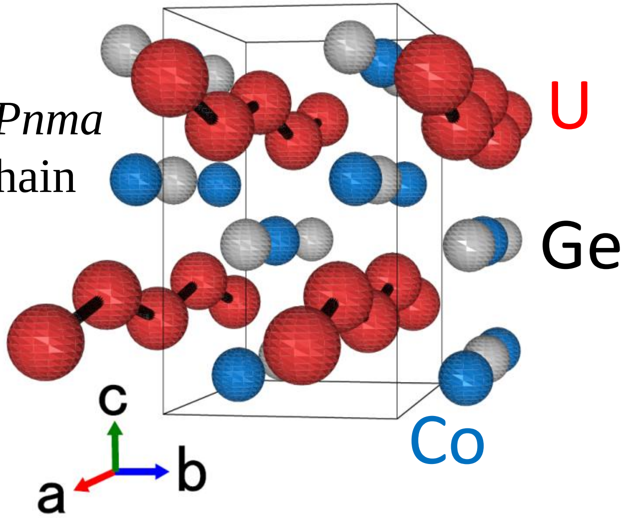


T_{Curie}=3K
m_0=0.03 μ_B : weak itinerant FM
T_{SC}=0.8K

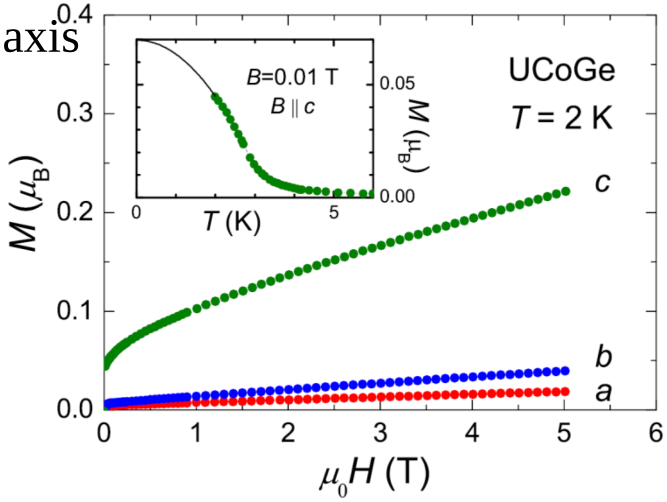
cf. UGe$_2$, P = 1.3 GPa T_{SC} = 0.7K

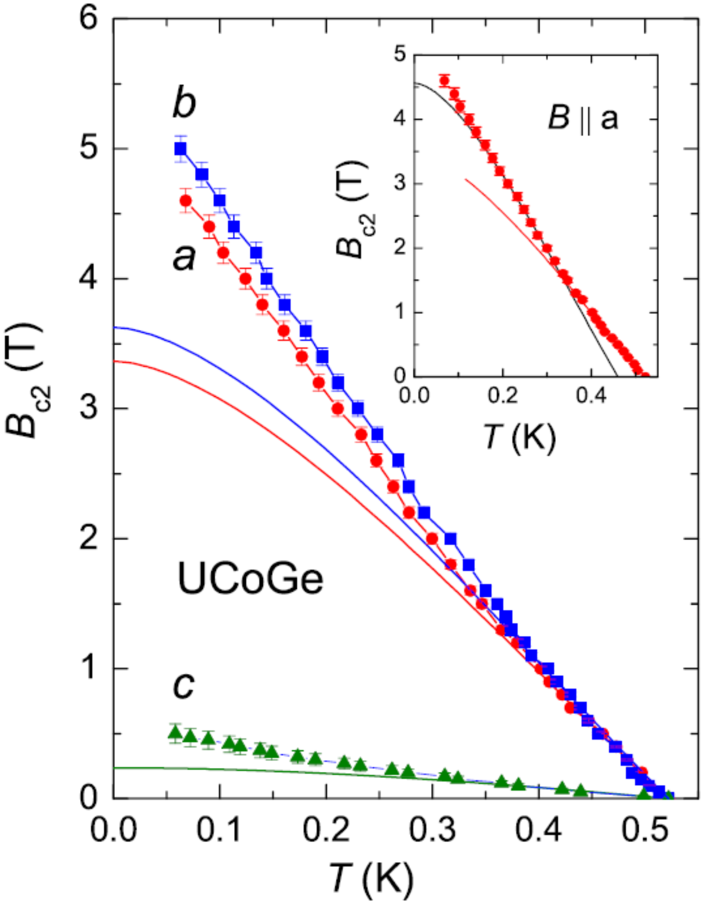
Introduction

- Othorhombic TiNiSi structure *Pnma*
- **U** forms zigzag chain



Easy axis: c axis





N. T. Huy, *et al.*, Phys. Rev. Lett. 100 077002 (2008)

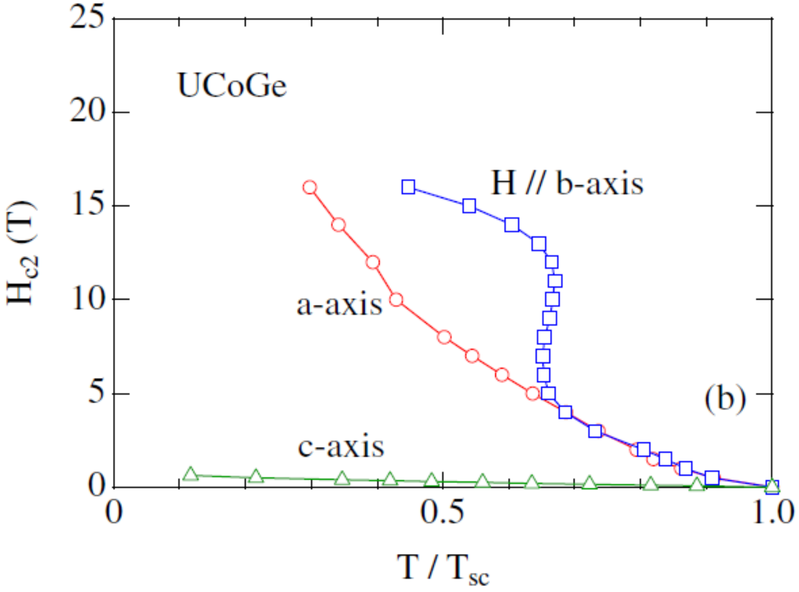
Large Upper Critical Field:

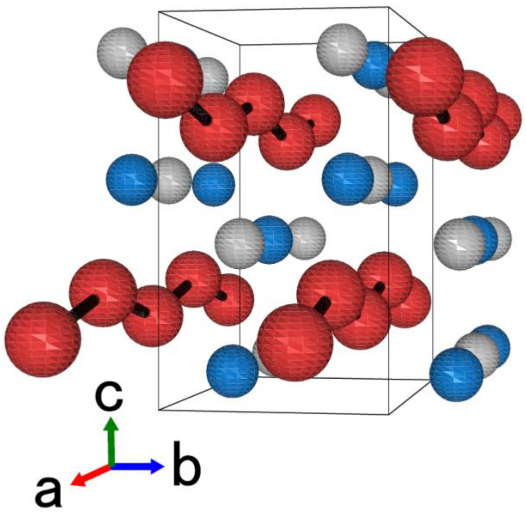
Greater than Pauli-Limit field (that is an expected H_{c2} in the spin-singlet SC)

$$\left(B_{c2}^{\mathrm{Pauli}}(0K) = 1.83T_{\mathrm{SC}} \sim 1\mathrm{T}\right)$$

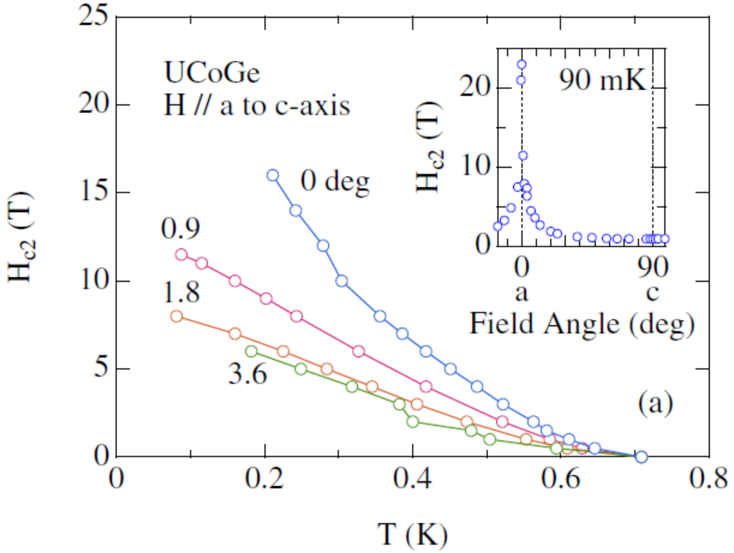
Spin-triplet superconductivity?

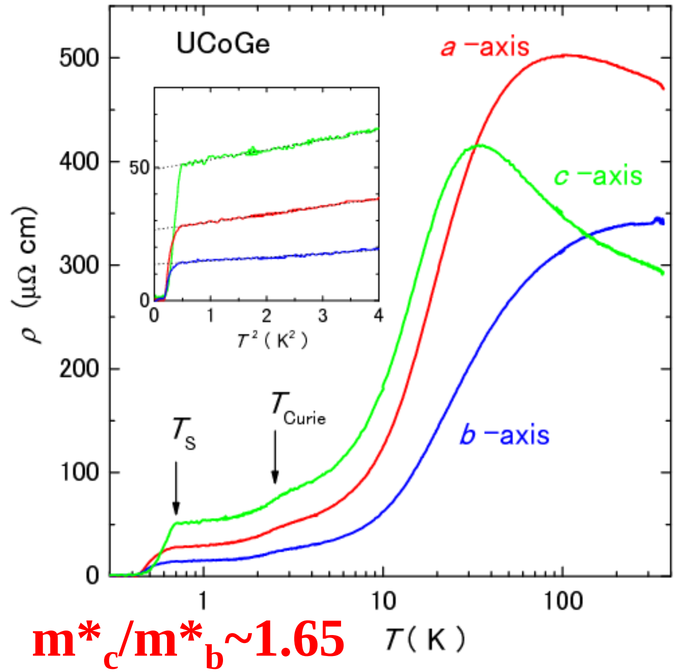
Characteristic Features of H_{c2} in UCoGe





D. Aoki et al.
J. Phys. Soc. Jpn. 78, 113709 (2009)

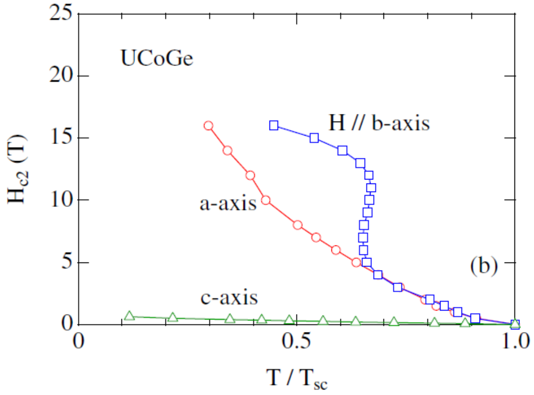




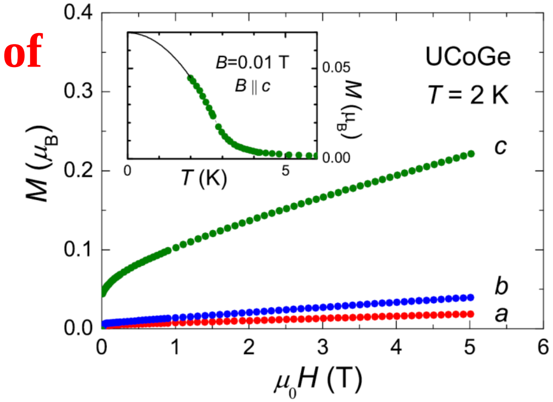
m^*_c/m^*_b~1.65

How do we understand Anisotropy of H_{c2}?

$H_{c2}^{c} << H_{c2}^{a,b}$



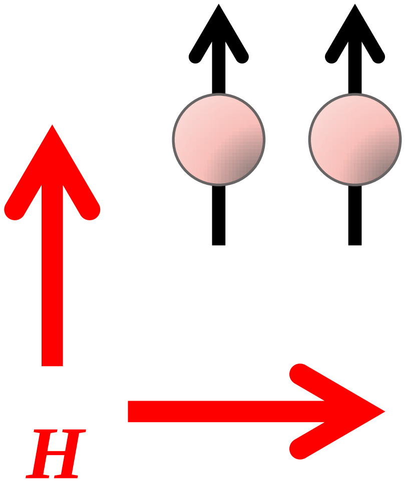
Ising anisotropy of magnetization



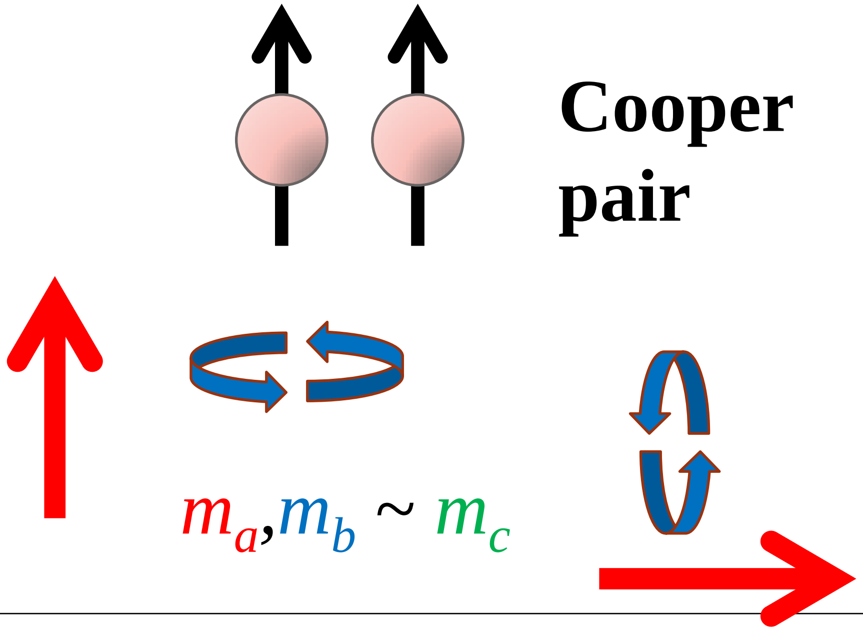
Pauli limit

c-axis

Orbital limit



Cooper pair



Cooper pair

$m_a, m_b \sim m_c$

Aims of Our Studies

Character of coexistence

⇒ coexistence is homogeneous or phase-separation?

Character of magnetic fluctuations

⇒ important for unconventional SC

Why superconductivity is so anisotropy?

Why H_{c2} along a and b are different in high H?

T_c seems to be enhanced when H // b.

To understand above issues,
we have performed Co-NQR and NMR in UCoGe

Single Crystal

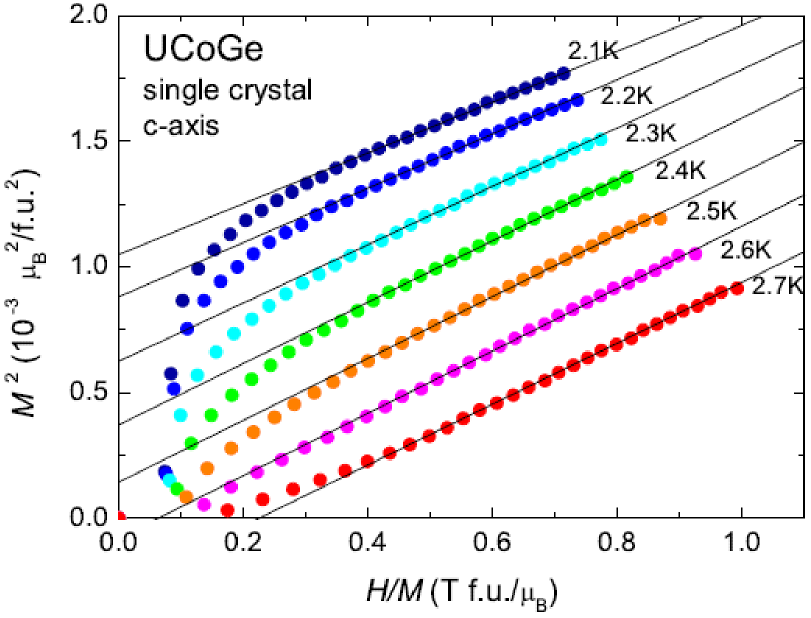
$T_C \sim 2.5K$

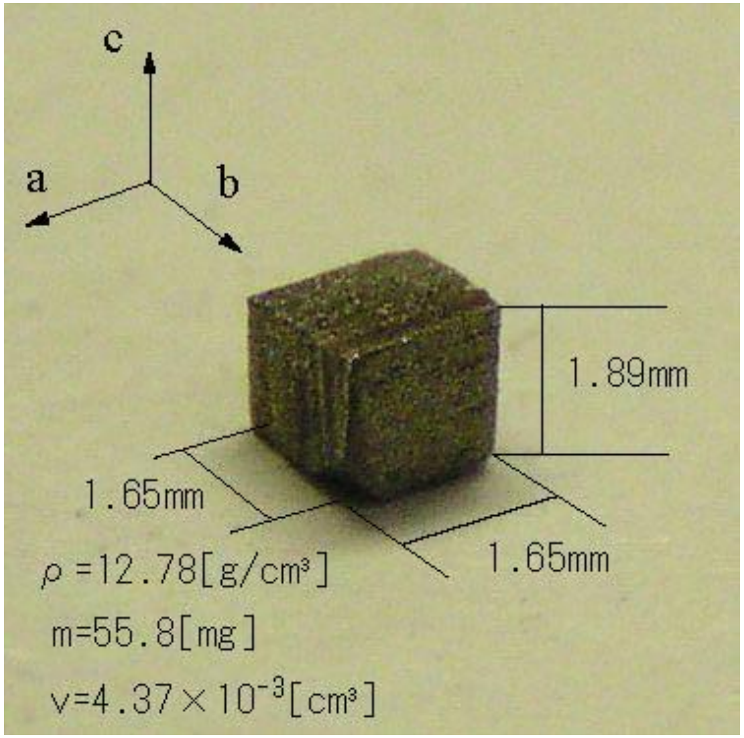
$T_S \sim 0.6K$

Magnetic easy axis: c axis

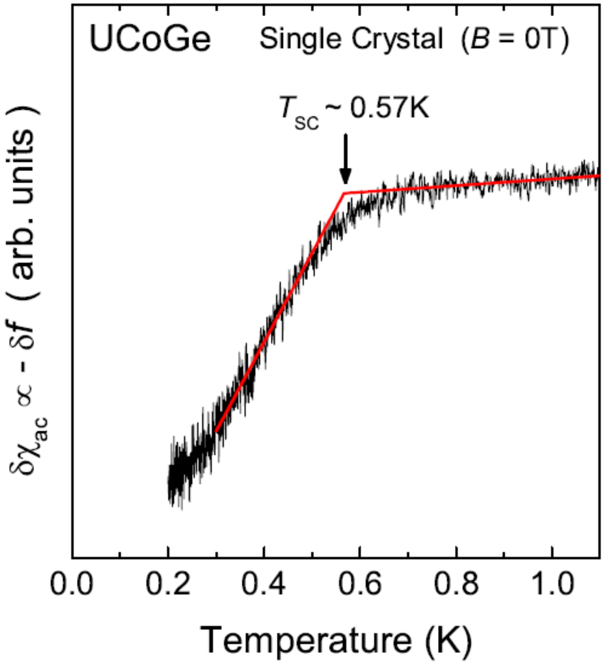
Good agreement with

Huy *et al.* Phys. Rev. Lett. **100** 077002





***RRR* ~ 20**



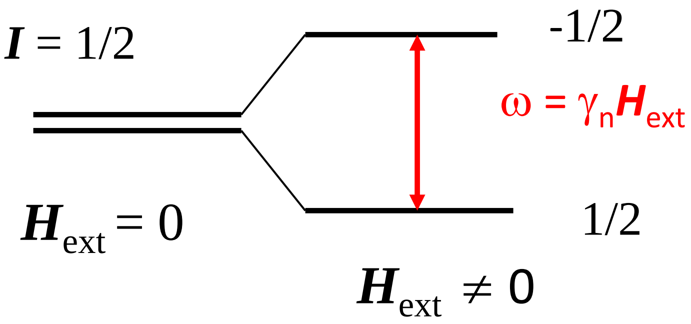
What are NMR and NQR ?

Degenerate nuclear spin levels are lifted by **an magnetic field** .

$$\mathcal{H}_z = -\gamma_{\mathrm{n}}\hbar \boldsymbol{I}\cdot\boldsymbol{H}_{\mathrm{ext}}$$

RF field induces the resonance between two nuclear spin levels

⇒ **Nuclear Magnetic Resonance**



$\boldsymbol{I} = 1/2$

-1/2

$\omega = \gamma_{\mathrm{n}}\boldsymbol{H}_{\mathrm{ext}}$

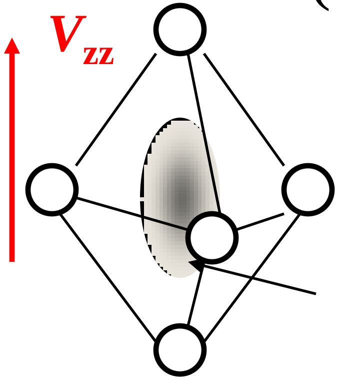
1/2

$\boldsymbol{H}_{\mathrm{ext}} = 0$

$\boldsymbol{H}_{\mathrm{ext}} \neq 0$

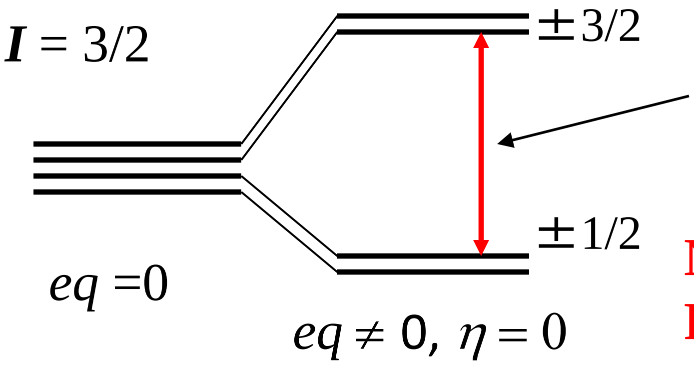
Degenerate nuclear spin levels are lifted by **an electric field gradient.**

$$\mathcal{H}_{eqQ} = \frac{e^2qQ}{4I(2I-1)}\left\{(3I_z{}^2 - \boldsymbol{I}^2) + \frac{1}{2}\eta(I_+{}^2 + I_-{}^2)\right\}, \quad eq \equiv V_{zz}, \eta \equiv \frac{V_{xx} - V_{yy}}{V_{zz}}$$



V_{zz}

Nuclear quadrupole moments



$\boldsymbol{I} = 3/2$

±3/2

±1/2

$eq = 0$

$eq \neq 0,\ \eta = 0$

$$\boxed{\hbar\omega_{\mathrm{Q}} = \frac{e^2qQ}{2}}$$

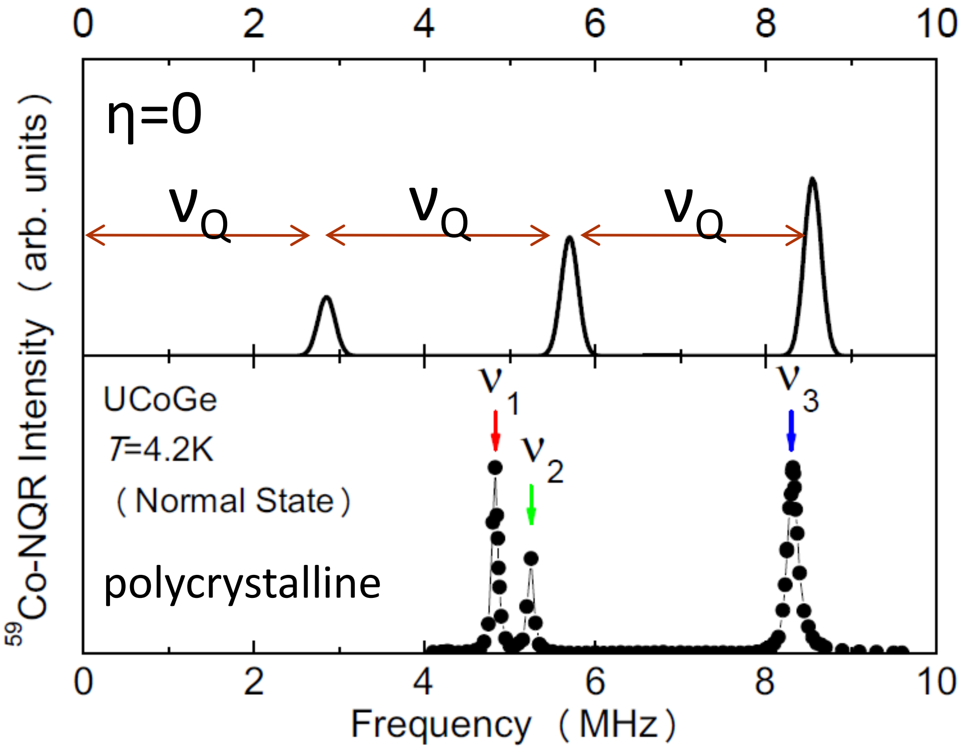
Nuclear quadrupole Resonance

^{59}Co-NQR spectrum 1

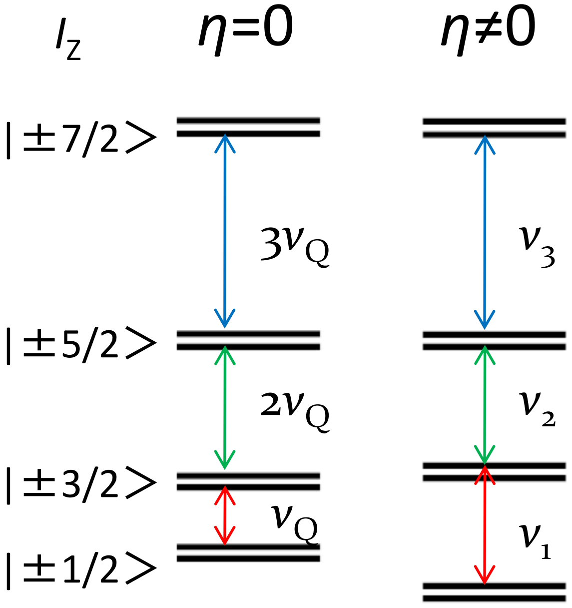
Electric quadrupole Hamiltonian

$$\mathcal{H}_{eqQ} = \frac{\nu_Q}{6}\left\{\left(3\hat{I}_Z^2 - \hat{I}^2\right) + \frac{1}{2}\eta\left(\hat{I}_+^2 + \hat{I}_-^2\right)\right\}$$

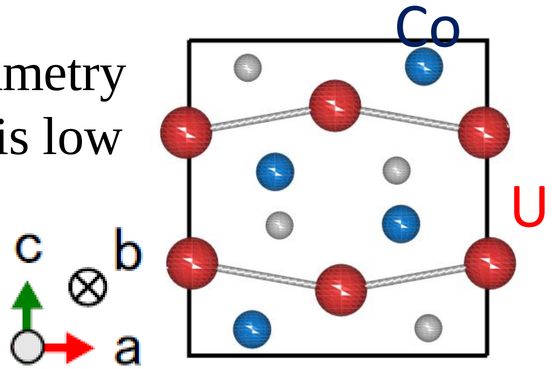
Asymmetric parameter $\eta = \frac{V_{XX} - V_{YY}}{V_{ZZ}}$



^{59}Co (nuclear spin I = 7/2)

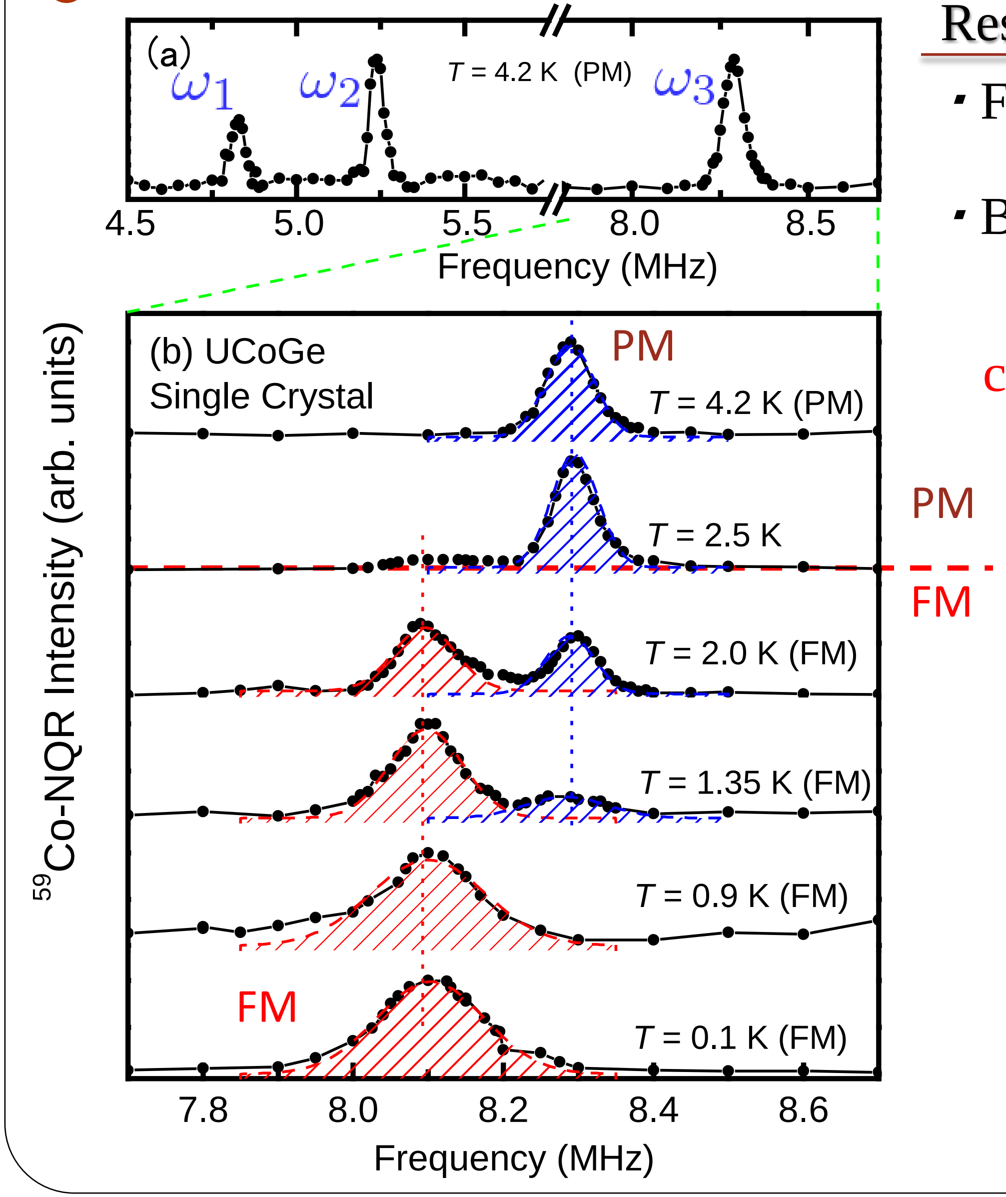


Local symmetry at Co site is low
⇒η ≠0



ν_Q=2.85MHz, η=0.52

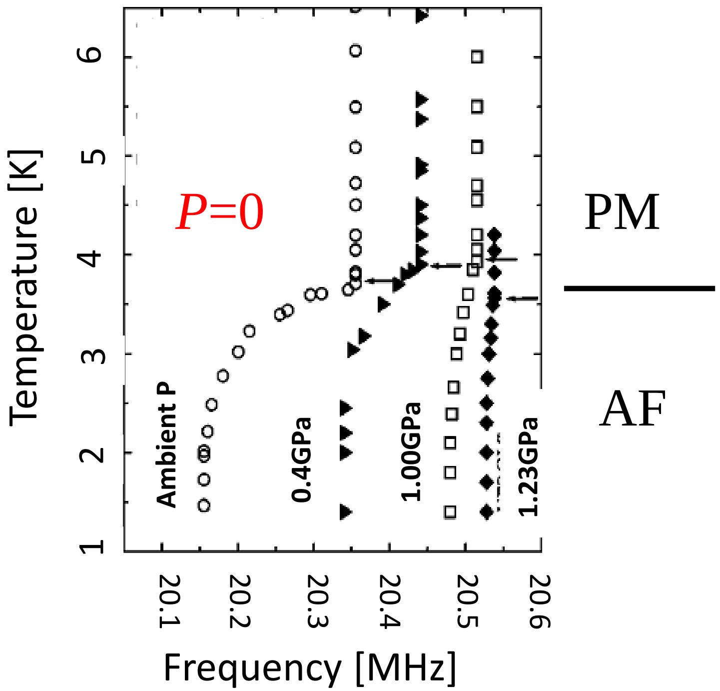
First-order like transition at T_{Curie} in the single crystal



Results

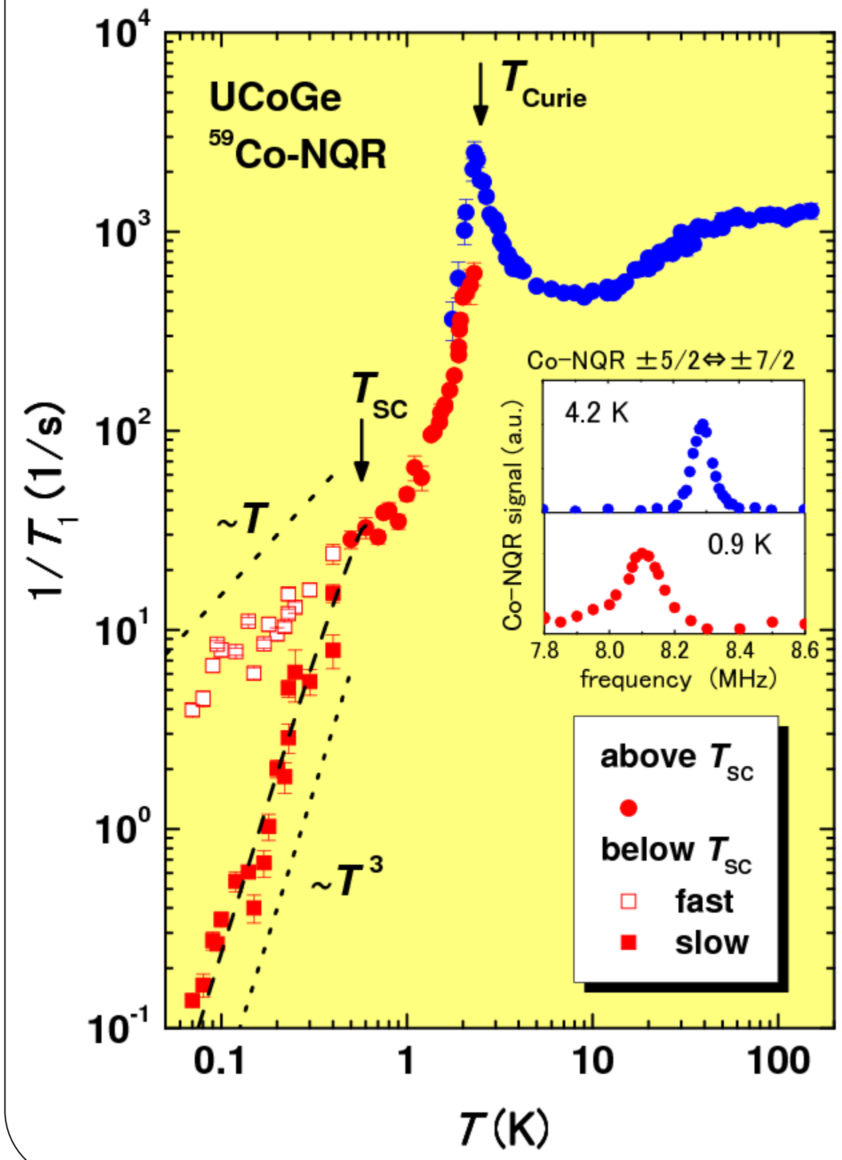
- FM signal and PM signal are observed between 2.5 K and 1 K
- Below 1 K, whole region of the sample is in the FM state

cf. CeRhIn$_5$ AF-Order 2$^{\mathrm{nd}}$-order



T.Mito *et al*,, Phys. Rev. B. 63(2001) 220507

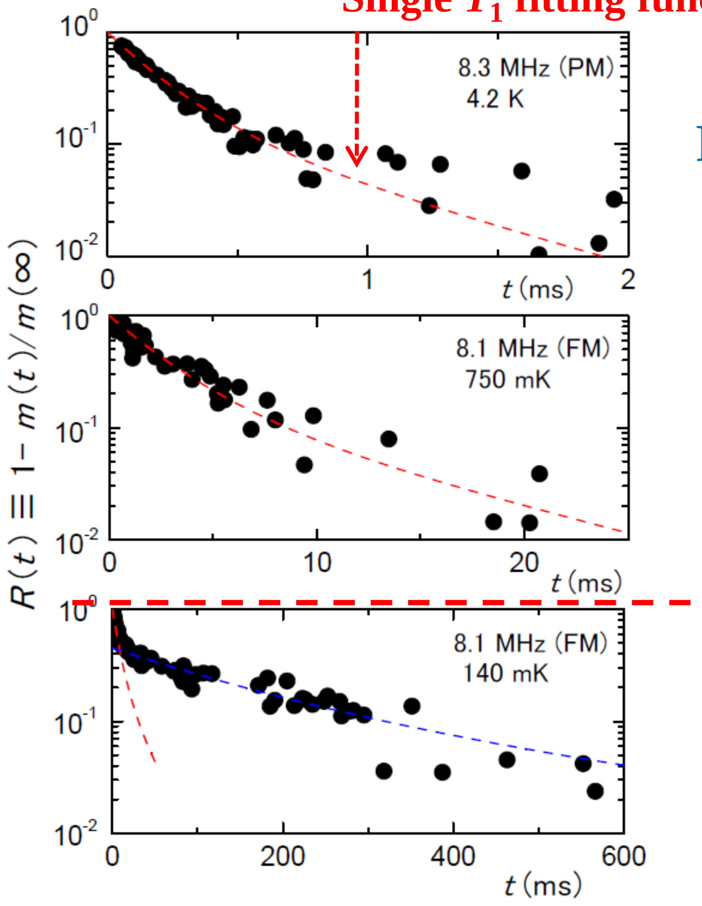
Summary of ^{59}Co-NQR on UCoGe (Zero field)



T. Ohta *et al.* JPSJ 79, 023707 ('10)

Single component of $1/T_1$ is determined above T_{SC}.⇒ **Ele. State is homogeneous**

Single T_1 fitting function



PM

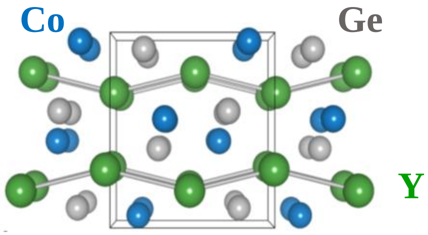
FM

SC

Two components of $1/T_1$ are observed below T_S

Reference compound YCoGe

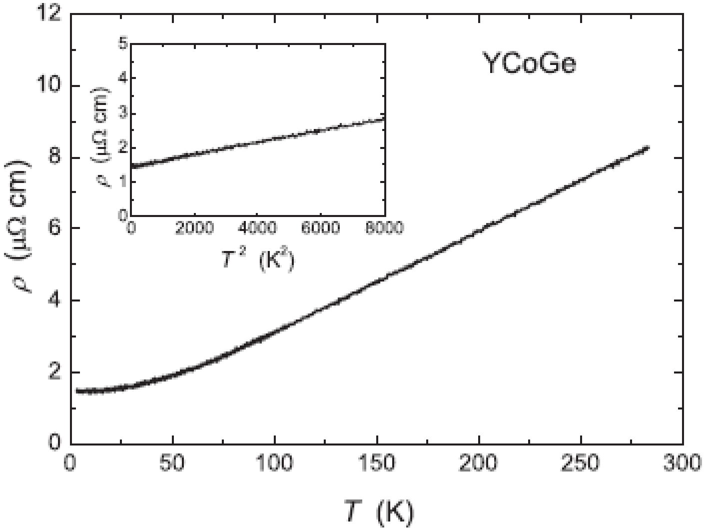
c.f. **reference: YCoGe** (P_{nma} with similar L.C.)

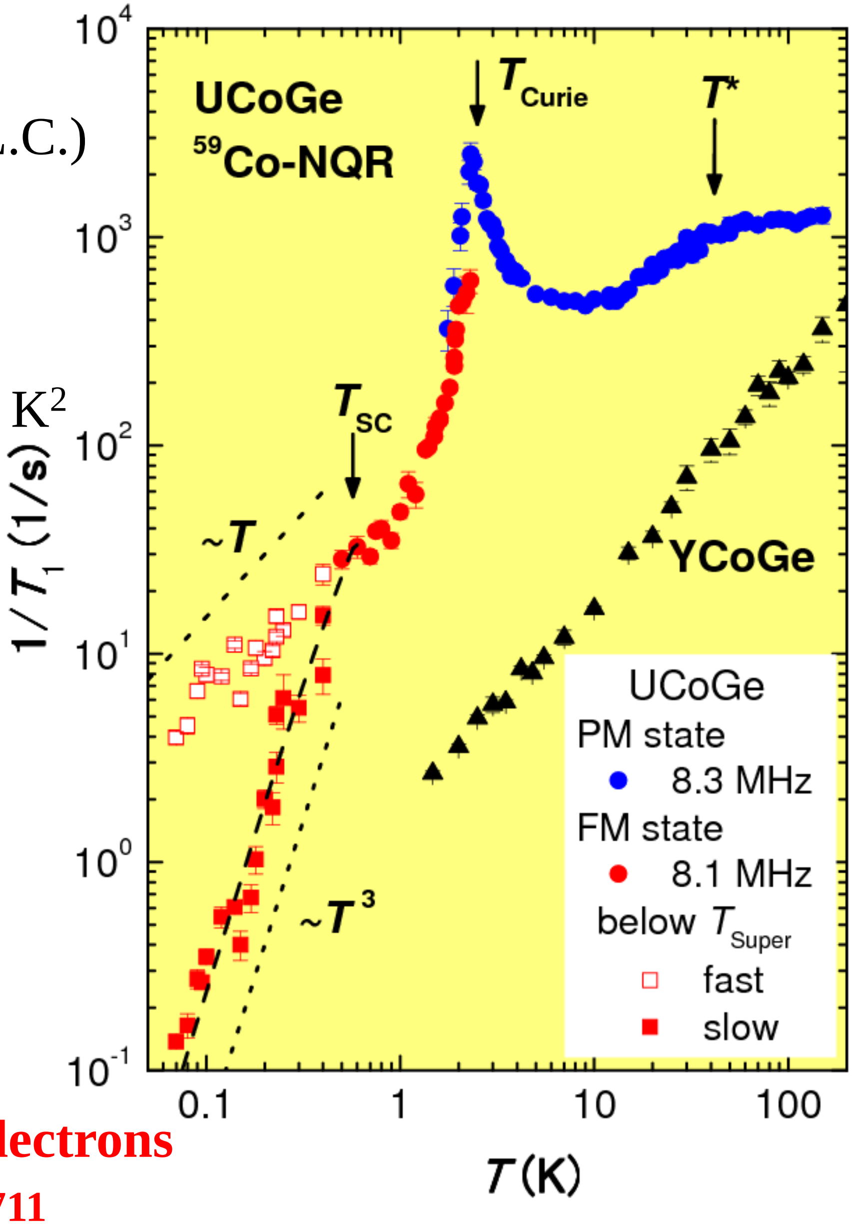


Y : [Kr] $4d^{1}5s^{2}$

U : [Rn] $5f^{n}$, $6d^{1}5s^{2}$

$^{\mathrm{YCoGe}}\gamma_{n}$ = 6.6 mJ /mol K^2





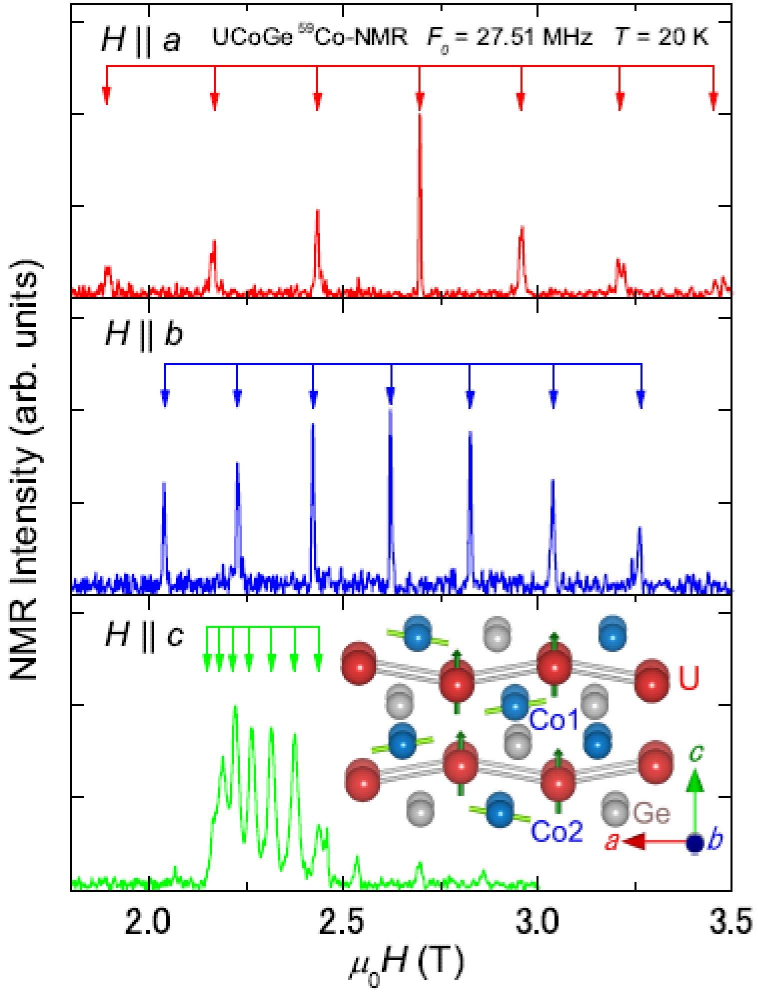
No SC & FM down to 100 mK

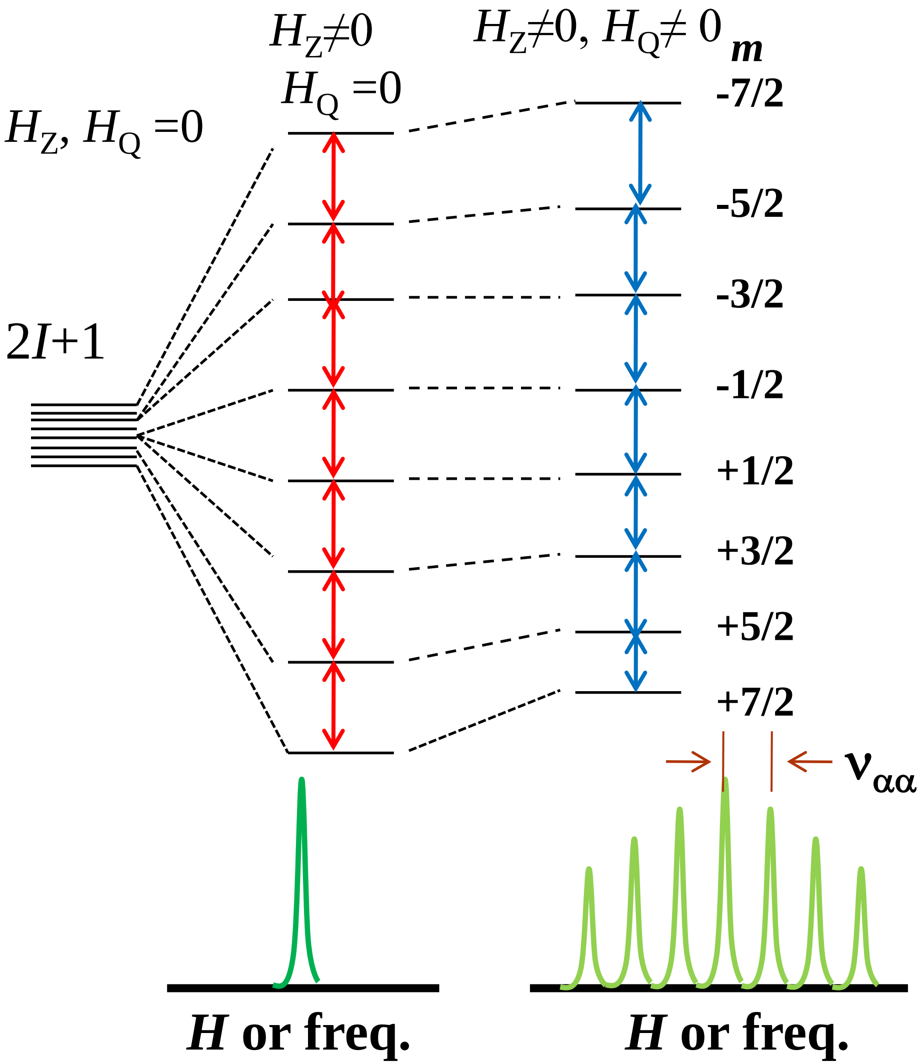
⇒ SC and FM originate from U-5f electrons

Karube J. Phys. Soc. Jpn. 80, 064711

Co NMR (Single Crystal) $H_z >> H_Q$

Zeeman Hamiltonian : $H_Z = -\gamma_n \hbar \boldsymbol{I} \boldsymbol{H}$





H_Z, H_Q =0

2I+1

$H_Z \neq 0$
H_Q =0

$H_Z \neq 0$, $H_Q \neq$ 0

m

-7/2

-5/2

-3/2

-1/2

+1/2

+3/2

+5/2

+7/2

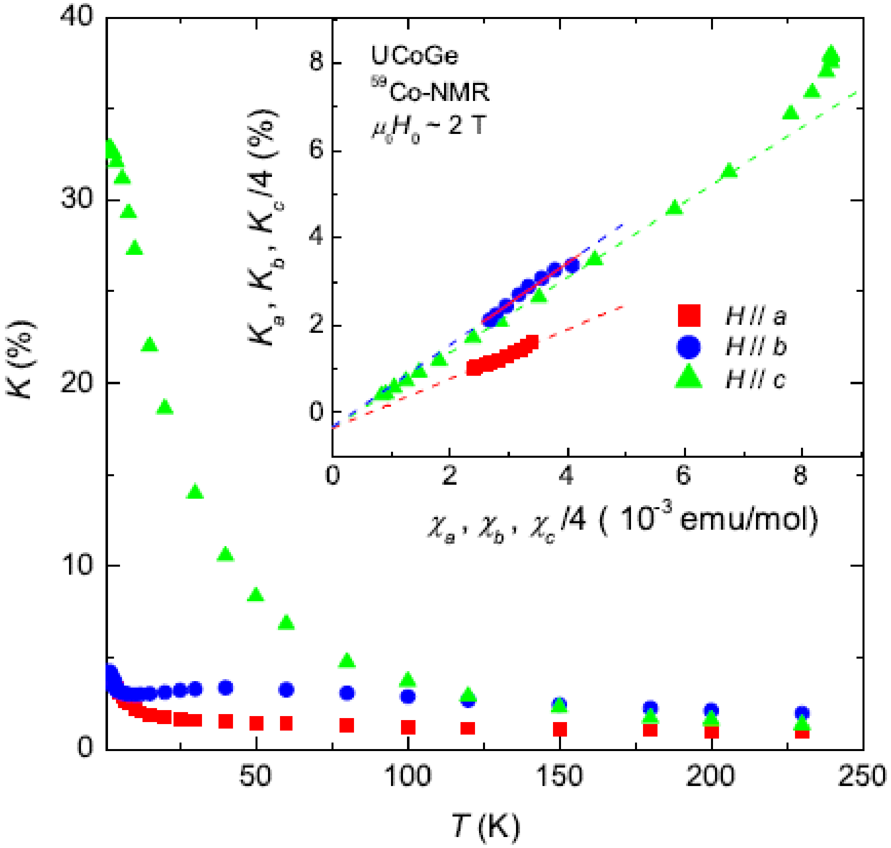
$\nu_{\alpha\alpha}$

H **or freq.**

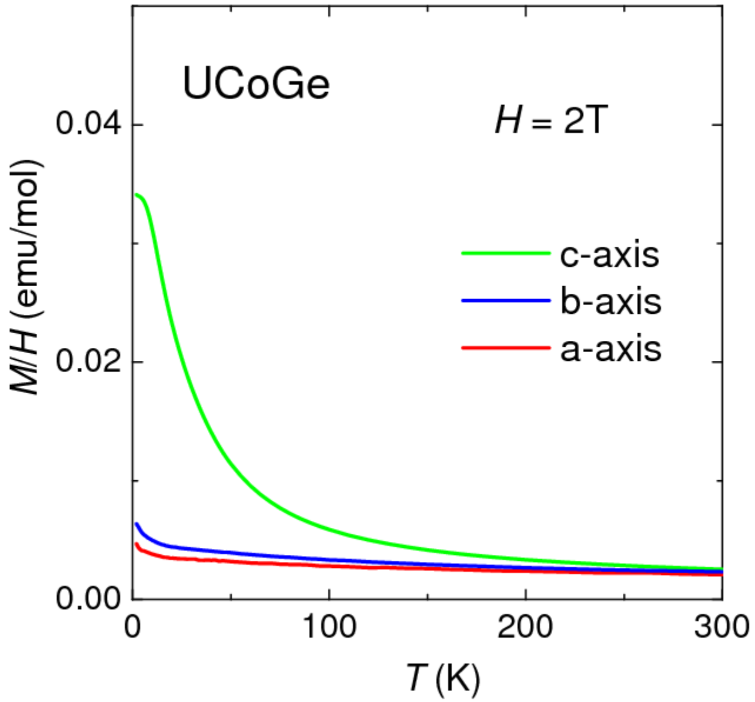
H **or freq.**

Direction dependence of Knight shift

Knight shift (Static, *q* = 0)



χ_{bulk}

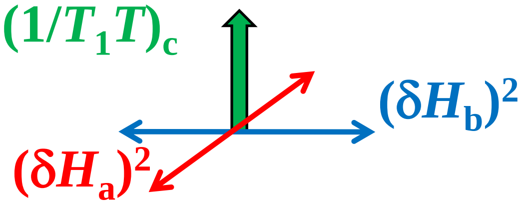


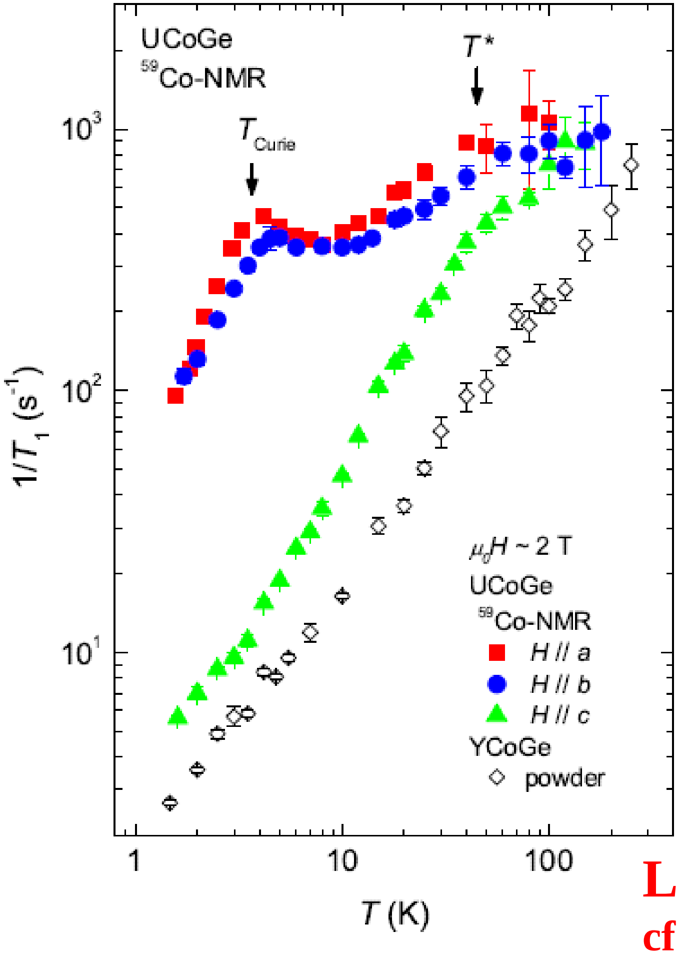
$$K_\alpha = A^\alpha_{\mathrm{hf}} \chi^\alpha(0) + K^\alpha_{\mathrm{orb}}$$

- **Knight shift is anisotropic, which is scale to bulk susceptibility.**

⇒ Magnetic properties at the Co site are affected by the U-5*f* state.

Direction-dependence of $1/T_1$ (Dynamic properties)



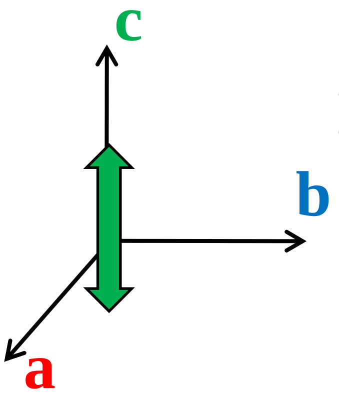


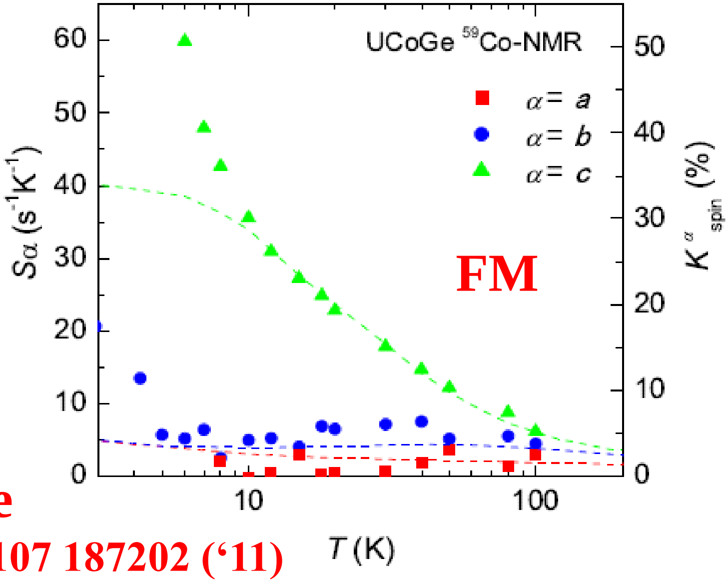
$$\left(\frac{1}{T_1T}\right)_a = \frac{\gamma_n^2 k_B}{(\gamma_e\hbar)^2}\sum_{\boldsymbol{q}}\left[\underline{\left|A_{hf}^{\,b}\right|^2\frac{\chi''_b(q,\omega_N)}{\omega_N}}+\underline{\left|A_{hf}^{\,c}\right|^2\frac{\chi''_c(q,\omega_N)}{\omega_N}}\right]$$

$$\left(\frac{1}{T_1T}\right)_b = \frac{\gamma_n^2 k_B}{(\gamma_e\hbar)^2}\sum_{\boldsymbol{q}}\left[\underline{\left|A_{hf}^{\,c}\right|^2\frac{\chi''_c(q,\omega_N)}{\omega_N}}+\underline{\left|A_{hf}^{\,a}\right|^2\frac{\chi''_a(q,\omega_N)}{\omega_N}}\right]$$

$$\left(\frac{1}{T_1T}\right)_c = \frac{\gamma_n^2 k_B}{(\gamma_e\hbar)^2}\sum_{\boldsymbol{q}}\left[\underline{\left|A_{hf}^{\,a}\right|^2\frac{\chi''_a(q,\omega_N)}{\omega_N}}+\underline{\left|A_{hf}^{\,b}\right|^2\frac{\chi''_b(q,\omega_N)}{\omega_N}}\right]$$

$$S_\alpha = \frac{\gamma_n^2 k_B}{(\gamma_e\hbar)^2}\sum_{\boldsymbol{q}}\left[\left|A_{hf}^{\,\alpha}\right|^2\frac{\chi''_\alpha(q,\omega_N)}{\omega_N}\right] \qquad (\alpha = a,\ b,\ \text{and}\ c)$$

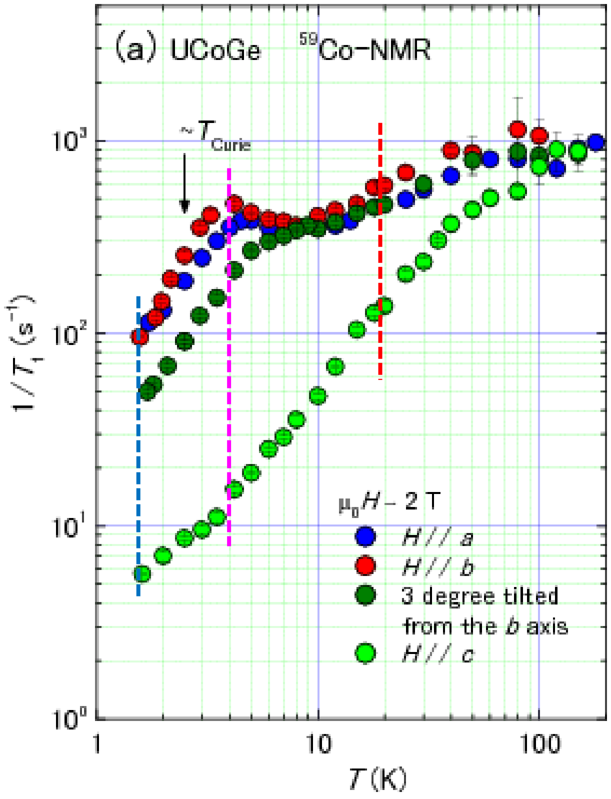


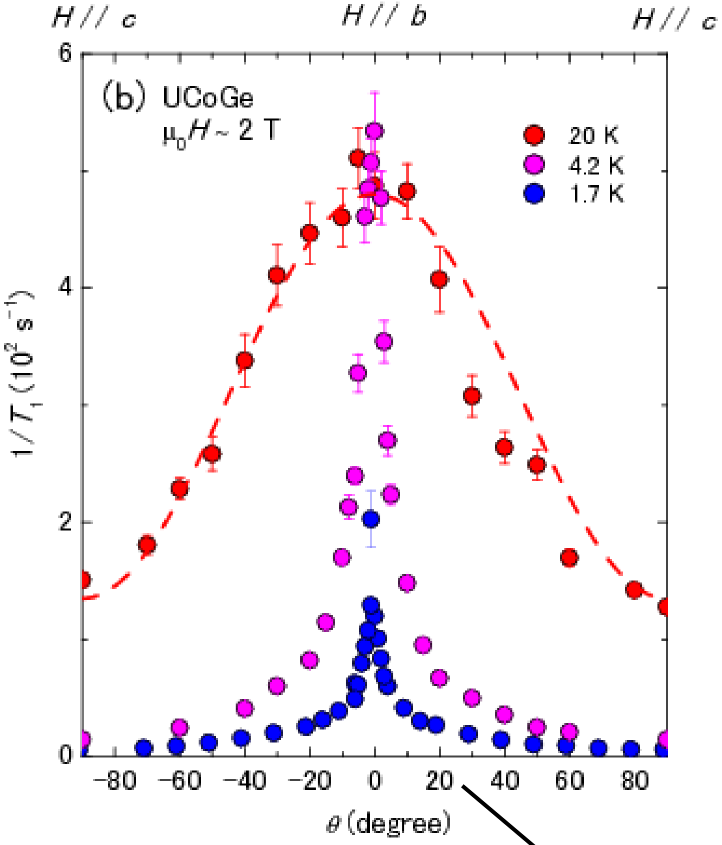


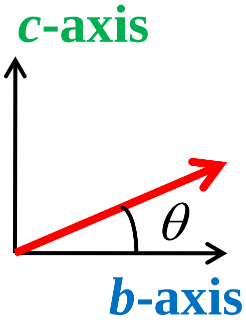
Longitudinal mode

cf. C. Stock *et al.* PRL 107 187202 ('11)

Angle dependence of $1/T_1$ at various temperatures

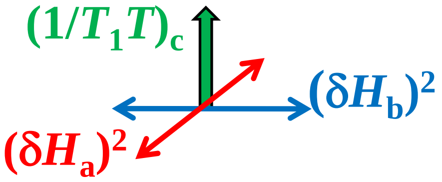


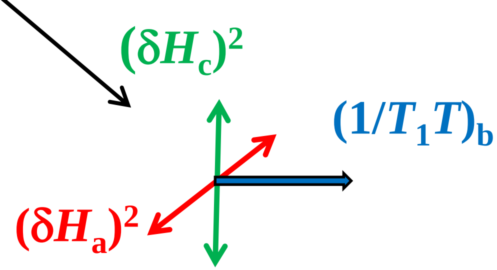




ab-plane dynamics are isotropic

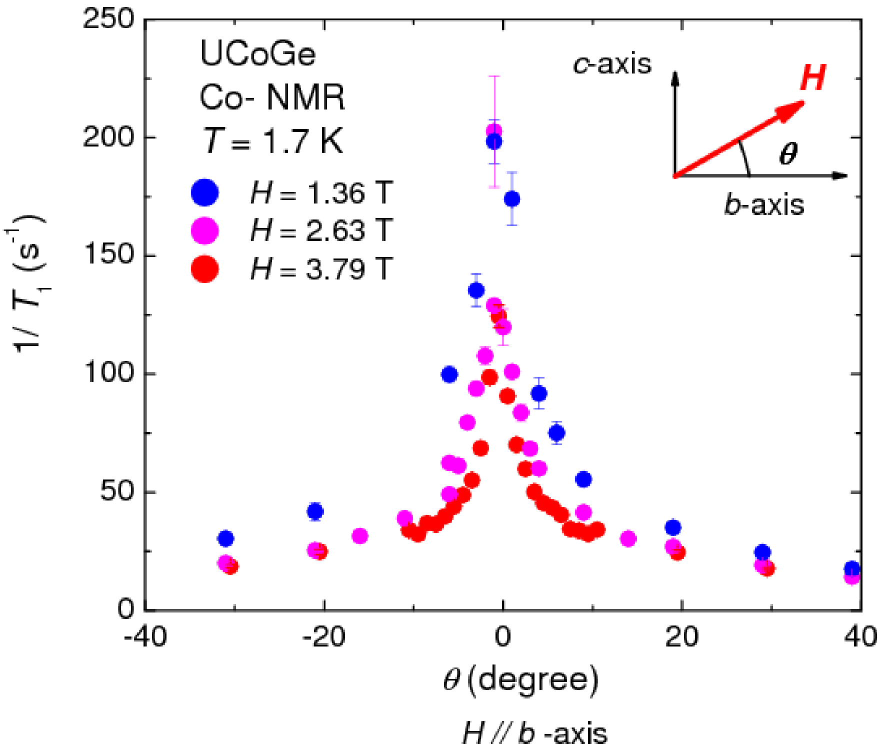
$$1/T_1(\theta) = (1/T_1)_b \cos^2\theta + (1/T_1)_c \sin^2\theta$$

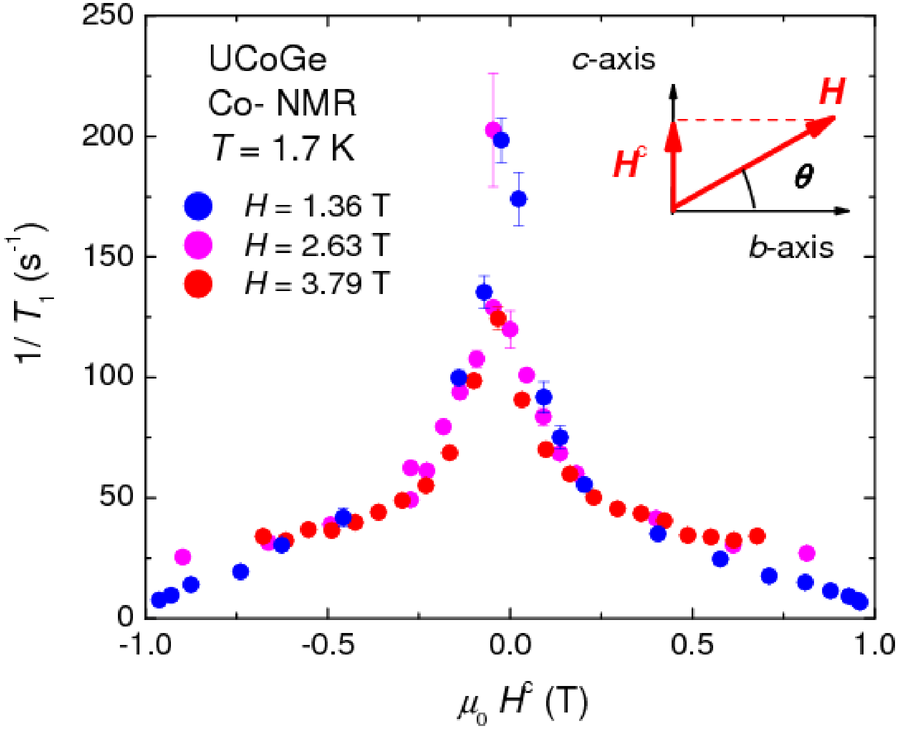




20 K data can be fitted by this relation.

Angle dependence of $1/T_1$ at 1.7 K under different H

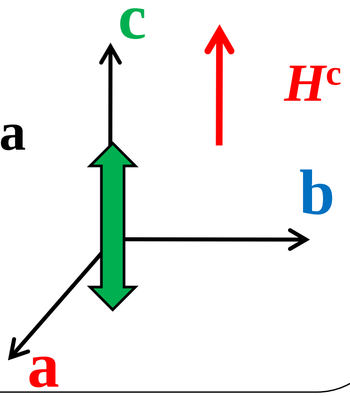




- Angle dependence of $1/T_1$ becomes steeper with higher fields.

- $(1/T_1)_c$ is independent of H.

If $1/T_1$ is plotted against H^c, the data fall on the same curve.

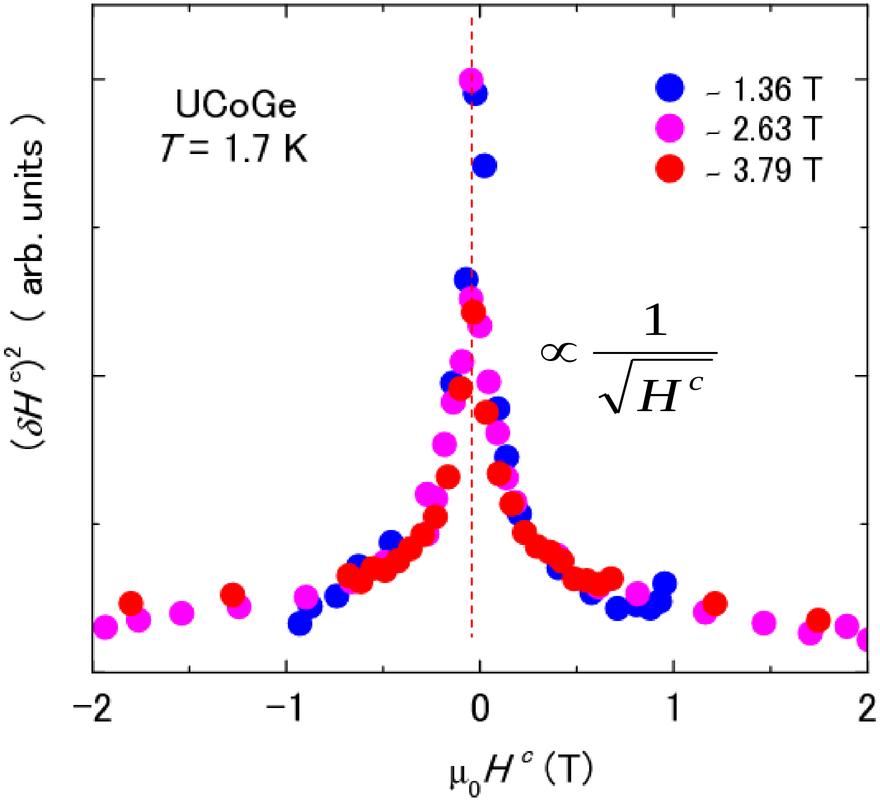


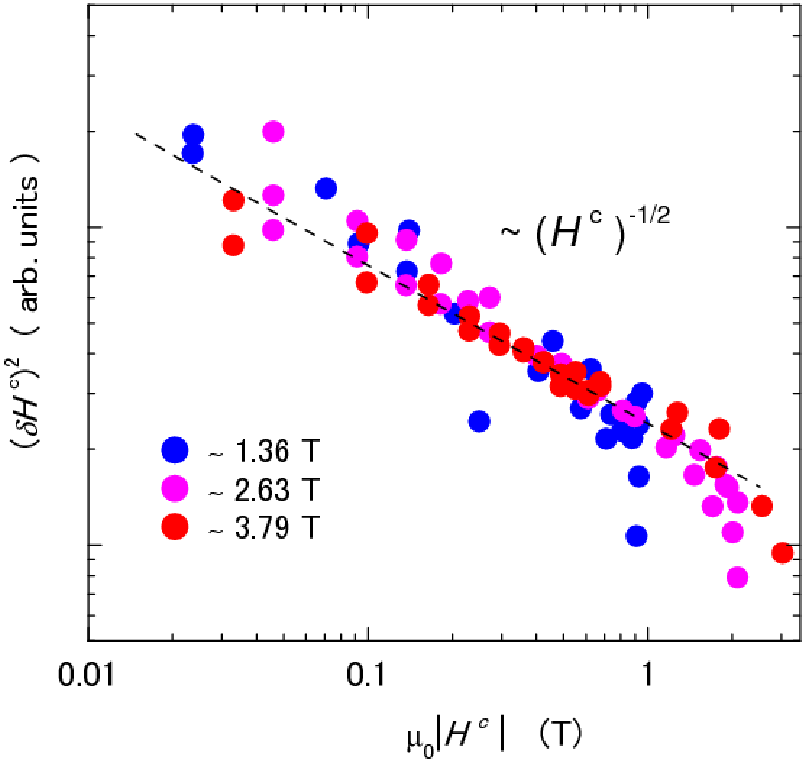
H^c dependence of the c-axis FM Fluctuations

$$\frac{1}{T_1}(\theta)=\frac{1}{T_1^{b}}\cos^2\theta+\frac{1}{T_1^{c}}\sin^2\theta=\left[(\delta H_a)^2+(\delta H_c)^2\right]\cos^2\theta+\frac{1}{T_1^{c}}\sin^2\theta$$

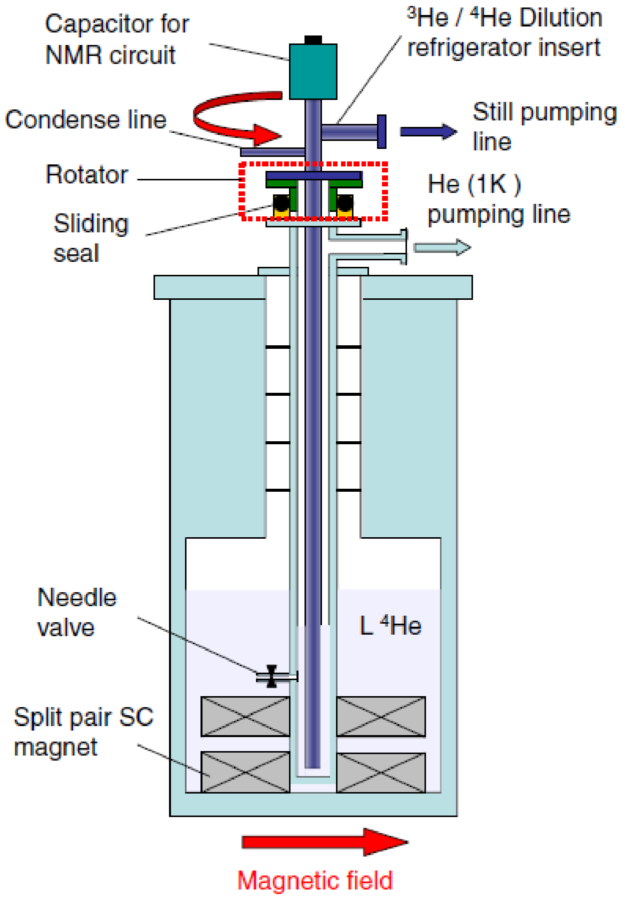
$$(\delta H^c)^2(\theta)\propto\frac{1}{\cos^2\theta}\left(\frac{1}{T_1}(\theta)-\frac{(1+\sin^2\theta)}{2}\frac{1}{T_1^{c}}\right)$$

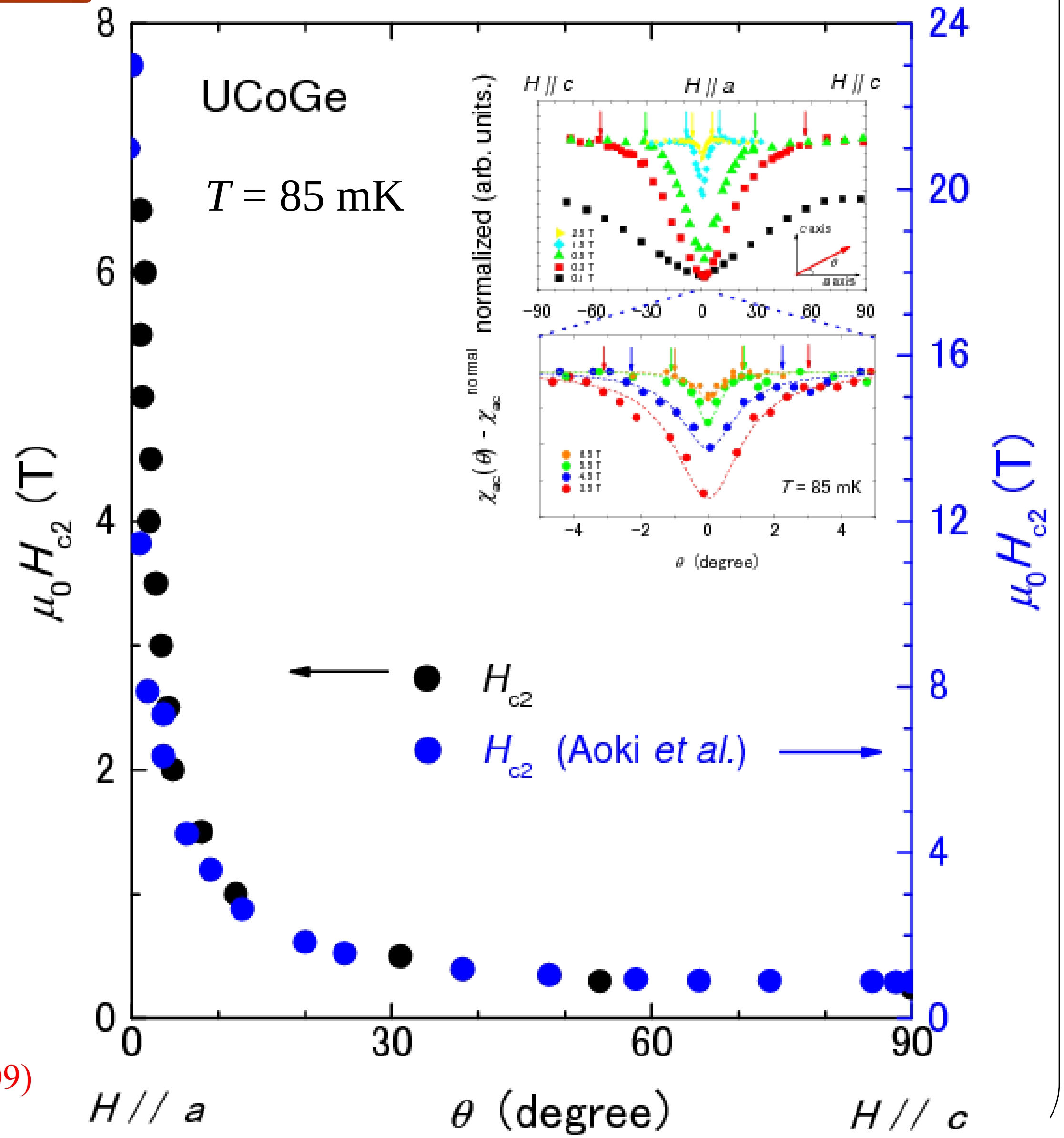
$$\boxed{(\delta H^c)^2\propto\frac{1}{\sqrt{H^c}}}$$





Critical Magnetic Field of Superconductivity



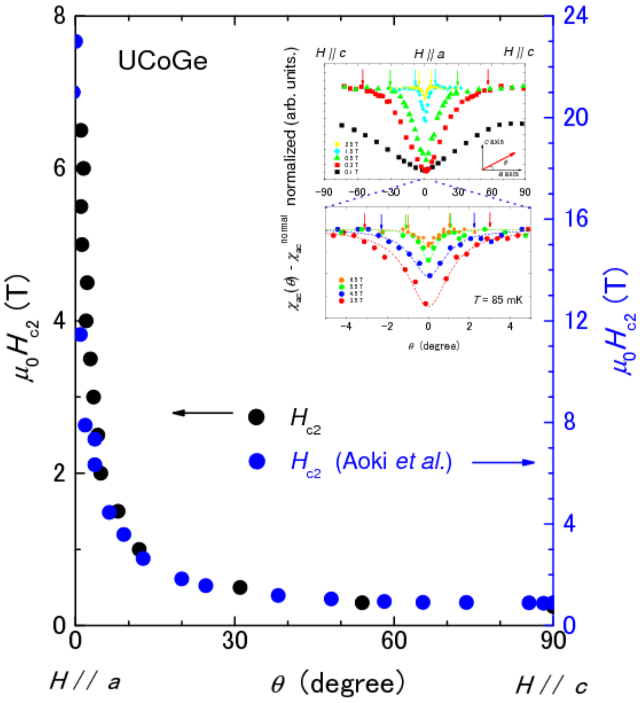


Anisotropy of H_{c2} is in good agreement with the Aoki's results

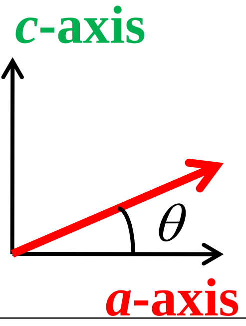
D. Aoki *et al*., J. Phys. Soc. Jpn. **76** 113709 ('09)

Relationship between Magnetic and SC Properties

Transport properties

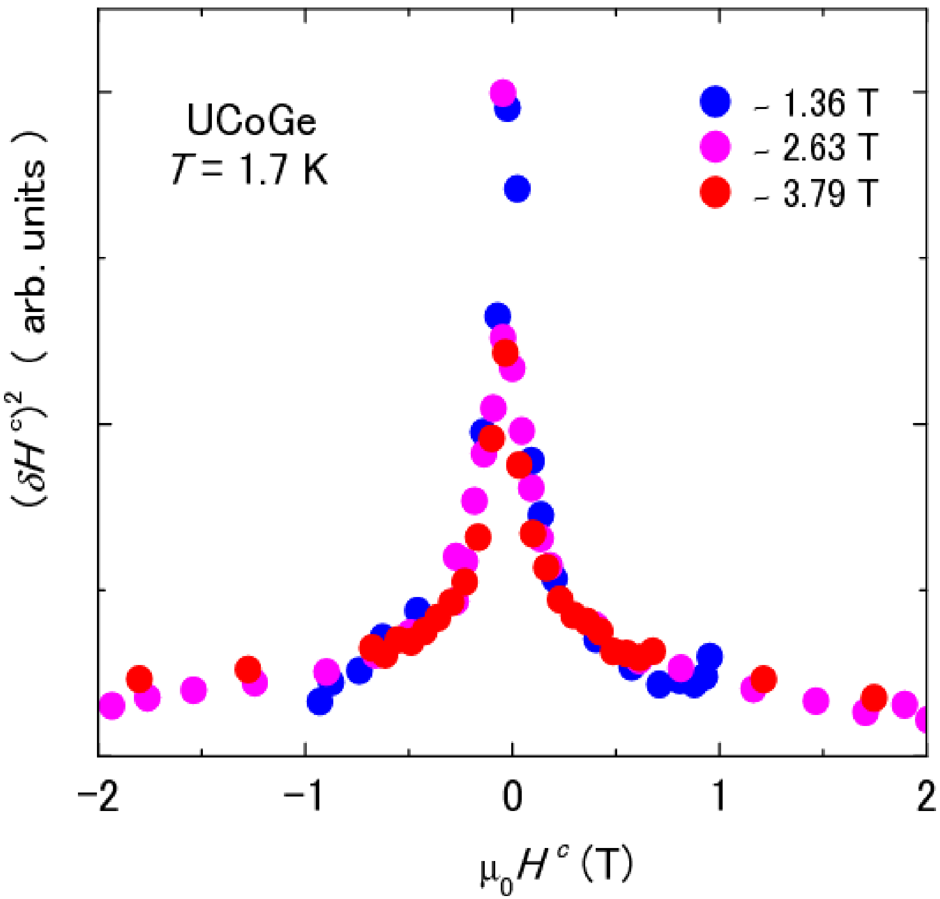


H_{c2} is plotted agaist c-axis component of *H*.


***c*-axis**

θ

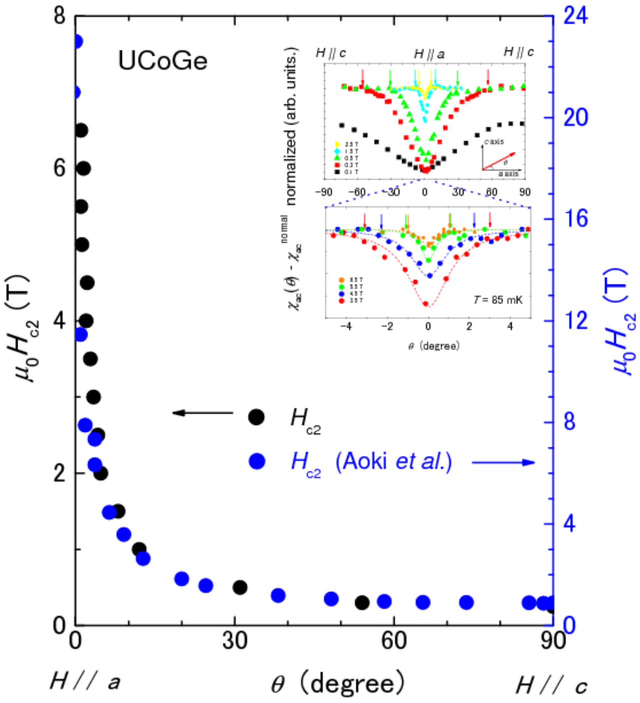
***a*-axis**



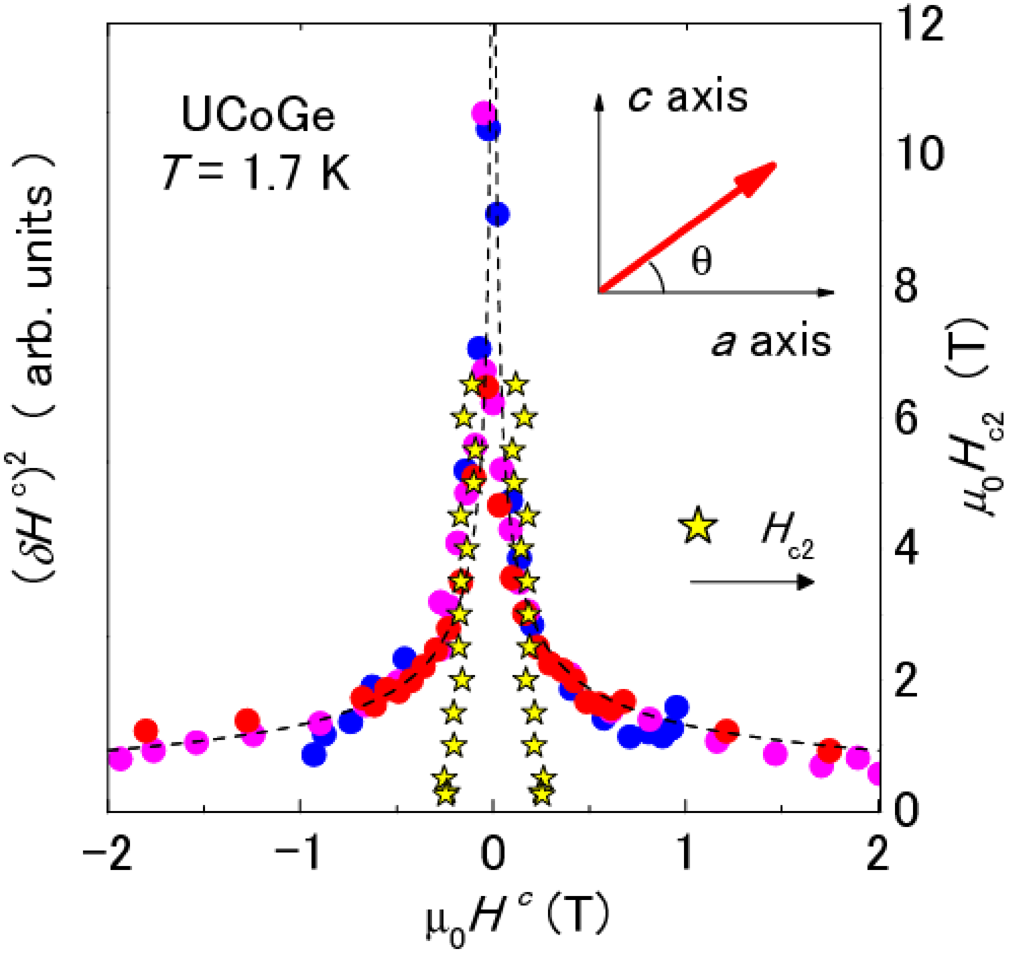
Longitudinal FM fluctuations are significantly suppressed by H^c

Relationship between Magnetic and SC Properties

Transport properties



H_{c2} is plotted agaist c-axis component of *H*.



Superconductivity is observed in the *H* range where $(\delta H^c)^2$ is enhanced

Longitudinal mode is related with SC

Theoretical Studies I (Magnetic Glue) Tada & Fujimoto

Monthoux & Lonzarich PRB’99

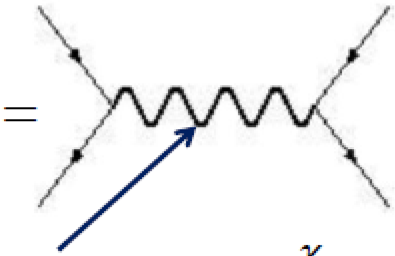
averaged interaction

$$v[\Delta] \equiv \frac{\left\langle \Delta_k V(\vec{k},\vec{k}')\Delta_{k'} \right\rangle_{FS}}{\left\langle \Delta_k^2 \right\rangle_{FS}}$$

v < 0 ; attractive

v > 0 ; repulsive

$$V(k,k') =$$



$$\chi(q) = \frac{\chi_0}{\delta + (\vec{q}-\vec{Q})^2 + |\omega_n|/\Gamma_q}$$

◆For singlet pairing

$$V_s(k,k') \approx g^2 \chi(k-k')$$

$$(ex)Q = (\pi,\pi)$$

$$d_{x^2-y^2} : \Delta_k = \Delta_0(\cos k_x - \cos k_y)$$

$$\Rightarrow v < 0$$

AF favors singlet pairing

◆For triplet pairing

$$V_t(k,k') \approx -\frac{1}{3} g^2 \chi(k-k')$$

$$(ex)Q = (0,0)$$

$$p_x : \Delta_k = \Delta_0 \sin k_x$$

$$\Rightarrow v < 0$$

FM favors triplet pairing

Theoretical Studies II (Magnetic Glue) **Tada & Fujimoto**

single band model on a cubic lattice

$$S = \underbrace{\sum c_k^+[-i\omega_n + \varepsilon_k]c_k}_{\text{kinetic}} - \underbrace{(\vec{h}_{exc} + \mu_B \vec{H}) \cdot \sum c_k^+ \vec{\sigma} c_k}_{\text{exchange and Zeemann}} - \underbrace{\sum \frac{2g^2}{3}\chi(q) S_q^z S_{-q}^z}_{\text{Ising spin fluctuations}}$$

nearly spherical FS

$h_{exc} >> T_{sc}$

$\chi(q)^{-1} \propto \delta + q^2 + |\Omega_n|/q$,

$\delta = \delta(h_{exc}, H_z)$

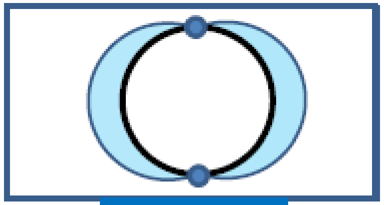
H_z dependent

$t = 50(K), a = 4(A)$

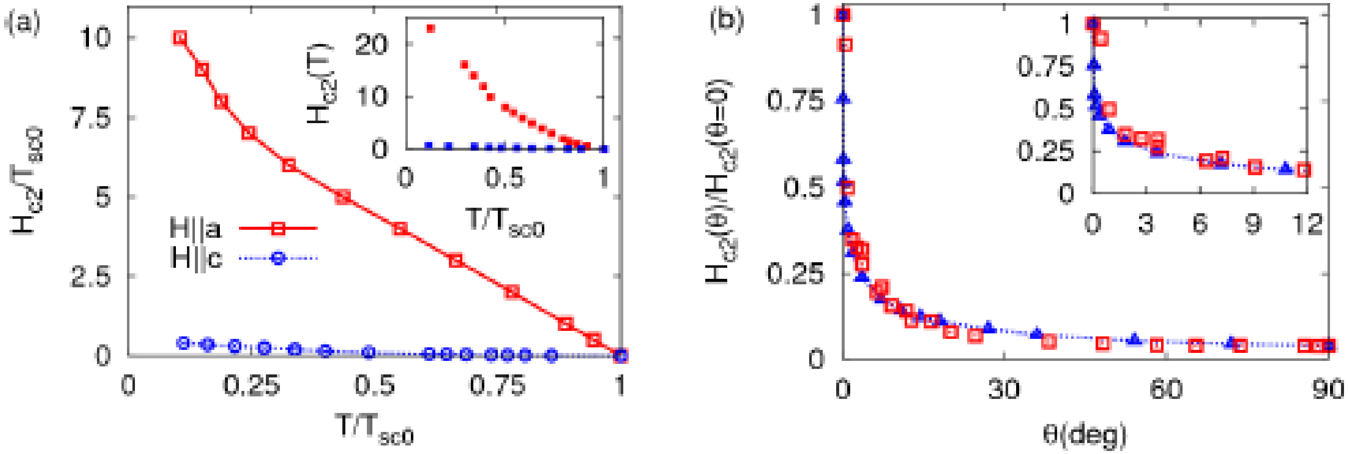
$g^2\chi_0 = 125, h_{exc} = 0.5$

$$\boxed{(\delta H^c)^2 \propto \frac{1}{\sqrt{H^c}}}$$

Spin-triplet superconductivity



A state



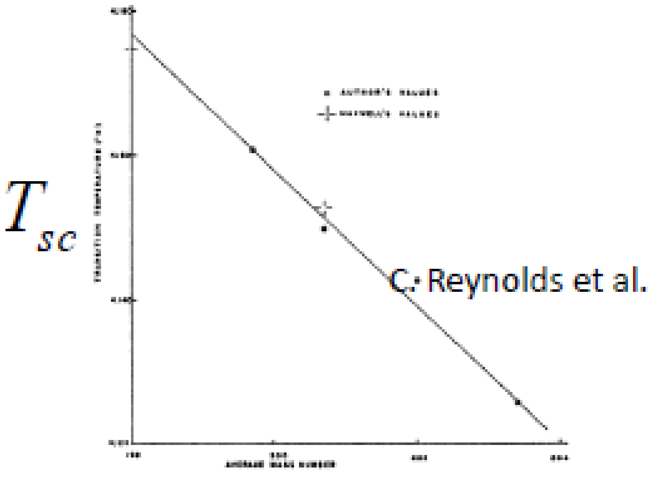
Implication

Phonon-mediated superconductivity

Direct evidence: *Isotope effect*

$$T_c \propto \hbar\omega_D \propto 1/\sqrt{M}$$

M can be changed with isotopes

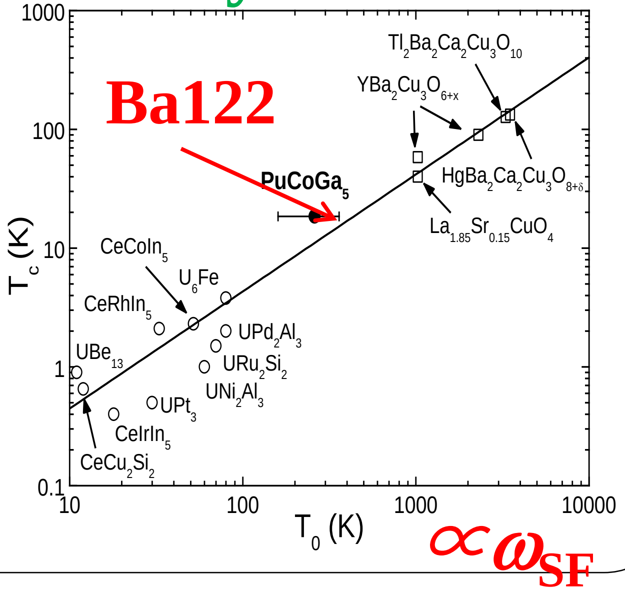


Isotope mass

Spin-fluctuation-mediated superconductivity

$$T_c \propto F\left[\chi''(q)\right]$$

T_{SC}



$\propto \omega_{SF}$

Most of case, AFM fluctuation,

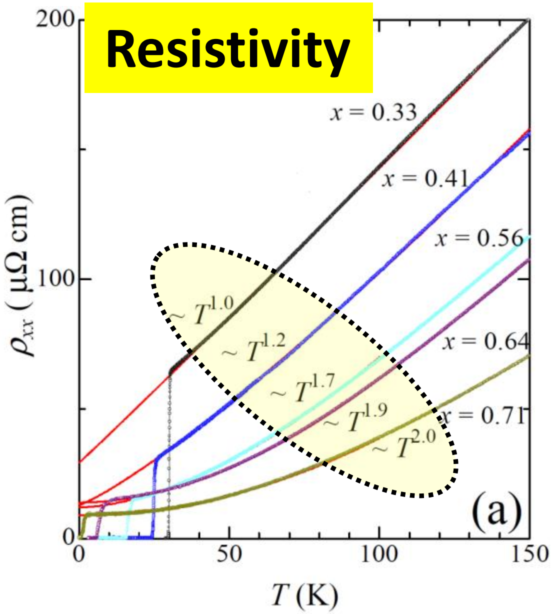
e.g. high-T_c, HF and iron-pnictide SC

There is no conjugate physical parameter.

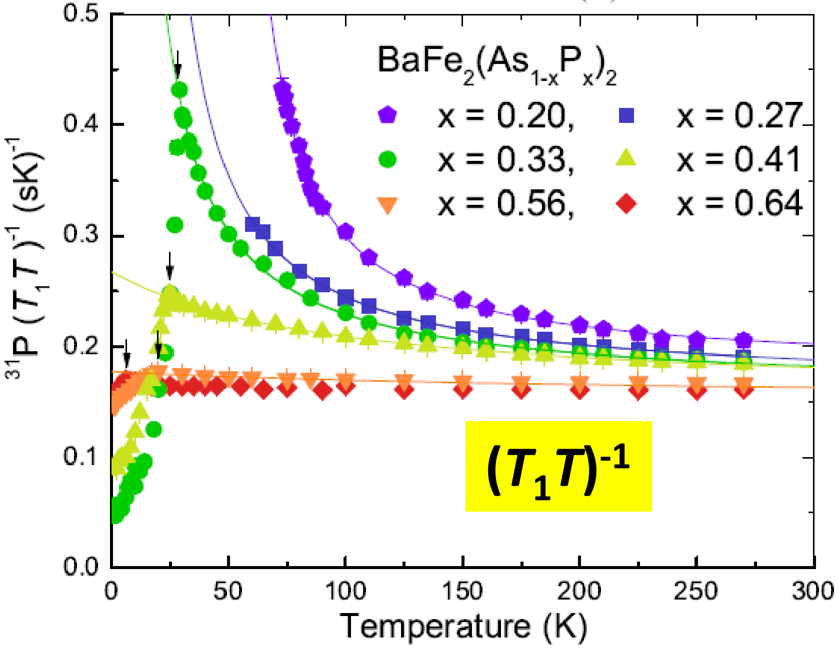
In FM SF, $\chi''(q=0) = f(H)$

Summary of Experimental Results : ρ , m^* and $(T_1T)^{-1}$

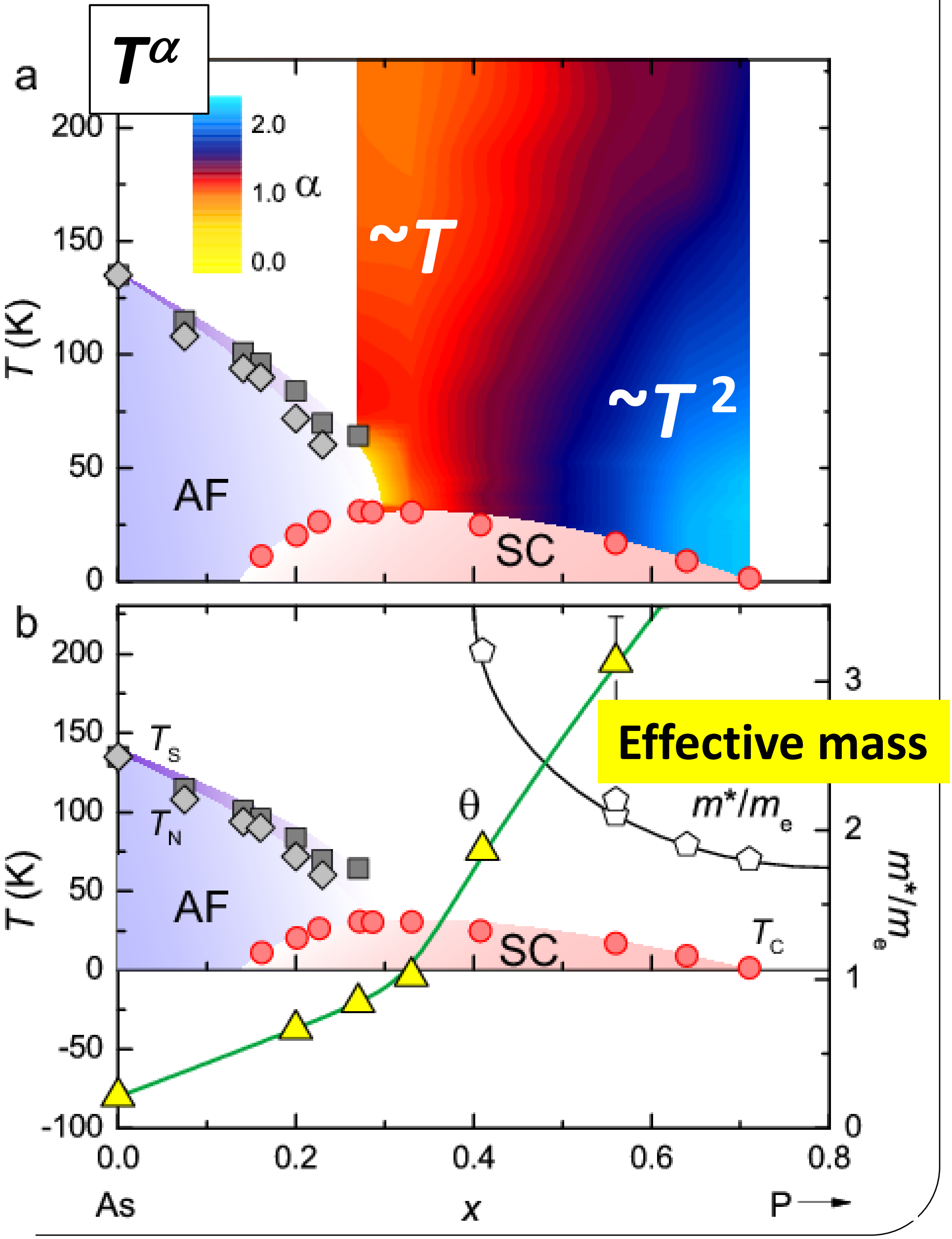
YN *et al.*,
PRL **105**
(2010) 107003.



Resistivity



$(T_1T)^{-1}$



T^α

Effective mass

Summary

T. Ohta *et al.* JPSJ 79, 023707 ('10)
Y. Ihara *et al*, PRL 105 206403 ('10)
T. Hattori *et al*, PRL 108, 066403 ('12)

From Co NQR/NMR studies on UCoGe, we show that

1) Superconductivity occurs in the FM region

⇒ Microscopic coexistence of FM and SC

2) FM signal and PM signal coexist between 2.5 and 1K.

⇒ 1st-order like transition at T_{Curie}

3) Ising-type FM fluctuations develop at low *T*.

⇒ Longitudinal (c-axis) mode of the moments

4) The FM fluctuations are very sensitive to H_c and are related with the SC properties.

We suggest that spin-triplet superconductivity is induced by the Ising-type FM fluctuations

5) T_{Curie} is suppressed by *H*// *b*, but is not by *H*//*a*.

Critical FM fluctuations enhance T_c

$$\boxed{\left(\delta H^{c}\right)^{2} \propto \frac{1}{\sqrt{H^{c}}}}$$

Consistent with the theoretical prediction,

P. Monthoux and G. G. Lonzarich, PRB 59, 14598 ('99)
S. Fujimoto, JPSJ 73 2061 (04)