The performance of NaI(Tl) scintillation detectors
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For determining the decay rate of radioactive sources in a modulation experiment.

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Abstract

This master thesis reports on the performance of the experimental setup for the Modulation experiment. The goal of this experiment is to falsify a claim that the activity of several radioactive sources modulates with an amplitude of 0.1%, a period of a year and a maximum in February. Some[1] scientists claim that these modulations are caused by the change in flux of solar neutrinos. An international Modulation collaboration will therefore measure the activity of four sources with NaI(Tl) detectors on four different locations for at least a year.

The performance of NaI(Tl) detectors is presented. The Photo Multiplier Tubes(PMT) reach a gain of $10^5$ and $10^6$ while operated between 500 and 1000 V. An energy resolution of 4.83-5.07(5) %, 7.29(5) % and 13.8(1) % is achieved for $^{60}$Co, $^{137}$Cs and $^{133}$Ba, respectively. The optimal operating voltages for the PMT’s are set to achieve the best energy resolutions. A total detection efficiency for $\gamma$-rays from $^{60}$Co, $^{137}$Cs and $^{133}$Ba decays ranges between 30 and 60% for a set of detectors. Using a set of two detectors reduces rate changes to 0.9 ± 0.04 % for shifts in the position of the source. After calibration the energy of the incoming $\gamma$-rays are identified within 0.2% accuracy. A 5 cm lead enclosure lowers the background rate from natural and cosmic origin to 20 Hz over the full spectrum.

To test the experimental setup, the activity of a $^{60}$Co source has been continuously measured over periods of 127 and 149 hours with a period of 37.5 days in between. The expected activity is compared to the measured activity and a 1 and 0.5 % lower detection rate is found for the total spectrum and peak region, respectively. A shift in voltage, a shifted source position and a change in background rate are investigated as possible causes. To suppress these effects improvements to the experimental setup are proposed.
Outline

Chapter one starts with a short history on and an explanation of the working of radiation. It is followed by the experimental results from two different institutes which are presented with both the original and new (controversial) explanations of the origin of a measured modulation. In chapter two the goal and setup of the modulation experiment, the working of scintillation crystals and PMTs and the signal processing with resulting spectrum are described. Chapter three focuses on the $\gamma$-ray interactions with special attention to the photoelectric effect, Compton scattering and pair production. In chapter four the results on the performance of the detectors (Gain, energy resolution, detector efficiency, geometric rate effects, energy calibration and background rate) can be found. A first test of the whole experimental setup is found in chapter five where the results on the activity of a $^{60}$Co source are shown. Chapter six concludes with the results and a discussion on possible improvements on the experimental setup. The Appendix contains extra figures as referenced in the other chapters.

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Chapter 1

Modulations in radioactive decay

In this chapter experimental results are described showing a modulation in the activity of several different radioactive isotopes. First a historical overview of how radioactivity was discovered is given with a short introduction into the theory of radioactivity.

1.1 Radiation

To understand what could cause a modulation in the activity of a radioactive source one first needs to understand the underlying process of radiation. In this section the discovery and the working process of radioactive decay is described. The goal here is to get a better understanding of how one classifies a decay and how certain we are of the accepted theory of radiation.

1.1.1 A short history of radiation

Radioactivity was first discovery by Henri Becquerel in 1896. His idea of testing the effect of Uranium salts on photographic plates was inspired by Röntgen’s discovery of Radiation (only) two and a halve months earlier. Wilhelm Conrad Röntgen was, like many physicists in his day, working with cathode ray tubes. In these vacuum tubes two electrodes where placed, by applying a voltage difference over them a glow could be seen coming from the anode and going into the cathode. When cathode rays (electrons) had enough energy and where directed towards the end the glass would exhibit fluorescence. It was demonstrated that these rays could not penetrate deep into a material. On the evening of November 8, 1895, Wilhelm darkened the
room, enclosed the tube in black cardboard so that no light or cathode rays could escape. Behind the cathode ray a plate covered with a layer of barium-platinum cyanide was placed. When he turned the cathode ray tube on he noticed that the screen gave off a fluorescent light. Since no cathode rays (electrons) or photons could have reached the screen he concluded that some other rays must travel from the tube to the screen. He then placed several different objects between the tube and the screen and found that his newly found 'Röntgen rays'(X-rays) still reached the screen and made it exhibit fluorescence. His saw his final proof when he placed his hand in front of the screen and saw a picture of his own bones, see figure 1.1.

![Figure 1.1: One of the first pictures Röntgen made of his own hand.][2]

Röntgen’s paper, including the first X-ray of a hand, reached Becquerel on January 20th, 1896. He immediately began testing the effect of other fluorescent materials on photographic plates to see if these also radiated the newly discovered 'X-rays'. His first success came when he placed a photographic plate, wrapped in black paper, with uranium salt on it in the sun for a day. After developing the plate he noticed that there was a light silhouette where the salt had been. Because he thought the sun was activating these rays and it was cloudy on the 26th and 27th on February he did not repeat his experiment. So he stored a new plate with uranium salt in his desk for later use. On the 1st of March, 1896, he decided to develop this ‘unused’ plate and discovered[3] that the markings where even brighter then the previous one, see figure 1.2. This was the discovery of the first radioactive process which could send out rays that could penetrate normal materials. Röntgen got the first Nobel prize in 1901 for discovering radiation while Becquerel (together with Pierre and Madam Curie) got his Nobel prize in 1903 for his
discovery of radioactivity. In 1902 Rutherford and Soddy discovered the exponential decay law (Equation 1.4). Since their discovery scientists believe that radioactivity is a well understood and in nature a random process. This is thus in clear contrast to the belief of a few physicists[1] who claim that this process is in fact not random but is influenced by external forces.

Figure 1.2: Photographic plate made by Becquerel showing the effects of exposure to radioactivity. One can clearly see a visible shadow on the photographic plate made by a metal Maltese cross that was accidentally placed between the plate and radioactive uranium salt. Made by H. Becquerel in 1896.[4]

1.1.2 Radiation from decays

The three most prominent radioactive decay types are $\alpha$ decay (which emits helium nucleus), $\beta$ decay (which emits/captures an electron) and $\gamma$ decay (which emits photons). The focus on this thesis lies on $\beta$ since no modulation has been found with alpha decays [5]. The sources used - $\gamma$ emitters - undergo $\beta$ decay or electron capture after which the nucleus sends out an $\gamma$-ray which can be detected. In equations 1.1 & 1.2 the process is shown for electron capture and beta decay, respectively. The sources $^{60}\text{Co}$ and $^{137}\text{Cs}$ decays by beta decay while $^{133}\text{Ba}$ decays by electron capture.

$$p^+ + e^- \rightarrow n + \nu_e$$  \hfill (1.1)

$$n \rightarrow p^+ + e^- + \bar{\nu}_e$$  \hfill (1.2)
The two forces that keep the nucleus together are the Coulomb interaction and the strong force. For two protons, which have a charge of +1, the Coulomb interaction pushing them away from each other is dependent on the distance between them:

\[ F = K_e \frac{Q_1 Q_2}{r^2} \]  

(1.3)

Here \( K_e \) is Coulomb’s constant, \( Q \) is the charge of the particles and \( r \) is the distance between the charged particles. The size of a typical atom is in first approximation proportional to the number of nucleons (\( A \)), these each take up a volume of around \( V_0 \simeq 7 \text{fm} \). The total size of the nucleus can thus be approximated as a sphere with a radius of \( R \simeq r_0 A^{2/3} \text{fm} \), with \( r_0 = 1.2 [6] \).

The size of the atom, which is defined by the extent of the electron cloud, is not proportional to \( A \) and is in the order of 0.1 to 0.3 nm. The Coulomb repulsion between two protons will force protons away while the strong force binds them together. The strong force works the same for protons as for neutrons (ignoring spin for the moment). This force is attractive between two nucleons which are separated more than 0.7 fm and less than 2 fm (center to center). For smaller separations the force repels them from colliding into each other while for larger separations the attraction vanishes (see figure 1.3a).

![Diagram](http://www.chem1.com/)

(a) Force between nucleons for the coulomb (top, red) and strong force (bottom, blue). The typical nucleon-nucleon separation distance is the point where the potential energy for the nucleon is lowest. Figure from http://www.chem1.com/

(b) Feynman diagram of β decay. The \( \bar{\nu}_e \) traveling backwards in the time line is the same as the \( \nu_e \) going forward in time.

Figure 1.3

One defines a nuclear unstable atom as one that can spontaneously decay. The heavier the atom is the more the Coulomb force grows in comparison
to the strong force. This phenomenon is explained by the fact that the protons at each end of the nucleus repel each other over large distances while the strong force can only bind the nucleons that are next to each other. By looking at the binding energy per nucleon one sees that after a certain size ($^{56}$Fe) of the atom the energy per added nucleus decreases. The atom becomes less stable since the total coulomb repulsion grows bigger than the total attraction of the strong force. For this unstable state only a small disturbance from outside will be enough to let it transition into a lower energy state. For an excited atomic nucleus the small disturbance can be from random quantum vacuum fluctuations. The transformation of the structure of the nucleus with the emission of either a photon($\gamma$) or a high-velocity particle that has mass ($\alpha$, $\beta$) is the outcome.

$\beta$ decay (or weak decay, see figure 1.3b) is the primary decay for atoms which have a low proton/neutron ratios. For electron capture, the electron (normally from its own k shell) is captured by the proton(e.q. 1.2). This process cannot happen for a free proton since the mass of the neutron (939.566 MeV) is higher than that of the proton (938.272). In $\beta$ decay the nucleus is excited by the freed energy, the nucleus can then release this energy by emitting a $\gamma$-ray. With electron capture the vacancy in the electron shell is filled by another electron which can also sends out a $\gamma$-ray. Since the energy of the parent and daughter states are well known, the precise energy of the detected outgoing $\gamma$-ray tells us which decay it came from.

The decay of an instable atom is a random process which can be described by the nuclear decay constant $\lambda$. From this constant the other nuclear parameters can be derived such as the half-life $t_{1/2} = \frac{\ln(2)}{\lambda}$ and the mean life time $\tau = \frac{1}{\lambda}$. Further more any radioactive source has

$$N(t) = N_0 e^{-\lambda t}$$

active atoms, where $N_0$ is the amount of active nuclei at $t=0$ and has an activity of $A(t) = N(t)\lambda$.

1.2 The nuclear decay constant $\lambda$

This section gives a review of available publications on the nuclear decay constant($\lambda$). Special attention is given to the unexpected conclusions on yearly modulating activities which point to a time depended decay constant. For those findings the modulation amplitude, modulation period and modulation phase are given. Even though the analyzed data comes from older measurements[7] [8], the conclusion that the activity is changed by solar neutrinos comes from newer publications from Jenkins and Fishbach[9] [5] [1].
1.2.1 Experimental results for several isotopes from PTB

Since 1989 the activity of several different radioactive sources has been monitored by the National Bureau of Standards group at the Physikalisch-Technische Bundesanstalt (PTB). Most measurements have been done with a pressured 4π ionization chamber (IG12/A20, Centronic 20th Century Electronics, Ltd.). The ionization current from the ionization chamber was measured by the Townsend balance-of-charge method[10]. The primary goal of the experiments was to measure the half life time of the following radioactive isotopes: \(^{85}\)Kr, \(^{90}\)Sr, \(^{108}\)Ag, \(^{133}\)Ba, \(^{137}\)Cs (from 1998), \(^{152}\)Eu, \(^{154}\)Eu. As a reference source \(^{226}\)Ra (\(T_{1/2} = 1635\)y) was used and all sources were measured at nearly the same time for all corresponding data points (Schötzig et al., 1992[7]; Siegert et al., 1998[8]; Schrader, 2004[11]).

To look for systematic errors in the measurements, a plot was made to depict the residual activity of an isotope. The residual defined as:

\[
R = A(t)_{\text{expected}} - A(t)_{\text{measured}} = -N_0 \cdot e^{\lambda t} - N(t)_{\text{measured}}
\]

(1.5)

here the activity \(A = N\lambda\), the decay constant is \(\lambda(t) = \lambda\), time is \(t\) and the number of active nuclei is \(N_0\). The residuals for the \(^{226}\)Ra reference source for a period of over 10 years are plotted in figure 1.4a (first published by Siegert et al., 1998[8]). An oscillation is seen with peaks in the order of \(N_{\text{mod}} \approx 0.15\%\) and with a period of a year (\(\omega_{\text{mod}} = 1\) y\(^{-1}\)). The maximum positive deviation occurs around February and the maximum negative deviation occurs around August (phase is \(\phi_{\text{mod}} = 1.44\) months, ref data: 1 Jan). A similar fluctuation is seen in the residuals of the \(^{154}\)Eu source (figure 1.5a). To compare the two modulations the residuals of the ratio of \(^{154}\)Eu/\(^{226}\)Ra are shown in figure 1.4b. No periodic fluctuation is seen in this figure which means they have identical amplitude and phase. The authors state that the oscillation may be explained by a discharge effect on the charge collecting capacitor, the cables and the insulator to the ionization chamber electrode caused by background radioactivity such as radon and daughter products which are known to show seasonal concentration changes[8].

Because the values of the life times at PTB are 0.4% lower compared to others the residuals for \(^{85}\)Kr, \(^{108}\)Ag, \(^{152}\)Eu and \(^{154}\)Eu where analyzed by Schrader et al., 2010[12]. In figure 1.5a the residuals on \(^{154}\)Eu (\(A_{\text{mod}} \approx 0.1\%, \omega_{\text{mod}} \approx 1\) y\(^{-1}\), \(\phi_{\text{mod}} \approx 0.12\)) are shown for the period of 1990 to 1996 (see figure 7.1 in Appendix A for all 4 residuals). Looking at the same source for the period of 1999 to 2009, as can be seen in figure 1.5b, the yearly fluctuations cannot be discerned anymore for \(^{154}\)Eu. The difference can be found in the fact that in October 1998 the Townsend measuring electronics
(a) "Residuals of the ionization chamber measurement data of $^{226}$Ra as a function of time from a fit with an exponential decay function. The vertical dotted lines are positioned at 1st January at an interval of 10 years. ($\lambda_{\text{mod}} \approx 0.15\%, \omega_{\text{mod}} \approx 1y^{-1}, \phi_{\text{mod}} \approx 30d$)

Figure 1.4: "The modulation is seen in the individual data, but disappears when one takes the ratio of these two sources. A data point is an average of the ratio of sample to reference source currents, containing about 30 individual sample measurements taken over 3 days and corrected for background." Figure and text from[8]

were replaced by a Keithley electrometer(model 6571A). This divides the data into two periods: 1990-1995 with the Townsend method and 1999-2008 with two identical Keithley electrometers. The authors state that a direct interaction on the corresponding nuclear decay rates does not take place, an effect of interaction related to the Earth-Sun distance can be excluded simply by the fact that the yearly fluctuations disappear or completely change their structure in the case that the current measuring technique is changed, for example, by using a Keithley electrometer instead of a Townsend balance current measuring method. If there is an effect on the PTB measuring techniques for half-lives which induces yearly fluctuations it must interact with the measuring electronics.[12]

In the described publications of the PTB data no other explanation for the modulation is given other than background or electronics, but this changes with the publication by Jenkins et al., 2012[5]. They show a power-spectrum analysis of the $^{137}$Cs, $^{133}$Ba and $^{226}$Ra PTB data for the possibility of intermittent oscillations. For the $^{137}$Cs data there is no evidence of an annual period for the period of 2002 to 2006 (see figure 1.6a). The spectrogram for the $^{226}$Ra reference source does show a modulation (see figure 1.6b). If the modulation on the $^{226}$Ra has an origin in natural background, in electronics or in any other factor that is not source-related, the same oscillation is expected in the $^{137}$Cs signal. The spectrograms in figure 1.6 do not show the
(a) Residuals of $^{154}\text{Eu}$ for the period of 1990 to 1995 (Townsend electrometer). ($A_{\text{mod}} \approx 0.1\%$, $\omega_{\text{mod}} \approx 1y^{-1}$, $\phi_{\text{mod}} \approx 0.12$) The decrease in signal is caused by small continuous decrease of the detector efficiency.

(b) Residuals of $^{154}\text{Eu}$ for the period of 1999 to 2009 (Keithley electrometer). No modulation with a $A_{\text{mod}} \approx 0.1\%$ is seen.

Figure 1.5: Residuals of fits of current raw data for $^{154}\text{Eu}$. Data is corrected for background and source impurities. A yearly fluctuation is seen in the first period but is not seen in the second period. Between these two periods the electronics that where used to measure the output current of the ionization chamber where changed. Figures from [12]

same oscillation for the period of 2002 to 2006. If these two sources have been measured and analyzed using the exact same procedure the effect must be source dependent. This is in clear contradiction to the statement that the yearly fluctuations must interact on the measuring electronics as stated in Schrader et al., 2010 [12]. The spectrogram for $^{133}\text{Ba}$ also shows evidence of oscillation periods with: $\omega_{\text{mod}} \approx 1y^{-1}0.5y^{-1}$, $\phi_{\text{mod}} \approx 0.12$ (see figure 7.3a in appendix A).

1.2.2 Experimental results for $^{32}\text{Si}/^{36}\text{Cl}$ from BNL

Between 1982 and 1986 the half-life of $^{32}\text{Si}$ was measured by Alburger et al., (1986)[13] at Brookhaven National Laboratory(BNL). The direct counting rate was measured as a function of time for both $^{32}\text{Si}$ and $^{36}\text{Cl}$ ($T_{1/2} = 301.000$ y), of which the last one functioned as a reference source. Both sources where counted for 30 minutes alternately with a precision sample changer (Harbottle et al., 1973[14] ), 10 times each. Those 10 counts combined to get the a data point of the ratio between $^{32}\text{Si}$ and $^{36}\text{Cl}$. For every week 3 full days of data where taken. By calculating the ratio of $^{32}\text{Si}/^{36}\text{Cl}$ systematic effects that would effect the electronics would largely be canceled out. They observed small periodic annual deviations of the data points from an exponential decay curve, but also state that they are of uncertain origin and had no significant
(a) Spectrogram of $^{137}$Cs. There is only slight suggestion of an annual oscillation from 2002 to 2003.

(b) Spectrogram of $^{226}$Ra. There is evidence of an annual oscillation from 2002 to 2005. This is the same source as seen in 1.4a

**Figure 1.6:** In color. Spectrograms of measurements of the decay-rates made at PTB over the time interval June 1999 to November 2008. The power, $S$, is represented by the colour bar.[5]

effect on the result.

A completely different conclusion is drawn in the paper of Fischbach et al. (2009)[1] where this BNL data is compared with respect to the Earth-Sun distance, see figure 1.7. The fluctuation shows $A_{\text{mod}} \approx 0.15\%$, $\Omega_{\text{mod}} \approx 1\text{yr}^{-1}$, $\phi_{\text{mod}} \approx 0.12$. Fischbach et al., 2009[1] found a correlation coefficient between the BNL data and $1/R^2$ (here $R$ is the distance between the Earth and the Sun) that is $r = 0.52$ for $N = 239$ points. They conclude that the formal probability that the indicated correlation could have arisen from uncorrelated data sets is $6 \times 10^{-18}$. This still does not mean there is a causal relation, for example a correlation can also be found between averaged monthly temperature and the (yearly) modulation. They state that the modulation in the activities could be caused by the change in flux from solar neutrinos.

### 1.2.3 List of different isotopes with modulation effect

During the last few decades different experiments have measured the decay of radioactive isotopes. Table 7.1 in the appendix shows an overview of isotopes which have shown to exhibit modulation in their decay. This shows not only isotopes that exhibit annual variations, but also sub-annual periodicities. The annual periods can be easily attributed to a seasonal influence with a clear annual variation, such as temperature. This cannot be done as easily with periodicities on the order of six months, one month, or less.[5]. Jenkins et al.[5], in light of Table 7.1, conclude the following:
Figure 1.7: "Plot of U(t) for the raw BNL $^{32}$Si/$^{36}$Cl ratio along with 1/R² where R is the Earth-Sun distance. U(t) is obtained by multiplying each data point by exp(+$\lambda$t) where $\lambda$ = ln(2)/T₁/₂ and T₁/₂ = 172 y for $^{32}$Si. The left axis gives the scale for the normalized U(t), and the right axis denotes the values of 1/R² in units of 1/(a.u.)² obtained from the U.S. Naval Observatory (USNO). The fractional change in $^{32}$Si counting rates between perihelion and aphelion is approximately 3 x 10⁻³. " Figure and text from [1]

1. Not all nuclide’s exhibit variability in decay constants
2. Among nuclide’s that do exhibit this variability, the patterns of variability (e.g., amplitude and phase of any oscillation) are not all the same
3. For nuclide’s that do exhibit variability, the patterns themselves may vary over time

1.2.4 Experimental results for $^{137}$Cs at Gran Sasso

At the time of writing the results of one dedicated modulation experiment is available. Starting in June 2011 the activity of a $^{137}$Cs source has been measured with a HPGe detector by Bellotti et al., (2012)[15]. The experiment is set up deep underground in the Gran Sasso Laboratory to reduce background influences. The cosmic ray flux and neutron flux is reduced by a factor $10^6$ and $10^3$ respectively. The temperature does not change more than 0.7K during a 7 month period of data acquisition. With the source being firmly fixed any changes in detector-source distance are reduced. From Monte
Carlo Simulations they conclude that a variation in the source-detector distance of 1 micron would result in a variation of the counting efficiency of \(5 \times 10^{-5}\). The background rate is reduced to 40 counts per hour above the 7 keV threshold (0.01Hz). Data has been collected during 217 days, spectra are collected every hour and the dead time stays around 5%. In Figure 1.8 the results are shown. At the bottom of the figure the residuals are shown, it is clear that no deviations above or below 0.001% have been measured.

![Figure 1.8: Detected activity of the \(^{137}\)Cs source. Dead-time corrected data are summed over 96 hours. The first two points correspond to the beginning of data taking, when the set-up was stabilizing, and they are not considered in the analysis. Dotted lines represent a 0.1% deviation from the exponential trend. Residuals (lower panel) of the measured activity to the exponential fit. Error bars include statistical uncertainties and fluctuations in the measured dead time." Figure and text from Bellotti et al., (2012)](image)

No significant improvement to the \(\chi^2\) fit is observed for fits with periods between 6 hours to 400 days. The analysis excludes any oscillation with amplitude larger than \(9.6 \cdot 10^{-5}\) at 95% C.L.. In particular, for an oscillation period of 1 year the amplitude is \(3.1(2.7) \cdot 10^{-5}\), which is well compatible with zero, and a limit of \(8.5 \cdot 10^{-5}\) at 95% C.L. on the maximum allowed amplitude is set independently of the phase for \(^{137}\)Cs.
1.3 Experimental results for the effect of solar flares on radioactive decay

During four different solar flares the decay rate of several different radioactive isotopes have been measured. One publication[9] show a correlation between the solar flare and an decrease of the decay rate of $^{54}$Mn while other publications show no effect for $^{40}$K, $^{137}$Cs and $^{nat}$Th[16].

Solar flares are huge and sudden eruptions of energy from the sun that occur on irregular intervals and are hard to predict. One solar flare can release up to $6 \times 10^{25}$ joules, which is equivalent to 1/6th of the total energy the Sun normally releases per second. This energy is released within minutes to tens of minutes and can send a plasma consisting primarily of electrons and protons towards Earth(Coronal mass ejection). The effect on Earth is largely reduced by the Magnetic field of the Earth and the outer atmosphere, still power cuts, radio blackout and aurora’s are reported after strong solar flares in the direction of Earth. The Sun goes through a solar cycle of 11 years with changing magnetic activity. It has to be noted that in the fall of 2013 the solar cycle is at its high point. One could expect a change in the solar neutrino flux on Earth produced by for example secondary pions from the flare that decay into electrons and electron neutrino’s. Even though a relation between Solar flares and an increase in the solar Neutrino flux has been postulated, it has not yet been observed[17].

In December 2006 two solar flares where detected by satellites by a sudden increase in high energy protons and X-rays. During these solar flares a decrease in the decay rate of $^{54}$Mn was reported by Jenkins en Fischbach[9]. A 1 µCi sample of $^{54}$Mn was monitored by a NaI(Tl) scintillator detector, shielded by lead, in an air conditioned lab at Purdue University. In each 4 hour live-time period ($\sim 4.25$ hours real-time) $\sim 2 \times 10^7$ 834.8 keV $\gamma$-rays where recorded (uncertainty per data point: $\sim 2 \times 10^{-4}$). In figure 1.9 the decay is plotted together with the expected exponential decay of $N(t) = N_0e^{-\lambda t}$, $\lambda = 0.002347(2)d^{-1}$. The counting rates show a dip corresponding with the solar flares on 13 December at 02:37 UT (21:37 EST on 12/12/06) and 17 December at 12:40 EST. Dips in the counting rate can also be observed around 14/12/16 and 22/12/06, the first has a corresponding X-ray detection while the second corresponded with solar storms. The spikes in X-ray and proton fluxes where seen[18] by all of the Geostationary Operational Environmental Satellites(GOES), the X-rays peaks from the GOES-11 is also shown in figure 1.9.

To see if the decrease in the counting rate could be explained by influences on the electronics the detectors setup have been exposed to magnetic
Figure 1.9: "December 2006 $^{54}$Mn data, and GOES-11 x-ray data, both plotted on a logarithmic scale. For $^{54}$Mn, each point represents the natural logarithm of the number of counts $\sim 2.5 \times 10^7$ in the subsequent 4 hour period, and has a $\frac{1}{\sqrt{N}}$ statistical error shown by the indicated error bar. For the GOES-11 x-ray data, each point is the solar x-ray flux in W/m² summed over the same real time intervals as the corresponding decay data. The solid line is a fit to the $^{54}$Mn data. The dates for other solar events are also shown by arrows." Figure and text from Jenkins et al., (2008)[9]

fields to twice that of the Earth’s magnetic field ($\sim 0.85$Gauss). No voltage differences outside the range 115-126V were occurring and the temperature was a constant 19.5(5)°C in the windowless lab. They state that the most compelling argument against this explanation of the $^{54}$Mn data is that the $^{54}$Mn decay rate began to decrease more than one day before any signal was detected in x-rays by the GOES satellites. The decrease in counting rate during the solar flares was measured to be below 0.015% which can be seen in figure 7.4 in appendix A.
Chapter 2
Gamma-ray interactions in matter.

For the measurement of the rate of decay of a radioactive source a good understanding of gamma-ray interactions in the scintillation crystal is essential. The focus of this chapter is on $\gamma$ interactions. For $\gamma$-rays below 10 MeV the three most prominent interactions are the Photoelectric absorption, Compton scattering and Pair production. The following paragraphs describe how the $\gamma$-ray interacts with the medium and how excited electrons and newly formed photons emerge.

2.1 Beer’s Law

The chance of an interaction between the incoming $\gamma$-ray and the material in our detector depends on several different variables. When one increase the dimensions of the detector it increases, it also increases when one increases the Z-value of the detection material. For higher energy incoming photon energies this chance of interaction decreases. To get a relation between the chance of detection and the cross section one can imagine the decrease in intensity $dI$ of a beam of particles traveling over a small distance $dx$ through a medium as:

$$dI = I(x + dx) - I(x) \equiv -\mu I(x)dx$$

(2.1)

Here $\mu$ is introduced as the attenuation or absorption coefficient. The larger $\mu$ is the more the beam gets attenuated (weakened) as it travels through the medium, this is caused by a higher probability of interaction between the photon and the material. Integrating the above equation from an initial intensity $I_0$ at $x = 0$ to a final intensity $I(x)$ at point $x$ one gets Beer’s law.
(also known as the Beer-Lambert-Bouguer law):

\[ I(x) = I_0e^{-\mu x} \]  

(2.2)

The total attenuation coefficient \( \mu \) for a \( \gamma \)-ray is a summation of the three separate coefficients (using the same notation as in figure 2.4):

\[ \mu = (\tau_{\text{Photo}} + \sigma_{\text{Compton}} + \kappa_{\text{Pair}})n \]  

(2.3)

The total cross section for an interaction and the attenuation coefficient are related as followed:

\[ \frac{dI}{I} = \frac{A_0}{A} \rho \sigma dx \]  

(2.4)

\[ \mu = \rho \frac{A_0}{A} \rho = n\sigma \]  

(2.5)

Here \( A \) is the atomic weight of the target, \( A_0 \) is Avagadro’s number of atoms per mole and \( \rho \) the density of the target material. Here one can argue that scattering an object from the beam drops the intensity of the beam, and thus the decrease in intensity of the beam is proportional to the cross section per nuclear scatterer (\( \sigma \)) and the thickness of the target material (\( dx \)). For a range of \( \gamma \)-rays energies, going through NaI(Tl), the total chance of interaction is shown in table 2.1. The relationship between the cross section, the Z-value and photon energy is different for all three types of interactions as seen in equations 2.6, 2.9 and 2.14. These equations result in the three different regions where one interaction dominates over the other two.

\section*{2.2 Photoelectric absorption}

Photoelectric absorption is the interaction between an incoming \( \gamma \)-ray and an atom of the detector material. The incoming photon is completely absorbed by the atom and the energy is transferred to an outgoing electron, from which shell the electron is excited depends on the energy. Electrons with an energy above 33 keV have the biggest chance to be excited from the K shell of the atom. The chances of exciting an electron from the L or M shell are shown in figure 7.5. The energy of the outgoing electron is the energy of the photon minus the energy of the electron binding energy.

\[ E_{e^-} = h\nu - E_b \]  

(2.6)

Here \( h \) is Planck’s constant and \( \nu \) is the frequency of the photon. The binding energies of the different shells of the iodine atom are shown in table 2.2.
In the NaI(Tl) crystals the iodine atoms are the primary absorbers. The vacancy in the electron shell can be filled in two ways. First the atom can capturing a free electron from the medium and secondly one of the electron in a higher electron shell can drop down to the vacant place the the lower shell. The potential energy gained by the lowering of the electron is released through the creation of an X-ray photon. These photons are mostly reabsorbed by the surrounding atoms which in turn excite new electrons. When an electron from a shell drops to a lower electron shell it is possible that instead of creating an X-ray the energy is transferred to another electron in that shell. This so called Auger electron is then excited from the atom. All these interactions together result in the deposited energy for the photoelectric absorption. This deposited energy is for Photoelectric absorption related to the total energy of the incoming photon.

For low energy gamma-rays the Photoelectric absorption is the dominant interaction. A rough approximation of the probability of photoelectric absorption per atom over all ranges of energy is given by[19]:

$$\tau \approx \text{constant} \cdot \frac{Z^n}{E_{\gamma}^{3.5}}$$

(2.7)

Here $n$ is a constant between 3 and 5 depending on the energy. In the NaI(Tl) crystal the photoelectric absorption is the dominant interaction for gamma-rays up to 400 keV. For a gamma-ray with an energy of around 400 keV (in NaI(Tl)) the probability of undergoing the photoelectric effect is equal to the chance it undergoes Compton scattering.

<table>
<thead>
<tr>
<th>Source</th>
<th>Energy(MeV)</th>
<th>$\mu_{\text{total}}$</th>
<th>$P_I$ (%)</th>
<th>$P_S$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>241Am</td>
<td>0.06</td>
<td>5</td>
<td>100.00</td>
<td>0.00</td>
</tr>
<tr>
<td>137Cs</td>
<td>0.66</td>
<td>0.07</td>
<td>85.81</td>
<td>14.19</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>0.06</td>
<td>81.24</td>
<td>18.76</td>
</tr>
<tr>
<td>60Co</td>
<td>1.17</td>
<td>0.055</td>
<td>78.43</td>
<td>21.57</td>
</tr>
<tr>
<td></td>
<td>1.33</td>
<td>0.05</td>
<td>75.21</td>
<td>24.79</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>0.04</td>
<td>67.23</td>
<td>32.77</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>0.035</td>
<td>62.33</td>
<td>37.67</td>
</tr>
</tbody>
</table>

Table 2.1: This table contains the chance of interaction $P_I$ and of escape without interaction $P_S$ for different photon energies. All $\mu$ values from figure 2.4 for NaI(Tl).

2.3 Compton Scattering

In Compton scattering the incoming photon interacts with a free or loosely bound electron and travels on afterwards. This results in the photon’s path
being altered with an angle $\theta$ as is shown in figure 2.1. The change in energy of the outgoing photon is dependent on $\theta$, the rest of the energy is transferred to the recoiling electron. The energy of the outgoing photon is given by:

$$h\nu' = \frac{h\nu}{1 + \frac{h\nu}{m_0c^2}(1 - \cos(\theta))}$$

(2.8)

Here $h\nu'$ is the energy of the outgoing photon and $m_0c^2$ is the rest mass of the electron (0.511 MeV). Since the photon loses a portion of its energy it can interact again and again with the detector material or leave it altogether on the other side. Any $\gamma$-ray that leaves the detector after interaction deposits a part of its total energy in the material. The chance of Compton scattering in the medium depends on both the energy of the incoming photon and the electron density. The higher the density of electrons ($Z$) in the absorbing material the higher the chance of Compton interaction, given by:

$$\sigma \propto \frac{Z}{E_\gamma}$$

(2.9)

In figure 2.4 one can see the range of energies in which the Compton interaction is dominant. Although the photon is totally absorbed for the photoelectric effect this is never the case for Compton scattering. The reason for this is that a free electron can not conserve both the energy and the momentum of the incoming photon on its own. The photon has zero mass, while the electron has a mass.
To understand this one could look at the energy in the hypothetical situation of total absorption of a photon by a free electron. One would find the following relation in the rest frame of the outgoing electron:

\[ E_\gamma + E_{e \text{ incoming}} = E_{e \text{ outgoing}} \]  \hspace{1cm} (2.10)

\[ E_\gamma + \sqrt{p_{e \text{ incoming}}^2 c^2 + m_e^2 c^4} = m_e c^2 \]  \hspace{1cm} (2.11)

\[ E_\gamma + \sqrt{E_\gamma^2 + m_e^2 c^4} > m_e c^2 \]  \hspace{1cm} (2.12)

The left side of this equation is always bigger than the right side of this equation for a photon momentum not equal to zero. So following conservation of energy and momentum there must always be a scattered photon in Compton scattering. With the photoelectric effect the photon can disappear since its momentum can be transferred to the system of the atom. The Angular distribution of scattering γ-rays for a differential cross section is predicted by the Klein-Nishina formula:

\[ \frac{d\sigma}{d\Omega} = Zr_0^2 \cdot \left( \frac{1}{1 + \alpha^2 (1 - \cos^2 \theta)} \right)^2 \cdot \left( \frac{1 + \cos \theta}{2} \right) \cdot \left( 1 + \frac{\alpha^2 (1 - \cos^2 \theta)}{(1 + \cos^2 \theta)(1 + \alpha (1 - \cos^2 \theta))} \right) \]  \hspace{1cm} (2.13)

Here \( \alpha = \frac{E_\gamma}{m_0 c^2} \) and \( r_0 \) is the classical e\(^-\) radius. For high gamma-ray energies there is a strong tendency to scatter in the forward direction. In Figure 2.2 the scatter angle dependence on the energy is shown in a polar plot.

### 2.4 Pair production

When a high energy γ-ray transfers its energy into the spontaneous creation of an electron-positron pair is called Pair production(figure 2.3). This phenomenon only happens when \( h \nu \) exceeds twice the creation energy of the electron(\( E_\gamma \geq 2M_e c^2 \approx 1.022 \text{ MeV} \)).

The conversion of a photon into the electron-positron pair does not conserve both energy and momentum without a third party involved. In the medium the photon can only disappear when it recoils to the nucleus. There the photon is subjected to the Coulomb field of the nucleus and can transfer a bit of its momentum to the system so that energy and momentum are conserved again in the final states. The excess photon energy above 1.022 MeV is converted into the kinetic energy of the electron-positron pair. Afterwards
Figure 2.2: Polar plot of the numbers of photons Compton scattered into a unit solid angle at the scattering angle $\theta$. The energies are shown in isobar lines with $\alpha = h\nu$ in MeV.

the positron will annihilate with an electron inside the detector medium into two annihilation photons. The pair production cross section scales as

$$\kappa_{\text{pair}} \propto Z^2$$

(2.14)

which is why its interaction probability dominates the other two possible interaction above 7 MeV in NaI(Tl) as can be seen in figure 2.4.
Figure 2.3: Schematic representation of pair production. The incoming photon interacts with the Coulomb field of the nucleus and disappears forming an electron-positron pair.

<table>
<thead>
<tr>
<th>Shell Label</th>
<th>Orbital</th>
<th>Binding energy (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>K</td>
<td>1s</td>
<td>33169</td>
</tr>
<tr>
<td>L I</td>
<td>2s</td>
<td>5188</td>
</tr>
<tr>
<td>L II</td>
<td>2p</td>
<td>4852</td>
</tr>
<tr>
<td>L III</td>
<td>2p3/2</td>
<td>4557</td>
</tr>
<tr>
<td>M I</td>
<td>3s</td>
<td>1072</td>
</tr>
<tr>
<td>M II</td>
<td>3p1/2</td>
<td>931</td>
</tr>
<tr>
<td>M III</td>
<td>3p3/2</td>
<td>875</td>
</tr>
<tr>
<td>M IV</td>
<td>3d3/2</td>
<td>630.8</td>
</tr>
<tr>
<td>M V</td>
<td>3d5/2</td>
<td>619.3</td>
</tr>
</tbody>
</table>

Table 2.2: The electron binding energies for iodine for the three inner electron shells. Data from [20] and [21]
Figure 2.4: Mass attenuation coefficient against energy for NaI(Tl). This graph depicts how for different energies the different interaction modes contribute to the total interaction change. The upper (bold) line is the total mass attenuation coefficient. The total mass attenuation coefficient is composed out of the three different interactions. The three lines are the photoelectric effect ("photo"), the Compton scattering ("Compton Total") and Pair production ("pair"). For γ-rays of 0.5-1.5 MeV it is seen that the Compton scattering is the dominant interaction.
Chapter 3

Experimental setup and expected signals

To study the rate of decay of radioactive sources scintillator detectors are used to detect outgoing $\gamma$-rays. An incoming $\gamma$-ray interacts with the scintillation material and produces an amount of light that is proportional to its energy. The light from the NaI(Tl) crystal reaches a Photo Multiplier Tube (PMT) where the light is turned into a proportional electric signal. For post processing of data every single outgoing pulse from the PMT can be saved. This chapter focuses on the goals and setup of the experiment, the working of the detector, the signal processing and the expected spectrum. To explain the shapes in the spectrum the working of NaI crystal and the PMT is explained.

3.1 Goals of the Modulation experiment

The goal of the Modulation experiment is to measure if radioactive sources exhibit a yearly modulation in their activity of the order of 0.1%. Past experiments had disadvantages in identifying the source of the observed modulation. These where that they measured the activity of a source only a few times per day or even per week, most detectors where not shielded from natural or cosmic radiation and they could not exclude effects from local environmental influences such as temperature and pressure. The Modulation experiment can save every single pulse coming out of the detector instead if only spectra. Detected pulses will be time-stamped, so changes in activity can be derived later and pinpointed to occurrences that happened concurrent. Four identical experimental setups will be build and placed at different location around the world. By comparing the results local influences will
be excluded while effects that influence the four boxes simultaneously (like neutrinos from the sun) can be identified.

To measure a change in the order of 0.1% of total activity Monte Carlo simulations have been performed for the setup with different sources. To measure a 0.1% modulation with a 5 sigma significance the minimal activity for the sources like $^{40}\text{K}$, $^{54}\text{Mn}$, $^{60}\text{Co}$, $^{133}\text{Ba}$ and $^{137}\text{Cs}$ need to be in the order of $10^1\text{Bq}$, $10^4\text{Bq}$, $10^1\text{Bq}$, $10^1\text{Bq}$, $10^7\text{Bq}$ respectively. This experiment will run for at least a year since the found Modulation has a period of a year.

### 3.2 The experimental setup

The four sources will each be monitored by two NaI(Tl) detectors placed opposite of each other (see figure 3.1). The sets of detectors will be enclosed in an so called inner box. To exclude local variation this inner box will be air tightly sealed, filled with an radio pure gas and kept on a steady temperature. To exclude as much natural radioactivity from the air the radon level will be measured. Sensors will also measure the pressure, temperature, humidity and magnetic field and this slow data will also be saved. To lower the cosmic ray contribution, natural radioactivity and the $\gamma$-rays of neighboring sources the detector sets will be enclosed by a lead shield with a width of 5 cm.

![Figure 3.1: Left: photo of inner box with 7 detectors, wiring and one of the lead shields. Right: drawing of one of the detector set with a radioactive source in between.](image)

An outer box will house the necessary electronics and is heat isolating to ensure the inner box keeps a constant temperature. The set of electronics used will consist of a high voltage supply, an NI-PXIe DAQ with FPGA module for the data acquisition, a PID heater controller, a gas flow system,
a Radon meter, a voltage supply and a data storage server. The box is connected from the outside by a power supply, an ethernet cable and gas inflow. The ethernet cable will be used to transport the data during the experiment to see externally if the data flow is stable. A safety system will cut off the main power if one of the temperature monitors inside the inner or outer box measures a problem with the temperature. These safety measures are to make sure the heaters and power supplies will not damage the whole setup in case of a malfunction. An overview of how the electronics fit in the outer box and how all the wiring works can be found in figure 7.6 and 7.7 in the appendix.

![Figure 3.2: Drawing of the setup of the outer box with isolation plates, the inner box and the readout electronics. Courtesy of R. Walet](image)

### 3.3 3”x3” NaI(Tl) scintillation detectors

Eight identical detectors are used with a built-in Voltage Divider and LED stabilization (build by Scionix). It has a Save High Voltage (SHV) connector for a typical positive high voltage of 500-1000V, a BNC connector for the output signal and a LEMO connector to operate the LED inside. A schematic of the detector is seen in figure 3.3 in the appendix. The LED has been engineered next to the crystal to send a light pulse directly onto the photocathode. For the LED no electric circuit is used and the LED is a 3mm blue water/clear lens (470nm) with a forward voltage of +3.5V. This calibration system can used to determine any changes in PMT amplification over time and to calculate the dead time.
Figure 3.3: Schematic picture of the detector with NaI(Tl) crystal, PMT and connectors. Image K. Heijhoff

<table>
<thead>
<tr>
<th>Scintillator material:</th>
<th>NaI(Tl)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density:</td>
<td>3.67 g cm$^{-3}$</td>
</tr>
<tr>
<td>Wavelength at max emission:</td>
<td>415 nm</td>
</tr>
<tr>
<td>Decay time:</td>
<td>230 ns</td>
</tr>
<tr>
<td>Refractive index:</td>
<td>1.85</td>
</tr>
<tr>
<td>After glow:</td>
<td>0.5-5%</td>
</tr>
</tbody>
</table>

Table 3.1: Characteristics of the NaI(Tl) scintillation crystal. Data from Gilmore.

3.3.1 Scintillation crystal

To determine the activity of the radioactive sources the choice of scintillation crystal is important to ensure that most $\gamma$-rays are detected and that the interactions give a clear identifiable signal. Sodium Iodine crystals have been used as scintillation detectors since the early 1950s because of the high light yield, linear energy response and widespread availability. Even though other materials such as High Purity Germanium (HPGe) have a much better energy resolution (1-2%) and lower energy threshold (∼KeV) the costs of production and operation do not outweigh these benefits for this experiment.

Every $\gamma$-ray that enters the crystal has a chance to interact with the crystal. To show the complexity of outcomes for the different interactions figure 7.5 (in the appendix) shows all possible outcomes for the photoelectric effect for an incoming photon with an energy above 33.2 keV (the binding energy of the K shell of Iodine) in NaI(Tl). The chance that the photon excites the K shell is 83%. It then sends out an X-ray (83% chance) or lowers another electron from the L or M shell. The freed electron creates electron-hole pairs which are captured by the the activator atoms (here Thallium). This happens because of the lower energy levels of the activator (they lie between the valance and conduction band of the Iodine). This light does not have enough energy to excite another Iodine electron and is able to reach the PMT unhindered. Only about 12% of the energy of the $\gamma$-ray is converted.
this way into light, the rest dissipates into lattice vibrations.

The electrons freed by the different interaction lose their energy through ionization (Coulomb interaction) and Bremsstrahlung (radiative loss). For electrons energies of 0.05 to 1 MeV they normally travel between $0.5 \times 10^{-3}$ and $0.5 \times 10^{-1}$ cm in sodium iodine[19] which means they normally stay within the scintillation crystal. The Coulomb interaction is mainly between the now moving electrons and the 'stationary' electrons in the medium, although they also have a small chance of colliding with the nucleus. These interactions result in an electron path being changed by a lot of small angle scatterings. The effect for a photo-electron ejected by a $^{137}$Cs γ-ray in sodium iodine is illustrated in figure 3.4.

![Figure 3.4](image)

Figure 3.4: Illustration of the range of an electron ejected by a 0.662 MeV $^{137}$Cs γ-ray giving up its kinetic energy in the production of ionization and excitation followed by the emission of light proportional to the kinetic energy of the electron and hence the energy of the incoming γ-ray. Image and text from [22]

Positrons which are created by pair production lose their energy in the same way as electrons. The difference is that when most of their kinetic energy is gone they will capture an electron to form a bound state, called a positronium. This is an unstable state that will decay again into two photons which, to conserve energy and momentum, leave in opposite directions and each have an energy of exactly 0.511 MeV. For electrons that are ejected in the material close to the edge of the detector there is a chance that a portion of the energy will escape by electrons or photons leaving the medium. Since the used NaI(Th) detectors have dimension(3” x 3” inch) which are bigger than the average path length of the electrons, there is a high chance that the full energy of the incoming γ-ray is transferred to the PMT.
3.3.2 Photo Multiplier Tube

A photon that hits the photocathode of the PMT has a chance to free an electron depending on its frequency. To get a high total light yield the sensitive frequency region of the scintillation material and photocathode must overlap as much as possible. The light yield of NaI(Tl) is \((39 \pm 1) \times 10^3\) photons per MeV. Because there is an overlap in both sensitive frequency regions (see figure 3.5) this setup has a high scintillation conversion efficiency. The electric field between the cathode and the first dynode accelerates the electron to create a small electron avalanche. This happens 10 times before the total avalanche hits the anode in the back of the PMT which gives the electric output signal, see figure 3.3.

![Figure 3.5: Quantum efficiency of photocathode. The wavelength at max emission for NaI(Tl), 415 nm, lies within the quantum efficiency peak. The Image from specsheet ET Enterprises PMT 9305KB](image)

The PMT used in the modulation experiment is an ETL 9305, it is an 78 mm (3) diameter, end window photomultiplier with blue-green sensitive bi-alkali photo-cathode and 10 high gain, high stability, SbCs dynodes of linear focused design for good linearity and timing. The specifications can be found in in table 3.2 and the electrical circuit in the appendix.

The mean life time of the excited activator - the time that it takes for an photon to be released again - is 230 ns. When an electron finds itself in a forbidden transition from the ground state it can take a much longer time to de-excite. NaI(Tl) has a 0.3-5% afterglow which means that this fraction of light is still emitted 6 ms after the initial fluorescence. In comparison to the primary scintillation time of NaI(Tl) of 230 ns this can give a problem with hight event rates. When another event takes place within the afterglow
Window Material | Borosilicate
--- | ---
Cathode Type | KCs
Diameter | 78 mm
Min Spectrum | 280 nm
Max Spectrum | 630 nm
Gain | 0.70 x 10^6
Dark Current Typical | 0.50 nA
Peak Current Linearity | 50.00 mA

Table 3.2: Specifications for the 9305KB PMt from ET Enterprises.

of the last event the light piles up and a higher output signal is measured.

### 3.4 Signal processing

The signal from the detector goes to an Analog to Digital Converter (ADC) and then to an Field Programmable Gate Array (FPGA), both are powered and controlled by the National Instruments PXIe-PCie8375. The FPGA is programmed using LabVIEW 2012 SP3. (3.6) The ADC has a resistor of 50 Ohm and converts a range of 2V into a 16-bit (unsigned) integer (2^{16} – 1 steps).

![Figure 3.6: The setup for two detectors. Used is a Caen SY-127 high voltage power supply, a NI-PXI with FPGA and ADC modules. The data is saved with LabVIEW to hard disk for post processing.](image)

The integral of a single signal pulse (figure 3.7) is proportional to the energy of the $\gamma$-ray. When the voltage from the anode of the PMT is higher than a set threshold value the pulse is saved. From the 10 data points before the threshold is reached the baseline is calculated. The time between two data points is set to $20 \cdot 10^{-9}$s. This captures the whole signal since the fall time (from 90% to 10% peak height) of these pulses is around 600 ns. This data, together with the channel number and timing information is saved in a binary file.
Figure 3.7: Single pulse from a detector. The baseline (green) is calculated from the first 10 data points. Time between two data points is \(20 \cdot 10^{-9}\) s. The integral of this pulse is proportional to the energy of the detected \(\gamma\)-ray. For every pulse this value is calculated and the histogram of that data is the spectrum.

For every signal the peak height, baseline, integral, time and channel number are calculated and saved. From all the integral values for a set period a spectrum can be made as seen in figure 3.9. Every count in the spectrum is the integral of a single pulse. To find the peak values of the photopeak a Gaussian fit is made with Root. Background and photopeak models are best fitted to data and manually checked for errors. See figure 3.8.

Figure 3.8: Spectrum of \(^{60}\)Co with Gaussian fits to both photopeaks. Background fitted with 2nd order polynomial (BG0-BG2). Values used are "Peak" and "FWHM%" 1 and 2. Areas of the peaks are calculated separately.
3.5 The resulting $\gamma$ spectrum

To determine the decay rate for a radioactive source all the signals from the detector need to be counted. The total detection rate is the sum of several different sources: gammas from the primary source, cosmics (muons) from secondary air showers, gammas from natural radioactive decay, gammas from secondary sources and noise. To select the signals from the source it is possible to make a cut in the energy range. In figure 3.9 an energy spectrum is shown taken with one of the used detectors. The contributions are specific for the geometry of the detector and are described below.

![Spectrum of Co-60](image)

**Figure 3.9:** Spectrum of $^{60}\text{Co}$ made by the NaI(Tl) detector (ID: SBJ593). Total count is 1 million, HV is on 580 V. Both photopeaks and the sum peak are clearly identifiable. The annihilation peak from the pair creation lies in the middle of the Compton edge of the first photopeak. The left region is empty because of the set threshold. This spectrum includes the background and no energy calibration has been made.
3.5.1 The photopeak

If every $\gamma$-ray would transfer its full energy ($h\omega - E_{\text{binding}}$) into the scintillation crystal a single delta function should be observed in the spectrum. This is called the photopeak and two of these are shown in figure 3.9. The photopeak is broader than a delta function and this energy resolution is described in chapter 4.

3.5.2 Compton continuum

The transferred energy in Compton scattering is dependent on the scattering angle. The kinetic energy of the recoil energy is given by:

$$E_{e^-} = h\omega_{in} - h\omega_{out} = h\omega_{in}\left(\frac{(h\omega/m_0c^2)(1 - \cos\theta)}{1 + (h\omega/m_0c^2)(1 - \cos\theta)}\right)$$  \hspace{1cm} (3.1)

Here two extremes can be identified. First the $\gamma$ can graze the electron in which case $\theta \cong 0$ and $E_{e^-} \cong 0$. Secondly the $\gamma$ can have a head on collision with the electron which results in the $\gamma$-ray backscattering towards its direction of origin and transfer max energy. In this case the energy of the recoil electron is given by:

$$E_{e^-}|_{\theta=\pi} = h\omega\left(\frac{2h\omega/m_0c^2}{1 + 2h\omega/m_0c^2}\right)$$  \hspace{1cm} (3.2)

Since for an incoming $\gamma$-ray every Compton scattering angle will occur in the crystal a continuum of energies can be transferred, ranging from 0 to the maximum $E$. This is shown in figure 3.10 and can also be seen in the $^{60}\text{Co}$ spectrum in figure 3.9. The size of the scintillation crystal changes the ratio between photopeak and Compton continuum. For an infinitely large spherical detector centered around a source no photons would be able to escape and only a photopeak would be seen on the spectrum. For very small detectors the chance for a photon to leave after Compton scattering is high and the Compton continuum would be large compared to the photopeak. For real size detectors there is also a chance that a $\gamma$-ray will Compton scatter multiple times, resulting in energy deposits that lie between the Compton edge and the photopeak. Not all signals are from the primary radioactive source in the spectrum since the natural and cosmic background has not been excluded here.

3.5.3 Other contributions to the spectrum

Other contributions to the spectrum are Compton backscatter peaks, X-ray peaks and annihilation peaks. A Compton backscatter peak is found when
\( \gamma \)-rays enter the material around the detector and are scattered back into the detector. For scattering angles greater than 110-120° the backscattered \( \gamma \)-rays have an energy of around 0.2-0.25 MeV. When the \( \gamma \)-rays undergoes photoelectric effect in surrounding materials (for example lead) the outgoing X-ray can be captured again by the detector. This gives an characteristic X-ray peak with an energy depending on the material it came from. In case of lead the characteristic X-ray energies is in the 72-84 keV range. Pair-production in surrounding material can send annihilations photons back into the detector. This results in more hits in the annihilation peak of 511 keV. These expected contributions to the spectra are shown in figure 3.10.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig3.10}
\caption{Expected peaks in spectra from external contributions. The Compton continuum with Compton edge and the Photopeak are shown as dotted lines. 1 is the characteristic X-ray peak, 2 is the Compton backscatter peak and 3 is the annihilation peak. Figure from [19]}
\end{figure}
Chapter 4
Performance of NaI(Tl) scintillation detectors

To measure the activity of a radioactive source with high precision over a period of a year the signal response of all eight detectors need to be known. Since a modulation could be in the order of 0.1% of the activity, all effects that influence the activity measurement should be distinguishable. Therefore the following detector characteristics are described in this chapter: gain of the PMT, energy resolution, detection efficiency, geometric effects, peak to total ratio, energy calibration and linearity. With these measurements the optimum working voltage per detector is determined and an estimate is made on the measurable change in detection rate for a modulation as discussed in chapter 1.

4.1 Gain of the Photomultiplier Tube (PMT)

The Gain ($G$) of the PMT is the ratio between the photoelectric current and the output current of the anode. $G = \frac{I_{\text{anode}}}{I_{\text{photocathode}}}$. The average secondary electron emission ratio $\delta$ per dynode step depends on the interstage voltage ($V_{\text{dyn}}$) and a material coefficient $\alpha$, given by:

$$\delta = A \cdot V_{\text{dyn}}^\alpha$$  \hspace{1cm} (4.1)

Here $A$ is a constant. The total Gain is defined as $G = \delta^n$ where the PMTs used have 10 dynode steps ($n=10$).

$$G = \delta^n = (A \cdot V_{\text{dyn}}^\alpha)^n = (A \cdot (\frac{V}{n+1})^\alpha)^n = \frac{A^n}{(n+1)^{\alpha n}} \cdot V^{\alpha n} = K \cdot V^{\alpha n}$$  \hspace{1cm} (4.2)

Here $K$ is a constant depending on the fabrication of the PMT.
Table 4.1: Detector setup with sources and voltage range.

<table>
<thead>
<tr>
<th>Detector</th>
<th>Source</th>
<th>$V_{\text{start}}$(V)</th>
<th>$V_{\text{end}}$(V)</th>
<th>$V_{\text{steps}}$(V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SBJ586</td>
<td>Cs-137</td>
<td>460</td>
<td>660</td>
<td>20</td>
</tr>
<tr>
<td>SBJ588</td>
<td>Ba-133</td>
<td>780</td>
<td>980</td>
<td>20</td>
</tr>
<tr>
<td>SBJ589</td>
<td>Ba-133</td>
<td>520</td>
<td>720</td>
<td>20</td>
</tr>
<tr>
<td>SBJ590</td>
<td>Co-60</td>
<td>470</td>
<td>670</td>
<td>20</td>
</tr>
<tr>
<td>SBJ591</td>
<td>Co-60</td>
<td>500</td>
<td>700</td>
<td>20</td>
</tr>
<tr>
<td>SBJ592</td>
<td>Co-60</td>
<td>500</td>
<td>700</td>
<td>20</td>
</tr>
<tr>
<td>SBJ593</td>
<td>Co-60</td>
<td>550</td>
<td>750</td>
<td>20</td>
</tr>
</tbody>
</table>

The PMT data sheet indicates how the gain should depend on the applied voltage (‘specsheet’ plotted in figure 4.1. To measure the gain of the PMT the photoelectric current is estimated and the anode output current was measured for 7 detectors placed in sets of 2 around one radioactive source (figure 3.6). A SY-127 Caen power supply was used to change the voltage (2V resolution) over a range of 200V with 20V increments. The starting voltages in table 4.1 were high enough to see a full photopeak over the threshold. In every spectrum the photopeak position (P) was found assuming a normal distribution (the errors are independent). The total output charge is given by:

$$Q_{\text{output}} = \int I_{\text{anode}} dt = \int \frac{V_{\text{anode}} dt}{R_{\text{ADC}}} = \frac{P}{50\Omega} \quad (4.3)$$

The photocathode current is derived from the amount of photoelectrons ($n_{\text{PE}}$) that are freed from the photocathode when a 1 MeV gamma ray has photoelectrically interacted. The total charge before the first dynode is $n_{\text{PE}} \cdot e$, where $e$ is the electron charge. The light yield is reported to be around $(40 \pm 2) \times 10^3$ photons per MeV in NaI(Tl). If one assumes a typical Collection Efficiency (CE) of 20% then 8000 photons will reach the photocathode. The Quantum Efficiency (QE) has a maximum of 30% for 350 nm (figure 3.5), since scintillation light from NaI has a typical wavelength of 415 nm, a 25% QE can be assumed over the total spectrum which results in a total amount of photoelectrons of $(2 \pm 1) \times 10^3$ per MeV. All gains have been calculated with this number of photoelectrons and a change in this number will shift all gain curves up or down in a linear fashion.

$$n_{\text{PE}} = E_{\gamma} \cdot 4 \cdot 10^5 \cdot QE \cdot CE = E_{\gamma} \cdot 4 \cdot 10^5 \cdot 0.25 \cdot 0.2 = \alpha \cdot E_{\gamma} \quad (4.4)$$

With $\alpha$ is $(2 \pm 1) \times 10^3$ [PE/MeV].
Now the gain of the PMT is:

\[
G = \frac{Q_{\text{anode}}}{Q_{\text{photocathode}}} = \frac{P[V_{\text{a}}]}{n_{\text{PE}} \cdot 1.60 \cdot 10^{-19}[C]}
\]  

(4.5)

**Figure 4.1:** Gain of the PMT for the detectors. Errors are due to the uncertainty in \(n_{\text{PE}}\). The expected gain from the PMT spec sheet[23] is also plotted.

The gain of the PMT of the detectors used is shown in figure 4.1. An exponential function, from equation 4.2, can be fitted to the gain plots. The \(\alpha\) and \(K\) values are calculated from the found fit parameters and shown in table 4.2. For 7 identical PMT’s the \(\alpha\) values vary between \((7.35 \pm 0.14)10^{-4}\) and \((1.88 \pm 0.07)e^{-3}\) and the \(K\) values vary between \(51.9 \pm 23\) and \(449 \pm 63\).

Six of the seven detectors have a higher PMT gain at the given voltage than indicated on the specification sheet. All PMTs reach a gain between \(10^5\) and \(10^6\) while operated between 500 and 1000 V. For the PMT of the SBJ586 detector 8 instead of 11 data points have been collected since for three data points no fit could be made to the photopeak. To test this hypothesis the gain of SBJ586 should be measured again with another source of lower activity. The SBJ587 detector did not have reach the specified gain in this voltage range and was sent back to the manufacturer for repair.
Table 4.2: Fit results for gain as seen in figure 4.1. Fit function = \([C_0] + [C_2] \cdot x^{10 \cdot C_1}\). Here \(C_1 = \alpha\) and \(C_2 = K\) as in equation 4.2

<table>
<thead>
<tr>
<th>Detector</th>
<th>(C_1)</th>
<th>(C_2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SBJ586</td>
<td>(1.88±0.07)e-3</td>
<td>51.9±23</td>
</tr>
<tr>
<td>SBJ588</td>
<td>(7.35±0.14)e-4</td>
<td>449±63</td>
</tr>
<tr>
<td>SBJ589</td>
<td>(1.33±0.04)e-3</td>
<td>105±28</td>
</tr>
<tr>
<td>SBJ590</td>
<td>(1.30±0.04)e-3</td>
<td>253±62</td>
</tr>
<tr>
<td>SBJ591</td>
<td>(1.26±0.04)e-3</td>
<td>183±45</td>
</tr>
<tr>
<td>SBJ592</td>
<td>(1.28±0.07)e-3</td>
<td>172±43</td>
</tr>
<tr>
<td>SBJ593</td>
<td>(1.06±0.04)e-3</td>
<td>209±56</td>
</tr>
</tbody>
</table>

4.2 Energy resolution

For a perfect detector the photopeak is expected to be a delta function. Which is in contrast to the widened peaks in the measured energy spectrum. This widening contains contributions from a range of separate effects such as charge collection statistics, electronic noise, fluctuations in PMT gain, variations in the detector response over its active volume and drifts in parameters over the course of the measurement. The energy resolution is defined as the Full Width at Half Max (FWHM) in percentage.

\[
R = \frac{FWHM}{H_0}\quad (4.6)
\]

where FWHM is the full width at half maximum of the full-energy peak and \(H_0\) is the mean pulse height corresponding to the same peak. In figure 4.2 this calculation is shown for the photopeak in the spectrum of \(^{137}\)Cs.

The total energy resolution has contributions from the PMT and scintillation energy resolutions:

\[
R^2 = R^2_{\text{scintillator}} + R^2_{\text{PMT}}\quad (4.7)
\]

By changing the voltage the energy resolution of the PMT behaves as:

\[
R^2_{\text{PMT}} = \frac{2.35^2}{N_{VUV} \cdot QE \cdot CE} \cdot \frac{\delta}{\delta - 2}\quad (4.8)
\]

Here \(N_{VUV}\) is the number of scintillation photons, QE and CE the Quantum Efficiency and Collection Efficiency, respectively. With changing the voltage on the PMT the mean secondary emission yield of each dynode(\(\delta\)) and the CE of photoelectrons at the first dynode change the total energy resolution.

The width of the energy resolution is heavily influenced by the statistical spread in the amount of photoelectrons created at the photo-cathode. Since
the amplitude of the output signal is proportional to the amount of electrons reaching the first dynode, the statistical variance in photoelectrons influences the statistical variance in the signal output. Here the standard deviation is $\sqrt{2000}$, or 2.2% of the mean energy assuming Poisson statistics. For a Gaussian shape the FWHM is 2.35 times the standard deviation and this

\[ f = e^{-0.5\left(\frac{x-\mu}{\sigma}\right)^2} \] with $\mu=5$.

For higher energy resolution the peak broadens and a higher contribution from Compton edge and background hits are expected to be found within the photopeak region.
makes the statistical contribution to the energy resolution—for a 1 MeV $\gamma$ ray—5.3%.

---

**Figure 4.4:** Energy resolution as a function of PMT gain for detectors used.

The energy resolution of the detector is shown in figure 4.14. For the lower voltage region it can be seen that the energy resolution improves for higher voltage (higher gain). This phenomenon can be caused an increase in the number of photoelectrons since then $\sqrt{n}$ improves. In the higher voltage region the energy resolution decreases for higher gain which can be caused by after pulsing and higher electron losses between dynodes.

To count the hits within the photopeak, which will be the goal when determining the activity of a radioactive source, a better (lower) energy resolution lowers other contributions to the peak. How the peak broadens from 4% to 6% energy resolution can be seen in figure 4.3. To lower this effect the operating voltage needs to be determined to minimize the energy resolution. Thus the optimum operating voltage ($V_{\text{work}}$) is where the energy resolution is lowest, these values are found in figure 4.14 and table 4.3.

The energy resolution for all detectors is shown in figure 4.4. The energy resolution for a particular source depends on the PMT voltage. The difference in resolution between sources is caused by the energy of the $\gamma$-ray and the
Table 4.3: Lowest energy resolution and working voltage per detector.

<table>
<thead>
<tr>
<th>Detector</th>
<th>FWHM (%)</th>
<th>$V_{work}$</th>
<th>$E_{\gamma}$ (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SBJ586</td>
<td>7.29(5)</td>
<td>522(2)</td>
<td>662</td>
</tr>
<tr>
<td>SBJ588</td>
<td>13.8(1)</td>
<td>990(2)</td>
<td>356</td>
</tr>
<tr>
<td>SBJ589</td>
<td>13.8(1)</td>
<td>646(2)</td>
<td>356</td>
</tr>
<tr>
<td>SBJ590</td>
<td>5.03(5)</td>
<td>578(2)</td>
<td>1173</td>
</tr>
<tr>
<td>SBJ591</td>
<td>4.83(4)</td>
<td>628(2)</td>
<td>1173</td>
</tr>
<tr>
<td>SBJ592</td>
<td>5.07(4)</td>
<td>586(2)</td>
<td>1173</td>
</tr>
<tr>
<td>SBJ586</td>
<td>4.87(3)</td>
<td>718(2)</td>
<td>1173</td>
</tr>
</tbody>
</table>

number of different $\gamma$-ray energies that a source will send out. For lower $\gamma$-ray energies the ratio between the photoelectric effect over the Compton scattering increases. The broad FWHM of the Ba-133 photopeak is caused by the peaks of energies close to the photopeak.

### 4.3 Detector efficiency and geometric effects

#### 4.3.1 Total detector efficiency

The detector efficiency is the percentage of $\gamma$ rays form a source passing though the scintillator that interact and are counted in the final spectrum. The total count rate in the spectrum is dependent on the set threshold and the background rate (natural radiation and cosmic particles). The percentage of interacting $\gamma$ rays is depending on the distance between source and detector and the size and shape of the NaI(Tl) crystal. The total activity ($A_t$) of the source is calculated with respect to the original calibration $A_0$, the half life ($t_{1/2}$) and the time since calibration ($t$).

$$A_t(t) = A_0 * e^{-\frac{\ln(2) \cdot t}{t_{1/2}}}$$

(4.9)

To calculate the total detection efficiency per detector set per source the total number of counts per spectrum minus the number of background hits for the same HV is compared to the total activity of the source. (see figure 4.5) For all three sources the total count is adjusted for the number of $\gamma$-rays per decay. The $^{137}\text{Cs}$ efficiency is from only one detector. For the $^{60}\text{Co}$ source the efficiency reaches a plateau. For higher HV (gain) the efficiency improves as is expected since more of the lower end of the spectrum goes over the threshold (trigger) level and is counted in the spectrum. The number of cosmic events has not been subtracted. Monte Carlo studies done by A. Kish on the detector setup show that the total detector efficiency should be 12.6
Table 4.4: Activities of radioactive sources used for performance measurements. All sources where calibrated on 1-12-1980 with the exception of Co-60 which was calibrated on 1-1-1980. Equation 4.9 was used. The $\gamma$/decay values from BNM-LNHB/CEA tables. $^{60}$Co (1) has source number: 1U546. All times in days. *Activity can be 50% higher or lower since the original source activity has not been documented.

and 41.6 for $^{137}$Cs and $^{60}$Co respectively. The $^{60}$Co efficiency is a few % higher than this data, the $^{137}$Cs efficiency is doubles the Monte Carlo values as is shown in figure 4.5.

![Figure 4.5](image)

Figure 4.5: Total detection efficiency for a detector set and a single detector(SBJ586). Legend in color. The Ba-133 efficiency is not adjusted for the number of $\gamma$-rays per decay.
4.3.2 Geometric effects

Small fluctuations in the position of the sources caused by for example temperature and humidity variations may result in apparent modulations mimicking ones found by others. Moving the source with respect to the two detectors changes the amount of $\gamma$ rays traveling through the scintillator. The distance between the two detectors is 6.5 mm while the radius for one detector is 42 mm. If the source is placed right in the middle the detectors will get equal amounts of $\gamma$ rays assuming the direction of decay is uniformly distributed over a sphere. For this setup the amount of $\gamma$ rays going through the spherical cap for of one detector is 47.5% of the whole sphere. Calculating the area of the detector ($A_{\text{cap}}$) with $R$ the radius of the sphere and $h$ the distance between the source and detector:

\[
A_{\text{cap}} = \int \int d\phi d\theta \sin(\theta) R^2 = 2\pi R^2 \int_0^{\theta_{\text{max}}} d\theta \sin(\theta) \quad (4.10)
\]

\[
\theta_{\text{max}} = \frac{\pi}{4} - \arcsin\left(\frac{h}{R}\right) \approx \frac{\pi}{2} - \frac{h}{R} \quad (4.11)
\]

\[
A_{\text{cap}} = 2\pi R^2 \cos\left(\frac{\pi}{2} - \frac{h}{R}\right) = 2\pi R^2 \sin\left(\frac{h}{R}\right) \approx 2\pi Rh \quad (4.12)
\]

\[
A_{\text{cap}} = 2\pi Rh = 2 \cdot \pi \cdot 42 \cdot (42 - 3,15) = 10225\text{mm}^2 \quad (4.13)
\]

\[
A_{\text{ratio \ 2 \ caps}} = \frac{2 \cdot A_{\text{cap}}}{4\pi r^2} = \frac{20451}{22167} = 0.92 \quad (4.14)
\]

If the source is placed directly against one of the detectors the geometric efficiency changes to 38% for the furthest detectors and to 50% for the closest.
detector.

\[ A_{\text{ratio 1 cap}} = \frac{\pi \cdot 43.2 \cdot (43.2 - 6.5)}{4 \cdot \pi \cdot 43.2^2} = 0.43 \]  

(4.15)

The conclusion is that the geometric efficiency for a set of detectors will stay close to 0.92\% when the relative source-detector distance changes. The other 7\% of all \( \gamma \) rays will escape the detector set. To suppress the rate change due a change in the distance between the source and the detector, a set of two detectors are used. In figure 4.7 the rate change is shown for the maximum change in the distance between the source and the detector. By setting up the experiment in this way the combined rate does not change more than 0.9 \( \pm \) 0.04 \%, even though the detection rates of the two individual detectors change by approximately 10\%.

![Figure 4.7: Rates for different distances between detector SBJ590 and a \(^{60}\text{Co}\) source. Setup is seen in figure 4.6](image)

### 4.3.3 Peak to total ratio

The ratio of \( \gamma \)-rays which undergo the photoelectric effect changes with the size of the NaI(Tl) crystal. A bigger crystal will for example have a higher chance of photoelectric absorption as well as double and triple Compton scattering. This is shown in the spectrum by the ratio between the counts in the photopeak and the counts in the total spectrum (see figure 4.8). The expected peak to total ratio for a 3x3 inch NaI crystal is 0.42 for an 1 MeV \( \gamma \)-ray and 0.68 for a 0.5 MeV \( \gamma \)-ray. Monte Carlo studies performed in the frame the of decay modulation experiment shows that the peak to total fraction for \(^{60}\text{Co}\) is 0.15 and 0.16 for the 1173 and 1332 keV peaks respectively. The
combined peak to total ratio of 0.31 is within the experimental values found in figure 4.8.

![Ratio_Peak_to_total_Spectrum_SBJ592](image)

**Figure 4.8:** Peak to total ratio for SBJ592 with $^{60}$Co source. The difference between the 1.173 MeV and 1.332 MeV peak ratios is explained by the different chance to photoelectrically interact. (see table 2.1)

### 4.4 Energy calibration and linearity

#### 4.4.1 Energy calibration

To calibrate the detector at a certain voltage spectra from radioactive sources with different $\gamma$-ray energies are used. The position of the photopeak is found (Gaussian fit) and this value is related to the known energy of the $\gamma$-ray of the source. If the integral of the pulse is proportional to the energy of the incoming $\gamma$-ray for all energies the calibrations points should be on the same line as seen in figure 4.9. In our setup the peak position is dependent on the detection rate, therefore the sources are placed at a certain distance from the detector to get a 200(6) Hz detection rate for all three sources. Why the gain increases (or peak position) for higher detection rates requires further research.

The observed energies of $\gamma$-rays, calculated from the position of the photopeaks, are within 0.1% of the expected values as can be seen in table 4.5.

With this energy calibration the spectra for $^{133}$Ba, $^{137}$Cs and $^{60}$Co can now be shown with their correct energy scale (see figure 4.10). The SBJ591 detector (at 628 V) can reproduce the energy of a $\gamma$ with a maximum deviation of 0.2% (spectra taken at 200(6) Hz with at least 200k events).
Figure 4.9: Energy calibration for SBJ591 at 628 V with linear fit. Error bars are within markers. Detection rate above threshold is 200(7) Hz for all three sources. The peak energies are shown in table 4.5.

Table 4.5: Difference between known and found γ energies for $^{133}$Ba, $^{137}$Cs and $^{60}$Co. Energy calibration from fit in figure 4.9. Known values from LNE-LNHB/CEA tables. Residuals are expected values minus measured values.

<table>
<thead>
<tr>
<th>Source</th>
<th>$E_\gamma$ (keV)</th>
<th>$E_{\text{found}}$ (keV)</th>
<th>Difference (%)</th>
<th>Residuals</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}$Ba</td>
<td>356.0</td>
<td>355.9(1)</td>
<td>0.03</td>
<td>0.1</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>661.7</td>
<td>662.3(2)</td>
<td>0.09</td>
<td>-4.6</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1173.2</td>
<td>1171.3(5)</td>
<td>0.16</td>
<td>1.9</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1332.5</td>
<td>1332.9(5)</td>
<td>0.03</td>
<td>-0.4</td>
</tr>
</tbody>
</table>

The energy resolution is inversely proportional to the γ-ray energy:

$$R \equiv \frac{\text{FWHM}}{\text{peak position}} = K \frac{\sqrt{E}}{E} = \frac{K}{\sqrt{E}} \quad (4.16)$$

Here $K$ is a constant of proportionality.

$$\ln R = \ln K - \frac{1}{2} \ln E \quad (4.17)$$

Thus the plot of ln R versus ln E should be a straight line with a slope of
Figure 4.10: Spectra of $^{133}$Ba(Red), $^{137}$Cs(Blue) and $^{60}$Co(Green). Detection rates are 196, 206 and 200 Hz respectively. Photopeak values differ less than 0.2% from values from literature. The 302 keV photopeak of $^{133}$Ba is also visible as a distinct peak.

In figure 4.11 we can see a slope of -0.64 instead. This shows the energy resolution is not completely dominated by the peak broadening.

Figure 4.11: Energy resolution vs $\gamma$-ray energy for the photopeaks of $^{133}$Ba, $^{137}$Cs and $^{60}$Co. With linear fit, $K = p_0$ from fit.
4.4.2 Linearity

The typical shape of a single pulse, like height and length, is related to the build of the detector. The NaI crystal has an decay time of 230 ns and the PMT has a rise time of around 7.5 ns. For an instant pulse the voltage will decay exponentially with a time constant of RC, here R = 50Ω and C = 1000pF. The decay time of the crystal, the rise and fall time of the PMT and the RC time should not change for different $\gamma$-ray energies. If the shape of the pulse stays the same for different $\gamma$-ray energies the ratio of the peak height over pulse integral should not change. In figure 4.12 this is shown for three different spectra.

![Figure 4.12: Pulse height plotted against pulse integral. A total of 600k events are plotted, all data from the $^{133}$Ba(Black), $^{137}$Cs(Red) and $^{60}$Co(Blue) spectra. A linear ratio is seen for $\gamma$-ray energies up to 6 MeV. Entries above 3 MeV are cosmic particles. The origin of the two regions with low integral and high peak height and the second linear line requires further research.](image)

In the region from 0 to 3 MeV and under 0.05V peak height all the $\gamma$-rays from nuclear decays are shown. Above 3 MeV the detected events must be a cosmic particles since it has more energy that a $\gamma$-ray from a nuclear decay. Till 6 MeV the ratio is linear, from 6 MeV till 9 MeV the height grows faster than the integral. After which a region comes where the peak height increases while the pulse integral decreases. In this region the response (pulse integral) of the detector for larger incoming signals (higher energetic $\gamma$-rays) is not proportional. Since the activity of the source is subject of interest, events with a higher energy than 3 MeV will be omitted in the data analysis. In the energy region of $\gamma$-rays from radioactive decays the ratio of peak height
over pulse integral is linear. This result concurs with the energy calibration that in the 0-3 MeV region the response of the detector is proportional to the incoming energy.

4.5 Background signals

The experiment is designed to detect \( \gamma \)-rays from both the radioactive source as well as from the natural background radiation. Another contribution to the detected event rate are cosmic particles (mostly muons\[25\] ). The background, both of natural and cosmic origin, can vary in activity during the year. To lower the background and to limit cross-talk between the detector stations all the detectors are encased in 5 cm lead (see figure 3.1), the high energetic cosmic signals are cut out of the data and the inner box is filled with a radio pure gas.

4.5.1 Natural radioactivity and cosmic rays

At the Nikhef institute the background(BG) rate has been measured without and with lead. Without any lead for SBJ591 (628 V) the BG rate is 223 Hz in the energy region 0 to 3 MeV. With the lead setup this rate drops to 20 Hz. The cosmic rate (above 3 MeV) changes from 0.65 to 0.5 Hz from no lead to lead respectively. The rate of cosmic particles leaving a signal in a slice of the detector is \( 1.70(5)10^{-3} \) Hz as measured by K. Heijhoff (2013)\[25\]. For both situations the spectrum is shown in figure 4.13.

With 5 cm lead(Black) low energy \( \gamma \)-rays are stopped by the lead and more high energy events are detected in comparison to the no lead(Red) spectrum. Some known natural radioactive isotopes have been identified. A small percentage, 0.012% (120 ppm), of naturally occurring Potassium in the form of \(^{40}\)K is radioactive, this is present in for example concrete and glass. The \(^{214}\)Bi and \(^{214}\)Pb are daughters from the \(^{238}\)U decay chain which enters the lab mostly in the form of heavy Radon gas(\(^{222}\)Rn). The \(^{208}\)Tl originates from the natural occurring \(^{232}\)Th series.

To exclude some of the background rate a 3 sigma region (99.7\% assuming Gaussian) can be taken centered around the photopeak. This would lower the background rate to 1.16, 2.13, 1.01 Hz for \(^{133}\)Ba, \(^{137}\)Cs and \(^{60}\)Co. These values are calculated from the fit values from the energy calibration in the background spectrum with lead. This BG rate can still be lowered by filling the inner box with a radio pure gas. With the characteristics of the detectors it is now possible to calculate the minimum activity of the sources to make the size of a possible modulation signal one order bigger than the background.
Figure 4.13: Background spectrum with (black) and without (red) 5 cm lead encasing. Both spectra have 1 million events. Measured with SBJ591 (628 V) on 11/21/2013.

Signal. For a 1kBq $^{60}$Co source with a minimum total detection rate of 40% and a peak to total ratio of 30% one would detect 120 events per second in the peak region. A detectable modulation is then a change in the rate of 0.12 Hz in this region. For a 4 hour measurement this would result in a change of 1700 counts out of a total of 1.7 million events in the peak region.
Figure 4.14: Energy resolution with best operating voltage ($V_{work}$).
Chapter 5

Activity of $^{60}$Co over 50 days

To test the experimental setup the activity of $^{60}$Co has been measured at two different time periods with 37.5 days in between. In this chapter the expected activity is compared to the measured activity. The analysis is done for the whole spectrum and for the photopeak region individually. Possible improvements on the experimental setup will be presented in the following chapter.

5.1 Setup of measurement

The activity of one $^{60}$Co source ((1) from table 4.4) has been measured by detectors SBJ592 and SBJ593 over a long period of time (127-150 hours). The Caen SY-127 power supply was used with 600(2) V on SBJ592 and 680(2) V on SBJ593. The voltage per channel and the temperature has been measured every few days. The voltage did not change more than 2 V and the temperature did not change more than 2 ±0.5 °C in the air conditioned room. Both detectors where encased with 5 cm thick lead blocks as described in chapter 2. The source was losely placed between the two detectors and has not been taken out between measurements.

The first data set started on 25/09/2013 and lasted 127 hours. The second data set started on 07/11/2013 and lasted 149 hours. The second data set started 1027.5(2) hours (= 42.8 days) after the first one started. Zero hour for both measurements has been set to 11 AM on 25/09/2013. Every used data point is one hour’s worth of counting.

The expected activity (from eq 1.4) of the source at time $t$ is:

$$A_t(t) = A_0 \cdot e^{-t_{1/2}(2)}$$  \hspace{1cm} (5.1)
with $A_t(t)$ the activity at time $t$, $A_0$ the activity at time $t = 0$ and $t_{1/2}$ the half-life.

### 5.2 Results on the activity of $^{60}$Co

The detection rate of the full spectrum for $^{60}$Co over 149 hours is shown in figure 5.1b. The maximum change in Rate over 149 hours is 0.8(1)% for both detectors. The maximum expected change due to decay over 149 hours is 0.25(1)%. The background rate would need to change ± 50% in rate over 149 hours to account for this difference. After 120 hours an overall decrease can be seen in rate of the SBJ593 detector accompanied by an overall increase in the rate of the SBJ592 detector.

In figure 5.1b the detection rate of the full spectrum for $^{60}$Co is shown for the two data sets. For detector SBJ592 the rate detection of the second data set is higher than the first one. As seen in chapter 4 the detection rate for a single detector increases when the source is placed closer to it. As the sources are not held into place by any source holder it is possible that the source shifted position between the two detectors due to normal maintenance of the experiment. In figure 5.2 the average detection rate for the two detectors shows no sudden increase in detection rate. It is thus likely that the sudden overall in- and decreases in detection rate seen in figures 5.1b and 5.1b where caused by a smaller and larger shift in the source position, respectively.

![Graph](image1.png)  
**Figure 5.1**

(a) Detection rate for SBJ592(red) and SBJ593(blue) detectors with $^{60}$Co over 149 hours. Every data point is one hour of counting over the full spectrum. Error = $\sqrt{\text{Rate}}$

(b) Detection rate for SBJ592(red) and SBJ593(blue) detectors with $^{60}$Co over 1200 hours (± 50 days). Rate of full spectrum. Error = $\sqrt{\text{Rate}}$. The shift in activity between the detectors could be caused by a placement shift in the position of the source.
5.2.1 Analysis of full spectrum

In figure 5.2 the averaged combined detection rate of detectors SBJ592 and SBJ593 for the full spectrum of $^{60}$Co over 50 days is shown. The fit is an exponential decay function with starting activity ($p_0$) at day 0 and an half-life ($p_1$) of 1116(4) days. This value is $42 \pm 0.2\%$ lower than the $^{60}$Co half life of 1925.20(25) days from literature. An exponential decay function with the fitted starting value and the half-life of 1925.2 is shown in black. The measurements show a lower detection rate for the second data set than expected.

\begin{center}
\begin{tabular}{|c|c|}
  \hline
  $\chi^2 / \text{ndf}$ & 513.3 / 273 \\
  \hline
  $p_0$ & 2438 $\pm$ 0.1771 \\
  \hline
  $p_1$ & 1116 $\pm$ 3.862 \\
  \hline
\end{tabular}
\end{center}

![Figure 5.2: Averaged combined detection rate (SBJ592+SBJ593) for the full spectrum of $^{60}$Co over 50 days. Data points in red and error bars in blue. A fit of the data is shown (red) and the expected activity is plotted (black). Both from eq. 5.1.](image)

The residuals of the activity of the full spectrum of $^{60}$Co from a fit with an exponential decay function with the known half-life is shown in figure 5.3. The residuals are the expected rate minus measured rate as defined in equation 1.5. If the data would fully agree with the expected activity the residuals will be randomly above and below the $R=0$ axis. In the second data set the activity is averaged 21.8 $\pm$ 2 Hz, or 0.9 $\pm$ 0.06 %, which is lower than expected. The measured background rate would need to decrease by $\pm$ 105% in rate to account for this difference, which probably means this
decrease must have at least one other origin. A maximum shift in the position of the source would change the detection rate by $0.9 \pm 0.04\%$ and could thus explain the difference.

![Graph: Difference between measured and expected rate](image)

**Figure 5.3:** Residuals of the activity of the full spectrum of $^{60}\text{Co}$ as a function of time from fit with an exponential decay function. Data points in red and error bars in blue. The right axis is the residuals in percentage of the activity.

### 5.2.2 Analysis of photopeak region

To lower the background contribution the rate within the photopeak region can be determined instead of the rate of the full spectrum. The background contribution is 1.01 Hz for the rate of the $E_\gamma = 1173$ keV peak region of $^{60}\text{Co}$. In figure 5.4 the averaged combined detection rate of detectors SBJ592 and SBJ593 for the $E_\gamma = 1173$ keV peak region of $^{60}\text{Co}$ over 50 days is shown. Again an exponential decay fit (red) and expected rate (black) is shown. The observed half-life value is 26(1)% lower than the reported $^{60}\text{Co}$ half life.

A plot of the residuals of the activity of the $E_\gamma = 1173$ keV peak region of $^{60}\text{Co}$ from a fit with an exponential decay function with the reported half-life is shown in figure 5.5. In the second data set the activity is averaged 3.2 ± 3 Hz, or 0.37 ± 0.35 %, which is lower than expected. The measured background rate would need to decrease by ± 320% in rate to account for this difference, which means this decrease must have at least one other origin. A shift in the position of the source could also explain the difference.
**Figure 5.4:** Averaged combined detection rate (SBJ592+SBJ593) for the $E_\gamma = 1173$ keV peak of $^{60}$Co over 50 days. Data points in red and error bars in blue. A fit of the data is shown (red) and the expected activity is plotted (black). Both from eq. 5.1.

**Figure 5.5:** Residuals of the activity of the $E_\gamma = 1173$ keV peak of $^{60}$Co as a function of time from fit with an exponential decay function. Data points in red and error bars in blue. The right axis is the residuals in percentage of the activity.
5.2.3 Stability of the setup

The stability of the measurement setup can have an influence on the measured activities. The position of the fitted $E_\gamma = 1173$ keV peak for all measurement points is shown in figure 5.6a. The right blue and red peak are from the first data set while the left red and blue peak are from the second data set. An decrease in peak position means an decrease in gain which in turn means a lower detection rate as shown in chapter 4. The gain change can be caused by a lower voltage of approximately 2 V. Since two volt is the precision of the Cean HV power supply this change could therefore not have been detected. The decrease of approximately 2 V would result in an efficiency increase of $\pm 0.15(7)\%$ which would result in a higher detection rate of $28 \pm 14$ Hz and $8 \pm 4$ Hz for the total spectrum and peak rate respectively. Therefore the decrease in voltage is likely the cause of the lower detected rates. The decrease in voltage did not change the FWHM (%) as can be seen in figure 5.6b.

![Distribution of fitted Peak Values](image1)

(a) Distribution of values of fitted $E_\gamma = 1173$ keV peak positions for SBJ592 (red) and SBJ593 (blue). A change in distribution can be seen between the different data sets.

![Distribution of FWHM](image2)

(b) Distribution of FWHM(%) of fitted $E_\gamma = 1173$ keV peaks for SBJ592 (red) and SBJ593 (blue). No change in distribution can be seen between the different data sets.

Figure 5.6
Chapter 6

Conclusion and discussion

6.1 Conclusions

Before the Modulation experiment commences, the performance of the detectors within the experimental setup are tested. When testing the seven detectors it was found that they perform according to their specifications. The PMT’s achieve high enough gain to be operated within their specified voltage range: gains of $10^5$ and $10^6$ while operated between 500 and 1000 V. At the optimum working voltage the energy resolution is good enough to identify single peaks within the spectrum: FWHM are $4.83-5.07(5)$ %, $7.29(5)$ % and $13.8(1)$ % is for $^{60}$Co, $^{137}$Cs and $^{133}$Ba respectively. With the total detection efficiency of between 30 and 60% for a set of detectors a large portion of the emitted $\gamma$-rays by the radioactive source will be detected. The choice of using a detector set instead of single detectors per source limits the change in detection rate for small shifts in the source position to $0.9 \pm 0.04$ %. For the Modulation experiment this is important since temperature changes can change the distance between the detector and source and also exhibits a yearly modulation. The 5 cm lead enclosure lowers the background rate from natural and cosmic origin from 223 Hz to 20 Hz. The background rate is lowered within the photopeak regions to 1.16, 2.13, 1.01 Hz for $^{133}$Ba, $^{137}$Cs and $^{60}$Co, respectively.

The activity measurement of $^{60}$Co over a period of 50 days shows a change in detection rate higher than expected. For the full spectrum the rate is $21.8 \pm 2$ Hz, or $0.9 \pm 0.06$ %, lower than expected. For the photopeak region of $E_\gamma = 1173$ keV the rate is $3.2 \pm 3$ Hz, or $0.37 \pm 0.35$ %, lower than expected. It was most likely caused by a decrease in the voltage, although a shift in the source position could also have been an influence. Further improvements on the experimental setup need to be made to exclude these shifts in the
order of 1% to reach a sensitivity to identify modulations in the order of 0.1%. The question if radioactive sources exhibit a yearly modulation is of course not yet answered and will hopefully be answered by the Modulation experiment in the following years.

6.2 Improvements

First of all the performance of the eight detector needs to be analyzed to see if it works according to the specifications. With a set of eight detectors and 4 sources some improvements can be made to understand and limit the possibility of rate shifts. For example building source holders will limit the possible shift in position of the sources and therefore limit the rate shift caused by maintenance of the setup. A power supply with a better voltage stability would lower changes in the efficiencies of the detectors. Another way to compensate for any changes in the voltage would be to use a power supply with a better precision. With a more precise measurements of the relation between voltage and efficiency around the optimum voltage value a change in voltage could be used to adjust the total detected rate afterwards accordingly.

Another problem that needs to be solved is the fact that in the current setup the peak position increases for higher detection rates. The detector’s manufacturer has shown that by placing an amplifier and shaper between the detectors and the data acquisition system this effect diminishes. The mechanism of this effect still requires further research since this experiment is build for measuring event rates with high precision.

Before starting the 24/7 data acquisition the activity measurement described in chapter five should be repeated after improvements have been made. Then it will be clear if the precision of determining the detection rate is better than 0.1% to identify yearly modulations of the source activity.
Figure 7.1: PTB data 1989-1996 with the Townsend method. "(ad) Relative residuals of fits of current raw data corrected for background and, if necessary, radionuclide impurities without using a ratio to any reference source. The yearly fluctuations from sources of the radionuclides: (a) $^{85}$Kr, (b) $^{108m}$Ag, (c) $^{152}$Eu and (d) $^{154}$Eu are depicted." Figure and text from [12]
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Decay type</th>
<th>Detector type</th>
<th>Radiation measured</th>
<th>Periodicity</th>
<th>Reference</th>
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<tr>
<td>$^3$H</td>
<td>$\beta^-$</td>
<td>Photodiodes</td>
<td>$\beta^-$</td>
<td>1 yr$^{-1}$</td>
<td>Falkenberg (2001)</td>
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<td>$^3$H</td>
<td>$\beta^-$</td>
<td>Liquid scintillator</td>
<td>$\beta^-$</td>
<td>1/d, 12.1 yr$^{-1}$</td>
<td>Shnoll et al. (1998)</td>
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<td>$^3$H</td>
<td>$\beta^-$</td>
<td>Liquid scintillator</td>
<td>$\beta^-$</td>
<td>~12.5 yr$^{-1}$</td>
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<td>$\beta^-$</td>
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<td>$^{22}$Na/$^{44}$Tia $\beta^+$</td>
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<td>$\gamma$</td>
<td>$\sim$12 yr$^{-1}$</td>
<td>O’Keefe (2012)</td>
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<td>$^{36}$Cl</td>
<td>$\beta^-$</td>
<td>Proportional</td>
<td>$\beta^-$</td>
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<td>$^{54}$Mn</td>
<td>$\kappa$</td>
<td>Scintillation</td>
<td>$\gamma$</td>
<td>Short term decrease</td>
<td>Jenkins and Fischbach (2009)</td>
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<tr>
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<td>$\gamma$</td>
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<td>$\beta^-\gamma$</td>
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<td>$\gamma$</td>
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<td>$\beta^-$</td>
<td>1, 11.7 yr$^{-1}$</td>
<td>Parkhomov, 2010a and Parkhomov, 2010b and Sturrock et al. (2012)</td>
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<td>$\gamma$</td>
<td>1 yr$^{-1}$</td>
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<tr>
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<td>Ion chamber</td>
<td>$\gamma$</td>
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<td>$\beta^-$</td>
<td>Scintillation</td>
<td>$\gamma$</td>
<td>1, d=1, 12.1 yr$^{-1}$</td>
<td>Baurov et al. (2007)</td>
</tr>
<tr>
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<td>$\beta^-\kappa$</td>
<td>Solid state (Ge)</td>
<td>$\gamma b$</td>
<td>1 yr$^{-1}$</td>
<td>Siegert et al. (1998)</td>
</tr>
<tr>
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<td>$\beta^-\kappa$</td>
<td>Ion chamber</td>
<td>$\gamma$</td>
<td>1 yr$^{-1}$</td>
<td>Schrader (2010)</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>$\beta^-\kappa$</td>
<td>Ion chamber</td>
<td>$\gamma$</td>
<td>1 yr$^{-1}$</td>
<td>Schrader (2010)</td>
</tr>
<tr>
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<td>$\alpha,\beta^-$</td>
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<td>$\gamma$</td>
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<td>$\gamma$</td>
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<td>Jenkins et al. (2009) and Sturrock et al., 2010b and Sturrock et al., 2011a</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>$\beta^-$</td>
<td>Solid state a</td>
<td>$\alpha$</td>
<td>1/d, 13.5, 1 yr$^{-1}$</td>
<td>Shnoll et al. (1998)</td>
</tr>
</tbody>
</table>

Table 7.1: Isotopes for which time-dependent decay rates have been observed. These results represent over sixty years of data collection. The $\kappa$ is electron capture. Table from Jenkins et al. (2012)[5]
Figure 7.2: PTB data 1989-1996 with the Townsend method. "(ac) Relative residuals of fits of current ratios using various combinations of radionuclide sources: (a) $R(\text{Kr}^{85})/R(\text{Eu}^{154})$, (b) $R(\text{Eu}^{152})/R(\text{Eu}^{154})$ and (c) $R(\text{Eu}^{154})/R(\text{Ag}^{108})$." Figure and text from \[12\]

(a) Spectrogram of $^{133}\text{Ba}$. There is evidence of an annual oscillation from 2003 to 2005. There is also evidence of the first harmonic of this oscillation.

(b) Time-phase display (phasegram) of measurements of the decay-rate of $^{133}\text{Ba}$. The phase of the annual oscillations is approximately 0.43, corresponding to a peak in the modulation on or about June 6.

Figure 7.3: On-line in colour. Spectrogram and phasegram of measurements of the decay-rates made at PTB over the time interval June 1999 to November 2008. The power, $S$, is represented by the color bar. Figures and text from \[5\]
Figure 7.4: "Normalized December 2006 $^{54}$Mn decay data along with GOES-11 x-ray data on a logarithmic scale. For $^{54}$Mn, each point represents the number of counts in the subsequent four hour period normalized to the average decay rate (see text), and has a fractional $\frac{1}{\sqrt{N}}$ statistical uncertainty of $\sim 2 \times 10^{-4}$. For the GOES-11 x-ray data, each point is the solar flux in W/m$^2$ summed over the same real-time intervals. The 12 December peak in the x-ray flux occurred at $\sim$21:37 EST." Figure and text from Jenkins et al., (2008)[9]
Figure 7.5: Complexity of interactions from photoelectric absorption in NaI(Tl). All possible origins of electrons and photons from a gamma-ray with energy above the binding energy of the K shell in sodium iodine. On the left the chances are given that the electron is excited from a K, L or M shell. Image from [19]
Figure 7.6: Electronics overview, all parts are to scale. The four ovals with wires pointing to them are the four holes in the isolation where the wires can go from the top of the outer box to the inner box.
Figure 7.7: Overview of the wiring schematics for all parts in the Modulation experiment. The UPS is an Uninterruptible Power Supply which can give power for a short while when the power from outside fails.
Bibliography


