

15<sup>TH</sup> TOPICAL SEMINAR ON INNOVATIVE PARTICLE AND RADIATION DETECTORS  
14–17 OCTOBER 2019  
SIENA, ITALY

## Comparison between photon detection efficiency and tetraphenyl-butadiene coating stability of photomultiplier tubes immersed in liquid argon

B. Burak,<sup>a</sup> P. Garcia,<sup>b</sup> C. Jesús-Valls,<sup>b</sup> T. Lux,<sup>b</sup> A. Menegolli,<sup>c,d</sup> M.C. Prata,<sup>c,d</sup> G.L. Raselli,<sup>c,d</sup> M. Rossella,<sup>c,d</sup> D. Vargas<sup>b,1</sup> and W. Vollenberg<sup>a</sup>

<sup>a</sup>European Organization for Nuclear Research (CERN), Geneva, Switzerland

<sup>b</sup>Institut de Física d'Altes Energies (IFAE), Barcelona, Spain

<sup>c</sup>Istituto Nazionale di Fisica Nucleare (INFN), Pavia, Italy

<sup>d</sup>University of Pavia, Pavia, Italy

E-mail: [dvargas@ifae.es](mailto:dvargas@ifae.es)

**ABSTRACT:** Liquid argon detectors are an interesting option for neutrino experiments. The high density of liquid argon allows using it directly as target for neutrino interactions. The relatively large abundance of argon in the atmosphere makes it a cost-effective medium, allowing the construction of detectors of several hundred tons, such as ICARUS, and even several kton detectors are foreseen for the next generation of experiments, such as DUNE. Besides being suitable to act as the target, liquid argon also serves as the detection medium for the charged particles coming out of the interaction vertex. The interaction time,  $t_0$ , can be determined by detecting the photons which are produced together with the electrons in the ionization process. To detect them, the integration of an efficient photon detection system into the liquid argon detector is necessary. One difficulty here is related to the fact that the emitted photons have a wavelength in the VUV range (128 nm). The classical approach for the photon detection system is, therefore, the usage of photomultiplier tubes coated with tetraphenyl-butadiene which shifts the VUV photons to about 430 nm, a wavelength to which the photomultiplier tubes are directly sensitive to. While the basic concept is well established and has been used in several experiments such as ICARUS, ArDM and DarkSide, some aspects of the coating are not well understood such as the preparation of the photomultiplier tube surface which can be either polished or sandblasted. To better understand the effect on the overall photon detection efficiency on one hand and the tetraphenyl-butadiene stability during the cooling down, on the other hand, the quantum efficiencies of sandblasted and polished photomultiplier tubes,

<sup>1</sup>Corresponding author.

coated following the same procedure, were measured with a setup at the Istituto Nazionale di Fisica Nucleare (INFN), in Pavia. In addition, the immersion of the photomultiplier tubes with different soak timings was tested, showing a significantly different behavior for fast and slow immersion for the polished photomultiplier tubes.

**KEYWORDS:** Large detector systems for particle and astroparticle physics; Neutrino detectors; Photon detectors for UV, visible and IR photons (gas); Time projection chambers

---

## Contents

|          |   |          |
|----------|---|----------|
| <b>1</b> | <b>Introduction</b>                                     | <b>1</b> |
| <b>2</b> | <b>Coating setup and procedure</b>                      | <b>3</b> |
| <b>3</b> | <b>Quantum efficiency of the VUV measurement system</b> | <b>5</b> |
| <b>4</b> | <b>Test of the coating resistance</b>                   | <b>7</b> |
| <b>5</b> | <b>Conclusion</b>                                       | <b>8</b> |

---

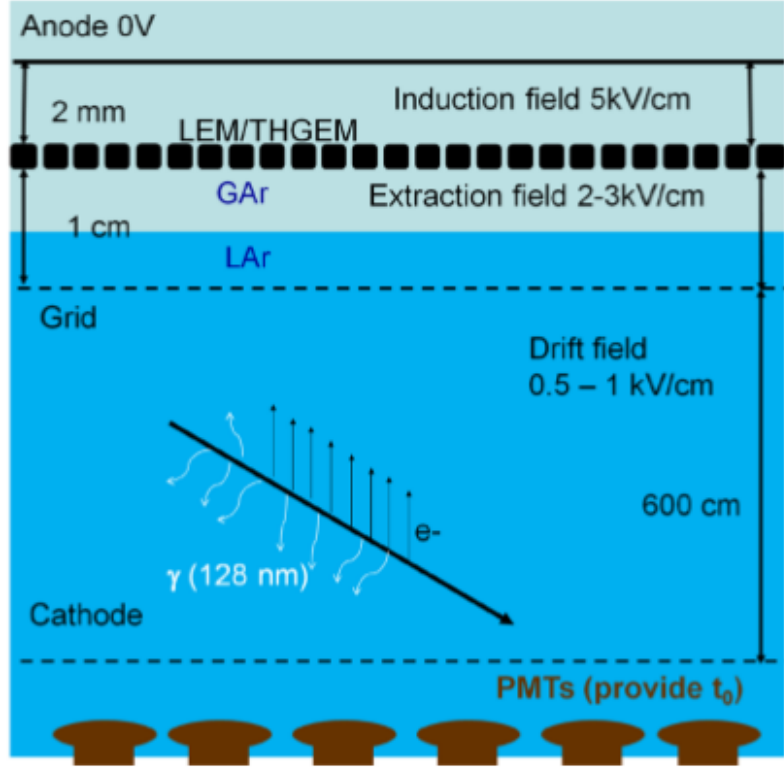
## 1 Introduction

The Deep Underground Neutrino Experiment (DUNE) [1] is the next generation of long baseline neutrino oscillation experiments. A wide band neutrino beam is produced at Fermilab and directed towards the Sanford Underground Research Facility (SURF), located 1300 km away where the far detectors of the experiment will be installed at a depth of around 1.5 km to reduce the cosmic ray background.

Liquid argon time projection chambers (LArTPC) have been chosen as the detector concept for the four far detectors, each with a fiducial mass of 10 kton. LArTPCs exist in two realisations: single and dual phase. In the single phase design, the anode is made up of different sets of wires that are immersed in the liquid on which the electrons create a signal, either due to induction or direct absorption. In the dual phase design the electrons are extracted from the liquid to a gaseous volume within a strong electric field of 2 to 3 kV/cm and are amplified in a Townsend avalanche afterwards, before the signal is read out on a segmented anode [2]. The dual phase concept has several advantages: it has excellent tracking and calorimetry capabilities, and due to the amplification in the gas phase the losses of electrons, that appear as a consequence of the oxygen impurities, can be partly compensated for, allowing a lower energy detection thresholds.

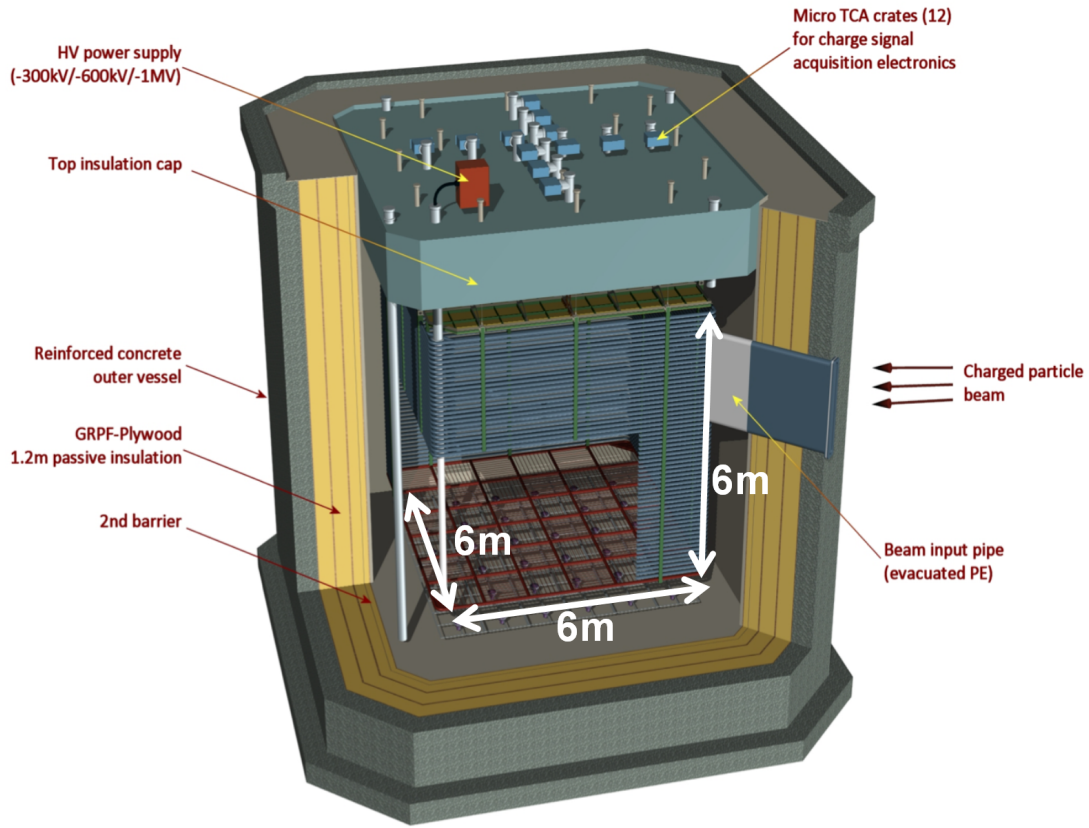
The basic principle of a LArTPC is shown in figure 1. A charged particle traversing the LAr ionizes the argon atoms creating a large amount of electron-ion pairs on its way through the liquid. By applying an external electric field (the drift field) the electrons and ions are separated and drift toward the anode and cathode respectively. Drift lengths up to several meters are feasible, corresponding to drift times of a few milliseconds. During this drift, the population of electrons is attenuated before they are extracted in the gas phase due to attachment to impurities. A precise measurement of the initial time  $t_0$  is required in order to correct for this effect, reconstructing the original charge as the measured charge weighted by a factor that can be computed knowing the attenuation length, the electrons drift length, and the drift velocity. Large Electron Multipliers (LEMs) provide amplification before the charge collection on an anode plane with strip readout. The primary scintillation light (S1) production for  $t_0$ + trigger is detected by an array of photomultiplier

tubes (PMTs). The interaction time,  $t_0$ , can be determined by detecting the photons produced together with the electrons in the ionization process. These photons provide a prompt signal on the nanosecond scale.



**Figure 1.** Basic principle of the dual phase LArTPC [2].

WA105/protoDUNE-DP is a large prototype of the dual-phase LArTPC (figure 2) which was constructed at the Neutrino Platform Hall at CERN (Preessin) in 2018 and commissioned in the second half of 2019. The inner volume has dimensions of  $8.3 \times 8.3 \times 8.1 \text{ m}^3$  ( $W \times L \times H$ ) containing about 700 tons of LAr. The fiducial volume has dimensions of  $6 \times 6 \times 6 \text{ m}^3$  and contains about 300 tons of active mass. Due to the large drift distance of several meters, a voltage of up to 300 kV is required at the cathode, which consists of steel tubes to achieve a high transparency for the photons of the primary scintillation light [3]. These photons provide a prompt signal on the nanosecond scale. To detect them, the integration of an efficient photon detection system (PDS) into the LAr detector is necessary, therefore a set of 36 PMTs (Hamamatsu R5912-02mod) are installed about 1 m below the cathode for the detection of this primary scintillation light. The PMTs used have a polished convex surface with a diameter of 8 inches, 14 linearly focused dynode stages, a narrow spread in transit time and a gain of up to  $1 \times 10^9$ . One difficulty here is related to the fact that the emitted photons have a wavelength in the VUV range (128 nm). The classical approach for the PDS is, therefore, the usage of PMTs coated with tetraphenyl-butadiene (TPB) as a wavelength shifter (WLS) shifting the VUV photons to about 430 nm, a wavelength to which the PMTs are directly sensitive to.

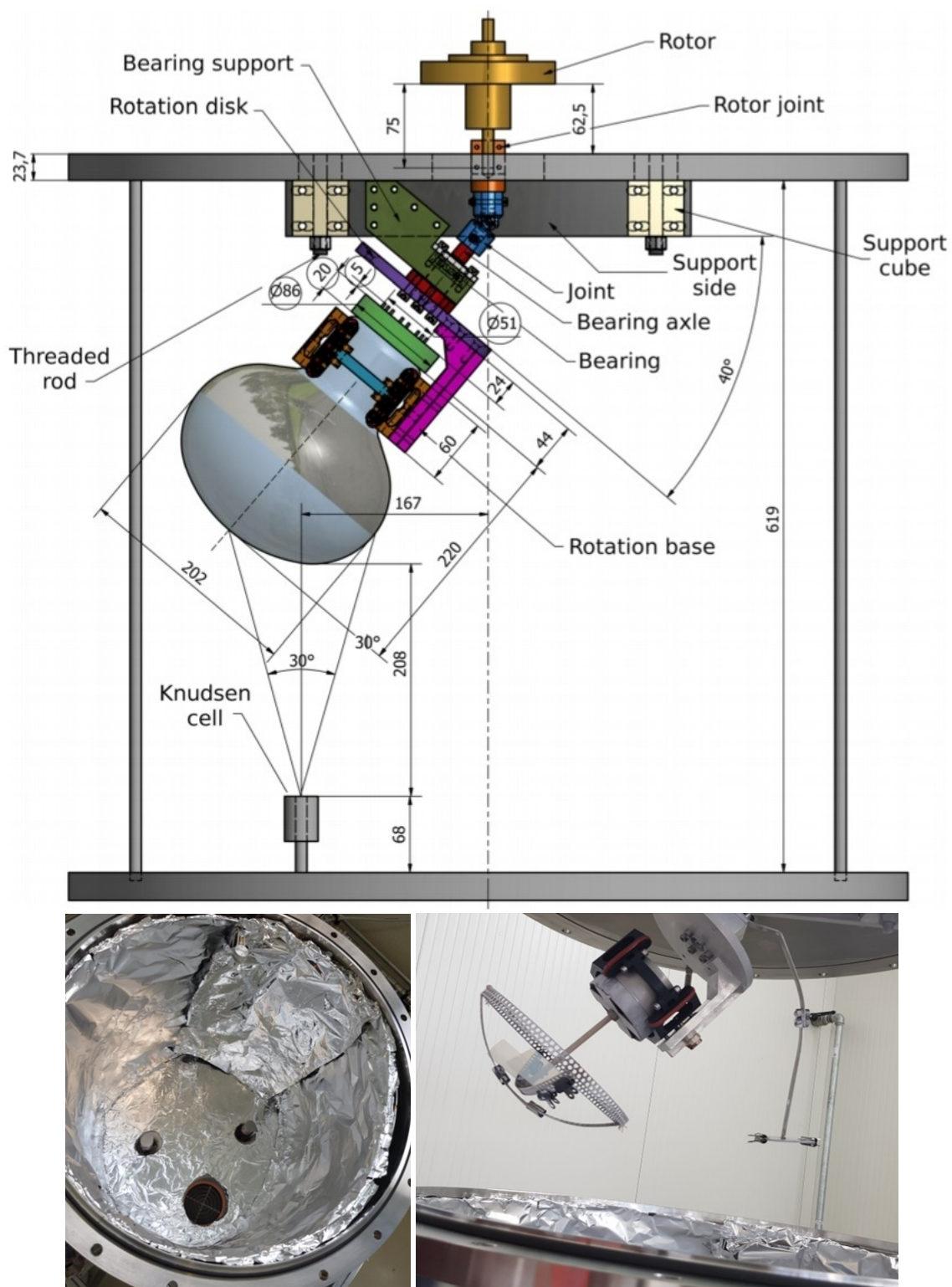


**Figure 2.** Schematic view of the WA105/protoDUNE-DP detector. The sensitive volume has dimensions of  $6 \times 6 \times 6 \text{ m}^3$  and contains about 300 ton of LAr [2].

## 2 Coating setup and procedure

The thermal evaporation system, shown in figure 3, used for this study was originally designed for the coating of the convex surfaces for the 360 PMTs deployed in the ICARUS T600 detector. The system consists of a motorized rotating feedthrough on the top of a vacuum chamber, in which the PMT is mounted at an angle of  $40^\circ$  with respect to the cover. A Knudsen cell (effusion evaporator source) is placed below the PMT, in which the WLS must be deposited in for the evaporation in vacuum. The angle and the distance from the Knudsen cell guarantee a geometrical uniformity of the expected evaporation process between center and border of the PMT surface [4].

The coating system was tested and calibrated and the most important parameters of the process have been evaluated by evaporating TPB over a PMT mock-up (figure 3 bottom right), on which small square Mylar foils were fixed at different positions. The uniformity of the evaporation over the entire convex photocathode is measured by weighting each sample before and after the process, with a high precision scale. Before the coating, the PMTs were cleaned by pouring acetone first and then alcohol over their surface and dried with two clean room tissues to ensure that the surface is completely dry. For the next step in the process, the PMT was placed in the support and the Knudsen cell on the bottom of the vacuum chamber was filled with about 0.8 g of TPB.



**Figure 3.** Schematic overview of the evaporation system [4] (top), a photo of the top view of the vacuum chamber (bottom left) and a photograph of the mock-up mounted on the feedthrough (bottom right).



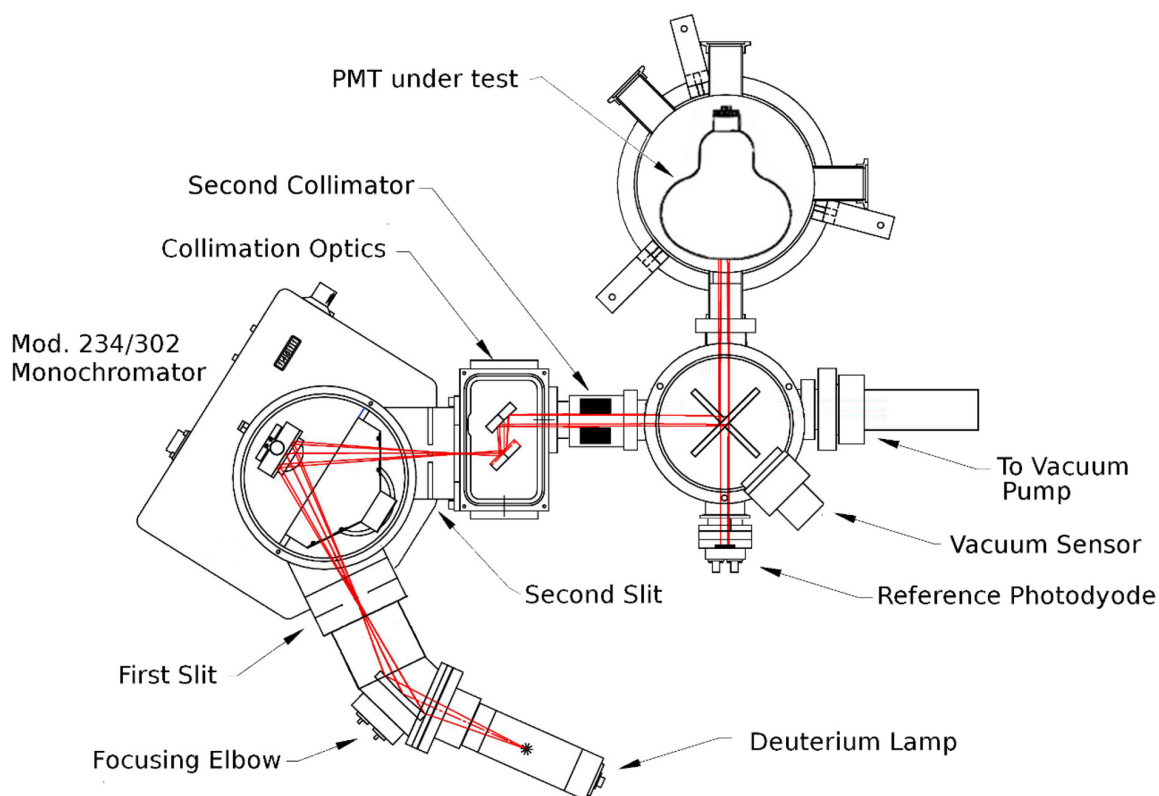
When the pressure inside the vacuum chamber is below  $3 \times 10^{-5}$  mbar, the motor is started, rotating at 10 turns/min, and the heater of the Knudsen cell is switched on with the temperature regulator set to 220 °C. When the temperature reaches 190 °C, the shutter over the Knudsen cell is opened, allowing the material to evaporate and deposit on the PMT surface. During the evaporation, a Sycon STM-100/MF Thickness/Rate Monitor allows to monitor the process. It uses an oscillating quartz crystal as the sensor device. The crystal, located inside the chamber beside the PMT, is externally connected to a Sycon OSC-100A Oscillator. The monitor displays the deposition thickness value both per unit of time (Å/s) and integrated over the whole process (kÅ) [4]. The procedure takes about 3 hours per PMT. After the coating is finished, a visual inspection with a commercial UV lamp (figure 4) was carried out for all PMTs as the last step of the process.

### 3 Quantum efficiency of the VUV measurement system

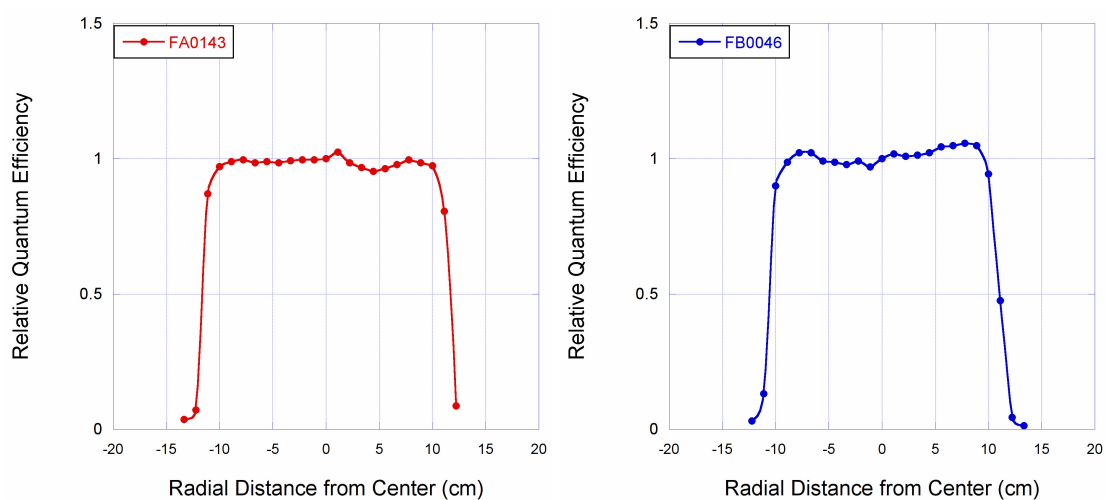
The optical characterization of the detector after the coating process was completed by means of a VUV measurement system developed at the INFN Pavia (Italy) [4]. It allows the characterization of detectors in terms of absolute quantum efficiency ( $Q_{\text{eff}}$ ) in the VUV range (figure 5). It consists of a vacuum chamber where the light produced by a continuous spectrum deuterium lamp is separated into different wavelengths by a MacPherson 234 monochromator and then directed alternatively toward a National Institute of Standards and Technology (NIST) calibrated reference photodiode or toward the device under test. A comparison between the current delivered from the two devices, measured with a Keithley picoammeter and corrected by the photodiode  $Q_{\text{eff}}$  given by NIST, provides the absolute  $Q_{\text{eff}}$  of the device under test. The available wavelength ranges from 120 to 250 nm with a maximum resolution of 1 nm. The mounting of the device on a rotating feedthrough allows the measurements of the  $Q_{\text{eff}}$  in various positions of the photocathode [4].



**Figure 4.** Photo of the visual inspection with a commercial UV lamp.



**Figure 5.** Schematic description of the VUV measurement system developed at the INFN Pavia (Italy) [4].



**Figure 6.** Relative quantum efficiency for different PMTs as a function of the radial distance from the center for a polished (left) and sandblasted (right) surface.



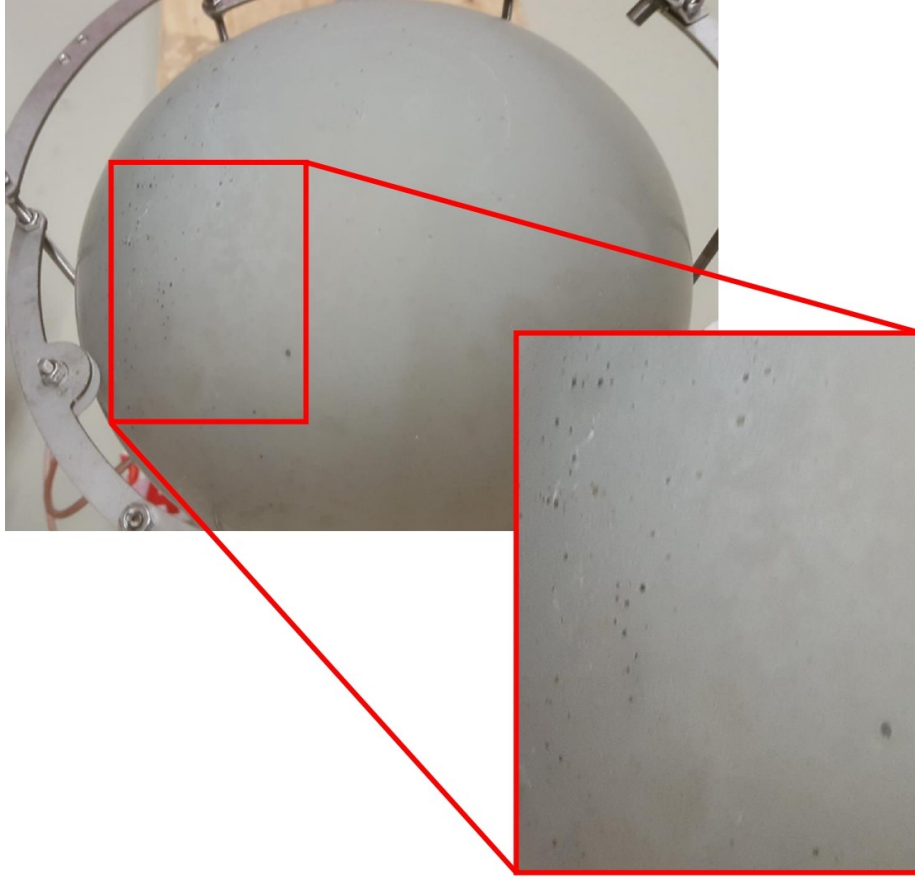
A sample of PMTs was characterized in terms of quantum efficiency at  $\lambda = 128$  nm. The distribution of the relative  $Q_{\text{eff}}$  is shown in figure 6. The study was done for 4 PMTs with polished surface (randomly chosen from 40 coated PMTs) and the results were compared to the one reported by ICARUS for PMTs with sandblasted surface. The obtained values are distributed in the 10% to 20% range, with an average value of  $0.14 \pm 0.02$  and  $0.12 \pm 0.01$  for polished and sandblasted PMTs respectively. Fluctuations are mainly due to different sensitivity of the bialkali photocathode on different devices [4].



**Figure 7.** The two types of surface used for the comparison were: sandblasted PMT (left) and polished PMT (right).

#### 4 Test of the coating resistance

We also performed some preliminary resistance tests for the coatings using the spare PMTs. A PMT including its base and support structure, which were designed to avoid the floating of the PMT in LAr, was placed on the bottom of a small open dewar. The dewar was then filled with LAr through a tube which was placed such that the LAr entered the Dewar at the bottom, close to the dewar wall to avoid that the LAr flows directly across the surface of the PMTs which were not cooled down before the filling. The filling was then performed quickly and the PMT was immersed completely in LAr within less than 1 hour. Since the Dewar and the PMT were not cooled the LAr started to boil, leading to drops of LAr falling on the TPB coating, mainly on the side of the PMT close to the tube. The PMT coating was inspected with a UV lamp directly after the coating process and after the immersion test. As is visible in the figure 8, after the immersion test small spots on the surface appeared, where the TPB was almost completely removed from the PMT surface. Further tests with even harsher immersion procedures with polished and sandblasted PMTs showed that the almost complete removal of TPB is restricted to the PMTs with polished surface. Immersing a polished PMT into LAr with soaking times close to the one in large detectors shows no indications of damage to the TPB coatings.



**Figure 8.** Coated PMT with small holes in the coating of the PMT after the test.

Further systematic tests with sandblasted PMTs which are more resistant to thermal shocks seem to be recommendable since the procedure to cool down the cryostats of future larger LArTPCs includes the usage of sprayers letting LAr rain down in the cryostat, and thus it might affect the design of these detectors.

## 5 Conclusion

For the next generation of neutrino oscillation experiments, detectors with about 10 ktons active mass will be constructed. The WA105/protoDUNE-DP detector is a necessary intermediate step for these detectors to prove their technical feasibility and will also provide a deeper understanding of the detector performance. The evaporation system guarantees a uniform evaporation of the wavelength shifter over the PMT's photocathode window, granting a good  $Q_{\text{eff}}$  even at the photocathode edge. For the WA105/protoDUNE-DP detector 40 PMTs were coated successfully. The performance of the coating was tested at INFN to study the impact of the PMT surface treatments. The  $Q_{\text{eff}}$  at  $\lambda = 128 \text{ nm}$  was measured for four polished PMTs, used in WA105/protoDUNE-DP, to be  $0.14 \pm 0.02$  while for the sandblasted surface a value of  $0.12 \pm 0.01$  is found, indicating that from the  $Q_{\text{eff}}$  point of view both surface treatments are suitable for high energy experiments. The coating of sandblasted PMTs are less sensitive to thermal shocks. The photon detection system of protoDUNE-DP was successfully commissioned.

## Acknowledgments

The author would like to thank the CERN Neutrino Platform for their help. We also acknowledge the support received from the Spanish Ministerio de Economía y Competitividad (SEIDI-MINECO) under grants FPA2016-77347-C2-2-P, TEC2012-39150-C02-02 and Centro de Excelencia Severo Ochoa SEV-2016-0588, some of which include ERDF funds from the European Union.

## References

- [1] DUNE collaboration, *Long-Baseline Neutrino Facility (LBNF) and Deep Underground Neutrino Experiment (DUNE)*, FERMILAB-DESIGN-2016-01 (2015) [[arXiv:1512.06148](#)].
- [2] WA105 collaboration, *WA105: A large LAr double phase TPC prototype*, *EPJ Web Conf.* **174** (2018) 01009.
- [3] WA105 collaboration, *WA105: A large demonstrator of a liquid argon dual phase TPC*, *J. Phys. Conf. Ser.* **888** (2017) 012202.
- [4] M. Bonesini et al., *Realization of a high vacuum evaporation system for wave-length shifter deposition on photo-detector windows*, *J. Vac. Sci. Tech.* **B 36** (2018) 01A101.
- [5] I. De Bonis et al., *LBNO-DEMO: Large-scale neutrino detector demonstrators for phased performance assessment in view of a long-baseline oscillation experiment*, SPSC-TDR-004 (2014) [[arXiv:1409.4405](#)].
- [6] C. Cantini et al., *Long-term operation of a double phase LAr LEM Time Projection Chamber with a simplified anode and extraction-grid design*, *2014 JINST* **9** P03017 [[arXiv:1312.6487](#)].
- [7] K. Mavrokoridis et al., *Optical Readout of a Two Phase Liquid Argon TPC using CCD Camera and THGEMs*, *2014 JINST* **9** P02006 [[arXiv:1401.0525](#)].