Perturbed Angular Correlations

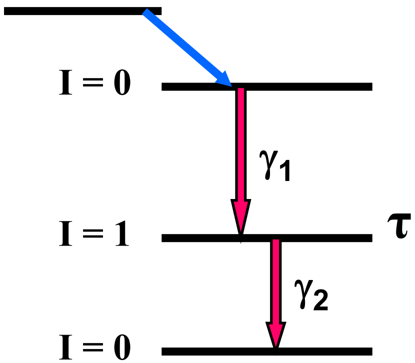
in nuclear physics and material science

- The phenomenon of γ-γ angular correlations
- Time Differential (TDPAC) and Time Integral (IPAC) technique
- Perturbation of Angular Correlations by Hyperfine Interactions
- Related techniques: TDPAD and SRPAC
- Applications
- Analysis of strengths and weaknesses
- Comparison with other hyperfine interaction techniques

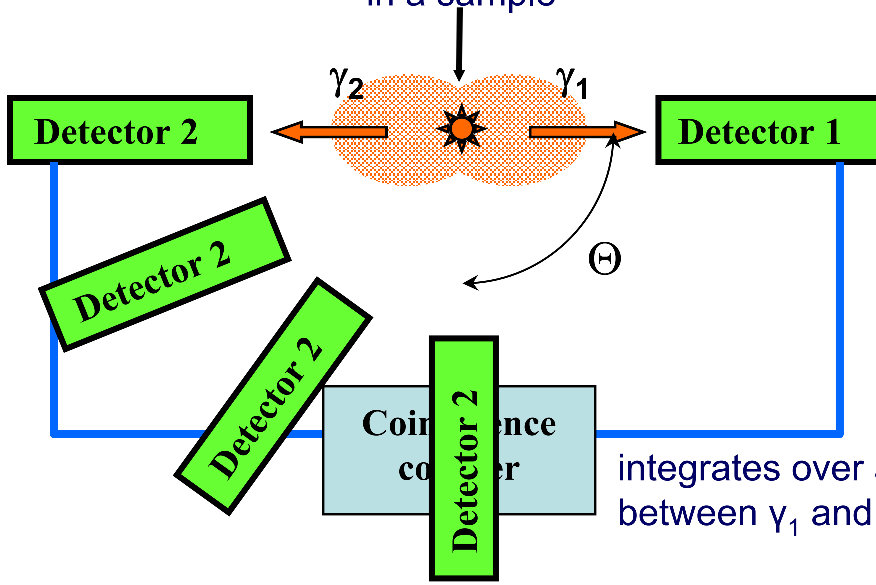
The phenomenon of unperturbed γ - γ angular correlations

The time-integral mode (IPAC)

Parent isotope

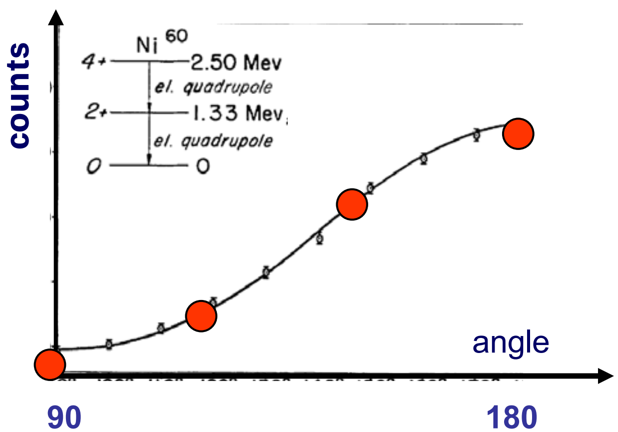


About 10^{11} parent isotopes in a sample



integrates over all times between γ_1 and γ_2

For spin sequence 0-1-0:
counts ≈ $\cos^2\Theta$



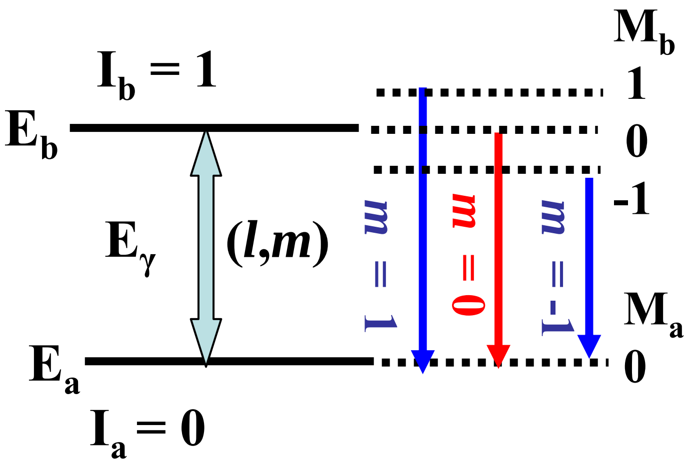
R.M. Steffen, Adv. In Phys. 4 (1955) 293

$$W(\Theta, t = 0 \rightarrow \infty) = 1 + A_{kk}P_k(\cos\Theta)\ ,\ k = 2,4$$

A_{kk} = angular correlation coefficients

depend on spins, parities and multipole order of the cascade

Gamma decay of excited nuclear states



Conservation of angular momentum:

$|I_a - I_b| \leq l \leq |I_a + I_b|$, $m = M_a - M_b$

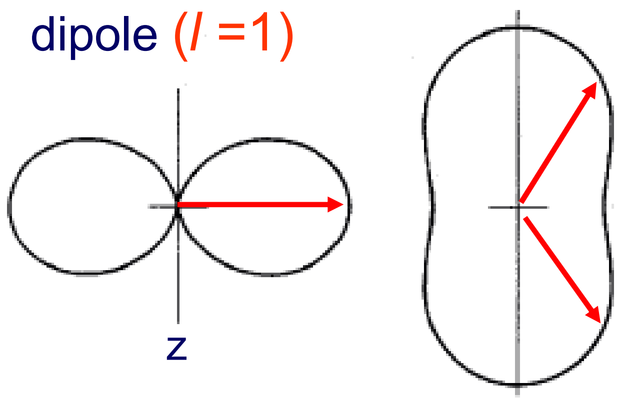
l : multipole order

$l = 1$ dipole radiation

$l = 2$ quadrupole radiation

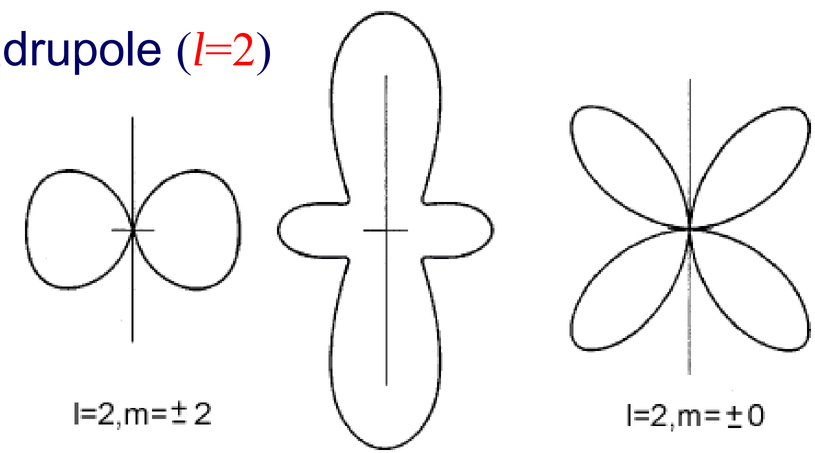
The angular characteristics $I_{l,m}(\Theta)$ of the gamma-radiation depends on (l, m)

dipole (l =1)



l=1,m=0 l=1,m=±1

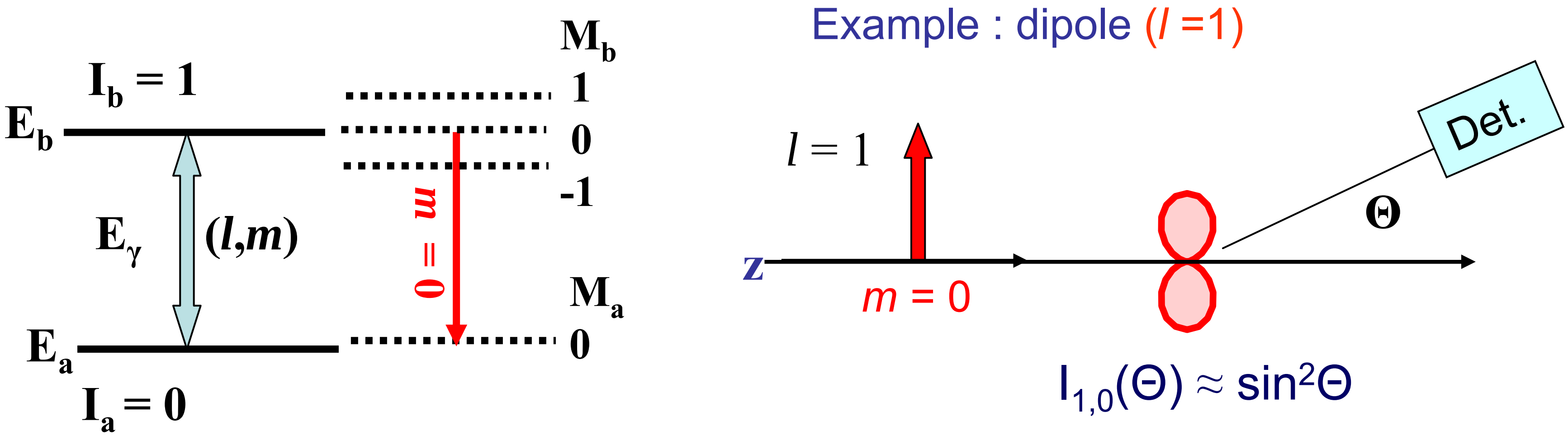
quadrupole (l=2)

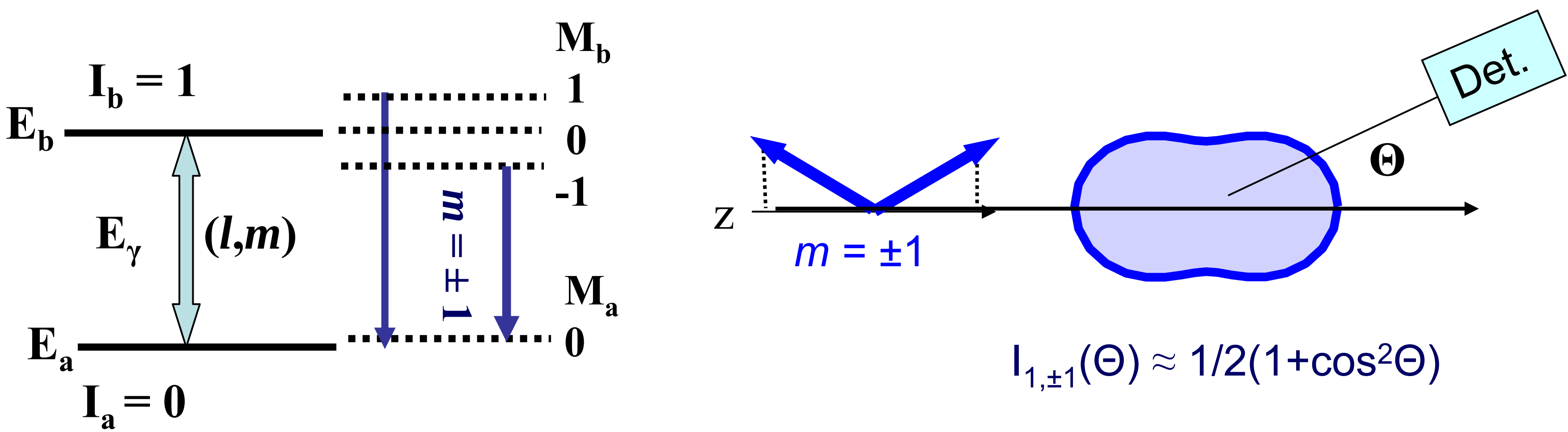


l=2,m=±2 l=2,m=±1 l=2,m=±0

Nuclear physics : Dermination of the multipole order and spins by measurement of $I_{l,m}(\Theta)$

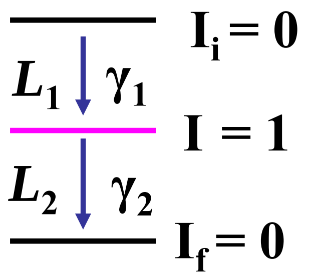
Gamma emission from aligned nuclear states





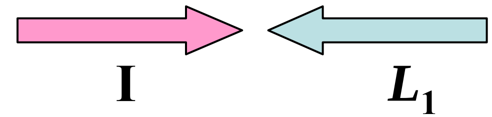
The radiation distribution informs on the spin orientation

Angular Correlations – Theoretical background

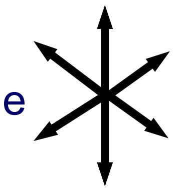


$||I_i - I|| \leq L_1 \leq ||I_i + I||$

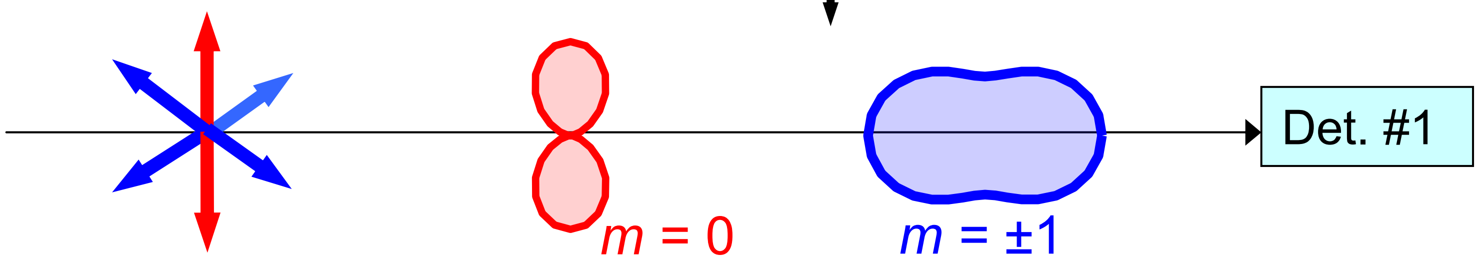
The first γ- transition with



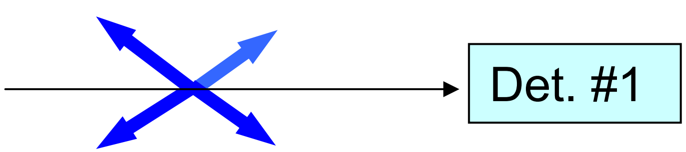
Radiation emitted by an ensemble

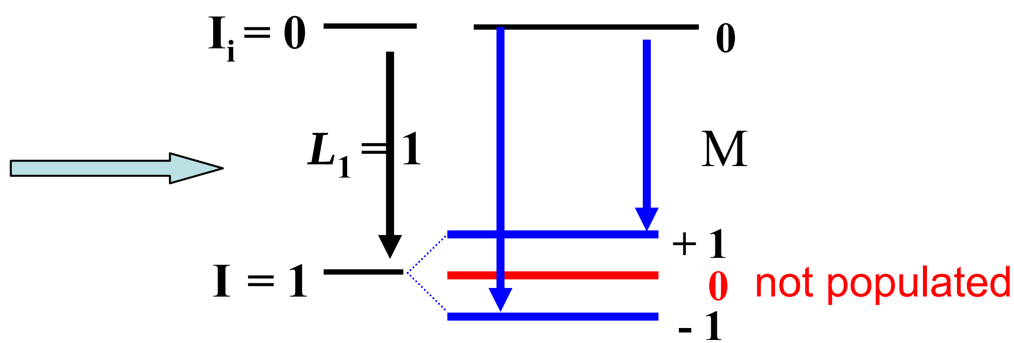


of randomly oriented nuclear spins:



Nuclear spins seen by Det. #1



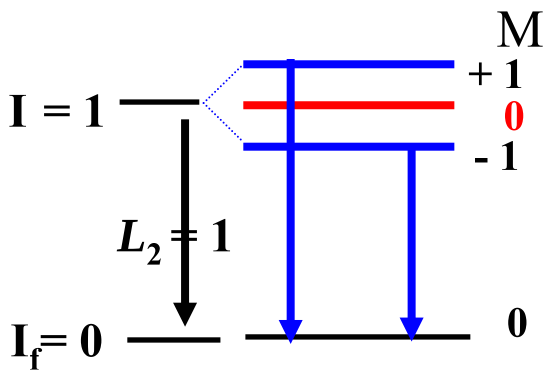


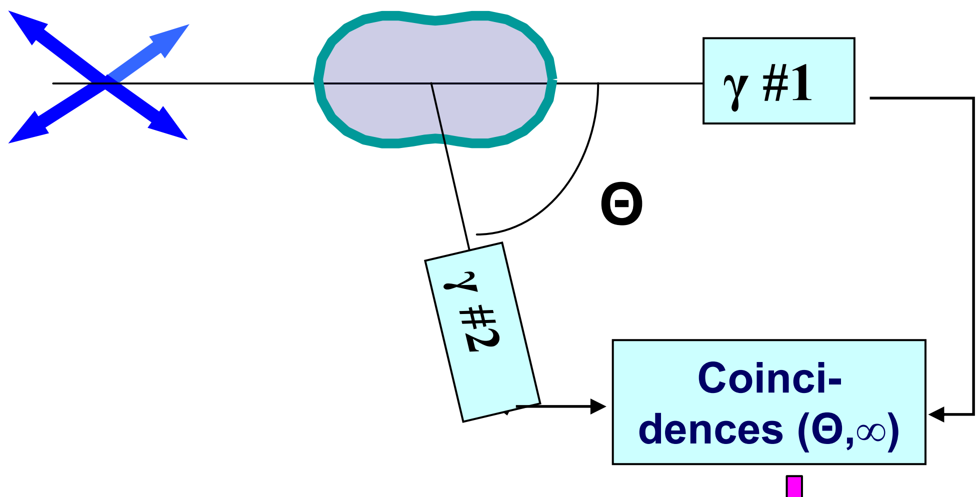
Observation of γ_1 = selection of a subgroup of spin orientations

Spin alignment !

The second γ- transition of the γγ –cascade

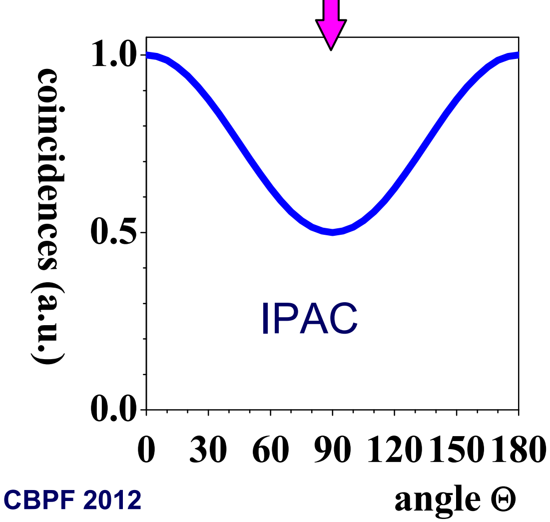
Emission from an aligned spin ensemble





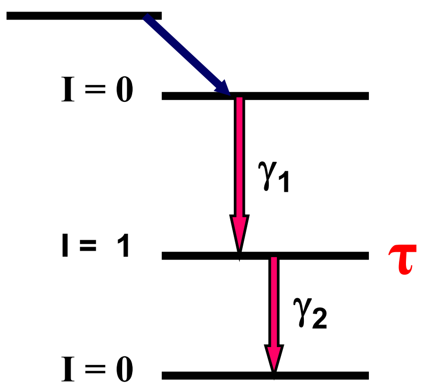
By observation of γ_1 we select
a specific orientation of the spin I.

The direction of γ_2 informs us
on changes in the spin orientation
while the nucleus is in the intermediate state.

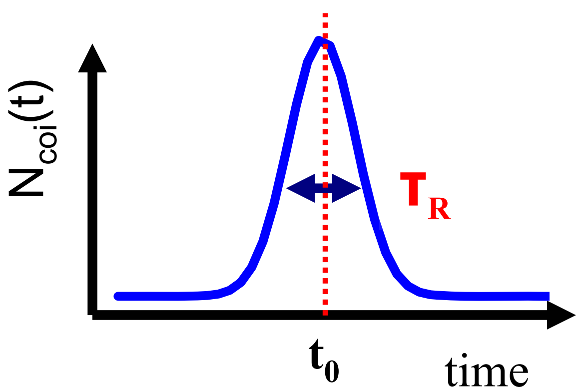


OBSERVATION MODES of Angular Correlations

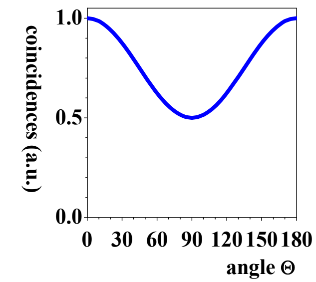
Parent isotope



Time resolution



Time integral mode: IPAC



Half-width $\tau_R \geq$ life time τ

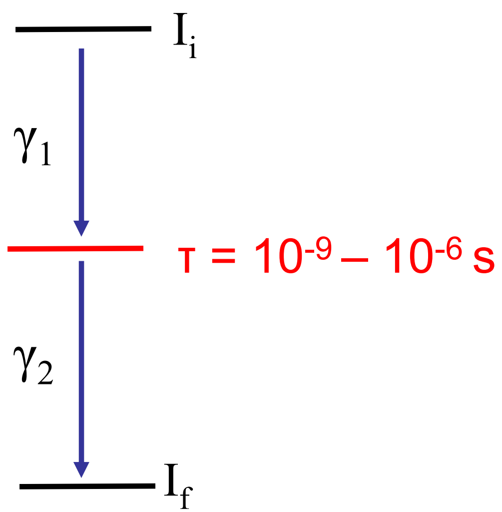
- Integration over all times of the intermediate state of the cascad
- Time-integrated observation of hyperfine interactíons

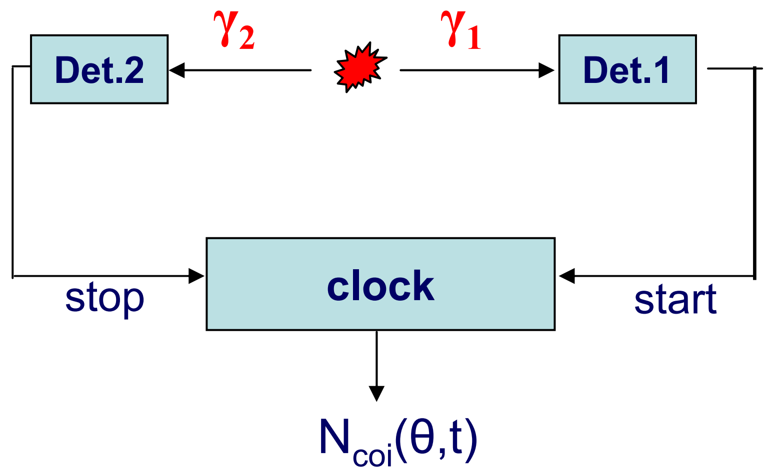
Time differential mode: TDPAC

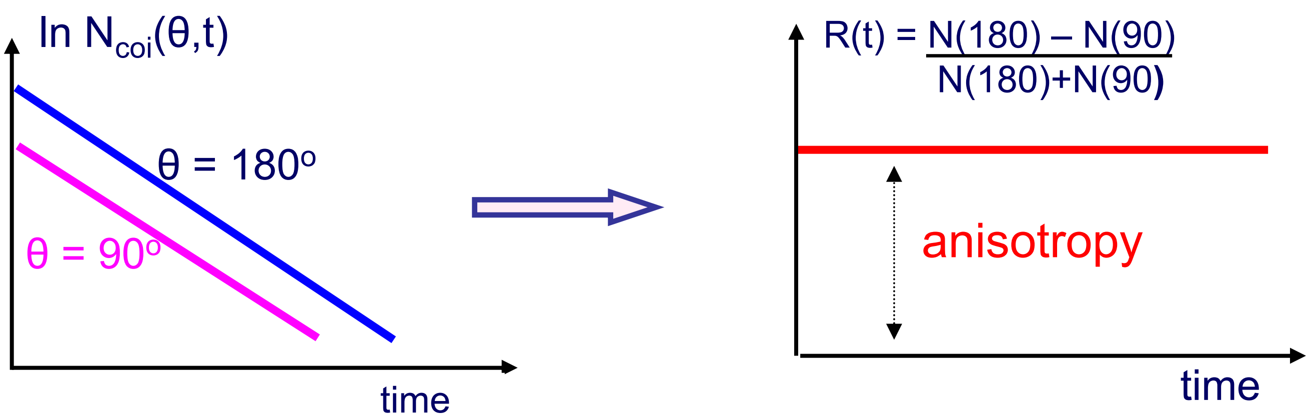
Half-width $\tau_R \leq$ life time τ

- Measurement of the anisotropy as a function of the time the nucleus has spent in the inter-mediate state of the cascade
- Time resolved observation of hyperfine frequencies $\nu_L \leq 1/\Delta\tau$

The time-differential mode (TDPAC) of unperturbed γ - γ angular correlations





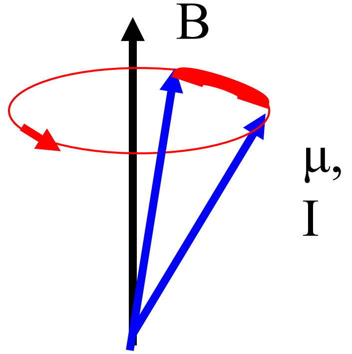


The perturbation of γγ angular correlations by hyperfine interactions

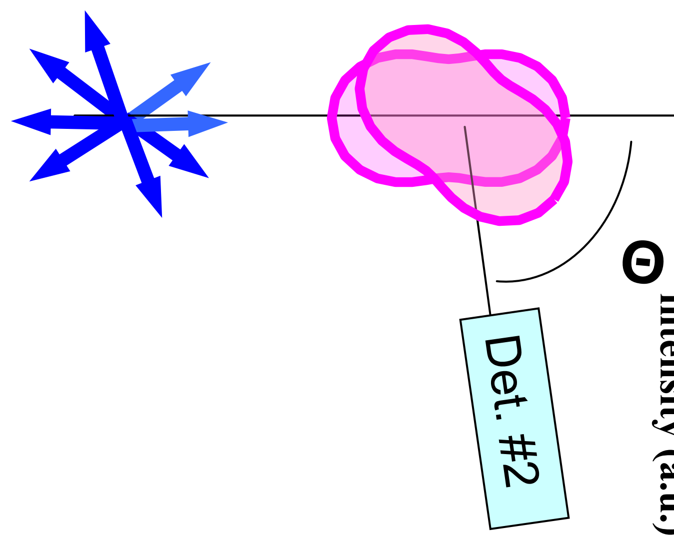
Basic aspects:

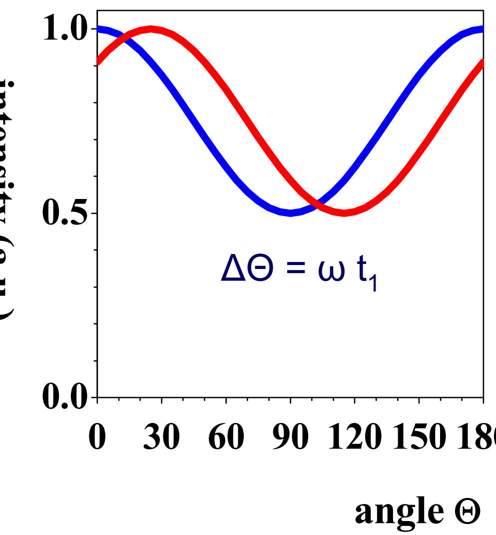
(i) Detection of γ_1 = selection of a subgroup of spin orientations
→ anisotropic intensity distribution of γ_2

(ii) Hyperfine interaction → Larmor precession of spins
= Precession of the intensity distribution of γ_2



$$\omega_L \propto (\vec{\mu} \cdot \vec{B})$$



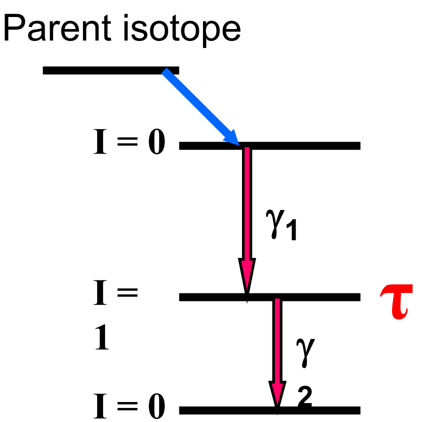


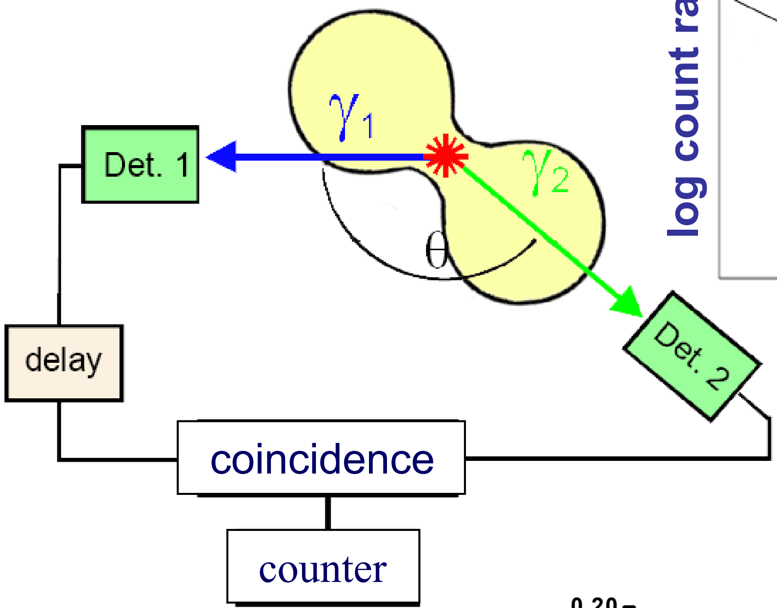
(iii) Detection of γ_2 = spin orientation at time t

The perturbation of γγ angular correlations by hyperfine interactions

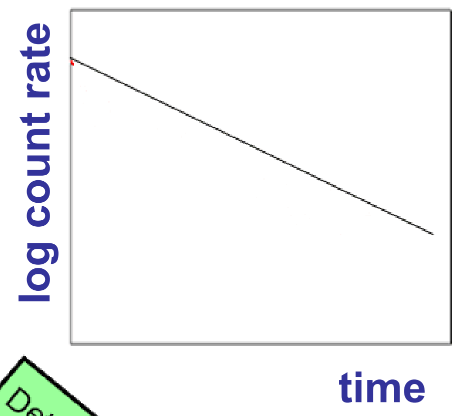
The time differential mode

Example : Magnetic field B ⊥ detector plane



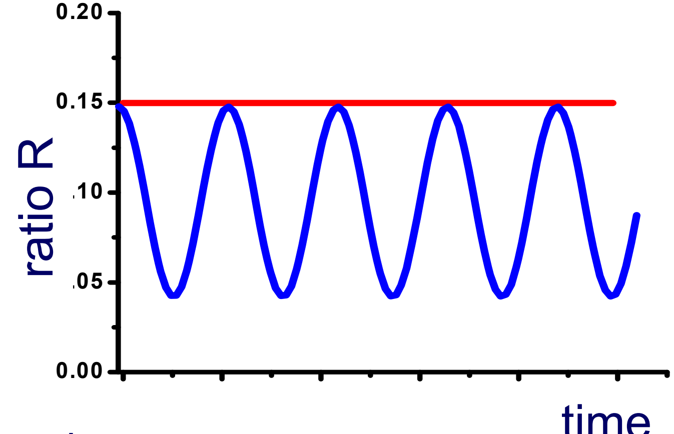


$N(\Theta,t) = N_0 \, e^{-\lambda t} \, W(\Theta,t)$



Elimination of the exponential decay

$$R(t) = \frac{N(180,t) - N(90,t)}{N(180,t) + N(90,t)}$$



Time differential Perturbed Angular Correlations – An Early Example (1965)

^{100}Rh in an external magnetic field of 0.22 T

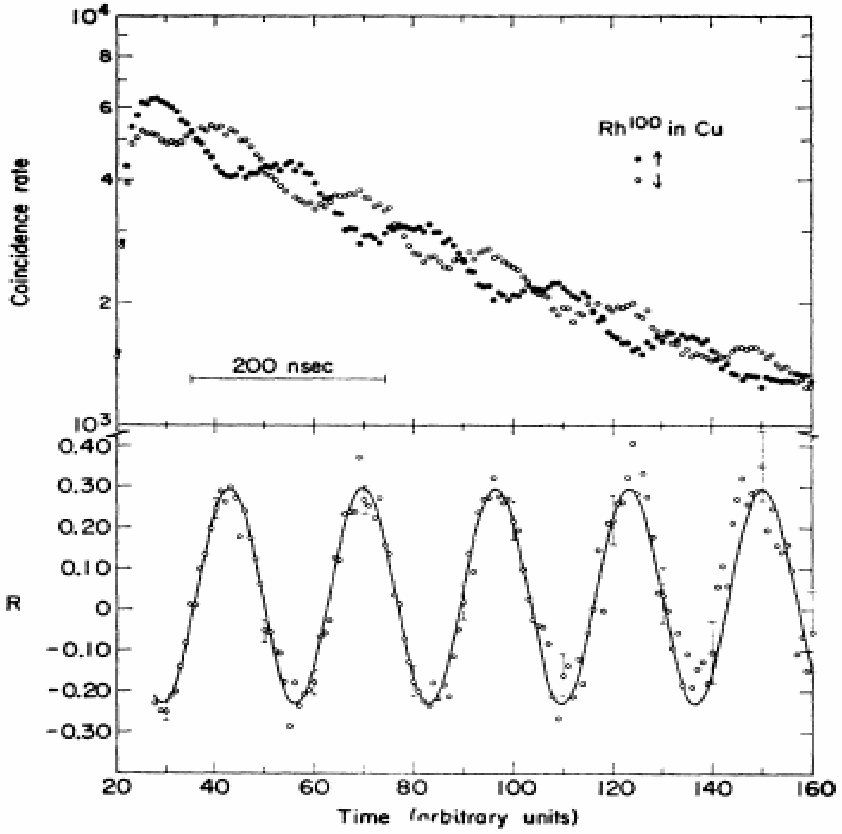


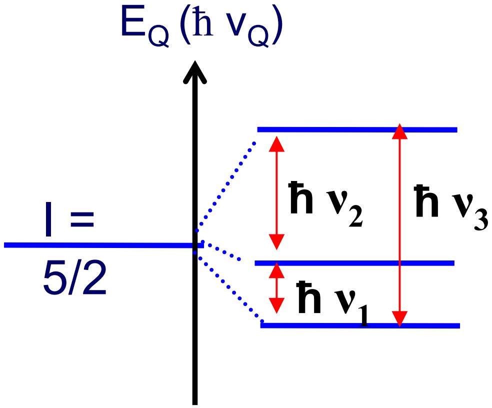
Fig. 4. Time-differential g-factor measurement with a source of Rh^{100} in copper in an external field of ± 2.22 kG. In the upper part of the figure the raw data of run No. 1 in Table II are shown without any background correction. In the lower part the corresponding ratios R_i are displayed. The full line represents the weighted least-squares fit to the data.

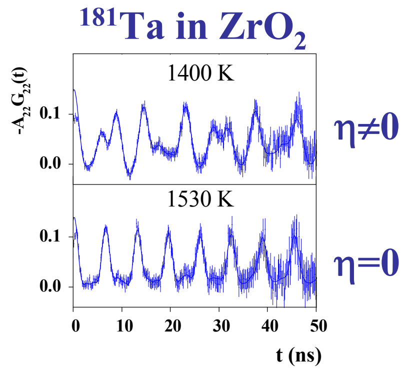
The perturbation of angular correlations by static hyperfine interactions

$$W(\theta,t) = \sum_{k=par} A_{kk} G_{kk}(t) P_k(\cos\theta)$$

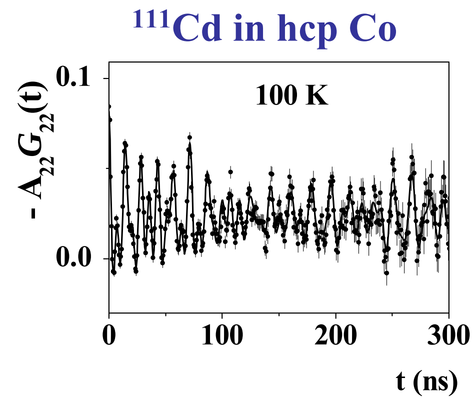
$G_{kk}(t)$ = perturbation factor

Electric quadrupole interaction

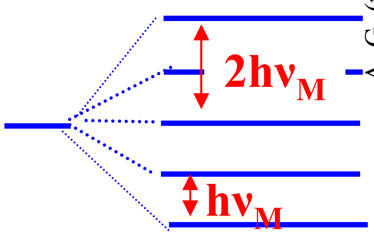


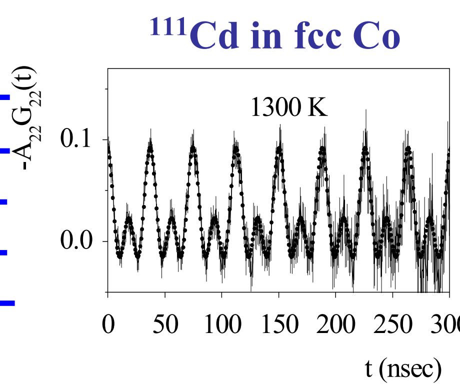


Combined MHFI +QI

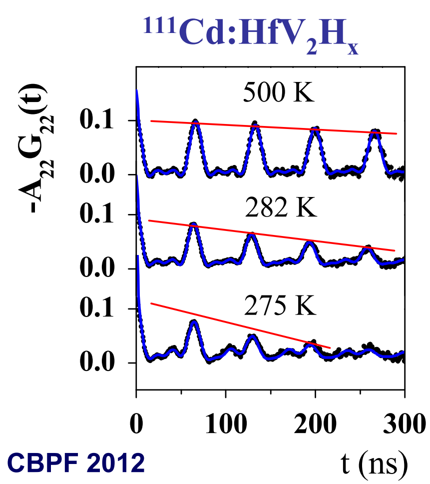


Magnetic dipole interaction



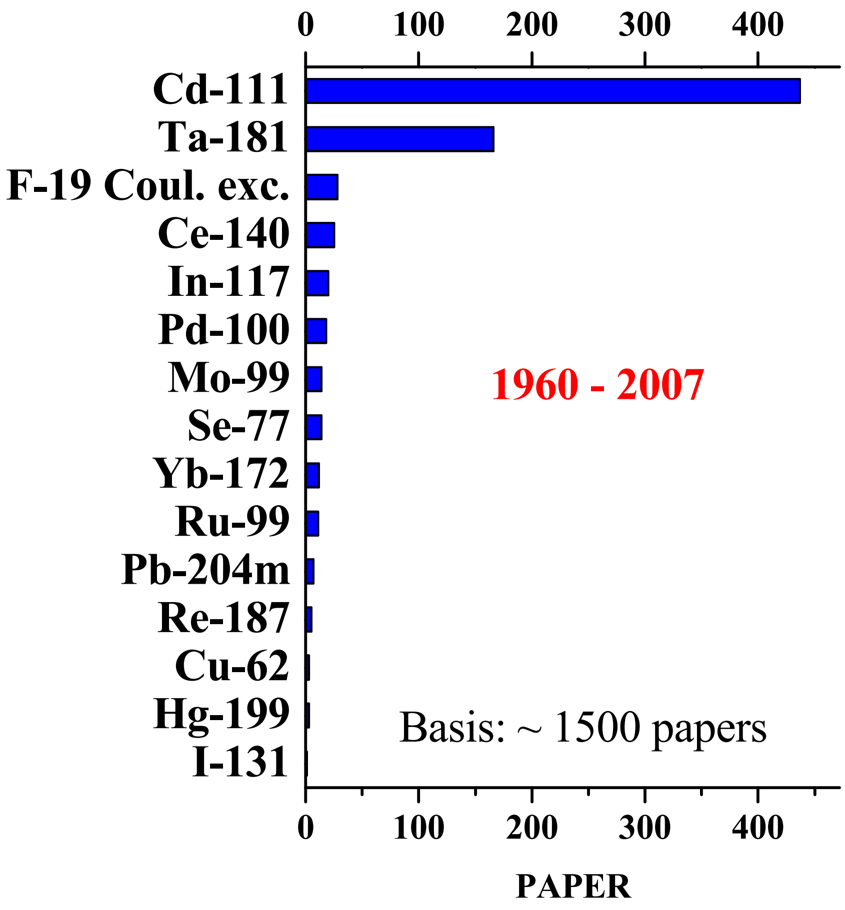


Dynamic QI



γγ-cascades for TDPAC measurements - required properties

- Mean life time τ of the intermediate state: 10^{-9} s $\leq \tau \leq 10^{-6}$ s
- Spin I of the intermediate state: $I \geq 1$
- Anisotropy: $A_{22} \geq 0.05$
- Intensity of the cascade
- Half- life of the mother isotope: $T_{1/2} \geq 1$ h (Laboratory experiments)
- Coincidence experiment: data accumulation:10 min to a few days

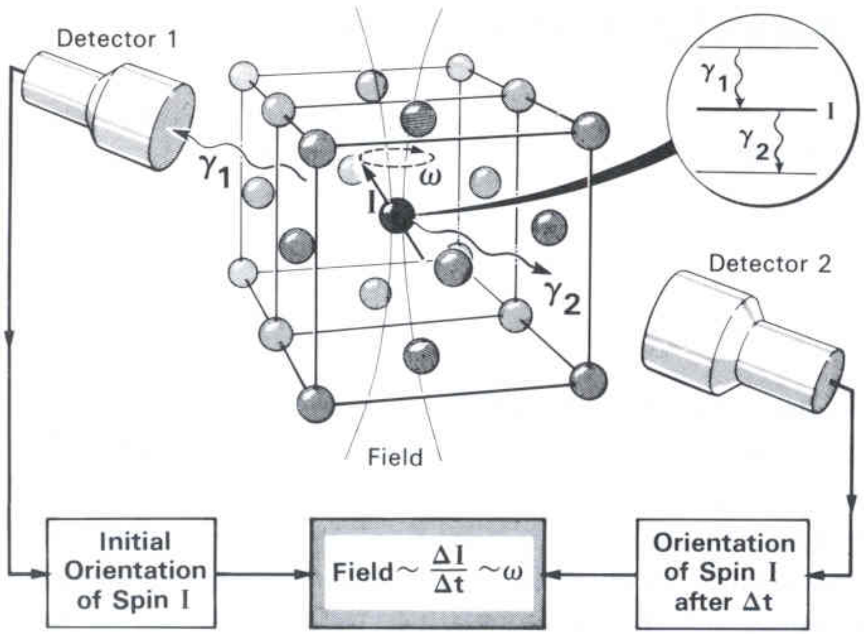


Number of suitable isotopes for laboratory experiments: ≤ 15

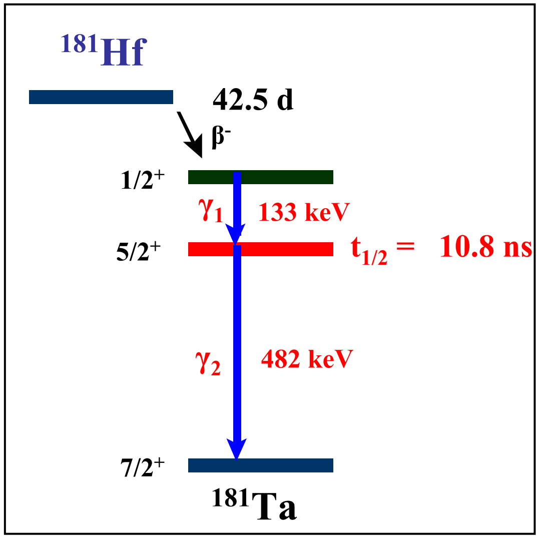
TDPAC spectrometer – basic aspects

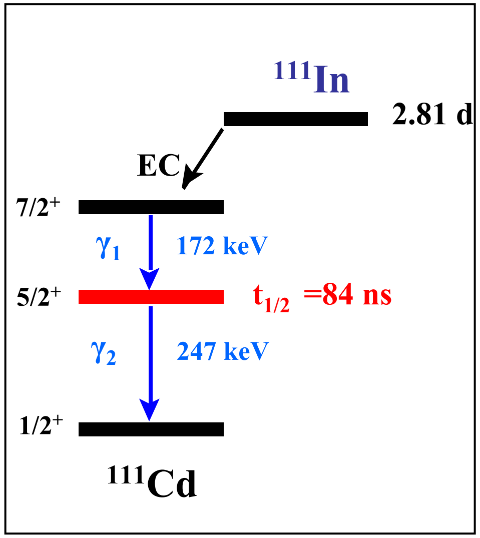
Requirements:

high statistics
high detection efficiency
energy resolution
time resolution



popular isotopes



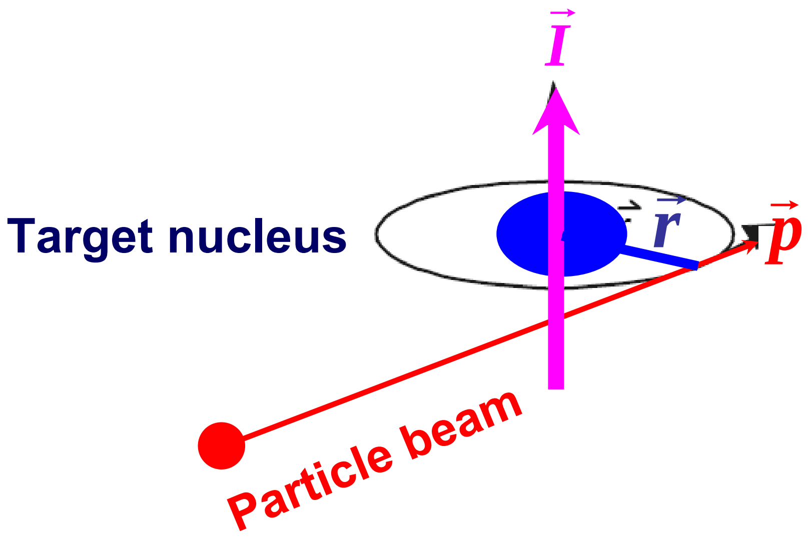


Present standard:

Multi-detector arrays: up to 6 detectors
Fast photomultiplier coupled to BaF_2 scintillators
Time resolution: ~ 100 ps FWHM at ^{60}Co energies

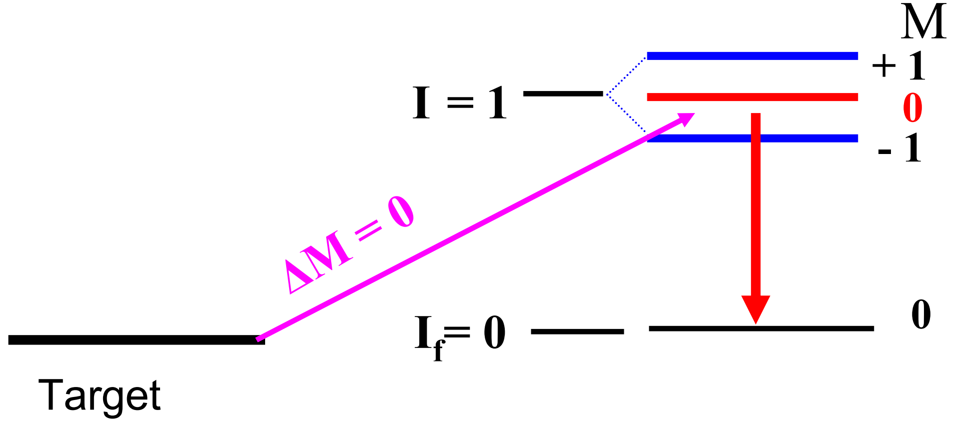
Perturbed angular distribution (TDPAD)

Production of spin alignment by nuclear reaction



In nuclear reaction the angular momentum transferred to the target nucleus is preferentially perpendicular to the beam axis

Nuclear reactions therefore favour $m = 0$ transitions, producing aligned nuclear states



The subsequent gamma-emission is anisotropic

Synchrotron radiation PAC (SRPAC)

Production of spin alignment by synchroton radiation

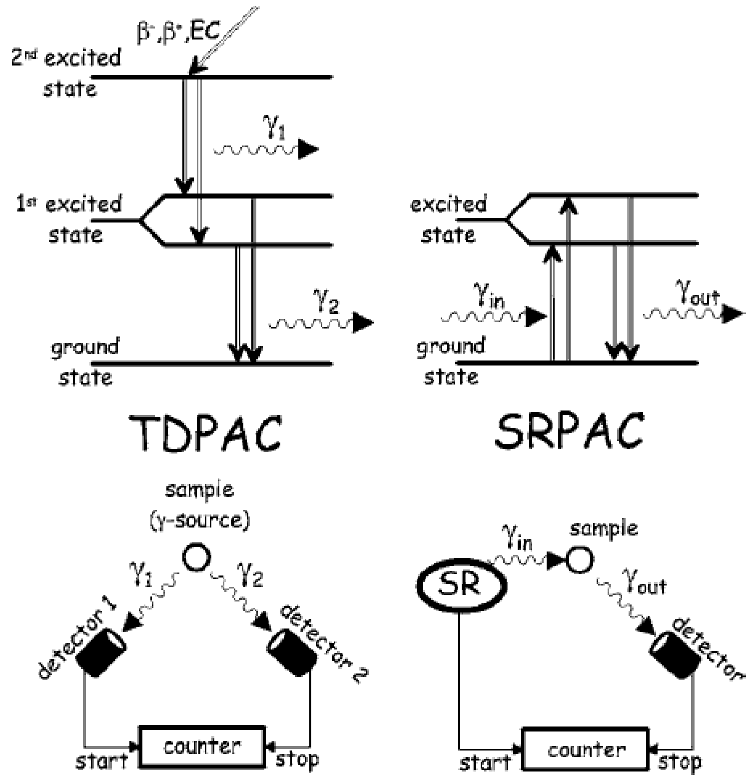


FIG. 1. Schemes of the principle and of the experimental setup for TDPAC (left side) and SRPAC (right side).

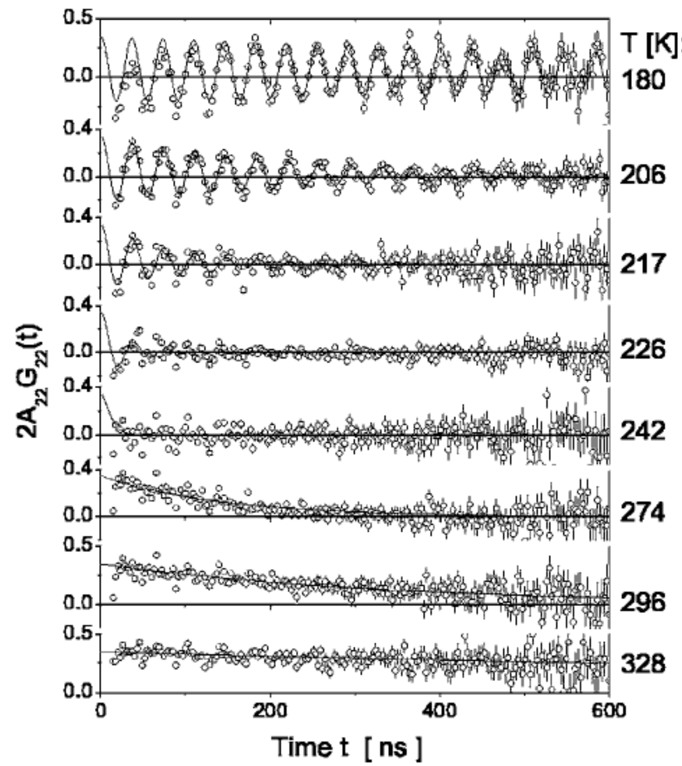


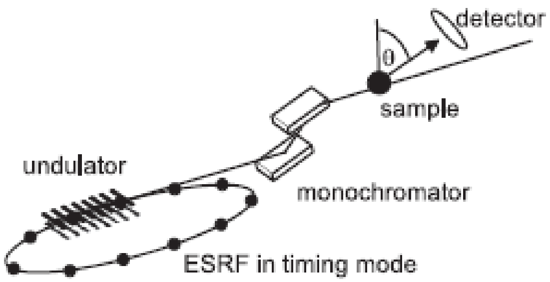
FIG. 8. Time evolution of the anisotropy $2A_{22}G_{22}(t)$ for several temperatures. The solid lines are fits by the theory given by Eqs. (3) and (10).

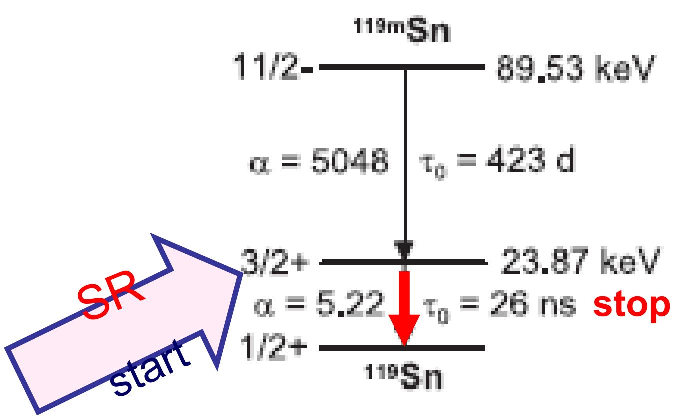
^{57}Fe in the molecular glass former dibutyl phthalate DBP doped with 5% mol of ferrocene FC enriched to 95% in ^{57}Fe.

Sergueev,et al, Phys. Rev. B 73 (2006) 024203

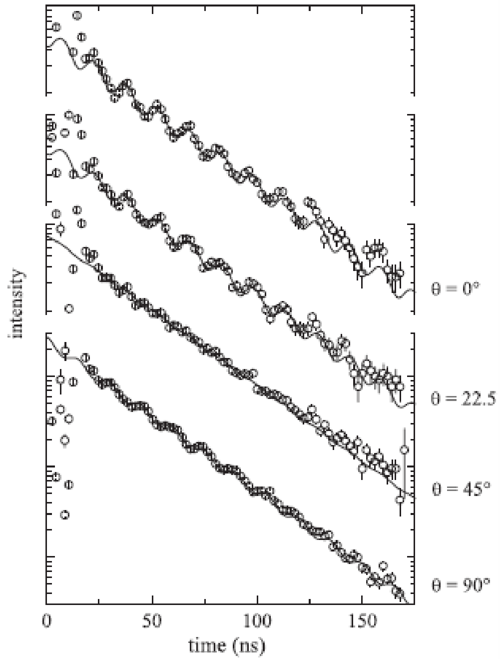
Synchrotron-radiation–based perturbed angular correlations from ^{119}Sn

C. Strohm, I. Serguეev and U. van Bürck, EPL, 81 (2008) 52001

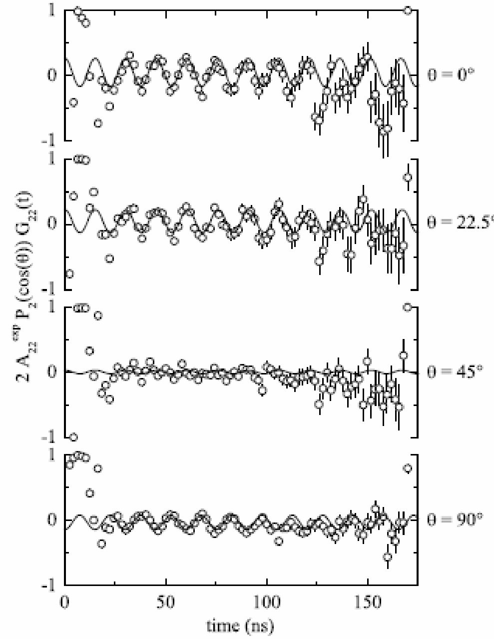




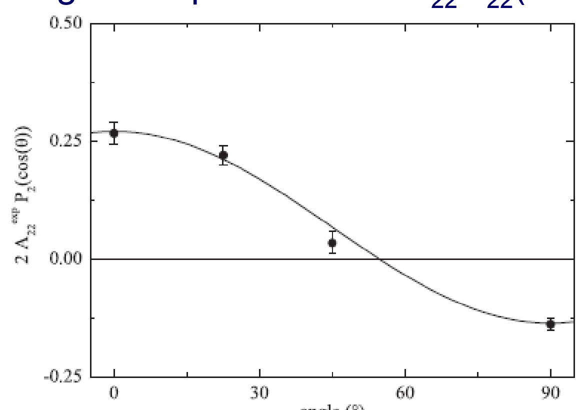
Counts *vs.* time



Anisotropy *vs.* time
(exp. decay eliminated)



Angular dependence of $A_{22}G_{22}(t=0)$



Main areas of PAC research

Nuclear physics

- Nuclear moments

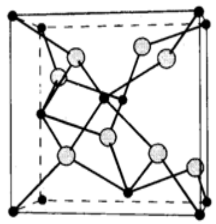
Condensed matter physics

- Magnetic properties of magnetically ordered systems
- Electric field gradients in non-cubic solids
- Dynamic processes: Atomic motion in solids, liquids , gases
- Phase transitions
- Defects in Solids
- Solid state reactions
- Biology; chemistry

Static QI in Solids: Phase identification and phase transformation

Example: ZrO_2

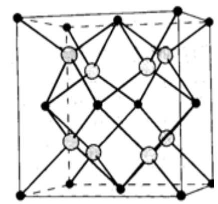
ZrO_2 phases



monoclinic

$\nu_q \neq 0$
$\eta \neq 0$

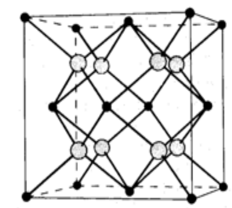
T ~ 1450 K →



tetragonal

$\nu_q \neq 0$
$\eta = 0$

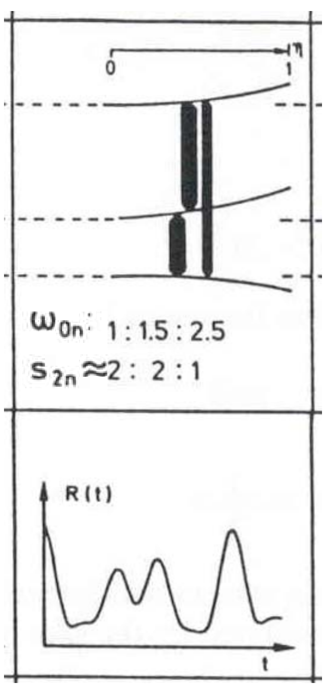
T ~ 2150 K →

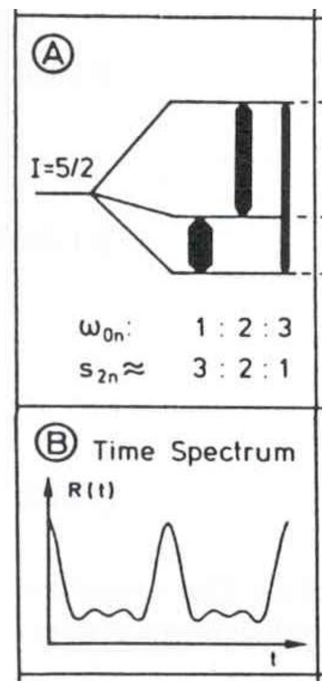


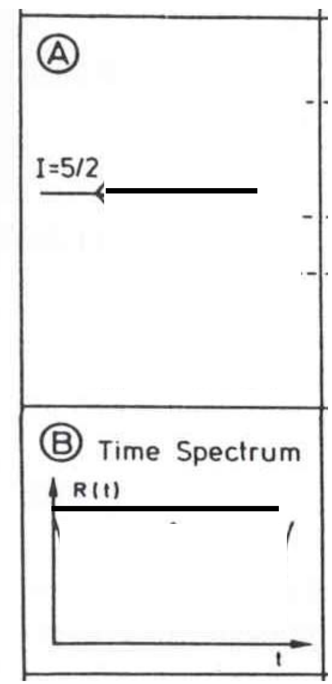
$\nu_q = 0$

cubic, EFG = 0

QI and PAC spectra

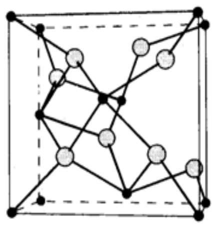






Phase identification and phase transformation by QI - Example: ZrO_2

ZrO_2 phases

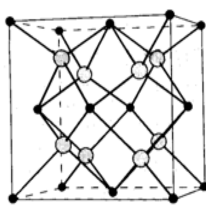


monoclinic

$\nu_q \neq 0$

$\eta \neq 0$

T ~1450 K

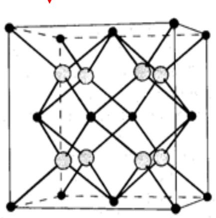


tetragonal

$\nu_q \neq 0$

$\eta = 0$

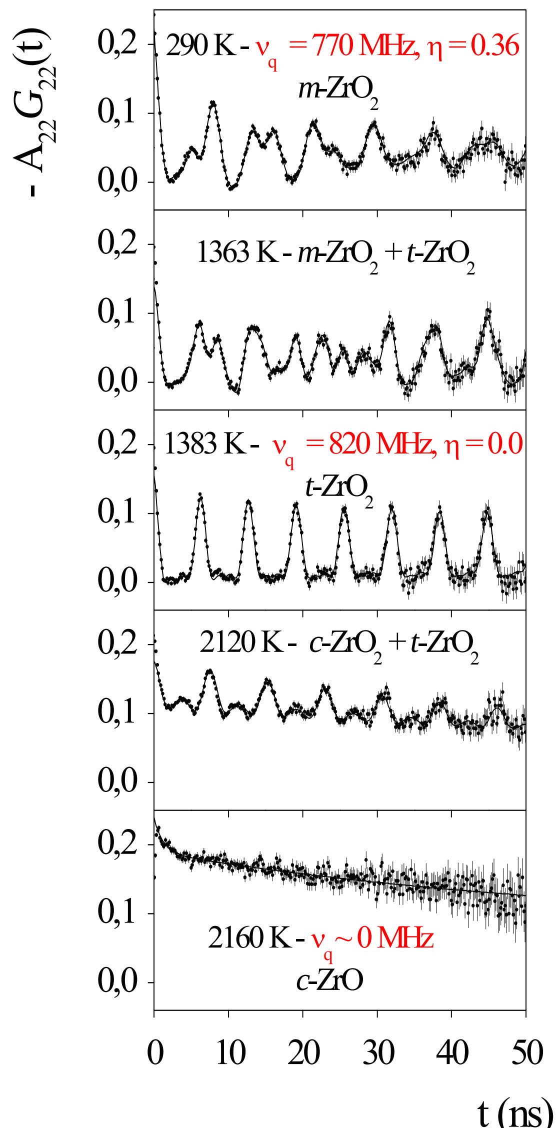
T ~2150 K



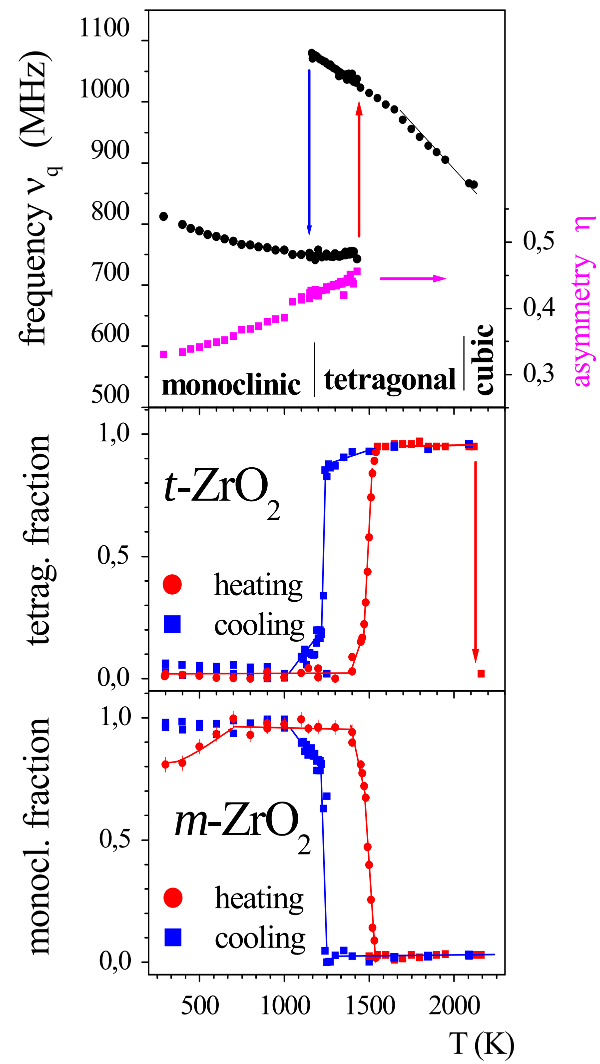
cubic, EFG = 0

$\nu_q = 0$

PAC spectra



Phase transformations

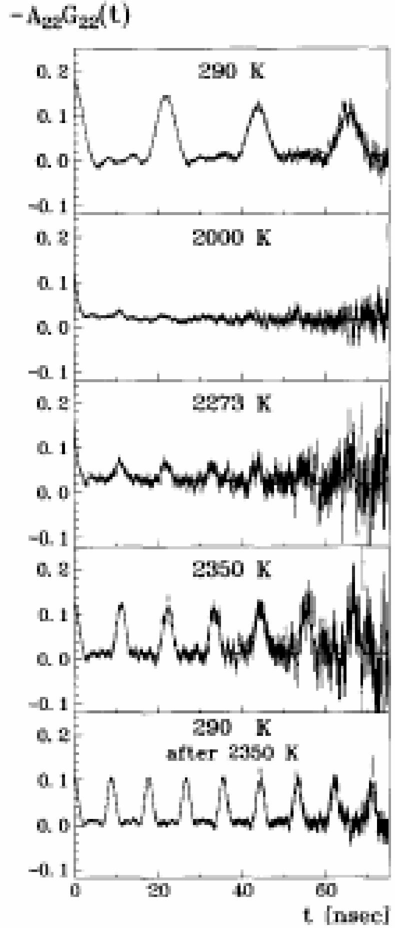


M. Forker, Nuclear Techniques in Solid State Resaerch , CBPF 2012

Static QI in solids:
Solid state reaction: Transformation of Zr metal into ZrB_2

$Zr + BN \xrightarrow{T} ZrB_2$

T ↓



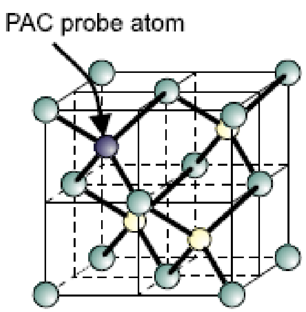
^{181}Hf:Zr metal

^{181}Hf:ZrB_2

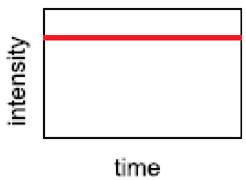
Fig. 4. PAC spectra of ^{181}Ta in Zr metal, heated to 2350 K in a boron nitride crucible.

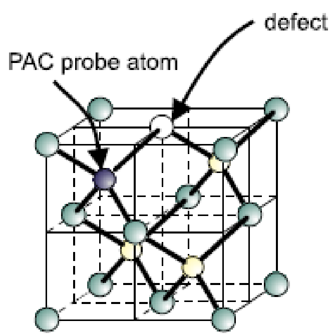
M. Forker, W. Herz, U. Hütten, M. Müller', R. Müßeler, J. Schmidberger, D. Simon, A. Weingarten and S.C. Bedi 2Nuclear Instruments and Methods m Physics Research A327 (1993) 456-462

Static QI in solids: Defects in metals and semiconductors studied by TDPAC

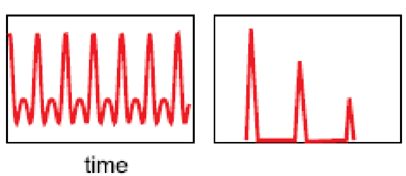


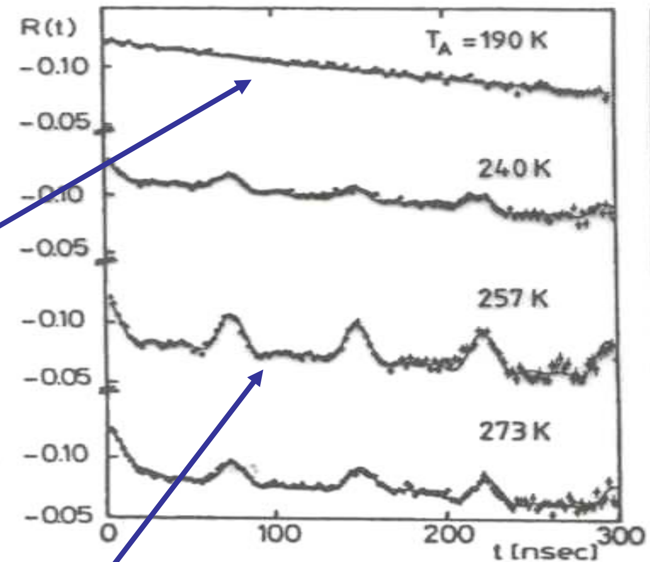
cubic unperturbed





nn defect sharp QI

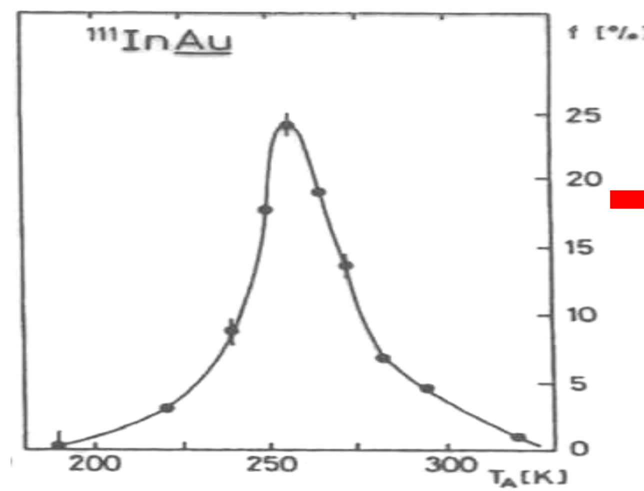




^{111}In in Au
e-irradiation
at low *T*

Subsequent
annealing

Probes with *nn* defect



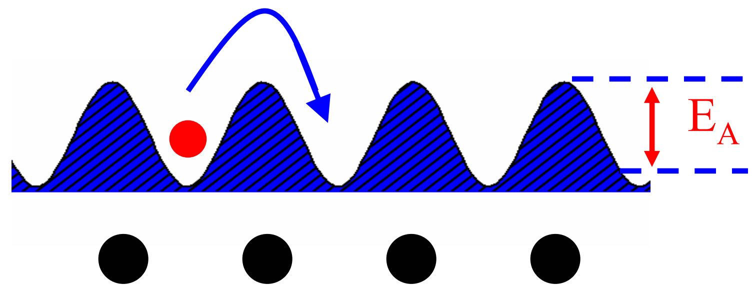
Defect migration
enthalpy

Probe-defect
binding energy

The perturbation of angular correlations by dynamic QI

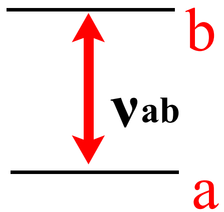
Example: Hydrogen diffusion in solids

Thermally activated H diffusion



Activation energy

Residence time
$\tau_R = \tau_R(0)\exp(E_A/k_BT)$



Time dependent QI → Nuclear relaxation; rate R $\approx \dfrac{\nu_f^2\,\tau_R}{1+(\nu_{ab}\,\tau_R)^2}$

fast fluctuations:

τ_R << precession period $1/\nu_{ab}$

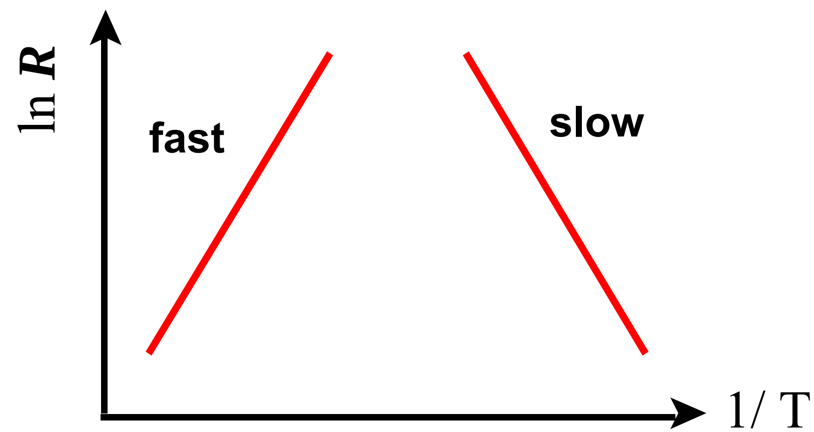
$R \approx \nu_f^2 \cdot \tau_R \approx \exp(E_A/k_BT)$

slow fluctuations:

τ_R >> precession period $1/\nu_{ab}$

$R \approx \nu_f^2 / \tau_R \approx \exp(-E_A/k_BT)$

Arrhenius plot: ln *R* vs. 1/T



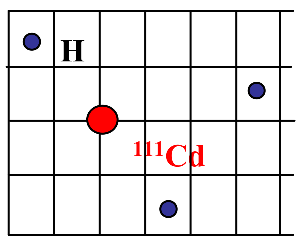
Hydrogen Diffusion studied by perturbed angular correlations

Characteristic times: Residence time τ_R and PAC time window 10 ns $\leq \Delta T \leq$ 1 μs

$\tau_R >> \Delta T$:

(quasi-) static,

frequency distribution

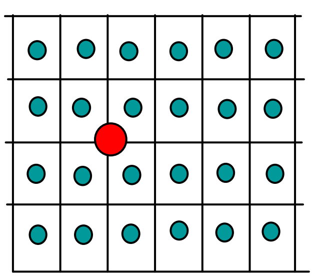


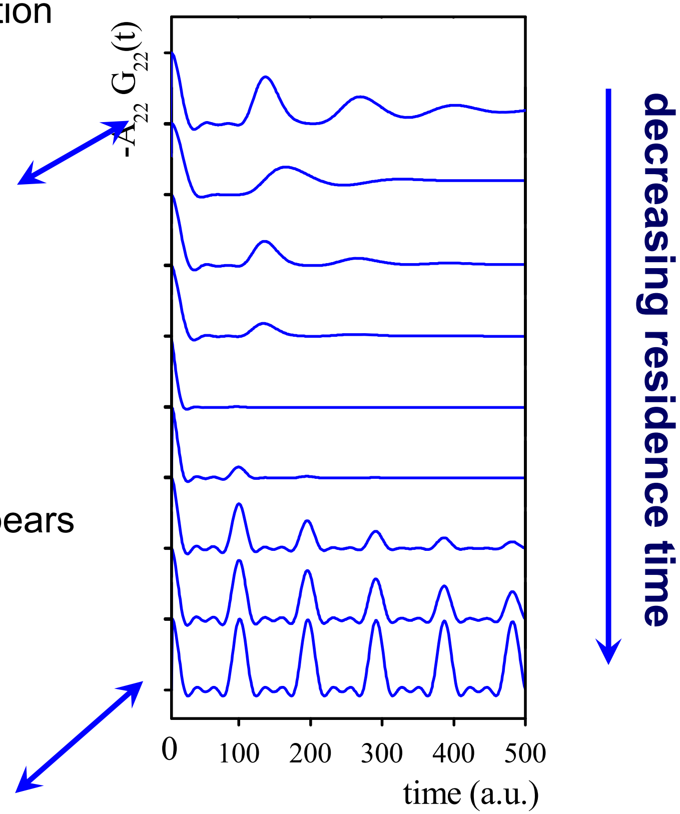
$\tau_R << \Delta T$:

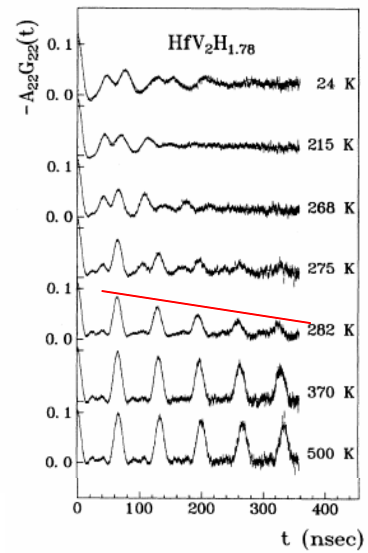
fast fluctuations

time average appears

to be static



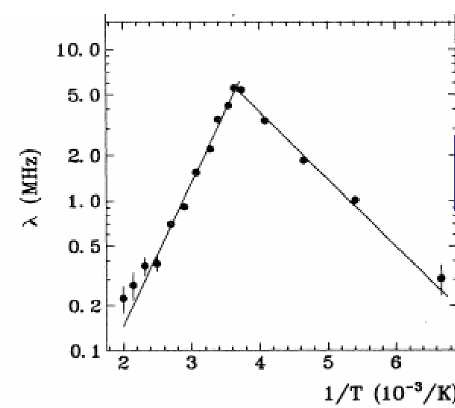




exp(-λt)

λ ≈ relaxation rate

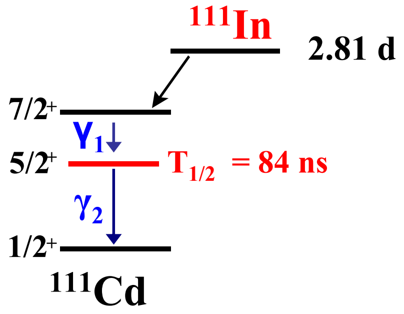
Arrhenius plot

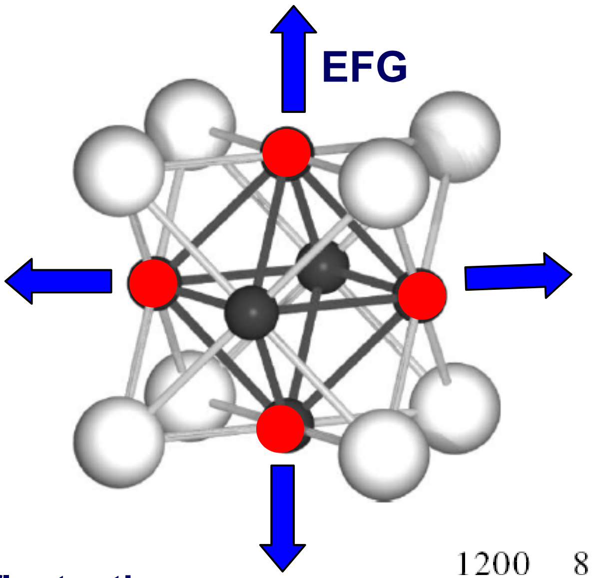


E_a = 2500 K

M. Forker, Nuclear Techniques in Solid State Resaerch , CBPF 2012

Dynamic QI in solids: Reorientation jumps of ^{111}In in In_3La





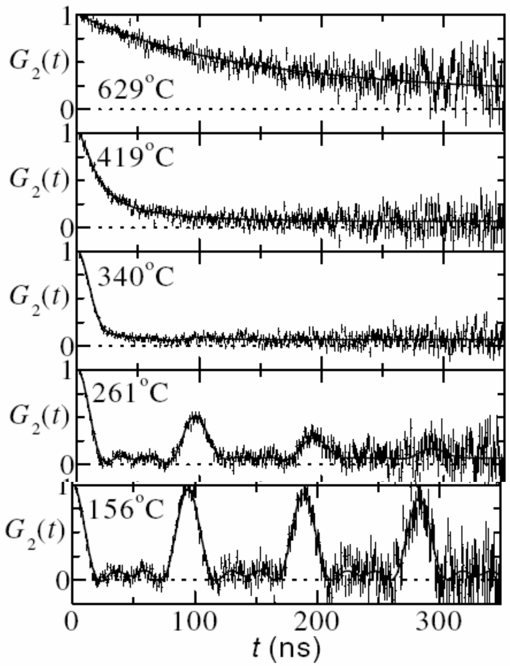


FIG. 2. Perturbation functions of ^{111}Cd tracer atoms in In_3La at the indicated temperatures.

fast fluctuations

static

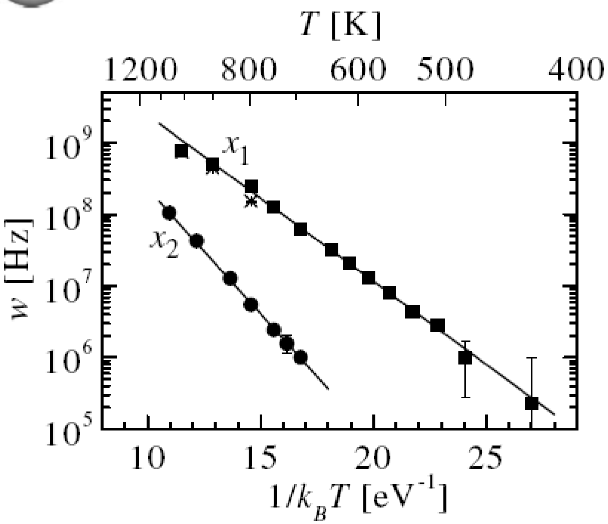


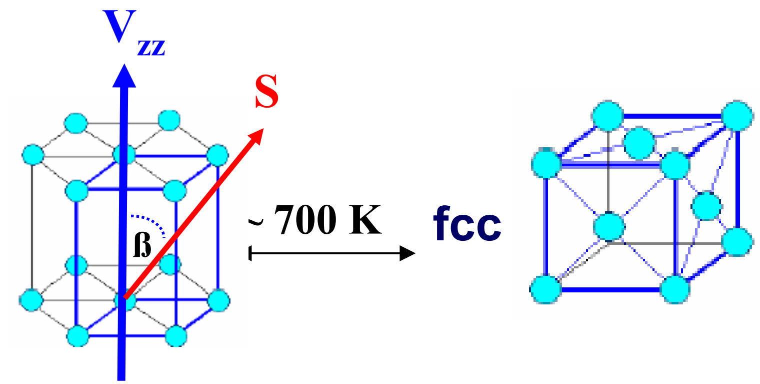
FIG. 3. Temperature dependence of the frequency of reorientation of the electric field gradient at the nuclei of Cd tracer atoms caused by jumps on the In sublattice in In_3La. The lines indicate fits of data for the two different compositions, with symbols and labels defined in the text.

M. Zacate, A. Favrot, G. Collins, PRL 92 (2004) 225901

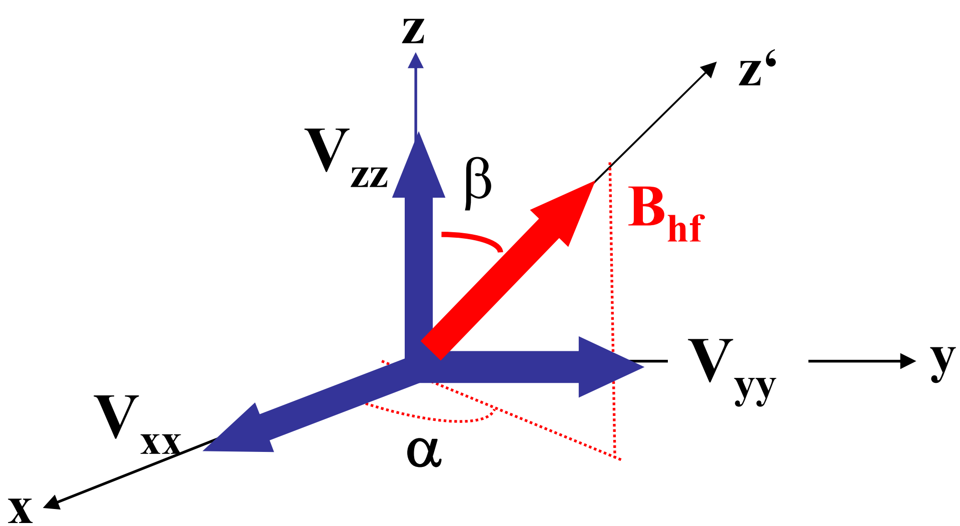
PAC study of magnetic properties

Example: ferromagnetic Cobalt

Cobalt: T_C = 1390 K, structure: hcp

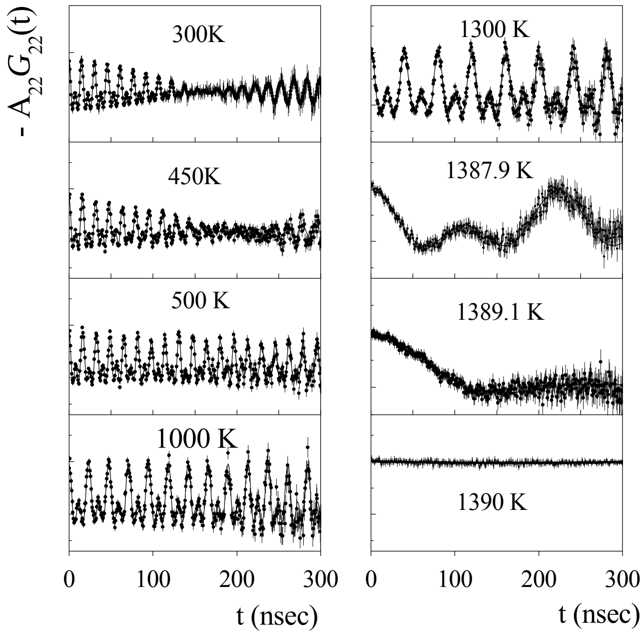


Combined magnetic + electric hfi

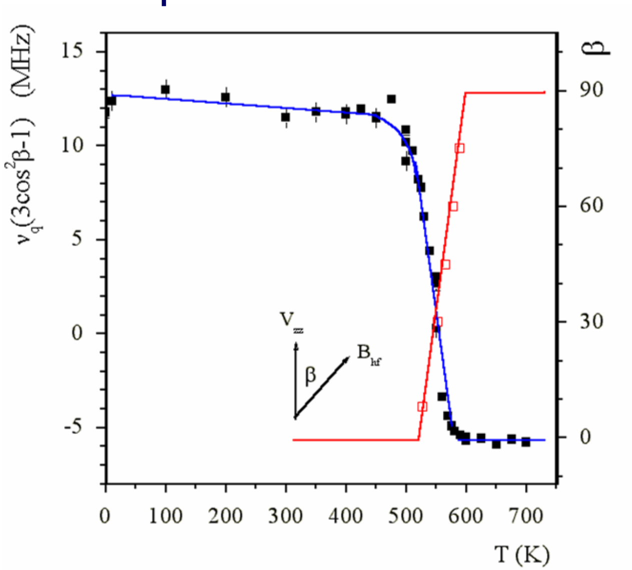


- $\nu_Q = eQV_{zz} / h$
- $\eta = \frac{V_{yy} - V_{xx}}{V_{zz}}$
- $\nu_M = g\mu_B B_{hf} / h$
- *Euler angles* β, γ

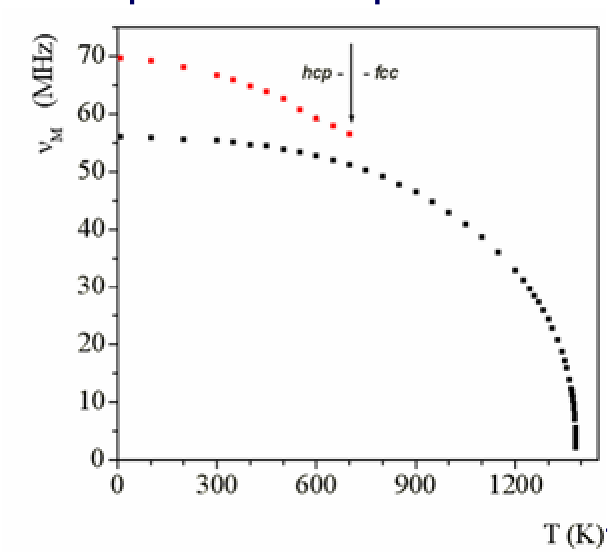
PAC spectra of ^{111}Cd in Co



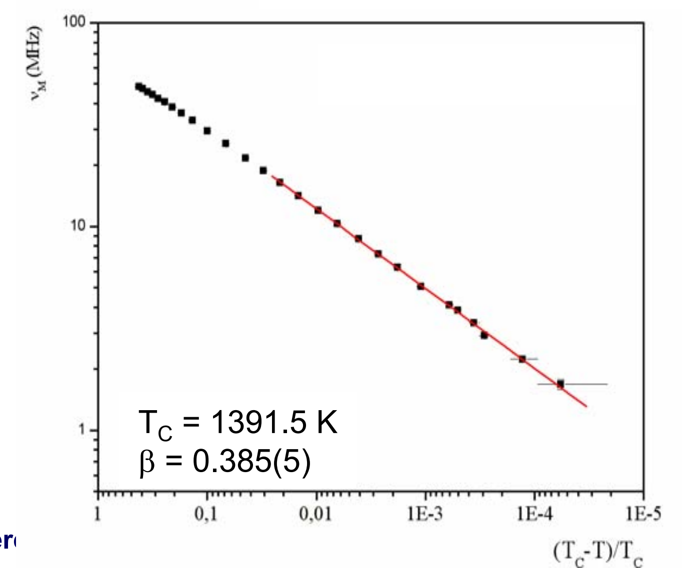
Spin reorientation



Temperature depedence of ν_M



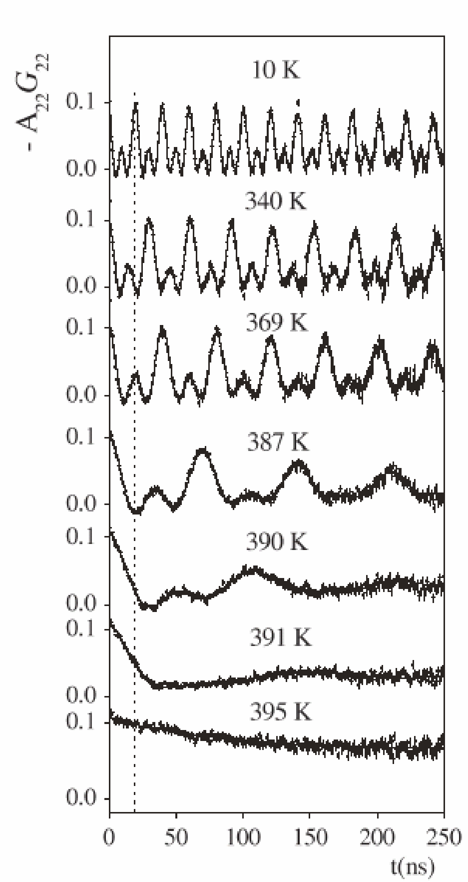
critical exponent: $\nu_M(T) = \nu_M(0)(1 - T/T_C)^\beta$



Phase transitions of RCo_2

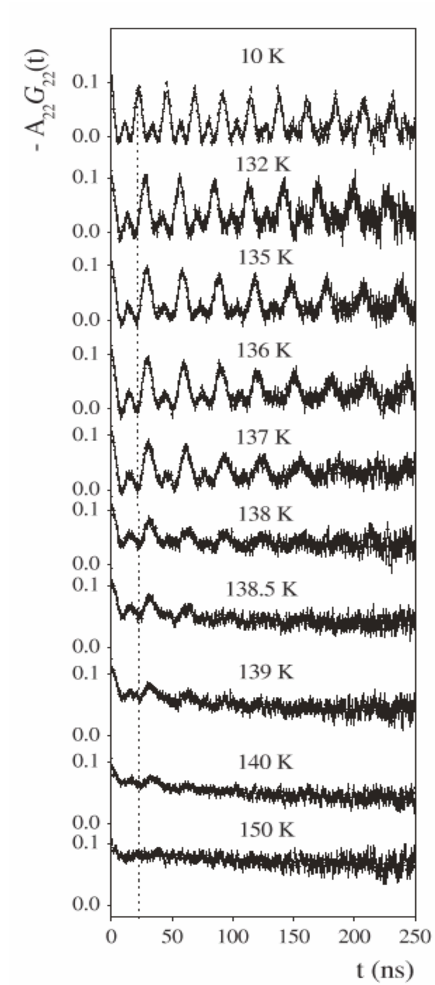
studied by measurement of the temperature dependence of the magnetic hyperfine field

*R*are earth = Pr, Nd, Sm, Gd, Dy, Ho, Er, Tm



$GdCo_2$

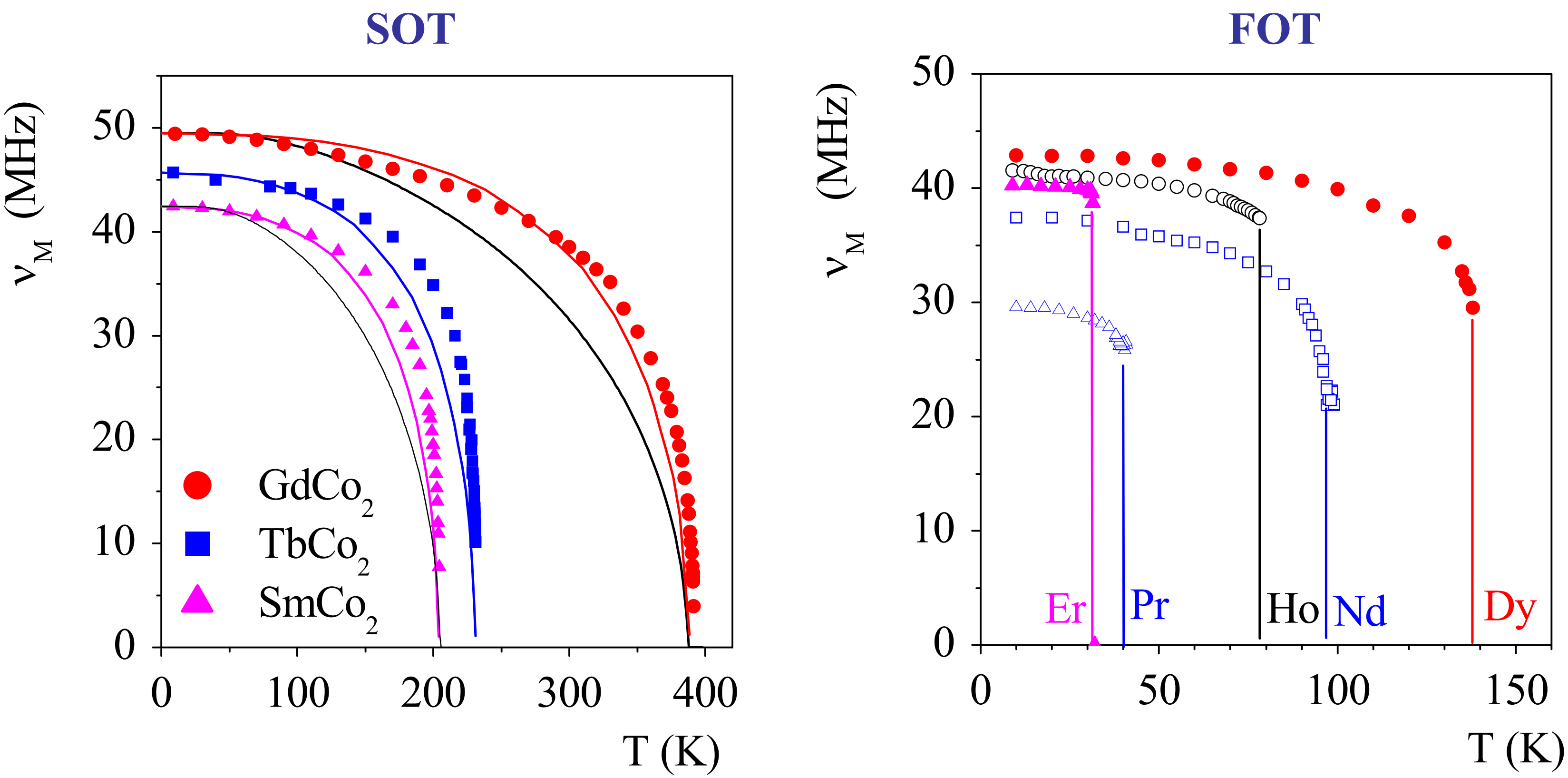
Second-order Transition SOT



$DyCo_2$

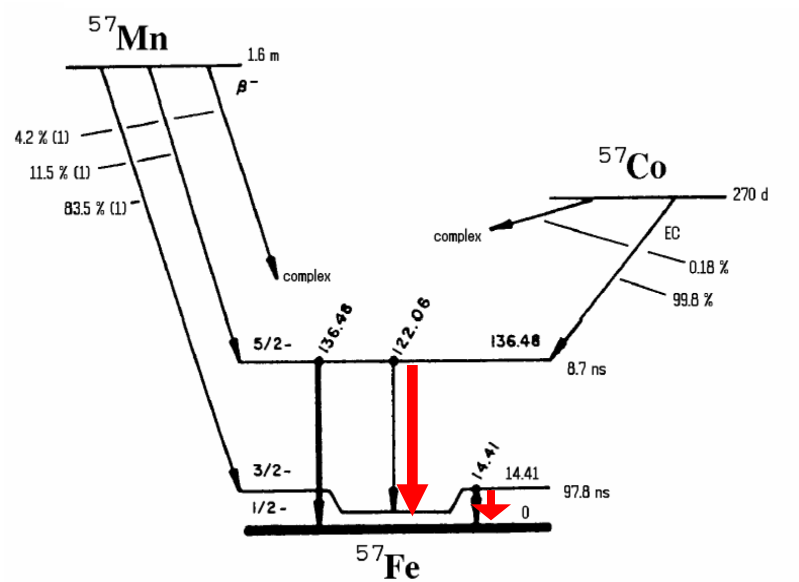
First-order Transition FOT

The order of the magnetic phase transitions of RCo_2 deduced from the magnetic hyperfine field at ^{111}Cd



Time-Differential Perturbed Angular-Correlation Experiment for ^{57}Fe in a Ni Host, and a Comparison with the Mössbauer Effect*

C. Hohenemser, R. Reno, H. C. Benski, and J. Lehr†

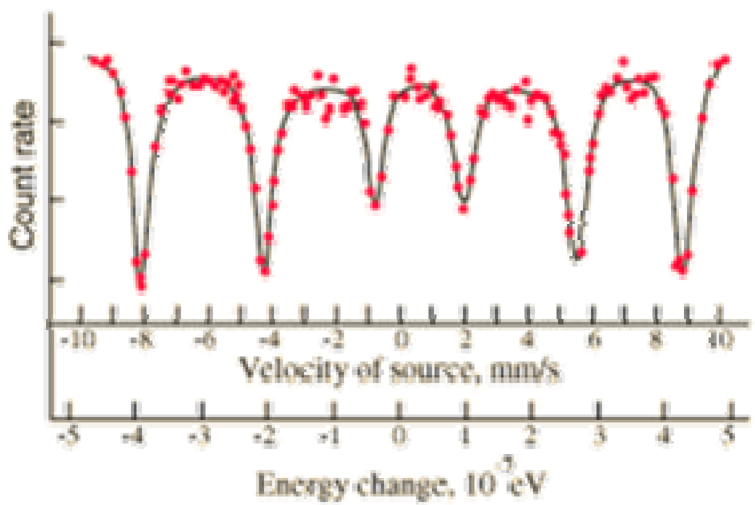


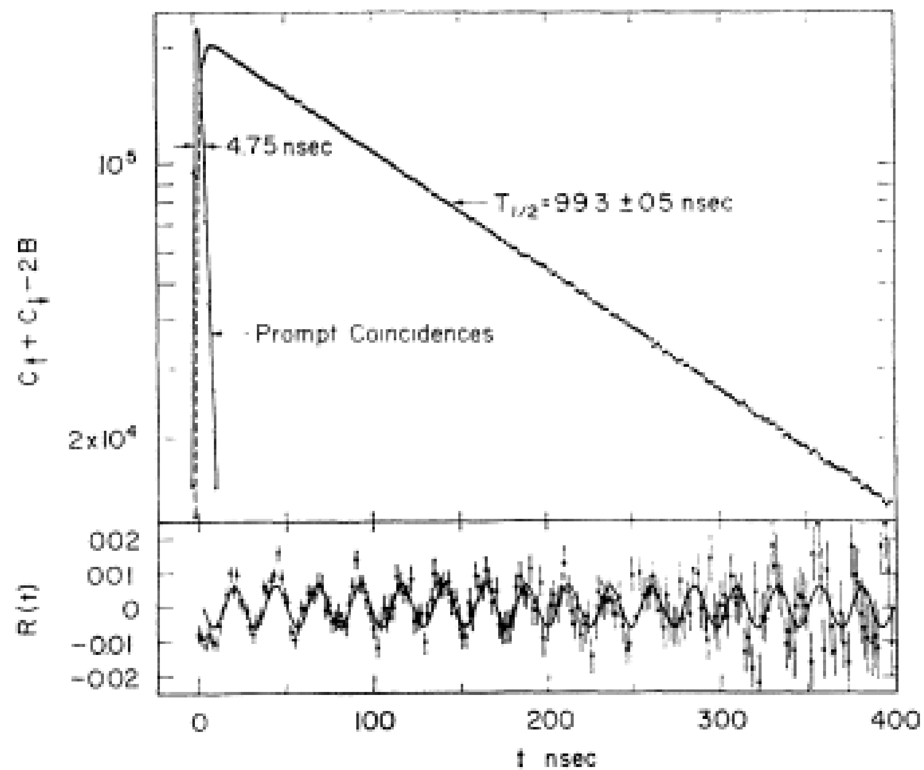
PAC source: ^{57}Co in Ni

$|B_{hf}(^{57}Fe:Ni)| = 267.8(2.7)$ kG

Mössbauer: 265(5) kG

$|B_{hf}(^{57}Fe:\alpha\text{-}Fe)| = 330$ kG





Comparison PAC – Mössbauer, NMR, NQR, NO, SH....

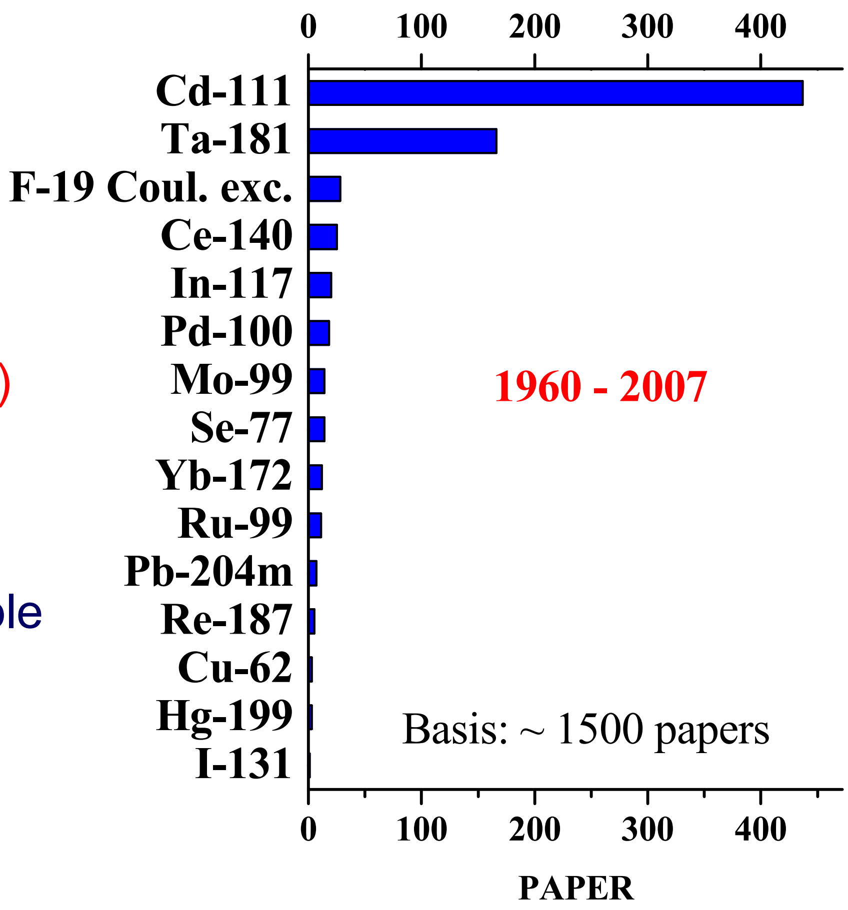
Advantages of PAC:

- Any temperature
- Any environment (solid, liquid, gas)
- Low concentration of probe nuclei: $10^{-12} – 10^{-6}$
- No restriction to low E_γ
- Small samples: 1-100 mg
- No external field or rf field required

Comparison PAC – Mössbauer, NMR, NQR, NO, SH,...

Disadvantages of PAC

- Limited number of probes for
  for laboratory experiment
- Source preparation
  (Radiochemistry installations required)
- Legal constraints
- Probe mostly an impurity in the sample
- Complex electronic set-up



Global PAC Activity