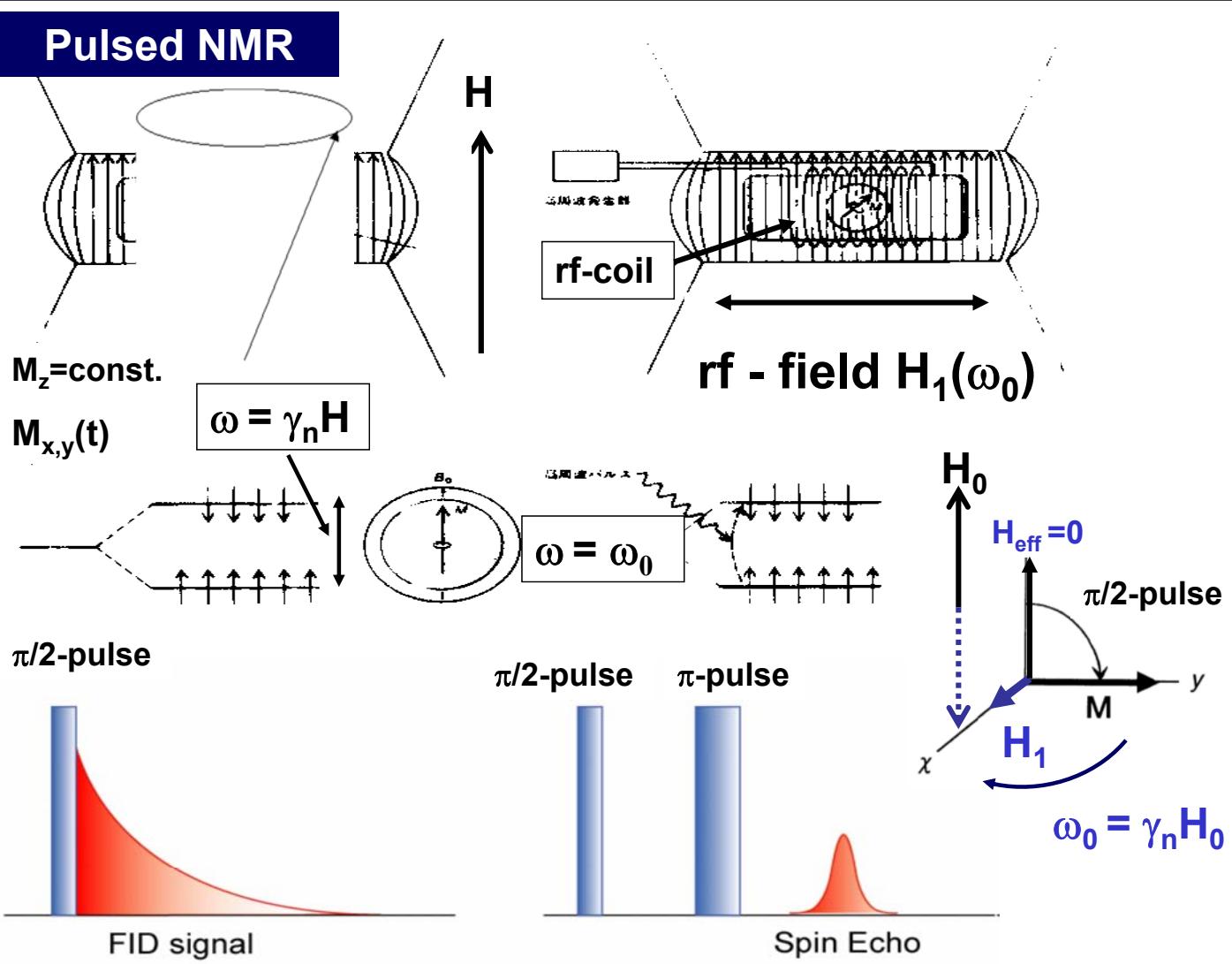
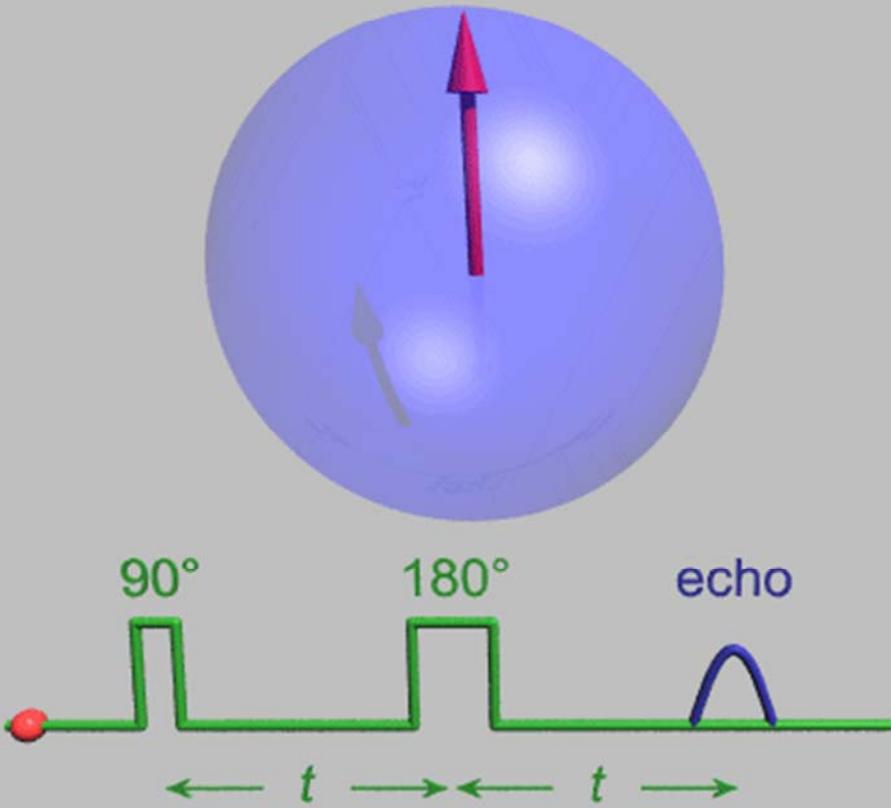


核磁気共鳴法(NMR)および核電気四極子共鳴法(NQR) の基礎 および 強相関電子系への応用

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

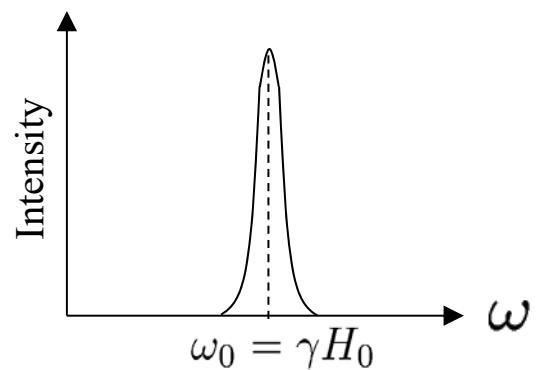
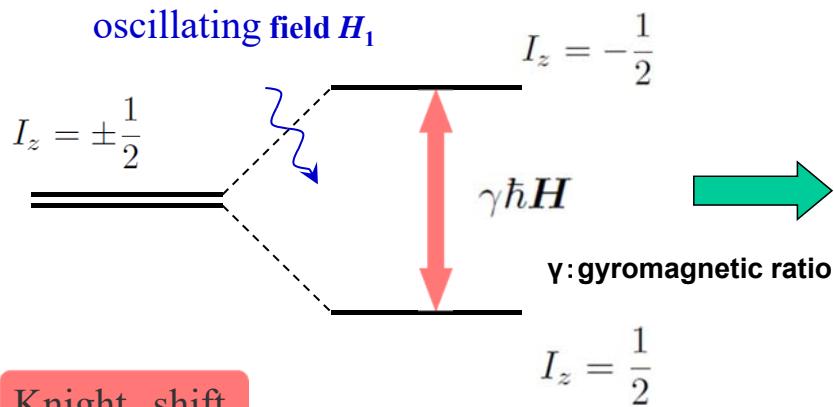
- Symmetry of the Cooper pair of either spin-singlet or spin-triplet
- SC gap with either isotropic or nodal structure
- Characters of spin fluctuations



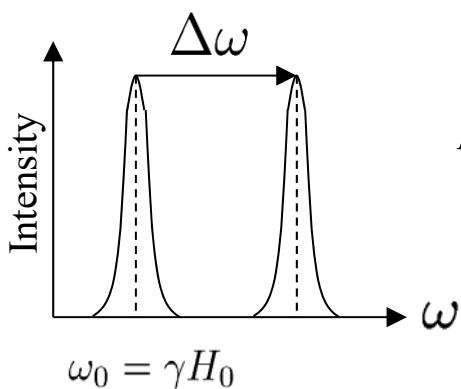


NMR – Nuclear Magnetic Resonance –

Zeeman splitting



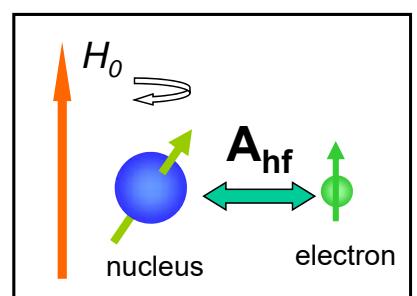
Knight shift



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

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

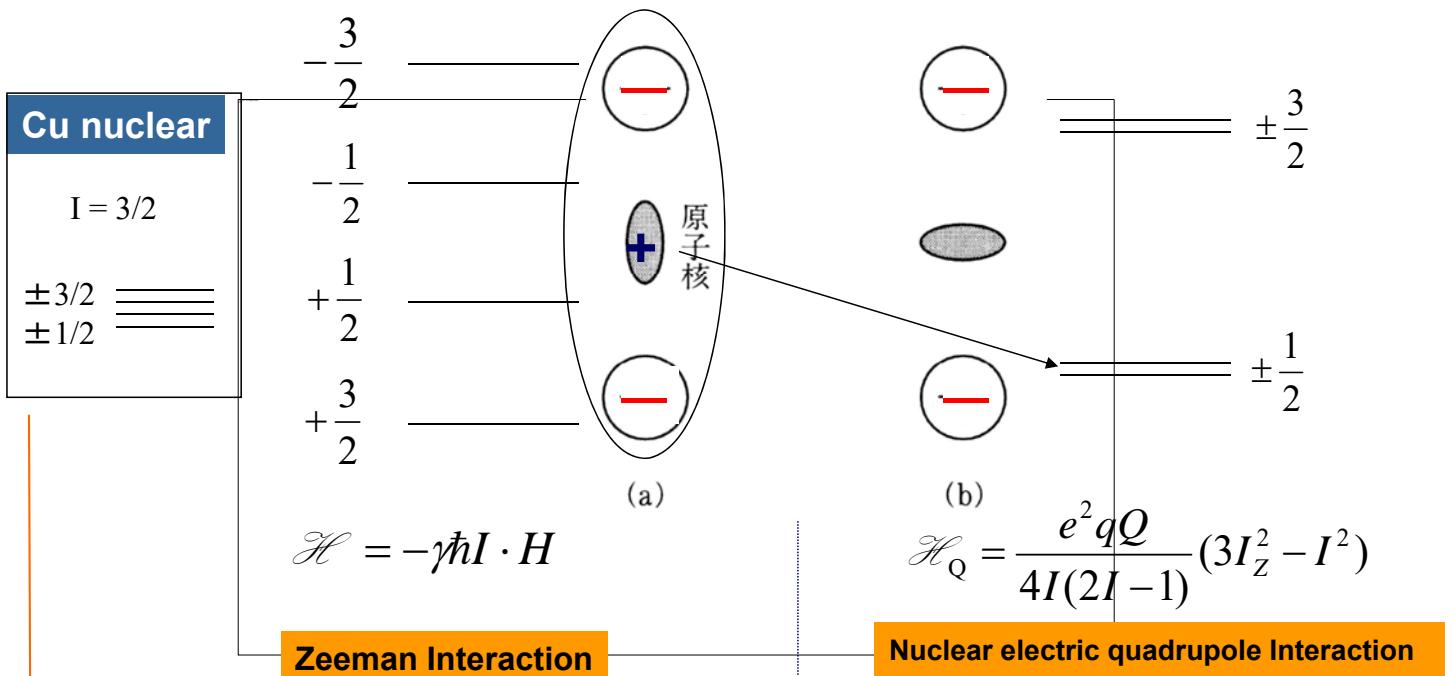
$$K_{spin} = A_{hf} \chi_{spin}$$



核電気四極子共鳴の原理

Principle of Nuclear Electric Quadrupole Resonance

Nuclear Magnetic Resonance (NMR) and Nuclear Quadrupole Resonance (NQR)



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

$$\omega = \gamma_n H$$

- NQR at zero field

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

Nuclear Hamiltonian of Internal Zeeman interaction and electric quadrupole interaction

$$\mathcal{H} = -\gamma_N \hbar I \cdot H_{\text{int}} + \frac{e^2 q Q}{4I(2I-1)} (3I_z^2 - I^2)$$

i) In the case of non-magnetic state

NQR at zero field → To characterize samples

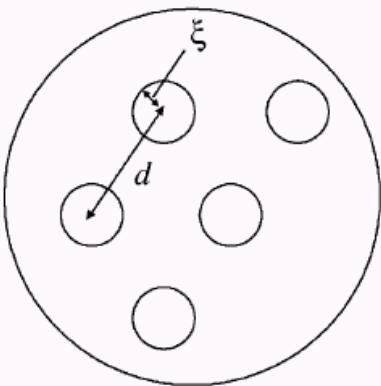
ii) In the case of antiferromagnetically ordered state

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

超伝導状態のNMRによる研究から分かること

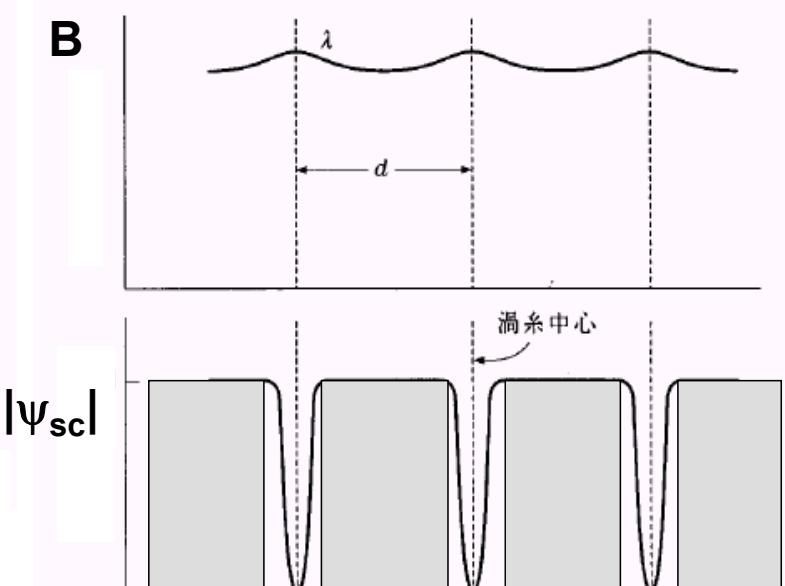
- ・ナイトシフト
- ・ T_1 測定

NMR in superconducting state under magnetic field



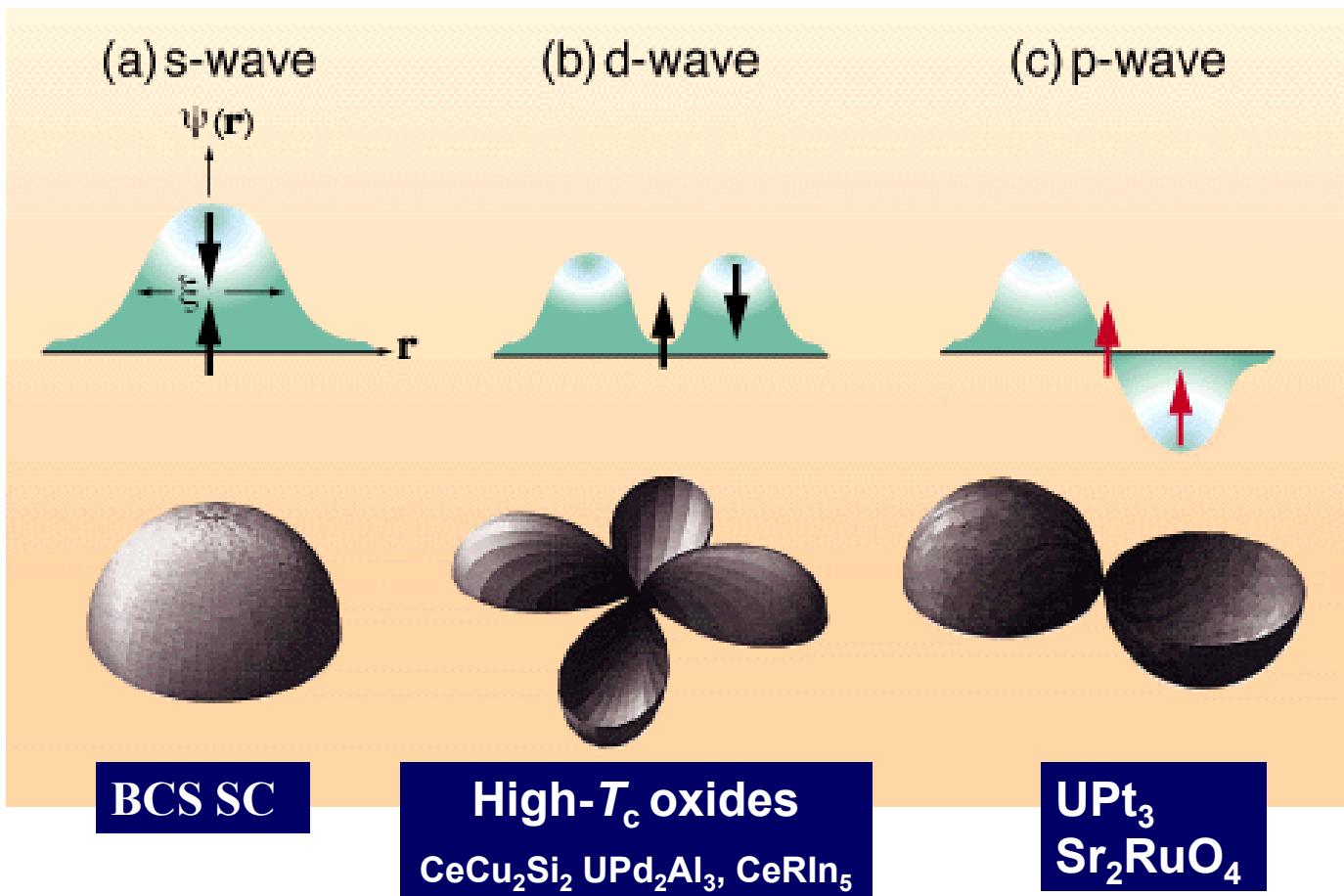
Distribution of Vortexes

$$\xi \ll d \leq \lambda$$



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

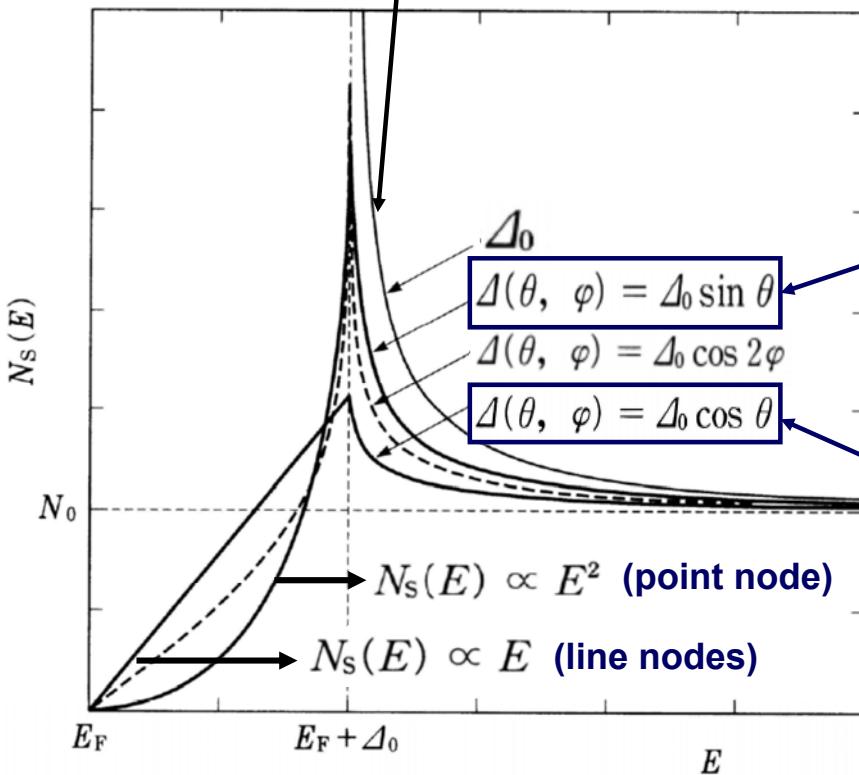
Possible SC order parameters and their spin-state



Quasi-particle DOS in SC state

s-wave with isotropic gap

$$N_s(E) = N_0 E / (E^2 - \Delta^2)^{1/2}$$



p-wave with point-node gap

$$\begin{aligned} N_s(E) &= \frac{N_0 E}{4\pi} \int_0^{2\pi} \int_0^\pi \frac{\sin \theta \, d\theta \, d\varphi}{\sqrt{E^2 - \Delta_0^2 \sin^2 \theta}} \\ &= \frac{N_0 E}{2\Delta_0} \ln \left| \frac{E + \Delta_0}{E - \Delta_0} \right| \end{aligned}$$

p-wave with line-node gap

$$\begin{aligned} N_s(E) &= \frac{N_0 E}{4\pi} \int_0^{2\pi} \int_0^\pi \frac{\sin \theta \, d\theta \, d\varphi}{\sqrt{E^2 - \Delta_0^2 \cos^2 \theta}} \\ &= \frac{\pi}{2} \frac{N_0 E}{\Delta_0} \quad (E < \Delta_0) \end{aligned}$$

Spin susceptibility

Spin polarization in superconducting phase

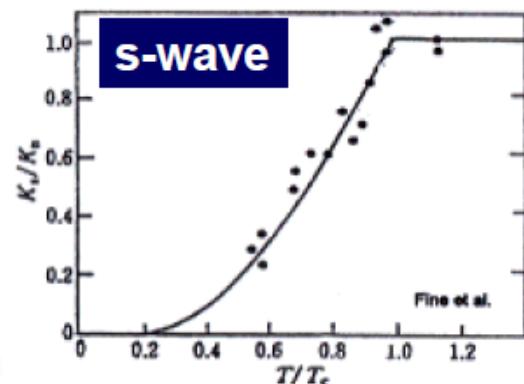
Spin singlet pairing:

- 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 by³⁸⁾

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

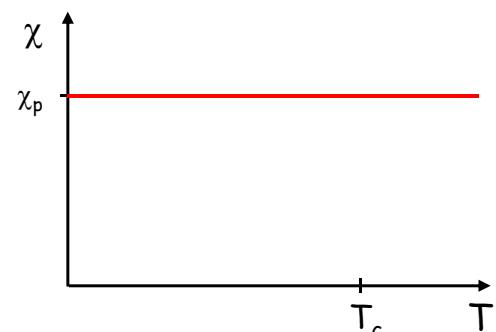


²⁷Al Knight shift

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

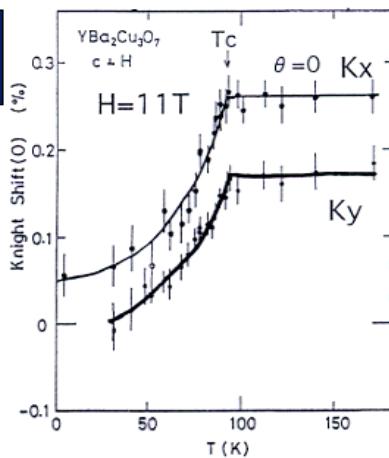
Spin polarization in superconducting phase

Spin triplet pairing:

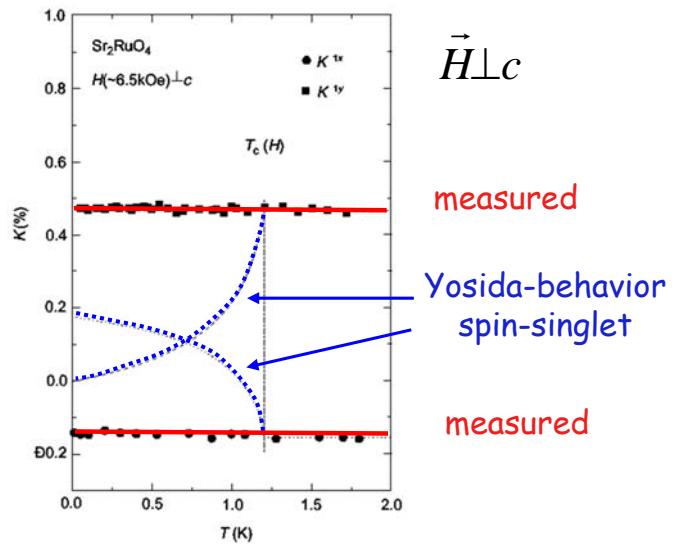
- polarization without pair breaking
- no reduction of spin susceptibility for equal-spin pairing

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

¹⁷O-Knight shift
(High- T_c oxides)



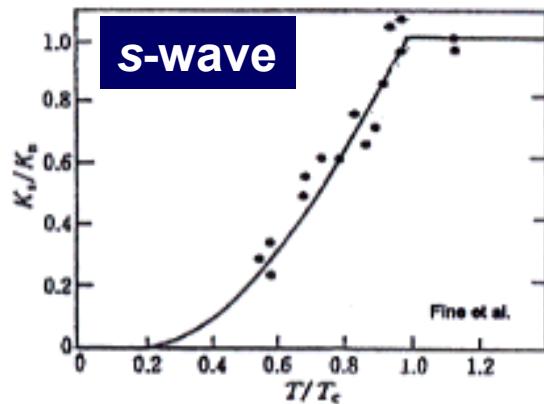
¹⁷O-Knight shift In Sr_2RuO_4



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

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

Summary1 : Knight shift

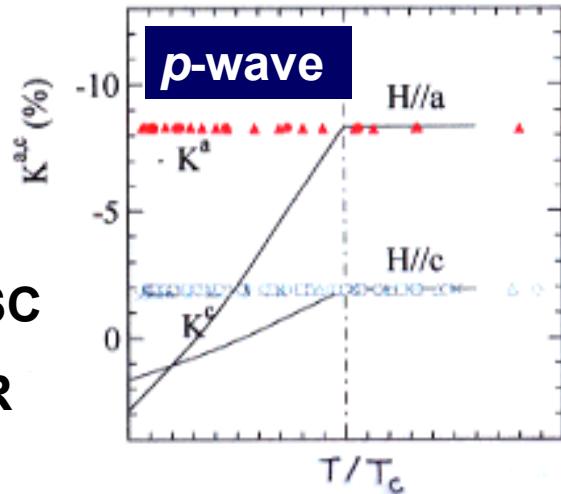
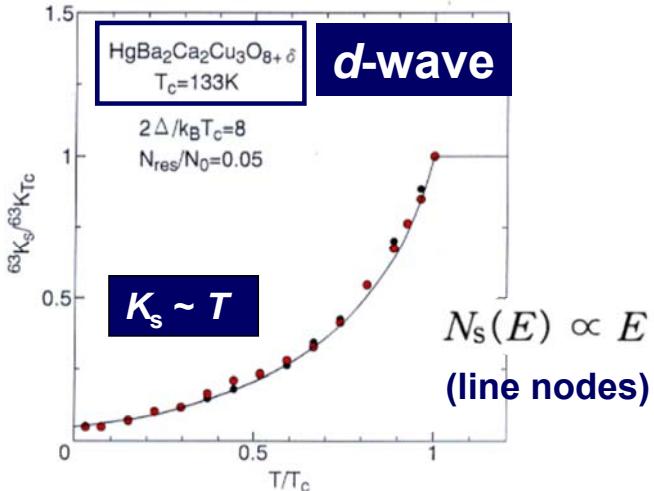


²⁷Al Knight shift

Heavy-electrons SC

UPt₃ : ¹⁹⁵Pt-NMR

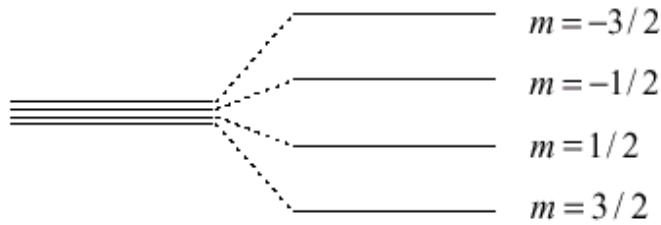
High- T_c SC : ⁶³Cu-NMR



Nuclear Magnetism

$$\begin{aligned}\vec{\mu} &= g_N \mu_N \vec{I} \\ &= \gamma \hbar \vec{I}\end{aligned}$$

$$\begin{aligned}\mathcal{H}_0 &= -\vec{\mu} \cdot \vec{H}_0 \\ &= -\gamma \hbar \vec{I} \cdot \vec{H}_0 \\ E_m &= -\gamma \hbar H_0 m\end{aligned}$$



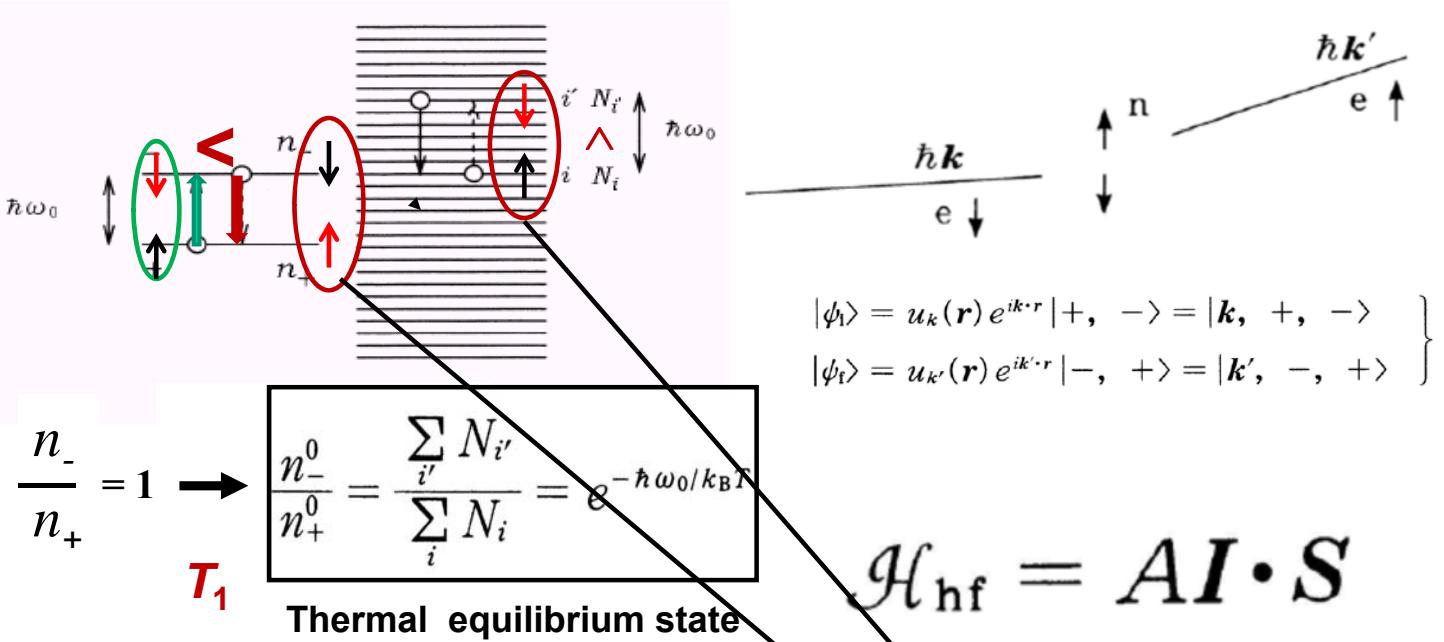
$$\begin{aligned}M(T, H) &= \frac{N_0 \gamma \hbar \sum_{m=-I}^I m \exp\left(\frac{\gamma \hbar H_0 m}{k_B T}\right)}{\sum_{m=-I}^I \exp\left(\frac{\gamma \hbar H_0 m}{k_B T}\right)} \\ &= N_0 \gamma \hbar I B_I(Ix)\end{aligned}$$

$$H_0 = 0 \quad H_0 \neq 0$$

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

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

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

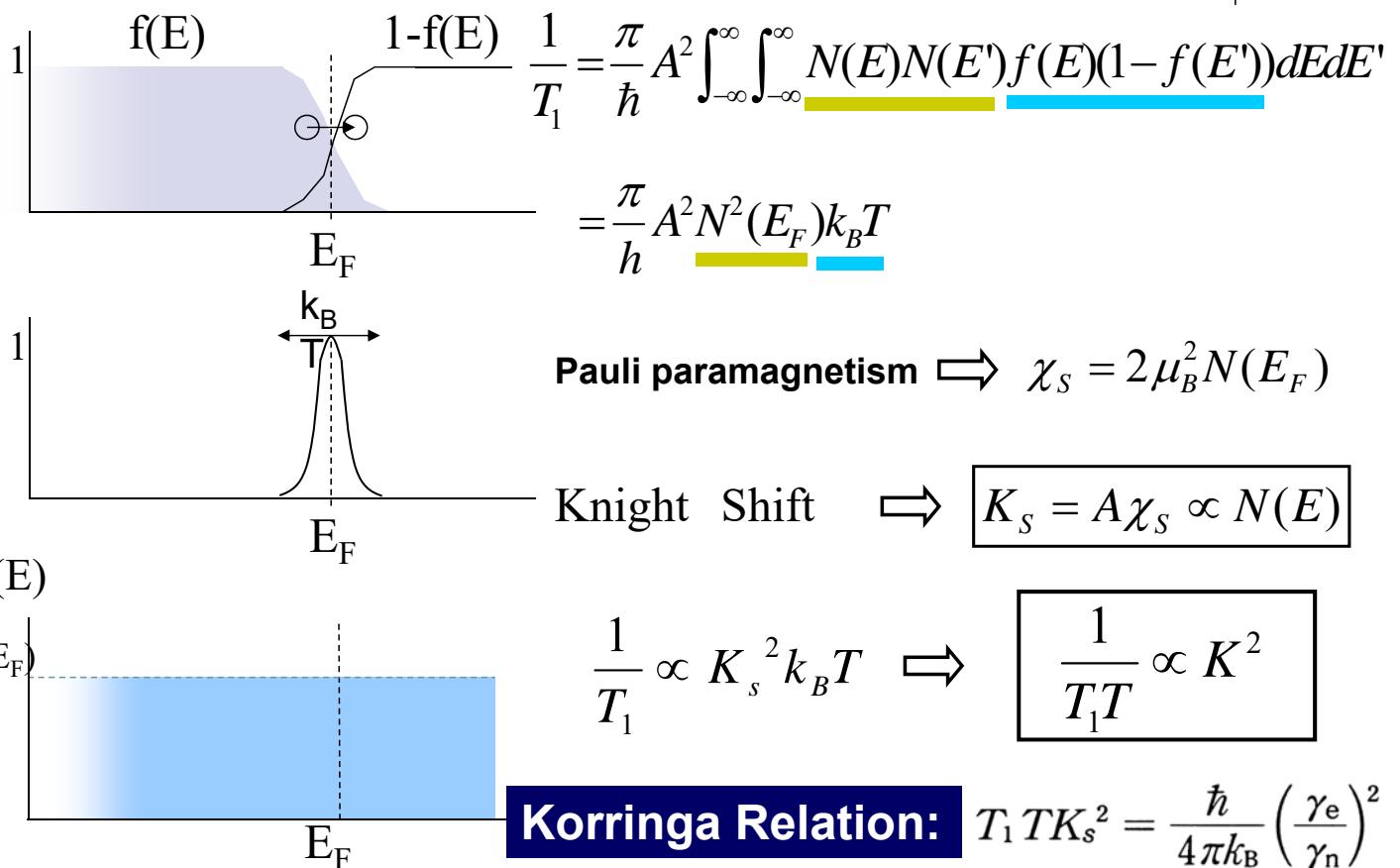


A : Fermi-contact hyperfine interaction (s-electrons)

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



T_1 in normal state of metals

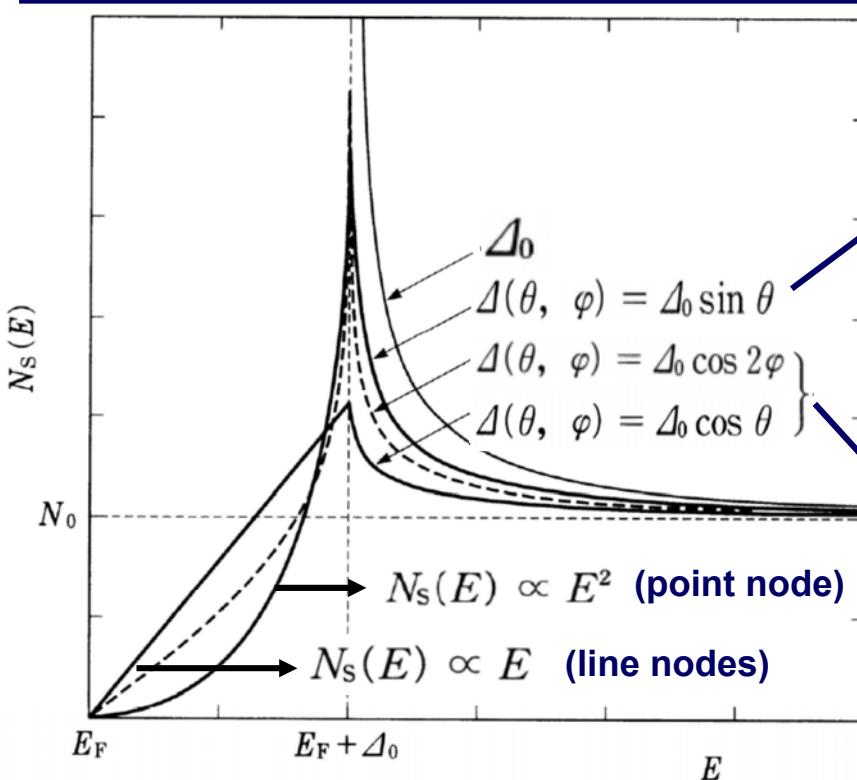


1/ T_1 in superconducting state

s-wave with isotropic gap

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

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



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:

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

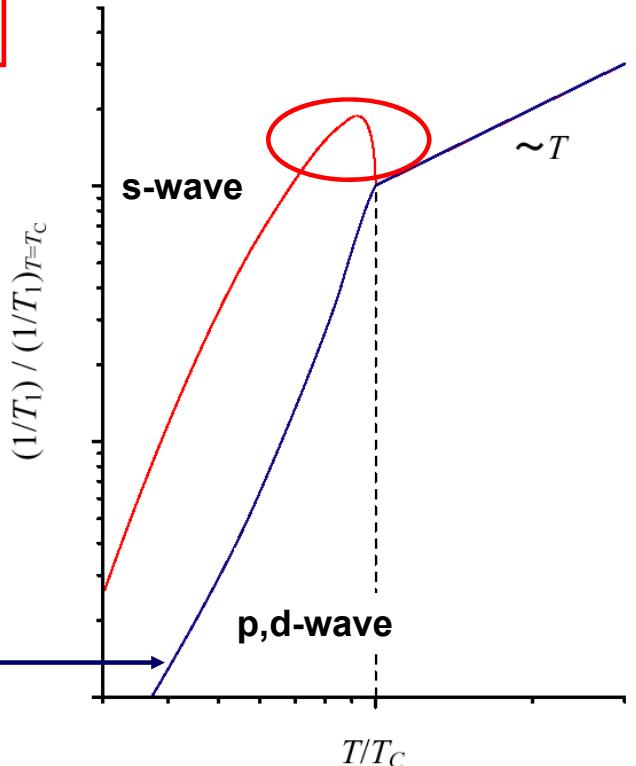
$$M_s(E) = \frac{\Delta}{\sqrt{E^2 - \Delta^2}}$$

Unconventional SC in most correlated-electrons systems

$E \rightarrow 0$

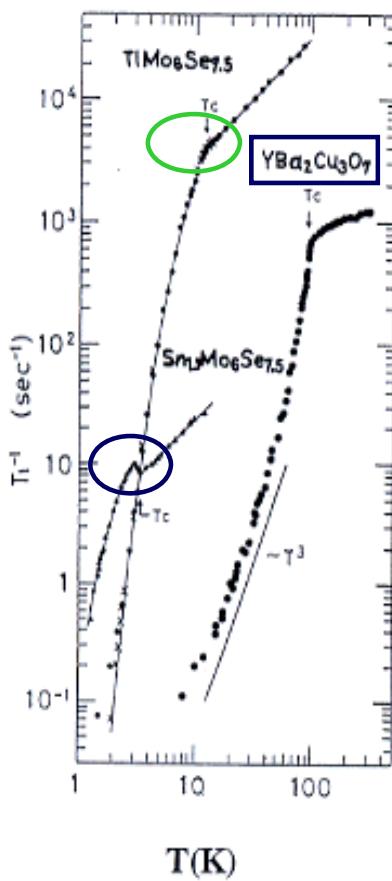
$$N_s(E) \propto E \quad [\int M_s(E, \theta) d\theta = 0]$$

(line nodes)



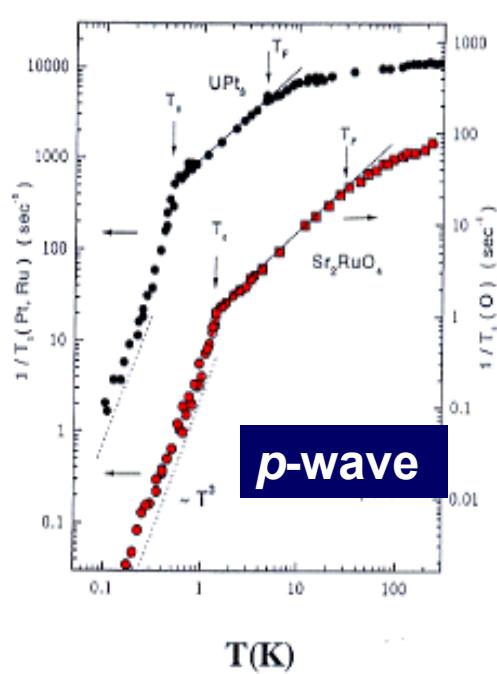
$$\frac{1}{T_1} \propto \int_0^\infty E^2 e^{-E/k_B T} dE = T^3 \int_0^\infty x^2 e^{-x} dx$$

Line-nodes gap SC in correlated electrons SC

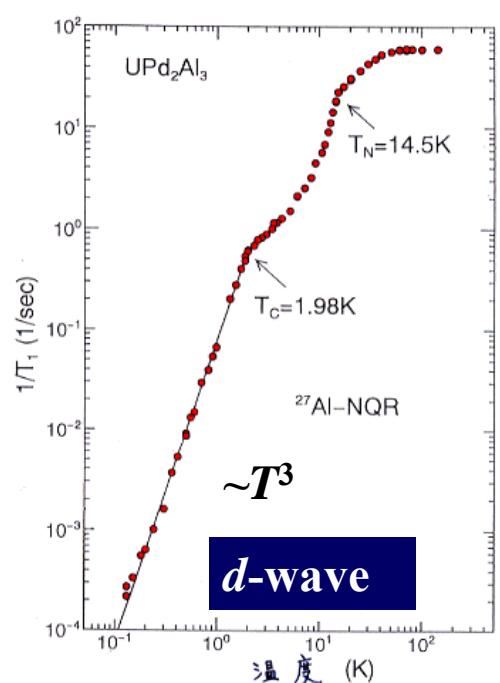


s-wave

Strong-coupling effect



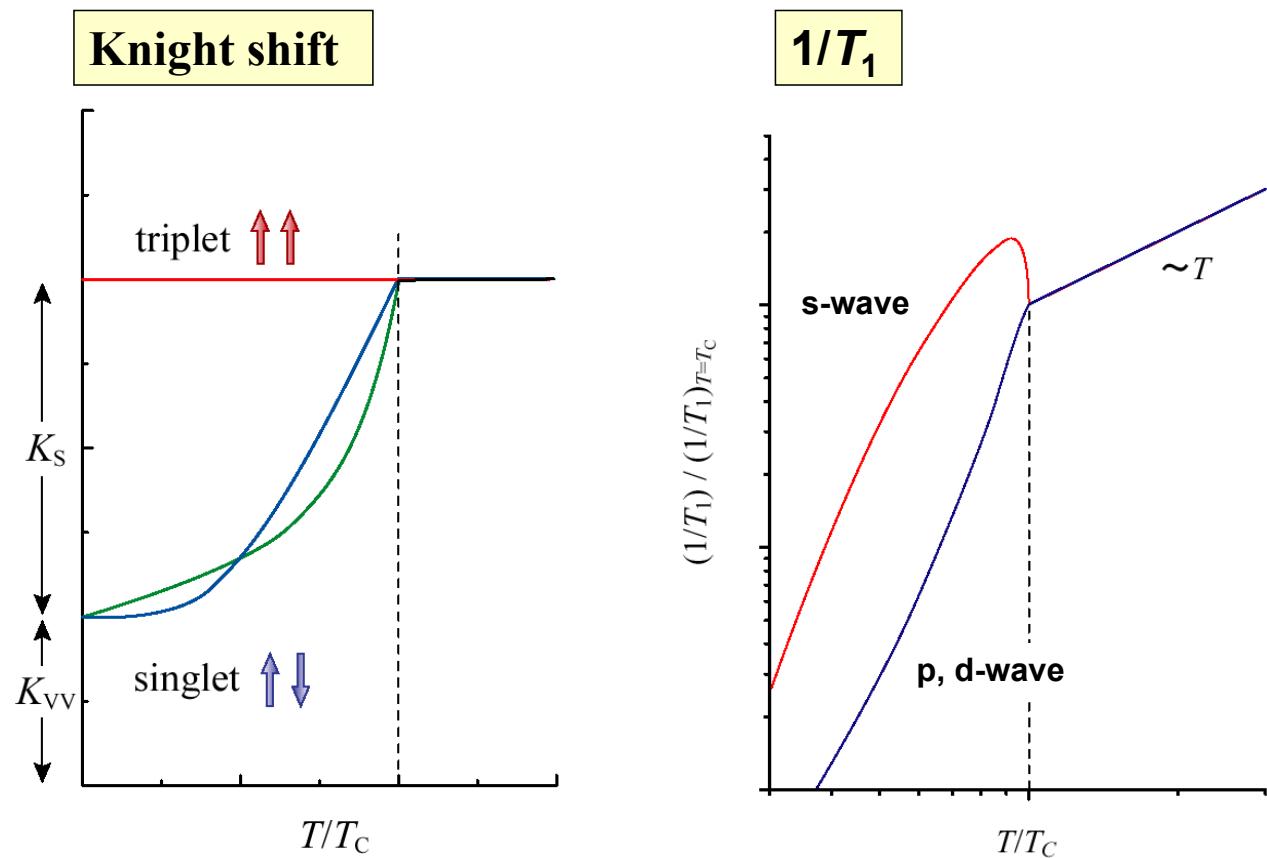
d-wave



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

AFM-SC
UPd₂Al₃

Summary: NMR probe for SC characteristics



NMRからみた「スピンのゆらぎ」と
「d波超伝導」

NMR Probe for 「Spin Fluctuations」 and
「d-wave superconductivity」

The formula of $1/T_1$

$-1/2$ ————— $N-$

$I_z = 1/2$ ————— N_+

Time-dependent
perturbation,
 $H' = -\gamma_N \hbar \vec{I} \cdot \vec{H}_{\text{loc}}(t)$
allows us to calculate

Relaxation rate $1/T_1$ due to fluctuations
of local magnetic field \mathbf{H}_{loc}

$$\frac{-\gamma_N \hbar}{2} (I^+ H_{\text{loc}}^- (t) + I^- H_{\text{loc}}^+ (t))$$

$$I^\pm = I_x \pm i I_y, \quad H_{\text{loc}}^\pm = H_{\text{loc}}^x \pm i H_{\text{loc}}^y$$

$$\begin{aligned} \frac{1}{T_1} &= \frac{2\pi}{\hbar} \left(\frac{\gamma_N \hbar}{2} \right)^2 \sum_{n,m} \exp(-\beta \varepsilon_n) \left(\langle m | H_{\text{loc}}^+ | n \rangle \right)^2 \delta(\varepsilon_m - \varepsilon_n + \hbar \omega_N) + \left(\langle m | H_{\text{loc}}^- | n \rangle \right)^2 \delta(\varepsilon_m - \varepsilon_n - \hbar \omega_N) \\ &= \frac{\gamma_N^2}{4} \sum_{n,m} \exp(-\beta \varepsilon_n) \int_{-\infty}^{\infty} \left[\langle m | H_{\text{loc}}^+ | n \rangle \right]^2 \exp\left(\frac{i(\varepsilon_m - \varepsilon_n)t}{\hbar}\right) + \left[\langle m | H_{\text{loc}}^- | n \rangle \right]^2 \exp\left(\frac{i(\varepsilon_n - \varepsilon_m)t}{\hbar}\right) \right] \exp(i\omega_N t) dt \\ &= \frac{\gamma_N^2}{4} \sum_{n,m} \exp(-\beta \varepsilon_n) \int_{-\infty}^{\infty} \left(\langle n | H_{\text{loc}}^- | m \rangle \langle m | e^{\frac{iHt}{\hbar}} H_{\text{loc}}^+ e^{-\frac{iHt}{\hbar}} | n \rangle + \langle n | e^{\frac{iHt}{\hbar}} H_{\text{loc}}^+ e^{-\frac{iHt}{\hbar}} | m \rangle \langle m | H_{\text{loc}}^- | n \rangle \right) \exp(i\omega_N t) dt \\ &= \frac{\gamma_N^2}{2} \int_{-\infty}^{\infty} \langle [H_{\text{loc}}^-, H_{\text{loc}}^+](t) \rangle \exp(i\omega_N t) dt \quad \left(\{A, B\} = \frac{1}{2}(AB + BA), \quad H_{\text{loc}}^+(t) = e^{\frac{iHt}{\hbar}} H_{\text{loc}}^+ e^{-\frac{iHt}{\hbar}} \right) \end{aligned}$$

Transition probability \longleftrightarrow Correlation function
(General principle , Neutron scattering)

In general, for magnetic fluctuations of correlated electrons

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

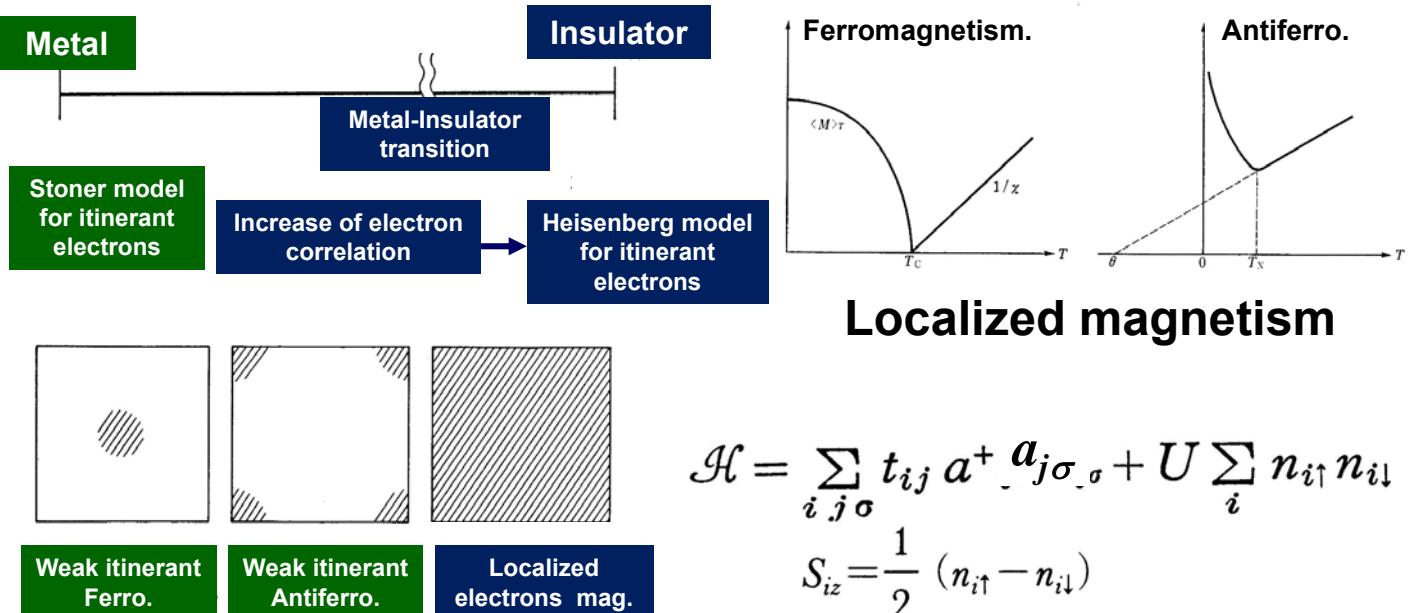
Using the fluctuations-dissipation theorem :

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

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

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

Itinerant Magnetism & Spin-fluctuations



Wave-number dependent susceptibility follows a Currie Weiss law in a different origin from the localized model

Self-consistent renormalization (SCR) theory:

$$\langle S_{iz}(t)S_{iz}(t') \rangle \sim \langle S_{iz}(t) \rangle \langle S_{iz}(t') \rangle$$

$$\chi(q, \omega) = \frac{\chi_0(q, \omega)}{1 - I\chi_0(q, \omega) + \lambda(q, \omega)}$$

$$\mathcal{H} = \sum_{i,j,\sigma} t_{ij} a^+ a_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

$$S_{iz} = \frac{1}{2} (n_{i\uparrow} - n_{i\downarrow})$$

This relationship replaces the above second term in Hamiltonian to the follows;

$$-2I \sum_i S_{iz}^2 + \text{const}$$

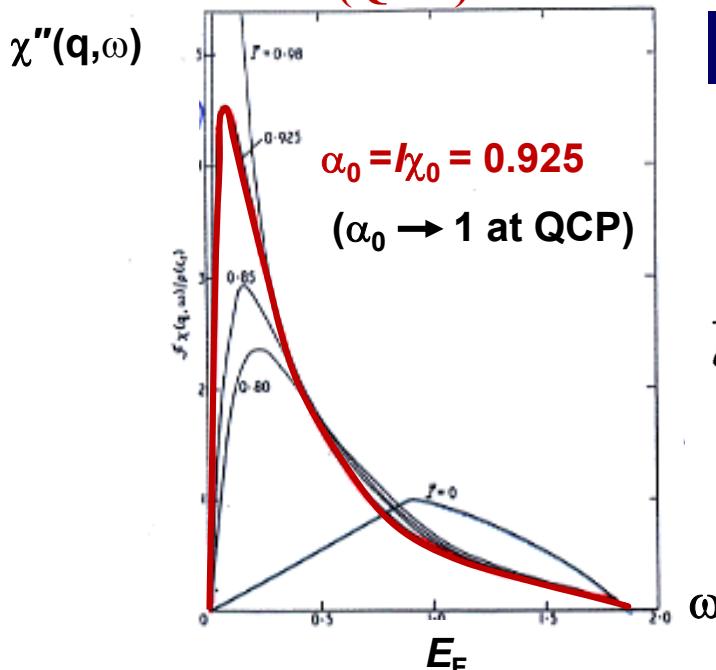
$$\chi = \frac{\chi_0}{(1 - \alpha_0)}$$

taking $\langle S_{iz} \rangle = \text{const.}$

$$\alpha_0 = I\chi_0$$

Dynamical susceptibility and NMR- $1/T_1$

Near ferromagnetic critical point (QCP)



$$1/T_1 T \propto \sum_q \chi''(q, \omega_0) / \omega_0$$

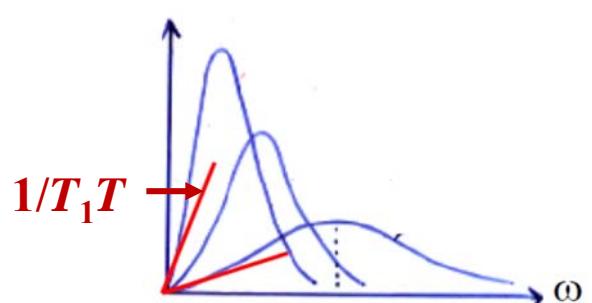
Neutron scattering

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

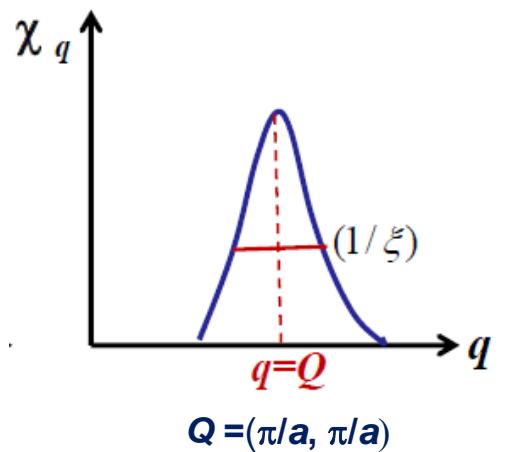
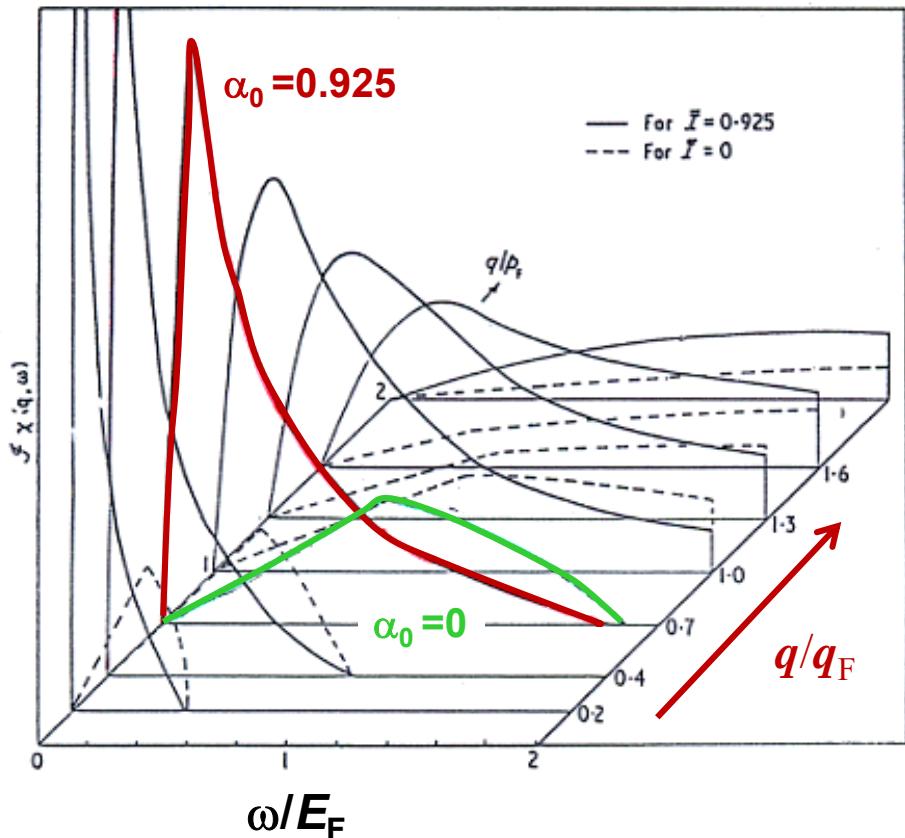
$$\chi''(q, \omega) / \omega \propto \chi_0 / (\alpha_0 - 1 + Aq^2)$$

$$1/T_1 T \sim \chi''(Q, \omega_0) / \omega_0$$

AFM-QCP near $q=Q$



Near ferromagnetic critical point

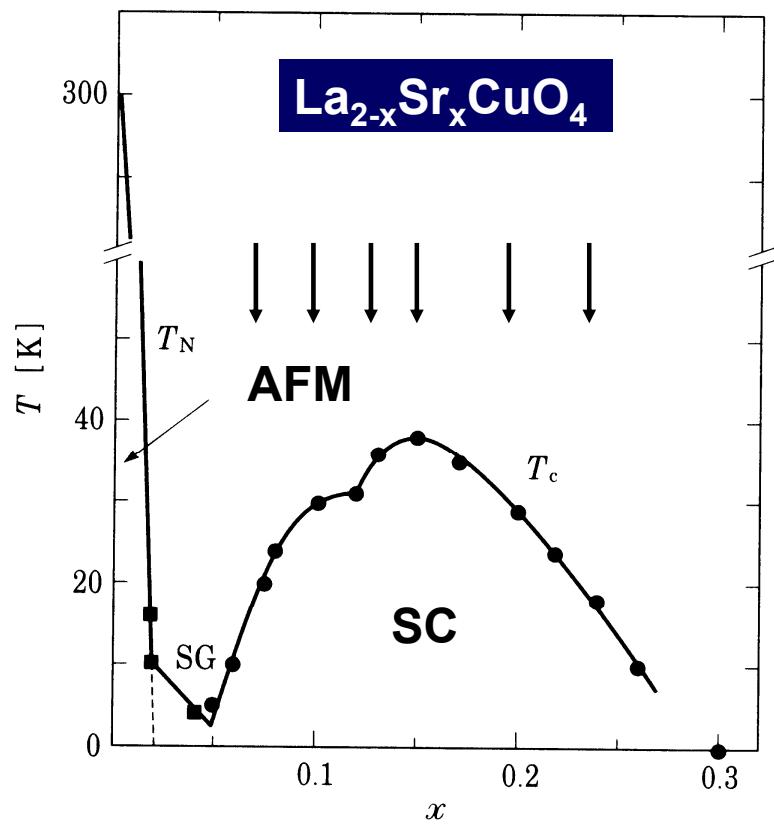


Near AFM critical point

$$\alpha_0 = I\chi_q \sim 1$$

$$1/T_1 T \sim \chi''(Q, \omega_0)/\omega_0 \\ \sim \chi_Q(T)$$

Spin Fluctuations of High-Temperature Superconductivity



Phase diagram of AFM and SC

$1/T_1 T$ of ^{63}Cu -NMR in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_3\text{O}_7$

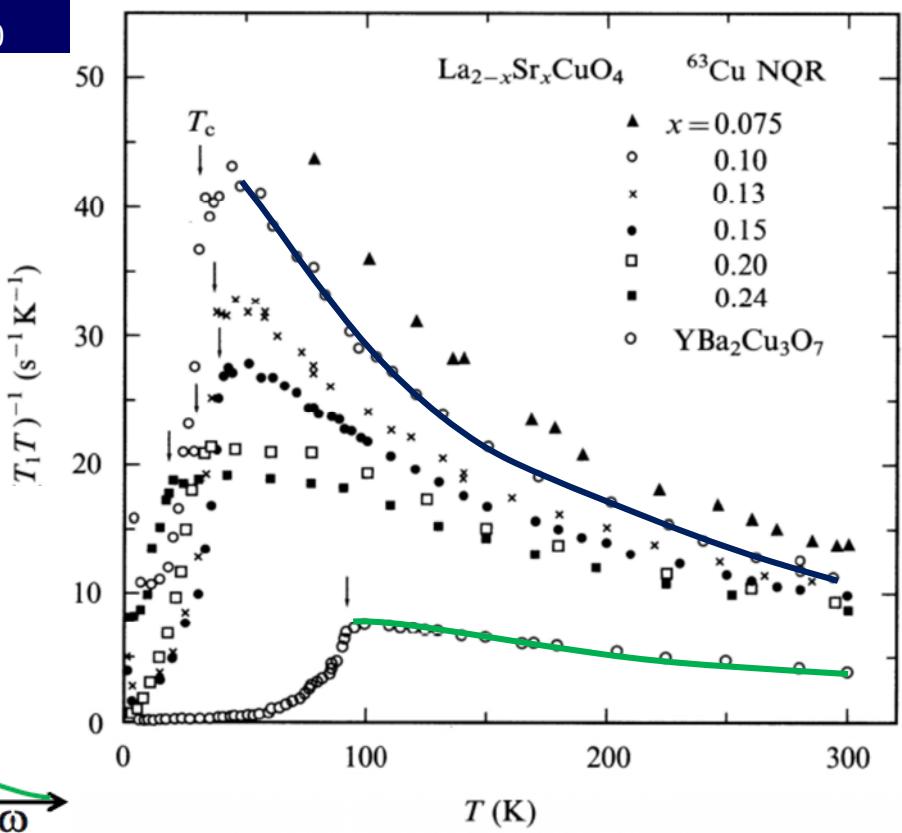
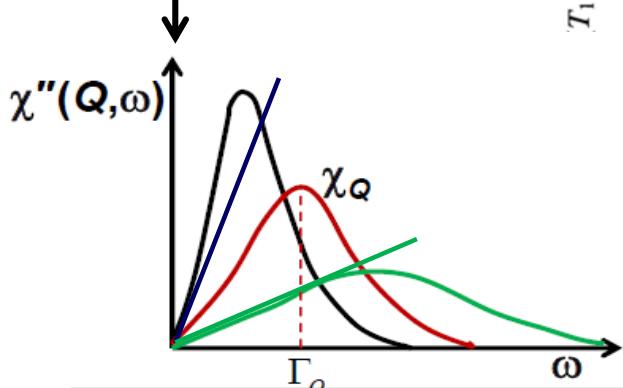
$$1/T_1 T \propto \chi''(\mathbf{Q}, \omega_0) / \omega_0$$

ω_0 : NQR frequency

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

$$\chi'(\mathbf{Q}, \omega=0) \sim \int [\chi''(\mathbf{Q}, \omega)/\omega] d\omega$$

$\rightarrow \infty$ (AFM order)



$1/T_1 T (x, T)$ probes AFM spin fluctuations

Antiferromagnetic Spin Fluctuations in LSCO

2D-AFM spin fluctuations :

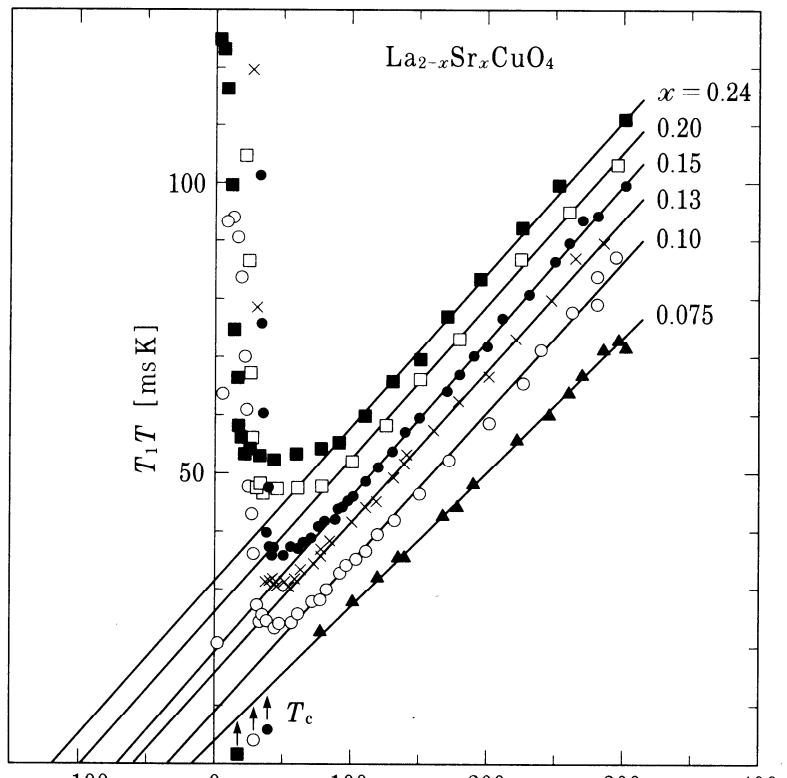
$$1/T_1 T \propto \chi_Q(T)$$

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



$$T_1 T \propto 1 / \chi_Q(T)$$

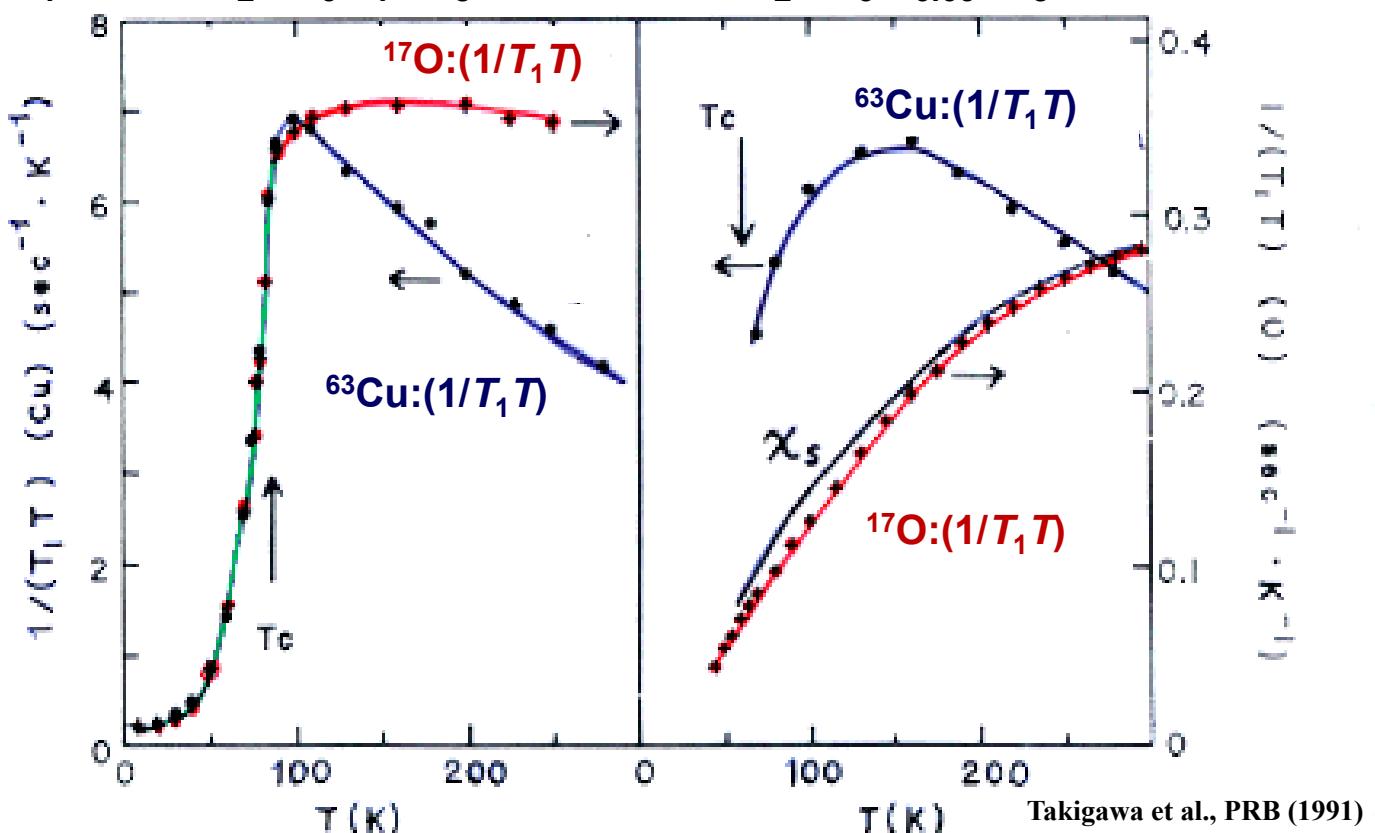
$$\propto (T+\theta)$$



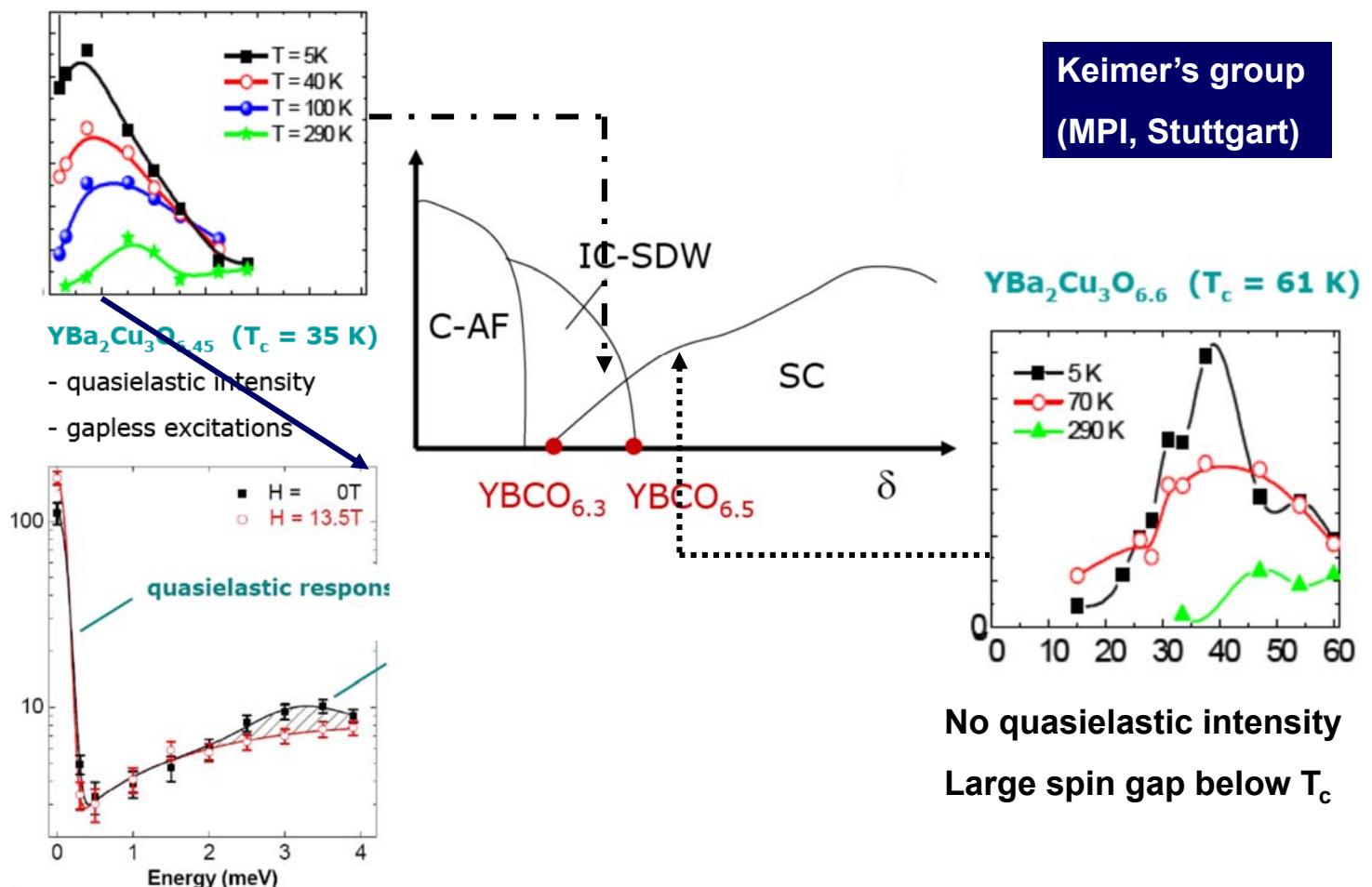
$\theta \rightarrow 0$ (QCP) at $x \sim 0.05$

Characteristics of Antiferromagnetic Spin Fluctuations in HTSC

$1/T_1 T$: $\text{YBa}_2\text{Cu}_3\text{O}_7$ ($T_c = 93$ K) $\text{YBa}_2\text{Cu}_3\text{O}_{6.63}$ ($T_c = 60$ K)



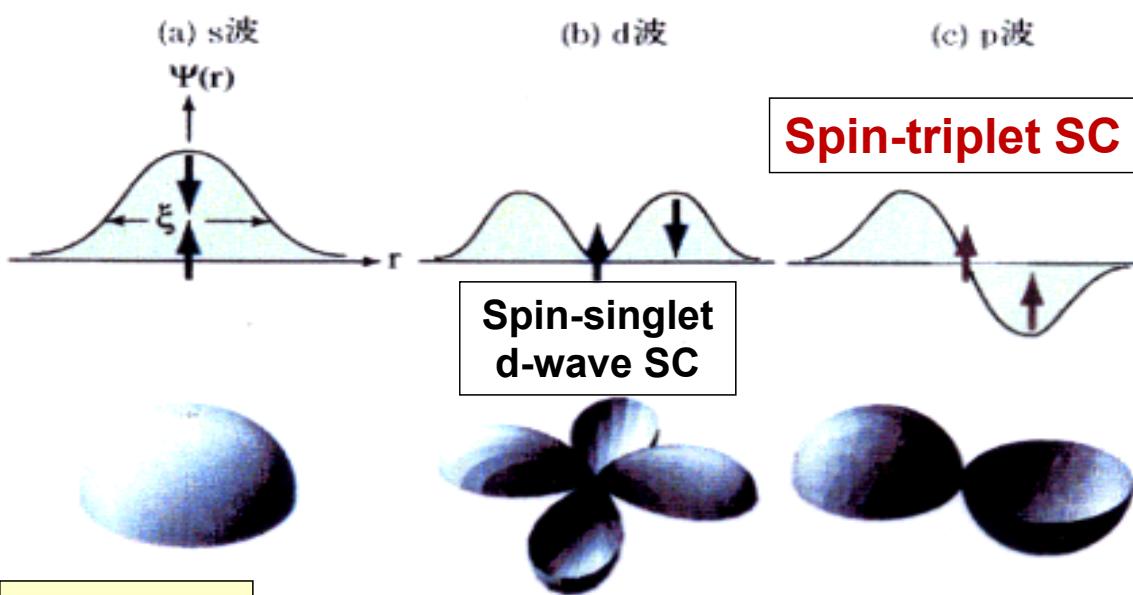
Spin Fluctuations in $\text{YBa}_2\text{Cu}_3\text{O}_x$ probed by neutron



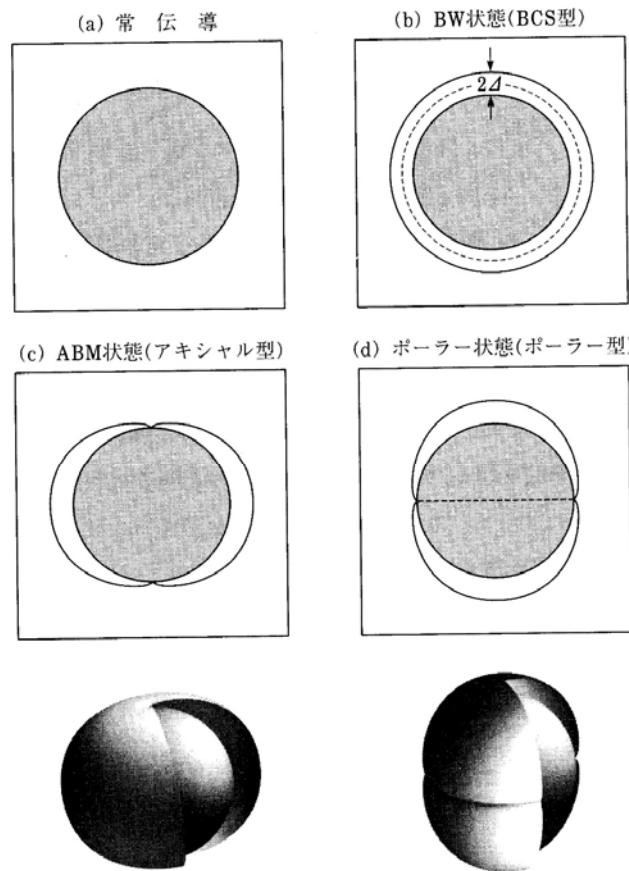
スピンゆらぎによって媒介されるd波超伝導

Spin-fluctuations mediated d-wave superconductivity

Various types of SC pairing states

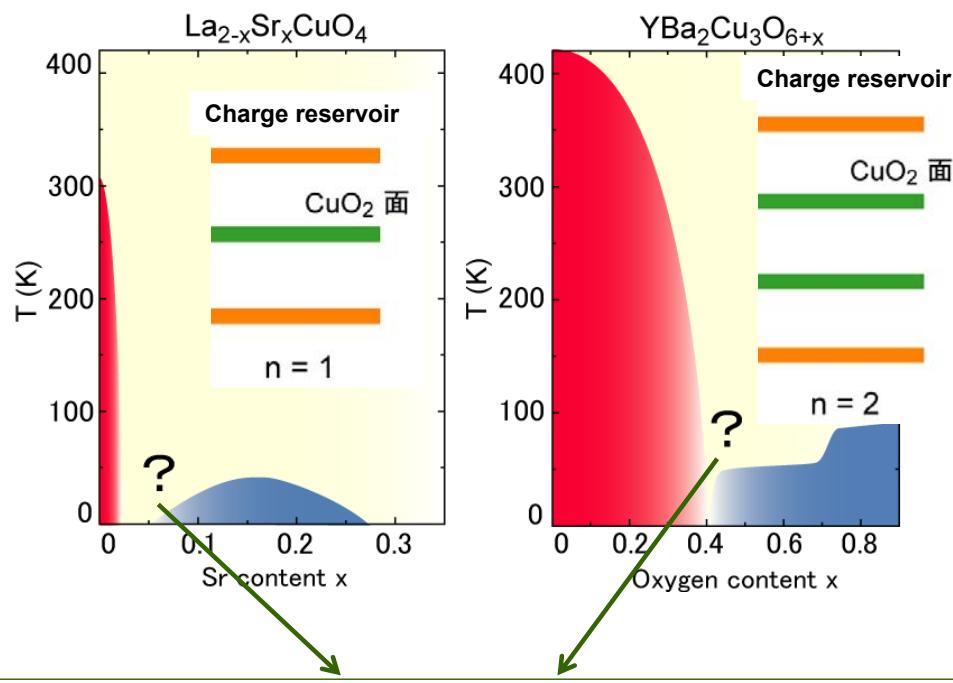


Energy gap structure of unconventional SC



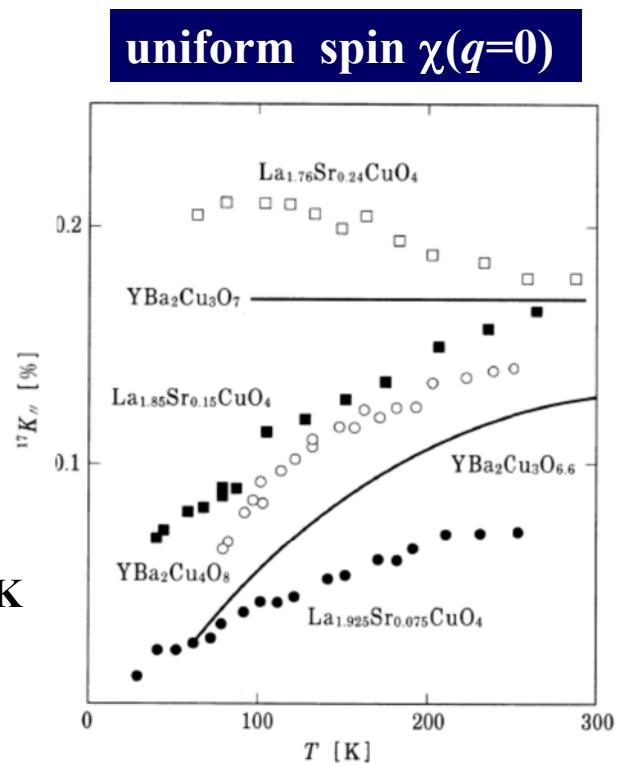
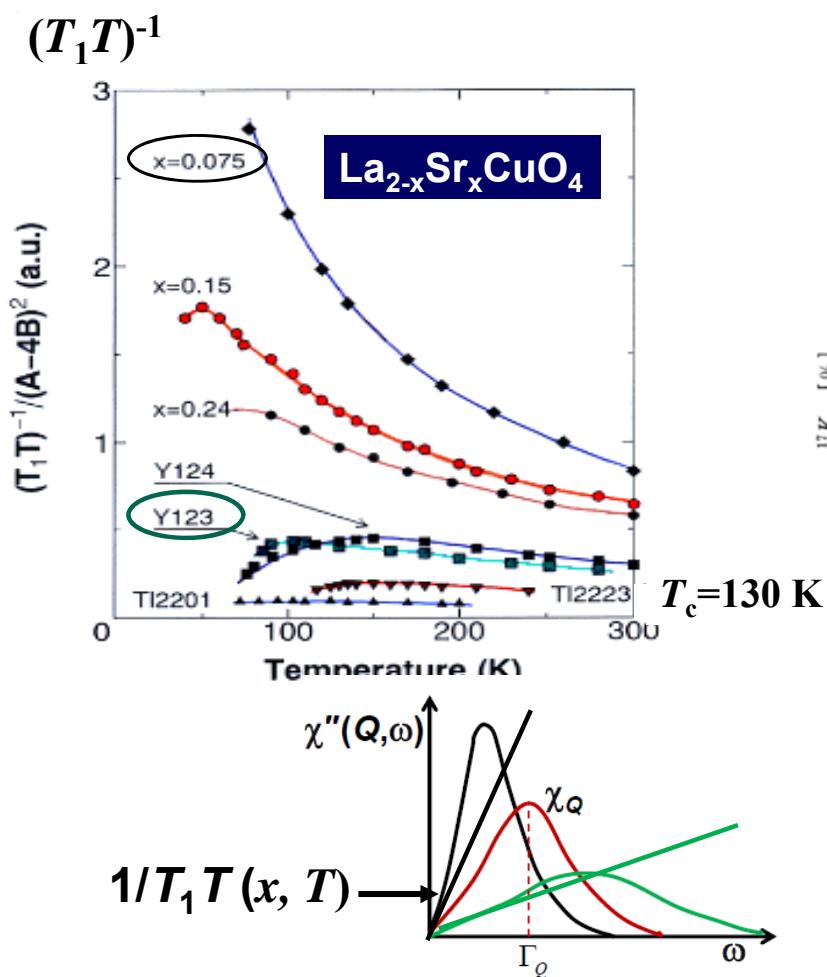
7-3図 BW, ABM, ポーラー状態に対するエネルギーギャップの様子

Phase Diagram of high-T_c copper oxides



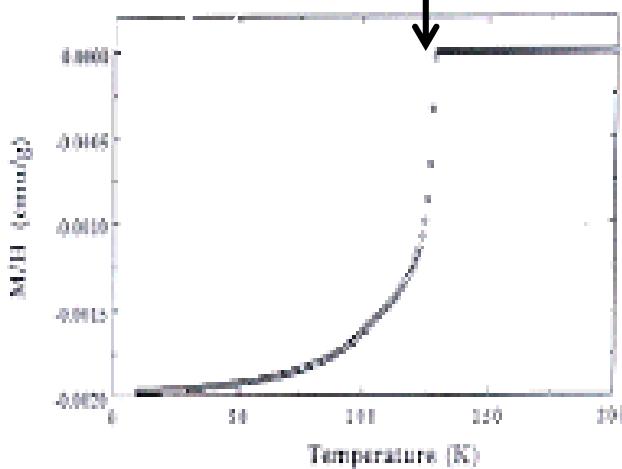
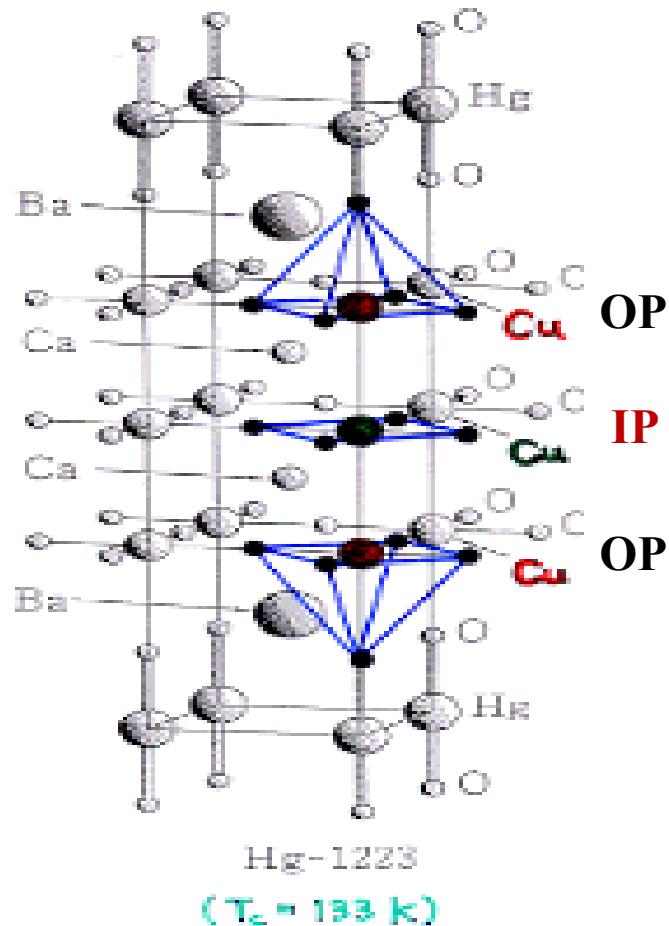
- i) Intrinsic phase in a underdoped region ?
- ii) Effect of a number of CuO_2 planes ?

Summary: Carrier-density Dependence of Spin Fluctuations

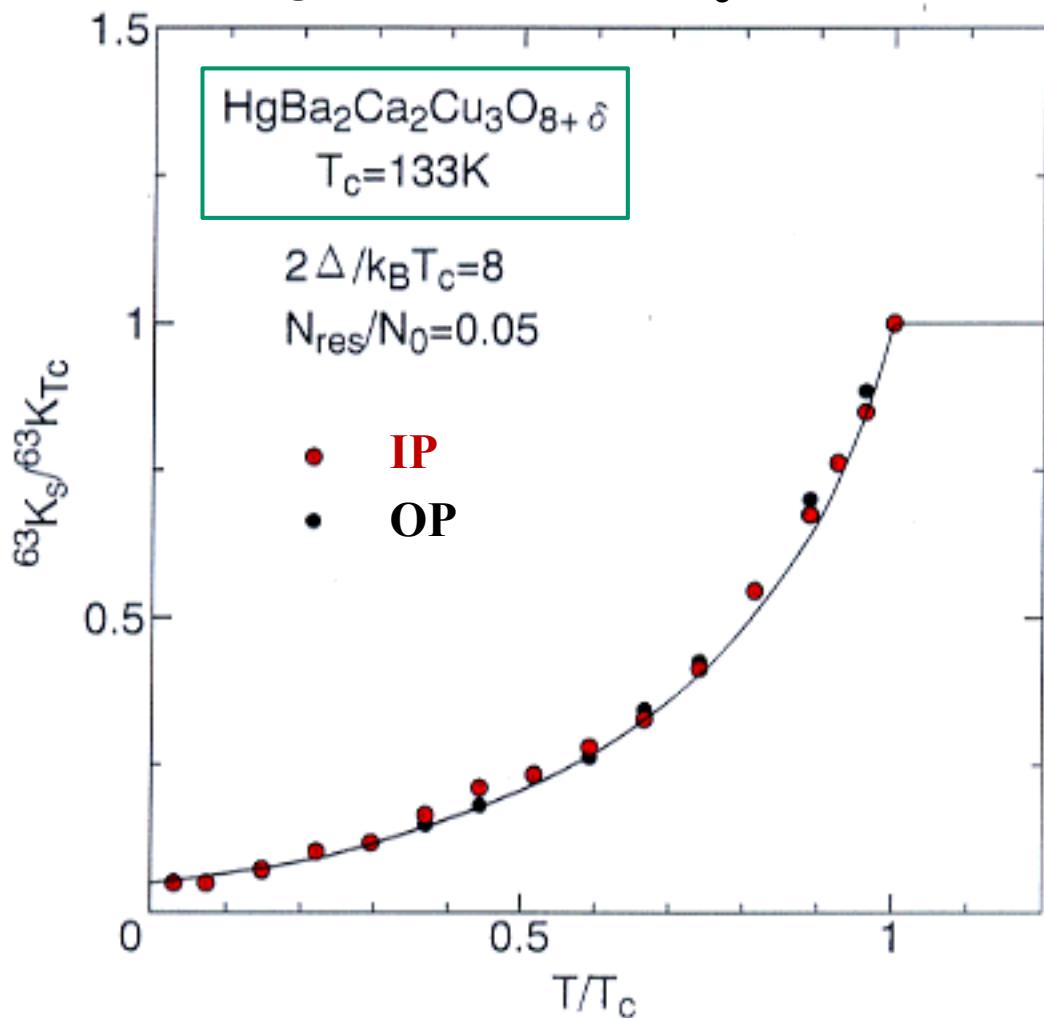


$\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$

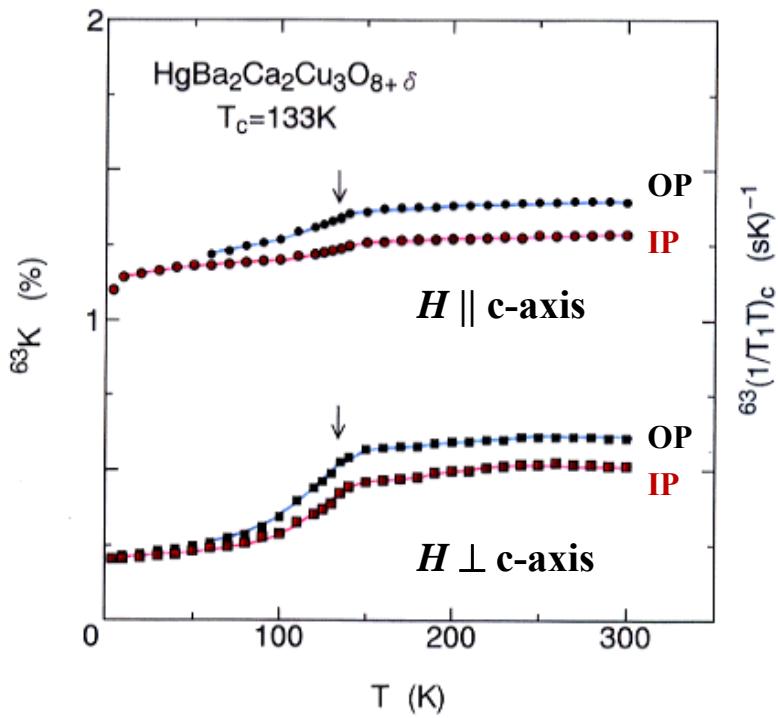
$T_c = 133 \text{ K}$



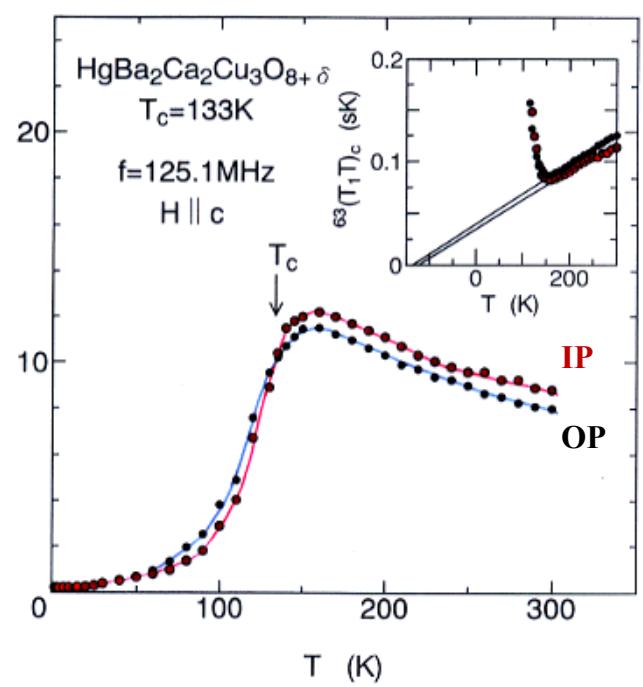
Knight Shift below T_c



Knight Shift

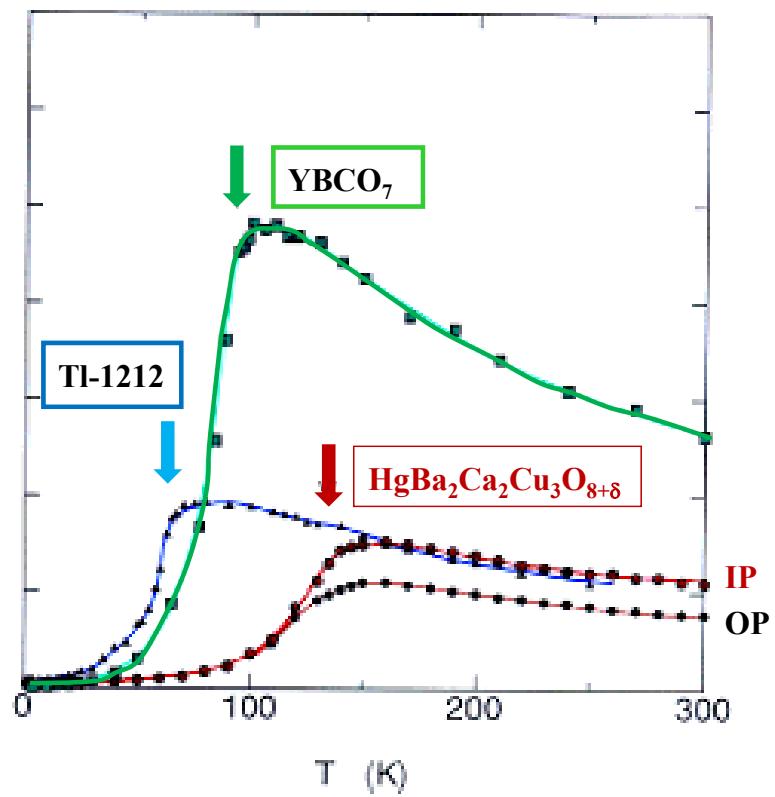


$1/T_1 T$



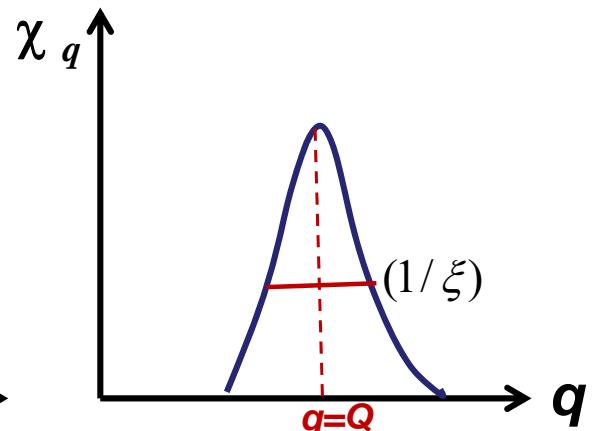
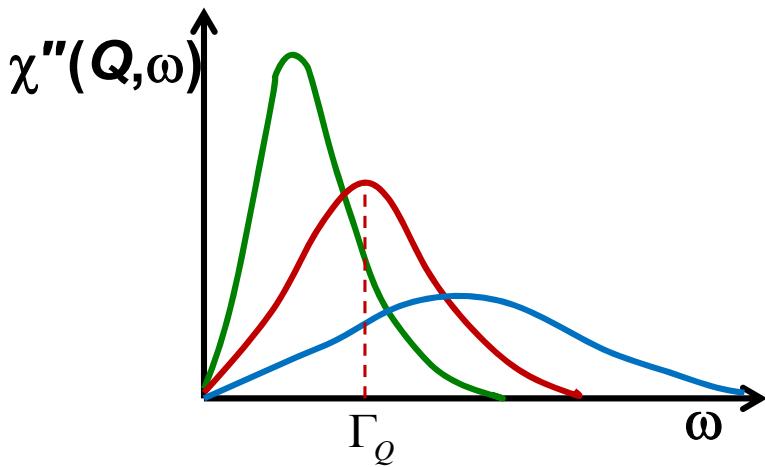
$$\boxed{\frac{1}{T_1 T}} \propto \frac{\chi''(Q, \omega_{NMR})}{\omega_{NMR}} = \frac{\chi_Q}{\Gamma_Q}$$

$$\frac{\chi''(q, \omega)}{\omega} = \frac{\chi_Q}{1 + (q - Q)^2 \xi^2} \frac{\Gamma_q}{\omega^2 + \Gamma_q^2}$$



When Γ_Q increases, T_c is enhanced.

The Spin-fluctuations theory: $T_c \propto \xi^2 \Gamma_Q \propto \frac{T_1 T}{T_{2G}^2}$

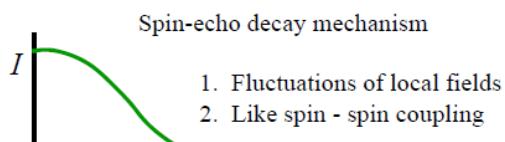


AFM spin fluctuations are described by

$$\frac{\chi''(q, \omega)}{\omega} = \frac{\chi_Q}{1 + (q - Q)^2 \xi^2} \frac{\Gamma_q}{\omega^2 + \Gamma_q^2} \quad \Gamma_q = \Gamma_Q [1 + (q - Q)^2 \xi^2]$$

Temperature dependence of spin-echo decay rate $1/T_{2G}$

Magnetic Correlation Length: ξ

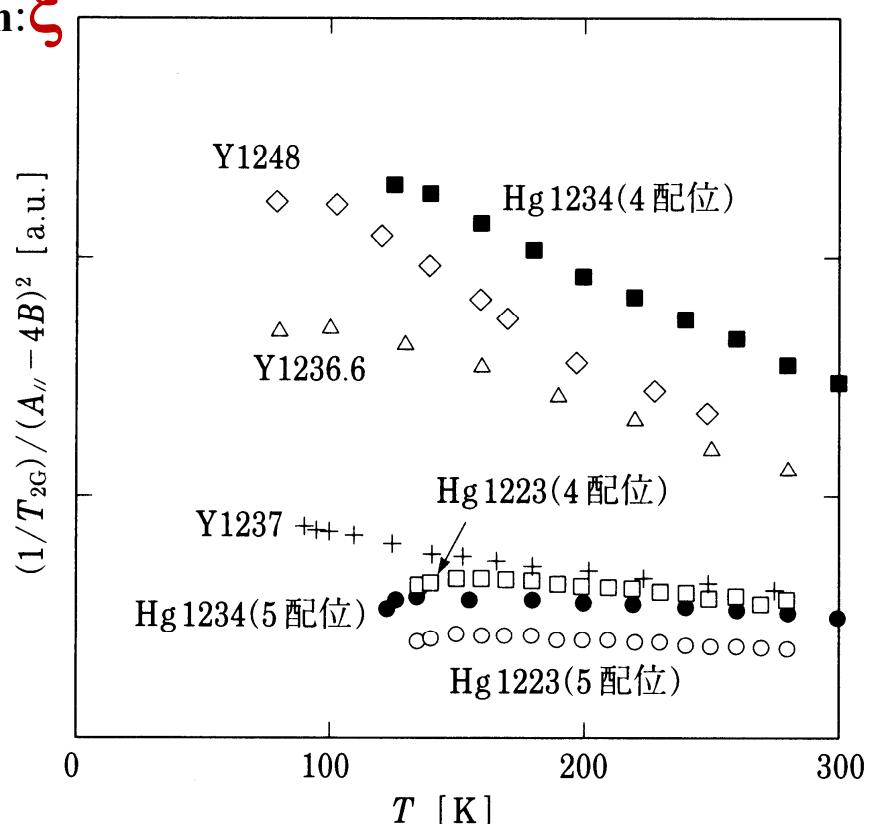
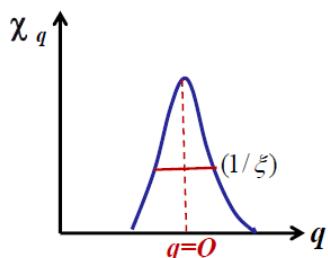


$$H_{\text{spin-spin}} = \sum_{j,k} \vec{I}_j \cdot \tilde{a}_{jk} \cdot \vec{I}_k$$

$$= \sum_j \hbar \gamma_N \vec{I}_j \cdot \vec{H}_j^{\text{loc}}$$

$$\vec{H}_j^{\text{loc}} = \frac{1}{\hbar \gamma_N} \sum_k \tilde{a}_{jk} \cdot \vec{I}_k \sim A_{\text{hf}}^2 \xi$$

indirect nuclear spin-spin coupling via spin fluctuations



6-26 図 種々の系の CuO_2 面内の ^{63}Cu の $1/(A_{||} - 4B)^2 T_{2G}$

Spin Fluctuations mediated d-wave superconductivity

Spin-fluctuations parameters derived from T_1 and T_{2G}

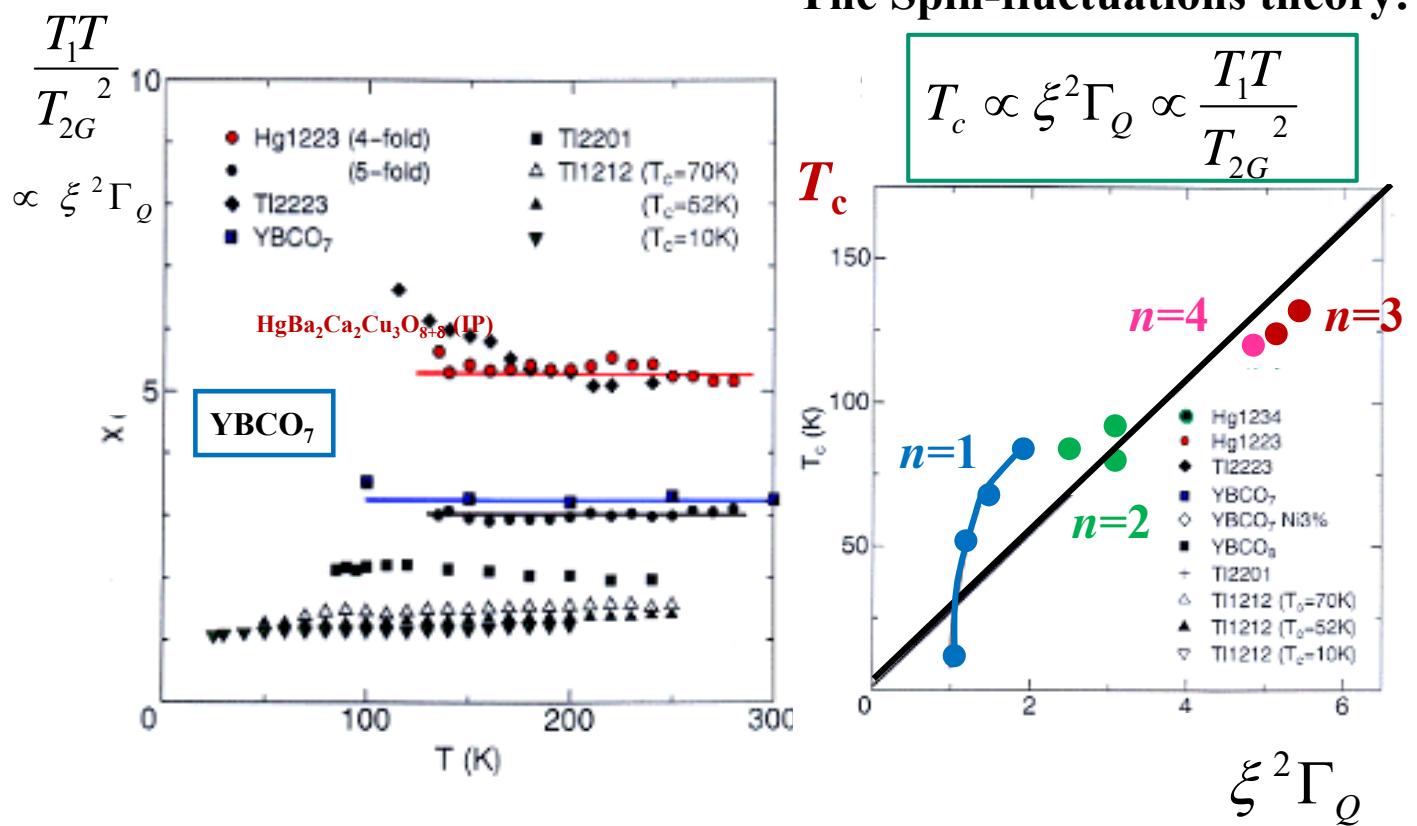
$$\left(\frac{1}{T_{2G}}\right)^2 = \frac{0.69(\gamma_N \hbar)^4 (A_c - 4B)^4}{32\pi \hbar^2} \frac{\chi_Q^2}{\xi^2} \sim \frac{\chi_Q^2}{\xi^2} \sim \xi^2$$

$$\left(\frac{1}{T_1 T}\right)_c = \frac{k_B(\gamma_N \hbar)^2 (A_c - 4B)^2}{2\hbar^2 (2R - 1)} \frac{\chi_Q}{\Gamma_Q \xi^2} \sim \frac{\chi_Q}{\Gamma_Q \xi^2} \sim 1/\Gamma_Q$$

$$\frac{(T_1 T)_c}{(T_{2G})^2} = \frac{0.69(\gamma_N \hbar)^2 (A_c - 4B)^2 (2R - 1)}{16\pi \hbar k_B} \frac{\chi_Q \hbar \Gamma_Q}{\Gamma_Q \xi^2}$$

Relationship between spin-fluctuation parameters and T_C

The Spin-fluctuations theory:



BCS theory predicts the many-body ground state for the superconducting state described by the following wave function as

$$\psi_{\text{BCS}} = \prod_k (u_k + v_k c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger) |0\rangle \quad (1)$$

The BCS Hamiltonian is given by

$$\mathcal{H} = \sum_{k,\sigma} \left(\frac{\hbar^2 k^2}{2m} - \mu \right) c_{k\sigma}^\dagger c_{k\sigma} - \sum_{k,k'} V_{kk'} c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger c_{-k'\downarrow}^\dagger c_{k'\uparrow}$$

Using (1), by minimizing the expectation value of the above BCS Hamiltonian, the parameters u_k and v_k are expressed as follows;

$$u_k^2 = \frac{1}{2} \left(1 + \frac{\xi_k}{E_k} \right), \quad v_k^2 = \frac{1}{2} \left(1 - \frac{\xi_k}{E_k} \right) \quad \text{with} \quad E_k = (\xi_k^2 + |\Delta_k|^2)^{1/2}$$

Here note that $\Delta_k = \langle c_{-k\downarrow} c_{k\uparrow} \rangle$ based ion the mean field approximation and when it is assumed as k -independent, we get the following relations using the density of state at the Fermi Level, $N(0)$ and the Deby frequency, ω_D .

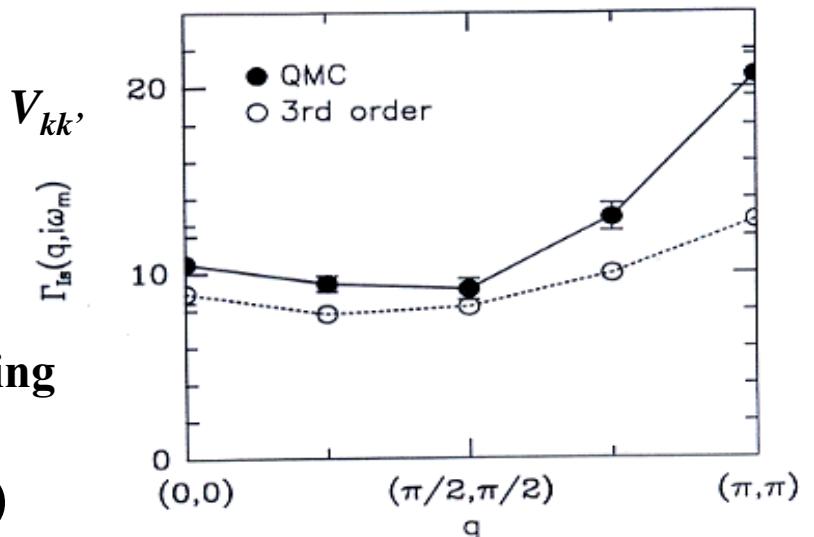
$$1 = \frac{1}{2} N(0) V \int_{-\hbar\omega_D}^{\hbar\omega_D} \frac{1}{\sqrt{\xi^2 + \Delta^2}} d\xi \quad \Delta = \frac{\hbar\omega_D}{\sinh\left(\frac{1}{N(0)V}\right)} \approx 2\hbar\omega_D \exp\left(-\frac{1}{N(0)V}\right)$$

$$\Delta_k = -\sum_{k'} V_{kk'} \frac{\Delta_{k'}}{2E_{k'}} \tanh\left(\frac{E_{k'}(T)}{2k_B T}\right)$$

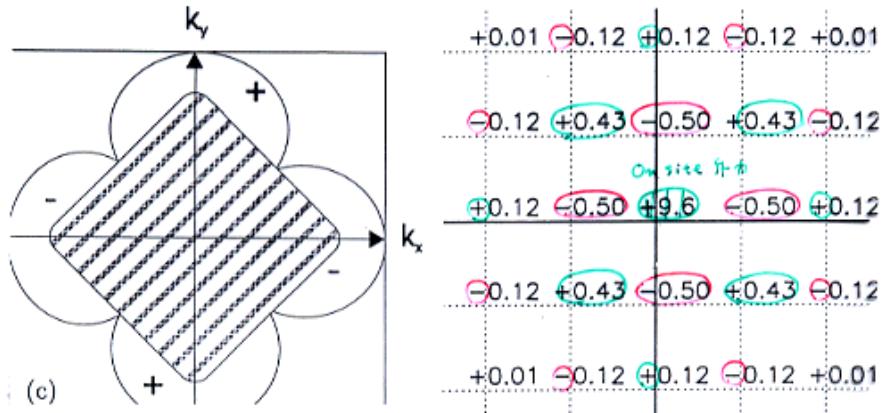
d-wave superconductivity due to the on-site Coulomb repulsive interaction

Gap equation for $d_{x^2-y^2}$ pairing

$$\Delta_{\mathbf{k}} = -\sum_{\mathbf{k}'} V_{\mathbf{k},\mathbf{k}'} (\Delta_{\mathbf{k}'}/E_{\mathbf{k}'})$$

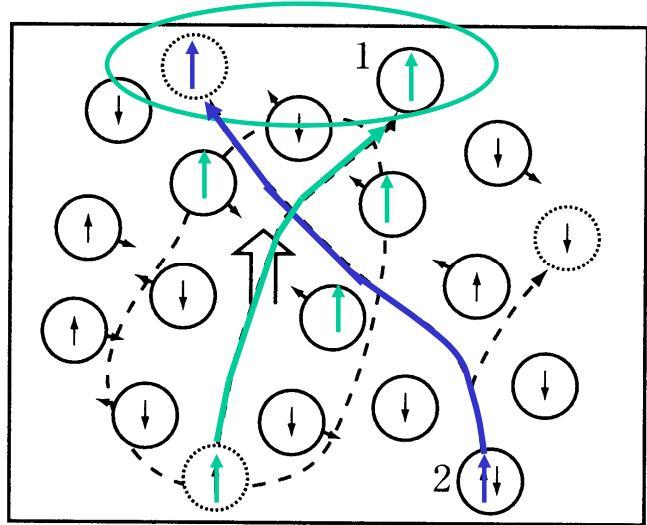


$d_{x^2-y^2}$ pairing

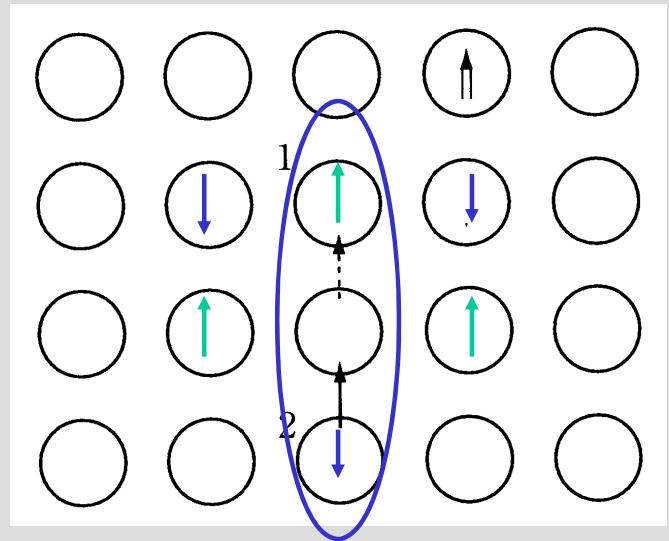


Magnetic fluctuations mediated SC mechanism

Ferromagnetic case



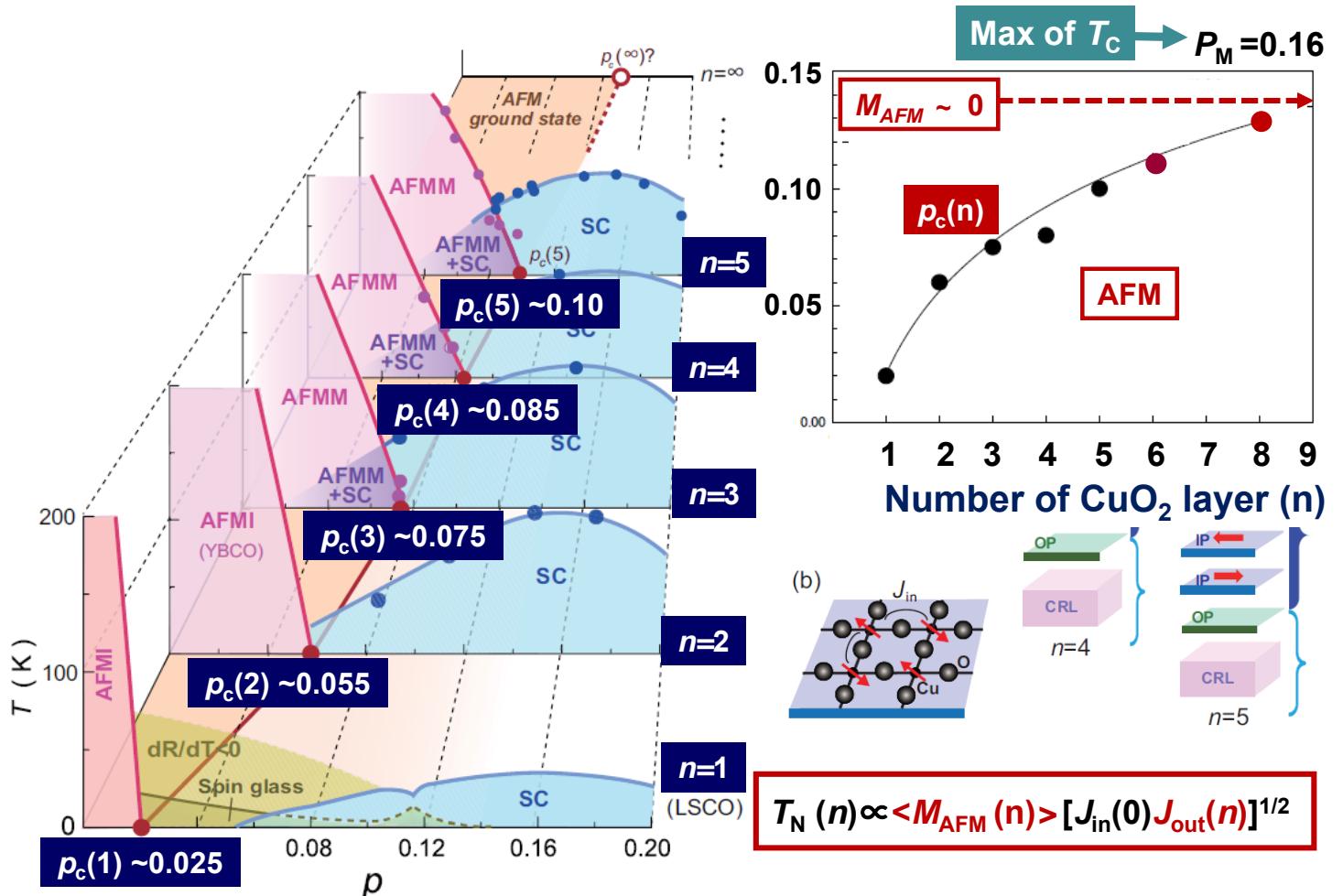
Antiferromagnetic case



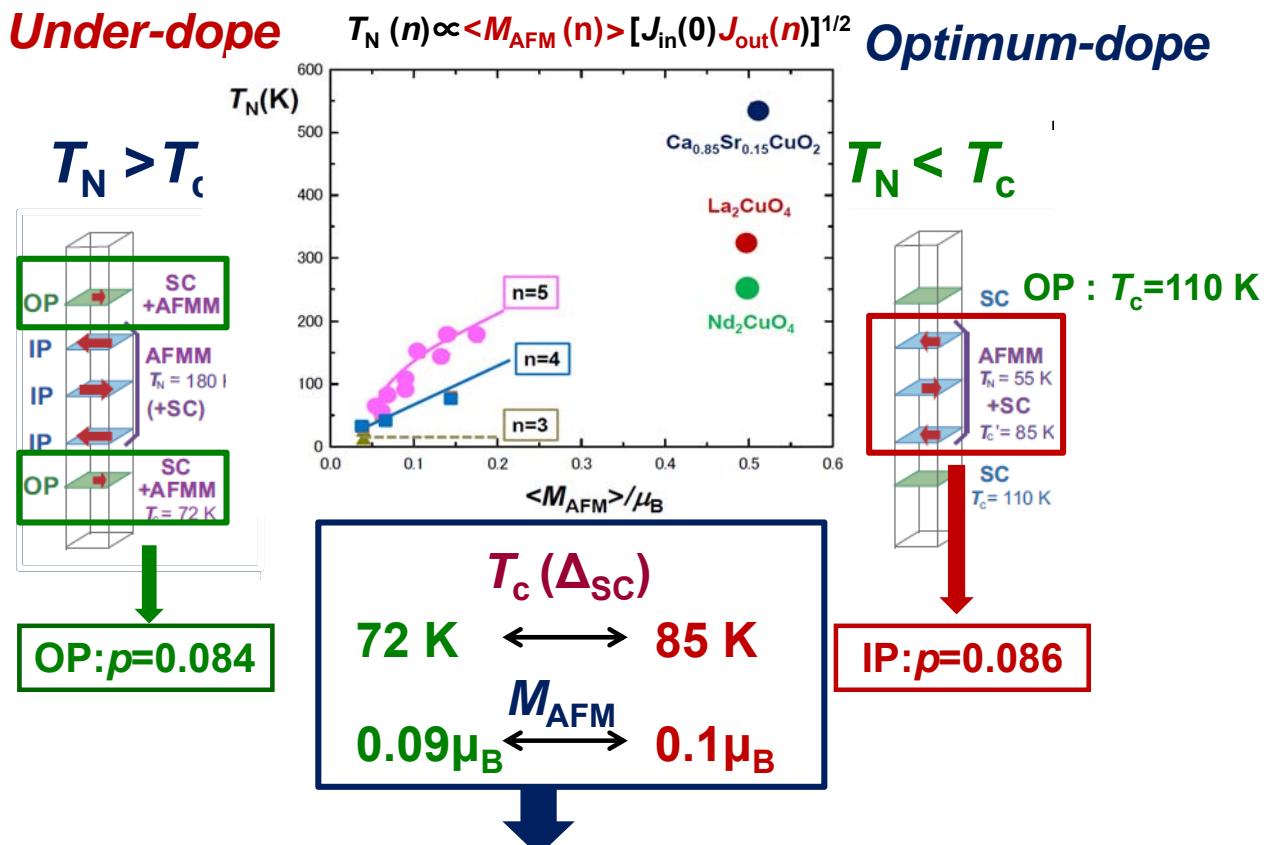
Recent Topics in Multilayer High- T_c Cuprates

- Novel phase diagram for single CuO₂ plane
deduced from n = 3, 4, 5 and 6 compounds (new)

CuO₂-layer number dependence of phase diagram



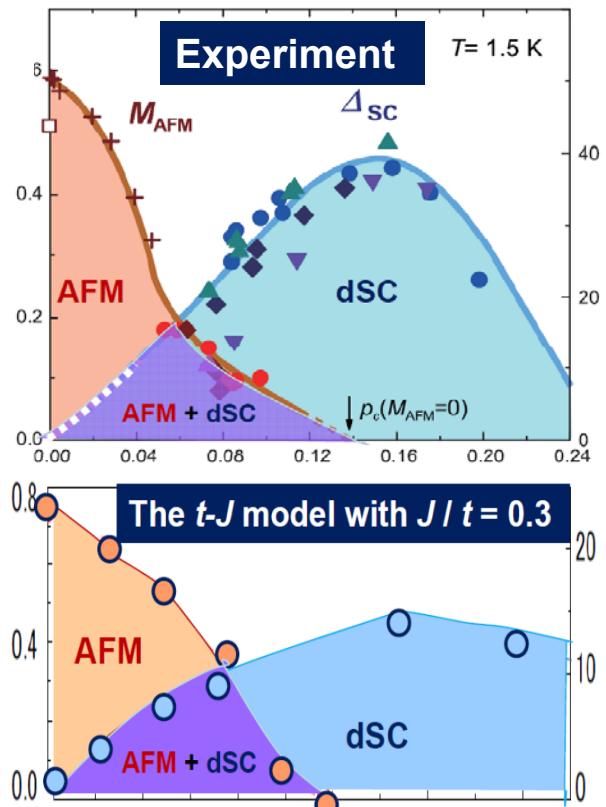
AFM and SC characteristics inherent to a single CuO₂ plane



Summary (II)

	Copper Oxides
Mother compound	AFM-Mott Insulators ($T_N \sim 500$ K)
Phase diagram	Carrier doping
Electronic state	Single band
SC symmetry	d wave ($T_c = 135$ K at $P = 0$)
Pairing interaction	AFM superexchange Interaction J

$$H = \sum_{\langle i, j \rangle} t_{ij} a_{i\sigma}^\dagger a_{j\sigma} + \sum_i J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$$



In strong coupling regime of electron correlation ($U > 8t$):
 Doped Mott Insulator is the superconductor, leading to the high T_c superconductivity mediated by the AFM superexchange interaction!

第5回おわり