SSRI (Nov.16, 2006): **京大炉 事務棟会議室**

ガンマ線摂動角相関(TDPAC)による物性研究

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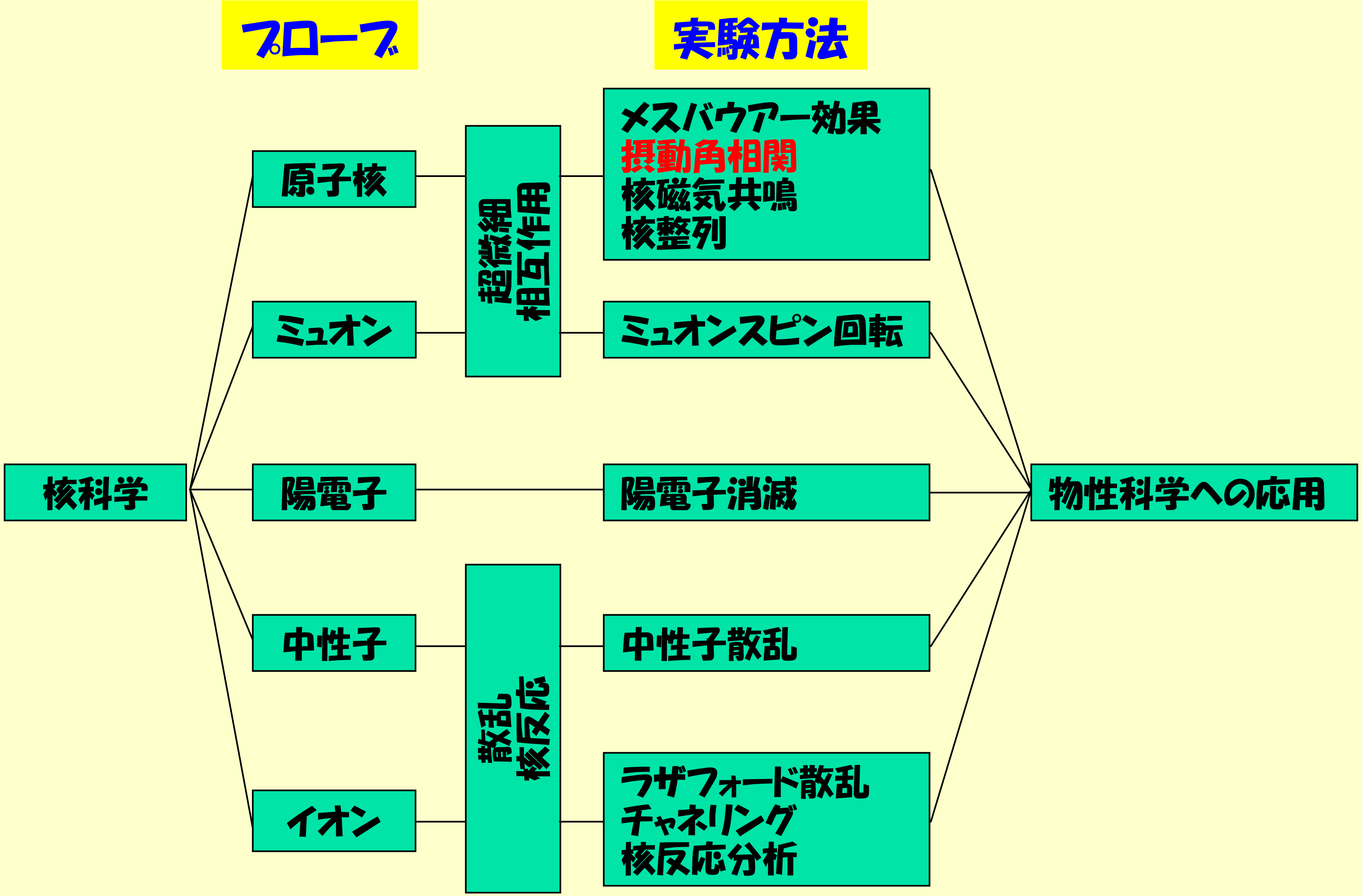
1. Outline of TDPAC

(Time-Differential Perturbed Angular Correlation)

2. TDPAC experiments at KURRI

- **Ferroelectric phase transition of $LiTaO_3$**
- **Local magnetic fields in the Mo layer of Mo/Fe multilayer**
- **Hopping motion of Ce in graphite**
- **Hyperfine fields in a protein, mavicyanin**

核物性学



プローブ

実験方法

核科学

原子核

ミュオン

陽電子

中性子

イオン

超微細
相互作用

散乱
核反応

メスバウアー効果
摂動角相関
核磁気共鳴
核整列

ミュオンスピン回転

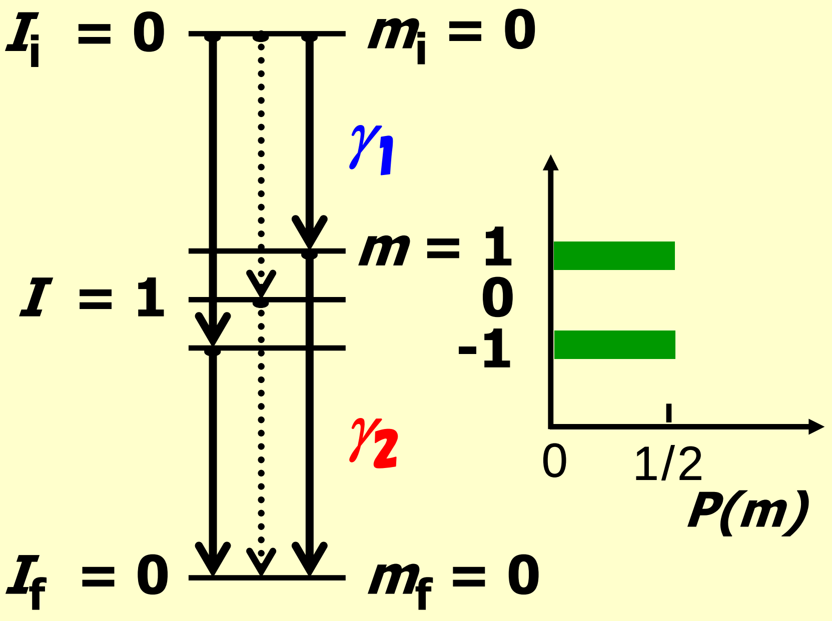
陽電子消滅

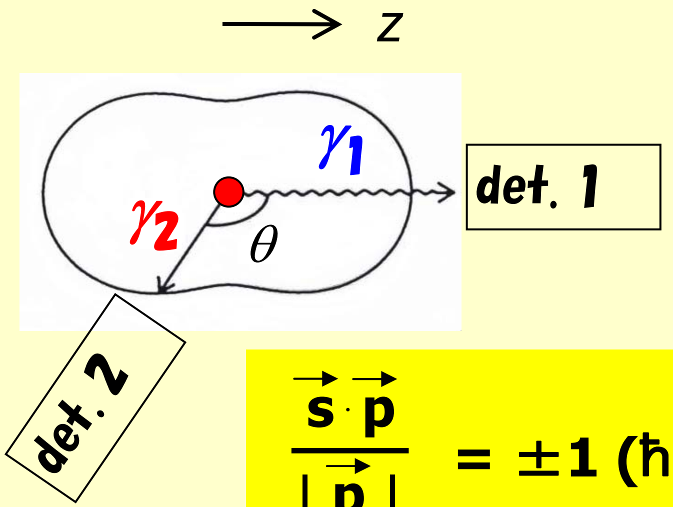
中性子散乱

ラザフォード散乱
チャネリング
核反応分析

物性科学への応用

Angular correlation of γ rays for the case of 0 → 1 → 0





$$\frac{\vec{s}\cdot\vec{p}}{|\vec{p}|} = \pm 1\,(\hbar)$$

$\vec{s}$: **photon angular momentum**

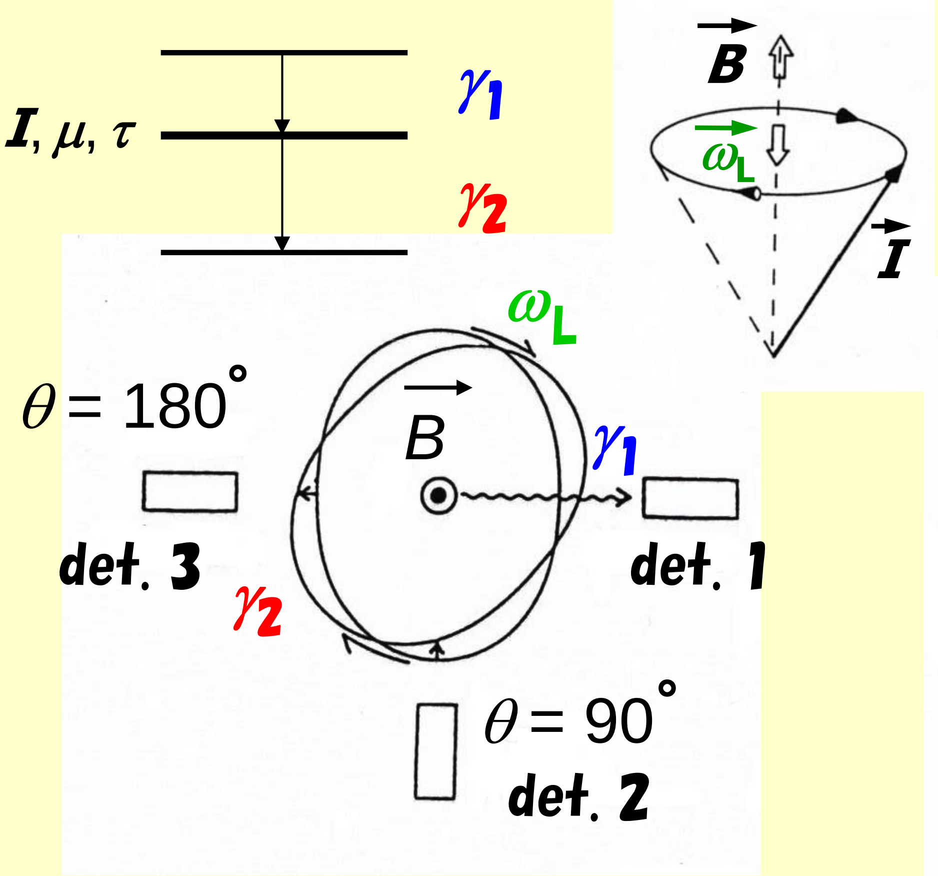
$\vec{p}$: **photon momentum**

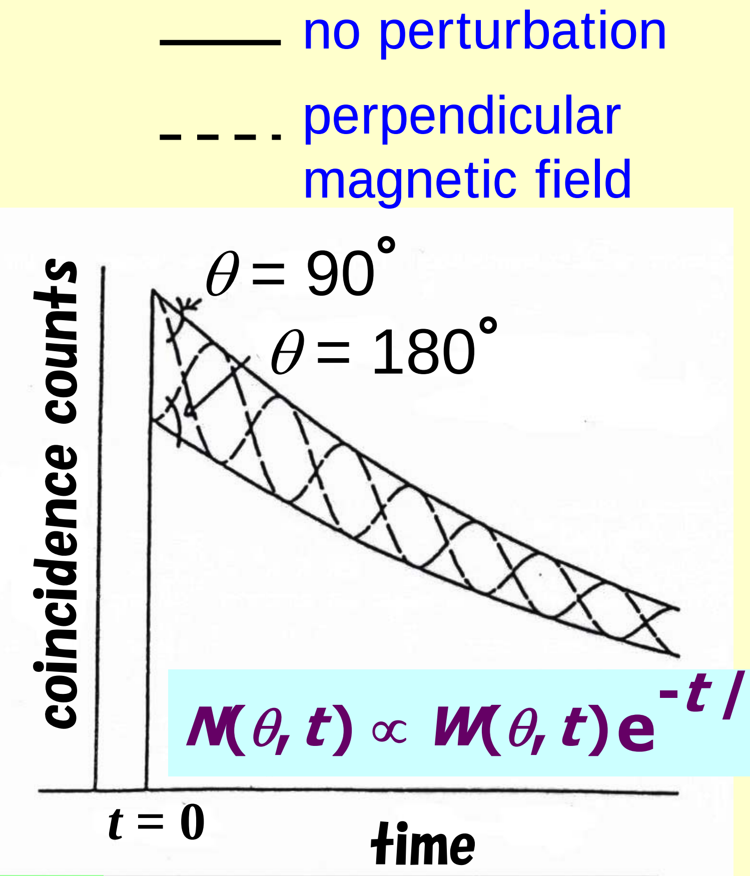
$$W(\theta) \propto 1 + \cos^2\theta$$

a method for producing a nuclear spin alignment:

$$P(m) \neq P(m'),\ P(m) = P(-m)$$

TDPAC for the case of a uniform static magnetic field perpendicular to the detector plane





$$N(\theta, t) \propto W(\theta, t)e^{-t/\tau}$$

Larmor precession frequency

$$\omega_L = -\frac{\mu B}{I\hbar}$$

$0 \rightarrow 1 \rightarrow 0$

$$W(\theta, t) \propto 1 + \cos^2(\theta - \omega_L t)$$

Interaction Hamiltonian

$$\mathcal{H} = \underbrace{- \vec{\mu}\cdot\vec{B}}_{\text{magnetic dipole interaction}} + \underbrace{\frac{eQV_{zz}}{4I(2I-1)}\left[3I_z^2 - I(I+1) + \frac{1}{2}\eta\left(I_+^2 + I_-^2\right)\right]}_{\text{electric quadrupole interaction}}$$

$$\eta = (V_{xx} - V_{yy})/V_{zz}$$

TDPAC for polycrystals

$$W(\theta, t) = \sum_{k:\,\text{even}}^{k_{\max}} A_k(1)A_k(2)G_{kk}(t)\,\mathrm{P}_k(\cos\theta) \approx 1 + A_{22}G_{22}(t)\,\mathrm{P}_2(\cos\theta)$$

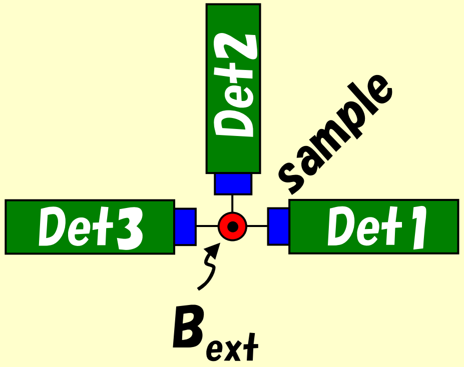
Attenuation factor for static interactions

$$G_{kk}(t) = \sum_{\substack{N,\, m_a,\, m_b \\ n,\, n'}} (-1)^{2I+m_a+m_b} \begin{bmatrix} I & I & k \\ m_a' & -m_a & N \end{bmatrix} \begin{bmatrix} I & I & k \\ m_b' & -m_b & N \end{bmatrix}$$

$$\times \exp[(-i/\hbar)(E_n - E_n')t]\langle n|m_b\rangle^* \langle n|m_a\rangle \langle n'|m_b'\rangle \langle n'|m_a'\rangle^*$$

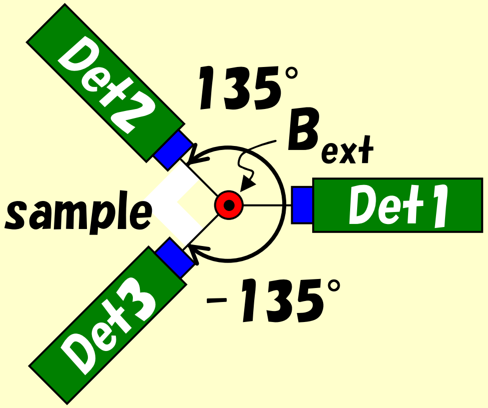
Outline of TDPAC

^{111}Cd($\leftarrow$^{111}In) in Fe_3O_4 at 300 K



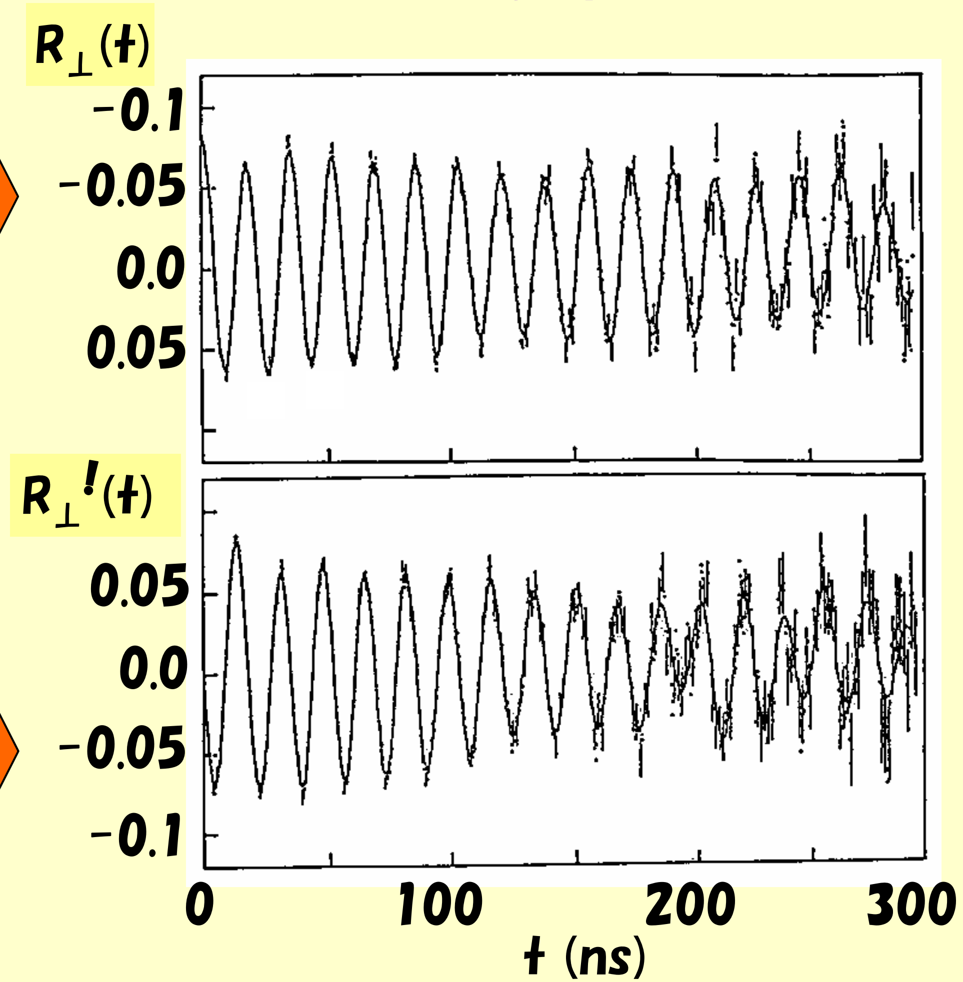
$$R_\perp(t) = \frac{N_\perp(180^\circ, t) - N_\perp(90^\circ, t)}{N_\perp(180^\circ, t) + N_\perp(90^\circ, t)}$$

$$\cong (3/4)A_{22}\cos(2\omega_L t)$$



$$R_\perp{}'(t) = \frac{N_\perp(-135^\circ, t) - N_\perp(135^\circ, t)}{N_\perp(-135^\circ, t) + N_\perp(135^\circ, t)}$$

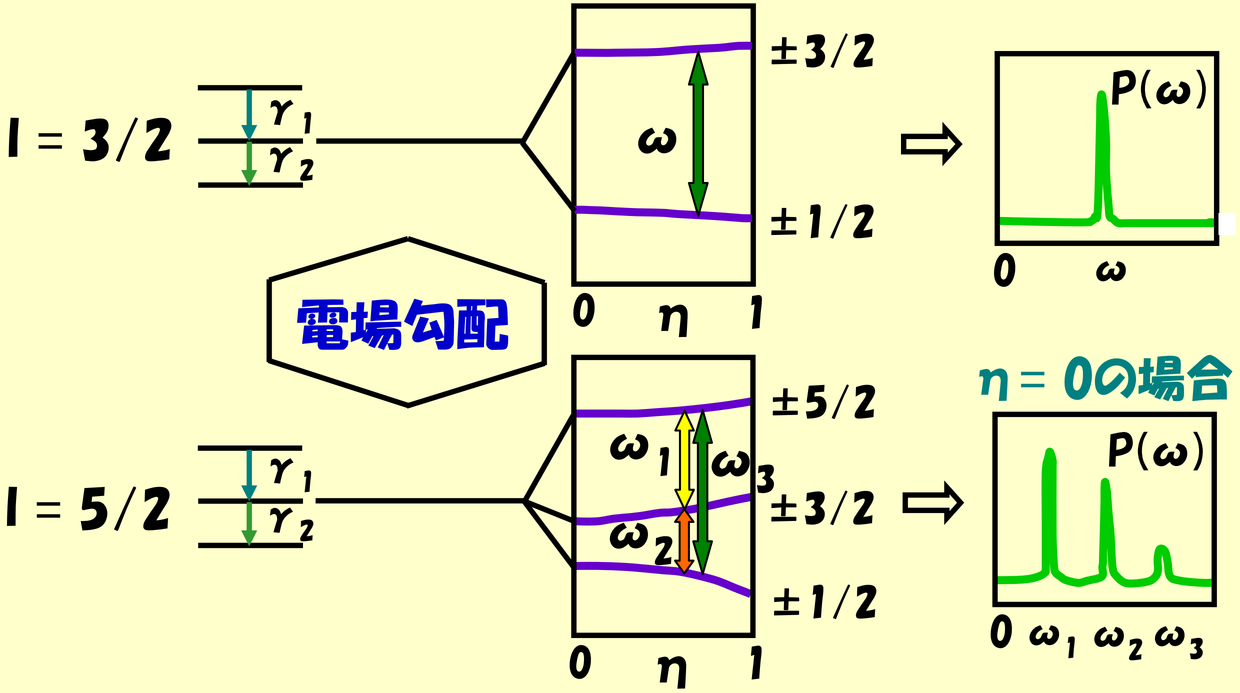
$$\cong (3/4)A_{22}\sin(2\omega_L t)$$



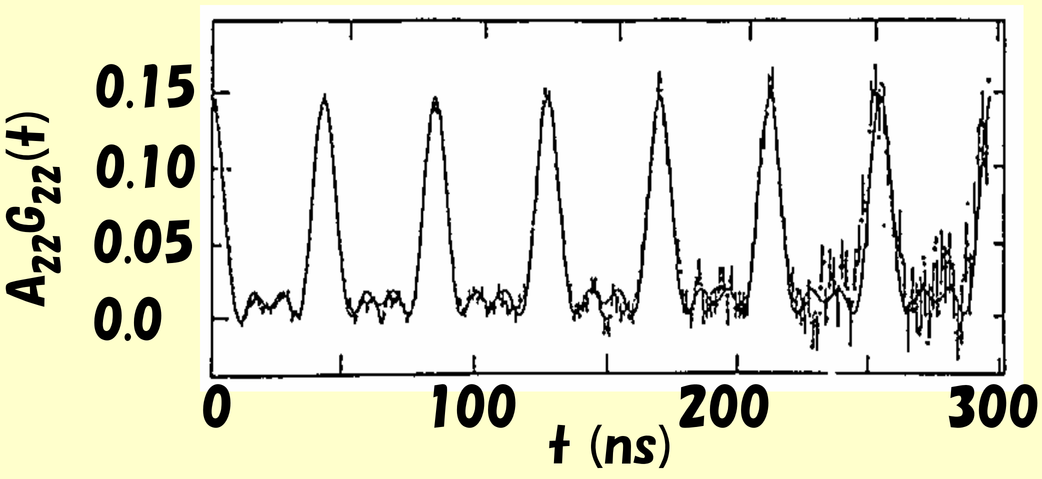
$$\omega_L = -\frac{\mu B}{I\hbar}$$

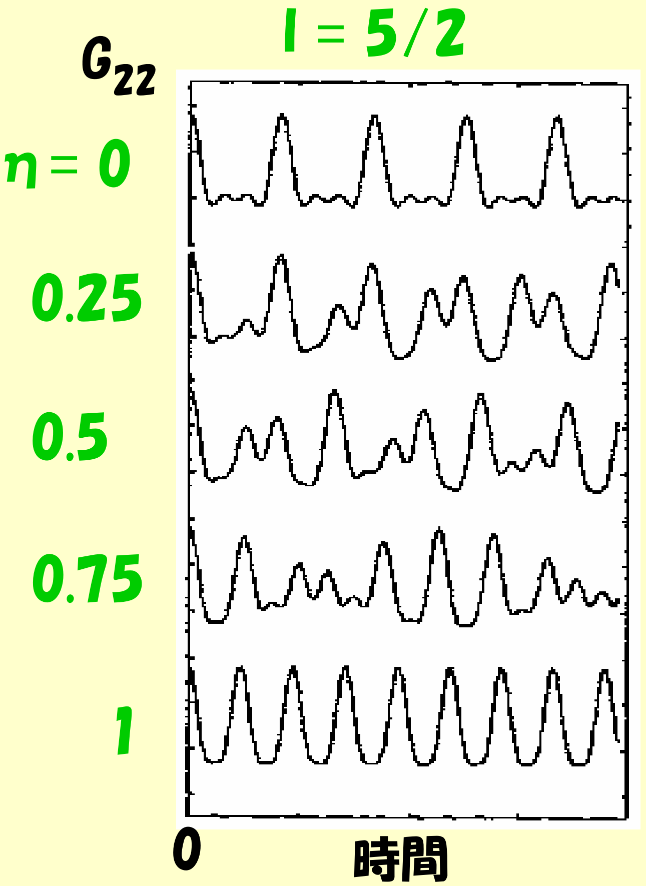
電場勾配: $|V_{zz}| \geq |V_{yy}| \geq |V_{xx}|$

非対称パラメター: $\eta = \dfrac{V_{xx} - V_{yy}}{V_{zz}}$



^{111}Cd(←^{111}In) in α-Fe_2O_3 at 987 K





$$A_{22}G_{22}(t) = \frac{2[N(180^\circ, t) - N(90^\circ, t)]}{N(180^\circ, t) + 2N(90^\circ, t)}$$

I = 5/2, η = 0の場合

$$G_{22}(t) = \frac{1}{5}\{1 + \frac{13}{7}\cos(\omega_0 t) + \frac{10}{7}\cos(2\omega_0 t) + \frac{5}{7}\cos(3\omega_0 t)\}$$

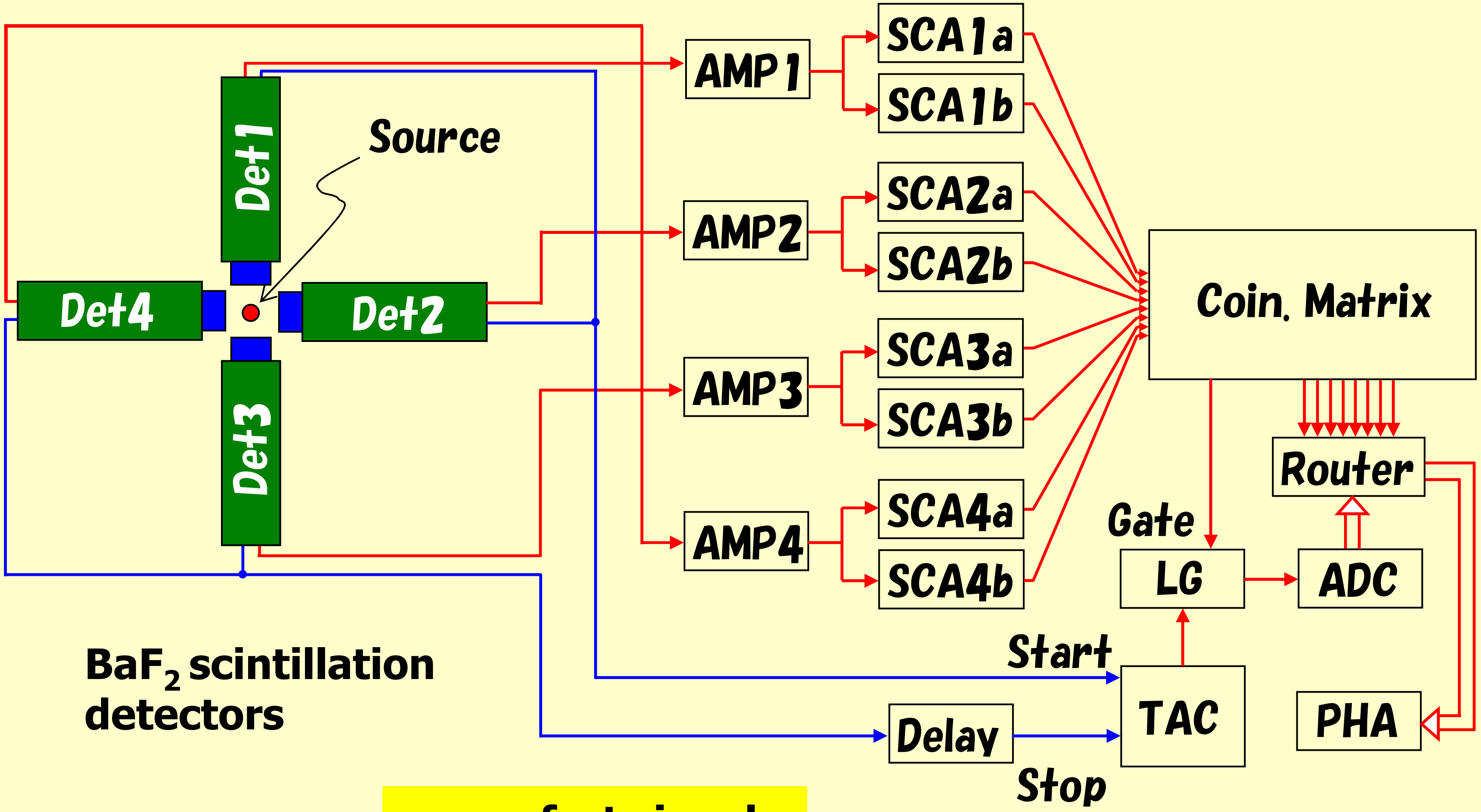
$$\omega_0 = 6\omega_Q$$

$$\omega_Q = \frac{eQV_{zz}}{4I(2I-1)\hbar}$$

Characteristics of TDPAC probes

	parent $\xrightarrow[\text{half-life}]{\text{decay mode}}$ probe	Intermediate state				γ_1(keV)	γ_2(keV)	A_{22}
		I^{π}	mean life(ns)	μ(nm)	Q(b)			
✓	$^{99}\mathrm{Mo} \xrightarrow[2.7\ \mathrm{d}]{\beta^-} {}^{99}\mathrm{Tc}$	$5/2^+$	5.2	+3.291		740	181	+0.10
	$^{99}\mathrm{Rh} \xrightarrow[16\ \mathrm{d}]{\mathrm{EC}} {}^{99}\mathrm{Ru}$	$3/2^+$	29.6	-0.284	+0.231	528 353	90 90	-0.19 -0.15
	$^{111}\mathrm{Ag} \xrightarrow[7.5\ \mathrm{d}]{\beta^-} {}^{111}\mathrm{Cd}$	$5/2^+$	123	-0.7656	+0.77	97	245	-0.13
✓	$^{111\mathrm{m}}\mathrm{Cd} \xrightarrow[49\ \mathrm{m}]{\mathrm{IT}} {}^{111}\mathrm{Cd}$					151	245	+0.18
	$^{111}\mathrm{In} \xrightarrow[2.8\ \mathrm{d}]{\mathrm{EC}} {}^{111}\mathrm{Cd}$					171	245	-0.18
✓	$^{117}\mathrm{Cd} \xrightarrow[2.5\ \mathrm{h}]{\beta^-} {}^{117}\mathrm{In}$	$3/2^+$	77.3	+0.938	(-)0.59	90	344	-0.36
✓	$^{140}\mathrm{La} \xrightarrow[1.7\ \mathrm{d}]{\beta^-} {}^{140}\mathrm{Ce}$	4^+	5.0	+4.35	0.35	329	487	-0.13
	$^{181}\mathrm{Hf} \xrightarrow[42\ \mathrm{d}]{\beta^-} {}^{181}\mathrm{Ta}$	$5/2^+$	15.6	+3.29	(+)2.35	133	482	-0.20

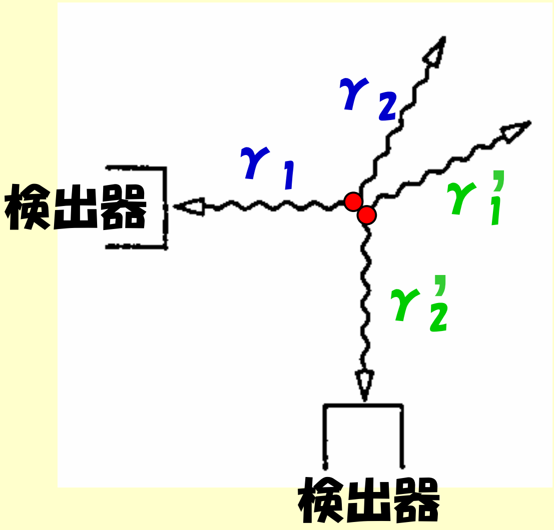
TDPAC measurement system



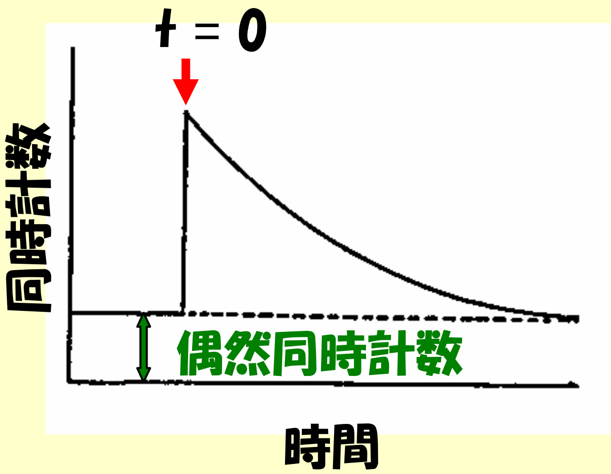
BaF$_2$ scintillation detectors

🟦 fast signal
🟥 slow signal

偶然同時計数



実験で得られる時間スペクトル



N： 単位時間に崩壊する核の数
α_i： γ_iの放出確率
ε_i： γ_iの検出効率
Ω_i： 立体角
Δt： 時間スペクトル1チャンネルあたりの時間

偶然同時計数

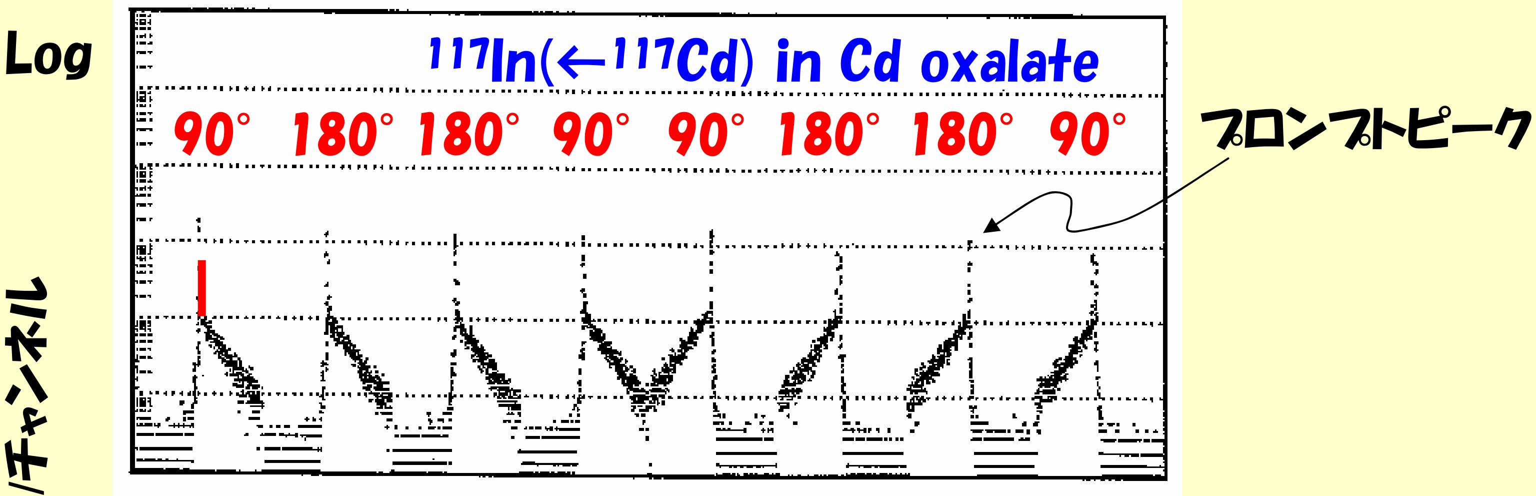
$$N^2 \alpha_1 \alpha_2 \varepsilon_1 \varepsilon_2 \Omega_1 \Omega_2 \Delta t$$

真の同時計数

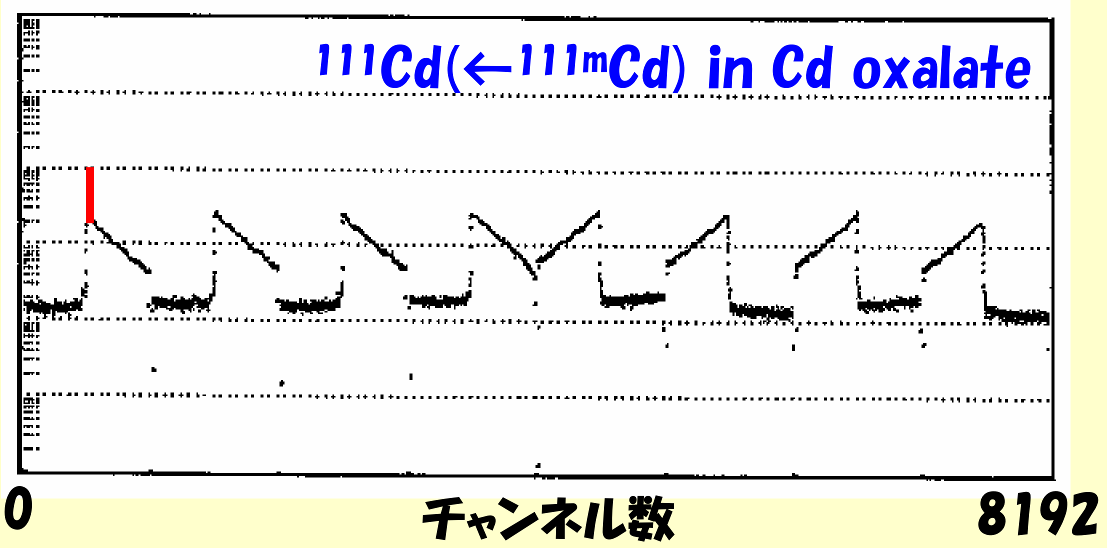
$$N \alpha_1 \alpha_2 \varepsilon_1 \varepsilon_2 \Omega_1 \Omega_2 e^{-t/\tau_N} \Delta t/\tau_N$$

真の同時計数/偶然同時計数 $= e^{-t/\tau_N} / N\tau_N$

時間スペクトルの例



カーソル Ch 515 142cts



カーソル Ch 515 2145cts

1 ch ≅ 0.45 ns

Ferroelectric phase transition of $LiTaO_3$

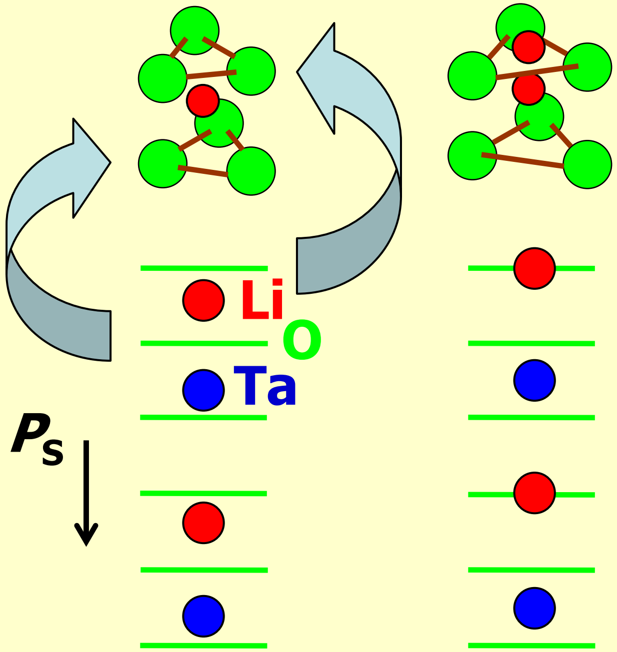
ferroelectric $LiTaO_3$ (T_C = 938 K)

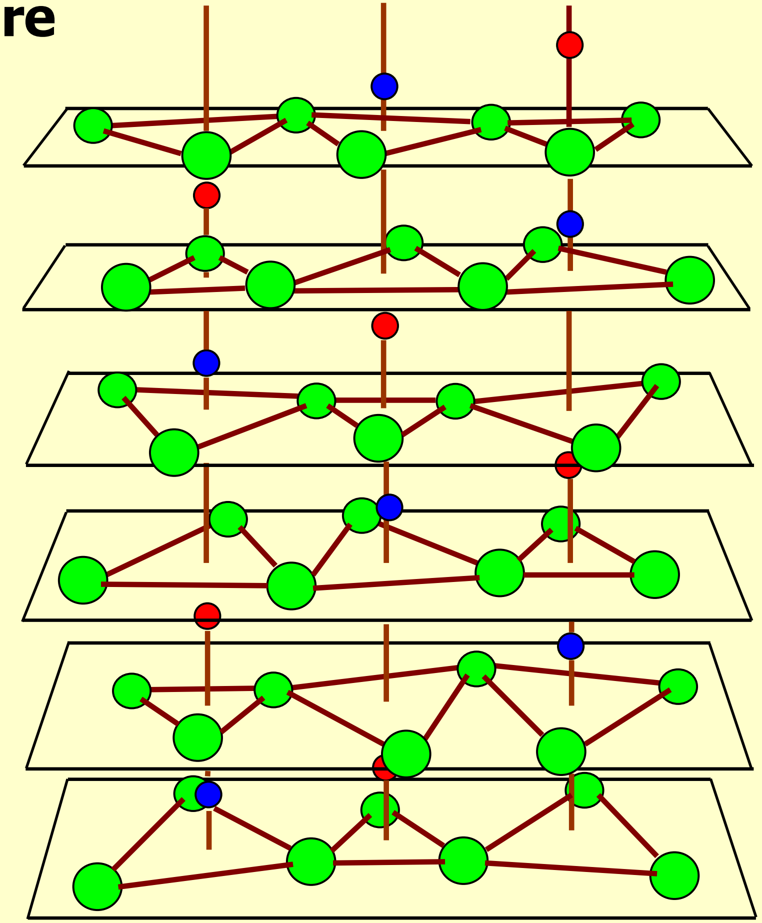
($LiNbO_3$: T_C = 1483 K)

ilmenite($FeTiO_3$)-related structure

$R3c$ ($T < T_C$) $R\bar{3}c$ ($T < T_C$)

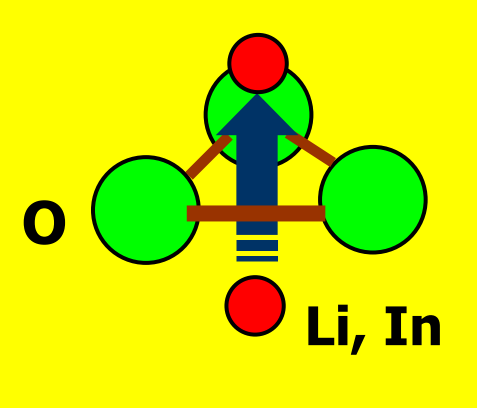
Ferroelectric paraelectric

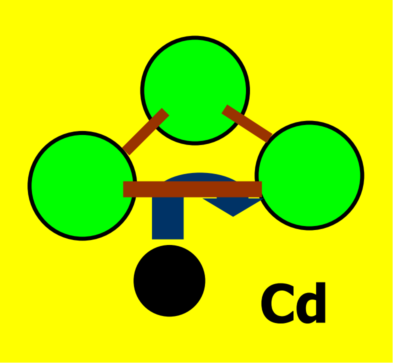




● Li^+
● Ta^{5+}
● O^{2-}

Expected behavior





	ionic radius (pm)
Li^+	76
In^{3+}	80
Cd^{2+}	95
O^{2-}	140

preparation of samples

^{116}CdO powder (96.5 at.%)
^{110}CdO powder (96 at.%)

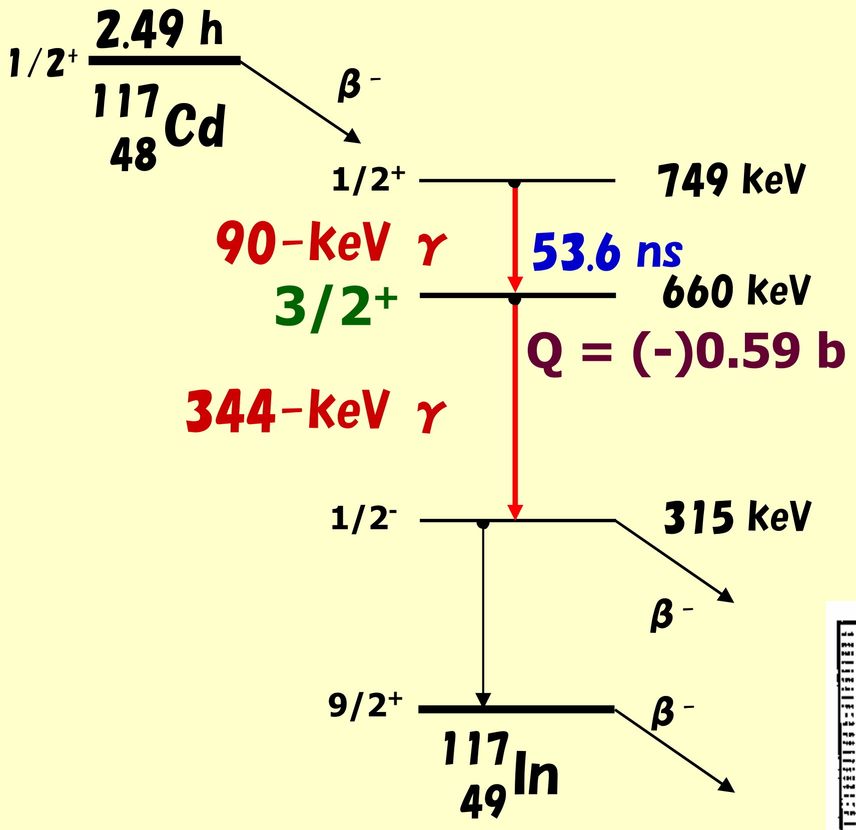
↓ *neutron irradiation*

^{117}CdO (^{116}CdO)
^{111m}CdO (^{110}CdO)

↓ *mixed with* Li_2CO_3, Ta_2O_5
pressed into pellet
heated in air

$LiTaO_3$ (^{117}Cd(^{111m}Cd))

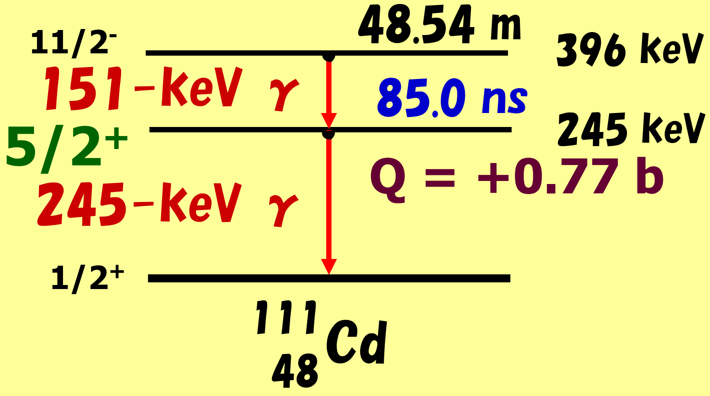
^{117}In(←^{117}Cd)



γ ray abundance

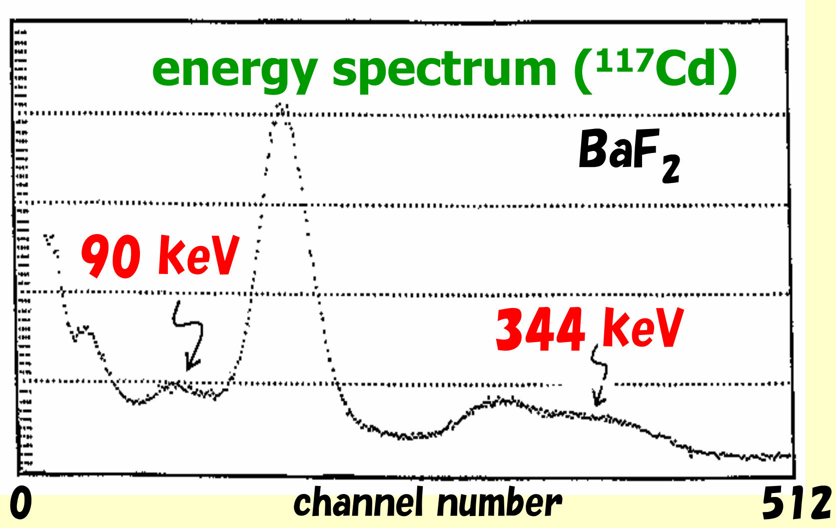
90 keV	3%
345 keV	18%

^{111}Cd(←^{111m}Cd)

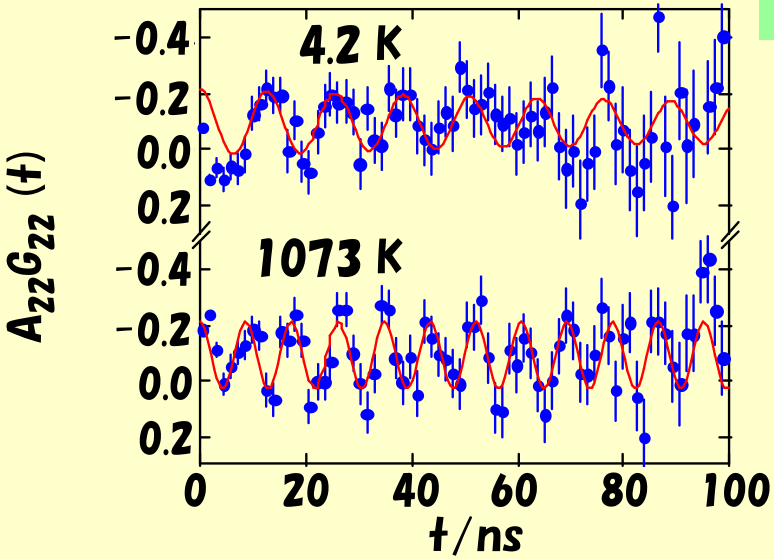


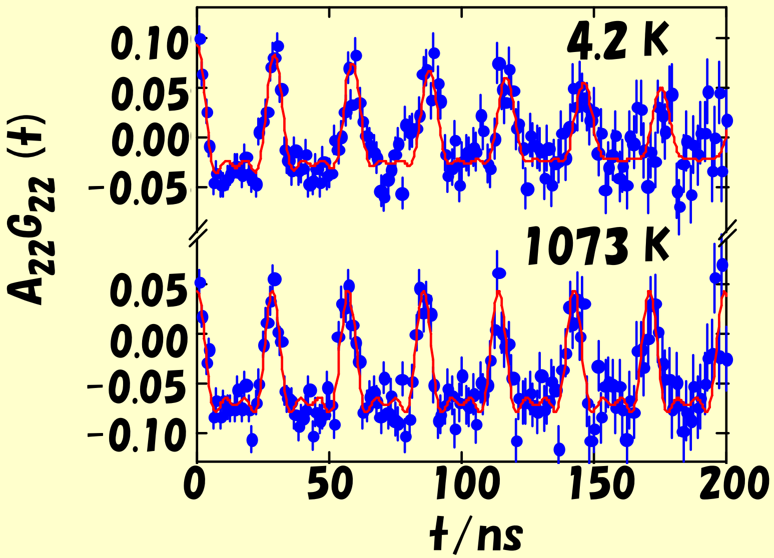
γ ray abundance

151 keV	32%
245 keV	94%



TDPAC spectra for $LiTaO_3$





axial symmetric electric field gradient

^{117}In ($I = 3/2$)

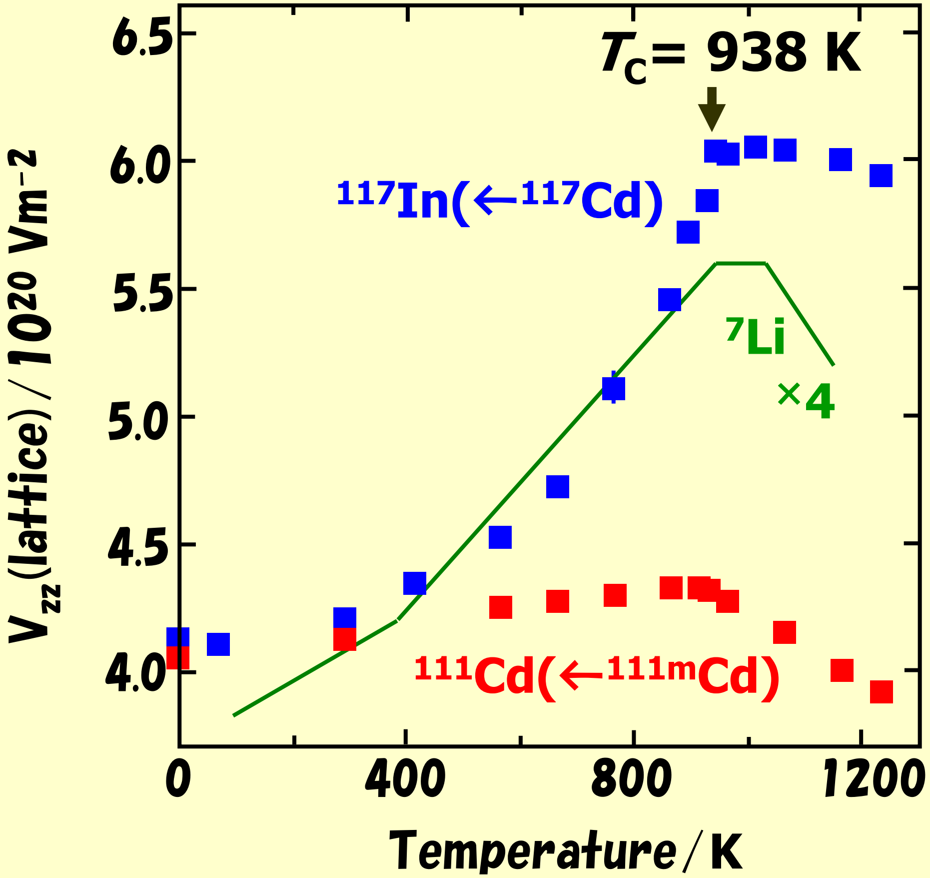
$$A_{22}G_{22}(t) = \frac{1}{5}\{1 + 4\cos(6\omega_Q t)\}$$

$$\omega_Q = \frac{eQV_{zz}}{4I(2I-1)\hbar}$$

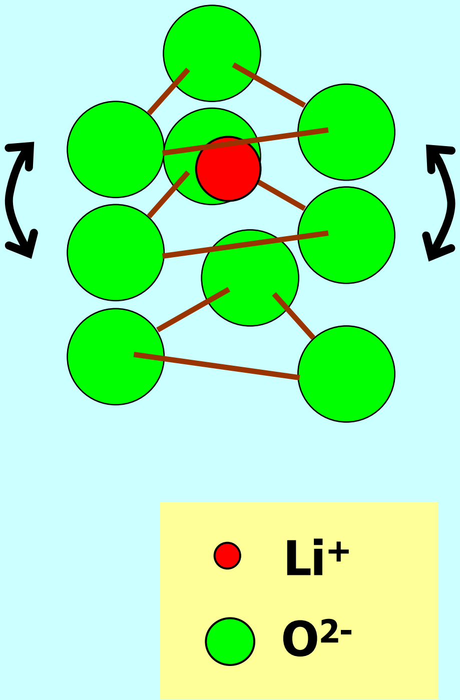
^{111}Cd ($I = 5/2$)

$$A_{22}G_{22}(t) = \frac{1}{5}\{1 + \frac{13}{7}\cos(6\omega_Q t) + \frac{10}{7}\cos(12\omega_Q t) + \frac{5}{7}\cos(18\omega_Q t)\}$$

Temperature dependences of V_{zz} (lattice) at ^{117}In, ^{111}Cd, and ^{7}Li in LiTaO$_3$

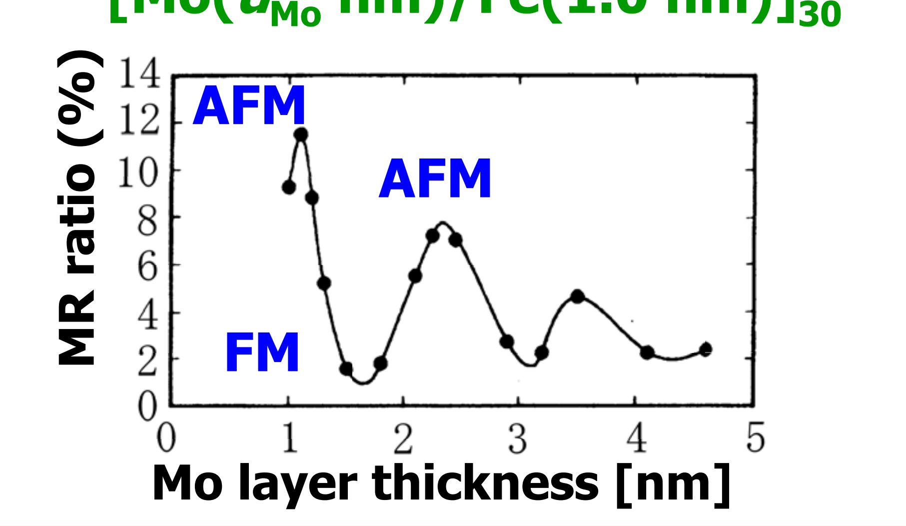


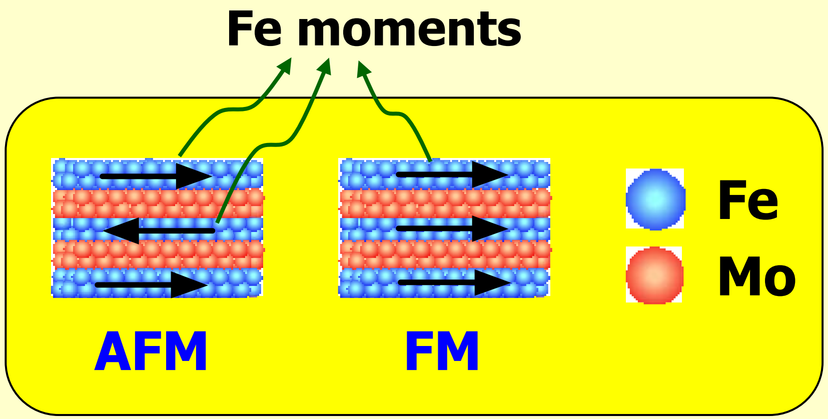
Paraelectric phase



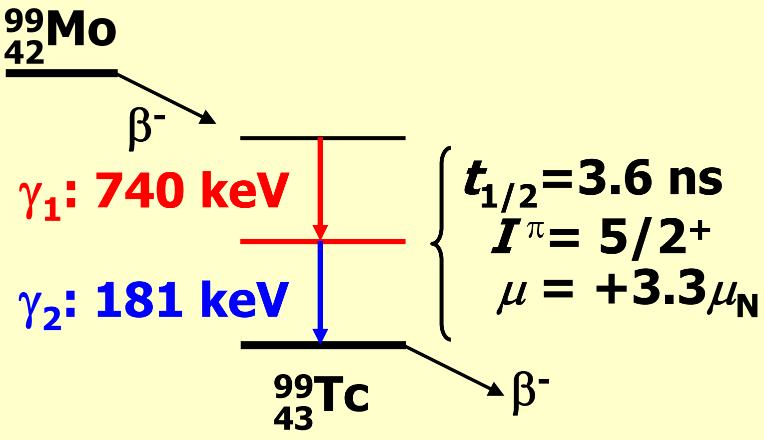
Local magnetic fields in the Mo layer of Mo/Fe multilayer

$[\mathrm{Mo}(d_{\mathrm{Mo}}\ \mathrm{nm})/\mathrm{Fe}(1.0\ \mathrm{nm})]_{30}$

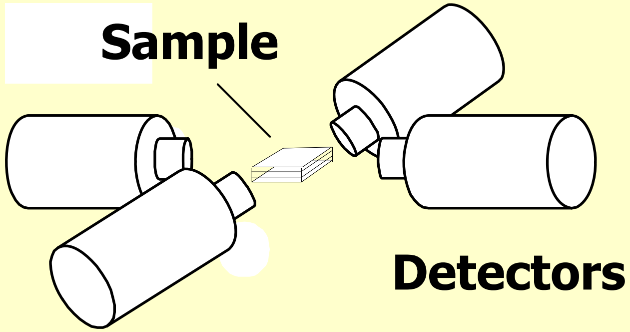


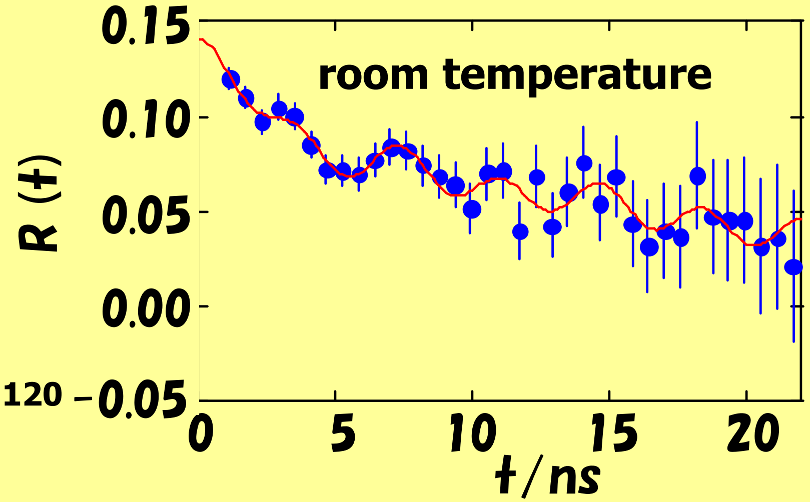


TDPAC probe: ^{99}Mo → ^{99}Tc



TDPAC spectra of ^{99}Tc(←^{99}Mo)

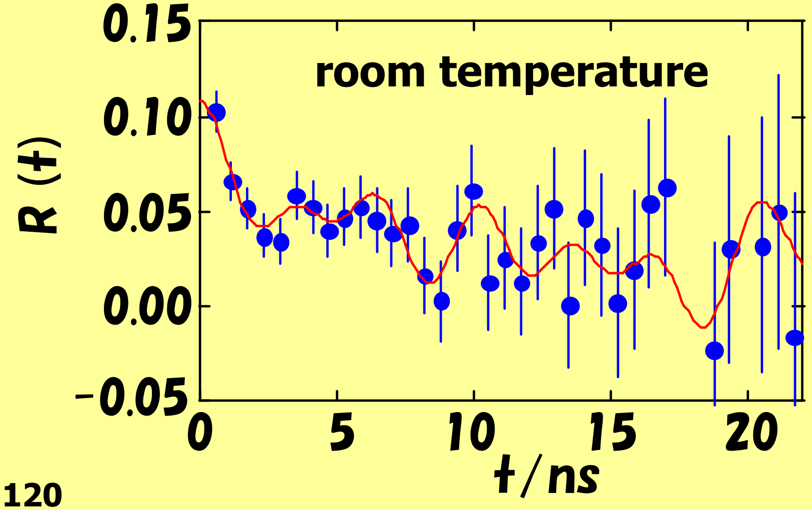




Fe(10nm)[Mo(**1.1**nm)/Fe(2.0nm)]$_{120}$

i =	1	2	3	4
B_i (T)	13.6	7.6	5.5	1.3
fraction	0.2	0.2	0.2	0.4

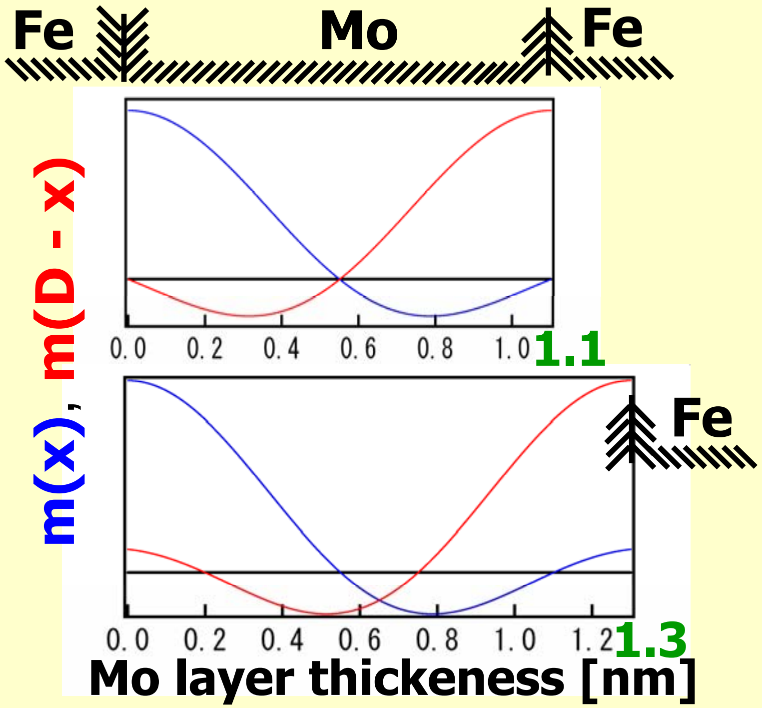
B_i (T)	14.9	9.6	7.0	1.4
fraction	0.2	0.2	0.4	0.2



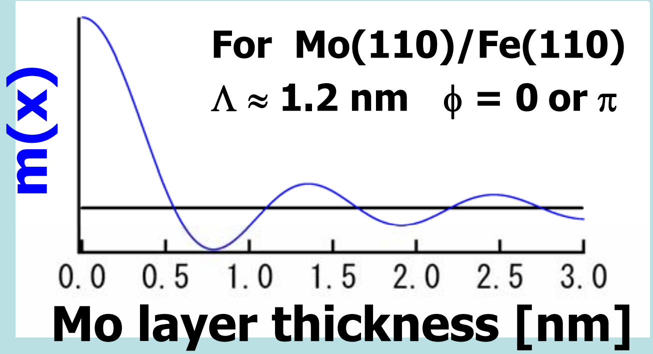
Fe(10nm)[Mo(**1.3**nm)/Fe(2.0nm)]$_{120}$

Also, Mo(0.4nm), Mo(0.7nm), Mo(0.9nm). All are FM systems.

superposed magnetic profile *M*(*x*)



magnetic profile *m*(*x*) from one of the two Mo/Fe interfaces due to the spin polarization of the conduction electrons



$$M(x) = m(x) + m(D - x),$$

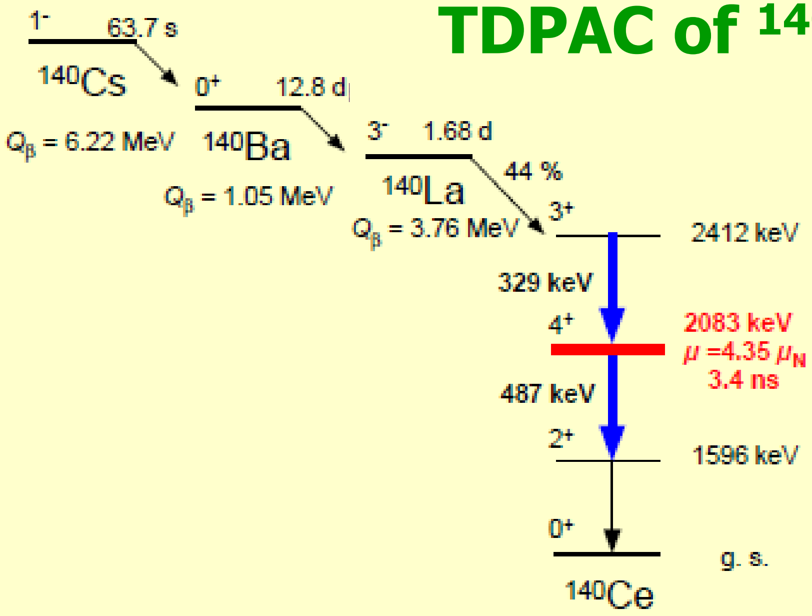
$$m(x) \propto x^{-\alpha} \sin(2\pi x/\Lambda + \phi)$$

x : distance from the interface;
D : Mo layer thickness; ϕ : initial phase
α : parameter; Λ : oscillation period of interlayer coupling (AFM→FM→AFM)

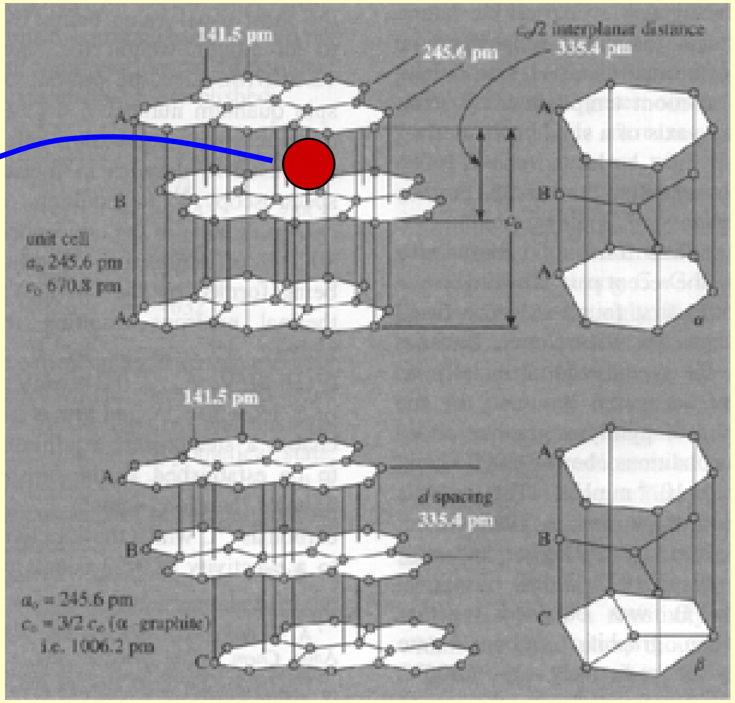
Using the TDPAC data, α was determined to be about 2. ➡ **RKKY interactions**

Hopping motion of Ce in graphite

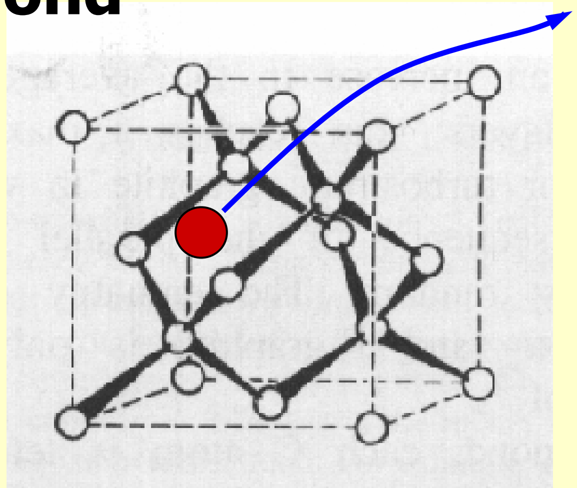
TDPAC of ^{140}Ce(←^{140}La←^{140}Ba←^{140}Cs)



HOPG (Highly Oriented Pyrolytic Graphite)

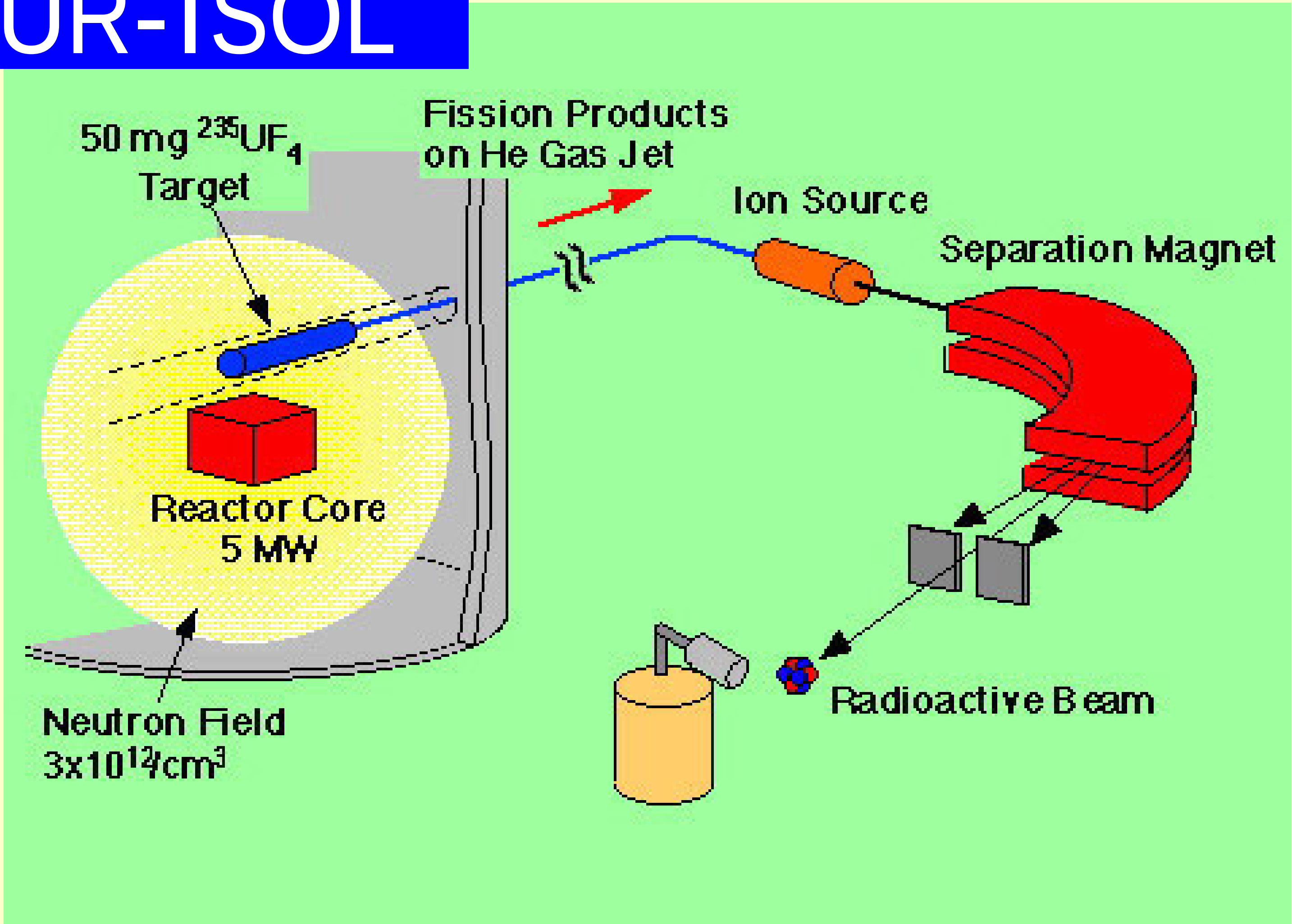


Diamond



^{140}Ce

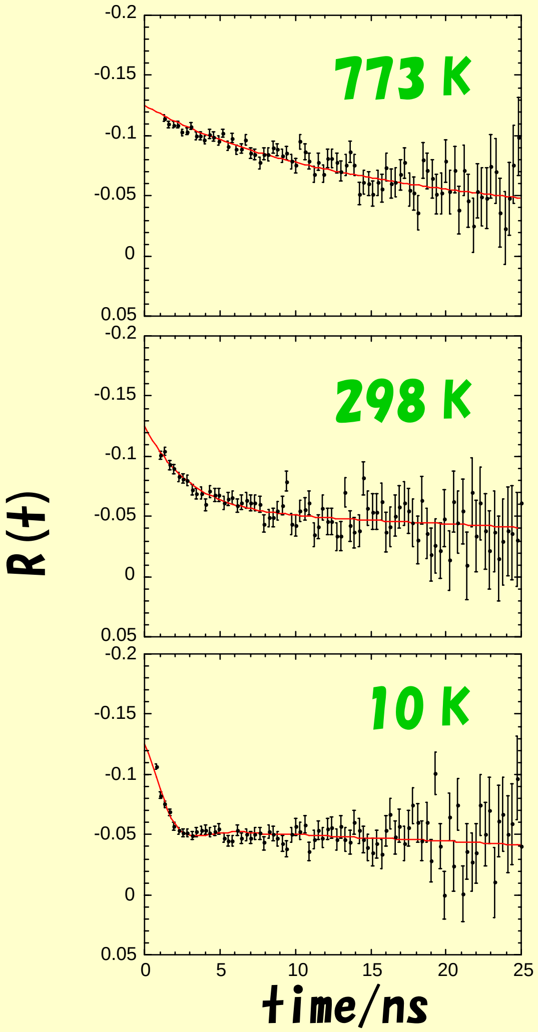
KUR-ISOL



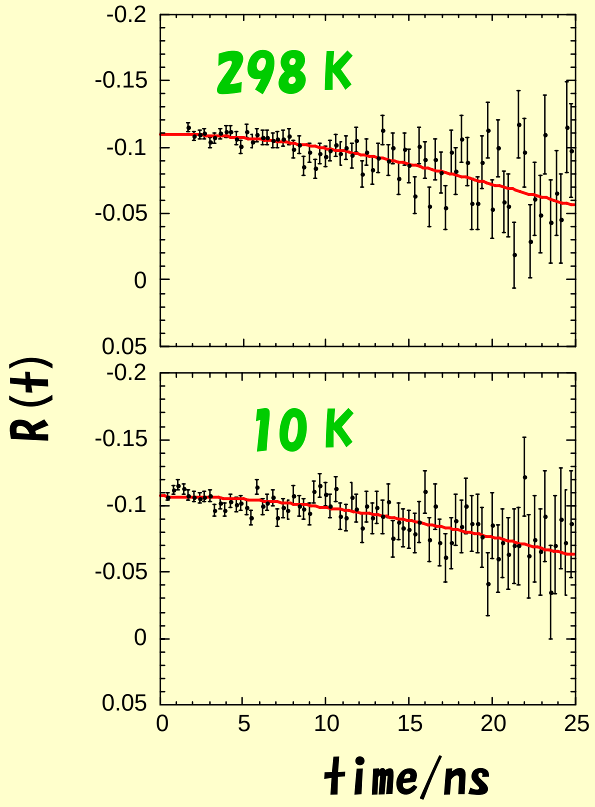
Fission fragments produced in the target chamber are transported to the ion source by gas-jet composed of He-N_2 mixed gas and PbI_2 aerosol.

TDPAC of ^{140}Ce

exponential behavior



cosine behavior



Graphite（HOPG)

dynamic perturbation（Ce^{3+})

Diamond

static perturbation（Ce^{4+})

Hyperfine fields in mavicyanin

Mavicyanin: a protein having a molecular weight of about 10,000, contained in zucchini

Cu atom at the active site

TDPAC of ^{117}In($\leftarrow$^{117}Cd) substituted for Cu