TEMPERATURE-DEPENDENT HYPERFINE INTERACTION AT THE ^{111}Cd NUCLEUS IN SILICON

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Abstract

Impurity atoms (indium and tellurium) were incorporated in silicon through an implantation process. The samples were annealed to remove radiation damage. The formation of Cd-Te complex and the temperature dependence of the quadrupole interaction frequency (QIF) were studied with help of perturbed $\gamma - \gamma$ angular correlation (PAC) technique. The observed pairing between Cd and Te was due to the interaction between acceptors and donor atoms in silicon. This explains how the presence of Te influences the lattice site of the Cd atoms. The unique quadrupole interaction frequency $\nu_Q = 444MHz$ was associated to this complex formation. The strong quadrupole interaction frequency, 444MHz at room temperature, decreases to 388MHz at $700^{\circ}C$ which is associated to the state $(Cd - Te)^{-}$ and $(Cd - Te)^{0}$ respectively.

0.1 Introduction

Nuclear techniques, such as *Mössbauer* Spectroscopy, Perturbed $\gamma - \gamma$ Angular Correlation Spectroscopy (PAC), Nuclear Magnetic Resonance and others have proven in the past to be versatile tools for studying a broad range of problems of nature [1, 2]. The interaction between the nuclear moments and the extranuclear electromagnetic fields is called hyperfine interactions [3].

The hyperfine interactions method involving the measurement of (PAC) is being used to investigate defects in intermetallic compounds, metal oxides, semiconductors, and insulators. PAC measurements heavily rely on radioactive probe atom like ^{111}In [4]. It measures the electric field gradient (EFG) which arises from the immediate lattice surrounding at the site of the probe atom. The observed effect is from the hyperfine interaction between electric field gradients and the quadrupole moment of the isotope ^{111}Cd, which is daughter of ^{111}In.

Acceptor-Donor pairs in semiconductors received much attention in the past because of their role in the deactivation of electrically active centers. PAC is well suited for the detection of such pairs because the probe atom, indium, is also an acceptor in silicon. Several group-V donors atoms implanted in silicon have been reported earlier. Recently a new defect complex has been reported after tellurium implantation in silicon. This defect complex was characterized by the observed unique quadrupole interaction frequency. This frequency is dependent on the temperature.

In this thesis discussion is given about the formation of indium-tellurium pair and the temperature dependance of the electric field gradient associated with the formed complex.

Chapter 1

Hyperfine interaction

1.1 Introduction

The observation of microscopic environment in solids is possible using nuclear methods whose working principles are based on the interaction between the nuclear moments of the probe nucleus and the surrounding electromagnetic fields. The interaction of moments (magnetic or electric) of the nucleus with electromagnetic fields is called Hyperfine Interaction. The hyperfine field affects the nucleus which makes possible the determination of internal fields in solids through the measurements of the nuclear properties

1.2 Classical Electrostatic Hyperfine Interaction

The nucleus of a solid is surrounded by electrical charges (clouds of charges) which interact with external electromagnetic fields. This interaction causes a potential $\Phi(\vec{r})$ at the site of the nucleus. Classically, the electrostatic interaction energy of a nuclear charge distribution $\rho(\vec{r})$ in an external potential $\Phi(\vec{r})$ is given by

$$E_{elec} = \int \rho(\vec{r})\Phi(\vec{r})d^3r \tag{1.1}$$

where $\int \rho(\vec{r})d^3r = Ze$, the total charge of the nucleus. Assuming the potential $\Phi(\vec{r})$ to range slowly over the region of the nuclear charge distribution, where $\rho(\vec{r})$ is non-negligible, the potential can be expanded in Taylor series around $\vec{r} = 0$ near the center of the nucleus as

$$\Phi(\vec{r}) = \phi(0) + \sum_{\alpha=1}^{3}\left(\frac{\partial\Phi}{\partial x_\alpha}\right)x_\alpha + \frac{1}{2}\sum_\alpha\sum_\beta\left(\frac{\partial^2\Phi}{\partial x_\alpha\partial x_\beta}\right)_0 x_\alpha x_\beta + \ldots. \tag{1.2}$$

Substituting equation (1.2) into equation (1.1) gives

$$E_{elec} = \underbrace{\Phi(0)Ze} + \underbrace{\sum_{\alpha=1}^{3}\left[\frac{\partial\Phi}{\partial x_\alpha}x_\alpha\right]_0\int\rho(r)x_\alpha d^3r} + \underbrace{\frac{1}{2}\sum_\alpha\sum_\beta\left[\frac{\partial^2\Phi}{\partial x_\alpha\partial x_\beta}\right]_0\int\rho(r)d^3r}$$

$$E_{elec} = E^{(0)} + E^{(1)} + E^{(2)}. \tag{1.3}$$

The first term is the Coulomb energy of point charges, which only contributes to the potential energy of the crystals lattice and thus it is of no interest in the discussion of the hyperfine interactions. The second term is the electric dipole interaction which vanishes because atomic nuclei possess no electric dipole moment (parity conservation). The third term $E^{(2)}$ contains the isomeric shift and quadrupole interaction. By introducing $r^2 = \sum_\alpha x_\alpha^2$, and adding and subtracting the same term $\frac{1}{6}\sum_\alpha\sum_\beta\Phi_{\alpha_\beta}\int\rho(r)r^2d^3r$, one can write $E^{(2)}$ as

$$E^{(2)} = \frac{1}{6}\sum_\alpha\sum_\beta\Phi_{\alpha_\beta}\int\rho(r)r^2d^3r + \frac{1}{6}\sum_\alpha\sum_\beta\Phi_{\alpha_\beta}\int\rho(r)d^3r\Big(3x_\alpha x_\beta - r^2\delta_{\alpha_\beta}\Big), \tag{1.4}$$

where the quantity Φ_{α_β} is

$$\Phi_{\alpha_\beta} = \left(\frac{\partial^2\Phi}{\partial x_\alpha\partial x_\beta}\right). \tag{1.5}$$

Of course, this result can also be obtained through the expansion of the term $\frac{1}{|r-r'|}$ using a binomial series expansion of the potential $\Phi(\vec{r})$, where

$$\Phi(\vec{r}) = \frac{1}{4\pi\epsilon_0}\int_{vol}\frac{\rho(\vec{r})dr}{|\vec{r}-\vec{r}'|}. \tag{1.6}$$

Therefore,

$$\frac{1}{|r-r'|} = \frac{1}{\left(r^2 + r'^2 - 2\vec{r}\cdot\vec{r}'\right)^{\frac{1}{2}}}$$

$$\frac{1}{|r-r'|} = \frac{1}{r}\Big[1 + \frac{\vec{r'}\cdot\vec{r}}{r^2} - \frac{r'^2}{2r^2} + \frac{3}{2}\Big(\frac{\vec{r'}\cdot\vec{r}}{r^2}\Big)^2 - \frac{3}{8}\Big(\frac{r'^2}{r^2}\Big)^2 + \frac{6(\vec{r'}\cdot\vec{r})^2}{2r^4} + \ldots\Big]$$

$$\frac{1}{|r-r'|} \simeq \frac{1}{r}\Big[1 + \frac{\vec{r}\cdot\vec{r'}}{r^2} + \frac{1}{2}(3\Big(\frac{\vec{r}\cdot\vec{r'}}{r^2}\Big)^2 - \frac{r'^2}{r^2})\Big]$$

or,

$$\frac{1}{|r-r'|} = \frac{1}{r} + \frac{\vec{r}\cdot\vec{r'}}{r^3} + \frac{1}{2}\Big[\frac{3\Big(\vec{r}\cdot\vec{r'}\Big)^2}{r^5} - \frac{r'^2}{r^3}\Big]. \tag{1.7}$$

With $\frac{\vec{r}\cdot\vec{r'}}{r} = r'\cos\theta$, substituting equation (1.7) into equation (1.6), one obtains,

$$\Phi(\vec{r}) = \frac{1}{4\pi\epsilon_0}\frac{q}{r} + \frac{1}{4\pi\epsilon_0}\int\frac{\rho(\vec{r})r'\cos\theta}{r^2}d^3r + \frac{1}{8\pi\epsilon_0}\int\frac{\rho(\vec{r})(3\cos^2\theta - 1)r'^2}{r^3}d^3r$$

This expression can be rewritten using tensor representation of the term

$$\frac{\vec{r}\cdot\vec{r'}}{r} = r'\cos\theta = \sum_\alpha \frac{x_\alpha X_\alpha}{r}$$

(with x_α=1,2,3 corresponding to X,Y,Z respectively). In the above expression, q is the total charge $\int\rho(\vec{r})d^3r$. Using the cartesian expansion we obtain,

$$\Phi(\vec{r}) = \frac{1}{4\pi\epsilon_0}\frac{q}{r} + \sum_\alpha\frac{x_\alpha}{4\pi\epsilon_0}\int\frac{\rho(\vec{r})X_\alpha}{r^3}d^2r + \frac{1}{8\pi\epsilon_0}\sum_{\alpha\beta}X_\alpha X_\beta\int\frac{\rho(\vec{r})(3x_\alpha x_\beta - r'^2\delta_{\alpha\beta})}{r^5}d^3r$$

$$\Phi(\vec{r}) = \Phi^0 + \sum_\alpha\frac{p_\alpha X_\alpha}{4\pi\epsilon_0 r^3} + \frac{1}{8\pi\epsilon_0}\sum_{\alpha\beta}\frac{Q_{\alpha\beta}X_\alpha X_\beta}{r^5} \tag{1.8}$$

with

$$\Phi^0 = \frac{1}{4\pi\epsilon_0}\frac{q}{r},$$

$$p_\alpha = \int\rho(\vec{r})x_\alpha d^3r$$

and

$$Q_{\alpha\beta} = \int\rho(\vec{r})(3x_\alpha x_\beta - r'^2\delta_{\alpha\beta})d^3r$$

p_α and $Q_{\alpha\beta}$ are the dipole and quadrupole tensors, respectively. The quadrupole tensor $Q_{\alpha\beta}$ can be expressed as its nine components in matrix from [5]. Hence,

$$Q_{\alpha\beta} \Rightarrow \begin{pmatrix} 3x^2 - r'2 & 3xy & 3xz \\ 3xy & 3y^2 - r'^2 & 3yz \\ 3xz & 3yz & 3z^2 - r'^2 \end{pmatrix}$$

Moreover, transforming the quadrupole tensor $Q_{\alpha\beta}$ to diagonal form, one can find a new coordinate system in which the non-diagonal terms vanish and one gets a new quadrupole tensor

$$Q'_{\alpha\beta} \Rightarrow \begin{pmatrix} 3x^2 - r'2 & & \\ & 3y^2 - r'^2 & \\ & & 3z^2 - r'^2 \end{pmatrix}$$

The electrostatic potential $\Phi(\vec{r})$ that causes the electrostatic interaction energy in equation (1.1) is the same as the potential $\Phi(\vec{r})$ given in equation (1.5). In addition, the expansion performed for $\Phi(\vec{r})$ in equation (1.2) is for a slowly varying potential over the region of the nuclear charge distribution and the expansion performed in equation (1.6) is also for $r' < r$, i.e. for a slowly varying potential. On the other hand, if one substitutes equation (1.6) into equation (1.2) and performs the necessary differentiation, equation (1.8) can be obtained. Therefore, equation (1.8) and equation (1.2) are expressions of the same potential based on different approaches. The electrostatic potential $\Phi(r)$ satisfies Poisson's equation $\Delta\Phi = -4\,\pi\rho_e(\vec{r})$, which holds true especially near $\vec{r} = 0$. Hence, the gradient of Φ at $\vec{r} = 0$

$$\Big(\Delta\Phi\Big)_0 = \sum_\alpha \Phi_{\alpha\alpha} = 4\pi e|\Psi(0)|^2 \tag{1.9}$$

where $-e|\Psi(0)|^2$ is the charge density of the atomic electrons at the nucleus. Applying a suitable choice of rotation of the coordinate axes to equation (1.4), the electrostatic hyperfine interaction may be written as

$$E^{(2)} = \underbrace{\frac{1}{6}\sum_\alpha \Phi_{\alpha\alpha} \int \rho(r) r^2 d^3r}_{} + \underbrace{\frac{1}{6}\sum_\alpha \sum_\beta \Phi_{\alpha\beta} \int \rho(r) d^3r \Big(3x_\alpha x_\beta - r^2\delta_{\alpha\beta}\Big)}_{}$$

$$E^{(2)} = E_s + E_Q. \tag{1.10}$$

The first term, the isomeric shift E_s, reflects the interaction between the extended charge of the nucleus and the charge density of the electrons at the nucleus.

$$E_s = \frac{1}{6}\sum_{\alpha}\Phi_{\alpha\alpha}\int\rho(r)r^2d^3r = \frac{4\pi e}{6}|\psi(0)|^2\int\rho(r)r^2d^3r = \frac{4\pi Ze}{3}|\psi(0)|^2\langle r^2\rangle$$

The isomeric shift does not explicitly depend on the angular momentum of the nucleus. Instead, it depends on the mean square of the radius of the nucleus. Thus, this leads only to a shift of the nuclear energy levels but not to a splitting of the degenerate m-substates. E_Q describes the interaction between the electric field gradient and the non-spherical part of the nuclear charge distribution, i.e. the quadrupole moment teusor.

1.3 Quantum Mechanical Treatment of the Electrostatic Hyperfine Interaction

The interaction between the quadrupole moment of the nucleus and the local electric field gradient results in a splitting of the energy levels of the different states of the nucleus. The interaction energy is defined as the matrix element of the electrostatic Hamiltonian (H_{el})

$$E_m = \langle Im|H_{el}|Im'\rangle \tag{1.11}$$

where I is local angular momentum with its projection m and m' along the Z-axis. The Hamiltonian that describes the interaction of a fixed electrostatic field gradient with the electric quadrupole moment of a nuclear state is given by [6, 7, 8]

$$H_Q = \frac{4\pi}{5}T^{(2)}V^{(2)} = \frac{4\pi}{5}\sum(-1)^q T_q^{(2)}V_{-q}^{(2)} \tag{1.12}$$

This relation results from the Hamiltonian

$$H_{el} = \sum_{c,p}\frac{e_p e_c}{|r_p - r_c|} \tag{1.13}$$

for $r_c > r_p$. The term $\frac{1}{|r_p - r_c|}$ of the above expression can be written using the expansion of Legender Polynomials [6],

$$\frac{1}{|r_p - r_c|} = \sum_{k=0}^{\infty} \frac{r_p^k}{r_c^{k+1}} P_k(\cos(r_p, r_c)) \tag{1.14}$$

where P_k is Legender polynomial. Using the addition theorem of spherical harmonics

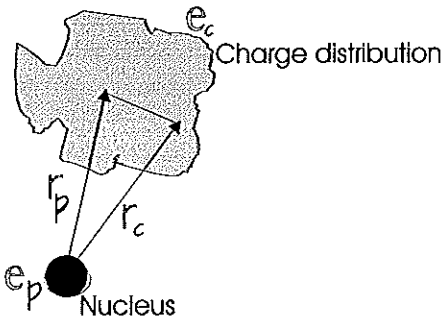

Figure 1.1: Representation of the position vectors r_c and r_p.

which states that; if two different directions (θ_1, ϕ_1) and (θ_2, ϕ_2) in spherical coordinate system are separated by an angle θ [9, 8] then

$$P_k(cos\theta) = \frac{4\pi}{2k+1} \sum_{q=-k}^{k} Y_k^q(\theta_1, \phi_1) Y_k^{-q}(\theta_2, \phi_2)$$

$$P_k(cos\theta) = \frac{4\pi}{2k+1} \sum_{q=k}^{k} Y_k^q(\theta_1, \phi_1) Y_k^{q*}(\theta_2, \phi_2) \tag{1.15}$$

where the asterisk may go on either spherical harmonic since

$$Y_k^{-q}(\theta, \phi) = (-)^q Y_k^{q*}(\theta, \phi) \tag{1.16}$$

and

$$Y_k^q(\theta, \phi) = Y_{k,q}(\theta, \phi) = (-1)^q \sqrt{\frac{(2k+1)(k-q)!}{4\pi(k+q)!}} P_k^q(\cos(\theta, \phi)).$$

Therefore, substituting equation (1.13) and equation (1.14) into equation (1.15) and then substituting the result into equation (1.12) one obtains

$$H_{el} = 4\pi \sum_{k=0}^{\infty} \frac{1}{2k+1} \sum_{q} (-1)^q \underbrace{\sum_{p} e_p r_p^k Y_{k,q}(\theta_p, \phi_p)} \underbrace{\sum_{c} \frac{e_c}{r_c^{k+1}} Y_{k,-q}(\theta_c, \phi_c)} \quad . \tag{1.17}$$

To write H_{el} more compactly, let

$$T_q^k = \sum_p e_p r_p^k Y_{k,q}(\theta_p, \phi_p)$$

and

$$V_q^k = \sum_c \frac{e_c}{r_c^{k+1}} Y_{k,-q}(\theta_c, \phi_c),$$

where e_p are the (point) charges in the nucleus at the points (r_p, θ_p, ϕ_p). T^k is the k-rank tensor operator of the nuclear quadrupole moment, e_c are point charges (ions in the crystal lattice) at positions (r_c, θ_c, ϕ_c) with respect to nuclear center, V_q^k is the tensor operator of the classical external field gradient. Therefore, for $k = 2$ equation (1.12) can be written as

$$H_Q = 4\pi \frac{1}{5} \sum_q (-1)^q T_q^{(2)} V_{-q}^{(2)}$$

$$H_Q = H_{el} = \frac{4\pi}{5} V^{(2)} T^{(2)}. \tag{1.18}$$

Now, calculating field tensors from Coulomb's law

$$V_c = \sum \frac{e_c}{r_c} = \sum \frac{e_c}{\left[x^2 + y^2 + z^2\right]^{1/2}}$$

$$\frac{\partial V_c}{\partial z} = \sum e_c \Big[- z(x^2 + y^2 + z^2)^{-3/2} \Big]$$

$$\frac{\partial^2 V}{\partial z^2} = \sum e_c \left[\frac{3z^2 - r^2}{r^5} \right]$$

and with

$$z = r cos\theta$$

$$\frac{\partial^2 V}{\partial z^2} = \frac{1}{r^3} \sum e_c (3\cos^2\theta - 1). \tag{1.19}$$

But from the general definition of the field tensor,

$$V_q^{(k)} = \sum_c \frac{e_c}{r_c^{k+1}} Y_{k,-q}(\theta_c, \phi_c)$$

$$V_0^{(2)} = \sqrt{\frac{5}{16\pi}} \frac{1}{r^3} \sum e_c (3\cos^2\theta - 1)$$

$V_0^{(2)} = \sqrt{\frac{5}{16\pi}} V_{zz}$ where, $V_{zz} = \frac{\partial^2 V}{\partial z^2}$

$$V_{\pm 2}^{(2)} = \frac{1}{r^3} \sum e_c \sqrt{\frac{15}{32\pi}} \sin^2\theta \exp(\pm i 2\phi)$$

Here we have used $Y_{2,\pm 2} = \sqrt{\frac{15}{32\pi}} \sin^2\theta \exp(\pm i 2\phi)$. Taking the real part only gives

$$V_{\pm 2}^{(2)} = \frac{1}{r^3} \sum_c e_c \sqrt{\frac{15}{32\pi}} \sin^2\theta \Big(\cos^2\phi - \sin^2\phi \Big).$$

But,

$$\frac{\partial^2 V_c}{\partial x^2} = \sum_c \frac{e_c}{r^3} \Big(3\sin^2\theta \cos^2\phi - 1 \Big) \tag{1.20}$$

and,

$$\frac{\partial^2 V_c}{\partial y^2} = \sum_c \frac{e_c}{r^3} \Big(3\sin^2\theta \sin^2\phi - 1 \Big). \tag{1.21}$$

Let $V_{xx} = \frac{\partial^2 V_c}{\partial x^2}$ and $V_{yy} = \frac{\partial^2 V_c}{\partial y^2}$

$$V_{xx} - V_{yy} = 3 \sum_c \frac{e_c}{r^3} \sin^2\theta \Big(\cos^2\phi - \sin^2\phi \Big)$$

and,

$$\frac{V_{xx} - V_{yy}}{3} = \sum_c \frac{e_c}{r^3} \sin^2\theta \Big(\cos^2\phi - \sin^2\phi \Big).$$

Therefore, $V_{\pm 2}^{(2)} = \sqrt{\frac{15}{32\pi}} \frac{V_{xx} - V_{yy}}{3} = \frac{1}{4}\sqrt{\frac{5}{6\pi}} \eta V_{zz}$ where

$$\eta = \frac{V_{xx} - V_{yy}}{V_{zz}} \tag{1.22}$$

$$V_{\pm 1}^{(2)} = \sum_c \frac{e_c}{r^3} Y_{2\pm 1}(\theta, \phi)$$

or

$$V_{\pm 2}^{(2)} = \sum_c \frac{e_c}{r^3} \sqrt{\frac{15}{8\pi}} \exp(\pm i\phi) \sin\theta \cos\theta \tag{1.23}$$

where, $Y_{2,\pm 1} = \sqrt{\frac{15}{8\pi}} \exp(\pm i\phi) \sin\theta \cos\theta$. If we consider only the real part,then

$$V_{\pm 1}^{(2)} = \sum_c \frac{e_c}{r^3} \sqrt{\frac{15}{8\pi}} \sin\theta \cos\theta \cos\phi. \tag{1.24}$$

Let us now calculate the mixed derivatives

$$\frac{\partial^2 V}{\partial x \partial y} = \frac{\partial \Big(\sum -e_c \, x \frac{1}{r^3} \Big)}{\partial y} = \frac{\sum e_c x \, y}{r^5} = \frac{\sum e_c (\sin^2\theta) \cos\phi \sin\phi}{r^3} \tag{1.25}$$

$$\frac{\partial^2 V}{\partial x \partial z} = \frac{\partial\left(\sum -e_c \frac{x}{r^3}\right)}{\partial z} = \frac{\sum e_c x\ z}{r^5} = \frac{\sum e_c (\sin\theta)\ \cos\phi\cos\theta}{r^3}$$
$$\frac{\partial^2 V}{\partial y \partial z} = \frac{\partial\left(\sum -e_c \frac{x}{r^3}\right)}{\partial z} = \frac{\sum e_c x\ z}{r^5} = \frac{\sum e_c (\sin\theta)\ \sin\phi\cos\theta}{r^3} \tag{1.26}$$

By symmetry

$$\begin{aligned}\frac{\partial^2 V}{\partial x \partial y} &= \frac{\partial^2 V}{\partial y \partial x} \\ \frac{\partial^2 V}{\partial x \partial z} &= \frac{\partial^2 V}{\partial z \partial x} \\ \frac{\partial^2 V}{\partial y \partial z} &= \frac{\partial^2 V}{\partial z \partial y}\end{aligned} \tag{1.27}$$

$$\frac{\partial^2 V}{\partial x_\alpha \partial x_\beta} = \sum \frac{e_c}{r^3} \begin{pmatrix} 3\sin^2\theta\cos^2\phi - 1 & \sin^2\theta\sin\phi\cos\phi & \sin\theta\cos\theta\cos\phi \\ \sin^2\theta\cos\phi\sin\phi & 3\sin^2\theta\sin^2\phi - 1 & \sin\theta\cos\theta\sin\phi \\ \sin\theta\cos\theta\cos\phi & \sin\theta\cos\theta\sin\phi & 3\cos^2\theta - 1 \end{pmatrix}$$

However, choosing a principal axis system such that the mixed derivatives of the potential V disappears, the elements of the tensor become

$$V_0^{(2)} = \frac{1}{4}\sqrt{\frac{5}{\pi}} V_{zz}$$

$$V_{\pm 1}^{(2)} = 0$$

$$V_{\pm 2}^{(2)} = \frac{1}{4}\sqrt{\frac{5}{6\,\pi}}(V_{xx} - V_{yy}) = \frac{1}{4}\sqrt{\frac{5}{6\,\pi}}\ \eta\ V_{zz}$$

Where η measures the symmetry of the field with respect to quantization axis. For axially symmetric field ($\eta = 0$), the fields tensor operator reduces to the form

$$V_0^{(2)} = \frac{1}{4}\sqrt{\frac{5}{\pi}} V_{zz} \tag{1.28}$$

Using this result the Hamiltonian H_Q can be written as

$$H_Q = \sqrt{\frac{\pi}{5}}\ V_{zz}\ T_0^{(2)} \tag{1.29}$$

$$E_m = \langle Im | H_Q | Im \rangle$$

or,

$$E_m = \sqrt{\frac{\pi}{5}}\, V_{zz}(Im|T_0^{(2)}|Im\rangle. \tag{1.30}$$

The quadrupole moment of a nucleus is usually defined as the $I = m$ component of the tensor [11, 7]. That is,

$$eQ = (II|\sum_p \rho_p(3z_p^2 - 1)|II\rangle = (II|\sum_p \rho_p r_p^2(3\cos^2\theta - 1)|II\rangle. \tag{1.31}$$

But,

$$T_0^{(2)} = \sum \rho_p r_p^2 Y_{2,0}(\theta, \phi)$$

$$T_0^{(2)} = \sum \rho_p r_p^2 \frac{1}{4}\sqrt{\frac{5}{\pi}}(3\cos^2\theta - 1),$$

where $Y_{2,0}(\theta, \phi) = \sqrt{\frac{5}{16\pi}}(3\cos^2\theta - 1)$. Hence,

$$T_0^{(2)} = \sqrt{\frac{5}{16\pi}} \sum_p e_p r_p^2(3\cos^2\theta - 1).$$

But $Z_p = r_p\cos\theta$, so

$$T_0^{(2)} = \sqrt{\frac{5}{16\pi}} \sum_p e_p(3r_p^2\cos^2\theta - r_p^2)$$

$$= \sqrt{\frac{5}{16\pi}} \sum_p e_p(3Z_p^2 - r_p^2)$$

or,

$$\frac{T_0^{(2)}}{\sqrt{\frac{5}{16\pi}}} = \sum_p e_p(3Z_p^2 - r_p^2) \tag{1.32}$$

Using equation (1.32) in equation (1.31) gives

$$eQ = \langle II|\sum_p e_p(3Z_p^2 - r_p^2)|II\rangle$$

$$= \langle II|\frac{T_0^{(2)}}{\sqrt{\frac{5}{16\pi}}}|II\rangle$$

$$eQ = 4\sqrt{\frac{\pi}{5}}\langle II|T_0^{(2)}|II\rangle \tag{1.33}$$

This equation is solved using Wigner-Eckert Theorem and the 3-j symbols [12, 13]

The Wigner-Eckert Theorem predicts that the matrix element $\langle jm|T_q^k|j'm'\rangle$ may

be separated into two parts, one of them containing information of magnetic quantum number and other containing characteristic of the wave vector and operator. It separates the vector of the form $\langle jm|T^k_q|j'm'\rangle$ as

$$\langle jm|T^k_q|j'm'\rangle = (-1)^{j-m}\begin{pmatrix} j & k & j' \\ -m & q & m' \end{pmatrix}\langle j||T^k||j'\rangle.$$

Using these definitions for j=I, k=2 and q=0, eQ can be written as

$$eQ = 4\sqrt{\frac{\pi}{5}}\langle II|T^{(2)}_0|II\rangle$$

Using the Wigner-Eckert theorem,

$$\langle II|T^{(2)}_0|II\rangle = (-1)^{I-(I)}\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}\langle I||T^2||I\rangle.$$

Hence,

$$eQ = (\ \ 1)^{I-(I)}4\sqrt{\frac{\pi}{5}}\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}\langle I||T^2||I\rangle$$

or,

$$= 4\sqrt{\frac{\pi}{5}}\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}\langle I||T^2||I\rangle. \tag{1.34}$$

Therefore, the expression for interaction energy becomes

$$E_m = \sqrt{\frac{\pi}{5}}\langle Im|T^2_0|Im'\rangle V_{zz}.$$

By use of the Wigner-Eckert theorem this expression can be written as

$$E_m = \sqrt{\frac{\pi}{5}}\begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix}\langle I||T^2||I\rangle V_{zz}. \tag{1.35}$$

Solving for $\langle I||T^2||I\rangle$ from equation (1.34) and substituting into equation (1.35) we obtain

$$E_m = \frac{1}{4}\frac{\sqrt{\frac{\pi}{5}}\begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix}eQV_{zz}}{\sqrt{\frac{\pi}{5}}\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}}$$

or,

$$= \frac{1}{4} \frac{\begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix} eQV_{zz}}{\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}}. \tag{1.36}$$

Inserting equation (A.6) into equation (1.36) the energy eigenvalues of the quadrupole interaction are

$$E_m = \frac{3m^2 - I(I+1)}{4I(2I-1)} eQV_{zz}. \tag{1.37}$$

Similarly for m'-substates

$$E_{m'} = \frac{3m'^2 - I(I+1)}{4I(2I-1)} eQV_{zz}. \tag{1.38}$$

The difference of the two substates gives

$$E_m - E_{m'} = \frac{3eQV_{zz}}{4(2I-1)} |m^2 - m'^2|, \tag{1.39}$$

where differences $|m^2 - m'^2|$ are integral numbers. Due to the m^2 dependence of the quadrupole interaction energy there is a two fold degeneracy of each energy levels. The term Q is the quadrupole moment of the nucleus that is subjected to the external field. The splitting of the levels is not equidistant in the electrostatic interaction. Hence, the fundamental frequency of the interaction ω_Q is defined as [11]

$$\omega_Q = \frac{eQV_{zz}}{4I(2I-1)\hbar}. \tag{1.40}$$

Using this result in equation (1.39)

$$E_m - E_{m'} = 3\hbar\omega_Q |m^2 - m'^2| \tag{1.41}$$

the frequency ω_0 corresponding to the smallest non-vanishing energy difference is

$$\omega_0 = \begin{cases} 6\omega_Q \; for \; half \; integer \; nuclearspin \; I \\ 3\omega_Q \; for \; integer \; nuclear \; spin \; I \end{cases}. \tag{1.42}$$

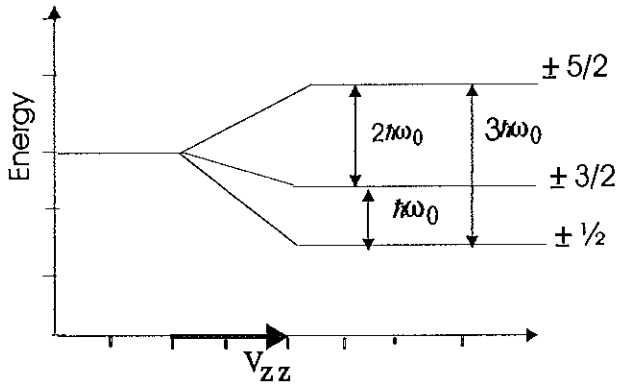

Figure 1.2: Schematic diagram of the splitting of the intermediate energy level for $I = 5/2$ and $\eta = 0$ according to equation (1.39)..

Fig.(1.2) shows the splitting of the energy level of the intermediate state of a nucleus with spin $I = \frac{5}{2}$, as the result of static electric quadrupole interaction. For $\eta = 0$ the ratio of the transition frequencies are $\omega_1 : \omega_2 : \omega_3 = 1 : 2 : 3$. The smallest transition frequency in this case would be $\omega_1 = \omega_0$ and the remaining components can be obtained by the relation $\omega_n = n\omega_0 (n = 1, 2, 3)$. In the case of non-axially symmetric field ($\eta \neq 0$), the Hamiltonian describing pure electrostatic quadrupole interaction is written in a form [14, 15, 16]

$$H_Q = \left\{ 3{I_z}^2 - I(I+1) + \frac{\eta}{2}(I_+^2 + I_-^2) \right\} \hbar\omega_Q, \tag{1.43}$$

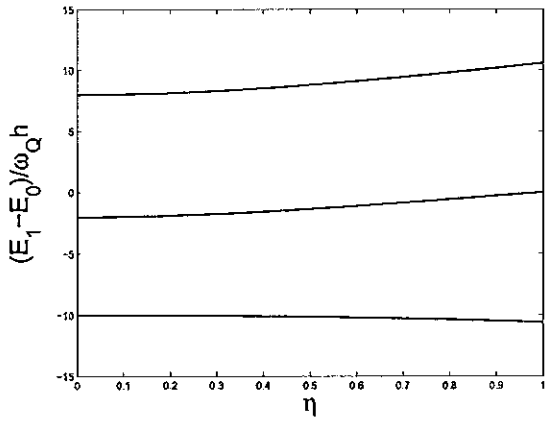

Figure 1.3: The splitting of the intermediate energy level as function of the asymmetry parameter η according to equation (1.44)

where I_z, I_+ and I_- are designated as the angular momentum operators. The quantities ω_Q and η are defined in equation (1.40) and equation (1.22), respectively.

In order to calculate the energy eigenvalues, the interaction matrix $\langle Im|H_Q|Im'\rangle$ has to be diagonalized. This can be achieved by the unitary transformation of the Hamiltonian (H_Q). For the spin $I = \frac{5}{2}$, the resulting energy eigenvalues are [15],

$$\begin{aligned} E_{\pm\frac{5}{2}} &= E_0 + 2\alpha\hbar\omega_Q(\frac{1}{3}\arccos\beta) \\ E_{\pm\frac{3}{2}} &= E_0 - 2\alpha\hbar\omega_Q(\frac{1}{3}(\pi + \arccos\beta)) \\ E_{\pm\frac{1}{2}} &= E_0 - 2\alpha\hbar\omega_Q(\frac{1}{3}(\pi - \arccos\beta)) \end{aligned} \tag{1.44}$$

where α and β are

$$\alpha = \sqrt{28(1 + \frac{\eta^2}{3})} \tag{1.45}$$

and

$$\beta = \frac{80(1 - \eta^2)}{\alpha^3}. \tag{1.46}$$

The corresponding expressions for the spin precision frequencies are,

$$\omega_1 = 2\sqrt{3}\alpha\omega_Q \sin(\frac{1}{3}\arccos\beta)$$

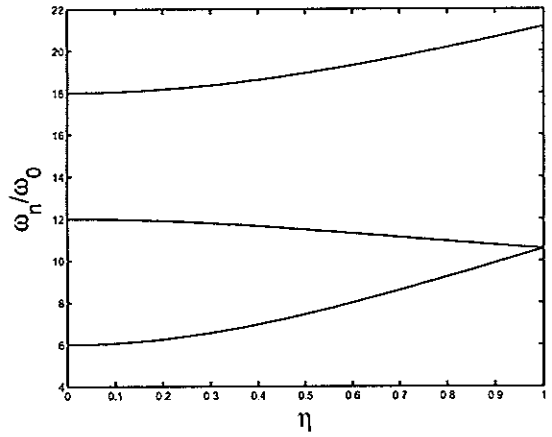

Figure 1.4: The splitting of the intermediate energy level as a function of the asymmetry parameter η according to equation (1.47)

$$\omega_2 = 2\sqrt{3}\alpha\omega_Q \sin(\frac{1}{3}(\pi - \arccos\beta))$$

$$\omega_3 = \omega_1 + \omega_2 = 2\sqrt{3}\alpha\omega_Q \sin(\frac{1}{3}(\pi + \arccos\beta)) \tag{1.47}$$

The value of the asymmetric parameter defined by equation (1.22), in the principal axes system with the convention $|V_{xx}| \leq |V_{yy}| \leq |V_{zz}|$, is restricted to $0 \leq \eta \leq 1$. Therefore, the electric field gradient tensor can be described for most applications by two parameters, i.e. V_{zz} and η. In most cases the quadrupole coupling constant ν_Q is defined instead of V_{zz}. This convention will be followed here:

$$\nu_Q = g(\eta)\frac{10}{3\pi}\omega_1. \tag{1.48}$$

When $\eta = 0$ the value of $g = 1$

Chapter 2

Theory of angular correlation

2.1 Introduction

Perturbed Angular Correlation (PAC) is a nuclear technique, which uses radioactive nuclei as a probe to extract information about their microscopic environments [17]. The method is based on the spatial orientation of nuclear spins. The radiation from a radioactive source has no unique direction in space under normal circumstances. But, if attempts made to align the spin nuclei, an anisotropic radiation distribution is observed. An effective spin alignment can be established by picking out only those nuclei whose spins align in a preferred direction.

In successive emission of two γ-rays the observation of the first emission in one direction determines the preferred spin alignment. From a give nuclei, one selects an ensemble of aligned nuclear spins such that the correlated second γ-ray of the cascade reflects anisotropic radiation pattern.

To observe the spin precession in angular correlation measurements, the nuclear moments (magnetic or electric) must be large enough in order to enhance A_{kk} (the anisotropic coefficients). Moreover, the life time τ of the intermediate state of the decaying nuclei should also be large enough. The reason for having large τ is in order to have enough

time for the interaction of the nuclear with electromagnetic fields moments at the site of the nucleus to take place. This results in perturbation of the correlation. In some cases the nuclear moment interacts with static extranuclear fields (static interactions) or it may interact with fields that periodically vary (periodic interactions), or it may interact with time dependent fields (time-dependent interactions).

2.1.1 Directional angular correlation

From the above introduction we see that an anisotropic radiation pattern can only be observed when there is a nuclear spin I alignment. This spin alignment is governed by conservation of angular momentum, and the angular distribution of electromagnetic radiation with respect to its angular momentum vector L. Fig.2.1 describes this situation.

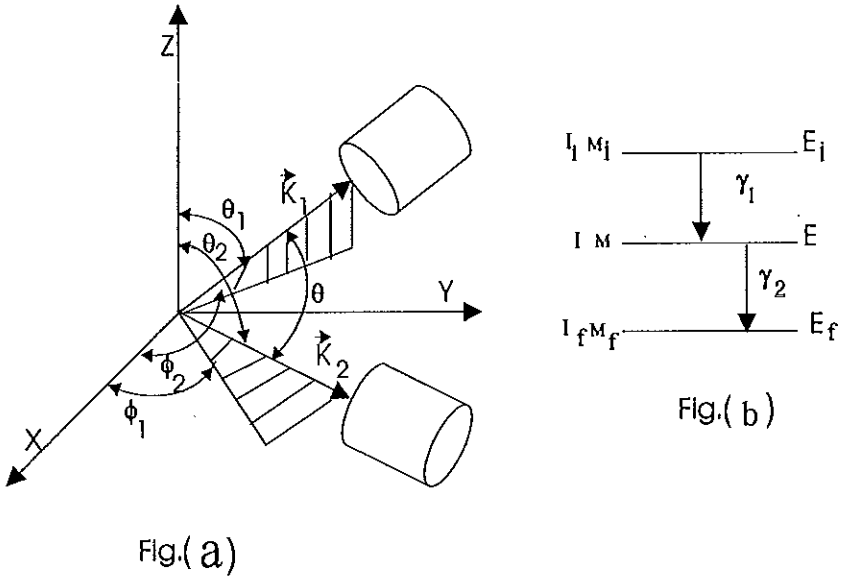

Figure 2.1: Fig.(a) shows emission direction of γ-rays in laboratory systems and Fig.(b) represents a decay scheme of a $\gamma - \gamma$ transition from an ensemble of nuclei.

In the figure a radioactive nuclei decays to the intermediate state by emission of γ_1 which carries an energy of E_1 and angular momentum L_1 in the direction of $\vec{k_1}$. The intermediate

state is now populated with nuclei of certain directions (say, γ_2 in the direction of $\vec{k_2}$). Then, the succeeding radiation of γ_2 with energy E_2 and angular momentum L_2 is anisotropic and reflects unequal sub-levels of the population of the intermediate state spin L. Therefore, angular correlation is the spatial distribution of the second quantum state.

The probability of measuring γ_2 in a particular direction relative to the direction of γ_1 depends on the angle between the directions of both radiations. When the angle dependence remain constant as a function of time, we have unperturbed angular correlation. Such directional angular correlation can be described by correlation function $W(\vec{k_1}, \vec{k_2})$. Here $W(\vec{k_1}, \vec{k_2})$ is the probability to detect γ_2 at an angle θ with respect to γ_1 when a nuclei decays through the cascade $I_i \rightarrow I \rightarrow I_f$. According to [11, 18] $W(\theta)$ is defined as

$$W(\vec{k_1}, \vec{k_2}) = W(\theta) = \sum_k A_{kk} P_k(\cos\theta) \tag{2.1}$$

where θ is the angle between $\vec{k_1}$ and $\vec{k_2}$. A_{kk} is the angular correlation coefficient, which describe the deviation of the coincidence probability from the isotropic case $W(\theta) = 1$. It will be derived in the next section. $P_k(\cos\theta)$ is Legender Polynomials and describes the spatial angular distribution of the γ-ray.

Since electromagnetic radiation have even parity, the sum in equation (2.1) includes only even values of k's. If the radiation patterns are pure multipole orders of L_1 and L_2, i.e. for case of no mixed state (mixed state is a state which has a dipole and quadrupole in the first or second states), one can write equation (2.1) as

$$W(\vec{k_1}, \vec{k_2}) = W(\theta) = 1 + \sum_{k=2}^{k_{max}} A_{kk} P_k(\cos\theta) \tag{2.2}$$

where $k = 2, 4, ..., k_{max}$. By selection rule for angular momentum, $k_{max} = min(2I, 2L_1, 2L_2)$. This show that $k_{max}/2$ is determined by the smallest of the three angular momenta. From equation (2.2) when $W(\theta) = 1$, the result is an isotropic radiation pattern. But when $k = 2 = k_{max}$, anisotropic radiation pattern takes place and therefore, according to selection rule $k_{max}/2 = 1$ which indicate that $I \geq 1$ for anisotropic radiation.

2.2 Extranuclear perturbation

In general the probability of measuring γ_2 in a particular direction relative to the direction of γ_1 depends on the angle between the direction of γ_1 and γ_2. However, as the nucleus remains in the intermediate state for a finite life time, the nuclear spins may interact with the environment. Accordingly, through magnetic dipole or electric quadrupole interactions between the nuclear spins and the extranuclear environment, the orientation of the nuclear spin is altered. The precession of the nuclear spin is reflected by a time dependant angular correlation of γ_1 and γ_2, and is called perturbed angular correlation. This perturbation depends on the magnitude of interaction and the mean life τ of the intermediate state. The magnitude can be described by a precession frequency ω for static perturbation. ω is equal to the Lamor frequency ω_B for magnetic interaction and is proportional to μ (magnetic dipole moment) and H(magnetic field). But, for quadropole case ω is proportional to Q and V_{zz}. The time dependence of directional angular correlation is contained in a perturbation function $G_{kk}(t)$.

For a randomly oriented nnclei (polycystalline) in the local field, seen by an ensemble of atoms, equation (2.1) becomes

$$W(\vec{k_1}, \vec{k_2}) = W(\theta) = \sum_k G_{kk}(t) A_{kk} P_k(\cos\theta) \tag{2.3}$$

Quantum mechanically, if one chooses the quantization axis to coincide with the direction of the first radiation, the interaction causes a transition among the m-state. The transition among m-state can be described by a time dependent density matrix of the intermediate state I. The interaction of the nucleus in its intermediate state I with extranuclear fields is described by Hamiltonian H. This interaction lasts from a time t=0 where a first radiation is emitted to a time t when the second radiation emitted. Within this time interval t the states $|m_a\rangle$ change to different states $|m_b\rangle$ because of the extranuclear perturbation. The transition from states $|m_a\rangle$ to states $|m_b\rangle$ may be represented by a unitary operator $\Lambda(t)$ that describes the evolution of the state vectors $|m_b\rangle$, and thus the

perturbed angular correlation can be expressed as [11]

$$W(\vec{k_1},\vec{k_2},t) = \sum_{m_i,m_f,m_a,m'_a} \langle m_f|H_2\Lambda(t)|m_a\rangle\langle m_a|H_1|m_i\rangle\langle m_f|H_2\Lambda(t)|m'_a\rangle^*\langle|m'_a|H_1|m_i\rangle^* \tag{2.4}$$

where H_1 and H_2 represent the interaction Hamiltonian that governs the two step γ-transition between the nucleus and the radiation field only. The state $|m\rangle$ form a complete set and the state vector $\Lambda(t)$ is expressed as

$$\Lambda(t)|m_a\rangle = \sum_{m_b} |m_b\rangle\langle m_b|\Lambda(t)|m_a\rangle$$

and,

$$\Lambda(t)|m'_a\rangle = \sum_{m'_b} |m'_b\rangle\langle m'_b|\Lambda(t)|m'_a\rangle \tag{2.5}$$

The expansion coefficients are the matrix elements of the time-evolution operator $\Lambda(t)$ in the m representation. The evolution operator satisfies the Schrödinger equation with

$$\begin{aligned}
\frac{\partial\Lambda(t)}{\partial t} &= \frac{-i}{\hbar}H\Lambda(t) \\
\frac{d\Lambda(t)}{\Lambda(t)} &= \frac{-i}{\hbar}Hdt \\
\ln\Lambda(t) &= \frac{-i}{\hbar}Ht \\
\Lambda(t) &= \exp(\frac{-i}{\hbar}Ht).
\end{aligned} \tag{2.6}$$

Using equation (2.5) and equation (2.6), one can write equation (2.4) as

$$\begin{aligned}
W(k_1,k_2,t) = \sum_{m_i,m_f,m_a,m_b,m'_a,m'_b} &\langle m_f|H_2|m_b\rangle\langle m_b|\Lambda(t)|m_a\rangle\langle m_a|H_1|m_i\rangle\langle m_f|H_2|m'_b\rangle^* \\
&\times\langle m'_b|\Lambda(t)|m'_a\rangle^*\langle m'_a|H_1|m_i\rangle^*
\end{aligned} \tag{2.7}$$

Based on the unitary matrix representation, which can be written in terms of the spherical harmonics $Y_k^N(\theta,\phi)$, and the special choice of the quantization axis Z, the time dependent correlation function can be derived as shown in appendix B and the required result is

$$W(k_1,k_2,t) = \sum_{k_1,k_2 N_1 N_2} A_{k_1}(\gamma_1) A_{k_2}(\gamma_2) G_{k_1 k_2}^{N_1 N_2}(t) [(2k_1+1)(2k_2+1)]^{-1/2} \times Y_{k_1}^{N_1}(\theta_1,\phi_1) Y_{k_2}^{N_2 *}(\theta_2,\phi_2) \tag{2.8}$$

where we define the perturbation coefficient $G_{k_1 k_2}^{N_1 N_2}(t)$ as

$$G_{k_1 k_2}^{N_1 N_2}(t) = \sum_{m_a m_b} [(2k_1+1)(2k_2+1)]^{1/2} \langle m_b|\Lambda(t)\rangle \langle m_b'|\Lambda(t)|m_a'\rangle^* \times \begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m_b' & -m_b' & N_2 \end{pmatrix}. \tag{2.9}$$

Hence, the influence of the external perturbation is completely described by the perturbation function $G_{k_1 k_2}^{N_1 N_2}(t)$. The expression (2.8) represents the time differential perturbed angular correlation, i.e. the correlation measured if the second radiation is observed within the time t and $t + \delta t$ after the emission of the first radiation.

2.3 Perturbation by static interaction

If the perturbation of angular correlation is caused by the interaction of the nuclear moments (magnetic or electric) with a stationary externally applied magnetic field or a crystalline electric field gradient, the matrix elements of $\Lambda(t)$ can be expressed in the m representation. We designate the unitary matrix which diagonalizes the interaction Hamiltonian H by U [11]

$$UHU^{-1} = E \tag{2.10}$$

where E is the diagonal energy matrix with the diagonal element E_n (energy eigenvalues).

$$\begin{aligned} U\exp(-i/\hbar Ht)U^{-1} &= U\{1-(i/\hbar)Ht + 1/2((i/\hbar)Ht)^2 + ...\}U^{-1} \\ &= 1-(i/\hbar)Et + 1/2((i/\hbar)Et)^2 + ... \\ U\exp(-i/\hbar Ht)U^{-1} &= \exp(-(i/\hbar)Et). \end{aligned} \tag{2.11}$$

provided that the perturbing Hamiltonian is very small. Using the time evolution operator

$$\Lambda(t) = \exp(-(i/\hbar)Ht),$$

we can write

$$UU^{-1}\exp(-(i/\hbar)Ht)U^{-1}U = U^{-1}\exp(-(i/\hbar)Et)U,$$

$$\exp(-(i/\hbar)Ht) = U^{-1}\exp(-(i/\hbar)Et)U,$$

or

$$\Lambda(t) = U^{-1}\exp(-(i/\hbar)Et)U. \tag{2.12}$$

Therefore, the matrix elements of $\Lambda(t)$ in the m representation are

$$\begin{aligned}\langle m_b|\Lambda(t)|m_a\rangle &= \sum |n\rangle\langle n|\langle m_b|\Lambda(t)|m_a\rangle \\ &= \sum \langle n|m_b\rangle^* \langle n|e^{-(i/\hbar)Ht}|m_a\rangle \\ &= \sum \langle n|m_b\rangle^* e^{-(i/\hbar)E_n t}\langle n|m_a\rangle \end{aligned} \tag{2.13}$$

where m and n are the matrix elements of the unitary matrix U that is obtained by solving eigenvalue equation (2.10). The perturbation

$$G^{N_1N_2}_{k_1k_2}(t) = \sum_{m_a m_b} (-1)^{2I+m_a+m_b}[(2k_1+1)(2k_2+1)]^{1/2}$$

$$\times \exp(-(i/\hbar)(E_n - E_{n'})t)\langle n|m_b\rangle^*\langle n|m_a\rangle\langle n'|m'_b\rangle\langle n'|m'_a\rangle^*$$

$$\times \begin{pmatrix} I & I & k_1 \\ m'_a & -m_a & N_1 \end{pmatrix}\begin{pmatrix} I & I & k_2 \\ m'_b & -m'_b & N_2 \end{pmatrix}$$

If the perturbing field is axially symmetric, then this guarantees that $N_1 = N_2 = N$, but k_1 need not be identical to k_2. The symmetry axis of the interaction can be then chosen parallel to the Z-axis and as the quantization axis for the eigenfunctions of the Hamiltonian H. The eigenfunction $|m\rangle$ and the evolution matrix $\Lambda(t)$ are then diagonal in this representation $U = 1$. [$\eta = 0$ and non diagonal eigenvalues of the Hamiltonian vanish]

$$\langle m_b|\Lambda(t)|m_a\rangle = \sum \langle m|m_b\rangle^* e^{-(i/\hbar)E_m t}\langle m|m_a\rangle$$

$$\langle m_b|\Lambda(t)|m_a\rangle = e^{-(i/\hbar)E_m t}\delta_{mm_a}\delta_{mm_b} \tag{2.14}$$

Therefore, the perturbation factor is reduced to

$$G_{k_1k_2}^{N_1N_2}(t) = \sum_m[(2k_1+1)(2k_2+1)]^{1/2}\begin{pmatrix} I & I & k_1 \\ m'_a & -m_a & N_1 \end{pmatrix}\begin{pmatrix} I & I & k_2 \\ m'_b & -m'_b & N_2 \end{pmatrix}$$
$$\times \exp(-i/\hbar(E_m - E_{m'})t) \tag{2.15}$$

an important relation can be derived from this choice of axes. If the axial symmetric field is parallel to the propagation direction of one of the two radiation (e.g; γ_1), then $Y_{k_1}^N(0,\varphi) = \sqrt{\frac{2k_1+1}{4\pi}}\delta_{N,0}$, and only the term $m = m'$, i.e. $E_m - E_{m'} = 0$, occur. The orthogonality relation of the $3-j$ symbols can be used in equation (2.15) and we obtain $G_{k_1k_2}^{N_1N_2}(t) = \delta_{k_1k_2}$. Therefore, the angular correlation is not influenced by this field. The influence of such a field can be interpreted as a precision of the intermediate nuclear state about $\vec{k_1}$. The projection of I on $\vec{k_1}$ does not change, and the population of the m-states with respect to $\vec{k_1}$ is not disturbed.

In most cases, angular correlations are observed by using a radioactive source that consists of an ensemble of randomly oriented microcrystals . The angular correlation is obtained by averaging over the random directions of the symmetry axes of the microcrytals. Therefore, the perturbation factor of the powder source is

$$G_{kk}(t) = \overline{G_{k_1k_2}^{N_1N_2}(t)}. \tag{2.16}$$

The perturbation factor $G_{kk}(t)$ of the sources that as a whole display no privileged direction is therefore called the *attenuation factor*. The time integrated attenuation factor of the static interaction of the powder source has finite lower limits or is described as " hard core ". For the hard core case the perturbation factor reduces to equation (2.19), i.e. from equation (2.15) one can write the averaged perturbation function as [11]

$$\overline{G_{kk}(\infty)} = \sum_m \begin{pmatrix} I & I & k_1 \\ m'_a & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_b & -m'_b & N_2 \end{pmatrix} \times \exp(-i/\hbar(E_m - E_{m'})t).$$

For this limiting case $m = m'$ and $k_1 = k_2 = k$, and

$$\overline{G_{kk}(\infty)} = \sum \begin{pmatrix} I & I & k \\ m & -m & N \end{pmatrix}^2. \tag{2.17}$$

The orthogonality of the 3-j symbols is given as

$$\sum_{m_1 m_2} \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \begin{pmatrix} j_1 & j_2 & j'_3 \\ m_1 & m_2 & m'_3 \end{pmatrix} = \frac{\delta_{j_3 j'_3} \delta_{m_3 m'_3}}{2j_3 + 1}. \tag{2.18}$$

Hence, $j_3 = j'_3$ and $m_3 = m'_3$ equation (2.17) becomes

$$\overline{G_{kk}(\infty)} = \frac{1}{2k+1}. \tag{2.19}$$

Thus, the values in the hard core cases are independent of the interaction.

For the second order $k = 2$ perturbation of the electrostatic interaction the hard core would be 1/5. The meaning of this lower limit of the static interaction is that the anisotropic distribution of the second γ-ray is never completely lost regardless of the field strength.

It is important to emphasize that the existence of the hard core case is subordinated to the following assumptions

a) The perturbing interaction are static; b) No privileged direction exist in the radioactive source considered as a whole. If either one of these assumptions is violated, the correlation can be quenched, i.e destroyed completely.

2.3.1 Perturbation factor for static electric quadrupole interaction

The interaction between the nuclear quadrupole moment and the electric field gradient at the site of the nucleus is referred as the electric quadrupole interaction. This interac-

tion results in the splitting energy levels of the decaying nucleus. Therefore, if there is splitting of the intermediate level, we can observe the perturbed angular correlation. The Hamiltonian that describes the axially symmetric electric quadrupole interaction and the resulting energy difference from this interaction for the degenerate m-states is given by equation (1.29) and (1.39) respectively. Rewriting the equations

$$H_Q = \sqrt{\frac{\pi}{5}}\, V_{zz}\, T_0^{(2)} \tag{2.20}$$

$$E_m - E_{m'} = \frac{3eQV_{zz}}{4(2I-1)}|m^2 - m'^2| \tag{2.21}$$

and substituting equation (2.21) into (2.15), one obtains the perturbations factor for the axially symmetric quadrupole interaction. That is

$$G_{k_1k_2}^{N_1N_2}(t) = \sum_m{}'[(2k_1+1)(2k_2+1)]^{1/2} \begin{pmatrix} I & I & k_1 \\ m'_a & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_b & -m'_b & N_2 \end{pmatrix}$$

$$\times \exp(-3i(m^2 - m'^2)\omega_Q t)$$

$$= \sum_m{}'[(2k_1+1)(2k_2+1)]^{1/2} \begin{pmatrix} I & I & k_1 \\ m'_a & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_b & -m'_b & N_2 \end{pmatrix} \cos(n\omega_0 t)$$

or

$$G_{k_1k_2}^{N_1N_2}(t) = \sum_n S_{nN}^{k_1k_2} \cos(n\omega_0 t), \tag{2.22}$$

where the summation index n is over all positives integers and zero, i.e.,

$$n = \begin{cases} |m^2 - m'^2| \ for\ integer\ I \\ 1/2|m^2 - m'^2| \ for\ half\ integer\ I \end{cases} \tag{2.23}$$

The coefficient $S_{nN}^{k_1k_2}$ is given by

$$S_{nN}^{k_1k_2} = \sum_{mm'}{}'[(2k_1+1)(2k_2+1)]^{1/2} \begin{pmatrix} I & I & k_1 \\ m'_a & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m'_b & -m'_b & N_2 \end{pmatrix} \tag{2.24}$$

where the prime on the summation sign indicates that the summation over m and m' should only include those terms where m and m' satisfies the condition (2.23).

POLYCRYSTAL SOURCE

In general the correlation functions of polycrstalline sources are given as according to the relation (2.3). For axially symmetric ($\eta = 0$) quadrupole interactions the time dependent perturbation term is represented by the attenuation coefficient $G_{kk}(t)$ [11].

$$G_{kk}(t) = \sum_{m,m'}{}' \begin{pmatrix} I & I & k \\ m' & -m & p \end{pmatrix}^2 \exp(-3i(m^2 - m'^2)\omega_Q t) \tag{2.25}$$

The quadrupole frequencies ω_Q is defined by equation (1.40) and $p = -m' + m$. This expression can also be written as

$$G_{kk}(t) = \sum_n S_{kn} \cos(n\omega_0 t) \tag{2.26}$$

with

$$S_{kn} = \sum_{m,m'} \begin{pmatrix} I & I & k \\ m' & -m & -m'+m \end{pmatrix}^2, \tag{2.27}$$

and n satisfies the condition in equation (2.23). The S_{kn} can be determined to all allowed m-levels of the intermediate state, m and m' (see appendix C). The time integrated coefficients from polycrystalline samples can be obtained from [11]

$$G_{kk}(\infty) = \frac{1}{\tau}\int_0^\infty \exp(-t/\tau) G_{kk}(t) dt$$

where $G_{kk}(t)$ is by equation (2.22).

$$\begin{aligned} G_{kk}(\infty) &= \sum_n S_{kn} \frac{1}{\tau}\int_0^\infty \exp(-t/\tau)\exp(-in\omega_0 t) dt \\ &= \sum_n S_{kn} \frac{1}{\tau}\int_0^\infty \exp(-t(\frac{1+in\omega_0\tau}{\tau})) dt \\ &= \sum_n S_{kn} \frac{1}{\tau}\tau \frac{-1}{1+in\omega_0\tau} \exp(-t(\frac{1+in\omega_0\tau}{\tau}))|_0^\infty \\ &= \sum_n S_{kn} \frac{1}{1+in\omega_0\tau} \\ &= \sum_n S_{kn} \frac{1-in\omega_0\tau}{1+(n\omega_0\tau)^2} \end{aligned}$$

or

$$G_{kk}(\infty) = \sum_n S_{kn}[\frac{1}{1+(n\omega_0\tau)^2} - \frac{in\omega_0\tau}{1+(n\omega_0\tau)^2}]$$

Taking the real part only

$$G_{kk}(\infty) = \sum_n S_{kn}\frac{1}{1+(n\omega_0\tau)^2}. \tag{2.28}$$

The limiting values for infinitely strong axially symmetric static quadrupole interaction are determined by the value of the hard core, which is given here in terms of the coefficient S_{kn} as

$$G_{kk}(\infty) = S_{k0}$$

In a non-axially symmetric magnetic or electric field the S_{kn} are polynomial function of η.

2.4 Measurement of the time spectra in perturbed angular correlation (PAC)

Up to now we have discuss only the theoretical back ground of PAC. In this section we will discribe how the data are measured experimentally and how the results compare with predictions based on the appropriate theoretical perturbation functions.

In measuring the angular correlation between γ_1 and γ_2 as a function of time elapsed between the arrival of both photons, at least two detectors are necessary. The simplest form of the experimental set up is a two detector system in which one is fixed and the other one is movable.

When both γ_1 and γ_2 are detected in coincidence unit (coincidence unit is a clock which detect both gamma rays and allow them to pass only if their energy has the desired value), information can be obtained on the orientations of the same nuclear spins I at the times of the emission of the two γ rays. The spin precession frequency ω is deduced from the measured changes of the spin orientation ΔI during the time t, between the emission of

both gamma rays for an ensemble of probe atoms. Thus, whenever a radioactive probe atom decays the frequency of the γ that is emitted provides information about the field strength at its lattice site which related to field strength according to equation (1.40).

The time differential coincidence count rate $N_{ij}(\theta, t)$ at a time t, where t is the elapsed time between the emission γ_1 and γ_2, is given by [19]

$$N_{ij}(\theta, t) = N_0 \exp(-t/\tau_N) W(\theta, t) + B \tag{2.29}$$

where B is the time independent background random coincident count rate, $W(\theta, t)$ represents the time dependent angular correlation, and θ is the angle between the two detectors. For k=2, the coincidence count rate recorded by detectors i and j is

$$N(\theta, t) = N_0 \exp(-t/\tau_N)\{1 + A_{22} G_2(t) P_2(\cos\theta)\} + B. \tag{2.30}$$

Since detector 2 is movable, eight coincidence spectra $N_{ij}(\theta, t)$ are possible similar to the arrangement of four fixed detectors that each form 90 degree to each other. The ratio function R(t) can be obtained from the relation [18]

$$R(t) = \frac{N(180°) - N(90°)}{N(180°) + 2N(90°)}. \tag{2.31}$$

and,

$$R(t) \simeq A_{22} G_2(t). \tag{2.32}$$

From equation (2.32) the experimental R(t) can be fitted with the perturbation function

$$R(t) = A_{22} G_2(t) = A_{22} \sum_{n=0}^{3} f_i S_{2n} \cos n\omega_0 t \tag{2.33}$$

Once the relation between the experimentally measured quantity R(t) and the perturbation factor $G_{22}(t)$ is known, it is possible to determine ν_Q and η from the measurable spectra. For polycrystalline sources the coefficient S_{2n} for $\eta = 0$ are already calculated according to equation (2.27) in appendix C. But for $0 < \eta \leq 1$, S_{2n} can be predicted from the Fourier transform graph of the perturbation function $G_2(t)$ [20].

The general situation observed in the $\gamma - \gamma$ decay of angular correlation is summarized in Fig 2.2. First a radioactive probe such as ^{111}In nucleus decays by emitting a photon with energy =171KeV and goes to the intermediate state. At this intermediate state the splitting of the energy level occur which can be described by its frequencies and EFG. The intermediate level inturn decays to the ground state through emission of the second gamma ray of energy 245KeV,with the formation of the daughter atom cadmium. Using the two detectors the $\gamma\gamma$-coincidence spectrum is measured and converted to PAC time spectra and its Fourier transform spectra.

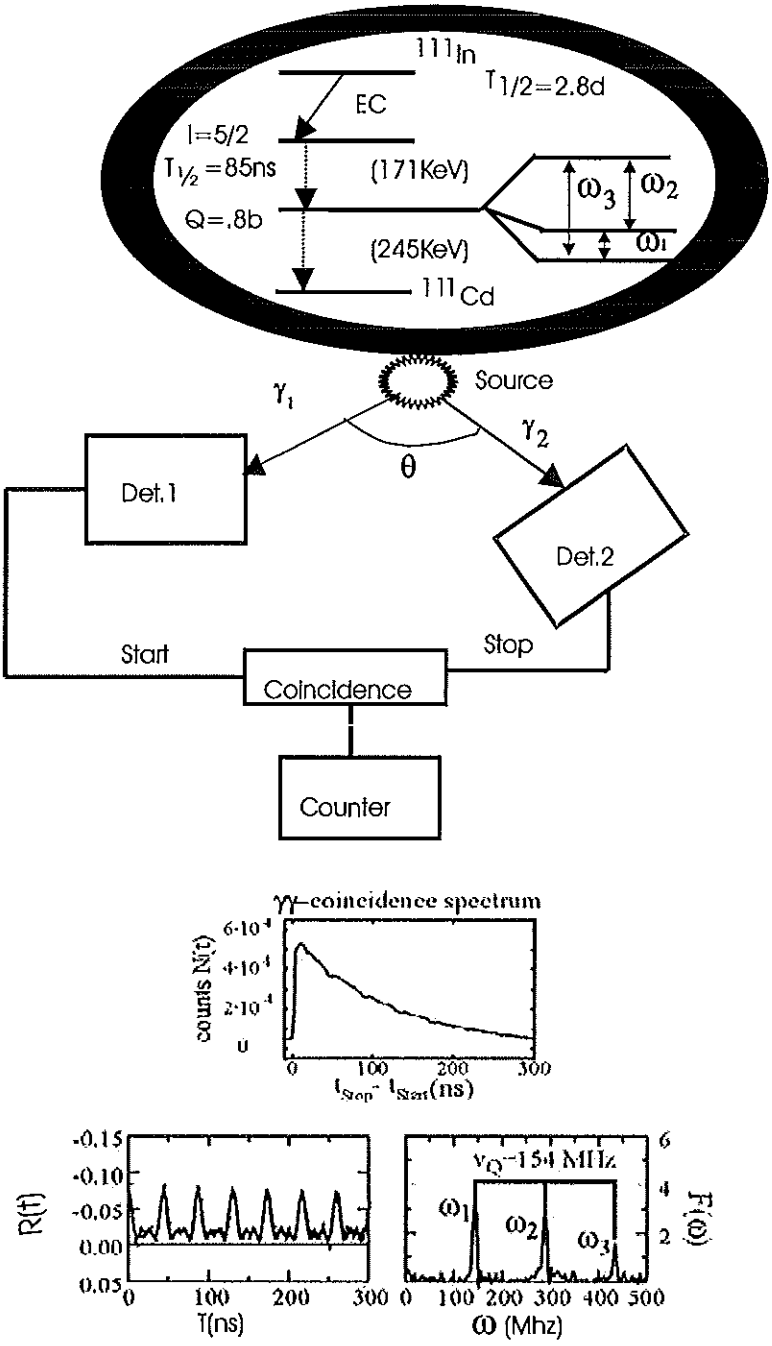

Figure 2.2: Illustration of coincidence measurements. The two lower figures are taken from [26]

entire charge distribution will be disturbed at the site of the impurity because of the size difference and change of the lattice parameters near the complex.

In some cases, it is customary to find a variety of impurity atoms simultaneously in the semiconductors. These impurities can be either acceptors or donors in the host lattice. At suitable thermodynamic conditions the impurities can form an acceptor-donor pair which in many cases contributes for the passivation of electrically active centers. Such pairs have been detected by different methods such as infrared absorption (IR) and perturbed $\gamma-\gamma$ angular correlation PAC. In the present discussion, results of the PAC measurement is presented. The PAC is very suitable for the detection of such pairs since it is sensitive to the nearest neighbour defect.

3.2 Formation and property of the $In-Te$ pair in silicon

In the current investigation of indium-donor defect interaction in silicon, we employed Czochralski-grown crystalline silicon with ⟨110⟩ surface orientation. The impurity atoms (indium and tellurium) were incorporated in silicon by ion implantation. Tellurium was implanted with dose $2.5\times10^{15} atoms/cm^2$ at 50KeV in the mass separator facility at University of Bonn. Following tellurium implantation and recovery from radiation induced damage, the probe atom indium (^{111}In) was implanted at 160KeV with dose ranging between (10^{12} *and* 10^{13}) $atoms/cm^2$. This dose range corresponds to a local concentration of probe atoms in the order of $10^{17} atoms/cm^3$. After the impurity implantation the samples was annealed at different temperatures in vacuum ($\simeq 0^{-6} mbar$) in an isochronal annealing program for 10 minutes holding time. Consequently, the time spectra where taken right after each annealing stage. Fig 3.1 shows the time spectra taken after different annealing temperatures . Severe radiation damage in the sample was observed immediately after implantation. Panel 1 of Fig 3.1 show the time spectrum after annealing the sample at $500°C$. This spectrum shows substantial improvement of the radiation damage

as indicated by the rise in the value of the anisotropic coefficient (A_{22}). The spectrum of course slightly modulated by a quadrupole interaction frequency due to the small fraction of the probe atoms. This frequency is highly damped because of the EFGs coming from defect complexes in close proximity of the probe atoms in the sample.

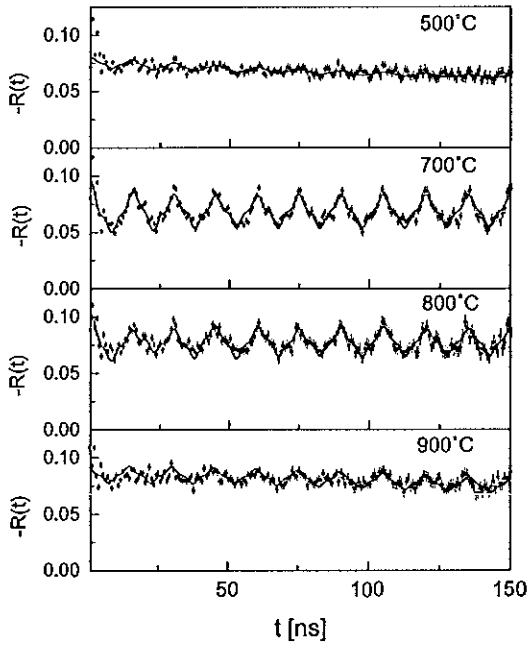

Figure 3.1: PAC spectra of (Cd-Te) doped in silicon at different annealing temperature

At $700°C$, one can clearly see a well defined frequency modulation resulting from the formation of a unique probe-defect complex in the sample. Again, at higher annealing temperature the frequency still exists but the amplitude of modulation decreases suggesting the dissociation of the formed complex.

The observed frequency is assigned to the formation of the indium- tellurium pair in the silicon. To our knowledge , this frequency has never been observed in any intrinsic or impurity doped silicon. The quadrupole interaction frequency of $\nu_Q = 444MHz$ is derived from the time spectra representing $In - Te$ pair in silicon. Moreover, orientation

measurements were also taken by pointing the major crystal axes toward the detectors (i.e. ⟨100⟩, ⟨110⟩ and ⟨111⟩). The measurements suggest that orientation of the major component of EFG tensor(V_{zz}) lies along ⟨100⟩ [20, 21]

The $\gamma - \gamma$ coincidence spectrum R(t) recorded as function of time is described by the perturbation function discussed in Chapter 2. Recall that

$$R(t) = A_{22}G_2(t),$$

$$R(t) = A_{22}\sum f_i G_2(t), \tag{3.1}$$

where

$$G_2(t) = \sum_0^3 S_n \exp(-\delta_n t)\cos(w_n t). \tag{3.2}$$

Here f_i is fractional population of site (i) characterized by ν_{Qi} and η_i. The anisotropy coefficient $A_{22} = -0.1025$ represents the effective spatial anisotropy of the $\gamma - \gamma$ angular correlation. δ_n is the Lorenzian distribution for the different EFG distribution around the mean value. The value of δ is 2.150% for annealing temperature of $500°C$ and 0.15% for all other cases.

The coefficient S_{2n} depends on the orientation of the EFG tensor with respect to the host lattice (for single crystal). The experimental data for S_{2n} is given in table 3.1. The strong dependence of S_{2n} values on angle θ is clearly seen from the table. For small change in angle θ large change in S_{2n} values is observed (see Fig. 2.1).

S_{2n}	$\theta = 2.72,\ \phi = 90$	$\theta = 0.03, \phi = 0.3$	$\theta = 90,\ \phi = 0.04$
S_{21}	.322152	.000023	.642857
S_{22}	0	.000079	0
S_{23}	.178974	.000005	.357143

Table 3.1: Direction dependence of the S_{2n} values for single crystal.

In this experiment, the Te^{+2} ions are assumed to occupy octahedral interstitial site in the diamond structure of silicon. The strong quadrupole coupling constant frequency

T_a	$\sum f_i\%$	unperturbed fraction	polycrystal	ω_0
500°c	9.9(1)	65(3)	25	418.0
600°c	26.7(1)	58(1)	15	418.0
650°c	29.5(2)	55(2)	15.3	418.0
700°c	32.6(2)	52(2)	15	418.0
800°c	28.1(1)	57(1)	14	418.0
900°c	15.5(2)	66.3(1)	18.2	418.0

Table 3.2: Relation between annealing temperature and fraction of the probe atoms exposed to electric field gradient

indicates that this interaction is related to associated EFG, V_{zz} through the relation

$$\nu_Q = eQV_{zz}/h \tag{3.3}$$

Since we are performing the experiment for the case of $I = 5/2$, then the fundamental interaction frequency $\omega_0 = 3\pi\nu_Q/10$. In panel 1 of the Fig 3.1, the sample is annealed for 10 min at $T_a = 500°C$ and 10(1)% of the probe atoms in the sample experiences an EFG, and a large population 65(3)% of them are still in the symmetric region. After annealing at $T_a = 700°C$, 32.6(2)% of the probe atoms experience unique EFGs and 52(2)% are unperturbed. The frequency modulation is slightly damped due to the presence of imperfection close to the Cd-Te complex.

As the annealing temperature is increased, more Cd-Te pairs are formed until the pair fraction reaches 32.6(2)% of the probe atoms at $T_a = 700°C$. When the annealing temperature is further increased above $T_a = 700°C$ the formed complex start to dissociate and completely vanishes above $1000°C$. For all cases the fraction of the probe atoms not mentioned here are situated in undefined lattice location.

The influence of the annealing temperature T_a on the population is shown in Fig.3.2. The Cd-Te implantation at $23°C$ causes radiation damage and/or amorphization of the

Si crystal. During annealing at temperatures up to 700°C, recrystallization occurred and Cd-Te complexes were formed, which lead to the observed quadrupole interaction frequency (QIF). Above this temperature the concentration of Te^{+2} ions decreases and the probability for the formation of Cd-Te complex decreases. Hence the fractional population that experiences the observed EFG will decrease.

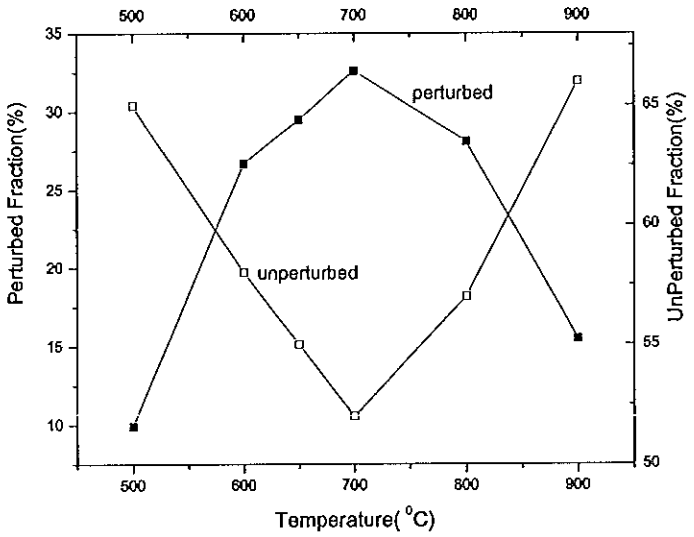

Figure 3.2: Fractional population of Cd-Te that experiences an EFG during annealing at different temperatures and unperturbed fraction during this annealing process.

3.3 Temperature-dependent Electric Field Gradient

In this section, the temperature dependence of the EFG at the ^{111}Cd nucleus in silicon is discussed. To examine the temperature dependence of the EFG another sample was prepared using the process given in section 3.2 but changing the annealing condition. Following the implantation of tellurium and indium the sample was annealed at 500°C and 700°C consecutively for 10minuetes, and then the sample was stored in an oven. These

annealing temperature were chosen in order to observe maximum fractional population of the In-Te complex. The temperature dependance of the QIF was examined between the temperatures $20°C$ and $725°C$. The time spectra taken at different sample temperatures showed a significant change in QIF. The QIF decreases from $\nu_Q = 444MHz$ at room temperature to $\nu_Q = 388MHz$ at the highest temperature.

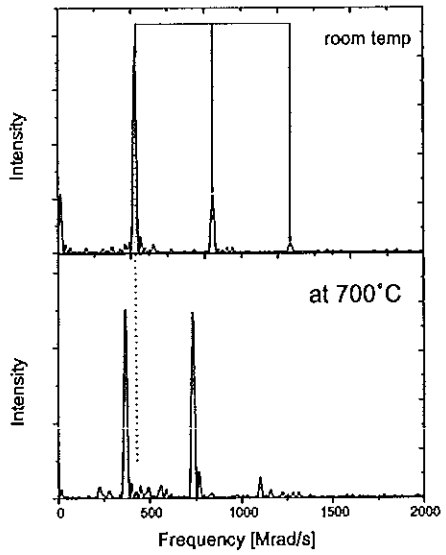

Figure 3.3: Fourier transform of the PAC time spectra taken at room temperature and $700°C$

The strong temperature dependence of the quadrupole interaction frequency is seen from Fourier transform of the PAC time spectra. The first peak the interaction frequency observed at room temperature is shifted to the left at $700°C$ indicating the decrease in frequency. According to equation (3.3) this indicate that the electric field gradient decreases.

At the present time, there is no clear theoretical understanding that explains the change of the electric field gradient at the site of the nucleus. Often the empirical $T^{3/2}$ relation is used to explain the same effects in metals. However, the change of the quadrupole

interaction frequency in semiconductors is belived to resulte from the existence of different charge states of the complex in the sample at the time of the measurements [17]. Thus, the observed complex would be formed by Columbic attraction between substitutional indium (In^-) and ionized tellurium (Te^+ *and* Te^{++}) during thermal treatment of the samples. The change of the charge state of the probe atom as it transform from In^- to Cd^{-2} would form complexes $(Cd-Te)^-$ and $(Cd-Te)^0$ which causing two different EFGs $V_{zz}(-)$ and $V_{zz}(0)$ respectively. However, the population of the states could vary with temperature resulting in different averaged EFG values.

The formation of indium-donor pairs in silicon using PAC technique have been studied by [4, 22, 23]. They measured EFG at the cadmium-donor pairs after electron capture decay of the ^{111}In probe atoms to ^{111}Cd. In this case the trapping and detrapping of impurities or defects is governed by the electronic properties of the indium atom. However, since the EFG is measured at the cadmium nucleus, it yields information about the electronic structure of the cadmium impurity complexes. In their paper [22] they indicate that the EFG of three indium donor pairs (donors: P, As and Sn) have similar step-like temperature dependence of the coupling frequency ν_Q. In their discussion the drastic change in the strength of the field gradient is attributed to the electronic effects, based on the fact that thermal lattice expansion or increases in the phonons amplitude cause comparatively small and smooth variation.

The electronic capture decay of ^{111}In transforms the electrically inactive In-Te pairs into a Cd-Te pair that can be regarded as an acceptor. Here the model which refers to the charge state of the Cd-donor complex Cd-D is used which was applied for first time to the semiconductor In_2Te_5 [22 and references therein].

At low temperature Te^+ is dominant because of high concentration of Te. Hence, most of the complexes are ionized to the $(Cd-Te)^-$ state that produces an electric field $V_{zz}(-)$. At high temperature more Te^{+2} state is populated. At higher temperatures the $(Cd-Te)^0$ state dominates and produce EFG, $V_{zz}(0)$. In semiconductors as the temperature increases the Fermi level E_F shifts to the center of the energy gap and the

ionization probability P(T) is dependant on the temperature. P(T) is described as, using the Fermi-Dirac probability distribution,

$$P(T) = \frac{1}{1 + g\exp\{[E_{Cd-D} - E_F(T)]/kT\}}, \tag{3.4}$$

where E_{Cd-D} is the energy level and g is the degeneracy factor of the Cd-D acceptor. The charge state of the acceptor is thus characterized by a dynamic equilibrium between $(Cd-Te)^-$ and $(Cd-Te)^0$ resulting in fluctuating field gradients. Assuming an identical principal axes system for V(-) and V(0), the time average of the EFG is given by [22], as

$$V_{zz}(T) = (1 - aT)\{V_{zz}(0) + [V_{zz}(-) - V_{zz}(0)]P(T)\}. \tag{3.5}$$

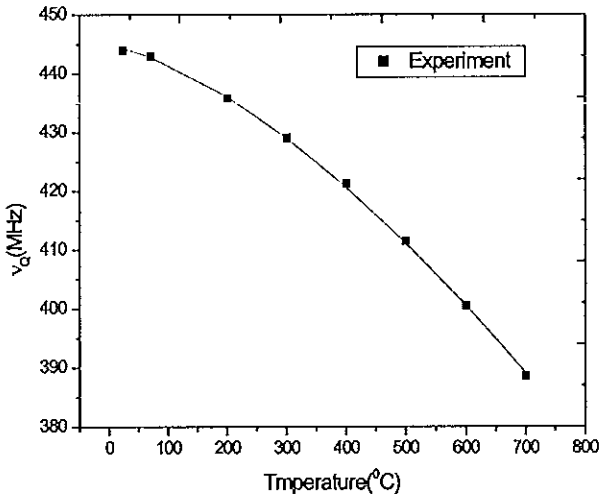

Figure 3.4: Temperature dependence of the QIF

The empirical factor $(1 - aT)$ is added to account for a small temperature dependence of the EFG caused by thermal effects. Our sample can be considered as n type semiconductor because of high concentration of Te (high Te dose). However, at the local site the Columbic interaction between the In^- ions and ionized Te forms Cd-Te states. The Cd-Te states

act as acceptors and form acceptor energy levels. Based on this the sample is assumed a P type semiconductor. For P-type semiconductors, the temperature dependant Fermi level is given by [24, 25] as,

$$E_F = E_v + kT\ln(N_v/N_A)$$

or,

$$E_F = E_v - kT\ln(N_A/N_V) \tag{3.6}$$

But, $N_v = 2\left(2\pi kTm_e^*/h^2\right)^{3/2}$

where N_v is the effective density of states in the valence band, k is Boltzmann constant $= 1.38\times10^{-23}J/K$, T is temperature in K, m_e^* is the effective mass of electrons $= m_0\times0.2$ for silicon (m_0 is mass of free electrons $= 9.11\times10^{-31}Kg$) and h is planck's constant $= 6.625\times10^{-34}$ J.sec. One can write N_v as,

$$N_v = P_0T^{3/2} \tag{3.7}$$

where, $P_0 = 2\left(2\pi km_e^*/h^2\right)^{3/2} = 3.66\times10^{20}/m^3k^{3/2}$. Therefore E_F can be written as

$$E_F = E_v - kT\ln(N_A/P_0T^{3/2}). \tag{3.8}$$

Substituting the value of E_F from equation(3.8) into equation(3.4) gives,

$$P(T) = \frac{1}{1+g\exp\{[E_a - E_v + kT\ln(N_A/P_0T^{3/2})]/kT\}},$$

$$P(T) = \frac{1}{1+g[\exp\Delta E_a/kTN_A/P_0T^{3/2}]},$$

or

$$P(T) = \frac{1}{1+g\{\frac{N_A}{P_0T^{3/2}}\exp\frac{\Delta E_a}{kT}\}}. \tag{3.9}$$

Now, substituting equation (3.9) into equation (3.5) and modifying the linear temperature dependence of equation (3.5) by $T^{3/2}$, we obtain

$$V_{zz}(T) = (1-aT^{3/2})[V_{zz}(0) + [V_{zz}(-) - V_{zz}(0)]\frac{1}{1+g[\frac{N_A}{P_oT^{3/2}}\exp\frac{\Delta E_a}{kT}]}],$$

or

$$V_{zz}(T) = (1 - P_1 T^{3/2})[P_2 + \frac{P_3}{1 + g[P_4 T^{-3/2} exp(\frac{P_5}{T})]}], \tag{3.10}$$

where,

$$P_1 = a,$$

$$P_2 = V_{zz}(0),$$

$$P_3 = V_{zz}(-) - V_{zz}(0),$$

$$P_4 = N_A/P_0,$$

$$P_5 = \Delta E_a/k,$$

$$N_A = P_4 \cdot P_0 \tag{3.11}$$

$$\Delta E_a = E_{Cd-Te} - E_v = P_5 k. \tag{3.12}$$

We use equation (3.10) to fit the data. During fitting P_1 was treated as free parameter and its value is obtained as 6.7×10^{-6}. This small value of P_1 clearly suggests that the empirical factor $(1 - aT^{3/2})$ has no significant effect on equation (3.5). The acceptor concentration N_A is also treated as a free parameter and is obtained as $N_A = 9 \times 10^{15}(1) atoms/cm^3$. The Cd-Te energy level is calculated from equation (3.12) and equals 0.21(1)eV. Moreover, the frequency due to the two EFGs of the given states $(Cd - Te)^-$ and $(Cd - Te)^0$ is found to be 444(1)MHz and 388(MHz), respectively.

Conclusions

In this thesis, crystalline silicon hosts were used to investigate $In - Te$ (donor-defect) interactions. In and Te impurities were incorporated in silicon by the implantation method with dose rates of $2.5 \times 10^{15} atoms/cm^3$ at 50KeV and $(10^{12} - 10^{13}) atoms/cm^3$ for Te and In, respectively.

The time spectra taken at various annealing temperatures showed a unique quadrupole interaction frequency associated with the presence of tellurium in silicon. From these spectra an interaction frequency of $\nu_Q = 444 MHz$ was derived. This frequency represent the formation of $In - Te$ in silicon. With small deviation from theoretical results of S_{2n} values, the measurements suggest that the orientation of the major component of EFG tensor (V_{zz}) lies along $\langle 100 \rangle$. The frequency damping (2.15(0)%) is determined for spectrum taken at $500°C$ and 0.15(1)% for all spectra measured at higher temperatures.

The measured interaction frequency showed strong dependence on temperature in the temperature range between $20°C$ and $700°C$. The drastic change of QIF can be explained by the model which assumes two different charge states at the time of the measurements. States such as $(Cd - Te)^-$ and $(Cd - Te)^0$ were assumed during the calculation. The results give two different magnitude of the EFG as $V_{zz}(-)$ and $V_{zz}(0)$ respectively. Based on calculation in this work the concentration of the charged complex (Cd-Te) is $9 \times 10^{15}(1) atoms/cm^3$ and its energy level above the valence band is 0.21(1)eV.

Appendix A

The 3-j symbols

Calculation of The 3-j symbols for $k = 2$ and spin $I = 5/2$

The 3-j symbols : for any angular momentum j_1, j_2, j_3 with projection m_1, m_2, m_3 respectively [12]

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = (-1)^{j_1-j_2-m_3}\Delta(j_1,j_2,j_3)W_3(j_1,j_2,j_3,m_1,m_2,m_3) \tag{A.1}$$

where

$$\Delta(j_1,j_2,j_3) = [\frac{(j_1+j_2-j_3)!(j_1-j_2+j_3)!(-j_1+j_2+j_3)!}{(j_1+j_2+j_3+1)!}]^{1/2} \tag{A.2}$$

$$\begin{aligned} W_3(j_1,j_2,j_3,m_1,m_2,m_3) = {} & [(j_1+m_1)!(j_1-m_1)!(j_2+m_2)!(j_2-m_2)!(j_3+m_3)!(j_3-m_3)!]^{1/2} \\ & \times \sum_{\nu} \frac{(-1)^{\nu}}{\nu!(j_1+j_2-j_3-\nu)!(j_1-m_1-\nu)!(j_2-m_2-\nu)!} \\ & \times \frac{1}{(j_3-j_2+m_1+\nu)!(j_3-j_1-m_2+\nu)!} \end{aligned} \tag{A.3}$$

and the summation is over zero and positive values of ν for which the factorial is not negative. Usiug this formula we can evaluate the 3-j symbols as $j_1 = j_3 = I, j_2 = 2, m_1 = -m, m_2 = 0$ and $m_3 = m$.

Hence,

$$\begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix} = \sqrt{\frac{(2)!(I-2+I)!(2)!}{(I+2+I+1)!}}[(I+m)!(I-m)!(2)!(2)!(I-m)!(I+m)!]^{\frac{1}{2}}$$

$$\times \sum_{\nu} \frac{(-1)^{\nu}}{\nu!(2+I-I-\nu)!(I+m-\nu)!(2-0-\nu)!} \frac{1}{(I-2-m+\nu)!(I-I-0+nu)!}$$

For convenience this equation can be grouped into three parts A, B, and C, with

$$A = \left[\frac{(2)!(I-2+I)!(2)!}{(I+2+I+1)!}\right]^{1/2},$$

$$B = [(I+m)!(I-m)!(2)!(2)!(I-m)!(I+m)!]^{1/2},$$

and

$$C = \sum_{\nu}\left[\frac{(-1)^{\nu}}{\nu!(2+I-I-\nu)!(I+m-\nu)!(2-0-\nu)!}\right] \times \frac{1}{(I-2-m+\nu)!(I-I-0+\nu)!}$$

$$A - 2\left[\frac{(2I-2)!}{(2I+3)(2I+2)(2I+1)(2I)(2I-1)(2I-2)!}\right]^{1/2}$$

$$= \frac{2}{\left[(2I+3)(2I+2)(2I+1)(2I)(2I-1)\right]^{1/2}}$$

$$B = \left[[(I+m)!]^2[2!]^2[(I-m)!]^2\right]^{\frac{1}{2}}$$

$$= 2(I+m)!(I-m)!$$

In C we have only three possible values of ν, namely $\nu = 0, 1, 2$ since the other terms make the factorials negative. For $\nu = 0$

$$C = \frac{1}{0!2!(I+m)!2!(I-m-1)!0!} = \frac{-1}{(I+m-1)!(I-m-1)!}.$$

For $\nu = 1$,

$$C = \frac{-1}{1!.1!(I+m-1)!1!(I-m-1)!1!} = \frac{-1}{(I+m-1)!(I-m-1)!}.$$

For $\nu = 2$,

$$C = \frac{1}{2!0!(I+m-2)!0!(I-m)!2!} = \frac{1}{4(I+m-2)!(I-m)!}$$

$$C = \frac{-1}{(I+m-1)!(I-m-1)!} - \frac{1}{(I+m-1)!(I-m-1)!} + \frac{1}{4(I+m-2)!(I-m)!}.$$

Using the above, the matrix elements of numerator in equation (1.36) gives

$$\begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix} = \frac{2(3m^2 - I(I+1))}{\left[(2I+3)(2I+2)(2I+1)(2I)(2I-1)\right]^{\frac{1}{2}}}. \tag{A.4}$$

Calculating the values of the matrix elements in the denominator of equation (1.36) gives

$$\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix} = A'.B'.C'$$

where A', B' and C' are

$$A' = \left[\frac{(I+2-I)!(I-2+I)!(-I+2+I)!}{(2I+3)!}\right]^{\frac{1}{2}} = \frac{2}{\sqrt{(2I+3)(2I+2)(2I+1)(2I)(2I-1)}}$$

$$B' = \left[(I-I)!(I+I)!(2!\cdot 2!)(I+I)!(I-I)!\right]^{\frac{1}{2}} = 2(2I)!$$

$$C' = \sum_{\nu} \frac{(-1)^{\nu}}{\nu!(2-\nu)!(2I-\nu)!(2-\nu)!(\nu-2)\nu!}.$$

The value of ν that gives only non negative factorial is 2. Using this value C' is given as

$$C' = \frac{1}{4(2I-2)!}.$$

Based on the above the values of the matrix element in denominator are

$$\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix} = A'B'C' = \frac{2}{\sqrt{(2I+3)(2I+2)(2I+1)(2I)(2I-1)}} \times 2(2I)! \times \frac{1}{4(2I-2)!}$$

$$= \frac{2I(2I-1)}{\sqrt{(2I+3)(2I+2)(2I+1)(2I)(2I-1)}} \tag{A.5}$$

Combining equation (A.4) and equation (A.5) results in,

$$\frac{\begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix}}{\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}} = \frac{\frac{2(3m^2 - I(I+1))}{\sqrt{(2I+3)(2I+2)(2I+1)(2I)(2I-1)}}}{\frac{2I(2I-1)}{\sqrt{(2I+3)(2I+2)(2I+1)(2I)(2I-1)}}}$$

which simplified gives

$$\frac{\begin{pmatrix} I & 2 & I \\ -m & 0 & m \end{pmatrix}}{\begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix}} = \frac{3m^2 - I(I+1)}{I(2I-1)} \tag{A.6}$$

Appendix B

Time dependent correlation function

Derivation of time the dependent correlation function and the perturbation function

For any two steps γ transition between two levels (such as level A and level B with corresponding eigenvalue 'a' and 'b' respectively) the density matrix is defined as [11]

$$\langle b|\varrho_B|b'\rangle = \sum_{a\ a'} \langle b|H|a\rangle\langle a|\varrho_A|a'\rangle\langle b'|H|a'\rangle^* \tag{B.1}$$

Therefore, the density matrix in the intermediate state is

$$\langle m|\varrho(k_1)|m'\rangle = \sum_{m\ m'} \langle m|H_1|m_i\rangle\langle m_i|\varrho_i|m'\rangle\langle m'|H_1|m_i'\rangle^* \tag{B.2}$$

In equations (B.1) and (B.2) the angle independent factors have been set equal to 1(one). The matrix $\langle m|H_i|m_i\rangle$ represents $\langle Im|H_1|I_i m_i\rangle$.
For the second transition $a = m_i, b = m_f$ in equation (B.1) and equation (B.2), where i and f indicates the initial and final states, respectively

$$\begin{aligned}\langle m_f|\varrho(k_1,k_2)|m_f'\rangle = \sum_{m\ m'\ m_i\ m_f} &\langle m_f|H_2|m\rangle\langle m|H_1|m_i\rangle\langle m_i|\varrho_i|m_i'\rangle \\ &\times\langle m'|H_1|m_i'\rangle^*\langle m_f'|H_2|m'\rangle^*\end{aligned} \tag{B.3}$$

Furthermore, the density matrix equation (B.2) can be written as

$$\langle m|\varrho(k_1)|m'\rangle = \sum_{m_i}\langle m|H_1|m_i\rangle\langle m_i|H_1|m_i'\rangle^* \tag{B.4}$$

$$\langle m'|\varrho(k_2)|m\rangle = \sum_{m_f}\langle m_f|H_2|m\rangle\langle m_f|H_2|m\rangle^* \tag{B.5}$$

$$\langle Im|H|I_i m_i'\rangle = \sum_{LM}|LM\rangle\langle ImLM|H|I_i m_i'\rangle \tag{B.6}$$

The eigenfunctions in the two systems are related by

$$\langle k\sigma|LM\pi\rangle = \sum_{\mu}\langle 0\sigma|LM\pi\rangle D^L_{\mu M}(k\rightarrow Z) \tag{B.7}$$

where $D^L_{\mu M}(k\rightarrow Z)$ are the rotation group operators which is given below. The matrix $\langle k\sigma|LM\pi\rangle$ describes the eigenfunction of the radiation R corresponding to eigenvalues L, M, and π of the angular momentum operators, the Z-component of angular momentum and parity respectively. σ is the index of polarization. The expression of this matrix holds for both directional and polarization correlation. However, we are not interested in the polarization part so π and σ are neglected. The rotational group operators are given as [11]

$$D^L_{\mu M}(k\rightarrow z) = D^{L*}_{M\mu}(z\rightarrow k) \tag{B.8}$$

$$D^L_{\mu 0}(\phi,\theta,\gamma) = \sqrt{\frac{4\pi}{2L+1}}Y_L^{\mu *}(\theta,\phi) \tag{B.9}$$

$$D^L_{0\mu}(\phi,\theta,\gamma\rangle = \sqrt{\frac{4\pi}{2L+1}}Y_L^{-M}(\theta,\phi) \tag{B.10}$$

$$D^L_{00}(\phi,\theta,\gamma) = P_L(cos\theta) \tag{B.11}$$

$$D^L_{M0}(z\rightarrow k) = D^{L*}_{\mu 0}(k\rightarrow z) = \sqrt{\frac{4\pi}{2L+1}}Y_L^{\mu}(\theta,\phi) \tag{B.12}$$

$$\sum_{M'} D^L_{\mu M'}(k\rightarrow z')D^L_{M'M}(z'\rightarrow k) = D^L_{\mu M}(k\rightarrow z). \tag{B.13}$$

The matrix element $\langle ImLM|H|I_i m_i\rangle$ can be written as

$$D^L_{\mu M}(k\rightarrow z') = \sum_{I_i' m_i'}(ImLM|I_i m_i'\rangle\langle I_i m_i IL|H|I_i m_i\rangle. \tag{B.14}$$

Here I and L must be retained since they are dependent on the other intermediate states. We now use the fact that H for physical reasons is invariant under rotations [11]. The matrix element $\langle I_i m_i IL|I_i m_i\rangle$ is non-zero only if $I_i' = I_i, m_i' = m_i$. Furthermore it is independent of the magnetic quantum number m_i.

Combining equation (B.6) and (B.7) and replacing the Clebsch-Gordan coefficient by more symmetric Wigner $3-j$ symbol (B.17) below and omitting uninteresting factors (like 4π which does not contain information about the intermediate state), we get the final expression for a typical matrix element

$$\langle m|H|m_i\rangle = \sum_{LM\mu}(-1)^{-I+L-m_i}\begin{pmatrix} I & L & I_i \\ m' & M & -m_i \end{pmatrix}\langle 0|L\mu\rangle\langle I||L||I_i\rangle D^{L*}_{M\mu}(z\to k) \qquad \text{(B.15)}$$

In addition to the above expressions we need the explicit form of the matrix element of the type $\langle m|\varrho(k)|m'\rangle$. Inserting equation (B.15) in to (B.4) and (B.5), we obtain

$$\langle m|\varrho(k)|m'\rangle = \sum_{m_i}\langle m|H|m_i\rangle\langle m'|H_1|m_i\rangle^*$$

$$= \sum_{m_i}\sum_{LM\mu,L'M'\mu'}(-1)^{-2I+L-2m_i+L'}\begin{pmatrix} I & L & I_i \\ m' & M & -m_i \end{pmatrix}\begin{pmatrix} I & L' & I_i \\ m' & M' & -m_i \end{pmatrix}$$
$$\times\langle 0|L\mu\rangle\langle I||L||I_i\rangle\langle 0|L\mu\rangle^*\langle I||L'||I_i\rangle^* D^{L'}_{\mu'M'}(z\to k)D^{L}_{\mu M}(k\to z)$$

But,

$$D^{L'}_{\mu'M'}(z\to k)D^{L}_{\mu M}(k\to z) = \sum_k\langle L\mu L'\mu'|k\tau\rangle\langle ML'L'M'|kN\rangle D^{k}_{\tau N}(z\to k\rangle. \qquad \text{(B.16)}$$

and the Celbsch-Gordan coefficients for vector addition $j_1 + j_2 = j_3$, $m_1 + m_2 = m_3$ are defined by the transformation

$$|j_1 j_2 j_3 m_3\rangle = \sum_{m_1 m_2}|j_1 m_1 j_2 m_2\rangle\langle j_1 m_1 j_2 m_2|j_3 m_3\rangle$$

$$(-1)^{j_1-j_2-m_3}\langle j_1 m_1 j_2 m_2|j_3 - m_3\rangle = \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix}\sqrt{2j_3+1} \qquad \text{(B.17)}$$

$$\langle j_1 m_1 j_2 m_2 | j_3 m_3 \rangle = \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & -m_3 \end{pmatrix} (-1)^{-j_1+j_2-m_3}.$$

Therefore,

$$\langle m|\varrho(k)|m'\rangle = \sum_{m_i} \sum_{LM\mu} \sum_{KL\tau} (-1)^{M'-\mu'} \begin{pmatrix} I & L & I_i \\ m' & M & -m_i \end{pmatrix} \begin{pmatrix} I & L' & I_i \\ m' & M' & -m_i \end{pmatrix}$$
$$\times \begin{pmatrix} I & L' & k \\ \mu & -\mu' & \tau \end{pmatrix} \begin{pmatrix} L & L' & k \\ M & -M' & N \end{pmatrix} (2k+1)$$
$$\times \langle 0|L\mu\rangle \langle I||L||I_i\rangle \langle 0|L\mu'\rangle^* \langle I||L'||I_i\rangle D^k_{\tau N}(z \to k) \tag{B.18}$$

In the above expression $N = M' - M$, $\tau = \mu' - \mu$ and angle independent factor have again been omitted. Moreover, we have used the fact that $L' - L$, N, τ, $2I$ and $I + L + m_i$ are integers. The sum m_i, M and M' over the product of the first $3-j$ symbols of equation (B.18) can be evaluated by using Racah relation, together with the symmetry relation given below. The $3-j$ symbols have the property that an even permutation of the columns leaves the numerical unchanged [12].

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = \begin{pmatrix} j_2 & j_3 & j_1 \\ m_2 & m_3 & m_1 \end{pmatrix} = \begin{pmatrix} j_3 & j_1 & j_2 \\ m_3 & m_1 & m_2 \end{pmatrix} \tag{B.19}$$

While an odd permutation is equivalent to multiplication by $(-1)^{j_1+j_2+j_3}$,

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = (-1)^{j_1+j_2+j_3} \begin{pmatrix} j_2 & j_1 & j_3 \\ m_2 & m_1 & m_3 \end{pmatrix} = (-1)^{j_1+j_2+j_3} \begin{pmatrix} j_3 & j_2 & j_1 \\ m_3 & m_2 & m_1 \end{pmatrix}. \tag{B.20}$$

In equation (B.20) the terms of the bottom row has been replaced their negative values:

$$\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = (-1)^{j_1+j_2+j_3} \begin{pmatrix} j_1 & j_2 & j_3 \\ -m_1 & -m_2 & -m_3 \end{pmatrix} \tag{B.21}$$

On the other hand the Wigner $6-j$ symbols can be defined as

$$\sum(-1)^{j_4+j_5+j_6+m_4+m_5+m_6}\begin{pmatrix} j_1 & j_5 & j_6 \\ m_1 & m_5 & -m_6 \end{pmatrix}\begin{pmatrix} j_4 & j_2 & j_6 \\ -m_4 & m_2 & m_6 \end{pmatrix}\begin{pmatrix} j_4 & j_5 & j_3 \\ m_4 & -m_5 & m_3 \end{pmatrix}$$

$$=\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix}\begin{Bmatrix} j_1 & j_2 & j_3 \\ j_4 & j_5 & j_6 \end{Bmatrix} \tag{B.22}$$

The term within $\{\}$ is the Wigner $6-j$ symbol and it is independent of magnetic quantum number m. Using equation (B.21) and (B.22) in equation (B.18) gives

$$\langle m|\varrho(k)|m'\rangle = \sum_{LM\mu L'}\sum_{KL\tau}(-1)^{2I-I_i+m-\mu}\begin{pmatrix} L & L' & k \\ \mu & \mu' & \tau \end{pmatrix}\begin{pmatrix} I & I & k \\ m' & -m & N \end{pmatrix}$$

$$\times\begin{Bmatrix} I & I & k \\ L & L' & I_i \end{Bmatrix}(2k+1)$$

$$\times\langle 0|L\mu\rangle\langle I||L||I_i\rangle\langle 0|L'\mu'\rangle^*\langle I||L'||I_i\rangle D^k_{\tau N}(z\rightarrow k) \tag{B.23}$$

The expression (B.23) contains factors that depend only on the properties of the radiation. To single out these factors clearly, we introduce with Racah radiation parameters $C_{k\tau}$, defined by [11]

$$C_{k\tau}(LL') = \sum_{\mu\mu'}(-1)^{L'-\mu'}\langle L\mu L'-\mu'|k\tau\rangle\langle 0|L\mu\rangle^*\langle 0|L'\mu'\rangle$$

$$=\sum_{\mu\mu'}(-1)^{L-\mu}\sqrt{2k+1}\begin{pmatrix} L & L' & k \\ \mu & \mu' & \tau \end{pmatrix}\langle 0|L\mu\rangle^*\langle 0|L'\mu'\rangle \tag{B.24}$$

Inserting equation (B.24) into (B.23)

$$\langle m|\varrho(k)|m'\rangle = \sum_{LL'}\sum_{kN\tau}(-1)^{2I-I_i+m-L'}\sqrt{2k+1}\begin{pmatrix} I & I & k \\ m' & -m & N \end{pmatrix}\begin{Bmatrix} I & I & k \\ L & L' & I_i \end{Bmatrix}$$

$$\times C_{k\tau}(LL')\langle I||L||I_i\rangle\langle I||L'||I_i\rangle D^k_{\tau N}(z\rightarrow k) \tag{B.25}$$

The matrix element $\langle m|\varrho(k_1)|m'\rangle$ is given by the expansion of equation (B.25) with the proper subscripts. For the matrix element $\langle m'|\varrho(k_2)|m\rangle$, one finds

$$\langle m'|\varrho(k_2)|m\rangle = \sum_{L_2L_2'}\sum_{k_2N_2\tau_2}(-1)^{k_2-I_f-m-L_2'}\sqrt{2k_2+1}\begin{pmatrix} I & I & k_2 \\ m' & -m & N_2\end{pmatrix}\begin{Bmatrix} I & I & k_2 \\ L_2 & L_2' & I_f\end{Bmatrix}$$
$$\times C^*_{k\tau}(LL')\langle I_f||L_2||I\rangle\langle I_f||L_2'||I\rangle^* D^{k_2*}_{N_2\tau_2}(z\rightarrow k) \qquad \text{(B.26)}$$

Let us define the coefficient A_{kk} (the anisotropic coefficient which depends on radiation pattern)

$$A_{kk} = A_k(L_1L_1'I_iI)A_k(L_2L_2'I_fI)$$

where

$$A_k(L_1L_1'I_iI) = \sum_{L_1L_1'}(-1)^{L_1}C^*_{k0}(L_1L_1')\begin{Bmatrix} I & I & k \\ L_1 & L_1' & I_i\end{Bmatrix}\langle I||L_1||I_i\rangle\langle I||L_1'||I_i\rangle \qquad \text{(B.27)}$$

and

$$A_k(L_2L_2'I_fI) = \sum_{L_2L_2'}(-1)^{L_2}C^*_{k0}(L_2L_2')\begin{Bmatrix} I & I & k \\ L_2 & L_2' & I_f\end{Bmatrix}\langle I||L_2||I_f\rangle\langle I||L_2'||I_f\rangle \qquad \text{(B.28)}$$

with

$$C^*_{k0}(LL') = C_{k0}(LL'). \qquad \text{(B.29)}$$

Using some of the expression that we have discused up to now, we can derive a formula for time dependent angular correlation function. Starting from

$$W(k_1,k_2,t) = \sum_{m_am_a'm_bm_b'}\langle m_f|H_2|m_b\rangle\langle m_b|\Lambda(t)|m_a\rangle\langle m_a|H_1|m_i\rangle\times$$
$$\langle m_f|H_2|m_b'\rangle^*\langle m_b'|\Lambda(t)|m_a\rangle^*\langle m_a'|H_1|m_i\rangle^* \qquad \text{(B.30)}$$

we introduce the density matrices by equation (B.4) and equation (B.5) above. writing the correlation function as:

$$W(k_1,k_2,t) = \sum_{m_a m_a' m_b m_b'} \langle m_a|\varrho(k_1)|m_a'\rangle\langle m_b'|\varrho(k_2)|m_b\rangle \times$$
$$\langle m_b|\Lambda(t)|m_a\rangle\langle m_b'|\Lambda(t)|m_a'\rangle^* \tag{B.31}$$

with $\tau_1 = \tau_2 = 0$, equation (B.9) or equation (B.12) can be used to express the D^k_{N0} as spherical harmonics $Y^N_k(\theta,\phi)$. Inserting the expansion (B.25) and (B.26) for the density matrices into equation (B.30) or equation (B.31) and using the definition (B.27) and (B.28) of the coefficient A_k gives

$$W(k_1,k_2,t) = \sum_{m_a m_a' m_b m_b'} \sum_{L_1 L_1'} \sum_{k_1 N_1 0} (-1)^{2I-I_i+m_a-L_1'} \sqrt{2k_1+1} \begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1 \end{pmatrix}$$
$$\times \begin{Bmatrix} I & I & k_1 \\ L_1 & L_1' & I_i \end{Bmatrix} C_{k0}(L_1 L_1')\langle I||L_1||I_i\rangle\langle I||L_1'||I_i\rangle^*$$
$$\times \sum_{L_2 L_2'} \sum_{k_2 N_2 0} (-1)^{k_2-I_f+m_b-L_2'} \sqrt{2k_2+1} \begin{pmatrix} I & I & k_2 \\ m_b' & -m_b & N_2 \end{pmatrix} \begin{Bmatrix} I & I & k_2 \\ L_2 & L_2' & I_f \end{Bmatrix}$$
$$\times C_{k0}(L_2 L_2')\langle I_f||L_2||I\rangle(I_f||L_2'||I\rangle^* D^{k_1}_{N_1 0}(z\to k_1) D^{k_2 *}_{N_2 0}(z\to k_2)\langle m_b|\Lambda(t)|m_a\rangle\langle m_b'|\Lambda(t)|m_a'\rangle^*$$
$$= \sum_{m_a m_a' m_b m_b'} \sum_{k_1 k_2 N_1 N_2} \sqrt{(2k_1+1)(2k_2+1)} A_{k_1}(1) A_{k_2}(2) \times \begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1 \end{pmatrix}$$
$$\begin{pmatrix} I & I & k_2 \\ m_b' & -m_b & N_2 \end{pmatrix} \langle m_b|\Lambda(t)|m_a\rangle\langle m_b'|\Lambda(t)|m_a'\rangle^* D^{k_1}_{N_1 0}(z\to k_1) D^{k_2 *}_{N_2 0}(z\to k)$$

where the definition of $A_{k_1}(1)$ and $A_{k_2}(2)$ in equation (B.27) and (B.28) have been used. Moreover from equation (B.9) or B.12)

$$D^{k_1}_{N_1 0} = \sqrt{\frac{4\pi}{2k_1+1}} Y^{N_1}_{k_1}(\theta_1,\phi_1)$$

$$D^{k_2}_{N_2 0} = \sqrt{\frac{4\pi}{2k_2+1}} Y^{N_2 *}_{k_2}(\theta_2,\phi_2)$$

$$W(k_1,k_2,t) = \sum_{m_a m_a' m_b m_b'} \sum_{k_1,k_2 N_1 N_2} \sqrt{(2k_1+1)(2k_2+1)} A_{k_1}(1) A_{k_2}(2)$$
$$\times \frac{4\pi}{\sqrt{(2k_1+1)(2k_2+1)}} \begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m_b' & -m_b & N_2 \end{pmatrix}$$
$$\times \langle m_b|\Lambda(t)|m_a\rangle \langle m_b'|\Lambda(t)|m_a'\rangle^* Y_{k_1}^{N_1}(\theta_1,\phi_1) Y_{k_2}^{N_2 *}(\theta_2,\phi_2)$$

$$W(k_1,k_2,t) = 4\pi \sum_{k_1,k_2 N_1 N_2} A_{k_1}(1) A_{k_2}(2) G_{k_1 k_2}^{N_1 N_2}(t) [(2k_1+1)(2k_2+1)]^{-1/2}$$
$$\times Y_{k_1}^{N_1}(\theta_1,\phi_1) Y_{k_2}^{N_2 *}(\theta_2,\phi_2) \tag{B.32}$$

$$= \sum_{k_1,k_2 N_1 N_2} A_{k_1}(1) A_{k_2}(2) G_{k_1 k_2}^{N_1 N_2}(t) P_k(\cos\theta) \tag{B.33}$$

where θ is the angle between k_1 and k_2.

In equation (B.32), we now replace $A_{k_1}(1)$ by $A_{k_1}(\gamma_1)$ and $A_{k_2}(2)$ by $A_{k_2}(\gamma_2)$. Finally, we obtain the required perturbation function as

$$W(k_1,k_2,t) = \sum_{k_1,k_2 N_1 N_2} A_{k_1}(\gamma_1) A_{k_2}(\gamma_2) G_{k_1 k_2}^{N_1 N_2}(t) [(2k_1+1)(2k_2+1)]^{-1/2}$$
$$\times Y_{k_1}^{N_1}(\theta_1,\phi_1) Y_{k_2}^{N_2 *}(\theta_2,\phi_2) \tag{B.34}$$

where we define the perturbation coefficient $G_{k_1 k_2}^{N_1 N_2}(t)$ as

$$G_{k_1 k_2}^{N_1 N_2}(t) = \sum_{m_a m_b} [(2k_1+1)(2k_2+1)]^{1/2} \langle m_b|\Lambda(t)|m_a\rangle \langle m_b'|\Lambda(t)|m_a'\rangle^*$$
$$\times \begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m_b' & -m_b & N_2 \end{pmatrix}. \tag{B.35}$$

Appendix C

Anisotropic Coefficients

Calculation of S_{kn} for $k = 2$ and spin $I = 5/2$

The Wigner 3-j vectors can be solved using equation (A.1), (A.2) and (A.3). Therefore, using these equations

$$\begin{pmatrix} 5/2 & 5/2 & 2 \\ m' & -m & m-m' \end{pmatrix} = A \times B \times C$$

where

$$A = [\frac{(j_1+j_2-j_3)!(j_1+-j_2+j_3)!(-j_1+j_2+j_3)!}{(j_1+j_2+j_3+1)!}]^{\frac{1}{2}}$$

$$B = [(j_1+m_1)!(j_1-m_1)!(j_2+m_2)!(j_2-m_2)!(j_3+m_3)!(j_3-m_3)!]^{\frac{1}{2}}$$

$$C = \sum_{\nu} \frac{(-1)^{\nu}}{\nu!(j_1+j_2-j_3-\nu)!(j_1-m_1-\nu)!(j_2-m_2-\nu)!} \times \frac{1}{(j_3-j_2+m_1+\nu)!(j_3-j_1-m_2+\nu)!}$$

Therefore,

$$A = \left[\frac{2! \times 3! \times 2!}{8!}\right]^{1/2} = \frac{2}{\sqrt{6720}}$$

The possible combinations of S_{20} is

m	1/2	-1/2	3/2	-3/2	5/2	-5/2	1/2	-1/2
m'	1/2	-1/2	3/2	-3/2	5/2	-5/2	-1/2	1/2

.

The other combinations like m = 3/2 and m' = -3/2 is not possible because it requires values greater than value of k. If we have a vector in the form $\begin{pmatrix} I & I & 2 \\ m & -m & 0 \end{pmatrix}$ the result can be written as

$$\begin{pmatrix} I & I & 2 \\ m & -m & 0 \end{pmatrix} = \frac{2[3m^2 - I(I+1)]}{\sqrt{(2I+3)(2I+2)(2I+1)(2I)(2I-1)}}.$$

For n = 0, m = 1/2 and m' = 1/2,

$$\begin{pmatrix} 5/2 & 5/2 & 2 \\ 1/2 & -1/2 & 0 \end{pmatrix}^2 = \left[\frac{2[(3\times 1/2)^2 - 5/2(5/2+1)]}{\sqrt{8\times 7\times 6\times 5\times 4}}\right]^2.$$

$$= 0.038095$$

For n = 0, m = -1/2 and m' = -1/2,

$$\begin{pmatrix} 5/2 & 5/2 & 2 \\ -1/2 & 1/2 & 0 \end{pmatrix}^2 = \left[\frac{2[(3\times -1/2)^2 - 5/2(5/2+1)]}{\sqrt{8\times 7\times 6\times 5\times 4}}\right]^2 = 0.038095.$$

For n = 0, m = 3/2 and m' = 3/2

$$\begin{pmatrix} 5/2 & 5/2 & 2 \\ 3/2 & -3/2 & 0 \end{pmatrix}^2 = \left[\frac{2[(3\times 3/2)^2 - 5/2(5/2+1)]}{\sqrt{8\times 7\times 6\times 5\times 4}}\right]^2 = 0.002381.$$

For n = 0, m = -3/2 and m' = -3/2

$$\begin{pmatrix} 5/2 & 5/2 & 2 \\ -3/2 & 3/2 & 0 \end{pmatrix}^2 = \left[\frac{2[(3\times -3/2)^2 - 5/2(5/2+1)]}{\sqrt{8\times 7\times 6\times 5\times 4}}\right]^2 = 0.002381.$$

For n = 0, m = 5/2 and m' = 5/2

$$\begin{pmatrix} 5/2 & 5/2 & 2 \\ 5/2 & -5/2 & 0 \end{pmatrix}^2 = \left[\frac{2[(3\times 5/2)^2 - 5/2(5/2+1)]}{\sqrt{8\times 7\times 6\times 5\times 4}}\right]^2 = 0.059524.$$

For n = 0, m = -5/2 and m' = -5/2

$$\begin{pmatrix} 5/2 & 5/2 & 2 \\ -5/2 & 5/2 & 0 \end{pmatrix}^2 = \left[\frac{2[(3\times -5/2)^2 - 5/2(5/2+1)]}{\sqrt{8\times 7\times 6\times 5\times 4}}\right]^2 = 0.059524.$$

For n = 0, m = 1/2 and m' = -1/2

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -1/2 & 1 & -1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{7720}} \times B \times C\right]^2.$$

$$B = \sqrt{2!\times 3!\times 3!\times 1!\times 3!\times 2!} = 12\sqrt{6}$$

$$C = \sum \frac{(-1)^\nu}{\nu!(2-\nu)!(3-\nu)!\nu!(\nu-1)!}$$

$\nu = 1, 2$

$$C = \frac{-1}{1!.1!.2!.2!.1!.0!} + \frac{1}{2!.0!.1!.1!.2!.1!}$$

$$C = \frac{-1}{4} + \frac{1}{4} = 0$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -1/2 & 1 & -1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{7720}} \times 12\sqrt{6} \times 0\right]^2 = 0$$

For n = 0, m = -1/2 and m' = 1/2, by symmetry

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -1/2 & 1 & -1/2 \end{pmatrix}^2 = 0$$

Therefore,

$$S_{20} = \sum_{m,m'} \begin{pmatrix} I & I & k \\ m' & -m & -m'+m \end{pmatrix}^2$$

$$= 2(0.038095 + 0.002381 + 0.059524) = 0.2000$$

The possible combination of S_{21} is,

m	1/2	3/2	-1/2	1/2	3/2	-3/2	-1/2	-3/2
m'	-3/2	-1/2	3/2	3/2	1/2	-1/2	-3/2	1/2

Therefore, for n = 1, m = 1/2 and m' = -3/2,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -1/2 & 2 & -3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[2! \times 3! \times 4! \times 0 \times 4! \times 1!\right]^{1/2} = 48\sqrt{3}$$

$$C = \sum \frac{(-1)^{\nu}}{\nu!(2-\nu)!(3-\nu)!(4-\nu)!(\nu)!(\nu-2)!}$$

$\nu = 2$ only for this case

$$C = \frac{1}{2!.0!.1!.2!.2!.0!} = \frac{1}{8}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -1/2 & 2 & -3/2 \end{pmatrix}^2 = \left(\frac{2}{\sqrt{6720}} 48\sqrt{3} \times \frac{1}{8}\right)^2 = 0.064285$$

For n =1, m = -3/2 and $m' = 1/2$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 3/2 & -2 & 1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[4!.1!.0!.4!.3!.2!\right]^{1/2} = 48\sqrt{3}$$

$$C = \sum \frac{(-1)^{\nu}}{\nu!(2-\nu)!(1-\nu)!(-\nu)!(2+\nu)!(2+\nu)!}$$

$\nu = 0$ is only possible

$$C = \frac{1}{0!.2!.1!.0!.2!.2!} = \frac{1}{8}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 3/2 & -2 & 1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{48\sqrt{3}}{8} = 0.064285\right.$$

For n = 1, m = 3/2 and m' = -1/2,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -3/2 & 2 & -1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[1!.4!.4!.0!.2!.3!\right]^{1/2} = 48\sqrt{3}$$

$$C = \sum \frac{(-1)^{\nu}}{\nu!(2-\nu)!(4-\nu)!(4-\nu)!(\nu-1)!(\nu-2)!}$$

$\nu = 2$ only,

$$C = \frac{1}{2!.0!.2!.2!.1!.0!} = \frac{1}{8}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -3/2 & -2 & -1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{48\sqrt{3}}{8}\right]^2 = 0.064285$$

For n = 1, m = -1/2 and $m' = 3/2$,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 1/2 & -2 & 3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[3!.2!.0!.4!.4!.1!\right]^{1/2} = 48\sqrt{3}$$

$$C = \sum \frac{(-1)^\nu}{\nu!(2-\nu)!(2-\nu)!(-\nu)!(1+\nu)!(2+\nu)!}$$

ν can be only 0,

$$C = \frac{1}{0!.2!.2!.0!.1!.2!} = \frac{1}{8}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 1/2 & -2 & 3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{48\sqrt{3}}{8}\right]^2 = 0.064285.$$

For n = 1, m = 1/2 and $m' = 3/2$,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -1/2 & -1 & 3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[2!.3!.3!.1!.4!.1!\right]^{1/2} = 24\sqrt{3}$$

$$C = \sum \frac{(-1)^\nu}{\nu!(2-\nu)!(3-\nu)!(1-\nu)!(\nu)!(1+\nu)!}$$

$\nu = 0, 1$

$$C = \frac{1}{0!.2!.3!.1!.0!.1!} + \frac{-1}{1!.1!.2!.0!.1!.2!} = \frac{1}{12} - \frac{1}{4} = \frac{-1}{6}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -1/2 & -1 & 3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{(-24\sqrt{3})}{6}\right]^2 = 0.028571$$

For n = 1, m = 3/2 and $m' = 1/2$,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -3/2 & 1 & 1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[1!.4!.3!.1!.3!.2!\right]^{1/2} = 24\sqrt{3}$$

$$C = \sum \frac{(-1)^{\nu}}{\nu!(2-\nu)!(4-\nu)!(3-\nu)!(\nu-1)!(\nu-1)!}$$

$\nu = 1, 2$ only

$$C = \frac{-1}{1!.1!.3!.2!.0!.0!} + \frac{1}{2!.0!.2!.1!.1!.1!} = \frac{-1}{12} + \frac{1}{4} = \frac{1}{6}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -3/2 & 1 & 1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{24\sqrt{3}}{4}\right]^2 = 0.028571$$

For n = 1, m = -3/2 and m' = -1/2,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 3/2 & -1 & -1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[4!.1!.1!.3!.2!.3!\right]^{1/2} = 24\sqrt{3}$$

$$C = \sum \frac{(-1)^{\nu}}{\nu!(2-\nu)!(1-\nu)!(1-\nu)!(\nu+2)!(\nu+1)!}$$

$\nu = 0$ and 1

$$C = \frac{1}{0!.2!.1!.1!.2!.1!} + \frac{-1}{1!.1!.0!.0!.3!.2!} = \frac{1}{4} + \frac{-1}{12} = \frac{1}{4} - \frac{1}{12} = \frac{1}{6}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 3/2 & -1 & -1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{24\sqrt{3}}{6}\right]^2 = 0.028571$$

For n = 1, m = -1/2 and m' = -3/2,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 1/2 & 1 & -3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[3!.2!.3!.1!.4!.1!\right]^{1/2} = 24\sqrt{3}$$

$$C = \sum \frac{(-1)^{\nu}}{\nu!(2-\nu)!(2-\nu)!(3-\nu)!(\nu+1)!(\nu-1)!}$$

$\nu = 1, 2$

$$C = \frac{-1}{1!.1!.1!2!.2!.0!} + \frac{1}{2!.0!.0!.1!.3!.1!} = \frac{-1}{4} + \frac{1}{12} = \frac{-1}{6}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 1/2 & 1 & -3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{(-24\sqrt{3})}{6}\right]^2 = 0.028571$$

Therefore,

$$S_{21} = \sum_{m,m'} \begin{pmatrix} I & I & k \\ m' & -m & -m'+m \end{pmatrix}^2$$

$$S_{21} = 4(0.028571 + 0.064285) = 0.371$$

To obtain n = 2, we have four possible combinations. These combinations

m	5/2	-5/2	-3/2	3/2
m'	5/2	3/2	-5/2	-3/2

Even though other combinations like those given in the following table give n = 2, because of the fact that $-m'+m = 4$ or -4 which are not allowed for k = 2, we cannot use them.

m	-3/2	-5/2	3/2	5/2
m'	5/2	3/2	-5/2	-3/2

For n = 2, m = 5/2 and $m' = 3/2$,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -5/2 & 1 & 3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[0!.5!.3!.1!.4!.1!\right]^{1/2} = 24\sqrt{30}$$

$$C = \sum \frac{(-1)^\nu}{\nu!(2-\nu)!(5-\nu)!(3-\nu)!(\nu-2)!(\nu-1)!}$$

$\nu = 2$ only

$$C = \frac{1}{2!.0!.3!.1!.0!.1!} = \frac{1}{12}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -5/2 & 1 & 3/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{24\sqrt{30}}{12}\right]^2 = 0.071429$$

For n = 2, m = 3/2 and $m' = 5/2$ by symmetry,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -3/2 & -1 & 5/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{(-24\sqrt{30})}{12}\right]^2 = 0.071429$$

For n = 2, m = -3/2 and m' = -5/2,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 3/2 & 1 & -5/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[4!.1!.3!.1!.0!.5!\right]^{1/2} = 24\sqrt{30}$$

$$C = \sum \frac{(-1)^\nu}{\nu!(2-\nu)!(1-\nu)!(3-\nu)!(\nu+2)!(\nu-1)!}$$

$\nu = 1$ only

$$C = \frac{-1}{1!.1!.0!.2!.3!.0!} = \frac{-1}{12}$$

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 3/2 & 1 & -5/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{(-24\sqrt{30})}{12}\right]^2 = 0.071429$$

For n = 2, m = -5/2 and m' = -3/2 by symmetry

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ 3/2 & 1 & -5/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times \frac{(24\sqrt{30})}{12}\right]^2 = 0.071429$$

Therefore,

$$S_{22} = \sum_{m,m'} \begin{pmatrix} I & I & k \\ m' & -m & -m'+m \end{pmatrix}^2$$

$$= 4 \times 0.071429 = 0.2857$$

Let us now calculate S_{23}. Four possible combinations that give n = 3 is given in the table.

m	5/2	1/2	-1/2	-5/2
m'	1/2	5/2	-5/2	-1/2

and the remaining four combinations are not possible for the reason discussed in the previous case. For n = 3, m = 5/2 and m' = 1/2,

$$\begin{pmatrix} 5/2 & 2 & 5/2 \\ -5/2 & 2 & 1/2 \end{pmatrix}^2 = \left[\frac{2}{\sqrt{6720}} \times B \times C\right]^2$$

$$B = \left[0!.5!.4!.0!.2!.3!\right]^{1/2} = 48\sqrt{15}$$

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