Exotic Magnetism and Superconductivity in Actinide compounds

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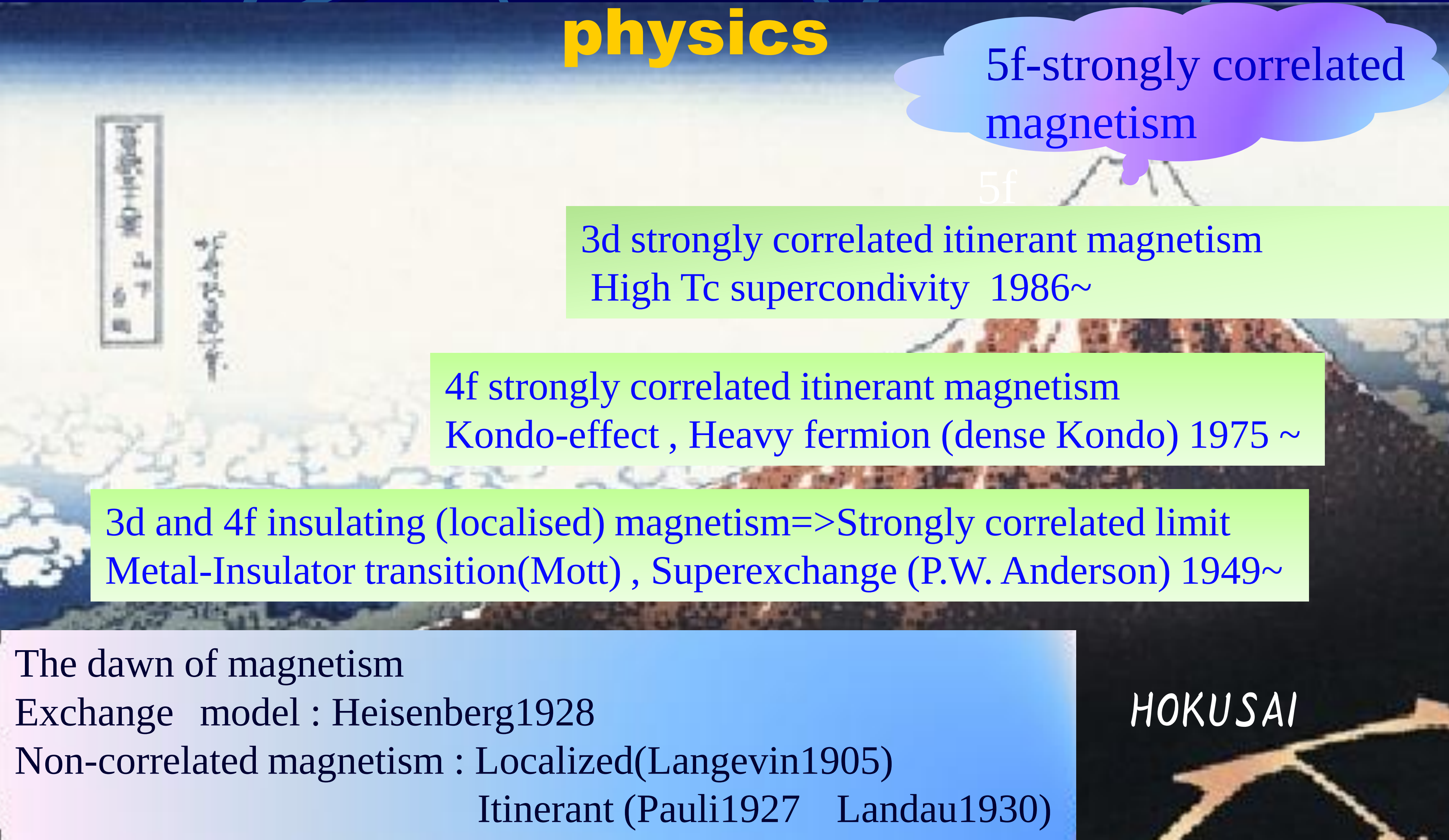
5f-system
Last unexplored summit for strongly correlated electron physics



5f-strongly correlated magnetism

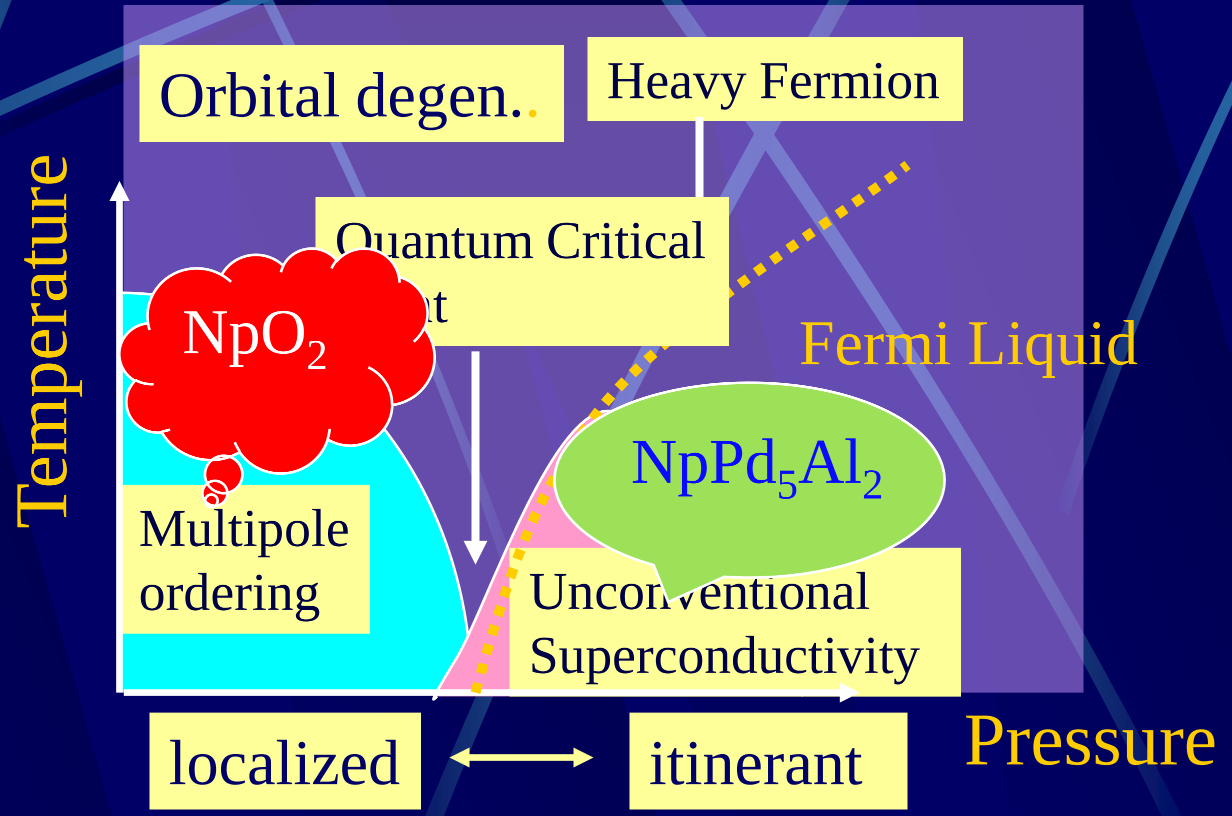
3d strongly correlated itinerant magnetism
High Tc supercondivity 1986~

4f strongly correlated itinerant magnetism
Kondo-effect , Heavy fermion (dense Kondo) 1975 ~

3d and 4f insulating (localised) magnetism=>Strongly correlated limit
Metal-Insulator transition(Mott) , Superexchange (P.W. Anderson) 1949~

The dawn of magnetism
Exchange model : Heisenberg1928
Non-correlated magnetism : Localized(Langevin1905)
Itinerant (Pauli1927 Landau1930)

Physical problems in strongly correlated 5f systems



Orbital degen..

Heavy Fermion

Temperature

Quantum Critical
t

NpO_2

Fermi Liquid

$NpPd_5Al_2$

Multipole
ordering

Unconventional
Superconductivity

localized ⟷ itinerant

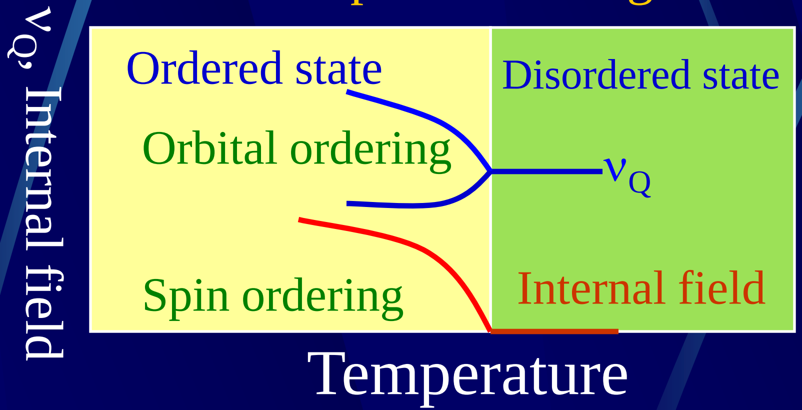
Pressure

Schematic Phase diagram of 5f systems

NMR for Identification of Exotic phases

Internal field =>Zeeman Interaction (Shift)
Orbital ordering=> Quadrupolar Interaction (ν_Q)
Fluctuation around Phase transition=> nuclear relaxations (T_1,T_2)

Multipolar ordering



ν_Q, Internal field

Ordered state

Disordered state

Orbital ordering

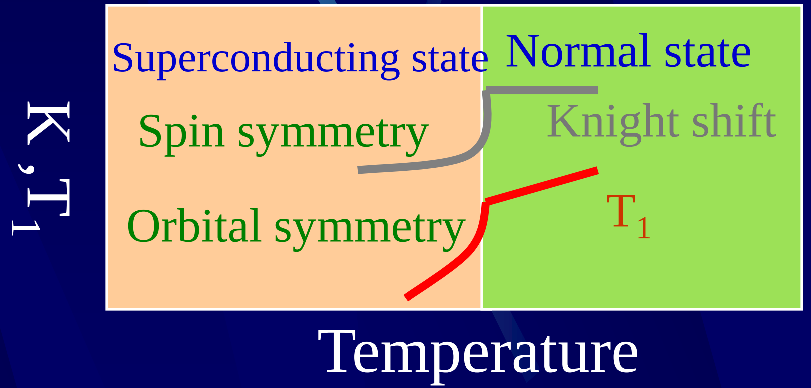
ν_Q

Spin ordering

Internal field

Temperature

Superconducting symmetry



K, T_1

Superconducting state

Normal state

Spin symmetry

Knight shift

Orbital symmetry

T_1

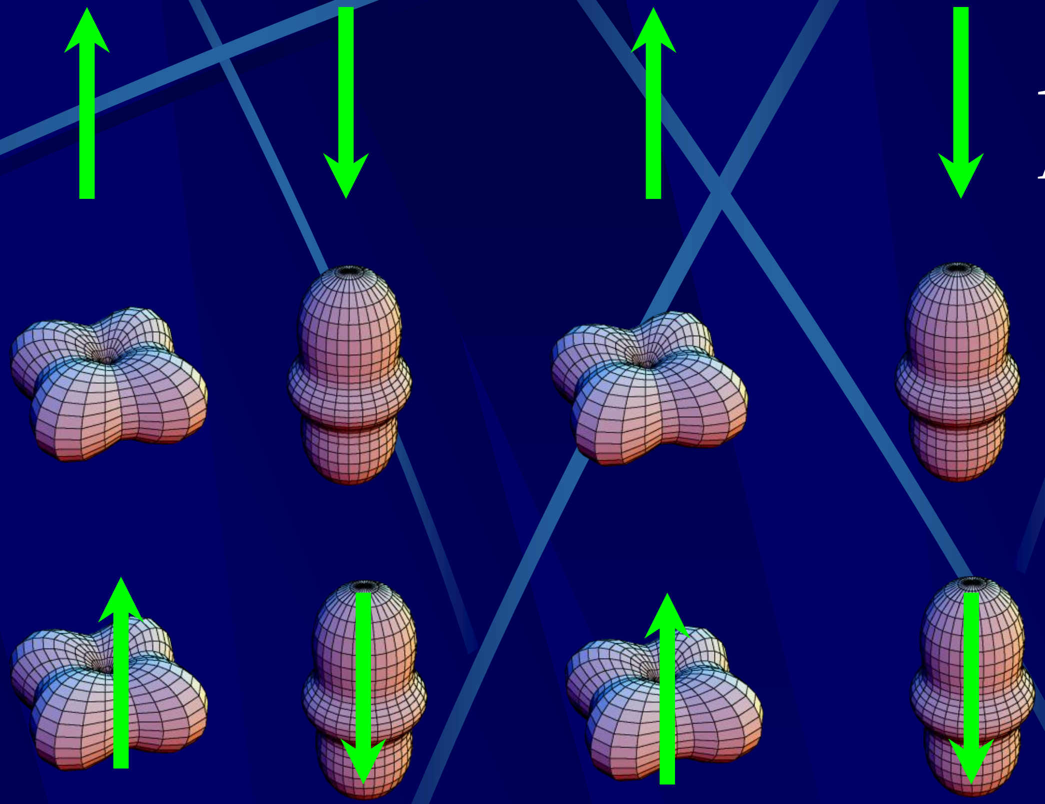
Temperature

Outline of talk

- Introduction to Multipolar Ordering
- ^{17}O-NMR study of Octupolar ordering in NpO_2 and AmO_2
- Introduction to Unconventional superconductivity
- Al-NMR study of d-wave superconductivity in $NpPd_5Al_2$

Part I Multipolar ordering

What is multipolar ordering?



Spin

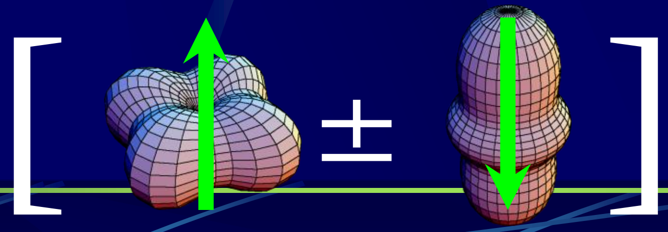
AFM
Antiferromagnet

Orbital

AFQ
Antiferro
quadruploar

Spin and
Orbital

That’s All？



No!

Multipole moments

electric multipoles

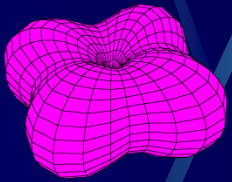
monopole

$$\int d\mathbf{r}\varphi^*(\mathbf{r})\varphi(\mathbf{r})$$

charge:
charge ordering

quadrupole

$$\int d\mathbf{r}\varphi^*(\mathbf{r})xy\varphi(\mathbf{r})$$



anisotropy in charge distribution:
usual orbital order → Jahn-Teller

magnetic multipoles

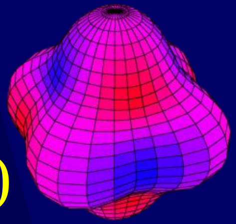
dipole

$$\int d\mathbf{r}\varphi^*(\mathbf{r})M_x\varphi(\mathbf{r})$$

spin:
usual magnetic order

octupole

$$\int d\mathbf{r}\varphi^*(\mathbf{r})xyM_z\varphi(\mathbf{r})$$



anisotropy in spin distribution

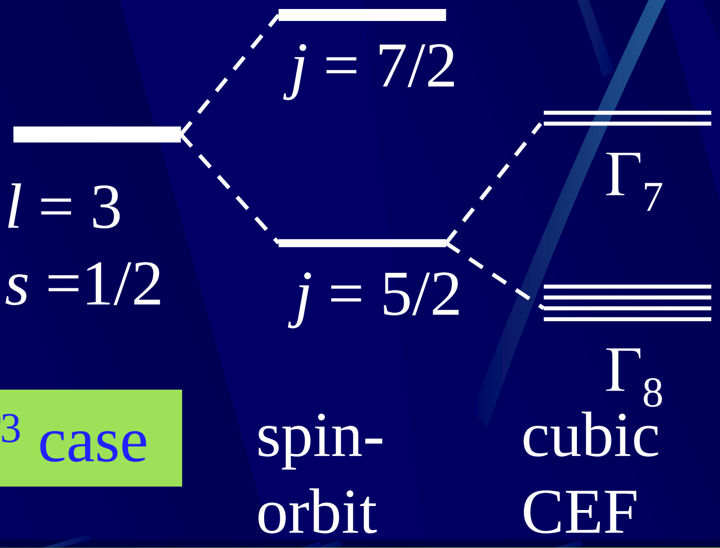
NpO_2

Octupolar ordering

Electronic state of AnO_2

- AnO_2 (UO_2, NpO_2, PuO_2…)
- Well studied as nuclear fuel, but low temperature properties are still mysterious!

Highly degenerated f-levels due to cubic symmetry



j = 7/2

Γ_7

l = 3

s =1/2

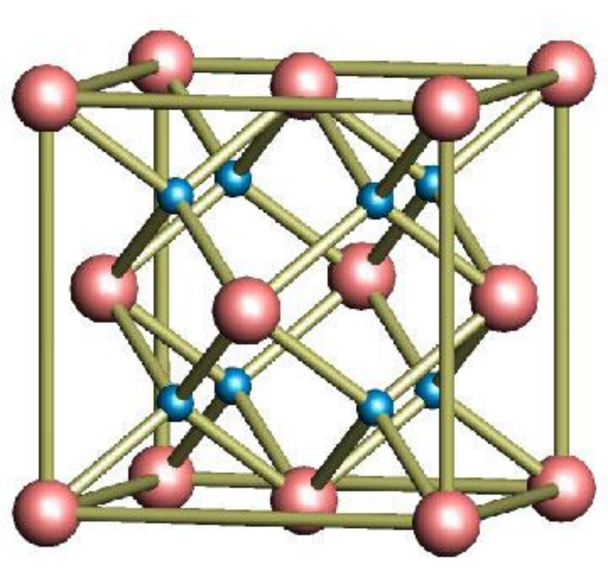
j = 5/2

Γ_8

spin-orbit

cubic CEF

Np^{4+} f^3 case



Crystal structure of AnO_2

Mysterious ordering in NpO_2

UO_2 is AFM PuO_2 is non magnetic

What is the order parameter of NpO_2 ?

AFM → No

dipole moment = 0

Neutron, Mössbauer

$\mu_0 < 0.01\mu_B$/Np.

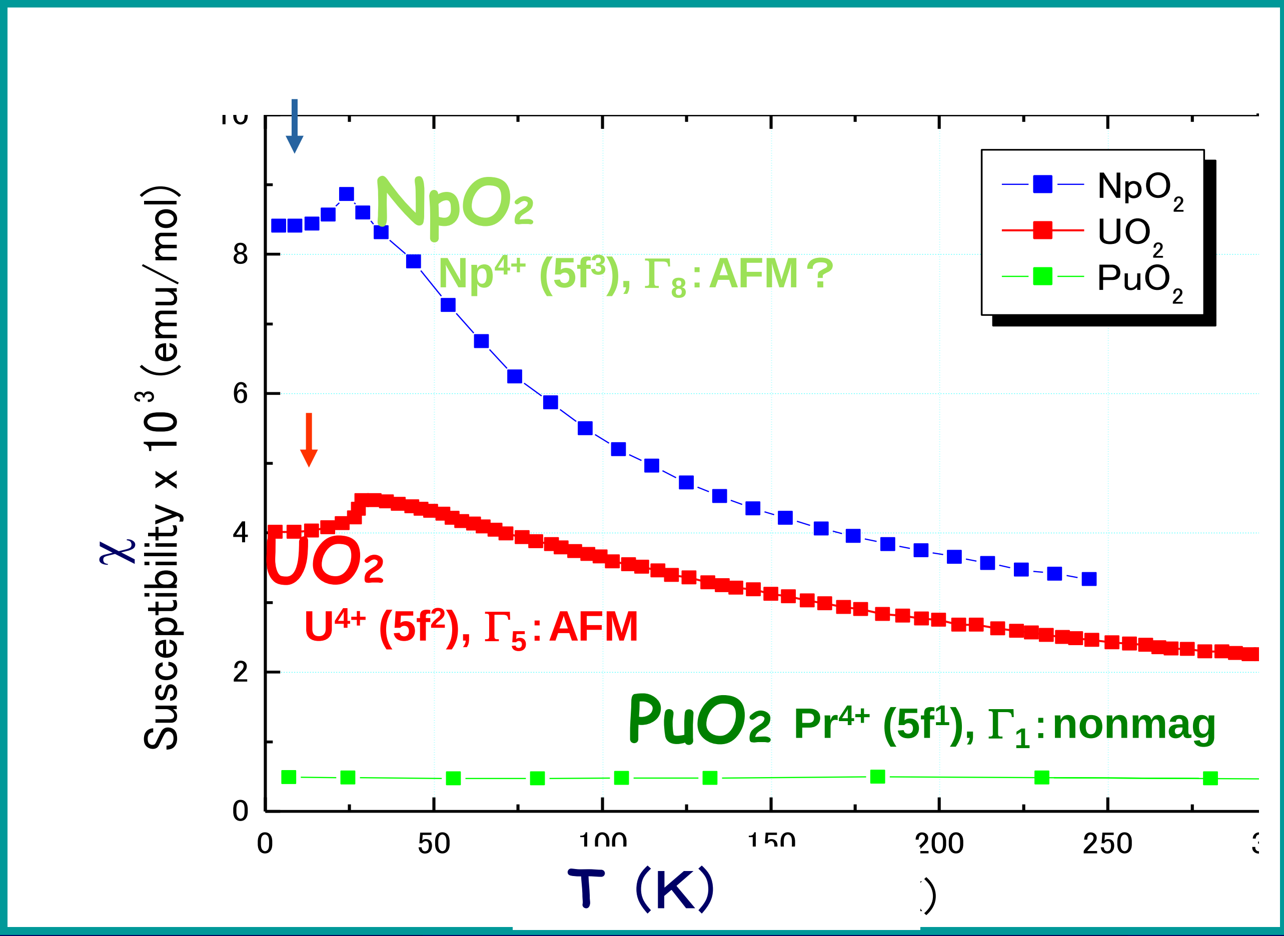
AFQ → No?

Broken TR sym.

Susceptibility, μSR

No lattice distortion at T_0

→Octupolar(AFO)?



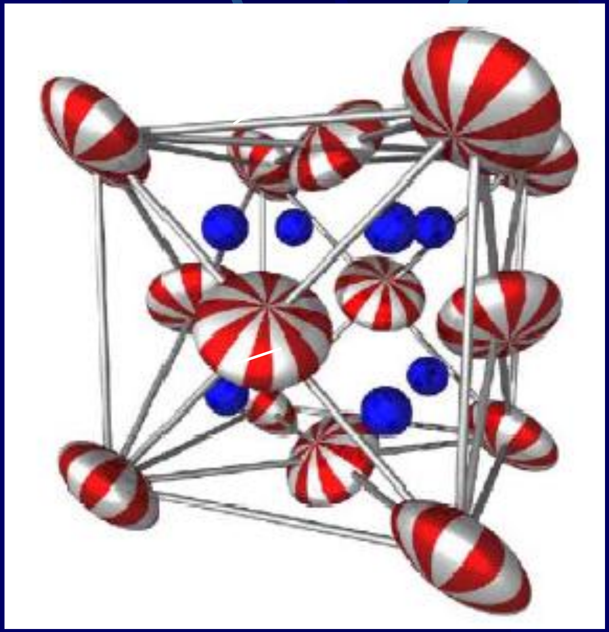
T-dependence of Magnetic susceptibility

Magnetic X-ray scattering

AFO(Γ_5): Primary order parameter induces AFQ

↓

AFQ(Γ_5) is observed: secondary order parameter



Triple-q AFQ

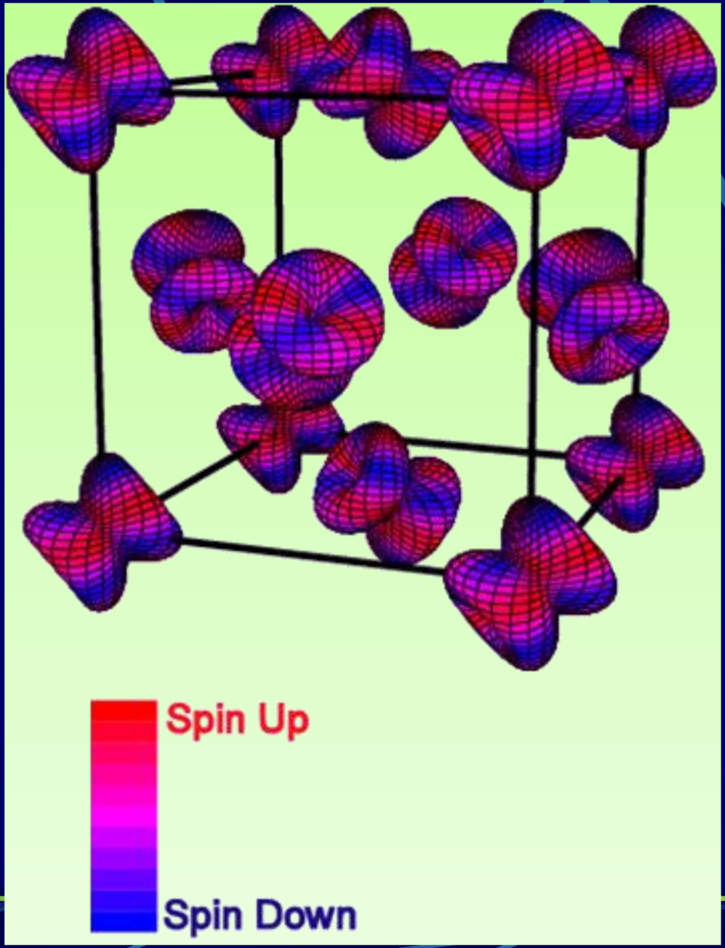
AFQ ordered structure from Magnetic X-ray scattering

J. A. Paixao et al, PRL**89** (2002)

Micro scopic j-j coupling model for NpO_2

K. Kubo and T. Hotta PRB **72**, 144401 (2005).

Fcc: Γ_{5u} longitudinal triple-q AFO



^{17}O-NMR in the ordered phase

NMR spectrum is splitted in the ordered phase

→**Hyperfine field due to ordered moment**

Emergence of two oxygen sites

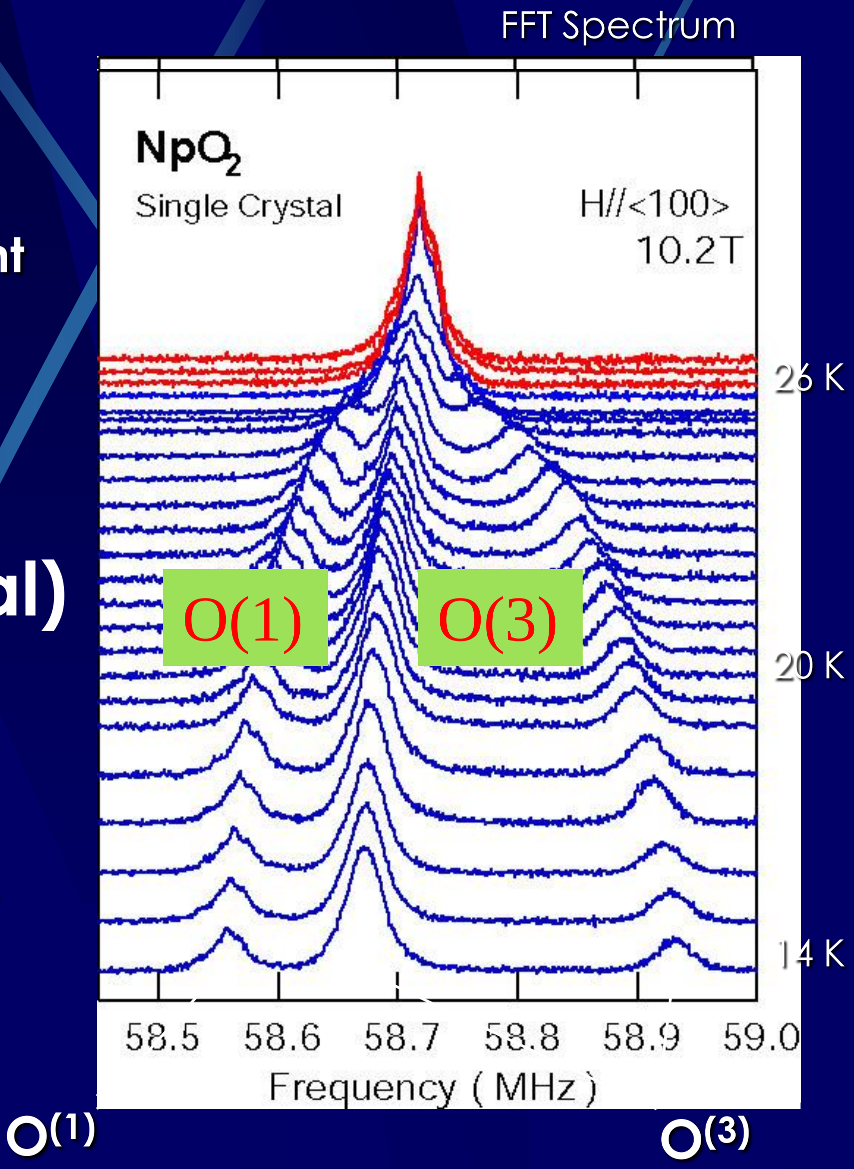
O(1) : **isotropic**

O(3) : **anisotropic(uniaxial)**

Sites number ratio

O(1):O(3)=1:3

Y.Tokunaga *et al.* , PRL 94(2005)

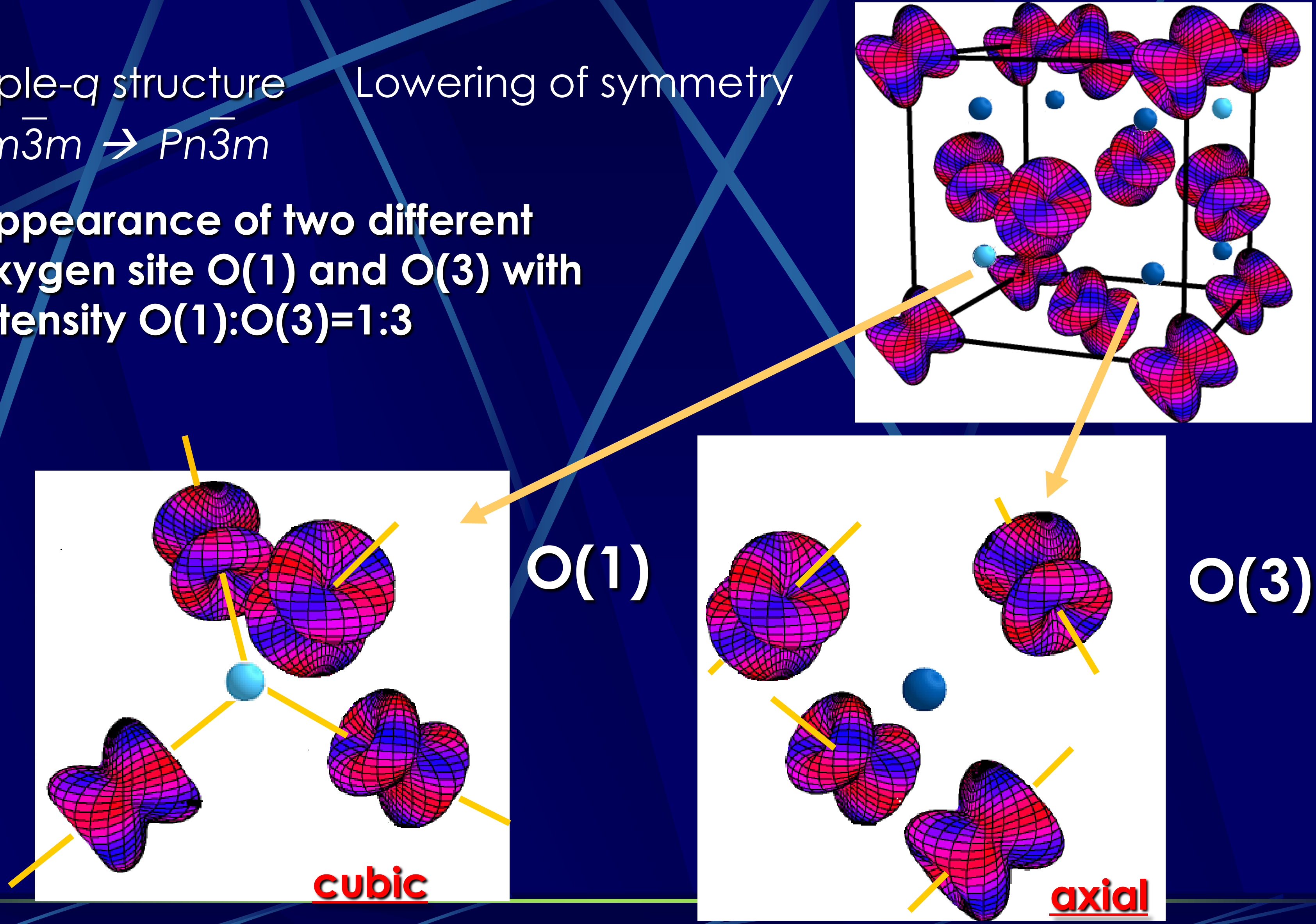


Origin of two Oxygen sites

Triple-*q* structure   Lowering of symmetry

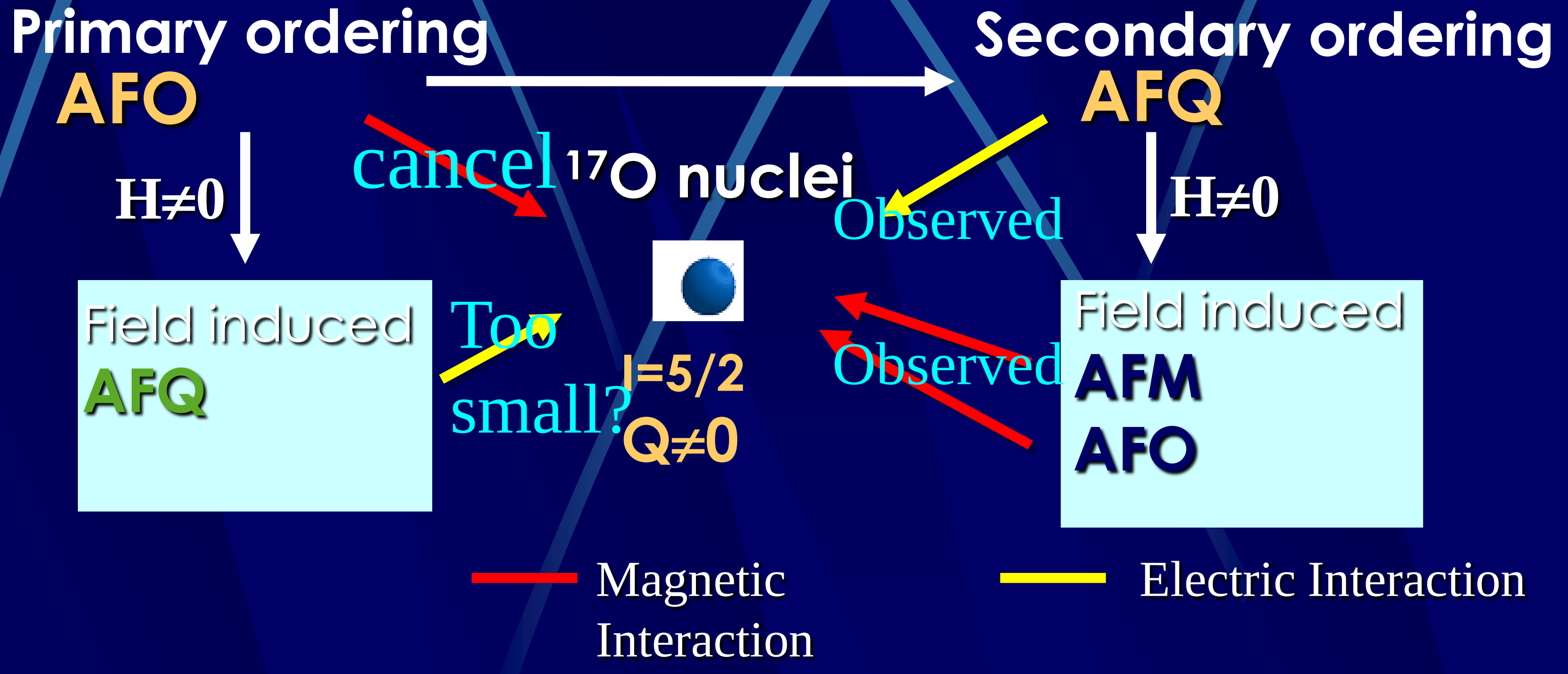
$Fm\bar{3}m \rightarrow Pn\bar{3}m$

Appearance of two different oxygen site O(1) and O(3) with intensity O(1):O(3)=1:3



Hyperfine coupling in NpO_2

O.Sakai *et al*, JPSJ 74 (2005)



Primary ordering

AFO

H≠0

Field induced

AFQ

cancel

Too small?

^{17}O nuclei

I=5/2

Q≠0

Observed

Observed

Secondary ordering

AFQ

H≠0

Field induced

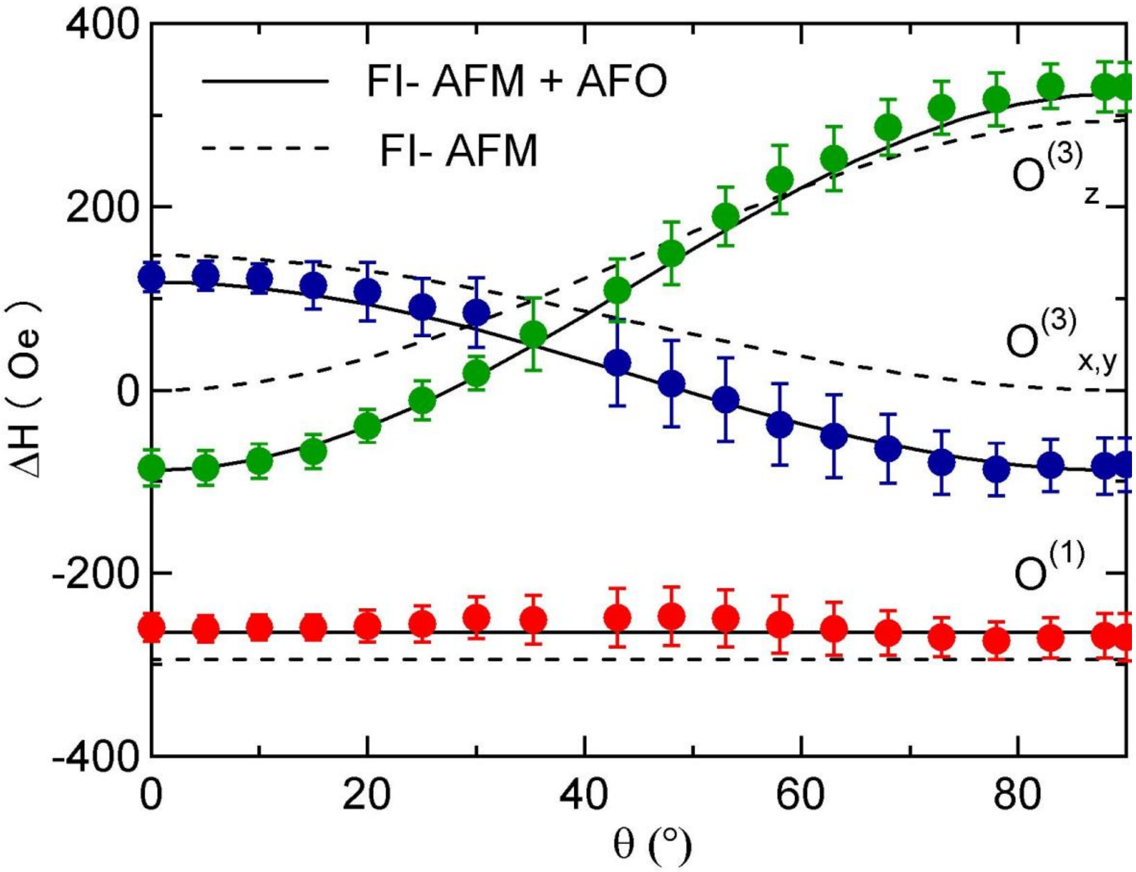
AFM

AFO

Magnetic Interaction

Electric Interaction

Comparison with model



$$
\begin{aligned}
H_{\mathrm{hf}}(\boldsymbol{\rho}) = {} & \frac{1}{\sqrt{N}} \sum_{\boldsymbol{q}} \mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{\rho}} I_x(\boldsymbol{\rho}) \\
& \times \Big[c_{1,1} \frac{4}{\sqrt{4}} \underline{J_{x,\boldsymbol{q}}} (c_x c_y c_z - \mathrm{i}\,\mathrm{sg}(\boldsymbol{\rho}) s_x s_y s_z) \\
& + c_{1,2} \frac{4}{\sqrt{16}} \Big\{ \underline{J_{y,\boldsymbol{q}}} (\mathrm{i}\,\mathrm{sg}(\boldsymbol{\rho}) s_z c_x c_y - c_z s_x s_y) \\
& + \underline{J_{z,\boldsymbol{q}}} (\mathrm{i}\,\mathrm{sg}(\boldsymbol{\rho}) s_y c_z c_x - c_y s_z s_x) \Big\} \\
& + c_{1,3} \frac{4}{\sqrt{8}} \Big\{ \underline{T^{\beta}_{y,\boldsymbol{q}}} (\mathrm{i}\,\mathrm{sg}(\boldsymbol{\rho}) s_z c_x c_y - c_z s_x s_y) \\
& - \underline{T^{\beta}_{z,\boldsymbol{q}}} (\mathrm{i}\,\mathrm{sg}(\boldsymbol{\rho}) s_y c_z c_x - c_y s_z s_x) \Big\} \\
& + c_{1,4} \frac{4}{\sqrt{4}} \underline{T_{xyz,\boldsymbol{q}}} (\mathrm{i}\,\mathrm{sg}(\boldsymbol{\rho}) s_x c_y c_z - c_x s_y s_z) \Big] \\
& + (\text{cyclic permutation of } x,\ y \text{ and } z).
\end{aligned}
$$

▬ AFM

▬ AFO

$H_{FI-AFM} + \alpha\, \underline{H_{FI-AFO}} \,(T^{\beta} + T_{xyz})$

$\alpha = -0.2$

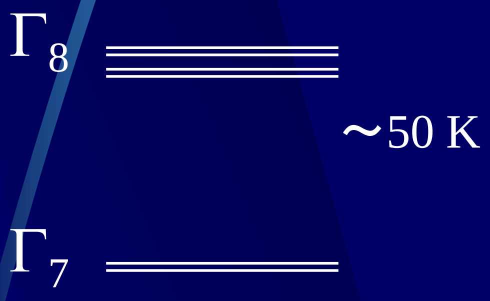
AFO contributions!

Beyond NpO$_2$: AmO$_2$ (Am^{4+} : 5f^{5})

Susceptibility : AFM-like phase transition at 8.5K

Neutron, Mössbauer : No-dipolar moment below 8.5 K.

→Multipolar ordering ? ^{17}O-NMR in progress



From *T*-dependence of susceptibility

D.G.Karraker, The Journal of Chemical Physics, Vol.63, 3174 (1975)

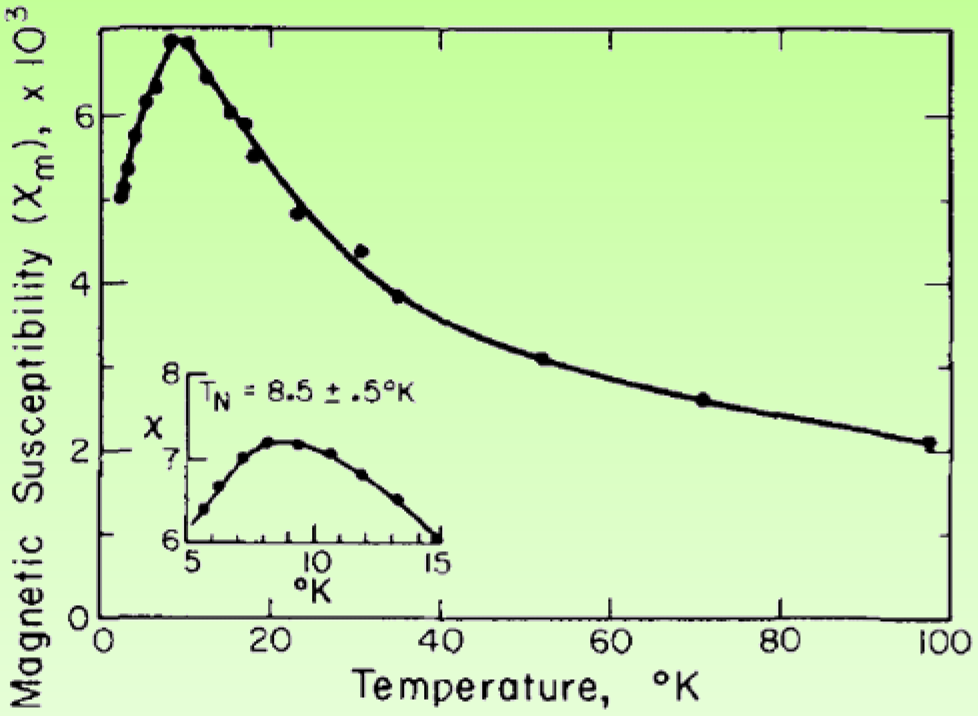


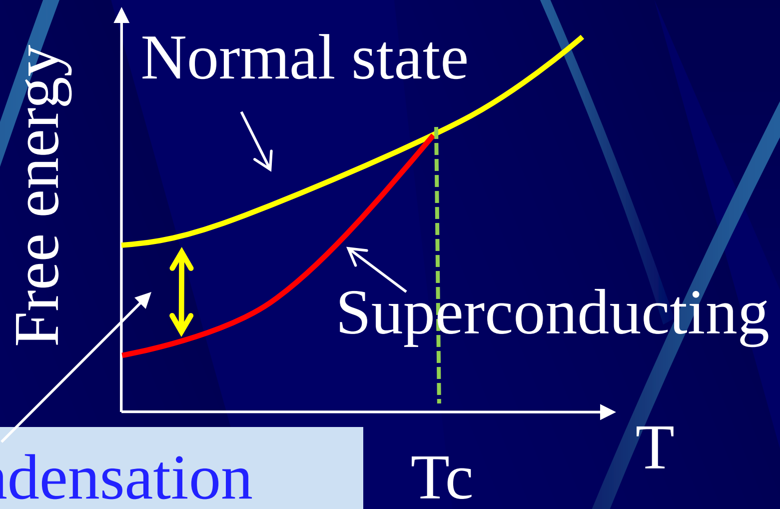
FIG. 1. Antiferromagnetic transition of ^{243}AmO$_2$.

Part II Unconventional Superconductivity

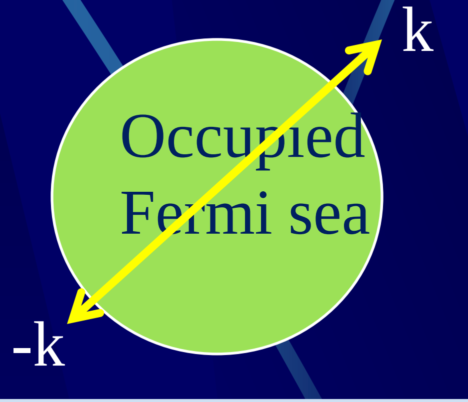
Superconductivity : New condensed state with formation of

1)Superconducting condensation energy (energy gap)

2) Copper paring of two electrons



Condensation energy below Tc



k and – k Cooper paring in k-space

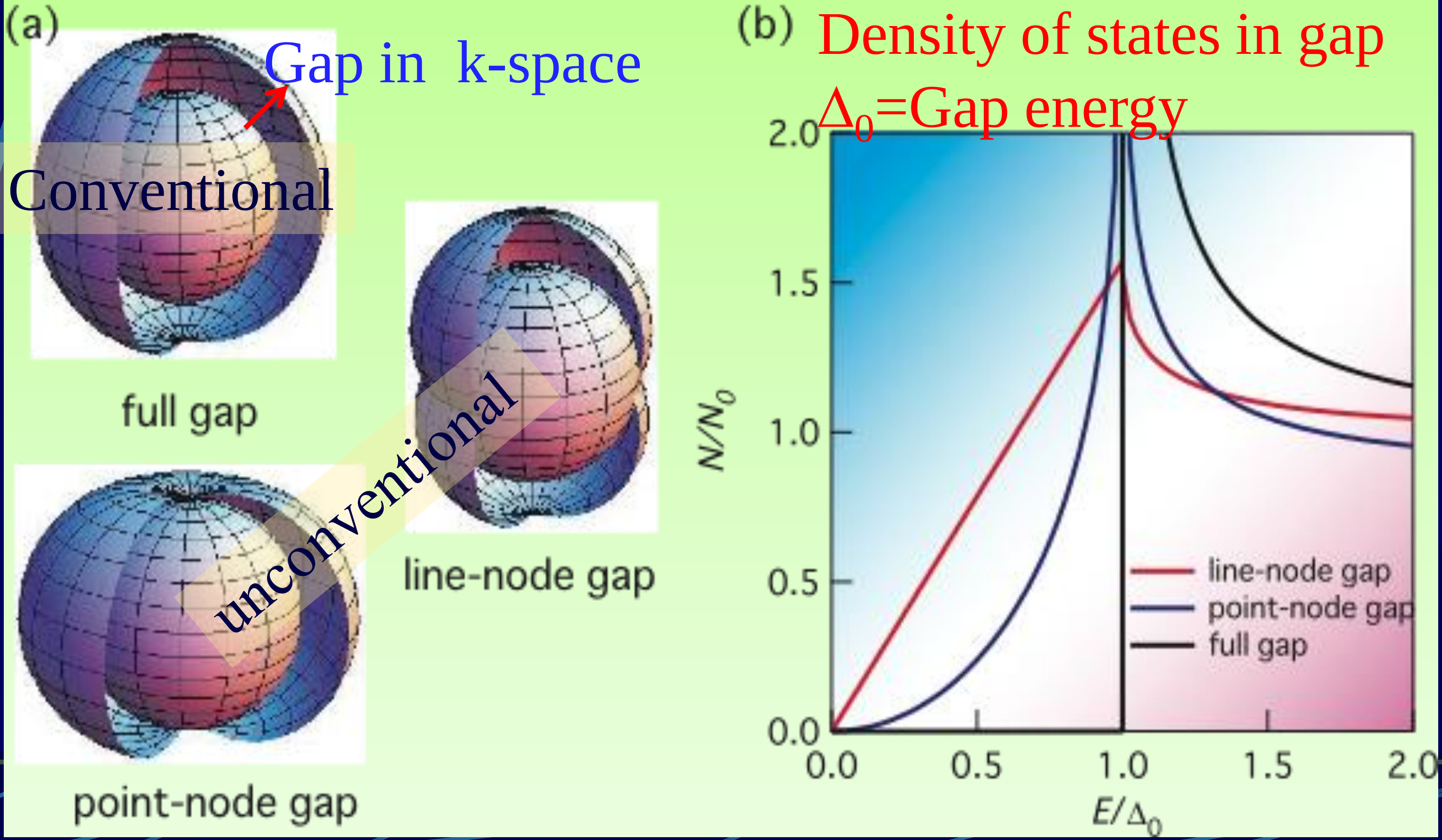
(Inversion symmetrical case)

What's happens in unconventional Superconductivity?

Anisotropic Superconducting gap

Conventional => isotropic full superconducting gap

Unconventional => anisotropic partial superconducting gap



Gap in k-space

Conventional

unconventional

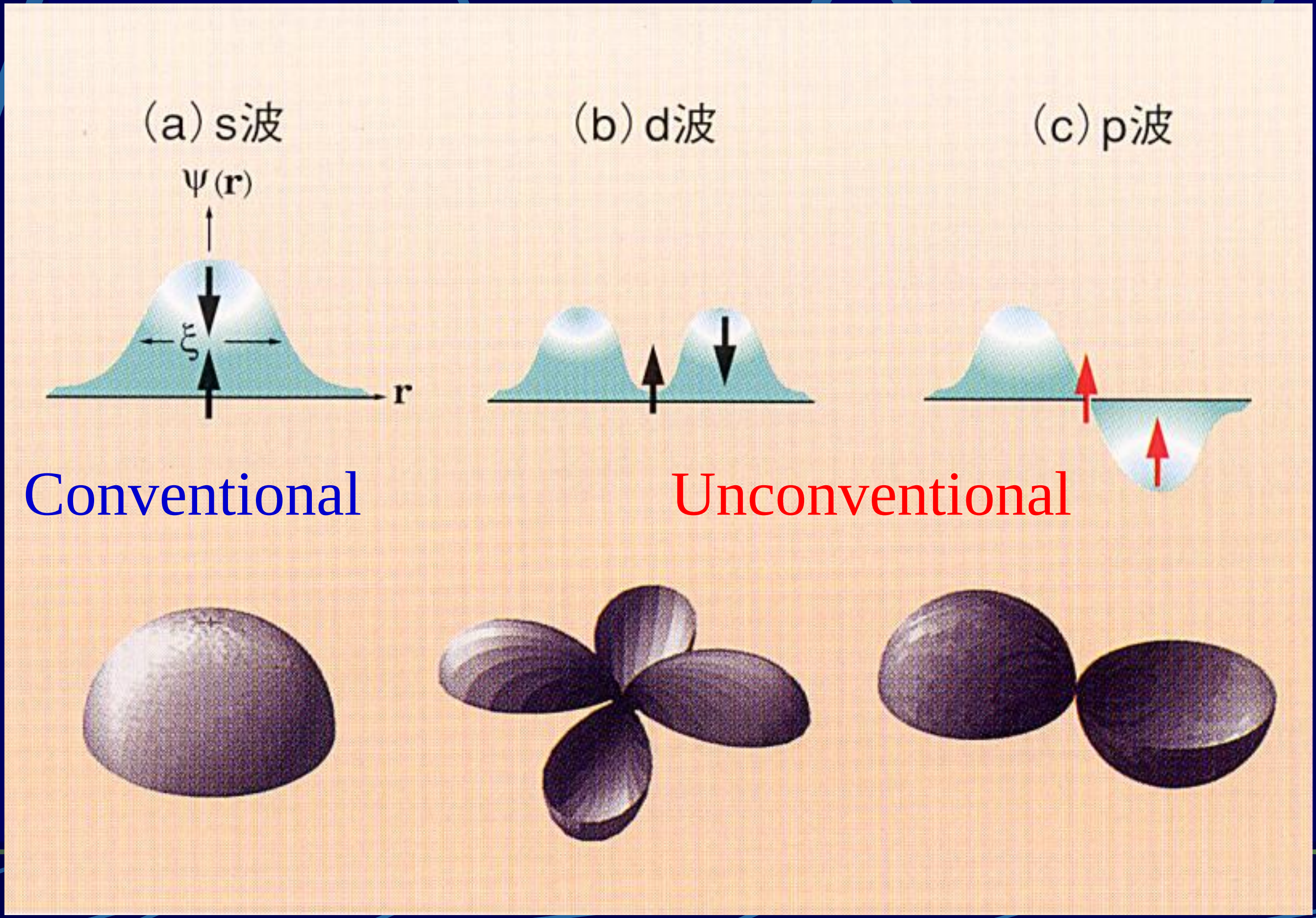
Density of states in gap

Δ_0=Gap energy

Alternative Spin Paring

Conventional => Singlet paring (s-wave)

Unconventional => Singlet (d-wave) or Triplet (p-wave)



Conventional

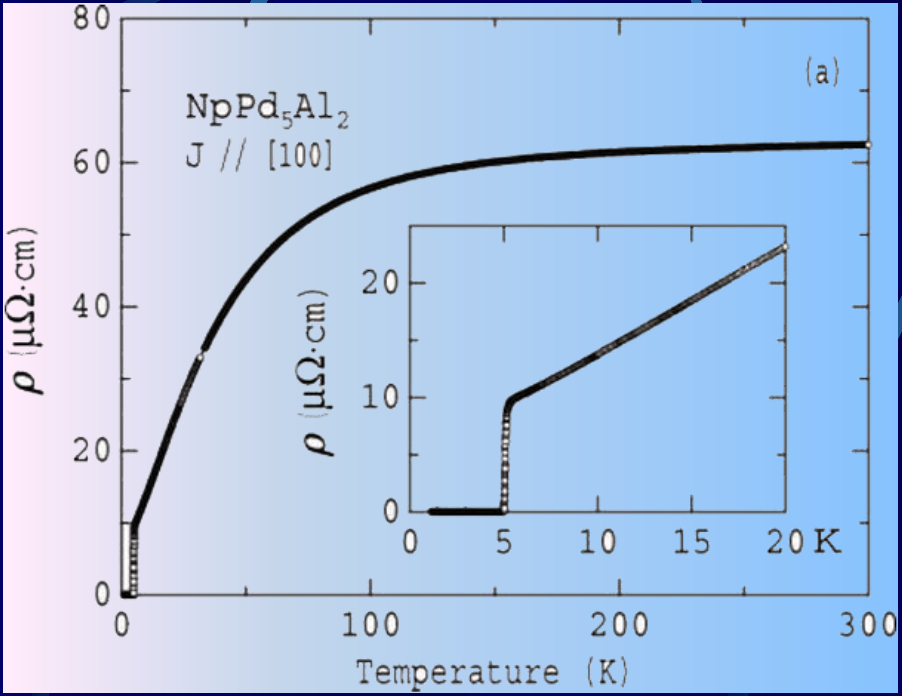
Unconventional

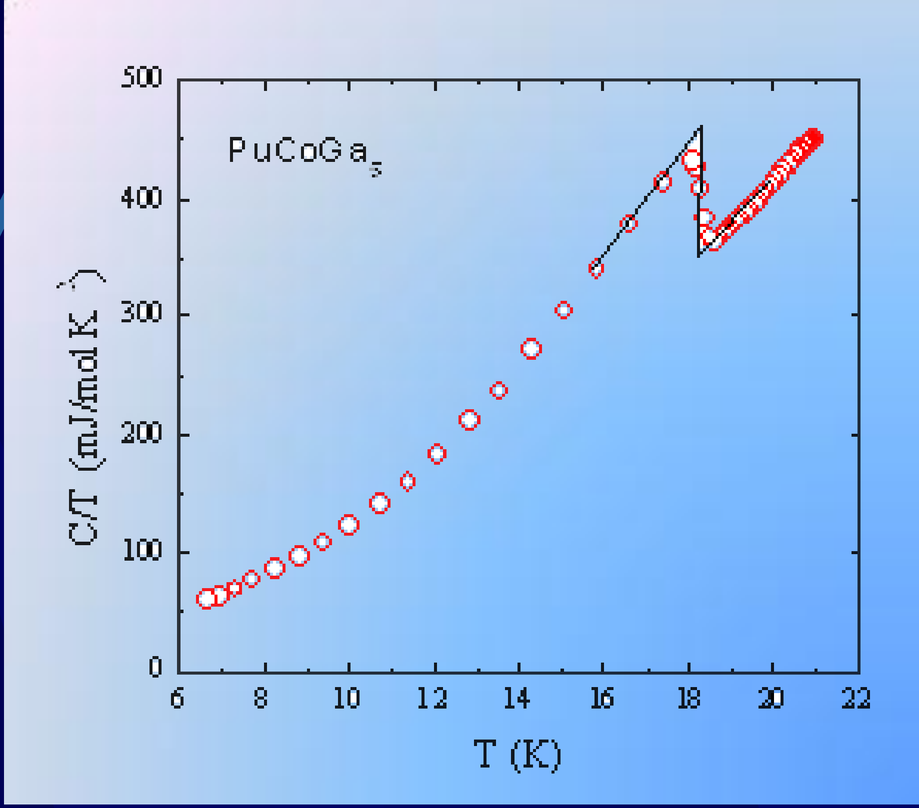
$NpPd_5Al_2$ $PuRhGa_5$ $PuCoGa_5$ d-wave superconductors

Np, Pu based New superconductors

Specific heat is very large $\gamma \sim 10^2 \mathrm{mJ/K^2 mol}$ => Heavy fermion

Tc is very high ~10K compared with ~ 1K in Ce heavy fermion systems





$NpPd_5Al_2$ Tc=5K
D. Aoki et al JPSJ 2007
Next talk !

$PuCoGa_5$ Tc=18K
J. Sarrao et al Nature 2002

Characteristics of Crystal Structures

- Similarities

  Tetragonal

  Lattice parameter of *a*-axis

  Layered structure

- Dissimilarities

  Lattice parameter of c-axis

  - ~2 times longer

  Actinide layers stacking in alternate phase along c-axis

  - bcc lattice

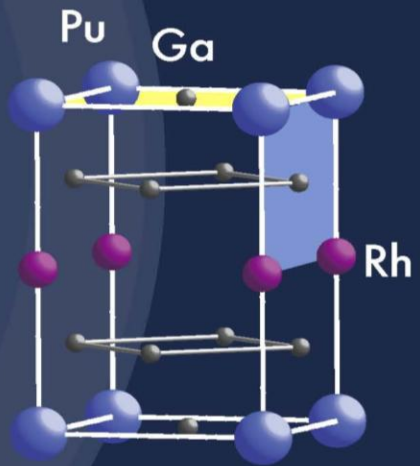
  Nearest hybridization path

  - 5*f* (Pu) - 4*p* (Ga)
  - 5*f* (Np) - 4*d* (Pd)

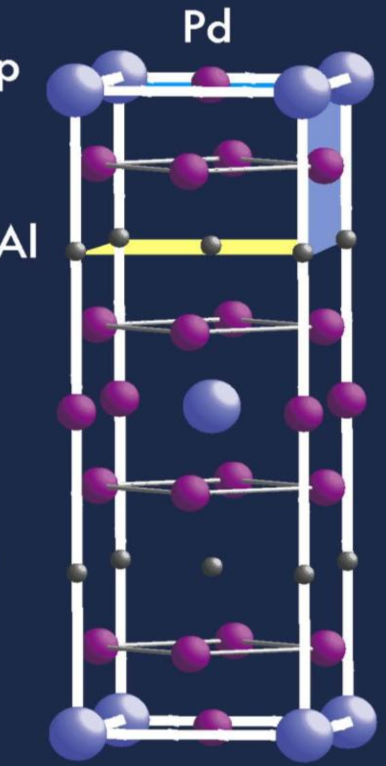
PuRhGa$_5$ & NpPd$_5$Al$_2$

a=4.30 Å
c=6.86 Å

a=4.15 Å
c=14.7 Å



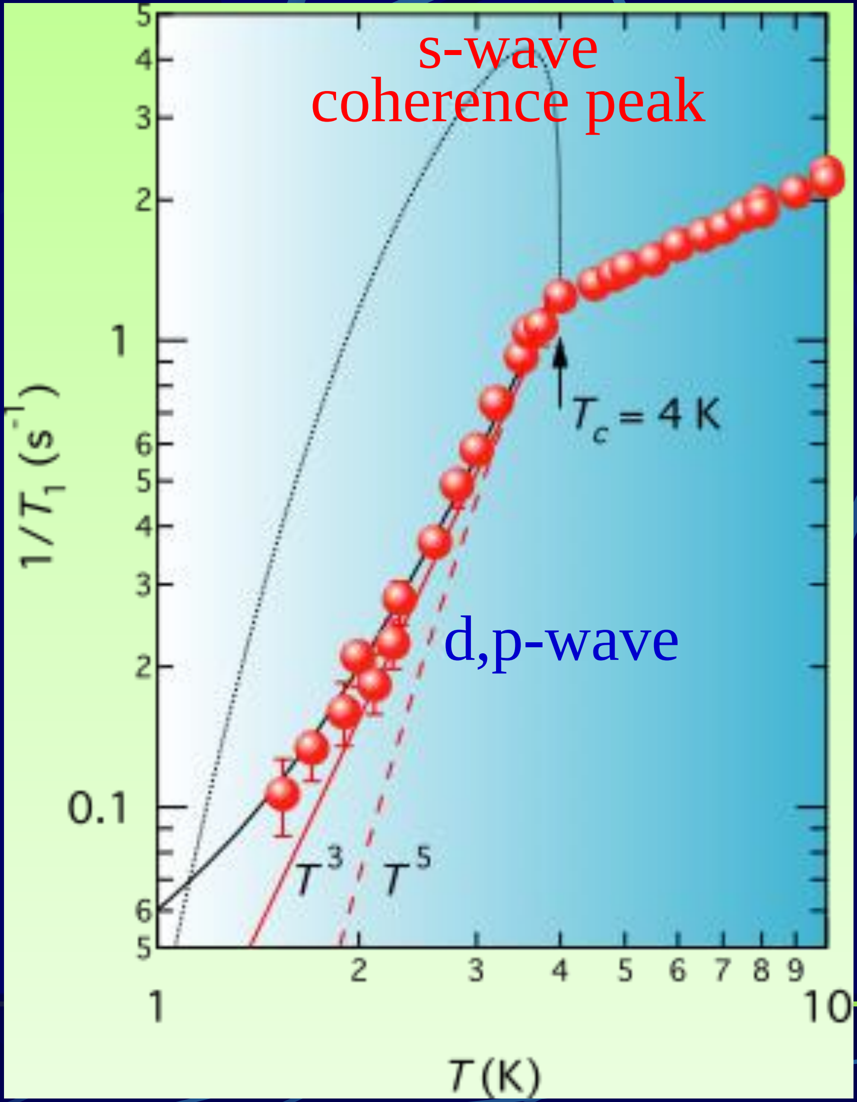
tetra. HoCoGa$_5$-type



tetra. ZrAl$_5$Ni$_2$-type

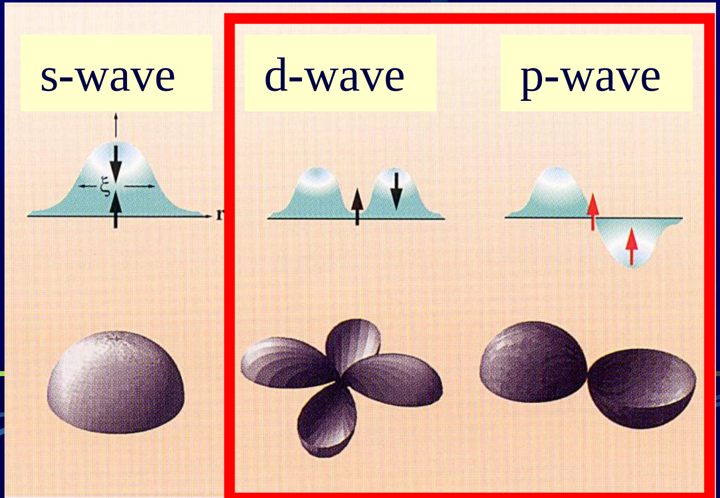
Spin-lattice relaxation rate $1/T_1$ in $NpPd_5Al_2$

H. Chudo et al
JPSJ 2008

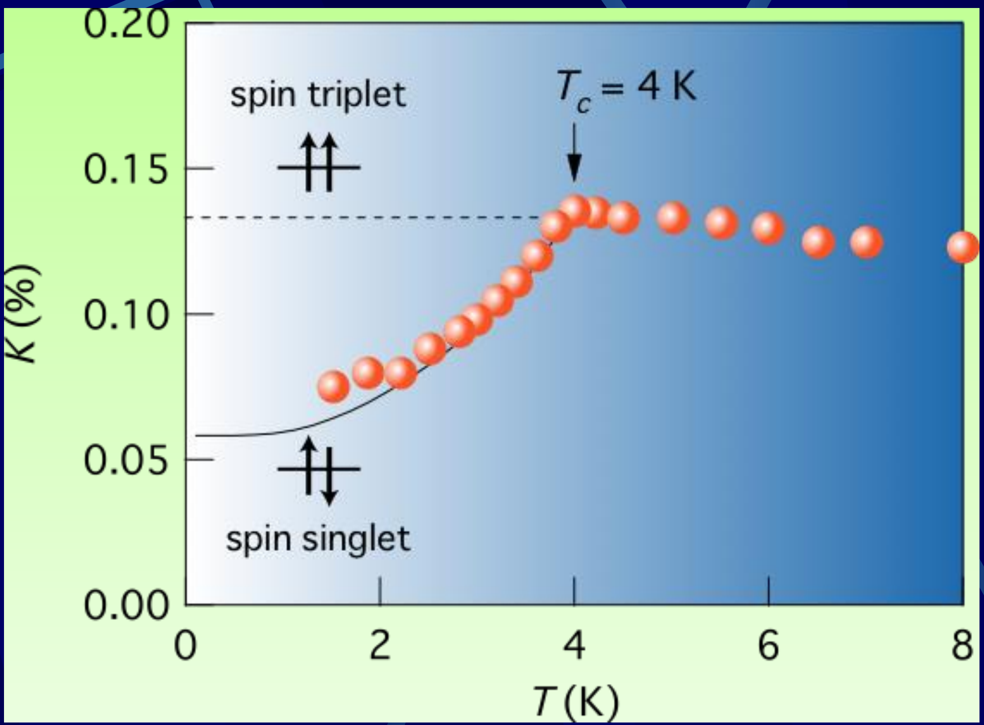


No coherence peak at Tc
$1/T_1 \propto T^3$ below T_c

anisotropic SC gap
(d or p-wave)



Knight shift in the superconducting state of $NpPd_5Al_2$



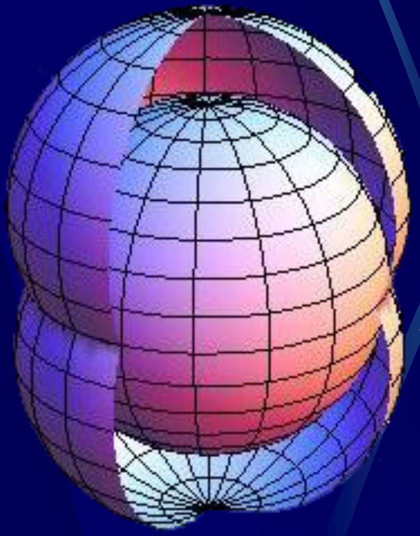
H. Chudo et al
JPSJ 2008

T-dependence of Knight shift

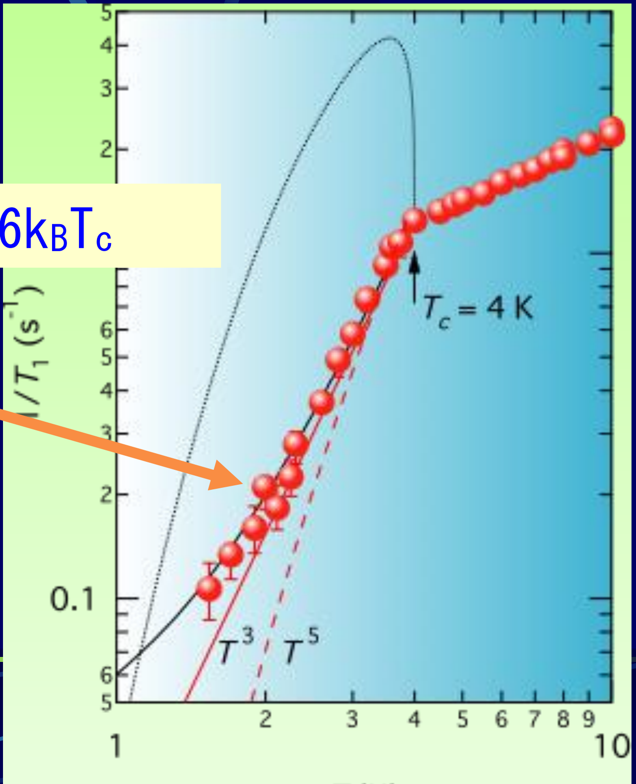
Spin susceptibility decreases below Tc＝＞Spin singlet sate

Symmetry of superconducting state in $NpPd_5Al_2$

Anisotropic gap and Spin-singlet state
=>d-wave state



SC gap $2\Delta_0 \approx 6k_BT_c$



Fermi surface with d-wave gap

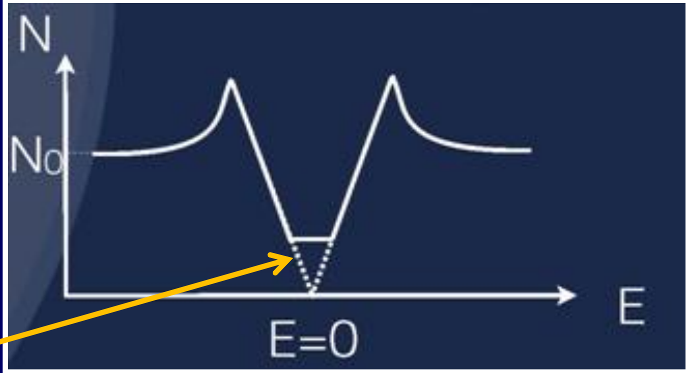
Superconducting gap and residual density of states

	$2\Delta_0/k_BT_c$	Residual DOS Nr/N(0)
$NpPd_5Al_2$	6.4	0.47
$PuRhGa_5$ a)	5	0.23
$PuCoGa_5$ b)	8	0.4
$CeCoIn_5$ b)	9	0.08

a) Sakai et al JPSJ2005 b) Yashima et al JPSJ2004

$2\Delta_0/k_BT_c>3.5$ => Strong coupling

Nr => radiation damage



Residual DOS Nr

Collaboration

NMR Group
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Michigan Univ. R.E.Walstedt

High quality Sample preparation
JAEA Y. Haga, T.D. Mastuda
Tohoku Univ. D. Aoki, Y. Homma, Y. Shiokawa
Osaka Univ. Y. Onuki

Perspectives

- Search for ^{235}U-NMR in paramagnetic state under very high field or in solution
- Investigations of AnO$_2$
  Ground states and defects