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APPLICATION OF ELECTROLUMINESCENCE IN A PROTOTYPE FOR THE MEASUREMENT OF A LIQUID ARGON SCINTILLATIONS

Sergei Pakhomov, Tatiana Kuzmina, Ekaterina Kurysheva, Gennady Shakhetov

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INTRODUCTION AND MAIN RESULTS

Development of the equipment for measuring of low levels argon-37 activities in soil and atmospheric air remains a priority task for improving nuclear test monitoring technologies for OSI and a promising direction for IMS.

The use of the electroluminescence effect in the prototype of the installation for the argon-37 measurements, based on the registration of liquid argon scintillations, allows for a significant improvement in its detection characteristics.

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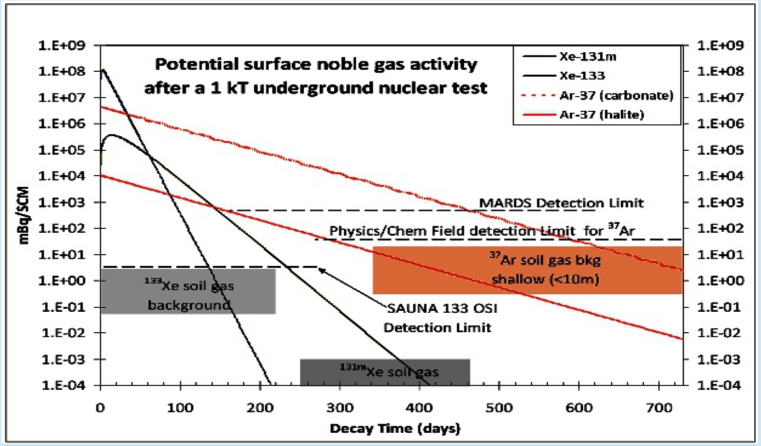
Relevance of ^{37}Ar measurements



Argon-37 and radioxenon levels expected at the surface after a 1 kT nuclear explosion ***/D.A. Haas, H.S. Miley, J.L. Orrelle et al. The Science Case for 37Ar as a Monitor for Underground Nuclear Explosions. Report PNNL-19458/.***

One of the key evidence of a violation of the CTBT is a detection in the subsoil air enlarged concentrations of ^{37}Ar which is formed owing to interaction of a nuclear explosion neutrons with soil calcium: ^{40}Ca (n, α)^{37}Ar.

Concentrations of argon-37 retains over-beackground levels in subsoil air for much longer time than xenon radionuclides, so it is the "ideal witness" of a nuclear test.

Therefore the development of the equipment for measuring of low levels argon-37 in air argon samples remains a priority task for improving nuclear test monitoring technologies for OSI and a promising direction of IMS elaboration.

Development of prototype for ^{37}Ar measurements

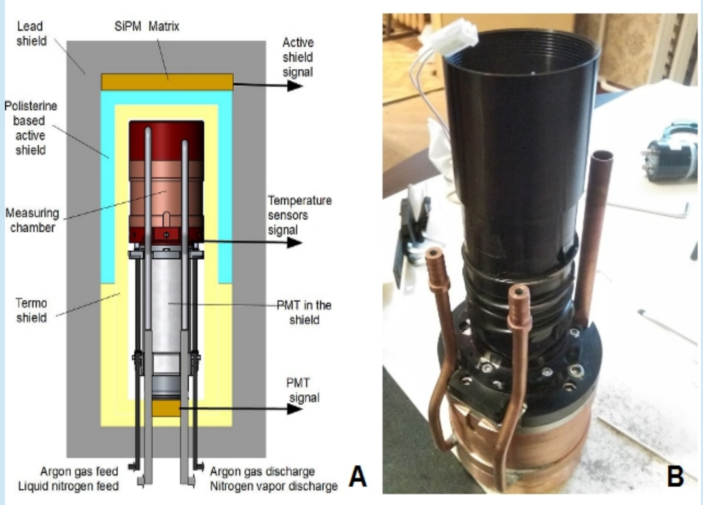
Traditionally, ^{37}Ar measurements are performed using low-background gas proportional counters. However, the capacity of these counters does not exceed several liters of argon, which limits the volume of an equivalent air sample to several hundred liters.

It is possible to significantly increase the sample volume by converting argon into a liquid state. Due to the good scintillation properties of liquefied noble gases, including argon, it is possible to measure the activity of ^{37}Ar in a sample by registering the scintillation of liquid argon itself.

The Radium Institute took the initiative to develop such a device and, having received support from the CTBTO, developed and manufactured a prototype for measuring scintillations in liquid argon caused by decays of ^{37}Ar contained in the atmospheric air.

A-General view of developed device, B-Detecting unit in a passive shield, C Ramp with argon cylinders, D-dosing system.

Detecting unit of installation prototype



A- Detecting unit in active, thermal and lead shield.
B- General view of the detecting unit.

A sample of ^{37}Ar in argon gas is fed into detecting unit where it is liquefied in a 100 cm^3 measuring chamber, which is equivalent to 78 liters of argon gas or 8,3 m^3 of atmospheric air.

Liquid argon scintillations are registered by a Hamamatsu R331-5 photomultiplier, and signals are processed by a CAEN DT5790 12-bit signal digitizer.

Testing of the prototype showed the functional operability of the device, but revealed low efficiency of ^{37}Ar decays registration, deficient for measurements at the background level.

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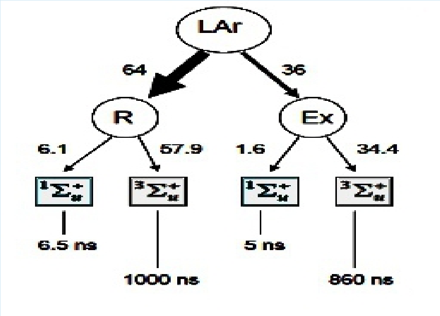
Features of ^{37}Ar scintillation

Scintillations in liquid argon are formed in interaction with ionizing radiation either as a result of ionization of the atom (R) and as a result of its excitation (Ex):



The mechanisms of argon scintillation

Thus, scintillations consist of a fast (1/4) and a slow (3/4) components, the slow component is subject to quenching due to the presence of impurities and as a result of ionization quenching.

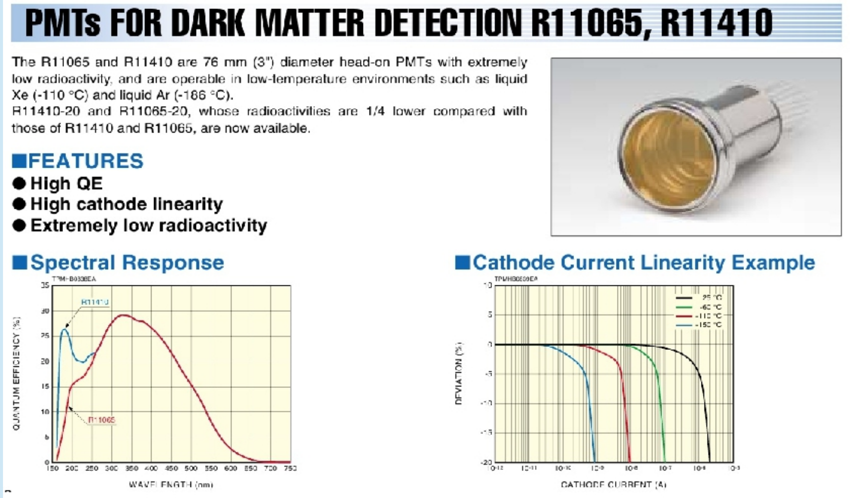
For high-energy electrons with low ionization capacity, the scintillation yield in LAr reaches 40 photons/keV, but when registering ^{37}Ar Auger electrons with low energy of 2.8 keV, the light yield decreases significantly and reaches only dozen photons/keV, or about 40 photons per decay event. ***/Sergei A. Pakhomov. Prototype of laboratory system for detecting of argon-37 scintillations in liquefied samples of atmospheric argon. INGE Workshop 2017./.***

For comparison, when measuring tritium using a liquid scintillator, the average scintillation yield is also typically about 40 photons per tritium decay. Given a light collection efficiency of 75% and a photocathode quantum efficiency of 20%, the number of photoelectrons produced is about 6 photoelectrons per tritium decay event.

Options for registration of ^{37}Ar scintillation

The wavelength of light emitted in LAr (128 nm) lies in the far vacuum ultraviolet (VUV) region, and quartz windows are no longer sufficient for its direct detection; fragile and expensive photomultiplier tubes (PMTs) with MgF2 windows, transparent in this wavelength region, are required. The use of PMTs with conventional windows is possible with the use of coatings with a spectrum shifter - Tetraphenyl-butadiene (TPB), which converts primary photons into photons with a wavelength of 420 nm. It is also possible to use xenon dissolved in LAr in small concentrations (<100 ppm), which provides a wavelength shift to the 173 nm region, where quartz windows are transparent.

Initially, when developing the project, we planned to use the new high-efficiency low-temperature PMT from Hamamatsu Photonics R11410 with a quartz window. Its efficiency at cryogenic temperatures reaches 26% at a wavelength of 175 nm.



PMTs FOR DARK MATTER DETECTION R11065, R11410

The R11065 and R11410 are 76 mm (3") diameter head-on PMTs with extremely low radioactivity, and are operable in low-temperature environments such as liquid Xe (-110 °C) and liquid Ar (-186 °C).
R11410-20 and R11065-20, whose radioactivities are 1/4 lower compared with those of R11410 and R11065, are now available.

■FEATURES
- High QE
- High cathode linearity
- Extremely low radioactivity

■Spectral Response

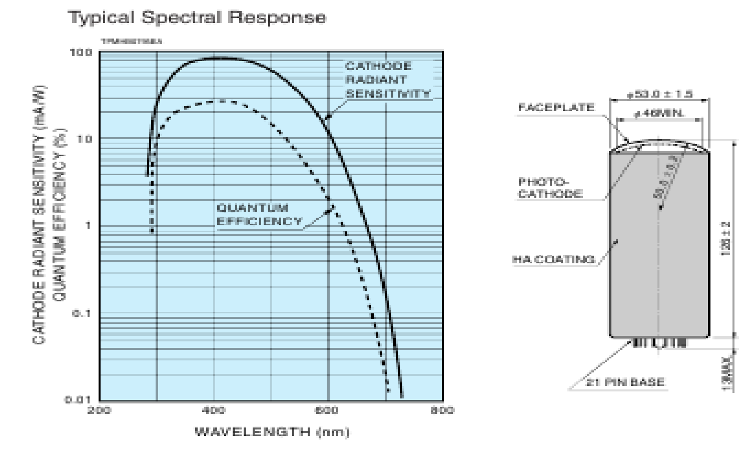
■Cathode Current Linearity Example

/HAMAMATSU PHOTONICS, Electron Tube Division, Photomultiplaer tubes and related products, catalog rev. Feb.2016, www.hamamatsu.com/.

Chose of PMT for installation prototype

The PMT with MgF2 window, the Hamamatsu R11410 low-temperature quartz windowed PMT, and even the Hamamatsu R331 tritium PMT were unavailable for a number of reasons. The MgF2 windowed PMT, the low-temperature Hamamatsu R11410 quartz windowed PMT, and even the Hamamatsu R331 quartz PMT were unavailable for a variety of reasons. Therefore, we chose the regular Hamamatsu R331-5 PMT, commonly used in tritium radiometers.

This PMT is sensitive in the range of 300 – 650 nm, At maximum sensitivity (420 nm) it provides a quantum efficiency of 25%. The operating temperature range is from -30 to +50 C, so it should be protected from low temperatures in the measuring chamber.



Spectral response of R331-95 PMT

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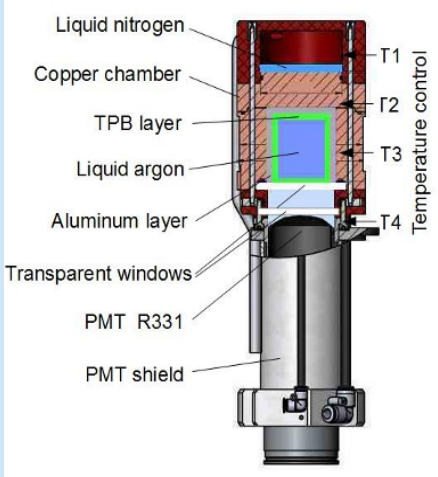
Detection unit based on PMT R331

The design of the measuring chamber of detection unit uses electrolytically purified copper and aluminum that do not contain significant amounts of natural and man-made radionuclides.

In the upper part of the measuring chamber, there is a cooler with liquid nitrogen. The thin-walled aluminum cylinder is located inside the thick-walled copper chamber. At the bottom, there is a protective transparent window optically connected to the Hamamatsu R331-05 PMT.

The inner surface of the aluminum cylinder and the surface of the window are covered with a layer of TPB, which converts the primary 128 nm photons of liquid argon scintillations into visible light. Local heating is provided to maintain an acceptable temperature of the photocathode of the photomultiplier at least -30 ^{0}C.



Since the tests of the prototype revealed low efficiency of registration of Auger electrons of 37Ar, additional research was carried out and work was carried out to improve its characteristics.

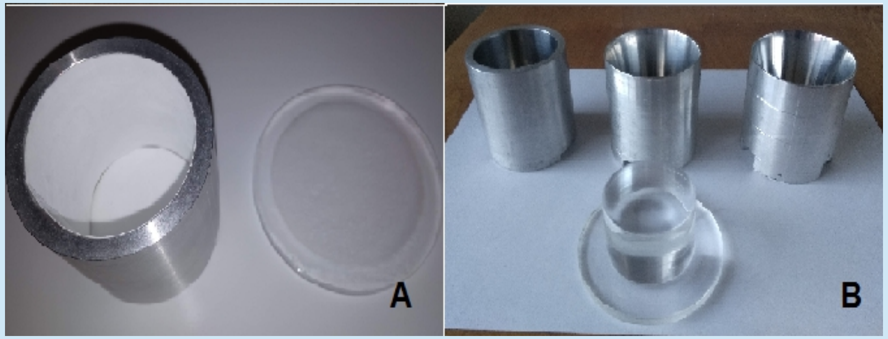
Sectional view of the detecting unit.

Main areas of improvement work

To eliminate the identified shortcomings of the installation prototype, the main directions of work were as follows:
- Optimization of the geometry of the measuring chamber;
- Manufacturing of a light guide;
- Carrying out experiments with a model source based on tritium in order to confirm the improvement in light collection;
- Optimization of the technology for coating the wavelength shifter;
- Improvement of the computer program in order to analyze the ratio of fast and slow components.

The geometry of measuring chamber optimization

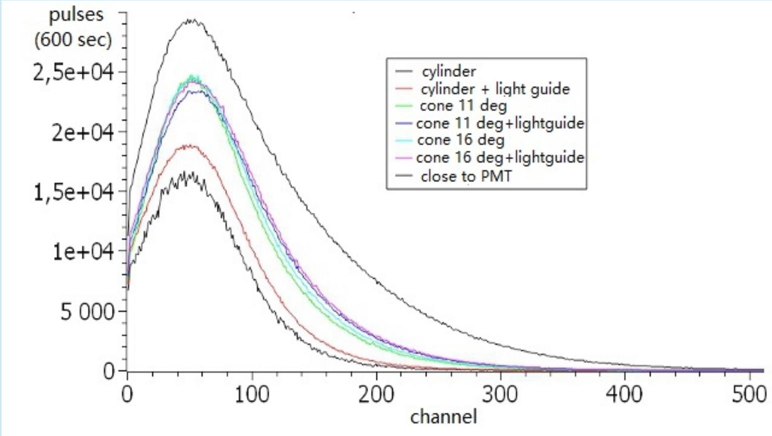
Three versions of light-collecting measuring chambers were made of high-purity aluminum: cylindrical and conical, with opening angles of 11 and 16 degrees, as well as a light guide made of organic (polyacrylic) glass.



(A) - Original view of the measuring chamber with a flat protective glass without a light guide. (B) - Various versions of measuring chambers with a profiled light guide.

Testing of optimized measuring chamber

The measurements were performed using a small-sized model tritium source (1.24 kBq in the Ultima Gold scintillator) and three variants of the light collection chamber: a cylindrical chamber and two conical chambers with different cone angles (11 and 16 degrees) with and without optical fiber. It is evident that the 16-degree conical chamber in combination with optical fiber leads to a 2-fold improvement in light collection relative to the original geometry - a cylindrical chamber without a light guide. The absolute value of the light collection efficiency reaches 70% of the light collection efficiency when the tritium source is located close to the PMT and covered with a reflective coating.



Results of measurements of various light collection for different variants of measuring chambers

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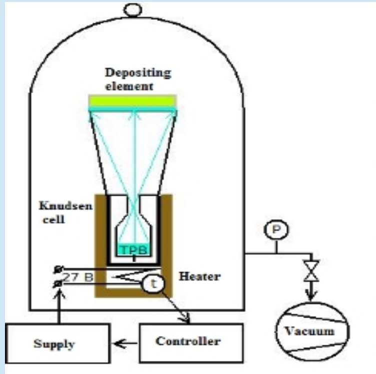
Optimization technology of TPB coating

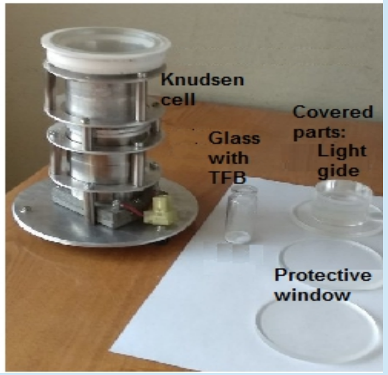
It is considered that the efficiency of applying the TPB coating by the thermal vacuum deposition (TVD) method (as well as the registration efficiency) is significantly higher than when applying by other methods (airbrushing or a brush with polymer plasticizers), ***/Christopher Benson, Gabriel D. Orebi Gann & Victor Gehma. Measurements of the intrinsic quantum efficiency and absorption length of TPB thin films in the vacuum ultraviolet regime The European Physical Journal C 78, N 329 (2018)/.***

Therefore, we have designed a laboratory stand for implementing this method. The device consists of the deposition device itself, placed in a vacuum volume, and components that ensure the deposition process.

Optimal thickness of the TPB coating is 2-3 µm (200-300 µg / cm²), this thickness was chosen as the target when applying TPB by the TVD method.



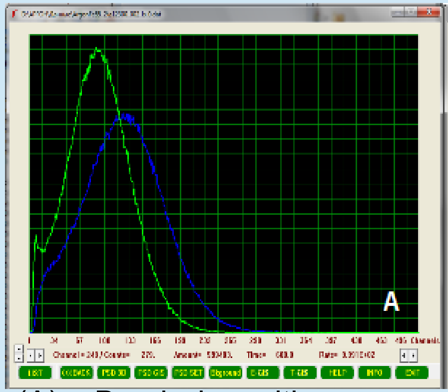
Functional schema of laboratory stand



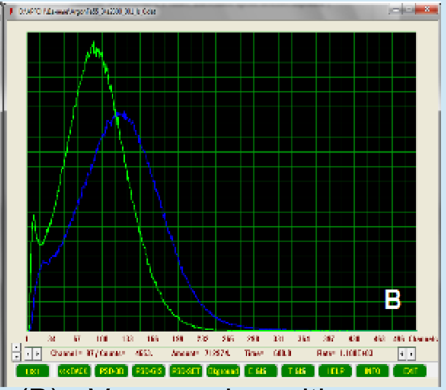
Knudsen cell, bottle with a TPB and elements to be deposited

Verification of the improvement after optimization

To experimentally verify the increase in the detection efficiency as a result of using vacuum deposition technology, measurements were carried out with a model Fe-55 source with an activity of 5 kBq. Spectrum (A) was obtained by applying TPB to a protective glass with a brush from a solution in toluene. Spectrum (B) was obtained by applying TPB by vacuum deposition using developed stand.



(A) – Brash deposition



(B) - Vacuum deposition.

As seen, in both cases all pulses are registered in the region of the single-photon peak, which indicates a small number of scintillation photons. The count rate in the left window was 1188 imp/s, and in the right - 991 imp/s, which indicates an improvement in the detection system registration efficiency by 20%, but this does not change the overall situation with the low registration efficiency of ^{37}Ar Auger electrons

Finally, as a result of optimizing the geometry of the measuring chamber, using a light guide and using the thermal vacuum deposition method for applying the TPB, the efficiency of scintillation registration in liquid argon was increased by 2.2 times. But, unfortunately, such an improvement is still insufficient for ^{37}Ar measurements..

Measures for signal/noise improvement

After the work carried out to improve the light collection and improve the technique for applying the TPB coating, further improvement of the sensitivity of prototype is determined by measures to increase the signal/noise ratio. Among these measures are:

- PMT noise reduction;
- increase the amplitude of the useful signal;
- selection of the area of interest (ROI) away from the noise area;
- rejection of false impulses by the PSD (pulse shape discrimination) parameter ;
- use of single-electron PMT capable of registering single photons.

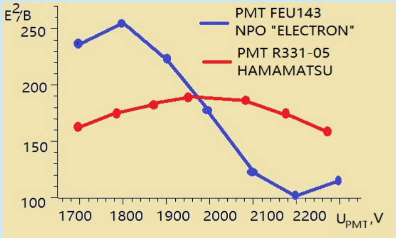
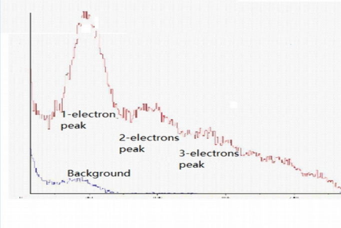


Figure of merit for PMT FEU143 and R331-05



Single-electron spectrum of FEU-143

As we can see, the characteristics of regular PMT R331-05 are not very good, they are inferior even to the old Soviet FEU143 (not produced). The best option would be to replace it with the originally planned R11410, but it is also not available now.

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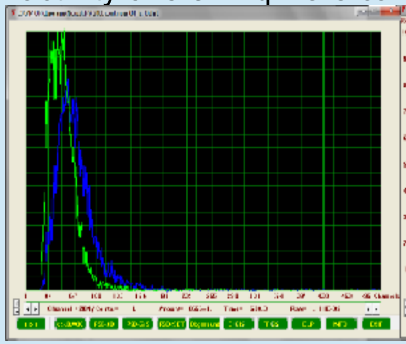
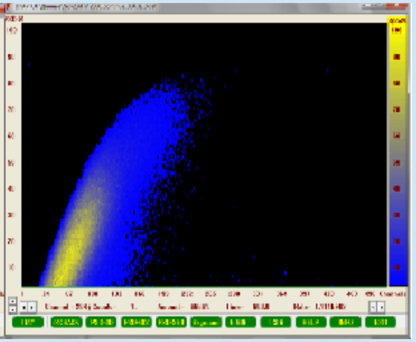
Final testing of installation prototype

The measures taken did not ensure the achievement of the required characteristics sufficient for measuring the activity of ^{37}Ar. This is due to the low energy of ^{37}Ar electrons (2.8 keV), insufficient quantum efficiency of the PMT (20%), losses due to wavelength shift by the TPB coating (10 - 40%) and incomplete collection of photons (70%). The effective resulting value of quantum efficiency in this case is 1.5%. Then 40 photons on average emitted during the absorption of an electron by ^{37}Ar cause the emission of only 0.6 photoelectrons by the PMT photocathode. This is 10 times less than when registering tritium beta particles and is completely insufficient for detecting 37Ar decays.

Test measurements of a 72 L sample with 37Ar activity of 8.3 kBq were carried out:



Spectrum of ^{37}Ar electrons



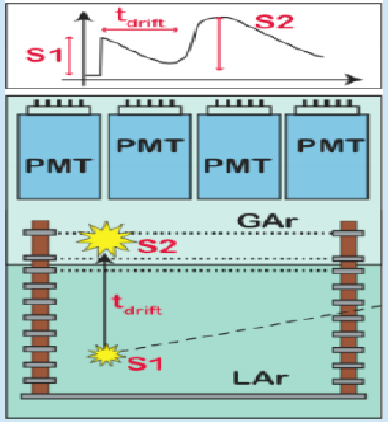
3D projection of PSD

The estimated registration efficiency was 1.3%. If it were possible to replace the R331-05 PMT with the R11410, all the problems listed above would be solved. But it is not available.

Increasing light output using EL

The most effective way to increase the amplitude of the useful signal when registering scintillations of inert gases is to use the electroluminescence (EL) effect.

These effect is widely used in experiments to search for dark matter ***/P. Agnes et al., "Search for low-mass dark matter WIMPs with 12 ton-day exposure of DarkSide-50," Phys. Rev. D 107 (2023), 063001/.***



Electroluminescent argon detector.

In the lower part of the detector, filled with liquid argon, interaction of charged particles or quanta of ionizing radiation with argon atoms occurs. As a result, some of the argon atoms go into an excited state (excitons are formed), and some of the atoms are ionized (ions and free electrons are formed). The ionization energy of atoms in liquid argon is 23.6 eV, the excitation energy is 11.55 eV

At the bottom of the detector there is a cathode, and a grid is located at the phase interface. The potential difference between the grid and the cathode creates a drift field that pulls free electrons from the liquid into the gas gap. In the upper part of the gas gap, in front of the photomultiplier, there is a second grid (anode), which provides an accelerating voltage sufficient to excite electroluminescence in argon gas.

Free electrons that enter the gas phase as a result of directed drift under the action of a pulling electric field, colliding with atoms of argon gas, these collisions occur

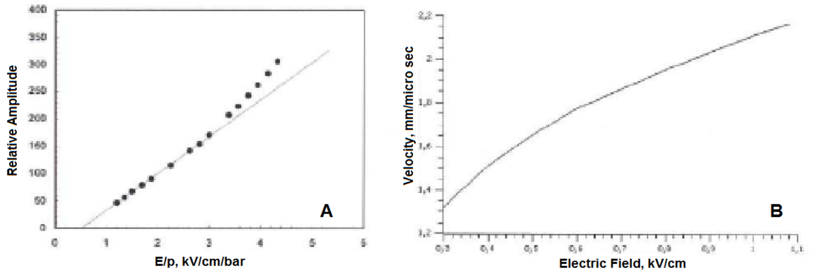
EL main characteristics

elastically, with almost no loss of energy. When the electrons reach an energy of 11.55 eV, the threshold for electroluminescence of argon, the electrons excite neutral atoms and lose energy, but can be re-accelerated and cause new excitation.

Excited argon atoms are in lower energy states, singlet ($^1\Sigma u+$) and triplet ($^3\Sigma u+$), which correspond to the fast and slow components of scintillations:

Argon phase	Singlet decay time	Triplet decay time	Wawelent h, nm
Liquid Ar	7.0±1.0 ns	1.6±0.1 μs	129.6
Ar gas	4.2 ns	3.1±0.1 μs	128

Below is the dependence of the amplitude and signal of the detector on the intensity of the applied electric field in the luminescence region. It is evident that at an intensity of about 2 - 3 kV/cm, the amplitude of the signal, and, consequently, the light output increases by 100 - 150 times.



Electroluminescent signal amplitude increase (A) and electron drift velocity (B) as a function of voltage.

Thus, electroluminescent signal amplitude enhancement is a promising direction for improving the prototype characteristics.

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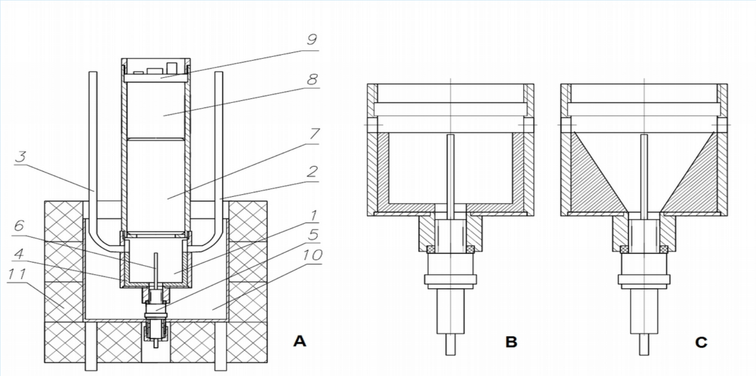
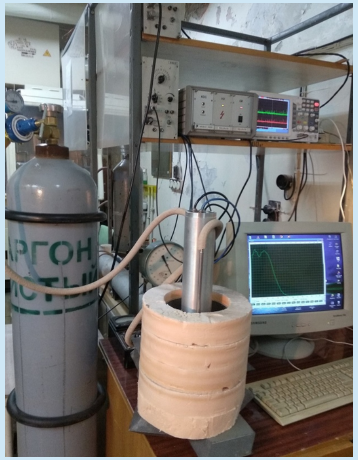
Laboratory stand for EL argon detector

An experimental prototype of the detection unit, was developed (A) with cylindrical(B) and conical © measuring chambers and a central electrode.



Drawing of laboratory stand measuring chamber

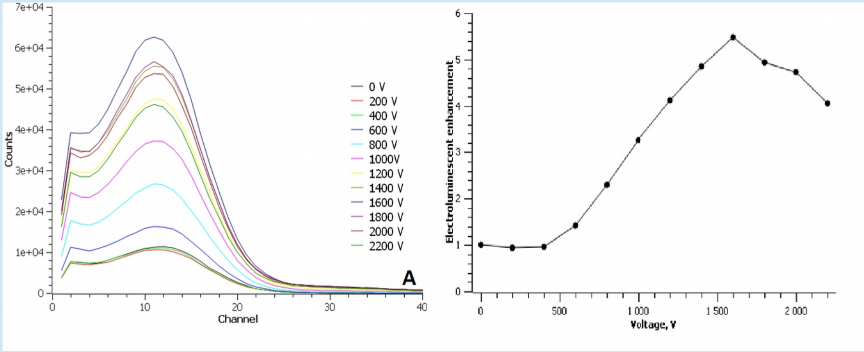


View of laboratory stand

Laboratori stend include: 1 - aluminium measuring chamber with TPB coating (replaceable), 2,3 - argon supply line, 4 - measuring chamber housing, 5 - high-voltage input, 6 - internal electrode, 7 - PMT, 8 - PMT electronics unit, 9 - panel with connectors, 10 - chamber for filling with liquid nitrogen, 11 - thermal insulation.

Testing of the laboratory stand of EL detector

A source of ^{63}Ni beta particles with an activity of about 3,7 kBq, having a ring shape coaxial with the central electrode, was placed in the lower part of the measuring chamber. The measurements were carried out, a positive voltage was applied to the electrode in the range from 0 to +2200 V.

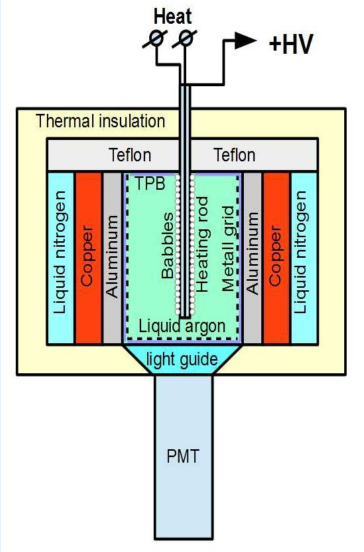


Amplitude spectra at different voltage (A); Relative increase in amplitude (B).

As seen, the voltage increases from 0 to 1700 V, the signal amplitude increases by 6 times, and with a further increase in voltage it begins to decrease. When the voltage increased above 2200 V, a breakdown occurred at the high-voltage input. It is likely that the decrease in the signal amplitude when the voltage reached 1700 V was caused by pre-breakdown phenomena at the input. (the limiting resistor began to heat up). However, the effect of electroluminescent signal amplification has been obtained.

EL application for improving a prototype

Measuring chamber has a volume of 100 cm3 and therefore, technical solutions developed for Dark Side Experiment not applicable in our case. The most logical option for developing a small-volume detection system is to constract an axially symmetric chamber with a central high-voltage electrode.



LAr scintillation detector with EL improving.

At the bottom, the photomultiplier, optically connected to the measuring chamber using a light guide. The internal surfaces of the chamber and the light guide are coated with a layer of TPB. Along the axis of the chamber there is a heated metal rod to which a high-voltage potential +HV is supplied. As a result of heating, argon bubbles on the surface of the rod, provides the gas phase necessary to realize the effect of gas EL in LAr.

Conclusions: 1)The developed prototype of the setup for registering scintillations in LAr currently does not provide the required sensitivity. 2) The prototype characterictics can be corrected by using a low-temperature UV photomultiplier, which is not available. 3) The characteristics of the prototype can also be improved by using EL reinforcement. 4) Work is underway to create an improved detection unit using the EL effect in combination with bubble technology..

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