Chapter 2
Theoretical Background of the Two-Photon Decay Experiment

2.1 The Two-Photon Decay Probability

The theory of the two-photon decay has been studied many times in literature [1–3]. A complete description is given by Friar [4]. Based upon his results the authors of Ref. [5] further extended the theoretical analysis with an emphasis on $0^+_2 \rightarrow 0^+_1$ transitions. The crucial differential two-photon decay probability is only given for the $0^+_2 \rightarrow 0^+_1$ case in Ref. [5]. J. Millener derived this quantity for the general case which is needed for the investigation of $^{137}$Ba [6, 7].

The interaction between the nucleus and an electromagnetic radiation field can be written as (Gaussian units with $\hbar = c = 1$ and $\alpha = e^2$) [4]

$$H_{int} = \int j_\mu(x) A^\mu(x) d^3x + \frac{1}{2} \int B_{\mu\nu}(x, y) A^\mu(x) A^\nu(y) d^3x d^3y.$$ (2.1)

where $j_\mu(x) = (\rho(x), \vec{j}(x))$ is the current operator and $A^\mu(x)$ is the vector potential. $B_{\mu\nu}(x, y)$ is the so called ‘seagull’ operator which results from the non-relativistic treatment of the hamiltonian [4]. As explained in Ref. [5] in a fully relativistic theory the ‘seagull’ operator corresponds to a sum over very high mass states which contain virtual nucleon-antinucleon pairs. These very complicated contributions are effectively taken into account by the ‘seagull’ operator.

If one neglects the second term in Eq. 2.1 and treats the first one in first order perturbation theory—which is well justified because the electromagnetic interaction is weak—one obtains the well known equations for the one-photon decay [8] which are extensively used in nuclear physics. The corresponding Feynman diagram of the one-photon decay is shown in Fig. 2.1a.

In order to obtain the differential two-photon decay probability one has to treat the first term in Eq. 2.1 in second order perturbation theory and the second term in first order perturbation theory. The Feynman diagrams which contribute to the total two-photon decay amplitude are shown in Fig. 2.1b–d. The so-called resonance
amplitudes from the second order perturbation theory of the $j \cdot A$ term in Eq. 2.1 are displayed in (c, d) and the ‘seagull’ amplitude, which is due to the first order perturbation theory of the $A^2$ term, is shown in (b).

Starting from Eq. (A.34.a) of Ref. [5] one obtains, for a transition from a state with spin $I_i$ to a state with spin $I_f$ after integrating over all angles, expect the one between the emitted photons $\theta$, the differential two-photon decay probability [6]

$$
\frac{d^2 \Gamma_{\gamma\gamma}}{d\omega d(\cos \theta)} = \frac{\omega \omega'}{(2I_i + 1)\pi} \sum_{LL'S'L} \sum_{SS'QJ} \lambda \lambda'
\cdot \left\{ \frac{\hat{J}^2}{L L'} \cdot \langle L - \lambda \hat{L} \lambda | Q0 \rangle \cdot \langle L' - \lambda' \hat{L}' \lambda' | Q0 \rangle \cdot U(\hat{L} J Q L', \hat{L}' L) \right.
\cdot \langle S L' | \tilde{S} L | 0 \rangle \cdot \langle S' L | \tilde{S}' L | 0 \rangle \cdot \langle \omega | \omega' \rangle \cdot P_J(\cos \theta) \cdot P_J'(S' L' SL, \omega' \omega) \cdot P_J(\tilde{S}' \tilde{L} L, \omega' \omega)' \bigg\},
$$

with $\hat{x} = \sqrt{2x + 1}$. A rather similar expression is presented in Ref. [2]. The equation is valid for the specialization that the spin of the initial nuclear state is randomly oriented and only the emission directions of the two photons are measured. Additionally Eq. 2.2 does not consider the ‘seagull’ amplitude. According to Ref. [5] this part contributes only if both transitions are magnetic which is not the case for the considered transition in $^{137}\text{Ba}$ (see Sect. 2.2).

The sums run over all unobserved quantum numbers. $L$ and $S$ ($S = E = 0$ and $S = M = 1$) are the multipoles and the multipole characters of the virtual transitions, respectively. $J$ denotes the total angular momentum transfer carried by the two photons. The helicity is $\lambda = +1(-1)$ for positive (negative) helicity photons. The angular distribution is determined by the Legendre polynomials $P_Q(\cos \theta)$ and the $Q$ dependent coefficients. The energies of the two photons $\omega$ and $\omega'$ have to satisfy the condition $\omega + \omega' = \omega_0$ with $\omega_0$ being the transition energy. The angular momentum coupling coefficient $U$ is defined in Eq. A.1 of the Appendix.

The nuclear structure information is contained in the so-called generalized polarizabilities $P_J$ which are given through [5]
The expression is symmetric under exchange of $\omega$ and $\omega'$. 

Furthermore the generalized polarizabilities have to obey the following parity selection rule

$$(-1)^{L'+S'+L+S} = \pi_i \pi_f,$$

and the spin selection rule

$$|I_i - I_f| \leq J \leq |I_i + I_f|,$$

$$|L' - L| \leq J \leq |L' + L|.$$  

In Eq. 2.3 the sum runs over all intermediate states $|I_n\rangle$ which can be connected by the transition operators $M(SL)$ to the initial and final states obeying electromagnetic selection rules. Hence the two-photon decay probability is a new integral quantity of the nucleus allowing to study special structural properties of the nuclear system. 

The total two-photon decay probability is obtained after integrating Eq. 2.2 over the relative angle of the two photons and their energies

$$\Gamma_{\gamma\gamma} = \frac{1}{2} \int_0^{\theta_0} \int_{-1}^{+1} \frac{d^2 \Gamma_{\gamma\gamma}}{d\omega d\omega'} d\omega d\omega' d\cos(\theta).$$

The factor $\frac{1}{2}$ is necessary to avoid double counting due to the bosonic character of photons [5].

In Ref. [5] the usefulness of this quantity was demonstrated for the special case of the two-photon decay of a $0^+_2 \rightarrow 0^+_1$ transition. It gave access to the electric dipole transition polarizability and magnetic dipole transition susceptibility which are not easily accessible through other experimental methods. The latter can be used to test the validity of $M1$-sum rules and to obtain information on the quenching factor of the $M1$-strengths [5].

According to Ref. [5] the magnetic dipole transition susceptibility is defined as

$$\chi^{12} = \chi_P^{12} + \chi_D^{12},$$

(2.7)
where $\chi_P^{12}$ and $\chi_D^{12}$ are the paramagnetic and diamagnetic transition susceptibilities (for a definition see Ref. [5]). The paramagnetic transition susceptibility is closely related to the diagonal paramagnetic susceptibility of the ground state [5]. Which is given through

$$\chi_P = \frac{4}{9}\pi \cdot 2 \sum_n \frac{\langle 0^+|\langle M(1)|n^- \rangle^2}{E_n}.$$ (2.8)

$\chi_P$ can be determined in inelastic electron scattering or in inelastic proton scattering experiments under zero degrees [9]. If one is able to measure $\chi^{12}$ with sufficient accuracy one can combine the results of both experiments to determine $\chi_D^{12}$ for the first time. This would provide a direct test of the ‘seagull’ operator [5] which is closely related to $\chi_D^{12}$ and give valuable information on the importance of mesonic degrees of freedom.

Although the discussion in Ref. [5] is limited to the $0^+_2 \rightarrow 0^+_1$ case one can do the same kind of investigations for other transitions like $2^+_1 \rightarrow 0^+_1$. Clearly, this requires that one finds experimentally a way to deal with the—then not forbidden—one-photon decay.

The diagonal electric dipole polarizability of the ground state is defined as [5, 10]

$$\alpha_{E1} = \frac{4}{9}\pi \cdot 2 \sum_n \frac{\langle 0^+_1|\langle iM(E1)|1^-_n \rangle^2}{E_n}. \quad (2.9)$$

It is used e.g. to restrict the parameters of the equation of state [11] or to determine the neutron skin thickness [12]. It is thinkable that one uses the electric dipole transition polarizability obtained from the two-photon decay probability for the same investigations. Whether this is possible has to be answered by a detailed theoretical analysis. One problem might be that the contributions of the various intermediate states cancel in the sum in Eq. 2.3. Such a cancellation is not possible for the diagonal electric dipole polarizability.

In general the idea to obtain a similar integral quantity like the diagonal electric dipole polarizability and the diagonal paramagnetic dipole susceptibility just from a transition between two low-lying states—as it is the case for the two-photon decay—is very appealing. To identify all states of one multipolarity and parity over the full energy range is experimentally very demanding and one can easily miss a part of the strength. Due to the sum over all intermediate states in Eq. 2.3 this is not possible in case of the two-photon decay probability.

### 2.2 The Case of $^{137}$Ba

In the following, the general results of the last section are applied to the case of $^{137}$Ba. In order to keep the discussion transparent a simple nuclear structure model is employed.
The following results are based on the work of D.J. Millener who calculated the relevant observables for the two-photon decay in the independent particle model. The discussion follows closely his article in Ref. [6].

The $\beta^-$-decay of $^{137}$Cs populates the $11/2^-$ state in $^{137}$Ba which decays via a $M4$-transition and the emission of a single photon to the $3/2^+$ ground state. The two-photon decay proceeds through higher-lying intermediate states. According to the spin selection rule of Eq. 2.5 the allowed values for the total angular momentum transfer are $J = 4, 5, 6, 7$. In a very good approximation one can restrict the sum in Eq. 2.2 to $J = 4$ [13]. Furthermore the discussion is limited to the case where the multipolarities $L, L'$ sum to $J = 4$, i.e. $L + L' = 4$. Hence the considered transitions are $E1 + M3, M3 + E1, M1 + E3, E3 + M1, M2 + E2$ and $E2 + M2$. The quality of this assumption has still to be verified e.g. an appreciable contribution of the $E2 + E3$ term in Eq. 2.2 cannot be ruled out [13].

In the following the two-photon decay probability is estimated in the independent particle shell model. The nucleus $^{137}$Ba is one neutron away from the $N = 82$ shell closure. The $3/2^+$ ground state as well as the $11/2^-$ state at 662 keV are assumed to be neutron hole states with quantum numbers $2d_{3/2}^+$ and $1h_{11/2}^+$ respectively.

Due to the approximations made above the intermediate states have to have angular momentum $j = 5/2, 7/2$ or $9/2$. The configuration of the $1h_{11/2}^+$ state is schematically shown on the right-hand side of Fig. 2.2. Within the same major shell the neutron hole can be excited to the the $2d_{5/2}^+$ and $1g_{7/2}^+$ single particle states. In the next major shell there is the possibility to go to the $1g_{9/2}^+$ and $1f_{5/2}^-$ single particle states. On the other hand an excitation of a neutron from the $1h_{11/2}^+$ single-particle state to the $2d_{5/2}^+$ ground state through intermediate neutron hole states ($left-hand side$). On the $right hand side$ the neutron configuration in the independent particle model of the $1h_{11/2}^+$ state is shown. A transition through the $1g_{9/2}^+$ state is allowed (red arrow). On the other hand an excitation from a neutron to the $2f_{7/2}^-$ state is not possible (blue arrow), since there is no one-step deexcitation back to the $2d_{3/2}^+$ ground state. Figure on the $left hand side$ is taken, and slightly changed, from Ref. [6].

The excitation energies of the single-particle states are given in MeV.

![Fig. 2.2](image-url)
above the \( N = 82 \) shell closure to e.g. the \( 2f_{7/2} \) single particle state is not allowed in this simple model, since it is not possible to obtain again the \( 2d_{5/2}^+ \) ground state configuration in one step. Some of the possible paths for the two-photon decay in this simple model are presented in Fig. 2.2 with the corresponding multipolarities.

The denominator of Eq. 2.3 depends on the energy difference between the \( 1_{\gamma 1/2} \) single particle state and the corresponding intermediate single particle state. The \( 1g_{9/2}^+ \) and \( 1f_{5/2}^- \) hole states are higher in energy than the \( 2d_{5/2}^- \) and \( 1g_{7/2}^+ \) hole states since they belong to the next major shell. Hence, to a good approximation, one can neglect the \( E1 + M3 \) paths in Fig. 2.2 and consider the \( M2 + E2 \) and \( M1 + E3 \) multipole pairs only.

After integrating Eq. 2.2 over \( d(\cos \theta) \) one obtains—under the made approximations—for one pair of multipoles

\[
\frac{d\Gamma_{\gamma \gamma}}{d\omega} = \frac{2I_f + 1}{2I_i + 1} \frac{1}{32\pi} \frac{L + 1 L' + 1}{L L'} \left[ \frac{1}{(2L + 1)!!(2L' + 1)!!} \right]^2 \cdot \omega' \frac{\omega}{E_i - E_n - \omega} \left\{ \sum_n \omega L' \omega' \langle I_f | |i^{L-S'} M(S'L')||I_n\rangle \langle I_n||i^{L-S} M(SL)||I_i\rangle \right\}^2 \\
+ \left\{ \sum_n \omega L' \omega' \langle I_f | |i^{L-S'} M(S'L')||I_n\rangle \langle I_n||i^{L-S} M(SL)||I_i\rangle \right\}^2 \\
\left(2.10\right)
\]

In principle one can integrate Eq. 2.10 numerically over \( d\omega \) to obtain the two-photon decay probability \( \Gamma_{\gamma \gamma} \). However, in order to obtain a deeper insight in the two-photon decay the denominator is assumed to be independent of \( \omega \): \( \Delta E = E_i - E_n - \omega_0/2 \) which allows to write Eq. 2.10 as

\[
\frac{d\Gamma_{\gamma \gamma}}{d\omega} \sim \frac{\langle I_f | |i^{L-S'} M(S'L')||I_n\rangle^2 \langle I_n||i^{L-S} M(SL)||I_i\rangle^2}{\Delta E^2} \frac{\omega^2 + \omega^2 L' + 1 + \omega^2 + \omega^2 L' + 1}{\omega^2 L + \omega^2 L' + 1 + \omega^2 + \omega^2 L' + 1}.
\left(2.11\right)
\]

This approximation is valid if \( E_i - E_n \) is large compared to \( \omega_0 \). If one integrates Eq. 2.11 over \( d\omega \) and divides the result with the expression for the \( M4 \)-one-photon decay radiation width one obtains simple equations for the branching ratios in the \( M2 + E2 \) case [6]

\[
\frac{\Gamma_{\gamma \gamma}}{\Gamma_{\gamma}} = 4.58 \times 10^{-2} \cdot \frac{\omega_0}{\Delta E} \frac{B(M2; 1h_{11/2}^- \rightarrow 1g_{7/2}^+)}{B(M4; 1h_{11/2}^- \rightarrow 2d_{3/2}^+)} \cdot \frac{B(E2; 1g_{7/2}^+ \rightarrow 2d_{3/2}^+)}{B(M4; 1h_{11/2}^- \rightarrow 2d_{3/2}^+)}
\left(2.12\right)
\]

and for the \( M1 + E3 \) case

\[
\frac{\Gamma_{\gamma \gamma}}{\Gamma_{\gamma}} = 5.82 \times 10^{-2} \cdot \frac{\omega_0}{\Delta E} \frac{B(E3; 1h_{11/2}^- \rightarrow 2d_{5/2}^+)}{B(M4; 1h_{11/2}^- \rightarrow 2d_{3/2}^+)} \cdot \frac{B(M1; 2d_{5/2}^+ \rightarrow 2d_{3/2}^+)}{B(M4; 1h_{11/2}^- \rightarrow 2d_{3/2}^+)}
\left(2.13\right)
\]
Table 2.1 Two-photon decay branching ratio $\Gamma_{\gamma\gamma}/\Gamma_{\gamma}$ calculated in the independent particle model (IPM) according to Eqs. 2.12 and 2.13

<table>
<thead>
<tr>
<th></th>
<th>$M2 + E2$</th>
<th>$M1 + E3$</th>
<th>Full</th>
</tr>
</thead>
<tbody>
<tr>
<td>IPM</td>
<td>$1.28 \times 10^{-6}$</td>
<td>$0.78 \times 10^{-6}$</td>
<td>$2.06 \times 10^{-6}$</td>
</tr>
</tbody>
</table>

In order to determine the quadrupole-quadrupole and dipole-octupole branching ratios one has to know the energy differences $\Delta E$ between the $1h_{11/2}$ single particle states and the $2d_{5/2}^+$ and $1g_{7/2}^-$ single particle states. The energies of the $1h_{11/2}$ and $1g_{7/2}^+$ single particle states can be determined approximately from $^{138}\text{Ba}(p, d)^{137}\text{Ba}$ data [14] and amount to 0.66 MeV and 2.97 MeV respectively. The energy of the $2d_{5/2}^+$ orbital at 2.22 MeV is taken from Ref. [15].

The $B(M4 : 1h_{11/2}^- \rightarrow 2d_{5/2}^+)$-value of $8.89 \times 10^4 \mu_N^2\text{fm}^6$ [6] is taken from experiment. The other transition strengths are calculated using harmonic oscillator wave functions. The effective charges are $e_n = 1.0$ for $E2$- and $E3$-transitions. For the $M1$-transition the effective operator of Ref. [16] was used and for the $M2$-transition the spin g-factor was assumed to be $g_{s}^{\text{eff}} = 0.7g_{s}^{\text{free}}$. More details are given in Ref. [6].

The resulting branching ratios in this simple independent particle model are given in Table 2.1. The two-photon decay branching ratio is small and amounts to $2.06 \times 10^{-6}$. It is experimentally very demanding to measure a decay mode which has such a small branching ratio. Furthermore the contributions of the quadrupole-quadrupole and dipole-octupole multipole pairs are predicted to be similar in size.

In order to understand the two-photon decay it is desirable to obtain information on the contributing multipole pairs. From the $Q$ dependent factors in Eq. 2.2 one can obtain the angular distribution of each multipole pair. For the quadrupole-quadrupole pair the angular distribution is given through

$$W(\theta) = 1 + \frac{5}{49} P_2(\cos \theta) + \frac{40}{441} P_4(\cos \theta),\quad (2.14)$$

and for the dipole-octupole case

$$W(\theta) = 1 - \frac{1}{8} P_2(\cos \theta).\quad (2.15)$$

The angular distributions for pure multipole pairs and fixed $\omega$ are presented on the left hand side of Fig. 2.3.

The variation of $W(\theta)$ with the relative angle between both emitted photons is rather weak. However this angular distribution can be more pronounced for other multipole pairs. Furthermore interference effects can alter the angular distribution significantly.

Another way to obtain information on multipole pairs is to investigate the dependence of $\Gamma_{\gamma\gamma}/d\omega$ on the photon energy $\omega$. According to Eq. 2.11 $d\Gamma_{\gamma\gamma}/d\omega \sim \omega^5\omega^5$ in case of quadrupole-quadrupole and $d\Gamma_{\gamma\gamma}/d\omega \sim \omega^3\omega^7 + \omega^7\omega^3$ in case of dipole-octupole. Both distributions are shown on the right hand side of Fig. 2.3 assuming...
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Fig. 2.3  The dependence of the two-photon decay probability on the relative angle between the two emitted photons (left-hand side) and on the photon energy $\omega$ (right-hand side). The distributions are shown for the quadrupole-quadrupole (red) and dipole-octupole (blue) multipole pairs.

$\omega + \omega' = 662$ keV. In Eq. 2.11 the denominator is assumed to be independent of $\omega$. If the energy difference of the corresponding single particle states $\Delta E$ is small (relative to $\omega_0$) this assumption is not justified and the distributions in Fig. 2.3 will slightly change [13].

The considerations in the independent particle model allow to draw some general conclusions on the design of an experiment to measure the two-photon decay branch.

- The two-photon decay branching ratio is of the order of $\sim 2 \times 10^{-6}$ i.e. background and Compton scattering of photons stemming from the one-photon decay (see Sect. 3.1) have to be suppressed sufficiently to measure a decay branch of this order of magnitude.
- It is important to obtain information on the angular distribution and energy sharing function to decide which multipole pairs contribute to the two-photon decay.
- The present experiment was conducted with five LaBr$_3$-detectors with a distance of 22 cm to the source. The source strength was 606 kBq (October 5th 2013). With a simulation of the full energy efficiency of the LaBr$_3$-detectors one obtains a rate of 3.4 two-photon events per detector pair and day. For simplicity the full energy efficiency at 331 keV was used for this rate estimate taken from a Monte-Carlo simulation.

The values in Table 2.1 are just rough estimates for the two-photon decay probabilities. The used model is simple and is restricted to two decay paths. A calculation in a more advanced nuclear structure model like the Quasi-Particle-Phonon model (see Sect. 5.3) is clearly required. This would also allow to study the influence of the giant resonances on the two-photon decay probability which can significantly alter the results in terms of the contribution multipole pairs as well as the absolute value of the two-photon decay branching ratio.
References

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