Perturbed Angular Correlations
in nuclear physics and material science

- The phenomenon of $\gamma$-$\gamma$ 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 $\gamma - \gamma$ angular correlations
The time-integral mode (IPAC)

Parent isotope

$\gamma_1$

$\gamma_2$

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

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

$A_{kk} = $ angular correlation coefficients

depend on spins, parities and multipole order of the cascade

About $10^{11}$ parent isotopes in a sample

integrates over all times between $\gamma_1$ and $\gamma_2$


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

**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

Nuclear physics: Dermination of the multipole order and spins by measurement of $I_{l,m}(\Theta)$
Gamma emission from **aligned** nuclear states

\[ E_{\gamma}^{(l,m)}(l_a, m_a) \]

**Example**: dipole \((l = 1)\)

\[ I_{1,0}(\Theta) \approx \sin^2 \Theta \]

\[ I_{1,\pm 1}(\Theta) \approx \frac{1}{2}(1 + \cos^2 \Theta) \]

The radiation distribution informs on the spin orientation
Angular Correlations – Theoretical background

The first $\gamma$-transition with $I = 1$

$\gamma_1$

$\gamma_2$

$I_f = 0$

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

Radiation emitted by an ensemble of randomly oriented nuclear spins:

Nuclear spins seen by Det. #1

Observation of $\gamma_1 = \text{selection of a subgroup of spin orientations}$

Spin alignment!
The second γ-transition of the γγ-cascade

Emission from an aligned spin ensemble

By observation of γ₁ we select a specific orientation of the spin I.

The direction of γ₂ informs us on changes in the spin orientation while the nucleus is in the intermediate state.
OBSERVATION MODES
of Angular Correlations

Time integral mode: IPAC
Half-width $\tau_R \geq$ life time $\tau$
- Integration over all times of the intermediate state of the cascade
- Time-integrated observation of hyperfine interactions

Time differential mode: TDPAC
Half-width $\tau_R \leq$ life time $\tau$
- Measurement of the anisotropy as a function of the time the nucleus has spent in the intermediate state of the cascade
- Time resolved observation of hyperfine frequencies $\nu_L \leq 1/\Delta\tau$
The time-differential mode (TDPAC) of unperturbed $\gamma - \gamma$ angular correlations

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

$\tau = 10^{-9} - 10^{-6}$ s
The perturbation of γγ angular correlations by hyperfine interactions

Basic aspects:

(i) Detection of $\gamma_1$ = selection of a subgroup of spin orientations  
→ anisotropic intensity distribution of $\gamma_2$

(ii) Hyperfine interaction → Larmor precession of spins  
= Precession of the intensity distribution of $\gamma_2$

(iii) Detection of $\gamma_2$ = spin orientation at time $t$

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

$$\Delta \Theta = \omega t_1$$
The perturbation of $\gamma\gamma$ angular correlations by hyperfine interactions

The time differential mode

Example: Magnetic field $B_\perp$ detector plane

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

![Graph showing 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 K_t are displayed. The full line represents the weighted least-squares fit to the data.](image)
The perturbation of angular correlations by static hyperfine interactions

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

\[ G_{kk}(t) = \text{perturbation factor} \]

**Electric quadrupole interaction**

\[ E_Q(\hbar \nu_Q) \]

\[ l = \frac{5}{2} \]

\[ \hbar \nu_1 \]

\[ \hbar \nu_2 \]

\[ \hbar \nu_3 \]

**Magnetic dipole interaction**

\[ 2\hbar \nu_M \]

\[ \hbar \nu_M \]

**Combined MHFI + QI**

111Cd in hcp Co

181Ta in ZrO₂

**Dynamic QI**

111Cd in fcc Co

111Cd:HfV₂Hₓ

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Mean life time $\tau$ 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: $\leq 15$

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**Basis:** ~ 1500 papers

**1960 - 2007**
TDPAC spectrometer – basic aspects

Requirements:

- high statistics
- high detection efficiency
- energy resolution
- time resolution

Present standard:

- Multi-detector arrays: up to 6 detectors
- Fast photomultiplier coupled to BaF$_2$ scintillators
- Time resolution: $\sim 100$ ps FWHM at $^{60}$Co energies

Popular isotopes:

- $^{181}$Hf
  - $\beta^-$
  - $1/2^+$
  - $5/2^+$
  - $7/2^+$
  - $t_{1/2} = 42.5$ d
  - $\gamma_1 = 133$ keV
  - $\gamma_2 = 482$ keV

- $^{181}$Ta
  - $\beta^-$
  - $1/2^+$
  - $5/2^+$
  - $7/2^+$
  - $t_{1/2} = 10.8$ ns

- $^{111}$In
  - $\text{EC}$
  - $7/2^+$
  - $5/2^+$
  - $1/2^+$
  - $t_{1/2} = 2.81$ d
  - $\gamma_1 = 172$ keV
  - $\gamma_2 = 247$ keV
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 synchrotron 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).

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

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

Counts vs. time

Anisotropy vs. time (exp. decay eliminated)

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

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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**: $\nu_q \neq 0$, $\eta \neq 0$, $T \sim 1450$ K
- **tetragonal**: $\nu_q \neq 0$, $\eta = 0$, $T \sim 2150$ K
- **cubic, EFG = 0**: $\nu_q = 0$

**QI and PAC spectra**

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Phase identification and phase transformation by QI - Example: ZrO₂

ZrO₂ phases

- **monoclinic**
  - $\nu_q \neq 0$
  - $\eta \neq 0$

- **tetragonal**
  - $\nu_q \neq 0$
  - $\eta = 0$

- **cubic, EFG = 0**

Phase transformations

- **m-ZrO₂**
  - $290 \text{ K} - \nu_q = 770 \text{ MHz}, \eta = 0.36$

- **t-ZrO₂**
  - $1383 \text{ K} - \nu_q = 820 \text{ MHz}, \eta = 0.0$

- **c-ZrO₂**
  - $2120 \text{ K} - \nu_q \sim 0 \text{ MHz}$

PAC spectra

- Heating and cooling curves for different phases.

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Static QI in solids:
Solid state reaction: Transformation of Zr metal into ZrB$_2$

Zr + BN $\rightarrow$ ZrB$_2$

$^{181}$Hf:Zr metal

$^{181}$Hf:ZrB$_2$
Static QI in solids: Defects in metals and semiconductors studied by TDPAC

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

Subsequent annealing

Defect migration enthalpy

$nn$ defect binding energy

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

\[ \tau_R = \tau_R(0) \exp \left( \frac{E_A}{k_B T} \right) \]

Fast fluctuations:

\[ \tau_R \ll \text{precession period } \frac{1}{\nu_{ab}} \]

\[ R \approx \nu_f^2 \cdot \tau_R \approx \exp \left( \frac{E_A}{k_B T} \right) \]

Slow fluctuations:

\[ \tau_R \gg \text{precession period } \frac{1}{\nu_{ab}} \]

\[ R \approx \frac{\nu_f^2}{\tau_R} \approx \exp \left( \frac{-E_A}{k_B T} \right) \]

Arrhenius plot: \( \ln R \) vs. \( 1/T \)

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Hydrogen Diffusion studied by perturbed angular correlations

Characteristic times: Residence time $\tau_R$ and PAC time window $10 \text{ ns} \leq \Delta T \leq 1 \mu s$

$\tau_R >> \Delta T$: (quasi-) static, frequency distribution

$\tau_R << \Delta T$: fast fluctuations, time average appears to be static

$\exp(-\lambda t)$
$\lambda \approx$ relaxation rate

Arrhenius plot

$E_a = 2500 \text{ K}$
Dynamic QI in solids: Reorientation jumps of $^{111}$In in $\text{In}_3\text{La}$

$^{111}\text{In}$ 2.81 d

$\gamma_1$

$\gamma_2$

$^{111}\text{Cd}$

$T_{1/2} = 84$ ns

$\gamma_1$

$\gamma_2$

FIG. 2. Perturbation functions of $^{111}\text{Cd}$ tracer atoms in $\text{In}_3\text{La}$ at the indicated temperatures.

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 $\text{In}_3\text{La}$. The lines indicate fits of data for the two different compositions, with symbols and labels defined in the text.
PAC study of magnetic properties

Example: ferromagnetic Cobalt

Cobalt: $T_C = 1390 \text{ K}$, 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 $\beta, \gamma$
PAC spectra of $^{111}$Cd in Co

Temperature dependence of $\nu_M$

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

$T_C = 1391.5$ K
$\beta = 0.385(5)$
Phase transitions of $R\text{Co}_2$

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

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

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**GdCo$_2$**
Second-order Transition
SOT

**DyCo$_2$**
First-order Transition
FOT
The order of the magnetic phase transitions of $R\text{Co}_2$ deduced from the magnetic hyperfine field at $^{111}\text{Cd}$
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

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

- Any temperature
- Any environment (solid, liquid, gas)
- Low concentration of probe nuclei: $10^{-12} – 10^{-6}$
- No restriction to low $E_\gamma$
- 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 laboratory experiment
- Source preparation (Radiochemistry installations required)
- Legal constraints
- Probe mostly an impurity in the sample
- Complex electronic set-up

**Comparison**

<table>
<thead>
<tr>
<th>PAC</th>
<th>Mössbauer</th>
<th>NMR</th>
<th>NQR</th>
<th>NO</th>
<th>SH</th>
</tr>
</thead>
</table>

**Graph**

- 1960 - 2007
- Basis: ~ 1500 papers

- Cd-111
- Ta-181
- F-19 Coul. exc.
- Ce-140
- In-117
- Pd-100
- Mo-99
- Se-77
- Yb-172
- Ru-99
- Pb-204m
- Re-187
- Cu-62
- Hg-199
- I-131

**Bar Chart**

- PAPER: 0 100 200 300 400

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Global PAC Activity