CHAPTER III

INSTRUMENTATION AND METHODS OF ANALYSIS

3.1 The X-ray and Gamma ray Intensity Measurements

The information regarding the levels of daughter nuclides can be extracted by precise energy and intensity measurements of gamma rays emitted by various radionuclides. Also, for a better understanding of the atomic processes following radioactive decay, there is a need for the measurement of intensities of X-rays emitted by daughter nuclides preceding internal conversion and electron capture processes. Precision measurements of X-ray and gamma ray intensities are also useful for the investigation of the nuclear fuel burn-up rate, nuclear fission cross-sections, activation analysis and mesic X-ray spectroscopy.

The relative intensity of gamma rays from the radioactive decays is usually measured with an accuracy of 2-5%. However, some workers\(^1\)\(^2\)\(^3\)\(^4\)\(^5\)\(^6\)\(^7\) have recently performed precision intensity measurements with errors better than 1% for gamma rays above 100 keV in various nuclides. Apart from this only a few measurements of K and L X-rays from radioactive decays using high resolution detectors are available in literature\(^5\)\(^6\)\(^8\).

The purpose of our experiments was to determine intensities for X-rays and gamma rays from radioactive decays in the energy region 5-1400 keV. To cover this energy region, we have used four detector set-ups (i) a 64.1 cm\(^3\) and a 95.0 cm\(^3\) coaxial HPGe detector (ii) a 28.27 mm\(^2\) x 5.0 mm vertical planar HPGe detector.
A brief discussion of different detector set-ups and compatible electronics (Fig. 3.1) used for singles intensity measurements is described below:

3.1.1 Photon Detector Set-ups

a) The coaxial HPGe detector set-up

A 64.1 cm$^3$ and a 96.0 cm$^3$ coaxial HPGe detectors of GEM series manufactured by EG & G ORTEC, U.S.A., were used for the gamma ray energy and intensity measurements in the energy region 80-1400 keV. The detector element was made of a p type germanium crystal with a lithium diffused outer contact (thickness 500-1000 micron) and inner contact is ion implanted (thickness < 0.3 micron). It was mounted in the cryostat and operated close to liquid nitrogen temperature. The specifications of these detectors along-with other coaxial semiconductor detectors used for directional correlation/Coulomb excitation studies, are given in Table 3.1. The detectors were shielded using lead cones and cylinders (wall thickness 2 cm) having inside lining of graded Compton shield of tin and copper to reduce the background radiations. For gamma ray intensity measurements, the source-to-detector distance was kept at 25 cm, by fixing the source on the axial line of the detector on a separate mount.

b) The vertical HPGe detector set-up

A small 28.27 mm$^2$ x 5.0 mm vertical HPGe detector of GLP series manufactured by EG & G ORTEC, USA, was used for K X-ray and
Fig. 3.1 GENERAL BLOCK DIAGRAM OF A PHOTON SPECTROMETER
TABLE 3.1
Specifications of Coaxial HPGe detectors used for gamma ray intensity and gamma-gamma directional correlation measurements.

<table>
<thead>
<tr>
<th>Item</th>
<th>Detector</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>64.1 cm³</td>
</tr>
<tr>
<td>Type</td>
<td>Coaxial</td>
</tr>
<tr>
<td>Crystal diameter</td>
<td>47.0 mm</td>
</tr>
<tr>
<td>Crystal length</td>
<td>37.1 mm</td>
</tr>
<tr>
<td>Inactive germanium</td>
<td>0.7 mm</td>
</tr>
<tr>
<td>Aluminium case</td>
<td>1.25 mm</td>
</tr>
<tr>
<td>Bias Voltage</td>
<td>3500 V, pos.</td>
</tr>
<tr>
<td>fwhm at 1332 keV</td>
<td>1.80 keV</td>
</tr>
<tr>
<td>fwhm at 122 keV</td>
<td>1000 eV</td>
</tr>
<tr>
<td>Peak/Compton ratio for 1332 keV Y-rays.</td>
<td>48</td>
</tr>
<tr>
<td>Amplifier shaping time (ORTEC 572)</td>
<td>6 μs</td>
</tr>
<tr>
<td>High voltage filter (ORTEC)</td>
<td>Model 138</td>
</tr>
<tr>
<td>Preamplifier (ORTEC)</td>
<td>Model 137CP2</td>
</tr>
<tr>
<td>Output pulse</td>
<td>pos.</td>
</tr>
<tr>
<td>Conversion gain (Ge)</td>
<td>400 mV/MeV</td>
</tr>
<tr>
<td>Maximum energy rate</td>
<td>180,000 MeV/s</td>
</tr>
<tr>
<td></td>
<td>(2000 MΩ feedback resistance)</td>
</tr>
</tbody>
</table>
gamma ray intensity measurements in the energy region 20-400 keV. The detector element (p type germanium crystal) was having a planar configuration with a thin, ion implanted front contact (thickness < 0.3 micron). It was mounted in the cryostat and operated close to liquid nitrogen temperature. The cryostat end cap was having a beryllium window and thickness 0.127 mm to maximise the transmission of low energy photons to the detector element. This detector cannot be used on intensity measurements below 15 keV because of a strong discontinuity at 11.1 keV due to K-absorption edge of germanium. The specifications of the detector are given in Table 3.2. For photon intensity measurements, the source-to-detector distance was kept 4 cm by placing the source on a perspex cup mounted on the detector.

c) The Si(Li) detector set-up

The X-rays and gamma rays intensity measurements in the energy region 4-90 keV, were performed by using a 28.27 mm² x 5.5 mm vertical planar Si(Li) detector (SLP series, EG & G ORTEC, USA). The detector element was lithium-drifted silicon crystal with ion implanted front contact (thickness < 0.3 micron). It was mounted in a cryostat and operated close to liquid nitrogen temperature. The detector was having beryllium window (thickness = 0.025 mm) to maximise its low energy efficiency. The specification of the Si(Li) detector are given in Table 3.2.

A special annular source geometry, generally used for X-ray fluorescence experiments, was mounted on Si(Li) detector. It consisted of an annular exciter source of $^{109}$Cd (15 mCi),
<table>
<thead>
<tr>
<th>Item</th>
<th>HPGe detector</th>
<th>Si(Li) detector</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type</td>
<td>Vertical planar</td>
<td>Vertical planar</td>
</tr>
<tr>
<td>Crystal diameter</td>
<td>6.0 mm</td>
<td>6.0 mm</td>
</tr>
<tr>
<td>Crystal thickness</td>
<td>5.0 mm</td>
<td>5.5 mm</td>
</tr>
<tr>
<td>Beryllium window thickness</td>
<td>0.127 mm</td>
<td>0.025 mm</td>
</tr>
<tr>
<td>Bias Voltage</td>
<td>1000 V, Neg.</td>
<td>1500 V, Neg.</td>
</tr>
<tr>
<td>fwhm at 5.9 keV</td>
<td>165 eV</td>
<td>165 eV</td>
</tr>
<tr>
<td>fwhm at 122 keV</td>
<td>459 eV</td>
<td>-</td>
</tr>
<tr>
<td>Amplifier shaping time</td>
<td>6 μs</td>
<td>10 μs</td>
</tr>
<tr>
<td>High Voltage Filter</td>
<td>Model 138 L</td>
<td>Model 138 L</td>
</tr>
<tr>
<td>Preamplifier (ORTEC)</td>
<td>Model 139</td>
<td>Model 139</td>
</tr>
<tr>
<td>Output pulse</td>
<td>Neg.</td>
<td>Neg.</td>
</tr>
<tr>
<td>Conversion gain(Ge)</td>
<td>1.7 mV/keV</td>
<td>1.7 mV/keV.</td>
</tr>
<tr>
<td>Maximum Energy Rate</td>
<td>2500 MeV/s</td>
<td>2500 MeV/s</td>
</tr>
<tr>
<td>(1x10^{11} feedback resistor)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
tungsten spacer and shield packed in an aluminium casing. Fig. 3.2 shows the annular source geometry for the present measurements. The tungsten alloy shield avoids the direct radiation exposure of the detector from $^{109}\text{Cd}$ exciter source. There is a perspex holder to mount a target for X-ray fluorescence studies. The main purpose of using this set-up was to use the knowledge of X-ray fluorescence parameters for efficiency calibration of the Si(Li) detector (section 3.1.4c).

For the intensity measurements, the uncovered radioactive source was placed at the target position (Fig. 3.2) with $^{109}\text{Cd}$ exciter source removed. A lead collimator of 5.6 mm dia with Cu and Al lining on the inside was used to collimate the photon beam to the detector. This reduced the scattering contribution at the low energy side of the peaks and consequently improves the true-to-background ratio.

3.1.2 Associated Electronics

a) Preamplifier and high voltage filter

The absorption of a photon by the detector element produces a current pulse at the preamplifier input and is integrated by the charge sensitive loop. It consists of an OPAMP (with liquid nitrogen cooled FET at input stage) with capacitive feedback. The pole zero cancellation network differentiates the charge loop output to give shorter decay time constant and was adjusted to eliminate any undershoots. For most of spectroscopy applications, the above mentioned charge sensitive preamplifiers are used because of their gain being
Fig. 3.2 SOURCE TO DETECTOR GEOMETRY SET-UP USED WITH Si(Li) DETECTOR
insensitive to stray capacitances (capacitance of the detector, cables etc.) and provide large output pulse heights. The detector bias is applied through a high voltage filter network to remove power supply ripple and interference picked up on the cable between the preamplifier and high voltage supply.

For the present work, the preamplifiers and high voltage filters used with different detectors are given in Tables 3.1 and 3.2. The detector element, preamplifier and high voltage filter were incorporated into a cylinder less than three inches in diameter. The d.c. power to the preamplifiers was applied from the rear panel of the spectroscopy amplifier (ORTEC model 572). Output pulses from preamplifiers used with different detectors (section 3.1.1) were having rise time < 200 ns and exponential fall with \( \sim 50 \, \mu s \) time constant. The output of each preamplifier is series terminated in 93 \( \Omega \) and is suitable for driving the 93 \( \Omega \) (RG62U) coaxial cable used for taking the output pulse to spectroscopy amplifier. The change in gain of the preamplifier with the change in temperature from 0° to 50°C is < 50 ppm/°C.

b) **Spectroscopy amplifier**

The outputs from the preamplifiers of semiconductor (HPGe/Si(Li)) detectors were amplified by a versatile spectroscopy amplifier (ORTEC model 572). It has an input impedance of approximately 1000 \( \Omega \) and accepts either positive or negative pulses with rise time < 650 ns and fall time > 40 \( \mu s \). Its gain is continuously variable from 1 to 1500. The shaping time constant on the amplifier is switch selectable in steps of 0.5, 1, 2, 3, 6 and 10 \( \mu s \). The choice of proper shaping time
constant is generally a compromise between operating at a shorter time constant for accommodation of high count rates (> 3000 counts/s) and operating with a large time constant for a better signal to noise ratio. At low count rates (< 1500 counts/s) used for photon intensity measurements, the optimum value for shaping time is 6 $\mu$s for HPGe detectors and 10 $\mu$s for Si(Li) detector.

The spectroscopy amplifier is having a variable pole zero cancellation network, which can be adjusted to match preamplifiers with decay time >40 $\mu$s. This drastically reduces the undershoot after the first differentiator and improves the overload and count rate characteristics. To compensate for the effect of baseline shift on a unipolar signal, caused by uncanceled RC interstages in the amplifier, base line restoration is used. The dc level of the output is adjustable up to ±100 mV. It provides unipolar and bipolar amplified output pulses with an output impedance of less than 1 $\Omega$.

c) Detector-bias supply

The ORTEC model 459 (0-5 kV, 0-100 $\mu$A) detector bias supply provides a bias voltage of either polarity for semiconductor detectors. The noise and ripple of this power supply is less than 10 mV peak-to-peak from 2Hz - 50 MHz. The temperature stability is 0.02% per °C through the operating range 0 to 50°C. Available voltage stability of this supply is <0.1% per hour in output voltage with constant voltage from bin supply, constant temperature and constant load.

d) Bin and Power Supply

Various ORTEC modular nuclear instruments used in the
e) Multichannel analyser (MCA) system

The positive unipolar pulses from the spectroscopy amplifier (ORTEC 572) are fed to a microcomputer based multichannel analyser ND-66B for pulse height analysis. The ND-66B combines the operational conversion of a conventional MCA with the computational power of an LSI-11/2 microcomputer. An analog-to-digital converter (ADC) ND 575 is used to digitise the analog pulses from spectroscopy amplifier. The ND 575 has input impedance of 1600Ω and accepts positive unipolar or initially positive bipolar pulses (0–8V). Since the 1Ω output of the spectroscopy amplifier (ORTEC model 572) was fed to the ADC through a 93Ω cable, the ADC was shunt terminated by 100Ω resistance at its input. The ADC ND575 is having conversion
gains 256, 512, 1024, 2048, 4096 or 8192 channels full scale and conversion rate of 80 MHz on all conversion gains. The conversion time per event is 2.5-9.0 μs at different conversion gains. The input pulses through the lower and upper level discriminator of ADC could be selected with coincident, anti-coincident or no gating requirements.

The MCA was having sufficient memory to store 8K, 24 bit data channels. The memory upto 8K channels could be assigned to ADC (ND 575) and it could contain upto 128 groups of memory storage. The ND-65BMCA performs a number of functions like data acquisition and storage, dead time correction, display manipulation, linear and quadratic energy calibration, spectrum stripping, smoothing etc. The stored spectrum data could be transferred to either on the Pertec model 7 x 40 magnetic tape or listed out onto the AT & T teleprinter. The 4K-channel memory of MCA could also be used for programming in Basic language.

3.1.3 Source Preparation

For the present intensity measurements, the radioisotopes were procured in the solution from Bhabha Atomic Research Centre, Bombay, Trombay, India. For intensity measurements, above 80 keV using Coaxial HPGe detector, the radioactive sources were prepared by drying the source solution on a mylar backing treated with insulin and supported on an aluminium annulus (external diameter = 5 cm). The sources were covered with mylar films of thickness 2 mg/cm². The source spread was kept in the range 2-4 mm diameter. The geometry of these sources was similar to that of
standard calibration sources used for efficiency calibration of
detectors. The count rate for different sources was kept around
1200 counts/s with the coaxial HPGe detector set-ups, to minimize
the pile up rejection rate of spectroscopy amplifier and dead
time correction of the multichannel analyser.

For the low energy measurements, using vertical HPGe/Si(Li)
detector, thin and uncovered sources were prepared by drying the
source solution on mylar backing treated with insulin and supported
on perspex ring (with external diameter 2.5 cm). The diameter for
these sources were kept in the range 2-3 mm. The count rate for
these sources was about 300 counts/s with the vertical Si(Li)/HPGe
detector set-ups.

1.4 Photopeak Efficiency Calibration of Photon Detectors

a) Coaxial HPGe detectors

The photopeak efficiency calibration of the 64.1 cm$^3$ and
96.0 cm$^3$ HPGe detectors over the energy region 80-1400 keV were
done by using a number of calibrated radioactive sources in fixed
and reproducible source-detector set-up (Section 3.1.1). The source-
to-detector distance was kept 25 cm for both the detectors. For
the present efficiency measurements, standard sources of $^{166m}$Ho,
$^{152}$Eu, $^{133}$Ba, $^{60}$Co, $^{137}$Cs, $^{54}$Mn, $^{94}$Nb and a mixed standard
containing $^{154}$Eu, $^{155}$Eu, $^{125}$Sb and $^{125m}$Te radionuclides were used.
These standard sources were procured from the National Bureau of
Standards, Washington, D.C., U.S.A. Emission rates of various
gamma rays and K X-rays were certified with uncertainties of 0.3-1.3%
These standard sources give count rates less than 500 counts/sec
with both the detectors.
Six spectra were taken for all these sources for time intervals ranging between 30,000-60,000 secs. The spectra were analysed to obtain areas of different photopeaks by using the computer code SAMPO (section 3.1.5) on a DEC-20 system at Regional Computer Centre, Chandigarh. The detector efficiency points at various energies were then determined by dividing the photopeak area per sec by the certified emission rate for the corresponding gamma ray after properly correcting for the radioactive decay.

The least square fitting of experimental efficiency for the 64.1 cm$^3$ HPGe detector over the different energy region; 80-400 keV, 300-460 keV and 420-1400 keV, was done by using the relation

$$\log_e(\text{eff}) = \frac{A(3)}{E^3} + \frac{A(2)}{E^2} + \frac{A(1)}{E} + A(0) + A(-1)E$$  \hspace{1cm} (3.1)$$

A computer program in double precision mode was used for this purpose. For the efficiency points for the 96.0 cm$^3$ HPGe detector, same relation (3.1) was least square fitted over the different energy regions; 90-300 keV, 250-465 keV, and 410-1400 keV.

For both the detector set-ups, the fitted efficiency curves were found to deviate from the experimental points by $\leq 0.8\%$ over all the energy regions and overlap very well over different energy regions (deviation is $\leq 0.3\%$). We estimate the calibration accuracy to be $0.8\%-1.0\%$ and $0.5\%-0.8\%$ over the energy regions 80-400 keV and 400-1400 keV, respectively. The large number of efficiency points and the small deviation of the fitted curves in the energy range 80-1400 keV encouraged us to give such low figures for error in efficiency calibration. The photopeak efficiency curves for the 64.1 cm$^3$ HPGe detector and 96.0 cm$^3$ HPGe detector at a source to
detector distance of 25 cm are shown in Figs. 3.3 and 3.4 respectively.

b) **Vertical planar HPGe detector**

The photopeak efficiency of the vertical planar HPGe detector over the energy region 20-400 keV was obtained by taking spectra of standard sources of $^{166}\text{Ho}$, $^{133}\text{Ba}$, $^{241}\text{Am}$ and a mixed standard containing $^{154}\text{Eu}$, $^{155}\text{Eu}$, $^{125}\text{Sb}$ and $^{125m}\text{Te}$, as explained in the section (3.1.4a). The absolute intensities of various gamma rays and X-rays from $^{241}\text{Am}$ were taken from Cohen and for $^{57}\text{Co}$ from NCRP report. The source-to-detector distance was kept 4 cm by using the set-up detailed in section (3.1.1). The experimental efficiency points over different energy region 20-105, 80-225 and 200-400 keV were least square fitted using eqns. (3.1) and (3.2).

$$\log_e(\text{eff}) = A(3)E^3 + A(2)E^2 + A(1)E + A(0)$$

These fitted curves were found to deviate from experimental points by less than 1% and overlap very well over different energy regions. The efficiency calibration errors for the vertical HPGe detector were estimated to be 1.0 - 1.5% and 1.5-2.5%, in the energy region 80-400 keV and 20-90 keV respectively. The photopeak efficiency curve for vertical HPGe detector is shown in Fig. (3.5).

c) **Vertical planar Si(Li) detector**

The relative efficiency curve for the Si(Li) detector in the energy region 4-90 keV, was obtained by putting radioactive standard sources of $^{55}\text{Fe}$, $^{241}\text{Am}$, $^{57}\text{Co}$ and $^{137}\text{Cs}$ and a mixed...
PHOTOPEAK EFFICIENCY CURVE OF THE 64.1 cm$^3$ HPGe DETECTOR (source-to-detector
Fig. 3.4 PHOTOPEAK EFFICIENCY CURVE OF THE 96.0 cm³ HPGe DETECTOR (source-to-detector distance = 25.0 cm)
Fig. 3.5 PHOTOPEAK EFFICIENCY CURVE OF THE VERTICAL PLANAR HPGe DETECTOR (source-to-detector distance = 4.0 cm)
standard containing $^{125}\text{Sb}$, $^{125m}\text{Te}$, $^{154}\text{Eu}$ and $^{155}\text{Eu}$ at the sample foil position with the $^{109}\text{Cd}$ annular source removed from the annular source geometry (Section 3.1.1c). As various sources (except the NBS mixed standard source) used for X-ray measurements were uncovered, therefore, the only absorption correction is due to self-absorption in source and is estimated to be less than 0.5% at 5 keV. Four spectra were recorded for each source for sufficiently long time to collect good statistics. The detector efficiency was calculated as already explained in the section (3.1.4a).

The number of efficiency points obtained by using radioactive sources were not sufficient for interpolation of detector efficiency in the energy region below 15 keV. More number of efficiency points in this energy region, were obtained by taking the $K_{\alpha}$ X-ray spectra from spectroscopically pure thin foils of Ti, Mn, Cu, Se, Ge and Y (thickness $\sim 100 \mu g/cm^2$) excited by 22.6 keV photons emitted from annular source of $^{109}\text{Cd}$ (15 mCi) using experimental set up detailed in section (3.1.1c). The efficiency of the Si(Li) detector ($\varepsilon_1$) at the $K_{\alpha}$ X-ray energies of various target elements was calculated by using the relation

$$ (I_0 G \varepsilon ) = \frac{N_{K_{\alpha}}}{\sigma_{K_{\alpha}} m_j \beta} $$

(3.3)

where $I_0 G$ is the intensity of the exciting radiations falling on the area of target foil visible to the detector. This factor was constant for all the foils. $N_{K_{\alpha}}$ is the number of counts/s under
the \( K_\alpha \) peak in the spectrum, \( m_j \) is the mass of the element in target in gm/cm\(^2\), \( \sigma_{K_\alpha} \) is the \( K_\alpha \) X-ray fluorescence cross-section (cm\(^2\)/gm) and was taken from Krause et al.\(^{11}\) \( \beta \) is the target self absorption correction for both the incident and the emitted \( K_\alpha \) X-ray radiation and is calculated by using the following expression

\[
\beta = \frac{1 - \exp(-1) \left[ \frac{\mu_{\text{inc}}}{\cos \theta} + \mu_{\text{emit}} \right] t}{\left( \frac{\mu_{\text{inc}}}{\cos \theta} + \mu_{\text{emit}} \right) t}
\]

(3.4)

where \( \mu_{\text{inc}} \) is the absorption coefficients for the incident photon energy, \( \mu_{\text{emit}} \) is the absorption coefficient for the emitted characteristic X-ray energy from the foil. The values of these absorption coefficients are taken from the table of Storm and Israel\(^{12}\). The effective angle of incidence in the present geometry works out to be 66°. The \( \beta \) correction factor for the above mentioned foils was found to be >0.993.

The estimated uncertainty in the relative values of the calculated fluorescent cross-section used for evaluation of efficiency is given to be 2.5 - 3.0%\(^{11}\). The uncertainty in the foil thickness was of the order of 2% and the error in the yield measurements under the K X-rays for different element was <0.7%. The factor \( I_0 G \) in eqn. (3.3) was evaluated by normalizing the product \( (I_0 G \cdot \epsilon) \) for Mn-\( K_\alpha \) and \( K_\beta \) X-rays to the efficiency points obtained from \( ^{55}\text{Fe} \) radioactive standard source. The final efficiency points obtained for Si(Li) detector at various energies in annular source geometry set up were least square fitted over different energy region 4.0 - 14.0, 12.0-32.0 and 30.0 - 86.0 keV.
by using the eqn. (3.1). The deviation of the fitted curve from the experimental points was found to be less than 1.5% over the whole energy region. Considering all the above mentioned errors, we have placed a very conservative error of 4% below 15 keV and 1.5 - 3.0% in the energy region 15-90 keV. The photopeak efficiency curve for the Si(Li) detector in annular geometry set-up is shown in Fig. 3.6.

3.1.5 Analysis of X-ray and Gamma ray Spectra

a) Gamma ray spectra

The information regarding the gamma ray energies and intensities is obtained by determining the position and area of individual photopeaks in a given spectrum obtained by using semi-conductor detectors. For the accurate determination of locations and areas of photopeaks, one must know the correct photopeak shapes and use these representations in analysis procedures. The primary factor determining the width of a photopeak is the statistical fluctuation in the division of the absorbed energy between ionization and heating of the detector crystal lattice. This gives rise to a Gaussian distribution with a small specified width. The material properties, the impurities in the detector and the electronic noise associated with leakage current affect the charge collection. The incomplete charge collection and random summing of pulses worsen the resolution and give rise to low energy tailing of the photopeaks. The continuum under the peaks is due to compton distributions from higher energy gamma rays and general background. The continuum in the short interval under the peak or a cluster
Fig. 3.6 PHOTONEAK EFFICIENCY CURVE FOR THE VERTICAL Si(Li) DETECTOR IN ANNULAR SOURCE GEOMETRY
of peaks can be assumed to be continuous and smoothly varying function of energy except for statistical fluctuations.

In the present work, a versatile computer program SAMPO\textsuperscript{13}) was used for the analysis of gamma ray spectra. This program describes the central part of a peak as gaussian and the tails of peak as simple exponentials which join the gaussian part so that the function and its derivatives are continuous at junction points. The continuum under the peak is approximated by a second order polynomial. The program searches the data for statistically significant peaks with fitting regions and performs single and multiple peaks fitting. The line shape calculations and fitting are performed by using a least square procedure with an iterative gradient minimization method with variable metric\textsuperscript{14}). The energy calibration of the spectrum is performed by a polynomial least square fit of the energy calibration points to account for the nonlinearities due to electric instabilities in collecting spectrum. The program also makes complete statistical and calibration error estimates.

b) \textbf{X-ray spectra}

The procedure discussed for the analysis of gamma ray spectra is not fully suitable for K X-ray photopeaks. The reason being that in case of gamma rays the natural line width is quite small in comparison with the full width at half maximum caused by the detector (by a factor of 10\textsuperscript{5}). However, the natural line width of characteristic K X-rays (sum of widths of the involved atomic levels) following EC or IC processes are not negligible
in comparison to resolution of the detector. For example, the line width of the Pb-K$_\alpha_1$ X-ray of energy 75 keV is about 62 eV and the fwhm of Si(Li) detector is 450 eV. According to the quantum mechanical theory, the natural line shape is described by Lorentzian function

$$L(E) = \frac{\Gamma/2}{(E-E_0)^2+(\Gamma/2)^2}$$

where $E_0$ and $\Gamma$ are the centre and the width of the line. Therefore, the observed peak shape for the K X-rays should be described by a Lorentzian function folded with Gaussian response function $G$

$$V(E) = \int_{-\infty}^{\infty} L(E') G(E-E') \, dE'$$

The detail studies of K X-ray line shapes from various elements have been studied recently by Campbell et al.\textsuperscript{15)}.

In case of L X-ray peaks, the Lorentzian correction can be discarded, as its natural line width of about 10 eV is very small as compared with the detector resolution of 240 eV at 10 keV. However, the L X-ray spectrum taken with Si(Li) detector is having complex peak distribution. For the nuclides studied in the present work, the L X-ray spectra were found to have four L X-ray groups $L_L$, $L_{\alpha \beta}$, $L_\beta$, and $L_\gamma$ each consisting of a cluster of strong and weak overlapping L X-ray peaks. The individual L X-ray peaks could not be easily resolved from the complex peak distribution by functional fitting.

For the analysis of present spectra, the photopeak areas were determined by graphical analysis. The spectrum was divided
into regions having well separated different groups of X-rays, namely, $L_x, L_\alpha, \eta_1, L_\beta, L_\gamma, (K_\alpha_1, K_\alpha_2), K_\beta_1, K_\beta_2$. Within these regions the peak area under each group of X-rays was determined by summing the counts. In each region, the continuum were approximated by smooth extrapolation of continuum from above and below the peaks. For the present intensity measurements, the energies of various K and L X-ray peaks were taken from the tables of Storm and Israel.

3.1.6 Coincidence Summing Corrections

There are two kinds of summing occurring in the detector which affect the area under the photopeaks in singles intensity measurements. First kind of summing effects are due to accidental coincidences of events detected in detector. The magnitude of random summing depends on the count rate of events being detected and the charge collection time in a semiconductor detector. These summing effects can be rendered negligible by using low counting rates and pile up rejection techniques. In the present work, the count rate for various standard calibration sources were less than 500 counts/s, therefore the corrections for random coincidence summing were not required. Also, since we were interested in relative intensity measurements, therefore, these corrections need not be applied in case of radioisotopes understudy.

Second kind of summing effects are due to detection of cascading events. Usually, the lifetimes of nuclear levels for gamma decay are much shorter than the charge collection time in a semiconductor detector. If two or more gamma rays are emitted
in cascade, any two or more of these may deposit energy in the
detector to form a composite pulse indistinguishable from that
due to single event. Thus, the area of peaks corresponding
to these gamma rays will get affected. Other radiations which
could also be in true coincidence include X-rays (from electron
capture, positron decay or internal conversion), $\beta^-$-particles
and their associated bremsstrahlung, and annihilation radiation
from positron decay. The magnitude of this effect depends upon
the specific features of the decay scheme and the solid angle
subtended by the detector at the source. In the present work, the
summing correction to various gamma rays detected by large volume
coaxial HPGe detectors were applied because of their high detection
efficiency.

Let us consider the decay scheme shown in Fig.(3.7a). The
rate of pulses resulting from full absorption of gamma rays of
energy $E_i$ in the detector is given by

$$N_i = A p_i \epsilon_i,$$

where $A$ is the activity of the source, $p_i$ is the emission
probability of gamma ray, $\gamma_i$, $\epsilon_i$ is the full energy peak effi­
ciency for $\gamma_i$-ray of energy $E_i$. Since each $\gamma_1$-photon emission
is followed by a $\gamma_2$-photon emission or internal conversion
process. It may happen that $\gamma_1$ and cascading $\gamma_2$ or converted
X-ray be detected simultaneously, thus leading to a single
sum pulse. If the energy of $\gamma_1$ is totally absorbed, this sum
pulse will be recorded at an energy between $E_1$ and $E_1 + E_2$. Thus
the event will be lost from full energy peak of $\gamma_1$. The rate
Fig. 3.7 SIMPLE DECAY SCHEMES TO DERIVE EXPRESSIONS FOR COINCIDENCE SUMMING CORRECTIONS
of pulses actually recorded under the photopeak of $Y_1$ will be given by

$$N'_1 = A p_1 \varepsilon_1 - A p_1 \varepsilon_1 \cdot \varepsilon_T^2 \cdot b_2 \overline{w}_{12}(0°) - A p_1 \varepsilon_1 \overline{\alpha}_k^T b_2 \alpha_k^T \omega_k,$$

(3.11)

where $b_1$ is the fraction of the decays from the level from which $Y_1$ originates which produce $Y_1$ photons. $\varepsilon_T^T, \varepsilon_k^T$ are the total efficiencies of the detector at the energy of $Y_2$ and mean K X-ray energy respectively. $\alpha_k^T$ is the K internal conversion coefficient for $Y_1$, $\omega_k$ is the fluorescent yield for the daughter element. (It is assumed that only K X-rays will enter the detector). $\overline{w}_{1j}(0°)$ is the directional correlation function of the gamma rays $Y_i$ and $Y_j$ averaged over the solid angle subtended by the detector, and is given by

$$\overline{w}_{1j}(0°) = 1 + a_2 A_{22} + a_4 A_{44},$$

(3.12)

where $A_{22}$ and $A_{44}$ are the directional correlation coefficients, and $a_2 = Q_{21} Q_{2j}$; $a_4 = Q_{41} Q_{4j}$; $Q$ is the solid angle corrections due to finite angular resolution of the detector (Section 3.2.5).

Similarly, the observed photopeak area for $Y_2$ will be lesser due to its summing with either $Y_1$ photons or with K X-rays produced due to internal conversion process competing $Y_1$-photon emission. The rate of pulses recorded under the $Y_2$-photopeak will be

$$N'_2 = A p_2 \varepsilon_2 - A p_2 \varepsilon_2 f_1 \overline{\varepsilon}_{1}^T \overline{w}_{12}(0°) - A p_2 \varepsilon_2 f_1 \overline{\alpha}_k^T \omega_k \overline{\varepsilon}_k^T,$$

(3.13)

Various symbols have similar meaning as in eqn. (3.11). $f_1$ is the fraction of the events populating the level to which $Y_1$ de-excites that are $Y_1$ photons.
The correction for the peak $Y_3$ is of negative type. Here, the summing of $Y_1$ and $Y_2$ leads to additional events in this peak. The observed area will include a term from the simultaneous deposition of $Y_1$ and $Y_2$ full energy and is given by

$$N_3' = A_p e_3 + A_p e_1 b_2 e_2 \bar{W}_{12}(0^\circ)$$

(3.14)

All the terms have usual meaning as detailed above.

For the decay scheme as shown in Fig.(3.7b), the number of pulses recorded under the photopeak of $Y_1$ is

$$N_1' = A p e_1 \left\{ 1 - b_2 e_2 \bar{W}_{23}(0^\circ) - e_k T b_2 x_w^k \right\}$$

$$b_2 e_3 \bar{W}_{13}(0^\circ) - b_2 b_3 x_w^k e_k$$

(3.15)

The observed area under the $Y_3$ peak will be

$$N_3' = A_p e_3 \left\{ 1 - f_2 e_2 \bar{W}_{23}(0^\circ) - e_k T f_2 x_w^k \right\}$$

$$f_2 f_3 e_3 \bar{W}_{13}(0^\circ) - f_2 f_3 x_w^k e_k$$

(3.16)

where $b_1 = (1 + T)$ is the fraction of the decays from the level depopulated by $Y_i$ that result in the transition corresponding to $Y_i$. $f_i (1+ T)$ is the fraction of the decays into the level populated by $Y_i$ that result from the transition corresponding to $Y_i$. The contribution due to triple coincidences being negligibly small is not considered in eqn. (3.15) and (3.16).

The area of sumpeak due to $Y_1$ and $Y_3$ is given by

$$N_{Y_1+Y_3} = N_1 b_2 e_3 \bar{W}_{13}(0^\circ)$$

(3.17)

The expressions for summing of gamma rays with X-rays following electron capture decays were written on the basis of two simple
3.1.7 **Total Efficiency of Coaxial HPGe Detector**

For applying the summing correction for cascading gamma-rays, the total efficiency of the detector as a function of energy is required. The total efficiency ($\varepsilon^T$) is defined as the ratio of the total number of counts observed to the number of gamma rays emitted. A general expression for total efficiency is given by

$$\varepsilon^T = \frac{1}{4\pi} \int (1 - \exp(-\mu S)) \, d\omega$$

(3.18)

where $S$ is the slant thickness of the detector in the direction of gamma ray (Fig. 3.8a). $\mu$ is the absorption coefficient in germanium and $d\omega$ is an element of the solid angle over which $S$ may be considered constant. For the coaxial HPGe detector, the expression for total efficiency derived by Griffiths\textsuperscript{18} (Fig. 3.8b) is

$$\varepsilon^T = k \frac{D}{Z} \left\{ \int_{R_1}^{R_2} \frac{\lambda}{(\lambda^2 + D^2)^{3/2}} \, d\lambda + \int_{R_1}^{R_2} \frac{\lambda}{(\lambda^2 + D^2)^{1/2}} \exp \left[ - \frac{\mu (R_2 - \lambda) (\lambda^2 + D^2)^{1/2}}{\lambda} \right] d\lambda \right\}$$

(3.19)
Fig. 3.8(a) GEOMETRY OF COAXIAL HPGe DETECTOR SHOWING CO-ORDINATE SYSTEM USED (b) CROSS-SECTION OF DETECTOR SHOWING REGIONS CONSIDERED FOR CALCULATING TOTAL EFFICIENCY
where \( D \) is the source to detector distance, \( R_1 \) is the radius of p type core, \( R_2 \) is the active radius of detector, \( t \) is the active length of the detector. \( K \) is the factor which corrects the efficiency for absorption of gamma ray in aluminium casing.

For the present work, the total efficiency of the 64.1 cm\(^3\) and 96.0 cm\(^3\) HPGe detectors were determined over the energy region 100-1500 keV using the above expression. The various dimensions of detectors used are given in Table (3.1). The absorption coefficients in germanium were taken from Chapman et al.\(^{19}\).

A computer program was written to evaluate various integrals in eqn. (3.19). Also, the total efficiency values of the coaxial detectors were measured using radioactive standard sources of \(^{60}\)Co, \(^{57}\)Co, \(^{54}\)Mn and \(^{94}\)Nb. The calculated total efficiency curves were normalized to the experimental points. Final total efficiency curves for the two detectors with 25.0 cm source-to-detector distance are shown in Fig. (3.9).

3.1.8 Calculation of X-ray Intensities in \( \beta^- \)-decay Nuclides

The deexcitation of an excited nucleus following \( \beta^- \)-decay can take place through gamma emission or internal conversion process. In the internal conversion process, the electrons from atomic shells are knocked out and the filling of the vacancies thus created by electrons from higher shells/subshells leads to emission of X-ray and \( \mu \)ger electrons. In the present work, the intensities of different K and L X-rays emitted as a result of internal conversion process competing the emission of a gamma ray of intensity \( I_\gamma \) were calculated using the formulation given below.
Fig. 3.9 TOTAL EFFICIENCY CURVES FOR HPGe DETECTORS
The intensities of different $K_i$ X-rays (where $i$ refers to $\alpha_1, \alpha_2, \beta_1'$ and $\beta_2'$) relative to gamma ray intensity $I_\gamma$ were calculated by using the equation,

$$I_{K_i} = I_\gamma \alpha_k \omega_k \frac{R_{K_i}}{\sum_{K_i}}$$

(3.20)

where $\alpha_k$ is the $K$-shell internal conversion coefficient, $\omega_k$ is the $K$-shell fluorescent yield and $R_{K_i}$ are the relative intensities of $K_i$ X-rays.

The number of vacancies produced in $L_i$-subshell ($i=1,2,3$) due to internal conversion process and from the decay of $K$-shell vacancies were calculated by

$$N_{L_i} = I_\gamma \alpha_{L_i} \left[ \frac{\alpha_{L_i}(IC)}{\alpha_k (IC)} + \eta_{KL_i} \right]$$

(3.21)

where $\alpha_{L_i}$ is the $L_i$ subshell internal conversion coefficient, $\eta_{KL_i}$ is the average number of primary $L_i$ subshell vacancies produced in the decay of one $K$ vacancy through radiative and Auger transitions.

The intensity of different $L$ X-rays following the filling of $N_{L_i}$ vacancies were calculated by using the following set of equations:

$$I_{L_1} = (N_{L_1} f_{13} + N_{L_1} f_{12} f_{23} + N_{L_1} f_{23} + N_{L_2} f_{23} + N_{L_3} ) \omega_3 F_{31}$$

$$I_{L_2} = (N_{L_1} f_{13} + N_{L_1} f_{12} f_{23} + N_{L_2} f_{23} + N_{L_3} ) \omega_3 F_{32}$$

$$I_{L_1} = (N_{L_1} f_{12} + N_{L_2} ) \omega_2 F_{2\eta}$$
where $\omega_1$, $\omega_2$, and $\omega_3$ are the fluorescent yields for $L_i$ ($i=1, 2, 3$) subshells; $f_{12}$, $f_{23}$, and $f_{13}$ are the Coster-Kronig transition probabilities of $L_1 \rightarrow L_2$, $L_2 \rightarrow L_3$, and $L_1 \rightarrow L_3$ subshells respectively, $F_{ij}$ ($i=1, 2, 3$; $j=1, \alpha, \eta, \beta, \gamma$) is the fraction of the X-rays originating from the $L_i$ subshell which contribute to the $L_j$ X-ray.

In case of $\beta$-radionuclides, emitting a number of gamma rays, the $K$ and $L$ X-ray intensities were calculated by adding the contributions due to internal conversion process competing with each the gamma ray. For the present calculations of X-ray intensities, the present measured gamma ray intensities were used. The $K$ X-ray relative intensities $R_{kL_i}$ based on relativistic-Hartree-Fock theory were taken from Storm and Israel\textsuperscript{12}. The values of $\alpha_{L_i}^{}$ and $\omega_1^{}$ were taken from ref.\textsuperscript{21}, $\eta_{kL_i}^{}$ from Rao et al.\textsuperscript{22}; $f_{ij}$ and $\omega_1^{}$ from Krause et al.\textsuperscript{11} and $F_{ij}$ from Scofield\textsuperscript{23} were used.

3.2 Gamma-gamma Coincidence Measurements

When nuclei undergo emission of two radiations in succession, the spatial and time correlation of emitted radiations are attractive and useful fields of study. To make such kind of studies, coincidence set-ups are required. A coincidence set-up is characterized by a parameter called resolving time ($2T$), i.e.
a coincidence can be detected if pulse due to one event lies within time $\pm \tau$ of the other. If there are two sources of uncorrelated pulses of rate $n_1$ and $n_2$ per second, the expected rate of accidental coincidences will be $2\tau n_1 n_2$ per second. If the recorded coincidence rate differs from expected chance rate, a correlation is established between the two sources of pulses. Both for the suppression of chance coincidences and for the measurement of short time intervals it is desirable to have coincidence resolving time as short as possible. This allows the use of relatively strong sources which further helps to have good statistics data collected in the measurements.

In the present work, the gamma-gamma coincidence studies were done for the following investigations.

i) to identify cascading gamma rays during the de-excitation of daughter nucleus.

ii) the gamma-gamma directional correlation measurements.

iii) the K-shell internal conversion measurements for certain transitions in various nuclei by $\gamma - \gamma - X$ coincidence method.

The present measurements were done by using following two coincidence set-ups.

1. A coincidence set-up involving $96 \text{ cm}^3$ HPGe and $90 \text{ cm}^3$ HPGe detectors.

2. A coincidence set-up involving $64.1 \text{ cm}^3$ Ge(Li) and
   $5.1 \text{ cm} \times 5.1 \text{ cm}$ NaI(Tl) detectors.

3.2.1 The HPGe-HPGe Detector Coincidence Set-up

For the coincidence and directional correlation studies in case of nuclides with complex decay schemes, a high resolution coincidence set-up involving $90 \text{ cm}^3$ and $96 \text{ cm}^3$ HPGe detectors,
was used. The energy resolution of both the detectors was 1.8 keV for 1332 keV gamma rays. The HPGe detectors were mounted on two firm arms of a versatile directional correlation table. The arms of the table were movable in all four quadrants in horizontal plane about the fixed central axis and can be locked at any position. The detectors were levelled and properly aligned with their horizontal axes intersecting at the central axis of the table. The angle between the two detectors can be changed up to an accuracy of 1°. The source was mounted on the fixed central axis of the table using a perspex holder. The source-to-detector distance could be varied up to 40 cm for both the detectors. The detectors were shielded with lead cylinders and cones with Compton graded shield of tin and copper on the inside to minimize the cross-scattering between the detectors and to reduce the detection of background radiations.

For the present measurements, the 90 cm$^3$ HPGe detector was used for gating gamma peaks and the 96 cm$^3$ HPGe detector was used as analog detector. The block diagram of the HPGe-HPGe detector coincidence set-up used in the present work is shown in Fig. 3.10. In channel 1 of the coincidence circuit set up, the energy output from the preamplifiers of both HPGe detectors were fed to two spectroscopy amplifiers (ORTEC 471) having switch selectable integration and differentiation time constants $\tau =0.5$, 1 or 2 $\mu$s. The bipolar output from each of these amplifiers was fed to two timing single channel analysers (TSCA)(ORTEC 420A). The TSCA on the 90 cm$^3$ HPGe detector side was used to gate the
Fig. 3.10 THE HPGe-HPGe COINCIDENCE SET-UP
desired photopeak (differential mode) and that on the 96 cm³ HPGe detector side was used to accept the entire spectrum (integral mode). The TSCA triggers at a time when the bipolar pulse crosses the baseline and provides output timing pulse (+5V, rise time ≤ 20 ns, width ~ 500 ns). The output signal can be delayed by 0.1 - 1.1 µs after the trigger. The positive outputs from the two TSCA's were fed to a fast coincidence circuit (FCC) (ORTEC 414A). The FCC gives coincidence output (+5V, width ~ 500 ns) if the leading edges of input pulses are within the resolving time (10-110 ns). The pulses from both the detectors were suitably delayed by using the 0.1 - 1.1 µs delays in the TSCA units to give maximum coincidence counts at the FCC output. The output of FCC was further used to strobe the time-to-pulse height converter in the second channel.

The timing measurements of signals from HPGe detectors were performed using the output from the charge sensitive preamplifier in order to maintain the energy resolution of these detectors. But the noise of the detector & preamplifier and the variation in charge collection time in the detector creates error in timing determination from the signal. Therefore the timing information was drawn by using constant fraction pulse height timing technique. In the second channel of the coincidence circuit, the energy outputs from preamplifier of both the HPGe detectors were fed to timing filter amplifiers (TFA) (ORTEC 454) for proper shaping and amplification for timing measurements. These amplifiers have RC-integration circuit (time constant 2, 5, 10, 20, 50, 100 and 200 ns) and RC-differentiation circuit (time constant
5, 10, 20, 50, 100, 200 and 5000 ns). The amplifiers have adjustable gain at any value between 2 to 200 and have output impedance of 50 ohms. The negative output pulses from TFA's are fed to two constant fraction discriminators (CFD) (ORTEC 473) with shaping mode set for Ge-detectors. The CFD was set to trigger at 50% amplitude for all input signals and eliminates time walk over wide range of amplitudes for input signals (50 mV-5V) of definite risetime. The output of the CFD's were negative pulses of fixed width \( \leq 10 \text{ ns} \) and fixed rise time \( \leq 2.5 \text{ ns} \).

The negative output signals from CFD on the 90 cm\(^3\) HPGe detector side used for gating purposes, were fed to start input of time-to-pulse height converter (TPHC) (ORTEC 457), whereas those from CFD on the 96 cm\(^3\) HPGe detector side were fed to its stop input after suitably delayed by using delay boxes (ORTEC 425). The TPHC produces an analog output pulse with amplitude proportional to the time interval between its start and stop inputs. The start-to-stop time conversion is accomplished after a valid start has been identified and only if a stop pulse arrives within the selected time range (50 ns-80 \( \mu \text{s} \)). The start input is disabled during the busy interval to prohibit pile up and a stop pulse is disabled after the first stop signal is accepted. The time-to-pulse height converter (TPHC) was further strobed by the FCC output from first channel i.e., the output of TPHC will appear only if the strobe signal is furnished within the selected interval (5 or 120 \( \mu \text{s} \)) after the stop pulse. The output of TPHC is a positive pulse (rise time \( \leq 300 \text{ ns} \), width 1.0-2.5 \( \mu \text{s} \)) and is linear between 0 to 10V. The positive output
pulse of the TPHC was fed to another TSCA (ORTEC 420A) which is used to select the prompt portion of the time spectrum. The width of the output pulse from TSCA was increased to about 4\(\mu\)s using a pulse stretcher (ORTEC 411) and was used for gating the multichannel analyser (ND 66B). The energy output of 90 cm\(^3\) HPGe detector, amplified through a spectroscopy amplifier (ORTEC 472A) and suitably delayed through a delay amplifier (ORTEC 427A) was fed to the signal input of MCA. The number of pulses from FCC, the counts under the gated peak and the counts used for gating MCA were simultaneously recorded by using a Quad Counter (ORTEC 872).

For the measurement of time resolution of the present coincidence set up, the 1173-1332 keV cascade from the decay of \(^{60}\)Co was used. The output of TPHC was fed to ND 66B MCA in singles mode to record the timing information. The MCA was calibrated for time (ns) by measuring the prompt peak shift corresponding to change in delay (ORTEC 425A) between start and stop pulses. The full width at half maximum of the prompt peak in units of time gave the time resolution of the coincidence circuit. For the present set-up, a time resolution of \(\sim 7\) ns was achieved for 1173-1332 keV cascade from \(^{60}\)Co decay.

3.2.2 The Ge(Li)-NaI(Tl) Coincidence Set-up

The coincidence and directional correlation measurements involving weak gamma rays require a coincidence set-up with good efficiency. Since the HPGe detectors are known to have low detection efficiencies as compared to NaI(Tl) detectors, therefore,
The HPGe-HPGe detector coincidence set-up is not very suitable for the above mentioned coincidence measurements. To make up the requirement of good efficiency, a coincidence set up involving 5.1 cm x 5.1 cm NaI(Tl) and 64.1 cm^3 Ge(Li) detector was also used for the present measurements. The obvious limitation of this set-up is the poor resolution of NaI(Tl) detector. The energy resolution of the Ge(Li) detector was 2.1 keV at 1332 keV.

The output from the NaI(Tl) detector was used for gating different gamma rays and HPGe detector was used as an analog detector. Both the detectors were shielded as described in section (3.2.1). The HPGe detector was kept fixed with source placed on its axis at a distance of 10 cm. The NaI(Tl) detector was mounted on the movable arm of the table, and could be rotated between 90° and 270° w.r.t. fixed detector with source placed at the axis of rotation. The NaI(Tl) detector could be locked at intervals of 15°. The distance between the NaI(Tl) detector and the source could be varied and it was kept 10 cm for the present measurements.

The block diagram of Ge(Li)-NaI(Tl) coincidence set-up used in present work is shown in Fig. (3.11). The coincidence circuit is very much similar to that used with HPGe-HPGe detector coincidence set-up detailed in previous section. In the first channel, the slow pulses from dynode of photomultiplier tube connected to NaI(Tl) crystal, were fed to preamplifier (ORTEC 113). The energy output from preamplifiers of NaI(Tl) and Ge(Li) detectors were fed to spectroscopy amplifier (ORTEC 471), TSCA (ORTEC 420A) and FCC (ORTEC 414A) as detailed in previous section.
Fig. 3.11 THE Ge(Li) – NaI(Tl) COINCIDENCE SET-UP
In the second channel, the anode output of the photomultiplier tube connected to NaI(Tl) crystal were fed to a preamplifier (ORTEC 113). The negative amplified anode output from preamplifiers of NaI(Tl) detector through TFA (ORTEC 454), CFD (ORTEC 473, shaping mode set for NaI(Tl) detectors) and delay boxes (ORTEC 425A) were used as stop input of the TPHC (ORTEC 457). Similarly, the output from Ge(Li) preamplifier through TFA and CFD (shaping mode set for Ge(Li) detector) were fed to TPHC as start input. The TPHC was further strobed by FCC output from upper channel. The selected output of TPHC through TSCA and pulse stretcher was used for gating ND-100 MCA. The main signal to the MCA was provided by energy output of 64.1 cm$^3$ Ge(Li) detector, through a spectroscopy amplifier and a delay amplifier.

The time resolution of NaI(Tl)-Ge(Li) coincidence set-up was achieved to be $\sim 6$ ns for 1173-1332 keV cascade from $^{60}$Co as explained in previous section.

3.2.3 Source Preparation

For the directional correlation studies, the radioactive sources of size (2mm dia and 3mm height) were prepared by putting source solution in a perspex rod (diameter 4 mm) with a vertical cavity. The source holder was sealed properly to keep the source in liquid form for long periods. The count rate for various sources was adjusted according to the requirements of the experiments.
3.2.4 Processing and Analysis of Gamma-Gamma Directional Correlation Data

For the present directional correlation measurements, the HPGe-HPGe and NaI(Tl)-Ge(Li) detector coincidence set-ups detailed in sections (3.2.1 and 3.2.2) were used. The source in cylindrical perspex holders (section 3.2.3) was mounted at the central axes of the correlation table. The centring of the source was done with an accuracy better than 1% by noting the counts under the gated peak of movable detector at different angles. The coincidence spectra were taken at seven angles between 90° and 180° at the intervals of 15°, for the time intervals 3000-5000 s at each angle to minimize the systematic errors due to electronic shifting, change in dead time correction with decay of source etc. The data were taken for a number of cycles to collect enough statistics under coincidence peaks at each angle. In each case the data were summed up over all the channels comprising a peak in the coincidence spectrum. The background under the peak was estimated to be linear under the peak and the same was subtracted from the gross coincidence counts.

The correction for miscentring of the source and the radioactive decay were applied by normalizing the coincidence counts by the corresponding singles counts under the gated peak of the movable detector. The chance coincidence counts were separately estimated by shifting the gate to the chance region of the time spectrum at the output of the TPHC. The contribution to coincidence counts due to compton counts accepted by the gated energy region were estimated from the coincidence spectrum taken by...
shifting the window of the respective TSCA to the compton region above the peak of interest.

After applying all the above mentioned corrections, the gross coincidence data at various angles were fitted to the directional correlation equation

\[ W(\theta) = 1 + A_{22} P_2(\cos \theta) + A_{44} P_4(\cos \theta) \]  

using the method of least squares, to evaluate the directional correlation coefficients \( A_{22} \) and \( A_{44} \). These directional correlation coefficients were further corrected for the finite size of the source and the finite angular resolution of the two detectors as explained in section (3.2.5). The corrections to these coefficients due to influence of extranuclear fields was applied, whenever necessary, as discussed in section (2.1.4).

The correlation coefficients \( (A_{kk}) \) are related to the directional distribution coefficient \( (A_k) \), the orientation parameter \( (B_k) \) and the reorientation parameter \( (U_k) \) of the involved cascading transition by eqns. (2.5) and (2.9) for the double and triple gamma correlations respectively. These coefficients \( A_k, B_k \) and \( U_k \) for the involved transitions are further related to the mixing ratio of these transitions, the spin of the involved nuclear levels by eqns. (2.6), (2.7) and (2.11) respectively. These equations were used to determine the multipole mixing ratios of the mixed transitions and in some cases to assign the spins of the nuclear levels from the measured correlation coefficients \( (A_{kk}) \). From the above analysis, two values of mixing ratio of a mixed transition are obtained and a unique value out of these is selected with the help of some other
measurements e.g. correlation measurements in a different cascade, internal conversion measurements or theoretical considerations.

3.2.5 Solid Angle Correction Factor

For a point source situated on the axis of symmetry of point detectors, the theoretical directional correlation function is given by eqn. (3.23). However, owing to the finite size of source and detector used in actual practice, the observed directional correlation function will differ somewhat from ideal distribution. As has been discussed by Rose\textsuperscript{25}, the effect of finite detector size is to introduce into each term in angular correlation function an attenuation factor $Q_k$. This will make the distribution to smear out somewhat without changing the form of $\hat{W}(\theta)$ function, for the detectors having axial symmetry about the propagation direction of the gamma ray. The ideal correlation coefficient $A_{kk}$ are related to the measured ones $A_{kk}^{\text{exp}}$ using detectors of finite size by the equation,

$$A_{kk} = \frac{A_{kk}^{\text{exp}}}{Q_k}$$  \hspace{1cm} (3.24)

Since the detector 1 accepts $Y_1$ and detector 2 accepts $Y_2$, the $Q_k$ includes the correction for both the detectors and is given by

$$Q_k = Q_k^1 (Y_1) Q_k^2 (Y_2)$$ \hspace{1cm} (3.25)

Because of the energy dependence of the gamma ray absorption coefficient, the $Q_k$ for a given detector depends on the gamma ray energy. In addition, they depend on the size and shape of the detector and on the source-to-detector distance.

Following the method of Rose\textsuperscript{25}, we take
$$Q_k(Y_i) = \frac{J_k(Y_i)}{J_0(Y_i)}$$  \hspace{1cm} (3.26)

where $J_k(Y_i) = \int P_k(\cos \beta)(1-e^{-\tau(Y)\chi(\beta)}) \sin \beta \, d\beta$  \hspace{1cm} (3.27)

The integration is carried out over the path length $x(\beta)$ through the active volume of the detector, $\beta$ is the angle between the propagation direction of the gamma ray (Fig. 3.12) and the detector symmetry axis. $\tau(Y)$ is the absorption coefficient for the full energy peak.

For the coaxial germanium detectors used in the present directional correlation set-up, the solid angle correction factors $Q_k^i(Y_i)$ were calculated using the detector size quoted by manufactures (Table 3.1) and the integration subroutine given by Krane\textsuperscript{26}. The values of absorption coefficients $\tau(Y)$ at different gamma ray energies computed by Krane\textsuperscript{26} were used in the present calculation of $Q_k^i(Y_i)$. For the NaI(Tl) detectors of different sizes, Yates\textsuperscript{27} has tabulated the values of correction factors $Q_k^i$ for different gamma ray energies and different source-to-detector distances. These values were used in present directional correlation measurements using Ge(Li)-NaI(Tl) detector set up.

The correction due to finite length of the source used in present work were found to be negligibly small by using the formulation given by Jagam et al.\textsuperscript{28}.

3.2.6 The Internal Conversion Coefficient Measurements

The K-shell internal conversion coefficient for certain transitions in the daughter nuclei following $\beta^-$-decay, were determined by $Y-YX$ coincidence method. This method is particularly
Fig. 3.12 GEOMETRY OF SOURCE DETECTOR CONFIGURATION
USED FOR CALCULATING SOLID ANGLE CORRECTIONS
FOR COAXIAL HPGe DETECTORS
useful for the low energy transitions where the energy of the internally converted K-electrons is low and their intensities cannot be measured accurately. The coincidence spectrum obtained by gating one of the gamma rays will contain peaks due to (i) all gamma rays cascading with the gated gamma ray (ii) X-rays produced due to internal conversion process competing the gamma transition, cascading the gated gamma ray. In a simple case, where only one gamma ray in the decay scheme is cascading the gated gamma ray, the K-internal conversion coefficient for that gamma transition can be determined by measuring the intensities of gamma and K X-rays in coincidence spectrum. The K conversion coefficient is deduced by using the relation

\[ \alpha_k = \frac{I_{kX}}{I_Y} \frac{1}{\omega_k} \]  

(3.28)

where \( \omega_k \) is the K-shell fluorescent yield for the daughter atom. However, this method becomes tedious if the gated gamma ray is in coincidence with more than one gamma ray as in such cases the coincidence X-ray intensity cannot be interpreted accurately.

In the present work, the coincidence set up involving 90 cm\(^3\) HFGe & 96 cm\(^3\) HFGe detectors was used for determination of internal conversion coefficients. The details of set up are given in section (3.2.1). The angle between the two detectors was kept 125°, \( P_2(\cos \theta) = 0 \) at \( \theta = 125^\circ \), so that angular correlation effects could be neglected. The source-to-detector distance was kept 15 cm for both the detectors. One of the gamma ray was gated through 90 cm\(^3\) HFGe detector and the
coincidence spectrum observed by 96 cm³ HPGe detector was recorded on 4K-channel, ND-66B analyser. The internal conversion coefficient is then deduced using the relation;

\[
\alpha_k = \left( \frac{N_{K\alpha}}{N_{\gamma}} \right)_{\text{coin}} \left( \frac{I_{K\alpha} + I_{K\beta}}{I_{K\alpha}} \right) \left( \frac{1}{\omega_k} \right) \left( \frac{\varepsilon_\gamma}{\varepsilon_{K\alpha}} \right) \tag{3.29}
\]

where \(N_{K\alpha}\) and \(N_{\gamma}\) are the areas under the \(K\alpha\) and \(\gamma\)-peaks in coincidence spectrum, \(\omega_k\) is the \(K\)-shell fluorescent yield and is taken from Krause et al. \^\textsuperscript{11} \(\varepsilon_\gamma\) and \(\varepsilon_{K\alpha}\) are the efficiencies of the 96 cm³ HPGe detector at \(\gamma\)-ray and \(K\alpha\) X-ray energies respectively. The ratio \((\varepsilon_\gamma/\varepsilon_{K\alpha})\) was determined by taking singles spectrum with 96 cm³ HPGe detector with the source detector geometry as for coincidence measurements.

\[
\frac{\varepsilon_\gamma}{\varepsilon_{K\alpha}} = \left( \frac{N_{K\alpha}}{N_{\gamma}} \right)_{\text{singles}} \left( \frac{I_{\gamma}}{I_{K\alpha}} \right) \tag{3.30}
\]

The intensity ratios \((I_{K\alpha} + I_{K\beta})/I_{K\alpha}\) in eqn. (3.29) and \(I_{\gamma}/I_{K\alpha}\) in eqn. (3.30) were determined precisely in singles intensity measurements as detailed in section (3.1). By this method of efficiency determination, the corrections due to absorption of photons in source geometry, summing at small distances and count rate effects etc. were automatically taken into account.

3.3 **Coulomb Excitation Measurements**

In the present work, the Coulomb excitation experiments were carried out using proton beam from Variable Energy Cyclotron at Panjab University, Chandigarh, India. The general experimental
set-up, electronics employed to record the data and the analysis procedure adopted for the treatment of data to extract E2 transition probability information about various levels of target nuclei, are described in the following sections.

3.3.1 Experimental Set-up

a) Accelerator

It is a single dee classical 23" dia cyclotron, which provides 2-4.5 MeV protons with energy resolution 30 keV. An analyzed, well collimated proton beam of 5 mm dia was available in the target chamber.

b) Target Chamber

The chamber was a hollow cylinder of brass 3" in dia., 8" in height and uniform wall thickness (1.5 mm) and was fixed at the end of the beam line. The chamber was having transparent perspex window at 135° in the plane of the beam.

c) Target holder

The target was placed in a target holder of aluminium, which was suspended at the centre of the target chamber, such that proton beam was falling at the centre of the target. The target was covered with a tantalum sheet having a hole of 1.1 cm diameter in it. A teflon ring was sandwiched between the target and the tantalum front. The tantalum sheet was grounded, so that the charge collected on it did not produce any potential on it.

d) Ge(Li) detector set-up

The deexcitation gamma rays from target were detected by using a 50 cm$^3$ Ge(Li) detector having an energy resolution of
2.5 keV for the 1332 keV gamma ray from $^{60}$Co. The Ge(Li) detector was mounted on a directional distribution table which was free to rotate on a levelled metallic heavy platform, such that its axis of rotation and vertical axis of the target chamber were coinciding with each other. The detector was shielded using lead cone and cylinder with tin and copper graded shield inside. The electronics used with Ge(Li) detector to measure gamma ray spectra was similar to that detailed in section 3.1.2. The detector bias of Neg. 3000 V was supplied to the Ge(Li) detector by ORTEC 459 bias supply. The power to the preamplifier of Ge(Li) detector was applied by power supply (ORTEC 114). Pulses from the preamplifier located inside the detector assembly were amplified by a spectroscopy amplifier (ORTEC 572, shaping time 6 $\mu$s) and then fed to a 4096-channel ND-100 analyser. The stored spectrum data were listed out onto the teleprinter (model 33 ASR).

3.3.2 Experimental Procedure and Data Analysis

The target under study, sufficiently thick to completely stop the proton beam was fitted in the target holder and was suspended at the centre of the target chamber facing the beam direction. The bombarding energy were chosen to be well below the Coulomb barrier of the target nucleus, so that no compound nuclear formation was expected at this safe energy (eqn.2.17). The Ge(Li) detector was placed at an angle of 55° to the beam direction in order to minimize the angular distribution effects ($P_2(\cos\theta) = 0$ at $\theta = 55^\circ$). The detector was kept at
a distance of 15 cm from the target. The absolute efficiency of the Ge(Li) detector in the target-detector geometry was determined by placing $^{152}$Eu standard source and mixed standard source containing $^{154}$Eu, $^{155}$Eu, $^{125}$Sb and $^{125m}$Te radionuclides, at the target position, in a similar way as discussed in section (3.1.4).

The deexcitation gamma rays observed by Ge(Li) detector were recorded on NEL100 multichannel analyser and the beam current falling on the target were simultaneously measured by a current integration (ORTEC 439). The real and live times of the multichannel analyser were noted for applying dead time correction. The gamma ray spectra were analysed to obtain area under the various photopeaks. Then, the thick target coulomb excitation gamma ray yield per incident proton $\gamma(Y)_{\text{corr}}$, was extracted using the following relation,

$$
\gamma(Y)_{\text{corr}} = \frac{1.602 \times 10^{-19}}{\gamma a} \left[ \frac{N(Y)}{\overline{W}(\theta) \epsilon_Y \tau_D} - T_c \right]
$$

where $q$ is the charge in Coulombs collected at the target during beam exposure, $a$ is the fractional isotopic abundance in target, $N(Y)$ are the number of counts in the full energy gamma peak, $\epsilon_Y$ is the detector efficiency in the target-detector set-up at a particular gamma-ray energy, $\overline{W}(\theta)$ is the factor that accounts for the anisotropic gamma ray angular distribution. In the present measurements $\theta$ is kept equal to 55° and therefore $\overline{W}(\theta) = 1$. $T_c$ is the factor which accounts for the population of the level of interest from the decay of higher excited levels and $\tau_D$ ($= \text{Live Time}/\text{Real Time}$) is the dead time correction.
3.3.3 Extraction of $E2$ Transition Probability $B(E2)$

The number of nuclei excited to a particular level $1$ per incident particle of energy $E_i$ by E2 excitation mode in a thin target of thickness $dx$ (assuming no energy loss of incident ions in target) can be written as

$$dN(1) = \sigma_{E2}(E_i) \frac{N_A}{M} a \rho \, dx$$

(3.32)

where $N_A$ is Avogadro's number, $M$ and $\rho$ are the molecular weight and the density of the target material, $a$ is the number of target nuclei per molecule, $\sigma_{E2}(E)$ is the total cross-section for the E2 excitation and is given by eqn. (2.33) i.e.

$$\sigma_{E2} = 4.819 \left( \frac{1}{1 + \frac{A_1}{A_2}} \right)^2 \frac{A_1}{Z^2} (E - AE') B(E2) \int_{E'}^{E} \eta_{E2}(E) \, dE$$

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where the various terms have the same meaning as detailed in section (2.2).

In case the target is thick enough to stop the incident beam completely i.e. $E_f = 0$, the number of nuclei excited to a particular level $1$ by E2 excitation mode per incident particle can be obtained by integrating the eqn. (3.32) over the trajectory of the incident particle i.e.

$$N(1) = \frac{N_A}{M} a \rho \int_0^{E_i} \frac{\sigma_{E2}(E)}{dE/dx} \, dE = \frac{N_A}{M} a \int_0^{E_i} \frac{\sigma_{E2}(E)}{dE/dx} \, dE$$

(3.33)

where $dE/dx$ is the stopping power of the target material for incident particles in the units of MeV cm$^2$/gm. Then the yield of gamma ray ($Y_g$) following deexcitation of level $1$ from target nuclei excited per incident particle is given by
\[ Y(Y_k) = \frac{N_A}{M} a \epsilon(Y_k) \int_0^{E_1} \frac{\sigma_{E2}}{dE/dx} \, dE + T_c \]  

(3.34)

where \( T_c \) is the contribution due to decay of higher excited levels to level 1, \( \epsilon(Y_k) \) is the decay fraction for gamma ray \( (Y_k) \) from level 1 and is given by

\[ \epsilon(Y_k) = \frac{f_k}{\sum_{j} (1 + \alpha_j^2) f_j} \]  

(3.35)

where \( f_k \) is the relative intensity of the gamma ray \( (Y_k) \).

The gamma ray yield per incident particle, after correcting for the contribution due to second term in eqn. (3.34) and using the expression for \( \sigma_{E2} \) (eqn. 2.33), can be written as

\[ Y(Y_k)_{corr} = \frac{N_A}{M} a \epsilon(Y_k) \int_0^{E_1} 4.819 \left( \frac{\alpha_2}{\alpha_1 + \alpha_2} \right)^2 \frac{A_1}{Z_2^2} (E - \Delta E') B(E2) f_{E2}(\eta_1, \xi) \, dE \]  

(3.36a)

Introducing \( K = \left[ \frac{4.819}{M} \frac{N_A}{a} \left( \frac{A_2}{\alpha_1 + \alpha_2} \right)^2 \frac{A_1}{Z_2^2} \right] \) and

\[ I_{E2} = \int_0^{E_1} \frac{(E - \Delta E') f_{E2}(\eta_1, \xi)}{dE/dx} \, dE, \]  

(3.36b)

the expression for the reduced E2 transition probability \( B(E2) \) can be written as

\[ B(E2) = Y(Y_k)_{corr} \frac{1}{(K \cdot I_{E2})} \]  

(3.37)

The gamma ray yield per incident particle, \( Y(Y_k)_{corr} \) was deduced from experimental measurements detailed in section (3.3.2).

A computer program was used to evaluate the integral \( I_{E2} \). The required values of \( f_{E2}(\eta_1, \xi) \) were interpolated from the tables given by Alder et al. [23]. For the \( dE/dx \) term, the
Bethe's formula\(^{28}\) for the stopping power was used, which holds good above energies 2 MeV/ a.m.u and for compound medium, it can be written as

\[
- \frac{dE}{dx} = (YZ_1)^2 \frac{0.307}{M \beta^2} \sum_i a_i I_i \ln \left( \frac{1.022 \times 10^6 \beta^2}{I_i} \right) \text{ MeV cm}^2 \text{ g}^{-1} \text{ m} \quad (3.38)
\]

where \(I_i = 9.1 \, Z_2 \left(1 + 1.9 \, Z_2^{-2/3}\right)\) is the mean ionisation potential of atom in units of eV, \(YZ_1\) is the effective charge of the incident ion, \(M\) is the molecular weight of the target material, \(a_i\) is the number of atoms of type \(i\) per molecule of target material; \(\beta = \frac{\nu}{c}\), \(\nu\) is the velocity of incident ions.
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