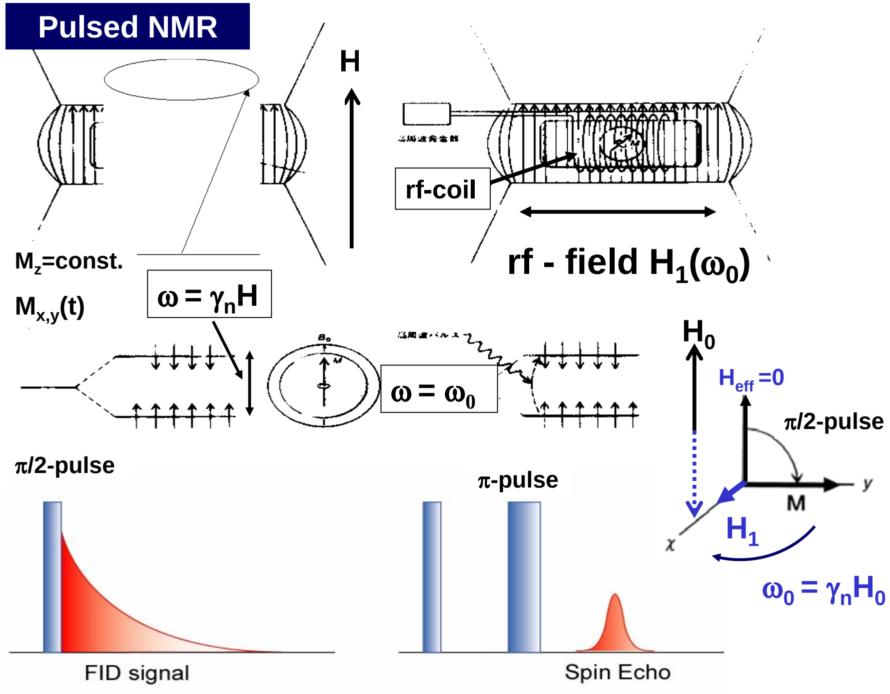
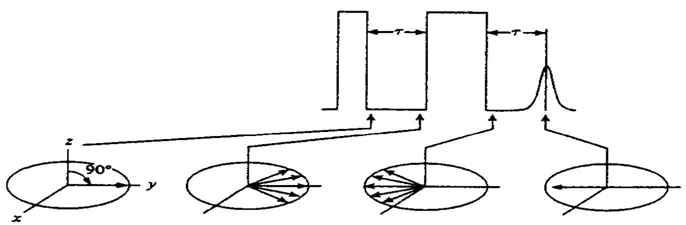
NMR/NQR probes of emergent properties in correlated-electron superconductors

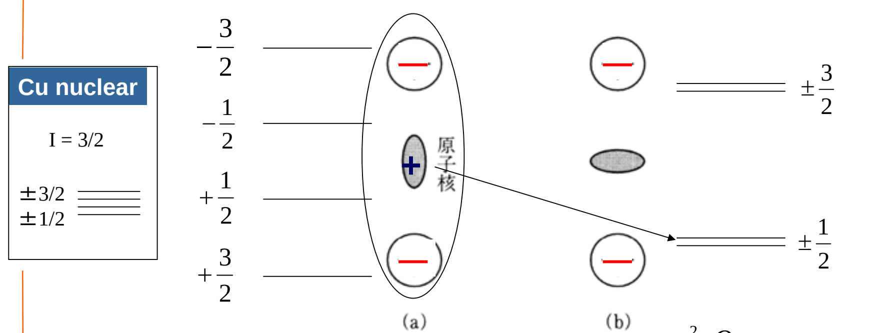
- Symmetry of the Cooper pair of either spin-singlet or spin-triplet
- SC gap with either isotropic or nodal structure
- Characters of spin fluctuations



Spin-Echo method



Nuclear Magnetic Resonance（NMR, Zero-field NMR, NQR）



$H = -\gamma\hbar I \cdot H$

Zeeman Interaction

$H_Q = \frac{e^2 qQ}{4I(2I-1)}(3I_z^2 - I^2)$

Nuclear electric quadrupole Interaction

- NMR at magnetic field
- Zero-Field NMR probing onset of magnetism

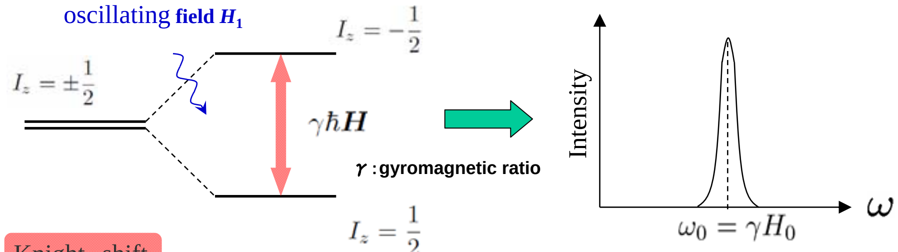
$\omega = \gamma_n H$

- NQR at zero field

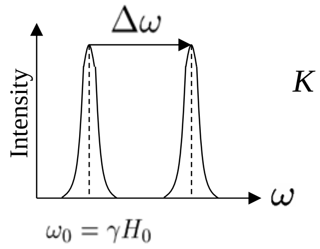
$f \equiv \nu_Q = \frac{3e^2 qQ}{2I(2I-1)h}$

NMR – Nuclear Magnetic Resonance –

Zeeman splitting

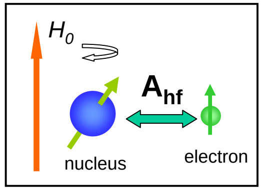


Knight shift



$$K = \frac{\Delta\omega}{\omega_0} = K_{spin} + K_{orb}$$

$$K_{spin} = A_{hf}\chi_{spin}$$



NQR and Zero-field NMR for nuclear spin I ≥ 1

Nuclear Hamiltonian of Internal Zeeman interaction and electric quadrupole interaction

$$H = -\gamma_N \hbar I \cdot H_{int} + \frac{e^2 qQ}{4I(2I-1)}(3I_Z^2 - I^2)$$

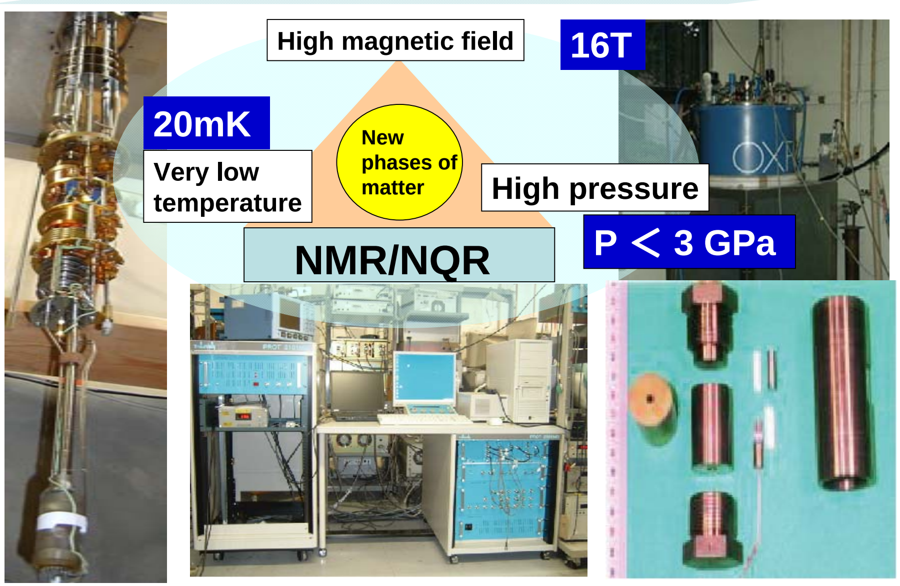
i) **In the case of non-magnetic state**

NQR at zero field → To characterize samples

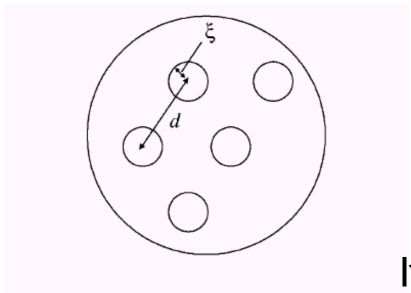
ii) **In the case of antiferromagnetically ordered state**

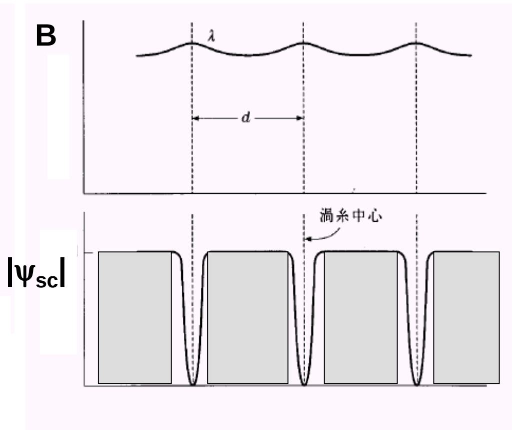
Observation of Zero-field NMR provides evidence for an onset of AFM and enables to estimate of AFM moments

NMR/NQR under multi-physical conditions



NMR in superconducting state under magnetic field



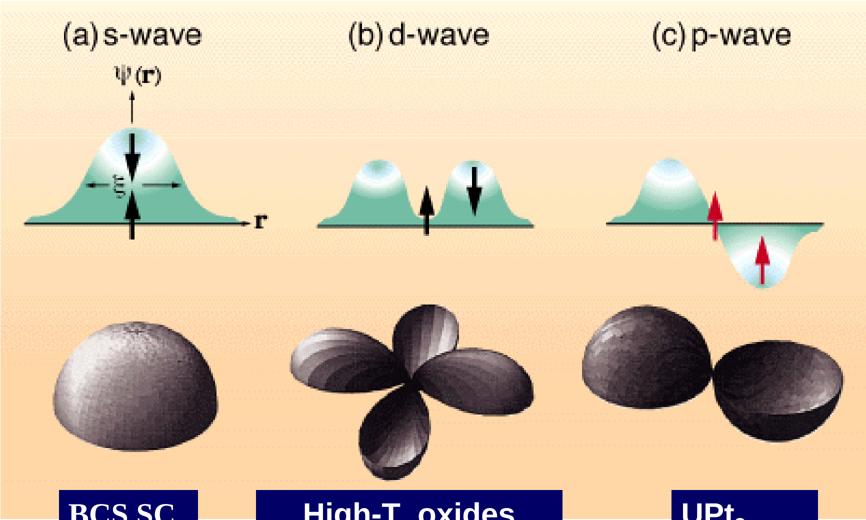


Distribution of Vortexes

$\xi << d \leq \lambda$

Knight-shift can measure the spin susceptibility below T_c, regardless of bulk-susceptibility being dominant by SC diamagnetism

Possible SC order parameters and their spin-state



BCS SC

High-T_c oxides

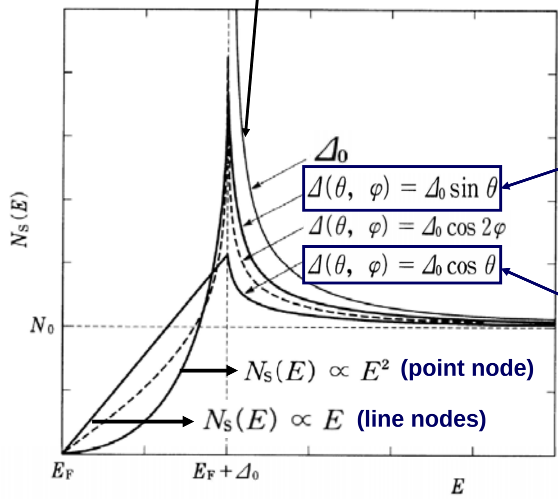
$CeCu_2Si_2$, UPd_2Al_3, $CeRIn_5$

UPt_3
Sr_2RuO_4

Quasi-particle DOS in SC state

s-wave with isotropic gap

$N_s(E)=N_0E/(E^2 - \Delta^2)^{1/2}$



p-wave with point–node gap

$$N_S(E) = \frac{N_0E}{4\pi}\int_0^{2\pi}\int_0^{\pi}\frac{\sin\theta\, d\theta\, d\varphi}{\sqrt{E^2-\Delta_0^2\sin^2\theta}} = \frac{N_0E}{2\Delta_0}\ln\left|\frac{E+\Delta_0}{E-\Delta_0}\right|$$

p-wave with line–node gap

$$N_S(E) = \frac{N_0E}{4\pi}\int_0^{2\pi}\int_0^{\pi}\frac{\sin\theta\, d\theta\, d\varphi}{\sqrt{E^2-\Delta_0^2\cos^2\theta}} = \frac{\pi}{2}\frac{N_0E}{\Delta_0} \quad (E < \Delta_0)$$

Spin susceptibility

Spin polarization in superconducting phase

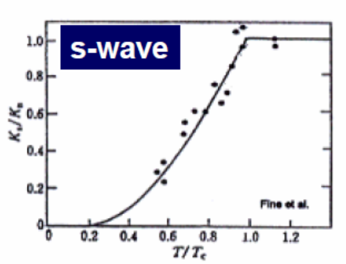
Spin singlet pairing:

- breaking up of Cooper pairs
- decrease of spin susceptibility
- vanishing susceptibility at T=0

$$\chi_s = 2\mu_B^2 N_0 Y(T),$$

where $Y(T)$ is the *Yosida function* defined by[38]

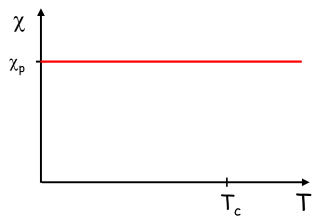
$$Y(T) = -\frac{2}{N_0}\int_0^{\infty} N_{BCS}(\varepsilon)\frac{df(\varepsilon)}{d\varepsilon}\,d\varepsilon,$$



^{27}Al Knight shift

Spin triplet pairing:

- polarization without pair breaking
- no reduction of spin susceptibility for equal-spin pairing



$\chi = \text{const.}$ for $\vec{d}(\vec{k})\cdot\vec{H} = 0$

Spin susceptibility

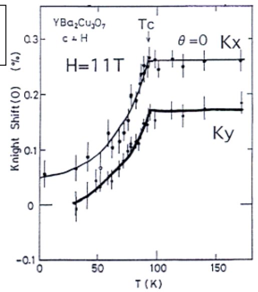
Spin polarization in superconducting phase

Spin triplet pairing:

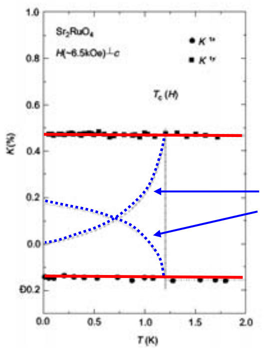
- polarization without pair breaking
- no reduction of spin susceptibility for equal-spin pairing

$\chi = \text{const.}$ for $\vec{d}(\vec{k})\cdot\vec{H} = 0$

^{17}O-Knight shift (High-T_c oxides)



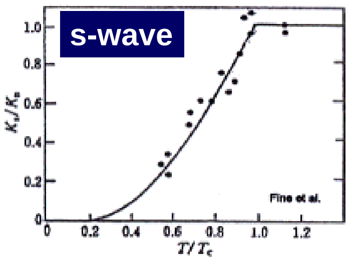
^{17}O-Knight shift In Sr_2RuO_4



Ishida et al., Nature 396, 242 (1998)

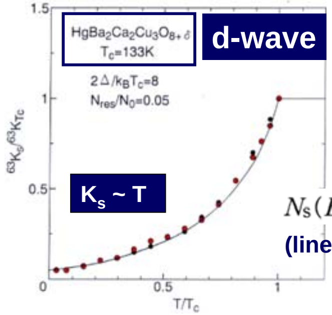
inplane equal-spin pairing $\vec{d} \parallel \hat{z}$

Summary1 : Knight shift



^{27}Al Knight shift

High-T_c SC : ^{63}Cu-NMR

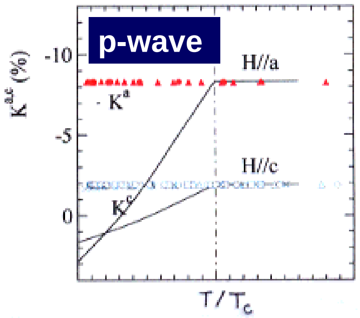


$N_s(E) \propto E$

(line nodes)

Heavy-Fermion SC

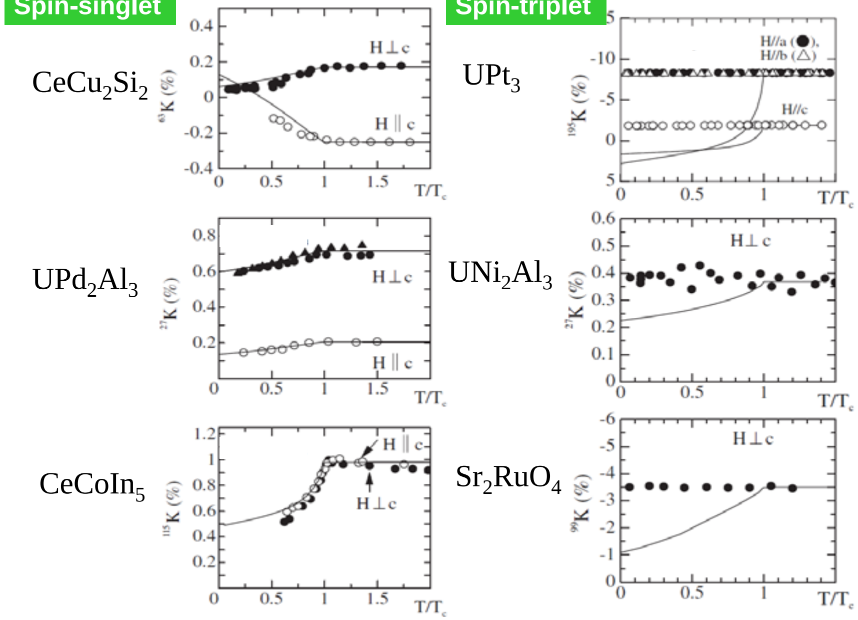
UPt$_3$: ^{195}Pt-NMR



Summary2 : pairing state in unconventional superconductors

Spin-singlet

Spin-triplet



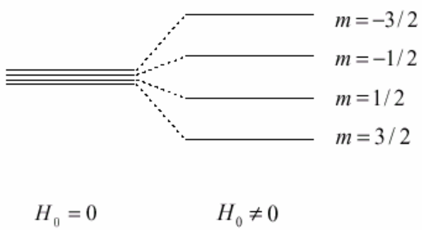
Nuclear-spin Lattice Relaxation Rate 1/T$_1$ probing SC and magnetic characteristics

Nuclear Magnetism

$$\vec{\mu} = g_N \mu_N \vec{I} = \gamma \hbar \vec{I}$$

$$\mathcal{H}_0 = -\vec{\mu}\cdot\vec{H}_0 = -\gamma\hbar\vec{I}\cdot\vec{H}_0$$

$$E_m = -\gamma\hbar H_0 m$$

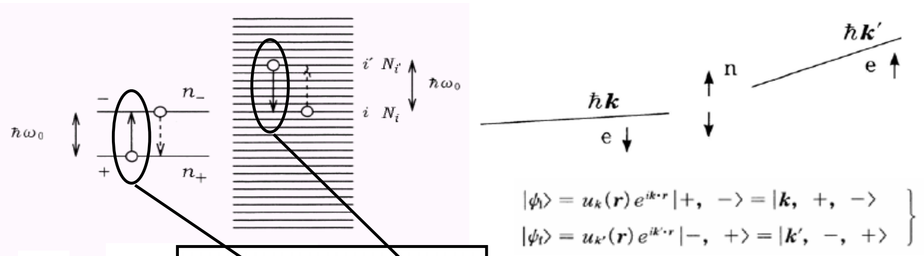


$$M(T,H) = \frac{N_0\gamma\hbar \sum_{m=-I}^{I} m \exp\left(\frac{\gamma\hbar H_0 m}{k_B T}\right)}{\sum_{m=-I}^{I} \exp\left(\frac{\gamma\hbar H_0 m}{k_B T}\right)} = N_0\gamma\hbar I B_I(Ix)$$

$$B_I(y) = \frac{2I+1}{2I}\coth\left(\frac{2I+1}{2I}y\right) - \frac{1}{2I}\coth\left(\frac{y}{2I}\right)$$

$$\chi_0(T) = \frac{M}{H} = \frac{N_0\gamma^2\hbar^2}{3k_BT} I(I+1)$$

Nuclear-spin relaxation ($1/T_1$) process



$\frac{n_-}{n_+} = 1 \rightarrow$ $\frac{n_-^0}{n_+^0} = \frac{\sum_{i'} N_{i'}}{\sum_i N_i} = e^{-\hbar\omega_0/k_BT}$

T_1 Thermal equilibrium state

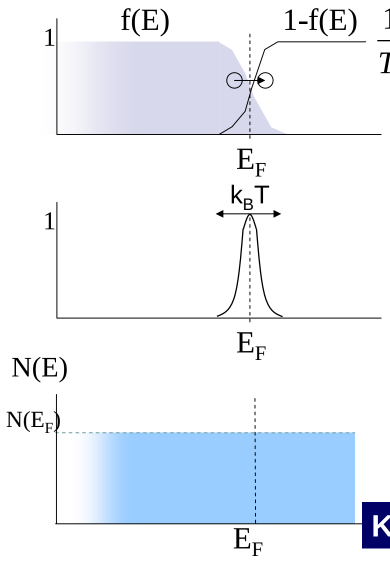
$$\mathcal{H}_{hf} = A\boldsymbol{I}\cdot\boldsymbol{S}$$

A **: Fermi-contact hyperfine interaction (s-electrons)**

$$\mathcal{H}_F = \frac{8\pi}{3}\gamma_n\gamma_e\hbar^2\delta(\boldsymbol{r})\left\{I_zS_z + \frac{1}{2}(I_+S_- + I_-S_+)\right\}$$

NMR – $1/T_1$ –

T_1 in normal state of metals



$$\frac{1}{T_1} = \frac{\pi}{\hbar}A^2\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} N(E)N(E')f(E)(1-f(E'))dEdE'$$

$$= \frac{\pi}{h}A^2N^2(E_F)k_BT$$

Pauli paramagnetism $\Rightarrow$ $\chi_S = 2\mu_B^2N(E_F)$

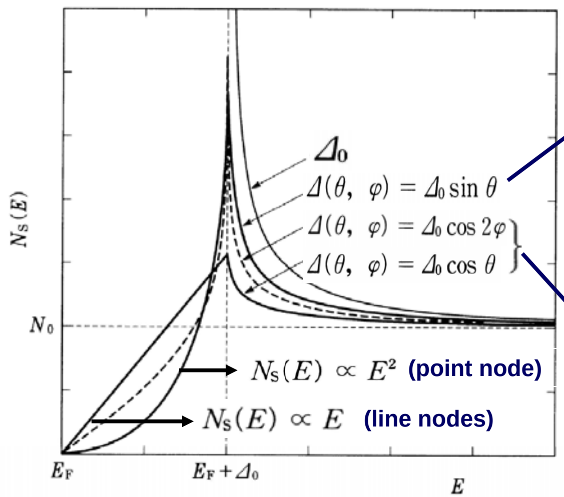
Knight Shift $\Rightarrow$ $K_S = A\chi_S \propto N(E)$

$\frac{1}{T_1} \propto K_s^2k_BT \Rightarrow \frac{1}{T_1T} \propto K^2$

Korringa Relation: $T_1TK_s^2 = \frac{\hbar}{4\pi k_B}\left(\frac{\gamma_e}{\gamma_n}\right)^2$

$1/T_1$ in superconducting state

$$\frac{1}{T_1} = \frac{\pi}{\hbar}\frac{A^2}{N^2}\int_0^{\infty}\int_0^{\infty}\left\{\left(1+\frac{\Delta^2}{EE'}\right)N_S(E)N_S(E')\right\} \times f(E)(1-f(E'))\delta(E-E')\,dE\,dE'$$



s-wave with isotropic gap

$$\left(1+\frac{\Delta^2}{E^2}\right)N_s^2(E) = N_s^2(E) + M_s^2(E)$$
$$M_s(E) = \frac{\Delta}{\sqrt{E^2-\Delta^2}}$$

p-wave with point–node gap

$$N_S(E) \propto E^2$$

SC with line–node gap

$$N_S(E) \propto E$$

BCS s-wave superconductors

$$\left(1+\frac{\Delta^2}{E^2}\right)N_s^2(E) = N_s^2(E) + M_s^2(E)$$
$$M_s(E) = \frac{\Delta}{\sqrt{E^2-\Delta^2}}$$

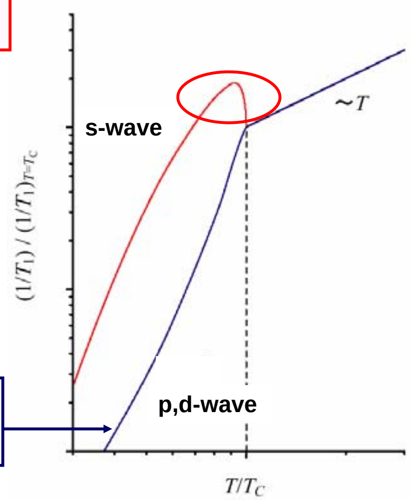
Unconventional SC in most correlated-electrons systems

$E \rightarrow 0$

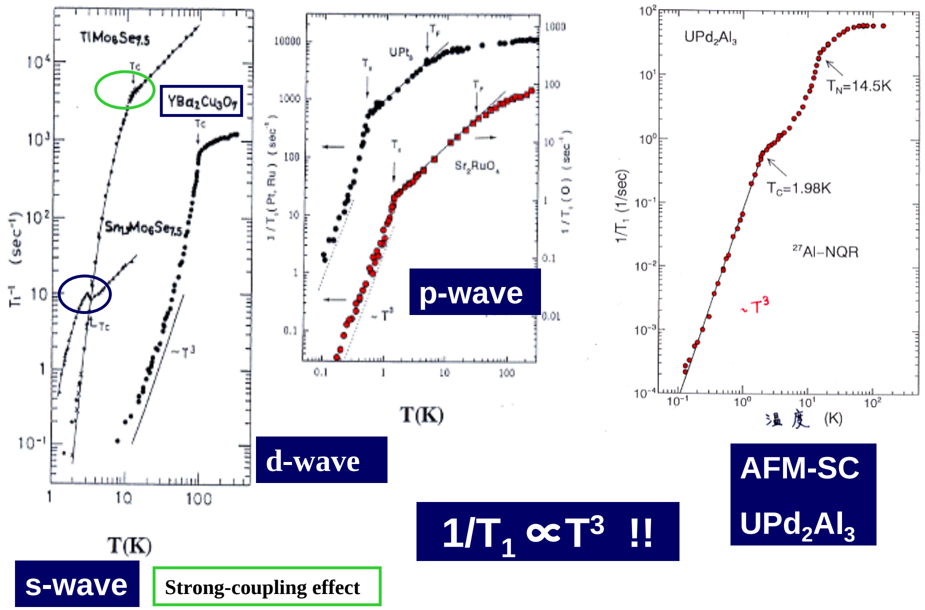
$N_S(E) \propto E$ $[\int M_s(E, \theta)\,d\theta = 0]$

(line nodes)

$$\frac{1}{T_1} \propto \int_0^{\infty} E^2e^{-E/k_BT}dE = T^3\int_0^{\infty} x^2e^{-x}dx$$

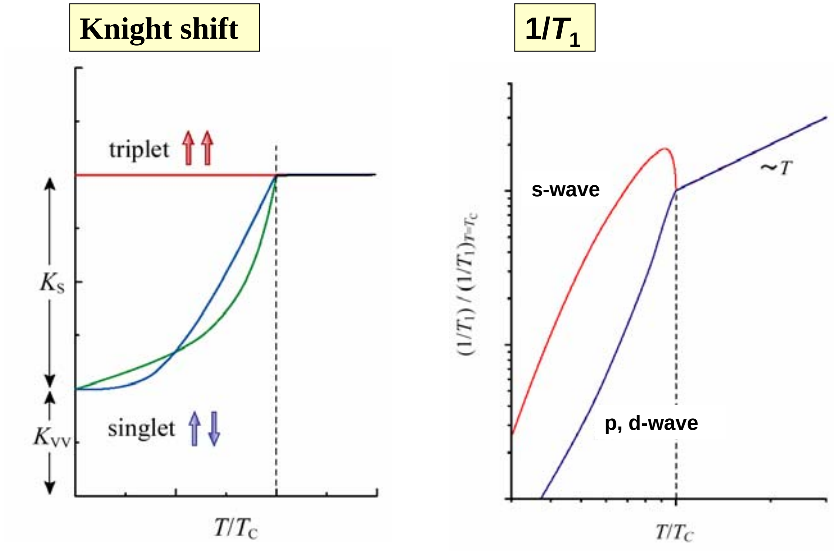


Line-nodes gap SC in correlated electrons SC



$1/T_1 \propto T^3$!!

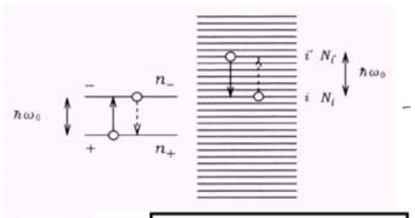
Summary: NMR probe for SC characteristics



Magnetic phases and Spin Fluctuations of Correlated Electrons

Probed by $1/T_1$ measurement

Hyperfine interactions:



$$\mathcal{H}' = -\gamma_n \hbar \boldsymbol{I} \cdot \delta \boldsymbol{H}$$

$\delta \boldsymbol{H}$: electrons spin fluctuations

$$\frac{n_-}{n_+} = 1 \rightarrow \frac{n_-^0}{n_+^0} = \frac{\sum_{i'} N_{i'}}{\sum_i N_i} = e^{-\hbar\omega_0/k_B T}$$

T_1 — Thermal equilibrium state

$1/T_1 \sim W_{a,b}$

$$W_{a,b} = \frac{2\pi}{\hbar} |\langle a|\mathcal{H}'|b\rangle|^2 \, \delta(E_a - E_b)$$

$$W_{m,\nu \to m+1,\nu'} = \frac{2\pi}{\hbar}\left(\frac{\gamma_n \hbar}{2}\right)^2 |\langle m|I_+|m+1\rangle\langle \nu|\delta H_-|\nu'\rangle|^2 \, \delta(E_{\nu'} - E_\nu - \hbar\omega_0)$$

Here,

$$\delta H_\pm = \delta H_x \pm i\, \delta H_y \qquad \delta(E_{\nu'} - E_\nu - \hbar\omega_0) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} e^{i\{(E_{\nu'} - E_\nu)/\hbar - \omega_0\}t}\, dt$$

$$W_{m,\nu\to m+1,\nu'} = \frac{\gamma_n^2}{2}\{I(I+1) - m(m+1)\}$$
$$\times \int_{-\infty}^{\infty} \langle \nu' | (e^{i\mathcal{H}_0 t/\hbar}\delta H_+ e^{-i\mathcal{H}_0 t/\hbar})\,\delta H_- | \nu'\rangle e^{-i\omega_0 t}\, dt$$

Along with $W_{m+1,\nu'\to m\nu}$

$$\frac{1}{T_1} = \sum_{\nu,\nu'} \frac{2W_{m,\nu\leftrightarrow m+1,\nu'}}{(I-m)(I+m+1)}$$
$$= \frac{\gamma_n{}^2}{2}\int_{-\infty}^{\infty} dt\, \cos\omega_0 t \left\langle \frac{\delta H_+(t)\,\delta H_-(0) + \delta H_-(t)\,\delta H_+(0)}{2} \right\rangle$$

using $\delta H_+(t) = e^{i\mathcal{H}_e t/\hbar}\delta H_+ e^{-i\mathcal{H}_e t/\hbar}$

$$\langle Q\rangle = \frac{\mathrm{Tr}\,[e^{-(\mathcal{H}_e/k_B T)}Q]}{\mathrm{Tr}\,[e^{-(\mathcal{H}_e/k_B T)}]}$$

$$A\boldsymbol{I}\cdot\boldsymbol{S} = -\gamma_n\hbar\boldsymbol{I}\cdot\delta\boldsymbol{H}$$

$$\frac{1}{T_1} = \frac{1}{2}\frac{A^2}{\hbar^2}\int_{-\infty}^{\infty}\cos\omega_0\tau\,\langle[S_+(\tau)S_-(0)]\rangle\, d\tau$$

Here, $[S_+(\tau)S_-(0)] = \dfrac{S_+(\tau)S_-(0) + S_-(\tau)S_+(0)}{2}$

Paramagnetic state in local-moment systems:

$$\frac{1}{2}\langle S_+(t)S_-(0)\rangle = \frac{1}{3}S(S+1)\exp(-\omega_e{}^2 t^2)$$

Here, $\omega_e \sim J$

$$\frac{1}{T_1} = \frac{\sqrt{2\pi}\,S(S+1)A^2}{3\hbar^2\omega_e} \sim \omega_n\frac{\omega_n}{\omega_e} \quad = \text{constant}$$

In general, for magnetic fluctuations of correlated electrons

$$\frac{1}{T_1} = \frac{\gamma_n{}^2}{2}\sum_q A_q A_{-q}\int_{-\infty}^{\infty} dt\,\cos\omega_0 t\,\langle[S_q^+(t),\ S_{-q}^-(0)]\rangle$$

Using the fluctuations-dissipation theorem :

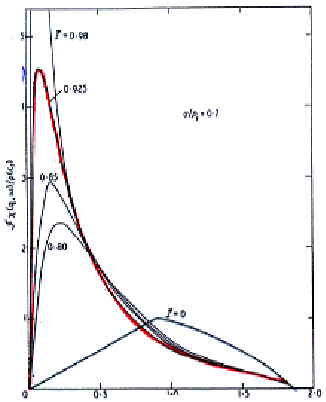
$$\frac{2\hbar\chi''_\perp(\boldsymbol{q},\ \omega_0)}{(\gamma_e\hbar)^2(1-e^{-\hbar\omega_0/k_BT})} = \frac{1}{2}\int_{-\infty}^{\infty} dt\,\cos\omega_0 t\,\langle[S_q^+(t),\ S_{-q}^-(0)]\rangle$$

$$\hbar\omega_0 \ll k_B T$$

$$\frac{1}{T_1} = \frac{2\gamma_n{}^2 k_B T}{(\gamma_e\hbar)^2}\sum_q A_q A_{-q}\frac{\chi''_\perp(\boldsymbol{q},\ \omega_0)}{\omega_0}$$

Dynamical susceptibility and NMR-$1/T_1$

$\chi''(q,\omega)$



ω

E_F

$1/T_1T \propto \Sigma_q\,\chi''(q,\omega_0)/\omega_0$

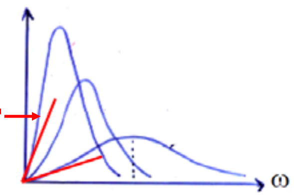
Neutron scattering

$$\frac{d^2\sigma}{d\Omega\, d\omega} \propto \frac{\chi''(q,\omega)}{1-\exp(-\omega/T)}$$

AFM-QCP at $q=Q$

$1/T_1T \sim \chi''(Q,\omega_0)/\omega_0$

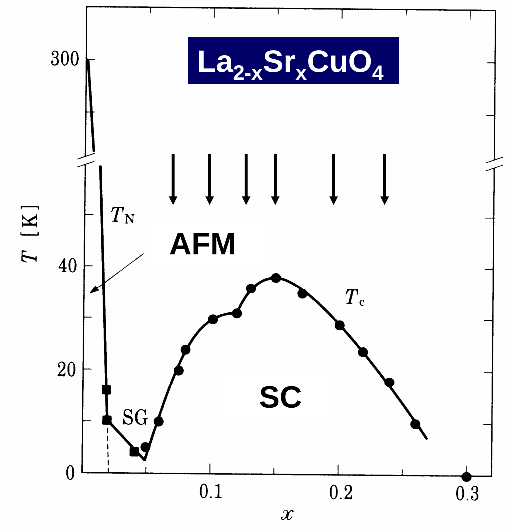
$1/T_1T$



$\chi''(q,\omega) \propto \omega\chi_0 / (\alpha - 1 + Aq^2)$

Near magnetic instability ($\alpha \to 1$ at QCP)

Spin Fluctuations of High-Temperature Superconductivity



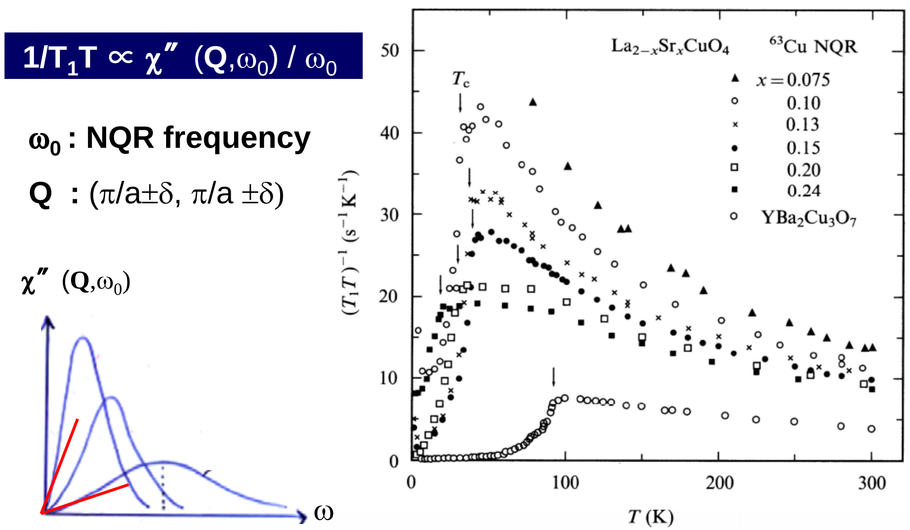
Phase diagram of AFM and SC

$1/T_1T$ of ^{63}Cu-NMR in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$

$1/T_1T \propto \chi''(Q,\omega_0)/\omega_0$

ω_0 : NQR frequency

Q : $(\pi/a\pm\delta, \pi/a \pm\delta)$



$1/T_1T$ (x, T) probes AFM spin fluctuations

Antiferromagnetic Spin Fluctuations in LSCO

2D-AFM spin fluctuations :

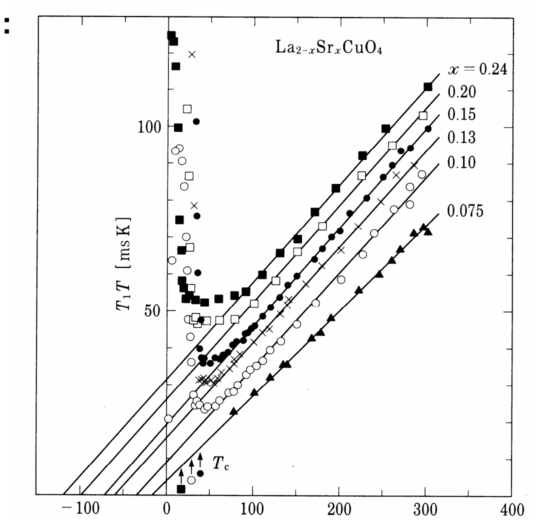
$1/T_1T \propto \chi_Q(T)$

$\propto C/(T+\theta)$

↓

$T_1T \propto 1/\chi_Q(T)$

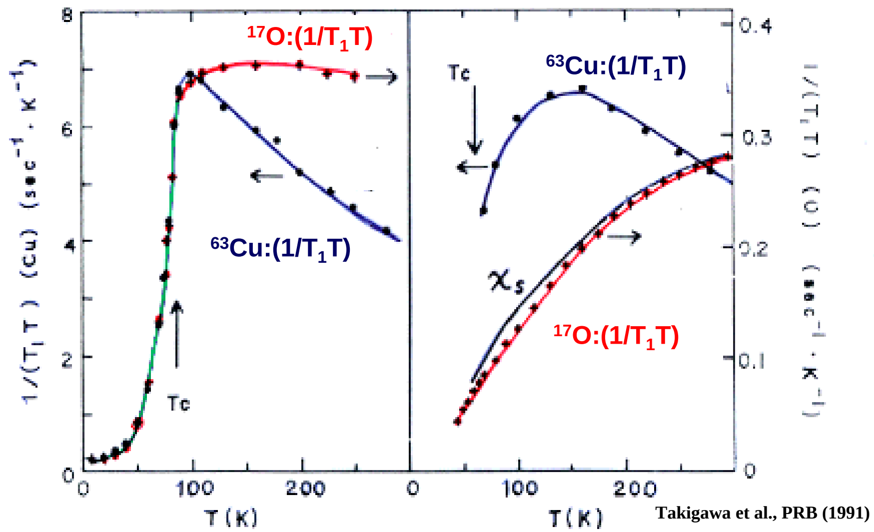
$\propto (T+\theta)$



θ → 0 (QCP) at $x \sim 0.05$

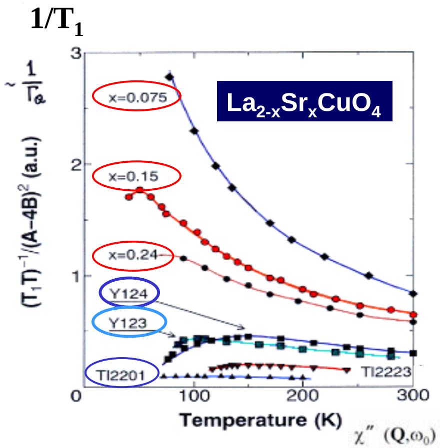
Characteristics of Antiferromagnetic Spin Fluctuations in HTSC

$1/T_1T$: $YBa_2Cu_3O_7$ (T_c=93 K) $YBa_2Cu_3O_{6.63}$ (T_c=60 K)

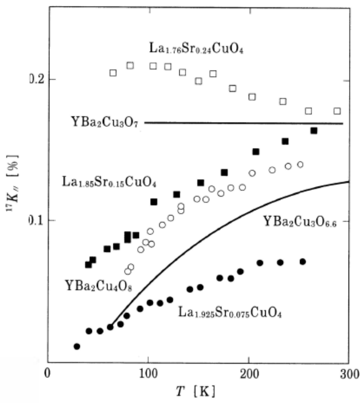


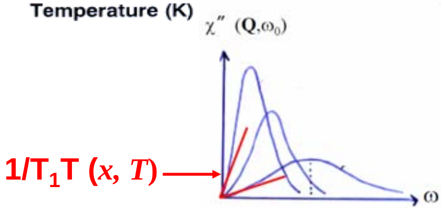
Takigawa et al., PRB (1991)

Summary: Carrier-density Dependence of Spin Fluctuations



uniform spin χ(q=0)





Spin Fluctuations in $YBa_2Cu_3O_x$ probed by neutron