Optical And Acoustical Studies
Of The Proposed PICO-250L
Dark Matter Detector

by

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Abstract

In the first part of this work I explore the possibility of using linear alkylbenzene, the buffer liquid for the future PICO-250L detector, configured as a liquid scintillator that could provide information on the background radioactivity levels of the inner vessel. I investigate what interactions could produce scintillation light and determine their intensity and likelihood. I design different devices that help light collection and assess quantitatively their advantages and disadvantages.

In the second part of this work, I develop a 2D acoustical model of the inner vessel of the detector that reproduces the discrimination between alpha and neutron events characteristic of backgrounds in superheated liquid experiments. Such a model is crucial for understanding the propagation and reception of the acoustic signals in the chamber. Subsequently, I exploit this model to show that piezoelectric devices located in different part of the vessel yield different discrimination capability between single and multiple bubble events.
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To possess a wide and deep knowledge, is surely admirable. To be willing and able to share it in a clear and brilliant way, shows great and mindful generosity. To guide with a patience that seems to be nourished by the difficulties of a student, is a gift that encourages creativity and strengthens self-confidence.

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Chapter 1

Introduction

The PICO collaboration is currently designing a large scale detector, based on the superheated liquid technology, that will contribute to the field of direct searches for dark matter.

The first part of this work focuses on exploring a latent potentiality of the detector, such as the use of the buffer liquid, linear alkylbenzene (LAB), as scintillator to study the background radioactivity inside the inner vessel.

I first investigate the compatibility of the scintillation property of LAB with the active fluids used in the experiment, while then considering the intensities of background radiations that can be detected using the liquid scintillator, and finally designing and simulating components that enhance the efficiency of this method for background assessment purposes.

Acoustical signals constitute a crucial part of superheated liquid experiments. Understanding how these signals are generated and then propagate in the detector provides fundamental knowledge in guiding the design of the experiment.

In the second part of this work I deal with the development of an acoustic model
of the PICO detector. I aim at building a reliable model that replicates the difference between alpha and neutron signals that is distinctively observed in superheated liquid experiments, while reproducing the physical mechanism believed to cause this difference.

Lastly, I use this model to determine which positions of the piezoelectric devices around the vessel can produce the best results for discriminating between alpha-like and neutron-like (i.e. dark-matter-like) events.

In Chapter 2 I present the evidence for dark matter and introduce the fields of direct and indirect searches with an emphasis on bubble chamber experiments. In Chapter 3 I discuss the studies on LAB as a scintillator in the PICO-250L detector, while in Chapter 4 I describe the development of a 2D acoustical model for the detector. In Chapter 5 I summarize and comment the results of this work.
Chapter 2

Dark matter

2.1 A short introduction to dark matter

According to the latest data provided by the Planck experiment [1], a space observatory operated by the European Space Agency which was dedicated to studying the early Universe and its evolution, all the particles that have been detected and studied so far represent only 4.9% of the total amount of matter and energy in the universe.

To understand the current lack of understanding of the rest of matter and energy in the Universe, we could start by realizing that if gravity is the only force that can act at the large scale of intergalactic distances, we would expect matter to clump and collapse together.

However, it has been shown by Hubble in 1929 [2] that the Universe is actually expanding, and galaxies are moving away from the Earth at a speed that is directly proportional to their distance (Hubble’s law: \( v = H_0 D \), where \( H_0 \) is the Hubble constant), as dots on the surface of a balloon that is being inflated. Even more surprisingly, recently it has been discovered that the Universe is not simply expanding at a linear rate, but the expansion rate is increasing with time. For "the discovery
of the accelerating expansion of the Universe through observations of distant supernovae\textsuperscript{a} the Nobel Prize in Physics 2011 has been awarded to Saul Perlmutter, Brian P. Schmidt and Adam G. Riess \textsuperscript{3}. To justify this enormous force that counterbalances gravity, a new type of energy has to be introduced, and since at the moment it has not been understood or detected, it has been called \textit{dark energy}. This dark energy contributes 68.3\% of the energy/matter budget in the Universe.

The last slice in the pie, 26.8\%, is attributed to a new type of matter that theoretically could solve several astronomical anomalies which escape our current understanding of the universe. Again, this type of matter has not been understood or detected yet, therefore the adjective "dark" well describes it, i.e. we call it \textit{dark matter}. In order to believe in something that we can not see or detect in any direct way, much evidence is needed. This is summarized in the next section.

2.1.1 Evidence for dark matter

\textbf{Velocity dispersions of galaxies and rotation curves anomalies}

The first hint for a new type of matter came with the observation by Fritz Zwicky in 1933 \textsuperscript{4} that the luminous mass of the Coma cluster was too small if compared with the value inferred through his measurements of the velocity dispersions of eight galaxies rotating around the center of the cluster. Zwicky understood that a non-luminous type of matter was present in the cluster, and he called it dark matter.

Clearly, another possible explanation for this anomaly could be an incomplete understanding of the laws of gravity, especially at the large scales that are encountered in intergalactic distances. This approach was the one followed by the father of the
Modified Newtonian Dynamics (MoND) theory, Mordehai Milgrom, in 1983 [5]. Unfortunately, it fails to reproduce important known properties of the Universe or leads to unobserved features, and therefore has limited support in the scientific community.

More compelling evidence for the introduction of dark matter came from the astronomer Vera Rubin in 1970's [6], who studied the Doppler shift of stars (more specifically, clouds of ionized hydrogen) rotating in spiral galaxies. Rubin realized that if a star with mass $m$ were bound via a gravitational interaction $F_g$ with the matter $M$ in the galaxy in a radius $r$, equating its equation to the corresponding centripetal force $F_c$, as in Eq. 2.1, can provide an expected curve (Eq. 2.2) for the velocity $v$ of the stars as a function of their distance from the center of the galaxy.

$$F_g = G \frac{mM(r)}{r^2} = F_c$$

$$v = \sqrt{G \frac{M(r)}{r}}$$

Rubin analyzed the Doppler shift of the light coming from the stars she selected, and found out that even though the mass of the galaxy was concentrated in the center, which yields $M(r) \approx$ const for the distances of these stars, the behavior of the velocity curve was not following the simple $\sqrt{1/r}$ dependence as expected, but was actually constant with increasing $r$, as shown in Fig. 2.1.

**Gravitational lensing**

As explained in the previous sections, despite having existed since the birth of our Universe, dark matter was unobserved and unsearched for until the end of the 20th century. Therefore, it not surprising that the second chronological piece of evidence
for this new type of particles comes from an effect due only to the gravitational force. From Einstein’s General Relativity (GR) we know that space-time is distorted by the presence of mass, and anything traveling in it will be affected by this gravitational distortion.

Not even light rays escape from this process, so that the light coming from, for instance, a galaxy, is bent by the gravitational field of masses and generates interesting optical effects. This effect, predicted by GR, is known as *strong gravitation lensing*, and turns out to be very useful to indirectly detect and locate distributions of forms of matter that emit a little amount of light or no light at all. It then perfectly applies to the case of dark matter.

In Fig. 2.2 we can see one of the most famous examples of strong gravitational lensing, the horseshoe Einstein ring named LRG 3-757 [7].

Unfortunately it is quite rare to find an alignment of a luminous object, such as a background galaxy, and a concentration of mass in the foreground that yields optical
2.1. A SHORT INTRODUCTION TO DARK MATTER

Figure 2.2: The Einstein ring LRG 3-757, as seen from the Hubble Telescope.

distortions such as arcs and multiple images, or Einstein rings.

Nevertheless, gravitational lensing also occurs in a lighter fashion, when light coming from far galaxies is simply sheared by large-scale structures. This second type of gravitational distortion is called \textit{weak gravitational lensing}, and when used to interpret a large enough number of galaxies to separate the lensing effect from the noise represented by their random orientation, it allows one to create a map of the total matter in the observed region of the Universe. The first detection of weak lensing by large structure occurred in the year 2000 [8].

A comparison of the mapped distribution of total matter to the distribution of baryonic matter given by optical and X-rays data reveals where dark matter is likely hiding. This has been done for the Bullet Cluster [9], (where two clusters of galaxies are colliding) using data provided from the NASA’s Chandra X-ray Observatory and other telescopes.

Superimposing the dark matter distribution (in blue), obtained by studying gravitational lensing effects, on the baryonic matter one (in pink), obtained by studying
the X-ray emission of the cluster, yields one of the most spectacular pieces of evidence for dark matter (Fig. 2.3). In this collision, the dark matter has passed through without interaction while the dust, comprising the majority of the baryonic material, remains in the centre. The bulk of the matter, as seen from gravitational lensing, is outside the centre of mass of the luminous material.

Figure 2.3: Composite image where dark matter and baryonic matter distributions have been superimposed on the optical image for the Bullet Cluster.

Cosmic Background Radiation

When in 1964 the two American radio astronomers Arno Penzias and Robert W. Wilson unexpectedly found that their 6-meter horn-reflector antenna was receiving a uniform signal in the microwave frequencies coming isotropically from the sky \[10\], their first thought went to some problem in their experimental apparatus, as “white


2.1. A SHORT INTRODUCTION TO DARK MATTER

dielectric substances" i.e. pigeon droppings. The reason why they received the Nobel Prize in Physics 1978 for this discovery is that an isotropic radiation had been predicted by Ralph Alpher and Robert Herman in 1948 [11], as a consequence of the Big Bang model described by their colleague George Gamow.

In fact, if our Universe was born in an extremely hot and dense state, a plasma would exist where the interactions between electrons, protons and photons could not allow light to travel freely in space (because the photons' energy was always being used to break apart newly formed hydrogen atoms) [12].

After approximately 300,000 years, when the Universe cooled down to a temperature that permitted the formation of hydrogen atoms, these photons were finally free to travel in space, and this is what Penzias and Wilson initially, and a long list of experiments subsequently, studied in precise detail, most recently by the ESA’s mission Planck, as in Fig. 2.4. These observations of the very young Universe have to be predicted by any model that proposes to explain how our Universe was born and evolved, and carry therefore fundamental information for the field of cosmology.

Given that this relic radiation reassembles with great precision the spectrum emitted by a black body at a temperature of 2.73 Kelvin [13], with a radiance that peaks in the microwave frequencies, it has been called the Cosmic microwave background (CMB). The CMB is very uniform and isotropic, with tiny fluctuations (visually enhanced in Fig. 2.4) of ten parts in one million, reflecting regions with different densities which are believed to be the seeds for the formation of galaxies.

One of the most important results that we can get from the CMB is the angular power spectrum (Fig. 2.5) [1], a measure of how much temperature fluctuates as a function of the angular scale. Precise measurements of the peaks in this graph, allows
2.1. A SHORT INTRODUCTION TO DARK MATTER

one to determine the geometry of the universe, the baryon density $\Omega_b$, and the total density of non-relativistic matter relative to critical, $\Omega_m$, and as a consequence the dark matter density fraction, $\Omega_{DM}$.

![Figure 2.4: The anisotropies of the CMB as observed by Planck.][14]

![Figure 2.5: Temperature fluctuations in the CMB detected by Planck at different angular scales on the sky.][1]

2.1.2 WIMPs

Extensions of the Standard Model

What has been discussed so far concerns mainly cosmology and astronomy, but since we know that 26.8% of the total matter and energy in the universe is a brand new type of matter, certainly particle physics plays a fundamental role in the field of dark matter.

The Standard Model (SM) of particle physics is the most reliable and up to date model that describes theoretically all the known particles, leading to good agreement with experiments. However, there are several unresolved puzzles in the SM, one of them being that it is really hard to explain why the mass of the Higgs boson \( m_h \approx 125 \text{ GeV} \) is so small if compared to the Planck mass \( M_P = \sqrt{\hbar c/G} \sim 1.2 \times 10^{19} \text{ GeV} \) as it would be expected \([15]\).

In fact, taking into account quantum corrections \( \Delta m_h^2 \) coming from loop-level diagrams the Higgs mass can be written as:

\[
m_h^2 = m_{h0}^2 + \Delta m_h^2 \tag{2.3}
\]

where:

\[
\Delta m_h^2 \sim \frac{\lambda^2}{16\pi^2} \int^{\Lambda} \frac{d^4p}{p^2} \sim \frac{\lambda^2}{16\pi^2} \Lambda^2 \tag{2.4}
\]

\( \Lambda \) being the energy scale at which the SM stops being a valid description of nature, so that’s where the integral stops.

As can be seen \( \Delta m_h^2 \propto \Lambda^2 \) and since in the SM \( \Lambda \sim M_P \), it is very hard for the two terms on the right-hand side of Eq.\((2.3)\) to cancel each other out to get the known value of \( m_h \). One possible explanation for this gauge hierarchy problem would be provided
by having $\Lambda \lesssim 1$ TeV which means new physics at the weak scale $m_{\text{weak}} < 10 \text{GeV-TeV}$ scale \[15\].

Primarily for this reason (and for many others) a new model has been proposed in the 70’s \[16\], called *Supersymmetry*, which postulates superpartners for each known particle of the Standard Model, with the same quantum numbers but different in spin by a half, i.e. bosons are turned into fermions and vice versa.

**Supersymmetry for dark matter**

The lightest supersymmetric particle (LSP) in the supersymmetry, which is a neutral and stable particle in models with R-parity conservation, is accidentally also the best candidate among the *weakly interacting massive particles* (WIMPs) that could make up for dark matter, the neutralino \[15\].

One important property for WIMPs is that they were created thermally during the Big Bang: in the thermal scenario we assume that matter and light were in thermal equilibrium in the earliest stages of the Universe, as simple energy equipartition requires.

However, as the Universe expanded and cooled down, WIMPs could not be created anymore but only self-annihilate, decreasing their density. At some point this density became low enough for these particles to be sufficiently dispersed in the Universe that self-annihilation also became very rare. This means that the WIMP density reached a constant value, i.e. the *freeze-out* equilibrium.

As we can see, the self-annihilation cross-section $\sigma v$, which means the likelihood for interactions, and density number, or better the relic abundance $\Omega_x$, are related to
2.2. DARK MATTER SEARCH

Each other, and with some assumptions this relation can be written as [17]:

\[ \Omega_\chi \approx 3 \times 10^{-27} \text{ cm}^3 \text{ s}^{-1} \]

and substituting for the measured value \( \Omega_\chi \approx 0.1 \) we obtain \( \sigma \approx 3 \times 10^{-26} \text{ cm}^3 \text{ s}^{-1} \), which means a strength of interaction at the electroweak scale, just as the property of supersymmetric particles.

Other candidates for dark matter, which do not benefit from the support of a strong theory such as supersymmetry and are thus considered less important, are: axion particles [18], hypothetical particles created as a potential explanation for why the strong force has not been found to violate charge-parity symmetry, even though the SM predicts so; sterile neutrinos [19], hypothetical right-handed neutrinos that interact only through the force of gravity and if existing could explain several puzzles in the SM, such as flavour violation in neutrino experiments.

2.2 Dark Matter Search

2.2.1 MACHOs

Before describing the different techniques used to search for WIMPs, it is important to mention that the possibility that dark matter consists of large objects made of baryonic matter, which do not emit light and therefore escape detection, called MACHOs (Massive Compact Halo Object), has been investigated with negative results and thus ruled out as an explanation for the dark matter problem.

The most effective way to detect these object is through gravitational microlensing effects, as explained in the previous sections. If the alignment between the Earth and
a MACHO (i.e. the lens) and a source star is perfect, the star would appear as an Einstein ring with radius:

\[ R_E = \left[ 4GmLx \frac{(1-x)}{c^2} \right]^{1/2} \] (2.6)

where \( G \) is Newton’s constant, \( m \) is the mass of the lens, \( L \) is the distance to the source star, and \( x \) is the distance to the lens divided by the distance to the source \([20]\). However, when the alignment is not perfect, as it more likely happens, we can define an impact parameter \( b \), and there will be two images of the source, too close to be resolved. The light from these two images will add, giving a total magnification of the source star by a factor:

\[ A = (u^2 + 2) \cdot u^{-1} \cdot (u^2 + 4)^{-1/2} \] (2.7)

where \( u = b/R_E \) \([20]\). Searches of MACHOs therefore involve observation of stars in nearby galaxies, such as the Large Magellanic Cloud hoping to see if any of them become magnified when a MACHO passes in front. Theoretical calculations provided a probability for this type of event, which for a full MACHO halo occurs for about one star every two million, while the duration of the event (magnification factor \( A > 1.34 \)) will be \( t = 130(m/M_\odot)^{1/2} \) days, where \( m/M_\odot \) is the mass of the MACHO in units of solar masses.

Given that it is possible to monitor stars on time scales of minutes to years, experiments of this type can have sensitivity to any compact dark matter objects in the \( 10^{-7}M_\odot \) to \( 10M_\odot \) range.
Gravitational Lensing projects include the MACHO project (America and Australia), the EROS project (France), and the OGLE project (America and Poland). The MACHO collaboration, claims that a fraction equal to 20% of the dark matter in the galaxy consists of MACHO objects, with an average mass of about \(0.5M_\odot\)\(^{[20]}\). This suggests that MACHOs could be white dwarfs or red dwarfs, objects that are not completely dark, and therefore could be searched for with the Hubble Telescope and with proper motion surveys. These searches have shown that these objects can not account for a significant fraction of dark matter in our galaxy.

Moreover, the EROS2 collaboration does not confirm the signal claims by the MACHO group. Observations using the Hubble Space Telescope’s NICMOS instrument showed that less than one percent of the halo mass is composed of red dwarfs. This corresponds to a negligible fraction of the dark matter halo mass.

To conclude MACHOs’ searches have not found evidence for these objects as dark matter and no experiments suggest MACHOs could constitute the entirety of the dark matter.

### 2.2.2 Indirect Searches

Even though the WIMPs density is low enough to yield a negligible self-annihilation rate, in regions of high density dark matter would still annihilate producing signals that could be detected on Earth or by spacecraft telescopes.

WIMPs interacting while going through the Earth, the Sun, or any massive body would lose kinetic energy, and some of them may not have enough energy to escape the gravitational field, ending up trapped in the center of these objects. In this scenario WIMPs annihilation could produce \(b\bar{b}\) or \(W^+W^-\) pairs that then would
decay to neutrinos with high energy, 1/2 or 1/3 of the WIMPs mass, which would make them easily distinguishable from solar neutrinos \[15\]. Experiments such as Super-Kamiokande, the Icecube Neutrino Observatory and the Antarctic Muon and Neutrino Detector Array (AMANDA) are searching for this signal and so far have only set upper limits compatible to direct measurements.

Other particles produced by dark matter annihilation, such as gamma rays or SM pairs of particles and anti-particles could provide signals that prove the presence of dark matter. The Payload for Antimatter/Matter Exploration and Light-nuclei Astrophysics (PAMELA) \[21\], launched in 2006, the Fermi Large Area Telescope \[22\], launched in 2008, and the Alpha Magnetic Spectrometer \[23\], all indicate an excess of gamma rays or positrons, however there may be other mechanisms that produced this excess and thus this can not be considered a definitive indirect detection of dark matter.

2.2.3 Direct Searches

The WIMP-target interaction rate

The field of dark matter direct searches deals with the efforts of numerous experiments using different technologies to detect dark matter particles. In this section only experiments searching for WIMPs will be considered.

From theoretical considerations we can infer an estimate for the order of magnitude of the mean recoil energy together with the rate of interactions per day per kg that we should expect in dark matter detectors. In the following discussion we follow the calculations presented in Ref. \[24\].

In the lab reference frame, WIMPs with mass $M_\chi$ and velocity $v$ will have an
energy $E_i = 1/2 \cdot M_\chi v^2$, so that in an elastic scattering with target nuclei having mass $M_A$, the recoil energy deposited in the detector would be:

$$E_R = E_i r \left( \frac{1 - \cos \theta}{2} \right)$$

(2.8)

where:

$$r = \frac{4M_\chi M_A}{(M_\chi + M_A)^2}$$

(2.9)

and $\theta$ is the angle between the final velocities of the WIMP and the target nucleus.

If we consider the local circular velocity for WIMPs, $v_0 = 220 \pm 20$ km s$^{-1}$, with associated energy of these particles $E_0$, the mean recoil energy can be defined as $\langle E_R \rangle = E_0 r$, so that using $M_\chi = M_A = 50$ GeV/c$^2$, we obtain $\langle E_R \rangle \approx 15$ keV. This recoil energy is much lower than what solar neutrinos experiments expect to detect, and therefore poses a new challenge for the design of dark matter detectors.

The total rate of interactions $R_T$ in a detector depends on the range of energies deposited in the target material, with a cut-off for energies lower than the threshold energy of the detector:

$$R(E_{\text{th}}) = \int_{E_{\text{th}}}^{E_{\text{max}}} R(E_R) \, dE_R$$

(2.10)

It turns out that it’s easier to think in terms of $E_i$ rather than $E_R$ to solve the above integral, as explained in [24] :

$$R(E_R) = \int_{E_{R/\text{th}}}^{E_{\text{max}}} \frac{R(E_i) \, dE_i}{E_i r}$$

(2.11)

The rate depends on how many WIMPs could hit a target nucleus in our detector each second, how many target nuclei we have in the detector, and how likely are
the interactions to happen, i.e the scattering cross-section $\sigma$. Therefore the rate per kilogram of target can be written as:

$$ R(\vec{v} + \vec{v}_E) \, d^3v = n_0 f(\vec{v} + \vec{v}_E) \, v \cdot \frac{N_0}{A} \cdot \sigma $$

(2.12)

where we have assumed that we can describe the WIMPs velocities with a distribution $f(\vec{v} + \vec{v}_E)$ that takes into account the Earth velocity $\vec{v}_E$ with respect to the Galaxy, $n_0 = \rho_\chi / M_\chi$ is the number of WIMPs per unit volume with the mass density of WIMPs in the galaxy (estimated as 0.3 GeV/cm$^3$, \cite{25}), while $N_0$ is Avogadro’s number and $A$ is the atomic weight of the target material.

For our Galaxy we can assume a Maxwellian distribution for WIMPs velocities:

$$ f(\vec{v} + \vec{v}_E) = \frac{e^{-(\vec{v} + \vec{v}_E)^2/v_0^2}}{k} $$

(2.13)

$k$ being a normalization factor.

Now we can tackle Eq. 2.11 integrating over the accepted incoming WIMP energies, i.e. their velocities, for the simplified case where we assume Earth at rest $\vec{v}_E = 0$ and an infinite galactic escape velocity which yields $E_{i,\text{max}} = \infty$. To simplify the notation we set:

$$ R_0 \equiv \frac{2}{\sqrt{\pi}} \frac{N_0}{A} n_0 \sigma v $$

(2.14)
2.2. DARK MATTER SEARCH

\[ R(E_R) = \int_{E_R/r}^{\infty} \frac{1}{(M\chi v^2/2)r} \frac{R_0}{2\pi v_0^2} v e^{-v^2/v_0^2}(4\pi v^2 dv) \]  
\[ = \frac{R_0}{(M\chi v_0^2/2)r} \int_{v_{\min}}^{\infty} \frac{2}{v_0^2} e^{-v^2/v_0^2} v dv \]  
\[ = \frac{R_0}{E_0 r} e^{-E_R/E_0 r} \]  

Now we can integrate Eq. 2.17 and finally calculate the total rate taking into account energies above the energy threshold. Using \( E_{th} = 0 \) we find that the total rate is simply \( R_0 \), which can be expressed as:

\[ R_0 \sim \frac{500}{M\chi (\text{GeV})} \frac{\sigma_{0WN}}{1 \text{ pb}} \frac{\rho_\chi}{0.4 \text{ GeV}/c^2\text{m}} \text{ events kg}^{-1}\text{day}^{-1} \]  

where \( \sigma_{0WN} \) is the WIMP-nucleon cross-section. Substituting for a 50 GeV WIMP with \( \sigma_{0WN} = 1 \) pb, we obtain a rate of approximately 10 events kg\(^{-1}\)day\(^{-1}\).

Taking into account the motion around the Sun, which affects the velocity in the Galaxy, the Earth's velocity can be expressed as:

\[ v_E(t) \approx 232 + 15\cos \left( 2\pi \frac{t - 152.5}{365.25} \right) \text{ km/s} \]  

It can be shown [24] that the 6 % annual modulation given by Eq. 2.19 is reflected in a 3 % annual modulation in the total WIMP rate, if one takes into account all non-zero recoil energies.
Spin-independent and spin-dependent interactions

The WIMP-nucleon cross-section can be divided in two components, given by spin-independent $\sigma_{0WN}^{SI}$ and spin-dependent $\sigma_{0WN}^{SD}$ contributions, as explained in [26]:

$$\sigma_{0WN} = \sigma_{0WN}^{SI} + \sigma_{0WN}^{SD}$$ (2.20)

$$\sigma_{0WN,SI} = \frac{4\mu_p^2}{\pi} [Zf_p + (A-Z)f_n]^2$$ (2.21)

$$\sigma_{0WN,SD} = \frac{32G_F^2\mu_A^2}{\pi} \frac{J+1}{J} (a_p\langle S_p \rangle + a_n\langle S_n \rangle)^2$$ (2.22)

where $f_p$ and $f_n$ are the effective spin-independent coupling of WIMPs with protons and neutrons, while $a_p$ and $a_n$ refer to the spin-dependent couplings. The target material is characterized by the atomic number $Z$, the total nuclear spin $J$ and the expectation values of the proton and the neutron spins within the nucleus $\langle S_{p,n} \rangle = \langle N|S_{p,n}|N \rangle$.

If we assume $f_p \approx f_n$, the spin-independent WIMP-nucleus cross-section can be written as:

$$\sigma_{0WN,SI} \approx \frac{4\mu_A^2}{\pi} f_n^2 A^2 = \sigma_{SI} \frac{\mu_A^2}{\mu_n^2} A^2$$ (2.23)

where we have set:

$$\sigma_{SI} = \frac{4\mu_n^2 f_n^2}{\pi}$$ (2.24)

Eq. 2.23 is particularly useful to convert limits on the WIMP-nucleus to limits on the WIMP-nucleon cross-section $\sigma_{SI}$ as a function of $M_{\chi}$, which allows to compare results from different experiments, as shown in Fig. 2.6.

We can see that the spin-independent WIMP-nucleus cross-section is proportional
2.2. DARK MATTER SEARCH

Figure 2.6: Spin-independent WIMP-nucleon cross-section limits vs WIMP mass showing results from CDMSlite, 2013 (black), XMASS, 2013 (magenta dashed), SIMPLE, 2012 (cyan), CRESST-II-TUM40, 2014 (blue), CRESST-II, 2009 (red dotted), SuperCDMS LT, 2014 (light red), COUPP, 2012 (light blue), Edelweiss II, 2011 (green), CDMS, 2009 (green dashed) XENON 100, 2012 (light black) LUX, 2013 (magenta). This figure has been generated with the online tool Dark Matter Limit Plot Generator [27].

to the square of the atomic mass of the target nucleus, since scattering is approximately coherent across all the nucleons for the range of momenta considered.

For experiments looking for spin-dependent interactions, contributions from proton spin and neutron spin couplings often cancel each other so that separate limits have to be calculated for each type of interaction, and what actually matters for the total spin-dependent cross-section is the net nuclear spin. Similarly to the
spin-independent scenario, spin-dependent experiments can normalize their results in order to allow for a comparison of different technologies that do not depend on the target material, providing the spin-dependent WIMP-proton cross-section, \( \sigma_{SDp} \equiv 24G_F^2\mu_p^2a_p^2/\pi \), and the spin-dependent WIMP-neutron cross-section, \( \sigma_{SDn} \equiv 24G_F^2\mu_n^2a_n^2/\pi \). However, it is important to bear in mind that such results are often not exactly adapted for a fair comparison, as different experiments have different thresholds for the energy deposited by WIMPs interactions and thus do not test the same velocity range of these particles.

2.3 Bubble Chamber Technology Experiments

A fluid that is expected to be in a gaseous phase according to its thermodynamical parameters such as pressure and temperature, but still survives in a liquid phase, is defined as *superheated*.

When superheated, a fluid is in a metastable state, so that any nucleation that
obeys specific conditions, would trigger a violent phase transition. Nucleation can occur when enough energy is deposited and heats up a short track in the bulk [29], allowing the formation of a proto-bubble that can overcome the combined liquid pressure and surface tension that tries to suppress it.

As a consequence, a chamber filled with a superheated liquid, a bubble chamber, can be used to study how particles interact and deposit energy in the fluid, and therefore collect information on their properties. For this reason the bubble chamber technology has been exploited in several dark matter experiments, as described in the following sections.

2.3.1 Theory

The dynamics of bubble formation is governed by the outer liquid pressure $p_l$, the gas pressure inside the bubble $p_b$, and the surface tension $\sigma$ that acts as the elastic membrane of a balloon and produces an effective pressure expressed as $p_c = 2\sigma/r$ where $r$ is the bubble’s radius.

In order to have expansion of a bubble we require $p_b > p_c + p_l$ which determines for what minimum radius, the critical radius $r_c$ we will observe bubble growth:

$$r_c = \frac{2\sigma}{p_b - p_l} \quad (2.25)$$

Bubbles that are generated with a radius smaller than the critical radius $r_c$ will be squeezed and collapse, otherwise they will grow indefinitely, evaporating the surrounding liquid. As discussed in the previous sections, the mass of a WIMP particle that could constitute dark matter, is allowed to vary in a wide range, and in turn, so does the recoil energy.
2.3. BUBBLE CHAMBER TECHNOLOGY EXPERIMENTS

Hence, knowing what minimum energy $E_{th}$ is necessary to generate a bubble in the chamber provides information on what energy is deposited by the interacting particle. Actually, any deposited energy greater than $E_{th}$, within a given length $L_{th}$, would trigger a phase transition, and it is for this reason that bubble chambers can perform as energy threshold devices.

Bubble formation occurs when enough energy is provided by an ionizing particle as a heat spike to account for the total bubble creation energy, i.e. surface energy, chemical potential and heat of vaporization of the vapor in the bubble [30]:

$$E_{th} = 4\pi r_c^2 \left( \sigma - T \frac{\partial \sigma}{\partial T} \right) + \frac{4\pi}{3} r_c^3 \rho_b (h_b - h_l)$$

(2.26)

in which $T$ is the temperature of the system, $\rho_b$ is the bubble vapor density, and $h_b$ and $h_l$ are the specific enthalpies of the bubble vapor and superheated liquid, respectively. The stopping power must therefore be $dE/dx > E_{th}/(2r_c)$, otherwise either not enough energy is deposited, or it is deposited sparsely along an extended track, and the proto-bubble created will collapse and disappear.

2.3.2 PICASSO

The PICASSO (Project In CAnada to Search for Supersymntric Objects) experiment [31], which ran for approximately 15 years, ending operations in 2013, has been one of the first experiments to use superheated liquids for a direct search dark matter experiment.

The target material has been $C_4F_{10}$, chosen for its thermodynamical properties and the fact that the spin $1/2$ of the $^{19}$F nucleus is mainly due to the unpaired proton. As seen in Table [2.1] more specifically in the last two columns which describe the
2.3. BUBBLE CHAMBER TECHNOLOGY EXPERIMENTS

Enhancement factors of the WIMP-proton and WIMP-neutron spin-dependent cross-section (Eq. 2.22), the target nucleus $^{19}\text{F}$ provides a WIMP-proton cross-section that is almost one order of magnitude larger than any other element.

Table 2.1: Values of the atomic number $Z$, the total nuclear spin $J$, and the expectation values of the proton and neutron spins within the nucleus $\langle S_p, n \rangle$ for nuclei commonly used in detectors, with odd numbers of protons or neutrons, which lead to the enhancement factors used to calculate spin-dependent cross-sections in Eq. 2.22.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Z</th>
<th>Odd Nuc.</th>
<th>J</th>
<th>$\langle S_p \rangle$</th>
<th>$\langle S_n \rangle$</th>
<th>$\frac{4(S_p)^2(J+1)}{3J}$</th>
<th>$\frac{4(S_n)^2(J+1)}{3J}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{19}\text{F}$</td>
<td>9</td>
<td>p</td>
<td>1/2</td>
<td>0.477</td>
<td>-0.004</td>
<td>$9.1 \times 10^{-1}$</td>
<td>$6.4 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{23}\text{Na}$</td>
<td>11</td>
<td>p</td>
<td>3/2</td>
<td>0.248</td>
<td>0.020</td>
<td>$1.3 \times 10^{-1}$</td>
<td>$8.9 \times 10^{-4}$</td>
</tr>
<tr>
<td>$^{27}\text{Al}$</td>
<td>13</td>
<td>p</td>
<td>5/2</td>
<td>-0.343</td>
<td>0.030</td>
<td>$2.2 \times 10^{-1}$</td>
<td>$1.7 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{29}\text{Si}$</td>
<td>14</td>
<td>n</td>
<td>1/2</td>
<td>-0.002</td>
<td>0.130</td>
<td>$1.6 \times 10^{-5}$</td>
<td>$6.8 \times 10^{-2}$</td>
</tr>
<tr>
<td>$^{35}\text{Cl}$</td>
<td>17</td>
<td>p</td>
<td>3/2</td>
<td>-0.083</td>
<td>0.004</td>
<td>$1.5 \times 10^{-2}$</td>
<td>$3.6 \times 10^{-5}$</td>
</tr>
<tr>
<td>$^{39}\text{K}$</td>
<td>19</td>
<td>p</td>
<td>3/2</td>
<td>-0.180</td>
<td>0.050</td>
<td>$7.2 \times 10^{-2}$</td>
<td>$5.6 \times 10^{-3}$</td>
</tr>
<tr>
<td>$^{73}\text{Ge}$</td>
<td>32</td>
<td>n</td>
<td>9/2</td>
<td>0.030</td>
<td>0.378</td>
<td>$1.5 \times 10^{-3}$</td>
<td>$2.3 \times 10^{-1}$</td>
</tr>
<tr>
<td>$^{93}\text{Nb}$</td>
<td>41</td>
<td>p</td>
<td>9/2</td>
<td>0.460</td>
<td>0.080</td>
<td>$3.4 \times 10^{-1}$</td>
<td>$1.0 \times 10^{-2}$</td>
</tr>
<tr>
<td>$^{125}\text{Te}$</td>
<td>52</td>
<td>n</td>
<td>1/2</td>
<td>0.001</td>
<td>0.287</td>
<td>$4.0 \times 10^{-6}$</td>
<td>$3.3 \times 10^{-1}$</td>
</tr>
<tr>
<td>$^{127}\text{I}$</td>
<td>53</td>
<td>p</td>
<td>5/2</td>
<td>0.309</td>
<td>0.075</td>
<td>$1.8 \times 10^{-1}$</td>
<td>$1.0 \times 10^{-2}$</td>
</tr>
<tr>
<td>$^{129}\text{Xe}$</td>
<td>54</td>
<td>n</td>
<td>1/2</td>
<td>0.028</td>
<td>0.359</td>
<td>$3.1 \times 10^{-3}$</td>
<td>$5.2 \times 10^{-1}$</td>
</tr>
<tr>
<td>$^{131}\text{Xe}$</td>
<td>54</td>
<td>n</td>
<td>3/2</td>
<td>-0.009</td>
<td>-0.227</td>
<td>$1.8 \times 10^{-4}$</td>
<td>$1.2 \times 10^{-1}$</td>
</tr>
</tbody>
</table>

The last phase of the PICASSO experiment consisted of 32 cylindrical modules where $C_4F_{10}$ droplets with an average diameter of 200 µm, in a superheated state at ambient temperature and pressure, were suspended in polymerized gel matrix.
Each droplet can be seen as a tiny bubble chamber, where WIMPs could deposit energy and cause the formation of a vapour bubble with an associated explosive sound generation, recorded by nine piezoelectric devices per detector. Each module contains around 68 g of active mass of $^{19}$F per module. The active part of each detector is topped by mineral oil, which allows for periodic recompression of the modules in order to reconvert to a liquid phase those droplets where nucleation has occurred.

The detectors were installed at SNOLAB, the deepest operational underground laboratory in the world, located in Sudbury, Ontario, Canada. A depth of 2070 m guarantees a shielding from cosmic radiation that could be provided equivalently by 6010 m of water [32].

Since the minimum energy required to trigger a phase transition decreases with increasing temperature of the fluid, minimum ionizing radiation, such as cosmic muons, photons and $\beta$ particles, will only cause nucleation in the detector for relatively high temperatures, as shown in Fig. 2.8. It is therefore possible to completely eliminate nucleation caused by these uninteresting interactions through accurate temperature selection.

Furthermore, it was discovered by the PICASSO collaboration that nucleation given by alpha particles has a distinct signature in its sound generation, which can be detected by the piezoelectric transducers [33]. These interactions are loud enough to be discriminated from neutron recoils, which are indistinguishable from the expected WIMPs recoils.

This tool, explained by assuming that $\alpha$ particles generate more than one nucleation along their track, greatly improves the capacity of any superheated detector to reject background events and only listen for extremely rare events such as WIMPs.
2.3. BUBBLE CHAMBER TECHNOLOGY EXPERIMENTS

Figure 2.8: Response to different types of particles in superheated $C_4F_{10}$. From left to right: 1.75 MeV $\gamma$-rays and minimum ionizing particles (dot-dashed); $^{19}$F recoils modeled assuming the scattering of a 50 GeV WIMP (red); poly-energetic neutrons from an AcBe source (dotted); $\alpha$ particles at the Bragg peak from $^{241}$Am decays (open triangles); and $^{210}$Pb recoil nuclei plus $\alpha$ particles from $^{226}$Ra spikes (full dots). [31]

interactions.

Another project that uses that same superheated droplet detector technology is the SIMPLE (Superheated Instrument for Massive ParticLe Experiments) experiment, which operates a small program at a depth of 500 m in the Laboratoire Souterrain à Bas Bruit near Apt, France [34].

2.3.3 COUPP

While the use of a conventional bubble chamber for dark matter experiments was considered hardly achievable, due to the difficulty of avoiding frequent nucleation from the chamber’s walls, the COUPP (Chicagoland Observatory for Underground
Particle Physics) collaboration has proved that this is indeed feasible.

The superheated target material, $CF_3I$, is contained in a high purity synthetic fused silica jar topped with propylene glycol as buffer liquid, used to apply pressure and recompress the chamber after each event. In addition to piezoelectric devices, CCD cameras also observe the active fluid, allowing not only for signal triggering and event localization but also for visualization of multiple bubbles caused by several neutron scatters. The COUPP experiment is located in the SNOLAB facility, in the past using a chamber with 4.0 kg of active fluid [35], and now operates a 30.0 kg chamber.

The COUPP experiment also takes advantage of the alpha-neutron discrimination technique invented by the PICASSO collaboration, defining an acoustic parameter (AP) that describes the acoustic energy of each event. The excellent discrimination results for the 4.0 kg chamber are shown in Fig. 2.9.

2.3.4 PICO

In 2012, the PICASSO and COUPP collaborations have decided to work jointly to scale up the bubble chamber technology with the goal of constructing and operating a ton-scale detector. The first two letters of each experiment’s name have been combined to create the name of the new collaboration, PICO.

The first step of the PICO collaboration has been to re-use the COUPP 4kg chamber with a new PICASSO-like target material, $C_3F_8$, and a new buffer liquid, water, which has been collecting data since the last months of 2013.

Large efforts of R&D are being undertaken to improve different aspects of the future PICO ton-scale detector, and some of them are the topics of this thesis.
Figure 2.9: Data from the COUPP 4 kg chamber, showing the distribution of the acoustic parameter (AP), in red for the WIMP search mode where alpha particles are observed. The blue histogram shows the identical analysis for data taken in the presence of an AmBe neutron source. Defining an acoustic cut allows to select nuclear recoils with an acceptance of 95.8% as determined by the AmBe calibration.
Chapter 3

LAB as a scintillator in the PICO-250L detector

3.1 Introduction

3.1.1 Scintillators

Materials, either gases, liquids or solids, that can convert the energy of ionizing particles into light are called *scintillators* and are divided into two categories: inorganic and organic scintillators.

Scintillation light is then used as input for a photodetector that converts it into an electrical signal. Photomultiplier tubes (PMTs) are the most commonly used devices to read out light from scintillators. Since one photon produces only one photoelectron in the PMT, the signal of scintillators is usually weak, thus PMTs have a large gain that permits them to amplify this signal and produce a detectable output.

**Inorganic scintillators**

Inorganic scintillators are generally crystalline in nature (most commonly formed from alkali metals) and usually containing a tiny amount of impurity, also known as *activator*. In a crystal, the electronic energy states of an atom are represented
by bands (Fig. 3.1): the valence band is the allowed band with highest energy in a ground state, and is completely filled. At higher energy, the conduction band is found, which contains electrons that are free to migrate throughout the lattice, but is normally empty. Between the valence band and the conduction band there is an energy gap where there are no electron allowed energy levels, called the forbidden band.

![Diagram of energy bands](image)

Figure 3.1: Discrete levels within the forbidden band caused by crystal impurities.

When radiation interacts with the atoms in the crystal, enough energy may be provided to raise electrons to the conduction band, while the absence of the same electrons in the valence bond can be seen as positive charges, called the holes, that are also free to move in the lattice. Direct decay to the ground state is an inefficient method to produce light, moreover the energy of the emitted photon would be too high to be in the visible range.
For this reason, it is necessary to add energy states in the forbidden band, so that any decay between these states would emit light with visible wavelength. For this purpose, an activator or impurity, is added in small amounts to the crystal.

In this configuration, a hole can encounter an activator atom and ionize it, given that the ionization energy of the impurity is lower than the lattice site. Subsequently, an electron wandering in the conduction band may find the ionized activator and join it. This process may produce an activator atom in a neutral configuration with its own set of excited states, which are located between the valence and conduction bands of the lattice.

If the activator atom is formed in an excited state, with an allowed transition to the ground state, quick de-excitation through emission of light can occur. The half-life for this decay is generally on the order of 50-500 ns and constitutes the characteristic time of scintillation light in inorganic scintillators.

In case the decay of the excited state of the activator happens in a radiationless mode, the activator is called a quenching center and energy is lost to other processes. If the transition to the ground state is forbidden by selection rules, an additional amount of energy is required to raise the activator to a higher energy state where a transition to the ground state is allowed. In this last case, these metastable states are called traps and this competing mechanism produces delayed light emission, which is known as phosphorescence.

A second mechanism that produces light in the crystal, is the formation of an electron-hole pair that is electrostatically bound, an exciton, that has an energy characteristic of the exciton band, a thin band whose upper limit overlaps with the lower limit of the conduction band. Formation of an exciton occurs when the energy
3.1. INTRODUCTION

imparted to an electron is insufficient to raise it to the conduction band. An exciton can move freely through the crystal, until it finds an activator site, where again excited states in the forbidden band can be created locally.

The advantage of inorganic scintillators is that they have a high density and high atomic number, therefore possessing a high stopping power for radiation.

Organic scintillators

Organic scintillators are aromatic hydrocarbon compounds that consist of planar benzene-ring structures \[37,38,39\]. Scintillation occurs when free valence electrons belonging to the \(\pi\) molecular electronic orbitals, which are not associated with a particular atom in the molecule, interact with the incident radiation, reach an excited state and then de-excite in a radiative mode. Since light production in organic scintillators occurs at the molecular level, these materials can be used in different physical states.

For singlet electronic states (spin \(S = 0\)), labeled as \(S_0, S_1, \ldots\), in Fig. 3.2 the energy needed for electronic states transitions is 3-4 eV, but these states are also split into vibrational levels with typical energy spacing in the order of 0.15 eV. However, the average thermal energy (0.025 eV) of the electrons is lower than these quantities, and thus they are found in the ground state \(S_{00}\), where the second subscript has been added to label the vibrational level.

After an interaction with the incident radiation, molecules are found in highly excited states, from where they de-excite almost immediately (\(\leq 10\) ps) through a radiationless process, called internal degradation, that causes these states to de-excite to a vibrational level of the \(S_1\) electron state, which being unstable relaxes to
Figure 3.2: Electronic states of an organic molecule. [39] (re-drawn with the package tikz for this thesis)

the lowest level of this state.

Then, de-excitation to one of the vibrational states of the ground state $S_0$ occurs through the emission of light, known as prompt fluorescence, in a few nanoseconds.

The slow component of the spectrum, comes from the triplet state ($S = 1$). In fact, an excited triplet state gets to the ground state only through internal degradation. Transitions from $T_1$ to $S_0$ are prohibited by angular momentum and parity selection rules, which is the reason for the long decay time, on the order of microseconds, for this direct process called phosphorescence.

However, excited triplet states can interact with each other producing an excited
singlet state that decays as previously explained:

\[ T_1 + T_1 \rightarrow S_1 + S_0 + \text{phonons} \]  \hspace{1cm} (3.1)

Except for the transition \( S_{10} - S_{00} \), all the other decay energies are lower than the energy needed to excite molecules, hence there the emission spectra barely overlaps with the absorption spectra of organic scintillators. This is a phenomenon known as Stokes shift and is what assures that the material is transparent to its own scintillation light.

**Liquid scintillators**

When a large volume of a scintillator is needed, for example to study extremely rare interactions, such as those produced by neutrinos, liquid scintillator (LS) turns out to be an inexpensive solution. Moreover, LS is transparent to self-radiation and can fill detectors of any shape.

The main ingredient in the cocktail that characterizes a liquid scintillator is the solvent \[39\], which interacts with the ionizing particles, absorbing their energy, and consequently reaching an excited state.

After the solvent completes a transition to the ground state, its energy is efficiently absorbed by the solute, a fluor, which in turn decays from an induced excited state, emitting light. The solute is introduced in the solution to take advantage of its high fluorescence quantum yield and to shift the wavelength when it re-emits light, thereby avoiding bulk self-absorption by the solvent.

A secondary solute can be added to the cocktail, the most widely used being bis-MSB, in order to further shift the emission spectrum to longer wavelengths, to
better match the quantum efficiency profile of the photomultipliers that collect the light from the LS.

The scintillation efficiency refers to the fraction of light absorbed by the scintillator that is re-emitted as light. When radiative energy is converted into forms other than light (mainly into heat), we talk about quenching. In many liquids, the presence of dissolved oxygen can behave as a strong quenching factor and can produce a strong reduction in the fluorescence efficiency [40].

3.1.2 LAB and its use as buffer liquid in PICO

In large scale neutrino detectors, in addition to radiopurity, low cost and high availability, also fire safety (i.e high flash point), low toxicity and compatibility with construction materials are requirements that an ideal LS has to fulfill.

One of the solvents that is most widely used in neutrino experiments, and reinvented by the SNO+ experiment, is linear alkylbenzene (LAB), a compound that is largely manufactured in the industry of synthetic detergents. The name LAB labels a family of organic compounds with the formula $C_6H_5C_nH_{2n+1}$, i.e. several monoalkyl-derivatives of benzene, with the main components having an $n$ value ranging from 10 to 13 for the carbons in the side chain.

The other ingredient of the LS cocktail used in the SNO+ experiment is PPO (2,5-diphenyloxazole) with a concentration of 2 g/l, a fluor that re-emits as light the energy transferred from LAB, thus avoiding self-absorption and matching the spectral response of the photomultipliers. Optical and thermophysical properties of the cocktail will be discussed in detail in Sections 3.3 and A.1, respectively.

As presented in Sec. 2.3.4, the PICO-2L chamber uses $C_3F_8$ (octafluoropropane) as
an active fluid and water as buffer liquid. This combination introduced an unexpected new source of nucleation, given by the coarse emulsion at the $C_3F_8$-water interface. These nucleations can be clearly identified but their presence limits the live-time of the experiment.

To eliminate the instability at this interface, LAB has been investigated as a new buffer liquid and will be used in the second phase of the PICO-2L project.

### 3.2 Compatibility of LAB scintillation and active fluids

#### 3.2.1 LAB and $C_3F_8$

In order to also take advantage of the scintillation capabilities of LAB+PPO, which could be used to assess background radioactivity in the fused silica vessel, it is necessary to test the scintillation properties of LAB+PPO while the cocktail is interfaced with the active liquid $C_3F_8$.

To make these studies, scintillation spectra of gamma radioactive sources obtained with pure LAB + PPO have been compared to those obtained with LAB used as buffer liquid over $C_3F_8$.

**Experimental setup**

The experimental setup is shown in Fig. 3.3. A photomultiplier tube (PMT) with a 2" diameter window (9266 KA - Electronic Tubes) has been used to collect the scintillation light from the LAB + PPO cocktail. This PMT has been chosen as its diameter well fits the dimension of the LAB+PPO container and its quantum efficiency matches well the fluorescence spectrum of the fluor, PPO (as shown in Fig. 3.4).
The output signal of the PMT is then fed into a pre-amplifier (ORTEC 113), which integrates the charge signal and shapes the signal that is input to the amplifier (ORTEC 572A), which plays the two roles of amplifying and shaping the signal and whose signals are then stored in bins by the multi-channel analyzer (ORTEC 920E).

![Experimental setup diagram](image_url)

Figure 3.3: Experimental setup for the compatibility test of LAB scintillation with $C_3F_8$.

$C_3F_8$ has a boiling point $T_b = -36.7^\circ C$ at 1 atm, and consequently a high vapor pressure builds up when this substance is in a liquid state at room temperature, $p_v (20.0^\circ C) \approx 110\text{psi}$ [42]. Therefore, a Chemglass pressure vessel with a volume of 150 ml, guaranteed to be safe for pressures up to 150 psi, has been used to contain the fluids for this test.
Given that the vessel will be filled with LAB+PPO only partially, the small volume does not permit for full absorption of the gamma rays emitted by the radioactive sources used in this test ($^{207}\text{Bi}$, $^{60}\text{Co}$, $^{137}\text{Cs}$). However, the Compton edges are clearly recognizable in the spectra collected and can provide answers to the questions that motivate this experiment.

For an incident gamma ray of energy $E$, the energy $E'$ of a $180^\circ$ Compton scattered photon is given by:

$$E' = \frac{E}{1 + 2E/m_e c^2}$$  \hspace{1cm} (3.2)

where $m_e c^2$ is the electron rest energy. Therefore the energy $E_c$ deposited in the detector by such an event is equal to $E_c = E - E'$.

This is the maximum energy that can be transferred by the Compton process and
so the energy spectrum has a sharp cut off at this point. A summary of radioactive sources used in the test, their gamma ray energies and the associated Compton’s edge energies is shown in Table 3.1.

Table 3.1: Summary of radioactive sources used in the test, their gamma ray energies and the associated Compton’s edge energies.

<table>
<thead>
<tr>
<th>Source</th>
<th>$\gamma$ energies (keV)</th>
<th>Compton edge energies (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{207}$Bi</td>
<td>569, 1063</td>
<td>395, 860</td>
</tr>
<tr>
<td>$^{60}$Co</td>
<td>1173, 1332</td>
<td>966, 1121</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>661</td>
<td>482</td>
</tr>
</tbody>
</table>

Procedure

The vessel has been filled with approximately 90 ml of LAB+PPO, coupled to the PMT window with a thin layer of mating compound such as high vacuum grease, with a refractive index close to the refractive indices of the scintillator and the glass, to minimize interface losses, and surrounded with aluminum foil in order to increase light collection.

Three scintillation spectra have been obtained with the three different radioactive sources. The relative positions of pressure vessel, PMT and radioactive sources were kept constant during the different phases of the test by means of a homemade wooden jig, providing good repeatability of the measurements. A dark box was used to contain the wooden jig in order to protect the PMT from external light.

In the second phase of the experiment, a bath mixture of ethanol and ethylene glycol in dry ice, whose temperature can vary in the range (-12 °C, -78 °C) depending
on the volume fraction of ethylene glycol \[43\], has been used to cool down the vessel and the LAB in it to below the boiling of $C_3F_8$, to allow the latter to freely flow from its high pressure bottle to the vessel without excessive boiling.

Since at room temperature the $C_3F_8$ density ($\rho_{C_3F_8} = 1354.0 \text{ kg/m}^3$) is larger that of LAB ($\rho_{LAB} = 854.4 \text{ kg/m}^3$), the $C_3F_8$ takes its position at the bottom of the vessel, displacing the LAB volume that was already in the vessel. This requires careful consideration of the volumes chosen for the experiment (Figure 3.5), in order to preserve the relative position of the LAB volume with respect to the PMT window, and not introduce other differences between the two phases of the experiment other than adding $C_3F_8$. Also the radioactive source is accordingly moved with respect to the LAB location.

Figure 3.5: The vessel containing only LAB+PPO (left), and after adding 25 ml of $C_3F_8$ (right). Colours have been added to highlight the volumes.
Finally, scintillation spectra for the three radioactive sources have been obtained for LAB+PPO interfaced with octafluoropropane.

Results

Degradation of the LAB+PPO scintillation could occur as a slow process when the cocktail is interfaced with $C_3F_8$. For this reason measurements have been taken after 1, 3, 7, 14 and 31 days after having added $C_3F_8$ to the pressure vessel.

Results are shown in Figures 3.6, 3.7 and 3.8, for measurements taken using, respectively, the radioactive sources $^{207}$Bi, $^{60}$Co and $^{137}$Cs. The data for the six spectra have been collected over the same amount of time. The legend is shown only in the first figure to better visualize the scintillation spectra in the following figures.

![Figure 3.6: Experimental results for the compatibility test of LAB scintillation with $C_3F_8$. Source used: $^{207}$Bi.](image)
3.2. COMPATIBILITY OF LAB SCINTILLATION AND ACTIVE FLUIDS

Figure 3.7: Experimental results for the compatibility test of LAB scintillation with $C_3F_8$. Source used: $^{60}$Co.

It can be seen that not only there is no significant difference between the scintillation spectrum of pure LAB+PPO and the one where the cocktail is used as buffer liquid for $C_3F_8$, but also spectra collected after different intervals of time, up to one month, don’t show any important variation. The small changes observed between the spectra were expected, as in order to change radioactive source it has been necessary to manually operate on the apparatus, altering inevitably the relative positions of the different components.

We can conclude that the capabilities of LAB+PPO are not affected by the presence of $C_3F_8$, and as a consequence LAB+PPO could be used as a scintillator to assess levels of radioactivity inside any PICO chamber.
3.2. COMPATIBILITY OF LAB SCINTILLATION AND ACTIVE FLUIDS

3.2.2 LAB and $C\text{F}_3\text{I}$

It is important to test compatibility of LAB also with the second active fluid that could be used in the PICO experiment, $C\text{F}_3\text{I}$, or trifluoriodomethane. The first step is to test the chemical compatibility of LAB and $C\text{F}_3\text{I}$.

Similarly to $C_3F_8$, this fluid is also superheated at room temperature and pressure, having a boiling point $T_b = -22.5^\circ\text{C}$ at 1 atm, and vapor pressure $p_v (20.0^\circ\text{C}) \approx 62$ psi when in a liquid state at room temperature [44].

For this test, 30 ml of trifluoriodomethane have been transferred to the pressure vessel, previously filled partially with 30 ml of LAB without PPO, from its high pressure bottle using the same procedure exploited for $C_3F_8$ explained in the previous

---

Figure 3.8: Experimental results for the compatibility test of LAB scintillation with $C_3F_8$. Source used: $^{137}\text{Cs}$.
3.2. COMPATIBILITY OF LAB SCINTILLATION AND ACTIVE FLUIDS

Figure 3.9: The vessel containing LAB and $CF_3I$. Merging is observed in the lower half of the vessel.

The result is shown in Fig. 3.9, where the absence of a net interface between the two fluids shows that LAB merges with $CF_3I$. Consequently, LAB cannot be used as a buffer liquid with $CF_3I$. 
3.3 Production of scintillation light

As previously discussed, the use of a liquid scintillator, such as LAB, as a buffer liquid in the PICO bubble chamber, potentially gives the opportunity to assess the levels of radioactivity in the inner vessel of the detector.

In this section I present a study on light production and collection in the PICO 250L detector, articulated in the following steps:

- Consider what background interactions could cause scintillation in the buffer liquid volume;
- Evaluate the intensity of the interactions with highest probability;
- Design different configurations for light collection and study quantitatively their strengths and disadvantages.

3.3.1 Background interactions

The two backgrounds that superheated liquid experiments seek to limit and identify are alpha particles and neutrons. Alphas, being charged particles would interact with the molecules of the liquid scintillator as previously described in Section 3.1.1, while for the detection of a neutron it is necessary to consider the phenomenon of neutron capture.

Superheated liquid experiments consider fast neutrons to be a significant source of background events as they can produce recoiling nuclei with energies indistinguishable from WIMP interactions. To independently measure the fast neutron flux, we can measure the thermal neutron capture rate, as many of the fast neutrons will thermalize.
3.3. PRODUCTION OF SCINTILLATION LIGHT

within the bulk fluid and become detectable by using liquid scintillator. The detection of these thermal neutrons will be the focus of the following sections.

**Neutron-capture in $C_3F_8$**

The short notation for neutron capture is, in the case of $^{12}C$ as target, $^{12}C(n, \gamma)^{13}C$, which means that the isotope $^{12}C$ absorbs a neutron, forming $^{13}C$ in an excited state, which then decays to the ground state through emission of one or more gamma rays, either in a prompt or delayed mode.

Given that the directions of these gamma rays are not correlated, it is necessary to consider them singularly to assess their intensity and emission cross-section.

Carbon is found on our planet in two stable isotopes, which differ largely in their natural abundances, 98.9 % and 1.1 % for, respectively, $^{12}C$ and $^{13}C$, so that for the following discussion the latter will be considered negligible. The other element in the active liquid, fluorine, has only one stable isotope, $^{19}F$.

The likelihood of thermal neutron capture, i.e. the elemental capture cross-section for neutrons in thermal equilibrium with the ambient medium (usually quoted for a neutron velocity of 2200 m/s at 293.6 K) is much larger for the process $^{19}F(n, \gamma)^{20}F$ (9.6(5) mb), than for $^{12}C(n, \gamma)^{13}C$ (3.51(5) mb) [45].

Neutron capture data quoted in this work use the notation value(uncertainty). For instance, 9.6(5) mb stands for $9.6 \pm 0.5$ mb.

However, to properly assess the probability of the emission of a gamma ray after neutron capture, it is necessary also to take into account the branching ratios for the different gamma rays.
Hence, the partial $\gamma$-ray production cross-section is defined by:

$$\sigma_{\gamma}(E_{\gamma}) = \theta \cdot P(E_{\gamma}) \cdot \sigma_0$$

(3.3)

where $\theta$ is the isotopic abundance, $P(E_{\gamma})$ is the emission probability of the given $\gamma$-line, and $\sigma_0$ is the isotopic capture cross-section for neutrons with a velocity equal to 2200 m/s.

Tables 3.2 and 3.3 summarize the energies and likelihoods for gamma rays with partial elemental capture cross-section $\sigma_{\gamma} \gtrsim 1$ mb that are produced after neutron capture in, respectively, $^{12}C$ and $^{19}F$.

Table 3.2: Thermal neutron capture gamma rays, with highest partial production cross-section, produced in $^{12}C(n, \gamma)^{13}C$.

<table>
<thead>
<tr>
<th>$E_{\gamma}$ (keV)</th>
<th>$\sigma_{\gamma}$ (mb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1261.765(9)</td>
<td>1.24(3)</td>
</tr>
<tr>
<td>3683 .920(9)</td>
<td>1.22(3)</td>
</tr>
<tr>
<td>4945.301(3)</td>
<td>2.61(5)</td>
</tr>
</tbody>
</table>
3.3. PRODUCTION OF SCINTILLATION LIGHT

Table 3.3: Thermal neutron capture gamma rays, with highest partial production cross-section, produced in $^{19}\text{F}(n,\gamma)^{20}\text{F}$.  

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$\sigma_\gamma$ (mb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>583.561(16)</td>
<td>3.56(12)</td>
</tr>
<tr>
<td>656.006(18)</td>
<td>1.97(7)</td>
</tr>
<tr>
<td>665.207(18)</td>
<td>1.49(6)</td>
</tr>
<tr>
<td>983.538(20)</td>
<td>1.16(4)</td>
</tr>
<tr>
<td>1056.776(17)</td>
<td>0.95(3)</td>
</tr>
<tr>
<td>1633.53(3)</td>
<td>9.6(4)</td>
</tr>
<tr>
<td>6016.802(16)</td>
<td>0.94(4)</td>
</tr>
<tr>
<td>6600.175(16)</td>
<td>0.96(3)</td>
</tr>
</tbody>
</table>

*a* delayed by 11.163 s

Neutron-capture in LAB

In linear alkylbene, neutron capture can occur on hydrogen and carbon. Given that deuterium, $^2\text{H}$, has a tiny natural abundance ($\theta = 0.0115(70)$ %), we consider only the gamma ray produced by the most common isotope of hydrogen, protium or $^1\text{H}$, with an energy of 2223.25 keV and a partial cross-section $\sigma_\gamma = 0.3326(7)$ b.

The probability of neutron capture in the scintillator can be enhanced by loading it with an element that has a high cross-section for this type of interaction. The element with the highest neutron capture cross-section ($\sigma_0 = 48770(150)$ b) is gadolinium, with the most important contributions from the isotopes $^{155}\text{Gd}$ and $^{157}\text{Gd}$, as summarized in Table 3.4.

This property of gadolinium makes it very attractive for LS experiments where
the detection of the neutron flux is crucial. Experiments that study the oscillation of antineutrinos produced in nuclear reactors, such as Daya Bay \[46\], RENO \[47\] and Double Chooz \[48\], are interested in the distinctive signature of the inverse beta decay \(\bar{\nu} + p \rightarrow e^+ + n\), given by a prompt positron signal followed by capture of the neutron which occurs after it has been thermalized in the LS.

Neutron capture in Gd causes a release of about 8 MeV through a cascade of gamma rays, with the highest intensity ones, \(\sigma_\gamma \gtrsim 1000\) b, listed in Table 3.5. Partial cross-sections are listed using barn as units, rather than millibarns as in the previous tables.

Even though the compatibility between an efficient superheated \(C_3F_8\) and a scintillating buffer liquid loaded with gadolinium is not addressed in this thesis work, the extremely large cross-section for neutron capture and the high total de-excitation energy emitted through gamma rays creates a new interesting possibility for detection of background neutrons in the inner vessel.

Table 3.4: Natural abundances and isotopic neutron capture cross-sections for the isotopes of \(^{64}Gd\). \[45\]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>(\theta(%))</th>
<th>(\sigma_0)(b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{152}Gd)</td>
<td>0.20(1)</td>
<td>735(20)</td>
</tr>
<tr>
<td>(^{154}Gd)</td>
<td>2.18(3)</td>
<td>85(12)</td>
</tr>
<tr>
<td>(^{155}Gd)</td>
<td>14.80(12)</td>
<td>60900(500)</td>
</tr>
<tr>
<td>(^{157}Gd)</td>
<td>15.65(2)</td>
<td>254000(800)</td>
</tr>
<tr>
<td>(^{158}Gd)</td>
<td>24.84(7)</td>
<td>2.2 (2)</td>
</tr>
<tr>
<td>(^{160}Gd)</td>
<td>21.86(19)</td>
<td>1.4 (3)</td>
</tr>
</tbody>
</table>
3.3. PRODUCTION OF SCINTILLATION LIGHT

Table 3.5: Thermal neutron capture gamma rays, with highest partial production cross-section, produced in $^{157}Gd(n, \gamma)^{158}Gd$.\[45\]

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$\sigma_\gamma$ (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>780.174(10)</td>
<td>1010(22)</td>
</tr>
<tr>
<td>897.502(10)</td>
<td>1200(50)</td>
</tr>
<tr>
<td>897.611(10)</td>
<td>1090(50)</td>
</tr>
<tr>
<td>944.174(10)</td>
<td>3090(70)</td>
</tr>
<tr>
<td>962.104(10)</td>
<td>2050(130)</td>
</tr>
<tr>
<td>977.121(10)</td>
<td>1440(21)</td>
</tr>
<tr>
<td>1107.612(9)</td>
<td>1830(40)</td>
</tr>
<tr>
<td>1119.163(10)</td>
<td>1180(30)</td>
</tr>
<tr>
<td>1183.968(10)</td>
<td>958(60)</td>
</tr>
<tr>
<td>1185.988(9)</td>
<td>1600(90)</td>
</tr>
<tr>
<td>1187.122(9)</td>
<td>1420(90)</td>
</tr>
<tr>
<td>6750.11(5)</td>
<td>965(30)</td>
</tr>
</tbody>
</table>

3.3.2 Optical properties of LS

The most important optical properties of a LS are its light yield and its attenuation length.

Light yield

From the light yield one can determine the number of photons that are produced by the interaction of gamma rays and charged particles in the LS.
In the case of the cocktail used for the SNO+ experiment, i.e. LAB + 2g/L PPO, the light yield is 12,400 photons/MeV \([49]\). Knowing the light yield \(Y\) allows one to calculate the efficiency \(\eta\) of a scintillator, defined as the total energy emitted as scintillation light over the total energy of the interacting particle, assuming some emission wavelength \(\lambda_e\) for the scintillator.

For LAB+PPO, given that PPO has an absorption band of 280-325 nm and an emission band of 350-400 nm, with a peak of the emission spectrum equal to about \(\lambda_e = 360\) nm, we obtain:

\[
\eta = \frac{(hc/\lambda_e) \cdot Y}{10^6 \text{ eV}} = 4.3\% \tag{3.4}
\]

Antineutrino reactor experiments, which use a LS loaded with 0.1 % gadolinium in weight, add a secondary fluor to the cocktail of LAB+PPO.

This wavelength shifter, bis-MSB (1,4-bis-(o-methyl-styryl)-benzene), has an absorption band equal to 320-370 nm and an emission band of 380-450 nm, which brings the peak of the cocktail emission spectrum to about 430 nm (Fig. 3.10). For instance, the Daya Bay experiment cocktail consists of LAB + 3g/L PPO + 15 g/L bis-MSB.

The light yield of this cocktail is about 50 percent of anthracene, a LS often used as a reference, and is therefore approximately 10,000 photons/MeV \([51]\).

**Attenuation length**

The transparency of the LS to its own light determines what fraction of the light will be transmitted to the photodetectors. A measure of the transmission capacity of a liquid is given by its attenuation length, which includes effects due to absorption
and scattering, and is defined by the length that light of a certain wavelength has to travel in the material in order to be attenuated by a factor $1/e$, according to Beer's law $I(x) = I_0 e^{-x/l}$. The attenuation length depends strongly on the radiation's wavelength and generally longer wavelengths yield a higher transmittance.

The LS discussed in this work have large attenuation lengths. Pure LAB has an attenuation length of $28.6 \pm 0.6$ m at 430 nm, which has a lower value for the cocktail LAB+PPO+bis-MSB, of $24.6 \pm 0.5$ m, decreasing finally to $19.9 \pm 0.4$ m after the LS is loaded with gadolinium [52].

### 3.4 Collection of scintillation light

The design of the PICO-250L detector doesn’t allow for the total collection of the scintillation light from a LS buffer liquid. In fact, bellows are attached to the top of the vessel to control the pressure in the chamber and allow for recompression after bubble events, and obviously the buffer liquid is located on top of the active liquid. Thus, light collection has to occur from the sides of the cylindrical volume of the buffer liquid.
Photomultiplier tubes, which would be used to read out the light signal of the liquid scintillator, are a source of radioactivity, and thus can not be situated too close to the vessel that contains the superheated liquid.

Moreover, since the design of the PICO-250L detector presents an inner vessel, made of fused silica, enclosed in a pressure vessel filled with the hydraulic fluid used to control the pressure in the silica jar, recompression of the active fluid presents a hazard for the PMT windows.

In the following sections, three different cases will be considered qualitatively and quantitatively:

- Light propagates freely in the detector to PMTs with windows located on the outer vessel walls;
- Lofted light guides concentrate light on the PMTs windows;
- Compound parabolic concentrators focus light on the PMTs windows.

### 3.4.1 Free light propagation

Figure 3.11 depicts the inner vessel of the PICO-250L detector, where the volume of the buffer liquid and the active fluid have been colored in, respectively, light blue and orange. The pressure vessel is drawn with dashed lines.

The simplest configuration for the collection of scintillation light presents PMTs just outside the pressure vessel. A photomultiplier at a distance $R$ from the light source, having a photocathode with effective diameter $L$, would receive light emitted in a cone with apex angle $\alpha = 2 \cdot \arctan(L/2R)$. Figure 3.11 shows this configuration for a light source in the center of the LAB volume, and the solid angle under study...
Figure 3.11: Schematic representation of the PICO-250L inner and outer vessels, on scale.

is highlighted in green.

The solid angle $\Omega_{\text{cone}}$ is determined by $\alpha$ and its magnitude can be calculated by:

$$\Omega_{\text{cone}} = 2\pi(1 - \cos \frac{\alpha}{2})$$  \hspace{1cm} (3.5)$$

Finally, the ratio $\Omega_{\text{cone}}/4\pi$ determines what fraction of the total number of photons is collected on a single PMT.
3.4.2 Lofted concentrators

Light guides provide a way to direct light that is originally emitted from a large surface to a smaller surface where we are interested in analyzing the light signal, such as at a PMT window. Since we have seen that photomultipliers can’t be located at the boundary of the light emission volume, light guides, or concentrators, can greatly increase the amount of light that reaches the PMTs.

In this section we analyze a light guide that covers a quarter (a coverage of the azimuthal angle equal to $\pi/2$) of the lateral surface of the buffer liquid volume, guiding light rays to four PMTs located around the surface of the pressure vessel, as shown in Fig. 3.12. Given that such a surface is created with the software Inventor using the loft tool, this type of device is called *lofted concentrator*.

![Design of the lofted concentrator attached to the buffer liquid volume and to the effective area of the PMT.](image_url)

Figure 3.12: Design of the lofted concentrator attached to the buffer liquid volume and to the effective area of the PMT.

The light guides are located around the inner vessel of the detector and are immersed in LAB, which, in this case, is used as the hydraulic fluid that fills the pressure
vessel. For this reason, it is important that acrylic features the great advantage of being chemically compatible with LAB \[50\], which constitutes one of its advantages for use in neutrino experiments.

Moreover, this material offers large attenuation lengths for wavelengths similar to the emission peak of the WLS bis-MSB, i.e. around 420 nm, while the transparency worsens as the wavelength decreases. Acrylic is therefore an optimal material for light guides.

The matching between a cocktail such as LAB+PPO+bis-MSB, and the materials commonly used for light guides, extends also to aluminium, which may be used to coat the external surfaces of the light guides. In fact, at a 420 nm wavelength, aluminium provides a reflectance having a value of around 93% \[53\].

The percentage of light emitted in a scintillation event in the buffer liquid, and therefore the portion of light that can be potentially concentrated by the light guides, changes with the position of the event in the cylindrical volume of the buffer liquid.

In order to study how this percentage changes with position, we only have to consider the \(z\) and \(r\) coordinates of light emission sources, as the detector presents rotational symmetry around the \(z\)-axis. Moreover since the cylindrical volume also possesses symmetry with respect to horizontal plane passing through its center point, the range of the \(z\) coordinate can be restricted to one half of the height of the cylinder. This symmetry is slightly altered at the top of the vessel, where the flange elongates the upper portion of the cylinder (as shown in Fig.3.11).

Fig. 3.13 shows what solid angle coverage can be achieved as a function of the \(z\) and \(r\) coordinates of a light source in the LAB volume. As expected the coverage increases as we get closer to the cylinder walls, i.e. closer to the escape surface,
reaching a value of about 75 %, while it decreases to about 40 % when approaching either the bottom or top surfaces of the LAB volume, where light can not be collected, with light emissions occurring at the flange height presenting the lowest values for the solid angle coverage (∼ 30%) .

Figure 3.13: Solid angle coverage of the buffer liquid volume, when light is only collected at the lateral surface, as a function of the $z$ and $r$ coordinates of a light source in the LAB volume. Symmetries permit to restrict the ranges of points considered to the red bordered area of the cross-section of the buffer liquid volume shown in the lower part of the figure.
Simulation

The 3D geometry of the light guide extends from a portion of the cylindrical surface of the LAB that corresponds to a $\pi/2$ azimuthal angle around the z-axis, to a flat circular surface with diameter equal to the effective diameter of the PMTs considered, located on the pressure vessel walls, as shown in Fig. 3.12.

In order to simulate light emission in the LAB volume and assess what amount of light this design permits to collect, a 3D model of the inner vessel filled with LAB, surrounded by the four concentrators described, has been developed with the software Inventor Autodesk, and subsequently imported to the software TracePro.

A measure of the usefulness of the concentrators is given by the amplification factor, defined for a single PMT as:

$$AF = \frac{\text{number of rays collected using the light guide}}{\text{number of rays collected using the free propagation model}}$$  \hspace{1cm} (3.6)

Several PMTs (Hamamatsu R5912, R877, R329-02, R374) with different effective diameters (190 mm, 111 mm, 46 mm and 25 mm), can offer great ($>30\%$) spectral response in the range of emission of bis-MSB, and are therefore a good match for acrylic light guides.

Results for the amplification factor for the conical concentrators designed for the four PMTs considered are summarized in Table 3.6.

### 3.4.3 Compound parabolic concentrators

Concentrators are widely used in applications where the formation of an image is not necessary, as for instance in solar energy concentration. In the field of nonimaging
optics, the simplest way to generate the profile of a concentrator is called the *string method* [54].

**The string method**

To illustrate how this method works, we can consider an ellipse, the locus of the points whose distances from the two foci sums up to a constant value.

This definition suggests that to draw an ellipse we can tie each end of a string to one focus of the ellipse (points $E$ and $A$ in Fig. 3.14), and then draw the whole shape using a pencil (point $P$) that keeps the string taut.

Moreover, we notice that in this way we’re drawing a curve whose normal at each point divides into two equal parts the angle given by the two segments of the string. Expressed in other words, we could see the string as a light ray coming from one of the foci, that hits the ellipse curve where the pencil is, and is reflected to the other foci. We have just drawn the shape of a concentrator for point-like source and receiver.

![Figure 3.14: Drawing an ellipse using the string method.][54]

With a variable amount of creativity that depends on the shapes of the sources and receivers considered, the string method can be adapted and applied to the generation of any concentrator’s profile. First, one side of the profile of the concentrator is obtained with the string method in 2D, and then this profile is revolved around an
appropriate axis to obtain a 3D surface.

In our case, since we aim at concentrating rays from the cylindrical surface of the inner vessel to the flat surface of a PMT window, we tie one end of the string to an end of the effective diameter, and the other end to an extreme of the cylinder, as in Fig. 3.15. The length of the string is chosen when, in this configuration, the string is kept taut by a pencil located at the end of the photocathode where the string is not tied \[55\].

Concentrators designed in this way generally have a profile given by a parabolic arc (\(AC\) in Fig. 3.16) reflected along the vertical axis to obtain \(BD\) and concentrate effectively only light rays that make an angle smaller than the acceptance angle \(\theta\). They are generally known as compound parabolic concentrators, or CPCs.

A 3D model of the CPC has been built using the software Inventor, and then imported in the software TracePro for a computation of the amplification factors. The results for the amplification factors for the CPC designed for the four PMTs considered are summarized in Table 3.6.

Table 3.6: Amplification factor for the lofted and CP concentrators designed for the PMTs (Hamamatsu R5912, R877, R329-02, R374) with different effective diameters (190 mm, 111 mm, 46 mm and 25 mm).

<table>
<thead>
<tr>
<th>Effective diameter</th>
<th>(190 mm)</th>
<th>(111 mm)</th>
<th>(46 mm)</th>
<th>(25 mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lofted concentrator</td>
<td>6.55</td>
<td>4.97</td>
<td>5.09</td>
<td>4.29</td>
</tr>
<tr>
<td>CPC</td>
<td>4.96</td>
<td>4.83</td>
<td>4.95</td>
<td>4.96</td>
</tr>
</tbody>
</table>
3.4. COLLECTION OF SCINTILLATION LIGHT

Figure 3.15: Illustration of the string method. From this top view of the PICO-250L detector we see how the string (thick red line) is initially tied at one extreme of the inner vessel (blue line) and at one extreme of the photocathode of the PMT (in this figure: Hamamatsu R5912, 190mm effective diameter, located around the pressure vessel (green line) ), while being taut by a pencil at the other extreme of the photocathode. Subsequently one side of the profile of the concentrator (black line) is drawn moving the pencil and still keeping the string taut (thin red line).

3.4.4 Results

In order to compute the number $N$ of photons that are effectively converted into photoelectrons at the photocathode of a PMT, for the interaction of a specific $\gamma$-ray, several factors have to be taken into account (Eq. [3.7]).
3.4. COLLECTION OF SCINTILLATION LIGHT

Figure 3.16: A CPC concentrator. Light that enters the entrance CD with an angle smaller than the acceptance angle $\theta$ is concentrated on the exit AB. \[54\]

We first determine the number of photons produced in the liquid scintillator following a $\gamma$-ray interaction, $N_p$, multiplying the energy $E$ of the $\gamma$-ray by the light yield $Y$.

The result is then multiplied by the geometrical factor $G$ (the fraction of the emitted photons that reach the PMT window) determined through simulation as described in the Sections \[3.4.2\] and \[3.4.3\].

Finally, a correction given by the quantum efficiency $Q$ of the PMT (30%) is applied to obtain the number of photons that effectively yield a photoelectron at the PMT photocathode.

\[ N = N_p \cdot G \cdot Q = (E \cdot Y) \cdot G \cdot Q \]  \hspace{1cm} (3.7)

Results corrected for the quantum efficiency of the PMT, obtained for the $\gamma$-rays with highest partial production cross-section, using the free light propagation model and for the two designs of concentrators and the four different models of PMT considered,
Table 3.7: Results for the number of photons effectively converted into photoelectrons at the PMT photocathode are presented for the three $\gamma$-rays with highest partial production cross-section for neutron-capture in $^{12}C$ and $^{19}F$. The first two rows contain information about the $\gamma$-rays considered (sequentially, energy and cross-section), while the first two columns present, sequentially, the effective diameter of the PMT considered and the design used to study light propagation in the detector. At the intersection of any row and line, one can find the number of photons obtained for that specific combination of $\gamma$-ray, PMT and light propagation design.

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>1663</th>
<th>583</th>
<th>4945</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cross-section (mb)</td>
<td>9.6</td>
<td>3.6</td>
<td>2.6</td>
</tr>
<tr>
<td>No conc.</td>
<td>23.8</td>
<td>8.4</td>
<td>70.9</td>
</tr>
<tr>
<td>190 mm</td>
<td>141.4</td>
<td>49.6</td>
<td>420.5</td>
</tr>
<tr>
<td>CPC</td>
<td>118.1</td>
<td>41.4</td>
<td>351.3</td>
</tr>
<tr>
<td>No conc.</td>
<td>8.2</td>
<td>2.9</td>
<td>24.3</td>
</tr>
<tr>
<td>111 mm</td>
<td>40.6</td>
<td>14.2</td>
<td>120.9</td>
</tr>
<tr>
<td>CPC</td>
<td>39.4</td>
<td>13.8</td>
<td>117.2</td>
</tr>
<tr>
<td>No conc.</td>
<td>1.3</td>
<td>0.5</td>
<td>4.0</td>
</tr>
<tr>
<td>46 mm</td>
<td>6.8</td>
<td>2.4</td>
<td>20.1</td>
</tr>
<tr>
<td>CPC</td>
<td>6.6</td>
<td>2.3</td>
<td>19.5</td>
</tr>
<tr>
<td>No conc.</td>
<td>0.4</td>
<td>0.2</td>
<td>1.3</td>
</tr>
<tr>
<td>25 mm</td>
<td>1.9</td>
<td>0.7</td>
<td>5.5</td>
</tr>
<tr>
<td>CPC</td>
<td>2.2</td>
<td>0.8</td>
<td>6.4</td>
</tr>
</tbody>
</table>

Table 3.6 shows that the designs of lofted concentrators and CPC offer comparable amplification factors, with a larger difference for the case of the PMT with a 190 mm effective diameter.
3.4. COLLECTION OF SCINTILLATION LIGHT

The higher the number of photons that produce a signal, the easier it is to discriminate this signal from the noise and to gain information on the gamma ray that has interacted in the LS. However, we can set a threshold of one photon for a signal that can be detected by the PMT.

Table 3.7 shows that for the PMT with largest diameter (190 and 111 mm) the free propagation model yields a remarkable number of photons collected. On the other hand, the importance of the concentrators increases for the PMT of small diameter (46 and 25 mm), where the free propagation model often fails to provide at least one photon.

The use of PMT of small size is preferred for the low radioactivity introduced inside the detectors, and for the convenience of installing such small devices around the pressure vessel. Given that the 25 mm diameter PMT can’t detect the low energy gamma-rays even when equipped with concentrators, the smallest PMT that would be able to detect all the gamma-rays with high cross-section is the 46 mm diameter PMT.
Chapter 4

Acoustic studies of the PICO-250L detector

4.1 A 2D acoustic model for PICO

As explained in Section 2.3, events that occur in a bubble chamber produce a detectable sound emission that is recorded by piezoelectric devices around the chamber. In order to extract as much information as possible about these events, it is necessary to study and understand not only what phenomena take place when a phase transition produces a bubble and what drives the bubble expansion, but also how the sound signal produced by such an event propagates in the chamber and is transmitted in different components of the detector, until it reaches the piezoelectric devices.

While the different theories provides insight on the first part of these questions, namely the formation and expansion of a bubble, the propagation of a sound signal in a volume and its reception at the piezoelectric device boundaries creates a complex problem that cannot be solved analytically.

As a consequence, our efforts focus on the development of a simulation that allows the study of sound propagation in a bubble chamber, such as a scaled version of the PICO-250L.
4.1. A 2D ACOUSTIC MODEL FOR PICO

Superheated liquid experiments exhibit a distinct difference in the acoustic energy of the signals recorded by piezoelectric devices for interactions of alphas and neutrons. This discovery, that allows discrimination between alpha and neutron events, is based on the fact that an alpha particle deposits its entire kinetic energy in a short track on the order of tens of \( \mu \text{m} \), triggering the formation of several proto-bubbles, while the shorter track produced by the scatter of neutrons (or WIMPs) on nuclei, on the sub-\( \mu \text{m} \) order, can only cause the formation of a single bubble \cite{56}. As a consequence, the acoustic traces of alpha events are observed as being louder than those of neutron events.

In this work, we aim at reproducing a clear difference in the intensity of sound signals produced by events where a variable number, \([1 : 4]\), of bubbles are placed along an alpha track of 35 \( \mu \text{m} \). The length of this track is in fact yielded by alpha particles with a 5.6 MeV energy, while the chosen range of number of multiple nucleations along an alpha track is characteristic of alpha interactions in superheated liquids \cite{33}.

Moreover, given the much larger size of the PICO-250L detectors with respect to the previous detectors used in the COUPP and PICO projects, it is of interest to consider where the piezoelectric devices should be located, thus we determine whether the location of the piezoelectric influences the efficiency of the discrimination technique, providing information on their optimal position around the inner vessel.

Simulations have been carried out with an Acoustic Module of the multiphysics software COMSOL, widely used in many areas of physics.
4.1. Details of the model

Mesh size and degrees of freedom

In order to assess the complexity of a model that is to be simulated, the number of degrees of freedom (DOF) of the model has to be carefully considered. In fact, the COMSOL Acoustic Guide warns that “even on a 64-bit system, models with more than a few million DOFs are cumbersome to handle.”

The number of degrees of freedom depends on the mesh size used to discretize the model. For an acoustic model, in order to resolve properly the acoustic propagation, the mesh size needs to be, in the least demanding of the cases, a quarter of the wavelength that is to be studied.

Therefore, since each mesh point has three degrees of freedom in a 3D model the number of DOFs in a sufficiently resolved mesh is about \((3 \cdot 4)^3\), or 1728, times the model volume measured in wavelengths cubed.

Sound waves in the model are generated through a Gaussian explosion, an explosion whose intensity varies according to a Gaussian curve, as described in Ref. [57]. The frequency and therefore the wavelength of these waves is determined by the rise time of the Gaussian explosion.

Theoretical calculations based on the Seitz model show that for the active fluids used in the PICO family experiments, the vast majority of the acoustic power of a bubble event is emitted in the first few microseconds [58]. We have therefore chosen a rise time \(t_0 = 1\,\mu\text{s}\), which, using a speed of sound \(c\) for \(C_3F_8\) selected from Ref. [42], yields a wavelength \(\lambda_0 = c \cdot 2t_0 = 664\,\mu\text{m}\).

The Fourier Transform of the Gaussian pulse \(g(t)\) (Eq. 4.5) is given by (neglecting
cutoff effects):

\[
G(\omega) \equiv \int_{-\infty}^{+\infty} g(t) e^{-i\omega t} dt = \frac{2A\sqrt{\pi}}{\omega_0} e^{-\frac{\omega^2}{\omega_0^2} - i\omega t_0} \tag{4.1}
\]

where:

\[
\omega_0 = 2\pi f_0 = 2\pi / t_0 \tag{4.2}
\]

Since the magnitude of the Fourier transform decreases quickly with increasing angular frequency \(\omega\), and most of it is concentrated between \(-\omega_0\) and \(\omega_0\), it is only necessary to resolve wavelengths corresponding to the angular frequency \(\omega_0\) or equivalently the frequency \(f_0\).

Taking into account the simpler case of the PICO-2L chamber, which has a volume of 3 liters, we can calculate the number of DOF as:

\[
DOFs = \frac{1728 \cdot V}{\lambda_0^3} = 1728 \cdot \frac{3 \cdot 10^{-3} \text{ m}^3}{664^3 \cdot 10^{-18} \text{ m}^3} \approx 18000 \cdot 10^6 \tag{4.3}
\]

which is much obviously more than the few million DOFs a 64-bit pc can handle. Thus, a 2D model of the PICO-250L chamber has been chosen for the initial simulations, with dimensions scaled down by a factor 15 (from a chamber full height of 145 cm to approximately 9.7 cm).

**Geometry**

The geometry of the model is shown in Fig. 4.1

Piezoelectric devices, with dimensions of 5 × 9 mm, such as those used in the PICO-2L detector, have been positioned in three different locations:
4.1  A 2D ACOUSTIC MODEL FOR PICO

Figure 4.1: 2D scaled model of PICO-250L detector studies in this work. Points chosen for the bubble events are shown on a grid with cell width equal to 5 mm.

- Upper part of the vessel, at half-height of the cylindrical volume of the buffer liquid [top piezo];
- Middle part of the vessel, at half-height of the cylindrical volume of the active liquid [middle piezo];
- Lower part of the vessel, at the bottom of the hemi-spherical volume of the active liquid [bottom piezo].
**Materials**

The COMSOL software provided a library of built-in materials that already includes all the properties needed for acoustic simulation, such as density and bulk modulus (Young’s modulus) for fluids (solids), but also the quantities that are necessary to build the equations that describe the physics of piezoelectric devices, such as mechanical properties (compliance or stiffness), electrical properties (permittivity), and piezoelectric coupling properties.

The active liquid, $C_3F_8$, was not part of the built-in materials and has been implemented using data on density and bulk modulus (speed of sound) as found in [42]. The other materials chosen were: water for the buffer liquid, fused silica for the jar, and lead zirconate titanate ceramic (PZT-5H) for the piezoelectric transducers.

**Physics domains**

The model is divided into several physics domains, each with a different set of variables and equations solved by the software COMSOL. For the domains where sound propagates, and the acoustic pressure is studied over a certain time range at quiescent background conditions, such as the active fluid and the buffer liquid, a Transient Acoustics Pressure model is applied. The wave equation solved by the simulation to obtain the acoustic pressure $p = p(x, t)$ is:

$$\frac{1}{\rho c^2} \frac{\partial^2 p}{\partial t^2} + \nabla \cdot \left(-\frac{1}{\rho}(\nabla p - q_d)\right) = Q_m$$

(4.4)

where $c$ is the speed of sound and $\rho$ is the equilibrium density, while $q_d$ and $Q_m$ are dipole and monopole sources, respectively. Eq. 4.4 can be derived by combining the...
4.1. A 2D ACOUSTIC MODEL FOR PICO

linear equation of state ($p = K \cdot s$), the linear equation of continuity ($\partial p / \partial t + \nabla \cdot (\rho_0 \vec{u}) = 0$) and the linear equation of force ($\partial u / \partial t + \nabla p / \rho_0 = 0$), where $K$ is the adiabatic bulk modulus, $s$ is the condensation, $\rho_0$ is the equilibrium density and $\vec{u}$ is the fluid particle velocity [59].

To simulate a nucleation event in the bulk of the active fluid, a perturbation is generated by a Gaussian pulse with amplitude $g(t)$ defined as a function of time as:

$$g(t) = \begin{cases} 
  A e^{-\pi^2 (t/\tau - 1)^2}, & \text{for } 0 < t < 2\pi, \\
  0, & \text{otherwise} 
\end{cases}$$

(4.5)

where $\tau = 1/f_0$, and $A$ is the maximum amplitude of the Gaussian explosion describing the water flow (measured in $m^2/s$) away from the source.

As previously discussed in Section 2.3.1, the energy of the interacting particle has to be deposited in a certain length in order to trigger the formation of a bubble. However, the subsequent growth of a proto-bubble is driven by the energy of the surrounding superheated liquid, rather than by the kinetic energy deposited in the fluid [33]. As a consequence, the amplitude of each explosion in the multiple bubble events has the same value as the one of the single bubble event.

We can now rewrite Eq. 4.4 as:

$$\frac{1}{\rho c^2} \frac{\partial^2 p}{\partial t^2} + \nabla \cdot \left( -\frac{1}{\rho} (\nabla p) \right) = S(x, t)$$

(4.6)

where the point-source term on the right-hand side is given by:

$$S(x, t) = \frac{dg}{dt}(t)\delta(2)(x - x_0)$$

(4.7)
4.1. A 2D ACOUSTIC MODEL FOR PICO

with the source being located at \( x = x_0 \).

The Transient Acoustics Pressure model deals with transmission and reflection of the sound signal at the interface between the buffer fluid and the active fluid, which are modeled with selected values of thermophysical properties such as bulk modulus and density. In order to allow an outgoing sound wave to leave the vessel with minimal reflections, the top boundary of the buffer fluid has been modeled with a radiation boundary condition called Spherical Wave Radiation. In fact, bellows (that transmit pressure allowing for recompression of the active fluid) will be attached at the top of the PICO-250L detector and offer a vertical spatial extension of the buffer fluid volume that eliminates reflections at the top of the quartz vessel.

The walls of the vessel are treated as a Linear Elastic Material, so that there is a linear relationship between the components of stress and strain in the material. As a consequence, a linear elastic equation for the displacements is solved computationally, in order to connect the acoustics pressure variation in the fluid domain with the structural deformation in the solid domain, therefore modeling the transmission of sound through the quartz vessel and the reflection of sound waves at the quartz-fluids interface.

Finally, the piezoelectric transducers are naturally modeled as a COMSOL defined Piezoelectric Material, where both the direct and inverse piezoelectric effects can be modeled and the piezoelectric coupling can be formulated using either of two equivalent formulations, the strain-charge (Eq. 4.8) and the stress-charge (Eq. 4.9) matrix form:

\[
S = s_E \cdot T + d^f \cdot E \\
D = d \cdot T + \varepsilon_T \cdot E
\] (4.8)
4.1. A 2D ACOUSTIC MODEL FOR PICO

\[
T = c_E \cdot S - e^t \cdot E \\
D = e \cdot S + \varepsilon_S \cdot E
\]  \tag{4.9}

which connects the mechanical strain \( S \), mechanical stress \( T \), electric field \( E \), and the electric displacement field \( D \) through the elastic compliance \( s \), stiffness coefficient \( c \), electric permittivity \( \varepsilon \), piezoelectric coupling coefficients \( d \) and \( e \). The subscripts in the equations describe the conditions under which the material property data was measured. For example, the subscript \( E \) on the compliance matrix \( s_E \) means that the compliance data was measured under at least a constant, and preferably a zero, electric field.

The coefficients are evaluated when the field in the subscript is kept constant and can be converted between the two forms as:

\[
c_E = s_E^{-1} \\
e = d \cdot s_E^{-1} \\
\varepsilon_S = \varepsilon_T - d \cdot s_E^{-1} \cdot d^t
\]  \tag{4.10}

which turns out to be particularly useful for converting the parameters of the piezoelectric materials provided by the manufacturers into the input quantities required by the COMSOL software.

Piezoelectricity is an effect given by the polarization of a crystal that occurs as consequence of a mechanical stress applied on the material, but the reverse mechanism falls under the same header. We can imagine that the piezoelectric properties of a material strongly depends on the direction of the applied stress.

The piezoceramic material PZT-5H is a transversely isotropic material, which is a special class of orthotropic materials. This material has the same properties in
one plane (isotropic behavior) and different properties in the direction normal to this plane.

However, COMSOL’s material library data is entered in a form which assumes that the crystal polarization is aligned with the global coordinate y axis. As a consequence, to get correct results from all the piezoelectric devices, the orientation of the upper and middle ones must be rotated so that the material polarization direction is aligned with the x direction.

**Convergence and time step**

It is possible to provide an upper limit on the time step using a fundamental necessary, but not sufficient, condition for convergence of any numerical scheme, called the CFL condition (from the names of Richard Courant, Kurt Friedrichs and Hans Lewy, who first identified it in 1928 [60]).

This condition requires that the physical domain of dependence must be contained in the numerical domain dependence, more simply expressed as keeping the time step small so that information has enough time to propagate through the space discretization and has the simple form:

\[
N_{CFL} \equiv \left| \frac{c \cdot \Delta t_{step}}{\Delta x_{mesh}} \right| \leq 1
\]  \hspace{1cm} (4.11)

where \( N_{CFL} \) is the CFL number, \( c \) is the velocity of information propagation in the model, \( \Delta t_{step} \) is the time step and \( \Delta x_{mesh} \) is the smallest segment in the space discretization.

In order to select the ideal \( N_{CFL} \), which in turn determines the upper limit on the
time step of the simulation, a simple convergence study has been carried out. Comparing outputs of the model, which have been obtained using different CFL numbers (see Fig. 4.2), we select the $N_{CFL}$ that yields an output which is essentially identical to the output obtained with the smaller $N_{CFL}$ studied. Following this procedure, we choose the CFL number $N_{CFL} = 0.05$.

Figure 4.2: The figure shows plots of the acoustic pressure along a representative portion of y-axis of the detector at a specific time, obtained for models with different CFL numbers. We select the CFL number, namely $N_{CFL} = 0.05$, that yields an output that only shows negligible differences if compared with the plot obtained using the next smaller CFL studied.
4.1.2 Results

The equations presented above only yield a unique solution for a specific set of initial conditions, thus, in order to produce a sufficient amount of data from all relevant locations in the detector, acoustic signals are studied for Gaussian explosions generated at different locations in the 2D surface of the active fluid.

These locations are spatially organized on a grid, as shown in Fig. 4.1. For each point in the grid we acquire the output signals of the three piezoelectric devices positioned around the vessel, repeating this process for events with 1, 2, 3 and 4 Gaussian explosions distributed along a track.

Non-ideal conditions such as those encountered in real life experiments, lead to a complex analysis of the acoustic traces, in order to eliminate noise but also isolate and study only the portion of data that contains physical meaning. However, in this simulation we can proceed with a straightforward analysis and define an acoustic parameter AP as the logarithm of the sum of the absolute values of the piezoelectric devices output (i.e. different from the AP definition used in the superheated liquid experiments):

\[
AP = \log \left( \sum_t |V(t)| \right)
\]  

(4.12)

Output signals are recorded for a 1 ms time interval, discretized with equidistant 500 points, i.e. using a 500 kHz sampling frequency. This frequency refers to the number of stored solutions that are used to visualize the piezo signal and calculate the AP, and is different from the frequency \( f_0 \) (see Eq. 4.2) of the acoustic signal that is studied in the simulation.
4.1. A 2D ACOUSTIC MODEL FOR PICO

For each piezo, we obtain four distributions of AP parameters, one for each simulated number of Gaussian explosions along an alpha track. They are shown in Figures 4.3a, 4.3b and 4.3c for respectively the top, middle and bottom piezoelectric device. These histograms show that there exists a clear separation between single bubble and multiple bubble events. Moreover, this separation does not increase linearly with the number of Gaussian explosions, as one would expect from the fact that the amplitude of explosions used for multiple bubble events is the same as the one used in the single-bubble events.

In order to assess quantitatively the separation between the distributions of AP parameters, produced by a piezo with label \( p \), it is useful to consider the Bhattacharyya distance \( B_p(j,k) \), a measure widely used in the engineering and statistical sciences (for example in the field of image processing and speaker recognition), which using mean and standard deviation calculated for the distributions \( j \) and \( k \) can be exploited in the form [61]:

\[
B_p(j,k) = \frac{1}{4} \ln \left( \frac{1}{4} \left( \frac{\sigma^2_{p,j}}{\sigma^2_{p,k}} + \frac{\sigma^2_{p,k}}{\sigma^2_{p,j}} + 2 \right) \right) + \frac{1}{4} \left( \frac{(\mu_{p,j} - \mu_{p,k})^2}{\sigma^2_{p,j} + \sigma^2_{p,k}} \right) \tag{4.13}
\]

It can be seen that for two identical Gaussian distributions (\( \sigma_{p,j} = \sigma_{p,k}, \mu_{p,j} = \mu_{p,k} \)) the distance goes to zero, while it increases indefinitely for an increasing difference between the means \( \mu_{p,j} \) and \( \mu_{p,k} \).

Bhattacharyya distances between the single bubble and the multiple bubbles distributions for a single piezo, allow one to determine how the positions of the piezos affect the discrimination between events. Bhattacharyya distances for the top \( (p = 1) \), middle \( (p = 2) \) and bottom \( (p = 3) \) piezos are summarized in Table 4.1.
4.1. A 2D ACOUSTIC MODEL FOR PICO

Figure 4.3: Distributions of the AP parameter obtained for the top, middle and bottom piezoelectric devices for events with, from top to bottom in this figures, 1, 2, 3 and 4 explosions. Each distribution contains 45 events.
Table 4.1: Bhattacharyya distances $B_p(1, i)$ for distributions between a single bubble and with a number $i$ of explosions are listed for the top, middle and bottom piezos.

<table>
<thead>
<tr>
<th></th>
<th>$B_p(1, 2)$</th>
<th>$B_p(1, 3)$</th>
<th>$B_p(1, 4)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top piezo</td>
<td>6.8</td>
<td>16.4</td>
<td>26.3</td>
</tr>
<tr>
<td>Middle piezo</td>
<td>0.7</td>
<td>1.9</td>
<td>3.0</td>
</tr>
<tr>
<td>Bottom piezo</td>
<td>1.0</td>
<td>2.5</td>
<td>4</td>
</tr>
</tbody>
</table>

Moreover, it is useful to compare different piezos by determining how many events that are contained in the multiple bubbles distributions fall also under the single bubble distribution. In other words, we want to know how many alpha like events could have the same acoustic parameter as the neutron events, therefore creating an undistinguishable source of background.

To do so, we set an acceptance cut at 96% for the neutron like events distributions (corresponding to a range $[-\infty : 1.75 \sigma]$ for a Gaussian probability density function), as obtained in the 4kg chamber of the COUPP experiment [35], and we determine what fraction $F_p(1, i)$ of the events with a number $i$ of bubbles exceed the acceptance cut and could be mislabeled as single bubble events, as shown in Fig. 4.4.
4.1. A 2D ACOUSTIC MODEL FOR PICO

Figure 4.4: The blue Gaussian curve represents the distribution of single bubble events, where the area under the curve corresponding to the 96 % of the events (a range \([-\infty : 1.75\sigma]\]) has been colored using a lighter blue. The area under the red Gaussian curve (the distribution of events with \(i\) bubbles) that overlaps with the highlighted area of the blue Gaussian determines \(F_p(1,i)\).

Table 4.2: Results for the fraction \(F_p(1,i)\) of events with a number \(i\) of explosions that overlap with the range \([-\infty : 1.75\sigma]\) of the single bubble Gaussian distributions. Results are shown for the top, middle and bottom piezo.

<table>
<thead>
<tr>
<th></th>
<th>(F_p(1,2))</th>
<th>(F_p(1,3))</th>
<th>(F_p(1,4))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Top piezo</td>
<td>0 %</td>
<td>0 %</td>
<td>0 %</td>
</tr>
<tr>
<td>Middle piezo</td>
<td>26.5 %</td>
<td>1.7 %</td>
<td>0.1 %</td>
</tr>
<tr>
<td>Bottom piezo</td>
<td>13.0 %</td>
<td>0.3 %</td>
<td>0 %</td>
</tr>
</tbody>
</table>

It is evident from these results that the top piezo offers the best separation between the distributions, and therefore the best discrimination capabilities, while the bottom
and middle provide worse and similar results.

We can explain the better performance of the top piezo by observing that, in the top part of the vessel, the acoustic signal is not reflected between the walls as intensely as it happens in the middle and bottom parts. In fact, acoustic waves can escape the vessel from the top part, limiting the number of reflections that are detected by the top piezo. Moreover, in this model, acoustic explosions are only located in the middle and bottom part of the vessel, so that sound waves reach the wall in the top part with an angle that allows them to easily escape after only a few reflections.

Table 4.2 shows that, for the middle and bottom piezos, the two-bubble event distributions show a considerable overlap with the single-bubble event distributions, while the three-bubble event distributions show only a tiny overlap that becomes completely negligible when we consider the four-bubble events. These results are clearly compatible with the discrimination capabilities shown in the superheated liquid experiments (see Fig. 2.9).
Chapter 5

Conclusion

In the first part of this work, I have investigated the possibility of using the buffer liquid of the PICO-250L detector, i.e. linear alkylbenzene, as a liquid scintillator to help assess the levels of radioactivity inside the fused quartz inner vessel. Given the extensive studies on the scintillation properties of this solvent mixed with fluors, I have focused on the specific questions that have to be answered in order to implement this potentiality in the PICO experiment.

At first, I have experimentally verified that LAB mixed with PPO retains its scintillation properties when interfaced with the active fluid $C_3F_8$. This test has also demonstrated that the scintillation mechanism in the cocktail does not show any degradation over a month, as it is necessary to assure that the scintillator efficiently performs during the long timescale of the experiment. Moreover, tests carried out by the PICO collaboration have demonstrated that the superheated properties of the active fluid $C_3F_8$ are not affected by the presence of LAB as a buffer liquid, providing the complementary information for the use of this combination of active and buffer fluids.

I have also discovered that LAB merges with the secondary active fluid $CF_3I$,
which eliminates the possibility of using this combination of buffer and active fluids.

Potential interactions between the LS and the radioactive particles that constitutes the background of superheated experiments, such as neutrons and alphas, have been identified and the scintillation light yielded by these interactions has been assessed. In particular, I have considered the intensity and likelihood of gamma rays produced by neutron capture in $^{12}C$ and $^{19}F$.

Research carried out by reactor antineutrino experiments, has proven that gadolinium, the element with the highest cross section for neutron capture, can be added in a clean way to a LAB based cocktail to generate a stable liquid scintillator. The research started in this work suggests to explore the possibility of exploiting a gadolinium-loaded LAB scintillator in the PICO detector.

To complete the study of a liquid scintillator in PICO, I have investigated several methods to collect the light produced by scintillation in the buffer liquid. Identifying photomultiplier tubes with 4 different effective diameters (190, 111, 46 and 25 mm) as the best candidates for the optical readout, I have designed two different types of concentrators that would increase the amount of collected light. The efficiency of these designs is determined through a comparison to the model where light is not guided to the PMTs.

Acrylic conical concentrators coated with aluminium guarantee high transparency and high reflectivity for the wavelengths emitted by the LAB+PPO+mis-MSB cocktail, in addition to increasing the amount of light collected by a factor of 6. Alternatively, CPC concentrators offer a comparable improvement in light collection, while requiring a much smaller volume of materials that scales down with the PMT’s size.

Future research on this topic could focus on building and testing a bubble chamber
where LAB is to be used as buffer fluid and liquid scintillator, in order to deal with
the experimental challenges presented by the construction of concentrators and the
mounting of light guides on the vessel, testing if it’s possible to achieve the results
obtained in this work.

In the second part of this work, I have developed a 2D acoustic model of the inner
vessel of the PICO-250L detector, using the COMSOL simulation software. Super-
heated experiments observe acoustic traces with higher intensity for events caused by
alpha particles than for those triggered by neutrons’ interactions. This difference is
explained by the fact that alpha particles deposit their energy on an extremely short
track where they trigger the formation of multiple bubbles, while recoils from neutron
interactions deposit energy in a very short track which only allows for the formation
of one bubble per event.

While being limited to a smaller, scaled version of the inner vessel, the model
has demonstrated that it is possible to reproduce this fundamental discrimination
technique, exploiting this difference in the physics of alpha and neutron interactions,
and confirming the connection between multiple-bubble events and signals with high
acoustic energy.

A model that can replicate this discrimination technique constitutes a reliable
framework with which to build simulations that can guide the design of the PICO-
250L detector. Following this reasoning, I have investigated how this discrimination
capability is affected by the choice of the position of the piezoelectric devices around
the inner vessel. I have discovered that piezoelectric devices located around the buffer
liquid generate an excellent discrimination between single and multiple-bubble events,
while a reduced discrimination is obtained by devices located around the active fluid.
Research that could follow from this work could aim at building a larger and more precise model that is efficient from a computational point of view. Given the importance of reflections of the acoustic signal in the vessel, and the increased complexity of the reflections behavior when going from two to three dimensions, the development of 3D model with an affordable number of degrees of freedom could be tackled by future projects. Moreover, an experimental setup could be built in order to obtain results that are comparable with the simulations’ outputs.
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Appendix A

A measurement of the speed of sound in LAB

A.1 Thermophysical properties of LAB

As previously discussed, LAB was the liquid scintillator of choice for neutrino experiments because of its thermophysical properties such as high flash point (147 °C) and boiling temperature (280-311 °C) [62], in addition to its high concentration of hydrogen atoms (6.29 × 10^{22} \text{cm}^{-3}) [50], its chemical compatibility with acrylic polymers, being environmentally safe and its optical properties.

However, the use of LAB as a hydraulic fluid in the PICO detectors requires an understanding of its properties as a fluid power medium, such as viscosity and fluid compressibility.

Values for thermophysical properties of LAB that are relevant to its use as hydraulic fluid as summarized in Table A.1.
A.2. A MEASUREMENT OF THE SPEED OF SOUND IN LAB

Table A.1: Summary of thermophysical properties of linear alkylbenzene at $p = 760$ mmHg [63].

<table>
<thead>
<tr>
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<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>10.0</td>
<td>862.04</td>
<td>7.183</td>
<td>32.36</td>
<td>1.69</td>
<td>0.130</td>
</tr>
<tr>
<td>20.0</td>
<td>854.37</td>
<td>5.446</td>
<td>31.46</td>
<td>1.75</td>
<td>0.128</td>
</tr>
<tr>
<td>30.0</td>
<td>845.80</td>
<td>4.280</td>
<td>30.57</td>
<td>1.80</td>
<td>0.127</td>
</tr>
<tr>
<td>40.0</td>
<td>836.54</td>
<td>3.465</td>
<td>29.68</td>
<td>1.85</td>
<td>0.125</td>
</tr>
</tbody>
</table>

One important property missing in the above table is the bulk modulus $K$ which provides information on the resistance that the fluid exerts as a reaction to compression.

As postulated by Newton in his Principia, published in 1687, the bulk modulus is related to density $\rho$ and speed of sound $c$ through the formula:

$$K = c^2 \rho \tag{A.1}$$

therefore a measurement of the speed of sound in LAB, combined with values of density would yield an indirect measurement of the bulk modulus.

A.2 A measurement of the speed of sound in LAB

A.2.1 Experimental setup and method

Piezoelectric transducers, can be used to transform a mechanical stress (caused by an acoustic wave, for instance) into a voltage that can be then read and analyzed, or inversely to generate an acoustic signal by applying a variable electrical field which
yields a deformation of the material, using respectively the direct and indirect piezo-
electric effect.

In this measurement, two piezoelectric devices similar to those used in the PI-
CASSO experiment have been used, i.e. two ceramic disks (Ferroperm PZ27) with a
diameter of 16 mm and 8.7 mm thickness and a pressure sensitivity of 27 µV/µbar.

The first piezo was located at the bottom surface of a cylindrical container, which
was then filled with the fluid that is to be studied. The second piezo was lowered in
the cylindrical container, its vertical position could easily be adjusted and measured
with a precision of 1 mm. It was used to produce an acoustic signal, produced by a
voltage pulse applied through a function generator (Agilent 33220A), that propagated
in the fluid and which was then received at the bottom piezo.

Since the goal of this measurement was to determine the speed of sound in LAB,
time measurements related to distance measurements were required.

The advantage of this experimental setup (Fig. A.1) is that only relative mea-
surements of time and distance are needed. In fact, measuring the time difference $\Delta t$
between emitting and receiving time for an acoustic signal traveling in the fluid, for a
set of vertical displacements $\Delta y$ of the top piezo from its original position $l_0$, a slope
can be determined from a displacement vs time difference graph, whose inverse then
yields a value for the speed of sound.

Expressed with formulas, time relates to displacement as:

$$\Delta t = \frac{l_0 - \Delta y}{c} + C_1$$  \hspace{1cm} (A.2)

where $C_1$ is a time offset introduced by instruments and cables’ lengths. Then Eq.
A.2. A MEASUREMENT OF THE SPEED OF SOUND IN LAB

\[ \Delta t = -\frac{\Delta y}{c} + C_1 + C_2 \]  

where \( C_2 = \frac{l_0}{c} \).

Time measurements are given by a Time-to-Amplitude-Converter, or TAC (ORTEC 566), which calculates the time difference between a start signal, produced by the function generator, which is simultaneously fed to the top piezo, and a stop signal which comes from the reception of the start signal at the bottom piezo.

For a precise and coherent measurement of the time differences, the stop signal is generated when the bottom piezo output, magnified by an ORTEC 572A amplifier, exceeds a voltage threshold set by a time threshold discriminator (LRS 623).

The output signal of the TAC is fed to a Multi-Channel-Analyzer, or MCA (ORTEC 920E), which sorts the time difference signals in bins, constructing a distribution that provides a statistical measurement of the time difference needed.

### A.2.2 Results

In order to calibrate the experimental setup, and in particular the TAC time conversion, I first measured the speed of sound in tap water. The results are shown in Fig. A.2, where the y-coordinate of each point refers to time difference between start and stop signal fed to the TAC and the x-coordinate to the vertical displacement of the top piezo from its initial position. Error bars are too small to be visible in this graph.

A linear regression has been used to fit the data with a dedicated MATLAB function and get a numerical value for the slope of the line and its uncertainty.
The value obtained for the speed of sound in water at \( T = 23.3^\circ C \) is:

\[
c_w,\text{mea} \pm \Delta c_w,\text{mea} = (1473.9 \pm 6.0) \text{ m/s} \tag{A.4}
\]

To calibrate the experimental setup, the measured value for the speed of sound in water \( c_{\text{mea}} \) has been compared with accepted value \( c_{\text{acc}} \) in the literature \[64\], in order to obtain a calibration factor \( F \) that will then be applied to the measurements for LAB, as:

\[
F = \frac{c_{w,\text{acc}}(T = 23.3^\circ C)}{c_{w,\text{mea}}} = \frac{1492.0 \text{ m/s}}{1473.9 \text{ m/s}} = 1.0123 \tag{A.5}
\]

The value obtained for the speed of sound in LAB at \( T = 23.7^\circ C \) (Fig. A.3) is \( c_{l,\text{mea}} \pm \Delta c_{l,\text{mea}} = (1354.7 \pm 5.5) \text{ m/s} \), while using the calibration factor we obtain:

\[
c_{l,\text{cal}} \pm \Delta c_{l,\text{cal}} = (1371.4 \pm 11.1) \text{ m/s} \tag{A.6}
\]
Figure A.2: Results for the speed of sound in water measurement, at $T = 23.3^\circ C$.

where

$$\frac{\Delta c_{l,\text{cal}}}{c_{l,\text{cal}}} = \frac{\Delta F}{F} + \frac{\Delta c_{l,\text{mea}}}{c_{l,\text{mea}}}$$

(A.7)

Consequently, for the bulk modulus $K$ of LAB, Eq. (A.1) yields:

$$K \pm \Delta K = (1.60 \pm 0.03) \text{ GPa}$$

(A.8)

where

$$\frac{\Delta K}{K} = 2 \cdot \frac{\Delta c_{l,\text{cal}}}{c_{l,\text{cal}}} + \frac{\Delta \rho}{\rho}$$

(A.9)

since an error on the density value was not provided.
Figure A.3: Results for the speed of sound in LAB measurement, at $T = 23.7^\circ$C.