

Implementation of "ab initio" Perturbed Angular Correlation Observables for Analysis of Fluctuating Quadrupole Interactions

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November, 2010



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Thesis submitted for the degree of Master in Physics

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Resumo

É apresentada uma revisão sobre as propriedades nucleares, nomeadamente os momentos nucleares (momento dipolar magnético e momento quadrupolar eléctrico) e a sua interacção com campos electromagnéticos externos ao núcleo (interacções hiperfinas), assim como a distribuição angular da radiação produzida por decaimento γ .

Uma descrição detalhada sobre a teoria de Correlações Angulares Perturbadas é feita, incluindo a comparação entre correlações $\gamma - \gamma$ e correlações $e^- - \gamma$.

Para interacções nucleares dinâmicas, foi feita uma introdução à teoria de estados estocásticos em PAC.

Focámo-nos na implementação *ab-initio* de observáveis para analisar flutuações nas interacções hiperfinas quadrupolares em experiências de correlações angulares perturbadas dependentes no tempo.

O desenvolvimento de códigos computacionais para resolver o problema, adaptados ao ajuste de dados obtidos em monocristais ou policristais para campos transitórios de dois estados com qualquer simetria axial e orientação foi o objectivo principal desde trabalho.

A última parte foi a aplicação a um primeiro caso teste: $^{181}\text{Hf} : \text{GaN}(n)$ e $^{181}\text{Hf} : \text{GaN}(\text{Mg})$.

Abstract

A review about the nuclear properties, namely the nuclear moments (magnetic dipole moment and electric quadrupole moment) and their interaction with electromagnetic fields external to the nucleus (hyperfine interactions), as well as the angular distribution of radiation produced by γ -decay, is presented.

A detailed description about the theory of Perturbed Angular Correlations was done, including the comparison between $\gamma - \gamma$ correlations and $e^- - \gamma$ correlations.

For dynamic nuclear interactions, an introduction to the theory of stochastic states in PAC was performed.

We focused on *ab-initio* implementation of observables for analysing fluctuating quadrupole hyperfine interactions on time dependent perturbed angular correlations experiments.

The development of computational codes solving the full problem, adapted to fit data obtained on single crystals or polycrystals for two-state transient fields with any axial symmetry and orientation was the main purpose of this work.

The final part was the application of a first test case: $^{181}\text{Hf}/^{181}\text{Ta} : \text{GaN}(n)$ and $^{181}\text{Hf}/^{181}\text{Ta} : \text{GaN}(\text{Mg})$.

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1. Motivation

The study of the nuclear electronic environment in matter gives us the knowledge about atomic scale phenomena and, consequently, provides a better understanding about the microscopic origin of the macroscopic features of materials.

The measurement of the interactions between the nuclear magnetic and quadrupole moments with the extranuclear electromagnetic fields, named Hyperfine Interactions, provides a very useful tool. By probing the local charge distribution and magnetic hyperfine fields, hyperfine interactions techniques are very sensitive and accurate to investigate condensed matter phenomena, given that subtle differences in the microscopic aspects of materials originate very distinct macroscopic properties.

There are several nuclear hyperfine methods, such as Mössbauer, nuclear magnetic resonance or muon-spin rotation, among other techniques, but we will focus on Perturbed Angular Correlations (PAC). In spite of the fact that PAC has been widely used for about 40 years to study a multitude of phenomena from point defect structure, electronic excitation, local deformations, etc., dynamic phenomena originating transient fields are scarcely understood due to the complexity of the analysis, even if potentially revealing the atomic and electronic dynamic at the nanoscopic scale. Recent examples are phenomena like polaron formation and dynamic distortions existing in complex, e.g., manganite systems that require real-lattice atomic scale studies with direct measurements on the local aspects, PAC being a perfect tool for such task given that it allows us to visualize local effects and not lattice properties. Due to the hyperfine interactions of magnetic or quadrupole electric moments of the nuclei with external fields, i.e., with the electronic environment and spin around the nuclei (the nuclear quadrupole electric moment interacts with the electric field gradient and the magnetic moment with external magnetic fields), it provides access to defects or local deformations even if they exist without any kind of periodicity.

PAC measurements are done and discussed most frequently when the electric field gradients (or magnetic fields) remain constant in time, i.e., constant during the characteristic measurement time, ranging from a few tens of nanoseconds to few hundreds of nanoseconds: static

1. Motivation

cases.

The particular case of slow recovery processes on the electronic environment after the loss of electrons from the lower atomic orbits, due to electron conversion or electron capture decays, or simply due to the dynamic of electrons, point defects or polarons (cell deformations) make different nuclei sense slightly different electric field gradients (EFG) due to charge density distortions in their close or remote neighborhood. This requires a dynamic description of PAC spectra, so we studied and used a description based on stochastic processes developed by Blume [1] and Winkler and Gerdau [2], which is appropriate to describe general cases of discrete jumps between several different states that might involve strong changes of the interaction.

The implementation of first principle calculation of PAC observable that accounts for time-dependent interactions, i.e., that enables the interpretation of situations where the electric field gradient (EFG) on the surroundings of probe nuclei fluctuates within the time range of the PAC measurement, was considered.

This work consisted of implementing a C++ program (TFPAC) that produces the PAC observable (firstly) and fits (secondly) the experimental data for dynamic cases (but it can also be used for static ones).

The following physics situations have been considered:

- We should be able to fit single-crystals and polycrystals like experiments
- General nuclear spin values must be possible to use
- We limited the case of two possible state transitions (due to limited computational resources).

Immediate situations where this program can be used to analyse are, e.g, the after-effects in materials and polaronic fluctuations like the ones which occur in manganites (studied with PAC).

Such program is the first, to our knowledge, to properly simulate single-crystals, to accept any EFG1 and EFG2 (e.g., $\eta_1 \neq \eta_2 \neq 0$) and where the principal diagonalization axis orientation does not need to be colinear (different Euler angles for each state).

Because it was conceived within an *ab initio* treatment, TFPAC calculates theoretical values without any approximations.

The case study to be briefly presented will be PAC experiments combining $\gamma - \gamma$ and $e^- - \gamma$ PAC measured on the $\tau = 10.8\text{ ns}$ intermediate state of ^{181}Ta obtained onto decay of ^{181}Hf implanted in $\text{GaN}(n)$ (n-type semiconductor) and $\text{GaN}(\text{Mg})$ (doped with magnesium).

The analysis with TFPAC will be discussed as well as the strategy for improving the fitting speed and the simulations capability (parallel processing), in particular justifying the necessity of implementing 3 state transition Hamiltonian.

Given that for this work it was necessary to understand with detail all the theoretical expressions involved in Perturbed Angular Correlations, in quadrupole interactions (the current Hamiltonian does not include magnetic interactions, by now), as well as the theory of Blume to the implementation of “stochastic” Hamiltonians, and because in some cases we didn’t find the explicit derivation of some steps necessary to implement the correct theory, we used this thesis as an opportunity to do a brief review and derivations when these were not easily found on literature of the essential steps to achieve the necessary results.

For this reason, we divide the thesis in these different chapters:

- **Chapter 2: Nuclear Properties**

Description of nuclear moments (magnetic dipolar moment and electric quadrupole moment), the energy terms due to hyperfine interactions (either electric or magnetic) and the angular distribution of radiation emitted by a nuclear γ decay.

- **Chapter 3: Perturbed $\gamma - \gamma$ Angular Correlation (PAC)**

Description of the general theory of PAC with the derivation of the theoretical expressions and the introduction to time-dependent interactions. Although not all the theoretical calculations are presented, the derivation of the theoretical expressions is still very detailed, so for those more interested in experimental results than in heavy mathematics, the important result is given by the perturbed angular correlation function $W(k_1, k_2, t)$ which contains the perturbation factor $G(t)$. All the information about the nuclei under consideration is given by the perturbation factor.

It is also explained how to get the expressions for $e^- - \gamma$ (electron-gamma) PAC experiments knowing the $\gamma - \gamma$ PAC theory.

- **Chapter 4: Theory of Stochastic Perturbations in Angular Correlations**

Presentation and explanation of Blume’s theory (ref.[1]) used to implement dynamic interactions in PAC experiments. The calculation performed by Winkler and Gerda (ref.[2]) was followed, adding some extra comments and descriptions.

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- **Chapter 5: Implementing the Time Dependent Perturbation Function $R(t)$**

After knowing the theory of PAC we need to know how to get information from experimental data, so we discuss the implementation of the experimental perturbation function $R(t)$ which gives relevance to the perturbation factor $G(t)$, allowing us to get information about the hyperfine interactions. We also introduce the concepts of static distributions, including frequency distributions, time resolution and attenuation coefficients due to corrections in the solid angles of the detectors.

- **Chapter 6: TFPAC Program**

At this point, we already know the theory, so we can take a look at the structure of the program and how it gives us the results we need.

The most important topics are explained, such as the rotation matrix to non-zero Euler angles, matrix diagonalization to find eigenvectors and eigenvalues of the Blume matrix and the implementation of frequency distributions. The last two points were the most painful to solve so they are explained in more detail.

- **Chapter 7: Experimental Data and Analysis**

Experimental data for the cases $^{181}\text{Hf}/^{181}\text{Ta} : \text{GaN}(n)$ and $^{181}\text{Hf}/^{181}\text{Ta} : \text{GaN}(\text{Mg})$ is showed and we explain the method used to fit the results. The physical phenomena behind the results are briefly discussed.

- **Chapter 8: Conclusions and Final Comments**

A review of the obtained results is done as well as a summary of the goals achieved during this work. Finally, we present the ideas about how we can continue this work and what are the important points which we should focus on first in future investigation.

2. Nuclear Properties

2.1. Nuclear Moments

2.1.1. Nuclear Magnetic Dipolar Moment

The magnetic moment $\boldsymbol{\mu}$ of a charged body with angular momentum $\mathbf{I}$ is given classically by

$$\boldsymbol{\mu} = \gamma \mathbf{I} \quad (2.1.1)$$

where γ is the gyromagnetic ratio.

In quantum mechanics, $\boldsymbol{\mu}$ and $\mathbf{I}$ are operators, thus the magnetic moment is defined as the component of $\boldsymbol{\mu}$ along the z -axis in the state $|I, m\rangle$ with $m = I$ (I is the quantum number for total angular momentum and m is the quantum number for the projection of I along z)

$$\mu = \langle I, I | \mu_z | I, I \rangle = \gamma \langle I, I | I_z | I, I \rangle \quad (2.1.2)$$

which gives

$$\mu = \gamma \hbar I \quad (2.1.3)$$

where we recall $I_z |I, m\rangle = m\hbar |I, m\rangle$.

The nuclear gyromagnetic ratio is defined as

$$\gamma = g \frac{\mu_N}{\hbar} \quad (2.1.4)$$

where g is an adimensional factor designated by g -factor and μ_N is the nuclear magneton

$$\mu_N = \frac{e\hbar}{2m_p} = 5.05 \times 10^{-27} A.m^2 \quad (2.1.5)$$

(m_p is the proton mass and e is the proton charge).

For nuclei with orbital angular momentum $\mathbf{L}$, the g -factor is given by

$$\begin{aligned} g_L(\text{proton}) &= 1 \\ g_L(\text{neutron}) &= 0 \end{aligned} \quad (2.1.6)$$

2. Nuclear Properties

but there is also a g -factor associated to the spin $\mathbf{S}$

$$\begin{aligned} g_S(\text{proton}) &= 5.59 \\ g_S(\text{neutron}) &= -3.83. \end{aligned} \quad (2.1.7)$$

Thus, for nuclei with total angular momentum $\mathbf{I}$ and magnetic moment $\boldsymbol{\mu}$, we have the relations

$$\boldsymbol{\mu} = g_L \mathbf{L} + g_S \mathbf{S}$$

with $\mathbf{I} = \mathbf{L} + \mathbf{S}$. $\boldsymbol{\mu}$ is expressed in units of μ_N and $\mathbf{I}$, $\mathbf{L}$ and $\mathbf{S}$ in units of $\hbar$.

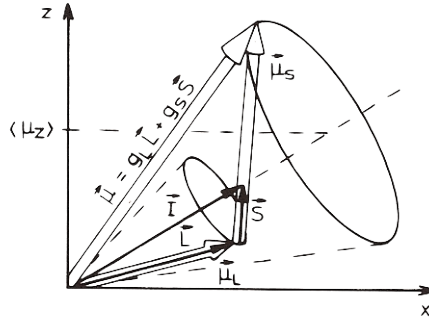


Figure 2.1.1.: Coupling scheme for angular momenta and magnetic moments. Only the component of $\boldsymbol{\mu}$ parallel to $\mathbf{I}$ is observable. Perpendicular components average zero (ref.[3])

2.1.2. Electric Quadrupole Moment

A volumic charge distribution $\rho(\mathbf{r})$ creates an electric potential given by

$$\phi(\mathbf{r}) = \frac{1}{4\pi\epsilon_0} \int \rho(\mathbf{r}') \frac{dv'}{|\mathbf{r} - \mathbf{r}'|} \quad (2.1.8)$$

where, according to ref.[3], its multipolar expansion can be written in terms of Legendre Polynomials as

$$\phi(\mathbf{r}) = \frac{1}{4\pi\epsilon_0 r} \int \rho(\mathbf{r}') \sum_{l=0}^{\infty} P_l(\cos \theta) \left(\frac{r'}{r}\right)^l dv' \quad (2.1.9)$$

being θ the angle between $\mathbf{r}$ and $\mathbf{r}'$.

Analysing only the quadrupolar term ($l = 2$) we get the expression

$$\phi_2(\mathbf{r}) = \frac{1}{4\pi\epsilon_0} \int \rho(\mathbf{r}') \frac{\sum_{i,j} x_i(3x'_i x'_j - r'^2 \delta_{ij})x_j}{2r^5} dv' \quad (2.1.10)$$

thus defining the electric quadrupole moment tensor

$$Q_{ij} = \frac{1}{e} \int \rho(\mathbf{r})(3x'_i x'_j - r'^2 \delta_{ij}) dv' \quad (2.1.11)$$

being e the proton charge.

In Nuclear Physics, the quadrupole interaction is of great interest.

The atomic nuclei may have electric quadrupole moments and its magnitude and sign reflect the nature of the forces between nuclei and protons, as well as the shape of the nuclei themselves. Its existence indicates a deformation in the spherical shape of the nucleus, however still existing a symmetry axis (Fig.(2.1.2)). Thus, a spherically symmetric charge distribution has a null quadrupole moment.

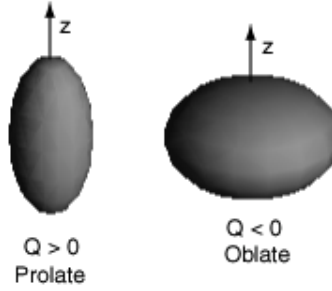


Figure 2.1.2.: Examples of non-zero electric quadrupole moments (ref.[4]).

The quadrupole moment of a nuclear state is defined as $Q = Q_{zz}$:

$$Q = \frac{1}{e} \int (3z^2 - r^2) \rho(\mathbf{r}) d^3r \quad (2.1.12)$$

Since $r^2 = x^2 + y^2 + z^2$, we can write $(3z^2 - r^2) = 3r^2 \cos^2 \theta - r^2 = r^2(3 \cos^2 \theta - 1)$, where θ is the angle between the position vector $\mathbf{r}$ and the z axis. As we know, the spherical harmonic for $l = 2$ and $m = 0$ is $Y_2^0 = \sqrt{\frac{5}{4\pi}}(\frac{3}{2} \cos^2 \theta - \frac{1}{2})$, thereby

$$Q = \frac{1}{e} \sqrt{\frac{16\pi}{5}} \int r^2 Y_2^0 \rho(\mathbf{r}) d^3r \quad (2.1.13)$$

In Quantum Mechanics, Q is defined as the expectation value of the quadrupole operator $\sqrt{\frac{16\pi}{5}} Z r^2 Y_2^0$ in the state $|I, m = I\rangle$

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$$Q = \sqrt{\frac{16\pi}{5}} \langle I, m = I | Z r^2 Y_2^0 | I, m = I \rangle = \langle Q_{20} \rangle \quad (2.1.14)$$

where Z is the atomic number. The quantity $Q_{20} = Z r^2 Y_2^0$ is a tensor operator. Using the Wigner-Eckart theorem (appendix A.2) we get

$$\langle I, m | Q_{20} | I, m \rangle = (-1)^{I-m} \begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix} \langle I || Q_2 || I \rangle \quad (2.1.15)$$

where the elements between brackets are called 3j-symbols (appendix A.1), whose properties give $Q = 0$ for $I < 1$, since the 3j-symbols are zero in this case.

Q has the dimensions of length squared, i.e., the dimensions of area. Normally, Q is expressed in units of barns where

$$1 \text{ barn} = 100 fm^2 = 10^{-28} m^2. \quad (2.1.16)$$

We still have to define the operator $\hat{Q}_{ij}$ (where i and j are cartesian coordinates), which we do in terms of Q . We realize that acting in the space of states defined by $| I, m \rangle$, the matrix elements of any operator in this space must be reproduced by an analogous operator formed from the angular momentum operator $\hat{I}_i$. The quadrupole operator is a rank 2, symmetric, traceless tensor, and hence the operator given in terms of the I_i must also have these properties. Therefore we find that the general form for $\hat{Q}_{ij}$ must be (ref.[5])

$$\langle I, m | \hat{Q}_{ij} | I, m \rangle = \alpha(I) \langle I, m | \frac{3}{2} (\hat{I}_i \hat{I}_j + \hat{I}_j \hat{I}_i) - \delta_{ij} \hat{I}^2 | I, m \rangle \quad (2.1.17)$$

where α is a yet to be determined constant. It is again convenient to examine the matrix element of $\hat{Q}_{zz}$ between states of $| I, m = I \rangle$

$$\begin{aligned} \langle I, I | \hat{Q}_{zz} | I, I \rangle &= \alpha(I) \langle I, I | \frac{3}{2} (\hat{I}_z \hat{I}_z + \hat{I}_z \hat{I}_z) - \hat{I}^2 | I, I \rangle \\ &= \alpha(I) (3I^2 - I(I+1)) \\ &= \alpha(I) I (2I-1) = Q \end{aligned} \quad (2.1.18)$$

and hence we have solved for the constant

$$\alpha(I) = \frac{Q}{I(2I-1)}. \quad (2.1.19)$$

We can then write the operator as

$$\hat{Q}_{ij} = \frac{Q}{I(2I-1)} \left[\frac{3}{2} (\hat{I}_i \hat{I}_j + \hat{I}_j \hat{I}_i) - \delta_{ij} \hat{I}^2 \right]. \quad (2.1.20)$$

2.2. The Hyperfine Interactions Energy

Up to now we have considered only the electromagnetic properties of free nuclei. We now consider the interaction between a nucleus and an external electric or magnetic field. The interaction of a nucleus with these fields is called a Hyperfine Interaction. Through the analysis of the hyperfine structure we can experimentally determine several inherent characteristics of the nucleus.

2.2.1. Electric Interactions

Let's consider a nuclear charge density $\rho(\mathbf{r})$ with an external electrical potential $\phi(\mathbf{r})$.

The electrostatic energy is given by

$$E = \int \rho(\mathbf{r})\Phi(\mathbf{r})d^3r \quad (2.2.1)$$

where

$$\int \rho(\mathbf{r})d^3r = Z.e \quad (2.2.2)$$

is the nuclear charge.

Expanding the potential in Taylor series around $\mathbf{r} = 0$, we get

$$\Phi(\mathbf{r}) = \Phi(0) + \sum_{i=1}^3 \left(\frac{\partial \Phi}{\partial x_i} \right)_0 x_i + \frac{1}{2} \sum_{i,j} \left(\frac{\partial^2 \Phi}{\partial x_i \partial x_j} \right)_0 x_i x_j + \dots \quad (2.2.3)$$

where x_i and x_j are Cartesian coordinates.

Inserting this expression in equation (2.2.1), we obtain the energy as a sum of several terms

$$E = E_0 + E_1 + E_2 + \dots \quad (2.2.4)$$

where

$$E_0 = \Phi_0 \int \rho(\mathbf{r})d^3r \quad (2.2.5)$$

$$E_1 = \sum_{i=1}^3 \left(\frac{\partial \Phi}{\partial x_i} \right)_0 \int \rho(\mathbf{r})x_i d^3r \quad (2.2.6)$$

2. Nuclear Properties

$$E_2 = \frac{1}{2} \sum_{i,j} \left(\frac{\partial^2 \Phi}{\partial x_i \partial x_j} \right)_o \int \rho(\mathbf{r}) x_i x_j d^3 r. \quad (2.2.7)$$

So, let's analyse each term separately.

By eq. (2.2.2), it's easily verified that $E_0 = \Phi_0$. *Z.e.* As Φ_0 is the potential at the origin ($\mathbf{r} = 0$), E_0 is the Coulomb energy of a point charge in an external potential. This term only contributes to the potential energy of the crystal lattice, being the same for every isotope of the same element and so it's not important for our discussion.

The term E_1 can be written as

$$E_1 = [\nabla \Phi]_{r=0} \int \rho(\mathbf{r}) x_i d^3 r \quad (2.2.8)$$

By definition, $\int \rho(\mathbf{r}) x_i d^3 r$ is the electric dipole moment $\mathbf{p}_i$ and $\mathbf{E} = -\nabla \phi$ is the electric field, thus

$$E_1 = -(\mathbf{E} \cdot \mathbf{p})_{r=0}. \quad (2.2.9)$$

This is the interaction energy of an electric dipole moment $\mathbf{p}$ of a nuclear charge distribution with an external electric field, at the origin ($\mathbf{r} = 0$).

Due to the well defined parity of the nuclear wave function, the electric dipole moment of the nuclear charge distribution is zero, thus $E_1 = 0$.

Let us focus then on the remaining term, E_2 .

For simplification, we do the substitution

$$\left(\frac{\partial^2 \Phi}{\partial x_i \partial x_j} \right)_0 = \Phi_{ij} \quad (2.2.10)$$

giving us

$$E_2 = \frac{1}{2} \sum_{i,j} \Phi_{ij} \int \rho(\mathbf{r}) x_i x_j d^3 r \quad (2.2.11)$$

$$= \frac{1}{6} \sum_i \Phi_{ii} \int \rho(\mathbf{r}) r^2 d^3 r + \frac{1}{2} \sum_{i,j} \Phi_{ij} \int \rho(\mathbf{r}) \left(x_i x_j - \frac{r^2}{3} \delta_{ij} \right) d^3 r \quad (2.2.12)$$

where $r^2 = x_1^2 + x_2^2 + x_3^2$ and the last separation in two terms is useful ahead.

2.2. The Hyperfine Interactions Energy

The electrostatic potencial $\Phi(\mathbf{r})$ obeys the Poisson equation, so at the nucleus we have

$$(\Delta\Phi)_0 = \sum_i \Phi_{ii} = \frac{e}{\epsilon_0} |\psi(0)|^2 \quad (2.2.13)$$

where $|\psi(0)|^2$ is the probability density of electrons at the nucleus (only nonnegligible for s electrons) and $-e|\psi(0)|^2$ is the charge density. Therefore, we have

$$E_2 = E_C + E_Q$$

with

$$\begin{aligned} E_C &= \frac{e}{6\epsilon_0} |\psi(0)|^2 \int \rho(\mathbf{r}) r^2 d^3r \\ E_Q &= \frac{1}{2} \sum_{i,j} \Phi_{ij} \int \rho(\mathbf{r}) \left(x_i x_j - \frac{r^2}{3} \delta_{ij} \right) d^3r \end{aligned} \quad (2.2.14)$$

Monopolar Term E_C

From eq. (2.2.14) we see that E_C only depends on the mean square nuclear radius, given by

$$\langle r^2 \rangle = \frac{1}{Ze} \int \rho(r) r^2 d^3r \quad (2.2.15)$$

resulting in

$$E_C = \frac{Z.e^2}{6\epsilon_0} |\psi(0)|^2 \langle r^2 \rangle \quad (2.2.16)$$

This monopolar term describes the electrostatic interaction of an expanded nucleus with electrons in the nuclear zone. E_C then triggers a shift in the levels but not its split into different sub-levels. Thus it gives rise to the isotope shift, causing slightly different spectral lines for isotopes with different nuclear radius, so, this term won't be important for us.

2. Nuclear Properties

Electric Quadrupole Interaction E_Q

Let us now give special attention to the term E_Q . From the definition of electric quadrupole moment, eq.(2.1.11), we can write

$$E_Q = \frac{e}{6} \sum_{i,j} \Phi_{ij} Q_{ij} \quad (2.2.17)$$

Because Φ_{ij} is a symmetric matrix, it is possible to find an eigenvectors basis where this matrix is diagonal, giving us

$$E_Q = \frac{e}{6} \sum_i \Phi_{ii} Q_{ii}. \quad (2.2.18)$$

Considering

$$\Phi_{ii} = V_{ii} + \frac{1}{3} Tr(\Phi) \quad (2.2.19)$$

being $Tr(\Phi) = \sum_i \Phi_{ii}$ the trace of Φ , V_{ii} is a traceless matrix. Q is also traceless ($\sum_i Q_{ii} = 0$), and because $\frac{1}{3} Tr(\Phi)$ is a constant, it doesn't contribute to E_Q .

This way we get

$$E_Q = \frac{e}{6} \sum_i V_{ii} Q_{ii} \quad (2.2.20)$$

V_{ii} is called the electric field gradient (EFG). Only charges not at the nuclear site contribute to V_{ii} .

For spherically symmetric charge distributions (s electrons ($l = 0$)), $V_{xx} = V_{yy} = V_{zz}$. Since $\sum_i V_{ii} = 0$, all components of the electric field gradient are zero and cannot contribute to the energy E_Q . In fact, only charges with $l \geq 1$ contribute to V_{ii} , and therefore to E_Q .

Because $\sum_i V_{ii} = 0$, the electric field gradient is completely described by two parameters. By appropriate choice of the principal axis system, one can have $|V_{zz}| \geq |V_{yy}| \geq |V_{xx}|$.

Generaly one chooses to describe the electric field gradient by V_{zz} and the asymmetry parameter η .

$$\eta = \frac{V_{xx} - V_{yy}}{V_{zz}} \quad (2.2.21)$$

The asymmetry parameter measures the deviation of the local charge distribution from axial symmetry (e.g. tetragonal, rhombohedral or hexagonal).

Thus, E_Q is the product of the nuclear electric quadrupolar moment with the electric field gradient in the nuclear site - nuclear quadrupole interaction.

The quadrupole energy can be written using the asymmetry parameter η explicitly, and EFG principal component, V_{zz} , yielding,

$$E_Q = \frac{eV_{zz}}{12} (3Q_{zz} + \eta(Q_{xx} - Q_{yy})) \quad (2.2.22)$$

To calculate the effect of the quadrupole interaction on a nuclear state characterized by its angular momentum $\hbar I$ the quantum formulation has to be used. The expression (2.2.22) remains valid for the interaction hamiltonian $\hat{H}_Q$ with the quadrupole tensor components, Q_{ii} , substituted by the correspondent quantum operators, $\hat{Q}_{ii}$.

From eq.(2.1.20) we have that the operators $\hat{Q}_{ii}$ are given by

$$\hat{Q}_{ii} = \frac{Q}{I(2I-1)} [3\hat{I}_i\hat{I}_i - \hat{I}^2] \quad (2.2.23)$$

thus the quadrupole interaction hamiltonian can be written as

$$\hat{H}_Q = \frac{eQV_{zz}}{4I(2I-1)} [3\hat{I}_z^2 - \hat{I}^2 + \eta(\hat{I}_x^2 - \hat{I}_y^2)] \quad (2.2.24)$$

and taking advantage of ladder operators - the raising operator $\hat{I}_+$ and the lowering operator $\hat{I}_-$,

$$\begin{aligned} \hat{I}_+ &= \hat{I}_x + i\hat{I}_y \\ \hat{I}_- &= \hat{I}_x - i\hat{I}_y \end{aligned} \quad (2.2.25)$$

we get

$$\hat{H}_Q = \frac{eQV_{zz}}{4I(2I-1)} \left[3\hat{I}_z^2 - \hat{I}^2 + \frac{\eta}{2} (\hat{I}_+^2 + \hat{I}_-^2) \right]. \quad (2.2.26)$$

Calculating $\langle I, m | \hat{H}_Q | I, m' \rangle$, the only non-zero matrix elements of $\hat{H}_Q$ are

$$\begin{aligned} H_{m,m} &= \hbar\omega_Q (3m^2 - I(I+1)) \\ H_{m,m\pm 2} &= \hbar\omega_Q \frac{1}{2} \eta \sqrt{(I \mp m - 1)(I \mp m)(I \pm m + 1)(I \pm m + 2)} \end{aligned} \quad (2.2.27)$$

where we defined the quadrupole frequency

$$\omega_Q = \frac{eQV_{zz}}{4I(2I-1)\hbar} \quad (2.2.28)$$

2. Nuclear Properties

that gives us the energy scale for the nuclear quadrupole interaction.

For the particular case of an axially symmetric interaction ($\eta = 0$), the eigenvalues of the hamiltonian are given by

$$E_Q = \hbar\omega_Q \left[3m^2 - I(I+1) \right] \quad (2.2.29)$$

The transition energy between two sublevels m and m' is given by

$$\Delta E_Q = \hbar\omega_Q \left| m^2 - m'^2 \right| \quad (2.2.30)$$

$|m^2 - m'^2|$ is always integer so all transition frequencies are multiples of ω_Q and the fundamental observable frequency ω_0 , the lowest transition frequency is:

$$\omega_0 = 6\omega_Q, \text{ for } I \text{ half-interger}$$

$$\omega_0 = 3\omega_Q, \text{ for } I \text{ integer}$$

Is also usual to define another quantity, the fundamental frequency:

$$\nu_Q = \frac{eQV_{zz}}{\hbar} = \omega_0 \frac{4I(2I-1)}{2\pi k} \quad (2.2.31)$$

where $k = 3$ (for I integer) and $k = 6$ (for I half-integer).

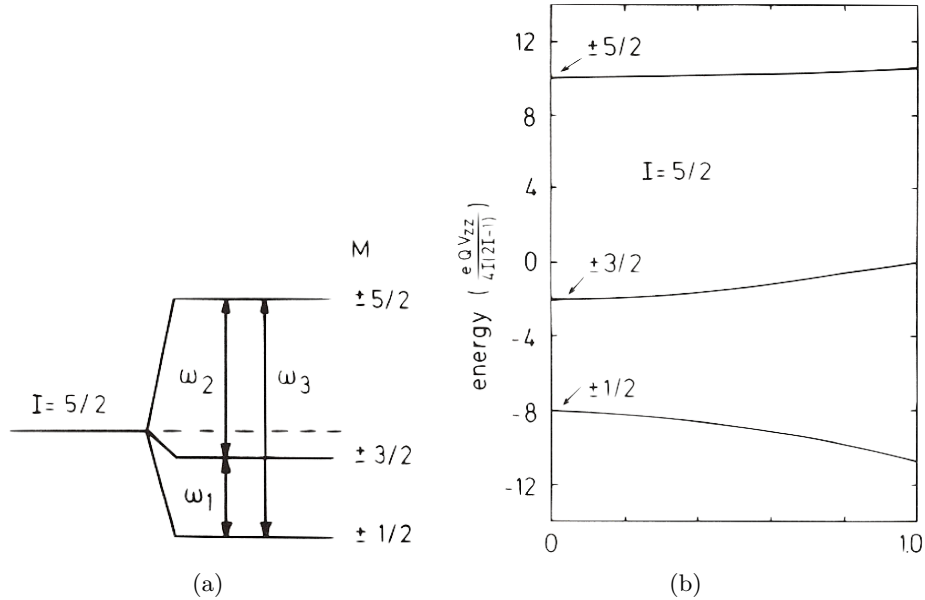


Figure 2.2.1.: (a) Energy splitting of an $I = 5/2$ nuclear level under the influence of an axially symmetric quadrupole interaction. (b) Energy splitting of an $I = 5/2$ nuclear state as a function of the asymmetry parameter η ($V_{zz} = \text{constant}$). The m values shown in the figure are strictly valid only for $\eta = 0$ (ref.[3]).

2.2.2. Magnetic Interactions

The magnetic nuclear dipole moment $\boldsymbol{\mu}$ interacts with a magnetic flux density $\mathbf{B}$ at the position of the nucleus. The interaction energy is

$$E_{magn} = -\boldsymbol{\mu} \cdot \mathbf{B} \quad (2.2.32)$$

This extra energy breaks the degeneracy of the nuclear m states and induces a precession of the nuclear spin. Here we are only interested in the first feature.

Choosing the z axis parallel to the magnetic field and considering the z -component of the magnetic moment, $\mu_z = \frac{g\mu_N}{\hbar} I_z$ (eq.2.1.1) we know that the interaction energy in the state $|I, m\rangle$ is given by

$$E_{magn} = \langle I, m | -\mu_z B_z | I, m \rangle = -\frac{g\mu_N}{\hbar} B_z \langle I, m | \hat{I}_z | I, m \rangle = -g\mu_N B_z m. \quad (2.2.33)$$

The energy difference between adjacent m levels is

$$E_{magn}(m+1) - E_{magn}(m) = -g\mu_N B_z. \quad (2.2.34)$$

This means the splitting is the same between all consecutive m levels (see Fig.(2.2.2)).

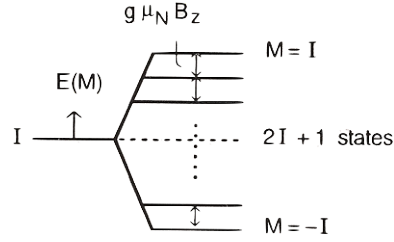


Figure 2.2.2.: Nuclear level splitting in a B magnetic field (nuclear Zeeman effect) (ref.[3])

2.3. Nuclear γ Decay

Quantum levels of nuclei are characterized by well-defined total angular momentum I and parity π .

In the transition from a higher to lower energy level, γ -rays are often emitted. The following conservation relations hold:

$$\begin{array}{ll} \text{energy} & E_i = E_f + \hbar\omega \\ \text{angular momentum} & I_i = I_f + l \\ \text{parity} & \pi_i = \pi_f \pi \end{array} \quad (2.3.1)$$

2. Nuclear Properties

During the transition from an initial state (I_i, M_i, π_i) to a final state (I_f, M_f, π_f) , a photon γ with quantum numbers (l, m, π) is emitted (Fig.(2.3.1)).

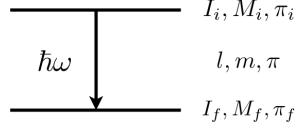


Figure 2.3.1.: Schematic representation of a γ transition between an initial state i and a final state f .

The emission of a photon is equivalent to the creation of an electromagnetic wave with energy $\hbar\omega$. The conservation of angular momentum and parity requires that the emitted wave, like the nuclear states, has well-defined angular momentum and parity. We therefore look for solutions of Maxwell's equations expressed in the form of multipole radiation where angular momentum and parity are explicit parameters.

In free space, Maxwell equations are:

$$\begin{aligned} \nabla \times \mathbf{E} &= -\frac{\partial}{\partial t} \mathbf{B} & \nabla \times \mathbf{B} &= \frac{1}{c^2} \frac{\partial \mathbf{E}}{\partial t} \\ \nabla \cdot \mathbf{E} &= 0 & \nabla \cdot \mathbf{B} &= 0 \end{aligned} \quad (2.3.2)$$

where $\mathbf{E}$ is the electric field, $\mathbf{B}$ is the magnetic field and c is the speed of light in free space.

Because $\mathbf{E}, \mathbf{B} \propto e^{-i\omega t}$, from equations (2.3.2) we get

$$\begin{aligned} \nabla \times \mathbf{E} &= i\omega \mathbf{B} = ikc \mathbf{B} \\ \nabla \times \mathbf{B} &= -\frac{i}{c^2} \omega \mathbf{E} = -i\frac{k}{c} \mathbf{E} \end{aligned} \quad (2.3.3)$$

where $\mathbf{k}$ is the wave vector. Using the identity relations between the rotational operator $(\nabla \times)$ and the laplacian (∇^2) , we can write equations (2.3.3) as

$$\begin{aligned} (\nabla^2 + k^2) \mathbf{E} &= 0 \\ (\nabla^2 + k^2) \mathbf{B} &= 0 \end{aligned} \quad (2.3.4)$$

We can see that these equations obey Helmholtz wave equation $((\nabla^2 + k^2)\psi(r, \omega) = 0)$ whose solutions are given as $\psi(r, \omega) = \sum_{l,m} f_l(kr) Y_{lm}(\theta, \phi)$ (ref.[6]), where $f_l(kr)$ only depend on r but not on angle and are proportional to the spherical Bessel functions, and $Y_l^m(\theta, \phi)$ are the spherical harmonics, but we should note that the solutions for $\mathbf{B}$ and $\mathbf{E}$ are limited by the conditions $\nabla \cdot \mathbf{E} = 0$ and $\nabla \cdot \mathbf{B} = 0$, given by eq.(2.3.2).

Following the explicit calculation done in ref.([6]), we can get the final solution for the multipole fields, having for the electromagnetic fields of the magnetic multipole of order (l, m)

$$\begin{aligned}\mathbf{B}_{lm}^{(M)} &= f_l(kr)\mathbf{L}Y_l^m(\theta, \phi) \\ \mathbf{E}_{lm}^{(M)} &= \frac{i}{k}\nabla \times \mathbf{B}_{lm}^{(M)}\end{aligned}\tag{2.3.5}$$

and for the electric multipole of order (l, m)

$$\begin{aligned}\mathbf{E}_{lm}^{(E)} &= f_l(kr)\mathbf{L}Y_l^m(\theta, \phi) \\ \mathbf{B}_{lm}^{(E)} &= -\frac{i}{k}\nabla \times \mathbf{E}_{lm}^{(E)}\end{aligned}\tag{2.3.6}$$

where we introduced the angular momentum operator $\mathbf{L} = -i\hbar(\mathbf{r} \times \nabla)$.

Thus, for the solutions of the desired multipole fields we have:

- Electrical ($2^l - poles$): equations (E);
- Magnetic ($2^l - poles$): equations (M).

These are the radiation fields of oscillating ($2^l - poles$).

So far we have considered only classical electromagnetic radiation. In quantum electrodynamics, the γ quantum corresponding to the multipole radiation l has an angular momentum with magnitude $\sqrt{l(l+1)}\hbar$ and z component $m\hbar$.

In addition, the multipole fields have a well defined parity:

$$\begin{aligned}\pi &= (-1)^l && \text{for } E \text{ radiation} \\ \pi &= (-1)^{l+1} && \text{for } M \text{ radiation}\end{aligned}\tag{2.3.7}$$

The multipole transitions will then obey to the angular momentum selection rules

$$l = I_i + I_f, I_i + I_f - 1, \dots, |I_i - I_f|\tag{2.3.8}$$

2. Nuclear Properties

Radiation Angular Distribution

Let us now calculate the angular distribution of emitted γ -rays based on the solutions of the Maxwell equations.

The energy flux density of an electromagnetic wave is given by the Poynting vector

$$\mathbf{S} = \frac{1}{\mu_0} (\mathbf{E} \times \mathbf{B}) \quad (2.3.9)$$

which is the direction of propagation of the electromagnetic wave.

Since the distance between the detectors and the source is much larger than the source dimensions (nuclear scale), the far-field solutions for $\mathbf{E}$ and $\mathbf{B}$ can be used

$$\epsilon_0 |\mathbf{E}|^2 = \frac{1}{\mu_0} |\mathbf{B}|^2 \quad (2.3.10)$$

and because $c^2 = \frac{1}{\epsilon_0 \mu_0}$,

$$|\mathbf{E}| = c |\mathbf{B}| \quad (2.3.11)$$

so, we get

$$|\mathbf{S}| = c \epsilon_0 |\mathbf{E}|^2 = \frac{c}{\mu_0} |\mathbf{B}|^2 \quad (2.3.12)$$

From equations (2.3.5) and (2.3.6), one obtains

$$\begin{aligned} |\mathbf{S}| &= c \epsilon_0 |\mathbf{E}|^2 \propto |\mathbf{L}Y_l^m(\theta, \phi)|^2 \\ |\mathbf{S}| &= \frac{c}{\mu_0} |\mathbf{B}|^2 \propto |\mathbf{L}Y_l^m(\theta, \phi)|^2 \end{aligned} \quad (2.3.13)$$

One sees that the electric and the magnetic radiations of the same multipole order have the same angular pattern, thus they are only distinguishable by the measurement of polarization.

Now, let us work the term $|\mathbf{L}Y_l^m(\theta, \phi)|$.

Remembering the ladder operators, L_+ and L_- , and the projection of the angular momentum operator in the z axis, L_z

$$\begin{aligned} L_+ &= L_x + iL_y & L_x &= \frac{1}{2}(L_+ + L_-) \\ L_- &= L_x - iL_y & L_y &= \frac{1}{2i}(L_+ - L_-) \end{aligned} \quad (2.3.14)$$

and using the relations of these operators when applied to the eigenvectors basis of angular momentum, $|l, m\rangle$, given by

$$\begin{aligned} L_+ |l, m\rangle &= \hbar\sqrt{l(l+1) - m(m+1)} |l, m+1\rangle \\ L_- |l, m\rangle &= \hbar\sqrt{l(l+1) - m(m-1)} |l, m-1\rangle \\ L_z |l, m\rangle &= m\hbar |l, m\rangle \end{aligned} \quad (2.3.15)$$

we obtain

$$\begin{aligned} |\mathbf{L}Y_l^m|^2 &= |L_x Y_l^m|^2 + |L_y Y_l^m|^2 + |L_z Y_l^m|^2 \\ &= \frac{1}{2} |L_+ Y_l^m|^2 + \frac{1}{2} |L_- Y_l^m|^2 + |L_z Y_l^m|^2 \\ &= \frac{\hbar^2}{2} (l(l+1) - m(m+1)) |Y_l^{m+1}|^2 + \frac{\hbar^2}{2} (l(l+1) - m(m-1)) |Y_l^{m-1}|^2 \\ &\quad + \hbar^2 m^2 |Y_l^m|^2. \end{aligned} \quad (2.3.16)$$

The expansion of the product of spherical harmonics is given by (ref.[7])

$$Y_{l_1}^{m_1}(\Omega) Y_{l_2}^{m_2}(\Omega) = \sum_{l=|l_1-l_2|}^{l_1+l_2} \sum_{m'=-l}^l \sqrt{\frac{(2l_1+1)(2l_2+1)}{4\pi(2l+1)}} \langle l_1, l_2; 0, 0 | l, 0 \rangle \langle l_1, l_2; m_1, m_2 | l, m' \rangle Y_l^{m'}(\Omega) \quad (2.3.17)$$

Since $|Y_l^m(\theta, \phi)|^2 = Y_{l,m}(\theta, \phi) Y_{l,m}^*(\theta, \phi)$, and $Y_{l,m}^*(\theta, \phi) = (-1)^m Y_{l,-m}(\theta, \phi)$, considering in eq.(2.3.17) $l_1 = l_2 = l$, $m_1 = m$, $m_2 = -m$, $l = k$ and only $m' = 0$, we get

$$|Y_l^m(\theta, \phi)|^2 = \sum_k \frac{2l+1}{\sqrt{4\pi(2k+1)}} \langle l, l; 0, 0 | k, 0 \rangle \langle l, l; m, -m | k, 0 \rangle Y_k^0(\theta, \phi) \quad (2.3.18)$$

Because (ref.[7])

$$Y_k^0 = \sqrt{\frac{2k+1}{4\pi}} P_k^0(\cos \theta) \quad (2.3.19)$$

substituting eq.(2.3.19) in eq.(2.3.18) and substituting the Clebsch-Gordan coefficients by the correspondent 3-j symbols (appendix A.1), we obtain the final expression

$$|Y_l^m(\theta, \phi)|^2 = \sum_k \frac{2l+1}{4\pi} (2k+1) \begin{pmatrix} l & l & k \\ m & -m & 0 \end{pmatrix} \begin{pmatrix} l & l & k \\ 0 & 0 & 0 \end{pmatrix} P_k(\cos \theta) \quad (2.3.20)$$

2. Nuclear Properties

After all this math, one can define the normalized angular distribution

$$F_{lm}(\theta) = \frac{|\mathbf{L}Y_l^m|^2}{\sum_m |\mathbf{L}Y_l^m|^2} \quad (2.3.21)$$

that can be calculated applying eq.(2.3.20) on eq.(2.3.16).

From the properties of the 3-j symbols (appendix A.1), the following condition must hold:

$k \leq 2l$ and k even, since $\begin{pmatrix} l & l & k \\ 0 & 0 & 0 \end{pmatrix} = 0$ for k odd.

As $|\mathbf{S}| \propto |\mathbf{L}Y_l^m(\theta, \phi)|^2$, we can easily see that $F_{lm}(\theta)$ gives us the probability of a photon γ to be emitted in the direction defined by the angle θ .

On Table 2.1 the angular distribution functions $F_{lm}(\theta)$ for dipole and quadrupole radiation are presented, with the respective emission patterns on Fig.(2.3.2).

Table 2.1.: Angular distribution functions $F_{lm}(\theta)$ for dipole and quadrupole radiation.

	$m = 0$	$m = \pm 1$	$m = \pm 2$
$l = 1$ (dipole)	$\frac{1}{2} \sin^2 \theta$	$\frac{1}{4} (1 + \cos^2 \theta)$	-
$l = 2$ (quadrupole)	$\frac{1}{2} \sin^2 \theta \cos^2 \theta$	$\frac{1}{4} (1 - 3 \cos^2 \theta + 4 \cos^4 \theta)$	$\frac{1}{4} (1 - \cos^4 \theta)$

Dipole

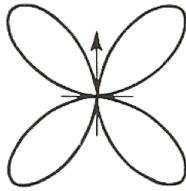


$l = 1, m = 0$

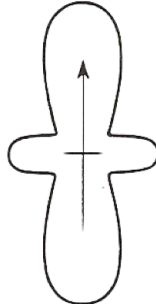


$l = 1, m = \pm 1$

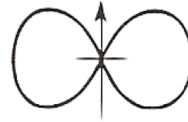
Quadrupole



$l = 2, m = 0$



$l = 2, m = \pm 1$



$l = 2, m = \pm 2$

Figure 2.3.2.: Emission patterns of pure dipole and quadrupole radiation (ref.[3]).

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

The probability of emission of a photon by a radioactive nucleus depends, in general, on the angle between the nuclear spin axis and the direction of emission. Under ordinary circumstances, the total radiation of a radioactive sample is isotropic because the nuclei are randomly oriented. Anisotropy is verified when a nuclear state is well oriented.

The basis of Perturbed Angular Correlations is the emission of two consecutive γ -rays (or $e^- - \gamma$) from internal decay of a probe nucleus. Due to the conservation of the angular momentum, I , there is a correlation between the emission directions, $\mathbf{k}_1$ and $\mathbf{k}_2$, of these two γ . Usually, in a set of nuclei the spins are randomly oriented in space and the emitted radiation is isotropic.

To obtain anisotropic γ angular distribution, the $2I + 1$ degenerate m sub-levels cannot be equally populated, i.e., the state from which the radiation is emitted has to be polarized or aligned. The nuclei are considered to be “aligned” if the density of states $\rho(m)$ of the sub-levels depends only on its absolute value, i.e, $\rho(m) = \rho(-m) \neq \rho(m')$. On the other hand, the set of nuclei is referred to as “polarized” when the density of states $\rho(m)$ depends on m with $\rho(m) \neq \rho(-m)$. (ref.[8])

In the case of angular correlations, the oriented set of nuclei is obtained by choosing only those nuclei whose spins happen to lie in a preferred direction. So, let us consider a nucleus in an initial state $|I_i, m_i, \pi_i\rangle$ that decays through a cascade emitting two successive γ -rays (Fig.(3.0.1)) and assuming that the initial state is randomly oriented, i.e., all the different m_i states are equally occupied, the observation of γ_1 in a fixed direction k_1 (which we choose as the z axis) selects an ensemble of nuclei in the intermediate state whose m magnetic sub-levels are no longer equally populated. This is a consequence of the angular momentum conservation¹ and the angular distribution of the electromagnetic radiation with respect to its angular momentum, L , being the angular distribution given by $F_{lm}(\theta)$, which was defined in eq.(2.3.21).

The angular correlation between the two photons (or $e^- - \gamma$) will be given by the correlation function $W(\mathbf{k}_1, \mathbf{k}_2) = W(\theta)$.

¹ $I_i = I + L$, where I_i and I are the angular momentum of the initial state and the intermediate nucleus state, respectively, and L is the photon angular momentum.

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

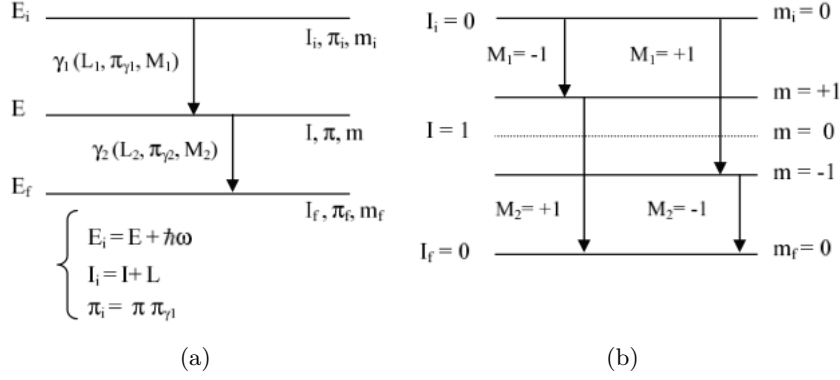


Figure 3.0.1.: (a) Schema representing the emission of two γ particles from a radioactive decay through a cascade between the nuclear states $E_i \rightarrow E \rightarrow E_f$ (i for initial and f for final states, respectively). The states are characterized by the angular momentum, I , and parity, π . The conservation are shown in the same figure. (b) Example of the alignment of the intermediate state, considering $I_i = 0 \rightarrow I = 1 \rightarrow I_f = 0$, i.e., the sublevel $m = 0$ is not populated (ref.[8]).

This is valid for free nuclei, but in the presence of extra-nuclear fields (electric or magnetic), an interaction between those fields and the nuclear moments appears. The resulting angular correlation will be perturbed, becoming time dependent. During the time t that a nucleus remains at the intermediate state (time elapsed between the emission of γ_1 and γ_2) the nucleus is subjected to hyperfine interactions, which will cause repopulations in the intermediate state, promoting changes in the occupation of the different magnetic sub-levels ($m - m'$ transitions) and consequently, the emission probability of γ_2 in a certain direction becomes time dependent. This gives result to the perturbed angular correlation function $W(\mathbf{k}_1, \mathbf{k}_2; t)$.

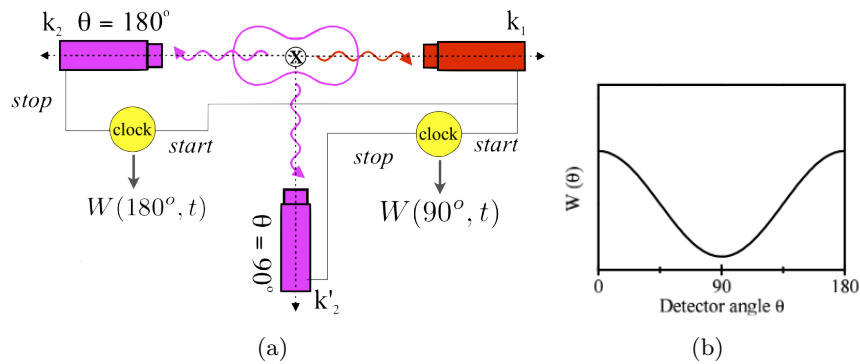


Figure 3.0.2.: (a) Experimental setup to measure $\gamma - \gamma$ angular correlations for detectors with angles of 180° and 90° between them. (b) Illustrative representation of $W(\theta)$.

3.1. General Theory of $\gamma - \gamma$ Angular Correlation (non-perturbed)

Let's consider a decay from an initial state $|I_i, m_i\rangle$ to an intermediate state $|I, m\rangle$ with the emission of a photon γ_1 , followed by a decay from $|I, m\rangle$ to a final state $|I_f, m_f\rangle$ with the emission of a photon γ_2 .

The transition amplitudes of emission of each photon are given by

$$\begin{aligned} \langle I, m, \mathbf{k}_1, \sigma_1 | \hat{H}_1 | I_i, m_i \rangle \\ \langle I_f, m_f, \mathbf{k}_2, \sigma_2 | \hat{H}_2 | I, m \rangle \end{aligned} \quad (3.1.1)$$

where $\mathbf{k}_i$ is the wave vector and σ_i is the polarization of the photon γ_i , and $\hat{H}_i$ is the interaction Hamiltonian for the emission of γ_i .

For a more easy writing, we will represent it as

$$\begin{aligned} \langle m | \hat{H}_1 | m_i \rangle \\ \langle m_f | \hat{H}_2 | m \rangle \end{aligned} \quad (3.1.2)$$

The unperturbed angular correlation will then be given by (ref.[3])

$$W(\mathbf{k}_1, \mathbf{k}_2) = \sum_{m_i, m_f, \sigma_1, \sigma_2} \left| \sum_m \langle m_f | \hat{H}_2 | m \rangle \langle m | \hat{H}_1 | m_i \rangle \right|^2 \quad (3.1.3)$$

It is important to notice that the intermediate state m is not observable, so we sum its transition amplitudes and not its transition probabilities. Initial and final states are observable, so we sum its transition probabilities.

Now, we can write eq.(3.1.3) in the following form:

$$W(\mathbf{k}_1, \mathbf{k}_2) = \sum_{m_i, m_f, m, m', \sigma_1, \sigma_2} \langle m_f | \hat{H}_2 | m \rangle \langle m | \hat{H}_1 | m_i \rangle \langle m' | \hat{H}_1 | m_i \rangle^* \langle m_f | \hat{H}_2 | m' \rangle^* \quad (3.1.4)$$

The Density Matrix

A very suitable tool for dealing with the theory of perturbation of angular correlations is the density matrix which is introduced to describe situations where we do not have an ensemble of pure quantum states, but rather “mixed” states (ref.[9]).

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

Assume that the eigenstates of a certain operator form a complete orthonormal set $|\varphi\rangle$. A pure state $|\Psi\rangle$ is given by the expansion

$$|\Psi\rangle = \sum_{\varphi} |\varphi\rangle \langle\varphi|\Psi\rangle \quad (3.1.5)$$

where, as usual, the expansion coefficients $\langle\varphi|\Psi\rangle$ satisfy the relationship

$$\langle\varphi|\Psi\rangle = \langle\Psi|\varphi\rangle^*. \quad (3.1.6)$$

The expectation value of any operator $\hat{a}$ in the state $|\Psi\rangle$ is given by

$$\langle\Psi|\hat{a}|\Psi\rangle = \sum_{\varphi, \varphi'} \langle\Psi|\varphi'\rangle \langle\varphi'|\hat{a}|\varphi\rangle \langle\varphi|\Psi\rangle. \quad (3.1.7)$$

If we have N possible different pure states $|\Psi_n\rangle$, with weights $g(n)$, the expectation value $\langle\hat{a}\rangle$ of the operator a is

$$\langle\hat{a}\rangle = \sum_{n=1}^N g(n) \langle\Psi_n|\hat{a}|\Psi_n\rangle \quad (3.1.8)$$

with the weights normalized by

$$\sum_{n=1}^N g(n) = 1 \quad (3.1.9)$$

and, of course, they are real numbers

$$0 \leq g(n) \leq 1. \quad (3.1.10)$$

Substituting (3.1.7) into (3.1.8), one can write

$$\langle\hat{a}\rangle = \sum_{n, \varphi, \varphi'} \langle\varphi|\Psi_n\rangle g(n) \langle\Psi_n|\varphi'\rangle \langle\varphi'|\hat{a}|\varphi\rangle \quad (3.1.11)$$

or

$$\langle\hat{a}\rangle = \sum_{\varphi, \varphi'} \langle\varphi|\hat{\rho}|\varphi'\rangle \langle\varphi'|\hat{a}|\varphi\rangle \quad (3.1.12)$$

where the matrix ρ with the elements

$$\langle\varphi|\hat{\rho}|\varphi'\rangle = \sum_{n=1}^N \langle\varphi|\Psi_n\rangle g(n) \langle\Psi_n|\varphi'\rangle \quad (3.1.13)$$

has been introduced. The state of the ensemble is completely characterized by the matrix ρ , whereas the matrix A with the elements $\langle\varphi'|\hat{a}|\varphi\rangle$ of the operator $\hat{a}$ depends *only* on the operator $\hat{a}$ and on the choice of the representation $\{\varphi\}$.

3.1. General Theory of $\gamma - \gamma$ Angular Correlation (non-perturbed)

The matrix ρ is the *density matrix* of the ensemble in the $\{\varphi\}$ representation. The density matrix is independent of the relative phases of the states labeled by different Ψ_n , since these phases cancel in the products $\langle \varphi | \Psi_n \rangle g(n) \langle \Psi_n | \varphi' \rangle$. This expresses the fact that the density matrix describes the ensemble in terms of inchoerent different states $|\Psi_n\rangle$.

The density matrix, ρ , provides all the significant information about the physical properties of the ensemble, since the average value of any observable represented by the operator $\hat{a}$ can be computed from eq. (3.1.12). This important equation can also be written in the more compact form

$$\langle \hat{a} \rangle = Tr(\rho A) \quad (3.1.14)$$

where the trace operation Tr corresponds to the sum of the diagonal elements of the product matrix ρA . Naturally, ρ and A must be in the same representation. A straighforward calculation shows that

$$Tr(\rho A) = Tr(A\rho). \quad (3.1.15)$$

It is a well known fact that the trace of a matrix is invariant under a unitary transformation U ($U^{-1} = U^\dagger$)

$$Tr(\rho A) = Tr(U(\rho A)U^\dagger). \quad (3.1.16)$$

Hence

$$Tr(\rho A) = Tr(U\rho U^\dagger UAU^\dagger) = Tr(\rho' A') \quad (3.1.17)$$

where

$$\rho' = U\rho U^\dagger \quad (3.1.18)$$

$$A' = UAU^\dagger \quad (3.1.19)$$

i.e., the trace of ρA is independent of the representation used for the matrices ρ and A .

The probability $P(\varphi)$ that any part of the ensemble is in a given state $|\varphi\rangle$ is given by

$$P(\varphi) = \sum_{n=1}^N \langle \varphi | n \rangle g(n) \langle n | \varphi \rangle = \langle \varphi | \hat{\rho} | \varphi \rangle \quad (3.1.20)$$

i.e., by the diagonal element $\langle \varphi | \hat{\rho} | \varphi \rangle$ of the density matrix in the representation $\{\varphi\}$. From its construction (eq. 3.1.13), it is obvious that $\hat{\rho}$ is Hermitian

$$\hat{\rho}^\dagger = \hat{\rho}. \quad (3.1.21)$$

The density matrix can be considered as a representation of the density operator

$$\hat{\rho} = \sum_{n=1}^N |\Psi_n\rangle g(n) \langle \Psi_n| \quad (3.1.22)$$

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

in the basis $\{\varphi\}$. The density operator $\hat{\rho}$ completely describes the state of the ensemble, whether the ensemble is in a mixed state (at least two $g(n)$ different from zero) or in a pure state (only one $g(n)$ non-vanishing).

Since the density matrix ρ is Hermitian, it can be diagonalized by a unitary transformation. In this diagonal representation, the basis states $\{\Psi\}$ can be chosen to be orthogonal and the density matrix is diagonal

$$\rho = \begin{pmatrix} g(1) & & & 0 \\ & \ddots & & \\ & & g(i) & \\ & & & \ddots \\ 0 & & & & g(N) \end{pmatrix} \quad (3.1.23)$$

because then

$$\begin{aligned} \langle \Psi_i | \hat{\rho} | \Psi_k \rangle &= \sum_{n=1}^N \langle \Psi_i | \Psi_n \rangle g(n) \langle \Psi_n | \Psi_k \rangle \\ &= \sum_{n=1}^N \delta_{in} g(n) \delta_{nk} = g(i) \delta_{ik}. \end{aligned} \quad (3.1.24)$$

The diagonal elements $g(i)$ represent the probability of finding a particle or quantum of the ensemble in the state $|\Psi_i\rangle$. The $g(i)$ are called the *populations* of the pure states $|\Psi_i\rangle$. Because all $g(i)$ are normalized, we have

$$\text{Tr } \rho = 1 \quad (3.1.25)$$

in any representation.

3.1.1. The Angular Correlation Function

Going back to the Angular Correlations, and applying the definition of density matrices, we get

$$\begin{aligned} \langle m | \hat{\rho}(\mathbf{k}_1) | m' \rangle &= \sum_{m_i} \langle m | \hat{H}_1 | m_i \rangle \langle m' | \hat{H}_1 | m_i \rangle^* \\ \langle m' | \hat{\rho}(\mathbf{k}_2) | m \rangle &= \sum_{m_f} \langle m_f | \hat{H}_2 | m \rangle \langle m_f | \hat{H}_2 | m' \rangle^* \end{aligned} \quad (3.1.26)$$

so, we can re-write the correlation function (3.1.4) as

$$W(\mathbf{k}_1, \mathbf{k}_2) = \sum_{m, m'} \langle m | \hat{\rho}(\mathbf{k}_1) | m' \rangle \langle m' | \hat{\rho}(\mathbf{k}_2) | m \rangle \quad (3.1.27)$$

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or simply

$$W(\mathbf{k}_1, \mathbf{k}_2) = \text{Tr} \{ \hat{\rho}(\mathbf{k}_2) \hat{\rho}(\mathbf{k}_1, 0) \} \quad (3.1.28)$$

Matrix Elements of the Type $\langle m | \hat{H} | m_i \rangle$

The matrix element $\langle m | \hat{H} | m_i \rangle \equiv \langle Im\mathbf{k}\sigma | \hat{H} | I_i m_i \rangle$ is represented in terms of the nuclear spins and the direction $\mathbf{k}$ and polarization σ of the emitted radiations. This representation is natural for the description of the observed radiation properties, but it does not provide us the desired information about the nucleus. Instead of linear momentum ($\hbar k$) and polarization (σ), we would like to represent the radiation in terms of its angular momentum (L) and its parity (π). These properties, described by the quantum numbers L , M and π , yield the desired data about the nuclear transition, since $I_i - I = L$, and for parity conserving radiation, $\pi_i \times \pi = \pi_{rad}$ (ref.[10]).

The connection between the 'observed' matrix element $\langle Im\mathbf{k}\sigma | \hat{H} | I_i m_i \rangle$ and the 'desired' matrix element $\langle ImLM\pi | \hat{H} | I_i m_i \rangle$ is then given by

$$\langle Im\mathbf{k}\sigma | \hat{H} | I_i m_i \rangle = \sum_{LM} \langle \mathbf{k}\sigma | LM\pi \rangle \langle ImLM\pi | \hat{H} | I_i m_i \rangle \quad (3.1.29)$$

The parity is conserved so we don't need to sum in π .

Now, let's analyse the factors on the right hand side of eq.(3.1.29) separately.

The first factor, $\langle \mathbf{k}\sigma | LM\pi \rangle$ describes the eigenfunction of the γ -radiation corresponding to eigenvalues L , M and π , of the operators of angular momentum, z-component of angular momentum and parity, respectively, and can be written as (ref.[10])

$$\langle \mathbf{k}\sigma | LM\pi \rangle = \sum_{\mu} \langle 0\sigma | L\mu\pi \rangle D_{M\mu}^{L*}(\mathbf{z} \rightarrow \mathbf{k}) \quad (3.1.30)$$

where $D_{M\mu}^L \equiv \langle \mu | D^L | M \rangle$ are the elements of a unitary matrix, called the rotation matrix. The argument ($\mathbf{z} \rightarrow \mathbf{k}$) of the rotation matrix D^L stands for the Euler angles (ϕ, θ, γ) (ref.[11]) describing the rotation that carries the arbitrary quantization system $\mathbf{z}$ over into the coordinate system of the radiation (assuming that the z-axis coincides with the direction $\mathbf{k}$ of emission of the γ -radiation we have $\theta = 0$); ($\mathbf{k} \rightarrow \mathbf{z}$) denotes the inverse rotation. Some important properties of $D_{M\mu}^L$ are presented in Appendix A.3.

The second factor in eq.(3.1.29), the matrix element $\langle ImLM\pi | \hat{H} | I_i m_i \rangle$ corresponds to the vector addition $I_i = I + L$, $m_i = m + M$. Because $\langle ImLM\pi | \equiv \langle Im | \langle LM\pi |$, using the properties of the Clebsch-Gordan coefficients we have

$$\langle ImLM\pi | \hat{H} | I_i m_i \rangle = \sum_{I'_i m'_i} \langle ImLM | I'_i m'_i \rangle \langle I'_i m'_i | IL\pi | \hat{H} | I_i m_i \rangle \quad (3.1.31)$$

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

We now use the fact that $\hat{H}$ for physical reasons is invariant under rotations and so the matrix element $\langle I'_i m'_i I L \pi | \hat{H} | I_i m_i \rangle$ is different from zero only if $I'_i = I_i$, $m'_i = m_i$, and furthermore it is independent of the magnetic quantum number m_i (ref.[10]), thus from the Wigner-Eckart theorem (A.2) we get

$$\langle I m L M \pi | \hat{H} | I_i m_i \rangle = \langle I m L M | I_i m_i \rangle \langle I || L \pi || I_i \rangle \quad (3.1.32)$$

defining the reduced matrix element $\langle I || L \pi || I_i \rangle$ of a scalar operator $\hat{H}$. If the interaction that induces the transition can be described by a Hamiltonian that commutes with the operator of time reversal, the reduced matrix elements may be chosen to be real

$$\langle I || L \pi || I_i \rangle^* = \langle I || L \pi || I_i \rangle. \quad (3.1.33)$$

Combining equations (3.1.29), (3.1.30), (3.1.32) and replacing the Clebsch-Gordan coefficients by the more symmetric Wigner 3-j symbol (appendix A.1), we get the final expression for a typical matrix element

$$\langle m | \hat{H} | m_i \rangle = \sum_{LM\mu} (-1)^{-I+L-m_1} \begin{pmatrix} I & L & I_i \\ m & M & -m_i \end{pmatrix} \langle 0\sigma | L\mu\pi \rangle \langle I || L\pi || I_i \rangle D_{M\mu}^{L*}(\mathbf{z} \rightarrow \mathbf{k}). \quad (3.1.34)$$

Matrix Elements of the Type $\langle m | \hat{\rho}(\mathbf{k}) | m' \rangle$

We turn now to expressions (3.1.26) for the matrix elements of the density operators $\hat{\rho}(\mathbf{k})$. Inserting the elements $\langle m | \hat{H} | m_i \rangle$ of eq.(3.1.34) into eq.(3.1.26), simplifying the result using eq.(A.3.4) and the properties of the Clebsch-Gordan coefficients and, after replacing the Clebsch-Gordan coefficients by 3-j symbols (A.1), we get

$$\begin{aligned} \langle m | \hat{\rho}(\mathbf{k}) | m' \rangle &= \sum_{m_i} \sum_{LM\mu L' M' \mu' k N \tau} (-1)^{M'-\mu'} (2k+1) \langle 0\sigma | L\mu\pi \rangle \langle 0\sigma' | L'\mu'\pi' \rangle^* \times \\ &\times \begin{pmatrix} I & L & I_i \\ m & M & -m_i \end{pmatrix} \begin{pmatrix} I & L' & I_i \\ m' & M' & -m_i \end{pmatrix} \begin{pmatrix} L & L' & k \\ M & -M' & N \end{pmatrix} \begin{pmatrix} L & L' & k \\ \mu & -\mu' & \tau \end{pmatrix} \times \\ &\times \langle I || L\pi || I_i \rangle \langle I || L'\pi' || I_i \rangle^* D_{N\tau}^k(\mathbf{z} \rightarrow \mathbf{k}). \end{aligned} \quad (3.1.35)$$

Here, we have set $N = M' - M$, $\tau = \mu' - \mu$, and to get the sign we used the fact that $L' - L$, N , τ , $2I$, $2m_i$ and $I + L + m_i$ are integers.

The sum over m_i , M , and M' over the product of the first three 3j-symbols can be evaluated by using eq.(A.1.11), together with the symmetry relations given in section A.1 and with

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$m_i = m + M$. The result is

$$\begin{aligned} \langle m \mid \hat{\rho}(\mathbf{k}) \mid m' \rangle &= \sum_{L\mu L'\mu'} \sum_{kN\tau} (-1)^{2I-I_i+m-\mu'} (2k+1) \langle 0\sigma \mid L\mu\pi \rangle \langle 0\sigma' \mid L'\mu'\pi' \rangle^* \times \\ &\times \begin{pmatrix} L & L' & k \\ \mu & -\mu' & \tau \end{pmatrix} \begin{pmatrix} I & I & k \\ m' & -m & N \end{pmatrix} \left\{ \begin{matrix} I & I & k \\ L & L' & I_i \end{matrix} \right\} \\ &\times \langle I \parallel L\pi \parallel I_i \rangle \langle I \parallel L'\pi' \parallel I_i \rangle^* D_{N\tau}^k(\mathbf{z} \rightarrow \mathbf{k}) \end{aligned} \quad (3.1.36)$$

This expression contains factors that depend only on the properties of the radiation. To single out these factors clearly, we introduce the Racah radiation parameters $c_{k\tau}(LL')$, defined by

$$c_{k\tau}(LL') = \sum_{\mu\mu'} (-1)^{L-\mu} (2k+1)^{\frac{1}{2}} \begin{pmatrix} L & L' & k \\ \mu & -\mu' & -\tau \end{pmatrix} \langle 0\sigma \mid L\mu\pi \rangle^* \langle 0\sigma' \mid L'\mu'\pi' \rangle \quad (3.1.37)$$

These parameters satisfy the relation

$$c_{k\tau}(LL') = (-1)^{L-L'-\tau} c_{k-\tau}^*(L'L). \quad (3.1.38)$$

They are characteristic of the emitted radiation through the transformation coefficients $\langle 0\sigma \mid L\mu\pi \rangle$. Formally, the $c_{k\tau}(LL')$ are eigenfunctions of the operators for the total angular momentum and the z-component of the angular momentum with eigenvalues k and τ .

Introducing the radiation parameters (3.1.37) into (3.1.36) and using the relation (3.1.38) yields for the matrix element

$$\begin{aligned} \langle m \mid \hat{\rho}(\mathbf{k}) \mid m' \rangle &= \sum_{LL'} \sum_{kN\tau} (-1)^{2I-I_i+m-L'} (2k+1)^{\frac{1}{2}} c_{k\tau}(LL') \times \\ &\times \begin{pmatrix} I & I & k \\ m' & -m & N \end{pmatrix} \left\{ \begin{matrix} I & I & k \\ L & L' & I_i \end{matrix} \right\} \langle I \parallel L\pi \parallel I_i \rangle \langle I \parallel L'\pi' \parallel I_i \rangle^* D_{N\tau}^k(\mathbf{z} \rightarrow \mathbf{k}). \end{aligned} \quad (3.1.39)$$

Finally, The Unperturbed Correlation Function $W(\mathbf{k}_1, \mathbf{k}_2)$

The matrix element $\langle m \mid \hat{\rho}(\mathbf{k}_1) \mid m' \rangle$ for the first step of an unperturbed cascade decay $I_i \rightarrow I \rightarrow I_f$ is given by the expression (3.1.39) with the proper subscripts. For the matrix element $\langle m' \mid \hat{\rho}(\mathbf{k}_2) \mid m \rangle$, one finds in the same way

$$\begin{aligned} \langle m' \mid \hat{\rho}(\mathbf{k}_2) \mid m \rangle &= \sum_{L_2 L'_2} \sum_{k_2 N_2 \tau_2} (-1)^{k_2 - I_f - m - L'_2} (2k_2 + 1)^{\frac{1}{2}} c_{k_2 \tau_2}^*(L_2 L'_2) \times \\ &\times \begin{pmatrix} I & I & k_2 \\ m' & -m & N_2 \end{pmatrix} \left\{ \begin{matrix} I & I & k_2 \\ L_2 & L'_2 & I_f \end{matrix} \right\} \langle I_f \parallel L_2 \pi_2 \parallel I \rangle \langle I_f \parallel L'_2 \pi'_2 \parallel I \rangle^* D_{N_2 \tau_2}^{k_2*}(\mathbf{z} \rightarrow \mathbf{k}_2). \end{aligned} \quad (3.1.40)$$

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The expressions (3.1.39) and (3.1.40) can now be inserted into eq.(3.1.27) for $W(\mathbf{k}_1, \mathbf{k}_2)$. The resulting formula can be simplified by the orthogonality relations of the 3-j symbols (A.1). With $k_1 = k_2 \equiv k$, and $N_1 = N_2 \equiv N$ it becomes possible to combine the two representations D^{k_1} and D^{k_2*} by using eq.(A.3.3). The final expression for the unperturbed angular correlation between two successive radiations thus is

$$\begin{aligned} W(\mathbf{k}_1, \mathbf{k}_2) = & (-1)^{2I-I_i-I_f} \sum_k \sum_{L_1 L'_1 L_2 L'_2} \sum_{\tau_1 \tau_2} (-1)^{k-L'_1-L'_2} \left\{ \begin{matrix} I & I & k \\ L_1 & L'_1 & I_i \end{matrix} \right\} \left\{ \begin{matrix} I & I & k \\ L_2 & L'_2 & I_f \end{matrix} \right\} \times \\ & \times \langle I_f \| L_2 \pi_2 \| I \rangle \langle I_f \| L'_2 \pi'_2 \| I \rangle^* \langle I \| L_1 \pi_1 \| I_i \rangle \langle I \| L'_1 \pi'_1 \| I_i \rangle^* \times \\ & \times c_{k\tau_1}(L'_1 L_1) c_{k\tau_2}^*(L_2 L'_2) D_{\tau_2 \tau_1}^k(\mathbf{k}_2 \rightarrow \mathbf{k}_1). \end{aligned} \quad (3.1.41)$$

The rotation $\mathbf{k}_2 \rightarrow \mathbf{k}_1$ carries the coordinate system of the second radiation (given by the propagation direction and the polarization of γ_2) into that of the first one.

Due to the properties of the 6-j symbols, we get the following restrictions to the sum in k

$$0 < k < \text{Min}(2I, L_1 + L'_1, L_2 + L'_2) \quad (3.1.42)$$

or for pure multipole radiation

$$0 < k < \text{Min}(2I, 2L_1, 2L_2). \quad (3.1.43)$$

Besides, the summation index k is an even integer as long as the measurements are restricted to the directions and the linear polarizations of the radiations (ref.[10] p.1023).

Experimentally we usually observe directions only, but not polarizations of the γ_1 -radiation and γ_2 -radiation. In this case, the correlation function must be independent of the Euler angles $(\gamma)_1$ and $(\gamma)_2$ (not to be confused with γ_1 and γ_2), which denote rotations about the directions of propagation $\mathbf{k}_1$ and $\mathbf{k}_2$. Only the functions c_{k0} then appear in $W(\mathbf{k}_1, \mathbf{k}_2)$, as can be seen immediately: $c_{k\tau}$ is an eigenfunction of the operator $L_z = -i\hbar(\partial/\partial\gamma)$, with eigenvalue τ . Independence of γ thus means $\tau = 0$.

With $\tau_1 = \tau_2 = 0$, from eq.(A.3.7) the representation D^k become Legendre Polynomials, and using the A'_{kk} coefficients defined on ref.[10] (p.1024) as

$$A'_{kk} = A'_k(L_1 L'_1 I_i I) A'_k(L_2 L'_2 I_f I) \quad (3.1.44)$$

where

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$$\begin{aligned} A'_k(L_1 L'_1 I_i I) &= \sum_{L_1 L'_1} (-1)^{L_1} c_{k0}^*(L_1 L'_1) \left\{ \begin{matrix} I & I & k \\ L_1 & L'_1 & I_i \end{matrix} \right\} \langle I \| L_1 \pi_1 \| I_i \rangle \langle I \| L'_1 \pi'_1 \| I_i \rangle^* \\ A'_k(L_2 L'_2 I_f I) &= \sum_{L_2 L'_2} (-1)^{L_2} c_{k0}^*(L_2 L'_2) \left\{ \begin{matrix} I & I & k \\ L_2 & L'_2 & I_f \end{matrix} \right\} \langle I \| L_2 \pi_2 \| I_f \rangle^* \langle I \| L'_2 \pi'_2 \| I_f \rangle \end{aligned} \quad (3.1.45)$$

the expression (3.1.41) simplifies to

$$W(\mathbf{k}_1, \mathbf{k}_2) = W(\theta) = \sum_{k \text{ even}} A'_{kk} P_k(\cos \theta) \quad (3.1.46)$$

being θ the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$.

The radiation parameters $c_{k\tau}(LL')$ are defined by eq.(3.1.37) and can be calculated by several ways (two of them are noted in ref. [10]). To point their physical meaning let's start from eq.(3.1.29) and note that only the factor $\langle \mathbf{k}\sigma | LM\pi \rangle$ on the right hand side depends on the direction of emission $\mathbf{k}$ of the γ -ray. The directional distribution function $F_L^M(\theta)$, defined by eq.(2.3.21), is proportional to the square of the transition probability $\langle Im\mathbf{k}\sigma | \hat{H} | I_i m_i \rangle$ and hence can be written as

$$F_L^M(\theta) \equiv F_L^M(\mathbf{k}) \propto \langle \mathbf{k}\sigma | LM\pi \rangle \langle \mathbf{k}\sigma | LM\pi \rangle^*. \quad (3.1.47)$$

The radiation parameters thus are connected to the functions F_L^M by²

$$c_{k0}(LL) \propto \sum_{\mu} (-1)^{L-\mu} \sqrt{2k+1} \begin{pmatrix} L & L & k \\ \mu & -\mu & 0 \end{pmatrix} F_L^M(0). \quad (3.1.48)$$

For mixed multipole transitions, we get the result (see ref.[10])

$$c_{k0}(LL') = (-1)^{L-1} \sqrt{2L+1} \sqrt{2L'+1} \sqrt{2k+1} \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix} \quad (3.1.49)$$

where, as previously said, only even k appear.

3.1.2. Anisotropy Coefficients A_{kk}

We have just seen that, by definition, the anisotropy coefficients A'_{kk} are given by expressions (3.1.45), which are not normalized. In applications, they are usually chosen so that $A_{00} = 1$. The normalized form is easily obtained by dividing each coefficient A'_{kk} by A'_{00} .

²Remember that because we only observe directions and not polarizations, we are only interested in the solutions of $c_{k\tau}(LL')$ with $\tau = 0$.

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

If we consider a $\gamma - \gamma$ cascade $I_i \rightarrow I \rightarrow I_f$ in which both γ -rays, of multipole order L_1 and L_2 , respectively, are pure, we have

$$A_{kk}(\text{pure}) = F_k(L_1 L_1 I_i I) F_k(L_2 L_2 I_f I) \quad (3.1.50)$$

where we introduced the coefficients F

$$F_k(LLI_i I) = \frac{A'_k(LLI_i I)}{A'_0(LLI_i I)} \quad (3.1.51)$$

given by (ref.[12])

$$F_k(LLI_i I) = (-1)^{I_i+I-1} (2L+1) \sqrt{2I+1} \sqrt{2k+1} \begin{pmatrix} L & L & k \\ 1 & -1 & 0 \end{pmatrix} \left\{ \begin{matrix} L & L & k \\ I & I & I_i \end{matrix} \right\} \quad (3.1.52)$$

In ref.[12] we can find tables of frequently used F -coefficients.

Now, let's consider a $\gamma - \gamma$ cascade $I_i \rightarrow I \rightarrow I_f$ in which two multipole components L_n and L'_n contribute to each of the two γ -transitions. Only two types of mixed γ -transitions have so far been observed experimentally, the rather frequent $M1 + E2$ and the rare $E1 + M2$ transitions³.

We define the (amplitude) mixing ratio of the transition 1 as the ratio of the reduced matrix elements

$$\delta_1(\gamma) \equiv \frac{\langle I || L'_1 \pi'_1 || I_i \rangle}{\langle I || L_1 \pi_1 || I_i \rangle} \quad (3.1.53)$$

and similarly for the second transition. The ratio of the total intensity of the L' -pole to that of the L -pole is then equal to δ^2 .

To write the directional correlation for mixed transitions in a convenient form, we generalize the F -coefficients to read (ref.[12])

$$F_k(LL'I_i I) = (-1)^{I_i+I-1} \sqrt{(2L+1)(2L'+1)(2I+1)(2k+1)} \begin{pmatrix} L & L' & k \\ 1 & -1 & 0 \end{pmatrix} \left\{ \begin{matrix} L & L' & k \\ I & I & I_i \end{matrix} \right\} \quad (3.1.54)$$

These coefficients satisfy

$$F_0(LL'I_i I) = \delta_{LL'}. \quad (3.1.55)$$

The normalized coefficients A_{kk} then can be written as

$$A_{kk} = A_k(L_1 L'_1 I_i I) A_k(L_2 L'_2 I_f I) \quad (3.1.56)$$

³ $M1$ relates to magnetic dipole transition, $E2$ to electric quadrupole transition, $E1$ to electric dipole transition and $M2$ magnetic quadrupole transition

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and one obtains easily from eqs. (3.1.45), (3.1.49), (3.1.54) and (3.1.55)

$$A_k(L_1 L'_1 I_i I) = \frac{F_k(L_1 L_1 I_i I) + 2\delta_1(\gamma)F_k(L_1 L'_1 I_i I) + \delta_1^2(\gamma)F_k(L'_1 L'_1 I_i I)}{1 + \delta_1^2(\gamma)} \quad (3.1.57)$$

and similar expression for $A_k(L_2 L'_2 I_f I)$.

The coefficients $F_k(LL'I_i I)$ are tabulated in ref.[12].

With the normalized coefficients A_{kk} , the directional correlation $W(\theta)$ between γ_1 and γ_2 given by eq.(3.1.46) is represented as

$$W(\mathbf{k}_1, \mathbf{k}_2) = W(\theta) = 1 + A_{22}P_2(\cos \theta) + \dots + A_{k_{max}k_{max}}P_{k_{max}}(\cos \theta). \quad (3.1.58)$$

In an experimental investigation of $\gamma - \gamma$ directional correlation we use, at best, $A_{k_{max}k_{max}} = A_{44}$, because the energies that normally are available in radioactive decays are such that multipoles with $L > 2$ possess a half life $> 10^{-6}sec$ and it is experimentally difficult (or even impossible) to measure a directional correlation function of a cascade involving an intermediate state with so long a half-life (ref.[10]).

3.1.3. From $\gamma - \gamma$ to $e^- - \gamma$ correlations

Instead of measuring the directional correlation between successive γ -rays, one can detect the corresponding conversion electrons and measure their correlation. This procedure becomes feasible if at least one of the γ -rays is moderately converted.

In contrast with $\gamma - \gamma$ correlation, the conversion correlation depends not only on the spins of the nuclear levels and the multipole orders of the transition, but also on the parity changes and the energies of the converted transitions and on the nuclear charge. The conversion correlation thus may yield more information than the $\gamma - \gamma$ directional correlation.

The detection of a e^- -particle instead of a γ -particle results in the introduction of a particle parameter in the correlation function.

Conveniently adopting the $\gamma - \gamma$ angular correlation function as the standard representation of the 'geometry' of the correlation process and remembering that the nature of the radiation entered only through the Racah radiation parameters $c_{k0}(LL')$, the particle parameter is defined as (ref.[10], ref.[12])

$$b_k(LL'; e^-) = \frac{c_{k0}(LL'; e^-)}{c_{k0}(LL'; \gamma)} \quad (3.1.59)$$

where we denoted the radiation parameter $c_{k0}(LL')$ for the emission of a "radiation" x ($x = e^-, \gamma$) as $c_{k0}(LL'; x)$.

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

For a mixed converted transition, from ref.[10], we have that the anisotropy coefficients $A_k(LL'I_iI)$ given by eq.(3.1.57) for a transition with emission of a e^- -particle will then be

$$A_k(LL'I_iI; e^-) = \frac{b_k(LL)F_k(LLI_iI) + 2\delta(e^-)b(LL')F(LL'I_iI) + \delta^2(e^-)b(L'L')F(L'L'I_iI)}{1 + \delta^2(e^-)} \quad (3.1.60)$$

with

$$\delta(e^-) = \delta(\gamma) \sqrt{\frac{c(L'\pi)}{c(L\pi)}} \quad (3.1.61)$$

$c(L'\pi)$ and $c(L\pi)$ being conversion coefficients of the two competing multipoles. The conversion coefficients are defined as $c(L\pi) = \frac{N(e^-)}{N(\gamma)}$, where $N(e^-)$, $N(\gamma)$ represent the probabilistic intensities of emission of a conversion electron or of emission of a γ in the transition from an excited state, respectively.

Particle parameters $b_k(LL'; e^-)$ and conversion coefficients $c(L\pi)$ are tabulated in ref.[12].

3.2. Theory of Perturbed $\gamma - \gamma$ Angular Correlation

Now we must consider that the intermediate state has a certain mean-life time, τ . This means that, if the nuclei are under the influence of hyperfine interactions, this may cause repopulations or state transitions before the second photon is emitted. This time evolution is represented by the propagation operator $\hat{\Lambda}(t)$, changing the intermediate state, $|m\rangle \rightarrow \hat{\Lambda}(t)|m\rangle$.

But let's take a better look at the temporal behavior of this process.

The time dependence of a density operator $\hat{\rho}(t)$ is governed by the von Neumann equation (ref.[13])

$$\frac{d\hat{\rho}(t)}{dt} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}(t)] \quad (3.2.1)$$

where $\hat{H}$ is the total Hamiltonian of the system. Note that the sign of the commutator is opposite to that of the corresponding equation for operators of observables (Heisenberg equation). The relation (3.2.1) is the equivalent of the time-dependent Schrödinger equation.

In fact, (3.2.1) can be derived from the Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} |\psi\rangle = \hat{H} |\psi\rangle. \quad (3.2.2)$$

The general solution of (3.2.2) can be expressed in the form

$$|\psi(t)\rangle = \hat{\Lambda}(t, t_0) |\psi(t_0)\rangle \quad (3.2.3)$$

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where $\hat{\Lambda}(t)$ is the unitary time-evolution operator. From the two previous equations, it is obvious that the operator $\hat{\Lambda}(t, t_0)$ must also satisfy the Schrödinger equation

$$\frac{\partial}{\partial t} \hat{\Lambda}(t, t_0) = -\frac{i}{\hbar} \hat{H} \hat{\Lambda}(t, t_0). \quad (3.2.4)$$

The time behaviour of the density operator (3.1.22) is thus given by

$$\hat{\rho}(t) = \sum_n \hat{\Lambda}(t, t_0) | \psi_n(t_0) \rangle g_n \langle \psi_n(t_0) | \hat{\Lambda}^\dagger(t, t_0) = \hat{\Lambda}(t, t_0) \hat{\rho}(t_0) \hat{\Lambda}^\dagger(t, t_0). \quad (3.2.5)$$

Taking the time derivative of $\hat{\rho}(t)$, one obtains

$$\begin{aligned} \frac{d\hat{\rho}(t)}{dt} &= \frac{\partial \hat{\Lambda}}{\partial t} \hat{\rho}(t_0) \hat{\Lambda}^\dagger + \hat{\Lambda} \hat{\rho}(t_0) \frac{\partial \hat{\Lambda}^\dagger}{\partial t} \\ &= -\frac{i}{\hbar} \hat{H} \hat{\Lambda} \hat{\rho}(t_0) \hat{\Lambda}^\dagger + \frac{i}{\hbar} \hat{\Lambda} \hat{\rho}(t_0) \hat{\Lambda}^\dagger \hat{H} \\ &= -\frac{i}{\hbar} [\hat{H}, \hat{\rho}(t)] \end{aligned} \quad (3.2.6)$$

because of (3.2.5).

The fundamental equation of motion (3.2.1) is particularly important in the interaction representation. The total Hamiltonian is

$$\hat{H} = \hat{H}_0 + \hat{H}_{int} \quad (3.2.7)$$

where $\hat{H}_0$ is responsible for the existence of the stationary states (nuclear states and radiation field) whereas $\hat{H}_{int}$ is responsible for inducing transitions between the eigenstates of $\hat{H}_0$. The interaction representation is now introduced by the transformations

$$\hat{H}_I(t) = \exp(i\hat{H}_0 t) \hat{H}_{int} \exp(-i\hat{H}_0 t) \quad (3.2.8)$$

and

$$\hat{\Lambda}_I(t, t_0) = \exp(i\hat{H}_0 t) \hat{\Lambda}(t, t_0) \exp(-i\hat{H}_0 t). \quad (3.2.9)$$

The statevectors in this representation are

$$| \psi_I(t) \rangle = \exp(i\hat{H}_0 t) | \psi(t) \rangle_{Sch} \quad (3.2.10)$$

where $| \psi(t) \rangle_{Sch}$ are the statevectors in the Schrödinger representation. In the interaction representation the von Neumann equation is

$$\frac{d\hat{\rho}_I(t)}{dt} = -\frac{i}{\hbar} [\hat{H}_I(t), \hat{\rho}_I(t)] \quad (3.2.11)$$

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and the time-evolution operator $\hat{\Lambda}_I(t, t_0)$ must satisfy the operator equation

$$\frac{\partial}{\partial t} \hat{\Lambda}_I(t, t_0) = -\frac{i}{\hbar} \hat{H}_I(t) \hat{\Lambda}_I(t, t_0) \quad (3.2.12)$$

which solution is written in the form

$$\hat{\Lambda}_I(t, t_0) = T \exp \left\{ -\frac{i}{\hbar} \int_{t_0}^t \hat{H}_I(t) dt \right\} \quad (3.2.13)$$

where T is the time-ordering operator, which defines the explicit expansion of the exponential, i.e.

$$\hat{\Lambda}_I(t, t_0) = 1 + \left(-\frac{i}{\hbar} \right) \int_{t_0}^t \hat{H}_I(t') dt' + \left(-\frac{i}{\hbar} \right)^2 \int_{t_0}^t \hat{H}_I(t') dt' \int_{t_0}^{t'<t} \hat{H}_I(t'') dt'' + \dots \quad (3.2.14)$$

The different-order terms in this expression correspond to the different orders of perturbation theory.

Note: Because the interaction Hamiltonian is the responsible for all the behaviour we want to study, everything is done in the interaction representation and, for simplicity, we remove the subscript I from the operator's notation.

Now that we know the time dependence of the density operator, let's focus again on the Perturbed Angular Correlation expression.

As it was said before, $|m\rangle \rightarrow \hat{\Lambda}(t) |m\rangle$, so eq.(3.1.3) transforms into

$$W(\mathbf{k}_1, \mathbf{k}_2, t) = \sum_{m_i, m_f, \sigma_1, \sigma_2} \left| \sum_m \langle m_f | \hat{H}_2 \hat{\Lambda}(t) | m \rangle \langle m | \hat{H}_1 | m_i \rangle \right|^2 \quad (3.2.15)$$

which can be written as

$$\begin{aligned} W(\mathbf{k}_1, \mathbf{k}_2, t) &= \sum_{m_i, m_f, m, m', m_a, m'_a, \sigma_1, \sigma_2} \langle m_f | \hat{H}_2 | m_a \rangle \langle m_a | \hat{\Lambda}(t) | m \rangle \langle m | \hat{H}_1 | m_i \rangle \\ &\times \langle m_f | \hat{H}_2 | m'_a \rangle^* \langle m'_a | \hat{\Lambda}(t) | m' \rangle^* \langle m' | \hat{H}_1 | m_i \rangle^* \end{aligned} \quad (3.2.16)$$

The influence of the perturbation in the intermediate state only appears in the time-dependent terms, $\langle m_a | \hat{\Lambda}(t) | m \rangle$ and $\langle m'_a | \hat{\Lambda}(t) | m' \rangle^*$.

From eq. (3.2.5) we get

$$\hat{\rho}(\mathbf{k}_1, t) = \hat{\Lambda}(t) \hat{\rho}(\mathbf{k}_1) \hat{\Lambda}^\dagger(t) \quad (3.2.17)$$

and remembering equations (3.1.26), eq.(3.2.16) is given by

$$W(\mathbf{k}_1, \mathbf{k}_2, t) = \sum_{m, m'} \langle m | \hat{\rho}(\mathbf{k}_1, t) | m' \rangle \langle m' | \hat{\rho}(\mathbf{k}_2) | m \rangle \quad (3.2.18)$$

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or alternatively

$$W(\mathbf{k}_1, \mathbf{k}_2, t) = Tr \left\{ \hat{\rho}(\mathbf{k}_2) \hat{\Lambda}(t) \hat{\rho}(\mathbf{k}_1) \hat{\Lambda}^\dagger(t) \right\} \quad (3.2.19)$$

We restrict the discussion of the influence of extranuclear fields to directional correlations and to circular polarization correlations. With $\tau_1 = \tau_2 = 0$, eq.(A.3.6) can be used to express the D_{N0}^k as spherical harmonics $Y_k^N(\theta, \phi)$. (The definition of the angles θ and ϕ with respect to the quantization z-axis is shown in Fig.(3.2.1))

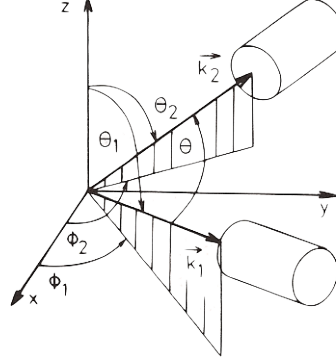


Figure 3.2.1.: Generalized coordinate system for the description of $\gamma - \gamma$ angular correlation. $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of γ_1 and γ_2 , respectively (ref.[10]).

Inserting expressions (3.1.39) and (3.1.40) for the density matrices into eq.(3.2.16), and using the definitions (3.1.45) of the coefficients A_k gives

$$W(\mathbf{k}_1, \mathbf{k}_2, t) = \sum_{k_1, k_2, N_1, N_2} A_{k_1}(1) A_{k_2}(2) G_{k_1 k_2}^{N_1 N_2}(t) [(2k_1 + 1)(2k_2 + 1)]^{-\frac{1}{2}} \times \\ \times Y_{k_1}^{N_1*}(\theta_1, \phi_1) Y_{k_2}^{N_2}(\theta_2, \phi_2) \quad (3.2.20)$$

where we define the perturbation factor

$$G_{k_1 k_2}^{N_1 N_2}(t) = \sum_{m_a, m_b} (-1)^{2I+m_a+m_b} [(2k_1 + 1)(2k_2 + 1)]^{\frac{1}{2}} \begin{pmatrix} I & I & k_1 \\ m'_a & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_b & -m_b & N_2 \end{pmatrix} \times \\ \times \langle m_b | \hat{\Lambda}(t) | m_a \rangle \langle m'_b | \hat{\Lambda}(t) | m'_a \rangle^* \quad (3.2.21)$$

The influence of extranuclear perturbation is completely described by the (in general, complex) perturbation factor $G_{k_1 k_2}^{N_1 N_2}(t)$.

For vanishing perturbation (indicated by $t \equiv 0$) the evolution matrix $\Lambda(t)$ reduces to the unit matrix and the orthogonality relations of the 3-j symbol given in appendix (A.1) give

$$G_{k_1 k_2}^{N_1 N_2}(t) \equiv \delta_{k_1 k_2} \delta_{N_1 N_2}. \quad (3.2.22)$$

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

3.2.1. Calculation of the Perturbation Factor for Static Cases

We have just reviewed the deduction of the general expression for perturbed angular correlations. However, for practical applications and for practical understanding, it is useful to derive analytic expressions for some common cases. A major simplification results if we restrict attention to static, axially symmetric interactions, and we make this restriction in the following discussion.

The time-evolution operator $\hat{\Lambda}(t)$ obeys to Schrödinger equation

$$\frac{\partial}{\partial t} \hat{\Lambda}(t) = -\frac{i}{\hbar} \hat{H} \hat{\Lambda}(t) \quad (3.2.23)$$

If $\hat{H}$ is time-independent (which is the case for static interactions), then the operator is given by

$$\hat{\Lambda}(t) = e^{-\frac{i}{\hbar} \hat{H} t}. \quad (3.2.24)$$

Choosing the z-axis in the direction of the axial field, the matrix elements of the time-evolution operator are

$$\langle m_b | \hat{\Lambda}(t) | m_a \rangle = \langle m_b | e^{-\frac{i}{\hbar} \hat{H} t} | m_a \rangle = e^{-\frac{i}{\hbar} E(m) t} \delta_{m, m_a} \delta_{m, m_b} \quad (3.2.25)$$

where $m_a = m_b = m$. A similar relation is valid for $\langle m'_b | \hat{\Lambda}(t) | m'_a \rangle$. Substituting eq.(3.2.25) in eq.(3.2.21) we obtain the perturbation factor for static axial interactions

$$G_{k_1 k_2}^{NN}(t) = \sum_m \sqrt{(2k_1 + 1)(2k_2 + 1)} \begin{pmatrix} I & I & k_1 \\ m' & -m & N \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m' & -m & N \end{pmatrix} \times \quad (3.2.26)$$

$$\times \exp \left\{ -\frac{i}{\hbar} [E(m) - E(m')] t \right\}$$

Only the energy differences between m sublevels appear in the perturbation factor, so a measurement of this factor allows one to determine these energy differences, and consequently the hyperfine interaction.

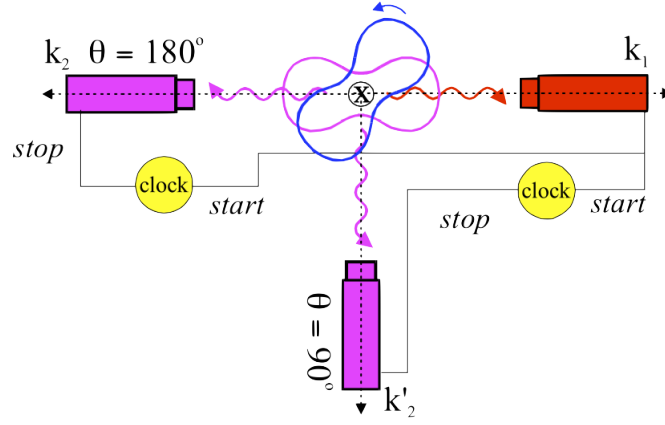


Figure 3.2.2.: Representation of a planar set of detectors. The effect of the perturbation by external fields is shown, being induced a “rotation” in the angular correlation given by the perturbation factor $G_{k_1 k_2}^{N_1 N_2}(t)$. The frequencies of rotation contain the information about the nuclei under consideration, thus the experimental purpose is to find them.

3.2.1.1. Magnetic Dipole Interaction

From eq.(2.2.34), the energy differences between the m levels due to a magnetic dipole interaction is

$$E_{\text{magn}}(m) - E_{\text{magn}}(m') = -(m - m') g \mu_N B_z = N \hbar \omega_L \quad (3.2.27)$$

with $N = m - m'$. Replacing in eq.(3.2.26) we get

$$G_{k_1 k_2}^{NN}(t) = \sum_m \sqrt{(2k_1 + 1)(2k_2 + 1)} \begin{pmatrix} I & I & k_1 \\ m' & -m & N \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m' & -m & N \end{pmatrix} e^{-iN\omega_L t} \quad (3.2.28)$$

and using the orthogonality relations of the 3-j symbols (appendix A.1), the sum over m gives $\frac{1}{\sqrt{(2k_1 + 1)(2k_2 + 1)}} \delta_{k_1 k_2}$, so we obtain for the magnetic dipole interaction

$$G_{kk}^{NN}(t) = e^{-iN\omega_L t} \quad (3.2.29)$$

N and k are summation indices with the restrictions $|N| \leq k$, and k restricted to the values shown in eq.(3.1.43). In particular, $|N| \leq 2I$ where I is the spin of the intermediate nuclear state. One sees from eq.(3.2.29) that the angular correlation may, in general, precess at the fundamental frequency ω_L as well as higher harmonics $N\omega_L$.

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

3.2.1.2. Electric Quadrupole Interaction

Once again, we restrict our attention to the special case of axially symmetric interactions. From eq.(2.2.30), the energy separation between two m states for the electric quadrupole interaction is

$$E_Q(m) - E_Q(m') = 3 |m^2 - m'^2| \hbar \omega_Q \quad (3.2.30)$$

Substituting into eq.(3.2.26)

$$G_{k_1 k_2}^{NN}(t) = \sqrt{(2k_1 + 1)(2k_2 + 1)} \sum_{m, m'} \begin{pmatrix} I & I & k_1 \\ m' & -m & N \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m' & -m & N \end{pmatrix} e^{-i3|m^2 - m'^2|\omega_Q t} \quad (3.2.31)$$

Since the factor multiplying the two 3-j symbols depends on the summation index m , the summation cannot be performed as simply as it was done in the magnetic case. There we employed the orthogonality of the 3-j symbols, which caused the disappearance of the interference terms with $k_1 \neq k_2$. In the case of quadrupole interactions these interference terms exist, in general, and no simple relationship can be established between the correlation factors $A_{kk} = A_k(1)A_k(2)$ of the unperturbed correlation and the factors

$$A_{k_1}(1)A_{k_2}(2)G_{k_1 k_2}^{NN}(t) \quad (3.2.32)$$

that are needed to describe the perturbed correlation. The 'interference' factors $A_{k_1}(1)A_{k_2}(2)$ with $k_1 \neq k_2$ must be computed from the factors $A_{k_1}(1)$ and $A_{k_2}(2)$ which must be individually known. This requires an accurate knowledge of the multipole expansion of the two radiations involved in the cascade. One must note that, from eq.(3.2.31), if $k_1 = 0$ or $k_2 = 0$, the perturbation factor vanishes and, because k_1 and k_2 only assume even values, $k_1 \neq k_2$ requires $k_{max} \geq 4$.

The perturbation factor can now be rewritten more clearly as

$$G_{k_1 k_2}^{NN}(t) = \sum_n S_{nN}^{k_1 k_2} \cos(n\omega_0 t) \quad (3.2.33)$$

where the summation index n takes all the positive integer values (including 0) $|m^2 - m'^2|$ for integer I , with $\omega_0 = 3\omega_Q$, and is $2|m^2 - m'^2|$ for half-integer I , with $\omega_0 = 6\omega_Q$. The exponential term can be transformed into $\cos$ because we only need the real component (that's the only part we can measure).

The coefficient $S_{nN}^{k_1 k_2}$ is given by

$$S_{nN}^{k_1 k_2} = \sum_{m, m'} \begin{pmatrix} I & I & k_1 \\ m' & -m & N \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m' & -m & N \end{pmatrix} \sqrt{(2k_1 + 1)(2k_2 + 1)} \quad (3.2.34)$$

remembering that the summation over m and m' should only include those terms where m and m' satisfy the condition $|m^2 - m'^2| = n$ for integer I , and $2 \mid m^2 - m'^2 = n$ for half-integer I .

Therefore, the result obtained in eq.(3.2.33) tells us that the angular correlation precesses with frequencies $\omega_0, 2\omega_0, \dots, n\omega_0$, where each frequency has the amplitude $S_{nN}^{k_1 k_2}$.

3.3. Time-dependent Interactions

There have been several attempts to deal with time-dependent interactions in perturbed angular correlations. For the investigation of this problem it is convenient and customary to introduce the so-called Liouville formalism, which mainly means that one has to work with superoperators (ref.[2]).

From equations (3.2.17) and (3.2.13) we have

$$\hat{\rho}(k_1, t) = \exp \left\{ -\frac{i}{\hbar} \int_0^t \hat{H}(t) dt \right\} \hat{\rho}(k_1, 0) \exp \left\{ \frac{i}{\hbar} \int_0^t \hat{H}(t) dt \right\} \quad (3.3.1)$$

and applying eq. (A.4.5) we get

$$\hat{\rho}(k_1, t) = \exp \left\{ -\frac{i}{\hbar} \int_0^t H^\times(t) dt \right\} \hat{\rho}(k_1) = \hat{\Omega}(t) \hat{\rho}(k_1, 0) \quad (3.3.2)$$

where we defined the time-evolution operator $\hat{\Omega}(t)$ represented in the Liouville formalism as

$$\hat{\Omega}(t) = \exp \left\{ -\frac{i}{\hbar} \int_0^t H^\times(t) dt \right\} \quad (3.3.3)$$

and $H^\times$ is the so-called Liouville operator, which is related to the Hamiltonian $\hat{H}$ of the system in the way that $H^\times$ acting on any operator $\hat{A}$ is equivalent to the commutator of $\hat{H}$ and $\hat{A}$, i.e.

$$H^\times \hat{A} = [\hat{H}, \hat{A}] \quad (3.3.4)$$

(A more complete description of Liouville operators is done in the appendix A.4)

This way, we can write eq. (3.2.19) as

$$W(\mathbf{k}_1, \mathbf{k}_2, t) = \text{Tr} \left\{ \hat{\rho}(\mathbf{k}_2) \hat{\Omega}(t) \hat{\rho}(\mathbf{k}_1, 0) \right\} \quad (3.3.5)$$

3. Perturbed $\gamma - \gamma$ Angular Correlation (PAC)

If the system under consideration is governed by a Hamiltonian $\hat{H}$, which does not depend explicitly on time, the evolution superoperator can be easily found by the solution of the Schrodinger-von Neumann equation

$$\frac{d}{dt}\hat{\Omega}(t) = \frac{i}{\hbar}H^\times\hat{\Omega}(t) \quad (3.3.6)$$

given by

$$\hat{\Omega}(t) = e^{-\frac{i}{\hbar}H^\times t}. \quad (3.3.7)$$

In the case that the corresponding system undergoes time-dependent perturbations, $\hat{\Omega}(t)$ is the solution of a complicated integro-differential equation. This is the point where we need the use of the Theory of Stochastic Perturbations. Blume [1] treated it in a little less general model, using obvious arguments of statistics. In the following section we will present a derivation based on Blume's calculation, but with slightly different aspects, following Winkler and Gerdau derivation [2].

4. Theory of Stochastic Perturbations in Angular Correlations

4.1. The Stochastic Model

Let's start by considering the time-dependent Hamiltonian of a system in which the interaction jumps at random between a finite number N of states:

$$\hat{H}(t) = \sum_{j=1}^N \hat{V}_j f_j(t) \quad (4.1.1)$$

Here $\hat{V}_j$ is the interaction Hamiltonian of state j and the set of functions $f_j(t)$, defined such that for any time t_1 , $f_{k \neq j}(t_1) = 0$ if $f_j(t_1) = 1$, describes the state in which the system is found at a given time.

In perturbed angular correlations, we deal with nuclei, which interact with their electronic and ionic environment. Thus, the various V_j are defined by the hyperfine interaction of the nucleus with the corresponding state of its environment.

It is supposed that in the course of time the nuclear environment changes its state in such a way that the probability $P_{a \rightarrow b}(\tau_1, \tau_2)$ of finding the system at a moment τ_2 in the state b when it was at $\tau_1 \leq \tau_2$ in the state a is independent of the “history”, i.e. of the states which were passed by the system before τ_1 , and depends only on the time interval $t = \tau_2 - \tau_1$. Therefore, we can consider it to be a Markov process (see appendix A.5).

Under these conditions the problem arises to derive an explicit expression for the time-evolution operator $\hat{\Omega}(t)$, which is the central element in the theory of perturbed angular correlation because it describes the evolution of the intermediate nuclear state of the $\gamma\gamma$ -cascade (or $e^- \gamma$ -cascade) under the influence of hyperfine interaction.

As it was previously seen, in angular correlations we are only interested in the nuclear spin variables, so we are occupied with the finite-dimensional vector space constituted by the direct product of the angular momentum eigenstates $|Im\rangle$ and the states $|a\rangle$ of the nuclear environment (the latter tell us in which of the N states the system is). Consequently the

4. Theory of Stochastic Perturbations in Angular Correlations

ket-bra products

$$|a\rangle |Im\rangle \langle Im'| \langle a'| \equiv |aa'mm'\rangle \quad (4.1.2)$$

form a complete orthonormal set of base operators in the corresponding Liouville space, i.e

$$\sum |aa'mm'\rangle \langle aa'mm'| = \hat{1} \quad (4.1.3)$$

Since an expression of the form (4.1.1) is only reasonable if the interaction is due to classical fields, the model is applicable merely to the so-called diagonal hyperfine interactions. In this special case the time-evolution of the combined system nucleus plus environment is restricted to the subspace of the $|aamm'\rangle \equiv |amm'\rangle$.

In the Liouville formalism not only the base operators but any operator, especially the density operator, can be characterized as “super-kets”. Remembering (4.1.3) we get

$$|\rho\rangle = \sum |amm'\rangle \langle amm'| \rho \quad (4.1.4)$$

where the coefficient $\langle amm'| \rho$ is equivalent to the matrix element $\langle m| \rho |m'\rangle$ of the nuclear density matrix under the condition that the environment is in the state a .

To derive a master equation for $\hat{\Omega}(t)$, we remember that from eq.(3.3.7), if the environment is in a state j , the time-evolution in the interval Δt is given by $\exp\left(-\frac{i}{\hbar} V_j^\times \Delta t\right)$, so it is physically reasonable to assume that, for sufficiently small Δt , we can consider that the time evolution of a system that goes from a state i to a state j is given by the transition probability $P_{i \rightarrow j}(\Delta t)$ times the time-evolution operator, i.e., $P_{i \rightarrow j}(\Delta t) \exp\left(-\frac{i}{\hbar} V_j^\times \Delta t\right)$. (ref.[14])

Thus, dividing the time interval t of a transition from a state $|a\rangle$ to a state $|c\rangle$ in $n + 1$ subintervals $\Delta t = t/(n + 1)$ we can write

$$\begin{aligned} \langle c| \hat{\Omega}(t) |a\rangle &= \lim_{n \rightarrow \infty} \sum_{i_1 \dots i_n} P_{a \rightarrow i_1}(\Delta t) P_{i_1 \rightarrow i_2}(\Delta t) \dots P_{i_n \rightarrow i_c}(\Delta t) \\ &\quad \cdot \exp\left(-\frac{i}{\hbar} V_c^\times \Delta t\right) \dots \exp\left(-\frac{i}{\hbar} V_{i_2}^\times \Delta t\right) \\ &\quad \cdot \exp\left(-\frac{i}{\hbar} V_{i_1}^\times \Delta t\right) \end{aligned} \quad (4.1.5)$$

where the sum amounts to all possible intermediate states i_k .

Because this is a Markov process (appendix A.5), the Chapman-Kolmogorov equation (A.5.4) allows us to write

$$P_{a \rightarrow b}(s + t) = \sum_i P_{a \rightarrow i}(s) P_{i \rightarrow b}(t) \quad (4.1.6)$$

so, after another time Δt we get:

$$(b|\hat{\Omega}(t + \Delta t)|a) = \exp\left(-\frac{i}{\hbar}V_b^\times \Delta t\right) \sum_c P_{c \rightarrow b}(\Delta t) (c|\hat{\Omega}(t)|a) \quad (4.1.7)$$

For sufficiently small time interval Δt , it is reasonable to consider the relations

$$\begin{aligned} P_{a \rightarrow a}(\Delta t) &= 1 - \gamma_a \Delta t + O(\Delta t) \\ P_{a \rightarrow b}(\Delta t) &= \omega_{a \rightarrow b} \Delta t + O(\Delta t) \end{aligned} \quad (4.1.8)$$

The quantities $1/\gamma_a = \tau_a$ give the mean lifetime of the corresponding stochastic states, whereas the $\omega_{a \rightarrow b}$ are the transition probabilities per unit time. We define $\omega_{a \rightarrow a} = 0$ in order to simplify some of the following formulas.

Since the total probability that any transition occurs is normalized to one,

$$\sum_b P_{a \rightarrow b}(\Delta t) = 1 \quad (4.1.9)$$

we get

$$\gamma_a = \sum_b \omega_{a \rightarrow b}$$

Expanding the exponential function in (4.1.7) into a power series up to the first order in Δt , we get

$$(b|\hat{\Omega}(t + \Delta t)|a) = \left(1 - \frac{i}{\hbar}V_b^\times \Delta t\right) \sum_c [\delta_{bc} (1 - \gamma_c \Delta t) + \omega_{c \rightarrow b} \Delta t] (c|\hat{\Omega}(t)|a) \quad (4.1.10)$$

and so

$$\lim_{\Delta t \rightarrow 0} \frac{1}{\Delta t} \left[(b|\hat{\Omega}(t + \Delta t)|a) - (b|\hat{\Omega}(t)|a) \right] = \sum_c \left[\left(-\frac{i}{\hbar}V_c^\times - \gamma_c \right) \delta_{bc} + \omega_{c \rightarrow b} \right] (c|\hat{\Omega}(t)|a) \quad (4.1.11)$$

By the definition of derivative, this means that the time-evolution operator $\hat{\Omega}(t)$ is determined by a differential equation of the form

$$\frac{d}{dt} \hat{\Omega}(t) = \left(-\frac{i}{\hbar}H_{st}^\times + \hat{R} \right) \hat{\Omega}(t) \quad (4.1.12)$$

which has to be compared with expression (3.3.6) that describes the situation when there are no fluctuations present. The formal solution of (4.1.12) is simply

$$\hat{\Omega}(t) = \exp \left[\left(-\frac{i}{\hbar}H_{st}^\times + \hat{R} \right) t \right] \quad (4.1.13)$$

4. Theory of Stochastic Perturbations in Angular Correlations

In the Liouville space representation, this is a matrix exponential function, where the elements of the argument matrix are those given by Blume in ref.[1]:

$$(bm_2m'_2 | \hat{R} | am_1m'_1) = \delta_{m_1m_2} \delta_{m'_1m'_2} \omega_{a \rightarrow b} \quad (4.1.14)$$

$$(am_2m'_2 | \hat{R} | am_1m'_1) = -\delta_{m_1m_2} \delta_{m'_1m'_2} \sum_b \omega_{a \rightarrow b} \quad (4.1.15)$$

$$(bm_2m'_2 | H_{st}^\times | am_1m'_1) = \delta_{ab} (m_2m'_2 | V_a^\times | m_1m'_1) \quad (4.1.16)$$

The index of $H_{st}^\times$ relates to the fact that this Liouville operator is constructed from the Hamiltonians of the different stochastic states.

Using the general definition of a Liouville operator, as discussed in the Appendix A.4, it can be verified easily that the following relation holds:

$$(m_2m'_2 | V_a^\times | m_1m'_1) = \delta_{m'_1m'_2} \langle m_2 | V_a | m_1 \rangle - \delta_{m_1m_2} \langle m'_1 | V_a | m'_2 \rangle \quad (4.1.17)$$

4.2. The perturbation factors

The a priori probability of finding the environment at $t = 0$ in the state a is p_a . If we assume that in the moment, when the first radiation is emitted, the nuclei are decoupled from their surroundings, we can write (ref.[2])

$$(am_1m'_1 | \rho(0)) = p_a \langle m_1 | \rho(k_1, 0) | m'_1 \rangle \quad (4.2.1)$$

Since we get no information about the particular state of the environment from our experimental arrangement, we have to sum over all possible initial and final states getting

$$\langle m_2 | \rho(k_1, t) | m'_2 \rangle = \sum_{a,b} p_a (bm_2m'_2 | \hat{\Omega}(t) | am_1m'_1) \langle m_1 | \rho(k_1, 0) | m'_1 \rangle \quad (4.2.2)$$

Substituting this result on eq.(3.2.18), we get the perturbed angular correlation function for stochastic states:

$$W(k_1, k_2, t) = \sum_{k_1, k_2, N_1, N_2} A_{k_1}(1) A_{k_2}(2) [(2k_1 + 1)(2k_2 + 1)]^{-\frac{1}{2}} \cdot G_{k_1 k_2}^{N_1 N_2}(t) \cdot Y_{k_1}^{N_1*}(\theta_1, \phi_1) Y_{k_2}^{N_2}(\theta_2, \phi_2) \quad (4.2.3)$$

where the perturbation factors are given by

$$\begin{aligned}
 G_{k_1 k_2}^{N_1 N_2}(t) = & \sum_{m_1 m_2} (-1)^{2I+m_1+m_2} [(2k_1+1)(2k_2+1)]^{\frac{1}{2}} \\
 & \cdot \begin{pmatrix} I & I & k_1 \\ m'_1 & -m_1 & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_2 & -m_2 & N_2 \end{pmatrix} \\
 & \cdot \sum_{a,b} p_a (bm_2 m'_2 | \exp \left[\left(-\frac{i}{\hbar} H_{st}^\times + \hat{R} \right) t \right] | am_1 m'_1) \quad (4.2.4)
 \end{aligned}$$

They will obviously be determined by the eigenvalues of the Blume matrix, which are generally complex since this matrix is not hermitian.

We denote the eigenvectors by $|M_q\rangle$ and $(N_q|$ indicating that they are kets and bras in Liouville space. They verify the relation $Tr \{N_r^\dagger M_q\} \equiv (N_r| M_q) = \delta_{rq}$ for all r, q and $\sum_q |M_q\rangle (N_q| = \hat{1}$

Defining the eigenvalues by

$$\left(-\frac{i}{\hbar} H_{st}^\times + \hat{R} \right) |M_q\rangle = (-\lambda_q + i\omega_q) |M_q\rangle \quad (4.2.5)$$

$$(N_q| \left(-\frac{i}{\hbar} H_{st}^\times + \hat{R} \right) = (-\lambda_q + i\omega_q) (N_q| \quad (4.2.6)$$

it can be shown, by using the properties of $H_{st}^\times$ and $\hat{R}$, that

- the Blume matrix has always the eigenvalue 0 with the corresponding eigenket $|M_0\rangle = (2I+1)^{-1} \sum_{ab} p_a |amm\rangle$ and eigenbra $(N_0| = \sum_{am} (amm|$ independent of the special choice of parameters;
- the real part of the eigenvalues is always less or equal 0;
- all eigenkets except $|M_0\rangle$ have vanishing trace;
- the complex eigenvalues appear in conjugate pairs: if $|M_q\rangle$ is the eigenket for the eigenvalue $(-\lambda_q + i\omega_q)$, then $|M_q^\dagger\rangle$ is the eigenket for the eigenvalue $(-\lambda_q - i\omega_q)$.

From these theorems we conclude that

$$\begin{aligned}
 G_{k_1 k_2}^{N_1 N_2}(t) = & \sum_{m_1 m_2} (-1)^{2I+m_1+m_2} [(2k_1+1)(2k_2+1)]^{\frac{1}{2}} \\
 & \cdot \begin{pmatrix} I & I & k_1 \\ m'_1 & -m_1 & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_2 & -m_2 & N_2 \end{pmatrix} \\
 & \cdot \sum_{a,b,q} p_a (bm_2 m'_2 | N_q) (M_q | am_1 m'_1) e^{(-\lambda_q + i\omega_q)t} \quad (4.2.7)
 \end{aligned}$$

consists of a number of exponentially damped trigonometric functions.

4. Theory of Stochastic Perturbations in Angular Correlations

If $\lambda_q \neq 0$ for all $q \neq 0$ we get relaxation to isotropy:

$$G_{k_1 k_2}^{N_1 N_2}(t \rightarrow \infty) = \delta_{N_1 0} \delta_{N_2 0} \delta_{k_1 0} \delta_{k_2 0} \quad (4.2.8)$$

For **single crystal** materials we have to use the general expression for the perturbation factor, eq.(4.2.7).

For **polycrystal** materials the electric field gradient doesn't have the same orientation for all nuclei, so we can consider the average of all possible orientations. Thus, let us start by noting that the base kets $|mm'\rangle$ are related to the eigenvectors $|\mu\rangle$ of $I_{z'}$ defined in a coordinate system (x', y', z') , which is rotated by the Euler angles α , β , and γ , by the formula

$$|mm'\rangle = \sum_{\nu\nu'} D_{\nu m}^{(I)}(\alpha, \beta, \gamma) D_{\nu' m'}^{(I)*}(\alpha, \beta, \gamma) |\mu\nu'\rangle \quad (4.2.9)$$

where $D_{\nu m}^{(I)}(\alpha, \beta, \gamma)$ (and $D_{\nu' m'}^{(I)*}(\alpha, \beta, \gamma)$) are the rotation matrices associated to the Euler angles α, β, γ , with the properties referred in appendix A.3.

Inserting (4.2.9) into (4.2.4) and averaging over the Euler angles yields the perturbation factors for the polycrystalline source:

$$\begin{aligned} G_{kk}(t) &= \overline{G_{k_1 k_2}^{N_1 N_2}}^{\alpha\beta\gamma}(t) = \sum_{m_1 m_2} (-1)^{2I+m_1+m_2} \\ &\cdot \begin{pmatrix} I & I & k \\ m'_1 & -m_1 & N \end{pmatrix} \begin{pmatrix} I & I & k \\ m'_2 & -m_2 & N \end{pmatrix} \\ &\cdot \sum_{a,b} p_a (bm_2 m'_2 | \exp \left[\left(-\frac{i}{\hbar} H_{st}^\times + \hat{R} \right) t \right] | am_1 m'_1) \end{aligned} \quad (4.2.10)$$

This way, we can re-write eq.(4.2.10) in the form

$$G_{kk}(t) = \sum_{q=1}^{(2I+1)^2 N} A_{kq} \cos(\omega_q t) \exp(-\lambda_q t) \quad (4.2.11)$$

with the amplitudes A_{kq} given by

$$\begin{aligned} A_{kq} &= \sum_{N, m_1, m_2} (-1)^{2I+m_1+m_2} \begin{pmatrix} I & I & k \\ m'_1 & -m_1 & N \end{pmatrix} \begin{pmatrix} I & I & k \\ m'_2 & -m_2 & N \end{pmatrix} \times \\ &\times \sum_{a,b} p_a (bm_2 m'_2 | N_q) (M_q | am_1 m'_1) \end{aligned} \quad (4.2.12)$$

As a final note, we can see that the $G_{k_1 k_1}^{N_1 N_2}$ factor, either for single crystal or polycrystal materials, has a component depending on ω_q that represents a set of oscillations, just as we had in the static cases, but it also has an extra term depending on λ_q given by $e^{-\lambda_q t}$, which represents a damping in our function. Without any kind of transitions between the different states (i.e., without dynamic behavior), this term disappears, so the existence of a damping is the signature of the presence of dynamic processes.

5. Implementing the Time Dependent Perturbation Function $R(t)$

So far we have only seen the PAC features in the theoretical point of view.

We learned that, studying the $W(\theta, t)$ function given by the correlation of two photons emitted with an angle θ between the directions of each other, we are able to analyse the perturbation factor $G(t)$ which contains all the information about the nuclear environment at a local scale, given by the hyperfine interactions.

Besides the theoretical predictions, let us now focus on how to experimentally obtain the perturbation factor $G(t)$ in order to allow us to study the properties of the nuclei under consideration.

5.1. The Experimental Perturbation Function $R(t)$

The γ -rays are emitted due to the radioactive decay of the nuclei, and as we know, radioactive decay is described by a function like $N(t) = N_0 e^{-\lambda t}$, where $N(t)$ is the counting rate, N_0 is the number of radioactive nuclei at $t = 0$ and $\lambda = \frac{1}{\tau}$, being τ the mean lifetime. Also, $\lambda = \frac{\ln 2}{t_{1/2}}$, where $t_{1/2}$ is the half-life time (when $t = t_{1/2}$, $N = N_0/2$).

In PAC experiments we have a set of detectors whose apparatus is usually done in such a way that each pair of detectors has an angle of 90° or 180° between them. If a certain γ -ray is detected by a detector i with the energy E_{γ_1} the clock starts counting until another γ -ray is detected by a detector j with the energy E_{γ_2} , making the clock to stop. This event will be stored in the coincidence counting rate of the pair of detectors i, j .

The coincidence counting rate of any pair of detectors i, j thus is given by

$$N_{ij}(\theta, t) = N_0 e^{-\lambda t} W(\theta, t) + B \quad (5.1.1)$$

where θ , as previously said, is the angle between the direction of emission of each γ , $t = t_{\gamma_2} - t_{\gamma_1}$, $W(\theta, t)$ is the perturbed correlation function given by eq.(4.2.3) and B is proportional to the background random coincidence count rate, which we assume to be time independent for simplicity.

5. Implementing the Time Dependent Perturbation Function $R(t)$

We can see an example of such an arrangement in Fig.(5.1.1).

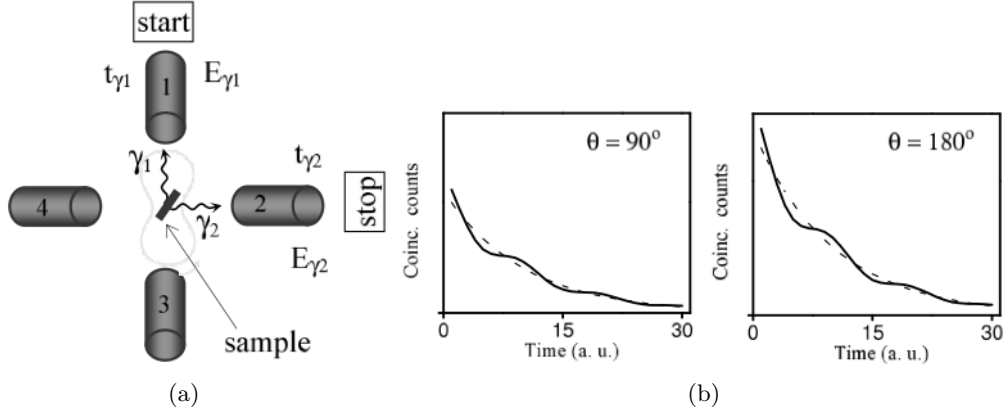


Figure 5.1.1.: (a) Schema of a in-plane four detectors $\gamma - \gamma$ PAC experimental setup. On (b) we have an illustrative representation of the coincidence count rate at 90° and 180° . The dashed line represents the count rate for a non perturbed system, i.e., the pure exponential histogram associated to the radioactive decay of the intermediate nuclear level (ref.[8]).

Looking at equation 5.1.1 it is easy to understand that this is not a good expression for us to be able to study the perturbed angular correlation function $W(\theta, t)$ from the experimental results.

The way to solve this starts by subtracting the background B from the experimental results. Then, we need to figure out a way to eliminate the exponential of the decay, leaving behind only the correlation function. To do that, first, we do an appropriate average of all the coincidence count rate $N_{ij}(\theta, t)$ coming from pairs of detectors with the same angular separation, given by

$$N(\theta, t) = \sqrt[N_\theta]{\prod_{ij} N_{ij}(\theta, t)} \quad (5.1.2)$$

N_θ being the total number of pairs of detectors separated by θ (which will have the possible values of 90° and 180° , as said before). This way, we can minimize the effect of different detector efficiencies.

After this, we construct the experimental perturbation function:

$$R(t) = 2 \frac{N(180^\circ, t) - N(90^\circ, t)}{N(180^\circ, t) + 2N(90^\circ, t)} \quad (5.1.3)$$

Because N_0 and $e^{-\lambda t}$ are the same for every $N(\theta, t)$, the function $R(t)$ is the perfect solution for us to eliminate these undesired terms, getting the final expression

$$R(t) = 2 \frac{W(180^\circ, t) - W(90^\circ, t)}{W(180^\circ, t) + 2W(90^\circ, t)}. \quad (5.1.4)$$

5.1. The Experimental Perturbation Function $R(t)$

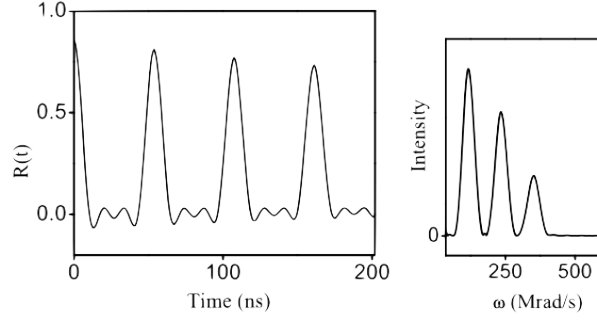


Figure 5.1.2.: Example of the experimental $R(t)$ function and the corresponding Fourier transform with the three frequencies $(\omega_1, \omega_2, \omega_3)$ characteristic from spin 5/2 and that fully characterize the EFG parameters V_{zz} and η (ref.[8]).

The $R(t)$ function is the best way to get the information about the hyperfine interaction since it gives relevance to the perturbation factor $G(t)$, which contains all this information. In Fig.(5.1.2) an example of the $R(t)$ function and its Fourier transform is shown.

By noting that

$$R(t) \sim \frac{\Delta W}{\langle W \rangle} \quad (5.1.5)$$

where $\Delta W = W(180^\circ, t) - W(90^\circ, t)$ and $\langle W \rangle = (W(180^\circ, t) + W(90^\circ, t)) / 2$, we can see that $R(t)$ not only eliminates $N_0 e^{-\lambda t}$ but it essentially is the ratio that represents a relative variation $W(180^\circ, t) - W(90^\circ, t)$ compared to its mean value $(W(180^\circ, t) + W(90^\circ, t)) / 2$.

The factor “2” inside the denominator of eq.(5.1.4) transforms $R(t)$ in such a way that it puts in evidence the angular anisotropy coefficients of the cascade, since $P_2(\cos(180^\circ)) = 1$ and $P_2(\cos(90^\circ)) = -1/2$, where $P_2(x)$ are the Legendre polynomials for $n = 2$.

This formalization of $R(t)$ is adopted since for polycrystalline samples the time-dependent terms in the denominator are eliminated. As the anisotropy coefficients A_{44} are usually smaller than the coefficients A_{22} , the $R(t)$ function is approximated to

$$R(t) \approx A_{22} G_{22}(t) \quad (5.1.6)$$

in this cases.

For monocrystal samples, the time-dependent terms in the denominator don't cancel, but they usually are less than 4% of the constant term in the denominator, so we still use the same expression (5.1.4) in this cases, for simplification.

The fit observable of TFPAC (chapter 6) fully reproduces eq.(5.1.4).

5.2. Static Distributions

5.2.1. Frequency Distribution

So far we assumed in our discussion of the electric quadrupole interaction that the electric field gradients (EFG) acting on the nuclear quadrupole moments are the same at every nuclear site¹. However, slight variations of the crystalline fields are expected, caused by lattice imperfections (e.g., implantations defects) and impurity centers and thus a distribution of frequencies around a central value is measured. This will induce an attenuation in the perturbation function $R(t)$, of the static type.

For low concentration of defects these distributions are generally represented by a gaussian or a lorentzian function in the frequency space.

For an infinite number of states, the averaged perturbation factor $G(t)$ in the static limit is given by the integral (ref.[15])

$$G(t) = \int_{-\infty}^{+\infty} D(\omega_Q) G(t; \omega_Q) d\omega_Q \quad (5.2.1)$$

where $D(\omega_Q)$ is the distribution of the interaction strength, expressed by the quadrupole frequency $\omega_Q = eQV_{zz}/4I(2I-1)\hbar$.

For a finite number N of states $|a\rangle$ with different quadrupole frequencies ω_{Qa} , the integral (5.2.1) has to be replaced by the sum

$$G(t) = \sum_{a=1}^N D(\omega_{Qa}) G(t; \omega_{Qa}) \Delta\omega_{Qa}. \quad (5.2.2)$$

In our study we considered $D(\omega_Q)$ to be a lorentzian function, representing the relative intensities of the discrete frequencies ω_{Qa} , centered at ω_Q , and rather than using a constant frequency interval $\Delta\omega_Q$, we have chosen the intervals $\Delta\omega_{Qa}$ such that the area covered by the distribution $D(\omega_{Qa})$ is divided into N equal parts.

This condition can be represented as

$$D(\omega_{Qa}) \Delta\omega_{Qa} = \frac{1}{N} \quad (5.2.3)$$

and so equation (5.2.1) transforms into

$$G(t) = \frac{1}{N} \sum_{a=1}^N G(\omega_{Qa}; t). \quad (5.2.4)$$

¹Are the same for static cases or, in dynamic cases, are the same for nuclei in the same state

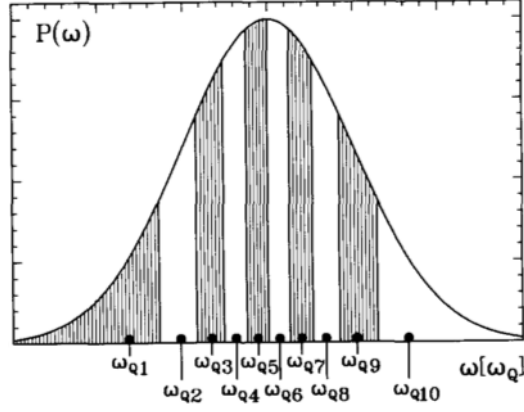


Figure 5.2.1.: Example of a frequency distribution divided into 10 sections of equal area (ref.[15]).

Now, we have to remember that we are not only studying the cases where there may be several possible states with different EFG but we may have random transitions between those states during time, thus we have to take into account the distribution for each possible EFG.

This forbids us to use equations (5.2.1) and (5.2.2) because they only work for static cases, as pointed, due to the fact that it only depends in one of the quadrupole frequencies at each time ($G_{static}(t) = G(t; \omega_Q)$ as given by eq.(3.2.31).

In dynamic systems, the perturbation factor $G(t)$ is given by eq.(4.2.4) and so it depends on the several values of ω_{Qa} from the different states entering at the same time to construct the Blume matrix $\left(-\frac{i}{\hbar}H_{st}^\times + \hat{R}\right)$, due to the possible transitions between them, which means that we must take into account the distribution of each state but we cannot do it for each one separately.

So, we need to consider that for every possible state, the correspondent ω_Q might have a distribution but, how to implement the distributions with the Stochastic Hamiltonian transitions?

To make it simple to understand, let's consider that we only have two possible states, ω_{Q1} and ω_{Q2} , each one with a frequency distribution $D(\omega)$ given by a lorentzian or a gaussian function with full width at half maximum Γ_1 and Γ_2 , correspondingly.

In both cases we divide the distribution in N intervals with equal area, as considered for the static cases (equation 5.2.3). For each interval $\Delta\omega$, it will correspond a frequency given by the mean value $\omega = \frac{\omega_{final} + \omega_{initial}}{2}$, where $\Delta\omega = \omega_{final} - \omega_{initial}$, and the relative intensity $D(\omega)$ can be seen as the probability of having each value of ω .

5. Implementing the Time Dependent Perturbation Function $R(t)$

This means that for every state we will have N pairs² of $(\omega_i; D(\omega_i))$.

Without distributions we could only have transitions between ω_{Q1} and ω_{Q2} , but with distributions we have to take into account all the possible transitions between the N_1 values $(\omega_1)_i$ and the N_2 values $(\omega_2)_j$

This is done calculating $W_{final}(180^\circ; t)$ and $W_{final}(90^\circ; t)$, to construct $R(t)$, as

$$\begin{aligned} W_{final}(180^\circ; t) &= \sum_{i,j} D((\omega_1)_i) D((\omega_2)_j) W(180^\circ; t; (\omega_1)_i; (\omega_2)_j) \\ W_{final}(90^\circ; t) &= \sum_{i,j} D((\omega_1)_i) D((\omega_2)_j) W(90^\circ; t; (\omega_1)_i; (\omega_2)_j) \end{aligned} \quad (5.2.5)$$

i.e., we calculate $W(180^\circ; t)$ and $W(90^\circ; t)$ given that the state "1" has $\omega_{Q1} = (\omega_1)_i$ and the state "2" has $\omega_{Q2} = (\omega_2)_j$, and we consider that the probability of this to happen is $D((\omega_1)_i) D((\omega_2)_j)$, doing the sum for all $i = 1, \dots, N_1$ and $j = 1, \dots, N_2$.

Our method to give the correspondent weight in probability to each $W(180^\circ; t)$ and $W(90^\circ; t)$ is assured by the fact that

$$\sum_{i,j} D((\omega_1)_i) D((\omega_2)_j) = \left[\sum_i D((\omega_1)_i) \right] \left[\sum_j D((\omega_2)_j) \right] = 1 \quad (5.2.6)$$

since $\sum_i D(\omega_i) = 1$ because we only use frequency distributions given by normalized lorentzian or normalized gaussian functions, so $D((\omega_1)_i) D((\omega_2)_j)$ has the usual behavior of a probability distribution.

The existence of frequency distributions induces a damping along the time in the observable $R(t)$ function.

5.2.2. Time Resolution (Prompt)

To get useful information from the results of a measurement using the PAC technique, a theoretical fit to the experimental data is performed. The construction of a theoretical function $R_{fit}(t)$ which better fits that data is performed by a computer, so it will be represented for discrete instants of time.

On the other hand, when we are doing experiments measuring the time between two signals, Start and Stop, the ideal picture would be that for a physical $\Delta t = 0$ (two simultaneous

²We don't necessarily need the same number N of intervals for all the possible states. They are completely independent

5.3. Attenuation in the Coefficients $A_{kk'}$ and the Multiplicative Constant in $R(t)$

events) our experimental measurement would be $\Delta t' = 0$, however this is not the obtained result.

The data acquisition system has a finite resolution in time. The time between the detection of the first γ -radiation (or conversion electron) and the second γ -radiation is registered with an uncertainty described by a probability distribution, and $R_{fit}(t)$ must take this into account instead of the ideal discrete instants of time.

The histogram of the number of coincidences on a function of the time measured between the nuclear transitions is convoluted with a function (“prompt”) that represents the response of the measurement system to the measure of two simultaneous events. Usually, this function is approximated by a Gaussian function.

The convolution in the time domain represents a multiplication in frequency domain. Analysing the Fourier spectrum it's important to know how the attenuation in frequency might be obtained by the Fourier transform of the function that represents the curve “prompt”.

Considering a Gaussian function with full width at half maximum $F_{whm(prompt)}$, the curve that describes the attenuation in frequency is given by

$$P(\omega, F_{whm(prompt)}) = \exp \left(-\frac{\omega^2 \left(F_{whm(prompt)} \right)^2}{16 \ln 2} \right) \quad (5.2.7)$$

where ω is expressed in $rad.s^{-1}$ and $F_{whm(prompt)}$ in s .

5.3. Attenuation in the Coefficients $A_{kk'}$ and the Multiplicative Constant in $R(t)$

The magnetic lens and the γ -detectors have a finite solid angle. Consequently, the angular correlation has an attenuation compared to the theoretical values calculated for a well defined angle between the detectors. Aiming to calculate this coefficients, it is necessary to do a summation of all the contributions to the anisotropy from the radiations inside the solid angle of detection. This way, we define the attenuation coefficients $Q_k(\gamma)$ and $Q_k(e^-)$ (ref.[16]) by:

$$Q_k(\gamma \text{ or } e^-) = \frac{\int \epsilon(\theta) P_k(\cos(\theta)) \sin(\theta) d\theta}{\int \epsilon(\theta) \sin(\theta)} \quad (5.3.1)$$

with $\epsilon(\theta)$ representing the efficiency of the detectors as a function of the azimuthal angle from the cylindrical symmetry axis of the detectors. $P_k(\cos(\theta))$ is the Legendre Polynomial of order k .

5. Implementing the Time Dependent Perturbation Function $R(t)$

In the final expression of $W(k_1, k_2; t)$ this will be taken into account by replacing the theoretical coefficients $A_{kk'}$ for “effective” coefficients $A_{kk'}^{eff}$ given by:

$$A_{kk'}^{eff} = A_k^{eff}(\gamma_1)A_{k'}^{eff}(\gamma_2) = Q_k(\gamma_1)A_k(\gamma_1)Q_{k'}(\gamma_2)A_{k'}(\gamma_2) \quad (5.3.2)$$

for $\gamma - \gamma$ correlations and

$$A_{kk'}^{eff} = A_k^{eff}(e^-)A_{k'}^{eff}(\gamma) = Q_k(e^-)A_k(e^-)Q_{k'}(\gamma)A_{k'}(\gamma) \quad (5.3.3)$$

for $e^- - \gamma$ correlations.

Occasionally, the results we find for the “effective” coefficients $A_{kk'}^{eff}$ concerning the used detectors have an error associated to calculation difficulties, so a correction to the $R(t)$ function must be considered. This is done by a simple approximation, considering a multiplicative constant a , resulting in the perturbation function: $a.R(t)$. The multiplicative constant should be ~ 1 . In our case we have $a \approx 0.9$.

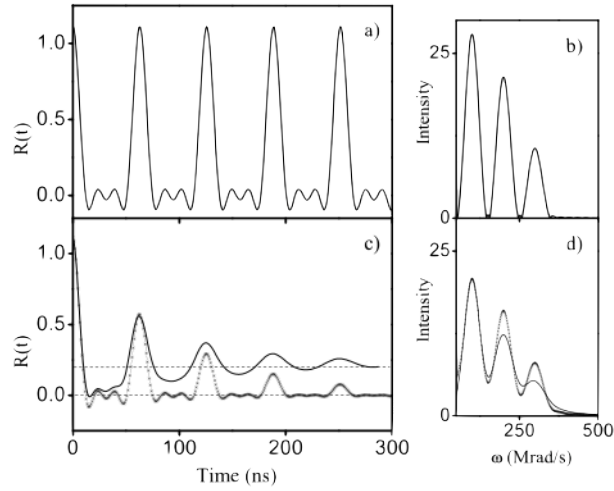


Figure 5.3.1.: (a) Schematic representation of $R(t)$ with no attenuation, for a EFG with $\eta = 0$, with the correspondent Fourier transform in (b). (c) Representation of $R(t)$ for the same frequency as in (a) with static attenuation, line curve, and with dynamic attenuation (for fast fluctuations), line plus symbol curve, with the correspondent Fourier transform in (d). In the frequencies space the attenuation to $\omega_1, \omega_2, \omega_3$ amplitudes diminish by the same factor in the case of damping due to dynamic interactions while for a static damping the attenuation is proportional to the frequency itself (ref.[8]).

6. TFPAC Program

The full program has a little more than 2000 lines (just counting code written by us and not the used external libraries, otherwise it would be a lot more) and is divided into the experimental PAC function, given by the simulation routine, and the fitting routine.

On Fig.6.0.1 we show the flow chart of TFPAC to help us correlate the different parts of the program with the theory presented before, for facilitation on understanding.

We need to divide our discussion in two sections: the simulation routine and the fitting routine.

The simulation routine aims to calculate the $R(t)$ function (eq. 5.1.4) for a set of input parameters. This way we can check our program to see if it is giving the expected results for some known ensembles. After that, we can use it to study some new setups and see what are the results we should expect to get in experimental measurements.

The fitting routine reads the experimental data from an external file and, using the simulation routine, it finds the best parameter values that give the theoretical $R(t)$ that best suits those results.

6.1. Simulation Routine

We can divide the simulation program in 3 big parts:

- Read and treatment of the input data
- Construction of the Blume matrix $\left(-\frac{i}{\hbar}H_{st}^{\times} + \hat{R}\right)$ and the calculation of its eigenvectors and eigenvalues
- Construction of the correlation functions $W(180^{\circ}, t)$ and $W(90^{\circ}, t)$, followed by the construction of $R(t)$.

Coupled to the main routine, we created a library with some useful subroutines. Some of the most important of them are:

- Calculation of the interaction Hamiltonian for the desired nuclear spin, with two

6. TFPAC Program

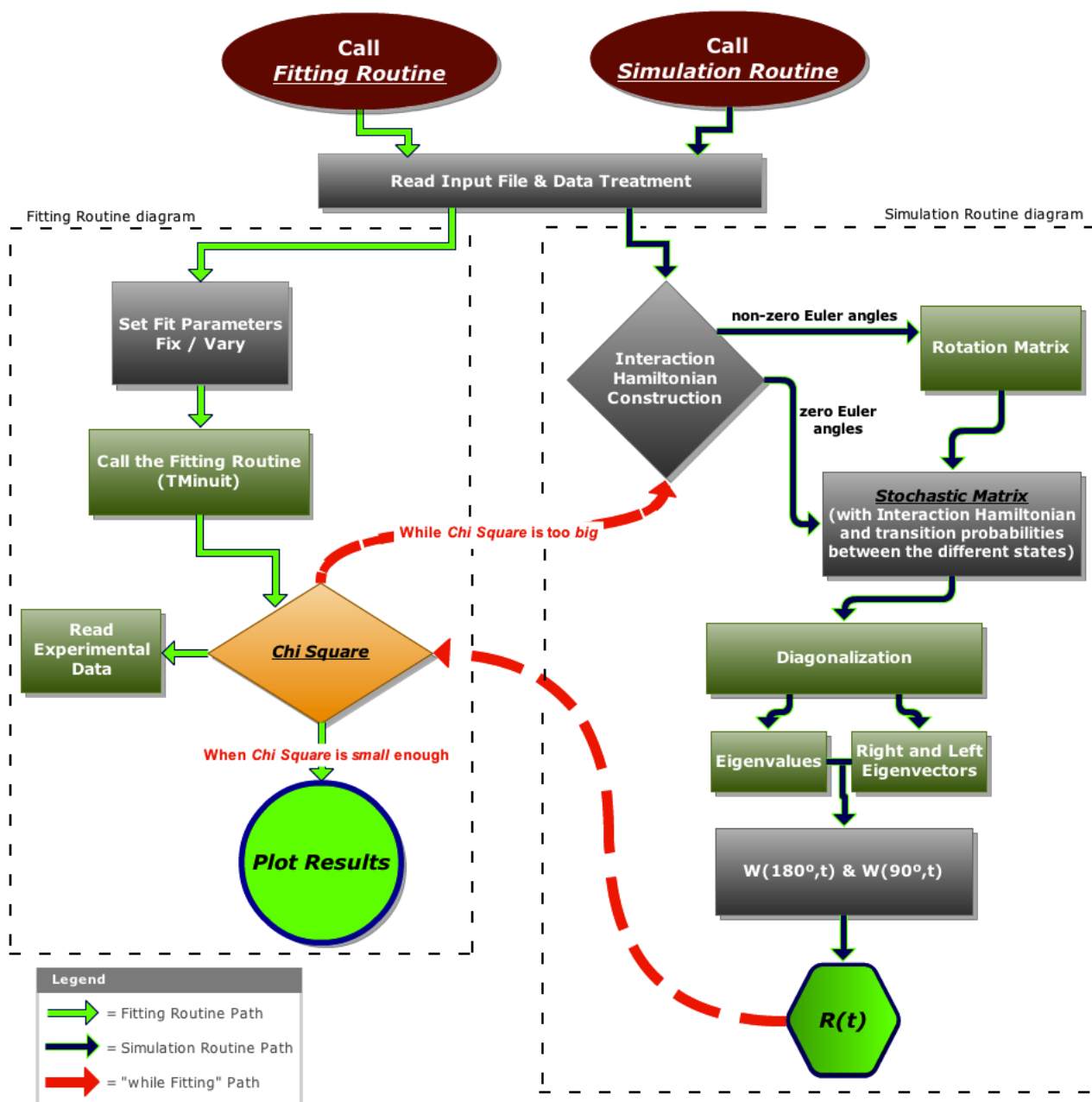


Figure 6.0.1.: Flow Chart of TFPAC

possible cases: zero or non-zero Euler angles

- Calculation of the rotation matrix to calculate the Hamiltonians in the case of non-zero Euler Angles
- The functions used in the matrix diagonalization
- Calculation of the frequency distributions and the attenuation function for the time resolution
- Calculation of 3j-symbols, spherical harmonics and Legendre Polynomials.

All of these are completely general and work for any given nuclear spin that we might want to work with.

By default, the interaction Hamiltonian is given for electric quadrupole interaction, but it is easily adapted for magnetic interaction or even join both of them, if one wants (however this is not a default option, since it is necessary to change lines of code to do this).

The treatment of the input data is very simple and basically it just does normalizations and transforms the values in such a way they can be better used in the program.

The difficult parts of the program are the construction and calculation of eigenvectors and eigenvalues of the Blume matrix and the implementation of the frequency distributions, as can be seen ahead.

The final part is just to put all the values calculated together to build $R(t)$ and make its output to an external file, followed by the plot of the results.

Now, we will describe in more detail the points of greatest relevance in TFPAC.

6.1.1. The particular Hamiltonian for spin $\frac{5}{2}$

In spite of the fact that TFPAC was built to run for every spin value, the system we are studying has nuclei with spin $5/2$ and axially symmetric EFG's of identical orientation only differing in their coupling constants.

The Hamiltonian of an electric quadrupole interaction expressed in terms of magnetic quantum numbers m , as we saw in expressions (2.2.27), is given by

$$\begin{aligned} H_{m,m} &= \hbar\omega_E \left(3m^2 - I(I+1) \right) \\ H_{m,m\pm 2} &= \hbar\omega_E \eta \frac{1}{2} [(I \mp m - 1)(I \mp m)(I \pm m + 1)(I \pm m)(I \pm m + 2)]^{\frac{1}{2}} \end{aligned} \quad (6.1.1)$$

All other elements vanish. Remember that $\omega_E = eQV_{zz}/\hbar 4I(2I-1)$, with eQ denoting the nuclear quadrupole moment, V_{zz} is the largest component (in magnitude) of the electric

6. TFPAC Program

field gradient tensor and the asymmetry parameter η is defined as $\eta = \frac{V_{xx}-V_{yy}}{V_{zz}}$, with the (arbitrary) prescription $|V_{xx}| \leq |V_{yy}| \leq |V_{zz}|$ giving result to $0 \leq \eta \leq 1$.

In our specific case of spin 5/2, the Hamiltonian obtained is

$$\hat{H} = \hbar\omega_E \begin{pmatrix} 10 & 0 & \eta\sqrt{10} & 0 & 0 & 0 \\ 0 & -2 & 0 & \eta\sqrt{18} & 0 & 0 \\ \eta\sqrt{10} & 0 & -8 & 0 & \eta\sqrt{18} & 0 \\ 0 & \eta\sqrt{18} & 0 & -8 & 0 & \eta\sqrt{10} \\ 0 & 0 & \eta\sqrt{18} & 0 & -2 & 0 \\ 0 & 0 & 0 & \eta\sqrt{10} & 0 & 10 \end{pmatrix} \quad (6.1.2)$$

All the values and constants that define our system are inserted by an input file, and that's where we can change and control all the dynamics of the system that we want to simulate.

The values that we have in the input file are:

- calibration (time scale)
- channel maximum (the number of points in time that we want to simulate)
- nuclear spin
- magnetic moment
- electric quadrupole moment
- time resolution
- and for each state we have
 - coupling constants (to give us $\hbar\omega_E$)
 - asymmetry parameter, η
 - Euler angles, α, β, γ
 - probability to be in the state i at $t = 0$, p_i
 - transition probabilities per time unit, $\omega_{a \rightarrow b}$
 - frequency distribution

Introducing the Hamiltonians of the different states, matricially represented as eq.(6.1.2), in eq.(4.1.17) and the transition probabilities in equations (4.1.14) and (4.1.15), we can define the Blume matrix and use it with the rest of the input values to construct the perturbation function $R(t)$.

6.1.2. Rotation Matrix

In the previous section we saw how the Hamiltonian of the system is built, but if we have non-zero Euler angles, this will change the Hamiltonian itself. The new Hamiltonian $\hat{H}'$ is related with the original one, $\hat{H}$, as:

$$\hat{H}' = \hat{D}^\dagger \hat{H} \hat{D} \quad (6.1.3)$$

where D is the rotation matrix and $D^\dagger$ is its conjugate.

For Euler angles, α, β, γ , as defined in Fig.(6.1.1), the rotation matrix is given by (ref.[11])

$$\begin{aligned} D &\equiv D_{mm'}^{(I)}(\alpha, \beta, \gamma) = e^{-i(m'\alpha + m\gamma)} \langle I, m' | \exp\left(-\frac{i}{\hbar} I_y \beta\right) | I, m \rangle \\ &= e^{-i(m'\alpha + m\gamma)} d_{mm'}^{(I)}(\beta) \end{aligned} \quad (6.1.4)$$

where we consider $d_{mm'}^{(I)}(\beta) = \langle I, m' | \exp\left(-\frac{i}{\hbar} I_y \beta\right) | I, m \rangle$, by definition.

The rotation matrix rank is given by the spin I as well as the rank of $d_{mm'}^{(I)}(\beta)$, of course. This way, in order to construct the rotation matrix for any spin we want, we need to use the explicit and general form of $d_{mm'}^{(I)}(\beta)$, given by (ref.[17])

$$\begin{aligned} d_{mm'}^{(I)}(\beta) &= \sum_k (-1)^{k-m+m'} \frac{\sqrt{(j+m)!(j-m)!(j+m')!(j-m')!}}{(j+m-k)!k!(j-k-m')!(k-m+m')!} \\ &\quad \times \cos\left(\frac{\beta}{2}\right)^{2j-2k+m-m'} \sin\left(\frac{\beta}{2}\right)^{2k-m+m'} \end{aligned} \quad (6.1.5)$$

taking the sum over k whenever none of the arguments of factorials in denominator are negative.

As was said before, the construction of the rotation matrix is made by a routine located in the library coupled to the main routine, and it's called everytime a non-zero Euler angle is considered.

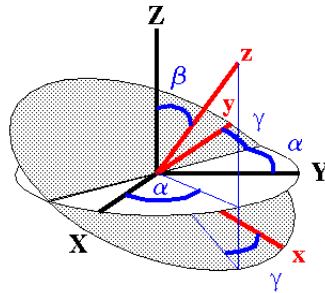


Figure 6.1.1.: Scheme representing the considered definition of Euler angles (ref.[18]).

6.1.3. Matrix Diagonalization

In this section it is shown how to find the eigenvalues and eigenvectors of the Blume matrix.

The diagonalization is made in four steps.

First, we balance the matrix. This involves two steps.

We permute the matrix H by a similarity transformation to block upper triangular form:

$$PHPT^T = H' = \begin{pmatrix} H'_{11} & H'_{12} & H'_{13} \\ 0 & H'_{22} & H'_{23} \\ 0 & 0 & H'_{33} \end{pmatrix}$$

where P is a permutation matrix.

Then, we apply a diagonal similarity transformation to H' to make the rows and columns of H' as close in norm as possible

$$H'' = DH'D^{-1}$$

This can improve the accuracy of the next process.

Second, we transform H'' in a upper Hessenberg matrix, which is zero below the first subdiagonal. Reduction of a matrix to Hessenberg form improves the calculation of its eigenvalues and eigenvectors.

$$H_{Hessenberg} = \begin{pmatrix} H_{11} & H_{12} & H_{13} & H_{14} \\ H_{21} & H_{22} & H_{23} & H_{24} \\ 0 & H_{32} & H_{33} & H_{34} \\ 0 & 0 & H_{43} & H_{44} \end{pmatrix}$$

Then, we calculate the eigenvalues and the eigenvectors of the Hessenberg matrix.

Finally, we transform it in the eigenvalues and the eigenvectors of the original matrix.

This process of diagonalization to find eigenvalues and eigenvectors was the first big problem and took some time to solve. The point is the Blume matrix has rank $N(2I+1)^2$, where N is the number of different possible stochastic states and I is the spin of the nuclei, and the matrix is complex, so it will have complex eigenvalues and eigenvectors. In the present

case, we are considering two possible states of $I = 5/2$, so we have a Blume matrix with dimension 72×72 .

For such big matrices (yes, they are not that big but they are complex, so it gets more complicated than usual), we need to be careful about the routines we use, because it's difficult to get results with good accuracy in such cases. This way, we decided not to build a routine from the beginning but to use packs of routines already built and well-tested.

After working with several methods and testing different packs of routines, our choice for diagonalization was the use of a LAPACK routine.

This routine is called ZGEEV (ref.[19]) and taking as input a complex matrix and its rank, it gives the right and left eigenvectors and the correspondent eigenvalues. It does all the steps described before for diagonalization, so we don't need to worry about them. It's very stable and works well and fast,

Now we have to make reference to a very tricky part that took us a lot of time to understand, simply because it's something we never needed to notice before.

First, let's notice that the eigenvectors and eigenvalues given by the diagonalization of the Blume matrix appear in the perturbation factors $G(t)$ (eq. 4.2.4) in the term

$$\sum_{ab} p_a (bm_2 m_2' | N_q) (M_q | am_1 m_1') e^{(-\lambda_q + i\omega_q)t} \quad (6.1.6)$$

where, as said, $-\lambda_q + i\omega_q$ are the eigenvalues, $|N_q\rangle$ are the right eigenvectors and $\langle M_q|$ are the left eigenvectors (see equations 4.2.6).

Second, we point out the notion of normal matrix:

A complex square matrix A is a normal matrix if

$$A^* A = A A^* \quad (6.1.7)$$

where A^* is the conjugate transpose of A . That is, $[A, A^*] = A A^* - A^* A = 0$ (they commute).

The important feature of normal matrices is that they are unitarily diagonalizable, i.e, there is a unitary matrix U ($U^* = U^{-1}$ by definition of unitarity) such that

$$U A U^* = \Lambda \quad (6.1.8)$$

where $\Lambda = \text{diag}(\lambda_1, \lambda_2, \dots)$, λ_i being the eigenvalues of A and the i -column of U the correspondent eigenvector of λ_i . For normal matrices, the eigenvectors form an orthonormal basis.

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Because $\hat{R}$ is a real matrix operator and by eq.(4.1.17), it is easily proved that the Blume matrix $\left(-\frac{i}{\hbar}H^\times + \hat{R}\right)$ is a normal matrix, which means that is always diagonalizable.

Now here is the tricky part. The Blume matrix is not hermitian. In fact, it will always have complex eigenvalues, as seen previously. But we are used to work with hermitian Hamiltonians so, for H hermitian, if λ is an eigenvalue with left eigenvector $\langle\chi|$ and right eigenvector $|\psi\rangle$ we have

$$\begin{aligned} H|\psi\rangle &= \lambda|\psi\rangle \\ \langle\chi|H &= \lambda\langle\chi| \end{aligned} \tag{6.1.9}$$

and if we do

$$\begin{aligned} (H|\psi\rangle)^* &= (\lambda|\psi\rangle)^* \iff \langle\psi|H^* = \lambda^*\langle\psi| \\ &\iff \langle\psi|H = \lambda\langle\psi| \end{aligned} \tag{6.1.10}$$

using the fact of H being hermitian, $H = H^*$ and $\lambda^* = \lambda$, we get that

$$|\psi\rangle^* = \langle\psi| = \langle\chi| \tag{6.1.11}$$

so, the left eigenvector $\langle\chi|$ is just the adjoint (the conjugate transpose) of the right eigenvector $|\psi\rangle$.

But if we have a non-hermitian normal matrix H' , doing the same approach

$$\begin{aligned} (H'|\psi\rangle)^* &= (\lambda|\psi\rangle)^* \iff \langle\psi|H'^* = \lambda^*\langle\psi| \\ &\iff \langle\psi|H' = \lambda^*\langle\psi| \neq \lambda\langle\psi| \end{aligned} \tag{6.1.12}$$

because λ can be complex, so $\lambda \neq \lambda^*$ if the imaginary part is non-null and H' is normal, so $H'^* = H'$.

This means that in this case

$$|\psi\rangle^* = \langle\psi| \neq \langle\chi| \tag{6.1.13}$$

so we cannot calculate the left eigenvectors just by doing the conjugate transpose of the right eigenvectors, as we were used in Quantum Mechanics.

Instead, we need to find the right eigenvectors and make them the columns of the unitary matrix U , which verifies eq.(6.1.8), and then calculate its inverse U^{-1} which is equal to U^* , and so the rows of U^{-1} will be the left eigenvectors.

As the Blume matrix is a non-hermitian normal matrix, this is the procedure we need to do to find the eigenvectors $|N_q\rangle$ and $|M_q\rangle$.

We were so used to the fact that usually in quantum mechanics we have $|\psi\rangle^* = \langle\chi|$ that it took us a lot of time and work to realize this detail about matrix diagonalization and be able to get the right results.

6.1.4. How To Do The Implementation of Frequency Distributions

In section 5.2.1 it was pointed the importance of inserting a frequency distribution in the perturbation function $R(t)$.

In our case, we considered the distribution of each state to be a Lorentzian (figure 6.1.2):

$$D(\omega) = \frac{1}{\pi} \frac{\frac{1}{2}F_{whm}}{(\omega - \omega_0)^2 + \left(\frac{1}{2}F_{whm}\right)^2} \quad (6.1.14)$$

where ω goes from $-\infty$ to $+\infty$, ω_0 is the central value where $D(\omega)$ has its maximum ($D(\omega_0) = \frac{2}{\pi F_{whm}}$), F_{whm} is the full width at half maximum and the distribution is normalized

$$\int_{-\infty}^{+\infty} D(\omega) d\omega = 1. \quad (6.1.15)$$

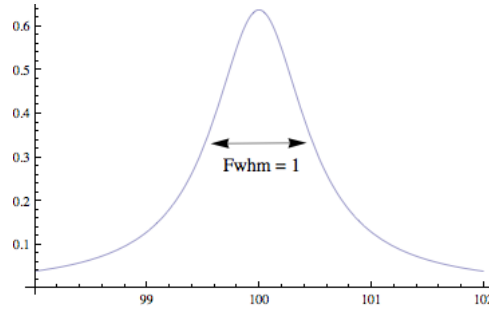


Figure 6.1.2.: Example of a Lorentzian with $\omega_0 = 100$ and $F_{whm} = 1$

In order to calculate $R(t)$ using equations 5.2.5, we know that we need to divide the distribution of each state in N intervals in such a way that, for integer¹ $k = -\frac{(N-1)}{2}, \dots, 0, \dots, \frac{(N-1)}{2}$, the N obtained quadrupole frequencies $(\omega)_k$ and the correspondent intervals' size $(\Delta\omega)_k$ obey eq.(5.2.3).

The problem is that from the start we only know the observable frequency ω_0 ($\omega_0 = 6\omega_Q$ for half-integer spin and $\omega_0 = 3\omega_Q$ for integer spin, where ω_Q is the quadrupole frequency (section 2.2.1)) and the value of F_{whm} , so we needed to figure out a way to find all the N frequencies for us to use.

¹In our distribution we always have a central value, corresponding to ω_0 , that we associate to $k = 0$ and an equal number of intervals with $\omega > \omega_0$ (associated to $k > 0$) and with $\omega < \omega_0$ (associated to $k < 0$), so N must be odd for all cases, and the limits of k are then justified.

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If we consider that for each frequency ω_k in the distribution, its area is delimited by $\omega_{initial} \equiv \omega_i$ and $\omega_{final} \equiv \omega_f$ such that $\omega_f > \omega_i$ and $\omega_k = \frac{(\omega_f + \omega_i)}{2}$, we have

$$\int_{\omega_i}^{\omega_f} D(\omega) d\omega = \frac{1}{N} \quad (6.1.16)$$

which basically represents the same as eq.(5.2.3), i.e., all intervals have the same area.

Substituting $D(\omega)$ by eq.(6.1.14) and solving the integral, it is straightforward to get

$$\omega_f = \frac{F_{whm}}{2} \tan \left(\frac{\pi}{N} + \arctan \left(\frac{\omega_i - \omega_0}{F_{whm}/2} \right) \right) + \omega_0. \quad (6.1.17)$$

Because ω_0 , F_{whm} and N are fixed values, we have an equation that gives us ω_f as a function of ω_i .

The procedure then will be:

ω_0 is the central frequency of the distribution ($\omega_0 = \omega_{k=0}$) and, from eq.(5.2.3) we know that

$$D(\omega_0) \Delta\omega_0 = \frac{1}{N} \quad (6.1.18)$$

so we can get $\Delta\omega_0$. Now, the value of ω_f for ω_0 will be

$$(\omega_f)_{k=0} = \omega_0 + \frac{\Delta\omega_0}{2} \quad (6.1.19)$$

Setting $(\omega_f)_{k=0}$ as $(\omega_i)_{k=1}$ for the next interval, by eq.(6.1.17) we can get the value of $(\omega_f)_{k=1}$.

The correspondent frequency of this interval will then be

$$\omega_{k=1} = \frac{(\omega_f)_{k=1} + (\omega_i)_{k=1}}{2} \quad (6.1.20)$$

with the correspondent probability $D(\omega_{k=1})$.

It's easy to see that this is an iterative method, where the values from one interval will give us the values of the next interval: $((\omega_f)_k = (\omega_i)_{k+1})$. Therefore, we get all the frequencies ω_k with $k > 0$ in the distribution but, because a Lorentzian is a symmetric function, to each $\omega_k > \omega_0$ will correspond a frequency $\omega_{-k} < \omega_0$ given by

$$\omega_{-k} = \omega_0 - |\omega_k - \omega_0|, \quad k > 0 \quad (6.1.21)$$

i.e., a frequency whose distance from ω_0 is the same that ω_k has: $|\omega_k - \omega_0| = |\omega_0 - \omega_{-k}|$, and so $D(\omega_{-k}) = D(\omega_k)$.

This procedure allows us to calculate all the N pairs $(\omega_k; D(\omega_k))$ from our distribution.

After calculating the frequency distributions for all different possible states a , we can use the N_a pairs $(\omega_k; D(\omega_k))_a$ in equations (5.2.5) to calculate the angular correlation functions $W(\theta, t)$.

6.2. Fit Routine

The concept of the fit routine is very simple.

First, it reads the parameter file, where we define what is the value of each parameter and if they vary or are fixed during the fitting procedure, and the file with the experimental data.

Then, it finds the parameters values that give the best $R(t)$ curve to fit the experimental data, using the simulation routine to calculate $R(t)$.

To do the fit, we make use of the so called “chi square” function, χ^2 .

6.2.1. χ^2 (chi-square function)

The chi-square function is defined as

$$\chi^2 = \frac{1}{N} \sum_{i=0}^N \left[\frac{(y_i - f(x_i))^2}{(\Delta y_i)^2} \right] \quad 0 < \chi^2 < \infty \quad (6.2.1)$$

where N is the number of data points, y is the independent variable, x is the dependent variable, f is the relationship between x and y , y_i is the “observed mean”, $f(x_i)$ is the “predicted mean” and Δy_i is the error in the experimental measurement of y_i . Of course f might have several variables (which means several different parameters), being represented as $f((x_1)_i, (x_2)_i, \dots)$.

In summary, the χ^2 function will compare the difference between our experimental data points y_i and the points given by the fitting curve $f(x_i)$ (that in our case is the $R(t)$ function) with the experimental error Δy_i .

If $\chi^2 \gg 1$, it means that $(y_i - f(x_i))^2 \gg (\Delta y_i)^2$ and so our experimental data is too different from the fitting curve, thus the fit is not good.

On the other hand, if $\chi^2 \ll 1$, it means that $(y_i - f(x_i))^2 \ll (\Delta y_i)^2$. At first glance we could think that this is the desired result, but the condition $(y_i - f(x_i))^2 \ll (\Delta y_i)^2$ doesn't necessarily mean that $(y_i - f(x_i))^2$ is small, making us believe that we have a good fit; it can also mean that Δy_i has really big values, which means the error in our measurements is

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so high that we can have lots of curves giving “good” results even if they are not even close to the experimental data.

Therefore, the best result we could have is to get a fitting curve and measurement errors good enough to give us $(y_i - f(x_i))^2 \approx (\Delta y_i)^2$, which means a predicted curve whose difference from our experimental data points is given by the measurement errors. With this condition we have

$$\left[\frac{(y_i - f(x_i))^2}{(\Delta y_i)^2} \approx 1 \right] \quad (6.2.2)$$

and so

$$\sum_{i=1}^N \left[\frac{(y_i - f(x_i))^2}{(\Delta y_i)^2} \right] \approx \sum_{i=1}^N 1 = N \quad (6.2.3)$$

yielding

$$\chi^2 = 1. \quad (6.2.4)$$

This way, when we are doing fits we aim to find the fitting curve that, compared to the experimental data, gives us $\chi^2 \approx 1$.

In TFPAC the fits are done using the minimization package TMinuit² from ROOT³. ROOT is an object-oriented program and library developed by CERN and written in C++. We used its functions the same way we used LAPACK in the diagonalization routine.

We define χ^2 as the function to be analysed and, considering the parameters that we defined as “fixed” or “variable”, the minimization package will find the values of the “variable” parameters that give the smallest value of χ^2 . When these values are found, they correspond to our fitting curve.

We are looking for the smallest value of χ^2 so we could have cases resulting in $\chi^2 \ll 1$, which are not good, as previously said. However, in this cases it means that the measurement error is not good, and so the problem is not the fit but the measurements. In this case we have to be very careful doing the fits or even think about re-doing the experiment.

Having this in mind, the minimization of χ^2 is a good tool to fit experimental data, and so it is the one we use in TFPAC.

²For more informations about TMinuit, <http://root.cern.ch/root/html/TMinuit.html>

³<http://root.cern.ch/>

7. Experimental Data and Analysis

Our purpose to create TFPAC was its potencial usefulness in real experiments, because currently it is the only program available to study dynamic interactions in PAC measurements without approximations in the construction of the perturbation function $R(t)$ in single crystals.

So, the last part of this work shows few examples of the use of TFPAC to analyse real experimental data.

7.1. Studying “after-effects” on doped GaN

Semiconductors are well known to be among the more studied worldwide materials due to their applications in daily life.

One major interest in semiconductors concerns their electric properties and, to take advantage of them, we need to dope these materials with electrically active impurity atoms. During the doping procedure there is always the possibility of including unintentional defects. For instance, doping by implantation is always accompanied by the creation of lattice defects which have to be removed by thermal treatment of the sample which, on the other hand, increases the risk of diffusion of impurity atoms into the material (ref.[20]). Therefore, the understanding of the nature and physical properties of these defects and the electrical environment of each site of a semiconductor is of key importance.

$\gamma - \gamma$ PAC has been intensively used to study semiconductors using different probe elements with isotopes of suitable cascades for the technique, as $^{111}In/^{111}Cd$, $^{111m}Cd/^{111}Cd$, $^{181}Hf/^{181}Ta$, etc. Today, at ISOLDE, semiconductors are actively studied combining $\gamma - \gamma$ and $e^- - \gamma$ PAC looking forward to understand defects and activation of In doped nitrides (ref.[21]).

In this experiment, we studied GaN (gallium nitride), a wide bandgap semiconductor material (3.4 eV bandgap at 300 K) widely used in transistors, solar cells and other optoelectronic devices, e.g., in blue light lasers.

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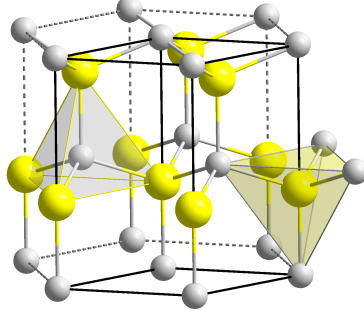


Figure 7.1.1.: Gallium Nitride

Two types of epitaxial thin films, grown over sapphire were used: GaN (intrinsic -n) and $GaN(Mg \text{ doped})$, performing measurements with $e^- - \gamma$ and $\gamma - \gamma$ Perturbed Angular Correlations. For such task the radioactive $^{181}Hf/^{181}Ta$ probe, which has $T_{1/2} = 42.4$ days, was implanted into the samples at the Institute für Strahlen-und Kernphysik, University of Bonn, Germany, and the PAC measurements were done at ISOLDE-CERN. In the sample preparation, ^{181}Hf was implanted at 80 keV with a final dose of around $3 \times 10^{13} \text{ ions/cm}^3$. Post-implant annealings were performed with $T_A = 1000^\circ C$, $120s$ RTA in nitrogen flow with proximity cap.

E. Alves et al. [22] showed that Hf is incorporated into substitutional Ga sites immediately after the implantation. Emission channeling experiments were also performed and they corroborate this result. The actual experiments have not been done during this work. We are using this data since it can only be properly analysed with TFPAC.

^{181}Hf decays by a cascade to ^{181}Ta , allowing us to do both $e^- - \gamma$ or $\gamma - \gamma$ PAC measurements, as can be seen in figure 7.1.2.

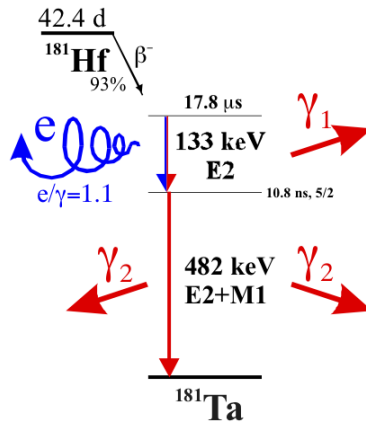


Figure 7.1.2.: $^{181}Hf/^{181}Ta$ cascade [23]

For $\gamma - \gamma$ experiments γ_1 has energy 133 keV and γ_2 has energy 482 keV . For the $e^- - \gamma$, the conversion electron $<L>$ with energy $(133 \text{ keV} - 11 \text{ keV}) = 122 \text{ keV}$ was used.

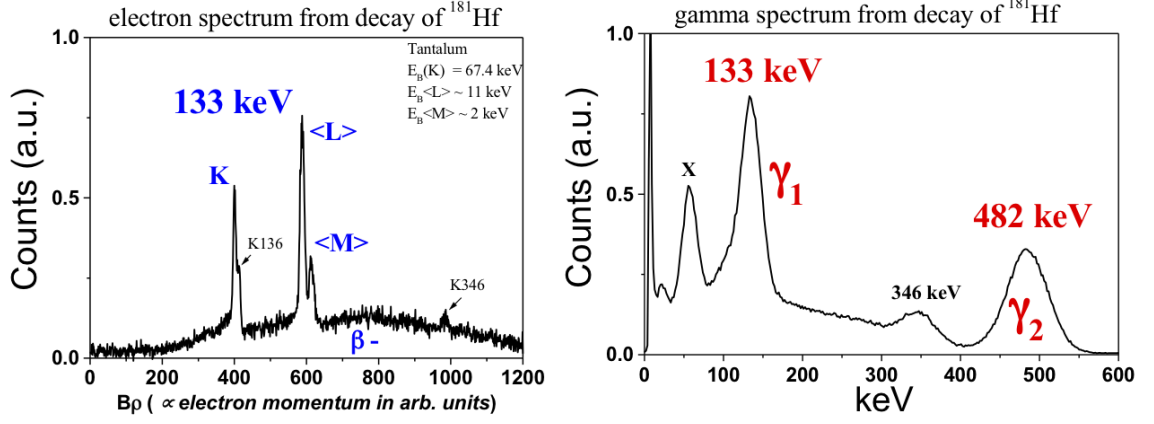


Figure 7.1.3.: e^- and γ spectrum from decay of ^{181}Hf [23]

The GaN epitaxial thin films are single crystalline materials and so the EFG from the lattice has a well defined orientation, which means that we need to know the correspondent Euler angles to represent the coordinate system of the crystal in the coordinate system of the detectors. In this case we have the z-axis of the crystal coordinate system in the detectors plane and forming a 45° angle with each detector.

Measurements were done at room temperature (about 25°C), 300 K and then the temperature was lowered, being registered values from several points down to 12 K .

Now, let's focus on the questions “What are after-effects?” and “Why to measure them combining $e^- - \gamma$ with $\gamma - \gamma$?”

After-effects of hole formation due to internal conversion, electron capture and α - and β -decay, present immediately after radioactive decay, have been studied since the sixties (ref. [24] (1967) and ref.[21]).

In this experiment, the aim is to create an electronic defect by the loss of a conversion electron from the lower atomic orbitals and see how the material will provide electrons to fill in the gap, eventually detecting deep excited states associated with the material doping - after effects. These conversion electrons are detected in $e^- - \gamma$ measurements, but not in $\gamma - \gamma$ so, to the latter the defects will be “invisible”.

Combining $e^- - \gamma$ with $\gamma - \gamma$ is the perfect tool to study this kind of phenomena whereas $\gamma - \gamma$ gives us the information about the defectless nuclei while $e^- - \gamma$ shows the existence and the features of the defects, created on purpose.

7. Experimental Data and Analysis

$\gamma-\gamma$ PAC only will be sensitive to the material-probe interactions, which are due to intrinsic properties and intrinsic defects changing as a function of temperature.

$e^- - \gamma$ PAC will provide extra information on electronic dynamics, since it creates ionized states just before the measurement starts, deep into the material.

7.1.1. Comparing $e^- - \gamma$ with $\gamma - \gamma$

The results of both $e^- - \gamma$ and $\gamma - \gamma$ experiments are presented, in Fig.(7.1.4(a)) for n-type *GaN* and in Fig.(7.1.4(b)) for *GaN* doped with *Mg*.

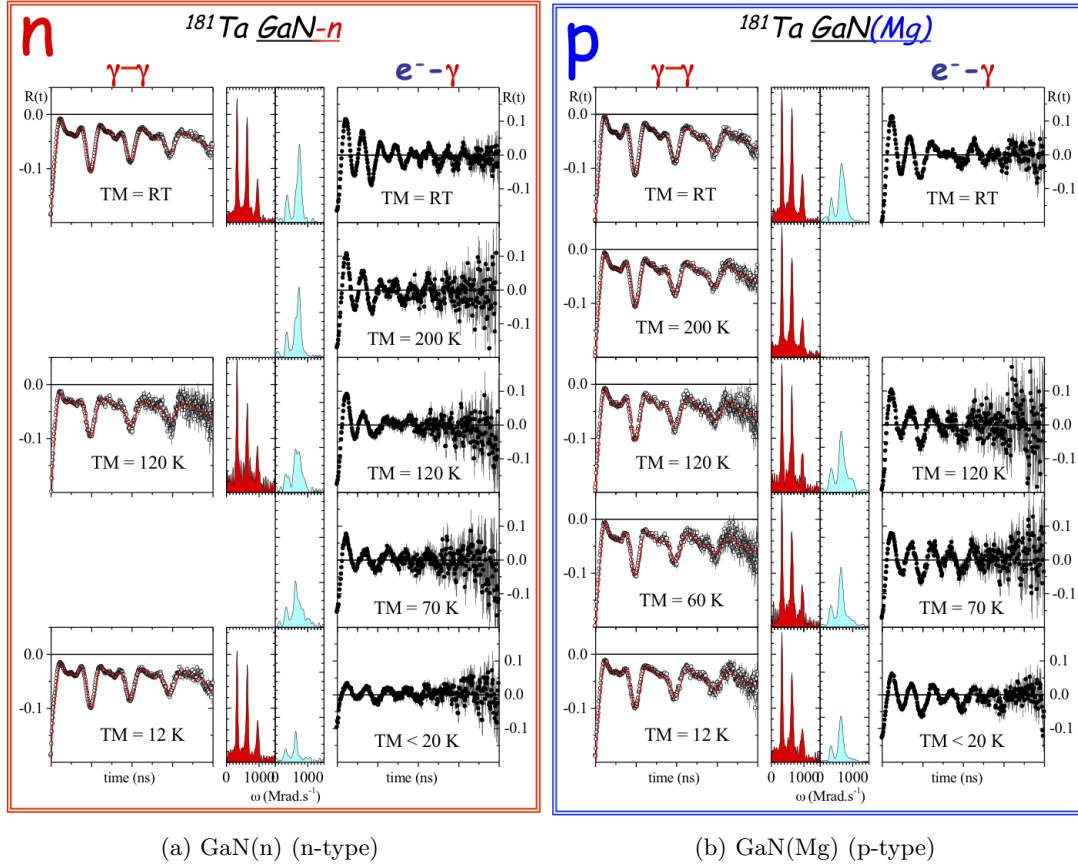


Figure 7.1.4.: $\gamma - \gamma$ and $e^- - \gamma$ results of *GaN*(*n*) and *GaN*(*Mg*) [23]

The middle column in each figure represents the Fourier transform of the data, thus giving us the frequencies that exist in every function, and by expression (4.2.7) we can see that this tells us the changes in the electric field gradient.

It is straightforward to observe that for either the n-type or the p-type, the $\gamma - \gamma$ results

are the same for every temperature. This happens because there is no creation of electronic “holes” in the emission of γ -radiation, thus we only observe the intrinsic behavior of the interaction between the probe and the host. If there are no changes in the lattice with the variation of temperature, there are no changes in the $\gamma - \gamma$ PAC spectrum. However, in the $e^- - \gamma$ measurements we definitely have big changes for different temperature values. This means that in the production of the conversion electron that we detect there are changes in the nuclear electronic environment whose response is highly sensitive to temperature changes. Although this process is invisible for γ -ray Perturbed Angular Correlations, the production of conversion electrons makes it possible to observe with $e^- - \gamma$ PAC measurements, so these are the results we really focused on.

7.2. Analysing the Experimental Data - “How have we done the fits?”

We started by the results obtained for $GaN(n)$.

Without any analysis, just looking to the results, we can see two things:

- We have dampings in the amplitudes of the $R(t)$ functions and
- the spectra reveal the presence of different frequencies, i.e, the presence of different electric field gradients (EFG).

As seen before, dampings in $R(t)$ have two possible sources: the distribution in the frequencies ω_Q (section 5.2.1) and the existence of dynamic interactions. For the distribution in the frequencies we observe a damping along the time, but for dynamic interactions we have dampings in the anisotropy coefficients, i.e., in the amplitudes of $R(t)$, which is the case observed in our results. Gathering it with the observed changes in the frequency, this makes us believe right from the start that we have a dynamic system¹.

Before trying any kind of fit, we need to think about what we know from our system in order to get some hints about how to do it.

After emitting the conversion electron, the electronic environment of the nucleus is in a very unstable state, with a hole in a low atomic orbit and the electrons of the higher orbits trying to recover it. This makes different nuclei to have slight differences in the EFG’s. As the conversion electrons give us the “start” on the clock of $e^- - \gamma$ PAC measurements, we know that at the time $t = 0$ we should have a state with a very big frequency distribution.

¹However, we always need to be careful when assuming that a damping reflects dynamic’s existence, because of the attenuations induced by big frequency distributions.

7. Experimental Data and Analysis

The nuclei will then recover to a stable state, so we should see a transition from the initial state to that final state.

With these ideas in mind, our method was:

- At high temperature (573 K) the transition must be fast so we just see the final state. Use that final state to all temperatures;
- Go to the lower temperatures and find the initial state with big frequency distribution that best describes the initial sharp climb of $R(t)$;
- Fit the results to find the activation frequency (the probability of transition from the initial state to the final state) for all the temperatures².

On our first attempt to do the fits we got different activation frequencies for each temperature, but also slightly different quadrupole frequencies in the final state.

The results for $GaN(n)$ are in Fig.(7.2.1(a)) and, as we can see, the fits are not good.

There are two important features to notice:

- The quadrupole frequencies we are getting in the final states correspond to the very first oscillations in the curve, but from the middle of the experimental time until the end, the frequency gets smaller. In fact, this smaller frequency seems to be very similar at all temperatures
- We should get a bigger decrease in the amplitudes and that is not happening.

At first glance, these results seem to be quite bad, but with the pointed features they turned out to be the origin of our resolution! Our interpretation is that there might exist an intermediate state that we were not counting, so this led us to our final approach:

We considered 2 fractions (a diagram explaining the configuration is shown on Fig.(7.3.2)), both of them have the same final state, with fixed quadrupole frequency given by the value we got at high temperature.

For the initial state:

1. In the first fraction we have exactly the same initial state considered before
2. In the second fraction we start in the “intermediate” state, considering that it has the quadrupole frequency that the first fit gave as the final state.

²This step included the higher temperature, because although in the beginning we considered the existence of only one state at this temperature and then used it as the final state for the other temperatures, if we consider also that there is a transition at this point, we get a very high activation frequency (which means a fast transition), but the fit is better.

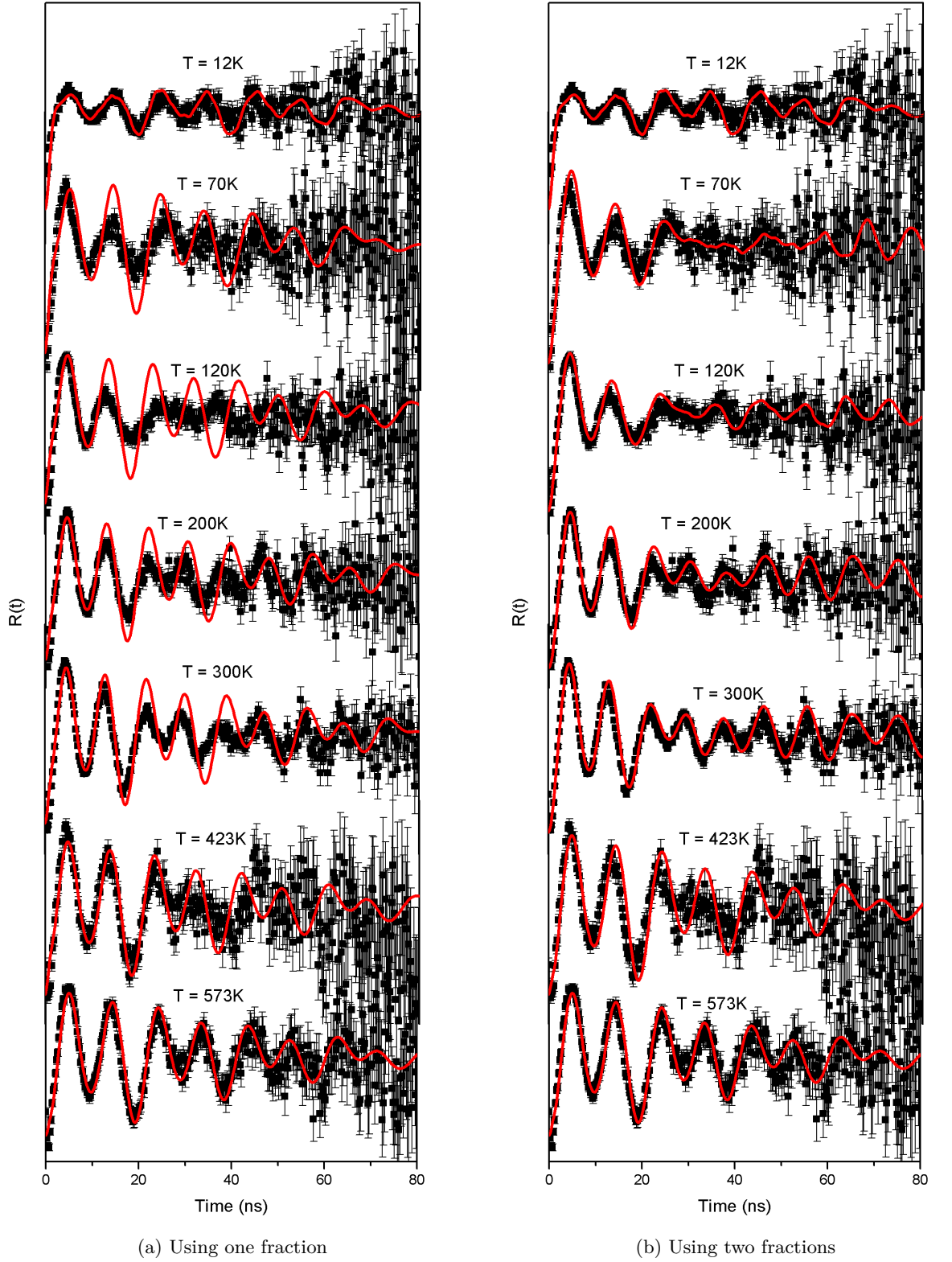


Figure 7.2.1.: Fits of $GaN(n)$. In *a*) we considered $I_{initial} \rightarrow I_{final}$ and in *b*) we have two fractions, one considering $I_{initial} \rightarrow I_{final}$ and the other considering $I_{intermediate} \rightarrow I_{final}$.

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Hereupon, we did the fits for each temperature and found the activation frequency for each fraction.

The results obtained are far better than the previous ones and are represented in Fig.(7.2.1(b)).

The initial state and the final state are considered to be the same for all temperatures and the correspondent values are shown on Table (7.1).

The intermediate state is different for each temperature, so on Table (7.2) we have its correspondent values depending on temperature, as well as the activation frequencies³ Γ_f given for fraction one ($I_{initial} \rightarrow I_{final}$) and for fraction two ($I_{intermediate} \rightarrow I_{final}$).

$\omega_0(i)$	$V_{zz}(i)$	$\eta(i)$	$F_{whm}(i)$	$\omega_0(f)$	$V_{zz}(f)$	$\eta(f)$	$F_{whm}(f)$	α	β	γ
526.0	18.68	0.0	1500.0	316.0	11.22	0.134	40.0	45°	90°	0°

Table 7.1.: Quadrupole frequency ω_0 ($Mrad.s^{-1}$), electric field gradient V_{zz} ($V.\text{\AA}^{-2}$) (see eq.(2.2.28), considering $I = 5/2$ and $Q = 2.36 \text{ barn}$), asymmetry parameter η , distribution width F_{whm} ($Mrad.s^{-1}$) and Euler angles α , β , γ for the initial state (i) and the final state (f), in $GaN(n)$. These values are the same for all temperatures.

	Fraction 1	Fraction 2				
T	$\Gamma_{f_1}(i \rightarrow f)$	$\omega_0(int.)$	$V_{zz}(int.)$	$\eta(int.)$	$F_{whm}(int.)$	$\Gamma_{f_2}(int. \rightarrow f)$
12 K	60.0	-	-	-	-	-
70 K	10.0	324.0	11.50	0.134	100.0	10.0
120 K	20.0	345.0	12.25	0.134	100.0	20.0
200 K	30.0	360.0	12.78	0.134	40.0	30.0
$RT (\sim 300 K)$	30.0	368.0	13.06	0.134	40.0	30.0
423 K	40.0	340.7	12.10	0.134	40.0	40.0
573 K	60.0	328.0	11.64	0.134	40.0	60.0

Table 7.2.: Activation frequencies Γ_f (MHz) for each fraction, correlated with the probability of transition: in f_1 from the initial state to the final state and in f_2 from the intermediate state to the final state. The quadrupole frequency ω_0 ($Mrad.s^{-1}$), the V_{zz} ($Mrad.s^{-1}$), the asymmetry parameter η and the distribution width F_{whm} ($Mrad.s^{-1}$) for the intermediate state in each temperature are also given. ($GaN(n)$).

³The definition of the activation frequencies is given from $\omega_{a \rightarrow b} = \Gamma_f(a \rightarrow b) \frac{4I(2I-1)}{2\pi k}$, where $k = 3$ (for spin I integer) and $k = 6$ (for I half-integer), being $\omega_{a \rightarrow b}$ the probability of transition from a state "a" to a state "b". This way, looking to eq.(2.2.31), we can compare $\omega_{a \rightarrow b}$ to ν_Q and $\Gamma_f(a \rightarrow b)$ to ω_0 .

7.2. Analysing the Experimental Data - “How have we done the fits?”

Even though it was the highest temperature (573 K) to initially give us information about the final state, we got a better fit considering also the existence of $I_{intermediate}$ in this point, and not just a fast transition $I_{initial} \rightarrow I_{final}$.

For all the temperatures, the fraction one is considered to be 40% of the ensemble and the fraction two 60%, except for the lowest temperature (12 K), where we just needed to consider the fraction ($I_{initial} \rightarrow I_{final}$). At this temperature we got a higher activation frequency, but this is due to the fact of not considering nuclei in the intermediate state, therefore, even the value of the activation frequency seeming to represent a quicker transition, at 12 K the recovery is slower.

For $GaN(Mg)$, the analysis of the experimental data couldn't be completely done, so the results must be seen cautiously.

Because of this lack of time we had to be pragmatic, so we based our analysis in the method used for $GaN(n)$, using the following considerations:

- The same two fractions were considered, the first referring to $I_{initial} \rightarrow I_{final}$ and the second referring to $I_{intermediate} \rightarrow I_{final}$;
- The initial and the final state are ideally similar for both samples so, for $GaN(Mg)$, we considered them to be equal to the results of $GaN(n)$ (Table 7.1). Thus, we only considered variations in the intermediate state and in the activation frequencies for the transitions.

The obtained results are represented on Fig.(7.2.2(a)).

The intermediate state was observed for all range of temperatures, except for the higher temperature (568 K), where the transition to the final state is fast enough.

Although the results seem to be considerably good, the percentage obtained for the fractions were 80% for the fraction ($I_{initial} \rightarrow I_{final}$) and 20% for the fraction ($I_{intermediate} \rightarrow I_{final}$), which means that for $GaN(Mg)$ the intermediate state is of pour interest, so we decided to redo the fits without “forcing” the existence of intermediate state and only considering the transition $I_{initial} \rightarrow I_{final}$. The results are shown on Table (7.3) and are represented on Fig.(7.2.2(b)).

The initial states obtained have a slight variation depending on temperature but the frequency distributions are much smaller than the obtained for $GaN(n)$, which means that the electronic recovery is much faster in this case. This is also verified by the bigger values on the activation frequencies, representing a faster transition to the final state.

In the higher temperature, due to the fast recovery, we cannot see the transition and so we only observe the static final state.

7. Experimental Data and Analysis

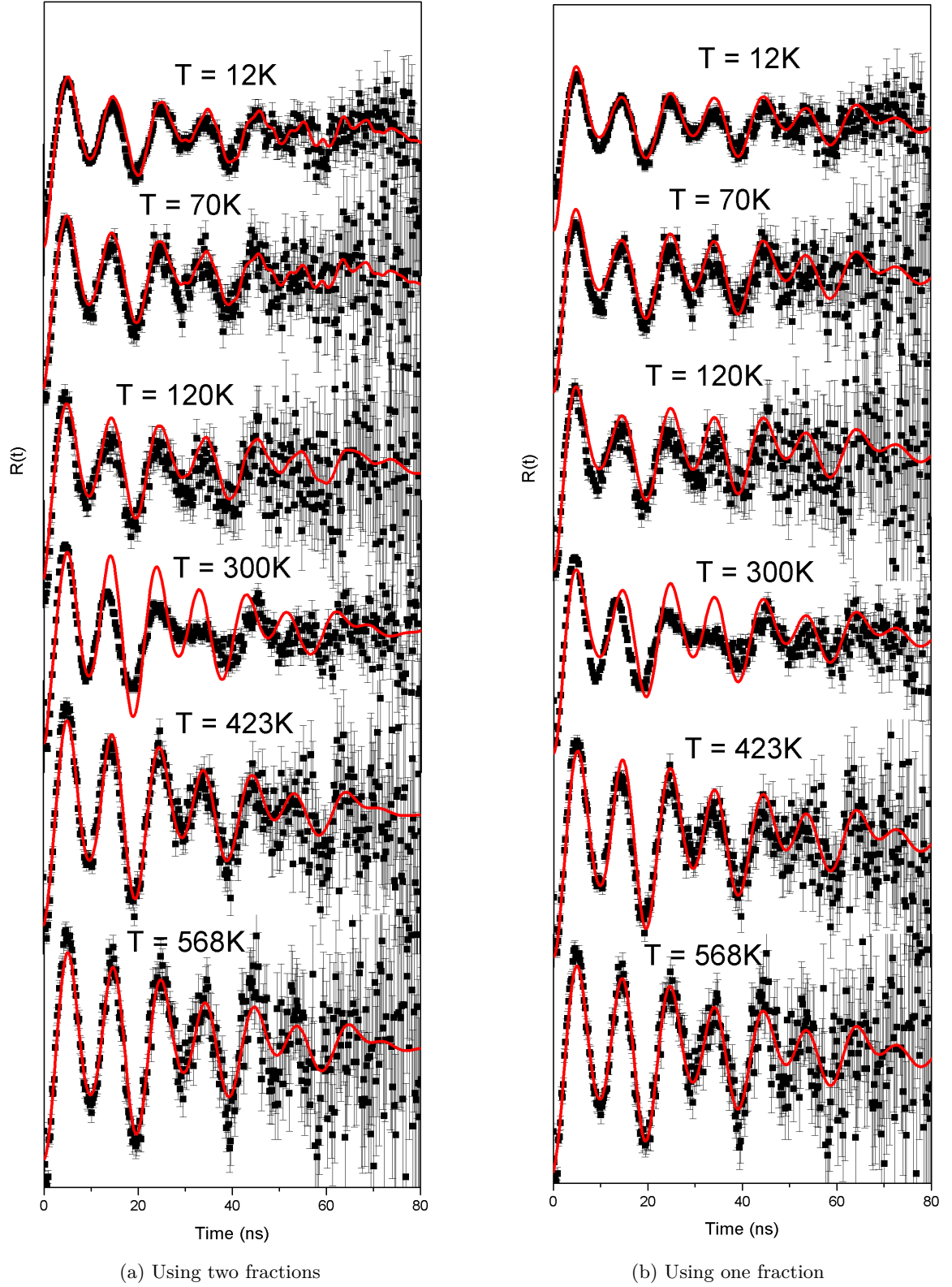


Figure 7.2.2.: Fits of $GaN(Mg)$. In *a*) we have two fractions, one considering $I_{initial} \rightarrow I_{final}$ and the other considering $I_{intermediate} \rightarrow I_{final}$. In *b*) we considered only $I_{initial} \rightarrow I_{final}$

7.2. Analysing the Experimental Data - “How have we done the fits?”

T	$\omega_0(i)$	$V_{zz}(i)$	$\eta(i)$	$F_{whm}(i)$	$\omega_0(f)$	$V_{zz}(f)$	$\eta(f)$	$F_{whm}(f)$	$\Gamma_{i \rightarrow f}$
12 K	321.0	11.40	0.134	260.0	321.0	11.40	0.134	40.0	80.0
70 K	323.0	11.46	0.134	260.0	321.0	11.40	0.134	40.0	95.0
120 K	328.0	11.64	0.134	260.0	321.0	11.40	0.134	40.0	105.0
RT ($\sim 300K$)	330.0	11.72	0.134	260.0	321.0	11.40	0.134	40.0	140.0
423 K	321.0	11.40	0.134	260.0	321.0	11.40	0.134	40.0	600.0
568 K	-		-	-	321.0	11.40	0.134	40.0	-

Table 7.3.: Quadrupole frequency ω_0 ($Mrad.s^{-1}$), electric field gradient V_{zz} ($V.\text{\AA}^{-2}$), asymmetry parameter η and distribution width F_{whm} ($Mrad.s^{-1}$) for the initial state (i) and the final state (f), in $GaN(Mg)$, with the correspondent activation frequency $\Gamma_{i \rightarrow f}$. The Euler angles are $\alpha = 45^\circ, \beta = 90^\circ, \gamma = 0^\circ$ for all states.

In the available time for analysis we were not able to do a proper fit to the results at room temperature for $GaN(Mg)$. Observing the intermediate state obtained for $GaN(n)$ we can see that its quadrupole frequency gets a higher value near room temperature, so the phenomena that gives origin to this state might be stronger at this point, and it might be present some how in $GaN(Mg)$ at this temperature. Unfortunately the current analysis doesn't provide us more information to understand this fact.

The Euler angles α, β, γ are defined according to Fig.(6.1.1) and the arrangement of the detectors was considered as having the detection of γ_1 (or e^-) given along the x axis and the detection of γ_2 given along the y axis for $W(90^\circ, t)$ and along the x axis, but in opposite direction of γ_1 (or e^-), for $W(180^\circ, t)$ (which means that in Fig.(3.2.1) we have $\theta_1 = 90^\circ, \phi_1 = 0^\circ$ and, for $W(90^\circ, t)$: $\theta_2 = 90^\circ, \phi_2 = 90^\circ$; for $W(180^\circ, t)$: $\theta_2 = 90^\circ, \phi_2 = 180^\circ$).

The used “effective” attenuation coefficients (defined by eq.(5.3.3)) are presented on table 7.4.

a_{22}	a_{24}	a_{42}	a_{44}
-0.178	-0.205	-0.00128	-0.00148

Table 7.4.: “effective” attenuation coefficients.

To implement the frequency distributions (section 6.1.4) we had to define the number of intervals for the distribution functions to be divided in. For the initial state, because the distribution width at half-maximum is very high we had to consider a great number of intervals, so it has been divided in 51 intervals. For the final state and the intermediate state the distribution width is much smaller, so we just needed to consider, for the initial state, 21 points, and for the intermediate state, 11 points.

It was considered a time resolution with $F_{whm(prompt)} = 0.9 ns$.

7.3. From The Results To The Physical Phenomena

$e^- - \gamma$ PAC measurements are the ones who reveal the dynamics we wanted to study, hereby we will only discuss the $e^- - \gamma$ PAC results.

After the radioactive probe is implanted in our sample, the decay from the nuclear excited state is done by γ -decay (used in the $\gamma - \gamma$ correlations) but we also have conversion electrons (appendix A.6) that are used in the $e^- - \gamma$ correlations.

When an innermost orbital electron is ejected by electron conversion, an electron from a higher orbital will drop to a lower state to fill the vacancy, accompanied by the emission of characteristic x-rays, whose photons can interact with other electrons and, by “Auger effect”, Auger electrons can be ejected (appendix A.6). To help visualizing the phenomena, a schematic representation is done in Fig.(7.3.1).

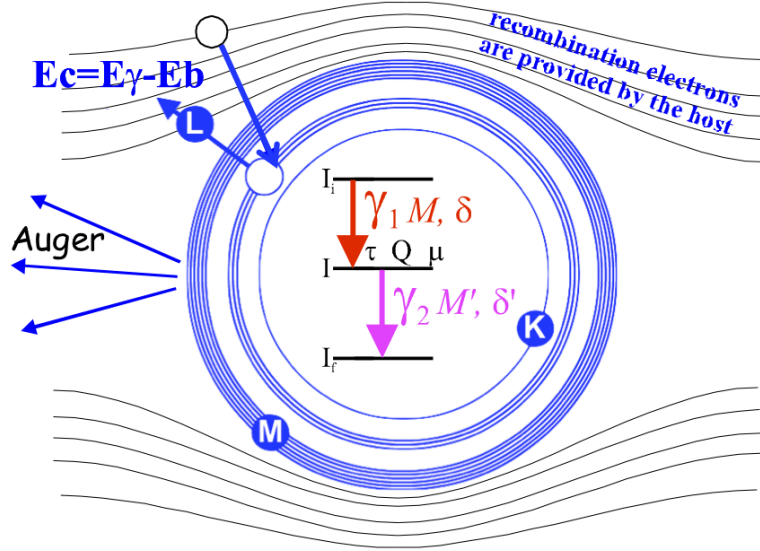


Figure 7.3.1.: Internal Conversion Electrons (in this case from L-shell) and Auger Electrons

All this processes together will create an external charge difference in each nucleus, so they will feel different electric potentials. Because the electric field gradient (EFG) is the second derivative of the electric potential (see section 2.2.1) this means that when we detect the electron e^- that gives the “start” in our PAC measurements, different nuclei will have slight different EFG’s, justifying the initial state with a huge distribution in the EFG.

After that, the system will recover until it gets to a final stable state, which is the final state that we observe, being the same for all temperatures.

The recovery process that contributes to the transition from the initial state to the final state only depends on the ease with which the electrons from the higher orbitals fill the vacancies left by the conversion electrons. Obviously, the easier the electrons “move” in the

material, the faster they recover those vacancies. Because the movement of the electrons is highly influenced by the temperature, the higher the temperature, the faster the recovery will be. Thus, we get an increase of the activation frequency when we have an increase in temperature, corresponding exactly to the experimental results we have got.

We should make a brief comment about the initial conditions: although we said that at $t = 0$ all nuclei are in the initial state, there might be a statistical transition to the final state faster than the transition we can see experimentally. This way, to do the fits we didn't consider that all the nuclei were in the initial state but only 90% of them were. The other 10% we considered to already be in the final state.

Now, we must compare the two samples: $GaN(n)$ and $GaN(Mg)$.

On the first one we have the intrinsic semiconductor GaN , but the lack of some atoms of Ga transforms it in a n-type semiconductor. On the second one, implanting Mg , we are supposed to obtain a p-type semiconductor, however this has not been proved.

For $GaN(n)$ we have an initial state with higher frequency distribution than for $GaN(Mg)$, which means that after the loss of the conversion electrons and the production of the Auger electrons, the recovery is much faster in the second sample. It is also observed that the transition $I_{initial} \rightarrow I_{final}$ is quicker for $GaN(Mg)$ (it has bigger activation frequencies), so both results mean that in $GaN(Mg)$ we have more electrons available. This is a contra-sense because if we had a p-type semiconductor we should have less electrons. Our idea is that when implanting the Mg atoms into our material, some defects might be formed, and these defects are the cause for the excess of electrons in this case.

Besides the initial state and the final state we also have the presence of an "intermediate" state in $GaN(n)$.

Thus, at $t = 0$ we considered a fraction of nuclei to be in the called initial state and the rest of the nuclei to be in the intermediate state (diagram in Fig.7.3.2).

The intermediate state has a bigger half-life time and a smaller distribution in frequency than the initial state but it's still not stable enough so, we also have a transition to the final state in these nuclei. This means that, in the end, all nuclei of the material will be in the same state.

According to the cascade represented on Fig.(7.1.2), the ion Hf^{4+} gives result to Ta^{4+} by β^- decay and, after the double cascade, we are suppose to have stable Ta^{3+} in the place of Ga^{3+} ions. Due to the lack of electrons, some Ta^{4+} ions might remain present during a longer interval of time, and so they are the reason for the existence of the intermediate state.

7. Experimental Data and Analysis

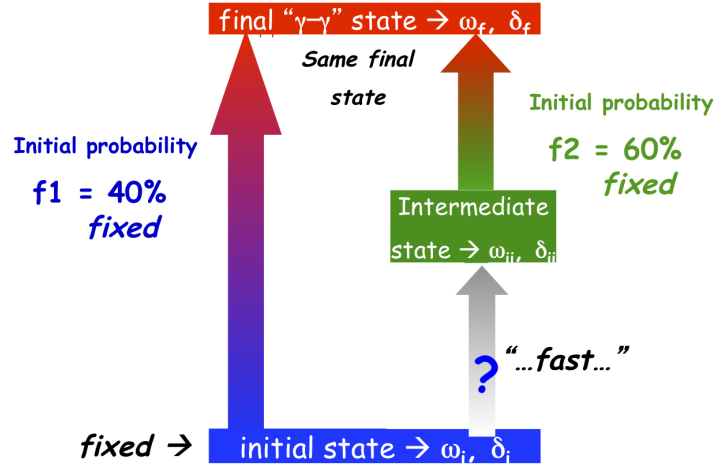


Figure 7.3.2.: Diagram explaining the configurations of the two different fractions for $GaN(n)$ [23].

In $GaN(Mg)$ we have more electrons available, thus there won't be Ta^{4+} ions remaining present enough time to give result to the intermediate state, so the latter is not observed (note that this is a conjecture because the analysis is not conclusive enough).

Because our starting point ($t = 0$) is given by the measurement of the conversion electron e^- and it is the loss of this electron that leaves the nucleus in an unstable state, the existence of this intermediate state in this first instant might tell us that some nuclei have a faster initial recover. This leaves the question: Is there a transition from the initial state to the intermediate state, $I_{initial} \rightarrow I_{intermediate}$, that is fast enough so we can barely see it in our measurements? (Fig.7.3.2)

To answer this question we should consider, not only the transitions $I_{initial} \rightarrow I_{final}$ and $I_{intermediate} \rightarrow I_{final}$, but also the transition $I_{initial} \rightarrow I_{intermediate}$ and use the three in the construction of our Blume matrix, considering that at $t = 0$ all the nuclei were at $I_{initial}$, instead of the use of two different fractions as we did, which is not a perfect approximation since we considered the same percentage of nuclei in the intermediate state for all temperatures and that number might vary in function of the temperature. Due to computational difficulties (speed and memory) this could not be done in time, so this is our leading point for future work.

We think that the obtained results allow us to understand what might be happening in the electronic recovery after the loss of a conversion electron, but our idea of the phenomena involved cannot be fully proved, so further work must be done.

8. Conclusion and Final Comments

We have started this thesis by referring to the nuclear properties. The definition of nuclear moments (magnetic dipolar moment and electric quadrupole moment) and its interaction with electromagnetic fields external to the nucleus was given, as well as the angular distribution of radiation produced by γ -decay.

Then we have introduced the Perturbed Angular Correlation technique, with a detailed description of the theoretical expressions leading to the angular correlation observables. We compared the $\gamma - \gamma$ correlations with the $e^- - \gamma$ correlations, referring the advantage of experimentally comparing both results.

Phenomena generally analysed with PAC, like defect structure or local deformations, are associated with static electric gradient fields, but atomic and electronic dynamics might be observed in angular correlations, thus a description of PAC using dynamic Hamiltonians must be used. For such purpose, we studied the theory of stochastic perturbations in angular correlations, presenting the fundamental calculations necessary for getting the correlation function in such cases.

The implementation of the experimental correlation function $W(\mathbf{k}_1, \mathbf{k}_2, t)$ was done, and used to construct the anisotropy ratio function, analogous of the experimentally used “perturbation function” $R(t)$.

The existence of static distributions given by distributions on the quadrupole frequencies (due to lattice imperfections and impurity elements distributions) was justified. To cope with experiments, a finite time resolution is mathematically incorporated in the simulated observable function.

The kernel of this work summarized the implementation of a complex C++ program (TF-PAC) that reproduces and fits the experimental data.

The most important steps of TFPAC, such as the rotation matrix for non-zero Euler angles, the diagonalization of the Blume matrix and the implementation of static distributions, were explained.

Finally, an experimental case study was presented and analysed, being justified the search

8. Conclusion and Final Comments

of “after-effects” on doped *GaN*. We tentatively draft an idea behind the observed physical phenomena.

Due to the short time left between the end of writing and debugging of TFPAC, we have only presented a first draft of the data analysis of the cases of $^{181}\text{Ta} : \text{GaN}(n)$ and $^{181}\text{Ta} : \text{GaN}(\text{Mg})$.

“After-effects” were observed only on $e^- - \gamma$ PAC measurements thanks to the produced conversion electrons from the innermost orbits. The electronic recovery was observed to be quicker on *GaN*(Mg) due to an excess of electrons, which we suppose to be caused by defects from the implantation of Mg but we cannot be sure with this brief analysis.

We found a “long lived” excited state for Ta on *GaN*(n) but we cannot much detail on its phenomenology without further progress by implementing a 3 state transition Hamiltonian.

Due to lack of time, we were not able to properly analyse the obtained results, specially for *GaN*(Mg). This lack is essentially due to computational difficulties. The calculations performed in this work are very heavy (specially during the fits), so other calculation methods might be considered in future work (e.g., parallel computing). The biggest problem is the way we do the frequency distributions (sections 5.2.1) because we are using 51 points in the distribution of the initial state, 21 points for the final state and 11 points for the intermediate state so, with all the possible transitions, we need to calculate $51 \times 21 + 21 \times 11 = 1302$ times the eigenvalues and eigenvectors of the Blume matrix (which is a complex matrix of rank 72) and keep them in memory until the final calculation of $R(t)$ for all time intervals (usually around 450 in this work). If we are fitting data, this process is done for about 40 times, thus it’s not difficult to see how time and memory consuming the all calculation is.

Although there is some work to be done for a full understanding of the physical phenomena involved, the results we have got so far are very encouraging for the work to come.

TFPAC is all done using C++ but a graphical interface was done using JAVA (with the aim of being “operative system”-independent). Unfortunately, the interface could not be finished, being in a very early stage, and it’s only working for simulations and not for fits.

To finish, the aim of this work was the development of a program that was capable to simulate and fit data of perturbed angular correlations for dynamic systems with different possible states of electric field gradients in single crystal systems, with any orientation for each state, by *ab initio* calculations and to use it to analyse experimental data. This goal was achieved succesfully and now the objective is to make TFPAC suitable for even more possible states, using not only electric interactions but also magnetic interactions, to be faster during calculations and to have a graphical interface easy for anyone to use, not only in simulations mode but also in fitting mode.

A. Appendix

A.1. Clebsh-Gordan Coefficients and 3j-Symbols

The coupling of two angular momenta j_1 and j_2 to the total angular momentum j_3 is given by

$$\mathbf{j}_3 = \mathbf{j}_1 + \mathbf{j}_2 \quad (m_3 = m_1 + m_2) \quad (\text{A.1.1})$$

The states of the entire system can be represented either by the products $|j_1, m_1\rangle |j_2, m_2\rangle$ or by $|j_3, m_3\rangle$, thus there is a linear relation between the two representations, which is given by

$$|j_3, m_3\rangle = \sum_{m_1, m_2, m_3=m_1+m_2} \langle j_1, j_2, m_1, m_2 | j_3, m_3 \rangle |j_1, m_1\rangle |j_2, m_2\rangle \quad (\text{A.1.2})$$

The expansion coefficients are called Clebsch-Gordan coefficients. Often one uses the the Wigner 3j-symbols since they possess higher symmetries. The 3-j-symbols are associated with the Clebsch-Gordan coefficients by the following relationship (ref.[3])

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = (-1)^{j_1-j_2-m_3} \frac{1}{\sqrt{2j_3+1}} \langle j_1, j_2, m_1, m_2 | j_3, -m_3 \rangle \quad (\text{A.1.3})$$

For non-null solutions, the following rules must be satisfied:

$$\begin{cases} m_1 + m_2 + m_3 = 0 \\ j_1 + j_2 + j_3 \quad \text{integer} \\ |m_i| \leq j_i \\ |j_1 - j_2| \leq j_3 \leq j_1 + j_2 \end{cases} \quad (\text{A.1.4})$$

The calculation of these coefficients was done by E. P. Wigner (1931) in a very extensive mathematical analysis, getting the result

A. Appendix

$$\begin{aligned}
\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} &= \sqrt{\frac{j_1 + j_2 - j_3!(j_1 - j_2 + j_3)!(-j_1 + j_2 + j_3)!}{(j_1 + j_2 + j_3 + 1)!}} \times \\
&\times \sqrt{(j_1 + m_1)!(j_1 - m_1)!(j_2 + m_2)!(j_2 - m_2)!(j_3 + m_3)!(j_3 - m_3)!} \times \\
&\times \sum_z \left(\frac{(-1)^{z+j_1-j_2-m_3}}{z!(j_1 + j_2 - j_3 - z)!(j_1 - m_1 - z)!(j_2 + m_2 - z)!} \times \right. \\
&\times \left. \frac{1}{(j_3 - j_2 + m_1 + z)!(j_3 - j_1 - m_2 + z)!} \right) \quad (A.1.5)
\end{aligned}$$

The sum is done for all integer values of z , however, the factorial of a negative number is zero so the sum only has a finite number of terms.

Because the calculation of 3j-symbols by this formula is complicated and hardworking, in this work we always tried to use the symmetry relations to give us simple results and were also used computational methods in some cases.

The 3j-symbol's symmetry relations can be consulted in ref. [25]. Some of the most important symmetry relations are

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = \begin{pmatrix} j_2 & j_3 & j_1 \\ m_2 & m_3 & m_1 \end{pmatrix} = \begin{pmatrix} j_3 & j_1 & j_2 \\ m_3 & m_1 & m_2 \end{pmatrix} \quad (A.1.6)$$

$$(-1)^{j_1+j_2+j_3} \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = \begin{pmatrix} j_2 & j_1 & j_3 \\ m_2 & m_1 & m_3 \end{pmatrix} \quad (A.1.7)$$

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = (-1)^{j_1+j_2+j_3} \begin{pmatrix} j_1 & j_2 & j_3 \\ -m_1 & -m_2 & -m_3 \end{pmatrix} \quad (A.1.8)$$

The 3-j symbol satisfy the following orthogonality relations:

$$\sum_{j_3 m_3} (2j_3 + 1) \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \begin{pmatrix} j_1 & j_2 & j_3 \\ m'_1 & m'_2 & m_3 \end{pmatrix} = \delta_{m_1 m'_1} \delta_{m_2 m'_2} \quad (A.1.9)$$

$$\sum_{m_1 m_2} (2j_3 + 1) \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \begin{pmatrix} j_1 & j_2 & j'_3 \\ m_1 & m_2 & m'_3 \end{pmatrix} = \delta_{j_3 j'_3} \delta_{m_3 m'_3} \quad (A.1.10)$$

In the calculation of the correlation function, sums over products of 3-j symbols appear.

Such sums can be performed by using the Wigner 6-j symbol:

$$\begin{aligned} \sum_{m_4 m_5 m_6} (-1)^{j_4+j_5+j_6+m_4+m_5+m_6} \begin{pmatrix} j_1 & j_5 & j_6 \\ m_1 & m_5 & -m_6 \end{pmatrix} \begin{pmatrix} j_4 & j_2 & j_6 \\ -m_4 & m_2 & m_6 \end{pmatrix} \begin{pmatrix} j_4 & j_5 & j_3 \\ m_4 & -m_5 & m_3 \end{pmatrix} \\ = \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \left\{ \begin{matrix} j_1 & j_2 & j_3 \\ j_4 & j_5 & j_6 \end{matrix} \right\} \end{aligned} \quad (\text{A.1.11})$$

Here, $\{\}$ is the Wigner 6-j symbol, which is independent of magnetic quantum numbers. More information about the Wigner 6-j symbol in ref.[25]

A.2. Wigner-Eckart Theorem

If T_{lm} is a spherical tensor operator of l th rank, from the angular momentum coupling rules we get (ref.[3])

$$\langle I', M' | T_{lm} | I, M \rangle = (-1)^{I'-M'} \begin{pmatrix} I' & l & I \\ -M' & m & M \end{pmatrix} \langle I' || T_l || I \rangle \quad (\text{A.2.1})$$

The quantity $\langle I' || T_l || I \rangle$ is called the reduced matrix element. Its value does not depend on the components M' , M , and m . This theorem is very useful for comparing amplitudes of transitions within a given manifold of states. The relative transition strenghts depend only on the 3j-symbol.

A.3. Useful Properties of the Unitary Matrix $D_{\mu M}^L$

$D_{\mu M}^L \equiv \langle \mu | D^L | M \rangle$ are the elements of a unitary matrix, called the rotation matrix. The argument ($z \rightarrow k$) of the rotation matrix D^L stands for the Euler angles (ϕ, θ, γ) describing the rotation that carries a coordinate system z to the coordinate system k . ($k \rightarrow z$) denotes the inverse rotation.

Some of the most useful properties of $D_{\mu M}^L$ are (ref.[10])

$$D_{\mu M}^L(k \rightarrow z) = D_{M\mu}^{L*}(z \rightarrow k) \quad (\text{A.3.1})$$

$$D_{\mu M}^{L*}(k \rightarrow z) = (-1)^{\mu-M} D_{-\mu-M}^L(k \rightarrow z) \quad (\text{A.3.2})$$

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$$\sum_{M'} D_{\mu M'}^L(k \rightarrow z') D_{M' M}^L(z' \rightarrow z) = D_{\mu M}^L(k \rightarrow z) \quad (\text{A.3.3})$$

The product of two matrix elements of the rotation group can be expressed in terms of a sum over matrix elements (Clebsch-Gordan series):

$$D_{\mu M}^L D_{\mu' M'}^{L'} = \sum_k \langle L \mu L' \mu' | k \tau \rangle \langle L M L' M' | k N \rangle D_{\tau N}^k \quad (\text{A.3.4})$$

The summation index k runs from $|L - L'|$ to $L + L'$ and the symbols N and τ stand for $N = M + M'$, $\tau = \mu + \mu'$.

For special choices of the quantum numbers M and μ , the matrix elements take particularly simple forms:

$$D_{\mu 0}^L(\phi \theta \gamma) = \left(\frac{4\pi}{2L+1} \right)^{\frac{1}{2}} Y_L^{\mu*}(\theta \phi) \quad (\text{A.3.5})$$

$$D_{0 M}^L(\phi \theta \gamma) = \left(\frac{4\pi}{2L+1} \right)^{\frac{1}{2}} Y_L^{-M}(\theta \phi) \quad (\text{A.3.6})$$

$$D_{00}^L(\phi \theta \gamma) = P_L(\cos \theta) \quad (\text{A.3.7})$$

where $Y_L^M(\theta \phi)$ are the spherical harmonics and $P_L(\cos \theta)$ are the Legendre Polynomials.

A.4. Liouville Operators

For each quantum-mechanical operator A we may associate the Liouville operator $A^\times$ which acts on other quantum-mechanical operators B , so that $A^\times B$ gives the commutator of A with B

$$A^\times B = AB - BA = [A, B] \quad (\text{A.4.1})$$

This appears to be no more than a notational device, and we might use it as such to simplify the mathematical problem, but there's also a physical significance as we will see further ahead.

In developing the mathematical properties of the Liouville operator, we note that it has the same relation to an ordinary linear operator as that ordinary operator has to a vector. The ordinary operator acting on a state vector gives a different vector. Similarly, a Liouville

operator acting on an ordinary operator gives a different operator. We may therefore write the matrix elements of the operator $A^\times B$ as a linear combination of the matrix elements of B :

$$\langle \mu | (A^\times B) | \nu \rangle = \sum_{\mu', \nu'} \langle \mu \nu | A^\times | \mu' \nu' \rangle \langle \mu' | B | \nu' \rangle \quad (\text{A.4.2})$$

where the coefficients $\langle \mu \nu | A^\times | \mu' \nu' \rangle$ are labeled by four indices just as the elements of B are labeled by two. These coefficients can be expressed in terms of the matrix elements of the operator A , since from eq.A.4.1 we find

$$\langle \mu | (A^\times B) | \nu \rangle = \langle \mu | (AB - BA) | \nu \rangle = \sum_{\mu'} \langle \mu | A | \mu' \rangle \langle \mu' | B | \nu \rangle - \sum_{\nu'} \langle \mu | B | \nu' \rangle \langle \nu' | A | \nu \rangle \quad (\text{A.4.3})$$

Comparing A.4.2 and A.4.3 shows that

$$\langle \mu \nu | A^\times | \mu' \nu' \rangle = \delta_{\nu \nu'} \langle \mu | A | \mu' \rangle - \delta_{\mu \mu'} \langle \nu | A | \nu' \rangle \quad (\text{A.4.4})$$

This relation thus defines the four-index “matrix elements” of the Liouville operator in terms of the matrix elements of the ordinary operator with which it is associated.

The principal property of the Liouville operator which we use is the relation

$$e^A B e^{-A} = \exp(A^\times) B \quad (\text{A.4.5})$$

To prove this we consider $e^{\lambda A} B e^{-\lambda A} = F(\lambda) B$, where $F(\lambda)$ is a four-index Liouville-type operator, and note that

$$\left(\frac{d}{d\lambda} \right) F(\lambda) B = e^{\lambda A} [A, B] e^{-\lambda A} = F(\lambda) A^\times B$$

the formal solution of which is $F(\lambda) B = \exp(\lambda A^\times) B$. This reduces to eq.A.4.5 for $\lambda = 1$. We may obtain some idea of the significance of (A.4.5) by considering the particular case $A = iHt$ where H is the system Hamiltonian. We find

$$e^{iHt} B e^{-iHt} = e^{iH^\times t} B = B(t) \quad (\text{A.4.6})$$

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The physical significance of the Liouville operator for the Hamiltonian $H^\times$ may be seen by asking for its eigenvalues and eigenoperators. These are easily found in terms of the eigenvalues and eigenfunctions of the Hamiltonian itself. If we have $H|\mu\rangle = E_\mu|\mu\rangle$, $H|\nu\rangle = E_\nu|\nu\rangle$, then the transition operators $|\mu\rangle\langle\nu|$ are seen to be the “eigenoperators” of $H^\times$:

$$\begin{aligned} H^\times |\mu\rangle\langle\nu| &= H|\mu\rangle\langle\nu| - |\mu\rangle\langle\nu|H \\ &= (E_\mu - E_\nu) |\mu\rangle\langle\nu| \end{aligned} \tag{A.4.7}$$

The eigenvalues of the Liouville operator $H^\times$ are therefore the differences $E_\mu - E_\nu$ of all of the energy levels of the Hamiltonian. These are physically observable quantities, unlike the energy levels E_μ themselves, which contain an arbitrary zero of energy. The differences $E_\mu - E_\nu$ represent the possible spectral lines emitted by the system.

A.5. Markov Process

Essentially, a Markov process is a stochastic process $X(t)$ with a short memory, that is a process which rapidly forgets its past history. It's a random process whose future probabilities are determined by its most recent values.

Markov processes play an important role in physics and the natural sciences. One reason for this fact is that many important processes, as for example the processes arising in equilibrium statistical mechanics, are Markovian provided one chooses an appropriate set of variables. Another reason is that many types of stochastic processes become Markovian by an appropriate extension of state space. Finally, Markov processes are relatively easy to describe mathematically (ref.[26]).

Considering that each X_n is a discrete random variable that takes one of N possible values, where $N = |S|$ (it may be the case that $N = \infty$), the process X is a **Markov chain** if it satisfies the Markov condition (ref.[27]):

$$\mathbb{P}(X_n = s \mid X_0 = x_0, X_1 = x_1, \dots, X_{n-1} = x_{n-1}) = \mathbb{P}(X_n = s \mid X_{n-1} = x_{n-1}) \tag{A.5.1}$$

for all $n \geq 1$ and all $s, x_1, \dots, x_{n-1} \in S$; S is some countable set.

The evolution of a chain is described by its 'transition probabilities' $\mathbb{P}(X_{n+1} = j \mid X_n = i)$; it can be quite complicated in general since these probabilities depend upon the three quantities n , i and j . We shall restrict our attention to the case when they do not depend on n but only upon i and j .

The chain X is called **homogeneous** if

$$\mathbb{P}(X_{n+1} = j \mid X_n = i) = \mathbb{P}(X_1 = j \mid X_0 = i) \quad (\text{A.5.2})$$

for all n, i, j . The transition matrix $P = (p_{ij})$ is the $|S| \times |S|$ matrix of the transition probabilities

$$p_{ij} = \mathbb{P}(X_{n+1} = j \mid X_n = i). \quad (\text{A.5.3})$$

The transition matrix P is a stochastic matrix, which is to say that:

- P has non-negative entries, or $p_{ij} \geq 0$ for all i, j
- P has row sums equal to one, or $\sum_j p_{ij} = 1$ for all i .

The n -step transition matrix $P(m, m+n) = (p_{ij}(m, m+n))$ is the matrix of n -step transition probabilities $p_{ij}(m, m+n) = \mathbb{P}(X_{m+n} = j \mid X_m = i)$.

By the assumption of homogeneity (defined above), $P(m, m+1) = P$. That $P(m, m+n)$ does not depend on m is a consequence of the following important fact

$$p_{ij}(m, m+n+r) = \sum_k p_{ik}(m, m+n) p_{kj}(m+n, m+n+r) \quad (\text{A.5.4})$$

which is called the **Chapman-Kolmogorov** equation.

The proof is given by

$$\begin{aligned} p_{ij}(m, m+n+r) &= \mathbb{P}(X_{m+n+r} = j \mid X_m = i) \\ &= \sum_k \mathbb{P}(X_{m+n+r} = j, X_{m+n} = k \mid X_m = i) \\ &= \sum_k \mathbb{P}(X_{m+n+r} = j \mid X_{m+n} = k, X_m = i) \mathbb{P}(X_{m+n} = k \mid X_m = i) \\ &= \sum_k \mathbb{P}(X_{m+n+r} = j \mid X_{m+n} = k) \mathbb{P}(X_{m+n} = k \mid X_m = i) \end{aligned} \quad (\text{A.5.5})$$

where we used the fact that $\mathbb{P}(A \cap B \mid C) = \mathbb{P}(A \mid B \cap C) \mathbb{P}(B \mid C)$.

A.6. Internal Conversion and Auger Electrons

Internal Conversion Electrons

The usual method for an excited nucleus to go from the excited state to the ground state is by emission of gamma radiation. However, in some cases, electrons from the innermost orbital shells, with greatest probability to be near the nucleus (s-electrons), interact with the nuclear charge and the energy of the excited nuclear state is transferred to the atomic electron. The conversion electron is ejected from the atom with kinetic energy equal to the transition energy minus the binding energy of the orbital electron. An orbital electron then drops to a lower energy state to fill the vacancy, and this is accompanied by the emission of characteristic x-rays.

In conversion electron decay processes there are no element transmutation (no change from one isotope into another), while upon beta decay, a proton or neutron transform each other upon emission of a positron and a neutrino or emission of an electron and an anti neutrino, respectively.

Because in the conversion electron decay the energy transfer is a two-body process (between the excited nucleus and the bound electron, and between the de-excited nucleus and the free electron), by conservation of momentum and energy, the conversion electrons have discrete values of energy, which make them useful for $e^- - \gamma$ PAC measurements since we can choose the energy value associated to the electrons that we want to use as “start” in our coincidences counting.

In a β -decay, it is usual for the daughter nucleus to still stay in an excited state, so internal conversion is one of the possible processes to make it decay to ground state and, therefore, the spectra of conversion electrons often appears mixed with the β spectra.

Auger Electrons

As it was said in the previous section, when an innermost orbital electron is lost by internal conversion (or by electron capture), an electron from a higher orbital will drop to a lower state to fill the vacancy, accompanied by the emission of electromagnetic radiation (usually x-rays) (ref.[28]). This radiation can be transferred to another electron which will be ejected. This is called “Auger effect”, thus the ejected electron is called Auger electron. The energy of the Auger electron is given by the difference between the energies of the involved orbitals, and is usually much smaller than the energy associated to the conversion electrons.

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