Chapter 3

Perturbed Angular Correlations

The term angular correlation refers to the relative angular distribution of radiations which depends on the orientation of the nuclear state at the time the radiation is emitted. Therefore, in nuclear spectroscopy, the study of gamma-gamma angular correlation is an important tool for determining level spins, gamma-ray multipolarities and transition probabilities of gamma radiations emanating from the excited levels of a nucleus. In some experimental cases the angular correlation is altered by extranuclear fields (external fields, hyperfine fields) acting on the nuclear moments of the nucleus. A measurement of magnetic moment/electric quadrupole moment of excited state then becomes possible. The existence of an anisotropy in the angular correlation of two successively emitted \( \gamma \)-rays was treated theoretically first by Hamilton [HAM40], and confirmed experimentally by Kikuchi et al. [KIK42] and Brady and Deutsch [BRA50]. Alder [ALD52] showed that perturbation resulting from the static fields could be expressed in the form of Legendre polynomial expansion of the correlation function by time integrated attenuation coefficients, which depend upon the life-time of the intermediate states and the interaction energy.

From its nuclear physics origin the perturbed angular correlation technique has been proved to be a very useful tool for Solid State Physics problems also and now it is well established technique in the Nuclear Solid State Physics. The theoretical as well as experimental aspects of the perturbed angular correlations have been discussed in detail in several review articles [FRA68, STE64, STE75, KAR67, GRO68]. The present write-up which has been adopted from refs. [FRA68, STE64] covers only certain important steps in
3.1 Unperturbed Angular Correlation

The importance of angular correlation technique lies in its straightforward theoretical basis and relatively simple experimental realization. The various aspects of angular correlation are briefly given below:

- Directional correlation, which yields the information about spins of nuclear levels and angular momenta carried by the radiations.
- Polarization correlation, which determines the parity of the nuclear level.

Normally, the radiation from a radioactive sample is isotropic because all spins are randomly oriented in space. An anisotropic radiation pattern (for \( I \geq 1 \)) can be accomplished by applying low temperatures and strong electromagnetic fields, thereby polarizing or aligning the nuclear spins, as is the case in nuclear orientation technique. In case of angular correlation technique, an effective spin alignment can be established by picking out

![Diagram](https://via.placeholder.com/150)

*Fig. 3.1: Nuclear levels and γ-transitions in cascade.*
only those nuclei whose spins happen to lie in a preferred direction. In any cascade of two nuclear radiations between two states \( I_i \to I_0 \to I_f \), let two radiations be emitted out in succession with angular momenta \( L_1 \) and \( L_2 \) in the directions \( k_1 \) and \( k_2 \) respectively. The observation direction of first radiation \( R_1 \) determines the preferred spin direction and thereby selects an ensemble of aligned nuclear spins, so that the correlated second radiation \( R_2 \) of the cascade displays an anisotropic radiation pattern. Such a situation is shown in fig. 3.1 [STE64] where \( L_1 \) and \( L_2 \) are the angular momenta of two cascading gamma rays. It is observed that this correlation is a direct consequence of the conservation of angular momentum and thus is governed by the angular momenta involved only (level spins and \( \gamma \)-ray multipolarities).

The angular correlation remains unperturbed if the nuclei in their intermediate state

![Fig.3.2: Experimental setup for detecting a \( \gamma-\gamma \) angular correlation, where 1 and 2 are the \( \gamma \)-detectors.](image_url)

have short lifetime or are subjected to vanishingly small extranuclear fields. For such a case the angular correlation function of two successively emitted radiations is given by
where $\theta$ is the angle between the direction of emission of two gamma rays (fig. 3.2), $A_{k}\text{'s}$ are the angular correlation coefficients that depend upon the spins of levels involved, the multipole orders $L, L'$ of radiations and their mixing ratios. The angular correlation coefficients $A_{k}$ describe the deviation of the coincidence probability from the isotropic case $W(\theta)=1$ and their values can be positive, negative or zero. In above equation $P_{k}(\cos\theta)$ are the Legendre polynomials. Hence $W(\theta)$ is the function of $\cos\theta$ which is sketched in fig. 3.3. A few remarks can be made about the above equation:

- Summation index $k$ is an even integer provided the measurements are restricted to directional correlations and linear polarizations of the radiations.
- $K_{max}$ is the smallest integer of the set $L, L_{1}$ and $L_{2}$: that means, an
anisotropic radiation pattern requires \( I \geq 1 \). \( L_1 \) and \( L_2 \) are the maximum values of the angular momentum carried away by the nuclear radiation in the first and second transition, respectively. The selection rules for \( k \) are

- \[ 0 \leq k \leq \min (2I, \ L_1 + L_1', \ L_2 + L_2') \quad \text{mixed transitions} \]
- \[ 0 \leq k \leq \min (2I, \ 2L_1, \ 2L_2) \quad \text{pure transitions} \]

where \( L_1' = L_1 + 1 \) and \( L_2' = L_2 + 1 \) are the angular momenta carried away by the radiations \( R_1 \) and \( R_2 \).

- The coefficients \( A_{ik} \) can be broken into two factors \( A_{1}(1)A_{2}(2) \), each factor depending upon the properties of only one transition of the cascade. These coefficients have been calculated and tabulated for nearly all cases of nuclear radiations [WAT91]. \( A_{ik} \) in the above equation are not normalized but a normalization condition can be obtained so as to give \( A_{00} = 1 \).

- Since the transition probabilities between nuclear states decrease with increasing angular momentum of the emitted ray, therefore, for majority of nuclear cascades \( K_{\text{max}} \leq 2 \).

- The angular correlation function assumes that between the time of formation and the time of decay of the intermediate state \( I \), the population of the \( m \)-states of \( I \) remains unchanged. The angular correlation in such cases is said to be unperturbed. For this condition to be valid either the lifetime of the intermediate level must be very short or the extranuclear fields must be vanishingly small.

For the pure multipole gamma radiations, eq. (3.1) can be written as

\[
W(\theta) = 1 + A_{22} P_2(\cos \theta) + A_{44} P_4(\cos \theta) + \ldots + A_{22} P_{2m}(\cos \theta) \quad \ldots(3.2)
\]

The coefficients \( A_{ik} \) are normalized by setting \( A_{00} = 1 \).

### 3.2 Perturbed Angular Correlation

The general correlation function \( W(k_1, k_2) \) can be derived by various methods. A density matrix formalism enables us to write for the unperturbed angular correlation function in the
following manner \cite{FRA68, STE64}:

\[ W(k_1, k_2, t) = \sum_{m, m'} \langle m' | H_2 | m \rangle \langle m' | H_1 | m \rangle \delta_{m, m}, X \]

where \( H_1 \) and \( H_2 \) represent the interaction between nucleus and radiation field only. If no extranuclear perturbations act on the nuclei, the final states \( \langle m_a \rangle \) and \( \langle m_a' \rangle \) of the first radiation coincide with the initial states \( |m_b\rangle \) and \( |m_b'\rangle \) of the second radiation. The presence of extranuclear fields, which can be purely magnetic, purely electric or combined magnetic and electric, gives rise to their interaction with the nucleus in its intermediate state \( I \). This interaction which changes the state \( |m_a\rangle \) to different states \( |m_b\rangle \) is described by a Hamiltonian \( K \) and is supposed to act from time \( t=0 \) at which \( R_1 \) is emitted until the time \( t=t \) at which \( R_2 \) is emitted. If we define a unitary operator \( A(t) \) that describes the evolution of the initially unperturbed \( m \)-states into the perturbed \( m \)-states during the time interval \( t \), then the perturbed angular correlation function can be written as:

\[ W(k_1, k_2, t) = \sum_{m, m'} \langle m' | H_2 | A(t) | m \rangle \langle m' | H_1 | m \rangle X \langle m' | H_2 | A(t) | m \rangle' \langle m_a' | H_1 | m \rangle' \] ...(3.4)

The states \( |m_a\rangle \) form a complete set and the state vector \( A(t)|m_a\rangle \) can be written as:

\[ A(t) | m_a \rangle = \sum_{m_b} | m_b \rangle < m_b | A(t) | m_a \rangle \] ... (3.5)

with a similar expression for \( A(t)|m_a'\rangle \).

The time evolution operator satisfies the equation:

\[ \frac{\partial}{\partial t} = -\frac{i}{\hbar} K A(t) \] ... (3.6)

For static interactions, which will be considered in our measurements, the formal solution of eq. (3.6) is:
A(t) = \exp \left[ -i \frac{K}{\hbar} t \right] \quad \text{(3.7)}

The perturbed angular correlation function can now be written in the following from:

\[
W(k_1, k_2, t) = \sum <m_f | H_2 | m_i> <m_i | A(t) | m_i> X <m_i | H_1 | m_i> \nonumber \\
<m_i | H_2 | m_i>^* X <m_i | A(t) | m_i>^* <m_i | H_1 | m_i>^* \quad \text{(3.8)}
\]

The matrix elements \(<m'|H_1|m>\) and \(<m'|H_2|m>\) are now substituted by their usual expressions [HOB56, HUB57] and summations over m-substates are performed using Racah-Algebra.

If the direction of the first radiation is assumed to be the quantization axis, the interactions cause transitions among the m-states and change the population distribution. This change is responsible for the attenuation of the correlation. For time dependent perturbations, the m-states approach a uniform population exponentially with an inverse time constant or relaxation constant which characterises the time dependent nature of the extranuclear fields and makes the angular correlation coefficients time dependent. The time dependence is then taken into account by inclusion of perturbation factor \(G_{\text{mt}}(t)\) into the eq. (3.8) i.e.

\[
W(\theta, t) = \sum_{k_1, k_2} A_{k_1}(1) A_{k_2}(2) G_{k_1, k_2}^{N_1, N_2} \left[ (2k_1 + 1)(2k_2 + 1) \right]^{-\frac{3}{2}} \times 

Y_{N_1}^{k_1}(\theta_1, \phi_1) Y_{N_2}^{k_2}(\theta_2, \phi_2) \quad \text{(3.9)}
\]

where \(k_1\) and \(k_2\) are the propagation directions of first and second gamma ray respectively and the functions \(Y_N^{k}\)'s are the spherical harmonics in directions \(k_1\) and \(k_2\) respectively. The arguments \(\theta\) and \(\phi\) of the spherical harmonics refer to the direction of observation of radiation with respect to an arbitrarily chosen axis of quantization \(z\) (fig. 3.4). For static and axially symmetric perturbing field, the perturbation factor is
\[ G^{NN}_{k_1, k_2} = \sum \left( (2k_1 + 1)(2k_2 + 1) \right)^{1/2} \begin{bmatrix} I & I & k_1 \\ m' & -N & m' \\ m' & -m & N \end{bmatrix} \begin{bmatrix} I & I & k_2 \\ m' & -N & m' \\ m' & -m & N \end{bmatrix} X \]

\[ \exp \left( -\frac{i}{\hbar} (E_m - E_{m'}) t \right) \]

with the symmetry axis of the interaction parallel to z, the angles \( \theta_1 \), \( \phi_1 \) represents the direction to the gamma-ray feeding the intermediate level in correlation experiment. The direction of the gamma-ray from the decay of the level is represented by \( \theta_2 \), \( \phi_2 \) and \( E_n \) are the energy eigenvalues to the interaction Hamiltonian.

If interacting fields in an individual micro-crystal are axially symmetric, then symmetry axis \( z' \) is specified by Euler angles \( (\alpha, \beta, 0) \) with respect to the earlier z system (fig. 3.5). The interaction Hamiltonian is diagonal in \( z' \) system with eigenvalues \( E_n \). The perturbation function for a powder source, then comes out to be:

![Diagram showing angular coordinates (\( \theta_1, \phi_1 \)) of the radiation observation directions \( k_1 \) and \( k_2 \) with respect to the perturbation coordinate system (x, y, z).](image)
Apart from the angular momentum $I$, nuclear states also possess a magnetic dipole moment $\mu$ and an electric quadrupole moment $Q$. If we place such a nucleus in an external magnetic or electric field, it interacts with these fields by virtue of its magnetic and electric moments.

Like PAC technique, unequal populations of substates can also be achieved by a nuclear reaction using a particle beam. If the particle beam is pulsed, a timing signal can be derived to indicate the moment the isomeric state is populated. In that case the direction of the particle beam replaces the direction of the feeding $\gamma$-ray and one measures the perturbed angular distribution (PAD) of $\gamma$-rays depopulating the isomeric level as a function of time elapsed since the moment at which the reaction occurred. Therefore, both PAC and PAD methods depend upon the change in population parameters of excited levels caused by hyperfine interactions and these nuclear levels can be populated either by radioactive...
decay (PAC) or by a nuclear reaction (PAD). The former method of populating a nuclear isomeric level is also known as "Off-beam" method and latter being as "In-beam" method. In "In-beam" method, a number of additional parameters like the nature of incoming and outgoing particles, their energies, directions and angular momenta in a nuclear reaction are required to determine the observed radiation. The sensitivity of a PAD experiment depends on the degree of orientation of the isomer resulting from the reaction. The high angular momenta transferred to the isomeric residue following a heavy-ion reaction also leaves it in a highly aligned state which produces sizable anisotropies of the de-exciting radiations. The main advantage of PAD method is that those isomeric states can be studied which are not populated via a cascade in the decay of radio-active nuclei with half-lives suitable for PAC experiments.

3.3 Perturbation by Magnetic Interactions

The interaction Hamiltonian required to describe the interaction of external static magnetic field acting along z-direction with the magnetic dipole moment $\mu$ of the intermediate state of the nucleus is given by

$$K_B = -\mu \cdot B$$

...(3.11)

The energy eigenvalues of the above Hamiltonian are given by

$$E_m = -B \frac{\mu}{\hbar}$$

...(3.12)

A static magnetic field $B$ exerts a torque on the nuclear magnetic dipole moment $\mu$ and tends to align the nuclear magnetic axis in the direction of $B$. The nuclear angular momentum $I^z$ will respond to this torque by a precession around $B$ as axis (shown in fig. 3.6). The Larmor frequency with which the magnetic dipole moment $\mu$ precesses around the field direction (z-axis) is defined as

$$\omega_B = \frac{(E_{m+1} - E_m)}{\hbar} = -\frac{g \mu \gamma B}{\hbar}$$

...(3.13)
Fig. 3.6: The precession of the nuclear angular momentum $I$ and the magnetic moment $\mu$ around a magnetic field.

where $g$ is Lande's $g$-factor of the intermediate state, $\mu_N$ is the nuclear magneton ($\mu_N = 5.054 \times 10^{-24}$ erg/gauss) and $B$ is the local magnetic field at the nucleus.

Quantum mechanically, the component of $I$ along field $B$ is equal to an integer $m_z$, the magnetic quantum number, and has $2I+1$ different orientations with respect to $B$ i.e. $I \geq m \geq -I$. Therefore the potential energy of a magnetic dipole in a magnetic field is

$$E(m_z) = -\frac{\mu \cdot B}{I} m_z$$

...(3.14)

and has $2I+1$ orientations which differ in their energy states. For magnetic splitting, the energy difference between two adjacent levels is constant (shown in fig. 3.7) i.e. $\Delta E = \mu B/I = \omega \frac{g I}{I} m_z$ and corresponds to Larmor frequency of the precessing magnetic dipole.

On writing the energy eigen values in terms of Larmor frequency as $E_n = \omega_n m_z$, we can express the perturbation factor (eq. 3.10) as

$$G_{k, k_z}^{NN} = \sum \left[ (2k_1 + 1)(2k_z + 1) \right]^\frac{1}{2} \left[ \begin{array}{ccc} I & I & k_1 \\ m' - m & N \end{array} \right] \left[ \begin{array}{ccc} I & I & k_2 \\ m' - m & N \end{array} \right] \chi e^{i\omega_n \mu t}$$

(3.15)
or after using the orthogonality relations of the 3-j symbols, we have

\[ G^{N
u}_{k_l}\kappa} = \exp\left[-i N \omega_B t\right] \delta_{k_l\kappa} \quad \ldots (3.16) \]

Thus, the perturbation of angular correlation function by static magnetic field acting along z-axis can be obtained by inserting this value of perturbation factor.

Further, the angular correlation function becomes simpler if the radiation detectors are held perpendicular to the direction of magnetic field B i.e. in the X-Y plane (fig. 3.8). Then the angular correlation depends only on the angle \( \theta = \phi - \phi_i \) between the directions of emission of the radiations. The simpler form of the angular correlation in this case is given by

\[ W(\theta, t, B) = \sum_{k = -N}^{N} B_k \exp\left[i N (\theta - \omega_B t)\right] \quad \ldots (3.17) \]

Here the terms with even values of \( k \) will occur because of directional correlation. A more convenient form of eq. (3.17) is:
Fig.3.8: Angular correlation of the radiation observation directions $k_1$ and $k_2$ in case of transverse magnetic field.

\[
W(\theta,t,B) = 1 + \sum_{N=2}^{\infty} b_N \cos(\theta - \omega_B t) \quad \ldots(3.18)
\]

where the coefficients $b_N = \frac{2B_N}{B_0}$, $N \neq 0$ and also

\[
b_2 = \frac{48A_{22} + 20A_{44}}{64 + 16A_{22} + 9A_{44}} \quad \ldots(3.19)
\]

\[
b_4 = \frac{35A_{44}}{64 + 16A_{22} + 9A_{44}} \quad \ldots(3.20)
\]

These angular correlation functions can also be expressed as:

\[
W(\theta,t,B) = \sum_A A_1 A_2 P_4 [\cos(\theta - \omega_B t)] \\
= 1 + \sum_{k \geq 2} b_k \cos(\theta - \omega_B t) \quad \ldots(3.21)
\]
The interpretation of these equations is that the static magnetic field causes the precession of the intermediate state of the nucleus I around the field direction with a frequency \( \omega \), the Larmor frequency. During the time \( t \) elapsed between the emission of the first radiation and that of the second radiation, the angular distributions pattern of the second radiation has rotated by \( \Delta \theta = \omega t \). The measurement of this rotation and its exploitation for the hyperfine field measurements will be discussed in the forthcoming sections.

### 3.4 Perturbation by Electric Quadrupole Interaction

Among the most common ways to observe quadrupole interaction of excited nuclei are the \( \gamma-\gamma \) time differential perturbed angular correlation / distribution methods [FRA68, STE75]. These methods have a drawback, however, that they yield only the magnitude of the product of quadrupole moment \( Q \) and EFG and not its sign. In order to determine the sign as well, measurements that are sensitive to the nuclear polarization are necessary. With pulsed heavy ion nuclear reactions, using high spin isomeric nuclear states, one can perform experiment on the polarized isomer recoil implanted into an oriented single crystal which yields information on the sign as well as the magnitude of \( Q \) [HAS84]. For this reason, in-beam experiments [HAU82, DAF85] are now performed for the observation of electric quadrupole interactions.

Basically two different physical quantities are accessible in these experiments: the strength and the symmetry of the EFG. When the EFG does not have axial symmetry, the analysis of pure quadrupole spectra is rather complex. Not only is the EFG tensor in the principal axis system to be determined, but also the orientations of the three principal axes \( x, y \) and \( z \) relative to the laboratory system (crystal lattice and detector system) must be found.

The Hamiltonian describing the interaction between quadrupole moment \( Q \) of a nucleus with the electric field gradient \( V_{zz} \) at its position due to surrounding charges is given by

\[
H_{\text{int}} = \frac{2}{3} Q V_{zz} \text{I}^2 \cos^2 \theta
\]
where \( Q^{(2)}_q \) is the tensor defining the quadrupole distribution in the nucleus and \( V^{(2)}_q \) is a symmetric trace less tensor operator of electric field gradient.

The field gradient tensor \( V_{ij} \) is defined as the second partial derivative of the classical electrostatic potential \( V \) due to the surrounding charges, evaluated at the nuclear site, that is,

\[
V_{ij} = \frac{\partial^2 V}{\partial x_i \partial x_j} \quad \ldots (3.23)
\]

The static electric interaction between quadrupole moment \( Q \) of the nucleus and electric field gradient of the lattice (fig. 3.9) perturbs the angular correlation / distribution and gives rise to an aligning torque exerted on the nucleus. The resulting precession of the
angular momentum about the z-axis of the field gradient has in general not one, but several characteristic frequencies, depending upon the relative orientation of the nuclear spin axis I with respect to the axis of the field (z-axis). This is also reflected in the non-equidistant splitting of the 2I+1 energy levels caused by the electric quadrupole coupling, a fact which is well known from pure quadrupole spectra.

The EFG is a tensor operator of rank two and nine components are required to specify it. Here we choose principal axis system in order to eliminate the next derivatives or off diagonal elements and we relate the diagonal elements $V_{xx}$, $V_{yy}$ and $V_{zz}$ to Poisson equation as

$$V_{xx} + V_{yy} + V_{zz} = 0 \quad \text{...(3.24)}$$

Therefore, it is convenient to define the axial symmetry parameter $\eta$ of the EFG as

$$\eta = \frac{V_{xx} - V_{yy}}{V_{zz}} \quad \text{...(3.25)}$$

The principal axis system is assumed to be chosen in such a way that $V_{xx} \leq V_{yy} \leq V_{zz}$ therefore $\eta$ can vary from 0 to 1, where $\eta = 0$ corresponds to axial symmetry around z with $V_{xx} = V_{yy} = -1/2 \ V_{zz}$ ; and $\eta = 1$ corresponds to $V_{xx} = 0; V_{yy} = -V_{zz}$. For axially Symmetric EFG ($V_{zz}$) whose symmetry axis is along z-axis, for a nuclear state with spin I and electric quadrupole moment Q, the level splitting is given by

$$E_{Q} = \langle \mu_{m} | H_{Q} | \mu_{m} \rangle$$

$$= \frac{3m^2 - I(I + 1)}{4I(2I - 1)} eQV_{zz}$$

$$= (3m^2 - I(I + 1))\hbar \omega_{Q} \quad \text{...(3.26)}$$

and

$$\omega_{Q} = \frac{eQV_{zz}}{4I(2I - 1)^2 \hbar} = \frac{\pi}{2I(2I - 1)} v_{Q} \quad \text{...(3.27)}$$

where $v_{Q} = (eQV_{zz})/\hbar$ is the quadrupole coupling constant.

We often describe the strength of the interaction in terms of $\omega_{Q}$ which is equivalent
to the smallest non-vanishing energy difference between two adjacent m-states and is given by the relation

\[ \omega_0 = 3\omega_q \] for integer I and
\[ \omega_0 = 6\omega_q \] for half-integer I.

For half-integral spins there are I+1/2 energy levels and all are doubly degenerate, while for integral spins there are I+1 energy levels, where I of these are doubly degenerate, and only one of them with m=0 is nondegenerate. Thus in the case of quadrupole interactions:

- The energy splitting are non-uniform and are given by \( E_m - E_{m'} = \pm n\omega_0 \) where \( n=|m^2-m'^2| \) for integral I and \( n=2|m^2 - m'^2| \) for half integral I.
- The states are two fold degenerate i.e. +m and -m give the same energy. This means that the influence of quadrupole interaction on the angular correlation can no longer be described as a unidirectional precession of the correlation pattern as in the case of a magnetic interaction and the sign of the quadrupole interaction cannot be determined by this method.

The perturbation factor for axially symmetric quadrupole interaction \( \eta = 0 \) can be described in terms of attenuation factor as

\[ G_{\alpha}(t) = \sum_n S_n \cos(n\omega_0 t) \] ...(3.28)

where \( \omega_0 \) is the basic frequency related to the interaction strength and \( S_n \) are the amplitudes defined as

\[ S_n = \sum_{m,m'} \left[ \begin{array}{ll} I & k \\ m' + m & m' - m \end{array} \right]^2 \] ...(3.29)

The amplitudes \( S_n \) depend on the nuclear spin and the radiation parameter \( k \). The numerical values of \( S_n \) for various I-values have been calculated and tabulated by Steffen and Alder [STE75]. The calculations of the attenuation factors become much more complicated in case of axially asymmetric field gradients i.e. \( \eta\neq0 \). In case of powder
sources, we diagonalize the interaction Hamiltonian using a unitary transformation \( \langle n|m \rangle \) and eigen-functions are calculated for various values of \( \omega_Q \) and \( \eta \) as has been done by Gerdau et al. [GER69]. In the case of axially asymmetric field gradients, the final expression for the perturbation factor is given by

\[
G_{\omega}(t) = S_{\omega_0} + \sum_{n=1}^{N} S_{\omega_n} \cos(\omega_n t) \tag{3.30}
\]

where \( N \) depends upon the spin of the intermediate state of cascade and \( \omega_n \) refers to the transition frequencies between hyperfine sublevels into which the intermediate state splits up because of the influence of the quadrupole interaction. For a non-zero asymmetry parameter \( \eta \), the interaction Hamiltonian must in principle be diagonalized but for small \( \eta \) it is possible to use an expansion in \( \eta \). For spin states \( I=2 \) and \( I=5/2 \) the perturbation functions are given in references [BEL76] and [BUD75] respectively and author has used the same functions in this thesis.

### 3.5 Combined Magnetic and Electric Interaction

The general theory of combined interaction for non-coaxial electric and magnetic fields has been made by Alder et al. [ALD63] and Bostrom et al. [BOS70] for a polycrystalline source. The calculations involve matrix diagonalization and summations of the products of 3j-symbols and eigenvectors. In most of the cases, the magnetic field \( B \) and the electric field gradient \( V_z \) are collinear and parallel to the \( z \)-axis or if electric interaction is much less than the magnetic interaction, allowing first order perturbation, a convenient analytic form is obtained. If \( \beta \) is the angle between \( B \) and \( V_z \) (fig. 3.10), the energy splitting \( E_m \) is given by

\[
E_m = -\frac{1}{2} \omega_B m + \frac{1}{2} \omega_Q \left[ 3m^2 - I(I+1) \right] \tag{3.31}
\]

with

\[
\omega_Q = \frac{1}{2} \frac{eQ \gamma}{4I(2I-1)} \left( 3 \cos^2 \beta - 1 \right) \tag{3.32}
\]
for a random source, the attenuation factor for combined interaction is

$$G_{ik}(t) = \sum S_{k}^{\text{ik}} \cos(q\omega_{i}t) \cos(n\omega_{i}t) \quad \text{(3.33)}$$

where coefficients $S_{k}^{\text{ik}}$ are given in review article of Steffen and Alder [STE75].

### 3.6 Method of Measurement

The experimental measurements are carried out by any of the two methods: IPAC/D and TDPAC/D, depending upon the resolving time of the coincidence system and the life time of the nuclear state. If the lifetime of the isomeric state $\tau_{N}$ is very much less than resolving time $\tau_{R}$ of the coincidence system, then effectively a perturbation factor integrated over the nuclear lifetime is obtained. If, however, $\tau_{N} \gg \tau_{R}$ then a time differential measurement can be made. Here we will restrict our discussion to TDPAC/D only which have been used for the experiments described in this thesis.
3.6.1 Measurement of Magnetic Hyperfine Field

In TDPAC technique the time differential observation of spin rotation in the presence of hyperfine fields provides a useful method for the determination of hyperfine fields [MAT63]. A pair of detectors is used to observe a cascade of two gamma rays and an external magnetic field perpendicular to or in the plane [RAG71, RAF83] of detectors is applied. Then the time differential angular correlation i.e. time spectrum of coincidence counts is determined as a function of angle $\theta$. Coincidence events are accepted only if the time delay is smaller than the resolving time and if only singles pulses have passed the windows of single channel analyzer (SCA). The decay of intermediate state is exponential which is modulated by the Larmor precession of the spins in the intermediate state and one can write

$$W(\theta, t, B) = N_0 e^{-\lambda t} \left[ \sum_{i=0}^{K} A_{ik} P_i \cos(\theta - \omega_B t) \right]$$  \hspace{1cm} (3.34)

neglecting the effect of perturbations other than magnetic ones. The coincidence counting rate is measured with the magnetic field in alternately positive and negative directions at some angle $\theta$ (135° or 225° if $A_{22}>>A_{44}$ and 157.5° if $A_{22}<<A_{44}$) and the symmetry ratio $R(t)$ is calculated as

$$R(t) = 2 \frac{N(135°, +B) - N(135°, -B)}{N(135°, +B) + N(135°, -B)}$$  \hspace{1cm} (3.35)

For $\theta = 135°$, $R(t)$ is related to $A_{22}$ and $A_{44}$ as

$$R(t) = \frac{(12A_{22} + 5A_{44}) \sin 2\omega_B t}{8 + 2A_{22} + 1/8A_{44}(9 - 35\cos 4\omega_B t)}$$  \hspace{1cm} (3.36)

for $A_{22} >> A_{44}$ this expression has the simplest form as

$$R(t) = 2b_2 \sin 2\omega_B t$$  \hspace{1cm} (3.37)

where $b_2 = 3A_{22}/(4 + A_{22})$. Since life time of the intermediate state is not involved in the
measurement of $R(t)$, $R(t)$ directly exhibits the Larmor period precisely. Instead of reversing the direction of magnetic field, same result can be obtained only by changing the angle $\theta$ from $135^\circ$ to $225^\circ$.

The Larmor precession frequency can be determined in the absence of polarizing field. It was shown by Matthias et al. [MAT65] that even if the magnetic domains are randomly oriented, the time spectrum of coincidences is still modulated by the spin rotation. The attenuation factors in that case can be expressed as

$$G_{0k}(t) = \frac{1}{2k+1} \sum_{x=0}^{k} \cos(N\omega_B t)$$  

Neglecting the terms higher than $k = 2$, we get

$$G_{22}(t) = \frac{1}{5} \left[ 1 + 2\cos(\omega_B t) + 2\cos(2\omega_B t) \right]$$  

Thus, $G_{22}(t)$ is clearly the superimposition of the two frequencies $\omega_B$ and $2\omega_B$ and experimentally it can be calculated more conveniently by making the observations at $\theta=90^\circ$ and $\theta=180^\circ$ such that

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

Two distinct features of measurements with randomly oriented domain technique are:

- The superposition of hyperfine field and a direct contribution from external field gives the effective field at the nuclear site. The externally applied field is difficult to determine because of the requirement of the knowledge of demagnetization factor for the special geometry of the ferromagnetic sample.
- The resolution of $2\omega_B$ modulations may become difficult if the hyperfine fields are very large in magnitude whereas $\omega_B$ modulations can be resolved easily because their period is twice as large compared to the case of the magnetized sample.
The above equations for $R(t)$ are valid only for an infinitely short time resolution of the electronic set-up. The finite resolving time causes an over-spreading of the amplitude of $R(t)$ but has no effect on the frequency. The theoretical expression for $R(t)$ should, therefore, be folded with experimental time response curve [BER69].

3.6.2 Measurement of Electric Field Gradient

In the case of measurement of electric field gradients, the experimental values of $G_{22}(t)$ are obtained by calculating the coincidence counting rate at $180^\circ$ and $90^\circ$ and using the following relation as in the case of randomly oriented magnetic interactions

$$G_{22}(t) = \frac{2}{A_{22}} \left[ \frac{N(180^\circ,t) - N(90^\circ,t)}{N(180^\circ,t) + 2N(90^\circ,t)} \right] \quad \text{...(3.41)}$$

Now we consider the theoretical expression (3.30) which is valid for perfect crystals where all the decaying nuclei experience exactly the same field gradient which is also applicable for the static electric interactions for the powder source. The various interaction parameters are determined by least squares fitting of the values of $G_{22}(t)$ with the above theoretical expression. The lattice imperfections which cause the distribution of EFG components is reflected as the damping of harmonic oscillations in the time spectra. Matthias et al. [MAT63a] have shown that the effect of these lattice imperfections can be taken into account by assuming a frequency distribution of Gaussian shape of relative width $\delta$ for interaction frequency. Thus, we can express the perturbation factor as

$$G_{ij}(t) = S_{ij} + \sum_n S_{in} e^{-\frac{\delta^2 \omega_i^2 t^2}{2}} \cos(\omega_n t) \quad \text{...(3.42)}$$

and for the frequency distribution of Lorentzian shape, the perturbation factor can be written as

$$G_{ij}(t) = S_{ij} + \sum_n S_{in} e^{-\delta \omega_i t} \cos(\omega_n t) \quad \text{...(3.43)}$$
which gives the interaction parameters $\omega_0$, $\eta$ and $\delta$ by least squares fitting of this expression with the experimental values.

Sometimes the behaviour of experimental $G_{22}(t)$ curve suggests the presence of two or more sites for the impurity nuclei in the host lattice and we cannot fit the data to the theoretical function given by eq. 3.17 but in such cases we can fit the data in a better way by considering the number of sites. According to this model e.g. in case of two fraction model, a fraction ($f$) of impurity nuclei occupying regular substitutional sites experience unique quadrupole interaction whereas the remaining fraction ($1-f$) is due to irregular site and subjected to a broad frequency distribution. The expression for perturbation factors in such cases is given by

$$G_{ik}(t) = f G_{ik}^{\text{reg}}(t) + (1-f) G_{ik}^{\text{irr}}(t)$$  \hspace{1cm} (3.44)

where $G_{ik}^{\text{reg}}(t)$ and $G_{ik}^{\text{irr}}(t)$ are regular and irregular site fractions of the perturbation factors.

For most of the measurements the author has adopted this two-fraction fitting procedure.

The hyperfine interaction measurements in the paramagnetic region involves the fluctuating hyperfine fields which cause the nuclear relaxation. The nuclear relaxation effects in the presence of fluctuating hyperfine fields are treated by several authors [ABR53, BLU68, SCH70, CLA71, MAR94]. In the Abragam and Pound [ABR53] approximation, $\omega_0$, $\tau_r$, and $\tau_\omega$, the attenuation of the modulated spectra can be described by a single exponential function as

$$G_{ik}(t) = e^{-\lambda_k t}, \quad k = 2, 4$$  \hspace{1cm} (3.45)

where $G_{ik}(t)$ is the perturbation factor of the angular correlation coefficient $A_{ik}$ and $\lambda_k$ is the nuclear relaxation time. The decay constants have been calculated by Abragam and Pound for the cases of pure magnetic and pure electric quadrupole perturbations. For magnetic case $\lambda_k$ is

$$\lambda_k(M) = \frac{1}{3} k(k + 1) \omega_0^2 \tau_\omega$$  \hspace{1cm} (3.46)

and for electric quadrupole interaction $\lambda_k$ is given by
\[ \lambda_4(Q) = \frac{3}{5} k(K + 1) \left[ 4I(I + 1) - k(k + 1) - 1 \right] \omega_e^2 \tau_j \] (3.47)

where \( \omega_m \), \( \omega_e \) are magnetic interaction and quadrupole interaction frequencies respectively, \( \tau_e \) is the electronic relaxation time and \( I \) is the nuclear spin. The nuclear relaxation is mainly due to the magnetic interaction and the quadrupole attenuation parameter \( \lambda_4(Q) \) is generally less than 10% of the magnetic attenuation parameter \( \lambda_4(M) \). Since the perturbing fields are known, the electronic relaxation time \( \tau_e \) can be determined by measurement of the attenuation parameter. We have used the following perturbation function \( G_4(t) \),

\[ G_4(t) = \left[ S_{s0} + \sum S_{sm} \cos(\omega_n t) \right] e^{-\lambda_4 t} \] (3.48)

for systematic investigation of electric field gradient in RE metals.