

Light yield of nuclear recoils in liquid argon induced by scattered neutrons

Diploma-Thesis

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Abstract

It is well established that the largest fraction of matter in the universe is non-baryonic and non-luminous, the so-called “dark matter”. Promising dark matter candidates are weakly interacting massive particles (WIMPs).

A competitive type of detector used in the WIMP search measures scintillation light and ionization, which are produced in elastic collisions between liquid Ar or Xe nuclei and WIMPs. With such a detector it is essential to know the exact amount of light as a function of the nuclear recoil energy. In this work, the dependency is measured in a scattering experiment with monochromatic neutrons, which are generated by deuterium-deuterium fusion, then scattered off liquid argon atoms in a cryostat. The detection of the scattered neutrons and measurement of the scattering angle is performed using organic liquid scintillators.

The VUV scintillation light (128 nm) of liquid argon must be converted to light of a longer wavelength before it can be detected by conventional photomultiplier tubes (PMTs). This conversion is performed by a tetraphenyl-butadiene coating on the PMT glass window and the side reflectors. The efficiency of the wavelength shifter coating was optimized to improve the light yield.

Furthermore, the PMT calibration method has been improved significantly. Subject of this thesis is also the setup of the trigger for the scattering experiment including a pulse shape discrimination module for organic liquid scintillators. In addition a new data acquisition system has been installed and commissioned. The calibration of the argon cell was performed with γ sources. A first result for a neutron scattering measurement at an angle of 65° (70 keVr) is consistent with other experiments. The energy resolution and the precision of the neutron scattering experiment was significantly improved, mainly by rebuilding the cryostat with less surrounding material and a better wavelength shifter coating.

A precise measurement of the light yield of nuclear recoils in liquid argon is expected within the next few months, after the installation of a continuously running cryogenic cooling system.

Zusammenfassung

Man geht davon aus, dass der überwiegende Anteil an Materie im Universum aus nicht-baryonischer, nicht-leuchtender, sogenannter „dunkler Materie“ besteht. Vielversprechende Kandidaten hierfür, sind massive, schwach wechselwirkende Teilchen: sogenannte WIMPs.

Eine sehr erfolgversprechende Art von Detektoren bei der Suche nach WIMPs nutzt das Szintillationslicht und die Ionisationsladung, die bei elastischer Streuung an Atomkernen in flüssigem Ar oder Xe erzeugt werden. Für diese Experimente ist es erforderlich die nichtlineare Lichtausbeute als Funktion der Rückstoßenergie des Kernes genau zu kennen. Diese Abhängigkeit wird mit Hilfe eines Neutronen-Streuexperimentes gemessen. In dem in dieser Arbeit beschriebenen Experiment werden die dazu notwendigen monochromatischen Neutronen durch Deuterium-Deuterium Fusion erzeugt und an Flüssig-Argon Atomen in einem Kryostat gestreut. Die Detektion der gestreuten Neutronen sowie die Messung des Streuwinkels geschieht mit Hilfe von organischen Flüssigszintillatoren.

Das im Flüssig-Argon entstehende VUV Szintillationslicht (128nm) muss zu Licht einer größeren Wellenlänge konvertiert werden, bevor es mit gängigen Photomultipliern (PMTs) detektiert werden kann. Diese Konvertierung geschieht durch eine Tetraphenyl-Butadiene Beschichtung auf dem PMT Glasfenster und auf den Seitenreflektoren. Um die Lichtausbeute bei möglichst kleinen Rückstoßenergien messen zu können, wurde in dieser Arbeit die Effizienz der wellenlängenschiebenden Beschichtungen optimiert. Des weiteren wurde eine verbesserte Methode zur Kalibrierung der PMTs entwickelt. Diese Arbeit beinhaltet auch den Aufbau des Triggers für das Streuexperiment, welcher ein Modul zur Pulsform-Diskriminierung von organischen Flüssigszintillatoren beinhaltet sowie die Installation und Inbetriebnahme eines neuen Datenerfassungssystems. Die Kalibrierung der Argon Zelle wurde mittels γ Quellen erzielt. Des weiteren wurde eine Streumessung mit Neutronen für einen Streuwinkel von 65° durchgeführt und ein erstes Ergebnis für die Lichtausbeute von Rückstoßkernen bei 70 keV erhalten, welches mit aktuellen Messungen konsistent ist. Zur Verbesserung der Energieauflösung und der Präzision des Neutronen-Streuexperimentes wurde der Messaufbau erheblich verbessert, hauptsächlich durch Reduktion von umgebendem Material und verbessertem Wellenlängenschieber.

Eine präzise Messung der Lichtausbeute von Rückstoßkernen in Flüssig-Argon wird in den folgenden Monaten erwartet, nach dem noch fehlenden Einbau eines kontinuierlich arbeitenden Kryo-Kühlsystems.

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Chapter 1

Introduction

This chapter gives a short introduction to dark matter and the methods for its direct detection. Special emphasis is given on the scintillation mechanism in liquid argon.

1.1 Dark matter

There are many astrophysical observations giving hints to the existence of matter which is not baryonic and does not emit or absorb light. The existence of “dark matter” was discussed already in 1933, when Fritz Zwicky discovered that the mass in the Coma cluster must be several times the luminous mass, in order to explain the observed velocity dispersion [1]. Today the existence of dark matter is well-established, observations show that the luminous part of some galaxies is embedded into a dark matter halo. The most promising candidates for dark matter particles are so called weakly interacting massive particles (WIMPs). If they exist, WIMPs should also interact with ordinary matter with a cross section on the order of the weak scale. According to most theories beyond the standard model, the WIMP mass is on the order of $10 \text{ GeV}/c^2$ to a few TeV/c^2 . A popular WIMP candidate is the neutralino from a constrained minimal supersymmetric standard model [2].

1.1.1 Hints to dark matter

A strong hint to the existence of dark matter comes from the rotational velocity of stars in the galactic disk of spiral galaxies as a function of the distance from the center of the galaxy. Such “rotational curves” are different from what is calculated using the visible mass and interstellar gases. A measurement of the rotational curve of the galaxy NGC 3198 together with a fit to a model which includes both dark and baryonic matter is shown in figure 1.1. Only if dark matter is included, the observed rotational curve shows good agreement with the model.

Another strong hint is given by observations of the cluster 1E 0657-558 known as the

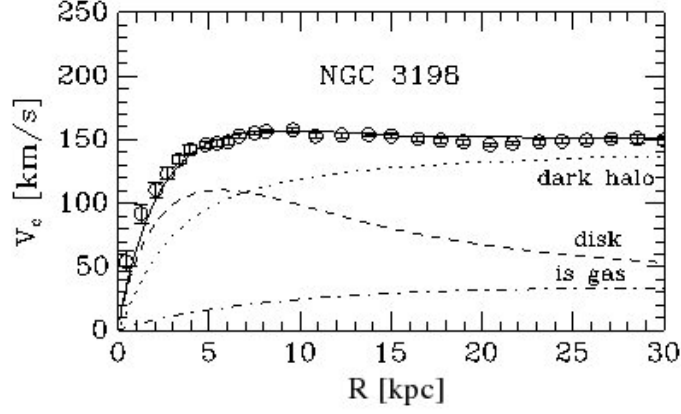


Figure 1.1: Measured rotational curve of the galaxy NGC 3198 (circles). The solid line shows the result from the fit of a model which includes dark matter. The single mass components (dark matter, stellar disk and interstellar gas) of the fit are also shown [3].

bullet cluster, where two clusters consisting of baryonic matter and dark matter collided. The dark matter components have little interaction whereas the interstellar gas of the two clusters, which yield the dominant part of the baryonic mass, did interact. This led to a slow down of the interstellar gas clouds compared to the dark matter distributions and resulted in a spatial separation of the both. This collision took place about 100 Myr ago [4]. The mass distributions from baryonic and dark matter of the bullet cluster have been measured using X-ray and optical imaging as well as weak gravitational lensing. The contour lines of the mass distribution are shown in green in figure 1.2. The image in figure 1.2 (b) shows the plasma density, corresponding to the baryonic mass distribution, which was measured using X-ray imaging. It is clear that the plasma clouds have been slowed down compared to the mass distributions seen by weak lensing. This shows that the dominant dark matter components of each cluster have passed through each other without interaction, as the baryonic components have slowed down.

1.1.2 Direct dark matter search

Dark matter particles are constantly crossing the earth. Their local density is $\sim 0.3 \text{ GeV}/\text{cm}^2$ [5]. As the mass of dark matter particles is expected to be on the order of the mass of a nuclei or higher, the interaction probability with nuclei is expected to be significantly higher than with electrons. Direct detection experiments employ different techniques to detect nuclear recoils from WIMP elastic scattering. The recoil energy is expected to be in the range of 0 to 100 keV. As the event rate caused by WIMP interaction is very small, it is important to have extremely low radioactive background. For this reason all direct detection dark matter experiments are installed in underground laboratories.

One type of detector uses scintillation crystals. If a WIMP hits a nucleus in such a crystal it causes the emission of photons which can be detected by photomultiplier

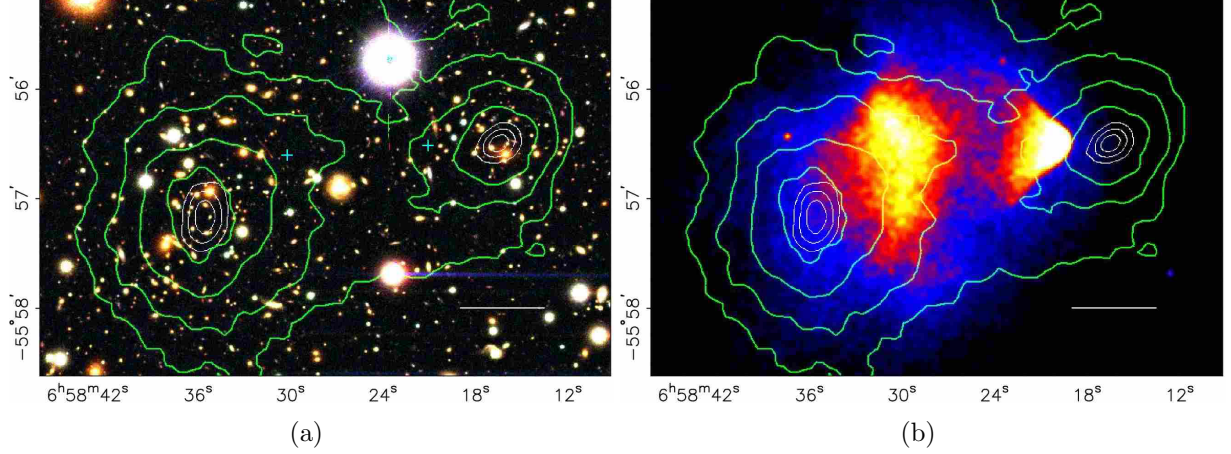


Figure 1.2: The Bullet Cluster. Mass density contour lines from weak gravitational lensing are superimposed to an optical image in (a) and an X-ray image in (b). The white contour lines show the center of mass at 68.3 %, 95.5 %, and 99.7 % confidence levels. The white bar corresponds to 200 kpc [4].

tubes. Such events can be caused as well by cosmic rays and radioactive impurities. DAMA/LIBRA is such an experiment using NaI crystals. They measure an annual modulation of the rate of a signal in an energy range of 2 to 6 keV [6]. This could be explained by the motion of the earth inside the dark matter halo. Our sun system is moving inside the milky way, but the dark matter halo is expected to be at rest. As the earth is moving around the sun, the speed of the earth relative to the dark matter halo and with this the flux of dark matter particles is annually modulated. A problem with the result is that the parameter space where DAMA/LIBRA measures the signal is excluded by other experiments (see figure 1.3).

The CoGeNT experiment uses a germanium detector. Their measurement is showing an excess of events at the low energy end of their detection range, which could be explained by a dark matter signal, but is hardly distinct from the background [7]. The parameter space favored by this signal is shown in cyan in figure 1.3.

Cryogenic semiconductor detectors like CDMS measure phonons and ionization. The exclusion limit obtained by CDMS is shown in figure 1.3.

Another very promising type of detector uses liquid noble gases as scintillation material. An important advantage of this type of detector is that they are easily scalable. The most sensitive noble liquid based dark matter detector currently existing is Xenon100. It has a target of 100 kg liquid Xe. Its current exclusion limit is shown in figure 1.3. An important detector using a liquid argon target is the ArDM detector. It is currently under construction and has an active volume of 850 l. The scintillation process in liquid Ar is described more detailed in section 1.2.

Exclusion limits on dark matter particles are based on dark matter properties like velocity, spacial distribution and interaction properties in addition to the detector response.

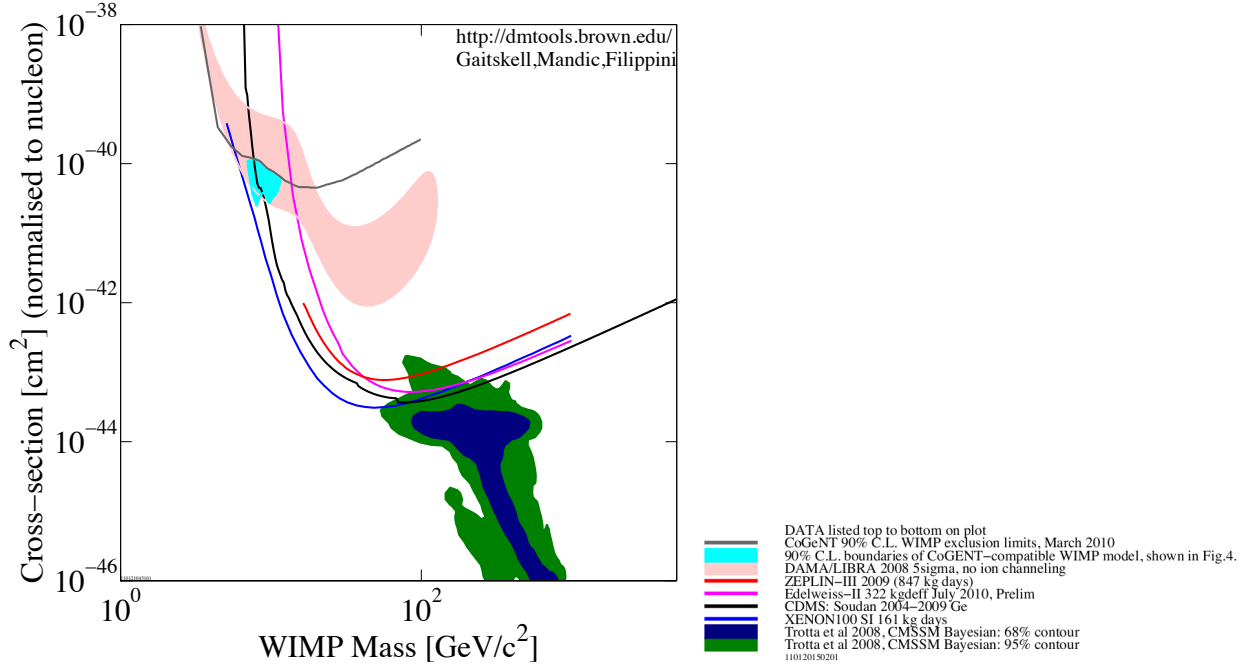


Figure 1.3: Exclusion plot for the spin independent WIMP-nucleon interaction cross section as a function of the WIMP mass[8]. The areas above the lines are excluded by the corresponding experiments. Shown as well is the region of the DAMA/LIBRA [6] and CoGeNT [7] signals and the region favored by a popular constraint minimal supersymmetric standard model (CMSSM) [9].

The errors on the limits are strongly dependent on the errors of the response of the detectors. Due to this it is very important to know the exact characteristics of the detection media and the detection mechanism.

The topic of this thesis is the light yield study of neutron induced nuclear recoils in liquid Ar, which show the same event signature as a WIMP interaction. In addition results from CoGeNT suggest a signal in the very low energy region motivating this improved study (compare to [10]).

1.2 Scintillation in liquid argon

The use of liquid Ar as detection medium for WIMPs or correspondingly to measure the energy of ionizing particles has several advantages. First it is possible to distinguish between electronic and nuclear recoils. In general in liquid Ar (like in any other particle detection medium) one can discriminate particles by looking at their different dE/dx , which determines the ionization density and in liquid Ar is strongly connected to the scintillation process. One group of particles are electronic recoils, these include γ rays, which are detected by their scattering or absorption by an electron, β radiation and muons. If dark matter particles exist, they will scatter off Ar nuclei (see as well section 1.1.2) and would hence produce nuclear recoil events, like scattered neutrons do. Electronic

recoil events form the main part of the background, even if the detector is situated in an underground laboratory. These events can be eliminated efficiently by pulse shape discrimination down to a certain detector dependent energy. This important property of Ar is not present in liquid xenon detectors.

Ionizing particles passing through liquid Ar produce excited argon atoms (Ar^*) and argon ions (Ar^+). These hit other Ar atoms and form excited molecules called excimers (excited dimer) (see figure 1.4) [11]. The ratio between excitation and ionization is independent of the ionizing particle [12] but the ionization density depends on dE/dx and is hence a lot higher for nuclear than for electronic recoils. The ionized molecules recombine with an electron to form neutral excimers, which eventually decay by emitting light with a wavelength of $(128 \pm 10) \text{ nm}$ [13]. This so called Vacuum Ultra Violet (VUV) light is not absorbed by neutral Ar atoms and can be detected with a technique described in section 1.3.

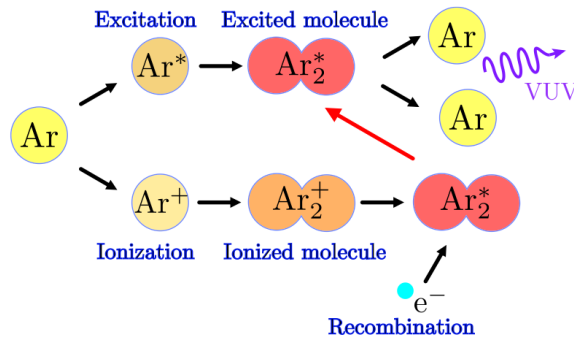


Figure 1.4: Illustration of the scintillation process in liquid Ar [14].

The excimers are produced in two different states, in the $^1\Sigma_u^+$ singlet and the $^3\Sigma_u^+$ triplet state. In liquid Ar the lifetime of the singlet state is $(7.0 \pm 1.0) \text{ ns}$ and $(1600 \pm 100) \text{ ns}$ for the triplet state [15] independent of the ionizing particle. The result is a signal with a high first component and a long second component. The ratio between light from the fast component and the total light produced is called component ratio (CR). It is ~ 0.8 for nuclear recoils and 0.28 for electron recoils, which makes it an important parameter for particle discrimination. The reason for the difference in the CR in liquid Ar is not well known, but might be due to super elastic collisions [12] where a singlet state excimer is changing the spin by colliding with an electron

$$^1\Sigma_u^+ + e^- \rightarrow ^3\Sigma_u^+ + e^-. \quad (1.1)$$

This process is expected to be more likely for electrons because of longer recombination times in lower ionization densities. The recombination probability is different for the same reason, which allows particle identification based on the light to charge ratio in addition to the CR.

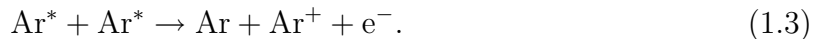
1.2.1 Quenching mechanisms in liquid argon

Ionizing particles convert a part of their energy into scintillation light, which is commonly expressed in units relative to light from electronic recoils. In general quenching can be expressed by the following relation:

$$q = \frac{\text{light output with quenching}}{\text{light output in absence of any quenching}}. \quad (1.2)$$

There are several types of quenching. In nuclear quenching a part of the energy of the ionizing particle is transferred into heat by elastic collisions with Ar atoms without exciting or ionizing them. The interaction cross section in an elastic collision is higher if the mass difference between the colliding particles is smaller, hence ions lose significantly more energy by this process than electrons do. Electrons can not lose their energy by heating up other electrons, simply because there are no free electrons to interact with.

Another quenching mechanism is the so called Bi-excitonic quenching and is a consequence of the so called “Penning process” [16], where two excited Ar atoms collide and produce a ground state Ar, an ionized Ar^+ and an electron



The probability for such an interaction depends on the density of excited Ar atoms. Both processes do not affect the lifetime of the molecular states, but reduce the light yield of nuclear recoils compared to electron recoils. As a result an Ar recoil produces approximately four times less light than an electron recoil with the same energy. This is described by the relative scintillation efficiency

$$\mathcal{L}_{eff} = \frac{\text{light output nuclear recoil}}{\text{light output electronic recoil}}, \quad (1.4)$$

usually measured relative to 122 keV γ -rays. Additionally there is a type of quenching related to impurities present in liquid Ar. Excimers colliding with impurities can decay non-radiatively by the process,



The longer an excimer exists the higher the probability to interact with an impurity. As the lifetime of the first component is short compared to the lifetime of the second component, the effect on the first component is negligible for relevant impurity concentrations, whereas the amount of light in the second component is significantly reduced [17]. This type of quenching provides an additional decay mode for the triplet state molecules which reduces the lifetime of the second component. This is an important property of liquid

Ar, which gives the opportunity to monitor the purity and to correct for this type of quenching. As the integral from zero to infinity of an exponentially decaying function is proportional to the decay time, the impurity quenching factor q_{τ_2} for the light emitted by decaying triplet state molecules B is given by the ratio

$$q_{\tau_2} = \frac{B_{\text{measured}}}{B_{\text{max purity}}} = \frac{\tau_2}{\tau_2^{\text{max}}}, \quad (1.6)$$

with τ_2^{max} the lifetime of the triplet state in absence of any impurities. For the quenching factor of the total integrated pulse height due to impurities q_{imp} one obtains

$$q_{\text{imp}} = \frac{A + B}{A + B_{\text{max purity}}} = \frac{A + B}{A + B/q_{\tau_2}}, \quad (1.7)$$

with A the total light emitted by singlet state molecules.

1.3 Detection of VUV light

The 128 nm scintillation light from liquid Ar would be absorbed by the PMT window and can not be detected directly using standard photomultiplier tubes (PMTs). To detect this VUV light, tetraphenyl butadiene (TPB) is used to convert the light to the spectrum shown in figure 1.5. This can be detected using conventional cryogenic PMTs. This wavelength shifter is situated in the experiment on the PMT windows as well as on reflector foils which surround the active volume (see as well chapter 3).

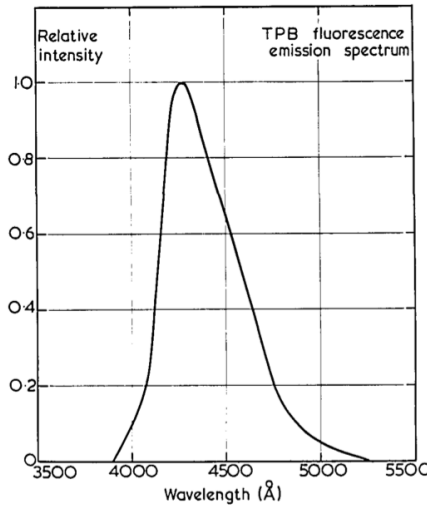


Figure 1.5: The emission spectrum of TPB illuminated by 253.7 nm light [18].

1.3.1 General properties of photomultiplier tubes

As the wavelength shifted light is detected using PMTs it is important to know some of their properties. If an incident photon hits the photocathode which is situated inside the PMT window it produces with a certain probability a photo-electron. The average number of produced photo-electrons divided by the number of incident photons is called quantum efficiency η . It depends on the wavelength of the incident light, the temperature and the material of the photocathode.

These primary electrons are accelerated onto the first dynode where they kick out secondary electrons (see figure 1.6). Some electrons are lost before hitting the first dynode. This is described using the collection efficiency β . The secondary electrons are accelerated towards the dynodes where they kick out further electrons. At the last stage the electrons are collected by the anode. An important value depending on the work function of the i^{th} dynode, the temperature and the energy of the incident electron is the secondary emission rate δ_i , which corresponds to the average number of secondary electrons divided by the number of primary electrons. The total gain G of the electron multiplication using n dynodes is given by:

$$G = \beta \cdot \prod_{i=1}^n \delta_i. \quad (1.8)$$

If all dynodes are identical and the applied voltage V in between the anode and the cathode is split equally between all dynodes, the gain is given by

$$G = \beta \cdot \delta^n = c \cdot V^{\alpha n}, \quad (1.9)$$

where c is a constant and α a value depending on the structure and the material of the dynodes [19]. The single photoelectron charge distribution is not symmetric.

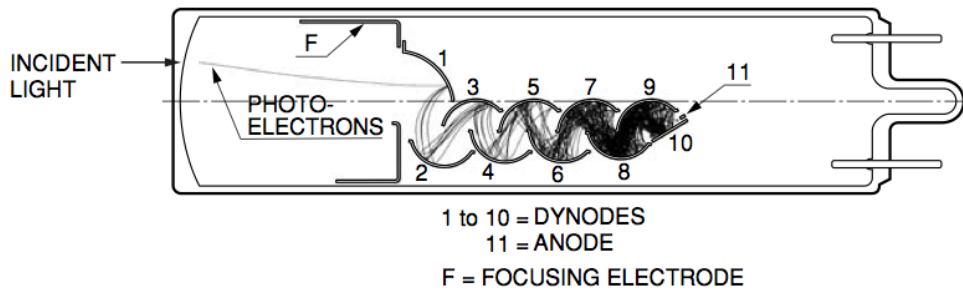


Figure 1.6: Functional principle of a PMT [19].

As equation 1.9 shows, the gain is very sensitive to fluctuations in the applied voltage. Fluctuations are produced by the difference between the number of incident and kicked-out electrons of one dynode. This smearing of the output signal is reduced using capacitors in parallel to the dynodes.

The energy resolution R of a PMT is given by:

$$R^2 \approx \frac{\delta}{N\eta\beta(\delta - 1)}, \quad (1.10)$$

with N the mean number of incident photons [19].

Thermally emitted electrons from the cathode produce single-photon like events. An electron thermally emitted from a dynode produces an event with a smaller pulse. These background events are randomly distributed in time.

Chapter 2

Experimental principles

The light yield of liquid Ar for nuclear recoils is studied using elastically scattered monochromatic neutrons. The scintillation light is detected by two photomultiplier tubes (PMTs) and the scattered neutrons by an organic liquid scintillator (LSc). A scheme of the basics of the experimental setup is shown in figure 2.1 (see also [20]).

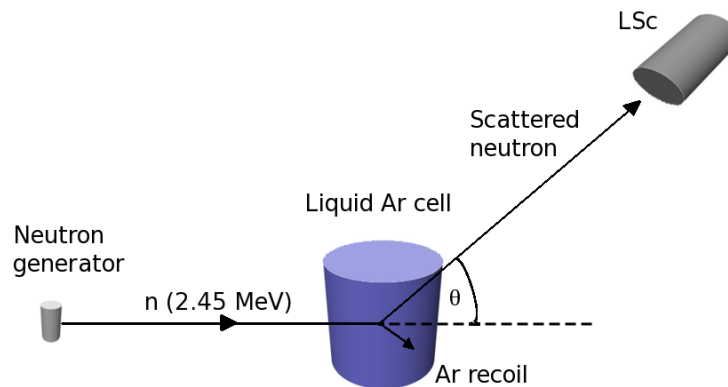
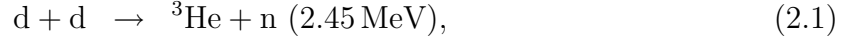


Figure 2.1: Principle setup of the scattering experiment.

2.1 The neutron generator

Monochromatic neutrons are generated by deuterium-deuterium fusion occurring in a plasma discharge in a volume of 36 mm height and 25 mm in diameter. The reaction chamber is filled with deuterium gas at a pressure of $\sim 10^{-2}$ mbar. This can be controlled by heating getter discs, which serve as deuterium reservoirs. By applying a voltage between 10 kV and 100 kV a deuterium plasma is created and fusion occurs by the tunnel effect. The decay

produces monochromatic neutrons or protons with equal rates by the reactions [21][22]:



Protons or neutrons are emitted isotropically. Both reactions occur with the same probability, but the produced protons are absorbed in the Al housing of the fusion zone. The amount of produced tritium (t) and ${}^3\text{He}$ can be neglected for further fusion.

The Al cylinder has cooling fins on its outer side and is actively cooled by air. At the direction of an exit collimator the fins are removed. At this “window” the mean thickness of the Al is 4.5 mm. Everything is covered by another Al cylinder with a 3 mm thick wall guiding the cooling air. The generator is surrounded by a 37 cm thick cylindrical neutron shield. It consists of 25 % $\text{CaB}_3\text{O}_4(\text{OH})_3\text{H}_2\text{O}$ and 75 % water extended polyester. A hole in the shield, placed in front of the Al window, collimates the neutrons for the scattering experiment. Outside the neutron shielding there is are layers of lead (1 mm) to stop Bremsstrahlung X-rays also produced by the plasma. In front of the collimator it has a thickness of 2 mm.

2.2 Nuclear recoils from neutron scattering

The neutrons with an energy of $E_n=2.45\text{ MeV}$ are elastically scattered on Ar nuclei. Knowing the scattering angle θ from the position of the LSc, the energy transferred to the nuclei E_{recoil} can be calculated using classical kinematics,

$$E_{\text{recoil}} = \frac{2E_n}{(1+A)^2} \left[1 + A - \cos^2\theta - \cos\theta\sqrt{A^2 + \cos^2\theta - 1} \right] \cong \frac{2E_n A}{(1+A)^2} (1 - \cos\theta), \quad (2.3)$$

where A is the atomic mass number of Ar (39.95 u). This dependency is shown in figure 2.2.

The interaction probability P_{int} for a 2.5 MeV neutron with an elastic cross section of $\sigma = 2\text{ b}$ [23] crossing the whole active volume ($l=74\text{ mm}$) of the liquid Ar cell is given by

$$P_{\text{int}} = 1 - \exp\left(-\frac{N_A \cdot \rho_{\text{LAr}} \cdot \sigma}{A_{\text{Ar}}} \cdot l\right) = 0.27 \quad (2.4)$$

with the Avogadro constant $N_a = 6 \cdot 10^{23}$ and the density of liquid Ar $\rho_{\text{LAr}}=1.4\text{ g/cm}^3$.

2.3 Fast neutron detection

Scattered fast neutrons are detected by a 5“ Scionix EJ301 organic liquid scintillator (LSc). The scintillator material contains 6 H atoms per 5 C atoms [24]. For a large

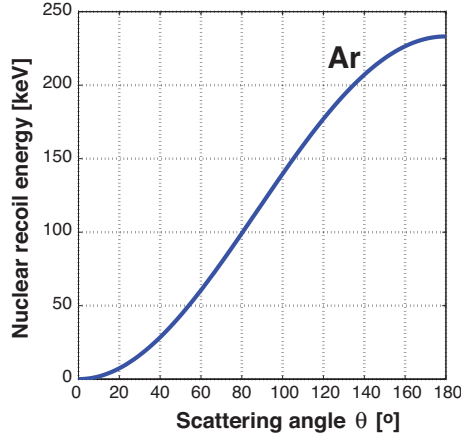


Figure 2.2: Ar recoil energy versus scattering angle for 2.45 MeV neutrons [20].

energy deposit it is important that the target has a mass similar to that of a neutron. A proton recoil from a neutron scattering on a hydrogen nucleus produces a light signal containing a slow and a fast component. This property is used to detect neutrons in the trigger stage by the pulse shape discrimination module MPD-4 described in section 2.5.1. In the contrary to liquid Ar, a nuclear recoil in the organic liquid scintillator has a higher fraction of light in the slow component than in the fast one in respect to electronic recoils [25]. The amount of light produced by C recoils is strongly suppressed due to the large mass difference and higher quenching [26].

The density of the scintillator is ρ_{LSc} 0.87 g/cm³ corresponding to a H density of

$$\rho_H = \rho_{\text{LSc}} \frac{6}{5 \cdot A_C + 6} = 0.079 \text{ g/cm}^3, \quad (2.5)$$

with $A_C = 12$ the atomic mass number of carbon. Using equation 2.4 the interaction probability of a neutron with a proton from the H atom in the 5" LSc ($l=13$ cm) is 0.84 using a cross section of $\sigma = 3$ b [27].

2.4 The liquid Ar cell

A scheme of the liquid Ar cell is shown in figure 2.3. The internal vessel contains very pure liquid Ar. The active volume with a diameter of 74 mm and a height of 78 mm is enclosed by two PMTs and a reflector. The PMTs from Hamamatsu of the type R6091-01 have a quantum efficiency of $\sim 15\%$ [28]. Between the reflector and the 2 mm thick stainless steel vessel wall are 40 mm of liquid Ar. The distance between the outside of the vessel and the inside of the external dewar is 45 mm.

Pure Ar gas is condensed into the inner vessel. The necessary cooling is provided by a mixture of liquid N₂ and liquid Ar in the external dewar. Ar has an evaporation

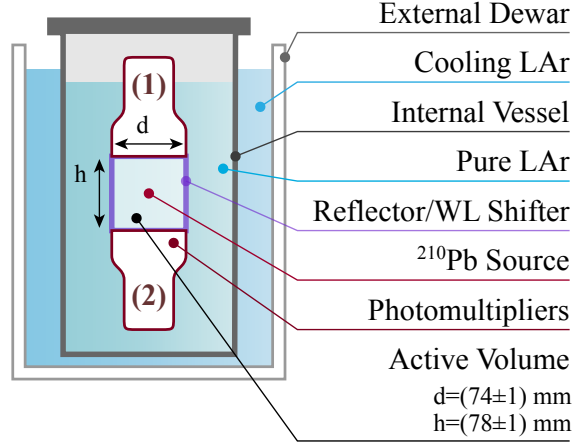


Figure 2.3: Scheme of the liquid Ar cell [35].

temperature of 87,3K and a melting point of 83.8K. The evaporation temperature of N_2 is 77.4K. For this reason Ar can not be liquified using only liquid N_2 . To achieve a temperature between the melting and the boiling point of liquid Ar, N_2 and Ar has to be mixed in the external dewar. The top of the dewar is covered by foam to prohibit condensation of water. Liquid N_2 has to be refilled manually, liquid Ar is automatically refilled using two platinum resistors as temperature sensors (PTs) inside the external dewar. One is $\sim 2,5$ cm below the border of the dewar and the other one $\sim 1,5$ cm lower. As soon as the liquid drops below both sensors (the resistances increase above an adjustable threshold) the Ar automatically refills until both are again covered by the liquid (resistance below the threshold).

Before filling, the cell was pumped for several days to a typical pressure of $\sim 5 \cdot 10^{-6}$ mbar. Then it was filled with Ar gas containing less than 0.5 ppm of H_2O , 0.1 ppm of O_2 , 0.1 ppm of H_2 , 0.1 ppm of CO_2 , 0.1 ppm of CO and 0.1 ppm of C_nH_m which is condensed into the inner vessel [29]. The level of the liquid Ar inside the cell is determined from the resistances of 7 PT sensors at different heights.

The active volume is surrounded by a reflector made of Tyvek (a polyethylene fibre membrane from DuPont). The reflector is covered with 1.0 ± 0.1 mg/cm² of the wavelength shifter TPB (see section 1.3). The PMTs are covered with TPB embedded in a Paraloid (a methyl-methacrylat-copolymer) matrix. Their photo cathods have a high sensitivity at the peak of the emission spectrum of TPB.

To perform gain calibrations of the PMTs (see section 2.7.1) an optical fibre connected to an LED (390 nm) is installed inside the active volume.

A spherical radioactive ^{210}Pb source with an activity of ~ 50 Bq is placed in the center of the sensitive volume. The decay scheme is shown in figure 2.4. This source is permanently installed in the setup.

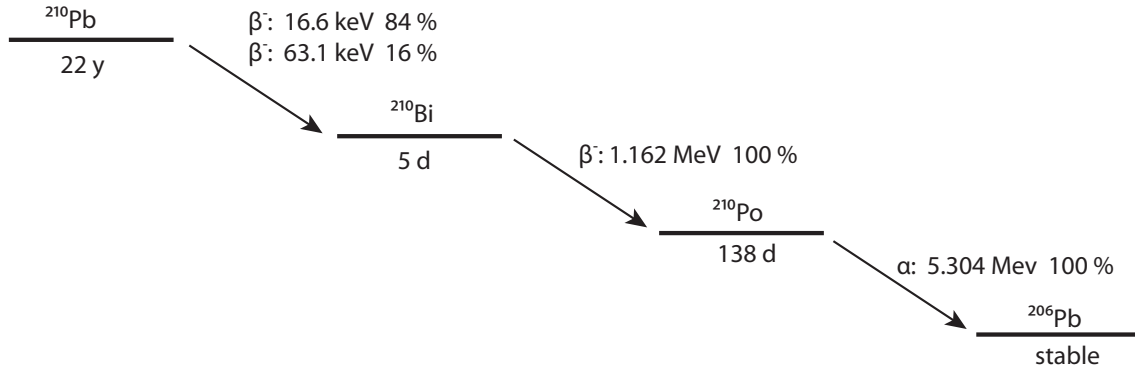


Figure 2.4: Decay scheme of the internal ^{210}Pb source. Values from [30].

2.5 DAQ principles

The DAQ system is based on the digital oscilloscope wavepro 735Zi from LeCroy with 4 input channels and 8 bit resolution running Windows. The input range of each channel can be set independently. The signal of each PMT is split by a passive splitter with an amplitude ratio of approximately 1:10. Both signals are recorded by a individual channels. This technique allows measurement of small events like single photo-electrons in one channel and at the same time big signals in the other channel (high dynamic range). Saturated (large) signals in the high gain trace are replaced by the corresponding signal from the low gain channel. This is done offline by a preprocessing program.

The oscilloscope software stores the data of all four channels of triggered events into a shared memory region. The principle concept of the DAQ software to access this shared memory was provided by the manufacturer. The code is written in C++ and has been extended by an optional event rejection algorithm and to write data in the desired format into a binary file. A header with fixed length is written at the beginning of each file containing the settings of the oscilloscope, which are read from the shared memory. The header contains as well information about the number of events rejected by the DAQ software, the threshold, the time when the file started and when it was completed, as well as a comment which can be entered when the software is started.

2.5.1 Trigger of the scattering experiment

A key device in the trigger for the scattering experiment is an NIM module of the type MPD-4 from MESYTEC, Germany. It is designed to discriminate proton recoils from γ events in organic liquid scintillators [31]. The component ratio (CR) as well as the integrated pulse height of each event are computed analogical. Here the CR corresponds to the ratio of light from the slow to light from the fast component, which inverts the bands compared to our definition used for Ar scintillation light.

The module can be set to generate a trigger if the CR of an event is above or below

a chosen value. Additionally a threshold for the pulse height can be set. The module has a fast and a standard mode of operation. In the standard mode it generates signals between 0 V and 8 V at the “TAC” and the “Ampl” output channel. The voltage of the TAC output corresponds to the CR and the Ampl output corresponds to the integrated pulse height. In this mode the dead time of the module is 1250 ns.

To improve the particle discrimination, the value of the CR measured by the MPD-4 module is internally modified, by the module itself, as a function of the integrated pulse height. This can be controlled by a parameter called walk, which was adjusted to obtain a strait upper edge of the n-band. The parameters of the module were adjusted using a LSc installed in front of the neutron generator. The result of the adjustments are shown in fig. 2.5.

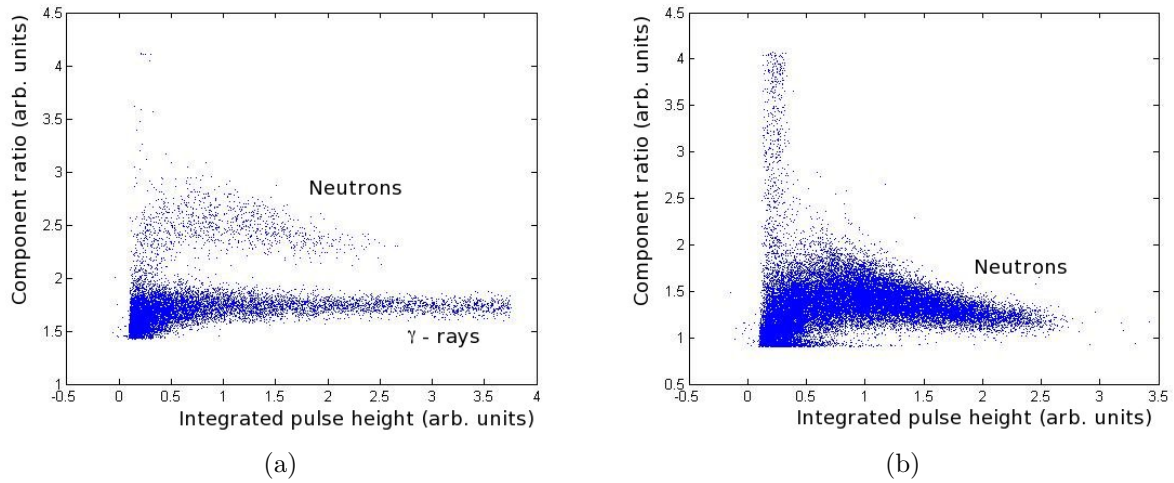


Figure 2.5: CR versus integrated pulse height in a 5“ liquid scintillator determined with the MPD-4 module. (a), The proton and the electron recoil bands after the adjustment of the “walk” parameter (band shape) but without a CR cut. (b), The neutron band after the adjustment of the CR threshold (higher total number of events than in (a)).

For the scattering experiment the module is used in the fast mode with a dead time of 250 ns. In this mode the TAC and Ampl. outputs are not available. The trigger output of the MPD-4 module delivers the external trigger for the DAQ (oscilloscope). Due to the processing time in the module this signal is delayed compared to the signals from the PMTs. The oscilloscope is set to accept an event if the signals of both PMTs are above a threshold at the same time and an external trigger occurs within 500 ns. The external trigger is always at the same position in the recorded time range. Due to this, information on the time between the event in the liquid Ar cell and the LSc is given by the position of the liquid Ar signal. This corresponds to the time of flight, which is used to reduce background in scattering experiments. The coincidence of both PMTs is used to suppress background originating from thermally emitted electrons in the PMTs.

Time jitter of the MPD-4 module trigger

It was tested if the trigger signal from the MPD-4 module has a time jitter which could induce an error on the time of flight. This was measured for pulses with different CRs and for different CR thresholds. To produce different pulse shapes the signals of two pulse generators have been added using a $50\ \Omega$ passive splitter. A triangular pulse with a width of 14 ns and a tail with a fall time of 100 ns was generated. Different CRs are obtained by changing the pulse height of the tail. Figure 2.6 shows superimposed trigger signals. The sampling rate of the oscilloscope was set to 10 GS/s. The full width half maximum of the trigger signal distribution is found to be ~ 0.2 ns independently of the pulse shape and the threshold on the CR. This is by far precise enough for our purposes, since already the transition time in the organic liquid scintillator accounts to a couple of ns.

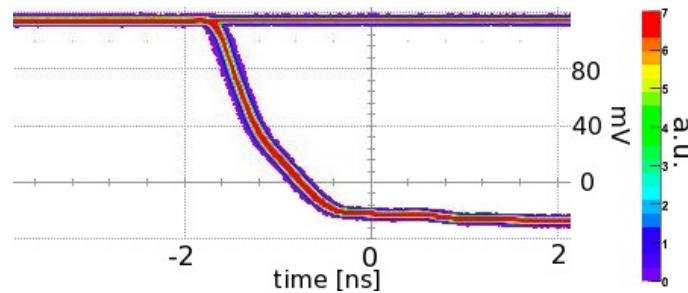


Figure 2.6: Several superimposed trigger signals of the MPD-4 module.

2.6 Offline data reconstruction

As described in section 2.5 the low and high gain traces are combined for each PMT. In the next stage important properties of the signals of both PMTs are extracted by a software written in C++ using the ROOT analysis package from CERN [32]. The obtained information is stored in ROOT trees. The further analysis is performed with ROOT macros specific for each analysis using the signal properties read from the ROOT files. The relevant properties of the software are described in this section.

2.6.1 Pedestal calculation and cluster finding

The pedestal is calculated for each event individually in a time range before the signal, which can be set in the configuration file. This calculation is done by an iterative method illustrated in figure 2.7:

- First the mean and the standard deviation of the signal trace in the time range are calculated.

- In the second step consecutive samples with an amplitude outside the range of $\pm 3 \cdot \sigma$ are accounted for as clusters. Additionally 10 points before and after the cluster are added to the cluster.
- A new mean and σ is calculated from the areas outside the signal clusters. This is done as long as the time interval is shortened by more than 1% compared to the previous loop.

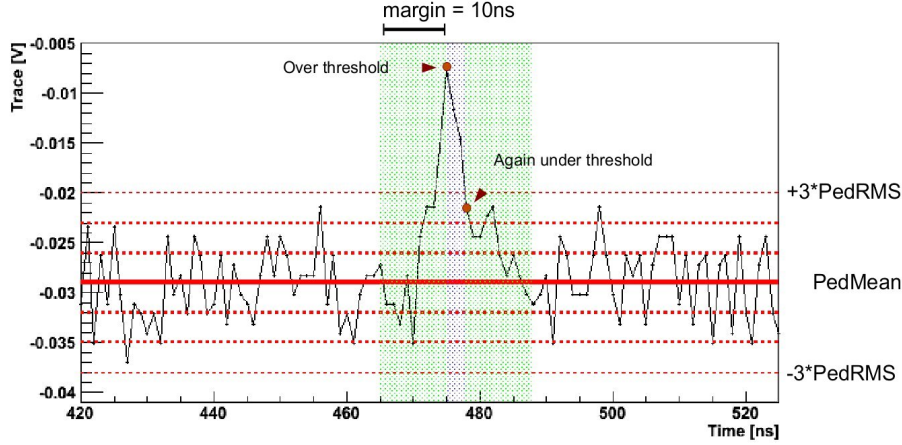


Figure 2.7: Illustration of the cluster finding algorithm (courtesy of Luca Scotto Lavina).

2.7 Measurements

Light yields are determined dividing integrated pulse heights by the mean single photo-electron charge (= gain). This can be measured using the integrated pulse height spectrum of pulses induced by an LED. Additionally, a method to perform the gain calibration using only information extracted from event tails is studied.

The light output per keV of electronic recoil events is measured using different radioactive γ sources. This electronic light yield determines as well the important low energy threshold. To indicate the type of the recoiling particle the unit keVr is used for recoiling nuclei and the unit keV electron equivalent (keVee) is used for electronic recoils. The end of this section describes a measurement of the scintillation efficiency of nuclear recoils $\mathcal{L}_{eff}$ at an energy of 69.8 keVr.

2.7.1 PMT gain calibration

The integrated pulse height spectrum of single photo-electron pulses is not symmetric, because of this the most probable value (MPV) is different from the mean. To determine the mean value of the spectrum it is important to have very little background contamination and no strong cuts into the single photo-electron distribution at the same time.

This is difficult, especially when using single photo-electrons from event tails. For this reason it is desirable to perform the calibration by determining the MPV of the single photo-electron spectrum and then correcting this value by the ratio between the mean of the background free single photo-electron spectrum and the MPV. This scaling factor might differ from PMT to PMT and is calculated for both PMTs individually as described in the two following subsections.

Both gain calibration methods are using the cluster finding algorithm described in section 2.6.1, especially the sum of the integral of all clusters in the selected time range. The integral of each individual cluster is not available but the number of clusters.

Every identified cluster with the wrong polarity originates from noise. Assuming that noise occurs with the same probability with positive and negative polarity, the number of single photo-electron clusters is calculated by subtracting the number of clusters with the wrong polarity from the number of clusters with the right polarity.

Gain calibration using an LED

The LED was powered by 10 ns pulses of 2.05 V with a rise and fall time of 3 ns. The voltage was set to create events of zero, one and two photo-electrons due to statistics (Poisson) without polluting the data too much with two photo-electron events. The number of clusters shown in figure 2.8 is calculated using a threshold of four times the standard deviation of the pedestal in the cluster finding algorithm. Overlaid is a normalized (to 10 000 events) Poisson distribution with an estimated expectation value. The number of events with one cluster is higher than the number of single photo-electrons expected from a poisson distribution, which is due the contribution of zero and and two photo-electron events. The readout was triggered by the pulse generator. The pedestal is calculated as described in section 2.6.1 starting the iteration with the whole event using a threshold of three times the standard deviation of the pedestal.

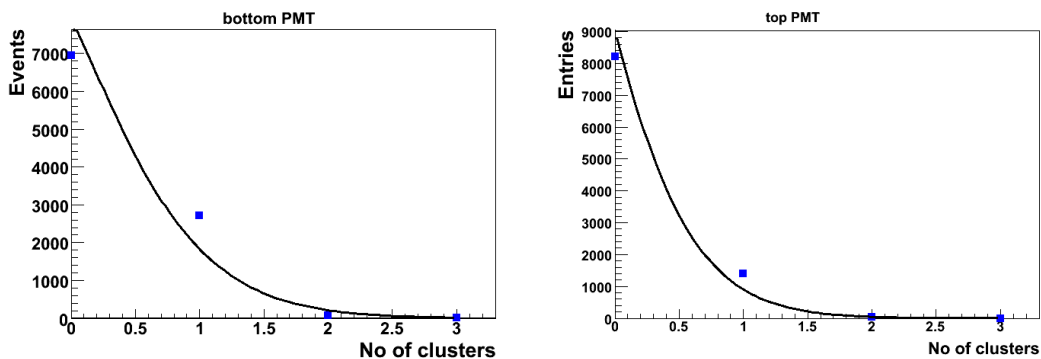


Figure 2.8: Number of clusters identified by the peak finding algorithm superimposed by normalized Poisson distributions. The mean values are 0.23 and 0.1 for the bottom and top PMT respectively (the bottom PMT is stronger illuminated).

The raw integrated pulse height spectrum for events with one cluster is shown in figure 2.9. To remove multiple and zero photo-electron events, only events with a cluster length ≥ 22 and ≤ 38 ns are selected. This integration time in ns is the number of samples over threshold + 20 (compare to figure 2.7). The cut is based on the cluster length histogram of both PMTs (figure 2.10). All cuts are summarized in table 2.1. The cut on the pedestal σ is relevant for the next subsection (compare figure 2.12).

No. of clusters	1
Cluster length	$\geq 22 \ \& \ \leq 38$
Pedestal σ	≤ 1 mV

Table 2.1: Summary of cuts for the gain calibration.

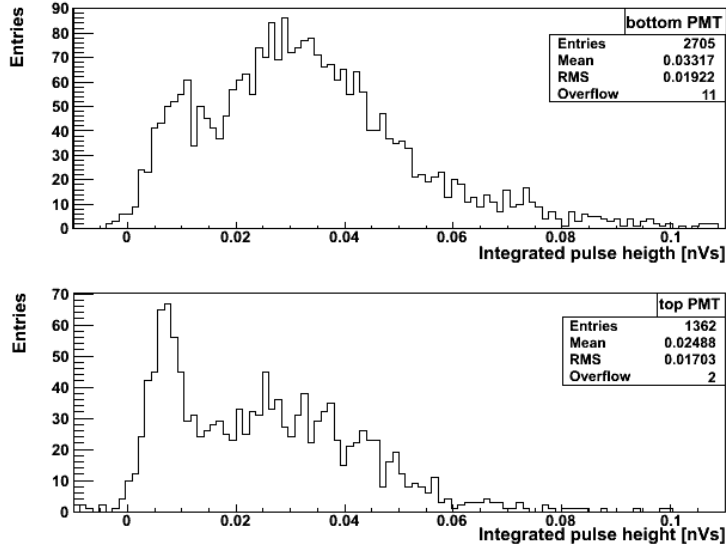


Figure 2.9: Integrated pulse height spectrum of single clusters from a LED measurement. The stronger illumination of the bottom PMT can be observed.

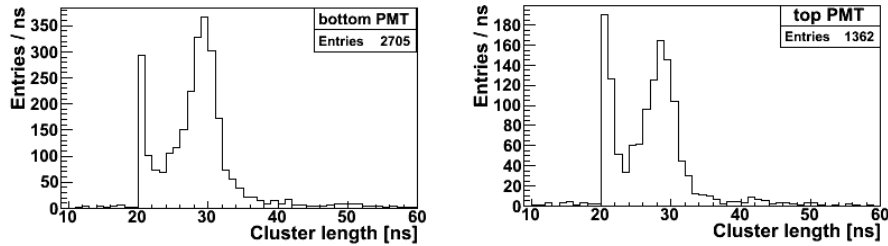


Figure 2.10: Cluster length of single cluster events.

The finally obtained spectrum for all events with one cluster is shown in figure 2.11. Six measurements with each 10 000 events were taken at different times during one day. The spectra of the different measurements were found similar to each other. The mean values of the histograms from each measurement are shown in table 2.2. The average value from the six LED measurement is (36.6 ± 0.45) pVs for the bottom and (31.5 ± 0.53) pVs

for the top PMT. The small fluctuations show that the gain of the PMTs did not change significantly. This is important to monitor, as a change in temperature (dynodes) can change the gain of a PMT.

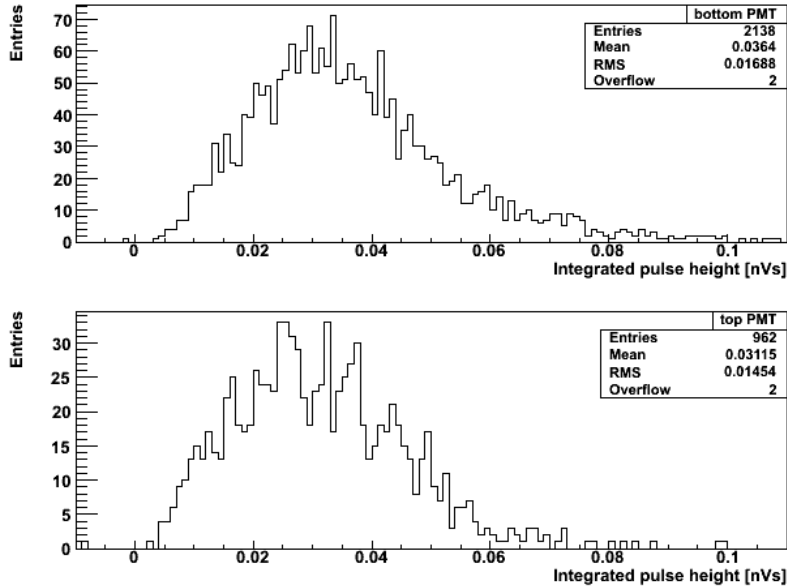


Figure 2.11: Integrated pulse height spectrum of mostly single photo-electron events from a LED measurement. The cuts are summarized in table 2.1.

Corrections on the gain calibration. The single photo-electron spectrum still contains some zero and two photoelectron events. Based on the histograms shown in figure 2.10 the number of zero photo-electron events n_{0pe} in the histograms in figure 2.11 was estimated to be 150 ± 60 events ($(7 \pm 2.8) \%$) for the bottom PMT and 160 ± 35 events ($(16.6 \pm 3.6) \%$) for the top PMT. The mean integrated pulse height of zero photo-electron events is estimated to be (10 ± 5) pVs for the bottom and (7 ± 3) pVs for the top PMT using figure 2.9 (due to noise).

bottom [pVs]	top [pVs]
37.0	32.1
36.4	31.2
36.4	30.7
37.2	32.0
36.4	31.7
36.0	31.3

Table 2.2: Mean values of the integrated pulse height spectra for both PMTs for six calibration measurements using a LED.

Assuming the number of two photo-electron events n_{2pe} in the single photo-electron spectrum is equal to the number of events with an integrated pulse height above twice the mean, one obtains a contamination of 122 ± 41 events ($(6 \pm 2) \%$) for the bottom and 36 ± 14 events ($(4 \pm 1.5) \%$) for the top PMT. The higher two photo-electron contamination of the bottom PMT can be explained with the LED pointing in the direction of the bottom

PMT. The single photo-electron mean is related to the measured mean by:

$$\text{mean}_{\text{measured}} = \frac{n_{0\text{pe}} \cdot \text{mean}_{0\text{pe}} + n_{1\text{pe}} \cdot \text{mean}_{1\text{pe}} + n_{2\text{pe}} \cdot 2 \cdot \text{mean}_{1\text{pe}}}{n_{\text{total}}}. \quad (2.6)$$

This gives for the real mean value of the single photo-electron spectrum

$$\text{mean}_{1\text{pe}} = \frac{n_{\text{total}} \cdot \text{mean}_{\text{measured}} - n_{0\text{pe}} \cdot \text{mean}_{0\text{pe}}}{n_{\text{total}} + n_{2\text{pe}} - n_{0\text{pe}}}. \quad (2.7)$$

Using the described estimations one obtains the corrected calibration values of

$$\text{mean}_{1\text{pe bottom}} = \frac{2138 \cdot 36.6 \text{ pVs} - 150 \cdot 10 \text{ pVs}}{2138 + 122 - 150} = (36.4 \pm 0.9) \text{ pVs}, \quad (2.8)$$

$$\text{mean}_{1\text{pe top}} = \frac{962 \cdot 31.5 \text{ pVs} - 160 \cdot 7 \text{ pVs}}{962 + 36 - 160} = (34.8 \pm 1.0) \text{ pVs}. \quad (2.9)$$

The errors on this values have been calculated according to the Gaussian error propagation law using the errors mentioned in the text. The effect of the 4σ cut from the cluster finding algorithm is neglected.

Gain calibration using single photo-electrons from event tails

The previously described LED measurements have been taken between measurements with different radioactive sources. In this subsection single photo-electron events are extracted from event tails. A time range for the extraction of single photo-electron events is chosen to contain exactly one photo-electron for as many events as possible. This selection is related to the cluster finding algorithm, which calculates only the sum of the integrals of all clusters (compare introduction of section 2.7.1). The best performance was found for a time interval of 3700 ns to 4200 ns and 1500 ns to 2500 ns after the peak for electronic recoil events and α events respectively.

The pedestal is calculated for the whole time interval excluding the clusters. Events with a standard deviation bigger than 1 mV are removed. The effect of this cut is small (see figure 2.12), for the LED calibration it is negligible. The effects on the spectra obtained from a measurement with a ^{109}Cd source of the number of clusters and cluster length cuts are shown in figure 2.13. The final spectra (figure 2.14) were obtained applying the same cuts as for the calibration with the LED (table 2.1). The MPV was determined from a Gaussian fit to the top part of the spectra. These values from five different measurements are summarized in table 2.3. The average MPV is $(31.4 \pm 0.2) \text{ pVs}$ and $(30.8 \pm 0.3) \text{ pVs}$ for the bottom and top PMT respectively.

The scaling factor is calculated using the corrected mean from equation 2.8 and 2.9 and the average of the MPVs in table 2.3 for each PMT separately. It is found to be 1.161 ± 0.029 for the bottom PMT and 1.129 ± 0.033 for the top PMT. The scaling factors of the two PMTs are the same within their errors. This shows that the effect of

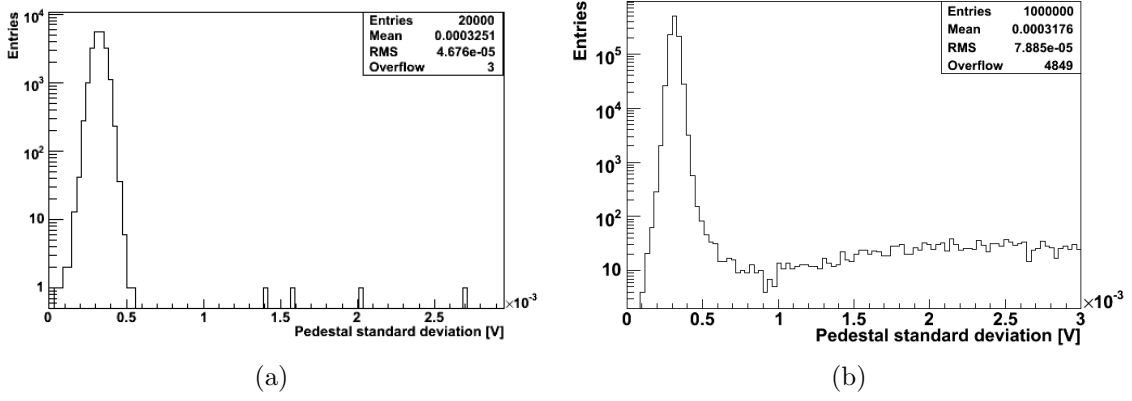


Figure 2.12: Histogram of the pedestal σ containing all events in a measurement with (a) the LED and (b) a ^{109}Cd γ source.

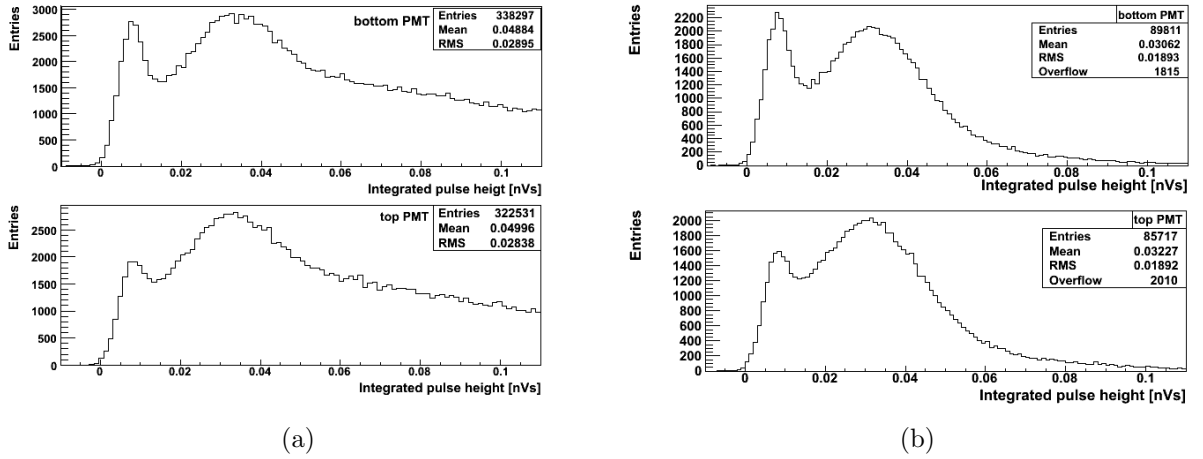


Figure 2.13: (a), Raw integrated pulse height spectra and (b) only events with one cluster.

possible differences in the hardware of the PMTs is negligible within the errors and allows to use the same scaling factor

$$s = \frac{1.129 + 1.161}{2} = 1.145 \pm 0.022 \quad (2.10)$$

for both PMTs.

These results show that it is possible to do the gain calibration with good precision using MPVs of the single photo-electron distributions extracted from event tails. This makes the calibration significantly easier and faster. A further improvement would be to store the integral of all clusters separately. This would significantly increase the statistics, which is especially important for scattering measurements with low rates.

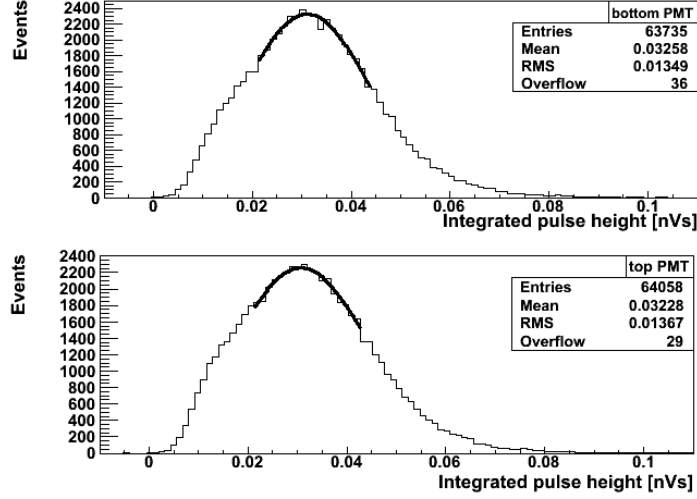


Figure 2.14: Final integrated pulse height spectra with a Gauss fit.

Source	MPV bottom PMT [pVs]	MPV top PMT [pVs]
Internal only	31.5	30.7
^{57}Co	31.3	30.6
^{57}Co	31.7	31.1
^{57}Co	31.2	31.2
^{109}Cd	31.1	30.5

Table 2.3: Most probable values of the integrated pulse height spectra from one cluster events.

2.7.2 Time calibration

For the pulse shape quality it is important to add the signals of both PMTs without a time shift. Both PMTs are connected with equal cable length. To correct differences in the propagation time the positions of the main peak (= highest sample) of all events of both PMTs are filled into histograms separately and fit with a Gaussian (see figure 2.15). The mean values of this two fits are subtracted and taken as the time shift in between the two PMTs. The time difference is rounded to a precision of 1 ns (the time resolution of the DAQ).

Only events with non saturated pulses, bigger integral than 0.2 nVs and a peak value higher than 0.02 V were used. The time difference obtained by this analysis is 2 ns. It was the same for all analyzed measurements taken with the present connection.

2.7.3 Light yield of electronic recoils – γ calibration

The light yield of electronic recoil events was determined using the photopeak from 60 keV γ rays of ^{241}Am and the Compton edges of 511 keV and 1275 keV γ rays from ^{22}Na and 662 keV from ^{137}Cs . These sources are placed outside the external dewar. The integrated

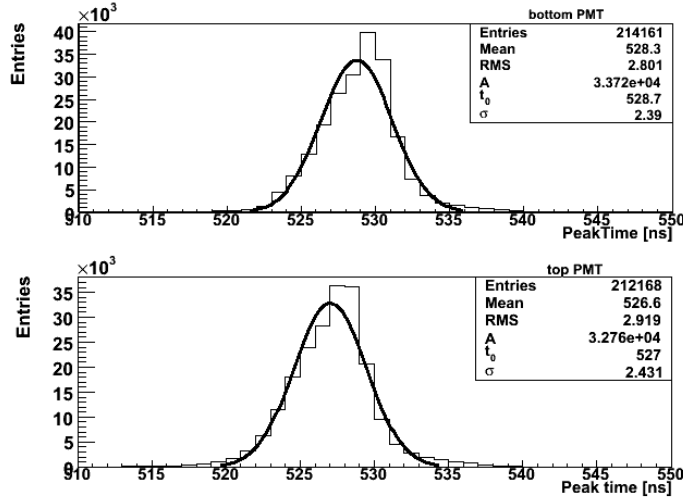


Figure 2.15: Peak times of both PMTs with Gaussian fits measured with the internal and a ^{57}Co source.

pulse height spectra are poluted by background and the energy resolution is low (see figure 2.16). Reasons for this are discussed in section 2.8.

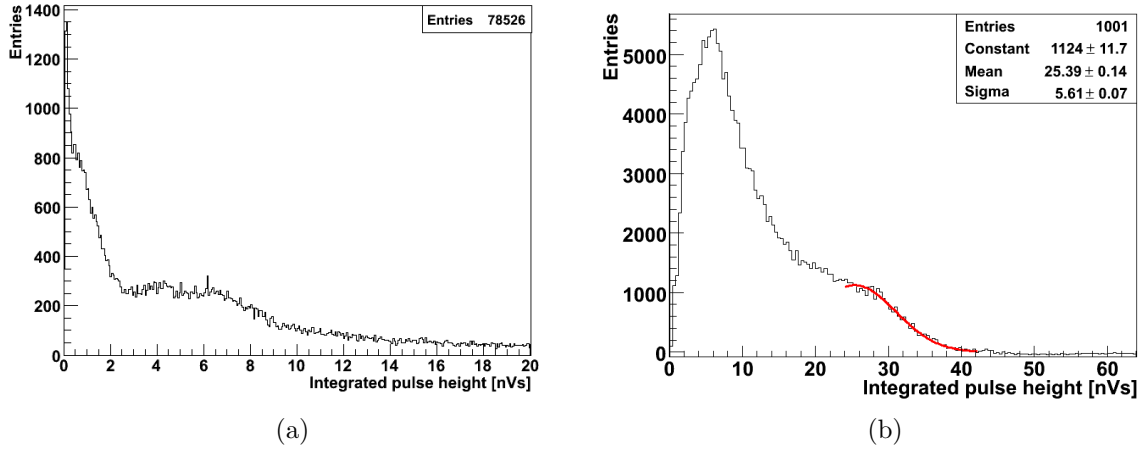


Figure 2.16: Integrated pulse height spectrum scaled to a gain of 28 pVs/pe from (a) 60 keV γ rays emitted by ^{241}Am and (b) from the 662 keV γ rays emitted by ^{137}Cs .

The Compton edge of the integrated pulse height spectrum is fit with half of a Gauss distribution. The integrated pulse height at 1/2 of the height of this fit is taken as the position of the Compton edge. Based on [33] the systematic error of this method is estimated to be 4 %. An error free mean value of the Gaussian fit function would imply, that the statistical error is the fit error of σ and vice versa. Neglecting the correlation, the total error on the position of the Compton edge is estimated to be the square root of the two fit errors and the systematic error quadratically summed. This total error is dominated by the systematic error.

The broad photo peak from the 60 keV γ rays (figure 2.16 (a)) could be interpreted as two peaks. For this reason the error on this value is estimated to be 15 %.

These measurements are taken at a τ_2 of (1366 ± 70) ns. The γ calibration values determined from the integrated pulse height spectra are extrapolated to a τ_2 of 1600 ns using equation 1.7 on p.9. The values of $A = 3.808 \pm 0.012$ and $B = 13.65 \pm 0.1$ are obtained from a fit to the mean trace of the ^{22}Na measurement¹. The fit function is the sum of two exponential decays describing the decay of the fast and the slow component convoluted with a Gauss distribution accounting for the time resolution of the PMTs (for more information see equation 3 on page 15 of [34]). The resulting impurity quenching factor is $q_{\text{imp}} = 0.882 \pm 0.036$.

The results from all 4 measurements are shown in figure 2.17. The light yield is obtained from the slope ($= (2.129 \pm 0.049)$ pe/keVee) of a linear fit passing the origin. Extrapolated to a lifetime of 1600 ns we finally obtain (2.41 ± 0.11) pe/keVee. The measured light output as a function of the electronic recoil energy is linear despite the low energy resolution of the integrated pulse height spectra.

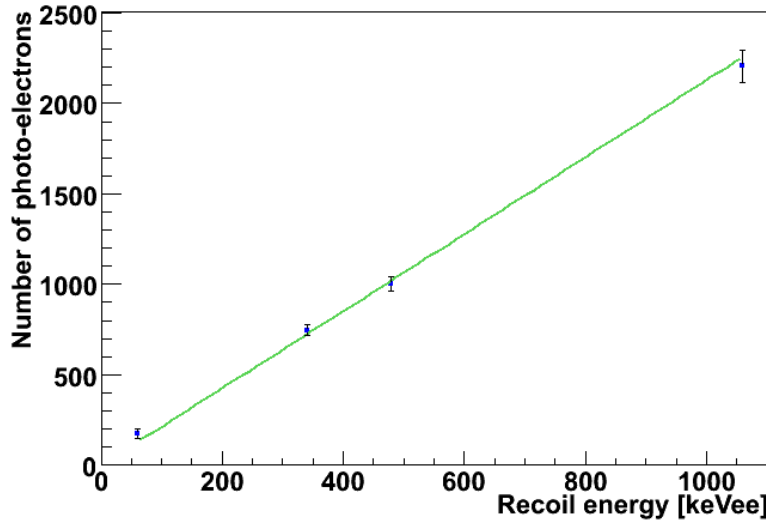


Figure 2.17: Measured no. of photo-electrons as a function of the electronic recoil energy from different γ sources (see text) together with a linear fit at $\tau_2 = (1366 \pm 70)$ ns).

2.7.4 A first measurement of the light yield of Ar recoils

In a first measurement of the nuclear light yield, the liquid Ar cell was set (67 ± 2) cm from the neutron generator (center to center). A 5" LSc was placed at an angle of $(65 \pm 3)^\circ$ and a distance of (50 ± 1) cm from the center of the liquid Ar cell to the face of the LSc. According to equation 2.3 on p.13 this corresponds to a mean recoil energy

¹Courtesy of William Creus

of (69.8 ± 5.5) keV. The speed of the scattered neutron (2.4 MeV) is $v_n=2.1$ cm/ns. This corresponds to a time of flight from the cell to the LSc in the range of 22 to 32 ns due to the dimensions of the active volume and the LSc.

The trigger was setup as described in section 2.5.1. The threshold of both PMTs was set to a value below the height of an average single photo-electron pulse.

Neutron events are selected by removing all events with a component ratio smaller than 0.6 (see figure 2.18 (a)). To remove random coincidences and multiple scattered neutrons (longer path and lower velocity) only traces having the peak within a time window of 13 ns are selected. This corresponds to a cut on the time of flight (compare section 2.5.1). Additionally events with an asymmetry,

$$asym = |(\text{integrated pulse height PMT 1} - \text{PMT 2}) / (\text{sum of both PMTs})|, \quad (2.11)$$

larger than 50% are removed (see figure 2.18 (b)). This cut is used to remove poor reconstructed events. The final spectrum is shown in figure 2.19. The measured value from the gaussian fit is (41.5 ± 2.8) pe at a τ_2 of (650 ± 100) ns assuming an error of 3% on the single photo-electron calibration. The extrapolation to a lifetime of 1600 ns is done using

$$q_{\text{imp nuclear recoils}} = 0.79 + \frac{0.131}{\mu s} \cdot \tau_2 = 0.875 \pm 0.013 \quad (2.12)$$

taken from equation 5.17 on page 54 of [35]. This equation is derived from equation 1.7 and uses the values of A and B at a τ_2 of 1600 ns determined in a separate measurement. The extrapolated light output is (47.4 ± 3.3) pe. The resulting relative nuclear scintillation efficiency (equation 1.4) is

$$\mathcal{L}_{eff} = \frac{47.4 \text{ pe}}{2.41 \text{ pe/keVee} \cdot 69.8 \text{ keVr}} = 0.282 \pm 0.090, \quad (2.13)$$

using the light yield of electronic recoils obtained in section 2.7.3. This result is consistent with the results presented in reference [10].

The error on the scattering angle has been estimated using a Monte-Carlo simulation². The geometries and materials of the neutron generator, the shielding, the liquid Ar cell and the LSc are included. Not included is the liquid Ar cooling bath and the external dewar. The scattering angle in the active volume of all neutrons with a time of flight in the range of 19 ns to 35 ns is shown in figure 2.20. The time of flight cut on the simulation data implies an interaction in the active volume and the LSc. The standard deviation of this histogram is taken as an estimation of the error on the scattering angle due to the geometries of the setup. The total error on the scattering angle is $\pm(\sqrt{9.5^2 + 3^2})^\circ = \pm 10.0^\circ$. This

²The simulation was written in the group, the author of this thesis was only using it.

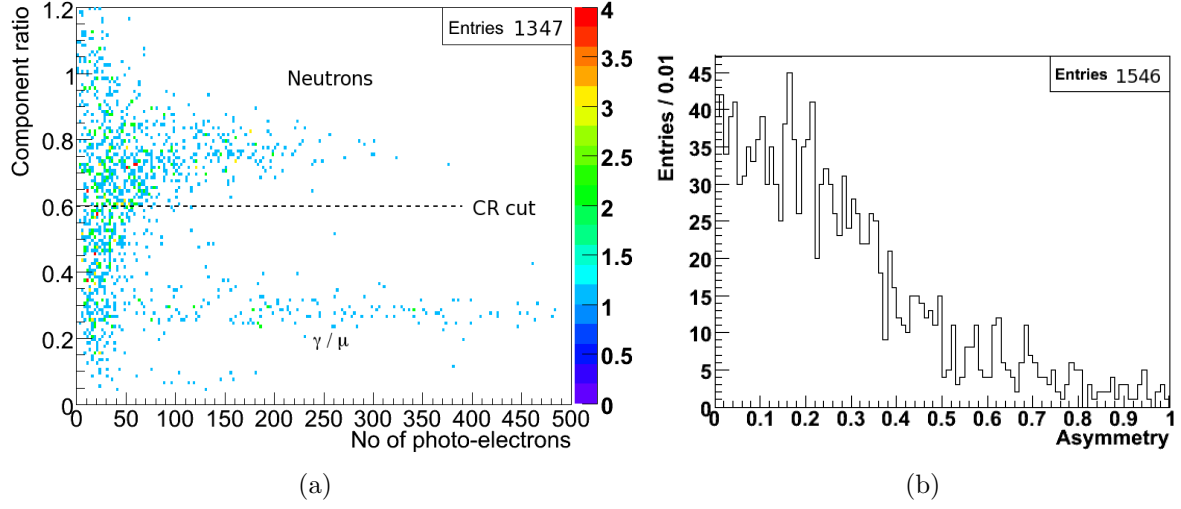


Figure 2.18: (a), The component ratio versus light yield and (b) a histogram of the relative asymmetry of the signal. Both plots are without previous cuts.

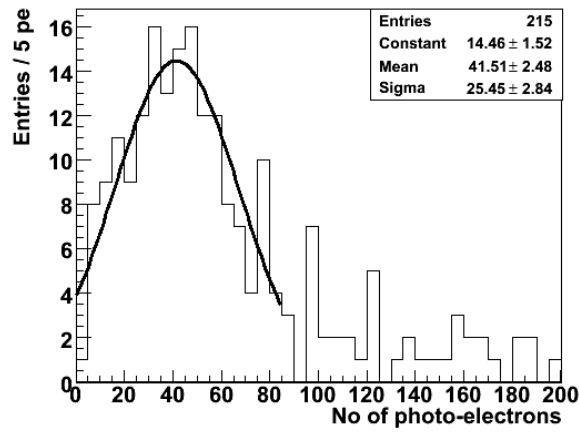


Figure 2.19: Light yield spectrum of recoil events from scattered neutrons measured at a purity of $\tau_2 = (650 \pm 100)$ together with a Gaussian fit, the fit values and fit errors.

corresponds to an error on the energy of

$$\Delta E_{\text{recoil}} = \frac{dE_{\text{recoil}}}{d\theta}(65^\circ) \cdot \Delta\theta = \pm 18.5 \text{ keV}. \quad (2.14)$$

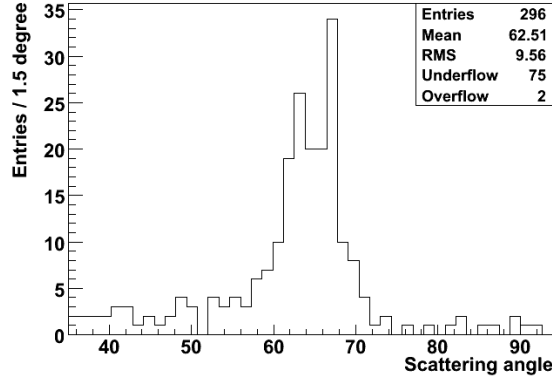


Figure 2.20: Scattering angle in the active volume for all events with an interaction in the active volume and the LSc and a time of flight in the range of 19 ns to 35 ns, obtained from a Monte-Carlo simulation.

The error on $\mathcal{L}_{eff}$ for recoil energies in the range of 55.3 keVr to 81.4 keVr ($57^\circ - 71^\circ$) amounts to 0.046 if the error on the scattering angle due to geometry is not taken into account (the error of 3° corresponding to 5.5 keV is still taken into account).

2.8 Conclusions

The measurements previously described show that the experimental setup is capable of measuring the light yield of scattered neutrons at low recoil energies. However it is important to increase the efficiency of the PMT coating and the side reflectors as much as possible to measure Ar recoils to as low energies as possible (compare section 1.1.2). This together with the uniformity of the coatings increases the energy resolution. The large amount of material surrounding the active volume has to be reduced to increase the angular resolution of the scattering experiment and to improve the γ calibration. Additionally the distances between the n-generator, the liquid Ar cell and the LSc must be increased. This reduction of the event rate together with the need of more statistics than in the measurement described in section 2.7.4 makes it necessary to measure several angles in parallel and to have a setup which can be run continuously (cryocooler). The PMT gain calibration procedure was significantly improved. Now it can be performed using the most probable value of a single photo-electron spectrum instead of the mean.

Chapter 3

Investigations on PMT coatings for liquid Ar scintillation light

The coatings of PMTs play an important role in the detection of VUV scintillation light (see as well section 1.3). The coating must be transparent for blue light emitted by the coated reflectors and must contain a wavelength shifter (TPB in our case) for direct light. Further more it has to resist cryogenic temperatures and ideally transports shifted light shifted to the photocathode. So far PMT coatings have not been sufficiently tested in liquid Ar. To measure the efficiency of coatings for scintillation light a special setup was designed and built using a 40 kBq ^{241}Am source producing 5.49 MeV α particles. Several coatings are described and analyzed in this chapter. The same PMT has been used for all coatings.

3.1 Measurement setup

The setup is shown in figure 3.1. A 3" square PMT (77 mm) is used, which faces downwards to a disc with a square hole of 74 mm edge length. The PMT is held in place by surrounding support thread rods, the filling pipe and two clamps. A sealed α source for cryogenic environments is situated (58 ± 2) mm below the PMT in a recess of the bottom disk. The two aluminum discs with a diameter of 88 mm have holes for the Ar flow. The whole setup is held by a cover situated on top of a stainless steel dewar with an inner diameter of 90 mm. The filling pipe for the liquid Ar ends below the lower disc. The gaseous Ar can leave the dewar through a 5 mm hole in the cover and through the exhaust pipe. During the beginning of the filling procedure the hole is covered by cloth. Air is lighter than Ar and should hence accumulate below the cover where it is blown out through this hole. This is expected to increase the purity. As soon as the source is covered by liquid, the hole is closed by several layers of black tape for a light tight setup and to avoid the diffusion of air into the cell. To reduce incoming light through

the exhaust pipe it has two bendings of 90° and a black rubber tube of ~ 50 cm at it's end. Dense black foam is glued to the base of the cover disk, which is pulled down by the holder. All feed-throughs are sealed with black silicone. Residual light from the outside is found to be less important than the dark count rate.

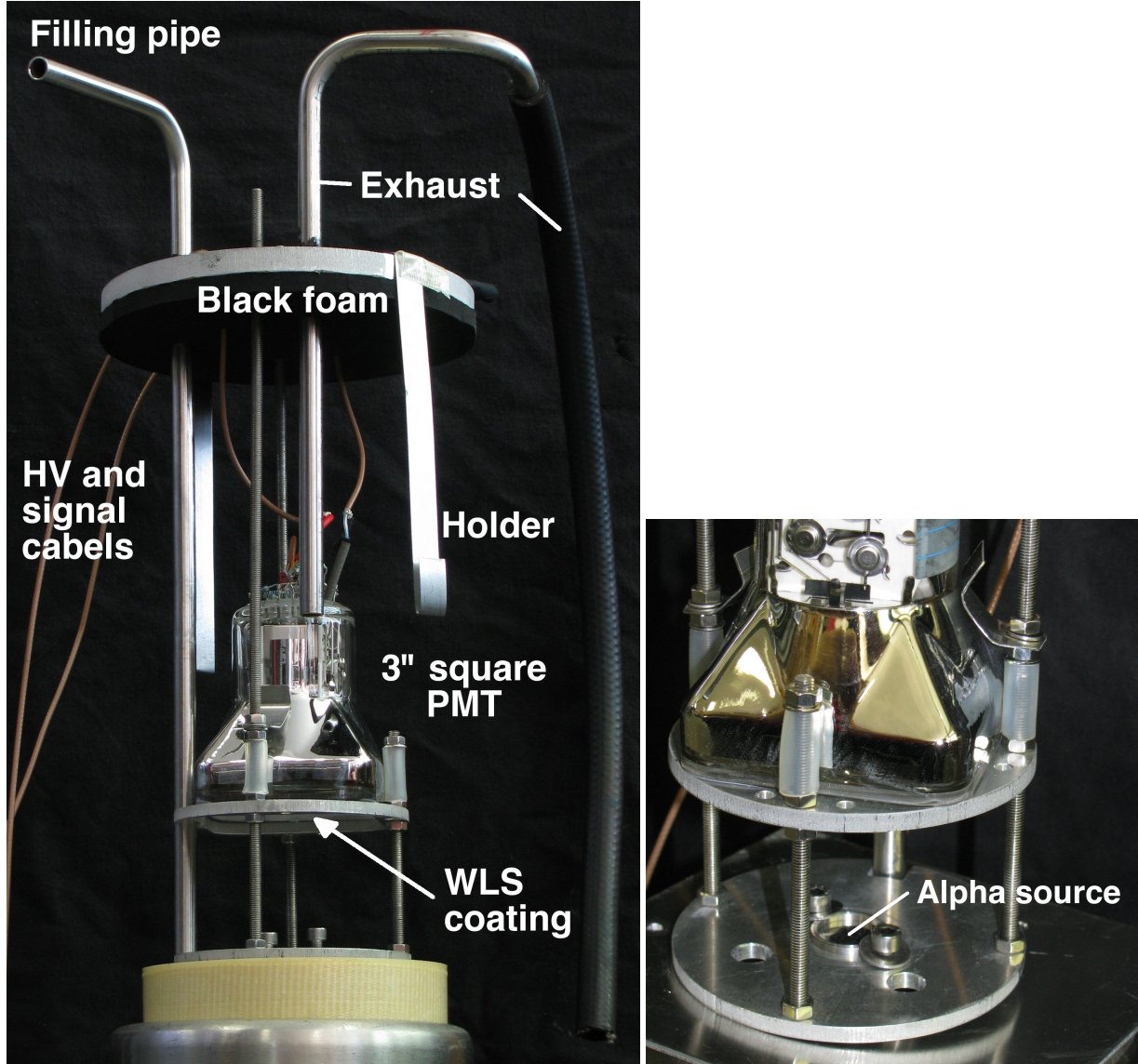


Figure 3.1: Setup to measure the efficiency of different PMT coatings.

3.2 Coatings

Two different coating techniques were studied. Firstly, TPB is directly evaporated on the PMT. Secondly, TPB was dissolved together with a plastic in an organic solvent. The coating was performed by dipping the PMT into the solution. This has been done using Paraloid or Makrolon as a host matrix.

3.2.1 Evaporation

TPB deposited by evaporation is a common coating used in liquid Ar detectors. The evaporation was done by the thin film workshop at CERN. The evaporated layer looks uniform but milky. With a microscope, using visible and UV light, structures and grains are observed (see figure 3.2). The grains are probably small crystals causing the milky appearance of the layer. Measurements were done at a thickness of 0.08 mg/cm^2 and 0.25 mg/cm^2 . According to [36] a thickness of 0.07 mg/cm^2 is favorable and [10] is using a thickness of 0.2 mg/cm^2 .

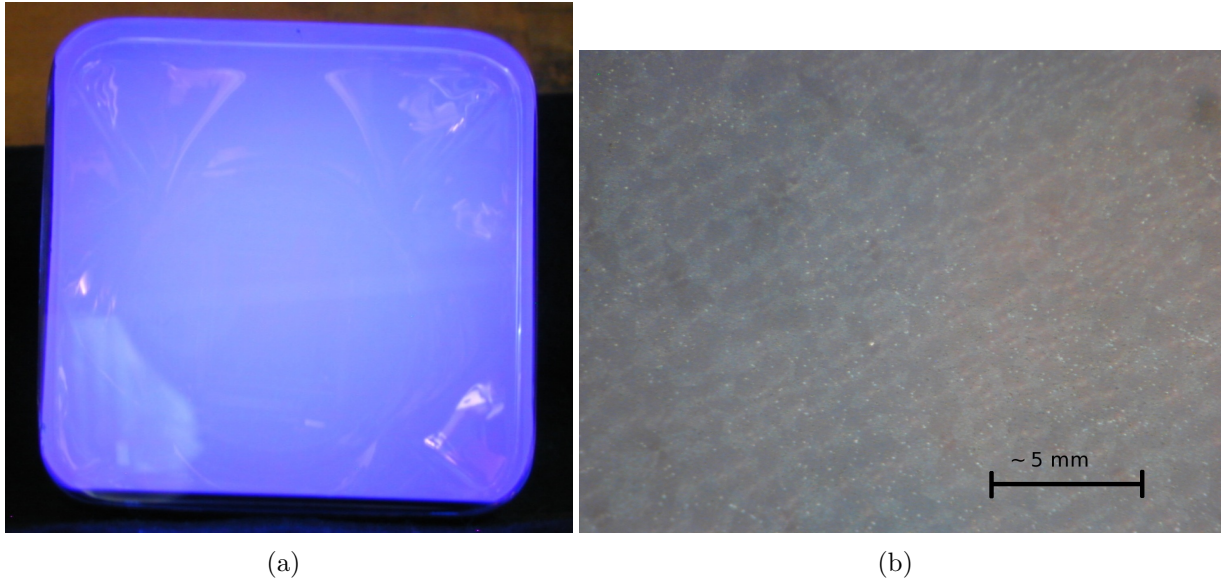


Figure 3.2: The PMT window coated with 0.08 mg TPB/cm^2 deposited by evaporation. (a), View of the whole coated PMT window illuminated by 254 nm light and (b) a close view of the coating illuminated by white light.

3.2.2 TPB in a Paraloid matrix

Dissolving TPB together with a plastic material acting as a host matrix might prevent TPB from forming small crystals. A clear plastic often used to produce thin films is Paraloid. 2.5 g of TPB and 1 g of Paraloid has been dissolved in 300 ml of Chloroform. Tests have been performed on glass slides to get control of the thickness. A good result can be obtained by pulling out the glass plates or the PMT relatively slow. One dipping deposits a layer of $(55 \pm 13) \mu\text{g/cm}^2$. Two layers can be obtained by dipping twice. The second dipping must be done quickly to avoid dissolving the first layer and to achieve uniformity.

The solvent evaporates quickly and leaves a uniform and very clear layer of TPB in Paraloid. A picture of the coated PMT is shown in figure 3.3. Illuminated by 254 nm light the layer looks less bright than the layers deposited by evaporation. One reason could be

that light shifted inside the layer is trapped by reflections on the surface of the coating. This would be a desirable property.

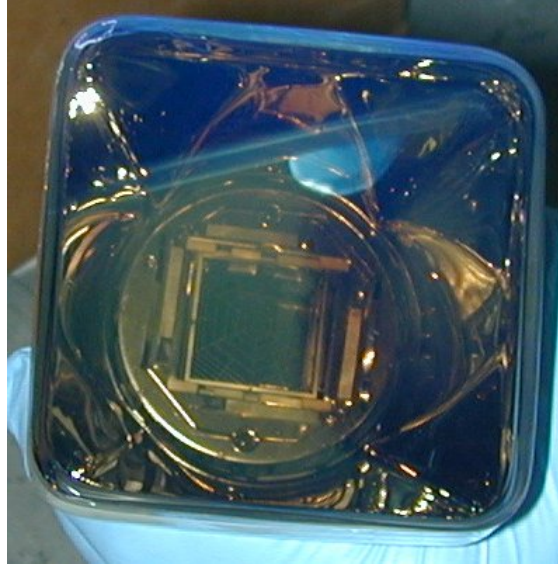


Figure 3.3: View of the PMT coated with one layer of TPB in a Paraloid.

3.2.3 TPB in a Makrolon matrix

Makrolon is a clear plastic from Bayer AG Germany known to be resistant against low temperatures. 1g of Makrolon and 2g of TPB has been dissolved in 300 ml of Dichloromethane. A uniform coating can be obtained if the test glass plates or the PMT are pulled out relatively fast and then held horizontally until dried. Right after pulling it out a tissue is held to the side of the lower corner for about two seconds to soak up a small accumulation of the solution. The resulting thickness of the coating is $(34 \pm 8) \mu\text{g}/\text{cm}^2$ for one dip and $(49 \pm 9) \mu\text{g}/\text{cm}^2$ for two dips. The coated PMT looks very similar to the PMT coated with TPB in a Paraloid matrix.

3.3 Measurement Procedure

The PMT was set under high voltage (1400 V) before filling with liquid Ar. Pulses were analyzed online in a small Matlab program. The values for τ_2 , pulse height and integrated pulse height were calculated. Figure 3.4 shows τ_2 and the integrated pulse height recorded over a full cool down and warmup procedure. Each entry corresponds to an average over 200 events. Before the cell was filled with liquid Ar it was slowly flushed with gaseous Ar (achieved by slightly opening the liquid Ar valve). At position 1 the flow was increased significantly. As a result the lifetime increases immediately. The α source has been covered with liquid Ar at position 2 leading to a sudden decrease of the lifetime. As soon as liquid

Ar starts to come out of the exhaust, the filling was stopped and the trace of each event was recorded (position M). The cell is warming up within several hours. At position 3 the level of the liquid drops below the PMT. The reduction of the amount of collected light is probably caused by reflections on the surface of the liquid. The scintillation process and hence τ_2 is not effected. At position 4 the source was not covered by liquid Ar anymore. At position 5 all the liquid was evaporated, which stops the Ar gas flow. This increases the diffusion of air into the cell.

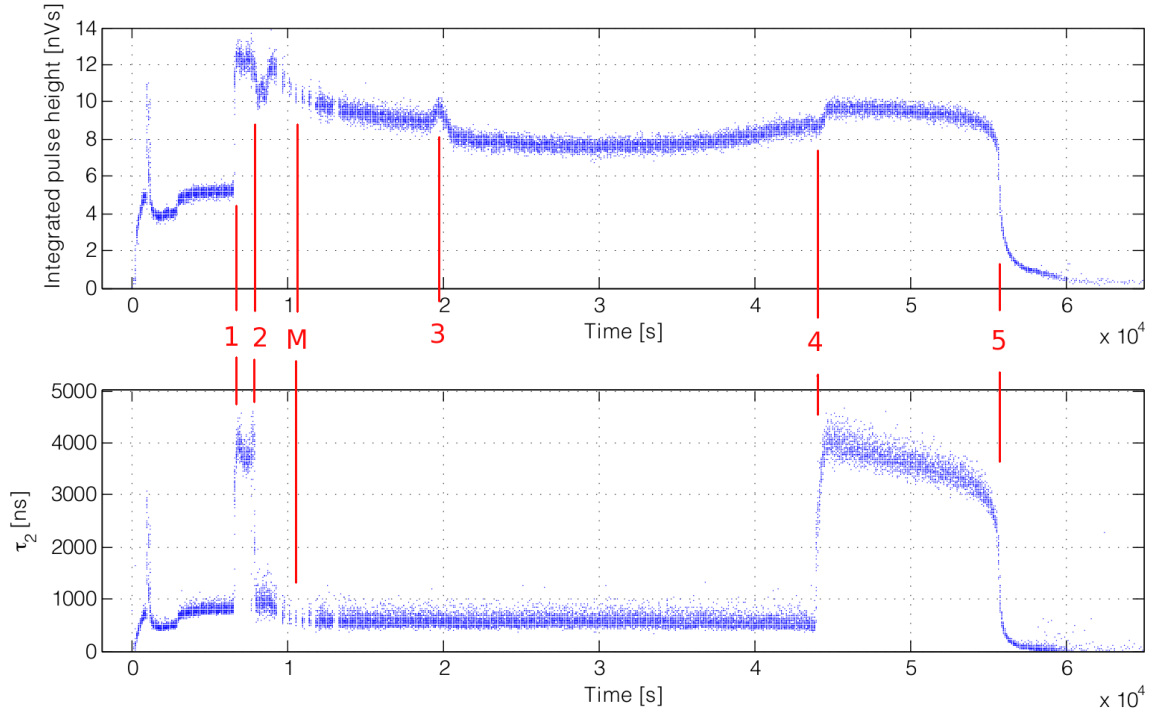


Figure 3.4: Online monitoring of the integrated pulse height and τ_2 during a measurement with a coating of 0.25 mg TPB/cm^2 deposited by evaporation.

The measurements have been taken in two series with about three month in between. A second batch of liquid Ar was necessary for the second series. This was delivered in a different dewar with which it was not possible to flush as slow as with the first one. This reduced the time of the filling procedure to about a half.

For each event 5000 ns are recorded with a sampling frequency of 1 GHz using the oscilloscope described in section 2.5. The readout is triggered if the signal exceeds a threshold of 5 mV, which is above the single photo-electron level.

3.4 Analysis

The efficiencies of the coatings are analyzed using pulse height spectra of α events. The pulse height corresponds to the amount of light in the first component. As described in section 1.2, the first component and especially the pulse height should be less sensitive to impurities than the total integrated pulse height and hence more accurate for an open system. However, absorption is not taken into account.

The pulse height of an event is determined by taking its maximum value. The error originating from this method is negligible for the used PMT with a rise time of ~ 10 ns and a DAQ sampling rate of 1 GHz.

In a first step pile up events produced by the α source (40 kBq) were removed. These can be seen in the two dimensional plot showing the integrated pulse height versus component ratio (CR) (figure 3.5). Events with a correct CR but higher integrated pulse heights are due to pile up events in the first component. In events with a smaller CR, the pile up is counted as light in the second component. It is necessary to remove pile up events for an accurate determination of τ_2 .

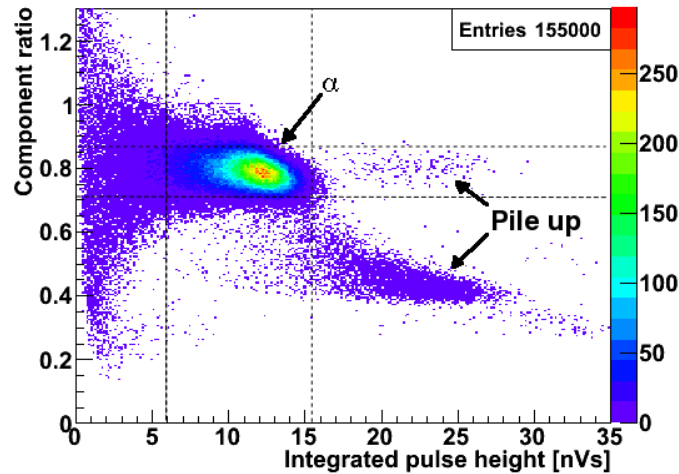


Figure 3.5: Component ratio versus integrated pulse height from the measurement of one layer TPB in Paraloid illustrating the applied cut.

3.4.1 Pile up removal

Most of the pile up events are removed by a cut on the CR and the integrated pulse height. For the measurement shown in figure 3.5 events with a CR in the range of 0.71 to 0.87 and an integrated pulse height between 6 nVs and 15.5 nVs are chosen. Additionally all events with a position of the main peak outside a time interval of 40 ns are removed. However, these cuts are not sufficient to remove small pile up events and a more sophisticated

method has been developed and implemented in the analysis software to determine τ_2 accurately.

The pile up removal method is the following: The trace of an event is divided in intervals of 10 samples ($=10\text{ ns}$) and the average of each interval is calculated. The difference between neighboring intervals is calculated and written into an array. This is done for each event. The idea behind this method is that if there is a pile up, the signal increases and after a short time decreases again. In contradiction to a change of the signal due to noise, the change due to a pile up has a characteristic time scale which is the rise time of the used PMT ($\sim 10\text{ ns}$). This is the reason to average over 10 samples. The method acts as a high and low frequency noise filter while being still sensitive to pile up events. The event is considered to contain a pile up if any difference more than 80 ns after the peak is bigger than 4 mV. An example of a trace containing two small pile up events and the corresponding differentiated trace is shown in figure 3.6.

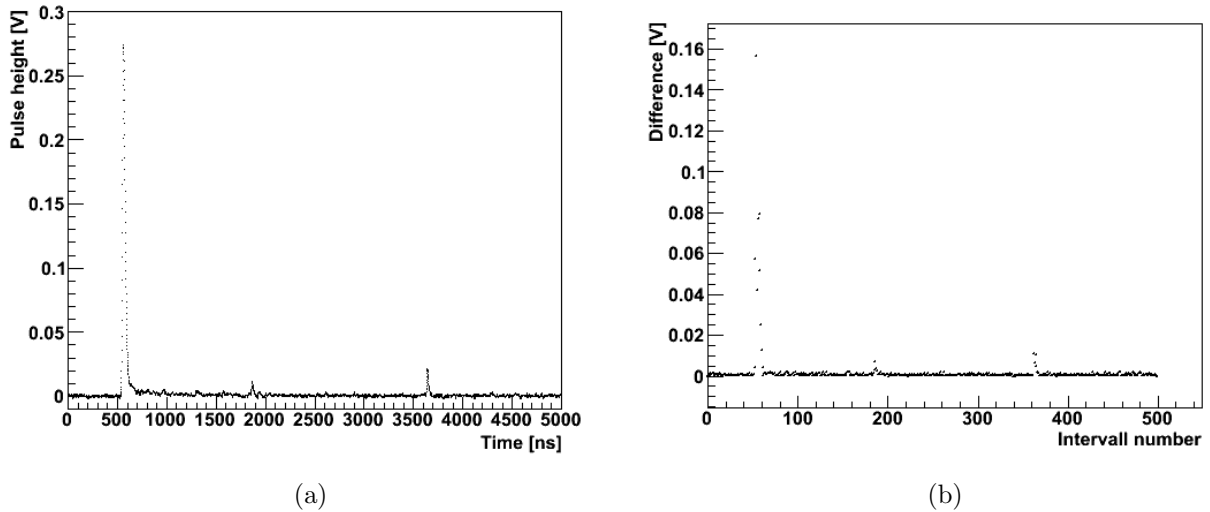


Figure 3.6: Example of a trace (a) containing two small pile up events and the corresponding differentiated trace (b).

3.4.2 Determination of τ_2

In a first step the measurement was divided in blocks of 5000 events. The mean trace of 5000 events is shown in figure 3.7 (a) and (b) without and with cuts (including the pile up removal method) respectively. It is calculated by summing up the traces of single events and dividing the result by the number of events. The time position of the peaks needs to be adjusted offline before calculating the mean trace.

The cuts are then applied and the mean trace is calculated for each block of 5000 events. τ_2 is obtained by fitting an exponential decay to the mean trace in the time range of 700 to 1700 ns after the peak. In contradiction to the expectations there is a correlation

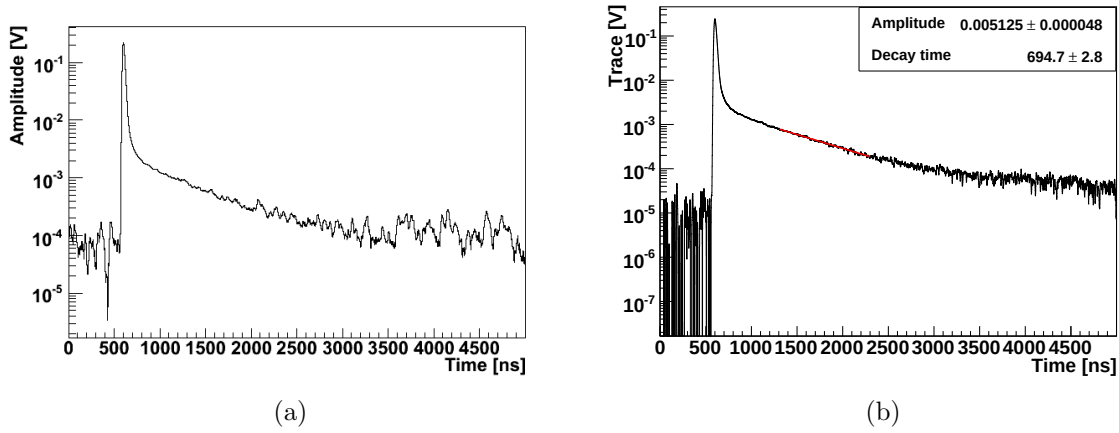


Figure 3.7: Mean trace of the first 5000 events with two layers of TPB in Paraloid. In (a) without cuts, in (b) with the final cuts described in the text and in red the fit of an exponential decay.

between the pulse height and τ_2 (see figure 3.8). This is a hint that impurities absorb light beyond the impurity quenching mechanism described in section 1.2.1. The total error on τ_2 is 36 ns. It has been estimated by determining τ_2 from mean traces obtained for different cuts on the component ratio ± 21 ns, the pile up removal method ± 13 ns and different fit ranges ± 26 ns. The error originating from the cut on the integrated pulse height is neglected. The statistical error of ~ 3.5 ns originating from the fit itself is calculated using an estimation on the error of the mean trace.

3.4.3 Results

To compare events with the same purity, all events from consecutive blocks with τ_2 in the range of 820 ns to 910 ns ((865 ± 45) ns) which are not removed by the previously mentioned cuts are used to produce the final pulse height spectra. An accurate value for comparison is the pulse height on the higher energy slope at half of the height of the spectrum (compare to figure 3.9). The spectrum was plot in the same range and 200 bins for each measurement. For the determination of the height, the spectrum is rebined to 50 bins. The maximum of this spectrum divided by four is taken as the height of the spectrum with 200 bins. One half of this value is equalized with a linear function obtained from a fit to the slope (blue line in figure 3.9). For this spectrum the equation is:

$$\frac{340}{2} = 3244 - 8480 \cdot \text{PH}. \quad (3.1)$$

The error on the pulse height value obtained by this method originating from the error on the height of the spectrum is less than 1 %. The results for all described coatings are presented in table 3.1.

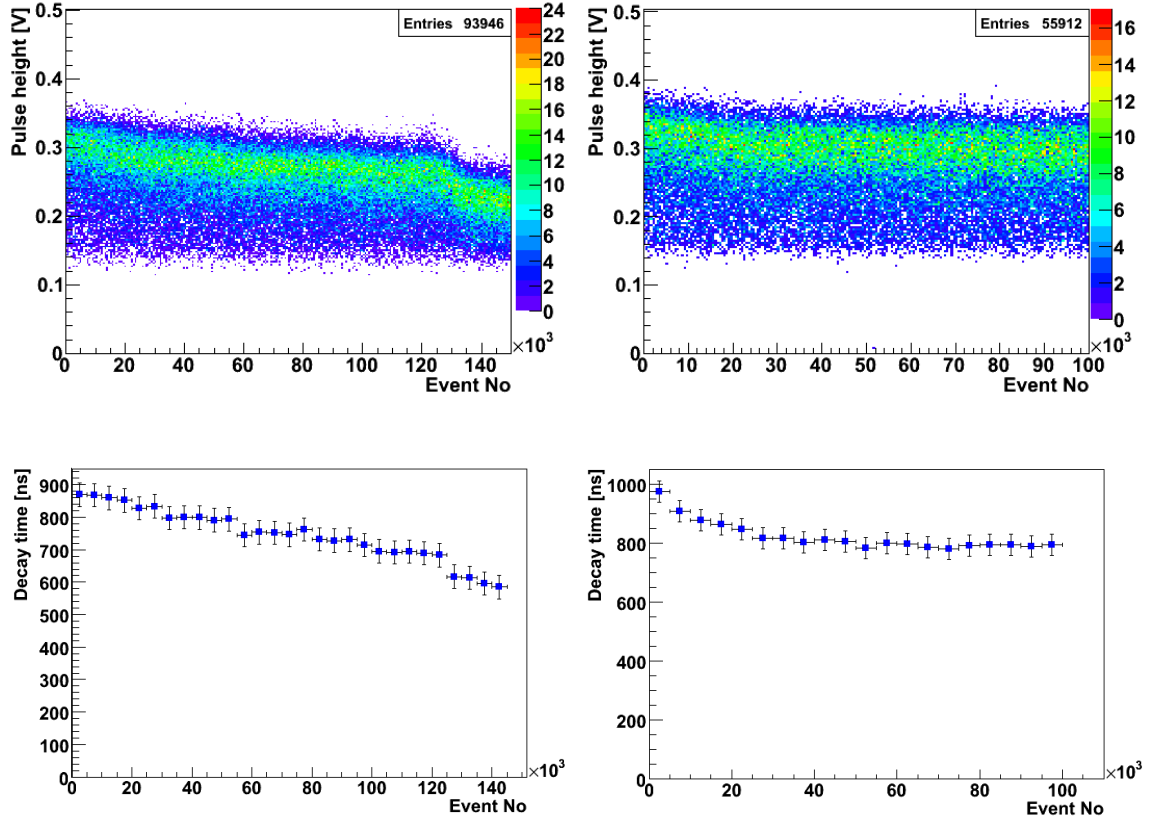


Figure 3.8: Correlation between the pulse height and the decay time of the triplet state for 2 layers of TPB in Paraloid (left) and 2 layers of TPB in Makrolon (right).

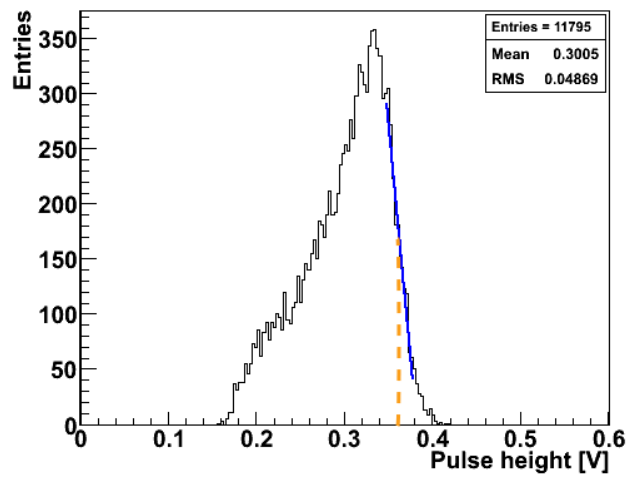


Figure 3.9: Pulse height spectrum (200 bins) for one layer TPB in Makrolon with a linear fit to the high energy slope (blue line). The determined pulse height value is indicated in orange.

TPB Coating	Batch	Range of τ_2 [ns]	No of events	Pulse height [V]
1 layer in Paraloid	1	854 - 880	28 643	0.332 ± 0.015
2 layers in Paraloid	1	828 - 870	18 650	0.327 ± 0.015
0.08 mg/cm ² evap.	1	831 - 899	8 822	0.491 ± 0.030
0.25 mg/cm ² evap.	1	824 - 839	7 733	0.362 ± 0.015
1 layer in Makrolon	2	820 - 856	11 795	0.362 ± 0.015
2 layers in Makrolon	2	847 - 907	11 251	0.344 ± 0.015

Table 3.1: Results of the coating analysis, together with the range of τ_2 of the chosen blocks and the number of events in the final spectra.

The errors given in table 3.1 are estimated from the pulse height versus event number plots. The gain of a PMT might change with a change of its the temperature. The same filling procedure was used for all measurements to keep this effect consistent. The presence of a gain change within the coating measurements can neither be excluded nor confirmed by comparing the trend of the integrated pulse height with the trend of τ_2 from the online monitor.

For the measurement with 0.08 mg TPB/cm² deposited by evaporation, the α source was partly covered by a washer to reduce the number of pile up events. Due to a gap between the source and the washer, the reduction in the number of pile up events was marginal. An undesired effect of this was that the pulse height spectrum was significantly smeared out to lower energies which results in a bigger error. The washer has therefore been removed.

The efficiency of one layer of TPB in Makrolon was measured twice. The result of a quick analysis of the second measurement is (0.37 ± 0.025) V. This is consistent with the result shown in table 3.1. This emphasizes the reproducibility of the measurement and analysis procedure.

3.4.4 Durability of coatings

The coating needs to be robust against cooling to a temperature of ~ 87 K. In addition a certain amount of mechanical stress occurring during the filling of liquid Ar detectors must be resisted. Results from investigations on these aspects are given in this section.

To test the durability of **evaporated TPB**, the layer with 0.25 mg TPB/cm² was used. The coated PMT was left in air but inside the measurement setup for three month. Afterwards, the coating layer looked more milky. For the measurement of the aged coating, the Ar flow during the flushing of the cell was strong compared to previous measurements. The result is an enormous reduction in the efficiency of the coating. The pulse height of the α signal was reduced by $\sim 40\%$ compared to the first measurement. The same measurement was performed again the next day leading to a reduction of $\sim 65\%$ compared to the first measurement. The reason for this enormous difference is that the TPB does

not adhere well to the PMT. This is clearly visible on the PMT window itself and as the whole cell was covered with TPB afterwards (see figure 3.10). The stickiness of the coating is possibly affected by the evaporation procedure and reactions with oxygen or water due to the storage of the coated PMT in air.

TPB in Paraloid has a good mechanical resistance but does not resist the temperature change. After a measurement in liquid Ar the coated PMT looks milky. The origin of this are many small cracks clearly visible using a microscope (figure 3.11). Illuminating the layer with 254 nm light it can be seen that shifted light escapes the layer at these cracks. The milkiness of a coating with one layer compared to two layers is similar. The cracks very likely occur due to a difference of the thermal expansion of Paraloid and glass. The granulate itself forms some cracks at liquid N₂ temperature (77 K, liquid Ar 87 K).

The mechanical resistance of **TPB in Makrolon** is good, but less than TPB in Paraloid. This has been tested by scratching the layer. After the measurement this coating is slightly milky. The origin are structures visible with the microscope (see figure 3.12). These were not present before the measurement. The density of these structures is smaller than the density of cracks in the coating using Paraloid. A thicker layer shows the same behavior. These structures can be removed by wiping over the coating. Illuminated with a UV lamp, the wiped areas are less bright than the areas with the structures. These structures might originate from TPB residue in the setup.

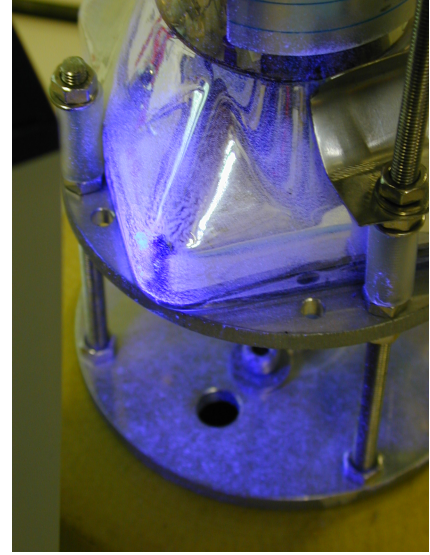


Figure 3.10: The setup illuminated by UV light after the measurements of the aged TPB coating. It is covered by TPB which had been on the PMT window before.

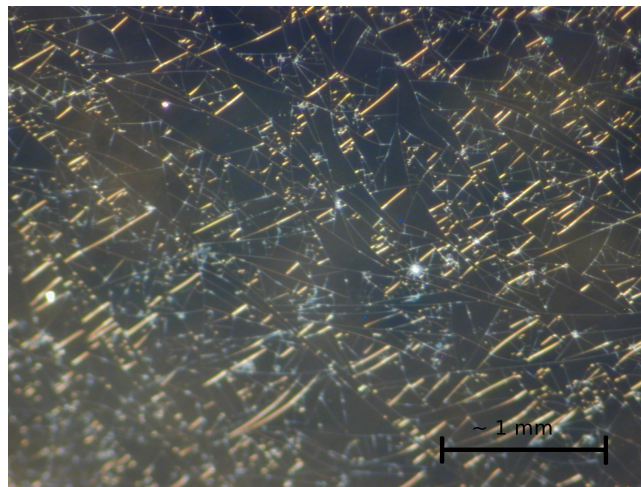


Figure 3.11: Cracked Paraloid TPB coating after a measurement in liquid Ar.

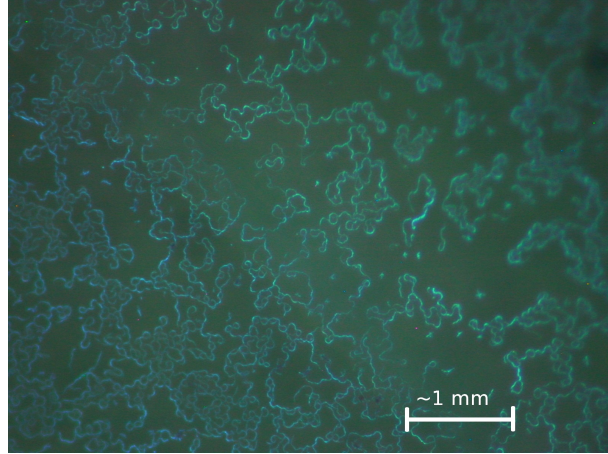


Figure 3.12: TPB in a Makrolon matrix after two cool downs, illuminated by 254 nm light.

3.5 Conclusions

In this chapter the results of different TPB coatings on a PMT window were described. Their efficiency was measured in gaseous and liquid Ar with α particles.

The liquid Ar cell for the scattering experiment contains side reflectors coated with TPB, unlike the coating measurement cell. This implies that the transparency to visible light must be considered in addition to the efficiency for direct VUV light.

Among coatings with a host matrix TPB in Makrolon yielded better results than TPB in Paraloid. The thickness of coatings using a plastic matrix seems to have a small effect on the efficiency. However the best results were achieved by directly evaporating TPB on the PMT windows in thin layers ($\sim 0.08 \text{ mg TPB/cm}^2$).

An important side result is the observation of the change of the pulse height as a function of τ_2 in liquid Ar. Investigations on this should be performed to extrapolate the light yield correctly.

Chapter 4

The improved scattering setup

The former liquid Ar cell has been modified for a better light yield and material surrounding the active volume was removed to reduce the fraction of multiple scattered neutrons. Additionally the new setup allows continuous running without permanent monitoring. To obtain better statistics in the scattering experiment, four angles can be measured at the same time using four 3" LScs (see figure 4.1). Recorded is the signal of the PMTs and the information which LSc has triggered. For this purpose a new DAQ system has been set up.

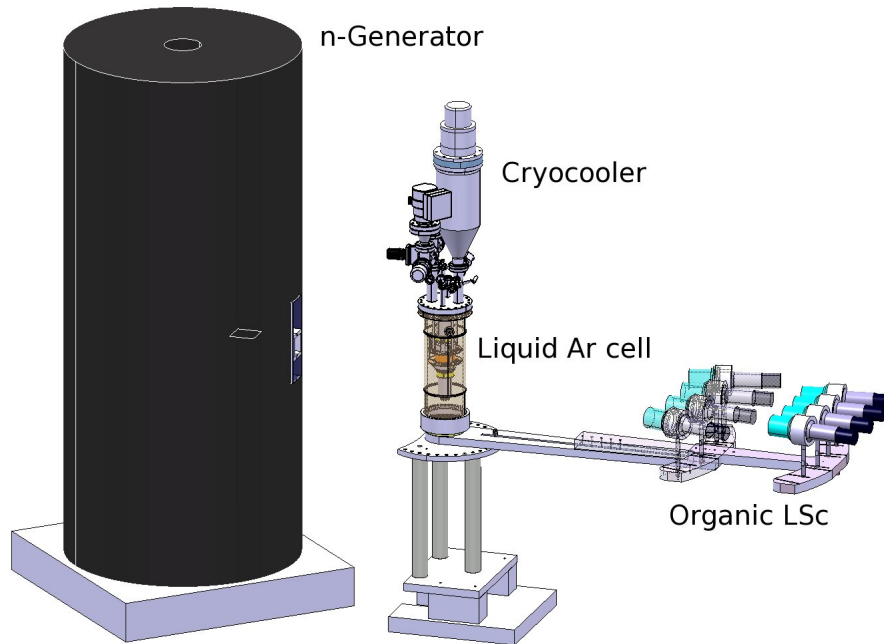


Figure 4.1: Setup for the scattering experiment measuring four angles at the same time. The liquid scintillators can be put at two different distances. The cryocooler will be installed in the future and will replace the Ar handling system described in section 4.1.1.

4.1 The new liquid Ar cell

The external dewar is not needed with the new cell and the inner vessel has been replaced by a stainless steel dewar with an inner diameter of 133 mm. This reduces the amount of liquid Ar surrounding the active volume to 30 mm ($\sim 1/3$ of the former cell) and the total amount of surrounding stainless steel to 1.5 mm. The height of the active volume is reduced to 49 mm. This improves the light collection and the resolution of the scattering angle.

Based on the results of [37] the Tyvek reflector is replaced by a Tetratex foil from the company Donaldson Membranes covered with 1 mg TPB/cm² deposited by evaporation. Corresponding to the results of the PMT coating tests (see chapter 3) the TPB Paraloid coating has been replaced by 0.08 mg TPB/cm² deposited by evaporation.

Additionally a ⁸³Rb source producing ⁸³Kr can be connected to the setup for a calibration at 32 and 9 keV.

4.1.1 The Ar handling system

A schematic of the Argon handling system is shown in figure 4.2. The liquid Ar is filled-in from a 200 l dewar. This is equipped with a rupture disc, an adjustable overpressure valve and two pipes, each having a manual valve. One pipe is connecting the liquid phase of the dewar and the liquid Ar cell. The other connects the two gas phases. The Ar cell has two overpressure valves, one is adjustable, the other opens at ~ 1.5 bar overpressure and is used as a safety valve.

Liquid Ar is filled into the cell by the following procedure: The cell is filled with Ar gas. The overpressure valve of the cell is adjusted to a slightly lower pressure than the pressure inside the 200 l dewar. The connection of the two gas phases is closed and the connection of the two liquid phases is opened. The filling speed can be controlled by the adjustable overpressure valve on the cell. This system allows the liquid to be filled back into the dewar in a similar way.

4.1.2 Test of the insulation properties of the cell dewar

To measure the heat leak rate into the dewar it was filled $\sim 2/3$ with liquid N₂. The top was covered by a cleaned stainless steel plate to reduce the heat transferred by radiation and convection. To measure the level a plastic ruler has been put into the dewar. The dewar has an inner diameter of 133 mm.

To ensure stable conditions, the measurement was started two hours after filling. The level of the liquid N₂ was 21.1 cm at this time. In the first three hours the level was checked every hour and decreased (1.3 ± 0.1) cm per hour. For the next 14.5 h the cover was not removed and the level decreased to 3.2 cm which corresponds to 0.97 cm/h. The

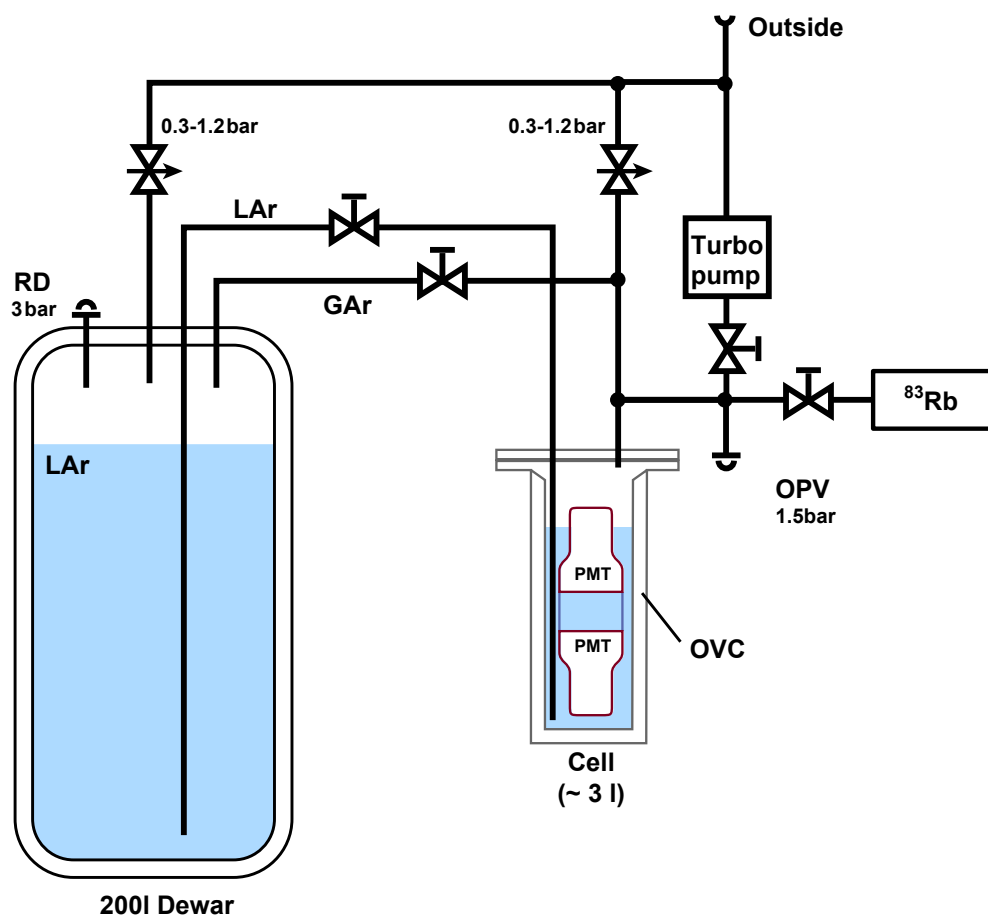


Figure 4.2: Scheme of the argon handling system.

enthalpy of vaporization of liquid N_2 is 198,6 J/g and the density at 77 K is 0.807 g/cm³. Using these values we obtain a heat transfer of 8.1 W in the first tree hours and 6.0 W in the last 14.5 hours. The insulation is sufficient for our needs.

4.2 High resolution DAQ

Using four LSCs at different angles at the same time requires recording at least one signal in addition to the PMT signals. This is not possible with the previously used Oscilloscope. For this reason we use an 8 channel 12 bit ADC module (V1720) from CAEN. The higher resolution allows measurement of single photo-electron events without saturation of the 5.3 MeV α particles from the internal source and without splitting the signals. The sampling frequency is 250 MHz. The V1720 is connected to a PC by an optical link. The data is recorded using a software called “wavedump”. The wavedump software reads a configuration file containing the settings of the V1720 an writes them into the memory of the module.

The basic version of the wavedump software provided by CAEN was modified to the specific needs of our experiment. Additionally an online display of the traces was setup using Matlab.

4.2.1 Measurement of the scattering angle

The four LSCs are connected to the four channels of the MPD4 module (see as well section 2.5.1). If an event is identified as a neutron, the corresponding channel will generate a digital trigger pulse. Four trigger outputs are connected to a passive analog adder with four signal attenuators to deliver one signal recorded by the V1720. The circuit diagram is shown in figure 4.3. The triggered channel is determined by the level of its output voltage.

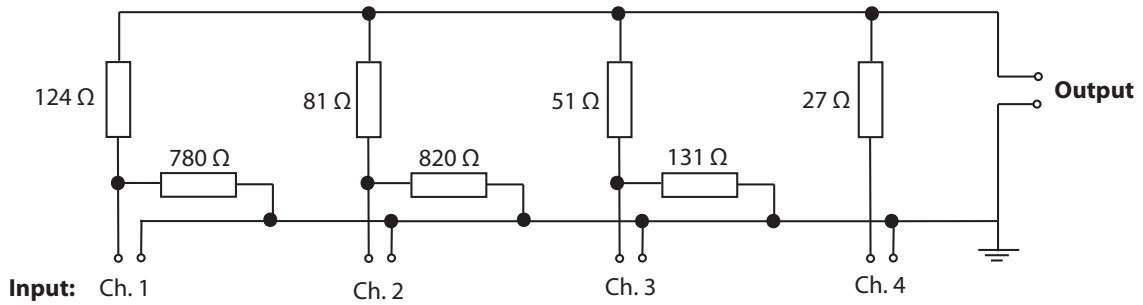


Figure 4.3: Analog adder with four signal attenuators.

4.3 Measurement of the improved light yield and results

The PMT gains were determined from the MPVs of single photo-electron spectra from event tails using the scaling factor derived in section 2.7.1. The light yield spectrum from a measurement with 60 keV γ rays from a ^{241}Am source is shown in figure 4.4. The energy resolution and the signal to background ratio has significantly improved compared to the measurement presented in section 2.7.3. The MPV of the Gauss fit to the photo peak is (60.9 ± 1.3) pe at a τ_2 of (308 ± 70) ns. The error on the MPV is originating from the PMT gain calibration. Extrapolated to 1600 ns using equation 1.7 on page 9 this corresponds to (182 ± 34) pe with¹ $A = 29.57$ pe and $B = 26.48$ pe obtained like described in section 2.7.3. The resulting light yield is (3.03 ± 0.57) pe/keV. The purity of the delivered Ar was very low, which results in a large error on the extrapolation. The measurements in chapter 3 indicate that the light in the first component is also affected by impurities. This would result in a light yield higher than 3.03 pe/keV for a τ_2 of 1600 ns.

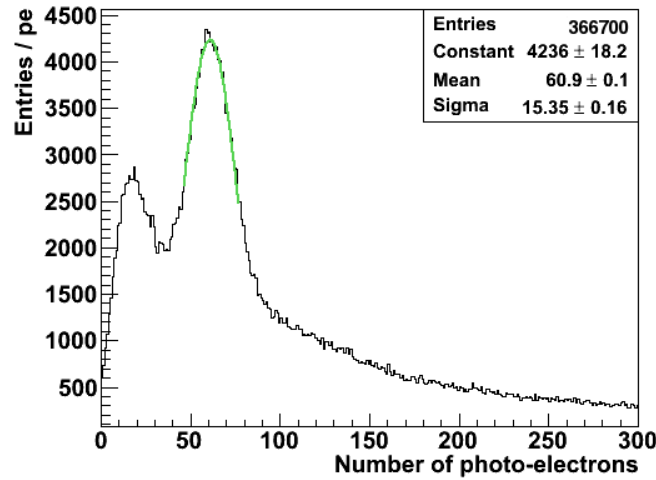


Figure 4.4: New spectrum of the 60 keV γ line from ^{241}Am demonstrating the improved energy resolution. A Gaussian fit is superimposed to the data.

Assuming a perfect energy resolution of the detector, the background-free spectrum from photo-effect γ rays is a poisson distribution. The deviation of the measured spectrum from the poisson distribution is a measure for the energy resolution of the setup. The σ of the poisson distribution is the square root of the expectation value. For a high number of photo-electrons a poisson distribution can be approximated by a gauss distribution with a σ equal to the square root of its mean value. To estimate the energy resolution of the improved liquid Ar cell, the spectrum of the 60 keV γ line is fit with a gaussian

¹Courtesy of William Creus

and the square root of the mean value of this fit is compared with the σ obtained from this fit. The σ is a factor of 1.97 bigger. This means that the energy resolution of the detector is worse than the intrinsic resolution due to Poisson statistics. This is probably predominantly caused by geometrical effects on the light collection.

4.4 Conclusions

The presented results show a significant improvement in light yield and light collection (energy resolution) compared to the previous setup. The purity of the Ar was low but will become better as soon as the cryocooler together with a recirculation and cleaning system will be installed. This is expected to give an electronic light yield better than 3 pe/keV. The setup is almost ready to take a measurement of the light yield of nuclear recoils for several angles with a precision competitive to the measurement in [10] or better.

Chapter 5

Final conclusions and outlook

In this work developments for measurements of the relative scintillation efficiency $\mathcal{L}_{eff}$ for nuclear recoils in liquid Ar were performed. They resulted in a largely improved setup ready to take data with neutrons from a monochromatic neutron generator. A first measurement was done in the range of 55 keVr to 81 keVr and yielded a result for $\mathcal{L}_{eff} = 0.282 \pm 0.046$. In particular the following results were found in the framework of this thesis.

The PMT gain calibration procedure was reworked. Now it is performed using the most probable values of single photo-electron spectra from event tails, which does not require calibration measurements anymore.

Investigations on wavelength shifter coatings showed the highest efficiency for a layer of 0.08 mg TPB/cm² deposited by evaporation. However, the adherence of this type of coating seems to be insufficient. In this aspect the best performance was found for a coating of TPB in a Makrolon matrix, with $(73.7 \pm 5.4)\%$ of the efficiency of the best evaporated layer.

The electronic light yield was improved by at least 25 %, primarily by changing the wavelength shifter coatings. The light yield of the improved setup was measured at a poor Ar purity. However, the purity was found to have an effect on the first component of the signal. The light yield measurements are corrected only for a reduction of the lifetime of the second component.

A precise measurement of the light yield of nuclear recoils in liquid argon is expected within the next few months, after the installation of a continuously running cryogenic cooling and cleaning system.

In a next stage 2" PMTs with a quantum efficiency of $\sim 30\%$ (compared to $\sim 15\%$ in the present setup) will be installed, which will significantly improve the low energy threshold of the setup. It will also include a charge readout system to measure the charge yield in addition to the light yield.

Chapter 6

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